

Synthesis and Properties of Cellulose-based Superabsorbent Hydrogel by a New Crosslinker

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Abstract: Cellulose-based superabsorbent hydrogel (SAH) has been successfully prepared by the grafting method with cellulose and sodium acrylate as well as aluminum hydroxide as a novel crosslinker. The factors influencing water absorption such as the neutralization degree of acrylic acid (AA), the amount of initiator ammonium persulfate (APS) and crosslinker, and the proportion of cellulose to AA were investigated. The chemical structure and thermal behavior of the SAH were analyzed by fourier-transform infrared spectrometry, field-emission scanning electron microscopy, and thermal gravimetry, respectively. Results showed that the water absorbency of the SAH in distilled water and 0.9 wt% NaCl solution reached 922 and 118 g/g when the mass ratio of cellulose to AA, APS to AA, Al(OH)₃ to AA, and the neutralization degree of AA was 39 wt%, 0.67 wt%, 0.89 %, and 76 %, respectively. Under the same condition, the hydrogel synthesized by traditional crosslinker N,N-methylenebisacrylamide was only 530 and 84 g/g in distilled water and 0.9 wt% NaCl solution, respectively. The mechanism of polymerization reaction was analyzed. And the cost and feasibility for the mass-producing the hydrogel were analyzed.

Keywords: Superabsorbent hydrogel, Crosslinker, Cellulose, Acrylic acid, Water absorption

Introduction

Superabsorbent hydrogel (SAH), which can absorb water at hundreds or even thousands of times of its weight, is widely applied in agriculture, horticulture, and sanitary goods such as disposable diapers and feminine napkins, sealing composites, artificial snow, drilling fluid additives, fighting fire, and drug delivery [1-4]. The SAH not only has many hydrophilic groups such as carboxyl, hydroxyl, and amino groups, but also exhibits a large number of spatial netted structures [5-7]. The SAH is mainly synthesized by using starch [8] or cellulose [9] with acrylonitrile [10], acrylate [11], citric acid [12], polyethylene glycol [13] and so forth. However, starch as a raw material for SAH also serves as food for humans, which threatens the supply of human food. By contrast, cellulose has many advantages, such as renewability, easy biodegradability, and highly abundant natural resources. In addition, cellulose also contains numerous hydroxyl groups, which is beneficial to the formation of more stable hydrophilic network structure with monomer. Therefore, it has been concerned by many researchers [14,15]. To establish the netted structure of SAH, many crosslinkers such as trimethylolpropane triacrylate, N,N-methyl-enebisacrylamide (MBA), polyethylene glycol, and maleic acid have been reported [5,16]. And the reaction system is conducted in atmosphere of nitrogen. These crosslinkers belong to organic compounds, which usually originates from fossils. It is necessary to search for a new crosslinker due to the nonrenewable fossils. Qi *et al.* [17] and Liu *et al.* [18] reported the inorganic compounds

(sodium tungstate and aluminum sulfate) as a crosslinker for the synthesis of SAH. However, the studies for the synthesis of SAH with aluminum hydroxide (Al(OH)₃) as crosslinkers are very little.

In this work, some inorganic compounds as a crosslinker have been investigated. It can be found that the cellulose-based SAH synthesized with Al(OH)₃ as a new and low-cost inorganic crosslinker performed a high absorbing water capacity compared with that of MBA. The maximum absorbing water capacity of SAH reached 922 and 118 g/g in distilled water and 0.9 wt% NaCl solution, respectively. Importantly, the preparation cost markedly decreased.

Experimental

Materials

Cellulose was purchased from Shandong Ehua Pharmaceuticals Co., Ltd. AA, Al(OH)₃, sodium chloride (NaCl), ammonium persulfate (APS), sodium hydroxide (NaOH), aluminum sulfate (Al₂(SO₄)₃), ferric hydroxide (Fe(OH)₃), acetone, and MBA were analytical grade and purchased from the Sinopharm Chemical Reagent Co. Ltd.

Hydrogel Preparation

Neutralization of AA

At an AA-to-NaOH ratio of 76 %, sodium acrylate (AcNa) can be prepared based on the requirement of the experiment. About 2.0 mol/l NaOH was slowly added to AA in an ice-water bath.

