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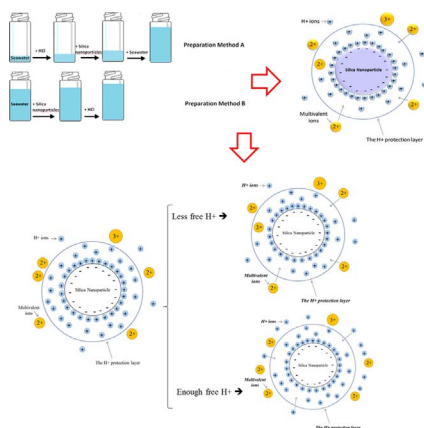
Insight into the stability of hydrophilic silica nanoparticles in seawater for Enhanced oil recovery implications

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GRAPHICAL ABSTRACT



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ABSTRACT

The stability of nanoparticles in the aqueous phase is a major challenge in the application of nanoparticles in Enhanced Oil Recovery (Nano-EOR) processes. Previous studies evaluated the performance of nanoparticles for EOR purposes; either deionized water or water at very low ionic strength was used. Nanoparticles can be easily dispersed in the deionized or low salinity water, whereas they are extremely unstable in high salinity seawater or formation water. Typically, seawater or formation brine is injected for water-flooding and EOR purposes. If we want to change the fluid–fluid or fluid–rock properties by injecting nanoparticle enhanced water, then the stability of the nanoparticles in high salinity water is extremely important. In this work, a novel and simple method to stabilize silica nanoparticles in seawater is proposed. First, the aggregation of silica nanoparticles in the presence of different ions is investigated. The results show that the presence of positive multivalent ions in the electrical double layer around nanoparticles can destabilize silica nanoparticles. In order to reduce the concentration of positive multivalent ions around silica nanoparticles, a theory based on “H⁺ protection” is proposed and its effectiveness is tested by particle size, turbidity, zeta-potential, and pH measurements. The effect of the concentrations of nanoparticles and HCl on the stability of silica nanoparticles in seawater is evaluated. Experimental results show that H⁺ protection, which can be obtained by adding HCl to the solution, can effectively stabilize silica nanoparticles in seawater. The experiments show that the size of nanoparticles in the seawater directly depends on the concentration of nanoparticles and inversely to the HCl concentration.

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1. Introduction

Recently, there has been a growing interest in application of nanoparticles in the Enhanced Oil Recovery (EOR) processes. Numerous experimental works has been published discussing the effect of nanoparticles on increasing oil recovery [1–5]. It is reported that nanoparticles can adsorb at liquid-liquid interfaces and reduce interfacial tension [6–8]. Whether nanoparticles adsorb at the interface or they change the oil-water interfacial tension is still an ongoing debate. It is accepted that this phenomenon occurs because the adsorption lowers the total energy of the system [9]. Furthermore, nanoparticles can alter surface wettability from oil-wet to water-wet [10–13] which is favorable for oil recovery.

One of the most important challenges in the application of nanoparticles for EOR methods is their stability in an aqueous solution. Nanoparticle dispersion in the aqueous phase is not a thermodynamically stable. Dispersed nanoparticles are always subject to Brownian motion with frequent collisions between them. The stability of a dispersion is thus determined by the nature of the interactions between the particles during such collisions [14]. Although the potential of silica nanoparticles in EOR processes is widely studied and their effectiveness is well-documented [3,15,16] the applications of hydrophilic silica nanoparticles are limited because the nanoparticles' high energetic hydrophilic surface, causes the silica nanoparticles to be easily agglomerated [17].

In most studies which evaluated performance of nanoparticles for EOR purposes with wettability, IFT measurement and core-flooding experiments, either deionized water or water at very low ionic strength (especially NaCl brine) is used [18–25]. However, nanoparticles are extremely unstable in seawater, formation water and concentrated ionic solutions of multiple types and charges of ions. Water flooding and EOR projects, especially for offshore reservoirs, use seawater. Even higher salinity formation water is present in the reservoirs as well. The stability of nanoparticles in these fluids is crucial for any successful nano-EOR processes. In this paper, nanoparticle stability in mixed ionic solutions is systematically investigated and a new method to stabilize hydrophilic silica nanoparticles in seawater is proposed. We first examined the effect of the most common ions in seawater on the stability of silica nanoparticles. The purpose of this step was confirmation of previous findings (see [46,66]). Based on the results of these experiments, a novel method, H^+ protection, is proposed to stabilize silica nanoparticles in seawater. The effectiveness of proposed method in the stability of silica nanoparticles in seawater is tested by particle size, turbidity, zeta-potential and pH measurements. Furthermore, in each part, the results of experiments are compared with DLVO theory.

