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Correlations between emulsification behaviors of crude oil-water systems and crude oil compositions

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ABSTRACT

The emulsification behavior of a crude oil-water system is greatly related to the compositions of a crude oil. However, there was lack of quantitative description on the relationship between emulsification behaviors and crude oil compositions. As the first part of this work which was published recently, full and partial emulsification were found which were dependent upon shear rate and water fraction; For the partial emulsification under high water fraction, the relationship between the emulsified water fraction under flowing conditions and entropy production rate was found to obey a power law equation; Moreover, the critical value of water fraction, the range of shear rate for full emulsification, and the coefficient and power law index were highly dependent on the compositions of crude oil. As a continued study, this paper further investigates quantitatively the relationship between the above-mentioned characteristic parameters of emulsification behaviors and the crude oil compositions. In terms of the mechanism on the emulsification behaviors, the crude oil compositions are classified into four categories, i.e. the content of asphaltenes and resins (c_{a+r}), the sum of the concentration of precipitated wax and the content of mechanical impurities (c_{w+m}), acid value (AV), and the average carbon number of crude oil (CN_{oil}). By data regression based on emulsification experiments for eight crude oils, correlations were developed between these emulsification parameters and the oil compositions, helping to understand quantitatively the relationship between emulsification behaviors and crude oil compositions. In addition, the influence of the four compositions was found to be ranked as $CN_{oil} > c_{a+r} > AV > c_{w+m}$.

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1. Introduction

The formation of emulsions is common during the production and transportation process of crude oil, which can greatly affect the flow characteristics (Grassi et al., 2008; Hwang and Pal, 1998; Hoshyargar and Ashrafizadeh, 2013; Plasencia et al., 2013; Sotgia et al., 2008; Wang et al., 2011) of multiphase flow. Recently, we published our first part of the work on the emulsification behaviors of crude oil-water systems (Wen et al., 2014), which reported a phenomenon of the full emulsification below a critical value of water fraction, and a phenomenon of partial emulsification at high water fractions. We also found that for high water fraction systems in which partial emulsification can only occur, the emulsified water fraction under flowing conditions correlates well with entropy production rate, and this correlation obeys a power law equation. Quantitatively, these emulsification behaviors varied from one crude oil to another. This stimulates us to further investigate the quantitative relations between

the characteristic parameters of emulsification and the crude oil compositions. This paper reports the results of our continued study.

The natural surfactants in the crude oil, such as asphaltenes, resins, and acidic compounds etc., reducing the interfacial tension of oil-water and forming an interfacial film with certain mechanical strength, plays a key role in the emulsion formation and stabilization. Therefore, the difference in crude oil compositions may result in large difference in the emulsification behaviors.

Asphaltene is a mixture comprising of numerous non-hydrocarbon compounds with complicated molecular structures. It improves the stability of emulsion due to its good interfacial activity (Acevedo et al., 1999; Kilpatrick, 2012). The molecular aggregation and concentration of asphaltenes in crude oil significantly affect the stability of emulsion. Jestin et al. (2007), Verruto and Kilpatrick (2008), Alvarez et al. (2009), and Liu et al. (2006) stated that the stabilizing interfacial film in asphaltenic emulsions was formed by asphaltenic aggregates with a size of 7–20 nm. Castillo et al. (2004) and Stark and Asomaning (2005) indicated that the stability of the emulsion was greatly affected by the aggregation of asphaltenes, i.e., that the asphaltenes dispersing in the crude oil in a state of aggregation led to a greater emulsification ability. Rondón et al.

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(2007) revealed that the concentration of asphaltenes played an important role in the emulsion stabilization, however, a stable emulsion was unable to be formed with the concentration of asphaltenes lower than a critical value. McLean and Kilpatrick (1997), Sullivan et al. (2007) also found that a higher concentration of asphaltenes but lower than the critical concentration of precipitation led to a more stable emulsion; while unstable emulsion occurred for the concentration of asphaltenes higher than the critical concentration at which asphaltenes began to precipitate.

Hydrocarbons with different carbon numbers have different effects on the aggregation of asphaltenes. Hammami et al. (2000) indicated that the addition of alkanes such as n-heptane into the oil containing asphaltenes would reduce the solubility of the asphaltenes and improve the aggregation of the asphaltene molecules. Hirschberg et al. (1984) used the n-alkanes (from n-heptane to n-hexadecane) to precipitate the asphaltenes in oil. It was found that with the increase of carbon number of the n-alkanes, the solubility of the asphaltenes decreased, and the precipitating rate of the asphaltenes increased. For different crude oils, the hydrocarbon components and the distribution of carbon number vary, and hence, the solubility and the aggregation of the asphaltenes are different, leading to various emulsification characteristics.

