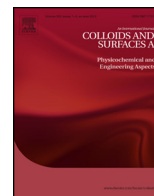




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Silica nanoparticles cationic surfactants interaction in water-oil system

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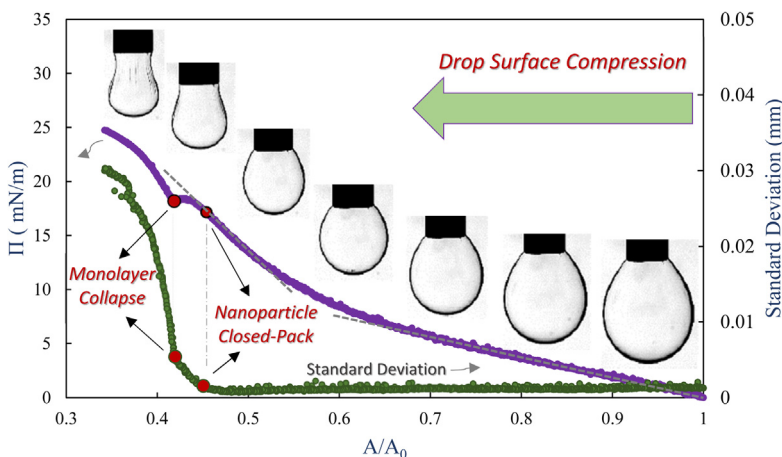
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HIGHLIGHTS

- Interfacial behavior of CTAB-Silica nanoparticle complexes are measured and discussed at low surfactant/nanoparticle ratio.
- Dynamic IFT and elasticity measurements can shed light on the CTAB-Silica nanoparticle interaction and complexes.
- Standard deviation of drop shape from Laplacian form is a powerful tool for evaluation of complexes structure at interface.
- The initiation of closed-pack state can be detected by the standard deviation values more accurately.
- Nanoparticle-surfactants complexes show surface activity higher than similar surfactant solution, however slower kinetic.

GRAPHICAL ABSTRACT

Deviation of drop shape from its Laplacian form can be an excellent parameter for indication of closed-pack state of nanoparticles and monolayer collapse.



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ABSTRACT

The aim of this experimental study is to get insight into the combined interfacial and bulk properties of silica nanoparticles and CTAB cationic surfactant mixture in water/oil systems. The study is focused on low surfactant/nanoparticle ratios to avoid extra free surfactants in the bulk. Surfactant concentration was fixed at 9.0×10^{-2} mM, (0.1 CMC) and the nanoparticle concentration varied between 0 and 5 wt.%. Drop profile analysis tensiometry (PAT) was utilized to measure the equilibrium and dynamic interfacial tension at water/heptane interface. To comprehend the layer structure of nanoparticle-surfactant complex at the interface, the interfacial rheology was also studied using drop oscillation experiments. The formation of the surfactant-nanoparticle complex layer at the drop surface and its different structures were explained using surface pressure vs. drop surface area curves and the standard deviations of experimental drop profiles from Young-Laplace equation during large compression/expansion cycles. The results demonstrate significant changes in the standard deviation at the onset of closed-packed state of surfactant-nanoparticle complex collapse. This illustrates the formation of irregular adsorbed layer (multilayer formation). The number of surfactant-nanoparticle complexes at drop surface was calculated

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by accurately assessing the surface coverage at the maximum packing of particles at the interface. For the studied concentration range, surfactant-nanoparticle complexes demonstrate surface activity higher than similar surfactant concentration, however with slower dynamics of adsorption. Elasticity measurements during large compression/expansion path can also provide more details about the structure of mono-/multi-layer formation.

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1. Introduction

Surfactant-nanoparticle mixtures also known as complex fluid systems have been an important growing research area due to numerous technological applications. Particles in nano-scale have unique properties which stem from surface specifications of particle (forces, chemistry, shape and patterns) in this scale. We are seeking for any surface active agents (Surfactants, Proteins and Polymers, etc.) that can significantly improve their intrinsic properties and behaviors. These can be either a new enhanced property or the ability to control an unexpected behavior. To do so, we need to better understand surfactant-nanoparticles interactions in the bulk and at the interface. For example, the surfactant-nanoparticle complex can act as a novel surface-modifier agent and can be used for stabilizing dispersed systems, such as foams and emulsions. Furthermore, in many situations such as chemical EOR processes in petroleum industry or blood composition in drug delivery systems, the co-existence of surfactants and fine solid particles and therefore interaction between them is inevitable.

