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Supercritical impregnation of pomegranate biomass as active packaging

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Abstract

This study explores the effect of supercritical solvent impregnation (SSI) time (1, 3 and 5 h; 300 bar and 60°C) of pomegranate peel extract on the physical and functional properties of low-density polyethylene (LDPE) films. The results indicate that SSI significantly increased both film thickness and mass. Impregnation time of 1 h maximised impregnation yield (mass gain 5.55 mg and impregnation yield 2.13%), increased film thickness to 102.0 μm and improved the light barrier properties as compared to the control film. Film opacity increased from 2.37 to 2.88 AU mm^{-1} (1 h) and to 3.39 AU mm^{-1} (5 h). Surface roughness also increased with impregnation time. The Fourier transform infrared spectra corroborated the presence of oxygenated phenolics in impregnated films. The highest total phenolic content (0.037 mg GAE g^{-1} film) was observed at 1 h, consistent with the results of DPPH and ABTS radical scavenging activities. SSI provided these improvements through a short, solvent-free and low-temperature process, which eliminates the need for toxic organic solvents, avoids high-energy solvent evaporation steps and achieves functionalisation within 1 h compared with multi-hour or multi-step conventional methods. Overall, the results demonstrate that SSI is an energy-efficient sustainable technique for producing active LDPE films with antioxidant properties.

KEYWORDS
active packaging, antioxidant properties, low-density polyethylene, pomegranate peel extract, supercritical solvent impregnation

1 | INTRODUCTION

Food loss and waste remain major global challenges, with significant economic, environmental and social implications. The Food and Agriculture Organization estimates that around one-third of all food produced is lost or wasted along the supply chain.[1] This issue is particularly critical for perishable commodities, where microbial spoilage and oxidation degradation account for substantial losses. Therefore, advances in food packaging technologies are essential to extend shelf life and preserve product quality. Within this context, active packaging has attracted considerable attention for its capacity to incorporate functional compounds that improve food preservation.

Active packaging enhances food preservation by embedding bioactive components into polymer matrices, allowing controlled

release of antioxidants and antimicrobials that slow spoilage while maintaining quality.[2,3] Among natural sources of bioactive compounds, pomegranate (*Punica granatum* L.) peel has attracted particular interest because its phenolic profile is rich in hydrolysable tannins (punicalagin, casuarinin and geraniin), flavonoids (catechin and epicatechin) and phenolic acids (gallic and ellagic acids) and significantly more concentrated than in the arils, seeds or juice.[4,5] Quantitative analyses show that the peel can contain up to 30% total phenolics by dry weight,[6] representing roughly half of the fruit's mass, and exhibits antioxidant capacities two to three times higher than the edible portions.[7,8]

Previous studies on diverse fruit-waste extracts, such as apple pomace, grape skins and mango kernel, consistently ranks pomegranate peel among the top performers for 2,2-diphenyl-1-picrylhydrazyl (DPPH) and 2,2′-azino-bis(3-ethylbenzothiazoline-6-sulphonic acid) (ABTS) radical scavenging activities[9–11] and broad-spectrum antimicrobial activity against *Listeria monocytogenes*, *Escherichia coli* and *Staphylococcus aureus*.[12,13] This superior efficacy is attributed to the synergistic action of its high-molecular-weight ellagitannins, which can disrupt microbial membranes and chelate metal ions that catalyse oxidative reactions.[14,15]

Given their bioactivity, pomegranate peel extract (PPE) has been widely explored for incorporation into biopolymer-based food packaging. For instance, chitosan-based films loaded with PPE exhibited a marked increase in tensile strength from 19.2 to 27.6 MPa, reduced water vapour permeability from 2.91 to 2.30 g mm day^{-1} kPa^{-1} and an increased antioxidant activity from 27% to 66%.[16] Similarly, pectin-based coatings enriched with PPE showed phenolic contents of 30–50 mg GAE g^{-1} and inhibited *S. aureus* and *E. coli* at 30–50 mg mL^{-1}, while preserving the sensory attributes of coated energy bars during 30 days of storage.[17] These studies demonstrate that PPE not only imparts functional bioactivity but also enhances mechanical and water vapour barrier properties when properly integrated.

Although biopolymers offer excellent compatibility with natural extracts,[18–20] they often suffer from limitations, such as low mechanical strength and high moisture and gas permeabilities, which can limit their commercial viability. To overcome these drawbacks, a hybrid strategy involves applying a thin PPE-enriched biopolymer coating onto conventional petrochemical-based substrates.[21,22] Although this approach combines the robustness of the base polymer with the active functionality of the natural extract, it raises concerns about the migration of water-soluble biopolymer components into food.[23]

To address these challenges, supercritical solvent impregnation (SSI) has emerged as a promising green technology for incorporating bioactive compounds into polymeric matrices. This technique utilises supercritical carbon dioxide ($scCO_2$) as a solvent to achieve uniform and non-destructive dispersion of active compounds within the polymer matrix. $scCO_2$ is advantageous because of its mild critical temperature (31.06°C) and pressure (73.8 bar) and its liquid-like density and gas-like diffusivity. It is also non-toxic, non-flammable and chemically inert, and can be recovered and reused, improving environmental sustainability.[24] Compared with conventional solvent casting, SSI offers several distinct advantages.[25] In solvent casting, active compounds are first dissolved in organic solvents and blended into the polymer solution, followed by solvent evaporation to form films. This approach often requires high processing temperatures (>80°C) to dissolve the polymers and bioactive compounds,[26] which can degrade thermolabile compounds, and it leaves residual solvents that necessitate additional drying steps. In contrast, SSI operates under relatively mild conditions (30–60°C)[27] using $scCO_2$, which possesses liquid-like density and gas-like diffusivity, enabling rapid penetration and uniform distribution of bioactive compounds into polymer matrices. Because CO_2 reverts to the gaseous state upon depressurisation, the resulting films are free from solvent residues,[28] eliminating concerns about toxicity or regulatory compliance. Beyond functional performance, SSI is also more environmentally sustainable, generating negligible effluent and reusing over 90% of the CO_2, whereas solvent casting relies heavily on volatile organic compounds and produces hazardous waste streams.[29,30] These comparative advantages highlight SSI as a greener, more efficient and technologically superior alternative to solvent casting for developing active food packaging materials.

