



Experimental evaluation of partially hydrolyzed polyacrylamide and silica nanoparticles solutions for enhanced oil recovery

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Received: 7 June 2019 / Accepted: 31 July 2019 / Published online: 3 August 2019
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Abstract

The establishment of oil production well is becoming a challenge with the increasing demand for energy. The fulfillment of energy need requires large production of oil and gas as it is a primary source of energy. EOR is also important because of the enhancement in oil production from thirty percent to more than fifty percent. The chemical EOR is one of the techniques for the increment in oil production. Chemical flooding using water-soluble polymers like partially hydrolyzed polyacrylamide (PHPA) has been industrially used as an EOR technique. The paper deals with the effect of nano-silica particles on viscosity as well as the shear rate of the polymer solution. The change in viscosity, as well as shear rate, was studied at variable concentrations of the nano particles in the different concentrations of PHPA solution. Mutual correlation between viscosity and other parameters like temperature, shear rate, salinity, nanoparticle concentration, and polymer concentration was established using the statistical method.

Keywords Enhanced oil recovery (EOR) · Nano-silica particles · Viscosity · Shear rate · Optimization

Introduction

Oil and gas are the primary requirement for energy resources throughout the world and it will continue in the future. The continuous production of the hydrocarbon leads to the depletion of oil and gas resources. Only thirty percent production of hydrocarbon is possible with the conventional method. Therefore, it is highly required to recover the remaining unreachable hydrocarbon from fulfilling the increasing demand. The recovery of those unreachable hydrocarbons is possible by using enhanced oil recovery (EOR) methods. Out of several EOR techniques, polymer flooding is the most common and effective chemical EOR methods for potential recovery of oil and gas (Wever et al. 2011; Sheng et al. 2015). This technique overcomes several disadvantages of the conventional waterflooding technique like the fingering and bypassing of water through oil. One of the critical

parameters in the flooding process is the mobility ratio, which is a measure of the combined effect of permeability and fluid viscosities on fractional flow. If the mobility ratio is one or slightly less than one, there will be an efficient and piston-like displacement of oil (Vossoughi 2000). Partially hydrolyzed polyacrylamide (PHPA) is the most frequently used polymer in this respect, owing to its low cost, viscosifying nature, and well-known physiochemical properties (Urbissinova et al. 2010). The molecule of this polymer is a flexible chain which remains in coil-like structure in the absence of a shearing effect. The application of PHPA is limited due to high temperature and salinity (Uddin et al. 2002). In such conditions of elevated temperature, the amide groups of the chain undergo extensive hydrolysis into a carboxylic acid and these hydrolyzed products get precipitated when they come in contact with cations commonly present in reservoir brines (Thorne et al. 2010; Taylor and Nasr-El-Din 1998). PHPA acts as shear-thinning polymer, i.e., it undergoes shear degradation and reduces viscosity at high shear rates. This is because, at high shear rates, the chains of the polymer get cutoff (Song et al. 2006).

An increasing number of research studies using nanoparticles are being carried out every year. This special attention is due to the unusual properties that these particles display because of their extremely small size and large surface area.

