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Sugar Palm Starch/Chitosan Bionanocomposite Films Incorporated with Anthocyanin and Curcumin – Thermal Properties and Release Kinetics

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ABSTRACT

The packaging industry responding to growing consumer demands for product safety, seeks active packaging that allows controlled antioxidant release through incorporating anthocyanin, curcumin, cinnamaldehyde, and other polyphenolic compounds to enhance functional properties of the film antimicrobial interfacial interaction. The research focuses on exploring the impact of adding curcumin and anthocyanin to sugar palm starch/chitosan bionanocomposite films, specifically examining the release kinetics of these bioactive compounds. The biocomposite film with added curcumin exhibits a smoother surface compared to the anthocyanin-based film. Although the thermal stability of the CH/SPS matrix remains unaffected by the addition of anthocyanin and curcumin, the inclusion of these compounds significantly reduces the melting enthalpy of the CH/SPS matrix. Specifically, the addition of curcumin decreases it from 142.96 J/g to 23.43 J/g, and the addition of anthocyanin reduces it to 33.22 J/g. Anthocyanin release from the CH/SPS matrix into water conforms to the Kosmeye-Peppas model (R^2 = 0.9808, n = 0.1177), while the release kinetics of curcumin compounds adhere to the Higuchi model (R^2 = 0.9968). These findings provide advantageous insights that potentially have implications for a variety of applications, particularly in areas such as sustainable food packaging.

Keywords: release kinetic, anthocyanin, curcumin, bionanocomposite film, sugar palm starch, chitosan.

INTRODUCTION

In the pursuit of environmentally friendly and sustainable packaging solutions, researchers have been actively engaged in extensive studies aimed at developing biodegradable biocomposite films (Zhao et al., 2023; Syarifuddin et al., 2023). These films hold immense promise as they combine natural components to create packaging materials that are not only eco-friendly but also possess enhanced functionalities (Yulianto et al., 2023; Bukit et al., 2023). With the global call for reducing plastic waste and environmental impact, these innovative bionanocomposite films represent a crucial step towards a greener and more sustainable future for the packaging industry (Wu et al.,2019; Sirait et al., 2023). Sugar palm starch and chitosan, two biopolymers with abundant availability, have emerged as prominent components in the design of bionanocomposite packaging films (Nazrin et al., 2021). Besides low cost, abundant in nature, and biodegradability, sugar palm starch exhibits excellent film-forming characteristics, while chitosan contributes barrier properties, nontoxic, and biocompatibility (Mousa et al., 2020). With the increasing consumer demand for packaging product safety, active packaging is highly desired by the packaging industry, where the controlled release of antioxidants within the packaging is possible

(Rossa et al., 2022). The release of active compounds in the packaging, such as anthocyanin, curcumin, cinnamaldehyde, and other polyphenolic compounds, will enhance the antimicrobial properties of the film through interfacial interaction (Ge et al., 2020). Curcumin, derived from turmeric rhizomes, has anti-inflammatory, antioxidant, and anti-cancer properties (Li et al., 2022), and has been investigated as a bioactive agent in functional packaging films for food applications (Filho and Egea, 2022). Anthocyanin, whether derived from mulberry or other sources, has UV barrier properties, enhances film color stability (Jamroz et al., 2022), pH-responsive properties for monitoring pH changes (Aliabbasi et al., 2021), and antibacterial properties that could be useful in inhibiting bacterial growth in food products (Roy and Rhim, 2020). Curcumin and anthocyanin both have the potential to improve the functionality and bioactivity of biocomposite packaging films, enabling them to be used in food packaging. The strategic combination of these biopolymers with curcumin and anthocyanin holds great promise in creating packaging films with enhanced functionalities and improved sustainability (Zhang et al., 2021).

