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DZIAŁANIE PRZEPUSZCZALNEJ BARIERY AKTYWNEJ (SYSTEM "FUNNEL AND GATE") Z ZASTOSOWANIEM BIOREAKTORA NA TERENIE BYŁEJ FABRYKI SMOŁY W OFFENBACH W NIEMCZECH

OPERATION OF A FUNNEL&GATE SYSTEM WITH BIOREAKTOR AT THE SITE OF A FORMER TAR PLANT IN OFFENBACH, GERMANY

STRESZCZENIE

Pełnowymiarowa biobariera w systemie „funnel – gate" (co można przetłumaczyć jako "parkan – bramka") została zastosowana na terenie byłej fabryki smoły w Offenbach w Niemczech w celu usunięcia zanieczyszczenia wód podziemnych olejami smołowymi. Budowa i pierwszy okres funkcjonowania przepuszczalnej bariery aktywnej w systemie „parkan – bramka" stanowiły część sieci badawczo-rozwojowej RUBIN.

Zastosowana metoda okazała się skutecznym sposobem ograniczenia zanieczyszczenia terenu. Pomyślność bariery „parkan – bramka" była wynikiem wnikliwych badań laboratoryjnych i terenowych, innowacyjnej konstrukcji bioreaktorów oraz bieżącej kontroli systemu i reakcji na jego działanie.

Pełnowymiarowa biobariera składa się z warstwowego separatora, w którym osadza się wytrącone żelazo, oraz trzech bioreaktorów wypełnionych żwirem. Bioreaktory oddzielone są stalowymi ekranami od stref „wolnej wody". W strefach tych do wody dodaje się nadtlenek wodoru H_2O_2 i substancje odżywcze w celu wzmocnienia procesu biodegradacji zanieczyszczeń. Działanie systemu „parkan –

bramka" z 30-sto metrowej długości parkanami po obydwu stronach rozpoczęło się w kwietniu 2007 r.

Pilotażowe badania terenowe potwierdziły wyniki eksperymentów laboratoryjnych oraz testów kolumnowych przeprowadzonych na terenie fabryki („on-site"). W obrębie „bramki", ponad 99% wszystkich obserwowanych na dopływie zanieczyszczeń organicznych zostało wyeliminowanych. W związku z uzyskanymi wynikami okazało się niepotrzebnym oczyszczanie wody na odpływie z reaktora przy użyciu węgla aktywnego, co pierwotnie zakładano. Ponadto, wydaje się, że przedawkowywanie azotu w reaktorze może przynosić wymierne korzyści w stymulowaniu wspomaganego samooczyszczania wody (ENA) na odpływie z "bramki".

Podczas fazy testów zidentyfikowano i zoptymalizowano najważniejsze czynniki kontrolne. Tym samym udowodniono, iż system jest wrażliwy na zróżnicowanie przepływu przez "bramkę", a także ilość dodanego nadtlenku wodoru H_2O_2 i azotu. Zaletą okazało się również utrzymywanie prędkości dopływu wody do "bramki" na, w miarę możliwości, stałym poziomie poprzez pompowanie wody gruntowej z pierwszej strefy "wolnej wody" do warstwowego separatora.

Ponadto, wyprzedzająco w stosunku do planowanej rozbudowy systemu w 2017 r., na potrzeby stadium wykonalności, przeanalizowano i oceniono szereg alternatywnych rozwiązań, takich jak przedłużenie „parkanu", czy zastosowanie dwóch „bramek".

SUMMARY

A full scale funnel-and-gate biobarrier was developed for the removal of tar oil pollutants in the groundwater at an abandoned tar factory site in the city of Offenbach, Germany. The development, construction and first period of operation of the funnel-and-gate system were part of the RUBIN research and development network.

The funnel-and-gate system has demonstrated its efficiency and is part of a reliable containment of the contaminated site. The successful performance was based on careful laboratory and field tests, an innovative reactor design and the active operational concept.

