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Superabsorbent hydrogel derived from hide trimming waste

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Abstract

Superabsorbent hydrogels were produced using a graft copolymerization technique, utilizing hydrolyzed collagen obtained from hide trimming waste and acrylic acid as the monomer. Methylenebisacrylamide (MBA) was employed as a crosslinker. The concentrations of acrylic acid and MBA were systematically optimized to obtain maximum swelling capacity. The maximum swelling capacity was 156 g/g in distilled water. The structure of the superabsorbent was verified using FTIR spectroscopy, and the morphology was identified using SEM. Various salt solutions (NaCl, KCl, $MgCl_2$, and $CaCl_2$) and solutions with pH levels spanning from 1 to 13 were also utilized to evaluate the swelling capacity of hydrogels.

Keywords: *hydrogels, swelling, graft copolymers, biopolymers, hide trimming waste.*

Data Availability: Research data is available upon request from the corresponding author.

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1. Introduction

Hydrogel, also known as a superabsorbent polymer, is a network of hydrophilic cross-linked molecules that can absorb, expand, and retain liquid several times its dry weight without dissolving[1-3]. Superabsorbent hydrogels have been employed in diverse sectors, including hygienic and agricultural product applications[1,4,5], coal dewatering[6], sealing[7,8], controlled drug delivery systems[9,10], and as additives for concrete[11]. The hydrophilic functional groups that attach to the polymer backbone of hydrogels are responsible for their water-absorbing properties. The presence of interconnections between network structures is the reason for their resistance to dissolution. When exposed to specific external factors, including temperature, solvent, pH, and electric field, hydrogels can undergo significant volume changes[12,13]. The hydrogel matrix can undergo volume changes either gradually throughout a range of stimulus levels or suddenly at certain stimulus levels.

Hydrogel structure and properties are influenced by various parameters, such as crosslinking agents, monomers, and the network chemical composition. The hydrogel exhibits great hydrophilicity, biocompatibility, and similarity to natural tissues due to hydrophilic functional groups (hydroxyl, amide, carboxylic), sulfonate groups, and polymer chains[14].

Concerns over hydrogels arise over their potential environmental impact and limitations due to their prolonged usage, toxicity, and inability to biodegrade. Today, hydrogel production focuses mainly on utilizing natural polymers due to their non-toxic nature, excellent biocompatibility, and inherent ability to undergo natural degradation. Hydrogels can be synthesized via graft copolymerization to combine synthetic monomers with natural polymers. Some of the most widely studied natural polymers are cellulose[15], starch[16,17], gelatin[18], chitosan[19], carrageenan[20] and protein[21].

Hydrolyzed collagen (h-collagen) is a natural polymer that can be derived from the hydrolysis of solid-tanned leather waste[22]. Several previous studies have shown that h-collagen can be developed into hydrogels. Pourjavadi et al.[23] produced hydrogels by graft copolymerization of h-collagen with AA; Pourjavadi and Kurdtabar[24] made hydrogels using graft copolymerization techniques using h-collagen, AA and Aam. Marandi et al.[25] obtained hydrogels from the graft copolymerization process of h-collagen with 2-acrylamido-2-methylpropane sulfonic acid and acrylamide. Graft copolymerization was chosen as the preferred method because it can modify the chemical and physical characteristic of natural polymers[26,27]. Hide trimming waste, a by-product of the tannery process, constitutes over 75% of the solid waste produced by tanneries. The utilization of hydrolyzed collagen derived from hide trimming waste may reduce the solid waste produced by this industry.

To our knowledge, research on the utilization of h-collagen derived from hide trimming waste has not been reported in the literature except by Purba et al.[22], who discovered that h-collagen derived from hide trimming waste has significant promise as a polymer for development in Indonesia. Therefore, this study aimed to synthesize a superabsorbent hydrogel from hydrolyzed collagen protein. In addition, this study also examined the impact of various reaction variables on the hydrogel's ability to absorb water and its swelling behavior in different salt solutions and pH levels.

