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Research Paper

Synthesis and Characterization of Superabsorbent Bio-Based Hydrogels from *Pandanus tectorius* Leaf Cellulose through Ester-Based Crosslinking for Water-Holding Applications

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Abstract
The development of sustainable hydrogels from renewable biomass has attracted increasing interest for environmental and agricultural applications. In this study, cellulose isolated from *Pandanus tectorius* leaves was utilized as a precursor for the synthesis of carboxymethyl cellulose (CMC), which was subsequently used to prepare hydrogels through ester-based crosslink. A dual-network hydrogel structure was formed by blending CMC with hydroxyethyl cellulose (HEC), followed by esterification with citric (CA), fumaric (FA), and malic (MA) acids. Structural characterization using FTIR confirmed successful carboxymethylation and ester crosslinking within the polymer network, while SEM revealed a porous morphology favorable for water diffusion. The synthesized hydrogels exhibited gel fractions of 67-89%, swelling ratios of 578-4163%, and maximum degradation temperatures of 311-428 °C. The CMC/HEC-MA hydrogel at a 15% crosslinker concentration demonstrated the most balanced combination of swelling capacity, gel fraction, and thermal stability. Owing to their high water-retention capability, porous structure, and biodegradability, these hydrogels show strong potential as superabsorbent materials for soil moisture conservation, controlled water release in agriculture, drought mitigation, and sustainable water management. These findings highlight the potential of biomass-derived hydrogels to support SDG 2 (Zero Hunger) by improving agricultural water retention, SDG 6 (Clean Water and Sanitation) by promoting efficient water management and treatment, and SDG 12 (Responsible Consumption and Production) by transforming renewable biomass resources into sustainable value-added materials.

Keywords
Carboxymethyl Cellulose (CMC), Hydrogels, Hydroxyethyl Cellulose (HEC), SDG 6 (Clean Water and Sanitation)

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1. INTRODUCTION

Hydrogels are three-dimensional hydrophilic polymer networks capable of absorbing and retaining large amounts of water while maintaining their structural integrity, making them suitable for biomedical, environmental, and agricultural applications (Yang, 2022; Priya et al., 2024). Their unique ability to combine high water uptake with tuneable physicochemical and mechanical properties has led to increasing interest in developing hydrogels for controlled water release, soil moisture retention, drug delivery, tissue engineering, wound dressing, and wastewater treatment. In particular, the growing demand for environmentally sustainable materials has accelerated research into bio-based hydrogels derived from renewable natural polymers (Altaf et al., 2021; MohanKumar et al., 2022; Segneanu et al., 2025). Most commercially available hydrogels are synthesized from petroleum-based polymers, which offer tunable physicochemical properties but often suffer from drawbacks such as poor biodegradability, limited environmental compatibility, and potential toxicity concerns (Jiang and Zhou, 2025; Shariatinia and Jalali, 2018). These limitations have motivated the replacement of synthetic polymers with biodegradable and renewable alternatives that can reduce environmental impacts without compromising hydrogel performance.

Natural polymers such as starch, carrageenan, chitosan, alginate, and cellulose have received considerable attention as sustainable hydrogel precursors because of their biodegradability, biocompatibility, low toxicity, abundant availability, and excellent water absorption characteristics (Li and Gong, 2024; Agustina et al., 2026; Noviza et al., 2026; Winarti et al., 2026). Among these materials, cellulose is particularly attractive be-

cause it is the most abundant renewable biopolymer, possesses excellent mechanical strength, chemical stability, and abundant hydroxyl groups that enable various chemical modifications (Adawiyah et al., 2022; Nurhaliza et al., 2022; Trisnawati et al., 2024). These characteristics make cellulose an ideal platform for designing functional hydrogels with tailored physicochemical properties.

Carboxymethyl cellulose (CMC), one of the most extensively studied cellulose derivatives, is a linear anionic polysaccharide containing hydroxyl and carboxyl functional groups that facilitate hydrogel formation through physical or chemical crosslinking (Suryanti et al., 2023; Salama et al., 2024, 2026). CMC exhibits excellent hydrophilicity, biodegradability, biocompatibility, and film-forming ability, making it a promising material for superabsorbent hydrogel preparation. Nevertheless, hydrogels prepared solely from CMC often suffer from inadequate mechanical strength, limited structural stability, excessive solubility, and insufficient network formation, resulting in poor dimensional stability during swelling (Demitri et al., 2008; Dolbow et al., 2005; Wulandari et al., 2016). Therefore, improving the crosslinked network structure is essential to obtain hydrogels with balanced swelling capacity, structural integrity, and thermal stability.

One effective strategy for overcoming these limitations is blending CMC with hydroxyethyl cellulose (HEC), a nonionic cellulose derivative capable of enhancing intermolecular hydrogen bonding and reinforcing the hydrogel network. The incorporation of HEC can improve network homogeneity while maintaining high water absorption capacity (Kim and Kang, 2026). In addition, the use of environmentally friendly crosslinking agents is increasingly preferred over conventional synthetic crosslinkers because they reduce toxicity and improve the sustainability of hydrogel production. Polycarboxylic acids such as citric acid (CA), fumaric acid (FA), and malic acid (MA) have emerged as promising green crosslinkers because they can react with cellulose hydroxyl groups through esterification to form stable three-dimensional covalent networks (Saputra et al., 2015; Reddy et al., 2015; Qiu et al., 2021). Besides enhancing network stability, the molecular structure and functionality of these polycarboxylic acids strongly influence crosslink density, swelling behavior, gel fraction, and thermal stability. However, comparative studies evaluating the influence of different ester-based polycarboxylic acids under identical preparation conditions remain relatively limited.

