


Article

Low-Cost H-Grade Polyacrylamide Gel with High-Temperature Resistance

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Abstract: This paper presents a low-cost, high-temperature-resistant and high-strength polyacrylamide gel system formed by secondary cross-linking. The gel system (named JM186) used phenolic resin and organic zirconium as cross-linking agents, and the performance of the gel system was systematically evaluated under high temperature. The gel properties studied include: gel formation time, gel strength, thermal stability, sand-filled pipe sealing efficiency, and its microstructure. The concentration of polyacrylamide in JM186 gel system was as low as 0.3%, which can control the gelling time in a range of 1–9 h by adjusting the ratio of two cross-linking agents. It can resist temperature up to 120 °C without dehydration, and its highest gel strength can reach H grade. The modulus of elasticity (G') and viscosity (G'') can reach 32.33 Pa and 3.25 Pa, respectively. DSC (differential scanning calorimetry) test indicated that the temperature of structural failure for this composite gel is 310.5 °C. The average sealing efficiency of the gel is 96.03% in sand-filled pipes. Finally, the gel microstructure was observed by cryo-scanning electron microscopy (Cryo-SEM). It was found that the gel system by secondary cross-linking has a dense and thickened network structure compared with the single cross-linker gel system. The gel is cross-linked by both the coordination bond and covalent bond, and the two cross-linking agents have a synergistic effect. This is the reason why the secondary cross-link gel system is better than the single-cross-linker gel system.

Keywords: secondary cross-linking; high-temperature-resistant; high-strength polyacrylamide gel; low-cost



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1. Introduction

Most old oil fields are facing high water content and low oil recovery. There are high permeability and preferential channels in heterogeneous reservoirs. Therefore, water returns to the surface through these preferential channels during water injection processes, which can result in high water content, low recovery, and high residual oil saturation [1]. Large amounts of ineffective water injection can waste water resources, pollute groundwater, aggravate equipment aging, and bring a series of problems [2]. Therefore, it is a challenge to reduce the water content and increase the oil recovery rate [3]. Improving wave efficiency is the key to improving oil recovery [4,5], and the use of gel to seal high-permeability channels is a common method to improve wave efficiency in oil fields. Polyacrylamide gel is a commonly used polymer in oilfield chemistry and also has good performance in water management and plugging [6]. However, the traditional polyacrylamide gel is insufficient in thermostability and difficult to apply in high-temperature reservoirs. Although there are several high-temperature-resistant polyacrylamide weak gel systems, many shortcomings still exist. For example, high-temperature-resistant gel systems commonly use high concentrations (0.5–3.0 wt%) of polyacrylamide. When the concentration of polymer is below 0.5 wt%, the addition of expensive nanoparticles or other reinforcing agents is required to enhance the gel strength [3,7–10]. Therefore, it

is urgent to develop a high-temperature-resistant, high-strength, and low-concentration polyacrylamide gel system for the EOR of high-temperature reservoirs.

Phenolic resin is a common organic cross-linker. Fu et al. developed a gel system using a modified phenolic resin with good salt resistance up to 8000 mg/L of salinity and good thermal stability up to 200 °C [11]. For high-temperature reservoirs (150 °C), Zhu et al. developed a stable ternary polymer gel using phenolic resin as a cross-linking agent, in which the polymer molecular weight was 3 to 5 million [12]. Wu et al. synthesized a water-soluble phenolic resin and studied its plugging capacity under three synthesized conditions [13]. The polyacrylamide and phenolic resin can cross-link under heating conditions to form a gel with good temperature resistance, which has been widely used for profile control for reservoir temperatures higher than 60 °C.

Inorganic cross-linking agents are also used to form polyacrylamide gels. Dai [14] et al. studied the influence of various factors on the gelling time and gel strength of zirconium cross-linked gel, such as pH of PAM solution, dosage of $ZrOCl_2$, mass fraction of PAM, temperature, degree of hydrolysis, molecular weight, and salinity. Ma [15] et al. demonstrated that when the concentration of polymer and cross-linker, temperature, and/or salinity increase, the gelling time and strength of the zirconium cross-linked gel increase. Zhang [16] et al. developed a polyacrylamide gel formed by Cr(III) and phenolic resin, which did not undergo any dehydration for 120 days under 140 °C and can reach a strength of H grade. Similarly, Yao et al. obtained an H-grade gel by Cr(III) and phenolic resin that can resist 95 °C at very low polymer concentration [17]. In addition, Liu et al. investigated silicone resin as organic–inorganic cross-linkers, and the gel system was stable in 90 days at 140 °C [18].

