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Edible Coloured Printing Paste for Food Packaging Materials Using Carboxymethyl Cellulose/Chitosan nanoparticles Composites as Thickeners

Entsar S. Abdou [1], H. M. El-Hennawi *[2], and K. A. Ahmed [2]

[1] Food Engineering and Packaging Department, Food Technology Research Institute, Agriculture Research Center, 12613 Giza, Egypt

[2] National Research Centre, Textile Research and Technology Institute, Dyeing, Printing and Intermediate Auxiliaries Department, 33 El-Behouth St. (former El-Tahrir str.), Dokki, P.O 12622, Giza, Egypt

Abstract

Chitosan is a biodegradable, biocompatible and non-toxic biopolymer that has the potential to produce edible film which possess antimicrobial properties. In this work, edible printing paste was developed using carboxymethyl cellulose (CMC) and chitosan nanoparticles (ChNPs) blends as thickeners with Hibiscus rosa-sinensis and Curcuma as natural dyes. CMC/ChNPs composites were prepared by adding different percent of chitosan nanoparticles (0.5, 1.00 %), with (1.5%, 2.0%) carboxymethyl cellulose. The elucidation of the prepared composites was established using, Fourier-transform infrared spectroscopy (FTIR) and Thermogravimetric analysis (TGA). The prepared composites were used as thickeners for printing paper and jute materials that are used for food packaging. The effect of ChNPs on the dyeing affinity of paper and jut was assessed using colorimetric data measurements reported as K/S values. The highest values of K/S were reached when using (2.0% CMC+1.0% ChNPs) thickener. Additionally, the printed samples were subjected to test their antimicrobial activity against both bacteria and fungi. The printed samples with CMC/ChNPs thickeners exhibited superior antimicrobial activity than that printed using (CMC) thickener only when tested against *Escherichia coli*, *Staphylococcus aureus* and *Candida albicans*. For all printed samples with CMC/ChNPs thickeners the reduction in CFU% reached to more than 96% for all tested microbes. The printed samples with (2.0% CMC+1.0% ChNPs) thickener have the best inhibition rate for tested bacteria and fungi. The colour assessment and rheological properties are also measured. The prepared thickeners exhibited non-Newtonian pseudoplastic behavior.

Keywords: Edible, printing paste, carboxymethyl cellulose, chitosan nanoparticles, thickeners

1. Introduction

Food contact materials and food packaging play a significant role in the attempt to supply people with healthy food across the globe [1]. Printing inks, laminating adhesives, and polymeric packaging materials all contain chemical compounds that enhance functionality [2]. These chemical compounds may interact with food components during processing or storage and drift into the food. When these substances exceed a predetermined threshold, the quality and safety of food may be compromised. Based on the toxicological risk evaluation of various compounds, migration limits have been established for numerus substances [3]. In order to limit the exposure to these contaminants and safeguard human health, these limits are incorporated into the Food Contact laws. Consumers are required to assess how packaging used in contact with food affects food safety. Manufacturers of printing inks and packaging, however, must be ready to deal with a complicated global regulatory environment, in which various jurisdictions have adopted multiple rules and regulations for the examination and authorization of food packaging and food contact materials, as well as for the issue of compliance statements.

* Corresponding author email: dr.heba.hennawi@gmail.com.; (Heba Mohamed)
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In response to the environmental risks posed by non-biodegradable packaging materials, recent research has focused on the development of biobased polymer packaging materials. Chitosan is one of the most widely used biopolymers because of its biocompatibility, biodegradability, antimicrobial characteristics, and simplicity of usage [4-7].

Carboxymethyl cellulose, a polysaccharide derivative, has shown a lot of interest in the textile printing paste business [8]. Because of its solubility in water and its ability to create films using CMC highly commendable [9]. It is employed in many printing pasts due to its stability in acids and alkalis within the pastes' pH range. In many recent researches CMC used for producing active and promising packaging materials [10-12].

This work exam developed printing paste using carboxymethyl cellulose (CMC) and chitosan nanoparticles (ChNPs) as edible composite multifunction thickener on Jute and printing paper that could overcome the disadvantages of using hazardous materials. Hibiscus rosa-sinensis is a fascinating natural dye that contains antibacterial compounds such as saponins, flavonoids, and tannins, Curcumin, a major bioactive constituent of turmeric (Curcuma longa L.), is testified as an antioxidant, anti-inflammatory, antibacterial, antifungal, and antiviral agent, these two natural dyes were used as colouring agents in the printing recipe.

2. Materials and Methods

2.1. Materials

Carboxymethyl cellulose (CMC) was obtained from Fluka Chemical Company. Hibiscus rosa-sinensis, Curcuma, white paper, and Jute fabric, were purchased from local market of Cairo.

2.2. Methods

2.2.1. Extraction of Chitosan

Chitosan was extracted from the shells of shrimp. Crushed shrimp exoskeletons were subjected to the standard procedures for extracting chitosan, which included treating with HCl (1M), NaOH (1-2M), and 40% NaOH. The degree of deacetylation (DDA%) of chitosan was determined by potentiometric titration [13] The molecular weight was established by using the value of intrinsic viscosity measured by an Ubbelohde viscometer [14]. Chitosan has a molecular weight of 3.98×10^4 gm/mol and a value of 92% for (DDA%).

