

Research Article

Rheological Study on ATBS-AM Copolymer-Surfactant System in High-Temperature and High-Salinity Environment

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Experimental studies were conducted to evaluate the rheological properties of surfactant-polymer (SP) system. This SP system consists of a copolymer of acrylamide (AM) and acrylamido tertiary butyl sulfonate (ATBS) and sodium dodecyl sulphate (SDS) surfactant. Effects of surfactant concentration, temperature, polymer concentration, and salinity on rheological properties of SP system were investigated by means of oscillation and shear measurements. Comparison with classical partially hydrolyzed polyacrylamide (HPAM) was made. For the same temperature range, the viscosity drop for HPAM was about four times higher than the viscosity drop for ATBS-AM copolymer. In deionized water, viscosity of both polymers and SP systems was very high as compared to viscosity in saline water. Viscosity reduction of ATBS-AM copolymer was higher for salts having divalent cations. The SP system showed precipitation in presence of divalent cations. It worked well with monovalent cations even at relatively high salinities. The addition of 0.1% surfactant to the polymer resulted in a 60% decrease in the viscosity. Some interfacial rheological experiments were also carried out to investigate the behaviors on the interface between SP solutions and oil. Addition of 0.1% surfactant showed a 65% decrease in G' at SP solution-oil interface. SP system consisting of ATBS-AM and SDS showed better performance at high temperature compared to HPAM-SDS system. Due to precipitation, the SP system should be restricted to environment having low divalent cations.

1. Introduction

In chemical enhanced oil recovery (cEOR), surfactants are used to reduce the interfacial tension between crude oil and water. Polymers are used to improve the mobility ratio by viscosifying displacing fluid. Enhanced viscosity of displacing fluids can increase macroscopic displacement efficiency by overcoming viscous fingering. Recently, some researchers have proved that these polymers not only improve macroscopic displacement efficiency but also microscopic efficiency due to elasticity [1–7].

HPAM which is widely used for cEOR applications [8] fails at high-temperature and high-salinity (HTHS) environment. Extensive hydrolysis of amide group at high temperatures may also cause precipitation of hydrolyzed product in the presence of divalent cations. Field applications of available polymers are limited to low-temperature and low-salinity reservoirs. HTHS reservoirs present a major challenge for implementation of cEOR techniques. Incorporation of some thermally stable and salt tolerant comonomers can enhance the performance of polyacrylamide in HTHS conditions. Recently, synthesized thermoviscosifying polymers [9–12]

showed some positive results in HTHS environment after some preliminary rheological tests.

Three major types of surfactant-polymer interactions may exist depending upon the nature and structure of polymer and surfactant head group. Firstly, attractive forces between surfactant and polymer may be stronger than mutual forces of surfactant molecule. Secondly, attractive forces among surfactant molecules may be greater than attractive forces between surfactant and polymer. Thirdly, repulsive forces between surfactant and polymer are very high as compared to attractive forces among surfactant molecules [13]. The system becomes more complex in the presence of oil, cosurfactant, alkali, and salts. Many approaches have been adopted to understand the interactions between surfactant-polymer systems. These include surface tension and interfacial tension measurement, rheology [14–17], fluorescence spectroscopy [18–20], potentiometry [21], light scattering [22, 23], and conductivity measurement [24–28].

There are some reports available in the literature on the rheology of classical HPAM/SDS [15] PAM/SDS [29] and hydrophobically modified PAM (HM-PAM)/SDS [14] system. Addition of the surfactant has no effect on the rheology of the polymer due to nonionic nature of PAM. Significant effect of surfactant addition has been reported for HPAM and HM-HPAM due to strong interactions between the surfactant and polymer. By addition of 30 mmol/L (0.86%) SDS, about 85% drop in the viscosity of HPAM is reported by Bu et al. [17]. SP solution consisting of SDS and vinylpyridine-AM copolymer in 0.1N NaCl solution has about 90% less viscosity than that of the viscosity of the SP solution in deionized water [16]. Temperature also plays an important role in surfactant-polymer interactions. For HM-HPAM/SDS system, Nystrom [29] found that the viscosity at 40°C is 80% less than that of viscosity at 10°C.

Rheology study can be used to screen many SP systems for cEOR applications. In addition, interfacial rheology can provide the insight on the interface between SP solution and oil. The objective of this work is to study the influence of salts, temperature, and surfactant concentration on rheological properties of SP system consisting of SDS and ATBS-AM copolymer. Some of the results were compared with HPAM.

2. Experimental

2.1. Materials. Copolymer of AM and ATBS (Flopaam An125SH) with a molecular weight of 8 million Dalton and 25% sulfonation degree was obtained from SNF FLOERGER in a powder form. Classical HPAM (Flopaam 3630S) with a molecular weight of 20 million Dalton and 30% hydrolysis degree was obtained from SNF FLOERGER. Sea water was prepared using laboratory grade sodium bicarbonate, sodium sulphate, sodium chloride, calcium chloride, and magnesium chloride with a total salinity of 57,638 mg/L (ppm). SDS with 99% purity was obtained from Sigma Aldrich. The oil used in interfacial rheology experiments has a density of 0.8767 g/cm³.

