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Synthesis and Characterization of a Resin/Acrylamide-2-acrylamide-2-methylpropane Sulfonate-Diallyl Dimethyl Ammonium Chloride-*N*-vinyl-2-pyrrolidinone Polymer Microcapsule Gelling Agent for Oil and Gas Field Transformation

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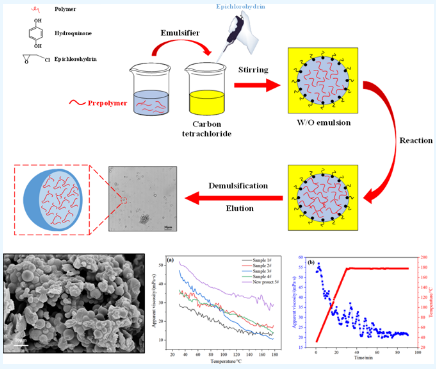

ABSTRACT: Deep carbonate rock oil and gas reservoir is an important support for increasing oil and gas storage and production at present. The environment of ultradeep and ultrahigh-temperature reservoirs has put forward higher technical requirements for reservoir modification acid technology. Moreover, gelling acid is the main acid solution for high-temperature reservoir acidizing transformation, with a temperature resistance of no more than 180 °C, and the gelling agent is one of the key factors restricting its high-temperature resistance performance. In this paper, AM, AMPS, DMDAAC, and NVP were used as monomers, oxidants, and reducing agents to prepare a high-temperature-resistant polymer gel through polymerization. At the same time, microcapsules were prepared by in situ polymerization using epoxy resin as the wall material. The indoor performance evaluation results indicate that the gelling agent is easily soluble in high-concentration acid solution and has good viscosity increasing effect. At 180 °C and 170 s^{-1} shear rate, 0.8% mass fraction of the gelling agent was dissolved in 20% mass fraction of hydrochloric acid. After shearing for 60 min, the viscosity remained at about 22.45 mPa·s, demonstrating good temperature resistance and shear resistance, and its performance was superior to existing commonly used gelling agent products.

1. INTRODUCTION

Acid fracturing is the most important stimulation measure for carbonate reservoirs, and the main factors restricting the effect of acid fracturing are the acid penetration distance and the conductivity of acid corrosion fractures.[1−3] At present, acid fracturing technology is faced with two major technical problems: on the one hand, acid rock reaction speed is fast, and the effective action distance of acid liquid is difficult to be improved, which leads to acid liquid unable to effectively communicate with the far well area, resulting in poor stimulation effect.[4] On the other hand, the actual conductivity of acid-etched fracture is not ideal.[5] Therefore, the gelling agent with excellent performance is added to the acid to improve the viscosity of the acid, effectively limit the convection inside the fluid, and limit the transmission of H^+ to diffusion. At the same time, the added polymer gelling agent forms a colloidal network structure in the acid, which also prevents the activity of H^+, thus effectively delaying the reaction rate of acid and rock.[6−8] At present, gelling agents mainly include synthetic polymers and natural polymers.[9−11] It is noteworthy that natural polymers have the advantages of low cost and wide sources, but their temperature resistance is poor.[12−14] Synthetic polymers can be designed according to the needs of the formation, and finally suitable gelling agent products can be obtained.[15] At the same time, research shows that the synthetic polymer gelling acid has the characteristics of small filtration, low friction, and certain suspension and is easy to inject at a high speed, which is helpful to remove insoluble particles after reaction.[16−18] However, the synthetic gelling agents suitable for high-temperature conditions are still few (i.e., temperature resistance ≥180 °C).

With the rise of formation temperature, the requirements for temperature resistance of gelling agents also increase.[19,20] With an increase in temperature, the molecular movement tends to be intense and the molecular chain structure is destroyed. With the decrease of the pH value, the degree of molecular chain destruction also increases.[9,21−23] Therefore, how to inhibit the degradation of polymer molecular chains at high temperatures has become a key scientific issue. At present, the ways to

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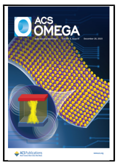

