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Aggregation of silica nanoparticles and its impact on particle mobility under high-salinity conditions



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ABSTRACT

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Keywords: Silica nanoparticles Aggregation Straining Breakdown CO_2 foams that are stabilized with nanoparticles are being considered to improve volumetric sweep efficiency for CO_2 enhanced oil recovery (EOR) processes. To better understand the nanoparticle retention/transport under high salinity conditions often associated with typical hydrocarbon reservoirs, the aggregation of the surface-treated silica nanoparticles was studied as a function of salinity and nanoparticle concentration. At pH 8.5, an enhancement in aggregation kinetics occurred with increasing calcium and particle concentration. However, when the pH was reduced to ca. 3, close to the CO_2 saturated condition, the aggregation was insignificant regardless of the salinity and the particle concentration over 10 days.

In the transport test of silica nanoparticles in the sandpack column, a negligible amount of nanoparticles was retained with non-aggregated nanoparticles and for aggregates less than 235 nm regardless of flow rate. However, the retention of larger aggregates (> ca. 1000 nm) was enhanced as Darcy velocity decreased. Modeling based on a modified version of clean-bed filtration theory was able to explain experimental breakthrough curves only for the non-aggregated and slightly aggregated particles, suggesting that further considerations are essential to understand the physical effect of aggregates due to the hydrodynamic forces, hence retrieving the mobility of silica nanoparticles in the porous media.

These observations demonstrate that silica nanoparticle aggregation can be prevented by increasing flow rate and lowering pH. The findings of this study would provide an insight on the behavior of nanoparticle in the reservoirs and a guideline for the application of the nanoparticles in enhanced oil recovery operations.

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1. Introduction

Nanoparticles have an important application potential in the oil and gas industry from exploration to production and distribution. Especially for the enhanced oil recovery (EOR) processes, active research efforts are ongoing to employ nanoparticle-stabilized foam and emulsion for mobility control purposes (Aroonsri et al., 2013; Espinoza et al., 2010; Mo et al., 2012; Nguyen et al., 2014; Zhang et al., 2009). The application of nanoparticles can be more promising than the use of surfactants because of (1) the robust stability of nanoparticles on the gas–liquid interface at high-salinity and elevated temperature conditions (Worthen et al., 2012), and (2) the low retention of nanoparticles in reservoir rock when compared to surfactants (Zhang et al., 2009). To take advantage of

http://dx.doi.org/10.1016/j.petrol.2015.06.019 0920-4105/© 2015 Elsevier B.V. All rights reserved. nanoparticle-stabilized foams/emulsions, that is, to improve volumetric sweep efficiency, it is critical to understand and control their mobility and long term transport required for successful field-scale displacement of oil. Therefore, an understanding of particle aggregation, and its influence on the retention characteristics of nanoparticles is essential to effectively designing a nanoparticle-mediated CO_2 foam flooding scheme.

To generate the nanoparticle-stabilized foams, the particle concentration needs to be above a certain value to maintain foam stability, especially in a high salinity reservoir condition (Espinoza et al., 2010; Yu et al., 2012). However, when the nanoparticles are concentrated, the collisions between nanoparticles could potentially result in greater aggregation (Holthoff et al., 1996). This change in the stability of nanoparticles would result in a high affinity to other surfaces they can contact with (Solovitch et al., 2010). Aggregation is normally derived by overcoming the energy barrier in electrostatic and entropic interaction between particles. In the case of nanoparticles whose transport behavior is dictated

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by Brownian motion, the initial particle concentration is a key factor governing the aggregation kinetics (Chen and Elimelech, 2006). For oil reservoir applications, the reservoir brine usually has a high salinity (Bagaria et al., 2013) so that the electrostatic repulsion of particles is virtually eliminated (Vandesteeg et al., 1992). Therefore, the possibility of nanoparticle aggregation, when they are considered for subsurface injection, must be carefully investigated. To prevent the instability of nanoparticles in the aggregation-inducing, high-salinity environment, the surface treatment of nanoparticles should provide steric repulsion between particles keeping them stable along their long travel distance thereby ensuring the longevity of nanoparticle-stabilized foam in the reservoir (Nguyen et al., 2014).

In this study, particle aggregation and its effects on the deposition and transport behavior of the surface-treated silica nanoparticles were studied at a range of salinities and particle concentrations representative of practical field conditions. A sandpack column study was conducted to investigate the coupled effect of flow velocity and aggregation on the retention of silica nanoparticles. Two existing colloidal filtration theory-based models of nanoparticle transport in porous media were adapted and applied to describe the experimental observations, necessary for field application scale-up purposes.