Among synthetic polymers, low-density polyethylene (LDPE) is widely used in food packaging due to its lightweight nature, mechanical durability and excellent moisture resistance.[31] Although its backbone is primarily composed of linear alkyl chains, the presence of branched chains reduces crystallinity (typically 45%–55%) and increases amorphous regions, facilitating the incorporation of small molecules into its matrix.[32] These structural characteristics makes LDPE particularly suitable for SSI [30] as it maintains its thermal and chemical stability throughout the impregnation process.[33,34]

Studies have shown that increasing impregnation pressure enhances the impregnation yield.[5] Additionally, process parameters, such as impregnation time, significantly influence the solvating capacity and diffusivity of $scCO_2$, which ultimately affects the polymer's ability to absorb active compounds.[25,35] Although SSI has been successfully applied for impregnating various bioactive compounds, including extracts from black walnut husk,[36] clove,[37] olive leaves[38] and green tea,[39] direct optimisation of impregnation time for PPE into LDPE under fixed pressure/temperature has not been reported. This study aims to determine the optimal SSI duration required to achieve ideal physical and functional properties in LDPE/PPE films, contributing to the advancement of active food packaging technology.

2 | MATERIALS AND METHODS

2.1 | Materials

Fresh pomegranate peels were sourced from a local supermarket (Cold Storage). LDPE films, with a thickness of 95 ± 10 μm, were obtained from Humi Pak Sdn. Bhd. Absolute ethanol (99.7%) and potassium persulphate ($K_2S_2O_8$) were supplied by from Chemiz (M)

Sdn. Bhd. Mueller–Hinton agar (MHA), nutrient agar, Folin–Ciocalteu reagent and gallic acid were procured from Merck Sdn. Bhd. Sodium carbonate (Na_2CO_3) and sodium chloride (NaCl) were procured from R&M Chemicals whereas DPPH and ABTS reagents were provided by Sigma-Aldrich.

2.2 | PPE preparation

The PPE was prepared following the method described by Maryam Adilah and Nur Hanani.[40] Fresh pomegranate peels were cleaned and oven-dried at 30°C for 24 h. The dried peels were then pulverised using an electronic blender (Panasonic MX-M200) to obtain pomegranate peel powder. To extract PPE, pomegranate peel powder was mixed with absolute ethanol in a 1:5 (w/v) ratio and shaken continuously in the dark for 24 h. Absolute ethanol was selected because it enhances the solubility of PPE in $scCO_2$ and may improve extract-polymer affinity.[26] The mixture was then filtered using a Whatman No. 1 filter paper (pore size = 11 μm). The filtrate was concentrated using a rotary evaporator (Eyela Pte. Ltd.) at 45°C until the final volume was reduced to 10% of the original. The resulting PPE was stored at −18°C until further use.

2.3 | LDPE films SSI

SSI of LDPE films was conducted in batch mode using a laboratory-scale SSI system (Taiwan Supercritical Technology Co. Ltd.) as reported in our previous study (Figure 1).[5] A 12 × 1.5 cm LDPE film was secured onto a steel support inside the impregnation vessel, ensuring exposure on both surfaces for uniform diffusion. Then, PPE (15 mL) was placed at the bottom of the vessel without direct contact with the film and the chamber was hermetically sealed. The impregnation process was carried out at 300 bar and 60°C conditions selected based on prior experimental findings.[5] The CO_2 flow was gradually increased to reach the target pressure, and the impregnation time (1, 3 and 5 h) commenced once both temperature and pressure stabilised. A non-impregnated LDPE film served as the control. After the designated impregnation time, the system was gradually depressurised and the heater was switched off. The impregnated film samples were carefully wiped with a dry cloth to remove any residual extract and stored at 4°C until further analysis.

2.4 | Film analyses

2.4.1 | Thickness

The thickness of LDPE films, both before and after impregnation, was measured using a digital thickness gauge (Mitutoyo) with a 1 μm sensitivity. Measurements were taking at 10 different points on each sample and the average thickness was calculated.

2.4.2 | Impregnation yield

The impregnation yield was determined gravimetrically by measuring the mass difference of the films before and after impregnation.[41] It was calculated using the following Equation (1):

$$\text{Impregnation yield } (\%) = \frac{m_f - m_o}{m_o} \times 100, \quad (1)$$

where m_f is the film mass after SSI and m_0 is the initial film mass.

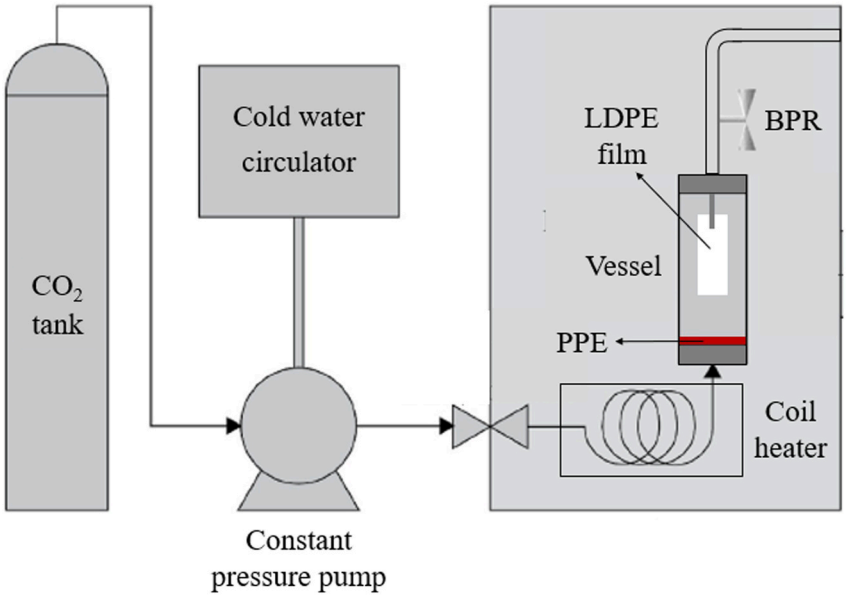


FIGURE 1 Experimental setup of the SSI equipment. BPE, back pressure regulator; LDPE, low-density polyethylene; PPE, pomegranate peel extract; SSI, supercritical solvent impregnation.

2.4.3 | Light transmittance and opacity

The light transmittance of each film sample at 200, 280, 400, 500, 600 and 800 nm was measured using a UV-Vis spectrophotometer (GENESYS 10S, Thermo Fisher Scientific) following the method described by Zeng et al.[42] Two rectangular film strips (4 × 1 cm) were placed on the opposite sides of a cuvette, whereas a blank cuvette was used as the reference. The opacity of film was calculated using the following Equation (2):

$$\text{Opacity } (\text{AU mm}^{-1}) = \frac{\text{Abs}_{600}}{x}, \quad (2)$$

where Abs_{600} is the absorbance value at $\lambda = 600$ nm (AU) and x is the thickness of the film sample (mm).

