Received: 29 May 2023 | Accepted: 30 June 2023

DOI: 10.1002/fbe2.12058

RESEARCH ARTICLE

Food Bioengineering WILEY

Development of a bilayer biodegradable packaging material enriched with coffee waste extract for cake preservation

Salimata Yakubu[1] | Hui Zheng[1] | Jingwen Chen[1] | Francis K. Amagloh[2] | Jingge Xu[1] | Xiaohan Chen[1] | Li Wang[3,4] | Li Li[1]

[1]College of Food Science and Technology, Shanghai Ocean University, Shanghai, P.R. China

[2]Department of Food Science and Technology, Faculty of Agriculture, Food and Consumer Sciences, University for Development Studies, Tamale, Ghana

[3]State Key Laboratory of Bioreactor Engineering, School of Biotechnology, East China University of Science and Technology, Shanghai, P.R. China

[4]Key Laboratory of Biobased Material Engineering, China National Light Industry, East China University of Science and Technology, Shanghai, China

Correspondence

Li Wang, State Key Laboratory of Bioreactor Engineering, School of Biotechnology, East China University of Science and Technology, Shanghai 200237, P.R. China.
Email: li.wang@ecust.edu.cn

Li Li, College of Food Science and Technology, Shanghai Ocean University, Shanghai 201306, P.R. China.
Email: l-li@shou.edu.cn

Funding information

Program of Special Capability Development for local Colleges and Universities in Shanghai, Grant/Award Number: 19050502000

Abstract

The study aimed to develop an active packaging, functionalized by spent coffee grounds extracts (SCGE) to preserve cake. The antioxidant extracts were obtained using three extraction methods: conventional extraction (CE), ultrasound-assisted extraction (UAE), and microwave-assisted extraction (MAE). The SCGE obtained with UAE and MAE produced the highest total phenolic contents and antioxidant activities, and were incorporated into bilayer films composed of polylactic acid, konjac glucomannan, and wheat gluten. The SCGE improved the film's antioxidant activity (8.40 to 90. 56%) and oxygen transmission rate (5.05 ± 0.04 to 2.22 ± 0.13). Active packaging with microwave extracts was more effective in preserving cake's lipids than the ultrasound extracts as measured by the peroxide value, thiobarbituric acid, and acid value during 21 days of storage. Overall, the study demonstrates the potential safe utilization of coffee waste in active packaging.

KEYWORDS

active packaging, cake, extraction method, oxidative deterioration, spent coffee grounds

1 | INTRODUCTION

Recently, the utilization of plant extracts for food preservation as an alternative to synthetic and chemical antioxidants and antimicrobials is a growing trend due to their ability to retard oxidative deterioration without potential health risks (Lourenço et al., 2019). Spent coffee grounds (SCG), a byproduct of the brewing of soluble coffee is an excellent source of bioactive components such as polyphenols, caffeine and tannins, showing a potential for valorization (Ballesteros et al., 2017; Oliveira et al., 2021). However, the majority of the waste is frequently deposited in landfills, generating considerable ecological issues and environmental damage. Estimates place the annual garbage production at six million tons worldwide. Therefore, valorizing this waste as a source of natural antioxidants for various applications, including active packaging, could be a better alternative for handling the bioactive waste, reducing environmental impact and being cost-effective. There are many recognized procedures for recovering bioactive compounds from SCG, but only a few produces good yields. The CE techniques have been reported with many limitations, including insufficient antioxidant recovery, thermal degradation of phenolic compounds, and an excessive requirement for large volumes of hazardous solvents, time and energy (Lourenço et al., 2019). To overcome the drawbacks of CE approaches, nonconventional extraction techniques have become increasingly popular in recent years. The intriguing benefits of MAE and UAE include reduced extraction time, less extractant consumption, suitability for heat-sensitive compounds, greater yields, and reproducibility (Lourenço et al., 2019), which we aim to

investigate in this study. Despite the considerable antioxidant activity of SCG, its application as a natural antioxidant in food preservation is limited to only a few studies. Furthermore, the major existing applications relate to the direct (without treatment) addition of spent coffee grounds extracts (SCGE) into food products (Aguilar-Raymundo et al., 2019; Hwang et al., 2019; Martinez-Saez et al., 2017).

However, the direct incorporation of active compounds into food encounters the limitation of immediate and short-term preservation of food once the active compounds are depleted at the beginning of storage (Budryn & Nebesny, 2013; Kozłowska et al., 2019). Therefore, the development of active packaging films and coatings as functional additives with antioxidant and antimicrobial activities is a novel alternative capable of improving the oxidative stability of oxidation-sensitive food products (Singh et al., 2022). The SCGE incorporation into active packaging is expected to maintain the quality and extend the shelf life of food products. However, little is known about the effectiveness of this approach in preserving cake products.

Cakes are a favorite baked food across the globe. However, cakes are prone to oxidation, influencing the color, texture, organoleptic properties, and nutritional value due to high fat content. Pound cake is especially prone to lipid oxidation due to the high content of fat (25%) coupled with high temperature during the baking process, generating primary oxidation products (Nhouchi et al., 2019). Oxidized fat reduces the quality of cake products and can be a threat to human health when consumed. Consequently, synthetic antioxidants such as butylated hydroxytoluene (BHT), butylated hydroxyl anisole (BHA), propyl gallate (PG), and tert-butyl hydroquinone (TBHQ) are often used to prevent/slow down lipid oxidation during cake preservation (Saranraj & Geetha, 2012). However, the use of synthetic antioxidants may be associated with toxicity, cancer and liver damage, urging researchers and industries to the development of alternative preservation strategies such as active packaging (Kornienko et al., 2019).

Among the biopolymers employed in active packaging, poly (lactic) acid (PLA) exhibits interesting characteristics for food packaging such as biodegradability, renewability, hydrophobicity, and low cost. Conversely, low gas barrier and poor mechanical properties precludes its extensive industrial application (Cacciotti et al., 2018).

