Synthesis and Antimicrobial Activity of Some Cross-linked Copolymers with Alkyl Chains of Various Lengths

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ABSTRACT: Amphoteric polymer hydrogels were prepared by the copolymerization of three kinds of *N*,*N*′-dimethyl-*N*-alkylmethacryloxylethyl ammoniumbromide (DMAEA) with different lengths of alkyl chains (DMAEA-RB) (*R*-ehtyl/hexyl/dodecyl), acrylic acid (AA), and acrylamide (AM). The water content of the AA-AM-(DMAEA-RB) terpolymers decreased with the increasing length of alkyl chains in quaternary ammonium group in the terpolymers and increased with the increasing molar ratio of DMAEA-RB to 2 mol % and then decreased. Their antibacterial activities against *Escherichia coli* and *Staphylococcus hyicus* were investigated by a colony count method. It was found that the copolymer exhibited higher antibacterial activity with increasing chain length of alkyl groups in ammonium groups. For P[AA-AM-(DMAEA-DB)], the DMAEA-DB content is higher and contact time is longer, its antibacterial activity is better. However, when the contact time and quaternary ammonium content were above 30 min and 2%, respectively, the amount of live cells $N(t)$ in a cell suspension increased in the presence of P[AA-AM-(DMAEA-EB)] or P[AA-AM-(DMAEA-HB)]. © 2010 Wiley Periodicals, Inc. J Appl Polym Sci 120: 1767–1773, 2011

Key words: superabsorbent; hydrogels; antibacterial activity; quaternary ammonium groups

INTRODUCTION

Quaternary ammonium salt (quats) has been known for over 100 years, and it plays an important role in industry, exerting surface antimicrobial activity.[1] At present, quats derivatives are widely used as disinfectants in agriculture, food processing industry, clinics, etc. Use of the organic cations as disinfectants is particularly important because they possess a high antibacterial activity and a broad spectrum of antimicrobial activity. Quats used as cationic biocides have a common structure of long alkyl chains in the molecule. Trimethyl-*n*-alkylammonium salts are representative of this group.[2] Interestingly, they kill bacteria and fungi by interaction with the constituents of the cell envelope, interaction with the negative charges of the cell wall, destabilization, and weakening of the cytoplasmic membrane (thanks to their lipophilic moiety) leading to a loss of cytoplasm constituents because of the very high osmotic pressure.[3] On these grounds, it is considered that electrostatic interaction of the positive charges on the molecules of the antibacterial agents with the negatively charged species present in the cytoplasmic membranes can play an important role in course of the killing of bacteria using these bacterial agents. This feature endows the synthesis of antibacterial polymers with a new direction.

The antibacterial activities of superabsorbent polymer by introducing antibacterial agents with low molecular weight into polymer chains like P(OAD-MAC-AM-MBAAm),[4] P[AM-*co*-4VP),[5] P(TRVB-AAm-MBAAm),[6] P(TRVB-NIPAAm-MBAAm),[7] and PS grafting quaternary ammonium groups[8] were widely studied. Undoubtedly, these results provide an important and scientific basis for developing a new generation of polymeric antibacterial superabsorbents, which have some advantages over low-molecular-weight agents because they are more stable against volatilization, dissolution, and diffusion to the surfaces of materials to be protected. But it is a shortcoming that the absorbency is deficient. In view of these, we introduced cationic groups ($-N^+R_4$) and anionic groups ($-COO^-$) into the polymer chains. The obtained resin is amphoteric polyacrylamide, which is representative of a special sort of polymers that contain both positively and negatively charged groups along the macromolecular backbone. The combination of $-N^+R_4$ and $-COO^-$ for water-insoluble polymer can be found to increase the water absorbency of antibacterial resin.

In our previous research article,[9] the preparation, characterization, water absorbency, and antibacterial activity of amphoteric terpolymers P[AA-AM-(DMAEA-

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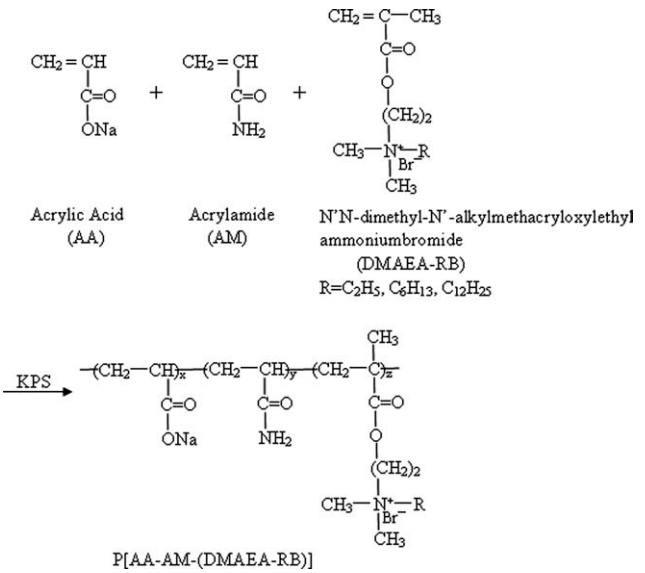

Figure 1 Synthesis of the AA-AM-DMAEA-RB (*R* = ethyl/hexyl/dodecyl).