This paper presents a low-polymer-concentration, high-temperature-resistant, high-strength, more environmentally friendly, and secondary cross-linked polyacrylamide gel. The phenolic resin and an organozirconium cross-linker are used for the cross-linking process. The gelling mechanism of secondary cross-linking was studied by rheological test, differential scanning calorimetry (DSC), and cryo-scanning electron microscopy (Cryo-SEM).

2. Experimental Part

2.1. Experimental Materials

Water-soluble polyacrylamide was used. It has an average molecular weight 2.5×10^7 g/mol and 30~40% degree of hydrolysis, and it was purchased from Aisen (China) Flocculants Co. Organic zirconium cross-linker and phenolic resin cross-linker were self-made in the lab, and the specific preparation steps are shown in Section 2.2. Formaldehyde solution (36%), phenol (analytical purity), thiourea (analytical purity), zirconium oxychloride (analytical purity), and propanetriol (analytical purity) were purchased from Shanghai Aladdin Biochemical Technology Co (No. 809, Chuhua Branch Road, Fengxian District, Shanghai). NaOH (purity $\geq 96\%$) was purchased from Xilong Science Co (No. 1-3, West Long Middle Street, Chaoshan Road, Shantou City, China).

2.2. Synthesis of Cross-Linkers

The synthetic method of phenolic resin cross-linker was as follows: (1) Phenol, NaOH solid particles, and water were added to the flask in pre-designed proportion. The temperature was set at 60 °C for 50 min. (2) Next, additional NaOH was added, and the solution was stirred at 70 °C for 20 min. (3) Then, formaldehyde solution was added, and the solution was stirred at 90 °C for 30 min. (4) Finally, a brown-red water-soluble phenolic resin was obtained after cooling.

The synthetic method of organic zirconium cross-linker was as follows: (1) Zirconium oxychloride, propanetriol, and tap water were added to the flask in pre-designed proportion. (2) Then, the solution was stirred at 60 °C for 5 h. (3) After cooling, a transparent organic zirconium cross-linker was obtained. It is worth noting that this part of the experiment used

a balance with an accuracy of ± 0.0001 g. The purpose of this was to ensure the accuracy and repeatability of the synthetic experiments.

2.3. Preparation of Gel Solution

- (1) A total of 1.5 g polyacrylamide was dissolved into 500 mL tap water. After strong stirring, a 0.3 wt% polyacrylamide solution was gained. This solution was hydrated under room temperature and placed away from light for 24 h.
- (2) A total of 50 mL of the solution was taken and added to 100 mL sample bottle. Then, 0.1 wt% of antioxidant (thiourea) and different concentrations of cross-linker were added and stirred for 25 min to obtain homogeneous gel solution. This stirring time and antioxidant dosage were determined according to the experiments.
- (3) The gel stood for 30 min to remove air bubbles. Then, the sample bottle was put into the oven at 120 °C to form the gel. This part of the experiments also used a balance with an accuracy of ± 0.0001 g, as in the previous section, so that the composition of the gel solution could be precisely controlled.

2.4. Measurement of Gel-Forming Time and Dosage Optimization of Cross-Linking Agent

The sample bottles were placed in the oven at 120 °C and then taken out at different times to observe the gel formation. The gel-forming strength is referred to using the GSC strength code method proposed by Sydansk [19]. When the gel strength reaches maximum and does not change, this time is regarded as the gel-forming time [20]. The gel strengths at each level are shown in Figure 1. The different levels of gel are described in Table 1.