Polysaccharide and protein biopolymers are conceivable good candidates for developing sustainable multilayer complexes with PLA. It is well known that polysaccharide and protein films present poor water vapor barriers, hence, producing bilayer films with hydrophobic and hydrophilic biopolymers may provide complementary effects in terms of barrier capacity and mechanical performance to meet food packaging applications (Rocca-Smith et al., 2019). Konjac glucomannan (KGM), a natural water-soluble polysaccharide extracted from the tubers of the *Amorphophallus konjac* plant, has been widely used in the development of packaging films. KGM has attained the Generally Recognized as Safe (GRAS) status along with interesting advantages in packaging, including excellent film-forming ability, nontoxicity, and biodegradability (Lei et al., 2019). Wheat gluten (WG) is a protein polymer capable of forming films with good mechanical and gas barrier characteristics (Chavoshizadeh et al., 2020). The blended films of KGM with other protein polymers resulted in complementary effects of improved tensile strength (TS), elongation at break (EAB) hydrophobicity, biocompatibility, and reduced oxygen uptake of the films (Wang et al., 2014). This study aimed to compare the effectiveness of PLA/KGM/WG bilayer films incorporated with SCGE, obtained from different extraction methods for cake preservation for 21 days. To the best of our knowledge, no work has been reported regarding the comparisons of the antioxidant activities of active films loaded with SCGE obtained by different extraction methods (CE, MAE, and UAE). Additionally, the polymer blend employed in this study and the utilization of SCGE in the preservation of cake has not been reported.

Some potential applications and performance of SCGE have been reported. For example, SCGE as an oxygen scavenger and diatomaceous earth as a reinforcing filler were incorporated in to PLA with the aim of improving the mechanical and gas barrier properties (Suaduang et al., 2019).

The results displayed improvement of both mechanical and oxygen barrier properties for systems characterized by the co-presence of diatomite and SCGE, suggesting a possible synergic effect of the two additives. Getachew et al. (2021) reported a high antioxidant and microbial activity of fish skin gelatine-based packaging films functionalized by subcritical water SCGE, with improved surface hydrophobicity and transparency of the developed films. Other studies report the advantages of SCG modified biodegradable films including promotion of PLA crystallization during solvent casting process and improved biodegradability (Cacciotti et al., 2018), increase in color, enhanced thermal stability, improved water vapor permeability, and preservation of physicochemical properties of films (Mendes et al., 2019).

2 | MATERIALS AND METHODS

2.1 | Materials

Arabica SCG was obtained from "canteen three" of Shanghai Ocean University. PLA (LX575) was supplied by Total Petrochemicals, Ltd, KGM (purity of 95%, viscosity: 1% solution, 30°C, ≥35,000 MPa) by BoMei Biotechnology Co., Ltd 149 and WG (gluten protein: ≥78% of dry weight) from XinTai Biotechnology Co., Ltd. All reagents: Folin-Ciocalteu's phenol reagent, anhydrous sodium carbonate (Na_2CO_3), gallic acid, 1,1-diphenyl-2- picrylhydrazyl (DPPH), 2,2-Azinobis-(3-ethylbenzothiazolin-6-sulfonicacid) ($ABTS^+$) trichloromethane, petroleum ether, glacial acetic acid, thiobarbituric acid (TBA), potassium iodide, phenolphthalein, butanol, sodium hydroxide, soluble starch, sodium thiosulfate, ethanol, and glycerol used were of analytical grade, and obtained from Aladdin Biochemical Technology Co., Ltd. Cake flour, sugar, eggs, and butter were all obtained from local commercial sources.

2.2 | Preparation and extraction of SCGE

The SCG was dried in an electrothermal constant temperature drying oven (Changge Weiheng Machinery Equipment Co., Ltd) at 50°C until constant weight. It was then passed through an 80 mm mesh sieve and stored in a dark, airtight, opaque bag until extraction. Three extraction methods, MAE, UAE, and CE, were studied and compared.

In preliminary experiments, the effect of process variables such as power (W) and time (min), were investigated for MAE and UAE to obtain the highly efficient extraction conditions. The efficiency was investigated in terms of the highest total phenolic content (TPC) and antioxidant activities. The MAE was performed in an ultrasonic microwave reaction system (Nanjing Xiaonou Instruments Manufacture Co., Ltd), equipped with a thin film transistor color monitor. The preliminary extraction experiments were performed with varying power (300, 600, 900 W) and time (5, 10, 15 min). In the UAE, the preliminary experiment was performed using an ultrasonic bath apparatus (Shenzhen Yujie Cleaning Equipment Co., Ltd) with varying power (240, 120 W) and time (30, 60, 90 min). Finally, the CE was performed using a constant temperature heating magnetic mixing hot water bath (Gongyi Yuhua Insteument Co., Ltd) at a stirring speed of 20 rmp. The effect of only time at 30, 60, and 90 min was investigated. The temperature, extraction solvent and solid/liquid ratio were kept constant at 80°C, 20% ethanol solution and 1:40 g/mL respectively, for all methods based on preliminary experiments. All extractions were performed in triplicate.

2.3 | Quantification of TPC

The TPC was measured following a previous method (Pavlović et al., 2013) with some modifications. Briefly, 400 µL of samples were strongly mixed with 400 µL of Folin-Ciocalteu's phenol reagent and allowed to react for 1 min. A 1.2 mL aliquot of 15% Na_2CO_3 was added and incubated for 30 min at 37°C.

Then 200 µL of the reactants were seeded in a 96-well microplate and the absorbance was measured at 765 nm using UV-spectrophotometer (Synergy 2 Automatic; BioTek). The TPC was calculated using the standard gallic acid calibration curve, and the results were expressed as milligram gallic acid equivalents per gram (mg GAE/g) dry basis.

2.4 | Determination of antioxidant activity

2.4.1 | DPPH radical scavenging activity

The DPPH radical scavenging assay was carried out following the method described by Pavlović et al. (2013) with some modifications. Samples of 50 µL were diluted with 3 mL of 20% ethanol solution, then 1 mL of 0.2 mmol DPPH solution was added and mixed with a vortex. The mixtures were incubated in the dark for 30 min at room temperature. A control sample was prepared in the same way except for replacing the coffee extract with distilled water. Absorbance was measured in a 96-well microplate reader at 517 nm. The DPPH radical scavenging activity was calculated with the following equation:

$$\%\text{DPPH Inhibition} = \frac{A_K - A_A}{A_K} \times 100, \quad (1)$$

where A_K is the absorbance of the control and A_A is the absorbance of the sample at 517 nm. All samples were performed in triplicate.