EB)] (acrylic acid (AA), acrylamide (AM), and *N*,*N*′-dimethyl-*N*-ethylmethacryloxylethyl ammoniumbromide (DMAEA-EB)) with varied compositions were investigated. It was demonstrated that the hydrogel exhibited high antibacterial activity against bacteria tested. Following previous work, in this study, we synthesized a family of three polyampholytic hydrogels with different quaternary ammonium groups. By investigating their swelling behavior and antibacterial activity, we designed and performed some antibacterial experiment tests using *Escherichia coli* and *Staphylococcus hyicus* as test bacteria and sterilized 0.9 wt % NaCl solution as the suspension medium of the bacterial cells. The results of these experiments are analyzed and discussed, and the behavior features of the antibacterial activity of three kinds of polyampholytic hydrogels are summarized. It was found that the insoluble structure and the adsorption of bacteria on the antibacterial agent play an important role in the antibacterial behavior. A novel model for antibacterial activity of insoluble ammonium salt was set up.

EXPERIMENTAL

Materials

AM was purchased from Tianjin Bodi Chemical Factory (Tianjin, China) and recrystallized in acetone solvent before use. AA was distilled under reduced pressure. Sodium acrylate was prepared from AA; *N*,*N*′-methylenebisacrylamide (NMBA) and potassium persulfate (KPS) were obtained from Shanghai Chemical Reagent Co. (Shanghai, China) and recrystallized in distilled water. 2-(Dimethylamino) ethylmethacrylate (DMAEA) was obtained from Xinyu Chemical Industries (Wuxi, China). Bromoethane, bromohexane, and dodecyl bromide were supplied by Guoyao Chemical Reagent Co. (Shanghai, China) and were used without further purification. Mono-octadecyl phosphate was synthesized in our laboratories.[10] All reagents were of analytical grade. Peptone and agar were purchased from Wuhan Tianyi, China. Yeast extract was procured from Guoyao Chemical Reagent Co. All reagents were of biochemical grade. *E. coli* (XL10-gold) and *S. hyicus* (NAT-SEL0502) were provided by the School of Life Sciences of Hubei University.

Synthesis of DMAEA-RB monomer

DMAEA and acetone with equal volume and 0.00034 g/mL hydroquinone (according to the dosage of DMAEA) were added into a 100-mL flask with a reflux condenser. The mixture was magnetically stirred until the inhibitor dissolved, then charged with equal volume of bromoethane, bromohexane, and bromododecane, respectively. After that, it was heated at 45°C in water bath for 10 h, then cooled in air, filtered, and cleaned with ethyl ether repeatedly. The product (DMAEA-RB) was dried under vacuum.[11]

DMAEA-RB with ethyl, hexyl, and dodecyl groups in ammonium groups were obtained and were abbreviated as DMAEA-EB (*N*,*N*′-dimethyl-*N*-ethylmethacryloxylethyl ammoniumbromide), DMAEA-HB (*N*′-dimethyl-*N*-hexylmethacryloxylethyl ammoniumbromide), and DMAEA-DB (*N*′-dimethyl-*N*-dodecyl methacryloxylethyl ammoniumbromide).

Inverse suspension polymerization

Polymer hydrogels (P[AA-AM-(DMAEA-RB)]) used in this study were prepared by inverse suspension polymerization of AA, AM, and DMAEA-RB (Fig. 1). DMAEA-RB with either a ethyl, hexyl or dedecyl group in their nitrogen groups respectively was used. Cyclohexane was used as the continuous phase and mono-octadecyl phosphate as nonionic surfactant. NaAAc solution, obtained by neutralizing 75% of AA, AM, DMAEA-RB, and 0.01 mol % NMBA were dissolved in 10 mL of deionized water containing KPS, and the mixture was bubbled with dry nitrogen to remove the dissolved oxygen. This solution was slowly poured into 30 mL of cyclohexane containing 0.15 g of mono-octadecyl phosphate, which was previously dissolved at 65°C ± 2°C and purged with dry nitrogen in a 100 mL three-necked round-bottom flask. The reaction mixture was mechanically stirred at 250 rpm under nitrogen atmosphere. The polymerization was allowed to proceed for 2.5 h at 72°C. After polymerization, the beads were separated from the oil phase and were washed several times with the mixture of water and

ethanol (1 : 9 v/v). The product was then dried in an oven at 60°C to constant weight. White fine particles were obtained.