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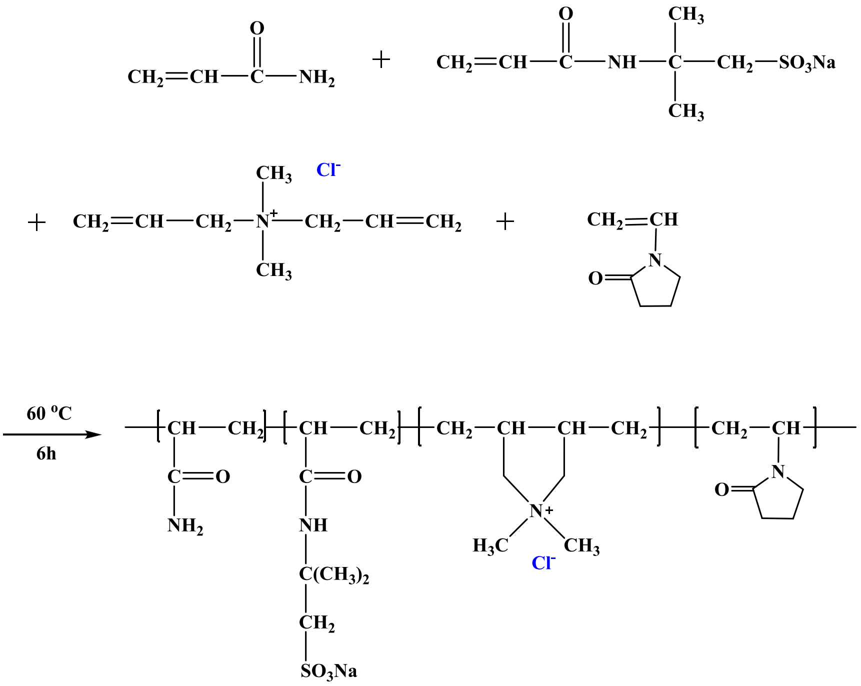

Figure 1. Preparation of the AM−AMPS−DMDAAC−NVP polymer.

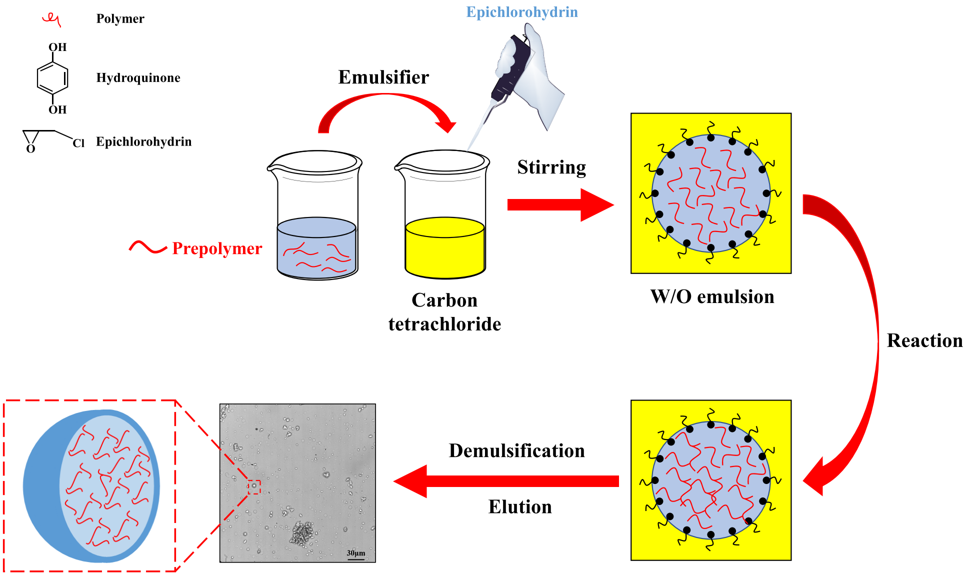

Figure 2. Preparation of the microcapsule gelling agent.

optimize the molecular structure of polymers include[24,25] (i) introducing adsorption groups and hydration groups to improve thermal stability and adsorption, (ii) introducing the rigid side chain or large side chain, such as the benzene ring, into the molecular chain to improve the steric hindrance of the molecular chain and enhance the rigidity of the main chain, and (iii) enhancing the stability of the polymer molecular backbone, such as carbon−carbon bond as the molecular backbone. At the same time, the temperature resistance of the polymer can also be improved by optimizing the molecular configuration of the polymer.[26] The results show that the molecular chain structure of the polymer will be enhanced with the increase of the complexity of the molecular chain branch.[27,28] On the one hand, this paper uses the above two methods to design a network polymer with rigid side chains to improve its temperature resistance from the point of view of the polymer molecule itself. On the other hand, using the protective outer wall to prevent the polymer molecule from contacting with the outside in advance can also inhibit the degradation of the polymer to a certain extent.[29−34]

