

Network Structure and Mechanical Properties of Hydrophobic Association Hydrogels: Surfactant Effect I

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ABSTRACT: Three series hydrophobic association hydrogels (HA-gels) with different concentration of sodium dodecyl sulfate (SDS) were prepared by free radical micellar copolymerization. The backbone of HA-gels was based on the copolymer of acrylamide and a little octylphenol polyoxyethylene ether acrylate with 10 ethoxyl units (OP10~AC). Their mechanical properties were determined by tension tests. It was found that the properties of all three series HA-gels presented a very similar variation tendency with increasing R , the molar ratio of SDS to OP10~AC. In the medium region of R (around 1.23), higher effective crosslinked density and proper match between shorter and longer chains were achieved, so that the HA-gels exhibited better mechanical strengths. In lower R region, it is prone to form the unstable aggregation of OP10~AC with little SDS, while in higher R region some of co-micelles could not act as crosslinkers. Both would reduce the effective crosslinked density and the mechanical strength of HA-gel. In the condition of fixed R , the change of OP10~AC content would remodel the topological structure of network. The HA-gels with better properties could be obtained as the OP10~AC content was varied from 1 to 2% at the proper R . © 2014 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* **2015**, *132*, 41222.

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INTRODUCTION

Hydrogels have been intensively studied as unique smart materials for several decades and widely applied in drug-delivery systems, tissue engineering, artificial actuators, colloid crystals, and separation devices.^{1–5} For some natural and chemically crosslinked hydrogels mechanical strengths and transparencies are weak generally, which restrict their applications largely in some situations.⁶ How to improve the mechanical properties of hydrogels has become an important project. Up to now, some kinds of novel hydrogels with high mechanical strengths have been reported, such as topological gels (TG gels),⁷ nanocomposite hydrogels (NC gels),^{8,9} double network gels (DN gels),¹⁰ and macromolecular microsphere composite gels (MMC gels).¹¹ These hydrogels respectively have extraordinary properties, i.e., high mechanical strengths, long elongations at break, or improved response rates, etc. However, there are some problems still need to be resolved, for instance, complicated synthesis processes, expensive original materials, and poor capabilities of secondary processing.

On the other hand, hydrophobically modified water-soluble polymers (HMWSP), consisting of a long hydrophilic backbone

to which low amounts of hydrophobic constituents are attached, have been comprehensively researched in recent years.^{12–16} Aqueous solutions of HMWSP always exhibit unusual rheological properties, due to the intermolecular reversible association of the hydrophobic groups. It is prone to form the mixed polymer/surfactant aggregations in the presence of ionic surfactants. However, the interaction between HMWSP and ionic surfactants is very intricate.^{17–20} Most researchers have focused on the rheological properties in dilute or semidilute solutions of hydrophobically modified polymers, but there are relatively few communications on their gels properties at higher concentrations. As a matter of fact, physical gels could be formed at concentrated regime, in which the mixed micelles of hydrophobic groups and surfactants would act as crosslinking points.^{21,22} On the basis of this point of view, novel hydrophobic association hydrogels (HA-gels) were synthesized via a free-radical micellar copolymerization technique in our laboratory.^{23–27} In the process, the octylphenol polyoxyethylene acrylates (OP~AC) as hydrophobic monomers were used to copolymerize with acrylamides (AM) in the presence of anionic surfactant sodium dodecyl sulfate (SDS). It was found that not only did the HA-gels exhibit a good toughness and rubber-like elasticity so

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that their network structures could be evaluated by using rubber elastic theory, but also they possessed the capability of self-healing and remolding.^{23,24,26} Compared with other gels, no external crosslinkers was used during the formation of network and the preparation process of HA-gels was relatively simple. Especially, HA-gels possessed the capability of self-healing and remolding under proper conditions, which must be due to the dissociation and re-association of hydrophobic associated cross-linking points.²⁶ Moreover, the properties of the HA-gels can be easily changed by varying the composition in the system.²⁵ These results show that the HA-gels can improve the properties of other hydrogels mentioned above.

Since there is an interaction between the surfactant and HMWSP, it is expected that the content of surfactant will directly influence the network structures and properties of HA-gels. In the present work, the octylphenol polyoxyethylene acrylate with 10 polyoxyethylene units (OP10~AC) was selected as a hydrophobically modified monomer which could dissolve in water even without SDS. Three sets of HA-gels containing different OP10~AC contents were synthesized within a wide range of SDS dosage. The effect of SDS on the network structure and mechanical strength of HA-gels was systematically discussed on the basis of cooperation of SDS and OP10~AC and rubber elastic theory.

