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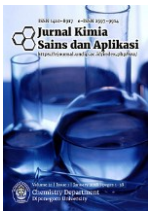

Synthesis and Characterization of Chitosan-HPMC-Methylparaben Hydrogel Film as an Antibacterial Wound Dressing Material

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Abstract

Chitosan-HPMC-methylparaben hydrogel film has been successfully synthesized. Chitosan, a key component with desirable characteristics for the production of hydrogel films, has some shortcomings that can be overcome by adding HPMC, a commonly used gelling agent that improves the physical properties of hydrogel films, and methylparaben, which enhances mechanical properties and antibacterial activity. The physical properties test of the hydrogel film, i.e., the solubility and swelling test, showed that the addition of HPMC reduced the solubility ratio and increased the swelling ratio in the chitosan hydrogel film. The addition of methylparaben relatively increases the antibacterial activity, tensile strength, and elongation of chitosan-HPMC-methylparaben hydrogel films. The chitosan-HPMC-methylparaben dry film exhibits a tensile strength of 19.56 MPa and an elongation of 1.98%. Surface morphological characterization of the hydrogel film through SEM tests showed that the chitosan-HPMC-methylparaben hydrogel film has granules on its surface, whereas the chitosan-HPMC hydrogel film has a smooth texture with a flat surface. The addition of methylparaben reduces the degree of crystallinity and produces an amorphous phase. The chitosan-HPMC-methylparaben hydrogel film shows strong activity against *S. aureus* and *E. coli*. Based on these results, the hydrogel film can be considered an antibacterial material for wound dressings, although elongation still needs to be improved.

1. Introduction

Skin is one of the largest organs, covering 15% of the body's surface area and receiving one-third of the blood supply in adults. The skin plays a major role as a barrier and protector from the environment. In addition, the skin functions in homeostasis and in the prevention of microbial invasion. This prevention requires that the wound be closed immediately to avoid damage to the skin's morphology or deeper tissues [1]. Wounds occur when the skin loses its tissues, resulting in damage to skin cells. Injuries can arise as a result of trauma caused by sharp objects or blunt objects, temperature changes, chemicals, explosions, electric shocks, or animal bites. Wound healing is closely related to the inflammatory process. The inflammatory process serves to protect damaged skin tissue, aiming to minimize the rate of damage and prevent bacterial infections [2]. Bacteria that cause skin infections include *Staphylococcus aureus*, *Staphylococcus epidermidis*, *Escherichia coli*, and *Pseudomonas aeruginosa*.

Skin damage caused by *Staphylococcus aureus* and *Escherichia coli* can be minimized with wound dressings [3]. Wound dressings aim to stop bleeding and protect the wound from environmental irritants such as water, bacteria, and electrolyte disturbances. Hydrogel is considered an ideal candidate for wound dressing due to its 3D structure, high permeability, superior biocompatibility, and ability to provide a moist environment that favors wound healing, thereby overcoming the disadvantages of traditional dressings [4,

5]. The hydrogel can absorb large amounts of biological fluids, making it highly effective as a wound dressing [6].

One ingredient with good absorption potential is chitosan, a copolymer of 2-glucosamine and N-acetyl-2-glucosamine, an environmentally friendly derivative of chitin [7]. It is supported by chitosan characteristics, including high resistance, biodegradability, good biocompatibility, low toxicity, liquid absorption ability, and antibacterial activity [1]. However, chitosan has drawbacks, including poor mechanical and swelling properties and low antibacterial activity against bacterial growth. To address these weaknesses, additional agents are needed, such as HPMC (hydroxypropyl methylcellulose) to improve mechanical and swelling properties and methylparaben to increase antibacterial activity. The addition of HPMC and methylparaben to chitosan hydrogel can occur via hydrogen bonding and entanglement processes. The entanglement of HPMC and chitosan molecular chains contributes to gel formation and enhances the network structure and physical integrity of the hydrogel formulation by connecting the polymer chains, thereby improving mechanical properties [8]. Chitosan, through the entanglement process, has a structure that contributes to good mechanical properties [9], affects its compatibility [10], and can significantly inhibit bacterial growth [11].

HPMC is a non-ionic derivative of cellulose with large hydrophobic segments, which results in strong surface activity. Previous studies have shown that using HPMC as a thickening agent yields stable formulations [12]. HPMC, formulated with sodium alginate, can form a polyelectrolyte complex used in drug delivery [13]. In general, HPMC has been used extensively in a variety of research applications as an emulsifier, suspension agent, and stabilizer for topical formulations such as gels and ointments. HPMC has also been researched to increase the tensile strength of films [14]. Therefore, this study aims to utilize HPMC in film form to investigate how its effects in film form, combined with chitosan, can expand its application potential.

Methylparaben is a preservative commonly used in cosmetic formulations due to its antibacterial and antifungal properties. Parabens are more effective against gram-positive bacteria (*Staphylococcus aureus*) compared to gram-negative bacteria (*Escherichia coli*) [15]. The addition of methylparaben as a preservative and antimicrobial agent aims to improve the quality of the hydrogel film produced. Methylparaben is commonly used as an antimicrobial preservative in cosmetic products and topical preparations at concentrations ranging from 0.02% to 0.3% [16]. Therefore, this study aims to leverage the characteristics of methylparaben to develop wound-dressing materials that increase the antibacterial activity of hydrogel films.

Based on the problems and background explained above, a study was conducted to synthesize chitosan hydrogel films incorporating HPMC and methylparaben as antibacterial agents. Chitosan hydrogel films entangled with HPMC and methylparaben were analyzed using FTIR to determine functional group structure, SEM for morphological analysis, and solubility, swelling, and tensile strength tests to evaluate the mechanical properties of the hydrogel films. The antibacterial activity of chitosan hydrogel films entangled with HPMC and Methylparaben was tested against *Staphylococcus aureus* (*S. aureus*) and *Escherichia coli* (*E. coli*) to determine their effectiveness.

2. Experimental

2.1. Materials and Equipment

The materials used in this study included food-grade chitosan supplied by CV ChiMultiguna Indonesia, glacial acetic acid (Merck), technical-grade hydroxypropyl methylcellulose (HPMC), technical-grade methylparaben, and distilled water. The equipment consisted of standard laboratory glassware and a magnetic stirrer. The characterization instruments included an ATR-FTIR spectrometer (Agilent Cary 630), a scanning electron microscope (SEM; Zeiss DSM 960), and an X-ray diffractometer (XRD; Rigaku).

