Geotechnical requirements for *in-situ* permeable reactive barriers

Conditions requises pour les barrières perméables réactives *in situ*

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ABSTRACT: Permeable Reactive Barriers (PRBs) represent an efficient alternative to pump-and-treat methods for remediation of contaminant plumes in groundwater. Research activities at many places concentrate on the development of innovative reactive materials to apply with this technology. In most cases geochemical aspects are of major concern. However, in this paper geotechnical test methods for successful application of reactive barrier materials are proposed. Furthermore, a chromate contaminated site is described where a PRB is to be installed to a depth of 40 m.

RÉSUMÉ: Les barrières perméables réactives (PRBs) représentent une alternative efficace aux méthodes de pompage et traitement pour la réhabilitation d'eaux souterraines polluées. Des recherches sur le développement de nouveaux matériaux réactifs ont été entreprises à plusieurs endroits dans le but de les appliquer à cette nouvelle technologie. Dans la plupart des cas, les aspects géochimiques sont le point principal des PRBs. Dans cet article, des tests géochimiques sont proposés pour une application réussie des matériaux relatifs aux barrières réactives. En outre, un site contaminé au chromate, où une PRB reste à installer jusqu'à une profondeur de 40 mètres, est décrite.

1 INTRODUCTION

Increasingly it is realized that, under economical and ecological aspects, conventional pump-and-treat systems for remediation of contaminant plumes do not lead to satisfying results. This is particularly true if remediation periods of several decades are anticipated, which means a continuous and, ultimately, high input of energy for pumping water from extraction wells and operating water treatment systems, as well as periodic maintenance and monitoring.

In the early nineties the method of "Permeable Reactive Barriers" (PRBs) was suggested in Canada and USA. PRBs are a passive in-situ technique that runs without any need of permanent and cost-intensive operations. Once installed in the subsurface, geochemical or geophysical reactions between the material in the PRB and ground water contaminants take place without any further external interference.

Research efforts at the Institute of Geotechnical Engineering (IGT) include the development of adsorptive and reductive barrier materials. The research programme will comprise both laboratory geochemical and geotechnical tests. A full scale project is being planned at a chromate contaminated site. From geotechnical point of view, the particular challenge at this site is the great depth that is to be reached with a PRB which is at more than 40 m below surface.

2 SPECIFICATION OF THE PERMEABLE REACTIVE BARRIER (PRB)-PROCEEDINGS

Permeable Reactive Barriers are installed in the aquifer across the flow path of a contaminant plume. As the contaminated groundwater moves through the PRB due to the natural gradient, the contaminants are removed by physical, chemical and/or biological processes. Depending on what processes take place, the reactive barrier material can remain permanently in the subsurface, or replaceable units can be provided. As the reactions that occur in such systems are affected by many parameters, successful application of this technology requires a sufficient characterisation of contaminants, ground water flow regime and subsurface geology. In case of instability of the subsoil or if great depths are to be reached, techniques of specialised heavy construction are essential for the implementation (i.e. sheetpiling, contiguous bored cased piles).

Important for the decision on the feasibility of a PRB is, apart from a financial point of view, the examination of:

- Installation facility and hydraulic conductivity ($k_{PRB} > k_{Aquifer}$)
- Efficiency and clean-up performance
- Reliability and potentiality to monitor the performance of the reduction-/adsorption processes
- Hydraulic and mineralogical long-term stability
- Environmental compatibility

Currently, two basic designs are being used in full-scale implementations of PRBs. A continuous trench, filled with the reactive media, is called "*reactive wall*" or "*treatment wall*". The combination of cut-off walls and permeable in-situ reactor(s) is known as the *"funnel-and-gate" system* (EPA 1998, Gavaskar et al. 1998, NATO/CCMS 1998, Starr and Cherry 1994, Teutsch et al. 1996).

