

## Supporting Information

### Interaction Between Graphene Oxide Nanoparticles and Quartz Sand

by

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## DLVO theory

**Sphere-plate.** For the case of two approaching surfaces, one with spherical and the other with planar geometries (sphere-plate), the  $\Phi_{vdW}$  [J] interactions were calculated with the following expression:<sup>1</sup>

$$\Phi_{vdW}(h) = -\frac{A_{123}r_p}{6h} \left[ 1 + \left( \frac{14h}{\lambda} \right) \right]^{-1} \quad (SI1)$$

where  $A_{123}$  [J] is the combined Hamaker constant for microscopic bodies of composition “1” and “3” in medium “2” [(1-GO particle)-(2-water)-(3-collector)],  $\lambda \approx 10^{-7}$  m is the characteristic wavelength of the sphere-plate or sphere-sphere interactions, and  $r_p$  [m] is the GO particle radius. In this study, the combined Hamaker constants for the system GO-water-quartz sand was set to  $A_{123}=6.26 \times 10^{-21}$  [J].<sup>2,3</sup> The  $\Phi_{dl}$  for sphere-plate interactions were calculated with the expression:<sup>4</sup>

$$\Phi_{dl}(h) = \pi \varepsilon_r \varepsilon_0 r_p \left[ 2\psi_p \psi_s \ln \left( \frac{1+e^{-\kappa h}}{1-e^{-\kappa h}} \right) + (\psi_p^2 + \psi_s^2) \ln(1-e^{-2\kappa h}) \right] \quad (SI2)$$

where  $\varepsilon_r = \varepsilon/\varepsilon_0$  is the dimensionless relative dielectric constant of the suspending liquid,  $\varepsilon$  [C<sup>2</sup>/(J·m)] is the dielectric constant of the suspending liquid,  $\varepsilon_0$  [C<sup>2</sup>/(J·m)] is the permittivity of free space,  $\psi_p$  [V] is the surface potential of the colloid particle,  $\psi_s$  [V] is the surface potential of the collector surface (plate), and  $\kappa$  [1/m] is the inverse of the diffuse layer thickness, known as the Debye-Huckel parameter:

$$\kappa = \left[ \frac{2000 I_s N_A e^2}{\varepsilon_r \varepsilon_0 k_B T} \right]^{1/2} \quad (SI3)$$

where  $I_s$  [mol/L] is the ionic strength,  $N_A = 6.02 \times 10^{23}$  [1/mol] is Avogadro's number,  $e = 1.602 \times 10^{-19}$  [C] is the elementary charge,  $k_B = 1.38 \times 10^{-23}$  [J/K] is the Boltzmann constant, and  $T$  [K] is the fluid absolute temperature. The  $\Phi_{Born}$  [J] for sphere-plate was estimated by the relationship:<sup>5</sup>

$$\Phi_{Born}(h) = \frac{A_{123} \sigma_{Born}^6}{7560} \left[ \frac{8r_p + h}{(2r_p + h)^7} + \frac{6r_p - h}{h^7} \right] \quad (SI4)$$

where  $\Phi_{Born}$  [m] is the Born collision parameter. For the commonly used value of  $\Phi_{Born} = 0.5$  nm,<sup>5</sup> the resulting acceptable minimum separation distance, at  $h=h_0$ , i.e. at “contact”, is estimated to be  $h_0 = 0.25$  nm, which compares well to  $h_0 = 0.4$  to 1.0 nm estimated by other investigators.<sup>6,7</sup> Note that  $\Phi_{Born}$  can easily be neglected if  $h > 1$  nm. The effect of Born interaction may not be of great significance in aqueous systems since the presence of any hydrated ions, which are likely to be present, will prevent surface-surface separation distances to approach  $h \sim 0.3$  nm.<sup>8</sup>