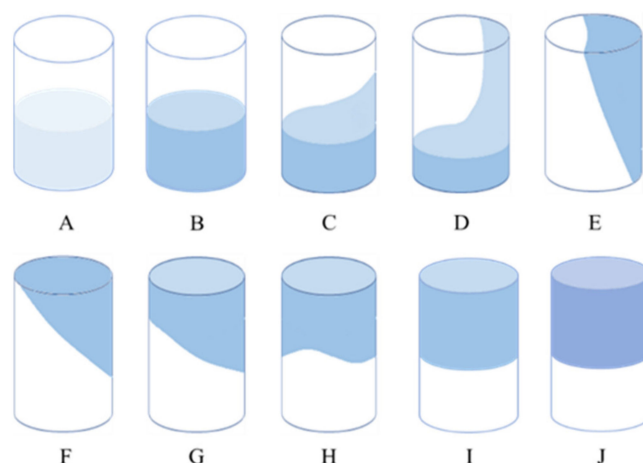


Figure 1. Schematic diagram of GSC strength code.

Table 1. GSC classification [16].

Strength Code	Gel Name	Corresponding Intensity Description
A	Non-detecting gel	The viscosity of the system is equivalent to the viscosity of the polymer, and the formation of gel cannot be observed with the naked eye
B	High flow gel	The viscosity of the gel system is slightly higher than the viscosity of the polymer
C	Flowable gel	The viscosity of the gel system is slightly higher than the viscosity of the polymer
D	Medium flow gel	When the glass bottle is turned over, a small amount of gel (mass fraction <15%) can not flow to the other end, often in the shape of a tongue
E	Hardly flowing gel	When the glass bottle is turned over, a few gels can slowly flow to the other end, and most of them (mass fraction > 15%) are not fluid
F	High deformation non-flowing gel	The gel cannot flow to the mouth of the bottle when the glass bottle is turned over
G	Medium deformation non-flowing gel	When turned over, it can only flow to the middle of the glass bottle
H	Slightly deformed non-flowing gel	When flipped, only the surface of the gel is deformed
I	Rigid gel	When turned over, the surface of the gel does not deform
J	Ringing gel	When shaking the glass bottle, you can feel the mechanical vibration like a tuning fork

A rheometer (HAAKE RS600) was used to evaluate the gel strength by comparing the G' and G'' of the sample at a frequency of 0.1 Hz [21]. The rotor type chosen for the experiment was Z41Ti. This part of the experiment was repeated 3–5 times according to the stability of the data. The average value was selected as the final experimental result, and the error bar was calculated and is shown on the pictures.

2.5. Gel Temperature Resistance Test

The gel system was tested for phase transition temperature using a simultaneous thermal analyzer (METTLER TOLEDO TGA/DSC 1). A small amount of sample was weighed and warmed from 30 °C to 400 °C in an air atmosphere, where the scanning frequency was 5 °C/min, and the temperature resistance of the gel was assessed by comparing the enthalpy peaks shown in the data [22,23]. The DSC test was repeated until the curves were similar. Then, one of the curves was selected and placed in the article.

2.6. Blocking Effect Test

The sand-filled pipe was selected to test the blocking effect of the gel system. Amounts of 40~50 mesh and 100~120 mesh quartz sands were mixed in a volume ratio of 1:4 and loaded into the sand-filling pipe (sand-filling pipe diameter $d = 25$ mm, effective length $L = 100$ mm). Then, it was connected to the displacement pump, as shown in Figure 2. Water was injected at a certain flow rate, and the permeability k_1 was recorded and calculated when the inlet and outlet pressure was stable at a constant value. Then, the gel solution was pumped in until a stable gel solution came out of the outlet. The sand-filled pipe was put into the oven (120 °C) to promote the formation of the gel. Again, water was injected into the sand-filled pipe to obtain the permeability k_2 when the pressure was stable. The blocking efficiency was evaluated by difference between k_1 and k_2 according to Equation (1).

