Foods and Raw Materials. 2026;14(2)

ISSN 2308-4057 (Print)
ISSN 2310-9599 (Online)

Research Article
Open Access

Available online at http://jfrm.ru/en
https://doi.org/10.21603/2308-4057-2026-2-681
https://elibrary.ru/ETGLCB

Developing composite films from carboxymethyl starch, polyvinyl alcohol, and kaolin for sustainable packaging applications

Bright A. Omoike[1,*], Felix E. Okieimen[2], Chinyere Imoisi[1]

[1] Mewar International University, Masaka, Nigeria
[2] University of Benin, Benin City, Nigeria

* e-mail: brightomoike@yahoo.com

Received 27.09.2024; Revised 21.11.2024; Accepted 03.06.2025; Published online 20.10.2025

Abstract:
Health and environmental problems are rising by the day due to an increasing use of synthetic plastics. However, biobased packaging from starch, with its numerous advantages, or its derivatives offers a promising solution to this problem. In this study, we aimed to explore a sustainable approach to developing a bioplastic film from carboxymethyl starch, polyvinyl alcohol, and kaolin to serve as a substitute for synthetic packaging.

The study objects included carboxymethyl starch, polyvinyl alcohol, glycerol, and kaolin. All the materials were heated in water to form viscous solutions. The solution was then cast into films using a mold and the water was evaporated through oven-drying. The cast films were characterized via scanning electron microscopy, X-ray diffraction, and thermogravimetric analysis. They were analyzed for their tensile mechanical, barrier, sorption, and biodegradability properties. We also investigated the effects of polyvinyl alcohol and kaolin on the morphology and functional properties of the films.

The micro-surface morphology of the carboxymethyl starch/polyvinyl alcohol blend revealed a smooth and homogenous structure, while the film reinforced with kaolin had a more compact structure with zones of particle aggregations. The highest thermal stability was observed in the composite films containing carboxymethyl starch, polyvinyl alcohol, and kaolin. Higher contents of polyvinyl alcohol and kaolin significantly improved the films' thermal, tensile mechanical, barrier, and sorption properties. The films also demonstrated a substantial rate of biodegradability. The best properties were observed in the films containing 40% of carboxymethyl starch, 60% of polyvinyl alcohol, and 4.5 per hundred resin (phr) of kaolin.

The composite films made from carboxymethyl starch, polyvinyl alcohol, and kaolin had good biodegradability, renewability, and improved functional material properties. Therefore, they can be considered a sustainable alternative to the traditional synthetic plastics in packaging applications.

Keywords: Biobased packaging, bioplastics, sustainable, mechanical tensile, barrier properties, biodegradability, composites

Please cite this article in press as: Omoike BA, Okieimen FE, Imoisi C. Developing composite films from carboxymethyl starch, polyvinyl alcohol, and kaolin for sustainable packaging applications. Foods and Raw Materials. 2026;14(2):344–356. https://doi.org/10.21603/2308-4057-2026-2-681

INTRODUCTION

In the face of escalating global concerns about environmental degradation and waste management, sustainable development has emerged as a paramount priority in the pursuit of a circular economy and waste minimization strategies [1]. This imperative has inspired researchers to seek innovative and greener solutions to conventional plastics in the packaging industry [2]. The plastics industry is a significant sector globally, boasting a massive market of $348 billion and playing a crucial role in shaping the global economy [3]. The demand for plastics is expected to continue rising, with a projected annual growth of 4.2% from 2021 to 2026 [4].

However, the environmental impact of conventional plastics has become a major concern, driving the need for sustainable alternatives. Moreover, the natural sources of these conventional plastics are not renewable and hence are becoming depleted as a result of the increasing demand for plastics [2]. Bioplastic is a novel, eco-friendly, and sustainable substitute for the traditional petroleum-based plastic [5]. It is obtained from renewable bio-natural resources such as starch, cellulose, gelatin, chitin, etc. [6]. The use of bioplastic offers advantages of biodegradability, biocompatibility, and renewability in addition to nearly comparable material strength properties of conventional plastics.

Starch-based bioplastics are promising alternatives to conventional plastics but further modifications are needed to enhance their mechanical and barrier properties for competitive use [7]. Previous studies have shown various

Copyright © 2025, Omoike *et al.* This is an open access article distributed under the terms of the Creative Commons Attribution 4.0 International License (https://creativecommons.org/licenses/by/4.0/), allowing third parties to copy and redistribute the material in any medium or format and to remix, transform, and build upon the material for any purpose, even commercially, provided the original work is properly cited and states its license.

techniques for modifying starch-based films. These include modifying native starch [8], blending with other biomaterials sources [9], or reinforcing with nanofillers [10].

Carboxymethyl starch (CMS) is a versatile green polymer with a wide range of applications in pharmacy, medicine, cosmetics, food, environmental protection, and other industries [11]. It is a chemically modified starch derived from native starch through a reaction with monochloroacetic acid. This process leads to the replacement of some hydroxyl groups with carboxymethyl groups and is known as the Williamson synthesis [12]. This modification enhances the properties of starch, making it a promising material for packaging applications. Introducing a bulky hydrophilic group into the starch chains results in improved thermal stability and film-forming properties, as well as in decreased susceptibility to microbial degradation [13, 14]. Additionally, CMS demonstrates a low tendency for retrogradation, a low gelatinization temperature, and a good freeze-thaw behavior, which further expands its potential applications [11, 15].

The blending of polyvinyl alcohol (PVA or PVOH) with starch has been reported as a good approach to improve the properties of starch-based films [16]. Polyvinyl alcohol is the most widely produced biodegradable synthetic polymer in the world because of its exceptional physical characteristics, resistance to chemicals, and full biodegradability [17]. It is a polymer with a linear structure that is non-toxic and non-carcinogenic. Its remarkable film-forming ability and good compatibility with other materials have led to a wide range of industrial applications [18], especially in food packaging. Due to their semicrystalline structure, PVOH films exhibit excellent oxygen and aromatic barrier qualities, a reduced capacity for water adsorption, high tensile strength, and flexibility [19]. Adding PVOH to biodegradable packaging enhances its mechanical characteristics and water resistance [16, 6]. Highly-polar hydroxyl groups (-OH) in starch and PVOH can create intra- and intermolecular hydrogen interactions, which can change the functional properties of blended films and enhance their integrity [20].

Kaolinite, also known as Kaolin clay, is a naturally occurring clay mineral with various applications in different industries, such as adhesives, paints, pharmaceuticals, fiberglass, paper, rubber, ceramics, electronics, plastics, coatings, and agriculture [21]. Its chemical composition consists of hydrated aluminum silicate with a structure of 1:1 dioctahedral asymmetric layers linked through apical oxygen [21]. Adding kaolin clay to starch films improves their mechanical and thermal stability. Kaolin clay also acts as an effective barrier to moisture transmission, which makes it suitable for use in packaging materials [22, 23]. This cost-effective and versatile material offers a wide surface area and reactive surface for OH-groups, making it an interesting alternative for incorporating into various products [4].

Despite extensive research on developing native starch-based composites for packaging applications, limited studies have explored the potential of carboxymethyl starch and Wilpiszewska *et al.* [25] have demonstrated the potential of CMS derivatives in pharmaceutical and food packaging applications. However, further research is urgently needed to improve the properties and viability of CMS for packaging uses.

In this study, we aimed to develop a CMS/PVA/kaolin composite film with enhanced properties for packaging applications through the synergistic effect of PVA and kaolin (Fig. 1). We also investigated the impact of PVA and kaolin on the morphological, thermal, mechanical, barrier, sorption, and biodegradability properties of CMS/PVA/kaolin composite films. A comprehensive review of the current literature revealed no previous research in this area, highlighting the need to explore the properties and potential applications of this composite material.

STUDY OBJECTS AND METHODS

Chemicals and materials. Carboxymethyl starch (degree of substitution = 0.71), polyvinyl alcohol, and glycerol of analytical grades were supplied by a chemical store in Benin City. Kaolin clay was obtained from a factory in Auchi, Edo State. All the other chemicals and reagents in this study were of analytical grade.

