Article

Swelling Behaviors of Superabsorbent Composites Based on Acrylic Acid/Acrylamide Copolymer and Attapulgite

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Abstract: The swelling behaviors and water retention of superabsorbent sand-fixing materials prepared from acrylic acid/acrylamide copolymer (AA/AM) and acid-modified attapulgite under ultrasonic treatments and different pH conditions were investigated. The results demonstrated that a suitable amount of attapulgite can effectively improve the absorption capacity and saltwater performance. The superabsorbent achieved the highest absorptions of 1257.54 g/g and 209.45 g/g in distilled water and a 0.9 wt% NaCl solution, and a higher water absorbency occurred over a wide pH range of 5~9 when the ultrasonic power was 200 W and the attapulgite content was 10%. The addition of attapulgite could significantly increase the water absorption and retention.

Keywords: polymers; acrylic acid/acrylamide; attapulgite; water absorption; water retention

1. Introduction

Superabsorbent polymers usually possess a three-dimensional, loosely cross-linked network structure containing hydrophilic and hydrophobic parts in an appropriate proportion. They are capable of absorbing and retaining a large amount of water or aquatic solution, from tens to thousands of times their own weight, in a relatively short period of time [1–4]. Due to their unique characteristics, such as swell ability, insolubility, hydrophilicity, and stimuli-responsiveness, applications of superabsorbent materials have been expanded to diverse fields, such as agriculture, horticulture, environmental protection, and desertification control, and these materials have attracted considerable attention. However, most superabsorbents are expensive and environmentally toxic [2,5,6]. Hence, in order to reduce their cost of production, as well as to improve their biocompatibility, renewability, eco-friendliness, non-toxicity, and biodegradability, natural polysaccharides and inorganic clay have often been incorporated into superabsorbents. For instance, they have been prepared with natural materials, such as starch, sodium alginate, chitosan, guar gum, and clay [7–13]. Cellulose is one of the most abundant natural polymers, with excellent biodegradability and biocompatibility. However, the strong intramolecular and intramolecular bonds among the hydroxyl groups along the chain backbones limit the water solubility of cellulose [10,14]. Carboxymethyl cellulose (CMC) is a representative cellulose derivative with carboxymethyl groups bonded to some of these hydroxyl groups [15]. The polar carboxyl groups result in high solubility, excellent chemical reactivity, and strong hydrophilicity. The application of CMC in superabsorbent materials has become an attractive solution to the above problems for various materials scientists.

Attapulgite is a type of natural inorganic mineral material. It possesses a chain structure in which there are a large number of hydrophilic Si-OH bonds, and graft copolymerization can occur with hydrophilic vinyl monomers onto the surface of attapulgite. When attapulgite has been added to a superabsorbent, it improves the salt tolerance and the swelling properties of the materials, and effectively reduces their cost [1,5,8,9,12].
Attapulgite has been shown to polymerize with two acrylamide type monomers and 2-acrylamido-2-methylpropane sulfonic acid, with the product having strong salt tolerance and excellent water absorption capacity, forming inorganic–organic composite superabsorbents [14,16–22].

Previous studies have mainly reported the synthesis of superabsorbents through bulk polymerization, solution polymerization, and reversed-phase suspension polymerization [3,6]. In the research, the present paper, which describes a solution polymerization method, was created to prepare a new sand-fixing material by reacting organic molecules with inorganic, non-metallic clay. The effects of attapulgite and ultrasonic treatment on the water absorption and retention properties of the sand-fixing material were analyzed.

2. Materials and Methods

2.1. Materials

Attapulgite clay was obtained from Tianshui City of Gansu Province, China. It contains minerals such as attapulgite, quartz, dolomite, and montmorillonite (Figure 1b), and the SEM images present an obvious rod-shaped crystal structure (Figure 1a). Acrylic acid (AA), acrylamide (AM), potassium persulfate, N,N-methylenebisacrylamide (MBA), sodium hydroxide (NaOH), and sodium bisulphite were all of analytical grade.