Preparation of SAH

SAH was prepared in a three-necked flask equipped with a stirrer. In a typical procedure, 3.5 g of cellulose, 0.08 g of Al(OH)₃, and 10 ml of distilled water were mixed in a flask

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using a magnetic stirrer. The mixture was well distributed and heated to 60 °C. Then, 0.06 g of APS and 9.0 g of AcNa (76 % neutralization degree) was added to the reaction system, which was continuously heated to 85 °C. The reaction was maintained under this condition. When some sticky matter appeared in the reaction system, heating was stopped. Finally, the samples were filtered and washed with distilled water several times, and dried at 85 °C overnight. Based on the same prepared procedure, the control experiments were conducted in the absence of Al(OH)₃, MBA, Al₂(SO₄)₃, and Fe(OH)₃ as a crosslinker, and the control experiment was conducted in the absence of cellulose.

Measurements and Analysis

Water absorption is defined as a water mass absorbed by a unit mass of hydrogel at a certain time [19]. The absorption capability of SAH was placed in an amount of distilled water or 0.9 wt% NaCl solution at room temperature for 24 h. The quantity of water absorption (Q) was calculated by the following equation (1):

$$Q = (m_2 - m_1)/m_1 \quad (1)$$

where m_1 and m_2 are the weight of dried hydrogel and swollen hydrogel (g), respectively. All values were the average of three determinations.

Water-holding Capacity (WHC) of SAH

To evaluate the WHC of SAH, acceleration experiments were conducted by the centrifugal method at a speed of 4000 r/min for 10 min. The WHC of the hydrogel after centrifugation was expressed as follows:

$$\text{WHC} = (M_2/M_1) \times 100 \% \quad (2)$$

where M_1 and M_2 are the weight of swollen SAH and the weight of SAH after centrifugation, respectively. All values were the average of three determinations.

Grafting Ratio (Gr) of SAH

The purified SAH was pretreated with pure acetone and distilled water. Then, the sample was dried at 110 °C overnight. According to the previous literatures [20,21], the Gr of SAH is determined by the following equation (3):

$$\text{Gr} = (w_1/w) \times 100 \% \quad (3)$$

where w_1 and w are the weight of purified graft polymer and crude graft polymer, respectively.

Characterization Methods

Fourier-transform infrared (FTIR) spectra were obtained within the wavenumber range of 4000-400 cm⁻¹ on a spectrophotometer (BRUKER TENSOR 27) operating at a resolution of 0.2 cm⁻¹ by averaging 16 scans. The KBr pellet method was used. Namely, 1.5 mg of SAH was uniformly ground with 100 mg of KBr (<2 μm sample size). Then, the mixture was pressed in a standard device. The background

spectrum of spectroscopy-pure KBr was subtracted from that of the sample spectrum. The morphology of SAH was analyzed using field-emission scanning electron microscopy (FESEM JEOL JSM-7001F). The thermal behavior of SAH was monitored with a NETZSCH STA409C analyzer. About 10 mg of sample was placed in a thermobalance. The temperature was increased to 800 °C at a heating rate of 10 °C·min⁻¹ under a nitrogen flow of 100 ml·min⁻¹. The percentage of weight loss was calculated for different temperature intervals, and the weight at the beginning of each interval was considered as the initial weight.

Results and Discussion

Comparison of Different Crosslinkers on Q Value

MBA is often used as a crosslinker to synthesize hydrogels [22,23]. Given the existence of unsaturated C=C and C=O bonds in the structure of MBA, it easily reacts with some groups such as hydroxyl and aldehyde groups in cellulose. In this work, the inorganic compounds Al(OH)₃, Fe(OH)₃, and Al₂(SO₄)₃ were used to act as crosslinkers. The results are shown in Figure 1.

It is easily found that the synthesized hydrogels have different Q value. They were affected by the type and amount of crosslinker. The SAH with Al(OH)₃ as a crosslinker exhibits the highest water absorbency. Considering that the crosslinkers differed, the grafting ratio is also different. The Gr of SAH affected the formation of spatial network structures. The high Gr of SAH leads to the increase of the degree of polymerization for the hydrogel formed. Therefore, the gap among the backbones became out of order, resulting in the inferior swelling capacity. By contrast, with the low Gr, the netted texture was ineffectively formed. The optimum Gr of SAH is 85.3 % and the maximum Q value of SAH is 922 g/g when the mass ratio of Al(OH)₃ to AA is 0.9 wt%. When the mass ratio of MBA to AA is 0.44 wt%, the Gr of SAH is 79.7 % and the Q value of SAH is 380 g/g, respectively. An optimum amount of crosslinker

