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To cite this article: Hongda Zhou *et al* 2019 *IOP Conf. Ser.: Earth Environ. Sci.* **218** 012153

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Tuning the Electrical Properties of Nanoparticles and Application in the EOR Process of Ultra-low Permeability Reservoirs

Hongda Zhou, Rui Cheng, Caili Dai*, Yuyang Li, Mingwei Zhao*, Wenjiao Lv

¹ School of Petroleum Engineering, State Key Laboratory of Heavy Oil, China University of Petroleum (East China), Qingdao, Shandong 266580, China

*Corresponding author's e-mail: daicl306@163.com, zhaomingwei@upc.edu.cn

Abstract. In this study, electronegative silica nanofluid and electropositive alumina nanofluid were prepared to enhance oil recovery of ultra-low permeability cores. Transmission electron microscopy and dynamic light scattering were employed to characterize their morphology and size. Nanofluids with different concentrations, ranging from 0.001wt% to 0.2 wt%, were used for spontaneous imbibition tests of sandstone cores to evaluate the performance of these nanofluids for EOR. The wettability alteration experiment and zeta potential measurements were employed to investigate the mechanism of nanofluids for EOR. As the concentration of alumina nanofluid increases, the adsorption of alumina nanoparticles could cause pore blocking. On the contrary, electronegative silica nanofluid could improve displacement efficiency and oil recovery as the increase of concentration of silica nanofluid. Electronegative silica nanofluid has great potential for EOR in ultra-low permeability reservoir.

1. Introduction

In recent years there has been an increasing interest in application of nanomaterials in petroleum industry. Nanofluids as a new kind of chemical agent have been widely applied in enhanced oil recovery because of their excellent characters.[1]

The nanoparticles dispersed in a liquid phase were proposed by Choi[2] to improve the heat transfer performance of the liquid. Nanofluid, by definition, is traditional fluid such as oil, water or ethylene glycol that is dispersed by nanoparticle which has average size less than 100 nm .[3] It has been shown that, recently, the application of nanoparticles (NPs) for EOR purposes has been tested.[4-6] Nanoparticles can improve some interaction characteristics between fluid and the rock such as wettability. Meanwhile, with the addition of nanoparticles, some of the fluid properties, such as density, particle size of the displacing phase (water) and interfacial tension (IFT) could be enhanced.

The most common type of NPs used for EOR is silica nanoparticles and alumina nanoparticles.[1, 7] In the past decade, many laboratory experiments were designed to investigate the performance of nanoparticles.[8] Ju et al.[9] reported that polysilicon with nanometer range could alter the wettability of porous media surfaces of reservoir rock, impacting the flow behavior of oil and water phase in porous media body. Hendraningrat, L et al.[10] has studied some parameters which affect EOR processes, such as particle size, concentration and original core wettability with the application of hydrophilic silica nanoparticles. Thus, oil recovery increases by decreasing nanoparticle size and



injection rate, they concluded. Onyekonwu et al.[11] investigated the ability of three different nanoparticles to improve oil recovery. They came to the conclusion that LHPN (dispersed in ethanol) and neutrally wet polysilicon nanoparticles are effective agents for increasing oil recovery in water-wet structure.

In this work, electronegative silica nanofluid and electropositive alumina nanofluid were compared and used to enhance oil recovery of ultra-low permeability cores. Different charges on the surface of nanoparticle can affect the adsorption of nanoparticles on the surface of rock. Electropositive alumina nanoparticles could be easier adsorbed on the electronegative pore wall, which may lead pore blocking. Electronegative silica nanofluid has great potential for EOR in ultra-low permeability reservoir.

2. Experimental Section

2.1. Materials and Apparatus

Silica and alumina nanoparticles were bought from Aladdin Reagent Co., Ltd., China, with an average diameter about 10 nm and 12 nm, respectively. The oil phase was the mixture of kerosene and dehydrated crude oil with a volume ratio of 18:1, with the density of 0.824 g/cm³, and its dynamic viscosity was about 5.5 mPa s at 25 °C. The NaCl solution (3 wt %) was prepared as reservoir brine. The density and dynamic viscosity of brine were 1.025 g/cm³ and 0.90 mPa s at 25 °C, respectively. Natural sandstone cores were bought from Haiyan Oil Scientific Research Apparatus Co., Ltd. The porosity and gas permeability of cores were 15% and 0.5 mD, respectively. The detailed parameters of cores were shown in Table 1.

