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Mgr inż. Magdalena Karolina Mikus

**Biopolimerowe folie aktywne  
i ich zastosowanie jako powłoki ochronne  
w monitorowaniu jakości pozbiorczej jabłek**

Biopolymer active films and their use as protective coatings  
in post-harvest quality monitoring of apples

Rozprawa doktorska

Doctoral thesis

Rozprawa doktorska wykonana pod kierunkiem

dr hab. inż. Sabiny Galus, prof. SGGW

Instytut Nauk o Żywności

Katedra Inżynierii Żywności

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## Streszczenie w języku polskim

### *Biopolimerowe folie aktywne i ich zastosowanie jako powłoki ochronne w monitorowaniu jakości pozbiorczej jabłek*

Celem pracy była charakterystyka jadalnych folii aktywnych wytworzonych z pektyny jabłkowej z dodatkiem wybranych kwasów fenolowych jako składników o działaniu przeciwutleniającym. Zakres badań obejmował analizę wpływu dodatku związków aktywnych na właściwości funkcjonalne oraz użytkowe badanych jadalnych folii pektynowych, w aspekcie oceny barwy, przepuszczalności światła, parametrów mechanicznych, stabilności termicznej oraz zwilżalności i zdolności do pochłaniania wilgoci z otaczającego środowiska (właściwości sorpcyjne). Jednocześnie oceniono potencjał praktyczny folii pektynowych poprzez aplikację roztworów filmotwórczych na powierzchnię jabłek odmiany *Golden Delicious* i zbadanie wpływu wytworzonych powłok ochronnych na zmiany masy, ekstraktu, kwasowości, pH, barwy, jędrności oraz aktywności oddechowej owoców, monitorowane w czasie ich przechowywania w temperaturze pokojowej przez 28 dni.

Analiza otrzymanych wyników wykazała, że kwasy fenolowe stanowią skuteczny środek sieciujący materiałów otrzymanych z pektyny jabłkowej. Dodatek kwasów fenolowych wpłynął na wzrost grubości folii i zmniejszenie zawartości wody. Ponadto wykazano, że proces adsorpcji pary wodnej zachodził najintensywniej w pierwszych 10 godzinach, a jego przebieg był podobny w przypadku wszystkich wariantów badanych materiałów. Folie otrzymane z pektyny jabłkowej i modyfikowane kwasem kawowym są obiecujące pod względem ich potencjalnego zastosowania, wykazując wyższe wartości kąta zwilżania wody, co może wpływać na ograniczenie niekorzystnego wpływu wilgoci na produkt. Oprócz tego wykazano, że folie jadalne z dodatkiem kwasu galusowego oraz protokatechowego charakteryzowały się wyższą barierowością wobec pary wodnej w porównaniu do pozostałych folii. Analiza mikroskopowa wykazała, że wszystkie otrzymane folie były gładkie i jednorodne, cechujące się dobrą kompatybilnością pektyny jabłkowej z kwasami fenolowymi.

Folia kontrolna bez dodatku kwasów fenolowych oraz pozostałe folie kompozytowe były wizualnie przezroczyste, gładkie, jednorodne oraz wykazywały odcień w kierunku barwy żółtej. Folia kontrolna charakteryzowała się najwyższą wartością transmitancji światła, natomiast po wprowadzeniu kwasów fenolowych zaobserwowano

zauważalne zmniejszenie przenikania światła we wszystkich wariantach folii i zakresie pomiarowym długości fali 200-800 nm. Można zatem stwierdzić, że dodatek kwasów fenolowych zwiększa barierowość folii dla światła. Ponadto, zaobserwowano, że folie z kwasami fenolowymi charakteryzowały się zmniejszonym połyskiem zarówno przy kącie 20°, jak i 60°. Dodatek kwasów fenolowych wpłynął na zwiększenie wytrzymałości na rozciąganie folii oraz modułu Younga w porównaniu z foliami kontrolnymi.

Badania przechowalnicze wykazały, że powlekanie świeżych jabłek wpłynęło na zmniejszenie ubytków masy oraz zachowanie jędrności w porównaniu do jabłek niepowlekanych. Zaobserwowano, że proces powlekania znacząco spowolnił szybkość zwiększania się ekstraktu w owocach przez cały okres przechowywania (28 dni). Kwasowość jabłek powlekanych zmniejszyła się nieistotnie w porównaniu do owoców niepowlekanych oraz między poszczególnymi wariantami nie odnotowano większych różnic w wartościach pH. Oprócz tego, jabłka poddane procesowi powlekania, charakteryzowały się mniejszą aktywnością oddechową, z czego wynikało wolniejsze dojrzewanie owoców oraz opóźnienie żółknięcia skórki jabłek, a tym samym zachowanie odpowiedniej jakości konsumpcyjnej owoców.

**Słowa kluczowe:** folie jadalne, powłoki jadalne, pektyna jabłkowa, kwasy fenolowe, jabłka, jakość pozbiorcza, pakowanie aktywne

## Streszczenie w języku angielskim

### *Biopolymer active films and their use as protective coatings in post-harvest quality monitoring of apples*

This study aimed to characterize edible active films produced from apple pectin with added phenolic acids as antioxidant components. The study analyzed the effect of these active compounds on the functional properties of the pectin films, assessing their color, light transmittance, mechanical parameters, thermal stability, wettability, and moisture sorption properties. Simultaneously, the practical potential of pectin films was assessed by applying film-forming solutions to the surface of *Golden Delicious* apples. The effect of the resulting protective coatings on changes in fruit weight, extract, acidity, pH, color, firmness, and respiratory activity was monitored during 28 days of storage at room temperature. Analysis of the results indicated that phenolic acids are effective cross-linking agents for materials derived from apple pectin.

The addition of phenolic acids increased film thickness and reduced water content. The water vapor adsorption process was most intense in the initial 10 hours, exhibiting a similar course across all tested material variants. Films derived from apple pectin and modified with caffeic acid show promise for potential applications, demonstrating higher water contact angle values that may mitigate the adverse effects of moisture on products. Edible films incorporating gallic and protocatechuic acids exhibited superior water-vapor barrier performance compared to other films. Microscopic analysis showed that all obtained films were smooth and uniform, demonstrating good compatibility between apple pectin and phenolic acids.

The control film, without phenolic acid addition, and the composite films were visually transparent, smooth, and uniform and exhibited a yellow tint. The control film had the highest light transmittance. The addition of phenolic acids significantly reduced light transmission across all film variants within the 200-800 nm wavelength range, indicating an increased light barrier. Furthermore, films containing phenolic acids showed reduced gloss at both 20° and 60° angles. The addition of phenolic acids increased the film's tensile strength and Young's modulus compared to the control films.

Storage studies showed that coating fresh apples reduced weight loss and retained firmness compared to uncoated apples. The coating process significantly slowed the rate of increase in fruit extract throughout the entire 28-day storage period. The acidity of coated apples decreased insignificantly compared to uncoated fruit, and no significant differences

in pH values were observed between the individual varieties. Coated apples showed reduced respiratory activity, resulting in slower fruit ripening and delayed skin yellowing, thereby maintaining the fruit's eating quality.

**Keywords:** edible films, edible coatings, apple pectin, phenolic acids, apples, post-harvest quality, active packaging

## WYKAZ PUBLIKACJI STANOWIĄCYCH ROZPRAWĘ DOKTORSKĄ

[P1] Mikus M., Galus S. (2025). Extending the Shelf Life of Apples After Harvest Using Edible Coatings as Active Packaging – A Review. *Applied Sciences*, 15(2), 767 (100 pkt, IF 2,5). <https://doi.org/10.3390/app15020767>

[P2] Mikus M., Galus S. (2025). The Effect of Phenolic Acids on the Sorption and Wetting Properties of Apple Pectin-Based Packaging Films. *Molecules*, 30(9), 1960 (140 pkt, IF 4,6). <https://doi.org/10.3390/molecules30091960>

[P3] Mikus M., Galus S. (2026). The Effect of Selected Phenolic Acids on the Functional Properties of Pectin-based Packaging Films. *Applied Sciences*, 16(2), 797 (100 pkt, IF 2,5). <https://doi.org/10.3390/app16020797>

[P4] Mikus M., Szulc K. Galus S. (2025). The Use of Bioactive Edible Coatings Based on Pectin and Phenolic Acids for Enhancing Quality Attributes of Golden Delicious Apples During Storage. *Processes*, 13(12), 3821 (70 pkt, IF 2,8). <https://doi.org/10.3390/pr13123821>

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## Wykaz skrótów

Symbol	Znaczenie i jednostka
$\Delta E$	bezwzględna różnica barwy/całkowita różnica barwy (-)
$\Delta l$	wydłużenie próbki, przy którym nastąpiło zerwanie folii (mm)
A	pole powierzchni przekroju folii (mm <sup>2</sup> )
$a^*$	współczynnik barwy czerwonej/zielonej (-)
$A_{600}$	wartość absorbancji przy długości fali 600 nm
ABTS	2,2'-azyno-bis(etylobenzotiazolino-6-sulfonian)
$b^*$	współczynnik barwy żółtej/niebieskiej (-)
$C_{f,\infty}$	stężenia równowagowe substancji czynnej w folii (mg/l)
$C_{S,\infty}$	stężenia równowagowe substancji czynnej w substancji symulującej żywność (mg/l)
$Ca^{2+}$	kation wapnia
$C_2H_4$	etylen
$CaCl_2$	chlorek wapnia
$CO_2$	dutlenek węgla
CFA	kwaskawowy, kwas 3,4-dihydroksycynamonowy
CMA	kwaskumarowy, 4-hydroxycinnamic acid
$C_t$	stężenie substancji czynnej w czasie w ośrodku uwalniania (mg/l)
$C_\infty$	stężenie substancji czynnej w stanie równowagi w ośrodku uwalniania (mg/l)
D	współczynnik dyfuzji substancji czynnej (m <sup>2</sup> /s)
DPPH	2,2-difenylo-1-pikrylohydrazyl
E	wydłużenie względne przy zerwaniu (%)
$F_b$	siła powodująca odkształcenie w punkcie b, stanowiącym granicę liniowej plastyczności (N)
$F_{max}$	siła powodująca zerwanie folii (N)
FRA	kwaskerulowy, kwas 4-hydrokso-3-metoksycynamonowy
FTIR	spektrometria w podczerwieni z transformacją Fouriera
GLA	kwaskalusowy, kwas 3,4,5-trihydroksybenzoesowy
IUPAC	Międzynarodowa Unia Chemii Czystej i Stosowanej
K	wskaźnik konsystencji (Pa·s <sup>n</sup> )
$l_0$	długość początkowa próbki (mm)

$L, l$	grubość folii (m lub mm)
$L^*$	parametr barwy określający jasność (-)
$m_0$	masa próbki przed rozpuszczeniem (g)
$m_1$	masa próbki przed pęcznieniem (g)
$m_2$	masa próbki po pęcznieniu (g)
$m_p$	masa próbki przed wysuszeniem (g)
$m_{p0}$	masa początkowa (dzień 0) (g)
$m_r$	masa próbki po rozpuszczeniu (g)
$m_s$	masa próbki po wysuszeniu (g)
$m_t$	masa w dniu $t$ (g)
$M_0$	początkowa zawartość wody (g/g s.s.)
$M_e$	zawartość wody w czasie $t$ (g/g s.s.)
$M_t$	całkowita ilość zaadsorbowanej wody (g/g s.s.) w czasie $t$ (s)
$n$	bezwymiarowy wskaźnik zachowania się pływnięcia
$O$	nieprzezroczystość (a.u./mm)
$O_2$	tlen
$P$	pęcznienie folii (%)
PCA	kwas protokatechowy, 3,4-dihydroxybenzoic acid
$q_n$	niezerowe, dodatnie pierwiastki z $\tan(q_n) = -\alpha q_n$ (wartość $n$ od 1 do 6)
$R$	rozpuszczalność folii w wodzie (%)
SD	odchylenie standardowe
SNA	kwas synapinowy, 3,5-dimethoxy-4-hydroxycinnamic acid
TS	wytrzymałość na rozciąganie (MPa)
TTA	kwaskowość miareczkowa (%)
$U_t$	procentowy ubytek masy po $t$ dniach (%)
$V_1$	zużyta objętość roztworu (ml)
$V_f$	objętość folii (m <sup>3</sup> )
WVP	przepuszczalność pary wodnej (g/g s.s.)
YM	moduł Younga/moduł sprężystości (MPa)
$\tau$	naprężenie ścinające (Pa)
$\dot{\gamma}$	szybkość ścinania (s <sup>-1</sup> )

## 1. UZASADNIENIE PODJĘCIA TEMATU PRACY DOKTORSKIEJ

Do opracowania rozdziału posłużyła pierwsza publikacja [P1] z cyklu publikacji wchodzących w skład niniejszej rozprawy doktorskiej: Mikus M., Galus S. (2025). **Extending the Shelf Life of Apples After Harvest Using Edible Coatings as Active Packaging – A Review. *Applied Sciences*, 15(2), 767.** W pracy przeglądowej dokonano kompleksowej charakterystyki jabłek, obejmującej ich wartość odżywczą, wymagania uprawowe oraz kluczowe czynniki determinujące jakość owoców, ze szczególnym uwzględnieniem warunków zbioru i przechowywania. Przeanalizowano również zagadnienia związane z powstawaniem odpadów jabłkowych oraz przedstawiono możliwości ich zagospodarowania. Istotną część pracy poświęcono jadalnym powłokom ochronnym, koncentrując się na metodach ich wytwarzania, stosowanych materiałach oraz rozwiązaniach znajdujących zastosowanie komercyjne w ochronie owoców. Dokonano przeglądu aktywnych folii i powłok jadalnych, ze szczególnym uwzględnieniem ich funkcji ochronnych. Omówiono właściwości wybranych związków aktywnych, takich jak ekstrakty roślinne, kwasy fenolowe, nanocząstki oraz nanomateriały, stosowanych jako dodatki do jadalnych folii w celu poprawy ich funkcjonalności. Ostatnim analizowanym zagadnieniem była rola jadalnych powłok biopolimerowych w utrzymaniu jakości jabłek po zbiorze, a także wyzwania technologiczne i perspektywy dalszego rozwoju tych rozwiązań w kontekście zrównoważonego przechowywania owoców.

Przeprowadzony przegląd literatury (etap 1 badań) pozwolił na kompleksową analizę aktualnego stanu wiedzy dotyczącego jakości jabłek po zbiorze, jadalnych powłok ochronnych oraz aktywnych dodatków stosowanych w ich formulacji. Analiza dostępnych publikacji wykazała znaczną różnorodność w zakresie doboru składników powłokotwórczych (biopolimerów), ich proporcji oraz metod otrzymywania roztworów powlekających, co utrudnia jednoznaczną ocenę skuteczności poszczególnych rozwiązań technologicznych. Jednocześnie nie zidentyfikowano badań dotyczących powlekania świeżych jabłek z zastosowaniem składników oraz ich proporcji odpowiadających rozwiązaniom zaproponowanym w niniejszej rozprawie. Zidentyfikowana luka badawcza stała się podstawą do sformułowania celu pracy oraz postawienia hipotez badawczych, zakładających możliwość opracowania jadalnej, aktywnej powłoki zdolnej do skutecznego utrzymania jakości jabłek po zbiorze w warunkach konsumpcyjnych. Wyniki przeglądu literatury stanowiły również podstawę do zaprojektowania kolejnego etapu badań (etap 2), obejmującego opracowanie składu roztworów powłokotwórczych oraz metod ich

otrzymywania. Efektem realizacji etapu 1 była publikacja przeglądowa [P1], która stanowi merytoryczne uzasadnienie podjęcia badań eksperymentalnych przedstawionych w dalszych rozdziałach rozprawy.

### **1.1. Determinanty jakościowe jabłek**

Polska jest europejskim liderem w produkcji jabłek i czwartym ich producentem na świecie (po Chinach, Stanach Zjednoczonych i Turcji). W 2024 roku produkcja jabłek w Polsce wyniosła około 3,4 miliona ton. Jabłka posiadają wiele cech korzystnie wpływających na zdrowie człowieka oraz są jednymi z ulubionych owoców spożywanych przez konsumentów (Ropelewska i wsp. 2025). Jedną z najpopularniejszych odmian jabłek jest odmiana *Golden Delicious*, charakteryzująca się cienką i delikatną zieloną skórką oraz słodkim smakiem (Sredojevic i wsp. 2025). Jabłka należą do owoców niskokalorycznych, o wysokiej zawartości wody. Ponadto są pożądanym składnikiem diety, ze względu na obecność różnych potencjalnych związków aktywnych, w tym węglowodanów, minerałów oraz witaminy C (Jeelani i wsp. 2025). Jabłka zawierają także liczne kwasy organiczne, takie jak kwas askorbinowy, jabłkowy, cytrynowy oraz cukry – glukozę, fruktozę i sacharozę. Dodatkowo są źródłem wielu makroelementów, do których można zaliczyć potas, sód, magnez, wapń, fosfor oraz śladowe ilości żelaza, cynku, manganu lub miedzi (Butkeviciute i wsp. 2021). Oprócz tego jabłka zapewniają ochronę przeciwutleniającą organizmu, będąc bogatym źródłem flawonoidów oraz kwasów fenolowych. W ostatnich latach obserwuje się wzrost rynkowego popytu na świeże owoce dostępne przez cały rok. Pomimo rosnącego zapotrzebowania na całoroczną dostępność świeżych jabłek, ich okres przydatności do spożycia ulega skróceniu, m.in. na skutek zachodzących zmian fizjologicznych, co prowadzi do zwiększenia strat jakościowych oraz generowania strat ekonomicznych (Oraee i wsp. 2025).

Straty owoców i warzyw po zbiorach stanowią poważny problem, którego rozwiązanie jest ściśle powiązane z zapewnieniem zrównoważonego gospodarowania zasobami oraz zagwarantowaniem globalnego bezpieczeństwa żywnościowego. Z doniesień naukowych wynika, że 30% owoców pochodzących z rocznych upraw, wynoszących około 656 milionów ton, stanowią straty. Aby ograniczyć marnotrawienie świeżych owoców po zbiorach, dąży się do wdrażania ulepszonych sposobów zrównoważonego zarządzania ich gospodarowaniem pozbiornym, unowocześniania infrastruktury przechowalniczej, a także wprowadzania nowych i zmodyfikowanych technologii utrwalania. Promowane są

również praktyki, zapewniające minimalizację ilości generowanych odpadów poprzez ich ponowne zagospodarowanie oraz gwarancję bardziej stabilnych dostaw żywności (Tarawneh i wsp. 2025). Do czynników wpływających na skrócenie okresu przydatności do spożycia owoców zalicza się m.in. niewłaściwą temperaturę i nieodpowiedni skład gazowy atmosfery ochronnej w czasie przechowywania, zbyt długi czas transportu, a także nieodpowiednio dobrany rodzaj opakowań, które nie spełniają swojej funkcji. Natomiast odpowiednia wilgotność względna otaczającego środowiska podczas przechowywania owoców w sposób korzystny wpływa na wydłużenie ich trwałości przechowalniczej. Oprócz tego ważnym aspektem jest kontrolowanie szybkości oddychania owoców oraz stężenia gazów, takich jak para wodna, tlen oraz ditlenek węgla. Należy również zwrócić uwagę na fakt, że podczas przechowywania oraz transportu świeżych jabłek dochodzi do wydzielania się etylenu, co sprzyja degradacji owoców. Jabłka należą do owoców klimakterycznych, które dojrzewają po zbiorze w wyniku nasilonego wydzielania etylenu, towarzyszącego oddychaniu komórkowemu. Proces ten determinuje zmiany cech sensorycznych owoców, takich jak smak, zapach, barwa i tekstura, a jego nadmierna szybkość może skutkować obniżeniem jakości jabłek. Pod wpływem etylenu może dochodzić również do zmiany zawartości cukrów oraz syntezy lotnych aromatów (Busatto i wsp. 2016). Występowanie zmian w zawartości licznych lotnych związków organicznych podczas przechowywania wiąże się z wydzielaniem nieprzyjemnego zapachu, wpływającego na obniżenie atrakcyjności konsumpcyjnej jabłek. Często obserwuje się także występowanie miękkich, wodnistych oraz brązowych plam, będących wynikiem rozwoju pleśni, głównie z rodzaju *Fusarium*, *Botrytis* i *Penicilium*. Dlatego istotnym stało się dążenie do opracowania nowych, przyjaznych dla środowiska sposobów zabezpieczania oraz materiałów, które mogłyby wpłynąć na wydłużenie okresu przydatności owoców do spożycia (Martínez i wsp. 2025). Utrata jędrności w czasie przechowywania stanowi podstawowy problem w ocenie konsumpcyjnej atrakcyjności jabłek. Wynika ona ze zmian strukturalnych ścian komórkowych, związanych z degradacją polisacharydów pod wpływem zwiększonej aktywności enzymów hydrolitycznych, takich jak poligalakturonazy i pektynometylsterazy, co prowadzi do osłabienia połączeń międzykomórkowych i zmiękczenia tkanek owoców. Zastosowanie powłok ochronnych umożliwia ograniczenie intensywności procesów metabolicznych, w tym oddychania i biosyntezy etylenu, a także redukcję strat wody, co przyczynia się do spowolnienia degradacji ścian komórkowych i zachowania jędrności owoców podczas przechowywania (Yi i wsp. 2020).

## 1.2. Powlekanie żywności

W ostatnim czasie jadalne folie i powłoki biopolimerowe są coraz częściej stosowane w przemyśle spożywczym do wydłużania okresu przydatności do spożycia różnego rodzaju produktów (Oraee i wsp. 2025). Folie jadalne to wstępnie uformowane cienkie arkusze materiału, które mogą stanowić samodzielne opakowania, natomiast powłoki są formowane bezpośrednio na produktach żywnościowych poprzez wstępne nanoszenie ich na powierzchnię w postaci płynnych roztworów, co ma miejsce w przypadku m.in. powlekania świeżych owoców oraz warzyw (Pawase i wsp. 2025). Zazwyczaj grubość folii, wytwarzanej jako samodzielna struktura z przeznaczeniem do wykorzystania jako materiał opakowaniowy, wynosi od 0,05 do 0,3 mm, natomiast grubość powłoki jadalnej, stanowiącej integralną część produktu, wynosi poniżej 0,025 mm (Yadav i wsp. 2025). Na grubość folii wpływa gęstość, napięcie powierzchniowe, lepkość mieszanin folio- lub/i powłokotwórczych, a także warunki otrzymywania tych jadalnych struktur, wytwarzanych z wykorzystaniem naturalnych biopolimerów. Stosowanie biopolimerów do folii jadalnych stanowi zrównoważoną alternatywę dla tworzyw sztucznych, mając znaczący wpływ na utrwalanie żywności, przy jednoczesnym zachowaniu jej aktywnych składników (Tasnim i Islam 2026).

Jedną z funkcji folii/powłok jadalnych jest ograniczenie utraty wilgoci oraz spowolnienie przebiegu niepożądanych reakcji chemicznych w celu zachowania bezpieczeństwa oraz jakości żywności. Powłoki nanoszone bezpośrednio na powierzchnię produktu, jak już wspomniano, mogą stanowić alternatywę materiałów opakowaniowych, ale także mogą być uzupełnieniem konwencjonalnych materiałów ochronnych (Haghighi i wsp. 2026). W celu rozszerzenia zakresu funkcjonalności jadalnych powłok do ich składu wprowadza się substancje bioaktywne, w tym związki przeciwdrobnoustrojowe, przeciwutleniacze, a także dodatki aromatyczne i barwniki. Częstym problemem, występującym w przypadku zastosowań materiałów opakowaniowych w formie folii/powłok jadalnych o zróżnicowanej funkcjonalności, jest ograniczona przepuszczalność oraz słabe właściwości mechaniczne, które nie są porównywalne w stosunku do powszechnie stosowanych folii z tworzyw sztucznych (Kumar i wsp. 2022).

Materiały wytworzone na bazie biopolimerów, które mają być zastosowane do powlekania świeżych owoców, powinny charakteryzować się niską przepuszczalnością pary wodnej, wpływając na ograniczenie wysychania. Ponadto powinny charakteryzować się niską przepuszczalnością tlenu i ditlenku węgla w celu spowolnienia oddychania

i aktywności metabolicznej, wpływając na proces dojrzewania. Najczęściej do powlekania świeżych owoców i warzyw testowane są głównie matrycotwórcze roztwory białkowe z kukurydzy, soi oraz glutenu pszennego, które przy niskich wartościach wilgotności względnej otaczającego środowiska wykazują pożądane właściwości barierowe wobec gazów i lipidów (Tiwari i wsp. 2022). Natomiast pokrywanie owoców i warzyw lipidowymi powłokami wpływa na niekorzystny wzrost zawartości ditlenku węgla w materiale i prowadzi do wzrostu mikroorganizmów oraz zwiększonej zawartości aldehydu octowego i etanolu (Yousuf i wsp. 2021). Według Amin i wsp. (2021) wprowadzony do folii kompozytowych dodatek wosku powoduje zminimalizowanie ubytku masy owoców i warzyw, a dodatkowo prowadzi do poprawy stabilności termicznej warstw ochronnych, stosowanych na surowcach roślinnych.

W przypadku owoców do głównych przyczyn występowania uszkodzeń oraz chorób pozbiorczych zalicza się niekorzystne oddziaływanie bakterii i grzybów. Opracowane strategie powlekania owoców, łączące zastosowanie aktywnych dodatków, takich jak naturalne ekstrakty, biologiczne inhibitory oraz związki o działaniu przeciwdrobnoustrojowym, ukierunkowane są na ograniczenie rozwoju mikroorganizmów oraz redukcję częstości występowania chorób pozbiorczych (Pham i wsp. 2023). W literaturze przedmiotu wykazano, że zastosowanie jadalnych powłok wzbogaconych o dodatki pochodzenia naturalnego może skutecznie przyczynić się do zachowania jakości pozbiorczej owoców i warzyw. Wyższą aktywność przeciwutleniającą powlekanych owoców i warzyw zaobserwowano m.in. w przypadku zastosowania powłok z dodatkiem propolisu. Proces powlekania umożliwił jednocześnie utrzymanie zawartości witaminy C oraz flawonoidów na odpowiednim poziomie, istotnym z punktu widzenia wartości żywieniowej produktu (Yong i Liu 2021). Znaczące właściwości przeciwutleniające oraz przeciwdrobnoustrojowe zaobserwowano również w przypadku zastosowania związków fenolowych jako składników funkcjonalnych w jadalnych materiałach opakowaniowych (Singh i wsp. 2022).

Powlekanie żywności stanowi łagodną i nieinwazyjną metodę utrwalania, która wpisuje się w założenia Europejskiego Zielonego Ładu oraz koncepcję gospodarki o obiegu zamkniętym. Technologia ta przyczynia się do zwiększenia trwałości przechowywalnej żywności, poprawy jej stabilności oraz ograniczenia strat i marnotrawienia, w szczególności w odniesieniu do świeżych owoców (Ali i wsp. 2025). Zastosowanie powłok ochronnych może nie tylko prowadzić do redukcji intensywności procesów oddychania, utraty wody oraz rozwoju mikroorganizmów, ale w konsekwencji

sprzyjać zmniejszeniu strat żywności na kolejnych etapach łańcucha dostaw (Iñiguez-Moreno 2026). Podsumowując, procesy dojrzewania, a następnie starzenia się owoców, jak również straty wynikające z niewłaściwego zagospodarowania plonów w okresie pozbiornym, stanowią istotne wyzwanie w kontekście zachowania jakości oraz trwałości przechowalniczej tych surowców. W związku z tym zasadne jest prowadzenie dalszych badań nad innowacyjnymi, przyjaznymi dla środowiska rozwiązaniami, umożliwiającymi wydłużenie okresu przydatności owoców do spożycia, wśród których szczególną uwagę zwraca się na zastosowanie materiałów w postaci jadalnych powłok ochronnych jako perspektywicznej metody ograniczania niekorzystnych zmian jakościowych.

### 1.2.1. Materiały powłokotwórcze

Do najczęściej stosowanych materiałów w otrzymywaniu folii/powłok jadalnych zalicza się polisacharydy (np. celuloza, skrobia, pektyna, chitozan), białka (np. białka mleka, żelatyna, soja, gluten), lipidy (np. oleje, воск carnauba, воск pszczeli) oraz ich połączenia (Aggarwal i wsp. 2025). Przykłady stosowanych materiałów folio/powłokotwórczych przedstawiono na **Rysunku 1**.



**Rysunek 1.** Przykłady materiałów folio/powłokotwórczych

Zastosowanie odpadów owocowo-warzywnych do produkcji pektyny, istotnego materiału powłokotwórczego, wpisuje się w ideę zrównoważonego gospodarowania zasobami oraz gospodarki o obiegu zamkniętym. Jednym z najczęściej stosowanych polisacharydów w powlekaniu żywności jest pektyna, pozyskiwana głównie ze skórek owoców cytrusowych (85,5%) oraz wyłoków jabłkowych (14,0%). Pomimo licznych właściwości funkcjonalnych, pektyna może być modyfikowana z wykorzystaniem plastyfikatorów, emulgatorów oraz substancji bioaktywnych w celu poprawy właściwości mechanicznych, stabilności, przyczepności i funkcjonalności, tak aby spełniać wymagania stawiane aktywnym opakowaniom (Barrera-Chamorro i wsp. 2025).

Według definicji Międzynarodowej Unii Chemii Czystej i Stosowanej (IUPAC) plastyfikatory są dodatkami materiałowymi, zapewniającymi matrycy folio/powłokotwórczej większą elastyczność, podatność na obróbkę oraz rozciągliwość. Do najczęściej stosowanych plastyfikatorów w jadalnych opakowaniach w formie folii/powłok zalicza się glicerol oraz sorbitol, które zapobiegają powstawaniu porów oraz pęknięć w wytwarzanych strukturach. Aby plastyfikatory charakteryzowały się skutecznością w działaniu, muszą być kompatybilne z polimerami stosowanymi w roztworach folio/powłokotwórczych (Syarifuddin i wsp. 2025).

Obecnie konsumenci poszukują produktów minimalnie przetworzonych o wydłużonym okresie przydatności do spożycia oraz posiadających wysoką wartość żywieniową. Wymagania te są bardziej rygorystyczne w przypadku produktów nietrwałych, takich jak owoce i warzywa, stanowiąc wyzwanie w opracowaniu odpowiednich i zrównoważonych technik utrwalania. Lepsze zrozumienie mechanizmu tworzenia funkcjonalnych folii/powłok oraz odpowiednia edukacja konsumentów może wpłynąć na rozszerzenie możliwości zastosowania tego rodzaju materiałów biokompozytowych do przedłużania okresu przydatności m.in. owoców i warzyw, a także zwiększenia biodostępności związków określanych jako bioaktywne (Mitelut i wsp. 2021). Ponadto stale zwiększający się standard życia oraz wynikający z tego rozwój przemysłu spożywczego wpływa na zmiany właściwości funkcjonalnych i użytkowych folii/powłok jadalnych. W związku z tym coraz częściej oczekuje się, że jadalne materiały opakowaniowe, przeznaczone do ochrony świeżych owoców i warzyw, będą posiadały odpowiednie właściwości hydrofobowe, większą wytrzymałość mechaniczną oraz właściwości barierowe w stosunku do gazów oraz światła. Ważnym aspektem jest także zaprojektowanie folii/powłok, które poza poprawą walorów smakowych produktów, będą zgodne z wymaganiami i preferencjami konsumentów. Przykładem mogą być powłoki

wzbogacone o dodatki, takie jak czosnek lub imbir, które oprócz nadawania cech sensorycznych będą doskonałym środkiem konserwującym (Sun i wsp. 2022).

### **1.2.2. Substancje dodatkowe w powlekanii żywności**

W celu nadania foliom i powłokom jadalnym pożądanych właściwości użytkowych do ich struktury wprowadza się różnego rodzaju substancje dodatkowe, m.in. plastyfikatory. Jednakże, w celu nadania takim materiałom właściwości funkcjonalnych coraz częściej wzbogaca się ich skład o związki aktywne różnego pochodzenia (Iñiguez-Moreno 2026). Wyniki badań wskazują na istotną rolę ekstraktów roślinnych w procesie wydłużania okresu przydatności żywności do spożycia, a także w utrzymaniu w wielu owocach i warzywach odpowiedniego poziomu składników bioaktywnych o znaczeniu żywieniowym (Nair i wsp. 2020).

W ostatnich latach obserwuje się rosnące zainteresowanie olejkami eterycznymi, jako dodatków do jadalnych folii/powłok. Olejki eteryczne, będące metabolitami wtórnymi wytwarzanymi przez rośliny, są badane jako potencjalne składniki aktywne stosowane w powlekanii oraz utrwalaniu świeżych owoców i warzyw. Wykazano, że mogą one również stanowić alternatywę dla chemicznych środków konserwujących oraz skutecznie przyczyniać się do zachowania jakości pozbiorczej produktów. Jednocześnie ich stosowanie wiąże się często z modyfikacją cech sensorycznych, w szczególności smaku, co nie zawsze jest akceptowane przez konsumentów (Yousuf i wsp. 2021).

Do istotnych grup substancji funkcjonalnych stosowanych w powlekanii żywności należą związki fenolowe, będące metabolitami wtórnymi syntetyzowanymi przez rośliny, charakteryzujące się obecnością co najmniej jednego pierścienia aromatycznego z przyłączoną jedną lub kilkoma grupami hydroksylowymi. Związki fenolowe wykazują szerokie spektrum aktywności biologicznej, w tym właściwości przeciwzapalne, przeciwdrobnoustrojowe oraz przeciwutleniające, wynikające z ich zdolności do neutralizacji reaktywnych form tlenu poprzez wychwytywanie wolnych rodników. Ponadto liczne polifenole przyczyniają się do poprawy właściwości fizykochemicznych jadalnych folii i powłok, m.in. poprzez modyfikację barierowości wobec pary wodnej, wytrzymałości mechanicznej, barwy, rozpuszczalności oraz właściwości sensorycznych (Pedreiro i wsp. 2021).

Związki fenolowe są powszechnie klasyfikowane m.in. jako flawonoidy, garbniki, kwasy fenolowe lub kumaryny, w zależności od źródła pochodzenia oraz zróżnicowania strukturalnego. W obrębie kwasów fenolowych wyróżnia się dwie podgrupy – kwasy

zawierające strukturę C<sub>6</sub>-C<sub>1</sub>, takie jak np. kwas protokatechowy, galusowy oraz syringowy, a także kwasy posiadające trójwęglowy łańcuch boczny (C<sub>6</sub>-C<sub>3</sub>), do których zaklasyfikować można kwas kumarowy, kawowy, ferulowy oraz synapinowy (Zhang i wsp. 2022). Szczególnie bogatym źródłem kwasów fenolowych w diecie człowieka są rośliny strączkowe, nasiona oleiste, owoce, warzywa, zboża oraz zioła i przyprawy. Ponadto antocyjany, należące do związków fenolowych, znajdują zastosowanie w foliach i powłokach jadalnych jako wskaźniki pH, umożliwiające monitorowanie zmian jakości produktów spożywczych. Należy jednak zaznaczyć, że wprowadzenie związków fenolowych do matrycy polimerowej biokompozytów opakowaniowych może prowadzić do zwiększenia dyfuzji tlenu do produktu, co jest konsekwencją zaburzenia ciągłości uporządkowanej struktury sieci polimerowej (Khin i wsp. 2024). Związki fenolowe, stosowane do utrwalania żywności, powinny charakteryzować się niską toksycznością oraz niewielkim kosztem pozyskania. Dodatkowo istotne jest, aby wykazywały wysoką aktywność biologiczną już przy niskich stężeniach, a jednocześnie charakteryzowały się dobrą stabilnością i nie wpływały negatywnie na jakość sensoryczną żywności.

Sun i wsp. (2014) oceniali właściwości folii chitozanowych modyfikowanych różnymi stężeniami kwasu galusowego i wykazali istotne działanie przeciwdrobnoustrojowe wobec *Escherichia coli*, *Salmonella typhimurium*, *Listeria innocua* oraz *Bacillus subtilis*. Dane literaturowe wskazują również, że kwas galusowy wykazuje aktywność przeciwbakteryjną wobec patogennych gatunków z rodzaju *Vibrio*, wskazując na jego potencjał w tworzeniu materiałów opakowaniowych, ograniczających rozwój patogenów oraz bakterii odpowiedzialnych za psucie się żywności. Podobne obserwacje przedstawili Fang i wsp. (2018), którzy odnotowali wzrost aktywności przeciwdrobnoustrojowej powłok chitozanowych po wprowadzeniu kwasu galusowego. Dodatkowo obecność kwasu galusowego w strukturze folii prowadzi do zwiększenia ich elastyczności, co potwierdza jego funkcję jako plastyfikatora, tym samym redukując kruchość materiału. Badania przeprowadzone przez Araghi i wsp. (2015) wykazały, że kwas kawowy charakteryzuje się wyższą skutecznością w porównaniu do kwasu ferulowego, co może przyczynić się do poprawy właściwości barierowych oraz fizykochemicznych biodegradowalnych materiałów opakowaniowych, a w konsekwencji do zwiększenia ich bezpieczeństwa.

Folie chitozanowe z dodatkiem kwasu syringowego charakteryzują się zwiększoną gęstością, rozpuszczalnością w wodzie oraz zmniejszoną nieprzezroczystością w porównaniu do folii kontrolnych, a także wykazują działanie ograniczające rozwój

mikroorganizmów. Kwas syringowy posiada również właściwości przeciwutleniające, co pozwala na wykorzystanie go w aktywnych materiałach opakowaniowych w celu wydłużenia okresu przydatności żywności (Yang i wsp. 2019).

Liu i wsp. (2017) wykazali, że folie chitozanowe wzbogacone kwasem protokatechowym charakteryzowały się zwiększoną grubością oraz zmniejszoną przepuszczalnością światła i wody w porównaniu do folii kontrolnych. Kwas protokatechowy, powszechnie obecny w owocach i warzywach, wyróżnia się silnym działaniem przeciwutleniającym. Związki fenolowe mogą oddziaływać z łańcuchami chitozanu poprzez wiązania estrowe, oddziaływania elektrostatyczne i wiązania wodorowe, co zwiększa wytrzymałość mechaniczną powłok.

Przy doborze odpowiednich związków fenolowych należy uwzględnić takie czynniki jak wielkość cząsteczek, polarność oraz mechanizmy uwalniania substancji aktywnych z matrycy polimerowej (Martins i wsp. 2024). Z dostępnych badań wynika, że folie zawierające kwas taninowy charakteryzowały się bardziej zwartą strukturą oraz zwiększoną wytrzymałością mechaniczną, natomiast dodatek kwasu galusowego wpływał na poprawę elastyczności folii, pełniąc również funkcję plastyfikatora (Chiralt i wsp. 2020). Kwas ferulowy wyróżnia się silnym działaniem przeciwutleniającym i przeciwdrobnoustrojowym, co przyczynia się do poprawy bezpieczeństwa mikrobiologicznego żywności powlekanej formulacji zawierającymi kwas ferulowy. W badaniach potwierdzono skuteczność połączenia alginianu sodu z kwasem ferulowym w powlekanii jabłek, wykazując istotne zmniejszenie liczebności *Listerii monocytogenes* po 7 dniach przechowywania, przy jednoczesnym braku wpływu na wzrost grzybów *Saccharomyces cerevisiae* (Nicolau-Lapeña i wsp. 2021).

### **1.2.3. Metody powlekania**

Jakość powłok jadalnych jest determinowana przede wszystkim przez zastosowaną metodę aplikacji oraz efektywność adhezji powłoki do powierzchni produktu. Jedną z najczęściej stosowanych technik nanoszenia powłok jadalnych jest metoda zanurzeniowa, polegająca na krótkotrwałym zanurzeniu produktu w uprzednio przygotowanym roztworze powlekającym, zwykle w czasie od 5 do 30 sekund. Po etapie aplikacji nadmiar roztworu jest usuwany, a następnie realizowany jest proces odparowania, w trakcie którego rozpuszczalnik oraz pozostała ciecz są eliminowane z powierzchni produktu w wyniku kontrolowanych procesów suszenia i/lub ogrzewania. Główną zaletą tej powszechnie stosowanej metody jest równomierne pokrycie produktu ze wszystkich stron, nawet

w przypadku występowania powierzchni o zwiększonej chropowatości. Z tego względu zarówno właściwości powierzchni powlekanego produktu, jak i charakter powlekanej żywności mają istotny wpływ na dobór odpowiedniej metody nanoszenia powłoki. W odniesieniu do owoców, przed naniesieniem powłoki jadalnej na powierzchnię, należy uwzględnić ich charakter fizjologiczny, w szczególności podział na owoce klimakteryczne oraz nieklimakteryczne. Z doniesień naukowych wynika, że grubość powłok jadalnych jest zależna od wielu właściwości roztworu powlekającego, m.in. gęstości, lepkości lub napięcia powierzchniowego. Oprócz tego, w przypadku powlekania pokrojonych owoców lub warzyw nakładanie powłok jadalnych jest trudniejsze, ponieważ hydrofilowy charakter powierzchni utrudnia żelowanie roztworów powłokotwórczych, szczególnie tych o hydrofobowym charakterze (Pandey i wsp. 2022).

Metoda zanurzeniowa stanowi uniwersalną technikę nanoszenia powłok, ponieważ może być stosowana w szerokim zakresie lepkości roztworów powlekających. Zazwyczaj jest metodą stosowaną do nakładania jednowarstwowych powłok, jednak rozwój technologiczny umożliwia również otrzymywanie powłok wielowarstwowych, nanoszonych naprzemiennie w celu uzyskania lepszej kontroli procesu oraz większej funkcjonalności powłoki. Powłoki wielowarstwowe mogą być formowane poprzez sekwencyjne nakładanie na powierzchnię owoców roztworów polisacharydowych lub białkowych, ich wysuszenie, a następnie aplikację kolejnej warstwy, np. lipidowej. W wyniku takich połączeń, które zachodzą w matrycy biopolimerowej na powierzchni owoców, otrzymana powłoka wykazuje właściwości zbliżone do kompozytowej powłoki emulsyjnej, która występuje w postaci jednej emulsji i nanoszona jest jednorazowo na powierzchnię produktu. Rozszerzeniem metody zanurzeniowej jest technika impregnacji próżniowej, w której owoce są zanurzane w roztworze powlekającym w hermetycznej komorze próżniowej, połączonej z pompami próżniowymi. Dotychczas metoda ta znajduje zastosowanie głównie w przypadku całych, świeżych owoców (Nor i Ding 2020).

W metodzie osadzania warstwa po warstwie istotną rolę odgrywają oddziaływania elektrostatyczne, zachodzące pomiędzy powierzchnią żywności a naładowanymi polielektrolitami, co sprzyja poprawie adhezji powłoki do powierzchni żywności. Zastosowanie tej metody umożliwia formowanie dwóch lub większej liczby cienkich warstw, połączonych ze sobą w sposób fizyczny lub chemiczny, co prowadzi do poprawy skuteczności powłok w porównaniu z powłokami konwencjonalnymi. Wykazano, że metoda ta jest szczególnie efektywna w okresie pozbiorczego przechowywania owoców, przyczyniając się do zwiększenia szczelności powłok ochronnych (Pham i wsp. 2023).

Dotychczasowe badania jednoznacznie wskazują, że zastosowanie powłok jadalnych może skutecznie przyczynić się do utrzymania jakości świeżych owoców oraz wydłużenia ich okresu przydatności do spożycia (Maringgal i wsp. 2020). Pomimo tego nadal prowadzone są intensywne prace badawcze ukierunkowane na opracowanie nowych, naturalnych materiałów powłokotwórczych. Równoczesny rozwój technologii powlekania oraz poszukiwanie innowacyjnych surowców umożliwia stopniowe zwiększanie efektywności działania powłok jadalnych, bez negatywnego oddziaływania na zdrowie konsumentów. Ponadto połączenie powłok jadalnych z innymi metodami przetwarzania może istotnie wydłużyć trwałość pozbiorczą owoców, sprzyjając zachowaniu ich ogólnej jakości konsumpcyjnej. Z ekonomicznego punktu widzenia wdrażanie innowacyjnych technologii powlekania na skalę przemysłową pozostaje wciąż wyzwaniem, dlatego producenci owoców wciąż preferują tradycyjne metody przechowywania, uznawane za bardziej praktyczne i sprawdzone. W związku z tym dalsze badania koncentrują się na opracowaniu rozwiązań, umożliwiających wydłużenie okresu przydatności do spożycia świeżych owoców, bez negatywnego wpływu na ich cechy sensoryczne i wartość odżywczą oraz na adaptacji technik, znajdujących się na wczesnym etapie rozwoju w skali laboratoryjnej, do zastosowań przemysłowych.

### **1.3. Metody przedłużenia jakości pozbiorczej jabłek**

Pozbiorcze zapewnienie odpowiedniej jakości jabłek polega przede wszystkim na ich przechowywaniu w niskotemperaturowych komorach z kontrolowaną atmosferą. Pomimo stosowania warunków chłodniczych, podczas magazynowania owoców może dochodzić do rozwoju *Penicillium expansum*, tak zwanego patogenu pozbiorczego, który produkuje patulinę, czyli wtórny metabolit o działaniu toksycznym, prowadzący do znacznych strat jakościowych i ilościowych jabłek w trakcie przechowywania. Zanieczyszczenie mikrobiologiczne minimalnie przetworzonych produktów spożywczych może pojawiać się na różnych etapach łańcucha produkcyjnego, m.in. w sadzie, przy zbiorze, przechowywaniu, przetwarzaniu lub dystrybucji. Oprócz tego wpływ ma również niewłaściwa temperatura przechowywania, zbyt duża lub mała wilgotność otaczającego środowiska oraz uszkodzenia mechaniczne (Vieira i wsp. 2019).

Jednym z podstawowych warunków odpowiedniego utrwalania jabłek zaraz po zbiorze jest utrzymanie obniżonej temperatury przechowywania w całym łańcuchu chłodniczym (Sena i wsp. 2019), która wpływa na ograniczenie pojawiania się dwóch najczęściej występujących pleśni, wywołanych przez *Penicillium expansum* oraz *Botrytis*

*cinerea*. W celu redukcji strat pozbiorczych tradycyjnie stosowano chemiczne fungicydy, jednak ich wykorzystanie zostało ograniczone ze względu na obawy konsumentów, dotyczące negatywnego wpływu na zdrowie oraz środowisko naturalne. Obecnie duże zainteresowanie wzbudzają folie i powłoki jadalne, zawierające dodatek naturalnych związków o działaniu przeciwdrobnoustrojowym lub przeciwutleniającym (Usman i wsp. 2026).

Istotnym czynnikiem, determinującym jakość owoców w trakcie przechowywania jest skład gazowy atmosfery, który wpływa m.in. na zmiany aktywności przeciwutleniającej jabłek. Odpowiedni dobór warunków przechowywania umożliwia zachowanie wysokiej jakości owoców, przy jednoczesnym ograniczeniu zmian w ich składzie chemicznym. Liczne doniesienia literaturowe wskazują na skuteczność przechowywania owoców w komorach o obniżonej zawartości tlenu. Takie środowisko prowadzi do spowolnienia procesów metabolicznych, ograniczenia syntezy etylenu oraz ograniczenia zachodzenia procesów fermentacji (Butkeviciute i wsp. 2022).

Etylen ( $C_2H_4$ ) jest prostym gazowym węglowodorem, który reguluje procesy fizjologiczne roślin, m.in. kwitnienie, dojrzewanie i starzenie się owoców. Nawet w niskich temperaturach indukuje przemiany biochemiczne, przyspieszając metabolizm owoców i warzyw (Montero-Prado i Morales 2022), a wzrost jego stężenia powoduje zmiękczenie tkanek oraz degradację chlorofilu, co przyspiesza utratę ich jakości i psucie się produktów. Jabłka, wrażliwe na etylen, mogą być chronione przez powłoki jadalne, które opóźniają dojrzewanie (Jafarzadeh i wsp. 2021). Dodatkowo przechowywanie owoców klimakterycznych w atmosferze o wysokim stężeniu  $CO_2$  i obniżonym  $O_2$  ogranicza produkcję etylenu (Cortellino i wsp. 2015). W celu ograniczenia niekorzystnych zmian oraz poprawy atrakcyjności wizualnej owoców poprzez nadanie im połysku, coraz częściej przed wprowadzeniem ich do obrotu handlowego stosuje się technologię powlekania. Najwyższą skuteczność wykazują jadalne powłoki o złożonym składzie, ponieważ pojedyncze komponenty nie są wystarczające do istotnej poprawy trwałości oraz jakości owoców. Do najczęściej stosowanych dodatków, nadających świeżym jabłkom połysk, należą wosk, szelak oraz carnauba, których podstawową funkcją jest ograniczenie utraty masy. Substancje te, stosowane samodzielnie lub jako składniki emulsji, umożliwiają kontrolę ubytków wody, co sprzyja zachowaniu jakości świeżych owoców oraz spowolnieniu procesów dojrzewania. Ocena jakości świeżych owoców przez konsumentów opiera się głównie na cechach takich jak wygląd powierzchni skórki, jędrność oraz połysk. Niespełnienie tych kryteriów przez producentów może prowadzić do

obniżenia akceptacji produktu przez konsumentów, a nawet do jego całkowitego odrzucenia, co przekłada się na zmniejszenie opłacalności ekonomicznej (Thakur i wsp. 2019).

Niewłaściwe dobranie temperatury przechowywania prowadzi do pogorszenia jakości produktów oraz skrócenia okresu ich przydatności do spożycia, co skutkuje zwiększonymi stratami żywności. Zastosowanie niskich temperatur podczas przechowywania wpływa na zmniejszenie aktywności fizjologicznej tkanek oraz hamuje rozwój drobnoustrojów, odpowiedzialnych za psucie się żywności, przyczyniając się do wydłużenia trwałości pozbiorczej. Widoczne zmiany pogorszenia się jakości, takie jak więdnienie, kurczenie się czy wysychanie, mogą występować już przy 3-6% utracie wilgoci, przyczyniając się do znacznych strat jakości wielu gatunków owoców oraz warzyw (Nunes i wsp. 2009). Natomiast w przypadku przechowywania w warunkach konsumpcyjnych (pokojowych) zmiany jakości jabłek następują szybciej. W związku z tym powlekanie jabłek jest obiecującą metodą na spowolnienie niekorzystnych zmian jakościowych w czasie przechowywania. Dodatkowo wprowadzenie substancji funkcjonalnych wpływa na zwiększenie efektu powlekania i większą stabilność owoców.

Przykładem wpływu powlekania z dodatkiem aktywnym na jakość jabłek są badania Synowiec i wsp. (2014), w których jabłka odmiany *Jonagored* powlekano jadalną powłoką pullulanową oraz pullulanową z dodatkiem ekstraktu z bazylii. Wykazano, że proces powlekania istotnie wpływał na cechy wizualne owoców – jabłka pokryte powłoką zawierającą ekstrakt z bazylii charakteryzowały się matową powierzchnią oraz wyraźnie wyczuwalnym posmakiem bazylii. Ponadto stwierdzono, że powłoka pullulanowa z dodatkiem ekstraktu z bazylii zapewniała skuteczną ochronę przeciwgrzybiczą wobec gatunku *Rhizopus arrhizus*. Po 30-dniowym okresie przechowywania w temperaturze chłodniczej masa jabłek powlekanych była istotnie mniejsza w porównaniu z owocami niepowlekаныmi, co wskazuje na ograniczenie strat masy podczas przechowywania.

De León-Zapata i wsp. (2017) opracowali nanostrukturalne powłoki na bazie wosku candelilla i ekstraktu z *Flourensia cernua*, których zastosowanie miało na celu spowolnienie procesu dojrzewania jabłek odmiany *Fuji* podczas 56-dniowego okresu przechowywania. Po zoptymalizowaniu właściwości zwilżalności nanopowłoki oraz sposobu jej aplikacji na powierzchnię owoców przeprowadzono ocenę zmian fizykochemicznych, przepuszczalności pary wodnej oraz stabilności mikrobiologicznej jabłek. Powlekanie ograniczyło utratę jędrności oraz spowolniło zmiany fizykochemiczne i mikrobiologiczne, w tym rozwój grzybów i drożdży, w porównaniu do owoców

niewpewnych.

Duong i wsp. (2022) prowadzili badania nad zastosowaniem folii jadalnych na bazie alginianu sodu do powlekania jabłek gatunku 'Tabtimchan' (*Syzygium samarangense*), które zawierają liczne związki biologiczne czynne, w tym przeciwutleniacze, wykazujące korzystny wpływ na zdrowie człowieka. Liczne hipotezy wskazują, że ograniczenie uszkodzeń owoców wrażliwych na przechowywanie w temperaturach poniżej 10-12°C może być związane ze wzmocnieniem systemów antyoksydacyjnych lub utrzymaniem integralności komórkowej, co prowadzi do obniżenia aktywności fosfolipazy oraz lipooksygenazy. Ponadto alginianowe folie rozpuszczalne w wodzie mogą zostać przekształcone w struktury nierozpuszczalne poprzez sieciowanie z udziałem jonów dwu- lub wielowartościowych, takich jak  $\text{Ca}^{2+}$ . Dodatek substancji sieciujących, w tym chlorku wapnia ( $\text{CaCl}_2$ ), zwiększa wytrzymałość mechaniczną folii, ogranicza przenikanie pary wodnej i tlenu. Chlorek wapnia jest powszechnie stosowany w przetwórstwie owocowym, ponieważ wpływa na utrzymanie jędrności owoców. Z badań wynika, że powłoka na bazie alginianu sodu usieciowana jonami  $\text{Ca}^{2+}$  stanowi obiecującą alternatywę dla zabiegów chemicznych, ograniczając występowanie brązowych plam i kurczenie się owoców. Dzięki tworzeniu wiązań z jonami  $\text{Ca}^{2+}$ , zastosowanie  $\text{CaCl}_2$  może również ograniczać aktywność enzymów, odpowiedzialnych za zmiękczenie i oddychanie, takich jak poligalakturonaza (Anjani i wsp. 2021).

Skuteczność zachowania składników bioaktywnych, w tym witaminy C oraz całkowitej zawartości flawonoidów i antocyjanów, uzyskano również poprzez powlekanie owoców żelem *Aloe vera*. Powłoki nakładano w różnych stężeniach (50, 75 i 100 v/v %) na woskowane jabłka, które następnie przechowywano przez 12 dni w temperaturze 10°C. Obniżone utlenianie kwasu askorbinowego było prawdopodobnie wynikiem niskiej przepuszczalności powłok aloesowych, co skutkowało zmniejszeniem aktywności enzymatycznej. Zaobserwowano także ograniczenie ubytku masy owoców oraz spowolnienie wzrostu kwasowości miareczkowej, co wskazuje na opóźnienie pogarszania się jakości przechowywanych jabłek. Utrata wilgoci mogła być związana z higroskopijnymi właściwościami żelu *Aloe vera*, który tworzył barierę ograniczającą migrację wody między owocem a otoczeniem (Mubarak i Engakanah 2017).

Belay i wsp. (2023) wykazali, że pokrywanie jabłek odmiany *Granny Smith* roztworami powłokotwórczymi, zawierającymi zeinę lub mieszaninę zeiny w połączeniu z niziną, spowolniło utratę masy owoców w porównaniu do jabłek niewpewnych. Po 21 dniach przechowywania owoców w temperaturze 15°C odnotowano 30% zmniejszenie

początkowej masy jabłek niepowlekanych, 25% zmniejszenie masy jabłek powlekanych zeiną i nizyną, natomiast 17% zmniejszenie masy wystąpiło w przypadku owoców pokrytych zeiną. Redukcja ubytku masy była związana ze zdolnością powłoki do tworzenia zmodyfikowanej bariery dla pary wodnej poprzez blokowanie porów naturalnie obecnych w skórce. Najwyższą jędrność zachowały owoce powlekane zeiną, co przypisuje się ograniczeniu aktywności enzymów rozkładających ściany komórkowe, w tym pektynesterazy i poligalakturonazy. Wszystkie owoce, zarówno powlekane, jak i niepowlekane, utrzymały akceptowalną barwę i nie wykazywały oznak psucia. Dodatkowo powlekanie spowolniło rozwój drobnoustrojów na powierzchni owoców w porównaniu z próbkami kontrolnymi.

Jung i Choi (2021) oceniali jadalne powłoki ochronne na jabłkach odmian *Tsugaru*, *Summer King* i *Shinano Gold*, oparte na monoestrach sacharozy z kwasami tłuszczowymi i etanolem. Po 28 dniach przechowywania w temperaturze pokojowej powłoka na owocach *Shinano Gold* wykazywała obniżoną adhezję i nieciągłość struktury, co przypisano naturalnej porowatości skórki, różnicom w składzie kutykuli oraz lokalnemu nagromadzeniu wosków. W przypadku odmian *Tsugaru* i *Summer King* powłoki pozostały ciągłe, a owoce utrzymały jędrność i stabilną barwę.