Therefore, AM, AMPS, DMDAAC, and NVP were used as monomers, oxidants, and reducing agents and a high-temperature-resistant polymer gel was prepared by polymerization. The microcapsule polymer gelling agent with a targeted release function was prepared with epoxy resin as the wrapping material. Then it was characterized by means of SEM, TEM, FTIR, and other test methods. At the same time, the temperature resistance and rheological properties were tested by a high-temperature and high-pressure rheometer.

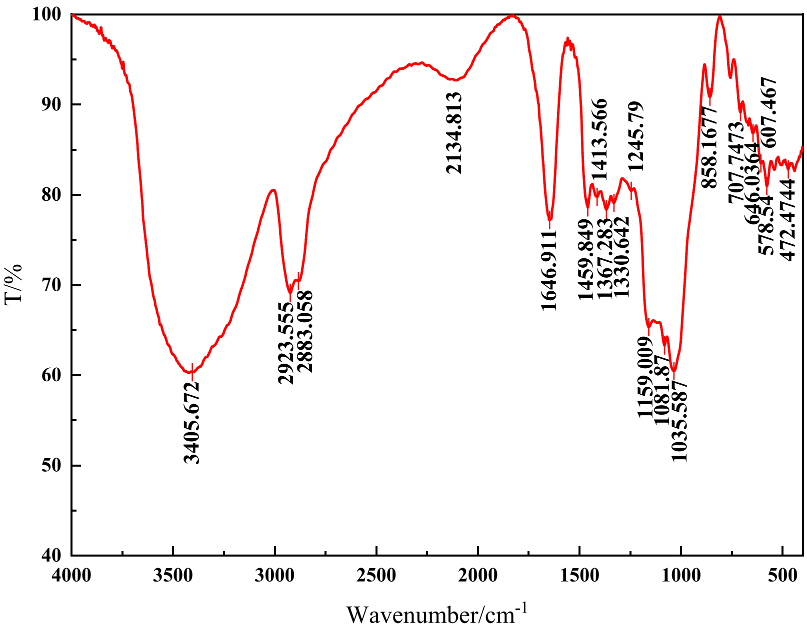

Figure 3. Infrared spectrum of the microcapsule gelling agent.

2. MATERIALS AND METHODS

2.1. Materials. Acrylamide, 2-acrylamide-2-methylpropanesulfonate sodium, diallyl dimethylammonium chloride, and *N*-vinyl-2-pyrrolidone are all analytical reagents provided by Sinopharm Chemical Reagents Co., Ltd. Epoxychloropropane, hydroquinone, ammonium persulfate, and sodium bisulfite are analytical reagents from Aladdin Reagent Co., Ltd.

2.2. Preparation Method. *2.2.1. Preparation of the AM–AMPS–DMDAAC–NVP Polymer.* Polymers were synthesized by the nucleophilic addition reaction, mainly through cross-linking of carbon–carbon double bonds between monomers. The main reaction steps are as follows (Figure 1): 13.5 g of acrylamide, 10.8 g of 2-acrylamide-2-methylpropanesulfonate, 16.65 g of diallyl dimethylammonium chloride, and 3.64 g of *N*-vinyl-2-pyrrolidone are slowly added to 100 mL of deionized water solution, and 0.05 g of ammonium persulfate and 0.05 g of sodium bisulfite are added. The polymer solution is obtained by reaction at 60 °C for 6 h.

2.2.2. Preparation of the Microcapsule Gelling Agent. The microcapsule gelling agent was finally obtained by in situ polymerization using epoxy resin to wrap the polymer solution (Figure 2).

i. 50 mL of prepolymer solution and 11.0 g of hydroquinone were mixed to obtain the dispersion phase. In addition, 18.4 g of epichlorohydrin was dissolved in 300 mL of carbon tetrachloride to obtain the continuous phase.
ii. 6.0 g of Span60 and 2.0 g of OP-10 were added into the continuous phase, and the dispersion phase was added, followed by reaction at 60 °C for 5 h to obtain the microcapsule gelling agent.

