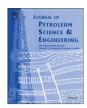
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Effects of concentration, salinity and injection scenario of ionic liquids analogue in heavy oil recovery enhancement



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ABSTRACT

Recently, Deep Eutectic Solvents (DESs) have been demonstrated to be an effective analogue of ionic liquid in many applications. In our previous report, for the first time, two Chloride-based DESs were shown to have significant effects on different oil recovery mechanisms in heavy oil/formation brine/Berea sandstone system. In this report, the same DESs were used. The effects of DESs concentration, secondary and tertiary DES injections and brine salinity on recovery performance were examined through IFT measurements, contact angle measurements and core flooding experiments.

Results of capillary number calculations indicated that although DESs solutions increased the IFT value, the capillary numbers in DESs flooding due to viscous forces and wettability alteration were higher than brine flooding case. Results showed that for both DESs, although the heavy oil recovery factors decreased by diluting the DES solution with brine from 50 vol% to 5 vol%, they were still promising between 6.3 and 8.7% for DES1 and DES2 respectively at lower concentrations of 5 vol%. Also, by reducing concentration from 50 to 25 vol%, there were no changes in recovery factors from 14 to 13.6% for DES1 and from 23.2 to 23.1% for DES2. The same trend was observed during wettability alteration study by contact angle measurements between oil droplet and aged surface of rock samples in the both DESs solutions at different concentrations. It verified that wettability alteration during DES injection has significant impact on oil recovery and it could be dominant and governing mechanism of heavy oil recovery enhancement. The DESs flooding in tertiary stage led to higher total recovery factors. The total recovery at tertiary mode compared to secondary mode were around 16% higher at 50 vol% concentrations and 3–6% higher at 5 vol% concentrations for both DESs. In addition, Results confirmed that the DESs can tolerate wide range of reservoir's salinities and there were no big changes in oil recovery at different brine salinity during tertiary DESs injection.

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1. Introduction

Previous reports have shown that more than two third of original oil in place will remain unproduced in the reservoirs after primary stage of conventional production (Saidi, 1996; Al-Hadhdarmi and Blunt, 2001; Kianinejad et al., 2013). Also, ever growing energy demands in the worlds motivates oil producing companies to find efficient enhance oil recovery (EOR) methods to recover the trapped oil from conventional and unconventional resources such as heavy oil reservoirs. Chemical flooding as an efficient EOR technique has attracted attentions to the wide range of investigations in past 30 years. Significant achievements in application of surfactants, alkaline and polymers have been made in term of recovery enhancement due to different mechanisms such as lowering interfacial tension (IFT) between trapped oil and displacing chemical, emulsification, improving sweep efficiency

and wettability alteration of reservoir's rock (Sheng, 2013). Chemical methods have been used not only for light oil recovery but also for heavy oil recovery enhancement, and several studies have reported successful field scale experience in utilizing chemicals for improving heavy oil recovery (Pei et al., 2012; Delamaide et al., 2014).

More recently, new group of chemicals called Ionic Liquids (ILs) have received interests as green alternative to conventional chemicals for EOR purposes (Abbott et al., 2007; Shahbaz et al., 2012; Pereira et al., 2014). Several investigations in the literature have reported concerns about surfactant adsorption on the rock surface which leads to chemical lost and formation damage in the reservoirs during chemical flooding (Dang et al., 2011; Somasundaran and Zhang, 2006). It is expected that ionic liquids, in form of diluted salt solution, could be injected through the reservoir without causing any mechanical plugging in the porous medium. Zeinolabedini et al. (2013) studied the use of 1-dodecyl-3-methylimidazolium chloride as an IL-based surfactant to enhance oil recovery. They showed promising results in terms of IFT reduction and tertiary oil recovery efficiency. Unlike the conventional surfactants, they also reported that the IL is

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more effective in high saline formation brine even at very low concentration of 100 ppm and it has minor adsorption in the rock surface. Pereira et al. (2014) applied ionic liquids for EOR of aromatic oil at laboratory scale using sand pack model. They showed that injecting just 2 wt% aqueous solution of 1-ethyl-3-methylimidazolium tosylate ([C2mim][OTs]) can double recovery factor compared to brine solution. The polar compounds of heavy oil such as asphaltene and resins would diffuse in the ILs which results to reductions in the heavy oil viscosity, asphaltene and sulfur contents. These reductions will lead to an increase in the heavy oil "API gravity as well as a decrease in asphaltene precipitation and subsequent plugging of rock formation. Hu and Guo (2005) simulated CO₂-flooded reservoir treated by ionic liquids and they have shown that using ILs leads to slight asphaltene precipitation and deposition in the reservoir during CO2 injection. Furthermore, the catalytic properties of ILs in crude oil oxidation, cracking and hydrocracking reactions, which are partly responsible for heavy oil upgrading have been reported (Nares et al., 2007).