This research aims to investigate the influence of incorporating curcumin and anthocyanin into sugar palm starch/chitosan bio-nanocomposite films, with a specific focus on studying the release kinetics of these bioactive compounds. Understanding the release behavior is vital to optimizing the film's functional properties and evaluating its potential applications in various industries, particularly in food packaging and preservation (Ma et al., 2022; Alinaqi et al., 2021). The study will employ comprehensive experimental methodologies to explore the release kinetics of curcumin and anthocyanin from the bionanocomposite films. The findings of this investigation hold significant implications for the packaging industry and the broader quest for sustainability. By elucidating the release kinetics of curcumin and anthocyanin in sugar palm starch/chitosan bionanocomposite films, we can advance our understanding of how these films can actively contribute to prolonging product shelf life, reducing food waste, and minimizing environmental impacts. Ultimately, this study endeavors to pave the way for the development of innovative, eco-friendly packaging materials that leverage the natural potential of bioactive compounds, steering us toward a more sustainable and health-conscious future.

EXPERIMENTAL

Material and methods

Nanoparticle chitosan (made using a ball milling technique), sugar palm starch (SPS) (Chimultiguna Co., Ltd., Indonesia, hexane, curcumin, and ethanol purchased from Sigma; anthocyanin from purple sweet potato extract (PSPE) (purchased from a local market in Banda Aceh).

Film preparation

The film was developed using the solvent-casting procedure. SPS was dissolved in 1.5% acetic acid to provide a 3% (w/v) SPS solution. Similarly, chitosan nanoparticles were dissolved in 30 mL of 1.5% acetic acid to generate a chitosan solution. Blending CH and SPS solutions (60:40 ratio), 30% w/w glycerol and 5% PSPE anthocyanin obtained the CH/SPS-Acy bio-nanocomposite. The CH/SPS-Cur bio-nanocomposite film was made in the same approach.

Atomic force microscopy

The surface roughness of the film samples was measured using an AFM (Veeco Instruments, Santa Barbara, CA, USA). The test was carried out on a 1.5 cm piece of the sample film, and the roughness of the film was calculated in terms of the root mean square roughness (Rq) and average roughness (Ra).

Thermal properties

Differential scanning calorimetry (DSC) analysis was used to examine the thermal properties of the films using a PT 1600 simultaneous thermogravimetric analyzer (Linseis Inc., USA). The samples were placed in an aluminum pan which was heated at a rate of 10°C/min under N_2 (20 mL/min) from 30°C to 600°C.

Release kinetic of anthocyanin and curcumin

The release of anthocyanins and curcumin from the bionanocomposite CH/SPS matrix was carried out following the procedure developed by (Cheng et al, 2021). Films sized 6×6 mm were placed in dark bottles containing 20 mL of water and soaked for specific time intervals (7.5, 15, 60, 120, 180, 240, and 280 minutes) while being continuously shaken at room temperature. The absorbance of the sample solutions was measured using a UV spectrophotometer at wavelengths of 530 nm and 450 nm, respectively, for the CH/SPS-Acy and CH/SPS-Cur samples. To determine the release profile involves calculating the percentage release at specific time points and creating a graph to illustrate the relationship between percentage release and time. Next, the release data of anthocyanin and curcumin from the CH/SPS film matrix is analyzed using different kinetic release models listed in Table 1. The interpretation of the results is based on the R^2 values obtained from the fitting. The model with the highest R^2 value indicates that the release mechanism aligns well with that particular kinetic model.

RESULTS AND DISCUSSION

Atomic force microscopy

AFM is frequently applied to evaluate morphology by assessing surface topography and film characteristics (Mwema et al., 2018). Figure 1 exhibits AFM images of CH-SPS-Acy and CH-SPS-Cur films. The films containing anthocyanins look porous and have a rougher surface (Ra = 110 nm) than the curcumin-based films (Ra = 78 nm). Nonetheless, the film developed in this work has a smoother surface than both native chitosan film (Ra = 210 nm) and starch film (Ra = 220 nm) (Hasan et al., 2022). No anthocyanin or curcumin residues were found in either film, showing good compatibility within the CS/CH blend (Dai et al., 2019). The lack of obvious clustering or agglomeration on the film surface showed the uniform distribution of anthocyanin and curcumin within the CS/CH matrix (Contardi et al., 2023; Carloz-Salazar, and Valderrama-Negron, 2017). This result is in line with the findings of (Ilyas et al., 2019) who revealed that adding sugar palm nanofibrillated cellulose to sugar palm starch resulted in similarly homogeneous film surfaces.

