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Study of Rheological Property and Flow Behavior for Nanoparticles Enhanced VES System in Porous Media

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In this paper, a composite sample (VES and SiO_2 nanoparticle) was used to overcome the deficiencies of polymer. The rheological character of the VES/nanoparticles hybrid and flow behavior in porous media were examined. It was found that SiO_2 nanoparticles exhibited viscosifying action and improved the oil tolerance. In addition, the VES solution without nanoparticles showed a lower capacity to recover oil, which might be attributed to the fact that wormlike micelles would be destroyed in crude oil. On the contrary, an enhanced oil recovery of 9.68% was achieved in the composited experiment for the VES sample with nanoparticles which is relatively stable with oil.

Keywords: worm-like, nano-SiO_2, rheology, enhanced oil recovery, viscoelastic surfactant, worm

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INTRODUCTION

Viscoelastic surfactant (VES) fluids, generally formed by wormlike micelles, have been utilized as completion or stimulation agents in the oil and gas industry (Jeffrey Giacomin et al., 2008). Durga P. Acharya discussed the formation of wormlike micelles and the evolution of rheological properties in different mixed surfactant systems (Zhang, et al., 2018; Chu, et al., 2010). Lstvan Lakatos evaluated the VES fluid as a mobility control agent. It shows that a slug-type injection protocol is more efficient than the continuous injection of any single displacement fluids (Lakatos, et al., 2007). Michael Golombok carried out experiments on inert glass cores in the range of 45–2,200mD. By analogy they considered their observations to correspond to permeability thickening, although it was understood that the permeability was fixed and the apparent viscosity increased. For the range of permeabilities that are applicable to oil reservoirs, the apparent viscosities observed at high permeabilities are around 10 times that of the low permeabilities (Golombok and van der Wijst, 2013).

Recent work has shown the advantageous use of nanoparticles in VES fluid systems, which included significantly increased thermal stability and fluid loss control properties in the fluid system (Huang and Crews, 2007; Qin, et al., 2017). It shows that when selected nanoparticles are added to a VES solution, they will associate or “pseudo-crosslink” the VES micelles together through charge attraction and surface adsorption (Huang, 2007). Ranjini Bandyopadhyay and A.K. Sood studied the effects of the addition of submicrometer-sized colloidal silica spheres on the linear and nonlinear rheology of semidilute solutions of a viscoelastic gel (Bandyopadhyay and Sood, 2005). The oscillatory rheological measurements for nanoparticles in viscoelastic surfactant fluids noted that the nanoparticles apparently strengthened the micelle-micelle interactions. Lab proppant settling tests demonstrated that the nanoparticle induced VES micelle network structures that dramatically increased the capacity of the surfactant fluid to suspend and transport proppant in well treatments

(Huang et al., 2008; Huang and Crews, 2007; Huang and Crews, 2008; Crews and Huang, 2008; Crews and Ahmed, 2012; Gurluk, et al., 2013). M.F. Fakoya thinks the nanofluids could be used for hydraulic fracturing (Fan et al., 2011; Fakoya and Shah, 2013; Fakoya and Shah, 2014). They also investigated the pyroelectric barium titanate ($BaTiO_3$) nanoparticle and found the viscosity of the MES micelle solution increased with the temperature within a certain range because of the pyroelectric effect of the nanoparticles (Luo, et al., 2012; Helgeson, et al., 2010).

For the majority of oil reservoirs, large amounts of oil are still left unrecovered after extensive water flooding. Chemical EOR technology is the most promising tertiary recovery technique to both improve sweep and displacement efficiency. The well-known process to improve sweep efficiency consists in injecting polymer solution. This process known as polymer flooding has been widely used at large scale especially at Daqing field in China (Morvan, et al., 2009). For conventional polymer flooding, the injection of concentrated polymer solutions raises injectivity issues and requires higher injection pressure with the risk of exceeding the formation fracturing pressure. Micelles behave like polymer chains. In particular, at high concentrations, they form a network of topological entanglements and, as a result, the solution acquires viscoelastic properties. However, compared to polymers, the micellar chains of surfactants can reversibly break and recover. However, unlike polymers in solutions, wormlike micelles undergo breaking and recombination (Chu et al., 2010). Mikel Morvan studied the viscoelastic behavior, thermal stability, and adsorption on sandstone. They suspected that WLMs could increase the 29% of oil recovery vs. water flooding (Morvan, et al., 2012a; Degre, et al., 2012; Morvan, et al., 2012b). Joris van Santvoort studied the retardation ratio of VES in the different permeability core. Flow resistance in a high permeability core was shown to be significantly higher than in a low permeability core. Increasing the concentration of surfactant and co-solute led to an enhanced resistance factor (Zhu et al., 2013; Joris van and Golombok, 2015).

