Hydraulic conductivity of zeolite-sand mixtures permeated with landfill leachate

Conductivité hydaulique de mélanges zéolithe-sable infiltrés par des écoulements de décharge de déchets

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ABSTRACT: The study presented in this paper has been undertaken to evaluate the influence of leachate from landfill collected the municipal solid wastes at Warsaw district onto the hydraulic conductivity of zeolite-sand mixtures (with 50% and 20% content of zeolite). The long-term hydraulic conductivity tests with distilled water and landfill leachate collected from the landfill drainge were carried out using Trautwein equipment. Results of study indicated that hydraulic conductivity of reactive material tested have changed almost by two orders of magnitude (from 9.23E-5 to 1.24E-6 m/s). The decrease of hydraulic conductivity can be caused by reduction of the effective porosity due to pore clogging. The analysis of the calcium carbonate content using Scheibler method showed no significant increase of carbonates in the samples, while research in scanning electron microscope showed increased calcium content and crystals of calcium carbonate in the samples. Moreover, the presence of microbial activity had been observed. The conclusions drawn based on tests results should be considered in designing of PRB.

RÉSUMÉ : L'étude présentée dans cet article a été entreprise afin d'évaluer l'influence des écoulements des sites d'enfouissement des déchets du district de Varsovie sur la conductivité hydraulique de mélanges de zéolithe et de sable (avec 50% et 20% de teneur en zéolithe). La conductivité hydraulique à long terme a été mesurée avec de l'eau distillée et un liquide provenant du drainage d'une décharge, à l'aide d'un appareil de Trautwein. Les résultats de l'étude ont montré que la conductivité hydraulique du matériel réactif testé a changé de deux ordres de grandeur (de 9,23E-5 à 1,24E-6 m/s). La diminution de la conductivité hydraulique peut être due à la réduction de la porosité effective due à l'obstruction des pores. L'analyse de la teneur en carbonate de calcium en utilisant la méthode de Scheibler n'a montré aucune augmentation significative de carbonates dans les échantillons, alors que l'analyse par microscopie électronique à balayage a montré une augmentation de la teneur en calcium et des cristaux de carbonate de calcium dans les échantillons. En outre, la présence d'une activité microbienne a été observée. Les conclusions basées sur le résultat de ces essais doivent être prises en compte dans la conception des barrières réactives de protection.

KEYWORDS: permeable reactive barrier, landfill leachate, reactive materials, hydraulic conductivity.

1 INTRODUCTION

Throughout the world the pollution of groundwater by hazardous substances (e.g. landfill leachate) is one of the most crucial environmental problems. To protect the natural groundwater environment many technologies were developed in the last decade. One of the most effective and simultaneously low-cost technology is a method based on permeable reactive barriers - PRBs - that improves natural attenuation processes (Fig. 1). PRBs can be an effective remedy in many environmental settings with varying hydrogeologic and geochemical conditions.

According to ITRC publication (2011), ideal case for PRB application is ground containing cohesive silts and sands with hydraulic conductivity values lower than 3.5E-6 m/s. However, ITRC proposes to extend this criteria and also check the ground with hydraulic conductivity and groundwater velocity in range of 3.5E-6 to 3.5E-5 m/s as the most suitable conditions for application of PRBs.

The proper functioning of PRBs depends on the hydraulic properties of reactive materials that fullfield reaction zone of the barriers. The hydraulic criterion in the evaluation of reactive materials such as zeolites and zeolite-sand mixtures is expressed as the ratio of hydraulic conductivity of the reactive material (k_s) to the hydraulic conductivity of the aquifer (k_g), which, according to common recommendation, should be higher than one. This parameter governs the rate of occurring processes (sorption, biological and chemical reduction etc.) and in some circumstances can lead to changing groundwater flow direction ever to wrap the PRBs by contaminated groundwater. In this regard, it is necessary to identify changes of hydraulic conductivity as a result of contact with liquid pollutants.

The hydraulic conductivity of reactive materials and their changes during exploitation should be taken into account for design of PRB, particularly their thickness (b), which can be estimated from the equation (Czurda and Haus 2002):

$$b = \frac{t_{PRB} \cdot k \cdot i}{R} \quad (1)$$

where:
t_{PRB} - "working time" (s), time required for contaminant reduction to acceptable level,
k - hydraulic conductivity (m/s),
i - hydraulic gradient,
R - retardation factor, that depend on sorption processes intensity.