Pandanus tectorius leaves are an abundant lignocellulosic biomass widely distributed throughout tropical regions and represent a sustainable source of cellulose (Trisnawati et al., 2023a,b; Suryanti et al., 2023). Although these leaves contain a high proportion of cellulose and are available throughout the year, they are largely treated as agricultural waste or remain underutilized. Converting this renewable biomass into high-value CMC for hydrogel production not only improves resource utilization but also supports circular bioeconomy principles by transforming low-value agricultural residues into environmentally friendly functional materials.

In CMC-based hydrogels, both the chemical structure and concentration of the crosslinker significantly influence the resulting network architecture and physicochemical properties, including swelling behavior, gel fraction, morphology, and thermal stability (Ahmed, 2015; Chang and Zhang, 2011; Huang et al., 2017). Accordingly, superabsorbent bio-based hydrogels were synthesized from CMC derived from *P. tectorius* leaf cellulose and blended with HEC, using CA, FA, and MA as green ester-based crosslinking agents. The effects of crosslinker chemistry and concentration on the structural, chemical, thermal, morphological, gel fraction, and swelling properties of the resulting hydrogels were systematically investigated. The novelty of this study lies in the use of cellulose derived from *P. tectorius* leaves as a sustainable CMC precursor and the systematic comparison of three environmentally friendly polycarboxylic acid crosslinkers to establish structure–property relationships in CMC/HEC hydrogels. These findings provide a sustainable strategy for developing high-performance bio-based superabsorbent hydrogels for water-retention and agricultural applications.

2. EXPERIMENTAL SECTION

2.1 Materials

Leaves of *P. tectorius* were collected from the coastal area of Gunung Kidul, Yogyakarta, Indonesia. All chemicals were of analytical grade (≥98% purity, unless otherwise specified) and used as received without further purification. The materials included sodium hydroxide, sodium hypochloride, isopropanol, sodium monochloroacetate, acetic acid, ethanol, HEC, CA, FA and MA. The reagents were purchased from Sigma-Aldrich, except hydroxyethyl cellulose was supplied by Natrosol (Shandong Guanwei-China).

2.2 Methods

2.2.1 Sample Preparation of *P. tectorius* Leaves

Leaves were collected and cleaned to remove thorns. The thorn-free leaves were washed thoroughly with water and cut into pieces measuring approximately 12 cm × 3 cm. The cut leaves were sun-dried until moisture content was reduced. After drying, the leaves were further cut into smaller pieces measuring 1 cm × 1 cm. The small pieces were then soaked in 10 L of water for 72 hours (3 × 24 h) with regular water changes. After soaking, the leaves were rinsed with clean water, dried, and ground into a fine powder for further use.

2.2.2 Isolation of *P. tectorius* Leaves Cellulose

Cellulose was isolated from *P. tectorius* leaves using an alkaline and bleaching treatment. Initially, 25 g of powdered leaves were mixed with 500 mL of 10% NaOH and stirred at approximately 100 rpm for 120 minutes. The mixture was then neutralized with distilled water, and the solid residue was collected and dried. The dried residue was treated with 4% NaOCl (pH 4.5) at a residue-to-NaOCl ratio of 1:20, and stirred at 100 rpm for 120 minutes. After the bleaching step, the mixture

was neutralized again with distilled water and dried to obtain purified cellulose.

2.2.3 Synthesis of Carboxymethyl Cellulose (CMC)

CMC was synthesized from the isolated *P. tectorius* cellulose via etherification. One gram of cellulose was suspended in a mixture of 3 mL of 20% NaOH and 20 mL of isopropanol and stirred at room temperature for 1 hour. Sodium monochloroacetate (1 g), dissolved in 4 mL of isopropanol, was added gradually to the mixture while refluxing at 45–50 °C. The reaction mixture was stirred for 3 hours, then filtered. The residue was neutralized with 10% acetic acid, washed with 95% ethanol, and dried at 60 °C to obtain the CMC-Na product.

2.2.4 Synthesis of CMC/HEC-Based Hydrogels

Hydrogels were prepared using a blend of CMC and HEC. A total of 1.2 g of polymer (0.9 g CMC-Na and 0.3 g HEC) was dissolved in 60 mL of distilled water with continuous stirring until completely homogeneous. Initially, HEC was dissolved in distilled water for 5 minutes to achieve slight viscosity. Subsequently, CMC-Na was added, and the solution was stirred at room temperature for 24 hours until a significant viscosity increase was observed. Crosslinkers, such as CA, FA or MA, were added at various concentrations of 10, 15, and 20% (w/w, based on the total polymer weight) (Table 1). These concentrations were selected to evaluate the effect of low, intermediate, and high crosslinking densities on hydrogel properties while remaining within the range commonly reported for polycarboxylic acid-crosslinked cellulose-based hydrogels. The resulting solution was poured into Petri dishes and allowed to stand at room temperature for 24 hours to remove excess water. The formed hydrogels were then dried at 50°C under for 16 h.

2.2.5 Functional Group Analysis by Fourier Transform Infrared (FTIR) Spectroscopy

FTIR spectroscopy was used to analyze hydrogel functional groups using a Shimadzu IR Prestige-21 FTIR instrument in KBr pellet mode. Spectra were recorded at a resolution of 2 cm^{-1} with 30 scans over 400-4000 cm^{-1}. Crosslinking was confirmed by the appearance of a new absorption band around 1700 cm^{-1}, corresponding to ester bonds (COO^{-}) formed between acid anhydrides and cellulose hydroxyl groups.

2.2.6 Morphological Analysis by Scanning Electron Microscope (SEM)

Hydrogel surface morphology was examined using a scanning electron microscope (SEM Jeol Benchtop JCM-7000) at 500–1000× magnification, 15 kV accelerating voltage, and 70 Pa chamber pressure. SEM images showed a porous structure, indicative of crosslinking that facilitates water absorption.