Measurement of water absorbency of the resin: Suction filtration method

Accurately weighed 300 mg dry superabsorbent polymers (W, the average size of copolymer gels were 150–180 μm) were immersed in a 500-mL beaker containing an excess amount (V_0) of deionized water or 0.9 wt % NaCl (aq) solutions for 24 h to reach the swelling equilibrium at room temperature. The completely swollen copolymer was filtered through a 100-mesh standard screen at least for 15 min until no water being filtered. Then, the volume of filtered water V_1 was measured.

Water content (Q_s) was calculated by the equation below:

$$Q_s = \frac{(V_0 - V_1)}{W} \times \rho_{H_2O} \quad (1)$$

where V_0 and V_1 are the initial and filtered volume of solution, respectively, W is the weight of the dry resin, and ρ_{H_2O} is the density of water.

Measurement of antibacterial activity

Preparation of nutrient and the initial cell suspension

All procedures in the antibacterial tests for polymers were carried out under aseptic conditions and performed using a batch method. Freeze-dried ampoules of *E. coli* and *S. hyicus* were opened, and a loopful of each culture was spread to give single colonies on nutrient agar and incubated at 37°C for 24 h. Three representative colonies were selected with a wire loop, placed in 5 mL nutrient broth (peptone, 1.0 g; NaCl, 1.0 g; and beef extract, 0.5 g in 100 mL sterile distilled water, pH 7.2), then incubated at 37°C at 150 rpm overnight. 0.5 mL of the cell suspension was pipetted out from the container and quickly mixed with 4.5 mL of sterilized physiological saline, and then decimal serial dilutions were prepared from this by taking 0.5 mL into 4.5 mL of sterilized physiological saline and mixing. The cell number was determined by a colony count method. At this stage, the culture of *E. coli* or *S. hyicus* contained about 10^9 to 10^{10} CFU/mL, respectively.[5,7,8,12–15]

Contact of polymer gels with bacteria

0.1 g copolymers with optimized experimental results and 20 mL sterile 0.9 wt % NaCl solution were placed in 100-mL conical flask with cotton stopper, and the polymers were swollen for 24 h. After that, 0.1 mL of the cell suspension was added to the flask, and the mixture was continually shaken at 37°C 150 rpm. At prescribed time intervals, 0.5 mL of the cell suspension was pipetted out from the container and quickly mixed with 4.5 mL of sterilized physiological saline, and then decimal serial dilutions were prepared according to the method described in the previous section. The colonies were counted after the inoculated plates were incubated at 37°C for 24 h, and the number of viable cells $N(t)$ was calculated from those of the colonies. Blank test was finished by the same means without polymer. The counting was done in triplicate every time.

Measurement of viable cell numbers after contacting with terpolymer gels

0.1 g copolymers with varying DMAEA-RB content and 20 mL sterile 0.9 wt % NaCl solution were placed in 100-mL flask, and the polymers were swollen for 24 h, After that, 0.1 mL of the cell suspension (*E. coli* and *S. hyicus*) were added to the flask, which was shaken for 30 min at 37°C at 150 rpm. The suspension pipetted was diluted several times and treated with means as reported in the previous section, and blank test was finished in the same manner without polymers.

Contact of an insoluble quaternary ammonium-type polymer with dead bacterial cells

An experiment was made to clarify whether or not an insoluble polymer can adsorb or adhere dead bacterial cells. The *E. coli* cell suspension only contacting dead cells was prepared by an *E. coli* cell suspension, in which the initial viable cell number was around 10^8 CFU/mL being heated at 121.3°C, 1.05 kg/cm^3 for 25 min. The contact study of an insoluble polymer with dead bacterial cells was performed with the *E. coli* cells suspension only containing dead cells. After the cells were in contact with a polymer sample for a prescribed time, an appropriate amount of the supernatant was pipetted out and diluted with suitable volume of nutrient. Its optical density at 660 nm (OD_{660}) was measured with a spectrophotometer. The OD_{660} value of a bacterial suspension indirectly reflects its cell concentration. The lower the OD_{660} value is, the lower the cell concentration in the bacterial suspension is, and *vice versa*.[16,17]

