

度及紫外屏蔽的橡仁粉/卡拉胶基可胶&热封的油脂包装膜的制备及性能研究

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摘要: 塑料的大量使用造成了白色污染,使生态环境被严重破坏。治理和消除白色污染已成为全球的共同目标。因此,研制出可生物降解的包装薄膜材料迫在眉睫。天然聚合物,如多糖和蛋白质,可用来生产包装膜并具有代替合成聚合物的潜力。目前,已有很多比传统塑料包装膜具有更好的阻隔性、抗菌性或抗氧化性的多糖基薄膜被研制出来。但由于多糖的熔融温度高于热解温度导致其无法通过热封形成有效地包装袋,难于实际应用。淀粉和蛋白质衍生的薄膜可热封并用作塑料的替代品。然而,这种利用方式可能会加剧全球粮食短缺。本研究中,我们利用废弃的橡子与卡拉胶制备了可胶&热封的复合包装膜。测试了橡仁粉/ κ -卡拉胶(AKM/ κ C)复合膜的机械强度,热稳定性,透光率,雾度和抗菌性能。结果表明,当甘油从30%增加到50%时,雾度和透光率分别从96.74%和3.50%下降到94.50%和2.25%。AKM/ κ C薄膜具有优异的紫外线屏蔽性能(380-200 nm处的透过率为0)。使用15g/L的壳聚糖季铵盐(CQAS)作为生物胶水对45g-AKM/ κ C薄膜进行胶&热封。在115 °C, 0.3MPa下热封1s的密封强度最大,为 5.09 ± 0.29 N/15mm。用45g-AKM/ κ C包装的牛油在25 °C贮存75天后,过氧化值(POV)从0.0187 g/100g增加到0.0336 g/100g,低于暴露在空气中的牛油的POV(0.5209 g/100g,已变质)。我们制备的薄膜具有较高的雾度和紫外线屏蔽性能,可用作富含脂肪食品的塑料包装替代品。

关键词: 橡仁粉; κ -卡拉胶; 复合膜; 高雾度; 紫外屏蔽; 可密封

Fabrication and characterization of a glue- and heat- sealable acorn kernel meal/ κ -carrageenan composite film with high-haze and UV-shield for packaging grease

Abstract: The extensive utilization of plastics has resulted in the emergence of white pollution, which poses a severe threat to the ecological environment. Consequently, tackling and eradicating this issue has become a shared objective worldwide. Therefore, it is imperative to expedite the development of biodegradable packaging film materials. Natural polymers, such as polysaccharides and proteins, possess the potential to replace synthetic polymers in producing packaging films. Numerous polysaccharide-based films have been developed with better barrier, antimicrobial or antioxidant properties compared to traditional plastic packaging films. However, their practical application is limited due to they pyrolyze before melting which hinders heat-sealing for effective bag formation. Starch- and protein-based films are viable substitutes for plastics, as they can be heat-sealed. However, the utilization of starches and proteins in this way may exacerbate global food shortages. In this study, we used discarded acorns to prepare a glue- and heat-sealable packaging film. We evaluated the mechanical characteristics, thermal stability, transmission, haze, and antimicrobial properties of an acorn kernel meal/ κ -carrageenan (AKM/ κ C) composite film. The results showed that the haze and transmittance decreased from 96.74% to 94.50% and from 3.50 to 2.25%, respectively, when glycerol increased from 30% to 50%. The AKM/ κ C film had excellent UV shielding properties (transmittance at 380-200 nm was 0). The 45 g-AKM/ κ C films were sealed through a combination of bio-glue composed of 15 g/L chitosan quaternary ammonium salt (CQAS) and heat at 115 °C for 1 s under a pressure of 0.3 MPa, resulting in a maximum sealing strength of 5.09 ± 0.29 N/15 mm. Beef tallow packaged in 45g-AKM/ κ C exhibited an increased peroxide value (POV) from 0.0187 to 0.0336 g/100 g after 75 days of storage at 25 °C, which was lower than that in air (0.5209 g/100g, which has deteriorated).

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Our developed film had high haze and UV shielding performance and can be used as a plastic packaging substitute for fat-rich foods.

Key words: acorn kernel meal; κ -Carrageenan; composite film; high-haze; UV-shield; sealability.