EXPERIMENTAL

Materials

Acrylamide (AM, Tianjin Fuchen Chemical Reagent Factory) and potassium peroxydisulfate (KPS) were purified by re-crystallization from distilled water and dried under vacuum at room temperature. Octylphenol polyoxyethylene acrylate (OP10~AC) was synthesized via the previous method (composition and structure analysis are given in Supporting Information Figure S1).²³ Other reagents were purchased from Tianjin Fuchen Chemical Reagent Factory, and used without further purification.

Synthesis of HA-Gels

HA-gels were synthesized by free radical micellar copolymerization. As a typical example, to prepare HA-OP4S3 gel, an aqueous solution consisting of water (19.02 mL), AM (2.5 g, 35 mmol), OP10~AC (0.9848 g, 1.4 mmol), and SDS (0.75 g, 2.6 mmol) was obtained after ultrasonic. Next, 1.74 mL KPS aqueous (10 mg/mL) was added to the former solution while stirring at room temperature. At the moment, the total mass of the reaction system was 25 g, the molar percentage of OP10~AC relative to AM was 4%, and the weight concentrations of AM and SDS against the total reaction solution were 10 and 3 wt %, respectively. The reaction mixture was purged with nitrogen for several minutes and then equilibrated for another 30 min at room temperature. Finally, the polymerization was allowed to proceed in water bath at 50°C for 5 h.

In this study, all the gel samples were expressed as HA-OP m Sn gels. Here, $m\%$ denotes the molar percentage of OP10~AC against AM, which was varied from 1 to 4%. The weight concentration of SDS relative to water was expressed as n , which was varied within the range of 0–7 wt %. For example, HA-OP2S3 indicated that the molar percentage of OP10~AC

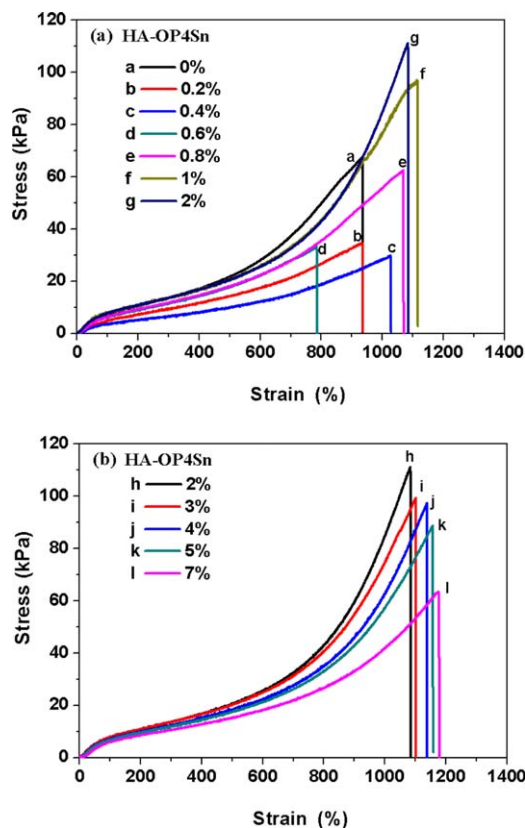


Figure 1. Stress–strain curves of HA-OP4Sn gels with various SDS contents. (a) 0–2 wt %, (b) 2–7 wt %. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

against AM was 2%, and the weight concentration of SDS relative to water was 3 wt %.

Mechanical Measurements

The tensile stress–strain curves of the HA-gel samples were measured using a universal tester (AG-I, 1KN, Shimadzu, Japan) under the following condition: 25°C; gauge length L_0 : 25 mm; crosshead speed: 100 mm/min. The cylindrical samples were 7 mm in diameter and 50 mm in length.

Optical Transmittance

Optical transmittances of the HA-OP2Sn gels were detected using a UV/vis spectrophotometer (UV-2450, Shimadzu Corporation, Kyoto, Japan) at 600 nm and 25°C.