The full scale biobarrier consists of a lamella separator for the sedimentation of the precipitated iron and three bioreactors filled with fine gravel, which are separated by steel screens from open-water areas. In these open-water zones H_2O_2 is added to enhance the *biodegradation of the pollutants. The operation of the funnel-and-gate biobarrier with a 30-m long funnel on each side of the gate started in April 2007.*

The pilot field test confirmed the main results of the laboratory experiments and the on-site column experiment. Within the gate, more than 99% of all monitored influent organic contaminants were eliminated. Due to these results, a treatment of the reactor outflow by activated carbon as a security barrier is not necessary. Moreover, it seems to be promising to stimulate ENA processes in the outflow area of the gate by overdosing with nitrate in the reactor.

During the test phase the most important controlling means were identified and optimized. Thereby the system proved to be sensitive to variations of the flow rate through the gate as well as the amount of H_2O_2 and nutrients added. It turned out to be an advantage to keep the inflow into the gate as constant as possible by actively pumping the groundwater from the first open-water zone into the lamella separator at a defined flow rate.

In advance of the planned system expansion in 2017, in the context of a feasibility study, various alternatives (e.g. length of the funnel extensions, two gates) were identified and evaluated.

1. Introduction

The funnel-and-gate system is located on the side of a former tar factory adjacent to the river Main in the city of Offenbach, Germany, which was in operation from 1914 until 1929. The area of the former property was about 15,000 m². After the termination of tar production, the buildings were demolished. During and after World War II the site was used as a dumping area for construction waste and rubble.

In 1991 site investigations started. About two years later in 1993 the regulatory authority (Regierungspräsidium Darmstadt) declared the site to be an official contaminated site (Altlast) and transferred it to the HIM-ASG for remediation. The HIM GmbH is responsible on behalf of the land Hesse for the remediation of contaminated sites in those cases, where responsible parties can not at all or not in time be charged with the cost for remediation.

Since 1994, HIM-ASG has conducted extensive soil and groundwater investigations and established a grid of groundwater monitoring wells in the Quaternary-, Tertiary- and Rotliegend sediments.

As a consequence of past activities, the Quaternary subsoil and aquifer was massively contaminated with tar oil, which also appears partly in phase. This tar oil especially spread at the base of the aquifer. A large plume of dissolved contaminants extends in the saturated zone down-gradient from the hot spot area. There is also pollution in the Tertiary aquifer.

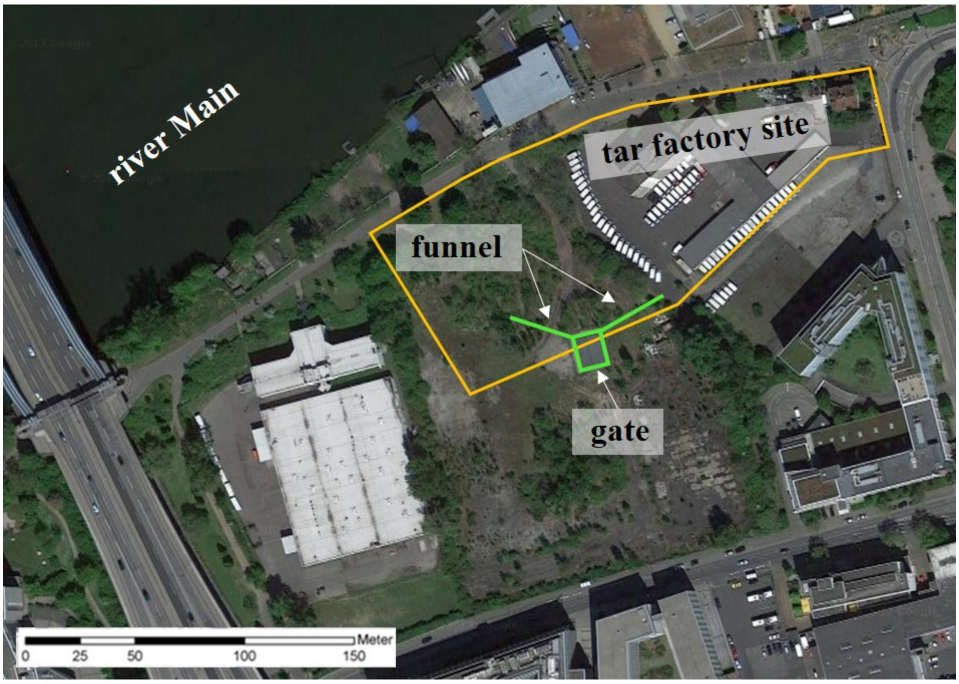
During the screening and development of remedial alternatives in 1997, the options excavation, containment and pump and treat were considered. In 1998 the planning of a funnel-and-gate system was initiated.

