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Arsenic removal from groundwater using iron waste materials

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ABSTRACT

Potentially toxic elements and other substances are present in many regions of Slovakia as a consequence of previous industrial activities. This study involved the laboratory testing of the effectiveness of cheap and widely available materials to decrease the arsenic concentration in groundwater. The materials are to be installed in a pilot permeable reactive barrier in the Nováky area, Slovakia. Two types of cheap iron waste material (iron powder and iron shavings) were evaluated as reactive media, which form the active part of the experimental column. The arsenic concentrations at the column inlet and outlet, the permeability of the reactive material, pH, and iron concentration in treated water were measured over the course of 60 d. Throughout this time, arsenic levels in the outlet of both columns were far below the permitted limit of 100 ppb for arsenic content in groundwater. Iron shavings were chosen as final reactive material for environmental beneficial usage due to their high permeability, easy handling and high efficiency.

Keywords: Groundwater; Arsenic; Iron waste material;Remediation; Permeable reactive barrier

1. Introduction

Contamination of groundwater in different regions of the Slovak Republic is caused by industrial and mining activities, which are the sources of many hazardous substances.

Arsenic is one of the main potentially toxic elements, and it exceeds the permitted groundwater concentration limits in some contaminated areas of Slovakia [1]. Naturally occurring arsenic in groundwater is now considered one of the most deleterious public health crises of the world.

Inorganic arsenic species are toxic and are found in groundwater around the world at levels higher than the maximum contaminant level of 10 μg l^{-1} that is suggested by the World Health Organization (WHO) for drinking water [2]. Acute and sub-acute arsenic toxicity has been known for a long time. The major symptoms of acute arsenic poisoning include burning and dryness of the mouth and throat, dysphasia, abnormal pain, and hematuria [3]. It is a cumulative poison which inhibits SH groups in enzymes. Chronic poisoning leads to loss of weight and appetite as well as with related gastrointestinal disorders [4].

The toxicological effects of arsenic depend on its chemical form. Arsenic may occur in the environment in inorganic forms with different oxidation states, such as As(V), As(III), As(0) and As(−III), and as various organic compounds of arsenic [5]. In aerobic environments, As(V) is dominant, and it is usually in the form of arsenate $(AsO_4)^{3-}$. It behaves as chelate and can coprecipitate with or adsorb into Fe oxyhydroxides under acidic conditions [6]. Under reducing conditions, As(III) dominates and takes the form of arsenite $(AsO_3)^{3-}$, which is water soluble and can be adsorbed/coprecipitated with metal sulphides [7]. The inorganic As species are usually more toxic than

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the organic As species. As(III) is considerably more toxic and soluble than As(V) [8].

Currently, remediation of contaminated areas is needed due to the dangers of arsenic contamination of groundwater. Permeable reactive barrier (PRB) technology is a relatively novel groundwater remediation method which involves a groundwater flow system barrier that transforms contaminants to environmentally acceptable forms [9]. The implementation of a PRB in an affected area can be done after consistent chemical, geological, hydrological and environmental evaluation of the particular area. A key condition for beginning the complex process of implementation is the testing of the initial interaction of the contaminants and the reactive material, i.e., the active part of the PRB.

At present, a number of full scale [10–12], pilot scale [13–15] and laboratory scale [16–20] studies have been performed to investigate arsenic removal from groundwater and wastewater. In almost all the mentioned cases, high-efficiency remediation of arsenic to below the permissible groundwater concentrations has been achieved. The type of Fe material [18,19], pH [17–19], redox potential [19], dissolved oxygen [19], adsorption capacity [18], specific surface area [16,18] and corrosion rate [16] are all stated as influential factors of arsenic groundwater remediation.

Zero valent iron (ZVI) is one of the most frequently used reactive materials both in laboratory and fields applications. It was used for the remediation purposes in many reports either alone, or in a mixture with other materials [20–25]. Nanoscale iron particles showed excellent performance in remediation of wide spectrum of common environmental contaminants. The comprehensive overview of iron nanoparticle applications was published by Zhang [26]. Removal of As(V) in groundwater using nanoscale iron particles was examined in batch reactors. SEM-EDX results indicated the adsorption of arsenic onto the iron surface, and XRD analysis found the formation of iron corrosion products including lepidocrocite, magnetite and/or maghemite at a reaction period of 7 d [27]. A pelllet type iron-cerium bimetal oxide adsorbent was tested in column and batch experiments for the removal of As in groundwater. Both tests showed that the Fe-Ce adsorbent could remove As from groundwater with high efficiency [28].

