The Swelling Behaviors and Network Parameters of Cationic Starch-*g*-Acrylic Acid/Poly(dimethyldiallylammonium chloride) Semi-Interpenetrating Polymer Networks Hydrogels

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ABSTRACT: Amphiphilic semi-interpenetrating polymer networks (semi-IPN) hydrogels were prepared by a sequential-IPN method by acrylic acid graft copolymerization into cationic starch in mild aqueous media of poly(dimethyldiallylammonium chloride). Some main factors were investigated to evaluate the swelling of hydrogels, and the network parameters M_c were given accordingly to elaborate the interaction between polymers. The chemical structure of the resulting hydrogel was confirmed using Fourier transform infrared spectroscopy. The cationic starch-based semi-IPN hydrogels achieved a high swelling capacity of 1070 g/g in deionized water and 94 g/g in 0.9 wt % NaCl solution, respectively) and high compressive stress in a high water content. Besides, a different pH-dependent behavior was found for this semi-IPN hydrogel. © 2008 Wiley Periodicals, Inc. J Appl Polym Sci 110: 1828–1836, 2008

Key words: amphiphilic; hydrogels; interpenetrating networks (IPN); mechanical properties; starch

INTRODUCTION

Hydrogels are three-dimensional physically and/or chemically bonded polymeric matrices that absorb and swell with a significant proportion of water without undergoing dissolution.[1] They undergo a volume phase change in response to a change in external stimuli, such as pH, ionic strength, temperature, electric field, solvent, and pressure.[2–5] All these properties of the hydrogels have aroused great interest and found widespread applications in adult incontinence pads, sanitary products, and pharmaceutical and medical applications[6,7] in addition to their use in the medical device industry as artificial muscles,[8,9] agriculture,[10] and so on.

Starch is the most abundant natural polymer and has been investigated actively to synthesize the hydrogel because of its advantages of renewable, biodegradable, and toxicologically harmless.[11,12] However, its high brittleness and insolubility in water limited the application.[13] In our previous study, cationic starch (CS)-based hydrogels were successfully prepared by graft copolymerization of acrylic acid (AA) and showed a good swelling reversibility in saline solution.[14] However, the swelling capacity of the hydrogels remained unsatisfactory.

Besides swelling capacity, mechanical strength is also a factor to decide the application of hydrogels. Poor mechanical properties of hydrogel are hardly to sustain their functions for a long time and are easily destroyed when even small force is applied.

The interpenetrating polymer networks (IPN) structure endows the hydrogels better mechanical properties.[15–17] IPN is defined as a physical mixture of two or more crosslinked networks synthesized or crosslinked within each other with no significant degree of covalent bonds between them. IPN could help improve the mechanical strength and resiliency of the polymer due to physical entanglements and network interactions when compared to individual crosslinking networks.[18–20] The semi-IPNs hydrogels with one linear polymer in the IPN have been the subject of many investigations.[21,22] Kim and Park[23] have synthesized the hydrogels based on poly (acrylic acid-*co*-acrylamide)/polyethylenimine which shows good mechanical strength of 29 kPa enhanced by their semi-IPN structure and charge attraction between cationic PEI and anionic PAA, but the absorbency of hydrogels does not exceed 100 g/g.

The semi-IPNs composed of polyacrylamide having entangled PNIPAAm which also have good mechanical properties but is low in water content.[24] It has been suggested that the hardness of the gels could be improved because of their amphiphilic characteristic.[25–27] However, the hydrogels usually exhibit enhanced mechanical strength at the sacrifice of swelling capacity.

In this article, a new amphiphilic semi-IPN hydrogel was developed by introducing cationic poly (dimethyldiallylammonium chloride) (PDMDAAC) into CS-g-AA polymer networks by a sequential IPN method, anticipating an improved hydrogel swelling and mechanical strength. In addition, the interaction between cationic groups and anionic groups was expected to improve the compatibility of the components of a polymer mixture. Network parameters were measured under various synthetic conditions in an attempt to give a detailed explanation on such swelling behaviors.

EXPERIMENTAL

Materials

Corn starch (Hutubi, Xingjiang), cationic starch (CS) with a degree of substitution (DS) of (DS = 0.11, 0.20, 0.37), was prepared by a hemi-dry process according to our previous work[28]; acrylic acid (AA) (Tianjin Guangfu chemical reagent factory, Tianjin, China), DMDAAC (60 wt % aqueous solution, Shandong Luyue Chemical, Shandong, China), N, N'-methylene bisacrylamide (MBAM, Shanghai Chemical Co., Shanghai, China), ammonium persulfate (APS, Xi'an Chemical Co., Xi'an, China). All chemicals were of analytical grade and used without further purification.

Preparation of PDMDAAC

One hundred gram 60 wt % dimethyldiallylammonium chloride (DMDAAC) aqueous solution was added to a 250-mL three-neck flask, followed by 0.3 g APS as an initiator. The mixture was stirred 2 h at 60°C. The resulting polymer was poured into acetone and absolute ethanol in turn to form precipitation and wash away monomer respectively. The precipitation was dried for 24 h at 60°C.

