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Effect of physicochemical factors on transport and retention of graphene oxide in saturated media

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ABSTRACT

Fate and transport of graphene oxide (GO) have received much attention recently with the increase of GO applications. This study investigated the effect of salt concentration on the transport and retention behavior of GO particles in heterogeneous saturated porous media. Transport experiments were conducted in NaCl solutions with three concentrations (1, 20, and 50 mM) using six structurally packed columns (two homogeneous and four heterogeneous) which were made of fine and coarse grains. The results showed that GO particles had high mobility in all the homogeneous and heterogeneous columns when solution ionic strength (IS) was low. When IS was high, GO particles showed distinct transport ability in six structurally heterogeneous porous media. In homogeneous columns, decreasing ionic strength and increasing grain size increased the mobility of GO. For the column containing coarse-grained channel, the preferential flow path resulted in an early breakthrough of GO, and further larger contact area between coarse and fine grains caused a lower breakthrough peak and a stronger tailing at different IS. In the layered column, there was significant GO retention at coarse-fine grain interface where water flowed from coarse grain to fine grain. Our results indicated that the fate and transport of GO particles in the natural heterogeneous porous media was highly related to the coupled effect of medium structure and salt solution concentration.

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1. Introduction

Graphite oxide (GO) is a layered carbonaceous nanomaterial, synthesized by treating graphite with strong oxidizers (Hummers and Offeman, 1958; Dreyer et al., 2010). Due to its extraordinary mechanical and electronic properties, GO has been widely applied in many fields, such as electronic devices, drug delivery, energy storage, and composite materials (Chen et al., 2012). Consequently, the production technology of GO is developing rapidly in recent years (Lanphere et al., 2014). With the wide application and production increase, GO particles will inevitably be released into the environment. As recent studies have reported that GO have toxic effects on living organisms, the released GO particles might eventually enter human body via food chain and pose a potential threat to human health (Ahmed and Rodrigues, 2013; Chen et al., 2015). Thus, it is of importance to investigate the transport pathways and fate of GO in the environment.

A number of studies recently have focused on GO mobility in homogeneous saturated porous media, and reported that the factors affecting GO transport include solution chemistry (e.g., ionic strength, ionic type, and pH), organic matter, flow rate, water content, particle concentration, and media size (Feriancikova and Xu, 2012; Liu et al., 2013; Qi et al., 2014; Fan et al., 2015; Sun et al., 2015). As the change of salt concentration is the most common in soil and subsurface environment, many studies focused on its effect on the transport and fate of GO particles in saturated porous media (Feriancikova and Xu, 2012; Lanphere et al., 2013; Qi et al., 2014; Wang et al., 2017; Dong et al., 2017). At low salt concentration, GO exhibited high mobility in saturated porous media (Feriancikova and Xu, 2012). However, high salt concentration had increased hydrodynamic sizes of GO, and the adhesive force between GO and grain surface which further inhibited GO transport

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(Feriancikova and Xu, 2012; Lanphere et al., 2013).

Natural soil and subsurface aquifer are very complex which may exhibit a variety of heterogeneities, such as the mixture of different sized grain, layered media, and fine media containing a coarse-grained channel. Porubcan and Xu (2011) studied the transport behavior of latex particles (diameters ranging from 0.46 to 6.06 μm) in heterogeneous sand mixtures (mean grain sizes: 0.78, 0.46, and 0.23 mm), and found fine sand mixed in coarse sand could significantly increase the straining of colloidal particles in saturated porous media. In a layered configuration, a pronounced colloid retention was observed at coarse-fine grain interface where water flow moved from coarse to fine media, and the increase in contrast between two grain sizes had caused more colloidal retention at the interface (Silliman, 1995; Bradford et al., 2004, 2006). The presence of the layered media and the media containing a coarse-grained channel in porous media resulted in a preferential flow (Allaire et al., 2009), which accelerated colloid transport under saturated flow condition (Saiers et al., 1994; Bradford et al., 2004; Wang et al., 2014; Lv et al., 2016). In the system composed of coarse-grained channels embedded in a fine medium, the mass exchange at the interface between the coarse and fine grains had also been observed (Liu et al., 2014), so interface was also an important factor affecting the migration of colloids. The above discussion suggested that structural heterogeneity would produce a significant effect on the transport and fate of GO in saturated subsurface system. However, according to our current knowledge, this is the first investigation on the coupled effect of structural heterogeneity and salt concentration on GO transport and fate in saturated porous media.

The specific objectives of this work were to: (i) investigate the effect of media size on GO transport and retention behavior at different ionic strengths (ISs) in homogeneous media; and (ii) compare the similarity and difference between GO transport and retention behaviors in homogeneous and heterogeneous columns under different ISs. In this study, the breakthrough curves and retention profiles of GO were determined at three ISs in six structurally packed columns (two homogeneous and four heterogeneous). GO aggregation profiles, DLVO calculations, and tracer transport data were used to explain the transport and retention behaviors of GO in the packed column.

