Impact of Flow Velocity on Transport of Graphene Oxide Nanoparticles in Saturated Porous Media

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Saturated column experiments were conducted to systematically examine transport of graphene oxide nanoparticles (GONPs) in sand porous media at different solution ionic strengths (ISs) with different flow velocities. Results show that deposition of GONPs increase with decreasing IS. However, the Derjaguin–Landau–Verwey–Overbeek (DLVO) interaction energy calculations using a surface element integration technique show that the energy barrier increases with increasing IS for the sheet-shaped colloids interacting with a planar sand surface. In contrast, the presence of nanoscale protruding asperities (NPAs) can cause a decrease in the energy barrier with increasing IS, which facilitates the attachment in primary minima at higher IS. At a given IS, increasing flow velocity increases attachment efficiency on the rough sand surfaces. Torque analysis shows that the maximum hydrodynamic torques are smaller than the adhesive torques, even for GONPs located on the NPAs where the adhesions are the lowest. Consequently, the attachment efficiency cannot be reduced by increasing flow velocity. Additional column experiments confirm that deposited GONPs cannot be detached by increasing flow velocity. Conversely, increase of flow velocity may enhance approaching of GONPs in low-flow regions such as concave areas of sand surfaces and subsequent attachment. Although it has been recognized in the literature that the presence of NPAs can cause decrease of attachment efficiency with increasing flow velocity for spherical colloids under both unfavorable and favorable chemical conditions, our results highlight a different role of surface roughness in attachment and detachment of sheet-shaped colloids under unfavorable conditions.

Abbreviations: BTC, breakthrough curve; CFT, colloid filtration theory; CW, cosine wave; DI, deionized; DLVO, Derjaguin–Landau–Verwey–Overbeek; GONP, graphene oxide nanoparticle; IFV, increasing flow velocity; IS, solution ionic strength; NPA, nanoscale protruding asperity; PV, pore volume; RIS, reducing solution ionic strength; SEI, surface element integration.

Graphene oxide is a two-dimensional carbon-based nanomaterial with hydroxyl and epoxy bridge functional groups on the basal plane and carbonyl and carboxyl groups on the edges (Gao et al., 2009; Chowdhury et al., 2014). The unique physicochemical properties of graphene oxide nanoparticles (GONPs) warrant their application in various fields such as biomedicine, energy storage, and water treatment (Peng et al., 2017). The wide production and use of GONPs elevate the likelihood of their release into subsurface environments such as soil and groundwater (Wang et al., 2011; Shih et al., 2012; Wang et al., 2017a). However, GONPs were found to be the most toxic graphene derivative (Liu et al., 2011). The toxicity of GONPs was found to exist even at trace concentrations (Zhang et al., 2017) and at different organizational levels of the living system from biomolecules to animals (Seabra et al., 2014). Therefore, understanding of transport of GONPs in soil is of significant importance for accurate assessment of their environmental fate and potential ecotoxicological risks.

Deposition is the primary factor that controls transport of colloids in soil porous media (Ryan and Elimelech, 1996). Hence, investigation of GONPs deposition in porous media has received considerable attention in the literature (Feriancikova and Xu, 2012;
Solution ionic strength (IS) is one of the most important factors that influence deposition of GONPs in porous media (Lanphere et al., 2013; Liu et al., 2013; Qi et al., 2014a, 2014b; Dong et al., 2017; Wang et al., 2017a; Xia et al., 2017). Although increasing the IS does not influence \( \eta_0 \), it can increase the value of \( \alpha \) and hence \( \eta \). This is because increasing IS can increase attachment at both primary and secondary minima by decreasing the repulsive Derjaguin–Landau–Verwey–Overbeek (DLVO) energy barrier and increasing secondary-minimum depth (Hahn and O’Melia, 2004; Hahn et al., 2004; Redman et al., 2004; Kuznar and Elimelech, 2007; Liu et al., 2009). In addition to IS, other factors that can influence the value of \( \alpha \) and \( \eta \) include temperature (Wang et al., 2017b), sunlight (Chowdhury et al., 2015), natural organic matter (Duan et al., 2017), and surfactant (Fan et al., 2015; Liu et al., 2015). For example, the presence of dissolved organic matter and surfactant in solutions can increase steric repulsions between GONP and collector surfaces and thus decrease the values of \( \alpha \) and \( \eta \).