2.4.4 | Scanning electron microscopy

The surface and cross-sectional microstructures of the film samples were analysed using a scanning electron microscope (SEM) (JSM-IT100 InTouchScope, JEOL). Each 1 × 1 cm film sample was mounted onto a metal stub and coated with a thin gold layer using a sputter coater (SCD 005, BalTec AG) to enhance conductivity. SEM images were captured at an accelerating voltage of 10 kV, with surface microstructures observed at 500× and 1500× magnifications, whereas cross-sectional microstructures were examined at 500× magnification.

2.4.5 | Atomic force microscopy (AFM)

The surface roughness and morphological characteristics of the films were analysed using a Dimension Edge atomic force microscope (Bruker GmbH). Each film sample was scanned over a 1 × 1 μm area, and a 3D image of the film surface was generated.

2.4.6 | FTIR spectroscopy

The chemical composition of the film was analysed using a Fourier transform infrared (FTIR) spectrophotometer (Bruker Alpha II) equipped with an attenuated total reflectance attachment. FTIR spectra were recorded at a resolution of 4 cm^{-1} over a scan range of 4000 to 500 cm^{-1}.

2.4.7 | Antioxidant properties

Total phenolic content

Film extract was prepared by immersing 25 mg of each film sample in 3 mL of ethanol for 3 h.[43] The film sample was then removed, and 0.3 mL of the extract was mixed with 2.5 mL of Folin–Ciocalteu reagent (10% v/v) and 2 mL of sodium carbonate solution (7.5% w/v). The mixture was incubated at 50°C for 5 min, and the absorbance was read at 765 nm. Total phenolic content (TPC) values were expressed in milligramme gallic acid equivalents per gram (mg GAE g^{-1}) of film based on a calibration curve of gallic acid.

DPPH RSA

DPPH radical scavenging activity (RSA) of the films was determined using a film extract prepared by immersing 1 × 1 cm film samples in 3 mL of ethanol for 3 h[44] After removing the film, 1 mL of 0.1 mM ethanolic DPPH solution was added. A control was prepared by substituting the film extract with ethanol. The mixture was stored in the dark for 30 min, and the absorbance was measured at 517 nm. The RSA was calculated using the following Equation (3):

$$\text{RSA } (\%) = \frac{\text{Abs}_{\text{control}} - \text{Abs}_{\text{extract}}}{\text{Abs}_{\text{control}}} \times 100, \quad (3)$$

where $\text{Abs}_{\text{control}}$ is the absorbance of control sample and $\text{Abs}_{\text{extract}}$ is the absorbance of the film extract.

ABTS RSA

The ABTS radical solution was prepared by mixing equal volumes of 7 mM ABTS stock solution and 2.45 mM potassium persulfate ($K_2S_2O_8$) solution.[40,42] The mixture was kept in the dark at room temperature for 16 h and then diluted with ethanol until the absorbance at 734 nm reached approximately 0.70. To initiate the reaction, 0.4 mL of film extract was mixed with 4 mL of ABTS radical solution. A control sample was prepared using ethanol instead of the film extract. The samples were incubated in the dark at room temperature for 6 min and absorbance was measured at 734 nm. ABTS RSA was calculated using Equation (3).

2.5 | Statistical analysis

All experiments were conducted in triplicate. Data were analysed using one-way analysis of variance in Minitab 19 at a significance level of 0.05 (α). Significant differences between means ($p < 0.05$) were determined using Tukey's test.

3 | RESULTS AND DISCUSSION

3.1 | Film thickness

The film thickness increased significantly ($p < 0.05$) after SSI (Table 1). However, extending the impregnation time beyond 1 h did not lead to further changes in thickness. This increase may be attributed to the plasticising action of $scCO_2$, which induced polymer swelling.[45] Similar findings were reported by Villegas et al.[45] who observed thickening in polylactic acid films after cinnamaldehyde impregnation, attributing it to the combined plasticising effects of

TABLE 1 Thickness and impregnation yield of control LDPE film (0 h) and LDPE films supercritically impregnated with pomegranate peel extract for different durations (1, 3 and 5 h).

Impregnation time (h)	Thickness (μm)	Mass (mg)	Impregnation yield (%)
0	90.50 ± 0.71^{b}	161.62 ± 0.31^{b}	0.00 ± 0.00^{b}
1	102.00 ± 2.83^{a}	167.17 ± 4.29^{a}	2.13 ± 0.88^{a}
3	100.50 ± 2.12^{a}	163.63 ± 0.90^{a}	1.25 ± 0.36^{a}
5	93.50 ± 0.71^{a}	163.52 ± 2.66^{a}	1.05 ± 0.03^{a}

Note: Means in the same column with different superscript letters are significantly different ($p < 0.05$).

Abbreviation: LDPE, low-density polyethylene.

$scCO_2$ and cinnamaldehyde. Conversely, Medeiros et al. found that linear low density polyethylene (LLDPE) films impregnated with clove essential oil showed no significant change ($p \geq 0.05$) in thickness, likely due to the reversible swelling behaviour of the polymer during the process.[46]

3.2 | Mass of the film and impregnation yield

The film mass increased significantly ($p < 0.05$) after SSI (Table 1), though a slight decline in mass gain was observed with prolonged impregnation times. The impregnation yield (Table 1) followed a similar trend, with the highest values recorded for films impregnated for 1 h (5.55 mg mass increase and 2.13% yield). This aligns with findings from a previous study, where olive leaf extract showed the highest antioxidant loading at 1 h of SSI (3%), surpassing other impregnation times (5 min, 30 min, 2 h, 5 h and 22 h).[26] The decline in PPE loading with extended impregnation times could be due to the desorption of phenolic compounds into the $scCO_2$ phase and a dynamic loading–equilibration balance during prolonged exposure at 60°C. In contrast, Belizón et al.[47] reported an increase in methyl gallate loading on PET/PP films with longer impregnation times (3–22 h), attributed to prolonged polymer swelling.[48] This is consistent with other reports suggesting that some polymers continue swelling beyond 2.5 h during SSI.[49]

