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Investigation of physical, antioxidant and structural properties of edible films based on carboxymethyl cellulose containing okra mucilage and black cumin seed (*Nigella sativa*) oil

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ABSTRACT

The purpose of this work was to establish the biocomposite films made from carboxymethyl cellulose (CMC) (1% w/v), okra mucilage (OM) (1-3% w/v) and black cumin seed oil (BSO) (0-0.5% v/v). The impact of OM and BSO concentrations on the properties of biocomposite films including the thickness, density, water vapor permeability, water solubility, antioxidant activity, color and microstructure properties was evaluated by response surface methodology. The results demonstrated that the water solubility and water vapor barrier properties of fabricated films increased at higher OM concentrations, whereas by increasing BSO concentrations, these parameters decreased. The parameters of redness (a*), total color difference value (ΔE) and yellowness index (YI) have incremental trend after incorporating higher OM and BSO concentrations, while the lightness (L*) and whiteness index (WI) displayed a decline. Antioxidant activity (DPPH) and total phenolic content (TPC) of films increased by increasing okra mucilage concentrations, on the contrary the incorporation of BSO at higher concentrations caused a diminish in antioxidant activity and total phenolic content. Meanwhile, the AFM images indicated that the roughness parameters (Ra, Rq) increased by increasing okra mucilage and black seed oil concentrations. According to the analysis of variance, the proposed model was significant for the all parameters ($p < 0.05$), except for the yellowness parameter (b*), for which none of the models were statistically significant ($p > 0.05$). Based on optimization, the optimal film was obtained with a proportion of 1% OM and 0.5% BSO, with a desirability score of 0.75. The recorded SEM images of the microstructural cross-sectional films indicated that the porosity and discontinuity increased by increasing okra mucilage amount, in contrast higher concentrations of black seed oil led to a decrease in porosity of cross-sectional films. Additionally, FTIR analysis confirmed the occurrence of new intermolecular interactions in the film matrix.

1- Introduction

In recent years, biodegradable packaging has attracted considerable attention as an alternative to plastic packaging. Environmental pollution caused by plastics and the problem of waste recycling is one of the biggest crises in today's societies. According to statistical reports, more than 340 million tons of plastic waste are produced globally, of which about 46% originates from the packaging sector. Packaging materials are largely non-recyclable, and approximately 95% of plastic packaging materials made from polyolefins and polyethylene terephthalate are not recycled; after single use, they end up in landfills, resulting in an annual loss of 80 to 120 billion dollars to the global economy [1, 2]. Due to reasons such as environmental issues, the dependence of plastic materials on petroleum products, and customer demand for safe and healthy food, researchers are seeking to find renewable biopolymers and their application in food packaging. Compounds such as carbohydrates, proteins, lipids, or a mixture of these are used in the preparation of edible films. Edible films prevent the depletion of petroleum resources while not causing environmental pollution [3, 4]. Furthermore, by incorporating active compounds such as natural antioxidants into edible films, the quality and shelf life of packaged food are improved [5, 6].

Carboxymethyl cellulose (CMC), as one of the important derivatives of cellulose, has a number of hydroxyl and carboxyl groups. Due to its polymeric structure, high molecular weight, high viscosity, good film-forming ability, low price, and easy availability, it has been widely used in the formulation of edible films. Also, to improve the mechanical and barrier properties of films, CMC has been combined with other biopolymers such as cassava starch, rice starch, and chitosan [7]. The okra plant, scientifically known as *Hibiscus esculentus* L., belongs to the Malvaceae family. Okra has a thick, viscous substance present in the fruit pod, which is called mucilage. The main compounds of okra mucilage are polysaccharides, including rhamnose, galactose, and galacturonic acid. Polysaccharides extracted from okra are used as texture improvers, thickeners, and emulsion stabilizers. Okra mucilage has antioxidant compounds, including flavonoids and quercetin [8, 9]. Black cumin, scientifically known as *Nigella sativa* L., is a plant from the Ranunculaceae family. Black cumin has been widely used in traditional medicine, and many studies have proven its therapeutic effects for many diseases. Various active compounds from black cumin, including thymoquinone, thymohydroquinone, carvacrol, and limonene, have been identified. Furthermore, the most abundant saturated fatty acids in black seed oil include palmitic, stearic, and myristic acid, while the main unsaturated fatty acids are linoleic, oleic, and alpha-linolenic acid [10, 11].

Despite numerous investigations to develop the application of biodegradable polymers in packaging, the application of biodegradable films is limited due to issues with oxygen permeability, water vapor, and mechanical properties. Extensive research is necessary regarding the methods of forming these films and improving their properties for the potential use of edible films in food packaging [2]. Film production requires at least one polymeric compound capable of creating a network structure with sufficient strength and coherence. Mixing two or more biopolymers is a common method for improving the properties of biocomposite films [2, 4]. Regarding the combination of oil with polysaccharide-based films, research has been conducted, including CMC-based films containing oleic acid [12], chitosan-based films containing various animal and vegetable oils by Akyuz et al. [13], and galactomannan films mixed with corn oil by Cerqueira et al. [14]. Research has also been conducted on adding polysaccharides to CMC-based edible films, such as CMC-based edible film enriched with chickpea hull polysaccharides and CMC-based film containing coffee pulp polysaccharides, studied by Akhtar et al. [15] and Ballesteros et al. [16], respectively. Also, in recent years, seed gums and plant gums have attracted the attention of researchers in the preparation of edible films, including plantain seed mucilage and basil seed mucilage, which were used to prepare edible films by Niknam et al. [17] and Mohammad Amini et al. [18], respectively.

Okra mucilage is a rich source of polysaccharides, and due to its hydrophilic nature, it has the ability to retain moisture, and in this respect, it can be considered a natural plasticizer. Therefore, the application of okra mucilage in the preparation of films, in addition to enhancing antioxidant properties, can play a role in improving the flexibility property of the film [19, 20]. On the other hand, polysaccharide films, due to their hydrophilic nature, do not have desirable barrier properties against water vapor. In these conditions, by adding lipids to polysaccharide films, the barrier properties against moisture can be improved due to the hydrophobic nature of lipids [3, 4, 6]. As a result, combining these materials helps establish a balance between hydrophobicity and hydrophilicity in the film matrix, thereby overlapping the weaknesses of each material and improving the properties of the prepared film [4]. Despite research on the combination of CMC with polysaccharides and oils, the effect of incorporating black seed oil and okra mucilage on the properties of CMC-based films has not been investigated. The aim of this research is to introduce a new edible film developed based on CMC containing okra mucilage and black seed oil, where the effect of different concentrations of okra mucilage and black seed oil on the physical, color, antioxidant, and structural properties of the prepared films was studied.

2-Materials and Methods

2.1. Materials

Fresh okra and black seeds were purchased from the local market (Mahabad, Iran). Carboxymethyl cellulose (300-1500 cp), Tween 80, choline chloride, and 2,2-diphenyl-1-picrylhydrazyl (DPPH) were purchased from Sigma-Aldrich, Germany. Other compounds, including glycerol, sodium carbonate, potassium sulfate, and calcium nitrate, were prepared from Merck, Germany.

2.2. Methods

2.2.1. Extraction of Okra Mucilage

Extraction of okra mucilage was performed according to the method of Lee et al. [21]. First, the seeds were separated from fresh okra, and then the pods were immersed in distilled water for 12 hours. The insoluble part was filtered through a cloth filter, and the obtained extract was dried in an incubator (Memmert, Germany) at 55°C and ground.

2.2.2. Extraction of Black Cumin Seed Oil

Extraction of black seed oil was performed using a cold screw press (Oil Press PR 500, Germany) at 45°C. After filtration, it was stored in dark glass bottles at 4°C until use.