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The size of these particles varies from 1 to 100 nm. Thus, the use of nanoparticles in EOR is highly appreciable for hydrocarbon production (Raghav Chaturvedi et al. 2018). Polymer nanoparticles solutions have been studied for potential applications in chemical flooding operations, and it has been found that the addition of nanoparticles may greatly alter the polymer properties of the solution along with properties like the rock wettability and interfacial tension. The smaller size and large surface area of NP make it suitable for application in polymer flooding. Owing to their small size (1–100 nm), NPs can be propagated through the formation using a stable aqueous polymer solution. The NPs exhibit thermal stability at higher reservoir temperature (Sohn et al. 1990; Seright et al. 2001; Pritchett et al. 2003; Ponnappati et al. 2011; Raghav Chaturvedi et al. 2018). Silica nanoparticles, in particular, have shown great potential to increase the oil recovery (Needham and Doe 1987; Modesto et al. 2009; Maghzi et al. 2013). Some studies have attributed the effect to the wettability and interfacial tension alteration (Koochi et al. 2010) and the enhancement in viscosity elsewhere (Kharrat and Vossoughi 1992). The rheological properties of any fluid to be used for flooding are of utmost importance. The increase in viscosity is what helps prevent viscous fingering (Kavanagh and Ross-Murphy 1998). Viscous fingering is the cause for excessive water production in a majority of wells being produced from under waterflooding technique. The increased viscosity of the injected fluid thwarts this problem and thus enhances oil recovery (Aslam et al. 1984). The most significant and obvious mechanism of polymer flooding is the increased sweep efficiency by reducing viscous fingering and improving the water injection profile (Sheng et al. 2015). Polymer flooding also creates a high-viscosity solution to control the mobility of the displacement process (Torrealba and Hoteit 2019). Therefore, the study of polymer flooding is important for the chemical EOR.

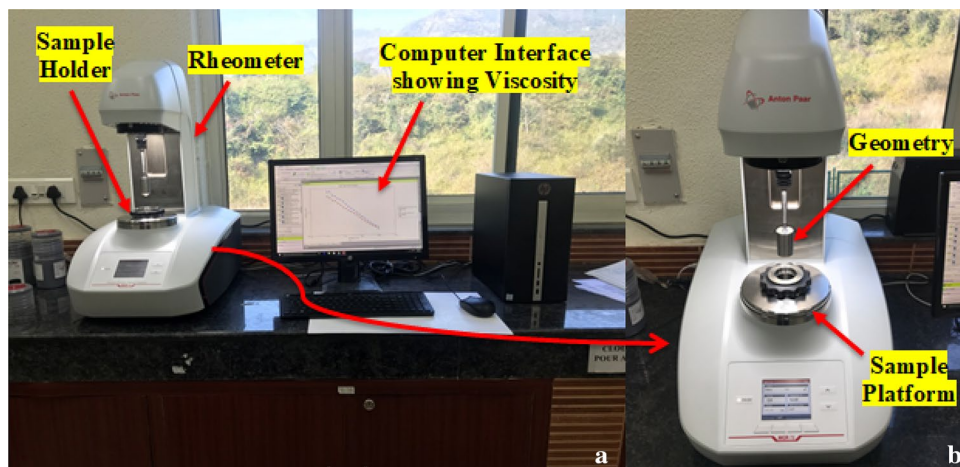
In this paper, the effects of various parameters like temperature, salinity, shear rate, nanoparticles concentration, and polymer concentration on the rheology of silica nanocomposite partially hydrolyzed polyacrylamide were studied. Some comparisons have also been drawn between the changes in the properties of the nanocomposites and those of the PHPA solution. The mutual correlation was established between different parameters using the statistical method.

Materials and methods

The silica nanoparticles used in this study were purchased from Nano Research Labs, Jamshedpur, Jharkhand, India. Partially hydrolyzed polyacrylamide (PHPA) polymer was procured from ONGC, Dehradun, India. The average particle size of silica nanoparticles varied from 20 to 30 nm. Polymer solutions of various concentrations were prepared using PHPA and brine. The solution was mixed using magnetic stirrer up to homogeneous condition followed by aging for 24 h at room temperature ($\sim 28^\circ\text{C}$). After 1 day of aging, silica nanoparticles were added into the solution and mixed until the nanoparticles fully dissolved in the solution and prepared a homogeneous solution.

Rheological parameters (viscosity and shear rate) were measured using Rheometer (Make: Anton Paar, MCR-72) (Fig. 1). The measurement was taken using cylindrical geometry to prevent the spilling of sample and skin formation. A CC27 bob (27 mm diameter) along with a flow cup (28.92 mm inner diameter) was used for measurement. Viscosity was measured at variable temperature and shear rate. Temperature control was achieved using a Peltier system. It allowed fast heating and cooling along with high-accuracy measurement. The mutual correlation was established between different parameters. The correlations were used to study the effects of parameters like salinity, polymer

Fig. 1 Measurement of viscosity using rheometer **a** placement of the sample in sample holder followed by computer interfacing **b** machine showing geometry and sample platform



concentration, and nanoparticle concentration on the solution viscosity.