2.2. Synthesis of Chitosan-HPMC-Methylparaben Hydrogel Film

Chitosan–HPMC–methylparaben hydrogel films were prepared according to the method described by Susilowati *et al.* [17] with slight modifications. First, a 1.5% (w/v) chitosan solution was prepared by dissolving 1.5 g of chitosan in 1% (v/v) acetic acid solution under continuous stirring until a homogeneous solution was obtained. Separately, a 1% (w/v) HPMC solution was prepared by dissolving 1 g of HPMC in distilled water and stirring at 60°C until completely dissolved. The chitosan and HPMC solutions were then mixed to obtain HPMC concentrations of 2.9%, 4.8%, and 6.7% (w/v), followed by continuous stirring until homogeneous. Next, a 0.1% (w/v) methylparaben solution was prepared by dissolving 0.1 g of methylparaben in 100 mL of distilled water under stirring. This solution was subsequently added to the chitosan–HPMC mixture to obtain final methylparaben concentrations of 0.02%, 0.04%, and 0.06% (w/v). The resulting mixtures were cast into molds and dried in an oven at 60°C for 22 h to obtain hydrogel films.

2.3. Solubility Test

Chitosan–HPMC hydrogel films containing HPMC concentrations of 2.9%, 4.8%, and 6.7% (w/v), as well as chitosan–HPMC–methylparaben hydrogel films containing methylparaben concentrations of 0.02%, 0.04%, and 0.06% (w/v), were cut into 3 × 3 cm specimens. Each specimen was weighed to obtain its initial dry weight (W_0). The specimens were then immersed in 30 mL of distilled water for 30 min. After immersion, the insoluble portions of the hydrogel films were removed, gently blotted with filter paper to eliminate excess surface water, and dried in an oven at 100°C for 10 min. The dried specimens were reweighed to determine the final dry weight (W_1). The solubility (S) of each hydrogel film was calculated using Equation (1).

$$S = \frac{W_0 - W_1}{W_0} \times 100\% \quad (1)$$

2.4. Swelling Test

Chitosan–HPMC hydrogel films containing HPMC concentrations of 2.9%, 4.8%, and 6.7% (w/v), as well as chitosan–HPMC–methylparaben hydrogel films containing methylparaben concentrations of 0.02%, 0.04%, and 0.06% (w/v), were cut into 3 × 3 cm specimens. Each specimen was weighed to determine its initial dry weight (W_0). The specimens were then immersed in 30 mL of distilled water for 2 h. After immersion, the films were carefully removed, gently blotted with tissue paper to remove excess surface water, and immediately weighed to obtain the swollen weight (W_1). The swelling ratio (S_w) was calculated using Equation (2).

$$S_w = \frac{W_1 - W_0}{W_0} \times 100\% \quad (2)$$

2.5. FTIR Analysis

Fourier-transform infrared (FTIR) spectroscopy was performed to identify the functional groups present in the prepared chitosan–HPMC–methylparaben hydrogel films. The analysis was carried out using an attenuated total reflectance Fourier-transform infrared (ATR-FTIR) spectrophotometer (Agilent Cary 630). Prior to analysis, the hydrogel films were cut into 3 × 3 cm specimens and placed directly on the ATR crystal. The FTIR spectra were recorded as absorbance versus wavenumber (cm^{-1}), and the characteristic absorption bands were analyzed to identify the functional groups and possible interactions among the hydrogel components.

2.6. XRD Analysis

X-ray diffraction (XRD) analysis was performed to evaluate the crystalline structure of the prepared chitosan–HPMC–methylparaben hydrogel films. Prior to analysis, the hydrogel films were cut into suitable specimens and mounted on the sample holder of an X-ray diffractometer (Rigaku). The diffraction patterns were recorded over a 2θ range of 5°–65° using Cu Kα radiation under the instrument's standard operating conditions. The obtained diffractograms were analyzed to identify the crystalline characteristics and structural changes of the hydrogel films resulting from the incorporation of HPMC and methylparaben.

2.7. SEM Analysis

Scanning electron microscopy (SEM) was performed to examine the surface morphology of the prepared chitosan–HPMC–methylparaben hydrogel films. Prior to analysis, the hydrogel film specimens were mounted on aluminum stubs using double-sided conductive adhesive tape [18] and sputter-coated with a thin layer of gold under vacuum to enhance surface conductivity. The coated specimens were then examined using a Zeiss DSM 960 scanning electron microscope. Surface micrographs were acquired and analyzed to evaluate the morphological characteristics of the hydrogel films.

2.8. Tensile Strength Test

The tensile properties of the prepared chitosan–HPMC and chitosan–HPMC–methylparaben hydrogel films were evaluated in accordance with ISO 527-3:1996. The hydrogel films were cut into rectangular specimens measuring 15 cm in length and 2 cm in width. Each specimen was mounted on a Universal Tensile Testing Machine, and a uniaxial tensile load was applied until the specimen fractured [19].

2.9. Antibacterial Activity Test

The antibacterial activity of the prepared chitosan–HPMC and chitosan–HPMC–methylparaben hydrogel films was evaluated against *S. aureus* and *E. coli* using the disc diffusion method. The hydrogel films were cut into circular discs using a sterile paper punch. A chloramphenicol disc was used as the positive control, while a blank disc impregnated with dimethyl sulfoxide (DMSO) served as the negative control. The sample and control discs were placed on Mueller–Hinton agar (MHA) plates that had been inoculated with the test bacteria. The plates were incubated at 37°C for 20–24 h. After incubation, the diameter of the bacterial growth inhibition zone surrounding each disc was measured using a digital caliper.

3. Results and Discussion

3.1. Hydrogel Films

Hydrogel films were prepared in three formulations: chitosan, chitosan-HPMC, and chitosan-HPMC-methylparaben. The films formed thin, whitish-yellow sheets. As shown in Figure 1, the chitosan hydrogel film is yellowish-white with a smooth texture, while the whiter chitosan-HPMC is almost transparent with a fine texture, similar to chitosan-HPMC-methylparaben (0.06%). The addition of methylparabens, which serve as antibacterial and softening agents, causes the yellowish color of chitosan to fade because of the colorless solution formed by HPMC and methylparaben. The interaction between chitosan and HPMC produced a homogeneous hydrogel film through polymer entanglement and hydrogen bonding between the $-NH_2$ groups of chitosan and the -OH groups of HPMC [20]. HPMC forms a strong hydrogen bond with chitosan [21].

