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Physicochemical and Catechin Release Properties of Carboxymethyl Cellulose/ κ -carrageenan's Composite as Precursor of Active Green Packaging: A Preliminary Study

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Abstract— The ongoing dependence on conventional petroleum-based plastic food packaging, and the increasing necessity to protect food quality against lipid oxidation, pose significant challenges that requires comprehensive scientific examinations. In response to these challenges, the development of green food packaging with biopolymer composites, enhanced with antioxidant substances, has become an urgent and promising area of investigation. This preliminary study demonstrated the novel fabrication of green food packaging films by solvent casting blended carboxymethyl cellulose (CMC) and κ -carrageenan (κ -Car) at specific ratios, resulting in a CMC/ κ -Car biocomposites. Catechin (CAT), a model of phenolic-rich antioxidant, was incorporated into the CMC/ κ -Car composite matrix to simulate the release of active compounds. The efficacy of the composite synthesis was assessed using spectrometric characterisation and physicochemical property evaluation. Infrared spectroscopy validated the successful synthesis and confirmation of the CMC/ κ -Car composite, both in its original form and with CAT. Physicochemically, increasing κ -Car content improved tensile strength (TS), elongation at break (EAB), and water absorption capacity (WAC), with optimal performance at an 8:2 CMC/ κ -Car ratio. The sustained release profile of CAT over a specific time interval revealed that the composite acted as a carrier matrix, with diffusion-driven release kinetics, as predicted by the Higuchi and Korsmeyer–Peppas equations. The findings of this preliminary study indicated that, the CMC/ κ -Car composite, might have potential as a green food packaging material, improving shelf life and reducing lipid oxidation due to CAT's antioxidant properties.

Keywords— Active green packaging; Carboxymethyl cellulose; Catechin release profile; κ -carrageenan; Physicochemical properties.

1. INTRODUCTION

Food packaging is crucial for preserving product quality, serving as a barrier to against external pollutants, and extending the shelf life of food items. Currently, most food packaging relies on plastic sourced from petroleum-based substances, which are frequently structurally altered and augmented with chemicals to enhance their efficacy. Previous reports have indicated that the recurrent use of conventional packaging presents considerable hazards of food contamination. The situation is especially alarming due to the emission of micro- and nanoplastics, which may adversely affect the health of the human digestive system [1]. Furthermore, essential constituents of petroleum-derived plastics, such as ethylene and

propylene, are notoriously challenging to decompose organically. They require activities, such as pyrolysis or thermal destruction, for effective decomposition, rendering them persistent pollutants that contribute to environmental pollution [2]. Despite excellent thermal, mechanical, and barrier qualities of low-cost standard plastic food packaging, the use of recycled plastic for food covering introduces additional difficulties. The use of mixed plastic components and synthetic dyes in recycled packaging increases the risk of food contamination [3]. Considering these issues, further investigation into the advancement of sustainable food packaging materials is required. These alternatives should work as effective substitutes for petroleum-

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based polymers while also providing enhanced benefits in maintaining food quality and prolonging shelf life.

Active food packaging has emerged as a promising strategy to address the growing concerns surrounding the widespread use of petroleum-based plastic food packaging. Over the past five years, a noticeable trend has emerged in the development of active food packaging derived from biopolymeric materials, particularly those incorporating antioxidant compounds, which has significantly driven research exploration in this field [4–8]. This increasing interest was primarily attributed to the inherent advantages of biopolymer-based materials, including their natural biodegradability within a short timeframe, non-toxicity, and the abundant availability of raw materials as precursors for active food packaging [9,10]. Polysaccharide-based biopolymers such as starch [11], alginate [8], chitosan [12], pectin [13], carboxymethyl cellulose (CMC) [14], polyvinyl alcohol (PVA) [15], and carrageenan [5] have been extensively reported as potential candidates for packaging precursor materials. Among CMC, a cellulose derivative with a linear anionic polysaccharide structure composed of anhydroglucose units linked via β -1,4 glycosidic bonds, is particularly notable for its high-water solubility, non-toxic nature, and biodegradability [16]. CMC exhibited several advantageous properties, including the ability to form transparent films, water barrier performance, biodegradability, and ease of processing into film-forming solutions. These attributes have resulted in its widespread application as a component in the synthesis of food packaging composites [5,9,14,17,18]. However, CMC-based biofilms still encounter limitations, particularly regarding their suboptimal mechanical strength, relatively high-water vapour permeability, and poor stability in humid environmental conditions [14,17]. These challenges highlight the necessity for further modification through the incorporation of complementary materials to enhance the physicochemical properties of the resulting packaging films.

