Research Article

Formulation of Microwave-Assisted Natural-Synthetic Polymer Composite Film and Its Physicochemical Characterization

Aman Ullah,[1,2] Nauman Rahim Khan ⬤ ,[2,3] Muaz Habib Khan,[1,2] Saima Mehmood,[1,2] Jahangir Khan,[4] Tayyaba Iftikhar,[5] Inam Ullah Gul,[1,2] Hafiz Muhammad Basit,[1,2] Mughal Qayum,[3] and Shefaat Ullah Shah[1,2]

[1]*Department of Pharmaceutics, Faculty of Pharmacy, Gomal University, DI Khan, 29050 KPK, Pakistan*
[2]*Gomal Centre for Skin/Regenerative Medicine and Drug Delivery Research (GCSRDDR), Faculty of Pharmacy, Gomal University, DI Khan, 29050 KPK, Pakistan*
[3]*Department of Pharmacy, Kohat University of Science and Technology, Kohat, 26000 KPK, Pakistan*
[4]*Department of Pharmacy, University of Malakand, Chakdara Lower Dir, 18800 KPK, Pakistan*
[5]*Department of Pharmacy, Abdul Wali Khan University Mardan, 23200 KPK, Pakistan*

Correspondence should be addressed to Nauman Rahim Khan; naumanpharma@gmail.com

This study is aimed at microwave-assisted synthesis of sodium carboxymethylcellulose and Eudragit L100 composite film and its physicochemical characterization. The film was developed with varying quantities of each polymer and treated with microwave at a fixed frequency of 2450 MHz with a power of 350 Watts for 60 and 120 s. All formulations were characterized for thickness/ weight uniformity, moisture adsorption, erosion and water uptake, tensile strength, and vibrational, thermal, and surface morphological analysis in comparison with untreated film samples. Results indicated that microwave treatment for 60 s significantly improved the tensile strength, reduced the water adsorption, delayed erosion, and reduced the water uptake in comparison with the untreated and 120 s treated film formulations. The vibrational analysis revealed rigidification of hydrophilic domains at OH/NH moiety and fluidization of hydrophobic domains at asymmetric and symmetric CH moieties, which is envisaged to be due to the formation of new linkages between the two polymers. These were later confirmed by thermal analysis where a significant rise in transition temperature, as well as enthalpy of the system, was recorded. The microwave treatment for 60 s is thus advocated to be the best treatment condition for developing sodium carboxymethylcellulose and Eudragit L100 composite polymeric films.

1. Introduction

Polymer composites were introduced to address the demerits associated with lone use of either natural and/or synthetic polymers where more than one polymer is mixed and/or treated with another organic or inorganic moiety to create a novel polymeric network with improved physicochemical attributes [1]. Various composites have been developed for modifying drug release [2]; in skin tissue engineering [3, 4]; to improve mechanical properties [5], bone tissue regeneration [6], implants [7, 8], dental applications [9], and skin tissue regeneration [10]; and as wound dressing material [11, 12], ureter prosthesis [13], sutures and catheters [14], and prosthetic limbs [15]. The lone use of synthetic polymer presented a high risk of rejection due to reduced bioactivity, inciting tissue necrosis upon degradation of polymer via hydrolysis [16], and natural polymers also are prone to demerits like strength inadequacy and uncontrolled material degradability [17]. Composites, made through the method of ionic cross-linking, involve the use of toxic chemical agents, whose complete removal from the polymer mixture after the reaction is difficult [18].

Sodium carboxymethylcellulose (Na-CMC) is a low-cost soluble and poly-anionic polysaccharide derivative of cellulose

which is a biocompatible and biodegradable polymer [19, 20]. Due to its polyelectrolyte nature, it is used as an excellent superabsorbent and acts as a viscosity modifier, thickener, and emulsifier [17]. It exhibits good thermal gelation and forms excellent films due to its polymeric structure and high molecular weight [21, 22]. It also finds widespread applications in drug delivery, textile printing, paper industry, nanocomposite materials, detergents and food industry, and wound healing applications [23, 24]. Na-CMC has numerous surface OH groups readily available to form H-bonds with other moieties for the purpose to improve its physicochemical properties [25, 26].

Eudragit®, prepared by the polymerization of acrylic and methacrylic acids or their ester, is a synthetic polymer [27]. The Eudragit L100 is a neutral copolymer of polyethacrylate, methacrylate, and trimethylammonium methacrylate chloride [28] with a 1 : 1 ratio of free carboxyl and ester groups [29]. It is hydrophobic in nature [27, 30] and also used as a film coating material [31], with sustained release [32], and has been used in drug delivery to the colon [33, 34] and vagina [35] and as an ophthalmic [36], transdermal [37], enteric [38], and gene medicine and vaccine [39].

Microwave is characterized by frequencies in the range of 300 MHz to 300 GHz [40, 41]. Microwave has been used in food processing [42], muscle and tendon injury treatments [43], and endometrial ablation [44]; as a transdermal permeation enhancer [45] and synthesis of carbon nitride dots from folic acid for cell imaging [46]; and for catalyzing proteolytic reactions [47] and enhancing skin drug retention for the local treatment of skin pathologies [48–50]. Microwave has been explored for cross-linking of polymers to prepare the polymeric microspheres [51], graft polymer synthesis for colon drug delivery [52], for hydrogel synthesis [53], as microwave coagulation treatment [53, 54], for tumor cell ablation [55, 56] and endometrial ablation [57, 58], and for transdermal penetration of drugs [59].

Composites are referred to inter- and intrapolymer cross-linking initiated by chemical and/or physical methods whose physicochemical properties are different than those of the individual polymers used in their fabrication [60, 61]. Composites, made using chemical and/or ionic cross-linking, involve the use of hazardous/toxic chemical agents whose complete removal from the polymer mixture is difficult [62], and also sometimes, the polymers may get exposed for a much higher amount of heat for a long time [63], which can deteriorate the polymers internally, necessitating the exploration of an alternate physical technique such as microwaves, which have the ability to physically cross-link polymers to fabricate a composite material. Hence, in this paper, a microwave-assisted composite polymeric film of synthetic and natural polymer comprising Eudragit L-100 and Na-CMC was fabricated and compared for their physicochemical attributes with and without microwave treatment.