The attapulgite ore was crushed and ground, then passed through a 200-mesh standard sieve. The obtained attapulgite was washed with distilled water and stirred until all attapulgite particles were dispersed. The attapulgite particles were allowed to stand still for 30 min to remove upper impurities of the suspension under the precipitate. The middle attapulgite was heated at 80 °C to evaporate the water totally, then ground to 100 mesh. The 100-mesh attapulgite powder was heated at 400 °C for 1 h to obtain heat-modified attapulgite.

The attapulgite was examined using X-ray diffraction (XRD) (D8 ADVANCE, Zeiss Supra 55VP, Germany) under Cu Kα radiation filtered by Ni. The surface morphology and particle size of the attapulgite were observed via scanning electron microscopy (SEM, Zeiss Supra 55VP, Germany).

2.2. Synthesizing Experiment

A sample of 5 g NaOH was first dissolved in 100 mL distilled water in a beaker. Then, 15 mL of AA and 4 g of AM were slowly added to the NaOH solution to obtain the mixed solution. The initiator–crosslinker solution was prepared with the addition of 0.1 g of
0.05% MBA (acting as the crosslinker), 0.5 g of potassium persulfate, and 0.5 g of sodium bisulfite (molar ratio = 1:1; acting as the reaction initiator) into 10 mL distilled water. The heat-modified attapulgite was added into 300 mL distilled water with 0%, 3%, 5%, 8%, 10%, and 15% mass fractions to obtain six attapulgite colloids; then, the attapulgite colloid and the mixed solution were mixed and reacted for 1 h under four ultrasonic treatments (powers of 200, 300, 400, or 500 W) at 80 °C. The products were dried at 90 °C in an oven and ground to 100 mesh to obtain six sand-fixing materials, denoted as ATP0, ATP3, ATP5, ATP8, ATP10, and ATP15.

2.3. Measurement of Swelling Properties

A dried sample of 0.5 g was mixed with 800 mL distilled water or 0.5% NaCl solution in a beaker. After 5, 10, 20, 30, 40, 50, 60, 70, 80, and 90 min, the material was taken out and filtered through a 100-mesh sieve until no water dripped from it. The water absorption degree (WAD) and saline absorption degree (SAD) were calculated using Equations (1) and (2):

\[
WAD = \frac{(M_2 - M_1)}{M_1} \quad (1)
\]

\[
SAD = \frac{(M_2 - M_1)}{M_1} \quad (2)
\]

where \(M_1\) is the dry mass of the sand-fixing materials before swelling (g), and \(M_2\) is the mass of the sand-fixing materials after swelling (g).

A sample of 0.5 g was weighed and put into a container. The container was placed in a constant-temperature oven at 90°C after saturation. The container was taken out of the oven at a certain time interval and weighed. The water retention value (\(\varphi\)) was calculated using Equation (3):

\[
\varphi = \frac{W_2}{W_1} \times 100\% \quad (3)
\]

where \(W_2\) signifies mass after dehydration (g) and \(W_1\) denotes mass after saturation of water (g). The samples (0.5 g) were also further added to 500 mL deionized water with 0.1 M HCl or NaOH used to adjust the initial pH value (6, 7, 8, 9 or 10) of the resultant solution, followed by shaking for 2 h until the equilibrium of swelling was reached. The influences of pH change on the water absorption were examined.

2.4. Statistical Analysis

The two-way analysis of variance (ANOVA), followed by a multiple comparison test (Duncan’s test), was used to analyze the differences among the treatments. All treatments had six repetitions.

3. Results and Discussion

3.1. Effect of Attapulgite Addition on Water Absorbency

When the attapulgite was used as a comonomer for the synthesis of sand-fixing materials, the ratio of attapulgite to resins was critical to the absorbent performance. The attapulgite addition obviously augmented the water and saline absorbency of the sand-fixing materials. The WAD and SAD of the sand-fixing materials increased with the addition of attapulgite, and reached maxima of 1222.72 g/g and 240.62 g/g at 60 min, respectively, for ATP10. The ATP10 had a higher WAD than other materials. The salinity significantly decreased the water absorbency of the sand-fixing material (Figures 2 and 3). Na⁺ and Cl⁻ might have been complexed on the surface of attapulgite crystal.