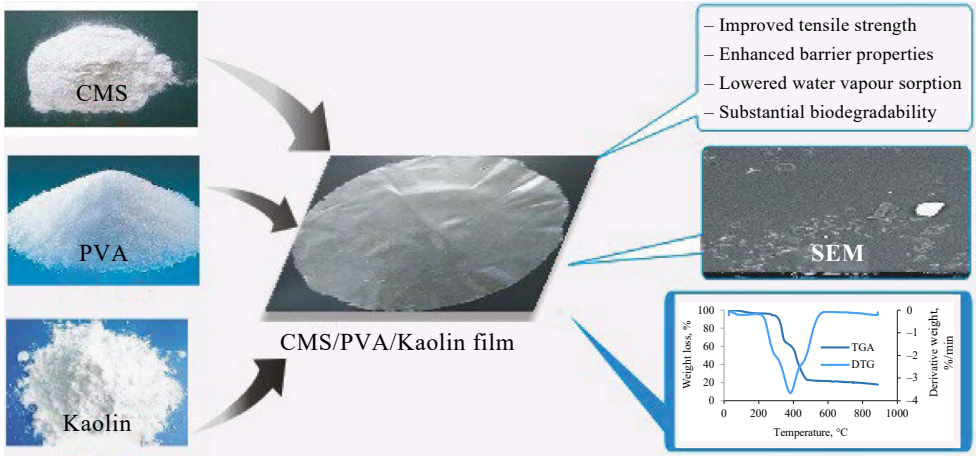


Figure 1 Graphical representation for the preparation and characterization of composite films

Preparation of a carboxymethyl starch/polyvinyl alcohol blend. Carboxymethyl starch (CMS)/polyvinyl alcohol (PVA) blend was prepared by the solution casting method as described by Patil *et al.* with slight modifications [16]. First, different masses of PVA were heated at 90°C in distilled water until complete dissolution. Then, we added a slurry of CMS to the PVA solution and heated at 80–90°C with continuous stirring to ensure complete gelatinization and homogenization. Glycerol was then added to the mixture and stirred for another 20 min. The viscous polymer solution was cast on a glass plate at room temperature for 24 h. The films were further dried in an oven at 60°C for 16 h before being peeled off the plates and kept in a desiccator for analysis. The composition of the films varied depending on the mass of PVA and CMS in the blend.

Preparation of carboxymethyl starch/polyvinyl alcohol/kaolin composites. Carboxymethyl starch (CMS)/polyvinyl alcohol (PVA) films reinforced with kaolin clay were prepared using a modified method described in the study by Garavand *et al.* [26]. First, kaolin clay was dispersed in distilled water and then placed in a mechanical shaker to ensure even distribution of the clay particles. Separately, a slurry of CMS was prepared in water. Meanwhile, PVA was heated at 90°C in distilled water until complete dissolution. Once PVA was fully dissolved, it was mixed with the kaolin clay/CMS mixture and then with glycerol. The entire mixture was then stirred using a magnetic stirrer at 80–90°C for 30 min to ensure complete gelatinization and homogenization. This step was crucial to achieve a uniform filmogenic solution. The filmogenic solution was then cast onto a glass plate and dried in an oven at 60°C for 16 h. The dried films were subsequently peeled off and stored in a desiccator for further analysis. To control the thickness of the films, a known volume of the film solution was measured and poured into the glass plate mold.

The composition of the films is presented in Table 1, which outlines the specific proportions of carboxymethyl starch, polyvinyl alcohol, kaolin clay, and glycerol used in the study.

Characterization of carboxymethyl starch-based composite films. The surface morphological properties of the carboxymethyl starch-based composite films were analyzed using a Phenom ProX scanning electron microscope (Phenom-World, Eindhoven, Netherlands) at an accelerating voltage of 10 kV. Prior to scanning, the samples were mounted on a brass stub and sputtered with gold to ensure electrical conductivity. The scanning electron microscopy (SEM) micrographs were taken at magnifications ranging from 1500 to 2000 times, with the microscope focused on the upper surface of the films. SEM allows for a detailed visualization of the surface morphology, topography, and structure of materials at a microscopic level, enabling researchers to observe the characteristics and arrangement of the components within the composite films.

X-ray diffraction. The crystallinity of the carboxymethyl starch-based composite films was determined using a Miniflex 600 X-ray diffractometer (Rigaku, Japan). This instrument utilizes copper (Kα) anode radiation, which has a wavelength of 1.5444 Å, to examine the crystal structure of the films. The emitted anode radiation produced at 40 kV and 50 mA was passed through a monochromator to produce a focused incident beam. This beam was directed onto the film sample through a one-degree (1°) divergence slit, ensuring a precise and controlled measurement. The diffraction intensity was measured over a range of 2θ = 2 to 70° at room temperature. This allowed for a comprehensive assessment of the crystal structure and order within the composite films. The level of crystallinity for each film was evaluated by visual inspection of the strongest peak in the X-ray diffraction pattern.

Table 1 Composition of the films used for preparation.

Films (Coded)	CMS, g	PVA, g	CMS, %	PVA, %	Kaolin, phr
Pure CMS	10.0	0.0	100	0	–
CMS/PVA1	8.0	2.0	80	20	–
CMS/PVA2	7.0	3.0	70	30	–
CMS/PVA3	6.0	4.0	60	40	–
CMS/PVA4	5.0	5.0	50	50	–
CMS/PVA5	4.0	6.0	40	60	–
CMS/PVA/K1	4.0	6.0	40	60	1.5
CMS/PVA/K2	4.0	6.0	40	60	2.5
CMS/PVA/K3	4.0	6.0	40	60	3.5
CMS/PVA/K4	4.0	6.0	40	60	4.5
CMS/PVA/K5	4.0	6.0	40	60	5.5

CMS is the carboxymethyl starch; PVA is the polyvinyl alcohol; Amount of glycerol used (1 mL = 10% of CMS & PVA) for all films, phr = per hundred resin of filmogenic solution

Thermal stability. The thermal stability of the films was determined via thermogravimetric analysis. This analysis was conducted using a TGA 4000 thermal analyzer (PerkinElmer, Netherland) in a nitrogen gas environment at 10°C/min in the temperature range of 30–950°C. The thermogravimetric and derivative thermogravimetric curves were obtained to assess the thermal stability of the composite films. These curves provide valuable information about the weight loss of the films over a range of temperatures, indicating their thermal degradation behavior.

Evaluation of properties of carboxymethyl starch-based composites. ***Tensile mechanical properties.*** The tensile mechanical properties of the composite films were determined using a universal testing machine (TecQuipment, India) following standard procedures as described in [7]. Prior to the test, the films were cut into rectangular 3 × 15 cm strips and conditioned in a desiccator (0% relative humidity) for 48 h. The films' thickness was measured by means of a digital caliper and was used to obtain the cross-sectional area of the films (the film's thickness multiplied by its width). Subsequently, the ultimate tensile strength, percentage of

elongation at break, and tensile modulus of the films were determined by subjecting them to a controlled tension of 1 kN load at a cross-head speed of 50 mm/min. Their responses to the applied force just before breaking were measured.

Barrier properties. The barrier properties of the carboxymethyl starch-based composite films were determined using ASTM E96-95 standard procedures as described in earlier publications with slight modifications [27]. The water vapor permeability (WVP) of the films was evaluated by a gravimetric approach. A permeation cup with specific dimensions (4 cm in diameter and 3.5 cm in height) was utilized for the measurement. The films were cut into circular discs and mounted on the top surface of the cup using a collar-type lid supported with a rubber washer. The cup contained activated silica gel to maintain 0% relative humidity inside. The cup was then placed in a desiccator containing a saturated solution of NaCl (75% relative humidity). The test cup with the film was periodically weighed at a 24 h (1 day) interval for 5 days under room temperature, and the amount of water vapor that permeated through the film was calculated from the weight gain. The water vapor transmission rate (WVTR) through the film was determined from the plot of water vapor permeated as a function of time (Eq. (1)). The WVP was then calculated by considering various factors such as the film's thickness, area, and the partial pressure difference across the film (Eq. (2)). This method provides a comprehensive understanding of the barrier properties of the composite films in terms of their ability to prevent the permeation of water vapor.

$$\mathrm{WVTR} = \frac{S}{A} \tag{1}$$

$$\mathrm{WVP} = \frac{\mathrm{WVTR}}{S^1\left(R_2 - R_1\right)} \times L \tag{2}$$

where S is the slope of a plot; A is the cross-sectional area of the films, L is the film's mean thickness, mm; S^1 is the saturation vapor pressure at test temperature, Pa; R_1 is the relative humidity in the test chamber expressed as a fraction, and R_2 is the relative humidity in the vapor sink expressed as a fraction.