2.4.2 | $ABTS^+$ radical scavenging activity

The $ABTS^+$ radical scavenging activity was performed according to the procedure reported by Ballesteros et al. (2017) with modifications. The $ABTS^+$ stock solution was prepared by mixing equal amounts of 7 mM $ABTS^+$ and 2.4 mM potassium persulfate solution and allowed to react in the dark for 14 h at room temperature. The working solution was then diluted with 20% ethanol to obtain an absorbance of 0.70 ± 0.02 at 734 nm using a UV-spectrophotometer.

Reactions of the sample with the working solution were performed as described for DPPH (Section 2.4.1). A 96-well microplate reader spectrophotometer was used to measure the absorbance at 734 nm. Calculations were also performed as described for DPPH.

2.4.3 | Freeze-drying of SCGE

The freeze-dryer (Ningbo Scientz Biotechnology Co., Ltd) was precooled to −40°C before use. The prefrozen samples in petri dishes were wrapped with thin punctured plastic films and placed in the freeze-dryer. It was then covered with an organic glass lid and the vacuum pump turned on. The freeze-drying process was carried out for about 72 h at a pressure of less than 20 Pa and a temperature of −40°C.

2.5 | Preparation of bilayer films

Pure PLA substrate film was produced using the melt blending technique with a single screw plastic extruder (model LSJ-20) according to Li et al. (2020). KGM of 0.7% (w/v) was continuously stirred in hot deionized water (45°C) for 2 h (Wu et al., 2019). The ratio of WG, absolute ethanol and deionized water used was 1:1:4 which was continuously stirred at 75°C for 20 min with an adjusted pH of 10 according to Chavoshizadeh et al. (2020) with modification. The film-forming solutions of KGM and WG were mixed and evenly stirred. Finally, 15% (w/v) each of the freeze-dried SCGE from UAE and MAE (efficient methods) and 5% glycerol with respect to the total mixture were added and homogenized. The control sample was prepared using the same method but without freeze-dried SCGE. The coating was achieved by

pouring about 58 mL of the film solution on to the substrate PLA film, on a coating bed and the thickness was controlled by a coating rod. Films were allowed to dry on the coating bed at 45°C. The resulting biocomposite films were labeled as PLA/KGM/WG, PLA/KGM/WG/UE, and PLA/KGM/WG/ME, representing the control and films incorporated with ultrasound extracts (UE) and microwave extracts (ME) respectively.

2.6 | Film characterization

2.6.1 | Thickness

An electronic micrometer (Qinghai Measuring & Cutting Tools Co., Ltd) with 0.001 mm accuracy was used to measure the thickness of the films. Measurements were taken at five different spots on each film, and the average value was computed to represent the response for each replication.

2.6.2 | Mechanical properties of films

The TS and EAB of the produced films were measured using a universal testing machine (PARAM XLW [EC], Labthink) (ASTM- D882-12., 2012). Each rectangular film strip (10 × 80 mm) was mounted between the grips of the machine and tested, with an initial grip separation of 5 cm and crosshead speed of 50 mm/min at 25°C and 90% RH.

Five replicates were measured for each film.

2.6.3 | Barrier properties of films

The oxygen transmission rate (OTR) of the films were determined by coulometric method (sensor in cell). The OTR measurements were performed in OX-TRAN model 1/50 (Mocon) at 23°C and 0% RH (relative humidity) according to (ASTM D3985-17 (2017). Triplicate measurements of three different randomly selected films were done for each treatment.

2.6.4 | Water contact angle (WCA)

The WCA of films was tested by carefully dropping distilled water (3 μL) on the film's surface using a microsyringe. The contact angles and images were recorded immediately using a JC2000c video-based contact angle meter (Shanghai Zhongchen Digital Technic Apparatus co., Ltd). Triplicate measurements of three different randomly selected films were done for each treatment.

2.6.5 | Scanning electron microscopy (SEM)

The surface and cross-sectional morphologies of the films were observed by SEM (SU5000; Hitachi). Before the observation, randomly chosen samples were freeze-fractured in liquid nitrogen and sputter-coated with gold under vacuum. Finally, the observation was performed at an acceleration voltage of 15 kV with a magnification of ×460.

2.6.6 | Fourier-transform infrared (FTIR) spectroscopy

The film's chemical composition was analyzed using the Attenuated Total Reflection-Fourier Transform Infrared (ATR-FTIR) spectroscopy (Thermo Nicolet Corporation). Each film was randomly scanned from 4000 to 500 cm^{-1} for 64 times with an interval of 4 cm^{-1}.

2.6.7 | Quantification of TPC and antioxidant efficacy of films

Films of 6 mg were cut into smaller pieces and immersed in 95% ethanol at 37°C for 4 h. The amount of antioxidants were measured by reacting 2 mL of each film extract with 2 mL of $ABTS^{+}$ and DPPH solutions in the dark for 30 min. The absorbances were measured at 734 and 517 nm respectively, using a microplate reader.

2.7 | Preparation and packaging of cake

Cake batter was made with 250 g each of cake flour, butter, sugar, and eggs. All ingredients were mixed and baked in cupcake molds at 150°C for 25 min using a laboratory kitchen oven (JUSTA Oven Co., Ltd). The cakes had an average weight of 11.5 ± 4.3 g and were randomly packed into the three types of packaging films produced.

Each film was sealed into packaging bags and 12 cake pieces were randomly packed inside and completely sealed. The exposed group was packed on a laboratory plate without any packaging film. All samples were stored at 25°C for 21 days.

2.8 | Evaluation of fat oxidation

A roughly grounded cake in the ratio of 1 g of cake/2 mL hexane was agitated for 1 h and filtered to obtain the cake's lipid. The supernatant was rotary evaporated at 50°C to remove the extractant. The extent of lipid oxidation was evaluated by the Chinese National Standards for PV (GB 5009.227-2016), TBA (GB/T 35252-2017), and AV (GB 5009.229-2016). Analyses were performed in triplicates on days 0, 7, 14, and 21.