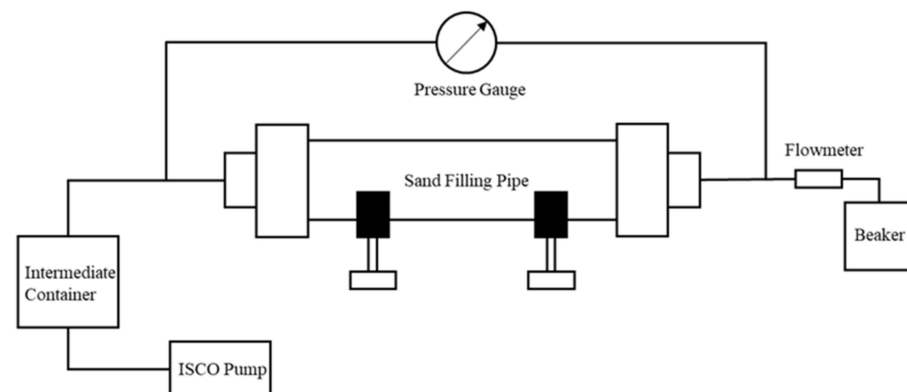


Figure 2. Sand-filled pipe displacement device.

The blocking efficiency in the experiment is calculated as follows.

$$\eta = \frac{k_1 - k_2}{k_1} = \frac{\Delta P_2 - \Delta P_1}{\Delta P_2} \times 100\% \quad (1)$$

2.7. Microstructure of the Gel

The microstructure of the gels was observed using a cryo-scanning electron microscope (SU3500 Hitachi, Gui Gu Technology Development Co., Ltd., Shanghai, China). A small piece of sample was stuck to a sample table with conductive carbon adhesive. Then, the sample stable was quick-frozen in liquid nitrogen slush for 30 s. It was transferred to the sample preparation chamber under vacuum using a cryogenic cryo-preparation transfer system and sublimated at −90 °C for 10 min. Then, the sample was sputtered gold at a current of 10 mA for 60 s. The sample was sent to the SEM sample chamber for observation,

the temperature was set as $-140\text{ }^{\circ}\text{C}$, and the accelerating voltage was 5 kv. Compared with conventional EM, cryo-electron microscopy can observe the 3D structure of the gel sample rather than the collapsed structures.

3. Results and Analysis

3.1. Optimal Concentration of Cross-Linking Agent and Gel-Forming Time

3.1.1. Phenolic Resin Cross-linker

Phenolic resin is a commonly used organic cross-linking agent. The hydroxyl group on phenolic resin and the amide group on polyacrylamide form covalent bonds through dehydration condensation reaction and grow into a 3D network. This 3D network structure helps the system to control the water molecules effectively and keep the gel stable at relatively high temperatures [17].

In order to minimize the gel cost, this paper uses 0.3 wt% polyacrylamide concentration and 0.1 wt% antioxidant. Phenolic resin cross-linker concentration should be between 0.1–0.7% in order to obtain the best gel. The experimental settings and results are shown in Table 2.

Table 2. The experimental settings and results of the cross-linking reactions.

Polyacrylamide Concentration/wt%	Antioxidant Concentration/wt%	Cross-Linker Concentration/%	Gel Grade	Gel-Forming Time/h	Dehydration
0.3	0.1	0.1	B	12	No
		0.2	B	10	
		0.3	C	9	
		0.4	C	8	
		0.5	C	8	
		0.6	D	7	Yes
		0.7	D	6	

With the increase in cross-linker concentration, the gel grade increased from B to D. While continuing to increase dosage of cross-linker to 0.7%, the phenomenon of dehydration appeared. The gel-forming time gradually decreased from 12 h to 6 h with the increasing of cross-linker dosage. The dehydration of the gel originated from excessive cross-linking [18]. The shorter gel-forming time with the increase in cross-linking agents means a greater probability that the hydroxyl group in the phenolic resin was bonded to the amide group in the polyacrylamide, and thus, a faster cross-linking rate was obtained.

However, compared with inorganic cross-linkers, it was difficult for an organic cross-linker to form a high-strength gel, such as an H-grade gel. Therefore, in the next section, an appropriate amount of inorganic cross-linker for secondary cross-linking was investigated to improve the gel strength.

3.1.2. Organic Zirconium Secondary Cross-linker

Organic zirconium is a widely used inorganic cross-linking agent. Organic zirconium can hydrolyze and release Zr^{4+} ions, which cross-link with COO^- in polyacrylamide and form multinuclear hydroxyl-bridged complex compound [24]. Therefore, it also makes polyacrylamide form a 3D network structure, namely a gel. The concentration of organic zirconium cross-linker used in this study was 1.0–2.0%. The experimental parameters and results are shown in Table 3.

