



Mechanistic study of wettability alteration of oil-wet sandstone surface using different surfactants



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ABSTRACT

Different analytical methods including Fourier transform infrared (FTIR), atomic force microscopy (AFM), zeta potential measurements, contact angle measurements and spontaneous imbibition tests were utilized to make clear the mechanism for wettability alteration of oil-wet sandstone surface using different surfactants. Results show that among three types of surfactants including cationic surfactants, anionic surfactants and nonionic surfactants, the cationic surfactant CTAB demonstrates the best effect on the wettability alteration of oil-wet sandstone surface. The positively charged head groups of CTAB molecules and carboxylic acid groups from crude oil could interact to form ion pairs, which could be desorbed from the solid surface and solubilized into the micelle formed by CTAB. Thus, the water-wetness of the solid surface is improved. Nonionic surfactant TX-100 could be adsorbed on oil-wet sandstone surface through hydrogen bonds and hydrophobic interaction to alter the wettability of oil-wet solid surface. The wettability alteration of oil-wet sandstone surface using the anionic surfactant POE(1) is caused by hydrophobic interaction. Due to the electrostatic repulsion between the anionic surfactant and the negatively charged surface, POE(1) shows less effect on the wettability alteration of oil-wet sandstone surface.

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1. Introduction

Currently, many of the world's reservoirs have low oil recoveries and different methods have been used to enhance oil recovery. Among these methods, wettability modification has played a huge role in the oil exploitation [1,2]. Wettability has long been defined as "the tendency that the fluid is attached to the solid, which is a nature of adsorption" [3]. More and more studies show that there are both water-wet and oil-wet reservoirs in the world. Moreover, there is a wide range of changes from strong water-wet conditions to strong oil-wet conditions for the reservoir wettability and there are also various degrees of neutral wetting reservoirs [4,5]. Wettability is a fundamental parameter of the physical property of the reservoir, affecting the relative permeability of the aqueous phase, the distribution of residual oil in the reservoir, characteristics of waterflooding exploitation and the recovery of crude oil [6–8]. Various processes [9–11] must be applied to alter the wettability of reservoirs for enhancing oil recovery [1,12–16].

So far two methods have been used to change the wettability of reservoirs: (1) chemical treatment for wettability alteration, and (2) thermally inducing changes in wettability [6]. This paper looks into

the wettability alteration of oil-wet solid surface using different surfactants.

Much work has been done on wettability alteration of the reservoir by different surfactants. Three types of surfactants including cationic surfactants [6,17–20], anionic surfactants [6,17,18,21,22] and nonionic surfactants [6,18,23] have been used to alter the wettability of oil-wet solid surface. Reservoir wettability has a great effect on oil recovery. Generally speaking, the stronger the hydrophilicity of the rock surface, the higher the spontaneous imbibition recovery. While for the waterflooding process, the lowest residual oil saturations could happen under near neutral-wet conditions after injection of many pore volumes of the displacing fluid [24]. To achieve the maximum oil recovery, a suitable surfactant could be chosen to alter the wettability of the solid surface. Although a series of problems will be produced by the adsorption of surfactants on the solid surface during the wettability alteration process, some methods could be used to alleviate this problem. Nonionic surfactants could be added into the surfactant solutions, which could reduce the adsorption amount of the surfactants and improve the performance of the surfactant system.

So far, the mechanism for wettability alteration of oil-wet solid surface using different surfactants has not been studied thoroughly and different assumptions have been proposed [17,25–28]. Standnes and Austad [17] suggested that the positively charged surfactant and the carboxylic acid group from crude oil could interact

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Nomenclature

L	length (cm)
D	diameter (cm)
k	core permeability ($10^{-3} \mu\text{m}^2$)
PV	pore volume (mL)
S_{oi}	initial oil saturation (fraction, %)
wt%	weight %
IFT	interfacial tension (mN/m)
CMC	critical micelle concentration
OOIP	original oil in place

to form an ion pair, then the ion pair is desorbed from the solid surface and solubilized into the micelle formed by the cationic surfactants. Thus, the wettability of oil-wet solid surface is changed toward water-wet conditions. This mechanism is called “ion-pair mechanism”, while another mechanism called “surfactant adsorption mechanism” was also proposed by them, stating that the adsorption of anionic or nonionic surfactants on oil-wet surface is caused by the hydrophobic interaction between tails of anionic or nonionic surfactants and hydrophobic substance from crude oil adsorbed on the solid surface. The wettability of the solid surface could also be altered toward water-wet state through the adsorption of surfactants on oil-wet solid surface.

