Ionic Strength and Composition affect the mobility of surface-modified Fe$^0$ Nanoparticles in water-saturated sand columns.

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DLVO calculation for energy barrier

Symmetric electrolyte case

Using Gouy-Chapman theory, for weak overlap limit, Derjaguin approximation yields electrostatic double layer interaction energy equation for two spherical surfaces.

\[
V_{\text{sphere-sphere}}^{dl} = \frac{128\pi R_1 R_2 k C_+^*}{(R_1 + R_2) \kappa^2} \tanh^2 \left( \frac{ze\phi_0}{4kT} \right) \exp(-\kappa h)
\] (S1)

Here, \( R_1 \) and \( R_2 \) are radius of spheres, \( k \) is the Boltzmann Constant, \( T \) is temperature, \( C_+^* \) is the salt concentration, \( \kappa \) is debye length, \( z \) is valency, \( e \) is electron charge (1.6E-19 amp-s), \( \phi_0 \) is the \( \zeta \) potential, and \( h \) is the sphere-sphere distance (not c/c distance rather edge to edge).

For obtaining particle-collector interaction, sphere-flat plate EDL equation is needed. Equation S1 can be modified by assuming one radius as \( R \) and the tanh term can be separated for sphere and the flat plate (i.e. using \( \zeta \) potential for sphere as \( \phi_0^1 \) and \( \zeta \) potential for flat plate as \( \phi_0^2 \) in the following equation.

\[
V_{\text{sphere-wall}}^{dl} = \frac{128\pi kTC_+^*}{\kappa^2} \tanh \left( \frac{ze\phi_0^1}{4kT} \right) \tanh \left( \frac{ze\phi_0^2}{4kT} \right) \exp(-\kappa h)
\] (S2)

Interaction energy due to van der Waals forces between a sphere and flat plate can be written as

\[
V_{\text{sphere-wall}}^{\text{vdl}} = -\frac{AR}{6h}
\] (S3)

Here, \( A \) is the Hamaker Constant. Because the interaction occurs between a particle and a silica surface having water as the intermediate medium, a combined Hamaker Constant needs to be calculated as follows.

\[
A_{132} = (A_{11}^{1/2} - A_{13}^{1/2})(A_{22}^{1/2} - A_{33}^{1/2})\text{ where } A_{11} = 6E-20J, A_{22} = 8E-21J, A_{33} = 1E-19J \text{ are Hamaker Constants for silica, water and iron respectively.} 
\]
Therefore, the DLVO interaction energy in terms of number of $kT$ can be calculated using the following equation.

$$V_{DLVO_{sphere-wall}} = -\frac{AR}{6h} + \frac{128\pi R kT \zeta^2}{\kappa^2} \tanh \left( \frac{ze\phi_0^1}{4kT} \right) \tanh \left( \frac{ze\phi_0^2}{4kT} \right) \exp(-\kappa h) \quad (S4)$$
Figure S1. Grain size distribution of silica sand used in column experiments. AFS Grain Number is reported as 35 and effective grain size is reported as 0.3 mm by the manufacturer Agsco Corp., Wheeling, IL.

Figure S2. Schematic of transport experiment setup.
Table S1. Equations of fitted calibration curves for all three surface modifiers at each ionic strength.

‘y’ corresponds to concentration and ‘x’ corresponds to UV absorbance.

