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Original Paper

A novel polymer gel with high-temperature and high-salinity resistance for conformance control in carbonate reservoirs

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ABSTRACT

Plugging agents have been widely used to enhance oil recovery in fractured-vuggy carbonate reservoirs. However, the harsh conditions of fractured-vuggy carbonate reservoirs yield a significant challenge in maintaining a long-term stabilization of plugging agents. In this work, we developed an anti-high-temperature and high-salinity polymer gel (APG) with excellent resistance to high temperature (140 °C) and ultra-high salinity (240000 mg/L). The rheology and microstructure of APG were characterized before and after gelation. Core plugging tests on fractured cubic cores were conducted to quantify the plugging performance of the gel system. Experimental results showed that the Sclerglucan and Cobalt (II) Chloride Hexahydrate filled the three-dimensional (3-D) network with various morphologies, providing extra protection to the cross-linking points of the 3D network structure of APG and thus, leading to a prolongation of the dehydration time. The dehydration rate of APG was only 5% within 30 days, and the strength of APG could be maintained at a rigid or near-rigid level over 150 days. Moreover, APG exhibited satisfactory shear and scour resistance. Core plugging tests showed that APG could achieve a plugging rate of 90% and demonstrate ignorable minor damage to the substrate. Our results indicate that APG can serve as a great candidate in channel plugging in fractured-vuggy carbonate reservoirs where fractures are fully developed.

1. Introduction

According to the rapid development of the oil industry, the exploration scale of oil and natural gas has been continuously expanded. Water production is becoming an inevitably severe problem in most mature oilfields globally, especially in sandstone and carbonate reservoirs. When the water cut exceeds a certain amount in the production well, oil production is enormously influenced. It is estimated that 2.8 barrels of water are produced on average with each barrel of oil worldwide (Buciak et al., 2013; Al-Nakhli et al., 2016; Rassenfoss and Jacobs, 2017; Kumar et al., 2020). Most mature oilfields have entered the mid to late stages of water flooding, and dominant channels are formed due to the long-term water scour and the enhanced reservoir heterogeneity, yielding a significant decrease in sweep efficiency of the injected fluids (Hu et al., 2015; Lamas et al., 2021; Zhong et al., 2021). Therefore, how to effectively control water and increase oil has gradually become the mainstream topic in the field of oil exploitation.

Since the 20th century, with a variety of experiments in lab and application in-situ demonstrates, elastic polymers, bulk gel system, and foams into sandstone reservoirs in low permeability oilfields were proved to be successful, which revealed that chemical methods for water production control have emerged (Allen and Robinson, 1993; Moradi-Araghi, 2000; Li et al., 2010; Sun and Bai, 2017). Particularly gels, such as different kinds of bulk gel systems, of which main advantage is the formations of low-cost polymer and crosslinker, which is the most widely used water plugging agents and its feasibility and practicality to plug high permeability channels in hydrocarbon reservoirs has been proved for decades (Allen and Robinson, 1993; Moradi-Araghi, 2000).

Gels, chemical water plugging is one of the commonly used

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treatments, which is mainly synthesized by polymer and cross-linker. For example, PAM is the representative polymer to crosslink metallic or organic cross-linkers to form the polyacrylamide-based gel system (Al-Muntasheri et al., 2006), such as the preformed particle gels (PPG) (Saghafi, 2018; Song et al., 2018; Wang et al., 2019), polymer microspheres (Tian et al., 2012; Yang et al., 2019), micro-gel (Yuan et al., 2019; Chen et al., 2020), and dispersed particle gels (DPG) (You et al., 2011) et al. Those profile control and water plugging agent were synthesized using non-ionic, cationic, amphoteric acrylamide monomers, polyacrylamide, cross-linkers, initiator or other additives by polymerization to present a perfect thermo-stability in the majority of medium and high temperature reservoirs at 60–120 °C, those reservoirs of which salinity no more than 150000 mg/L can be exploited at the same time (Li et al., 2015; Esfahlan et al., 2021).

However, almost all gel plugging agents cannot be satisfactory in harsh reservoir conditions. For example, ultra-deep carbonate reservoirs, which exist in the oilfields in Western China and Central Asia, those reservoirs with ultra-high temperature (≧ 140 °C) and ultra-high salinity (≧ 240000 mg/L). So that the development of water plugging agents for these reservoirs is very challenging (Gao et al., 2016; Bai et al., 2018; Ding et al., 2020). When the temperature is over 140 °C, the abovementioned gels quickly dehydrate and cannot form effective plugging. Cecilia et al. proceed nano-composite modification treatment; the PPG can only satisfy high temperature (130 °C) and high salinity (84000 mg/L) reservoirs. Buddha Bhushan et al. developed HT-PPG which is resistant in 150 °C high temperature for more than 18 months (Durán-Valencia et al., 2014; Salunkhe et al., 2021). Khoshkar et al. and Paprouschi et al. found the effectiveness and performance of the Nano-PPG on increasing the sweep efficiency of water injection and improving the oil production only in Low Salinity Water flooding (Khoshkar et al., 2020; Paprouschi et al., 2021). Yang et al. introduced soft microgel with salinity resistance of only 5000 mg/L (Yang et al., 2016). Kang et al. proved that plugging agents such as in-situ formed monomer gels, polymer gels using metal cross-linkers, organic gel systems, PPG, DPG and so on, none of them can be satisfactory in high temperature (140 °C) and formation water (240000 mg/L) at the same time (Kang et al., 2021). The majority of those chemical water plugging agents cannot be satisfactory with ultra-deep carbonate reservoirs. Therefore, plenty of polymer molecular chains break under high temperature and high salinity, which leads to the destruction of the gel grid structure and degradation of the polymer, facilitating gel dehydration. (Zhu et al., 2017).

Recently, some of the polymer gels with much higher thermal resistance have been developed. However, among them only a few are used in oilfields because of their high costs. Particular polymer gels must overcome problems like costs and develop the ability to plug carbonate permeable channels for sufficiently long times (Kang et al., 2021; Song et al., 2021). Therefore, finding a high temperature and high salinity resistant bulk gel system, which is an effective conformance control method to reduce water production is urgent when exploiting fractured-vuggy carbonate reservoirs, such as Tahe Oilfield.

In this paper, an anti-high-temperature and high-salinity polymer gel (APG) system applied for high temperature and ultra-high salinity reservoirs was be synthesized. APG is fabricated using the formation water with high salinity. Anionic polyacrylamide molecular chain contains a certain number of polar groups that do not adsorb metal cations in the formation water, thereby remaining charge neutralization and condensation. To evaluate the bulk gel system, the strength, shearing time, shearing rate, and viscosity of the bulk gel system are analyzed by static characterizations. Core plugging test are conducted to assess the plugging performance of APG.

2. Experimental section

2.1. Materials

The chemicals used for APG fabrication include Anionic polyacrylamide SNF 55 (HPAM) with an average molecular weight of 700–800 kDaltons (SNF Co. Ltd., China), methenamine (AR, 99.99%), hydroquinone (AR, 99.99%), thiourea (AR, 99.99%), sclerglucan (USP) and CCH ($CoCl_2 \cdot 6H_2O$) (AR, 99.99%).

The formation brine used in our experiments was provided by the Tahe Oilfield. The main compositions of formation brine is presented in Table 1. The ion composition of formation brine was determined by SINOPEC standard titration method. The presence of SO_4^{2-} in the formation brine may generate salt crystals that may affect the observation using SEM (Scanning electron microscopy). Therefore, NaCl solution (240000 mg/L) was also prepared for SEM imaging of the gel system.

