Development and Performance Evaluation of a High-Temperature Profile Control System

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ABSTRACT: In the late stage of the steam flooding development of heavy oil reservoirs, the contradiction between layers is prominent, which makes the development of the reservoir difficult. To obtain a profile control agent suitable for steam flooding in heavy oil reservoirs, the modification technology was used to improve the temperature resistance of the traditional NH-1 main agent, as the conjugation effect may enhance the stability of the molecular structure. Thus, we obtained the modified NH-1 main agent, which was used in combination with graphite particles. To explore the oil displacement effect of the profile control system, the single-variable method was used to optimize the profile control system and evaluate its performance. Moreover, the multimedium steam flooding experiment was carried out to further verify the profile control capability of the profile control system. The results show that the formula of the graphite particle–gel compounding system is 0.03% coagulant BK-A05 + 2.2% cross-linking agent I + 1.8% cross-linking agent II + 6% modified NH-1 main agent (prepared by reacting NH-1 main agent with 65% concentrated nitric acid at a solid–liquid ratio of 1:6) + 0.7% graphite particles (10 000 meshes) + 0.3% suspending agent CMC. The gel viscosity of the profile control system can reach 2 × 10^6 mPa-s, the gel temperature range is wide (160–280 °C), and the temperature resistance is 300 °C. The profile control system has good injection performance, plugging effect, washing resistance, and thermal stability and is significantly better than the gel system alone. The experiment results also show that the profile control system has a strong profile control capability and expands the swept volume. The crude oil recovery increases by 8.89%, and it can be effectively applied to the profile control operation of heavy oil multimedium steam flooding.

1. INTRODUCTION

Low-permeability and ultralow-permeability reservoirs are widely distributed in oilfields throughout the world. Low permeability, low porosity, and poor reservoir properties lead to water injection difficulties and low oil recovery in these oilfields.1 At present, the water content in many oilfields is up to 95%. In heterogeneous reservoirs, water channeling results in only oil displacement in high-permeability zones, but unswept oil needs to be recovered to improve the recovery efficiency.1-5 Based on the profile control technology of water injection reservoirs, the high-temperature profile control technology has gradually developed and high-temperature profile control technologies based on gels, particles, foams, and other media have been developed.1,2,6 At present, preformed particle gel has been successfully synthesized and applied to control excess water production in most of the mature, waterflooded oilfields in China.10,11 Zhao et al. successfully prepared phenolic resin dispersed particle gel (PDPG) by a mechanical shearing method and systematically analyzed the effects of bulk gel strength, shearing time, shearing rate, and bulk gel–water ratio on the PDPG particles. Zhao et al. believed that the shearing rate is one of the most critical factors in the preparation. In addition, the bulk gel–water ratio has a slight impact on the prepared PDPG particles. The experiment indicated that PDPG particles can easily enter the deep formations, with a temperature resistance of 150 °C and a pH value of 4–8.11 Al-Muntasheri et al. mixed polyvinylamide with aldehyde and phenolic cross-linking agents, with low initial viscosity and easy injection, and its gel temperature is around 205 °C; it can be used for profile control operations in steam flooding reservoirs.12 Micron-size polyacrylamide elastic microsphere (MPEM) is a newly developed profile control and oil displacement agent for enhanced oil recovery in heterogeneous
For high-salinity reservoirs, Yang et al. researched a hydrophobic groups of PAMA. Tang et al. synthesized to reduce surface tension and the association function between a higher hydrophobic group content could enhance the ability of a polymer microsphere bulk gel to a certain extent and be used like polymer microspheres. The study showed that the addition of compound high-temperature polymer/nano SiO₂ composite microspheres (PNSCMs) via inverse suspension polymerization with 3-methacryloxypropyl-triethoxysilane (MPS)-modified SiO₂ as the reinforcing filler. Many enhanced oil recovery (EOR) techniques have been applied like polymer flooding to decrease the mobility of water. Similarly, another widespread example of EOR is polymer gel that can be used as a plugging agent in high-permeability zones to transfer the displacing fluid toward the low-permeability zone, thus displacing the existing reservoir and increasing the oil production.

With the increasing profile control requirements of different formation conditions and interlayer characteristics, the compound high-temperature profile control technology is increasingly favored for its advantages of strong plugging ability and deep profile control. The compound profile control agent is generally compounded by the principle of bridging between the gel and the particles. The traditional gel profile control agent has good selectivity and deformability, while the main agent plays a vital role in the temperature resistance of the gel system.