Herein, a novel hybrid sample (VES and SiO_2 nanoparticle) has been studied to overcome the deficiencies of HPAM. We investigated the effects of nano-SiO_2 on the rheology of VES solutions and the influencing parameters, including surfactant concentrations, nanoparticle concentrations, particle diameters, and NaCl concentrations. We also compared the ability of the VES system in enhancing oil recovery in the presence and absence of nanoparticles. Our investigation may provide a new idea for the development of an oil recovery agent.

EXPERIMENTAL

Materials

The VES solutions used during the experiments are composed of sodium dodecyl sulfate (chemical pure) and lauroylamidopropyl betaine (industrial product) purchased from Sinopharm. SiO_2 nanoparticles with different particle diameters including 7nm, 12nm, and 22nm are those of Ludox SM, HS, and TM, were received as a gift from W. R. Grace. Sodium chloride (NaCl) was purchased from the Xilong Scientific Company. Deionized water with an electrical resistivity of 18.2MΩ·cm was used to prepare the solutions. Crude oil was obtained from the Shengli Oilfield in China, with a density and viscosity of 0.89g/cm^3 and 80.1mPa·s, respectively, at 50°C. Component analysis results show that this oil is composed of 70.66wt% saturated hydrocarbons, 20.92wt% aromatic hydrocarbons, 5.83wt% resions, and 2.59wt% asphaltene.

Rheology Test

Rheological experiments were performed using an Anton Paar MCR301 rotational rheometer. The sample temperature was adjusted using a Peltier thermostat (Chen, et al., 2012). The viscosity of the samples was measured at different temperatures and various shear rates. During the angular frequency scan, elastic and storage moduli were determined at 50°C with the same rheometer (Zhu et al., 2013).

Interfacial Tension Measurements

The dynamic IFT values were measured using a Texas-500 spinning drop tensiometer (Temco, United States) (Jiang, et al., 2014). The instrument was equipped with an image-acquisition module and the IFT value could be automatically calculated according to the length and the width of the oil droplet. The temperature for all the measurements was maintained at 50°C using a semiconductor thermostat.

Sandpack Flooding Experiment

Flooding experiments were performed at 50°C. A steel cylinder of 10 inches in height and 1 inch in inner diameter was filled with quartz sands of different sizes. The porosity of the porous sandpack was about 35.8%. The sandpack was initially saturated with synthetic brine, and then displaced by dehydrated crude oil. Water flooding was performed until the water proportion of the output fluid was higher than 98%. Afterward, the composite VES solution was injected. The difference between water flooding recovery and total recovery was calculated as the tertiary recovery increased by nanoparticle/surfactant composite flooding. The injection rate was maintained at 0.5ml/min (Zhu et al., 2013).

RESULTS AND DISCUSSION

Rheological Properties of VES

The viscosity of injected solutions plays a significant role in displacing crude oil during oilfield development. Consequently, it is necessary to clarify the rheological behaviors of nanoparticle/surfactant solutions (Zhu et al., 2013).

Figure 1 shows the viscosities of VES at the shear rate of 7.34s^{-1} against the percentage of LAB in the LAB/SDS mixture. The viscosity increases with the increasing LAB percentage on composite surfactants until the mass ratio of LAB/SDS is 3:1, after which the viscosity begins to drop. Therefore, this proportion is used in the following experiments.

Figure 2 shows that the viscosity of VES is significantly affected by salinities. The VES viscosity increases with a

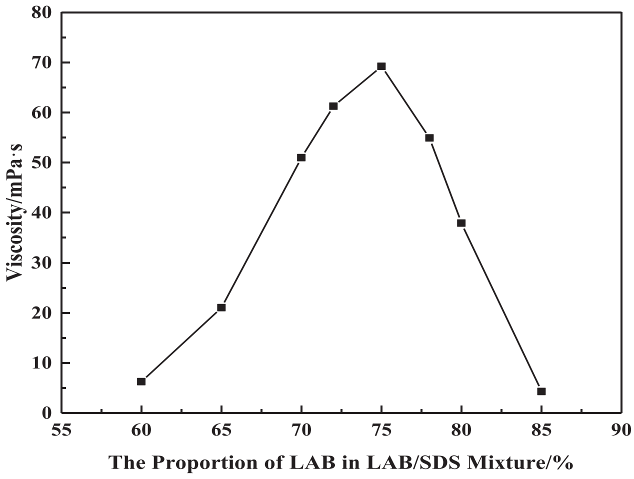

FIGURE 1 | Effect of LAB percentage on apparent viscosity (*w* (LBA + SDS) = 1%, *w* (NaCl) = 1%, T = 50°C, $\gamma = 7.34\ s^{-1}$).