2.2.7 Thermal Stability Analysis by Thermogravimetric Analyzer (TGA)

Thermal stability was analyzed using a STA Linseis PT 1600 thermogravimetric analyzer under a nitrogen flow of 20 mL/min. Samples were heated from room temperature to 600 °C at 10 °C/min. Three degradation stages were observed at: (i). 50-140 °C: dehydration of surface-bound water, (ii). 180-428 °C: degradation of crosslinking agents, and (iii). 428 °C: decomposition of the CMC polymer backbone.

2.2.8 Gel Fraction Measurement

Hydrogel films (3 × 3 cm) were immersed in distilled water for 14 hours, with water replaced twice to remove unreacted components such as residual crosslinking agents. The hydrogel was then dried and weighed. The gel fraction (%) was calculated using Equation (1).

$$\%\text{Gel Fraction} = \frac{W_g}{W_0} \times 100\% \quad (1)$$

where:
- Wg = weight of the hydrogel after immersion and drying
- W_0 = weight of the hydrogel before immersion

2.2.9 Swelling Ratio Measurement

Hydrogel films (1 × 5 cm) were immersed in distilled water at room temperature for 1 hour. Excess water on the hydrogel surface was gently removed using filter paper. The hydrogel was weighed before and after immersion, and the swelling ratio (SR) was calculated using Equation (2).

$$\%SR = \frac{W_t - W_0}{W_0} \times 100\% \quad (2)$$

where:
- Wt = weight of the wet hydrogel
- W_0 = weight of the dry hydrogel

2.2.10 Statistical Analysis

All experiments were performed in triplicate, and the results are presented as mean ± standard deviation (SD). Statistical analysis was conducted using one-way analysis of variance (ANOVA), followed by Tukey's post hoc test to determine significant differences among the samples. Differences were considered statistically significant at $p \leq 0.05$.

3. RESULTS AND DISCUSSIONS

3.1 Isolation of Cellulose from *P. tectorius* Leaves

The valorization of underutilized lignocellulosic biomass as a renewable precursor for functional polymer materials has attracted increasing interest. In this study, leaves of *Pandanus tectorius*, an abundant tropical plant resource, were explored as a novel cellulose feedstock for hydrogel production. Cellulose isolation was performed through sequential alkalization and bleaching treatments to remove lignin, hemicellulose, and other extractives. During alkalization, the extract gradually darkened, indicating dissolution of chromophoric compounds and effective delignification of the pandanus fiber matrix (Trisnawati et al., 2023a,b).

Table 1. Characteristics of CMC/HEC-CA, CMC/HEC-FA, and CMC/HEC-MA Hydrogels.

Crosslinker Agent	Hydrogels	Weight (g)	pH	Thickness (mm)
Citric Acid	CMC/HEC-CA10	1.24	6	0.18
	CMC/HEC-CA15	1.20	5	0.14
	CMC/HEC-CA20	1.26	4	0.30
Fumaric Acid	CMC/HEC-FA10	1.27	6	0.31
	CMC/HEC-FA15	1.23	5	0.20
	CMC/HEC-FA20	1.19	3	0.12
Malic Acid	CMC/HEC-MA10	2.07	5	0.20
	CMC/HEC-MA15	1.59	4	0.18
	CMC/HEC-MA20	1.44	3	0.27

Compositional analysis revealed a cellulose content of 40.26 %, demonstrating that *P. tectorius* leaves represent a promising alternative lignocellulosic source for the preparation of cellulose-derived functional materials. The successful extraction of cellulose provides a sustainable starting point for subsequent chemical modification and hydrogel synthesis.

3.2 Synthesis of Carboxymethyl Cellulose (CMC)

The isolated cellulose was converted into CMC via an etherification reaction using a carboxymethylating agent (Suryanti et al., 2023). During this process, hydroxyl (-OH) groups along the cellulose backbone are partially substituted by carboxymethyl groups, introducing anionic functionality and significantly improving water solubility and reactivity. This modification is particularly advantageous for hydrogel formation, as the introduced carboxyl groups facilitate intermolecular interactions and crosslinking reactions (Asl et al., 2017).

FTIR spectroscopy was employed to verify successful carboxymethylation. As shown in Figure 1, the appearance of a characteristic absorption band at 1609 cm^{-1}, corresponding to the C=O stretching vibration of carboxylate groups, confirms the formation of carboxymethyl functionality (Huang et al., 2017; Asl et al., 2017). Additional peaks at 1024 cm^{-1} and 1059 cm^{-1} are attributed to C–O–C ether stretching vibrations characteristic of CMC. These spectral features confirm the successful conversion of pandanus-derived cellulose into CMC and are consistent with previously reported CMC spectra (Tufan et al., 2016). The successful synthesis of CMC from *P. tectorius* highlights the potential of this biomass as a renewable source for producing functional polysaccharide derivatives suitable for advanced material applications.

3.3 Synthesis of CMC/HEC-Based Hydrogels

CMC obtained from *P. tectorius* cellulose was utilized as the primary polymer matrix for hydrogel synthesis through chemical crosslinking with three green polycarboxylic acids, namely CA, FA, and MA. Hydrogels were prepared using a CMC/HEC blend at a weight ratio of 3:1, in which HEC was incorporated to enhance intermolecular interactions, improve network formation and reinforce the hydrogel structure. The incorporation of HEC is advantageous because CMC alone may exhibit limited crosslinking efficiency due to electrostatic repulsion between negatively charged polymer chains and the high degree of hydroxyl substitution (Demitri et al., 2008).