The traditional NH-1 main agent is a nontoxic high-molecular natural organic substance; its molecules contain oxygen-containing functional groups such as phenolic hydroxyl groups and quinone groups, and its temperature resistance is only 180 °C. Aiming at the harsh profile control environment caused by steam channeling in high-temperature reservoirs with steam flooding, as the conjugation effect may enhance the stability of the molecular structure, the modification technology was used to improve the temperature resistance of the traditional NH-1 main agent in which nitro was added. The modified NH-1 main agent can be copolymerized with coagulant (HPAM, BK-A05), aldehyde, and phenolic cross-linking agents to produce a gel with temperature resistance of 300 °C. The gel was used in combination with high-temperature graphite particles to further improve the scouring resistance of the profile control system and extend the use period of the profile control agent. Through the laboratory evaluation experiment, the applicable limit of the profile control agent was obtained from the perspective of formation condition adaptability. Through the high-temperature dynamic performance evaluation, the plugging rate, scouring resistance, and thermal stability of the profile control system were investigated. The purpose of this work is to develop a high-temperature profile control agent suitable for steam flooding, to improve the contradiction between layers in the later stage of steam flooding, and to increase the economic benefit.

2. RESULTS AND DISCUSSION

2.1. Optimization of the Modified High-Temperature Gel System. 2.1.1. Content of Coagulant BK-A05. The mass fraction of the modified NH-1 main agent was 6%, that of the cross-linking agent I was 2.2%, and that of the cross-linking agent II was 5%. The effect of the mass fraction of coagulant BK-A05 (0.015, 0.02, 0.025, 0.03, and 0.035 wt %) on the gelling performance of the gel solution is shown in Table 1. With the increase in the content of coagulant BK-A05, the gel viscosity of the gel solution increased, and after the content of coagulant BK-A05 was higher than 0.015%, the gel time was shortened slightly.

The content of BK-A05 has a greater influence on the initial viscosity of the solution. As the content of BK-A05 increases, the initial viscosity of the solution increases. As can be seen from Figure 1, as the content of BK-A05 increased, the gel viscosity increased and the gel time decreased. Therefore, considering the gelling performance and time, the mass fraction of coagulant BK-A05 was selected as 0.03% (Figure 2).

2.1.2. Content of the Modified NH-1 Main Agent. The mass fraction of coagulant BK-A05 was 0.03%, that of cross-linking agent I was 2.2%, and that of cross-linking agent II was 1.5%. The gel viscosity of the gel solution at different modified NH-1 main agent contents (4, 6, 8, 10, and 12%) is shown in Table 2. When the content of the modified NH-1 main agent increased from 4 to 12%, the gel viscosity increased positively. The phenolic hydroxyl group in the molecular structure of the modified NH-1 main agent and the aldehyde cross-linking agent formed a phenolic pre-condensate. However, with low reaction, a long-molecular-chain coagulant was needed to provide the amide group. Furthermore, it should be

Table 1. Change of the Gel Viscosity of Gel Solution with Different Mass Fractions of BK-A05

| time (h) | 0.015 | 0.02 | 0.025 | 0.03 | 0.035 |
|---------|-------|------|-------|------|------|
| Viscosity (mPa·s) |
| 2.31 | 3.01 | 3.92 | 4.33 | 5.21 |
| 1 | 48 587 | 39 382 | 43 831 | 50 511 | 69 015 |
| 2 | 71 183 | 88 324 | 88 124 | 87 654 | 91 134 |
| 3 | 102 000 | 133 000 | 137 000 | 151 000 | 173 000 |
| 4 | 272 000 | 261 000 | 303 000 | 397 000 | 392 000 |
| 5 | 379 000 | 485 000 | 515 000 | 685 000 | 868 000 |
| 6 | 490 700 | 745 000 | 778 000 | 998 000 | 1 140 000 |
| 7 | 878 000 | 912 000 | 962 000 | 1 090 000 | 1 110 000 |
| 8 | 889 000 | 933 000 | 982 000 | 1 020 000 | 1 150 000 |
| 9 | 880 000 | 908 000 | 974 000 | 1 050 000 | 1 160 000 |
| 10 | 882 000 | 918 000 | 968 000 | 1 050 000 | 1 160 000 |
copolymerized with the active functional groups in other crosslinking components, so that the gel net structure becomes denser and stronger. As the content of the modified NH-1 main agent increased, the probability of copolymerization and cross-linking reaction among the four molecules increased, resulting in a decrease in gel time.

