



Carbonaceous nanomaterial-initiated reductive transformation of silver ions in the aqueous environment under sunlight

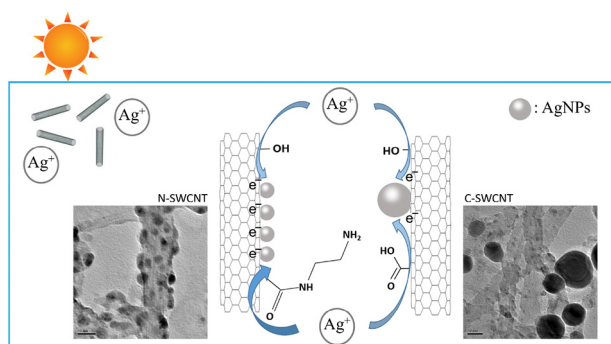
Chiaying Chen ^{*}, Yu Huang

Department of Environmental Engineering, National Chung Hsing University, Taichung City 402, Taiwan

HIGHLIGHTS

- Co-existing engineered nanomaterials could alter the potential risks of Ag^+ .
- Nitrogen-functionalized CNTs greatly enhanced the reductive transformation of Ag^+ .
- The photoreduction of Ag^+ by CNTs involves a charge transfer process.
- The formation kinetics, size, and morphology of the AgNPs depend on CNT properties.

GRAPHICAL ABSTRACT



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ABSTRACT

The aquatic systems are among the major sinks for discharged substances, and these substances will likely associate with each other. The present work, therefore, aims to study the transformation of metal ions to nanoparticles by discharged carbonaceous materials of emerging concern (e.g., carbon nanotubes (CNTs)) coexisting in the aqueous environment. Here we undertook a systematic study of the reduction of silver ions by CNT suspensions under sunlight irradiation. The formation rate of silver nanoparticles (AgNPs) is suppressed by an increasing amount of dissolved oxygen or strong solution acidity, as well as the presence of other cations. The photoreduction of Ag^+ by CNTs involves a charge transfer process between Ag^+ and the CNTs. The way in which carbonaceous nanomaterial properties influence the formation kinetics, size, and morphology of the AgNPs was examined. An enhanced sunlight-driven formation of AgNPs with highly monodispersity was observed in CNTs with nitrogen-containing functional groups due to their active electrochemical and stabilizing nature. The compiled results reveal the importance of an understanding of not only the inherent environmental behaviors of individual substances but also their interactions with concurrent substances in the environment. We demonstrated that the transformation of silver under sunlight by carbonaceous materials with different characteristics could alter the properties and potential risks of metallic species in aquatic environments.

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1. Introduction

With the extremely active development of emerging materials, a parallel effort of understanding their biological and environmental fate and behavior becomes essential to ensure their ecological safety across all applicable fields. A variety of engineered nanomaterials, including

^{*} Corresponding author.
E-mail address: chiayingchen@nchu.edu.tw (C. Chen).

carbonaceous and metallic materials, are contained in a wide range of consumer and commercial products (Benn et al., 2010; Benn and Westerhoff, 2008; Geranio et al., 2009; Smith and Rodrigues, 2015; Xu et al., 2015; Zhang et al., 2016) and are likely to enter into the environment including terrestrial and aquatic systems (Keller et al., 2014; Maurer-Jones et al., 2013), which cause both regulatory and public health concerns. Under natural conditions, it has been suggested that only a limited portion of emerging materials can persist for long periods (Gottschalk et al., 2013; Isaacson et al., 2009), and the potential transformation ultimately changes these materials from their initial discharged form. A growing number of studies have reported the transformation of materials, under the influence of the constituents in the surrounding environmental matrices. For example, environmental processes (e.g., adsorption, aggregation, dissolution, and surface reactions) of carbon or metal-based engineered nanoparticles such as silver and gold nanoparticles (AgNPs and AuNPs) may be altered through their physicochemical and biological interactions in aquatic systems (Dwivedi et al., 2015) or in the presence of natural organic matter (NOM) (Wang et al., 2016). Discharged AgNPs may undergo a series of physical and chemical transformations, such as transformed to ionic silver in biological or aqueous (Liu and Hurt, 2010) environment, or sulfidation to Ag_2S (Liu et al., 2011; Lowry et al., 2012).

Apart from anthropogenic discharge during manufacturing and application processes to aquatic ecosystems, their natural occurrence has been suggested as another source of these metallic nanoparticles. Redox-active dissolved organic matter (DOM) such as humic acid (HA) and peat fulvic acid (FA) act as reducing agents that induce the formation of AgNPs via the reduction of Ag^+ and have attracted increasing attention. Ag^+ could originate from bulk sources (e.g., effluents from textiles, plastics, and medical industries, and municipal wastewater treatment plants) (Eckelman and Graedel, 2007) or from the dissolution of AgNPs (Liu and Hurt, 2010; Liu et al., 2010). The reduction of Ag^+ by peat fulvic acid at high temperatures (70–90 °C) and high concentration (500 mg/L) under dark condition was first reported (Sal'nikov et al., 2009). Then, Akaighe et al. described AgNPs formation in the presence of humic acid (1–100 mg/L) from various sources under environmentally relevant conditions (22–90 °C) (Akaighe et al., 2011). It was suggested that the AgNPs characteristics were affected by the initial concentration of HA and the operating temperature. Aquatic HA more readily reduced Ag^+ than soil HA did, likely due to its stronger reducing ability and the accessibility of the functional groups on the surface (Akaighe et al., 2011; Struyk and Sposito, 2001). Moreover, the photochemical reduction of ionic Ag and Au to their metallic nanoparticles in natural water samples under sunlight exposure has been demonstrated in recent studies (Adegbayega et al., 2013; Hou et al., 2013; Liu et al., 2016; Yin et al., 2012). Several photochemical reaction pathways have been described, including processes mediated by superoxides from irradiated phenol group in DOM (Yin et al., 2012), reactions involving ligand-to-metal charge transfer (Hou et al., 2013), and mechanisms indicating the critical role of carbonyl-containing moieties via superoxide and charge transfer reactions (Liu et al., 2016).

