

Rheology and Polymer Flooding Characteristics of Partially Hydrolyzed Polyacrylamide for Enhanced Heavy Oil Recovery

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ABSTRACT: Polymer flooding characteristics of partially hydrolyzed polyacrylamide (HPAM) solution with the addition of NaOH were examined in homogeneous glass-bead packs. The heavy oil recovery in unconsolidated sandstone formations by applying the alkali-polymer flooding was observed. Experimental results showed that HPAM solution was sensitive to temperature, salinity, and alkali, finding that alkali-polymer solutions are more effective in improving viscosity than conventional polymer solutions. The solution of 0.5 wt % NaOH mixed with 1500 ppm HPAM (12 mol % hydrolysis degree) was found to be the optimal choice, which gives rise to the highest viscosity on the rheological characterization. Flood tests using the alkali-polymer solution showed an increase in oil recovery by 30% over water-flooding when the water-cut reached 95%, indicating that alkali-polymer could be more effective in improving sweep efficiency than polymer flood. © 2012 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* 000: 000–000, 2012

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INTRODUCTION

As the production from conventional oil reservoirs starts declining, the need for heavy oil development processes becomes increasingly important. Polymers have been successfully used in enhanced oil recovery (EOR) as flooding agents for a long time in the commercially proven polymer flooding technology, in which the aqueous polymer solution as a displacement fluid stabilizes a water–oil interface. As a main mechanism for polymer flooding, a water-soluble polymer is added into the flooding water, resulting in the increase of its shear viscosity. Three potential ways by which a polymer flooding makes the oil recovery process more efficient are generally accepted: (1) the effects of polymers on fractional flow, (2) decrease in the water/oil mobility ratio, and (3) diversion of injected water from zones that have been swept.¹ Polymers have a variety of applications in the petroleum industry because of their remarkable ability to increase sweep efficiency and to decrease mobility ratio. This process may be applicable to many reservoirs. Field tests also indicate that flooding with low-concentration partially hydrolyzed polyacrylamide (HPAM) solutions has a potential of commercial application to various fields.^{2,3} However, it has been also reported by many laboratory studies and field tests that polymer flooding is not recommended for the viscous oil having a viscosity of over 200 mPa s.⁴ Mobility reduction is one of the critical parameters in polymer flooding. Viscous fingering

during polymer flooding of heavy oil leaves large amount of oil in the reservoir untouched.^{5,6} Moreover, both technical and economic factors restrict the application of polymer flooding to heavy oil reservoirs. On the other hand, high level screening criteria shows that the polymer flood technology implies greater potential than thermal, solvent or microbial EOR methods.⁷ Higher polymer concentration and larger slug size may be profitable for some heavy oils with higher viscosity than the viscosity limitations usually recommended in the screening criteria. There are some field tests where polymer flood was considered for heavier oil reservoirs. Both fields of Pelican lake in Canada and the East Bodo were successfully performed with polymer solution in 600–1000 cp oil reservoirs.⁷

Earlier research results suggest that polymer efficiency is subject to variables such as polymer type, concentration, salinity, pH, rock type, and crude oil compositions.⁸ Especially polyacrylamide polymers are known to be sensitive to shear degradation and shear thickening at high fluxes.⁹ Furthermore, the efforts to increase viscosity in the polymer solution resulted in an alkali-polymer flooding technology with enhanced sweep efficiency. Lowering viscosity of HPAM solution by adding alkali is mainly attributed to high ionic strength condition rather than high pH. Instead, an optimal alkali condition at the same ionic strength tends to increase polymer solution viscosity.¹⁰ Alkalis can also alter rock–fluid and fluid–fluid properties such as wettability