Carrageenan is a biopolymer obtained from red seaweed extract and distinguished by a sulphate functional group on its anionic polysaccharide backbone [19]. κ -carrageenan (κ -Car), a biodegradable, non-toxic, readily available, and cost-effective biopolymer, is one of six main structural types distinguished by the position and amount of sulphate groups, and has garnered significant attention for its broad applicability as a film-forming material [9,14]. Prior research has shown that the incorporation of CMC with κ -Car improved the mechanical properties of the resultant biofilm, as indicated by an increase in tensile strength from 20.31 MPa (CMC alone) to 28.54 MPa [14]. Moreover, the integration of κ -Car has demonstrated a reduction in water solubility and water vapour permeability in CMC/ghatti gum-based composites, making them appropriate as precursor materials for coffee

packaging [9]. κ -Car has been reported to enhance the heat stability and barrier characteristics of CMC-based composites, hence promoting their application as packaging films for strawberries [5]. The sulphate groups of κ -Car were thought to be crucial in establishing hydrogen bonds with the hydroxyl groups found in polymers like chitosan [19] and CMC. The incorporation of κ -Car into CMC-based biofilm systems is expected to improve their physicochemical characteristics, therefore broadening their potential use as active food packaging precursors.

Active food packaging utilizing a composite biofilm matrix of CMC/ κ -Car, enriched with antioxidant chemicals, constitutes a viable option for the development of functional food wrapping materials. The integration of antioxidants, which inhibit lipid oxidation and prevent the development of rancidity in food products, is a vital area of investigation for CMC/ κ -Car-based packaging systems. Antioxidant compounds are effective in reducing lipid peroxidation, therefore prolonging the shelf life of food goods [20,21]. Previous research has revealed the effective incorporation of natural antioxidants including tannic acid (TA) [19], gallic acid (GA) [20], thyme essential oil (TEO) [21], watercress oil [8], rose petal anthocyanins [4], wood vinegar [12], cannabidiol [13], and ferulic acid (FA) [15]. These compounds exhibited aromatic cores abundant in hydroxyl (–OH) groups, characteristic of phenolics or polyphenols, which function through hydrogen atom or electron donation mechanisms to neutralize free radicals, especially reactive oxygen species (ROS), within antioxidant defense frameworks [12,20–22]. Catechin (CAT), a natural phenolic chemical of the flavan-3-ol class, exhibits significant solubility in water and ethanol owing to its many hydroxyl groups. It demonstrated redox potential through hydrogen or electron transfer and stabilizes radical species via resonance delocalization across its aromatic ring structures [22–24]. These characteristics contributed substantial antioxidant properties to catechin, rendering it an optimal choice for integration into biopolymer-based food packaging matrices [23]. The release of catechin was delayed when incorporated into an amylose-rich biopolymer composite compared to its free form, due to interactions with the hydroxyl-rich polymer network [24]. As of now, and according to our comprehensive review of reputable literature, the incorporation of CAT into CMC/ κ -Car-based biofilm composites as a precursor for sustainable green food packaging systems has not been documented. This unexamined method offers a significant prospect for future investigation.

Despite the promise of CMC and κ -Car as sustainable packaging materials, existing studies often focus on single components or complex composites, leaving a gap in understanding the fundamental properties and practical viability of simple, binary blends of CMC and κ -Car fabricated by accessible

methods like solvent casting. To address this gap and offer a novel solution, the present work pioneers the development of green biocomposite films based exclusively on blends of CMC and κ -Car at systematically varied ratios. This study investigates preliminary efforts to reduce dependency on conventional petroleum-based polymers while simultaneously improving the shelf life and quality of food. The specific objective is to fabricate these CMC/ κ -Car films via solvent casting and comprehensively evaluate how their composition (ratio) influences key characteristics including spectroscopic analysis and physicochemical characterizations. Ultimately, the purpose of this preliminary study is to establish the feasibility and functional potential of tailor-made CMC/ κ -Car biocomposites as a straightforward yet effective route towards sustainable food packaging with improved performance.