Determination of water sorption characteristics. The method used for this study is in accordance with that described in previous publications [28]. The films were first cut into pieces of 3 × 3 cm and dried in an oven at 60°C for 4 h to obtain a constant weight. One side of each film was covered with aluminum foil to allow absorption only on one side and prevent moisture permeation from the other side. The moisture sorption was measured at 25°C (75% relative humidity) in a desiccator containing a saturated solution of NaCl. The weight of the films was measured every 6 h for 7 days to reach the equilibrium weight. A control was set up to account for any moisture adsorbed on the aluminum foil surface. The mass of the water vapor absorbed was expressed as a weight fraction and plotted against the square root of time. The sorption, diffusion, and permeability coefficients were calculated using the Fickian equations (Eq. 3). The sorption coefficient was determined using the simplified Henry's law, where the concentration of absorbed water vapor in the film is linearly dependent on the water vapor partial pressure, and was calculated as a proportionality constant (Eq. 4). The sorption, diffusion, and permeability coefficients were calculated to understand the sorption properties of the composite films.

$$\frac{m_t}{m_\infty} = 1 - \sum_{n=0}^{\infty} \frac{8}{(2n+1)^2 x n^2} \times e^{\frac{-D x (2n+1)^2 x n^2 x t}{l^2}} \tag{3}$$

For short exposure times, the simplified Fickian equation (Eq. 4) holds:

$$\frac{m_t}{m_\infty} = \frac{4}{h}\sqrt{\frac{Dt}{\pi}} \tag{4}$$

where $\frac{m_t}{m_\infty}$ is the mass fraction of absorbed water vapour (i.e m_t is the mass absorbed at time t and m_∞ is the mass of moisture absorbed at equilibrium), h is the thickness of the film, π = 3.142, D is the diffusion coefficient, t is the time. The application of these equations to polymer composite materials involves simplifications, and thus, D should be considered as an effective diffusion coefficient, D_{eff} [29] can be determined experimentally from the initial slope (s) of the sorption curve (Eq. (5)).

$$D_{eff} = \pi\left[\frac{s}{4}\right]^2 \tag{5}$$

The sorption or solubility coefficient was determined using a mathematical expression based on Henry's law which assumes a linear relationship between the concentration of absorbed water vapour, C_{water} (i. e. in the film and the partial pressure of water vapour ($C_{water} = m_\infty \rho$) as described in Eq. (6) [18, 31].

$$S_{eff} = \frac{C_{water}}{P_{water}} = \frac{m_\infty}{\Delta P}\rho \tag{6}$$

where S_{eff} is the sorption coefficient; ρ is the density of the film and ΔP is the saturation vapour pressure at 75% relative humidity.

The water vapour permeability coefficient (P_{eff}) of the films as determined by sorption kinetics can be calculated using the relationship:

$$P_{eff} = D_{eff} \times S_{eff} \tag{7}$$

Determination of biodegradation. The biodegradability of the composite films was evaluated using the soil burial degradation method, as described by Chaisuwan *et al.* [30] with slight modifications. This method involved burying the film samples in compost soil to assess their degradation over time. The composite films were cut into rectangular pieces of 3 × 5 cm. These pieces were then buried in garden buckets filled

with compost soil at a depth of 10 cm. The buckets were kept at room temperature, and the soil was maintained in a moist state through regular watering. This ensured a consistent environment for the degradation process. The degradation of the films was assessed by measuring their weight at regular intervals of 7 days over a total period of 8 weeks (56 days). After removal from the soil, the films were gently washed to remove adhered soil, and the weight of the dry residual film was determined. The degree of soil burial degradation (DSBD), %, was calculated from the percentage weight loss of each film sample over time (Eq. 8). This equation provided a numerical value representing the extent of degradation, allowing for a comprehensive evaluation of the films' biodegradability.

$$\text{DSBD} = \frac{W_i - W_n}{W_i} \times 100 \qquad (8)$$

where W_i is the initial weight of the films before burial and W_n is the residual weight of the films after soil burial degradation over 7 successive days.

Tools and equipment manufacturers. The instruments used in this study, such as the digital caliper, oven, analytical balance, desiccator, and other equipment, were obtained from well-known manufacturers, including Hanna Instruments (Woonsocket, Rhode Island, USA), Thermo Fisher Scientific (Waltham, Massachusetts, USA), and Mettler Toledo (Columbus, Ohio, USA), among others.

Statistical analysis. Statistical analysis was carried out using the BMDP 2R software (version 7.0) for stepwise multiple regressions. The data were presented as a mean of three replicate measurements. The findings were considered statistically significant at $p < 0.05$.

RESULTS AND DISCUSSION

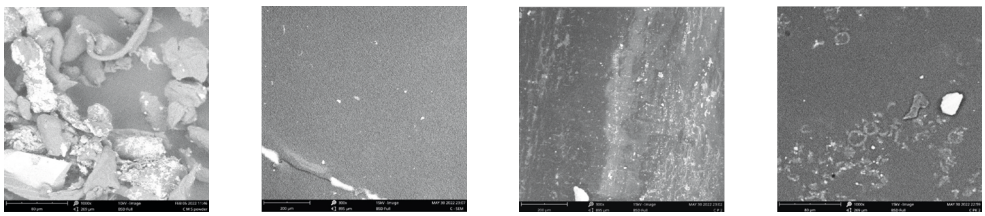
Scanning electron microscopy. Scanning electron microscopy (SEM) reveals important insights into the surface structural characteristics of composite materials in terms of the topography, distribution, and interaction of the components within the material [31]. The SEM image of carboxymethyl starch (CMS) powder (Fig. 2a) exhibited a porous granular structure, whereas the CMS film (Fig. 2b) displayed a smooth and continuous surface. This indicates effective gelatinization and plastification of CMS upon dispersion and heating in water during preparation [7].

The SEM image of the CMS/polyvinyl alcohol (PVA) blend (Fig. 2c) revealed a smooth and continuous surface structure, with particle domains having diameters in the micron range, possibly due to phase separation. However, the predominantly continuous phase indicated good miscibility and homogeneity between CMS and PVA. The indistinct interface between the two polymers suggested strong intermolecular interactions between CMS and PVA molecules, which were possibly facilitated by hydrogen bonding [16]. This observation indicates a high degree of compatibility and interaction between the two polymers [9].

The SEM images of the CMS/PVA/kaolin composite (Fig. 2d) exhibited a smooth surface homogenous morphology with no cracks or defects. However, there were some visible particle aggregates and zones of agglomeration due to the presence of kaolin in the blend. This distribution of kaolin aggregates in the homogenous matrix of CMS/PVA had a positive impact on the interfacial interaction between the kaolin particles and the polymer phase [4, 22].

X-ray diffraction (XRD) is a powerful technique used to assess the structural properties of materials by analyzing the diffraction intensity. It provides valuable information about the crystalline regions within the material [32]. In this study, XRD was employed to investigate the crystallinity and structural changes induced by PVA and kaolin additives in carboxymethyl starch (CMS)-based composite films. The results revealed significant changes in the crystalline peak patterns upon plasticization of CMS and blending with polyvinyl alcohol (PVA), indicating modifications of the material's crystalline structure. Specifically, the XRD diffractogram of the CMS powder (Fig. 3a) exhibited a characteristic peak intensity at 20.4°, indicating a high degree of crystallinity. In contrast, the CMS film (Fig. 3b) showed a decrease in peak intensity, suggesting disruption of the granular structure during heating and gelatinization [9].

When CMS was blended with PVA, an increase in peak intensity was observed in the XRD diffractogram of the film (Fig. 3c). This enhancement in peak intensity suggested strengthening of intermolecular bonding via additional hydroxyl groups on the PVA chains [9, 33]. The hydroxyl groups on the PVA chains can form hydrogen bonds with CMS, leading to improved crystallinity and structural stability.



a b c d

Figure 2 SEM micrographs of CMS powder (a), CMS film (b), CMS/PVA film (c), and CMS/PVA/kaolin film (d)

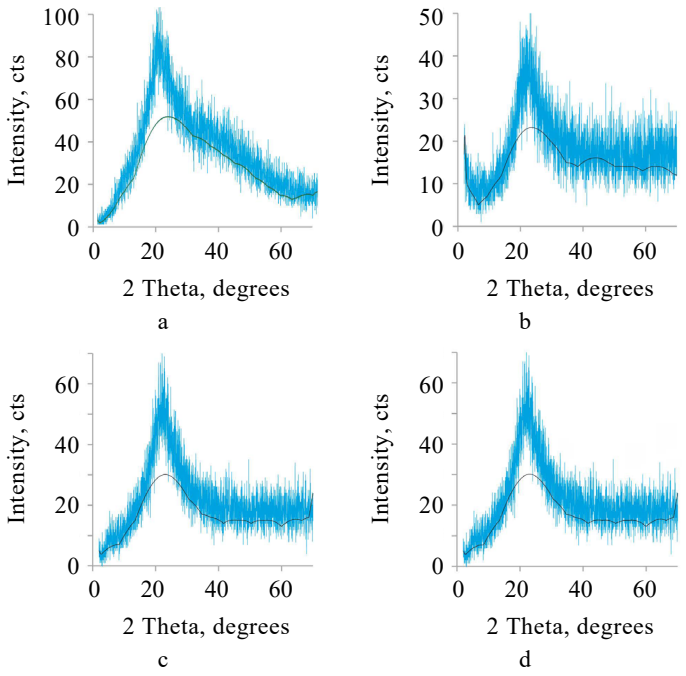


Figure 3 X-ray diffraction patterns of CMS powder (a), CMS film (b), CMS/PVA film (c), and CMS/PVA/kaolin film (d)