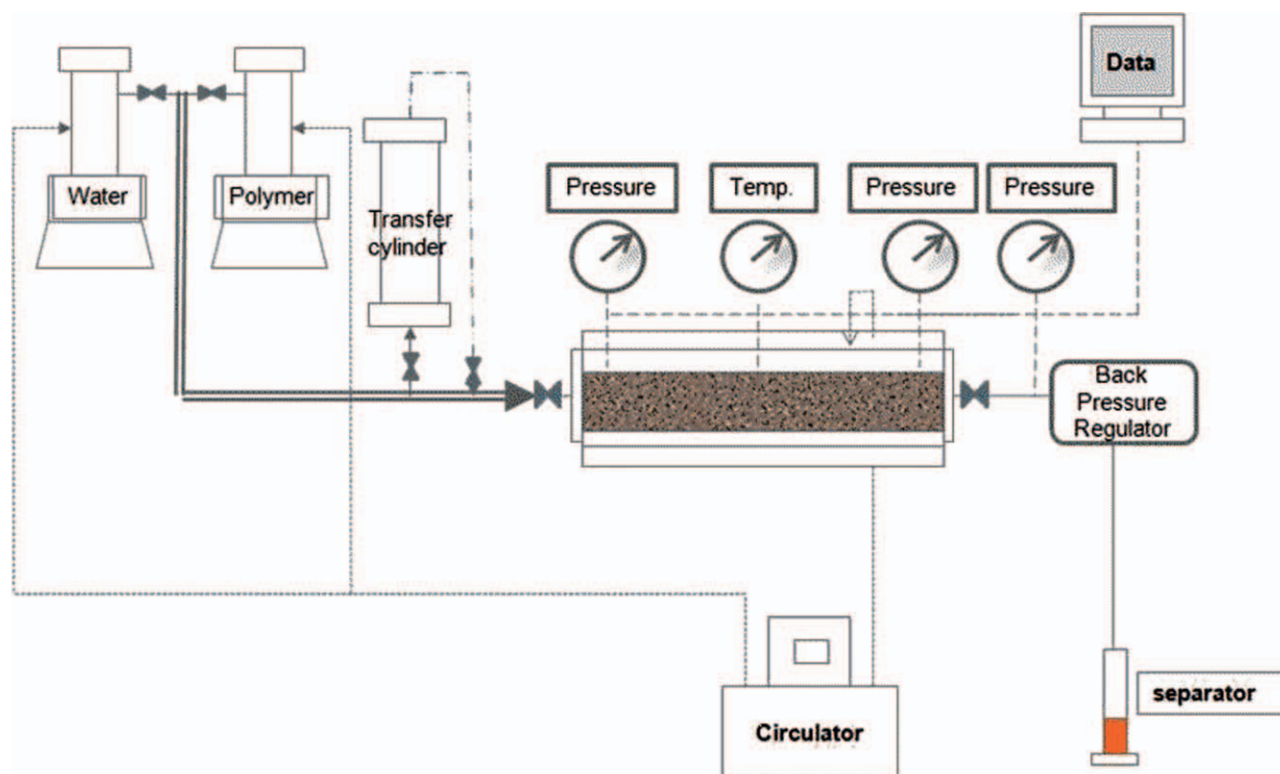


Figure 1. Schematic diagram of experimental apparatus for polymer flooding. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

and oil–water interfacial tension, which could improve oil recovery under proper conditions.¹⁰ Therefore, alkalinity is versatile with improving oil recovery in petroleum industry. It is important to clearly identify rheological parameters of a polymer solution in different environments, i.e., reservoirs.

In this study using the HPAM, test procedures were established to investigate the effect of alkali, salinity, and temperature on polymer solutions in various polymer concentrations. Both the polymer flooding and alkali-polymer flooding tests were performed at same residual oil saturations to investigate the effect of each test on the recovery efficiency. These performances were correlated with rheological properties of the polymer solutions at various experimental conditions.

EXPERIMENTAL

The HPAM (A-132PH, OCI-SNF) with molecular weight of 13×10^6 Dalton and hydrolysis degree of 12 mol % was used, which is linear, water-soluble polymer having negative charges along its chain. The stock solution (0.5 wt %) of HPAM was prepared in advance under mild stirring for 1 week at room temperature. The preprepared polymer solution with the desired concentration was injected in homogeneous glass-bead packs containing heavy oil in the pore space, in which the shear viscosity of heavy oil (SK energy, Korean) is 450 mPa s at 25°C. Porous medium used in the experiment was glass beads ranging from 63 to 106 μm in diameter, which were packed in cylindrical-type core holder with 32 cm in length and 7.8 cm in diameter. When producing glass-bead pack, glass beads were first