As part of the RUBIN research and development network focused on permeable reactive barriers (PRB), a funnel-and-gate system was developed and constructed at the site. This system is intended to be used as a hydraulic containment for the contaminated upper aquifer.

Originally the concept for water clean-up in the gate was based on a three-step process involving sedimentation of ferric iron, aerobic biodegradation of the aromatic hydrocarbons in bioreactors, and a subsequent zone with granular activated carbon (GAC) removing remaining pollutants.

In the bioreactor, the availability of suitable electron acceptors and nutrients had to be considered as an important factor affecting biodegradation efficiency. Due to the high pollutant concentrations in the groundwater, hydrogen peroxide (H_2O_2) was selected as oxygen carrier due to its higher water solubility as compared to oxygen. Additionally, nitrate and phosphate were added as an alternative electron acceptor and as a nutrient, respectively.

Laboratory and pilot scale experiments were conducted to determine the site-specific sequence and efficiency of pollutant biodegradation, the optimum locations and concentrations for hydrogen peroxide addition, and the suitable operating parameters for the in-situ biobarrier. Based on the results of the preliminary tests, between October 2006 and March 2007 a large-scale pilot plant with one gate and two funnel wings (each 30 m long) was designed and installed on the site. The system has been operated successfully since April 2007.



Rys.1. Mapa terenu z lokalizacją system "parkan – bramka"

Fig. 1. Site map with location of the funnel & gate-system

2. Laboratory and onsite columns tests

Laboratory column experiments were carried out over a period of 300 days. The column experiments were performed using groundwater samples taken from a well up-gradient of the projected reactor. The samples of the groundwater were filled into 30-liter Teflon bags enclosed in nitrogen flushed vessels and stored under anaerobic conditions. The laboratory column (d = 25 cm, length 100 cm, volume 12.7 L) was filled with silica sand (grain size 1 – 2.5 mm) and flowed through from top to button. In order to remove precipitated ferric iron, reverse flushing was performed discontinuously. H_2O_2 and nitrate were added at the column inlet. During column operation, a second addition port for H_2O_2 was installed in the upper part of the column.

Pilot tests were carried out to test process steps (sedimentation, microbiological degradation, degasification, adding electron acceptors and nutrients, activated carbon adsorption) and to identify relevant parameters for the design of the reactor under in-situ conditions,.

The pilot-test columns were operated on-site in an air-conditioned container. Two bioreactors (stainless steel columns, filled with silica sand) were operated with groundwater taken from a well on the site. A sedimentation tank with a lamella separator to remove ferric iron after the first H_2O_2-addition was incorporated in front of the bioreactors. H_2O_2 was added to the sedimentation tank and the two bioreactors. Nitrate was added to the first bioreactor. Phosphate was dosed discontinuously at both bioreactors. A GAC column was installed at the bioreactor effluent to remove remaining pollutants. The on-site column experiments were operated for a period of 270 day.

The microbiological investigations at the Offenbach site had revealed that sufficient numbers of microorganisms capable of degrading BTEX and PAH under aerobic conditions were present in the contaminated soil and groundwater. Denitrifying bacteria were also detected. Therefore, additional inoculation of allochthonous microorganisms was not required to establish aerobic and denitrifying consortia in the bioreactor.

In addition laboratory tests were conducted to select an acceptable type of activated carbon considering hydraulic, sorptive and economical aspects.

The result of the laboratory tests and the pilot plant field study confirmed the feasibility of the biobarrier system.