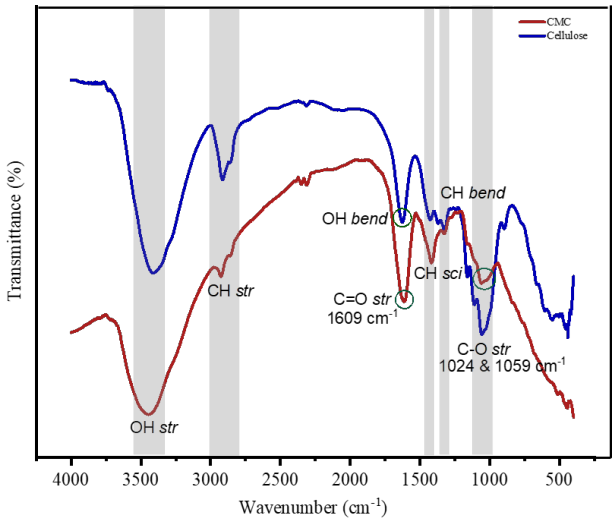

Figure 1. FT-IR Spectra of Cellulose and CMC

Figure ?? illustrates the proposed esterification reaction between the hydroxyl groups of the CMC/HEC blend and the carboxyl groups of CA, FA, and MA, resulting in the formation of a covalently crosslinked three-dimensional hydrogel network. During thermal curing, CA forms cyclic anhydride intermediates that subsequently react with the hydroxyl groups of CMC or HEC to form ester linkages (de Lima et al., 2020). In contrast, FA and MA undergo direct esterification with the polymer hydroxyl groups (Priya et al., 2019). These ester bonds act as permanent covalent crosslinks that stabilize the hydrogel network, while the combination of anionic CMC and nonionic HEC simultaneously forms an interpenetrating polymer network (IPN), further improving network integrity and structural stability (Hashem et al., 2013). The resulting interconnected porous structure, enriched with hydroxyl and carboxyl groups, facilitates efficient water diffusion and retention.

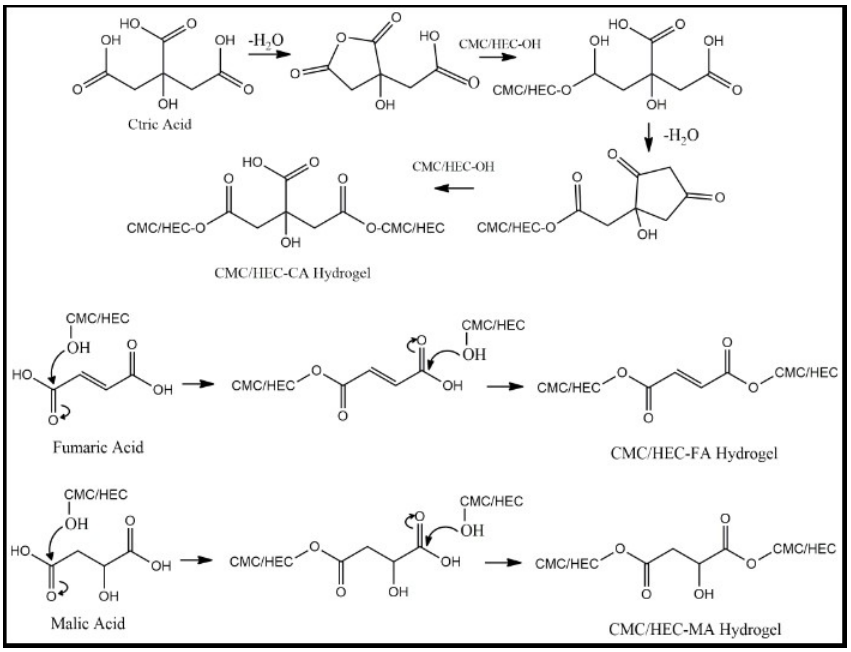

Figure 2. Proposed Esterification Reaction Between the CMC/HEC Blend and CA, FA, and MA Leading to the Formation of a Crosslinked Hydrogel Network

The molecular structures of the three crosslinkers play a crucial role in determining the hydrogel network architecture. CA contains three carboxyl groups, providing multiple reactive sites that promote the formation of highly crosslinked networks with excellent biocompatibility (Franklin and Guhanathan, 2014). FA contains two carboxyl groups linked by a rigid carbon–carbon double bond, producing comparatively rigid and structurally stable polymer networks (Kabir et al., 2018). In contrast, MA possesses two carboxyl groups together with an additional hydroxyl group, which increases the hydrophilicity of the polymer network while maintaining effective crosslinking through esterification (Qiu et al., 2021). Consequently, differences in the molecular structures and functionalities of these crosslinkers influence crosslink density, network architecture, and ultimately the gel fraction, swelling behavior, and thermal stability of the synthesized hydrogels. The synthesized hydrogels exhibited a yellowish-white, flexible appearance, with weights ranging from 1.20 to 2.07 g and thicknesses between 0.12 and 0.31 mm (Table 1). Increasing the crosslinker concentration had little effect on hydrogel weight or thickness but significantly decreased the hydrogel pH because of unreacted carboxyl groups at higher crosslinker loadings (Nasution et al., 2022).

3.4 Functional Group Analysis by FTIR Spectroscopy

FTIR analysis confirmed the formation of ester crosslinks between the polymer chains and the organic acid crosslinkers (Figure 3, Table 2). A new absorption band observed at approximately 1720 cm^{-1} corresponds to the carbonyl stretching vibration of ester groups, indicating successful covalent bonding between hydroxyl groups of CMC/HEC and the carboxyl groups of CA, FA, and MA. Hydrogels prepared with 10% crosslinker displayed weaker absorption at 1720 cm^{-1}, suggesting incomplete crosslinking due to limited crosslinker availability. In contrast, hydrogels synthesized with higher crosslinker concentrations exhibited stronger absorption bands, reflecting increased esterification and greater crosslink density. This trend is consistent with previous studies on ester-crosslinked cellulose hydrogels, in which increasing concentrations of polycarboxylic acid crosslinkers resulted in enhanced ester bond formation and higher crosslinking densities (Reddy et al., 2015; Qiu et al., 2021). Additionally, a broad –OH stretching band near 3621 cm^{-1} was observed, indicating the presence of hydrophilic functional groups capable of forming hydrogen bonds with water molecules. Similar broad –OH absorption bands have also been reported CA- and other polycarboxylic acid-crosslinked cellulose hydrogels, reflecting the retention of hydrophilic hydroxyl groups that facilitate water absorption while participating in intermolecular hydrogen bonding within the polymer network (Hashem et al., 2013; Qiu et al., 2021). Overall, the close agreement between the FTIR spectra obtained in this study and those reported for ester-crosslinked cellulose hydrogels further confirms the successful formation of covalently crosslinked CMC/HEC hydrogel networks derived from P. tectorius cellulose. Although crystallinity changes were not evaluated in this study, the successful hydrogel formation was supported by SEM, TGA, gel fraction, and swelling analyses.

