



Full Length Article

Salinity-dependent adhesion of model molecules of crude oil at quartz surface with different wettability



Fanghui Liu^{a,b}, Hui Yang^{a,*}, Jingyao Wang^c, Minghui Zhang^a, Ting Chen^a, Guangxin Hu^a, Wei Zhang^a, Jiangzhong Wu^c, Shijing Xu^c, Xu Wu^d, Jinben Wang^{a,*}

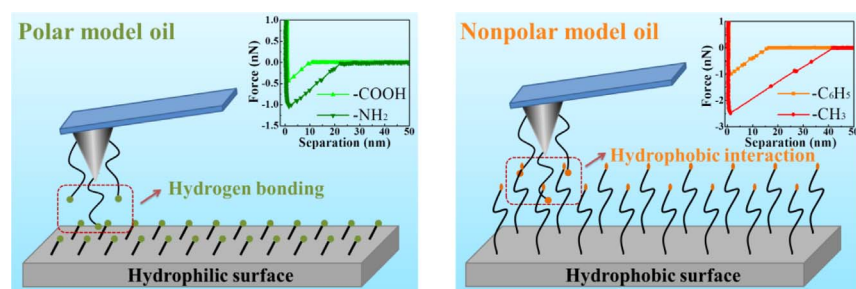
^a CAS Key Lab of Colloid, Interface and Chemical Thermodynamics, Institute of Chemistry, Chinese Academy of Sciences, Beijing 100190, PR China

^b University of Chinese Academy of Sciences, Beijing 100049, PR China

^c State Key Laboratory of Enhanced Oil Recovery, Research Institute of Petroleum Exploration and Development of PetroChina, Beijing 100083, PR China

^d Department of Chemistry and Chemical Engineering, Guangzhou University, Guangzhou, Guangdong 510006, PR China

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:

Low salinity effect
Specific functional group
Adhesion forces
Wettability

ABSTRACT

The adhesion forces between model chemical moieties of oil molecules (CH_3 -, C_6H_5 -, COOH -, and NH_2 -) and quartz surfaces with different wettability were investigated by the aid of chemical force microscopy at low and high salinity (10 mM and 100 mM). Our results showed that the adhesion force on hydrophilic surfaces decreased as in the order: $-\text{NH}_2 > -\text{COOH} > -\text{C}_6\text{H}_5 \approx -\text{CH}_3$, due to the contribution of hydrogen bonding interactions between polar groups of functionalized tips and Si-OH groups of quartz surfaces. By comparison, the adhesion force decreased almost in an opposite order: $-\text{CH}_3 > -\text{C}_6\text{H}_5 > -\text{COOH} \approx -\text{NH}_2$, in the case of hydrophobic surfaces, attributed to the strong trend of hydrophobic interactions between nonpolar oil groups and hydrophobic surface (modified with alkyl chains). A high potential of low salinity effect on improving oil recovery can be found in polar oil system in the presence of hydrophilic surfaces, as well as in nonpolar oil system in the presence of hydrophobic surfaces, because of the significantly reduced adhesion forces from high to low salinity environment. Thus it can be concluded that for hydrophilic reservoirs, the low salinity effect may be remarkable in the case of crude oil containing more polar components. And for hydrophobic reservoirs, crude oil with more nonpolar components would exhibit a significant low salinity effect. All these provide a better understanding in the molecular level for low salinity water flooding applications.

* Corresponding authors.

E-mail addresses: yanghui@iccas.ac.cn (H. Yang), jbwang@iccas.ac.cn (J. Wang).

1. Introduction

In recent decades, low salinity waterflooding has become one of the most successful enhanced oil recovery (EOR) techniques because it shows unique advantages compared with conventional chemical EOR methods, such as efficient [1–3], cost-effective [2,4,5], and environmentally friendly [4–6]. Based on published literatures, different mechanisms, including multi-component ionic exchange (MIE) [7–9], pH modification [10,11], double layer effects [12–14], osmotic pressure [10,15,16], and wettability alteration [16–18], have been proposed to explain the low salinity effect (LSE). Although, tremendous efforts have been made to understand such effect, controversial issues have emerged and molecular level processes that control EOR have not been completely revealed [19–23]. Some researchers believed that wettability alteration is the main contributor to LSE in oil/brine/rock systems; [24–26] however, other reports do not support the previous observation. For example, it was reported that the potential for EOR by low salinity flooding both through laboratory measurements and a field test was low, attributed to different wetting conditions of reservoirs [27,28]. Until now, the gap between such experimental observation and the mechanism(s) to trigger LSE in the presence of different pore surface chemistry has not been fully understood, arousing broad interest both in technological and theoretical fields.

To address the above questions, we designed a series of experiments to investigate the adhesion behaviors of oil/brine/rock systems at a molecular level. Hydroxyl and methyl terminated self-assembled monolayer (SAM) surfaces were prepared to create hydrophilic and hydrophobic surfaces, respectively [29,30]. Hydrocarbon petroleum model compounds with different polarity (including $-\text{CH}_3$, $-\text{C}_6\text{H}_5$, $-\text{COOH}$, and $-\text{NH}_2$) were used to chemically modified atomic force microscope (AFM) tips. NaCl solutions at low and high salinity were chosen to be the injected bulk. By the aid of chemical force microscopy, forces between the functionalized tip and substrate at the scale of nN (NanoNewtons) can be directly measured. The difference in adhesion behaviors of model molecules representative of crude oil on the surfaces with different wettability as a function of salinity can be achieved, providing a new insight into the mechanisms behind low salinity effects and a scientific basis for improving oil recovery.