**Plate-plate.** For the case of two approaching surfaces, both with planar geometries (plate-plate), the  $\Phi_{vdW}$  [J] interactions were calculated with the following expression:<sup>1,9</sup>

$$\Phi_{vdW}(h) = -\frac{A_{123}}{12\pi h^2} \quad (SI5)$$

The  $\Phi_{dl}$  [J] for plate-plate interactions were calculated with the expression:<sup>4</sup>

$$\Phi_{dl}(h) = \frac{\epsilon_r \epsilon_0 K}{2} \left\{ 2\psi_{p1}\psi_{p2} \frac{1}{\sinh(\kappa h)} + (\psi_{p1}^2 + \psi_{p2}^2) [1 - \coth(\kappa h)] \right\} \quad (SI6)$$

where  $\Psi_{p1}$  [V] is the surface potential of the GO nanoparticle, and  $\Psi_{p2}$  [V] is the surface potential of the collector surface. The  $\Phi_{Born}$  [J] for plate–plate interactions were estimated with the relationship:<sup>10</sup>

$$\Phi_{Born}(h) = \frac{A_{123} h_o^6}{45\pi h^9} \quad (SI7)$$

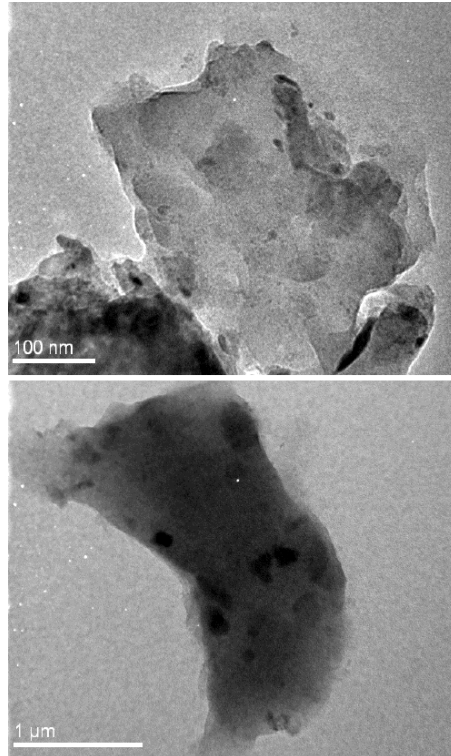
### Pseudo-second-order kinetic model

The analytic expression (5) can be rearranged as follows:

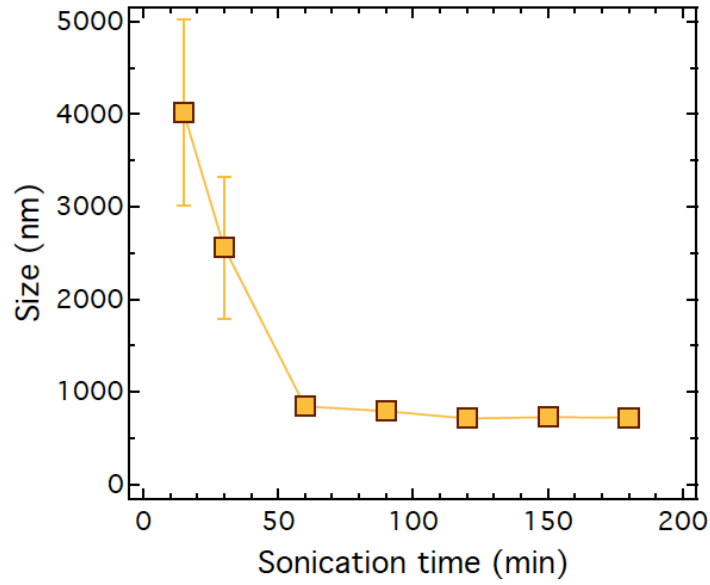
$$C_t^* = \frac{t}{\left[ 1/k_{p2} (C_{eq}^*)^2 \right] + \left[ t/C_{eq}^* \right]} \quad (SI8)$$

or can also be written in the following linear form:

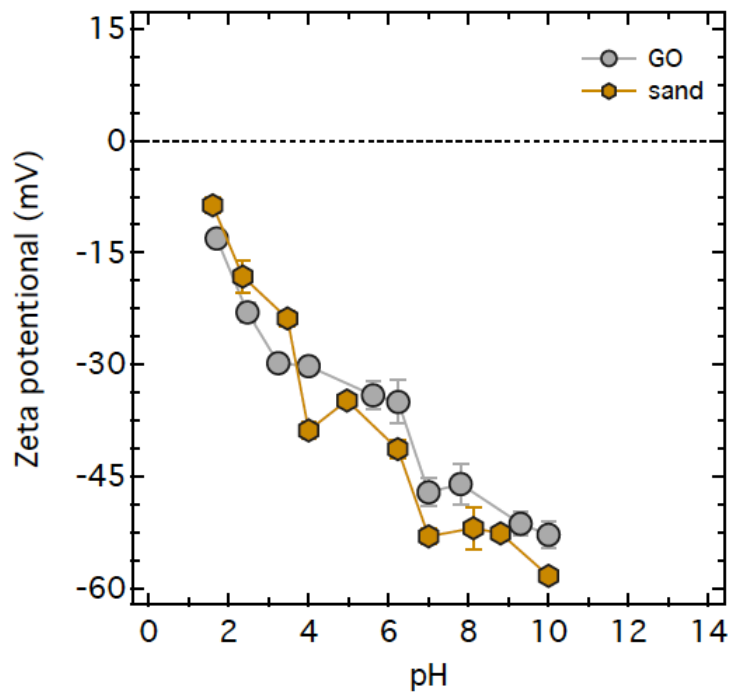
$$\frac{t}{C_t^*} = \frac{1}{k_{p2} (C_{eq}^*)^2} + \frac{t}{C_{eq}^*} \quad (SI9)$$



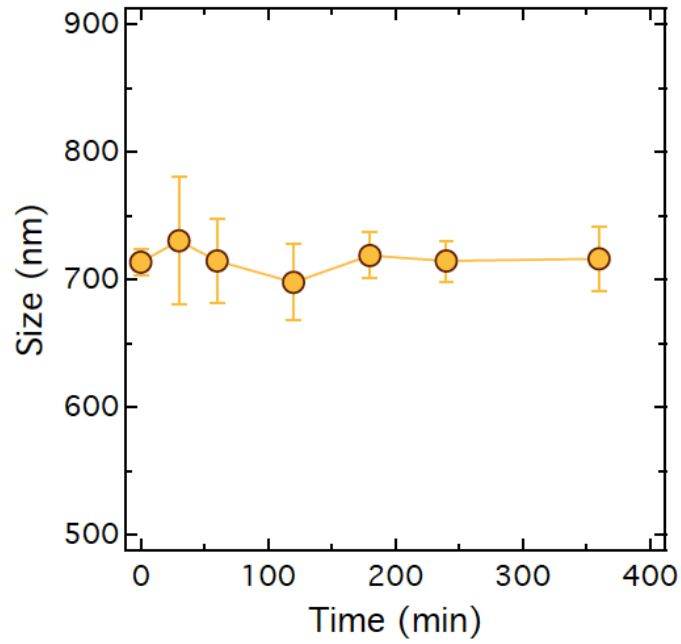
**Figure SI1:** Transmission electron micrograph images of GO flakes.