3. Design and Construction of the Pilot Gate

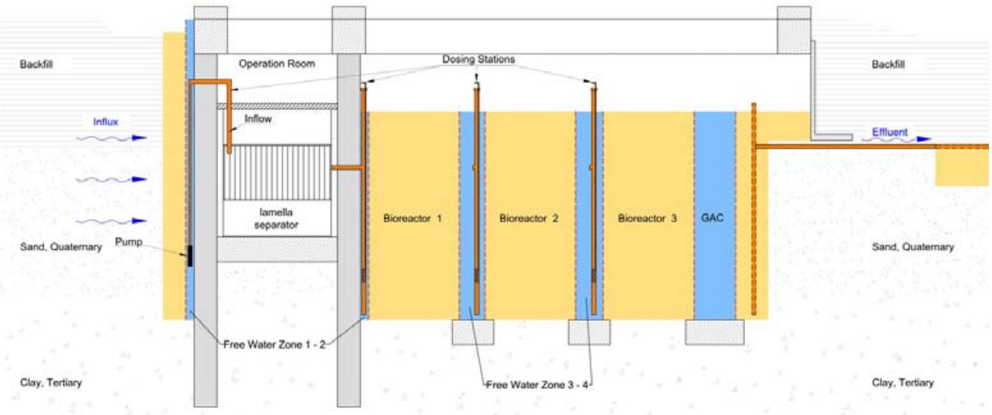
The results of the laboratory and field tests led to the general design of the pilot gate. The construction of the gate with a 30-m long funnel on each side was completed in April 2007 and the operation of the funnel-and-gate biobarrier started. For the construction of the funnel, a mixed-in-place construction technique was used.

The modular design of the bioreactor with three separated bioreactors follows the concept of permeable reactive barriers with extended control (EC-PRB) instead of passive, less controllable systems. The reactor consists of a sedimentation zone, 3 bioreactors and a GAC-zone (figure 2) .

The different zones are separated by free-water zones with dosing ports for the addition of H_2O_2 and nutrients to achieve complete degradation of the organic contaminants.

The free-water zones and the GAC-zone are built up of two rows of perforated I-beams to allow cross-flow. Inflow and outflow chambers of the free-water zone are separated by sheets of HDPE. Water leaves the inflow chamber via an outlet, which is also used as a dosing station, to reach the outflow chamber. Bridge slot filters were mounted on the I-beams as limits of the bioreactors.

The contaminated groundwater is directed to the gate by the two funnel wings and is collected from the aquifer in a first free-water zone with an upstream gravel filter. Within the free-water zone 1 the groundwater flow is directed to a connecting pipe from where the groundwater flows to the sedimentation zone (lamella separator, 200 m² sedimentation area). In the sedimentation stage, iron is extracted from the groundwater using oxidation and precipitation in a plate clarifier in order to prevent blocking in the later bioreactor levels. As oxidation agent hydrogen peroxide is dosed into groundwater at the inlet to the plate clarifier (dosing port 1).



Rys.2. Przekrój podłużny przez system biobarier

Fig. 2. Longitudinal section of the biobarrier system

After leaving the lamella separator the groundwater passes to the second free-water zone where hydrogen peroxide are added (dosing port 2). In the free-water zone uniform mixing between H_2O_2 and groundwater and the distribution to the entire cross-section is achieved. From there the water is released to the first of in total three bioreactor.

Each bioreactor is filled with about 50 m^3 of fine to coarse gravel (grain size 2 – 8 mm) which serves as a medium on which the microorganisms can grow. Assuming a porosity of 35% in the bioreactors and a flow rate of 300 L/h, an average retention time in each stage of 4 – 5 days can be calculated.

Due to the planned system expansion, the system constructed included two parallel identical free-water zone and bioreactor system, from which one part is active (contaminated groundwater flows through it) and the other part inactive (no water flows). The two systems can connect together by removing membrane sheets, which are separated the two parts.