As the mass fraction of the main agent increased, the gel viscosity increased. When the mass fraction of the main agent was 6%, the gel time was up to 7 h. When the mass fraction of the main agent was 12%, the generated gel had the highest viscosity, with short gel time and fast reaction. When the mass fraction of the main agent further increased, the main agent reached saturation in the solution and was difficult to continue to dissolve. Based on the gel time and gel viscosity, the mass fraction of the main agent was selected as 6%.

### 2.1.3. Content of Cross-Linking Agent I

The mass fraction of the modified NH-1 main agent was 6%, that of coagulant BK-A05 was 0.03%, and that of cross-linking agent II was 1.5%. The variation of the gel viscosity of the gel solution with different mass fractions of cross-linking agent I (1.65, 2.2, 2.75, 3.3, and 3.85%) is shown in Table 3.

As the mass fraction of the cross-linking agent I increased, the gel time of the system decreased. When the mass fraction of cross-linking agent I was 2.2%, the gel had the maximum viscosity, which was $1.2 \times 10^6$ mPa·s. According to Table 3 and Figure 3, the gel viscosity increased first and then decreased as the mass fraction of the cross-linking agent I increased, that is, when the mass fraction ranged from 1.65 to 3.85%. Therefore, considering the gel time and gel viscosity, the mass fraction of the cross-linking agent I was selected as 2.2%.

### 2.1.4. Content of Cross-Linking Agent II

The mass fraction of the modified NH-1 main agent was 6%, that of coagulant BK-A05 was 0.03%, and that of cross-linking agent I was 2.2%. The variation of gel viscosity of the gel solution with different mass fractions of cross-linking agent II (1.2−2.4%) is shown in Table 4.

As shown in Table 4 and Figure 4, with the increase of the content of cross-linking agent II, the system’s gel viscosity increased and the gel time decreased. The hydrolysate of cross-linking agent II was able to react with the other three agents. When the content of cross-linking agent II exceeded 1.8%, the gel time was short. The formula of the modified high-temperature gel system finally obtained through the ratio optimization experiment was as follows: 0.03% coagulant BK-A05 + 2.2% cross-linking agent I + 1.8% cross-linking agent II + 6% modified NH-1 main agent.

### 2.2. Optimization of the Graphite Particle−Gel Compounding System

#### 2.2.1. Optimization of CMC Content of the Suspending Agent

As shown in Figure 5, when the mass fraction of CMC was 0.3%, the suspension rate of graphite particles reached more than 98%, and there was almost no sedimentation at the bottom after 48 h. Graphite particles have good fluidity and dispersibility, without water precipitation. Therefore, the CMC mass fraction of the suspending agent was determined to be 0.3%.

#### 2.2.2. Optimization of Graphite Particle Content

The content of graphite particles was optimized based on the plugging rate of the sand-filled pipe and the plugging rate after 30 PV steam flushing. As shown in Figure 6, with the increase of graphite particle content, the plugging rate of the compounding system to the sand-filled pipe increased, reaching more than 98.4%. When the graphite particle content was greater than 2%, the injection became difficult. As the graphite particle content increased, the plugging rate of the sand-filled pipe after 30 PV steam flushing increased until it became stable. The inflection point appeared when the content was 0.7%. Therefore, the content of graphite particles (10,000 meshes) in the compounding system was determined to be 0.7%.

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**Table 2. Variation of the Gel Viscosity of the Gel Solution with Different Mass Fractions of the Modified NH-1 Main Agent**

| time (h) | NH-1 main agent mass fraction (%) | Viscosity (mPa·s) |
|---------|----------------------------------|------------------|
| 0       | 4 3.31 3.89 4.01 4.33 4.74       |                  |
| 1       | 43 4.52 5.2 3.12 3.8 4.94 5.8 5.15 7.8 8.9 5                  |
| 2       | 77 2.74 7.8 3.24 8.8 285 8.8 315 135 000                  |
| 3       | 141 000 149 000 173 000 171 000 1 080 000                  |
| 4       | 387 000 391 000 523 000 538 000 2 000 000                  |
| 5       | 595 000 662 000 758 000 778 000 1 890 000                  |
| 6       | 773 000 823 000 1 340 000 1 410 000 2 000 000                  |
| 7       | 788 000 1 100 000 1 340 000 1 430 000 1 910 000                  |
| 8       | 810 000 1 090 000 1 330 000 1 490 000 1 950 000                  |
| 9       | 798 000 1 110 000 1 350 000 1 410 000 2 000 000                  |
| 10      | 790 000 1 100 000 1 330 000 1 410 000 2 000 000                  |

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**Figure 1.** Effect of the mass fraction of coagulant BK-A05 on gelling performance.