Thermal properties

Thermal evaluation techniques are crucial for characterizing structure-property correlations for single polymers and polymer composites (Frida et al., 2023). Figure 2 shows the thermal characteristics of the CH/SPS films. The DSC curves of the films demonstrated two consecutive endothermic peaks and one exothermic peak. The initial endothermic peak ranging from 137.6 °C to 156.4 °C, was attributed to the dehydration of the chitosan nanocomposite films (Shahbazi et al., 2017). The dehydration of the CH/SPS-Acy film takes place at a higher temperature than that of other films. This is affected by the presence of anthocyanins, which are hydrophilic due to the abundance of hydroxyl groups that interact with moisture (Amaregouda, et al., 2022). The second endothermic peak, which corresponds to the melting point, is reached at temperatures ranging from 352.1 °C to 356.7 °C. This CH/SPS film has a higher melting point than karaya gum film containing cinnamaldehyde (211.63 °C to 221.09 °C) (Cao and Song, 2018). This is due to nano-sized chitosan raw materials, which allow chitosan molecules to interact with starch effectively through hydrogen bond formation (Kumar et a., 2020). As a result, the molecular structure will become more crystalline (Qin et al., 2019). The addition of anthocyanin and curcumin to the chitosan/starch matrix has no significant effect on melting temperature, only slightly decreasing for the CH/SPS-Cur sample. However, adding these substances to the polymer matrix

Table 1. Release kinetic models

No.	Kinetic models	Equations ($y = ax + b$)	Type of linear plot
1	Zero-order	$\frac{M_t}{M_\infty} = k_0 t$	$\frac{M_t}{M_\infty}$ vs. t
2	First order	$ln\frac{M_t}{M_\infty} = k_1 t$	$ln\frac{M_t}{M_\infty}$ vs t
3	Kosmeyer-Peppas	$ln\frac{M_t}{M_\infty} = lnk_1 + n\ ln\ t$	$ln\frac{M_t}{M_\infty}$ vs *ln* t
4	Higuchi	$\frac{M_t}{M_\infty} = k_H t^{1/2}$	$\frac{M_t}{M_\infty}$ vs $t^{1/2}$