2. EXPERIMENTAL SECTION

2.1. Materials

The materials used in the preliminary study were procured from Sigma-Aldrich and included CMC (average Mw ~250,000, degree of substitution 0.7, CAS No. 9004-32-4), κ -Car (CAS No. 1114-20-8), and CAT (CAS No. 18829-70-4). Supporting materials, specifically double-distilled water, were supplied by CV. ARD Pratama, Sleman, Indonesia.

2.2. Instrumentation

The successful synthesis of CMC/ κ -Car/CAT-based biofilm composite was verified via Infrared (IR) spectroscopy employing an Attenuated Total Reflectance (ATR) accessory (Spirit QATR-S, Shimadzu) within the wavenumber range of 400–4000 cm^{-1} . The mechanical performance, encompassing tensile strength (TS) and elongation at break (EAB), was assessed using a Universal Testing Machine (Zwick 0.5). The peak absorption wavelength and release profile of CAT were subsequently analyzed using a UV-Visible spectrophotometer (Shimadzu UV-1280).

2.3. Synthesis of CMC/ κ -Car/CAT-based Biofilm Composite

The synthesis of CMC/ κ -Car composites was performed according to a modified procedure derived from the literature [19]. CMC and κ -Car powders, totaling 100 mg, were mixed in different proportions (10:0; 9:1; 8:2; 7:3; and 0:10) and dissolved in 10 mL of double-distilled water. The mixture was continuously agitated with a magnetic stirrer at 60 °C for 1 h to ensure complete homogeneity. After obtaining a homogeneous solution, the composite mixture was transferred into a 6 cm diameter glass petri dishes using the solvent casting technique and then dried at 60 °C until a stable mass was reached, leading to the

creation of a transparent biofilm. The CMC/ κ -Car composite product was subsequently analyzed using ATR-IR and underwent physicochemical evaluation. To prepare the active compound-enriched composites, 10 mg of CAT was integrated into the homogenized CMC/ κ -Car solution before the casting process. The subsequent processes followed a similar solvent casting and drying procedure, succeeded by further characterization to evaluate the effects of CAT incorporation.

2.4. Mechanical Properties

The mechanical properties of CMC/ κ -Car biofilm composite, namely TS and EAB, were characterized. The CMC/ κ -Car biofilm composite was sectioned to dimensions of 30 mm in length and 5 mm in width. Each composite sample was positioned in the tensile grips of the UTM instrument and underwent TS and EAB testing at a rate of 1 mm/s, with a trigger force of 4.5 g and a deflection of 60 mm. TS and EAB were determined according to Equations (1) and (2), respectively, as outlined in the referenced methodology [17]:

$$\text{TS (MPa)} = \frac{F}{w \times d} \quad (1)$$

$$\text{EAB (\%)} = \frac{L_1}{L_0} \times 100\% \quad (2)$$

where F denotes the maximum force applied at break (N), w represents the width of the biofilm (mm), d is the thickness of the biofilm (mm), L_1 corresponds to the elongation at break (mm), and L_0 indicates the initial gauge length (mm). Each test was performed three times ($n=3$) on independent samples to ensure data reliability. Reported values are mean $\pm$ standard deviation (SD).

2.5. Water Absorption Capacity (WAC) Test

WAC analysis was conducted by immersing the CMC/ κ -Car composite films in 100 mL of double-distilled water for 1 h, following a method adapted from reference [19]. The WAC percentage (%) was calculated using Equation 3, where m_1 represents the wet mass of the biofilm (g) and m_0 denotes its dry mass (g). Each test was performed three times ($n=3$) on independent samples to ensure data reliability. Reported values are the mean $\pm$ standard deviation (SD).

$$\text{WAC (\%)} = \frac{m_1 - m_0}{m_0} \times 100\% \quad (3)$$

2.6. CAT Release Profile

The release profile of CAT was evaluated following the method described in reference [19]. The CMC/ κ -Car/CAT biofilm composite was immersed in 96% ethanol, as an organic solvent model that can perfectly dissolve the antioxidant CAT compared to water

