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## Laboratory Evaluation Of High-Temperature Resistant Lysine-Based Polymer Gel Systems For Leakage Control

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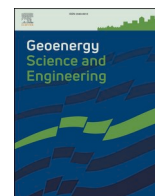
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## Laboratory evaluation of high-temperature resistant lysine-based polymer Gel systems for leakage control

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### ABSTRACT

In-situ crosslinking gel known for its cost-effectiveness, has been employed for decades to plug high-permeability features in subsurface environments. However, some commonly used crosslinkers are being phased out due to the increasingly rigorous environmental regulations. As a newly discovered environmentally friendly crosslinker, lysine can crosslink the partially hydrolyzed polyacrylamide through transamidation reaction. The present work aimed to study the effect of polymer composition and concentration on the gelation behavior of lysine and high molecular weight acrylamide-based polymers. Several commercial high molecular weight polymers with different contents of 2-Acrylamido-2-methyl-1-propane sulfonic acid (AMPS) including AN-105/125, SAV-55/37/28, and SAV-10 were deployed in this work. High-temperature/pressure-resistant glass tubes were used to study the crosslinking behavior. The plugging efficiency test used a high permeability sandstone core (1600 mD). Besides, the effect of salinity and pH on the crosslinking behavior, gel strength, and long-term thermal stability was also studied. Results showed that increasing the AMPS content could prolong the gelation time, but the polymer and lysine mixture failed to form gels when the AMPS content was excessively high. Additionally, increasing the AMPS content negatively affects the gel strength but positively impacts long-term thermal stability. Polymer gel with a relatively high content of AMPS could be stable in 0.5 % CaCl<sub>2</sub> solution at 130 °C for over 200 days. The polymer/lysine gel system can efficiently plug the high permeability matrix, and the plugging efficiency is higher than 99 %. The polymer/lysine gel system is a promising green leakage control system in treating the high conductivity features in geothermal and oil reservoirs.

### 1. Introduction

Polymer gels have been developed and extensively applied to plug high permeable zones in the geothermal and oil reservoirs to ameliorate the short circulation and excessive water production problems (Chen et al., 2023; Chen et al., 2022; Seright, 1995; Seright and Brattekas, 2021; Song et al., 2022b; Tschoepe III, 1982; Wang and Seright, 2021; Yu et al., 2022). Two kinds of polymers, including polysaccharides and partially hydrolyzed polyacrylamide (HPAM), are often used to produce gels in situ (Moradi-Araghi et al., 1988). It was reported that 22 types of metal ions could crosslink the polysaccharides, and some of these crosslinkers, such as Al (III), Cr (III), and Zr (IV) could also crosslink HPAM and its derivatives (Conway et al., 1983; Sydansk, 1993). Besides, organic crosslinkers such as phenol/formaldehyde could also crosslink

polysaccharides and acrylamide-based polymers (Bai et al., 2022; Dovan et al., 1997; Moradi-Araghi, 2000; Song et al., 2022a, 2023). However, the metallic or phenolic crosslinked polysaccharides and HPAM gels are limited to low temperatures and hardness reservoirs (Moradi-Araghi and Doe, 1987). HPAM gels undergo substantial syneresis under harsh conditions because the hydrolysis of amide groups can generate carboxylate groups, making the gel extremely vulnerable to divalent cations (DiGiacomo, 1983; Moradi-Araghi et al., 1993). In addition, polysaccharide gels have limited long-term thermal stability under harsh conditions due to the inherently weak ether bonds (Moradi-Araghi et al., 1993).

The temperature and salt resistance of polymer gels can be improved through polymer optimization and using organic crosslinkers. In terms of polymer optimization, the hydrolytic stability could be improved by

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**Table 1**  
Polymers used in this study.

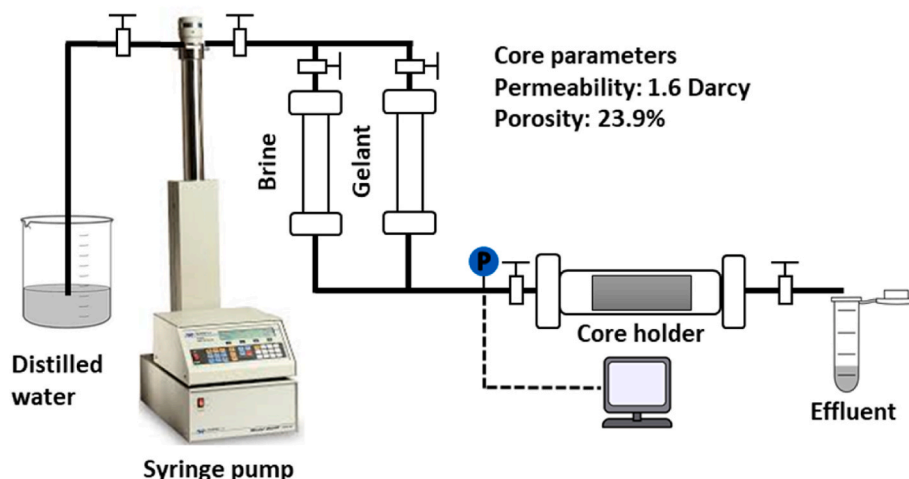
Polymer Name	MW/million dalton	AM mol %	AMPS mol %	NVP mol %
AN-105	8–10	95	5	0
AN-125	6–8	75	25	0
SAV-55	–	–	–	0
SAV-37	–	–	–	0
SAV-28	–	–	–	0
SAV-10	–	<10	>90	0
SAV-225	3–5	50–60	20–25	20–25

adding monomers meeting the criteria of inactive to divalent cations and resistance to hydrolysis. Some commonly used monomers are N, N'-dimethyl acrylamide, N-vinyl acetamide, 2-Acrylamido-2-methyl-1-propane sulfonic acid (AMPS), and N-vinyl pyrrolidone (NVP) (Doe et al., 1987; Gulden and Halasz, 1988; Moradi-Araghi et al., 1987; Salunkhe et al., 2021; Wu et al., 2009). For example, NVP and AMPS-rich polymers such as SAV-505/333 and SAV-10 were reported to have excellent thermal stability at 120–140 °C in brines with concentrated divalent cations (Dupuis et al., 2017; Skauge et al., 2022). The viscosity retention ratio of AMPS-rich and NVP-rich polymers prepared in seawater after 1-year exposure at 140 °C were 75 % and 68 %, respectively (Dupuis et al., 2017; Jouenne, 2020). AMPS-rich polymers have been more prevalent during the polymer flooding project than NVP-based and hydrophobic polymers because of their excellent injectivity, reasonable cost and low adsorption/retention (Sandengen et al., 2017, 2018; Seright and Wang, 2022; Seright et al., 2021).

Switching metallic-based crosslinker to organic crosslinkers could also improve the hydrolytic thermal stability of the crosslinked gel. Several commercial organic crosslinker-based gel systems have been developed to reach the goal of high-temperature conformance control, such as H2Zero, HE, and Unogel (Seright). Some commonly used crosslinkers are phenol formaldehyde and its derivatives, which can crosslink acrylamide-based polymers through condensation reaction. For example, Unogel which is a hydroquinone and hexamethylenetetramine crosslinked cationic polymer was proven to have excellent thermal stability at high temperatures (Guo et al., 2022; Hutchins et al., 1996). Field application in New Mexico, and the North Sea showed good results in improving the reservoir conformance (Hutchins et al., 1996; Norman et al., 2006). Daoyi studied the effect of crosslinker types on the gel stability and he found that the gel crosslinked by phenol and hexamethylenetetramine has limited thermal stability compared with gel crosslinker by hydroquinone, resorcinol and hexamethylenetetramine (Zhu et al., 2017). Other crosslinkers such as furan alcohol and melamine are also deployed to crosslink the polymer (Fjelde and Stavland,

1995; Huang and Jones, 1996; Hubbert, 1956). Apart from these aldehyde-based crosslinkers, polyamine, such as trimethylene tetra amine and polyethyleneimine, is a new type of crosslinker that can crosslink acrylamide-based polymers in a wide temperature range. It has been proved that polyethyleneimine can crosslink the polymer from 4.4 to 176 °C, and the crosslinked gels have excellent thermal stability (Ghriga et al., 2019, 2020; Reddy et al., 2012; Vasquez, 2005). For example, the polyethyleneimine crosslinked gel was proved to be stable for over 300 days at 176 °C (Vasquez, 2005). It should be noted that high-temperature resistant gel can also be achieved by using concentrated low molecular weight polyacrylamide and chromium acetate, such as Maraseal gel (Sydansk, 1993, 2000). The high thermal stability was attributed to the dense and highly crosslinked gel network structure. Chitosan is another newly founded crosslinker that is considered to be more environmentally friendly than PEI and aldehyde-based crosslinker. It can also crosslink the acrylamide-based polymers through transamidation reaction. It can crosslink acrylamide-based polymer at temperatures higher than 65 °C, and the gel strength can be manipulated from highly flowing gel to rigid or even ringing gel (Mendes et al., 2023; Shamlooh et al., 2022; Vasquez, 2003). The crosslinked gels also have excellent hydrolytic thermal stability at 121 °C (Vasquez, 2003).