2.2. Preparation of Polymer Solutions. A polymer solution was prepared by using magnetically driven stirrer. The polymer was added slowly on the shoulder of the vortex formed by deionized water, surfactant solution, or salt solution to avoid formation of slubs. As soon as the entire polymer was added the rotor speed was reduced from 300 to 80 rpm to avoid mechanical degradation. Stirring was turned off after 3 hours and the solution was kept at room temperature for the next 48 hours for complete hydration.

2.3. Rheological Measurements. Rheological measurements were carried out using discovery hybrid rheometer (DHR-3) from TA Instrument. Concentric cylinder geometry was used to measure both steady state and oscillation measurements. The range of shear rate for steady shear viscosity measurements was from 0.01 to 1000 s⁻¹. Frequency from 0.1 to 100 rad/s was applied for oscillatory measurements. All reported data points are within torque limits (± 5 mN·m–2000 mN·m). Frequency sweep experiments were conducted in the linear viscoelastic region. Oscillation time experiment was performed to check the thermal stability of HPAM and ATBS-AM copolymer. Interfacial rheology experiments were performed using AR-G2 platinum double wall ring interfacial geometry. Frequency was kept at 1 rad/s in oscillation time and temperature ramp experiments unless it is stated otherwise.

3. Results and Discussion

3.1. Effect of Surfactant Concentration. Copolymer concentration was fixed to 0.25% for experiments with varying surfactant concentration. Unless otherwise specified, the term copolymer in this discussion refers to ATBS-AM copolymer. Figure 1 shows the viscosity versus shear rate plots of 0.25% copolymer in deionized water for different concentrations of surfactant. It is evident from Figure 1 that by increasing the surfactant concentration steady viscosity is decreasing. The viscosity of the SP system is low compared to the viscosity of copolymer. By adding 0.3% surfactant, the viscosity is 80% less than that of the viscosity of the copolymer. Similar degree of viscosity drop by adding SDS has been reported for HPAM [15, 17]. This decrease in viscosity by the addition of surfactant is due to charge shielding mechanism as reported by Mandal and Ojha [15] for HPAM. At very high shear rate, viscosity is almost the same for all surfactant concentrations due to the dominance of the effect of shear in comparison to charge interactions. Variation of complex viscosity at a frequency of 0.1 rad/s is also shown in Figure 2. Storage modulus, G' , versus surfactant concentration is shown in Figure 3. G' decreases with the increase in surfactant concentration. As G' is a measure of elastic nature, it is obvious that addition of surfactant is causing loss in network structure of ATBS-AM copolymer. Interfacial rheological properties were also evaluated at oil/SP solution interface to examine the influence of surfactant concentration. Figure 4 shows the interfacial storage modulus (G'_I) versus frequency curves at different surfactant concentrations. At all frequencies, G'_I decreases by increasing surfactant concentration. We can conclude that

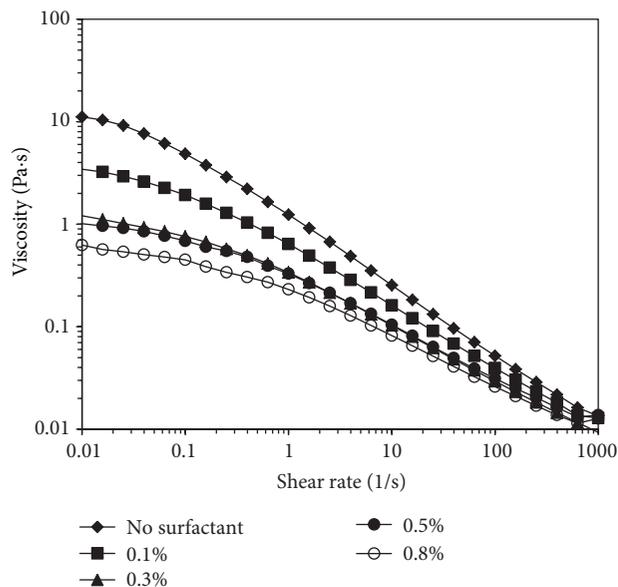


FIGURE 1: Effect of surfactant concentration on steady shear viscosity of copolymer at different shear rates in deionized water at 50°C.

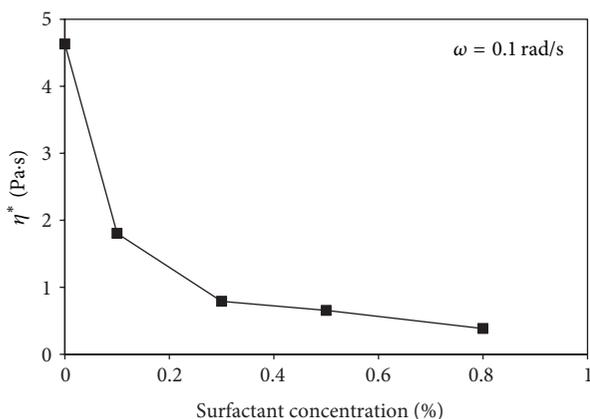


FIGURE 2: Effect of surfactant concentration on complex viscosity of copolymer in deionized water at 50°C.

addition of surfactant is weakening the interface between oil and SP solution. Time sweep measurements were also carried out to determine G'_I . These measurements were performed for 5 minutes and within experimental time no change in storage modulus was observed. Results of time sweep experiment are shown in Figure 5. A major drop in G'_I due to addition of 0.1% surfactant is observed, while, further addition of the surfactant has little effect.