Transmission electron microscope (TEM, Tecnai-G20, FEI, U.S.A.) images were obtained to observe the structure of nanoparticles. Dynamic light scattering (DLS) and Zeta potential measurements were conducted using a NanoBrook Omni laser particle size analyzer (Brookhaven, U.S.A.). The interfacial tension was measured at 60 °C by TX-500C spinning drop interfacial tension meter (Bowling, Stafford, TX, U.S.A.). The contact angle measurement was conducted by a contact angle measuring system (Tracker, Teclis, France).

Table 1. parameters of cores

Silica nanofluid	Core numbers	Permeability (mD)	Porosity (%)	alumina nanofluid	Core numbers	Permeability (mD)	Porosity (%)
0.2 wt %	A11	0.42	15.2	0.2 wt %	B11	0.55	14.8
0.1 wt %	A12	0.51	14.7	0.1 wt %	B12	0.47	14.2
0.05 wt %	A13	0.47	15.6	0.05 wt %	B13	0.52	15.1
0.01 wt %	A14	0.53	15.1	0.01 wt %	B14	0.46	14.6
0.005 wt %	A15	0.56	15.7	0.005 wt %	B15	0.44	14.9
0.001 wt %	A16	0.41	14.9	0.001 wt %	B16	0.43	14.7
brine	A17	0.55	15.3				

2.2. Preparation of Nanofluids

First, two kinds of nanoparticles were added to water to obtain nanofluids, ranging from 0.001 % to 1 %. Then, pH of nanofluids were adjusted to optimum value with 1 mol/L NaOH solution. After 2 h of ultrasonic vibration, the clear nanofluids were obtained.

2.3. Spontaneous Imbibition Tests

These cores (length 25 mm and diameter 25 mm) were dried in an oven to remove bound water at 80 °C for 24 h. Then, the dried cores were saturated with oil under the pressure of 15 MPa for 24 h. These prepared cores were immersed in oil at 60°C for 24 h to prevent the high temperature influence on the oil volume. These cores were then immersed in brine and nanofluids with different

concentrations in different imbibition devices at 60°C. The volumes of oil discharged from the cores were recorded against time.

3. Results and Discussion

3.1. Characterization of Nanoparticles

Figure 1 shows the TEM images of silica and alumina nanoparticles. These two kinds of nanoparticles showed similar spherical-like microstructure. Meanwhile, the sizes of nanoparticles ranged from 10 to 20, which indicated that the nanoparticles have good ability to disperse in water.

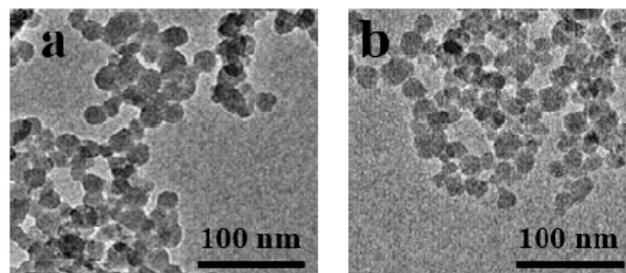


Figure 1. TEM images of silica nanoparticles (a) and alumina nanoparticles (b)

3.2. Dispersion of Nanoparticles

The clear and transparent nanofluids based on nanoparticles were obtained after ultrasonic vibration (Figure 2). The size distribution of these two kinds of nanoparticles in water was measured by DLS. The

silica nanofluid has an average size of 13 nm, while the average particle size of alumina nanoparticles in water was 20 nm. Although there were no nanoparticles aggregation in both nanofluids, the silica nanoparticles have a better dispersion in water. Meanwhile, the Zeta potential of silica nanoparticles or alumina nanoparticles in water was -42 mV and -37 mV, which was indicative of well-dispersed nanofluids.