2.2.3. Preparation of Films

Films were prepared by the casting method according to the method of Taqi et al. [22]. Initially, okra mucilage powder (1-3% w/v) and carboxymethyl cellulose (1% w/v) were dissolved separately in 30 mL of distilled water in a beaker using a magnetic stirrer at 60°C for 30 minutes. The prepared solutions were mixed together, and 30% glycerol (w/w of CMC) was added, and stirring was continued by a magnetic stirrer for 20 minutes. Then, black seed oil (0-0.5% v/v) was mixed with 40 mL of distilled water containing 0.1% Tween 80 (based on oil weight) and homogenized for 2 minutes at 5000 rpm using a homogenizer (IKA T25 Digital Ultra Turrax, Germany) and added to the previously prepared solution. The prepared emulsion solutions were poured into plastic containers and placed in an oven at 50°C to dry. After drying, the film samples were separated from the container surfaces and conditioned at 25°C in a desiccator containing a saturated solution of calcium nitrate (50% relative humidity).

2.3. Properties of the Prepared Biocomposite Film

2.3.1. Thickness and Density

The thickness of each film sample was measured using a manual micrometer (Alton M820-25, China) with an accuracy of 0.01 mm at 10 different points, and the average was reported. To determine density, film samples were cut into dimensions of 2 cm x 2 cm, and density was calculated according to the following formula:

$$\rho=\mathrm{mA}\times\mathrm{d}\rho=A\times dm$$

Where A is the surface area of the film (4 cm^2), d is the film thickness (cm), and m is the film mass (g), resulting in ρ, the film density (g/cm^3).

2.3.2. Solubility

Film solubility was determined using the method presented by Riaz et al. [23]. Film samples of 2 x 2 cm^2 were dried at 105°C for 24 hours to reach constant weight, and their initial weight was measured (M1). The dried film samples were placed in beakers containing 50 mL of distilled water for 24 hours. After this period, the film pieces were removed from the beaker and dried in an oven at 105°C for 24 hours (M2). The solubility percentage of the films was calculated using the following equation:

Solubility (%)=W1−W2W1×100Solubility (%)=$W_1W_1-W_2\times100$

Where W1 and W2 are the initial and final weights of the films in grams, respectively.

2.3.3. Water Vapor Permeability (WVP)

The water vapor permeability of the films was determined according to the method of Ghanbarzadeh and Almasi [12]. In this test, a glass cell with a diameter of 1 cm and a depth of 4 cm was used. The cells contained 3 grams of silica gel to create 0% relative humidity. Then, films were cut circularly to the size of the cell opening and, after being placed on the cell opening, were sealed with Parafilm. The initial weight of the cells was measured with an accuracy of 0.0001 g, and then they were placed in a desiccator containing potassium sulfate to provide 97% relative humidity at a temperature of 25°C. Weight changes of the cells were measured at specific intervals for 7 days. The curve of weight gain of the cells over time was plotted, and the slope of the line was determined by linear regression. Water vapor permeability was calculated by the following formula:

WVP=Δm×xΔt×A×ΔpWVP=$\Delta t\times A\times\Delta p\Delta m\times x$

Where Δm is the weight gain of the cell (grams), x is the film thickness (mm), Δt is the time duration of water vapor transmission (seconds), A is the surface area of the film exposed to water vapor (m²), Δp is the water vapor pressure difference across the film (at 25°C, the partial pressure difference between the inside and outside of the cell is 3073.93 Pa), resulting in WVP (grams of water per meter per Pascal per second).

2.3.4. Measurement of Color Properties

Samples were evaluated using a HunterLab colorimeter (Minolta, CR-410, Japan). Color was determined based on parameters L (lightness-darkness), a (redness-greenness), b (yellowness-blueness), and the total color difference (ΔE), whiteness index (WI), and yellowness index (YI) were calculated from the following relationships:

ΔE=(L∗−L)2+(a∗−a)2+(b∗−b)2$\Delta E=(L_*-L)_2+(a_*-a)_2+(b_*-b)_2$
WI=100−(100−L)2+a2+b2$WI=100-(100-L)_2+a_2+b_2$YI=(142.86×b)/L$YI=(142.86\times b)/L$

Where L, a, b are the color factors of the sample and L, *a*, b* are the color factors of the standard white plate.

2.3.5. Evaluation of Free Radical Scavenging Activity (DPPH)

The antiradical property was assessed using the discoloration method of the purple solution of 2,2-diphenyl-1-picrylhydrazyl (DPPH) in methanol. According to the method of Shojaee-Aliabadi et al. [24], initially, 25 mg of film was dissolved in 0.1 mL of distilled water, then 3.9 mL of DPPH solution (0.1 mM in methanol) was added to 3.9 mL of the film extract solution. The samples were kept in a dark place at ambient temperature for 60 minutes. The absorbance was measured by a spectrophotometer (Perkin-Elmer, model Lambda 25, USA) against pure methanol at a wavelength of 517 nm, and the percentage of radical scavenging activity was calculated by the following formula:

Radical scavenging activity (%)=Areference−Asa mpleAreference×100Radical scavenging activity (%)=Areference$A$reference−Asample×100

Where Areference and Asample represent the absorbance of the DPPH solution without film extract and the absorbance of the DPPH solution containing film extract, respectively.

2.3.6. Total Phenolic Content (TPC)

The total phenolic content of the films was measured using the method of Shojaee-Aliabadi et al. [24]. 25 mg of film was placed in 3 mL of deionized distilled water and stirred gently for 5 minutes. Then, 0.1 mL of the film extract was poured into a test tube with 7 mL of deionized distilled water, followed by the addition of 0.5 mL of Folin-Ciocalteu reagent and mixing. After 8 minutes, 0.9 mL of saturated sodium carbonate solution (7% w/v) and 1.5 mL of deionized distilled water were added to the tubes. The tubes were kept at room temperature for 2 hours, and then the absorbance was read at a wavelength of 765 nm using a spectrophotometer (Perkin-Elmer, model Lambda 25, USA). The total phenol content was obtained using the following formula:

T=(C×V)/M$T=(C\times V)/M$

Where T is the total phenol content (mg gallic acid equivalent/g film weight), C is the gallic acid concentration obtained from the standard curve (mg/mL), V is the extract volume (mL), and M is the film weight (g).

2.3.7. Study of Microstructure with Scanning Electron Microscopy (SEM)

A scanning electron microscope (SEM, MIRA3 FEG-Tescan, Czech Republic) was used for the films. The film samples were coated with a layer of gold, and then SEM images were recorded from the surface and cross-section of the film samples.

2.3.8. Atomic Force Microscopy (AFM)

The surface morphology of the prepared biocomposite films was studied using an atomic force microscope (Nanosurf Mobile S, Switzerland). Quantitative parameters for determining surface roughness, including average roughness (Ra) and root mean square roughness (Rq), were calculated from the surface profiles of the film samples.

2.3.9. Fourier Transform Infrared Spectroscopy (FTIR)

The study of structural interactions of the prepared films was carried out using a Fourier transform infrared spectrometer (Tensor 27, Bruker, Germany). The film samples were compressed inside KBr pellets, and the pellets containing the sample were placed in the device cell. Infrared spectroscopy was performed in the range of 400-4000 cm^{-1}.

2.4. Statistical Analysis

Statistical analysis of data in this research was performed using Design Expert software version 13 and Response Surface Methodology (RSM) based on a Central Composite Design (CCD). As shown in Table 1, the independent variables included okra mucilage (OM, X1 = 1-3% w/v) and black seed oil (BSO, X2 = 0-0.5% v/v), and 13 combinations were obtained according to the central composite design. The effect of independent variables on the responses, including thickness, density, solubility, water vapor permeability, color properties, antioxidant activity, total phenolic content, and roughness parameters, was investigated using analysis of variance at a significance level of 0.95.