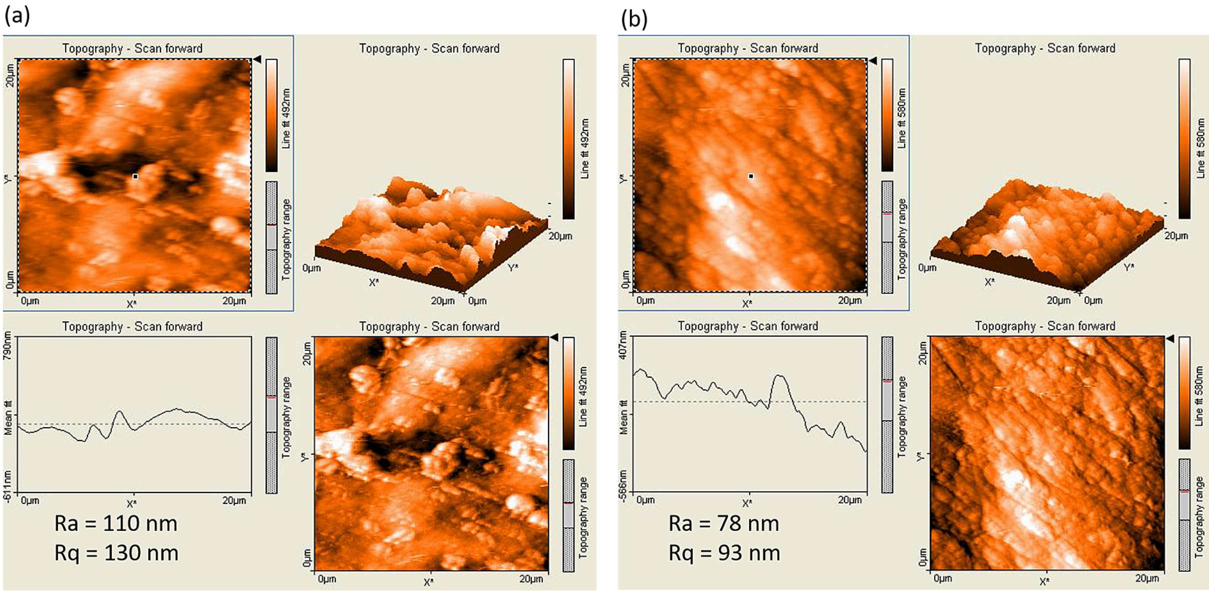

Figure 1. AFM images of (a) CH/SPS-Acy and (b) CH/SPS-Cur films

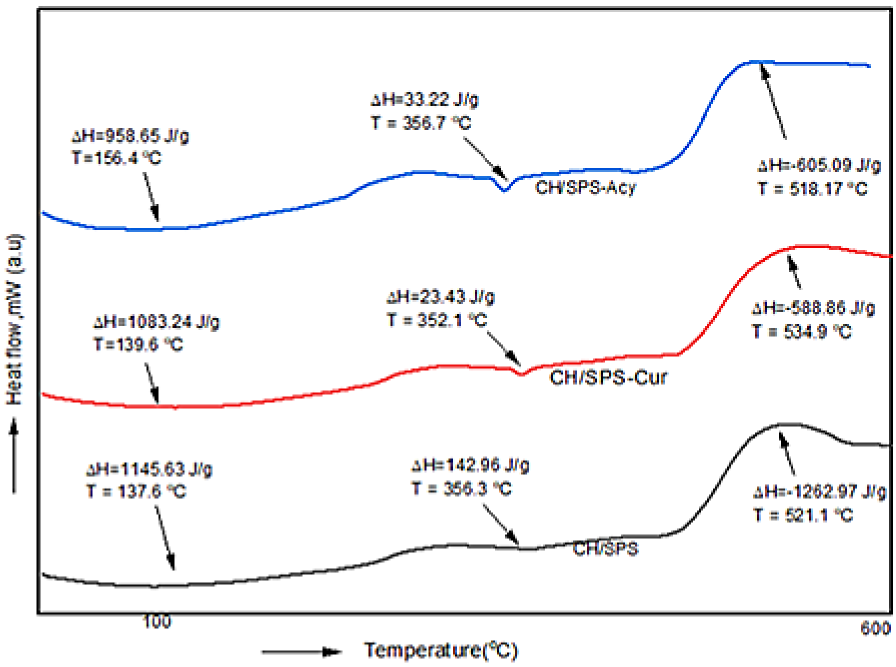

Figure 2. DSC thermogram of film samples

significantly reduces the enthalpy of fusion. This is due to structural changes in the film becoming more amorphous, which has a lower enthalpy of fusion than crystalline structures (Liu et al., 2021). The decomposition of the films can be observed by the exothermic peak between 518.17 and 534.4 °C. This result is consistent with corn starch and chitosan films with the addition of clove oil at degradation temperatures ranging from 473–600 °C (Hasan, et al., 2023).