Results and discussion

The variation in viscosity, as well as shear rate, was determined at variable polymer concentration, nanoparticle concentration as well as temperature. Results were obtained from the experimental investigation and has broadly discussed in the following subsections.

Effect of polymer concentration on viscosity

A solution was prepared with polymer concentration varied from 0.5 to 2 wt% and a fixed brine concentration of 1 wt%. The viscosity of the solution was determined at a constant shear rate of 110 s^{-1} and temperature $87.4 \text{ }^\circ\text{C}$. Increase in viscosity was observed with polymer concentration following nonlinear logarithmic correlation with correlation coefficient (R^2) of 0.93 (Fig. 2). Increase in viscosity was very fast between 0.5 and 0.9 wt%, stable between 1 and 1.6 wt%, and moderate from 1.7 to 2 wt% of polymer concentration. This is due to the fast formation of a long chain in the initial stage and stability of the macro-molecular structure with increasing concentration of the polymer that leads to an increase in viscosity. This increase in viscosity can be related to higher entanglement of polymeric chains, which is the governing factor in viscosity of polymers. The increase in viscosity is attributed to the long chains of bonded monomers. The entanglement of these chains makes the structure bulky, and as a result, the viscosity increases. It is the major factor to decrease the mobility ratio and sweeps the oil efficiently. Increase in viscosity with polymer concentration at higher temperature and saline environment confirms beneficial effect for a polymer flooding operation.

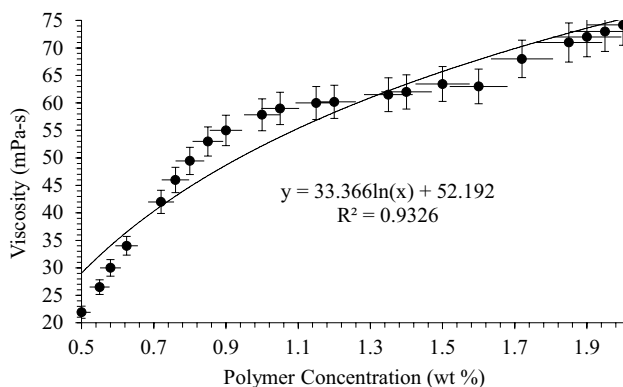


Fig. 2 Correlation between polymer concentration and viscosity at constant brine concentration, shear rate, and working temperature

Effect of silica nanoparticle concentration on viscosity

Viscosity was measured with varying silica nanoparticles from 0.1 to 2.5 wt% at constant PHPA conc. (1.0 wt%), brine conc. (1.0 wt%), shear rate (100 s^{-1}), and temperature: $87.4 \text{ }^\circ\text{C}$. The silica nanoparticles concentration and viscosity exhibit positive linear correlation with correlation coefficient (R^2) of 0.99 (Fig. 3). The addition of nanoparticles significantly increased the viscosity of the polymeric solution. The increase in viscosity is due to hydrogen bonding between the silanol groups in the nanoparticles and the amide groups in the PHPA. The ion–dipole interaction of cation and oxygen of the silica reduces the molecular size due to thermal conductivity, and as a result, the viscosity increases. The increment in viscosity was observed favorable for polymer flooding operations. The increase in the viscosity is reflected in terms of enhanced oil recovery and improvement in the rate of the production.