The changes of permeability of zeolite-sand mixtures, proposed as low-cost reactive materials, caused by landfill leachate are presented in this paper. The leachate samples were taken from municipal solid waste site in Warsaw (Radiowo site).

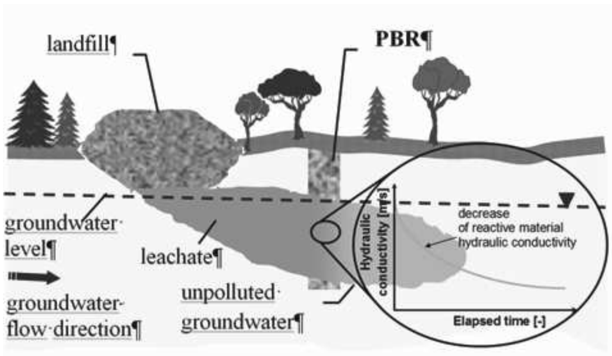

Figure 1. A concept of permeable reactive barriers.

1 MATERIALS AND TEST METHODS

Tests, result of which are presented in this paper, were carried out on zeolite-sand mixtures with 50% - ZS50, 20% - ZS20 content of Na-form of Slovak zeolite having 0.5-1.0 mm particle sizes. The mineral composition of the Slovak zeolite is as follows:

$$(Ca;K_2;Na_2;Mg)_4Al_8Si_{40}O_{96}\cdot 24H_2O \quad (2)$$

The zeolite-sand mixtures considered as a reactive materials have specific surface areas from 3,65 m^2/g for ZS20 to 29,04 m^2/g for ZS50. The crystal structure of zeolite is presented by 3-dimentional aluminosilicate framework with the developed system of micropores and channels occupied by water molecules and exchangeable cations. Crystal habits of zeolite include blocky and thin-tabular crystals with good monoclinic crystal forms (Sprynsky et al. 2004). According to the results of scanning electron micrographs study unit cell parameters of crystals are close to each other (Fig. 2).

To determine the effect of synthetic leachates (containing 360 mgNH_4^+/L, 100 mgCa/L, 200 mgMg/L and 100 mgCu/L) and landfill leachate on the hydraulic conductivity of ZS50 and ZS20, constant-head permeability tests were performed using flexible wall permeameter (Fig. 3). In the tests the natural leachate from municipal landfill in Warsaw was used. Chemical composition of leachate is listed in table 1. Due to the character of these studies, it was necessary to use equipment made from materials not reacting with contaminated water. The hydraulic gradient of 2 was obtained by establishing an elevation difference between the liquid surface of inflow and outflow ends. Before leachate tests, specimens were pre-saturated with distilled water until establishing the constant flow through the samples. Samples

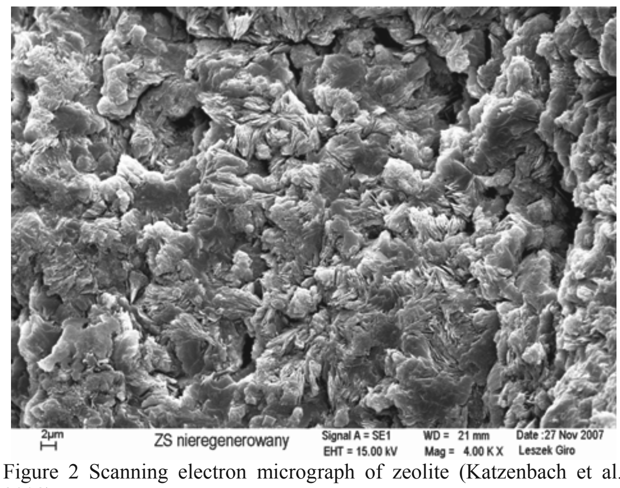

Figure 2 Scanning electron micrograph of zeolite (Katzenbach et al. 2008)