1 Introduction

Plastics are widely used in packaging due to their low-cost, stability, light weight and durability; however, plastics do not decompose contributing to environmental pollution (Cárdenas-Alcaide et al., 2022; Shen et al., 2020). Plastic pollution has become a global problem that urgently requires a solution (Chu et al., 2023; Walker and Fequet, 2023). A viable solution is to replace plastics with renewable biomass (Mohamed et al., 2020). Natural polymers, which possess adequate film-forming ability, such as chitosan (El Mouzahim et al., 2023), sodium alginate (Guo et al., 2023), pectin (Xie et al., 2023), cellulose (Huang et al., 2022a), hemicelluloses (Peng and She, 2014), carrageenan (Han et al., 2023), soybean isolate protein (Liu et al., 2021) and starch (Ortega et al., 2023), etc., are potential plastic substitutes for packaging. For example, chitosan and curry leaf essential oil have been used as packaging materials with insect repellency (Nazurah et al., 2022). Sodium alginate film and *Clitoria ternatea* extract have been used to maintain and monitor food freshness (Santos et al., 2022). Citron peel pectin and glycerol as a plasticizer has been developed into a superior food packaging film via a response surface method (Asfaw et al., 2023). Waste cotton textiles and tea polyphenols have been used to produce transparent cellulose-based films with antibacterial, antioxidant and UV resistance properties (Xia et al., 2022). κ -Carrageenan and mulberry polyphenol extracts were mixed to prepared a packaging film with pH-responsive and antioxidant properties (Liu et al., 2019). Even though these food packaging films have better properties such as oxygen barrier, antimicrobial or antioxidant than traditional plastic packaging, they are not used in food packaging because polysaccharide-based films can't be heat-sealed into bags due to the fact that they pyrolyze before melting (Chi et al., 2023b). Starch- and protein-derived films have been heat-sealed into bags. For instance, soybean isolate protein-based films incorporated with diatomite/thymol complex were used to package blueberries for extended shelf life (Lu et al., 2021). Chicken protein isolate/fish gelatin composite film containing phenolic compounds was used to package chicken skin oil via heat sealing (Nilsuwan et al., 2021). However, those proteins are not only key nutrients for human health (Rawiwan et al., 2022), but they are also troublesome to extract and expensive. Sago starch-based film can be formed into bags by heat-sealing with a sealing strength of 375 N/m (Abdorrezza et al., 2011). Cassava starch-based films were used to package pork and retard its spoilage (Katekhong et al., 2022). These starches are derived from staple foods; hence, the use of these foods for the preparation of plastic substitutes might exacerbate the global food shortage (Botreau and Cohen, 2020). Therefore, there is a need for a starch-rich biomass that can be used as a plastic substitute.

Starch-rich acorns are the fruit of oaks. China has an annual yield of 6 to 7 million metric tons starch-rich acorns (Chao et al., 2017). However, it is barely eaten or even fed to livestock in most areas of China, which is due to the acorn kernels content plenty of tannins with astringent flavor and anti-nutrients (Pasqualone et al., 2019). As a result, acorns are discarded (Sardão et al., 2021). Acorn kernel is an ideal film-forming biomass; its tannin content might contribute UV-shielding properties to the film (Huang et al., 2022b). Tannins were incorporated into sodium alginate/silk protein film that exhibited exceptional UV-blocking capabilities (Zhang et al. 2023). The nanocomposite films of *Eucommia ulmoides* rubber containing Fullerene C60 exhibited a 100% shielding effect against the UV light (Wei et al., 2021). A chitosan film blended with low molecular weight poly(limonene) also

demonstrated outstanding UV shielding properties (Goncalves et al., 2023). The incorporation of acorn kernel meal (AKM) containing tannins as a direct raw material would streamline the film preparation process by eliminating the need for additional UV shielding agents. Nonetheless, starch-based films are often brittle and have poor mechanical properties, which can be improved by adding more hygroscopic κ -carrageenan, which is a linear anionic polysaccharide composed of galactose and dehydrated galactose extracted from red algae (Jiang et al., 2023; Wang et al., 2023). Our previous study has suggested that κ -carrageenan film could be glue-sealed into bags by electrostatic action (Chi et al., 2023b). Currently, there is no report on AKM blended with κ -carrageenan (κ C) to prepare a glue- and heat-sealable food packaging film.

In this study, we investigated the effect of glycerol addition on AKM/ κ C composite films. We further studied the films using scanning electron microscopy (SEM), Fourier-transform infrared (FTIR) spectroscopy, and thermogravimetric analysis (TGA). We measured light transmittance, haze, oxygen permeability (OP), water vapor permeability (WVP), mechanical properties. We selected 45g-AKM/ κ C to investigate the heat-sealing parameters and used it to package beef tallow.

2. Materials and methods

2.1. Materials

Acorn was acquired from Anguo Fangcaojia Trading Co., Ltd. (Baoding, China). κ C was purchased from Qingdao Bairen Biotechnology Co., Ltd. (Shandong, China). Chitosan quaternary ammonium salt (CQAS) was bought from Nantong Lushen Biological Engineering Co., Ltd (Jiangsu, China). Glycerol was offered by Yongda Chemical Co., Ltd. (Tianjin, China). Beef tallow was obtained from a local market (Harbin, China).

2.2. Methods and equipment

2.2.1 Determination of components in AKM

The 180 mesh AKM was obtained by milling the acorns after peeling, cleaning and drying. The starch, tannin, lipid, protein and fiber contents in the AKM were determined according to Chinese GB 5009.9-2016 (the 1st method: enzymatic hydrolysis), Chinese GB 5848-86, Chinese GB 5009.6-2016 (the 1st method: soxhlet extraction), Chinese GB 5009.5-2016 (the 2nd method: spectrophotometry) and Chinese GB 5009.10-2003, respectively.

2.2.2 Preparation of films

AKM (6.0 g, dry weight) was dispersed into 200 mL of distilled water (room temperature) and then stirred at 80 °C for 30 min. Subsequently, κ C (20%, w/w of AKM basis) was added and stirred for 30 min. Glycerol (30%, 35%, 40%, 45%, 50%, w/w of AKM basis) was incorporated into the AKM/ κ C solution and further stirred for 30 min. Finally, the mixture was poured into a mould (30.0 cm $\times$ 29.0 cm $\times$ 4.0 cm) and dried about 12 h at 50 °C. The obtained films were denoted as 30g-AKM/ κ C, 35g-AKM/ κ C, 40g-AKM/ κ C, 45g-AKM/ κ C and 50g-AKM/ κ C corresponding to the glycerol amounts.

