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To cite this article: Mieczysław Połoński et al 2017 IOP Conf. Ser.: Mater. Sci. Eng. 245 072017

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## **Optimization Model for the Design of Multi-layered Permeable Reactive Barriers**

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Abstract. Permeable reactive barriers (PRBs) are employed as in situ groundwater remediation technology. The installation of PRBs is usually a major investment, where one of the biggest cost drivers are material costs. PRBs are barriers against contaminants moving under the natural gradient, however not against groundwater contaminants. The most common construction of a PRB is a single barrier, but in the case of contaminant mixtures a multi-layered construction, i.e. a combination of different reactive materials and removal processes, is required. The most important parameters for PRB design are dimensions. The barrier must be long enough to treat the entire width of the plume (dimension perpendicular to groundwater flow) and should extend to and be keyed into an impermeable layer. The problem is to determine the optimal thickness of a PRB, which should provide a residence time appropriate for reducing the concentration of contaminants to the desired effluent concentration. In PRBs, design is accomplished using numerical methods or simulators, which are useful to predict the scenarios and evaluate the resulting groundwater flow systems to specific site conditions. On the other hand, numerical methods are complicated and may have significant errors if the discretization is too coarse or is incorrectly aligned. This paper deals with a simple, conceptual model of a one-approach optimization method for multi-layered PRB design. Based on literature and laboratory test results (residence time, density and hydraulic coefficient), a selection of layers of reactive materials was determined. Considering the lowest cost of the reactive materials, the required thicknesses of activated carbon, zeolite and zero valent iron were calculated using two different algorithms. The simple model may be used for preliminary barrier design and cost calculations. Using the optimization model in a preliminary design stage, it is possible to reject the PRB concept and avoid losing time for the complicated analysis.

#### 1. Introduction

Groundwater contamination is becoming a serious problem in the world since its sources are not controlled in an effective manner. This is a major issue on the world agenda, because groundwater is one of the most important sources of drinking water. Groundwater contaminants come from two categories of sources: point sources (landfills, leaking gasoline storage tanks, leaking septic tanks, and accidental spills) and distributed, or non-point sources (infiltration from agricultural land treated with fertilizers and pesticides, contaminants in rain, snow, and dry atmospheric fallout). Contaminants come from natural sources (e.g. erosion processes) or numerous types of human residential, municipal,

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commercial, industrial, and agricultural activities. The presence of contaminants in groundwater at concentrations exceeding background levels demonstrates a high potential for health and environmental risks. Major contaminants include a variety of volatile hydrocarbons (such as benzene, toluene, ethylene, and xylene), heavy paraffin and chlorinated organic compounds (polychlorinated biphenyls PCB), and inorganic compounds (heavy metals, arsenic and mercury, radionuclides, such as tritium) [1].

The nature of groundwater is to move slowly, thus contamination often remains undetected for long periods of time. This makes the clean-up processes very difficult and in many cases even not possible. In these cases, remediation may cost thousands to millions of euros. If the contaminant source has been controlled or removed, contaminated groundwater may be treated in one of the many ways ([2]: containing the contaminant to prevent migration; pumping the water, treating it, and returning it to the aquifer; leaving the groundwater in place and treating either the water or the contaminant (stabilization/solidification, soil washing, air stripping, precipitation, vitrification, thermal desorption); allowing the contaminant to attenuate (reduce) naturally (with monitoring), following the implementation of an appropriate source control. The most traditional, active remediation technique (energy-intensive) is characterized by good efficiency but high operational costs [3]. In the past decades, few in situ passive technologies have also been proposed, including permeable reactive barriers (PRBs). PRBs are engineering constructions filled with reactive materials in the subsurface, which use the natural hydraulic gradient of the groundwater plume to move the contaminants through the reactive zone, which is advantageous over traditional technologies by being more cost effective and lower maintenance in the long-term [4,5]. In this remediation technology, one of the most important factors in the investment process is the longevity of PRBs connected with dimension parameters and total capital and operating costs including field and laboratory tests, reactive materials, installation, and monitoring costs. Compared to traditional technology remediation, PRBs may be more costly in the initial stages, especially during installation. However, since PRBs are a passive system, long-term costs are lower than traditional remediation operations and maintenance [6]. The total cost of a PRB system may be at least sixty percent cheaper than the equivalent pump-and-treat system. According to cost data obtained from the US federal agency sources, including case studies and reports, at least one third of these costs represented material cost and thus the materials volume may also influence the overall cost of the construction [6-8]. The most common construction of PRBs is a single barrier. Often, groundwater is contaminated with a mixture of contaminants and in this case a multi-layered construction (MPRB), i.e. a combination of different reactive materials, is more plausible. In PRB design, long-term procedures that have complicated calculation methods (numerical methods or simulators) are used. Besides numerous advantages, which are useful to predict the scenarios and evaluate the resulting groundwater flow systems for specific site conditions, these methods have also many faults, which include a large amount of input data or significant errors if the discretization is too coarse or is incorrectly aligned.