2.2.2. Preparation of chitosan nanoparticles (ChNPs)

Following previous descriptions [13-18], nanoparticles were generated by ionic gelation of tripolyphosphate (TPP) and chitosan. A 2% solution

of TPP aqueous basic solution and a 2% solution of chitosan acidic solution, added dropwise in a 1:1 ratio under magnetic stirring at room temperature, resulted in the spontaneous formation of nanoparticles.

2.2.2. Chitosan nanoparticle (ChNPs) size distribution and average particle size determination

Using a Zetasizer nano series nano ZS (Malvern Instruments Ltd., Worcestershire, UK) at a wavelength of 633 nm and a scattering angle of 173° with a size range from 0.6 to 6000 nm, we were able to determine the average particle size, size distribution, and polydispersity indexes (PDI) of chitosan nanoparticles, Using ultra-purified water to dilute the samples ten times helped to avoid repeated light scattering occurrences [19].

2.2.3. Preparation of Carboxymethylcellulose/ chitosan nanoparticles thickeners

The stock paste of carboxymethyl cellulose was prepared by adding two different concentrations 1.5 and 2 gm of the powder to 100 ml of distilled water. The powder was stirred continuously in the water to prevent lump formation. After the formation of viscous stock, the chitosan nanoparticles powder was added by 0.5% and 1%. Finally, we have 5 thickeners samples 1.5% CMC (blank), 1.5% CMC+0.5% ChNPs, 1.5% CMC+1% ChNPs, 2% CMC+0.5% ChNPs and 2% CMC+1% ChNPs

2.2.4. Rheological properties

Using a Brookfield Digital Rheometer, model Brookfield AMETEk (Brookfield Engineering Laboratories INC.), the rheological characteristics of the prepared thickeners were examined at room temperature. The samples were measured using the RV-7 measuring equipment spindle. Spindle speeds between 10 and 100 rpm were used to study the rheological characteristics. The shear rate was determined using the following equation to display shear stress/shear rate rheograms:

$$\gamma = \left[\frac{2\pi R_c^2}{60\,(R_c^2 - R_b^2)}\right] \text{RPM} \qquad (1)$$

Where: γ is the shear rate, s^{-1}, R_c is the radius of the container, cm, R_b is the radius of spindle, cm

2.2.5. Fourier transform infrared (FTIR) spectroscopy.

The prepared CMC/ChNPs thickeners were characterized with FTIR spectra. The thickeners were measured in the transmission mode using KBr pellets in the range 400–4000 cm^{-1}. Perkin-Elmer 2000 spectrophotometer was used.

2.2.6. Thermogravimetric Analysis (TGA)

Thermogravimetric analysis (TGA) of the prepared CMC/ChNPs thickeners was carried out using Shimadzu TGA-50 H instrument at a heating rate of 10°C/min under nitrogen atmosphere.

2.2.7. Colour measurements and fastness properties

At λ_{max}, Hunter Lab Ultra scan PRO assessed the printed samples' colour strength. To further understand the variation in colour between the blank and CMC/ChNPs printed samples, we additionally used the following relationships.

$$\Delta E = \sqrt{(\Delta L)^2+(\Delta a)^2+(\Delta b)^2} \quad (2)$$

where $\Delta L = L - L^*$; $\Delta a = a - a^*$; $\Delta b = b - b^*$. Lightness is described by L, whereas redness or greenness is measured by a and yellowness or blueness by b. L*, a*, and b* are the colour parameters of the control samples. Fabrics.

Fastness properties of all printed jute samples against washing, rubbing perspiration and light were measured according to the standards methods [20-24]

2.2.8. Antimicrobial measurements

The antimicrobial activity of printed jute and paper samples tested against gram +ve bacteria (*Staphylococcus aureus*), gram –ve bacteria (*Escherichia coli*) and fungi (*Candida albicans*). The antimicrobial efficiency was evaluated according to the standard test method, AATCC 100–1999 for bacterial counting [25, 26]. Equation (3) was used to calculate the reduction percentage of bacterial colonies. [27, 28]

$$R\% = ((B - A) / B) \times 100 \quad (3)$$

In this equation, B is the number of bacterial colonies from unprinted fabrics and A is the number of colonies from printed samples. The antimicrobial activity of the printed samples was then assessed.

3. Result and Discussion

3.1. Size and size distribution of chitosan nanoparticles (ChNPs)

Figure 1 displays the findings of the particle size and particle size distribution of ChNPs. As shown in Figure 1 ChNPs have a mean size of 19.18 nm.

The homogeneity of the synthesized nanoparticles expressed by the polydispersity index (PDI), which is the main factor for evaluating the nanoparticles stability. A value closer to 1 denotes a broad range of particle sizes , whereas a lower PDI value depicts a monodisperse particle size distribution [29]. PDI of the prepared ChNPs was found to be 0.626.

3.2. Rheological properties

3.2.1. Flow behavior of CMC/ChNPs thickeners

Figure (2a and 2b) illustrates the Rheograms (shear rate, s^{-1} versus shear stress, Pa) of different concentrations of CMC/ChNPs thickener fresh prepared (2a) and after 3 days of storage (2b). Rheological investigation of all blend pastes showed non-Newtonian pseudoplastic behavior for fresh prepared samples and after 3 days of storage [30-35] and following the power law equation:

$$\tau = k\gamma^n \quad (4)$$

The variables τ (shear stress), γ (shear rate), k (consistency index), and n (flow behavior index).