With the increase in cross-linker concentration, the gel-forming grade gradually increased from grade F to grade I, but the phenomenon of dehydration also appeared. The gel-forming time can be controlled from 1 h–9 h by adjusting the concentration of the two cross-linkers, as shown in Figure 3. When organic zirconium was used alone, the gel-forming time was usually 1–4 h. After adding phenolic resin cross-linker, the gel-forming time increased slightly. It is proposed that the spatial site resistance effect of the benzene ring in phenolic resin prevents organic zirconium from approaching polyacrylamide, which makes the gel-forming time increase. After comprehensive consideration of gel strength,

dehydration, and gel-forming time, the optimal formulation was selected as: 0.3 wt% polyacrylamide +0.1 wt% antioxidant +0.3 wt% phenolic resin +1.6% organic zirconium. At the optimal formulation, an H grade of gel was achieved without dehydration, and the gel-forming time was 4 h. This formulation was named JM186 and was used for further study. Samples #1 and #2 formed with a single cross-linker were used as contrasts. The composition of the three samples used for gel performance evaluation in this paper is shown in Table 4.

Table 3. The experimental settings and results of the secondary cross-linking reactions.

Polyacrylamide Concentration/wt%	Antioxidant Concentration/wt%	Phenolic Resin Concentration/%	Organic Zirconium Concentration/%	Gel Grade	Dehydration
0.3	0.1	0.1	1.0	F	No
			1.3	F	
			1.6	G	
			2.0	G	
		0.3	1.0	F	No
			1.3	G	
			1.6	H	
			2.0	I	
		0.5	1.0	F	No
			1.3	G	
			1.6	H	
			2.0	I	
		0.7	1.0	F	Yes
			1.3	G	
			1.6	H	
			2.0	I	

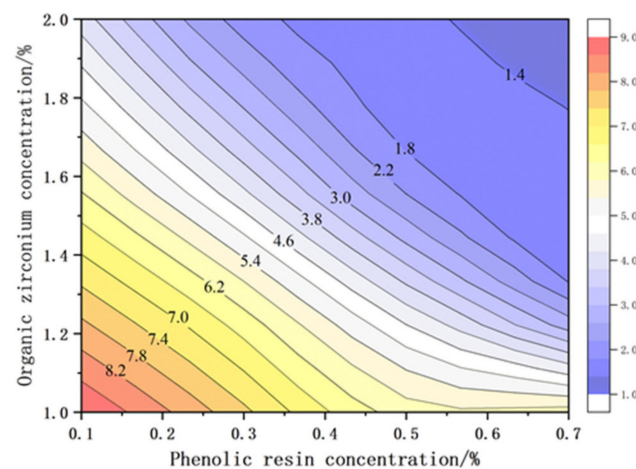


Figure 3. Gel formation time of the secondary cross-linking reactions.

Table 4. The recipe compositions of the samples JM186, #1, and #2.

No	Polyacrylamide Concentration/wt%	Antioxidant Concentration/wt%	Phenolic Resin Concentration/%	Organic Zirconium Concentration/%
JM186			0.3	1.6
#1	0.3	0.1	0	1.6
#2			0.3	0

3.2. Analysis of Rheological Results

The strength and toughness of a gel can be evaluated by its viscoelastic properties [25]. The viscoelasticity of gels includes two factors: the elastic modulus (G') and the viscous modulus (G''). G' represents the energy stored in the gel during deformation, and G'' represents the viscosity of the gel. The larger the value of the two, the greater the gel

strength, which is conducive to water shutoff operations [26]. The G' and G'' of samples JM186, #1, and #2 and the inversion photos of the gel in bottles are shown in Figures 4 and 5, respectively. It should be noted that the G' and G'' data are the average of three parallel tests.