2.2.3 Characterizations

Morphologies of the AKM and the cross section of the films were photographed by a SEM of Apreo S Hivac (Thermo Fisher Scientific Inc, USA). FTIR spectra of κ C, AKM and films were recorded by a Nicolet iN10 spectrometer (Thermo Fisher Scientific Inc, USA) within the wavenumber from 550 to 4000 cm^{-1} . The thermostability of glycerol and films were performed via a Q500 thermogravimetric analyzer (TA Company, USA)

from 20 °C to 600 °C under 10 °C /min in N₂. The gelatinization temperature of 45g-AKM/κC was measured after adequate wetting using a Q2000 differential scanning calorimeter (TA Instruments – Water LLC, USA) from 30 °C to 160 °C under a nitrogen atmosphere at a heating rate of 10 °C /min. The haze (400~700 nm) and light transmittance (200~800 nm) of films were obtained via a haze meter (CS-700, CHNSpec, China) and spectrophotometer (UV-2600, Shimadzu, Japan), respectively.

2.2.4. Performance measurements

The OP of the films with 33% RH and 53% RH was measured via an oxygen transmission rate tester (OX/230, Labthink, China). The WVP of the films under 75% RH (25 °C) was obtained using the gravimetric method. The average thickness of the films was randomly obtained at 240 points by an ID-C112XBS micrometer (Mitutoyo Corp., Japan). After the rectangular films (1.5 cm × 8.0 cm) were conditioned at 25 °C with different humidity (11% RH, 33% RH, 53% RH and 75% RH) for 48 h, the TS and EB were detected by an auto-tester (XLW-PC, Labthink, China) with a strain rate of 30.0 cm/min. Each sample was examined in 4 replicas. The bacteriostatic activity of the film was assessed via plate counts. 9 mL of bacterial suspension (10000 CFU/mL, *E. coli* or *S. aureus*) was cocultured with 0.2 g of broken films in a constant temperature shaker incubator at 37 °C for 4 h. Afterwards, 0.2 mL of the mixture was evenly coated on nutrient agar medium and cultured at 37 °C for 1 d. Finally, the count of colonies on the nutrient agar medium was recorded using an automatic colony counter (Icont-50, Xunda Technology Co., Ltd., China). A control experiment was carried out without broken film addition. The bacteriostatic rate of the films can be calculated by Eq. (1):

$$\text{Bacteriostatic rate} = (C - X) / C \times 100\%$$

(1)

where C and X are the colony counts of the control experiment and the experiment, respectively.

2.2.5. Synergized glue- and heat-sealing strength measurement

Rectangular 45g-AKM/κC (7.5 cm × 11.5 cm) was glue and heat synergistically sealed using a heat seal tester (AT-RF, Anmc, China). 0.2 mL of 15 g/L CQAS solution was coated along a 1.0 cm width of overlap of the two rectangular edges and pressure sealed at 0.10~0.40 MPa and 100~120 °C for 0.8~1.2 s. For measurement of sealing strength, the sealed film was cut to 1.5 cm × 6.5 cm and then stored at 33% RH for 12 h. The glue- and heat-sealing strength were measured by using an XLW-PC electronic tensile machine (Labthink, China) at a strain rate of 5 mm/s. Four replicates of each sample were tested.

2.2.6. Packaging beef tallow

A square 45g-AKM/κC (12.0 cm × 12.0 cm) was folded in half, CQAS solution was coated on the 1.0 cm width of the remaining edge, and the pressure sealed for 1.0 s at 0.30 MPa and 115 °C using a heat seal tester to result a bag contain 10.0 g of beef tallow. After storage at 25 °C for 75 days, the beef tallow was taken out and measured the peroxide value (POV) by following Chinese GB 5009.227-2016 (Titration method). Triplicate measurements of each sample were performed.

2.2.7. Statistics analysis

The SPSS v17.0 (SPSS Inc., USA) was utilized for data analysis with the Duncan multiple range test ($P < 0.05$).

3. Results and discussion

3.1. Acorn kernel meal component analysis

AKM contains $86.18 \pm 0.02\%$ starch, $7.90 \pm 0.22\%$ tannin, $2.47 \pm 0.04\%$ lipids, $0.92 \pm 0.03\%$ protein, and $0.13 \pm 0.00\%$ fiber (Table 1). Starch and tannin can provide favorable heat sealability and UV shielding for AKM-based films, respectively.

Table 1. Content of components in AKM.

Sample	Starch (%)	Tannin (%)	Lipids (%)	Protein (%)	Fiber (%)
AKM	86.18 ± 0.02	7.90 ± 0.22	2.47 ± 0.04	0.92 ± 0.03	0.13 ± 0.00

3.2. AKM and films morphology

Fig. 1 shows the morphology of AKM and the AKM/ κ C composite film by SEM. The AKM granules were irregular, polygonal and elliptical with some voids and debris on the surface could be observed in Fig. 1 (a). The AKM/ κ C composite film presented several pits and protrusions (Fig. 1 b~f) formed by the lipids evaporation during high vacuum. Similar results were observed in composite films of black corn seed powder/ κ C (Chi et al., 2023a) and olive pomace/chitosan (de Moraes Crizel et al., 2018). Tannin and proteins form precipitates that uniformly disperse in the composite films (Khongkliang et al., 2023). After quenching in liquid nitrogen, random pits and protrusions were formed in the cross-sections of the films. Furthermore, with increasing glycerol addition, these particles became smaller and more numerous. It is possible that glycerol prevented their aggregation during the film-forming and drying processes.