2. Experimental

2.1. Materials

2.1.1. AFM tips functionalization

The AFM tips (NPG-10, Bruker Corporation) with a cantilever that have a nominal spring constant of 0.06 N/m, were coated with gold. Tip parameters, such as tip radius, were provided by Bruker Corporation and the sectional area was calculated as suggested by previous reports [31,32]. To mimic different kinds of crude oil, four model oil samples having functional groups were used to prepare the functionalized tips. Before creating the functionalized tips, new tips were cleaned with Milli-Q grade water made by Milli-Q grade water machine with a resistivity of 18.2 M Ω -cm for 3 times and rinsed with ethanol (AR, 99%) for 5 min, and then cleaned in plasma (CIF CPC-A, 220 V, 50 Hz) for 10 min after nitrogen drying to remove any contaminations. The clean probes were submerged in an ethanol solution, including 11-mercaptoundecanoic acid, 11-amino-1-undecanethiol hydrochloride, 1-octadecanethiol, and 10-phenyl-1-dodecanethiol at 1 mM, for at least 24 h to get the adequately functionalized COOH-terminated, NH_2 -terminated, CH_3 -terminated, and C_6H_5 -terminated tips, respectively [33,34]. Finally, the functionalized tips were rinsed in ethanol (AR, 99%) and Milli-Q grade water for 3 times, separately, and dried with pure nitrogen.

2.1.2. Aqueous solutions

In all experiments, the force curves were performed in NaCl aqueous solutions. The high salinity (HS) aqueous solution was prepared by dissolving NaCl (AR, 99.5%) with Milli-Q grade water at the concentrations of 100 mM. The low salinity (LS) solution was obtained through diluting the HS solution by 10 times. All the pH values of the solutions were adjusted to 6.5 ± 0.05 using HCl or NaOH by pH meter (Mettler Toledo FE20).

2.1.3. Substrates functionalization

To ensure the desired smoothness and controllable surface wettability, quartz was chosen to be the substrate. The hydroxyl structure at quartz surface makes it easy to modify the surface by self-assembled monolayer (SAM) [35,36]. Before creating the functionalized substrates, all quartz slides were cleaned with piranha solution (7:3 $\text{H}_2\text{SO}_4/\text{H}_2\text{O}_2$) at 70 °C for 30 min, and rinsed with ethanol (AR, 99%) and Milli-Q grade water for more than 3 times, and then cleaned the front and back in plasma for 10 min after nitrogen drying. To produce hydrophilic surfaces, the cleaned quartz slides were placed in plasma for 10 min before each experiment. For hydrophobic surfaces, the cleaned quartz slides were plasma-treated and timely reacted with octadecyltrichlorosilane (OTS, 95%) at 5% (w/v) solution in chloroform (AR, 99.8%) for 4 h. OTS-treated substrates were rinsed in a 5 min sonication bath of chloroform and then heated at 120 °C for 1 h [37,38]. Plasma-treated quartz was fully water wettable ($\theta \approx 0^\circ$), while OTS-modified quartz was poorly water wettable ($\theta = 102.5^\circ \pm 2.1^\circ$), as assessed by contact angle measurements, as shown in Fig. S1.

2.2. Methods

2.2.1. Atomic force microscope

Forces between model oil molecules and surfaces in the presence of different salinity water were measured with an O-ring liquid cell in contact mode by a Multimode VIII atomic force microscope (Digital Instruments, Bruker, USA) as previously described [39,40]. Before measuring force curves, 1 mL NaCl solution was injected and kept for 5 min. To ensure the credibility of the experimental data, force curves of each group were repeated more than 100 times in different regions. Obtained curves were analyzed by NanoScope Analysis software provided by Bruker and one tip-surface force curve can be achieved through fitting and computing. The adhesion forces can calculate from the deflection on the y axis of each force curve through the “Find Peak” function of the software [41]. Subsequently, the histograms of adhesion force were obtained by statistics.

2.2.2. Contact angle

The water contact angles (CA) of functionalized substrates were measured by the CA goniometer (OCA 20L, Dataphysics Inc.) with a sessile-drop method. During the experiment, 2 μL water drop gradually approached and remained on the surfaces with an aging time of about 5 min. The contact angle values can be obtained from video snapshots using a tangent-fitting method in data analysis software (SCA 22). Contact angles can be determined with a relative accuracy of $\pm 0.1^\circ$.

3. Results and discussion

3.1. Interactions between functionalized tips and hydrophilic surfaces

Fig. 1 shows the approach force curves between different tips and hydrophilic surfaces in the presence of NaCl solution at different concentrations. At low salinity, repulsive force can be observed in the case of CH_3 -terminated or C_6H_5 -terminated tips, but for COOH-terminated or NH_2 -terminated tips, the attractive force acts as a leading force at the

Download English Version:

<https://daneshyari.com/en/article/6631300>

Download Persian Version:

<https://daneshyari.com/article/6631300>

[Daneshyari.com](https://daneshyari.com)