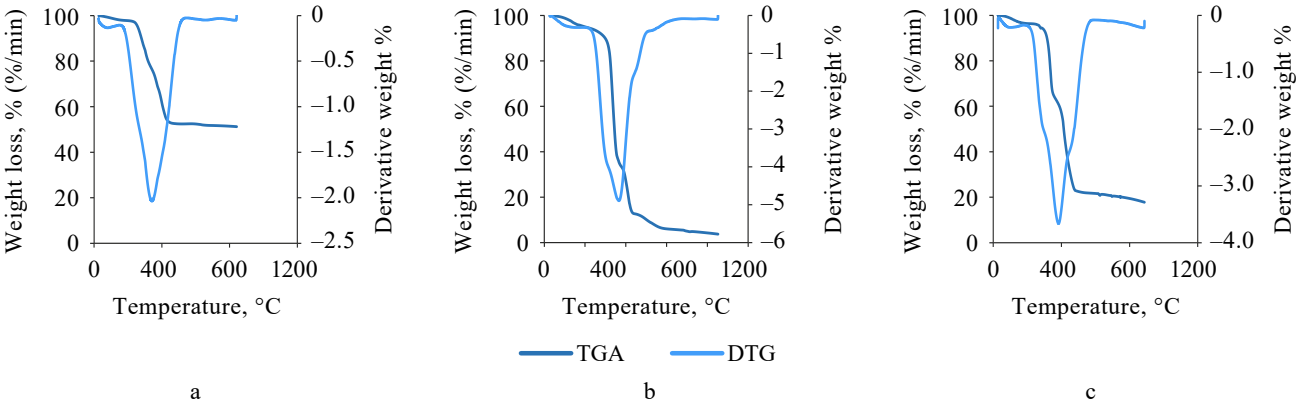


Figure 4 Thermogravimetric and derivative thermogravimetric curves of CMS powder (a), CMS film (b), CMS/PVA film (c), and CMS/PVA/kaolin film (d)

However, the incorporation of kaolin into the CMS/PVA films did not show any observable change in structural integrity. This can be seen from the non-distinguishable XRD peaks of the CMS/PVA and CMS/PVA/kaolin films, which was contrary to the scanning electron microscopy results. Neither did this finding agree with those reported by Yang *et al.* [33], who investigated the effect of additives on cross-linked CMS/PVA composite films. The authors observed that the addition of $CaCl_2$ and nano-SiO_2 to the cross-linked and PVA films improved their crystallinity and structural stability.

Thermal stability. Thermogravimetric analysis and derivative thermogravimetry were used to assess the thermal stability of carboxymethyl starch (CMS)-based composite films (Fig. 4). The thermal stability of CMS-based composite films is crucial to determine the maximum thermal conditions they can withstand without degrading or altering their structural characteristics [7].

The degradation process of the composite films occurred in three steps. The first step occurred at 25–200°C, with a weight loss of 0–6% for all the materials. The derivative thermogravimetry peaks were observed at 103, 160, and 105°C for the pure CMS film, CMS/PVA film, and CMS/PVA/kaolin film, respectively. The CMS film exhibited the lowest weight loss, while the CMS/PVA blend showed the highest weight loss. This initial degradation step corresponds to the loss of water molecules within the polymer matrix [34, 35]. The second step of degradation occurred at 200–350°C, with a weight loss of 5–10%. This step is attributed to the evaporation of volatile components like glycerol (melting point 290°C) [36]. The subsequent stage of degradation occurred at > 350°C and was attributed to the decomposition of composite components such as CMS, PVA, and/or kaolin [37]. This ultimate degradation step resulted in maximum weight loss for all the materials (Table 2).

Notably, the CMS film exhibited the lowest weight loss in this third stage apparently due to the absence of PVA and kaolin components.

PVA in the CMS film increased the maximum degradation temperature (DT_{max}) from 341.0 to 366.2°C, as can be observed in the derivative thermogravimetric peaks. However, the percentage weight loss for the CMS/PVA blend was significantly higher than that for the pure CMS film. In contrast, the CMS/PVA blend reinforced with kaolin exhibited a higher DT_{max} (383.3°C) and a lower percentage weight loss (39.1%) compared to the pure CMS and CMS/PVA films, indicating improved thermal stability.

Effect of polyvinyl alcohol on tensile mechanical properties of films. Figure 5 shows the effects of polyvinyl alcohol (PVA) on the tensile strength, percentage of elongation at break, and tensile modulus of the carboxymethyl starch (CMS) films. As can be seen, the addition of PVA (0–60%) into the CMS films resulted in a notable increase in tensile mechanical properties, with a 2.08-fold increase in tensile strength (Fig. 5a), a 1.31-fold increase in percentage of elongation at break (Fig. 5b), and a 1.58-fold increase in tensile modulus (Fig. 5c). The pure CMS film exhibited the lowest tensile strength, percentage of elongation at break (% EB), and tensile modulus, with values of 1.56 MPa, 25.40%, and 6.14 MPa, respectively. In contrast, the CMS/PVA film containing 60% of PVA demonstrated the highest values, with a tensile strength of 3.24 MPa, % EB of 33.3%, and tensile modulus of 9.32 MPa. This significant enhancement is attributed to the strengthening of intermolecular bonding between the CMS and PVA chains via hydrogen bonding [38]. The increased % EB values also indicate improved flexibility and toughness of the composite films. Similar findings were reported by Patil *et al.* [16], who investigated the effect of PVA incorporation (0–90%) into starch-based films. They observed a significant increase in tensile strength and the percentage of elongation at break of 1.24–1.93 and 1.93–3.74 fold, respectively. This consistency in results highlights the positive impact of PVA on the mechanical properties of CMS-based composite films.

Effect of kaolin on tensile mechanical strength of films. Figure 6 demonstrates the impact of kaolin on the tensile mechanical properties of the composites of carboxymethyl starch (CMS), polyvinyl alcohol (PVA), and kaolin. As can be seen, the introduction of 1.5 phr kaolin into the CMS/PVA film significantly increased its tensile strength by 18.8% from 3.24 to 3.85 MPa. Further increments in kaolin up to 4.5 phr resulted in a remarkable increase in tensile strength, reaching a maximum of 5.93 MPa (Fig. 6a). This enhancement may be attributed to a high degree of exfoliation and dispersion of kaolin in the polymer matrices of CMS and PVA [22]. However, a decrease in tensile strength to 5.82 MPa was observed at 5.5 phr kaolin. This reduction may be due to the kaolin content exceeding its compatibility limit and leading to the agglomeration and reduced interaction with the polymer matrix. Ariffin *et al.* [39] reported similar findings, where the flexural strength of polypropylene/kaolin composites exhibited an increase with rising kaolin contents, reaching an optimal value at 5 phr kaolin. However, further increases in kaolin beyond this threshold resulted in a decline in the mechanical properties of the composites.

The percentage of elongation at break (% EB) showed a slight decrease from 36.67 to 35.33% at 4.5 phr of kaolin (Fig. 6b), followed by a minor increase to

Table 2 Degradation steps of carboxymethyl starch-based composite films

Films	Step 1		Step 2		Step 3	
	DT peak, °C	Weight loss, %	DT peak, °C	Weight loss, %	DT_{max} peak, °C	Weight loss, %
CMS	103	1.14	256.4	4.8	341.0	23.6
CMS/PVA	160	4.05	294.5	9.6	366.2	64.6
CMS/PVA/Kaolin	103	1.33	302.3	6.9	383.3	39.1

DT is the degradation temperature; CMS is the carboxymethyl starch; PVA is the polyvinyl alcohol

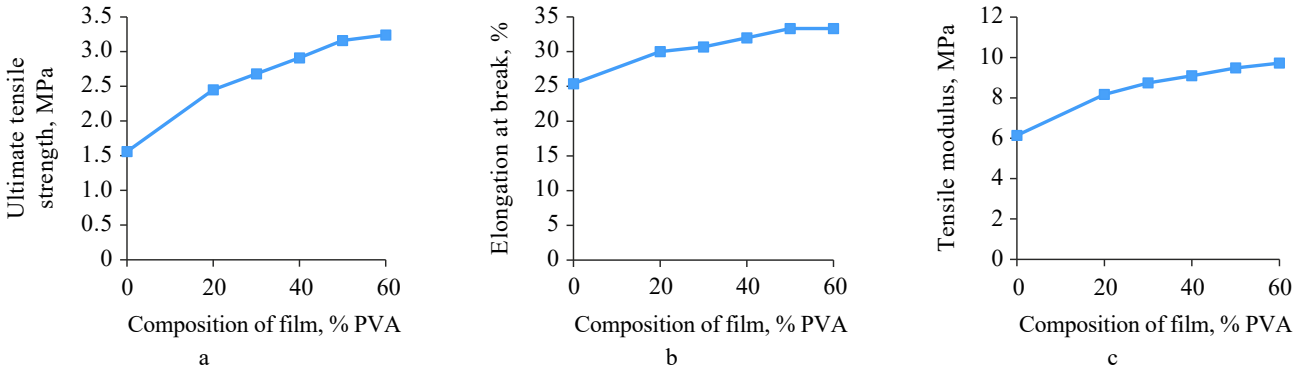


Figure 5 Plots of ultimate tensile strength (a), elongation at break (b), and tensile modulus (c) of carboxymethyl starch-based films versus percentage of polyvinyl alcohol

36.00% at 5.5 phr kaolin. This suggests that the addition of kaolin has a minimal impact on the flexibility of the composite films.