2.2. APG preparation

The polyacrylamide powder SNF 55 was dissolved in the formation water containing sclerglucan (SG). The SG solution is a typical non-Newtonian fluid with high stability (El Asjadi et al., 2018), therefore, it was selected as a stabilizer. What needs to be mentioned is that, SG has to be added into the aqueous solution at first, and then stirred at a high rotation speed to make sure SG can be dissolved in aqueous solution. The polymer powder was added slowly to ensure that the solution was transparent and fish-eye block substances were not observed. The prepared solution was well-mixed by agitating for 1.5 h. Then the crosslinkers (0.6 wt% methenamine and 0.6 wt% hydroquinone) and tackifier (0.3 wt% thiourea) were added into the prepared polymer solutions. The concentrations were optimized in preliminary experiments (Zhu et al., 2017b; Gu et al., 2018; Zhang et al., 2020). The obtained solution was well stirred for 15–20 min until all powders were fully dissolved. The high temperature stabilizer CCH was then added to the solution of which pH ranged within 5.8–6.5 and stirred the solution for 5 min, followed by placing the container into an oven at 140 °C until APG was gelated. It is crucial to notice that the liquid level of the polymer solution should be around 1/2–2/3 of each ampoule. An extra solution may cause rupture of the ampoules due to the volume expansion of the solutions in the process of gelation. In comparison, solution volume of less than half of the ampoule may result in difficulty in observation. APG solutions of various concentrations of SNF 55 (0.2 wt%, 0.4 wt%, 0.6 wt%, 0.8 wt% and 1.0 wt%), SG (0.02 wt%, 0.04 wt%, 0.06 wt%, 0.08 wt% and 0.1 wt%) and CCH (0.02 wt%, 0.04 wt%, 0.06 wt%, 0.08 wt% and 0.1 wt%) were prepared to determine the optimal proportion of each component.

According to C. Gu's research (Gu et al., 2018), phenol-

Table 1
Ion composition of formation brine sample.

Ions	$K^+ + Na^+$	Ca^{2+}	Mg^{2+}	SO_4^{2-}	Cl^-	Total salinity
Concentration, mg/L	88157.84	10420.56	2278.4	350	129282.25	230489.05

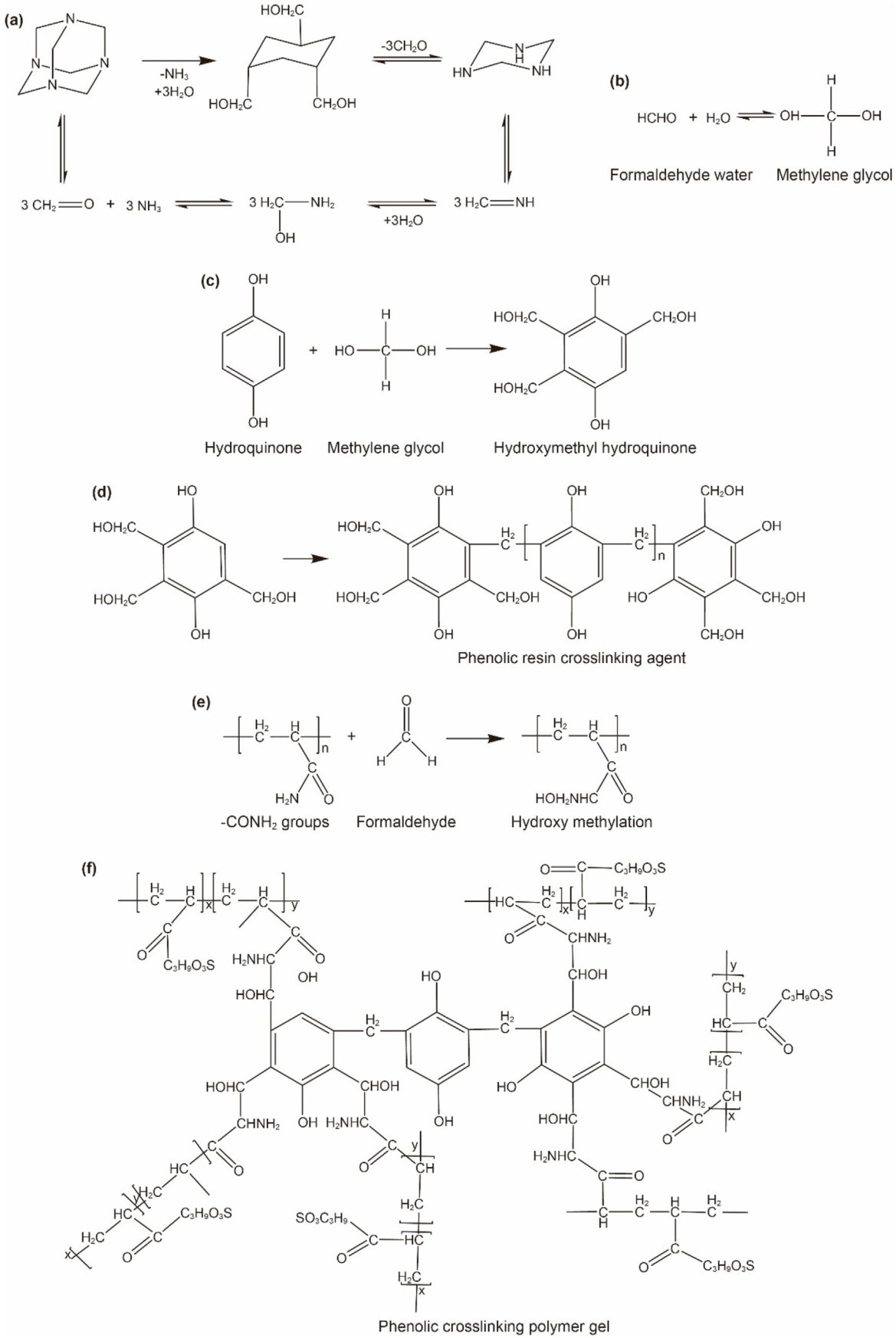

Fig. 1. Gelation mechanisms of the cross-linking reaction (Sun et al., 2016; Zhang et al., 2020).

formaldehyde is directly cross-linked with polyacrylamide to become pre-crosslinked solution. Phenol, hydroquinone, and resorcinol are commonly used as phenols, while formaldehyde and methenamine are commonly used as aldehydes. In this experiment, methenamine should be used as crosslinking agent, and it will release formaldehyde and NH_3 slowly during high temperature or acidic conditions (Fig. 1a); then formaldehyde is hydrolyzed to form methylene glycol (Fig. 1b), methylene glycol and hydroquinone can produce hydroxymethyl hydroquinone (Fig. 1c), phenolic resin crosslinking agent will be formed after dehydration condensation (Fig. 1d). Meanwhile, part of the $-CONH_2$ groups of polymer SNF 55 react with formaldehyde the perform hydroxy-methylation (Fig. 1e). Finally, phenolic crosslinking polymer gel was synthesized from phenolic resin crosslinking agent and hydroxymethylation polymer SNF 55 (Fig. 1f). The primary chemical reaction process as follows:

2.3. Rheology tests

The shear viscosity of APG with different concentrations was measured by the rotational rheometer (HAAKE-RS600, Thermo Electron Co., Germany) in fixed shear time at a shear rate of 7.34 s^{-1} under the temperature of 60 °C.

The storage modulus (G′) and loss modulus (G″) were also measured using the rotational rheometer with fixed shear stress (the scan frequency varies from 0.1 to 10 Hz) under the constant shear (CR) mode. The temperature was fixed at 60 °C due to the high volatility of the water content in the gel under high temperature, given that the measuring system is not closed. Therefore, although the measuring system is not closed and the moisture in the gel is extremely volatile, vaporization or weight loss has been restrained to the largest extent.

2.4. Sydansk gel strength tests

The flowing statuses of the gel system were observed via a test tube to justify the gel strength, which could be categorized into nine levels marked by alphabets from A to I (Table 2).