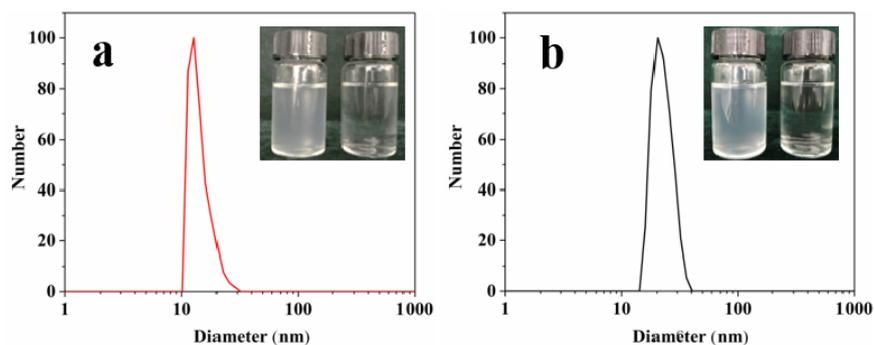


Figure 2. Size distribution of silica nanoparticles (a) and alumina nanoparticles (b) in water

3.3. Interfacial Activity of Nanofluids

To evaluate the influences of silica and alumina nanoparticles on oil-water interfacial properties, oil-water interfacial tensions between the silica nanofluid or alumina nanofluid and prepared oil were measured (Figure 3). Compared with alumina nanofluid, silica nanofluid owned a greater ability to reduce interfacial tension. The oil-water interfacial tension was 29.8 mN/m at 60 °C, 0.2 wt % silica nanofluid and alumina nanofluid could reduce it to 21.2 and 25.6 mN/m, respectively. The results indicated that electronegative silica nanoparticles have better ability to reduce oil-water interfacial tension.

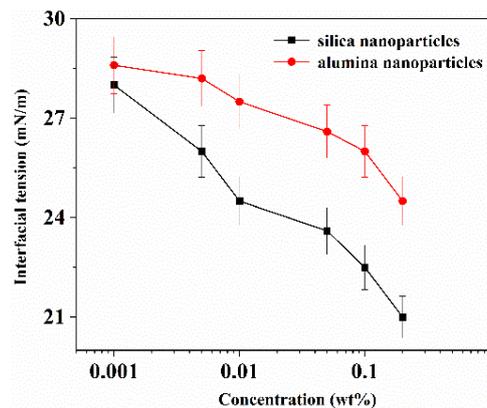


Figure 3. The oil-water interfacial tension of different nanofluids with different concentrations at 60 °C. Error bar = RSD (n = 5)

3.4. Oil Displacement from a Solid Surface

Different liquid phases (0.2 wt % silica nanofluid, 0.2 wt % alumina nanofluid and brine) were used to verify oil displacement from a solid surface. The glass substrate was treated by paraffin to obtain an oil-wet surface, the contact angle of the oil droplet captured on the surface was recorded with time. Figure 4 shows the influence of different fluids for the oil contact angle on the treated glass surface. The results indicated that the contact angle could be obviously changed by 0.2 wt % silica nanofluid. The angle was changed from 55° to 102°, while the contact angle in 0.2 wt % alumina nanofluid was changed from 60° to 83°. Figure 4b and 4c shows the wettability alteration of the surface treated by different fluids. According to the images, electronegative silica nanoparticles have a greater ability to alter wettability than electropositive alumina nanoparticles.