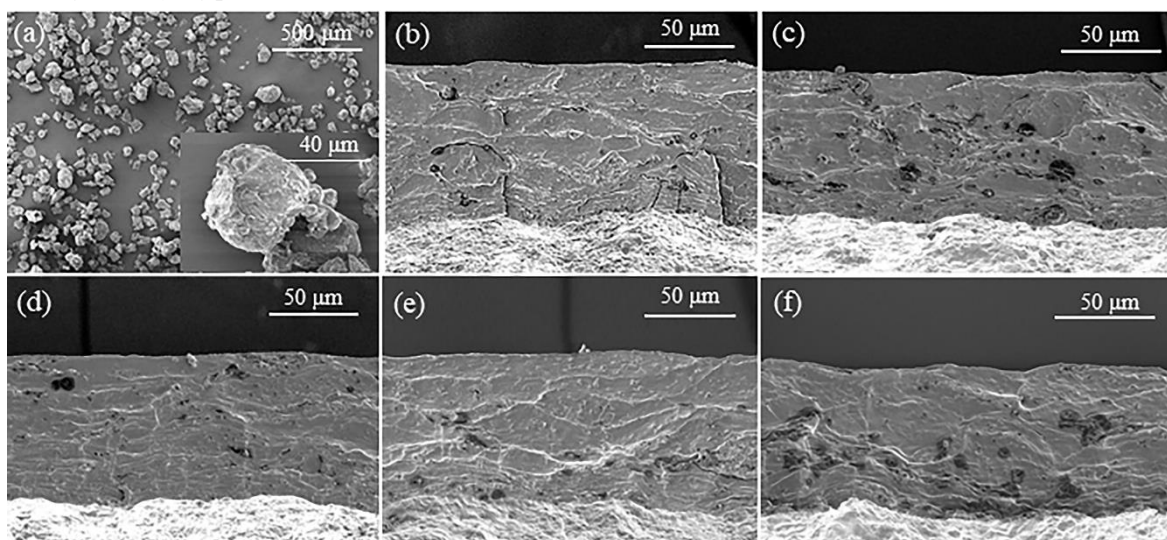


Fig. 1 SEM images of AKM (a), cross-sections of 30g-AKM/ κ C (b), 35g-AKM/ κ C (c), 40g-AKM/ κ C (d), 45g-AKM/ κ C (e) and 50g-AKM/ κ C (f).

3.3. FTIR analysis

Fig. 2 shows the FTIR spectra of κ C, AKM and composite films. Bands at 3305, 2924, 2853 and 1638 cm^{-1} are associated with the O–H and N–H stretching vibrations, C–H stretching vibrations, C–H bending vibrations, and water absorption, respectively (Daoud et al., 2019; Sun et al., 2019). In κ C, the bands at 1230 cm^{-1} , 925 cm^{-1} and 842 cm^{-1} are attributed to sulfate ester, 3,6-anhydrogalactose and galactose-4-sulfate ester, respectively (Ili Balqis et al., 2017). AKM displayed the bands at 1742 cm^{-1} (C=O of lipids) and 1240 cm^{-1} (N–H from protein)

(Rozenberg et al., 2019). Those bands could be easily observed in the composite film spectra (Fig. 2 b). The intensity of the band at 3305 cm⁻¹ increased with the addition of glycerol, which might be caused by glycerol obstructed regeneration of starch. Furthermore, there were no significant differences in the spectra of the composite films, demonstrating that no chemical reaction occurred among the components in the films. The deconvolution spectra of AKM and composite films are illustrated in Fig. 2 (c, d). The intensities of ~1040 cm⁻¹ and ~995 cm⁻¹ are correlated with starch crystalline phases, whereas intensity of ~1020 cm⁻¹ is associated with the amorphous phase (Chen and Sopade, 2013). The 995/1020 and 1040/1020 ratios can be evaluated the starch crystallinity; higher ratios suggest a more crystalline structure. The absorption intensity of AKM was slightly higher at 995 cm⁻¹ than at 1020 cm⁻¹, and 995/1020 ratio was 1.03. That for AKM/ κ C composite films were less than 1.03, and the band at 1040 cm⁻¹ disappeared. This result might be attributed to the fact that κ C and glycerol prevent the starch retrogradation during the films formation process, thereby reducing crystallinity. When glycerol increased from 30% to 50%, the 995/1020 ratio decreased from 0.94 to 0.86, which suggested the amorphous region of starch in the composite films increased. This result (Fig. 2d) was consistent with those in Fig. 2 (b).