Preparation of amphiphilic superabsorbents with semi-IPN

Amphiphilic superabsorbents with semi-interpenetrating polymer networks (semi-IPN) were prepared by free-radical crosslinking copolymerization at nitrogen gas condition. Typically, 1 g CS was added to 10 g deionized water in 250-mL three-necked flask and stirred for 20 min for a full dissolution at room temperature. Then 15 g AA, neutralized by 40 wt % NaOH solution to 75 mol % neutralization degree in ice bath before was added, followed by 2 g PDMDAAC under stirring for another 30 min to get a clear solution. MBAM (0.035 wt % based on the weight of AA) and APS (2.5 wt % based on the weight of AA) aqueous solutions were added to the mixture solution, respectively. After 15 min, the water bath was heated slowly to 55°C and kept for 3 h. Then the resulting product was immersed in excess deionized water overnight to remove the unreacted monomers in the gels, then dehydrated with absolute ethanol and dried at 80°C to a constant weight. The dried product was milled to 20–40 mesh.

Measurement of water swelling capacity

The swelling capacity of the hydrogels was assessed by filtration method. A powdered semi-IPN hydrogel sample (0.1 g) was immersed in 250 mL of deionized water or 100 mL of 0.9 wt % NaCl in a breaker at room temperature for 12 h to reach a swelling equilibrium. The swollen sample was then separated from swelling solutions by filtering through a 100-mesh screen. The swelling capacity of the absorbent hydrogel, Q_{H_2O} (g/g) was calculated using the eq. (1):

$$Q_{H_2O} = \frac{m_2 - m_1}{m_1} \quad (1)$$

m_1 and m_2 are the weights (g) of the dry sample and the swollen sample, respectively.

Fourier transform infrared spectroscopy

The dried samples were powdered and mixed with KBr to make pellets. The IR spectra were taken using a Fourier transform infrared (FTIR) spectrophotometer (BRUKER EQUATIONINOX55).

Network parameters M_c

The swelling behavior of the polymer network was mainly depends on the average molar mass between the crosslinks, M_c, which is reversely proportional to crosslink density. According to Flory-Huggins theory, M_c can be given as follows when the hydrogel with a highly crosslinking density and a swelling more than 10 g/g[29]:

$$Q^{5/3} = M_c(0.5 - \chi_1)/D_2 V_1 \quad (2)$$

where Q is the swelling capacity of the polymer; D_2 is the density of the polymer; V_1 is the molar volume of the solvent used for swelling studies, and the χ_1 is the Flory-Huggins interaction parameter between solvent and polymer.

Considering it is difficult to determined χ_1 by experiments, Li et al. think when methanol and deionized water are mixed with a volume fractions of methanol C, there lies a linear relationship between χ_1 and C. So χ_1 can be determined as[30]:

$$\chi_1 = K_1C + K_2 \qquad (3)$$

Thus, if K_2 is much less than 0.5, we obtain

$$D_2V_1Q_{\mathrm{CH_3OH-H_2O}}^{5/3} = M_c(0.5 - K_1C) \qquad (4)$$

Then $D_2V_1Q_{\mathrm{CH_3OH-H_2O}}^{5/3}$ is replaced by Y, eq. (4) can be simplified as

$$Y = M_c(0.5 - K_1C) \qquad (5)$$

The above equation indicates that there is a liner relationship between Y and C. So M_C can be calculated according to the slope of resulting line by plotting Y to C.

Determination of nitrogen content of hydrogels

Nitrogen content of hydrogel is determined by Kjeldahl method using a KDY-9820 Kjeldahl instrument (Beijing, China). The samples were analyzed three times with deionized water as blank. The nitrogen content was calculated by the following eq. (6):

$$N(\%) = 1.401 \times C \times (V - V_0)/W \qquad (6)$$

where C was the concentration of the hydrochloric acid (mol/L); W was the weight of the hydrogel (g), V_0, V were the volume of HCl consumed by the blank sample and the sample, respectively, (mL).

Mechanical strength

The hydrogel compressive stress was performed on columnar samples of the same size (20 mm diameter × 10 mm length) in homemade equipment according to the reference.[31] Fifteen measurements were performed at constant temperature (25°C), and the average value was taken as the ultimate compressive strength.

RESULTS AND DISCUSSION

The proposed method used to produce semi-IPN hydrogel is shown in Scheme 1.

