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## Influence of the wettability of particles on the morphology and stability of crude oil–particle aggregates in synthetic produced water

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## ABSTRACT

Handling and treatment of produced water is becoming an increasing part of oil and gas production. A major challenge in this respect is removal of oil drops stabilised by particles. Understanding the influence of interfacial and wetting properties of crude oil–particle+ systems on the formation of particle stabilised emulsions is therefore essential. Elucidating these relationships was the aim of this investigation. Synthetic produced water containing different particles and crude oil were studied. The morphology of aggregates formed by crude oil drops and particles was strongly related to the contact angle ( $\theta_{wo}$ ) at the water–oil–particle interfaces. High  $\theta_{wo}$  resulted in formation of particle stabilised emulsions, while less defined aggregates were formed at lower  $\theta_{wo}$ . Furthermore, coalescence of drops was related to the coverage of particles at the interface. Stable drops covered by particles were only seen when the amount of particles was significant. Otherwise drops tended to coalesce, likely due to particle bridging.

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## 1. Introduction

Vast amounts of water are produced during oil and gas recovery (Lee et al., 2011). The produced water contains a complex mixture of dissolved and dispersed components. The latter includes crude oil drops as well as various inorganic particles originating from the reservoir or appearing as scale and corrosion products. Removal of dispersed oil is considered to be crucial in order to reduce the environmental impact of produced water. Therefore current regulations in the North Sea require that the amount of oil in water must be less than 30 ppm prior to discharge to sea (OSPAR Commission, 2010).

Pressure drops and shear forces during transport from the reservoirs to production facilities can lead to extensive mixing of oil drops and particles. The properties of both the oil phase and water phase can alter the surface properties, wettability and stability of the particles (Dudášová et al., 2014, 2009a, 2009b, 2008; Marczewski and Szymula, 2002; Sullivan and Kilpatrick, 2002). Furthermore, if particles become trapped at oil–water interfaces, particle stabilised emulsions can form. Oil drops stabilised by particles can be very difficult to remove by typical water treatment processes such as hydrocyclones and compact flotation units due to hindered coalescence and reduced Stokes velocity (Finborud, 2010; Markoff, 2009; Yan and Masliyah, 1997).

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Effective coalescence of oil drops is beneficial for ensuring good quality of the produced water. It is well known that indigenous interfacially active components in the crude oil, like asphaltene class, can cause formation of mechanically strong, viscoelastic films at the interface (Horváth-Szabó et al., 2005; Pawar et al., 2011; Poteau et al., 2005; Spiecker et al., 2003). High elasticity of such films can slow down coalescence of oil drops (Gawęł et al., 2014b). Furthermore, emulsions containing inorganic particles can be even more stable than those stabilized by asphaltenes only (Hannisdal et al., 2006; Nianxi and Masliyah, 1996; Sullivan and Kilpatrick, 2002; Yan and Masliyah, 1994, 1997; Yan et al., 1999).

The ability of particles to stabilise emulsions is attributed to the large free energy of adsorption of solids which are partially wetted by both the oil and water phases (Aveyard et al., 2003; Binks, 2002). This means that particle layers adsorbed at drop interfaces can result in extremely stable emulsions (Binks and Lumsdon, 1999; Binks and Whitby, 2004; Dong et al., 2014). The wettability is a key factor affecting the properties of the particle layers and subsequently the emulsion behaviour (Gu et al., 2003; Yan et al., 2001). The contact angle for oil–water systems is related to interfacial tensions ( $\gamma$ ) according to the Young equation:

$$\gamma_{so} = \gamma_{sw} + \gamma_{ow} \cos \theta_{ow} \quad (1)$$

where the subscripts s, o, and w represent solid, oil and water, respectively. The oil–water contact angle ( $\theta_{ow}$ ) is measured through the water phase. The  $\gamma_{ow}$  is related to the chemical composition of the water phase and crude oil, while  $\gamma_{so}$  is related

to the composition of the crude oil and the adsorption of components at the solid surface (Abdallah et al., 2007).

The free energy of adsorption of particles is dependent on the contact angle by the following expression:

$$\Delta G_d = \pi r^2 (1 - |\cos(\theta)|)^2 \quad (2)$$

where  $r$  is the diameter of the particles. This expression states that adsorption of particles is strongest when  $\theta_{ow}$  is 90°. It is clear from this that the wettability of particles is a very important factor for emulsions stability. Another central parameter for particle stabilization emulsions is the surface coverage of the particles (Leal-Calderon and Schmitt, 2008; Pawar et al., 2011). However, there are extensive variations in the literature with respect to how many particles are required at the interface in order to constrain coalescence (Binks and Lumsdon, 1999; Dong et al., 2014; Frelichowska et al., 2010; Pawar et al., 2011; Tambe and Sharma, 1993; Vignati et al., 2003). Many studies related to particle stabilisation of emulsions have been focused on relatively simple model systems. During crude oil production, however, the fluid stream consists of crude oil, water and particles which become mixed during transport and can form various types of agglomerates like particles stabilised emulsions, schmoos etc. In this work the produced water stream was mimicked by mixing emulsions and particles under mild conditions. The main objective was to investigate the relations between wetting properties of particles in crude oil – synthetic produced water systems and the stability of the corresponding aggregates. Five different types of particles, typically present in produced water streams were used. The wetting properties of the particles, morphology of particle–oil aggregates and the corresponding stability in synthetic produced water were followed by contact angle, microscopy and Turbiscan measurements.

