



Improving the sustainability of granular iron/pumice systems for water treatment

Stefania Bilardi^a, Paolo S. Calabrò^a, Sabine Caré^b, Nicola Moraci^a, Chicgoua Noubactep^{c,d,*}

^a *Università degli Studi Mediterranea di Reggio Calabria, MECMAT, Mechanics and Materials Department, Faculty of Engineering, Via Graziella, Loc. Feo di Vito, 89122 Reggio Calabria, Italy*

^b *Université Paris-Est, Laboratoire Navier (UMR 8205), CNRS, ENPC, IFSTTAR, F-77455 Marne-la-Vallée, France*

^c *Angewandte Geologie, Universität Göttingen, Goldschmidtstraße 3, D-37077 Göttingen, Germany*

^d *Kultur und Nachhaltige Entwicklung CDD e.V., Postfach 1502, D-37005 Göttingen, Germany*

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ABSTRACT

Metallic iron (Fe^0) is currently used in subsurface and above-ground water filtration systems on a pragmatic basis. Recent theoretical studies have indicated that, to be sustainable, such systems should not contain more than 60% Fe^0 (vol/vol). The prediction was already validated in a Fe^0 /sand system using methylene blue as an operational tracer. The present work is the first attempt to experimentally verify the new concept using pumice particles. A well-characterized pumice sample is used as operational supporting material and is mixed with 200 g of a granular Fe^0 , in volumetric proportions, varying from 0 to 100%. The resulting column systems are characterized (i) by the time dependent evolution of their hydraulic conductivity and (ii) for their efficiency for the removal of Cu^{II} , Ni^{II} , and Zn^{II} from a three-contaminants-solution (about 0.3 mM of each metal). Test results showed a clear sustainability of the long term hydraulic conductivity with decreasing Fe^0 /pumice ratio. In fact, the pure Fe^0 system clogged after 17 days, while the 25% Fe^0 system could operate for 36 days. The experimental data confirmed the view that well-designed Fe^0 PRBs may be successful at removing both reducible and non-reducible metal species.

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1. Introduction

Filter materials for water treatment are ideally used in small quantities. The high required affinity of used aggregates for efficient water treatment is not always readily available in natural materials. On the other hand, efficient filters should be designed to make the best use of these latter with the minimum of processing (Smith et al., 2001). Alternatively, readily available natural materials (e.g. anthracite, gravel, pumice, sand) may be mixed to low cost synthetic aggregates/materials (activated carbon, blast furnace slag, metallic iron) for improving the performance of the resulting water treatment systems. The key properties determining the permeability, the stability and the longevity (sustainability) of granular filters include porosity/texture of used particles, particle size, particle shape and particle size distribution or material sorting (Haarhoff and Vessal, 2010; Kubare and Haarhoff, 2010; Miyajima, 2012; Btatkeu et al., 2013; Caré et al., 2013). Two key interrelated

properties required for a sustainable filter include: (i) high permeability combined with resistance to internal erosion of fines and (ii) low susceptibility to chemical attack (prerequisite p).

Granular metallic iron (Fe^0), as currently used in water treatment, is a reactive material and its oxidative dissolution by water is a volumetric expansive process (Pilling and Bedworth, 1923; Caré et al., 2008). This means that Fe^0 is highly susceptible to chemical attack and the products of this chemical reaction are fines/precipitates (iron hydroxides and oxides). In other words, 'prerequisite p' is not satisfied as the sustainability of Fe^0 filters is impaired by the same properties making Fe^0 an attractive material: the chemical reactivity of iron (Liu et al., 2013). However, without considering these key properties, Fe^0 permeable reactive barriers (Fe^0 PRBs) have become an established technology for the treatment of contaminated groundwater (O'Hannesin and Gillham, 1998; Li et al., 2006; Bartzas and Komnitsas, 2010; Li and Benson, 2010; Comba et al., 2011; Gheju, 2011; Giles et al., 2011; Hashim et al., 2011; Ruhl et al., 2012). Currently, about 180 Fe^0 PRBs have been installed worldwide (ITRC, 2011).