3. Results and Discussion

3.1. Thickness and Density

Based on Table 1, the film composed of 1.29% mucilage and 0.07% oil had the lowest thickness and density. According to Figure 1, with increasing concentrations of okra mucilage and black seed oil, the thickness and density of the films increased, and based on Table 2, the proposed model for thickness and density parameters was linear ($p < 0.05$). The increase in film thickness could be due to the increase in solid content in the film formulation [22, 25]. In previous studies, the addition of marjoram essential oil to CMC-based edible films by Hasheminya et al. [26] and the incorporation of green tea extract into pomelo peel-based films by Wu et al. [27] resulted in increased film thickness, which is consistent with the findings of this study. In a study conducted by Aydogdu et al. [28], density increased with increasing orange oil concentration in guar gum-based films, and they stated that the increase in film density could be due to the increased molecular weight of the compounds, increased solid content, and reactions between polymer chains leading to a more compact molecular structure.

3.2. Solubility

Solubility is considered an indicator of the hydrophilicity of films, and water resistance is an important factor in the preparation of commercial edible films [29]. Based on the data in Table 1, the film formulated with 1.29% okra mucilage and 0.43% black seed oil had the lowest solubility. According to Figure 1, with increasing concentration of okra mucilage in the film formulation, solubility increased, while with increasing oil concentration, a decrease in solubility was observed, and based on Table 2, a linear model was proposed for solubility ($p < 0.05$). Hydrophilic groups of polysaccharides and phenolic compounds react easily with water molecules, and with increasing okra mucilage concentration, the capacity of the film matrix to bond with water improved, hence the increase in solubility occurred. According to the report by Nawab et al. [30, 31], an increase in the solubility of mango starch-based films with the addition of guar and xanthan gums was observed. Similar results have been obtained by other researchers [15, 23]. The decrease in solubility with increasing amount of black seed oil in the film formulation is related to the reaction between the hydroxyl groups of the biopolymer and the oil, leading to reduced accessibility of hydroxyl groups and the creation of a hydrophobic nature in the film matrix [18, 24]. In a study by Akyüz et al. [13], they reported that the addition of olive oil to chitosan-based films led to a decrease in solubility, which was attributed to the dense and continuous structure of chitosan and olive oil. The results of this research are consistent with studies conducted by Cerqueira et al. [14], Galus and Kadzińska [29], and Ma et al. [32].

3.3. Water Vapor Permeability (WVP)

Based on Table 1, the lowest water vapor permeability was attributed to the film containing 1.29% okra mucilage and 0.43% black seed oil (5.22×10^{-8} g.mm/Pa.h.cm²), and the highest water vapor permeability was assigned to the film produced with a concentration of 2.71% okra mucilage and 0.07% black seed oil (8.96×10^{-8} g.mm/Pa.h.cm²). Figure 1 shows that with increasing okra mucilage concentration, water vapor permeability increased, whereas with increasing black seed oil concentration, a decrease in water vapor permeability occurred, and according to Table 2, a linear model was proposed for this parameter, which was significant ($p < 0.05$). Based on previous studies, the ratio of crystalline to amorphous regions and the hydrophilicity to hydrophobicity ratio of films are effective in water vapor transmission [33]. Increasing the concentration of okra mucilage in the film formulation caused the hydrophilic nature to dominate, increasing the number of hydroxyl groups and greater absorption of water molecules into the film structure, which resulted in increased mobility and fluidity of side chains and the creation of a less dense structure, thus water vapor transmission increased [31]. Also, in the prepared biocomposite films, with increasing okra mucilage concentration, amorphous regions increased, and a non-dense, disordered structure emerged, which facilitated the penetration of water molecules into the film, therefore WVP increased [33]. Jang et al. [34] reported that in biocomposite films prepared based on rapeseed protein-gelatin containing grapefruit seed extract, WVP increased with increasing extract concentration, and they attributed the cause to a decrease in intermolecular interactions between film components, leading to increased porosity and a weak structure in the films. Also, Cerqueira et al. [35] stated that extracts of seeds and plants containing phenolic acids and flavonoids are hydrophilic compounds, and these compounds lead to increased hydrophilic properties of films, resulting in increased WVP. Similar results have been reported by Kannani and Rhim [25] and Wu et al. [27]. In contrast to the effect of okra mucilage, the presence of black seed oil in the prepared biocomposite films, due to the hydrophobic nature of oils, led to improved barrier properties of the films. This behavior can be attributed to the discontinuity of the hydrophilic phase, creating a tortuous path for the passage of water vapor molecules [36, 37]. Furthermore, the reaction between biopolymer chains and black seed oil compounds caused a reduction in the mobility of polymer chains and a more compact structure, such that the free volume of the film matrix decreases, and therefore WVP decreased [37]. The reported results by Niknam et al. [17], Taqi et al. [22], Nisar et al. [38] are consistent with the results of this research.

3.4. Color Properties

Based on Table 1, the film formulated with 1.29% okra mucilage and 0.07% black seed oil exhibited the highest lightness (53.23) and the lowest redness (11.72) (Figure 1). With increasing concentrations of black seed oil and okra mucilage, an increase in redness (a*), total color difference (ΔE), and yellowness index (YI) was observed, while lightness (L*) and whiteness index (WI) decreased. According to the results obtained in Table 2, all color factors significantly followed a linear model ($p < 0.05$), except for the yellowness parameter (b*), for which none of the models were statistically significant ($p > 0.05$). These results are consistent with the findings of Kannani and Rhim [21], who reported in agar-based films containing grape seed extract a decrease in lightness (L*) and an increase in redness (a*) and yellowness (b*) with increasing extract concentration. Also, in another study by Sun et al. [30], with increasing concentration of young apple polyphenol extract, a decrease in lightness occurred in chitosan-based films. Regarding the effect of oil on the color properties of films, Ghanbarzadeh and Almasi [12] reported that increasing the concentration of oleic acid in carboxymethyl cellulose-based films led to increased turbidity and yellowness index, and this phenomenon could be related to the light scattering effect by the distribution of fat particles during the film drying process. Also, in a study by Nisar et al. [38], they found that the addition of clove essential oil to pectin-based films caused a decrease in film brightness. These findings are consistent with the results obtained by Saberi et al. [37] and Hasheminya et al. [26].

3.5. Evaluation of Free Radical Scavenging Activity (DPPH) and Total Phenolic Content (TPC)

According to Table 1, the highest antioxidant activity was assigned to the film with a formulation of 0.27% okra mucilage and 0.07% black seed oil. Figure 1 shows that antioxidant activity and total phenolic content of the films increased with increasing okra mucilage concentration, while increasing black seed oil concentration decreased the antioxidant activity and total phenolic content. Based on Table 2, both antioxidant activity and total phenolic content parameters significantly followed a quadratic model ($p < 0.05$). The increase in antioxidant activity of films with increasing okra mucilage concentration is related to the increase in hydroxyl groups of polyphenolic compounds present in okra mucilage, which act as electron donors to free radicals and, by donating electrons to free radicals, prevent the propagation chain of free radicals. Therefore, the antioxidant activity increased with increasing okra mucilage concentration [15]. The increase in antioxidant activity of films following the increase in okra mucilage concentration is consistent with the findings of other researchers [23, 30]. The decrease in antioxidant activity with increasing black seed oil concentration could be because black seed oil, compared to other vegetable oils, has a high propensity for oxidation due to its high content of unsaturated fatty acids; it is sensitive and easily oxidizes to produce peroxide compounds. Therefore, since black seed oil has a high peroxide value, the oxidation rate increases, and the antioxidant activity decreases with increasing oil concentration [10]. On the other hand, volatile and antioxidant compounds are oxidized during oil extraction and the film preparation and drying process, and due to the oxidation of these compounds, the antioxidant capacity of black seed oil decreases, so the antioxidant activity of the films showed a decrease [39]. In confirmation of these results, Dinagaran et al. [40] reported that with increasing concentration of black seed oil, the antioxidant activity of films decreased. The decrease in total phenolic content with increasing black seed oil concentration could be due to the reaction of phenols with other compounds present in the film, which prevents the complete release of phenolic compounds from the film structure; consequently, the measured total phenolic content decreased.