The effect of kaolin on tensile modulus (Fig. 6c) revealed a significant increase with rising kaolin concentrations, from 10.50 to 16.17 MPa. This enhancement in tensile modulus can be attributed to the presence of well-distributed rigid kaolin particles, as evident in the scanning electron microscopy results, which impose a mechanical restraint on the matrix by restricting its mobility and deformability [40].

Barrier properties. For a composite film to be considered suitable for packaging applications, it must possess sufficient barrier properties to prevent water vapor or air permeation, thereby ensuring the integrity of the packaged product [41]. The water vapor barrier property is typically measured in terms of water vapor permeability (WVP), where films with lower WVP values exhibit higher barrier properties.

Effect of polyvinyl alcohol on the water vapor permeability of films. Figure 7 shows the effect of polyvinyl alcohol (PVA) on the water vapor permeability (WVP) of carboxymethyl starch (CMS) films. As can be seen, the WVP of the CMS/PVA blend decreased by 28.9% with a PVA content from 0 to 60% (Fig. 7a). This is because the blending of PVA with CMS increases the intermolecular bonding between CMS and PVA via additional hydroxyl groups from PVA. This enhances their compatibility and lowers the penetration of water vapor molecules through the blend. The CMS/PVA blend containing 60% of PVA showed the lowest WVP (2.04×10^{-11} gm/m^{2}s·Pa), while the CMS film without PVA had the highest WVP (2.87×10^{-11} gm/m^{2}s·Pa).

Effect of kaolin on the water vapor permeability of films. The composite films showed a decrease in the water vapor permeability (WVP) from 1.5×10^{-11} to 0.945×10^{-11} gm/m^{2}s·Pa with the incorporation of 1.5–5.5 phr of kaolin (inorganic filler) (Fig. 7b). This result was in line with the studies by Yang *et al.* [33], who reported lower WVP values for cross-linked carboxymethyl starch-based composites with an increase in the inorganic filler (nano-SiO_2 particle). It might be that the polymer matrix system was reinforced by the exfoliation and intercalation of kaolin particles with carboxymethyl starch/polyvinyl alcohol chains. Consequently, this created a more tortuous pathway for water vapor molecules to diffuse through, thereby enhancing the barrier properties of the composite film [42].

Water vapor sorption characteristics. The water vapor sorption properties of carboxymethyl starch-based composite films were studied in terms of their sorption, diffusion, and permeability coefficients (Figs. 8 and 9). The effective diffusion coefficient was calculated from the slope of the early linear parts of the water vapor sorption curve.

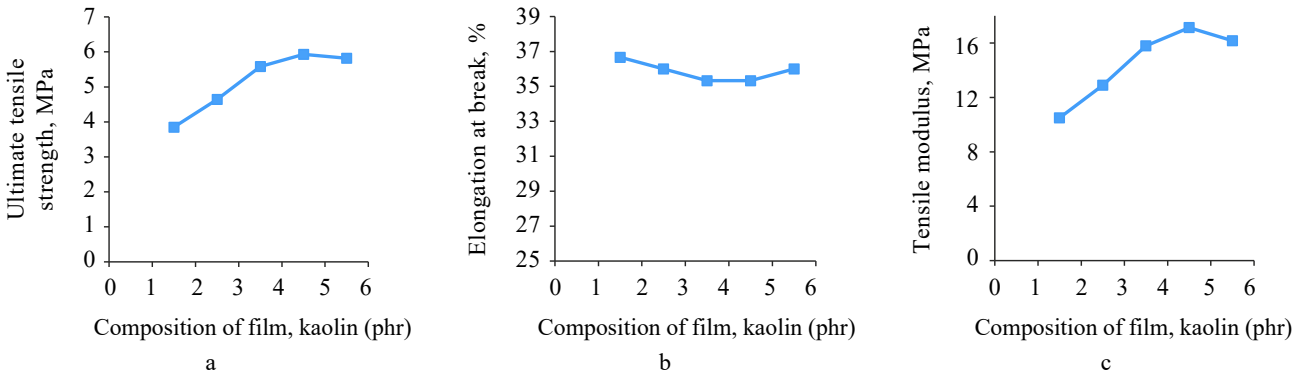


Figure 6 Plots of ultimate tensile strength (a), elongation at break (b), and tensile modulus (c) of carboxymethyl starch-based films versus kaolin content

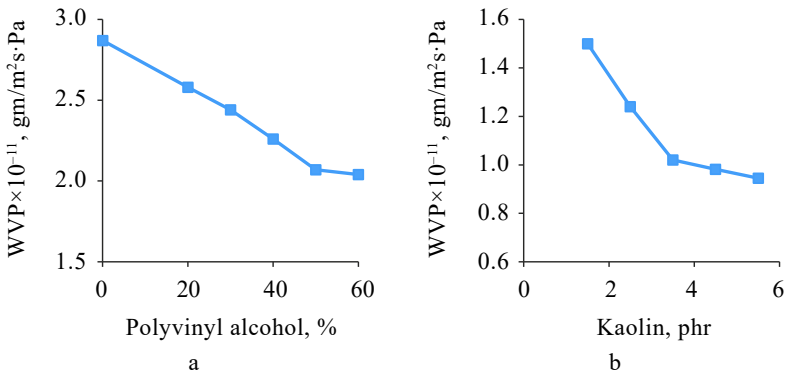


Figure 7 Effect of polyvinyl alcohol (a) and kaolin (b) on the water vapor permeability of carboxymethyl starch-based composite films

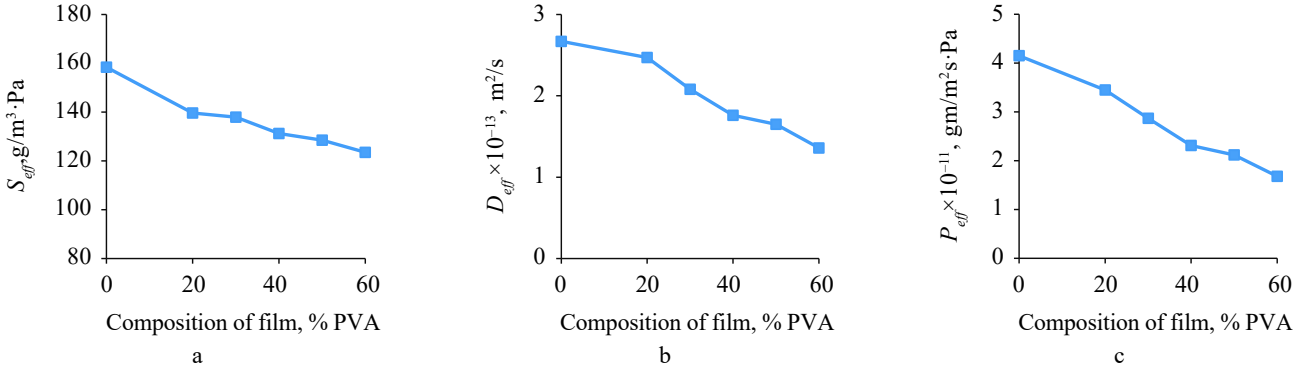


Figure 8 Plots of effective sorption S_{eff} (a), diffusion D_{eff} (b), and water vapor permeability P_{eff} (c) coefficients versus polyvinyl alcohol content