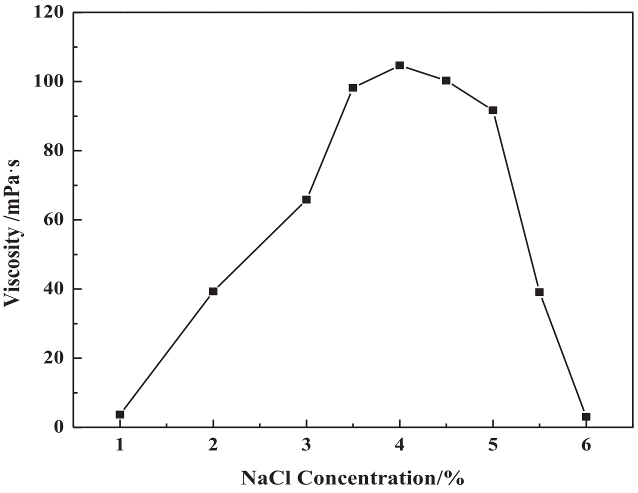

FIGURE 2 | Effect of salinity on apparent viscosity (*w* (LBA + SDS) = 0.8%, LBA:SDS = 3:1, T = 50°C, $\gamma = 7.34s^{-1}$).

salinity until 4%, before which the NaCl could compress the diffuse electric double-layer of the surfactants. And it causes more surfactant molecules to go into the micelle promoting the growth of entangled wormlike micelles. But if the salinity is more than 4%, the diffuse electric double layer would be over compressed. The surface charge of the micelle is too low, making the micelles coil and decreases the fluid dynamics radius.

The viscosity variation of the VES solution against surfactant concentration is shown in **Figure 3**. The solution viscosity increases as the concentration rises especially when the surfactant concentration is more than 0.4%. The viscosity increases from 13.79mPa·s to 185.49mPa·s as the concentration increases from 0.4% to 1%, at which concentration the micelles could form a large space structure with higher viscosity.

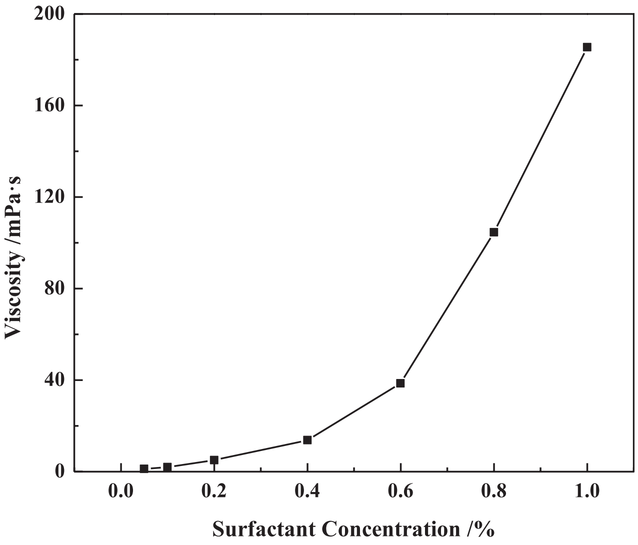

FIGURE 3 | Effect of surfactants concentration on apparent viscosity (LBA:SDS = 3:1, T = 50°C, *w* (NaCl) = 4%, $\gamma = 7.34s^{-1}$).

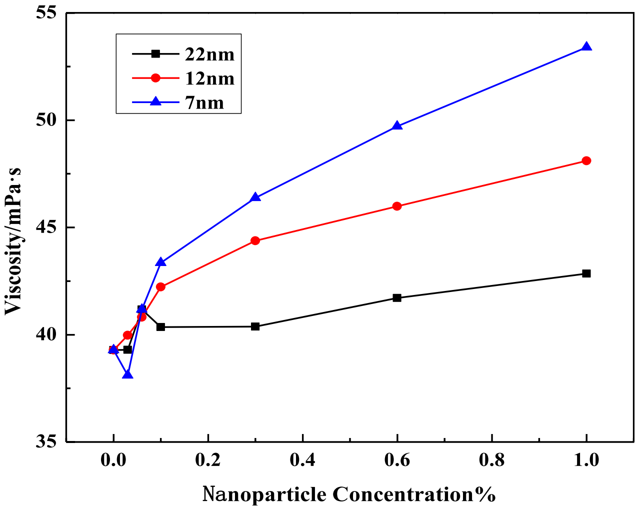

FIGURE 4 | Effect of SiO_2 concentration on solution viscosity at different nanoparticle diameters (*w* (LBA + SDS) = 0.8%, LBA:SDS = 3:1, T = 50°C, *w* (NaCl) = 2%, $\gamma = 7.34s^{-1}$).