Profile release

Figure 3 shows the profiles of the kinetic release of curcumin and anthocyanin from the CH/SPS-based active films into water. Curcumin has a greater release profile than anthocyanin due to its high hydrophobicity, which is caused by a relatively big phenolic core and hydrocarbon ring structure Contardi et al., 2023). Hydrophobic compounds, such as curcumin, tend to release more easily from hydrophilic matrices (e.g., chitosan/starch) due to lower hydrophobic-hydrophobic interactions (Hezma et al., 2019. Because of the presence of hydroxyl groups and oxygen atoms in its structure, anthocyanin is hydrophilic, resulting in a delayed release process via hydrophilic-hydrophilic contact (Cheng et al., 2022; Jiang et al., 2019). Profile of anthocyanin and curcumin release at different biocomposite matrices are depicted in Table 2, while anthocyanin and curcumin interaction among the bionanocomposite matrices are displayed in Figure 4. Primarily, the type of extracted food simulant, swelling rate, water solubility of the polymer, and diffusion of active compounds from the film to the simulant all affect the release of active compounds [Kuai et al., 2018].

Kinetic release

Figure 5 and Table 3 describe the results of the kinetic studies of anthocyanin and curcumin release from the CH/SPS matrix for zero-order, first-order, kosmeyer-peppas, and Higuchi. According to the plot in Figure 5, all models demonstrate extrapolation results with a tendency for a straight line. However, based on the R^2 values provided in Table 2, the CH/SPS-Cur sample has the highest R^2 value (0.9968) for the Higuchi model, while the CH/SPS-Acy sample has the highest R^2 value (0.9808) for the Kosmeyer-Peppas model. This suggests that the Higuchi model regulates the release mechanism of curcumin compounds in the CH/SPS matrix, while the Kosemeyer-Peppas model specifies the release mechanism of anthocyanin compounds in the CH/SPS matrix (Wojcik-Pastuszka et al., 2019;

Tabel 2. Profile of anthocyanin and curcumin release at different biocomposite matrices

Films formulation	Medium release	Findings	References
$Fe_3O_{4/}$Anthocyanin	water	21.1% anthocyanin released	Jiang et al., 2019
Red Cabbage Extract Anthocyanin + Acetylated distarchs phosphate	water	32.08 % anthocyanin released	Cheng, et al., 2022
Chitosan/ anthocyanin	Buffer phosphate pH = 6.5	28% anthocyanin released	Carloz-Salazar, and Valderrama-Negron, 2017
Sugar palm starch/chitosan + anthocyanin	water	27.5% anthocyanin released	This work
PVP + Zein powder + curcumin	Water at temperature 29 °C	39% curcumin released	Contardi et al., 2023
Nano hidroxy apatite/ PLA + curcumin	Saline	50% curcumin released	Hezma et al., 2019
Casein nanoparticles + curcumin	water	24.8% curcumin released	Jenifer and Upputuri, 2022
Sugar palm starch/chitosan + curcumin	water	33.95% curcumin released	This work

