



Interfacial dynamic and dilational rheology of polyelectrolyte/surfactant two-component nanoparticle systems at air–water interface

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ABSTRACT

The interfacial characteristics of nanoparticles and consequent inter-particle interactions at the interface are poorly understood. In this work, the interfacial dynamic and corresponding dilational surface rheology of self-assembled polyelectrolyte/surfactant nanoparticles at the air–water interface are characterized. The nanoparticles are prepared from dodecyltrimethylammonium (DTAB) and poly (sodium 4-styrene-sulfonate) (PSS) by mixing them in aqueous solution. The interfacial dynamic characteristics have been carried out by comparing the surface pressure with the dilational rheological response of these nanoparticles at interface. The results indicate that this type of nanoparticles can adsorb at the interface forming a nanoparticle monolayer, which leads to the surface tension decreased markedly. The dependence of surface pressure on time shows the instability and disassembly process of nanoparticles at the interface. On the basis of these observations, it is proposed that the nanoparticles undergo a dynamic process that interface induced nanoparticles disassembly into DTAB/PSS complexes. The presence of PSS in the subphase can promote the process of nanoparticles disassembly. A transition point in dilational elasticity and viscosity response of the nanoparticles versus oscillation frequency further validate the micro dynamic process of nanoparticles and the formation of polyelectrolyte/surfactant complex monolayer at the interface.

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1. Introduction

Understanding the dynamic behavior of nanoparticles at interfaces is becoming important in recent years due to their increased applications in industrial and biomedical fields [1]. The hydrophilic–lipophilic balance of colloidal nanoparticles plays a key role in their affinity for fluid interfaces [2,3]. Nanoparticles are often employed like surfactants and used as stabilizing additives of a lot of disperse systems in different fields of practical interests [4–6]. Understanding the dynamic behavior of nanoparticle at interfaces has also become increasingly important in biology due to the closely related fields of self-assembly of biological macromolecules at biologically relevant interfaces [7–11]. By manipulating the size, zeta potential and surface hydrophobicity, nanoparticles can absorb and form self-assembled microstructures at air/water interfaces. Examples of nanoparticle adsorption, desorption, disassociation or wettability

alterations at the air–water interface have been reported [12–14]. In a few recent works interfacial rheology has been applied to liquid system containing nanoparticles in order to understand the particle/fluid interface interactions and the effect on the macroscopic interfacial properties. Various studies were focused on silica/surfactant nanoparticles [15,16]. However, the interfacial behavior of nanoparticles prepared from oppositely charged polyelectrolyte/surfactant two-component system and their corresponding dilational surface viscoelasticity are relatively scarce. In order to successfully engineer potentially transformative technologies based on the interfacial adsorption and self-assembly of the oppositely charged two-component nanoparticles at interfaces, a better understanding of the interfacial behavior for polyelectrolyte/surfactant nanoparticles is required. While some studies have been carried about on the properties of a self-assembled two-component nanoparticle system [11], little is known so far about their dynamic surface properties and dilational surface viscoelasticity.

In this study, the interfacial behavior of poly (sodium 4-styrene-sulfonate) (PSS)/dodecyltrimethylammonium (DTAB) nanoparticles adsorbing at the air–water interface is investigated.

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These nanoparticles form as a result of electrostatic interactions between the positively charged surfactant and negatively charged polyelectrolyte. Once charges on the polyelectrolyte are neutralized, a hydrophobic interaction becomes dominant. In recent years, the interactions between PSS and DTAB at air–water interface have been widely investigated using interfacial dilational viscoelasticity method [17,18]. The results led to the conclusion that the hydrophobic nature of the polystyrene backbone influenced strongly the dynamic surface properties of DTAB/PSS solutions and resulted in a modification of the classic model of polyelectrolyte/surfactant adsorption layer where only electrostatic interactions were taken into account. A peculiar property for DTAB/PSS adsorption films at the air–water interface is that the microparticles formed with DTAB concentration increased in solution. The formation of microparticles in the surface layer produced a strong drop of the dynamic surface elasticity, thereby leading to concentration dependencies of this quantity. The adsorption and complexation of PSS and DTAB at the air/water interface also can lead to interfacial gels that strongly influence foam-film drainage and stability [19,20]. Indeed, DTAB/PSS adsorption layers have also been investigated in detail by X-ray technique [21], neutron reflectivity [22], scanning probe microscopy [23] and ellipsometry [24]. Besides, the dilational surface viscoelasticity of PSS adsorbed films were studied by oscillating barrier method in broad frequency and concentration range [25]. However, these studies focus on the adsorption and dynamic surface elasticity of mixed DTAB/PSS solutions from bulk solution to the interface as a function of DTAB concentration.