This result may have been achieved because the attapulgite was easily dispersed into the AA/AM polymer gel and enhanced its hydrophilicity, and the active -OH groups of attapulgite reacted and crosslinked with the polymeric chains, resulting in the
construction of a three-dimensional hydrophilic network. The nanoparticles of attapulgite reduced the entanglement of the grafted polymeric chains and weakened the hydrogen-bonding interaction among -COOH groups, and then decreased the physical crosslinking degree and facilitated the formation of a polymeric network structure. However, for the highest attapulgite addition (15%), the water absorbency was reduced; this was attributed to the fact that the attapulgite acted as additional crosslinking points in the polymeric network; thus, increasing the amount of attapulgite induced a larger crosslink density, resulting in decreased space in the network for penetration of water molecules. On the other hand, the excess attapulgite might have also filled in the physical space of the network, decreasing the number of hydrophilic groups per unit volume, inducing a decrease of hydrophilicity of the composite, and causing the water absorbency to diminish.

**Figure 2.** Water absorption degree (WAD) of sand-fixing materials synthesized with acrylic acid/acrylamide copolymer and heat-modified attapulgite.

**Figure 3.** Saline absorption rate (SAD) for salt solution (0.9 NaCl) of sand-fixing materials synthesized with acrylic acid/acrylamide copolymer and heat-modified attapulgite.
3.2. Effect of Ultrasonic Power on Water Absorbency

A two-way ANOVA showed that the WAR and SAR of the sand-fixing materials were significantly affected by ultrasonic treatments and the addition of attapulgite, as well as their interaction (Table 1). The WAR was significantly different among five ultrasonic treatments for all six sand-fixing materials to which attapulgite was added. Attapulgite addition significantly accelerated the rise of WAD. The lowest WAD always appeared in ATP0, ATP3, and ATP5 for all ultrasonic treatments, suggesting that the lower dosages of attapulgite had no significant influence on the WAD of the sand-fixing materials. With 200 W of ultrasonic treatment, ATP10 and ATP15 had the largest WAD compared with other materials, whereas the 300 W of ultrasonic treatment significantly improved the WAD of ATP8, ATP10, and ATP15. The WAD of ATP10 reached a maximum value of 1236.14 g/g in distilled water (Table 2).

**Table 1.** Analyses of variance for the effects of attapulgite addition with ultrasonic power, attapulgite addition, and solution pH on the water absorption rate (WAD) and water retention capacity (φ) of sand-fixing materials synthesized with acrylic acid/acrylamide copolymer and heat-modified attapulgite.

<table>
<thead>
<tr>
<th>Traits</th>
<th>Source of Variation</th>
<th>df</th>
<th>F-Value</th>
<th>p</th>
</tr>
</thead>
<tbody>
<tr>
<td>WAD</td>
<td>attapulgite</td>
<td>5</td>
<td>31.73</td>
<td>&lt;0.001</td>
</tr>
<tr>
<td></td>
<td>ultrasonic power</td>
<td>4</td>
<td>9.43</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td></td>
<td>attapulgite × ultrasonic power</td>
<td>20</td>
<td>137.20</td>
<td>&lt;0.001</td>
</tr>
<tr>
<td></td>
<td>attapulgite</td>
<td>5</td>
<td>28.73</td>
<td>&lt;0.001</td>
</tr>
<tr>
<td></td>
<td>pH</td>
<td>4</td>
<td>37.95</td>
<td>&lt;0.001</td>
</tr>
<tr>
<td></td>
<td>attapulgite × pH</td>
<td>20</td>
<td>79.53</td>
<td>&lt;0.001</td>
</tr>
<tr>
<td>φ</td>
<td>attapulgite</td>
<td>5</td>
<td>28.73</td>
<td>&lt;0.001</td>
</tr>
<tr>
<td></td>
<td>ultrasonic power</td>
<td>4</td>
<td>37.81</td>
<td>&lt;0.001</td>
</tr>
<tr>
<td></td>
<td>attapulgite × ultrasonic power</td>
<td>20</td>
<td>79.15</td>
<td>&lt;0.001</td>
</tr>
</tbody>
</table>

**Table 2.** Water absorption rate (WAD) of the sand-fixing materials synthesized with acrylic acid/acrylamide copolymer and attapulgite under different ultrasonic power treatments for 60 min each. Values with the same capital letters were not significantly different among six materials, and those with the same lowercase letters were not significantly different among four ultrasonic power treatments at p < 0.05 or according to Duncan’s multiple comparison tests. * significant difference at 0.05 level, ** at 0.01 level, *** at 0.001 level according to ANOVA.

