

Influence of Composition and Synthesis Conditions on Microstructure and Properties of Acrylonitrile-Chlorinated Polyethylene-Styrene Copolymer

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ABSTRACT: Acrylonitrile-chlorinated polyethylene-styrene copolymer (ACS resin) was prepared by a suspension polymerization and the structure of the ACS resin was characterized by IR and SEM. The influence of the chlorinated polyethylene (CPE) content, mass ratio of monomer, reaction temperature, and time on the properties of the ACS resin was investigated. With the increase of the CPE content, the thermal stability of the ACS resin decreased, the flexural strength and modulus increased, whereas the heat distortion temperature, tensile modulus, and impact strength first enhanced, and then decreased. The increase of the mass ratio of Styrene (St) to Acrylonitrile (AN) contributed to an improvement in the elongation at break, but played a negative role in the flexural strength and modulus. Increasing the reaction time and temperature could improve the performances of the ACS resin. The morphology of the ACS resin demonstrated the transformation from a brittle fracture to ductile fracture of the ACS resin with various CPE contents and supported the results of the mechanical properties. The ACS resin with good comprehensive performances could be obtained when the content of the CPE was 30%, the mass ratio of St to AN was 2/1, and the reaction condition was 80°C/3 h, 110°C/3 h, and 130°C/2 h. © 2013 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* 130: 2136–2142, 2013

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INTRODUCTION

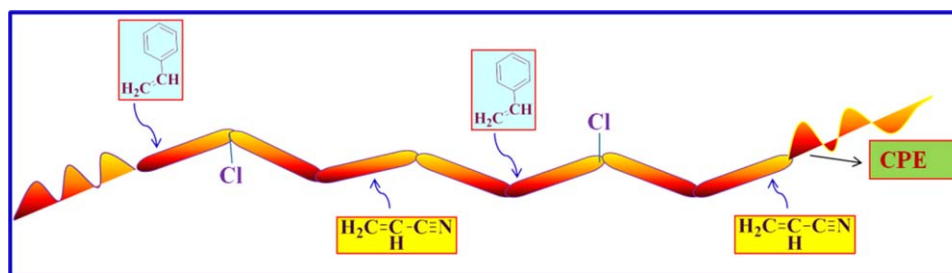
Acrylonitrile–butadiene–styrene (ABS) resin has been extensively applied to the field of engineering materials because of their excellent mechanical, electrical, physical, and chemical properties. ABS consists of butadiene rubber dispersed in a matrix of poly(styrene-*co*-acrylonitrile) (SAN).^{1,2} The ABS resin has the poor weatherability and aging resistance because of unsaturated double bond in the ABS molecule, resulting in limitations to industrial applications. Therefore, much research work has been carried out on improving the weatherability and aging resistance of the ABS resin and the acrylonitrile-chlorinated polyethylene-styrene copolymer (ACS resin) emerges, as the times require.

The ACS resin is usually produced by the blending method of the copolymer poly-Styrene (St)-Acrylonitrile (AN) (PAS) with chlorinated polyethylene (CPE) or graft copolymerization.³ The

general properties of the ACS resin are similar to the ABS, whereas the mechanical, thermal, and electrical properties are generally somewhat superior.⁴ In the ACS resin, the rubbery phase is CPE, which represents an important class of commercial polymers and constitutes the main polymer chains.⁵ The glassy phase is made from St and AN grafted onto the chlorinated polyethylene. The monomers (St and AN) in the ACS could interact with the CPE to improve the compatibility between compositions of the copolymer, thereby enhancing mechanical properties of the resin. The CPE without unsaturated double bond is substituted for the polybutadiene in the ABS resin as an elastomeric component in the ACS resin, leading to good weatherability and resistance to electrostatic deposition of dust of ACS resin. Moreover, the ACS resin has excellent flame resistance, because the CPE contains the chemical bond of carbon–chlorine. The ACS resin is of significant commercial interest.