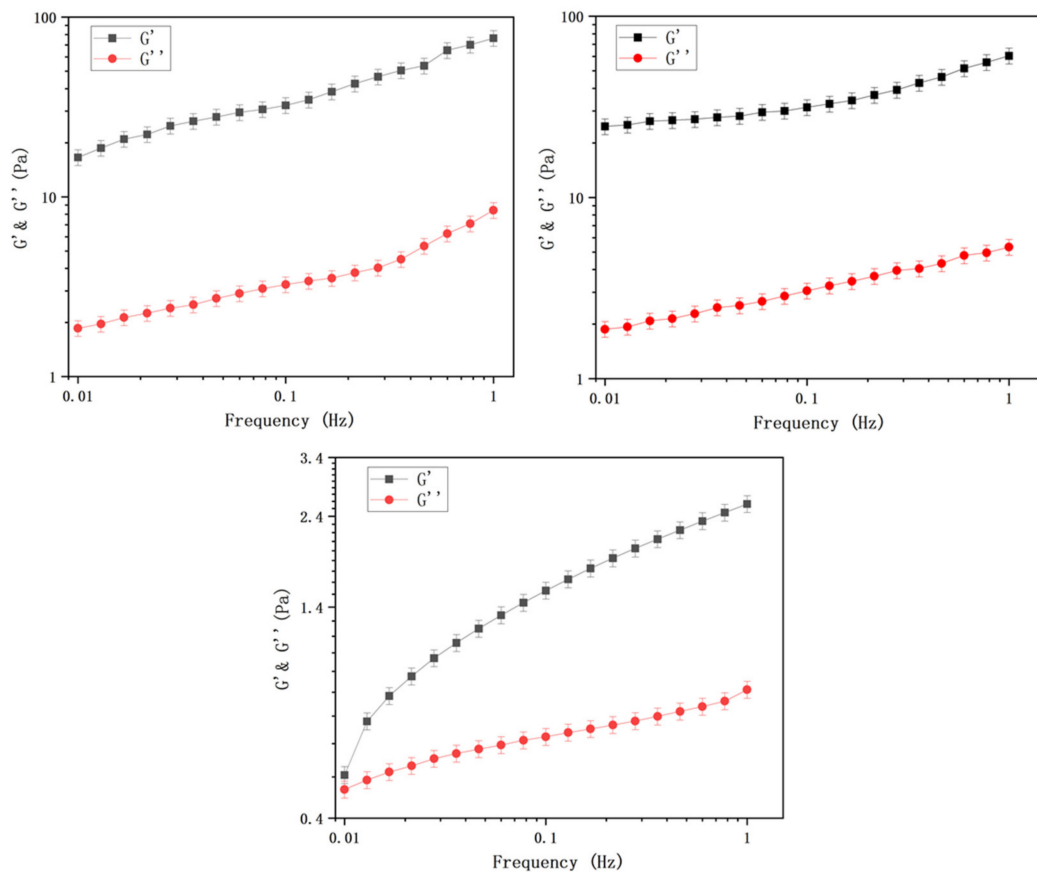


Figure 4. Plots of G' and G'' data for sample JM186, #1, and #2.



Figure 5. The pictures of inverted bottles containing JM186 (left), #1 (middle), and #2 (right).

From Figure 4, G'' and G' of JM186 reach 32.33 Pa and 3.25 Pa at 0.1 Hz, respectively. G'' and G' for sample #1 are only 31.45 Pa and 3.06 Pa at 0.1 Hz, respectively. For sample #2, G'' is 1.544 Pa and G' is 0.648 Pa at 0.1 Hz. The G' and G'' of JM186 were similar to those of #1 and much larger than those of #2. This indicates that the organic zirconium cross-linker made the G' and G'' of the secondary cross-linked gel system much better compared with that of the phenolic resin gel system.

3.3. Thermal Stability of Gel

DSC analyzes the enthalpy change of the sample according to the peak shape of the thermal curve, which allows the analysis of thermal stability of the gel system [27]. DSC test was performed on three samples to analyze the temperature resistance of the secondary cross-linking gel system, and the test results are shown in Figure 6. The thermal curve of the sample JM186 gradually decreased between 35 °C and 310.5 °C due to a small amount of water evaporation and other physical changes [28]. A clear exothermic peak started to appear at 310.5 °C, which can indicate that the structure of the gel starts to break down, until the gel structure was completely destroyed at 340.4 °C. Therefore, the maximum tolerated temperature of this gel system was no more than 310.5 °C. The two samples #1 and #2 have similar DSC curves, and their temperatures of structure destruction were 193.9 and 207.2 °C, respectively. Based on where the exothermic peak started, samples #1 and #2 should be used under 144.6 and 146.7 °C to ensure the structural integrity of the gel. In a word, the temperature resistance of the gel system had been greatly improved by the secondary cross-linking.