3.2. Solubility of Hydrogel Films

The solubility of hydrogel films is influenced by the composition and concentration of their constituent polymers, as these factors determine the extent of intermolecular interactions and the stability of the polymer network [22]. As shown in Table 1, increasing the HPMC concentration significantly decreased the solubility of the chitosan hydrogel films. The solubility values decreased from 89.69% for the chitosan film without HPMC (0%) to 7.72%, 5.97%, and 3.06% for films containing HPMC at 2.9%, 4.8%, and 6.7% (w/v), respectively.

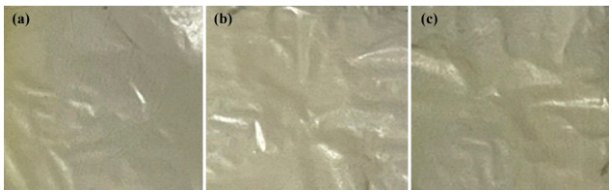

Figure 1. Hydrogel films: (a) chitosan, (b) chitosan-HPMC, (c) chitosan-HPMC-methylparaben

Table 1. Effect of HPMC concentration on the solubility of hydrogel films

Hydrogel film	Solubility (%)
Chitosan	89.69 ± 0.72
Chitosan-HPMC(2.9%)	7.72 ± 0.51
Chitosan-HPMC(4.8%)	5.97 ± 0.30
Chitosan-HPMC(6.7%)	3.06 ± 0.05
Chitosan-HPMC(6.7%)-Methylparaben(0.06%)	10.37 ± 0.10

The high solubility of the pure chitosan film can be attributed to the presence of protonated amino groups associated with acetate ions, which increase the hydrophilicity of the polymer and facilitate chain dispersion upon immersion in water. As the polymer chains become hydrated, they are more readily released into the surrounding medium, resulting in a high degree of solubility. In contrast, the incorporation of HPMC promotes the formation of intermolecular hydrogen bonds and physical chain entanglements between chitosan and HPMC molecules. These interactions generate a more compact and stable polymer network, thereby restricting polymer chain mobility and reducing chain dissolution in water. Consequently, increasing the HPMC concentration enhances the density of intermolecular interactions, making the hydrogel film more resistant to dissolution and resulting in progressively lower solubility [23].

As shown in Table 1, the solubility decreased from 89.69% for pure chitosan to 3.06% after HPMC incorporation, then increased to 10.37% following methylparaben addition. Methylparaben increases solubility by weakening hydrogen bonds and polymer chain entanglements [24].

3.3. Swelling Ratio of Hydrogel Films

The results of the swelling test on chitosan hydrogel films with varying HPMC concentrations are listed in Table 2. The addition of HPMC results in a homogeneous interaction with chitosan, as evidenced by the observed increase in the degree of swelling. The interaction between the amine group ($-NH_2$) of chitosan and the hydroxyl group (-OH) of HPMC forms a hydrogen bond and entanglement, which affects the swelling of the hydrogel [25]. The bonds formed during the reaction occur as the distance between molecules increases and the hydrogen bonds of the reaction weaken. Swelling in the chitosan hydrogel film with HPMC occurs when the hydration of the hydrophobic groups forms a swelling skeleton over the soaking period [26]. HPMC, as a non-ionic cellulose derivative containing -OH group that promotes polymer hydration, explains the increased degree of swelling at higher HPMC concentrations [27].

Table 2. Effect of HPMC concentration on the swelling ratio of hydrogel films

Hydrogel film	Swelling (%)
Chitosan	9.47 ± 1.50
Chitosan-HPMC(2.9%)	817.34 ± 32.92
Chitosan-HPMC(4.8%)	971.24 ± 14.57
Chitosan-HPMC(6.7%)	1005.53 ± 5.29
Chitosan-HPMC(6.7%)-Methylparaben(0.06%)	763.62 ± 6.99

Swelling tests were performed on chitosan, chitosan–HPMC, and chitosan–HPMC–methylparaben hydrogel films, and the results are presented in Table 2. The swelling ratio increased markedly with increasing HPMC concentration. Pure chitosan exhibited the lowest swelling ratio (9.47%), whereas chitosan–HPMC hydrogel films containing 2.9%, 4.8%, and 6.7% HPMC showed swelling ratios of 817.34%, 971.24%, and 1005.53%, respectively. The highest swelling ratio was therefore observed for the chitosan–HPMC hydrogel film containing 6.7% HPMC.

The increase in the swelling ratio with increasing HPMC concentration is attributed to the greater number of hydrophilic hydroxyl groups introduced by HPMC, which readily interact with water molecules through hydrogen bonding. Compared with chitosan, HPMC has a higher affinity for water; therefore, increasing its proportion enhances the hydrophilicity of the polymer matrix, facilitating water diffusion into the hydrogel network and resulting in greater expansion of the film structure. In addition, the interactions between chitosan and HPMC are primarily governed by hydrogen bonding and physical chain entanglement, producing a polymer network that remains sufficiently flexible to absorb a large amount of water [28]. These findings are consistent with previous studies [29, 30], which reported that hydrogel films with good swelling properties are capable of absorbing water several times their dry weight.

3.4. Surface Morphology of Hydrogel Films

The surface morphology of the hydrogel films was examined using SEM. The SEM micrographs of chitosan, chitosan–HPMC, and chitosan–HPMC–methylparaben hydrogel films are presented in Figure 2. The SEM analysis revealed that the pure chitosan hydrogel film exhibited a smooth and homogeneous surface. The incorporation of 6.7% (w/v) HPMC produced a hydrogel film with a similarly smooth and compact surface, whereas the addition of 0.06% (w/v) methylparaben resulted in a rougher and more heterogeneous surface characterized by the presence of fine particles.

