

# The biofiltration permeable reactive barrier: Practical experience from Synthesia

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## Abstract

The paper refers to utilization of biological elements within permeable reactive barriers. The concept of a biofiltration permeable barrier has been tested in the laboratory and in pilot-scale. Oxyhumolite (oxidized young lignite) was examined as an absorption material and a biofilm carrier. Laboratory tests performed before the pilot verification confirmed that oxyhumolite adsorbs organic pollutants at a minimum value, but that it can be used for biofilm attachment. An experimental barrier was built on premises of a chemical factory contaminated mainly by various organic pollutants [benzene, toluene, ethylbenzene, and xylenes (BTEX), chlorobenzenes, naphthalene, nitro-derivatives, phenols, trichloroethylene (TCE), and total petroleum hydrocarbon (TPH)]. Before the barrier was installed, a preliminary survey of the unsaturated zone, hydrogeological investigation, and a microbiological survey had been performed. The barrier was designed as a trench-and-gate system with an in situ bioreactor. During the year 2004, measurements of groundwater flux and retention time under current hydrological conditions, together with chemical and microbiological monitoring, were carried out on the site. The results showed high effectiveness of organic contamination removal. Average elimination varied from 57.3% (naphthalene) to 99.9% (nitro-derivatives, BTEX); microbial density in the bioreactor was approx.  $10^5$  CFU mL<sup>-1</sup>.

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## 1. Introduction

Permeable reactive barriers (PRBs) can provide cost-effective and long-term solutions for many groundwater contamination problems. They are constructed below ground to intercept groundwater flow. As contaminated groundwater moves through the reactive filling, contaminants are treated and transformed into harmless products and/or their by-products.

Up to today, more than 60 PRBs have been installed throughout the world. Most of them have been designed to treat halogenated aliphatic hydrocarbons (mainly

chlorinated) or heavy metals (e.g., hexavalent chromium). These barriers generally work on the principle of chemical reduction on a reactive medium, which is mainly zero-valent iron (e.g., Puls et al., 1995; Reardon, 1995; Orth and Gillham, 1996; Gavaskar et al., 1997). Only a small number of barriers has been installed for other pollution treatments, such as petroleum substances, aromatics, benzene, toluene, ethylbenzene, and xylenes (BTEX), or some explosives [e.g., cyclotrimethylenetrinitramine (RDX)]. These pollutants are usually treated in PRB by sorption on granulated activated carbon, or by aerobic degradation enhanced by intensive aeration in a reactor (USEPA, 2002; Alvarez et al., 2004; Dries et al., 2004; Birke et al., 2005). Special categories of these barriers are so-called air-sparging barriers, which are designed as single or more lines of injecting wells

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built across groundwater flow. These wells are used for injection of atmospheric air as an electron acceptor. In the case of extensive injection this method is also called biosparging.

Some of the first biosparging barriers were built on sites in East Garrington, Alberta, Canada, in 1995 (Bowles, 1997) and in Alameda, California, USA, in 1997 (USEPA, 2002). Biological barrier technology was also successfully applied in Europe, e.g., on sites in Italy, the Netherlands, and the United Kingdom (Bakker et al., 2000; Shields et al., 2000). During a pilot-scale application of a biological PRB on a site in Italy contaminated by various aromatic hydrocarbons, a decrease of total hydrocarbon concentration from  $8000 \mu\text{g L}^{-1}$  to less than  $200 \mu\text{g L}^{-1}$  was observed. It was also confirmed, by measurement of hydrocarbon concentrations in soil air, that the major process of contaminant removal was aerobic biological mineralization and not stripping of volatile organic substances. Further, a contamination level below  $10 \mu\text{g L}^{-1}$  was achieved by a full-scale application (Bakker et al., 2000).

At the same time as the first biosparging barriers were being developed, PRBs, working on the biofiltration principle, were also designed and tested. These biobarriers are generally designed as in situ reactor biofilter units, which contain suitable media with a bacterial biofilm. Unlike in a 'classical' PRB, pollutants are treated in biobarriers by two ways: first they are absorbed on reactive media and then specific microorganisms decompose them. As biological elements are used in the biofiltration PRB, a choice of 'bioreactive' media is considered one of the most important aspects of its design. A suitable 'bioreactive' medium has to not only fulfil all requirements of the 'classical' PRB (e.g., compatibility with underground environment, grain size, permeability, stability, low purchase price), but it must also ensure optimal growth conditions for microorganisms (i.e., it must not inhibit or negatively influence metabolic activity of microbes, and it must ensure satisfactory medium colonization and creation of an active biofilm). An important factor is also ensuring optimal conditions for growth (suitable pH, nutrient content, and, in the case of aerobic barriers, sufficient dissolved oxygen).