While the two-dimensional structures formed from PSS and DTAB is now relatively well understood, some important questions on the dynamic interfacial characteristics and microstructure of DTAB/PSS nanoparticles accumulated at the interface and consequent interparticle interactions remain poorly understood. A question is that how the accumulated nanoparticles at the interface undergo dissociation caused by particle–particle interactions. Therefore, the aim in this work is to study the stability and dynamic structure transformation process of DTAB/PSS nanoparticles from interface to the bulk solution over time, as well as the disassociation process of these nanoparticles under surface oscillations.

Although the properties of nanoparticles formed from polyelectrolyte/surfactant and the mechanisms through which they stabilize the interfaces are not yet completely understood, it is expected that the efficiency of this type of nanoparticles in the stabilization process is related to their interfacial dynamics and dilational surface rheology. Therefore, it is necessary to understand the fundamental processes of these nanoparticles at the interface. With these premises, the goal of the current study is to look into the interfacial dynamics, stability, disassociation and possible inter-particle interactions between nanoparticles at the air–water interface. Recently, more and more attention has been paid to the nanoparticles modified by surfactant [26]. The first challenge is that the conditions in which nanoparticles can adsorb to the air–fluid interface from suspension. The ability of a nanoparticle to adsorb to the air–fluid interface is determined by a delicate balance of surface forces. The second challenge in interfacial nanoparticles is the lack of control over the microstructures that form at the interface, which limits our ability to exploit nanoparticle monolayers more broadly in advanced materials applications [27,28]. The interactions between ionic surfactants and polyelectrolyte may add significant complexity to the structure and dynamics of the laden interfaces, and in particular to the mechanical properties of the interface. It should be noted that the interfacial macroscopic mechanical properties may be strongly modified by the attachment of nanoparticles onto the interface. However, little is known about the mechanism of formation and the microstructure of nanoparticle attached to the air–water interface. Studies

in this direction may shed light on the application of these systems.

Langmuir monolayer technique is a typical two-dimensional (2D) surface chemistry approach, which is widely applied for the structure and property studies of amphiphilic molecules at the interface, such as surfactants, polymer, proteins and lipids [29–33]. One striking feature of this method is the conformation and molecular interactions can be controlled and tuned by adjusting the interfacial area or by altering the components of the subphase solution. The dilational surface viscoelasticity of monolayer allows for a better understanding of the effect of nanoparticles on the mechanical properties of such monolayers [4]. In fact, the dependence of viscoelasticity on the characteristics of the dilatational perturbation contains information about the kinetics of the relaxation processes occurring in the interfacial layer [34]. The dilational rheology approach has been shown to be effective to obtain information on the dynamic processes occurring within nanoparticle monolayers at the air–water interface [1]. Recent application of the dilational surface rheology to polyelectrolyte/surfactant solutions has allowed the discovery of surface heterogeneities in these systems [35]. The combination of these two experimental methods provides evidence of crucial interfacial dynamic behavior of polyelectrolyte/surfactant nanoparticles.

In the present study, we aim to characterize and understand the interfacial dynamics and dilatational surface viscoelasticity of DTAB/PSS nanoparticles at the air–water interface using a Langmuir monolayer approach. The adsorption, nanoparticles disassembly and dilatational viscoelasticity response of nanoparticles have been reported. Based on the experimental observations, an interfacial dynamic process and structure instability mechanism of nanoparticles formed from DTAB and PSS at the air–water interface is proposed.

2. Materials and methods

2.1. Chemicals

The cationic surfactants DTAB was purchased from Adamas Reagent Co., Ltd., with a purity of $\geq 99\%$, and used without further purification. PSS of molecular weight $M_W = \sim 75\,000\text{ g mol}^{-1}$ was purchased from Sigma–Aldrich and used without further purification. Water used in these experiments is obtained from a Millipore water purification system, having a resistivity higher than $18\text{ M}\Omega\text{ cm}$.

2.2. Methods

2.2.1. Nanoparticle synthesis

Stock solutions of DTAB and PSS at desired concentration were prepared by dissolving the required weighed amount in Millipore water. To make the nanoparticles, PSS (0.5 wt%) was added dropwise to a solution of DTAB in a glass beaker being stirred at 1200 rpm at room temperature for about 5 min. The final concentration of PSS in bulk solution is in the range of 0.03 ~ 0.06 wt%. The nanoparticles were formed as a result of electrostatic interactions between the positively charged DTAB and negatively charged PSS. Under different mixing ratios, the experimental results may appear three situations: the turbid solution, the solution with a slight blue and transparent solution. The transmittance of light through the nanoparticles solutions was measured as a function of mixing mass ratios to obtain the turbidity of the solution on an Ultraviolet Spectrophotometer (Shanghai lab-spectrum instruments Co. Ltd., China). Excitation wavelength 600 nm was used. The surfactant solution with the same concentration was used as

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