2. Materials and Methods

2.1 Tools and material

H-collagen was synthesized following the method described by Purba et al.[22]. Acrylic acid (AA), 96% ethanol, potassium persulfate (KPS), NaCl, KCl, $MgCl_2$, $CaCl_2$, $BaCl_2$, NaOH, methylene bisacrylamide (MBA), HCl, and distilled water were purchas from Merck Co., Germany, and were utilized without undergoing additional purification. The tools used were a three-neck reactor, magnetic stirrer, thermostatic water bath, vacuum machine, oven, nylon cloth, Scanning Electron Microscopy (FEI Inspect S50), pH meter, and Fourier-transform Infrared Spectroscopy (Shimadzu IR Prestige-21).

2.2 Hydrogel preparation

H-collagen (1.33 g) was dissolved in 40 mL of distilled water, put into a three-neck reactor with a mechanical stirrer (300 rpm), and submerged in a thermostatic water bath at 80 °C. Next, a specific quantity of 70% neutralized AA (1.5 - 8.0 g in 5 mL of H_2O) was introduced and agitated for 10 minutes. Subsequently, the crosslinker solution, consisting of 0.1 - 0.2 g of MBA dissolved in 5 mL of H_2O, and the initiator solution, including 0.01-0.35 g of KPS dissolved in 5 mL of H_2O, were sequentially introduced. The reaction occurred for 60 minutes and a speed of 300 rpm at 80 °C. All reactions were carried out under vacuum. The formed superabsorbent was submerged in 200 mL ethanol and left overnight. The product was then reduced in size using scissors and washed using fresh ethanol. The superabsorbent was dried at 50 °C for 12 hours in a drying oven. Once the sample was dried, it was ground to obtain a powdered product and kept in a location shielded from moisture, high temperatures, and light.

2.3 Swelling capacity in distilled water

A total of 0.2 g superabsorbent was put in a tea bag constructed from nylon fabric with a mesh size of 100. The tea bag was fully submerged in distilled water (200 mL) for 12 hours at ambient temperature. After that, the tea bag was suspended in air for 15 minutes to remove the excess solution. The formula in Equation 1 was utilized to measure equilibrium swelling (ES).

$$ES\left(g/g\right)=\frac{Weight\ of\ expanded\ gel-Weight\ of\ dry\ gel}{Weight\ of\ dry\ gel} \quad (1)$$

2.4 Swelling capacity in various salt solutions

The impact of charge screening and ionic crosslinking on the disappearance of intense swelling in various salts has been investigated[24]. Hydrogel's capacity to expand in KCl, NaCl, $CaCl_2$, and $MgCl_2$ solutions at a concentration of 0.15 mol/L was calculated using Equation 1.

2.5 Swelling capacity in various pH

Solutions with pH values of 1 to 13 were achieved by diluting NaOH (pH 13) and HCl (pH 0.1) solutions to obtain pH values of 6.0 or lower. The pH value's precision was assessed using a pH meter. A total of 0.1 g of dry sample was utilized to assess swelling capacity using Equation 1.

2.6 Instrumental analysis

Fourier Transform Infrared Spectroscopy (FTIR) was employed to ascertain the existence of cross-linking between monomers in the superabsorbent. Observations of the superabsorbent morphology were also conducted using SEM. A coating of gold-palladium alloy was applied to the dry hydrogel powder and then examined using an SEM instrument.

2.7 Experimental design

Various variables influenced hydrogel's swelling capacity. The variables in question pertain to the crosslinker (MBA) and the monomer (AA) concentration. Optimizing each of these variables is necessary to obtain a hydrogel with maximum water absorption capacity. First, a search was conducted for the best MBA crosslinker concentration with a fixed monomer variable value. Second, a search for the best AA monomer concentration with a value for the crosslinker concentration variable derived from the optimal value in the first stage. Once the ideal value for each variable had been determined, the hydrogel product was produced and analyzed.

3. Results and Discussions

3.1 H-collagen

The extraction process of h-collagen from hide trimming waste has been published in previous studies[22]. The h-collagen used has a molecular weight ranging from 23-26 kDa and consists of various types of amino acids with the largest concentrations of glycine (30.01%), proline (15.26%), arginine (10.19%), and glutamic acid (8.96%). The IC_{50} value of h-collagen was 238.5 ppm.