Fourier transform infrared spectra

Fourier transform infrared (FTIR) spectra were carried out to characterize the chemical structure of semi-IPN hydrogels [Fig. 1(a)]. The peaks at 3472.8

cm^{-1} could be assigned to O—H stretching vibrations of semi-IPN hydrogel, and an absorption band at 1708.5 cm^{-1} was associated with stretching vibrations of —COOH of AA, whereas the adsorption was found at 1728.8 cm^{-1} for CS-g-AA [Fig. 1(c)], suggesting a different environment of —COOH groups. When PDMDAAC was entrapped into CS-g-AA network to fabricate semi-IPN structure, the interaction between —COOH and cationic ammonium groups of polymer chains is not ignorable. Ammonium groups partly distracted the charge density of —C=O in —COOH groups, resulting in a low-wavenumber shift. The peaks of —COO$^-$ asymmetric and symmetric stretching were found at 1561.4 cm^{-1} and 1404.4 cm^{-1}. Besides, the characteristic peak of starch [Fig. 1(b)] at 927 cm^{-1} became smaller and even disappeared compared with semi-IPN hydrogel and CS-g-AA, which indicated that AA was graft copolymerized onto CS.[32] It was difficult to recognize the existence of PDMDAAC because of its low content and unobvious characteristic peaks in this case. However, N analysis confirmed the existence of PDMDAAC in the polymer networks since the experimental nitrogen content of 0.6534 wt % the hydrogel was close to the theoretical values 0.6879 wt %.

Effect of DS of the cationic starch on swelling capacity

Cationic modification of starch endowed starch with the soluble ability in cold water when DS was more than 0.1. Moreover, the introduction of cationic hydrophilic groups might contributes to the salt tolerance of the resulting hydrogels.[33] However, it was found a high DS did not always increase the swelling capacity in our experiment. Table I showed that the swelling capacity increased to 1070.5 g/g and 94.0 g/g in deionized water and 0.9 wt % NaCl solution, respectively, under a DS of 0.11, compared with original starch based hydrogel with the swelling capacity of 723.6 g/g and 63.5 g/g accordingly. However, further increased DS resulted in the decline on the swelling capacity. This phenomenon can be explained as that more cationic ammonium groups provide more chance to interact with carboxylic groups under a higher DS. As a result, the network shrunk and swelling capacity became worse. This further confirmed the existence of salt bonds between carboxyl groups and ammonium groups.

Effect of crosslinker dose on swelling capacity and M_c

In this case, MBAM was used as a crosslinker because of its high reactivity and benign hydrophility. The crosslinker dose is one of the most
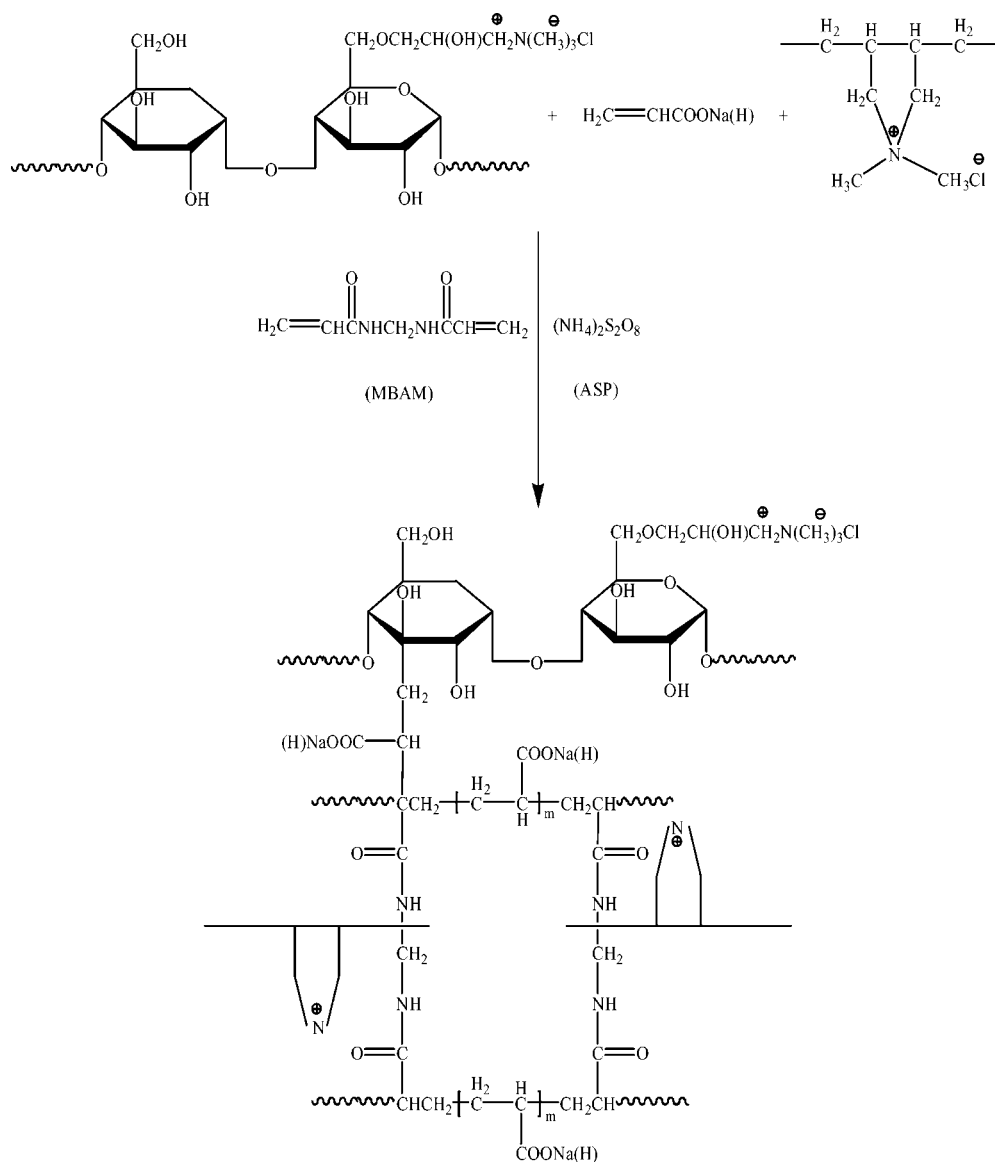
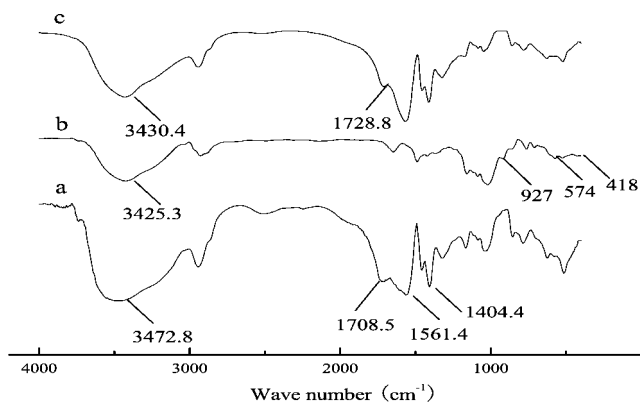**Scheme 1** Scheme of the method used to produce semi-IPN hydrogel.

