



Mechanisms and kinetics study on the trihalomethanes formation with carbon nanoparticle precursors



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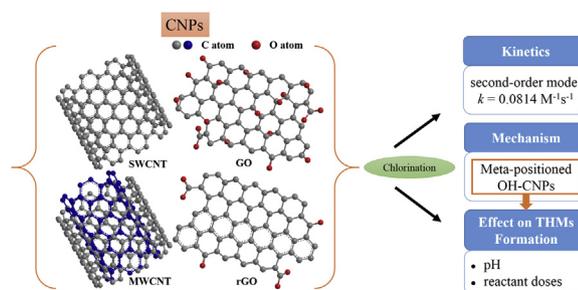
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HIGHLIGHTS

- THMs were formed during chlorination of CNPs.
- THMs formation kinetics with GO was higher than most of that with SRHA.
- OH functional groups on the CNPs were important on the THMs formation.
- CNPs structure and their functional groups may affect the formation of THMs.

GRAPHICAL ABSTRACT



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ABSTRACT

With lots of carbon nanoparticles (CNPs) applied in the industry, the possibilities of their environmental release have received much attention. As the CNPs may enter drinking water systems, and persist in water and wastewater treatment systems, their possible reaction with disinfectants should be studied. In this study, the formation of trihalomethanes (THMs) with 5 types of carbon nanotubes (CNTs), graphene oxide (GO) and reduced graphene oxide (rGO) was investigated. All CNPs could act as precursors of THMs in aqueous phase. Total concentrations of THMs formed with CNPs varied from 0.24 to 0.95 μM , much lower than that formed from chlorinated Suwannee River Natural Organic Matter (SRNOM) (approximately 9 μM). The kinetics of THMs formation with GO was $0.0814 \text{ M}^{-1} \text{ s}^{-1}$, which is higher than most of the chlorinated humic acid obtained from different natural waters. The study indicates that during chlorination, C–Cl bond could be formed on the surface of CNPs. However, carbon atoms at the middle of two *meta*-positioned OH groups on the benzene ring are more active and may prefer to form THMs with chlorine oxidation. The influences of pH and reactant doses on the formation of THMs were also discussed.

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1. Introduction

Carbon nanoparticles (CNPs) (e.g. fullerene (C₆₀), carbon nanotubes (CNTs), graphene oxide (GO), and reduced graphene oxide

(rGO)) are a class of promising carbon-based nanomaterials with a wide range of applications, such as energy-related materials, biomedicines, mechanic resonators and ultracapacitors (Robinson et al., 2008; Stoller et al., 2008; Yin et al., 2011). The possibilities of their environmental release and the associated implications have received much attention. To date, a considerable amount of research focused on the fate, transport, adsorption, dispersion and toxicity of CNPs in the environment (Chen et al., 2007; Klaine et al., 2008; Yang and Xing, 2010; Adeleye and Keller, 2014). However, knowledge of their chemical interactions with organic or inorganic materials is still very limited.

Many anthropogenic sources, such as direct discharge, vehicle emission, and industrial wastewater effluents, can release significant amounts of CNPs into the environment (Mueller and Nowack, 2008; Liu and Cohen, 2014). After their release into natural waters, CNPs may enter drinking water and wastewater treatment systems (Mueller and Nowack, 2008; Perez et al., 2009). Gottschalk et al. (2009) predicted that the concentration of C₆₀ and CNTs in sewage treatment plant effluents could be up to 32.6 ng/L and 18.4 ng/L Wang et al. (2012c) also found that residual CNPs remained in the effluent after simulated wastewater treatment. In addition, CNPs are considered a viable class of nanoparticles for environmental remediation, which may lead to their direct release into drinking water during treatment processes (Amin et al., 2014; Adeleye et al., 2016). Hyung and Kim (2009) demonstrated that CNPs could not be completely removed by conventional water treatment; hence, it is highly possible for the CNPs to remain in the drinking water system just like natural organic matter (NOM).

Disinfection is done in drinking water treatment plants to remove or inactivate bacteria and viruses that may otherwise cause waterborne diseases. However, disinfection byproducts (DBPs) are typically formed with the residuals of NOM in the water system (Rook, 1974; Dressman and Stevens, 1983; Richardson et al., 2007; Richardson and Ternes, 2014). After many years of study, the genotoxic and carcinogenic potential of DBPs to human being have received much attention (Villanueva et al., 2004; Richardson et al., 2007). Most of the existing studies on DBPs focused on examining the formation of DBPs under different source water characteristics and disinfection conditions (Glaze et al., 1977; Hua et al., 2006; Hua and Reckhow, 2008), the associations between DBPs exposure and toxicity (Savitz et al., 2006), and the determination of new halogenated DBPs (Krasner et al., 2006; Zhang and Minear, 2006; Zhang et al., 2008; Krasner, 2009; Zhai and Zhang, 2009). However, with CNPs entering the drinking water system, the possibilities of DBPs formation with CNPs remain unknown. Only a few studies have investigated the formation of DBPs with CNPs, with limited results (Wang et al., 2012a; Alpatova et al., 2013). The abilities of different CNPs to form DBPs, and the mechanism of DBPs formation with CNPs need to be well-understood (Richardson and Ternes, 2014).