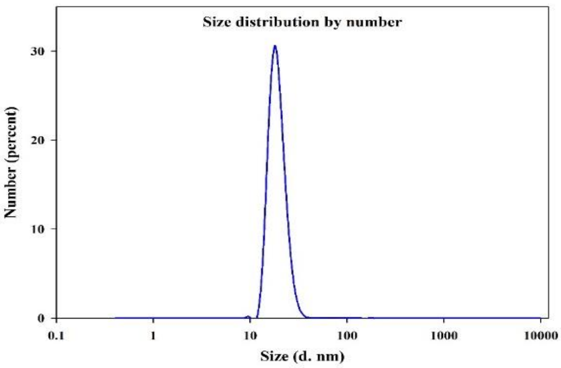

Figure 1. Particle size (nm) and Particle size distribution of chitosan nanoparticles

Table 1 presents the parameters obtained by fitting to the power law model for CMC/ChNPs thickeners fresh prepared and after 3 days of storage. The consistency index (k, $Pa.s^n$) and flow behaviour index (n). The degree to which the fluid exhibits non-Newtonian properties is indicated by the flow behaviour index (n) when the fluid becomes more shear thinning (n) decreases. For all prepared thickeners $n < 1$, which corresponds to pseudoplastic behavior as shown in table 1.

3.2.2. Shear rate effect on the viscosity of CMC/ChNPs thickeners

Figure (3a and 3b) represents the relation between viscosity values (Pa.s) and shear rate (s^{-1}) for fresh prepared samples (3a) and after 3 days of storage

(3b). As shown in figure (3a and 3b) the shear rate has a significant impact on viscosity when the shear rate values increased, the viscosity values decreased for both fresh and stored samples. Figure (3a and 3b) also illustrated that viscosity decreased significantly with storage.

Addition of ChNPs within concentration 0.5 to 1.0 wt% to different concentration of CMC caused a slight increase in the overall viscosity of the prepared thickeners and accordingly the viscosity of the printing paste. Thus, ChNPs in small proportions can act as a benign co-thickener to CMC in edible printing paste.

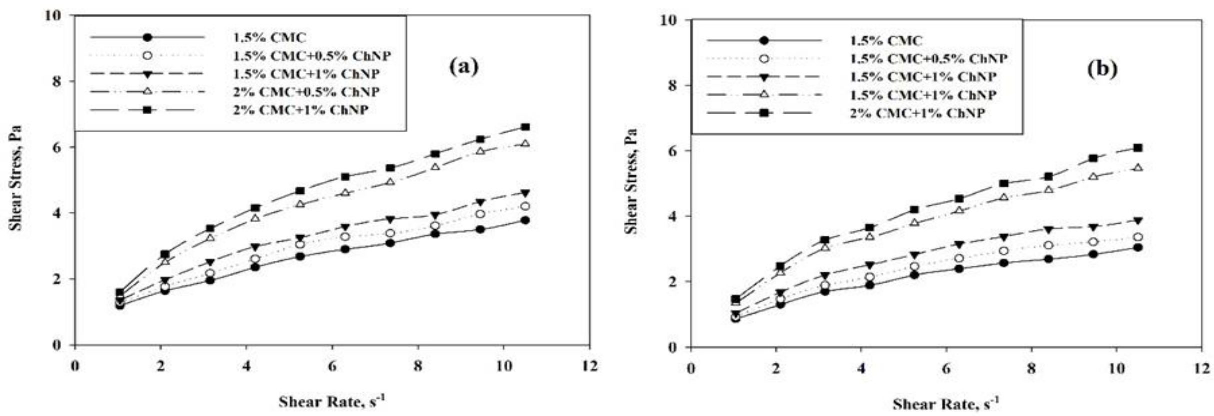

Figure 2: Rheograms of different concentrations of (a) fresh prepared CMC/ChNPs thickener (b) stored CMC/ChNPs thickeners for 3 days

Table 1: The consistency index K ($Pa.s^n$), flow behavior index (n) and Coefficient of Determination (R^2) of CMC/ChNPs thickeners fresh prepared and after 3 days of storage.

Sample	Fresh			After 3 days		
	n	k ($Pa.s^n$)	R^2	n	k ($Pa.s^n$)	R^2
Blank 1.50 %CMC	0.506	1.1353	0.9978	0.5393	0.8716	0.9949
1.5% CMC+0.5% ChNPs	0.5237	1.1353	0.9963	0.5571	0.9521	0.9927
1.5 %CMC+1.0% ChNPs	0.5237	1.3507	0.9963	0.5656	1.0841	0.9896
2.0 %CMC+0.5% ChNPs	0.5955	1.5463	0.9915	0.587	1.0841	0.9894
2.0 %CMC+1.0% ChNPs	0.5929	1.6904	0.9885	0.5931	1.5361	0.9909