thrown into a cylindrical core holder, then the core holder was vibrated for more than 1 h to ensure tight packing, and the core holder packed with glass bead was filled with formation brine in upright position and remained for 24 h for wettability restoration. After that, the porosity and permeability were measured to be 37% and 3.4 Darcy, respectively. Note that reservoir in different areas has different permeability due to various geological structures such as average permeabilities of East Bodo reservoirs and Turkey reservoirs which are low-permeability reservoirs around 1.0 Darcy⁷ and 0.05 Darcy (50 md),¹¹ respectively. On the other hand, the heavy oil reservoirs in California and Alaska (Ugnu Field, North field) have the average permeability from 1.0 to 10 Darcy, while the permeabilities of Canadian reservoirs are in the range 0.5–5 Darcy and Venezuelan reservoirs also show a large range of permeabilities from 2 to 15 Darcy.¹² We prepared the packed bed with the permeability of 3.4, which is in the range of permeability of the most reservoirs in the oil industry. Therefore, it is believed that the results obtained in this study can provide useful references for application of the polymer into the EOR in the unconventional reservoirs. In addition, heavy oil was also injected vertically using ISCO pump until the water production ceased. The initial oil saturation generated in this manner ranged from 88 to 92%. After generating the initial oil saturation, both the polymer flooding and alkali-polymer flooding tests were performed at same residual oil saturations in horizontal flow condition to examine the effect of additives on the recovery efficiency of oil. Summary of pressure on glass-bead pack flood tests is shown in Table IV. Although the glass-bead pack for each test was produced by the

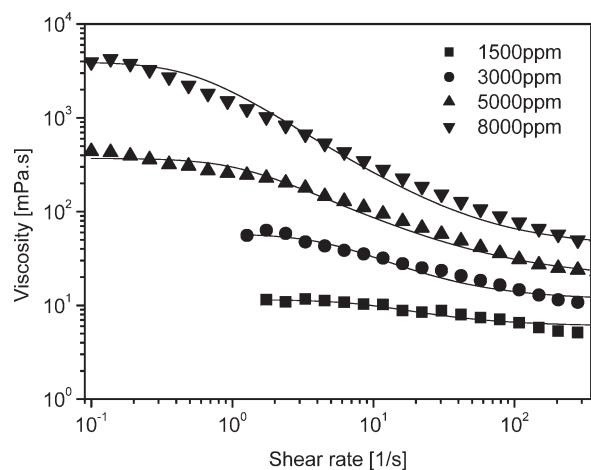


Figure 2. Effect of concentration of HPAM on shear viscosity at 25°C. The solid line obtained from Carreau model.

same method, there still remains an error (The error is in the allowable range), thus leading to the difference in initial inlet pressure.

Injection rate of 4 mL/min was maintained in all flood tests. Water flood tests called as secondary recovery were continued until the water-cut reached about 95% at 0.6 pore volume (PV) of injection. Tertiary flood tests were performed at same residual oil saturations after water floods to investigate the effect of chemical flooding on the recovery efficiency. Even though the increased shear viscosity of the polymer solution improved the oil production in a tertiary oil recovery, there existed a viscosity range of polymer solution in which the tertiary oil recovery had an evident increase in oil production with the increase in the effective viscosity of polymer solution. When the viscosity of polymer solution was out of the range, the increase of the polymer solution viscosity resulted in only a small incremental oil recovery.¹³

The experimental apparatus consisted of cylindrical core holder, ISCO syringe pump, transfer cylinder, pressure transducers, and a computer with data collection board. Figure 1 shows the schematic diagram of the experimental apparatus used for the polymer flooding experiments. The temperature of the cylindrical core holder is controlled at 25°C by circulator, and the outlet pressure is adjusted by back pressure regulator. The water used in all of the flood tests was 3.0 wt % NaCl at 25°C. After the flood test was completed, the photos of the glass-bead packs were taken to compare the residual oil saturation distributions in the cores.

