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Geochemical modelling for predicting the long-term performance of zeolite-PRB to treat lead contaminated groundwater

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ABSTRACT

The feasibility of using geochemical modelling to predict the performance of a zeolite-permeable reactive barrier (PRB) for treating lead (Pb^{2+}) contaminated water was investigated in this study. A short-term laboratory column experiment was first performed with the zeolite (clinoptilolite) until the elution of 50 PV (1 PV = ca. 283 mL). Geochemical simulations of the one-dimensional transport of the Pb^{2+}, considering removal processes including: ion-exchange, adsorption and complexation; the concomitant release of exchangeable cations (Ca^{2+}, Mg^{2+}, Na^{+}, and K^{+}) and the changes in pH were subsequently performed using the geochemical model PHREEQC. The results showed a reasonable agreement between the experimental results and the numerical simulations, with the exception of Ca^{2+} for which a great discrepancy was observed. The model also indicated the formation of secondary mineral precipitates such as goethite and hematite throughout the experiment, of which the effect on the hydraulic conductivity was found to be negligible. The results were further used to extrapolate the long-term performance of the zeolite. We found the capacity would be completely exhausted at PV = 250 (ca. 3 days). The study, thus, generally demonstrates the applicability of PHREEQC to predict the short and long-term performance of zeolite-PRBs. Therefore, it can be used to assist in the design and for management purposes of such barriers.

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1. Introduction

Contamination of groundwater with heavy metals such as lead can pose a serious threat to drinking water supply systems and ecosystem health (Evanko and Dzombak, 1997). Therefore their removal from groundwater is necessary to avert these problems.

Permeable reactive barriers (PRBs) that utilize carefully selected reactive media (based on factors including: reactivity, permeability, environmental compatibility, availability, cost, and long-term stability) are a passive remediation technology that offer a cost-effective alternative to the conventional pump and treat (P&T) systems for remediating contaminated groundwater. In the PRB system, the attenuation of contaminants is achieved by their sequestration or biotic/abiotic transformation to less hazardous forms by the reactive materials via processes such as sorption, precipitation and biodegradation (Blowes et al., 2000; Carey et al., 2002; Henderson and Demond, 2007; Komnitsas et al., 2007; Obiri-Nyarko et al., 2014).

Numerous laboratory and short-term field studies that demonstrate the effectiveness of PRBs in attenuating a broad spectrum of contaminants are available (Baker et al., 1998; Bartzas et al., 2006; Blowes et al., 1997; Bone, 2012; Komnitsas et al., 2004, 2007; Obiri-Nyarko et al., 2015; Wilkin and McNeil, 2003). Long-term studies are, however, necessary to evaluate the longevity of PRBs due to some issues regarding their long-

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term hydraulic performance and reactivity (Kouznetsova et al., 2007). For example, Schipper et al. (2004) reported the diversion of flow beneath a nitrate-reducing PRB due to reduction in the hydraulic conductivity of the PRB. Li et al. (2005) observed the reduction of porosity ranging from 0.0007 to 0.03 per year in nine PRB installations. Mushovic et al. (2006) also observed the formation of precipitates, which resulted in the reduction of the reactivity and hydraulic conductivity by 3 orders of magnitude. Gibert et al. (2013) have also reported the reduction of residence time in a sulphate-reducing barrier due to fouling of the pore spaces and heterogeneities in the reactive material. Multi-tracer testing in the oldest commercially-installed ZVI-PRB in Europe also revealed a restricted groundwater flow in the upper 25 cm of the ZVI-PRB (Phillips et al., 2010). Wilkin et al. (2014) have also reported a similar observation in the oldest PRB in the US. Barton et al. (2004) reported the remobilization of UO_2^{2+} after the capacity of the reactive material was exhausted.