3.2. Effect of Temperature. Viscosity versus shear rate plots for HPAM and ATBS-AM copolymer in the absence of surfactant is obtained at two different temperatures (Figure 6). It is evident that increasing temperature from 50°C to 90°C causes a major drop in steady shear viscosity of HPAM as compared to the copolymer. At low shear rate, the viscosity of HPAM at 90°C is about 60% less than its viscosity at 50°C. However,

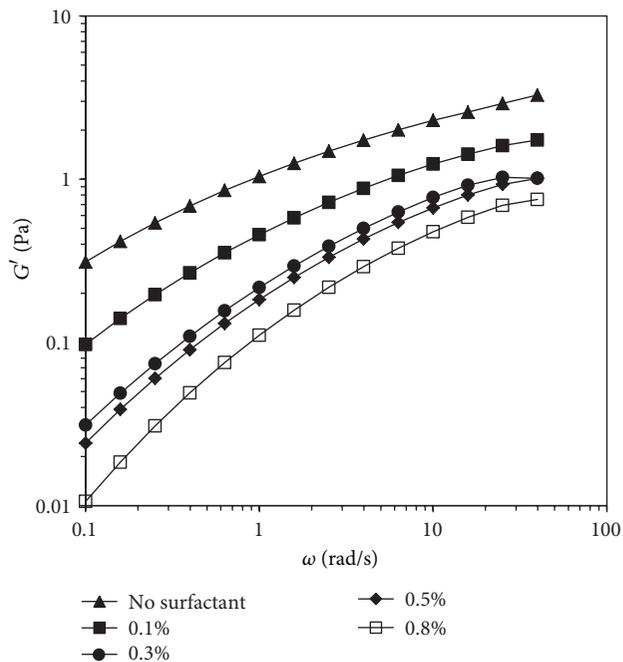


FIGURE 3: Effect of surfactant concentration on storage modulus of copolymer at different frequencies in deionized water at 50°C.

for ATBS-AM copolymer, small change in viscosity (15%) was observed in the same temperature range. This suggests that the viscosity of ATBS-AM copolymer is less sensitive to temperature in comparison with HPAM. Time sweep experiments were run for two hours for both polymers at 50°C and 90°C and results are shown in Figure 7. Both polymers were stable at 50°C within the experimental time. Within the experimental time, G' and viscosity (not shown) did not change for both polymers. At 90°C, completely different results were found. Unlike ATBS-AM copolymer a continuous decrease in storage modulus of HPAM was observed as experiment proceeds. From the above results it can be concluded that at 50°C both polymers show good stability and can be used at this temperature. However, at 90°C HPAM shows a continuous decrease in rheological properties while AM-ATBS copolymer is stable.

Figures 8 and 9 present results of temperature ramp experiments at 2°C/min for different surfactant concentrations. Viscosity versus temperature plots are shown in Figure 8. At all temperatures, the viscosity of the polymer and SP solutions is decreasing with increasing surfactant concentrations. However, the decrease in viscosity depends on the concentration of surfactant. With no surfactant added in the solution, 20% decrease in viscosity was observed in the temperature range 30°C to 85°C. With the addition of 0.1% surfactant, the viscosity decreases by 33% for the same temperature range. For 0.3% and higher surfactant concentrations about 58% drop in viscosity was observed. For HM-HPAM/SDS system 80% viscosity decrease was observed when temperature was increased from 10 to 40°C [29]. This shows that ATBS-AM/SDS system perform much better in high temperature compared to HM-HPAM/SDS system.

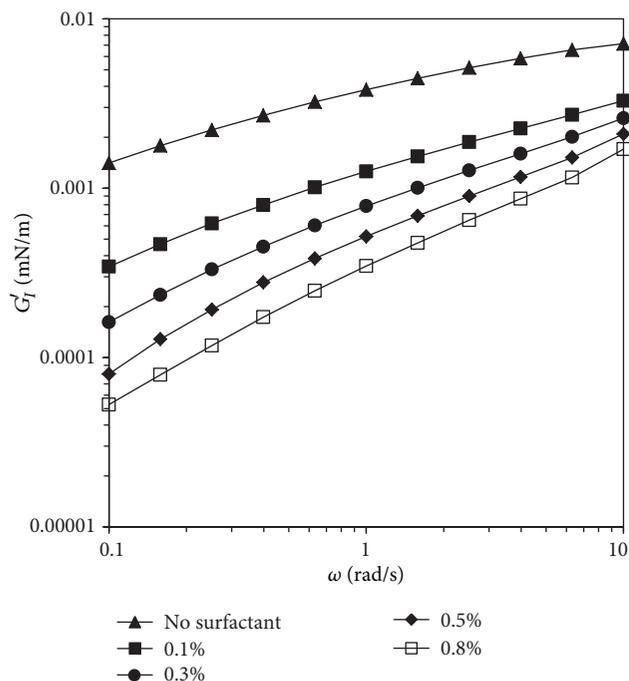


FIGURE 4: Effect of surfactant concentration on interfacial storage modulus of copolymer at different frequencies in deionized water at 25°C.