Addition of polymer can increase the viscosity of the aqueous phase and control mobility ratio, thus improve sweep efficiency during EOR processes. The viscosity of polymer solution is strongly dependent on different parameters, such as shear rate, temperature, salinity, hardness, pH, and active polymer concentration.^{1–10,14–16} The effects of concentration, temperature, alkali, and salinity on polymer solutions in various polymer concentrations were investigated via a rotational rheometer (Physica MCR 300, Anton Paar, Germany).

RESULTS AND DISCUSSION

Figure 2 shows the shear viscosity of different concentrations of HPAM solution versus shear rate at 25°C. The shear viscosities at a low shear rate region show an obvious increase with concentration. The apparent viscosities show the shear-thinning behavior with an increasing shear rate as the result of the uncoiling and aligning of polymer chains^{17,18} under exposed shear flow. The relationship of viscosity and shear rate was fitted with Carreau model [eq. (1)], which is one of the shear viscosity models consisting four parameters and can describe the viscosity behavior over the whole range of shear rates not only including a shear-thinning “power-law” range but also involving a low shear rate Newtonian region and a high shear rate Newtonian plateau.¹⁹

$$\frac{\eta - \eta_{\infty}}{\eta_0 - \eta_{\infty}} = [1 + (\lambda \dot{\gamma})^2]^{(n-1)/2} \quad (1)$$

where η is the steady-shear viscosity, η_0 is zero-shear-rate viscosity, η_{∞} is the limiting Newtonian viscosity at the high shear limits, λ is a time constant, and n is the “power-law” index ($n - 1$ is the slope of $\frac{\eta - \eta_{\infty}}{\eta_0 - \eta_{\infty}}$ versus $\dot{\gamma}$ in log–log plot). The parameters in the Carreau model are summarized in Table I, representing that η_0 and λ increase and n decreases when increasing polymer concentrations. The Carreau model is considered to fit the viscosity data in the regions above the critical values of shear rate $\dot{\gamma}$ in both 3000 and 5000 ppm. It can also be noted that very dilute solutions of flexible polymers including the HPAM in this study possess high-extensional viscosities, which affect the pressure losses in the polymer flooding²⁰ and drag reduction in the turbulent drag reduction phenomenon.²¹

It was also observed that the addition of sodium chloride (NaCl) into HPAM solution (1500 ppm) at 25°C significantly reduced the polymer solution viscosity as shown in Figure 3. Nasr-EI-Din et al.²² and Samanta et al.²³ explained that this phenomenon is due to the addition of Na⁺ effectively screening the negative charges (carboxyl groups) to reduce the electrostatic repulsion within the polymer chains. Thus conformational transition of the polymer from a stretched state to a shrinkable state decreases the hydraulic radius of the chain and the degree of polymer chain entanglement, resulting in reduction of viscosity of polymer solution. Although the observation from Nasr-EI-Din et al.²² indicates that the apparent viscosity decreases with increasing salinity concentration at a given shear rate, the difference between the apparent viscosities is small even at low shear rates when the concentrations are higher than 1.4 wt %. Our observation is similar to those observed by Nasr-EI-Din

Table I. The Optimal Parameters in the Carreau Model Obtained from the Flow Curve of HPAM Solution at Various Concentrations

	η_0	η_{∞}	λ	N
1500 ppm	11.5	6.0	0.1	0.04
3000 ppm	58.0	11.5	0.2	0.03
5000 ppm	370.0	19.0	0.9	0.25
8000 ppm	3950.0	43.0	1.9	0.02

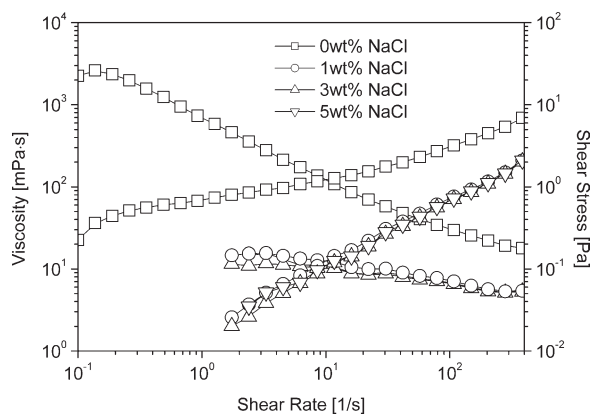


Figure 3. HPAM (1500 ppm) viscosities at different NaCl concentrations at 25°C versus shear rate.

et al.²² The limitation is called “critical salinity”¹⁰ above which the viscosity of solvent is slightly influenced by the increase of salinity. The critical salinity existing is considered to be the saturation of the shielding effect of salt cations around the polymer molecules.