solvents, at predetermined time intervals (1, 3, 6, 24, 48, 72, 96, and 120 h). At each interval, a 2 mL aliquot of the release medium was withdrawn and analyzed using UV-Visible spectrophotometry at the maximum absorbance wavelength of CAT (278 nm). To maintain the sink condition, an equal volume of fresh 96% ethanol was replenished into the system after each sampling. Each test was performed three times ($n=3$) on independent samples to ensure data reliability. Reported values are the mean $\pm$ standard deviation (SD). The release kinetics of CAT from the biofilm matrix were subsequently analyzed using the Higuchi and Korsmeyer–Peppas models, aiming to elucidate the diffusion-controlled mechanism of the active agent release from the polymeric film matrix. The Higuchi model, described by the Equation 4, with Q is the cumulative amount of drug released per unit area, k_H is the Higuchi release constant, and t is time, assumes a diffusion-controlled release from a homogeneous and non-swelling matrix under sink conditions, making it suitable for describing the square root time-dependent release of active compounds from solid matrices. The Korsmeyer–Peppas model, expressed as Equation 5, with M_t/M_∞ is the fraction of CAT released at time t , k is a kinetic constant, and n is the release exponent, provides a semi-empirical approach to evaluate drug release mechanisms from polymeric systems, with the value of n indicating whether the release follows Fickian diffusion, anomalous transport, or erosion-controlled kinetics.

$$Q = k_H t^{1/2} \quad (4)$$

$$\frac{M_t}{M_\infty} = k \cdot t^n \quad (5)$$

3. RESULT AND DISCUSSION

3.1. Structural Analysis of CMC/ κ -Car/CAT Biofilm Composite

The structural investigation of the CMC/ κ -Car composite and its interaction with CAT was performed using infrared spectroscopy, as seen in Fig. 1. The infrared spectrum of CMC presented in Fig. 1a exhibited distinct absorption bands at 3334 cm^{-1} (O–H stretching), 2903 cm^{-1} (asymmetric C–H stretching from methylene groups), 1590 cm^{-1} (carboxylate COO^- stretching), 1414 cm^{-1} (CH_2 bending), 1315 cm^{-1} (C–H bending), and 1022 cm^{-1} (C–O stretching within the HOC-O-CH_2 moiety) [25]. The functional groups of κ -Car, depicted in Fig. 1b, were characterized by absorption peaks at 3385 cm^{-1} (O–H stretching), 2956 cm^{-1} (asymmetric C–H stretching of methylene groups), 1635 cm^{-1} (H–O–H bending from water molecules, potentially indicating trace C=O contamination), 1224 cm^{-1} (O=S=O stretching of the SO_3^- group), 1033 cm^{-1} (C–O–C stretching from glycosidic linkages in the polysaccharide), 918 cm^{-1} (C–O–C

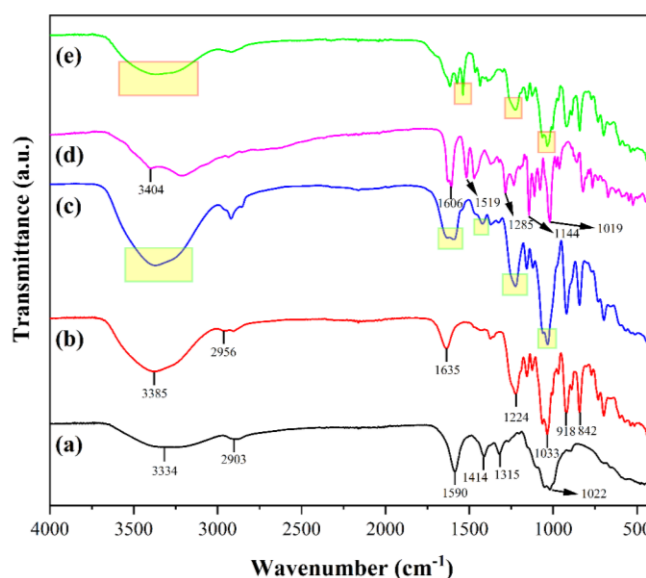


Fig. 1. Infrared spectra of CMC (a), κ -Car (b), CMC/ κ -Car (c), CAT (d), and CMC/ κ -Car/CAT (e)