Generally, untreated nanoparticles such as hydrophilic silica are not surface active at liquid interface but their presence in a system containing surfactant can strongly affect the interfacial properties of the liquid [1]. They can influence surfactant surface activity or be surface-activated by adsorbing surfactants and modifying their hydrophilic/hydrophobic character. This has been observed in several studies for mixture of nanoparticle and surfactant, also for some cases that particles and surfactants were located in different phases [2–7]. It has been shown that a significant change in dynamics of adsorption and interfacial rheology can occur depending on the surfactant to nanoparticle ratio.

Some systematic studies on the mixture of silica nanoparticles and CTAB for water/air systems are available [8–14]. However, liquid-liquid systems are much less studied. In general, most of available works done in this field imply that there is a synergistic effect on the stability of foam/emulsions in the presence of nanoparticle. According to these results, adsorption of the particles at the interface is attributed to the electrostatic interaction between the negatively charged surface of the particles and the positively charged surfactants. The electrostatic interaction promotes the adsorption of the surfactant onto the particle surface and increases its hydrophobicity [14,15]. Ravera et al. [7,11–15] have extensively studied complex systems containing silica nanoparticle and CTAB. They found that the charged silica nanoparticles significantly reduce the effectiveness of the CTAB solutions at water-air and water-hexane interfaces. In their experiments the surfactant to nanoparticle ratio was very high. However, there are a few studies that show the silica nanoparticles can reduce surface tension of CTAB solutions at low or all surfactant concentrations [16,17].

It seems that this contradictory observation is due to the differences between the utilized surfactant to nanoparticle ratios that highly depend on the nanoparticle size and the surfactant and nanoparticle concentrations. At constant surfactant concentration, the surfactant to nanoparticle ratio is lower for smaller nanoparticles due to higher number of nanoparticles in the system. The aim of this study is to get insight into the combined effects of sil-

ica nanoparticles and CTAB on liquid/liquid interfacial properties at low surfactant to nanoparticle ratios. Therefore, we have performed various experiments on the system containing CTAB and silica nanoparticles with the size smaller than used in the most other works. The dynamic interfacial tension (IFT) values have been measured for different particle-surfactant compositions. In comparison with other previous works in which the surfactant to nanoparticle ratios were mostly one order of magnitude higher than those used in the present work, only a few surfactants molecules adsorb at nanoparticles surface.

In addition to the dynamic interfacial tension, the response of the interfacial layer to low and high amplitude perturbations (interfacial area compression/expansion) can provide valuable information about the structure of layer of particles at liquid interfaces. At low amplitude surface area changes, the dilational visco-elasticity values can be measured. This can represent the reorganization of the mixed particle-surfactant complex layer [8,12,13,15,18]. However, the high amplitude surface area compression/expansion can also provide information about the interaction of particles and complexes at interface and add better understanding of the macroscopic structures of the particulate monolayers formed at the interface [12,19]. The traditional method of Langmuir film balance has been widely applied to obtain the surface pressure vs. surface area isotherms of particulate monolayers that consist of spherical nano- or micro-particles [20–24]. It has been observed that the composite system of nanoparticles and surfactant are able to form non regular solid structures at the water/air interface depending on the particles' hydrophobicity and the presence of these structures influence the trends of isotherms for both compression/expansion path [3,12,25]. In the course of the surface compression, the adsorbed layer continuously reorganizes its structure until particles become closed-pack and form a solid-like structure. Further compression leads to the collapse of monolayer at a certain surface area at which the particles are pushed and swiped out of the interface and form a three-dimensional structure [26]. In certain cases, some useful parameters such as particle size, the force-distance curve for pairs of approaching particles and the three-phase contact angle of particle can be estimated by analyzing the data obtained from the surface pressure vs. surface area curves [27]. These calculations depend on the accurate assessment of the surface coverage at maximum packing of particles at the surface. This closed-pack state is usually detected through the knee in the surface pressure curve which can be estimated by determining the intersection point of the two tangents at the inflection points on its either side [24].

Most of the available works presenting nanoparticle structure at interface using surface area compression/expansion are performed via Langmuir Trough method considering surface tension measurements during surface area variation. Such results have been generally correlated with microscopic image observations. In this study, we have investigated the structure and collapse of the CTAB-Silica nanoparticle complex layer on the drop surface. We have used the PAT technique which is more accurate for measurement of dynamic surface/interfacial tension in such systems. In addition, the PAT technique applied in this research can also provide us the

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