Journal of Contaminant Hydrology 166 (2014) 34–51

Contents lists available at ScienceDirect

Journal of Contaminant Hydrology

journal homepage: www.elsevier.com/locate/jconhyd

Guar gum solutions for improved delivery of iron particles in porous media (Part 2): Iron transport tests and modeling in radial geometry

Tiziana Tosco, Francesca Gastone, Rajandrea Sethi *

DIATI — Dipartimento di Ingegneria dell'Ambiente, del Territorio e delle Infrastrutture — Politecnico di Torino, C.so Duca degli Abruzzi 24, 10129 Torino, Italy

ARTICLE INFO

Article history:
Received 11 February 2014
Received in revised form 26 June 2014
Accepted 27 June 2014
Available online 6 July 2014

Keywords:
Microscale iron transport in porous media
Guar gum solutions
Porous medium flow and transport in radial domains
Column transport tests
Porous medium clogging

ABSTRACT

In the present work column transport tests were performed in order to study the mobility of guar-gum suspensions of microscale zero-valent iron particles (MZVI) in porous media. The results were analyzed with the purpose of implementing a radial model for the design of full scale interventions. The transport tests were performed using several concentrations of shear thinning guar gum solutions as stabilizer (1.5, 3 and 4 g/l) and applying different flow rates (Darcy velocity in the range $1 \cdot 10^{-4}$ to $2 \cdot 10^{-3}$ m/s), representative of different distances from the injection point in the radial domain. Empirical relationships, expressing the dependence of the deposition and release parameters on the flow velocity, were derived by inverse fitting of the column transport tests using a modified version of E-MNM1D (Tosco and Sethi, 2010) and the user interface MNMs (www.polito.it/groundwater/software). They were used to develop a comprehensive transport model of MZVI suspensions in radial coordinates, called E-MNM1R, which takes into account the non Newtonian (shear thinning) rheological properties of the dispersant fluid and the porous medium clogging associated with filtration and sedimentation in the porous medium of both MZVI and guar gum residual undissolved particles. The radial model was run in forward mode to simulate the injection of MZVI dispersed in guar gum in conditions similar to those applied in the column transport tests. In a second stage, we demonstrated how the model can be used as a valid tool for the design and the optimization of a full scale intervention. The simulation results indicated that several concurrent aspects are to be taken into account for the design of a successful delivery of MZVI/guar gum slurries via permeation injection, and a compromise is necessary between maximizing the radius of influence of the injection and minimizing the injection pressure, to guarantee a sufficiently homogeneous distribution of the particles around the injection point and to prevent preferential flow paths.

1. Introduction

Nanoscale and microscale iron particles (NZVI and MZVI) show a positive potential for the remediation of contaminated aquifers, in both laboratory and field-scale tests, in particular for the treatment of organic compounds, including widely spread chlorinated hydrocarbons (He and Zhao, 2005; Lien and Zhang, 2001; Liu et al., 2005; Zhang, 2003), pesticides (Elliott et al., 2003; Joo et al., 2004), polycyclic aromatic hydrocarbons (PAHs) (Chang et al., 2005), poly-chloro-biphenyls PCBs (Hadnagy et al., 2004; Lowry and Johnson, 2004), and azo-dyes (Freyria et al., 2011). Compared to other remediation techniques based on the use of millimetric zerovalent iron (namely permeable reactive barriers, PRBs) (Di Molfetta and Sethi, 2006; Zolla et al., 2009), MZVI and NZVI exhibit beneficial

* Corresponding author. Tel.: +39 011 564 7735.
E-mail address: rajandrea.sethi@polito.it (R. Sethi).

http://dx.doi.org/10.1016/j.jconhyd.2014.06.014

peculiarities. In particular, they are characterized by a high specific surface area, orders of magnitude higher than millimetric iron, which results in high degradation rates (Nurmi et al., 2005; Sun et al., 2006). Moreover, they can be injected into the contaminated area dispersed in water-based slurries, thus resulting in a limited occupation of the surface and in a higher flexibility of the technique, compared to other remediation approaches, e.g. permeable reactive barriers (Di Molfetta and Sethi, 2006; Elliott and Zhang, 2003). Finally, it is possible to modify the properties of the particles and of the dispersant fluid, for a tuned delivery and reactivity (Berge and Ramsburg, 2009; Dalla Vecchia et al., 2009b; Quinn et al., 2005; Tiraferri and Sethi, 2009).

