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A new physical model based on cascading column experiments to reproduce the radial flow and transport of micro-iron particles

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A R T I C L E I N F O

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A B S T R A C T

In the field of aquifer restoration and civil engineering, fluids and suspensions are delivered in porous media via well injection. The flow field resulting around the well is three-dimensional. Since two and three-dimensional experiments which can be used to reproduce such flow fields are, although accurate, extremely elaborate and time consuming, the authors suggest to substitute them by a cascade of interdependent, one-dimensional (column) experiments. The new method is used to simulate the injection in aquifers of micro-iron particles dispersed in a shear thinning gel of guar gum and allows the prediction of iron particle distribution around the well and of injection pressure. The method respects the mass balance at the field scale and its predictions are in good agreement with those of a mathematical model proposed in the literature.

The method is consistent with the Triad Approach, a way to improve remediation efficiency proposed by the U.S. Environmental Protection Agency. The representativeness of the proposed tests combined with their simplicity and relative inexpensiveness make this new method applicable and useful in the planning and design of real remediation.

1. Introduction

Zero-valent iron particles in permeable reactive barriers (PRB) can be used to effectively remove groundwater contaminations from an aquifer (Gillham and O'Hannessin, 1994). However, PRB require a high initial investment which increases with depths. Moreover, they cannot be installed underneath buildings. To overcome these obstacles it has been suggested to inject iron particles with dimensions ranging from tens of nanometers to some micrometers (Zhang, 2003). The main advantages of these particles are that they are very reactive due to their large specific surface area, they can be applied directly into a source zone, and they can be injected via wells to remediate the soil under buildings, with little or no reduction in building functionality. Field application of such particles has been documented at more than 100 sites around the word (Comba et al., 2011b; Müller et al., 2011)

To be injected, iron particles are dispersed in a carrier fluid which can be either a gas, an aerosol or a liquid. In this work the fluid phase is a liquid, which together with iron particles forms a slurry or a colloidal suspension. Injection is advantageous with respect to economy and invasivity, provided that iron particles are effectively delivered to the contaminants to ensure proximity to the contaminant, and hence chemical reactions (Illinois Environmental Protection Agency, 2001).

After a successful injection in a homogeneous porous medium, the zone saturated by the slurry has roughly the shape of a cylinder with height *H* depending on the length of the screened well section and on the soil stratification (or bedding) and with radius ROI or radius of influence depending on the injected volume, on soil porosity, and on *H*. Depending on the volume to be treated and site-specific

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parameters, several vertically or horizontally overlapping injections may be necessary. Failure to obtain such an effective distribution of the reagents in the treatment zone results in pockets of untreated contaminants. This will reduce the effectiveness of the remediation. It can also happen that the existence of untreated zones is discovered months after the injection by observing contaminant concentration rebound. This could entail injection repetition with consequent negative effects on costs and site availability. Cases where the outcome did not meet expectations due to a bad distribution of iron particles are reported by Gavaskar and Condit (2005) and by Henn and Waddill (2006).

To effectively distribute iron particles via injection two conditions are to be warranted: (i) the liquid phase has to reach and contact the target area, and (ii) the liquid phase needs to be capable of carrying the solid particles sufficiently long to ensure delivery. The first condition concerns many applications besides the use of iron particles for aquifer remediation: in civil engineering various chemical solutions are delivered into porous media to seal or improve the mechanical resistance of soils; in aquifer restoration mixtures of surfactants and solvents are used to mobilize and recover oil or solvent phase; in reservoir engineering polymeric solutions enhance the recovery of oil from the oil bearing strata. The phenomenon depends both on the soil and on the liquid properties. The second condition concerns only the injection of colloidal suspensions, like iron slurries or cementitious grouts. It is basically a reverse application of the filtration laws since conditions with the least filtration efficiency allow for maximum particle transport. The condition is hard to satisfy when using iron particles, since the high density of iron results in rapid particle sedimentation in porous media (Phenrat et al., 2007; Tiraferri et al., 2008). Also the tendency of iron particles to aggregate in large clusters determines their accumulation in porous media, as it further increases particle sedimentation and as it determines the phenomenon of ripening (Elimelech et al., 1998).