3.5 Morphological Analysis by SEM

SEM images (Figure 4) reveal that the synthesized hydrogels possess a porous and interconnected microstructure, a char-

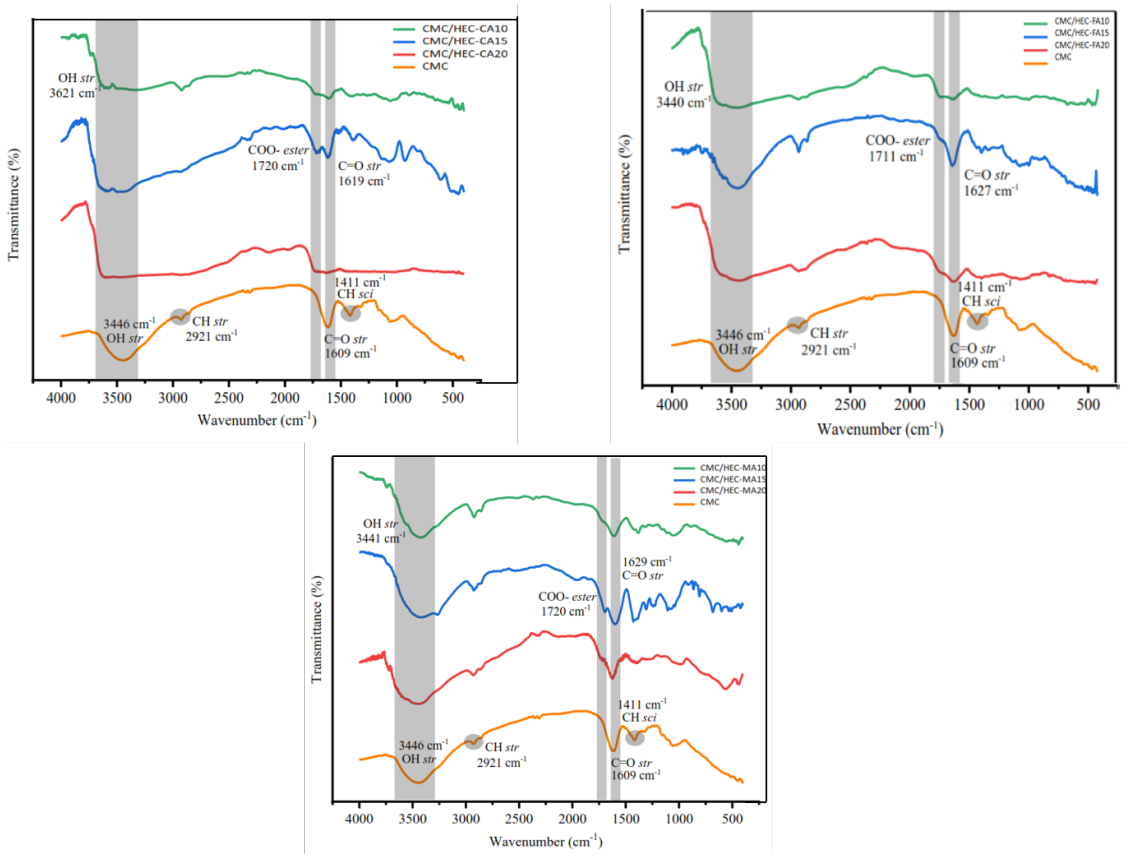

Figure 3. FTIR Spectra of CMC, CMC/HEC-CA, CMC/HEC-FAA, and CMC/HEC-MA Hydrogels

acteristic feature of chemically crosslinked hydrogel systems. Such porous architecture plays a crucial role in determining water absorption performance, as interconnected pores facilitate the diffusion and retention of water molecules within the polymer matrix. The pore structure is strongly influenced by both the type and concentration of crosslinker, reflecting the direct relationship between crosslinking chemistry and hydrogel morphology (Hashem et al., 2013). Although the SEM observations are qualitative, they consistently support the swelling behavior of the hydrogels. Further physicochemical characterization, including BET surface area analysis and EDS elemental mapping, will be conducted in future studies to better understand the relationship between the porous structure, elemental distribution, and the water absorption behavior of the synthesized hydrogels.

3.6 Thermal Stability Analysis by TGA

TGA was conducted to evaluate the thermal behaviour of the synthesized hydrogels. All samples exhibited three major degradation stages (Figure 5). The first stage, occurring below 215°C, corresponds to the evaporation of absorbed and bound water. The second stage, between 180-428°C, is attributed to the decomposition of the crosslinking agents. The final stage, above 311°C, corresponds to the degradation of the CMC polymer backbone. Crosslinking significantly improved the thermal stability of the materials, as the covalent network structure delayed polymer decomposition (Shen et al., 2026). Among the tested formulations, CMC/HEC-MA hydrogels exhibited the highest thermal stability, indicating stronger network formation with MA. Notably, the maximum degradation temperatures observed in this study exceed those reported for CMC hydrogels crosslinked with FA or MA in previous studies (298-317°C), suggesting that the hydrogel networks derived from *P. tectorius*-based CMC possess enhanced thermal stability (Seki et al., 2014). TGA was performed as a single instrumental measurement for each hydrogel formulation. The thermal degradation temperatures are presented as representative values and were interpreted qualitatively rather than subjected to statistical analysis.