Despite the positive characteristics of MZVI and NZVI, some critical aspects are to be addressed before the technology can be applied at the field scale as an established technique. A major issue is associated to the colloidal instability of aqueous dispersions of MZVI and NZVI (Kocur et al., 2013; O'Carroll et al., 2013; Tiraferri and Sethi, 2009; Tosco et al., 2014). Colloidal suspensions of iron particles in the typical concentrations required for field injections (in the order of several grams per liter) are not stable, being NZVI prone to aggregation and consequent sedimentation of the aggregates, and MZVI to sedimentation of the primary particles, with extremely negative impacts on the overall mobility in porous media in both cases (Dalla Vecchia et al., 2009a, 2009b; Saleh et al., 2007; Tiraferri and Sethi, 2009). The lack of stability can be successfully overcome by modifying the surface properties of the particles (approach effective for NZVI) or increasing the viscosity of the dispersant fluid (for MZVI) (Comba et al., 2011; Dalla Vecchia et al., 2009b; Fatisson et al., 2010; Oostrom et al., 2007; Saleh et al., 2007; Tiraferri and Sethi, 2009). In particular, the use of shear thinning solutions of biopolymers was found successful in improving colloidal stability, and consequently mobility, of both MZVI and NZVI in porous media. Among these, visco-elastic fluids (e.g. guar gum–xanthan mixtures) provide an almost indefinitely stable suspension (Xue and Sethi, 2012), while shear thinning solutions without yield stress (e.g. single polymer solutions of guar gum, xanthan, or carboxymethyl cellulose) form suspensions defined as meta-stable, in which the increased viscosity of the polymeric solution reduces the sedimentation rate of the particles (Dalla Vecchia et al., 2009b; Johnson et al., 2013; Kocur et al., 2013; Velimirovic et al., 2012).

The second critical issue to be addressed is related to the mobility of the particles in the porous medium. The suspensions of MZVI and NZVI can be delivered into the subsurface under pressure, resulting in the formation of preferential flow paths and porous medium fracturing (when the injection pressure overcomes the critical one of the porous medium) or permeation injection, which provides a less heterogeneous distribution of the slurry in the porous medium. The comprehension of the transport mechanisms and the development of a transport model are crucial for field applications. It is necessary to provide an estimation of the expected travel distance and iron distribution, and to identify whether or not the particles will be mobile under the natural flow field. If fracturing delivery is used, the overall distribution of the particles in the subsurface is highly heterogeneous. In this case the estimation of the iron distribution is not based on any transport model, and can be approximated with empirical relationships, since position, extent and width of the fractures in a field application cannot be predicted by numerical modeling (Abdel-Salam and Chrysikopoulos, 1995). Conversely, for permeation injection, modeling approaches based on modified advection–dispersion equations can be adopted (Kocur et al., 2013; Tosco and Sethi, 2010). Great efforts have been devoted so far to the identification of the mechanisms controlling the transport of NZVI (namely physical–chemical interactions with the porous medium, magnetic attraction among particles, filtration and straining of aggregates, sedimentation, etc), and several approaches have been proposed to model its transport and retention in porous media (Kanel et al., 2007; O'Carroll et al., 2013; Petosa et al., 2010; Tiraferri and Sethi, 2009; Torkzaban et al., 2012; Tosco and Sethi, 2010; Yan et al., 2013). On the other hand, fewer studies focused on the mobility of microscale iron (Dalla Vecchia et al., 2009b; Tosco and Sethi, 2010; Velimirovic et al., 2014). In this case, the shear thinning properties of the dispersant fluid, along with the colloidal stability or instability of the suspension, play a major role. The rheological characterization of the MZVI slurries and particle sedimentation was investigated in details by the authors in a previous work (Gastone et al., 2014).

In this work, a study on guar gum-based slurries of MZVI particles is presented. The work is composed by two papers. In Part I, the flow of guar gum solutions in porous media was assessed, evidencing the effects of the shear thinning rheological properties on the pressure build up, and the porous medium clogging caused during injection by residual undissolved particles of guar gum. A model was derived from experimental column filtration tests for the quantification of such processes. In the present Part II, a modeling approach based on the results of laboratory column transport tests is proposed for the simulation of large-scale injection of MZVI via permeation. In field applications fluids are typically injected into the subsurface via wells or direct push systems, generating a radial or radial-like flow, with decreasing velocity with increasing distance from the delivery point (Fig. 1). Therefore a simplified radial geometry was assumed here. The development of a transport model in radial geometry requires a detailed knowledge of the influence of flow velocity on the kinetics of the particle-porous medium interactions (deposition and release processes), the eventual clogging, and the viscosity of the shear thinning carrier fluid (Ciriello and Di Federico, 2012; Longo et al., 2013; Wexler et al., 2013). However, injection tests at a laboratory scale, which guarantee the well controlled conditions necessary for the development of a mathematical model, are complex and expensive to be realized in a radial flow field. For this reason, in this work the flow velocity dependence of transport processes was investigated in a set of one-dimensional column transport tests, performed at different flow rates, each representative of the flow velocity expected at a different distance from the delivery point. The one-dimensional transport tests were analyzed using a modified formulation of the transport software E-MNM1D (Tosco and Sethi, 2010), the processes governing MZVI transport were identified, and the dependence of the transport parameters on the flow rate and on the fluid viscosity was derived. The obtained relationships were then implemented in the radial model. The radial transport model was then developed integrating (i) the correlations for the kinetics of MZVI interactions with the porous medium (deposition and release processes) derived from the analysis of column tests, (ii) the results of the companion paper Part I on guar gum flow in porous media (clogging due to undissolved guar gum particles