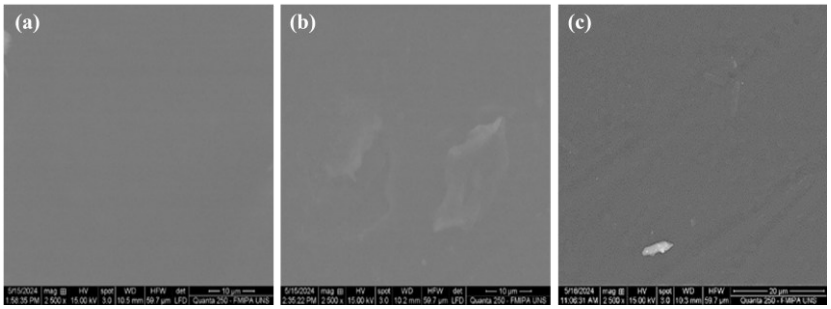

Figure 2. SEM micrographs of (a) chitosan, (b) chitosan–HPMC, and (c) chitosan–HPMC–methylparaben hydrogel films

The observed morphological differences indicate that the interactions among the film components influence the surface structure of the hydrogel films. The smooth and pore-free morphology of the chitosan–HPMC hydrogel film is consistent with previous studies, which reported that hydrogen bonding and polymer chain entanglement between chitosan and HPMC contribute to the formation of a dense and homogeneous film structure without visible pores or cracks [31]. In addition, electrostatic interactions, hydrogen bonding, and hydrophobic interactions between chitosan and HPMC further contribute to the structural organization and stability of the hydrogel film [32, 33].

3.5. FTIR Analysis of Hydrogel Films

FTIR spectroscopy was performed to identify the functional groups and investigate the interactions among the components of chitosan, chitosan–HPMC, and chitosan–HPMC–methylparaben hydrogel films. The FTIR spectra are presented in Figure 3. The FTIR spectrum of the chitosan hydrogel film (Figure 3a) exhibited characteristic absorption bands at 3192 cm^{-1} corresponding to O–H stretching, 2920 cm^{-1} to CH_2 stretching, 1543 cm^{-1} to NH_2 bending, 1401 cm^{-1} to C–H bending, 1341 cm^{-1} to C–OH stretching, 1254 cm^{-1} to C–O stretching, 1151 cm^{-1} to C–N stretching, 1019 cm^{-1} to O–H bending, and 896 cm^{-1} to glycosidic bond vibrations [25, 34, 35, 36, 37].

The FTIR spectrum of the chitosan–HPMC hydrogel film (Figure 3b) showed absorption bands at 3235 cm^{-1} (O–H stretching), 2920 cm^{-1} (CH_2 stretching), 1543 cm^{-1} (NH_2 bending), 1407 cm^{-1} (C–H bending), 1341 cm^{-1} (C– OH stretching), 1261 cm^{-1} (C–O stretching), 1151 cm^{-1} (C–N stretching), 1019 cm^{-1} (O–H bending), and 896 cm^{-1} (glycosidic bond vibrations) [31, 34, 35, 36, 37, 38, 39].

The FTIR spectrum of the chitosan–HPMC–methylparaben hydrogel film (Figure 3c) exhibited absorption bands at 3274 cm^{-1} (O–H stretching), 2920 cm^{-1} (CH_2 stretching), 1543 cm^{-1} (NH_2 bending), 1407 cm^{-1} (C–H bending), 1341 cm^{-1} (C–OH stretching), 1261 cm^{-1} (C–O stretching), 1151 cm^{-1} (C–N stretching), 1019 cm^{-1} (O–H bending), and 836 cm^{-1} corresponding to the aromatic ring vibration of methylparaben [40, 41, 42].

As summarized in Table 3, the shift of the O–H stretching band from 3192 cm^{-1} in the chitosan film to 3235 cm^{-1} in the chitosan–HPMC film and subsequently to 3274 cm^{-1} in the chitosan–HPMC–methylparaben film, accompanied by peak broadening, suggests enhanced intermolecular hydrogen-bonding interactions among chitosan, HPMC, and methylparaben [37]. The O– H and NH_2 functional groups facilitate hydrogen-bond formation between chitosan and HPMC, contributing to the stability of the hydrogel network. The characteristic aromatic band at 836 cm^{-1}, assigned to the benzene ring of methylparaben, confirms its successful incorporation into the chitosan–HPMC hydrogel matrix through intermolecular interactions [43].

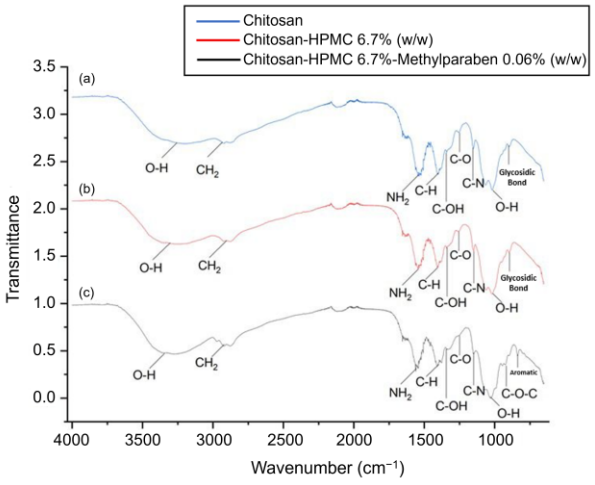

Figure 3. FTIR spectra of chitosan and hydrogel films

Table 3. Characteristic FTIR absorption bands of hydrogel films

Wavenumber (cm^{-1})			Functional group/ Vibration	Reference
Chitosan	Chitosan-HPMC	Chitosan-HPMC-methylparaben		
3192	3235	3274	O-H stretching	[32]
1401	1407	1407	C-H bending	[37]
1254	1261	1261	C-O stretching	[11]
896	896	-	Glycosidic bond vibration	[44]
-	-	836	Aromatic ring vibration	[45]

3.6. XRD Analysis of Hydrogel Films

XRD analysis was performed to investigate the crystalline structure and degree of crystallinity of chitosan, chitosan–HPMC, and chitosan–HPMC–methylparaben hydrogel films. The XRD patterns are presented in Figure 4. The chitosan hydrogel film (Figure 4a) exhibited a broad diffraction peak, indicating a predominantly amorphous structure [44]. The chitosan–HPMC film (Figure 4b) showed a similar pattern with lower peak intensity, suggesting that HPMC incorporation promoted hydrogen bonding with chitosan and reduced the structural order of the polymer matrix [16]. In addition, a characteristic diffraction peak attributed to HPMC was observed in the chitosan–HPMC hydrogel film [23].

The XRD pattern of the chitosan–HPMC–methylparaben hydrogel film (Figure 4c) exhibited a predominantly amorphous halo without distinct crystalline peaks, indicating that methylparaben further disrupted the ordered arrangement of the polymer chains. Consequently, the film became less crystalline due to increased intermolecular interactions and reduced molecular packing.