Deep Eutectic Solvents (DESs) have been determined as a class of ILs which has more advantages than the common ILs. They have shown more "green" behavior such as biodegradability and biocompatibility, chemical compatibility with water, easy preparation process, non-toxicity and very low prices (Abbott et al., 2007). Generally, the DESs are a eutectic mixture at suitable temperatures which can be synthesized by the combination of various quaternary ammonium/ phosphonium salts such as Choline Chloride (2-hydroxyethyl-trimethylammonium chloride, (ChCl)) with different Hydrogen Bond Donors (HBD) such as amines and carboxylic acids. DESs have currently found many applications in the different industries including extraction/separation, catalytic processes, electrochemistry, CO2 absorption and pharmaceutical, solvent development/reaction medium and hydrometallurgy. In addition, DESs have potential for EOR applications due to their excellent physicochemical properties such as high viscosity, polarity and better surface active ability (Mohsenzadeh et al., 2014a, 2014b; Shuwa et al., 2014; Zhang et al., 2012).

In our previous study (Mohsenzadeh et al., 2014a,b), Choline Chloride-based DESs were examined experimentally for the first time to study their effects on different oil recovery mechanisms in heavy oil/formation brine/Berea sandstone rock system. The evaluated mechanisms were emulsification, IFT reduction, wettability alteration and oil recovery factor by core flooding experiments. Although the DESs increased the IFT and could not make emulsion with heavy oil, it was found that wettability alteration of the rock surfaces from strongly oil-wet to neutral-wet condition as well as viscous forces were the main mechanisms leading to 14–30% enhanced heavy oil recovery (depends on DES components and temperature) at tertiary stage after water flooding.

In this study, we report results of further investigation to cover other important parameters affecting the performance of heavy oil recovery by DESs injection. The same DESs including Choline Chloride:Glycerol (1:2 M ratio) and Choline Chloride:Urea (1:2 M

ratio) were used and the effects of DESs concentrations, injection sequences by secondary and tertiary DES injection and brine salinity on heavy oil recovery performance were examined. The IFT measurements by spinning drop tensiometer were conducted to determine the effects of DES's concentration and brine salinity on interfacial tension. Contact angle measurements were performed to study the effects of DESs concentration on rock wettability alteration. Finally, a systematic core flooding experiments were designed and performed to examine the effects of DESs concentration, brine salinity and injection sequence.

2. Experimental

2.1. Material

Heavy crude oil with 15 °API and formation brine from an Omani oilfield were used for the experimental work. The heavy oil and brine properties are shown in Table 1. Homogenized consolidated Berea sandstone core plugs of 1.5″ diameter and 3″ length were utilized for core flooding experiments.

Generally, DESs are categories into 4 types, each consisting of at least binary mixture as follows—(a) type1: metal salts+organic salts, (b) type2: metal salt hydrate+organic salt, (c) type 3: organic salts+ hydrogen bond donor, and (d) type 4: metal salts (hydrate)+ hydrogen bond donor (Alotaibi and Nasr-El-Din, 2009). Among these four groups, types 1, 2 and 4 have very high viscosities and they contain metal compounds which are often very expensive. Therefore, type 3 was selected for this study. This is because it is relatively cheaper and has moderate range of viscosities. In addition, it consists of biodegradable components, which makes it environmentally friendlier than others. Two different Deep Eutectic Solvents (DESs) form type3 namely Choline Chloride/Glycerols (DES1: molar ratio 1:2; molecular weight: 107.93 g/mol) and Choline Chloride/Urea (DES2: molar ratio 1:2; molecular weight: 86.56 g/mol) were synthesized. These selected DESs are much cheaper compared to typical ionic liquids. The cost of typical ionic liquids is about 20,000–30,000 USD/mt whereas for choline chloride the cost is around 300-400 USD/mt, for glycerol is 700-800 USD/mt and for urea is 250-350 USD/mt. The synthesis of the DESs was started by mixing choline chloride (C₅H₁₄ClNO) powder which was dried in oven with appropriate amounts of glycerol (C₃H₈O₃) or urea (CH₄N₂O) powder based on their molecular weight to make DES1 or DES2. Then the mixtures were kept in the heater/shaker for 4 h at 80 °C to make transparent uniform liquid phase. These DES solutions remained liquid at room temperature after cooling down. Different dilutions of both DESs in formation brine were used also ranging from 50 vol% (equal to 17 and 21 mol% for DES1 and DES2 respectively) to 5 vol% (equal to 0.75 mol% for DES1 and 0.93 mol% for DES2). The physicochemical

Table 1 Heavy oil and formation brine properties.

Oil properties							Brine properties	
Chemical properties		Physical properties					Density @ 25 °C	1.05 g/cm ³
Molecular weight (MW)	519	Temperature (°C)	Viscosity (cP)	Density (g/cm ³)	Specific Gravity (SG)	°API	Total salinity	7.7 %wt
Total acid no. (mgKOH/mg)	1.82	25	10,970	0.965	0.968	14.68	Sodium	25.083 kg/m ³
Sulphur content (wt%)	3.10	30	9,713	0.961	0.965	15.13	Calcium	3.672 kg/m^3
Asphaltene content (wt%)	1.6	40	5,851	0.951	0.960	15.90	Magnesium	0.878 kg/m ³
Total nitrogen (mg/kg)	1948	45	2,350	0.941	0.957	16.36	Iron	0.045 kg/m^3
Total salt (Lbs/1000 bbls)	11.0	60	1,086	0.934	0.956	16.51	Chloride Sulphate	47.722 kg/m ³ 0.247 kg/m ³
Characterization factor UOP375		70	540	_	_	-	Bicarbonate	0.079 kg/m^3
		80	330	-	-	-	Filteration unit	0.45 μm

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