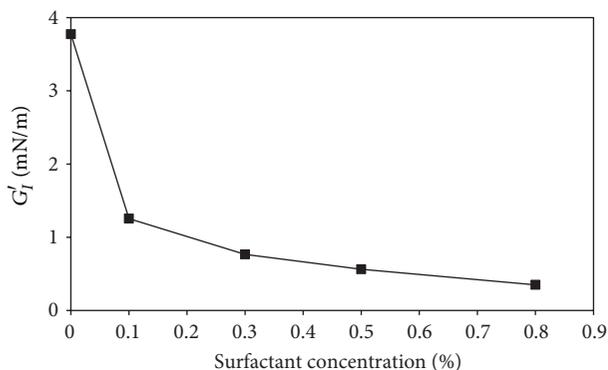


FIGURE 5: Effect of surfactant concentration on interfacial storage modulus of copolymer in deionized water at 25°C.

Arrhenius model was used to obtain activation energy of viscous flow and pre-exponential factor for solution of different surfactant concentrations. Activation energy of viscous flow increased while pre-exponential factor decreased with the addition of surfactant (Table 1). The percentage decrease in the pre-exponential factor is high as compared to the percentage increase in the activation energy. The above data indicate that in the presence of a surfactant, temperature has more effect on the viscosity of SP solutions as compared to copolymer solution having no surfactant. Similarly, for polymer solution without surfactant, a 26% drop in G' was noted. For 0.3% and higher surfactant concentrations about 70% decrease in storage modulus was observed in the same temperature range. Effect of temperature on G'_I is shown in

TABLE 1: Activation energy and pre-exponential factors obtained from Arrhenius model for different surfactant concentrations.

Concentration (%)	E_a^a (kJ/mole)	A^b (mPa-s)
0	5.020	210.74
0.1	7.010	45.177
0.3	14.54	1.2797
0.5	15.38	0.8152
0.8	15.877	0.5497

^a E_a is activation energy of viscous flow.

^b A is pre-exponential factor.

TABLE 2: Activation energy and pre-exponential factors obtained from Arrhenius-type relation for different polymer concentrations.

Concentration (%)	E_a^a (kJ/mole)	A^b (mPa-s)
0.15	18.32	0.4266
0.20	16.94	1.1205
0.25	14.54	1.2797
0.30	14.90	5.0728
0.40	13.86	12.891

^a E_a is activation energy of viscous flow.

^b A is pre-exponential factor.

Figure 10. With the increase in temperature G'_I also decreases. This shows that temperature is also weakening the interface, but its effect is not as much when we compare it to the effect of surfactant concentrations. The surfactant concentration is a major source of reduction in interfacial rheological properties between SP solution and oil interface.

3.3. *Effect of Polymer Concentration.* Surfactant concentration was fixed to 0.3% for all experiments and polymer concentration was varied from 0.1 to 0.4%. Figures 11 and 12 summarize the effect of polymer concentration on SP system. No unusual results were observed by changing the polymer concentration. It is clear from Figures 11 and 12 that increasing the concentration of polymer will cause increase in steady shear viscosity. This increase is more prominent at low shear rate. Results of temperature ramp experiments at a rate of 2°C/min for different copolymer concentrations are shown in Figures 13 and 14. Both storage modulus and viscosity decrease with increasing in temperature at all polymer concentration. However, the decrease in the viscosity and storage modulus depends on the polymer concentration. Maximum drop in viscosity (58%) and storage modulus (70%) was found for SP solution having minimum polymer concentration (0.15%) and minimum drop in viscosity (49%) and storage modulus (57%) was found with SP solution of highest polymer concentration (0.4%). Activation energy of viscous flow and pre-exponent factors for SP solutions of different polymer concentrations were calculated and tabulated in Table 2. Pre-exponent factor and activation energy of viscous flow of the SP solutions do not change significantly by changing the polymer concentration. However, slight change in surfactant concentration significantly changes the pre-exponent factor and activation energy of the SP solutions.

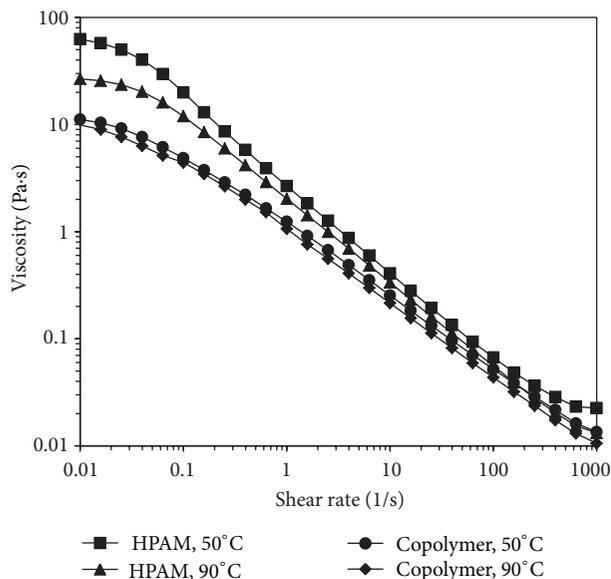


FIGURE 6: Steady shear viscosity profile of HPAM and ATBS-AM copolymer solutions in salt-free water without surfactant at 0.25% polymer concentration.