The degree of crystallinity was calculated from the ratio of the crystalline area to the total area of the diffractogram [46], and the results are presented in Table 6. The degree of crystallinity decreased from 27.01% in the chitosan hydrogel film to 17.69% after incorporating HPMC, indicating that HPMC reduced the crystalline regions of the polymer matrix. The degree of crystallinity of the chitosan–HPMC–methylparaben hydrogel film could not be determined because the diffraction pattern showed a predominantly amorphous profile with no distinguishable crystalline peaks.

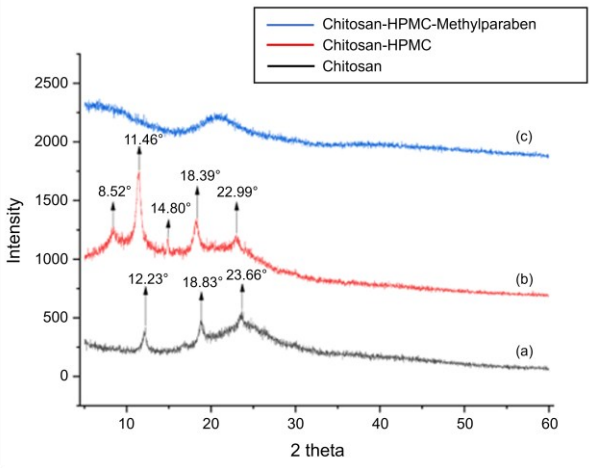

Figure 4. XRD pattern of hydrogel films

Table 6. Degree of crystallinity of the hydrogel films

Hydrogel film	Degree of crystallinity (%)
Chitosan	27.01
Chitosan-HPMC	17.69
Chitosan-HPMC-Methylparaben	-

3.7. Tensile Strength and Elongation of Hydrogel Films

The mechanical properties of chitosan, chitosan–HPMC, and chitosan–HPMC–methylparaben hydrogel films were evaluated by measuring their tensile strength and elongation at break, as presented in Table 7 [44]. Pure chitosan exhibited the highest tensile strength (26.06 ± 4.42 MPa). The incorporation of HPMC reduced the tensile strength to 16.83 ± 3.03 MPa, which may be attributed to hydrogen-bonding interactions between chitosan and HPMC that altered the intermolecular interactions within the polymer matrix [47, 48]. The addition of methylparaben slightly increased the tensile strength to 19.65 ± 1.38 MPa, suggesting that methylparaben contributed to improving the structural integrity of the hydrogel film through intermolecular interactions [49].

The elongation at break decreased from 2.49 ± 0.33% for the chitosan film to 1.51 ± 0.59% after HPMC incorporation, indicating reduced flexibility. The addition of methylparaben slightly increased the elongation to 1.98 ± 0.24%, indicating a modest improvement in film flexibility. However, the elongation values remained relatively low, indicating that the hydrogel films remained brittle under dry conditions [16].

3.8. Antibacterial Activity of Hydrogel Films

The antibacterial activity of chitosan, chitosan–HPMC, and chitosan–HPMC–methylparaben hydrogel films containing different concentrations of methylparaben (0.02%, 0.04%, and 0.06%), designated as S1, S2, S3, S4, and S5, was evaluated against *S. aureus* and *E. coli* using the disc diffusion method. The antibacterial activity was determined by measuring the diameter of the inhibition zone, where a larger inhibition zone indicates stronger antibacterial activity. The inhibition zones are presented in Figure 5.

As shown in Table 8, antibacterial activity against *S. aureus* was observed only for chitosan–HPMC–methylparaben hydrogel films containing 0.04% (S4) and 0.06% (S5) methylparaben, with inhibition zones of 11.54 mm and 12.25 mm, respectively. In contrast, the chitosan hydrogel film (S1), chitosan–HPMC hydrogel film (S2), and chitosan–HPMC–methylparaben hydrogel film containing 0.02% methylparaben (S3) did not exhibit significant antibacterial activity against *S. aureus*. These findings are consistent with previous studies reporting that the antibacterial activity of chitosan is influenced by its molecular weight [45].

Table 7. Mechanical properties of hydrogel films

Hydrogel film	Tensile strength (MPa)	Elongation (%)
Chitosan	26.06 ± 4.42	2.49 ± 0.33
Chitosan-HPMC	16.83 ± 3.03	1.51 ± 0.59
Chitosan-HPMC-Methylparaben	19.65 ± 1.38	1.98 ± 0.24

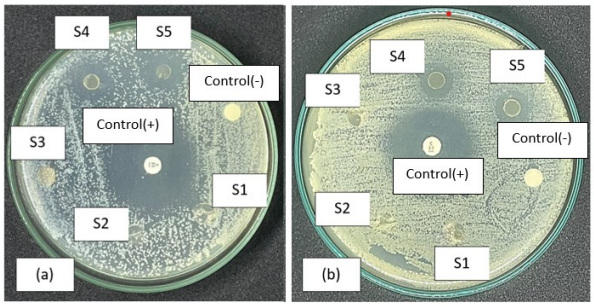

Figure 5. Inhibition zones against (a) *S. aureus* and (b) *E. coli*. S1 = chitosan; S2 = chitosan–HPMC; S3 = chitosan–HPMC–methylparaben (0.02%, w/v); S4 = chitosan–HPMC–methylparaben (0.04%, w/v); S5 = chitosan–HPMC–methylparaben (0.06%, w/v)

Table 8. Inhibition zones of hydrogel films against *S. aureus* and *E. coli*

Hydrogel film	*S. aureus* (mm)	*E. coli* (mm)
Chitosan	-	-
Chitosan-HPMC(6.7%)	-	-
Chitosan-HPMC(6.7%)-Methylparaben(0.02%) (w/v)	-	-
Chitosan-HPMC 6.7% Methylparaben 0.04% (w/v)	11.54 ± 0.81	11.29 ± 0.84
Chitosan-HPMC 6.7% Methylparaben 0.06% (w/v)	12.25 ± 0.70	12.03 ± 0.69
Positive control	31.54 ± 0.72	30.54 ± 0.68
Negative control	-	-

Chitosan and chitosan–HPMC hydrogel films did not exhibit antibacterial activity against either *S. aureus* or *E. coli*. This may be attributed to the limited diffusion of chitosan from the film matrix into the agar medium, thereby reducing its antibacterial effect in the disc diffusion assay. A similar result was observed for the chitosan–HPMC–methylparaben hydrogel film containing 0.02% (w/v) methylparaben (S3), which did not produce an inhibition zone.