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Scheme 1. Schematic representation of structure of ACS resin. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Actually, some literatures and patents had already reported properties and applications of the ACS resin.^{6–9} Qiu¹⁰ and Zhu¹¹ studied the preparation and application of flame-resistant ACS resin. Gao⁴ researched the apparent viscosity of modified ACS in the melt with a shear rate at 180°C and Weng¹² improved the mechanical performance of PVC by the addition of the ACS resin as a strengthening agent and studied the influence of preparation method on the property of ACS/Sb₂O₃ composite. Whether the ACS resin is used as a modifying agent or base resin, the structure of the resin itself is a key factor affecting the performance of the resin. However, there is no report on systematically studying the influence of the composition and synthesis conditions on the microstructure and performance of the resin. In this article, we mainly investigated the influence of the CPE content, monomer ratio (St/AN), reaction temperature and reaction time on the microstructure and performances of the ACS resin, including the thermal property, tensile modulus, elongation at break, flexural strength, flexural modulus, heat distortion temperature and the impact strength. The morphology of the ACS resin was also studied by the scanning electron microscopy (SEM).

EXPERIMENTAL

Materials

CPE, with weight-average molecular weight (*M_w*) of 4.70×10^5 and 35% of the weight content of chloride, was supplied by Weifang Chemical Factory (Weifang, China). St and AN were supplied by Fuchen Factory (Tianjin, China). Benzoyl peroxide and azobisisobutyronitrile were donated by Xilong chemical (Shantou, China). Polyvinyl alcohol was purchased from Jingchun reagent (Shanghai, China). Calcium phosphate was gift of Shisihewei chemical (Shanghai, China). 1-Dodecanethiol was purchased from Sinopharm Group Chemical Reagent (Beijing, China). NaNO₂ as a water phase inhibitor agent was kindly given by Yilongxin industrial Trade (Beijing, China). The monomers of AN and St were freshly distilled before use.

Synthesis of ACS Resins

A suspension polymerization for preparing the ACS resin can be described as follows. In a magnetically stirred autoclave, aqueous solution of polyvinyl alcohol (16 g, 5 wt %) was mixed with deionized water (75 mL), NaNO₂ (0.01 g) and Ca₃(PO₄)₂ (1.4 g), and the mixture was stirred for 10 min at room temperature. Then CPE (4 g) and Ca₃(PO₄)₂ (0.6 g) were added into the mixture under stirring to form a suspension system. Subsequently, the mixture of St (12 g), AN (8 g),

azobisisobutyronitrile (0.051 g), and benzoyl peroxide (0.009 g) was dropwise added over 15 min into the CPE suspension system at room temperature. Then the reaction temperature was raised to 40°C after stirring for 10 min. The suspension system was stirred at 40°C for 15 min to fully swell CPE by monomers. Subsequently, the reaction temperature was raised slowly to 80°C at a heating-up rate of about 1°C/min and the heating process spent about 40 min. The reaction was continued under stirring at 80°C for 3 h. Then the reaction temperature was raised to 110°C. After the reaction was continued at 110°C for 3 h, the reaction temperature was raised to 130°C, and the reaction was continued for 2 h. Subsequently, the suspension was cooled down and filtered, and the product was washed with dilute hydrochloric acid and deionized water for three times, respectively, and dried naturally in the air. Similarly, the ACS resin was prepared under different reaction temperature. The experimental operation are the same as the one mentioned above and the only difference is that after the reaction was carried out at 80°C for 3 h, the reaction temperature was raised to 90°C, and the reaction was continued at 90°C for 3 h, then the reaction was conducted at 99°C for 2 h. The schematic representation of structure of the ACS resin was showed in Scheme 1.

Characterization and Measurements

The FTIR spectra were obtained on a Nicolet 50XC spectrometer (Nicolet, USA) and scanned between 500 and 4000 cm⁻¹

The impact-fractured surfaces of the ACS resin were examined by a SEM(S-4700 Hitachi) by using an accelerating voltage of 20.0 kV. The specimens were fractured under cryogenic conditions with liquid nitrogen. Before viewing the observed surfaces were coated with gold to make the surface conductible and not subjected to staining or any other chemical treatments. The observed location lay in the central regions of the surfaces.

Thermal stability of the ACS resins with different CPE contents was analyzed by means of a thermogravimetric analyzer (TGA, a Mettler Toledo TGA/SDTA/851). The measurements were performed using 2–10 mg of the samples in the temperature interval from 25°C to 500°C at a heating rate of 20°C/min in a nitrogen atmosphere.

