



How do metal oxide nanoparticles influence on interfacial tension of asphaltic oil-Supercritical CO₂ systems?



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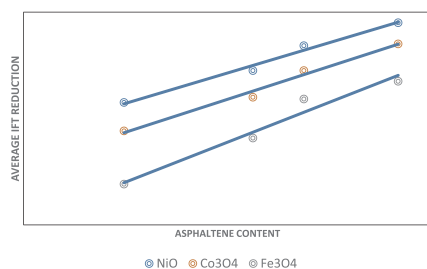
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GRAPHICAL ABSTRACT



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ABSTRACT

Often, supercritical CO₂ is injected into oil reservoirs as a miscible gas. CO₂ injection might force asphaltene particles to precipitate. To reduce asphaltene precipitation, it might be a good solution to use asphaltene adsorbents like metal oxide nanoparticles. Diffusion of supercritical CO₂, containing suspended nanoparticles, into oil slugs would decrease asphaltene precipitation as a result of nanoparticles high adsorption capacity. In this research, IFT values between the supercritical CO₂ and different crude oil types, which are saturated with NiO, Co₃O₄, and Fe₃O₄ nanoparticles, have been measured. Furthermore, effects of different crude oil properties such as API gravity, asphaltene content, and sulfur content on the nanoparticles performance are also investigated. Accordingly, nanoparticles performances order is NiO > Co₃O₄ > Fe₃O₄. In addition, it is found that oil properties effects on IFT reduction are not synergistic; so, the overall effect must be evaluated quantitatively to understand IFT reduction of the system.

1. Introduction

CO₂ is injected into underground hydrocarbon reservoirs both for storage and Enhanced-Oil-Recovery (EOR) purposes [1]. CO₂ injection into heavy oil reservoirs is problematic from the asphaltene damages aspect [2]. Presence of capillary forces in porous media is an important reason of oil trapping [3]. Near-zero IFT values between two flowing

fluids in porous media would minimize the capillary force [4]. Turning this value to zero is only possible if a miscible injection is conducted [5]. Hence, to minimize the trapped oil and maximize oil recovery, IFT behavior between reservoir fluids must be investigated carefully [6–9].

One of the new Minimum-Miscibility-Pressure (MMP) estimation methods is based on IFT measurements. According to this theory, when two fluids IFT becomes zero, these two fluids would completely

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construct a single phase [10,11]. In other words, to achieve to zero IFT, miscibility conditions of the two fluids should be provided [12–15]. pressure and IFT have inverse relation [15–18]. Extrapolation of IFT versus pressure curves would be helpful to estimate MMP. Presence of asphaltene during IFT measurements between a heavy oil sample and a gas sample would complicate these determinations [10–12,19].

Zolghadr et al. used kinetic solutions to measure IFT and MMP of different solutions and CO₂. They measured equilibrium IFT values for CO₂-C₇₊, hexadecane-CO₂, and a hydrocarbon mixture (diesel fuel and CO₂) at the equilibrium conditions using pendant drop method. They found that during the gas injection process, mass transfer between the injected gas and the reservoir fluid would be continued until the equilibrium conditions become satisfied. At the equilibrium conditions, CO₂ and reservoir oil densities would change compared to their primary state, and this, as a result, affects their IFT values [20]. Moreover, they recognized that the plot of diesel fuel-CO₂ IFT versus pressure would deviate from its original slope in the presence of non-paraffinic substances at the interface of the two phases [21].

Hemmati-Sarapardeh et al. investigated the temperature effect on CO₂-crude oil MMP. According to their results, CO₂-crude oil MMP would increase linearly with temperature. If a crude oil sample contains low amount of heavy components, its heavy components such as asphaltene would remain much more stable by increasing the pressure; consequently, asphaltene would precipitate at higher pressures. They used two crude oil samples with different resin to asphaltene ratios. Regarding the IFT versus pressure plot for the crude oil sample with higher resin to asphaltene ratio, two regions could be observed. By increasing the pressure, IFT in the first region (i.e. low-pressure region) would decline sharply, and it, however, in the second area, would decline much more gently compared to the first region. They introduced two pressure values by extrapolating each curve to the zero IFT point. The first region evaluated pressure is called MMP, and the other one is named maximum pressure (P_{max}). Besides, both values would increase linearly with temperature especially in the case of P_{max}; interestingly, in the plot of equilibrium IFT versus pressure, lower temperature results in lower slope of MMP and P_{max} curves. Considering IFT versus pressure plot for the crude oil sample with lower resin to asphaltene ratio, three regions could be distinguished. The slope turning point would occur at lower pressures compared with the last crude oil sample due to the presence of much more heavy components in this crude oil sample [22].