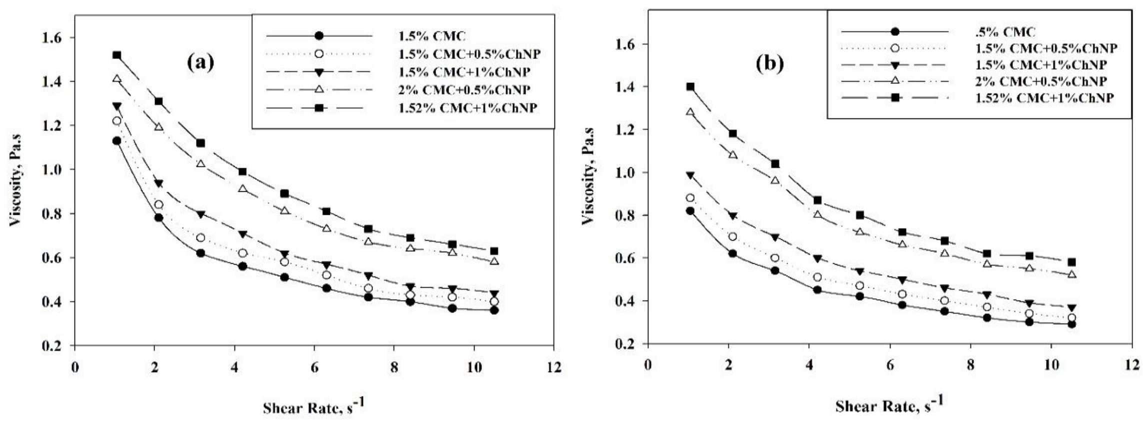

Figure 3: Effect of shear rate on viscosity of (a) fresh prepared (CMC)/(CNCs) thickener (b) stored (CMC)/(ChNPs) thickeners for 3 days

3.2.3. Fourier transform infrared (FTIR) spectroscopy.

Figure 4 illustrates FTIR spectra for CMC and CMC/ChNPs blends they have almost identical spectra showed broad band around 3293 cm^{-1} coexisted in CMC and all blends this band caused by –OH stretching and intermolecular/intramolecular hydrogen bonds. The band near 2919 cm^{-1} due to −CH stretching observed for CMC and all other blends. The sharp peak around 1585 cm^{-1} could be due to −N-H bending of the primary amine of chitosan this peak appears in all CMC/ChNPs blends [36]. The asymmetry stretching peak of −COO located around 1413 cm^{-1} may be due to the strong electrostatic attraction between [$-NH_3^+$] of ChNPs and [−COO] of CMC [37]. The band around 1060 cm^{-1} is generally thought to have arisen from polysaccharide glycosidic linkages [38]. Blending ChNPs with CMC led to decrease in peaks intensity as illustrated in figure 4. Generally, because both components are polysaccharides and so display bands in the same wavenumber interval, it is difficult to get more details about the presence of ChNPs in the nanocomposite blends using FTIR analysis.

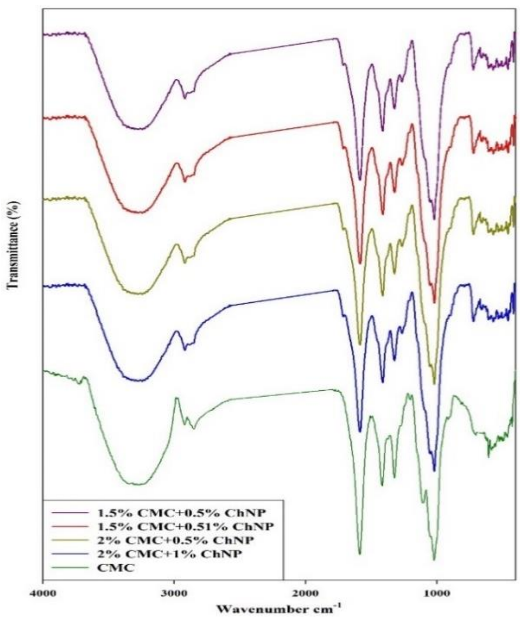

Figure 4: FTIR of CMC and CMC/ChNPs blends

3.2.4. Thermogravimetric Analysis (TGA)

The TGA thermogram for CMC and its blends with chitosan nanoparticles at different concentration 0.5 and 1 % are illustrated in figure 5. Multiple steps are seen in the breakdown pattern. In the first stage, weight loss of around 11% is seen for all blend thickeners between 35 and 158°C, and a loss of roughly 14% for CMC between 35 and 171°C. In the second stage, the prolonged weight loss (about 84%) at 184 °C to 784°C for CMC thickener while the other blends loss (about 45-57 %) at 159 to 177 °C this is could be related to the removal of the residual water of the nanoparticles and degradation of the polymer. Over all the CMC/ChNPs blends are thermally stable.