The tensile behavior of the specimens was determined using an Instron tensile tester (model 1185) at room temperature. Standard dumbbell specimens (ASTM D638) with 25 × 6.0 × 4.0 mm³ neck were used. A cross-head speed of 5 mm/min was

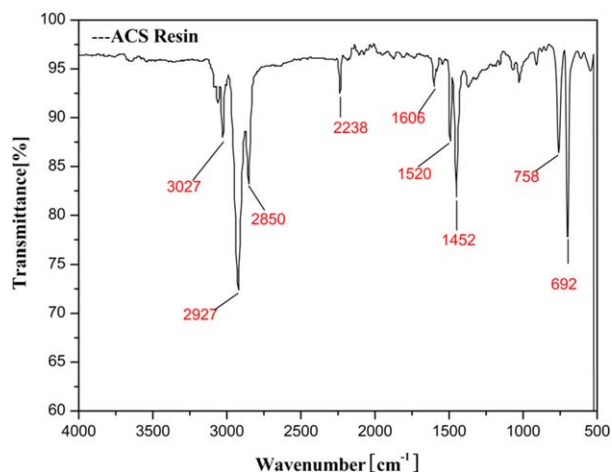


Figure 1. FTIR spectrum for ACS resin. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

used in the tests. Five specimens of each composition were tested and the average values were reported.

Flexural tests were carried out on a universal testing machine (model 1185) at 20°C following GB1042-79. Standard rectangular specimens with $55 \times 6.0 \times 4.0 \text{ mm}^3$ neck were used. At least five samples were tested and the average results were reported.

Notched Izod impact strength tests were conducted with an Izod impact tester (UJ-4, Chengde Machine Factory, China) at room temperature in accordance with ISO 180. Standard dumbbell specimens with $40 \times 3.0 \times 2.0 \text{ mm}^3$ neck were used. Five specimens were tested and the average values were reported.

Vicat/HDT equipment (XRW-300, Chengde Jinjian Testing Machine, China) was used to measure the heat distortion temperature at a heating rate of 120°C/h in accordance with GB1633-2000. Standard rectangular specimens with $10 \times 10 \times 5.0 \text{ mm}^3$ neck were used. The results were taken as an average from measurements of at least five specimens.

RESULTS AND DISCUSSION

Infrared Analysis of ACS Resin

As shown in Figure 1, the absorption peaks of methyl and methylene at about 2927 and 2850 cm^{-1} , respectively, appeared in the curve of ACS resin. The narrow band observed at 3027 cm^{-1} is attributed to the C–H stretching of benzene ring, whereas the bands at 1606 and 1520 cm^{-1} are assigned to C–C stretching of benzene ring. The characteristic absorption peaks of $\text{C}\equiv\text{N}$ and $\text{CH}_2\text{--Cl}$ appeared at 2238 and 1452 cm^{-1} , respectively. The absorption peaks at 758 and 692 cm^{-1} in the fingerprint area are the characteristic absorption for the monosubstituted benzene ring. In addition, the absorption peak of $\text{C}=\text{C}$ at 1670 cm^{-1} did not appear in the curve of the ACS resin, indicating that monomers of St and AN were successfully linked with CPE.

Thermogravimetric Analysis of ACS Resin

The effect of the CPE content in the ACS resin on the heat resistance was investigated and the results are clearly shown in Figure 2. Pure CPE did not exhibit good heat-durability and

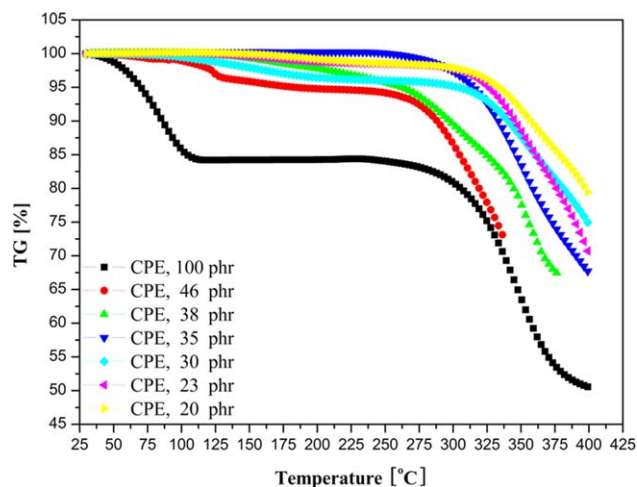


Figure 2. TG curves of the ACS resin with various CPE contents. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