3.7 Gel Fraction and Network Formation

The degree of crosslinking within the hydrogel network was evaluated through gel fraction analysis, which reflects the proportion of polymer chains successfully incorporated into the insoluble three-dimensional network. The gel fraction ranged from 66.5±0.9 to 89.1±1.3%, with the highest values observed for hydrogels prepared with 20% crosslinker exhibiting significantly higher gel fractions ($p < 0.05$) than those prepared with lower crosslinker concentrations (Figure 6). Increasing the crosslinker concentration provided a greater number of

Table 2. Infrared Spectral Bands (cm-1) for CMC/HEC-CA, CMC/HEC-FA and CMC/HEC-MA Hydrogels.

Functional Group Assigned	Absorbance Bands (cm^{-1})			
	CMC/HEC-CA	CMC/HEC-FA	CMC/HEC-MA	Reference
O-H Stretching (Hydrogel)	3404-3508	3421-3440	3419-3441	3347-3450a
COO- Ester (Hydrogel)	1718-1720	1708-1724	1708-1720	1713-1734b
C=O Stetching (CMC)	1612-1619	1613-1627	1599-1629	1595-1605c

[a](Morán et al., 2008) [b](Seki et al., 2014) [c](Durpekova et al., 2020)

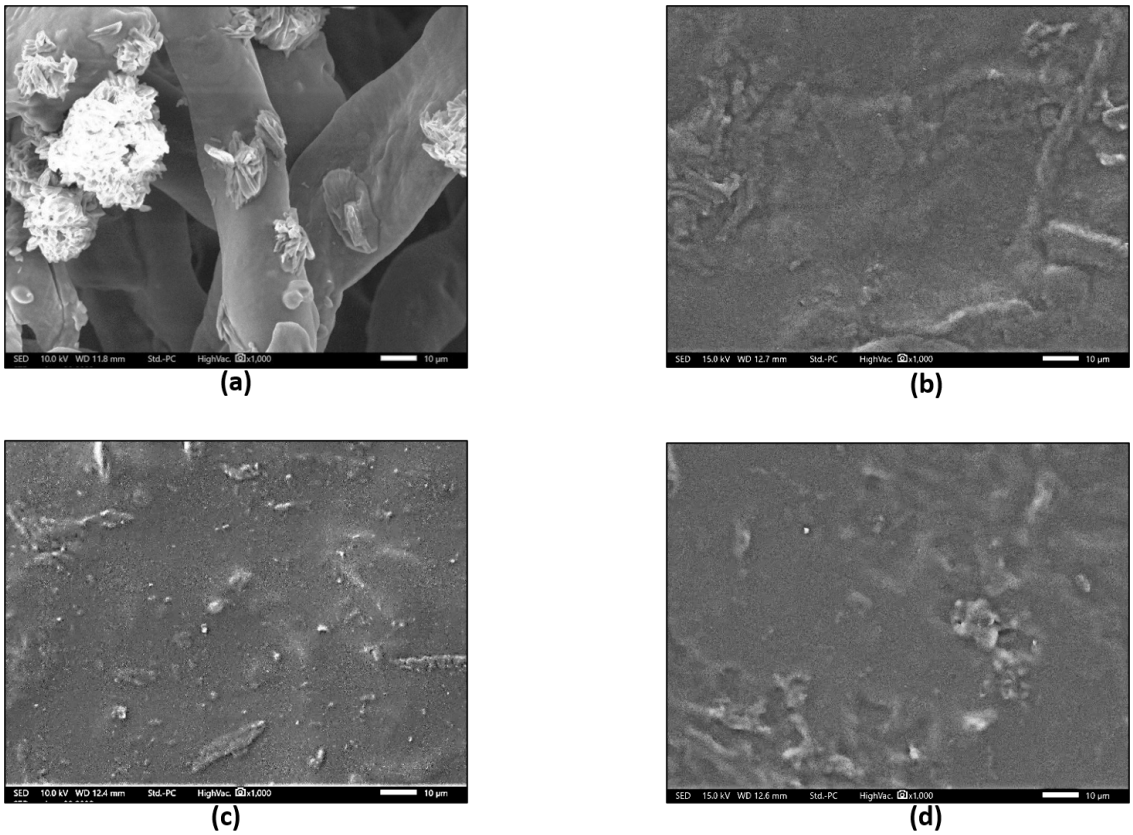

Figure 4. SEM Images of (a). CMC, (b). CMC/HEC-CA15, (c). CMC/HEC-FA15, and (d). CMC/HEC-MA15 Hydrogels

carboxyl groups available for esterification with the hydroxyl groups of CMC and HEC, thereby promoting the formation of additional covalent crosslinks and increasing network integrity (Saputra et al., 2015). Conversely, at lower crosslinker concentrations, incomplete crosslinking resulted in less stable polymer networks, allowing a larger fraction of un-crosslinked polymer chains to dissolve during water immersion and consequently lowering the gel fraction (Bashir et al., 2020). Among the different crosslinkers, the CMC/HEC-CA hydrogels consistently exhibited the highest gel fractions, increasing from 70.3 ± 0.7% at 10 wt% crosslinker to 89.1 ± 1.3% at 20 wt%, whereas the CMC/HEC-FA and CMC/HEC-MA hydrogels reached 81.5 ± 1.1% and 81.2 ± 1.0%, respectively, at the same crosslinker concentration. The superior gel fraction obtained with CA is attributed to its three carboxyl groups, which provide more reactive sites for esterification than FA and MA, each containing only two carboxyl groups. Consequently, CA generated the highest crosslinking efficiency and produced the most densely interconnected polymer network.