RESULTS AND DISCUSSION

Tensile Behavior

Figure 1 shows the stress–strain curves of HA-OP4Sn gels. It can be seen that the mechanical properties change greatly as SDS concentration (C_{SDS}) varying. In the low concentration range, the mechanical strength decreases along with the increase of C_{SDS} , and the lowest strength occurs when C_{SDS} is 0.4 wt %. With further adding SDS, the mechanical strengths of HA-gels are improved gradually. Up to 2 wt % C_{SDS} , the obtained gel (HA-OP4S2 gel) achieves the highest tensile strength and a better elongation at break. Once C_{SDS} is more than 2 wt %, the mechanical strengths tend to drop again. For

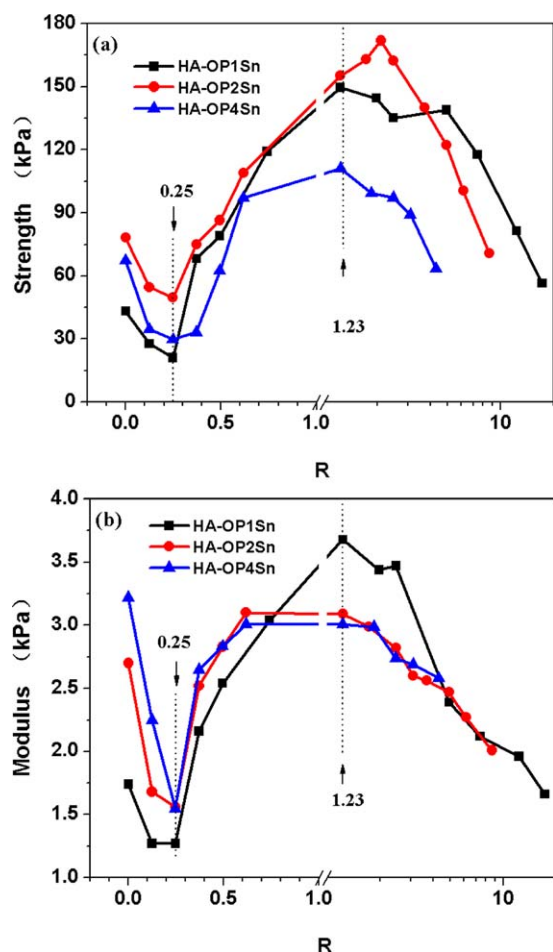


Figure 2. Mechanical properties as a function of R for HA-gels with different OP10~AC contents. (a) Tensile strength and (b) modulus. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

convenience, the whole variation tendency was denominated as "down-up-down".

For HA-OP1Sn and HA-OP2Sn gels, the OP10~AC contents have decreased to 1 and 2 %, respectively, in the same range of SDS. Their mechanical strengths similarly exhibit "down-up-down" tendency with the increase of C_{SDS} , but the C_{SDS} at which the lowest strength of gel occurs tends to fall along with decreasing OP10~AC content (Supporting Information Figure S2). By adopting the molar ratio (denoted by R) of SDS to OP10~AC as a variable, we can find that all three series HA-gels exhibit the lowest strengths when R value equals to 0.25, and the highest strengths appear at $R = 1.23$ for HA-OP1Sn and HA-OP4Sn gels [Figure 2(a)]. Based on stress-strain curve, the modulus can be estimated in low strain range. The results are shown in Figure 2(b). It can be seen that the minimum modulus also appears at $R = 0.25$ for all HA-gel samples and the maximum modulus again appears around $R = 1.23$ for HA-OP1Sn gels. For HA-OP2Sn and HA-OP4Sn gels, higher moduli appear in a range of R from 0.6 to 2 in which the values of moduli have no significant differences. Above phenomena suggest that R , the molar ratio of SDS to OP10~AC, is an important parameter determining the properties of HA-gels no matter how much OP10~AC unit is inducted.

Cooperation of SDS and OP10~AC

The chemical structure of monomer OP10~AC is very like it of octylphenol polyoxyethylene with 10 ethoxy units, a nonionic surfactant (Triton X-100), but the end hydrogen is replaced by acryloyl group. It is found that the OP10~AC itself do is a surfactant with critical micelle concentration (CMC) 0.045 mmol/mL which is determined via surface tension method (Supporting Information Figure S3). Therefore, it is expected that OP10~AC and SDS will tend to form mixed micelles or co-micelles, rather than solubilized micelles of SDS in aqueous solution beyond CMC. The aggregation behavior of the mixture of Triton X-100 and SDS in aqueous solution has been investigated in detail by Shannigrahi and Bagchi,^{28,29} which is helpful for us to analyze the cooperation of SDS and OP10~AC by means of comparing the construction of co-micelle formed by SDS/Triton X-100 and the R value which affected mechanical strengths of present HA-gels.

