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Effect of injection velocity and particle concentration on transport of nanoscale zero-valent iron and hydraulic conductivity in saturated porous media

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A B S T R A C T

Successful groundwater remediation by injecting nanoscale zero-valent iron (NZVI) particles requires efficient particle transportation and distribution in the subsurface. This study focused on the influence of injection velocity and particle concentration on the spatial NZVI particle distribution, the deposition processes and on quantifying the induced decrease in hydraulic conductivity (K) as a result of particle retention by lab tests and numerical simulations. Horizontal column tests of 2 m length were performed with initial Darcy injection velocities (q_0) of 0.5, 1.5, and 4.1 m/h and elemental iron input concentrations (Fe^0_{in}) of 0.6, 10, and 17 g/L. Concentrations of Fe^0 in the sand were determined by magnetic susceptibility scans, which provide detailed Fe^0 distribution profiles along the column. NZVI particles were transported farther at higher injection velocity and higher input concentrations. K decreased by one order of magnitude during injection in all experiments, with a stronger decrease after reaching Fe^0 concentrations of about 14–18 g/kg (sand). To simulate the observed nanoparticle transport behavior the existing finite-element code OGS has been successfully extended and parameterized for the investigated experiments using blocking, ripening, and straining as governing deposition processes. Considering parameter relationships deduced from single simulations for each experiment (e.g. deposition rate constants as a function of flow velocity) one mean parameter set has been generated reproducing the observations in an adequate way for most cases of the investigated realistic injection conditions. An assessment of the deposition processes related to clogging effects showed that the percentage of retention due to straining and ripening increased during experimental run time resulting in an ongoing reduction of K. Clogging is mainly evoked by straining which dominates particle deposition at higher flow velocities, while blocking and ripening play a significant role for attachment, mainly at lower injection velocities. Since the injection of fluids at real sites leads to descending flow velocities with increasing radial distance from the injection point, the simulation of particle transport requires accounting for all deposition processes mentioned above. Thus, the derived mean parameter set can be used as a basis for quantitative and predictive simulations of particle distributions and clogging effects at both lab and field scale. Since decreases in K can change the flow system, which may have positive as well as negative implications for the in situ remediation technology at a contaminated site, a reliable simulation is thus of great importance for NZVI injection and prediction.

1. Introduction

Nanoscale zero-valent iron (NZVI) particles have a great potential for in-situ groundwater remediation since they can be

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Nomenclature

A_j	attachment multiplier for interaction site j
a_0	specific surface of the sand $[L^{-1}]$
a_p	specific surface of the particles $[L^{-1}]$
C	species concentration in the mobile phase $[ML^{-3}]$
D	diffusion-dispersion tensor $[L^2T^{-1}]$
d_{50}	median sand grain diameter [L]
Fe^0_{in}	elemental iron input concentrations $[ML^{-3}]$
Fe^0_{tot}	total elemental iron concentration of the mobile and immobile phases $[MM^{-1}]$
$Fe^0_{tot(max)}$	maximum elemental iron concentration $[MM^{-1}]$
K	hydraulic conductivity $[LT^{-1}]$
K_0	initial hydraulic conductivity $[LT^{-1}]$
k_a	attachment rate coefficient $[T^{-1}]$
k_d	detachment rate coefficient $[T^{-1}]$
L	travel distance [L]
n	porosity
n_0	initial porosity
q	Darcy velocity $[LT^{-1}]$
q_0	initial Darcy injection velocities $[LT^{-1}]$
Q	flow rate $[L^3T^{-1}]$
S	species concentration in the solid phase $[MM^{-1}]$
S_{max}	maximum sediment retention capacity $[MM^{-1}]$
t	time [T]
β_j	attachment exponent for interaction site j
ρ_b	bulk density of sand $[ML^{-3}]$
ρ_p	density of the particles $[ML^{-3}]$
Θ	fraction of the surface area of the deposited particle that contributes to an increase of the total matrix surface area
χ	magnetic susceptibility

used to treat a wide range of contaminants, including chlorinated hydrocarbons (Wei et al., 2010; Zhang, 2003). The main advantages are flexible injection e.g. via direct push and the high specific surface area of up to 100 m^2/g (Crane and Scott, 2012), which makes them very reactive compared to granular zero-valent iron. A considerable body of research on NZVI applications has been conducted over the last decade. The results of lab- and field-scale experiments have supported the further development of NZVI technologies, in particular to enhance particle transport and mobility.

The mobility of the particles is generally controlled by stabilization of the particles (e.g. Liang et al., 2014), injection velocity (e.g. Raychoudhury et al., 2010), input particle concentration (e.g. Phenrat et al., 2009), ionic strength (e.g. Li et al., 2006; Saleh et al., 2008) and the pH-value of the groundwater (e.g. Kim et al., 2012). The main adjustable parameters to optimize particle propagation during NZVI injection for remediation purposes are the concentration of NZVI particles in the injected suspension and the injection flow rate. Since the deposition of particles during the transport through the porous medium can lead to hydraulic clogging effects, the effect intensity and quantity dependent on the injection conditions have to be considered. Therefore, an understanding of how these issues influence an NZVI injection is of great importance for site applications.

Some studies focused on the influence of the flow velocity on NZVI particle or colloid transport. In the range of about 0.01 up to about 3 m/h a higher mobility and lower retention of NZVI particles for upper velocity values have been stated by means of particle breakthrough at the column outflow (He et al., 2009; Hosseini and Tosco, 2013; Kanel and Choi, 2007; Phenrat et al., 2010a; Raychoudhury et al., 2010). Additionally, more homogeneous colloid distributions along the column length were found at high velocities (Veerapaneni and Wiesner, 1997). Zhuang et al. (2004) based this effect on the assumption that colloids remain in streamlines at higher velocities, because of a decreased thickness of shear interfaces and increased hydrodynamic forces, which consequently reduces the number of particles approaching the sediment surfaces as a precondition for attachment.

Investigations with different injection concentrations showed, that higher particle concentrations lead to an increasing NZVI retention (e.g. Hosseini and Tosco, 2013; Phenrat et al., 2007; Phenrat et al., 2010a; Phenrat et al., 2010b; Raychoudhury et al., 2010; Saleh et al., 2007). Phenrat et al. (2007, 2010b) concluded that increased aggregation at higher concentration is caused by magnetic forces between the NZVI particles. Bradford and Bettahar (2006) observed that higher colloid concentrations promote clogging of smaller pore throats causing an earlier particle breakthrough at the outflow boundary relative to the injected mass, while lower colloid concentrations lead to an increased mass retardation and deposition near the column inlet.

Some of the studies mentioned above revealed a decrease in hydraulic conductivity (K) of the porous media due to particle deposition. Hosseini and Tosco (2013) reported that nano Fe/