#### **1.4. Podsumowanie**

Biorąc pod uwagę ilość generowanych odpadów i poziom marnotrawienia żywności podejmowane są intensywne prace nad poszukiwaniem materiałów opakowaniowych przyjaznych środowisku oraz metod poprawy stabilności żywności. Folie z biopolimerów i powlekanie żywności wpisują się w aktualne założenia gospodarki cyrkularnej. W związku z czym niniejsza praca dotyczy wytworzenia, analizy właściwości fizykochemicznych i zastosowania jadalnych folii opakowaniowych wytworzonych z pektyny jabłkowej. Pektyna jako naturalny polisacharyd występujący w ścianach komórkowych owoców cechuje się dobrymi właściwościami foliotwórczymi i powłokotwórczymi oraz kompatybilnością w połączeniu z substancjami funkcjonalnymi. Na podstawie studium literaturowego postawiono hipotezy badawcze i zdecydowano o planie badań nad zastosowaniem wybranych kwasów fenolowych o działaniu przeciwutleniającym jako związki aktywne w foliach pektynowych oraz ich zastosowaniu w formie powłok ochronnych w zachowaniu jakości jabłek w czasie przechowywania.

## 2. CEL PRACY I HIPOTEZY BADAWCZE

Celem głównym niniejszej rozprawy doktorskiej było opracowanie oraz kompleksowa charakterystyka fizykochemiczna folii na bazie pektyny jabłkowej, w tym folii modyfikowanych dodatkiem wybranych kwasów fenolowych. Zakres badań obejmował określenie wpływu zastosowanych kwasów fenolowych na właściwości optyczne, sorpcyjne, zwilżające, strukturalne, mechaniczne, termiczne oraz przeciwutleniające otrzymanych materiałów, a także analizę zależności pomiędzy strukturą folii, jej właściwościami użytkowymi i zakładaną funkcją. Ponadto dokonano oceny możliwości zastosowania opracowanych materiałów biokompozytowych jako aktywnych powłok ochronnych do monitorowania jakości pozbiorczej jabłek odmiany *Golden Delicious* podczas przechowywania, jak również scharakteryzowano właściwości reologiczne roztworów powłokotwórczych oraz przeanalizowano kinetykę uwalniania wybranych kwasów fenolowych.

### **Przeprowadzone badania miały na celu weryfikację następujących hipotez:**

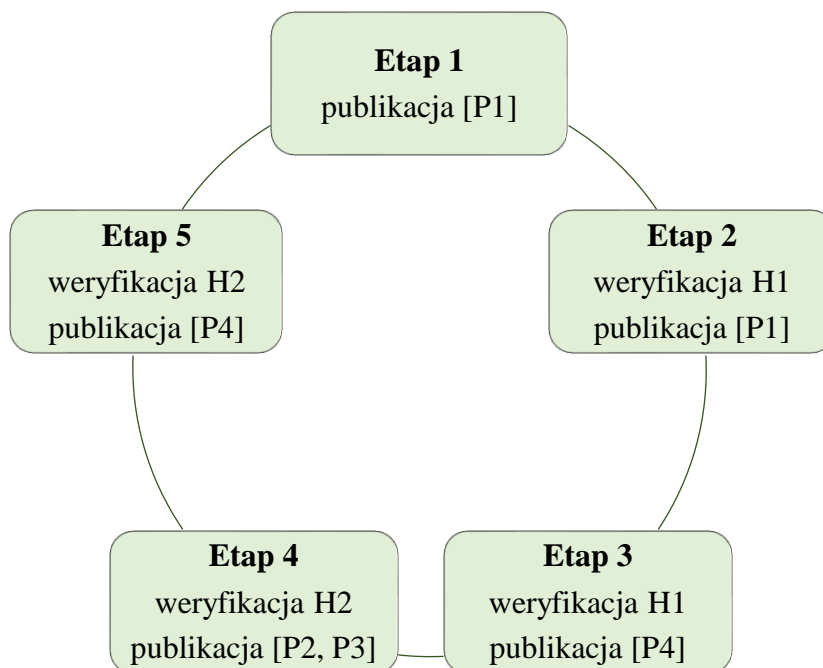
**H1.** Wprowadzenie substancji aktywnych w postaci kwasów fenolowych wpłynie pozytywnie na poprawę właściwości funkcjonalnych i użytkowych folii pektynowych. Odpowiedni dobór składników folii, takich jak polisacharydy, glicerol i związki o właściwościach przeciwutleniających, oraz ich właściwe proporcje umożliwiają otrzymanie aktywnego materiału ochronnego w postaci jadalnej powłoki o pożądanych właściwościach użytkowych, m.in. przezroczystości, wytrzymałości mechanicznej i barierowości wobec gazów, o wysokiej aktywności przeciwutleniającej oraz równomiernym uwalnianiu składnika aktywnego [P2, P3, P4].

**H2.** Zastosowanie roztworów powłokotwórczych o zoptymalizowanym składzie do ochrony jabłek w czasie przechowywania w warunkach pokojowych wpłynie na spowolnienie procesu dojrzewania owoców oraz spowoduje zachowanie ich odpowiedniej jakości i przydatności konsumpcyjnej [P4].

### 3. ORGANIZACJA BADAŃ, MATERIAŁY I METODYKA

#### 3.1. Organizacja badań

Badania będące podstawą naukową niniejszej rozprawy były prowadzone w pięciu głównych etapach, które pozwoliły na weryfikację postawionych hipotez badawczych. Ogólny plan badawczy przedstawiono na **Rysunku 2**.



**Rysunek 2.** Organizacja etapów badań oraz publikacji, w których weryfikowano poszczególne hipotezy badawcze oraz prezentowano wyniki: **etap 1** – analiza bibliograficzna dostępnej literatury na temat pektynowych folii jadalnych wzbogacanych w kwasy fenolowe; **etap 2** – opracowanie i wyselekcjonowanie materiału badawczego podczas badań wstępnych oraz wybór wariantów recepturowych do dalszych badań; **etap 3** – przeprowadzenie badań dotyczący reologii roztworów powłokotwórczych; **etap 4** – analiza właściwości fizycznych i przeciwutleniających folii oraz uwalniania substancji w czasie; **etap 5** – przeprowadzenie badań, dotyczących kontrolnych oraz powlekanych owoców oraz analizy sensorycznej, a także próby przechowalnicze w warunkach pokojowych, mające na celu określenie okresu przydatności do spożycia powlekanych jabłek

**Etap I** obejmował przegląd i opracowanie literaturowe. Proces zbierania danych opierał się na kompleksowym zgromadzeniu najbardziej istotnej i najnowszej literatury, dotyczącej powlekania świeżych jabłek różnego rodzaju roztworami powłokotwórczymi. Rezultaty przeglądu badań naukowych przeprowadzonych w kraju i na świecie skutkowały

powstaniem publikacji przeglądowej [P1]. Oprócz tego nakreślono założenia do postawienia hipotez badawczych, których weryfikację przedstawiono w rozdziale 4 niniejszego opracowania, opisując wyniki uzyskane podczas przeprowadzonych badań.

W **etapie II** celem badań wstępnych było opracowanie metodyki otrzymywania pektynowych jadalnych folii z dodatkiem wybranych kwasów fenolowych oraz dobór składników w odpowiednich proporcjach, tj. biopolimeru i plastyfikatora, jak również dobranie odpowiednich warunków suszenia, z uwzględnieniem możliwości zastosowania aplikatora warstw do tworzenia powierzchni matrycy materiału biokompozytowego. Założeniem było wybranie odpowiedniego rodzaju kwasu fenolowego oraz jego stężenia na takim poziomie, by aktywny dodatek nie modyfikował w złym kierunku właściwości mechanicznych wytwarzanych jadalnych folii, a jednocześnie inhibitował wzrost drobnoustrojów epifitycznych owoców pokrytych powłoką ochronną i zachowywał ich walory jakościowe. W celu doboru odpowiedniego stężenia kwasów fenolowych wykonano szereg badań modelowych, oznaczając aktywność przeciwdrobnoustrojową folii wobec wybranych gatunków bakterii i drożdży, w podłożach stałych i płynnych. Dodatkowo oceniono aktywność przeciwutleniającą folii wzbogaconych kwasami fenolowymi. Wyżej opisane eksperymenty stanowiły dodatkowy element badawczy pracy doktorskiej i nie były uwzględnione w jej planie. Stanowią one podstawę publikacji naukowej, która nie wchodzi w skład niniejszej rozprawy doktorskiej i jest obecnie w recenzji. W ramach dalszych badań tego etapu opracowano podstawową recepturę jadalnych folii z dodatkiem wybranych kwasów fenolowych, pełniących funkcję składników aktywnych. W trakcie optymalizacji parametrów procesu, w tym grubości folii, przeprowadzono ocenę wizualną otrzymanych materiałów biokompozytowych, na podstawie której wykluczono dwa warianty modyfikacji z udziałem kwasu wanilinowego i syryngowego. Materiały te nie zostały zakwalifikowane do dalszych badań ze względu na tworzenie niejednorodnych struktur folii ochronnych. Ocena możliwości zastosowania kwasów fenolowych pozwoliła na wybranie sześciu kwasów fenolowych, stanowiących składnik roztworów powłokotwórczych, w połączeniu z niskometylowaną pektyną jabłkową (nazywaną dalej również pektyną jabłkową i/lub pektyną). Ostatecznie, bazę powłokotwórczą do wytwarzania folii stanowiła pektyna jabłkowa w stężeniu 5%, jeden z sześciu wybranych kwasów fenolowych (ferulowy, galusowy, kawowy, kumarowy, protokatechowy, synapinowy) w stężeniu 5% względem pektyny, stanowiący czynnik zmienny, woda destylowana oraz plastyfikator, jakim był glicerol (w ilości 50% w stosunku do pektyny jabłkowej).

**Etap III** polegał na określeniu właściwości reologicznych przygotowanych roztworów powłokotwórczych oraz zbadaniu kinetyki uwalniania wybranych kwasów fenolowych na przykładzie kwasu kawowego i protokatechowego, zastosowanych do powlekania jabłek. Wyniki z tego etapu badań zostały opublikowane w artykule [P4] i posłużyły do uzupełnienia weryfikacji **hipotezy 1**.

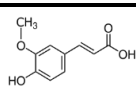

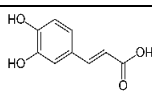
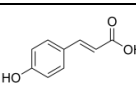
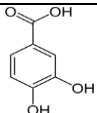
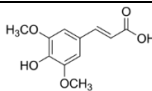
**Etap IV** obejmował realizację zadań badawczych, dotyczących materiału doświadczalnego opracowanego w ramach etapu II. Otrzymane jadalne folie poddano analizie podstawowych właściwości fizycznych, a wyniki przeprowadzonych badań wybranych wariantów próbek przedstawiono w artykułach [P2] oraz [P3]. Uzyskane rezultaty posłużyły do weryfikacji **hipotezy badawczej 1**. Analiza uzyskanych wyników badań polegała na określeniu wpływu dodatku kwasów fenolowych na właściwości fizykochemiczne folii jadalnych na bazie pektyny. W tym celu zostały przeprowadzone podstawowe badania cech charakteryzujących biopolimery kompozytowe w formie jadalnych folii, tj. grubość, zawartość wody, pęcznienie i rozpuszczalność w wodzie, oraz oceniono właściwości zwilżające (pomiar kąta zwilżania wody) i sorpcyjne (kinetyka i izotermy adsorpcji wody), optyczne, barierowe, mechaniczne i strukturalne (mikrostruktura i spektrometria w podczerwieni z transformacją Fouriera) folii.

**Etap V** polegał na zbadaniu wpływu powlekania świeżych jabłek pektynowymi roztworami powłokotwórczymi z dodatkiem wybranych kwasów fenolowych (kawowego i protokatechowego) na wyróżniki jakościowe owoców w czasie przechowywania. Ponadto, zbadano wpływ procesu powlekania na właściwości świeżych jabłek i możliwości wydłużenia ich okresu przechowywania, a także przydatności konsumpcyjnej. Wybrane wyróżniki jakościowe owoców badano zarówno bezpośrednio, jak i co 7 dni w trakcie 28-dniowego okresu przechowywania w temperaturze  $21 \pm 1^\circ\text{C}$  oraz przy względnej wilgotności powietrza wynoszącej  $35 \pm 5\%$ . Analizy obejmowały ocenę ubytku masy owoców w czasie, zmian kwasowości, pH i zawartości ekstraktu, a także jędrności oraz intensywności procesu oddychania. Zarówno jabłka stanowiące próbę kontrolną, jak i owoce powlekane poddano ocenie sensorycznej, obejmującej opiniowanie smaku, zapachu, barwy, jędrności oraz ogólnej jakości owoców. Wyniki badań umieszczono w publikacji [P4] i na ich podstawie zweryfikowano **hipotezę 2**.

### 3.2. Materiał badawczy

W zależności od etapu prowadzonych badań materiał stanowiły: roztwory powłokotwórcze; folie jadalne oraz jabłka odmiany *Golden Delicious*, pochodzące z Sadu Doświadczalnego SGGW w Warszawie, które w celu przeprowadzenia doświadczeń przechowalniczych pokryto odpowiednio zoptymalizowanymi powłokami ochronnymi na bazie pektyny z dodatkiem wybranych aktywnych składników. Folie wytworzono z wodnych roztworów pektyny jabłkowej, glicerolu (plastyfikator) i wybranych kwasów fenolowych. Charakterystykę zastosowanych kwasów fenolowych przedstawiono w Tabeli 1.

**Tabela 1.** Ogólna charakterystyka kwasów fenolowych zastosowanych do otrzymywania aktywnych folii pektynowych.

Kwas fenolowy	Skrót	Nazwa	Wzór sumaryczny	Wzór strukturalny	Producent
kwasy ferulowy	<b>FRA</b>	kwasy 4-hydroksy-3-metoksycynamonowy	$C_{10}H_{10}O_4$		POL- AURA
kwasy galusowy	<b>GLA</b>	kwasy 3,4,5-trihydroksybenzoesowy	$C_7H_6O_5$		POL- AURA
kwasy kawowy	<b>CFA</b>	kwasy 3,4-dihydroksycynamonowy	$C_9H_8O_4$		POL- AURA
kwasy kumarowy	<b>CMA</b>	4-hydroxycinnamic acid	$C_9H_8O_3$		POL- AURA
kwasy protokatechowy	<b>PCA</b>	3,4-dihydroksybenzoic acid	$C_7H_6O_4$		Thermo Scientific
kwasy synapinowy	<b>SNA</b>	3,5-dimethoxy-4-hydroxycinnamic acid	$C_{11}H_{12}O_5$		Acros Organics

### 3.3. Przygotowanie roztworów folio/powłokotwórczych

Wodne roztwory folio/powłokotwórcze zostały wytworzone o stężeniu 5% pektyny jabłkowej (Pektowin S.A., Jasło, Polska) o stopniu estryfikacji od 27 do 32% i stopniu amidacji od 18 do 23% wraz z dodatkiem plastyfikatora, jakim był glicerol (Avantor Performance Materials, Gliwice, Polska) w ilości 50% w stosunku do masy pektyny jabłkowej. Po połączeniu pektyny jabłkowej, wybranego kwasu fenolowego oraz

wody destylowanej roztwory mieszano przez 20 minut, podgrzewając je do temperatury 60°C z zastosowaniem płyty grzejnej RCT basic IKAMAG (IKA – Werke Gmn & Co., Staufen, Niemcy) i mieszadła magnetycznego, obracającego się z prędkością 600 obr./min. Po ostudzeniu roztworów dodawano glicerol (w ilości 50% w stosunku do pektyny jabłkowej).

### 3.3.1. Właściwości reologiczne roztworów foliotwórczych

Właściwości reologiczne roztworów określono z użyciem reometru Haake MARS 40 (Thermo Scientific Inc., Waltham, MA, USA). Roztwory badano w temperaturze 25°C w układzie współosiowych cylindrów (CC25DIN/Ti) przy szybkości ścinania rosnącej liniowo do 100 s<sup>-1</sup>. Oznaczenie przeprowadzono w trzech powtórzeniach dla roztworów powłokotwórczych zastosowanych w powlekanii jabłek, czyli dla mieszaniny kontrolnej oraz z dodatkiem kwasu kawowego lub protokatechowego. Krzywe płynięcia dopasowano za pomocą modelu Ostwalda de Waele według poniższego wzoru:

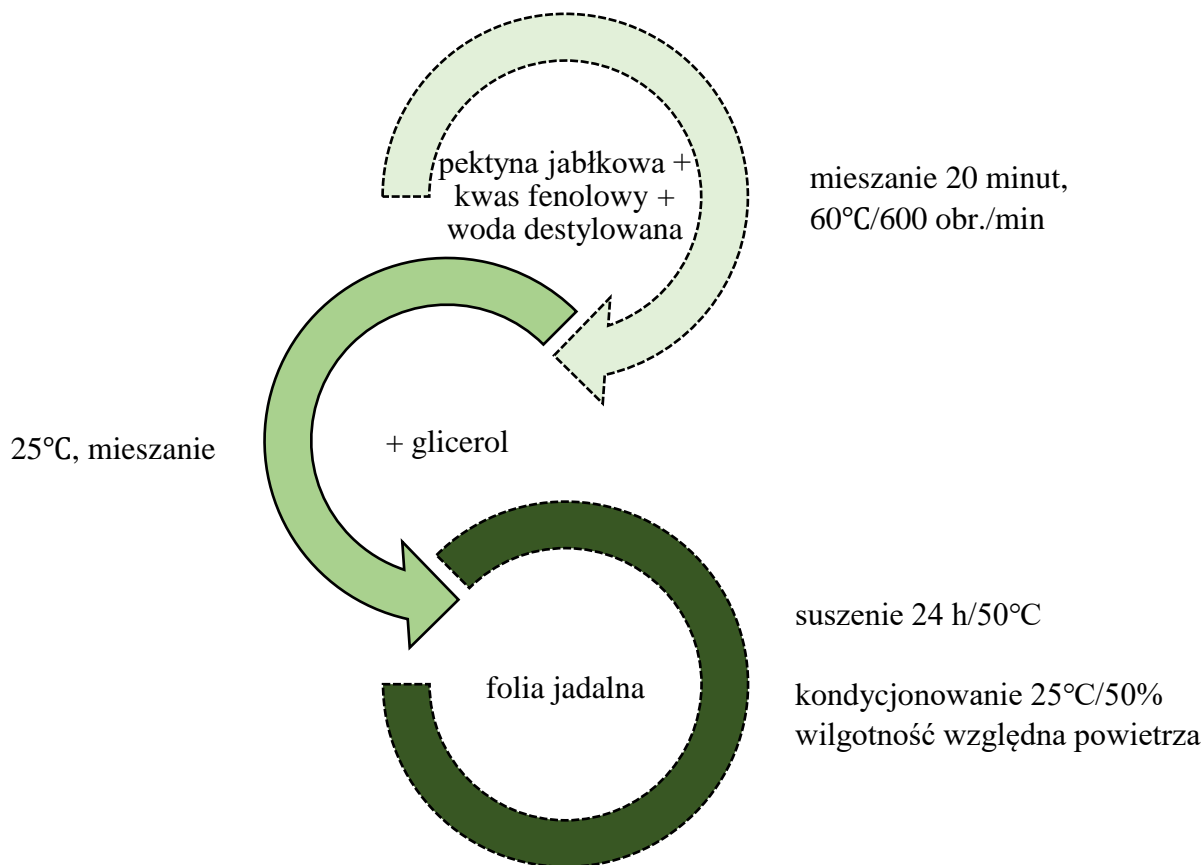
$$\tau = K \cdot \dot{\gamma}^n$$

gdzie:  $\tau$  – naprężenie ścinające (Pa),  $\dot{\gamma}$  – szybkość ścinania (s<sup>-1</sup>),  $K$  – współczynnik konsystencji (Pa·sn),  $n$  – bezwymiarowy wskaźnik płynięcia.

### 3.4. Wytworzenie folii pektynowych

Roztwory folio/powłokotwórcze, zarówno kontrolne, jak i z dodatkiem wybranych kwasów fenolowych, nakładano w warstwie o grubości 2500 μm na arkusze przy użyciu automatycznego aplikatora do folii Zehntner ZAA 2300 (Zehntner GmbH Testing Instruments, Sissach, Szwajcaria), poruszającego się z prędkością 10 mm/s. Folie suszono w suszarce laboratoryjnej SUP-65W (Wamed, Warszawa, Polska) przez 24 h w temperaturze 50°C. Następnie wysuszone na arkuszach aplikacyjnych folie kondycjonowano w komorze termostatycznej typu KFB 720 (Binder, Tuttlingen, Niemcy) w temperaturze 25°C i wilgotności względnej powietrza wynoszącej 50%. Schemat otrzymywania folii jadalnych przedstawiono na **Rysunku 3**.

Przed przystąpieniem do badania kinetyki oraz izoterm adsorpcji pary wodnej folie ponownie dosuszono w temperaturze 30°C i ciśnieniu 1,5 kPa przez 48 godzin, używając suszarki próżniowej (Memmert V0 500, Memmert GmbH, Schwabach, Niemcy). Wysuszone próbki przechowywano następnie w ekssykatorze z pięciotlenkiem difosforu (P<sub>2</sub>O<sub>5</sub>).



**Rysunek 3.** Schemat otrzymywania aktywnych folii pektynowych

### 3.5. Metody badania folii pektynowych

#### 3.5.1. Pomiar grubości

Grubość folii pektynowych mierzono za pomocą grubościomierza (Thwing-Albert, ProGage Thickness Tester, USA) z dokładnością do 1  $\mu\text{m}$ . Pomiary wykonywano w kilku miejscach próbki, w co najmniej trzech powtórzeniach, w zależności od badanych właściwości.

#### 3.5.2. Oznaczenie zawartości wody

Oznaczenie zawartości wody w foliach pektynowych przeprowadzono metodą suszarkową (105°C) do stałej masy w suszarce laboratoryjnej (SUP 65 WG, WAMED, Warszawa, Polska). Pomiar masy przeprowadzono z dokładnością  $\pm 0,0001$  g, stosując wagę analityczną (RADWAG PS 600/C/2, Radom, Polska).

### 3.5.3. Oznaczenie pęcznienia w wodzie

Przygotowane próbki z folii pektynowych o wymiarach  $2 \times 2$  cm zważono na wadze analitycznej (RADWAG PS 600/C/2, Radom, Polska) z dokładnością do  $\pm 0,0001$  g. Następnie umieszczono je w 25 ml wody destylowanej na czas wynoszący 2 minuty, po upływie którego folie odsączono z nadmiaru wody przy użyciu bibuły filtracyjnej i ponownie zważono. Pomiar został powtórzony trzykrotnie. Zdolność do pęcznienia badanych materiałów w formie folii ( $P$ ) obliczono według wzoru:

$$P = \frac{m_2 - m_1}{m_1} \cdot 100\%$$

gdzie:  $m_1$  – masa próbki przed zanurzeniem w wodzie (g),  $m_2$  – masa próbki po zanurzeniu w wodzie (g).

### 3.5.4. Oznaczenie rozpuszczalności w wodzie

Próbki folii pektynowych o wymiarach  $2 \times 2$  cm umieszczono w szklanych naczynkach, ważono na wadze analitycznej (RADWAG PS 600/C/2, Radom, Polska) z dokładnością do  $\pm 0,0001$  g i suszono w suszarce laboratoryjnej (SUP 65 WG, WAMED, Warszawa, Polska) przez 24 godziny w temperaturze  $105^\circ\text{C}$ . Po wystudzeniu folie ponownie ważono, umieszczono w 25 ml wody destylowanej i przechowywano przez 24 h, mieszając kilkakrotnie w tym czasie. Następnie nadmiar wody z próbek usunięto przy pomocy bibuły filtracyjnej i suszono je ponownie przez 24 h w suszarce laboratoryjnej (SUP 65 WG, WAMED, Warszawa, Polska) w temperaturze  $105^\circ\text{C}$ . Pomiar został wykonany w trzech powtórzeniach, a rozpuszczalność w wodzie ( $R$ ) folii wyznaczono według wzoru:

$$R = \frac{m_0 - m_r}{m_0} \cdot 100\%$$

gdzie:  $m_0$  – sucha masa próbki przed rozpuszczeniem (g),  $m_r$  – sucha masa próbki po rozpuszczeniu (g).

### 3.5.5. Kinetyka adsorpcji pary wodnej

Kinetykę adsorpcji pary wodnej określono na podstawie zmian masy próbek folii pektynowych ( $\pm 0,25$  g, dokładność  $\pm 0,0001$  g; RADWAG PS 600/C/2, Radom, Polska) w zakresie od 0 do 120 h przy 100% RH (woda destylowana). Krzywe kinetyczne przedstawiały zależność ilości zaadsorbowanej wody (g/g suchej substancji) w funkcji

czasu  $t$  (h), a współczynnik dyfuzji pary wodnej obliczono na podstawie drugiego prawa Ficka (Crank, 1975):

$$\frac{M_t - M_0}{M_e - M_0} = 1 - \sum_{n=0}^{\infty} \frac{8}{(2n+1)^2 \pi^2} \exp \left[ -\frac{(2n+1)^2 \pi^2}{4L^2} Dt \right]$$

gdzie:  $M_t$  – całkowita ilość zaadsorbowanej wody (g/g s.s.) w czasie  $t$  (s),  $M_0$  – początkowa zawartość wody (g/g s.s.),  $M_e$  – zawartość wody w czasie  $t$  (g/g s.s.),  $L$  – grubość folii (m).

### 3.5.6. Izotermy adsorpcji pary wodnej

Izotermy adsorpcji pary wodnej folii pektynowych wyznaczono przy użyciu aparatu do dynamicznego pomiaru adsorpcji pary wodnej (Aquadyne DVS-2HT, Quantachrome Instruments by Anton Paar sp. z o.o., Warszawa, Polska). Pomiary dokonano w dwóch powtórzeniach w zakresie wilgotności względnej środowiska od 0 do 75%. Dane eksperymentalne opracowano przy zastosowaniu programu Microsoft Excel 2000 i oprogramowania aquaWIN (Air3).

### 3.5.7. Pomiar kąta zwilżania wody

Analizę kąta zwilżania folii pektynowych przeprowadzono przy użyciu goniometru OCA 25 (DataPhysics Instruments, Filderstadt, Niemcy) metodą naniesionej kropli. Na powierzchnię folii aplikowano 10  $\mu$ l wody destylowanej z prędkością 10  $\mu$ l/s. Pomiar wykonywano w co najmniej sześciu powtórzeniach, w czasie 0 s oraz po 60 s. Wyniki opracowano przy użyciu oprogramowania SCA20\_U.

### 3.5.8. Obserwacje mikrostruktury

Obserwacji powierzchni ( $\times 600$ ) oraz przekrojów ( $\times 800$ ) otrzymanych folii pektynowych dokonano przy zastosowaniu skaningowego mikroskopu elektronowego TM3000 (Hitachi High Tech, Tokio, Japonia). Przygotowane próbki o wymiarach 5  $\times$  5 mm umieszczono na stole pomiarowym, mocując je przy użyciu taśmy węglowej PELCO o średnicy 9 mm (Pik Instruments Sp. z o.o., Piaseczno, Polska). Pomiar został przeprowadzony w warunkach niskiej próżni, wynoszącej 0,35-1 tora.

### 3.5.9. Pomiar barwy

Barwę folii zmierzono kolorymetrem CR-400 (KONICA MINOLTA, INC., Tokio, Japonia) w systemie CIE Lab\*, wykonując sześć powtórzeń. Standardem odniesienia była

folia otrzymana z pektyny jabłkowej bez aktywnych dodatków. Dla poprawności przeprowadzonej interpretacji uzyskanych wyników, oceny współrzędnych chromatycznych barwy z uwzględnieniem materiału odniesienia, obliczono bezwzględną różnicę barwy zgodnie ze wzorem, zaproponowanym przez Sobrala i wsp. (2005):

$$\Delta E = \sqrt{(L^* - L)^2 + (a^* - a)^2 + (b^* - b)^2}$$

gdzie:  $\Delta E$  - całkowita różnica barwy,  $L^*$ ,  $a^*$ ,  $b^*$  - pomiary dla standardu (folia z pektyny jabłkowej),  $L$ ,  $a$ ,  $b$  - pomiary dla folii.

### 3.5.10. Pomiar nieprzezroczystości

Do obliczenia nieprzezroczystości folii pektynowych dokonano pomiaru średniej grubości folii oraz absorbancji przy długości fali 600 nm, którą zmierzono za pomocą spektrofotometru EVOLUTION 220 UV (Thermo SCIENTIFIC, Waltham, USA) i oprogramowania Thermo INSIGHT (wersja 2.5). Pomiar wykonano w sześciu powtórzeniach, a nieprzezroczystość folii ( $O$ ) obliczono przy użyciu wzoru:

$$O = \frac{A_{600}}{l}$$

gdzie:  $A_{600}$  – wartość absorbancji przy długości fali 600 nm,  $l$  – grubość folii (mm).

### 3.5.11. Określenie transmitancji światła UV-VIS

Transmitancja światła UV-VIS (%) przez folie pektynowe została określona poprzez pomiar w zakresie długości fali wynoszących 200-800 nm, przy zastosowaniu spektrometru EVOLUTION 220 (Thermo Electron Corporation, Waltham, MA, USA) oraz adaptera do folii. Wyniki zostały opracowane z zastosowaniem oprogramowania Thermo INSIGHT.

### 3.5.12. Pomiar połysku

Połysk folii pektynowych badano pod trzema różnymi kątami, wynoszącymi 20, 60 i 85°, w dziesięciu powtórzeniach z dokładnością wynoszącą  $\pm 0,2^\circ$  przy użyciu urządzenia Multi Gloss 268A (Konica Minolta, Tokio, Japonia). Czułość widmową stanowiła przybliżona funkcja CIE  $y(2^\circ)$  dla źródła CIE C.

### 3.5.13. Badanie właściwości mechanicznych

Cechy wytrzymałościowe folii pektynowych, takie jak wytrzymałość na rozciąganie ( $TS$ , MPa), wydłużenie względne przy zerwaniu ( $E$ , %) oraz moduł Younga ( $MY$ , MPa), określono za pomocą teksturometru TA-XT2i (Stable Micro Systems Ltd, Surrey, Wielka Brytania). Pomiedzy szczękami teksturometru, znajdującymi się w odległości 25 mm, umieszczano próbki folii o wymiarach  $25 \times 100$  mm. Szczęki teksturometru rozsuwały się z prędkością 1 mm/s. Dla każdego rodzaju folii wykonano minimum sześć powtórzeń, a obliczenia przeprowadzono według poniższych wzorów:

$$TS = \frac{F_{max}}{A}$$

gdzie:  $F_{max}$  – siła powodująca zerwanie folii (N),  $A$  – pole powierzchni przekroju folii ( $\text{mm}^2$ ).

$$E = \frac{\Delta l}{l_0} * 100\%$$

gdzie:

$\Delta l$  – wydłużenie próbki, przy którym nastąpiło zerwanie folii (mm),  $l_0$  – długość początkowa próbki (mm).

$$MY = \frac{F_b}{A} * \frac{\Delta l}{l_0}$$

gdzie:  $F_b$  – siła powodująca odkształcenie w punkcie b, stanowiącym granicę liniowej plastyczności (N),  $\Delta l$  – wydłużenie próbki, przy którym nastąpiło zerwanie folii (mm),  $A$  – pole powierzchni przekroju folii ( $\text{mm}^2$ ),  $l_0$  – długość początkowa próbki (mm).

### 3.5.14. Badanie właściwości barierowych

Przepuszczalność pary wodnej folii pektynowych zbadano w trzech powtórzeniach metodą grawimetryczną z zastosowaniem analizatora CupMaster FX 3180-CM15 (Textest AG, Schwerzenbach, Szwajcaria). Pomiar odbył się w temperaturze  $25^\circ\text{C}$  z zastosowaniem miseczek testowych o powierzchni wymiany masowej  $28,3 \text{ cm}^2$  przy różnicy wilgotności względnych 50-100% i prędkości powietrza 2 m/s. Przepuszczalność folii pektynowych dla tlenu i ditlenku węgla badano w trzech powtórzeniach przy użyciu testera przepuszczalności gazów C130 (Labthink Instruments Co., Ltd., Jinan, Chiny) metodą manometryczną, zgodnie z normą ASTM D1434-82. Z folii wycięto krążki o średnicy 12 cm, a następnie zmierzono ich grubość. Próbki umieszczono na filtrze w systemie próżniowym, w którym tlen lub ditlenek węgla dyfundował przez folię do komory

pomiarowej. Badania prowadzono w warunkach  $22 \pm 1^\circ\text{C}$  i przy względnej wilgotności  $35 \pm 5\%$ .

### **3.5.15. Analiza termograwimetryczna**

Do przeprowadzenia analizy termograwimetrycznej folii pektynowych polegającej na ocenie stabilności termicznej oraz degradacji próbek zastosowano analizator termiczny (Mettler Toledo, Warszawa, Polska). Próbki (5 mg) zostały ogrzane z szybkością  $5^\circ\text{C}/\text{min}$ . przy zmianie temperatury z 30 na  $600^\circ\text{C}$  w atmosferze azotu. Prędkość przepływu azotu ( $\text{N}_2$ ) wynosiła 50 ml/min. Krzywe TGA i dTG zostały wyznaczone na podstawie różniczkowych wartości TGA. Pomiar został wykonany w dwóch powtórzeniach.

### **3.5.16. Spektroskopia w podczerwieni z transformacją Fouriera**

Przy użyciu spektrometru Cary-630 (Agilent Technologies, Cary, USA) wyznaczono grupy funkcyjne w foliach pektynowych. Do analizy widm z analizy spektroskopii w podczerwieni z transformacją Fouriera (FTIR) zastosowano metodę absorpcji w zakresie  $4000\text{-}650\text{ cm}^{-1}$  z rozdzielczością  $4\text{ cm}^{-1}$ . Widma zostały przedstawione jako średnia z 32 interferogramów.

### **3.5.17. Oznaczenie aktywności przeciwutleniającej**

Aktywność przeciwutleniającą folii pektynowych wobec rodników ABTS i DPPH oceniono w trzech powtórzeniach i wyrażono jako mg troloxu/g s.s. Absorbancję próbek folii dla ABTS zmierzono przy długości fali 734 nm, natomiast dla DPPH przy 515 nm, wykorzystując spektrometr UV-Vis EVOLUTION 220 (Thermo Electron Corporation, Waltham, MA, USA).

### **3.5.18. Kinetyka uwalniania kwasów fenolowych**

Uwalnianie wybranego kwasu fenolowego (kawowy i protokatechowy zastosowane w mieszaninach do powlekania jabłek) z folii pektynowych przeprowadzono z zastosowaniem 96% roztworu etanolu (Chempur, Piekary Śląskie, Polska) w trzech powtórzeniach. Próbki folii pektynowych o masie  $60 \pm 5\text{ mg}$  umieszczono w zlewkach, które następnie zostały napełnione etanolem do całkowitej objętości 100 ml etanolu. Następnie próbki poddano mieszaniu z prędkością 150 obr./min przy użyciu mieszadła magnetycznego RCT basic IKAMAG (IKA Polska, Warszawa, Polska). Stężenie kwasu fenolowego w próbce oznaczano metodą spektrofotometrii UV-VIS przy długości fali 310

nm dla kwasu kawowego i 315 nm dla kwasu protokatechowego, z zastosowaniem spektrofotometru Evolution 220 UV-Visible Spectrofotometr (Thermo SCIENTIFIC, Warszawa, Polska). Przez jedną godzinę pobierano 4 ml roztworu zawierającego uwolnione substancje w odstępach czasu - co minutę przez pierwsze 10 minut i co 5 minut przez pozostały czas. Po wykonaniu każdego z pomiarów zebrany roztwór umieszczano ponownie w zlewce w celu utrzymania stałej objętości. Krzywe kalibracyjne każdego związku wyznaczono w zakresie stężeń od 0,5 do 2,5 mg/100 ml 96% etanolu, otrzymując następujące równania:  $y = 0,9637x + 0,2184$  ( $R^2 = 0,944$ ) dla kwasu kawowego oraz  $y = 0,805x - 0,0511$  ( $R^2 = 0,973$ ) dla kwasu protokatechowego. Otrzymane wyniki posłużyły do obliczenia uwalniania kwasów fenolowych z zastosowaniem równania drugiego prawa Ficka w stanie przejściowym według poniższego wzoru:

$$\frac{Ct}{C_{\infty}} = 1 - \sum_{n=1}^{\infty} \frac{2\alpha(1+\alpha)}{1+\alpha+\alpha^2q_n^2} \exp\left(-\frac{Dq_n^2t}{L_0^2}\right)$$

gdzie:  $Ct$  – stężenie substancji czynnej w czasie w ośrodku uwalniania (mg/l),  $C_{\infty}$  – stężenie substancji czynnej w stanie równowagi w ośrodku uwalniania (mg/l),  $D$  – współczynnik dyfuzji substancji czynnej ( $m^2/s$ ),  $L_0$  – połowa grubości folii (m),  $q_n$  – niezerowe, dodatnie pierwiastki z  $\tan(q_n) = -\alpha q_n$  (wartość  $n$  od 1 do 6).

$\alpha$  określa się na podstawie wzoru:

$$\alpha = \frac{V_s}{K_{f,s}xV_f} (m^3)$$

gdzie:  $V_f$  – objętość folii ( $m^3$ ),  $K_{f,s} = \frac{C_{f,\infty}}{C_{s,\infty}}$  – objętość naważki i współczynnik podziału substancji czynnej między folię a ośrodek uwalniania w stanie równowagi,  $C_{f,\infty}$  i  $C_{s,\infty}$  – stężenia równowagowe substancji czynnej (mg/l) odpowiednio w folii i w ośrodku uwalniania.

## 3.6. Metody badania jablek

### 3.6.1. Powlekanie jablek

Przed procesem powlekania jabłka, charakteryzujące się podobną wielkością oraz dojrzałością, zostały umyte pod bieżącą wodą i osuszone. Następnie owoce zanurzone w roztworze powlekającym na 15 sekund i kolejno w 1% roztworze chlorku wapnia

(Avantor Performance Materials Poland S.A., Gliwice, Polska) przez 5 sekund w celu usieciowania pektyny. Próbę kontrolną stanowiły jabłka niepowlekane, które umieszczano w wodzie destylowanej na 20 sekund, a następnie osuszano. Dla każdego wariantu doświadczenia przechowalniczego przygotowano po 3 owoce o średniej masie  $150 \pm 20$  g. Po powlekanii owoce osuszono w celu usunięcia nadmiaru roztworu powlekającego, umieszczono na bibule filtracyjnej (lub plastikowych tackach w celu przeprowadzenia badań sensorycznych) i przechowywano w temperaturze pokojowej ( $22 \pm 1^\circ\text{C}$ ) i wilgotności względnej powietrza  $35 \pm 5\%$  przez 28 dni, poddając owoce badaniom po 7, 14, 21 i 28 dniach od momentu powlekania.

### 3.6.2. Zmiana masy jabłek

Ubytki masy jabłek w czasie przechowywania określono na podstawie różnicy między masą początkową i końcową przy użyciu półanalitycznej wagi laboratoryjnej Radwag S.A. (Radom, Polska) co 7 dni w 28-dniowym okresie przechowywania. Procentowy ubytek masy obliczono według wzoru:

$$U_t = \frac{m_{po} - m_t}{m_{po}} \cdot 100\%$$

gdzie:  $U_t$  – procentowy ubytek masy po  $t$  dniach (%),  $m_{po}$  – masa początkowa (dzień 0) (g),  $m_t$  – masa w dniu  $t$  (g).

### 3.6.3. Pomiar barwy jabłek

Do oznaczenia parametrów barwy owoców ( $L^*a^*b^*$ ) zastosowano kolorometr barwy Minolta CR-5 (Konica Minolta, Tokyo, Japonia). Pomiar został przeprowadzony przy użyciu standardowego obserwatora  $2^\circ$  i źródła światła D65. Oznaczenie wykonywano w dwóch przeciwległych miejscach trzech jabłek, otrzymując sześć powtórzeń dla każdego wariantu po każdym czasie przechowywania.

### 3.6.4. Pomiar zawartości ekstraktu

Zawartość ekstraktu w jabłkach wyrażoną w  $^\circ\text{Brix}$  zbadano w świeżo pozyskanym soku z jabłek w trzech powtórzeniach przy użyciu refraktometru ATAGO PAL-3 Pocket (PAL-3, Atago Instruments, Tokio, Japonia).

### 3.6.5. Pomiar kwasowości

W celu zbadania kwasowości miareczkowej jabłek pozyskano świeży sok z zastosowaniem gazy. Następnie pobierano 10 ml soku i dodawano 100 ml wody destylowanej. 25 ml tak przygotowanej próbki umieszczano w zlewce o pojemności 50 ml i umieszczano w niej elektrodę pH-metru (SHOTT Instruments, Lab 850, Warszawa, Polska). Pomiaru kwasowości dokonano poprzez miareczkowanie przygotowanych roztworów 0,25 mol/l NaOH do pH = 8,1, stanowiącego punkt zobojętnienia. Pomiar wykonywano w trzech powtórzeniach z zastosowaniem mieszania. Kwasowość miareczkową (*TTA*), wyrażoną w milimolach H<sup>+</sup> na litr produktu, obliczono według poniższego wzoru i przeliczano w celu określenia całkowitej kwasowości miareczkowej:

$$TTA = \frac{100 \cdot V_1 \cdot c}{V_0}$$

gdzie:  $V_1$  – objętość zużytego roztworu NaOH (ml),  $V_0 = 25$  ml,  $c = 0,25$  mol/l NaOH, czyli  $C_{H^+} = V_1 \cdot 10$ .

### 3.6.6. Pomiar pH

pH jabłek zmierzono w świeżym soku w trzech powtórzeniach przy użyciu pH-metru (SHOTT Instruments, Lab 850, Warszawa, Polska).

### 3.6.7. Pomiar jędrności

Pomiar jędrności jabłek dokonano z zastosowaniem teksturometru TA-TX2i (Stable Micro Systems Ltd, Haslemere, Wielka Brytania), wyposażonego w oprogramowanie Texture Expert (wersja 2.3). Przed pomiarem boki owoców były odcinane na głębokość ok 0,5 cm. Test penetracji został wykonany 9-milimetrowym trzpieniem przesuwającym się z prędkością 2 mm/s. Pomiar wykonywano w sześciu powtórzeniach – dwa nakłucia po dwóch przeciwległych stronach trzech jabłek. Miarą jędrności była maksymalna siła wyrażona w niutonach (N) i otrzymana z zależności siły od czasu penetracji.

### 3.6.8. Pomiar aktywności oddechowej jabłek

Badanie zawartości wydzielanego przez owoce etylenu (C<sub>2</sub>H<sub>4</sub>) i ditlenku węgla (CO<sub>2</sub>) wykonano w dwóch powtórzeniach przy użyciu analizatora gazów F-950 (Felix Instruments Inc., Camas, WA, USA). Trzy jabłka umieszczano na 1 h w szczelnie

zamkniętym słoju o pojemności 2 litrów, bezpośrednio połączonym z analizatorem gazów, umożliwiającym pobieranie 34 µl przestrzeni gazowej do pomiaru oddychania.

### 3.6.9. Analiza sensoryczna

Analiza sensoryczna jabłek została przeprowadzona po 7 i 28 dniach przechowywania. Zastosowano 5-stopniową skalę ocen, w której oceniono cechy jakościowe owoców, takie jak barwa, smak, aromat, jędrność i ogólna akceptowalność (**Tabela 2**). Oceny dokonano w grupie 40 osób, którą stanowili studenci oraz pracownicy Instytutu Nauk o Żywności, Szkoły Głównej Gospodarstwa Wiejskiego w Warszawie, w przedziale wiekowym wynoszącym od 20 do 45 lat. Przed przystąpieniem do oceny jabłka zostały umyte pod bieżącą wodą, a następnie pokrojone na kawałki i odpowiednio zakodowane.

**Tabela 2.** Definicje i określenia brzegowe wyróżników sensorycznych

Wyróżnik	Określenie wyróżnika	Określenia brzegowe
Barwa	Barwa jabłek (wybarwienie), występowanie rumieńca	5 pkt. – pożądana, równomierna 0 pkt. – niepożądana, nierównomiernie
Smak	Smak odczuwany po rozgryzieniu i przeżuciu	5 pkt. – charakterystyczny dla jabłek, 0 pkt. – niewyczuwalny, obcy
Zapach	Intensywność wyczuwanego zapachu	5 pkt. – charakterystyczny dla jabłek 0 pkt. – niewyczuwalny, obcy
Tekstura	Jędrność	5 pkt. – pożądana, jędrna 0 pkt. – niepożądana, niejędrna
Pożądalność	Ogólna ocena jakości	5 pkt. – bardzo pożądaną 0 pkt. – nieakceptowalne

### 3.7. Analiza statystyczna

Uzyskane wyniki badań przedstawiono w postaci średnich  $\pm$  odchylenie standardowe. Wszystkie analizy przeprowadzono przynajmniej w dwóch powtórzeniach. Otrzymane wyniki poddano analizie statystycznej przy użyciu programu Statistica 13.3 (StatSoft Polska Sp. z o.o., Kraków, Polska) z zastosowaniem testu Tukey'a oraz jednoczynnikowej analizy wariancji ANOVA przy poziomie istotności wynoszącym  $p=0,05$ .

## 4. OMÓWIENIE I DYSKUSJA WYNIKÓW

### 4.1. Wpływ kwasów fenolowych na właściwości strukturalne, sorpcyjne i zwilżalność folii pektynowych

Optymalny dobór składników do przygotowania roztworów folio/powłokotwórczych, a tym samym materiałów ochronnych, jest procesem złożonym, wymagającym uwzględnienia wielu czynników, takich jak wybór surowców, ich proporcje i właściwości, aby uzyskane materiały spełniały przewidziane funkcje. **Hipoteza 1** weryfikowana w pracy zakładała, że przygotowanie pektynowych roztworów foliotwórczych z dodatkiem wybranych kwasów fenolowych umożliwi wytworzenie aktywnego materiału ochronnego w postaci folii i powłoki o pożądanych właściwościach użytkowych wpisując się tym samym w założenia gospodarki w obiegu zamkniętym i spełniając założenia idei zrównoważonego rozwoju.

W artykułach [P2] i [P3] dokonano analizy oraz porównania właściwości fizykochemicznych wytworzonych folii pektynowych, zarówno niemodyfikowanych, jak i wzbogaconych o składniki aktywne w postaci związków fenolowych, na przykładzie kwasu wanilinowego oraz syryngowego. Badania potwierdziły możliwość włączenia obu związków wraz z pektyną jabłkową do składu roztworu tworzącego matrycę strukturotwórczą badanych materiałów. Pomimo że kwas wanilinowy oraz syryngowy pozwoliły na uzyskanie folii, to zbyt matowa oraz porowata struktura otrzymanych materiałów spowodowała rezygnację z tych wariantów w dalszych badaniach. W związku z powyższym, w kolejnych etapach badań scharakteryzowano właściwości folii jadalnych z dodatkiem sześciu innych kwasów fenolowych, spośród których wytypowano dwa kwasy przeznaczone do dalszych zastosowań. Wybrane warianty charakteryzowały się najkorzystniejszymi właściwościami powłokotwórczymi oraz użytkowymi w kontekście ich wykorzystania jako materiałów ochronnych do powlekania świeżych jabłek przeznaczonych do badań przechowalniczych [P4]. Uwzględniane w dalszych badaniach folie o zoptymalizowanych składach, cechowała gładka i jednolita struktura, barwa od lekko żółtej do delikatnie brązowej oraz transparentność (**P2-Rysunek 1**). Ponadto, dodatek plastyfikatora, jakim był glicerol, wpłynął pozytywnie na wytworzone folie poprzez zwiększenie ich plastyczności.

**Hipoteza 1** postawiona w pracy zakładała, że wprowadzenie do matrycy jadalnych folii aktywnych związków w postaci dodatku kwasów fenolowych spowoduje utworzenie

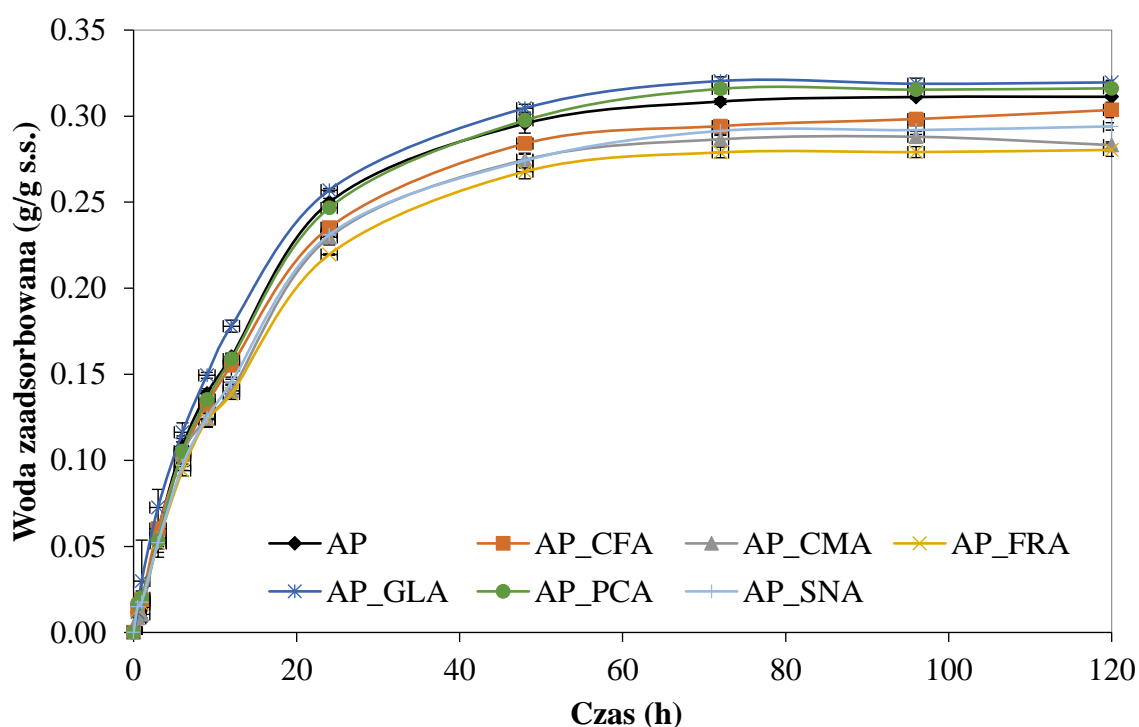
wzmocnionej oraz trwałej struktury. Analiza mikrostruktury folii jadalnych metodą skaningowej mikroskopii elektronowej (**Tabela 3**) wykazała, że zarówno folie kontrolne, jak i modyfikowane kwasami fenolowymi charakteryzowały się gładką, jednorodną i zwartą strukturą. Obserwacje przeprowadzone z wykorzystaniem skaningowej mikroskopii elektronowej wykazały brak pęknięć zarówno w przekrojach folii, jak i na ich powierzchni. Analiza struktury wskazuje na wysoką integralność materiałów oraz dobrą kompatybilność pektyny z zastosowanymi kwasami fenolowymi. Ponadto dodatek związków fenolowych przyczyniał się do zmniejszenia chropowatości powierzchni folii, potwierdzając ich istotny wpływ na proces formowania oraz organizację struktury matrycy foliotwórczej.

Ocena właściwości strukturalnych otrzymanych folii pektynowych obejmowała m.in. analizę zawartości wody, stopnia pęcznienia oraz ocenę wytworzonej struktury matrycy pektynowej wykonaną za pomocą obrazowania skaningową mikroskopią elektronową (SEM). Analiza mikrostruktury wykazała, że dodatek kwasów fenolowych wpłynął znacząco na strukturę otrzymanych kompozytów, sprzyjając powstawaniu struktur bardziej zwartych i jednorodnych (**P2-Rysunek 4**), a także przyczyniał się do zmniejszenia zawartości wody w foliach pektynowych. Zaobserwowane różnice grubości materiałów mogły wynikać ze zwiększonej zawartości frakcji stałej oraz zaburzeń pierwotnej struktury folii. Na wartość parametru grubości (**P2-Tabela 1**) wpływała również dynamika dyspersji folii, zachodząca podczas etapu jej przygotowywania oraz suszenia (Yerramathi i wsp. 2021). Dodatkowo wykazano, że dodatek kwasu synapinowego sprzyjał uzyskaniu najmniejszej zawartości wody w foliach, co można wiązać z modyfikacją struktury matrycy folii oraz tworzeniem dodatkowych wiązań, które prawdopodobnie przyczyniły się do zwiększenia szybkości parowania wody. Największą zawartość wody odnotowano w folii kontrolnej, niezawierającej dodatków aktywnych, a wprowadzenie kwasów fenolowych do matrycy pektynowej skutkowało zmniejszeniem zawartości wody w wytworzonych foliach. Zaobserwowane różnice można tłumaczyć odmienną kompatybilnością poszczególnych kwasów fenolowych z biopolimerem, stopniem ich dyspersji w strukturze folii oraz zróżnicowaną kinetyką rozpuszczania. Istotny wpływ na kształtowanie zawartości wody miały również warunki formowania struktury folii w trakcie przechowywania, w szczególności przebieg procesu odparowania wody, a także grubość otrzymanych materiałów.

Folie kontrolne oraz folie z dodatkiem kwasu galusowego, kumarowego i synapinowego wykazywały skłonność do rozpuszczania i utraty integralności

w środowisku wodnym (**P2-Tabela 1**), co wiąże się z obecnością hydroksylowych i nieestryfikowanych karboksylowych grup funkcyjnych pektyny (Athanasopoulou i wsp. 2024). Z kolei dodatek kwasu ferulowego, kawowego oraz protokatechowego zwiększał stabilność folii w wodzie, co może być związane z tworzeniem struktur o ograniczonej rozpuszczalności.

Analiza kinetyki adsorpcji pary wodnej (**Rysunek 4, P2-Rysunek 2**) wykazała, że wszystkie badane warianty folii osiągnęły stan równowagi w ciągu 24 godzin. Krzywe kinetyki adsorpcji pary wodnej charakteryzowały się zbliżonym przebiegiem, a w fazie początkowej nie stwierdzono istotnych różnic zawartości wody, co wskazuje na porównywalną siłę napędową procesu adsorpcji. Największą intensywność adsorpcji obserwowano w ciągu pierwszych 10 godzin. Zgodnie z doniesieniami Cheng i wsp. (2015), dodatek kwasów fenolowych do matrycy folii może prowadzić do zmniejszenia przepuszczalności pary wodnej, szczególnie przy niskich ich stężeniach, co wiąże się z obecnością hydrofobowych grup, ograniczających dyfuzję wody przez matrycę polimerową.



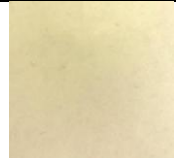
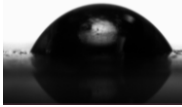

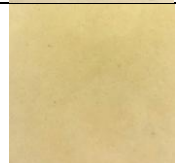

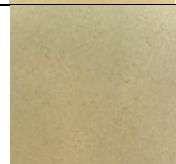
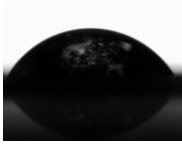

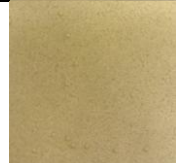

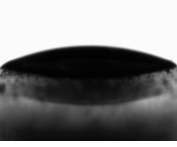
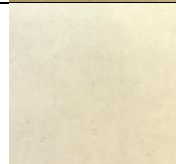



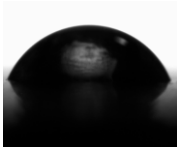
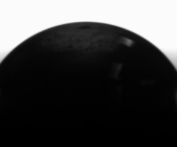
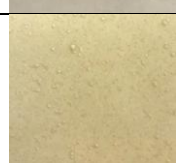
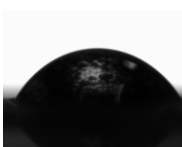
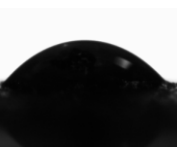
**Rysunek 4.** Woda zaadsorbowana przez folie z pektyny jabłkowej (AP) otrzymane z dodatkiem wybranych kwasów fenolowych: CFA – kwas kawowy, CMA – kwas kumarowy, FRA – kwas ferulowy, GLA – kwas galusowy, PCA – kwas protokatechowy, SNA – kwas synapinowy

W niniejszych badaniach zaobserwowano, że dodatek kwasów fenolowych do roztworów foliotwórczych wpływał jednak na wzrost współczynnika dyfuzji pary wodnej (**P2-Tabela 2**). Zjawisko to można tłumaczyć zależnością współczynnika dyfuzji od polarności zarówno matrycy polimerowej, jak i substratu, a także od oddziaływań molekularnych pomiędzy substancją czynną, matrycą i środowiskiem (Ordoñez i wsp. 2022). Na obserwowane różnice mogły również wpływać masa cząsteczkowa i struktura związków fenolowych, mikrostruktura folii oraz siła ich wiązania w matrycy, a także rozpuszczalność związków i modyfikacja kohezji łańcuchów polimerowych.

