



Utilization of Co_3O_4 nanoparticles for reducing precipitation of asphaltene during CO_2 injection



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ABSTRACT

Injection of CO_2 , as a redundant gas, is considered as one of the most viable options to enhance oil recovery. The performance of this process may though be compromised in the event of severe precipitation and deposition of asphaltene. In this study, the effect of Co_3O_4 nanoparticles with various concentrations of 0.01, 0.1 and 1.0 wt% on precipitation of asphaltene during CO_2 injection was investigated by means of interfacial tension (IFT) measurements. The addition of Co_3O_4 nanoparticles to synthetic solutions comprising of n-heptane, toluene and 5 wt% asphaltene and crude oil caused the IFT to decrease linearly as a function of pressure with two different slopes. In the first region, CO_2 dissolved in the oil phase while in the second region, asphaltene was prone to precipitation. The IFT reduced at lower slope in the second region. It was found that the addition of nanoparticles to the synthetic and crude oils resulted in more asphaltene particles adsorption on the surface of nanoparticles. In addition, more stable dispersion of asphaltene nano-aggregates could be attained. Consequently, the ultimate recovery of asphaltene oil would take place in reservoir conditions during CO_2 injection due to less aggregation and precipitation of asphaltene particles. Finally, the optimum concentration of dispersed Co_3O_4 nanoparticles was found to be 0.1 wt%.

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1. Introduction

Enhanced oil recovery (EOR) processes are frequently required since, for instance, residual oil saturation in light reservoirs is typically about 50–60 percent of original oil in place (OOIP) (Moritis, 2006; Nobakht et al., 2008; Nabipour et al., 2007). The crude mobility resistance, among other reasons, is primarily attributed to the precipitation and deposition of asphaltenes. Asphaltene, in general, is insoluble in light alkanes such as n-pentane and n-heptane while it can dissolve in aromatics such as toluene. It has also a complex structure which, in turn, tends to become insoluble and get precipitated (Borton et al., 2010; Dashti et al., 2011). During primary oil recovery operations, due to the reservoir pressure drop, asphaltenes tend to precipitate (Hirschberg et al., 1984; Aquino-Olivos et al., 2003).

The precipitation and deposition of asphaltene would cause severe operating problems in various stages of oil production either

in reservoir rocks or production equipment (Dashti et al., 2011; Sheu and Mullins, 1995). Asphaltene precipitation in a reservoir could happen for several reasons among them i) asphaltene accumulation caused by a pressure drop in oil production and ii) injected fluids (e.g. solvents) that are used to displace oil within the reservoir during EOR processes. The latter includes, but not limited to, natural gas liquids, liquefied petroleum gas, natural gas and environmentally redundant CO_2 (Leontaritis et al., 1994; Mousavi Dehghani et al., 2007).

The gas injection EOR projects account for 40% of EOR production in the USA and the remaining 60% of EOR production are carried out by steam injection projects (Moritis, 1998; Rao and Lee, 2002). Miscible displacement is a process in which there is no phase boundary and contact surface between the displaced and displacing fluids (Rao and Lee, 2002; Benham et al., 1965). Minimum miscibility pressure (MMP) is the pressure at which the injected gas becomes completely miscible with the reservoir crude oil thus forming a single phase. The process is prevailed by the dynamic mass transfer effects between the crude oil and the injected gas. At miscibility conditions, there is no capillary forces that would result in the release of trapped crude oil. When the

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displacement pressure is more than MMP then the conditions for reaching a full miscibility can be achieved. As a result, reliable information about MMP is required to design and complete miscible gas injection EOR projects (Saini and Rao, 2010). Theoretically MMP is estimated using analytical methods based on calculations of phase equilibrium and numerical methods. These methods are based on combined simulation of flooding processes of miscible gas and equations of state (Saini and Rao, 2010; Johns et al., 1993; Wang and Orr, 1997; Jessen et al., 1998; Yuan and Johns, 2005; Rao, 1997). The approach is quick which calculates MMP based on IFT for oil and gas at different pressures and reservoir temperatures. It can also be estimated by plotting IFT against pressure and the extrapolation of this graph to IFT with zero value (Saini and Rao, 2010). In 2008, Sequeira et al. (Sequeira et al., 2008) applied Vanishing Interfacial Tension (VIT) technique to calculate first-contact miscibility pressure (FCMP) and multiple-contact miscibility pressure (MCMP) in a combined live oil/CO₂ system.

IFT between solutions of toluene and normal paraffin containing asphaltenes in the presence of carbon dioxide decreases linearly with two different slopes. The first slope of this graph corresponds to dissolution of carbon dioxide in the oil phase, while the second slope corresponds to the asphaltene precipitation at the interface of gas/oil two phases (Zolghadr et al., 2013; Hemmati-Sarapardeh et al., 2014; Mahdavi et al., 2014; Escrochi et al., 2013). It should also be noted that from the first slope, MCMP and from the second slope FCMP, can be estimated. By reducing IFT, the rate of getting miscibility versus the pressure around the onset of asphaltene precipitation decreases. In fact, the aggregation of asphaltenes in the gas/oil interface for asphaltenic crude oils would alter the slope of IFT and also reduces the VIT slope which consequently makes it difficult to attain miscibility conditions (Escrochi et al., 2013).

