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Guar gum solutions for improved delivery of iron particles in porous media (Part 1): Porous medium rheology and guar gum-induced clogging

Francesca Gastone, Tiziana Tosco, 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

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ABSTRACT

The present work is the first part of a comprehensive study on the use of guar gum to improve delivery of microscale zero-valent iron particles in contaminated aquifers. Guar gum solutions exhibit peculiar shear thinning properties, with high viscosity in static conditions and lower viscosity in dynamic conditions: this is beneficial both for the storage of MZVI dispersions, and also for the injection in porous media. In the present paper, the processes associated with guar gum injection in porous media are studied performing single-step and multi-step filtration tests in sand-packed columns. The experimental results of single-step tests performed by injecting guar gum solutions prepared at several concentrations and applying different dissolution procedures evidenced that the presence of residual undissolved polymeric particles in the guar gum solution may have a relevant negative impact on the permeability of the porous medium, resulting in evident clogging. The most effective preparation procedure which minimizes the presence of residual particles is dissolution in warm water (60 °C) followed by centrifugation (procedure T60C). The multi-step tests (i.e. injection of guar gum at constant concentration with a step increase of flow velocity), performed at three polymer concentrations (1.5, 3 and 4 g/l) provided information on the rheological properties of guar gum solutions when flowing through a porous medium at variable discharge rates, which mimic the injection in radial geometry. An experimental protocol was defined for the rheological characterization of the fluids in porous media, and empirical relationships were derived for the quantification of rheological properties and clogging with variable injection rate. These relationships will be implemented in the second companion paper (Part II) in a radial transport model for the simulation of large-scale injection of MZVI-guar gum slurries.

1. Introduction

The injection of water-based slurries of nanoscale and microscale iron particles (NZVI and MZVI) represents a promising in situ remediation technology for contaminated aquifer systems (O'Carroll et al., 2013; Tosco et al., 2014; Yan et al., 2013). Micro and nanoscale iron particles proved effective in reducing a variety of metal ions (DeVor et al., 2006; Jegadeesan et al., 2005; Kanel et al., 2005; Ponder et al., 2000) and degrading a broad range of organic pollutants in groundwater systems, in particular chlorinated hydrocarbons and recalcitrant compounds (Chang et al., 2005; Freyria et al., 2011; He and Zhao, 2005; Joo et al., 2004; Lowry and Johnson, 2004; Zhang, 2003). Since NZVI and MZVI dispersions in pure water are not stable, due to fast aggregation and sedimentation, the use of stabilizers is necessary. Amendments to the particle surfaces which increase inter-particle repulsion forces are effective for NZVI, which is prone to attractive interactions, mainly magnetic in nature (Dalla Vecchia et al., 2009a; Hosseini and Tosco, 2013; Hydutsky et al., 2007; Kocur et al., 2013; Saleh et al., 2007; Schrick et al., 2004;

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

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Tiraferri et al., 2008), while modifications of the dispersant fluid is mainly applied for MZVI, which is subject to fast sedimentation (Cantrell et al., 1997; Dalla Vecchia et al., 2009b; Oostrom et al., 2007; Xue and Sethi, 2012). In particular, the increased viscosity of the dispersant fluid resulted in a significantly improved colloidal stability, when "green" biopolymers, like xanthan gum, starch, guar gum and carboxymethyl-cellulose, were used (Comba et al., 2011; Kocur et al., 2013; Krol et al., 2013; Xue and Sethi, 2012). These polymers, dispersed in water, form non-Newtonian, shear thinning solutions, characterized by a non constant viscosity: in static conditions the viscosity is higher, which is beneficial for storage of the solutions, and in dynamic conditions it is lower, which helps in limiting the pressure buildup during injection in the subsurface (Gastone et al., 2014; Xue and Sethi, 2012).

Iron particles are typically delivered to the contaminated area via gravity in wells and piezometers (Elliott and Zhang, 2001; Elliott and Zhang, 2003), or more often via pressure injection applying direct push techniques (Quinn et al., 2005; Mueller et al., 2012; Varadhi et al., 2005; Velimirovic et al., 2014) or using injection wells (Henn and Waddill, 2006). Pressure injection can be performed by applying a high injection pressure, thus resulting in the formation of preferential flow paths and porous medium fracturing, or injecting the slurries at low pressure (permeation injection), generating a fairly homogeneous flow through the soil pores (Christiansen et al., 2010; Suthersan, 1999; Tosco et al., 2014). The fracturing approach is used in low to medium permeability formations and when injecting particles whose dimension is in the order of the pore size. Conversely, permeation delivery is preferred in highly permeable formations and, more generally, when particles are small compared to pore size. The threshold between permeation and fracturing injection is related to the critical pressure, that is, the pore pressure which overcomes the porous medium lithostatic load and thus gives rise to preferential flow paths. If permeation is identified as the target delivery method, it is extremely important to limit the pressure buildup as much as possible, in order to prevent the possible formation of undesired preferential flow paths (Tosco et al., 2014).