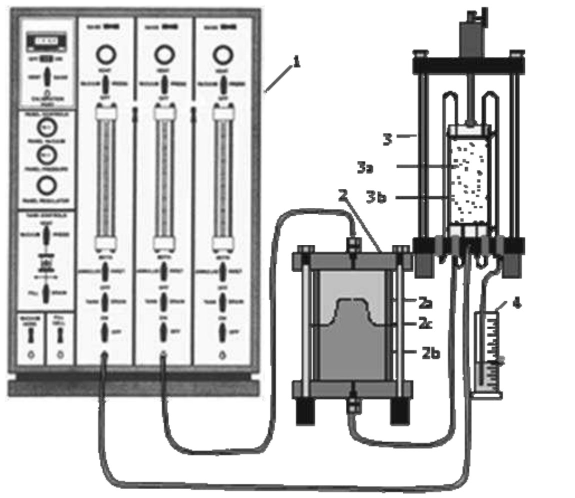

Figure 3. Scheme of a flexible-wall permeameter: 1-control panel, 2-bladder accumulator, 2a-water, 2b-liquid other than water, 2c-elastic membrane, 3-chamber, 3a-sample, 3b-latex membrane, 4- measuring cylinder (acc. to Trautwein Soil Testing Equipment Co.).

Table 1. Chemical composition of leachate from municipal landfill in Warsaw.

Parameter	*Value*
BOD (mg/l)	127.0
COD (mg/l)	960.0
Ammonia – N (mg/l)	52.0
Total P (mg/l)	3.94
Chloride (mg/l)	1400.5
Sulfate (mg/l)	419.0
Sodium (mg/l)	917.0
Potassium (mg/l)	396.0
Calcium (mg/l)	81.2
Magnesium (mg/l)	88.8
Iron (mg/l)	1.28
Chromium (mg/l)	0.13
Zink (mg/l)	1.83
Copper (mg/l)	0.2
Conductivity (µS/cm)	6480
pH	8.21

were placed between two porous plates, that's hydraulic conductivity was more than 1.0E-3 m/s. The samples were making with mixed materials having 10% of water content. The specimens tested were 0,07 m in diameter and 0,12 m in height. In this study specimens were compacted to relative density equal 0.6. The confining pressure was 50 kPa.

The hydraulic conductivity was calculated using equation (ASTM D 5084 - 00):

$$k = \frac{V}{i \cdot A \cdot \Delta t} \tag{3}$$

where:
k – hydraulic conductivity (m/s),
V – volume of effluent (m^3),
i – hydraulic gradient,
A – area of the specimen (m^2),
t – permeation time period (s).

2 RESULTS AND DISCUSSION

The hydraulic conductivity tests were performed with distilled and tap water, synthetic and landfill leachate. The results obtained are presented in Fig. 4. The hydraulic conductivity of samples is presented in relation to pore volume of flow, which may be calculated using equation:

$$PVF = \frac{k \cdot i \cdot t}{n_e \cdot L} \tag{4}$$

where:
PVF – pore volume of flow,
k – hydraulic conductivity (m/s),
i – hydraulic gradient,
t – elapsed time (s),
n_e – effective porosity,
L – length of the sample (m).

The hydraulic conductivity of materials tested using tap water was 9.8 E-5 m/s and 8.7E-5 of ZS50 and ZS20, respectively. The hydraulic conductivity slightly decrees with time at the beginning of the permeation with synthetic leachates but stabilized after 100 pore volume of flow. After 800 pore volume of flow hydraulic conductivity of ZS50 was 3.4E-5m/s and of ZS20 6.8E-5 m/s. This represented about a 2.90- and 1.30-fold decrees as compared with the results of hydraulic conductivity to tap water. The test results indicated that the permeation synthetic leachate influence negligible the hydraulic conductivity of zeolite-sand mixtures. Franciska and Glatstein (2010) observed the similar relation for compacted soil liners permeated with landfill leachate. Results of study with landfill leachate indicated that hydraulic conductivity of ZS50 have changed almost by two orders of magnitude (from 9.23E-5 to 1.24E-6 m/s). Observed permeability changes can be caused through inorganic compounds precipitation, mainly $CaCO_3$, and bioactivity (VanGulck and Rowe 2004, Asadi et al. 2011). The analysis of the calcium carbonate content using Scheibler method showed no significant increase of carbonates in the samples, while research in scanning electron microscope showed increased calcium content and crystals of calcium carbonate in the sample. Achieved results allow to conclude, that permeability changes of zeolie-sand mixtures may influence the operation conditions of PRB.