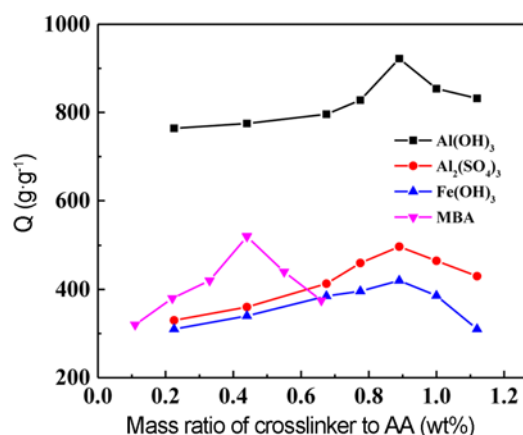


Figure 1. Effect of type of crosslinkers on Q value of SAH.

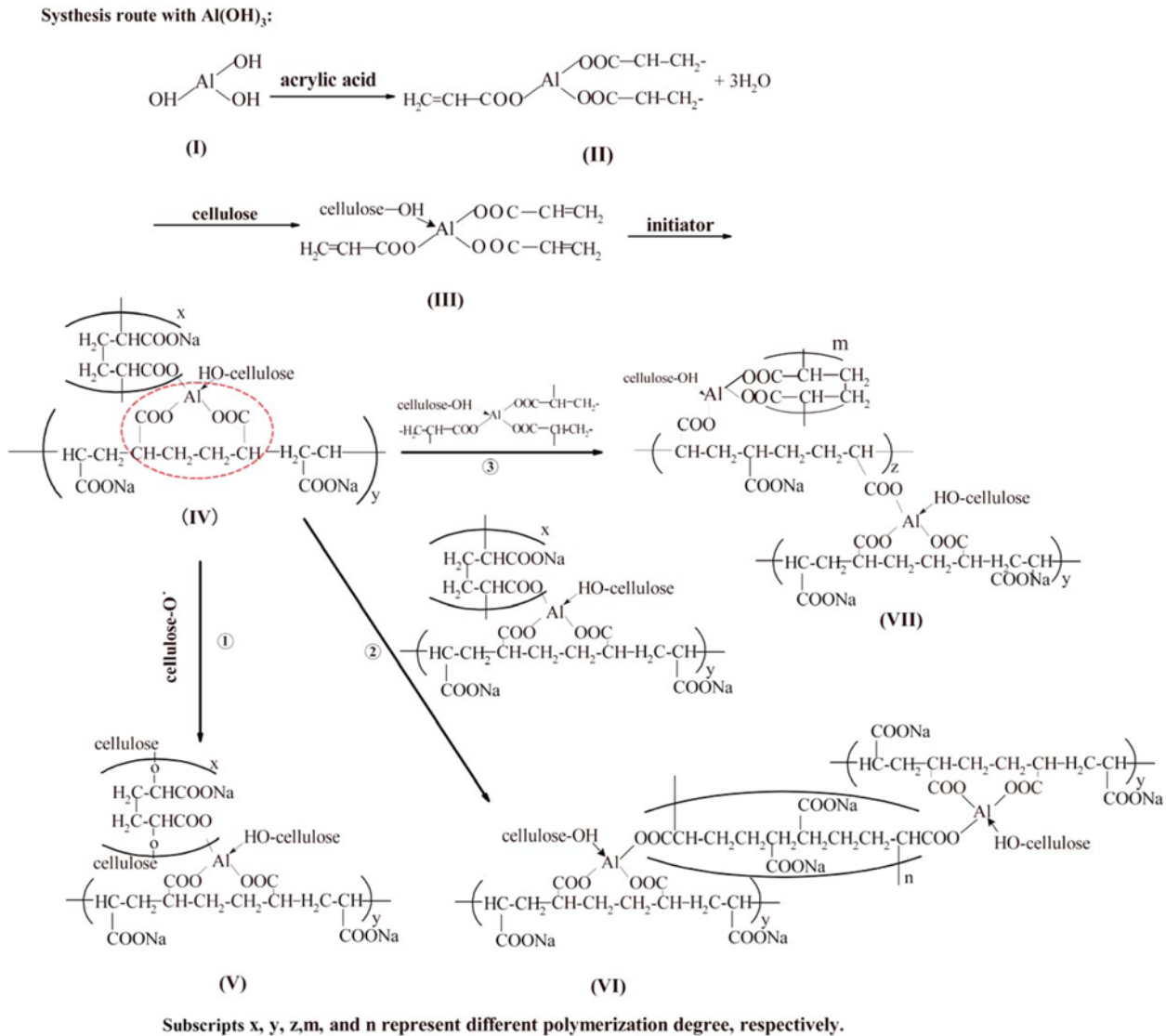


Figure 2. Proposed reaction mechanism for the synthesis of SAH.

was necessary [24]. In addition, the control sample indicated that the SAH could not be synthesized in the absence of $\text{Al}(\text{OH})_3$ and other crosslinkers.