3.2 Synthesis and characterization

The superabsorbent was produced by grafting copolymerization of AA monomer onto the hydrolyzed collagen backbone. KPS was used as an initiator and MBA as a crosslinking agent. KPS is a thermal dissociation initiator that converts to sulfate anion radicals at 80°C[28]. Subsequently, the anion radical removes a hydrogen atom from one of the functional groups (COOH, NH_2, OH, and SH) present on the substrate side chain, forming the corresponding radical. The macro-radical initiates the grafting process of 70% neutralized acrylic acid onto the h-collagen backbone. Furthermore, MBA facilitates a crosslinking reaction, forming a three-dimensional network, i.e. a crosslinked hydrogel copolymer.

FTIR was utilized to verify the chemical structure of the h-collagen and superabsorbent (Figure 1). The stretching of the hydroxyl groups in h-collagen results in the wide peak observed in a wavelength range of 3200-3600 cm^{-1}.

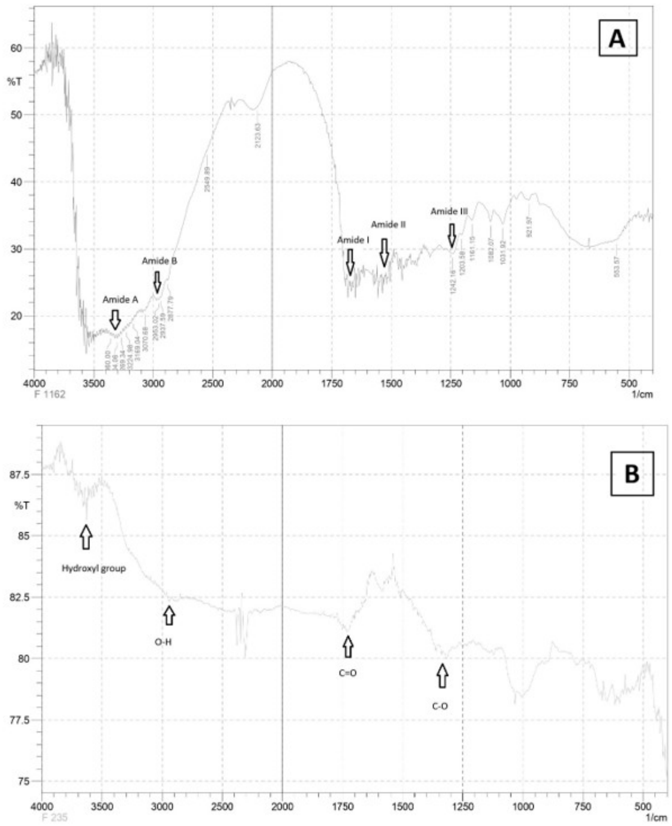

Figure 1. FTIR spectrum of h-collagen (A) and superabsorbent hydrogel (B).

The presence of these absorption bands at 1320-1210 cm^{-1} for C-O, 1760-1690 cm^{-1} for C=O, and 3300-2500 cm^{-1} for OH. confirm that AA monomer grafting has occurred on h-collagen.

FTIR spectra show a wide peak in the 3200-3600 cm^{-1} band, indicating O–H stretching vibrations from hydroxyl groups generating hydrogen bonds. These groups are mostly hydroxyproline residues and water molecules intercalated in the triple-helix structure. Hydroxyl groups affect collagen's solubility, viscoelasticity, and biomolecule interactions. Shoulders and Raines (2009) showed that hydroxyproline-mediated hydrogen bonding increases collagen heat stability[29]. Knot and Bailey (1998) found that hydroxylation levels affect collagen crosslinking density and mechanical strength[30].

3.3 Effect of MBA concentration

The effect of the crosslinker concentration on the ability of the hydrogel to swell is presented in Figure 2. The highest absorption capacity of 156 g/g was attained at a crosslinker concentration of 0.14 g. MBA serves primarily as a crosslinking agent, facilitating the formation of covalent bonds between polymer chains to establish a three-dimensional network. An increase in MBA concentration enhances crosslinking density, resulting in improved mechanical strength and elasticity[31]. A higher crosslinking density diminishes polymer chain mobility, leading to the formation of stronger hydrogels that exhibit increased resistance to deformation. In this study swelling capacity and MBA concentration have a negative correlation. A higher concentration of crosslinker increased the crosslink density making the rigid crosslinked structure inextensible and retaining significant water. Furthermore, enhanced crosslinking imposes further limitations within the polymer network, diminishing the capacity of hydrophilic groups to engage with water. This phenomenon has been reported by several researchers[2,32,33].