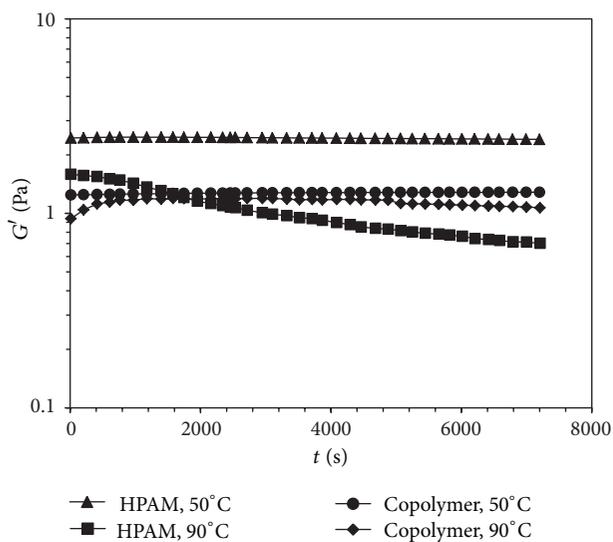


FIGURE 7: Comparison of G' of HPAM and ATBS-AM copolymer solutions in deionized water by time sweep measurements at 0.25% polymer concentration.

Decrease in the viscosity with temperature depends mainly on surfactant concentration and polymer concentration has little effect.

3.4. Effect of Salts. High temperatures and high salinities are real challenges for polymer flooding for the Middle East carbonate reservoirs. In the presence of salts and higher temperatures, viscosity of HPAM decreases. Synthetic sea water with a total salinity of 57638 mg/L containing 2732 mg/L divalent cations was prepared in the lab. It is important to see the effect of each individual ion on the SP system used for

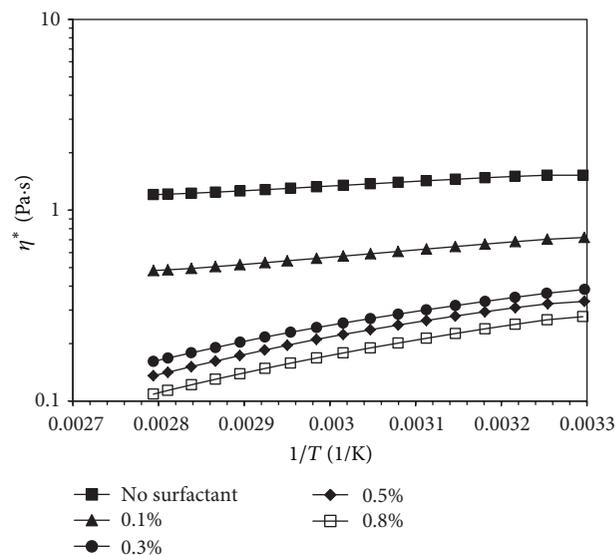


FIGURE 8: Effect of temperature on viscosity of 0.25% copolymer at different surfactant concentration.

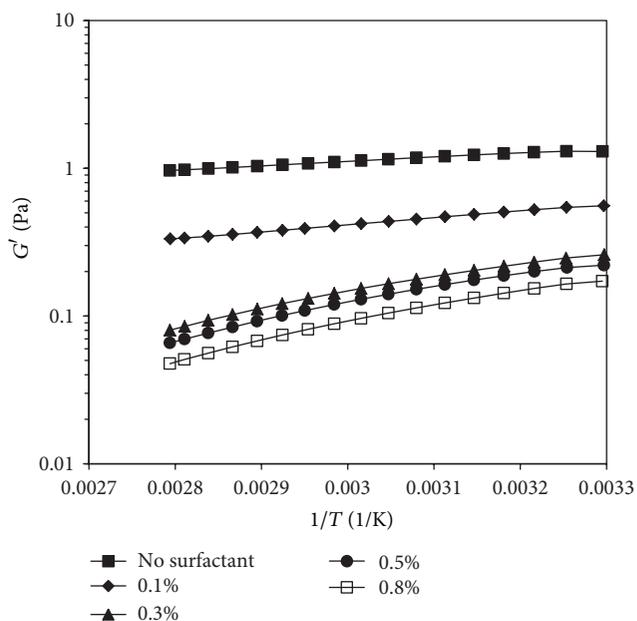


FIGURE 9: Effect of temperature on G' of 0.25% copolymer at different surfactant concentration.

chemical flooding. Besides sea water salt solutions of NaCl, Na₂SO₄, MgCl₂, and CaCl₂ were prepared with different molarities. SP solutions of 0.25% polymer and 0.3% surfactant concentration were used to evaluate the performance in different salts. Figure 15 shows the comparison of viscosity of both polymers in deionized water at 50°C without a surfactant. A major reduction in viscosity was observed for both polymers by adding synthetic sea water. Figures 16 and 17 show the effect of sodium chloride concentration on rheological properties of SP system. Steady shear viscosity reduces as the concentration of sodium chloride is increased.