2.5. SEM characterization

The microstructure of APG particles was observed by SEM (Carl Zeiss Co., Germany). APG was placed into the Petri dish, followed by putting the Petri dish in a sealed vacuum freeze dryer for 10–12 h. In order to observe APG microstructure and measured its size better, it is necessary to sputter APG with gold-palladium.

2.6. Core flooding tests

A series of core flooding tests were performed on fractured marble cores to examine APG's plugging performance. The fractured cubic cores were 45 cm in length, 4.5 cm in height, and 4.5 cm in width (Fig. 2). The fracture apertures are 0.1 mm, 0.3 mm and 0.5 mm in different cores, respectively. The core fabrication method used in the experiment is illustrated as follows: the core holder compacts fine sand into a single homogeneous core, then the single homogeneous core is splited horizontally into two cores of the same volume and size. The two separated cores are filled with large sand and then cemented together with strong glue and wrapped using ethoxy resin.

Oil saturation was not carried out in this experiment, the cores saturated with AFW were placed into an oven and heated to reservoir temperature. The dry weight and water saturation of core samples were recorded. Then the AFW was continuously injected at a constant injection rate of 0.5 mL/min until the water cut reached 98%, followed by injecting 0.5 PV APG solution at the same injection rate, after that, the system was standing for over 72 h so that APG was gelatinized. Finally, the subsequent AFW was injected at 0.5 mL/min until the water cut reached 98%. The injection volumes, inlet pressure, and outlet pressure were recorded. The schematic diagram of water plugging experiment was shown by Fig. 3.

Table 2
Sydansk gel strength code standard table (Sydansk and Southwell, 2000).

Strength Codes		The name of the gel	The intensity of description
A	0	No detectable gel formed	The gel appears to have the same viscosity as the original polymer solution
B	2	Highly flowing gel	The gel appears to be only slightly more viscous than the initial polymer solution
C	4	Flowing gel	Most of the gel flows to the bottle cap by gravity upon inversion
D	6	Moderately flowing gel	Only a small portion (5%–10%) of the gel does not readily flow to the bottle cap by gravity upon inversion (usually characterized as a tonguing gel)
E	8	Barely flowing gel	The gel can barely flow to the bottle cap and/or a significant portion (> 15%) of the gel does not flow by gravity upon inversion
F	10	Highly deformable nonflowing gel	The gel does not flow to the bottle cap by gravity upon inversion
G	12	Moderately deformable non flowing gel	The gel deforms about half way down the bottle by gravity upon inversion
H	14	Slightly deformable nonflowing gel	Only the gel surface slightly deforms by gravity upon inversion
I	16	Rigid gel	There is no gel surface deformation by gravity upon inversion

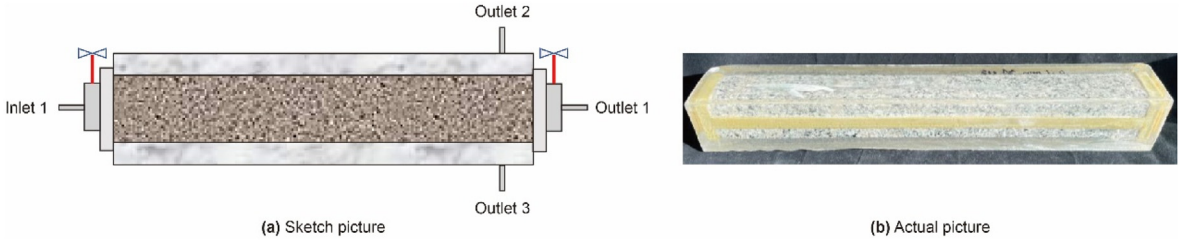

Fig. 2. The cubic fractured core samples.

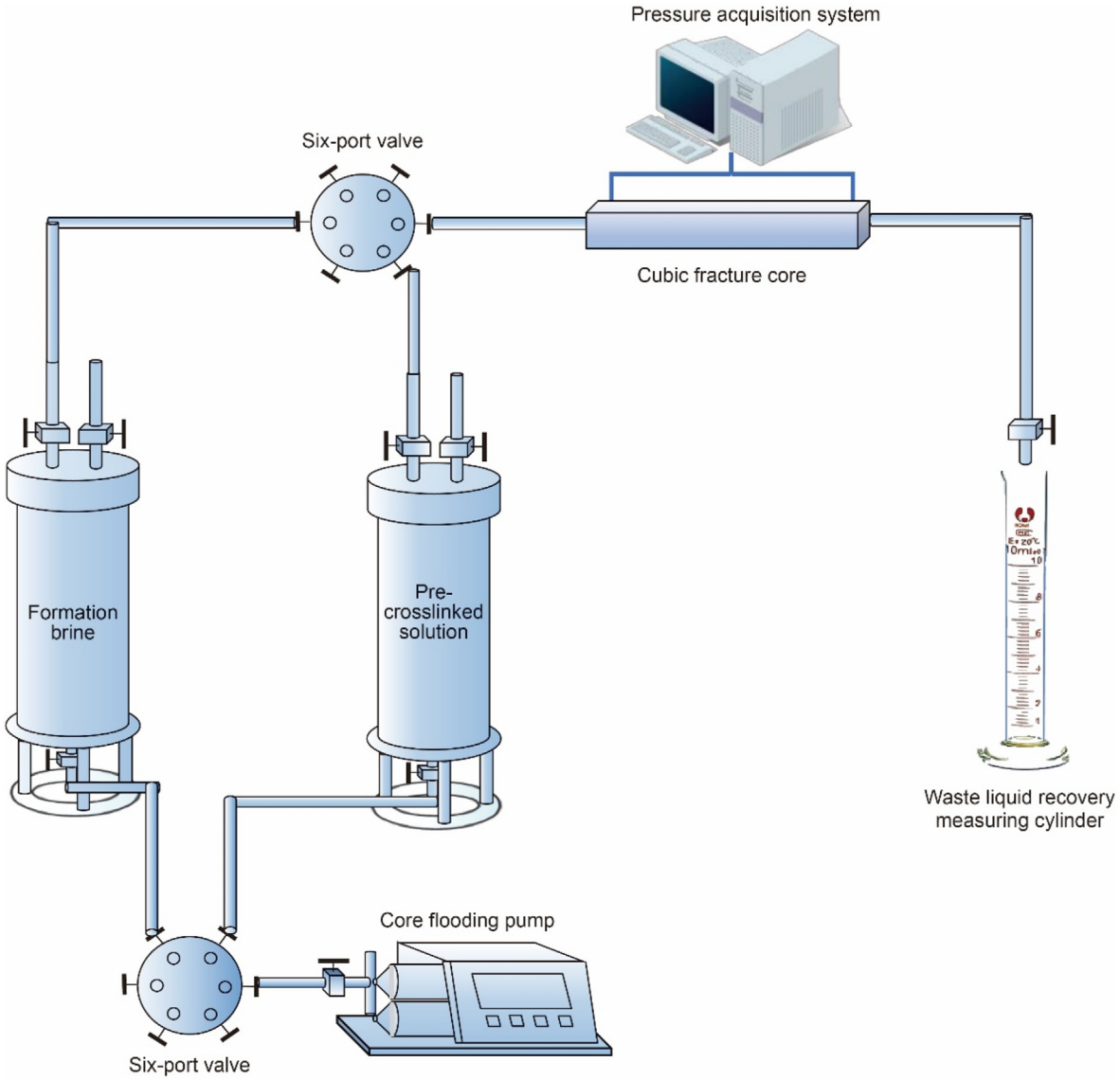

Fig. 3. Schematic diagram of water plugging experiment.