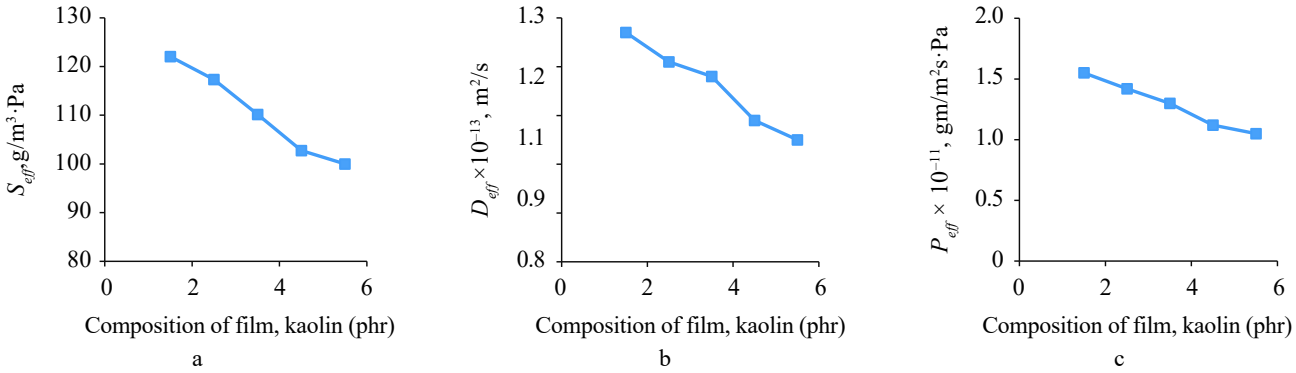


Figure 9 Plots of effective sorption S_{eff} (a), diffusion D_{eff} (b), and water vapor permeability P_{eff} (c) coefficients versus kaolin content

Effect of polyvinyl alcohol on the water vapor sorption. The influence of polyvinyl alcohol (PVA) on the water vapor sorption properties of the carboxymethyl starch composite films is illustrated in Fig. 8. The results revealed a progressive decrease in the effective sorption coefficients, diffusion coefficients, and water vapor permeability by 22% (Fig. 8a), 49.1% (Fig. 8b) and 59.5% (Fig. 8c), respectively, with increasing PVA concentrations (0–60%) in the films. Notably, the film containing 60% of PVA exhibited the lowest values for these parameters (S_{eff} 123.53 g/m³·Pa, D_{eff} 1.36 × 10^{-13} m²/s, and P_{eff} 1.68 × 10^{-11} g/m·Pa, respectively). This decrease can be attributed to the strong intermolecular interaction between carboxymethyl starch and PVA, which reduces the film's tendency to absorb moisture [43]. The formation of hydrogen bonds between the hydroxyl groups of starch and PVA leads to a more compact and dense film structure, thereby lowering the sorption, diffusion, and permeability coefficients [44]. The reduced water vapor permeability of the CMS/PVA composite films is desirable for packaging applications, as it helps maintain the quality and freshness of the packaged product by minimizing moisture absorption [45].

Effect of kaolin on the water vapor sorption. Figure 9 illustrates the impact of kaolin on the water vapor sorption properties of carboxymethyl starch (CMS) composite films. As can be seen, the incorporation of kaolin at concentrations of 1.5–5.5% resulted in a significant decrease in the sorption, diffusion, and permeability coefficients by 18.1% (Fig. 9a), 17.3% (Fig. 9b), and 32.3% (Fig. 9c), respectively. This reduction in the water vapor sorption properties of the blends indicated a decrease in the material's affinity for moisture [42]. This modification enhances the material's suitability for packaging applications, where minimal water vapor permeation is desired. The kaolin concentrations between 4.5 and 5.5 phr resulted in a slight decrease in water vapor sorption (2.7%), diffusion (3.7%), and permeability (6.3%) values. This subtle decline may indicate the peak kaolin concentration, beyond which further additions may not yield significant improvements. The dispersion of kaolin in the CMS/PVA polymer matrix creates a more tortuous pathway for water vapor molecules to diffuse through, thereby enhancing the material's water vapor resistance and barrier properties [42].

Biodegradation of carboxymethyl starch-based composite films. The biodegradability of polymers is a crucial aspect to consider in polymer applications, as these materials will ultimately be disposed of after their intended use. The ability of polymers to biodegrade naturally is essential for reducing the environmental impact and promoting sustainability [46]. The biodegradability of carboxymethyl starch (CMS)-based composite films was analyzed using the soil burial method (Figs. 10 and 11). This method provides a realistic environment for assessing biodegradability, as it simulates natural conditions and allows for the evaluation of microbial degradation [47]. The degree of biodegradability of the films

was studied by evaluating their weight loss for 56 days at periodic intervals of 7 days. As can be observed in Figure 10, all the film samples showed a significant increase in weight loss within the initial 7-day interval of burial. Specifically, the CMS/PVA blends (Fig. 10a) showed a 38–23% weight loss (decreasing with higher PVA contents, 0–60%), while the CMS/PVA/kaolin films exhibited lower weight loss (16–20%) dependent on the kaolin content (Fig. 10b). This rapid biodegradation can be attributed to the initial microbial colonization and degradation of the polymer surface [47].

However, the rate of biodegradation began to decrease during the subsequent days until the completion of the 56-day period. This decline can be due to the reduced availability of easily degradable materials and the increased complexity of the polymer structure [28]. Particularly, the rate of biodegradation of the CMS/PVA blends decreased by 22.2% with increasing PVA over the 56 days, while the pure CMS films exhibited the highest degradation rate of 90% (Fig. 11a). This suggests that CMS degrades at a faster rate than PVA, possibly due to its higher moisture absorption affinity. The absorbed moisture leads to swelling, creating a conducive environment for microbial growth on the film's surface [48]. In contrast, the incorporation of PVA into the CMS film appears to reduce biodegradability, potentially due to the formation of a more stable and compact polymer structure. This reduced biodegradability may result from PVA's ability to restrict the accessibility of microbes to the CMS chains, thereby slowing down the degradation process [49, 50].

In the CMS/PVA films reinforced with kaolin, the rate of biodegradation decreased by 30.4% with increasing kaolin contents (0–5.5 phr) (Fig. 11b). This is due to the formation of interfacial bonds between well-dispersed kaolin and the CMS/PVA matrix, which reinforces the polymer chain and renders it less accessible to the soil microbial attack [51]. Within the 56-day period, the films degraded quite significantly, if not completely, as evidenced by their substantial weight loss. This finding is crucial for the development of sustainable plastic packaging solutions, as it demonstrates the potential of these bioplastic films to degrade over time, reducing their environmental impact and promoting a more circular economy.

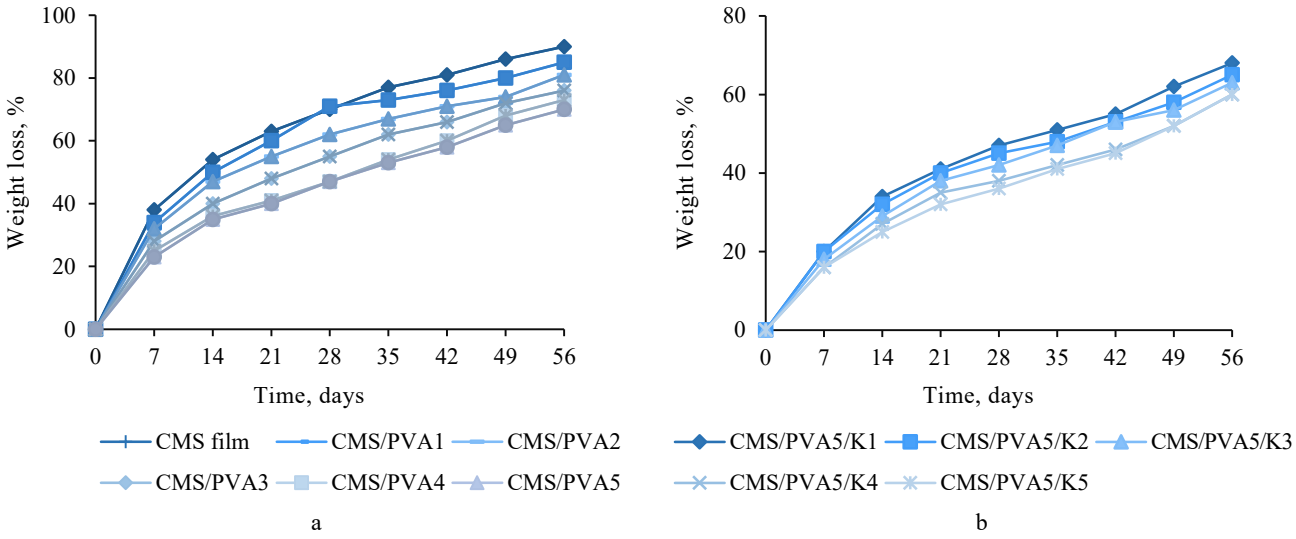


Figure 10 Soil burial biodegradability rates of carboxymethyl starch/polyvinyl alcohol (a) and CMS/PVA/kaolin composite (b) films for 56 days