The CFT shows that increasing flow velocity (IFV) reduces \( \eta \) by decreasing \( \eta_0 \), whereas it does not influence \( \alpha \) because the CFT only considers attachment in primary minima, where the adhesive torque that a colloid experiences is significantly larger than the hydrodynamic torque (Ryan and Elimelech, 1996). However, Shen et al. (2014) and Rasmuson et al. (2017) showed that the primary minimum attachment efficiency \( (\alpha_{\text{pri}}) \) can decrease with IFV for spherical colloids under both favorable and unfavorable chemical conditions (i.e., absence and presence of a repulsive DLVO energy barrier, respectively) when surface roughness is considered. Specifically, the adhesive torque that a spherical colloid experiences at primary minima can be reduced by nanoscale protruding asperities (NPAs) and overcome by hydrodynamic torque. The influence of flow velocity on the attachment efficiencies for the sheet-shaped GONPs, however, is still unclear to date. It has been shown that both adhesive and hydrodynamic torques are influenced by colloid shape (Salerno et al., 2006; Seymour et al., 2013).

Porosity

Porous Media

Quartz sand (Sinopharm Chemical Company), with size ranging from 210 to 430 \( \mu \)m, was used as a collector for packing the columns. Similar to Zhou et al. (2011) and Wang et al. (2012), the sand was cleaned by using a sequential acid–base procedure (i.e., 1 M HCl and 1 M NaOH) to remove impurities. Surfaces of the cleaned sand were imaged by a scanning electron microscope (Hitachi S4300). To determine zeta potentials of sand surfaces, the cleaned sand was sonicated for 5 min in electrolyte solutions, and samples of the supernatants were diluted for the measurements (Tufenkji and Elimelech, 2005).

Column Transport Experiments

Column transport experiments were implemented in acrylic columns (2 cm in diameter and 9 cm long) with a top and bottom plate. The cleaned sand was wet packed in deionized (DI) water to minimize any layering and air entrapment in the columns. Using the mass of packed sand and assuming a sand density of 2.65 g cm\(^{-3}\), the porosities \( (f) \) of the sand packed columns were determined to be 0.51.

Materials and Methods

Graphene Oxide Nanoparticle Influent Suspensions

Graphene oxide nanoparticle stock suspension (2 g L\(^{-1}\)) was obtained from Sigma-Aldrich. The manufacturer reported that the density and layer thickness of the GONPs were 2.26 g cm\(^{-3}\) and 0.8 to \( \sim \)1.2 nm, respectively. The method of Wang et al. (2017a) was used to prepare GONP influent suspensions for column transport experiments, and details about this method can be found in the supplemental material. The GONPs in the prepared influent suspensions were imaged by a transmission electron microscope (Hitachi). Supplemental Fig. S1 shows that the GONPs were sheet shaped with slight angular features. Using a Zetasizer Nano ZS (Malvern Instruments), the average hydrodynamic diameters (\( D_\text{H} \)) for the GONPs in different electrolyte solutions were determined by dynamic light scattering, and zeta potentials were obtained by measuring electrophoretic mobilities.