Rheological Behaviors of VES Samples With SiO_2 Nanoparticles

The curves of VES viscosity against the nanoparticle concentration at different salinities is shown in **Figures 4–6**. It shows that the viscosity of the VES solution at the same surfactant concentration could be apparently improved. These results can be interpreted in terms of the growth of entangled wormlike micelles, followed by the formation of bilayers due to the adsorption of the headgroups on SiO_2 surfaces (Bandyopadhyay and Sood, 2005). It also shows that the SiO_2 nanoparticles with a smaller diameter have a higher viscosifying

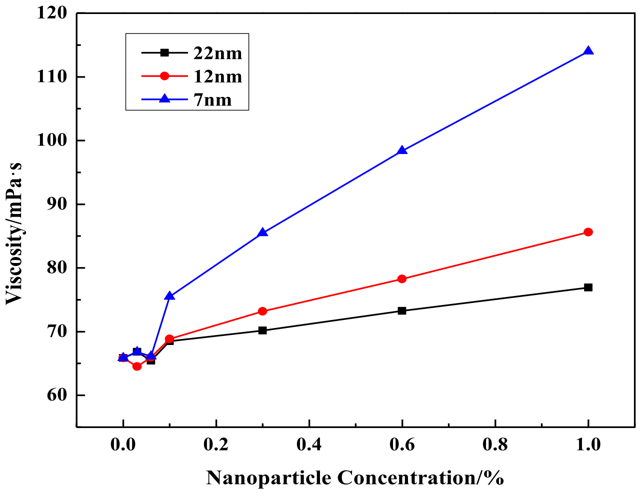

FIGURE 5 | Effect of SiO_2 concentration on solution viscosity at different nanoparticle diameters (*w* (LBA + SDS) = 0.8%, LBA:SDS = 3:1, T = 50°C, *w* (NaCl) = 3%, γ = 7.34s^{-1}).

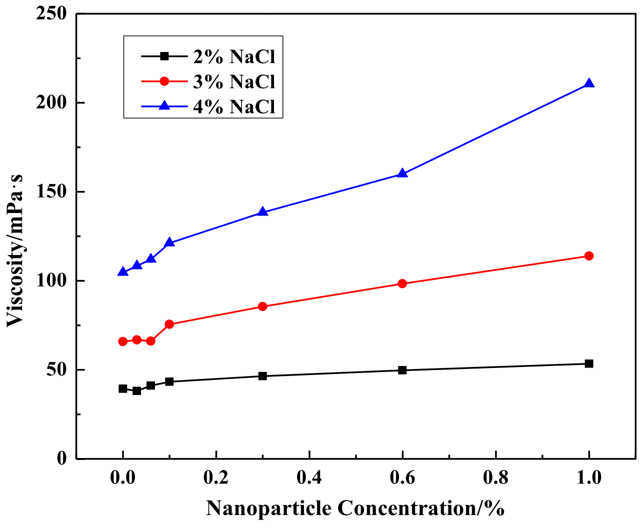

FIGURE 7 | Effect of SiO_2 concentration on solution viscosity at different salinities (*w* (LBA + SDS) = 0.8%, LBA:SDS = 3:1, T = 50°C, γ = 7.34s^{-1}).

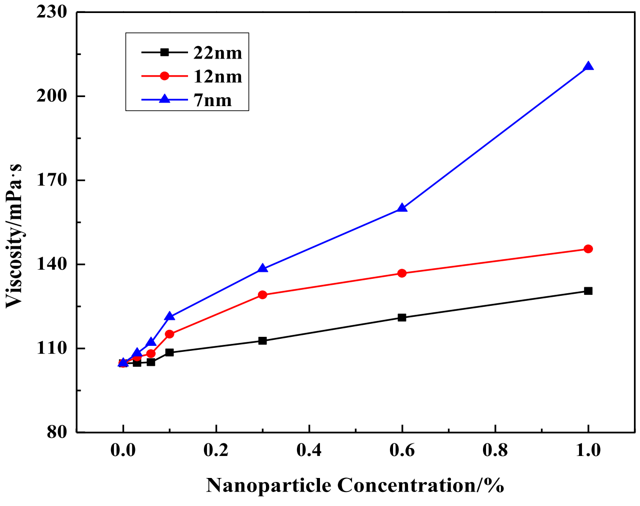

FIGURE 6 | Effect of SiO_2 concentration on solution viscosity at different nanoparticle diameters (*w* (LBA + SDS) = 0.8%, LBA:SDS = 3:1, T = 50°C, *w* (NaCl) = 4%, γ = 7.34s^{-1}).