Izotermy adsorpcji wody wykazały istotną zależność pomiędzy zawartością wilgoci a aktywnością wody w badanych materiałach (**P2-Rysunek 3**). W przypadku hydrofilowych folii biopolimerowych parametr ten ma kluczowe znaczenie, gdyż odzwierciedla wrażliwość materiału na obecność wody (Veras i wsp. 2025). Izotermy adsorpcji wody folii pektynowych z dodatkiem kwasów fenolowych charakteryzowały się mniejszą wilgotnością przy niskiej aktywności wody oraz jej wzrostem wraz ze zwiększeniem aktywności wody.

Dodatek kwasów fenolowych wpłynął także na wartości kąta zwilżania wody (**Tabela 3, P2-Tabela 3**). Początkowe wartości wahały się od  $47,00^\circ \pm 4,47$  do  $58,44^\circ \pm 5,62$ . Po upływie 60 s najmniej hydrofilowy charakter powierzchni odnotowano w przypadku folii z dodatkiem kwasu kawowego oraz protokatechowego. Efekt ten można wiązać z modyfikacją oddziaływań międzycząsteczkowych na granicy faz oraz występowaniem lekkiej chropowatości folii, jak również ze zwiększonej hydrofobowości warstwy spowodowanej wzmocnieniem struktury i występowaniem oddziaływań międzycząsteczkowych między matrycą a kwasem kawowym. W przypadku materiałów opakowaniowych odporność na adsorpcję wody jest bardzo ważną właściwością użytkową. Hydrofilowość folii jadalnych można określić na podstawie kąta zwilżania wody folii, który dostarcza również informacji o interakcjach zachodzących na granicy faz. Materiały o wartościach kąta zwilżania wody mniejszych niż  $90^\circ$  są hydrofilowe, natomiast powierzchnie o kącie zwilżania wodą większym niż  $90^\circ$  charakteryzują się hydrofobowością i całkowitym lub częściowym zwilżaniem (Giridhar i wsp. 2017). W związku z czym wszystkie badane folie pektynowe charakteryzowały się hydrofilowością powierzchni, jednak zaobserwowano, że pochłanianie wody w czasie przebiegało z różną intensywnością.

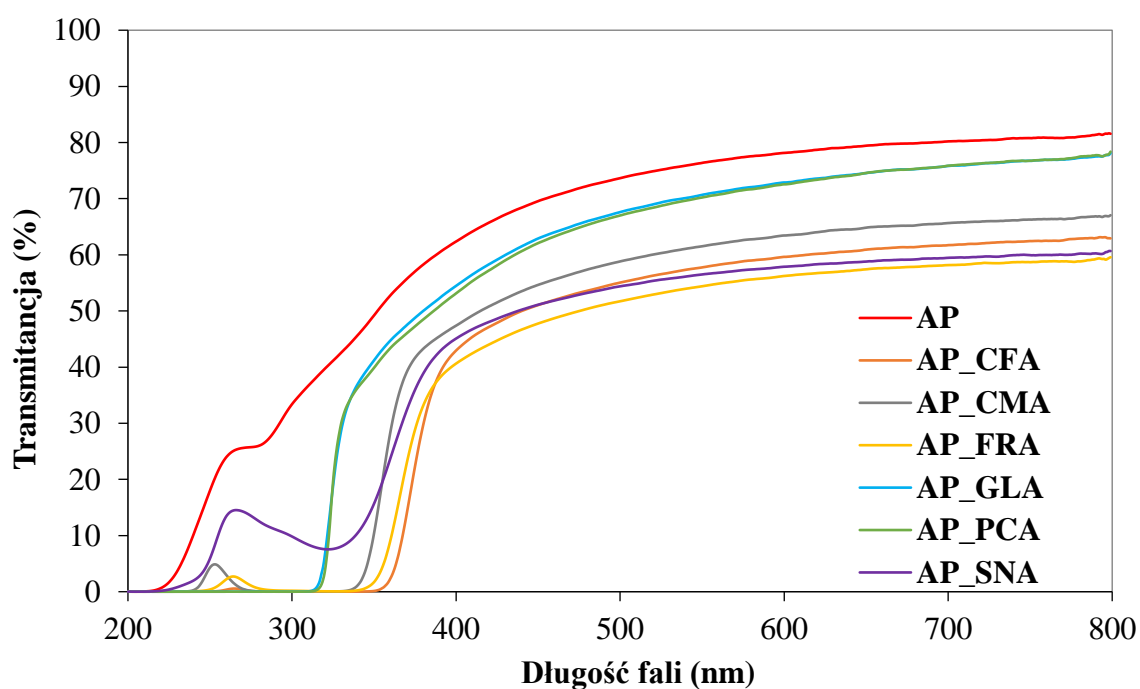
**Tabela 3.** Zdjęcia i wartości kąta zwilżania wody na początku pomiaru (0 s) i po 60 sekundach folii z pektyny jabłkowej (AP) z dodatkiem wybranych kwasów fenolowych: CFA – kwas kawowy, CMA – kwas kumarowy, FRA – kwas ferulowy, GLA – kwas galusowy, PCA – kwas protokatechowy, SNA – kwas synapinowy

Rodzaj folii		$\theta$ (°)			
		Czas (s)			
		0		60	
AP		$58,44 \pm 5,62^b$		$25,71 \pm 1,98^a$	
AP_CFA		$58,40 \pm 4,47^a$		$60,92 \pm 4,93^c$	
AP_CMA		$52,29 \pm 2,38^{ab}$		$26,88 \pm 2,12^a$	
AP_FRA		$47,00 \pm 4,47^a$		$25,03 \pm 9,23^a$	
AP_GLA		$47,61 \pm 4,02^a$		$24,81 \pm 3,82^a$	
AP_PCA		$56,51 \pm 5,91^b$		$56,42 \pm 4,27^{bc}$	
AP_SNA		$55,36 \pm 2,89^b$		$43,11 \pm 3,95^b$	

W tabeli przedstawiono średnie wartości z odchyleniem standardowym. Różne litery a-c przy wartościach w kolumnach oznaczają różnice statystyczne pomiędzy wynikami otrzymanymi dla poszczególnych wariantów.

## 4.2. Wpływ kwasów fenolowych na wybrane właściwości funkcjonalne folii pektynowych

Analiza parametrów optycznych wykazała, że dodatek kwasów fenolowych istotnie modyfikował właściwości barierowe badanych materiałów wobec światła, a także wpływał na zmianę barwy, połysk oraz stopień nieprzezroczystości. Na podstawie analizy widm transmitancji zaobserwowano, że folia kontrolna charakteryzowała się największą wartością transmitancji w całym analizowanym zakresie długości fal od 200 do 800 nm, co wskazuje na najniższą zdolność do absorpcji światła (**Rysunek 5, P3-Rysunek 1**). Natomiast we wszystkich wariantach folii zawierających kwasy fenolowe zaobserwowano wyraźne obniżenie transmitancji. Najmniejsze wartości tego parametru odnotowano w przypadku folii z dodatkiem kwasu ferulowego, co świadczy o jej największej skuteczności w ograniczaniu przenikania światła.



**Rysunek 5.** Widma światła UV-VIS folii z pektyny jabłkowej (AP) otrzymanych z dodatkiem wybranych kwasów fenolowych: CFA – kwas kawowy, CMA – kwas kumarowy, FRA – kwas ferulowy, GLA – kwas galusowy, PCA – kwas protokatechowy, SNA – kwas synapinowy

Obecność kwasów fenolowych: kawowego, kumarowego, ferulowego, galusowego oraz protokatechowego w strukturze folii pektynowych skutkowałą niską transmitancją w zakresie promieniowania UV (250–300 nm), przy jednoczesnym wzroście transmitancji

w zakresie światła widzialnego (350–800 nm) (**P3-Rysunek 1**). Uzyskane wyniki jednoznacznie wskazują, że dodatek tych związków zwiększał barierowość wobec światła, szczególnie w odniesieniu do promieniowania ultrafioletowego. Gwałtowny wzrost transmitancji obserwowany w zakresie 310–370 nm mógł być związany z charakterystycznymi pasmami absorpcji kwasów fenolowych. Materiały wykazujące niską transmitancję w zakresie UV-VIS mogą stanowić skuteczną ochronę dla produktów wrażliwych na działanie światła. Na uwagę zasługuje fakt, iż kwas ferulowy wykazywał silniejszy wpływ na redukcję transmisji promieniowania UV niż światła widzialnego. W konsekwencji folie zawierające związki bioaktywne mogą ograniczać powstawanie wolnych rodników w pakowanej żywności, zapewniając ochronę przed procesami utleniania indukowanymi światłem oraz opóźniając degradację składników odżywczych.

Dodatek kwasów fenolowych wpłynął na zmniejszenie jasności folii pektynowych (parametr  $L^*$ ) (**P3-Tabela 1**). Wszystkie badane folie wykazały tendencję w kierunku barwy zielonej (ujemne wartości parametru  $a^*$ ), jedynie folia pektynowa z dodatkiem kwasu ferulowego, charakteryzująca się dodatnią wartością parametru  $a^*$ , wykazywała tendencję do przesunięcia barwy w kierunku czerwieni. Jednakże wartości były stosunkowo niskie (od -1,49 do 0,77). Wszystkie badane folie pektynowe charakteryzowały się tendencją do żółknięcia (parametr  $b^*$  w zakresie od 17,43 do 26,22). Spośród analizowanych próbek folia z dodatkiem kwasu ferulowego wyróżniała się największą zmianą barwy, co przekładało się na jej najbardziej wyrazisty efekt wizualny.

Analiza właściwości barierowych wykazała istotne różnice w przepuszczalności pary wodnej (**Tabela 4, P2-Tabela 4**), tlenu i ditlenku węgla (**Tabela 4, P3-Tabela 3**) w zależności od rodzaju folii. Najmniejsze wartości współczynnika przepuszczalności pary wodnej odnotowano w przypadku folii kontrolnej, natomiast największe, gdy folię wzbogacono kwasem synapinowym. Uzyskane wyniki wskazują, że modyfikacje struktury kompozytowej folii istotnie determinują ich właściwości barierowe, co ma kluczowe znaczenie dla kształtowania właściwości użytkowych biopolimerowych materiałów opakowaniowych. Duża wartość wskaźnika przepuszczalności pary wodnej stanowi istotny czynnik sprzyjający utracie wilgoci przez owoce i warzywa po zbiorach, prowadząc do obniżenia ich jakości oraz skrócenia okresu ich przydatności do spożycia. Folie pektynowe z dodatkiem kwasów fenolowych, co wynika z przeprowadzonych badań, charakteryzowały się większą przepuszczalnością pary wodnej w porównaniu z folią kontrolną. Wzrost ten mógł wynikać z modyfikacji struktury matrycy wytworzonej folii, w której obecność kwasów fenolowych ułatwiała migrację pary wodnej.

**Tabela 4.** Przepuszczalność pary wodnej (*WVP*), tlenu (*O<sub>2</sub>P*) i ditlenku węgla (*CO<sub>2</sub>P*) folii z pektyny jabłkowej (*AP*) otrzymanych z dodatkiem wybranych kwasów fenolowych: *CFA* – kwas kawowy, *CMA* – kwas kumarowy, *FRA* – kwas ferulowy, *GLA* – kwas galusowy, *PCA* – kwas protokatechowy, *SNA* – kwas synapinowy

Folia	<i>WVP</i> ( $\times 10^{-10}$ g/m·s·Pa)	<i>O<sub>2</sub>P</i> ( $\times 10^{-16}$ g/m·s·Pa)	<i>CO<sub>2</sub>P</i> ( $\times 10^{-16}$ g/m·s·Pa)
AP	7,16 ± 0,42 <sup>a</sup>	0,84 ± 0,02 <sup>a</sup>	1,15 ± 0,07 <sup>a</sup>
AP_CFA	9,76 ± 0,06 <sup>b</sup>	0,46 ± 0,03 <sup>a</sup>	0,70 ± 0,01 <sup>a</sup>
AP_CMA	10,68 ± 0,12 <sup>c</sup>	0,89 ± 0,06 <sup>a</sup>	1,33 ± 0,09 <sup>a</sup>
AP_FRA	10,46 ± 0,16 <sup>c</sup>	0,92 ± 0,12 <sup>a</sup>	1,26 ± 0,17 <sup>b</sup>
AP_GLA	7,45 ± 0,17 <sup>a</sup>	0,88 ± 0,00 <sup>a</sup>	1,46 ± 0,09 <sup>a</sup>
AP_PCA	7,46 ± 0,22 <sup>a</sup>	0,45 ± 0,08 <sup>b</sup>	0,60 ± 0,09 <sup>b</sup>
AP_SNA	10,48 ± 0,22 <sup>c</sup>	1,65 ± 0,10 <sup>c</sup>	2,33 ± 0,12 <sup>c</sup>

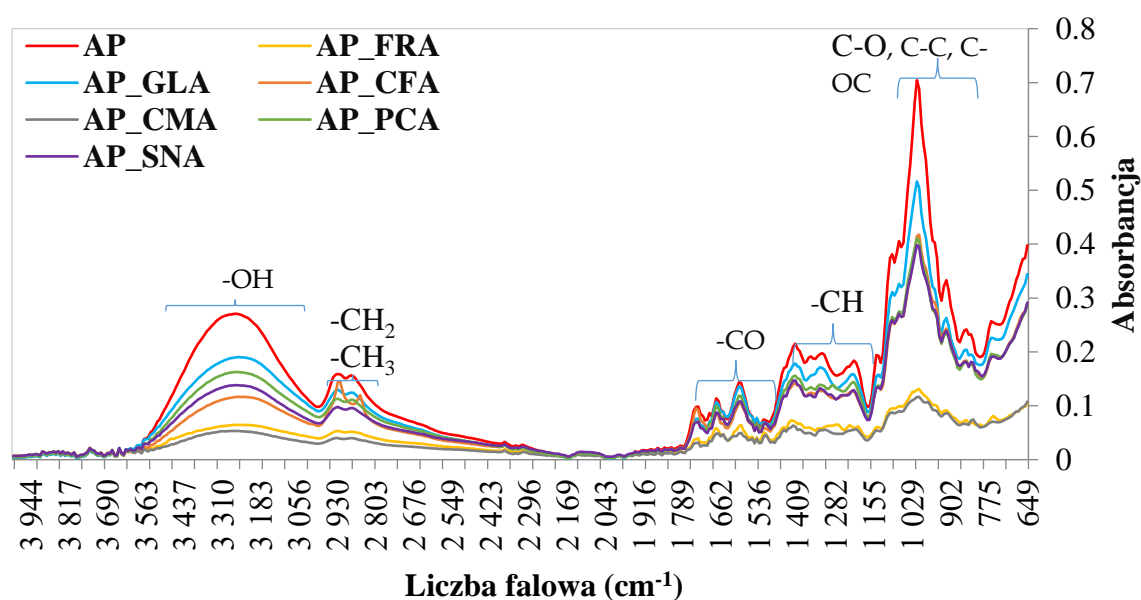
W tabeli przedstawiono średnie wartości z odchyleniem standardowym. Różne litery a-c przy wartościach w kolumnach oznaczają różnice statystyczne pomiędzy wynikami otrzymanymi dla poszczególnych wariantów.

Największą przepuszczalność tlenu oraz ditlenku węgla spośród folii pektynowych z dodatkiem kwasów fenolowych odnotowano w przypadku wariantu zawierającego pektynę jabłkową i kwas synapinowy (**Tabela 4, P3-Tabela 3**). Zwiększona przepuszczalność obu gazów może wynikać z zaburzenia struktury polimerowej folii na skutek inkorporacji kwasu synapinowego. Przepuszczalność gazów przez folie jadalne jest determinowana przez szereg czynników, w tym integralność strukturalną materiału, równowagę pomiędzy obszarami hydrofobowymi i hydrofilowymi oraz interakcje pomiędzy komponentami polimerowymi (Kocira i wsp. 2021). Uzyskane wyniki wskazują, że zarówno obecność kwasów fenolowych, jak i ich rodzaj odgrywają kluczową rolę w modyfikacji właściwości barierowych folii pektynowych.

Folie charakteryzujące się podwyższoną stabilnością termiczną mogą skuteczniej chronić pakowaną żywność, zachowując jej integralność strukturalną i właściwości funkcjonalne. Folie pektynowe wykazują relatywnie niską stabilność termiczną, co jest związane z efektem plastyfikującym wynikającym z hydratacji matrycy polimerowej. Dodatek bipolimerów lub związków o wysokiej stabilności termicznej może istotnie modyfikować parametry termiczne materiałów na bazie pektyny (Huang i wsp. 2021). Jednakże zaobserwowano degradację termiczną folii kontrolnych przy wyższych

temperaturach z zakresu 30-100°C i niższych z zakresu 100-280°C i 280-600°C w porównaniu z foliami zawierającymi kwasy fenolowe (**P3-Tabela 4**), wskazując, że w wyższych zakresach temperatur folie zawierające kwasy fenolowe były bardziej stabilne termicznie. Wyjątek stanowiła folia zawierająca kwas synapinowy, której degradacja następowała w niższej temperaturze z zakresu 280-600°C w porównaniu pozostałymi foliami. Zaobserwowano również mniejszą utratę masy folii kontrolnych w porównaniu do tych zawierających kwasy fenolowe, poza zakresem 280-600°C. Biorąc pod uwagę analizę termogravimetryczną wszystkich badanych próbek, największe zmniejszenie masy odnotowano w drugim etapie ogrzewania (**P3-Tabela 4**, **P3-Rysunek 2**), co można przypisać rozkładowi termicznemu polimerów o niskiej masie cząsteczkowej obecnych w matrycy folii, a także odparowaniu glicerolu i degradacji łańcuchów polisacharydowych (Cao i Song 2019).

Tworzenie wiązań wodorowych pomiędzy grupami O–H a innymi grupami funkcyjnymi obniża energię drgań rozciągających wiązania O–H, powodując przesunięcie pasm absorpcyjnych ku mniejszym częstotliwościom (**Rysunek 6**, **P3-Rysunek 3**). Zjawisko to, w połączeniu ze zmniejszeniem liczby cząsteczek wody związanych z pektynami, wskazuje na niższy stopień uwodnienia folii zawierających kwasy fenolowe w porównaniu z próbą kontrolną (Serrafi i wsp. 2025)



**Rysunek 6.** Widma w podczerwieni z transformacją Fouriera folii z pektyny jabłkowej (AP) otrzymanych z dodatkiem wybranych kwasów fenolowych: CFA – kwas kawowy, CMA – kwas kumarowy, FRA – kwas ferulowy, GLA – kwas galusowy, PCA – kwas protokatechowy, SNA – kwas synapinowy

Właściwości mechaniczne folii oceniono na podstawie wytrzymałości na rozciąganie, wydłużenia przy zerwaniu oraz modułu Younga. Dodatek kwasów fenolowych prowadził do wzrostu wytrzymałości na rozciąganie, przy czym największe wartości odnotowano w przypadku folii zawierających kwasy kawowy, protokatechowy oraz synapinowy (**P3-Tabela 5**). Efekt plastyfikujący był szczególnie widoczny w przypadku kwasów kawowego, ferulowego i protokatechowego, co przejawiało się istotnym zwiększeniem wydłużenia przy zerwaniu. Jednocześnie wartości modułu Younga wszystkich folii modyfikowanych kwasami fenolowymi były istotnie większe w porównaniu z próbkami kontrolnymi.

Wprowadzenie kwasów fenolowych do struktury pektyny umożliwia otrzymanie nowych matryc polimerowych o rozszerzonych właściwościach funkcjonalnych, w tym zadowalającej aktywności przeciwutleniającej. Zdolność przeciwutleniającą folii oceniano z wykorzystaniem powszechnie stosowanych metod, takich jak test DPPH oraz test ABTS (**Tabela 5, P3-Tabela 6**). Uzyskane wyniki jednoznacznie potwierdzają, że wszystkie zastosowane kwasy fenolowe przyczyniały się do zwiększenia aktywności przeciwutleniającej folii pektynowych. Zaobserwowane różnice pomiędzy poszczególnymi wariantami można wiązać z odmienną budową chemiczną kwasów fenolowych oraz stopniem ich oddziaływań i kompatybilności z matrycą pektynową w trakcie formowania folii.

**Tabela 5.** Aktywność przeciwutleniająca folii z pektyny jabłkowej (AP) otrzymanych z dodatkiem wybranych kwasów fenolowych: CFA – kwas kawowy, CMA – kwas kumarowy, FRA – kwas ferulowy, GLA – kwas galusowy, PCA – kwas protokatechowy, SNA – kwas synapinowy

Folia	ABTS (mg TE/g s.s.)	DPPH (mg TE/g s.s.)
AP	0,36 ± 0,12 <sup>a</sup>	0,83 ± 0,02 <sup>a</sup>
AP_CFA	10,13 ± 0,45 <sup>b</sup>	12,65 ± 0,36 <sup>a</sup>
AP_CMA	18,60 ± 0,91 <sup>c</sup>	62,66 ± 3,51 <sup>c</sup>
AP_FRA	38,08 ± 0,69 <sup>f</sup>	131,48 ± 7,57 <sup>e</sup>
AP_GLA	17,38 ± 0,23 <sup>c</sup>	39,84 ± 0,84 <sup>b</sup>
AP_PCA	23,13 ± 0,23 <sup>d</sup>	57,99 ± 0,04 <sup>c</sup>
AP_SNA	32,64 ± 0,42 <sup>e</sup>	93,47 ± 5,69 <sup>d</sup>

W tabeli przedstawiono średnie wartości z odchyleniem standardowym. Różne litery a-f przy wartościach w kolumnach oznaczają różnice statystyczne pomiędzy wynikami otrzymanymi dla poszczególnych wariantów.

### 4.3. Wpływ wybranych kwasów fenolowych na właściwości reologiczne pektynowych roztworów powłokotwórczych

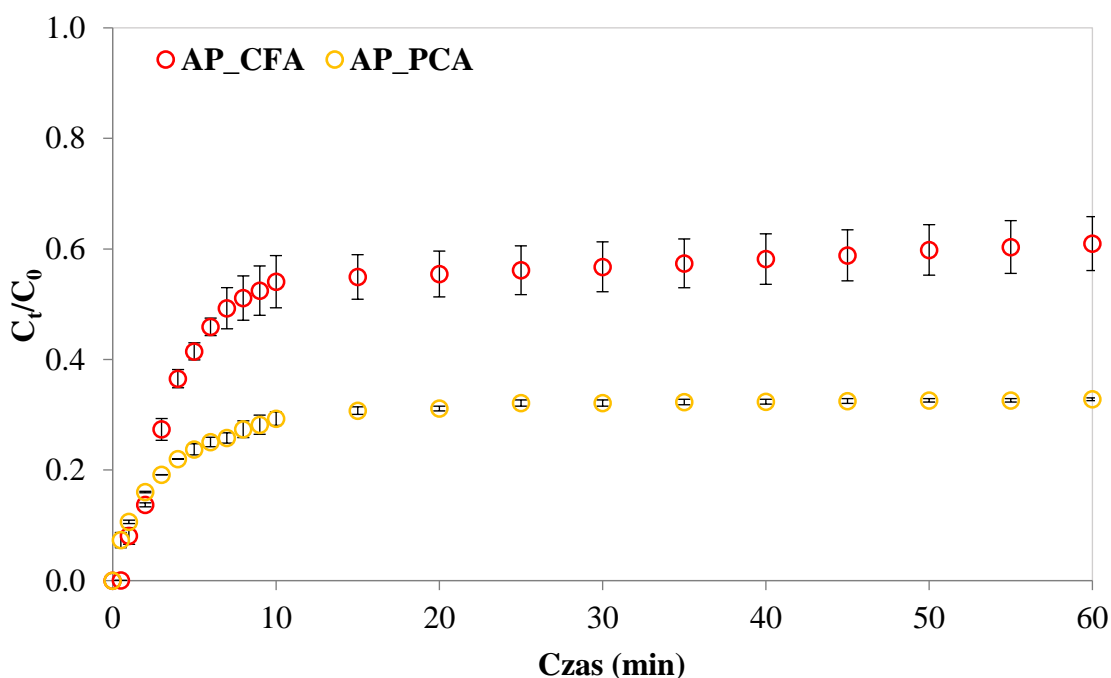
Analiza właściwości reologicznych roztworów powłokotwórczych stanowi istotne narzędzie do oceny parametrów determinujących ich przydatność aplikacyjną, takich jak zdolność do rozprowadzania, jednorodność warstwy, grubość powłoki, właściwości mechaniczne oraz mikrostruktura uzyskiwanych folii (Rashid i wsp. 2020; Janowicz i wsp. 2023). Krzywe płynięcia i lepkości roztworów na bazie pektyny jabłkowej oraz ich mieszanin z wybranymi kwasami fenolowymi (kawowym i protokatechowym) pokazały wyraźne różnice w zachowaniu reologicznym badanych układów (**P4-Rysunek 2**). Niezależnie od składu, wszystkie próbki wykazywały charakter nienewtonowski z wyraźnym efektem rozrzedzania przy ścinaniu, co pozostaje w zgodzie z doniesieniami literaturowymi dotyczącymi układów pektynowych (Zhai i wsp. 2021).

Dodatek kwasu protokatechowego wpłynął na zmniejszenie lepkości pozornej przy wszystkich poziomach zastosowanego naprężenia ścinającego. Efekt ten mógł być związany ze zmianami w strukturze cząsteczkowej pektyny, a w szczególności ze sposobem, w jaki kwas protokatechowy wpływa na stopień dysocjacji grup karboksylowych i potencjalnie zakłóca tworzenie sieci między łańcuchami polisacharydowymi. Obserwacje te są zbliżone z wynikami przedstawionymi przez Karaki i wsp. (2016), którzy wykazali, że dodatek kwasu ferulowego zmniejsza gęstość upakowania cząsteczkowego i oddziaływanie międzycząsteczkowego w układach pektynowych, co skutkuje zmniejszeniem lepkości. W przypadku kwasu kawowego lepkość pozostała niezmienną w porównaniu z próbką kontrolną. Można to przypisać słabszym oddziaływaniom tego kwasu z łańcuchami pektynowymi i mniejszej zdolności do tworzenia wiązań wodorowych.

Różnice w zachowaniu reologicznym można również interpretować w kategoriach zmian hydrofobowości cząsteczek pektyny po dodaniu związków fenolowych. Według Zhang i wsp. (2013), modyfikacja karboksylowych grup bocznych i zawartości ramnozy ma kluczowe znaczenie dla właściwości płynięcia pektyny jabłkowej. Obecność kwasu protokatechowego może zaburzać strukturę i długość agregatów pektyny, zmieniając tym samym lepkość. Zaobserwowane różnice właściwości reologicznych mogły wynikać m.in. ze zmian charakteru hydrofobowości makrocząsteczek pektyny, zachodzących w wyniku inkorporacji związków fenolowych.

#### 4.4. Wpływ rodzaju kwasu fenolowego na kinetykę uwalniania z folii pektynowych

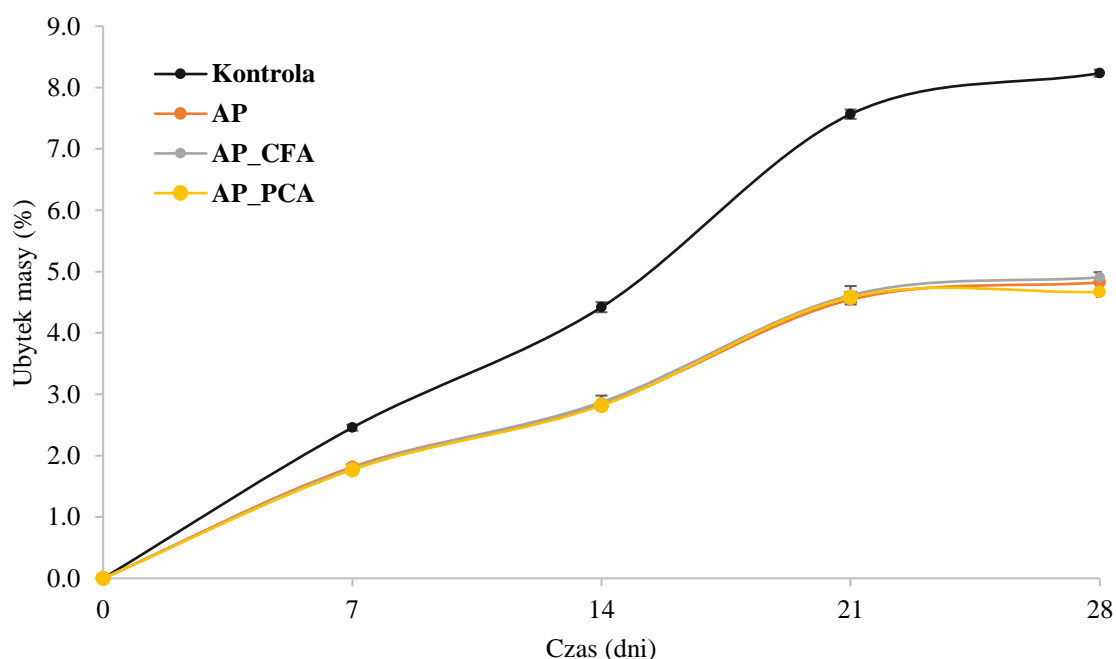
Kinetykę uwalniania kwasu kawowego i protokatechowego z folii pektynowych w środowisku 96% etanolu przedstawiono na **Rysunku 7 (P4-Rysunek 3)**. Analizowane folie wykazywały doskonałą rozpuszczalność w wodzie, w związku z tym woda destylowana lub 50% roztwór etanolu nie wykazywały odpowiedniej kinetyki uwalniania ze względu na natychmiastowe rozpuszczanie. Różne zachowania kwasów fenolowych w etanolu prawdopodobnie wynikają z różnego składu chemicznego i właściwości kwasów. Ponadto, proces uwalniania może być zależny od interakcji między składnikami, w szczególności pektyną, kwasem, glicerolem i etanolem. Ponadto wszystkie testowane warianty wykazywały współczynniki korelacji liniowej bliskie jedności, co wskazuje na silne zależności liniowe między zmiennymi.



**Rysunek 7.** Kinetyka uwalniania kwasu kawowego (CFA) i kwasu protokatechowego (PCA) z folii z pektyny jabłkowej (AP) do etanolu (96%)

















#### 4.5. Zastosowanie bioaktywnych powłok jadalnych z pektyny i kwasów fenolowych w celu poprawy cech jakościowych jabłek odmiany *Golden Delicious* podczas przechowywania

Zjawisko zmniejszania masy jabłek, które występuje podczas przechowywania ma istotny wpływ na zmiany wyglądu zewnętrznego owoców, a tym samym jest czynnikiem decydującym o ich jakości. Podczas przechowywania zaobserwowano, że w pierwszym tygodniu wystąpiło największe zmniejszenie masy (**Rysunek 8, P4-Rysunek 4**). Natomiast w kolejnych tygodniach przechowywania odnotowano większe zmiany masy w próbkach kontrolnych, które prawdopodobnie były spowodowane oddychaniem i odparowywaniem wody przez cienką skórę. Można zatem stwierdzić, że zastosowanie powłok jadalnych, zarówno otrzymanych z samej pektyny jabłkowej, jak i z dodatkiem kwasów fenolowych, zmniejszyło zmiany masy jabłek w kolejnych tygodniach przechowywania, czego nie można stwierdzić w przypadku jabłek bez powłok ochronnych. Oprócz tego stwierdzono, że czas był czynnikiem wpływającym na zmianę jasności barwy i pozwolił na dłuższe zachowanie zielonej barwy jabłek *Golden Delicious* podczas przechowywania w porównaniu do jabłek kontrolnych.



**Rysunek 8.** Ubytek masy jabłek niepowlekanych (Kontrola) i powlekanych roztworem z pektyny jabłkowej (AP) oraz z dodatkiem kwasu kawowego (CFA) i protokatechowego (PCA)

Barwa jabłek, stanowiąca jeden z kluczowych parametrów akceptacji konsumenckiej, ulegała wyraźnym zmianom w zależności od zastosowanej powłoki (**P4-Tabela 2**). Owoce powlekane charakteryzowały się bardziej zielonkawym odcieniem w porównaniu z próbkami kontrolnymi, które wykazywały tendencję do żółknięcia (**Rysunek 9, P4-Rysunek 5**). Nie stwierdzono istotnych różnic pomiędzy jabłkami pokrytymi powłoką pektynową bez i z kwasami fenolowymi, wskazując, że warstwa pektynowa pełni funkcję ochronną niezależnie od obecności związków aktywnych.

		Rodzaj powłoki			
		Kontrola	AP	AP_CFA	AP_PCA
Czas (dni)	7				
					
28					
					

**Rysunek 9.** Wygląd jabłek niepowlekanych i powlekanych roztworem powlekającym z pektyny jabłkowej (AP) z dodatkiem kwasu kawowego (CFA) i protokatechowego (PCA) po 7 i 28 dniach przechowywania

Zaobserwowano istotne zwiększenie zawartości ekstraktu w jabłkach kontrolnych w czasie przechowywania w porównaniu z jabłkami powlekаныmi (Tabela 6, P4-Tabela 3), wskazując na spowolnienie dojrzewania owoców, co było spowodowane obecnością powłok pektynowych i stanowi potwierdzenie założeń hipotezy 2.

**Tabela 6.** Zawartość ekstraktu, kwasowość miareczkowa i pH jabłek niepowlekanych (Kontrola) i powlekanych roztworem powlekającym na bazie pektyny jabłkowej (AP) z dodatkiem kwasu kawowego (CFA) i protokatechowego (PCA) w czasie przechowywania

Czas (Dni)	Typ analizy			
	Kontrola	AP	AP_CFA	AP_PCA
	<b>Ekstrakt (°Brix)</b>			
0	13,47±0,37 <sup>a,A</sup>			
7	13,77±0,11 <sup>a,A</sup>	13,50±0,10 <sup>a,A</sup>	13,48±0,23 <sup>a,A</sup>	13,53±0,40 <sup>a,A</sup>
14	14,03±0,39 <sup>a,A</sup>	13,50±0,24 <sup>a,A</sup>	13,52±0,21 <sup>a,A</sup>	13,50±0,37 <sup>a,A</sup>
21	14,35±0,19 <sup>b,B</sup>	13,53±0,07 <sup>a,A</sup>	13,52±0,18 <sup>a,A</sup>	13,55±0,36 <sup>a,A</sup>
28	14,95±0,48 <sup>b,B</sup>	13,53±0,19 <sup>a,A</sup>	13,52±0,32 <sup>a,A</sup>	13,57±0,27 <sup>a,A</sup>
	<b>Kwasowość miareczkowa (% kwasu jabłkowego)</b>			
0	1,37±0,02 <sup>b,A</sup>			
7	0,82±0,05 <sup>a,A</sup>	0,93±0,05 <sup>a,A</sup>	0,91±0,02 <sup>a,A</sup>	0,89±0,11 <sup>a,A</sup>
14	0,90±0,07 <sup>a,B</sup>	0,79±0,08 <sup>a,AB</sup>	0,77±0,03 <sup>a,A</sup>	0,77±0,09 <sup>a,A</sup>
21	0,88±0,04 <sup>a,B</sup>	0,75±0,05 <sup>a,A</sup>	0,76±0,08 <sup>a,A</sup>	0,71±0,05 <sup>a,A</sup>
28	0,80±0,05 <sup>a,A</sup>	0,79±0,05 <sup>a,A</sup>	0,76±0,05 <sup>a,A</sup>	0,73±0,02 <sup>a,A</sup>
	<b>pH</b>			
0	3,59±0,04 <sup>a,A</sup>			
7	3,84±0,02 <sup>b,A</sup>	3,84±0,01 <sup>b,A</sup>	3,83±0,03 <sup>a,A</sup>	3,79±0,05 <sup>a,A</sup>
14	3,90±0,03 <sup>b,A</sup>	3,98±0,09 <sup>b,A</sup>	3,98±0,06 <sup>b,A</sup>	4,04±0,01 <sup>b,A</sup>
21	4,00±0,02 <sup>b,B</sup>	4,09±0,06 <sup>b,A</sup>	4,12±0,08 <sup>b,A</sup>	4,16±0,01 <sup>b,A</sup>
28	4,05±0,06 <sup>b,A</sup>	4,11±0,07 <sup>b,A</sup>	4,12±0,03 <sup>b,A</sup>	4,26±0,04 <sup>b,A</sup>

W tabeli przedstawiono średnie wartości z odchyleniem standardowym. Różne litery a-b przy wartościach w kolumnach lub A-B w wierszach oznaczają różnice statystyczne pomiędzy wynikami otrzymanymi dla poszczególnych wariantów.

Największe zmniejszenie kwasowości zaobserwowano w jabłkach niepowlekanych oraz w próbkach pokrytych powłoką bez dodatku kwasów fenolowych (**Tabela 6, P4-Tabela 3**). Analiza zmian zawartości kwasu jabłkowego wskazuje, że jabłka powlekane wykazywały bardziej wyraźne zmiany tego parametru w trakcie przechowywania, co przekładało się na modyfikację profilu smakowego. Zjawisko to można wiązać z procesami dojrzewania, obejmującymi hydrolizę skrobi do cukrów prostych oraz jednoczesny spadek zawartości kwasów organicznych (Ahmad i wsp. 2021). Zwiększenie wartości wskaźnika pH owoców w trakcie przechowywania jest bezpośrednio związane z procesami oddychania, podczas których dochodzi do utleniania kwasów organicznych (Cofelice i wsp. 2019). W ciągu 28 dni przechowywania pH systematycznie wzrastało, przy czym nie stwierdzono istotnych różnic pomiędzy poszczególnymi wariantami (**Tabela 6, P4-Tabela 3**). Wartości pH mieściły się w zakresie od 3,59 do 4,26.

Głównym wyzwaniem w przechowywaniu świeżych jabłek jest zachowanie jędrności, która decyduje o pozytywnym odbiorze sensorycznym wśród konsumentów i ich wartości handlowej, a występowaniem tzw. mięknięcia owoców, które jest procesem niepożądanym, zachodzącym podczas dojrzewania jabłek (Ferreira i wsp. 2024). Jabłka charakteryzujące się znaczną twardością są bardziej odporne na uszkodzenia mechaniczne oraz lepiej nadają się do dłuższego przechowywania (Lara, Graell i Ortiz 2024). Stosowanie jadalnych powłok może wpłynąć na zahamowanie działania enzymów odpowiedzialnych za rozkład pektyny, w wyniku czego procesy metaboliczne ulegają spowolnieniu, a jabłka stają się twardsze (Rashid i wsp. 2023). Twardość owoców, silnie skorelowana z ich soczystością, chrupkością i ograniczeniem mączystości, stanowi istotny wyznacznik akceptacji konsumenckiej (Ferreira i wsp. 2024). W przypadku jabłek powlekanych szybkość obniżania twardości była wyraźnie mniejsza, przy jednoczesnym braku istotnych różnic pomiędzy poszczególnymi rodzajami powłok (**Tabela 7, P4-Tabela 4**).

Największy spadek twardości odnotowano w próbkach niepowlekanych po 14, 21 oraz 28 dniach przechowywania. Na podstawie przeprowadzonych badań można stwierdzić, że powlekanie wpłynęło na zachowanie większej jędrności jabłek w porównaniu do owoców niepowlekanych, co stanowi potwierdzenie założeń **hipotezy 2**.

**Tabela 7.** Jędrność jabłek kontrolnych niepowlekanych (Kontrola) i powlekanych roztworem powlekającym na bazie pektyny jabłkowej (AP) z dodatkiem kwasu kawowego (CFA) i protokatechowego (PCA) w czasie przechowywania

Czas (Dni)	Rodzaj powłoki			
	Kontrola	AP	AP_CFA	AP_PCA
	<b>Jędrność (N)</b>			
0	58,03±6,50 <sup>c,A</sup>			
7	50,77±2,40 <sup>b,B</sup>	49,01±1,60 <sup>b,AB</sup>	48,32±2,60 <sup>a,A</sup>	48,03±1,60 <sup>a,A</sup>
14	42,80±3,70 <sup>a,A</sup>	45,86±2,70 <sup>b,AB</sup>	47,40±2,60 <sup>a,B</sup>	46,45±2,80 <sup>a,B</sup>
21	39,73±2,50 <sup>a,A</sup>	43,94±3,70 <sup>a,B</sup>	44,70±3,30 <sup>a,B</sup>	45,33±2,90 <sup>a,B</sup>
28	36,84±1,30 <sup>a,A</sup>	43,64±3,50 <sup>a,B</sup>	44,20±4,10 <sup>a,B</sup>	44,61±4,80 <sup>a,B</sup>

W tabeli przedstawiono średnie wartości z odchyleniem standardowym. Różne litery a-c przy wartościach w kolumnach lub A-B w wierszach oznaczają różnice statystyczne pomiędzy wynikami otrzymanymi dla poszczególnych wariantów.

W celu utrzymania odpowiedniej jakości jabłek podczas przechowywania konieczne jest ograniczenie metabolizmu etylenu i oddychania owoców (Khera i wsp. 2024). Od początku okresu przechowywania jabłka niepowlekane wytwarzały znacznie więcej etylenu niż jabłka powlekane (**Tabela 8, P4-Tabela 5**). Można zatem stwierdzić, że powłoki ograniczały produkcję etylenu, a tym samym spowalniały proces dojrzewania owoców, co stanowi również potwierdzenie założeń **hipotezy 2**. Zaobserwowano również zwiększoną produkcję ditlenku węgla w pierwszym tygodniu przechowywania w próbie kontrolnej, co wskazuje na intensywną aktywność oddechową jabłek (**Tabela 8, P4-Tabela 5**). Zastosowanie powłok pektynowych spowolniło ten proces niezależnie od obecności kwasów fenolowych. Powłoka niezawierająca dodatków funkcjonalnych również wykazała istotny wpływ na ograniczenie zmian jakościowych. Jednakże, wprowadzenie kwasów fenolowych wzmocniło jej działanie. Zaobserwowana wyższa zawartość etylenu i ditlenku węgla w komorze analitycznej w przypadku jabłek kontrolnych w porównaniu z powlekanymi jednoznacznie wskazuje na ograniczenie aktywności oddechowej jabłek. Dodatkowo, różnice w obserwacjach pomiędzy wariantami powłok wskazują również na modyfikację właściwości barierowych powłok wytworzonych z dodatkiem kwasów fenolowych.

**Tabela 8.** Produkcja etylenu i ditlenku węgla przez jabłka niepowlekane (Kontrola) i powlekane roztworami powlekającymi na bazie pektyny jabłkowej (AP) z dodatkiem kwasu kawowego (CFA) i protokatechowego (PCA) w czasie przechowywania

Czas (Dni)	Rodzaj powłoki			
	Kontrola	AP	AP_CFA	AP_PCA
<b>Etylen (ppm C<sub>2</sub>H<sub>4</sub>)</b>				
0	60,40±3,00 <sup>b,A</sup>			
7	63,80±7,10 <sup>b,B</sup>	18,25±0,65 <sup>b,A</sup>	8,85±1,15 <sup>a,A</sup>	6,80±0,10 <sup>a,A</sup>
14	67,00±1,70 <sup>b,B</sup>	15,50±0,55 <sup>a,A</sup>	13,70±2,30 <sup>b,A</sup>	11,85±1,00 <sup>a,A</sup>
21	67,65±7,45 <sup>b,B</sup>	28,75±1,05 <sup>d,A</sup>	25,25±3,45 <sup>c,A</sup>	23,25±1,85 <sup>a,A</sup>
28	52,75±3,05 <sup>a,B</sup>	25,70±0,70 <sup>c,A</sup>	23,90±0,50 <sup>c,A</sup>	23,55±1,65 <sup>a,A</sup>
<b>Ditlenek węgla (mg CO<sub>2</sub>/kg/h)</b>				
0	0,23±0,13 <sup>c,A</sup>			
7	0,27±0,05 <sup>d,B</sup>	0,21±0,01 <sup>d,A</sup>	0,16±0,01 <sup>c,A</sup>	0,17±0,04 <sup>c,A</sup>
14	0,14±0,00 <sup>a,A</sup>	0,15±0,02 <sup>b,A</sup>	0,15±0,02 <sup>b,A</sup>	0,18±0,02 <sup>d,A</sup>
21	0,16±0,02 <sup>b,A</sup>	0,18±0,04 <sup>c,A</sup>	0,14±0,01 <sup>a,A</sup>	0,14±0,03 <sup>b,A</sup>
28	0,14±0,02 <sup>a,A</sup>	0,13±0,02 <sup>a,A</sup>	0,15±0,01 <sup>b,A</sup>	0,13±0,01 <sup>a,A</sup>

W tabeli przedstawiono średnie wartości z odchyleniem standardowym. Różne litery a-d przy wartościach w kolumnach lub A-B w wierszach oznaczają różnice statystyczne pomiędzy wynikami otrzymanymi dla poszczególnych wariantów.

Ocena sensoryczna wykazała, że akceptowalność wszystkich badanych wariantów obniżała się wraz z wydłużeniem czasu przechowywania, co znalazło odzwierciedlenie w niższych ocenach po 28 dniach (**P4-Rysunek 6**). Po 7 dniach jabłka powlekane były oceniane jako bardziej atrakcyjne wizualnie niż próbki kontrolne, co można wiązać z mniejszą utratą wody, zachowaniem jędrności oraz zwiększonym połyskiem powierzchni. Nie stwierdzono istotnych różnic w ocenie zapachu pomiędzy wariantami, natomiast jabłka niepowlekane uzyskały największe noty za smak. Po 28 dniach najwyższe oceny ogólne, uzyskały jabłka pokryte powłoką zawierającą kwas protokatechowy. Różnice pomiędzy tym wariantem a próbką kontrolną mieściły się jednak w przedziale 0,5–1 punktu, co wskazuje raczej na subiektywne preferencje konsumenckie niż istotne różnice jakościowe. Wyniki oceny sensorycznej potwierdziły ponadto, że jabłka powlekane były postrzegane jako bardziej soczyste, chrupiące przy pierwszym kęsie i jędrne w przekroju, podczas gdy owoce niepowlekane oceniano jako bardziej miękkie i mączyste.

## 5. PODSUMOWANIE I WNIOSKI

Badania przeprowadzone w ramach pracy doktorskiej pozwoliły na weryfikację założonych hipotez i potwierdziły zasadność stosowania kwasów fenolowych w tworzeniu folii pektynowych. Zastosowanie opracowanych roztworów powłokotwórczych w powlekanii jabłek odmiany *Golden Delicious* umożliwiło wydłużenie trwałości i przydatności konsumpcyjnej owoców podczas 28-dniowego przechowywania. Na podstawie wyników sformułowano następujące wnioski:

- folie pektynowe uplastycznione glicerolem i wzbogacone w kwasy fenolowe (ferulowy, galusowy, kawowy, kumarowy, protokatechowy, synapinowy) tworzyły jednorodne, gładkie struktury o dobrej kompatybilności składników, co umożliwiała ich dalszą charakterystykę materiałową;
- wprowadzenie kwasów fenolowych zwiększyło grubość folii pektynowych, zmniejszyło ich zawartość wody oraz wpłynęło na właściwości fizyczne, w tym wrażliwość na wilgoć i parę wodną a jednocześnie wszystkie folie wykazywały porównywalną zdolność do adsorpcji wilgoci;
- kwasy fenolowe zwiększyły dyfuzyjność matrycy pektynowej w stosunku do pary wodnej i charakteryzowały się mniejszą jasnością i połyskiem oraz zwiększoną barierowością wobec światła, w tym UV i światła widzialnego, jednakże barierowość folii pektynowych wobec pary wodnej, tlenu i ditlenku węgla była determinowana rodzajem zastosowanego dodatku kwasu fenolowego;
- obecność kwasów fenolowych zwiększyła wytrzymałość mechaniczną folii i wykazywała zróżnicowany efekt uplastyczniający w porównaniu z foliami kontrolnymi;
- dodatek kwasu protokatechowego zmniejszył lepkość pozorną pektynowych roztworów powłokotwórczych, co można wiązać ze zmianami hydrofobowości cząsteczek pektyny, wpływającymi na aplikacyjność i rozprowadzanie powłoki;
- powłoki pektynowe, zwłaszcza wzbogacone w kwasy fenolowe, skutecznie pełniły funkcję bariery ochronnej, ograniczając zmiany jakościowe jabłek i spowalniając procesy fizjologiczne w czasie przechowywania w warunkach konsumpcyjnych;
- proces powlekania wpływał na stabilizację składu chemicznego jabłek w czasie przechowywania, w tym utrzymanie wartości ekstraktu i kwasowości, zmniejszenie utraty jędrności oraz ograniczenie produkcji etylenu, co potwierdza wydłużenie okresu przydatności do spożycia.

### **Praktyczne zastosowanie w przemyśle:**

Badania w ramach niniejszej pracy doktorskiej wykazały, że wprowadzenie kwasów fenolowych do matrycy pektynowej umożliwia otrzymanie folii o jednorodnej strukturze, zwiększonej wytrzymałości mechanicznej, zmniejszonej zawartości wody oraz poprawionej barierowości wobec światła, pary wodnej i gazów. Dodatkowo kwasy fenolowe wpływały na reologię roztworów powłokotwórczych, ułatwiając aplikację i rozprowadzanie powłoki, oraz nadawały foliom aktywność przeciwutleniającą, co zwiększa ich funkcjonalność jako materiału ochronnego.

Zastosowanie opracowanych folii pektynowych jako powłoki ochronne w powlekanii jabłek odmiany *Golden Delicious* skutkowało zmniejszeniem ubytków masy, spowolnieniem dojrzewania i ograniczeniem zmian jakościowych owoców podczas 28-dniowego przechowywania w warunkach konsumpcyjnych. Uzyskane wyniki wskazują, że folie pektynowe kontrolne i z dodatkiem badanych kwasów fenolowych (kawowy i protokatechowy) mogą stanowić efektywną, naturalną powłokę ochronną w przemyśle spożywczym, przedłużając trwałość i przydatność jabłek do spożycia.

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## 7. DOROBEK NAUKOWY

### WYKAZ PUBLIKACJI STANOWIĄCYCH ROZPRAWĘ DOKTORSKĄ

[P1] Mikus M., Galus S. (2025). Extending the Shelf Life of Apples After Harvest Using Edible Coatings as Active Packaging – A Review. *Applied Sciences*, 15(2), 767 (100 pkt, IF 2,5). <https://doi.org/10.3390/app15020767>

[P2] Mikus M., Galus S. (2025). The Effect of Phenolic Acids on the Sorption and Wetting Properties of Apple Pectin-Based Packaging Films. *Molecules*, 30(9), 1960 (100 pkt, IF 4,6). <https://doi.org/10.3390/molecules30091960>

[P3] Mikus M., Galus S. (2026). The Effect of Selected Phenolic Acids on the Functional Properties of Pectin-based Packaging Films. *Applied Sciences*, 16(2), 797 (100 pkt, IF 2,5). <https://doi.org/10.3390/app16020797>

[P4] Mikus M., Szulc K., Galus S. (2025). The Use of Bioactive Edible Coatings Based on Pectin and Phenolic Acids for Enhancing Quality Attributes of Golden Delicious Apples During Storage. *Processes*, 13(12), 3821 (70 pkt, IF 2,8). <https://doi.org/10.3390/pr13123821>

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Pakulska, A., Mikus, M., Karwacka, M., Galus, S. (2025). The Potential of Bilberry and Blackcurrant Juices as a Source of Colorants in Intelligent Pectin Films. *Applied Sciences*, 15(19), 10789, 1-19. <https://doi.org/10.3390/app151910789>

Mikus, M., Galus, S. (2024). Zastosowanie procesu powlekania w monitorowaniu jakości pozbiorniczej jabłek. *Przemysł Spożywczy*, 78(12), 34-36. <https://doi.org/10.15199/65.2024.12.4>

Balcerzak, J., Mikus, M., Galus, S. (2024). Badanie migracji substancji aktywnych z biopolimerowych folii opakowaniowych. *Ważenie. Dozowanie. Pakowanie*, 2, 40-43.

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Wiśniewski, J., Zasada, K., **Mikus, M.**, Galus, S. (2022). An attempt to develop fast dissolving biopolymer-based pouches for instant coffee®. *Technological Progress in Food Processing*, 2, 70-78.

Galus, S., **Mikus, M.**, Ciurzyńska, A., Janowicz, M. (2022). Whey Protein Isolate-Based Edible Coatings Incorporated with Jojoba Oil as a Novel Approach for Improving the Quality of Fresh-Cut Root Parsley during Refrigerated Storage. *Applied Sciences*, 12, 9023. <https://doi.org/10.3390/app12189023>.

Twardowska, A., **Mikus, M.**, Galus, S. (2022). Barwniki naturalne i ich rola w opakowaniach inteligentnych do żywności. *Ważenie. Dozowanie. Pakowanie*, 1(85), 34-39.

Galus, S., **Mikus, M.**, Ptak, K. (2021). Biotworzywa – charakterystyka i zastosowanie do żywności. *Ważenie, Dozowanie, Pakowanie*, 82, 2, 38-43.

Galus, S., **Mikus, M.**, Ciurzyńska, A., Domian, E., Kowalska, J., Marzec, A., Kowalska, H. (2021). The Effect of Whey Protein-Based Edible Coatings Incorporated with Lemon and Lemongrass Essential Oils on the Quality Attributes of Fresh-Cut Pears during Storage. *Coatings*, 11, 745. <https://doi.org/10.3390/coatings11070745>.

**Mikus, M.**, Galus, S., Ciurzyńska, A., Janowicz, M. (2021). Development and Characterization of Novel Composite Films Based on Soy Protein Isolate and Oilseed Flours. *Molecules*, 26, 3738. <https://doi.org/10.3390/molecules26123738>.

**Mikus, M.**, Galus, S. (2020). Powlekanie żywności – materiały, metody i zastosowanie w przemyśle spożywczym. *Żywność. Nauka. Technologia. Jakość.*, 27, 4(125), 5-24. <https://doi.org/10.15193/zntj/2020/125/355>.