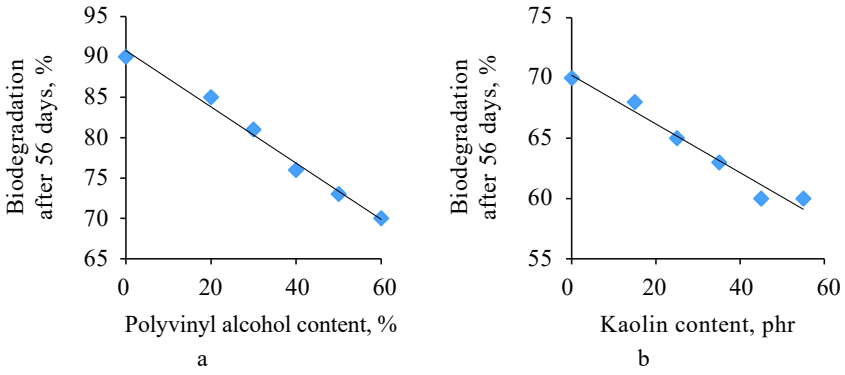


Figure 11 Degree of soil burial biodegradation of carboxymethyl starch-based films after 56 days versus polyvinyl alcohol (a) and kaolin (b) contents

CONCLUSION

In this study, we developed a composite film from carboxymethyl starch, polyvinyl alcohol, and kaolin for sustainable and eco-friendly bioplastic packaging. Blending carboxymethyl starch with polyvinyl alcohol at various concentrations and reinforcing the blend with kaolin yielded a bioplastic material with suitable properties for packaging applications.

The characterization results from scanning electron microscopy, X-ray diffraction, and thermogravimetric analysis revealed the composite's good surface microstructure, satisfactory crystallinity, and improved thermal stability, which affirmed the effective integration of the components.

The tensile mechanical test demonstrated significant improvements in the film's tensile strength and elongation at break with the addition of polyvinyl alcohol and kaolin, indicating enhanced durability. Furthermore, the inclusion of polyvinyl alcohol and kaolin substantially reduced the film's barrier properties in terms of water vapor permeability, making the bioplastic more effective at preventing moisture transfer.

Additionally, the water sorption ability of the composite film decreased with the incorporation of polyvinyl alcohol and kaolin, indicating improved resistance to water vapor absorption. Most importantly, the biodegradability test showed a significant level of biodegradation during the study period, which confirmed the composite's eco-friendly nature aligning with the principles of sustainability.

Conclusively, the carboxymethyl starch/polyvinyl alcohol/kaolin composite, with its improved functional properties, offers a viable alternative to conventional plastics in combating environmental concerns and plastic waste accumulation.

CONTRIBUTION

All the authors contributed equally and substantially to this research. Each author played a vital role in the conception, design, implementation, and interpretation of research findings, as well as in the overall preparation of this manuscript.

CONFLICT OF INTEREST

The authors state that there is no conflict of interest.

ACKNOWLEDGEMENTS

The authors wish to express their gratitude to the Head of Department of Chemistry, University of Benin for granting permission to use their laboratory facilities for this research and also to Mr. Isa Yakubu (Ahmadu Bello University) and Mr. Henry (University of Benin) for their technical assistance.

REFERENCES

1. Buryndin VG, Artyemov AV, Savinovskih AV, *et al*. Biostability of binder-free wood and plant plastics protected with antiseptics. Foods and Raw Materials. 2022;10(1):148–154. https://doi.org/10.21603/2308-4057-2022-1-148-154
2. Eremeeva NB. Nanoparticles of metals and their compounds in films and coatings: A review. Foods and Raw Materials. 2024;12(1):60–79. https://doi.org/10.21603/2308-4057-2024-1-588
3. Agarwal S. Major factors affecting the characteristics of starch-based biopolymer films. European Polymer Journal. 2021;160:11078. https://doi.org/10.1016/j.eurpolymj.2021.110788
4. Tabassum N, Rafique U, Qayyum M, Mohammed AAA, Asif S, *et al*. Kaolin–polyvinyl alcohol–potato starch composite films for environmentally friendly packaging: Optimization and characterization. Journal of Composite Science. 2024;8(1):29. https://doi.org/10.3390/jcs8010029
5. Shanmathy M, Mohanta M, Thirugnanam A. Development of biodegradable bioplastic films from Taro starch reinforced with bentonite. Carbohydrate Polymer Technologies and Applications. 2021;2:100173. https://doi.org/10.1016/j.carpta.2021.100173
6. Gómez-Aldapa CA, Velazquez G, Gutierrez MC, Rangel-Vargas E, Castro-Rosas J, *et al*. Effect of polyvinyl alcohol on the physicochemical properties of biodegradable starch films. Materials Chemistry and Physics. 2020;239:122–127. https://doi.org/10.1016/j.matchemphys.2019.122027
7. de Azevedo LC, Rovani S, Santos JJ, Dias DB, Nascimento SS, *et al*. Biodegradable films derived from corn and potato starch and study effect of silicate extracted from sugarcane waste ash. ACS Applied Polymer Materials. 2020;2(6):2160–2169. https://doi.org/10.1021/acsapm.0c00124
8. Compart J, Singh A, Fettke J, Apriyanto A. Customizing starch properties: A review of starch modifications and their applications. Polymers. 2023;15(16):3491. https://doi.org/10.3390/polym15163491
9. Omoike BA, Okieimen FE, Imoisi C, Abubakar MA. Characterization and evaluation of properties of cassava starch/poly(vinyl alcohol) films for food and pharmaceutical packaging applications. Singapore Journal of Scientific Research. 2024;14(1):34–42. https://doi.org/10.3923/sjsr.2024.34.42
10. Prasad J, Dixit A, Sharma SP, Mwakosya AW, Petkoska AT, *et al*. Nanoemulsion-based active packaging for food products. Foods and Raw Materials. 2024;12(1):22–36. https://doi.org/10.21603/2308-4057-2024-1-585
11. Spychaj T, Wilpiszewska K, Zdanowicz M. Medium and high substituted carboxymethyl starch: Synthesis, characterization and application. Starch – Stärke. 2013;65:22–33. https://doi.org/10.1002/star.201200159