Table1. Experimental data from impacts of independent variables (OM, BSO) on responses including THI, D, WS, WVP, L*, a*, b*, ∆E, WI, YI, DPPH, TPC, Ra and Rq by central composite design

Independent variables	Coded levels	-1	0	1
Okra mucilage (OM)	A	1	2	3
Black cumin seed oil (BSO)	B	0	0.25	0.5

	Variables		Responses													
Run	OM	BSO	THI	D	S	WVP	L*	a*	b*	∆E	WI	YI	DPPH	TPC	Ra	Rq
1	2.71	0.43	0.194	4.68	55.38	7.258	39.14	36.99	43.88	82.72	12.63	183.62	26.82	66.15	52.64	65.31
2	2.71	0.07	0.173	3.92	65.04	8.961	41.11	31.69	49.3	78.79	16.91	171.34	39.8	62.18	38.44	48.41
3	2.00	0.25	0.145	2.62	55.14	7.511	46.15	24.98	51.23	74.24	21.58	158.59	14.86	65.12	34.25	43.6
4	1.00	0.25	0.11	1.58	49.97	5.698	51.94	13.46	49.79	66.35	29.49	136.95	11.61	60.35	19.39	24.38
5	2.00	0.25	0.14	2.73	54.96	7.814	47.13	23.95	52.22	73.27	23.07	156.56	14.12	65.22	32.25	40.54
6	2.00	0.25	0.146	2.61	54.68	6.923	45.51	25.26	51.01	75.29	21.99	159.61	13.91	64.97	31.55	39.21
7	2.00	0.50	0.157	2.97	50.53	6.777	42.4	29.19	50.25	77.53	18.17	169.3	19.21	61.06	42.12	51.46
8	1.29	0.43	0.121	1.79	45.31	5.223	49.29	16.15	48.52	67.71	27.98	140.6	8.78	56.64	22.09	27.78
9	2.00	0.25	0.151	2.81	55.51	7.51	45.98	23.69	50.25	74.82	20.85	154.55	15.2	65.29	36.55	45.91
10	2.00	0.00	0.135	2.33	59.96	7.77	48.3	17.82	52.68	71.78	24.06	155.82	26.4	59.17	29.85	36.94
11	2.00	0.25	0.139	2.45	56.1	7.009	46.19	25.97	51.27	73.96	21.81	159.83	14.5	64.89	33.15	42.17
12	3.00	0.25	0.185	4.35	60.8	7.986	39.42	33.35	48.9	80.36	15.26	177.2	34.11	70.52	46.38	61.83
13	1.29	0.07	0.102	1.38	55.02	6.017	53.23	11.72	50.07	65.42	30.49	134.36	16.21	54.62	18.11	23.06

OM: Okra mucilage, BSO: Black seed oil, THI: Thickness, D: Density, S: Solubility, WVP: Water vapor permeability, L*: Lightness, a*: Redness, b*: Yellowness, ∆E: Total color difference, WI: Whiteness index, YI: Yellowness index, TPC: Total phenolic content, Ra: Average roughness, Rq: Root Mean square roughness

Table 2. Variance analysis of the CMC/OM/BSO biocomposite film responses including THI, D, WS, WVP, L*, a*, b*, ∆E, WI, YI, DPPH, TPC, Ra, Rq

Factor	Suggested model	R^2	p-value
Thickness (THI)	Linear	0.96	0.0001*
Density (D)	Linear	0.94	0.0001*
Solubility (S)	Linear	0.97	0.0001*
Water vapor permeability (WVP)	Linear	0.84	0.0001*
Redness (a)	Linear	0.95	0.0001*
Yellowness (b*)	Quadratic	0.7	0.0787 ^{ns}
Lightness (L*)	Linear	0.97	0.0001*
Total color difference (∆E)	Linear	0.95	0.0001*
Whiteness Index (WI)	Linear	0.96	0.0001*
Yellowness Index (YI)	Linear	0.95	0.0001*
DPPH	Quadratic	0.97	0.0001*
TPC	Quadratic	0.92	0.0009*
Ra	Linear	0.94	0.0001*
Rq	2FI	0.97	0.0001*

The sign of * shows the significant state at $P < 0.05$, ^{ns} shows the not significant state.
2FI: Two factor interactions