<table>
<thead>
<tr>
<th>Ultrasonic</th>
<th>ATP0</th>
<th>ATP3</th>
<th>ATP5</th>
<th>ATP8</th>
<th>ATP10</th>
<th>ATP15</th>
<th>F-Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>CK</td>
<td>607.3</td>
<td>746.01</td>
<td>C</td>
<td>880.96</td>
<td>bB</td>
<td>971.24</td>
<td>cB</td>
</tr>
<tr>
<td>200 W</td>
<td>603.16</td>
<td>889.56</td>
<td>aC</td>
<td>830.26</td>
<td>cC</td>
<td>1015.16 bAB</td>
<td>1216.58 bA</td>
</tr>
<tr>
<td>300 W</td>
<td>656.42</td>
<td>881.36</td>
<td>aC</td>
<td>928.36</td>
<td>aB</td>
<td>1057.68 aB</td>
<td>1236.14 aA</td>
</tr>
<tr>
<td>400 W</td>
<td>601.98</td>
<td>890.16</td>
<td>aC</td>
<td>932.4 aB</td>
<td>1014.72 bB</td>
<td>1137.88 aA</td>
<td>1032.98 bB</td>
</tr>
<tr>
<td>500 W</td>
<td>602.84</td>
<td>830.02</td>
<td>bC</td>
<td>885.96</td>
<td>bC</td>
<td>975.38 cB</td>
<td>1057.68 bA</td>
</tr>
<tr>
<td>F-value</td>
<td>2.39</td>
<td>9.38</td>
<td>*</td>
<td>7.37</td>
<td>*</td>
<td>10.23</td>
<td>**</td>
</tr>
</tbody>
</table>

The results suggested that the reaction caused the low water absorbency of superabsorbent composites, and could not been responded to completely. When the ultrasonic power was increased to 300 W, the number of active radicals distinctly increased, which resulted in an increase in grafting percentage and the growth of grafted polymer long chains to form a regular three-dimensional network. Thus, the water absorbency of the superabsorbent composites was increased. However, when the ultrasonic power was more than 300 W, there was no increase in the water absorbency of the superabsorbent composites (Table 2); the enhanced ultrasonic power could cause self-cross-linking between the end groups in the polymer and attapulgite, which in turn caused the enhancement of the cross-linking degree. In addition, the large number of free radicals might have terminated the growth of the grafted chain in the three-dimensional network; this might be an additional
reason for the absorbency loss at higher ultrasonic power. Moreover, the high power could cause liquid splashes easily, which would lead to difficulty in controlling the reaction. According to the result, the ultrasonic power at 300 W was chosen as the optimum ultrasonic power for water.

3.3. Effect of Solution pH on Water Absorbency

The sensitivity of sand-fixing materials to pH value was explored by investigating the WAR for various pH values ranging from 5.0 and 9.0 in water. A two-way ANOVA showed that the water absorbency was significantly different among five pH treatments of water for all six materials (Table 3). Under acidic conditions (pH < 7), most of the carboxylate anions were prorogated, which restricted the electrostatic repulsion and intensified the hydrogen-bonding interaction among the polymer chains, so the network in the composite tended to shrink and, consequently, the water absorbency was decreased. With increasing external pH (5 < pH < 6), we suggested that the ionization level of the graft chains increased and the hydrogen bonding interactions were broken, which induced an increase in the osmotic swelling pressure between the hydrophilic network and the external solution, as well as the electrostatic repulsions among the -COO- functional groups of resin. As a result, the network space of the composite superabsorbent was likely to expand and hold more water for slightly acidic water (pH = 6). These behaviors of the sand-fixing materials would be very advantageous for the use of a superabsorbent in various soils for agricultural application and the stabilization of sand. However, in alkaline water (pH > 8), a charge screening effect of the excess Na cations from the NaOH shielded the -COO- functional groups, inhibiting effective anion–anion repulsion, and thus brought down the water absorbency [23,24].