2. Materials and Methods

2.1. Materials. Sodium carboxymethylcellulose (Na-CMC, high-viscosity grade with average molecular weight of ~250,000 Da Sigma-Aldrich, USA) and Ethanol (≥95%, Sigma-Aldrich, USA). Eudragit L-100, glycerol, calcium sulphate, and potassium sulphate were generously gifted by BioLab Islamabad, Pakistan. All chemicals were used without any further purification.

2.2. Methods

2.2.1. Composite Polymeric Film Preparation. Composite polymeric film was fabricated via the solution casting method. Briefly, accurately weighed 1 gram of Na-CMC was dispersed in enough distilled water with continues magnetic stirring (Gallenkamp, England) for 2 h till a homogenous solution was obtained followed by the addition of glycerol (1% of the formulation) as a plasticizer and making the final weight of 100 g with distilled water (solution-A). Similarly, accurately weighed 1 gram of Eudragit L-100 was dispersed in enough ethanol (95%) and kept stirred until a homogenous solution was obtained and the final weight was made up to 100 g with ethanol (solution-B).

Both polymer solutions were then mixed at specific concentrations followed by casting of 50 grams of each mixture into petri dishes (internal diameter: 88 mm). The dishes were air dried in an oven (SH-DD-100NG, Korea) at 40°C for 4 days and/or until complete drying of the films. Following complete drying, films were peeled off from the petri dishes and subjected to physicochemical characterization tests described in later sections. The formulation and mixing pattern are given in Table 1. When required, the bipolymer solution mixture when added into petri dishes was subjected to microwave treatment at a fixed frequency of 2450 MHz and an average power of 350 Watts (LG, MS2022D/00, Japan) for two different time intervals, i.e., 60 s and 120 s. The petri dishes after microwave treatment were then subjected to the same drying process as the other plates and handled as described above. The formulations and microwave conditions are given in Table 1.

2.2.2. Physicochemical Characterization Tests. The methods reported by Ali et al. [64] were followed for the physicochemical characterization tests performed on untreated and microwave-treated composite polymeric film in this project.

(1) Thickness and Weight Uniformity. The untreated and microwave-treated dried composite polymeric films were peeled/scratched off from the petri dishes after drying to determine the thickness measured using a micrometer screw gauge at different positions of the film, and the average thickness was calculated. This established the uniformity of the film thickness, which is directly related to the accuracy of the dose in the strip. The average and the standard deviation (SD) of the three readings were recorded.

The untreated and microwave-treated dried composite polymeric films after drying were peeled/scratched off from the petri dishes and cut into appropriate sizes to determine their dry weight on a weighing balance (M124A, Italy). The average and the standard deviation (SD) of the three readings were recorded.

TABLE 1: Composite film formulations.

(a)

Formulations	Solution A 1% (w/w)	Solution B 1% (w/w)	Glycerol 1% (w/w)	Drug 0.5% (w/w)	Remarks
F1 (2 : 1)	66.66gm	33.33gm	1gm	0.5gm	Very good, easily removable
F2 (3 : 1)	75gm	25gm	1gm	0.5gm	Excellent, easily removable
F3 (1 : 1)	50gm	50gm	1gm	0.5gm	Good, easily removable

(b)

Formulations	Microwave treatment		Remarks
	Ratios	Treatment time (sec)	
F1 (60)	2 : 1	60	Excellent easily removable
F1 (120)	2 : 1	120	Excellent easily removable
F2 (60)	3 : 1	60	Excellent easily removable
F2 (120)	3 : 1	120	Excellent easily removable
F3 (60)	1 : 1	60	Fair and not removable
F3 (120)	1 : 1	120	Fair and not removable

2 : 1 (2-part solution A, 1-part solution B), 3 : 1 (3-part solution A and 1-part solution B), 1 : 1 (1-part solution A and 1-part solution B), 1 : 3 (1-part solution A and 3-part solution B), and 1 : 2 (1-part solution A and 2-part solution B).

(2) Moisture Adsorption. The individually weighed-untreated and microwave-treated polymeric films (30 mm × 30 mm) were placed for complete drying in a desiccator with filled anhydrous calcium sulphate for 48 h. Following this time, the films were weighed individually and placed again in another desiccator for 48 h with filled saturated solution of potassium sulphate at virtual RH of 97% to let the films rehydrate again. After complete rehydration, the films were removed from the desiccator, individually weighed again, and percent moisture content determined by using the following relation.

$$\text{Moisture uptake (\%)} = \frac{W - W0}{W0} \times 100, \qquad (1)$$

where W is the weight of the rehydrated films and $W0$ is the initial weight.

At least triplicates were conducted, and results averaged.

(3) Erosion and Water Uptake. An accurately weighed untreated and microwave-treated (initial weight Wi) and known dimension (25 mm) of the film samples were placed in a petri dish containing 20 ml phosphate buffer (pH 7.4) simulating the pH of the wound bed conditions and incubated in an oven (DO-100NG, Korea) at 37°C ± 0.2°C for a duration of 30 minutes. At specific time intervals, film samples were removed from buffer solution and blot dried by gently sliding them on the surface of petri dish to remove adhered moisture and weighed (Wt). Afterwards, the same film samples were placed in an oven at 40°C ± 0.2°C for at least 5 days. The dried films were then transferred to a des-

iccator maintained at 25°C enabling the films to achieve a constant final weight (Wd).

$$E = \left(\frac{\text{Wi} - \text{Wt}}{\text{Wi}}\right) \times 100,$$
$$\text{WU} = \left(\frac{\text{Wt} - \text{Wd}}{\text{Wd}}\right) \times 100. \qquad (2)$$

Wi is the initial dry weight of the film sample, Wt is the wet weight of film sample after specific time interval, and Wd is the final oven dry weight of the film sample. Tests were performed in triplicate and results averaged with SD calculated as well.

