



Retention and transport of graphene oxide in water-saturated limestone media



Shunan Dong^a, Yuanyuan Sun^a, Bin Gao^b, Xiaoqing Shi^{a,*}, Hongxia Xu^a, Jianfeng Wu^a, Jichun Wu^{a,**}

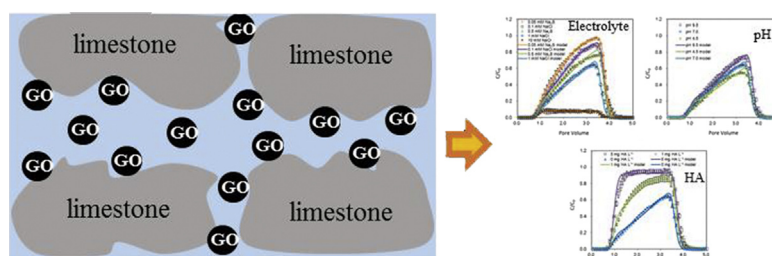
^a Key Laboratory of Surficial Geochemistry, Ministry of Education, School of Earth Sciences and Engineering, Nanjing University, Nanjing, 210023, China

^b Department of Agricultural and Biological Engineering, University of Florida, Gainesville, FL, 32611, USA

HIGHLIGHTS

- GO showed lower mobility in limestone media than that in sand and soil media.
- GO transport decreased with the increasing ionic strength and decreasing pH in limestone media.
- GO transport increased with presence of S^{2-} and HA in limestone media.
- One-site kinetic deposition model described GO breakthrough curves very well.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 4 January 2017

Received in revised form

27 March 2017

Accepted 12 April 2017

Available online 18 April 2017

Handling Editor: Jian-Ying Hu

Keywords:

Limestone

Graphene oxide

Chemical factors

Nanoparticles

Model

ABSTRACT

In this work, column experiments were conducted to investigate the transport characteristics of graphene oxide (GO) nanoparticles in limestone media under various electrolytes, solution pH, and humic acid (HA) concentration conditions. In the limestone media, GO exhibited relatively low mobility with the mass recovery rate lower than 65.2%, even when solution ionic strength was low. The presence of HA enhanced its mobility. In addition, the presence of S^{2-} , a divalent anion, also promoted GO transport in limestone media compared to Cl^- under similar ionic strength conditions through neutralizing more positive charge and thus diminishing the cation bridging. Solution pH showed slight effect on the transport of GO in limestone with the mass recovery range from 40.3% to 51.7%. Over all, decreases in solution pH, HA concentration and increases in solution ionic strength reduced the mobility of GO in the limestone media under the tested conditions. These results indicated both environmental conditions and media characteristics played important roles in controlling GO fate and transport in porous media. The one-site kinetic deposition model was applied to describe the interactions between the GO and limestone media and model simulations fitted the observed experimental data very well. As limestone is an important component of aquiferous media in subsurface, findings from this study elucidated the key factors and processes controlling the fate of GO particles in limestone media, which can inform the prediction and assessment of the risks of GO in groundwater environment.

© 2017 Elsevier Ltd. All rights reserved.

1. Introduction

Graphene oxide (GO) as a new carbon nanomaterial has shown various potentials in several areas ranging from nanomedicine to

* Corresponding author.

** Corresponding author.

E-mail addresses: shixq@nju.edu.cn (X. Shi), jcwu@nju.edu.cn (J. Wu).

energy materials areas (Allen et al., 2009; Liu et al., 2012; Zhou et al., 2015; Dong et al., 2016; Nodeh et al., 2017). With the rapidly increasing applications and production, GO will unavoidably be released into the environment, including soil and groundwater systems, and recently the emerging environmental impact of GO also has received much attention. Previous studies have indicated that GO nanoparticles can decrease the bioactivity of cells and have biotoxicity to organism (Zhao et al., 2014; Du et al., 2016; Xie et al., 2016). On the other hand, an important property of GO is that it contains abundant of functional groups and thus can serve as an effective carrier to facilitate the transport of other contaminants in porous media, which increase the potential risk of GO in the environment (Qi et al., 2014b; Zhou et al., 2016). As a result, understanding the fate and behaviors of GO in porous media comprehensively is thus critical to evaluate its environmental impact and the potential risk.

Several worthwhile laboratorial studies controlled by multiple physicochemical factors have examined the transport and retention behaviors of GO in porous media. The results have demonstrated that the decreasing of ionic strength (IS), increasing of flow rate and input particle concentration as well as the presence of nature organic matter or special kind of surfactant (e.g. anionic) can enhance the mobility of GO; while the reduction of media grain size and moisture content, and also the presence of biofilms can reduce GO mobility in porous media (Feriancikova and Xu, 2012; Lanphere et al., 2013; Liu et al., 2013; Qi et al., 2014a, 2014c; Fan et al., 2015a; Fan et al., 2015b; He et al., 2015; Liu et al., 2015; Sun et al., 2015; Xia et al., 2015; He et al., 2016). Furthermore, findings from these studies also exhibited that the established theories and models of colloid/nanoparticle can be employed for describing the transport and retention behaviors of GO particles in soil and sand porous media (Feriancikova and Xu, 2012; Liu et al., 2013; Qi et al., 2014a, 2014c; Fan et al., 2015a; Fan et al., 2015b; He et al., 2015; Liu et al., 2015; Sun et al., 2015). However, only soil or simple quartz sand system were used as the porous media for investigating the transport of GO in the previous studies, which may not represent the complex natural earth surface media systems (Christensen and Mooney, 1995; Hans Wedepohl, 1995).