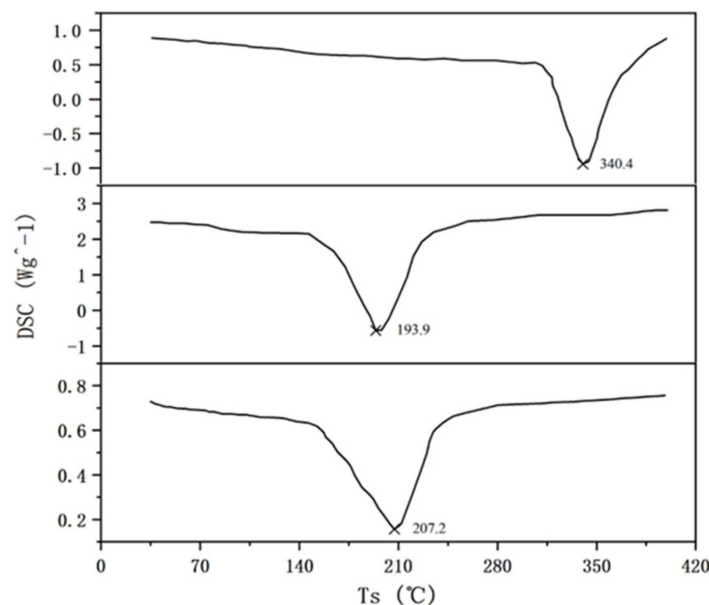


Figure 6. DSC thermal spectra of the samples JM186, #1, and #2.

3.4. Gel Microstructure and Cross-linking Mechanism Analysis

The strength and water-blocking properties of the gel system are inevitably linked to the microstructure of the gel. Therefore, to figure out the microstructure of the secondary cross-linking gel system, freezing environment scans were performed on three samples, and the results are shown in Figure 7.

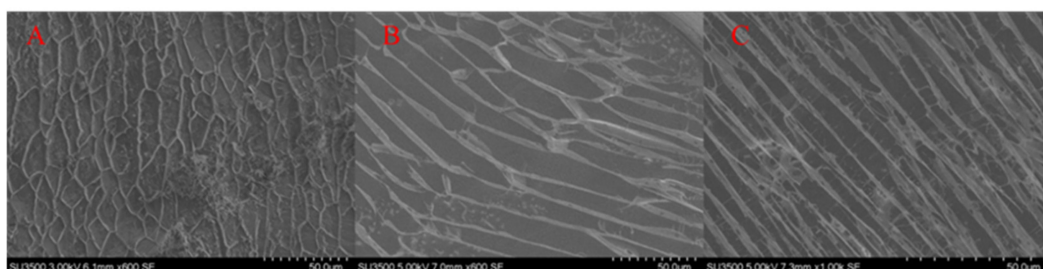


Figure 7. The structures of the gel samples JM86 (A), #1 (B), and #2 (C).

Figure 7C shows the structure of #2 gel cross-linked with phenolic resin alone. It can be clearly observed that there are many filamentous connections within the polyacrylamide skeleton, which are much finer compared to those of JM186 and #1 gels. This is the reason why the #2 gel is weaker than its companions. Figure 7B shows the #1 gel cross-linked with organozirconium alone. The #1 gel has a relatively thicker connection and a more pronounced grid compared to the #2 gel, which explains the higher strength of the #1 gel. Figure 7A shows the JM186 gel. JM186 has a more dense and uniform pore structure compared to #1 and #2 gels. The dense, multi-branch, and uniform porous network structure contributes to the low dehydration rate and good thermal stability of the secondary cross-linked gel system.