For $\text{Al}(\text{OH})_3$, the optimal mass ratio to monomer AA was 0.89 wt%. The effect of $\text{Al}_2(\text{SO}_4)_3$ as a crosslinker was close to that of MBA. $\text{Al}(\text{OH})_3$ contains three hydroxyl groups and is an electron-defect compound, thereby leading to the easy reaction with AA and AcNa and formation of hydrogel with a stable network structure containing lots of polar groups. The possible reaction route is given in Figure 2.

As shown, the coordination bond between hydroxyl in cellulose and Al atom can be formed. Since the product (III) and (IV) is unstable, polymerization reaction such as path 1, 2 and 3 may take place in the existence of initiator. Finally, possible structures, such as (V), (VI) and (VII) with highly developed network structure, were formed. It can be seen

from Figure 2 that there are numerous polar groups ($-\text{COOH}$ and $-\text{COONa}$) in the SAH structure. By this way, the backbone of carbon chain from cellulose is made great use of. For $\text{Fe}(\text{OH})_3$, it is not an electron-defect compound, thereby resulting in the fact that the coordination bond cannot be formed through oxygen in cellulose and Fe. The skeleton structure of carbon chain from cellulose is not effectively used. That is to say, the network structure of SAH with abundant space cannot be effectively formed. Li et al [25] reported that the hydrogel synthesized by acrylic acid and acrylamide in the presence of attapulgite has a high Q value (1400 g in distilled water). From the composition of attapulgite, we knew that it also contains Al atom [25]. Thus, Al plays an important role in enhancing the Q.

In order to examine the synthetic effect of MBA as a crosslinker on Q value of hydrogel in the existence of

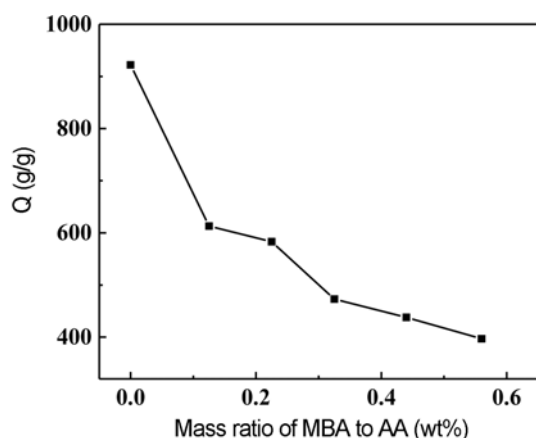


Figure 3. Influence of combination MBA and $\text{Al}(\text{OH})_3$ on Q value of SAH (the mass ratio of $\text{Al}(\text{OH})_3$ to AA is 0.89 wt%).

$\text{Al}(\text{OH})_3$, the Q of hydrogel synthesized by adding different amount of MBA was also investigated when the mass ratio of $\text{Al}(\text{OH})_3$ to AA was 0.89 wt%. The results are depicted in Figure 3. As shown, Q of hydrogel decreased as MBA was increased. It indicated that there is no synthetic effect between MBA and $\text{Al}(\text{OH})_3$.

Influence of Initiator Amount on Q Value

Free radicals that initiate the chain reaction during polymerization are produced by the initiator. In this work, APS was chosen as an initiator. The influence of APS amount on the Q value of the synthesized SAH is depicted in Figure 4. The maximal Q value of SAH reached 922 g/g when the mass ratio of APS to AA and Gr were 0.67 wt% and 85.3 %, respectively. However, when the mass ratio of APS to AA was 0.78 and 0.89 wt% being higher than 0.67 wt%, the Q value of SAH decreased although the Gr of SAH was 87.6 % and 89.8 %, respectively. This phenomenon was attributed to the fact that excessive free radicals would accelerate the formation of tight networks of SAH [26].