### **Rozdziały w monografiach**

**Mikus, M.**, Galus, S. (2022). Biopolymers from Agriculture Waste and By-Products. In A. K. Nadda, S. Sharma, & R. Bhat (Eds.), *Biopolymers: Recent Updates, Challenges and Opportunities* (pp. 111-128). Springer International Publishing. [https://doi.org/10.1007/978-3-030-98392-5\\_6](https://doi.org/10.1007/978-3-030-98392-5_6)

### **Udział w projektach badawczych**

Wykonawca w projekcie BIOSTRATEG 3 pt. „Opracowanie innowacyjnej metody obliczania śladu węglowego dla podstawowego koszyka produktów spożywczych” (BIOSTRATEG3 / 343817/17 / NCBR / 2018.), finansowanym przez Narodowe Centrum Badań i Rozwoju przy zadaniu „Rozwój technologii produkcji zdrowej żywności z uwzględnieniem zarządzania odpadami odżywczymi i metodami obliczania śladu węglowego”, 02.2019-06.2020

### **Doniesienia konferencyjne**

**Mikus, M., Galus, S.** Wpływ powłok polisacharydowych z nanocząstkami tlenu cynku na wybrane wyróżniki jakościowe jabłek w czasie przechowywania. XXIX Sesja Naukowa Sekcji Młodej Kadry Naukowej PTTŻ "Żywność w oczach młodego pokolenia, Kraków, 29-30.05.2025 r., prezentacja ustna

**Mikus, M., Galus, S.** Zastosowanie powłok pektynowych wzbogaconych w kwasy fenolowe w monitorowaniu jakości pozbiorniczej jabłek, VIII Symposium Inżynierii Żywności, Warszawa, 1-3.07.2024 r., prezentacja ustna

**Mikus, M., Galus, S.** Wpływ wybranych kwasów fenolowych na właściwości optyczne, sorpcyjne i zwilżające jadalnych folii pektynowych. XXVII Sesja Naukowa Sekcji Młodej Kadry Naukowej „Rozwój Nauk o Żywności. Zrównoważona przyszłość.” Warszawa, 11.05.2023 r., prezentacja ustna

**Mikus M., Galus, S.** Wybrane właściwości fizyczne folii pektynowych wzbogaconych o kwasy fenolowe”, XXVIII Sesja Naukowa Sekcji Młodej Kadry Naukowej, Gdańsk, 16-17.05.2024 r., prezentacja ustna

**Mikus M., Galus, S.** Udział w konferencji „II edycja Konferencji Młodych Naukowców” – Wpływ powłok jadalnych na wyróżniki jakościowe wybranych warzyw korzeniowych, 01.06.2022 r., prezentacja online

**Mikus M., Galus, S.** Barwa i analiza sensoryczna krojonych jabłek powlekanych wybranymi powłokami hydrokoloidowymi, IV Konferencja Doktorantów „Cztery Żywioly – współczesne problemy w naukach o życiu”, 14.12.2021 r., prezentacja online

### **Odbyte staże naukowe**

E.Wedel, Warszawa, 01.06.2022-31.08.2022 r. (3 m-ce)

Instytut Biotechnologii Przemysłu Rolno-Spożywczego im prof. Wacława Dąbrowskiego,  
Państwowy Instytut Badawczy, Warszawa, 05-07.2025 r. (3 m-ce)

### **Nagrody i wyróżnienia**

Nagroda zespołowa II stopnia JM Rektora SGGW za osiągnięcia badawcze, 2022

Nagroda zespołowa II stopnia JM Rektora SGGW za osiągnięcia badawcze, 2024

### **Pozostały dorobek naukowy i organizacyjny**

- szkolenia, warsztaty i webinaria związane z tematyką pracy doktorskiej;
- szkolenia z zakresu doskonalenia umiejętności miękkich, w tym m.in. umiejętności interpersonalnych, zarządzania projektami, wyboru czasopism naukowych i pisanie artykułów naukowych;
- czynny udział w Festiwalu Nauki w latach 2022-2024;
- czynny udział w Dniach SGGW w latach 2022-2024;
- członek Polskiego Towarzystwa Technologów Żywności;
- opieka nad pracami dyplomowymi pod kierunkiem dr hab. Sabiny Galus, prof. SGGW

Łączna liczba cytowań wg bazy Scopus 172

Indeks Hirscha 7


**8. PUBLIKACJE STANOWIĄCE ROZPRAWĘ DOKTORSKĄ ORAZ  
OŚWIADCZENIA WSPÓLAUTORÓW**

## **Publikacja 1**

**Mikus M., Galus S.** (2025). Extending the Shelf Life of Apples After Harvest Using Edible Coatings as Active Packaging – A Review. *Applied Sciences*, 15(2), 767, 1-34.  
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Review

# Extending the Shelf Life of Apples After Harvest Using Edible Coatings as Active Packaging—A Review

Magdalena Mikus and Sabina Galus \* 

Department of Food Engineering and Process Management, Institute of Food Sciences, Warsaw University of Life Sciences, 159c Nowoursynowska St., 02-776 Warsaw, Poland; magdalena\_mikus@sggw.edu.pl

\* Correspondence: sabina\_galus@sggw.edu.pl

**Abstract:** Extending the shelf life of perishable food, such as apples, and storing them in cold conditions and/or controlled atmospheres have been of great interest in the last decades. Apples are very valuable fruits with many health benefits, but during storage at ambient conditions, they ripen quickly and lose moisture, causing lower crispness or other negative effects, resulting in waste problems. There has been growing attention to protective edible coatings or active packaging films based on biopolymers and natural bioactive substances. Edible coatings and films allow for combination with functional ingredients or compounds, affecting the maintenance of the postharvest quality of fruits and vegetables. They also ensure the preservation of the sensory characteristics of food, and they can have antimicrobial or antioxidant properties. All these aspects play a significant role in the storage of apples, which can also help prevent waste, which is in line with the circular economy approach. The functionality of coatings and films is closely related to the type, content, and composition of active compounds, as well as their interaction with biopolymers. Active coatings with the addition of different functional compounds, such as plant extracts, phenolic acids, and nanoparticles, can be an alternative solution affecting the postharvest quality of apples during storage, maintaining the fruit's stability, and thus minimising their waste. The most important issues related to the latest reports on improving the postharvest quality of apples using edible coatings incorporated with various active substances were evaluated. Agricultural conditions and factors that affect the postharvest quality of apples were described. The requirements for protective coatings for apples should be focused on low-cost materials, including waste-based resources, good miscibility, and compatibility of components. Those factors combined with the storage conditions may result in shelf life extension or retention of the postharvest quality of apples, regardless of the variety or cultivation techniques.

**Keywords:** apples; *Malus* spp.; edible coatings; edible films; active packaging; postharvest quality



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## 1. Introduction

Current estimates suggest that by 2050 the global human population will reach 9 billion, which would require an increase in food production of 100–110%. This will be accompanied by an urgent need to address critical issues, including reducing food waste, especially perishable fruits, and vegetables. This increase in production must be sustainable and consider the environmental, social, and economic impacts of production and distribution. Therefore, fruit production presents certain challenges, including adapting to climate change, maintaining sustainable production practices, and maintaining quality and storage

efficiency after harvest [1]. Food waste is a major problem today. According to the Food and Agriculture Organisation (FAO), about 40% of fruit and vegetables are postharvest losses before retail, and about 50% are post-retail losses [2]. Recently, due to the growing awareness of a healthy lifestyle, intensive research has been carried out on innovative ways to improve the shelf life of food without the use of preservatives. Edible films and coatings used for food have become a particularly important aspect [3]. Innovations and technological progress in the area of food enable the use of edible coatings to reduce the loss of quality and quantitative losses of fruit and vegetables, the demand for which has been constantly increasing in recent years [4]. This is due to consumer concerns about implementing healthier eating habits and practical food products that can be consumed easily and quickly. Due to the instability of fruits resulting from the destructive action of microorganisms, respiration, ripening, and water loss, it is possible to modify and control the internal atmosphere by coating, thus extending their shelf life and preserving their organoleptic and nutritional properties [5]. It is possible to minimise the reduction in nutritional value and unpleasant taste and odour [6,7].

Food packaging is of great importance for fresh food products, as it aims to maintain the quality after harvest. When optimised, food packaging increases the convenience, safety, and quality of food, which is very important for society, while reducing additives, food waste, and the incidence of food poisoning. Modified Atmosphere Packaging (MAP) plays a significant role in packaging, especially for fresh fruits and vegetables. Its main advantages include the ability to reduce the respiration rate, delay ripening and discolouration, prevent the occurrence of unpleasant odours and flavours, and can also inhibit the growth of pathogens and spoilage organisms [8]. Refrigeration technologies used in the food supply chain are designed to maintain the appropriate temperature and humidity environment for perishable products, such as fruit. An integral and efficient cold chain system must maintain perishable food within the appropriate temperature and humidity range from the point of harvest to the point of final consumption. Any deviation from these standards can render the entire logistics chain useless and generate losses. For example, low humidity can accelerate the deterioration of sensory quality, while high humidity causes bacterial growth [9].

Fruits and vegetables occupy a significant position in the human diet due to their large consumption being linked to various health and nutritional benefits [10]. However, fresh produce is characterised by a relatively short post-harvest shelf life. This is mostly because they function as living tissues until they are consumed [11]. According to the Food and Agriculture Organisation of the United Nations (FAO), the total global production of apples in the last years was about 90 million tonnes [12]. Despite the occurrence of weather anomalies, i.e., drought, hail, and unfavourable weather conditions occurring during the flowering period, the harvest of apples, thanks to the introduced modernisation, shows an upward trend. These modernisations include irrigation systems and installations protecting against frost and hail. During storage, apples are susceptible to a myriad of biotic and abiotic diseases, which also affect the occurrence of economic losses. Usually, a number of phytopathogenic microorganisms are responsible for the decrease in fruit quality after harvest, which in turn affects the occurrence of significant losses in storage and distribution networks. It is estimated that about 90 species of fungi are responsible for apple rot occurring during storage. Even in developed countries with advanced storage solutions, fungal rot can be the cause of up to 25% of the total apple yield loss [13].

A shorter shelf life of fruit is caused by, among other things, improper storage temperature, gas composition of the atmosphere, too long transport time, or improperly selected type of packaging, which does not fulfil its function and affects the loss of fruit quality [14]. When storing fresh fruit, high relative humidity has a positive effect on extending the stor-

age life [15]. The fruit's respiration rate and the concentration of gases present between the internal and external atmospheres (water vapour, oxygen, carbon dioxide) must be taken into account and controlled. Ethylene evolution during storage and transport favours faster degradation of fresh products, generating product losses and, thus, financial losses [16]. Apples, belonging to climacteric fruits, i.e., ripening after harvesting under the influence of ethylene secretion, change their taste, smell, and colour—contributing to the yellowing of the green parts and texture, causing them to wilt [13]. In addition, under the influence of ethylene, apples can change the content of sugars and synthesise volatile aromas [17,18]. Due to the complexity and difficulty of detecting volatile aroma components, detecting ethylene content can be a simple way to assess the flavour quality of apples. So far, there have been few studies on the relationship between ethylene and the occurrence of volatile aroma compounds. In the study conducted by storing “Fuji” apples at 4 °C for 42 days, it was found that detecting ethylene level can reflect the aroma level, which is a reference to the intrinsic quality of apples [19]. Exposure of fruit to low temperatures stimulates ethylene synthesis, which explains why many apple varieties require chilling to induce ripening. Therefore, transport to distant markets should take place in refrigerated conditions (cold rooms, containers, specialised semi-trailers), with particular attention to determining the appropriate size of the chamber to ensure air circulation. There is a lot of research on the use of edible coatings in improving the quality of fresh-cut apples as minimally processed and convenient-to-eat fruits [20–24]. Changes in the quality attributes are different, and colour changes are the most important. Thus, edible coatings are incorporated with different functional compounds to maintain their quality during refrigerated storage [25]. However, these types of products packaging, which provides stability and protection from microbial contamination. Taking into account the environmental issue and the waste problem of packaging, there is a great interest in mild technologies for whole apples minimalising usage of packaging, such as protective edible coatings produced from natural materials.

Therefore, this work aimed to characterise the most important issues related to the latest reports on improving the postharvest quality of apples with the use of edible coatings enriched with various active substances. Agricultural conditions and factors that affect the postharvest quality of apples were described, as were coating materials and food coating technology. The potential of active packaging in the emerging food industry and the goals and benefits of reducing fruit waste and extending its shelf life were demonstrated.

## 2. Characteristics of Apples

### 2.1. Nutritional Values

Apples are very valuable fruits because they are attributed to many health benefits. Studies have shown that the polyphenols present in apples positively affect the prevention of many diseases, including type 2 diabetes, cancer, hypertension, asthma, infections, cardiovascular diseases, and many others. In addition, they are a source of vitamin C, which is a powerful antioxidant responsible for neutralising free radicals. Moreover, apple fibre has a positive effect on health, including preventing the occurrence of colon cancer [26]. A new area of interest for scientists is obtaining fibre from plant residues, which are a valuable source of polyphenols and carotenoids. According to scientific reports, 100 g of apple pomace contains from 4.4 to 47.3 g of fibre, and its content depends on the variety of apples and the method of their extraction [27].

Despite the consumption of apples being still insufficient, at the beginning of the previous decade in the European Union, about 2.2 million children were overweight, and over 5 million were obese. Fruit and vegetables play a significant role in the human diet, and the World Health Organisation (WHO) recommends eating five servings of fruit and vegetables a day, with a total weight of about 400 g [28]. Unfortunately, statistical data

published by Eurostat show that the average daily intake is lower than recommended. Too little consumption of fruits and vegetables can lead to many serious diseases. According to data published by the WHO in 2017, about 3.9 million deaths worldwide were caused by insufficient fruit and vegetable consumption, leading to 14% gastrointestinal cancers, 11% cardiovascular diseases, and 9% stroke [29]. Apples contain phenolic and triterpene compounds, one of the most important biologically active compounds responsible for the prevention of various diseases. It is suggested that the health-promoting effect of polyphenolic compounds and dietary fibre components may reduce the incidence of cardiovascular diseases by lowering cholesterol and limiting lipid oxidation [30].

Apples also contain organic acids, which include ascorbic, malic, citric, and maleic acids, as well as sugars—glucose, fructose, sucrose, and xylitol. In addition, apples also contain vitamins, macronutrients (potassium, sodium, magnesium, calcium, phosphorus), and trace amounts of elements such as iron, zinc, manganese, and copper [31]. The advantage of apples is also their low caloric value—a medium-sized apple has approx. 80 kcal [32]. The health benefits of apples depend mainly on their antioxidant activity, conditioned by the presence of phenolic compounds, which are, among others, secondary metabolites in apples. Depending on abiotic and biotic factors, the quantitative and qualitative composition of phenolic compounds may vary, and the highest content of phenolic compounds is found in apple skins. It has been proven that daily consumption of fruits containing large amounts of biologically active complexes reduces the risk of developing chronic diseases [33]. Due to the resulting large health benefits, it is recommended to increase the amount of apples consumed in the diet, because the studies show that eating apples has a beneficial effect on circulatory system diseases and reduces the risk of cancer. In addition, eating apples has a beneficial effect on alleviating Alzheimer's disease, the deterioration of cognitive functions in the ageing process, diabetes, controlling body weight, or the digestive tract [34].

## 2.2. Agricultural Conditions and Factors Affecting the Quality of Apples

The most suitable areas for the cultivation of apple orchards are elevated areas, slopes, or gentle slopes and plains. The apple tree is a temperate climate tree, while differences in varieties are related to specific thermal requirements during the growing season [14,35]. It is recommended to grow apples whose ripening time does not exceed 140 days after flowering. Late varieties of apple trees, grown in the southern and southwestern parts of Poland, reach full maturity after about 150 days. The soil, which should be fertile, slightly acidic, and airy, plays an important role in ensuring proper growing conditions. The least productive and yielding fruit of poor quality are orchards located in a climate with warm days and nights [36]. The quality of stored apples is closely related to the complex process of their storage, which affects the achievement of the appropriate result. The course of storage is affected by many factors, such as the initial characteristics of the raw material, agrotechnical conditions, degree of maturity at the time of harvesting, storage time and temperature, the concentration of carbon dioxide, oxygen, ethylene content, and humidity in the chamber. In addition, the course of apple storage also depends on the agricultural practices used [37]. The cost of fruit harvesting can be from 20 to 40% of the total farm production costs. Losses on the farm may also result from irregularities during the key stage, which is the harvest itself: hitting, packing, and overflowing of containers in the orchard. Bruises on apple fruit, characterised by soft and susceptible-to-damage skin, may occur in up to 35% of the harvested and transported fruit. Harvesting fruit during the warmer part of the day also affects the susceptibility of fresh fruit to the formation of unsightly bruises, causing faster ageing, flaccidity, and wilting [17].

A threat that has been noted in recent years, causing loss of quality of apples, is the pathogen that causes mycosis (*Colletotrichum fructicola*). As it is a pathogen whose

development depends on the climate, it will not survive the conditions during the winter in Poland, outside the fruit storage system. The spread of the pathogen can occur through direct contact with the mycelium, which can be carried over short distances by insects or wind. According to reports, this is a species of fungus that is not subject to mandatory eradication in European Union countries [38]. On the other hand, according to Popescu et al. [39], due to the presence of phenolic and flavonoid compounds in the skin, which includes catechins, phloridins, tannins, and chlorogenic acid, as well as its thickness and structure, apples are fruits that are quite resistant to microorganisms that cause rot.

The occurrence of unfavourable weather conditions, e.g., frost, generates losses in apple production. To prevent this, orchard owners use innovative fruit cultivation and storage methods, e.g., modern storage rooms. A tightly closed hall enables the storage of fruit even in the conditions of overselling in the amount of approx. 13 tonnes, thanks to which an extended sales period is possible. The innovative storage system provides a controlled temperature and atmosphere (oxygen—1.2%, carbon dioxide—2%), inhibiting life processes. The cost of building such a hall is approx. EUR 115,000 [40]. The establishment of new orchards and their cultivation are necessary to maintain the continuity of production on the farm and to maintain the high position of Poland in terms of the number of apples produced on global markets [41].

### 2.3. Storage and Harvesting Conditions

The selection of the right temperature and storage in optimal conditions of the gas composition of the atmosphere must meet the requirements of individual varieties [17]. Typically, apples are stored at a temperature of 0 to 4 °C, which allows them to maintain their quality for several months. The relative humidity should be maintained at 85 to 95%, and the weight loss of apples due to transpiration and respiration should not exceed 3.5% at the end of storage. After harvesting, the fruit is usually placed in large plastic packages (1.20 × 1.00 × 0.78 m) of approx. 300 kg, is stacked close together and up to a maximum of 12 levels. For this reason, there is a change in the thermal conditions in the storage rooms, caused by differences in air flow, to which the top layers of apples are most exposed. Refrigeration systems in storage chambers that cause temperature fluctuations can cause the temperature to drop below the dew point, resulting in condensation of some of the water vapour in the humid air. Condensed water manifests itself in the presence of single droplets or a film on the surface of apples, hurting the skin, structure, and surface texture and also providing excellent conditions for the proliferation of microorganisms [42]. Each change in temperature by 10 °C causes a 2- or 3-fold change in the rate of biological reactions [43]. Nevertheless, an important factor during fruit storage is the gas composition of the atmosphere, affecting, among others, changes in the antioxidant activity of apples. Determining the optimal storage conditions allows for maintaining the appropriate quality of apples with minimally changed chemical composition. In the literature, there are many reports on storing fruit in chambers with low oxygen content (approx. 1 kPa) or very low oxygen content (0.5 and 0.7–0.8 kPa), which is associated with slowing down the metabolism, production of ethylene, and reduction of fermentation processes [44].

Ethylene (C<sub>2</sub>H<sub>4</sub>) is a simple hydrocarbon that occurs in gaseous form. It is also a naturally occurring substance that affects the development of flowering, ripening, and ageing of fruits. By affecting even at low temperatures, it can cause many biochemical changes, accelerating the metabolism of fruits and vegetables [45]. Therefore, this hydrocarbon significantly extends the shelf life of fruits and vegetables after harvest. Increasing the amount of ethylene causes fruit and vegetables to soften, as well as the degradation of chlorophyll, resulting in progressive spoilage. Apples, sensitive to ethylene content, can be protected by coating to delay the ripening process [46]. The use of edible coatings delays

ethylene production during apple storage compared to detecting the amount of ethylene produced during the storage of uncoated apples. The use of edible coatings also delays microbiological degradation [47]. The degradation of chlorophyll under the influence of ethylene is commercially used to remove the green colour of, e.g., lemon peel [48]. Ethylene removal systems contained in packaging can effectively slow down the undesirable effects of ethylene during storage. The research shows that the ethylene volume in the packaging of fresh products can vary from 0.017–0.035  $\mu\text{L}$  in shopping centres to 0.06  $\mu\text{L}$  in distributor centres. In addition, using potassium permanganate ( $\text{KMnO}_4$ ) removes ethylene—causing the breakdown of the double bond, while zeolite and activated carbon can adsorb it. Potassium permanganate is used for its cost-effectiveness, but some studies indicate its toxic effects on the human body [49,50]. Due to the role of ethylene during fruit ripening, they can be divided into climacteric and non-climacteric. The first group concerns fruits that are capable of secreting a large amount of ethylene, which is conducive to the ripening process, e.g., peaches, apples, bananas, or kiwi. Non-climacteric fruits produce only a small amount of ethylene during ripening, and the ageing process does not depend on its production. This group includes strawberries, grapes, watermelons, pineapples, or citrus. Both in the case of fruit and vegetables, technological processes taking place during production, such as peeling or cutting, also have an impact on the increased production of ethylene. Changes can be noticed in just a few minutes. In addition, there are reports of reduced ethylene production when storing climacteric fruits (e.g., apples) in an atmosphere with increased carbon dioxide  $\text{CO}_2$  and reduced oxygen  $\text{O}_2$  atmospheres [51]. Hussain et al. [52] studied the degradation of bioethylene under refrigeration conditions ( $3^\circ\text{C}$ ) using a nanoparticle photocatalyst,  $\text{TiO}_2$ . The authors showed that nanoparticles can increase the capture capacity due to the synergistic system of ethylene gas released from fruits and vegetables. There are reports of inducing or inhibiting the production of ethylene during various processes in plants. Unfortunately, too little information has been reported so far on the effect of melatonin on the quality of apples during storage. The research conducted by Onik et al. [53] shows that melatonin delayed the ripening of apples by inhibiting the biosynthesis of ethylene.

One of the varieties of packaging that controls the amount of oxygen and carbon dioxide between the packaging and the environment is the so-called “smart films”. They are mainly used for fruit and vegetables, allowing for specific regulation of oxygen flow and controlling the ongoing respiration and ripening process. In addition, they make it possible to limit the development of undesirable anaerobic bacteria [54]. Intelligent packaging can also be used to monitor temperature changes or the generation of volatile amines and hydrogen sulphide during the shelf life of food products. Monitoring factors related to microbial growth, which is influenced by the formation of volatile compounds, temperature changes, and the presence of oxygen, is essential to ensure food safety and quality. With technological advances and the increasing demand for this type of packaging, the bakery industry is showing increasing interest in this type of packaging. The use of such packaging is associated with the possibility of adding antimicrobial and antioxidant compounds that can prevent the formation of undesirable flavours by microorganisms. Another great advantage is the possibility of adhesion of probiotics, which inhibit the growth of fungi [55]. Future research should also focus on recovering and extracting bioactive compounds from food processing waste while maintaining innovative “green” technologies. So far, there are too few studies and reports on using edible coatings and films produced from fruit processing waste for packaging fresh meat or poultry products. In addition, using nanotechnology is a promising solution to improve the functional properties of edible films and coatings. For example, edible coatings with 0.5% nano chitosan lowered the maximum ethylene production of apples and markedly eliminated the climacteric

respiration peak. The coating slowed down the natural fruit ripening, thus enhancing the quality and storage life of the apple [56]. Therefore, future work should focus on the use of inexpensive and unmanaged waste to reduce the production costs of edible films and scale up production from a commercial scale while maintaining economic viability [57]. Due to growing demand and global concerns about food safety, nanotechnology is gaining interest as a solution to increase food supply. Expansive surface area and antimicrobial efficacy are versatile in traditional packaging and in integration with biopolymer matrices. Studies have shown that using an edible coating based on a nanoemulsion containing citrus essential oil and chitosan nanoparticles for packaging seafood extends its shelf life. In contrast, the use of an edible coating based on sodium alginate containing a nanoemulsion of lemongrass essential oil on fresh-cut apples inhibited the growth of *E. coli* while maintaining moisture, reducing browning, and reducing respiration [58]. In summary, intelligent edible coatings can adjust their permeability properties or release bioactive compounds that are dependent on the needs of the fruit. The result is optimal preservation conditions throughout the supply chain [1].

#### 2.4. Waste Problem of Apples

The main environmental factors affecting consumer acceptance and maintaining the sensory quality of fresh food products include temperature and relative humidity. Incorrect selection of temperature reduces the quality and shelf life of, generating waste. The use of low temperatures during storage reduces the physiological activity of tissues and the activity of microorganisms causing food spoilage, extending the postharvest life of the product. Visible changes such as wilting, shrinkage, and desiccation occur at 3–6% moisture loss, contributing to significant quality losses in many types of fruit and vegetables. In addition, the occurrence of water loss affects the generation of economic losses, because many goods from the products industry are sold by weight [59]. Preservation of apples after harvest consists mainly of storage in low-temperature chambers with a controlled atmosphere. Despite the use of refrigerated storage conditions, it is possible to develop *Penicillium expansum* on apples, the so-called postharvest pathogen that produces patulin, a secondary metabolite with toxic effects that can lead to huge losses during the storage of apples. The reports of the Food and Agriculture Organisation (FAO) also show that nearly 50% of all fruit and vegetables produced worldwide are lost during the supply chain, generating large amounts of waste and causing environmental pollution [60]. Also, according to the FAO, world fruit production reached 870 million tonnes in 2018, with estimated post-harvest losses of fruits and vegetables throughout the supply chain estimated at between 25 and 50%, highlighting the need to develop systems to reduce losses in order to ensure sustainable development [1]. Large areas of agricultural land needed to produce food that is never eaten require large amounts of water and energy and lead to greenhouse gas emissions during processing, storage, and transport [61].

### 3. Edible Coatings

#### 3.1. Formation

Edible coatings are thin layers or layers of edible material that are applied directly to the surface of the food product, while edible films are placed on top of the product after the film has been formed outside of it [62]. The main functions of edible coatings include protection against mechanical damage, light, or contamination, and acting as a semipermeable membrane and a barrier against gases or moisture [63]. In addition, edible coatings reduce the rate of food respiration, enzymatic browning, and the release of volatile compounds into the environment [64]. Edible coatings are created mostly from aqueous solutions of edible materials such as biopolymers, which provide film-forming capacity

and continuous structure when applied to food products. Thus, during the drying process, a uniform layer is developed. Different functional compounds can be added, including plasticisers, antimicrobials, or antioxidants [62].

A better understanding of the mechanism of functional coatings and consumer awareness can contribute to expanding their use to extend the shelf life of fruits and increasing the bioavailability of compounds referred to as bioactive [65]. The primary role of edible coatings is to control moisture loss as well as reduce the rate of undesirable chemical reactions to maintain food freshness, safety, and quality [66]. Also, adding antimicrobials, antioxidants, flavours, or dyes extends the range of their applications [67]. It is increasingly expected that the edible coatings used for fresh fruit will be characterised by high strength, appropriate hydrophobicity, and barriers to gases and light. An important aspect is also the design of coatings that, in addition to improving the taste of products, will comply with the requirements and preferences of consumers; e.g., coatings made with the addition of garlic or ginger, in addition to imparting sensory characteristics, will be an excellent preservative [68].

In the case of edible coatings, two main factors affect their quality: the method of application and the degree of adhesion of the coating to the fruit. One of the methods of applying edible coatings to the product is the immersion method, which consists of placing the sample in a previously prepared coating solution for a certain time (usually from 5 to 30 s), then removing the excess solution and evaporation [69]. In the evaporation stage from the surface of the products, the solvent and excess liquid are removed by heating and drying processes. The advantage of this widely used method is that the product is evenly coated on all sides, even when there is a rough surface. Therefore, the surface properties of the coated product and the nature of the food to be coated also influence the choice of coating method. In the case of fruit, consideration must be given to whether the fruit is climacteric or non-climacteric before applying the edible coating. The conducted research shows that the value of the thickness of edible coatings depends on many properties of the coating solution, e.g., density, viscosity, or surface tension. Observations show that in the case of coating sliced fruit or vegetables, the application of edible coatings is more difficult because the hydrophilic nature of the surface makes it difficult to gel with the coating material of a hydrophobic nature [70,71]. The immersion method is versatile as it can be used over a wide range of coating solution viscosities. It is usually a method used to apply single-layer coatings, but thanks to technical progress, it is possible to obtain multi-layer coatings, applied alternately for more efficient control and greater coating functionality. A multi-layer coating can be obtained by applying, for example, a polysaccharide or protein solution to the surface of the fruit, drying it, and applying another layer, e.g., a lipid. Thanks to this, due to the combination taking place in the biopolymer matrix on the surface of the fruit, the resulting coating has similar properties to the composite emulsion coating, which is one emulsion and is applied as a single one on the surface of the fruit [72]. For solutions of low viscosity, which allows spraying at high pressure (60–80 psi) solution droplets with a diameter of up to about 20 microns, the spray method is practiced [73]. This technique ensures uniform coating, thickness control, and the ability to achieve multi-layer coatings. Automation and continuous production are also possible, as well as temperature control, which affects the stability of highly volatile antimicrobials. In addition, controlling the thickness parameter makes it possible to determine the amount of antimicrobial used to release the substance. The spray technique can combine hydrophilic and hydrophobic substances but requires four stages: two spray coatings and two drying processes. The advantages of this method also include cost reduction, because the spray method is a process that takes place in a short time and is also efficient in terms of solvent and material consumption [57,74]. The electrospray method is also practiced, which involves the use

of a strong electric field to obtain charged droplets with narrow size distribution and micrometric sizes. During this process, it is possible to adjust the size or thickness of the layer by being able to control the flow rate and viscosity of the solution. In the layer-by-layer deposition method, electrostatic interactions occurring on the food surface with charged polyelectrolytes play an important role in improving the adhesion of the coating to the food surface. Due to this method, it is also possible to create two or more thin layers, joined together physically or chemically, thus improving the effectiveness of coatings compared to conventional coatings [57]. This method has become effective during postharvest fruit storage and increased the tightness of coatings. Another coating technique is cross-linking, defined as a complex process involving the linking of polymer chains using covalent and non-covalent bonds. Cross-linked coatings can be obtained by spraying, dipping, or coating a food surface with a coating solution, adding a cross-linking agent to achieve stable coating materials, especially for biopolymer materials derived from proteins or polysaccharides. Due to the presence of more functional groups in the structure of proteins, they are more often used in this method compared to polysaccharides. The advantages of the cross-linking method include good mechanical properties, chemical and thermal stability, as well as improved molecular migration [75]. In the brushing method, the film-forming solution is placed directly on the products' surfaces and to a large extent, this process depends on human factors. The brushing technique requires expertise in obtaining a proper and uniform layer distribution [76].

### 3.2. Materials

Edible coatings and films can be made of polysaccharides, proteins, lipids, and their combinations to obtain composite materials with better physicochemical properties compared to packaging materials obtained based on a single material (Figure 1). Due to their biodegradable and non-toxic properties, they are intended to replace the existing synthetic polymers [77–79]. In general, polysaccharide edible coatings are characterised by their hydrophilic properties as a poor barrier to moisture, and some of them are also less permeable to oxygen. Coatings made from cellulose, amylose, or chitosan are resistant to fats and oils, with considerable strength, flexibility, and transparency [80]. Disadvantages of polysaccharide coatings include difficulties in processing and performance, especially when coating moist products. Edible coatings made from protein sources, on the other hand, are hydrophilic. Usually, the proteins must be denatured by heat, acid, alkali, and/or solvent. The aim is to create a more elaborate structure, which is required for coating formation. The most commonly used plant proteins for coating production are, e.g., corn zein, soy protein, and wheat gluten, while animal proteins include gelatin, casein, and keratin. Lipid coatings are used mainly for their hydrophobic properties, which help create a good barrier against moisture loss. Additionally, coatings made from them are thicker and more brittle [81].

Polysaccharides	Proteins	Lipids	Composite materials
<ul style="list-style-type: none"> <li>• cellulose and derivatives               <ul style="list-style-type: none"> <li>• starch</li> </ul> </li> <li>• chitosan</li> <li>• pectin</li> <li>• alginate</li> </ul>	<ul style="list-style-type: none"> <li>• whey proteins               <ul style="list-style-type: none"> <li>• collagen</li> <li>• gelatine</li> </ul> </li> <li>• soy protein isolate</li> <li>• wheat gluten</li> </ul>	<ul style="list-style-type: none"> <li>• beeswax</li> <li>• candelilla wax</li> <li>• carnauba wax               <ul style="list-style-type: none"> <li>• shellac</li> </ul> </li> <li>• vegetable oils</li> </ul>	<ul style="list-style-type: none"> <li>• mixtures of lipids and hydrocolloids</li> </ul>

**Figure 1.** Division of coating materials.

Edible coatings can act as a barrier against various atmospheric gases, moisture, water vapour, carbon dioxide, and microbes and also slow down respiration and the rate of oxidation reactions in food. However, to ensure adequate protection, they are used in combinations of more than one material, with the addition of low-molecular compounds, showing active and plasticising properties, providing additional properties. Plasticisers are usually polyols such as glycerol, honey, mannitol, propylene glycol, sucrose, or fatty acid esters, which impart strength and flexibility to the coatings, as well as improve the water vapour and gas permeability of the coatings. Active substances added to edible coatings include antimicrobial, antioxidant, taste, colour, or nutritional enhancers, as well as essential oils, dyes, flavours, or nanoparticles [82]. All biopolymers used for edible coatings and films must be recognised as safe by the U.S. Food and Drug Administration (FDA). It is expected that the creation of biodegradable films and edible coatings cannot completely replace artificial packaging materials, but they can be a substitute for basic, conventional packaging [83]. Coatings applied to fruit can be produced from different materials, also obtained from agricultural by-products [84], and they should be characterised by low water vapour permeability, preventing drying, as well as low oxygen and carbon dioxide permeability to slow down respiration and metabolic activity, affecting the ripening process [85]. Pectins (e.g., apple pectin, citrus pectin), a non-starch polysaccharide, are by-products of agricultural processing and are often used to create edible coatings. Pectin is obtained from the cell walls of almost all types of fruit and vegetables, and due to its diverse structure, it has a wide range of applications [86]. Currently, pectin-based edible coatings are commonly used to coat low-moisture fruits due to their hydrophilic nature and in food production as stabilisers, thickeners, and gelling agents, as well as encapsulation agents [87]. Pectin gelation and film formation largely depend on the degree of esterification, which is why modifications are introduced consisting of combining the hydrophilic structure with a hydrophobic lipid compound [3]. Pectins are generally classified as high and low methoxy, having a degree of esterification above and below 50%, respectively, making pectin one of nature's most complex macromolecules. Pectin-based coatings applied to the fruit retain their sensory properties and quality, in addition to being films with poor mechanical properties but resistant to the presence of oils and fats. In addition, they are biocompatible with selective gas permeability, are transparent and have the ability to reduce the loss of nutrients and volatile substances, which is an important aspect from a nutritional point of view. Due to their high availability and cheap cost, they find more and more applications for food coating [88]. In connection with the above-mentioned properties defining pectin, it is possible to produce compatible polymer matrices that are used in the development of active packaging, in addition to the ongoing process of coating food products, which is designed to extend their shelf life and storage quality. One of the popular polysaccharides used in food coatings is alginate, obtained from brown seaweed, which has long been used in the food industry as a thickening and stabilising agent and can retain large amounts of oxygen and carbon dioxide, affecting the good quality of obtained edible coatings [89]. Reducing the respiration rate caused by using edible coatings reduces the spoilage of food, especially fruits containing a large amount of water and thus susceptible to water loss, which makes them soft and easier to shrink. In addition, edible coatings can affect the preservation of firmness and reduce the loss of volatile fragrance components [90]. However, alginate tends to gel and becomes insoluble when polyvalent cations (e.g., calcium ions) are present, forming cross-linked structures [91].

Counteracting fruit waste through the coating, a gentle food processing technology, is now part of the circular economy, which includes, among others, optimisation of production and consumption in the agri-food sector. Thus, the circular economy is a development

strategy that aims to introduce innovative technologies, and organisations, transform industrial systems and production chains, as well as broadly understood changes in consumption habits [92]. The introduction of such a strategy by the European Commission is caused by the continuous depletion of natural resources and the consequences that result from it. The circular economy issue primarily concerns the reduction of the production of large amounts of waste, including losses resulting from food waste, as well as the continuous development of environmentally friendly production and protection of agricultural land [93]. At the level of the European Union, a component affecting the achievement of a sustainable future is the implementation of the assumptions of the European Green Deal. This assumes the reduction of food losses and waste, as well as the use of sustainable consumption and a balanced diet. To this end, mandatory nutrition, food origin, and environmental footprint labelling have been introduced [94]. The European Green Deal has the appropriate tools to eliminate imported food from the European Union market that does not meet specific environmental standards. Also due to the wealth of valuable compounds found in food residues, often referred to as waste, efforts are made to better manage them. To achieve sustainable development and a circular economy, it is necessary to reduce food losses and food waste by at least half. Therefore, the use of edible films and coatings, which will be produced based on by-products from the food industry, may be a promising alternative to improve the efficiency of packaged food, due to the bioactivity of their compounds [95–97]. Therefore, scientists are increasingly focusing on developing and creating greener materials using various “green technologies”, and their introduction to the market aims to improve the delivery of functional bioactive ingredients. Effective valorisation of biodegradable materials allows for an improvement in economic value. The effective replacement of non-biodegradable materials with environmentally sustainable materials contributes to the promotion of a range of economically viable biomaterials. In addition, it is possible to convert them into value-added products, making waste treatment more efficient and reducing the generation of residues [98].

Edible coatings based on pullulan, a microbiologically obtained biopolymer, can form transparent films that dissolve in water. They can be applied directly to the surface of the food along with colour or flavour additives. They are recommended primarily for minimally processed fruits and vegetables due to their limiting effect on fungi [99]. Currently, the commercial pullulan market is estimated to be USD 130 million in 2023, where pullulan is mainly to be used for food packaging systems and pharmaceuticals. It arouses great interest in Japan, China, and the USA, mainly due to its good hydrophilic properties. Pullulan is often combined with other biopolymers, essential oils, sodium alginate, and whey protein isolate to provide improved physicochemical and physicomaterial properties [100].

The proteins used to obtain film-forming solutions are materials of plant origin, e.g., wheat gluten, and zein and animal origin, e.g., whey protein, casein, albumin, or gelatin [64]. Protein coatings are characterised by greater fragility and susceptibility to damage compared to coatings obtained from other materials [101], as well as lower tensile strength compared to polysaccharide films [102]. Maize, soybean, and wheat gluten proteins are mainly used to coat fruit and vegetables because they exhibit gas and lipid barrier properties at low relative humidity [103].

Corn zein, extracted from corn gluten, due to the high content of non-polar amino acids (proline, leucine, alanine), can contribute to obtaining hydrophobic coating materials. On the other hand, using gelatine for edible films results in good resistance to gases and oils but poor mechanical properties and low thermal stability. To remedy this, the addition of appropriate starch materials is recommended. Packaging films made based on wheat gluten are characterised by other properties. Thanks to the ability to cross-link after heating, low production costs, and easy availability, they are one of the favourite sources of protein

used for edible packaging. Unfortunately, edible films and coatings containing gluten are brittle, tend to absorb water after processing, and are highly sensitive to pH values. To improve their physicochemical properties, various proteins, polysaccharides, and organic acids are added [67].

Milk proteins can form plastic, transparent, and tasteless films; e.g., casein in edible films affects their stability at different pH values, temperature, and salt content. To improve the properties of caseinate edible films, polyphenolic compounds such as tannic acid, which affect the achievement of better cross-linking of casein, are added. However, using collagen with the addition of other biopolymers (alginate, agar) and silver nanoparticles to film-forming solutions shows antimicrobial activity against *L. monocytogenes* and *E. coli*. The presence of functional groups in the amino acid side chain of proteins causes covalent and non-covalent cross-linking, which leads to increased insolubility and elongation of the film. It is probable that the conformational changes of proteins after cross-linking cause an increase in the hydrophobic surface at a contact angle greater than 90 °C. An increase in surface hydrophobicity was also observed during the heat treatment of food proteins, resulting from changes in the secondary and tertiary structure by breaking hydrogen bonds [104]. Currently, proteins from many other sources are also used, obtained as by-products or for desirable properties and nutritional value. They include, e.g., peanut protein, lentil protein, sesame protein, pumpkin seed protein, egg protein, or rice protein [67]. Mikus et al. [105] studied films based on soy protein isolate with the addition of oilseed flour. The coatings with the addition of flours were mostly characterised by irregularly occurring droplets of fat, which were visible on microscopic images. The best tensile parameters were recorded for coatings made of evening primrose, linseed, and sunflower flour, while the weakest for coatings were obtained with sesame flour and pumpkin seeds. Thermogravimetric analysis showed that combining soy protein and oilseed flours resulted in coatings with lower stability, probably due to the destruction of hydrogen bonds between the protein and other molecules.

Lipids are defined as small, hydrophobic molecules, which can include, for example, fats, waxes, or sterols, that are potential film-forming materials which are mainly used to reduce water vapour transmission. Covering fruit and vegetables with lipid coatings hurts the increase in carbon dioxide concentration, leading to an increase in the presence of microorganisms and an increase in the content of acetaldehyde and ethanol. Compared to protein or polysaccharide films, films made only of lipids lack structural integrity; therefore, to improve their properties, the lipid component is most often combined with the base structure of the hydrocolloid [106]. The advantage of using lipid films is also to achieve an attractive gloss on the product's surface [107]. In addition, lipid coatings protect against the adverse effects of low temperatures and changes resulting from hypothermia, most often occurring during the cold storage of food products [64]. Waxes used to create edible coatings are substances that, due to their source of origin, can be divided into synthetic (e.g., fatty acids, fatty acid esters and glycerides, hydrogenated oils) and natural (e.g., beeswax, carnauba wax, candelilla wax), corresponding to improving moisture barrier properties, but their use causes deterioration of the mechanical properties of edible films and coatings due to separation between the continuous and dispersed phases [79]. The resulting barrier against water vapour penetration depends on the type of added lipid [108]. Petroleum waxes (paraffin and polyethylene wax) also have a significant potential to influence hydrophobicity, but due to the safety of products intended for consumption, they are not used in them. Lipid coatings are currently used to improve the functionality and mechanical properties of coatings. A greater moisture barrier was observed when oil was added to the coating than when oil was added in the emulsification process [107]. On the other hand, the addition of wax to composite films minimises the weight loss of fruit and

vegetables and also affects the thermal stability of edible coatings and films [79]. Currently, there are also coating composites on the market, e.g., carboxymethylcellulose, fatty acids, and mono-diacylglycerols (TAL Chemicals Company, Great Britain), which, thanks to their properties, effectively affect the shelf life of fruit [83]. Continuous development, as well as progress in materials science and processing technology, creates the opportunity to discover new coating and film-forming materials with improved functional properties. Some studies have shown the promising film-forming potential of flours obtained from whole grain raw materials, e.g., amaranth, quinoa, and chia, or flours obtained from legumes, e.g., peas, lentils, chickpeas, or mung beans. To reduce the amount of waste generated during production, by-products (peel, pomace, seed fractions) are the material for producing flours, which are then a component of films and edible coatings [109]. This way of managing post-production residues generates a low energy cost produced using established techniques, which is important. New materials used to obtain film-forming ingredients are also fruit peels (e.g., pomelo, banana), which are a rich source of nutrients. On the other hand, residues formed during wine production, which are a potential film-forming material, when added to the film, decreased their hydrophilicity [77].

### 3.3. Commercial Edible Coatings for Fruits

The beginnings of edible coatings date back to the 12th and 13th centuries when they were used as waxes applied to oranges and lemons to extend the storage time and delay water loss. In the 1930s, hot-melt paraffin waxes were already used commercially as edible coatings for fresh apples and pears [110]. The first edible “Yuba” films made using soy proteins were recorded in the 15th century in Japan, while in the 16th century in England, the practice of coating fruit and vegetables in lard to prevent moisture loss began. Due to consumers’ growing quality requirements, the market of edible coatings and films is rapidly developing, leading to commercial production. This fact caused the development of many companies towards the packaging industry, and edible coatings produced by many foreign companies are used on the market, including BioEnvelop<sup>®</sup>, COGIN<sup>®</sup>, ENAK<sup>®</sup>, Freshseel<sup>™</sup>, Fry Shield<sup>™</sup>, GREENSOL<sup>®</sup>, Nature Seal<sup>™</sup>, Nutrasave<sup>™</sup>, Opta Glaze<sup>™</sup>, Seal gum, Spray gum<sup>™</sup>, Semperfresh<sup>™</sup>, SHELLAC (E904), and Z-Coat<sup>™</sup> [111]. Also in Poland, in 2016, a Polish researcher, Róża Rutkowska, developed eco-friendly packaging by Scoby Packaging Materials<sup>™</sup> (SPM) with the so-called tea mushroom (kombucha). SCOPY (Symbiotic Culture of Bacteria and Yeast) is an innovative method of obtaining packaging that does not need light, sterile conditions, and advanced technologies for production, and after use, it can be thrown into compost, where it will decompose naturally or simply eaten [112,113].

Coatings may be applied to food surfaces by dipping, spraying, fluidised-bed, and panning. The application of these methods depends on the type of food to be coated, the surface characteristics, and the main purpose of the coating. Dipping is a popular food-coating method that involves immersing the product in a dispersion that forms a coating. This method involves three main steps. The first step involves immersion in an emulsion/coating solution in an amount that allows for complete interaction between the substrate and the coating matrix, deposition. The second step removes excess liquid from the product by deposition and creates a thin coating layer on the surface. The third step involves evaporating the solvent from the product surface by heating or drying. The coating solution can be sprayed using three techniques: a high-velocity air jet is sprayed around a low-velocity fluid; air-assisted spray guns partially atomise the fluid and then use small amounts of compressed air to complete the atomisation. The last spraying method, the so-called pressure atomisation, involves applying an edible coating to the product using pressure. A widely used method in food processing is the fluidisation method, which applies thin layers of coating material to dry particles of low density and/or small size. In

this process, the solution is sprayed through a series of nozzles to create a structure. The panning method involves placing the coated food product in a large rotating bowl called a pan. In the next step, the coating solution is sprinkled or sprayed into the rotating bowl while the product is constantly rotated to distribute the solution evenly. Drying of such a coating occurs using air at a minimum room temperature or higher. Extruded articles with a round or oval structure (e.g., nuts) are suitable for this method [70].

### 3.4. Active Edible Packaging

#### 3.4.1. Active Systems

Food packaging has many tasks: it protects the contents of the package, protects it from contamination and spoilage, and also enables easier movement and storage of food. In addition, they perform an informative function, providing useful information to customers, marketing and advertising, and increasing the share of the product on the market. All these attributes are interrelated and important already at the packaging design stage [104]. Traditional food packaging is currently being modified by incorporating nanotechnology into production or creating active packaging with improved functionality. Edible nanomaterials and coatings are usually a better solution than conventional packaging because they provide better preservation and longer quality retention [114]. An example is the edible coating “Naturen™” developed in Thailand, which is specially designed to preserve the quality of freshly cut fruit, extending its shelf life by three times [115]. Regulations EC 1935/2004 [116] and EC 10/2011 [117] provide detailed information on the declarations of conformity that food products must comply with. These guidelines concern, among others, the use of active packaging materials, the release of substances that are referred to as food additives (accredited), and materials that are used during processing, in direct or indirect contact with food during the entire production process [39].

Active packaging is defined as packaging systems that are intended to extend the shelf life of the product and improve the safety, quality, and durability of food [118], and according to the definition contained in Regulation (EC) No 450/2009 [119], they are defined as “materials and articles whose function is to extend the shelf life or to maintain or improve the condition of packaged food that is intentionally designed so that their ingredients release or absorb substances into or around the packaged food”. Active packaging can be biodegradable films, edible coatings, or sachets, e.g., absorbing ethylene, moisture, or inserts [120]. It is estimated that active packaging systems that cause changes in the environmental conditions of packaged food during its storage in 2024 in the US market are expected to reach USD 6 billion in revenue. In addition, due to the growing interest in the development of active food packaging, it is possible to continuously improve the monitoring and control of environmental conditions to increase the durability of products and ensure their greater integrity. The main systems used as active packaging are, e.g., oxygen and ethylene scavengers, carbon dioxide scavengers/emitters, systems that release or absorb the food, as well as antioxidants, antimicrobials, and regulators to control changes in moisture. In addition, the development and manufacturing of active systems used in food packaging are constantly monitored by various regulatory agencies, such as the Food Safety Authority (EU) and the Food and Drug Administration (USA). This is their correct and safe use, thus constituting the legal basis [121]. The Regulation EC 1935/2004 [116] on materials and articles intended for consumption and contact with food contains information on the requirements for classifying substances responsible for shaping the active functions of the material, classifying them as safe and meeting all the requirements.

The physical size of the active substance diffusing through polymer macromolecules is of great importance in active packaging and release rate effectiveness. In addition, in active release systems, attention should be paid to the volatility of the active substance

present, as, for example, non-volatile antioxidants are suitable for packaging that comes into direct contact with food, releasing the active substances on the surface of the food through stimulus reactions. On the other hand, volatile antioxidants such as natural essential oils, which are substitutes used to provide added value, are characterised by rapid diffusion through the polymer matrix, affecting the retardation of food oxidation in a more effective way than non-volatile compounds due to their high diffusivity [115,122]. Due to the market's changing needs, a constant change in the traditional role of food packaging is noticeable, which is primarily influenced by the changing consumer demand. Other drivers of the evolving changes in packaging include the emergence of many new products on the market, consumer preferences for convenient and clear packaging, and the increasing demand for "safer" food with higher quality and longer shelf life. Increasing requirements for packaging materials have led to the development of new technologies, called active packaging [123]. After conducting market research, sales of active and intelligent packaging increased from 4.95 to 6.0 billion dollars (USD) from 2015 to 2019 (approx. 5.1%). This allowed for an approximate estimation of the total sales level in 2025 at approximately USD 8.6 billion, including active packaging at approximately USD 6.1 million. In addition, the number of scientific publications on this subject increased from 2000 to 2020 by approx. 1200% [45].

Various types of antioxidants are added to edible film-forming preparations, such as organic acids (ascorbic and citric), phenolic acids and compounds, essential oil compounds, volatile phenols, phospholipids, and carotenoids. By incorporating antioxidants into active packaging materials, including sachets, surface coatings, and single- or multi-layer films, it is possible to increase the shelf life of food products [124]. The extension of the shelf life in active forms of packaging results primarily from protecting food against oxidative rancidity, degradation, and discolouration. To achieve such an effect, it is necessary, first of all, to choose the right active agent. In addition, the antioxidant compounds should be compatible with the film [125]. Moreover, it is crucial to improve standard packaging technologies with antioxidant compounds that make it possible to slow down the oxidation processes on the food surface, thus affecting the protection of nutrients against oxidative degradation. Also, the conducted research shows that the addition of ferulic acid in the amount of 100 mg per 100 g of the film-forming solution to the soy protein isolate reduces the oxygen permeability by 18.8%, or by 32% in the case of caseinate films. This phenomenon is probably related to the level of cross-linking between ferulic acid and amino acid residues of the protein, which makes the matrix network more closed, affecting more controlled oxygen transfer, as well as limiting oxidation [126]. In the case of sachets used to control the ethylene in the voids of the packaging or around the food, the metabolic rate is reduced. Ethylene absorbers that extend the shelf life of e.g., fruit include potassium permanganate ( $\text{KMnO}_4$ ), used at 4 to 6%, which is introduced into the sachet. On the other hand, the moisture controller sachet is designed to prevent the accumulation of liquid in the packaging, which can be a favourable environment for developing fungi and bacteria. For dehydrated food, the so-called Tyvek<sup>®</sup> sachet, manufactured by Dupont Chemicals, contains desiccants (e.g., silica gel, calcium oxide, natural clay) and is characterised by excellent mechanical strength [120].

#### 3.4.2. Active Compounds from Plant Extracts as Additives for Edible Coatings

Increasing research interests in plant extracts aim to develop new edible coatings for food preservation. For this reason, in recent years, active packaging containing natural biopolymers and plant extracts that can effectively delay the oxidation and microbial spoilage of food has been developed [127]. Antioxidants and antimicrobial compounds that occur naturally in some plants play a significant role in maintaining food quality [128]. In the case of fruits, the main causes of postharvest diseases are bacterial and fungal infections,

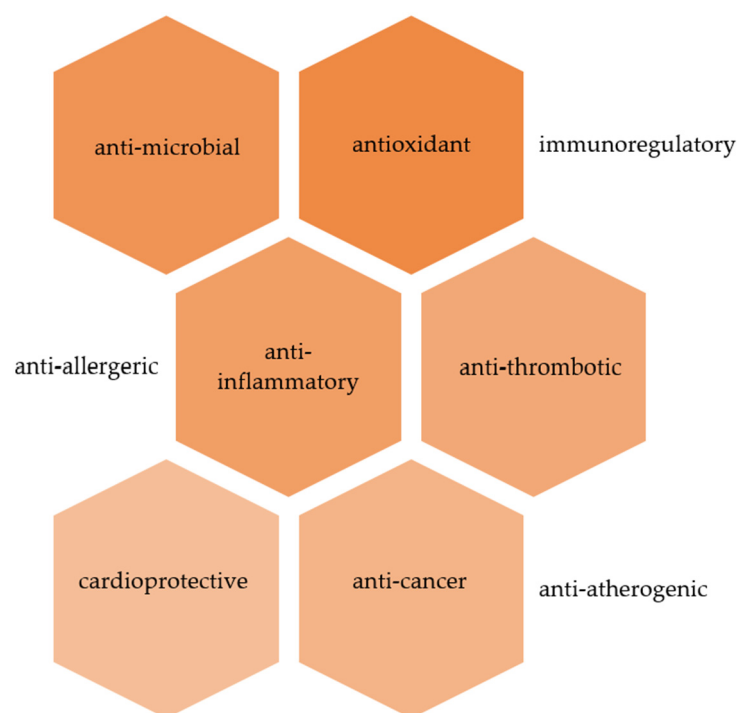
as well as damage. The developed strategy of covering the fruit with edible coatings to prevent and reduce postharvest diseases combines active additives such as natural extracts, biological inhibitors, or antimicrobial chemicals [75]. The conducted research shows that the use of pomegranate peel extract for edible coatings made based on alginate and chitosan reduced the development of *Colletotrichum gloeosporioides* strains and also inhibited the growth of *Penicillium digitatum* on oranges covered with a coating made of chitosan, locust bean gum, and pomegranate peel extract [129]. On the other hand, the moringa leaf extract present in coatings containing carboxymethylcellulose reduced the rate of ripening and inhibited the growth of postharvest avocado pathogens [130]. Synowiec et al. [131], covering apples of the “Jonagored” variety with a pullulan and a pullulan coating containing basil extract, noticed that the coating had an impact on visual factors: apples covered with a coating with the addition of basil extract were dull and had a noticeable basil aftertaste. In addition, the pullulan coating with basil extract showed good antifungal protection against *Rhizopus arrhizus*. In addition, it was observed that the weight loss of apples was significantly lower compared to uncoated apples after 30 days of storage at refrigerated temperature. Fruits and vegetables coated with propolis additives retain the appropriate level of vitamin C or flavonoids during storage, contributing to higher antioxidant activity of fruits and vegetables compared to uncoated ones. In addition, relatively higher peroxidase activity is maintained in postharvest fresh produce [132]. Significant antioxidant and antimicrobial properties were also observed when using phenols as functional compounds in edible coatings, showing the important role played by plant extracts in extending the shelf life of food, as well as maintaining the appropriate level of nutrients in many fruits and vegetables [133].

The last few years have also seen an increasing trend of adding essential oils as active compounds in edible coatings, which are secondary metabolites produced by plants, to edible coatings, which have been applied to, for example, fresh fruits and vegetables. Essential oils are an alternative to chemical preservatives and are described as effective methods to preserve the postharvest quality of fruits and vegetables, but they modify their taste, which is often not preferred [134]. However, mixing essential oils with other substances or using multi-layers may reduce unfavourable odours and create active coatings. Jamróz et al. [135] created an active and intelligent triple-layered edible packaging material based on the combination of furcellaran, chitosan, gelatin hydrolysates, curcumin, and lemon-grass essential oil that can be used as indicators of food product freshness. Plant extracts, which are pure compounds, as well as standardised extracts, affect the control of microbial growth [136].

### 3.4.3. Phenolic Acids as Active Compounds

Phenolic compounds are powerful antioxidants that can “scavenge” and recombine radicals and chelate metal ions. They act as electron transfers, resulting in stable products. For this reason, plant extracts containing phenolic compounds are increasingly used in developing active food packaging, and recent studies have shown the effectiveness of incorporating such extracts into products. Phenolic acids are found in a variety of plant sources such as fruits, vegetables, spices, cereals, and beverages. A promising source of phenolic compounds is also extracted from grain milling, which are by-products, e.g., bran and husks. Essential oils, mainly used as flavouring agents, include phenolic and terpenoid compounds with powerful antioxidant properties. The research shows that the films containing tannic acid had a more rigid structure and were more durable, and the addition of gallic acid affects, among others, the flexibility of the film because it acts as a plasticiser [137]. Phenolic acids, belonging to phenols, contain hydroxyl and carboxyl groups in their structure and are distinguished by their bioactive properties. Other proper-

ties of phenolic acids are shown in Figure 2. One of the classes of secondary metabolites includes phenolic compounds that are involved in the basic biochemical processes of the plants from which they are obtained, affecting the regulation of growth, development, and reproduction. Ferulic acid, a derivative of cinnamic acid, is obtained mainly from the leaves, seeds, and bark of coniferous trees, but another source confirming the high content of phenolic acids is the outer layer of cereal grains. It was observed that the differences in the antioxidant activity of phenolic acids, such as ferulic acid, caffeic acid, or p-coumaric acid, result from the number of hydroxyl groups present in the molecule. A higher level of antioxidant activity occurs when they are esterified [138]. A frequent phenomenon is the division of phenolic acids, classifying them as two subgroups: an acid based on the C<sub>6</sub>-C<sub>1</sub> structure, which includes acids such as pro catechin, gallic, and syringic, and also as aromatic compounds with a 3-carbon side chain (C<sub>6</sub>-C<sub>3</sub>), which includes coumaric, caffeic, ferulic, and sinapic acids [139]. De León-Zapata et al. [140] developed nanocoatings based on candelilla wax and tarbush (*Flourensia cernua*) extract to delay the ripening of “Fuji” apples during 56 days of storage. After developing appropriate values of wettability of the nano-coating on the surface of apples and covering the fruit with them, the physicochemical changes, water vapour permeability, and microbiological stability of apples were evaluated. It was observed that the coating reduced the loss of firmness and reduced the occurrence of physicochemical and microbiological changes (decreased growth rate of fungi and yeast) compared to apples not subjected to coating.