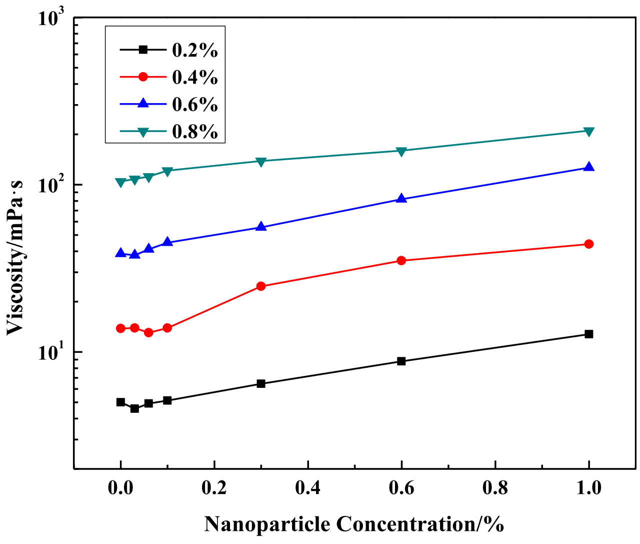

FIGURE 8 | Effect of SiO_2 concentration on solution viscosity at different surfactant concentrations (LBA:SDS = 3:1, T = 50°C, *w* (NaCl) = 4%, γ = 7.34s^{-1}).

ability. This could be attributed to the fact that smaller nanoparticles have a higher specific surface area and higher amounts at a certain concentration, which may provide more crosslinking points. Therefore, 7nm SiO_2 nanoparticles were used in the following experiments.

The influence of silica particle concentration, salinity, surfactant concentration, and temperature on VES viscosity is shown in **Figures 7–9**. In **Figure 7**, the solution viscosity increases with the rise of SiO_2 concentration and salinity. Especially, the test sample containing 4% NaCl presents a wide range of viscosity increase with the nanoparticle addition compared with the low salinity solution, which means that nanoparticles enhanced the viscosity of VES solutions by increasing the entanglements of wormlike micelles. As expected, the viscosity of the VES solution continuously decreases at increasing temperatures, which is similar to that of the system without particles.

Rheological evaluation of the viscoelastic nature of the surfactant fluids with and without nanoparticles was carried out. **Figure 10** shows the dependence of viscosity of the VES solution against the silica particle concentration on shear rate. At low shear rates, the viscosity of the VES solution without particles remains unchanged (Newtonian viscosity); then, its value

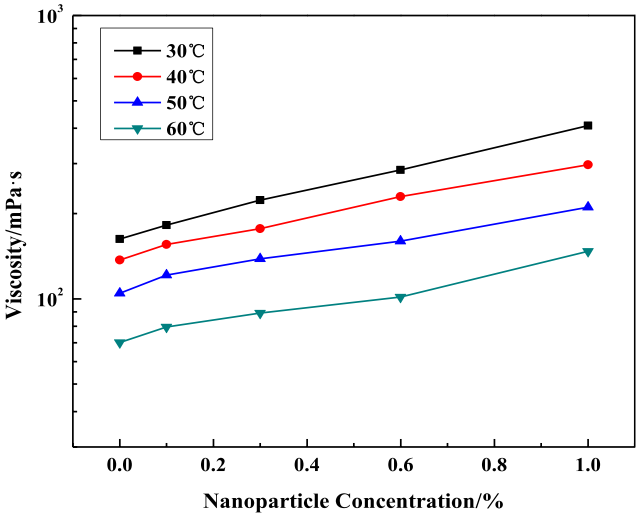

FIGURE 9 | Effect of SiO_2 concentration on solution viscosity at different temperatures (w (LBA + SDS) = 0.8%, LBA:SDS = 3:1, w (NaCl) = 4%, γ = $7.34s^{-1}$).

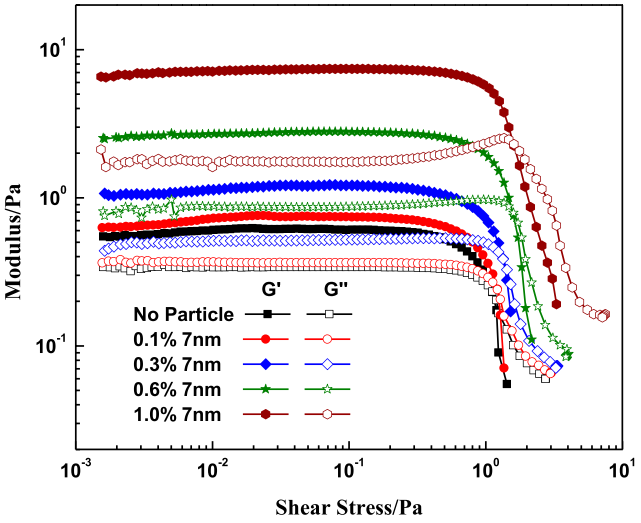

FIGURE 11 | Effect of SiO_2 concentration on the modulus with different shear stress. (w (LBA + SDS) = 0.8%, LBA:SDS = 3:1, T = 50°C, w (NaCl) = 4%, frequency = 1Hz).