Table 3. Water absorption degree (WAD) of the sand-fixing materials synthesized with acrylic acid/acrylamide copolymer and heat-modified palygorskite for water at different pH values at 60 min. Values with the same capital letters were not significantly different among six materials, and those with the same lowercase letters were not significantly different among four pH treatments at p < 0.05 or according to Duncan’s multiple comparison tests. * significantly different at 0.05 level, ** at 0.01 level, *** at 0.001 level according to ANOVA.

<table>
<thead>
<tr>
<th>pH</th>
<th>ATP0</th>
<th>ATP3</th>
<th>ATP5</th>
<th>ATP8</th>
<th>ATP10</th>
<th>ATP15</th>
<th>F-Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>315.04 aE</td>
<td>652.08 aD</td>
<td>1040.12 aC</td>
<td>1197.04 aB</td>
<td>1204.84 aA</td>
<td>1192.50 ab</td>
<td>56.32 ***</td>
</tr>
<tr>
<td>6</td>
<td>321.66 aD</td>
<td>656.60 aC</td>
<td>1051.46 aB</td>
<td>1208.66 aA</td>
<td>1213.66 aA</td>
<td>1201.40 aA</td>
<td>136.54 ***</td>
</tr>
<tr>
<td>7</td>
<td>314.40 aD</td>
<td>653.56 aC</td>
<td>1040.58 aB</td>
<td>1197.14 aA</td>
<td>1204.44 aA</td>
<td>1192.18 aA</td>
<td>214.25 ***</td>
</tr>
<tr>
<td>8</td>
<td>328.74 aC</td>
<td>614.12 bB</td>
<td>1012.7 aA</td>
<td>1097.00 aA</td>
<td>1166.62 abA</td>
<td>1170.08 aA</td>
<td>284.23 ***</td>
</tr>
<tr>
<td>9</td>
<td>279.38 bD</td>
<td>491.06 cC</td>
<td>857.14 bB</td>
<td>932.1 bAB</td>
<td>976.4 bA</td>
<td>983.14 bA</td>
<td>68.34 ***</td>
</tr>
<tr>
<td>F-value</td>
<td>8.36 *</td>
<td>12.37 **</td>
<td>23.47 **</td>
<td>10.43 *</td>
<td>9.34 *</td>
<td>6.85 *</td>
<td></td>
</tr>
</tbody>
</table>

3.4. Effect of Attapulgite Addition and Ultrasonic Power on Water Retention

The WRC was an important indicator for the sand-fixing materials in view of practical applications. In this study, sand-fixing materials prepared with various dosages of attapulgite were used to assess the water retention capacity under various ultrasonic treatments in order to evaluate the influences of attapulgite on the water retention of the sand-fixing materials (Table 4). The results showed that the ATP8 and ATP10, under 200 W and 300 W, of ultrasonic treatment showed outstanding water retention capacity compared to the other materials. The WRC of ATP10 was significantly higher than that of the other materials, and no (ATP0) and lower addition of attapulgite (ATP3) always reduced the WRC. Moreover, the swollen water in sand-fixing materials could be classified into bound water, half-bound water, and free water; compared to the bound water and half-bound water, the free water had a high transferability and could be easily lost under ultrasonic treatments [15,25]. The numbers of hydrophilic function groups, such as -COOH and -COO-, were assumed to be higher in the composites with attapulgite; this could result in a higher percentage of bound water and half-bound water in the sand-fixing materials.
compared to free water. In this way, the incorporation of a certain amount of attapulgite in the polymeric network could significantly improve the water retention capacity.

Table 4. Water retention capacity ($\phi$) of sand-fixing materials synthesized with acrylic acid/acylamide copolymer and heat-modified attapulgite under different ultrasonic treatments for 5 h. Values with the same capital letters were not significantly different among six materials, and those with the same lowercase letters were not significantly different among four ultrasonic power treatments at $p < 0.05$ or according to Duncan’s multiple comparison tests. * significant difference at 0.05 level, ** at 0.01 level, *** at 0.001 level according to ANOVA.