**Figure 2.** Properties of phenolic acids.

Phenolic acids can be extracted from fresh, frozen, or dried plant biomass. However, plant biomass is usually air-dried or lyophilised before phenolic acid extraction and then ground into a uniform powder. The extraction method from dry biomass is more advantageous because enzymes found in fresh cells can cause degradation of phenolic acids. Scientific reports indicate that the lyophilisation process allows for the preservation of a higher level of phenolic content in plant biomass compared to air-drying. The extraction process is influenced by many factors, including the chemical properties of the acid, polarity, concentration levels in plant tissue and the desired level of purity of the extract. The first step of the process is the crude extraction of phenolic compounds using 20–50%

ethanol, methanol, acetone, or a combination of these solvents. The next important step is alkaline hydrolysis using sodium hydroxide (1–4 M), the main purpose of which is to release insoluble phenolic acids into their soluble forms [141].

Organic phenolic acids, characterised by the presence of phenolic rings, arouse great interest among researchers due to their antioxidant and antibacterial properties. It has been proven that adding ferulic acid or gallic acid makes it possible to achieve a beneficial effect on the water vapour permeability of edible films and improve mechanical strength, antioxidant, and antibacterial activity. When food is exposed to light, oxidative degradation accelerates, which leads to loss of nutrients, discolouration, and a change in taste. Liu et al. [142] observed that chitosan films with the addition of phenolic acid used for coating shrimps had higher opacity than films without the addition of acid, i.e., they were characterised by a greater ability to absorb light. This is probably due to aromatic groups found in phenolic acids. In addition, the addition of phenolic acid caused the obtained films to have a lower moisture content, and the lowest value was obtained in the case of chitosan films with the addition of gallic acid, which could be caused by the presence of three phenolic hydroxyl groups present in this acid. Low moisture content affecting the solubility of the film in water is an important factor determining the biodegradability of the films used as food packaging materials. Current research development is also focused on combining ferulic acid and edible coatings to improve food performance. The distinguishing feature of ferulic acid is its strong antioxidant and antimicrobial effect, increasing the durability and safety of food. The study proved the positive effect of the combination of sodium alginate and ferulic acid in apple coating on reducing the *Listeria monocytogenes* population after 7 days of storage. In addition, no effect was observed on the fungal species *Saccharomyces cerevisiae* [143].

Caffeic acid, synthesised by plants as a secondary metabolite, as well as its natural derivatives (mainly esters), are referred to as strong antioxidants because they can protect cells from oxidative stress even in low concentrations. Oxidative changes can occur in both fats and proteins, affecting the quality of many food products. In food production, synthetic antioxidants are increasingly replaced by natural compounds, and phenolic acids are increasingly attracting interest. Caffeic acid is found in many foods, such as legumes, tomatoes, nuts, cereal grains, sunflower seeds, and various types of fruit [144]. Caffeic acid was used to produce active composite films consisting of N, O-carboxymethyl chitosan and methyl cellulose in various concentrations. During the agar test, the effect of the presence of caffeic acid on the occurrence of pathogenic bacteria was examined. It was proved that the inclusion of acid in composite films resulted in a 6-fold increase in the effectiveness of reducing the development of *Escherichia coli* strains, from 16.32% for the control film to 97.73% for the film containing caffeic acid. In addition, antimicrobial efficacy was also doubled when *Staphylococcus aureus* growth was reduced from 49.41 to 94.59%. These results show that the addition of caffeic acid to composite films has a positive effect on reducing the occurrence of food-borne pathogens [126]. The presence of caffeic and ferulic acids enables the formation of cross-linking bonds. Moreover, ferulic acid can react with some amino acids present in proteins, e.g., tyrosine, lysine, and cysteine, resulting in the formation of a cross-link. The research shows that caffeic acid is more effective than ferulic acid and, therefore, can increase the safety of biodegradable packaging by improving its barrier and physicochemical properties [145]. Sun et al. [146] studied the properties of chitosan films by adding various concentrations of gallic acid. The authors observed a beneficial antimicrobial effect against *Escherichia coli*, *Salmonella typhimurium*, *Listeria innocua*, and *Bacillus subtilis*. Literature reports also show that gallic acid has valuable antibacterial properties against pathogenic *Vibrio* species, indicating its great potential for creating packaging materials that limit the development of pathogens and bacteria

responsible for food spoilage. In addition, the content of gallic acid in foil-forming materials increases their flexibility, acting as a plasticiser, thus eliminating their brittleness. Also, Fang et al. [147] observed that the antimicrobial activity of the chitosan coating increased with the inclusion of gallic acid. On the other hand, films based on chitosan and syringic acid are characterised by higher density, water solubility, and opacity and have properties that limit the development of various microorganisms. In addition, syringic acid has antioxidant properties, and its addition can extend the shelf life of food by being an active packaging material [148]. Liu et al. [149] conducted research on the properties of chitosan films with the addition of protocatechuic acid, which was characterised by, among others, increased thickness, lower light, and water permeability compared to chitosan films without the addition of protocatechuic acid. Protocatechuic acid is characterised by strong antioxidant properties and is most often found in fruits and vegetables. It has been proven that phenolic compounds can interact with chitosan chains due to ester bonds, electrostatic interactions, and hydrogen bonds, resulting in increased mechanical strength of the film. Hydrogels are also used in active packaging, but this is only the initial phase, requiring further research to improve the cross-linking properties of the hydrogel. In addition, the challenge for researchers and scientists has also become the development of biodegradable packaging materials containing antimicrobial agents, the release of which will be controlled to a greater extent. For this purpose, studies have already been conducted on the release of gallic acid after 15 min, 1.5 h, and 16 h with films containing green tea extract. A slight increase in the release of phenolic compounds was observed in the approximate amount of 84 mg during 1.5–16 h [79].

Phenolic acids may be an interesting group of compounds for the development of functional foods. However, there are still some gaps that should be better explored to increase the possibilities of using these compounds on a large industrial scale and to make their use as effective and efficient as possible. The goal is also to provide the benefits of phenolic acids without causing side effects. There is still a need to investigate methods to increase the stability and preservation of the bioactivity of these compounds during the shelf life of food products.

#### 3.4.4. Nanoparticles and Nanomaterials as Active Compounds

The growing demand is due to consumers' concerns about food safety and quality, and the so-called "ecological films" involve using nanotechnology to improve their properties. In contrast, nanotechnology in food coatings plays an important role in developing a new generation of coatings with improved properties. Biodegradable and environmentally friendly biopolymers have gained enormous potential in food packaging, which, to improve barrier and mechanical properties, are used in combination with nanoparticles, defined as particles of 1 to 100 nm. In active packaging, bioactive agents (antimicrobial agents, enzymes, antioxidants) are used as components of polymer-based packaging that destroy microbes (bacteria, fungi, protozoa) or inhibit their penetration. For this reason, nanocomposites play a key role due to their antimicrobial properties [150]. The migration of nanometres from nanocomposites depends on many factors, including size, concentration, solubility, diffusion, type of food, pH, or interaction time. Some studies confirm the lack of migration of nanomaterials to food products after appropriate encapsulation of the polymer used. The main advantages of nanomaterials include high absorption and bioavailability, dispersion, solubility, and improved stability during food processing, improving efficiency during production, and reducing waste and financial burden [151]. The use of nanoparticles in packaging aims to improve the mechanical, thermal (stability), and barrier properties by introducing them into a polymer matrix. In addition, the packaging material influences the conditions inside the package. A significant role is played by the interaction of the material

from which the packaging is made—with the food product or with the atmosphere in the packaging, affecting the quality of the food. The ability to monitor changes in packaging conditions should also be considered. The most commonly used nanoparticles in food packaging are nanoparticles of metals or their oxides, such as silver, gold, copper, zinc, or magnesium [152,153]. Lan et al. [154] studied films containing red apple pomace and TiO<sub>2</sub> nanoparticles. The research that was conducted shows that the addition of nanoparticles significantly improved the water vapour barrier properties, mechanical properties, and thermal stability of the film. The film's excellent antioxidant and antibacterial properties were also observed, which may be a potential strategy for transforming apple processing waste into active packaging materials.

An important aspect is the legal regulations regarding food packaging materials, which may be accepted in one country and not in another. These discrepancies result from the classification of edible films and coatings as food ingredients, food products, substances that come into direct contact with food, as well as food additives or packaging substances, by the provisions of American and European directives. Edible coatings, defined as an integral ingredient of food, must meet several food standards, including should not alter the taste, smell, colour, or texture of the food, must be made by manufacturing practice, and must not hurt the composition of the food or pose a risk to human health. However, Regulation (EC) No 1935/2004 does not specify the safety assessment of nanoparticles in packaged food and the biological effects [64]. The addition of functional compounds to film-forming solutions, e.g., phenolic compounds, is recognised as safe (GRAS) and can be practiced to improve the functionality of edible coatings and films by the FDA Regulation 2019 [3]. In addition, guidelines and protocols for risk assessment in the aspect of nanotechnology are developed by the International Organisation for Standardisation (ISO), and the characterisation and identification of nanomaterials have been included by the European Food Safety Authority [155].

The greatest development of nanotechnology occurs in China, Japan, the European Union, and the United States, where the National Nanotechnology Initiative allocates USD 3.7 billion for a 4-year investment plan for research in the field of nanotechnology [151]. Due to the difficult specificity and complex structure of nanomaterials, the REFINE project of the Horizon 2020 program was created, which included information on regulatory challenges and the development of appropriate tools. The surveys and interviews showed that the current regulatory guidelines are difficult to access and not always understandable [156]. Recently, nanotechnology has offered the food industry many new approaches to improve food quality, durability, and safety. Unfortunately, there are concerns from consumers, regulatory agencies, and the food industry regarding the potential for side effects from using modified nanoparticles in food, e.g., through leakage from packaging material. There are reports that many of the nanoparticles used do not hurt human health, but there are also reports of their harmful effects, causing damage to cells or organs, as well as damage to microbial cells that inhabit the human digestive tract [157]. However, according to de Oliveira Filho et al. [158], the large specific surface of the nanoemulsion causes rapid digestion by enzymes from the gastrointestinal tract, limiting the occurrence of a possible toxic effect that may occur due to the accumulation in organ cells.

Several papers report that zinc oxide (ZnO) is not toxic to human cells and thus could be used as an antibacterial agent in food packaging. Nanomaterials are characterised by a high ratio of the specific surface area of nanoparticles to their volume, attributed to their different antibacterial mechanisms [159]. In addition to the huge benefits, nanotechnology also carries some risks that must be considered in everyday use. According to Ashfaq et al. [114], this ratio may have an adverse effect and cause a potential threat to human health due to the emerging dissimilarity of nanoparticle forms from the original forms.

Differences in the toxicity of nanoparticles may vary depending on their type, concentration, exposure time, and sensitivity to given conditions. A migration test should be performed to determine the risk of ingredients from any new food packaging material, which means “determining the release of substances from a material or article either into food or into food simulant”. In addition, it is a promising film-forming material approved by the US Food and Drug Administration (FDA). Coatings used on the surface of fruit and vegetables containing silver nanoparticles have an antimicrobial effect, the permissible level of which is 0.06 mg/L, conditioned by EU regulations. Unfortunately, consumers react reluctantly to emerging innovations, treating them as a threat to their health and the environment. The main reason for this is assumed to be legislative barriers and emerging uncertainty regarding the effectiveness of applying such systems, taking into account economic and environmental aspects. At present, the legislation is still in its early stages, and only five Member States of the European Commission (EU) have adopted legislation on nanomaterials or goods containing nanomaterials, i.e., Belgium, Denmark, France, Switzerland, and Sweden [160]. Although nanotechnology has been developed primarily for food packaging, it has not yet found widespread use. This was mainly due to too few reports on the uncertainty of the impact on human health and the environment. In addition, detecting nanomaterials in food products when they are released or react with food content is very complicated [46].

#### 4. The Role of Edible Coatings in the Enhancement of the Postharvest Quality of Apples

The basic idea about the edible active coating on the surface of apples is presented in Figure 3, which indicates that the main purpose is to maintain moisture inside the fruit while letting it respire. Microbial contamination present in minimally processed food products can arise at various stages of production, e.g., in the orchard, at harvest, storage, processing, or distribution. In addition, improper storage temperature, too high humidity, and mechanical damage also have an impact [161]. One of the basic conditions for the proper preservation of fresh fruit immediately after harvesting is the use of reduced storage temperature throughout the cold chain, which reduces the appearance of the two most common moulds caused by *Penicillium expansum* and *Botrytis cinerea*. Chemical fungicides have been used to reduce postharvest spoilage of fresh fruit and vegetables, but their use has been restricted due to emerging consumer concerns and their adverse health and environmental impacts. Currently, edible coatings and films containing the addition of natural antimicrobial compounds are of great interest [162].

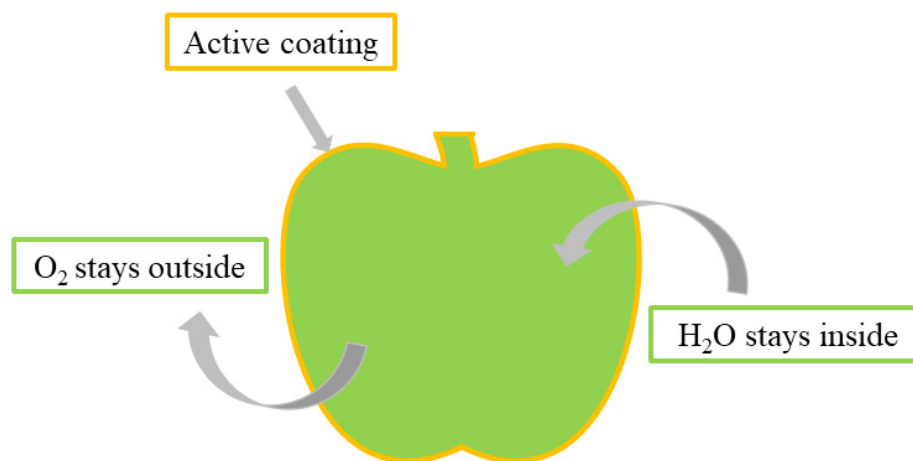


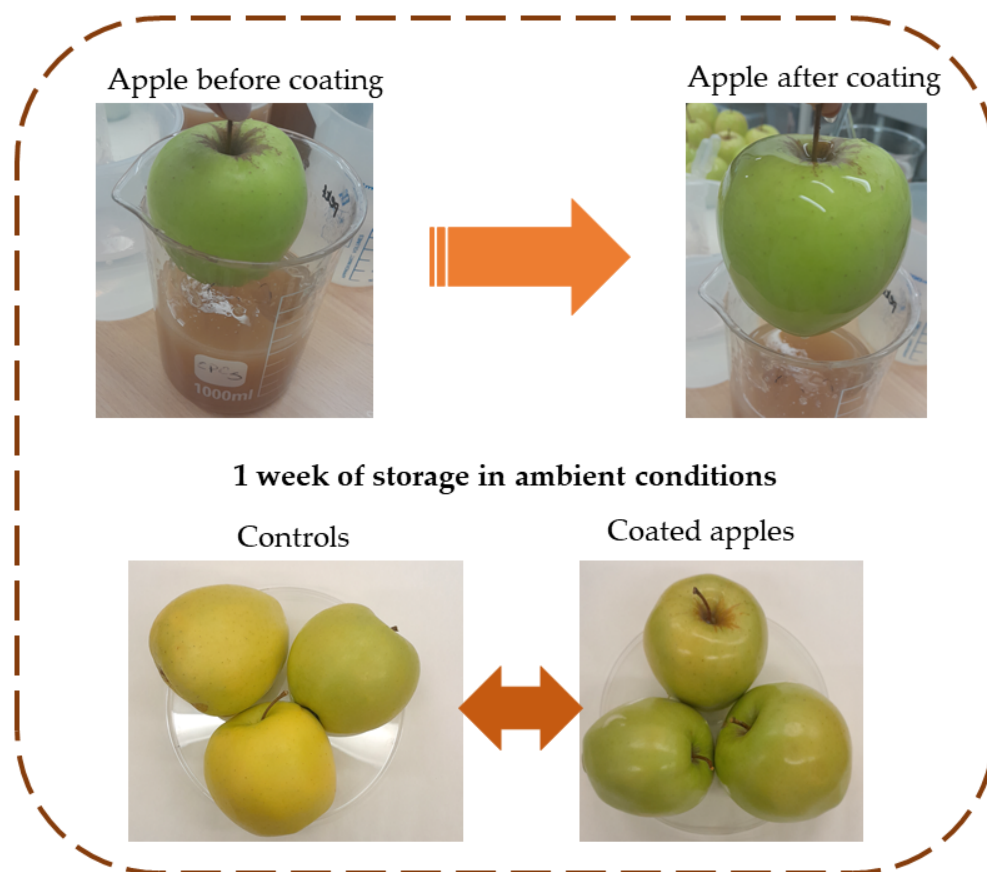
Figure 3. The mechanism of active coating for apples.

The results of the study showed that the mixture of *Bifidobacterium lactis* and *Lactobacillus rhamnosus*, which was on the surface of apples with an alginate coating, sufficiently reaches the human small intestine, exerting a beneficial effect on health. Fresh apples retained their microbiological and nutritional properties, making the alginate-based solution a good probiotic carrier, which is important in the aspect of developing new functional food products [163]. Also, according to Hashemi et al. [164], adding probiotics to edible films, in addition to ensuring their good stability, reduces the growth of microorganisms, and thus food spoilage. Thanks to the consumption of fresh fruit covered with a probiotic film, it becomes possible to supply beneficial bacteria to the human body, which are protected by the cellulose contained in the fruit. Recent studies show that probiotic bacteria contained in edible membranes help maintain physicochemical parameters and nutritional and sensory factors during storage. The use of an edible coating based on pullulan and basil on the “Jonagored” apples resulted in antifungal activity against *Rhizopus arrhizus*, while edible films based on potassium sorbate, pullulan, and gelatin showed effectiveness in terms of antibacterial properties against *E. coli*, *K. apiculata*, and *B. cinerea* [100].

To prevent contamination and enhance the visual appeal by adding a gloss, the coating is increasingly used before marketing fruit. Composite edible coatings are most effective because single ingredients are ineffective enough to improve fruit shelf life and quality (Table 1). The most commonly used additives that provide additional gloss to fresh apples are shellac wax and carnauba wax, which primarily prevent weight loss. These substances, alone or as components of an emulsion, simultaneously control water loss and thus preserve the quality of fresh fruit and delay their ripening, which can be observed visually. The example of uncoated and coated apples with apple pectin-based edible coatings before and after 1 week of storage are presented in Figure 4. Consumers judge fresh fruit by changes in the surface of the skin, firmness, and greasiness. Failure to meet these standards by producers reduces the acceptance of the product by the consumer or its complete rejection and thus reduces the financial return [165]. In a study on the use of zinc nanoparticles for coating climacteric fruits, it was proven that they effectively delay ripening, reduce weight and moisture loss, and preserve fruit firmness. Due to its excellent mechanical, barrier, and biocompatibility properties, zinc oxide has gained a lot of interest in the scientific community [160]. Susilowati, Mahardiani and Sulistyowati [166] coated apples with films made based on Suweg starch, with a high content of amylose (approx. 24.5%). The water vapour barrier of the obtained starch films was insufficient for their use. WVP was determined gravimetrically by placing the foil on an acrylic plate containing a moisture absorber, which was silica gel. The foil was clamped with a ring with four screws and placed in a desiccator with salt at a relative humidity of 75% and a temperature of 28 °C. Measurements were taken every hour for 8 h. It is necessary to add a hydrophobic biopolymer that allows low water permeability values to be achieved. Such a biopolymer that improves the properties of the film is chitosan, which, thanks to the strengthening of hydrogen bonds, also improves the mechanical properties of the resulting films, which become flexible and difficult to break. Coatings with better migration properties and reduction of transpiration losses from the surface of coated apples were observed by Bisen [167]. The fruits were coated with different concentrations of  $\text{CaCl}_2$ ,  $\text{CaNO}_3$ , aloe, sago, and arrowroot gel solutions and then dried at room temperature. After a nine-day observation, it was found that the weight loss of apples during storage was limited. Apples (*Syzygium samarangense* cv. Citra) with a relatively short shelf life and susceptible to spoilage microorganisms (*Klebsiella penumoniae*, *Penicillium purpurogenum*) were coated with an edible coating based on alginate with the addition of vanilla essential oils and the addition of green grass. The action of vanilla essential oils affects less membrane damage compared to the phenolic compound carvacrol, and it has also been confirmed that these oils have a bactericidal effect.

**Table 1.** Examples of the use of edible coatings in improving the postharvest quality of apples, including active compounds, storage conditions, and the effects achieved.

Apples or Variety	Coating or Biopolymer	Active Compound	The Storage Conditions	Effect	References
<i>Malus domestica</i> cv. 'Jonagored'	pullulan	extract <i>Satureja hortensis</i> L. (SH)	16 °C, 14 days and 2 °C, 28 days.	the reduction of weight loss; extending storage time; reduced wilting and wrinkling of the surface	[168]
<i>Malus domestica</i> cv. 'Jonagored'	pullulan	extract <i>Bergenia crassifolia</i> (LBL)	2 °C, RH 70% for 28 days, and 16 °C, RH 60 for 14 days.	antimicrobial effect; preserving the hardness of apples; extending storage time; the reduction of weight loss	[169]
<i>Malus domestica</i> cv. 'Jonagored'	pullulan	sweet basil ( <i>Ocimum basilicum</i> L.)	16 °C, RH 55–60% for 14 days, and at 2 °C, RH 70–75% for 30 days in refrigerators	extending the shelf life of apples; low protection against mesophilic bacteria, but good antifungal properties against <i>R. arrhizus</i> ; reduction of colour changes; reduced weight loss during storage	[131]
Not indicated	xanthan gum	zinc oxide (ZnO)	room temperature, 13 days	maintained overall quality of apples; protection against microbial contamination	[170]
Apple fruit variety Cripps Pink	starch, $\iota$ -carrageenan	sucrose ester	20 °C and 5 °C, then 1 day at 20 °C	reduced weight loss, firmness, and skin colour	[165]
Custard apple ( <i>Annona squamosa</i> )	chitosan	ascorbic acid	15 ± 1 °C, 12 days	delay in the degradation of post-harvest quality; the composite coating delayed the reduction of hardness, appearance, and loss of moisture	[171]
Apple ber ( <i>Ziziphus mauritiana</i> L.)	chitosan, guar gum	aloe vera gel, garlic extract	ambient temperature	extended the shelf life of the fruit compared to the control sample (uncoated)	[172]
Apples Golden Delicious	arabic gum, candidella wax, and jojoba oil	a fermented extract of tarbush	27 °C, 8 weeks	maintaining water activity; improvement of firmness, quality, and durability	[173]
Malang's Apples	starch-chitosan	essential oil of sweet orange peel	22 days	improvement of colour, smell, and texture of fruit; the essential oil had the maximum effect on antibacterial activity	[166]
Apples Kala kulu	guar	fenugreek, flaxseed	25 °C, 35 days	reduction of microbial contamination while preserving the quality attributes of fruits without significantly altering their nutritional value	[11]



**Figure 4.** The example of uncoated and coated apples with apple pectin-based edible coatings before and after 1 week of storage (The own study).

A slowed rate of respiration was also observed, and apples treated with coatings with the highest concentrations of essential oils were characterised by the most favourable sensory and visual characteristics [174]. Also, Duong et al. [18] researched the use of edible films made based on sodium alginate, covering apples of the species ‘Tabtimchan’ (*Syzygium samarangense*), which contain numerous biologically active compounds (antioxidants), having a positive effect on human health. Many hypotheses indicate that the reduction of damage to fruits sensitive to storage at temperatures below 10–12 °C is influenced by the strengthening of antioxidant systems or maintaining cell integrity, which allows for achieving lower phospholipase and lipoxygenase activity. Moreover, water-soluble alginate films can become insoluble material by cross-linking with divalent or polyvalent cations, e.g.,  $\text{Ca}^{2+}$ . On the other hand, adding cross-linking agents such as calcium chloride ( $\text{CaCl}_2$ ) improves the mechanical properties of edible alginate films and reduces the penetration of water vapour and oxygen.  $\text{CaCl}_2$  is also a commonly used solution to achieve fruit firmness in fruit processing. Research shows that a sodium alginate-based coating cross-linked with  $\text{Ca}^{2+}$  is a promising alternative to chemical treatments, reducing the occurrence of brown spots and fruit shrinkage. The use of  $\text{CaCl}_2$  by forming bonds with  $\text{Ca}^{2+}$  may reduce the activity of enzymes responsible for softening and respiration, such as polygalacturonase [175]. The effectiveness of preserving bioactive ingredients, including vitamin C, and the total content of flavonoids and anthocyanins in stored apples was obtained by coating the fruit with the addition of aloe vera gel. Edible coatings in various concentrations (50, 75, and 100 v/v%) were applied to waxed apples and then stored for 12 days at 10 °C. The limited oxidation of ascorbic acid could be due to the low permeability of the aloe-based gel coatings, and thus the enzyme activity was reduced. There was also a tendency to reduce the weight loss of apples and to delay the increasing titratable acidity, which suggests a

delay in the deterioration of the quality of stored fruit. The moisture loss of apples could be due to the hygroscopic properties of the aloe vera gel, which probably formed a barrier between the fruit and the environment, thus preventing moisture from entering [176].

Apples (*Anacardium occidentale* L.) were subjected to four treatments: hydro-cooling at 5 °C, hydro-cooling at 5 °C combined with the application of calcium chloride, coated with solutions obtained based on corn starch, and coated based on corn starch with the addition of calcium, were stored in refrigerated conditions for 25 days at 5 °C. Hydrocooling is a postharvest treatment to reduce residues and microbes by spraying or dipping in cold water. It was observed that hydrocooling delayed weight loss, reduced peroxidase activity, and reduced the decomposition of vitamin C, dye, carotenoids, and anthocyanins. As a consequence, hydrocooling had a beneficial effect on limiting unfavourable visual changes. On the other hand, adding calcium affected the preservation of apple firmness and lower activity of pectin methylesterase and polyphenol oxidase. In the research conducted by Belay et al. [177], it was proved that coating “Granny Smith” apples with coating solutions containing zein or zein in combination with nisin delayed their weight loss compared to uncoated apples. After 21 days of fruit storage at 15 °C, a 30% loss in the initial weight of uncoated apples was noted, a 25% loss in weight of apples coated with zein and nisin, and a 17% loss in weight occurred in the case of fruits covered with zein. The decrease in apple mass loss could be due to the ability of the coating to constitute a modified water vapour barrier by blocking the pores present in the skin. The highest retention of apple firmness during storage was found in the case of samples coated with zein, which could be caused by the limitation of the metabolic activity of enzymes affecting the decomposition of the cell wall—pectinesterase and polygalacturonase. The appropriate colour of all coated and uncoated fruits was also preserved under conditions acceptable to consumers without visual changes in colour and spoilage. In addition, a delay in the growth of microorganisms on the surface of apples was achieved for the coated samples compared to the control samples. Reduced weight loss and preservation of firmness of “Fuji” apples during storage were also observed in the study conducted by Chen et al. [178]. Fruits were coated with a combination of nanoemulsion coating (NC-1) and 1-methylcyclopropene (1-MCP). The samples were then stored for 280 days at 4 °C and inspected every 35 days. “Tsugaru”, “Summer King”, and “Shinano Gold” apples were coated with an edible coating consisting of sucrose monoesters of fatty acids and ethanol. The authors observed a weaker coating adhesion on Shinano Gold apples after 28 days of storage at room temperature, probably due to the presence of porosity and a different composition of the fruit skin. In addition, an increase in the effect of greasiness and thickness of the skin was observed, which was influenced by the accumulation of cuticular waxes. However, in the case of “Tsugaru” and “Summer King” apples, the appropriate firmness and colour of the fruit were preserved during storage [179].

## 5. Challenges and Future Perspectives

Edible coatings based on polysaccharides or proteins are distinguished by good mechanical strength, but they absorb moisture very quickly and easily. To improve these properties, scientists are developing the concept of hydrogels, which are introduced in biodegradable packaging systems and consist of a hydrophilic polymer that does not dissolve in water. An important aspect is optimising the release rate of active compounds through modifications, using different combinations of biopolymers, or using a binder. In addition, in the development of biodegradable packaging, agricultural waste gains more and more potential [79].

Numerous studies have been conducted on the impact of the use of edible coatings on maintaining the quality and extending the shelf life of fresh fruit, but efforts are still

being made to acquire and introduce new natural coating materials. There is a need for cheap and available film-forming materials, also from waste sources. At the same time, progress in the field of coating methods and the acquisition of new coating materials leads to continuous improvement of their effectiveness without any negative impact on human health. The discovery of the combination of edible coatings and irradiation revealed significant importance in extending the shelf life of fresh fruit, as it maintains the overall quality of postharvest fruit and vegetables but creates distrust among consumers regarding food safety. The postharvest technologies, supplementing the MAP effect, also include the use of silk fibroin coatings, a structural protein comparable to collagen, which has a positive effect on extending the shelf life of the fruit and is environmentally friendly. Unfortunately, due to the risk of residual amounts of metal elements and their level of toxicity, the commercial value of the fruit is reduced and may not be suitable for human consumption [180]. For this reason, further research in this direction is expected. In addition, for economic reasons, innovative coating techniques are still unprofitable at the industrial level, which is why exporters and fruit retailers rarely use innovative methods, sticking to traditional methods. It is still expected to research achieving an extended shelf life of fresh fruit, without interfering with their sensory and nutritional properties, as well as further development of research at the initial stage of development and their application on a larger industrial scale.

## 6. Conclusions

Edible coatings applied to fresh apples may extend their shelf life, and the use of appropriate components together with them may also have antimicrobial or MAP-enhancing effects. The use of coating is in line with the assumptions of a mild technology that does not require specialised equipment but is still not used on a large scale. Phenolic compounds have antioxidant and antimicrobial properties, and by introducing them into the biopolymer matrix, they can act as an active ingredient whose gradual release affects the product, improving its quality. The use of film-forming solutions on the surface of climacteric fruit, i.e., ripening after harvest, has an impact on slowing down the fruit ripening process and thus reducing their waste. For this reason, food coating complies with the circular economy approach, which assumes, among other things, that the activities carried out in the agri-food sector are to reduce food waste, thus affecting the protection of the natural environment and preventing the depletion of natural resources. The requirements for protective coatings for apples should be focused on many aspects. First of all, low-cost materials are desired, including waste-based resources such as pectin from fruit or vegetable pomace or agricultural by-products. Good miscibility between the components, biopolymers, solvents, plasticisers, additives, or other functional substances is necessary to obtain a smooth and continuous structure. Thus, their compatibility is also important. When combined with the storage conditions, those factors may result in shelf life extension or retention of the postharvest quality of apples. The designed protective coatings should be effective regardless of the variety of cultivation techniques.

In conclusion, further research is needed to ensure the cost-effectiveness of edible coatings and to expand their application scope. Biopolymers have the potential to completely change the future of food packaging and preservation practices through further research and development. Despite the great benefits, there is still a problem related to the scale of application of edible coatings and the production costs. By using modern production systems, significant progress can be achieved in packaging and preservation for the food industry, and edible coatings can be adopted as the standard for environmentally friendly food packaging. Furthermore, research should focus on improving the performance of

biopolymers under different environmental conditions in order to maintain reliability throughout the supply chain.

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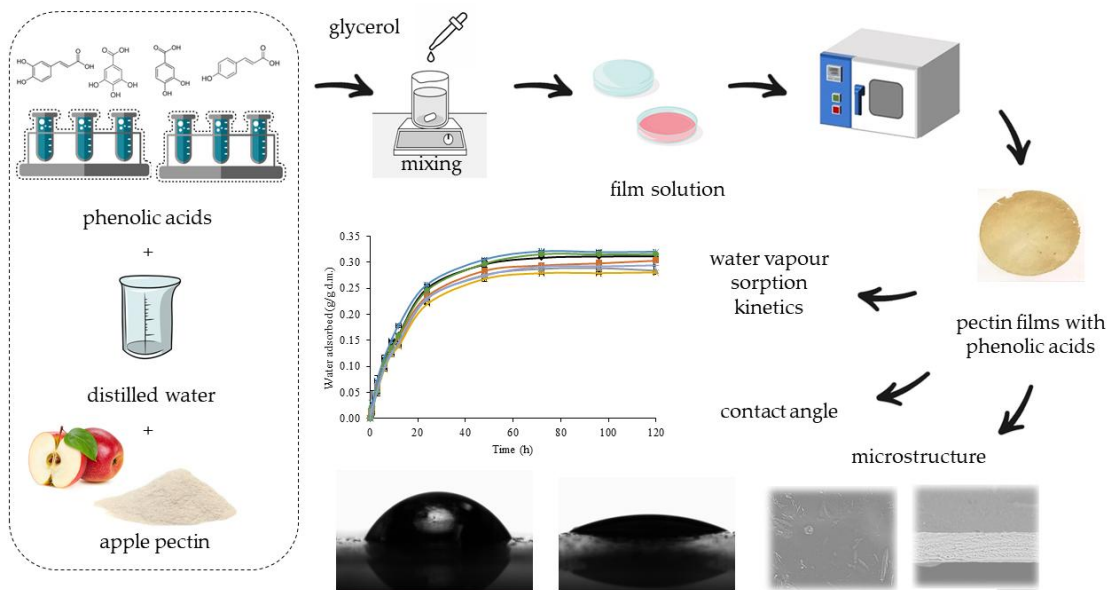
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## Publikacja 2

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### Abstrakt graficzny



# The Effect of Phenolic Acids on the Sorption and Wetting Properties of Apple Pectin-Based Packaging Films

Magdalena Mikus and Sabina Galus \*

Department of Food Engineering and Process Management, Institute of Food Sciences, Warsaw University of Life Sciences, 159c Nowoursynowska St., 02-776 Warsaw, Poland; magdalena\_mikus@sggw.edu.pl

\* Correspondence: sabina\_galus@sggw.edu.pl; Tel.: +48-22-593-75-79

**Abstract:** In this article, the effects of different phenolic acids, such as ferulic, gallic, caffeic, coumaric, protocatechuic, and sinapic, as active compounds on the sorption and wetting properties of apple pectin-based edible films were evaluated. The control pectin films and those with added phenolic acids differed in appearance and physical properties. The water content of the films was reduced and ranged from  $8.91 \pm 0.01\%$  to  $13.44 \pm 0.01\%$  for films containing phenolic acids compared to the control films ( $14.31 \pm 0.01\%$ ). The swelling index value of the films ranged from 86.63% for films with protocatechuic acid to 88.33% for films with the addition of caffeic acid. It was observed that the sorption isotherms had a similar shape for all the obtained films, while scanning electron microscopy (SEM) allowed for the observation of changes in the structure resulting from the film composition. It was shown that the lowest water contact angle values at the initial time (0 s) were observed for pectin films with ferulic acid ( $47.00^\circ \pm 4.47$ ) and the highest for the control films ( $58.44^\circ \pm 5.62$ ). After 60 s, the highest water contact angle value was recorded for the film with caffeic acid ( $66.39^\circ \pm 5.18$ ) and the lowest for the film with ferulic acid ( $14.72^\circ \pm 5.70$ ). Films containing gallic acid and protocatechuic acids showed the lowest water vapour permeability values among active films. The edible films developed in this study showed desirable features that could be used as bioactive packaging for food industry applications, both as protective edible coatings and active packaging films.

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**Keywords:** edible films; apple pectin; phenolic acids; sorption; contact angle

## 1. Introduction

Packaging is necessary to maintain the organoleptic, nutritional, and hygienic properties of food products. Due to the wide variety of packaging films, they can be classified as synthetic or biodegradable [1,2]. Nowadays, films made from plastics very often constitute an environmental problem [3]. Therefore, more and more often, research is being conducted to find alternative materials that will reduce the consumption of films made from non-renewable raw materials and replace synthetic polymers [4,5]. Their use in the food sector can reduce environmental problems related to accumulating non-renewable synthetic materials [6]. In addition, the use of edible coatings provides protection of the product against mechanical damage during transport, handling, and storage [7], thus ensuring an extension of shelf life, with no or minimal noticeable impact on the texture, taste, and nutritional value of the food [8]. This form of edible packaging is an edible coating, a thin layer of material placed on the surface of food products. The coating is a semi-permeable barrier to gases and affects the integrity of the food [9]. In

addition, the coating can be a carrier for bioactive compounds that act on the surface of food or in the human intestines, which include probiotics, antimicrobials, antivirals, and antioxidants. On the other hand, an edible film is a layer of material formed outside the food product, which is then applied to the product [10].

Edible films can be prepared based on proteins, polysaccharides, lipids, or a combination of these materials. Proteins and polysaccharides are characterised by their ability to create coatings with good oxygen barrier properties but are a poor moisture barrier [11]. In addition, films made based on polysaccharides are characterised by good permeability to carbon dioxide and have appropriate optical properties. They can be transparent and do not cause changes in the taste and smell of food [12].

Pectin is a water-soluble polysaccharide that consists mainly of linked esterified units of  $\alpha$ -D-galacturonic acid. It is characterised by a high molecular weight and the ability to form a firm gel, e.g., in the presence of calcium cations [13]. Pectin is one of the main components of plant cell walls and has different structures depending on the source [14].

Pectin can be extracted from food industry waste, particularly apple pomace, transforming what would otherwise be a throwaway by-product into valuable packaging materials. Furthermore, advances in pectin extraction and the use of state-of-the-art techniques such as supercritical water extraction, ultrasound, or microwave-assisted extraction are improving the quality and functionality of the resulting material, leading to new possibilities in food packaging. Due to its biodegradability, edibility, and ability to form films in combination with other materials, pectin is an attractive option for developing environmentally friendly alternatives to non-biodegradable plastics. Films based on pectin provide a barrier against oxygen, making them suitable for packaging fresh fruits. They are also characterised by their ability to limit respiration and extend the shelf life of perishable products [15]. Pectin-based films are also resistant to oils and fats, dissolve in water, and have excellent mechanical properties [16]. However, it should be noted that pectin films, without adding any other polymers, are characterised by high water permeability and high water solubility and have limited use in high-humidity atmospheres or the case of packaging food products with high moisture content [17]. Various modifications are used to improve these properties and increase the use of pectin-based films [18,19].

One of the modifications used is adding phenolic compounds to edible films, which provide antioxidant properties. This innovative approach, gaining popularity worldwide to obtain active packaging, can improve food products' oxidative state and antimicrobial properties. Another advantage of phenolic compounds is that they are natural and bioactive compounds found in various food products, e.g., fruits, vegetables, herbs, oils, and spices [20]. In addition, their occurrence has also been noted in agricultural and industrial by-products. Phenolic acids are defined as non-flavonoid polyphenolic substances, the characteristic feature of which is the connection of a carboxyl group with a benzene ring. They are a desirable group of compounds in the food industry because they inhibit harmful bacteria and fungi, including *E. coli*, *Bacillus cereus*, *Staphylococcus aureus*, or *Aspergillus flavus* [21].

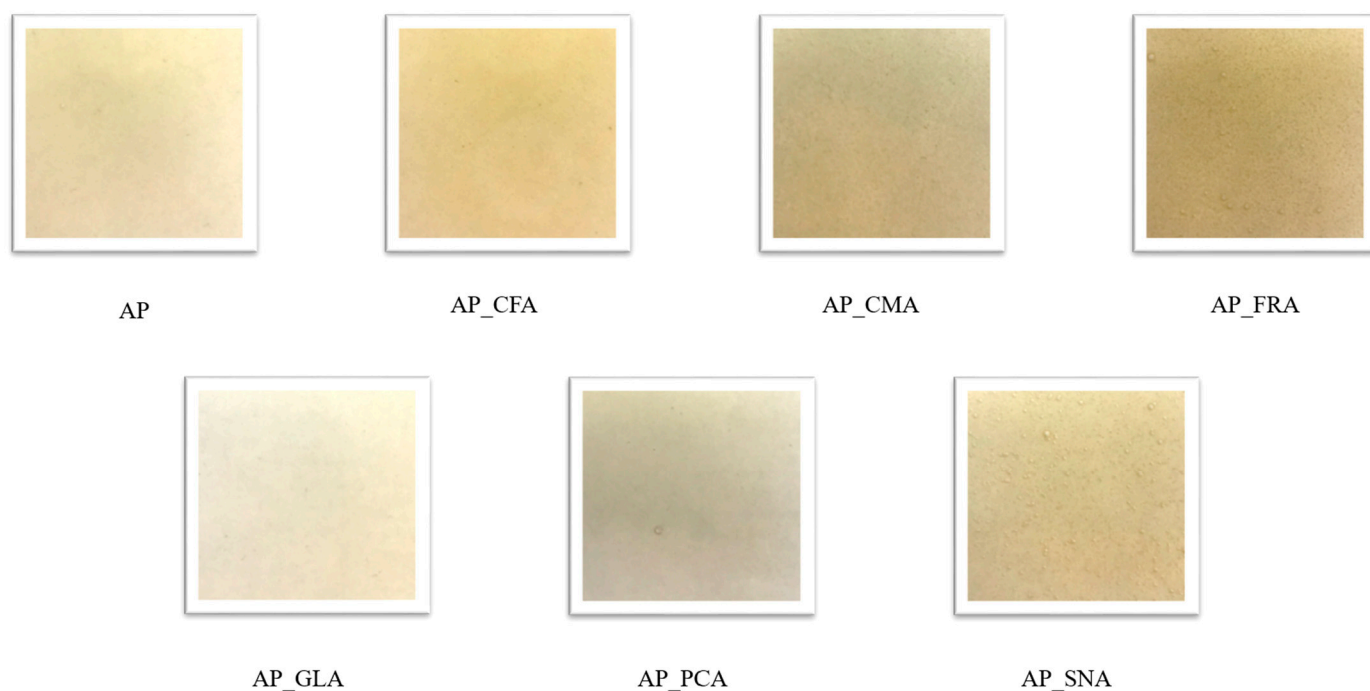
For example, phenolic acids, including ferulic acid, which is one of the most common acids in plants, are added to improve various properties. In addition to low toxicity and good cross-linking properties, ferulic acid has several other desirable properties, such as antioxidant, antimicrobial, anti-inflammatory, or antithrombotic effects. In addition, it is characterised by anticancer effects, protects against coronary artery disease, and increases sperm viability. It has been shown that in addition to ferulic acid, caffeic and gallic acids exhibit cross-linking effects [22]. Tannic acid also has excellent antioxidant, antimicrobial, and antiviral effects. In addition, it is considered safe in the food sector and prevents food spoilage and the development of pathogenic microorganisms [23].

The main objective of this study was to evaluate the sorption and wetting properties of active films based on apple pectin containing phenolic acids. In addition, it was determined which phenolic acid had the greatest impact on the properties of the films and their potential use as an edible coating or packaging film for food applications. The studies included thickness, water content, the swelling index, water solubility, water vapour sorption kinetics, water vapour sorption isotherms, water contact angle, the morphology of film measurements, and the water vapour permeability of active pectin films.

## 2. Results

### 2.1. Film Characterisation

An essential role in the context of considering edible films as packaging materials is played by their physicochemical properties. A smooth, uniform structure characterised the produced pectin films with the addition of selected phenolic acids. Photographs of the prepared films are presented in Figure 1. All the films obtained were transparent or slightly yellow to slightly brown. The addition of a plasticiser positively affected the obtained films by increasing their plasticity.



**Figure 1.** Photographs of the apple pectin (AP) edible films obtained with the addition of caffeic (CFA), coumaric (CMA), ferulic (FRA), gallic (GLA), protocatechuic (PCA), and sinapic (SNA) acids.

Thickness, moisture content, and the swelling index are important characteristics concerning using edible films as food packaging materials. Considering the thickness parameter, it was observed that the control films characterised the lowest value for this parameter. In contrast, the highest was characterised by the films adding coumaric acid. A similar relationship was observed by Yang et al. [24], who studied the thickness of films obtained from pectin and with the addition of tannic acid and  $\text{Fe}^{3+}$ . The thickness of the control pectin film was  $86 \mu\text{m}$ , while it increased with the addition of tannic acid and  $\text{Fe}^{3+}$ , reaching a maximum value of  $97 \mu\text{m}$ . The increase in the solid content and disruption of the original structure of the film probably caused this increase. According to Yerramathi et al. [25], the thickness of the film most often depends on the dynamics of film dispersion during preparation and drying, which later affects the achieved physical and mechanical

properties of edible composite films. The pectin films obtained by Liu et al. [26] reached a thickness of  $66 \pm 3.61 \mu\text{m}$ , while a thickness of  $68.67 \pm 2.40 \mu\text{m}$  characterised the films with the addition of gallic acid.

The water content indicates the water retention capacity of the produced film matrix. The results of the water content of the tested films are presented in Table 1. The values were in the range of 8.91–14.31%. These values are similar to other biopolymeric films obtained via casting [27–29] or electrospinning methods [30]. The highest water content was observed for the control film, while the lowest was obtained with the addition of sinapic acid. This could be attributed to the film structure and the formation of bonding in the film matrix that probably affected the higher rate of water evaporation when sinapic acid was added. Edible films containing high moisture content and water solubility are unsuitable packaging materials for food products requiring high water resistance. The water content of pectin films was reduced after adding phenolic acids due to many reasons, mostly due to differences in compatibility between acids and biopolymers, the acid distribution in the film matrix due to the various rates of solubility, film structure creation during storage (evaporation of water), and the film thickness.

**Table 1.** The water content, swelling index, and solubility in water for the apple pectin (AP) edible films obtained with the addition of caffeic (CFA), coumaric (CMA), ferulic (FRA), gallic (GLA), protocatechuic (PCA), and sinapic (SNA) acids.

Film Type	Thickness ( $\mu\text{m}$ )	Water Content (%)	Swelling Index (%)	Solubility in Water (%)
AP	$76.76 \pm 5.39^a$	$14.31 \pm 0.01^d$	100 <sup>ab</sup>	$77.39 \pm 8.00^a$
AP_CFA	$109.79 \pm 7.81^b$	$10.70 \pm 0.01^{ab}$	$88.33 \pm 1.06^{ab}$	$70.75 \pm 0.29^a$
AP_CMA	$129.49 \pm 7.37^c$	$11.36 \pm 0.01^{abc}$	100 <sup>ab</sup>	$70.22 \pm 0.39^a$
AP_FRA	$121.88 \pm 7.67^c$	$13.44 \pm 0.01^{cd}$	$88.18 \pm 0.71^a$	$79.11 \pm 7.92^a$
AP_GLA	$85.31 \pm 4.05^a$	$10.93 \pm 0.00^{abc}$	100 <sup>ab</sup>	$69.73 \pm 2.91^a$
AP_PCA	$78.26 \pm 5.10^a$	$12.87 \pm 0.01^{bcd}$	$86.63 \pm 0.24^a$	$66.26 \pm 1.20^a$
AP_SNA	$112.16 \pm 6.07^b$	$8.91 \pm 0.01^a$	100 <sup>ab</sup>	$69.11 \pm 3.29^a$

Mean values  $\pm$  standard deviations. Different superscript letters (<sup>a–d</sup>) within the same column indicate significant differences between the films ( $p < 0.05$ ).

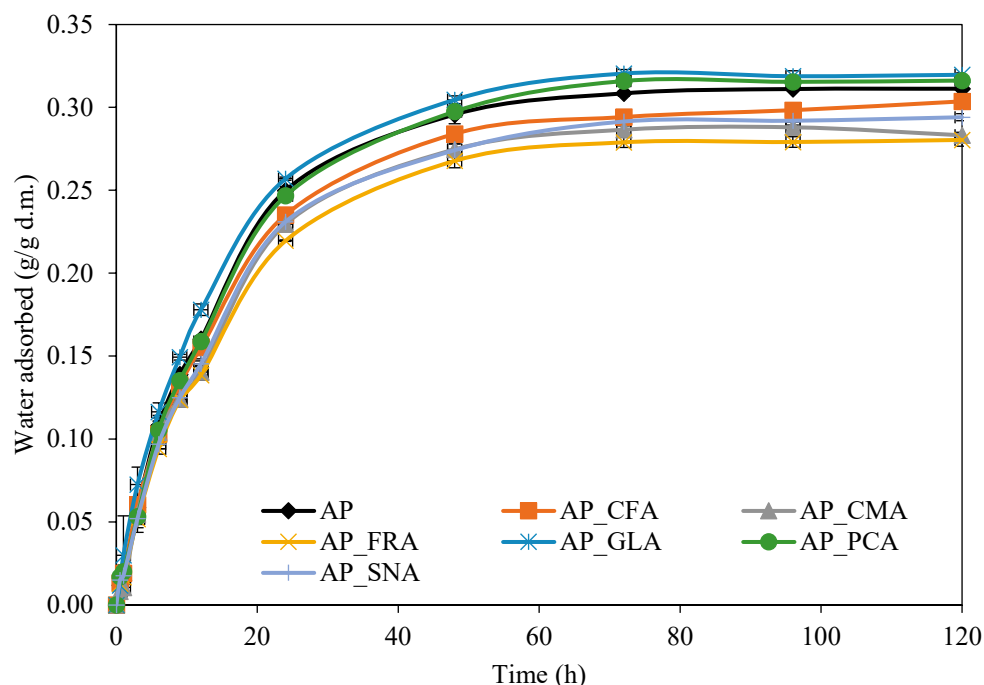
## 2.2. The Effect of Phenolic Acids on the Swelling Index of Apple Pectin Edible Films

The swelling parameter of the control and pectin films with added phenolic acids after being placed in a beaker with distilled water was also analysed (Table 1). The solubility parameter allows for determining the extent to which the edible film can act as a barrier and provide resistance to moisture. The swelling index indicates the film's ability to retain water resulting from a hydrophilic group in its structure in the film matrix. Groups that easily interact with water include the hydrophilic and hydroxyl groups [31]. A moment after being placed in water, the apple pectin films and those with added gallic, coumaric, and sinapic acids began to dissolve in water and lose their integrity. This phenomenon can be attributed to the hydrophilic hydroxyl groups present in the structure of the films. Pectin films dissolve in water due to the pectin structure's hydroxyl and non-esterified carboxyl functional groups, which can form hydrogen bonds [32]. The control films obtained without the addition of phenolic acids dissolved the fastest after being placed in distilled water (100%), which could be due to the good solubility properties of the film and biodegradability. In addition, it was found that the use of a plasticiser, glycerol, had no effect on the water solubility parameter because the same amount was used in each variant. The addition of ferulic, caffeic and protocatechuic acids influenced the stability of the film after being placed in water, which meant that they did not decompose. The lower water solubility of the film may also be due to the formation of

long-chain molecules with low water solubility. The highest value of the swelling index in water was recorded for the film with the addition of caffeic acid ( $88.33 \pm 1.06\%$ ). Phenolic acids contain many hydroxyl groups that are capable of binding water. According to reports by Rahmawati et al. [33], adding more gallic acid reduces the thickness and water solubility of edible films. Polysaccharides and phenolic compounds are characterised by their easy formation of complexes in food systems, which are usually driven mainly by hydrogen bonds and, to a lesser extent, by hydrophobic interactions, affecting phenolic compounds' chemical stability and bioavailability. In addition, gallic acid has a hydrophobic nature, which may result in film swelling [31].

### 2.3. The Effect of Phenolic Acids on the Water Vapour Sorption Kinetics

The water vapour transport rate through edible films depends on the adsorption, diffusion, and desorption rates. This phenomenon involves water molecules dissolving on one side of the film, moving into the empty space between polymer segments, and then desorbing from the polymer surface on the other side of the edible film [31]. The physicochemical properties of the films largely depend on the intermolecular interactions occurring in the film matrix. Figure 2 shows the kinetics of the water vapour sorption of the film. The highest water adsorption value was observed for the pectin film with the addition of gallic acid ( $0.320 \pm 0.001$  g/g d.m.), and the lowest was for the film obtained with ferulic acid ( $0.280 \pm 0.004$  g/g d.m.). All the analysed film variants reached equilibrium within 24 h. Moreover, the obtained films' water vapour sorption kinetics curves were characterised by a similar shape and course. In the initial phase, all the films did not differ significantly in water content, which allows us to state that the driving force of the sorption process was similar and was most intensive during the initial 10 h.



**Figure 2.** Water adsorbed by edible films for the apple pectin (AP) edible films obtained with the addition of caffeic (CFA), coumaric (CMA), ferulic (FRA), gallic (GLA), protocatechuic (PCA), and sinapic (SNA) acids.

The barrier properties of films are mainly influenced by the materials from which the edible films are obtained, the additives, and the film preparation process itself. According to Cheng et al. [34], adding phenolic acids to the film matrix reduces the films' water

vapour permeability (WVP). Moreover, it was found that this tendency is dominant at low concentrations because the presence of hydrophobic phenolic compounds limits the diffusion of water through the matrix.

The values of the water vapour diffusion coefficient in edible films are shown in Table 2. It was observed that adding phenolic acids to film-forming solutions influenced the increase in the water vapour diffusion coefficient. The values ranged from  $0.87 \pm 0.04 \times 10^{-14} \text{ m}^2/\text{s}$  for control films (AP) to  $2.80 \pm 0.18 \times 10^{-14} \text{ m}^2/\text{s}$  for films containing phenolic acids. The lowest increase occurred for films with the addition of protocatechuic acid ( $0.88 \pm 0.03 \times 10^{-14} \text{ m}^2/\text{s}$ ), while the highest was for coumaric and ferulic acids. Ordoñez et al. [35] found that the active compound's diffusion coefficient and release percentage highly depend on the polarity of the food substrates and the polymer matrix. This is due to different molecular interactions and the chemical affinity between the active compound, the polymer matrix, and food substrates. Various values may also result from differences in the molecular weight of the compound and its structure, the microstructure of the film, and the strength of the compound's bond in the matrix. In addition, the factors influencing the value of the diffusion coefficient are the compound's solubility and the interactions between the substance and the polymer, which affect the modification of the cohesive forces of the polymer chain.

**Table 2.** Water vapour diffusion coefficient for the apple pectin (AP) edible films obtained with the addition of caffeic (CFA), coumaric (CMA), ferulic (FRA), gallic (GLA), protocatechuic (PCA), and sinapic (SNA) acids.

Film Type	Water Vapour Diffusion Coefficient ( $\times 10^{-14} \text{ m}^2/\text{s}$ )
AP	$0.87 \pm 0.04^a$
AP_CFA	$1.72 \pm 0.03^c$
AP_CMA	$2.80 \pm 0.18^e$
AP_FRA	$2.23 \pm 0.13^d$
AP_GLA	$1.23 \pm 0.13^b$
AP_PCA	$0.88 \pm 0.03^a$
AP_SNA	$1.82 \pm 0.08^c$

Mean values  $\pm$  standard deviations. Different superscript letters (<sup>a-e</sup>) within the same column indicate significant differences between the films ( $p < 0.05$ ).

