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Aqueous stability and mobility of C_{60} complexed by sodium dodecyl benzene sulfonate surfactant

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ABSTRACT

Surfactant complexation may have significant effects on the environmental behavior of nano-particles. In order to understand the ecological exposure of nano-materials, it is important to determine the stability and mobility of surfactant-complexed nano-materials in aqueous systems. In this study, the aggregation and transport of C_{60} complexed by the surfactant sodium dodecyl benzene sulfonate (SDBS) were investigated. It was found that SDBS-complexed C_{60} had a ζ-potential of −49.5 mV under near-neutral pH conditions and remained stable during an aging period of 15 days. It had a critical coagulation concentration of 550 mmol/L for NaCl, which was higher than common natural colloids and many kinds of raw nano-materials, and was comparable to those of many kinds of surface-modified nano-materials. SDBS enhanced the stability of C_{60} colloid; however, at the same time, it also enhanced the colloidal particle aggregation rate. Much higher mobility was found for SDBS-complexed C_{60} than C_{60} colloid. Increase in ionic strength, Ca^{2+} concentration or Al^{3+} concentration decreased the mobility. In general, SDBS-complexed C_{60} had high stability and mobility.

Introduction

Among many kinds of nanomaterials developed, C_{60} (Fullerene) was a landmark discovery for carbon nanomaterials and has attracted wide attention since its discovery. C_{60} has a spherical cage-like molecular structure and its discovery has led to the equally significant subsequent discovery of other fullerenes, including carbon nanotubes (CNTs) (Chen and Elimelech, 2006). C_{60} is a classical engineered material with potential applications in the areas of biomedical technology, electronics, optics, *etc.* C_{60} is highly hydrophobic, tends to aggregate and is not readily dispersed in the aqueous phase. However, the complexation of C_{60} with manufactured surfactants may enhance the aqueous stability and thus enhance its mobility in environmental media. The complexation of C_{60} with surfactant to enhance aqueous stability may occur in two main ways, *i.e.*, (1) the complexation of C_{60} discharged into the environment with residual surfactants discharged to the environment by human activities; and (2) the complexation

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of C_{60} with surfactants in industrial processes to enhance the aqueous solubility of C_{60}.

Surfactants are a diverse group of chemicals that are best known for their wide use in detergents and other cleaning products. Annual production of surfactants worldwide has risen to 12.5 million tons (Tan et al., 2010). No doubt this figure will grow with the ever growing detergent and cosmetic industries. After being used, residual surfactants are discharged into sewage systems or directly into surface waters. Most of them finally disperse in different environmental compartments such as soil, water or sediment (Ivankovic and Hrenovic, 2010). Although most surfactants are readily biodegradable, the existence of surfactants in the environment is obvious in many regions due to the continual discharge (Sanderson et al., 2006; Ying, 2006). For example, the concentration/mass fraction of one of the most common surfactants, linear alkylbenzene sulphonic acid (LAS), reached 1.1 mg/L in sewage effluents (Holt et al., 1998) and 30.2 g/kg dry mass of treated sludge (Berna et al., 1989). Up to 0.4 mg/L of LAS was measured in surface waters (Fox et al., 2000). The elevated levels of surfactants in the environment can greatly affect the transport and fate of environmental pollutants in the environment. With the possible discharge of C_{60} into the aqueous environment, the complexation of C_{60} in the aquatic environment with residual surfactants may enhance the stability and mobility of C_{60}.

On the other hand, in the application of nano-materials, it is often required that C_{60} be debundled and made hydrophilic in order to be solubilized in the aqueous phase. In these cases, surfactant treatment is usually employed to modify C_{60}. Solubilizing C_{60} by attaching surfactants to the surface through non-covalent interactions is an important approach to enhance the hydrophilic properties for solubilization. For example, C_{60} can be solubilized in water after being wrapped with triptycene-based surfactants (Torres et al., 2011). Surfactants such as Tween 20, Tween 60, Tween 80, Triton X-100, polyoxyethylene (10) lauryl ether, *n*-dodecyl trimethylammonium chloride, myristyl trimethylammonium bromide and sodium dodecyl sulfate can be used to solubilize C_{60} to facilitate its biomedical application (Hammershøj et al., 2012).