Figure 8 shows the schematic diagram of the proposed secondary cross-linking mechanism. The gel formed by phenolic resin and the polyacrylamide showed a looser but elastic network structure. A more rigid and stronger network structure was formed between organic zirconium and polyacrylamide, which made up for the lower strength of the phenolic resin gel system. The denser and more thickened network structure made the gel system more stable. Therefore, the water-blocking of the secondary cross-linked gel system was stronger than that of the single inorganic or organic cross-linker gel system.

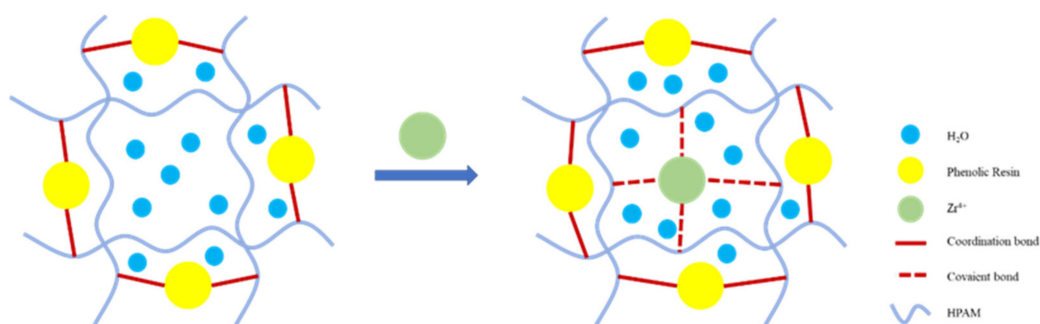


Figure 8. Schematic diagram of the secondary cross-linking reactions.

3.5. Evaluation of Blocking Effect

The JM186 gel system was evaluated for blocking effect, and the blocking efficiency was used to evaluate the lab blocking effect of this sample [29]. The results are shown in Table 5, and the average blocking efficiency of the JM186 gel system is 96.03%. After massive water injection, a high-permeability channel appeared in the sand-filled pipe, which could simulate the high-permeability channels in the reservoir. Then, the gel-forming liquid was injected and passed through the high-permeability channel. A gel was formed under high temperature to achieve the blockage of the high-permeability channel. The final permeability reduction can be 10–20 times lower than its initial state. This indicates that the water was blocked successfully.

Table 5. The water-blocking results of the JM186 gel system.

Sample No.	Permeability before Injection (mD)	Permeability after Gel Injection (mD)	Blocking Rate (%)
JM186	167.564	6.367	96.2
	208.643	10.849	94.8
	230.724	6.691	97.1

4. Conclusions

In this paper, a low-cost, high-temperature-resistant, and high-strength gel system JM186 was obtained by secondary cross-linking of polyacrylamide. The performance of the gel system was evaluated in terms of dosage, rheology, structure, and water-blocking efficiency. The main conclusions were obtained as follows:

1. Under the economic dosage, 0.3 wt% polyacrylamide can form gels regardless of organic zirconium or phenolic resin cross-linker. However, the phenolic resin gel system had low strength compared with the organic zirconium gel system, which lacks good water-blocking performance. The organozirconium gel system had poor water-blocking capability and lacked temperature resistance ability.
2. After initial cross-linking by phenolic resin (0.3%) and secondary cross-linking by organic zirconium (1.6%), a JM186 gel system can be obtained with temperature resistance up to 120 °C. The average elastic modulus and viscous modulus can reach 32.33 Pa and 3.25 Pa, which are significantly greater than those of the single cross-linker systems.
3. By changing the dosage of cross-linking agents, the JM186 gel system gel-forming time can range from 1 h to 9 h, and the strength of the gel can reach H grade. The water-blocking efficiency of the sand-filled pipe can reach 96.03%.
4. After secondary cross-linking by organic zirconium, the microscopic grid structure of the JM186 was denser and more thickened compared with the gel system of the single cross-linker, which is the reason why JM186 had the good thermal stability and greater gel strength.
5. The JM186 gel system is expected to achieve low cost and high water-blocking efficiency in high-temperature reservoirs. It will increase the wave efficiency of water injection and thus enhance the oil recovery rate.

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Conflicts of Interest: The authors declare no conflict of interest.

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