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Dynamic adsorption of water soluble crude oil components at air bubbles

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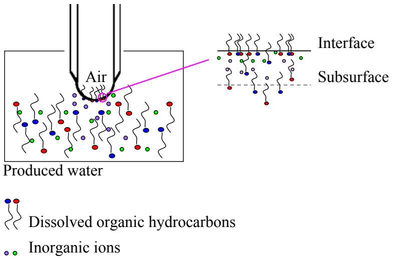
HIGHLIGHTS

- Nine different crude oils were used for synthetic produced water (PW) preparation.
- Adsorption of dissolved crude oil hydrocarbons onto air bubbles is investigated.
- The adsorption barrier appeared faster for the PW samples with higher TOC values.
- Crude oils acids and bases seemed to be partially present in PW samples.
- Water solubility and surface affinity is related to molecular structure of hydrocarbons.

GRAPHICAL ABSTRACT



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ABSTRACT

A detailed analysis of the dynamic adsorption of crude oil components dissolved in produced water onto air bubble surfaces is presented. Nine different crude oils were used to prepare synthetic produced water samples. The total organic carbon content, molecular mass and dynamic surface tension was determined for the samples. The experimental data were applied in a modified version of the Ward–Tordai model, in order to evaluate the adsorption mechanism. This allowed for determination of characteristic diffusion coefficients and investigation of adsorption mechanisms. All the crude oils contained water soluble components within a relatively narrow range of molecular size, but different polarity. The adsorption mechanism was purely diffusion controlled at short time scales regardless of the chemical composition of the crude oil. Further analyses were performed on two of the crude oils by selectively extracting interfacially active components (acids and bases) from the crude oils. Both acidic and basic compounds were present in the produced water samples, but their affinity for the air/water interface varied. There were no correlation between the amount of acids and bases in the crude oils and the solubility and surface activity of the respective water soluble fractions.

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1. Introduction

Produced water is a by-product of petroleum processing and can be a mixture of formation water (i.e., water naturally present in reservoirs), water previously injected into the formation and condensed water (Faksness et al., 2004; Wang et al., 2012). Its physical and chemical properties are strongly field dependant and will also vary within the field life time. Generally, the composition of produced water can be identified as dispersed oil and solids, dissolved organic and inorganic compounds, chemical production additives and traces of heavy metals (Silset et al., 2010). Some installations at the Norwegian Continental Shelf re-inject produced water into the reservoir for pressure support, but in most cases it is discharged to sea. Removal of dispersed oil as well as

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soluble and semi-soluble hydrocarbons is specifically targeted in order to minimize the environmental impact in the latter case (OSPAR, 2010).

Gas flotation is a widely used process for removing dispersed oil from produced water streams. The principle of induced gas flotation is attachment of oil droplets onto gas bubbles (Grattoni et al., 2003; Moosai and Dawe, 2003). This involves film rupture, followed by spreading of oil droplets around the gas bubbles. The properties of both the oil–water and water–gas interfaces will have an effect on each of these steps. Consequently, it is anticipated that fundamental understanding of the interfacial phenomena is necessary to facilitate optimal operating conditions during flotation processes.

Several studies have focused on detailed characterization of organic components dissolved in produced water, and both polar and non-polar compounds have been identified (Grewer et al., 2010; Headley and McMartin, 2004; Røe Utvik, 1999; Stanford et al., 2007). The partitioning of semi-soluble organic compounds between the water phase and oil droplets in produced water has been studied using conventional methods such as GC–MS and GC–FID (Faksness et al., 2004; Røe Utvik, 1999). In these studies, the produced water samples from different oil fields in North Sea were characterized with respect to benzene, toluene, ethylbenzene and xylene (BTEX), Poly aromatic hydrocarbons, phenols and their alkylated fractions. Research on polar water soluble compounds preliminary focused only on naphthenic acids. A review regarding the origin of these acids, their chemistry and available analytical methods for aquatic sampling has been done by Headley and McMartin (2004). However, naphthenic acids are not the only polar water soluble compounds which can be found in produced water and, N–S–O containing compounds can exhibit considerable water solubility. Stanford et al. (2007) identified water soluble N–S–O containing acidic, basic and neutral crude oil hydrocarbons from North and South American and Eastern crude oils in both pure water and sea water. They used ESI–FT-ICR MS technique for their analysis. Using the same technique, several studies have recently been performed on characterization of polar organics, naphthenic acids and acid extracts from different water samples, mostly from oil sand ponds and tailings (Grewer et al., 2010; Headley et al., 2011, 2012).