Among various DBPs, trihalomethanes (THMs) are often used as surrogates for halogenated DBPs in general, to ensure that no adverse health effects have to be anticipated (Richardson et al., 2007; Sebastien et al., 2015). Therefore, this study used THMs as typical DBPs, to investigate (1) the capacity of different CNPs to act as precursors in the formation of DBPs during water chlorination, and (2) the kinetics and mechanisms of DBPs formation with CNPs. pH and reactants effects on the formation of THMs were also evaluated.

2. Materials and methods

2.1. Reagents and materials

Sodium hypochlorite (NaClO) solution (5%) was purchased from Sigma-Aldrich Corporation and diluted to 1000–2000 mg/L as Cl₂

for the further use. THMs mixture standard (Product No. 48140-U), an internal and surrogate standards were obtained from Supelco (Bellefonte, PA, USA). Suwannee River Natural Organic Matter (SRNOM) (Cat. No. 1R101N), which has carbon content of 52.47% (wt:wt), was obtained from the International Humic Substances Society (IHSS, St. Paul, MN).

Single-walled carbon nanotube (SWCNT), graphitized multi-walled carbon nanotube (G-MWCNT), aminated multi-walled carbon nanotube (NH₂-MWCNT), hydroxylated multi-walled carbon nanotube (OH-MWCNT) and carboxylated multi-walled carbon nanotube (COOH-MWCNT) were purchased from Organic Chemicals Co. (Chengdu, Sichuan Province, China). GO and rGO were purchased from Plannano Technology Co. (Tianjin, China). The synthesis methods of the materials are described in the Supplementary Material (SM). Further chemical information of the materials used in the experiment was summarized in Table S1.

2.2. Preparation of aqueous nanoparticles suspensions

A stock suspension of GO or rGO nanoparticles was prepared by adding 30 mg GO or rGO powder to 300 mL ultrapure water, followed by ultrasonication (Vibra-Cell VCX800, Sonics & Material, Newtown, CT) for 4 h. The stock suspension of GO was then kept in the dark at 4 °C until use. The rGO suspension was directly ultrasonicated for 4 h before use. The CNT powders were directly added to solutions of phosphate buffer and ultrasonicated for 1 h before use.

2.3. THMs formation with CNPs particles

All the nanoparticle samples used in the experiment were approximately 10 mg/L. The pH of suspensions was adjusted to 7 with 10 mM phosphate buffer, and spiked with 2 mg/L of NaBr as Br⁻ (a high level of bromide is found in several drinking water supplies in different parts of the world (Grossman et al., 1999; Magazinovic et al., 2004). 10 mg/L NaClO (as Cl₂) was then added to the nanoparticle suspensions. A relatively high concentration of Br⁻ was used in the study as brominated DBPs were reported to be dozens to hundreds times more toxic than their chlorinated analogues (Richardson et al., 2003).

Seven chlorinated CNP samples were formed, including a chlorinated SWCNT sample (CSNT), a chlorinated G-MWCNT sample (CMNT), a chlorinated NH₂-MWCNT sample (CMNT-NH₂), a chlorinated OH-MWCNT sample (CMNT-OH), a chlorinated COOH-MWCNT sample (CMNT-COOH), a chlorinated GO sample (CGO) and a chlorinated rGO sample (CrGO). A chlorinated SRNOM sample was also prepared, to compare with the chlorinated CNP samples. 3 mg/L of SRNOM (as TOC) was used to simulate the residual concentration in the water system. After chlorination, all the samples were stored in the dark at room temperature (22 ± 1 °C) for 3 d after which they were quenched with ascorbic acid, and then immediately extracted with Methyl *tert*-butyl ether (MtBE) and analyzed via gas chromatography. The detailed extraction procedures and adsorption experiments are provided in the SM.

The effects of various factors, including pH (6, 7 and 8), free chlorine dosages (2, 10 and 20 mg/L as Cl₂), and CNP concentrations (5, 10 and 50 mg/L) on the formation of THMs were tested. All experiments were done in duplicates.

2.4. Kinetics experiment

Similarly with the THMs formation experiment, kinetics experiment was set up as follows: 10 mg/L GO suspensions were added into the water, which was buffered to pH 7 with 10 mM phosphate. 10 mg/L NaClO (as Cl₂) was then added to the GO

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