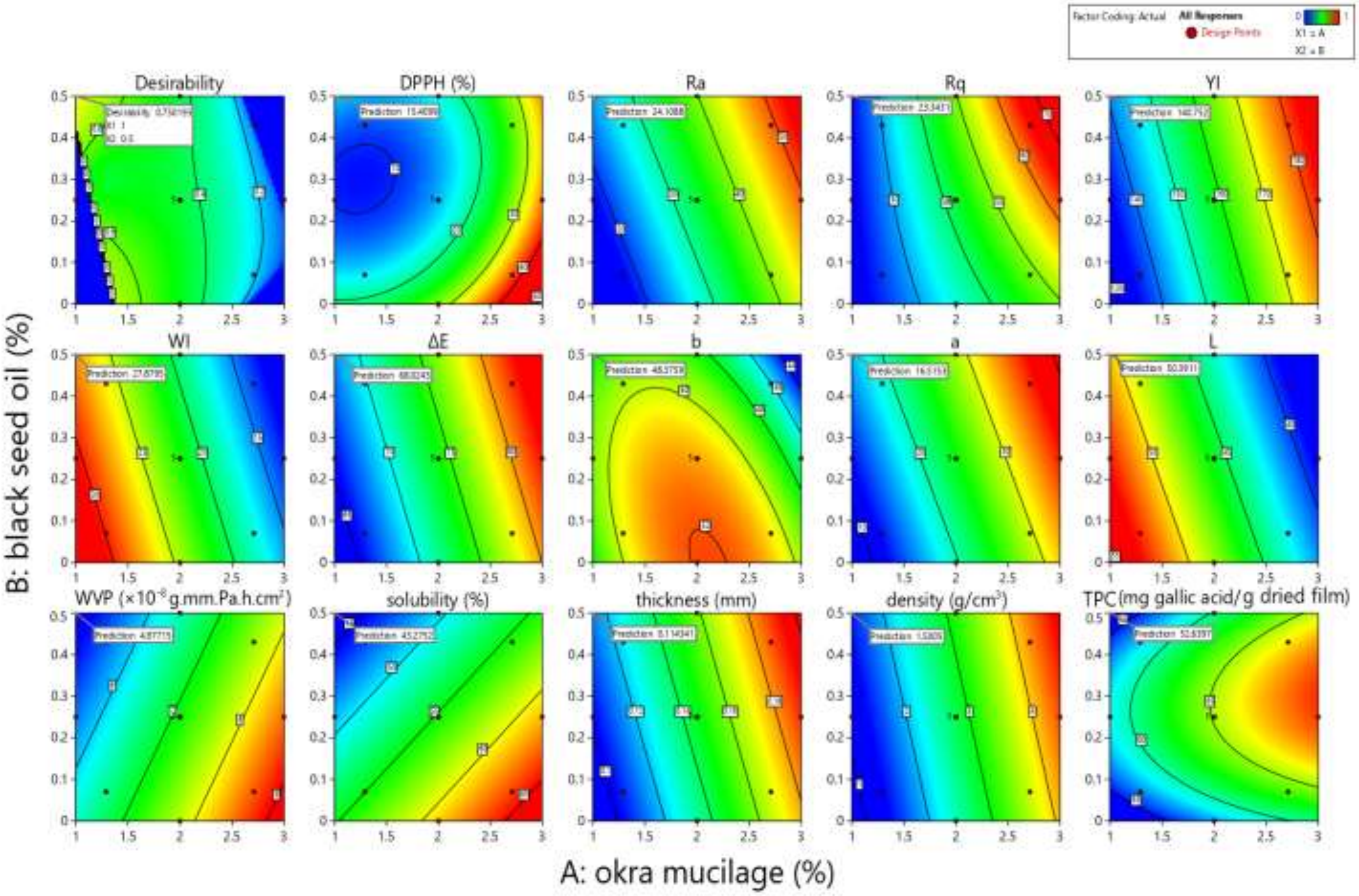
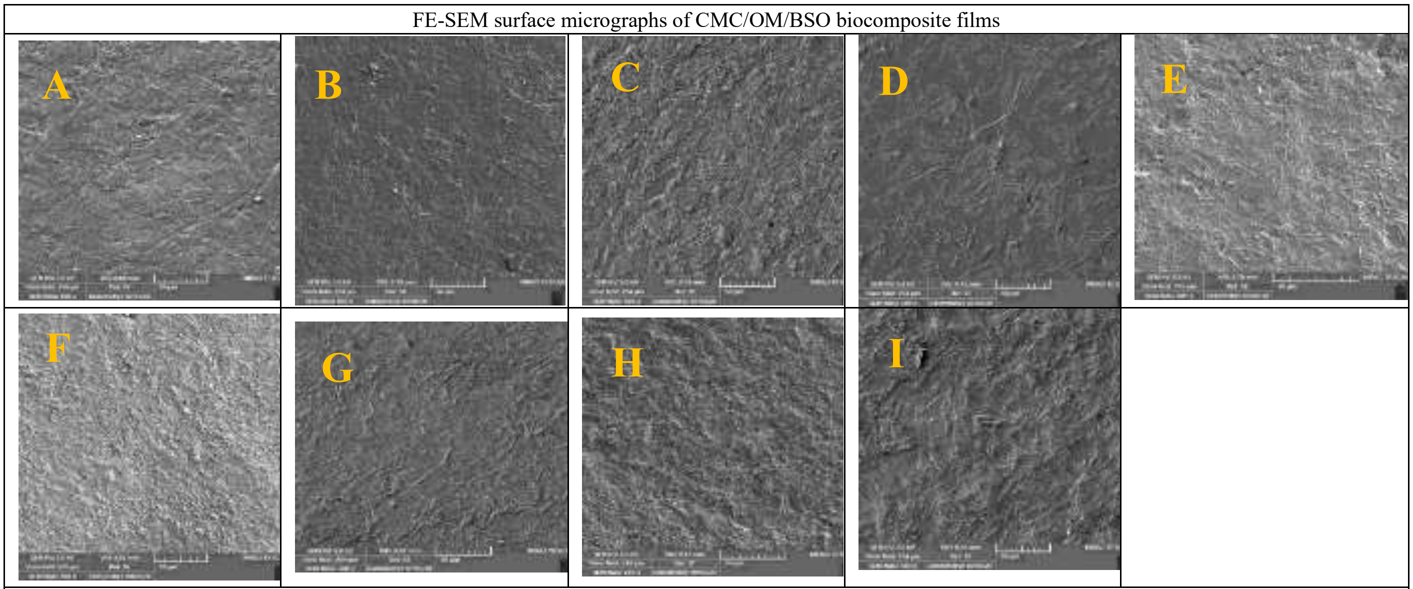


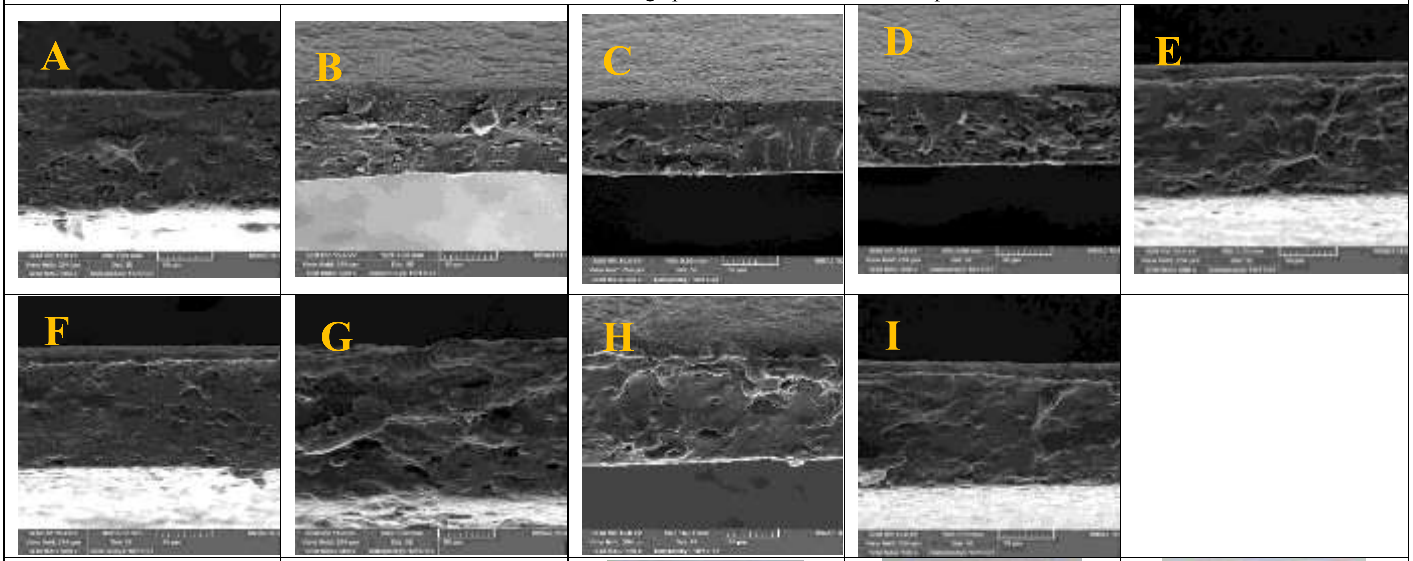
Fig 1. Desirability plot and contour plots of responses of the CMC/OM/BSO biocomposite films at various OM and BSO concentrations.

3.6. Microstructure Study with Scanning Electron Microscopy (SEM)

To better analyze the microstructure of the prepared films, a scanning electron microscope (SEM) was used. The SEM micrographs provide valuable information about the microstructure of films in relation to the distribution pattern and arrangement of particles and play an important role in the mechanism of water vapor transfer and film opacity [42]. The SEM micrographs obtained from the surface and cross-section of the film samples are shown in Figure 2. As observed, the film surface prepared with a concentration of 1.29% okra mucilage and 0.07% black seed oil showed a more uniform and integrated structure, whereas at higher concentrations of okra mucilage (2.71%) and black seed oil (0.43%), the films exhibited more accumulation, non-uniformity, and rough texture due to the formation of knots and aggregates of okra mucilage and black seed oil particles in the film structure. The presence of polyphenolic compounds in okra mucilage can improve the network structure due to their hydrophilic nature and absorb more water content in the matrix, leading to the aggregation and accumulation of insoluble polyphenolic compounds and consequently causing more irregularities, roughness, and disorder in the films [23, 27]. In addition to the effect of polyphenol accumulation, the low miscibility of the oil phase in the polymer matrix influences the film's microstructure. The concentration of oil particles on the film surface during the drying step increases particle accumulation, oil phase separation, and finally, crystallization of fatty acids in the film matrix, thereby causing disorder and non-uniformity of the structure in the films [29, 42]. Akyuz et al. [13] demonstrated that adding sunflower oil and animal fat to chitosan-based films resulted in a non-uniform and discontinuous structure in the polymer network due to the formation of two separate phases of oil and biopolymer. Rougher, more uneven surfaces and particle accumulation with the incorporation of oil in the film matrix have been reported by other researchers [17, 22, 32].