It has been attested that the aggregation number of molecules in a SDS/Triton X-100 co-micelle (N_{agg}) decreases very rapidly when the ratio (R') of SDS to Triton X-100 increases from zero to about 0.5, while N_{agg} remains almost constant (~ 62) after R' is more than 1.0. Correspondingly, both tensile strengths and modulus of the HA-gels accordingly reach the maximum values at around $R = 1.23$, then gradually decrease with further increase of R (Figure 2). That is, the HA-gels with better strength can be obtained only when R has a suitable value before which N_{agg} just reaches constant. Since the OP10~AC remains an aptotic content in each system, the number of co-micelle will increase with the increase of R , whereas, the strengths of the HA-gels get weaker and weaker after R is more than 2. It means that not every co-micelle can play a job of crosslinking point if SDS is over introduced. The OP10~AC units must be asymmetrically distributed among co-micelles in HA-gels. There are some co-micelles containing less OP10~AC units. They append only a single chain and contribute nothing to form network. The similar effect of surfactants on hydrophobic association was also found in HMWSP solutions systems and investigated with several methods.³⁰⁻³²

In the region of R from zero to about 0.5, the modulus and strengths of HA-gels experienced a V-shaped change. A turning point appears when R reaches about 0.25 (SDS/OP10~AC = 1 mol/4 mol). Two opposite trends about aggregation behavior of co-micellar are worthy of note. On the one hand, the introduction of SDS results in rapidly increasing co-micellar number and the co-micelles tend to be stable because of repulsion of charges. It will enhance the modulus of HA-gel due to the increase of crosslinked density. On the other hand, adding small amount of SDS maybe disturbs the aggregation manner of OP10~ACs, so that some unstable and complex aggregation micro-domains with larger size are formed.^{33,34} It will decrease the homogeneity and the apparent crosslinked density of the network. As a result, the modulus and strengths of HA-gels will become weak. It seems that the latter effect is predominant before the turning point, while the former becomes predominant after this value. On the basis of these two effects as well as the discussion for adding more SDS mentioned above, the "down-up-down" variation tendency about the mechanical properties of HA-gels can be reasonably explained.

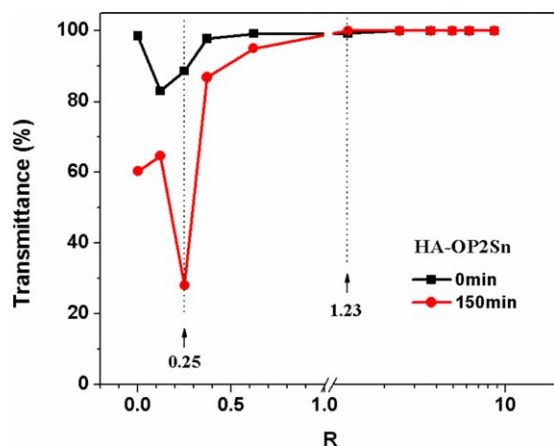


Figure 3. Changes in optical transmittances as a function of R for both original solutions and hydrogels of HA-OP2Sn gels. Optical wavelength: 600 nm. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

It should be mentioned that the behavior of disturbing aggregation by adding a little SDS was not observed in the pure Triton X-100 system. This special phenomenon occurs at present system maybe due to the end acryloyl group of OP10~AC. It is merely a conjecture on the basis of the experimental result. More direct evidences and its detailed mechanism are still unknown.

Transparency

The aggregation behavior of surfactants can be reflected at the turbidity in its dispersion system when the size of association domain is big enough.³⁵ Figure 3 shows the transmittances of the HA-OP2Sn system before and after polymerization. It can be seen that the transmittance is degraded as R is less than 1, and the worst transmittance occurs at $R = 0.25$ for the HA-gel. These indicate that association domains with larger size have been formed in the situations of reducing SDS, which is in agreement with above conjecture for the aggregation behavior. In this R range, all transmittances become worse after polymerization, meaning that structure reorganization have taken place. The micelles formed by OP10~ACs themselves, especially, are also unstable in the polymerization process although the original solution is transparent. In fact, the transmittance reflecting structure evolution is a function of reaction time, which will be discussed elsewhere.