Similar to asphaltene, resin is also a mixture comprising of a number of macromolecular non-hydrocarbon compounds, but with relative low molecular weight and polarity. There is no clear boundary between resin and asphaltene, and they are usually classified according to their solubility in a certain solvent such as n-heptane. Generally, the strength of the interfacial film formed by resins is lower than by asphaltenes and hence the stability of the emulsion (Mohammed et al., 1993; Alvarez et al., 2008). Yang et al. (2007) indicated that a stable emulsion could be formed if only asphaltenes presented, and with the addition of resins the stability of the emulsion deteriorated. Al-Sahhaf et al. (2008) and Nour et al. (2008) also found that an increase of the resin/asphaltene ratio in the simulated oil could lead to a decrease in the stability of emulsion.

The adsorption of wax particles in the crude oil onto the oil-water interface is able to enhance the strength of the film, leading to a more stable emulsion. Bobra (1991) discovered that a stable emulsion could not be formed with the simulated oil containing only asphaltene or paraffin but with the simulated oil containing both asphaltene and paraffin, indicating that the wax particles put a significant impact on the emulsion stabilization. It is also indicated in the study by Haj-shafiei et al. (2013) that the wax particles with appropriate size enhanced the stability of the W/O emulsion.

Mechanical impurities in crude oil such as solid particles, just like wax particles, can also adsorb onto the oil-water interface and hence help to the stabilization of emulsion (Gelot et al., 1984; Li et al., 2009). Moreover, the steric stabilization given by the solid particles may prevent the movement and coalescence of the emulsion droplets and as such increase the stability of the emulsion (Tambe and Sharma, 1993).

The adsorption of acids in the crude oil onto oil-water interface dramatically lowers the interfacial tension and hence stabilizes interfaces and emulsions (Arla et al., 2007; Alvarado et al. 2011). Guo et al. (2006) found that the adsorption of acids and their soaps onto oil-water interface stabilized the O/W emulsions. The interfacial activity is not proportional to the acid acidity, but is related to the property of the acid itself. Ovalles et al. (1998) found that the interfacial activity of acid was highly dependent on its structure and properties. Li et al. (2006) studied the effect of acidic compounds in crude oils on the emulsion with different crude oils, and found that the acids with lower molecular weight, such as the carboxylic acid and aliphatic acid reduced the interfacial tension of oil-water, while the acids with higher molecular weight stabilized

the emulsion. Kilpatrick (2012) indicated that the capability of carboxylic acids to lower interfacial tension and stabilize emulsion varied significantly with chemical structures and molecular weights. Naphthenic acids, which own at least one cyclic alkane functional group, also contribute to the stabilization of emulsion (Shepherd et al., 2010). Horváth-Szabó et al. (2003) indicated that sodium naphthenates can lead to form the lamellar liquid crystalline film, which stabilize W/O emulsion. Ese and Kilpatrick (2004) found that a naphthenic acid called β -cholanic acid may stabilize not only W/O emulsion but also O/W emulsion, which depends on the concentration of the acids.

Although a large body of research has been conducted to examine the effect of crude oil compositions on the emulsification of crude oil-water systems, studies on quantitative relationship between emulsification behaviors and crude oil compositions are generally scarce.

In this further work, we examined the factors of crude oil composition that may affect the emulsification of crude oil-water system, and determined the representative compositions and then took the values of these compositions by analytical chemistry methods. Finally we developed correlations of emulsification parameters and crude oil compositions.

2. Review of our previous work and additional results of continued study

This section briefly reviews our previous work (Wen et al., 2014) so that provides indispensable information for further study of this continued work. And of course, additional experimental results in the present work are presented.

2.1. Experimental materials

The crude oil specimens used in the present work are the same as our previous work. In order to facilitate the understanding of this work, some necessary properties showed in the previous work and some new properties of the oils are showed in Table 1.

The added properties, including the acid value, mechanical impurities of the crude oil, concentration of precipitated wax and wax appearance temperature (WAT), carbon number distributions of the crude oil, and the average carbon number of alkanes (C_8 to C_{40}) were determined by following approaches.

The acid values of crude oils were determined according to ASTM D664-11a. The contents of mechanical impurities of crude oils were determined according to ASTM D473-07. The concentration of precipitated wax and WAT were determined according to ASTM D4419-90. The carbon number distributions (C_1 to C_{40}) of the crude oils were determined using the gas chromatograph Agilent 6890, and the average carbon number of alkanes (C_8 to C_{40}) was calculated as follows (Yi and Zhang, 2011).

$$CN_{oil} = \frac{1}{14} \left[\sum_{n=8}^{40} \frac{(14n+2)w_n}{100} - 2 \right] \quad (1)$$

where w_n is the mass fraction of the alkanes (C_8 to C_{40}) (wt%).

In the present work, the emulsification experiments were conducted at two preset temperatures, i.e., (1) a temperature of 5 °C below the WAT of each oil, (2) and a temperature above the WAT of each oil. The concentration of precipitated wax at experimental temperature below WAT was measured correspondingly, as shown in Table 2.

2.2. Emulsification experiment results

The emulsified water fraction under flowing conditions was

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