The objectives of this paper are to illustrate the optimization of a MPRB design including working time and cost effectiveness. In the conceptual model of the single approach optimization method, the lowest cost of reactive materials and the required thicknesses of material layers were calculated. This simple model may be used for preliminary barrier design and cost calculations. Using the optimization model in a preliminary design stage, it is possible to reject the PRB concept and avoid losing time for the complicated analysis.

### 2. PRB design

The main aims of PRB design are to ensure that the contaminant plume is intercepted for treatment (there is no contaminant flow beneath, around, or above the treatment zone) and provide sufficient PRB dimensions to achieve the relevant contact time between contaminants and reactive media needed for the reduction of contaminant concentrations to acceptable levels.

Selection of reactive materials for PRBs is generally predicted by: the type of contaminants to be removed (organic and/or inorganic), their concentrations and mechanisms needed for their removal (e.g. biodegradation, sorption, precipitation); the hydrogeological and biogeochemical conditions of the aquifer; the environmental/health impacts; the materials' mechanical stability (capacity to maintain hydraulic conductivity and reactivity over time), and their availability and cost [8,9]. The basic reactive materials applied include: zero valent iron (ZVI), activated carbon (AC), zeolite (Z), and peat [10-15]. Single-layered constructions were frequently applied in the early stages of PRB technology. In addition, over the past few years, multi-layered systems have increasingly been used due to several advantages. They improve permeability, reduce costs, increase the number of removal mechanisms, enhance and accelerate removal rates, and considerably sustain the long-term performance of barriers [14-16].

After selection of reactive materials, the dimensions, location, and orientation of PRBs must be determined. The most important parameters include the residence time and the capture zone, which refers to the width of the barrier required to intercept the entire contamination plume [15]. The residence time is defined as the contact time between the contaminated groundwater and the reactive material required to achieve the treatment goals [17-20], or the time that the contaminated groundwater needs to pass through the reactive materials in a PRB [21,22]. A proper PRB design must assure that the residence time is sufficient to treat all contaminants in the polluted environment. It is mainly determined by groundwater velocity and thickness of the reactive materials in the PRB.

Dimensions of a single PRB are shown in figure 1 and include: length (L), which is perpendicular to groundwater flow, flow-through thickness, or width (b), and depth (h). The PRB should be long enough to treat the entire width of the plume (dimension perpendicular to groundwater flow) and should extend to and be keyed into an impermeable layer (bedrock) to reduce the chance of contaminant bypass under the barrier. If there is no sufficient bedrock, the depth of a PRB should extend to below the depth of contamination. The monitoring system should be sensitive enough to notice that contaminant bypass has not occurred [8].