RESULTS AND DISCUSSION

Effect of DMAEA-RB content on the water absorbency of the AA-AM-(DMAEA-RB) copolymer gels

In general, the swelling ratio, which corresponds to water content, of hydrogels can be expressed by eq. (2)[7]

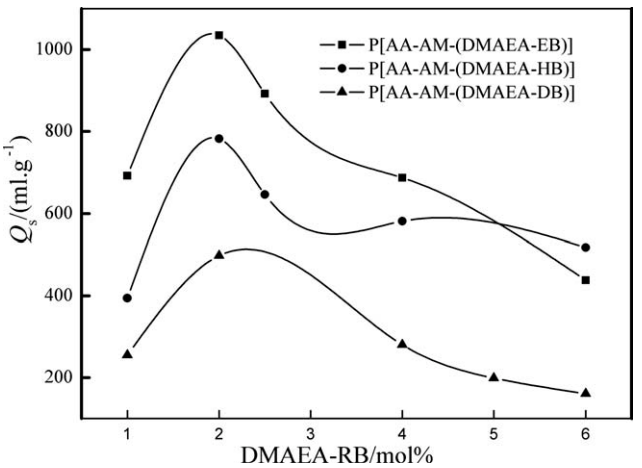

Figure 2 Effect of DMAEA-RB content in the gels on the absorbency of copolymer in deionized water. [Conditions: AA/AM, 1 : 1 (mol); NMBA, 0.01 mol %; $K_2S_2O_8$, 0.08 mol %; neutralization degree, 75%; monomer concentration, 5 mol/L.]

$$Q^{5/3} = \left\{ \left[\frac{1}{2} \times \frac{i}{V_u} \times \frac{1}{S^{1/2}} \right]^2 + \frac{1/2 - X_1}{V_1} \right\} \Big/ \frac{v}{V_0} \quad (2)$$

where Q is the swelling ratio, i/V_u is the charge density attached to the polymer matrix, $(1/2 - X_1)$ is the affinity between the polymer matrix and water, $S^{1/2}$ is the ionic strength of the outer solution, and v/V_0 is the cross-linking density.

This equation indicates that the water content of the hydrogels depends on the hydrophilicity, cross-linking density, charges of the copolymers, and concentration of salts in the outer solution. To reveal the effect of DMAEA-RB on the swelling behavior, the water content of AA-AM-(DMAEA-RB) copolymer, which was obtained using a constant amount of the cross-linking agent and various amounts of DMAEA-RB in the feed, was measured. The results are shown in Figure 2.

The water absorbency of copolymer has a maximum with the increase of the DMAEA-RB content. The water absorbency of gels is dependent on ionic osmotic pressure, cross-linking density, and the affinity of the gel for water. The cross-linking density of copolymer and the ionic concentration in the external solution were fixed. The water absorbency for the gel is dependent on the concentration of the fixed charge in the gel and the affinity of the gel for water. When the cationic monomer DMAEA-RB was introduced into the copolymer, the bromide ion (Br^-) was dissociated and the quaternary ammonium group (R_4N^+) with a positive charge was formed. The affinity of the quaternary ammonium group (R_4N^+) for water is stronger than that of the carboxylate group (COO^-). However, the quaternary ammonium group (positively charged) would bind with the carboxylate group (negatively charged); thus, the fixed charge concentration of the polymer network decreases. This behavior reduces the negative charge repulsion of the polymer network, and the water absorbency of the gel decreases.

On the other hand, the order of water content of the copolymers is as follows: AA-AM-(DMAEA-EB) > AA-AM-(DMAEA-HB) > AA-AM-(DMAEA-DB) copolymers. This indicates that the AA-AM-(DMAEA-RB) copolymer with a shorter alkyl chain has a higher affinity for water. This can be explained by the following: the longer the alkyl chain, the higher the hydrophobic interaction, the littler affinity for water, and so the fewer water absorbency of copolymer.

Effect of initiator, KPS, concentration on the water absorbency of the AA-AM-(DMAEA-RB) copolymer gels

The effect of the initiator amount on the water absorbency of the superabsorbents is obviously shown in Figure 3. The water absorbency increases with the increase in the total initiator amount up to 0.08 mol % and decreases with the further increase in the initiator amount. The maximum absorbency is obtained at KPS 0.08 mol %.