Effect of salinity on viscosity

The test was carried out with a 0.5 wt% silica nanoparticles concentration, 1 wt% PHPA concentration at a constant temperature of $87.4 \text{ }^\circ\text{C}$, and shear rate of 100 s^{-1} . Variation in viscosity was determined at different brine concentrations from 1 to 3 wt%. Negative linear correlation between viscosity as well as brine concentration was observed with R^2 of 0.99 (Fig. 4). Reduction in viscosity was observed with brine concentration. The screening of polymer site or chain with an increase in brine solution increases the origination period, and as a result, viscosity period also increases, and thus, the viscosity of polymer decreases. The delay in the viscosity of the polymer is due to the formation of sodium cation shield on amide group. This shield causes shrinkage and masking of the polymer chain or polymer site. The shrinking and

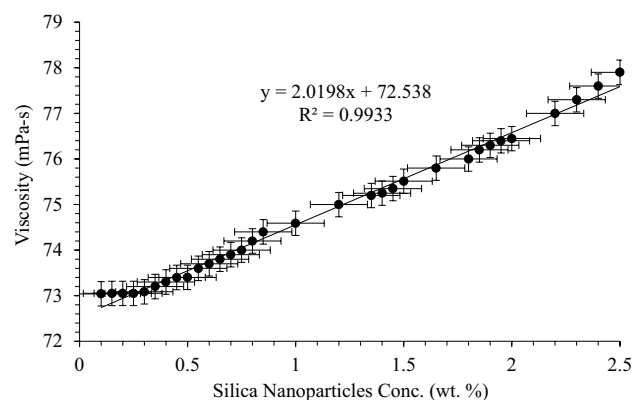


Fig. 3 Correlation between silica nanoparticle concentration and viscosity at constant PHPA conc., brine conc., shear rate, and temperature

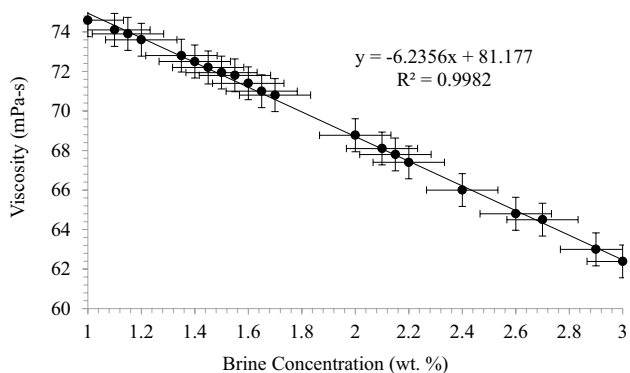


Fig. 4 Correlation between salt concentration and viscosity at constant PHPA conc., silica nano particles conc., shear rate, and temperature

masking effect in the polymer site leads to the reduction in active cross-linking site and minimization of the intensity of active site. Thus, the viscosity of the polymer solution with salinity decreases. The tendency of salinity to reduce the viscosity of polymer solution makes it as a retardation agent in high-temperature reservoirs and help in EOR.

Effect of temperature on viscosity

The relation between temperature as well as the viscosity of the solution was determined at a variable concentration of silica nanoparticles and a constant shear rate of 100 s^{-1} . The temperature and silica nanoparticles concentration varied from 80.15 to 110.68 °C and 0.1 to 2 wt%, respectively (Table 1). The change in viscosity with variation in temperature as well as silica nanoparticles concentration is shown in Fig. 5. Reducing the trend of viscosity was observed with the

Table 1 Variation in viscosity with temperature and silica nanoparticles concentration