In contrast, chitosan–HPMC–methylparaben hydrogel films containing 0.04% (S4) and 0.06% (S5) methylparaben exhibited antibacterial activity against *E. coli*, with inhibition zones of 11.29 mm and 12.03 mm, respectively. The increased antibacterial activity with increasing methylparaben concentration indicates a concentration-dependent antibacterial effect. Based on the results presented in Table 8, the antibacterial activity of the hydrogel films was primarily attributed to methylparaben. Moreover, the inhibition zones against *S. aureus* were slightly larger than those against *E. coli*, indicating that methylparaben was more effective against Gram-positive bacteria than Gram-negative bacteria. The antibacterial activity of methylparaben has been associated with its ability to disrupt bacterial membrane transport and interfere with DNA and RNA synthesis.

4. Conclusion

Chitosan–HPMC–methylparaben hydrogel films were successfully prepared. The incorporation of HPMC into chitosan hydrogel films decreased solubility, increased the swelling ratio, and reduced both tensile strength and elongation due to the formation of hydrogen bonds and polymer chain entanglements between chitosan and HPMC. In contrast, the incorporation of methylparaben increased the solubility and the mechanical properties of the hydrogel films, as indicated by the increased tensile strength and elongation, while reducing the swelling ratio. The chitosan–HPMC–methylparaben hydrogel film exhibited a tensile strength of 19.65 MPa, an elongation at break of 1.98%, a swelling ratio of 763.62%, and a solubility of 10.37%. FTIR analysis confirmed the presence of O–H, C–H, C–N, and aromatic functional groups, while SEM analysis revealed a rougher surface with fine particle formation following the incorporation of methylparaben. The chitosan–HPMC–methylparaben hydrogel film exhibited antibacterial activity against *S. aureus* and *E. coli*, with inhibition zones of 12.25 ± 0.70 mm and 12.03 ± 0.69 mm, respectively. The antibacterial activity was primarily attributed to the presence of methylparaben.

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References

[1] R. Jayakumar, M. Prabaharan, P. T. Sudheesh Kumar, S. V. Nair, H. Tamura, Biomaterials based on chitin and chitosan in wound dressing applications, *Biotechnology Advances*, 29, 3, (2011), 322-337 https://doi.org/10.1016/j.biotechadv.2011.01.005

[2] Sibusiso Alven, Blessing Atim Aderibigbe, Chitosan and Cellulose-Based Hydrogels for Wound Management, *International Journal of Molecular Sciences*, 21, 24, (2020), 9656 https://doi.org/10.3390/ijms21249656

[3] Wesley A. Volk, Margaret F. Wheeler, *Basic Microbiology*, 5th ed., Harper & Row, 1984,

[4] Yawen Xu, Yong Xu, Bo Bi, Mengjie Hou, Lin Yao, Qiran Du, Aijuan He, Yu Liu, Chunlei Miao, Xiaoqin Liang, Xulin Jiang, Guangdong Zhou, Yilin Cao, A moldable thermosensitive hydroxypropyl chitin hydrogel for 3D cartilage regeneration *in vitro* and *in vivo*, *Acta Biomaterialia*, 108, (2020), 87-96 https://doi.org/10.1016/j.actbio.2020.03.039

[5] Yu Shrike Zhang, Ali Khademhosseini, Advances in engineering hydrogels, *Science*, 356, 6337, (2017), https://doi.org/10.1126/science.aaf3627

[6] Yi Hou, Nan Jiang, Dan Sun, Yiping Wang, Xianchun Chen, Songsong Zhu, Li Zhang, A fast UV-curable PU-PAAm hydrogel with mechanical flexibility and self-adhesion for wound healing, *RSC Advances*, 10, 9, (2020), 4907-4915 https://doi.org/10.1039/C9RA10666A

[7] Chunyu Chang, Si Chen, Lina Zhang, Novel hydrogels prepared *via* direct dissolution of chitin at

low temperature: structure and biocompatibility, *Journal of Materials Chemistry*, 21, 11, (2011), 3865-3871 https://doi.org/10.1039/c0jm03075a

[8] Yanni Gao, Yuzhou Zhao, Ting Wang, Preparation and Characterization of Chitosan/Hydroxypropyl Methylcellulose Temperature-Sensitive Hydrogel Containing Inorganic Salts for Forest Fire Suppression, *Gels*, 10, 6, (2024), 390 https://doi.org/10.3390/gels10060390

[9] Yaqian Zhang, Min Zhang, Haoyang Jiang, Jinli Shi, Feibo Li, Yanhong Xia, Gongzheng Zhang, Huanjun Li, Bio-inspired layered chitosan/graphene oxide nanocomposite hydrogels with high strength and pH-driven shape memory effect, *Carbohydrate Polymers*, 177, (2017), 116-125 https://doi.org/10.1016/j.carbpol.2017.08.106

[10] Haihong Li, Xiaochen Gao, Yan Wang, Xiaobo Zhang, Zhiwei Tong, Comparison of chitosan/starch composite film properties before and after cross-linking, *International Journal of Biological Macromolecules*, 52, (2013), 275-279 https://doi.org/10.1016/j.ijbiomac.2012.10.016

[11] Bin Li, Chang-Lin Shan, Qing Zhou, Yuan Fang, Yang-Li Wang, Fei Xu, Li-Rong Han, Muhammad Ibrahim, Long-Biao Guo, Guan-Lin Xie, Guo-Chang Sun, Synthesis, Characterization, and Antibacterial Activity of Cross-Linked Chitosan-Glutaraldehyde, *Marine Drugs*, 11, 5, (2013), 1534-1552 https://doi.org/10.3390/md11051534

[12] Lusi Nurdianti, Evaluasi Sediaan Emulgel Anti Jerawat Tea Tree (*Melaleuca alternifolia*) Oil sengan Menggunakan HPMC sebagai *Gelling Agent*, *Journal of Pharmacopolium*, 1, 1, (2018), 23-31

[13] Jing-Ting Lin, Yi-Chan Chiang, Po-Hsien Li, Po-Yuan Chiang, Structural and Release Properties of Combined Curcumin Controlled-Release Tablets Formulated with Chitosan/Sodium Alginate/HPMC, *Foods*, 13, 13, (2024), https://doi.org/10.3390/foods13132022