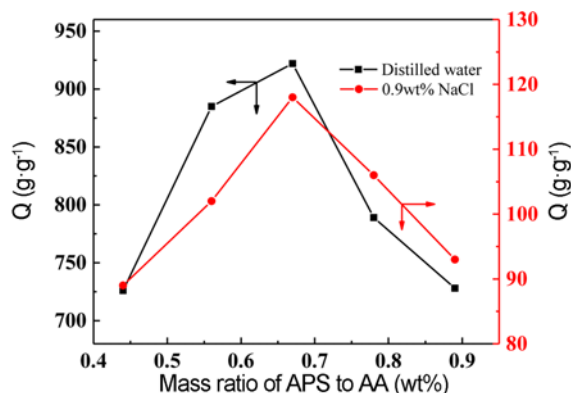


Figure 4. Effect of amount of APS on Q value of SAH.

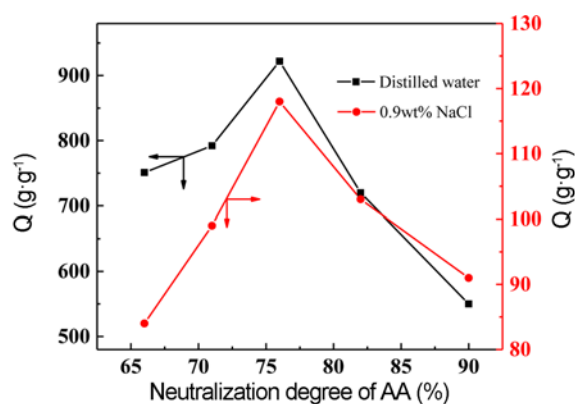


Figure 5. Effect of neutralization degree on Q value of SAH.

Table 1. Influence of different of pH values for the synthesis SAH on Q value

Sample	Neutralization degree of AA (%)	Gr (%)	pH	Q ^a (g/g)	Q ^b (g/g)
SAH	66	81.7	5.5	751	84
	71	82.4	5.7	792	99
	76	85.3	6.6	922	118
	82	87.1	6.8	720	103
	90	89.2	6.9	550	91

^aIn distilled water and ^bin 0.9 wt% NaCl solution.

Influence of Neutralization Degree of AA on Q Value

The influence of different neutralization degree on the Q of the hydrogel was observed. The results are shown in Figure 5. Appropriate neutralization degree of AA improved the Q value of the hydrogel. Figure 5 shows that the Q value of the hydrogel was the highest when the neutralization degree of AA was 76 %.

Considering that AA was neutralized, the number of hydrogen bonds formed among AA greatly decreased. This effect has also been observed by the researchers [27]. As shown in Table 1, when the neutralization degree of AA was 66 %, 71 %, 76 %, 82 %, and 90 %, the Gr of SAH was 81.7 %, 82.4 %, 85.3 %, 87.1 %, and 89.2 % in turn. The Gr of polymerization increased with the increase of neutralization degree of AA, thereby leading to the high inside osmotic pressure of the crosslinking network for SAH [9]. When the pH value increased from 5.5 to 6.6, the Q value of SAH increased initially from 751 to 922 g/g and 84 to 118 g/g in distilled water and 0.9 wt% NaCl solution, respectively. When SAH was synthesized under the acid condition, the Al^{3+} ion was the main existing form, resulting that the structure of SAH was ineffectively formed. When the pH value was under the neutral condition, the $\text{Al}(\text{OH})_3$ was the major existing form which is beneficial to the formation of netted texture of SAH. The reaction mechanism

can be found in Figure 2.

Effect of the Mass Ratio of Cellulose to AA on Q Value

The mass ratio of cellulose to AA significantly influences the Q value of SAH. Figure 6 shows that the maximum Q value of SAH was 922 and 118 g/g in distilled water and 0.9 wt% NaCl solution when the mass ratio of cellulose to AA was 39 wt%. When the mass ratio of cellulose to AA was varied, the amount of hydrophilic groups in the hydrogel was markedly affected. Consequently, the Q value of the hydrogel was also affected due to the low grafting rate of the polymerization reaction, resulting in the formation of loose networks of SAH [28,29]. The control experiment demonstrated that SAH could not be synthesized without cellulose under the same synthesis condition. Therefore, cellulose played an important backbone role for SAH.