(4) Tensile Strength. The ultimate tensile strength (UTS) of the untreated and microwave-treated polymeric films was determined at 25 ± 1°C using a universal testing machine Testometrics (United Kingdom). Three rectangular shaped strips with 7.5 cm length and 3.5 cm in width were cut from each of the film samples and were fixed between the grips of the machine. The initial grip separation and crosshead speed were set to 50 mm and 5 mm/min, respectively. Samples were pulled with 50 N loads. The maximum force to break the sample was recorded. Triplicates were conducted and the results averaged.

(5) Vibrational Analysis. The characteristic peaks of the dried untreated and microwave-treated composite polymeric films were recorded by an ATR-FTIR spectrophotometer (UATR TWO, Perkin Elmer, UK). Each film and/or powdered

polymer was placed on to the surface of the diamond crystal and clamped to ensure close contact and high sensitivity. All the samples were scanned over a wave number range of 400 to 4000 cm^{-1} with an acquisition time of 2 minutes. Each sample was analyzed three times and results averaged.

(6) Thermal Analysis. The thermal properties of untreated and microwave-treated films were determined by differential scanning calorimetry (DSC 200 F3 Maia Therma Analyzer, Netzsch-Gerätebau GmbH, Germany). A 2-3 mg film sample was cut and placed into an aluminum pan and crimped with the help of DSC pan compression assembly and placed in DSC furnace, which was preequilibrated with empty pans with ΔY value less than 0.3. The analysis was performed with a continuous purge of nitrogen gas at a flow rate of 40 ml/min with a heating rate of 10°C/min in the temperature range of 30 to 500°C. The characteristic transition temperatures and enthalpy values were recorded. At least triplicates were carried out and results averaged.

(7) Scanning Electron Microscopy. The surface and cross-section microstructure of untreated and microwave-treated films were observed by a Field-Emission Scanning Electron Microscope (S-570, Hitachi, Japan). For surface morphology, a sample of film 3 × 3 mm was cut and adhered to the SEM stub with a carbon tape. The samples were sputter coated with gold for 5 min. The samples were then viewed at an accelerating voltage of 10 kV and at magnifications of 1 k to 40 kx, and representative sections were photographed.

(8) Statistical Analysis. All experimental results are expressed as the mean of at least three experiments with corresponding standard deviation. Statistical data analysis was carried out using SPSS software version 18.0, and a statistically significant difference was denoted by $p < 0.05$. Student's t-test and analysis of variance (ANOVA)/post hoc analysis by Tukey HSD test were employed where applicable.

3. Result and Discussion

3.1. Physicochemical Characterization. The combination of Na-CMC and Eudragit L-100 exhibited good film forming properties with an average thickness in the range of 0.17 ± 0.01 to 0.19 ± 0.01 mm for untreated and 0.17 ± 0.01 to 0.20 ± 0.01 mm for microwave-treated formulations, respectively, where the difference was found to be insignificant (Student's t-test, $p > 0.05$). The trend of thickness is reflecting a slight increase in the case of untreated formulations (Table 2), which can be attributed to the fact of nonhomogeneous drying of the polymer solution, while in the case of microwave-treated samples, the same can be due to microwave irradiation, which involves heat generation. This might be changing the pattern of polymer fiber deposition and hence result in the formation of a nonhomogeneous composite matrix [65]. Similarly, no significant difference (Student's t-test, $p > 0.05$) in the average weight was observed when untreated and microwave-treated samples were compared (Table 2). By increasing the concentration of Na-CMC in composite mixture, the average weight experiences

TABLE 2: Thickness and weight uniformity of untreated films and microwave-treated films.

Formulations	Thickness (mm)	Weight uniformity (g)
F1	0.11 ± 0.01	0.20 ± 0.01
F2	0.14 ± 0.01	0.11 ± 0.00
F3	0.16 ± 0.01	0.11 ± 0.01
F2 (60)	0.15 ± 0.01	0.17 ± 0.01
F2 (120)	0.14 ± 0.01	0.17 ± 0.01
F3 (60)	0.16 ± 0.01	0.21 ± 0.02
F3 (120)	0.16 ± 0.01	0.18 ± 0.01

an increasing trend in the same way. Na-CMC is a high viscosity polymer, which depicts the possible reason behind the increasing weight trend in the weight uniformity in those formulations having high Na-CMC content [66].

3.2. Moisture Adsorption. Moisture adsorption provides information about the water and/or moisture retaining affinity of a matrix. Higher moisture content leads to softening of the matrix and hence reduced shelf life. The results of moisture content of untreated and microwave-treated polymeric composite films are shown in Figure 1. In the case of untreated samples, the significantly lowest water content was found to be that of the F2 formulation (Student's t-test, $p < 0.05$, $90.24 \pm 2.12\%$) compared to F1 and F3. The F2 formulation contains the highest Na-CMC content. This higher Na-CMC content would have high moisture content due to there being more hydrophilic polymer in the mixture. The excellent amalgamation of Eudragit L100 with Na-CMC resulted in enhancing the hydrophobicity of the resulting formulation through development of interpolymer linkages between the Na-CMC and Eudragit moieties and hence leaving less room for water molecules to interact with the material [67, 68]. The higher chain network flexibilities could enhance the maximum solvent uptake of the polymer and increasing the crosslink density could decrease the permeation rate [69]. Water absorption behavior is highly dependent on the voids available in the composite itself; thus, water will first fill up the empty voids rapidly through capillary action until the voids become limited; thus, the void content in the composite that allows water absorption can affect the degradation and its interfacial bond [70]. Additionally, occupation of surface OH groups in CMC chain by the Eudragit moieties reduces the hygroscopicity of the CMC moiety [71]. In case of microwave treatment, a significant decrease in the moisture content of the composite film was observed in a treatment time-dependent manner. Higher treatment times resulted in lower moisture adsorption of the film. Owing to the fact that, microwave involves heat generation during treatment, leading to evaporation of excess moisture from the polymer mixture, and hence, lesser moisture adsorption was observed upon complete drying [72–74].