2. Materials and methods

2.1. Silica nanoparticles

Spherical silica nanoparticles (nominally 5 nm diameter; see below) were provided by Nissan Chemical (Houston, TX) as a 20% w/v aqueous dispersion. The surface of the nanoparticles was treated to be hydrophilic as well as to provide its steric stabilization in water. The size distribution and the zeta potential of the nanoparticles were measured by dynamic light scattering (DLS, Malvern zetasizer); and transmission electron microscopy (TEM, FEI Tencai) was employed to supplement the size measurement. The stock suspension was diluted in the synthetic brine of different compositions to obtain the desired nanoparticle concentration for the study of the aggregation, deposition, and transport in sandpacks.

2.2. Aggregation test

Silica nanoparticle suspensions varying in concentration (0.5-4.0% w/v) were prepared in three different brines (8% NaCl; 8% NaCl+1% CaCl₂; and 8% NaCl +2% CaCl₂) and two different pH levels (3 and 8.5). The low pH value, created by the addition of HNO₃, is close to the pH of CO₂-saturated water (Peng et al., 2013). Without pH control, the high pH level was achieved when the silica nanoparticle stock solution was diluted in the brine. 40 mL of brine was transported to the vials which were kept in a stationary condition without any agitation. The particle aggregate size change over the test period was monitored by sampling 1 mL of dispersion to the cuvette at each sampling time and measuring it with DLS. Sampling was continued until the aggregate sizes became over 1000 nm.

2.3. Deposition test

2 wt% silica nanoparticles were kept in API brine (8% NaCl+2% CaCl₂) for 72 h, during which the particle aggregation slowly occurred. While the aggregation continued, the deposition test was carried out at three different times of aggregation stage using quartz crystal microbalance with dissipation (QCM-D, Biolin Scientific) in which silica Q-sensor was mounted, as a supplemental

tool to the study of nanoparticle transport (Zies et al., 2012). Before the test, the QCM-D was flushed with DI water until the frequency and dissipation signals were stabilized at zero. For each test, the aggregate suspension was injected for 10 min. The flow rate was fixed at 0.15 mL/min during the entire test period. Based on the frequency data, the mass of the adsorbed silica nanoparticle aggregates onto the silica surface were calculated using the Sauerbrey model embedded in the QCM-D software (QTool). After each test, the silica Q-sensor was rinsed with DI water and immersed in 2% sodium dodecyl sulfate solution for 30 min and rinsed with DI water and dried with nitrogen gas.

2.4. Viscosity measurement

The viscosity of the aggregate suspension was measured using a rheometer (ARES-LS1, TA Instruments) with the double-wall couette geometry (Yu, 2014). Samples of different aggregate sizes were prepared by keeping different nanoparticle concentrations in API brine for at least 48 h. All the measurements were performed in the steady rate sweep test mode between 1 and 1000 s⁻¹ in shear rate and the temperature was controlled by a circulator at 25 °C.

2.5. Transport test

For the transport test, the nanoparticle dispersion was flowed through an Ottawa sand (U.S. Silica, mean diameter: 350 µm) packed in a glass cylindrical column (2.54 cm in diameter and 30.48 cm in length. Kontes Chromaflex column. Kimble-Chase). The sand pack column was flushed with the background brine at least 30 pore volumes before the test started. The nanoparticle suspension was stored in a 400 mL floating-piston accumulator and injected from the accumulator to the column by pumping water into the accumulator. The pumping rate (2.5, 5, and 25 mL/ min) was controlled to yield a nanoparticle residence time in the column ranging from 2 to 24 min. The nanoparticle suspension was injected for ca. 4.5 pore volumes and followed by rinsing with the background particle-free brine solution for another 4.5 pore volumes. To monitor the pressure drop across the column, pressure transducers were installed at the inlet and outlet of the column. Different aggregate states of silica nanoparticles in API brine (8% NaCl and 2% CaCl₂) were tested. The influent and effluent samples were diluted in 3% trace metal grade HNO₃ and analyzed for silica concentration measurement using inductively coupled plasma-optical emission spectrometry (ICP-OES, Varian). The schematic test setup is shown in Fig. 1 and the experimental conditions are summarized in Table 1.

2.6. Modeling

Two commonly used and extensively studied colloidal transport models were adapted and employed to describe the particle retention and transport: (i) a clean-bed colloid filtration theory



Fig. 1. Schematic view of transport test setup.

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