The impregnation yields in this study (1.05%–2.13%) align with previous findings. Wenzel et al.[36] reported mass gains of 0.026%–1.712% for walnut husk extract in LDPE films, whereas Goñi et al. observed yields ranging from 2.0% to 4.8% for carvone-loaded LDPE films.[41] Similarly, eugenol-loaded LLDPE films showed yields from 1.31% to 5.67%,[50] whereas thymol impregnation in LLDPE ranged from 1.48% to 3.81%.[51]

The relatively low impregnation yields compared with other polymers reported in the literature are consistent with the limited polar functionality of LDPE,[41] which reduces the strength of interactions with phenolic solutes. Additionally, highly soluble solutes in $scCO_2$ may increase the partition coefficient between the polymer and the supercritical phase, further reducing loading efficiency.[52]

3.3 | Light transmittance and opacity

Table 2 shows the percentage of light transmittance of LDPE films across wavelengths from 200 to 800 nm, covering both UV and visible light regions. The UV spectrum is categorised into UV-C (100–280 nm), UV-B (280–315 nm) and UV-A (315–400 nm), whereas visible light extends from 400 to 800 nm.

A significant ($p < 0.05$) reduction of light transmittance was observed at 400, 500, 600 and 800 nm for impregnated films compared to the control. This decrease is attributed to anthocyanins and other phenolic compounds in pomegranate peel, which effectively absorb UV light.[42,53] Similar findings have been reported by Miranda-Villa et al. in a study on polylactic acid (PLA) films impregnated with carvone where increased crystallinity and UV-absorbing properties contributed to reduced light transmittance.[54] Similarly, Cerro et al. demonstrated a substantial reduction in UV transmittance in PLA films impregnated with cinnamaldehyde.[55]

All impregnated films displayed significantly ($p < 0.05$) higher opacity compared to the control film, with 5 h impregnated film showing the highest opacity (Table 2). Natural pigments in pomegranate peel, such as granatonine and tannin,[56] imparted a reddish-brown hue to the LDPE films (Figure 2), further contributing to opacity. These modifications enhanced the UV-visible light barrier properties of PPE-impregnated LDPE films, highlighting their potential to delay photooxidation in food and mitigate issues such as nutrient loss, rancidity, off-flavours and discolouration.

3.4 | Scanning electron microscopy

The surface microstructures of the control and impregnated LDPE films at magnifications of 500× and 1500× are shown in Figure 3. The control film exhibited a homogeneous, smooth and poreless surface, whereas all impregnated films displayed a heterogeneous distribution of blister-like undulations and shallow depressions distributed heterogeneously across the surface. These features can be attributed to $scCO_2$-induced swelling of amorphous regions near the polymer surface, followed by CO_2 nucleation and release during depressurisation.[46] Rather than forming open cavities, the rapid gas

TABLE 2 Light transmittance and opacity of control LDPE film (0 h) and LDPE films supercritically impregnated with pomegranate peel extract for different durations (1, 3 and 5 h).

Impregnation time (h)	Light transmittance (%) at different wavelengths						Opacity (AU mm^{-1})
	200 nm	280 nm	400 nm	500 nm	600 nm	800 nm	
0	53.62 ± 12.21[a]	51.62 ± 12.05[a]	57.12 ± 3.53[a]	61.48 ± 2.76[a]	65.37 ± 2.32[a]	70.19 ± 1.20[a]	2.37 ± 0.04[c]
1	48.85 ± 2.34[a]	31.00 ± 3.72[a]	35.16 ± 3.80[b]	54.27 ± 1.34[b]	58.77 ± 1.24[b]	63.84 ± 0.86[b]	2.88 ± 0.05[b]
3	47.04 ± 3.83[a]	38.42 ± 8.41[a]	43.56 ± 1.55[b]	53.04 ± 2.77[b]	57.78 ± 2.46[b]	62.97 ± 1.93[bc]	2.82 ± 0.02[b]
5	51.44 ± 2.84[a]	40.21 ± 4.74[a]	40.40 ± 3.69[b]	49.16 ± 2.27[b]	53.64 ± 2.20[b]	59.13 ± 2.02[c]	3.39 ± 0.03[a]

Note: Means in the same column with different superscript letters are significantly different ($p < 0.05$).
Abbreviation: LDPE, low-density polyethylene.

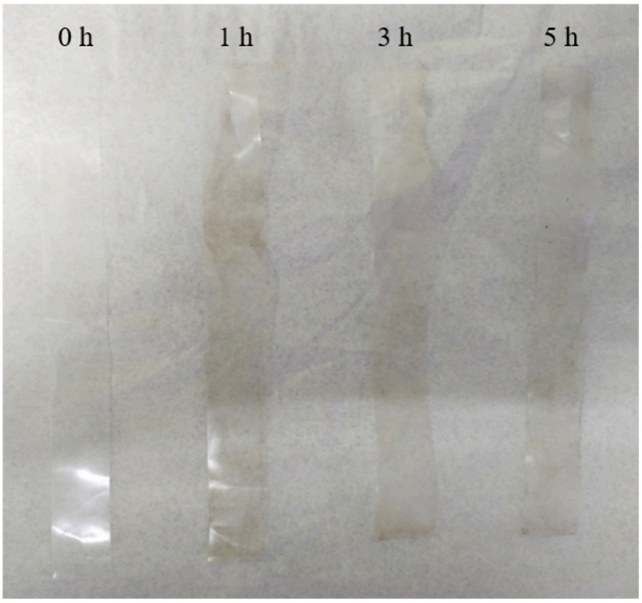


FIGURE 2 Appearance of control LDPE film (0 h) and LDPE films supercritically impregnated with pomegranate peel extract for different durations (1, 3 and 5 h). LDPE, low-density polyethylene.

release produced superficial blisters, with occasional partial collapse, consistent with the viscoelastic response of LDPE at the impregnation temperature. Increasing impregnation time from 1 to 5 h intensified surface swelling, resulting in more numerous and irregular features, particularly under 1500 × magnification. Despite these alterations, no cracks or physical damage were detected, consistent with reports from other SSI studies.[26,46]

Cross-sectional views of the films (Figure 4) further supported these findings. The control film presented a homogeneous and compact appearance, whereas the impregnated films exhibited rougher cross-sections. Grainy contrasts observed near the surface are more likely to reflect blistering or preparation artefacts than incompatibility, suggesting nanoscale roughening without discrete particles as confirmed by AFM. This suggests that PPE is predominantly incorporated within the superficial layers of the polymer matrix, consistent with earlier findings where mango leaf extract localised mainly in the surface regions of thermoplastic polyurethane films.[57]

3.5 | Atomic force microscopy

AFM images (1 × 1 μm) of LDPE films impregnated with PPE for different durations and their corresponding average surface roughness (Ra) values are shown in Figure 5. The Ra values increased significantly ($p < 0.05$) with impregnation time showing a clear increase in roughness with longer impregnation times. The Ra values increased from 4.40 nm in the control to 6.74 nm (1 h), 13.23 nm (3 h) and 15.40 (5 h), indicating progressive surface heterogeneity ($p < 0.05$). These nanoscale roughness trends are consistent with SEM observations, where films subjected to longer impregnation displayed more pronounced blister-like features and shallow depressions. Importantly, no particulate aggregates were detected by AFM, confirming that PPE was incorporated in a dispersed state rather than as visible clusters.