With increasingly stringent environmental regulations, most offshore oil reservoirs have phased out the metallic crosslinkers, polyethyleneimine, and phenol/formaldehyde-based crosslinkers because of their toxicity to aquatic creatures (Reddy and Eoff, 2003). Furthermore, phenol and its derivatives can readily dissolve in crude oil and partition from the gelant into the remaining oil causing drift in gelant composition (Bryant et al., 1997). Therefore, during the gelant injection, the gelation time and gel strength can not be well controlled. Besides, the crosslinking mechanism was based on a reversible condensation reaction, and field test has demonstrated that the phenol-formaldehyde crosslinked gel can de-crosslink at high temperatures generating free phenol and formaldehyde. Chitosan is the only commercially available, environmentally-friendly crosslinker that can produce gels in high-temperature reservoirs (Reddy and Eoff, 2003; Shamlooh et al., 2022). However, chitosan has poor solubility in neutral or alkaline solutions, and precipitation could be observed when the polymer/chitosan mixture was diluted by the alkaline formation water (Shamlooh et al., 2022). Besides, chitosan is a potential bug food and biocide preflush is needed to keep the chitosan from biodegradation (Reddy and Eoff, 2003). Fortunately, our group found that L-Lysine could crosslink polyacrylamide at high temperatures through the transamidation reaction. Lysine is an environmentally friendly material, which is nontoxic to both terrestrial and aquatic organisms and can dissolve in brine readily. For example, the LD<sub>50</sub>(rat) and EC<sub>50</sub>(fish) of lysine and



**Fig. 1.** Core flooding setup.

**Table 2**  
Effect of temperature on the crosslinking behavior.

Time	3 %, Lys-HCl 1 %, 5 % NaCl, pH 5.8–6.4																							
	AN-105						AN-125						SAV-55						SAV-225					
	80	90	100	110	120	130	80	90	100	110	120	130	80	90	100	110	120	130	80	90	100	110	120	130
0/hour	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	
4	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	
6	A	A	A	A	A	D	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	
8	A	A	A	A	C	F	A	A	A	A	A	C	A	A	A	A	A	A	A	A	A	A	A	
10	A	A	A	A	D	G	A	A	A	A	B	D	A	A	A	A	A	A	A	A	A	A	A	
12	A	A	A	A	F	H	A	A	A	A	C	E	A	A	A	A	A	A	A	A	A	A	B	
18	A	A	A	A	G	I	A	A	A	A	D	F	A	A	A	A	A	A	A	A	A	A	D	
22	A	A	A	A	G	I	A	A	A	A	E	G	A	A	A	A	A	A	A	A	A	A	E	
24	A	A	A	D	G	I	A	A	A	A	F	H	A	A	A	A	A	A	A	A	A	B	F	
28	A	A	C	F	G	I	A	A	A	A	F	H	A	A	A	A	A	A	A	A	A	B	F	
32	A	C	D	G	H	I	A	A	A	A	H	I	A	A	A	A	A	A	A	A	A	C	G	
40	A	D	I	I	I	I	A	A	A	D	H	I	A	A	A	A	A	B	A	A	A	A	H	
2/day	A	I	I	I	I	I	A	A	A	F	H	I	A	A	A	A	A	D	A	A	A	A	I	
3	A	I	I	I	I	I	A	A	B	G	I	I	A	A	A	A	C	F	A	A	A	A	I	
5	A	I	I	I	I	I	A	A	D	H	I	I	A	A	A	A	B	D	H	A	A	A	I	
7	A	I	I	I	I	I	A	A	G	H	I	I	A	A	C	D	G	I	A	A	C	D	I	
8	A	I	I	I	I	I	A	A	H	H	I	I	A	A	C	E	G	I	A	A	D	E	I	
10	A	I	I	I	I	I	A	A	I	H	I	I	A	A	D	F	H	I	A	A	E	F	I	
12	A	I	I	I	I	I	A	A	I	I	I	I	A	A	F	H	H	I	A	A	F	G	I	
16	A	I	I	I	I	I	A	A	I	I	I	I	A	A	G	H	H	I	A	A	G	I	I	
20	A	I	I	I	I	I	A	A	I	I	I	I	A	A	H	H	H	I	A	A	G	I	I	
22	A	I	I	I	I	I	A	B	I	I	I	I	A	A	H	H	H	I	A	A	H	I	I	
35	A	I	I	I	I	I	A	C	I	I	I	I	A	A	H	H	H	I	A	A	I	I	I	
40	A	I	I	I	I	I	A	D	I	I	I	I	A	A	H	H	H	I	A	A	I	I	I	
50	D	I	I	I	I	I	A	F	I	I	I	I	A	A	H	H	H	I	A	B	I	I	I	
61	F	I	I	I	I	I	A	I	I	I	I	I	A	A	H	H	H	I	A	D	I	I	I	
80	I	I	I	I	I	I	A	I	I	I	I	I	A	A	H	H	H	I	A	F	I	I	I	
100	I	I	I	I	I	I	A	I	I	I	I	I	A	A	H	H	H	I	A	I	I	I	I	
150	I	I	I	I	I	I	A	I	I	I	I	I	A	A	H	H	H	I	A	I	I	I	I	
200	I	I	I	I	I	I	A	I	I	I	I	I	A	A	H	H	H	I	A	I	I	I	I	
220	I	I	I	I	I	I	A	I	I	I	I	I	A	A	H	H	G	I	A	I	I	I	I	



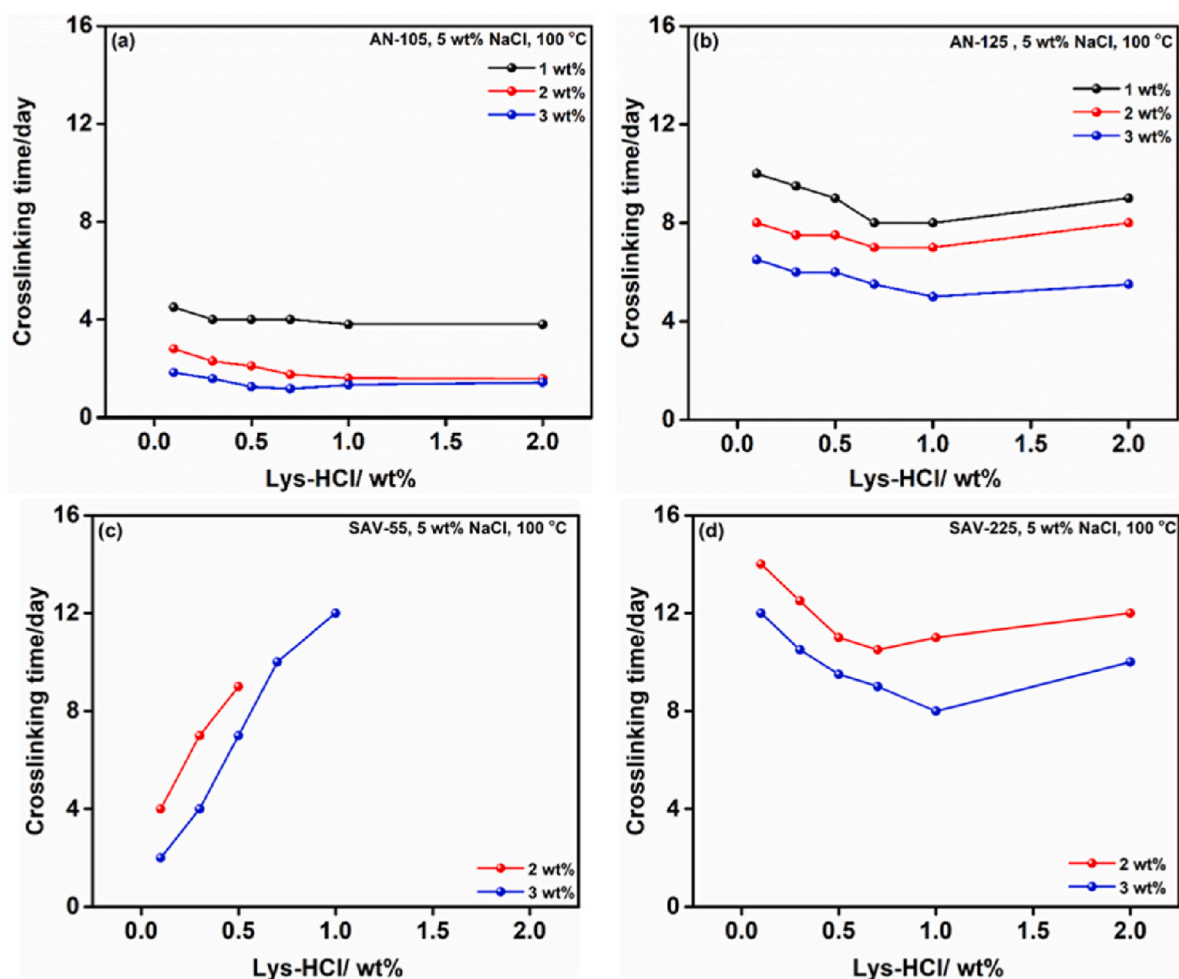


Fig. 2. Effect of polymer and Lys-HCl concentration on the crosslinking time of (a) AN-105, (b) AN-125, (c) SAV-55 based gelant; (d) SAV-225 based gelant at 100 °C.

polyethyleneimine are >10000, >100, 3300 and 1–10 mg/kg, respectively (Song et al., 2023). Lysine has two amine groups and can act as a di-functional crosslinker to build a 3-D network with polyacrylamide and its derivatives at high temperatures. In our previous work, we found that a gelant composed of 7 % of low molecular weight polymer (acrylamide-co-acrylic acid, SNF AB-055) and 1 % of Lysine-monohydrochloride (Lys-HCl) has a crosslinking time of 7 days at 100 °C (Song et al., 2023). However, the crosslinked gel suffers from syneresis extensively when the  $\text{CaCl}_2$  concentration is higher than 0.5 %. For example, the crosslinked gel formed at 100 °C suffered from severe syneresis after 10 days of aging when the  $\text{CaCl}_2$  concentration was 0.5 wt %, and the volume loss was over 60 %.

In the real scenario, high salinity usually comes along with high temperature, and thus the salt resistance of polymer gel must be improved to meet the plugging requirement. Besides, most field applications prefer to use high molecular weight polymers with a relatively low concentration (0.5–3 %) (Moradi-Araghi et al., 1988). Therefore, this work deployed several salt-resistant AMPS-rich high molecular weight acrylamide-based polymers and systematically evaluated the gelation behavior of these polymers and Lys-HCl to provide gelation time, gel strength and thermal stability data for the field application. The effect of Lys-HCl and polymer concentrations, temperature, salinity and pH on the gelation behavior, gel strength and thermal stability were studied. In addition, the plugging efficiency was studied using high-permeability sandstone cores. One thing that should be noted is that we systemically evaluate the effect of  $\text{CaCl}_2$  concentration and temperature on the gels' thermal stability. The testing temperature ranges from 80 to 130 °C, and the  $\text{CaCl}_2$  concentration ranges from 0.5

to 2 %.