In previous investigations, the environmental aspects of raw carbon nano-materials (Prylutskyy et al., 2013; Wang et al., 2014) and oxidized carbon nano-materials (Li and Huang, 2010) have been documented. Recently, the stability and mobility of surfactant complexed carbon nano-materials have become the concern of several researchers. The aggregation and transport of one kind of carbon nano-materials, single-walled carbon nanotubes, after surfactant complexation were investigated (Bouchard et al., 2012). Due to the fact that surfactant complexation may increase the aqueous stability and mobility of C_{60}, which may subsequently facilitate the transport of environmental pollutants and cause enhanced harm to organisms, it seems necessary to understand the fate of surfactant-complexed C_{60} in the natural environment. Wang et al. (2012a) also investigated the effect of different kinds of surfactants on the mobility of C_{60}.

In this investigation, the stability and mobility of surfactant-complexed C_{60} was studied as affected by electrolytes.

1. Experimental

1.1. Materials

The C_{60} used in this study, which was claimed to have a purity of 99.9% by the producer, was purchased from Nanjing Jicang Nano-Material Company (Nanjing, China). It was used as received without further purification. Surfactant sodium dodecyl benzene sulfonate (SDBS) of analytical grade was provided by Sinopharm Chemical Reagent Co. Ltd. Tetrahydrofuran (THF) of HPLC grade was purchased from Dikma Technologies INC, USA. Glass beads with an average diameter of 1.0 mm were employed as the porous media. Before being used, the beads were soaked in 0.01 mol/L NaOH for 24 hr and then in 0.01 mol/L HNO_3 for 24 hr. After being washed with ultra-pure water, the beads were dried at 105°C for 12 hr and stored in a closed desiccator. Ultra-pure water was used throughout the experiments.

1.2. Characterization and concentration measurement

The ζ-potentials of SDBS-complexed C_{60} dispersions were determined at various pH values, and the pH values of the dispersions were adjusted using either HCl or NaOH. The C_{60} concentration was analyzed spectrophotometrically using a Hach DR-5000 UV–visible spectrophotometer at 800 nm using matched 10 mm quartz cells. It was observed that SDBS had no absorbance at 800 nm, and there was a good correlation between the absorbance at this wavelength and the C_{60} concentration. A similar phenomenon was also observed by Lin et al. (2009), who found a good correlation between the absorbance and the concentration of multi-walled carbon nanotubes (MWCNTs) at 800 nm in the presence of tannic acid.

1.3. Preparation of SDBS-complexed C_{60}

Before the preparation of SDBS-complexed C_{60} colloid, C_{60} colloid was prepared employing the method of Fortner et al. (2005) with modifications. 0.02 g C_{60} was added to 400 mL THF and N_2 was sparged to remove oxygen. Then the bottle was sealed to prevent contact with air. Sonication was employed for the solubilization of C_{60} and a sonication time of 15 min was employed. After sonication, the mixture was filtered using a 0.45 μm nylon membrane, and a transparent pink liquid was obtained. Then 400 mL water was added to the solution and a yellow solution was obtained. A three-step evaporating procedure using a rotary evaporator was then employed for removal of THF from the mixture, with an evaporating temperature of 75°C. First, the obtained 800 mL liquid was evaporated to 350 mL. Then 100 mL water was added and the obtained 450 mL liquid was evaporated to 350 mL. Finally, 100 mL water was added, the liquid was evaporated to 400 mL and the obtained liquid was cooled to room temperature and employed as C_{60} colloid in subsequent study. For the preparation of SDBS-complexed C_{60} colloid, a prescribed amount of SDBS was added to the C_{60} colloid, stirred for 12 hr and stored at room temperature for later use. Every time before being used, the C_{60} colloid was sonicated for 10 min.