Elastic Parameters

The reduced stress σ^* is defined as following equation:

$$\sigma^* = \frac{\sigma}{\lambda - \lambda^{-2}} \quad (1)$$

where σ and λ denote stress and elongation ratio, respectively, calculated by the tensile curve. We may get more information concerning the network structure of HA-gels via investigating variation of σ^* on the basis of the rubber elastic theory.³⁶ Figure 4 shows a typical plot of the reduced stress against the reciprocal elongation ratio for HA-OP2S1 gel. It can be seen that the σ^* exhibits a large and abrupt increase at high elongation region, which reflects the limited chain extensibility and significant toughness of the network. The similar behavior is also observed for other HA-gel samples (Supporting Information Figure S4). This inflection point directly gives us two interesting

parameters, λ_{infl} and σ_{infl} , in the circumstance of limited extension of the network chains. Moreover, other two parameters, C_1 and C_2 , can be obtained from the intercept and slope of the fitting line showed in Figure 4, which is according to the semiempirical Mooney–Rivlin equation:

$$\sigma^* = 2C_1 + 2C_2\lambda^{-1} \quad (2)$$

In medium elongation region before reaching λ_{infl} , the behavior of σ^* decrease with λ have been theoretically interpreted by Flory and other researchers.^{37–42} According to their point of view, the σ^* was decreased results from the effect of chain constraints which tends to becoming weak in larger λ . On the other hand, Mark has detailed investigated the influences of network structures and swelling on the Mooney–Rivlin parameters.^{36,43,44} For the swollen samples of crosslinked rubber, it was found that C_2 rapidly fell with the increasing of swelling degree while C_1 was no significantly changed.⁴⁴ By using Flory theory reviews Mooney–Rivlin parameters, it is suggested that C_1 mainly reflects the permanently effective crosslinking degree and C_2 reflects the extents of chain constraint and its release from entanglement, because swelling does not destroy the crosslinking point but do reduce the chain constraint in the rubber network.

Figure 5 shows the variations of parameters C_1 and C_2 versus R for the three series HA-gels. The "down-up-down" variation tendency appears again and the turning point or turning region reasonably occurs at the same place as mentioned above. Since all HA-gel samples nearly possess the same total concentration or swelling degree, the variation of C_2 must mainly result from the network structure. The fact of similar variation tendencies of C_1 and C_2 with R reveals that the more degree of permanent crosslinking, the more extent of chain constraint in the state of no or small deformation. In other words, the crosslinking will enhance the chain constraint. In the less R range, the data of C_2 are relatively small, which relates to the instability of micelles mentioned above. This instability will result in the chain concentration fluctuation of HA-gels in larger scale although the total concentration is constant. The high concentrated domains are formed around the complex co-micelles with larger size, which maybe move like rigid bodies in the deformation process. At the same time lower concentration is induced in the other

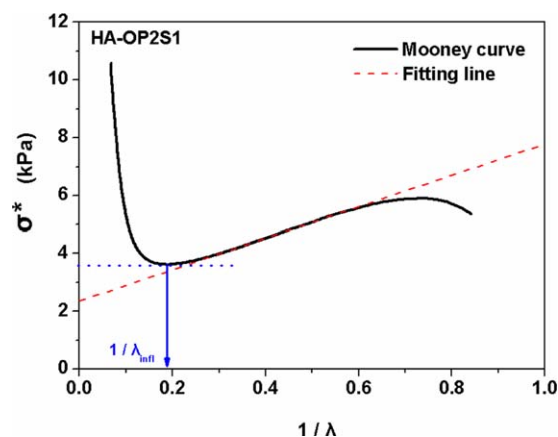


Figure 4. Dependence of σ^* on λ^{-1} for HA-OP2S1 gel. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