3 REACTIVE MATERIALS

Generally, the interest in PRBs has increased eminently over the past years, whereas implementations of full-scale projects have been realised especially in North America. So far, the predominant filling material has been *zero-valent iron (Fe^0)* in form of chips, jet blasting media, ironfoam or Fe-filler material for concrete etc. These materials can often be found as by-products in metalworking industry. Additionally, some highly reactive metals and metal alloys are being developed and modified with complex and expensive procedures. Fe^0 is very suitable for degrading chlorinated organic compounds (Dahmke 1997, Dahmke et al. 1999, Gillham et al. 1994, Gillham & O'Hannesin 1994, Johnson & Tratnyek 1994). Hereby, an abiotic, heterogeneous surface process occurs, which involves adsorption of the contaminant and hereafter a progressing, normally complete reductive dehalogenation of the chlorinated compound. In the end, essential degradation products are ethene or ethane, respectively. In small quantities, also some problematic low-grade chlorinated compounds, such as cis-dichloroethylene (cDCE) and vinylchloride (VC) occur as metabolic products. Generally speaking, Fe^0-reactive barriers represent an important tool for remediation of groundwater contaminated with halogenated compounds. Nevertheless, from scientific point of view, there are still several unsettled questions (e.g. degradation processes; influence of further substances in the groundwater; performance of contaminant mixtures with diverse degradation- or sorption-characteristics; processes occurring downgradient of the PRB, concerning subsequent reactors and, not to forget, water protection rights). Recent research studies show that using this degradation method, serious problems, such as clogging effects and/or inhibition of the chemical processes might occur in presence of HCO_3^-, SO_4^{2-}, PO_4^{3-} or NO_3^- or other competing oxidants (Dahmke et al. 1999). Furthermore, the influence of microbiological activities on the degradation process is widely unknown.

Zero-valent metals seem to be suitable for a series of heavy metals as well (Cr^{VI}, As^{VI}, As^{III}, Se^{VI} and TC). Soluble forms of these metals are transferred into insoluble forms by the reduction process, which then precipitate as hydroxide, for example (Blowes et al. 1996).

Significantly less distributed is the application of *adsorptive PRBs*. That is to achieve an immobilisation of the pollutants by chemically attaching them to mineral surfaces. With potable water, treatment by adsorption of organic components on solid surfaces is established. For this, activated carbon is used predominantly. Activated carbon has also been used as filling material in first applications of adsorptive PRBs (Grathwohl & Peschik 1997, Teutsch et al. 1996). This process provides just a retardation of the contaminant transport. With the reactive material showing a finite capacity, in most cases frequent replacement of the adsorptive medium must be considered.

Beside these commercially applied technologies, further ones are being developed, in order to optimise known processes as well as to develop new methods. The US Department Of Energy (US DOE) provides an extensive overview of research and application of Permeable Reactive Barriers (DOE 1998).

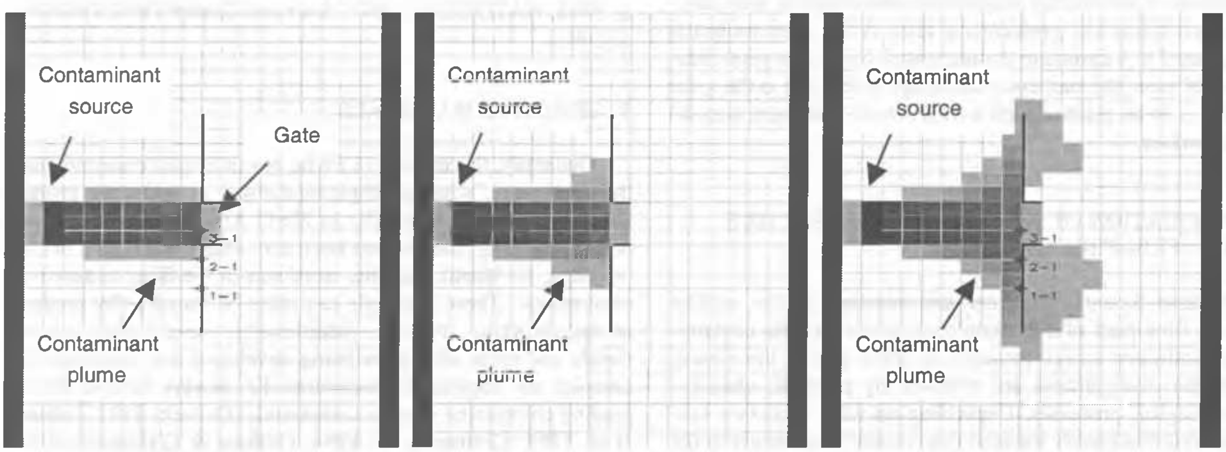
4 GEOTECHNICAL REQUIREMENTS FOR REACTIVE MATERIALS

Development of novel media to apply with Permeable Reactive Barriers (PRBs) is the objective of a present research project at the Institute of Geotechnical Engineering (IGT) at ETH Zurich. One aim is in particular to define general geotechnical criteria for PRB filling materials and carry out appropriate experiments.

