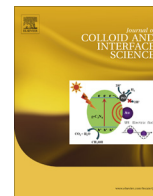




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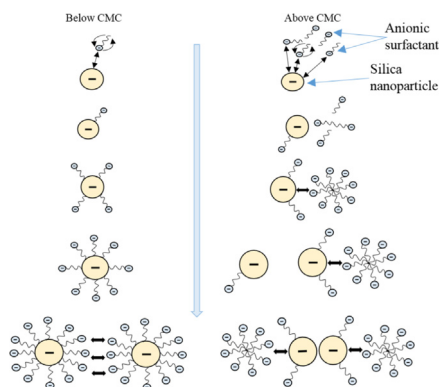
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Regular Article

Stabilising nanofluids in saline environments

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GRAPHICAL ABSTRACT



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ABSTRACT

Nanofluids (i.e. nanoparticles dispersed in a fluid) have tremendous potential in a broad range of applications, including pharmacy, medicine, water treatment, soil decontamination, or oil recovery and CO₂ geo-sequestration. In these applications nanofluid stability plays a key role, and typically robust stability is required. However, the fluids in these applications are saline, and no stability data is available for such salt-containing fluids. We thus measured and quantified nanofluid stability for a wide range of nanofluid formulations, as a function of salinity, nanoparticle content and various additives, and we investigated how this stability can be improved. Zeta sizer and dynamic light scattering (DLS) principles were used to investigate zeta potential and particle size distribution of nanoparticle-surfactant formulations. Also scanning electron microscopy was used to examine the physicochemical aspects of the suspension.

We found that the salt drastically reduced nanofluid stability (because of the screening effect on the repulsive forces between the nanoparticles), while addition of anionic surfactant improved stability. Cationic surfactants again deteriorated stability. Mechanisms for the different behaviour of the different formulations were identified and are discussed here.

We thus conclude that for achieving maximum nanofluid stability, anionic surfactant should be added.

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1. Introduction

Nanoparticles (NPs) have been widely investigated for many scientific and industrial applications, spanning from drug delivery

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[1], medicine [2,3], polymer composites [4], lubrication [5], and metal ion removal [6] to carbon geosequestration [7] and enhanced oil recovery [8–11]. Typically, thermodynamic properties of the base fluids are significantly modified by the suspended nanoparticles; thus specific and attractive properties can be tailored, including viscosity, rheology [5], thermal conductivity [12,13] and interfacial tension [14].

The successful application of NPs in saline environments (e.g. subsurface operations) requires dispersible, stable, inexpensive and injectable nano-suspensions to facilitate a uniform transport and migration of nanofluids in porous medium. However, in subsurface formations, many factors including temperature, pressure, heterogeneity, and complexity of reservoirs can dramatically impact the effectiveness of nanofluids. Increased temperature, for example, increases the kinetic energy of nanoparticles and consequently the collision rate between nanoparticles and eventually reducing nanofluid stability [15]. Another important pertinent challenge is the nanofluid stability in saline brine. It is well established that the brine salinity in subsurface formations and deep saline aquifers varies significantly and can reach very high levels [16,17]. Under such saline environments, electrolytes (e.g. NaCl) can dramatically reduce the repulsive forces between NPs and consequently accelerate particles flocculation and coagulation due to the increased rate of collision and coalescences of NPs in the suspension [18] leading to phase separation. In addition, it is known that the dispersion and stability of NPs in the base fluid can be improved by adding surface active agents such as surfactants [10,19], polymers [4], or surfactant–polymer combination [20] to the base fluid to adjust their properties for a specific application through the formation of surfactant coated nanoparticles.

A number of studies investigated the adsorption of surfactants onto NPs that were dispersed in DI water or dilute brine using contact angle measurements, adsorption isotherms of surfactant on nanoparticles, zeta potential measurements and dispersion stability in terms of nanoparticles and surfactant concentrations [19–24]. Despite the published data in the previous studies, there is no reported data about surfactant–nanosuspension dispersibility and stability at high salinity condition which is, nevertheless, very important. Thus in this study, we investigate the ability of anionic and cationic surfactants to disperse and stabilize silica NPs at high salinity conditions by measuring zeta potential and particle size of various nanofluid suspensions as a function of brine salinity (ranging from 0 wt% NaCl to 5 wt% NaCl). Sodium Dodecylsulfate (SDS) and Hexadecyltrimethylammonium Bromide (CTAB) are used as anionic and cationic surfactants respectively. The results demonstrate that anionic surfactants lead to better stability of nanofluids in comparison to cationic surfactants. This work thus leads to recognition of suitable conditions which promote better stability of nanofluids in saline environments which in turn lead to better transport of nanoparticles in porous media.