Figure 4 shows the effect of temperature on viscosity of pure HPAM solution (1500 ppm). It is found that the viscosity of polymer solution is strongly dependent on temperature, that is, the viscosity decreased as the temperature increased.²⁴ When temperature increased, the intermolecular interaction is considered to be decreased due to the increase in thermal motion of molecules,²³ while Muller et al.²⁵ indicated that the influence of temperature on HPAM polymer viscosity is very complicated because the solution viscosity can be increased due to the enhanced hydrolysis of HPAM at high temperature range.

The shear viscosity of HPAM is strongly dependent on the variation of pH due to the carboxyl groups (pH-sensitive) along its backbone chain. Thus, the viscosity of HPAM with different NaOH concentration was observed in Figure 5. It can be seen that the viscosity of HPAM increases with NaOH concentration at lower concentrations, and then it slightly declines and changes to be steady at higher concentration. At low NaOH concentrations, the polymer molecules of HPAM in the solution

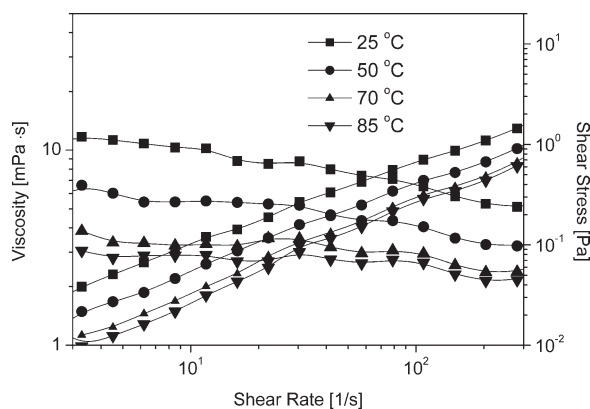


Figure 4. Viscosity of HPAM aqueous solutions (1500 ppm) at different temperatures.

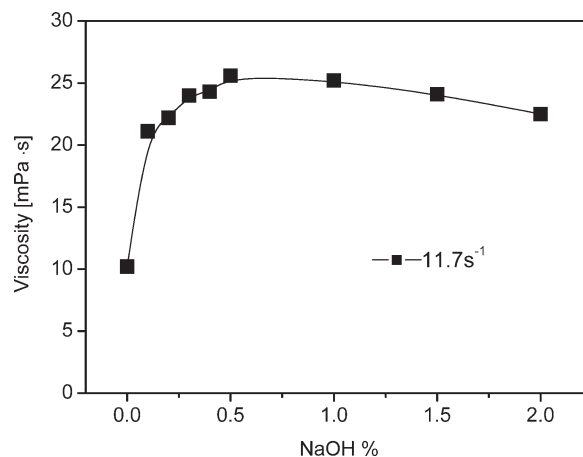


Figure 5. Effect of NaOH concentration on shear viscosity of HPAM solution (1500 ppm) with the shear rate of 11.7 s⁻¹.

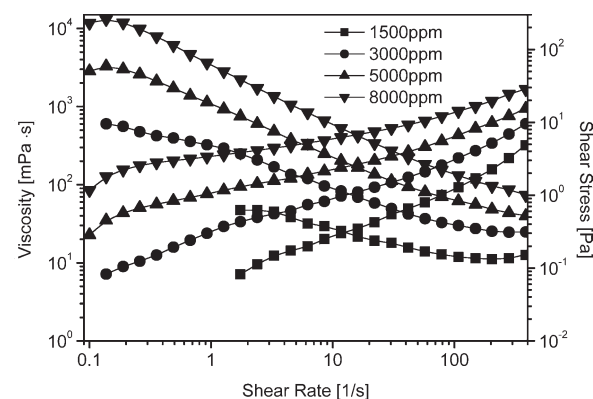


Figure 6. Effect of concentration of HPAM on shear viscosity with 0.5 wt % of NaOH at 25°C.