The latest research in this area is devoted to the study of new and progressive materials. For the first time a continuously accelerated Fe corrosion driven by common oxidants (i.e., NaClO, $KMnO_4$ or H_2O_2) and thereby the rapid and efficient removal of heavy metals by zero-valent iron under the experimental conditions was demonstrated by Guo et al. [29].

A facile freeze-drying method was presented to fabricate three dimensional honeycomb-like structured nanoscale zero-valent iron/chitosan composite foams (ICCFs) for effective removal of inorganic arsenic in water [30]. The mesoporous iron/aluminium bimetallic particles were synthesized and employed for the removal of aqueous As(III). Particles exhibited its great potential as an effective and environmental friendly agent for arsenic because of a good As(III) removal capability and stability [31]. Results of work [32] indicated that nanoscale zero-valent iron supported onto pumice and modified by chitosan (CS-P-NZVI) might be an effective material for both in situ and ex situ remediation. As(III) could be removed by adsorption on CS-P-NZVI in a very short time, with high removal rates, over a wide range of pH and concentration.

Finding low-cost and readily available adsorbents or reactive media to remove heavy metal ions has become a main research focus. The reviews by Fu and Wang [33] and Mohan and Pittman, Jr. [34] are among the very comprehensive reference publications for these materials. Choi et al. [18] demonstrated the potential of waste cast iron as the reactive material to treat wastewater and groundwater containing arsenic. Simple and appropriate methods for removing of arsenic from drinking water using low cost methods have also been described in detail by Malik et al. [35].

Among many advantageous solutions, PRB technologies are not without challenges. One is its possible clogging and thus, uncertainty regarding the longevity of the PRB [36]. Various studies deal with physical [37], biological and chemical clogging [38]. Moraci et al. [39] studied ZVI long-term hydraulic conductivity, which often decreases during treatment and so potentially compromises the long-term efficiency of the barrier. Long-term column tests demonstrated how iron-pumice granular mixtures were efficient in contaminant removal and were able to maintain constant permeability of the PRB.

The main objective of this work was to test two types of iron waste materials and determine the more suitable one for the installation of a pilot permeable reactive barrier near an industrial area of Nováky. Use of waste materials and the lower costs associated with the huge amount of material were major environmental priorities for the pilot project. Laboratory analyses were used to measure arsenic removal efficiency and the permeability of the membranes. The results of this analysis may be used to implement the optimal material in a PRB at a particular location.

2. Material and methods

Laboratory experiments were performed in cylindrical columns. The active part of each column was 30 cm long and 4.6 cm in diameter, and it was filled with reactive material. The inlet and outlet of each active part were closed with perforated Teflon filters and permeable geotextiles. At the inlet of each column, a manometer was installed to measure the eventual pressure increase due to clogging. The general arrangement of the experiment is shown in Fig. 1.

A peristaltic pump was used for water transport into the columns and to keep the water flowing through the columns at a rate of 0.22 ml min^{-1} throughout the experiment. This rate was chosen based on the observed mean groundwater flow rate at a study site for potential future permeable reactive barrier installation.

Waste iron in this study was obtained from industrial company Tatravagónka, Poprad, Slovakia. Columns were filled with two different materials:

- waste iron powder with sand (fraction up to 4 mm), volume ratio 1:1, column marked as IPS
- iron shavings (lathe turning waste), column marked as IS