2. Experimental methodology

2.1. Materials

SiO₂ nanoparticles (porous spheres, $\rho = 2.2\text{--}2.6\text{ g cm}^{-3}$) with a purity of 99 mol% and a primary particle diameter of 5–10 nm were supplied as nano-powder by SIGMA-ALDRICH, Australia. Two surfactants, (a) anionic [Sodium Dodecylsulfate, SDS, Sigma-Aldrich, $\geq 98.5\text{ mol}\%$, Mol.wt = 288.38 g mol^{-1} , CMC = 2450 mg/l] and, (b) cationic [Hexadecyltrimethylammonium Bromide, CTAB, Sigma-Aldrich, $\geq 98\text{ mol}\%$, Mol.wt = 364.45 g mol^{-1} , CMC = 350 mg/l] were used in this study. These two surfactants were chosen for their commercial availability and the widely known properties. Binks and Rodrigues [25] reported that the particular structure of ionic surfac-

tants have no effect on the electrical properties of silica particles and thus the adsorption of mono or di-chain ionic surfactant on silica surface gives similar effects on nanoparticles surface charge.

Deionized (DI) water (Ultrapure from David Gray; conductivity = 0.02 mS cm^{-1}) was used to prepare NaCl ($\geq 99.5\text{ mol}\%$ purity, from Scharlan) solutions, nanofluids, and surfactant solutions.

2.2. Nanofluid formulation

Surfactant coated nanoparticles were prepared by sonicating NPs in surfactants formulation with appropriate ratios. Various 100 mL surfactant solutions with varying surfactant concentrations (0, 245, 735, 980, 1125, 2450, 4900, and 7350 mg/l) and NaCl concentrations (0, 0.1, 0.5, 1.0, 1.5, 2.0, 2.5, 3.0 and 5.0 wt%) were prepared by adding the surfactant powder to brine and mixing with magnetic stirrer for 2 h [10]. Note that the measured critical micelles concentrations (CMCs) with DI water were 2380 and 355 mg/l for SDS [24,26] and CTAB [27], respectively.

Subsequently, various nano-suspensions were prepared by mixing a range of silica dioxide NPs concentrations (0.05 g, 0.10 g, 0.50 g, 1.00 g, 1.25 g, 1.50 g and 2.00 g) with the aqueous phase (brine, DI water or surfactant solution) and sonicating (with a 300 VT Ultrasonic Homogenizer/BIOLOGICS instrument) for 15 min to homogenize the dispersion [28–31]. Such homogenisation is crucial for chemical stability as it is required for the zeta potential measurements, otherwise results may be biased [32]. The appearance of the dispersion was photographed at varied times when required to check the phase stability (further information in [Supplementary material](#)).

2.3. Particle size, zeta potential and SEM measurements

The physicochemical characteristics of NPs were studied using scanning electron microscopy (SEM, Zeiss Neon 40EsB FIBSEM), particles size distribution (PSD), and zeta potential (ζ) measurements. A dynamic light scattering (DLS), Zetasizer Nano ZS (Malvern Instruments, UK), was used to determine particles size distribution and the zeta potential of the nano-suspension. The direct observation is the intensity fluctuation due to the diffusion of particles undergoing Brownian motion by a laser beam [33], and this diffusion coefficient is then interpreted to a hydrodynamic diameter. Meanwhile, the surface electric charge can be estimated by zeta potential which is the measurable parameter related to the charge and electrical double layer of a solid surface in aqueous solution [34] and it is totally based on displacement of the charge in the electrical double layer due to a tangential shifting of liquid phase against the solid using external force [35].

In this study we kept the pH of the suspension at pH = 6.25 for all tested formulations. Three measurements were taken for each test, and the average value was evaluated. The standard deviation of measurements was $\pm 3\text{ mV}$ however at relatively high salinity ($>1\text{ wt}\%$ NaCl) or around the isoelectric point (IEP), the standard deviation was higher (e.g. $\pm 6\text{ mV}$).

3. Result and discussion

Improving the stability of silica nanodispersion at high salinity conditions is a key in subsurface applications. Ionic surfactant can significantly affect the surface charges of NPs and its aggregation process, and in turn the stability of nanofluids. Thus, despite the potential changes in nanofluids compositions upon injection into the treated medium in the particle field, we investigated the influence of cationic and anionic surfactants in an attempt to address the effectiveness of surfactants to improve the nanofluids stability in a saline environment.

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