12. Omoike BA, Okieimen FE, Imoisi C. Development of lemongrass oil-based starch/PVA/kaolin films: Antimicrobial properties and biodegradability. Science International. 2025;13(1):1–12. https://doi.org/10.17311/sciintl.2025.01.12
13. Bangar SP, Whiteside WS, Ashogbon AO, Kumar M. Recent advances in thermoplastic starches for food packaging: A review. Food Packaging and Shelf Life. 2021;30:100743. https://doi.org/10.1016/j.fpsl.2021.100743
14. Lee S, Kim ST, Pant BR, Song HH, Lee SK, *et al*. Carboxymethylation of corn starch and characterization using assymetrical flow field-flow fractionation coupled with multiangle light scattering. Journal of Chromatography A. 2010;1217:4623–4628. https://doi.org/10.1016/j.chroma.2010.04.082
15. Bhattacharyya D, Singhal RS, Kulkarni PR. Physicochemical properties of carboxymethyl starch prepared from corn and waxy amaranth starch. Carbohydrate Polymers. 1995;27:167–169. https://doi.org/10.1016/0144-8617(95)00062-C
16. Patil S, Bharimalla AK, Mahapatra A, Dhakane-Lad J, Arputharaj A, *et al*. Effect of polymer blending on mechanical and barrier properties of starch-polyvinyl alcohol based biodegradable composite films. Food Bioscience. 2021;44 (Part A):101352. https://doi.org/10.1016/j.fbio.2021.101352
17. Phattarateera S, Xin L, Amphong C, *et al*. Comparative studies of starch blends on the properties of PVA films. Carbohydrate Polymer Technologies and Applications. 2023;6:100340. https://doi.org/10.1016/j.carpta.2023.100340
18. Yurong G, Dapeng L. Preparation and characterization of corn starch/PVA/glycerol composite films incorporated with ε-polylysine as a novel antimicrobial packaging material. e-Polymers. 2020;20(1):154–161. https://doi.org/10.1515/epoly-2020-0019
19. Idris A, Muntean A, Mesic B, Lestelius M, *et al*. Oxygen barrier performance of poly(vinyl alcohol) coating films with different induced crystallinity and model predictions. 2021;11(10):1253. https://doi.org/10.3390/coatings11101253
20. Deng H, Su J, Zhang W, Khan A, Sani MA, *et al*. A review of starch/polyvinyl alcohol (PVA) blend film: A potential replacement for traditional plastic-based food packaging film. International Journal of Biological Macromolecules. 2024;273(Part 1):132926. https://doi.org/10.1016/j.ijbiomac.2024.132926
21. Gierszewska M, Jakubowska E, Olewnik-Kruszkowska E. Effect of chemical crosslinking on properties of chitosan montmorillonite composites. Polymer Testing. 2019;77:105872. https://doi.org/10.1016/j.polymertesting.2019.04.019
22. Rammak T, Boonsuk P, Kaewtatip K. Mechanical and barrier properties of starch blend films enhanced with kaolin for application in food packaging. International Journal of Biological Macromolecules. 2021;192:1013–1020. https://doi.org/10.1016/j.ijbiomac.2021.10.081
23. Kumari P, Kumari N, Mohan C, *et al*. Augmenting barrier efficiency in clay-based starch composite films for enhanced packaging sustainability. Polymers for Advanced Technologies. 2024;35(6):e6458. https://doi.org/10.1002/pat.6458
24. Labelle MA, Ispas-Szabo P, Vilotte F, *et al*. Carboxymethyl starch films as enteric coatings: Processing and mechanistic insights. Journal of Pharmaceutical Sciences. 2024;113(3):725–734. https://doi.org/10.1016/j.xphs.2023.09.002
25. Wilpiszewska K. Hydrophilic films based on starch and carboxymethyl starch. Green Sciences. 2019;21(2):26–30. https://doi.org/10.2478/pjct-2019-0016
26. Garavand Y, Taheri-Garavand A, Garavand F, Shahbazi F, Khodaei D, *et al*. Starch-polyvinyl alcohol-based films reinforced with chitosan nanoparticles: Physical, mechanical, structural, thermal and antimicrobial properties. Applied Sciences. 2022;12(3):1111. https://doi.org/10.3390/app12031111
27. Omoike BA, Okieimen FE, Imoisi C. Design and optimization of eco-friendly biocomposite films for packaging applications using response surface methodology. Tanzania Journal of Science. 2024;50(5):961–974.
28. Turan D. Water vapor transport properties of polyurethane films for packaging of respiring foods. Food Engineering Reviews. 2021;13:54–65. https://doi.org/10.1007/s12393-019-09205-z
29. Omoike BA, Okieimen FE, Imoisi C. Water vapour transport properties of cassava starch/poly(vinyl alcohol) films via experimental and fickian methods. Trends in Applied Sciences Research. 2024;19(1):225–232. https://doi.org/10.3923/tasr.2024.225.232
30. Chaisuwan K, Anurakumphan D, Hemmanee S, Ruamcharoen I, Leelakriangsak M. Soil burial degradation of starch-based films on microbial load and plant growth. Journal of Sustainability Science and Management 2023;18(3):110–124. https://doi.org/10.46754/jssm.2023.03.008
31. Rydz J, Šišková A, Eckstein AA. Scanning electron microscopy and atomic force microscopy: Topographic and dynamical surface studies of blends, composites, and hybrid functional materials for sustainable future. Advances in Materials Science and Engineering. 2019;2019:681785. https://doi.org/10.1155/2019/6871785
32. Nath D, Singh F, Das R. X-ray diffraction analysis by Williamson-Hall, Halder-Wagner and size-strain plot methods of CdSe nanoparticles- a comparative study. Materials Chemistry and Physics. 2020;239:122021. https://doi.org/10.1016/j.matchemphys.2019.122021
33. Yang L, Xie M, Fang J, Zhang T, Wang X, *et al*. Effect of additives on properties of cross-linked carboxymethyl starch/polyvinyl alcohol composite films. Journal of Applied Polymer Science. 2022;139(4):51546. https://doi.org/10.1002/app.51546

34. Manimaran M, Norizan MN, Kassim MHM, Adam MR, Norrrahim MNF, *et al*. Critical assessment of the thermal stability and degradation of chemically functionalized nanocellulose-based polymer nanocomposites. Nanotechnology Reviews. 2024;13(1):20240005. https://doi.org/10.1515/ntrev-2024-0005

35. Quilez-Molina AI, Meins JFL, Charrier B, Dumon M. Starch-fibers composites, a study of all-polysaccharide foams from microwave foaming to biodegradation. Carbohydrate Polymers. 2024;328:121743. https://doi.org/10.1016/j.carbpol.2023.121743

36. Luchese CL, Benelli P, Spada JC, Tessaro IC. Impact of the starch source on the physicochemical properties and biodegradability of different starch-based films. Journal of Applied Polymer Science. 2018;135(33):46564. https://doi.org/10.1002/app.46564

37. Basiak E, Lenart A, Debeaufort F. Effect of starch type on the physico-chemical properties of edible films. International Journal of Biological Macromolecules. 2017;98:348–356. https://doi.org/10.1016/j.ijbiomac.2017.01.122

38. Tan L, Sun B, Luo W, Liu S, Qiu B, *et al*. Enhanced mechanical property and water solubility of polyvinyl alcohol film via constructing hydrogen-bond network by adding carboxymethyl chitosan. Journal of Macromolecular Science, Part A. 2023;61(1):31–39. https://doi.org/10.1080/10601325.2023.2283045

39. Ariffin A, Ariff ZM, Jikan SS. Effects of kaolin and maleic anhydride contents on melt elasticity and flexural behaviour of polypropylene/kaolin and unplasticised poly(vinyl chloride)/kaolin composites. e-Polymers. 2010;10(1):095. https://doi.org/10.1515/epoly.2010.10.1.1069

40. Essabir H, Raji M, Bouhfid R, Qaiss AE. Hybrid nanocomposites based on graphene and nano-clay: Preparation, characterization, and synergistic effect. In: Qaiss AK, Bouhfid R, Jawaid M, editors. Graphene and Nanoparticles Hybrid Nanocomposites. Singapore: Springer; 2021, pp. 153–181. https://doi.org/10.1007/978-981-33-4988-9_5

41. Luo Q, Hossen MA, Zeng Y, Dai J, Li S, *et al*. Gelatin-based composite films and their application in food packaging: A review. Journal of Food Engineering. 2022;313:110762. https://doi.org/10.1016/j.jfoodeng.2021.110762

42. Dharini V, Selvam SP, Jayaramudu J, Emmanuel RS. Functional properties of clay nanofillers used in the biopolymer-based composite films for active food packaging applications – Review. Applied Clay Science. 2022;226:106555. https://doi.org/10.1016/j.clay.2022.106555

43. Ma N, Dong W, Qin D, Dang C, Xie S, *et al*. Green and renewable thermoplastic polyvinyl alcohol/starch blend film fabricated by melt processing. International Journal of Biological Macromolecules. 2024;279(Part 2):134866. https://doi.org/10.1016/j.ijbiomac.2024.134866

44. Castro JM, Montalbán MG, Martínez-Pérez N, Domene-López D, Pérez JM, *et al*. Thermoplastic starch/polyvinyl alcohol blend modification by citric acid–glycerol polyesters. International Journal of Biological Macromolecules. 2023;244:125478. https://doi.org/10.1016/j.ijbiomac.2023.125478

45. Abedi-Firoozjah R, Chabook N, Rostami O, Heydari M, Kolahdouz-Nasiri A, *et al*. PVA/starch films: An updated review of their preparation, characterization, and diverse applications in the food industry. Polymer Testing. 2023;118:107903. https://doi.org/10.1016/j.polymertesting.2022.107903

46. Moshood TD, Nawanir G, Mahmud F, Mohamad F, Ahmad MH, *et al*. Sustainability of biodegradable plastics: New problem or solution to solve the global plastic pollution? Current Research in Green and Sustainable Chemistry. 2022;5:100273. https://doi.org/10.1016/j.orgsc.2022.100273

47. Zee MV. Methods for evaluating the biodegradability of environmentally degradable polymers. In: Bastioli C. editor. Handbook of Biodegradable Polymers. Berlin, Boston: De Gruyter; 2020, pp. 1–22. https://doi.org/10.1515/9781501511967-001

48. Mittal A, Garg S, Premi A, Giri AS. Synthesis of polyvinyl alcohol/modified starch-based biodegradable nanocomposite films reinforced with starch nanocrystals for packaging applications. Polymers and Polymer Composites. 2020;29(5):405–416. https://doi.org/10.1177/0967391120922429

49. Lin YP, Dhib R, Mehrvar M. Recent advances in dynamic modeling and process control of PVA degradation by biological and advanced oxidation processes: A review on trends and advances. Environments. 2021;8(11):116. https://doi.org/10.3390/environments8110116

50. Lv Z, Meng X, Sun S, Jiang T, Zhang S, *et al*. Biodegradable carboxymethyl chitosan/polyvinyl alcohol hymexazol-loaded mulch film for soybean root rot control. Agronomy. 2023;13(9):2205. https://doi.org/10.3390/agronomy13092205

51. Abdullah ZW, Dong Y. Biodegradable and water resistant poly(vinyl) alcohol (PVA)/starch (ST)/glycerol (GL)/halloysite nanotube (HNT) nanocomposite films for sustainable food packaging. Frontiers in Materials. 2019;6:58. https://doi.org/10.3389/fmats.2019.00058

ORCID ID

Chinyere Imoisi https://orcid.org/0000-0001-9680-1635