## 2. Experimental section

### 2.1. Materials

Lysine monohydrochloride (Lys-HCl) was purchased from Sigma-Aldrich (St. Louis, MO). SNF Floerger kindly donated AN-105 and 125, SAV-10, 28, 37, 55 and 225. Detailed information about these polymers is shown in Table 1 (Seright et al., 2021). The SAV-10 has the highest AMPS content, followed by SAV-28, SAV-37, SAV-55, AN-125 and AN-105. The core flooding test used sandstone cores with a brine permeability of 1600 mD.

### 2.2. Crosslinking time evaluation and thermal stability test

The bottle test method was adopted to determine the crosslinking time and evaluate the thermal stability (Sydansk, 1988). High-pressure glass tubes with temperature-resistant O-rings (USA Sealing, size 110, Material: 80 Shore A AFLAS, BATCH No: 180806003) were deployed to prevent leakage. The crosslinking time was defined as the time when the gelant reached gel code D (Zhu et al., 2019). Factors affecting crosslinking time, such as polymer and crosslinker concentration, gelant pH, salinity and temperature, were evaluated. The pH of the gelant was adjusted by dilute HCl and NaOH solution. A typical gelation time/thermal stability test process is as follows: 20 g of gelant solution was added into a high-pressure glass tube, followed by sparging argon for 20 min before sealing. After that, the glass tube was put into the preheated

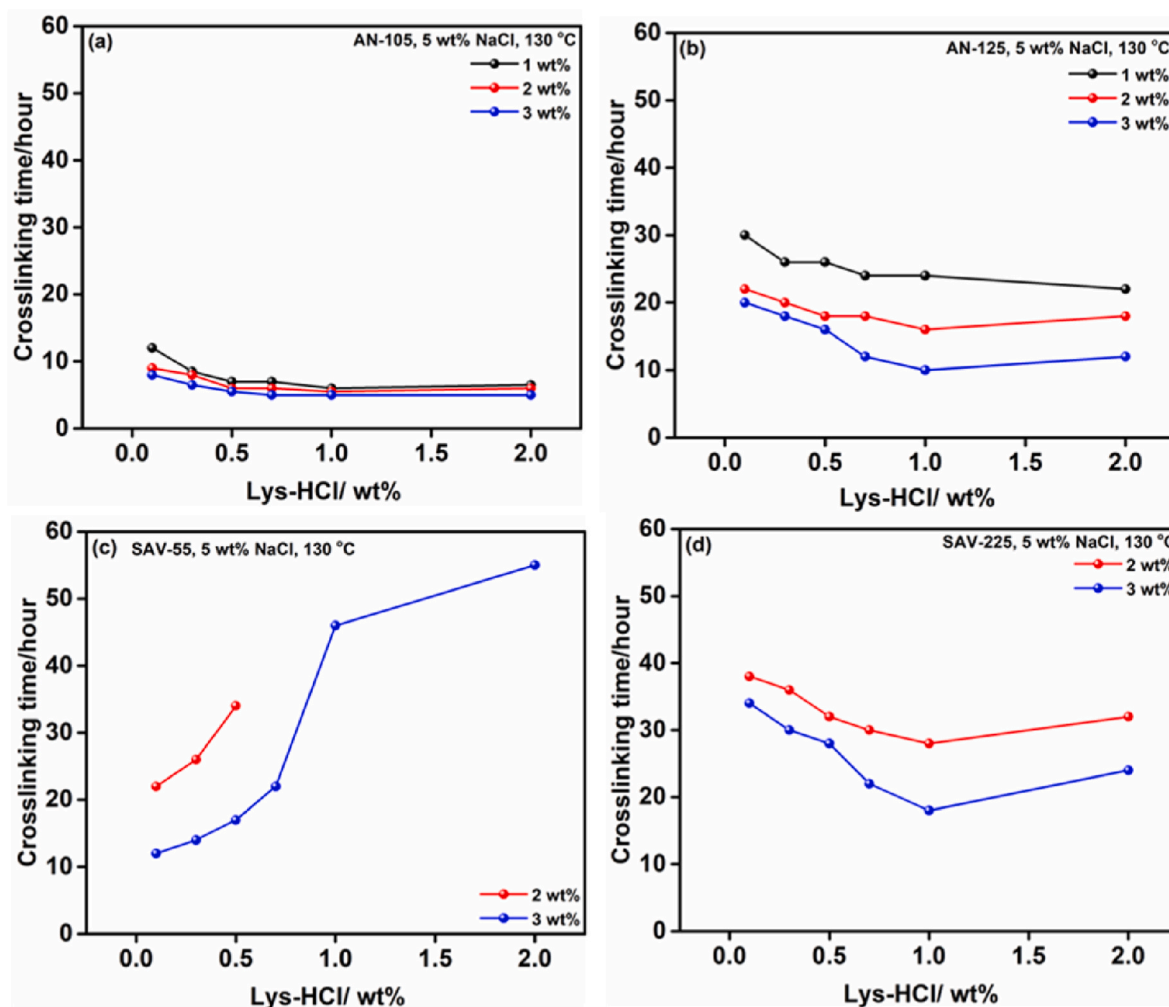


Fig. 3. Effect of polymer and Lys-HCl concentration on the crosslinking time of (a) AN-105, (b) AN-125, (c) SAV-55 based gelant; (d) SAV-225 based gelant at 130 °C.

oven. The glass tube was inverted for inspection periodically. The gel volume and gel code changes were monitored during the gelation and stability test.

### 2.3. Rheology tests

After crosslinking, the bulk gel strength was evaluated via HAAKE MARS III Rheometer using a parallel plate geometry (PP35L Ti L) with a gap of 1 mm. All the rheology tests were carried out in the linear viscoelastic region. In addition, the elastic and viscous modulus of the bulk gel was tested through a time-dependent oscillation experiment at a fixed frequency of 1 Hz and a controlled strain of 1 %.

### 2.4. Thermal stability test

The polymer gel's thermal stability was evaluated in various temperature and salinity conditions. The gelation time is defined as the time when the gelant reaches gel code D according to the Sydansk gel code method. The gel strength was evaluated using the Sydansk code method, and the gel strength and gel volume were monitored during the aging test (Sydansk, 1988). The gel volume and gel code changes were monitored during the stability test.

### 2.5. Thermogravimetric analysis (TGA)

TGA (STA 409 PC, Netzsch, Germany) test was deployed to compare the difference between different gel systems. Samples were scanned

from ambient temperature to 600 °C under a nitrogen atmosphere with a heating rate of 10 °C/min. The crosslinked gel samples were vacuum freeze-dried for 3 days and ground into particles before the test.

### 2.6. Core flooding test

The plugging efficiency was evaluated through the core-flooding test. The polymer and Lys-HCl concentrations were fixed at 3 and 1 %. 5 % NaCl brine was used to prepare the gelant. The experiment setup and core parameters are shown in Fig. 1. First, the gelant was injected into the core with a constant injection rate of 0.5 mL/min till the injection pressure became stable. Afterward, the core was sealed and aged at 130 °C for five days. Then the step-wise breakthrough test was performed to get the breakthrough pressure. Last, after the water breakthrough, the injection rate was changed to 0.1, 0.25, 0.5, 0.75 and 1 mL/min, and the stable pressure was recorded to calculate the  $F_{rr}$ . The residual resistance factor,  $F_{rr}$ , was calculated by the following equations (Hubbert, 1956; Seright, 1995):

$$F_{rr} = \frac{k_{before}}{k_{after}} \quad (1)$$

$$k = \frac{q \times \mu \times L}{A \times \Delta P} \quad (2)$$

Where  $k_{before}$  is the initial core permeability;  $k_{after}$  is the core permeability after the water breakthrough and calculated based on Darcy's