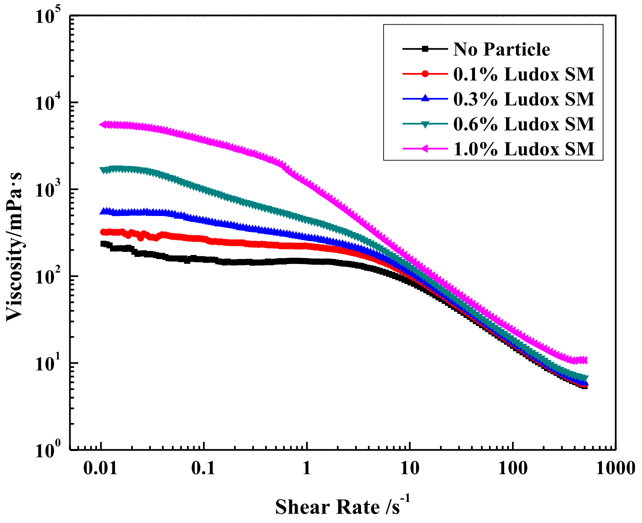

FIGURE 10 | Effect of SiO_2 concentration on solution viscosity at different shear rates. (w (LBA + SDS) = 0.8%, LBA:SDS = 3:1, T = 50°C, w (NaCl) = 4%).

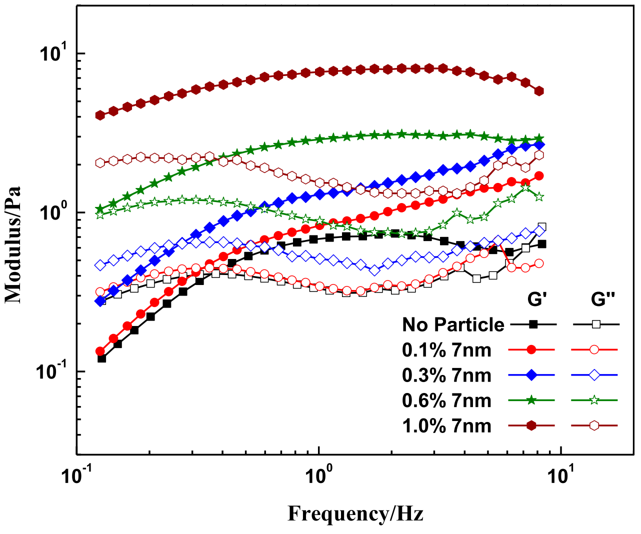

FIGURE 12 | Effect of SiO_2 concentration on the modulus at different frequencies. (w (LBA + SDS) = 0.8%, LBA:SDS = 3:1, T = 50°C, w (NaCl) = 4%, shear stress = 0.01Pa).

decreases as the shear rate grows above $2s^{-1}$, which is associated with the orientation of micellar chains along the flow direction and possibly with their partial break. And adding particles has a significant effect on the viscosity of VES solutions especially at low shear rates. Note that 1% nanoparticles increased the viscosity of the VES fluid by more than 23 times at the shear rate of $0.1s^{-1}$, which may be caused by the influence of particles on the network structure of micelles. But at high shear rates, the effect of the nanoparticles is weakened.

The G' and G" viscous-elastic behavior between the VES fluids with and without nanoparticles at different shear stress and frequency are shown in **Figures 11, 12. Figure 11** shows that G' and G" of the VES solution with or without particles were constant at low shear stress and significantly decreased at high shear stress above 0.8Pa. Within the plateau area, the system shows strong elastic properties. And in the range of high shear stress, the loss modulus exceeds the value of the storage modulus. Meanwhile, adding nanoparticles increases both G' and G", which indicated that the addition of small amounts of nanoparticles to the VES solution resulted in the strengthening of the micelle-micelle associations and elongated micelle structures in the fluids. The intersection point of the G' and