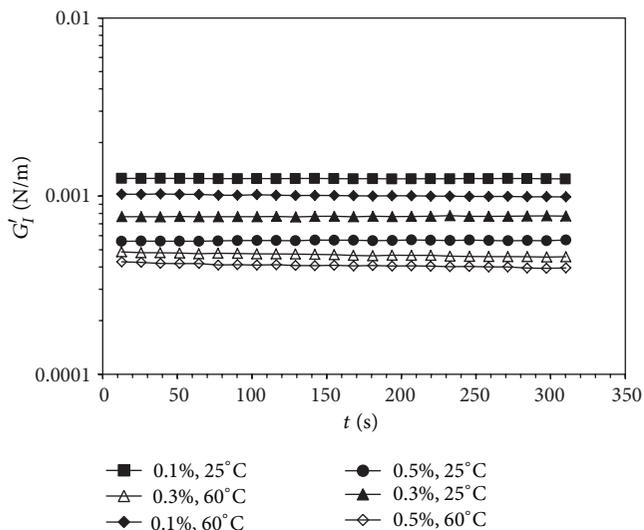


FIGURE 10: Effect of temperature on G'_I of 0.25% copolymer at different surfactant concentration.

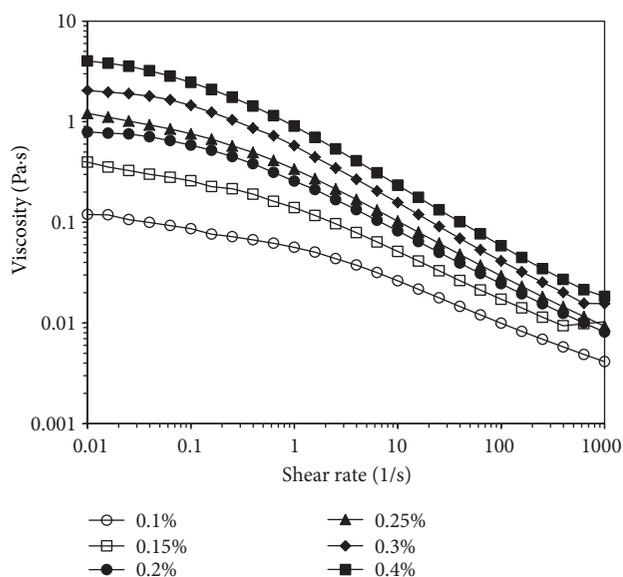


FIGURE 11: Steady state viscosity profile of SP solutions with varying concentration of copolymer at 50°C.

Viscosity at zero shear decreases with the increase in NaCl concentration. Similar trend was also observed for storage modulus. Addition of salts brings counterions in the system which reduces the stretching in polymer chains by charge screening and resulting in reduction in viscosity and storage modulus. A reduction in viscosity was also observed for sodium sulphate as shown in Figure 18. With calcium chloride and magnesium chloride precipitation of SP system was observed. With sea water there was also precipitation, but dilution of sea water to 25% sea water and 75% DW gives a clear solution. This observation shows that this SP system should be limited to environment having low divalent cations. In the absence of divalent cations this SP system

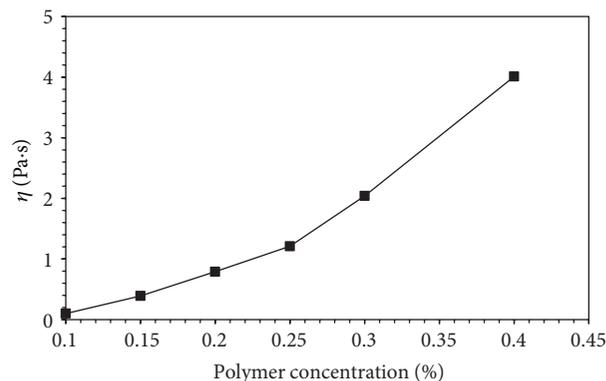


FIGURE 12: Effect of copolymer concentration on viscosity of SP solution at 50°C and shear rate of 0.01 s^{-1} .

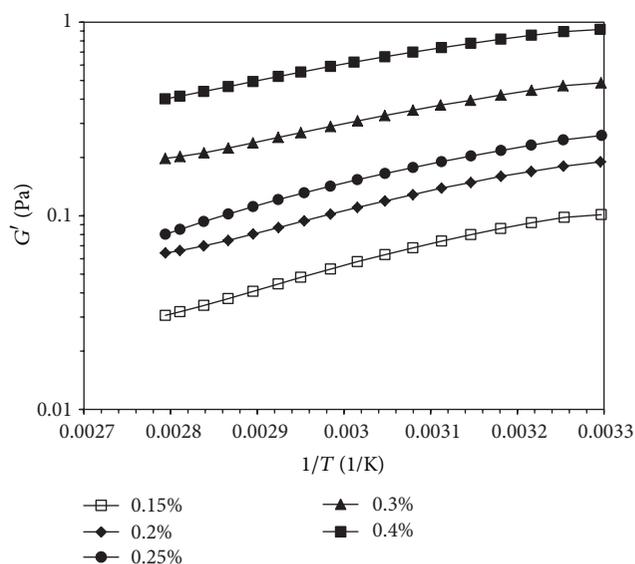


FIGURE 13: Effect of copolymer concentration on storage modulus of SP solution at different temperatures.