**Figure 2.** Effect of the mass fraction of NH-1 main agent on gelling performance.
Therefore, the formula of the graphite particle–gel compounding system was determined as follows: 0.03% coagulant BK-A05 + 2.2% cross-linking agent I + 1.8% cross-linking agent II + 6% modified NH-1 main agent + 0.3% CMC + 0.7% graphite particles (10,000 meshes).

### 2.3. Evaluation of Adaptability of the Modified High-Temperature Gel System

#### 2.3.1. Temperature

The actual injection wells needed to be shut down for 1 week for artificial water injection to reduce the temperature before manual profile control to simulate the temperature near the well zone (160, 180, 200, 240, and 280 °C). The influence of temperature on the gelling performance of the modified high-temperature gel system (0.03% coagulant BK-A05 + 2.2% cross-linking agent I + 1.8% cross-linking agent II + 6% modified NH-1 main agent, the same below) is shown in Figure 7.

In the above temperature range, the profile control agent is a semisolid gel with a dark reddish-brown appearance. The gel has a certain elasticity, and the gel surface is relatively smooth. Except for 160 °C, no liquid appeared around the gel at other temperatures. This shows that the gel reaction of this system is fully carried out and the gelling effect is good.

### Table 4. Variation of the Gel Viscosity of the Gel Solution with Different Mass Fractions of Cross-linking Agent II

| time (h) | 1.2 | 1.5 | 1.8 | 2.1 | 2.4 |
|----------|-----|-----|-----|-----|-----|
| 0        | 2.84| 3.27| 3.15| 3.34| 3.15|
| 1        | 2.69| 45.46| 86.78| 101.00| 98.905|
| 2        | 1620| 85.15| 153.00| 188.00| 1010.00|
| 3        | 2932| 101.00| 1030.00| 1560.00| 1470.00|
| 4        | 2696| 175.00| 1920.00| 2000.00| 2000.00|
| 5        | 3099| 577.00| 2000.00| 2000.00| 2000.00|
| 6        | 6764| 910.00| 1980.00| 1960.00| 2000.00|
| 7        | 8938| 986.00| 2000.00| 1890.00| 1950.00|
| 8        | 37392| 912.00| 2000.00| 2000.00| 2000.00|
| 9        | 36890| 998.00| 2000.00| 2000.00| 2000.00|
| 10       | 37487| 942.00| 1880.00| 2000.00| 2000.00|
As shown in Table 5, in the simulated formation temperature range, the modified high-temperature gel system has a gel viscosity of $4.3 \times 10^5$ mPa·s or more, which can meet the plugging needs, and has the advantages of wide gelling temperature range and high utilization rate.

### Table 5. Effect of Temperature on the Gelling Performance of the Gel System

| Temperature (°C) | 160 | 180 | 200 | 240 | 280 |
|------------------|-----|-----|-----|-----|-----|
| Gel Viscosity (10^5 mPa·s) | 0.43 | 1.25 | 2.00 | 1.35 | 2.00 |

2.3.2. **pH Value.** In the experiment, the reaction temperature was set at 200 °C, and the water used for preparation was changed to aqueous NaHCO₃ solution, with a salinity range of 1600–4800 mg/L.

### Table 6. Effect of pH on the Gelling Performance of the Gel System

| pH | Gel Time (h) | Gel Viscosity (mPa·s) |
|----|--------------|-----------------------|
| 6  | 18           | $2 \times 10^6$       |
| 7  | 16.5         | $2 \times 10^6$       |
| 8  | 17           | $1.57 \times 10^6$    |
| 9  | 24           | $1.15 \times 10^4$    |
| 10 | non-adhesive |                       |

2.3.3. **Salinity.** The reaction temperature was set at 200 °C, and the water used for preparation was changed to aqueous NaHCO₃ solution, with a salinity range of 1600–4800 mg/L.

As shown in Figures 8 and 9, the experimental results show that when the salinity was between 1600 and 3200 mg/L, the viscosity of the plugging agent changed a little and the gel time increased. When salinity was more than 4800 mg/L, the gel time became significantly shorter.