[14] Shujuan Jiang, Mengyuan Zhang, Shengnan Jiang, Yanfeng Tuo, Fang Qian, Guangqing Mu, Transglutaminase and hydroxypropyl methyl cellulose enhance mechanical properties of whey protein concentrate film, *International Journal of Food Science and Technology*, 57, 8, (2022), 5472-5478 https://doi.org/10.1111/ijfs.15880

[15] João Lincho, Rui C. Martins, João Gomes, Paraben Compounds—Part I: An Overview of Their Characteristics, Detection, and Impacts, *Applied Sciences*, 11, 5, (2021), 2307 https://doi.org/10.3390/app11052307

[16] M. G. Soni, S. L. Taylor, N. A. Greenberg, G. A. Burdock, Evaluation of the health aspects of methyl paraben: a review of the published literature, *Food and Chemical Toxicology*, 40, 10, (2002), 1335-1373 https://doi.org/10.1016/S0278-6915(02)00107-2

[17] Endang Susilowati, Sri Retno Dwi Ariani, Lina Mahardiani, Luthfia Izzati, Synthesis and Characterization Chitosan Film with Silver Nanoparticle Addition as a Multiresistant Antibacteria Material, *JKPK (Jurnal Kimia dan Pendidikan Kimia)*, 6, 3, (2021), 371-383 https://doi.org/10.20961/jkpk.v6i3.57101

[18] Dawei Zhang, Shuai Yang, Yuanqing Chen, Siyu Liu, Hongtao Zhao, Jiyou Gu, ^{60}Co γ-ray Irradiation Crosslinking of Chitosan/Graphene Oxide Composite Film: Swelling, Thermal Stability, Mechanical, and Antibacterial Properties, *Polymers*, 10, 3, (2018), 294 https://doi.org/10.3390/polym10030294

[19] Nurul Hasanah, Alimin Mahyudin, Pengaruh Variasi Massa Gliserol Terhadap Sifat Mekanik Film Plastik Pati Umbi Talas Berpenguat Nano Serat Pinang, *Jurnal Fisika Unand*, 11, 2, (2022), 194-200

[20] Guanyu Cai, Jing Sun, Fei Kang, Qilin Lv, Jin Liu, Jie Wang, Zideng Gao, Xueqin Ren, Chitosan Gel Hydroxypropyl Methylcellulose Membranes: A Novel Approach for the Remediation of Cadmium in Aqueous Solutions and Soils, *Coatings*, 14, 4, (2024), 421 https://doi.org/10.3390/coatings14040421

[21] A. Pawlak, M. Mucha, Thermogravimetric and FTIR studies of chitosan blends, *Thermochimica Acta*, 396, 1-2, (2003), 153-166 https://doi.org/10.1016/S0040-6031(02)00523-3

[22] Heidi Birch, Aaron D. Redman, Daniel J. Letinski, Delina Y. Lyon, Philipp Mayer, Determining the water solubility of difficult-to-test substances: A tutorial review, *Analytica Chimica Acta*, 1086, (2019), 16-28 https://doi.org/10.1016/j.aca.2019.07.034

[23] Jefferson Rotta, Edson Minatti, Pedro Luiz Manique Barreto, Determination of structural and mechanical properties, diffractometry, and thermal analysis of chitosan and hydroxypropylmethylcellulose films plasticized with sorbitol, *Food Science and Technology*, 31, 2, (2011), 450-455 https://doi.org/10.1590/S0101-20612011000200026

[24] K. S. Alexander, B. Laprade, J. W. Mauger, A. N. Paruta, Thermodynamics of Aqueous Solutions of Parabens, *Journal of Pharmaceutical Sciences*, 67, 5, (1978), 624-627 https://doi.org/10.1002/jps.2600670513

[25] Jianhui Li, Shaoling Zhuang, Antibacterial activity of chitosan and its derivatives and their interaction mechanism with bacteria: Current state and perspectives, *European Polymer Journal*, 138, (2020), 109984 https://doi.org/10.1016/j.eurpolymj.2020.109984

[26] Hafiz U. Ali, Dure N. Iqbal, Munawar Iqbal, Safa Ezzine, Aysha Arshad, Rabia Zeeshan, Aqif A. Chaudhry, Samar Z. Alshawwa, Arif Nazir, Ather F. Khan, HPMC crosslinked chitosan/hydroxyapatite scaffolds containing Lemongrass oil for potential bone tissue engineering applications, *Arabian Journal of Chemistry*, 15, 7, (2022), 103850 https://doi.org/10.1016/j.arabjc.2022.103850

[27] Evdokia Vassiliadi, Evgenia Mitsou, Spyridon Avramiotis, Christos L. Chochos, Franz Pirolt, Martin Medebach, Otto Glatter, Aristotelis Xenakis, Maria Zoumpanioti, Structural Study of (Hydroxypropyl)Methyl Cellulose Microemulsion-Based Gels Used for Biocompatible Encapsulations, *Nanomaterials*, 10, 11, (2020), 2204 https://doi.org/10.3390/nano10112204

[28] N. Vishal Gupta, H. G. Shivakumar, Investigation of Swelling Behavior and Mechanical Properties of a pH-Sensitive Superporous Hydrogel Composite, *Iranian Journal of Pharmaceutical Research*, 11, 2, (2012), 481-493

[29] Waseeq Ur Rehman, Muhammad Asim, Shah Hussain, Shahid Ali Khan, Sher Bahadar Khan,

Hydrogel: A Promising Material in Pharmaceutics, *Current Pharmaceutical Design*, 26, 45, (2020), 5892-5908 https://doi.org/10.2174/1381612826666201118095523

[30] Vicky Wong, Hydrogels Water Absorbing Polymers, *Catalyst*, 18, 1, (2007), 18-21

[31] Jinxing Song, Huafeng Feng, Mengqi Wu, Linxiao Chen, Wei Xia, Wenqing Zhang, Preparation and characterization of arginine-modified chitosan/hydroxypropyl methylcellose antibacterial film, *International Journal of Biological Macromolecules*, 145, (2020), 750-758 https://doi.org/10.1016/j.ijbiomac.2019.12.141

[32] Linling Li, Wenxin Guo, Lu Wang, Shuiyuan Cheng, Hua Cheng, Chitosan derived nano-selenium based coatings for postharvest safety of cherry tomato, *LWT*, 217, (2025), 117441 https://doi.org/10.1016/j.lwt.2025.117441