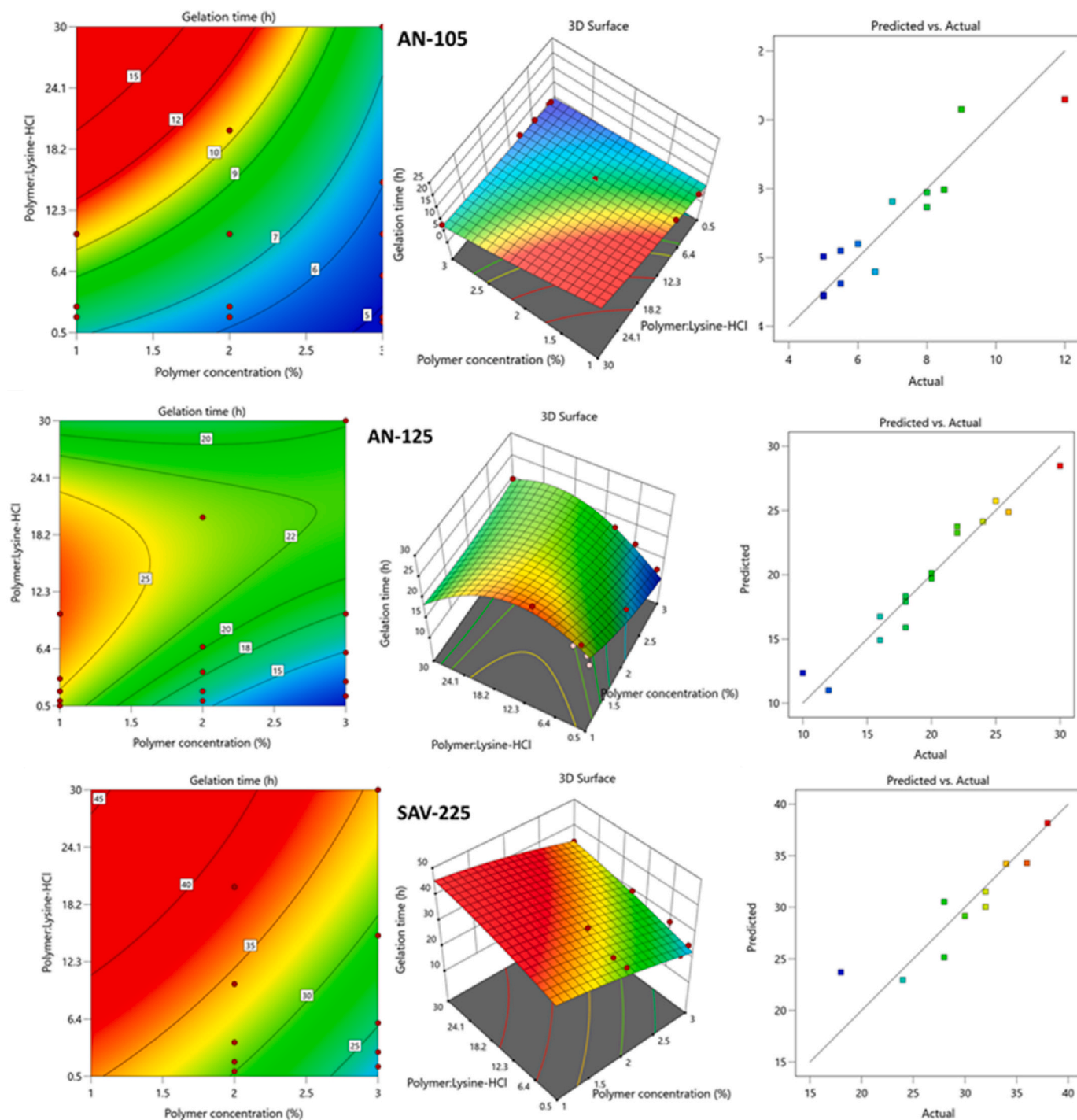


Fig. 4. Central composite design analysis result of AN-105/125 and SAV-225.

law.

### 3. Results and discussion

#### 3.1. Factors affecting the gelation time

##### 3.1.1. AMPS effect

Table 2 shows the crosslinking behavior of AN-105/125, SAV-55 and 255-based gelant at 80, 90, 100, 110, 120 and 130 °C. It should be noted that the AMPS-rich SAV-10/28/37-based gelants failed to reach gel code D during the 200-day gelation test at 80–130 °C, regardless of the polymer and Lys-HCl concentration, pH and salinity. PEI was also

reported to have a compatibility issue when mixed with polymers having high AMPS content due to the strong electrostatic interaction (Juárez et al., 2020). SAV-10 was demonstrated to have excellent thermal stability at high temperatures and high salinity reservoirs owing to the high fraction of AMPS units (>90 mol %) (Jouenne, 2020).

However, the crosslinking mechanism between HPAM and Lys-HCl was attributed to the transamidation reaction, and Lys-HCl is inactive toward the AMPS unit (Song et al., 2023). Besides, the hydrolysis rate of amide groups at high temperatures is relatively fast, and only a trace amount of amide groups might remain after several days of aging at high temperatures (Sandengen et al., 2017). Therefore, considering these two factors, the SAV-10/28/37-based gelant can not reach gel code D due to

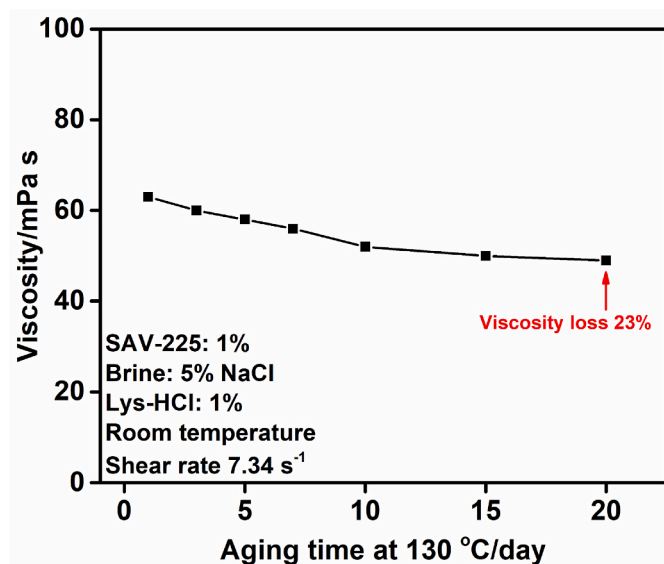


Fig. 5. Gelant viscosity changes at 130 °C.

strong charge repulsion and the limited amount of amide groups. Our result is in good accordance with Aqcheli’s research, they also found that AMPS could delay the crosslinking between PEI and HPAM (Aqcheli et al., 2020, 2021). They also found that the G’ of PEI crosslinked poly (AM-co-AMPS) gel decreases as AMPS content increases which also reveals AMPS negatively affects the crosslinking integrity (Aqcheli et al., 2021).

For other gelant systems, the crosslinking time increases with decreasing temperature, and the crosslinking time is extended

significantly when the temperature is lower than 120 °C. The gelation time of AN-105-based gelant at 130, 120, 110, 100, 90 and 80 °C were 6, 10, 24, 32, 40 h and 50 days, respectively. However, for the gelation test at 80 °C, AN-125, SAV-55 and SAV-225 gelants failed to form any visible gels or noticeable viscosity increase during the 200-day aging test. The AN-125 and SAV-225 gelants have a much longer crosslinking time than the AN-105 gelant, especially at 90 °C. The crosslinking time of the AN-125 and SAV-225 gelant at 90 °C were 40 and 61 days, respectively, whereas the AN-105 gelant had a crosslinking time of 40 h. This is because AN-125 and SAV-225 have a significant fraction of crosslinking inactive moieties such as AMPS and NVP. Besides, AMPS and NVP have bulky pendant groups, further hindering inter-chain crosslinking (Vasquez, 2005). In addition, SAV-55 is less reactive than AN-125 and SAV-225, and it cannot form gels at 90 °C.

Compared with our previous research, the crosslinking time of AN-125 and SAV-225 gelant is similar to the AB-055 gelant, but the crosslinking time of AN-105 gelant is much shorter than other gel systems. For example, at 100 °C, the crosslinking time of AB-055, AN-125 and SAV-55, SAV-225 gelant ranges from 5 to 14 days, and the AN-105 gelant has a crosslinking time of 32 h (Song et al., 2023). SAV-55 and SAV-225 have a similar amide ratio, and SAV-225 is an AM-AMPS-NVP terpolymer. However, SAV-55 has a longer gelation time than SAV-225. For example, the gelation time of SAV-55 and 225 at 130 °C is 2 days and 18 h, respectively. Therefore, it can be concluded that the AMPS unit has a more significant effect in delaying the gelation speed than the NVP unit.

### 3.1.2. Polymer and Lys-HCl concentration effect

We also studied the effect of polymer and Lys-HCl concentration on the crosslinking time at 100 and 130 °C, as shown in Figs. 2 and 3. The central composite design method (Salehi et al., 2012) was deployed to further analyze the effect of polymer concentration and polymer to

Table 3  
Effect of salinity on the crosslinking behavior at 100 °C.

Time	Polymer 3 %, Lys-HCl 1 %, 100 °C, pH 5.8–6.4															
	AN-105				AN-125				SAV-55				SAV-225			
	NaCl/wt %		CaCl <sub>2</sub> /wt % 5 wt % NaCl		NaCl/wt %		CaCl <sub>2</sub> /wt % 5 wt % NaCl		NaCl/wt %		CaCl <sub>2</sub> /wt % 5 wt % NaCl		NaCl/wt %		CaCl <sub>2</sub> /wt % 5 wt % NaCl	
0/hour	5	0.5	1	2	5	0.5	1	2	5	0.5	1	2	5	0.5	1	2
24	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A
28	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A
32	B	D	D	D	A	A	A	A	A	A	A	A	A	A	A	A
36	D	F	E	E	A	A	A	A	A	A	A	A	A	A	A	A
40	G	G	G	H	A	A	A	A	A	A	A	A	A	A	A	A
48	I	I	H	I	A	A	A	A	A	A	A	A	A	A	A	A
88	I	I	I	I	A	A	A	C	A	A	A	A	A	A	A	A
96	I	I	I	I	B	A	B	D	A	A	A	A	A	A	A	A
104	I	I	I	I	C	B	D	F	A	A	A	A	A	A	A	A
112	I	I	I	I	C	D	E	G	A	A	A	A	A	A	A	D
120	I	I	I	S	D	F	F	H	A	A	A	C	A	A	A	F
6/day	I	I	I		F	G	G	I	A	A	A	D	A	A	A	G
7	I	I	I		F	I	I	I	A	A	B	F	B	C	D	H
8	I	I	I		G	I	I	I	A	C	D	G	C	D	F	I
9	I	I	I		H	I	I	I	C	C	E	H	D	F	H	I
10	I	I	I		H	I	I	I	C	D	F	H	E	G	I	I
12	I	I	S		I	I	I	I	D	F	G	H	E	H	I	I
14	I	I			I	I	I	I	F	G	H	H	G	I	I	I
16	I	I			I	I	I	S	I	H	H	H	G	I	I	I
20	I	I			I	I	I		G	H	H	H	G	I	I	I
22	I	I			I	I	I		H	H	H	H	G	I	I	I
30	I	I			I	I	I		H	H	H	H	I	I	I	I
50	I	I			I	I	I		H	H	H	H	I	I	I	I
70	I	I			I	I	S		H	H	H	H	I	I	I	I
100	I	I			I	I			H	H	H	H	I	I	I	I
150	I	I			I	I			H	H	H	H	I	I	I	I
200	I	I			I	I			H	H	H	H	I	I	I	I
220	I	I			I	I			H	H	H	H	I	I	I	I