stretching in 3,6-anhydro-D-galactose), and 842 cm^{-1} (C–O–S stretching of galactose-4-sulfate) [4,19]. The combined spectra (Fig. 1c) showed a shift to lower wavenumbers in the 3400–3200 cm^{-1} range, indicating hydrogen bonds between the –OH groups of CMC and the –OH/– SO_3^- groups of κ -Car [9]. Moreover, a reduction in intensity at approximately 1600 and 1410 cm^{-1} , associated with COO^- vibrations, indicated the existence of electrostatic interactions between CMC carboxylate and κ -Car sulfate groups. The augmented absorption at 1224 cm^{-1} was typically ascribed to the intensified asymmetric stretching of sulfate ester groups (SO_3^-), suggesting either an elevated sulfate concentration or a heightened engagement in intermolecular interactions, such as hydrogen bonding or electrostatic attractions with –OH or COO^- groups. The heightened intensity at 1030 cm^{-1} , linked to C–O–C stretching, suggested a more structured and compact glycosidic network, possibly established through interpolymeric hydrogen bonding or interactions with functional additives, indicating improved integrity and network durability. This improvement indicates a strengthened polymeric structure and enhanced structural integrity.

Fig. 1d and 1e depicted the spectra of CAT and the CMC/ κ -Car/CAT composite, respectively. The CAT spectrum (Fig. 1d) displayed significant absorption bands at 3404 cm^{-1} (O–H stretching from phenolic chains), 1606 cm^{-1} (aromatic C=C stretching), 1519 cm^{-1} (aromatic C–H stretching in methylene groups), 1285 cm^{-1} (C–O stretching from aromatic esters), 1144 cm^{-1} (aromatic in-plane C=C deformation), and 1019 cm^{-1} (C–O stretching) [26]. Upon the integration of CAT into the CMC/ κ -Car matrix (Fig. 1e), a significant decrease in intensity in the 3400–3200 cm^{-1} range was noted, signifying strong hydrogen bonding between the phenolic –OH groups of CAT and the –OH/– COO^- /– SO_3^-