## 2. Materials and methods

### 2.1. Crude oil

The crude oil used in this study has previously been characterized with respect to its physicochemical and interfacial properties. Details about the results and experimental procedures can be found elsewhere (Gawel et al., 2014a, 2014b). Some of the properties are presented in Table 1 for convenience.

### 2.2. Particles

The inorganic particles were chosen to represent minerals typically present in produced water (i.e. clay, scale and corrosion products): kaolin (Aldrich, USA/Germany); CaCO<sub>3</sub> (98.2%) (Specialty Minerals Inc., USA); FeS (99.7%) (DLFTZ, Chang Hing, China); BaSO<sub>4</sub> (99%) and Fe<sub>3</sub>O<sub>4</sub> (98+%) (Nanoamor, USA). Physical properties of the particles are listed in Table 2. The particle shape,

**Table 1**  
Physicochemical properties of the crude oil (Gawel et al., 2014a).

Property	Value
Density at 20 °C (g/cm <sup>3</sup> )	0.81
Viscosity at 20 °C (cP)	3
Saturates (wt%)	80.0
Aromatics (wt%)	18.0
Resins (wt%)	1.9
Asphaltenes (wt%)	0.1
TAN (mg/g)	0.4
TBN (mg/g)	0.6

**Table 2**  
Physical properties of the particles.

Particle	Shape	Specific surface area (BET) (m <sup>2</sup> /g)	Density (g/cm <sup>3</sup> )	Particle size (from suppliers) (μm)	Average aggregate size (μm)*
Kaolin	Plate like	19.7 ± 3	2.65	0.1–4 μm	2.0
CaCO <sub>3</sub>	Rhombic	19.9 ± 2	2.71	70 nm	1.3
BaSO <sub>4</sub>	Prismatic	6.5 ± 1	4.5	80–450 nm	1.1
Fe <sub>3</sub> O <sub>4</sub>	Spherical	45.2 ± 3	4.8–5.1	20–30 nm	1.7
FeS	Spherical	6.9 ± 1	4.6–4.8	2 μm	2.1

\* Measured by Coulter Counter in 3.5 NaCl solutions.

densities and particle size/size distributions were provided by the suppliers, while surface area and average aggregate sizes were measured previously (Dudášová et al., 2009a, 2009b).

### 2.3. Brine composition

The brine solution was made by dissolving appropriate amounts of NaCl (99.5%, Merck, Germany), CaCl<sub>2</sub> · 2H<sub>2</sub>O (99.5%, Merck, Germany), MgCl<sub>2</sub> · 6H<sub>2</sub>O (99.5%, Merck, Germany), NaHCO<sub>3</sub> (99.5%, Merck, Germany) and Na<sub>2</sub>SO<sub>4</sub> (98.5%, Acros Chemicals) in MQ water from a Millipore Simplicity System. The ionic composition is listed in Table 3

### 2.4. Contact angles measurements

The particles were pressed into flat pellets (13 mm in diameter) under high pressure using a hydraulic press (Compac, Denmark). The pellets were put in a custom made holder and immersed in the brine solution before an oil drop was placed on the pellet surface by a syringe. Images were taken of the oil drop immediately after attachment using an Optical Contact Angle Metre equipped with a computer-controlled high-speed camera (CAM200, KSV Instruments). Subsequently, the contact angles were determined by fitting the Young Laplace equation to the drop profiles.

### 2.5. Microscopy imaging

Crude oil was weighted into 50 ml Schott bottles and the brine was added to give oil concentration of 10,000 ppm. The samples were emulsified with an Ultra Turrax mixer (IKA, S25N-10G with 10 and 7.5 mm stator and rotor diameter respectively) at 20,000 rpm for 2 min. Subsequently, particles were added to the o/w emulsion to give a concentration of 5000 ppm. The system was dispersed using an automatic shaker (HS 501 digital IKA) at 300 rpm for 1 h. The shaking was used to mimic mixing under mild shearing conditions.

Images of the dispersions were taken by a Nikon LV 100D microscope. To avoid any influence of glass slides on the oil drops, the emulsions were poured into rectangular glass capillaries

**Table 3**  
Ionic composition of the brine solution.

Ions	Concentration (ppm)
Cl <sup>-</sup>	62,810
Na <sup>+</sup>	35,393
Ca <sup>2+</sup>	3253
Mg <sup>2+</sup>	909
HCO <sub>3</sub> <sup>-</sup>	218
SO <sub>4</sub> <sup>2-</sup>	49

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