Based on general kinetics, the rate of polymerization depends on the concentration of monomers and initiators for bimolecular termination. With increasing the KPS content, large number of free radicals was produced; these primary radicals will initiate the monomers to form free radicals of monomers, which propagate monomer molecules in succession

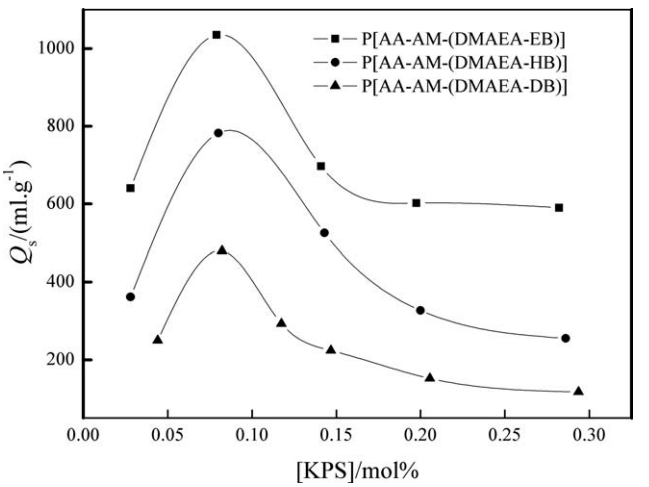

Figure 3 Effect of KPS concentration on water absorbency of copolymer in deionized water. [Conditions: AA/AM/(DMAEA-RB), 49 : 49 : 2 (mol); NMBA, 0.01 mol %; neutralization degree, 75%; monomer concentration, 5 mol/L.]

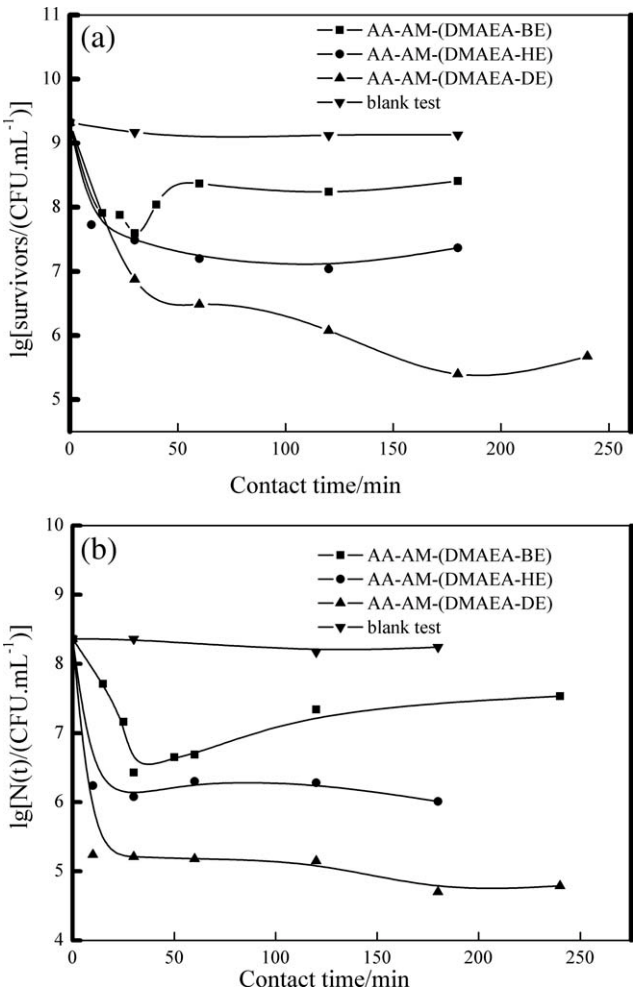

Figure 4 The effect of contact time on antibacterial activity of AA-AM-(DMAEA-RB) copolymers against (a) *E. coli* and (b) *S. hyicus*. [Conditions: A/AM/(DMAEA-RB), 49 : 49 : 2 (mol); NMBA, 0.01 mol %; $K_2S_2O_8$, 0.08 mol %; neutralization degree, 75%; initial viable cell concentration of *E. coli* suspension, 4.69×10^{10} CFU/mL; initial viable cell concentration of *S. hyicus* suspension, 4.78×10^9 CFU/mL; copolymer, 0.100 g.]

to form a large polymeric radical and a dead polymer in the termination step. Thus, in free-radical polymerization, the initiator has an effect on the polymerization; it also affects the cross-linking degree and molecular weight between the two cross-linking points. With increasing the KPS content, which led to more chain ends in the network and short average of kinetic chain length, swelling capacity will be increased.