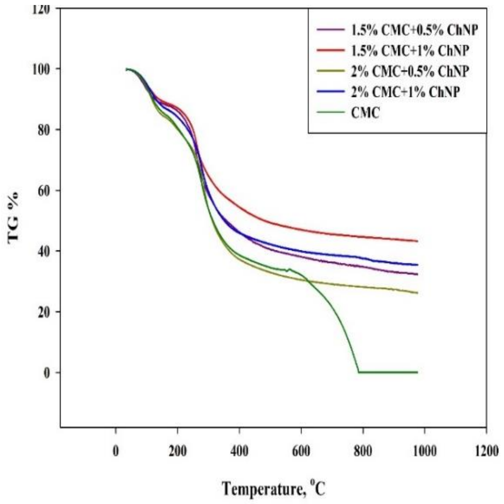

Figure 5: The thermogravimetric curves of CMC and CMC/ChNPs blends

3.2.5. Colour assessments

It's clear from table 2 to 5 that all printed samples using 1.5% CMC as thickener poses colour strength value ranging from 2.01 to 3.93 for printed jute overall both dyes and for printed paper colour strength ranging from 0.26 to 0.31 overall both dyes, these K/S values less than that obtained while using CMC/ChNPs blends as thickeners increase the rate of dye uptake of jute samples in the case of Hibiscus rosa-sinensis and Curcumin. Chitosan nanoparticles act as a mordant as metal salt due to amino groups which get protonated and offer the sites for attachment of the natural dye which is anionic by nature. Previous research used 10 to 20 percent of chitosan in the printing paste as mordant to enhance the printability of cotton fabric with Catechu, turmeric, and marigold natural dyes [39]. The used chitosan nanoparticles in this study has decrease the concertation to 0.5 and 1%. Addition of urea to printing paste slightly increase the K/S values where, urea accelerates the migration of dye from the thickener film into the fibres but overall, it can be illuminated without effect on the net result. the highest K/S value is obtained with using 2% CMC and 1% ChNPs. Table 6 state the fastness properties of printed jute fabric using the new thickeners blends with the used natural dyes. the results indicate the expected improvement in the all fastness tested compared with that produce by using CMC only.

In addition, Figure 6 shows pictures for Jute printed with curcumin using CMC as thickener (a), Jute printed with curcumin using 2% CMC+1% ChNPs (b), White paper printed with Hibiscus rosa-sinensis using CMC thickener (c) and White paper printed with Hibiscus rosa-sinensis dye using the new thickener blend 2% CMC+1% ChNPs (d)

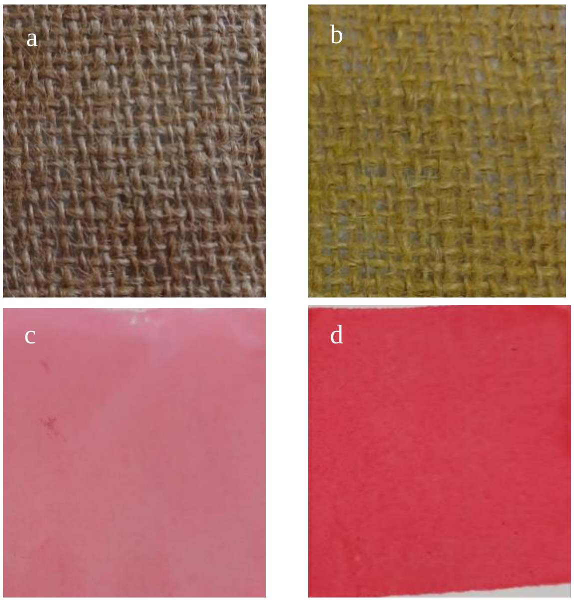

Figure 6: Pictures for printed jute and paper using CMC as thickener (a and c) and 2% CMC+1% ChNPs as thickener (b and d)

3.2.6. Antibacterial

Chitosan (Ch) demonstrates cationic characteristics in acidic conditions as a result of NH_2 being converted to NH_3^+. Because of its cationic properties, chitosan may penetrate bacterial cell walls and effectively break them down. Additionally, Ch acts on fungi in the same way [38]. Compared to chitosan, chitosan nanoparticles exhibit greater antibacterial efficacy.

Antimicrobial efficiency for jute printed samples were determined for all different prepared thickeners against two different types of gram +ve (*Staphylococcus aureus*) bacteria, gram –ve bacteria (*Escherichia coli*) and fungi (*Candida albicans*). From tables 7 and 8 we can notice the improvement more than 96% reduction for all tested samples against all tested microbes. Sample with composition 2.0% CMC+1.0% ChNPs poses the highest reduction % it could be safely confirmed that the prints have very good antimicrobial activity this could be due to that the ChNPs' nanostructure defeats the structures of bacteria and fungi. Similar result was reported by Mosaad et al. (2022) [40].they found that ChNPs had a stronger effect on both gram-positive and gram-negative bacteria.

Table 2: colour parameters of printed jute fabric using the CMC and its blends with ChNPs as thickener in the printing paste using Curcuma longa

Samples	without urea				Using urea			
	K/S value	L*	a*	b*	K/S value	L*	a*	b*
Blank 1.50 %CMC	3.72 ± 0.05	40.62	2.98	32.44	3.93	39.67	3.11	32.21
1.5% CMC+0.5% ChNPs	12.96 ± 0.17	43.32	4.01	34.32	12.83	13.1	3.88	33.01
1.5 %CMC+1.0% ChNPs	13.2 ± 0.17	49.91	4.08	37.36	13.9	49.71	3.95	37.36
2.0 %CMC+0.5% ChNPs	15.89 ± 0.21	49.33	4.54	39.13	15.89	49.91	3.73	37.60
2.0 %CMC+1.0% ChNPs	15.3 ± 0.13	49.73	3.69	38.35	15.92	49.28	3.24	38.58

Table 3: colour parameters of printed white papers using the CMC and its blends with (ChNP) as thinker in the printing paste using Curcuma longa