**Table 4**  
Effect of salinity on the crosslinking behavior at 130 °C.

Time	Polymer 3 %, Lys-HCl 1 %, 130 °C, pH 5.8–6.4																	
	AN-105				AN-125				SAV-55				SAV-225					
	NaCl/wt %		CaCl <sub>2</sub> /wt % 5 wt % NaCl		NaCl/wt %		CaCl <sub>2</sub> /wt % 5 wt % NaCl		NaCl/wt %		CaCl <sub>2</sub> /wt % 5 wt % NaCl		NaCl/wt %		CaCl <sub>2</sub> /wt % 5 wt % NaCl			
0/hour	5	0.5	1	2	5	0.5	1	2	5	0.5	1	2	5	0.5	1	2		
4	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A	A		
6	D	C	C	D	A	A	A	A	A	A	A	A	A	A	A	A		
8	F	D	D	G	C	B	C	C	A	A	A	A	A	A	A	A		
10	G	F	G	H	D	C	C	D	A	A	A	A	A	A	A	A		
12	H	H	H	I	E	D	D	F	A	A	A	A	B	B	C	C		
14	I	I	I	I	E	F	E	H	A	A	B	D	C	C	D	D		
16	I	I	I	I	F	G	G	I	A	C	D	F	C	C	E	F		
18	I	I	I	I	F	H	H	I	A	A	E	G	D	C	F	G		
20	I	I	I	I	G	H	I	I	A	D	F	H	E	D	G	H		
24	I	I	I	I	H	I	I	I	A	E	G	H	F	F	G	I		
32	I	I	I	I	I	I	I	I	A	F	H	H	G	H	H	I		
40	I	I	I	I	I	I	I	I	B	G	H	H	H	H	I	I		
2/day	I	I	S	S	I	I	I	I	D	G	H	H	I	I	I	I		
3	I	S			I	I	I	S	F	G	H	H	I	I	I	I		
4	I				I	I	S		H	H	I	H	I	I	I	I		
5	I				I	S			H	H	I	H	I	I	I	I		
6	I				I				I	I	I	H	I	I	I	I		
10	I				I				I	I	I	H	I	I	I	I		
12	I				I				I	I	I	S	I	I	I	I		
17	I				I				I	I	I	I	I	I	I	I		
20	I				I				I	I	I	I	I	I	I	S		
40	I				I				I	I	I	I	I	I	I			
50	I				I				I	I	S	I	I	I	I			
68	I				I				I	I		I	I	I	S			
100	I				I				I	I		I	I	I				
150	I				I				I	I		I	I	I				
200	I				I				I	I		I	I	I				
220	I				I				I	I		I	I	I				

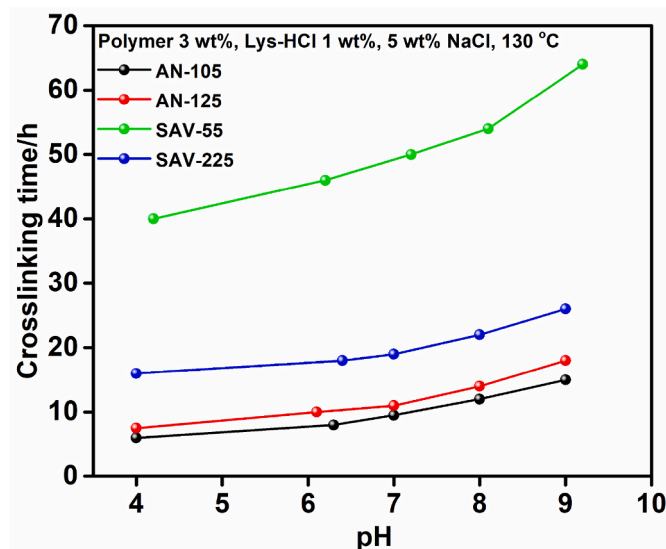


Fig. 6. Effect of gelant pH on the crosslinking time.

lysine-HCl weight ratio on the gelation time at 130 °C, as depicted in Fig. 4. The crosslinking time can be shortened by increasing temperature, polymer feeding amount and using polymers with high amide content. The crosslinking time at 100 °C ranges from 28 h to 14 days, while the gelation time range was significantly shortened to 5–55 h at 130 °C. To be more precise, at 130 °C, the crosslinking time of AN-105 gelant ranges from 5 to 12 h, while the crosslinking time of AN-125, SAV-55 and SAV-225 gelants are 10–30, 12–55 and 18–38 h, respectively. For the AN-105/125 gels, the crosslinking time decreases with

increasing Lys-HCl concentration. But in the case of SV-225 gelant, the crosslinking rate was somehow delayed when the Lys-HCl concentration was higher than 1 %. The gelation time of the SAV-55-based gelant increases with Lys-HCl concentration, which is different from the previous gelants. In addition, SAV-55 and 225 gels failed to reach gel code D when the polymer concentration was 1 %, regardless of the Lys-HCl concentration, and gelants composed of 2 % SAV-55 cannot form gels when the Lys-HCl concentration is higher than 0.7 %.

In the case of gelant SAV-225 (1 % polymer and 0.05 % of Lys-HCl, 5 % NaCl), which failed to form visible gels at 130 °C, we monitored the viscosity changes for 20 days, as shown in Fig. 5. We prepared 140 mL of gelant and packed in the 7 glass tubes after sparging argon for 20 min. We took one sample out from the oven each time to test the viscosity at room temperature. The SAV-225 has relatively good thermal stability at high temperatures (Gaillard, 2014). The viscosity decreases as the aging time increases, and the viscosity loss is 23 % after 20 days of aging. Therefore, the lower critical polymer concentration to form gels for the SAV-225-based gel system is between 1 and 2 %.

### 3.1.3. Salinity

We also studied the effect of salinity on the crosslinking time at 100 and 130 °C. The polymer and Lys-HCl concentrations were fixed at 3 and 1 %, respectively. The CaCl<sub>2</sub> concentration ranges from 0 to 2 %. Tables 3 and 4 show the impact of CaCl<sub>2</sub> concentration on the crosslinking time. The crosslinking time gradually decreases as the CaCl<sub>2</sub> concentration increases. At 100 °C, the crosslinking time of AN-105/125, SAV-55/225 gelants decreases from 32 to 28 h, 120 to 88 h, 10 to 5 days, and 8 to 4.4 days, respectively, as the CaCl<sub>2</sub> concentration increases from 0 to 2 %. This is because Ca<sup>2+</sup> could efficiently compress the polymer chains and reduce the chain repulsion, facilitating inter-chain crosslinking.

However, the stability of the crosslinked gel decreases sharply with

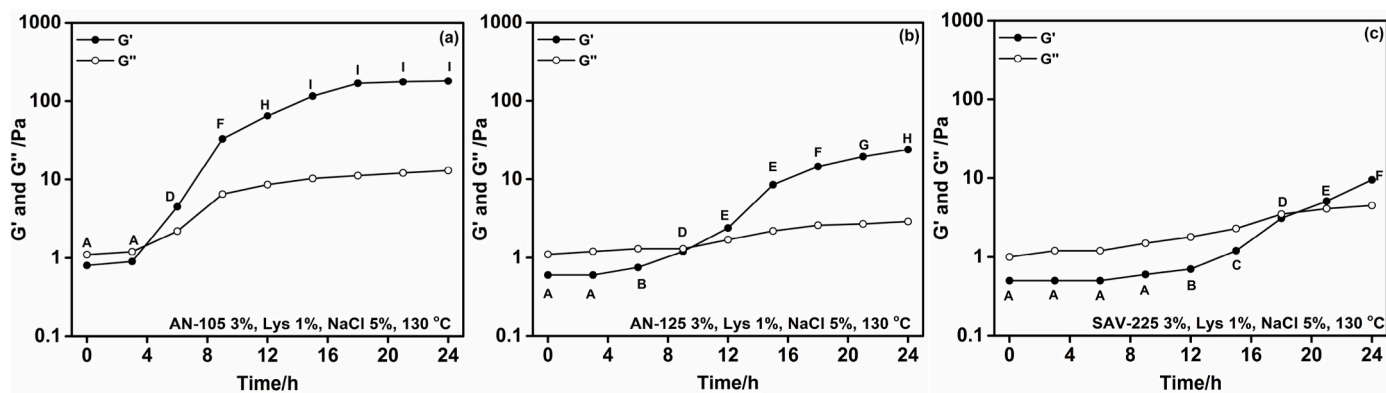


Fig. 7. Gel code and strength changes.