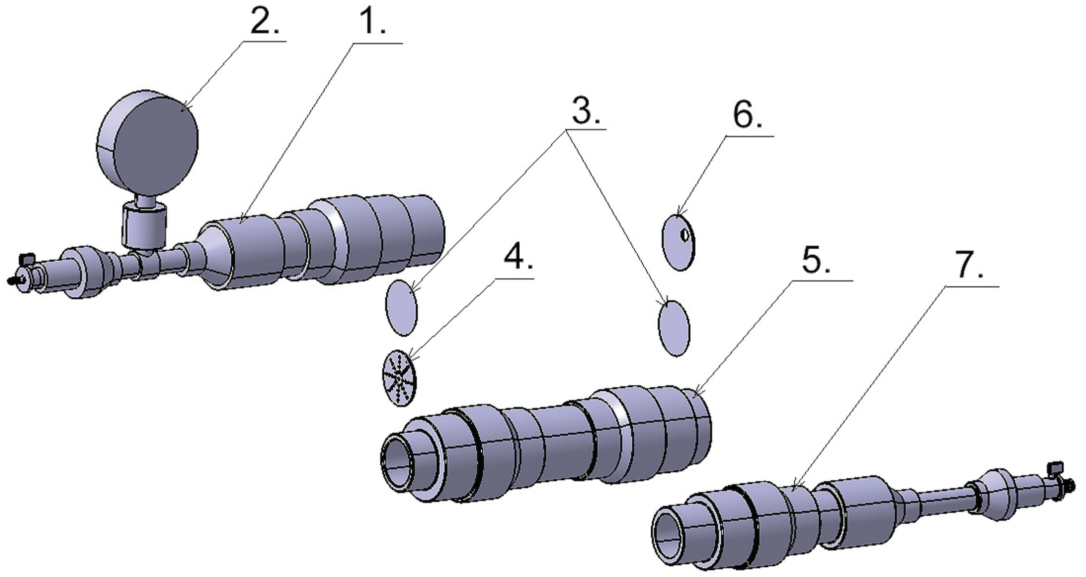

Fig. 1. Column scheme: 1. column inlet, 2. manomater, 3. permeable geotextile, 4. perforated inlet teflon filter, 5. column filled with reactive material, 6. perforated outlet teflon filter, 7. column outlet.

Table 1
Composition of the reactive media

Column mark	Volume of reactive material/cm^3		
	Iron powder	Sand	Iron shavings
IPS	310 (798 g)	310	–
IS	–	–	600 (520 g)

Table 2
The chemical composition of contaminated water

Element	Limit values		Sample of groundwater
	ID	IT	
As/µg l^{-1}	50	100	2580
Mo/µg l^{-1}	180	350	62
B/mg l^{-1}	0.5	5	3.1

The precise amounts of reactive and additional materials are summarized in Table 1.

The chemical composition of waste iron was determined using XRF spectrometry (Niton XL3t GOLDD+, Thermo Scientific, USA). pH measurements were taken daily using a pH meter (Orion3Star, Thermo Electron Corporation, USA).

The duration of experiment was 60 d for both columns. The real groundwater samples were collected from the Nováky area. Treated water was sampled after each 24 h period for arsenic and iron measurements. Sample volumes were 250 ml with 1 ml of additional concentrated nitric acid for sample conservation. Samples were refrigerated between sampling and analysis. An atomic absorption spectrometer (AA240Z, Agilent Technologies, USA) was used for As and Fe measurements. Analyses were performed by GEOLAB, Ltd. Košice, an accredited laboratory.

Leaching tests were performed on the reactive materials after the two months by the State Geological Institute of Dionýz Štúr, Slovakia, in accordance with the regulations of Annex. 22nd Decree of the Ministry of the Environment, Slovak Republic, n. 310/2013, to limit the values of the aqueous extracts of waste.

3. Results and discussion

The main groundwater contaminants present are summarized in Table 2. The established values were correlated with permitted limits of each element (according to Directive of the Ministry of Environment of the Slovak Republic from January 28, 2015 no. 1/2015–7) to carry out risk assessments of contaminated sites. The ID value is defined as a critical concentration of a pollutant in the soil, rocks or groundwater, above which there is endangerment of human health and the environment. This finding calls for monitoring of contaminated sites. The IT parameter is the critical value of concentration of a pollutant specified for the soil, rocks and groundwater, above which a given type of land use implies a high probability of risk to human health and the environment. The current situation means it is necessary to carry out a detailed geological survey of the local environment for risk analysis of polluted areas. According to Table 2, the IT criterion for arsenic concentration is far exceeded, indicating the necessity of remediation.

The chemical composition from XRF analysis showed that the major element in the waste iron material was Fe

(Table 3). IPS and IS contained 96.74% and 97.81% of Fe, respectively, indicating almost the same Fe content in both waste materials.

This experiment, with controlled water flow, was a continuation and extension of our previous study [40] that focused on short-duration laboratory experiments of arsenic removal. During the experiment, both the PRB parameters (permeability, stated by flow column) as well as water parameters (pH, concentration of arsenic and iron ions) were evaluated.