Table 1. Overview of geotechnical criteria and suitable test methods

Requirements for mineral filling materials	Test method
Hydraulic and mineralogical long term stability	One-dimensional flow tests in columns for quantification of reduction / adsorption characteristics Swelling tests (with clayey materials)
Small deformations Minimal consolidation	Oedometer tests for determination of time dependent stress-deformation behaviour Stress-dependent conductivity tests in oedometer apparatuses
Erosion stability of the pellets	Use of filter materials in the columns to be examined at the end of the tests
High grain strength (abrasion behaviour)	Los-Angeles test (modified procedure)

For application in PRBs, relatively coarse grained materials are eligible, categorically, which guarantee high hydraulic permeability. To be able to use the reductive or adsorptive properties of natural or synthetic minerals for example, they must show several soil mechanical and mineralogical characteristics that are unusual and even undesirable in many conventional geo-environmental fields dealing with contaminant retention. Primarily, only such minerals are suitable that are available in fractured form without any fine grained material or in granulated form (pellets). Hydraulic and mineralogical long term stability are essential. If the hydraulic conductivity of the barrier material becomes less over time and drops below the value of the aquifer, the groundwater will tend to pass around the whole remediation system instead of passing through it. In the worst case, neighbouring areas lying laterally to the barrier system and the natural ground water flow can get polluted, (Fig. 1). The shown effect might not necessarily occur over the whole extent of the plume but might vary with depth. This must be considered planning the monitoring system of a PRB system. Clogging effects might be caused by precipitants, biomass, swelling or simply if filter criteria are not fulfilled with respect to the aquifer soil. With regard to the long term performance of a barrier material, these effects – apart from the contaminant retention/reduction



a) $k_{Gate} : k_{Aquifer} = 2:1$ b) $k_{Gate} : k_{Aquifer} = 1:2$ c) $k_{Gate} : k_{Aquifer} = 1:200$

Figure 1. Numerical simulation of the migration behaviour of a contaminant plume depending on the ratio of the reactive gate material conductivity to the aquifer conductivity (plane view)

development – must be monitored in one-dimensional flow tests (column tests) over a long period.

Table 1 gives an overview of the geotechnical criteria and a proposal of suitable test methods. In view of normal ground pressure, only small deformations should occur. Pore volume must remain stable with regard to hydraulic conductivity and flow velocity. The candidate material must not show any consolidation characteristics. To quantify the stress-deformation behaviour, oedometer tests are performed.

Thinking of the construction works, the granular barrier material will experience a high mechanical impact during transport and especially by filling the trench. With the groundwater flow, abrasives might be eroded and clog the downgradient section of the PRB or the subsequent soil. To find out about the abrasion behaviour in laboratory tests, the material is exposed to a certain amount of mechanical energy, and its loss of weight is determined afterwards. The testing method is derived from the Los-Angeles test (ASTM standard for road construction). Erosion effects are monitored in the obligatory column tests by simply using filter materials in the columns which are examined at the end of the tests.

5 COMPARISON OF THE STRESS-DEFORMATION BEHAVIOUR OF SELECTED BARRIER MATERIALS

Considering current research activities on laboratory scale as well as commercially utilized barrier materials, currently some geotechnical tests are being carried out at the IGT with respect to the feasibility in PRBs. First results from oedometer tests are shown in figure 2. Table 2 gives an overview of relations between the reactive materials and contaminants.

From Figure 2 it can be seen that activated carbon AQ30 and Coke encounter relatively high settlements (13 to 14 % at 1600 kN/m^2), activated carbon F100 little less (10%), well graded iron ore about half of the amount of coke (7%) and quartz sand as well as fine grained ore are least compressible (3 to 4 %). Transferring these results into the natural environment, the different material density must be considered, though. Assuming that the