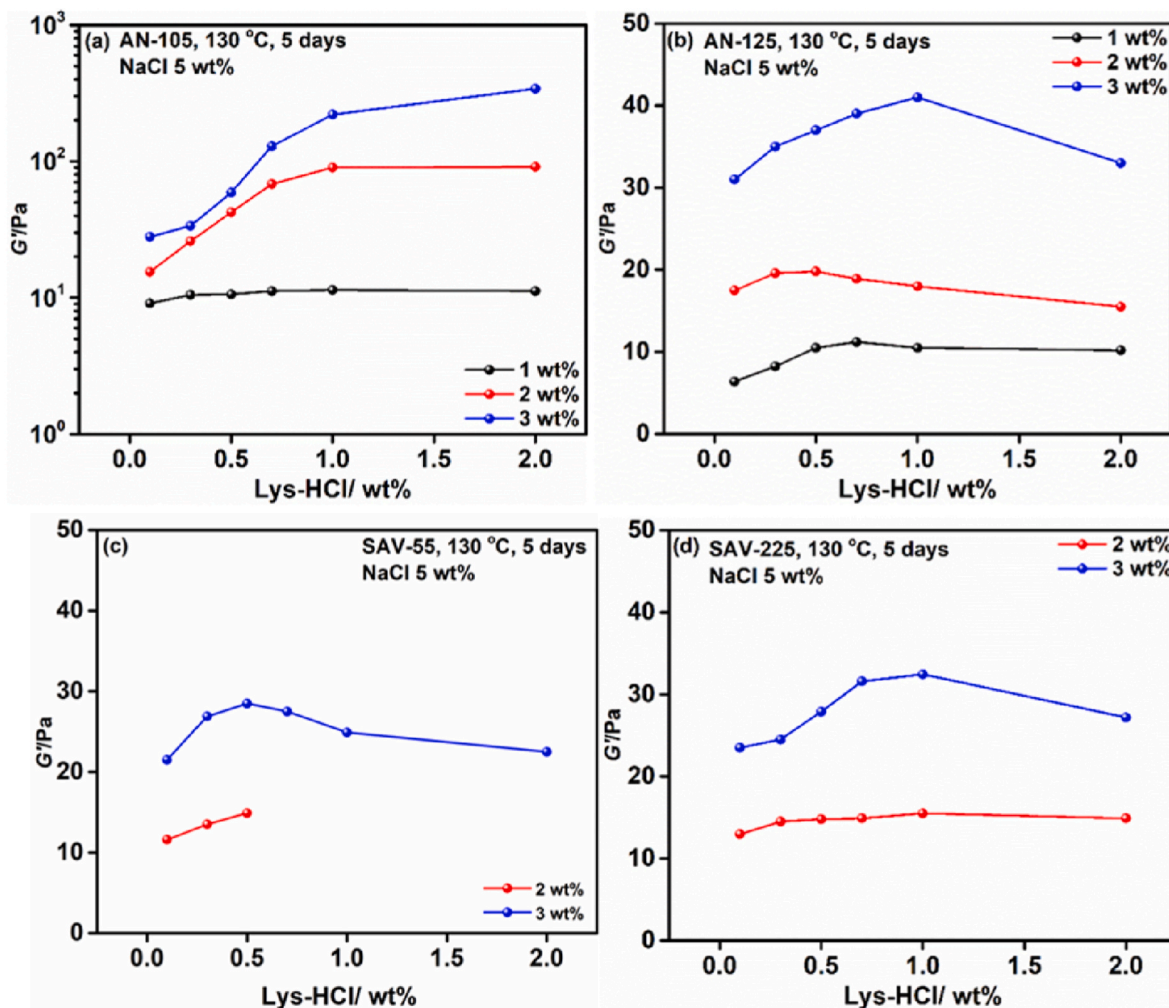


Fig. 8. Effect of polymer and Lys-HCl concentration on the gel strength (a) AN-105, (b) AN-125, (c) SAV-55, (d) SAV-225.

the rise in  $\text{CaCl}_2$  concentration, especially at 130 °C. This is because the  $\text{Ca}^{2+}$  can bond the carboxylate groups and negatively affect the phase stability of the polymer gel. The carboxylate groups may come from the hydrolysis product of AM and AMPS, and Lys-HCl. Therefore, the Lys-HCl-based gels are very sensitive to divalent cations at high temperatures.

3.1.4. pH

Fig. 6 shows the effect of gelant pH on the crosslinking time at

130 °C. Similar to our previous research, decreasing the solution pH can shorten the crosslinking time (Song et al., 2023). Gelant with high pH need a much longer time to reach gel code D because of the high hydrolysis rate of amide groups at high pH conditions. SAV-55, with the longest gelation time, is extremely sensitive to pH changes than other gelants. The gelation time was almost increased by 1.6 times as the gelant pH increased from 4 to 9.

We further studied the relationship between pH and salinity through the CCD method, and the results were analyzed using “Design Expert”

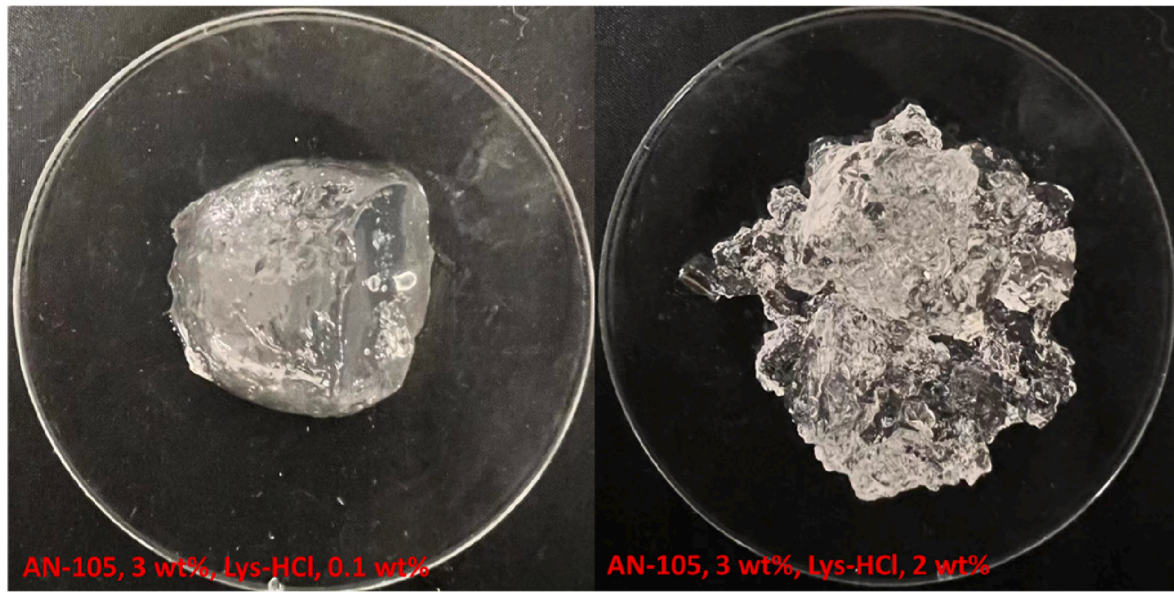


Fig. 9. The appearance of crosslinked gel with different Lys-HCl feeding amount.

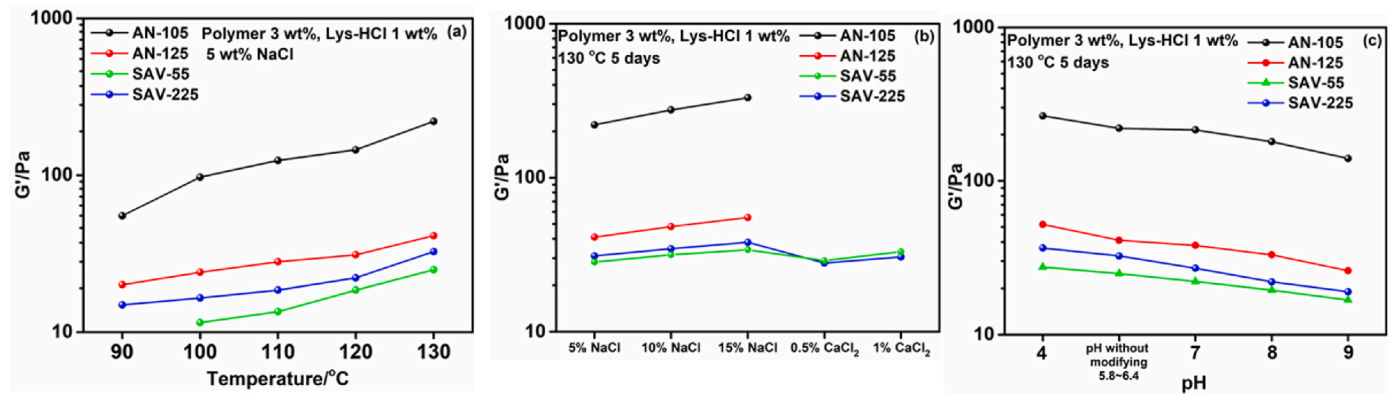


Fig. 10. Effect of (a) temperature, (b) salinity, (c) pH on the gel strength.

software with a quadratic model (Salehi et al., 2012). This model successfully predicts the gelation time and generates the following equations, Where  $GT$  is gelation time,  $A$  is pH, and  $B$  is salinity.

$$GT_{AN-105} = 6.36 + 2.94 \times A - 1.9 \times B - 1.46AB + 1.41A^2 + 0.625B^2 \quad (3)$$

$$GT_{AN-125} = 8.82 + 4.12 \times A - 2.03 \times B - 0.9534AB + 1.26A^2 + 0.258B^2 \quad (4)$$

$$GT_{SAV-225} = 42.55 + 10.19 \times A - 5.04 \times B - 0.8686AB + 3.63A^2 + 0.1277B^2 \quad (5)$$

### 3.2. Factors affecting the gel strength

In this section, the elastic modulus of the crosslinked gel was measured to evaluate the gel strength changes. We assessed the effect of several factors on the gel strength, such as polymer and crosslinker concentration, gelant pH, salinity and temperatures. To study the effect of polymer and Lys-HCl concentration, pH and salinity on the gel strength, we aged the gelant at 130 °C for five days before the rheology test.

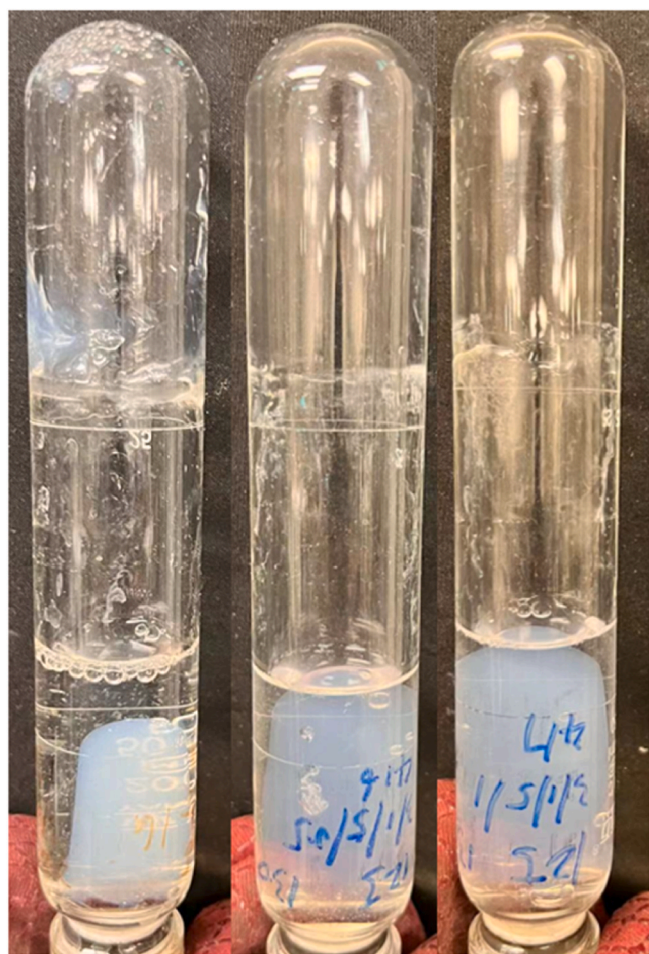
Fig. 7 illustrates the relationship between gel code and gel strength changes during the gelation test at 130 °C. For each gelant system, we prepared eight samples, subjecting all of them to a 130 °C oven.