Si. no.	Temp. (°C)	Silica nanoparticles conc. (wt%)	Viscosity (mPa-s)	Si. no.	Temp. (°C)	Silica nanoparticles conc. (wt%)	Viscosity (mPa s)
1	80.15	0.1	78.94	31	80.15	1	81.882
2	83.98	0.1	75.641	32	83.98	1	78.8
3	87.4	0.1	73.085	33	87.4	1	75.514
4	90.72	0.1	70.829	34	90.72	1	74.021
5	94.04	0.1	68.838	35	94.04	1	72.543
6	97.35	0.1	66.201	36	97.35	1	70.037
7	100.69	0.1	58.03	37	100.69	1	67.378
8	103.88	0.1	58.386	38	103.88	1	60.187
9	107.2	0.1	58.058	39	107.2	1	59.057
10	110.68	0.1	61.948	40	110.68	1	66.674
11	80.15	0.3	78.878	41	80.15	1.5	82.335
12	83.98	0.3	75.674	42	83.98	1.5	79.227
13	87.4	0.3	73.402	43	87.4	1.5	76.448
14	90.72	0.3	71.53	44	90.72	1.5	76.021
15	94.04	0.3	68.748	45	94.04	1.5	74.58
16	97.35	0.3	66.86	46	97.35	1.5	72.703
17	100.69	0.3	58.208	47	100.69	1.5	67.299
18	103.88	0.3	58.7	48	103.88	1.5	64.314
19	107.2	0.3	58.775	49	107.2	1.5	60.125
20	110.68	0.3	64.813	50	110.68	1.5	64.389
21	80.15	0.5	81.446	51	80.15	2	84.91
22	83.98	0.5	77.108	52	83.98	2	80.511
23	87.4	0.5	74.59	53	87.4	2	77.902
24	90.72	0.5	71.556	54	90.72	2	77.438
25	94.04	0.5	69.54	55	94.04	2	76.134
26	97.35	0.5	67.185	56	97.35	2	72.949
27	100.69	0.5	58.707	57	100.69	2	67.042
28	103.88	0.5	58.993	58	103.88	2	64.113
29	107.2	0.5	58.944	59	107.2	2	62.286
30	110.68	0.5	66.475	60	110.68	2	66.697

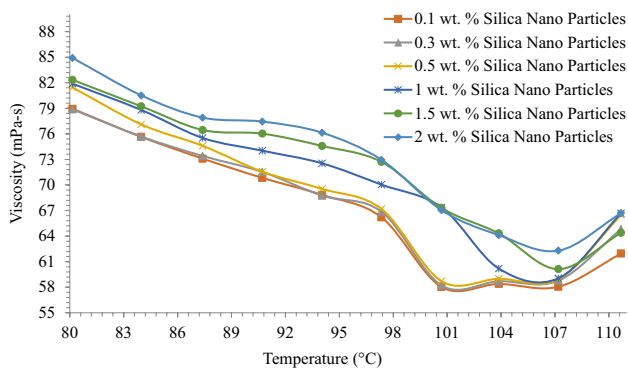


Fig. 5 Correlation between silica nanoparticle concentration and viscosity at constant PHPA conc., brine conc., shear rate, and temperature

temperature at different silica nanoparticle concentrations. The minimum and maximum values of viscosity varied from 58.03 to 62.26 mPa s and from 78.94 to 84.91 mPa s for silica nanoparticles concentration from 0.1 to 2 wt%, respectively (Fig. 5). The correlation in Fig. 5 indicates an increased viscosity with silica nanoparticles concentration at a higher temperature. This is due to the formation of long-chain bonded monomers and bulky structure. The long-chain formation increases the cohesive force in the solution, and hence, the viscosity increases. Therefore, it is confirmed that enhanced oil recovery is possible at elevated temperature with a higher concentration of silica nanoparticles.

Effect of shear rate on viscosity

Experimental investigation of prepared partially hydrolyzed polyacrylamide–silica nanoparticle solutions was carried out using stress-controlled rheometer. A total of 30 ml of prepared solution was used for the testing

purpose. The relation between viscosity, as well as the shear rate, was determined at different silica nanoparticle concentrations varied from 0.1 to 2 wt%, a constant polymer of 1 wt%, and at temperature 90 °C. The minimum and maximum values of viscosity varied from 21.492 to 32.129 mPa s and 557.69 to 1082.46 mPa s, respectively, at the shear rate from 0.99 to 1000 s⁻¹ (Table 2). The decrease in viscosity was observed with shear rate. The increased viscosity was observed with the inclusion of silica nanoparticles concentration in the solution at a higher shear rate (Fig. 6). The decrease in viscosity with shear rate is due to the formation of an intermolecular or intramolecular hydrogen bond between long-chain intermolecular or intramolecular associations or hydrophobic cross-linking in the long-chain polymer structure. The correlation between viscosity and shear rate clearly shows the non-Newtonian and shear-thinning behavior of the polymer solution.