The fundamental mechanisms of contaminant removal in Fe^0 filtration systems are adsorption, co-precipitation and adsorptive size-exclusion (Noubactep, 2008, 2010, 2011). Contaminant removal also implies iron corrosion (Lavine et al., 2001; You et al.,

* Corresponding author. Angewandte Geologie, Universität Göttingen, Goldschmidtstraße 3, D-37077 Göttingen, Germany. Tel.: +49 551 39 3191; fax: +49 551 39 9379.

E-mail address: cnoubac@gwdg.de (C. Noubactep).

2005; Jiao et al., 2009; Ghauch et al., 2011; Gheju and Balcu, 2011). Therefore, due to the volumetric expansive nature of this process ('prerequisite p'), the remediation of contaminated groundwater necessarily results in the gradual clogging of the Fe⁰ PRB, and thus in the deterioration of the permeable barrier hydraulic conductivity (permeability loss) over time (Zhang and Gillham, 2005; Courcelles et al., 2011; Knowles et al., 2011; Jeen et al., 2012; Miyajima, 2012; Noubactep, 2013a, 2013b, 2013c).

The gradual clogging (permeability loss) of Fe⁰ filtration systems has several origins: (i) biological activities like biofilm growth or biocorrosion, (ii) chemical processes like (hydr)oxide or calcite precipitation, (iii) physical processes allowing the retention of fine particles in the PRB pores, and (iv) production and accumulation of gases (mainly H₂). Pores clogging could generate a decrease in treatment performance and the bypass of untreated contaminated groundwater (Rangsvik and Jekel, 2005; Courcelles et al., 2011; Knowles et al., 2011; Jeen et al., 2012). Therefore, PRBs clogging issues will require cost-intensive reactive material substitution, if satisfactory operational performance has to be maintained. The present work is focused on the characterization of PRB clogging due to pore filling by in-situ generated iron corrosion products neglecting the other possible phenomena that could contribute to permeability reduction (i.e. gas retention, biocorrosion, biofouling) (Henderson and Demond, 2011; Caré et al., 2013; Noubactep, 2013a).

The objective of the present work is to characterize the efficiency of Fe⁰/pumice granular mixtures for contaminant removal in column experiments containing 0–100% Fe⁰ (vol/vol). Fe⁰ is admixed to a well-characterized pumice specimen (Moraci and Calabrò, 2010; Calabrò et al., 2012; Bilardi et al., 2013a), in different volumetric ratios. The model oxalic solution (about 8 mg/L O₂) contained about 0.3 mM of Cu^{II}, Ni^{II}, and Zn^{II}. The evolution of the systems is characterized by determining the (i) extent of contaminant removal (or retention), and (ii) variation of hydraulic conductivity.

2. Materials and methods

2.1. Chemicals

Copper(II) nitrate hydrate (purity 99.999), nickel(II) nitrate hexahydrate (purity 99.999) and zinc(II) nitrate hexahydrate (purity 99.000) were obtained from Sigma–Aldrich. The three heavy metals are used for their different affinity to iron oxides (Wang and Qin, 2007; Moreira and Alleoni, 2010; Vodyanitskii, 2010). In addition, a survey of the electrode potential (E^0) of involved couples indicated differential redox behaviours. In fact, Zn^{II} ($E^0 = -0.763$ V) can not be reduced by Fe⁰ ($E^0 = -0.440$ V) while Cu ($E^0 = 0.337$ V) is readily reduced. The electrode potential of Ni ($E^0 = -0.250$ V) is relatively close to that of Fe ($\Delta E^0 = 0.19$ V) such that quantitative reduction can not be expected.