3. Results and discussion

3.1. Influence of different concentrations of materials

3.1.1. Influence of different concentrations of polymer

Polymer solutions containing different amounts of SNF 55 (0.2 wt%, 0.4 wt%, 0.6 wt%, 0.8 wt% and 1.0 wt%) were prepared to test its optimal concentration. The prepared solutions were placed in an oven set at 140 °C, and the dehydration rates and gel strengths were recorded. The dehydration rates of the polymer solutions at different aging times were listed in columns 4–8 in Table 3, calculated by

$$S = \frac{V - V_t}{V} \times 100\% \quad (1)$$

where V denotes the initial gel volume and V_t represents the gel volume after a certain aging time t (Karimi et al., 2014).

The initial strengths of the gel systems are presented in the second column in Table 3. Increasing polymer concentration yields a higher collision probability between the long chains of molecules.

Table 3
Influence of polymer concentration on the gelation time, gel strength and dehydration rate.

Concentration	Gelation time	Strength code	Dehydration rates at 140 °C, %				
			24 h	7 d	30 d	90 d	150 d
0.2%	/	B	0%	0%	0%	0%	0%
0.4%	4–4.5 h	E-F	0%	5%	20%	50%	50%
0.6%	3–4 h	F-G	0%	5%	10%	30%	50%
0.8%	3 h	G-H	0%	0%	10%	30%	50%
1.0%	2–2.5 h	G-H	0%	0%	10%	30%	50%

The twisted structures entangle with each other and therefore, enhances gel strength. As shown in Table 3, the gel strength upgrades from level B at 0.2 wt% to level G-H at 0.8 wt% and 1.0 wt% of SNF 55. Moreover, a higher polymer concentration results in a shorter gelation time and slower dehydration. It is clear that of 0.8 wt% and 1.0 wt% of SNF 55 provide comparable gel quality.

The viscosity corresponding to each polymer solutions of various polymer concentrations (0.2 wt%, 0.4 wt%, 0.6 wt%, 0.8 wt%

and 1.0 wt%) are 9.1 mPa s, 21.1 mPa s, 44.7 mPa s, 77.1 mPa s and 125.1 mPa s, respectively (Fig. 4). Apparently, a larger polymer concentration yields higher viscosity yet lower injectivity. Considering the dehydration time and economic viability, although the gelation time at 0.8 wt% is slightly longer than that at 1.0 wt%, we select 0.8 wt% as the optimal concentration for the polymer SNF 55. It is worth mentioning that SG and CCH show negligible influence on viscosity (data not shown).

3.1.2. Influence of different concentrations of polymer

APG solutions containing 0.8 wt% polymer, 0.6 wt% methenamine, 0.6 wt% hydroquinone, 0.3 wt% thiourea and SG of various concentrations (0.02 wt%, 0.04 wt%, 0.06 wt%, 0.08 wt% and 0.1 wt %) were prepared to determine the optimal concentration of the anti-acid stabilizer SG. The prepared solutions were placed in an oven set at 140 °C, and the dehydration rates and gel strengths were recorded. As showed in Table 4, the SG concentration does not significantly influence the strength and stability of APG solutions. However, the dehydration rate is 5% lower after 150 days in the presence of 0.1 wt% SG. As a result, we select 0.1 wt% as the optimal SG concentration.

3.1.3. Influence of different concentrations of polymer

APG solutions containing 0.8 wt% polymer, 0.6 wt% methenamine, 0.6 wt% hydroquinone, 0.3 wt% thiourea, 0.1 wt% SG and CCH of various concentrations (0.02 wt%, 0.04 wt%, 0.06 wt%, 0.08 wt% and 0.1 wt%) were prepared to determine the optimal concentration of CCH. The prepared solutions were placed in an oven set at 140 °C, and the dehydration rates and gel strengths were recorded.

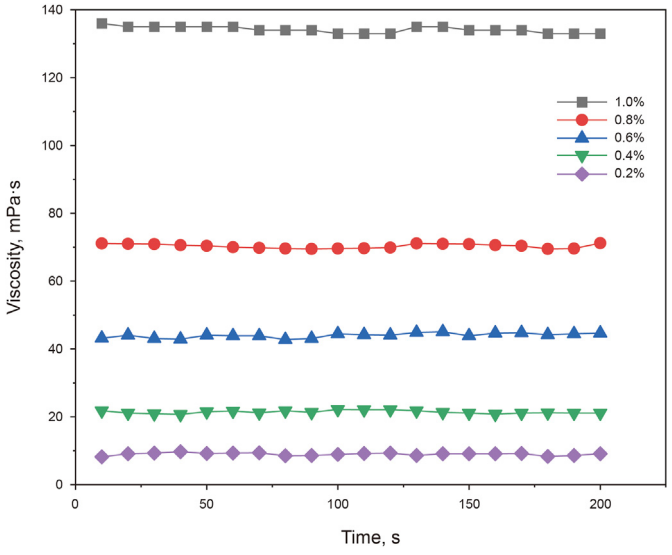

Fig. 4. Viscosity of different concentration before gelation of APG.

Table 4
Influence of SG concentration on the gelation time, gel strength and dehydration rate.

Concentration	Gelation time	Strength code	Dehydration rates at 140 °C, %				
			24 h	7 d	30 d	90 d	150 d
0.02%	3 h	G-H	0%	0%	5%	30%	50%
0.04%	3 h	G-H	0%	0%	5%	30%	50%
0.06%	3 h	G-H	0%	0%	5%	30%	50%
0.08%	3 h	G-H	0%	0%	5%	25%	50%
0.1%	3 h	G-H	0%	0%	5%	25%	45%

Table 5
Influence of CCH concentration on the gelation time, gel strength and dehydration rate.

Concentration	Gelation time	Strength code	Dehydration rates at 140 °C, %				
			24 h	7 d	30 d	90 d	150 d
0.02%	3 h	G-H	0%	0%	5%	25%	45%
0.04%	3 h	G-H	0%	0%	5%	25%	45%
0.06%	3 h	G-H	0%	0%	5%	25%	45%
0.08%	3 h	G-H	0%	0%	5%	25%	45%
0.1%	3 h	G-H	0%	0%	5%	20%	40%

As shown in Table 5, the CCH concentration shows no significant influence on the strength and stability of APG solutions. However, it should be noted that the dehydration rate is 5% lower after 90 days and 150 days in the presence of 0.1 wt% SG. As a result, we select 0.1 wt% as the optimal CCH concentration.

3.2. Thermal stability of samples prepared by high salinity AFW

Previous studies have shown that polymer solution is easy to hydrolyze at high temperatures because the cross-linked structure can be destroyed rapidly after reacting with the phenolic resin cross-linked system (Zhu et al., 2017). During this experiment, 0.8% polymer concentration was used to synthesis APG. Then, the thermal stability of APG can be improved in the presence of 0.1% SG and 0.1% CCH. The introduction of SG and CCH leads to a certain steric hindrance to inhibit the thermal hydrolysis of amide groups into carboxyl groups. Moreover, the energy that emerged in the process of partial thermal vibration may dissipate, and the presence of SG and CCH can alleviate the degree of thermal decomposition, hence, the number of fracture points on the polymer molecular chain is thus reduced (Fig. 5). As a result, adding SG and CCH to the bulk gel system could help the molecules form a stable structure and effectively mitigate the thermal decomposition of polymers. Fig. 6a and Fig. 6b show the initial microstructures of APG with and without adding SG and CCH, respectively. Generally, adding SG and CCH can inhibit the syneresis of gel, because the bound water is tightly bonded with the network of the gel via hydrogen bonds. Therefore, it is hard to be extruded from the gel (Wu and Ge, 2021). Gel strength codes were ranged from G to H for the samples shown in Fig. 6a and b. Fig. 6c and d demonstrate the microstructures of APG after 90 days of gelation with and without adding SG and CCH, the gel strength codes were F-G and E-F, respectively.