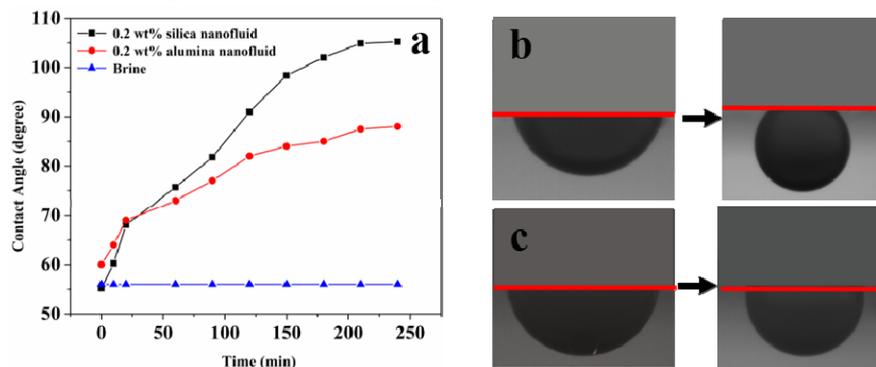


Figure 4. Influence of different liquid phases for the contact angle (a); oil droplets on oil-wet glass surface treated by silica nanofluid (b) and alumina nanofluid (c)

3.5. Spontaneous Imbibition Tests

To evaluate the performance of these nanofluids with different electrical properties for EOR, different nanofluids (0.2 wt % silica nanofluid, 0.2 wt % alumina nanofluid and brine) were used for spontaneous imbibition tests of sandstone cores. As shown in Figure 5, Oil recovery using different nanofluids increased with the increasing of nanofluid concentration. Compared with alumina nanofluid, silica nanofluid had a better to improve oil recovery and the speed of oil recovery was higher. As the concentration increases, the speed of oil recovery by alumina nanofluid reduced.

According to the results, different charge on the surface of nanoparticle can affect the adsorption of nanoparticles on the surface of rock. Electropositive alumina nanoparticles could be easier adsorbed on the electronegative pore wall. As the concentration of alumina nanofluid increases, the adsorption

of alumina nanoparticles could cause pore blocking. On the contrary, electronegative silica nanofluid could improve displacement efficiency and oil recovery as the increase of concentration of silica nanofluid. Hence, electronegative silica nanoparticles have a greater potential for EOR in ultra-low permeability reservoir.

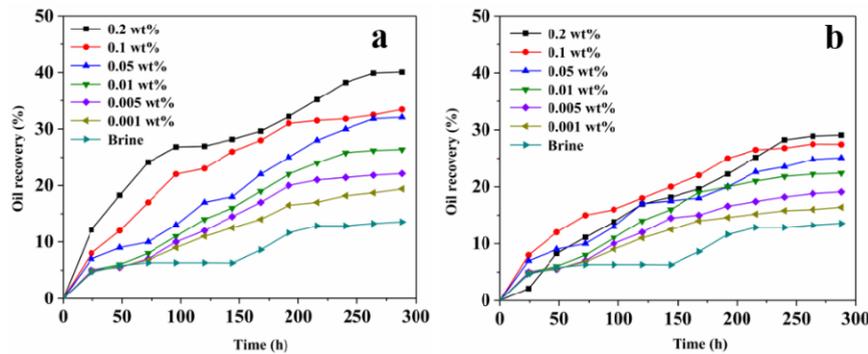


Figure 5. Influence of the concentration of silica nanofluid (a) and alumina nanofluid (b) for oil recovery in cores at 60 °C

4. Conclusions

In this study, electronegative silica nanofluid and electropositive alumina nanofluid were compared and used to enhance oil recovery of ultra-low permeability cores. Dispersing experiments indicated that silica nanoparticles and alumina nanoparticles had well dispersity and stability. The results of spontaneous imbibition tests indicated that oil recovery increased with the increase of silica nanofluid concentration or the decrease of alumina nanofluid concentration. Different charge on the surface of nanoparticle can affect the adsorption of nanoparticles on the surface of rock. Electropositive alumina nanoparticles could be easier adsorbed on the electronegative pore wall, which may lead pore blocking. Electronegative silica nanofluid has great potential for EOR in ultra-low permeability reservoir.

Acknowledge

This work was financially supported by the National Key Basic Research Program (No. 2015CB250904), the National Science Fund (U1663206, 51425406), the Chang Jiang Scholars Program (T2014152), Climb Taishan Scholar Program in Shandong Province (tspd20161004).

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