A viscosity increase of the liquid phase is beneficial to satisfy both conditions, since it reduces fingering of the injected suspension and stabilizes the injection front (Martel et al., 1998, 2004: Zhong et al., 2008). It also decreases the aggregation of the iron particles while increasing their sedimentation time (Comba and Sethi, 2009). The viscosity increase can be achieved using special polymers with shear thinning properties such as guar gum or xanthan gum (Cantrell et al., 1997a, 1997b; Comba and Sethi, 2009; Comba et al., 2011a; Dalla Vecchia et al., 2009; Oostrom et al., 2007).

Another major factor controlling colloidal transport is the flow (seepage) velocity of the liquid phase. In order to satisfy continuity, flow velocity decreases hyperbolically with distance from an injection well. According to filtration theory, such a decrease in seepage velocity results in a reduction of particle mobility or an increase of the fraction of iron particles lost from the slurry and deposited in inter-granular voids. When using shear thinning fluids, the velocity decrease around the well has the additional effect of reducing the shear rate, which causes an increase in the apparent viscosity of the liquid phase. This increase in viscosity may retard colloidal particle sedimentation, and thus decrease the fraction of iron particles lost from the slurry and deposited in the porous medium.

In addition, at the field scale, synergistic effects between the abovementioned factors can occur (as all phenomena occur simultaneously at the large scale). Site-specific and secondarily phenomena are also likely to appear (as an example the degradation of guar gum in real injection conditions, with variable injection time from preparation and temperature; Comba and Braun, 2012).

To predict iron distribution around a well, the transport behaviour is usually attributed to a number of physical mechanisms taking place at the microscopic level, and to their interdependencies. Among these mechanisms are sedimentation (Rajagopalan and Tien, 1976; Tufenkji and Elimelech, 2004; Yao et al., 1971), interception (Rajagopalan and Tien, 1976; Tufenkji and Elimelech, 2004; Yao et al., 1971), Brownian diffusion (Rajagopalan and Tien, 1976; Tufenkji and Elimelech, 2004; Yao et al., 1971), surface attachment (Elimelech, 1993), ripening (Darby and Lawler, 1990; Tobiason, 1989; Tobiason and Vigneswaran, 1994), blocking (Hunt et al., 1993; Johnson and Elimelech, 1995; Ko and Elimelech, 2000), straining (McDowell-Boyer et al. 1986, Bradford et al., 2006a, 2006b), and hydrodynamic bridging (Ramachandran and Fogler, 1999). Sedimentation or retention of the particles is also influenced by surface roughness of soil particles (Kretzschmar et al., 1997; Redman et al., 2001), charge heterogeneity (Johnson and Elimelech, 1995), and variability in colloid characteristics (Bolster et al., 1999). To find these dependencies, colloidal transport tests in columns filled with porous medium are performed and constitutive relations are derived therefrom. However, as the number of relevant physical mechanisms increases, it becomes difficult to isolate the effect of each mechanism in the results of column tests.

Another approach that predicts iron micro-particle distribution around a well based on empirical mathematical modeling was adopted by Comba and Braun (2012). For this approach column tests are also performed and interpreted based on the main mechanisms proposed by colloid filtration theory. However, this approach is not based on constitutive relations derived from phenomena occurring at the micro scale, but is an empirical mathematical relationship that links transport behaviour to field variables (such as injection rate or volume of slurry injected).

The large number and the complexity of the phenomena described above suggest that prior to a field application it is necessary to verify the prediction of iron distribution generated by the mathematical models with laboratory experiments. Such experiments should reproduce as closely as possible the real flow situation expected during a field application. Ideally, porous medium extracted from the concerned zone should be used. Also, the guar gum utilized should come from the same supplier and should be prepared as in the field. In this way all phenomena and interactions influencing transport will be taken in account, including the ones not yet well known or unexpected for the specific conditions.

Currently, the only way to reproduce radial flow is to conduct three-dimensional or at least two-dimensional, radially symmetrical, experiments (Chao et al., 2000; Kobus et al., 1995; Müller and Nowack, 2010). Since these physical models are extremely elaborate and time consuming, the authors suggest substituting them by a cascade of interdependent, one-dimensional (column) experiments. This cascading column method is applied to the injection of micro-iron particles dispersed in a shear thinning gel of guar gum into aquifers. The method is used to select suitable injection parameters (flow