2.3. Structural Characterization. The infrared spectrum of the microcapsule gelling agent was measured by the KBr compression method using a NEXUS Fourier transform infrared spectrometer (United States), with a scanning range of 4000−400 cm^{-1}. A JEM 2100 transmission electron microscope was used to observe the morphology of the microcapsule gelling agent. A Hitachi S-4800 scanning electron microscope was used to observe the apparent morphology and particle size of the microcapsule gelling agent. The thermal stability of the microcapsule gelling agent was tested by a TGA thermogravimetric analyzer (Mettler Toledo, Switzerland). The heating rate was 10 °C/min, the atmosphere was nitrogen, and the heating range was 25−800 °C. The particle size distribution of the microcapsule gelling agent was measured by a Bettersize2000 laser particle size analyzer.

2.4. Evaluation of the Temperature Resistance. 3% w/v of microcapsule gelling agent was dispersed in 20% hydrochloric acid solution and heated at different temperatures for 16 h; the mass change of the microcapsule gelling agent before and after heating was recorded. The mass retention rate was calculated using eq 1.

$$\eta = \frac{m_1}{m} \times 100\% \tag{1}$$

where η is the quality retention rate. m_1 is the mass after heating for 16 h. m is the mass before heating.

2.5. Rheological Property Evaluation. The rheological properties of the microencapsulated gelling agent were evaluated with reference to the oil and gas industry standard SY/T 6214-2016 "Thickener for Thickening Acid". Different dosages of the microcapsule gelling agent were added into 20% hydrochloric acid solution, and the mixture was stirred and dispersed at different temperatures for 60 min. The viscosity was measured at 170 s^{-1}, and the effect of the dosage of the microcapsule gelling agent on the viscosity of the acid solution was investigated.

2.6. Acid Solubility Evaluation. The acid solubility of the microencapsulated gelling agent was evaluated with reference to the oil and gas industry standard SY/T 6214-2016 "Thickener for Thickening Acid". 1% microcapsule gelling agent was added into 20% hydrochloric acid solution and stirred at different temperatures for dispersion. The viscosity of the acid solution at 170 s^{-1} was measured at 10 min intervals. When the viscosity started to increase and the viscosity tested at two consecutive time points was the same, the recording was

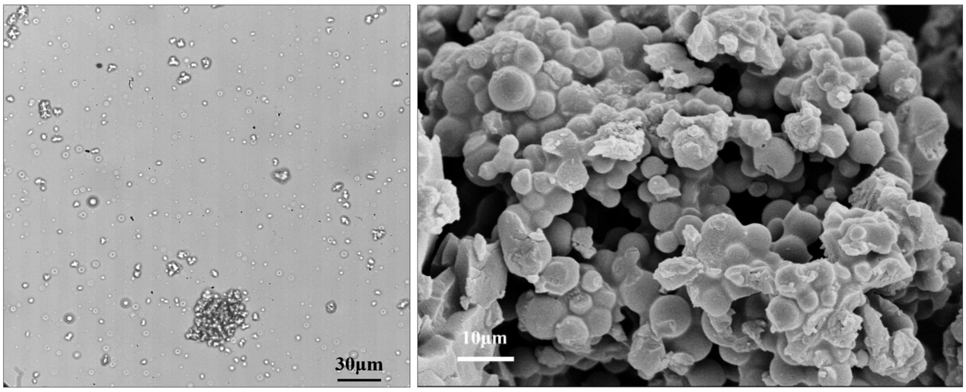

Figure 4. SEM image of the microcapsule gelling agent.

stopped and the corresponding time point of this point was taken as the acid dissolution time.

3. RESULTS AND DISCUSSION

3.1. FTIR Analysis. The infrared spectrum of the microcapsule gelling agent is shown in Figure 3. 3405.672 cm^{-1} is the stretching vibration absorption peak of N−H, 2923.555 cm^{-1}, 2883.058 cm^{-1} is the stretching vibration absorption peak of methyl and methylene, 2134.813 cm^{-1} is the water peak, 1646.911 cm^{-1} is the absorption peak of C=O, 1367.288 cm^{-1} is the characteristic peak of the C−N bond, 1159.009 cm^{-1} is the characteristic absorption peak of the sulfonic group, and 858.1677 cm^{-1} is the characteristic peak of the benzene ring. According to the infrared spectrum analysis, the target monomer was successfully synthesized, and the characteristic peak of the benzene ring can also be seen in the infrared spectrum, indicating that the epoxy resin was successfully prepared and coated with relevant polymers, meeting the expected design requirements.[18]