possess tightly coil conformation, leading to a low viscosity. However, by increasing NaOH concentrations, the negatively charged carboxyl groups along the chains make the polymer molecules tend to coil up due to the electric repulsion showing large hydrodynamic radius, which leads to a large increase in

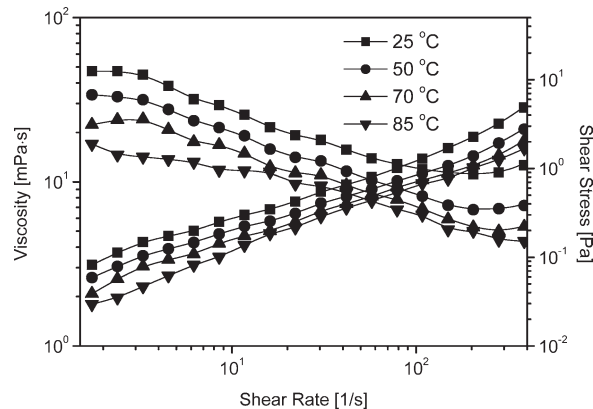


Figure 7. Effect of temperature on shear viscosity of HPAM (1500 ppm) with 0.5 wt % NaOH at 25°C.

Table II. Properties of the Glass-Bead Packs

Flood types	Porosity (%)	Absolute permeability (Darcy)	Soi (%)	Sor after water flood (%)
Water flood	36.9	3.79	91.0	–
Polymer flood	37.8	3.15	91.9	76.0
Alkali-polymer flood	36.3	3.33	91.2	75.5

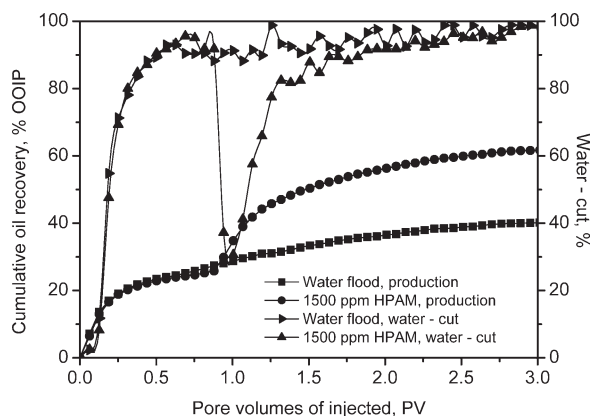


Figure 8. Comparison of oil recoveries in water and polymer flood tests.

solution viscosity. This phenomenon is similar to the salinity effect on viscosity as shown in Figure 3. According to the results obtained in Figure 5, we prepared different polymer solutions with 0.5 wt % NaOH for another rheological measurement.

In Figure 6, it can be observed that increasing the concentration of HPAM polymer solution also drives the viscosity to increase when NaOH (0.5 wt %) is added, while the increase extent in viscosity is higher than “alkali-free” solution (Figure 5) at the same concentration, indicating that addition of NaOH significantly increase the viscosity because of the increase of ionic strength.¹⁰ The similar results of the increase in viscosity of HPAM polymer solution (1500 ppm) can be found with increasing temperature when NaOH (0.5 wt %) is added (Figure 7). Kurenkov et al.²⁶ reported that polyacrylamide can be hydrolyzed in the presence of sodium hydroxide to form copolymer of acrylamide (AA) and acrylic (AAc) salts. Thus, in our experiment, the new generation of carboxylic side groups along HPAM chain might be obtained due to hydrolysis in alkali solution, which causes the polymer chain more expanded, resulting in increase of viscosity.