Subsequently, we extracted one sample every 3 h to assess the elastic and viscous moduli. It is widely acknowledged that the point of cross-over between  $G'$  (elastic modulus) and  $G''$  (viscous modulus) signifies the establishment of three-dimensional (3-D) network structures, and the cross-over time for AN-105, AN-125 and SAV-225 gel systems are 3–6, 9–12 and 18–21 h, respectively (Grillet et al., 2012). Notably, the gelation time determined via the rheology test aligns well with that obtained from the bottle test method. and the existence of a cross-over point confirmed the formation of a 3-D network structure.

As shown in Fig. 8, the gel strength increases with polymer concentration. The gel strength increases with Lys-HCl concentration in the case of the AN-105 gel system. An abrupt increase in elastic modulus can be observed when the Lys-HCl was higher than 0.7 %. However, the crosslinked gel becomes more brittle as the increase of Lys-HCl feeding amount. Fig. 9 shows the gel appearance of AN-105 gels with 0.1 and 2 % of Lys-HCl. The phase angle of gels crosslinked by 0.1 and 2 % Lys-HCl is 24 and 2.3°, respectively, meaning the gels gradually become rigid and lose elasticity as the Lys-HCl increases. We failed to recover the crosslinked gel from the tube as a bulk gel when the Lys-HCl feeding amount was 2 %. The crosslinked gel is very brittle and can easily be broken into small particles.

In the case of AN-125 and SAV-55/225 gelants, the gel strength first increases with Lys-HCl concentrations, but after reaching a certain point, the gel strength decreases as the Lys-HCl concentration increases.





**AN-105, 3 wt%, AN-125, 3 wt%, AN-125, 3 wt%,  
Lys-HCl, 1 wt% Lys-HCl, 1 wt% Lys-HCl, 1 wt%  
0.5 wt% CaCl<sub>2</sub> 0.5 wt% CaCl<sub>2</sub> 1 wt% CaCl<sub>2</sub>  
130 °C, 3 days 130 °C, 5 days 130 °C, 4 days**

Fig. 11. Poor phase stability of AN-105/125 based gels at 130 °C.

The optimal Lys-HCl concentration for 3 % of AN-125 and SAV-55/225 are 0.7–2, 0.3–0.7 and 0.7–1 %, respectively. One possible reason for this is that intra-crosslinking plays the dominant role when excessive Lys-HCl is added to the gelant, deteriorating the crosslinking density and network integrity. This phenomenon is extremely obvious when using AMPS-rich polymers. For example, the SAV-10/28/37-based gelants failed to reach gel code D at 130 °C, regardless of the Lys-HCl and polymer concentration, pH and salinity. The limited amount of crosslinking points (amide group) in AMPS-rich polymer can be consumed rapidly by the excessive Lys-HCl, prohibiting inter-chain crosslinking.

Fig. 10 shows the effect of temperature, salinity and pH on the gel strength. Similar to our previous research, the gel strength increases with temperature and salinity. While pH negatively affects the gel strength. AN-105 gelant, with the highest molecular weight and amide group density, has the highest gel strength among all the testing conditions, followed by AN-125, SAV-225 and SAV-55. To be more precise AN-105 gel is 6–8 times stronger than SAV-55/225, and 5–7 times stronger than AN-125 gels. The gel strength of AN-105 gel at 90, 100, 110, 120, and 130 °C was 55, 97, 124, 145, and 220 Pa, respectively. While the gel strength of SAV-55 at 100, 110, 120, and 130 °C was 11.5, 13.4, 18.6 and 24.9 Pa, respectively.

It should be noted that AN-105 and 125-based gels are not stable in 0.5 % CaCl<sub>2</sub> brine at 130 °C, and severe dehydration was observed after

five days of aging, as shown in Fig. 11. Therefore, we only tested the gel strength in different NaCl solutions. The gel strength of AN-105/125 increases significantly with salinity, but SAV-55-based gel is relatively insensitive to salinity owing to the excellent salt resistance endowed by the high AMPS content. In addition, the gel strength of SAV-55 only fluctuates in a small range as pH increases from 4 to 9. While, AN-105/125 is very sensitive to pH, and the gel strength decreases significantly as pH increases.

### 3.3. Thermal stability

The hydrolytic thermal stability test lasted for over 220 days. We define the lifetime of the crosslinked gel as the time when the syneresis ratio reaches 10 %. The effect of temperature and salinity on the thermal stability of gels was evaluated. Fig. 12 shows the effect of temperature and salinity on the thermal stability of the polymer gels at different temperatures. The polymer and Lys-HCl concentrations were fixed as 3 and 1 % for the AN-105/125 and SAV-225. The Lys-HCl concentration was reduced to 0.5 % in the case of SAV-55 gels. All these gels are transparent and stable in 5 % NaCl brine after 220 days of aging at 100 and 130 °C. HPAM, even has experienced severe hydrolysis, can still maintain the solution phase stability in deionized water or low salinity brines at high temperatures (Doe et al., 1987; Moradi-Araghi et al., 1987; Seright et al., 2010, 2021).

The upper critical safe application temperature of AN-105/125, SAV-55/225 gelants in the presence of 0.5 % CaCl<sub>2</sub> are 100 and 130 °C, respectively. This is because the divalent cations play the most dominant role in the polymer solution's thermal and phase stability at high temperatures (Doe et al., 1987; Moradi-Araghi et al., 1987; Seright et al., 2021). The hydrolysis rate of the amide group under high temperature is high and the hydrolysis product carboxylate could be chelated by Ca and generate less soluble salt, Ca and carboxylate domain, which in macro-view is the gel color changes from transparent to white and syneresis (DiGiacomo, 1983). AMPS can delay the hydrolysis rate of the amide group but it is still vulnerable to hydrolysis at high temperatures, and it also generates carboxylate groups after hydrolysis (Guo et al., 2022; Sandengen et al., 2018). In addition, the crosslinker Lys-HCl also introduces a large number of carboxylate groups, making the gels more sensitive to divalent cations than gels crosslinked by other organic crosslinkers such as PEI and phenolic resins. The stability of the polymer is significantly improved when the AMPS content is higher than 70 mol % (Sandengen et al., 2018). Therefore, SAV-55-based polymer gels perform better than AN-105/125 gel at 130 °C in 0.5 % CaCl<sub>2</sub>. Besides, NVP is hydrolysis resistant and can also inhibit the hydrolysis of amide groups (Doe et al., 1987; Moradi-Araghi et al., 1987; Sandengen et al., 2017). Therefore, the SAV-225 gels also have a relatively good performance at high temperatures. Fig. 13 presents the TGA analysis results of two crosslinked gel samples with varying AMPS content. The weight loss up to 220 °C was caused by the free water loss, and the weight loss between 200 and 340 °C was ascribed to the intra/intermolecular imidization (Van Dyke and Kasperski, 1993). The weight loss at temperatures higher than 340 °C was attributed to chain degradation. Besides, the weight loss of AMPS-rich polymer AN-125 gel is lower than the AN-105 gel, and the weight loss of AN-125 gel and AN105 gel are, 36.2 % and 33.3 % respectively (Ullah et al., 2019). It has been proved that adding monomers with a bulky group such as AMPS and NVP, can increase the degradation activation energy, and thus improve the thermal stability of polymer gels (Zhang et al., 2022). The TGA test conclusively showcased the superior thermal stability of the AN-125 gel over the AN-105 gel, and the addition of AMPS unit can improve the thermal stability of the crosslinked gel (Aqcheli et al., 2020).

### 3.4. Core flooding test

The sandstone cores with a permeability of 1600 mD were used to test the gelant plugging efficiency. AN-125 has similar thermal stability

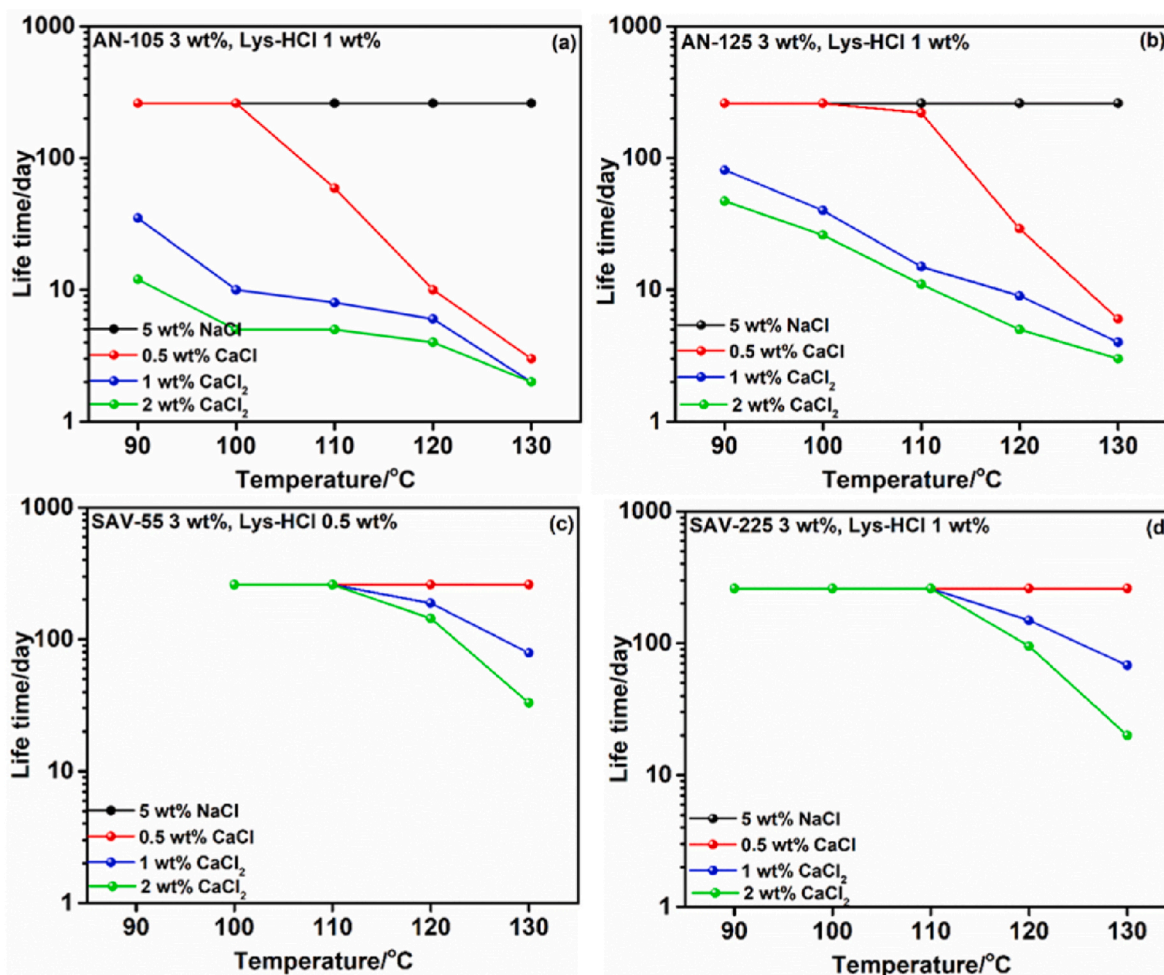


Fig. 12. Effect of temperature and salinity on the thermal stability of (a) AN-105 gels, (b) AN-125 gels, (c) SAV-55 and (d) SAV-225 gels.