Figure 1 FTIR spectra of CS, CS-g-AA, and the semi-IPN hydrogel (a) semi-IPN hydrogel; (b) CS; (c) CS-g-AA.

TABLE I
Effect of Degree of Substitution on Swelling Capacity

Cationic starch Degree of substitution	Swelling capacity (g/g)	
	In Deionized Water	In 0.9 wt % NaCl Solution
0	723.6	63.5
0.11	1070.5	94.0
0.20	786.7	86.7
0.37	626.5	67.1

Crosslinker = 0.035 wt %; Initiator = 2.5 wt %; T = 55°C; Mass ratio of CS : AA : PDMDAAC = 1 : 15 : 2.
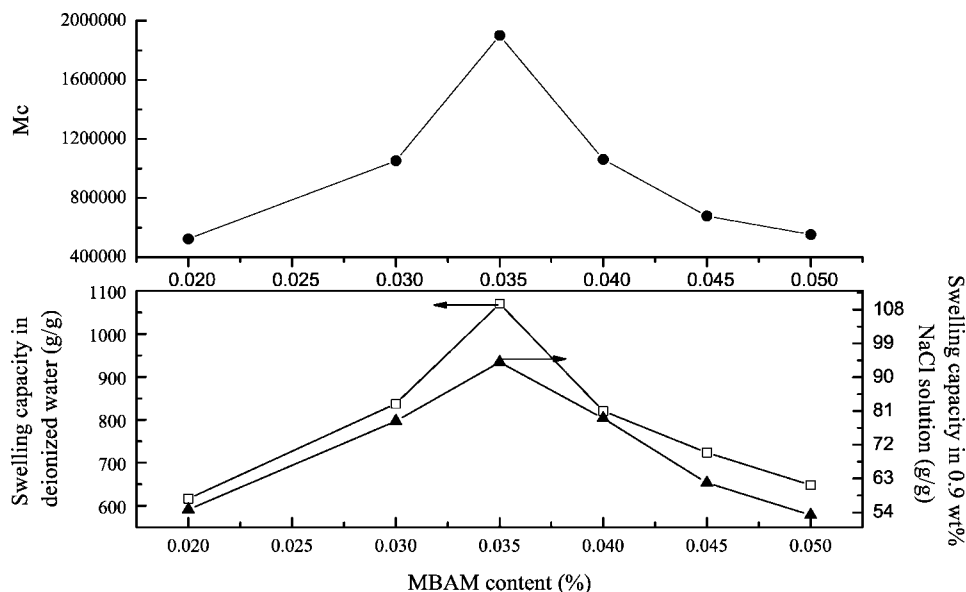
Figure 2 Effect of MBAM dose on swelling capacity and Mc of CS-*g*-AA/PDMDAAC semi-IPN hydrogel: cationic starch/AA/PDMDAAC (w/w/w) is 1 : 15 : 2; initiator is 2.5 wt %; the neutralization of AA is 75 mol %. □ in deionized water; ▲ in 0.9 wt % NaCl solution.

important factors to affect the swelling behavior of hydrogels. The relationship between the crosslinker dose and the swelling capacity was shown in Figure 2(bottom). The swelling capacity reached the maximum when the crosslinker dose was 0.035 wt %, then decreased greatly when the crosslinker dose exceeded the optimum value. A main reason for the observed decrease was that increasing crosslinker dose resulted in a rise of crosslinking density within the hydrogel. This can be confirmed by the decreased M_c in a range of crosslinker dose from 0.035 wt % to 0.050 wt % [Fig. 2(top)]. A decrease of M_c of the polymeric chain between crosslinking points obviously reduced the free volumes accessible to the water molecules, therefore, it was difficult for hydrogels to expand, and the swelling capacity was lower than the optimum.