3.3. Erosion and Water Uptake. "Erosion" designates the loss of material owing to monomers and oligomers leaving the polymer [75]. The erosion process is advantageous during
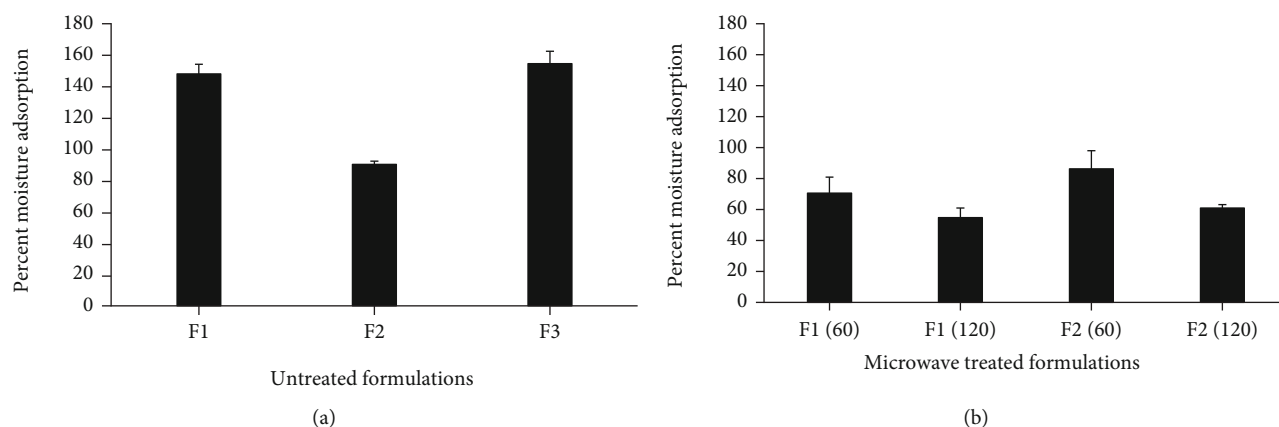
(a)

(b)

Figure 1: Percent moisture adsorption of (a) untreated films and (b) microwave-treated films.
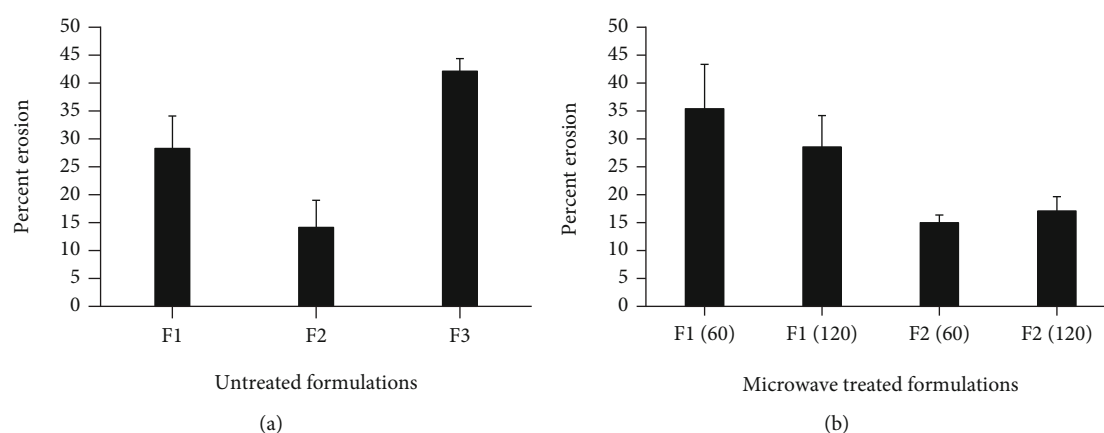

(a)

(b)

Figure 2: Erosion of (a) untreated films and (b) microwave-treated films.

the usage of such polymers for drug delivery, where the release of a drug and the rate of polymer erosion are directly interrelated [76], but in the case of film formulation, where it is mostly applied onto the skin, the delayed erosion is detrimental for an ideal topically applied plate form [25, 77]. The percent erosion results for untreated composite films are given in Figure 2(a). The results show that a significant decrease (Student's t-test, $p < 0.05$) in the time to erode was observed with the F2 formulation compared to F1 and F3. Despite the increasing hydrophilic Na-CMC moieties, the percent erosion for F2 decreases compared to F1 and F3, which could partly be explained by the increasing concentration of Na-CMC, which increases the availability of surface functional groups for interaction with other Eudragit moieties. This is a hydrophobic polymer and hence delays the time required for the composite film to erode compared with those films which contain less Na-CMC concentration [32]. Similarly, the increase in the cross-linking density also decelerates the affinity of rapid erosion [78]. When the formulations F1 and F2 were exposed to microwave irradiation, for 60 s and 120 s, the F1 formulation experienced a slight increase in erodibility with 60 s treatment while remained almost unaffected with 120 s treatment. In case of F2 formulation, the 60 s treatment did not produce any changes compared to untreated film while the 120 s treatment expe-