3.2. SEM Analysis. The apparent morphology and particle size of the microcapsule gelling agent were observed by a Hitachi S-4800 scanning electron microscope. It can be seen from Figure 4 that the particle size distribution of the microcapsule gelling agent is relatively uniform, there is little adhesion between the particles, the small particles have good sphericity, and some particles are agglomerated to form large particles. This may be due to the collision and combination of wall materials during the mixing process. The JMicroVision was used to count the particle size of the microcapsule gelling agent, and the particle size range is 2.85−6.86 μm. The average particle size is 4.05 μm.

3.3. TEM Analysis. Deionized water was used to prepare microcapsule gelling agent suspension with a concentration of 1% w/v. A JY92-IIDN ultrasonic cell pulverizer was used to disperse for 10 min, and a JEM 2100 transmission electron microscope was used to observe the morphology of the microcapsule gelling agent.

It can be seen from Figure 5 that the microcapsule gelling agent is a near-spherical particle and there is an aggregation phenomenon, which is connected into a piece. At the same time, there is a thin layer on the surface of the microcapsule gelling agent, which was the epoxy resin wall material structure, indicating that the epoxy resin successfully coated the polymer.

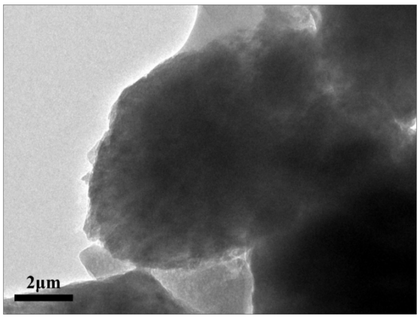

Figure 5. TEM image of microcapsule gelling agent.

3.4. TGA. A TGA 5500 thermogravimetric analyzer (Mettler Toledo, Switzerland) was used to test the thermal stability of the microcapsule gelling agent produced in situ. The heating rate was 10 °C·min^{-1}, the atmosphere was nitrogen, and the heating range was 25−800 °C. The test results are shown in Figure 6.

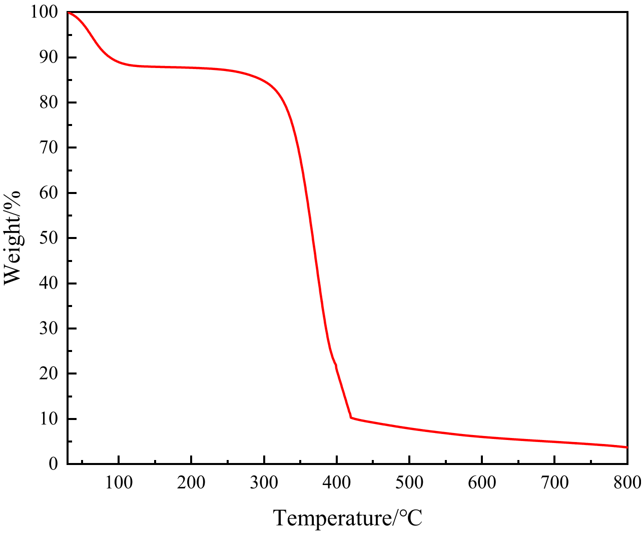

Figure 6. Thermogravimetric curve of the microcapsule gelling agent.

Figure 6 shows the thermogravimetric curve of the microcapsule gelling agent. In general, the thermal decomposition of the microcapsule gelling agent is mainly divided into three stages. The first stage is from room temperature to 100 °C, mainly for the adsorption of water molecules and the removal of crystal water. The weight loss of the microcapsule gelling agent at this stage is 10.54%. The second stage is 100−400 °C, and the corresponding weight loss is mainly the decomposition of the wall material of the microcapsule gelling agent and the pyrolysis of the polymer. The weight loss in this stage is 79.03%. At the same time, it could be seen that at the stage of 100−300 °C, the thermogravimetric curve changes smoothly, with less weight loss. When the temperature is higher than 300 °C, obvious degradation weightlessness begins to occur. The third stage is the degradation of residual carbon. According to the thermogravimetric analysis, the microcapsule gelling agent has relatively excellent thermal stability.[24,26]

3.5. Particle Size Distribution. 0.5% microcapsule gelling agent suspension was prepared with deionized water, and its particle size distribution was measured with a Bettersize2000 laser particle size analyzer after ultrasonic dispersion for 10 min. The result is shown in Figure 7.

