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Sunlight affects aggregation and deposition of graphene oxide in the aquatic environment

Indranil Chowdhury ^{a,**}, Wen-Che Hou ^{a,b}, David Goodwin ^c,
Matthew Henderson ^d, Richard G. Zepp ^d, Dermont Bouchard ^{d,*}

^a National Research Council Associate, National Exposure Research Laboratory, Ecosystems Research Division, U. S. Environmental Protection Agency, Athens, GA 30605, USA

^b Department of Environmental Engineering, National Cheng Kung University, Tainan City 70101, Taiwan, ROC

^c Department of Chemistry, Johns Hopkins University, Baltimore, MD, USA

^d National Exposure Research Laboratory, Ecosystems Research Division, U. S. Environmental Protection Agency, Athens, GA 30605, USA

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ABSTRACT

In this study, we investigate the role of simulated sunlight on the physicochemical properties, aggregation, and deposition of graphene oxide (GO) in aquatic environments. Results show that light exposure under varied environmental conditions significantly impacts the physicochemical properties and aggregation/deposition behaviors of GO. Photo-transformation has negligible effects on GO surface charge, however, GO aggregation rates increase with irradiation time for direct photo-transformation under both aerobic and anaerobic conditions. Under anaerobic conditions, photo-reduced GO has a greater tendency to form aggregates than under aerobic conditions. Aggregation of photo-transformed GO is notably influenced by ion valence, with higher aggregation found in the presence of divalent ions versus monovalent, but adding natural organic matter (NOM) reduces it. QCM-D studies show that deposition of GO on surfaces coated with organic matter decreases with increased GO irradiation time, indicating a potential increase in GO mobility due to photo-transformation. General deposition trends on Suwannee River Humic Acid (SRHA)-coated surfaces are control GO > aerobically photo-transformed GO ≈ anaerobically photo-transformed GO. The release of deposited GO from SRHA-coated surfaces decreases with increased irradiation time, indicating that photo-transformed GO is strongly attached to the NOM-coated surface.

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1. Introduction

Graphene, an atomically thin, two-dimensional carbon-based nanomaterial, has been receiving great attention recently in

research and applications due to its unique electronic, optical and mechanical properties (Dreyer et al., 2010; Geim, 2009; Geim and Novoselov, 2007). Potential applications of graphene-based nanomaterials include numerous electronic

* Corresponding author. Tel.: +706 355 8333.

** Corresponding author. Tel.: +706 355 8341.

E-mail addresses: Chowdhury.Indranil@epa.gov (I. Chowdhury), Bouchard.Dermont@epa.gov (D. Bouchard).

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devices, energy storage, biomedicine, drug delivery, imaging, and even in various environmental pollution management strategies (Compton and Nguyen, 2010; Segal, 2009; Stankovich et al., 2006; Yang et al., 2013a). Besides exceptional properties and potential applications, usage of graphene-based materials is rapidly increasing due to simple mechanical and chemical process for preparation of bulk quantities of graphene (Compton and Nguyen, 2010). To facilitate the aqueous dispersion of graphene, the surface is often modified to generate graphene oxide (GO). Among different graphene materials, GO has been found to be the most toxic (Akhavan and Ghaderi, 2010; Duch et al., 2011; Hu and Zhou, 2013), which indicates how important it is to understand GO environmental exposures for safe implementation (Hu and Zhou, 2013).

The environmental fate and transport of GO has been investigated and the studies (Chowdhury et al., 2013a; Wu et al., 2013) show that GO can be highly stable against aggregation and deposition in the natural aquatic environment, and it is thus likely that GO remains in the water column where interacting sunlight can result in transformations that alter its properties. Moreover, nitrate and natural organic matter (NOM) in the natural environment can facilitate highly reactive hydroxyl radical formation under sunlight which can further react with graphene nanomaterials.