However, when the initiator concentration is higher, the synthesized copolymer has a higher cross-linking density and increased number of produced radicals led to terminating step via bimolecular collision, resulting in shortening the macromolecular chains and reducing the available free volumes within the hydrogel, which affects the water absorbency.[9,18,19]

Antibacterial activity of AA-AM-(DMAEA-RB) copolymer

The effect of contact time on antibacterial activity of copolymer gels

Figure 4(a,b) shows the changes in viable cell numbers of three kinds of AA-AM-(DMAEA-RB) (49 : 49 : 2 mol ratio) copolymers containing DMAEA-EB, DMAEA-HB, and DMAEA-DB, respectively. The insoluble quats have good ability to remove bacteria in suspension. The order of antibacterial activity of the AA-AM-(DMAEA-RB) copolymers is as follows: AA-AM-(DMAEA-DB) > AA-AM-(DMAEA-HB) > AA-AM-(DMAEA-EB). It has been reported that the antibacterial activity of water-insoluble polymers containing cationic groups such as quaternary ammonium groups or phosphonium groups is affected by both electrostatic interaction and hydrophobic interaction between the surface of water-insoluble polymers and bacteria.[7] Kanazawa et al.[20] also studied the antibacterial activity of phosphonium salts with different alkyl chains as substituent against *E. coli*. They found that the surface antibacterial activity of tested sample increased with increasing hydrophobicity of the substituents.

Related article reported that the penetration of the insoluble ammonium polymers into the viable cells is intensively affected by the flexibility of the polymer backbone and mobility of the functional group. With more flexible backbone, the ammonium group is easier to reach the cytoplasmic membrane.[8] That is, the AA-AM-(DMAEA-RB) copolymers with longer alkyl chains had higher antibacterial activity than those with shorter alkyl chains in the ammonium groups.

The antibacterial activity of AA-AM-(DMAEA-RB) copolymers against *E. coli* and *S. hyicus* were investigated after contacting with different time. In addition, the viable cell number ($N(t)$) decreases within 30 min of contact, and kept this level afterward. But for AA-AM-(DMAEA-EB) copolymers, the behavior were more complex. $N(t)$ increases from 30 to 180 min. This result is similar to that reported in reference,[16] in which the antibacterial process was summarized as chemical reactions in the following way:

$$\mathrm{L} + \mathrm{P} \longleftrightarrow \mathrm{LP} \text{ physical adsorption of viable cells} \quad (3)$$

$$\mathrm{D} + \mathrm{P} \rightarrow \mathrm{DP} \text{ physical adsorption of dead cells} \quad (4)$$

$$\mathrm{L} + \mathrm{P} \rightarrow \mathrm{DP} \text{ killing of the viable cells} \quad (5)$$

where, L is the viable cells, P is the insoluble ammonium polymers, D is the dead cells, LP is the living cells adsorbed on polymers, and DP is the dead cells adsorbed on polymers. Equation (3) directly decreases the number of viable cell in suspension, whereas eq. (5) is the key step in killing the bacteria.

TABLE I
Determination of OD_{660} in Living and Heat-Killed *E. coli* Cell Suspension by Contact of *E. coli* with AA-AM-(DMAEA-DB) Copolymer[a]

E. coli cell suspension	OD_{660} of cell suspension				
	Before contact	After contact at the following contact times, t (min)			
		2	30	60	90
Heat-killed	0.060	0.013	0.008	0.006	0.007
Living	0.044	0.018	0.015	0.013	0.012

[a] Initial viable cell concentration is 1.03×10^8 CFU/mL, 0.5 mL of *E. coli* suspension, copolymer 0.1 g, 39.5 mL 0.9 wt % NaCl solution.

It is assumed that the adsorption of viable cells is a reversible process, but the adsorption of the dead cells is an irreversible one because dead cells lose their mobility and the interaction between dead cells and ammonium groups is strong.

Because the bacteria cannot incubate under a low nutrient concentration or within such short period of time, the recovery of $N(t)$ may be due to the release of the viable bacterial cells from the polymer surface.

As shown in Figure 4, the AA-AM-(DMAEA-EB) copolymer may first absorb the living bacterial cells rather than kill them, but keep them alive in the first 30 min. This may result in the decrease in $N(t)$. As shown in reaction (3), the absorption and release of the viable cells is a reversible process. The agent may also absorb the dead cells, as shown in reaction (4); the process is supposed to be an irreversible one. When the dead cells accumulate on the surface of the agent and shield the cationic center, the ammonium salt cannot absorb the living cells any more, and the previous absorbed viable cells are released from the insoluble ammonium salt. Therefore, the number of viable cells in suspension would increase after contacting with copolymer.

Interaction between an insoluble quaternary ammonium type polymer and killed bacterial cells

To verify whether an interaction between the polymer and dead cells can occur, it is desirable to do further research into the interaction between an insoluble quaternary ammonium type polymer and dead *E. coli* cells.