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Column transport experiments were performed in KCl (1, 10, or 50 mM) or CaCl$_2$ (0.1, 0.5, or 1 mM) electrolyte at 25°C. For a given chemical condition, three flow velocities (9.45 × 10$^{-5}$, 1.89 × 10$^{-4}$, and 3.78 × 10$^{-4}$ m s$^{-1}$) were used. For each transport experiment, the column was first flushed with background electrolyte solution upward for at least 10 pore volumes (PVs) to standardize the IS and pH of the system. Then, 10 PVs of GONPs influent suspension were injected into the sand column (Phase A), followed by elution with GONP-free electrolyte solution (Phase B), and finally elution with DI water to release the deposited GONPs in Phase A (Phase RIS). The concentrations of GONPs in the effluents at different phases were determined by ultraviolet-visible spectrophotometer (DU Series 800, Beckman Instruments) at 227 nm using calibration curves. For selected experiments, an additional phase (i.e., flushing with the same electrolyte solution as those in Phases A and B but at a higher flow velocity) was conducted after Phase RIS to explore whether IFV can further release the deposited GONPs (denoted as Phase IFV). In addition, we have completed experiments with phases following the sequence Phase A → Phase B → Phase IFV → Phase RIS and Phase A → Phase B → Phase RIS → Phase RIS-IFV. Phase RIS-IFV means flushing the column using DI water with a higher flow velocity than that of previous phases.

Calculation of Experimental Attachment Efficiency

Measured BTCs from transport experiments were adopted to calculate experimental attachment efficiency (α$_{exp}$) using the following equation (Yao et al., 1971; Shellenberger and Logan, 2002):

$$\alpha_{exp} = \frac{-2d_A}{3(1-f)I} \ln(M_{AB})$$  

[1]

where $d_A$ is the diameter of collector, $s$ is the length of the column, and $M_{AB}$ is the fraction of GONPs recovered from Phase A and Phase B. The value of $\eta_0$ was determined using the correlation equation of Tufenkji and Elimelech (2004).

Determination of Derjaguin–Landau–Verwey–Overbeek Interaction Energies

The total DLVO interaction energy ($U_T$) between a GONP and sand surface was considered to be a sum of van der Waals attraction, electrostatic double layer energy, and Born repulsion (Ryan and Elimelech, 1996; Torkzaban and Bradford, 2016). Similar to Wang et al. (2017a), we assumed the shape of GONPs to be cuboid. Supplemental Fig. S2 shows that the sand surfaces exhibit wavy roughness. Therefore, cosine waves (CWs) were used to represent the rough sand surfaces (Fig. 1). The surface element integration (SEI) technique (Bhattacharjee and Elimelech, 1997; Bhattacharjee et al., 1998; Duffidar and Davis, 2007) was used to calculate the energy for a cuboid-shaped particle interacting with a CW surface. Briefly, the Cartesian coordinate system was used for the interaction configuration, with the $xy$ plane superposing the zero plane of the CW surface, and the $yz$ plane bisecting an arch (e.g., $A_2A_3B_3$ in Fig. 1). The CW surface was represented by $z = (1/2)D\cos(x/N)$, where $D/2$ is amplitude and $2\pi N$ is period. The CW surface was discretized into small area elements (dS). The total interaction energy was calculated as a sum of differential interaction energy between each area element on the CW surface and the particle surface. The expression used to calculate the value of $U_T$ can be expressed as

$$U_T(H) = \sum_S E(b)\mathbf{n}dS$$

[2]

where $H$ is separation distance between the particle and the surface, $\mathbf{n}$ is the unit outward normal to the sine wave surface, $k$ is the unit vector directed toward the positive $z$-axis, points $A_1$ to $A_4$ are the highest and $B_1$ to $B_3$ are the lowest points on an intersection of the cosine wave surface.