### 2.4. High-Temperature Dynamic Performance Evaluation of the Profile Control System

#### 2.4.1. Plugging Ratio and Residual Resistance Factor.**

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alone. The blocking rate at high temperature is more than 99%, and the blocking effect changes little with the reaction temperature. This shows that the profile control agent can still react and block channeling in the formation environment. Therefore, the profile control agent has a perfect plugging performance.

2.4.2. Scouring Resistance. We evaluated the scouring resistance of the modified high-temperature gel system, the graphite particle, and the compounding system. Table 7 shows the permeability and plugging rate of the sand-filled pipe after steam flushing with different PV number. In the compounding system, after injection with 30 PV high-speed and high-temperature steam flushing, the system showed excellent scouring resistance and the plugging rate only dropped by 0.35%. The sand-filled pipe using the modified high-temperature gel system or graphite particles alone had poor scouring resistance. The compounding system can improve the retention ability of the plugging agent in the formation and extend the validity period.

2.4.3. Thermal Stability. At 300 °C, the thermal stability of the profile control agent was evaluated by the change in the plugging rate of the sand-filled pipe with profile control by the compounding system over time. The thermal stability test results of the three systems are shown in Figure 12. High-temperature aging had basically no effect on the stability of graphite particles. The blocking rate dropped to 85.9% on the 50th day. At the 50th day of aging, the blocking rate of the sand-filled pipe by the compounding system was 97.63%, only down 1.63%. The compounding system had good thermal stability; the graphite particles and the gel system supported each other. When the gel degraded, the graphite particles can still block the dominant pores with high strength.

Table 7. Scouring Resistance of Different Profile Control Systems for Plugging Sand-Filled Pipes

| system            | water permeability measurement (10⁻³ μm²) | before blocking | after blocking | 15 PV | 30 PV | 30 PV blocking rate % |
|-------------------|------------------------------------------|-----------------|---------------|-------|-------|-----------------------|
| gel system        |                                          | 680             | 14            | 71    | 336   | 50.59                 |
| graphite particles|                                          | 703             | 14            | 34    | 101   | 85.63                 |
| compounding       |                                          | 912             | 8             | 9     | 11    | 98.78                 |

Figure 9. Gelling type of the profile control agent under different salinity conditions.

Figure 10. Blocking rate curve of a sand-filled pipe with different systems.

Figure 11. Variation curve of the plugging rate and the residual resistance factor of the graphite particle—gel compounding system at different reaction temperatures.

Figure 12. Comparison curves of the thermal stability of different systems.
Through the above performance evaluation, the graphite–gel compounding system has excellent scouring resistance and good thermal stability. The blocking rate can reach more than 99% at high temperature.

2.5. Evaluation of the Effect of Profile Control System. In the experiment of profile control agent evaluation, the relationship between the cumulative volume of injected pore volume and the cumulative recovery factor of heavy oil is assessed and is shown in Figure 13. It can be found that before the injection of the profile control agent, the cumulative recovery rate of the multimedium steam flooding for heavy oil was 53.03%. At this time, the water content was as high as 98%, and there was no longer any heavy oil production (Figures 14 and 15).

After injection of the profile control agent, aging was performed according to the optimal aging time 5 days, and then the multimedium steam flooding was resumed. It can be seen from Figure 13 that the recovery factor and injection pressure of heavy oil increased gradually with the increase of pore volume multiple. It shows that the profile control agent forms a blockage on the original steam channeling channel, so that the injected hot fluid starts to advance to the unswept region and displaces the heavy oil not produced previously. When the water content reached 98%, the cumulative recovery of heavy oil was 61.92%.

After calculating the increase in the rate of the recovery factor, the results show that after injection of the profile control system, the recovery factor of heavy oil increases by 8.89%, with significantly enhanced oil recovery. It shows that the compounding system can have a good profile control and plugging effect in the process of heavy oil multimedium steam flooding. It can improve the oil displacement efficiency, expand the swept volume, and improve the crude oil recovery. This system can be used in profile control operations of heavy oil multimedium steam flooding.