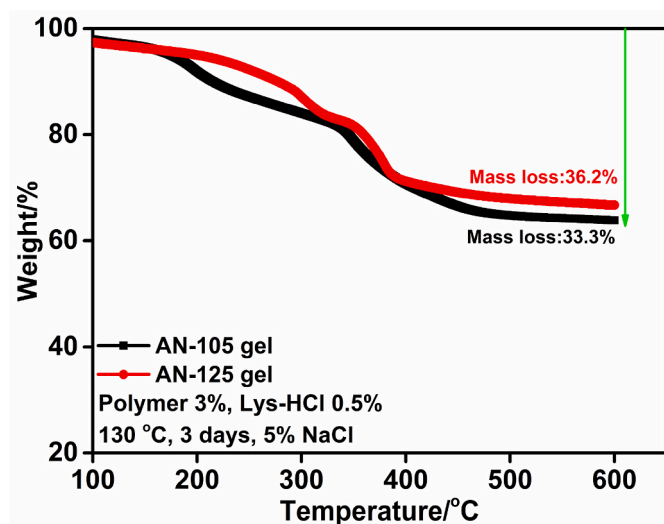


Fig. 13. TGA analysis results of crosslinked gels.

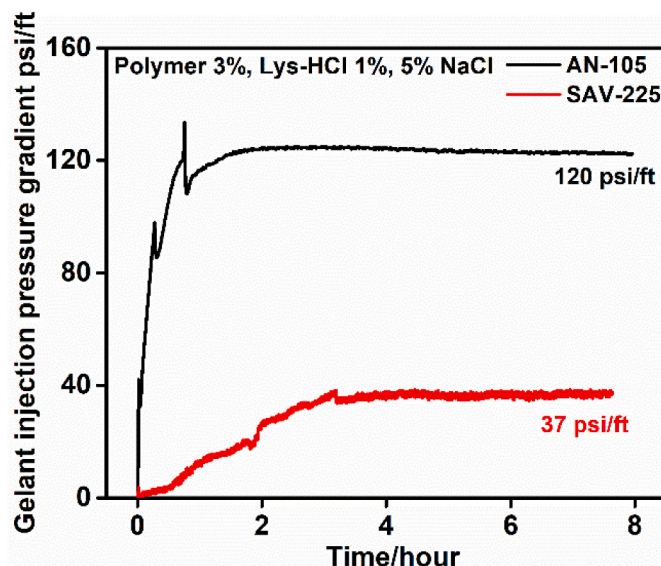


Fig. 14. Injection pressure gradient.

to AN-105 but is much more expensive than AN-105. Therefore, considering the gelation speed, gel strength and stability, we tested the plugging efficiency of AN-105 and SAV-225 gels.

Fig. 14 shows the gelant injection pressure gradient and the stable injection pressure gradient of AN-105 and SAV-225 gelants were 120

and 37 psi/ft, respectively. Fig. 15 (a) shows the breakthrough pressure gradient of different gel systems. AN-105 gel has higher water breakthrough pressure than SAV-225 gel, and the water breakthrough pressure gradient of AN-105 and SAV-225 gelant were 196 and 106 psi/ft,



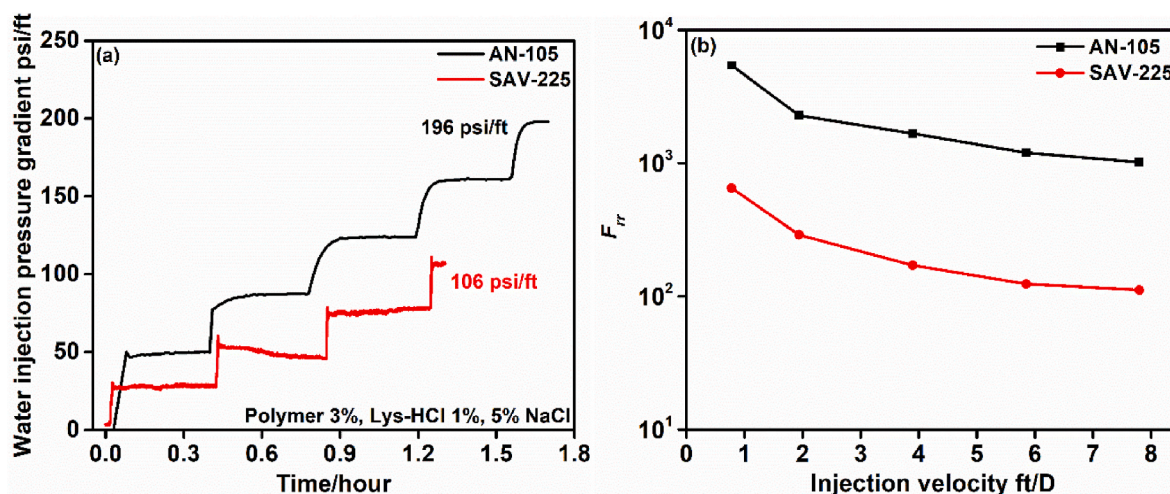


Fig. 15. (a) Water breakthrough test (b)  $F_{rr}$  test after the water breakthrough.

respectively. Fig. 15 (b) shows the  $F_{rr}$  data after the water breakthrough. The resistance factor of AN-105 gel is 3.3 times higher than the SAV-225 gel. For the AN-105 gel system, the  $F_{rr}$  decreases from 5447 to 1017 as the flow rate increases from 0.1 to 1 mL/min. However, the  $F_{rr}$  of SAV-225 gel only ranges from 111 to 650. Both gel systems can reduce the rock permeability to several millidarcys, and the plugging performance is comparable to the commonly used Marcit gel system (Sydansk and Argabright, 1987), but lower than the concentrated low molecular AB-055/lysine system (Song et al., 2023). AN-105 gel has better plugging performance than SAV-225 gelant in terms of the water breakthrough pressure and residual resistant factor. Combining the thermal stability and plugging efficiency, AN-105 gel can be applied to medium-temperature reservoirs (90–110 °C) with  $\text{CaCl}_2$  concentrations lower than 0.5 %. SAV-225 can be applied to high-temperature reservoirs (100–130 °C) with  $\text{CaCl}_2$  concentrations lower than 0.5 %.

#### 4. Conclusion

This work demonstrated that the L-Lysine monohydrochloride could crosslink several commercial high molecular weight acrylamide-based polymers. Lysine as an environmentally friendly crosslinker is a promising product to replace the current widely used crosslinkers such as Cr (III) and phenolic resins. We found that the crosslinking time and gel strength are closely related to the polymer and crosslinker concentration, polymer composition, pH, salinity and temperature. The crosslinking time decreases with the increase in temperature, salinity, polymer concentration, and the percentage of the amide group in the polymer chain. Besides, the crosslinking time can be extended by increasing gelant pH and using polymers with high AMPS concentrations. All the crosslinked polymer gels have excellent long-term thermal stability at 90–130 °C in 5 % NaCl. SAV-55 and 225 gels can be stable at 130 °C for over 200 days in 0.5 %  $\text{CaCl}_2$ . AN-105 gel has better plugging performance than SAV-225 gel, and both gel systems can decrease rock permeability by more than 99 %. Lysine crosslinked gel exhibits considerable potential for high-temperature reservoir conformance control.

However, the Lysine and polyacrylamide-based gel system falls into the category of in-situ gel, which shares the same drawbacks as the commonly used in-situ gels. The gelation process might also be sensitive to shearing, pH changes, adsorption of polymer and crosslinker, and the reaction with minerals. All these factors significantly increase the difficulty in controlling the gelation time, and the viscous gelant may also penetrate the low permeability oil zone causing unwanted plugging and severe formation damage. Further research is imperative to ensure successful field applications. This includes understanding the

transportation and adsorption behavior of the gelant in both sandstone and carbonate formations, as well as determining the lower critical gelant concentrations through bottle tests and core-flooding experiments. This is because the lysine and polymer cannot move at the same pace in the rock causing drift in gelant composition, and the changes in gelant composition may affect the gelation time, gel strength and cause insufficient crosslinking or over crosslinking which eventually affects the long-term thermal stability. More core flooding test is needed to see how far the gelant can penetrate while maintaining the crosslinking ability, reasonable crosslinking density, gel strength, thermal stability, and plugging performance.

#### CRediT authorship contribution statement

**Tao Song:** Writing – review & editing, Writing – original draft, Validation, Resources, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Xuyang Tian:** Investigation. **Baojun Bai:** Writing – review & editing, Project administration, Funding acquisition. **Yugandhara Eriyagama:** Investigation, Formal analysis. **Mohamed Ahdaya:** Methodology, Investigation. **Adel Alotibi:** Methodology, Investigation. **Thomas Schuman:** Writing – review & editing, Investigation.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Data availability

Data will be made available on request.

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