The observed roughening is attributed to $scCO_2$ swelling and rapid depressurisation, which disrupted chain rearrangement and produce local undulations. Similar topographical modifications in polyolefin films after SSI have been reported by Medeiros et al.[46] The increase in surface roughness also explains the reduced light transmittance of impregnated films as both nanoscale irregularities and internal blistering enhance light scattering and absorption, thereby contributing to increased opacity with longer impregnation times.[58]

3.6 | FTIR spectroscopy

Figure 6 shows the FTIR spectra of untreated and impregnated LDPE films. The characteristic peaks of LDPE were observed at 2915, 2847, 1463 and 720 cm^{-1} corresponding to -CH_2 asymmetric stretching, -CH_2 symmetric stretching, bending deformation and rocking deformation, respectively.[59,60]

In the impregnated films, a broad peak appeared between 3650 and 3000 cm^{-1}, indicative of the hydroxyl (-OH) stretching vibration from polyphenols, suggesting the presence of alcohol compounds and carboxylic acids.[36] The band between 1750 and 1650 cm^{-1} likely corresponded to carbonyl (C=O) stretching vibrations, which are characteristics of aldehydes, ketones and carboxylic acids.[36] Additionally, a peak at 1620 cm^{-1} suggested the presence of unsaturated compounds, such as alkenes, due to C=C stretching vibrations.[61]

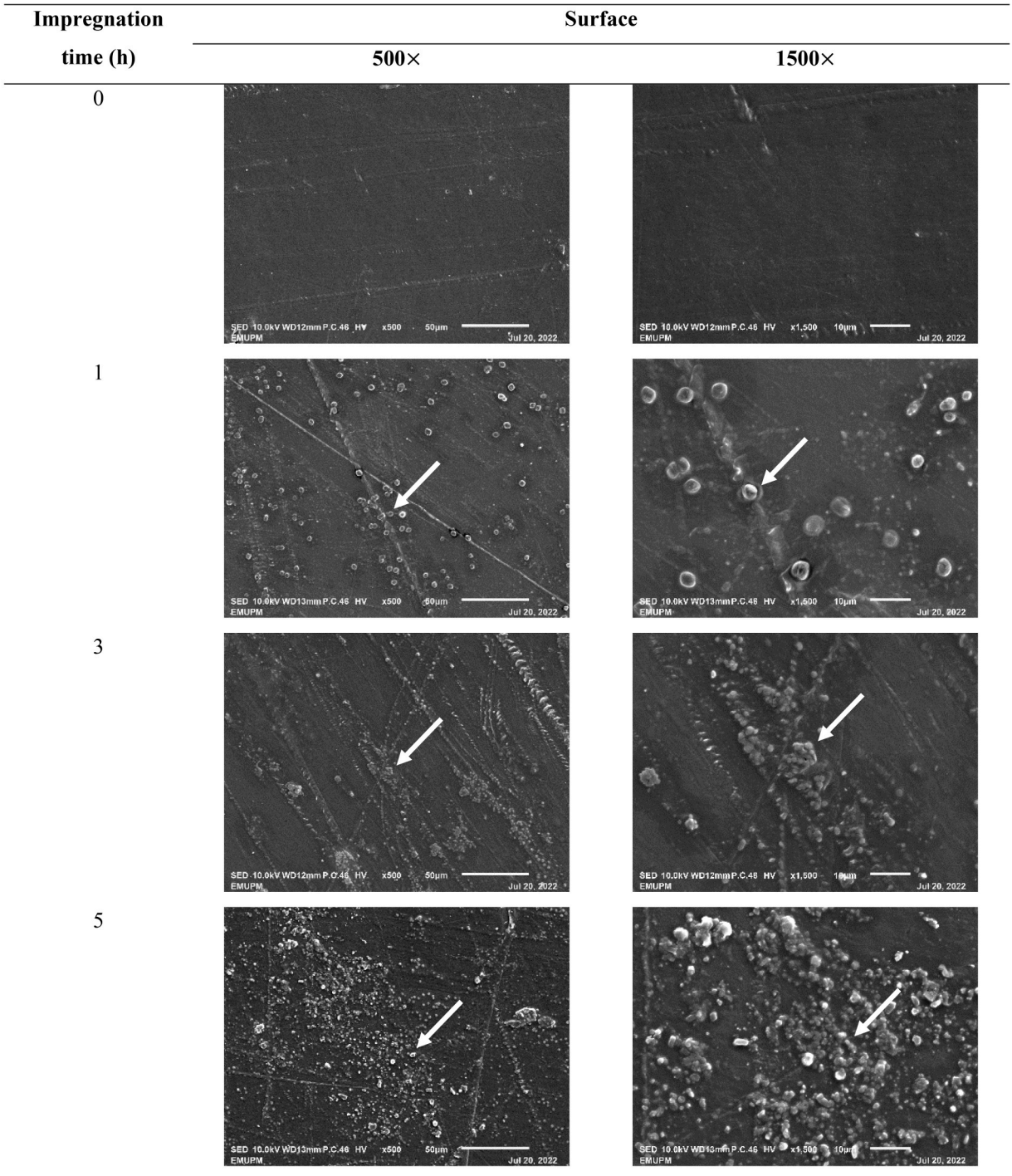


FIGURE 3 SEM images showing the surface microstructures of control LDPE film (0 h) and LDPE films supercritically impregnated with pomegranate peel extract for different durations (1, 3 and 5 h) under magnifications of 500 × and 1500 ×. LDPE, low-density polyethylene; SEM, scanning electron microscopy.