Moreover, in order to demonstrate whether SG and CCH were contained in the gel, EDX analysis was performed using SEM. In Fig. 7a, element cobalt and element chloride were found from EDX spectra analysis. On the contrary, element cobalt and element chloride cannot be observed in Fig. 7b from the EDX spectra analysis. As a consequence, the results of EDX spectra verified analysis about microstructure of SEM images. The cobalt and chloride ions adsorbed on the 3D mesh of APG make the gel have more coordination bonds and its thermal stability is improved.

3.3. Microstructure of APG

Fig. 8 shows the SEM images of the microstructures of APG with different polymer concentrations dissolved in NaCl solutions, APG were synthesized by each concentration (0.4 wt%, 0.6 wt%, 0.8 wt% and 1.0 wt%) and aged for 90 days, 0.2 wt% was excepted because it cannot be formed bulk gel. Since the number of amide groups ($-CONH_2$) on the polymer chains increases with the polymer concentration, the total number of active cross-linking points between the polymer molecule and the phenolic resin cross-linking agent increases. The regular 3D mesh structure is formed by

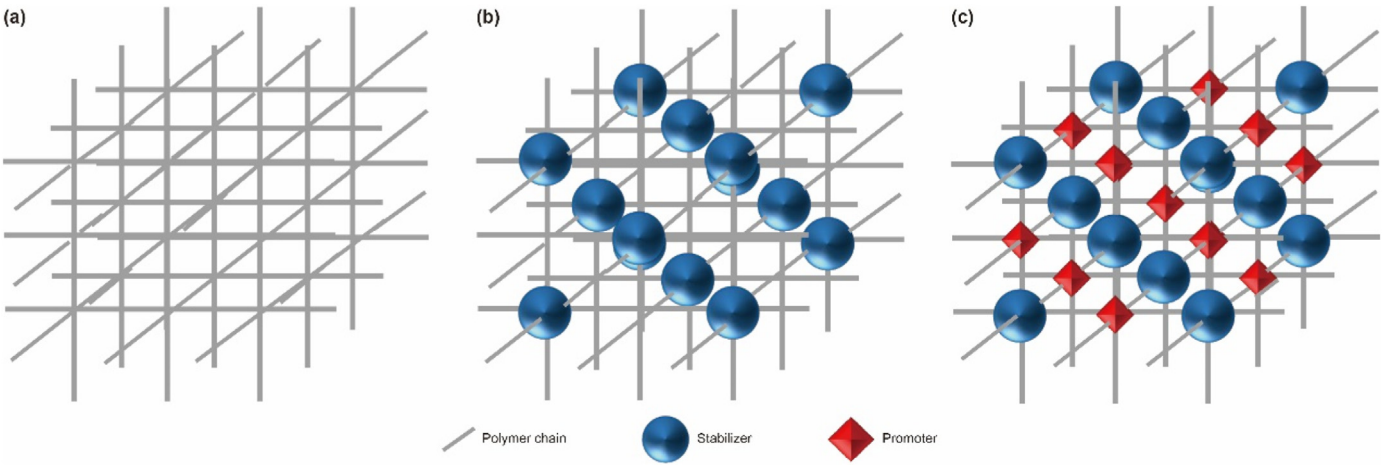

Fig. 5. Schematic of polymer molecules in the presence of different agents: (a) polymer; (b) polymer and SG; (c) polymer, SG and CCH.

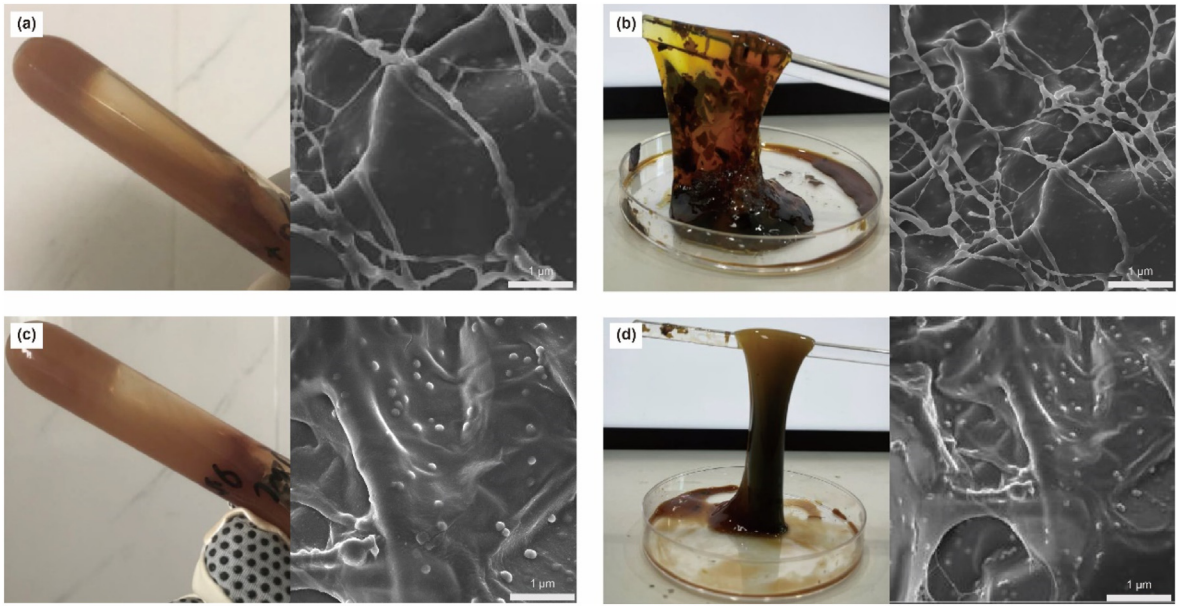

Fig. 6. Photos and SEM images of APG microstructures: (a) with SG and CCH; (b) without SG and CCH; (c) with SG and CCH after 90 d; (d) without SG and CCH after 90 d.

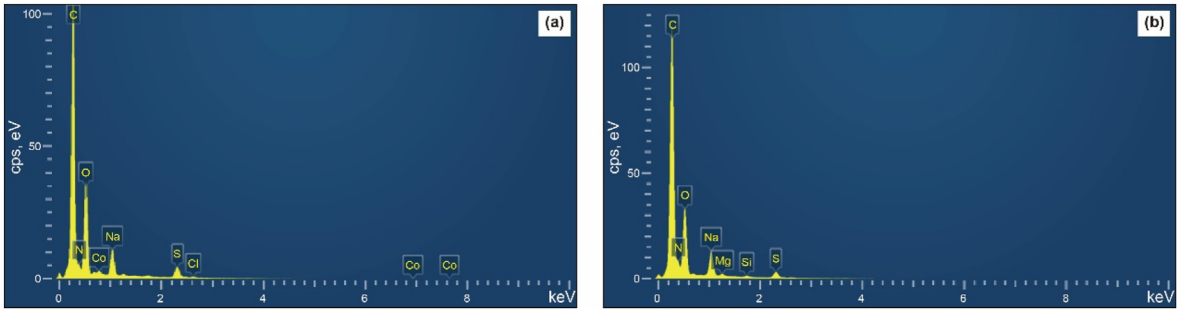

Fig. 7. EDX spectra analysis of APG after 90 d: (a) with SG and CCH; (b) without SG and CCH.

cross-linking amide groups and hydroxymethyl groups. Due to the high percentage of polymer and cross-linking agents, the generated 3D mesh is relatively compact. Even under the condition of high temperature and ultra-high salinity, it could still hold water, which indicates its good thermal stability. Meanwhile, when the sealed test tube containing gels is shaken violently in hand, the

appearance of the gel does not change, and the gel remains stable. Then the tube is placed in the oven again, and the dehydration period does not change either.