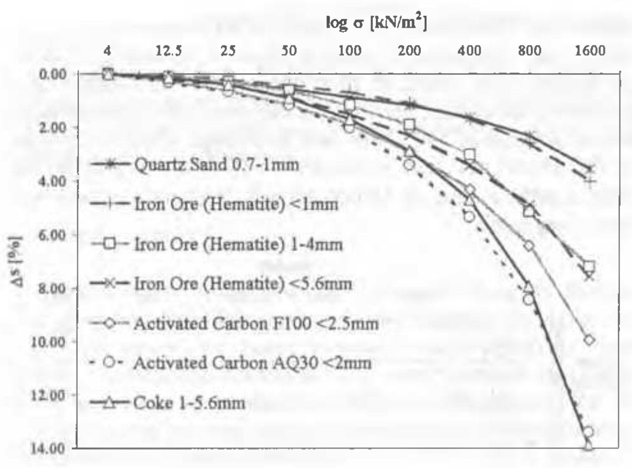


Figure 2. Results from oedometer tests with selected barrier materials and quartz sand as well known reference

Table 2. Relation between barrier materials and contaminants

PRB FILLING-MATERIAL	REFERRING CONTAMINANT CATEGORY
Iron Ore (Hematite)	Chromate (Cr^{VI})
Activated Carbon	Organic Compounds
Coke	PAH

Table 3. Unit weights of selected materials and resulting ground pressure at a depth of 10 m (buoyancy over the entire depth assumed)

PRB Filling material	Dry unit weight* γ_d [kN/m^3]	Specific unit weight γ_s [kN/m^3]	Pore-volume* n [%]	Buoyant unit weight γ' [kN/m^3]	Ground pressure at 10 m [kN/m^2]
Quartz Sand (0.7-1 mm)	15.7	26.2	40	9.8	98
Iron Ore (Hematite) (<1 mm)	31.4	48.8	36	25.1	251
Iron Ore (Hematite) (1-4 mm)	23.3	48.8	52	18.6	186
Iron Ore (Hematite) (<5.6 mm)	28.3	48.8	42	22.6	226
Activated Carbon F100	6.9	18.5	63	3.3	33
Activated Carbon AQ30	5.4	18.8	71	2.6	26
Coke	8.1	20.2	60	4.2	42

* Initial values from oedometer tests

whole pore volume is filled with water, table 3 gives a rough idea of normal ground pressure within the grain skeleton for 10m of filling level.

Depending on the reactive barrier material, a PRB reaching to a depth of 10 m would result in different ground pressure and thus different compression behaviour. For example, activated carbon F100 will encounter less than 1% settlement under a pressure of 33 kN/m^2 whereas iron ore, under a pressure of 226 kN/m^2, would settle for about 2.5%.

Hydraulic conductivity was measured at varying loading levels in the oedometer cells. With all examined materials, compression did not seem to affect hydraulic conductivity significantly.

6 PERMEABLE REACTIVE BARRIERS IN GREAT DEPTH – A SITE INVESTIGATION

Within the scope of this research project, a full scale PRB is to be installed as a demonstration object. The project is funded by the SWISS FEDERAL AGENCY FOR THE ENVIRONMENT, FORESTS AND LANDSCAPE (BUWAL) in collaboration with the INSTITUTE OF GEOTECHNICAL ENGINEERING at ETH ZURICH, GEOLOGICAL CONSULTING SERVICES DR. SCHENKER, MEGGEN (LU) and BATI GROUP AG CONSTRUCTION COMPANY, ZURICH.

The concerning site, a wood impregnations work, is located upstream of the drawoff zone of a waterworks facility in Canton Lucerne (Switzerland). Since about 1920 freshly impregnated timber had been dried and stored on the ramp and on the unsealed storage area (fig. 3). For this reason, yet in 1959/60 and again in 1979 soil- and water samples were taken to measure chromium and copper concentrations which form the main components of the impregnation substance. Both investigations showed a high contamination level of the soil with chromium and copper down to a depth of 1.2 m. Whereas copper can be toxic to fish at high concentrations, chromate is known to be toxic and carcinogen and thus is of much more environmental concern. The concentration level which indicates an impact from contaminated sites on groundwater is at 1.5 mg Cu/l and 0.02 mg Cr^{VI}/l, according to Swiss regulations.