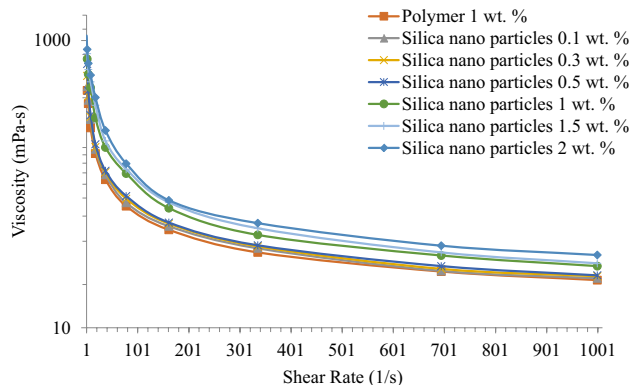


Fig. 6 Correlation between shear rate and viscosity at variable silica nanoparticles concentration and constant polymer concentration

Table 2 Variation in viscosity with shear rate and silica nanoparticles concentration at 90 °C

Shear rate (1/s)	Viscosity with 1 wt% polymer (mPa-s)	Viscosity with 0.1 wt% silica nanoparticles (mPa s)	Viscosity with 0.3 wt% silica nanoparticles (mPa s)	Viscosity with 0.5 wt% silica nanoparticles (mPa s)	Viscosity with 1 wt% silica nanoparticles (mPa s)	Viscosity with 1.5 wt% silica nanoparticles (mPa s)	Viscosity with 2 wt% silica nanoparticles (mPa s)
0.99	557.69	589.44	834.57	997.77	1024.71	1055.81	1082.46
2.07	450.11	477.35	567.13	688.53	745.28	809.79	868.92
4.28	363.97	384.22	410.21	447.39	583.52	629.15	694.33
8.86	246.72	282.19	294.13	302.17	470.98	516.24	575.43
18.3	163.56	174.78	181.28	189.75	289.76	354.22	401.28
37.9	107.53	116.28	121.37	124.02	180.25	200.35	236.71
78.5	70.384	74.121	79.339	82.664	118.65	127.14	139.17
162	48.106	50.962	53.146	54.039	68.129	74.668	77.213
336	33.493	35.758	36.873	37.532	44.332	49.312	53.516
695	24.647	24.892	25.712	26.891	31.875	33.483	37.287
1000	21.492	22.115	22.865	23.183	26.889	28.125	32.129

Conclusion

In this study, the effects of various parameters on the rheology of a nanocomposite solution were studied. Increase in viscosity was observed with polymer concentration following nonlinear logarithmic correlation. Increase in viscosity with polymer concentration at higher temperature and saline environment confirms beneficial effect for a polymer flooding operation. Significant increase in viscosity was observed with the addition of the silica nanoparticles. The silica nanoparticles concentration and viscosity exhibit positive linear correlation. Increase in the viscosity with silica nanoparticles is reflected in terms of enhanced oil recovery and improvement in the rate of oil production. The negative linear correlation between the viscosity of nanoparticle composite and brine concentration was observed. The tendency of salinity to reduce the viscosity of polymer solution makes it as a retardation agent in high-temperature reservoirs and help in EOR. Reducing the trend of viscosity was observed with the temperature at variable silica nanoparticle concentration. The addition of silica nanoparticles was found to increase the viscosity of the polymer solution at a higher temperature also. The decrease in viscosity was observed with shear rate. The increased viscosity was observed with the inclusion of silica nanoparticles concentration in the solution at a higher shear rate.

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