Samples	without urea				Using urea			
	K/S value	L*	a*	b*	K/S value	L*	a*	b*
Blank 1.50 %CMC	0.3 ± 0	86.45	-0.44	13.10	0.31	86.11	-0.41	11.10
1.5% CMC+0.5% ChNPs	1.2 ± 0.02	83.18	-2.13	32.70	1.1	84.02	-1.43	26.24
1.5 %CMC+1.0% ChNPs	1.95 ± 0.03	84.97	-0.89	22.71	1.87	83.91	-2.16	26.30
2.0 %CMC+0.5% ChNPs	1.93 ± 0.03	81.02	1.86	37.59	1.69	81.67	0.22	37.91
2.0 %CMC+1.0% ChNPs	2.4 ± 0.02	79.71	2.78	38.35	2.29	80.51	-2.18	35.40

Table 4: colour parameters of printed jute fabric using the CMC and its blends with (ChNPs) as thinker in the printing paste using Hibiscus rosa-sinensis

Samples	without urea				Using urea			
	K/S value	L*	a*	b*	K/S value	L*	a*	b*
Blank 1.50 %CMC	2.01 ± 0.03	52.98	5.48	17.16	2.12	51.76	3.89	16.8
1.5% CMC+0.5% ChNPs	10.86 ± 0.14	39.62	28.70	12.67	11.01	39.89	28.62	12.56
1.5 %CMC+1.0% ChNPs	13.92 ± 0.18	36.38	29.82	13.19	13.07	39.85	26.48	11.95
2.0 %CMC+0.5% ChNPs	12.37 ± 0.16	36.71	33.64	13.93	12.43	33.92	33.61	14.19
2.0 %CMC+1.0% ChNPs	18.13 ± 0.16	35.32	33.49	14,06	18.44	36.76	34.05	13.88

Table 5: colour parameters of printed white paper using the CMC and its blends with ChNPs as thinker in the printing paste using Hibiscus rosa-sinensis

Samples	without urea				Using urea			
	K/S value	L*	a*	b*	K/S value	L*	a*	b*
Blank 1.50 %CMC	0.26 ± 0.02	68.21	-2.18	0.05	0.31	67.91	-2.01	0.08
1.5% CMC+0.5% ChNPs	3.12 ± 0.04	58.67	47.96	9.48	3.22	58.51	49.01	9.11
1.5 %CMC+1.0% ChNPs	4.64 ± 0.06	60.62	47.10	7.82	4.00	54.82	47.30	10.09
2.0 %CMC+0.5% ChNPs	4.04 ± 0.05	55.76	50.31	12.64	4.30	55.51	51.67	12.76
2.0 %CMC+1.0% ChNPs	5.93 ± 0.05	56.28	50.65	12.84	5.07	52.83	50.35	13.77

Table 6: Fastness properties of printed jute fabrics using the new thickeners without urea

Printed jute Samples with	Rubbing fastness		Washing fastness		Perspiration fastness				Light fastness
	Dry	Wet	Alt.	St.	Acid		Alkaline		
					Alt.	St.	Alt	St.	
Curcuma longa									
Blank 1.50 %CMC	3	2-3	3	3		3	3	2-3	3-4
1.5% CMC+0.5% ChNPs	4	3-4	4-5	4	4-5	4	4	4-5	5-6
1.5 %CMC+1.0% ChNPs	4	4	4-5	4	4-5	4	4	4-5	5-6
2.0 %CMC+0.5% ChNPs	4	4-5	4-5	4	4-5	4	4	4-5	5-6
2.0 %CMC+1.0% ChNPs	4-5	4-5	4-5	4	4-5	4	4	4-5	5-6
Hibiscus rosa-sinensis									
Blank 1.50 %CMC	3	2-3	3	3	3-4	3	3	2-3	3-4
1.5% CMC+0.5% ChNPs	4-5	4	4	3-4	4	4	4-5	4	5
1.5 %CMC+1.0% ChNPs	4-5	4-5	4	4-5	4-5	4	4	3-4	5-6
2.0 %CMC+0.5% ChNPs	4-5	4	4	4-5	4	4-5	3-4	4	5
2.0 %CMC+1.0% ChNPs	4	4-5	4	4-5	4-5	4-5	4-5	4	5-6

Table 7: CFU reduction (%) of bacterial strains after incubation applying the Curcuma longa on jute printed samples

Printed samples with	Test strain		
	Escherichia coli (-ve)	*Staphylococcus aureus (+ve)*	*Candida albicans*
Blank 1.50 %CMC	55.07 ± 0.72	57.24 ± 0.75	52.32 ± 0.69
1.5% CMC+0.5% ChNPs	96.6 ± 1.27	96.09 ± 1.26	96.91 ± 1.27
1.5 %CMC+1.0% ChNPs	98.62 ± 1.29	97.91 ± 1.28	98.7 ± 1.29
2.0 %CMC+0.5% ChNPs	98.21 ± 1.29	97.12 ± 1.27	96.92 ± 1.27
2.0 %CMC+1.0% ChNPs	99.29 ± 0.87	99.52 ± 0.81	98.59 ± 0.86

Table 8: CFU reduction (%) of bacterial strains after incubation applying the Hibiscus rosa-sinensis on jute printed samples

Printed samples using	Test strain		
	Escherichia coli (-ve)	Staphylococcus aureus (+ve)	Candida albicans
Blank 1.50 %CMC	58.01 ± 1.37	48.44 ± 1.14	52.63 ± 1.24
1.5% CMC+0.5% ChNPs	97.96 ± 2.31	97.75 ± 2.31	96.34 ± 2.28
1.5 %CMC+1.0% ChNPs	99.85 ± 2.36	98.42 ± 2.33	96.18 ± 2.27
2.0 %CMC+0.5% ChNPs	99.68 ± 2.36	97.28 ± 2.3	97.91 ± 2.31
2.0 %CMC+1.0% ChNPs	99.85 ± 2.36	98.42 ± 2.33	97.19 ± 2.3