thermal stability, and presented a remarkable two-stage degradation behavior with two turning points at 80°C and 180°C. The significant degradation process at 80°C involved the elimination of side polar groups with the formation of small molecule HCl and of polyene chain, and the CPE lost 16% approximately through this degradation process. When the temperature rose above 180°C, the second degradation process of the CPE appeared that could be attributed to the break of CPE main chain. Moreover, the hydrogen chloride produced by the degradation process probably resulted in the stronger acceleration of the degradation of the CPE. By contrast, The ACS resins with less CPE content showed more excellent heat resistance because of the monomers grafting.^{13–15} The heat resistance of the ACS resins increased and the thermal degradation process gradually converted from two-step degradation process into single-step degradation with the decrease of the CPE content. We speculate that the grafting of the monomers onto the CPE could enhance the stability of the carbon–chlorine bond, thereby prohibiting dehydrochlorination reaction.

Mechanical Properties of ACS Resins

Effect of CPE Content on Mechanical Properties. Figure 3 shows that the tensile modulus varied in an inverted V-shaped curve with the increase of the CPE content. When CPE content increased from 20% to 30%, the tensile modulus increased gradually. But when the CPE content was over 30%, the tensile modulus began to drop with further addition of CPE. In contrast, the elongation at break appeared to reverse the trend. At the beginning, the resin molecular weight augmented with the increase of the CPE content, leading to an increase in the tensile modulus. On the other hand, CPE belongs to an elastomer with the lower strength and modulus, whereas the St and AN monomers belong to a rigid body.¹⁶ Hence, a further increase in the content of CPE in the resin resulted in a decrease in the tensile modulus and an increase in the elongation at break.

It can be seen from Figure 4 that increasing the CPE content in the resin led to a considerable boost in the flexural strength and flexural modulus. The curve showing the dependence of the

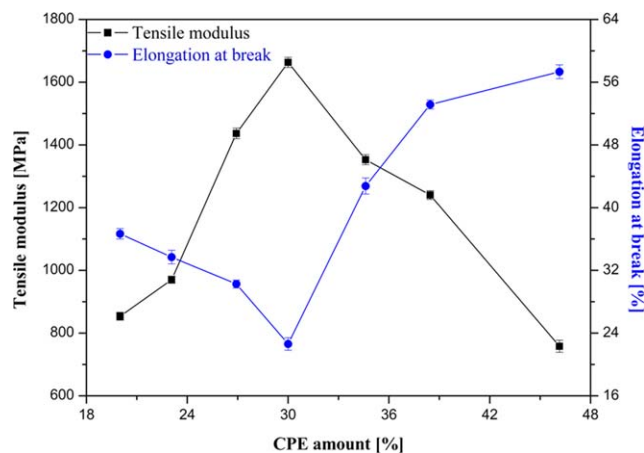


Figure 3. Effect of CPE content on the tensile modulus and the elongation at break. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

flexural strength on the content of CPE was S-shaped approximately. According to the position of inflexions, the curve could be divided into three regions. The first was the brittle region with the low additive content (< 23% CPE), where the flexural strength followed a gradual rise with the increase of the CPE content, and the resin exhibited a low ductility. The second was the brittle–ductile transition region edged by the CPE content from 23% to 30%, where the flexural strength was greatly influenced by the content of the CPE. When the content exceeded 30%, the resin exhibited superhigh toughness, and the influence of the CPE on the flexural strength became stronger. This was the ductile region.

Figure 5 shows that the heat distortion temperature and the impact strength presented a similar trend with the increase of the CPE content. They all first increased and then decreased, and reached a maximum value at the CPE content of about 30%. Notched Izod impact strength is used to characterize toughness. The impact strength of the ACS resin was very low when the CPE content was less than 27%, indicating poor

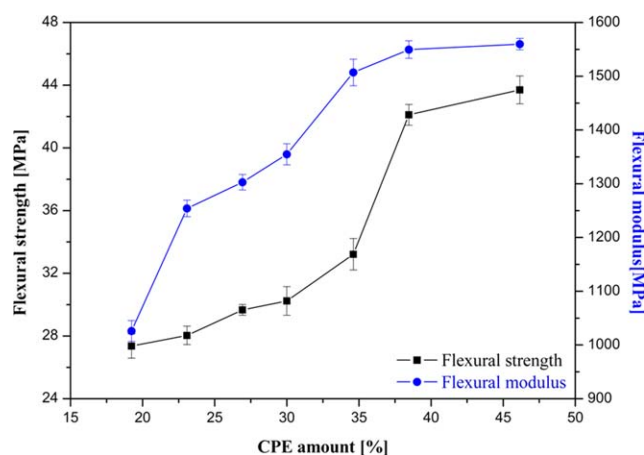


Figure 4. Effect of CPE content on the flexural strength and flexural modulus of ACS resin. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