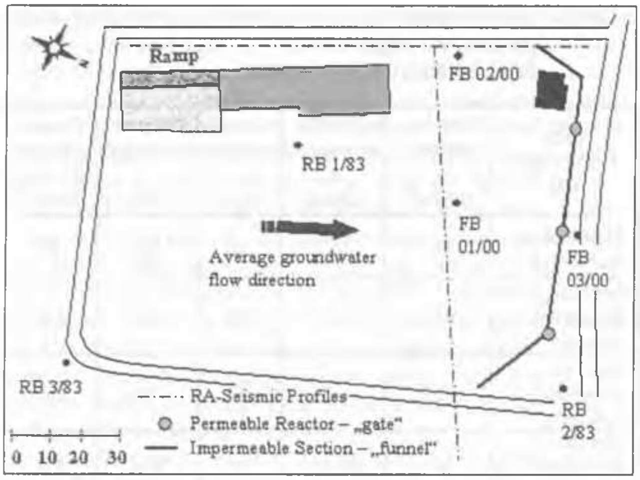


Figure 3. Possible configuration of a PRB at the wood impregnation work

In the early 1980s, chromate concentrations up to 0.023 mg/l were measured in the waterworks located about 500 m downstream of the site which is slightly above the cleanup criterion for groundwater. Maximum copper concentrations were slightly above detection level. In 1983 three screen wells were installed (RB 1-3/83) reaching down to 18 m. The groundwater level was observed to vary between 14 and 16 m below surface within the first year. The boreholes did not reach the aquitard. Surprisingly, the chromate concentrations from the screen wells were only slightly above detection level.

In order to stop further seepage of pollutants, the storage area was sealed and rainwater flowing off the area was collected separately. Only in 1999, the hotspot area around the ramp was removed down to a depth of 1.2 m.

Still assuming a certain contamination of the unsaturated zone, in summer 2000 the above mentioned organizations in arrangement with the property-owner decided to erect a PRB as part of a research project on this site. The tentative location and shape of the PRB as a funnel-and-gate system is shown in figure 3.

Successful application of a PRB requires extensive site characterisation. The following conditions have to be known:

- Soil mechanical and geochemical behaviour of the reactive material that will be applied in a PRB at a chromate contaminated site
- Macroscopic hydrogeology
- Exact elevation of the aquitard around the future funnel-and-gate location
- Ground water chemistry and background concentrations

Planning a funnel-and-gate system, a numerical groundwater model is essential. Efficient operation of the barrier system depends very much on aquifer characteristics like heterogeneities, lateral boundaries of the aquifer, elevation of the aquitard and on seasonal fluctuations of the groundwater flow as well as on the interaction of the groundwater system with the barrier structure itself.

First investigations within the scope of this project involved refraction seismic measures of the subsurface to find the aquitard and potential heterogeneities. By this, two perpendicular profiles of the subsurface were obtained, the position of which is shown in figure 3. The recorded data was confirmed by three additional boreholes (FB 01-03/00) brought down in January 2001. The aquitard was found at depths of 43 m (FB 01/00), 40.5 m (FB 02/00) and 32.2 m (FB 03/00).

Soil samples were collected from these boreholes for measuring the total chromium and copper concentration. The measurements were accomplished by AAS, the results of FB 01/00, FB 02/00 and FB03/00 are shown in figures 4, 5 and 6.

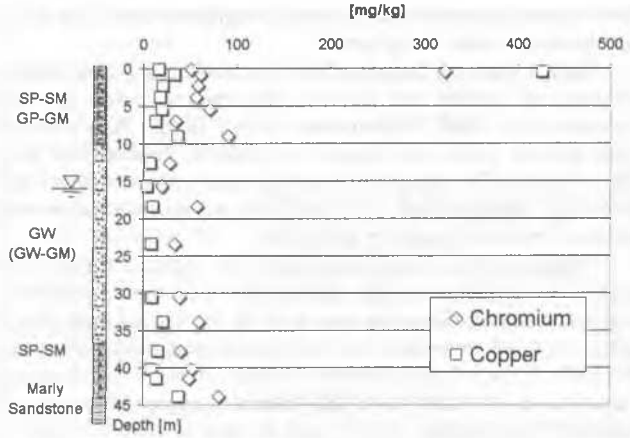


Figure 4. Concentration data of total copper and chromium from FB (01/00)

Regarding the chromium concentrations the soil can be classified as slightly contaminated (threshold value: 50 mg/kg according to Swiss Regulations for soil excavation – "Aushubrichtlinie, AHR"). Copper concentrations are below the threshold value of 40 mg/kg for copper. An exception are the remarkably high values in the very upper layer of FB 01/00 and FB 02/00, where the soil with concentrations of 322 mg/kg (Cr, FB 01/00), 427 mg/kg (Cu, FB 01/00) and up to 490 mg/kg (Cu, FB 02/00) can be characterised as heavily contaminated (threshold value: 250 mg/kg for both chromium and copper, according to the Swiss "AHR").