Different methods have been proposed to solve the asphaltene precipitation problem, i.e., adsorbing asphaltenes by nanoparticles in order to minimize their precipitation or aggregation [23]. Crystal methods, mechanical methods, using exothermal chemical substances, and using ultrasonic phenomena, which cause separation and elimination of precipitated asphaltenes, are some of these methods [11]. Using solvents is another approach against the asphaltene precipitation concern. However, Operational conditions do not allow engineers to implement these methods. disadvantages of these traditional methods Being expensive, resulting in environmental issues, and having low performance are [11]. Implementing nanotechnology seems to be applicable and appropriate to overcome these disadvantages [23,24–27]. It is worthy of attention that improving the reservoir properties is also one of the main objectives to reach to the final goal of nanofluid flooding.

Asphaltene adsorption will result in removing corrosive, fouling, coke forming, and viscous building components from heavy oil reservoirs. Asphaltene adsorption is an irreversible phenomenon regards to the solvent or oil system in which adsorption happened, and desorption could be occurred only in the presence of solvents and mixtures that dissolve asphaltenes with ease and are chromatographically strong enough to readily compete for surface adsorption sites [28]. Asphaltene adsorption have been attracting many researchers to investigate the effect of nanoparticles, especially metal oxide nanoparticles, on the heavy oil upgrading. Studying the behavior of asphaltene molecules in the presence of metal oxide nanoparticles and the adsorption

phenomenon is the main objective of such investigations [29–40].

Nassar et al.'s work was one of the pioneer studies that presented the applicability of using metal oxide nanoparticles for the purpose of asphaltene adsorption, followed by its oxidation for the purpose of heavy oil upgrading. They stated that the adsorption capacity of different metal oxide nanoparticles was in the order of CaO > Co₃O₄ > Fe₃O₄ > MgO > NiO > TiO₂ [41]. Franco et al. determined the adsorption isotherms of twelve nanoparticles, and, as a result, they concluded that isotherms were fitted with Langmuir and Freundlich models. Additionally, they reported that using these nanoparticles (metal oxide nanoparticles) at reservoir condition would be helpful to overcome the asphaltene deposition problem [42]. Abu Tarboush et al. used NiO nanoparticle to find out its asphaltene adsorption capacity in heavy oil reservoirs. They published that nanoparticles which are prepared in-place (prepared in oil matrix) have an adsorption capability of 8.2 g asphaltene per a nanoparticle gram; while, the commercial NiO adsorption capacity equals to 15% of this value [43]. Recently, Kazemzadeh et al. studied the metal oxide nanoparticles effect on the asphaltene precipitation behavior in a heavy oil reservoir and their subsequent effect on oil recovery. They concluded that presence of iron oxide nanoparticle in the solution would result in asphaltene particles adsorption onto the nanoparticles surface. This phenomenon significantly reduces the asphaltene flocculation and its subsequent damages in the porous media [44]. In continue, Kazemzadeh et al. conducted an experimental study to evaluate the effect of NiO nanoparticles on the interface of methane gas and different heavy oil samples by means of surface to volume ratios of oil drops. They demonstrated that NiO nanoparticles have high adsorption capacity; therefore, they would prevent from asphaltene aggregation and precipitation around each other and at the interface of two phases [45]. Lu et al. conducted an experimental investigation to understand the effect of Al₂O₃ nanoparticles on asphaltene adsorption during CO₂ injection. Their experiments have two sets: 1- changing the asphaltene amount at fix nanoparticle concentration, and 2- changing nanoparticle concentration at fix amount of asphaltene. They reported that increasing the nanoparticle concentration would adsorb more asphaltene and would result in better oil flow through the porous media [46]. Taborda et al. uses nanoparticles to analyze their performance to reduce the heavy oil viscosity or to improve mobility characteristics of the oil phase. They determined that increasing the nanoparticles concentration would decrease the oil viscosity up to 90%. At the end, they tested the nanoparticles in a dynamic test (core-flood), and it is found that oil recovery would increase about 16% as a consequence of using nanoparticles [47]. Hassanpour et al. examined the applicability of using Co₃O₄ nanoparticles to reduce the asphaltene problems using IFT-pressure curves. They concluded that two region exist in which the first region relates to the condition of CO₂ dissolution into the oil phase and the second region appears when asphaltenes start to precipitate. So, the slope of second region curves would be lower than the first region one [48].

In this research, IFT behavior between different asphaltenic oil samples, saturated with metal oxide nanoparticles, and supercritical CO₂ is investigated; in addition, the effects of several oil properties, namely API gravity, asphaltene content, and sulfur content on IFT values are discussed. To clarify, it is the first time that the performance of metal oxide nanoparticles to adsorb asphaltenes by means of IFT curves is investigated by considering the oil properties variation. Moreover, comparative behavior of IFT versus pressure curves is scrutinized as the composition of crude oil types varies with and without nanoparticles.

2. Methodology and material

2.1. Experimental setup

The setup by which IFT values are measured consists of an observable high pressure chamber and a high pressure glass, placed in

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