3. CONCLUSIONS

(1) The formula of the graphite particle–gel compounding system is 0.03% coagulant BK-A05 + 2.2% cross-linking agent I (phenols) + 1.8% cross-linking agent II (aldehydes) + 6% modified NH-1 main agent + 0.3% CMC suspending agent + 0.7% graphite particles (10 000 meshes). The gel temperature range is wide (160–280 °C), and the temperature resistance is 300 °C. The system is suitable for formation conditions with a pH range of 6–8 and alkaline water salinity below 8000 mg/L.

(2) The compounding system has good injection performance and a high blocking rate, greater than 99%. After 30 PV high-temperature steam flushing, the blocking rate can still reach 96.78%. After aging at 300 °C for 50 days, the blocking rate is 97.63%. The scouring resistance and thermal stability of the compounding system are significantly better than those of the gel system alone.

(3) The compounding system contains a small amount of graphite particles. The gel can degrade with steam flushing. Due to the lubricity of particles, the system has no damage to the pump during injection and production and has broad development prospects.

(4) The results of multimedium steam flooding experiments show that the profile control system has a strong profile control capability and expands the swept volume. The crude oil recovery increases by 8.89%. Therefore, this system can be effectively applied to the profile control operation of heavy oil multimedium steam flooding.

4. EXPERIMENTAL MATERIALS AND METHODS

4.1. Development of the High-Temperature Profile Control System. 4.1.1. Experimental Agents. The exper-
imental agents used were as follows: NH-1 main agent, chemically pure, Tianjin Fine Chemical Research Institute; cross-linking agent I (phenols), cross-linking agent II (aldehydes), and coagulant BK-A05 (HPAM relative molecular mass: 12 million), Henan Baike Company; graphite particles (2000–10 000 meshes), Qingdao Jiririle Graphite Co., Ltd.; suspending agent CMC (content ≥ 95%), Renqiu City Yanxing Chemical Co., Ltd.; NaOH, NaCl, and NaHCO₃, analytically pure and commercially available; HNO₃ (65–70%) commercially available.

Experimental water was distilled water. The sand-filled pipe was formed by pressing high-temperature-resistant hydraulic powder and quartz sand. The permeability of water measurement ranges from 600 × 10⁻³ to 1000 × 10⁻³ μm², with a diameter of 2.5 cm and a length of 30 cm.

4.1.2. Experimental Instruments. The following instruments were used: A YZHR-type high-temperature and high-pressure hydrothermal synthesis reactor (50 mL), Beijing Yanzheng Biotechnology Co., Ltd.; a 60227iec53 pressure monitoring device, Tianzhou Electric Group Co., Ltd.; a Brookfield rotational viscometer, American Brookfield Corporation; a SIN-PH100 pH test pen, Hangzhou Sinomasure Automation Technology Co., Ltd.; a PL4002-IC electronic balance, Shanghai Youyi Instrument Co., Ltd.; a constant-temperature magnetic stirrer, Shanghai Sile Instrument Co., Ltd.; a beaker, test tube, glass rod, condenser tube, three-necked flask, and thermometer.

4.1.1.1. Development of the Modified NH-I Main Agent. The quantitative NH-1 main agent and concentrated nitric acid were added into a three-necked flask for reflux condensation in a constant-temperature water bath at 90 °C. After 3 h of reaction, the product was taken out and placed in a flask. Then, the product was evaporated to dryness using a constant-temperature magnetic stirrer to obtain the modified NH-1 main agent.

The substituent nitro group was added to the benzene ring structure of the NH-1 main molecule. Based on the conjugation effect, the strong electronegativity generated by the nitro group changed the molecular electron cloud density and improved the molecular stability. The mass ratio of the NH-1 main agent to nitric acid (solid–liquid ratio) was 1:6. The modified NH-1 main agent was synthesized with different nitric acid mass fractions (10, 40, and 65%). The gel solution was prepared with 0.015% coagulant (HPAM, BK-A05) + 2% cross-linking agent I + 1.6% cross-linking agent II + 6% modified NH-1 main agent. The gel solution was gelled at 300 °C and placed for 24 h. The comparison shows that the solid–liquid ratio was 1:6, the dehydration amount of the gel tended to be stable, no scorch phenomenon was observed on the gel surface, and the structure was still compact.

Therefore, when synthesizing the modified NH-1 main agent, the mass fraction of nitric acid was 65% and the solid–liquid ratio was 1:6. The aqueous solution of the modified NH-1 main agent was dark red, and it was saturated when the mass fraction was 14%. At this time, the viscosity reached 2112 mPas. Considering that the initial viscosity of the field system should not be too large, the content of the modified NH-1 main agent in the following optimization experiment should be controlled within 12%.