2.9 | Statistical analysis

Measurements were repeated in triplicates, unless stated otherwise under each experiment. Statistical analyses were performed by One-way ANOVA analysis with post hoc pairwise analysis and *t* test using SPSS (version 16 software Inc). The results were considered statistically

significant when $p \leq 0.05$. All data were presented as mean ± standard deviation.

3 | RESULTS AND DISCUSSION

3.1 | Effects of extraction methods on the TPC and amount of antioxidants

The maximum extraction conditions for CE, UAE, and MAE, were 60 min, 240 W/60 min and 900 W/5 min, respectively. The highest TPC from the three methods were 18.24 ± 0.8, 43.66 ± 0.4, and 62.63 ± 0.5 mg GAE/g on average for CE, UAE, and MAE respectively, as shown in Figure 1a. The highest DPPH and $ABTS^+$ radical scavenging activities are shown in Figure 1b. The MAE conditions produced significantly ($p \leq 0.05$) higher values for TPC, DPPH, and $ABTS^+$ scavenging activities than the UAE and CE methods. Other studies reported varying results for TPC and antioxidant activities for SCGE (Ballesteros et al., 2017; Mussatto et al., 2011). This could be due to the various factors that affect TPC and amount of antioxidants such as coffee species, roasting degree, storage conditions, the extraction method, the composition and amount of the extraction solvent, processing/extraction time, and other extraction conditions (Lourenço et al., 2019).

The TPC is strongly correlated with antioxidant activity as the structural features of phenolic compounds are responsible for antioxidants (da Rosa et al., 2019). In MAE, electromagnetic waves interact with the solvent molecules, generating rapid heat and pressure that is transmitted homogeneously to the sample ionically or via dipole rotation. The solvent penetration within the sample is thus increased through cell wall rupture, causing a faster release of cellular components into the solvent, reducing extraction time, improving yield, and being suitable for thermolabile components (Bachtler & Bart, 2021; da Rosa et al., 2019). In the same vein, the proliferation of ultrasonic waves results in cavitation, creating chemical and mechanical effects that intensify mass transfer and close interaction between the solvent and plant tissues, producing high yields (Kumar et al., 2021). However, the efficiency of the UAE remained inferior to that of the MAE, which can be explained by the lower rate of cell structural rupture due to the lower frequency levels of ultrasonics as compared to microwaves (Chemat et al., 2017). da Rosa et al. (2019) compared scanning electron microscopic pictures of MAE, UAE and CE of olive leaf extracts after extraction with an unextracted leaf sample. The images that underwent extraction showed damaged cell structures compared to unextracted ones. The authors reported that MAE effectively raptured the cell structure, leading to a high yield of TPC and antioxidant activity. The images of the UAE showed minimal disruption compared to MAE. In contrast, CE images presented tiny pores on the surface, resulting in the lowest TPC and antioxidant activities. This observation agrees with an earlier work (Gavahian et al., 2015; Hiranvarachat et al., 2013).

In this study, the TPC and antioxidant activities of the MAE was higher than the UAE and CE, which is similar to other results (Bachtler & Bart, 2021; da Rosa et al., 2019; Tabaraki & Ghadiri, 2016). The CE recovered a very small amount of extracts compared to the green extraction methods. Therefore, nonconventional extraction techniques constitute more promising technologies for recovering antioxidants than CE methods.

3.2 | Film characterization

3.2.1 | Thickness

The thickness of films is essential in evaluating the mechanical and barrier properties of packaging materials. The average thicknesses of the coated films with or without SCGE ranged from 0.11 ± 1.61 to 0.15 ± 2.30 mm, as shown in Table 1. The thickness of the films incorporated with 15% SCGE was significantly higher ($p \leq 0.05$) than that without the extracts. This observation is mainly due to the increase in the solid content of films with the addition of SCGE (Getachew et al., 2021). This result is consistent with

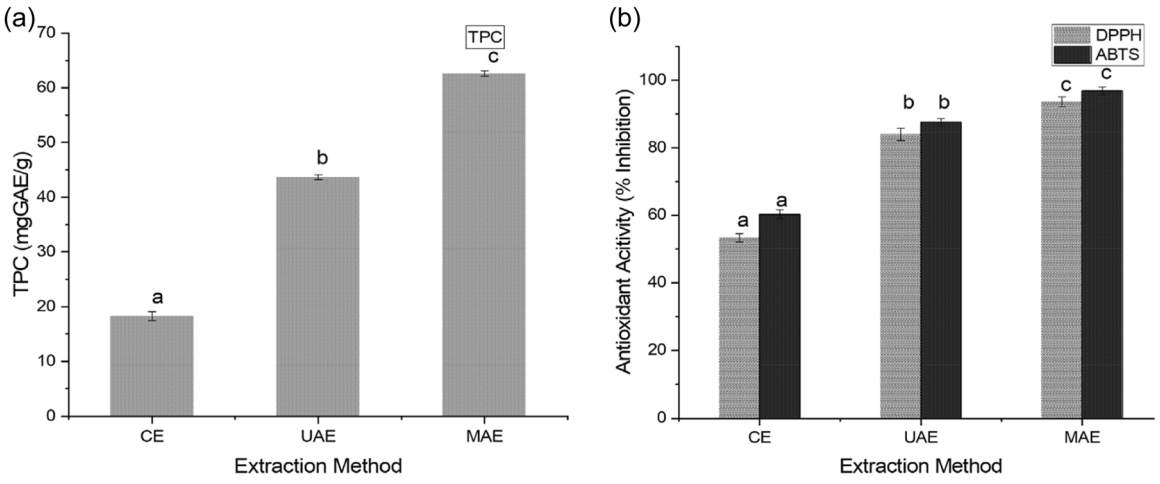

FIGURE 1 (a) (TPC) and (b) (DPPH and $ABTS^+$) radical scavenging activities of the extracts from different methods. Bars with different letters show significant differences ($p \leq 0.05$) between the groups. TPC, total phenolic content.

TABLE 1 Thickness, mechanical and barrier properties and, WCA of films.