3 DESIGN CONSIDERATION

Considering the test results obtained the hydraulic criterion in the evaluation of reactive materials such as zeolites and zeolite-sand mixtures should be expressed as follows:

the ratio of hydraulic conductivity of the reactive material (k_s) to the hydraulic conductivity of the aquifer (k_g)

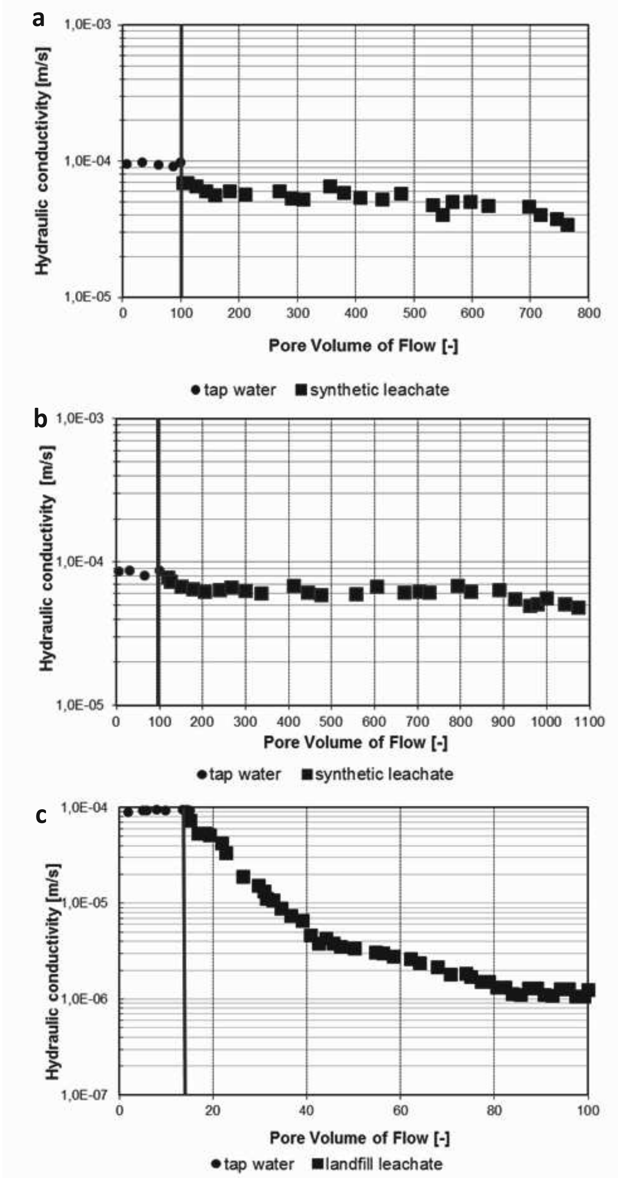

Figure 4. Hydraulic conductivity of ZS50 (a) and ZS20 (b) to tap water and synthetic leachate and ZS50 to landfill leachate (c).

$$\frac{k_s}{k_g} \geq 10 \tag{5}$$

Criterion (5) results from the necessity of taking into account changes of the hydraulic properties of materials during PRB exploitation. Laboratory studies showed a decrease of hydraulic conductivity values of zeolite and zeolite-sand mixtures under the influence of landfill leachate. In the case of zeolite-sand mixtures with a zeolite content Z (dimensionless value), estimation of hydraulic conductivity k_s in m/s at relative density I_D requires the application of the following equation (Fronczyk 2008):

$$k_s = A \cdot Z \cdot I_D^{-0.328} \tag{6}$$

For mixtures made of zeolite with particle size of 1.0–2.5 mm, A = 3.0 · 10-4, whereas at particle size of 0.5–1.0 mm, A = 2.6 · 10-5.

4 CONCLUSION

The choice of the reactive material for PRBs requires determining the hydraulic conductivity of reactive materials with regard to the relative density and strain conditions (effective stress) in the barrier. It is necessary to carry out "compatibility tests", i.e. test of contaminated groundwater influence on hydraulic properties of the selected material.

Achieved results for synthetic leachate showed less than 3-fold decrease of zeolite-sand mixtures permeability. Landfill leachate caused decrease of hydraulic conductivity almost by two orders of magnitude due to inorganic precipitation and biological growth.

5 ACKNOWLEDGEMENTS

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