4.1.1.2. Preparation of the Modified High-Temperature Gel System. An appropriate amount of coagulant (HPAM, BK-A05) was weighed and dissolved in distilled water until it was fully swollen, and quantitative cross-linking agent I and cross-linking agent II were added in sequence. Under sealed conditions, it was stirred at a speed of less than 100 r/min for 10 min until it mixed well, and then, a certain amount of modified NH-1 main agent dissolved in distilled water was added to obtain the gel solution after mixing well.

4.1.1.3. Preparation of the Gel–Graphite Particle Compounding System. Graphite particle solution with CMC as the suspending agent and the modified high-temperature gel system solution were prepared. The dosage of each agent was twice the target ratio content. The gel–graphite particle compounding system was obtained by mixing two solutions at a mass ratio of 1:1.

4.2. Optimization of the High-Temperature Profile Control System. 4.2.1. Optimization of the Modified High-Temperature Gel System. The experiment was aimed to optimize the content of each composition, and we used the gelling properties (gel time and gel viscosity) as the screening criteria. The temperature of the simulated formation was 200 °C, and the pH of the plugging agent solution was 7.4.

4.2.1.1. Experimental Agents. The experimental agents used were as follows: coagulant, modified main agent, cross-linking agent I, cross-linking agent II, pH adjusting agent, and formation water.

4.2.1.2. Experimental Instruments. The experimental instruments used were as follows: a hydrothermal synthesis reactor, an electric balance, a Brookfield rotary viscometer, an incubator, an electric blender, a pH meter, a measuring cylinder, a beaker, and so on.

4.2.1.3. Experimental Method. An appropriate modified NH-1 main agent, cross-linking agent I, and coagulant were weighed and sufficiently dissolved in water, respectively, and the pH value was adjusted to 7. The three kinds of solutions were mixed and added with the cross-linking agent II. The mixed solution was divided into 10 portions approximately, each of which was put into the hydrothermal synthesis reactor, sealed, and placed in the reaction environment (200 °C). One reactor was taken out every 1 h and cooled to room temperature in the air. Then, the reactor was opened (the reactor cannot be opened at a high temperature); the routine method was used to measure the viscosity of gel in the reactor with a viscometer. The appropriate rotor was selected for rotation. When the number fluctuated in a range with a small amplitude, the average should be recorded. The ratio of each

Table 8. Effect of the Nitric Acid Mass Fraction on the Gelling Performance of the Gel Solution (Solid–Liquid Ratio, 1:6)

| mass fraction of nitric acid (%) | gel viscosity (mPa·s) | gelling                              |
|---------------------------------|----------------------|-------------------------------------|
| 0                               | 20.1                 | ungelled                            |
| 10                              | 32.5                 | ungelled                            |
| 40                              | 32 558               | gel with medium mobility, low strength |
| 65                              | 2 000 000            | rigid gel with compact gel structure |
composition in the solution using the above procedure should be changed.

4.2.2. Optimization of the Graphite Particle–Gel Compounding System. 4.2.2.1. Optimization of CMC Content of the Suspending Agent. Taking the suspension rate as the criterion for evaluating suspension performance, the suspension rate is calculated as follows

\[ Q = \frac{(h_1 - h_2)}{h_1} \]  

(1)

where \( Q \) is the suspension rate in \%, \( h_1 \) is the volume of suspension solution in mL, and \( h_2 \) is the volume of water precipitated from the suspension solution at 48 h in mL.

Furthermore, weigh a certain amount of CMC solution and dissolve it in water using an electric mixer to stir at 1000 r/min until it is completely dissolved. Weigh the graphite particles with a mass fraction of 2% and a mesh range of 2000–10 000, disperse them in a beaker, and stir at high speed for 30 min. Pour suspension solutions with different meshes into the corresponding 100 mL measuring cylinders and allow them to stand for 48 h. Record the amount of precipitated water, and calculate the suspension rate.

4.2.2.2. Optimization of Graphite Particle Content. A certain amount of 10 000 mesh graphite particles was added to the CMC solution with a mass fraction of 0.3% and then mixed with the gel system at a mass ratio of 1:1 to obtain the graphite–gel compounding system. The content of graphite particles was optimized based on the plugging rate of the compounding system to the sand-filled pipe and the plugging rate after 30 PV steam flushing.