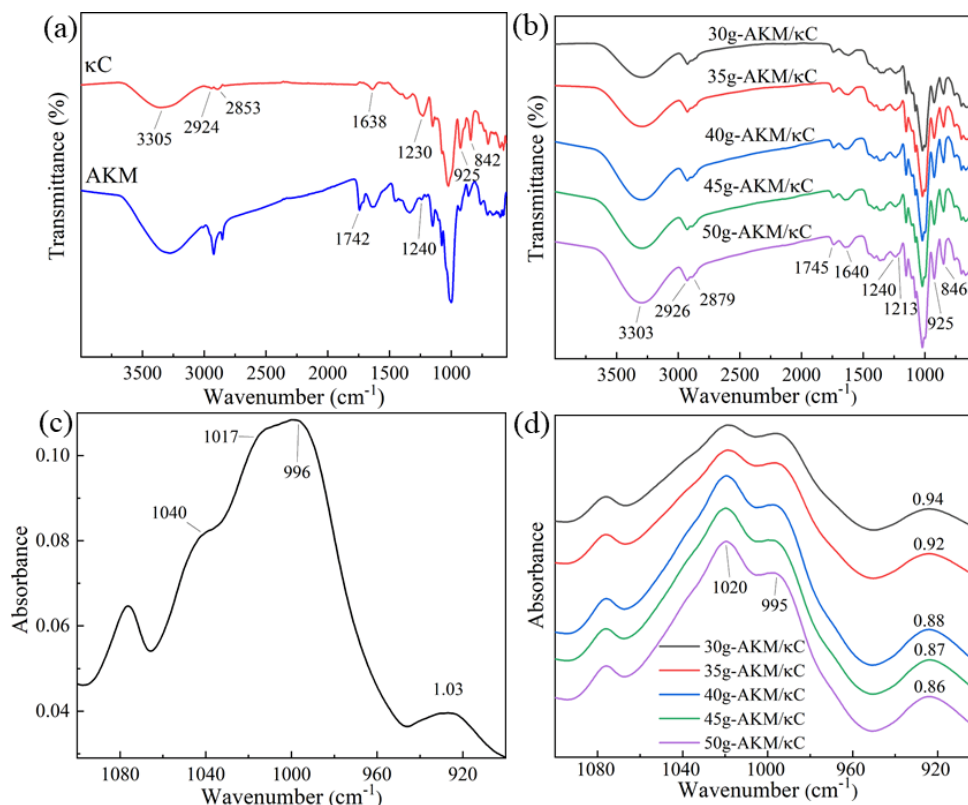


Fig. 2 Original (a,b) and deconvoluted (c,d) FTIR spectra of κ C, AKM and composite films.

3.4. Optical performance and antibacterial activity

Fig. 3 (a, b) presents the optical performance of the composite films. When glycerol addition increased from 30% to 50%, transmittance decreased from 3.50% to 2.25%. According to SEM images, the particles in the composite film became smaller and more numerous with increasing glycerol addition, which resulted in more reflection as light pass through the film. In addition, the composite films have excellent UV shielding (0% transmittance at 200-380 nm), which is due to the tannin with abundant phenolic hydroxyl groups that can adsorb

UV (Huang et al., 2022b). The composite film haze decreased from 96.74% to 94.50%, due to glycerol that reduced the regeneration of starch. This speculation is supported by FTIR result. The antibacterial properties of the composite films are shown in Fig. 3 (c). The bacteriostatic effects of composite film against *E. coli* and *S. aureus* decreased from 66.09% to 11.40% and from 56.63% to 27.63%, respectively, when glycerol increased from 30% to 50%. With increasing glycerol addition, the content of tannin, which possess antibacterial properties, decreases. The composite films had a stronger antibacterial effect against *S. aureus* than *E. coli*.

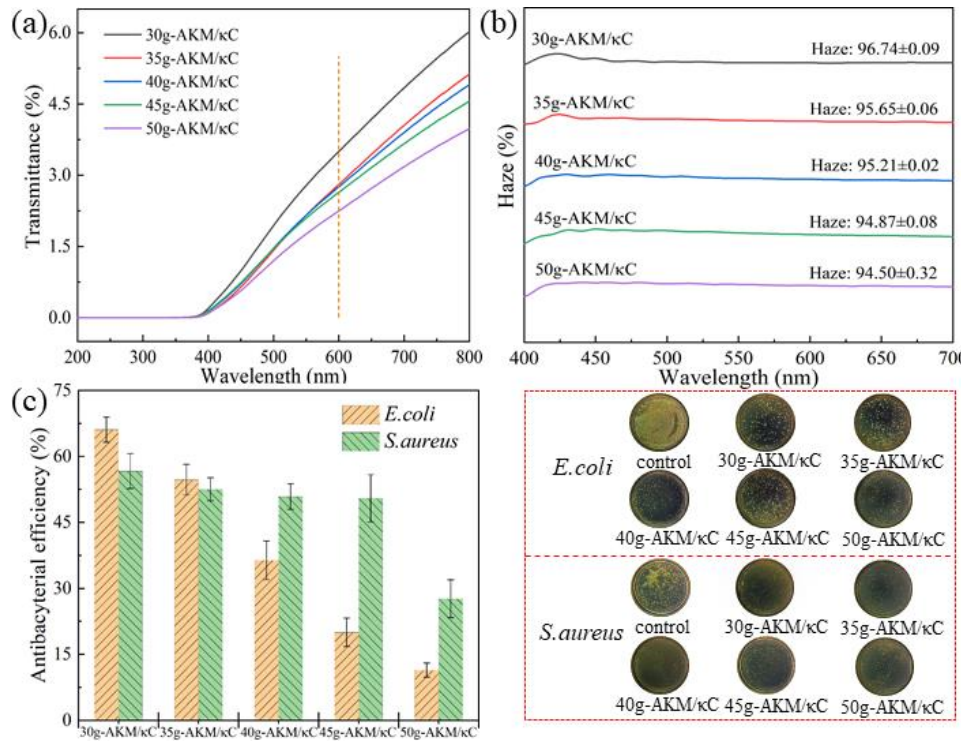


Fig. 3 Optical performance (a.transmittance, b.haze) and antibacterial properties of the composite film (c).

