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Hydrogeochemical Preconditioning Improves DNA-based Silica Nanoparticle Mobility (for Tracer Testing) in Carbonate Aquifer.

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DNA-based silica (SiDNA) nanoparticles are increasingly used as groundwater tracers, yet their behavior in fractured (carbonate) aquifers remains poorly characterized, limiting their effective application. To address this gap, we investigated hydrogeochemical controls on SiDNA nanoparticle mobility through a sequence of field experiments and laboratory analyses. An initial dipole tracer test was conducted in a fractured Silurian dolostone aquifer using magnetite-core silica nanoparticles (SiDNAMag). Under near-natural groundwater conditions, the test showed a marked delay in breakthrough and only 1.1% recovery, suggesting strong electrostatic retention at mineral surfaces or physical filtration. To identify the specific geochemical drivers of this retention, batch laboratory experiments and zeta potential measurements were performed. Results showed that dolostone surfaces maintain a negative charge across a wide pH range, with the surface potential becoming increasingly negative (from -11.9 mV to -19.9 mV) as ionic strength decreased from 5.4 mM to 0.9 mM. Ca^{2+} and Mg^{2+} were identified as the primary regulators of surface charge, whereas pH exerted only a secondary influence. Derjaguin–Landau–Verwey–Overbeek (DLVO) modeling revealed that reduced ionic strength enhances the stability and mobility of SiDNAMag. This effect is driven by the expansion of the electrical double layer, which provides a sufficient electrostatic barrier to inhibit attachment to mineral surfaces. Informed by these findings, a second field test incorporated a 48-hour deionized water flush. This preconditioning phase was designed to lower the ambient groundwater ionic strength below 10 mM before injecting the SiDNAMag nanoparticles. This intervention resulted in a 15-fold increase in recovery (17.4%) and faster transport velocities than in the initial test. This study provides a mechanistic framework for "tuning" groundwater chemistry to enhance nanoparticle recovery, thereby significantly improving the viability of colloid-tracer tests in fractured carbonate rock systems.