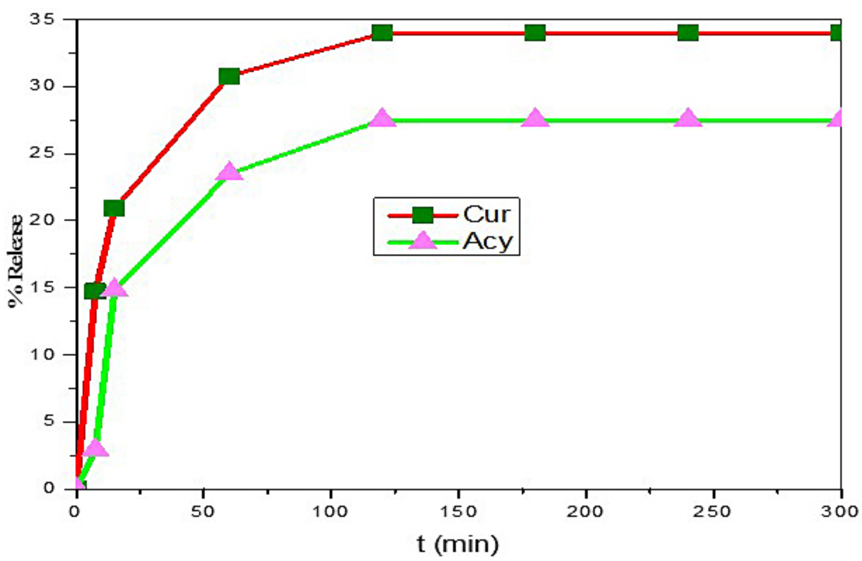

Figure 3. Profile release of anthocyanin and curcumin from CH/SPS matrices

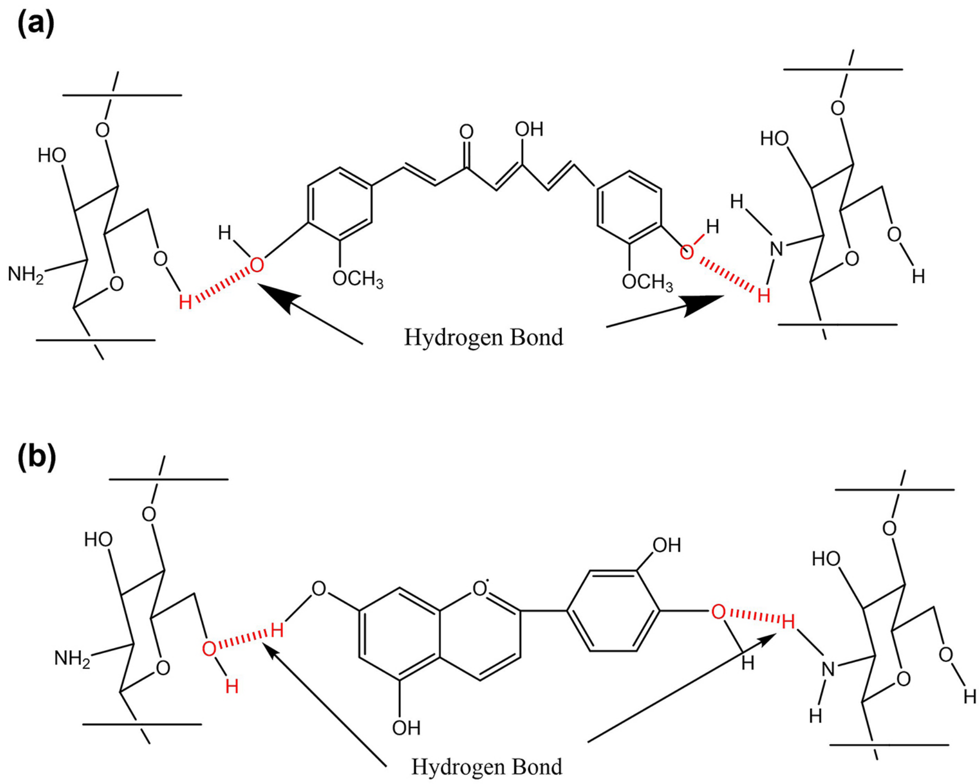

Figure 4. Bionanocomposite interaction through hydrogen bond formation between curcumin (a), and anthocyanin (b)