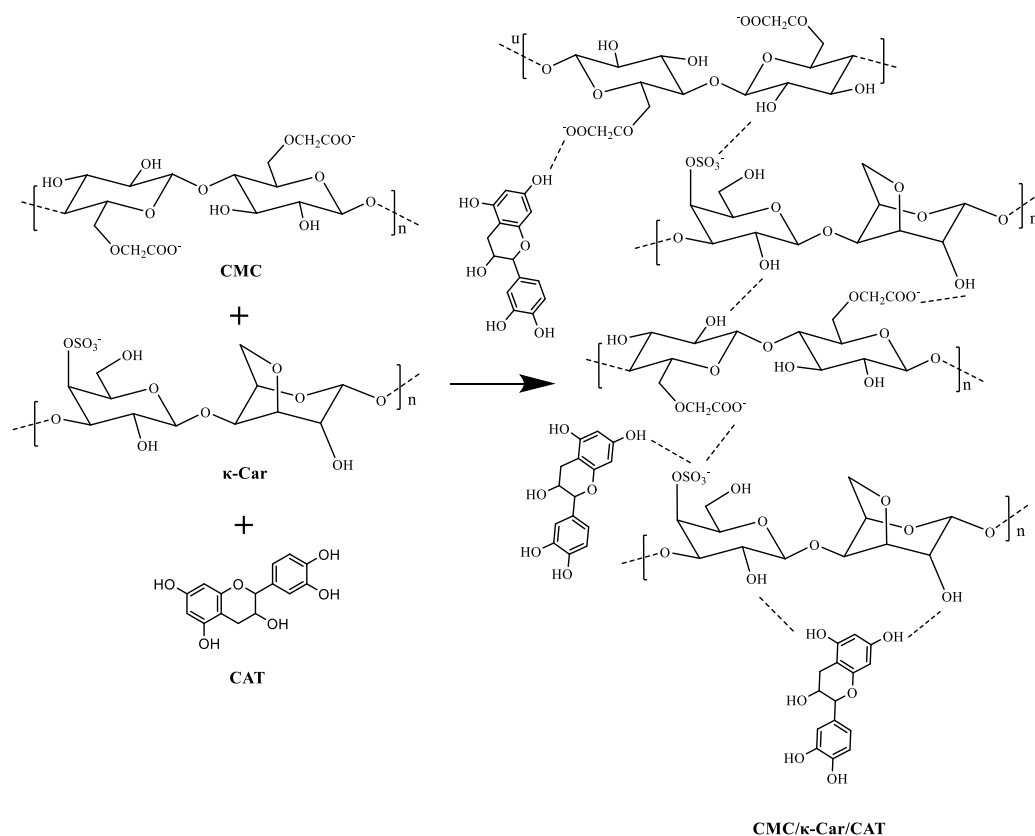


Fig. 2. Hypothesized interactions among CMC, κ -Car, and CAT

functionalities of the polymer matrix. The retention of the aromatic stretching bands within the range of ~ 1600 – 1500 cm^{-1} indicated that the aromatic structure of CAT was preserved, an essential characteristic for its antioxidant activity. Alterations in the C–O band at 1285 cm^{-1} indicated distinct interactions between the electron-donating groups of CAT and the electron-accepting moieties in the CMC/ κ -Car composite. Additionally, the overlapping absorption bands in the 1030 – 1070 cm^{-1} range indicated structural interactions between the C–O groups of CAT and the glycosidic bonds in the CMC/ κ -Car matrix. These interactions probably promoted network cohesiveness and structural integrity. These findings collectively suggested strong intermolecular hydrogen bonding and possible physical trapping of CAT within the polymeric structure, hence enhancing the composite's structural integrity and potentially improving its antioxidant efficacy. Based on the preceding structural analysis and literature review [27], the hypothesized interactions among CMC, κ -Car, and CAT are illustrated in Fig. 2.

3.2. Characterization of Structure and Morphology

The physicochemical characteristics of the CMC/ κ -Car composite were systematically analyzed through TS, EAB, and WAC evaluations. TS and EAB are critical mechanical parameters that reflect the structural integrity and flexibility of biocomposite films or bioplastics, directly affecting their suitability for various

packaging and biomedical applications [19]. **Fig. 3** presents the TS and EAB values as influenced by the incorporation of κ -Car, highlighting its impact on the mechanical performance of the resulting composite system. At a composition ratio of 10:0 (pure CMC), the resultant biofilm had a TS of $22.5 \pm 2.1\text{ MPa}$. The mechanical performance resulted from the efficient film-forming ability of CMC, which established a strong polymeric network via substantial interchain hydrogen bonding. Nonetheless, the EAB was low ($8.2 \pm 1.1\%$), signifying restricted flexibility, presumably attributable to the inherent rigidity of the CMC matrix. The addition of 10% κ -Car (ratio 9:1) resulted in an enhancement of both TS ($23.06 \pm 1.3\text{ MPa}$) and EAB ($10.5 \pm 0.9\%$). This improvement indicates that κ -Car contributed to strengthening the polymeric network, presumably via ionic interactions and synergistic effects between the two biopolymers. Significantly, at a ratio of 8:2, the mechanical parameters attained their maximum, with TS peaking at $25.7 \pm 1.5\text{ MPa}$ and EAB at $12.4 \pm 0.75\%$. These values indicate a balanced synergistic ratio in which structural reinforcement and chain mobility are optimally harmonized. Nonetheless, elevating the κ -Car content to a 7:3 ratio led to a reduction in TS to $23.9 \pm 2.3\text{ MPa}$, but the EAB remained comparatively high at $11.6 \pm 1.2\%$. The decrease in tensile strength may be due to phase heterogeneity or excessive plasticization caused by the surplus κ -Car, which could undermine the structural integrity of the biofilm. At the extreme

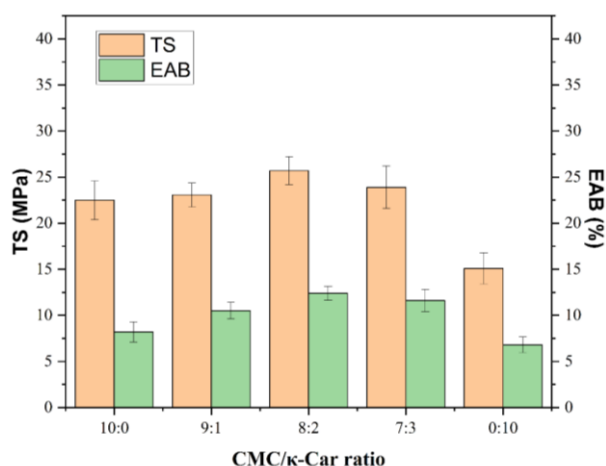


Fig. 3. TS and EAB value of CMC/κ-Car composite

end of the formulation range (0:10, pure κ-Car), both TS and EAB drastically decreased to 15.1 ± 1.7 MPa and $6.8 \pm 0.85\%$, respectively. The reduction is likely attributable to the inherent brittleness and inflexibility of κ-Car when utilized independently, along with the lack of crosslinking reinforcement provided by CMC. Furthermore, the addition of κ-Car as a modifier to CMC significantly improved the TS and flexibility of the resultant biofilm, with peak performance achieved at a CMC/κ-Car ratio of 8:2. This ratio established a compromise between a compact structural configuration and advantageous deformability. Excessive κ-Car content above this level negatively impacted the mechanical capabilities, likely due to the structural deficiencies and heightened brittleness caused by the imbalanced polymer network [17,19].