Different to the trend above, the swelling capacity of the hydrogel was enhanced while M_c increased with increasing of MBAM amount from 0.02 wt % to 0.035 wt %. The abnormal phenomenon can be explained as follows: the chemical crosslinking usually leads to uneven crosslinking, which would cause partly high crosslinking density in some domains whereas the low crosslinking density in other domains. As a result, more sols were deduced in lightly crosslinked domains. On the other side, highly crosslinker domains lead to a low swelling. So it can be concluded that the effective crosslinking density was not always proportional to the crosslinking dose.

In our investigation, the crosslinker dose was less than that of other semi-IPN hydrogels composed of

poly(acrylamide-*co*-styrene) and poly(vinyl alcohol).[34] It can be interpreted as carboxyl groups and quaternary ammonium groups in the amphiphilic hydrogele was inclined to engender electrostatic attraction, resulting in an increase of crosslinking density. This phenomenon was also found in other amphiphilic hydrogels by our group.[35–37]

Effect of AA dose on swelling capacity and M_c

The swelling capacity and M_c were plotted versus the AA dose varying from 5 g to 20 g, as shown in Figure 3. The polymerization was performed under the same amount of PDMDAAC varying the dose of AA. As a result, the concentration of PDMDAAC was higher under a low AA dose and inclined to interact with negatively charged AA. This can be confirmed by the N content in the resulting hydrogel (Table II). So the crosslink density was high (accordingly, M_c was 1.7 times lower than the one in a dose of 15 g AA), which finally caused a decrease in the free space available between crosslinks, thus providing less space for accommodation of water molecules in the network. Besides, the swelling capacity strongly depended on the number of hydrophilic groups, so increased carboxylate ions (COO^-) can increase the swelling capacity due to more operating electrostatic repulsions between chains, resulting in loosening of the cage structure causing decline of crosslink density, thus the swelling capacity increased. Similar swelling results were found in earlier reports.[37] However, when the dose of the AA increased further from 15 to 20 g, the swelling
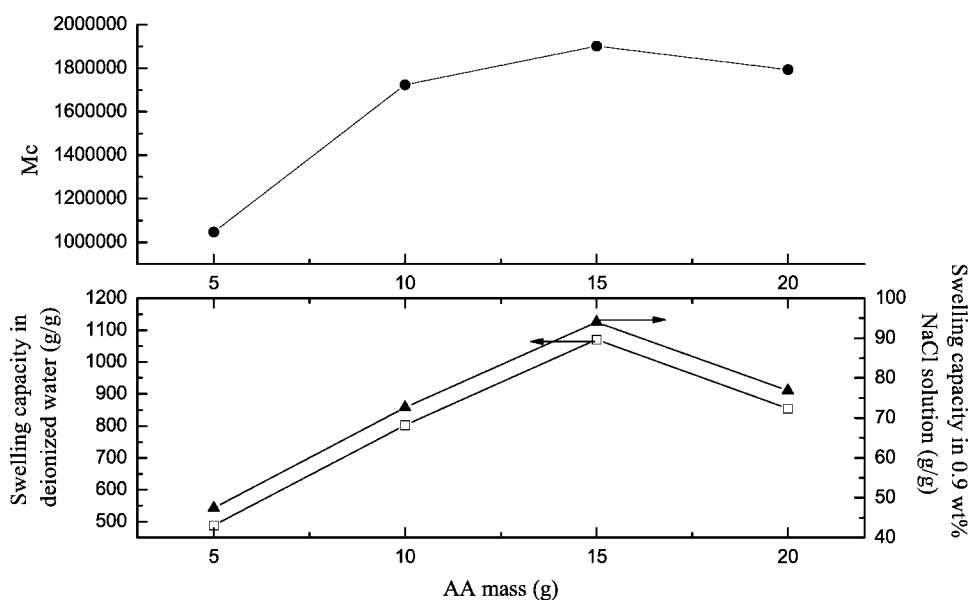
Figure 3 Effect of AA mass on swelling capacity and M_c of CS-*g*-AA/PDMDAAC semi-IPN hydrogel: cationic starch/ PDMDAAC (w/w) is 1 : 2; initiator is 2.5 wt %; crosslink content is 0.5 wt %; the neutralization of AA is 75 mol %. □ in deionized water; ▲ in 0.9 wt % NaCl solution.

decreased markedly. Excessive AA led to popcorn polymerization,[38] which produced short polymer chains and smaller molecular weight distribution. As a result, M_c decreased instead due to increased possibility for the electrostatic interaction between anionic and cationic groups.