Many studies have been focused on the wettability alteration of carbonates caused by different surfactants in the past few years [6,15,18,29,30]. Many useful experimental methods including contact angle measurement, spontaneous imbibition test, zeta potential measurement, IR determination and AFM studies have been utilized to study the mechanisms of wettability alteration of oil-wet carbonate surfaces. Functional groups of organic compounds on the solid surface can be detected using IR. Morphology and roughness changes of solid surfaces after treatment with different substances could be observed by AFM. The results of contact angle measurements, spontaneous imbibition tests and zeta potential measurements could help to analyze and to confirm the results of IR and AFM. Golabi [6,18] studied the effect of various surfactants on wettability alteration in the oil–water–limestone system through contact angle measurements. Gupta [30] studied the effect of salinity, surfactant concentration, electrolyte concentration and temperature on the wettability alteration of the calcite surface and identified underlying mechanisms through the contact angle measurement, ATR-IR determination and spontaneous imbibition tests. Jarrahian [29] utilized different experimental methods to verify the proposed mechanisms for the wettability alteration of carbonates. In their studies, the IR determinations detected different chemical bonds after the aging and surfactant treatment process while the AFM studies confirmed the morphology changes of solid surfaces and the zeta potential measurements indirectly verified the presence of different components on the solid surface. However, very few emphases have been laid on wettability alteration of sandstones caused by different surfactants and experimental methods have been rarely used to verify the above mechanisms.

The purpose of this paper is to explore the mechanism for wettability alteration of oil-wet sandstones by different surfactants through laboratory means. Several materials, such as quartz plate, mica plate and quartz sand (to simulate sandstones), cationic surfactant CTAB, nonionic surfactant TX-100 and anionic surfactant POE(1) were used in this study. Different analytical methods including FTIR, AFM and zeta potential measurements were utilized to study the mechanism of the wettability alteration of oil-wet sandstone surface. Contact angle measurements and spontaneous imbibition tests were used to verify the above proposed mechanisms.

Table 1

Basic data of cores.

Core number	L (cm)	D (cm)	Porosity (%)	k ($10^{-3} \mu\text{m}^2$)	S_{oi} (%)
h-1	5.02	2.50	22.50	92.10	70.10
h-2	5.07	2.50	22.30	91.95	70.05
h-3	5.05	2.50	22.40	92.05	70.20

The table shows the basic data of cores and the cores that were used in the following imbibition studies.

2. Experimental

2.1. Materials

2.1.1. Solid surface

For the solid materials, quartz plates and mica plates used in the present study have dimensions of $20 \times 20 \times 2$ mm and $15 \times 15 \times 0.15$ mm, respectively. The analytical-grade quartz powder is purchased from Sinopharm Chemical Reagent Co., Ltd. and the basic data of the cores used in this study are shown in Table 1.

2.1.2. Oil phase

Crude oil used in the aging process was obtained from Shengtuo Oilfield, China. The density of the crude oil was 0.86 g/cm^3 and the crude oil viscosity determined at ambient temperature was 61.5 mPa s . In addition, the acid number of the crude oil was 1.42 mg of KOH/g.

2.1.3. Aqueous phase

For the reagents, *n*-heptane, NaCl, CaCl_2 and $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ are analytical-grade. Deionized water was used to prepare different solutions and the ionic composition of the formation water used in this study is shown in Table 2.

2.1.4. Surfactants

For the surfactants, CTAB and POE(1) were analytical-grade and TX-100 was chemically pure grade. All the surfactants used in the present study were obtained from Sinopharm Chemical Reagent Co., Ltd. The surfactants concentrations were fixed at 0.30 wt% during IR, AFM and imbibition tests. The structures of surfactants used in this study are shown in Fig. 1.

2.2. Methods

2.2.1. Samples handling

Quartz (mica) plates or quartz sand handling: The samples were first soaked in the chromic acid for 24 h and then washed by deionized water. After that, the samples were poured into the oil and aged in an oven at 75°C for 5 days [31]. After being aged, the quartz sand was vacuum-filtered. Then the plates or sand were washed with *n*-heptane until the solution was visually colorless. Finally, the samples were dried overnight.

Cores handling: New cores were first dried and then saturated with the formation water under vacuum. The porosity and permeability of the cores were then determined at room temperature.

Table 2

Ionic composition of the formation water.

Component	Concentration (mg/L)
Na^+	6907.6
Ca^{2+}	421.2
Mg^{2+}	223.5
Cl^-	12521.0

The table shows the ionic composition of the formation water and the formation water that were used to prepare different solutions.

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