![Fig. 1. Schematic illustration of a cuboid-like particle with a cosine wave surface. dS is differential area element on the cosine wave surface. n is unit outward normal to the cosine wave surface, and k is unit vector directed toward the positive z-axis. Points A1 to A4 are the highest and B1 to B3 are the lowest points on an intersection of the cosine wave surface.](image)

The adhesive and hydrodynamic torques were calculated to determine whether GONPs can be successfully attached in primary minima or whether attached GONPs can be detached from primary minima at a given flow velocity (Bergendahl and Grasso, 2018).
The adhesive torque \( (T_A) \) is represented by the adhesive force \( (F_A) \) acting on a lever arm \( I_c \):

\[
T_A = F_A I_c
\]

The value of \( F_A \) is taken as the maximum attraction that is obtained through calculating the derivative of DLVO interaction energies with respect to \( b \). For a cuboid-shaped particle interacting with a sine wave surface, the lever arm is dependent on the contact between the particle and the surface. For example, if two arches on the CW surface are in contact with the particle, the lever arm is taken as half of the distance between the two contact lines. We considered that the cuboid-shaped particle interacted with at least two arches so that the particle can be attached stably. When the flow direction is perpendicular to a side of the cuboid-shaped particle, the hydrodynamic drag torque \( (T_D) \) that acts on the particle in the vicinity of a collector surface is given by

\[
T_D = 8 \mu R V_f P / 3
\]

where \( \mu \) is the viscosity of water, \( V_f \) is the relative velocity between the fluid and the particle at the center of the particle, and \( R \) is side length of the particle.

**Results and Discussion**

**Physicochemical Properties of Graphene Oxide Nanoparticle Sand**

Supplemental Table S2 shows measured zeta potentials of GONPs and sand at pH 7 with different ISs. The zeta potentials of GONPs and sand were negative at all ISs and were less negative with increasing IS. Similar results were reported for the GONPs in previous studies (Feriancikova and Xu, 2012; Chowdhury et al., 2014; Dong et al., 2017). For a given IS, the zeta potentials were less negative in CaCl\(_2\) than those in KCl due to the greater ability of Ca\(^{2+}\) for charge screening than K\(^+\) and charge neutralization by the Ca\(^{2+}\) (Chen and Elimelech, 2006).

Supplemental Table S2 also presents measured values of \( D_H \) for the GONPs in KCl and CaCl\(_2\) at different ISs. The values of \( D_H \) were similar in \( \leq 10 \) mM KCl and \( \leq 0.5 \) mM CaCl\(_2\) (the \( P \)-value of Student’s \( t \) test is 0.51, which is significantly \( >0.05 \)). However, the values of \( D_H \) increased significantly in 50 mM KCl and 1 mM CaCl\(_2\) (the \( P \)-value is 0.0016, which is significantly \( <0.01 \)), indicating that aggregation occurred in these cases. In addition, the value of \( D_H \) in CaCl\(_2\) at 1 mM was even larger than that in KCl at 50 mM because Ca\(^{2+}\) can significantly promote aggregation of GONPs by cross-linking of GONPs sheets via bridging of carboxylic functional groups such as COOH\(^-\) at the edges (i.e., edge-to-edge aggregation; Pham et al., 2009; Ren et al., 2014; Wang et al., 2017a).

**Deposition of Graphene Oxide Nanoparticles**

Figure 2 presents BTCs for the GONPs in sand porous media in KCl and CaCl\(_2\) at different ISs with different flow velocities \( (U) \). The normalized effluent GONP concentration \((C/C_0, \text{ where } C \text{ and } C_0 \text{ are effluent and influent concentrations of GONPs, respectively}) \) was plotted as a function of PV. Each BTC contains three segments corresponding to the three experimental phases, as described above. In Phase A, more GONPs were deposited at higher ISs. Our calculations in Supplemental Fig. S3, however, show that the interaction energy barrier increases with increasing solution IS for the sheet-shaped colloids (i.e., GONPs). Consequently, the theoretical calculations cannot explain the experimental observations. Lin and Wiesner (2012) obtained similar results for spherical nanoparticles by using the SEI technique to adequately evaluate the interaction energies. Conversely, if the Derjaguin approximation approach is used for the calculations, increasing IS will cause a decrease in the repulsive energy barrier (Hahn and O’Melia, 2004; Hahn et al., 2004; Shen et al., 2007, 2017; Torkzaban and Bradford, 2016). This is because the Derjaguin approximation approach only considers the screening effect of indifferent ions on the electric field between interacting surfaces (Lin and Wiesner, 2012). Increasing solution IS decreases the range of double layer interaction and accordingly the repulsive energy barrier. The SEI technique considers both screening effect and the osmotic pressure between interacting surfaces. Increasing IS increases osmotic pressure and thus the repulsive energy barrier, and a net increase of the energy barrier with increasing IS will occur if the enlargement of the energy barrier by the osmotic pressure dominates over the energy barrier reduction by the screening effect. Such a net increase in the energy barrier with increasing IS occurs for spherical nanoparticles and sheet-shaped colloids.