can be used even at high salinities. Figure 19 presents a comparison of the impact of different counterions on the steady shear viscosity of copolymer. Compared to deionized water, viscosity of polymer is lower in 0.01 M NaCl solution. But this viscosity is still higher than that of SP solution in 0.01 M NaCl having a surfactant. Both surfactant and salts are bringing counterions. System having surfactant has more counterions and introducing additional chain collapse. It is also clear from Figure 19 that decrease in viscosity is higher for polymer solution having calcium ions as compared to monovalent sodium ions. Divalency is the major cause of reduction in viscosity.

4. Conclusions

About 60% reduction in viscosity for HPAM was observed by increasing temperature from 50°C to 90°C. However,

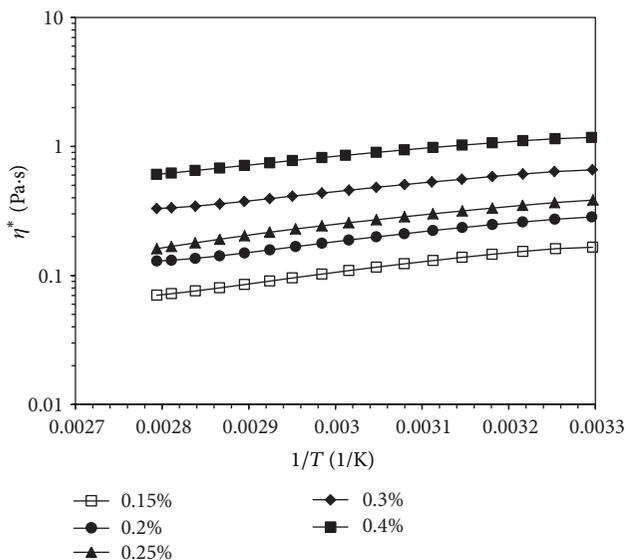


FIGURE 14: Effect of copolymer concentration on viscosity of SP solutions at different temperatures.

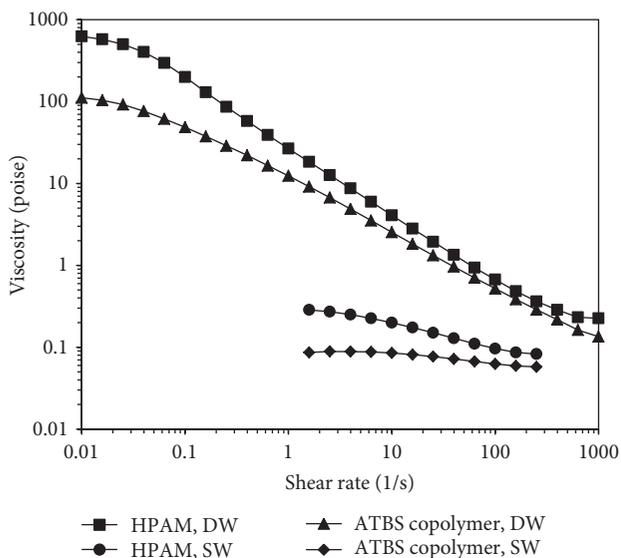


FIGURE 15: Effect of sea water on steady shear viscosity of HPAM and ATBS-AM copolymer without any surfactant at 50°C.

decrease in viscosity for ATBS-AM copolymer was 15% for the same temperature range. It can be seen from temperature sweep experiments that at 90°C there was a continuous decrease of storage modulus of HPAM. Copolymer did not show significant decrease in storage modulus and viscosity within the experimental time. Copolymer of acrylamide and acrylamido tertiary butyl sulfonate (ATBS) has better performance at higher temperatures as compared to partially hydrolyzed polyacrylamide. SP system consisting of AM-ATBS copolymer and SDS showed better performance from rheological point of view at high temperature compared to previously reported HPAM/SDS system. Presence of anionic surfactant SDS is also a source of reduction in the viscosity

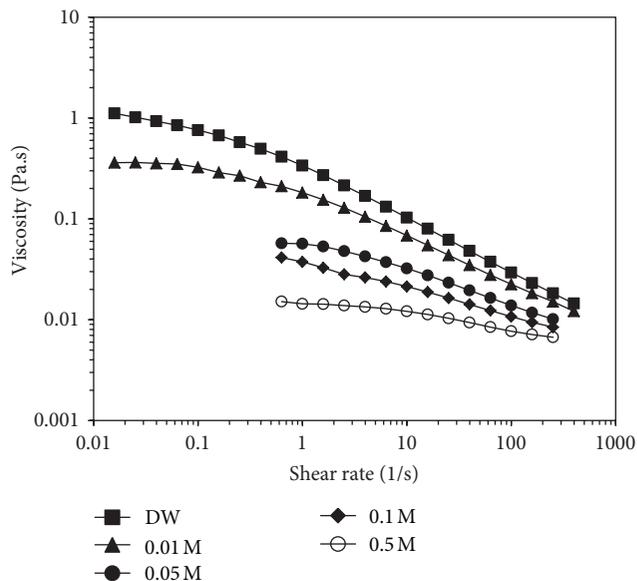


FIGURE 16: Effect of sodium chloride concentration on steady shear viscosity of copolymer at 50°C.