While significant progress has been made towards understanding metallic nanoparticle formation triggered by ubiquitous environmental constituents, mainly NOM, in the natural aquatic system, little is known about the transformation of metal ions induced by other discharged substances. These interactions are imperative to understand because aquatic systems are one of the major sinks for discharged substances, and those substances will likely associate with each other in aqueous environments. Several recent studies highlight the important role of the coexistence of emerging pollutants/materials on the physicochemical properties and toxicity of suspended nanoparticles, or vice versa. Li et al. indicated that the dissolution and aggregation rates of AgNPs were affected in the presence of perfluorocarboxylic acids (Li et al., 2014). The presence of C_{60} -aggregates influenced the aquatic toxicity (determined by algal and *D.magna* tests) and the bioaccumulation of environmental contaminants (atrazine, methyl parathion,

pentachlorophenol (PCP), and phenanthrene), (Baun et al., 2008) as well as promoted the intake of benzo[a]pyrene and decreased cell viability (Ferreira et al., 2014). Indeed, such interactions and interplays are critical factors in evaluating the impact of industrial discharge and establishing the extent of the environmental reactivity and risks of released species. Additionally, material properties that govern the observed influence need to be identified to elucidate their contributions to the underlying mechanisms.

Furthermore, NOM has been reported to mediate and facilitate various reactions, and several studies have shown that carbonaceous materials with surface functionalization (e.g., CNTs) reveal a reactivity reminiscent of NOM (Chen and Zepp, 2015; Chen and Jafvert, 2010; Verdugo et al., 2014). For example, similar to NOM, CNTs with nitrogen-containing surface groups act not only as a precursor to but also as a source of *N*-nitrosodimethylamine (Verdugo et al., 2014). Additionally, it was demonstrated that functionalized CNTs photosensitize the production of reactive oxygen species (Chen and Zepp, 2015; Chen and Jafvert, 2010), and act as sorbents for organic pollutants (Pan and Xing, 2008) or as extraction agents for metals (Latorre et al., 2012). Considering the structure and similarities in reactivity between functionalized CNTs and NOM, including conjugated aromatic structures (Garg et al., 2011) and functionalities contained on the surface, it is reasonable to believe that CNTs that enter the aquatic environment may also play a role in metallic nanoparticles formation or transformation.

This study aims to examine the potential formation of AgNPs via reduction of silver ions induced by other discharged nanoparticles (e.g., single-walled carbon nanotubes (SWCNTs)) in the aqueous environment. While the behaviors of pristine AgNPs have been well studied, the secondary AgNPs formed via photo-reduction of Ag^+ by potentially co-existing engineered nanomaterials should also be considered. We demonstrate the potential interaction pathways and determine the correlation of CNT properties with the observed Ag^+ reductive reactivity. The formation kinetics and morphological properties of the photoinduced generation of AgNPs by CNTs were found in relation to the CNT physicochemical characters (e.g., functionality and electrochemical activity).

2. Materials and methods

2.1. Reagents and sample preparation

The chemicals were of the highest purity available from Sigma Aldrich (St. Louis, MO) and Acros Organics (Morris Plains, NJ) unless stated otherwise and were used without further purification. Aqueous solutions were prepared in water purified to $\geq 18.2 \text{ M}\Omega \text{ cm}$. Carbon nanotubes were obtained from NanoLab, Inc. (D1.5L1-5-COOH and D1.5L1-5-NH₂, Waltham, MA). According to NanoLab, Inc., pristine CNTs produced by chemical vapor deposition were refluxed in concentrated sulfuric/nitric acid and then reacted with ethylenediamine, resulting in CNTs hereafter referred to as C-SWCNTs (CNT-COOH) and N-SWCNTs (CNT-C(=O)-NH-CH₂-CH₂-NH₂), respectively (Table S1). The N-SWCNTs have surface groups with an amide linkage and a primary amine group (Table S1). Aqueous dispersions of these two types of SWCNT were prepared by probe-sonicating a mixture of 5 mg CNTs with 50 mL of pure water in an ice-water bath until an energy of ~23,400 J (15 W for a duration of 26 min) was delivered. After probe-sonication, CNT suspensions were centrifuged at 10000g for 30 min, after which 70–80% of the supernatant was collected. The stability and consistency between the batches of CNT suspensions were checked by monitoring their UV-visible absorption spectra (HITACHI, U-3900 spectrophotometer). The CNT concentrations used in the present study were approximately 6 mg/L determined using a pre-determined calibration curve of CNTs based on the UV-Vis absorbance (Fig. S1 and Text S1). For the pH-dependence study, the pH was adjusted by adding HClO_4 or NaOH solutions. No buffer solutions were added to the reaction solutions in order to minimize colloidal instability or precipitation.

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