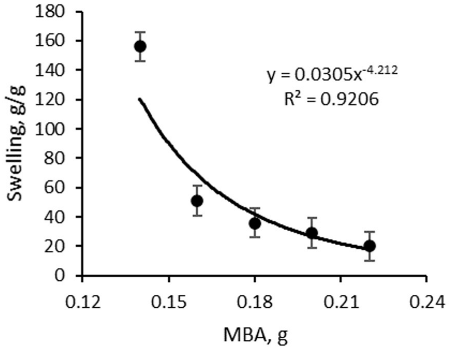

Figure 2. The effect of crosslinking concentration on the swelling capacity of superabsorbent hydrogel. Reaction conditions: h-collagen 1.33 g, AA 4.7 g, KPS 0.15 g, 80 °C for 60 minutes.

The concentration of MBA directly impacts the porosity of the hydrogel matrix, thereby influencing its diffusion characteristics. Lower MBA concentrations lead to increased pore sizes, facilitating the diffusion of molecules such as nutrients or drugs, whereas higher concentrations yield smaller pores, limiting diffusion[34]. Over-crosslinking with agents like methylene bisacrylamide (MBA) restricts hydrogel expansion, reducing swelling, while under-crosslinking affects mechanical stability[35-37].

Table 1 shows the swelling capacity of several hydrogels produced from natural materials. Compared to other study the swelling capacity of our hydrogel is lower. It is believed to be attributed to disparities in the animal species and tissues employed. Furthermore, these variations may result from varying levels of partial neutralization of AA. Neutralizing carboxylic groups prior to grafting enhances swelling capacity by improving ionic interactions and osmotic pressure[36,46,47]. Various crosslinking compositions and monomer ratios. The optimal concentrations of crosslinker and acrylic acid ensure a balance between network flexibility and structural integrity[35,46]. Varied reaction conditions. The polymer uniformity is significantly influenced by temperature, initiator concentration, and reaction time, which help to prevent undesirable side reactions like phase separation[35,43,44]. Other researchers utilized co-monomers like AMPS. The application of IA and AMPS enhanced salt tolerance and water absorption[42,46,48].

Figure 2 shows a power law link between ES capacity and MBA concentration. K and n represent constants specific to each superabsorbent hydrogel. The *n* shows the hydrogel's sensitivity to crosslinker concentration. The *K* value functions as a standard for assessing the extent of swelling at a constant level of crosslinker concentration. In other words, a higher *K* value corresponds to a greater capacity for swelling. The curve fit to Equation 2 yielded the values of $K = 80.621$ and $n = 4.2$.

$$ES = K\left[MBA\right]^{-n} \tag{2}$$

3.4 Effect of monomer concentration

The correlation between the monomer and swelling capacity was examined by altering the concentration of AA (Figure 3). The findings indicated a significant rise in absorption as the concentration of AA increased, followed by a subsequent decline. There was an increase in water absorption capacity in the first half of the experiment. This may be due to the increased hydrophilicity of the superabsorbent and the AA molecules around the h-collagen macro-radicals. There is a decrease in the water absorption capacity after the maximum point. This is thought to occur due to: (a) increased chain transfer to the AA molecule, (b) increased viscosity, which causes limited movement of the reactants and deactivates the growing macro-radical chains immediately after their formation, and (c) increased graft copolymerization reactions. Other researchers have also reported similar conclusions[49,50]. High grafting introduces rigid structures, limiting network expansion and water uptake[41,43].

3.5 Swelling capacity in various salt solutions

Understanding the swelling behavior of hydrogels in salt solutions is crucial, particularly in relation to their uses in agriculture and horticulture. The properties of the external solution, specifically its charge and ionic strength, primarily influence the swelling ratio. The polymer's inherent characteristics, such as its ability to form a flexible network, hydrophilic functional groups, and cross-linking density, also influence it. There is a negative correlation between the water absorption capacity and the ionic strength of a salt solution.