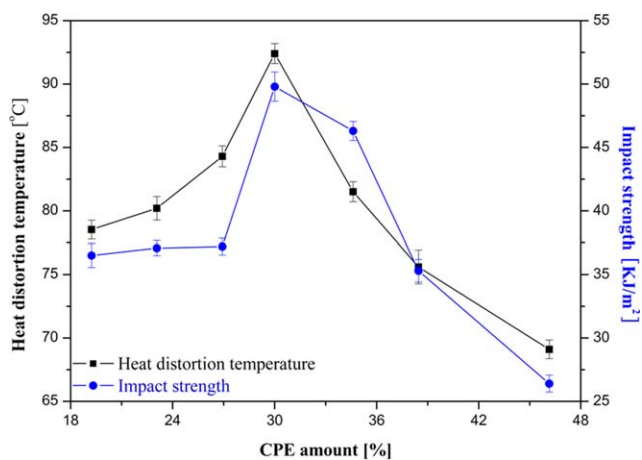


Figure 5. Effect of CPE content on the heat distortion temperature and the impact strength of ACS resin. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

toughness. Increasing amount of the CPE in the ACS resins could also give rise to a continuous network, inducing shear bands to absorb energy,¹⁷ thereby enhancing toughness. However, when the CPE content is too high, whereas the content of the rigid monomers (St and AN) is relatively too low, high elastic characteristics of the CPE would hold a dominant position, and more rubbery CPE dispersed in the rigid and glassy matrix resulting in the decrease in the heat distortion temperature and the impact strength. To summarize, when the CPE content was up to 30%, the increase of the CPE played a negative role in the impact strength and heat distortion temperature.

Effect of Mass Ratio of St to AN on the Mechanical Properties.

The effect of the mass ratio of St to AN on the mechanical properties of the resin is shown in Figure 6–8. Every point exhibited is the average data of the tests. Increasing the mass ratio of St to AN led to an increase in the elongation at break. When the St/AN in the ACS was increased to 3/1, the elongation at break was two-fold higher than that of 1/1. With regard to the tensile modulus, heat distortion temperature, and

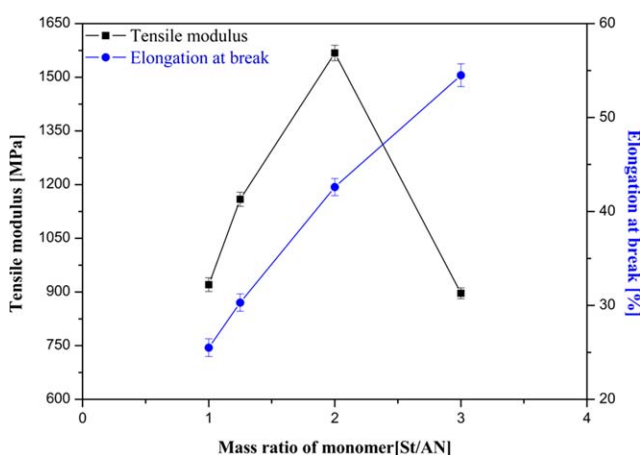


Figure 6. Effect of the mass ratio of St to AN on the tensile modulus and the elongation at break of ACS resin. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

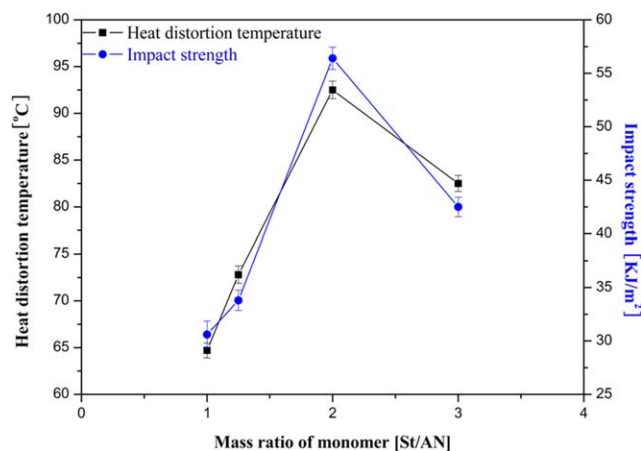


Figure 7. Effect of the mass ratio of St to AN on the heat distortion temperature and the impact strength of ACS resin. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