3.8 Swelling Behaviour

The swelling ratio, defined as the relative increase in hydrogel mass after water absorption, is a key parameter for evaluating hydrogel performance in water-retention applications. Wa-

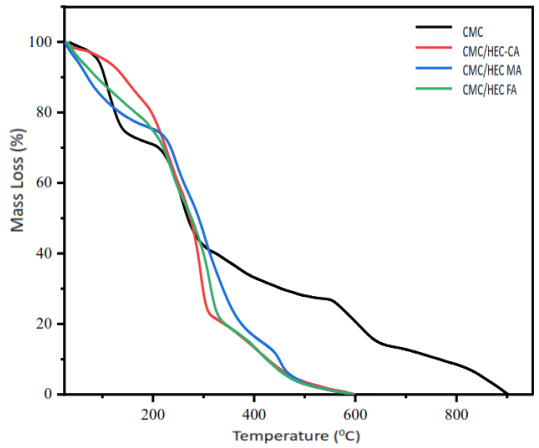

Figure 5. TGA Graphic of CMC, CMC/HEC-CA15, CMC/HEC-FA15, and CMC/HEC-MA15 Hydrogels

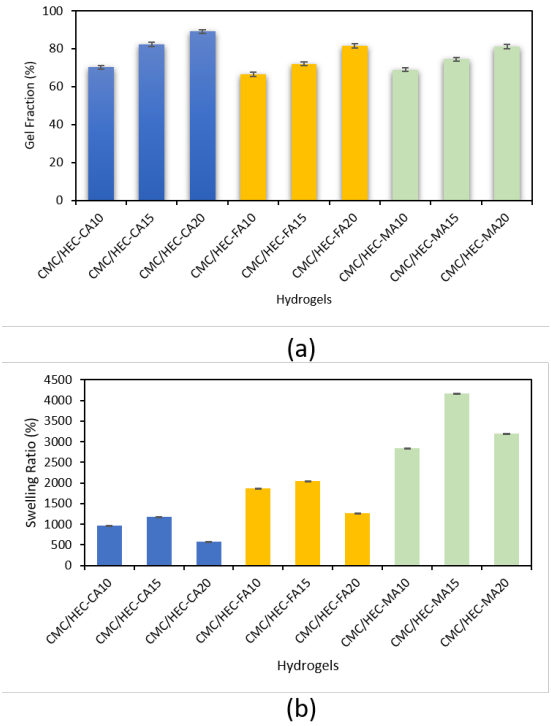

Figure 6. (a). Gel Fraction and (b). Swelling Ratio of CMC/HEC-CA, CMC/HEC-FA, and CMC/HEC-MA Hydrogels

ter molecules diffuse into the hydrogel network through hydrophilic sites and the interconnected pore structure created during crosslinking (Feng and Wang, 2023). The swelling behavior was evaluated in distilled water to determine the intrinsic water absorption capacity of the synthesized hydrogels under standardized conditions. Distilled water was selected to eliminate the influence of dissolved ions and pH variations, thereby allowing direct comparison of the intrinsic swelling behavior among the different hydrogel formulations. The effects of pH and ionic strength on swelling will be investigated in future work to better evaluate hydrogel performance under practical environmental conditions.

The swelling ratios of the synthesized hydrogels ranged from 577.8 ± 1.8% to 4163.2 ± 2.8%, indicating excellent water absorption capacity (Figure 6). Hydrogels prepared with 15 wt% crosslinker exhibited significantly higher swelling ratios ($p < 0.05$) than those prepared with 10 and 20 wt% crosslinker concentrations. At 10 wt%, the crosslinking reaction was insufficient to form a well-developed three-dimensional network, resulting in limited water absorption. Conversely, increasing the crosslinker concentration to 20 wt% reduced the swelling capacity because excessive crosslink density produced a more compact and rigid network that restricted polymer chain mobility and hindered water diffusion (Saputra et al., 2015; Seki et al., 2014). Among the three crosslinkers, the CMC/HEC-MA hydrogels exhibited significantly higher swelling ratios ($p < 0.05$) than the CMC/HEC-CA and CMC/HEC-FA hydrogels, despite exhibiting lower gel fractions than the CMC/HEC-CA formulations.

The observed differences in hydrogel performance are closely associated with the molecular structures of the three polycarboxylic acid crosslinkers. CA, containing three carboxyl groups, provides the greatest number of reactive sites for esterification with the hydroxyl groups of CMC and HEC, resulting in the highest gel fraction and the most densely crosslinked network. In contrast, FA contains two carboxyl groups linked by a rigid carbon–carbon double bond, which reduces molecular flexibility and limits the formation of an efficient three-dimensional polymer network. MA also contains two carboxyl groups but possesses an additional hydroxyl group that increases the hydrophilicity of the polymer network. Consequently, the CMC/HEC-MA hydrogels exhibited lower gel fractions than the CMC/HEC-CA hydrogels but achieved significantly greater swelling because the additional hydroxyl functionality promoted stronger polymer–water interactions. This enhanced affinity for water facilitated greater penetration and retention of water molecules within the hydrogel matrix despite the lower crosslink density.

These findings demonstrate that hydrogel swelling is governed not only by crosslink density but also by the balance between network stability and hydrophilicity. Although a high gel fraction generally indicates improved structural integrity and resistance to dissolution, excessive crosslinking produces a compact network with reduced free volume, limiting polymer chain relaxation and water diffusion (Hashem et al., 2013). Conversely, increased network hydrophilicity promotes stronger polymer–water interactions and enhances water uptake. Therefore, the superior swelling performance of the CMC/HEC-MA hydrogel can be attributed to an optimal balance between sufficient network stability and enhanced hydrophilicity rather than to crosslink density alone.