3.5. Physical properties

Thickness, WVP, OP and moisture content of the composite films are presented in Table 2. The thickness, WVP and OP increased with glycerol content. It was possible that glycerol disrupted the dense structure of the polymers, created more space. The increase in WVP may also be related to more hydroxyl groups (evidenced by the FTIR spectra). Similar results have been obtained in several researches (Cao et al., 2018; Gomes et al., 2022; Sun et al., 2018). OP increased with humidity, because moisture acted as a plasticizer, further weakening the film structure. Moisture content increased with glycerol addition; the highly hydrophilic glycerol increased the hydrophilicity of the composite film. The space between the polymers contributed to more water. In addition, composite films had a higher moisture content at a higher relative humidity.

Table 2. Thickness, WVP, OP and moisture content of the composite films.

Samples	Thickness (mm)	WVP (g m ⁻¹ s ⁻¹ Pa ⁻¹ × 10 ⁻³)	Moisture Content (%)				OP (cm ³ mm ⁻² day ⁻¹ atm ⁻¹)	
			11%RH	33%RH	53%RH	75%RH	35%RH	53%RH
30g-AKM/κC	0.088±0.007 ^a	0.91±0.03 ^a	8.38±0.58 ^a	10.73±0.80 ^a	15.76±0.44 ^a	27.17±1.75 ^a	3.43±0.68 ^a	445.29±1.00 ^a
35g-AKM/κC	0.091±0.010 ^b	1.02±0.08 ^a	10.52±0.21 ^b	13.79±0.17 ^b	18.31±0.12 ^b	30.56±0.30 ^b	29.68±0.98 ^b	476.65±11.05 ^a

40g-	0.093±0.	1.28±0	11.56±0	16.90±0	21.39±0	32.57±0	44.86±2.	566.00±15
AKM/κC	009c	11h	13c	34c	85c	35h	74c	61h
45g-	0.095±0.	1.73±0	16.17±0	18.81±0	24.94±0	35.02±0	84.47±2.	840.50±26
AKM/κC	009c	04c	05d	90d	65d	69c	83d	27c
50g-	0.098±0.	1.81±0	18.12±0	22.50±0	26.17±0	38.32±1	163.28±	1354.87±2
AKM/κC	007d	02c	17e	59e	37e	05d	132e	171d

Fig. 4 (a) shows the monthly average absolute humidity in 34 major cities in China from 2009 to 2018 (www.weather.com.cn). In this study, we measured the mechanical properties of the composite films in a humidity range (11% to 75% RH) that is representative of 70% of major cities in China. As shown in Fig. 4 (b), the EB of the composite films increased with the content of glycerol and moisture, which was attributed to easier slide between polymers caused by plasticizers. The EB of 50g-AKM/κC at 75% RH was lower than that at 53% RH, which was due to fewer hydrogen bonds between polymers resulting in an easier fracture of the composite film. At lower relative humidity (11% RH), the TS of the composite film was determined by a combination of intermolecular hydrogen bonding of the polymers and film flexibility. At 30% glycerol, the film was brittle and susceptible to fracture, resulting in a low TS. At 45% glycerol, TS increased from 1.53 ± 0.18 to 18.95 ± 0.85 MPa, which was due to increased flexibility, causing the films to withstand more tensile strength. At 50% glycerol, the intermolecular hydrogen bonds decreased, thereby reducing TS to 9.81 ± 0.67 MPa. In addition, the TS of the composite film decreased with increased glycerol and moisture content at $RH \geq 33\%$. This can be explained by a reduction in hydrogen bonding between the polymers due to glycerol and moisture. Therefore, we used 45g-AKM/κC for subsequent research, due to its better mechanical properties at lower humidity (11% RH).

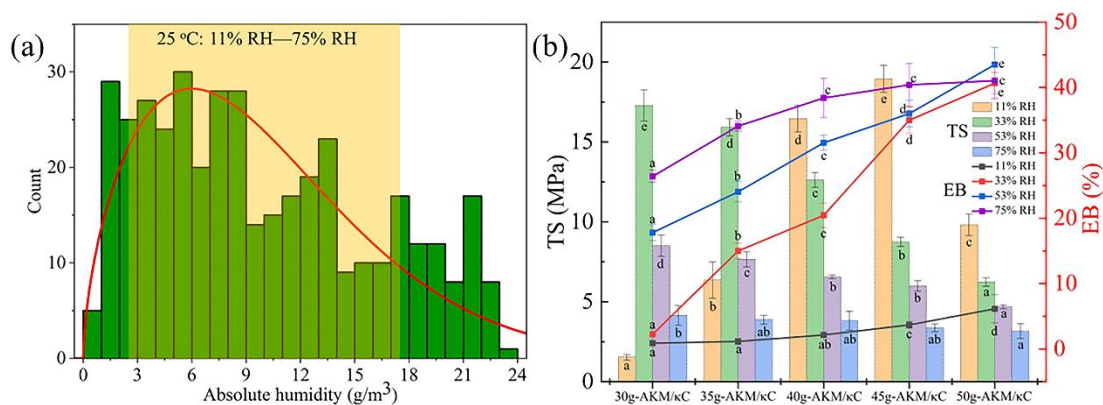


Fig. 4 Data distribution of the average monthly absolute humidity in 34 major cities in China from 2009-2018 (www.weather.com.cn, a); TS and EB of the composite films at different relative humidity (b).