Determining the PRB length and depth is relatively simple, whereas indicating the optimal thickness is more demanding. The PRB thickness must be designed based on the required residence time  $(t_R)$  of the contaminants and the groundwater flow velocity to allow for effective remediation. According to the recommendations of the Interstate Technology & Regulatory Council (ITRC), the required PRB flowthrough thickness (b) may be determined as follows:

$$b = v \cdot t_R \tag{1}$$

where v is the groundwater flow velocity and  $t_R$  is the residence time. Moreover, the thickness of the PRB may be calculated from estimates of flow velocities, residence times and removal efficiencies per unit length of the reactive materials obtained from column test data or hydrological modelling and direct measurements [8,15]. In practice, it is assumed that PRBs filled with ZVI must work efficiently for at least 10 years to be environmentally more effective in relation to other remediation technologies [23]. In the case of using parameters estimated from laboratory studies, longevity is considered to be approximately proportional to the thickness (*b*) of a barrier [24]. Finally, in calculating the optimal thickness, workable longevity (10 years) and evaluation of the minimum thickness (the thinnest layer of reactive materials that may reduce concentrations of contaminates to target levels) should be examined. Moreover, ITRC has recommended the application of safety factors to the result obtained to account for seasonal groundwater flow variations, field uncertainties, and potential loss of media reactivity [8]. Various unpredictable changes that may affect the barrier's longevity have been studied in several papers [4, 8, 15, 25-28].

#### 3. Optimization task of the PRB dimensions

The design of thickness in a single-layered PRB is a difficult and complex task, since there are various initial parameters, which should be taken into account in the decision-making process. It is more complicated if the barrier has a non-standard structure, such as multiple layers of reactive materials. The optimal values for a set of decision variables in a specific system should be determined by optimization. Optimality is defined with respect to a specified objective function and is subject to a set of constraints. For a single objective optimization, the objective function is the measure of the effectiveness to be optimized, such as the maximization of residence time and minimization of total costs, or associated with the design of a PRB.

For this purpose, the optimization task consisting of identifying the thickness of a single layer in a multi-layered barrier (MPRB) was proposed for two options of calculations using a single objective algorithm. The composition of MPRB materials for remediation of groundwater heavily contaminated by a mixture of heavy metals (Cd, Cu, Ni, Pb, Zn) using different layers of activated carbon (GAC), zeolite (Z) and zero-valent iron (ZVI) basing on laboratory test results and literature [16, 29, 30] was determined. In groundwater contaminated by mixtures of heavy metals (e.g., Cd, Cu, Ni, Pb, Zn), several processes such as sorption, ion-exchange, reductive-oxidative degradation, reduction and/or precipitation processes offered by ZVI, GAC, and zeolite may be used [8, 31, 32].

For a single objective optimization, the maximization of residence time in the first option and the minimization of costs of reactive materials in the second option were applied as optimization criteria. For the calculations, it was assumed that the barrier had height (h) and length (L) equal to 1 m, respectively. Unknown values were thicknesses of single layers  $(b_m)$  in the MPRB. In the barrier, the total number of used materials (M) was limited to 3. In addition, the following restrictions were implemented:

- minimum thickness of one layer (0.1 m);
- maximum width of the entire barrier (2 m);
- minimum thickness of entire barrier (1 m);
- minimum retardation time of WPBR (4000 days).

Inputs to the optimization model were based on the cost of buying 1 Mg of reactive material and the parameters determined from laboratory tests performed in the Department of Geotechnical Engineering of the Warsaw University of Life Sciences [33-35] and were as follows: flow velocity v [m/s], retardation factor R for mixtures of heavy metals [-], and density  $\rho$  [kg/m3]. The values of the input parameters are shown in Table 1.