In the case of contaminated water (initial pH = 7.3) applied to the IPS and IS columns, the pH of the water exiting the columns was essentially stable, and it oscillated around a value of 7.7 throughout the 60 d (Fig. 2). The pH conditions during the experiment were comparable to those occurring in the environment, and no notable changes in this value were observed.

In spite of water flow adjustments by the peristaltic pump, the volume of flowing water was measured to detect any column clogging. In the experiment arrangement, there was a manometer included to signalize any potential blockage of the column. Over the course of experiment, the water flow experienced no complications and no flow disturbance was observed. The measured volumes passing through the columns are given in Fig. 3. Both of the used waste materials have sufficient permeability to allow the continuous passage of groundwater through the columns without interruption.

The corrosion of Fe in the reactive media was evaluated through measurement of Fe content in samples at the outlet of columns. A slow corrosion of reactive material, which is evident in oscillating iron concentrations between values 0–1 mg l^{-1}, indicates the gradual process of corrosion as well as the gradual stripping of ferrous or ferric ions from the iron, as shown in Fig. 4. It is a significant parameter because both the dissolution and the passivation of iron belong to the mechanisms that could cause a PRB becoming ineffective over time. The reaction mechanism of As removal has been intensively investigated by many research teams [41–44]. Fe corrosion is a key step in the reaction mechanism, which produces Fe oxides/hydroxides under the oxic condition. Thus As removal is attributed to adsorption onto iron oxides/hydroxides [45]. Heterogeneous reactions of corroding iron are complex and they could result in different variations of the adsorption surfaces for As(III) and As(V). Products are inner spherical surface complexes of As(III) and As(V) on Fe^{3+} hydroxide-oxide [41]. Considering that our experiment was conducted under the oxic conditions, above mentioned mechanism of As adsorption was expected.

Much attention has been paid to decrease arsenic concentration in groundwater. Because of its aqueous nature,

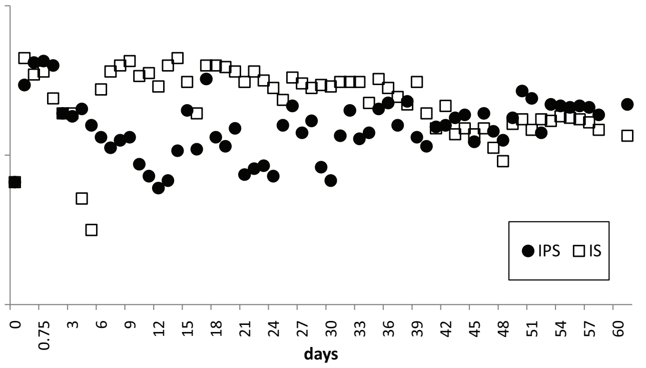

Fig. 2. Time dependence of groundwater pH on columns IPS and IS.

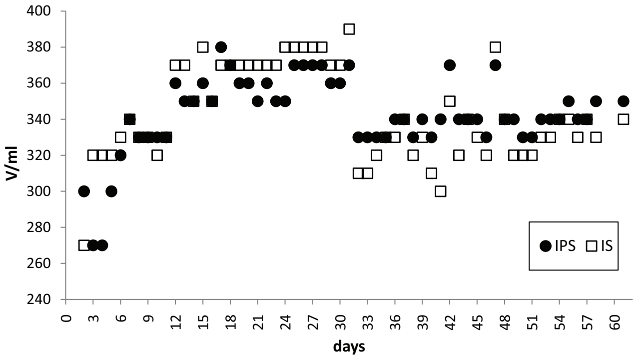

Fig. 3. Time dependence of groundwater flow volumes per day on columns IPS and IS.

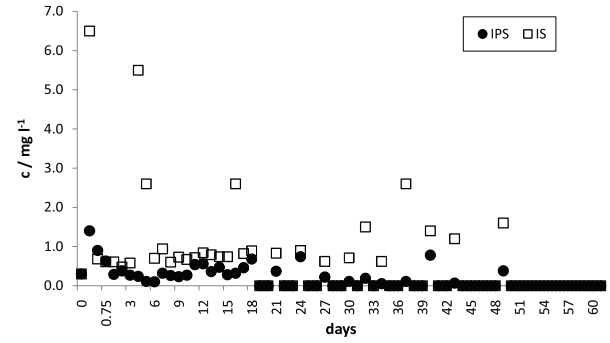

Fig. 4. Time dependence of iron concentration in water after IPS and IS column passing.