2.2. Solid materials

2.2.1. Pumice

The used pumice originates from Lipari (Aeolian Islands, Sicily – Italy); its mineralogical composition was determined as follows: SiO₂: 71.75%; Al₂O₃: 12.33%; K₂O: 4.47%; Na₂O: 3.59%; Fe₂O₃: 1.98%; moreover it contains about 4% of bound water (structural water) and traces of other compounds (e.g. CaO, SO₃, MgO, TiO₂, FeO, MnO, P₂O₅). Although pumice exhibited a non-negligible removal capacity for heavy metals (Moraci and Calabrò, 2010; Calabrò et al., 2012), it was used here as an operational inert material with the virtual capacity of storing corrosion products in its pores and retarding clogging (Moraci and Calabrò, 2010; Noubactep and Caré, 2010; Noubactep et al., 2012a, 2012b). The material is characterized by uniform grain

size distribution. The mean grain size (d_{50}) is about 0.3 mm and the coefficient of uniformity (U) is 1.4 (see Supporting information).

2.2.2. Metallic iron

The used Fe⁰ is of the type FERBLAST RI 850/3.5, distributed by Pometon S.p.A., Mestre – Italy. The material contains mainly iron (>99.74%). Identified impurities included mainly Mn (0.26%), O, S and C. The material is characterized by uniform grain size distribution. The mean grain size (d_{50}) is about 0.5 mm and the coefficient of uniformity (U) is 2 (see Supporting information).

The microstructure of used Fe⁰ and pumice was characterized using Mercury Intrusion Porosimetry (MIP) measurements and by Scanning Electron Microscopy (SEM) observations (see Supporting information).

2.3. Columns experiments

The used solutions were obtained by dissolving copper nitrate, nickel nitrate and zinc nitrate in distilled water. The molar concentration of the resulting solution was as follows: 0.27 mM Cu, 0.29 mM Ni and 0.37 mM Zn. The corresponding mass concentrations are 17 mg/L Cu, 17 mg/L Ni, and 23 mg/L Zn.

No attempt was made to control the mass of dissolved oxygen (DO) present during the column experiments. The main source of molecular oxygen is the air in the headspace of the PE bottles. It can be assumed that the model solutions contained up to 8 mg/L DO. The role of dissolved oxygen in accelerating the kinetics of aqueous iron corrosion is well-documented (e.g. Cohen, 1959; Stratmann and Müller, 1994). Using an oxalic solution is a tool to enable the characterization of clogging under relevant conditions at reasonable experimental durations.

Simplified model solutions (no carbonates, bicarbonates and relevant cations) were used as this work is a seminal one focused on the impact of molecular O₂ on the clogging process of Fe⁰ systems as influenced by pumice addition in various proportions. Testing more complex solutions relevant to simulate natural situation could be built on the results from these simplified systems.

Laboratory scale polymethyl methacrylate (Plexiglas) columns were operated in up-flow mode. The influent solution was pumped upwards from a single PE bottle using a precision peristaltic pump (Ismatec, ISM930). In all the tests the flow rate was maintained constant at a value of 0.5 mL/min. Tygon tubes were used to connect inlet reservoir, pump, columns and outlet. Six Plexiglas columns (50 cm long, 5.0 cm inner diameter) were used in the experiments (Fig. 1).

The ratio column diameter (D) to average material particle size (d) ensured the prevention of channelling and wall effects. In fact, used D/d ratio (actually 100–165) is by far larger than the threshold value of 50 (Badruzzaman and Westerhoff, 2005).

Six different systems were investigated (Systems A through F) (Table 1). System A was the operational reference system containing only pumice (0% Fe⁰) and system F was a pure iron column (100% Fe⁰). The volumetric proportion of Fe⁰ in the 4 other systems was 10, 25, 50 and 75% following a procedure recently presented (Noubactep and Caré, 2011; Noubactep et al., 2012b). In systems B to F, the mass of iron was fixed to 200 g. This mass represented either 100% of the reactive zone (rz) or the relevant volumetric proportion of rz (Fig. 1, Table 1). Table 1 summarizes the theoretical (rz_{theor} ; i.e. the height of the column occupied by the reactive medium when Fe⁰ and pumice were used in series and not as a mixture) and measured (rz_{eff} ; i.e. the height of the reactive zone in the column effectively measured) reactive zone for each individual systems.

The hydraulic conductivity was determined during the column tests, by either constant-head ($k > 10^{-6}$ m/s) or variable-head ($k < 10^{-6}$ m/s) permeability methods (Head and Keeton, 2008), at

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