3.4. Viscoelastic analysis of APG after static formation

Viscoelasticity is an essential property for evaluating the gel system. Commonly, the energy storage modulus G′ is used to characterize gel elasticity, and the loss modulus G″ is applied to describe gel viscosity. The shear stress which is a function of frequency, was measured at a constant shear rate of 7.34 s^{-1}. G′ and G″ can be measured according to the change of frequency. Through the variation of G′ and G″, the viscoelastic change of gel can be analyzed. A higher G′ indicates lower gel deformability, stronger recovery force, greater resistance to impact and local damage after deforming. The lower the loss modulus, the weaker the internal friction resistance and scour resistance. Fig. 9 presents the measured G′ and G″ of the APG with different polymer concentrations (0.4 wt%, 0.6 wt%, 0.8 wt% and 1.0 wt%) dissolved in NaCl solutions. Viscoelastic measurements were not performed for 0.2 wt% because it could not form gel.

It can be seen from Fig. 9, when the shear stress is fixed within the range of 0.1–1 Hz at low frequency, storage modulus G′ and loss modulus G″ of APG synthesized by different polymer concentrations do not change significantly. Which represents macromolecule aggregates of APG deform according to the action of external force, the deformation relaxes and recovers after the external force is eliminated. In comparison, APG deforms under the action of external forces within the range of 1–10 Hz. After the elimination of external forces, the deformation does not recover, and part of the deformation is stored, leading to increased storage modulus G′ and loss modulus G″.

In addition, as observed from Fig. 9, both the energy storage modulus and loss modulus increase with increased shear frequency, and the energy storage modulus of the APG is higher than the loss modulus under the same polymer concentration,

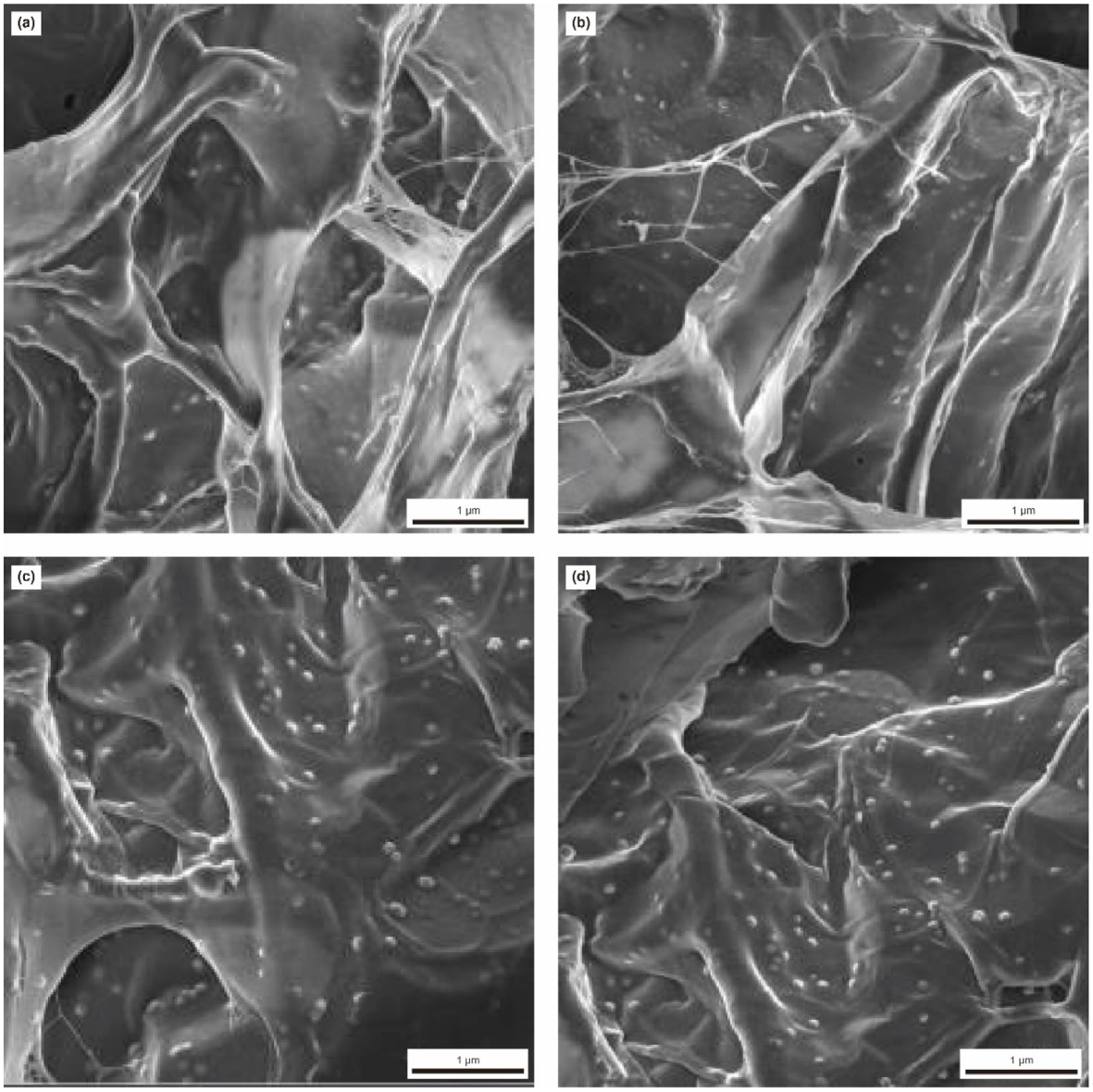

Fig. 8. Microstructures of APG dissolved in NaCl solutions: (a) 0.4% concentration polymer; (b) 0.6% concentration polymer; (c) 0.8% concentration polymer; (d) 1.0% concentration polymer.

indicating that the APG is mainly elastic and has certain deformability and restorability. The continuous augment of G′ and G″ suggest strong resistance to impact and local damage, good shear recovery after deformation. Therefore, it can be observed from Fig. 10 that the gel molecular chain breaks to form two pieces and stick together to form a piece of gel rapidly when APG was synthesized by 0.8% polymer concentration (see Fig. 10).

3.5. Plugging performance of APG in fracture core

The variations of the pressure drop of core floods conducted on cores of various fracture apertures are presented in Fig. 2. The detailed values are summarized in Table 6. The plugging rate of APG is calculated based on the following formula (Imqam et al., 2017):

$$Plugging\ Rate = \left(1 - \frac{1}{Frrw}\right) \times 100\% \quad (2)$$

where *Frrw* is the residual resistance factor, defined as the ratio of the injection pressure after gel plugging over the injection pressure before gel plugging.

The formation water was firstly injected into the lower layers with different fracture apertures (0.1 mm, 0.3 mm and 0.5 mm). After 2 PV injection, the pressures at pressure tap 1 are 0.81 kPa, 0.46 kPa and 0.21 kPa, respectively, corresponding to each fracture aperture from small to large. The pressures were relatively low due to the presence of fractures. A larger fracture opening leads to lower flow resistance and consequently higher-pressure difference across the core. Whereafter, 0.5 PV APG solutions were injected into the three lower fractured layers, respectively, with outlet valves 2 and 3 closed and the outlet valve 1 open as the only exit so that APG solutions can stay in the fractures to reach its largest extent and generate high-strength gels. During APG injection, the pressures at tap 1 show a slight increase to 3.44 kPa, 2.41 kPa, 1.46 kPa, respectively. After aging seven days in the closed oven under 140 °C, the subsequent formation water was injected at 0.5 mL/min. At this stage, outlet valve 1 was closed, and outlet valves 2 and 3 were open to achieve a detour flow.