impact strength, they all first increased, then decreased with the increase of the mass ratio of St to AN and the turning point appeared at the mass ratio of 2/1. The ACS resin was converted into a quite ductile material. Although the impressive improvement in the elongation at break was observed, it was accompanied by a loss in the flexural strength and flexural modulus, as shown in Figure 8. When the mass ratio of St to AN was increased to 3/1, the flexural strength and flexural modulus decreased to 32.0 and 1120 MPa, which were lower than that at the mass ratio of 1/1 (43 and 1580 MPa). It could be attributed to that the AN containing chemical bond of $C\equiv N$ can endow the resin with the strength, heat resistance, and rigidity, whereas the St plays an important role in the improvement of the processing liquidity and luster. Considering all those factors, the optimum mass ratio of St to AN was 2:1.

Effect of Reaction Temperature on the Mechanical Properties.

The influence of the reaction temperature on the mechanical properties of the resin was investigated. From Table I, we can clearly see that the performances of the resin can be improved

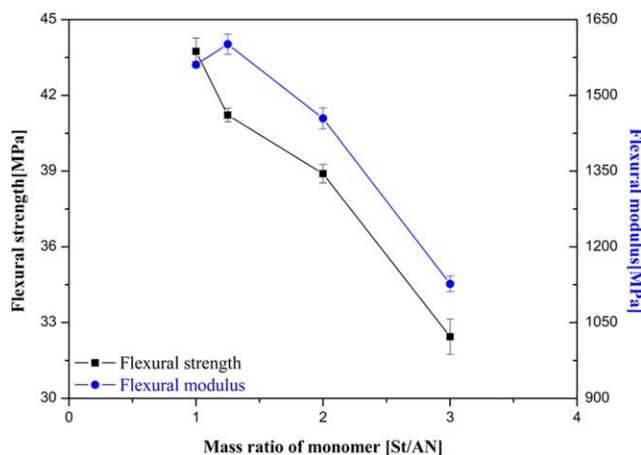


Figure 8. Effect of the mass ratio of St to AN on the flexural strength and flexural modulus of ACS resin. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Table I. Effect of Reaction Temperature on the Mechanical Properties

| Reaction temperature (°C) | 80°C/3 h, 90°C/3 h, 99°C/2 h | 80°C/3 h, 110°C/3 h, 130°C/2 h |
|--------------------------------------|------------------------------------|--------------------------------------|
| Reaction time (h) | 8 | 8 |
| Tensile modulus (MPa) | 1465 | 1663 |
| Elongation at break (%) | 28.9 | 26.4 |
| Flexural strength (MPa) | 30.7 | 38.5 |
| Flexural modulus (MPa) | 1052 | 1460 |
| Impact strength (kJ/m ²) | 26.5 | 56.3 |
| Heat distortion temperature (°C) | 68.5 | 92.6 |

under the high reaction temperature condition, especially the impact strength and heat distortion temperature. The impact strength was increased from 26.5 to 56.3 kJ/m², and the heat distortion temperature was also improved from 68.5°C to 92.6°C. It could be attributed to that monomer molecules had the high reaction activity at a relatively high temperature, which could improve the conversion of the monomers and the molecular weight of the resin,¹⁸ thereby being more easily linked with CPE and benefiting the growth of the molecular weight of the ACS resin. Our experimental results showed that the conversion of the monomers was 76.3% and the weight-average molecular weight of the ACS resin was 6.85×10^5 under low temperature condition (80°C/3 h, 90°C/3 h, and 99°C/2 h), whereas the conversion of the monomers was 96.5% and the weight-average molecular weight of the ACS resin was 8.72×10^5 under high temperature condition (80°C/3 h, 110°C/3 h, and 130°C/2 h), which also demonstrated the high reaction activity of the monomers at a high temperature. To some extent, the resin with high molecular weight has better mechanical properties.

Effect of Reaction Time on the Mechanical Properties.