Carbonates (e.g. limestone and dolomite) is an important component of sedimentary rock in the earth crust, which occupy approximately 40% of hydrocarbon resources in the world (Salehi et al., 2008). In addition, the groundwater developed in these areas is regarded as a significant section in global groundwater circulation (Bayat et al., 2015a). The fate and transport of nanoparticles in carbonates media thus are significant in the evaluation of nano-environmental risks. Nanoparticles have exhibited dynamic transport behaviors among various types of porous media (e.g. soil, quartz sand and sand coated with iron or biofilm) (Wang et al., 2012a; Neukum et al., 2014; Qi et al., 2014a; Wang et al., 2014a; Wang et al., 2014b; He et al., 2015; Pachapur et al., 2016; Wang et al., 2016a), indicating media type can be a key factor in controlling nanoparticle fate and transport in porous media (Wang et al., 2016b). Nevertheless, most of the previous studies of nanoparticle transport were in sand porous media; while only few have used limestone. The surface properties of limestone are different from sand. Previous studies have shown that limestone has a wide range of surface potential with both positive and negative values (Vdovic, 2001; Sondi et al., 2009; Chen et al., 2014; Kasha et al., 2015; Alroudhan et al., 2016). Solution chemistry, such as pH, ion type, and organics, can strongly affect the surface potential of limestone, and thus affect the mobility of nanoparticles in limestone media. The zeta potential of limestone generally decreases with the increasing solution pH, divalent anion concentration (e.g. SO_4^{2-}), and organics content; however, it increases with the increasing of cationic surfactant (e.g. octadecyl trimethyl

ammonium chloride) and the cation (e.g. Ca^{2+} , Mg^{2+} and Na^+) concentrations (Vdovic, 2001; Sondi et al., 2009; Chen et al., 2014; Kasha et al., 2015; Alroudhan et al., 2016). It has been reported that Al_2O_3 , TiO_2 and SiO_2 nanoparticles all have higher mobility in limestone and dolomite media than that in the quartz sand, meanwhile the SiO_2 nanoparticles (negative surface charge) has the relatively low mobility as compared to Al_2O_3 and TiO_2 nanoparticles (positive surface charge) (Bayat et al., 2015c). The retention of TiO_2 nanoparticles in limestone media can be enhanced with the increasing solution IS. Further, divalent Mg^{2+} has been found to be more effective than Na^+ in promoting TiO_2 nanoparticle deposition in limestone media (Bayat et al., 2015a). On the other hand, the presence of clay particles in limestone media can significantly reduce the mobility of Al_2O_3 and TiO_2 nanoparticles (Bayat et al., 2015b). However, none of the previous studies has systematically investigated the fate and transport of GO nanoparticles in limestone media, which extremely limits the prediction and monitoring on the fate of GO in the environment. Further investigations therefore are critically needed to fill the knowledge gap.

The overarching objective of this work is to understand the transport behaviors of GO nanoparticles in limestone media under various chemical conditions. Columns packed with water-saturated limestone grains were used as porous media to determine the retention and transport behaviors of GO with different combinations of experimental conditions. Mathematical models were then applied to simulate and interpret experimental data. The specific objectives are as follows: 1) determine the effect of solution electrolyte (NaCl and Na_2S used as the electrolyte separately) on the transport of GO in saturated limestone media; 2) determine the effect of pH on the transport of GO in saturated limestone media; 3) determine the effect of humic acid (HA) concentration on the transport of GO in saturated limestone media; and 4) develop and test mathematical models for fate and transport of GO in limestone media.

2. Materials and methods

2.1. Limestone media

A limestone sample collected from an outcrop in Hengyang County, Hunan Province, China was used as the porous media in this study. It was crushed and sieved into the size ranged from 0.70 to 0.90 mm for better elucidating the interactions between GO and limestone media under various conditions (Bayat et al., 2015a, 2015b; 2015c). The limestone grains were then washed sequentially with tap water and deionized (DI) water to remove ash and powder, followed by oven drying at 40 °C. The surface morphological images and properties of the limestone were determined with a Scanning Electron Microscope equipped with Energy Dispersive X-ray (SEM-EDX, JEOL JSM-6490, Japan). The limestone composition was characterized with the X-ray diffraction (XRD, DMX-III A, Japan) analyses. The major elements of the used limestone sample was determined with the X-ray fluorescence (XRF, ARL-9800, Switzerland) analyses. The specific surface area (SSA) of the limestone samples was detected with the Micropore & Chemisorption Analyzer (ASAP 2020 HD88, Micromeritics). The organic content of the limestone samples was determined from the CHN Elemental Analyser (Vario Macro, Elementar, Germany). The Fourier Transform Infrared Spectrometer (FTIR, NICOLET6700, Nicolet Continuum Microscope) was used to analyze the functional group of the limestone sample. The colloidal limestone suspensions were obtained by ultrasonication of limestone in different solution chemistry conditions for 5 min, following the method developed by Johnson et al. (1996). And then zeta potential values of the used limestone samples under varying solution chemistry conditions

Download English Version:

<https://daneshyari.com/en/article/5746129>

Download Persian Version:

<https://daneshyari.com/article/5746129>

[Daneshyari.com](https://daneshyari.com)