The experimental results show that after the APG solution generates a stable plugging in the fractures (Fig. 11). The pressures at different aperture (0.1 mm, 0.3 mm, 0.5 mm) in fractured cores valve 3 were 103.85 kPa, 96.86 kPa, and 96.17 kPa, valve 2 were 100.82 kPa, 96.42 kPa, 92.05 kPa, respectively, suggesting high plugging intensity. The bulk gel system remains a satisfactory plugging performance even after one-week aging at 140 °C. According to Eq. 2, the plugging rates in the three fractured cores were 95.46%, 94.27%, and 90.27%, respectively.

4. APG application in high temperature and ultra-high salinity reservoirs in-situ

Before APG was used in TK681, the effect of water shutoff in oil wells can rarely last for over three months in Tahe Oilfield. Those developed gels of high storage modulus generally hold a short

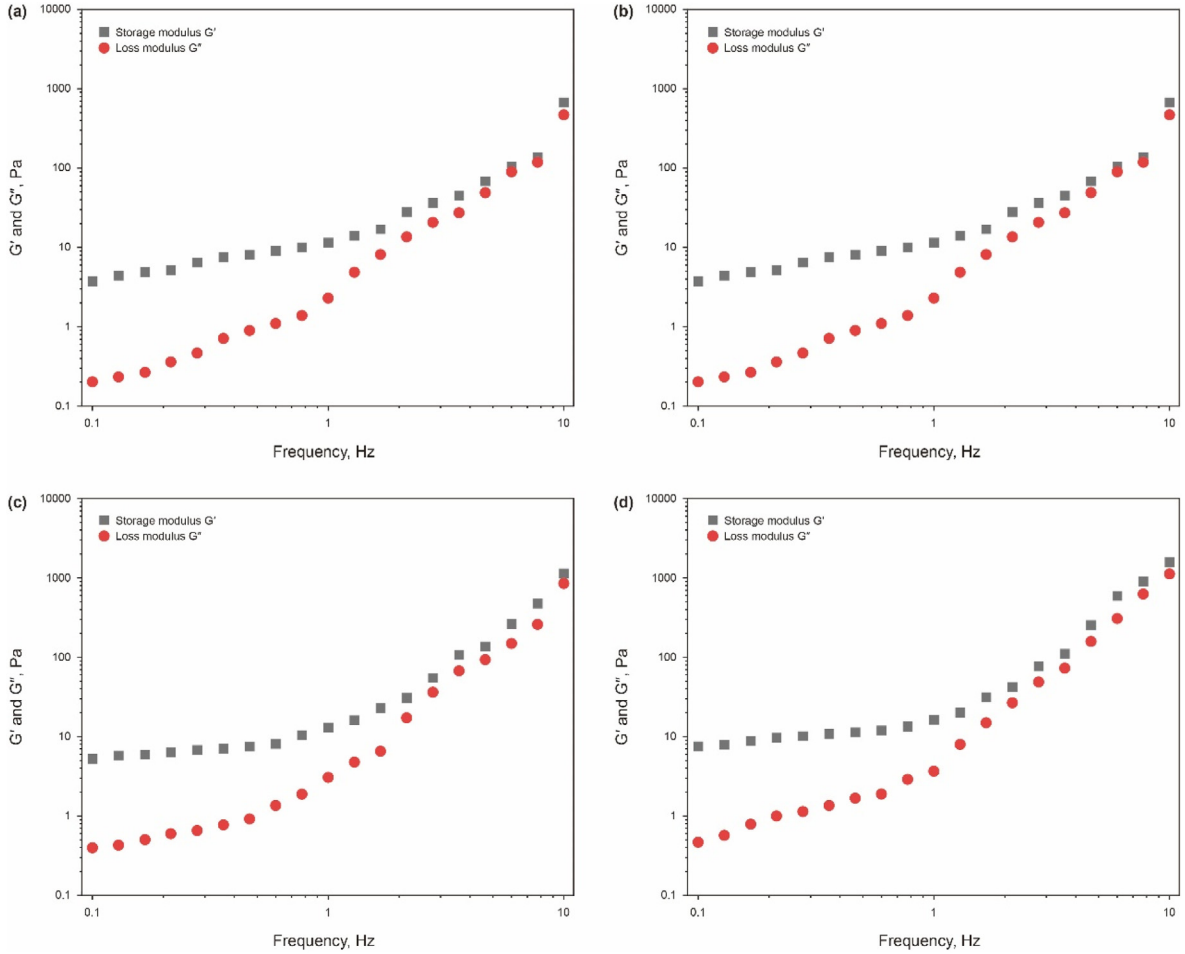

Fig. 9. The storage modulus and loss modulus of APG synthesized by different concentration polymer: (a) 0.4%; (b) 0.6%; (c) 0.8%; (d) 1.0%.

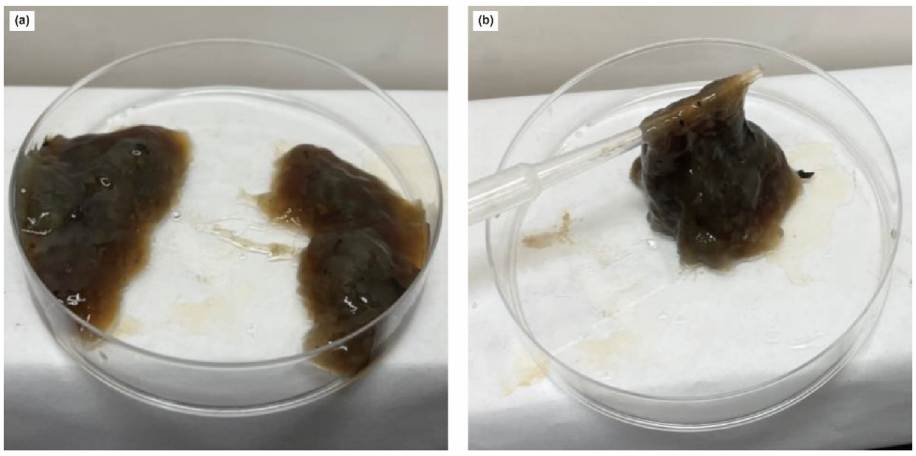

Fig. 10. APG with different morphologies: (a) separated state; (b) combined state.