Another band between 1150 and 1050 cm^{-1} was attributed to C-O stretching vibrations, further supporting the presence of oxygenated phenolic compounds.[36] To fully characterise the phenolic constituents retained in the films, detailed compositional analysis, such as ^{1}H NMR spectroscopy or chromatographic profiling, shall be performed.

Notably, the films impregnated for 1 and 3 h exhibited broader bands and more intense peaks in all the aforementioned regions (3650–3000, 1750–1650, 1620 and 1150–1050 cm^{-1}) compared to those impregnated for 5 h. This suggests a higher impregnation yield and phenolic content in the 1- and 3-h

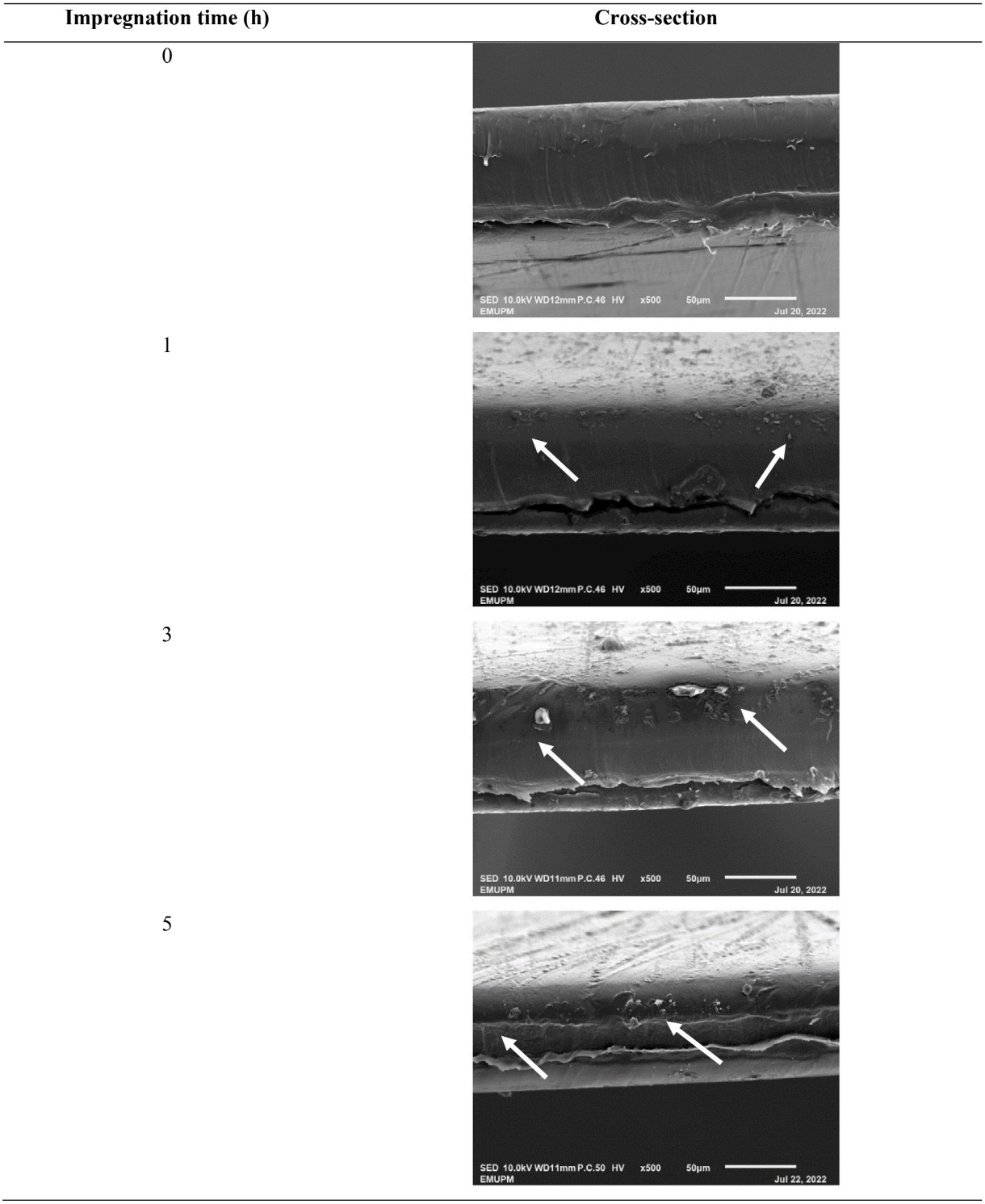


FIGURE 4 SEM images showing the cross-sectional microstructures of control LDPE film (0 h) and LDPE films supercritically impregnated with pomegranate peel extract for different durations (1, 3 and 5 h) under magnification of 500×. LDPE, low-density polyethylene; SEM, scanning electron microscopy.

samples. Similar FTIR spectra have been reported in SSI studies on LDPE films impregnated with black walnut husk,[36] carvone[41] and eugenol,[50] further validating the observed spectral trends.

3.7 | Antioxidant properties

Oxidation is a major contributor to food spoilage, leading to the development of off-flavours and discolouration due to radical

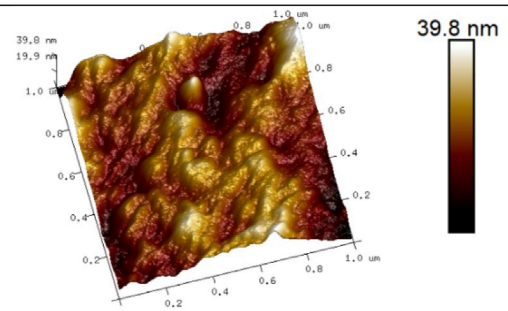
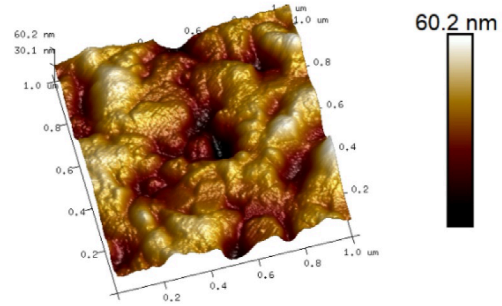
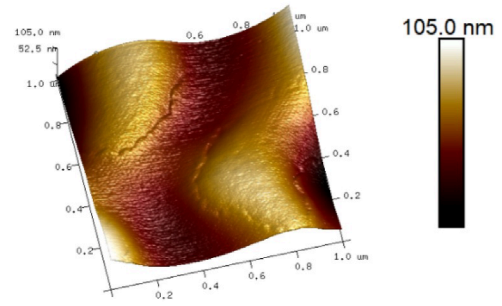
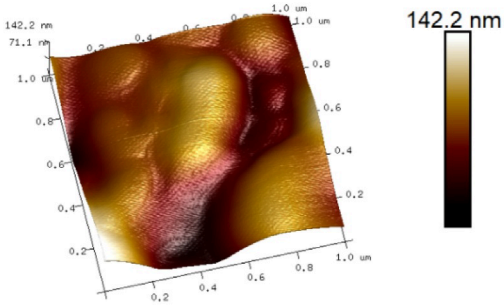
Impregnation time (h)	AFM images	Ra (nm)
0		4.40 ± 0.13[d]
1		6.74 ± 0.09[c]
3		13.23 ± 0.21[b]
5		15.40 ± 0.20[a]