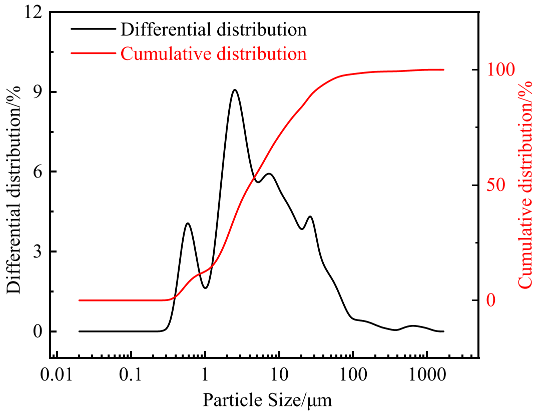

Figure 7. Particle size distribution of the microcapsule gelling agent.

It can be seen from the analysis that the microcapsule gelling agent has a multipeak distribution in deionized water, and the D50 particle size is 4.537 μm. The D90 particle size is 42.28 μm. The specific surface area is 62,500 $\mathrm{cm^2 \cdot g^{-1}}$. At the same time, there are some fine particles in the particle size distribution curve of the microcapsule gelling agent; the particle size is about 500 nm. At the same time, according to Figure 4, this is due to the existence of a large number of nanoscale particles in the suspension, as well as some irregular large particles, which may be small particle aggregates caused by the interaction between the coated polymers. It shows that a microcapsule gelling agent is composed of particles with particle sizes from hundreds of nanometers to several microns.

3.6. Temperature Resistance Evaluation. The quality changes of the microcapsule gelling agent before and after heating at different temperatures are shown in Table 1. It can be seen from the vernier that with the increase in temperature, the mass retention first remains basically unchanged and then decreases rapidly. When the temperature is lower than 150 °C, the mass retention rate is basically 99%. The mass loss after heating may be caused by the error of drying, weighing, and other experimental operations. Therefore, when the temperature is lower than 150 °C, the microcapsule gelling agent is basically not damaged by heat. When the temperature is >150 °C, the mass retention rate decreases rapidly. When the temperature is greater than 180 °C, the mass retention rate is 0%, which means that the microcapsule has complete pyrolysis and released polymer.[19] Therefore, it can be considered that the microcapsule gelling agent could resist the temperature of 180 °C. Its mechanism is that when the external temperature is lower than 180 °C, the microcapsule remains intact; when the external temperature is greater than or equal to 180 °C, the microcapsule is destroyed by heat, releasing polymer to adjust the rheological properties. Finally, the goal of an intelligently targeted release is achieved.

Table 1. Temperature Resistance Evaluation of the Microencapsulated Gelling Agent at Different Temperatures

temperature (°C)	weight before heating (g)	weight after heating (g)	quality retention rate (%)
30	15	14.98	99.87
60		14.99	99.93
90		14.98	99.87
120		14.86	99.07
150		10.75	71.67
160		6.58	43.87
170		2.85	19
180		0	0
190		0	0

3.7. Rheological Property Evaluation with Different Contents. The influence of the dosage of the microcapsule gelling agent on the viscosity of the acid solution at different temperatures is shown in Figure 8. At the same temperature, the viscosity of the acid solution will increase with the increase in the dosage of the gelling agent. This is because the polymer is released into the acid after the epoxy resin shell is damaged by heat. The amide group in the polymer has excellent hydrophilicity and can form hydrogen bonds with water molecules, thus forming a dense space grid structure, thus improving the viscosity of the acid solution.[24] At the same time, the pyrrolidone contained in the polymer is a macromolecular side chain, which can inhibit the aggregation of the polymer by increasing the spatial repulsion between the polymer molecules and improve the viscosity and temperature resistance of the polymer to a certain extent. This effect is called hydrophobic interaction.[25] It is noteworthy that the sulfonate group in the polymer molecular chain also improves the temperature resistance of the polymer.