It should be noted that the results in Supplemental Fig. S3 were obtained for the interaction of a GONP with a planar sand surface. If surface roughness is considered, the interaction energy barrier decreases with increasing solution IS for the GONPs. For example, Fig. 3 shows interaction energy profiles between a GONP and a CW surface with only one arch of different heights \((D)\) and widths \((L = 2\pi N)\) in KCl at different ISs. The obtained maximum energy barriers, primary minimum depths, and secondary minimum depths are presented in Supplemental Table S3. The energy barrier decreases with increasing IS for \( D = 10 \) and 20 nm. Consequently, more colloids can be attached in primary minima at higher IS in these cases, which explains the aforementioned experimental observations. The energy barrier increases with increasing IS for \( D = 100 \) nm or the number of arches with \( D = 20 \) nm on the CW surface (Supplemental Table S3). These results indicate that the variation of interaction energy barrier with IS is dependent on both height and number of arches. Figure 3 also shows that decreasing the value of \( L \) (i.e., sharper arch) reduces the energy barrier and increases attachment in primary minima. Similarly, Shen et al. (2015) represented rough surfaces as planar surfaces covered by a hemispheroid and showed that the hemispheroid with a large ratio of height to equatorial radius (i.e., sharp asperity) is more effective to reduce energy barriers and more favorable for attachment in primary minima. The presence of surface charge heterogeneity on the arches can further decrease the interaction energy barrier.
and increase attachment in primary minima (Santore and Kozlova, 2007; Pazmino et al., 2014; Bendersky et al., 2015). In addition, the aggregation of GONPs can also increase the attachment at 50 mM KCl and 1 mM CaCl₂ because aggregated GONPs have larger values of $h_0$. Consequently, more aggregated GONPs can transport from bulk solution to the vicinity of the collector surface and then attach.

Figure 2 shows that the deposition was greater in CaCl₂ than in KCl for a given IS in Phase A because Ca²⁺ is more efficient to reduce the electrostatic repulsion between the GONP and sand surfaces by screening the negative charges on the GONP and sand surfaces (Haque et al., 2017). Moreover, Ca²⁺ can increase the attachment by linking the GONPs and sand by cation bridging (Schijven and Hassanizadeh, 2000). For a given chemical condition, the deposition of GONPs decreased with IFV. Equation [1] shows that the decrease of GONP deposition (i.e., the increase of $C/C_0$) is due to reduction of $\beta h_0$. As mentioned previously, IFV decreases the value of $h_0$. To understand whether the decrease