<table>
<thead>
<tr>
<th>Modifier Type</th>
<th>Calibration Equation: Na⁺</th>
<th>Calibration Equation: Ca²⁺</th>
</tr>
</thead>
<tbody>
<tr>
<td>Triblock copolymer</td>
<td>1mM: y=117.70x+0.48; R²=0.9986</td>
<td>1mM: y=119.97x+3.8; R²=0.9898</td>
</tr>
<tr>
<td></td>
<td>10mM: y=115.68x+0.35; R²=0.9995</td>
<td>5mM: y=116.70x+2.35; R²=0.9928</td>
</tr>
<tr>
<td></td>
<td>100mM: y=100.52x+0.07; R²=0.9956</td>
<td>10mM: y=130.17x+3.2; R²=0.9868</td>
</tr>
<tr>
<td></td>
<td>500mM: y=83.04x+1.96; R²=0.9988</td>
<td>50mM: y=95.76x+3.14; R²=0.9922</td>
</tr>
<tr>
<td></td>
<td>1000mM: y=59.34x+3.46; R²=0.9983</td>
<td></td>
</tr>
<tr>
<td>Polyaaspartate (MRNIP)</td>
<td>1mM: y=160.60x+0.77; R²=0.9991</td>
<td>0.1mM: y=189.48x+3.21; R²=0.9868</td>
</tr>
<tr>
<td></td>
<td>10mM: y=99.98x+1.44; R²=0.9984</td>
<td>0.5mM: y=118.54x+1.41; R²=0.9981</td>
</tr>
<tr>
<td></td>
<td>25mM: y=138.52x+0.87; R²=0.9973</td>
<td>0.75mM: y=106.25x+2.32; R²=0.9998</td>
</tr>
<tr>
<td></td>
<td>40mM: y=97.60x+0.74; R²=0.9999</td>
<td>1mM: y=76.07x+1.74; R²=0.9997</td>
</tr>
<tr>
<td></td>
<td>100mM: y=39.35x+3.7; R²=0.9906</td>
<td></td>
</tr>
<tr>
<td>SDBS</td>
<td>1mM: y=138.35x+0.57; R²=0.9977</td>
<td>0.25mM: y=-139.51x²+125.58x+1.708; R²=0.9857</td>
</tr>
<tr>
<td></td>
<td>10mM: y=110.5x-0.30; R²=0.9938</td>
<td>0.5mM: y=-145.14x²+128.16x+1.888; R²=0.9859</td>
</tr>
<tr>
<td></td>
<td>40mM: y=90.87x+0.80; R²=0.9996</td>
<td>1mM: y=69.722x²+35.044x+1.2479; R²=0.9858</td>
</tr>
<tr>
<td></td>
<td>100mM: y=67.45x+1.31; R²=0.9973</td>
<td></td>
</tr>
</tbody>
</table>
Figure S3a. UV-vis calibration data of triblock copolymer modified particles as function of Na+ concentration. Data obtained at 508nm.

Figure S3b. UV-Vis calibration data of SDBS modified particles as a function of Na+ concentration. Data obtained at 508nm.
**Figure S3c.** UV-Vis calibration data of MRNIP particles as a function of Na$^+$ concentration. Data obtained at 508nm.

**Figure S3d.** UV-Vis calibration data of polymer modified particles as function of Ca$^{2+}$ concentration. Data obtained at 508nm.
Figure S3e. UV-Vis calibration data of SDBS modified particles as a function of Ca2+ concentration. Data obtained at 508nm.

Figure S3f. UV-Vis calibration plot of MRNIP particles as a function of Ca2+ concentration. Data obtained at 508nm.
Figure S4

Figure S4a. Breakthrough curves of triblock copolymer modified RNIP as a function of [Na⁺].

Figure S4b. Breakthrough curves of MRNIP as a function of [Na⁺].
Figure S4c. Breakthrough curves of SDBS modified RNIP as a function of [Na$^+$].
Figure S5a. Breakthrough curves of triblock copolymer modified RNIP as a function of [Ca$^{2+}$].

Figure S5b. Breakthrough curves of MRNIP as a function of [Ca$^{2+}$].
Figure S5c. Breakthrough curves of SDBS modified RNIP as a function of [Ca$^{2+}$].
Figure S6a. Sticking coefficient plot of all the three particles as a function of Na+ concentration.

Figure S6b. Sticking coefficient plot of all the three particles as a function of Ca2+ concentration.