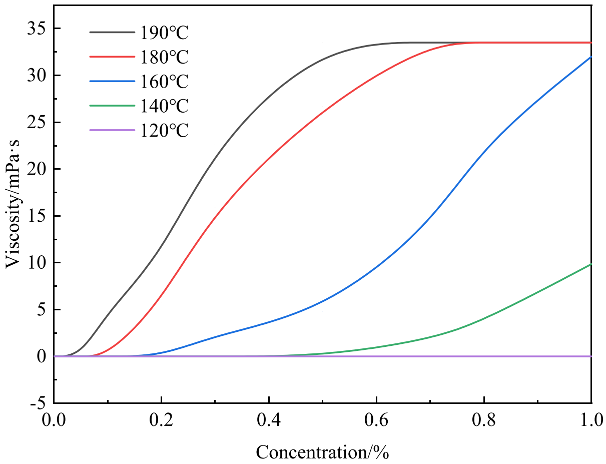

Figure 8. Effect of the dosage of the microencapsulated gelling agent on the acid viscosity at different temperatures.

Therefore, the microcapsule gelling agent has good performance in improving the viscosity of the acid solution. This is not only due to the grid structure formed between the polymer itself and water molecules but also due to the role of other rigid side chains or functional groups in maintaining the stability of the polymer molecular chain structure.[26,28]

3.8. Acid Solubility Evaluation. The change of viscosity of the microcapsule gelling agent with time at different temperatures is shown in Figure 9. When the temperature is lower than 160 °C, the viscosity of the acid solution basically does not change, which is due to a layer of epoxy resin coating on the surface of the microcapsule gelling agent. When the temperature is low, the epoxy resin will not be damaged and the core material cannot be released, so the viscosity of the acid solution cannot be increased. When the temperature is greater than 160 °C, the viscosity of the acid liquor shows a trend of basically unchanged at first, then rapidly increasing, and then reaching equilibrium with the increase of time. At this time, it only takes about 40 min to increase the viscosity of the acid to 42 mPa·s, which means that after the epoxy resin is destroyed, the released core material can be completely dissolved in the acid in a short time and form a space grid structure to improve the viscosity of the acid. At present, the acid dissolution time of the gelling agent is generally required to be less than 120 min in the field construction, so the microcapsule gelling agent meets the requirements of field construction.

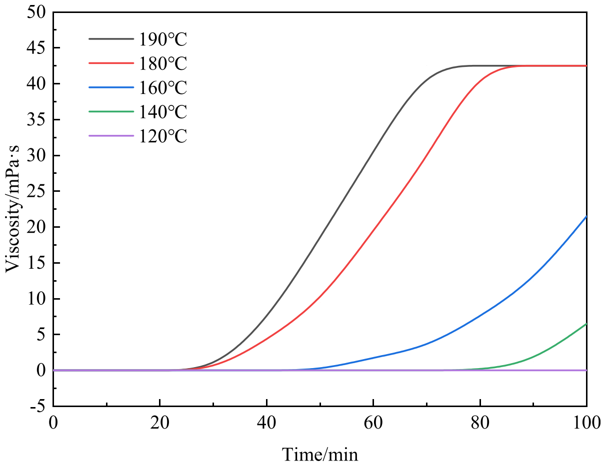

Figure 9. Change of viscosity of the microcapsule gelling agent with time.

3.9. Temperature and Shear Resistance Evaluation. Temperature and shear resistance are key performance indicators of high-temperature-gelled acids. Four types of gelling agent products with good application effects in different blocks were selected for the experiment, including acrylamide cationic polymer, polymer, and ethylene polymer, and compared with the newly developed microcapsule polymer. 0.5 wt % corrosion inhibitor and 0.8 wt % gelling agent were added to 20% HCl to prepare a uniform acid solution. The temperature and shear resistance of different acid solutions were evaluated using a HAAKE Viscotester iQ. The experimental results are shown in Figure 10.

From the results in Figure 7a, it can be seen that the apparent viscosity of different acid solutions gradually decreases with the increase of testing temperature; among them, when the temperature is constant, the apparent viscosity value of the newly developed gelling agent acid solution is relatively high, indicating that the temperature resistance performance of the new product is better than that of existing commonly used gelling agents under the same conditions. When the temperature reaches 180 °C, the viscosity of the new product acid is still close to 30 mPa·s, while the apparent viscosity values of commonly used and commercially available gelling agents on site are both lower than 20 mPa·s. The results indicate that the newly developed microcapsule polymer gelling agent has excellent temperature resistance. From the results in Figure 7b, it can be seen that as the temperature increases, the viscosity of the acid solution initially rapidly decreases. When the temperature rises to 180 °C, the viscosity decreases slowly, and after shearing at 180 °C for 60 min, the apparent viscosity can still reach 21.45 mPa·s. Research has shown that the viscosity of the acid solution prepared by existing polymer-based gelling agents after shearing under the same conditions is less than 15 mPa·s. The self-made microcapsule polymer gelling agent has better temperature and shear resistance than commonly used polymer gelling agents under strong acid concentration conditions.