Technological design of biofiltration barriers (e.g., continual, funnel-and-gate or trench-and-gate) and also the type of a reactive segment (e.g., reactive gate, keson, in situ reactor) are similar to the 'classical' PRB. However, it is necessary to realize that implementation of biological elements into this technology and the necessity of ensuring optimal growth conditions for microorganisms require certain technological amendments of PRB design and can cause some difficulties, for example, while ensuring sufficient oxygen level and optimal pH. So-called 'biofouling' is caused by biofilm overgrowth and can lead to decrease of biobarrier permeability. This can further change groundwater flow and cause biobarrier overflowing (i.e., bypassing).

Practical applications of biological PRB working on the biofiltration principle have been relatively few so far. Successful realization of this PRB type can be shown, for example, in a so-called 'denitrification' biobarrier, which was composed of a reactive medium of organic carbon and a bacterial culture of *Pseudomonas* sp. The 'denitrification' biobarrier was built on a pilot-scale in Canada in 1995. The results showed that it is possible to achieve fast denitrification in this system and decrease nitrate content from concentrations typical for wastewater below limits for drinking waters (Robertson and Cherry, 1995).

This paper refers to pilot-scale testing results of a biofiltration permeable barrier. Oxyhumolite (oxidized young lignite) was used as an absorption material and a biofilm carrier.

## 2. Materials and methods

### 2.1. Tested reactive medium: oxyhumolite

Oxidized young lignite (oxyhumolite) contains up to 90% biochemically active humic substances. In the Czech Republic, one can find some of the highest-quality oxyhumolites in the world. Some oxyhumolites from North Bohemia, for example, contain more than 80% humic acids and have a low content of bitumen ( $<0.1\%$ ) and inorganic compounds (Novak et al., 2001; Vesela et al., 2005b). At present, oxyhumolites are turned mainly into sodium and potassium humates, and partially also into humic acids and humates of other metals. Owing to the low price of oxyhumolites (they are waste products from coal mining) and their ion exchange properties, it is believed that another use for them could be as sorbents for wastewater treatment or removal of heavy metals (Novak et al., 2001; Cezikova et al., 2001; Madronova et al., 2001). The fact that humic acids extracted from oxyhumolites have similar or better ion exchange properties in comparison to humic acids extracted from peat and brown coal speaks for their utilization in this field.

During laboratory work performed before the pilot testing, it was found that oxyhumolite adsorbs organic pollutants at a minimum value (Vesela, 2005; Vesela et al., 2005a). Results of the batch sorption test confirmed the generally expected assumption of a large surface area of the tested oxyhumolites (up to  $22.344 \text{ m}^2 \text{ g}^{-1}$ ). It was further found that all tested samples adsorbed model organic substances (chlorobenzenes, aniline, and nitrobenzene) to a small extent (in the range of  $0.84\text{--}22.68 \text{ mg g}^{-1}$ ) and that experimentally determined maximal adsorption capacities did not correspond to the surface area of the tested materials. Also, based on literature research, it is believed that the binding strength of oxyhumolite to organic substances is very low (mainly hydrogen bonds, Van der Waals forces) and therefore this material is not suitable as an organic contaminant sorbent (Senesi, 1992; Piccolo, 1994; Senesi et al., 1995; Vesela et al., 2005b). Thus it was concluded that sorption of organic substances on oxyhumolites can be considered as low, compared to oxyhumolite sorption ability for heavy metals (Cezikova et al., 2001; Madronova et al., 2001), and will be neglected during further testing.

The results of laboratory biofiltration experiments showed the possibility of using oxyhumolite as the biofilm carrier (Siglova et al., 2004; Vesela, 2005; Vesela et al., 2005a). It was also confirmed that oxyhumolite is not suitable as the carrier itself, mainly because of its ability to decrease pH, and therefore it is necessary to combine this material with limestone.

### 2.2. Pilot-scale testing

The biofiltration PRB was built on the premises of a chemical plant called Synthesia. The company was established in 1920 and is situated in a

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