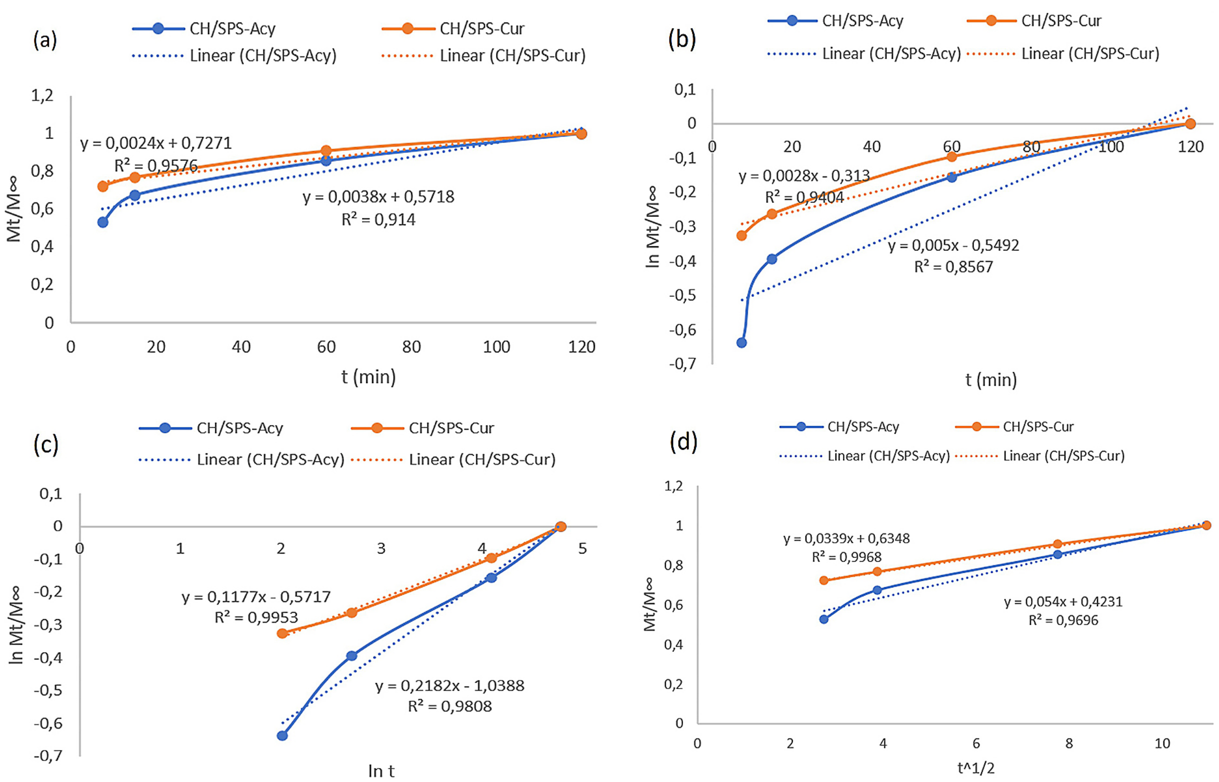

Figure 5. Release kinetics of anthocyanin and curcumin from CH/SPS bio-nanocomposite matrices, (a) zero order; (b) first order; (c) Kosmeyer-Peppas, and (d) Higuchi models

Table 3. Rate constant and determination coefficient

Film sample	Parameter	Kinetic model			
		Zero order	First order	Kosmeyer-Peppas	Higuchi
CH/SPS-Acy	R^2	0.9140	0.8567	0.9808	0.9696
	k	0.0038	0.0050	0.3539	0.0540
CH/SPS-Cur	R^2	0.9576	0.9404	0.9953	0.9968
	k	0.0024	0.0028	0.5646	0.0339

Hasan et al., 2022). As demonstrated in Figure 4.c, the coefficient value in 'n' for anthocyanin release from the CH/SPS matrix diverged significantly from Fickian behavior (n = 0.1177). The low 'n' values can be attributed to the partial solubilization of films in solvents, resulting in a non-Fickian process that contributes to compound delivery [Talon et al., 2017].

CONCLUSIONS

A bio-nanocomposite film based on sugar palm starch chitosan matrix has been synthesized. The surface of the biocomposite film added with curcumin is smoother than anthocyanin-based film. The addition of anthocyanin and curcumin did not affect the thermal stability of the CH/SPS matrix; however, the addition of these active compounds significantly decreased the melting enthalpy of the CH/SPS matrix, namely from 142.96 J/g to 23.43 J/g for the addition of curcumin and from 142.96 J/g to 33.22 J/g for the addition of anthocyanin. The release kinetics of anthocyanin compounds from the CH/SPS matrix into water follow the Kosmeye-Peppas model with a value of (R2 = 0.9808 and n 0.1177), while the release kinetics of curcumin compounds follow the Higuchi model (R^2= 0.9968). These findings provide valuable insights that may have implications for a wide range of applications, particularly in areas such as not just being environmentally friendly, but also functional and cost-effective sustainable food packaging.

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