rienced a slight increase in percent erosion ability of the film (although insignificant statistically, Figure 2(b)). The reason could be that microwave heating effected the polymer chain arrangement, which could not successfully develop a strong hydrophobic pattern in the composite to prevent sufficient water penetration into the polymer matrix, thus preventing the erosion. The lower viscosity polymer eroded faster than the higher viscosity one. Thus, the higher molecular weight Na-CMC polymers had a higher intrinsic water holding capacity, and the matrices formed from such polymers were less prone to erosion [79]. Processes that have so far been identified to be responsible for erosion behavior are autocatalysis [80] and percolation phenomenon [81]. Autocatalysis arises from the uptake of water by the polymers which are shown in Figures 3(a) and 3(b) where increasing amounts of carboxylic acid end groups are created due to the degradation of the polymer [82, 83]. As the surface of the polymer is kept at neutral pH, a pH gradient develops that slows down the degradation of the polymer matrix surface compared to the center. The surface layer breaks at some point when a critical osmotic pressure builds up inside the matrix due to the accumulation of degradation products. Percolation phenomenon is because degradation products can apparently not leave the matrices prior to the erosion onset. Only after a critical degree of degradation is reached, the polymers
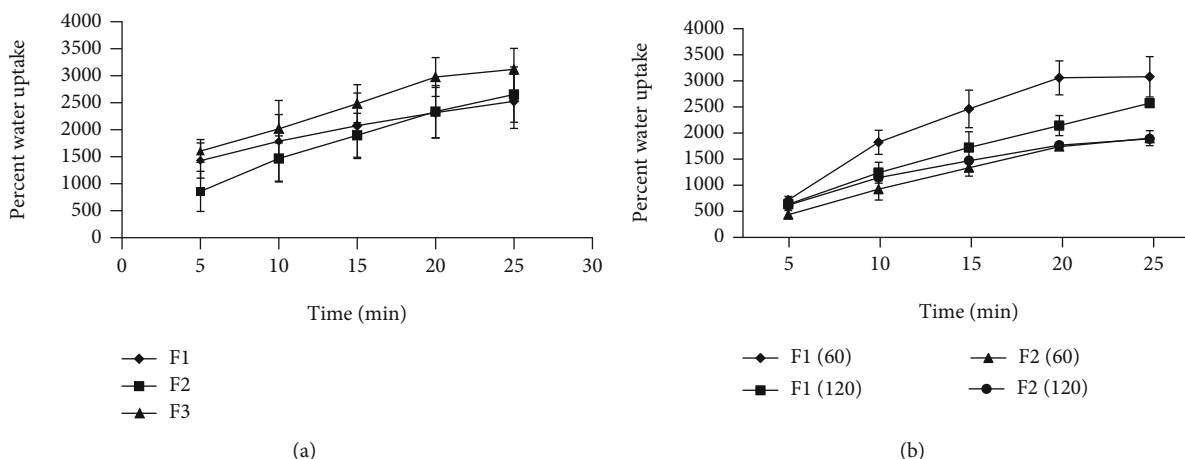
(a)

(b)

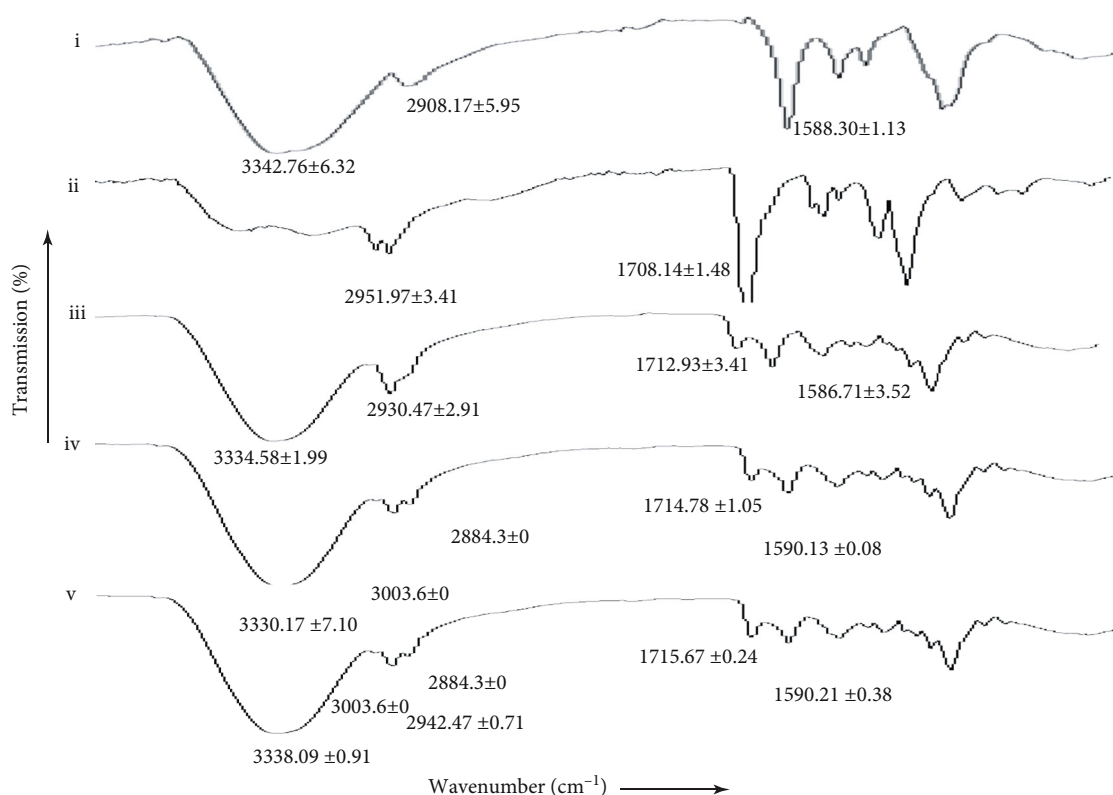FIGURE 3: Percent water uptake of (a) untreated films and (b) microwave-treated films.

FIGURE 4: FTIR spectra of pure powder (i) Na-CMC (ii) Eudragit L100, (iii) untreated film F2 (iv) microwave F2 (60) and (v) microwave F2 (120).

form a network of pores that allows for the release of monomers and oligomers [84].