Table 2 summarizes the previous results obtained hydrogel with the use of different crosslinker. It is worth noting that the SAH using $\text{Al}(\text{OH})_3$ as crosslinker is superior to previously reported hydrogel. In this work, the maximum Q value is 922 and 118 g/g in distilled water and 0.9 wt% NaCl solution, respectively. However, the Q value of SAH with

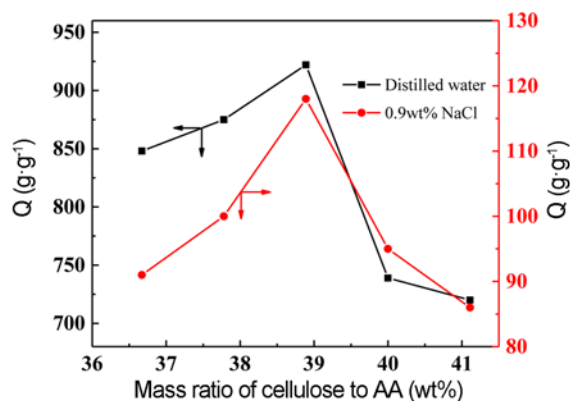


Figure 6. Effect of ratio of cellulose to AA on Q value of SAH.

MBA as crosslinker is 532 and 84 g/g in distilled water and 0.9 wt% NaCl solution, respectively, which is much smaller than that of $\text{Al}(\text{OH})_3$.

FTIR Analysis of SAH

The FTIR spectra of the cellulose and SAH are shown in Figure 7. A considerable difference existed between the FTIR spectra of SAH and cellulose. The characteristic absorption peaks of cellulose were found at 3420, 1430, 1117, and 1068 cm^{-1} , which represented the stretching vibration of O-H, antisymmetric stretching vibration of C-H of the methyl group, and stretching vibration of C-O [14]. For the FTIR spectra of SAH, characteristic absorption peaks appeared at 1645, 1569, 1412, 1451, and 1045 cm^{-1} , which were assigned to the stretching vibration of C=O for carboxylate, bending vibration of C-H, and stretching vibration of C-O [34,35]. The existence of $-\text{COO}^-$ mutual variable structure weakened the absorption peak of C=O, which caused the C=O stretching vibration absorption peak to move to a low wavenumber at 1645 and 1569 cm^{-1} . Thus, the copolymer was grafted through AA and cellulose.

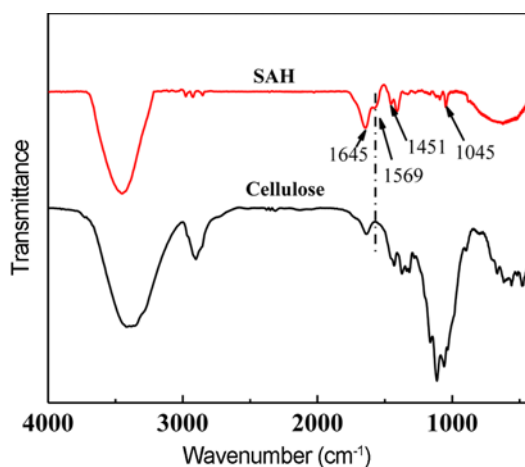


Figure 7. FTIR spectra of cellulose and SAH.

Table 2. Q value of hydrogel by using different crosslinker

Substrate	Cross-linker	Q ^a (g/g)	Q ^b (g/g)	Reference
Wheat straw	MBA	266.8	34.3	[22]
Wheat straw	MBA	417	45	[30]
Wheat straw	MBA	198.5	26.3	[31]
Carboxymethyl cellulose	MBA	750	85	[32]
Carboxymethyl cellulose	MBA	859	72.5	[33]
Chitosan	MBA	770	88	[23]
Cellulose	MBA	390	39.5	[21]
Cellulose	MBA	532	84	This work
Cellulose	$\text{Al}(\text{OH})_3$	922	118	This work

^aWater absorption in distilled water and ^bwater absorption in 0.9 wt% NaCl solution.

Thermoanalysis of SAH

Mass loss can provide useful information on the stability of SAH at different temperatures and reflect the difference in thermal stability between SAH and cellulose. Figure 8(a) shows that the thermal decomposition of SAH occurred in two parts at 197–351 °C and 401–489 °C. This phenomenon indicated that the first stage was due to the dehydration process and decomposition of carboxylic graft chain [36]. The second stage was mainly due to the breakage of C-C bonds and formation of coke structure [16,37]. For cellulose, only one stage of thermal decomposition occurred at 323–401 °C. For SAH, the residue mass accounting for about 46 wt% remained constant after the temperature reached 489 °C.