Generally, polar compounds are considerably more soluble than non-polar hydrocarbons in saline aqueous solutions, and the solubility will depend on molecular weight and type of heteroatoms present in the molecules (Headley et al., 2011, 2012; Stanford et al., 2007). In spite of all the effort which has been done on identifying the water soluble crude oil hydrocarbons, their adsorption behavior at air–water interfaces has received very little attention (Valsaraj, 1994).

Previously, we have reported on how the ionic composition of the water influences the adsorption of water soluble crude oil components onto air–water interfaces (Eftekhardadkhah and Øye, submitted for publication). In this paper the focus is on the dynamic adsorption of water soluble components in synthetic produced water prepared from nine crude oils. Furthermore, acidic and basic components were selectively extracted from two of the crude oils and synthetic produced water was prepared from these fractions as well. The aim was to obtain better understanding of the type of crude oil components that partition into the water phase and, in particular, how they influence properties of the air–water interfaces. The amount and molecular weight distribution of the dissolved hydrocarbons in the samples were determined by Total Organic Carbon (TOC) analysis and Electro-spray Ionization Mass Spectroscopy (EIMS), respectively. The dynamic surface tension was measured by maximum bubble pressure tensiometer, and a modified version of Ward–Tordai model was applied for detailed evaluation of adsorption mechanisms.

2. Materials and methods

2.1. Preparation of brine

Simulated brine was prepared by dissolving the analytical grade of NaCl (99.5%, Merck, Germany), Na_2SO_4 (99%, Acros, USA), $NaHCO_3$ (99.5%, Merck, Germany), $MgCl_2{\bullet}6H_2O$ (99%, Merck, Germany) and $CaCl_2{\bullet}2H_2O$ (99.5%, Fluka, Czech Republic) in water provided by a Millipore ultrapure water system. The ionic composition of the brine is listed in Table 1. In order to prevent precipitation of carbonate, the brine was stored in a refrigerator and brought to the experimental temperature just before use.

2.2. Preparation of synthetic produced water

The water soluble hydrocarbons of the crude oils were partitioned into the aqueous phase by mixing 50% oil and 50% simulated brine with a shaker (HS 501 digital IKA). The mixing speed was 250 rpm and the mixing was maintained for 24 h at room temperature. Subsequently, the oil and water phases were separated by centrifugation. The resulting aqueous phase was denoted synthetic produced water samples, and were prepared from nine crude oils. The Physicochemical properties of the crude oils are listed in Table 2, and the crude oils were denoted A–I. More detailed information about the oils can be found elsewhere (Gawel et al., in preparation).

Here, the synthetic produced water samples were named PW-A to PW-I, in accordance with the naming of the crude oils. Three parallel samples were prepared in each case.

2.3. Acid and base extracted fractions

Selective extractions of acidic and basic components were carried out for two crude oils according to methods developed by Simon et al., and detailed descriptions of the procedures can be found elsewhere (Nenningsland et al., 2010; Simon et al., 2008). Samples of synthetic produced water were prepared from these

Table 1
Average concentration of brine components.

Ions	Concentration (ppm)
Cl^-	62,810
Na^+	35,393
Ca^{2+}	3253
Mg^{2+}	909
$HCO_3{}^-$	218
$SO_4{}^{2-}$	49

Table 2
Crude oils physicochemical characteristics.

Crude oils	Density (g/cm^3)	Viscosity@ 50 °C (cP)	*SA* (%)	*AR* (%)	*R* (%)	*AS* (%)	TAN[a]	TBN[a]
A	0.80	1.4	80.0	18.0	1.9	0.1	0.3	0.5
B	0.91	4.6	44.3	38.8	16.3	0.4	2.2	4.4
C	0.94	17.8	25.6	49.6	10.9	13.9	0.50	1.3
D	0.91	73.4	58.1	30.7	9.5	1.7	0.3	1.5
E	0.81	4.5	67.6	27.1	4.4	0.7	0.2	1.1
F	0.82	4.6	62.1	31.2	5.8	0.6	0.2	1.4
G	0.92	59.6	53.5	29.9	14.4	2.5	2.1	2.8
H	0.88	11.1	60.0	36.0	6.7	0.3	2.6	0.7
I	0.89	32.7	52.0	31.9	13.8	2.1	1.4	3.0

[a] TAN and TBN are given in (mg of KOH/g of oil).

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