Film type	Thickness (mm)	TS (Mpa)	EAB (%)	OTR (cm^3/m^2 24 h 0.1 Mpa)	WCA (°)
PLA/KGM/WG	0.11 ± 0.01^a	40.73 ± 1.45^b	5.44 ± 0.73^a	5.05 ± 0.04^a	70.19 ± 0.96^a
PLA/KGM/WG/UE	0.15 ± 0.01^b	34.07 ± 0.87^a	5.64 ± 0.87^a	2.26 ± 0.38^b	47.49 ± 1.19^b
PLA/KGM/WG/ME	0.15 ± 0.00^b	33.95 ± 0.63^a	5.82 ± 0.87^a	2.22 ± 0.13^b	44.15 ± 1.48^b

Note: Data is presented as mean ± standard deviation. Values with different letters in the same column show significant differences ($p \leq 0.05$).
Abbreviations: EAB, elongation at break; OTR, oxygen transmission rate; PLA, poly (lactic) acid; TS, tensile strength; WCA, water contact angle; WG, wheat gluten.

(Getachew et al., 2021), where the incorporation of SCGE up to 15% and 20% into gelatin films significantly increased the thickness of the films.

3.2.2 | Mechanical properties of films

The mechanical (TS and EAB) and barrier parameters (OTR) of biodegradable films show their ability to withstand stress and oxygen vapor transmission respectively, during different stages of handling, transportation and storage (Mir et al., 2018). In this study, the TS of the SCGE-coated films showed a notable decrease ($p \leq 0.05$) without significantly affecting the EAB. It is worth noting that the SCGE-coated films displayed slightly higher but not significant ($p > 0.05$) EAB values than the films without SCGE. Furthermore, the EAB of PLA/KGM/WG/ME was relatively higher ($p > 0.05$) than that of the PLA/KGM/WG/UE. This could be due to the presence of oil and phenolic compounds in the SCGE acting as plasticizers and crosslinking agents respectively, slightly increasing the flexibility of the films. Similar trends were observed as the SCG concentration increased to 20% in pectin films (Mendes et al., 2019) and 10% wt. in PLA (Suaduang et al., 2019).

3.2.3 | Barrier properties of films

Table 1 depicts the results of the OTR of the developed films. Compared to PLA/KGM/WG films, the addition of SCGE significantly ($p \leq 0.05$) decreased the OTR of the packaging films. The PLA/KGM/WG/ME displayed a lower but not significant ($p > 0.05$) OTR value than that of PLA/KGM/WG/UE film. This observation could be attributed to the amount of extracted solids added to the inner coated layer with effective antioxidant scavenging compounds, limiting the mass transfer of oxygen through the films (Mir et al., 2018).

3.2.4 | Water contact angle of films (WCA)

The WCA of packaging films estimates whether films are hydrophilic or hydrophobic. WCA values greater than 90° on the surface are considered hydrophobic, while values less than 90° are hydrophilic (Wu et al., 2018). The WCA values of all packaging films produced in this study are considered hydrophilic, as shown in Table 1. This could be attributed to the hydrophilicity of all the constituents of the inner layer. Furthermore, the incorporation of SCGE into the polymeric matrix notably reduced the WCA of the films ($p \leq 0.05$). Yet again, there were no significant differences between the PLA/KGM/WG/UE and PLA/KGM/WG/ME films, although there was a slight increase in the PLA/KGM/WG/UE film. Variations in the WCA values of the films is probably induced by the introduction of polyphenols contained in the extracts into the polymeric matrix. Polyphenols possess many polar groups including—OH and =O, magnifying the wettability of films' surfaces (Mir et al., 2018). The small variation between the groups with the SCGE may be explained by the fact that the ME contained more polyphenols than UE, but not enough to cause significant changes in the developed packaging materials. Identical results were reported (Moraczewski et al., 2020) with the incorporation of PLA with 10% coffee extracts via extrusion. Another study reported contradictory results when 20% of SCGE was incorporated with fish skin gelatin via solution casting (Getachew et al., 2021).

The contact angle increased from 77.79° to 104.20° with SCGE addition from 0% to 20%. The researchers explained that the increment was due to increased crosslinking between the phenolic compounds and the polar groups of the gelatine molecules, turning the polar groups toward the inner structure of the film, leading to a more hydrophobic film surface. These inconsistencies highlight the effects of extract concentration, polymeric matrix, and method of film preparation on the water uptake of films.

3.2.5 | Scanning electron microscopy (SEM)

The films' cross-section (a–c) and surface (d–f) micromorphologies are shown in Figure 2a. The SEM micrographs of the PLA/KGM/WG and SCGE-coated films clearly show different structures. The substrate film (PLA) presented a smooth and homogeneous surface, as seen in the cross-sectional views (a–c) of all films.

The bilayer film of PLA/KGM/WG displayed partial immiscible structures with small aggregates, whereas films loaded with SCGE imparted larger aggregates to the film's microstructure. This phenomenon could be due to the polarity difference between the hydrophobic PLA substrate film and the hydrophilic inner KGM/WG blend (Rocca-Smith et al., 2019), with further agglomeration of the SCGE in the polymer matrix (Getachew et al., 2021). The larger voids observed in the SCGE incorporated films may explain the decrease in TS of the films and increased water uptake of the films as observed in the WCA analysis. The SEM observations of the bilayer films agree with the SEM micromorphologies of pectin