The present work is the first part of a comprehensive study, composed by two papers, on the use of guar gum solutions as dispersant fluids for MZVI delivery via permeation. Guar gum was selected due to its efficacy in improving the colloidal stability of MZVI particles, its environmental compatibility (it is usually adopted in the food industry as a thickening agent), limited cost, and easy degradation in subsurface conditions, especially in the presence of specific enzymes, which can be used to promote the polymer breakage and degradation (Burke and Khan, 2000; Cheng and Prud'homme, 2000; Di Molfetta and Sethi, 2006; Gastone et al., 2014; Velimirovic et al., 2012; Zolla et al., 2009). In the present Part I, the processes associated with the injection of guar gum solutions in porous media via permeation, as well as approaches and methods for the quantitative analysis and prediction of such processes, are investigated. In Part II, the injection of guar gum-stabilized MZVI in porous media will be studied, and a model for the simulation of field injection of MZVI slurries will be developed and discussed.

When studying the permeation of guar gum in porous media, it is important to predict the pressure buildup during injection. However, the application of Darcy's law (Darcy, 1856) to guar gum slurries is not straightforward, since neither fluid viscosity nor permeability is a constant parameter: the viscosity is affected by the flow velocity, due to the shear thinning nature of guar gum solutions, and the presence of residual undissolved polymeric particles results in a progressive decrease in permeability during injection (clogging). In this paper, experimental tests and a modeling approach are presented for the prediction of the pressure buildup via a modified Darcy's law, which takes into account variable viscosity and permeability. An experimental approach, based on step injection tests, is proposed for the characterization of the rheological properties of guar gum solutions in porous media. Provided that the rheological behavior of the guar gum solutions in the bulk is known (eg. from classical rheological measurements), applying the method herein presented the viscosity of guar gum solutions in the porous medium can be fully characterized. The reduction in permeability, associated with the presence of undissolved polymeric residuals, is considered as well. It is known that the presence of residual particles can be decreased by selecting a proper dissolution procedure (eg. dissolving guar gum powder at a temperature in the range 40 °C–70 °C), but cannot be completely avoided (Casas et al., 2000; Chaudemanche and Budtova, 2008; Chauveteau and Kohler, 1984; Gastone et al., 2014). Their presence was shown irrelevant for the overall colloidal stability of the suspension (Gastone et al., 2014), and it has no negative impact on the injectability of guar gum-based slurries via fracturing. Conversely, when delivery via permeation is desired, the undissolved guar gum particles can clog the porous medium. Moreover, when guar gum solutions are used as delivery fluids for MZVI injection, clogging due to undissolved guar gum produced effects similar to clogging due to the retention of iron particles and aggregates (which will be investigated in detail in Part II), both contributing to the overall reduction in porosity and permeability. The quantification of the maximum acceptable pressure buildup is not trivial, since the critical pressure for a given porous medium is related to a number of parameters and conditions, discussion of which is beyond the purpose of this work. A control of the overall pressure buildup is therefore necessary to prevent fracturing. To this aim, the retention of guar gum particles is to be limited as much as possible, to prevent excessive pressure buildup.

In the present paper, clogging due to injection of guar gum solutions is investigated in laboratory column tests (filtration tests), exploring the influence of the guar gum dissolution procedure, and the operating conditions which can minimize clogging are identified. An approach for the modeling of the process is proposed, based on the software E-MNM1D (Tosco and Sethi, 2010), previously developed by the authors. The model is validated against the experimental data of column filtration tests, and simple empirical relationships linking permeability reduction, polymer concentration and injection rate are derived, which will be used in Part II in the MZVI transport model.

2. Background

2.1. Guar gum flow in porous media

The flow of polymeric shear thinning fluids in porous media has been investigated mainly for applications in oil engineering, where these fluids are used in enhanced oil recovery techniques (Muskat and Wyckoff, 1937; Zeinijahromi et al., 2012; Littmann,