Water uptake is a prime variable that can affect the performance of biodegradable polymers [85], by affecting the degradation, swelling and release of drugs [86] and biological response [87], and changes in the mechanical properties of the matrixes [31]. For wound healing, the water preserving ability of the films is very important in skin tissue engineering [88]. The water uptake results are shown in Figure 4. In case of untreated polymeric films (Figure 3(a)), the moisture uptake capacity of the films decreases with an increase in the Na-CMC contents, but no significant difference among the samples was observed (ANOVA, $p > 0.05$). Despite being a hydrophilic polymer, an increase in Na-CMC content provides more polymer for cross-linking with Eudragit moiety leading to formation of denser composite matrix which hindered the water penetration ability into it [32, 78, 89]. In case of microwave-treated samples, a significant fall in water uptake capacity of the film was observed for both time treatments: i.e., 60 s and 120 s, for F2 formulation while F1 remained almost unaffected (ANOVA, $p < 0.05$, Figure 3(b)). The F2 formulation contained more Na-
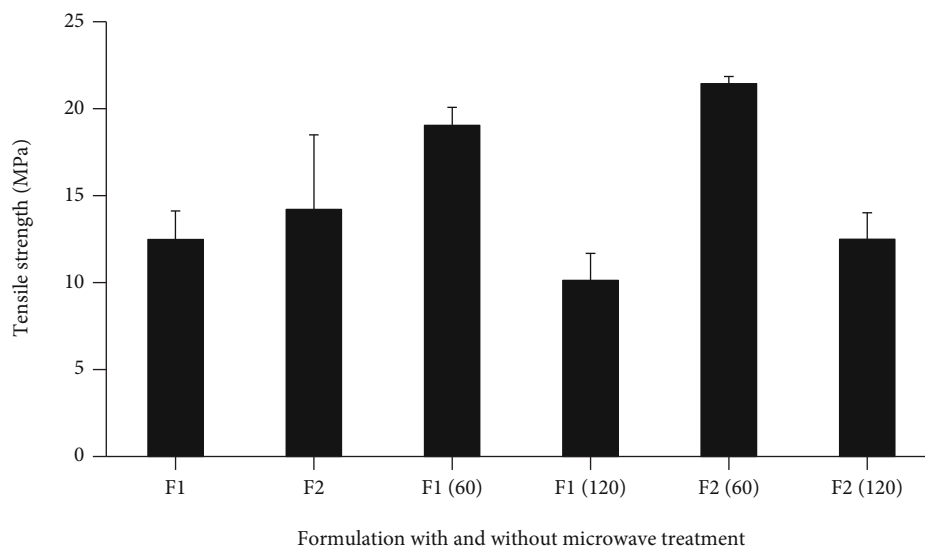
FIGURE 5: Tensile strength of the untreated and microwave-treated films.

CMC content compared to the F1 formulation. The efficient coverage of surface hydrophilic functional groups of Na-CMC by the Eudragit moiety shifted the hydrophilic nature of composite to hydrophobic thus mutual repulsion between the water molecules, and composite matrix prevented the water penetration and hence a significant fall in water uptake capacity was observed for F2 microwave-treated films [90]. Furthermore, microwave treatment increases the compactness of the composite film hence leaving less or no voids inside the mixed polymer matrix for water molecules to penetrate [91, 92]. Generally, in a polymer, the level of the hydrolysis depends on the number of the groups available in the polymer macromolecular chains that could react chemically with water molecules. The interaction level of the penetrant-polymer pair and the interaction effect on the polymer network will affect the amount of moisture absorbed in a solvent-solute system [93]. Diamant et al. [94] suggested that higher levels of cross-linking might remove possible interaction sites, and additional linking could retard the equilibration of the polymer due to the increased difficulty for the water molecules to access to the polymer sites.

3.4. Tensile Strength. Elasticity and strength of dressing materials (films) are among the primary factors that protect the wound surfaces from external factors [95]. The tensile strength analysis results of untreated and microwave-treated composite polymeric films are given in Figure 5.

In case of untreated samples, despite the increasing trend in the tensile strength of the film, the results were insignificantly different from each other (Student's t-test, $p > 0.05$) where it was increased from 12.03 ± 1.64 to 14.21 ± 4.28 MPa (megapascals) which was directly related to an increase in the Na-CMC ratio in the films and hence to the density of cross-linking in the film [78]. Furthermore, Na-CMC has an internal sugar ring structure which has a good skeleton effect on the composite structure leading to improved internal structural stability and hence increased mechanical properties [96]. Composite polymeric film composed of chitosan

and Na-CMC revealed increasing the content of one or another polymer was having positive impact on the mechanical properties of the film due to strengthening of cellulose crystals due to mutual interaction of polar functional groups [71]. Formulations exposed to 60 s microwave irradiation resulted in a statistically significant (Student's t-test, $p < 0.05$) rise in the TS where it was found to increase for F1 from 12.47 ± 1.64 MPa to 19.04 ± 1.03 MPa and similarly from 14.21 ± 4.28 MPa to 21.44 ± 0.40 MPa for F2 formulation. The reason for the increase in TS upon 60 s irradiation might be because microwave is able to rearrange the polymer fibers and effectively release the residual stress due to enhanced interfacial adhesion between the two polymer matrixes via the uniform heating characteristics of microwave energy [97, 98].

When the composite treatment time with microwave was increased to 120 s, a statistically significant fall in the mechanical attributes of the films was observed (Student's t-test, $p > 0.05$), where the TS for F1 reduced to 10.12 ± 1.64 and for F2 to 12.49 ± 2.10 MPa. Microwave involves transfer of energy from electromagnetic waves into the treated material in the form of heat. Owing to the fact that exposure of polymeric materials to microwave for higher time interval possesses the danger of thermal induced degradation, hence, 120 s treatment might have resulted in degradation of strength providing cellulose moiety and rupturing of intermolecular polymer chains in the matrix which led to reduction in the mechanical properties of the composite films [91, 99]. Based upon the comparative data analysis of F1 and F2 formulations treated with microwave for 60 s and 120 s, where F2 formulation proved significantly well and optimized in all physicochemical tests performed, therefore, it was decided to take only F2 formulation from this stage onward for further physicochemical analysis tests like ATR-FTIR, DSC, and SEM (discussed in later sections).