![Breakthrough curves for transport of graphene oxide nanoparticles (GONPs) in sand in KCl and CaCl₂ at different solution ionic strength with different flow velocities. Phase A was the injection of 10 pore volumes (PVs) of GONP suspension at 5 mg L⁻¹. Phase B was the injection of GONP-free electrolyte solution. Phase RIS (reducing solution ionic strength) was the injection of deionized water. The y-axis shows the normalized effluent GONP concentration ($C/C_0$, where $C$ and $C_0$ are effluent and influent concentrations of GONPs, respectively).](image)
of GONPs deposition is also due to reduction of $\alpha$, the values of $\alpha_{\text{exp}}$ at different ISs and flow velocities were calculated using Eq. [2] and are presented in Fig. 4. Surprisingly, the calculated value of $\alpha_{\text{exp}}$ increased with IFV for the GONPs, revealing that the decrease of deposition with IFV is solely due to the reduction of $h_0$. The result also illustrates that attachment in secondary minima should not play a significant role for the GONPs because colloids attached at secondary minima are readily swept away by hydrodynamic shear (Li et al., 2005; Torkzaban et al., 2007; Shen et al., 2010), causing a decrease of $\alpha$ with IFV. Note that we did not calculate $\alpha_{\text{exp}}$ at 50 mM KCl and 1 mM CaCl$_2$ because aggregation occurred under these chemical conditions and the CFT does not consider aggregation.

Previous studies (Torkzaban et al., 2007; Shen et al., 2014; Rasmuson et al., 2017) showed that the value of $\alpha_{\text{exp}}$ decreased with IFV for spherical particles because the presence of NPAs can significantly reduce the depths of primary minima and, accordingly, the adhesive torques. Colloids cannot be attached on the NPAs where the adhesive torques are exceeded by the hydrodynamic torques. There are more unfavorable sites at higher flow velocity, resulting in a decrease of $\alpha$ with IFV. Our theoretical calculations also show that the primary minimum depths between...
the GONPs and the CW surfaces are significantly smaller than those between the GONPs and the planar surface (Supplemental Table S3). The reduction of primary minimum depth is more significant for larger $D$ and smaller $L$. Specifically, the primary minimum depth reaches its minimum ($0.6kT$, where $k$ is the Boltzmann constant and $T$ is absolute temperature) for the arch of $D = 100$ nm and $L = 4\pi$ nm at 10 mM. However, even if the GONPs interact with only two arches of $D = 100$ nm and $L = 4\pi$ nm, the calculated adhesive torque ($5.12 \times 10^{-19}$ N m) is still orders of magnitude larger than the maximum hydrodynamic torque ($5.76 \times 10^{-23}$ N m) in sand porous media. Therefore, IFV cannot reduce the value of $\alpha$ for the sheet-shaped GONPs. The increase of $\alpha_{\text{exp}}$ with IFV is probably because both convex asperities and concave valleys (e.g., areas between arches) are present for a rough surface, and IFV may enhance the GONPs approaching the concave valleys (low flow regions) and then cause more deposition.

**Release of Graphene Oxide Nanoparticles**

In Phase B of Fig. 2, the unattached GONPs in the pore water of the sand porous media were displaced by introducing colloid-free electrolyte solution. In Phase RIS, detachment of GONPs occurred when DI water was introduced to reduce the IS, as denoted by the peaks in the BTCs. Particularly, the value of $\alpha$ of the GONPs initially attached in 50 mM KCl at 9.45 $\times$ $10^{-5}$ m s$^{-1}$. The theoretical calculations in Supplemental Table S3 show that if the sand surface is planar, the GONPs cannot be detached by RIS due to the large primary minimum depths (or detachment energy barrier from primary minima) at all ISs considered. In contrast, the presence of nanoscale arches on the surfaces can significantly reduce primary minimum depth and result in a decrease of primary minimum depth with increasing IS. The detachment of GONPs will occur if the primary minimum depth is reduced enough (e.g., <5$kT$) for the GONPs to escape by Brownian diffusion (Bradford et al., 2017). Such detachment occurs mainly on the arches with large values of $D$ and small values $L$. Therefore, the long and sharp asperities facilitate both attachment and subsequent detachment of GONPs. The facilitated attachment and detachment by NPAs have also been observed for spherical colloids (Fang et al., 2014; Shen et al., 2015). The detachment of GONPs was generally greater in Phase RIS at lower flow velocity because more GONPs were attached in Phase A. However, more detachment occurred in Phase RIS at higher flow velocity in 1 mM CaCl$_2$. This is probably because IFV did not cause a significant decrease in attachment in Phase A under this chemical condition, and the aggregated GONPs were smaller at higher flow velocity and were more readily to be detached on IS reduction.