For cellulose, the residue mass accounting for about 16.8 wt% did not change after the temperature reached 401 °C. The polymerization degree of SAH was much higher than that of cellulose. Therefore, SAH was the multipolymer obtained by the reaction of cellulose and AA using $\text{Al}(\text{OH})_3$ as initiator. The heat variations of the SAH at different heating stages are shown in Figure 8b, and obvious differences can be observed. The maximum heat change occurred at 323–385 °C for cellulose and at 347–388 °C, 423–449 °C, and 614–718 °C for SAH. In other words, endothermic processes almost always occurred for SAH.

This finding indicated that bonded polar groups on the cellulose structure continually decomposed.

Morphology of the Cellulose and SAH

The surface morphology of cellulose and SAH were presented in Figure 9. As shown in Figure 9(a), the surface of cellulose had irregular shape and little wrinkles. The FESEM image (Figure 9(b)) indicated that the surface of SAH was rough and exhibited some porous microstructures. This structure was helpful for the diffusion of water into the polymer network easier and faster, thereby enhancing the water absorption capacity.

Water Absorption Mechanism of SAH

Accurately weighted sample of 0.1 g was immersed into a certain amount of distilled water. Figure 10(a) and 10(b) show the appearance of SAH before and after swelling. The volume of SAH after swelling increased by approximately 280 times compared with that of SAH before swelling. Obviously, the bulk of SAH before swelling was smaller than that of SAH after swelling, demonstrating that the SAH with preferable water absorption was successfully synthesized.

At 4000 r/min centrifugal speed for 10 min, the WHC of SAH was investigated. Results showed almost no change in the Q value of SAH even if SAH was centrifuged. The WHC

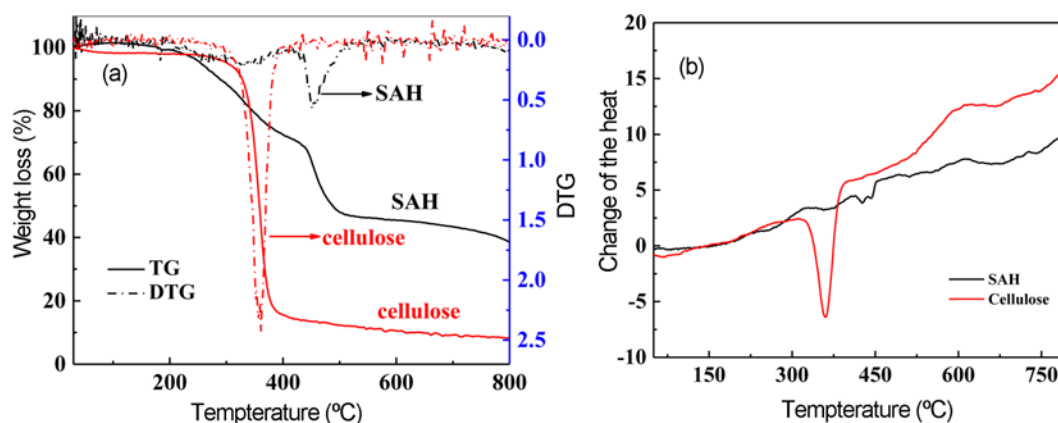


Figure 8. Thermogravimetric curves (a) and variation of the heat (b) for cellulose and SAH.

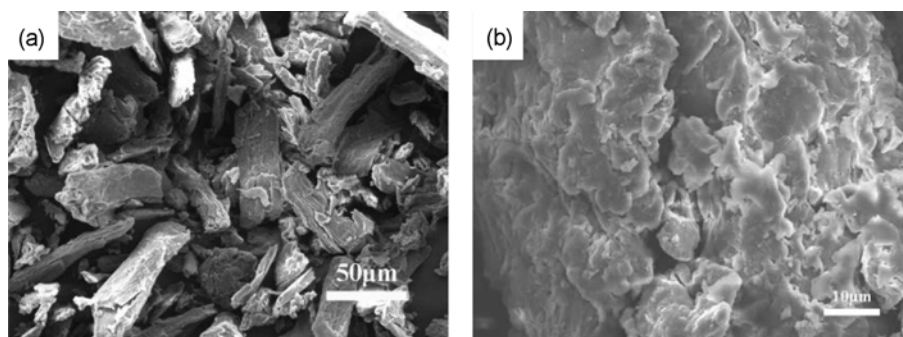


Figure 9. FESEM micrographs of SAH; (a) cellulose and (b) SAH.