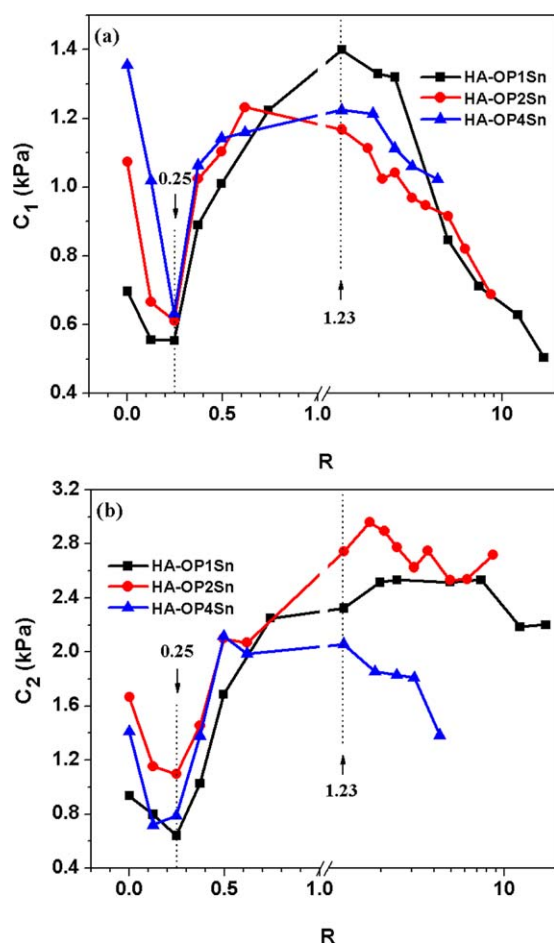


Figure 5. Variations of Mooney parameters (a) C_1 and (b) C_2 versus R for the three series HA-gels. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

section of HA-gel so that the movement of chains becomes relatively easy. Both will weaken the effect of chains constraint or drop C_2 . In the high R range, C_1 is reduced due to over introducing SDS. However, C_2 does not drop so quick as C_1 for HA-OP1Sn and HA-OP2Sn gels, reflecting more homogenized state of the chains concentration have been reached.

The ratio C_2/C_1 is a function of crosslinked functionality. For the end-linked poly (dimethylsiloxane), it has been found that C_2/C_1 decreases to near zero if the functionality is more than 10.^{35,45} In present HA-gels, the co-micelle can act as a crosslinker. If each OP10~AC unit in a co-micelle could link a different chain, its functionality would be very high. For instance, the crosslinked functionality could reach to 56 when $R = 1.23$ (suppose $N_{agg} = 62$). It can be observed that the ratios of C_2 to C_1 are always more than 1 (Supporting Information Figure S5), indicating that the crosslinked functionality of the co-micelle has a lower value. Therefore, it is suggested that many OP10~AC units in the co-micelle have formed a micro-block embedded on the AM backbone.

Limited Extensibility of the Network Chains

As mentioned above, λ_{infl} is a measure of the extension degree of network chains in the state of limited extension. If the distribu-

tion of crosslinking points and the lengths of network chains were uniform, it could be predicated that λ_{infl} would gradually reduced with increasing R , and the ratios of $\lambda_{infl(HA-OP1Sn)}/\lambda_{infl(HA-OP2Sn)}/\lambda_{infl(HA-OP4Sn)}$ would be $1/2^{1/3}/4^{1/3}$ at a same R . Figure 6(a) shows the dependence of λ_{infl} to R for three series HA-gels. In the lower R range, the anomalous variation of λ_{infl} is corresponding to the complex aggregation of co-micelles. In the higher R range, the increase of λ_{infl} with increasing R can be due to the effect of over introduction of SDS. It is noticed, especially, that λ_{infl} still slightly increases with R in the medium range of R .