2. Materials and methods

2.1. GO suspension preparation

GO material (>99%) was purchased from Jicang Nano Co., Ltd (Nanjing, China). GO particles have a planar structure, their lateral diameter is 0.8–3 μm, and their layer thickness is 0.8–1.2 nm. To prepare GO stock suspension, 100 mg of GO powder was added into 100 mL distilled (DI) water, and the suspension was stirred and sonicated for 0.5 h using ultrasonic machine (Kunshan Ultrasonic Instrument Ltd., China). Before each experiment, the stock solution was diluted to 12 mg L^{-1} using DI water and NaCl solution, and the solution was also adjusted to a desired NaCl concentration. Three ISs (1, 20, and 50 mM) were selected in this study to elucidate the interaction between GO and porous media at different IS. The hydrodynamic size and electrophoretic mobility of GO at different IS were measured using a Nano Zetasizer (Nano-ZS90, Malvern Instruments Ltd., Malvern, UK). The morphologies of GO particles at 1 and 50 mM IS were determined using a transmission electron microscope (TEM JEM-2100, Japan).

2.2. Porous media

Two sizes of glass beads (Huayu Glass Beads Co.,Ltd, Hebei, China) were selected as the model porous media in this study: fine beads (size range 53–70 μm) and coarse beads (600–850 μm). The average particle diameters of the two glass beads were denoted as 62 and 725 μm for simplicity. Saturated hydraulic conductivities of fine and coarse glass beads were 0.623 and 0.054 cm min^{-1}, respectively, which were determined by constant head method. To remove metal oxides and other impurities from the glass bead surface, the glass beads were soaked in 2 M HCl at 90 °C for 24 h. Then the glass beads were repeatedly rinsed with DI water until the electric conductivity of the cleaning solution approached that of DI water. Finally, the cleaned glass beads were dried in an oven at 105 °C and stored in PTFE bottles. To measure the electrophoretic mobility of the glass bead surfaces under different ISs, the clean glass beads were grinded with a mortar, the powder was suspended into the salt solutions, and the obtained supernatants were taken for electrophoretic mobility measurements using a zetasizer (Yang et al., 2017).

2.3. Column experiments

Transparent acrylic columns (12.4 cm long and 2.2 cm inside diameter) were used in the transport experiments. The bottom and top of the columns were equipped with two layers of nylon screens (50 μm) to seal the glass beads and distribute the flow in the columns. The column experiments were run in an upward mode at a constant flow rate of 1 mL min^{-1} using a peristaltic pump (DHL-A, Huxi Analytical Instrumentation Factory, Shanghai, China). The columns were wet-packed in DI water with vibration to minimize any layering and air entrapment. The columns were packed in six different configurations (Fig. 1): (1) homogeneous column packed with fine glass beads (53–70 μm); (2) homogeneous column packed with coarse glass beads (600–850 μm); (3) column packed with the homogeneous mixture made of 50% fine and 50% coarse glass beads; (4) column packed with coarse glass beads surrounded by fine beads; (5) column packed with two layers of fine and coarse glass beads parallel to the flow; (6) column packed with two layers of fine and coarse glass beads perpendicular to the flow. Column 1 and 2 were wet-packed with fine and coarse glass beads respectively, and column 3, 4, 5, and 6 were structurally packed with the mixtures which contains 50% coarse and 50% fine glass beads based on different configurations. During the packing process, a Poly Vinyl Chloride (PVC) pipe (1.6 cm in diameter) was placed in the center of column to separate coarse and fine glass beads for column 4, and a thin plastic sheet (<1 mm) was used to separate fine and coarse glass beads for column 5. During the wet-packing process, the PVC pipe and plastic sheet were slowly pulled out and the columns were gently tapped to assure tight packing.

For each experiment, the column was first flushed with more than 5 pore volumes (PVs) of DI water to establish a steady-state saturated flow condition. Nonreactive tracer (3 mM $NaNO_3$) was then introduced into the column for 3PVs, which was followed by 5 PVs of DI water to flush the column until that no tracer was detected in the effluents. Afterward, the column was equilibrated by injecting 5 PVs of the desired electrolyte solutions (1, 20 or 50 mM NaCl). Following the pre-equilibration, 3 PVs of the GO suspension (12 mg L^{-1}) in the desired electrolyte solution was injected into the column, followed by 5 PVs elution of the desired electrolyte solution. The concentrations of nitrate and GO suspension in the effluents were determined real-time with an in-line flow cell connected to a UV–vis spectrophotometer (PERSEE TU-1900, China). The concentrations of nitrate and GO suspension were measured at the wavelengths of 235 nm and 290 nm, respectively. Linear calibration curves ($R^2 > 0.999$) were prepared to establish the relationship between absorbance and GO concentrations (Fig. S1). The tracer transport experiments were used to

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