Table III. Summary of Glass-Bead Pack Flood Tests

Flood types	Water flood recovery		Tertiary recovery		Final recovery OOIP (%)
	OOIP (%)	Injected (PV)	OOIP (%)	ROIP (%)	
Water flood	40.2	3.0	–	–	40.2
Polymer flood	24.0	0.6	37.9	81.3	61.9
Alkali-polymer flood	24.5	0.6	46.8	94.4	71.3

Note that the main goal of this study was to investigate the technical feasibility of the proposed variations of an improved polymer flood and to examine their effectiveness when compared with a conventional water flood. In order to achieve this goal, a few parameters had to be established. Table II summarizes petrophysical properties of glass-bead packs for flood experiment.

The main purpose of using polymer in the EOR process is to increase solution viscosity and reduce rock permeability, both of which help to increase sweep efficiency by reducing the mobility of displacing fluid.¹⁰ In order to investigate the effect of polymer flooding on oil recovery, a comparison of cumulative oil recoveries between water flooding and polymer flooding was made as shown in Figure 8. The flow rate in all the flood tests was maintained at 4 mL/min by using syringe pump.

The first test was with the oil recovery made by water flood test with an injection of 3.0 PV. The second test was the oil recovery made by the polymer flood test with an injection from 0.6 to 3.0 PV after the water-cut reached about 95% at 0.6 PV of water injection. The polymer solution of both 1500 ppm solution of HPAM and 3 wt % NaCl was injected. As shown in Figure 8, experimental results represented that 61.9% of original oil in place (OOIP) was produced by polymer flood test, whereas only 40.2% of OOIP was produced by water flood test with an injection of 3.0 PV. Therefore polymer flooding proved to be more efficient in oil recovery than water flooding by 21.7%. It would be recognized that the more PVs of polymer solution is injected the more oil is recovered due to the reduction in mobility ratio of water to oil. The oil recovery was recorded as a function of PV of injected fluids in each test. The test results are summarized in Table III. Since the oil recovery is affected by viscous fingering phenomenon, the low viscous fingering at the water front would increase oil recovery. In water flooding, on the other hand, further injection of water would not produce any significant amount of oil after the water-cut ratio of 95% at 0.6 PV. Wassmuth et al.⁷ applied polymer flooding of the HPAM to improve heavy oil recovery at East Bodo with the permeability of 1 Darcy, finding that the oil recovery reached 20% OOIP and the water cut was approached to 95% after water flooding. In addition, Wassmuth et al.²⁷ also reported that the oil recoveries ranging from 16 to 23% OOIP was achieved by polymer flooding in different oil core floods with the viscosities from 280 (with the core permeability of 1.35 Darcy) to 1600 cp (with the core permeability of 5.7 Darcy) after the water-cut at a minimum of 1 PV.

As shown in Figure 9, an unstable pressure drop can be seen after breakthrough in water flood test. Such phenomenon can be explained by the viscous fingering occurrence at outlet face.

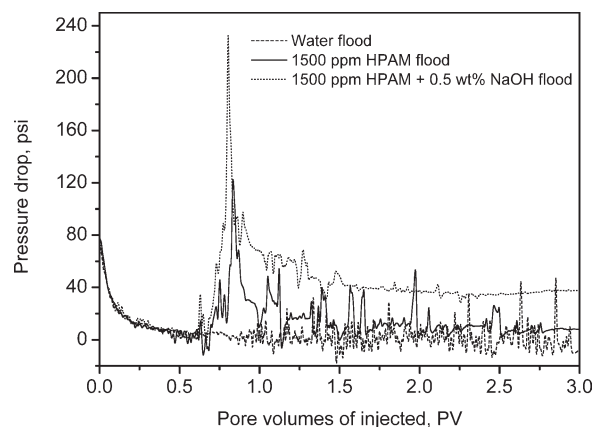


Figure 9. The shape of pressure drops in each test.