The change in the OD_{660} value of the heat-killed cell suspension that was in contact with the polymer AA-AM-(DMAEA-DB) was measured at different prescribed contact times. For the sake of convenient comparison and analysis, a control test was made with the living-cell suspension, the same suspension used to prepare the heat killed-cell suspension. The results are compiled in Table I.

Table I indicates that the corresponding OD660 value after AA-AM-(DMAEA-DB) copolymer is brought into contact with the living-cell suspension and the heat-killed-cell suspension. The OD_{660} of heat-killed *E. coli* cell suspension is 0.06 before the contact, and it decrease to 0.008 after the contact with P[AA-AM-(DMAEA-DB)] 0.1 g for 0.5 h. The OD_{660} value of viable cell suspension is 0.044 before the contact, and after contact with the copolymer for 0.5 h, it is 0.015. The OD_{660} value of living-cell suspension decreases distinctly, and so does the OD_{660} value of the heat-killed-cell suspension. This fact reflects the reduction in the dead cell number in the heat-killed cell suspension. It is concluded that the P[AA-AM-(DMAEA-DB)] polymer is also able to capture dead bacterial cells.

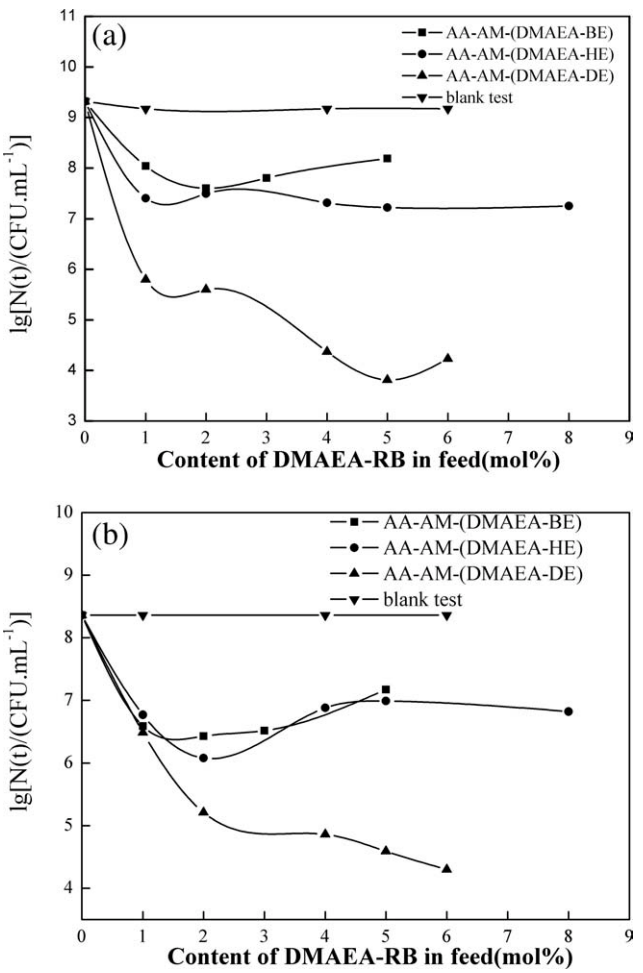

Figure 5 Change in number of viable cells after contact with AA-AM-(DMAEA-RB) copolymer with different monomer ratio in feed. Bacteria suspension: (a) *E. coli*, (b) *S. hyicus*. [Conditions: AA/AM, 1 : 1 (mol); NMBA, 0.01 mol %; $K_2S_2O_8$, 0.08 mol %; neutralization degree, 75%; initial viable cell concentration of *E. coli* suspension, 4.69×10^{10} CFU/mL; initial viable cell concentration of *S. hyicus* suspension, 4.78×10^9 CFU/mL; copolymer, 0.100; contact time, 30 min.]