3.6. Thermal stability

The thermogravimetric analysis of glycerol and AKM/κC composite films are illustrated in Fig. 5 (a, b). The weight loss of glycerol at 125-220 °C could be attributed to its evaporation. The composite film had three weight loss stages. The first stage below 100 °C was ascribed to the evaporation of water. The second stage resulted from the evaporation of glycerol, consistent with the results on glycerol-plasticized cassia gum film (Cao et al., 2018). The third stage was the pyrolysis of polysaccharides. In addition, when glycerol increased from 30% to 50%, pyrolysis temperature decreased from 262.56 °C to 257.97 °C. It is likely that glycerol disrupted the polymer interactions, thereby reducing the thermal resistance of the composite films. Similar results were obtained by using glycerol to plasticize the *Dioscorea hispida* starch film (Hazrati et al., 2021). Fig. 5 (c) shows that the gelatinization

temperature of 45g-AKM/ κ C ranged from 98.87 to 144.77 °C, indicating that the composite film could be heat-sealed.

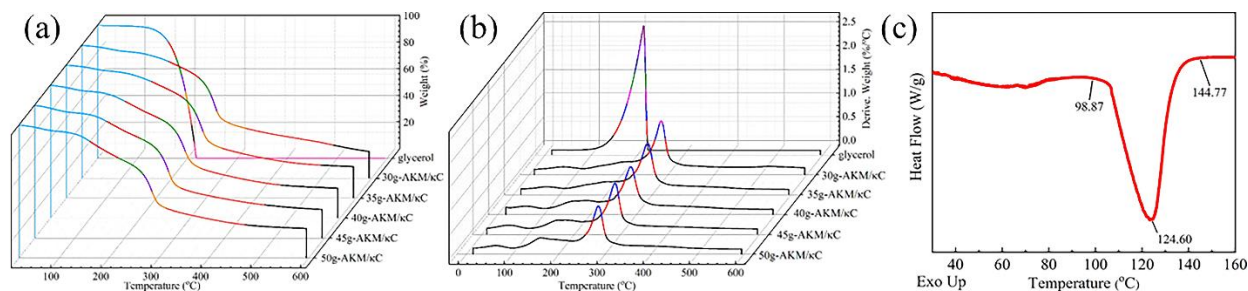


Fig. 5 TGA (a) and DTG (b) of glycerol and the composite film, and DSC of 45g-AKM/ κ C (c).

3.7. Synergistic heat & glue-sealing property

Fig. 6 (a) shows the variations in the sealing strength of 45g-AKM/ κ C at different heat-sealing temperature and time with 0.3 MPa of heat-sealing pressure. With prolonged heat-sealing time (1.2 s) or high heat-sealing temperature (120 °C), sealing strength decreased probably due to the thinning of the heat-sealing area. In addition, the fracture mode was II at 1.2 s (except at 115 °C), which may be due to the non-uniform bonding between the layers of the heat-sealing region by the combined effect of heat-sealing temperature and time. At 0.8 s and 100 °C, the fracture mode was I, which may be due to weak bonding in the heat-sealing area. The fracture mode was III at 115 °C, indicating that the strength of the bond in the sealed region is higher than that of the film. Fig. 6 (b) presents the variations in sealing strength of the composite film with heat-sealing pressure for different heat-sealing time at 115 °C. The sealing strength of 45g-AKM/ κ C increased with pressure because the appropriate pressure promotes the bonding of the heat-sealing layers. However, with increasing pressure, the sealing area became thinner, resulting in a reduction in heat-sealing strength. Sealing strength reached a maximum value of 5.09 ± 0.29 N/15 mm at 115 °C and 0.3 MPa for 1 s. As shown in Fig. 6 (d), both CQAS and heat could seal the composite film. The glue-sealing strength of 45g-AKM/ κ C was lower than the heat-sealing strength, which was attributed to the lower κ C content. Moreover, sealing strength of 45g-AKM/ κ C was > 2 N/15 mm for either sealing method, which is satisfactory for daily use according to Chinese GB/T 10003-2008.