3.10. Discussion on the Temperature Resistance Mechanism. When the temperature is lower than 160 °C, the viscosity of the acid solution basically does not change, which is due to a layer of epoxy resin coating on the surface of the microcapsule gelling agent. When the temperature is low, the epoxy resin will not be damaged and the core material cannot be released, so the viscosity of the acid solution cannot be increased. When the temperature is greater than 160 °C, the viscosity of the acid liquor shows a trend of basically unchanged at first, and then rapidly increasing, and finally

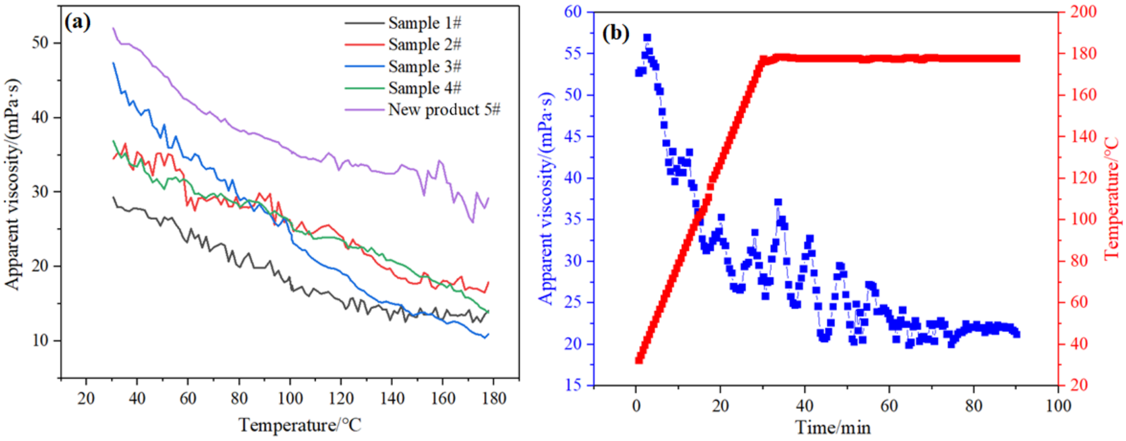

Figure 10. Evaluation of temperature resistance and shear resistance.

reaching equilibrium with the increase of time. After the epoxy resin is destroyed, the released core material can be completely dissolved in the acid in a short time and form a space grid structure to improve the viscosity of the acid. At present, the acid dissolution time of the gelling agent is generally required to be less than 120 min in the field construction, so the microcapsule gelling agent meets the requirements of field construction.

4. CONCLUSIONS

High-temperature and strong acidic conditions will destroy the structure of the polymer, resulting in poor rheological properties. In this article, AM, AMPS, DMDAAC, and NVP were used as monomers, oxidants, and reducing agents and a high-temperature-resistant polymer gel was prepared by polymerization. At the same time, using epoxy resin as the wall material, the microcapsule gelling agent was prepared by in situ polymerization. It has excellent acid solubility and tackiness. At 180 °C and 170 s^{-1} shear rate, 0.8% mass fraction of the gelling agent is dissolved in 20% mass fraction of hydrochloric acid, and the viscosity is maintained at about 21.45 mPa·s after shearing for 60 min. It has good temperature and shear resistance. The excellent temperature resistance of the microencapsulated gelling agent was due to the excellent temperature resistance of epoxy resin, which protects the core material from the influence of H^+. When the microcapsule gelling agent reaches the target temperature formation, the epoxy resin degrades the targeted release core material. At this time, the polymer is connected with acid and other polymer materials through hydrogen bonds so as to improve the viscosity of acid and achieve the goal of targeted release and temperature resistance.

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Author Contributions

All authors have accepted responsibility for the entire content of this manuscript and approved its submission.

Notes

The authors declare no competing financial interest.

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