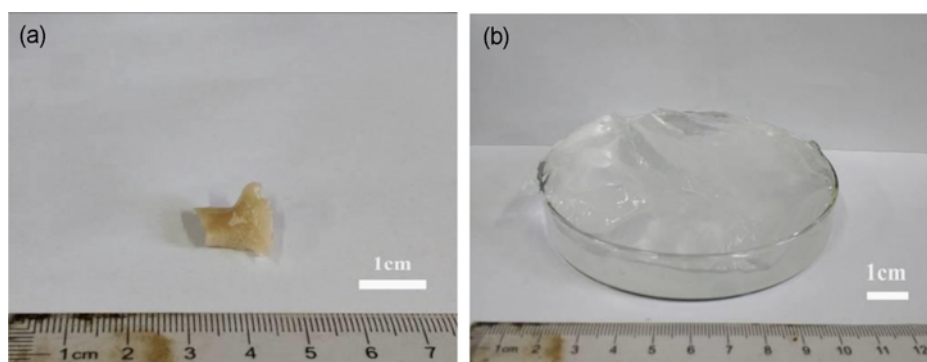


Figure 10. Appearance of SAH before (a) and after swelling (b).

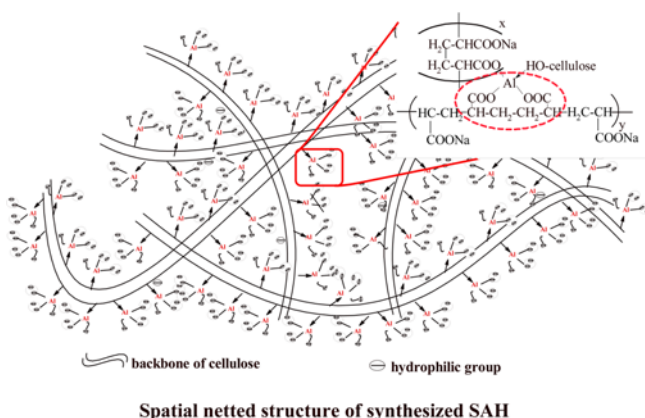


Figure 11. Spatial netted structure of SAH.

remained at 100 %, indicating the excellent water retention for SAH.

SAH is a functional high-molecular-weight material with an appropriate cross-linking degree. Whether the netted structure was formed by mutual cross-linking molecular chains is the key to preparing SAH with a high Q. Therefore, cellulose must be swelled before SAH synthesis so that the crosslinker and AA access and react with the hydroxy in cellulose. The key reason for high Q is that the existence of a great deal of exposed polar groups such as -OH, -CHO, -COOH, and -NH₂, which interacted with water molecules. The relationship between SAH and water is described in Figure 11. As seen in Figure 11, lots of ring structures are a basis of building spatial netted structure, which contains a great deal of oxygen-containing group. These groups distributed on the surface of SAH are beneficial to enhancing the Q value of SAH.

Analysis of Cost and Feasibility of SAH

To analyze the cost and feasibility of SAH, the synthesis conditions were made identically except for the initiators. Based on the market prices of Al(OH)₃ (about ¥1,000 per ton) and MBA (about ¥60,000 per ton), their synthesis

costs were compared. Obviously, the cost with MBA as an initiator for producing SAH was much higher than that with Al(OH)₃. However, the Q value of SAH obtained with Al(OH)₃ as a crosslinker was twice than that of SAH with MBA. Moreover, the percentage of the mass ratio of cellulose to AA was 39 wt%. Thus, cellulose accounted for almost one-third of all components. In addition, no protection gas was used in the synthesis reaction, which differed from the reaction with MBA as a crosslinker. Therefore, the cost of SAH produced by the above description markedly decreased.

Conclusion

With cellulose and AA as materials, SAH was successfully prepared with initiator APS and crosslinker Al(OH)₃. The maximum Q value of SAH in distilled water and 0.9 wt% NaCl solution reached 922 and 118 g/g, when the mass ratio of cellulose to AA, APS to AA, Al(OH)₃ to AA, and the neutralization degree was 39 wt%, 0.67 wt%, 0.89 %, and 76 %, respectively. The grafting ratio of SAH reached 85.3 % under the optimum synthesis condition. SAH was also environmentally and friendly because cellulose can be naturally degraded under atmospheric pressure without any additive. The low-cost Al(OH)₃ was used as a crosslinker alternative to the expensive MBA, and 27.6 % of cellulose was used. Therefore, the cost of the synthesized SAH was markedly reduced. This method has the potentiality to be applied in the mass production of SAH to meet the huge market demand like oilfield development, forestation of the hilly area.

Acknowledgements

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