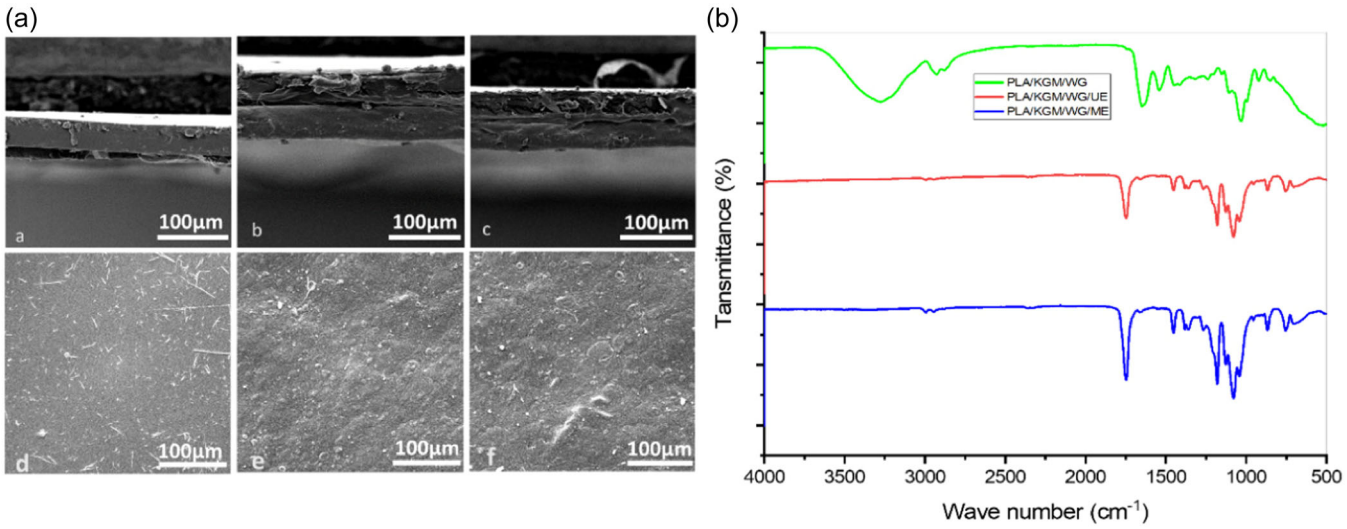

FIGURE 2 (a) SEM images of bilayer films; cross-section of PLA/KGM/WG (a), PLA/KGM/WG/UE (b), and PLA/KGM/WG/ME (c) and surface views of PLA/KGM/WG (d), PLA/KGM/WG/UE (e), and PLA/KGM/WG/ME (f). (b) FTIR spectroscopy of films. FTIR, fourier-transform infrared; PLA, poly (lactic) acid; WG, wheat gluten.

(PEC)/KGM films loaded with tea polyphenols (TP) (Lei et al., 2019). Using pure PEC/KGM films and a 2% addition of TP revealed high compatibility and stacked compact layers. In contrast, the addition of TP up to 5% created discontinuous zones with visible cavities along with the polymer network.

3.2.6 | FTIR spectroscopy

The FTIR results are shown in Figure 2b. Generally, all films presented similar patterns of peaks with different intensities, signifying no covalent bonding in the molecular structure of the bilayer films. The representative peaks observed for PLA at 2874 and 2935 cm^{-1} correspond to symmetric and asymmetric —CH_3 deformation vibrations, C—O stretching at 1082 cm^{-1}, and —C—O—C stretching of ester groups at 1005 cm^{-1} (Muller et al., 2017). In the absorption range of 700–1745 cm^{-1} in all films, similar peaks indicative of amides I, II, and III were observed (Getachew et al., 2021). Amide I shows C═O stretching and hydrogen bonding coupled with C—N stretching corresponding to the strong peak at 1659 cm^{-1}, amide II corresponds to C—N stretching and N—H bending at 1546 cm^{-1}, and amide III indicates the characteristic N—H and C—H groups in-plane vibration along with the vibration of the CH_2 group of proline side chains and glycine backbone at 1234 cm^{-1} (Arfat et al., 2017a). The peaks at 889 and 1566 cm^{-1} are assigned to the mannose unit stretching vibration and the benzene ring's unsaturated vibration peak respectively.

The broad peak at 3200 cm^{-1} observed in the films without SCGE corresponds to the stretching vibration of hydroxyl (—OH) and amino (—NH) groups (Arfat et al., 2017). The intensities and spectral patterns of the absorption bands slightly shifted or disappeared with the incorporation of the bilayer films with SCGE. The disappearance of the peak at ~3200 cm^{-1} after the addition of SCGE may have resulted in internal bond changes in some functional groups (Agarwal et al., 2021). This could also be due to the decreased stretching of free OH and/or NH as a result of the binding interactions between polyphenols and the inner layer KGM and WG as seen in the decreased OTR of the packaging films. Sun et al. (2017) reported that a broad peak at 3259 cm^{-1} became more flattened with the increasing concentration of young apple polyphenols in chitosan films at concentrations of 0.25%, 0.50%, 0.75%, and 1% than the peak in pure chitosan films.

It is worth noting that, the concentrations of polyphenols incorporated in that study is much less compared to the concentration in our study (15%), and if the concentrations were increased further, they could have noticed the disappearance of the broad peak around 3259 cm^{-1}. For the SCGE films, the intensity of the bands between 1000 and 1300 cm^{-1} was found to increase, which is related to C—O and C—C bands. Furthermore, the intensity of the stretching vibration of C═O and bending vibration of N—H in the amino group were amplified for peaks between 1400 and 1800 cm^{-1} in the SCGE-coated films, indicating the occurrence of hydrogen bonds between C═O and N—H with the O—H bands in polyphenol compounds (Li et al., 2014).

3.2.7 | TPC and antioxidant activity of films

The results of TPC, DPPH, and $ABTS^+$ are presented in Figure 3a,b, respectively. As expected, films with SCGE showed notably higher TPC, DPPH, and $ABTS^+$ values than films without extracts ($p \leq 0.05$). The higher TPC and antioxidant activity observed in the films is due to the presence of SCGE in the polymeric matrix, which imparts significant antioxidant activity to the films. The films without extracts showed minimal activities for TPC (0.4 ± 0.01 mg GAE/g), DPPH (7.53%), and $ABTS^+$ (8.40%). The antioxidant activity of the SCGE films was 9.5 times more than the films without extracts. Additionally, the PLA/KGM/WG/ME still maintained significantly higher TPC, DPPH