<table>
<thead>
<tr>
<th>Ultrasonic</th>
<th>ATP0</th>
<th>ATP3</th>
<th>ATP5</th>
<th>ATP8</th>
<th>ATP10</th>
<th>ATP15</th>
<th>F-value</th>
</tr>
</thead>
<tbody>
<tr>
<td>CK</td>
<td>8.63 C</td>
<td>9.34 cC</td>
<td>15.34 bA</td>
<td>11.24 cB</td>
<td>12.34 cB</td>
<td>16.57 bA</td>
<td>4.65 *</td>
</tr>
<tr>
<td>200 W</td>
<td>11.43 C</td>
<td>22.16 aB</td>
<td>23.49 aB</td>
<td>30.21 aA</td>
<td>32.65 aA</td>
<td>27.46 aA</td>
<td>27.45 ***</td>
</tr>
<tr>
<td>300 W</td>
<td>12.46 C</td>
<td>24.71 aB</td>
<td>25.78 aB</td>
<td>26.34 bB</td>
<td>31.86 aA</td>
<td>26.54 aB</td>
<td>8.28 *</td>
</tr>
<tr>
<td>400 W</td>
<td>9.64 C</td>
<td>16.24 bB</td>
<td>15.24 bB</td>
<td>16.32 cB</td>
<td>22.16 bA</td>
<td>15.21 bB</td>
<td>24.71 ***</td>
</tr>
<tr>
<td>500 W</td>
<td>8.34 C</td>
<td>12.34 cB</td>
<td>15.37 bB</td>
<td>13.24 cB</td>
<td>21.46 bA</td>
<td>11.34 cBC</td>
<td>14.87 **</td>
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<td>29.68 **</td>
<td>16.49 **</td>
<td>18.64 ***</td>
<td>14.37 **</td>
<td>9.34 **</td>
<td></td>
</tr>
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</table>

4. Conclusions

The attapulgite significantly improved the water absorbency and retention capacity of the sand-fixing material for both distilled water and saline solution, and 10% addition always achieved the optimal efficiency. A spatial network structure in the sand-fixing materials was formed with attapulgite and other materials. Ions released from attapulgite into the network were mutually exclusive, made the materials more extensible, and gave them more porosity. The water absorption capacity of the materials was the affinity of the hydrophilic groups with water [26–28]. The attapulgite was exposed to more adsorbent centers and reactive groups. High water-absorbing resins could absorb more water because their hydrogen bond groups were easy to combine with functional groups [29–31]; these composites combined with attapulgite and resins had greater water absorbency and retention capacity.

The pH value of water significantly influenced the water absorbency of the sand-fixing materials with attapulgite added. The sand-fixing materials always possessed greater water absorbency for slightly acidic water and absorbed less alkaline water. Acidic ions could replace more ions from attapulgite, resulting in more ions being released from attapulgite into the composite to increase the water absorbency and retention capacity of the sand-fixing materials. More alkaline ions may have blocked the pore canals in composites combined with attapulgite and resins due to the glue combination effect.

The water absorbency and retention of the sand-fixing materials manifested an increasing trend with the increase in ultrasonic power. However, in the range of 300–500 W, the water absorption capacity of all sand-fixing materials gradually decreased. The higher ultrasonic power caused liquid splashes. Thus, the ultrasonic power at 300 W was chosen as the optimum treatment.

The preparation method of efficient sand-fixing materials always needs further optimization, and the control of environmental conditions during the preparation process is a key factor affecting the absorption. The results of this study provide a possible basis for the use of attapulgite clay in desert control, and also provide new materials for the large-scale application of efficient sand-fixing materials for desertification control.

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Data Availability Statement: The authors confirm that the data supporting the findings of this study are available within the article.

Conflicts of Interest: Authors Hanru Ren and Ling Tao were employed by the company Gansu Hanxing Environmental Protection Co., Ltd. The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

References


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