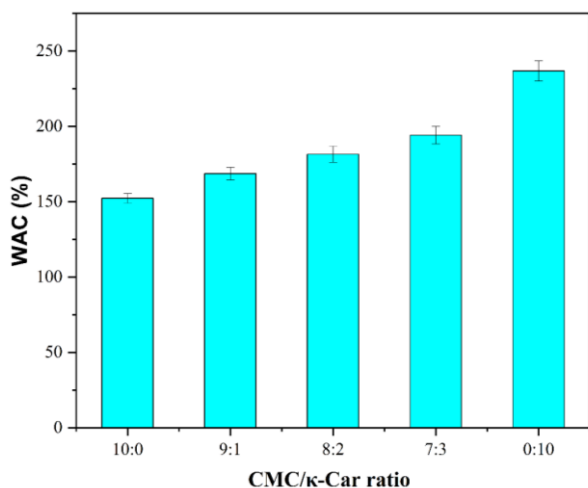


Fig. 4. WAC value of CMC/κ-Car composite

3.3. Release Study of CAT

The release profile of CAT (Figure 5) from the optimized CMC/κ-Car (8:2) biofilm matrix in 96% ethanol revealed a markedly sustained release behaviour compared to free CAT, which exhibited a rapid initial burst reaching over 90% within 24 h. In contrast, the

