

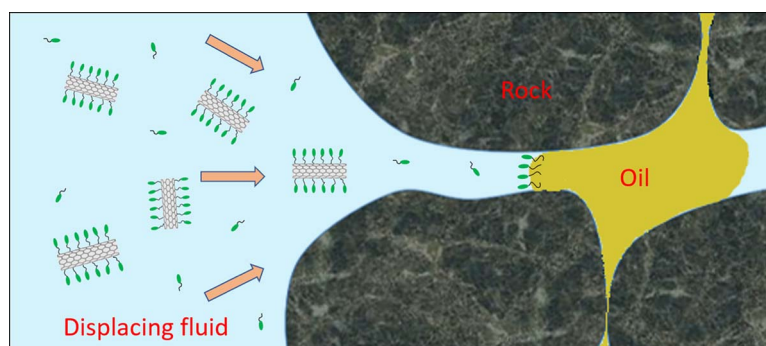


Full Length Article

Using carbonaceous nanoparticles as surfactant carrier in enhanced oil recovery: A laboratory study

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GRAPHICAL ABSTRACT



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ABSTRACT

Carbonaceous nanoparticles multi-walled carbon nanotubes (MWNTs) and carbon blacks (CBs) exhibit promising properties for potential applications in crude oil production. The combination of large specific surface area and the strong affinity toward surfactants of nanoparticles mark their candidacy for delivering surfactant deep inside the reservoir. This study is aimed to assess the feasibility of surfactant carriers in tertiary oil recovery. Stable dispersions of aqueous-phase MWNTs or CBs that are formulated and able to propagate through the reservoir medium (3 wt% brine and 60 °C) were first examined as a prerequisite for reservoir application. Competitive adsorption of surfactant on nanoparticles was beneficial to decrease adsorptive loss on Ottawa sand at equilibrium concentration below critical micelle concentration (CMC). As a proof of concept, phase behavior of a ternary surfactant microemulsion system confirmed that the chosen nanoparticles (100 mg/L) successfully delivered surfactants and spontaneously released them to the O/W interface. The observed phenomenon is in accordance with calculation of the Gibbs free energy associated with oil/water/surfactant system. Besides, surfactants carried by nanoparticles achieved equilibrium ultralow interfacial tension between excess oil and aqueous phase similar to the value of surfactant-only formulation (0.007–0.009 mN/m). In one-dimensional sand pack tests, injection of MWNT-surfactant blend achieved faster and higher tertiary recovery than surfactant-only formulation, with cumulative tertiary oil recovery of 42.7% versus 38.1%. It has been noticed that once surfactant been released, destabilization of nanoparticle dispersion occurred and thus increased their retention in porous medium. In case of tight formations, further improvements may be addressed by applying functionalized carbonaceous nanoparticles to assure their transport in porous media after release of surfactant.

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1. Introduction

Surfactant flooding is an enhanced oil recovery (EOR) technique, which mobilizes trapped oil by injecting finite slug of surfactant-only or mixture of surfactant/polymer solution into reservoir mainly to dramatically reduce the residual oil/water interfacial tension (IFT) [1–3]. Surfactant flooding was proven for decades a technically viable method from extensive laboratory efforts and numerous pilot tests, however, large field scale implementations have yet been realized due to the complexity of chemical flooding design and in many instances excessive adsorption of surfactant on formation rocks which adversely challenges economic viability of the projects [2,4].

The controlling mechanisms of adsorptive losses onto rock surfaces are complex phenomena depending on various parameters, including surfactant characteristics, e.g. type of surfactant, functional group, alkyl chain length, molecular weight; physicochemical properties of solution, such as solution pH, electrolytes concentration, divalent ions level, reservoir temperature; as well as formation mineral composition and clay content [1,5–9]. Among these, the dominant mechanisms governing surfactants adsorption onto formation rocks from aqueous solution include ion exchange, ion pairing, hydrophobic bonding, adsorption by polarization of π electrons, and adsorption by dispersion forces [10,11]. Thus, a proper control of surfactant adsorption over the course of chemical flooding is of great essence of successful recovery operations.

Normally surfactants with same charge as formation rocks are preferred owing to the electrostatic repulsion between surfactant head and rock surface which would mitigate the surfactant adsorption, for instance, anionic surfactants are preferably used in negatively charged sandstones formations at neutral pH [5,12]. In reality, however, it is impossible to completely eliminate surfactant adsorptive loss by changing the type or electrical property of surfactants [13], simply due to the inherent heterogeneity of formation rocks. The complex mineral compositions and their organic contents allow their surface properties to be altered along with sudden changes in dissolved constituents, pH of the solution, as well as advance or recession of wetting phase, therefore leave behind patchy surfactant adsorption.

Addition of polyelectrolytes, e.g. sodium polyacrylate, polystyrene sulfonate, have been considered serving as sacrificial agents for their capability to reduce adsorption of surfactants in reservoirs rocks [12–14]. These high-molecular weight sacrificial agents are believed to irreversibly occupy adsorptive sites on the substrate thus render a competition against surfactant molecules. Nevertheless, a typical pre-flush pattern of sacrificial agent may require similar injected volume as a chemical slug, which not only offset potential cost savings on surfactants but also drag field operations up to months, leading to less viability in a full-scale field project.