The effect of the reaction time on the properties of the resin was investigated in detail and the results were shown in Figures 9–11. As shown in Figure 9, the tensile modulus and the impact

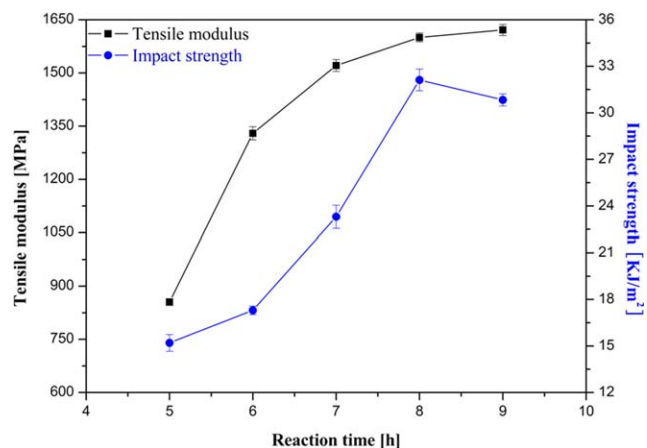


Figure 9. Effect of the reaction time on the tensile modulus and the impact strength of ACS resin. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

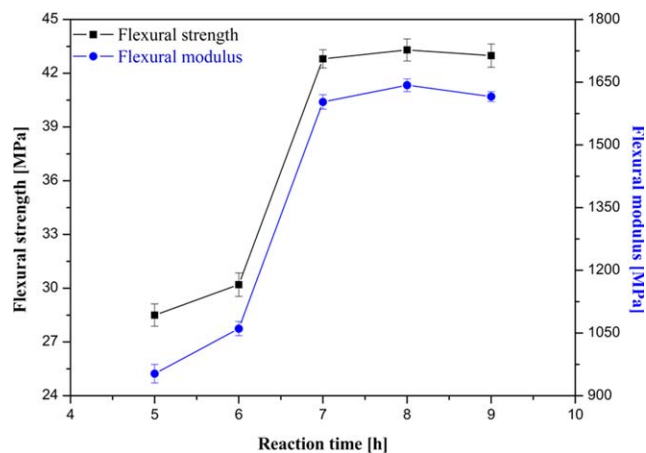


Figure 10. Effect of the reaction time on the flexural strength and flexural modulus of ACS resin. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

strength of the ACS resin were significantly improved by extending the reaction time. The tensile modulus of 1602 MPa and impact strength of 46.5 kJ/m² can be achieved when reaction time was 8 h. Meanwhile, It is further observed from Figure 10 that when the reaction time was extended from 6 to 7 h the flexural strength and flexural modulus were increased significantly, indicating the toughness of the resin was improved and the brittle–ductile transition was exhibited. In addition, Figure 11 shows that the heat distortion temperature was also boosted obviously with the extension of the reaction time. Extending reaction time is favorable for the increase of the grafting degree and the molecular weight of the resin. Therefore, the increase of the reaction time had a positive effect on the mechanical properties of the resin.

Morphology of ACS Resin

To establish the structure–property relation, the morphology of the ACS resin was observed by SEM. The SEM images of the fractured surfaces of these specimens with various CPE contents

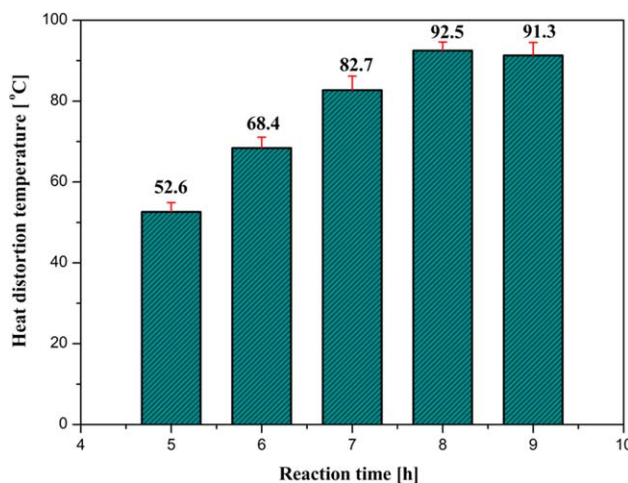


Figure 11. Effect of the reaction time on the heat distortion temperature of ACS resin. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

are shown in Figure 12. For the ACS resin with the CPE of 20% [see Figure 12(a)], a smooth surface without any stress whitening areas was observed, which is typical of a rigid and glassy surface, indicating brittle fracture.¹⁹ A transition from brittle to ductile fracture was observed from Figure 12(b,c,d), when the addition of the CPE was increased from 23% to 30%. The rough and coarse surfaces with domain distortions and areas of whitening reveal the improvement in the toughness.¹⁷ For the ACS resin with the CPE of 30% or more, the surfaces are composed of several separated tiny platelets, and the separated platelets begin to overlap each other at high levels of the CPE to dissipate more energy in the fracture.^{20,21} Increasing the content of the CPE could lead to an increase in the asperity and cloud-shaped platelet surface, which could dissipate more energy in the fracture, and improve the toughness (Figure 5). However, the CPE belongs to the rubbery phase and has no good rigidity