[33] Evdokia Vassiliadi, Marta Tsirigotis-Maniecka, Henry E. Symons, Pierangelo Gobbo, Frédéric Nallet, Aristotelis Xenakis, Maria Zoumpanioti, (Hydroxypropyl)methyl Cellulose-Chitosan Film as a Matrix for Lipase Immobilization—Part II: Structural Studies, *Gels*, 8, 9, (2022), 595 https://doi.org/10.3390/gels8090595

[34] Raada Dowais, Sarah Al Sharie, Mohammad Araydah, Sahar Al Khasawneh, Fadi Haddad, Anas AlJaiuossi, Pearl-white gallstones: A report of a case and a chemical analysis by FTIR and XRD, *International Journal of Surgery Case Reports*, 87, C, (2021), 106449 https://doi.org/10.1016/j.ijscr.2021.106449

[35] Wenjie Wang, Changhu Xue, Xiangzhao Mao, Chitosan: Structural modification, biological activity and application, *International Journal of Biological Macromolecules*, 164, (2020), 4532-4546 https://doi.org/10.1016/j.ijbiomac.2020.09.042

[36] Qiong Jiang, Wei Zhou, Jun Wang, Rupei Tang, Di Zhang, Xin Wang, Hypromellose succinate-crosslinked chitosan hydrogel films for potential wound dressing, *International Journal of Biological Macromolecules*, 91, (2016), 85-91 https://doi.org/10.1016/j.ijbiomac.2016.05.077

[37] Salvatore Lombardo, Bernard Cathala, Ana Villares, Adsorption of biopolymers onto nanocelluloses for the fabrication of hollow microcapsules, *Nordic Pulp & Paper Research Journal*, 36, 4, (2021), 651-661 https://doi.org/10.1515/npprj-2021-0040

[38] Getnet Abera, Belay Woldeyes, Hundessa Dessalegn Demash, Garret Miyake, The effect of plasticizers on thermoplastic starch films developed from the indigenous Ethiopian tuber crop Anchote (*Coccinia abyssinica*) starch, *International Journal of Biological Macromolecules*, 155, (2020), 581-587 https://doi.org/10.1016/j.ijbiomac.2020.03.218

[39] Furong Huang, Han Song, Liu Guo, Peiwen Guang, Xinhao Yang, Liqun Li, Hongxia Zhao, Maoxun Yang, Detection of adulteration in Chinese honey using NIR and ATR-FTIR spectral data fusion, *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy*, 235, (2020), 118297 https://doi.org/10.1016/j.saa.2020.118297

[40] Fatemeh Mahdiyeh Boroujeni, Gabriella Fioravanti, Ronald Kander, Synthesis and Characterization of Cellulose Microfibril-Reinforced Polyvinyl Alcohol Biodegradable Composites, *Materials*, 17, 2, (2024), 526 https://doi.org/10.3390/ma17020526

[41] Hongfeng Chen, Qi Li, Mingxia Wang, Daobin Ji, Wenfeng Tan, XPS and two-dimensional FTIR correlation analysis on the binding characteristics of humic acid onto kaolinite surface, *Science of The Total Environment*, 724, (2020), 138154 https://doi.org/10.1016/j.scitotenv.2020.138154

[42] Fang-Da Zhang, Chang-Hua Xu, Ming-Yu Li, An-Min Huang, Su-Qin Sun, Rapid identification of *Pterocarpus santalinus* and *Dalbergia louvelii* by FTIR and 2D correlation IR spectroscopy, *Journal of Molecular Structure*, 1069, (2014), 89-95 https://doi.org/10.1016/j.molstruc.2014.01.085

[43] Wenlong Liang, Yang Zhao, Douxin Xiao, Jingli Cheng, Jinhao Zhao, A biodegradable water-triggered chitosan/hydroxypropyl methylcellulose pesticide mulch film for sustained control of *Phytophthora sojae* in soybean (*Glycine max* L. Merr.), *Journal of Cleaner Production*, 245, (2020), 118943 https://doi.org/10.1016/j.jclepro.2019.118943

[44] Meryiem Derraz, Abdelaziz Elouahli, Chouaib Ennawaoui, Mohamed Aymen Ben Achour, Abdelkader Rjafallah, El Mehdi Laadissi, Hamza Khallok, Zineb Hatim, Abdelowahed Hajjaji, Extraction and Physicochemical Characterization of an Environmentally Friendly Biopolymer: Chitosan for Composite Matrix Application, *Journal of Composites Science*, 7, 6, (2023), 260 https://doi.org/10.3390/jcs7060260

[45] Arno Verlee, Stein Mincke, Christian V. Stevens, Recent developments in antibacterial and antifungal chitosan and its derivatives, *Carbohydrate Polymers*, 164, (2017), 268-283 https://doi.org/10.1016/j.carbpol.2017.02.001

[46] Liwei Gao, Xinjie Zhao, Meng Liu, Xiangzhong Zhao, Characterization and Antibacterial Activities of Carboxymethylated Paramylon from *Euglena gracilis*, *Polymers*, 14, 15, (2022), 3022 https://doi.org/10.3390/polym14153022

[47] Jinfeng Cao, Yang Wang, Chen He, Yanhui Kang, Jinping Zhou, Ionically crosslinked chitosan/poly(acrylic acid) hydrogels with high strength, toughness and antifreezing capability, *Carbohydrate Polymers*, 242, (2020), 116420 https://doi.org/10.1016/j.carbpol.2020.116420

[48] Muhammed L. Sanyang, Salit M. Sapuan, Mohammad Jawaid, Mohamad R. Ishak, Japar Sahari, Effect of Plasticizer Type and Concentration on Tensile, Thermal and Barrier Properties of Biodegradable Films Based on Sugar Palm (*Arenga pinnata*) Starch, *Polymers*, 7, 6, (2015), 1106-1124 https://doi.org/10.3390/polym7061106

[49] Zihang Shen, Jie Ma, Yijie Cai, Siyang Li, Dongrui Ruan, Shufen Dai, Zhi Sheng, Jiabao Bai, Daochen Yin, Jianfeng Ping, Yibin Ying, Canhui Yang, Shaoxing Qu, Zheng Jia, Low-water-content polyelectrolyte hydrogels inspired by human epidermal stratum corneum, *Cell Reports Physical Science*, 4, 12, (2023), 101741 https://doi.org/10.1016/j.xcrp.2023.101741