



Full Length Article

Understanding desorption of oil fractions from mineral surfaces

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ABSTRACT

Separation of heavy hydrocarbons from mineral surfaces is highly dependent on the oil composition and their host rock surface properties. Herein, the petroleum is divided into SARA fractions (saturates, aromatics, resins, asphaltenes) to investigate their desorption behaviors on different types of mineral surfaces (silica (SiO₂), kaolinite (Al₂Si₂O₅(OH)₄) and calcium carbonate (CaCO₃)). The Quartz Crystal Microbalance with Dissipation (QCM-D) tests show that the saturates and aromatics could desorb from the mineral surfaces spontaneously even in water, while no desorption was observed for the asphaltenes and resins. Although the above desorption could be enhanced by alkaline or surfactant solutions, great difference still appears to different oil fractions. Oil characterization shows that the heavy fractions (i.e., asphaltenes, resins) possess richer acid groups than those of light fractions, allowing the stronger affinity of heavy fractions to the mineral surfaces through polar and chemical interactions. Additionally, the heavy fractions dominate in determining the desorption properties of bitumen (the mixed fractions), and lead to more significant wettability alteration to the mineral surfaces. Furthermore, the oil fractions desorption is also found to be highly influenced by the mineral types. Compared with silica, kaolinite has stronger affinity to the heavy oil fractions, leading to smaller amount of desorption. While on the calcium carbonate surface, less than 11% of the coated oil fractions are observed to be desorbed. Surface characterizations summarize that, due to the difference in mineral composition, the affinity of minerals to oil components in aqueous solutions is given as: calcium carbonate (positively charged calcium ions) > kaolinite (containing –AlOH, –SiOH groups and heavy metal elements) > silica (–SiOH group). The above results reveal the differences among bitumen subfractions and mineral types, allowing potential insights to the development of enhanced oil recovery, such as aqueous-nonaqueous hybrid extraction process, solvent extraction, CO₂-enhanced oil recovery, etc.

1. Introduction

Separation of heavy hydrocarbons from mineral surfaces is the key step for unconventional oil production as well as the remediation of oil-contaminated soils (e.g., oil sludge). Here, take the unconventional oil production as an example, the unconventional oils (including heavy oil, oil/tar sands bitumen, shale oils, asphalt rocks, etc.) are considered as important alternative fuels to traditional crude oil [1,2]. However, differing from the traditional crude oil, the unconventional oils are more complex with higher content of heavy components, higher molecular weight, higher polarity, higher viscosity and more heteroatoms. Besides, these unconventional oils are found to coexist with different kinds of mineral solids, such as quartz sands, carbonate rocks, clays (e.g., kaolinite), etc. [1]. These natural properties lead to much more difficulty in the exploitation of unconventional oils, which is also true during the remediation of oil-contaminated soils.

The most widely used method to recover the unconventional oils in industry, such as bitumen, is water-based extraction processes. In this process, the separation of heavy oil from their host rocks is considered as the rate controlling step. The separation process is highly influenced by the water chemistry, oil composition and mineral composition, etc. [1,3,4]. During the past few decades, many investigations have been conducted to understand and enhance the oil separation from minerals by: controlling water chemistry [5–7], adding external chemicals [8–11], changing temperature, modifying solid wettability [11], etc.

It is well known that the oil properties exert significant influence to the oil-solid separation. For examples, some model studies show that the light petroleum components (e.g., PAHs, light hydrocarbons of kerosene, gasoline) could be relatively easily removed from the solids, while the heavy hydrocarbons (e.g., asphaltenes) tend to stay attached on the solid surfaces [12]. To enhance the oil-solid separation, some hydrocarbon solvents or light maltenes are added into the

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Nomenclature		CMC	critical micelle concentration
Abbreviations		Symbols	
QCM-D	Quartz Crystal Microbalance with Dissipation	Δm	shift of the adsorbed mass
SARA	saturates, aromatics, resins, asphaltenes	Δf	shift of the harmonic frequency
PAH	polycyclic aromatic hydrocarbon	ΔD	shift of the energy dissipation
SDS	sodium dodecyl sulfonate	γ	surface energy component
PS	polystyrene	θ	contact angle
TAN	total acid number	M	desorbed mass
TBN	total base number	k	desorption rate

unconventional oil ores, allowing the reduction of oil viscosity and the oil-water interfacial tension [8,11,13,14]. Generally, most of the previous studies have been focused on the adsorption and desorption behaviors of asphaltenes [12,15,16]. With regards to the effect of oil composition on its recovery, only rough comparison between light and heavy petroleum, as well as the dilution effect of solvents have been concerned. However, little knowledge has been obtained on how bitumen subfractions work in the process of oil-solid separation. From the aspect of oil composition, petroleum oil is generally divided into SARA fractions including saturates, aromatics, resins and asphaltenes. The differences among them are in the H/C ratio, molecular weight, element composition, solubility, polarity, etc. [17–21]. The oil heterogeneities exert significant influence on its liberation from mineral surfaces. The uneven distribution of these subfractions could probably affect the position and required condition of forming pin-holes which expand, leading to bitumen recession to form distinct bitumen droplets on the sand grains [3]. Accordingly, understanding the desorption properties of the SARA fractions from mineral surfaces is of much significance to provide profound implications for the oil recovery industry.

