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Invited article

Water chemistry controlled aggregation and photo-transformation of silver nanoparticles in environmental waters

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ABSTRACT

The inevitable release of engineered silver nanoparticles (AgNPs) into aquatic environments has drawn great concerns about its environmental toxicity and safety. Although aggregation and transformation play crucial roles in the transport and toxicity of AgNPs, how the water chemistry of environmental waters influences the aggregation and transformation of engineered AgNPs is still not well understood. In this study, the aggregation of polyvinylpyrrolidone (PVP) coated AgNPs was investigated in eight typical environmental water samples (with different ionic strengths, hardness, and dissolved organic matter (DOM) concentrations) by using UV-visible spectroscopy and dynamic light scattering. Raman spectroscopy was applied to probe the interaction of DOM with the surface of AgNPs. Further, the photo-transformation and morphology changes of AgNPs in environmental waters were studied by UV-visible spectroscopy, inductively coupled plasma mass spectrometry, and transmission electron microscopy. The results suggested that both electrolytes (especially Ca²⁺ and Mg²⁺) and DOM in the surface waters are key parameters for AgNP aggregation, and sunlight could accelerate the morphology change, aggregation, and further sedimentation of AgNPs. This water chemistry controlled aggregation and photo-transformation should have significant environmental impacts on the transport and toxicity of AgNPs in the aquatic environments.

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Introduction

Silver nanoparticles (AgNPs) are one of the most commonly used engineered nanomaterials due to their high electrical conductivity

and optical and anti-microbial properties (Jain et al., 2008; Kim et al., 2007). Because of the toxicity of AgNPs to microorganisms, algae, plants, and animals (Navarro et al., 2008; Marambio-Jones and Hoek, 2010; Asharani et al., 2008), the inevitable release of AgNPs into aquatic environments has drawn great concern about their environmental toxicity and safety (Benn and Westerhoff, 2008; Gottschalk and Nowack, 2011). Once released into aquatic environments, AgNPs transform physicochemically in terms of

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JOURNAL OF ENVIRONMENTAL SCIENCES WWW.jesc.ac.cn aggregation and chemical speciation, which not only modify their properties but also have great impacts on their transport, fate, and toxicity (Levard et al., 2012). Aggregation of AgNPs could decrease their toxicity to bacteria (Kvitek et al., 2008) and rainbow trout (Kalbassi et al., 2013). The oxidative dissolution of AgNPs to silver ion (Ag⁺) will increase the toxicity of AgNPs to organisms (Xiu et al., 2012; Pratsinis et al., 2013), while the reduction of Ag⁺ into AgNPs decreases the toxicity to protozoan *tetrahymena thermophila* (Juganson et al., 2013). Therefore, in recent years, study on the aggregation and chemical transformation of AgNPs.

Besides the characteristics of AgNPs such as size (Liu et al., 2010; Ma et al., 2012; Zhang et al., 2011a) and coating (Ma et al., 2012; El Badawy et al., 2010; Tejamaya et al., 2012), various environmental factors also play crucial roles in the aggregation and chemical transformation of AgNPs in aquatic environments. Generally, increasing ionic strength could destabilize AgNPs, and divalent electrolytes (Ca²⁺ and Mg²⁺) are more efficient in destabilizing AgNPs than monovalent electrolytes (i.e. Na⁺) (Huynh and Chen, 2011; Baalousha et al., 2013; El Badawy et al., 2012), while addition of dissolved organic matter (DOM) (i.e. humic or fulvic acids) could provide additional electrosteric repulsion and enhance the stability of AgNPs (Huynh and Chen, 2011; Baalousha et al., 2013). In natural waters, electrolyte type and concentration also play important roles in the aggregation of AgNPs (Piccapietra et al., 2012; Chinnapongse et al., 2011). However, the combined effects of electrolytes and DOM in natural waters on the stability of AgNPs are still not well understood (Gao et al., 2009). In addition to aggregation, AgNPs also encounter chemical and morphology changes in simulated or natural waters. Decreasing particle size (Ma et al., 2012; Zhang et al., 2011a) and solution pH (Liu et al., 2010) could accelerate the oxidative dissolution of AgNPs to Ag⁺. In ecotoxicology media (standard OECD media), morphology change, including formation of smaller or larger new AgNPs, was observed for engineered AgNPs with various coatings (Tejamaya et al., 2012). Importantly, it was found that light irradiation has great impact on the inter-species-transformation of Ag⁺ and AgNPs. Light exposure could accelerate the oxidation and dissolution of AgNPs to Ag⁺ (Grillet et al., 2013). On the other hand, Ag⁺ could also be reduced into zero-valent AgNPs by DOM under sunlight (Yin et al., 2012; Hou et al., 2013). Because of the redox instability of silver ($\varphi^{\parallel}(Ag^{+}/Ag^{\circ}) = 0.8 \text{ V}$), in our previous study, the cycle of oxidation of AgNPs to Ag⁺ and reduction of Ag⁺ to form new AgNPs was observed for AgNPs in DOM-solution under simulated and natural sunlight, which could induce changes in the morphology of AgNPs in aquatic environments (Yu et al., 2014). A related study also discovered that under sunlight, AgNPs with different coatings irreversibly aggregated and formed much larger nanoparticles or "chain"-like nanostructures in de-ionized water (Cheng et al., 2011). However, since most of these previous studies were performed in de-ionized water or synthesized water (i.e. DOM solution), there is still a lack of knowledge on the aggregation and photo-transformation of AgNPs in natural waters (Li and Lenhart, 2012).