The swelling capacity of semi-IPN hydrogels in this experiment was directly proportional to hydrophilic groups under certain dose of AA, which was similar in the case of graft copolymer hydrogels.[14] In comparison, semi-IPN hydrogel exhibited a extraordinary swelling capacity of 1070.5 g/g in the deionized water under a dose of AA of 15 g whereas the graft-copolymer hydrogel showed a maximum swelling of 460 g/g with a AA dose of 30 g. This difference indicated that the dosage of the AA and the hydrogel structure played a great role in the swelling. A large number of polymerization heat pro-

duced by excessive AA lead to smaller molecular weight and the low swelling capacity. Besides, polymerization in the presence of PDMDAAC also made a difference on the swelling.

Effect of PDMDAAC dose on swelling capacity and M_c

PDMDAAC as a linear polymer was used to fabricate the semi-IPN hydrogel by entrapping into CS-*g*-AA network. FTIR gave an indirect proof for the existence of PDMDAAC as well as the interaction between PDMDAAC and CS-*g*-AA network. It was also evident from Table III that the content of nitrogen in semi-IPN hydrogel was approaching the theoretical values with varying the dose of PDMDAAC. Besides, PDMDAAC dose dependence of the swelling behavior was shown in Figure 4. The swelling

<table>
<tr><td colspan="3" align="center">**TABLE II**
Effect of AA Content on the Content of Nitrogen</td></tr>
<tr><td></td><td colspan="2" align="center">Nitrogen Content (wt %)</td></tr>
<tr><td>Dose (g)
AA</td><td>Theoretical
Values</td><td>Experimental
Values</td></tr>
<tr><td>5</td><td>1.5479</td><td>1.4135</td></tr>
<tr><td>10</td><td>0.9526</td><td>0.8095</td></tr>
<tr><td>15</td><td>0.6879</td><td>0.6534</td></tr>
<tr><td>20</td><td>0.4630</td><td>0.4621</td></tr>
</table>

Crosslinker = 0.035 wt %; Initiator = 2.5 wt %; T = 55°C; Mass ratio of CS : PDMDAAC = 1 : 2.

<table>
<tr><td colspan="3" align="center">**TABLE III**
Effect of PDMDAAC Content on the Content of
Nitrogen and Swelling Capacity</td></tr>
<tr><td></td><td colspan="2" align="center">Nitrogen Content (wt %)</td></tr>
<tr><td>Dose (g)
PDMDAAC</td><td>Theoretical
Values</td><td>Experimental
Values</td></tr>
<tr><td>0.5</td><td>0.1876</td><td>0.1759</td></tr>
<tr><td>1</td><td>0.3642</td><td>0.3526</td></tr>
<tr><td>2</td><td>0.6876</td><td>0.6534</td></tr>
<tr><td>3</td><td>0.9777</td><td>0.8055</td></tr>
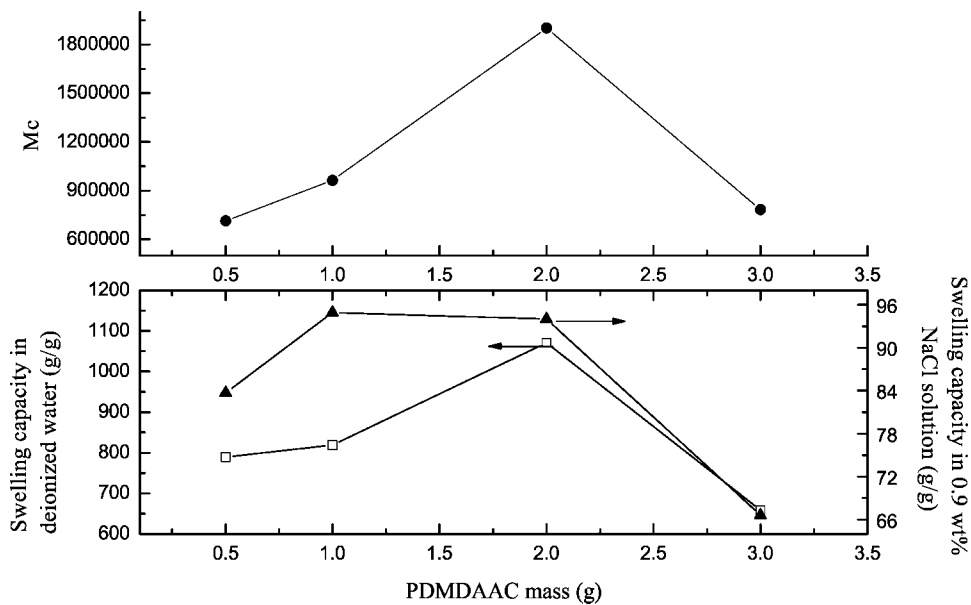</table>

Crosslinker = 0.035 wt %; Initiator = 2.5 wt %; T = 55°C; Mass ratio of CS : AA = 1 : 15.