The effect of DMAEA-RB content on antibacterial activity of copolymer

At certain contact time (30 min), the antibacterial activity of P[AA-AM-(DMAEA-RB)] with different content of DMAEA-RA (mol %) against *E. coli* and *S. hyicus* was investigated. As shown in Figure 5, the concentration of DMAEA-RB has a special effect on the antimicrobial activity of polyampholytic hydrogels, which is different from normal polycation. In general, the higher the content of antimicrobial agent in normal polycation, the better the antibacterial activity.[8,10–15] The result of AA-AM-(DMAEA-DB) copolymer followed this rule; the hydrophobicity of flexible longer alkyl chains plays an important role in the ability of disinfection. However, the antibacterial activity of AA-AM-(DMAEA-EB) and AA-AM-(DMAEA-HB) copolymers in our experiment is dissimilar, and this result can be confirmed by its influence on the water absorption. This result indicates that the alkyl chain length strongly affects the antibacterial activity of copolymers, and bacterial cells were not adsorbed on AA-AM-(DMAEA-RB) copolymers with shorter alkyl chains by hydrophobic interaction but by electrostatic interaction. That is, because the DMAEA-RB (*R*-ethyl/hexyl) content in the feed is above 2 mol %, the quaternary ammonium group (positively charged) would bind with the carboxylate group (negatively charged), and the fixed charge concentration of the polymer network decreases. This behavior reduces the charge repulsion between the polymer chains, and the network slack is restricted, and the electrostatic interaction between the positively charged polymer surfaces and the negatively charged cell surfaces weaken; this contributes to the reversion of the viable cell number $N(t)$.

CONCLUSIONS

On the basis of this investigation into the swelling and antibacterial activity behavior of insoluble AA-AM-(DMAEA-RB) terpolymers with alkyl chains of different lengths (DMAEA-RB) (*R*-ehtyl/hexyl/dodecyl), several important conclusions can be drawn:

1. The water content of the AA-AM-(DMAEA-RB) terpolymers decreased with the increasing length of alkyl chains in quaternary ammonium group in the terpolymers and increased with the increasing molar ratio of DMAEA-RB to 2 mol % and then decreased.
2. The copolymer exhibited higher antibacterial activity with increasing chain length of alkyl groups in ammonium groups. For P[AA-AM-(DMAEA-DB)], the DMAEA-DB content is higher and contact time is longer, and its antibacterial activity is better. However, when the contact time and quaternary ammonium content were above 30 min and 2%, respectively, the amount of live cells $N(t)$ in a cell suspension increased in the presence of P[AA-AM-(DMAEA-EB)] or P[AA-AM-(DMAEA-HB)].

It can be concluded that the cationic polymer of quats is a soluble copolymer with multiple functions, has antimicrobial activity and water absorbency ability concurrently, and will have wide application foreground in water purification treatment field.

References

1. Juliusz, P. Eur J Med Chem 2003, 38, 1035.
2. Kanazawa, A.; Ikeda, T.; Endo, T. Antimicrob Agents Chemother 1994, 5, 945.
3. Li, G. J.; Shen, J. R.; Zhu, Y. L. J Appl Polym Sci 1998, 67, 1761.
4. Yin, G. Q.; Cui, Y. D. J Funct Mater [in Chinese] 2004, 35, 368.
5. Gao, B. J.; He, S. X.; Guo, J. F. et al. J Appl Polym Sci 2006, 100, 1531.
6. Nonaka, T.; Yanada, K. J.; Watanabe, T. et al. J Appl Polym Sci 2000, 78, 1833.
7. Nonaka, T.; Yanada, K. J.; Watanabe, T. et al. J Appl Polym Sci 2001, 79, 115.
8. Jiang, S.; Wang, L.; Yu, H. J. et al. J Appl Polym Sci 2006, 99, 2389.
9. Li, X. H.; Dong, Q. Z.; He, P. X. J Appl Polym Sci 2009, 112, 439.
10. He, P. X.; Xiao, W. D.; Huang, H. Polym Mater Sci Eng [in Chinese] 1999, 15, 65.
11. Huang, S. Q.; Xun, Z. S.; Chen, Z. G. et al. Chem Adhes [in Chinese] 1997, 3, 128.
12. Peng, X. H.; Peng, X. C. J Appl Polym Sci 2006, 101, 1381.
13. Omidian, H.; Zohuriaan-Mehr, M. J.; Bouhendi, H. Eur Polym J 2003, 39, 1013.
14. Lee, W.-F.; Yang, L. G. J Appl Polym Sci 2004, 92, 3422.
15. Huang, J.; Huang, Z. M.; Bao, Y. Z. et al. J Appl Polym Sci 2006, 100, 1594.
16. Li, G. J.; Shen, J. R. J Appl Polym Sci 2000, 78, 676.
17. Tashiro, T. J Appl Polym Sci 1991, 43, 1369.
18. Zhang, Y. H.; Wang, L. M.; Li, X. H.; He, P. X. J Polym Res. DOI 10.1007/s10965-010-9402-8.
19. Pourjavadi, A.; Harzandi, A. M.; Hosseinzadeh, H. Eur Polym J 2004, 40, 1363.
20. Kanazawa, A.; Ikeda, T.; Endo, T. J Appl Polym Sci 1994, 54, 1305.