Based on the SEM images recorded from the cross-section of the film samples, with increasing okra mucilage concentration, the porosity and heterogeneity of the film matrix increased, creating a sponge-like structure in the cross-section of the film samples. In contrast, with increasing black seed oil concentration, the polymer network became more densely packed, and the amount of porosity in the cross-section of the samples decreased, so that the film prepared with low mucilage concentration (1.29%) and high oil concentration (0.43%) had the lowest porosity. The reduction of pores and increased continuity in the cross-sectional microstructure of films with increasing oil concentration can be attributed to the occurrence of reactions and the formation of new bonds between the phenolic compounds of okra mucilage and the lipid fraction (black seed oil) [41]. Wu et al. [27] showed that adding green tea polyphenols at high concentrations to pomelo peel-based films led to increased porosity and decreased film continuity, which is consistent with the findings of this research. In a study, Rubilar et al. [33] reported that the addition of carvacrol as a hydrophobic agent to a chitosan film matrix resulted in a cross-section observed as stacked sheets within compressed layers. In another study, Fabra et al. [41] investigated the effect of soybean oil and oleic acid along with green tea and grape seed extract on alginate-based films and reported that adding oil alone to the alginate film caused the formation of two separate phases of oil and alginate, whereas after adding the extracts to the film matrix, the lipid compounds were randomly distributed throughout the film cross-section, and no clear lipid phase separation was visible. Also, Ma et al. [43] studied the structure of Tara gum-based film containing oleic acid, and based on images obtained from the film cross-section, it was proven that at higher concentrations of oleic acid, the pore size decreased, as in this study, with increasing oil concentration, smaller pores appeared in the cross-section of the films. The SEM images obtained from the cross-section of the films confirm the results of the permeability measurements.


FE-SEM surface micrographs of CMC/OM/BSO biocomposite films


FE-SEM cross-sectional micrographs of CMC/OM/BSO biocomposite films

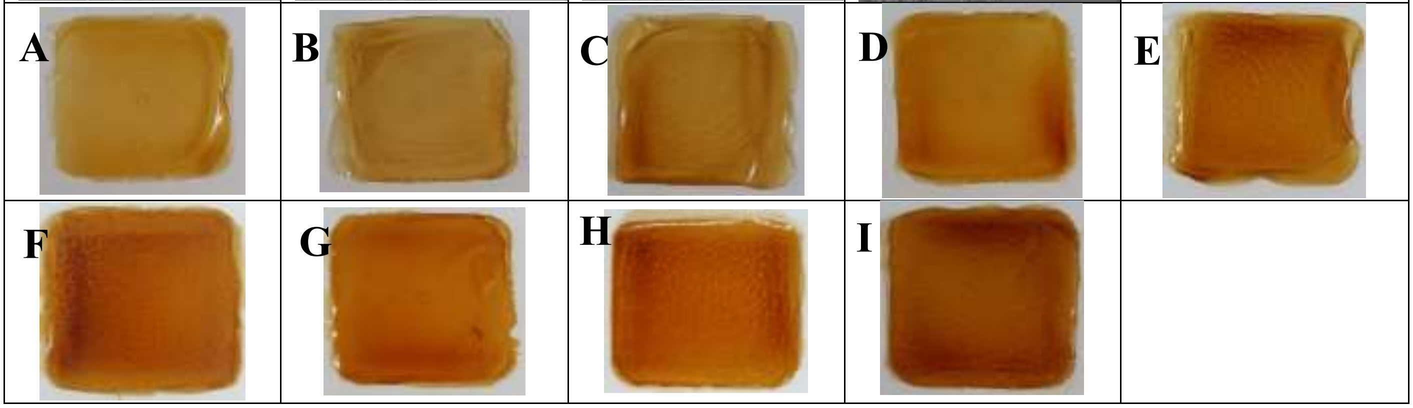
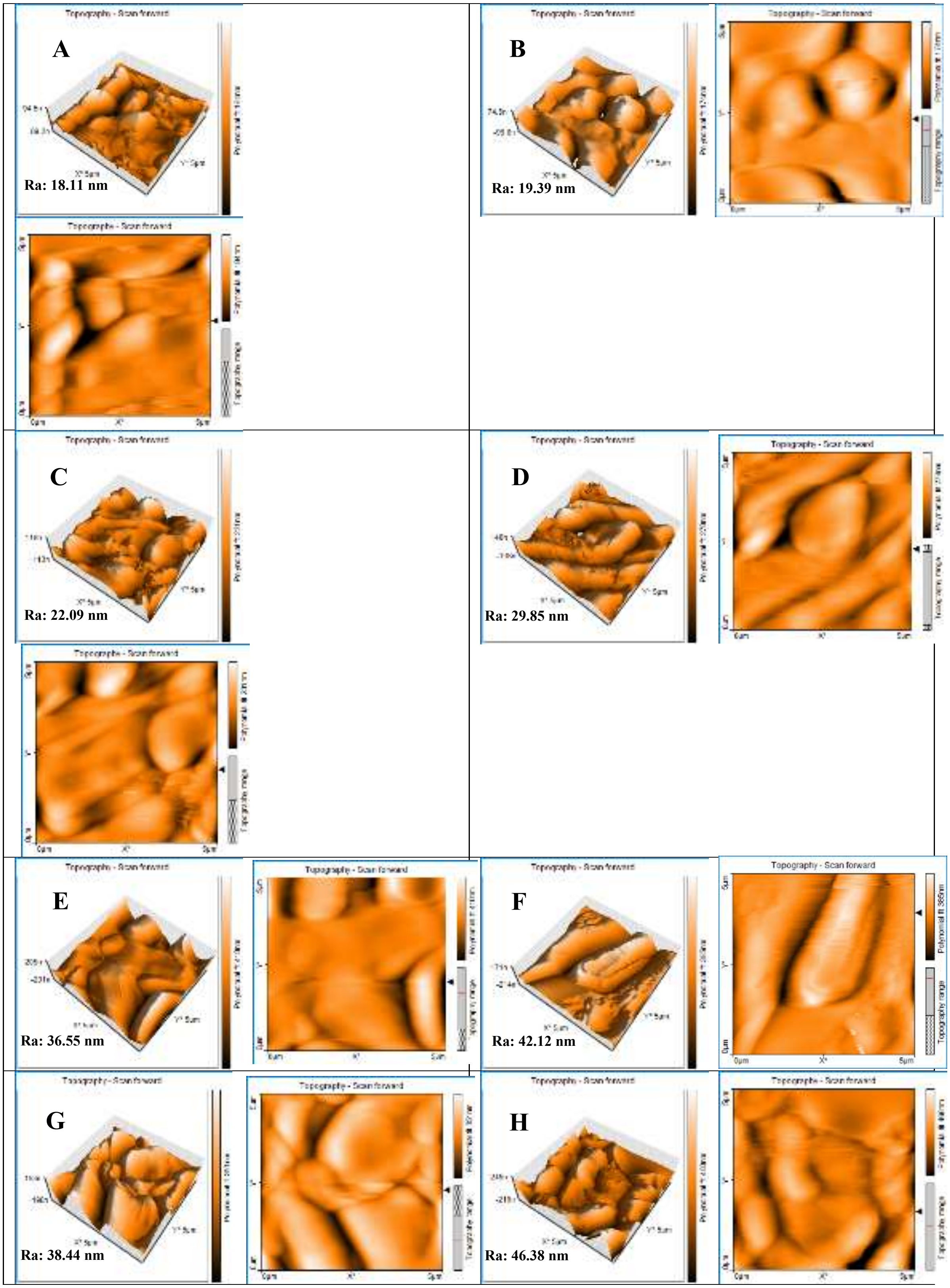


Fig 2. Photographs and FE-SEM surface and cross-sectional micrographs of CMC/OM/BSO biocomposite films at various OM and BSO concentrations, A (OM:1.29%, BSO:0.07%), B (OM:1%, BSO:0.25%), C (OM:1.29%, BSO:0.43%), D (OM:2%, BSO:0%), E (OM:2%, BSO:0.25%), F (OM:2%, BSO:0.5%), G (OM:2.71%, BSO:0.07%), H (OM:3%, BSO:0.25%), I (OM:2.71%, BSO:0.43%).