Figure 4 Effect of PDMDAAC mass on swelling capacity and Mc of CS-g-AA/PDMDAAC semi-IPN hydrogel: cationic starch/AA (w/w) is 1 : 15; initiator is 2.5 wt %; crosslink content is 0.5 wt %; the neutralization of AA is 75 mol %. □ in deionized water; ▲ in 0.9 wt % NaCl solution.

increased as the dose of the PDMDAAC increased from 0.5 g to 2 g, then decreased beyond 2 g. This phenomenon can be ascribed to the functions of PDMDAAC in the semi-IPN hydrogel: the hydrophilicity and electrostatic attraction. The former would make a contribution to the swelling capacity, whereas the latter would reduce the swelling by increasing the crosslinking density. However, it was found, under a low dose of PDMDAAC, M_c increased instead with the increasing of PDMDAAC (Fig. 4). This behavior was probably because of the entanglement of polymer chains, which produced the shielding of charges. When PDMDAAC was added even more, a higher crosslinking density would cause the network of hydrogel denser due to electrostatic attraction inevitably.
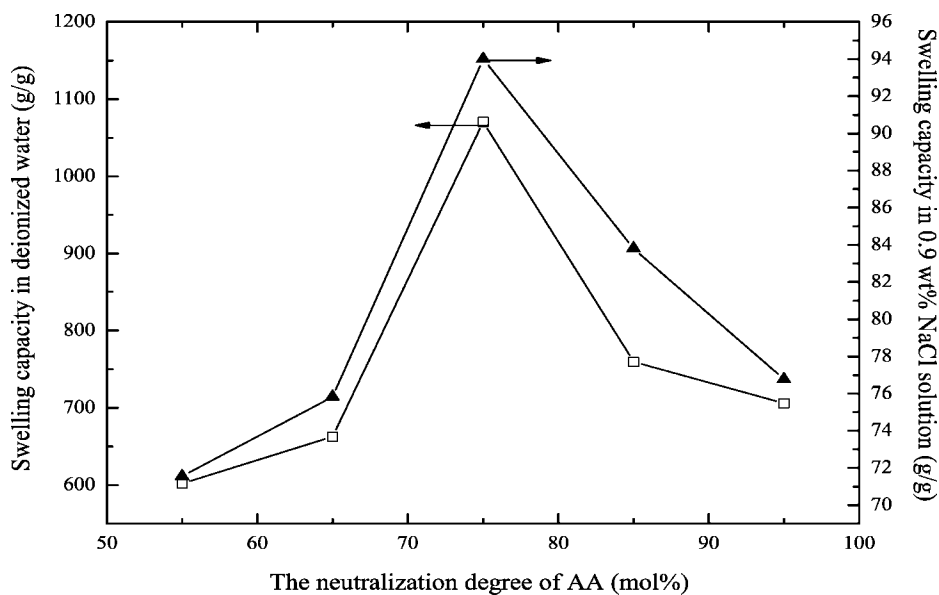

Figure 5 Effect of neutralization of AA on swelling capacity of CS-g-AA/PDMDAAC semi-IPN hydrogel: cationic starch/AA/PDMDAAC (w/w/w) is 1 : 15 : 2; initiator is 2.5 wt %; crosslink content is 0.5 wt %. □ in deionized water; ▲ in 0.9 wt % NaCl solution.
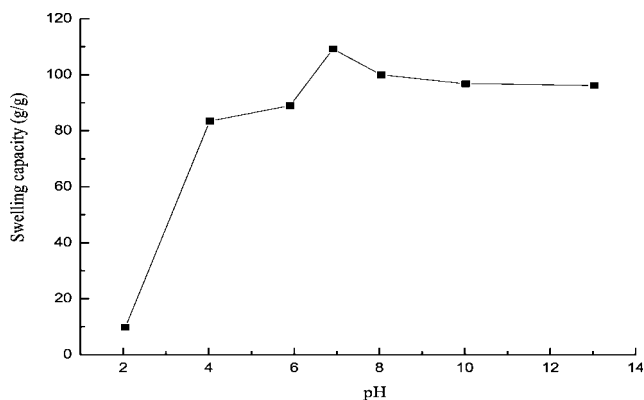
Figure 6 Effect of the different pH value on swelling capacity of CS-*g*-AA/PDMDAAC semi-IPN hydrogel.