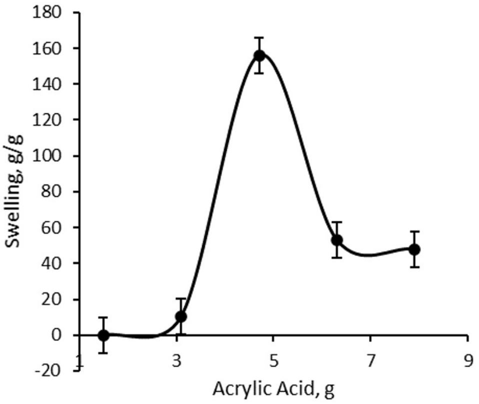

Figure 3. Effect of monomer concentration on swelling capacity of superabsorbent hydrogel. Reaction conditions: h-collagen 1.33 g, MBA 0.14 g, KPS 0.15 g, 80 °C for 60 minutes.

Table 1. Swelling capacity of several hydrogels produced from natural materials.

References	Backbone	Monomer	Water absorbency
Wu et al.[38]	Potato starch	Acrylamide (AM)	4.000 g/g
Yoshimura et al.[39]	Chitin	Succinic anhydride	300 g/g
Pourjavadi et al.[23]	H-collagen	AA	920 g/g
Zhang et al.[40]	Chitosan	AA	159.6 g/g
Pourjavadi et al.[41]	H-collagen	AMPS	342 g/g
Pourjavadi et al.[42]	H-collagen	AA, AMPS	~500 g/g
Pourjavadi & Salimi[43]	H-collagen	AA, HEMA	364 g/g
Sadeghi & Hosseinzadeh[44]	Kappa carrageenan	HEMA	Not tested
Marandi et al.[25]	H-collagen	AMPS, AM	~650 g/g
Saarai et al.[45]	Sodium alginate, gelatin	-	Swelling capacity ~400%
Sadeghi et al.[20]	Carrageenan	AA	370 g/g
Ibrahim et al.[18]	Gelatin	AM	Swelling capacity >800%
This research	H-collagen	AA	156 g/g

Higher levels of ions in a solution reduce the osmotic pressure between the hydrogel and the salt solution. Hydrolyzed collagen possesses charged functional groups, such as carboxyl and amine, which are essential for ionic interactions in the hydrogel matrix. Salt solutions introduce cations and anions that interact with charged groups, thereby altering electrostatic interactions and the organization of the matrix. Furthermore, the extra cation screening effect on the anionic group results in poor electrostatic repulsion of anions and lowers water absorption[51]. In addition, ionic cross-linking formed when the hydrogel is in a solution containing multivalent cations causes a decrease in water absorption capacity.

Various chloride salt solutions have different ionic strengths. Figures 4 and 5 present their effects on the hydrogel swelling capacity. An elevated cation charge leads to a higher level of cross-linking. Consequently, the ability to swell reduces. Hydrogels have a higher capacity to absorb monovalent cations than divalent cations in salt solutions. Consequently, the absorption in salt solutions is greater for K^+ and Na^+ than Ca^{2+} and Mg^{2+}. Monovalent ions primarily influence swelling by disrupting hydrogen bonding, whereas divalent ions enhance the matrix through ionic bridging. Monovalent salts decrease swelling via charge screening, while multivalent ions (Ca^{2+}, Al^{3+}) cause ionic crosslinking, leading to significant swelling reductions[36,41,46,51,52].

A dimensionless salt sensitivity factor (*f*) was also calculated for a salt solution at a concentration of 0.15 mol/L using Equation 3[53]. In this equation, *Sg* represents the swelling in a specific fluid, while *Sd* represents the swelling in deionized water. Table 2 displays the results. The *f* factor illustrates that the salt sensitivity is greatly influenced by the charge and radius of the cations added into the medium. The degree of cross-linking is directly proportional to the charge and radius of the cation, thus increasing the *f* factor. This experiment demonstrates that a hydrogel's ability to expand in a salt solution is consistent with its ability to expand in distilled water.

$$f = 1 - \frac{S_g}{S_d} \qquad (3)$$

3.6 Effect of pH on ES

Figure 6 displays the equilibrium swelling behavior of the superabsorbent material when subjected to different pH levels. The swelling capacity of the anionic superabsorbent exhibited a decrease in the presence of cations in the medium. Consequently, buffer solutions were not employed. The desired basic and acidic pH levels were achieved by diluting stock solutions of NaOH (pH 13.0) and HCl (pH 1.0).