3.7. Microstructure Study with Atomic Force Microscopy (AFM)

By examining the topographic images of the film surfaces using Atomic Force Microscopy (AFM), parameters such as Ra (average roughness) and Rq (root mean square roughness), which are indicators of surface roughness, can be obtained. The AFM images of the prepared biocomposite films are shown in Figure 3. The film prepared with the formulation of 2.71% okra mucilage and 0.43% black seed oil had the highest surface roughness. According to Figure 1, it was determined that with increasing concentrations of okra mucilage and black seed oil, the Ra and Rq parameters increased, and based on Table 2, a linear model and a two-factor interaction model were proposed for the Ra and Rq parameters, respectively, both of which were significant ($p < 0.05$). The increase in roughness parameters can be attributed to the insoluble particles of okra mucilage in the film matrix, which cause the polymer chains to be irregularly placed next to each other, creating a non-uniform and uneven structure on the film surface. The increase in film surface roughness and unevenness, in addition to the accumulation and agglomeration of insoluble mucilage particles, is also related to the non-uniform distribution of oil particles during film drying, which increases with increasing oil concentration [32]. Atarés et al. [44] observed that in sodium caseinate-based edible films containing cinnamon and ginger essential oils, with increasing essential oil concentration, more irregularities and unevenness were observed on the film surfaces. Jiménez et al. [45], studying the surface microstructure of hydroxypropyl methylcellulose (HPMC)-based films containing lauric, myristic, palmitic, stearic, and oleic fatty acids, found that roughness parameters increased with increasing fatty acid concentration, which is consistent with the findings of this research.


Topography - Scan forward
A
Ra: 18.11 nm
B
Ra: 19.39 nm
C
Ra: 22.09 nm
D
Ra: 29.85 nm
E
Ra: 36.55 nm
F
Ra: 42.12 nm
G
Ra: 38.44 nm
H
Ra: 46.38 nm
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X*
5µm
Topography range

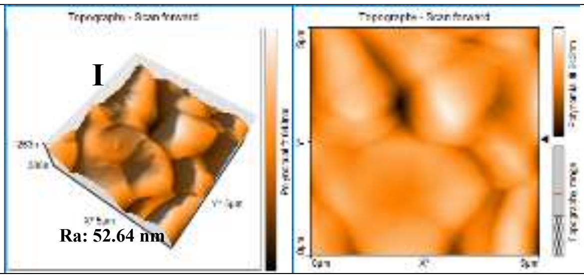

Fig 3. AFM surface micrographs of CMC/OM/BSO biocomposite films at various OM and BSO concentrations, A (OM:1.29%, BSO:0.07%), B (OM:1%, BSO:0.25%), C (OM:1.29%, BSO:0.43%), D (OM:2%, BSO:0%), E (OM:2%, BSO:0.25%), F (OM:2%, BSO:0.5%), G (OM:2.71%, BSO:0.07%), H (OM:3%, BSO:0.25%), I (OM:2.71%, BSO:0.43%).

3.8. Fourier Transform Infrared Spectroscopy (FTIR)

Different frequencies of IR radiation are absorbed by different chemical groups and bonds in the sample structure; therefore, infrared spectroscopy can be used to investigate interactions and structural changes in the prepared biocomposite films. The FT-IR spectra of CMC-based films are shown in Figure 4. The spectrum of the CMC film showed a prominent peak at 3430 cm^{-1}, which corresponds to O-H stretching vibrations. Also, absorption peaks were recorded at wavelengths of 2925, 1624, and 1047 cm^{-1}, which refer to aliphatic C-H stretching vibrations, carboxyl C=O groups, and C-O stretching vibrations, respectively, which are consistent with previous studies [16]. In all films, a broad band was observed in the 3430-3445 cm^{-1} region, indicating O-H stretching vibrations. It was determined that with increasing okra mucilage concentration, the width of the O-H band decreased, and this change indicates an increase in free hydroxyl groups [23]. Regarding the effect of oil on the film structure, it was proven that with increasing oil concentration, changes occurred in the film's structural bonds. In the film with the formulation of 2.7% okra mucilage and 0.07% black seed oil, O-H stretching vibrations appeared at a wavenumber of 3445 cm^{-1}. With increasing black seed oil concentration in the film formulation to 0.25% and 0.43%, the peaks shifted to lower wavelengths of 3434 cm^{-1} and 3433 cm^{-1}, respectively. These changes confirm the formation of hydrogen bonds between the carboxyl groups of CMC and the hydroxyl groups of phenolic compounds present in okra mucilage and black seed oil in the film matrix [23]. The absorption peak of O-H in the film formulation of 2% mucilage and 0% oil appeared at 3432 cm^{-1}, which shifted to lower wavelengths of 3430 cm^{-1} with increasing oil concentration to 0.5%. In the film containing 1.29% mucilage and 0.07% oil, the peak was observed at 3437 cm^{-1}, and with increasing oil concentration to 0.25% and 0.43%, it shifted to lower wavelengths of 3439 cm^{-1} and 3442 cm^{-1}, respectively. Observing these changes, similar to other films, proves the occurrence of intramolecular reactions and the formation of hydrogen bonds with increasing oil concentration [38]. These results are consistent with the findings of Nisar et al. [38] and Moghadam et al. [46]. The peak in the 2900-3000 cm^{-1} range recorded for all film samples is related to C-H stretching vibrations. As observed, in samples containing higher oil concentrations, the intensity and strength of the peak at the wavenumber of 2926 cm⁻¹ increased, which is consistent with the findings of Hasheminya et al. [26] and Ma et al. [43]. Furthermore, films containing higher oil concentrations exhibited an absorption band at 1745 cm⁻¹ with greater intensity, which arises from C=O stretching vibrations. Absorption bands in this range belong to the ester group of fatty acids and are attributed to the presence of oil [13]. Also, bands in the wavelength ranges of 1427-1460 cm⁻¹ and 1125 cm⁻¹ were recorded for all samples, which belong to CH_2 bending vibrations and carbohydrate C-O-C groups, respectively [26].