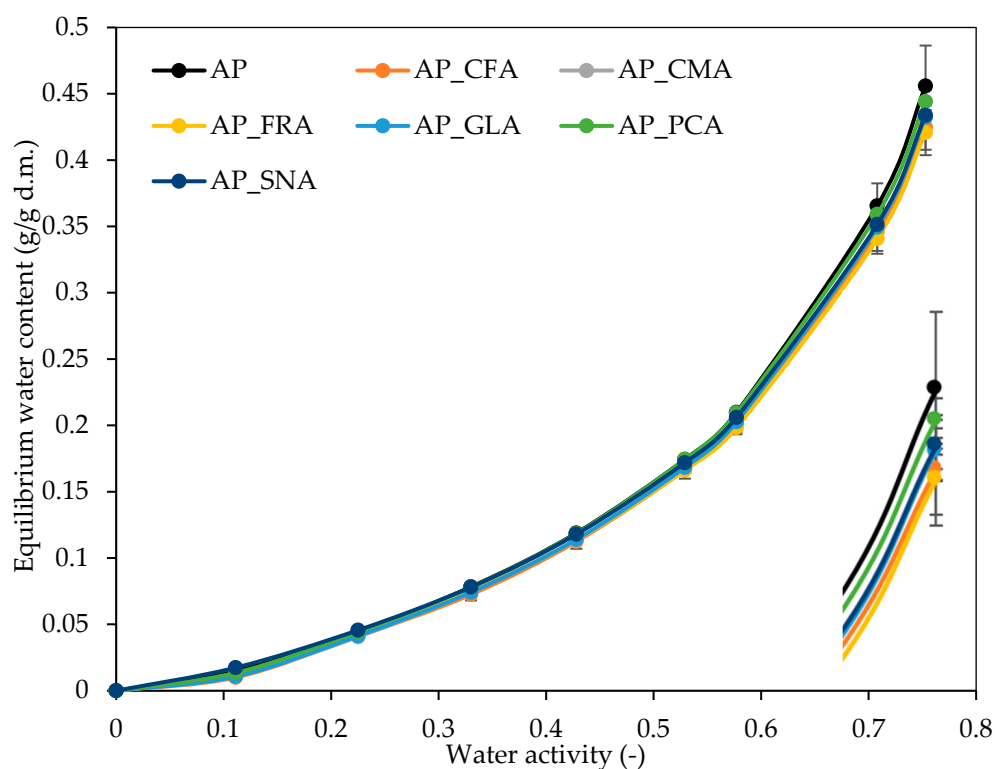
#### 2.4. The Effect of Phenolic Acids on the Water Vapour Sorption Isotherms

Water sorption isotherms are widely used to express the relationship between moisture content and water activity for various materials. In the case of hydrophilic biopolymer films, this is of great importance because the characteristics of such films indicate the sensitivity of the films to water, which is tantamount to the existence of a relationship between their functionality and water [36]. Isotherms can be represented as adsorption or desorption isotherms. Adsorption isotherms are obtained by measuring the increase in weight due to moisture uptake. This phenomenon occurs when a completely dry material is placed in an environment of increasing relative humidity. Desorption isotherms are obtained when a wet material is placed in an environment of constant relative humidity. In the case of adsorption isotherms, five types are distinguished, which depend on the shape of the curve and the process [37].

Moisture adsorption is a very important indicator that determines the sensitivity of the material to moisture. Figure 3 shows the water vapour sorption isotherms for the control film and with the addition of selected phenolic acids. The shape of the isotherms for pectin films with phenolic acids was smaller at lower water activity, and with each increase in water activity, there was an increase in relative humidity. In addition, the

sorption process was most intensive in the initial 10 h. The isotherms shown in Figure 3 represent a type of isotherms called Flory–Huggins isotherms, which are characterised by convexity at all points. According to Nazreen et al. [38], this type of graph occurs due to the weak interaction between the adsorbent surface and the adsorbate, which is necessary to determine the stability of the film packaging. This type of isotherms is often assigned to edible films, e.g., chitosan films. The determination of the film's sensitivity to moisture and the type of environment is necessary to maintain the shelf life of the package, as well as the quality of the food product and the stability during transport and storage. Furthermore, the isothermal moisture sorption of the material represents the predicted amount of water that will be retained in the material under specific conditions of relative humidity and temperature.

Also, Veras et al. [36] obtained similar shape sorption isotherms of pectin films with propolis. It was observed that the equilibrium water content in the film decreased with increasing temperature. According to Othman et al. [39], higher temperatures are closely related to lower water content in the film in the monolayer, which results in fewer sorption sites. Therefore, it is stated that storing the film at high temperatures and relative humidity reduces the possibility of avoiding autoxidation. This increases the probability that the edible film will be characterised by instability during storage.



**Figure 3.** Water vapour sorption isotherms of the apple pectin (AP) edible films obtained with the addition of caffeic (CFA), coumaric (CMA), ferulic (FRA), gallic (GLA), protocatechuic (PCA), and sinapic (SNA) acids.

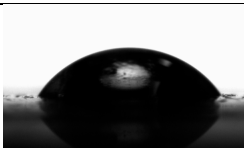
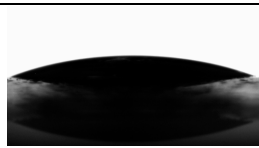


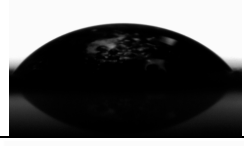


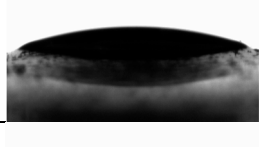
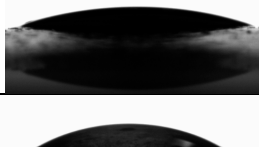
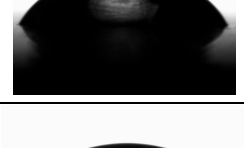

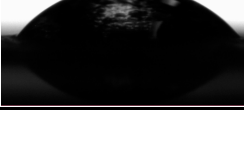
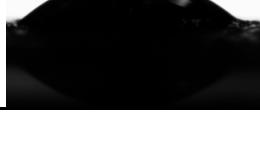
### 2.5. The Effect of Phenolic Acids on the Water Contact Angle

In the case of packaging materials, resistance to water absorption is a very important property. The hydrophilicity of edible films can be determined based on the water contact angle of the film surface layer, which also provides information on the interactions occurring at the phase boundary. The results of water contact angles for edible films are presented in Table 3. It is stated that surfaces with a water contact angle of less than  $90^\circ$

are hydrophilic, while surfaces with a water contact angle greater than  $90^\circ$  are characterised by hydrophilicity and complete or partial wetting [40].

As can be seen from Table 3, the initial water contact values for the films ranged from  $47.00^\circ \pm 4.47$  to  $58.44^\circ \pm 5.62$ . After 60 s, the most hydrophobic surface was observed in films with the addition of caffeic acid ( $60.92^\circ \pm 4.93$ ) and protocatechuic acid ( $56.42^\circ \pm 4.27$ ). This may be due to the increased hydrophobicity of the film due to the strengthening of the structure and the occurrence of intermolecular interactions between the matrix and caffeic acid. In addition, the water contact angle can also be related to the occurrence of slight surface roughness of the film. This statement results from the experiments which showed that the water contact angle depends not only on the interfacial energies but also on the surface structure, its pretreatment, and the existing contaminants [41]. According to Tavassoli-Kafran et al. [42], a high affinity for water characterises hydrophilic surfaces with a water contact angle of less than  $90^\circ$ . According to Żelaziński [43], the most hydrophobic materials defined as biodegradable can achieve a water contact angle of up to  $158^\circ$ . It is also worth noting that pure polylactic acid (PLA) has a water contact of  $75^\circ$ .

**Table 3.** Water contact angle ( $\theta$ ) results and contact angle photos at 0 and 60 s for the apple pectin (AP) edible films obtained with the addition of caffeic (CFA), coumaric (CMA), ferulic (FRA), gallic (GLA), protocatechuic (PCA), and sinapic (SNA) acids.

Film Type	$\theta$ ( $^\circ$ )			
	Time (s)			
	0		60	
AP	$58.44 \pm 5.62^b$		$25.71 \pm 1.98^a$	
AP_CFA	$58.40 \pm 4.47^b$		$60.92 \pm 4.93^c$	
AP_CMA	$52.29 \pm 2.38^{ab}$		$26.88 \pm 2.12^a$	
AP_FRA	$47.00 \pm 4.47^a$		$25.03 \pm 9.23^a$	
AP_GLA	$47.61 \pm 4.02^a$		$24.81 \pm 3.82^a$	
AP_PCA	$56.51 \pm 5.91^b$		$56.42 \pm 4.27^{bc}$	
AP_SNA	$55.36 \pm 2.89^b$		$43.11 \pm 3.95^b$	

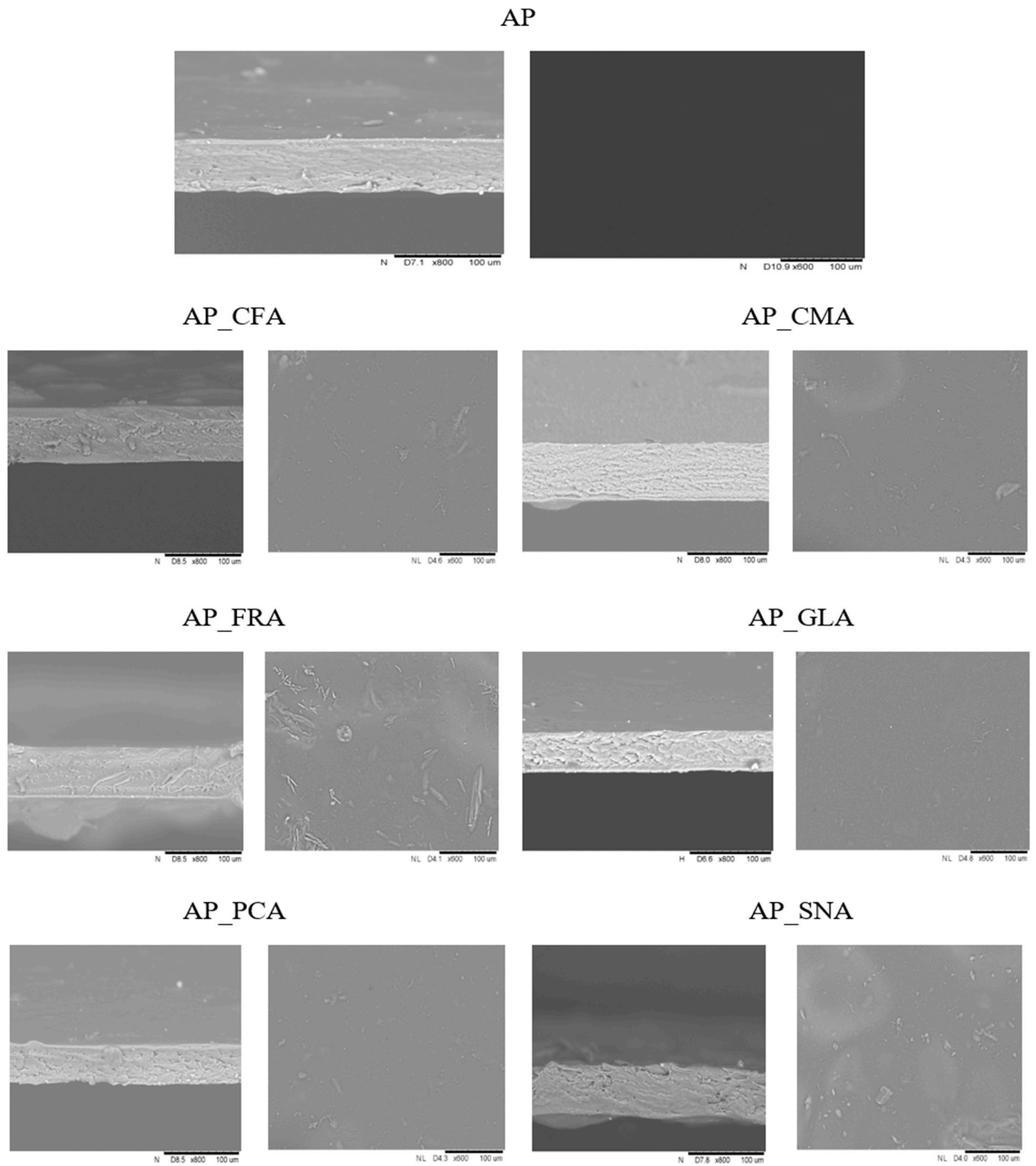
Mean values  $\pm$  standard deviations. Different superscript letters (a-c) within the same column indicate significant differences between the films ( $p < 0.05$ ).

### 2.6. The Effect of Phenolic Acids on the Microstructure of Pectin Films

Microstructure is a key parameter for controlling the mechanical and barrier properties of edible films and is essential for understanding the fundamentals of materials science from a practical point of view [44]. Scanning electron microscopy (SEM) is performed to determine the morphological features of the surface and the cross-sections of edible films. The study of the microstructure of films allows for the determination of the influence of modifiers on the structure formation processes, as well as the simultaneous identification of cracks, porosity, roughness, or homogeneity of edible films. SEM is recommended for studying the microstructure primarily in composite films, nanoemulsions, or when using functional additives, e.g., nanoparticles [45]. In addition, the microstructure of edible films depends on how the edible films are prepared, the ingredients used, and the interactions between the plasticiser and the components [7].

The observation of the surface (600 $\times$ ) and cross-sections (800 $\times$ ) of the obtained pectin films with the addition of selected phenolic acids was carried out using scanning electron microscopy (SEM) (Figure 4). Based on different additions of phenolic acids, the obtained films can have different structures. No cracks were found in the cross-section. Microphotographs of the control films and those with the addition of phenolic acids are smooth, uniform, and compact, which confirms their high compatibility, good adhesion, and integrity between them. The layer adjacent to the sheet of the obtained films was shiny, while the other side was more matt. This may be due to the drying conditions or the addition of various types of phenolic acids. The control film showed a smooth and uniform structure on the outer and substrate sides.

The smooth surface of the film indicates good compatibility between pectin and phenolic acids. According to Yerramathi et al. [25], controlled cross-linking of ferulic acid results in an edible biopolymer with a homogeneous and stable structure, which is used for preserving food products and other production applications. In addition, the studies conducted by Yong et al. [46] show that the hydroxyl groups present in hydroxycinnamic acids (e.g., p-coumaric acid, caffeic acid, and ferulic acid) can interact with chitosan skeletons and glycerol through the hydrogen bonds that occur, ensuring a homogeneous structure of the film. The tested caffeic acid had two hydroxyl groups, allowing it to interact with chitosan skeletons and the plasticiser (glycerol). Adding phenolic acids to the film matrix reduces the roughness parameters of the pectin films [47].



**Figure 4.** Photographs of cross-sections and surfaces of edible films for the apple pectin (AP) edible films obtained with the addition of caffeic (CFA), coumaric (CMA), ferulic (FRA), gallic (GLA), protocatechuic (PCA), and sinapic (SNA) acids. Magnifications: 600× (cross-sections) and 800× (film surfaces).

### 2.7. The Effect of Phenolic Acids on the Water Vapour Permeability of Pectin Films

In general, when applied to food products, the main function of an edible film or coating is to reduce moisture transfer between the coated food and the surrounding atmosphere or between two components of a heterogeneous food product. Therefore, it is essential that the water vapour permeability is as low as possible. The results for the water vapour permeability of the developed active packaging films are presented in Table 4. The values ranged from 7.16 to  $10.48 \times 10^{-10}$  g/m·s·Pa. A decrease in water vapour barrier efficiency can be observed due to the addition of phenolic acids. However, the values were similar for films containing gallic and protocatechuic ( $\times 10^{-10}$  g/m·s·Pa) acids when compared to the control films ( $7.45\text{--}7.46 \times 10^{-10}$  g/m·s·Pa), and the differences were not statistically significant ( $p < 0.05$ ). On the other hand, the rest of the films revealed significantly higher water vapour permeability values of  $9.76 \times 10^{-10}$  g/m·s·Pa for caffeic acid and  $10.45\text{--}10.68 \times 10^{-10}$  g/m·s·Pa for films containing other phenolic acids. This was probably attributed to the differences in compatibility between acids and biopolymers, the acid distribution in the film matrix due to the various rates of solubility, film structure creation during storage (evaporation of water), and the film thickness. Moreover, the hydrophilicity of the plasticiser (glycerol used in this study) is one of the main factors that affect water vapour permeability. Glycerol causes mobility in the film matrix by reducing intermolecular forces and creating free volume. In this context, the motion of water vapour becomes easier in biopolymer-based materials.

The obtained results of water vapour permeability of pectin films containing phenolic acids are similar to other biopolymeric films. However, the values vary due to biopolymer type or different methods used for film preparation, as well as differences in methods conducting water vapour permeability test. Thus, various conditions (temperature or humidity differentials in the gravimetric method) are also used. Hager et al. noted that the addition of tannic acid (1–30%) could potentially decrease the water vapour permeability of wheat gluten films. The authors explained that the improvement of barrier properties is probably related to the tight gluten–tannic acid network, which likely occupies previously hydrophilic hydroxyl groups and traps small air bubbles inside the matrix, resulting in reduced water molecule sorption in the cross-linked film. On the other hand, for wheat gluten films containing gallic acid, an increase in values at lower concentrations (1–2%) followed by a decrease at higher concentrations (5–10%) was observed [48]. The same tendency phenomenon was observed for chitosan films incorporated with gallic acid [49]. Similar observations were noted by Yang et al. [24]. The addition of tannic acid to pectin films resulted in a decrease in the water vapour transmission rate, which the authors attribute to the crosslinking between tannic acid and pectin, leading to a denser network structure within the pectin film and consequently reducing the water vapour permeability.

**Table 4.** Water vapour permeability (WVP) of edible films based on apple pectin (AP) with the addition of caffeic (CFA), coumaric (CMA), ferulic (FRA), gallic (GLA), protocatechuic (PCA), and sinapic (SNA) acids.

Film Type	WVP ( $\times 10^{-10}$ g/m·s·Pa)
AP	$7.16 \pm 0.42^a$
AP_CFA	$9.76 \pm 0.06^b$
AP_CMA	$10.68 \pm 0.12^c$
AP_FRA	$10.46 \pm 0.16^c$
AP_GLA	$7.45 \pm 0.17^a$
AP_PCA	$7.46 \pm 0.22^a$
AP_SNA	$10.48 \pm 0.22^c$

Mean values  $\pm$  standard deviations. Different superscript letters (<sup>a-c</sup>) within the same column indicate significant differences between the films ( $p < 0.05$ ).

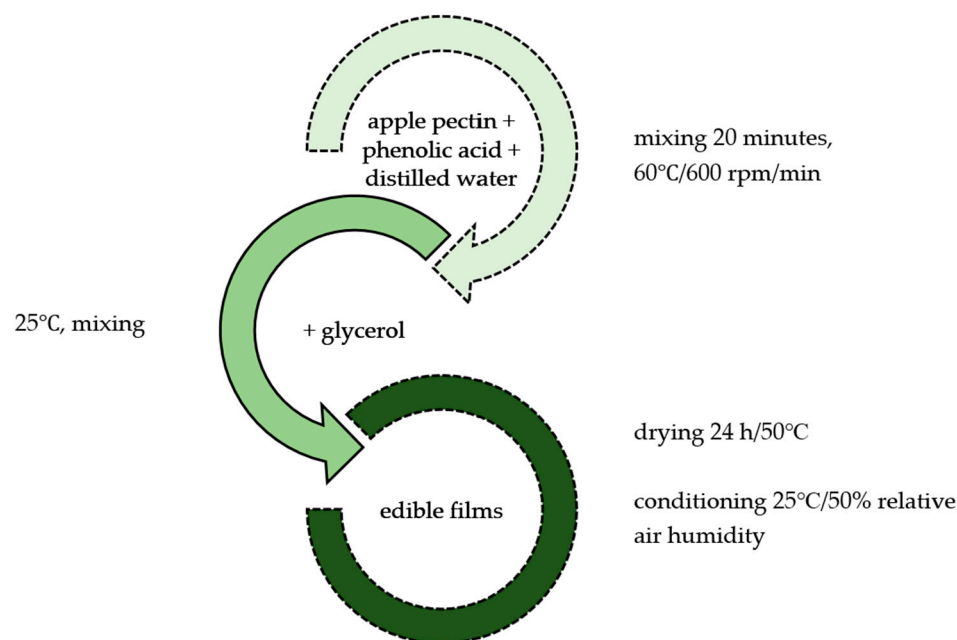
### 3. Materials and Methods

#### 3.1. Materials

The research material consisted of edible films produced based on apple pectin (Pektowin S.A., Jasło, Poland), selected phenolic acids (POL-AURA) (Warsaw, Poland), such as ferulic acid, gallic acid, caffeic acid, coumaric acid, protocatechuic acid (Thermo Scientific) (Gdańsk, Poland), and sinapic acid (Acros Organics) (Poznań, Poland). Glycerol (Avantor Performance Materials, Gliwice, Poland) was a plasticising agent.

#### 3.2. Film Preparation

The film-forming preparation steps are presented in Figure 5. Aqueous film-forming solutions were prepared with a concentration of 5% apple pectin, selected phenolic acids (5%), and the addition of a plasticiser (50% in relation to the weight of apple pectin).



**Figure 5.** Scheme of film preparation.

The solutions were mixed for 20 min using an RCT basic IKAMAG hot plate (60 °C) (IKA Werke Gmn & Co., Staufen, Germany) and an RCT basic IKAMAG magnetic stirrer (IKA Poland, Warsaw) rotating at a speed of 600 rpm. The film-forming solutions were poured onto sheets at 10 mm/s and a layer thickness of 2500  $\mu\text{m}$  using a Zehntner ZAA 2300 automatic film applicator (Zehntner GmbH Testing Instruments, Sissach, Switzerland). The films were dried in a SUP-65W laboratory dryer (Wamed, Warsaw, Poland) for 24 h at 50 °C. Then, the obtained films were conditioned in a KFB 240 thermostatic chamber (Binder, Tuttlingen, Germany) at 25 °C and 50% relative air humidity for 48 h prior to testing.

#### 3.3. Thickness

The thickness was determined using a thickness tester (Thwing-Albert, ProGage Thickness Tester, West Berlin, NJ, USA) with an accuracy of 1  $\mu\text{m}$ . The thickness of the films was measured for each experiment at least in 3 repetitions.

### 3.4. Water Content

The determination of the water content in the edible films was carried out by drying samples in a laboratory dryer (SUP 65 WG, WAMED, Warsaw, Poland) at 105 °C for 24 h with an accuracy of ±0.0001 g using an analytical balance (RADWAG PS 600/C/2, Radom, Poland). Measurements were made in 3 repetitions, and the dry matter was calculated according to the following equation:

$$d.m. = \frac{m_s - m}{m_p - m} \cdot 100\%$$

where  $m_s$  is the sample weight after drying (g),  $m_p$  is the sample weight before drying (g), and  $m$  is the mass of the empty weighing vessel (g).

### 3.5. Swelling Index

The prepared foils in squares with dimensions of 2 × 2 cm were weighed on an analytical balance with an accuracy of ±0.0001 g; then, the samples were placed in 25 mL of distilled water for 2 min. Then, the films were filtered to remove excess water using filter paper and weighed again. Each measurement was performed in 3 repetitions. The swelling of the foil was calculated based on the following formula [50]:

$$P = \frac{m_2 - m_1}{m_1} \cdot 100\%$$

where

$P$  is the swelling of the edible films (%);

$m_1$  is the sample weight before swelling (g);

$m_2$  is the sample weight after swelling (g).

### 3.6. Water Solubility

The films in squares with 2 × 2 cm sides were placed in glass vessels, weighed on an analytical balance with an accuracy of ±0.0001 g, and dried for 24 h at 105 °C. After this time, the samples were cooled in a silica gel desiccator. The films were weighed again and placed in 25 mL of distilled water. After 24 h of storage and occasional mixing, excess water was removed using filter paper. The films were again placed in the laboratory dryer for 24 h at 105 °C and weighed. The measurement was performed in 3 repetitions, and the solubility in water ( $R$ ) was determined based on the following formula [51]:

$$R = \frac{m_0 - m_r}{m_0} \times 100\%$$

where  $m_0$  is the dry mass of the sample before dissolving (g) and  $m_r$  is the dry mass of the sample after dissolving (g).

### 3.7. Water Vapour Sorption Kinetics

The water vapour sorption kinetics was determined based on the change in mass of the film samples with a mass of ±0.25 g and with an accuracy of ±0.0001 g (Radwag, Radom, Poland), with weighing conducted over a period of time. The determination was performed at a relative humidity of 100% (distilled water) at times 0, 0.5, 1, 3, 6, 9, 12, 24, 48, 72, 96, and 120 h. Based on the obtained kinetic curves, the results of water vapour sorption kinetics were interpreted, which were graphs of the dependence of the change in the amount of water that was adsorbed (g/g dry matter) on the time of the process (h). Using Fick's second law, the diffusion coefficient ( $D$ ) of water vapour was calculated based on the following formula:

$$\frac{M_t - M_0}{M_e - M_0} = 1 - \sum_{n=0}^{\infty} \frac{8}{(2n+1)^2 \pi^2} \exp \left[ -\frac{(2n+1)^2 \pi^2}{4L^2} Dt \right]$$

where  $M_t$  is the total amount of adsorbed water (g/g dry matter) during the process (s),  $M_0$  is the initial water content (g/g d.m),  $M_e$  is the water content over time  $t$  (g/g d.m), and  $L$  is the film thickness (m).

### 3.8. Water Vapour Sorption Isotherms

The water vapour adsorption isotherms of the coatings were determined in 2 repetitions using the dynamic water vapour sorption apparatus Aquadyne DVS-2HT (Quantachrome Instruments by Anton Paar Sp. z o.o., Warsaw, Poland) in the range of environmental relative humidities from 0 to 75%. Experimental data points were analysed using Microsoft Excel 2020 and aquaWIN Software (latest version Air3). The water vapour adsorption isotherms were presented as curves of the dependence of equilibrated water content on water activity.

### 3.9. Water Contact Angle Measurement

The water contact angle analysis was performed using the drop-by-drop method with an OCA 25 goniometer (DataPhysics Instruments, Filderstadt, Germany). The contact angle was measured after applying a 10  $\mu$ L drop of distilled water at a rate of 10  $\mu$ L/s to the film surface. The analysis was performed in at least 6 repetitions at 0 and 60 s, and the results were processed using the SCA20\_U software (Version 5.0.37).

### 3.10. Scanning Electron Microscopy

The observations of the cross-sections' structure and the films' surface were made using a TM3000 table scanning electron microscope (Hitachi High Tech, Tokyo, Japan). The 5  $\times$  5 mm films were placed on the measuring table using the carbon paste PELCO with a diameter of 9 mm (Pik Instruments Sp. z o.o., Piaseczno, Poland). The measurement was carried out in a low vacuum condition of 0.35–1 torr with the cross-sections magnified at 800 $\times$  and the surface magnified at 600 $\times$ .

### 3.11. Water Vapour Permeability

A gravimetric method was used to evaluate the water vapour permeability of the analysed films using the Mater Cup FX-3180 equipment (Textest AG, Schwerzenbach, Switzerland). Three samples were cut from each film, and their thickness was measured. The samples were placed between two rubber-based rings on top of cells containing distilled water, using a relative humidity gradient of 50–100% and a permeation surface of 28.3 cm<sup>2</sup>.

### 3.12. Statistical Analysis

Statistical analysis was performed using the Statistica 13.3 programme by analysing the variance in the system with repeated measurements and one-way analysis of variance (ANOVA) with the Tukey post hoc test, with a significance level 0.05.

## 4. Conclusions

The studies confirmed that phenolic acids are effective cross-linking agents for materials based on apple pectin. The analysis of the thickness and water content of the films showed that the addition of phenolic acids increased their thickness and reduced the water content. In addition, the sorption process was most intensive in the first 10 h and was similar for all film variants. Films obtained based on apple pectin modified with

caffeic acid are promising in terms of potential use as packaging because they achieved the highest values of the water contact angle and, thus, the highest hydrophobicity, limiting water absorption. Using edible films with the addition of caffeic acid could minimise the adverse effect of moisture on the product. On the other hand, films containing gallic acid and protocatechuic acids showed the lowest water vapour permeability values among active films, thus indicating the potential for applications for food products where water barrier efficiency is crucial. Microscopic analysis proved that all the films obtained were smooth and homogeneous, with good compatibility between apple pectin and phenolic acids. The obtained edible films could have potential applications in the food industry, as they prevent changes in colour and texture. However, more research is needed for coated food products or the application of developed films as edible pouches. This need arises from evaluating the mechanical strength and durability of the materials, as well as the compatibility of the edible films with food products.

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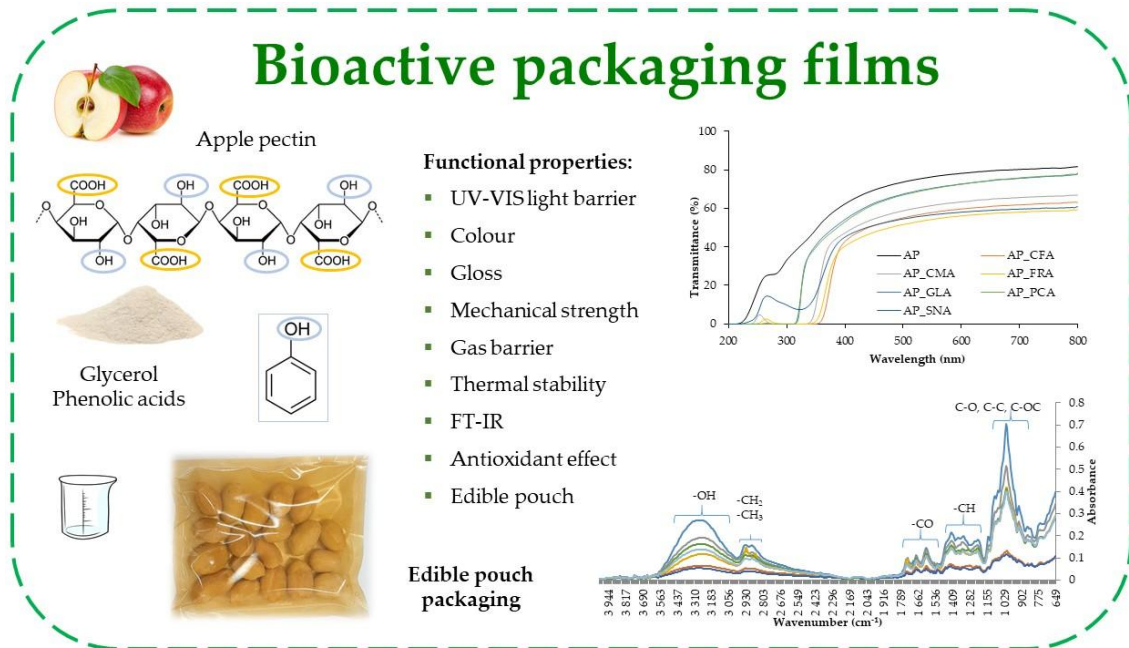
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# Publikacja 3

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## Abstrakt graficzny



## Article

# The Effect of Selected Phenolic Acids on the Functional Properties of Pectin-Based Packaging Films

Magdalena Mikus and Sabina Galus \* 

Warsaw University of Life Sciences, Institute of Food Sciences, Department of Food Engineering and Process Management, 02-776 Warsaw, Poland; magdalena\_mikus@sggw.edu.pl

\* Correspondence: sabina\_galus@sggw.edu.pl

## Abstract

In this study, pectin packaging films were enhanced with selected phenolic acids, including caffeic, coumaric, ferulic, gallic, protocatechuic, and sinapic acids. Edible films were created from apple pectin aqueous solutions that were plasticised with glycerol. The evaluation covered various properties, including optical, barrier, mechanical, thermal, structural, and antioxidant activity. The findings showed that phenolic acids are beneficial and compatible components for pectin films. A higher barrier against UV-VIS light and mechanical strength, as well as a more resilient structure, was observed. All the films exhibited a compact and uniform structure, along with transparency and a light colour. The addition of phenolic acids caused greater permeability to oxygen and carbon. Except for caffeic and protocatechuic acids, which resulted in lower values of permeability for both gases, the other acids improved gas transmission. Fourier transform infrared spectroscopy (FT-IR) analysis confirmed several functional groups, including hydroxyl (–OH) and carbonyl (C=O) groups. All films containing phenolic acids demonstrated increased antioxidant activity, with variations depending on the specific compound.

**Keywords:** edible films; apple pectin; phenolic acids; colour stability; mechanical properties

## 1. Introduction

Many packaging materials used for food products are also composed of petrochemical polymers, which, when discarded as waste, can have a harmful consequence on both the environment and human health due to the migration of certain substances [1,2]. The emphasis on using environmentally friendly alternative packaging solutions is growing. Therefore, there is a need for new resources for packaging films, mainly those made from renewable sources [3]. These materials offer a viable alternative to synthetic packaging, especially because of their biodegradability [4,5]. Additionally, utilising coatings and films derived from polysaccharides presents new opportunities for developing innovative packaging systems [6–8]. Plasticisers are used to correct mechanical and barrier limitations in edible films, increasing their flexibility. This allows for the preparation of packaging materials for various food products [9]. This approach offers alternatives to traditional packaging and the opportunity to meet consumer expectations for more sustainable and environmentally friendly solutions [10–12].

Edible packaging films are thin layers created from bio-based materials such as polysaccharides, proteins and lipids [13]. They can be placed on food products or sandwiched between food ingredients [14] and used as self-standing edible pouch packaging for different applications [15]. The properties of edible packaging films depend mainly on the



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physical and chemical properties of the biopolymers used [16,17]. It is important to select edible packaging materials that align with the food product category and storage conditions while paying particular attention to temperature [18].

Edible films not only serve protective purposes but can also be designed to offer antimicrobial and antioxidant benefits. They can enhance consumer sensory experiences by incorporating various flavours or colours for personalisation. Furthermore, edible films can be fortified with vitamins or minerals, enabling products to be customised to meet specific consumer needs [19,20]. Integrating innovative biomaterials enables continuous improvement of edible packaging and is in line with circular approach [14]. The creation of biodegradable and functional materials that incorporate bioactive compounds derived from plants and agricultural products offers a sustainable alternative to conventional packaging [12,21]. These materials provide important functional and antioxidant advantages that improve food preservation, minimise environmental impact, and inhibit food spoilage and microbial growth [22]. Embedding active compounds within polymer matrices can effectively resolve challenges such as inadequate mechanical strength, subpar water resistance, and restricted antioxidant capabilities. To overcome these issues, natural bioactive compounds like phenolic acids are employed to improve both functional and protective characteristics [23,24].

Pectin, a polysaccharide found primarily in plants, is considered ideal for producing composite coatings or films. It has a branched structure and is made of  $\beta$ -(1,4)-D-galacturonic acid [25]. Pectin is particularly appreciated for its natural characteristics and its sustainable character, as it can be obtained from byproducts. The primary waste that is used for production of pectin include fruit peels, mainly from oranges and lemons, and fruit pomace, both of which can be converted into valuable packaging materials [18,26]. One advantage of pectin films is their high transparency and resistance to moisture and gases. Additionally, controlling gas exchange helps preserve the colour, texture, and nutritional value of products [25]. Applying pectin as a coating material provides numerous advantages, such as its nutraceutical and probiotic benefits [27].

Polysaccharide coatings and films have limitations compared to plastic-based options, including poor mechanical properties and a higher risk of microbial contamination [28]. One method is to incorporate bioactive compounds, like phenolic compounds or extracts that are high in polyphenols. The incorporation of these ingredients aims to formulate multifunctional films and coatings to enhance their physical and chemical properties [12,29]. By combining pectin with a selected phenolic acid, a new substance can be synthesised that exhibits improved biological activity, including enhanced antioxidant and antibacterial properties [30]. Additionally, the inclusion of phenolic acids enhances the amphiphilic properties of hydrophilic pectin, improving its emulsifying capabilities and broadening its applications in food preservation [31]. Karaki et al. [32] In their study showed that incorporating ferulic acid units into the pectin structure enhanced its hydrophobicity and antioxidant activity.

Phenolic compounds are naturally occurring bioactive substances present in various plant sources, including fruits, vegetables, oils, herbs, agricultural waste, and industrial byproducts. They exhibit considerable functional and structural diversity, which enhances oxidative stability and imparts antimicrobial properties to food products [33]. Incorporating phenolic acids enhances the amphiphilic characteristics of hydrophilic pectin, leading to improved emulsifying properties. As a result, modifying pectin with phenolic acids could greatly broaden its potential uses within the food industry [31,34]. Phenolic compounds are substances that contain one hydroxyl group directly (at least) bonded with benzene ring [35].

Gallic acid as a bioactive compound is a crucial antioxidant compound that scavenges free radicals, and when incorporated into a film matrix, it improve the film properties. As

a secondary plant metabolite, it possesses antioxidant and antimicrobial properties and contributes to improved thermal stability. However, incorporating larger amounts of gallic acid into edible films may lead to decreased water solubility and diminished elongation at breaks [36]. Gamma-hydroxypropyl acid is recognised for its capacity to change the mechanical resistance of biopolymers. It functions as a compatible component, serving as both a natural cross-linking agent and a plasticizer. Because of this property, it is frequently used as an additive in food packaging materials [37]. Sinapic acid has several attributes such as antioxidant and antibacterial capacity. Although it is a common compound in the plant world and has widespread applications, there are still few reports on its use as a modifier for biopolymers. Additionally, there is limited research on how sinapic acid affects the properties of biopolymer materials [38].

The study aimed to analyse the effect of selected phenolic acids, including caffeic, coumaric, ferulic, gallic, protocatechuic, and sinapic acids, on the functional properties of edible films based on pectin. The evaluation covered optical, barrier, mechanical, thermal, structural, and antioxidant activity.

## 2. Materials and Methods

### 2.1. Materials

Apple pectin was supplied by Pektowin S.A. (Jasło, Poland). The chosen phenolic acids included caffeic acid, coumaric acid, ferulic acid and gallic acid from POL-AURA (Warsaw, Poland), along with protocatechuic acid from Thermo Scientific (Gdańsk, Poland) and sinapic acid from Acros Organics (Poznań, Poland). Glycerol (Avantor Performance Materials, Gliwice, Poland) served as a plasticising agent. Radicals for the analysis of antioxidant activity were sourced from Sigma Aldrich (Poznań, Poland).

### 2.2. Film Preparation

The method for film preparation has been presented in our previous study [39]. Briefly, aqueous film-forming solutions containing pectin (5%), various phenolic acids, and plasticiser (50% apple pectin by weight). were prepared and dried at 50 °C in a SUP-65W dryer (Wamed, Warsaw, Poland).

### 2.3. Optical Properties

#### 2.3.1. UV-VIS Light Transmittance

The UV-VIS light transmittance was evaluated based on the method described by Łyczak et al. [40].

#### 2.3.2. Colour Stability

The colour of edible films was measured with a CR-400 colorimeter (KONICA MINOLTA, Inc., Tokyo, Japan). The CIE  $L^* a^* b^*$  system was used and the measurement were made in 6 repetitions. The total colour difference ( $\Delta E$ ) between the films containing phenolic acids and the control films was obtained using the formula presented by Sobral et al. [41].

#### 2.3.3. Gloss

The gloss of the films at angles of 20°, 60°, and 85° was evaluated in ten replicates using a Multi Gloss 268A (Konica Minolta, Tokyo, Japan). The precision was  $\pm 0.2^\circ$ . Spectral sensitivity was an approximate function of CIE  $y(2^\circ)$  for a CIE C source.

#### 2.3.4. Opacity

The opacity of the edible films was determined using the average values of thickness and absorbance at 600 nm based on the formula:

$$O = \frac{A_{600}}{l}$$

where  $O$  is opacity (a.u./mm),  $A_{600}$  is absorbance value at a wavelength of 600 nm, and  $l$  is the film thickness (mm).

#### 2.4. Barrier Properties

Gas permeability was determined against oxygen and carbon dioxide using C130 gas permeability tester (Labthink Instruments Co., Ltd., Jinan, China) based on the manometric method, in accordance with ASTM D1434-82 [42].

#### 2.5. Thermogravimetric Analysis

Thermogravimetric analysis was performed based on the method described by Łyczak et al. [40] using a TGA thermal analyser (Mettler Toledo, Warsaw, Poland).

#### 2.6. Fourier Transform Infrared Spectroscopy (FT-IR)

FT-IR was determined in triplicate using the ATR method using a Cary-630 spectrometer (Agilent Technologies, Cary, NC, USA). The spectra of the analysed samples were analysed using absorption in the range of 4000–650  $\text{cm}^{-1}$  with a resolution of 4  $\text{cm}^{-1}$ . The spectra were presented as the average of 32 interferograms.

#### 2.7. Mechanical Properties

Mechanical properties were measured using a TA-XT2i texture analyser. Edible films measuring 25 × 100 mm were tested. TextureExpert software (version 2.3) was utilised to determine the mechanical properties. This software recorded parameters related to the load and elongation of the film, which were then used to generate curves depicting the maximum force at break and the elongation of the film. Each determination was performed with a minimum of six repetitions.

#### 2.8. Antioxidant Activity

The antioxidant activity of the analysed films was evaluated in triplicate against ABTS and DPPH radicals and expressed as mg of Trolox/g d.m. The absorbance of the samples was measured at wavelengths of 734 nm for ABTS and 515 nm for DPPH radicals.

#### 2.9. Statistical Analysis

Statistical analysis was done in Statistica 13.0. A one-way analysis of variance (ANOVA) with repeated measures and Tukey's post hoc test were conducted, with significance set at 0.05.

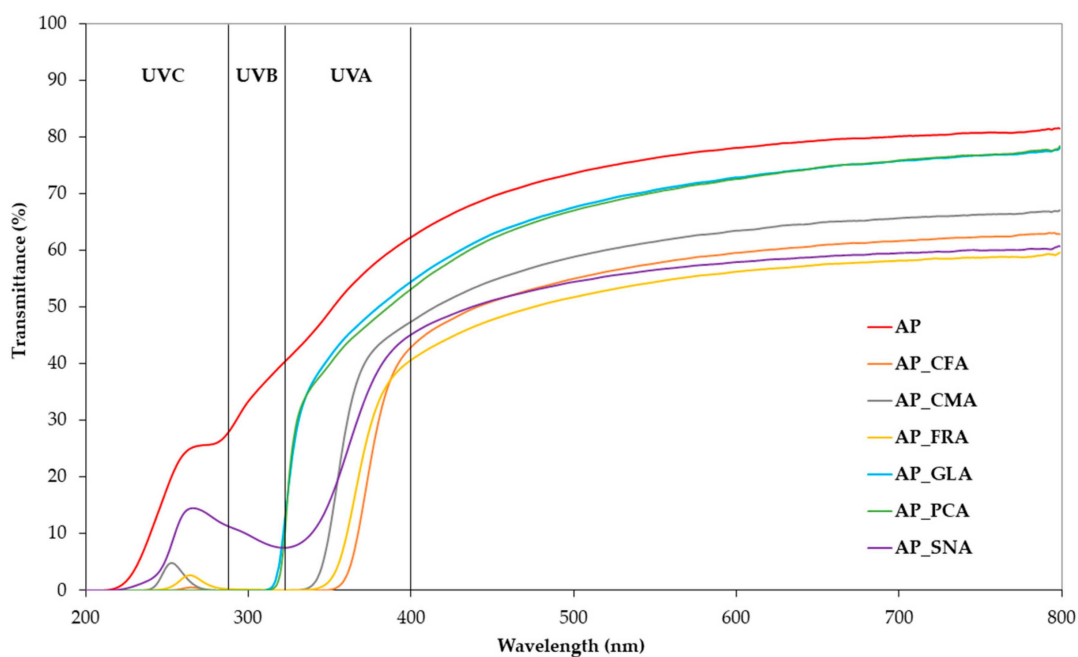
### 3. Results and Discussion

#### 3.1. The Effect of Phenolic Acids on the Optical Properties of Pectin Packaging Films

##### 3.1.1. UV-VIS Transmittance

The effectiveness of a film in blocking visible and ultraviolet (UV) light is important for preserving packaged food. For food products, this blocking ability is essential for evaluating the level of radiation that reaches the food's surface. This exposure can affect photooxidative reactions, which can influence the overall quality and shelf life of the food [43]. The results presented in Figure 1 show the transmittance of pectin films, including enhancement by the addition of various phenolic acids. This analysis measures the amount

of light the film transmits at specific wavelengths. Transmittance measurements were conducted using wavelengths from 200 to 800 nm, allowing for the assessment of optical properties within the ultraviolet range together with visible light. Figure 1 reveals that the films without phenolic acid, treated as controls (AP), had the highest transmittance across the entire wavelength range tested, indicating the lowest level of absorbance. In contrast, a noticeable decrease in transmittance was observed in all variants after the addition of phenolic acids. Specifically, the film with ferulic acid (AP\_FRA) exhibited the lowest transmittance values, indicating that it transmitted the least amount of light within the specified range. The phenolic acids—caffeic, coumaric, ferulic, gallic, and protocatechuic—present in the coatings displayed low transmittance within the 250–300 nm spectrum while showing improved transmittance in the 350–800 nm range. Therefore, it can be concluded that the addition of these acids enhances the film's light barrier. Moreover, the low transmittance observed in the wavelength range below 300 nm suggests an effective capacity for absorbing ultraviolet radiation. Above this range, absorbance gradually decreases, which is characteristic of materials with UV barrier properties that still partially transmit light in the UV-VIS spectrum. Cai et al. [44] observed that the transparency of edible films, measured between 350–800 nm, gradually stabilises as the wavelengths of ultraviolet light increase. Films that exhibit low transmission in the UV-VIS range can be effective materials for protecting light-sensitive products. Therefore, controlling transparency in the visible light spectrum is crucial when designing edible packaging films to achieve specific barrier properties. Moll and Chiralt [43] also found that incorporating ferulic acid into PHBV films significantly reduced light transmission in both visible and UV light. This effect is likely due to the light-absorbing properties of the newly added molecules. Notably, ferulic acid had a more substantial impact on reducing UV light transmission than on visible light. Consequently, films containing active compounds may help to minimise the formation of free radicals in packaged foods, providing protection against light-induced oxidation and delaying the degradation of food components.



**Figure 1.** UV-VIS light spectra of packaging films incorporated with selected phenolic acids: CFA—caffeic acid, CMA—coumaric acid, FRA—ferulic acid, GLA—gallic acid, PCA—protocatechuic acid, SNA—sinapic acid.

### 3.1.2. Colour

Analysing the colour of packaging film is crucial for consumer acceptance of food products [45]. All samples were analysed colourimetrically to determine their colour parameters, and the results are presented in Table 1. The  $L^*$  parameter indicates the contrast between darkness and lightness, with a range from 0 to 100. The  $a^*$  value reflects the shift in film colour from green to red, increasing from negative to positive values, while a rise in the  $b^*$  parameter signifies that the film colour becomes more yellow [46]. Additionally, the total colour difference ( $\Delta E$ ) relative to a control film (pectin films without phenolic acids) was calculated to assess the colour change in the films.

**Table 1.** Optical properties of packaging films incorporated with selected phenolic acids: CFA (caffeic acid), CMA (coumaric acid), FRA (ferulic acid), GLA (gallic acid), PCA (protocatechuic acid), and SNA (sinapic acid).

Film	$L^*$	$a^*$	$b^*$	$\Delta E$
AP	85.65 ± 0.70 <sup>e</sup>	−1.33 ± 0.10 <sup>a</sup>	20.68 ± 2.26 <sup>bc</sup>	-
AP_CFA	82.02 ± 0.87 <sup>b</sup>	−0.49 ± 0.20 <sup>c</sup>	26.22 ± 2.94 <sup>de</sup>	4.94 ± 1.94 <sup>ab</sup>
AP_CMA	83.74 ± 1.02 <sup>c</sup>	−1.02 ± 0.20 <sup>b</sup>	25.13 ± 2.78 <sup>de</sup>	3.29 ± 1.93 <sup>bc</sup>
AP_FRA	80.65 ± 0.66 <sup>a</sup>	0.77 ± 0.33 <sup>d</sup>	27.21 ± 1.18 <sup>e</sup>	6.24 ± 1.38 <sup>a</sup>
AP_GLA	84.94 ± 0.44 <sup>de</sup>	−1.49 ± 0.05 <sup>a</sup>	18.23 ± 1.37 <sup>ab</sup>	4.94 ± 1.44 <sup>ab</sup>
AP_PCA	84.96 ± 0.83 <sup>de</sup>	−0.95 ± 0.09 <sup>b</sup>	17.43 ± 1.58 <sup>a</sup>	5.78 ± 1.66 <sup>a</sup>
AP_SNA	84.36 ± 0.70 <sup>cd</sup>	−0.77 ± 0.16 <sup>b</sup>	23.19 ± 2.11 <sup>cd</sup>	2.08 ± 1.13 <sup>c</sup>

Mean values ± standard deviations. Different superscript letters (<sup>a–e</sup>) within the same column indicate significant differences between the films ( $p < 0.05$ ).

The control and composite films were visually transparent, smooth, and homogeneous. The  $L^*$  values ranged from 80.65 ± 0.66 to 85.65 ± 0.70. The pectin film with ferulic acid (AP\_FRA) exhibited the lowest  $L^*$  value, indicating lightness. The addition of phenolic acids reduced the  $L^*$  parameter in all film variants containing them. The  $a^*$  values ranged from −1.49 ± 0.05 to 0.77 ± 0.33. Therefore, it can be noted that only the films with ferulic acid, characterised by a positive value, showed a tendency towards redness, while all other films showed a slight tendency towards greenness. According to Ngo et al. [47], colour changes in pectin films suggest that an increase in the  $a^*$  colour parameter translates into a shift from green to red colours as the value changes from negative to positive. An increase in the  $b^*$  parameter value indicates a more yellow appearance of the film. All films showed a tendency to yellow. Furthermore, the most pronounced change in yellow colour was observed for pectin films with gallic, caffeic, and coumaric acids. Similar observations were presented by Insaward et al. [48], who observed that the addition of ferulic, caffeic, and gallic acids significantly increased the yellow content of soy protein films. Liu et al. [49], however, observed a slight colour change in chitosan films after the addition of p-coumaric acid, from colourless to light yellow.

The differences in colour are especially apparent in the total colour difference ( $\Delta E$ ). A value above 1 means no noticeable colour difference, while a trained observer may detect differences between values of 1 and 2. Results ranging from 2 to 3.5 are visible to an untrained people, and values above 5 signify a high colour difference compared to the control films [47]. The values obtained indicate significant colour differences that can even be noticed by non-experts. The total colour difference ( $\Delta E$ ) in films with added phenolic acids ranged from 2.08 to 6.24. Among these, the pectin films containing gallic and protocatechuic acids exhibited the most similar values. Notably, the film with the ferulic acid additive (AP\_FRA) was the most visually distinct, showing a greater colour difference compared to the other films.

### 3.1.3. Gloss and Opacity

The transparency and gloss of edible films are essential to their suitability when used as edible coatings, as these properties directly impact the appearance of coated products [50]. Currently, there is a widespread misinterpretation of opacity and transparency phenomena. Transparency describes a material's ability to transmit light, while opacity refers to the degree to which the material avoids light transmission. However, this does not play a crucial role for light reflectance, which may be affected semi-transparent films [51]. The shine of edible packaging films plays a crucial role in enhancing the visual appeal of food products. It not only makes them look more appealing to consumers but also conveys a sense of quality and freshness. Glossy finishes can be imparted by lipids, waxes, and other ingredients incorporated into the film, contributing to the product's overall aesthetic and influencing consumer purchasing decisions [52,53]. The gloss were measured at various angles of incidence (20°, 60°, and 85°) and are presented in Table 2. According to Fabra et al. [50], values at 45° are sufficient for assessing properties of medium-gloss surfaces. However, high-gloss films are better assessed at lower angles, and low-gloss surfaces are better distinguished at higher angles, due to increasing specular reflection with increasing angle of incidence. The incorporation of phenolic acids reduced the gloss of the films at both 20° and 60° angles, from  $73.50 \pm 2.47^\circ$  to  $25.38 \pm 1.38^\circ$  and from  $100.48 \pm 1.69^\circ$  to  $45.45 \pm 1.40^\circ$ , respectively. At 85°, a similar reducing tendency has been observed, from  $76.60 \pm 0.50^\circ$  to  $27.12 \pm 0.80^\circ$ , except for films containing protocatechuic acid. For this sample, higher values were obtained ( $76.66 \pm 0.52^\circ$ ); however, the differences between values were not statistically significant ( $p < 0.05$ ). Based on the results of the gloss measurements, the most significant changes were observed between the control films (AP) and those containing caffeic, ferulic, and sinapic acids. These findings indicate that films containing phenolic acids exhibited reduced glossiness, suggesting that these acids significantly altered the surface appearance of the pectin films. These results align with other optical parameters, such as colour values presented in Table 1 and the observations from the UV-VIS spectra shown in Figure 1.

**Table 2.** The gloss and opacity of packaging films incorporated with selected phenolic acids: CFA—caffeic acid, CMA—coumaric acid, FRA—ferulic acid, GLA—gallic acid, PCA—protocatechuic acid, SNA—sinapic acid.

Film	Gloss (°)			Opacity (a.u./mm)
	20	60	85	
AP	$73.50 \pm 2.47^e$	$100.48 \pm 1.69^d$	$76.60 \pm 0.50^c$	$2.15 \pm 0.17^{ab}$
AP_CFA	$25.38 \pm 1.38^a$	$45.45 \pm 1.40^a$	$27.12 \pm 0.80^a$	$2.53 \pm 0.15^b$
AP_CMA	$39.81 \pm 4.40^c$	$64.56 \pm 4.42^c$	$35.53 \pm 2.07^b$	$1.78 \pm 0.16^a$
AP_FRA	$26.69 \pm 2.67^a$	$48.66 \pm 2.87^a$	$27.67 \pm 1.64^a$	$2.17 \pm 0.27^{ab}$
AP_GLA	$67.96 \pm 3.02^d$	$97.34 \pm 1.50^d$	$75.02 \pm 1.63^c$	$2.35 \pm 0.51^b$
AP_PCA	$70.42 \pm 4.00^{de}$	$100.22 \pm 1.42^d$	$76.66 \pm 0.52^c$	$1.78 \pm 0.25^a$
AP_SNA	$35.15 \pm 2.00^b$	$55.02 \pm 1.79^b$	$27.90 \pm 1.64^a$	$3.03 \pm 0.46^c$

Mean values  $\pm$  standard deviations. Different superscript letters (a–e) within the same column indicate significant differences between the films ( $p < 0.05$ ).

Table 2 also presents the opacity of the films with added phenolic acids, referring to the degree of light blocking at a wavelength of 600 nm in the visible range. The film with sinapic acid exhibited the highest value (3.03 a.u./mm), indicating it was the most opaque sample tested. The lowest values were for films with coumaric and protocatechuic acids,  $1.78 \pm 0.16$  and  $1.78 \pm 0.25$  a.u./mm, respectively, which is even lower than that of the control films ( $2.15 \pm 0.17$  a.u./mm). The other phenolic acids showed higher values, suggesting an enhancement of the film's light barrier properties. The higher the visible light

transmittance value, the greater the film's transparency. Higher opacity values indicate good light barrier properties. Li et al. [54] demonstrated that the opacity of composite films containing ferulic acid was higher than that of the control film made from chitosan and sodium alginate. These results indicate that the addition of ferulic acid enhanced the interaction between chitosan and sodium alginate, resulting in a denser film structure and reduced transparency.

### 3.2. The Effect of Phenolic Acids on the Barrier Properties of Pectin Packaging Films

Edible films are characterised by possessing crucial barrier properties that can regulate the migration of gases such as oxygen, carbon dioxide, and water vapour. However, when water vapour permeability is high can cause significant weight loss due to moisture evaporation in post-harvest fruits and vegetables, affecting their quality decline and shortening their shelf life. Therefore, effective gas barrier properties are essential for edible films and coatings to address these issues [55]. These properties play a significant role in protecting food products quality from oxidation, moisture loss, and other processes that can negatively impact their shelf life and quality during storage [56]. Table 3 contains the results for oxygen and carbon dioxide permeabilities of edible films obtained from apple pectin (AP) and selected phenolic acids. The oxygen permeability for the apple pectin film was  $0.84 \pm 0.02 \times 10^{-16}$  g/m·s·Pa, and for carbon dioxide, it was  $1.15 \pm 0.07 \times 10^{-16}$  g/m·s·Pa. The highest values for the pectin film with added phenolic acid were observed for the variant with apple pectin and sinapic acid:  $1.65 \pm 0.10$  for oxygen and  $2.33 \pm 0.12 \times 10^{-16}$  g/m·s·Pa for carbon dioxide. The increase in permeability of both gases may be caused by the disruption of the film's polymer structure resulting from the addition of sinapic acid. Caffeic acid is characterised by low stability in the presence of oxygen [57]. Sun et al. [58] investigated the oxygen permeability of edible chitosan films with added gallic acid. The introduction of gallic acid improved oxygen permeability. The high value was probably due to the presence of non-crosslinking gallic acid molecules dispersed in the film, which reduced intermolecular forces between polymer chains and led to pore formation.

**Table 3.** The oxygen ( $O_2P$ ) and carbon dioxide ( $CO_2P$ ) permeability of packaging films incorporated with selected phenolic acids: CFA—caffeic acid, CMA—coumaric acid, FRA—ferulic acid, GLA—gallic acid, PCA—protocatechuic acid, SNA—sinapic acid.