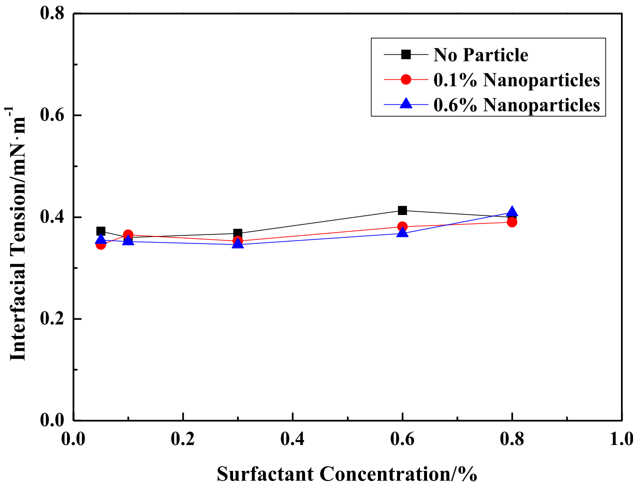

FIGURE 13 | IFT between the VES solution and Zhuangxi crude oil. **(A)** (*w* (LBA + SDS) = 0.8%, LBA:SDS = 3:1, T = 50°C, *w* (NaCl) = 4%, K = 500mD). **(B)** (*w* (LBA + SDS) = 0.8%, LBA:SDS = 3:1, T = 50°C, *w* (NaCl) = 4%, K = 1500mD). **(C)** (*w* (SiO_2) = 0.8%, *w* (LBA + SDS) = 0.8%, LBA:SDS = 3:1, T = 50°C, *w* (NaCl) = 4%, K = 400mD). **(D)** (*w* (SiO_2) = 0.8%, *w* (LBA + SDS) = 0.8%, LBA:SDS = 3:1, T = 50°C, *w* (NaCl) = 4%, K = 1900mD).

G" right shifts with the increase of the nanoparticle concentration, which indicates that the SiO_2 has a greater contribution to G'.

Figure 12 shows that at low frequency the loss modulus exceeds the value of storage modulus, and the elastic properties dominate at high frequency. Meanwhile, adding nanoparticles increases both G' and G", and the intersection point of the G' and G" left shift with the increases of the nanoparticles concentration indicates that the SiO_2 has a greater contribution to G' which is similar to the results of **Figure 11**.

Interfacial Tension Between VES Solution and Oil

Figure 13 shows the interfacial tensions between Zhuangxi crude oil and surfactant/silica solutions of various concentrations at 50°C. The interfacial tension is about 0.38mN/m within a wide range of surfactant concentrations. And adding SiO_2 has less effect on the IFT.

Oil Displacement Test

Variations of recovery factors, water contents, and displacement pressures with injected volumes are plotted in **Figure 14** of the VES solutions with and without nanoparticles in different permeability media. Sandpack parameters, flooding processes, and the oil recovery results are summarized in **Table 1**.

Less than 5% oil recovery was achieved by the VES solution without particles (**Figures 14A,4B**). As shown in **Figures 14C, 4D**, the oil recovery factor of the hybrid sample (VES solution with silica particles) was 9.68% in the high permeability zone, which was larger than that in low permeability media. In order to find the reason why VES has no effect on oil recovery, the apparent viscosity of VES with different oil ratios was measured, as shown in **Figure 15**.