М	Reactive material	v <sub>m</sub> [m/s]	ρ <sub>m</sub> [kg/m3]	R <sub>m</sub> [-]	cost <sub>m</sub> [euro]
1	GAC	0.0000016	0.45	39.20	2325.58
2	Zeolite	0.00000213	1.05	1438.10	30.23
3	ZVI	0.00000476	6.70	61.30	1627.91

In order to solve the optimization task, equation (2) can be used to determine the required resident time for contamination removal [36]:

$$t_{Rm} = \frac{b_m R_m}{v_m} \tag{2}$$

where:  $b_m$  – thickness of a single layer of WPBR [m],  $R_m$  – retardation factor of a single layer [–],  $v_m$  – flow velocity through a single layer m [m/s]. Moreover, the cost of a single layer of a MPRB *cost<sub>m</sub>* (h & L = 1 m) can be examined using the following formula:

$$cost_m = b_m \cdot \rho_m \cdot cost_m \tag{3}$$

The total purchase cost of reactive materials (*cost*) filling a WPBR is the sum of the costs of single layers and is as follows:

$$cost = \sum_{m=1}^{M} b_m \cdot \rho_m \cdot cost_m \tag{4}$$

The optimization calculations were performed in Microsoft Office Excel add-in program SOLVER using the "LP Simplex" algorithm. Similar calculations of optimizing dimensional parameters of PRBs in Microsoft Office Excel were conducted in the reports by Painter [37] and Craig [38].

#### 4. Results and discussions

Table 2 summarizes the main results of the single objective optimization. The results clearly show minor differences between the two options. Differences occurred in the calculation of the thickness of the zeolite layer M2. In option I, the optimal thickness was 1.55 m, and in option II – 0.55 m. The parameter, which had crucial impact on the calculation result was factor *R*. Among reactive materials, zeolite was characterized by a several dozen times longer delay of the flowing contaminants through the reactive zone than the other materials. Therefore, in the case of resolving the objective function for the maximization of residence time  $t_R$ , the thickness of zeolite was several times greater than for the other materials. The workable longevity of MPRB in this variant was estimated at 34 years (12,206 days), which is three times longer than the assumed limit (more than 10 years).

М	Reactive	Variant I			Variant II			
	material	$b_m$ [m]	$t_{Rm}$ [days]	$cost_m$	b <sub>m</sub> [m]	$t_{Rm}$ [days]	$cost_m$ [euro]	
				[euro]				
1	GAC	0.35	100.00	369.06	0.35	100.00	369.06	
2	Zeolite	1.55	12091.59	49.12	0.55	4277.19	17.38	
3	ZVI	0.10	14.91	1090.70	0.10	14.91	1090.70	

Table	2.	Cal	cul	lation	resul	lts
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In option II, where the objective function was minimization of the total costs, the calculated thickness of the zeolite layer was reduced by 1 m. Despite the reduction of the total thickness of the MPBR to 1 m, the time of effective work in accordance with the assumptions was retained and amounted to more

than 12 years (4,392 days). The buying cost of reactive materials to fill a MPRB with the following dimensions: layer thickness calculated in the options, height h and length L equal to 10 m, was 150,887.25 euros in option I and 147,712.83 euro in option II.

#### 5. Conclusions

Remediation of contaminated groundwater is a long-term and expensive process; in consequence, careful PRB design based on data for a specific site, the contaminant plume and the characteristics of the reactive material is required. Furthermore, optimization of workable longevity and dimension of a construction should be performed in order to assure cost-effective and widely applicable technology.

In this work, a single object task for the optimal design of a multi-layered PRB for groundwater remediation was proposed. Comparing the results of optimization calculation covering total residence time  $t_R$  and the total cost of purchasing reactive materials  $cost_m$ , it can be assumed that, despite the higher cost of more than 3000 euro, option I should be applied in the field, due to the fact that it assures a three times longer effective work in the environment. In the case of option II, the total thickness of a MPRB would not be sufficient enough for the removal of all contaminants, which would result in the need for replacement of reactive materials. This operation would significantly increase the total cost of the investment by increasing the costs of material and the cost of additional work and equipment. In future works on the optimization of PRB thickness or/and dimensions with simultaneous consideration of two divergent criteria, a multi-criteria algorithm should be applied.

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