4. Test Phase

During the test phase (May 2007 – September 2009) sampling was done on the basis of a detailed monitoring program. The samples for the monitoring of the reactor performance were taken from monitoring wells some meters upgradient and downgradient of the gate, from all free water zones in front of the dosing ports, from sampling wells in the bioreactor 3 and downgradient of the gate. The samples in the gate were taken with low flow pumps in order to minimize the disturbance of the system by the sampling process.

Monitoring parameters were BTEX, PAH, heterocyclic compounds, dissolved iron, redox conditions, numbers of microorganisms, and toxicity.

The zone which was foreseen to be filled with activated carbon remained empty during the adaptation phase of the bioreactors, thus being another open water zone. If the bioreactors had not been able to degrade contaminant concentrations sufficiently to reach the compliance values of the authorities, activated carbon would have been filled into the last chamber of the reactor to remove the remaining pollutants after bioreactor 3.

5. Operation Phase and Experiences

5.1. Establishing microbiological processes and dosing

To stimulate the aerobic and aerobic/denitrifying degradation, oxygen and nitrate are added. Nitrogen, which is another essential nutrient for microorganisms, was available from ammonium which is naturally found in the groundwater.

As carrier of O_2, a H_2O_2-solutions of 10% and 35% and Na-nitrate were added.

The column test studies showed that the successful adaption of the aerobic/denitrifying degradation needs a step by step increase of dosing rates.

Establishment of the microbiological processes took place from the first to the last bioreactor, following the impact by pollutants and microorganisms. Degradation first started in the sedimentation unit and biorector 1. After an operation time of 600 days, significant microbiological activity was observed in bioreactor 2.

A constant degradation of residual pollutants could be observed after increasing the flow rate and optimizing the nutrient dosage. After 800 days of degradation processes, the complete system was established.

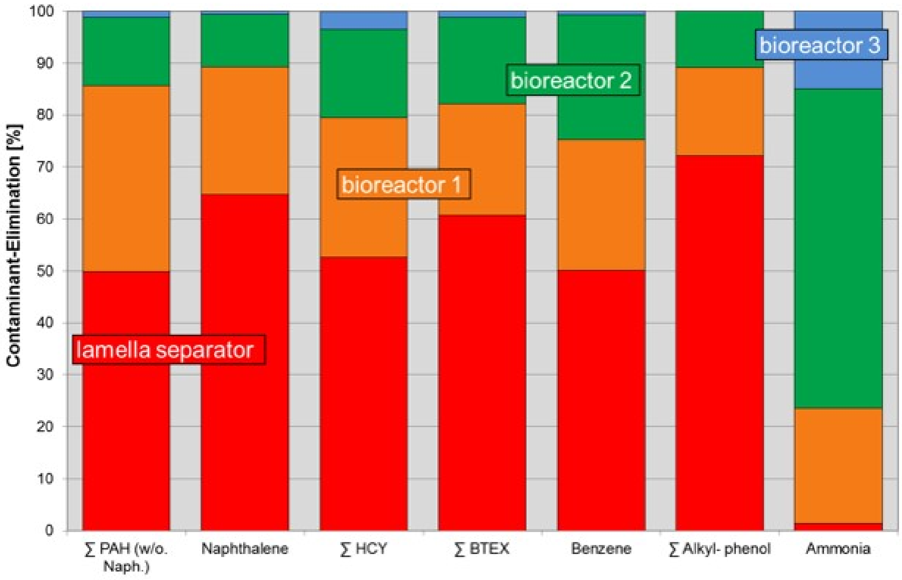
5.2. Operation of the bioreactor

Given the small and naturally varying groundwater flow rates through the gate under normal site conditions, the active pumping turned out to be an advantage because the flow rates could be kept constant or varied in defined steps. A constant input flow rate also allows constant dosing rates and degradation rates, and it was assumed that by this the operational cost could be reduced. During the testing phase this operational mode allowed the monitoring of microbiological degradation processes under constant inflow conditions and the study of the influence of variations of the dosing rates of H_2O_2 and the nutrients. Based on these results, a better evaluation of degradation processes and the effectiveness of the whole system were possible.