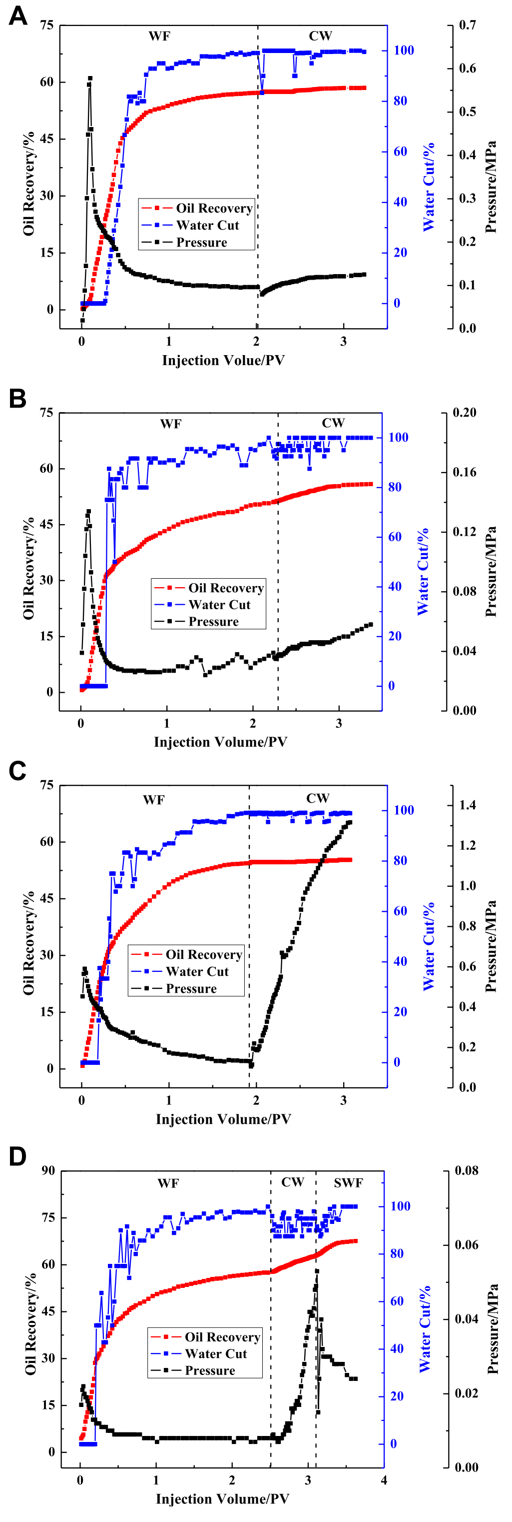

FIGURE 14 | Recovery factor, water cut, and flooding pressure plotted as a function of injected volume of different samples (WF = water flooding; CW = chemical flooding; SWF = subsequent water flooding).

It shows that oil has great influence on the apparent viscosity of the VES solution. Note that only 1% oil decreased the viscosity of the VES solution without nanoparticles by more than 97%

TABLE 1 | Sandpack parameters, displacement process, and the results of these oil displacement tests.

	0.8% (LAB:SDS = 3:1) + 4% NaCl		0.8% (LAB:SDS = 3:1) + 4% NaCl + 0.8% 7nm SiO_2	
Permeability/mD	500	1,500	400	1900
Initial oil saturation S_o/%	88.56	88.16	89.90	88.45
Primary recovery efficiency R_o/%	57.36	51.37	51.64	57.89
Secondary recovery factor R_o/%	1.14	4.52	0.25	9.68
Overall recovery efficiency R_o/%	58.50	55.89	51.89	67.57

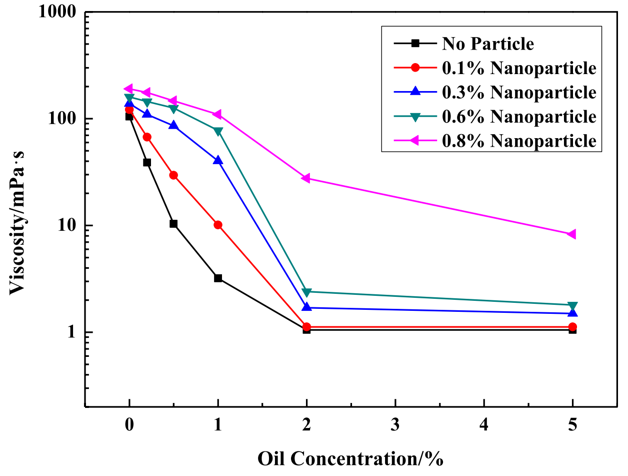

FIGURE 15 | Effect of oil ratio on apparent viscosity in different systems. (w (LBA + SDS) = 0.8%, LBA:SDS = 3:1, T = 50°C, w (NaCl) = 4%, $\gamma = 7.34s^{-1}$).

which indicates the oil break of the structure of WLMs. But the presence of particles could weaken the influence of oil. The viscosity of the system with 0.8% SiO_2 only decreased about 43% when mixed with 1% crude oil.

The poor oil recovery efficiency of the VES solution without particles may be attributed to the fact that VES micelles could be easily destructed while in residual crude oil. On the contrary, the structure of VES in the presence of nanoparticles could be significantly enhanced, and thus the water/oil mobility ratio could be improved, resulting in a higher oil recovery (Zhu et al. 2013). The main reason for the invalidity of the hybrid samples in low permeability sandpacks can be ascribed to the bridging of nanoparticles at the inlet, a consequence of the continuously increasing injection pressures.

CONCLUSION

The rheological behaviors of the VES/SiO_2 nanoparticle hybrid and sandpack flooding experiments were examined. It was found that the SiO_2 nanoparticle exhibited viscosifying action and improved oil tolerance. In addition, the poor oil recovery efficiency of the solution without nanoparticles may be attributed to the destruction of the VES micelles upon contacting the residual oil. On the contrary, 9.68% of oil recovery was achieved from the VES and nanoparticle samples in the high permeability sandpack flooding test for the VES sample with nanoparticles which is relatively stable with oil in order to produce more oil. However, the nanoparticles bridging off the sandpack inlet restrict its use in a low permeability reservoir.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/Supplementary Material, further inquiries can be directed to the corresponding author.

AUTHOR CONTRIBUTIONS

ZL, QW, and HC contributed to the conception and design of the work; QW, MG, and WL contributed to the acqusition and analysis of data for the work; QW, MG, and WL drafted the work; ZL and HC revise the work critically; All the five authors made the final approval of the version to be published; All the five authors made the agreement to be accountable for all aspects of the work in ensuring that questions related to the accuracy or integrity of any part of the work are appropriately investigated and resolved.

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