The highest degree of swelling was achieved at a pH of 7. At pH levels below acidity, most carboxylate anions become protonated, causing the elimination of the principal repulsive forces between anions. Consequently, the superabsorbent's ability to swell is reduced. Some carboxylate groups are ionized at pH levels between 5 and 7. This results in the electrostatic repulsion of the COO- groups, which in turn enhances their swelling capacity. Cations' presence restricts the material's expansion when the pH is over seven due to the charge-screening phenomenon. Comparable findings have been documented in other alternative hydrogel systems[23,54-56].

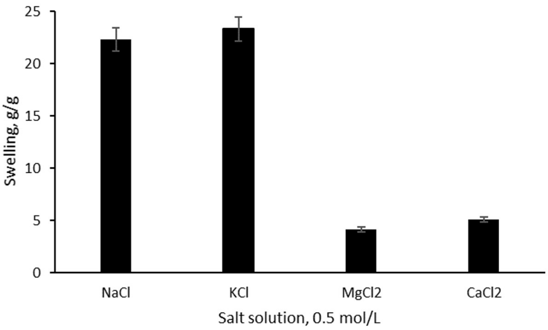

Figure 4. Swelling capacity of superabsorbent hydrogel in several salt solutions of equal concentration (0.15 mol/L).

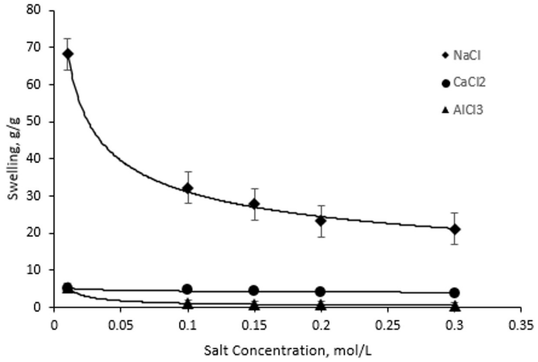

Figure 5. Swelling capacity of superabsorbent hydrogel in various salt solutions with different concentrations.

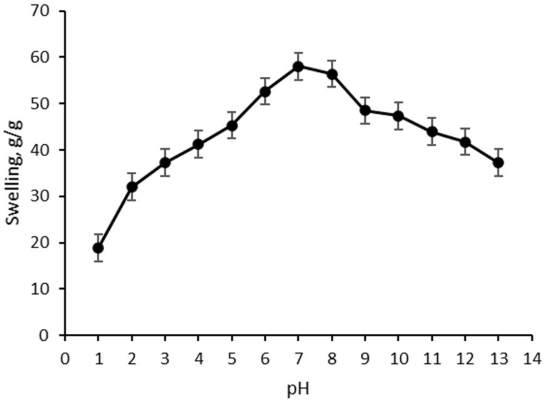

Figure 6. Effect of pH on the swelling capacity of superabsorbent hydrogels.

Table 2. Swelling capacity in distilled water and various salt solutions (0.15 mol/L) and salt sensitivity factor (*f*).