Transformation is a major factor controlling nanomaterials' fate in the environment (Lowry et al., 2012). Sunlight photolysis is one of the primary routes by which carbonaceous nanomaterials react in natural waters. Previous studies (Hou and Jafvert, 2009; Hou et al., 2010; Kong et al., 2013, 2009) have shown that sunlight exposure can photochemically transform fullerene (C_{60}), and its derivative fullerol, into CO_2 and products with significant oxygen-containing functionalities. For other nanomaterials such as carbon nanotubes, photo-transformation is strongly dependent on the involvement of reactive oxygen species (ROS) such as hydroxyl radical (Chen and Jafvert, 2010; Hou et al., 2014). Transport properties of nanomaterials have also been found to be affected by exposure to sunlight (Cheng et al., 2011; Qu et al., 2010). While nC_{60} s stability has been reported to increase after UVA irradiation due to an increased negative charge (Qu et al., 2010), oxidized multi-walled carbon nanotubes form aggregates under UVC irradiation due to loss of functional groups (Bitter et al., 2014).

Recent studies have shown that graphene is photoreactive (Gengler et al., 2013; Koinuma et al., 2012; Matsumoto et al., 2011, 2010; Zhou et al., 2012). One study (Matsumoto et al., 2011) reported that UV irradiation can reduce GO which creates many holes and defects due to photoreactions of oxygen-containing functional groups and carbon. (Matsumoto et al., 2011) and (Koinuma et al., 2012) reported formation of nanopores in the vicinity of oxygen-containing functional groups as a result of GO photoreactions in the presence of oxygen under UV irradiation. Another study (Gengler et al., 2013) observed an ultrafast photo-induced chain reaction responsible for GO reduction during UV pulsing that photo-ionizes the solvent, liberating hydrated electrons which trigger reduction (Gengler et al., 2013). Our recent study (Hou et al., 2015) on the transformation of GO under simulated sunlight found that GO readily photo-reacts under simulated sunlight

exposure, forming fragmented photoproducts similar to reduced GO (rGO) as well as low molecular-weight species. We further showed that GO photo-reactivity involves simultaneous formation of oxidative and reductive transient species, and is also dependent on dissolved oxygen level.

The objective of this study was to determine the effect of sunlight on the aggregation and deposition behavior of GO nanomaterials in aquatic environments. To our knowledge, this is the first study investigating effects of sunlight on the processes that are important in determining GO transport in the environment. Our findings indicate that photo-transformation can significantly alter transport characteristics of GO which are also a function of oxygen level during GO photo-transformation.

2. Materials and methods

2.1. Photo-transformed GO sample preparation

2.1.1. Materials

GO was obtained from Cheap Tubes Inc. (Brattleboro, VT) in a 2 mg/mL dispersion in pure water; according to the manufacturer, GO was synthesized using the modified Hummer's Method (Hummers and Offeman, 1958). All other chemicals used are the highest purity available from Sigma Aldrich (St. Louis, MO). All aqueous samples were prepared using water purified with an Aqua Solutions 2121BL system (≥ 18.0 M Ω).

2.1.2. Irradiation

The photo-transformation of GO was carried out in an Atlas SunTest CPS + solar simulator equipped with a 1 kW xenon arc lamp; details have been described elsewhere (Hou et al., 2015) and described briefly in SI. Briefly, photoreactions of GO in water were examined under two distinct reaction conditions (air equilibrated (aerobic) and oxygen deficient (anaerobic)) over a period of time (0–200 h).

2.2. Characterization of photo-transformed GO

A range of characterization was conducted to determine the physicochemical properties of photo-transformed GO. Physical dimensions were determined using a Veeco Multimode Atomic Force Microscopy (AFM) with a Nanoscope V controller and an E scanner (Bruker AXG Inc., Madison, WI). Images were taken under ScanAsyst-Air mode with a Silicon Nitride cantilever (ScanAsyst-Air, Bruker AXG Inc., Madison, WI). AFM images were further analyzed for size distribution using Nanoscope Analysis software (Bruker AXG Inc., Madison, WI). Surface functional groups on photo-transformed GO were determined by X-ray photoelectron spectroscopy (XPS). Electrokinetic and hydrodynamic properties of photo-transformed GO were measured with a ZetaSizer Nano ZS (Malvern Instruments, Worcestershire, U.K.) as a function of irradiation time and NaCl concentration at $pH\ 5.5 \pm 0.3$. Prior to measurement of electrophoretic mobility and hydrodynamic size, the sample was vortexed for 10 s. All solutions were filtered through a 100-nm filter (Anotop 25, Whatman, Middlesex, UK). Error bars describe the standard deviation based on at least three replicates.

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