Film	$O_2P$ ( $\times 10^{-16}$ g/m·s·Pa)	$CO_2P$ ( $\times 10^{-16}$ g/m·s·Pa)
AP	$0.84 \pm 0.02^a$	$1.15 \pm 0.07^a$
AP_CFA	$0.46 \pm 0.03^a$	$0.70 \pm 0.01^a$
AP_CMA	$0.89 \pm 0.06^a$	$1.33 \pm 0.09^a$
AP_FRA	$0.92 \pm 0.12^a$	$1.26 \pm 0.17^b$
AP_GLA	$0.88 \pm 0.00^a$	$1.46 \pm 0.09^a$
AP_PCA	$0.45 \pm 0.08^b$	$0.60 \pm 0.09^b$
AP_SNA	$1.65 \pm 0.10^c$	$2.33 \pm 0.12^c$

Mean values  $\pm$  standard deviations. Different superscript letters (<sup>a-c</sup>) within the same column indicate significant differences between the films ( $p < 0.05$ ).

The permeability of both oxygen and carbon dioxide is essential because respiratory processes can affect the quality of fresh fruits, vegetables, and other products. Various factors play a role in the gas permeability of edible films, such as the integrity of the film, the balance between hydrophobic and hydrophilic areas, and the interactions between the polymers that comprise the film [7]. The presence of plasticisers or other additives also plays a role [59,60]. Moreover, in our previous study, we observed that the inclusion of phenolic acids increased the water vapour permeability from  $7.16 \pm 0.42 \times 10^{-10}$  for

control films to  $10.46\text{--}10.68 \times 10^{-10}$  g/m·s·Pa for films with coumaric, ferulic, and sinapic acids. However, films with gallic acid and protocatechuic acids showed similar values to control pectin films [39]. Those observations indicate that the presence of phenolic acid and its type are crucial in modifying the barrier properties of pectin films.

### 3.3. The Effect of Phenolic Acids on the Thermal Properties of Pectin Packaging Films

Thermal property assessment is essential to understanding the limitations of films in various applications. Films characterised by higher thermal stability can improve the integrity and functional properties of packaged foods, thus preserving them. Thermal stability assessment is most often performed using thermogravimetric analysis (TGA) [61]. These methods can reflect thermal behaviour, degradation rates, and variability in intermolecular interactions. Pectin films exhibit lower thermal stability due to the plasticising effect resulting from the hydration process. The addition of biopolymers with greater thermal stability may influence and modify the thermal parameters of pectin-based films [62]. Thermogravimetric analysis, on the other hand, can provide information on the thermal stability and can provide information about the degradation profiles of biopolymer-based films. This allows for the assessment of how functional compounds, such as plasticisers or bioactive substances, affect the thermal properties or overall stability of the film [63]. Table 4 presents the percentage weight loss of edible films during heating.

**Table 4.** Temperatures and weight losses related to stages of TG/DTG curves of packaging films incorporated with selected phenolic acids: CFA—caffeic acid, CMA—coumaric acid, FRA—ferulic acid, GLA—gallic acid, PCA—protocatechuic acid, SNA—sinapic acid.

Film	30–100 °C		100–280 °C		280–600 °C	
	°C	%	°C	%	°C	%
AP	63.59	4.27	178.92 226.27	48.95	331.92	19.06
AP_CFA	60.44	4.45	182.01 220.00	52.16	349.44	16.32
AP_CMA	58.75	5.00	183.13 219.74	53.23	351.57	16.64
AP_FRA	56.19	6.95	179.02 221.59	50.19	349.03	17.29
AP_GLA	61.55	5.68	180.53 214.77	52.78	345.23	16.03
AP_PCA	54.88	4.37	188.28 220.07	52.41	354.28	16.94
AP_SNA	58.52	4.94	187.55 220.74	54.79	309.54	15.45

The film obtained from apple pectin exhibited the lowest percentage weight loss in the first two heating stages (30–100 °C and 100–280 °C), but the highest weight loss in the final heating stage at the highest temperature. The reduced weight loss of films containing added phenolic acids suggests increased hydrophobicity, a finding supported by the results on moisture content and water solubility [39]. For all films, the highest weight loss occurred in the second heating stage. The primary weight loss during the second heating stage can be attributed to thermal decomposition of polymers with low-molecular-weight. Additionally, glycerol evaporation and decomposition of polysaccharide chains are likely to have occurred [64]. However, according to Giz et al. [65], the dense network structure formed by cross-linking can increase the energy that is required to rupture the molecular chains. This phenomenon slows the rate that characterises thermal degradation and enhances the film's thermal stability. Glycerol and sorbitol have been shown to have no significant effect on the thermal properties of the films when compared to plasticisers,

materials with no plasticisers, or with other plasticisers. Yerramathi et al. [66] demonstrated that the addition of ferulic acid increased the thermal stability of the films, likely due to the interaction of polyphenols with sodium alginate. This enhancement was probably due to the presence of hydrogen bonds, as well as a denser film network structure.

Investigating the thermal properties of the packaging films is crucial, as they may undergo various processes at elevated temperatures during packaging, transport, and storage [67]. The thermal properties of analysed films with selected phenolic acids were examined using thermogravimetric analysis to quantitatively measure the change in sample mass with increasing temperature (Figure 2). The first stage of thermogravimetric analysis, in the temperature range of 25 °C to 200 °C, is due to the evaporation of water as well as volatile components. Pectin, has side chains that responsible for steric hindrance with adjacent molecules, affecting higher intermolecular distances. This minimise molecule-molecule interactions and lowers degradation temperatures [68]. Cruz et al. [69] observed a pectin film mass loss of approximately 12% and 5% in the first and second stages, occurring in the temperature ranges of 10–136 °C and 136–204 °C, respectively. These stages were associated with water evaporation and the release of low-molecular-weight substances, such as volatile compounds. Moreover, the third stage (204–308 °C) was responsible for a greater mass loss, resulting from depolymerisation and polysaccharide degradation.

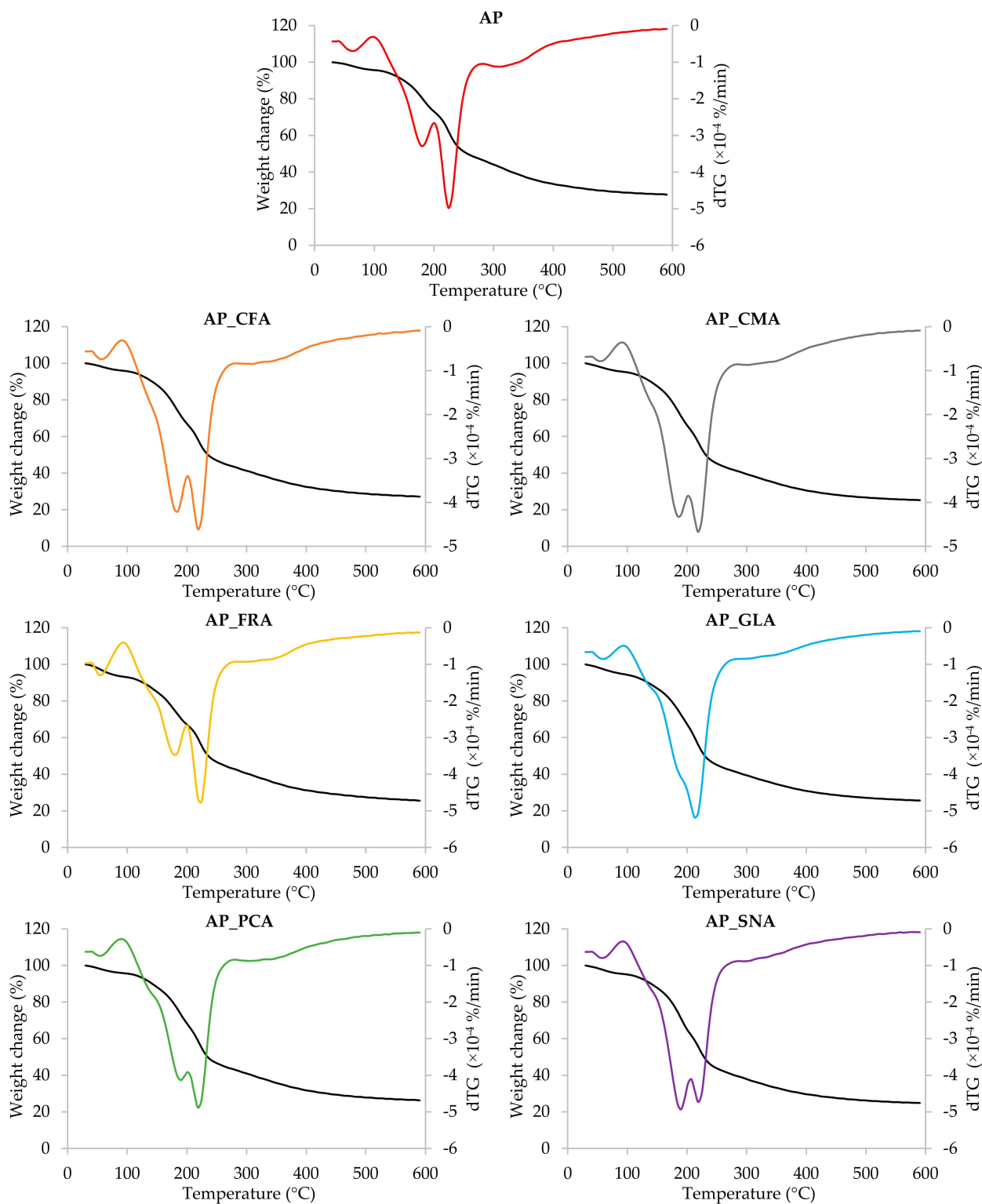
The low thermal stability of packaging materials hinders their use in packaging material. The thermal stability of biopolymer films is crucial for maintaining continuous structure and limiting degradation at various temperatures that may occur during processing, transport, and storage, which translates into protecting food quality and safety. Investigating the interactions between processing conditions and thermal behaviour supports the development of stable edible films. Furthermore, this allows for the preservation of the structural integrity and functionality of films [61]. Thermal degradation of polysaccharides up to approximately 200 °C results in the removal of bound and unbound water molecules (dehydration). This is succeeded by the breaking apart and separation of hydrocarbon chains, resulting in a notable reduction in weight and the creation of volatile products [70].

### 3.4. The Effect of Phenolic Acids on the Structural Properties of Pectin Packaging Films

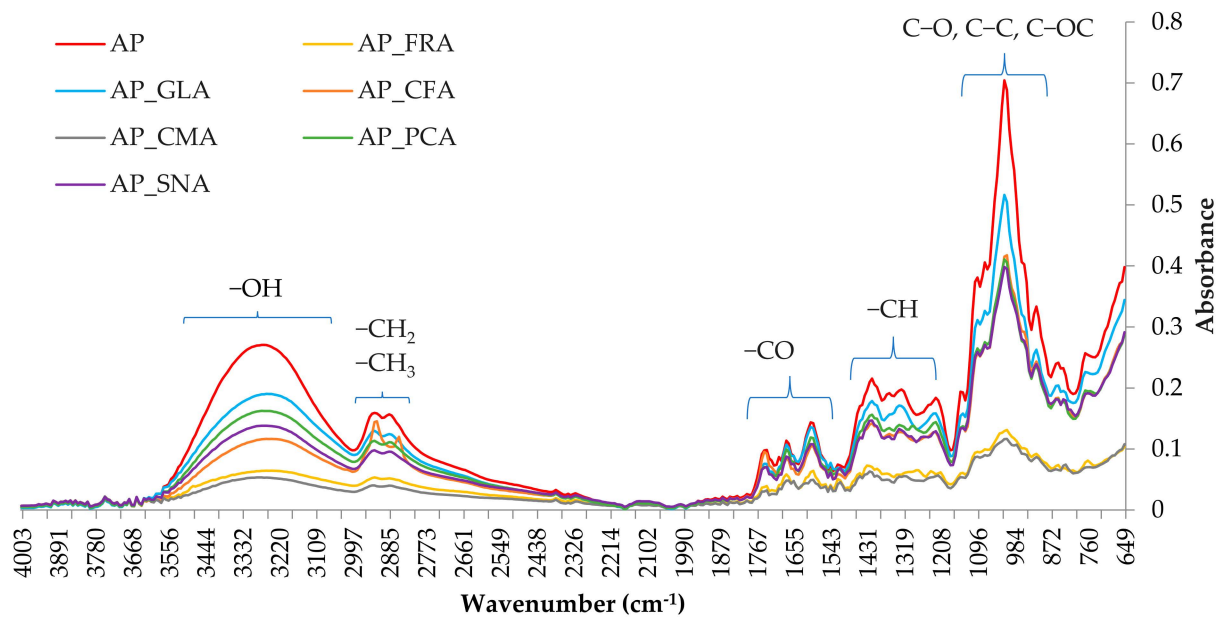
Fourier transform infrared spectroscopy (FT-IR) is based on the interaction of infrared radiation with matter. Infrared radiation is absorbed by functional groups present in the analyte, influencing the vibrations of covalently bonded atoms. This method enables the identification of specific functional groups (hydroxyl, carbonyl, and ether), which are important for predicting the chemical behaviour and interactions of films [63]. Furthermore, it is possible to analyse chemical changes occurring during processing, assess the degree of cross-linking, and monitor interactions with other compounds that may influence the changes in barrier, optical and mechanical properties of films [71]. FT-IR analysis was performed to investigate functional and intermolecular interactions. Furthermore, these interactions were assessed by incorporating phenolic acids into edible film formulations. This analysis identified specific wavelengths assigned to different functional groups in the 4000–650  $\text{cm}^{-1}$  spectrum. The results are presented in Figure 3.

The transmittance of the films showed important variations in the UV range (403–649 nm). The introduction of ferulic and coumaric acids led to a significant attenuation of UV radiation, indicating that their addition effectively enhanced the UV-blocking properties of the films. However, the inclusion of gallic acid significantly reduced transmittance of visible light, which increased light scattering and reflection, and consequently increased the opacity value. The main peaks in the spectral range of 3220–3332  $\text{cm}^{-1}$  characterise the stretching of hydroxyl groups. Cabrera-Barjas et al. [72] described characteristic bands

between 3000 and 3700  $\text{cm}^{-1}$  as associated with  $-\text{OH}$  groups, between 3000 and 2800  $\text{cm}^{-1}$  with  $-\text{CH}$  groups, and around 1640  $\text{cm}^{-1}$  as attributed to  $\text{C}=\text{C}$  groups. Moreover, the area between 1455 and 1400  $\text{cm}^{-1}$  is characteristic of the bending vibration of  $-\text{CH}$  groups, and bands in the region between 1150 and 1000  $\text{cm}^{-1}$  are related to the antisymmetric stretching vibrations of the  $\text{C}-\text{O}-\text{C}$  bridge and  $\text{C}-\text{O}$  groups.



**Figure 2.** TGA (black) and (dTG) (coloured) curves of packaging films incorporated with selected phenolic acids: CFA—caffeic acid, CMA—coumaric acid, FRA—ferulic acid, GLA—gallic acid, PCA—protocatechuic acid, SNA—sinapic acid.



**Figure 3.** The Fourier transform infrared spectra of packaging films incorporated with selected phenolic acids: CFA—caffeic acid, CMA—coumaric acid, FRA—ferulic acid, GLA—gallic acid, PCA—protocatechuic acid, SNA—sinapic acid.

Serrafi et al. [73] discovered that the shift in O-H vibration bands in pectin to lower frequencies is due to the production of hydrogen bonds and alterations in the chemical environment of the hydroxyl groups. The formation of hydrogen bonds between the hydroxyl group (O-H) and other functional groups reduces the energy required to stretch the O-H bond, resulting in a shift in the absorption band toward lower frequencies. Additionally, a lower amount of water molecules associated with pectin may also contribute to this change. In conclusion, films that contain phenolic acids may have lower hydration levels than the apple pectin film sample. The study by Bhatia et al. [60] showed that the peak observed at  $2923\text{ cm}^{-1}$  corresponded to C-H vibrations, specific to the presence of glycerol. However, Chaves et al. [68] reported that intermolecular interactions, particularly hydrogen bonds between polymers, plasticiser, and water, broaden and shift the bands characteristic of the absorption maximum. Dobrucka et al. [74] also observed a significant peak at  $1000\text{ cm}^{-1}$ , which was attributed to the stretching of the saccharide structure of pectin in the C-O-C direction.

### 3.5. The Effect of Phenolic Acids on the Mechanical Properties of Pectin Packaging Films

Assessing the mechanical properties of films is important because it indicates their durability and ability to maintain food integrity during handling, transport, and storage. The tensile strength parameter indicates the maximum elongation of the film when stretched [1,75]. An ideal edible packaging film should exhibit flexibility and elasticity, conforming to the shape of the packaged food while also being resistant to mechanical abrasion [76]. The higher molecular weight and dry matter of pectin is responsible for longer polymer chains that increase intermolecular entanglement, as well as film-forming solution viscosity. Compared to protein films, they typically exhibit good tensile strength but lower elasticity [25]. The mechanical resistance of the films was evaluated by characterising tensile strength, elongation, and Young's modulus, with the results shown in Table 5. The values of tensile strength for films ranged from 2.78 (AP) to 4.45 MPa. The addition of phenolic acids increased the tensile strength, with significantly higher values observed in films containing caffeic, protocatechuic, and sinapic acids. A plasticising effect of the phenolic acids was noted for caffeic, ferulic, and protocatechuic acids, leading to a

significant increase in elongation at break from  $9.71 \pm 1.16\%$  to a range of 12.51–15.07%. Furthermore, the Young's modulus values for all films containing phenolic acids were significantly higher, ranging from 5.28 to 5.98 MPa, compared to the control films, which had a Young's modulus of  $3.48 \pm 0.73$  MPa.

**Table 5.** The tensile strength (*TS*), elongation at break (*E*) and Young Modulus (*YM*) of packaging films incorporated with selected phenolic acids: CFA—caffeic acid, CMA—coumaric acid, FRA—ferulic acid, GLA—gallic acid, PCA—protocatechuic acid, SNA—sinapic acid.

Film	<i>TS</i> (MPa)	<i>E</i> (%)	<i>YM</i> (MPa)
AP	$2.78 \pm 0.40^a$	$9.71 \pm 1.16^a$	$3.48 \pm 0.73^a$
AP_CFA	$4.09 \pm 0.91^{bc}$	$12.51 \pm 1.83^b$	$5.52 \pm 0.68^b$
AP_CMA	$3.37 \pm 0.63^{ab}$	$9.55 \pm 1.62^a$	$5.33 \pm 0.63^b$
AP_FRA	$3.61 \pm 0.52^{abc}$	$15.07 \pm 1.74^c$	$5.82 \pm 0.69^b$
AP_GLA	$3.32 \pm 0.65^{ab}$	$9.71 \pm 1.45^a$	$5.28 \pm 0.94^b$
AP_PCA	$4.45 \pm 0.66^c$	$12.51 \pm 1.67^b$	$5.47 \pm 0.58^b$
AP_SNA	$2.92 \pm 0.40^a$	$9.38 \pm 1.80^a$	$5.98 \pm 0.58^b$

The same superscript letters within the same column (<sup>a-c</sup>) indicate no significant differences between the samples ( $p < 0.05$ ).

Gupta et al. [77] characterise ferulic acid by its ability to increase the mechanical strength and barrier properties of films, in addition to its strong antioxidant properties. Kaczmarek et al. [78] reported that adding ferulic acid to collagen films significantly increased mechanical resistance. An improvement in the tensile properties of chitosan–alginate films was also observed with the addition of ferulic acid [79]. Furthermore, ferulic acid was found to play a key role as a compatible agent and enhance the physical, chemical, and biological properties of the films. Wu et al. [80] reported that the increased tensile strength of films prepared with chitosan and gallic acid was attributed to cooperative cross-linking. This cross-linking results from the formation of hydrogen bonds between gallic acid and the chitosan chain, which influences the structure of the resulting films. Woranuch et al. [81] reported that the development of chitosan films with ferulic acid showed minimal impact on tensile strength and elastic modulus. However, elongation at break increased to 16.24%, compared to control films, which had a tensile strength of 19.09 MPa, an elastic modulus of 1103.72 MPa, and an elongation at break of 10.95%. This phenomenon can be attributed to the plasticising effect of small phenolic acid molecules, which made the film more flexible after the addition of ferulic acid. Yerramathi et al. [62] developed sodium alginate films with the addition of ferulic acid and also observed an improvement in mechanical properties. The addition of phenolic acids (ferulic acid, caffeic acid, tannic acid, and gallic acid) also improved the mechanical properties of chitosan films [82]. Liu et al. [83] observed an improvement in the mechanical properties of the films with the introduction of gallic acid, increasing tensile strength (TS) from 6.00 MPa to 15.97–20.06 MPa and elongation at break from 3.49% to 6.37–7.29%. Other researchers also observed a significant improvement in mechanical strength obtained through the formation of hydrogen bonds and covalent interactions with pectin molecules, resulting in a denser and stiffer polymer network. The use of tannic acid increased the tensile strength of the films by 40.14% compared to films made with pure pectin. This increased mechanical strength is closely related to the film's barrier properties, as it reduces water vapour permeability. Furthermore, it enhances the protective effect when used for perishable foods [84,85]. Improved mechanical strength of pectin-based films was also obtained by adding tannic acid, which influences the formation of hydrogen bonds and covalent interactions with pectin molecules, resulting in a denser and stiffer polymer network [25]. Liu et al. [49]

observed increased tensile strength and elongation at break in edible chitosan films with the addition of coumaric acid. This phenomenon was likely due to an appropriate grafting rate, which could have reduced molecular speed and free volume of the chains, thus creating a denser film structure.

### 3.6. The Effect of Phenolic Acids on the Antioxidant Properties of Pectin Packaging Films

Introducing phenolic acids into the pectin structure offers the possibility of obtaining new matrices with satisfactory antioxidant properties and extending the functional properties of pectin-based films. Antioxidant capacity has been widely studied using common assays such as electron or radical capture detection, known as the DPPH (2,2-diphenyl-1-picrylhydrazyl) assay and the ABTS (2,2'-azinobis-3-ethylbenzothiazoline-6-sulfonic acid) assay. Direct measurement of a film formulation requires first isolating (extracting) the antioxidant from the matrix (material of coating or film) using an appropriate solvent [86]. The results for antioxidant activity for composite pectin films are in Table 6. The ABTS free radical scavenging capacity in the tested edible films ranged from  $0.36 \pm 0.12$  for control films to  $38.08 \pm 0.69$  mg TE/g d.m for films containing ferulic acid. The antioxidant activity against DPPH radical ranged from 0.83 mg TE/g d.m. for the control films to 131.48 mg Trolox/g d.m. for the films containing ferulic acid. In general, all types of phenolic acids affected the antioxidant activity of pectin films. Phenolic compounds, naturally occurring and extracted from plants, exhibit various properties, such as antioxidant anti-inflammatory effects, as well as anticancer property. They are effective antibacterial agents due to their ability to destabilise bacterial membranes and inhibit protein synthesis in microbial cells [87,88]. Therefore, the addition of phenolic acids may enhance both antioxidant and antibacterial properties. Conversely, edible films created from pectin offer antioxidant benefits and help slow down the decline of various types of methanogens, thereby maintaining of food quality and safety [18]. In general, antioxidants convert the stable free DPPH radicals, which are purple, into their reduced form, DPPH-H, which are yellow. The ability to scavenge the DPPH radical is closely related to the antioxidant's capacity to donate hydrogen [89,90]. According to Dey et al. [91], the antioxidant capacity of phenolic acids correlates with their ability to donate hydrogen through the phenolic hydroxyl group, thereby terminating the radical oxidation of lipids or other biomolecules.

**Table 6.** The antioxidant activity of pectin films incorporated with caffeic (CFA), coumaric (CMA), ferulic (FRA), gallic (GLA), protocatechuic (PCA) and sinapic (SNA) acids.

Film	ABTS (mg TE/g d.m.)	DPPH (mg TE/g d.m.)
AP	$0.36 \pm 0.12^a$	$0.83 \pm 0.02^a$
AP_CFA	$10.13 \pm 0.45^b$	$12.65 \pm 0.36^a$
AP_CMA	$18.60 \pm 0.91^c$	$62.66 \pm 3.51^c$
AP_FRA	$38.08 \pm 0.69^f$	$131.48 \pm 7.57^e$
AP_GLA	$17.38 \pm 0.23^c$	$39.84 \pm 0.84^b$
AP_PCA	$23.13 \pm 0.23^d$	$57.99 \pm 0.04^c$
AP_SNA	$32.64 \pm 0.42^e$	$93.47 \pm 5.69^d$

Mean values  $\pm$  standard deviations. Different superscript letters (<sup>a-f</sup>) within the same column indicate significant differences between the films ( $p < 0.05$ ).

The DPPH activity of hydroxycinnamic acids is often presented in the following order: caffeic acid > sinapic acid > ferulic acid > coumaric acid [92]. The variations in the outcomes of this study could be linked to the distinct structures of phenolic acids and how well they interact and blend with the pectin matrix during the formation of the film. Thus, although the presence of two aromatic hydroxyl groups in caffeic acid is responsible for higher scavenging activity compared to ferulic acid, which contains a single hydroxyl group, this

did not affect the results in the case of pectin films [93]. The presence of a CH=COOH group in cinnamic acid derivatives contributes to greater antioxidant activity than the COOH groups in benzoic acids [94].

#### 4. Conclusions

This study showed that various phenolic acids are suitable compatible materials for apple pectin-based films. Each film exhibited a consistent, uniform structure along with a slight yellow tint. UV-VIS analysis revealed that the control film achieved the highest transmittance value. However, incorporating phenolic acids resulted in a reduction in this value, enhancing the film's effectiveness as a barrier against UV-VIS light. The incorporation of phenolic acids affected lower values of lightness (parameter  $L^*$ ) in all film variants. For all films, the highest mass loss was observed in the second heating stage, ranging from 100 to 280 °C. The lower mass loss of films containing phenolic acids, compared to pectin films, indicates their higher hydrophobicity. The analysis using Fourier transform infrared spectroscopy indicated that there were no interactions that may occur between the phenolic acids and the film matrix. It showcased the functional groups found in pectin, including carbonyl (C=O) and hydroxyl (–OH) groups. Phenolic acids enhanced both the mechanical strength and antioxidant properties of the pectin films. Consequently, it can be noted that the resulting edible films possess significant potential when used for food products.

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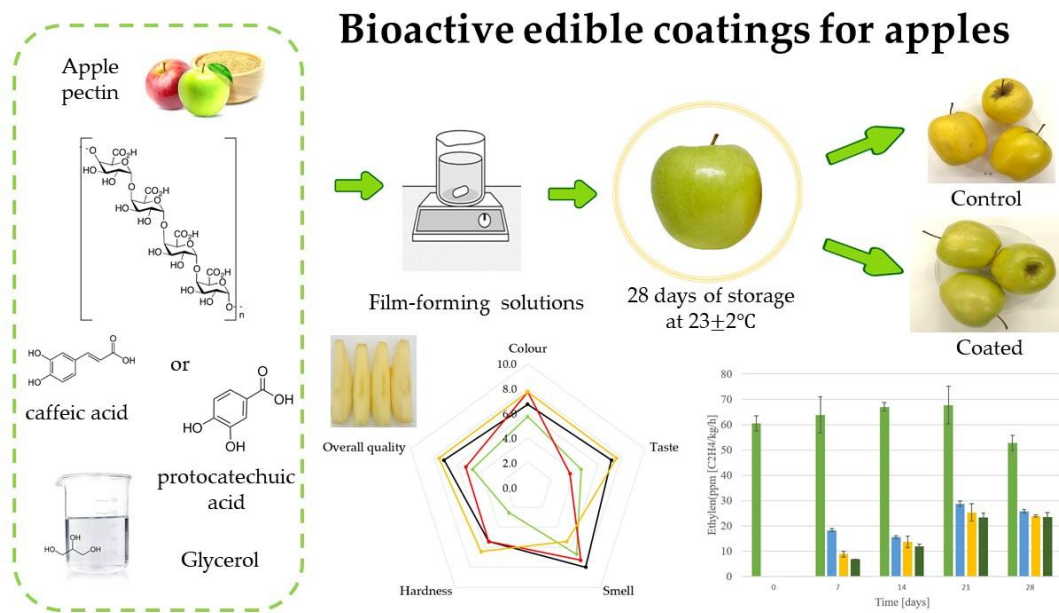
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# Publikacja 4

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## Abstrakt graficzny



## Article

# The Use of Bioactive Edible Coatings Based on Pectin and Phenolic Acids for Enhancing Quality Attributes of Golden Delicious Apples During Storage

Magdalena Mikus, Karolina Szulc  and Sabina Galus \* 

Warsaw University of Life Sciences, Institute of Food Sciences, Department of Food Engineering and Process Management, 02-776 Warsaw, Poland; magdalena\_mikus@sggw.edu.pl (M.M.); karolina\_szulc1@sggw.edu.pl (K.S.)

\* Correspondence: sabina\_galus@sggw.edu.pl

## Abstract

This research study investigated the effect of edible coatings made from apple pectin, incorporating caffeic and protocatechuic acids, on the quality attributes of Golden Delicious apples during 28 days of storage at ambient conditions. The study evaluated the rheological properties of the coating solutions, the release of phenolic acids from the edible films, and various quality characteristics of the apples. These characteristics included weight loss, colour, total soluble solids, total titratable acidity, pH, firmness, respiration rate, ripeness level, and sensory analysis. The results showed that all coating solutions exhibited non-Newtonian, shear-thinning flow behaviour, with the sample containing protocatechuic acid demonstrating a decrease in apparent viscosity. Additionally, both phenolic acids were released rapidly from the film into a 96% ethanol medium. The study found that bioactive edible coatings, both with and without phenolic acids, were significantly effective in reducing weight loss, colour changes, firmness, and ripening of apples during storage. The total soluble solids were higher in control apples ( $14.95 \pm 0.48$  °Brix at 28 days) compared to the coated samples (13.52–13.53 °Brix at 28 days), indicating that the control apples were riper and contained a higher amount of sugars. Ethylene production decreased after 4 weeks of storage, from 60.40 ppm for the apples before storage to 23.55–25.70 ppm for the coated samples, and only to 52.75 ppm for the control apples. Overall, this study confirmed that the use of developed bioactive coatings extends the shelf life of apples by preserving their quality and sensory attributes during storage.

**Keywords:** edible coatings; Golden Delicious apple; apple pectin; phenolic acids; colour changes; firmness; ripening; sensory analysis



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## 1. Introduction

Effectively managing post-harvest fruit poses numerous challenges that must be addressed to preserve fruit quality during extended storage under various conditions [1]. Apples are classified as climacteric fruits, meaning they continue to ripen after being harvested. This ongoing metabolic activity makes them sensitive to quality loss. To preserve their desired quality, apples are typically stored in cold storage or in a controlled atmosphere for long-term storage [2,3]. Apples are typically harvested in August and September, but they are available for sale and consumption throughout the entire year. To ensure that consumers have access to apples during the off-season, they are often stored for 4 to 6 months [4]. This is particularly advantageous in regions where fruit is only available

or cultivated during specific seasons. To address the challenge of storing and preserving apples after harvest, it is essential to develop a new preservation method, particularly for fruits stored at room temperature. Cold storage can negatively affect the texture, flavour, and colour of fruit. Thus, edible coating offers a better solution by protecting against gases, moisture, pathogens, and light [5].

In recent years, there has been a growing interest in environmentally friendly edible coatings that can extend the shelf life of fresh fruits and vegetables [6–8]. Edible coatings are thin layers of material applied to products, typically through immersion [9]. These coatings act as a semi-permeable membrane, providing protection against moisture, oxygen, and carbon dioxide. They can help reduce the rate of oxidation reactions, respiration, and moisture loss in the products, thereby prolonging their storage period [10,11]. Apples, due to their biological activity after harvest, become increasingly susceptible to infections caused by pathogenic fungi, such as *Penicillium expansum*, *Penicillium digitatum*, or *Botrytis cinerea*, as they ripen and age. These fungi are responsible for post-harvest decay [12–14]. Commercial storage can negatively impact the emission of apple aroma, which may persist throughout the retail chain, resulting in poor consumer acceptability [15]. The use of coatings reduces undesirable changes in fruit, preserving a fresh appearance that is characterised by good quality and sensory appeal [16]. Research indicates that nearly 71% of fruit is consumed fresh, while around 20% is processed into various value-added products [10]. Effective storage is essential to reduce post-harvest losses of fresh apples, which can reach 25–28%. New solutions need to be developed to extend the shelf life of fruits and increase the food supply [17]. Edible coatings have emerged as a highly effective and safe technology for preserving fresh fruits. They work by limiting gas exchange, which delays the ageing process of fruits during storage [6,18,19]. Edible coatings containing antimicrobial compounds, such as plant extracts or essential oils, inhibit the growth of microorganisms that can lead to the spoilage of fruits [20]. Many coatings are enriched with antimicrobial compounds or chitosan, which directly inhibit bacterial and fungal growth by disrupting cell membranes or carrying out other direct antimicrobial actions [21].

Edible coatings can be made from proteins, polysaccharides, lipids, or a combination of these ingredients. Additionally, edible coatings can serve as carriers for various food additives, including antimicrobial agents, anti-browning agents, antioxidants, as well as dyes, flavours, nutrients, and spices [22,23]. The main purpose of applying edible coatings to fresh fruit is to reinforce the existing natural barrier or replace it where it has been partially removed. Other benefits of edible coatings include improved mechanical properties by maintaining the structural integrity of coated products. By allowing the coatings to be consumed with fruit, it is possible to reduce packaging waste [24]. However, the disadvantages include the formation of an unpleasant aftertaste if the coating is too thick, as well as the occurrence in some cases of a hygroscopic nature, which favours the growth of microorganisms [25]. Different types of polysaccharides, like pectin, are commonly used to develop edible coatings for the preservation of various fruits. Pectin coatings offer excellent resistance to moisture and gases and are highly transparent, helping to preserve the sensory qualities and overall quality of the fruit [5,26]. Edible coatings that contain lipids can substantially reduce moisture loss in fruits and contribute to a lighter skin colour. These coatings serve as semipermeable membranes, which limit the flow of gases and water vapour [27]. As a result, they slow down the rates of respiration and moisture loss in fruits, helping to maintain fruit quality and delay physiological deterioration after harvest [28]. The estimated growth in biopolymer production, the main components of edible coatings, is expected to reach up to 2.41 million tons by 2030 [5]. The rising popularity of using natural polymers for coating and preserving food can be attributed to their potential benefits and the fact that they can be safely consumed alongside

fruits. Furthermore, unlike synthetic or petroleum-based coatings, natural polymers are environmentally friendly and help reduce packaging waste, aligning with the principles of sustainable development [21,29].

*Malus domestica* Borkh., known as Golden Delicious apples, is among the most commonly consumed and cultivated apple varieties in temperate regions of the world. They are valued for their shape, texture, nutritional benefits, and excellent taste [30,31]. They are prone to post-harvest softening because they ripen in summer [32]; however, for optimal storage, the conditions of 0–4 °C and 90–95% relative humidity, with a maximum storage duration of 2 to 6 months, are indicated [33]. Their storage life is estimated to be approximately four weeks [17]. Proper storage of apples is essential for economic processes, as it minimises food waste and promotes more sustainable food consumption [34].

Pectin is an effective coating material for protecting the quality of fresh apples. It can also serve as a carrier for functional ingredients, thereby enhancing antioxidant and antimicrobial properties [35]. Pectin's amphiphilic properties can improve stability by reducing surface tension. The presence of methoxy groups influences the hydrophobic nature of pectin, endowing it with surface-active properties. Furthermore, coating fresh fruit with edible coatings helps maintain firmness due to the coatings' antagonistic effect on microflora, which can contribute to the progressive softening of tissue [36]. Studies have shown that edible coatings and films are more effective in extending the shelf life of fresh produce when formulated as composite formulations with other ingredients, rather than using a single component [28]. Various coatings were applied to fresh apples to extend their shelf life. De León-Zapata et al. [37] observed that applying a candelilla wax-based edible coating, which included fermented tarbush extract as a natural antioxidant source, positively affected the quality and shelf life of Golden Delicious apples. Rashid et al. [38] demonstrated that composite coatings containing 2.5 g of fenugreek and 1.5 g of flaxseed polysaccharides, combined with stearic acid, monoglycerides, and canola oil, were the most effective for maintaining fruit mass, firmness, total soluble solids, acidity, and pH of apples. The effectiveness of these coatings can be attributed to their ability to delay the respiration rate and inhibit ethylene production in the fruit. Soppelsa et al. [9] observed that thyme and clove essential oils, when encapsulated in chitosan coatings, effectively control postharvest diseases of apple fruits stored at 20 °C and 100% relative humidity. Conversely, the quality of fresh-cut apples, which are prone to rapid deterioration, may also be extended by bioactive coatings. In this context, Nicolau-Lapeña et al. [39] noted the beneficial effect of the incorporation of ferulic acid into sodium alginate-based coatings in a reduction in browning and the population of *Listeria monocytogenes* after 7 days of refrigerated storage of apple slices.

The findings indicate that encapsulated active ingredients can be a valuable tool for managing apples after harvest. However, before widespread application, we must further investigate the economic sustainability of available biopolymers and their potential negative impact on fruit aroma and taste. Phenolic acids act as antioxidants by neutralising free radicals, thereby protecting against oxidative damage. Their antioxidant properties stem from a hydroxyl group attached to an aromatic ring, which allows them to donate a hydrogen atom to stabilise free radicals [40]. Abundant in fruits, vegetables, and beverages, they are considered a major class of natural antioxidants in the human diet and have potential applications in medicine, cosmetics, and the food industry [41]. Caffeic acid is a promising antioxidant that can be incorporated into edible coatings to protect food by scavenging free radicals, chelating metal ions, and preventing lipid oxidation. Research indicates that the addition of caffeic acid enhances antioxidant activity, UV protection, and antimicrobial properties in films composed of biopolymer-based materials [42]. Protocatechuic acid is another effective antioxidant compound for edible packaging, enhancing the film's ability

to protect food from oxidative damage and serving as a UV light barrier [43]. The addition of phenolic acids to film-forming solutions can improve properties like flexibility, while also helping to extend the shelf life of different food products by delaying spoilage.

The objective of this study was to preserve the quality of fresh Golden Delicious apples by applying a pectin coating that included selected phenolic acids, specifically caffeic and protocatechuic acids, during storage at room temperature. To assess the effectiveness of the edible coating treatment, several parameters were measured over a 28-day period while storing the apples at room temperature. These parameters included changes in weight, colour, total soluble solids, titratable acidity, pH, firmness, respiration rate, degree of ripeness, and sensory attributes. Further research is required to investigate the combination of caffeic acid and protocatechuic acid formulations and determine any additive or synergistic effects on antioxidant or antimicrobial activity, including release kinetics. It may also be important to determine the likelihood of antagonistic interactions resulting from the combination of these two phenolic acids.

## 2. Materials and Methods

### 2.1. Materials

Apple pectin (Pektowin S.A., Jasło, Poland) and caffeic (Pol-Aura Sp. z o.o., Zawroty, Poland) or protocatechuic acids (Thermo Scientific, Gdańsk, Poland) were used for the production of aqueous coating solutions. Glycerol (Avantor Performance Materials Poland S.A., Gliwice, Poland) was used as a plasticising agent. Golden Delicious apples from the experimental orchards of the Warsaw University of Life Sciences were harvested in 2024 and used for experiments.

### 2.2. Preparation of Coating Solutions

The selected phenolic acids were combined with apple pectin before hydration at a concentration of 5% (acids relative to pectin). The solutions were heated at 60 °C for 20 min using an RCT basic IKAMAG magnetic stirrer (IKA Poland, Warsaw, Poland) with a rotation speed of 600 rpm to obtain a uniform solution. After cooling the solutions, glycerol was added at a concentration of 50% relative to apple pectin (2.5 g). For the experiments, three coating solutions based on apple pectin were used: one without phenolic acids, coded AP; one with caffeic acid, coded AP\_CFA; and one with protocatechuic acid, coded AP\_PCA. Distilled water was used instead of the coating solution for control samples. The characteristics of the substances used and their proportions are presented in Table 1.

**Table 1.** Characteristics of the substances used to obtain film-forming solutions.

Substance	Quantity (g)
Apple pectin (AP)	5
Caffeic acid (CFA)	0.25
Protocatechuic acid (PCA)	0.25
Glycerol (GLY)	2.5
Water	92.25

### Rheology of Coating Solutions

A Haake MARS 40 rheometer (Thermo Scientific Inc., Waltham, MA, USA) was used to study the flow behaviour of the solutions at 25 °C in triplicate in a coaxial cylinder

system (CC25DIN/Ti) with a linearly increasing shear rate up to  $100 \text{ s}^{-1}$ . The flow curves were fitted using the Ostwald de Waele model [44]:

$$\tau = K \cdot \dot{\gamma}^n$$

where  $\tau$  is the shear stress (Pa),  $\dot{\gamma}$  is the shear rate ( $\text{s}^{-1}$ ),  $K$  is the consistency index ( $\text{Pa} \cdot \text{s}^n$ ), and  $n$  is the dimensionless flow behaviour index.

### 2.3. Film Preparation

The films were obtained based on the method described in our previous study [45]. Briefly, the coating solutions were poured onto sheets at a speed of  $10 \text{ mm/s}$  and a layer thickness of  $2500 \text{ }\mu\text{m}$  using a Zehntner ZAA 2300 automatic film applicator (Zehntner GmbH Testing Instruments, Sissach, Switzerland) and dried in a laboratory dryer SUP-65W (Wamed, Warsaw, Poland) for 24 h at  $50 \text{ }^\circ\text{C}$ . The obtained films were conditioned in a KFB 240 thermostatic chamber (Binder, Tuttlingen, Germany) at  $25 \text{ }^\circ\text{C}$  and 50% relative humidity for 48 h prior to testing.

#### 2.3.1. Film Thickness

The film thickness was determined at least in three replicates, and the values were used to determine the kinetics of phenolic acid release. A thickness tester, ProGage (Thwing-Albert, West Berlin, NJ, USA), with an accuracy of  $1 \text{ }\mu\text{m}$ , was used.

#### 2.3.2. The Release Kinetic Measurements of Phenolic Acids

Phenolic acid release was conducted in three replicates using a 96% ethanol solution (Chempur, Piekary Śląskie, Poland) based on the method described by Benbettaieb et al. [46]. Developed films dissolved instantly in water, so 96% ethanol was used to evaluate release over time. Film samples weighing approximately  $60 \pm 5 \text{ mg}$  were placed in beakers, which were then filled to a total volume of  $100 \text{ mL}$  with ethanol. The prepared samples were stirred at  $150 \text{ rpm}$  using an RCT basic IKAMAG magnetic stirrer (IKA Poland, Warsaw, Poland). Absorbance measurements were taken using an Evolution 220 UV-Visible Spectrophotometer (Thermo SCIENTIFIC, Warsaw, Poland). For one hour,  $4 \text{ mL}$  of the solution containing the released substances was collected at specific time intervals: every minute for the first 10 min, and every 5 min for the remaining duration. The concentrations of phenolic acids in the samples were determined by UV-VIS spectrophotometry at wavelengths of  $310 \text{ nm}$  for caffeic acid and  $315 \text{ nm}$  for protocatechuic acid. After each measurement, the collected solution was returned to the beaker to maintain a constant volume. The calibration curve was prepared for each compound at concentrations from  $0.5$  to  $2.5 \text{ mg/100 mL}$  of 96% ethanol, giving the equations of  $y = 0.9637x + 0.2184$  ( $R^2 = 0.944$ ) for caffeic acid and  $y = 0.805x - 0.0511$  ( $R^2 = 0.973$ ) for protocatechuic acid. The obtained results were used to calculate the release of active substances using Fick's second law equation in the transient state [46,47]:

$$\frac{C_t}{C_\infty} = 1 - \sum_{n=1}^{\infty} \frac{2\alpha(1 + \alpha)}{1 + \alpha + \alpha^2 q_n^2} \exp\left(-\frac{Dq_n^2 t}{L^2}\right)$$

where  $C_t$  (mg/L) is the concentration of the active substance over time in the release medium;  $C_\infty$  (mg/L) is the concentration of the active substance at equilibrium in the release medium;  $D$  is the diffusion coefficient of the active substance ( $\text{m}^2/\text{s}$ );  $L$  is half the thickness of the film (m);  $q_n$  are non-zero, positive roots of  $\tan(qn) = -\alpha qn$  ( $n$  value from 1 to 6);  $\alpha$  is determined on the basis of  $\alpha = \frac{V_s}{K_{f,s} V_f}$ ;  $V_f$ , film volume ( $\text{m}^3$ );  $K_{f,s} = \frac{C_{f,\infty}}{C_{s,\infty}}$ : volume of the medium, and the partition coefficient of the active substance between the foil and the simulant (solution) in equilibrium;  $C_{f,\infty}$  and  $C_{s,\infty}$  are the equilibrium concentrations of the active substance (mg/L) in the film and the food simulant, respectively.

#### 2.4. Coatings of Apples

Apples of similar size and ripeness were washed with tap water, followed by drying using a paper towel. The apples were then immersed in coating solutions for 15 s, followed by a 5 s immersion in a 1% calcium chloride solution (Avantor Performance Materials Poland S.A., Gliwice, Poland) for pectin cross-linking. Control apples were immersed in distilled water for 20 s. Coated fruits were blotted to remove excess solution, placed on filter paper, and stored at room temperature ( $22 \pm 1$  °C) and  $40 \pm 5\%$  relative humidity for 28 days. Testing occurred at 7-day intervals. There were four variants of samples in three repetitions: control apples treated with water (Control), apples coated with solutions without phenolic acids (AP), and apples coated with solutions containing caffeic (AP\_CFA) and protocatechuic (AP\_PCA) acids. Each sample of the variant consisted of three apples per repetition, with an average weight of  $150 \pm 20$  g, for which separate analyses were performed at 0, 7, 14, 21, and 28 days post-treatment.

### 3. Physicochemical Analyses

#### 3.1. Weight Loss

The determination of weight loss in all apple samples was conducted for each variant before coating for the control sample and after coating for the other variants. The samples were weighed every seven days throughout the 28-day storage period. Weight loss was calculated by subtracting the final weight of the apples from their initial weight. All measurements were recorded using a semi-analytical balance (Radwag S.A., Radom, Poland). Weight loss was calculated using the following formula:

$$U_t = \frac{m_0 - m_t}{m_0} \times 100\%$$

where  $U_t$ —percentage weight loss after  $t$  days,  $m_0$ —initial mass (day 0), and  $m_t$ —mass on day  $t$ .

#### 3.2. Colour

Colour parameters were measured in two opposite places of three apples (six repetitions) using a Minolta CR-5 colourimeter (Konica Minolta, Tokyo, Japan) in the  $L^*$ ,  $a^*$  and  $b^*$  colour space. Measurements were performed with the standard observer  $2^\circ$  and D65 light source.

#### 3.3. Total Soluble Solids

The analysis of the total soluble solids in apples was performed in triplicate using the refractometric method by squeezing the juice from the fruits. The measurement was performed using a refractometer (PAL-3, Atago Instruments, Tokyo, Japan). The results were obtained in °Brix.

#### 3.4. Total Titratable Acidity

To determine the total titratable acidity (TTA) expressed as a percentage of malic acid, 10 mL of apple juice was measured into a beaker. Then, 100 mL of distilled water was added. Using a pipette, 25 mL of this diluted apple juice was transferred to a 50 mL beaker. A magnetic stirrer and electrode were placed in the solution for testing. The titration was carried out by adding a 0.25 mol/L NaOH solution until a pH of 8.1 was reached, which was designated as the neutralisation point. The titratable acidity was calculated in three repetitions using the appropriate formula.

$$TTA = \frac{(100 * V_1 * c)}{V_0}$$

where  $V_1$  = amount of solution used,  $V_0 = 25$  mL and  $c = 0.25$  mol/L NaOH.

### 3.5. pH

The fruit tissue was cut, ground, and filtered through a sieve to collect the juice. The pH of the fruit juice was measured in three repetitions using a pH meter (SHOTT Instruments, Lab 850, Warsaw, Poland) with three replicates.

### 3.6. Firmness

The texture of the apples was analysed in nine repetitions using a TA-TX2i texturometer (Stable Micro Systems Ltd., Haslemere, UK) equipped with Texture Expert software (version 2.3), which recorded the force values during the testing process. A penetration test was conducted using a 9 mm pin at a speed of 2 mm/s. Measurements were taken in six replicates, with two punctures made on opposite sides of three apples. The measure of firmness was determined by the maximum force expressed in newtons (N), calculated from the relationship between force and penetration time.

### 3.7. Respiration Rate

The emissions of ethylene ( $C_2H_4$ ) and carbon dioxide ( $CO_2$ ) from the fruits were analysed in duplicates over the course of one hour using the F-950 Analyser (Felix Instruments Inc., Camas, WA, USA). During the analysis, three apples were placed in a sealed 2L jar that was directly connected to the gas analyser, which allowed for an air space of 34  $\mu$ L for each measurement.

### 3.8. Sensory Analysis

The sensory analysis of apples was conducted after 7 and 28 days of storage. The apples were washed with water, then cut into pieces and coded accordingly. A sensory evaluation was conducted using a 5-point scale with a group of 40 trained participants, comprising students and staff from the Institute of Food Sciences at Warsaw University of Life Sciences in Poland. Participants were aged 20 to 45. The following qualitative characteristics were assessed: colour, taste, aroma, firmness, and overall fruit acceptability. Excel 2013 software was also used to process the results.

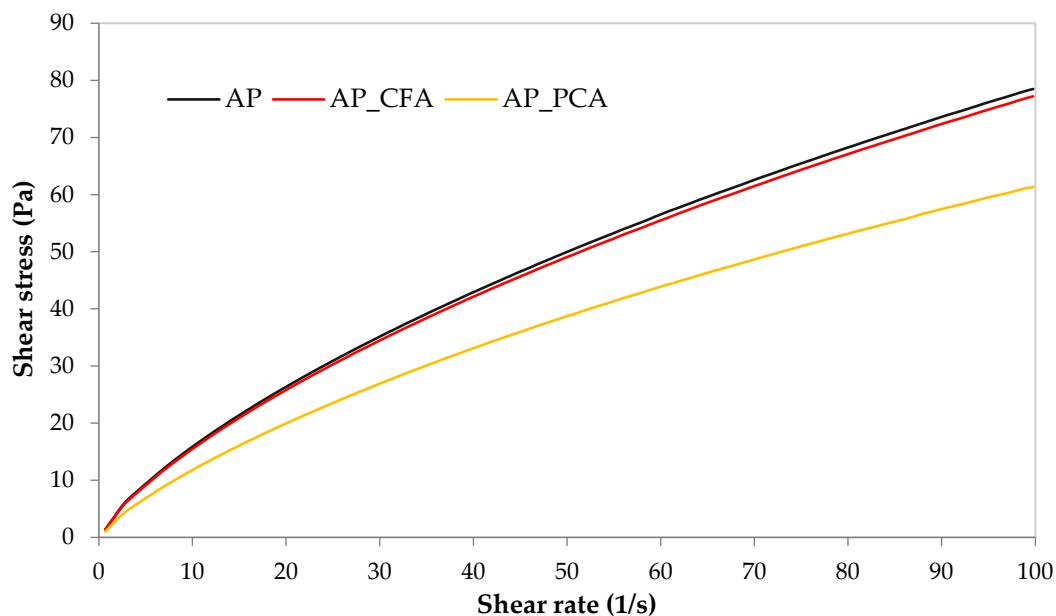
### 3.9. Statistical Analysis

Statistical analysis of the obtained results was performed using Statistica 13.0 (StatSoft Polska Sp. z o.o., Kraków, Poland), utilising one-way ANOVA, Pearson's correlation and Tukey's HSD post hoc test at a significance level of 0.05. Excel 2013 software was also used to process the results. Error bars represent standard deviation (SD).

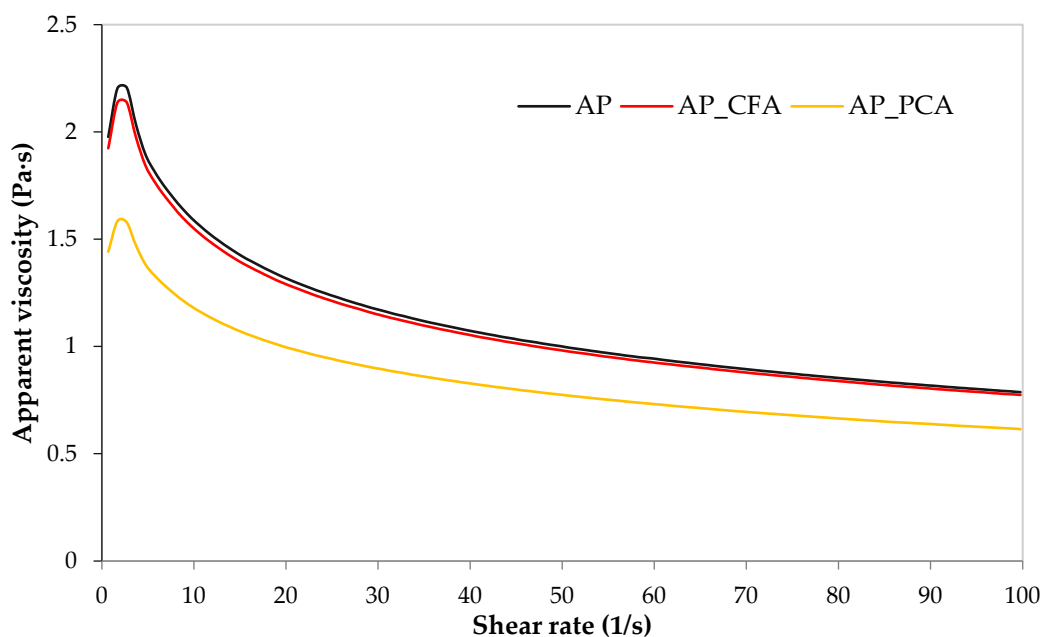
## 4. Results and Discussion

### 4.1. The Effect of Phenolic Acids on the Rheological Properties of Coating Solutions

Rheological properties concern the study of material flow and deformation under stress, which enables better optimisation of processing conditions and desired product attributes [48]. The flow properties of coating solutions enable the analysis of factors related to spreadability, thickness, homogeneity, mechanical properties, microstructure, and application methods [38,49]. The flow (Figure 1) and viscosity curves (Figure 2) of the prepared coating solution, based on apple pectin and its mixtures with phenolic acids (caffeic and protocatechuic), revealed differences in the rheological behaviour of the tested samples. All samples demonstrated non-Newtonian, shear-thinning flow behaviour, which is consistent with previous observations of pectin solutions [50,51].



**Figure 1.** Flow curves of coating-forming solutions based on apple pectin (AP) with the addition of caffeic acid (AP\_CFA) and protocatechuic acid (AP\_PCA).



**Figure 2.** Viscosity curves of coating solution based on apple pectin (AP) with the addition of caffeic acid (AP\_CFA) and protocatechuic (AP\_PCA) acid.

The addition of protocatechuic acid led to a decrease in apparent viscosity across all levels of applied shear stress. This effect may be related to changes in the molecular structure of pectin, particularly how protocatechuic acid influences the dissociation degree of carboxyl groups and potentially disrupts the network formation between polysaccharide chains. These findings align with those reported by Karaki et al. [52,53], who demonstrated that the addition of phenolic acids, such as ferulic acid, reduces molecular packing density and intermolecular interactions in pectin systems, resulting in decreased viscosity. In the case of caffeic acid, the viscosity remained unchanged compared to the control sample (coating solution without phenolic acids). This may be attributed to weaker interactions of this acid with pectin chains and a lower capacity to form hydrogen bonds. The differences

in rheological behaviour can also be interpreted in terms of changes in the hydrophobicity of pectin molecules after adding phenolic compounds. According to Zhang et al. [54], modifying carboxyl side groups and rhamnose content is crucial for the flow properties of apple pectin. The presence of protocatechuic acid may disrupt the structure and length of pectin aggregates, thereby altering viscosity.