	ES (g/g)	*f*
H_2O	156	-
NaCl	27.80	0.82
$CaCl_2$	4.56	0.97
$AlCl_3$	0.78	1.00

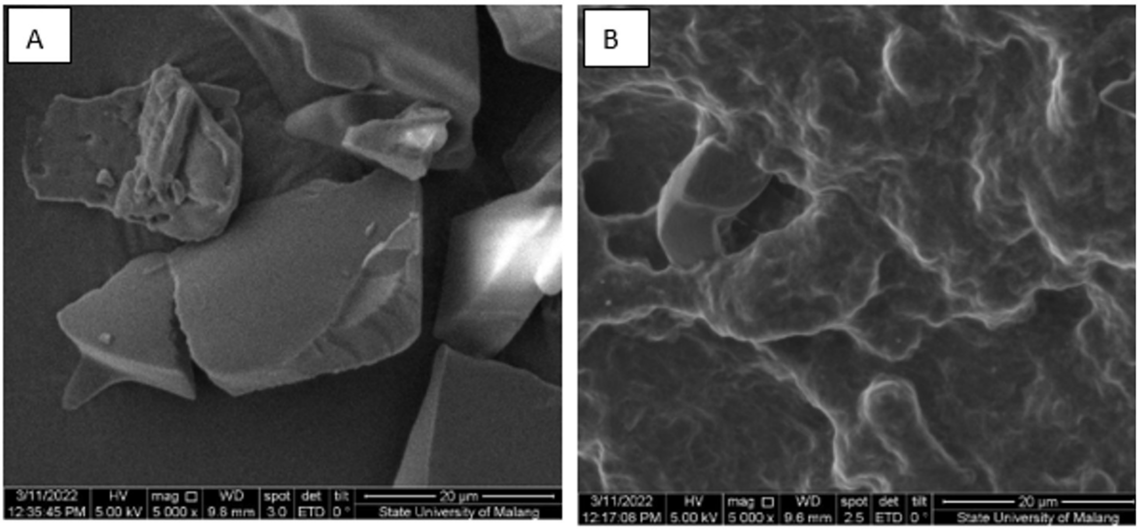

Figure 7. SEM results of hydrolyzed collagen (A) and superabsorbent hydrogel (B).

3.7 Scanning Electron Microscopy (SEM)

Microstructural morphology is a crucial property that must be considered when evaluating hydrogels. SEM identifies pore size, surface roughness, and network uniformity as critical structural parameters that influence water and nutrient retention and release rates[57]. Research indicates that smaller, denser pores are associated with a slower release rate but enhanced encapsulation, whereas porous networks promote a more rapid swelling-mediated release[57].

Figure 7 depicts the SEM results of h-collagen and optimized hydrogel. The h-collagen and hydrogel that were evaluated were in dry preparations. This study examined hydrogel specimens in their dry state before any expansion or swelling. The drying process of the swollen hydrogel influences the results of the SEM test. Freeze-drying effectively maintains the porous microstructures in hydrolyzed collagen-derived or similar protein-based superabsorbent materials, yielding superior water retention properties relative to air-drying or vacuum-drying, which lead to pore collapse and diminished swelling capacity[58].

Despite the absence of commonly employed drying procedures such as lyophilization or vacuum drying, SEM analysis confirms that the superabsorbent has a porous structure. These pores were most likely produced during the ethanol dewatering step and subsequent evaporation of ethanol from the hydrogel matrix during the drying process[59]. They are thought to be regions of water permeation and points of contact between environmental stimuli and the graft copolymer's hydrophilic groups.

4. Conclusion

A new superabsorbent hydrogel has been successfully manufactured via graft copolymerization of AA to h-collagen obtained from hide trimming waste. The optimal reaction conditions for achieving maximum swelling capacity of 156 g/g were a dosage of 0.14 g of MBA and 4.7 g of AA. The hydrogel's swelling capacity was found to be maximum in the KCl solution, as observed by measuring hydrogel swelling in various salt solutions. The hydrogel displayed significant pH sensitivity, leading to observable changes in swelling across a broad pH spectrum (1-13). The study using the FTIR instrument confirmed that the grafting of AA monomer had taken place on the h-collagen backbone. Simultaneously, SEM verified that the hydrogel produced had a porous architecture.

5. Author's Contribution

- **Conceptualization** – Febriani Purba.
- **Data curation** – Febriani Purba.
- **Formal analysis** – Alan Dwi Wibowo.
- **Funding acquisition** – Arief Rahmad Maulana Akbar.
- **Investigation** – Febriani Purba; Raihan Sari Afifah.
- **Methodology** – Febriani Purba.
- **Project administration** – Agung Cahyo Legowo.
- **Resources** – Hairu Suparto.
- **Software** – NA.
- **Supervision** – Agung Nugroho.
- **Validation** – Agung Nugroho.
- **Visualization** – Hairu Suparto.
- **Writing – original draft** – Febriani Purba.
- **Writing – review & editing** – Arief Rahmad Maulana Akbar; Agung Cahyo Legowo; Alan Dwi Wibowo; Agung Nugroho; Hairu Suparto; Raihan Sari Afifah.

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