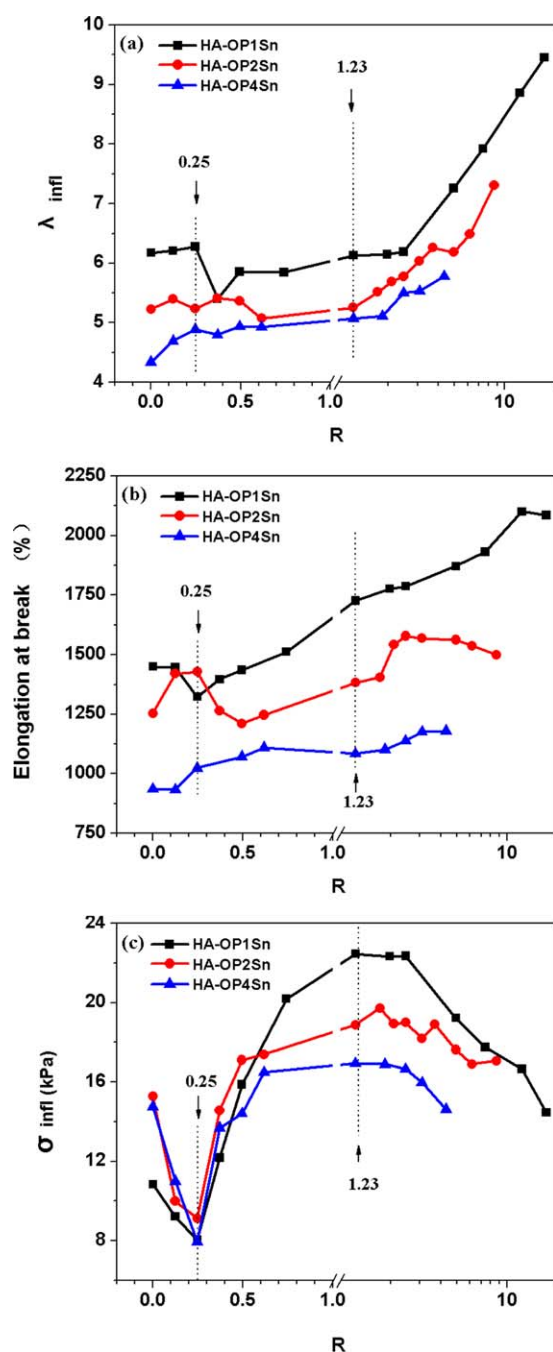


Figure 6. Variations of parameters (a) λ_{infl} , (b) elongations at break, and (c) σ_{infl} versus R for the three series HA-gels. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

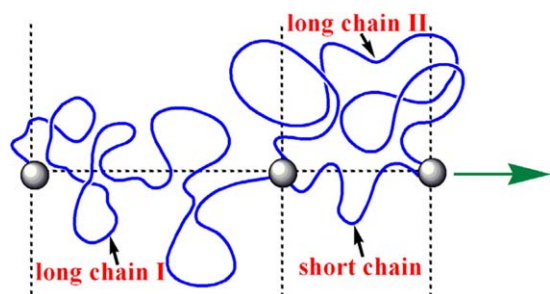


Figure 7. Schematic illustration of two types of long chain in the network. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Furthermore, the ratios between two λ_{infl} s are always larger than the values predicated in the uniform condition (Supporting Information Figure S6). These results indicate that the lengths of network chains are rather heterogenous or polydispersed.

Figure 6(b) shows the variations of elongation at break for three series HA-gels. It is interesting that the variation of elongation at break has a similar tendency with λ_{infl} , especially in the region of R being more than 0.5. Because the stress tends to concentrate in those stretched chains at $\lambda = \lambda_{\text{infl}}$, this comparability implies that the rupture of HA-gels starts from the stress concentration part. Figure 6(c) shows the variation of σ_{infl} with R for three series HA-gels. Because σ is proportional to the elastic force, while the force is mainly given by stretched chains at $\lambda = \lambda_{\text{infl}}$, it can be deduced that σ_{infl} roughly reflects the number of stretched chains. The more the stretched chains at limited extension state, the higher the σ_{infl} as well as the strength of HA-gel. It is reasonable that the variation of σ_{infl} with R presents the "down-up-down" trend similar with the mechanical strength showed in Figure 2(a).

Network Structure Character

Since λ_{infl} and σ_{infl} are, respectively, corresponding to the length and number of stretched chains, we may estimate the structural character of the network via comparing these two parameters. There is a chain length distribution from the shortest one to the longest one, which depends on the R value. For convenience, let us divide the network chains into two parts: shorter chains and longer chains. Figure 7 sketches a simplified image of a local network containing three crosslinking points and three representative chains. When elongation reaches to λ_{infl} , it is obvious that the elastic force will be given mainly by the short chain in the condition of affine deformation. If nonaffine deformation occurs, the middle crosslinking point illustrated in Figure 7 will move over the position predicated by affine model; namely, the extension of the short chain is less than it in the affine deformation, so that the cooperation of the short chain and the long chain I will mainly contribute the elastic force.^{46,47} Moreover, if the short chain moves as a rigid stick, the elastic force will be given mainly by the long chain I. In any case, little contribution to elastic force is given by the long chain II.

Because the λ_{infl} s are large enough [Figure 6(a)], it is expected that the nonaffine deformation must take place and most constrained chains in the original network have released at this elongation. In this case, the amount of elastic force ought to depend on the matching between shorter and longer chains.^{46,47}