Table 3
XRF analysis of the elements content

	Element %								
	Fe	Mn	Cr	Ni	Si	P	Cu	Zn	Rest
IPS	96.74	1.06	0.22	0.13	0.48	0.14	0.19	0.02	Nb, Mo, C
IS	97.81	1.36	0.36	0	0.18	0.30	0	0	C

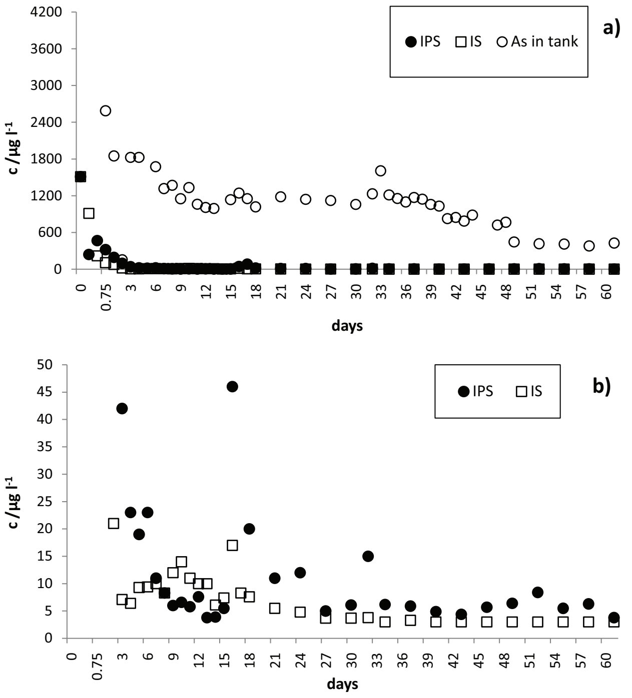

Fig. 5. Time dependence of arsenic concentration in groundwater after: (a) IPS and IS column passing, (b) cut chart of 5(a) with arsenic concentration up to 50 µg l^{-1}.

it was necessary to measure the content of As in the tanks before the water entered the columns. Fig. 5a compares the As concentrations before and after passing through the IPS and IS columns. For better illustration, Fig. 5b shows a magnification of the chart up to c_{As} = 50 µg l^{-1} (ID criteria). After 60 d, the effectiveness of water remediation was 99.11% and 99.29% for IPS and IS, respectively. These results, after laboratory tests, were satisfactorily sufficient and allowed the use of both materials for the pilot application. Nevertheless, IS reactive material was selected because it is easier to handle, and it was not necessary use additives in its preparation.

Exhausted reactive material used in the PRB requires either the reclamation, if possible, or its replacement. Wasted reactive material will be stored in a landfill broken down by the severity of the material toxicity. To examine the possibilities for the landfill material used, the favored IS reactive material was subjected to a leaching test in an aqueous solution according to assess the risks of material used at the landfill after the experiment. None of the evaluated elements (As, Al, Ba, Cd, Co, Cr, Cu, Hg, Mo, Ni, Pb, Sb, Se, Sn, V, Zn, Cl$^-$, F$^-$, SO_4^{2-}, DOC, CRL, CN$^-$) exceeded the leaching values for inert waste, i.e., the 1st class for leaching, which is not dangerous for the environment.

4. Conclusions

Two types of iron waste materials were evaluated in the laboratory as PRB materials for the remediation of groundwater from the Nováky site, which exceeds the state environmental standards for arsenic pollution. The lifetime of the reactive materials in natural water conditions was sufficient and had a negligible effect on groundwater pH; the permeability of both materials are also favourable for use.

The aim of this work was to test different waste materials under different conditions to determine the suitability for use in permeable reactive barriers for arsenic contaminated groundwater treatment, and based on the results, we conclude the following:

1. Iron powder an iron shavings are both suitable materials for application in groundwater at the Nováky locality due to the slow corrosion of these waste materials and their minimal effect on groundwater pH in natural conditions.
2. According to the arsenic concentration decrease in the original groundwater, both of the tested materials are very efficient, demonstrating over 99% effectiveness in arsenic removal over 60 d.
3. As a final recommendation for permeable reactive barrier installation in the environment, iron shavings are recommended because of their high effectiveness, good permeability, easy handling and simple replacement in the PRBs.

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