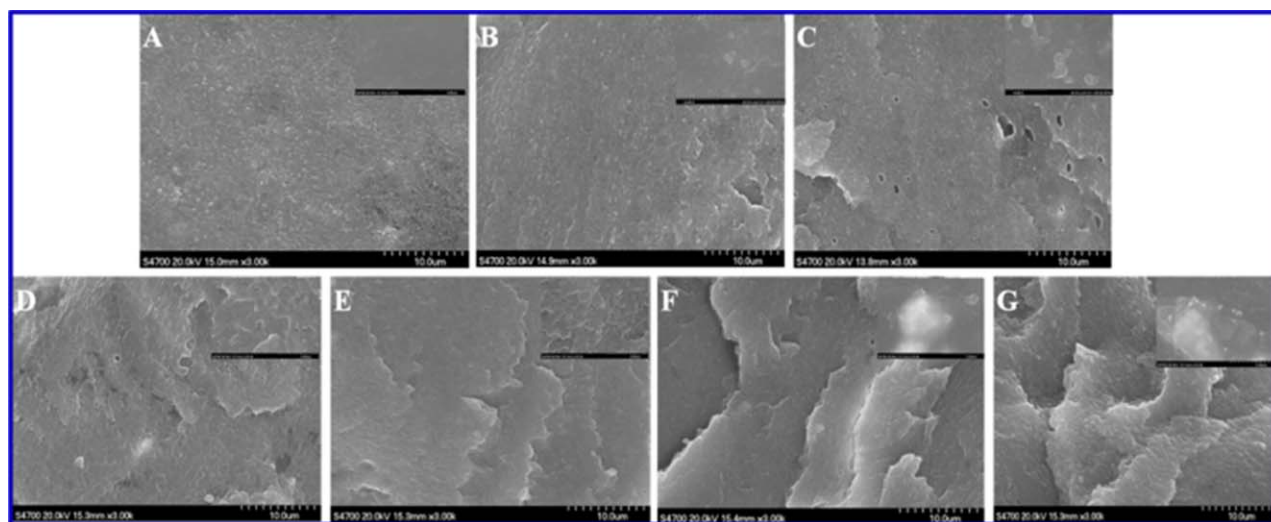


Figure 12. Effect of CPE content on the fractured surface morphology of ACS: (A) 20 phr, (B) 23 phr, (C) 27 phr, (D) 30 phr, (E) 35 phr, (F) 38 phr, and (G) 46 phr. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

in itself, when the CPE content exceeded 30%, some voids and stress whitening areas were apparent on E, F, and G that led to the stretching and ductility of the resin (Figures 3, 4). All the results of the morphology are in good agreement with the mechanical properties.

CONCLUSIONS

The ACS resin was synthesized by a suspension polymerization and its structure was characterized by IR and SEM. The effect of the CPE content, the mass ratio of St to AN, and reaction conditions on the microstructure and properties of the ACS resin was systematically studied. With the increase of the CPE content, the thermal stability of the ACS resin was decreased, whereas the flexural strength and flexural modulus were increased. The heat distortion temperature, the tensile modulus and the impact strength of the ACS resin presented a similar trend with the increase of the CPE content; they all first were enhanced, and then decreased. However, the elongation at break varied in a v-type curve with the increase of the CPE content. Increasing the mass ratio of St to AN could remarkably improve the elongation at break, but played a negative role in the flexural strength and flexural modulus. The tensile modulus, heat distortion temperature, and impact strength all had a maximum value at the mass ratio of 2/1. The mechanical properties of the ACS resin were enhanced by increasing the reaction time and temperature. The morphology of the ACS resin demonstrated the transformation from a brittle fracture to ductile fracture of the ACS resins with various CPE contents and supported the results of the mechanical properties.

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