In the lower R region, it is expected that large amount of shorter chains will be formed in the HA-gels due to the unstable micelle aggregation mentioned above. However, the λ_{infl} s are not too small except the HA-OP4S0 gel, while σ_{infl} s are relatively low in this region. It maybe imply that the elastic force is contributed mainly by a part of longer chains (type I chains), while the shorter chains in the large size aggregation region are too many to be stretched, and the other part of longer chains belong to type II. The worst mechanical strength and σ_{infl} appear at $R = 0.25$, reflecting that the amount of type I chains have reduced to the lowest value. In the medium R region, the micelles are stable and tend to separate from each other because of charge effect. It is expected that the length of short chains will become longer than it in the lower R region. It can be seen that the λ_{infl} s are relatively low while σ_{infl} s are relatively high in this region, which reveals that the chain length distribution has altered so that proper matching between shorter chains and longer chains has been achieved. The best mechanical strength and σ_{infl} appear around $R = 1.23$, reflecting that the amount of type II chains have reduced to the lowest value. In the higher R region, the λ_{infl} distinctly increases with the increase of R , indicating that the average length of the network chains becomes larger and larger. At the same time, the σ_{infl} tends to be reduced, which is corresponding to the fact that the amount of chains contributing the elastic force or crosslinked density has decreased. It confirms that some co-micelles are attached indeed on a single chain in HA-gels when SDS is over introduced.

Effect of OP10~AC Content

Above results have revealed that the properties of all three series HA-gels present a very similar variation tendency with R , the ratio of SDS to OP10~AC. That is, there is no significant difference in the aggregation behavior of surfactants or the construction of association micro-domains for all three systems so long as their R s keep a same value. However, the network structure of HA-gels should be influenced by the content of OP10~AC even though R is fixed, which should be embodied in the differences of physical parameters showed in above figures. In the condition of fixed R , it is expected that the amount of co-micelle or potential crosslinker will increase with the augment of OP10~AC content, so that the average length of network chains will tend to shorten. The differences of λ_{infl} s and elongations at break have confirmed this argumentation [Figure 6(a,b)]. Because of the unstable aggregation of surfactants, it is acceptable that the exceptive overlap or crossover occurs for HA-OP1Sn and HA-OP2Sn gels in the lower R region. In the medium R region, it can be observed that the σ_{infl} s [Figure 6(c)] and the moduli [Figure 2(b)] become low with the augment of OP10~AC content, and the strengths of HA-OP2Sn gels are near or higher than those of HA-OP1Sn gels while the strengths of HA-OP4Sn gels always have much lower values [Figure 2(a)]. These phenomena imply that the topological structure of intricate network must depend upon the OP10~AC content in the condition of same R . If the topological structures of the three systems were same, in other words, if the chain length merely reduced in an ordinary manner but its distribution character did not change when more OP10~AC was added, the cross-linked density would exponentially increase in the order of HA-OP1Sn to HA-OP4Sn gels, above abnormal phenomena could

not appear. In the basis of contrast of the physical parameters, it seems that too many OP10~AC used, HA-OP4Sn gels for instance, tends to reduce the effective crosslinked density or increase type II chains (Figure 7) in the HA-gels. Maybe 2% is the upper limit of OP10~AC content. Any way the effect of OP10~AC contents at same R is not as regular as the effect of R s at same OP10~AC content, which can be attributed to the intricacies of the topological structures.

It has been found that too few OP10~AC used is also disadvantageous. For instance, the HA-gel with 0.8% OP10~AC content was synthesized in our lab. Except its elongation at break became long, its strength and stability immersed in water became poor. Summarily, in order to get HA-gels with better mechanical properties, it is suggested that OP10~AC content should be in the region between 1 and 2% besides R region between 1 and 2.

CONCLUSIONS

Taking SDS contents as variables, three series HA-gels based on polyacrylamide with different contents of OP10~AC monomers were prepared by free radical micellar copolymerization. It was observed that the properties of all three series HA-gels presented a very similar variation tendency, "down-up-down", with increasing R , the molar ratio of SDS to OP10~AC. When R was approximately lower than 0.5, the tensile strengths and the optical transparencies of HA-gels were relatively poor, and the worst appeared at $R = 0.25$. It was speculated that the unstable aggregation of OP10~AC monomers and such little amount of SDS had taken place. If R was more than 2, some co-micelles contain less OP10~AC units and even append a single chain, so that the effective crosslinked density and the mechanical strength of HA-gel were reduced while the elongation at break became long. In the medium region of R (around 1.23), higher effective crosslinked density and proper match between shorter and longer chains were achieved, so that the HA-gels exhibited better mechanical strengths. In the condition of fixed R , the change of OP10~AC content would remodel the topological structure of network. Introducing more OP10~AC units tended to reduce the effective crosslinked density besides reducing the average length of the network chains. The HA-gels with better properties could be obtained as the OP10~AC content was between 1 and 2% and the R was between 1 and 2.

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