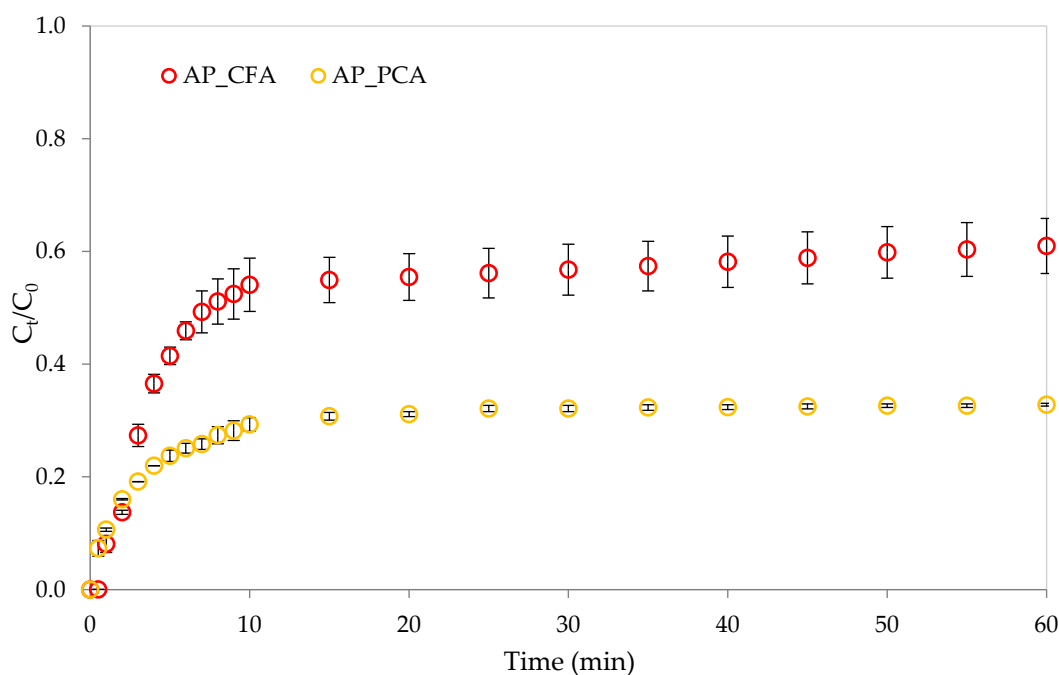
#### 4.2. Release Kinetics of Caffeic and Protocatechuic Acids from Pectin Films

Active packaging is a method used for food preservation that helps maintain the quality, safety, and integrity of food throughout its shelf life [55]. According to Regulation (EC) No. 450 [56], active packaging refers to packaging systems that interact with food to deliberately incorporate components that either release or absorb substances into the packaged food or its surrounding environment. The inclusion of bioactive substances can improve the ability of the packaging material to preserve the physicochemical properties of food, thereby extending its shelf life [57,58]. Some types of packaging films utilise materials that are sensitive to oxygen or contain antioxidants. To achieve a controlled or targeted release of bioactive compounds with specific functional properties, it is essential to consider various factors, including the type of bioactive compound, its delivery method, and the environmental conditions present [59]. In packaging films, parameters such as release rate, diffusion coefficient, and cumulative release are utilised [60]. There are two primary mass transport mechanisms that regulate the release of molecules: internal (diffusion within the material) and external (transport from the material to the surrounding atmosphere or food). Besides facilitating the mass transport of active substances, diffusion can also lead to swelling and/or plasticization of the polymer, which may disrupt the release process [61].

The release kinetics of caffeic and protocatechuic acids from pectin films in a 96% ethanol medium are illustrated in Figure 3. The analysed films displayed excellent solubility in water, as discussed in our previous work [36]. Consequently, distilled water or a 50% ethanol solution did not show suitable release kinetics due to immediate dissolution. The different behaviours of the phenolic acids in ethanol are likely attributed to the varying chemical compositions and properties of the acids. Additionally, the release process may be affected by interactions among the components, specifically pectin, acid, glycerol, and ethanol. Pearson's linear correlation coefficients for both acids were notably high, with protocatechuic acid exhibiting a higher value of 0.9733. Furthermore, all tested variants showed linear correlation coefficients close to unity, indicating strong linear relationships between the variables.

Hernández-García, Vargas, and Chiralt [62] performed methanolic extraction of ferulic, p-coumaric, and protocatechuic phenolic acids from PLA-PHBV blend films. Subsequently, spectrophotometric determinations yielded similar values ( $83.0 \pm 0.03$ ,  $80.0 \pm 0.1$ , and  $79.0 \pm 0.04$  g of retained compound/100 g of incorporated compound, respectively). After conducting kinetic studies on the release of active substances into food simulants, it was found that phenolic acids behaved differently in both simulants (10% *v/v* aqueous ethanol solution and 50% *v/v* aqueous ethanol solution). This difference depended on the chemical affinity of the compounds for the polymer matrix and their solubility. Furthermore, disintegration occurred more rapidly for films containing acids compared to acid-free polyester film, which remained intact for 20 min in contact with both types of simulants. This confirms that the release of phenolic acid and the subsequent pH drop in the aqueous environment significantly influence film disintegration. The highest initial release rate and equilibrium release coefficient were achieved for ferulic acid in both simulants, while protocatechuic acid exhibited the slowest release from films in both simulants. It was also observed that the maximum amount of released acids was significantly lower compared to the solubility of the compounds in water, which increases with the ethanol content in

the simulants. Differences in kinetic parameters are attributed to variations in chemical interactions between the compounds and the polyester matrix, as well as differing degrees of relaxation. The release rate and equilibrium release amount of all phenolic acids were higher in the simulant with a greater proportion of ethanol. Benbettaïeb et al. [63] studied the release kinetics of ferulic acid and tyrosol from chitosan-gelatin-based films, both non-irradiated and irradiated at a dose of 60 kGy, in an aqueous environment with a pH of 7 or lower. The non-irradiated films contained a significantly higher amount of ferulic acid, likely due to ferulic acid's greater ability to interact with the polymer network or promote cross-linking. This interaction leads to improved mechanical properties and permeability of the films. However, there is limited research focusing on the antioxidant activities of phenolic acids and their relationship to the mechanisms of their release.



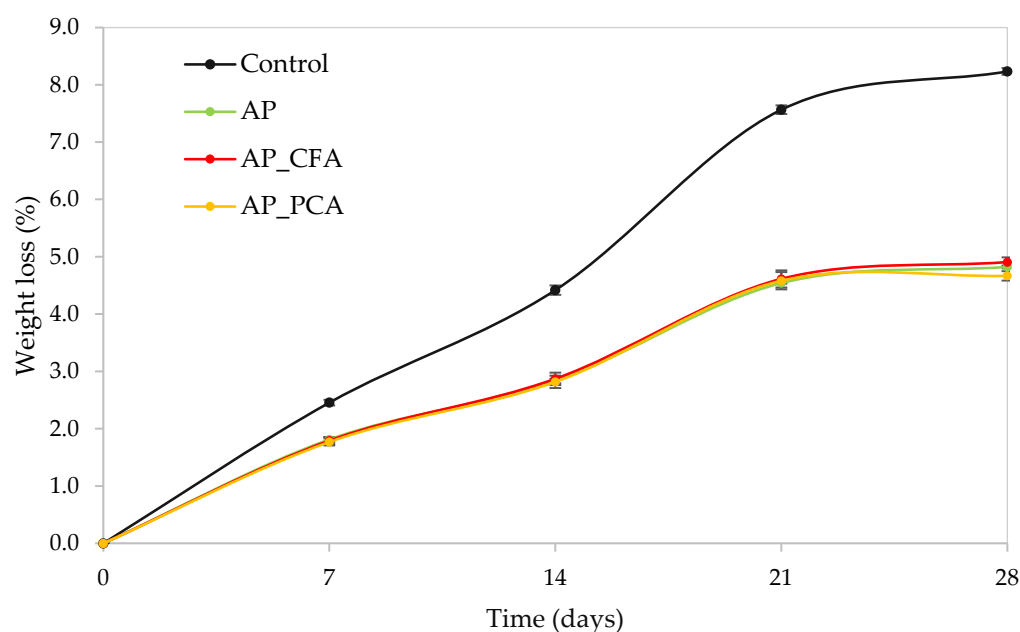
**Figure 3.** Release kinetics of caffeic acid (CFA) and protocatechuic acid (PCA) from apple pectin (AP) packaging films into aqueous medium (96% ethanol). ( $C_t$  is the antioxidant concentration in the medium at time  $t$ ;  $C_0$  is the initial antioxidant concentration in films prior to release).

Controlled-release packaging provides several advantages over traditional methods for extending the shelf life of food. This technology enables the adjustment of antioxidant activity according to the food's storage conditions within the package, ensuring maximum freshness [59]. Typically, the active ingredient can be introduced through methods such as adsorption, covalent immobilisation on the polymer surface, or direct incorporation into the polymer structure. When controlled-release technology involves entrapment within a polymer matrix, the film thickness is critical, as it significantly influences the diffusion characteristics of the active ingredient [60]. Generally, this means that the release rate will decrease as the film thickness increases. Moreover, when nanofillers are added, there is a reduction in the release rate due to an increased diffusion path length, which may result from a decrease in polymer flexibility and film elasticity [64].

#### 4.3. The Effect of Bioactive Coatings Based on Apple Pectin and Phenolic Acids on the Weight Loss of Apples During Storage

The loss of mass in fruit during storage significantly affects its external appearance, which is a key factor in determining quality [65]. Thus, a primary objective of applying edible coatings to fresh fruit is to minimise this loss of mass during storage [1]. Figure 4 shows

the weight losses of uncoated and coated apples with pectin, both with and without caffeic or protocatechuic acids, over a 28-day storage period. It is evident that uncoated apples experienced the highest weight loss, ranging from  $2.46 \pm 0.2\%$  at 7 days to  $8.23 \pm 0.06\%$  at 28 days. This trend indicates that the coating effectively reduced moisture loss caused by evaporation. Considering the effects of bioactive coatings, there was not much difference between the coatings with and without the addition of phenolic acid. The weight loss values ranged from 1.77% to 1.81% at 7 days, and from 4.67% to 4.82% at 28 days. Moreover, the highest weight losses for all samples were observed up to 21 days, after which there were lower differences noted between days 21 and 28. The rapid loss of water results in the drying and degradation of fruit [66]. Studies have shown that the application of edible coatings, whether made from apple pectin alone or mixed with phenolic acids, significantly reduces the mass loss of apples during storage over subsequent weeks. In contrast, uncoated apples do not show this behaviour. Therefore, it can be concluded that developed edible coatings serve as a barrier, preventing moisture loss from the fruit's surface and helping to maintain its quality during storage. Kassebi et al. [17] noted a higher value (approximately 17.5%) of weight loss of uncoated Golden Delicious apples stored at room temperature ( $24 \pm 1\text{ }^{\circ}\text{C}$ ) and 60% air humidity for 4 weeks. The average weight reduction in apples was 5.29% in the first week of measurement and 25.32% at the end of storage. The authors suggested that increased weight loss indicates the internal transpiration of the fruit, mainly due to moisture loss. In general, changes in the fruit during ripening are related to an increase in ethylene synthesis, the respiration process, and the process of pectin degradation that plays an essential role in cell wall degradation, affecting shelf life. However, the mechanisms of these processes are related to many factors, including fruit variety, harvesting time, and storage conditions (temperature and relative humidity). In the case of coated apples, the coating acts as a permeable membrane, which affects biological processes such as respiration and the migration of water vapour [67]. Therefore, edible coatings significantly reduce apple weight loss by forming a barrier that controls moisture and gas exchange, and they inhibit microbial growth by reducing water activity and oxygen availability, thereby extending shelf life and maintaining quality [68].



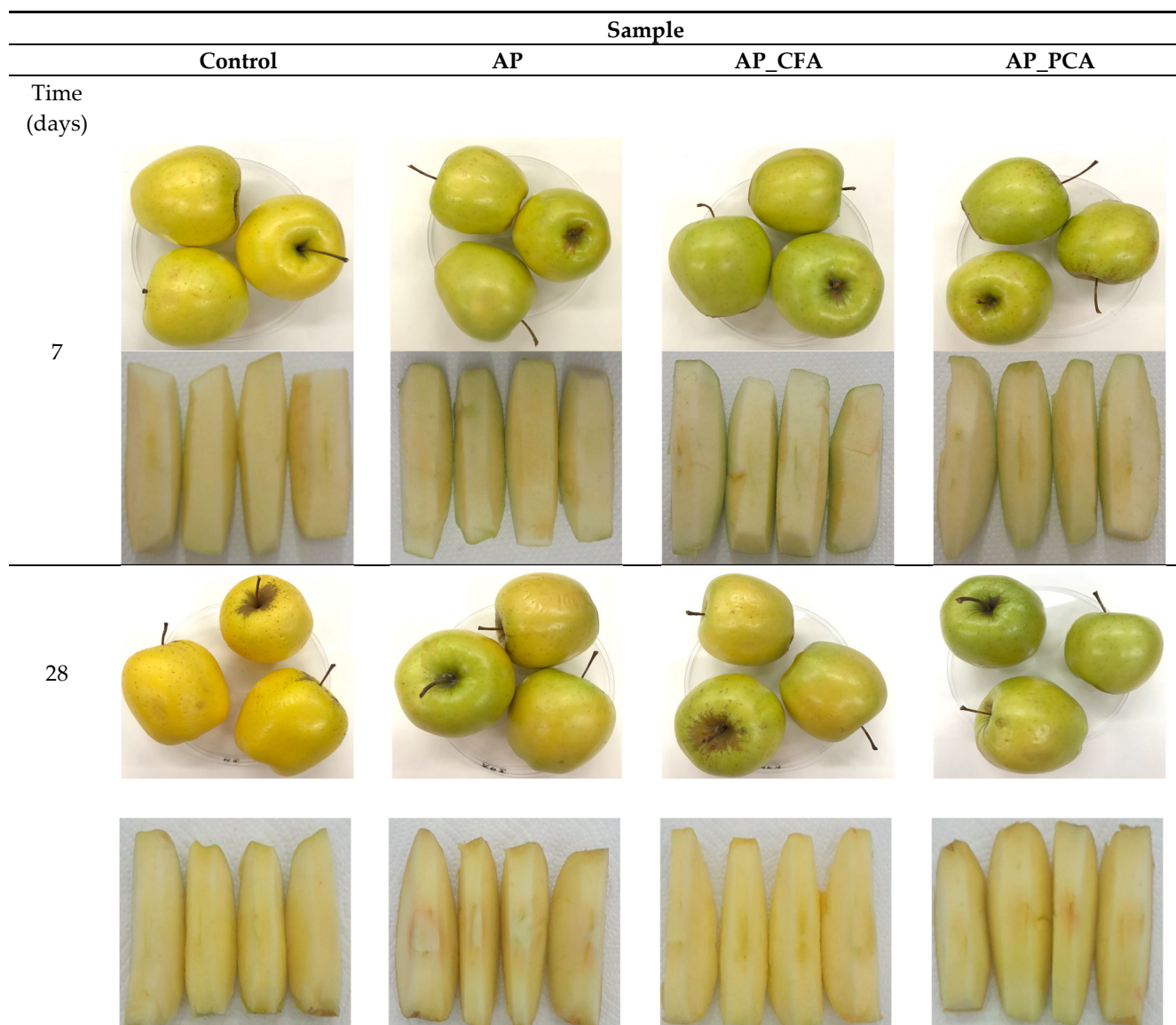
**Figure 4.** Weight loss of uncoated (Control) and coated apple with the coating solution based on apple (AP) incorporated with caffeic (AP\_CFA) and protocatechuic (AP\_PCA).

Storing fruit at room temperature may lead to reduced juice content due to increased moisture loss [69]. Studies by Trebar, Žalik, and Vidrih [70] demonstrate that temperature has a significant effect on weight loss in apples. Notable differences arise when uncoated Golden Delicious apples are stored at ambient temperature (approximately 25 °C) compared to the recommended storage temperature of 5 °C. Due to the high water content in the fruit, which is about 80–90% by weight, rapid evaporation can lead to spoilage and a reduced shelf life. Additionally, the natural barrier on the fruit, along with the use of an appropriate type and amount of edible coating, can influence the extent of weight reduction [71]. Even a small loss of 3 to 10% can negatively impact the appearance of apples, resulting in fruit shrinkage and a decline in their visual appeal [72]. Fruits become unsuitable for sale if their weight loss results in visual changes that are unacceptable to consumers [73]. A 59% reduction in weight loss was observed in apples of the Golab Kohanz variety when they were coated with a nanochitosan solution containing a 0.5% chitosan concentration during the entire storage period. These findings demonstrate the potential of using nanochitosan coatings for climacteric fruits, thereby extending their shelf life [74]. Apples coated with potato starch and polyvinyl alcohol demonstrated better weight retention during storage compared to uncoated apples [75]. Furthermore, coating Golden Delicious apples with water walnut starch-based coatings infused with rosemary essential oil (REO) at concentrations of 0.10%, 0.25%, and 0.50% (*w/v*) positively influenced their post-harvest stability during 120 days of cold storage. The control apples, which were not coated, experienced the highest weight loss, which gradually increased throughout the storage period, in contrast to the coated apples [10]. Maintaining moisture in apples enhances their appearance, preserves firmness, and increases marketable weight, positively impacting growers' income [65]. Jahanshahi et al. [76] demonstrated the potential of using tragacanth gum as a coating to improve postharvest shelf life and maintain the quality of apple fruits, thereby reducing postharvest losses. The results showed that, in both Red Delicious and Golden Delicious cultivars, the shelf life was significantly increased by applying a coating composed of 10 kg/m<sup>3</sup> of gum. This was evidenced by a reduced decrease in firmness and considerably reduced fruit weight loss during 120 days of storage at low temperature (0 ± 1 °C) and 85–95% relative humidity. The study suggests minimising postharvest losses by using an appropriate edible coating.

#### *4.4. The Effect of Bioactive Coatings Based on Apple Pectin and Phenolic Acids on the Colour of Apples During Storage*

In the view of consumers, one of the most important quality features of apples is their colour, which is influenced by cultural preferences and consumer choices. When cultivating apples, factors such as the consistency of colour, the intensity of blush (if applicable), as well as the presence of dents or bruises and the stage of maturity are often considered. Additionally, after harvesting, colour remains a significant quality attribute, affected by storage conditions, which can greatly influence the final quality of the fruit [77,78]. Enzymatic browning in fresh fruit causes skin and flesh discolouration, which reduces the product's commercial value. This browning is highly noticeable to consumers, influencing their perception of freshness and significantly affecting their purchasing decisions [14]. Recent studies indicate that coatings effectively regulate the critical physiological and biochemical changes that lead to enzymatic browning [66]. Figure 5 illustrates both the external and internal appearances of the analysed uncoated and coated apples during storage. It can be observed that the coated apples appear greener compared to the control samples, which are more yellow. There is no visible difference between the samples coated with formulations containing or lacking phenolic acids, indicating that the apple pectin coating serves as a protective layer during storage, regardless of the presence or absence of active compounds.

However, the internal tissue of the uncoated apples appears lighter, while the coated apples show a rapid onset of enzymatic browning compared to the controls.



**Figure 5.** The appearance of uncoated (Control) and coated apples with the coating solution based on apple (AP) incorporated with caffeic (AP\_CFA) and protocatechuic (AP\_PCA) at 7 and 28 days of storage.

The yellow colour of Golden Delicious apples comes from pigments in their skin, which mainly consist of flavonoids, particularly flavonols and flavones, as well as carotenoids. Meanwhile, chlorophyll is responsible for the green colour of the skin [79]. The colour change from green to light green and then to yellow is likely the result of pigment degradation during storage. This process is influenced by the synthesis of carotenoids and the decomposition of chlorophyll. Moreover, these changes are affected by the activity of polyphenol oxidase, the most common enzyme involved in the browning process of apple flesh [17]. Chlorophyll synthesis is a continuous process that occurs before fruit ripening, but it gradually decreases over time. This decline initially leads to a reduction in the green colour of the fruit. Eventually, chlorophyll is replaced by other pigments, resulting in the appearance of visible yellow shades [80].

The results for the colour parameters  $L^*$ ,  $a^*$ , and  $b^*$  for the evaluated apples are presented in Table 2. In the CIELAB colour space,  $L^*$  represents lightness; the  $a^*$  parameter indicates colour on a red-green scale, where positive values reflect red and negative values denote green. The  $b^*$  parameter indicates colour on a yellow-blue scale, with positive values corresponding to yellow and negative values to blue. A high  $L^*$  value indicates that the fruit has a light skin colour. After 28 days of storage, the colour of uncoated apples remained stable, although some changes in the colour components were observed throughout their shelf life.

**Table 2.** The  $L^*$ ,  $a^*$ , and  $b^*$  colour parameters of uncoated (Control) and coated apples with the coating solution based on apple (AP) incorporated with caffeic (AP\_CFA) and protocatechuic (AP\_PCA).

Time (Days)	Sample			
	Control	AP	AP_CFA	AP_PCA
$L^*$				
0	75.77 ± 1.07 <sup>a,A</sup>			
7	75.40 ± 2.45 <sup>a,A</sup>	72.03 ± 5.56 <sup>a,A</sup>	72.05 ± 1.26 <sup>b,A</sup>	72.21 ± 1.76 <sup>a,A</sup>
14	75.01 ± 1.98 <sup>a,B</sup>	70.45 ± 1.50 <sup>a,A</sup>	69.49 ± 1.40 <sup>a,A</sup>	68.64 ± 2.12 <sup>a,A</sup>
21	74.70 ± 1.59 <sup>c,B</sup>	70.30 ± 1.50 <sup>a,A</sup>	68.79 ± 2.44 <sup>a,A</sup>	68.01 ± 1.87 <sup>a,A</sup>
28	74.67 ± 1.79 <sup>a,B</sup>	64.64 ± 7.31 <sup>a,A</sup>	67.37 ± 2.46 <sup>a,A</sup>	64.66 ± 7.17 <sup>a,A</sup>
$a^*$				
0	−11.29 ± 0.68 <sup>a,A</sup>			
7	−7.81 ± 2.32 <sup>b,A</sup>	−9.18 ± 0.94 <sup>a,A</sup>	−9.26 ± 1.20 <sup>a,A</sup>	−8.11 ± 1.49 <sup>a,A</sup>
14	−4.64 ± 2.00 <sup>b,B</sup>	−8.57 ± 1.38 <sup>a,A</sup>	−8.51 ± 1.35 <sup>a,A</sup>	−8.04 ± 1.54 <sup>a,A</sup>
21	−2.75 ± 2.16 <sup>b,B</sup>	−6.12 ± 1.60 <sup>b,A</sup>	−5.96 ± 2.09 <sup>b,A</sup>	−5.58 ± 1.91 <sup>b,A</sup>
28	−2.16 ± 1.28 <sup>b,A</sup>	−4.35 ± 1.38 <sup>b,A</sup>	−3.72 ± 1.64 <sup>b,A</sup>	−3.96 ± 1.65 <sup>b,A</sup>
$b^*$				
0	46.83 ± 0.63 <sup>a,A</sup>			
7	55.21 ± 2.33 <sup>b,B</sup>	46.29 ± 2.74 <sup>a,A</sup>	46.28 ± 1.73 <sup>a,A</sup>	46.13 ± 1.93 <sup>a,A</sup>
14	56.18 ± 1.47 <sup>b,B</sup>	44.51 ± 1.68 <sup>a,A</sup>	43.68 ± 1.41 <sup>a,A</sup>	44.15 ± 2.14 <sup>a,A</sup>
21	57.22 ± 2.43 <sup>b,B</sup>	44.39 ± 2.26 <sup>a,A</sup>	43.39 ± 2.04 <sup>a,A</sup>	43.93 ± 2.03 <sup>a,A</sup>
28	57.76 ± 3.08 <sup>b,B</sup>	42.97 ± 4.25 <sup>a,A</sup>	42.69 ± 2.64 <sup>a,A</sup>	42.59 ± 1.43 <sup>a,A</sup>

Mean values ± standard deviations. Different superscript letters (<sup>a-c</sup>) within the same column or rows (<sup>A,B</sup>) indicate significant differences between the samples ( $p < 0.05$ ).

An analysis of the  $L^*$  parameter revealed that it fluctuated between  $64.64 ± 7.31$  and  $72.21 ± 1.76$  for coated apples, while uncoated apples ranged from  $74.67 ± 1.79$  to  $75.77 ± 1.07$  (Table 2). This suggests that time had a significant impact on changes in lightness. The decrease in lightness observed on the 28th day of storage for apples coated with a pectin solution containing added caffeic acid and protocatechuic acid was attributed to the development of brown spots on the apples' surface. A decrease in the  $L^*$  parameter was observed, which corresponds to a reduction in lightness as the skin surface yellowed and darkened due to spoilage. The results showed the significant differences between the control samples and the coated apples after 14 days of storage. There were no significant differences among the types of coatings used, suggesting that all pectin coatings, with and without the phenolic acids, were effective in the reduction in lightness ( $p < 0.05$ ). This change may be attributable to various alterations in the fruit's tissues, including enzymatic browning, dehydration, and ripening. Additionally, the colour change in apples is linked to weight loss, which affects ethylene production and respiration. Notably, the smallest reduction in lightness values was observed in the control group of uncoated apples. Kassebi et al. [17] investigated the changes in colour of Golden Delicious apples over a six-week storage period at 24 °C and 60% relative humidity. They found that the  $L^*$

parameter also decreased slightly, from 74.81 to 71.27, as storage time increased from 1 to 6 weeks.

The  $a^*$  and  $b^*$  colour parameters are closely related and indicate the ripening process of fruit. As fruit ripens, the amount of green colour (indicated by negative  $a^*$ ) decreases, while the amount of yellow colour (indicated by positive  $b^*$ ) increases. This colour change occurs due to the decomposition of chlorophyll, which leads to the synthesis of other pigments, primarily anthocyanins or carotenoids. The most noticeable change is in the colour of the skin, which typically transitions from green to a lighter shade. The brown-green colour observed in apples can be attributed to the degradation of chlorophyll, particularly the loss of the  $Mg^{2+}$  ion from chlorophyll, resulting in the formation of pheophytin. Several factors can influence chlorophyll degradation, including exposure to heat, light, and oxidising agents. Research indicates that the pheophytin reaction occurs more rapidly in the presence of heat due to protein denaturation [81]. In conclusion, using edible coatings that contain phenolic acids may help reduce chlorophyll degradation and preserve the green colour of Golden Delicious apples.

Negative  $a^*$  parameter values were observed for all samples (Table 2), indicating a green skin colour, which is characteristic of the Golden Delicious cultivar. The highest increase in parameter  $a^*$ , from  $-11.29 \pm 0.68$  to  $2.16 \pm 1.28$ , was observed for uncoated apples, indicating their advanced ripeness compared to the other samples. Pectin coatings mitigated this increase, resulting in values of  $-4.35 \pm 1.38$  for coated samples without phenolic acids,  $-4.72 \pm 1.64$  for those with caffeic acid, and  $-3.96 \pm 1.65$  for those with protocatechuic acid. It is observed that all bioactive coatings effectively affected the lower values of parameter  $a^*$  after 14 and 21 days of storage. However, among the types of coatings used, there were no significant differences in the values ( $p < 0.05$ ). This is probably attributed to the lack of effect of phenolic acids on this parameter for pectin films.

The  $b^*$  parameter values for control apples ranged from  $46.83 \pm 0.63$  to  $57.76 \pm 3.08$ , demonstrating an increasing trend with each week of storage (Table 2). In contrast, the  $b^*$  colour parameter for coated apples decreased as storage time increased. All  $b^*$  values remained positive, indicating that the apples consistently fell within the yellow colour range throughout each storage period. Storing the apples for an additional two months did not significantly change the fruit's colour; however, slight differences were noted across all colour quality classes. As weight loss increased, the  $b^*$  value also increased, suggesting that the apples became more yellow. This water loss may lead to a concentration of pigments in the apples, enhancing the yellow colour. Time had an impact on colour change in all combinations, but the application of coatings helped limit this change. The experiment confirmed that using coatings delays the yellowing of apple skin. This effect was observed for each coating formulation and at all times tested, indicating that all coated apples were significantly affected by bioactive coatings in a similar manner ( $p < 0.05$ ). There were no differences between the types of coatings used.

Coatings with added antioxidants, such as ascorbic acid, calcium chloride, or calcium lactate, are particularly effective at maintaining colour, mostly in fresh-cut apples. Incorporating antimicrobial agents, such as chitosan or essential oils, significantly reduces the overall microbial load, both for minimally processed and non-processed fruits [82].

#### 4.5. The Effect of Bioactive Coatings Based on Apple Pectin and Phenolic Acids on the Total Soluble Solids of Apples During Storage

Both coated and uncoated apples showed an increase in total soluble solids when comparing storage periods of 7 and 28 days (Table 3). For control samples, the values increased from  $13.47 \pm 0.37$  to  $14.95 \pm 0.48$  °Brix at 28 days of storage. In contrast, for the coated sample, the increase in this parameter was within the range of 13.52–13.57 °Brix. Thus, the coating process significantly decreased the rate of increase in the total soluble

solids in fruit throughout the period, as only a slight increase in extract was observed in the coated samples after 28 days of storage. The highest extract content was recorded in the control sample on day 28 of storage, measuring  $14.95 \pm 0.48$  °Brix. These results indicate that, from a physicochemical perspective, significant changes occurred during the storage of Golden Delicious apples, depending on their state of ripeness. Kassebi et al. [17] also observed an increase in extract during the storage of the same variety of apples. The initial extract value was  $13.61$  °Brix in the first week of storage at room temperature, which increased to  $14.78$  °Brix after six weeks. This increase may be attributed to a decrease in fruit moisture, leading to a higher concentration of sugars within the fruit, the breakdown of starch into sugars, or the hydrolysis of cell wall polysaccharides. Regarding the type of coating, there were similar tendencies among the samples. A significant increase in total soluble solids was observed in control samples after 21 and 28 days of storage ( $p < 0.05$ ).

**Table 3.** Total soluble solids, titratable acidity and pH of uncoated (Control) and coated apples with the coating solution based on apple (AP) incorporated with caffeic (AP\_CFA) and protocatechuic (AP\_PCA).

Time (Days)	Sample			
	Control	AP	AP_CFA	AP_PCA
<b>TSS (°Brix)</b>				
0	$13.47 \pm 0.37$ a,A			
7	$13.77 \pm 0.11$ a,A	$13.50 \pm 0.10$ a,A	$13.48 \pm 0.23$ a,A	$13.53 \pm 0.40$ a,A
14	$14.03 \pm 0.39$ a,A	$13.50 \pm 0.24$ a,A	$13.52 \pm 0.21$ a,A	$13.50 \pm 0.37$ a,A
21	$14.35 \pm 0.19$ b,B	$13.53 \pm 0.07$ a,A	$13.52 \pm 0.18$ a,A	$13.55 \pm 0.36$ a,A
28	$14.95 \pm 0.48$ b,B	$13.53 \pm 0.19$ a,A	$13.52 \pm 0.32$ a,A	$13.57 \pm 0.27$ a,A
<b>Titratable acidity (% of malic acid)</b>				
0	$1.37 \pm 0.02$ b,A			
7	$0.82 \pm 0.05$ a,A	$0.93 \pm 0.05$ a,A	$0.91 \pm 0.02$ a,A	$0.89 \pm 0.11$ a,A
14	$0.90 \pm 0.07$ a,B	$0.79 \pm 0.08$ a,AB	$0.77 \pm 0.03$ a,A	$0.77 \pm 0.09$ a,A
21	$0.88 \pm 0.04$ a,B	$0.75 \pm 0.05$ a,A	$0.76 \pm 0.08$ a,A	$0.77 \pm 0.05$ a,A
28	$0.80 \pm 0.05$ a,A	$0.79 \pm 0.05$ a,A	$0.76 \pm 0.05$ a,A	$0.73 \pm 0.02$ a,A
<b>pH</b>				
0	$3.59 \pm 0.04$ a,A			
7	$3.84 \pm 0.02$ b,A	$3.84 \pm 0.01$ b,A	$3.83 \pm 0.03$ a,A	$3.79 \pm 0.05$ a,A
14	$3.90 \pm 0.03$ b,A	$3.98 \pm 0.09$ b,A	$3.98 \pm 0.06$ b,A	$4.04 \pm 0.01$ b,A
21	$4.00 \pm 0.02$ b,B	$4.09 \pm 0.06$ b,A	$4.12 \pm 0.08$ b,A	$4.16 \pm 0.01$ b,A
28	$4.05 \pm 0.06$ b,A	$4.11 \pm 0.07$ b,A	$4.12 \pm 0.03$ b,A	$4.26 \pm 0.04$ b,A

Mean values  $\pm$  standard deviations. Different superscript letters (a,b) within the same column or rows (A,B) indicate significant differences between the samples ( $p < 0.05$ ).

#### 4.6. The Effect of Bioactive Coatings Based on Apple Pectin and Phenolic Acids on the Acidity of Apples During Storage

Acidity in fruits is determined by the amount of organic acids they contain, which significantly impacts both their physiological metabolism and sensory qualities. Additionally, acidity tends to decrease over time during storage and ripening. This decline is due to enzymatic processes that occur, leading to an increase in the fruit's sweetness [10]. Understanding how the storage process affects acidity is crucial for improving the quality of fruit after harvest. Fruit acidity is primarily determined by the presence of one or two acids, such as citric, tartaric, and malic, which contribute to the sour taste. Conversely, the sweet taste of apples depends on the amount of soluble solids. For apples, approximately 90% of all organic acids consist of malic acid, at a concentration ranging from 1.72 to 29.27 mg/g. During apple growth, malate content increases; however, it decreases during ripening and storage. Citric acid, present in small amounts, also contributes to the acceptability of apples.

Conventional storage of apples at  $0 \pm 0.1$  °C has been found to be ineffective in maintaining fruit acidity, an essential quality parameter [83].

It was observed that the acidity of uncoated apples decreased the most significantly (Table 3), from  $1.37 \pm 0.02$  to  $0.80 \pm 0.05\%$ , similar to apples coated with the formulation without phenolic acids ( $0.79 \pm 0.05\%$ ). The lowest values of acidity were observed for the sample coated with solutions containing caffeic and protocatechuic acids,  $0.76 \pm 0.05$  and  $0.73 \pm 0.02\%$ , respectively. After analysing the obtained results, it can be concluded that coated apples exhibited the most significant change in malic acid content during storage, leading to a considerable alteration in their taste. This phenomenon may arise from the fact that, during storage, the starch content decreases while the sugar content increases, resulting in a decrease in the acidity of the apples [84]. However, considering the coating type, similar tendencies were observed among the samples. A significant increase in titratable acidity was observed in uncoated apples after 14 and 21 days of storage ( $p < 0.05$ ). In a study conducted by Rashid et al. [85], apples were coated with solutions containing fenugreek and linseed, then stored at  $25 \pm 2$  °C for 35 days. The experimental results indicated that the titratable acidity values decreased for both coated and uncoated fruits. This reduction in acidity can likely be attributed to the coatings, which slowed down the respiration process and, consequently, the metabolic processes, including acid decomposition. It may be related to a reduction in gas exchange with the external environment [86].

#### *4.7. The Effect of Bioactive Coatings Based on Apple Pectin and Phenolic Acids on the pH of Apples During Storage*

The occurrence of an increase in the pH of fruits during storage is related to their respiration, during which the oxidation of organic acids occurs and the decrease in acids present in the fruits with the passage of time [87]. The pH of the fruit systematically increased in all samples over 28 days of storage (Table 3). No significant fluctuations were observed between combinations regarding storage time, and all samples exhibited similar pH values, ranging from 3.59 to 4.26. The values slightly increased from  $3.59 \pm 0.04$  to  $4.05 \pm 0.06$  for control samples. Coated apples showed higher values:  $4.11 \pm 0.07$  for samples coated with the solution without phenolic acids, and  $4.12 \pm 0.03$  and  $4.26 \pm 0.04$  for samples coated with solutions containing caffeic and protocatechuic acids, respectively. However, after 21 and 28 days of storage, a significant increase in pH was observed for uncoated apples ( $4.00 \pm 0.02$ ) and apples coated with a solution containing protocatechuic acid ( $4.26 \pm 0.04$ ), respectively ( $p < 0.05$ ). An increase in pH in apples during storage is a natural part of the ripening process, where apples become less acidic and more alkaline as organic acids, like malic acid, decrease due to respiration [88]. Coated apples were less ripe; however, their pH values were higher than those of the controls. This may suggest that the increase in pH is related to changes in the fruits' chemical composition. In contrast, Vieira et al. [89] reported a much higher pH of  $4.97 \pm 0.04$  for fresh Golden Delicious apples, while Alegre et al. [90] found an apple pH of 4.16. Variations in pH can be attributed to plant growth conditions. During storage, organic acids in fruits act as respiratory substrates and are slowly utilised, which increases pH and decreases acidity [4]. Moreover, pH may be related to increased dry matter content and polysaccharide depolymerisation with extended storage [85].

#### *4.8. The Effect of Bioactive Coatings Based on Apple Pectin and Phenolic Acids on the Firmness of Apples During Storage*

The primary challenge in apple storage is maintaining firmness, which significantly influences consumer sensory perception and commercial value. The loss of firmness, or softening, is an undesirable process during apple ripening. Higher fruit hardness and

firmness correlate with increased juiciness, crunchiness, and reduced mealiness, leading to greater consumer acceptance [91]. The firmness results, presented in Table 4, ranged from  $58.03 \pm 6.50$  to  $36.84 \pm 1.30$  N for fresh apples and after 28 days of storage. For the coated apples, a lower rate of decrease in values was observed, ranging from 43.64 to 44.61 N, with minimal differences among the types of coatings. Taking into account the coating type, no significant effect was observed between formulations, indicating that a significant reduction in firmness was achieved for uncoated apples after 14, 21, and 28 days of storage ( $p < 0.05$ ) compared to coated samples. In general, apples characterised by greater hardness are more resistant to mechanical damage and are more suitable for longer storage [92]. Softer fruit, due to enzymatically induced modifications of polysaccharide cell walls and their networks, is more susceptible to fungal infections [93]. In contrast, the loss of firmness and softening of apples are primarily attributed to modifications in pectin within the cell walls, with minor changes in hemicellulose and cellulose, as well as cellulase activity [94]. Edible coatings limit the action of enzymes responsible for pectin decomposition, thereby slowing metabolic processes and maintaining apple firmness [85]. Polysaccharide-based coatings are used as structural reinforcement to ensure cell wall integrity and mitigate mechanical damage that may occur during transport, thereby ensuring appropriate hardness and texture [5]. The hardness of Golden Delicious apples may also be influenced by soil and climatic conditions [70].

**Table 4.** Firmness of uncoated (Control) and coated apples with the coating solution based on apple (AP) incorporated with caffeic (AP\_CFA) and protocatechuic (AP\_PCA).

Time (Days)	Sample			
	Control	AP	AP_CFA	AP_PCA
	Firmness (N)			
0	$58.03 \pm 6.50$ <sup>c,A</sup>			
7	$50.77 \pm 2.40$ <sup>b,B</sup>	$49.01 \pm 1.60$ <sup>b,AB</sup>	$48.32 \pm 2.60$ <sup>a,A</sup>	$48.03 \pm 1.60$ <sup>a,A</sup>
14	$42.80 \pm 3.70$ <sup>a,A</sup>	$45.86 \pm 2.70$ <sup>b,AB</sup>	$47.40 \pm 2.60$ <sup>a,B</sup>	$46.45 \pm 2.80$ <sup>a,B</sup>
21	$39.73 \pm 2.50$ <sup>a,A</sup>	$43.94 \pm 3.70$ <sup>a,B</sup>	$44.70 \pm 3.30$ <sup>a,B</sup>	$45.33 \pm 2.90$ <sup>a,B</sup>
28	$36.84 \pm 1.30$ <sup>a,A</sup>	$43.64 \pm 3.50$ <sup>a,B</sup>	$44.20 \pm 4.10$ <sup>a,B</sup>	$44.61 \pm 4.80$ <sup>a,B</sup>

Mean values  $\pm$  standard deviations. Different superscript letters (<sup>a-c</sup>) within the same column or rows (<sup>A,B</sup>) indicate significant differences between the samples ( $p < 0.05$ ).

One method of limiting the loss of apple firmness is to use an appropriate concentration of coating containing calcium and wax combined with hydrocooling, which delays fruit ripening and maintains quality during long-term storage [95]. It was observed that using 2% potato starch and 2% apricot kernel oil maintained higher apple hardness, measuring 62.2 N and 62.8 N, respectively, during storage at room temperature and in a cold store [96]. The loss of firmness and limitation of internal browning of Granny Smith apples were also obtained by covering the apples with a wax coating [97]. On the other hand, the factor contributing to fruit softening is the production and accumulation of ethylene, which may induce undesirable physiological and biochemical changes in fruits, including the expansion of cell walls and the activation of proteolytic and pectolytic enzymes [98]. If the hardness drops below 44 N, it is considered to be the beginning of the occurrence of flouriness [99].

#### 4.9. The Effect of Bioactive Coatings Based on Apple Pectin and Phenolic Acids on the Respiration of Apples During Storage

Fruit ripening processes involve the exchange of gases, such as carbon dioxide and oxygen, with the external atmosphere during storage. To maintain the appropriate quality of apples during storage, reducing ethylene metabolism and fruit respiration is neces-

sary [100]. Apple skin is covered with a complex mixture of lipids that protects the fruit from environmental stress and affects gas exchange. The hydrophobic layer allows controlled diffusion of oxygen and carbon dioxide, facilitating proper fruit respiration and preventing the accumulation of carbon dioxide and ethylene. This layer is essential for maintaining the freshness and quality of apples during storage [101]. Unfortunately, cuticular wax can change during ripening and storage, increasing the content of alcohols, esters, and fatty acids in its composition, affecting the formation of a greasy or oily layer on the fruit's surface. The increase in skin greasiness after harvesting and storage at room temperature decreases their commercial value. Additionally, as apples ripen, increased ethylene production affects the fruit's physiological and metabolic functions, supporting ripening and the synthesis and degradation of cuticular wax. Due to increased wax synthesis with ethylene, the fruit skin becomes oily [102]. Reaching an oxygen level below 8% in the fruit limits endogenous ethylene production and extends its shelf life [66]. Lower oxygen content can also lead to the initiation of anaerobic respiration, causing ethanol accumulation and the development of undesirable flavours, which negatively affect the sensory experience of consumers [103]. Recent reports indicate that various types of edible coatings inhibit the exchange of carbon dioxide, oxygen, and ethylene, effectively delaying ripening and maintaining quality after harvest [66]. As noted from Table 5, differences in respiration rates were observed between uncoated and coated apples. From the beginning of storage, control apples produced significantly more ethylene, approximately twice as much, as coated apples, suggesting that the coatings limited ethylene production and thus slowed the ripening process. It is known that the rapid increase in ethylene synthesis, which can occur in climacteric fruit, causes changes in colour, aroma, texture, and flavour. This increase can also reduce fruit firmness and, therefore, consumer acceptability [104]. A gradual increase in ethylene release was also observed for all coated samples up to 28 days of storage, whereas for uncoated samples, only up to 21 days, suggesting the limit of ripeness. The effectiveness of reducing ethylene production may depend on the concentration and type of phenolic acid contained in the coating. However, taking into account the coating type, all coated apples showed significantly lower respiration rates in comparison to control samples for each week of analysis ( $p < 0.05$ ).

**Table 5.** Ethylene and carbon dioxide production of uncoated (Control) and coated apples with the coating solution based on apple (AP) incorporated with caffeic (AP\_CFA) and protocatechuic (AP\_PCA).

Time (Days)	Sample			
	Control	AP	AP_CFA	AP_PCA
<b>Ethylene (ppm C<sub>2</sub>H<sub>4</sub>/kg/h)</b>				
0	60.40 ± 3.00 <sup>b,A</sup>			
7	63.80 ± 7.10 <sup>b,B</sup>	18.25 ± 0.65 <sup>b,A</sup>	8.85 ± 1.15 <sup>a,A</sup>	6.80 ± 0.10 <sup>a,A</sup>
14	67.00 ± 1.70 <sup>b,B</sup>	15.50 ± 0.55 <sup>a,A</sup>	13.70 ± 2.30 <sup>b,A</sup>	11.85 ± 1.00 <sup>a,A</sup>
21	67.65 ± 7.45 <sup>b,B</sup>	28.75 ± 1.05 <sup>d,A</sup>	25.25 ± 3.45 <sup>c,A</sup>	23.25 ± 1.85 <sup>a,A</sup>
28	52.75 ± 3.05 <sup>a,B</sup>	25.70 ± 0.70 <sup>c,A</sup>	23.90 ± 0.50 <sup>c,A</sup>	23.55 ± 1.65 <sup>a,A</sup>
<b>Carbon dioxide (mg/kg/h)</b>				
0	0.23 ± 0.13 <sup>c,A</sup>			
7	0.27 ± 0.05 <sup>d,B</sup>	0.21 ± 0.01 <sup>d,A</sup>	0.16 ± 0.01 <sup>c,A</sup>	0.17 ± 0.04 <sup>c,A</sup>
14	0.14 ± 0.00 <sup>a,A</sup>	0.15 ± 0.02 <sup>b,A</sup>	0.15 ± 0.02 <sup>b,A</sup>	0.18 ± 0.02 <sup>d,A</sup>
21	0.16 ± 0.02 <sup>b,A</sup>	0.18 ± 0.04 <sup>c,A</sup>	0.14 ± 0.01 <sup>a,A</sup>	0.14 ± 0.03 <sup>b,A</sup>
28	0.14 ± 0.02 <sup>a,A</sup>	0.13 ± 0.02 <sup>a,A</sup>	0.15 ± 0.01 <sup>b,A</sup>	0.13 ± 0.01 <sup>a,A</sup>

Mean values ± standard deviations. Different superscript letters (<sup>a-d</sup>) within the same column or rows (<sup>A,B</sup>) indicate significant differences between the samples ( $p < 0.05$ ).

Carbon dioxide is a compound released during fruit respiration. In the experiment, increased production of this gas was observed in the first week for the control sample, indicating vigorous respiration (Table 5). The use of coatings slowed down this process. The study also noted the impact of coatings on limiting the fruit's respiration process, as higher oxygen content in the chamber meant less absorption by the apple of this gas, which is used in the respiration process. This fact again confirms the improved barrier properties of coatings using phenolic acids. However, the coating type has no significant effect on carbon dioxide production compared to control samples at each time of analysis ( $p < 0.05$ ), except at 7 days, when the highest value was observed for uncoated apples.

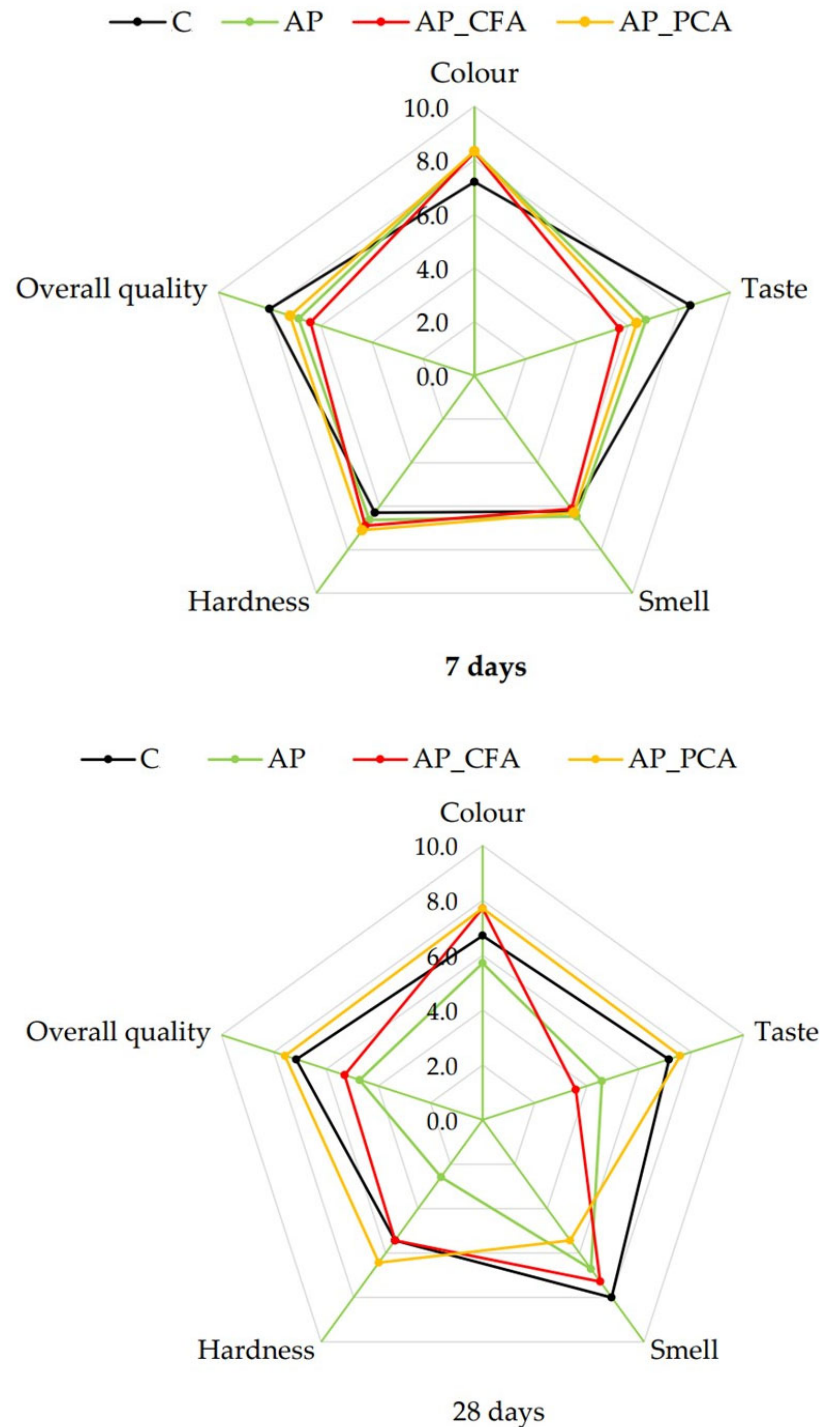
Yellow apples of the cultivars 'Tsugaru', 'Summer King', and 'Shinano Gold' (*Malus × domestica* Borkh.) were coated with a mixture of edible sucrose monoesters of fatty acids and ethanol and stored for up to 28 days at room temperature. A significant reduction in apple respiration rate was observed after both 14 and 28 days of storage, along with the preservation of skin colour and firmness [105]. Softening is also strongly correlated with increased respiration and ethylene production, as evidenced in a study of seven apple cultivars [98]. Coating 'Golden Reinders' and 'Granny Smith' apples with hydroxypropylmethylcellulose coating delayed fruit ripening by inhibiting ethylene production [79]. Ou et al. [106] demonstrated that the addition of ferulic acid to soy protein isolate reduced oxygen permeability by 18.8%. Similarly, Fabra et al. [107] observed a 32% decrease in oxygen permeability through sodium caseinate films after the addition of ferulic acid.

#### 4.10. The Effect of Bioactive Coatings Based on Apple Pectin and Phenolic Acids on the Sensory Attributes of Apples During Storage

The sensory quality of apples is influenced by various factors, including hardness, texture, sweetness, tartness, aroma, and taste. A significant portion of the fruit's aroma is attributed to volatile compounds produced by the apples. For Golden Delicious apples, butyl acetate, hexyl acetate, 2-methylbutyl acetate, and ethyl 2-methylbutanoate are the most crucial compounds determining sensory quality [108]. Apple varieties with crisp flesh are assumed to have a higher concentration of water-soluble pectin, whereas less firm fruits contain more ion-bound pectin. Changes in cell wall hydrolase activity and related gene expression levels significantly affect fruit firmness. During fruit softening, polysaccharide modification depends on various enzymes, such as polygalacturonase, which is responsible for pectin decomposition [109]. In addition to regulating the sensory quality of fruits, the goal is to obtain edible coatings that are tasteless, odourless, and colourless [110]. Improved sensory characteristics, including texture, taste, aroma, and sweetness, were observed in apples treated with composite coatings made from arabinoxylan and  $\beta$ -glucan stearic acid ester, compared to uncoated apples. The fruits were stored at 22 °C for 45 days [111].

Figure 6 presents the sensory analysis results of control apples and apples coated with edible coatings containing phenolic acids after 7 and 28 days of storage. The results indicate that the sensory acceptability of all analysed variants decreased over time, as evidenced by lower scores after 28 days. After 7 days of storage, coated apples exhibited a better appearance than control apples. This was probably due to lower water loss, preservation of fruit firmness, and greater gloss resulting from the application of coating solutions, which enhanced the fruit's attractiveness. No major differences were noted in the smell between the samples, while the control apples received the best scores for taste. After 28 days of storage, apples coated with an edible coating containing protocatechuic acid received the highest scores, excluding the smell variant. However, the differences between this variant and the control were 0.5–1, suggesting no significant differences but only consumer preferences. Sensory data showed that coated apples were rated as juicier than uncoated apples. Coated apples were rated as crispy on the first bite and firm internally, whereas uncoated apples were rated as mushy and floury. Internal texture was defined by the

degree to which the apple disintegrated or remained intact. Control apples and those coated with coatings containing phenolic acids were harder than apples coated solely with apple pectin solution, which were the most crisp and floury. Therefore, it can be concluded that the coatings reduced the respiration rate of the apples, thereby preventing degradation. After 28 days of storage, apples coated with protocatechuic acid received the highest scores. Only their aroma was lower, possibly because the ripest control apples had a more intense aroma. The control apples exhibited a softer, drier texture and a matte appearance.



**Figure 6.** Sensory attributes of uncoated (C) and coated apples with the coating solution based on apple (AP) incorporated with caffeic (AP\_CFA) and protocatechuic (AP\_PCA) after 7 and 28 days of storage.

## 5. Conclusions

Bioactive coatings, based on apple pectin with and without the addition of selected phenolic acids, were developed and applied to Golden Delicious apples for preservation during storage at ambient conditions. Coated apples maintained appropriate fruit quality for 28 days of storage, without visible microbial mould growth. The coating process helped maintain fruit firmness and limited changes in apple mass during storage at  $22 \pm 1$  °C and  $40 \pm 5\%$  relative humidity. Apples treated with edible coatings exhibited a slower respiration process, resulting in delayed fruit ripening. This was confirmed by differences in total soluble solids, acidity, and pH of the apples. Coatings made of apple pectin alone, or apple pectin with added phenolic acids, preserved the apples' appropriate colour and visual appearance. All samples showed good stability over time. The coating reduced the rate of respiration and ethylene production, thereby delaying fruit ripening. Sensory analysis revealed that respondents consistently perceived coated apples in a positive light throughout the entire storage period. The highest taste scores were observed after 28 days of storage for the apple-treated solution containing protocatechuic acid. The differences in the results among the coated samples suggest that apple pectin coatings, even without the addition of active ingredients, were effective in maintaining the quality of apples. More research is needed to provide a more comprehensive understanding of the effects of edible coatings, including bioactive compounds in apples such as vitamin C and polyphenols, as well as radical scavenging activity and microbiological analyses. Further research is also required to investigate the combination of both phenolic acids to determine synergistic effects on antioxidant or antimicrobial activity, including release kinetics.

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mgr inż. Magdalena Mikus  
e-mail: magdalena\_mikus@sggw.edu.pl

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Szkoły Głównej Gospodarstwa Wiejskiego w Warszawie**

**Oświadczenie o współautorstwie**

Niniejszym oświadczam, że w pracy Mikus M., Galus S. (2025). Extending the Shelf Life of Apples After Harvest Using Edible Coatings as Active Packaging—A Review. *Applied Sciences*, 15(2), 767 <https://doi.org/10.3390/app15020767> mój indywidualny udział w jej powstaniu polegał na przeprowadzeniu przeglądu literatury, analizie zebranych danych, przygotowaniu manuskryptu i współudziale w edycji manuskryptu i przygotowaniu odpowiedzi na recenzje.

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Magdalena Mikus

Podpis

Warszawa, 12.02.2026 r.

dr hab. inż. Sabina Galus, prof.  
e-mail: sabina\_galus@sggw.edu.pl

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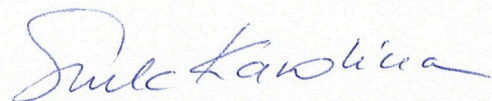
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dr hab. inż. Karolina Szulc  
e-mail: karolina\_szulc1@sggw.edu.pl

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Szkoły Głównej Gospodarstwa Wiejskiego w Warszawie**

**Oświadczenie o współautorstwie**

Niniejszym oświadczam, że w pracy Mikus M., Szulc K., Galus S. (2025). The Use of Bioactive Edible Coatings Based on Pectin and Phenolic Acids for Enhancing Quality Attributes of Golden Delicious Apples During Storage. *Processes*, 13(12), 3821 <https://doi.org/10.3390/pr13123821> mój indywidualny udział w jej powstaniu polegał na współdziałaniu w wykonaniu i opracowaniu wyników właściwości reologicznych roztworów foliotwórczych oraz współdziałaniu w redagowaniu i przygotowaniu odpowiedzi na recenzje dotyczące rozdziału o właściwościach reologicznych.



Podpis