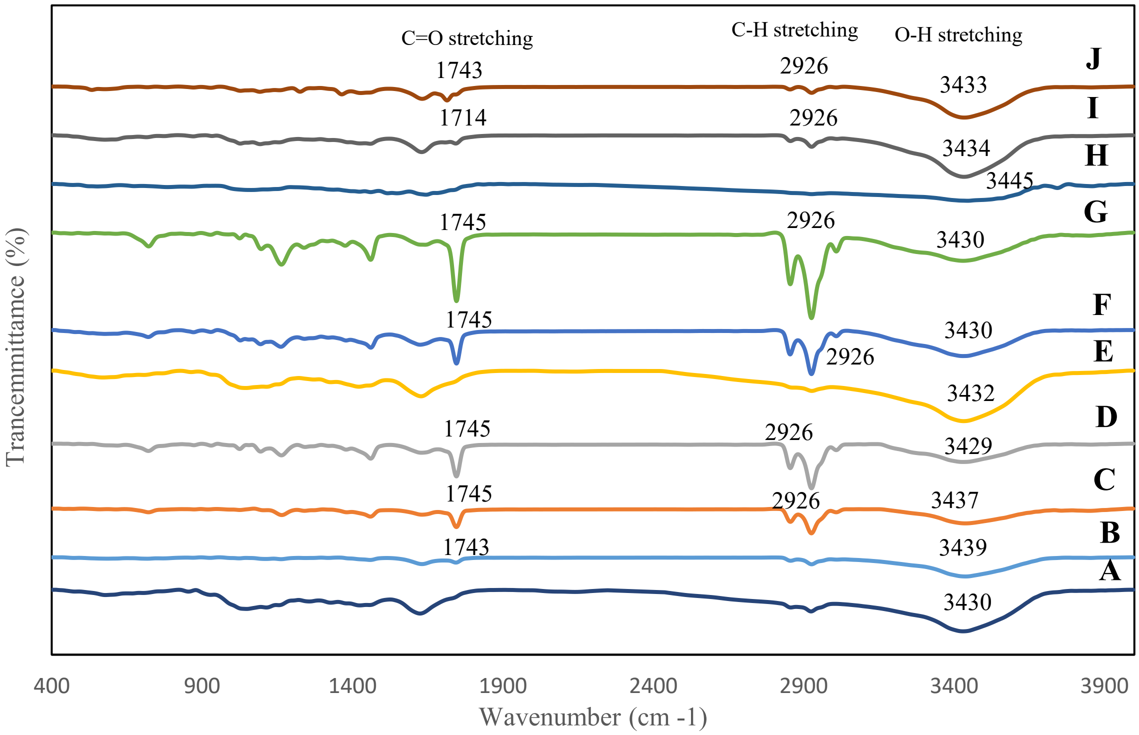

Fig 4. FT-IR spectra for all film samples, A (CMC control), B (OM:1.29%, BSO:0.07%), C (OM:1%, BSO:0.25%), D (OM:1.29%, BSO:0.43%), E (OM:2%, BSO:0%), F (OM:2%, BSO:0.25%), G (OM:2%, BSO:0.5%), H (OM:2.71%, BSO:0.07%), I (OM:3%, BSO:0.25%), J (OM:2.71%, BSO:0.43%).

3.9. Optimization of Carboxymethyl Cellulose-based Biocomposite Film Containing Okra Mucilage and Black Seed Oil

Optimization was performed to achieve the best formulation of the carboxymethyl cellulose-based film containing okra mucilage and black seed oil. The optimization conditions for producing a film with the best properties are shown in Table 3. In this optimization, the parameters of solubility, water vapor permeability, and antioxidant activity were maximized, while color properties and surface roughness parameters were minimized. The yellowness index (YI) parameter, being non-significant, was not considered for optimization. Also, the parameters were ranked according to their importance for film quality, with the number 5 chosen for the most important parameters. The order of importance of the parameters is shown in Table 3. The contour plots in Figure 1 show the relationship between the independent variables and the response variables. Also, according to Figure 1, the optimal combination of 1% okra mucilage and 0.5% black seed oil was obtained with a desirability of 0.75.

Table 3. Optimum conditions for achieving optimal CMC/OM/BSO biocomposite film

Factor	Goal	Importance
Okra mucilage	is in range	3
Black seed oil	is in range	3
Thickness (THI)	is in range	3
Density (D)	is in range	3
Solubility (S)	maximize	5
Water vapor permeability (WVP)	maximize	5
Redness (a)	is in range	3
Lightness (L*)	minimize	3
Total color difference (ΔE)	minimize	3
Whiteness Index (WI)	minimize	3
Yellowness Index (YI)	minimize	3
DPPH	maximize	4
Ra	minimize	2
Rq	minimize	2

4- Conclusion

In this research, the production of CMC-based biocomposite films containing okra mucilage and black seed oil was studied. Based on the obtained findings, with increasing okra mucilage concentration, the solubility and water vapor permeability parameters increased, while with increasing black seed oil concentration, they decreased. The total color difference (ΔE) and yellowness index (YI) parameters increased with increasing concentrations of okra mucilage and black seed oil. The images obtained from the film microstructure by AFM and SEM confirmed that with increasing concentrations of okra mucilage and black seed oil, the roughness, unevenness, and particle accumulation on the film surface increased. Also, the study of film structure by FTIR revealed that with increasing black seed oil concentration, hydrogen bonds formed between the free hydroxyl groups present in the film matrix. Furthermore, increasing the concentration of okra mucilage increased the antioxidant activity of the films, while increasing the concentration of black seed oil decreased the antioxidant activity of the films. Therefore, overall, the CMC-based film containing okra mucilage and black seed oil can be considered as a nature-friendly alternative for synthetic food packaging.

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Conflict of Interest

The author confirms that there are no financial conflicts of interest or competing interests in this study.

Author Contributions

All activities were performed by the author.

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مقاله علمی_پژوهشی

بررسی خصوصیات فیزیکی، آنتی اکسیدانی و ساختاری فیلم‌های خوراکی تولید شده بر پایه کربوکسی متیل سلولز حاوی موسیلاژ بامیه و روغن سیاه دانه

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چکیده

هدف از این تحقیق تهیه فیلم خوراکی بر پایه کربوکسی متیل سلولز (w/v ۱٪)، موسیلاژ بامیه (w/v ۳٪ –۱) و روغن سیاه دانه (v/v ۰/۵٪ –۰) بود. تأثیر غلظت‌های مختلف موسیلاژ بامیه و روغن سیاه دانه بر ویژگی‌های فیلم خوراکی تهیه شده شامل ضخامت، چگالی، انحلال‌پذیری، نفوذپذیری به بخار آب، فعالیت آنتی‌اکسیدانی، خصوصیات رنگی و ساختاری به روش سطح پاسخ (RSM) مورد بررسی قرار گرفت. نتایج نشان داد که پارامترهای انحلال‌پذیری و نفوذپذیری به بخار آب با افزایش غلظت موسیلاژ بامیه افزایش یافتند، در صورتیکه با افزایش غلظت روغن سیاه دانه کاهش در این پارامترها مشاهده شد. پارامترهای قرمزی (*a)، اختلاف رنگ کل (ΔE) و شاخص زردی (YI) با گنجاندن غلظت‌های بالاتر موسیلاژ بامیه و روغن سیاه دانه روند افزایشی داشتند، درحالیکه پارامترهای روشنایی (*L) و شاخص سفیدی (WI) روند کاهشی نشان دادند. همچنین فعالیت آنتی‌اکسدانی (DPPH) و محتوای فنل کل (TPC) در نمونه‌های فیلم با افزایش غلظت موسیلاژ بامیه افزایش یافتند، در مقابل گنجاندن غلظت‌های بالاتر روغن سیاه دانه باعث کاهش فعالیت آنتی‌اکسیدانی و ترکیبات فنلی فیلم‌ها شد. ضمن اینکه تصاویر AFM نشان داد که با افزایش غلظت موسیلاژ بامیه و روغن سیاه دانه پارامترهای زبری (Ra و Rq) افزایش یافتند. مطابق با آنالیز واریانس مدل پیشنهادی برای همه پارامترها معنی‌دار بود (p <۰/۰۵) به جز پارامتر زردی (*b) که هیچ یک از مدل‌ها معنی‌دار نبود (p > ۰/۰۵). بر اساس بهینه‌سازی، ترکیب بهینه موسیلاژ بامیه و روغن سیاه دانه به ترتیب ۱٪ و ۰/۵٪ با مطلوبیت ۰/۷۵ به دست آمد. تصاویر SEM ثبت شده از میکروساختار مقطع عرضی فیلم‌ها نشان داد که با افزایش غلظت موسیلاژ بامیه تخلخل در مقطع عرضی فیلم‌ها افزایش یافت، در مقابل افزایش غلظت روغن سیاه دانه منجر به کاهش تخلخل در مقطع عرضی فیلم‌ها شد. علاوه بر این آنالیز FTIR، وقوع واکنش‌های درون مولکولی در ماتریس فیلم را تأیید نمود.