The major objectives of this study were to evaluate how the water chemistry (i.e. ion composition and concentration, DOM) of real environmental waters influences the aggregation and photo-transformation of engineered AgNPs. To this end, eight environmental water samples with different ion compositions

and DOM concentrations were collected, and the aggregation of polyvinylpyrrolidone (PVP) coated AgNPs in these environmental waters was investigated by UV–visible spectroscopy and dynamic light scattering (DLS). The aggregation behavior of AgNPs was probed by Raman spectroscopy in simulated and natural waters. Further, the photo-transformation of AgNPs was studied in selected environmental waters by using UV–visible spectroscopy, inductively coupled plasma mass spectrometry (ICP-MS), and transmission electron microscope (TEM) characterization.

1. Materials and methods

1.1. Chemicals

Silver nitrite (AgNO₃), sodium hypophosphite (NaH₂PO₂·H₂O), and sodium hexametaphosphate ((NaPO₃)₆), of analytical grade, were purchased from Beijing Chemicals (Beijing, China). NaClO₄, Ca(ClO₄)₂, Mg(ClO₄)₂, PVP (molecular weight 3500), and benzotriazole were from Sinopharm Chemical Reagent Co. Ltd. (Shanghai, China). Suwannee River Natural Organic Matter (SRNOM), from an aquatic source, was purchased from the International Humic Substances Society (IHSS, St. Paul, MN, USA).

1.2. Synthesis and characterization of AgNPs

Considering PVP is widely used as a stabilizer and capping agent to control the shape of AgNPs and other nanoparticles (Moran et al., 2011), PVP-coated AgNPs were selected as a model to study the aggregation and photo-transformation of AgNPs in environmental waters. The AgNPs were synthesized as follows: AgNO₃ (3.40 g) was added into 20 mL de-ionized water and pre-heated at 50 °C in a water bath. The reductant was prepared by adding 1.325 g NaH₂PO₂·H₂O, 0.43 g sodium hexametaphosphate, 3.40 g PVP and 2.5 mL H₂SO₄ (1.0 mol/L) into 125 mL de-ionized water. Then, at 50 °C in the water bath, the reductant was stirred vigorously and the above prepared AgNO₃ solution was added at 20-30 drops per minute. Then, the mixture was stirred vigorously for another 30 min, and a reddish brown AgNP solution was formed. For purification of AgNPs, 150 mL of 2% (W/V) benzotriazole was added into the AgNP solution, and after 20 min incubation, the solution was centrifuged at 12,000 r/min (14,972 \times g) for 30 min. The precipitate was washed by acetone and ethanol, and dried in vacuum at 35 °C. Before use, the AgNP powder was re-dissolved in de-ionized water by ultrasonic dispersion (600 W for 5 min, KQ-600DE, Kunshan Ultrasonics, Shanghai, China). The AgNPs were then characterized by UV-visible spectroscopy, zeta potential measurement, and TEM.

TEM images were obtained from a Hitachi H-7500 (Hitachi, Japan) or JEOL 2011 (Herts, England) transmission electron microscope. The specimen for TEM was prepared by slow evaporation of solution onto a carbon-coated copper grid (T11031, Xinxing Bairui, Beijing, China) in a vacuum desiccator at room temperature.

1.3. Sampling and characterization of environmental water samples

Environmental waters, including river water, lake water, and spring water samples, were collected from Beijing. All the Download English Version:

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