Returning to the principles of stress-deformation behaviour of the reactive material, an average depth of 40 m must be taken into account for normal ground pressure within the grain skeleton. First batch tests showed that iron ore is principally suitable to reduce Cr^{VI} to the harmless and rather immobile form of Cr^{III}. Considering the accessibility of the reactor within the barrier structure, the reactive material would be placed in perforated tubes in the subsurface from a depth of 40 m up to about 13 m without any (unsaturated) material above it. Assuming an average buoyant unit weight of 20 kN/m^3 (i.e. 1-4mm, taking some abrasives into account, see table 3), this results in a maximum effective pressure of 540 kN/m^2 and an average effective pressure of 270 kN/m^2 and thus settlement of 1.3% or roughly 36 cm (with a stiffness ratio of 19'000 kN/m^2). Reduction of pore volume is negligible.

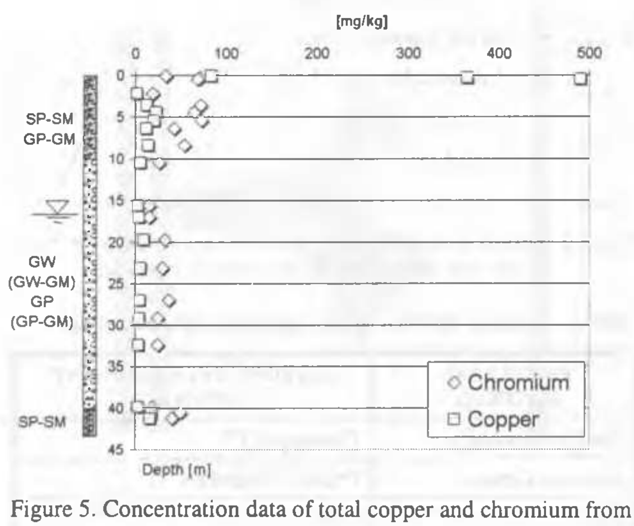


Figure 5. Concentration data of total copper and chromium from FB (02/00)

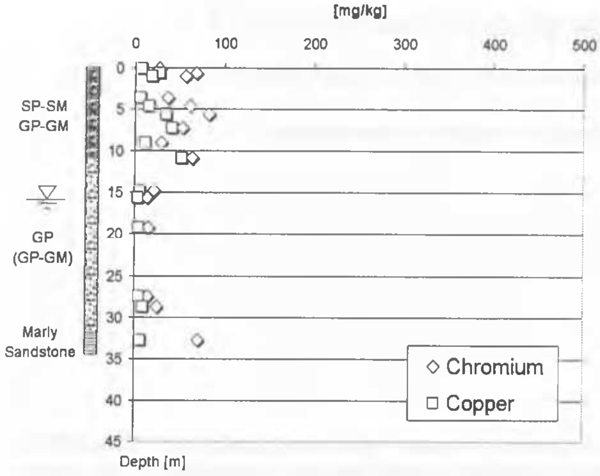


Figure 6. Concentration data of total copper and chromium from FB (03/00)

7 OUTLOOK

The described project represents an example of a PRB application in great depth. Site investigation is still far from being completed. Besides oedometer tests with further candidate reactive barrier material, abrasion behaviour will be examined. Apart from material testing, aquifer data is collected with the intention to implement a numerical groundwater model. This is essential for planning a funnel-and-gate system, as strong interaction between the subsurface structure and the groundwater flow occurs. Flow velocity within the gates will increase eminently due to a higher hydraulic gradient. This again influences the barrier design as the geochemical kinetics are affected.

To find out about the reduction-/adsorption kinetics, column tests with original site groundwater are essential. By this, concentration development can be observed along the flowpath of the columns and as a result, the necessary reactor thickness can be determined. It must be pointed out that numerical modelling must precede the column tests to be able to determine the proper flow velocity.

In addition to these theoretical considerations, methods for practical implementation of the funnel-and-gate system down to a depth of 40 m in gravel soil have to be found. It is a difficult job but a great challenge, too.

8 CONCLUSIONS

Research and development of Permeable Reactive Barriers and of appropriate filling materials in particular, as well as onsite applications, are being promoted continuously at many places. Geotechnical criteria are very rarely considered, though. With respect to applications of PRBs in great depth, the described aspects become important, thinking of efficient operation process and long term performance.

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