3.5. Vibrational Analysis. The vibrational analysis of Na-CMC powder showed characteristic hydrophilic regimens
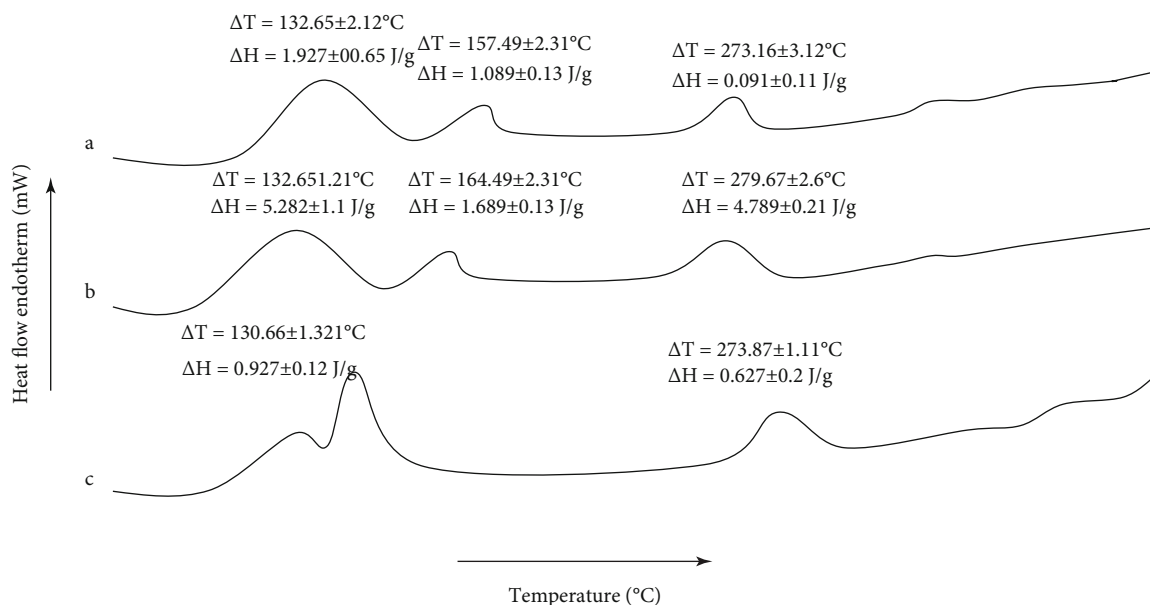
FIGURE 6: DSC thermograms of untreated and microwave-treated films: (a) F2, (b) F2 (60), and (c) F2 (120).

(OH/NH) in the range of 3350 to 3340 cm^{-1}, asymmetric CH band in the range of 2910 to 2905 cm^{-1}, and amide 1 band in the range of 1590 to 1585 cm^{-1}. Any further bands were not of interest, so they were not studied in later discussion, while in the case of Eudragit L-100 exhibited asymmetric CH bands (2960-2950 cm^{-1}) and carboxylic acid functional group band (1715 to 1700 cm^{-1}) only, owing to its hydrophobic nature where no OH/NH bands were observed.

In a comparative analysis of ATR-FTIR spectra of the F2 composite formulation with Na-CMC powder and Eudragit L100 powder spectra (Figure 4(i)–(iii)), a rigidification of hydrophilic domains was observed where the corresponding wave number reduced in case of films. Rigidification of domains of OH/NH represents compaction of the matrix in a way to reduce the entrapment and/or increased loss of moisture from the film structure [96, 100]. Another change in F2 was observed in terms of hydrophobic domain interaction where symmetric CH (2890-2885 cm^{-1}) appeared which was not found in Na-CMC and Eudragit L100 powder ATR-FTIR spectra. Owing to the fact that, Eudragit L100 may have covered up the Na-CMC surface OH functional groups, and hence, an increase in elasticity was induced due to formation of a new alkyl structure in the polymer matrix which led to splitting of only asymmetric CH band observed in a powder polymer into asymmetric and symmetric CH bands in the film [32, 53]. Bonatti et al.'s [101] study revealed the fact that nonwood plants incorporate cellulose which shows homogenous FTIR spectral profiles having the same frequency, i.e., in the range of CH region (2700-3000 cm^{-1}), by giving a similar molecular conformation. Furthermore, by passing from sorghum to kenaf, hemp, and flex, the width of band at half height decreases, which suggests that an increase in size and more perfection comes in the cellulose crystallites, so we can say that there would be an increase rigidification in the structure, while no significant difference in the wave number of C=O moiety of the Eudragit L100

(1715-1705 cm^{-1}, $p > 0.05$) was observed which depicts this moiety remained unaffected by the interactive capability of Na-CMC.

Treatment of F2 composite film with microwave for 60 s (Figure 4(iv)) brought dramatic changes in all hydrophilic and hydrophobic domains of the composite film, where the corresponding wave number of OH/NH significantly reduced showing a higher degree of rigidification of the polymer matrix which could be attributed to rapid loss of moisture and formation of new bonding (hydrogen, electrostatic, and/or Wander Waal's) between the Na-CMC and Eudragit L100 chains [102–104]. Furthermore, the hydrophobic domains too undergo dramatic changes where an additional weak CH stretching was observed (3100-3000 cm^{-1}) in addition to asymmetric and symmetric bands and a significant rise in C=O band (1715-1705 cm^{-1}). Owing to the fact that microwave interacts with moieties in volumetric manner, the rearrangement of hydrophobic moieties of Na-CMC and/or Eudragit L100 upon microwave treatment might be the reason for enhanced tensile strength observed (as discussed in earlier sections) and/or removal of absorbed water due to microwave heating which resulted in fluidization of the hydrophobic domains of the composite film formulations [70], while microwave treatment for 120 s (Figure 4(v)) does not produce any significant changes in the hydrophobic and hydrophilic domains of the composite films.