The alkali is directly related to the high viscosity of polymer solution. The pH effect on viscosity was confirmed by rheological analysis. Thus, it was designed to find the effect of alkali-polymer flood on tertiary recovery. The mixed solution of 12% HPAM, 3 wt % NaCl, and 0.5 wt % NaOH was prepared for the flood tests. The third test was the oil recovery made by alkali-polymer flooding with an injection from 0.6 to 3.0 PV. Water flooding was conducted until the water-cut ratio reached to 95% at 0.6 PV before the third test. Alkali-polymer flooding was followed after the water flooding to compare the recovery efficiency of alkali-polymer flooding with that of polymer flooding. As shown in Figure 10, 71.3% of OOIP was recovered by the alkali-polymer flood test. It represents almost 10% more oil recovery over HPAM solution when using HPAM solution with alkali. It can be explained that pressure drop after breakthrough held stable at 37.6 psi in the alkali-polymer flood, which is five times higher than that of polymer flooding stable at 7.3 psi as shown in Figure 9. The combination of alkalis and polymer strengthened the block role of displacing fluid in channel zones so that alkali-polymer flooding pressure drop was improved.²⁸ Moreover oil recovery efficiency increased more rapidly in the early stage of alkali-polymer injection due to the effect of mobility control. This indicates that more oil is recovered due to the improved sweep efficiency and mobility ratio reduction. Samanta et al.²⁹ presented the mechanism of alkali in EOR which showed that alkali can react with the organic acid existed in crude oil to form the surfactant and emulsification,³⁰ thus to decrease the surface tension. In the process, the variation of surface tension is suggested to be strongly dependent on the

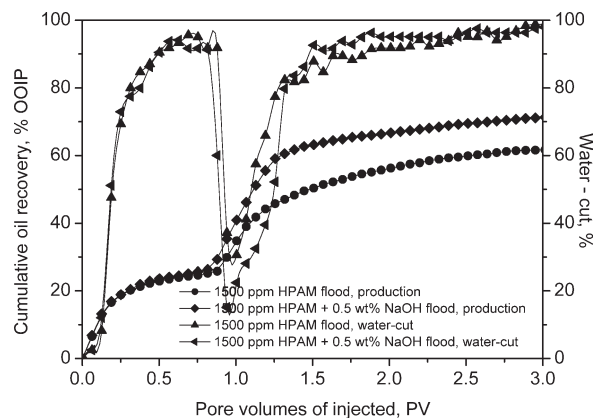


Figure 10. Comparison of oil recoveries in polymer and alkali-polymer flood tests.

concentration of alkali, which is increased with an increase in the concentration of alkali and then decreased. In addition, Kazempour et al.³¹ reported that the effect of alkalinity on oil recovery during polymer floods in sandstone, indicating that the alkali could react with polymer to decrease its viscosity, allowing for better injectivity. And, addition of alkali also allows for decreasing adsorption of HPAM on the rock surface and polymer retention in the core. In the experiment, 95% water-cut was obtained at 1.85 PV in alkali-polymer flood, while 95% water-cut was obtained to 2.33 PV in polymer flood. It could be explained that improved sweep efficiency led to the acceleration of oil recovery.

CONCLUSION

Throughout the experiments, the potential impact of an alkali-polymer flood on heavy oil recovery was investigated. A full-scale rheological characterization of the polymer solutions was studied and test results were compared. Shear viscosity behavior was examined by rheological test results as well as viscous modulus measurements at different polymer concentrations, salinities, temperatures, and alkalis. Both the oil recovery by the polymer flood and by the alkali-polymer flood were attempted and compared as shown in Table IV. Rheological measurements show that HPAM solution is sensitive to temperature, salinity, and alkali. The solution of 0.5 wt % NaOH mixed with 1500 ppm HPAM was the optimal choice, giving rise to the highest viscosity on the rheological characterization.

Table IV. Summary of Pressure on Glass-Bead Pack Flood Tests

Flood types	Initial inlet pressure (psi)	Pressure at water flood recovery		Pressure at tertiary recovery		
		Initial pressure drop (psi)	Injected (PV)	Maximum inlet pressure (psi)	Initial pressure drop (psi)	Stable pressure (psi)
Water flood	190.7	74.3	3.0	-	-	1.8
Polymer flood	214.1	77.8	0.6	190.4	125.8	7.3
Alkali-polymer flood	219.7	75.2	0.6	431.6	260.2	37.6

The flood tests performed on glass-bead packs indicated that alkali-polymer could improve oil recovery up to 10–30% of OOIP over the water flooding or polymer flooding. The application of polymer solution was more effective with 0.5 wt % NaOH. The results can be applied to the polymer flooding projects to evaluate the feasibility at the planning stage.

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