4. Conclusion

Combination of chitosan nanoparticles and carboxymethyl cellulose blends were used as thickeners to prepare edible printing paste. The prepared thickeners characterized by TGA, FTIR and rheological properties. ChNPs acted as environmentally acceptable mordant allowed for the effective printing of jute and paper with powdered natural colours. The novel thickeners were thermally stable and exhibited distinct rheological characteristics with non-Newtonian pseudoplastic behavior. They also have two effects: first, they created antimicrobial printed samples (jute and paper) appropriate for food packaging; second, the printed samples have increased colour strength and associated properties. 2%CMC+1% ChNPs sample has the highest antimicrobial activity and colour strength.

5. Fund

The authors have no fund

6. Conflict of interest

The authors have no conflict of interest

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8. References

[1]. M.A. Pascall, K. DeAngelo, J. Richards, M.B.J.F. Arensberg, Role and importance of functional food packaging in specialized products for vulnerable populations: Implications for innovation and policy development for sustainability, 11(19) (2022) 3043.

[2]. M. Alamri, A.A. Qasem, A.A. Mohamed, S. Hussain, M.A. Ibraheem, G. Shamlan, H.A. Alqah, A.S.J.S.J.o.B.S. Qasha, Food packaging's materials: A food safety perspective, 28(8) (2021) 4490-4499.

[3]. E. Galbiati, L. Jacxsens, B.J.F.A. De Meulenaer, C.P. A, Hazard prioritisation of substances in printing inks and adhesives applied to plastic food packaging, 38(9) (2021) 1608-1626.

[4]. N.A. Al-Tayyar, A.M. Youssef, R.J.F.c. Al-Hindi, Antimicrobial food packaging based on sustainable bio-based materials for reducing foodborne pathogens: A review, 310 (2020) 125915.

[5]. M. Asgher, S.A. Qamar, M. Bilal, H.M.J.F.R.I. Iqbal, Bio-based active food packaging materials: Sustainable alternative to conventional petrochemical-based packaging materials, 137 (2020) 109625.

[6]. J. Wang, M. Euring, K. Ostendorf, K.J.J.o.B. Zhang, Bioproducts, Biobased materials for food packaging, 7(1) (2022) 1-13.

[7]. Y. Pan, M. Farmahini-Farahani, P. O'Hearn, H. Xiao, H.J.J.B.B. Ocampo, An overview of bio-based polymers for packaging materials, 1(3) (2016) 106-113.

[8]. F. Alabi, L. Lajide, O. Ajayi, A. Adebayo, S. Emmanuel, A.J.A.J.o.P. Fadeyi, A. Chemistry, Synthesis and characterization of carboxymethyl cellulose from musa paradisiaca and tithonia diversifolia, 14(1) (2020) 9-23.

[9]. C.M. Obele, M.E. Ibenta, J.L. Chukwuneke, S.C.J.C. Nwanonenyi, Carboxymethyl cellulose and cellulose nanocrystals from cassava stem as thickeners in reactive printing of cotton, 28 (2021) 2615-2633.

[10]. R. Ramakrishnan, J.T. Kim, S. Roy, A.J.I.J.o.B.M. Jayakumar, Recent advances in carboxymethyl cellulose-based active and intelligent packaging materials: A comprehensive review, (2024) 129194.

[11]. A.G. Hassabo, N.A. Mohamed, N.A. Abd El-Salam, N.Z. Gouda, H. Othman, Application of modified xanthan as thickener in the printing of natural and synthetic fabrics, Journal of Textiles, Coloration and Polymer Science 20(1) (2023) 41-56.

[12]. N. Ahmed, A. Shahin, H. Othman, A.G. Hassabo, Neem tree extracts used in textile industries, Egy. J. Chem. 67(13) (2024) 159-169.

[13]. E. Abdou, H. El-Hennawi, K.J.J.o.C. Ahmed, Preparation of novel chitosan-starch blends as thickening agent and their application in textile printing, 2013 (2013).

[14]. A.A. Shahin, S.A. Mahmoud, H. El-Hennawi, A. Ragheb, Enhancement of dyeability and antibacterial characteristics of silk fabrics using chitosan nano-particles, Egy. J. Chem. 63(9) (2020) 3199 -3208.