Recently, nanotechnology have gained increasing attention in the petroleum industry. Successful applications of nanotechnology have been reported in reservoir characterization, drilling and completion, hydraulic fracturing, and acid diversion [15–20]. Oil and gas researchers are also exploring use of nanotechnology for solving some EOR challenges more effectively. Nyankson et al. [21] studied halloysite clay nanotubes loading with different surfactants for remediation of crude oil spill. It was stated that controlled release of surfactant from the lumen of the halloysite nanomaterial could reduce the amount of chemical wastes and cost associated with the cleanup efforts. Neves Libório De Avila et al. [22] used crosslinked polystyrene nanoparticles as surfactant carriers by trapping surfactant molecules in the nanoparticles' microstructure. Once in contact with oil phase, swelling of these nanoparticles occurred, as a result, surfactants were released into the medium before partitioning at oil/water interface. Romero-Zerón and Kittisrisawai [23] developed a sugar-based complexation formulation with surfactant/ β -cyclodextrin to prevent surfactant adsorption onto porous media. A total of 61% reduction of surfactant dynamic adsorption onto sand/kaolin blend media was evidenced for an equimolar surfactant/ β -cyclodextrin inclusion complex.

Carbonaceous nanoparticles e.g. carbon nanotubes (CNTs), fullerene, carbon blacks (CBs) are potential candidates for various applications in petroleum industry. For instance, Berlin et al. [24] explored the application of engineered CBs as carriers of hydrophobic compound (2,2',5,5'-tetrachlorobiphenyl) in detecting presence of hydrocarbons in oil reservoirs. Drexler et al. [25] reported that amphiphilic nano-hybrids of CNT/silica and CNT/alumina can simultaneously act as emulsion stabilizers as well as carriers for catalysts. The superior characteristics of carbonaceous nanoparticle involve that other hydrophobic compounds or the hydrophobic moiety of an amphiphilic molecule, e.g. long surfactant tail, will strongly attract to these nanoparticle surfaces because of the entropy-driven hydrophobic interactions. Besides, enormous specific surface area of these nanoparticles (close to 250 m²/g for nanotubes in this study) could afford substantial amount of hydrophobic molecules to be loaded onto their surfaces. Previously, Martaredona et al. [26] documented the net adsorption of sodium dodecyl benzene sulfonate (SDBS) on single-walled nanotube (SWNT) surface reaching as high as 11.6 molecules per nm².

Based on our prior studies [27], we focused on two carbonaceous nanomaterials, multi-walled nanotubes (MWNTs) and CBs to investigate their potential serving as surfactant carriers, in particular covering three aspects in dealing with the feasibility in EOR applications. First, the influence of MWNT in surfactant adsorption was quantified through the batch adsorption tests of anionic surfactant alpha olefin sulfonate (AOS) on Ottawa sand. Second, example of ultra-low-IFT microemulsions phase behavior as well as Gibbs free energy change associated with oil/water/surfactant system were examined to affirm whether the loaded surfactants could be released from the treated MWNTs surfaces to oil/water interface once met the oil phase. Third, sand pack column tests were carried out to compare the extent of oil recovery for an optimum surfactant formulation between cases of presence and absence of nanoparticles added. Moreover, stability of nanoparticle dispersion under mimic reservoir conditions (3 wt% brine and 60 °C) as well as transport and fate of these stable nano-fluids in porous medium were first examined as a prerequisite for any field applications.

2. Experimental

2.1. Materials

2.1.1. Nanomaterials

The MWNTs samples used in this study were purchased from US Research Nanomaterials, Inc (Houston, TX). The nanotube length is approximately 0.5–2 μm (μm) with an average outer diameter (OD) of approximately 5–15 nm (nm). The selected CBs nanoparticles were provided by Cabot Corporation (Billerica, MA) with primary particle size around 24 nm, and specific surface area (SSA) around 110 m²/g. Examples of electron microscopy images of these MWNTs as well as CBs have been reported previously by others [28,29].

2.1.2. Surfactants

Three anionic EOR surfactants were used in this study. Alpha olefin sulfonate (C₁₄₋₁₆SO₃Na, AOS) was manufactured and provided by Stepan Company (Northfield, IL) as 39 wt% active aqueous solution. Internal olefin sulfonate (IOS) with an average alkyl chain length of 19–23 carbons, was received from Shell Chemicals (Houston, TX) as 26.8 wt% active solution. Dioctyl sodium sulfosuccinate (AOT) was supplied by Fisher Scientific (Hampton, NH) as 99 wt% active wax. Among nonionic surfactant dispersants used, nonylphenol polyethoxylates with 30, and 40 ethylene oxide (EO) groups (NP30EO, and NP40EO, respectively) were both provided by Huntsman (Salt Lake City, UT) as 100% active. Linear alcohol ethoxylates with 40 ethoxylate groups (LA40EO) was provided by Dow Chemical Company (Midland, MI) as 100% active. All surfactants were used as received without further purification. Sodium chloride (NaCl) and calcium chloride

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