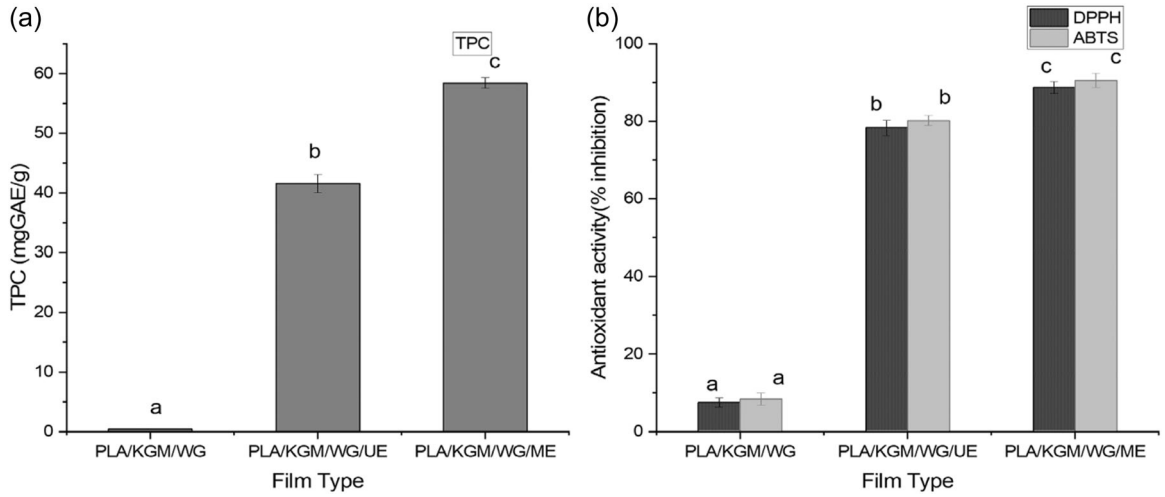

FIGURE 3 (a) TPC and (b) antioxidant activity of films. Bars with different letters show significant differences ($p \leq 0.05$) between the groups. TPC, total phenolic content.

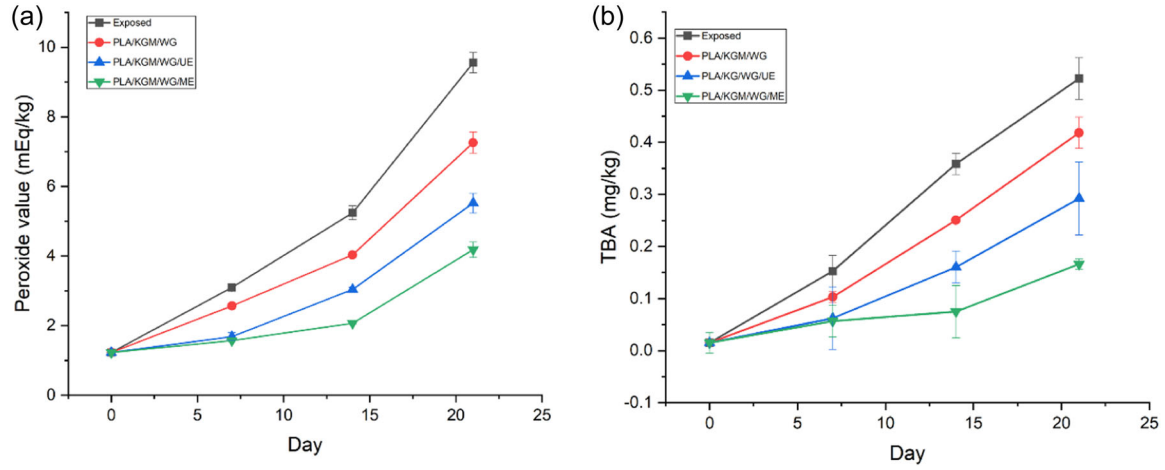

FIGURE 4 (a) PV and (b) TBA values of cake products during 21 days of storage. TBA, thiobarbituric acid.

and ABTS$^+$ activities than those of PLA/KG/WG/UE ($p \leq 0.05$). Similarly, the in vitro antioxidant capacity of a coffee-cocoa-cassava starch active film was reported (Getachew et al., 2021; Veiga-Santos et al., 2018). The developed active film showed a protective effect up to 6.09, 60.4, and 6.88 times against peroxide index increase, hexanal production, and conjugated dienoic acid production respectively, likened to a commercial polymer.

3.3 | Evaluation of fat oxidation

3.3.1 | Peroxide value (PV) and TBA of cake during storage

PV is a very important indicator used in food quality control. It is a commonly used method to determine the primary oxidation of lipids in food products. Hydroperoxides, the main byproducts of lipid peroxidation are highly labile and can undergo degradation, generating diverse secondary products like aldehydes, ketones, alcohols, and esters responsible for the deterioration of the sensory properties of foods rich in fat (Kusnandar et al., 2020). The PV in the range of 10–20 mEq/kg is considered rancid but still acceptable, and unacceptable for consumption above 20 mEq/kg (Izzreen & Noriham, 2011). The changes in the PV of the cake samples during the storage period are presented in Figure 4a. All cake samples showed a gradual increasing trend throughout the storage period except the samples exposed to open air, which rapidly increased after Day 14. All treated samples in this study are considered nonrancid and are still acceptable for consumption throughout the storage period. In comparison to the films without extracts, the packaging films with SCGE resulted in an evident decrease in PV, with the exposed group presenting the highest values. There were no observed significant differences among the groups with SCGE on Day 7, but all groups were significantly different at the end of storage time, evidence of the higher antioxidant ability of ME than UE ($p \leq 0.05$). Similarly, the integration of TP into cake preserved at 63°C during 28 days of storage observed the same trends for the first 14 days.

However, very sharp increments were observed for the rest of the storage time, where the PV of samples

enriched with green tea extract exceeded the control samples at the end of storage. This observation may be related to the fact that the green tea extracts might have acted as prooxidants (Kozłowska et al., 2019). Thus, it highlights the sustained activity of the SCGE bilayer films in retarding lipid oxidation, thereby contributing to the oxidative stability of cakes throughout the storage time.

The efficacy of controlled release of various active compounds from active packaging over direct incorporation of active ingredients into food is summarized in a critical review (Chen et al., 2019).

Secondary oxidation products such as malonaldehydes, formed during the breakdown of lipids contributes to the off-flavors of oxidized oil and are commonly quantified with the TBA index (Kusnandar et al., 2020). The higher the TBA value, the greater the degree of lipid degradation, and oxidation in the food product (Ozogul et al., 2005). As shown in Figure 4b, the secondary oxidation of cake lipids followed the trend of the primary oxidation products. During the 21 days of storage, the control samples displayed the highest TBA values, whereas the SCGE films showed the lowest values. An earlier research report suggests that a TBA value of less than 0.567 mg/kg is considered nonrancid, 0.65−1.44 mg/kg as rancid but still acceptable, while above 1.5 mg/kg as rancid and unsafe for consumption (Izzreen & Noriham, 2011). All samples were still acceptable at the end of storage. Correspondingly, the effect of SCG as a natural antioxidant on soybean oil and fish oil with a high concentration of omega-3 fatty acids has been demonstrated (Hwang et al., 2019). The SCGE at 0.25% and 0.5% concentrations were equal to or more effective than synthetic BHT in retarding oxidative deterioration of stored oils at 30°C and 50°C for 14 days. Other studies have demonstrated the feasible antioxidant activity of SCG in palm oil (Veiga-Santos et al., 2018), salted mackerel (Song et al., 2009) and frozen cooked pork patties (Jully et al., 2016).