3.6. Thermal Analysis.
The DSC thermograms of the untreated F2 formulation and the F2 treated with microwave for 60 and 120 s are given in Figure 6. A significant increase in the transition as well as the enthalpy of the system was recorded for 60 s microwave-treated composite film while 120 s treatment remained statistically insignificantly different from the untreated formulation. An increase in the transition temperature depicts change in the physicochemical nature of the final product, and an increase in enthalpy
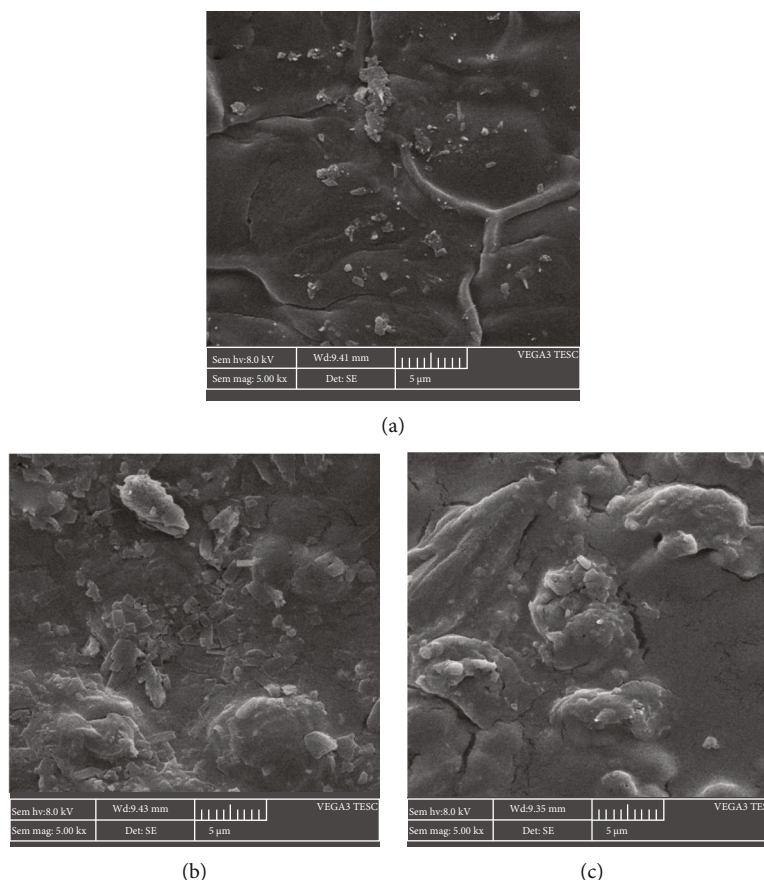
(a)

(b)

(c)

FIGURE 7: Surface morphology of untreated films: (a) F2, (b) F2 (60), and (c) F2 (120).

represents more compactness of the composite film [105]. Treatment of F2 formulation with microwave for 60 s ensured development of new bonds between the Na-CMC and Eudragit moiety which resulted in shifting of the transition temperature of the Na-CMC moiety from 273.16 ± 3.12 ˚C to 279.67 ± 2.6 with a significant increase in the corresponding enthalpy of up to 4 J/g (Figure 6(b), Student's t-test, $p < 0.05$) [106]. Similarly, the Eudragit moiety also under significant changes in the transition temperature as well as enthalpy value with microwave 60 s treatment where the corresponding peak temperature increased to 164.49 ± 2.31˚C from 157.49 ± 2.31˚C and enthalpy from 1.089 ± 0.13 J/g to 1.689 ± 0.13 J/g (Figure 6(b), Student's t-test, $p < 0.05$). Microwave 60 s treatment ensured excellent amalgamation of two polymers with each other by activating polar functional groups on both chains, which resulted in the formation of new bonds between both polymers hence changing the overall physicochemical properties of the composite matrix. While microwave 120 s treatment again did not produce any significant change either in Na-CMC moiety and Eudragit moiety (Figure 6(c), Student t-test, $p > 0.05$), rather, the corresponding peak temperature for Eudragit L100 disappeared which means 120 s treatment degraded the Eudragit moiety to enough extent that it is no more available in active state to form any additional/new bond with the Na-CMC moiety [107].

3.7. Scanning Electron Microscopy. The surface morphologies of composite polymeric films were observed under a scanning electron microscope (SEM) which is shown in Figure 7. The untreated microwave composite F2 formulation (Figure 7(a)) showed that it has much more homogenous organization of the matrix with most of the entities being present in a well-mixed amorphous state [96]. The microwave treatment for 60 s produced widespread crystalline structures on the surface of the composite film (Figure 7(b)) which could be the reason for delayed erosion, high tensile strength, and significantly different vibrational and thermal behavior of the composite film. On the other hand, the microwave treatment for 120 s and physical changes in the morphology of film surface are evident due to formation of cracks induced due to overheating process during 120 s treatment (Figure 7(c)) [108].

4. Conclusions

This paper focused on the development of natural and synthetic polymer composite films through microwave treatment. The physicochemical analysis results of untreated films helped in selection of optimized formulations for microwave treatment. The treatment of F1 and F2 formulations with microwave for 60 s significantly enhanced the mechanical properties of the films, with significant reduction

in water adsorption, water uptake, and percent erosion attributes. The mechanistic investigation revealed such enhancement resulted from rigidification of the hydrophilic and fluidization of the hydrophobic domains of the composite films with significant rise in the transition temperatures and enthalpy of the system enabling sustained release pattern of the drug from the composite, while 120 s treatment revealed degradation of the Eudragit moiety. Thus, microwave treatment of Na-CMC and Eudragit L100 at frequency of 2450 MHz at power of 350 Watts for 60 s time interval advocated the best and optimized treatment conditions for formulation of composite polymeric films using Na-CMC and Eudragit L100.

Data Availability

The data presented in the instinct manuscript are open to the public as per journal policy, and no restrictions to access are hereby placed by all authors.

Conflicts of Interest

The authors declare no conflict of interest, and all authors confirm agreement with the final statement.

Acknowledgments

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