Another essential factor affecting the separation efficiency is the mineral surface [22]. There are tens of different minerals existing in unconventional oil ores and soils, including the silica, silicate minerals, carbonate minerals, and some other metal oxides, etc. [1]. These minerals differ in physicochemical properties, influencing the oil adsorption and desorption [23]. Although no global trend of oil adsorption has been obtained among varying sorbents, the adsorption capacity and kinetics in non-aqueous solution are found to be sensitive to the mineral types [12]. The adsorption of oil on host rock is controlled by the interactions between them, which is also considered as the governing factor of oil displacement [23,24]. It is established that the oil can be more easily desorbed from a water-wetted surface than an oil-wetted surface [11,25]. However, there is still lack of systematic quantitative comparison of water-based desorption of different oil fractions from the silica, kaolinite and calcium carbonate surfaces.

To address the above issues, the quantitative desorption of different oil fractions on different host mineral surfaces has been performed in this study. Specific objectives were to: i) systematically investigate the desorption behaviors of SARA fractions and bitumen from mineral surfaces; ii) understand the effect of solid type on the desorption behaviors of different oil components; iii) understand the difference of oil and mineral composition in the oil-solid separation and potential suggestions for the enhancement of oil separation from real mineral surfaces.

Table 1
Composition of silica, kaolinite and calcium carbonate.

Element (wt%)	SiO ₂	Al ₂ O ₃	CaO	Fe ₂ O ₃	K ₂ O	MgO	ZrO ₂	TiO ₂	P ₂ O ₅	Min ^b
Silica	98.04	0.63	0.28	– ^a	–	–	–	–	1.00	0.05
Kaolinite	52.77	39.90	0.75	2.10	0.42	–	0.40	1.91	1.62	0.13
Calcium carbonate	–	–	99.02	–	–	0.23	–	–	0.55	0.20

^a – And Min are referred to the elements less than 0.1% of the minerals by weight.

2. Experimental section

2.1. Chemicals and oil fractions

All chemicals (at analytical-grade) were provided by Tianjin Jiangtian Technology Co., Ltd., China. Toluene, n-heptane, methanol and trichloroethylene were used as solvents. Three different kinds of mineral nanoparticles, silica, kaolinite and calcium carbonate, were used in preparation of the three mineral surfaces. The mineral composition of the solids was analyzed using X-ray fluorescence (XRF, Bruker S4 Pioneer, Germany), as shown in Table 1. All of the elements are listed in the form of oxides.

The solutions of sodium hydroxide, sodium dodecyl sulfonate (SDS) were used to facilitate the oil desorption. Potassium hydroxide, glycerol and diiodomethane were used in the acid number measurement and surface energy analysis, respectively. The water used in the whole experiment was at an ultrapure level (18 M Ω).

The bitumen sample was extracted from Athabasca oil sands (containing 12.88 wt% bitumen) by toluene using the standard Dean Stark method [26]. The reference method of SARA fractionation is the ASTM D-4124 [27]. The mass fraction of each fraction in bitumen is shown in Table 2.

2.2. Substrates preparation

2.2.1. Preparation of mineral basal planes

The solid suspensions in ethanol (~0.4 wt%) were prepared using high-purity silica, kaolinite and calcium carbonate nanoparticles (< 500 nm) respectively. The procedure to obtain the suspensions is given as follows: firstly, high concentration of suspensions were prepared with smashed mineral particles. Secondly, the suspensions were stirred and sonicated to obtain homogenous dispersion. Then, the suspensions were centrifuged at 640g for 3 min, followed by the decanting to extract the suspended upper phase. These procedures of stirring, sonication, centrifugation (1000g for over 3 min), and decanting, were repeated at least twice [28].

To prepare basal surfaces consisting of silica and kaolinite particles, the gold wafer was initially placed on a spin coater (KW-4A, Institute of Microelectronics, CAS). A total of 1 mL suspension was pipetted as 25 drops at the stirring rate of 2000 rpm within 20 s, followed by a speed of 5000 rpm for 60 s to obtain a relatively smooth and uniform silica/kaolinite deposited surface. The same procedure was used to spin-coat oil components on the substrates in other experiments. After being

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