The advantages of the modified system are better controllability and more flexibility, which allows the modification of flow rates and the adaption to changing hydraulic conditions.

5.3. Degradation performance

The contaminant removal along the individual process steps in the gate is shown in figure 3. The aerobic stimulation already begins in the precipitation unit. Significant amounts of BTEX, naphthalene, heterocyclic compounds (HCY) and other aromatic compounds (about 50-70%), other PAH (about 50%) are eliminated (or transformed) there. The biological degradation is indicated by a significant increase in the number of pollutant-degrading bacteria. In the bioreactors 1 and 2, a sequential aerobic and denitrifying degradation takes place. Almost all remaining BTEX and other AHC, PAH, and HCY are degraded. The degradation of ammonia mainly occurs in the second bioreactor. In bioreactor 3 the remaining contaminants are eliminated (figure 3).



Rys.3. Spadek stężenia zanieczyszczeń organicznych I amoniaku w "bramce" w 2016

Fig. 3. Decrease of the concentration of organic pollutants and ammonia in the gate on 2016

Tablica 1. Wydajność eliminacji zanieczyszczeń w bioreaktorach po całkowitym dostosowaniu procesów mikrobiologicznych w "bramce" (2016)

Table 1. Elimination performance of the bioreactors after the complete adaptation of the microbial processes in the gate in 2016

	untreated water	**lamella separator**		**bioreactor 1**		**bioreactor 2**		**bioreactor 3**		**total**	**clean-up target value**
parameter	[µg/l]	effluent conc. [µg/l]	reduction [%]	effluent conc. [µg/l]	reduction [%]	effluent conc. [µg/l]	reduction [%]	effluent conc. [µg/l]	reduction [%]	reduction [%]	conc. [µg/l]
∑ PAH (w/o. Naph.)	280	141	50	40	36	3,4	13	0,0	1	100,0	2
naphthalene	2.325	819	65	250	24	14,7	10	0,3	1	100,0	10
∑ HCY	505	239	53	103	27	18,0	17	0,7	3	99,9	-
∑ BTEX	2.694	1.060	61	479	22	32,1	17	0,1	1	100,0	120
benzene	1.575	787	50	391	25	11,9	24	0,1	1	100,0	10
∑ alkyl-phenol	683	190	72	74	17	0,2	11	0,1	0	100,0	80

The GAC-zone (after the third bioreactor) was initially conceived to retain contaminants that degrade poorly. However, the results show that the biological removal has been well established within the system. The overall reduction of the organic contaminants is better than 99.9 %. All concentrations in bioreactor 3 were below the compliance values (Table 1). Therefore, a treatment of the outflow of bioreactor 3 by activated carbon is not necessary.

6. Full-scale expansion and commissioning of the inactive part

The lessons learned in operating the system were incorporated into the subsequent design of the remediation measures. In this context the technical and hydraulic feasibility of different alternatives was investigated. In order to capture the entire groundwater discharge from the former industrial site, it is planned to extend both guide walls of the existing Funnel-and-Gate System in 2017. Due to the very good degradation efficiency it will not be necessary to install a second gate, as was originally planned. Groundwater flow to the gate will be hydraulically supported by wells located at the ends of the extended funnel walls.

In addition to an increase in the amount of water flowing through the gate, the system expansion also involves an increase in the quantities of pollutants to be removed/transformed.

The system constructed in 2007 included two parallel identical free-water zone and bioreactor system, from which one part is active (contaminated groundwater flows through it) and the other part inactive (no water flows). The inactive part could be utilized if required. In the current layout the two systems (active part and inactive part) are separated by membrane sheets. Removing the membranes will connect the two systems together.

For the full-scale expansion of the system, the inactive part was put into operation at the end of 2015. From the experience of the pilot phase, it could be deduced that the commissioning will take about half a year. During this period, the microorganisms in the bioreactors of the second part can become established to such an extent that there is also an almost complete reduction of pollutants. The commissioning of the second part was completed in April 2016. The construction of the extended funnel walls is planned to start in mid-2017.

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