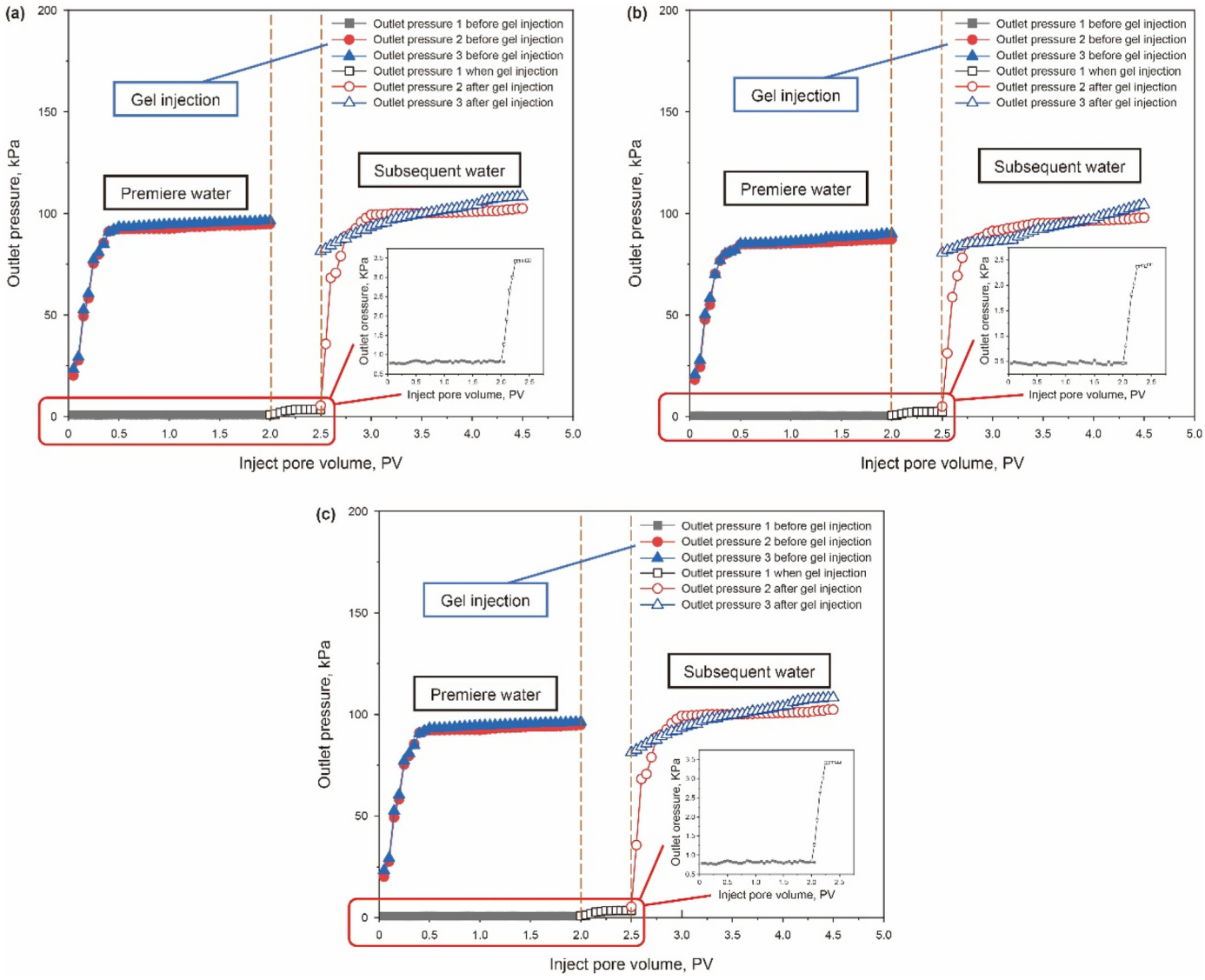

Fig. 11. Gel plugging performance in fractured cores with different apertures: (a) 0.1 mm; (b) 0.3 mm and (c) 0.5 mm.

Table 6
Plugging capacity of gel system in fractured cores.

No.	Aperture, mm	Permeability, mD	Plugging agent	Δp_1	Δp_2	Δp_3	Plugging rate
F1	0.1	48	APG	0.81	100.82	103.85	95.46%
F2	0.3	42		0.46	96.42	96.86	94.27%
F3	0.5	51		0.21	92.05	96.17	90.27%

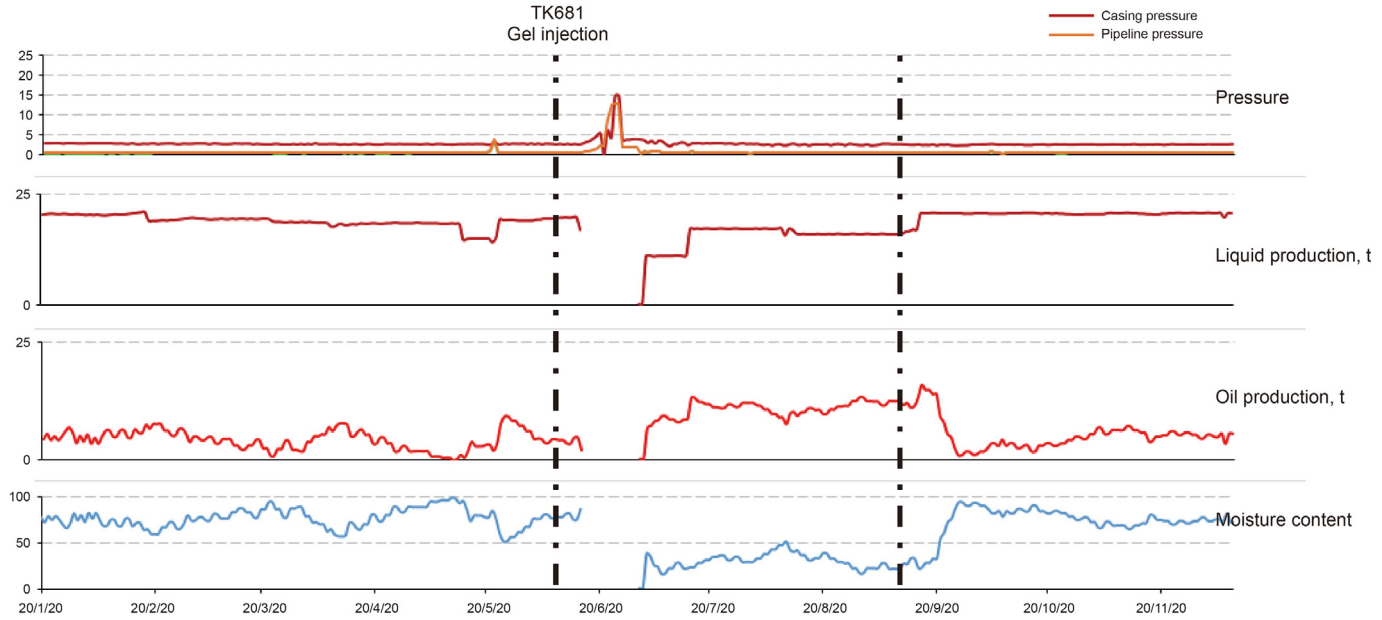

Fig. 12. Schematic diagram of production curve of Well TK681.

gelation time, which may cause wellbore or near-wellbore blockage in the ultra-deep, ultra-high temperature fractured-vuggy carbonate reservoir. The APG system was injected as a plugging agent for water shutoff in Well TK681 of Tahe Oilfield from June 20 to 24, 2020. This well was located in a typical carbonate fracture-vuggy reservoir where fractures were fully developed. The depth of the well was 5703 m, and the brine salinity of the oil reservoir was 207608 mg/L. As soon as the APG was completely gelated underground, the water cut was reduced from 90% to 50% without changing the fluid production, and the oil production increased from 5 t/d to 15 t/d. Later records of the well showed a stable oil production lasting for over three months. Because the formation water of the fracture-vuggy reservoir in Tahe Oilfield has great energy and high salinity, it has a strong ability of scouring and dilution to plugging agents. Moreover, the injection volume is less than 100 m^3 during the on-site application, and therefore, the thickness of the underground "colloidal wall" will not be too large. Consequently, APG exhibited a shorter stable period when applied on-site. However, the accumulated oil production reached 1000 t (Fig. 12), the economic input-output ratio reached 1:5 at least, which means it has good application potential.

5. Conclusions

An anti-high-temperature and high-salinity polymer gel (APG) for conformance control was developed. SG and CCH were utilized as stabilizers to provide more cross-link points for the 3D mesh structure of the bulk gel system, aiming at achieving satisfactory high-temperature resistance and high-salinity resistance. The APG was prepared within a brine solution of 240000 mg/L. The stability of the gel exhibited a dehydration rate of less than 5% after 30 days, and the strength was stabilized at a level ranging from G to H within 150 days. Besides, unlike the majority of polymer phenolic cross-linked gels, the APG showed satisfactory shear resistance and scour resistance, which remained stabilized with incremental shear time. Therefore, the APG demonstrated outstanding performance in blocking water channels. Meanwhile, APG injection showed minor damage to the substrate. After fully gelated, the plugging rate achieved 90%, and the breakthrough pressure of subsequent water flooding decreased with increased fracture aperture. APG was also successfully applied in the well TK681, exhibiting an effective period of over three months, which is much longer than other industrial gel systems that had been used in the same well.

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