Effect of the neutralization degree of AA

As shown in Figure 5, the absorbent ability increased as increasing the neutralization degree of AA from 55 mol % to 75 mol % and decreased with further increasing the neutralization degree of AA. The variation of water absorbency with increasing AA can be explained as follows: when neutralization degree of AA was low, both the polymerization rate and exothermic rate would be quicker, and the temperature of reaction system would be higher, which would lead to higher crosslinking degree,[39] that made the absorbency of hydrogels low, and the other reason was that the carboxylate anions were protonated under low neutralization degree of AA and would form hydrogen bond, reducing the electrostatic repulsion forces and consequently decreasing the water absorbency; with the increase of neutralization degree, the electrostatic repulsion in carboxylate anions of the hydrogel networks would increase, and the stretching degree of the hydrogel network would also increase,[40] which would lead to the increase of hydrophilicity of hydrogel. However, a further increase in neutralization of the carboxylic acid groups could reduce the swelling capacity. This is due to the charge screening effect of excess Na$^+$ ions in the swelling media and negative interactions of carboxylate ions.[29] The excess cations shield the carboxylate groups and prevent effective electrostatic

repulsion. Besides, the unprotonated carboxylic groups were inclined to form salt bond with ammonium groups on PDMDAAC, which could make the crosslinking density higher, so the swelling capacity was low.[41]

Effect of pH on swelling capacity

CS-*g*-AA/PDMDAAC is an amphiphilic polyelectrolyte hydrogel with carboxylic and ammonium groups in polymer chains. The protonation degree is closely dependent of the pH value of the medium, which results in pH-sensitivity of the product. To investigate the influence of pH on the swelling capacity of the hydrogel, the ionic strength of buffer solution with varying pH values is kept constant ($I = 0.15M$) by adding NaCl. The samples were placed in buffer solutions of desired pH (2–13) at room temperature, allowed to swell to equilibrium and then weighed. The swelling trend of hydrogel was shown in Figure 6. It was found that the swelling capacity was increased with increasing the pH value, but decreased a little when the pH value exceeded 6.91. Besides, the swelling degree of amphiphilic semi-IPN hydrogel in the basic medium was higher than in the acid medium. These results clearly indicated that, under low pH conditions (pH < 4), the carboxyl groups in the hydrogel associated and the electrostatic repulsion was reduced, thereby the contracted hydrogel network caused low swelling capacity. However, the swelling capacity in this experiment was still higher than the Li et al.'s work[42] reported in the same pH. In that case, semi-IPN hydrogel composed of guar gum/poly(acrylic acid) was synthesized. This was due to the fact that the quaternary ammonium salt played the hydrophilic role in the acid solution. As the pH of the solution increased, the swelling ratio was sharply increased, there were two reasons: one was the deprotonation of carboxylic groups in the hydrogels developed an internal ion osmotic pressure and induced the hydrogel to swell; and the other was that when the pH value of the solution increased, the ionization of the carboxylic groups in the hydrogel also increased correspondingly, which increased

TABLE IV
The Mechanical Strength of Three Hydrogels

Hydrogels	Swelling Capacity		Mechanical Strength	
	In Deionized Water (g/g)	In 0.9 wt % NaCl (g/g)	Compressive Stress (kg/m^2)	Water Content (%)
Semi-IPN	1071	90	1398	99.56
CS-*g*-AA-DMDAAC	873	84	986	99.41
Starch-*g*-AA	529	73	354	98.81

electrostatic repulsions among the macromolecular chains. This condition enlarged the free volume in the matrix which rapidly increased the swelling ratio subsequently. This phenomenon was similar to the other semi-IPN hydrogel.[42] When the pH value of the buffer solution further increased, the swelling capacity decreased but not in a sharp style, but this phenomenon was usually found in other hydrogels.[43] This is attributed to the charge screening to excessive Na^+ or Cl^- ions because of the addition of cationic PDMDAAC.

Mechanical strength of the semi-IPN hydrogel

All mechanical tests were carried out on swollen hydrogels at room temperature. Table IV showed ultimate compressive stress data of the CS-g-AA/PDMDAAC amphiphilic semi-IPN hydrogels with CS-g-AA-PDMDAAC and starch-AA as controls. Compared with grafted polymers, semi-IPNs hydrogels showed a higher resistance to compression (1398 kg/m^2) even under higher water content of 99.56%. This suggests that the mechanical performances are more related to the structure of the hydrogels. The highest mechanical strength of the semi-IPN is due to its chains distribution and molecular noncovalent interaction which resulted from the entanglement of linear PDMDAAC and the network of CS-g-AA.

CONCLUSIONS

Amphiphilic CS-g-AA/PDMDAAC semi-IPN hydrogels were successfully prepared by AA graft polymerization onto CS in the presence of PDMDAAC. The swelling capacity is substantially affected by the crosslinking density which varied with different dose of crosslinker, AA and PDMDAAC. And the crosslinking density in lower crosslinker dose showed strange phenomenon, which can be explained by uneven chemical crosslinking. The salt linkage produced by AA and PDMDAAC also played an important role in varying crosslinking density. The amphiphilic hydrogels showed especial characteristics in different pH solutions and exhibited relatively high absorbency in a wide pH range of solution. They also represented high-compressive strength because of the special amphiphilic semi-IPN structure. The relatively high water absorbent ability and compressive strength of CS-g-AA/PDMDAAC semi-IPN hydrogels showed a promise in applications for moisture and manure maintenance materials in agriculture/horticulture field.

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