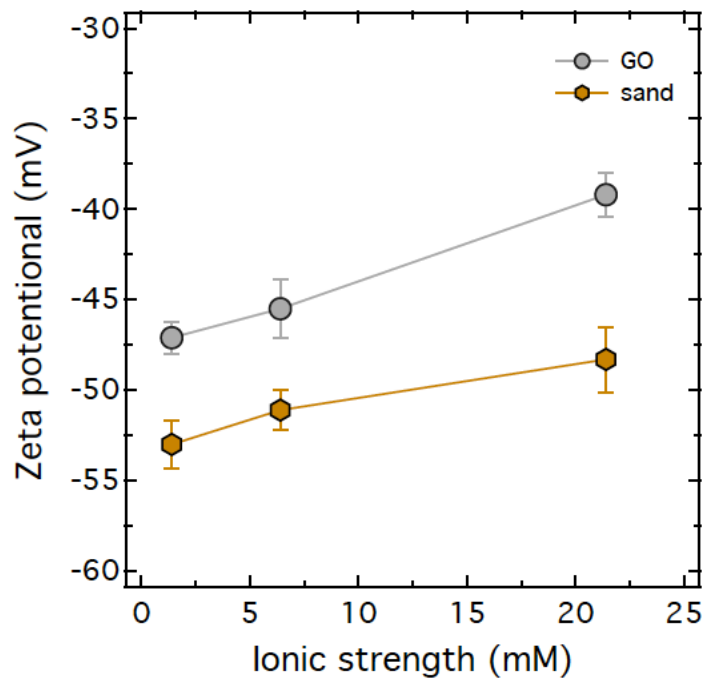
**Figure S12.** Effect of sonication time on the size of GO aggregates (Here pH=7,  $C_{GO}=20$  mg/L,  $I_s= 1.4$  mM).



**Figure S13:** Effect of solution pH on the zeta potential of GO and quartz sand suspensions (Here  $I_s=1.4$ ,  $C_{GO}=20$  mg/L,  $C_{sand}=2$  mg/L).



**Figure S14.** Effect of time on the size of GO aggregates (Here pH=7,  $C_{GO}$ =20 mg/L,  $I_S$ = 1.4 mM).



**Figure S15.** Effect of ionic strength on the zeta potential of GO and quartz sand suspensions (Here pH=7,  $C_{GO}$ =20 mg/L,  $C_{sand}$ =2 mg/L).

**Table S11.** Size of GO aggregates under the different experimental conditions (Here  $C_{GO}=20$  mg/L).

Experimental Conditions			Size (nm)
pH	$I_s$ (mM)	T (°C)	
4	1.4	25	758
7	1.4	25	714
10	1.4	25	716
7	6.4	25	734
7	21.4	25	801

**Table S12.** Experimental conditions of the various kinetic batch experiments conducted in this study.

Experiment	Experimental Conditions		
	pH	$I_s$ (mM)	T (°C)
1	4	1.4	25
2	7	1.4	25
3	10	1.4	25
4	7	6.4	25
5	7	21.4	25
6	7	1.4	12
7	7	1.4	4

**Table S13.** Comparison of the  $R^2$  values of the three isotherm models tested in this work.

$R^2$ of Isotherm Model			
T (°C)	Linear	Langmuir	Freundlich
Static			
4	0.860	0.550	0.976
12	0.814	0.408	0.947
25	0.851	0.449	0.957
Dynamic			
4	0.807	0.557	0.955
12	0.765	0.591	0.934
25	0.795	0.675	0.954

**Table SI4.** Amount of GO attached onto quartz sand at equilibrium in the various kinetic batch experiments conducted for this work.

Experimental Conditions			$C_{eq}^*$ ( $\mu\text{g GO/g sand}$ )
pH	$I_S$ (mM)	T ( $^{\circ}\text{C}$ )	
Static			
4	1.4	25	0.093
7	1.4	25	0.054
10	1.4	25	0.157
7	6.4	25	0.305
7	21.4	25	0.236
7	1.4	4	0.117
7	1.4	12	0.041
Dynamic			
4	1.4	25	1.812
7	1.4	25	0.896
10	1.4	25	0.145
7	6.4	25	2.112
7	21.4	25	3.361
7	1.4	4	0.777
7	1.4	12	0.902

## References

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