[15]. L.-J. Yin, B.-S. Chu, I. Kobayashi, M. Nakajima, Performance of selected emulsifiers and their combinations in the preparation of β-carotene nanodispersions, Food Hydrocolloids 23(6) (2009) 1617-1622.
[16]. P. Calvo, C. Remunan-Lopez, J.L. Vila-Jato, M.J.J.o.a.p.s. Alonso, Novel hydrophilic chitosan-polyethylene oxide nanoparticles as protein carriers, 63(1) (1997) 125-132.
[17]. R.M. Mosaad, M.H. Alhalafi, E.-A.M. Emam, M.A. Ibrahim, H. Ibrahim, Enhancement of antimicrobial and dyeing properties of cellulosic fabrics via chitosan nanoparticles, Polymers 14(19) (2022) 4211.
[18]. H. Ibrahim, M. El-Bisi, G. Taha, E. El-Alfy, Chitosan nanoparticles loaded antibiotics as drug delivery biomaterial, Journal of Applied Pharmaceutical Science (2015) 085-090.
[19]. E.S. Abdou, H.M. El-Hennawi, K.A. Ahmed, Application of multi-function thickener from chitosan/ starch blend in textile pigment printing, International Journal of ChemTech Research 9(12) (2016) 393-403.
[20]. H. El-Hennawi, N.A. Hanafy, Eco-friendly dyeing of natural-mordanted viscose fabrics with natural turmeric dye, J. Text. Color. Polym. Sci. 19(2) (2022) 349-355.
[21]. AATCC Test Method (8-2016), Colorfastness to crocking, crockmeter method, Technical Manual Method American Association of Textile Chemists and Colorists, 2018, pp. 17-19.
[22]. AATCC Test Method (15-2013), Colour fastness to perspiration, Technical Manual Method American Association of Textile Chemists and Colorists, 2017, pp. 30-32.
[23]. AATCC Test Method (16.1-2014), Colour fastness to light: Outdoor, Technical Manual Method American Association of Textile Chemists and Colorists, 2015, pp. 33-48.
[24]. AATCC Test Method (61-2013), Color fastness to laundering: Accelerated, Technical Manual Method American Association of Textile Chemists and Colorists, 2017, p. 108.
[25]. E.A. Ahmed, G. Elgemeie, K.A.-E.J.P. Ahmed, R. Technology, Microwave-assisted synthesis of disazothiazolone dyes as antibacterial agents and their application in polyester printing, 51(1) (2021) 1-5.
[26]. E.C. Svensson, A. Madar, C.D. Campbell, Y. He, M. Sultan, M.L. Healey, H. Xu, K. D'Aco, A. Fernandez, C.J.J.c. Wache-Mainier, Tet2-driven clonal hematopoiesis and response to canakinumab: An exploratory analysis of the cantos randomized clinical trial, 7(5) (2022) 521-528.
[27]. AATCC Test Method (147-2016), Antibacterial activity assessment of textile materials: Parallel streak, Technical Manual Method American Association of Textile Chemists and Colorists, 2017, pp. 275-276.
[28]. AATCC Test Method (100-2019), Assessment of antimicrobial finishes on textile materials, Technical Manual Method American Association of Textile Chemists and Colorists, 2019.
[29]. S.Y. Tang, S. Manickam, T.K. Wei, B. Nashiru, Formulation development and optimization of a novel cremophore el-based nanoemulsion using ultrasound cavitation, Ultrason. Sonochem. 19(2) (2012) 330-345.
[30]. F. Saad, A.G. Hassabo, H.A. Othman, M.M. Mosaad, A.L. Mohamed, Improving the performance of flax seed gum using metal oxides for using as a thickening agent in printing paste of different textile fabrics, Egy. J. Chem. 64(9) (2021) 4937 - 4954.
[31]. F. Saad, A.L. Mohamed, M. Mosaad, H.A. Othman, A.G. Hassabo, Enhancing the rheological properties of aloe vera polysaccharide gel for use as an eco-friendly thickening agent in textile printing paste, Carbo. Polym. Technol. App. 2 (2021) 100132.
[32]. F. Saad, A. Hassabo, H. Othman, M.M. Mosaad, A.L. Mohamed, A valuable observation on thickeners for valuable utilisation in the printing of different textile fabrics, Egy. J. Chem. 65(4) (2022) 431 – 448.
[33]. S.A. Ebrahim, A.G. Hassabo, H. Othman, Natural thickener in textile printing (a mini review), Journal of Textiles, Coloration and Polymer Science 18(1) (2021) 55-64.
[34]. S.A. Ebrahim, H.A. Othman, M.M. Mosaad, A.G. Hassabo, A valuable observation on pectin as an eco-friendly material for valuable utilisation in textile industry, Egy. J. Chem. 65(4) (2022) 555 – 568.
[35]. S.A. Ebrahim, H.A. Othman, M.M. Mosaad, A.G. Hassabo, Eco-friendly natural thickener (pectin) extracted from fruit peels for valuable utilization in textile printing as a thickening agent, Textiles 3(1) (2023) 26-49.
[36]. M.F. Queiroz, K.R. Teodosio Melo, D.A. Sabry, G.L. Sassaki, H.A.O. Rocha, Does the use of chitosan contribute to oxalate kidney stone formation?, Marine drugs 13(1) (2014) 141-158.
[37]. X. He, Cr (vi) removal from aqueous solution by chitosan/carboxylmethyl cellulose/silica hybrid membrane, World Journal of Engineering and Technology 3(03) (2015) 234.
[38]. M.R. de Moura, M.V. Lorevice, L.H. Mattoso, V. Zucolotto, Highly stable, edible cellulose films incorporating chitosan nanoparticles, Journal of Food Science 76(2) (2011) N25-N29.
[39]. M. Teli, J. Sheikh, P. Shastrakar, Exploratory investigation of chitosan as mordant for eco-friendly antibacterial printing of cotton with natural dyes, Journal of Textiles 2013(1) (2013) 320510.
[40]. A. Jiang, R. Patel, B. Padhan, S. Palimkar, P. Galgali, A. Adhikari, I. Varga, M. Patel, Chitosan based biodegradable composite for antibacterial food packaging application, Polymers 15(10) (2023) 2235.