4.3. Evaluation of Adaptability of the Modified High-Temperature Gel System. The gelling performance of the gel system was measured by simulating different formation conditions in the mine (changing the reaction conditions of the modified high-temperature gel system).

4.4. High-Temperature Dynamic Performance Evaluation of the Profile Control System. Preparation of the sand-filled pipe: Turn the sand-filled pipe into vacuum with the vacuum pump and saturate it with water after half an hour. Then, measure the pore volume and permeability before plugging.

4.4.1. Seal Test. Connect the experiment devices according to Figure 16. Turn on the valves c and d, turn off the valves a and b. Inject water through the pump to find the place that is not sealed. Change or tight the connection until it is well sealed.

4.4.2. Plugging Experiment. Fill the profile agent solution in the piston container. Turn on the valves a, b, and d, and turn off the valve c. Displace 3 PV profile control agent into the sand-filled pipe at a constant speed of 1 mL/min, keep it sealed, and put it into the thermostat at 200 °C for 12 h until it is gelled.

4.4.3. Plugging Ratio and Residual Resistance Factor. After the profile agent gels, measure the water phase permeability in the air. Inject water at speeds of 1, 3, 5, 7, and 9 mL/min; calculate the water phase permeability, respectively; and then get the average. The plugging ratio and residual resistance factor are calculated by formulas 2 and 3.

Plugging ratio \[ E = \frac{(K_1 - K_2)}{K_1} \times 100\% \]  

(2)

Residual resistance factor \[ RRE = \frac{K_1}{K_2} \]  

(3)

where \( K_1 \) is the water phase permeability before plugging, \( K_2 \) is the water phase permeability after plugging.

4.5. Evaluation of the Effect of Profile Control System. To evaluate the effect of the graphite–gel compounding system on the oil displacement in the process of heavy oil multimedium steam flooding, we simulated the multimedium steam flooding process by injecting air and steam mixed in a certain proportion. The experimental process is shown in Figure 17. The heavy oil used in the experiment was taken from Block Jin 91 in the Liaohe Oilfield, and the viscosity at 80 °C was 598.4 mPa·s.

The multimedium steam flooding process was simulated by mixed injection of air and steam at a mass ratio of 98:2. After flooding to 98% of the limit water content, the graphite–gel compounding system was injected and aged for a period of time before observing the change in recovery factor. The description of specific experimental steps is as follows.

1) Fill two sand-filled pipes with a permeability of 2000 × 10\(^{-12}\) μm\(^2\) with 50–70 mesh quartz sand, and conduct gas permeability measurements first and then water permeability measurements.
2) Saturate the sand-filled pipe with formation water by the vacuum method and record the pore volume.
3) Saturate the sand-filled pipe with heavy oil at 80 °C and record the saturated oil volume.
4) Connect the experimental instruments according to Figure 16. Check the air tightness of the instrument. Set the thermostat temperature to 80 °C. Pressurize the back pressure valve by a hand-operated metering pump, which is always 2 MPa higher than the injection pressure.
5) Set the internal temperature of the steam generator to 200 °C. Set the steam injection rate to 2 mL/min using the ISCO pump. Open the piston container with

![Figure 16. Flow chart for determination of the residual resistance factor.](https://dx.doi.org/10.1021/acsomega.0c02642)

![Figure 17. Flow chart of multimedium steam flooding.](https://dx.doi.org/10.1021/acsomega.0c02642)
compressed air, adjust the injection pressure through the pressure regulating valve, and read the air injection rate by the wet gas meter. The air injection rate was controlled at 34.91 mL/min, so that the injection mass ratio of steam to air was 98.2.

6) At the steam—air mass ratio of 98:2, carry out air-steamed flooding on the sand-filled pipe with saturated heavy oil. Record the pressure, the volume of the liquid, and the volume of produced water in the measuring cylinder every 5 min.

7) When the water content reached 98%, the profile control system was injected into the sand-filled pipe and aged at 80 °C for 5 days.

8) After the aging was over, continue the air-steamed flooding for the sand-filled pipe in the same manner. Record the pressure and the volume of oil and water in the measuring cylinder. Stop the experiment when the water content reached 98%.

9) Calculate the cumulative recovery of heavy oil at different times. Observe the cumulative recovery of heavy oil before and after injection of the profile control system. Calculate the increase in the rate of recovery factor. Evaluate the effect of this profile control system on the oil displacement during the multimedia steam flooding process.

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Notes
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