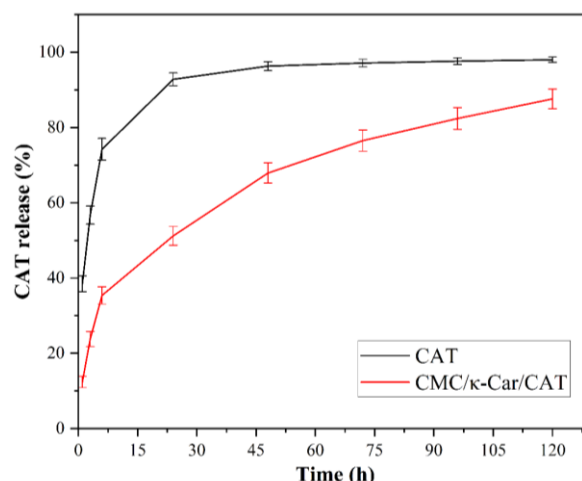


Fig. 5. Release profile of the CAT from the CMC/κ-Car composite

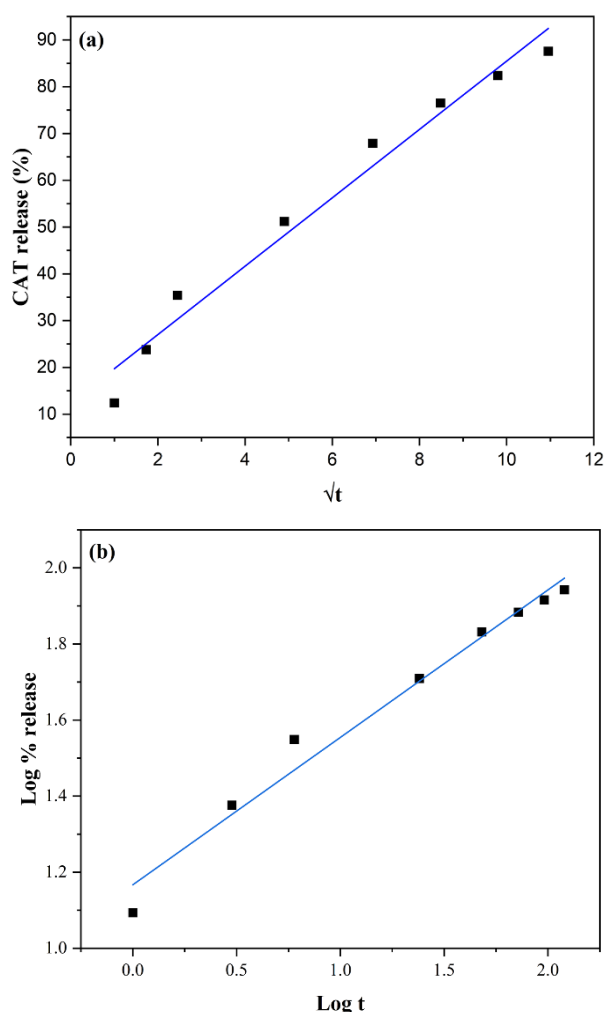


Fig. 6. Release kinetics of CAT from CMC/κ-Car composites based on the Higuchi (a) and Korsmeyer-Peppas (b) models

composite system demonstrated a gradual and slow-release, with only 51.2% released at 24 h and 87.6% after 120 h. This sustained release can be attributed to the polymeric network formed by hydrogen bonding and

ionic interactions between CMC and κ -Car, which likely restrict CAT diffusion and reduce its mobility within the matrix. The delayed release kinetics suggested that the composite film act as a diffusion barrier, modulating CAT release through a matrix-controlled mechanism. Standard deviation values remained within acceptable ranges, indicating good reproducibility across measurements. These results highlight the potential of the CMC/ κ -car matrix as a promising carrier for slow-release of polyphenolic compounds in hydrophobic environments.

Based on the release data of CAT from the CMC/ κ -Car (8:2) composite film in Figure 5, kinetic modeling revealed a strong fit to both the Higuchi and Korsmeyer–Peppas models (Figure 6), with R^2 values of 0.9744 and 0.9781, respectively. The Higuchi model (Figure 6a) yielded a linear regression equation of $y = 7.30x + 12.39$, indicating a diffusion-controlled release mechanism with a Higuchi release constant (k) of 7.31. Meanwhile, the Korsmeyer–Peppas model (Figure 6b) produced the equation $y = 0.38x - 0.83$, with a release exponent (n) of 0.388 and a kinetic constant (k) of 0.147, suggesting Fickian diffusion ($n < 0.45$) as the dominant release mechanism. These results collectively demonstrate that CAT release from the CMC/ κ -Car biofilm is primarily governed by a diffusion-controlled process through a polymeric matrix, confirming the suitability of the composite as a sustained release system [19]. In the future, CMC/ κ -Car composites may serve as viable eco-friendly food packaging materials, potentially prolonging shelf life and mitigating lipid oxidation due to the incorporation of CAT antioxidant properties.

CONCLUSION

The CMC/ κ -Car composite, both in its native form and when incorporated with CAT, was successfully synthesized and confirmed via infrared spectroscopy, as evidenced by the observable shifts in absorption bands corresponding to $-OH$, $-COO^-$, and $-SO_3^-$ functional groups. From a physicochemical profile, increasing the κ -Car content enhanced the TS, EAB, and WAC, with optimal performance achieved at a CMC/ κ -Car ratio of 8:2. The sustained release profile of CAT over a defined time interval suggested that the composite functions effectively as a carrier matrix, with release kinetics governed primarily by diffusion, as modeled by Higuchi and Korsmeyer–Peppas equations. The overall test results in this preliminary study had not yet been subjected to statistical analysis or in-depth testing parameter additions because the focus was more on finding the optimum formulation for the composite. However, this study would be a priority in the next phase of research. In the future, CMC/ κ -Car composites might be promising as green food packaging materials, capable of extending shelf life and

reducing lipid oxidation through the added CAT antioxidant action.

SUPPORTING INFORMATION

This article contains no supporting material. Requesting the relevant author (KSR), the data supporting the conclusions of this study are accessible.

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CONFLICT OF INTEREST

The authors declare that they possess no known conflicting financial interests or personal relationships that have the potential to impact this study findings.

AUTHOR CONTRIBUTIONS

KSR: Conceptualization, Formal analysis, Project administration, Supervision, Validation, and Writing – original draft. **W:** Supervision and Validation. **VM:** Conceptualization and Methodology. **EAP:** Conceptualization and Investigation. **AR:** Methodology and Investigation. **ES:** Methodology and Investigation. **BI:** Conceptualization, Supervision, Validation, and Writing – original manuscript.

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