2. Theory of nanoparticle stability

Colloidal systems consist of one or more dispersed phases and one continuous phase. On the nano-scale, due to an increase in the surface area and possible changes in the structure and composition of the surface, surface energy, and consequently the total energy of the system, increases. Nanoparticles tend to aggregate to reduce the surface energy, thereby making a colloidal dispersion at the nano-scale non-thermodynamically stable. Particles in the colloidal systems are always subject to Brownian motion and collisions frequently occur between particles. The nature of interaction between the particles during these collisions determines their stability in the solution. Van der Waals, electrical double layer, steric interaction, bridging, hydrophobic and hydration-solvation interaction are six main types of particle-particle interaction forces that can exist in the dispersion medium [26]. The sum of the attractive (van der Waals, bridging, and hydrophobic forces) and repulsive (the electrical double layer force, steric effect, and hydration force) forces between individual particles govern the stability and aggregation of particle dispersions. In general, to prepare a stable dispersion or to kinetically slow the aggregation, repulsive forces between

particles should overcome attractive forces [27].

Derjaguin, Landau, Overbeek and Verwey [28,29] proposed the DLVO theory to explain the stability of colloids in the absence of any polymer or surfactant. This theory combined two independent van der Waals attraction and electrostatic repulsion forces, explain dispersion mechanisms of colloids in the polar solution.

2.1. Electrical double layer

Helmholtz [30] first introduced and termed the idea of the electrical double layer, which was later extended by Gouy-Chapman and Stern [31,32]. The electrical double layer is a structure that appears on the surface of a charged surface when it is exposed to a fluid. The first layer, the surface charge (either positive or negative), is comprised of ions adsorbed onto the surface due to chemical interactions. This layer, which consists of a dense layer of ions of the opposite charge (counter-ions) that form around the nanoparticle, is known as the “Stern layer”. The second layer is composed of ions attracted to the surface charge via the Coulomb forces, electrically screening the first layer. This second layer is loosely associated with the surface. It is made of free ions that move in the fluid under the influence of electric attraction and thermal motion rather than being firmly anchored. It is thus called the “diffuse layer” [33]. The high concentration of counter-ions within the diffuse layer gradually decreases with increasing distance from the nanoparticle until equilibrium is reached with the ion concentration in the bulk of the solvent [31]. The distribution of ions in the electrical double layer around negatively charged nanoparticles is illustrated in Fig. 1.

The thickness of the double layer that forms at the charged surface is called “Debye Length”. Based on the electrolyte theories, interactions in the low ionic strength solutions decrease exponentially with distance or the Debye screening length. By increasing the ion concentration in the solution, due to effective screening of charges over short distances, this length decreases monotonically [35]. Greater nanoparticle surface charge and longer Debye length leads to increasing nanoparticle stability in the aqueous solution [36,37]. The thickness of the double layer is a function of ionic strength. The ionic strength can be defined as [38]:

$$I = \frac{1}{2} \sum_i z_i^2 c_i \quad (1)$$

where z and c are the charge number and molar concentration of i th ion, respectively. The Debye length (k^{-1}) in nanometer can be calculated as [39]:

$$k^{-1} = \sqrt{\frac{\epsilon_r \epsilon_0 K_B T}{e^2 \sum \rho_{\infty i} Z_i^2}} \quad (2)$$

where e is the elementary charge of an electron (C), T is the temperature (K), ϵ_0 is (F/C), ϵ_r is absolute and solution relative dielectric constant, K_B is the Boltzmann's constant, and $\rho_{\infty i}$ is the number density of ion i in the bulk solution. The electrostatic repulsion between two equally sized spherical particles with $ka > 5$ (where k is the reciprocal of Debye length (nm^{-1}) and a is the radius of spherical nanoparticles in nanometer) can be calculated by [40]:

$$V_{EL} = \frac{32\pi K_B T \epsilon_r \epsilon_0 \rho_{\infty} r \gamma^2}{k^2} e^{-kd} \quad (3)$$

where γ is the reduced surface potential and can be calculated as [40]:

$$\gamma = \tanh\left(\frac{zeE_0}{4K_B T}\right) \quad (4)$$

where E_0 is the potential on the surface. For a surface charge (E_0) below 30 mV or $ka < 5$, the electrostatic potential can be calculated by linear Poisson-Boltzmann approximation [39]:

$$V_{EL} \approx 2\pi\epsilon_r \epsilon_0 r E_0^2 k e^{-kd} \quad (5)$$

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