FIGURE 5 AFM images and Ra of control LDPE film (0 h) and LDPE films supercritically impregnated with pomegranate peel extract for different durations (1, 3 and 5 h). Means in the same column with different superscript letters are significantly different ($p < 0.05$). AFM, atomic force microscopy; LDPE, low-density polyethylene; Ra, average surface roughness.

formation.[20] Food products with high lipid content are particularly susceptible as oxidation often results in rancidity. The antioxidant activity of the PPE/LDPE films were evaluated using DPPH and ABTS assays, which employ different mechanisms to assess RSA. The DPPH assay measures the ability of PPE to neutralise the DPPH radical by donating a hydrogen atom, leading to a colour change from purple to yellow.[62] In contrast, the ABTS assay quantifies antioxidant capacity based on the reduction of the ABTS radical cation, which results in a colour shift from blue–green to colourless.[63]

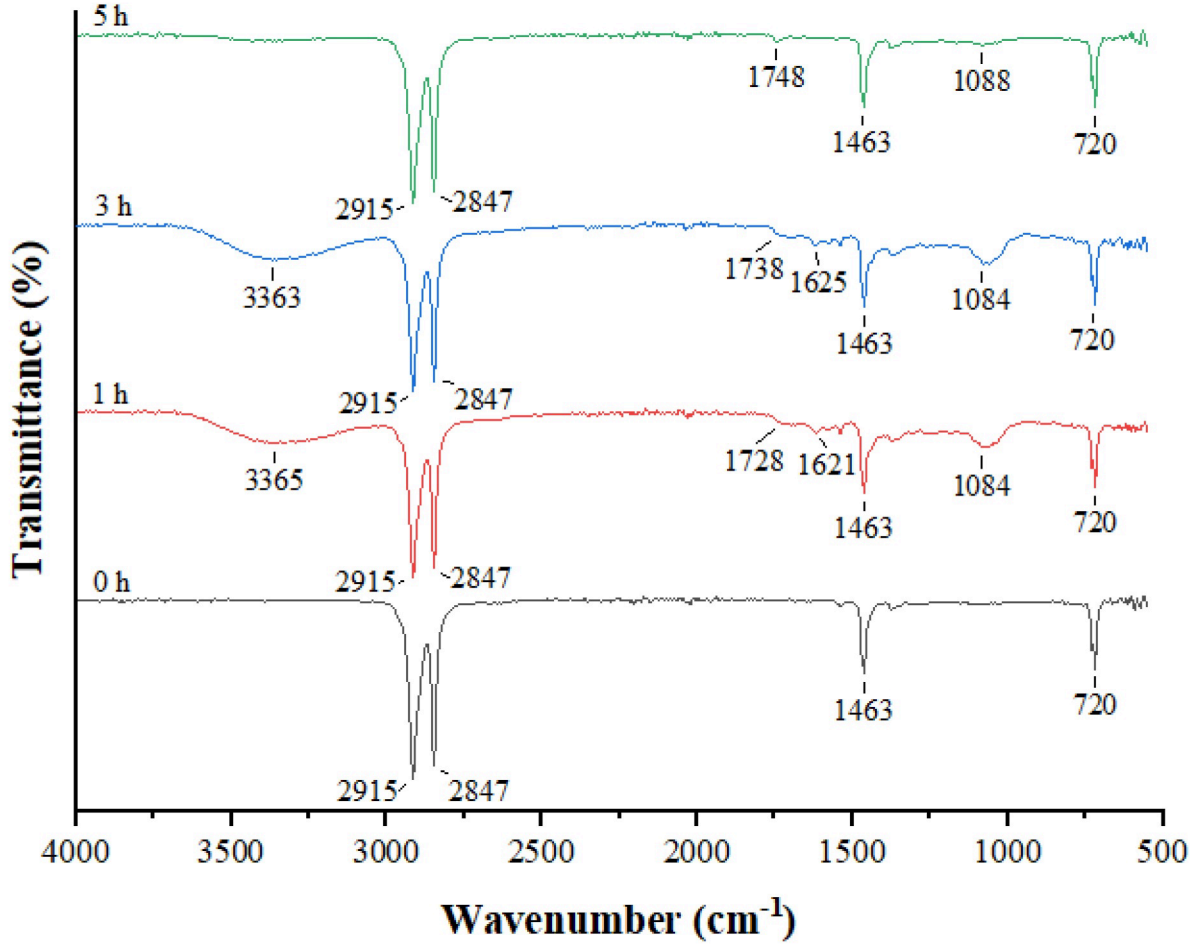


FIGURE 6 FTIR spectra of control LDPE film (0 h) and LDPE films supercritically impregnated with pomegranate peel extract for different durations (1, 3 and 5 h). FTIR, Fourier transform infrared; LDPE, low-density polyethylene.