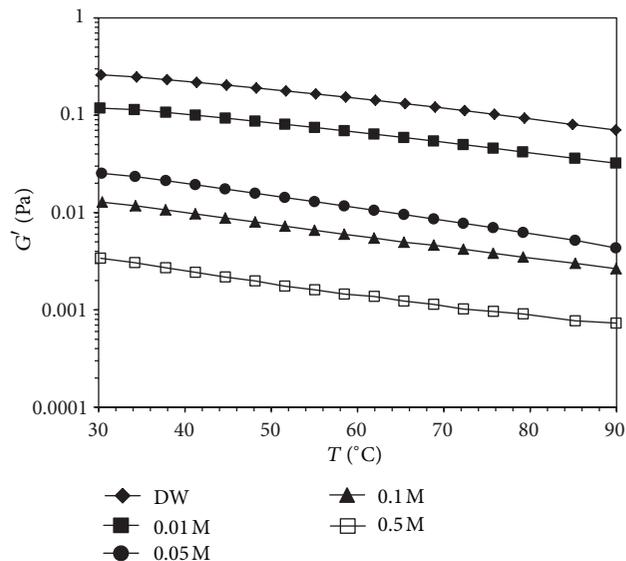


FIGURE 17: Effect of sodium chloride concentration on storage modulus of copolymer at different temperatures.

and storage modulus of polymer solution. Interfacial rheology experiments showed that this surfactant is significantly weakening the interface between SP solution and oil. Effect of each salt on the rheological properties of SP solution was investigated. There is a decrease in the viscosity in the presence of monovalent cations. In the presence of divalent cations precipitation was observed. There was precipitation with sea water. Dilution of this sea water by a ratio of 1 : 3 with DW yields a solution with no precipitation. Both polymers can be potential candidate for low temperature and low salinity reservoirs. HPAM is not suitable for high temperature

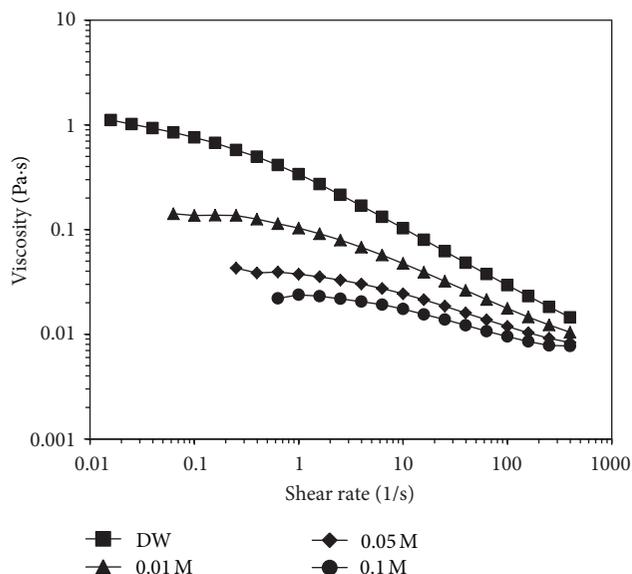


FIGURE 18: Effect of sodium sulphate concentration on steady shear viscosity of copolymer at 50°C.

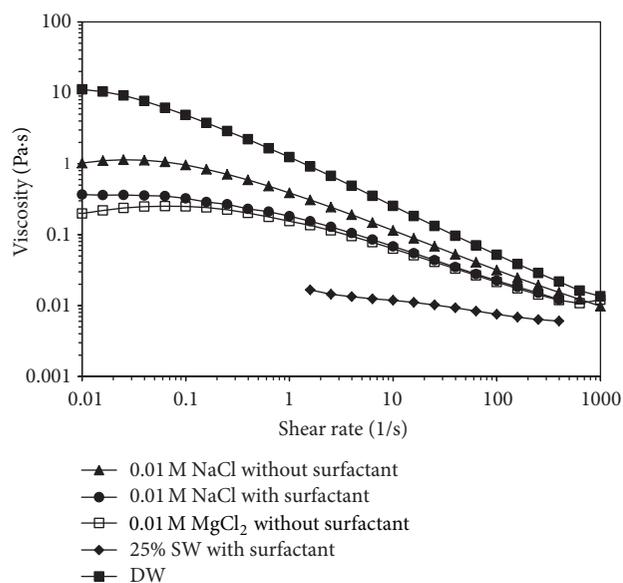


FIGURE 19: Effect of different salt on steady shear viscosity of copolymer at 50°C.

reservoirs while ATBS based copolymer can be a potential candidate for high temperature reservoirs.

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