3.3.2 | Acid value (AV)

The AV is an essential indicator of fat quality, characterizing the hydrolytic fat deterioration. It is the needed amount of potassium hydroxide (KOH) in mg to neutralize the organic acids present in 1 g of oil, and it is a measure of free fatty acids in the oil (Kusnandar et al., 2020).

The AV for all samples increased throughout the storage period, with the highest values recorded in the exposed group, followed by the films without SCGE and the SCGE incorporated bilayer films (Figure 5). There were no statistical differences between SCGE incorporated films during the storage time ($p > 0.05$). Budryn and Nebesny (2013) reported opposing results when freeze-dried coffee extracts were directly incorporated into cookies and chocolates with high-fat content. The AV increased with the increase in the concentration of coffee extracts as compared to the control samples. The authors stated the high content of organic acids in coffee composites such as triacylglycerols could be more prone to hydrolysis. This phenomenon emphasizes the advantages of packaging films in food preservation over direct addition to the food product. The PV, TBA, and AV evaluations of the cake products under different treatments indicate the potential of SCGE-coated bilayer film to maintain food products with high-fat content.

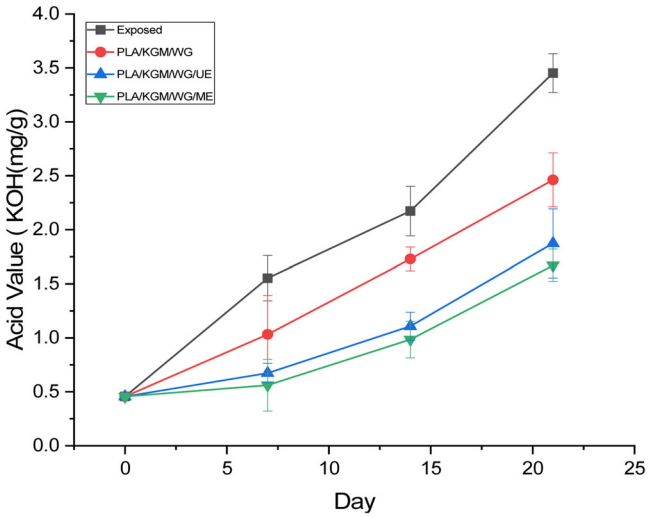

FIGURE 5 Acid value of cake samples during 21 days of storage.

4 | CONCLUSION

The study investigated the effects of different extraction methods and conditions on the recovery of phenolic compounds and antioxidants from SCG, and its application in the development of a packaging material for the shelf-life extension of cake. The MAE and UAE methods produced extracts with high TPC and antioxidants as compared with the well-established conventional heating method, highlighting the effectiveness of the emerging green extraction methods. The extracts produced were freeze dried and incorporated into bilayer films made of PLA, KGM, and WG via extrusion and coating methods.

The 15% SCGE integration offered a significant antioxidant activity to the films compared to the control films. Additionally, the OTR was enhanced for SCGE-containing films. Generally, cake samples packaged with SCGE-containing films considerably delayed lipid oxidation in terms of PV, TBA, and AV compared to the control film and, exposed group. Moreover, packaging with PLA/KGM/WG/ME film was more effective in preserving cake lipids than with PLA/KGM/WG/UE, highlighting MAE as a more suitable method to recover bioactive constituents from plant sources. Unlike some traditional methods such as the direct addition of the SCGE into the food which limits its long-term preservation effect, the current study proves that SCGE loaded with PLA/KGM/WG bilayer film is an improved method for long-term preservation purposes. Overall, the study demonstrates SCGE as a valuable source of antioxidant material capable of delaying lipid oxidation in food.

However, the remarkable contribution of the hydrophilic lignocellulose component of the SCGE in addition to the hydrophilic components of the inner KGM and WG layer coating increased the water uptake of the films.

Hence, it is recommended that future studies need to focus on reducing the hydrophilicity of SCGE before incorporation into polymeric matrices along with a good blend of polymers.

AUTHOR CONTRIBUTIONS

Salimata Yakubu: Conceptualization; data curation; formal analysis; investigation; methodology; software; writing—original draft; writing—review & editing. **Hui Zheng**: Investigation; methodology; software. **Jingwen Chen**: Investigation; methodology. **Francis Kweku Amagloh**: Conceptualization; investigation; writing—review & editing. **Jingge Xu**: Investigation; methodology. **Xiaohan Chen**: Investigation. **Li Wang**: Conceptualization; investigation; methodology; resources; supervision; validation; writing—review & editing. **Li Li**: Conceptualization; funding acquisition; methodology; project administration; resources; supervision; validation.

ACKNOWLEDGMENTS

The authors would like to thank Mr. Dominic Yellezuome and Miss Walker Anita Nyarkoa for proofreading the article, and Mr. Matthew Atongbiik Achaglinkame and Sabina Ackah for making valuable inputs in the methodology. The research was kindly supported by the Program of Special Capability Development for local Colleges and Universities in Shanghai (19050502000).

CONFLICT OF INTEREST STATEMENT

The authors declare no conflicts of interest.

DATA AVAILABILITY STATEMENT

Data available on request from the authors.

ETHICS STATEMENT

None declared.

ORCID

Salimata Yakubu http://orcid.org/0000-0002-2026-5263

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How to cite this article: Yakubu, S., Zheng, H., Chen, J., Amagloh, F. K., Xu, J., Chen, X., Wang, L., & Li, L. (2023). Development of a bilayer biodegradable packaging material enriched with coffee waste extract for cake preservation. *Food Bioengineering*, *2*, 212–222. https://doi.org/10.1002/fbe2.12058