Recent studies indicate that the global oil and gas recovery will have to be increased up to 10% by nanotechnology due to rapid technological advancements and applications in the oil and gas industry (Kong and Ohadi, 2010; Tippee, 2009). In a study about the adsorption of asphaltenes in a heavy oil by NiO nanoparticles, it was shown that these nanoparticles have profoundly improved the adsorption capacity (Abu Tarboush and Husein, 2012). It was also reported that 1 g of NiO nanoparticles adsorbed 2.8 g of asphaltene, while adsorption capacity of commercial nickel oxide at laboratory conditions was only 15%. The type of asphaltene and particularly the asphaltene molecular weight may also influence the absorption capacity, absorption rate and tendency of asphaltene to be absorbed on the surface of nanoparticles. Nanoparticles such as NiO, Fe₂O₃, Fe₃O₄, and Co₃O₄ are reported to act as catalysts for the adsorption of asphaltene particles (Nassar et al., 2011a, 2011b). The advantages and disadvantages of using nanoparticles in EOR processes are extensively discussed by Doryani et al. (Doryani et al., 2015; Nassar et al., 2011a, 2011b). They reported that type of nanoparticles and asphaltene and their content in the oil phase are among parameters that would have profound impact on the utilization of nanoparticles to mitigate the precipitation of asphaltene.

In this study, IFT changes were analyzed for two synthetic oil solutions of toluene and n-heptane with a volumetric percentage of 60 to 40 without and with 5 wt% of asphaltene. Carbon dioxide was also used as injecting gas. With the help of VIT technique, the impact of different concentrations of Co₃O₄ nanoparticles on IFT and MMP as a function of pressure was studied. For the sake of comparison, similar experiments were also conducted for a crude oil. These results were then compared with those of the investigated synthetic oil solutions with respect to the retardation of asphaltene precipitation.

2. Materials and methods

2.1. Experimental set-up

In this study, the pendant drop technique was used to measure the equilibrium interfacial tension between different oil samples and CO₂ as a function of pressure ranging from 2.8 to 8.4 MPa for a constant temperature of 323.15 K. In the investigated oil/CO₂ mixtures, the oil droplets would form in the range of 2.8–7.7 MPa for the synthetic oil and 3.5–8.4 MPa for the investigated crude oil. In pressures below and above these ranges, the droplets either would not form or are unstable. Fig. 1 illustrates a simple schematic of the experimental apparatus IFT700.

The test facility was mainly made of a stainless steel cell with the internal volume of 20 cm³. There were two glass view windows on both sides of the cell in order to capture images of droplets, an imaging system including a camera with suitable frame rate to capture multiple digital photos of the pendant drop with dynamic movements per second, two piston-cylinder systems that both were connected to a high pressure pump for gas injection and were used for the displacement of oil and gas to the cell. The test facility also included a needle made of stainless steel with internal diameter of 1.2 ± 0.001 mm to form droplets. The needle was installed on top of the cell. An image analyzing software was also utilized based on axisymmetric drop shape analysis (ADSA) which enabled the calculation of IFT between the gas/liquid phases.

The apparatus was working such that the gas was injected into the cell at a desired pressure. Afterward ample time was allowed to reach steady-state conditions of constant pressure and temperature in the cell. The oil in the chamber that reached to a steady-state at a desired temperature was then slowly injected in a form of droplet from the top of the cell by opening a valve. Then IFT of two fluids was shown over time. The IFT measurements continued till it became constant at thermodynamic equilibrium conditions. To achieve a quicker thermodynamic equilibrium, several droplets of the synthetic oil (about 10 droplets) were injected into the cell. In these experiments, the equilibrium was considered to reach as soon as IFT did not change over time. Due to a milli Newton fluctuation of system, at first the IFT changes were plotted against the time and then the average measured IFT in the last 100 s was considered as the IFT at any desired temperature and pressure.

The density of synthetic solutions and crude oil at desired temperature and pressure was measured by using a density meter (DMA HPM Anton-Paar) and the CO₂ density at the desired temperature and pressure was obtained from the NIST data bank. The density diagram of oils that was used in this study at temperature 323.15 K and different pressures are shown in Fig. 2.

As it can be seen from Fig. 2, at a constant temperature, the crude oil density is considerably larger than the one for the synthetic oil containing asphaltene. For the same pressure, the density of synthetic oil containing asphaltene is larger than the one without asphaltene. Curve-fitted equations and R² values for Fig. 2 are presented in Table 1. The correlations, listed in Table 1, for the estimation of densities as a function of pressure would be required as input in the IFT device for measuring the interfacial tension between the oil phase and CO₂ gas.

2.2. Test materials

The investigated CO₂ gas in this study had a purity of 99.99% and the hydrocarbonaceous liquids of toluene and n-heptane were of Merck purity. The required amount of asphaltene was extracted from one of the oil fields in southern Iran (with a molecular weight of 174.8), its compositions are detailed in Table 2. The metal nanoparticles with magnetic properties are shown to have an

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