It is worthwhile mentioning that the use of RIS did not release all attached GONPs in Phase A at all ISs considered (Supplemental Table S4). Although attachment on the planar sand surface or on arches with large values of $L$ is chemically irreversible due to large primary minimum depths, the GONPs cannot overcome the large energy barriers and attach at the primary minima in these cases. The irreversibly attached GONPs were probably those retained on charge heterogeneities and/or on the concave surfaces. Shen et al. (2013) showed that the presence of surface charge heterogeneities can eliminate the repulsive energy barrier and increase primary minimum depth, causing chemically irreversible attachment in primary minima. Li et al. (2017) demonstrated that chemically irreversible attachment in primary minima can also occur at concave regions where the repulsive energy barrier is reduced or even disappears and the primary minimum depth increases with decreasing IS. Colloids attached on these sites are much harder to detach by IFV than those on arches. Indeed, by conducting column experiments where an additional phase (i.e., Phase IFV) was used after phase RIS of Fig. 2, we did not observe the release of GONPs (Supplemental Fig. S5). In fact, no release of GONPs occurred even if Phase IFV was conducted after Phase B (Fig. 5), implying that even the GONPs attached on NPAs cannot be released by IFV. This is because the hydrodynamic torques that act on the GONPs is small compared with the adhesive torques, as was discussed above. The results also support the aforementioned observation that the value of $\alpha$ did not decrease with IFV. Interestingly, Fig. 5 illustrates that when Phase RIS was conducted after Phase IFV, considerable GONPs were detached (i.e., due to
detachment from nanoscale protruding asperities, as mentioned above). The detachments were similar to those in Fig. 2, indicating that adding Phase IFV between Phase B and Phase RIS had a minor influence on the detachment of GONPs by IFV.

Conclusions

It has been recognized in the literature that IFV decreases attachment efficiency for spherical colloids because colloids attached at secondary minima and atop of NPAs via primary minimum association are readily influenced by hydrodynamic shear. Our column experiments, however, showed that the attachment efficiency increased with IFV for the sheet-shaped GONPs. By modeling the roughness of sand surfaces as CWs, our theoretical calculations showed that the hydrodynamic torques that act on the GONPs are orders of magnitude smaller than the adhesive torques for the GONPs, even on nanoscale protruding asperities where adhesion is significantly reduced. Accordingly, IFV cannot decrease the value of attachment efficiency for GONPs. Additional column experiments verify that deposited GONPs cannot be detached by IFV. The increased attachment efficiency with IFV was attributed to the enhanced approaching and subsequent deposition of the GONPs at concave surfaces. These results indicate that IFV is not an effect method for detachment of sheet-shaped colloids, which is desired in a variety of engineered applications such as cleaning of metal surfaces and regeneration of deep bed filters.

Supplemental Material

The supplemental material includes the method used to prepare GONP suspensions, expressions used for calculating van der Waals attraction, electrostatic double layer, and Born repulsion differential interaction energies, zeta potentials of GONPs and sand and average hydrodynamic diameters, calculated primary minimum depth, maximum energy barrier and secondary minimum depth between a GONP and a planar surface or CW surface, fraction of GONPs recovered from Phase A and Phase B, phase RIS, total recovered GONPs, and irreversible attached GONPs for column experiments in Fig. 2, transmission electron microscopy (TEM) image of a GONP, scanning electron microscopy (SEM) image of a sand surface, DLVO energy profiles for a GONP interacting with a planar sand surface or CW surface, and BTCs for transport of GONPs in sand in KCl and CaCl₂ at different ISs.

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References