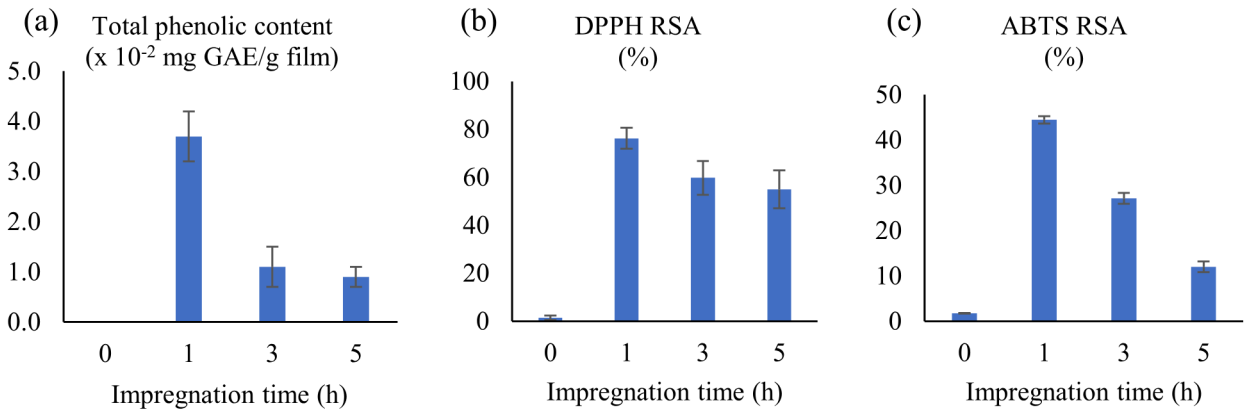


FIGURE 7 TPC, DPPH and ABTS RSA of control LDPE film (0 h) and LDPE films supercritically impregnated with pomegranate peel extract for different durations (1, 3 and 5 h). ABTS, 2,2′-azino-bis(3-ethylbenzothiazoline-6-sulphonic acid); DPPH, 2,2-diphenyl-1-picrylhydrazyl; LDPE, low-density polyethylene; RSA, radical scavenging activities; TPC, total phenolic content.

A time-dependent relationship between SSI and antioxidant activity was observed in the PPE/LDPE films (Figure 7). As expected, the control films exhibited no TPC. However, a slight RSA was detected in the control films, potentially due to the natural decrease in DPPH absorbance upon oxygen exposure.[50] All impregnated films demonstrated RSA, confirming the successful incorporation of antioxidant compounds from PPE. Notably, the PPE/LDPE film impregnated for 1 h exhibited the highest RSA ($p < 0.05$) among all samples, corroborating the results of the impregnation yield. Interestingly, the samples exhibited higher DPPH RSA than ABTS. This discrepancy may be attributed to the different extract volumes applied in each assay (1 mL for DPPH vs. 0.4 mL for ABTS). Solvent polarity likely also played a role: DPPH radicals are generated in an ethanolic medium, which enhances the solubility and reactivity of the moderately polar phenolics present in PPE.[64,65] In contrast, ABTS radicals are produced in an aqueous medium, where certain phenolic compounds may display reduced reactivity or slower diffusion. Despite these differences, both assays revealed the same overall trend, with maximum activity observed after 1 h of impregnation, consistent with the impregnation yield and TPC.

The TPC of impregnated films ranged from 0.009 to 0.037 mg GAE/g film, representing 6.67%–27.41% of the TPC of PPE (0.135 ± 0.006 mg GAE/g DW). This trend was consistent with RSA results, with the highest phenolic yield observed at an impregnation duration of 1 h. Previous studies have similarly reported a strong correlation between the radical scavenging properties of active films and their TPC.[66] For instance, Cejudo Bastante et al. found that PET/PP films impregnated with olive leaf extract retained approximately 0.06%–0.09% of the original extract's TPC.[38] Another study reported TPC values of 148.79–195.83 μg/g corresponding to a loading efficiency of 9.03%–11.89%.[67] In comparison, Lukic et al. reported significantly higher TPC values (118.06–128.05 mg GAE/g film) in polylactic acid/poly(ε-caprolactone) films impregnated with thymol and carvacrol, likely due to is the higher concentration of active compounds in thymol and carvacrol compared to PPE.[68]

A decline in RSA and TPC was observed after 1 h, consistent with the impregnation yield results. Similar trends have been reported in the literature. Albuquerque et al. found that essential oil-loaded gelatin films exhibited the highest DPPH RSA (41.63%) with a 60-min impregnation time compared to the films with longer impregnation duration (90 and 120 min).[69] Similarly, Cejudo Bastante et al. found that a 1-h impregnation time resulted in the highest antioxidant activity in olive leaf extract-impregnated PET/PP films.[26]

The antioxidant properties of PPE are primarily attributed to its rich composition of phenolic acids, such as gallic acid and ellagic acid, flavonoids, such as catechin and epicatechin and hydrolysable tannins such as geraniin, casuarinin and punicalagin.[5,70] Punicalagin, a major ellagitannin in pomegranate peel, plays a crucial role in its antioxidant capacity.[71] The inhibition of oxidation by phenolic compounds is linked to their redox properties, allowing them to act as reducing agents and hydrogen donors. Furthermore, phenolic compounds may exhibit metallic chelating activity, thereby preventing metal ions from catalysing oxidative reactions.[72]

In summary, the results indicate that PPE/LDPE possess strong antioxidant properties demonstrating their potential as active food packaging materials to mitigate oxidative deterioration in food products.

4 | CONCLUSION

This study demonstrated that SSI is an effective technique to incorporate PPE into LDPE films. Impregnation for 1 h at 300 bar and 60°C yielded the highest loading efficiency, TPC and radical scavenging activities, while also improving opacity and light-barrier properties. Extended impregnation times led to reduced PPE retention, highlighting the importance of time optimisation. FTIR confirmed the presence of oxygenated phenolic functionalities in the films, and surface analyses revealed increased roughness associated with CO_2 depressurisation. To enhance the practical applicability of LDPE/PPE films, future research should investigate the long-term stability and release kinetics of active compounds under various storage conditions as well as evaluating the barrier, mechanical and antimicrobial properties of the films, which are essential for practical applications. In addition, the film size produced here was limited by the capacity of the laboratory-scale SSI apparatus, though this constraint can be addressed through scale-up and equipment optimisation. The robust antioxidant properties of the films suggest potential for preserving foods that are vulnerable to oxidation and rancidity, including edible oils, nuts, snacks and cured meats. Overall, SSI provided a rapid solvent-free method to develop bioactive LDPE films with enhanced functional properties, demonstrating the potential of pomegranate peel as a sustainable source of antioxidants for active food packaging applications.

AUTHOR CONTRIBUTIONS

Chan Lee Ting: Investigation; writing—original draft; methodology. **Foong Han Lyn**: Writing—review and editing; validation; data curation. **Chong Gun Hean**: Conceptualization; validation. **Nor Afizah Mustapha**: Validation. **Nur Hanani Zainal Abedin**: Validation; conceptualization; funding acquisition; writing—review and editing; project administration; data curation; supervision; resources; visualization.

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CONFLICTS OF INTEREST STATEMENT

The authors declare no conflicts of interest.

DATA AVAILABILITY STATEMENT

Data will be made available on request.

ETHICS STATEMENT

Not applicable.

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