Due to the paucity of long-term studies/data on the reactive and hydraulic performance of PRBs (Wilkin et al., 2003, 2014), some researchers have used numerical modelling to predict the long-term performance of PRBs, based on data from short term lab-scale column experiments or field studies. The majority of these studies have, however, focused on ZVI due to its frequent application for the treatment of a wide range of groundwater contaminants (e.g., Agrawal et al., 2002; Bartzas and Komnitsas, 2010; Bartzas et al., 2006; Carniato et al., 2012; Jeen et al., 2007; Kouznetsova et al., 2007; Li et al., 2005, 2006; Liang et al., 2003; Mayer et al., 2001; Wanner et al., 2011; Yabusaki et al., 2001). Predictive modelling is not only useful for forecasting the longevity, but also for the optimization of the long-term performance of the PRBs, as well as enhancing the understanding of the complex interactions of physical transport and geochemical reactions that occur within the barriers. The successful development of these models, however, largely relies on the understanding and accurate estimation of the model parameters, as well as the inclusion of the appropriate (bio)-geochemical and physical transport processes (Dusek et al., 2015).

Natural zeolites are among the materials commonly used as an alternative to ZVI in PRBs for the removal of heavy metals due to their wide availability and cost-effectiveness (Lai, 2005; Li et al., 1999; Obiri-Nyarko et al., 2015; Rabideau et al., 2005). They are aluminosilicate minerals with high cation-exchange capacity and large surface area with different pore structures, allowing for selective adsorption of the metals (Perić et al., 2004). The long-term performance of zeolites depends on their continued effectiveness in precipitating, providing exchange sites or surface for the adsorption of the metals, as well as in maintaining the permeability of the PRB. Lai (2005) and Rabideau et al. (2005) employed numerical modelling to simulate the transport of contaminants through a zeolite-PRB. Rabideau et al. (2005) conducted laboratory studies to assess the performance of a zeolite treatment wall for removing Sr-90 from groundwater and extended their work further to predict the long-term performance of their zeolite-PRB using the K_d-based “reactive” transport and cation-exchange transport models. Although the K_d-based transport models are mathematically simple, they are limited in terms of their capability to sufficiently describe the complex geochemical processes occurring within the system (Bethke and Brady, 2000). The consideration of only ion-exchange reactions in the cation-exchange transport model also excludes other possible reactions such as precipitation. Natural zeolites are rarely pure as they may contain other mineral fractions which may allow contaminant (heavy metals) removal to occur via other processes in addition to the ion-exchange reactions. The work of Lai (2005), on the other hand, suggests that the combination of removal mechanisms such as adsorption, complexation and cation exchange could improve the performance of such transport models.

The aim of the present study was to investigate the feasibility of predicting the long-term performance of a zeolite-PRB. The study was divided into two parts. The first part consisted of accelerated short-term column experiment with zeolite to remove Pb^{2+} from contaminated acidic water. In the second part of the study, geochemical modelling was performed to simulate the 1D transport of Pb^{2+} through the zeolite following the approach of Lai (2005). The model was then extended further to predict the long-term performance of the zeolite-PRB.

2. Materials and methods

2.1. The zeolite

The zeolite used in this study was dominated by clinoptilolite. Selected properties of the zeolite determined in the laboratory are presented in Table 1.

2.2. Column experiments

The column experiments were carried out in duplicate using stainless steel cylinders (Fig. 1). Two ports were located at the base of the columns, one for feeding the column with the input solution and the other for influent sample collection. Two similar ports were also located at the top of the column for the discharge of the effluent solution and for effluent sample collection. The inlets and outlets of the columns were fitted with nylon mesh filters to prevent/minimize the outflow of the zeolite grains. The columns were clamped to an upright position to prevent the development of preferential flow paths. The total pore volumes of the columns were determined by the difference in dry and saturated weights of the columns. The total porosity was determined gravimetrically by dividing the total pore volume by the total volume of the columns. The

Table 1
Selected properties of the studied zeolite.

Chemical composition	Weight (%)
SiO_2	69.9
TiO_2	0.3
Al_2O_3	12.5
Fe_2O_3	1.6
CaO	2.4
MgO	0.9
MnO	1.1
Na_2O	0.3
K_2O	3.1
H_2O^+	8.0
Physical properties	
CEC	435.5 mmol(+)/kg
$pH_{(H2O)}$	7.1
Specific weight	2.3 g/mL

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