The exceptionally high swelling capacity obtained in this study indicates that the developed hydrogels have consider-

Table 3. Properties of CMC/HEC-CA, CMC/HEC-FA, and CMC/HEC-MA Hydrogels

Gel Properties	Hydrogels			
	CMC/HEC-CA	CMC/HEC-FA	CMC/HEC-MA	Reference [a]
Colour	Yellowish White	Yellowish White	Yellowish White	White
pH	4-6	3-6	3-5	4.5
Swelling Ratio (%)	577-1172	1273-2043	2841-4163	1000-1700
Hydrogel Degradation (°C)	>311	>340	>428	300-320

[a](Durpekova et al., 2020)

able potential for water-retention applications. Nevertheless, additional studies involving swelling kinetics, pH-responsive swelling, repeated swelling–deswelling cycles, soil water-retention performance, and biodegradation are required to comprehensively evaluate their long-term stability and practical applicability under agricultural conditions.

3.9 Comparative Performance Evaluation and Agricultural Potential of CMC/HEC Hydrogels

The swelling behavior of hydrogels varies considerably depending on the polymer composition and network architecture. Typical swelling ratios of synthetic hydrogels range from approximately 100–600% for poly(ethylene glycol) (PEG), 500–1500% for poly(acrylamide) (PAAm), and 1000–3000% for poly(acrylic acid) (PAA) (Son and Lee, 2016). In comparison, CMC-based hydrogels generally exhibit swelling ratios ranging from about 100% to over 1000% in distilled water, although these values are strongly influenced by crosslinking density, polymer composition, pH, and ionic strength (Kowalski et al., 2020). Increasing the CMC content generally enhances water absorption because of the higher concentration of hydrophilic carboxyl groups until an optimum network structure is achieved.

The hydrogels synthesized in this study exhibited swelling ratios ranging from 577.8 to 4163.2%, with the CMC/HEC-MA hydrogel prepared using 15 wt% crosslinker achieving the highest water uptake. This swelling capacity exceeds that reported for many conventional CMC-based hydrogels prepared from commercial cellulose sources and is also higher than the upper range commonly reported for several synthetic superabsorbent hydrogels. As summarized in Table 3, the synthetic CMC/HEC-FA and CMC/HEC-MA hydrogels also demonstrated superior thermal stability compared with similar CMC-based systems reported by Durpekova et al. (2020), indicating that CMC isolated from *P. tectorius* cellulose can serve as an effective and sustainable precursor for high-performance hydrogel networks. Although mechanical properties were not evaluated in the present study, the relatively high gel fraction together with enhanced thermal stability suggests the formation of a stable three-dimensional crosslinked network capable of maintaining structural integrity during repeated swelling and drying cycles. Such characteristics are particularly desirable for agricultural superabsorbent materials, which require both high water absorption and long-term network stability under fluctuating environmental conditions.

Hydrogels designed for agricultural applications generally require expansion capacities in the range of 200-800 times their original volume, together with environmental stability and biodegradability without generating toxic by-products (Neethu et al., 2018). Based on the swelling behavior observed in this study, the developed CMC/HEC hydrogels satisfy these fundamental requirements and therefore show considerable potential for applications including soil moisture retention, improved water-use efficiency, soil conservation, and drought stress mitigation (Zhang et al., 2025; Ali et al., 2024; Agbna and Zaidi, 2025). The exceptionally high swelling capacity obtained in this study places the developed hydrogel among the upper range of biodegradable natural polymer-based superabsorbent materials reported in recent literature. Importantly, this high absorbency was achieved while maintaining a high gel fraction and improved thermal stability, indicating that increased water uptake was not accompanied by excessive deterioration of the polymer network. Nevertheless, the proposed agricultural application is based primarily on laboratory measurements of swelling behavior, gel fraction, and thermal stability. Soil water-retention performance, mechanical durability under field conditions, biodegradation, and soil degradation behavior will be investigated in future work to comprehensively validate the practical applicability and environmental sustainability of these hydrogels.

These results suggest that the dual-network CMC/HEC hydrogel system derived from *P. tectorius* cellulose provides a structurally robust and highly absorbent alternative to conventional CMC-based hydrogels prepared from commercial precursors. The utilization of renewable *P. tectorius* biomass further enhances the sustainability of the developed material by replacing commercially produced cellulose with an abundant lignocellulosic resource. Such materials can contribute to Sustainable Development Goal (SDG) 2 (Zero Hunger) by improving soil water retention, SDG 6 (Clean Water and Sanitation) through more efficient water management, and SDG 12 (Responsible Consumption and Production) by promoting the utilization of renewable biomass resources. Overall, the present study demonstrates that naturally derived CMC can be engineered into high-performance superabsorbent hydrogels with physicochemical properties comparable to or exceeding those of many previously reported CMC-based systems.

4. CONCLUSIONS

This study demonstrates the successful valorization of *P. tectorius* leaves as a renewable source for producing CMC-based hydrogels. Cellulose was effectively isolated (40.26%) and converted into carboxymethyl cellulose, as confirmed by FTIR analysis. The resulting CMC was used to synthesize hydrogels in a CMC/HEC system crosslinked with CA, FA and MA, forming stable three-dimensional polymer networks with porous structures. The synthesized hydrogels exhibited gel fractions of 66.5±0.9 to 89.1±1.3% and swelling ratios ranging from 577.8±1.8 to 4163.2±2.8%. Among the tested formulations, the CMC/HEC-MA hydrogel with 15% crosslinker concentration showed the most optimal balance between swelling capacity, network stability, and thermal resistance. Overall, hydrogels derived from *P. tectorius* cellulose demonstrated excellent water absorption and structural stability, suggesting their potential as sustainable superabsorbent materials for agricultural water-retention applications. However, their practical applicability should be further validated through soil-based performance evaluations under realistic agricultural conditions. Future studies should focus on evaluating the mechanical properties, long-term biodegradability, swelling–deswelling durability, and performance under realistic soil conditions to further validate the practical applicability of the developed hydrogels.

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