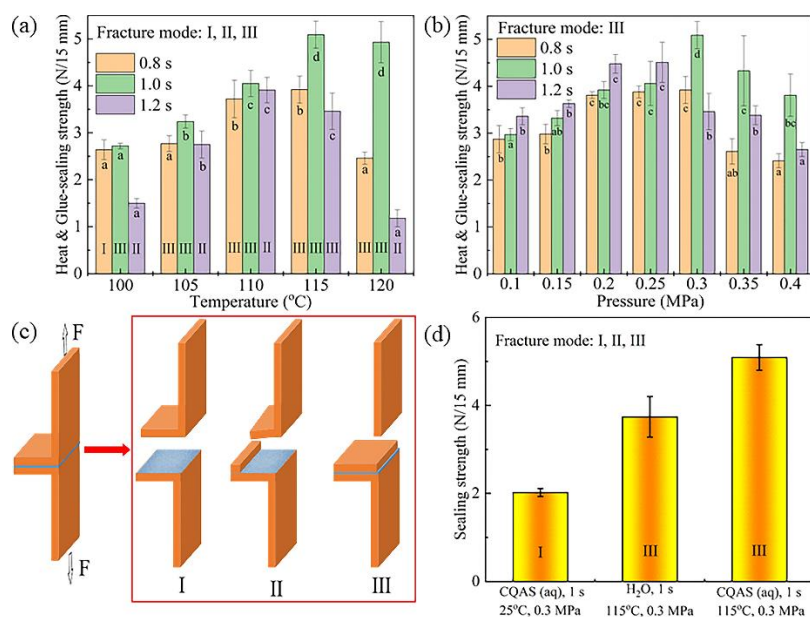


Fig. 6 Effect of heat-sealing temperature (a) and pressure (b) on the sealing strength of composite films at different heat-sealing time; Fracture mode (c); Effect of heat and CQAS on sealing strength (d).

3.8. Application in packaging beef tallow

Beef tallow is susceptible to rancidity during storage because it is rich in unsaturated fatty acids. Fig. 7 (b) shows that the air-exposed beef tallow had spoiled after 75 d since the POV rose from 0.0187 to 0.5209 g/100 g. The POV of beef tallow packaged in 45g-AKM/ κ C and PE film corresponded 0.0336 and 0.1035 g/100 g, showing that both hadn't spoiled. Compared with our previous research (Chi et al., 2023b), the POV of beef tallow packaged in 45g-AKM/ κ C increased by 79.68%, while that of beef tallow packaged in κ C film increased by 627.55% after storage of the same days. This is probably explained by the delayed oxidation of beef tallow in the composite film with excellent UV resistance. Therefore, 45g-AKM/ κ C has the potential to be used as a material for packaging fat-rich food.

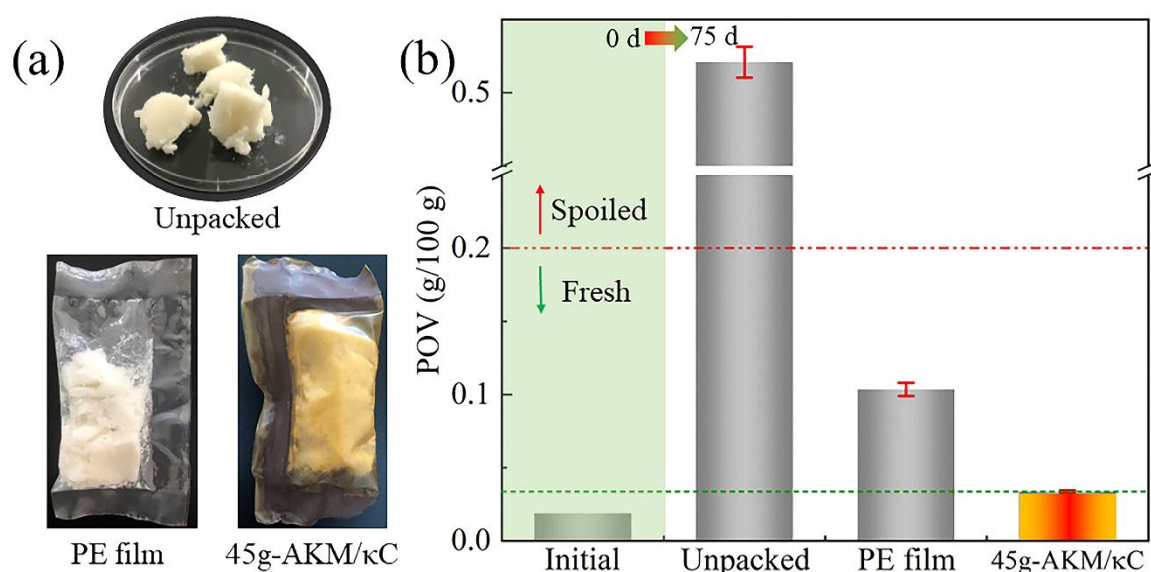


Fig. 7 Images of beef tallow exposed to air and packaged with PE film and 45g-AKM/ κ C (a), and POV before and after 75 d

(b).

4. Conclusions

We used acorns to prepare a glue- and heat-sealable AKM/ κ C composite film with high haze and UV resistance. AKM has good compatibility with κ C. With increased glycerol, thickness, OP, and WVP increased, but light transmission, haze, thermal stability, and antimicrobial properties decreased. With increased humidity, oxygen barrier decreased and flexibility increased. The glue- and heat-sealing strength of 45g-AKM/ κ C was 5.09 ± 0.29 N/15 mm at 0.3 MPa for 1 s (115 °C). After 75 days, exposed beef tallow had $15.5 \times$ the POV of beef tallow packaged in 45g-AKM/ κ C, indicating that 45g-AKM/ κ C was effective in extending the shelf life of grease. Our glue- and heat-sealable biomass-based film can be a substitute to plastic packaging. Additionally, our study provides a novel method for utilizing acorns.

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