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ACID MINE DRAINAGE CLEANUP IN A URANIUM DEPOSIT BY MEANS OF A PASSIVE TREATMENT SYSTEM

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Acid drainage waters generated in the Curilo uranium deposit, Bulgaria, were treated by means of a pilot-scale multibarrier with a total volume of 23 m³ and consisting of an alkalizing limestone drain and an anoxic section for microbial dissimilatory sulphate reduction, biosorption and additional chemical neutralization. The waters had a pH in the range of about 2.5 – 4.0 and contained radionuclides (U, Ra), heavy metals (Cu, Zn, Cd, Pb, Ni, Co, Fe, Mn), arsenic and sulphates in concentrations usually much higher than the relevant permissible levels for waters intended for use in agriculture and/or industry. The water flow rate through the multibarrier varied in a large range (approximately from 1 to 17 m³/24 h), reflecting water residence times of about 300 – 18 hours. An efficient removal of pollutants was achieved by the multibarrier during different climatic seasons, even during cold winter days at external air and water temperatures close to 0 °C. However, some essential changes in the composition and properties of the multibarrier arised during the 28 months since the start of its operation.

Key words: drenage, cleanup, uranium, waters

INTRODUCTION

The acid mine drainage waters generated in the Curilo uranium deposit, Bulgaria, are a heavy environmental problem, especially since the end of the mining operations in this deposit sixteen years ago. The fractured ore body and several dumps consisting of mining wastes are, especially after rainfall, the main sources of these waters. The waters have a low pH (usually in the range of 2.5 – 4.0) and contain radionuclides, heavy metals, arsenic and sulphates in concentrations usually much higher than the relevant permissible levels for waters intended for use in the agriculture and/or industry. The solubilization of these pollutants from the residual ore in the deposit is connected mainly with the oxidative activity of the indigenous acidophilic chemolithotrophic bacteria [Groudev et al., 2003].

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Since the summer of 2004 a portion of the above-mentioned polluted waters is treated by means of a multibarrier constructed in the deposit. The structure of this multibarrier and its work have been described in several papers, the most recent of which are those by Groudev et al., [2006a, 2006b]. The present paper summarizes the data obtained during the 28 months of multibarrier operation and contains the main conclusions based on these data.

MATERIALS AND METHODS

The multibarrier was constructed in a ravine collecting a portion of the acid drainage waters generated in the deposit. The multibarrier was a pond dug into the ground and its bottom and walls were isolated by an impermeable plastic sheet. The multibarrier consisted of two sections: an alkalizing limestone drain and an anoxic section for microbial dissimilatory sulphate reduction, biosorption and additional chemical neutralization. The alkalizing drain had a volume of about 2.5 m³ and was filled by a mixture of crushed limestone and gravel pieces (in a ratio of about 1:2 as dry weight) with a particle size less than 12 mm. The second section of the multibarrier had a volume of about 20.4 m³ (8.0 m long, 1.7 m wide, and 1.5 m deep) and was filled by a mixture of biodegradable solid organic substrates (cow manure, plant compost, straw), crushed limestone and zeolite saturated with ammonium phosphate. This section of the multibarrier was inhabited by a microbial community consisting mainly of sulphate-reducing bacteria and other metabolically interdependent microorganisms (Table 1).

Table 1. Microorganisms in the acid mine drainage and in the effluents from the permeable reactive multibarrier

Microorganisms	In the acid mine drainage	In the multibarrier effluents
	Cells/ml	
Fe ²⁺ -oxidizing chemolithotrophs (at pH 2)	10 ⁴ – 10 ⁷	0 – 10 ²
Aerobic heterotrophs (at pH 2)	10 ¹ – 10 ⁴	10 ¹ – 10 ³
S ₂ O ₃ ²⁻ -oxidizing chemolithotrophs (at pH 7)	0 – 10 ³	10 ¹ – 10 ⁴
Aerobic heterotrophs (at pH 7)	0 – 10 ²	10 ¹ – 10 ⁴
Anaerobic heterotrophs (at pH 7)	0 – 10 ¹	10 ⁴ – 10 ⁷
Sulphate-reducing bacteria	0 – 10 ¹	10 ⁴ – 10 ⁷
Cellulose-degrading microorganisms	ND	10 ³ – 10 ⁶
Bacteria fermenting sugars with gas production	ND	10 ³ – 10 ⁷
Ammonifying bacteria	ND	10 ² – 10 ⁵
Denitrifying bacteria	ND	10 ² – 10 ⁵
Fe ³⁺ -reducing bacteria	ND	10 ³ – 10 ⁶
Methane-producing bacteria	ND	10 ¹ – 10 ⁴

Note: ND = not detected

The quality of the waters was monitored at different sampling points located at the inlet and the outlet of the alkalizing drain and of the section for dissimilatory sulphate reduction and biosorption, as well as at different depths within these two sections of the multibarrier.

The parameters measured in situ included: pH, Eh, dissolved oxygen, total dissolved solids and temperature. Elemental analysis was done by atomic adsorption spectrophotometry and induced coupled plasma spectrophotometry in the laboratory. The radioactivity of the samples was measured, using the solid residues remaining after their evaporation, by means of a low background gamma-spectrophotometer ORTEC (HpGe – detector with a high distinguishing ability). The specific activity of ^{226}Ra was measured using a 10 dm³ ionization chamber.

Elemental analysis of solid samples from the sediments and the plant biomass was performed by digestion and measurement of the ion concentration in solution by atomic adsorption spectrophotometry and induced coupled plasma spectrophotometry. Mineralogical analysis was carried out by X – ray diffraction techniques. The mobility of the pollutants was determined by the sequential extraction procedure [Tessier et al., 1979]. The fraction analysis of the solid organic component in the multibarrier was performed using air dried samples after their prior washing with distilled water to remove the carbonates and the water – soluble organic compounds. The total amount of crystalline polysaccharides (cellulose) and non-crystalline polysaccharides (hemicellulose) was determined by hydrolysis with sulphuric acid [Ryan et al., 1990]. The non-crystalline polysaccharides (hemicellulose) were determined by hydrolysis with hydrochloric acid under anaerobic conditions [Lowe, 1993]. For determination of lignin the samples were initially treated by 1 M HCl for 12 h to remove the solid precipitates from the surface of the organic matrix and to make it well exposed for the subsequent solubilization of the lignin. This solubilization was performed by means of solution containing 2 M NaOH, CuO, Fe(NH₄)₂(SO₄)₂·6H₂O under anaerobic conditions, at 170 °C for a period of 4 h in the Soxhlet apparatus [Kogel & Bochter, 1985]. The suspensions of dissolved organic compounds obtained by the above-mentioned hydrolytic treatments were clarified by centrifugation at 1700 rpm for 30 min. The clarified supernatants were used for determination of the concentration of the respective dissolved organic compounds by their chemical oxidation to CO₂ at high temperature.

The isolation, identification and enumeration of microorganisms were carried out by methods described elsewhere (Karavaiko et al., 1988; Widdel & Hansen, 1991; Widdel & Bak, 1991; Groudeva & Tzeneva, 2001; Hallberg & Johnson, 2001).

RESULTS AND DISCUSSION

An efficient removal of pollutants from the acid drainage waters was achieved by means of the multibarrier (Table 2). The water flow rate through the multibarrier was changed considerably during the operation period (within the range of about 1 – 17

m³/24 h, reflecting water residence times of about 300 – 18 hours). These changes were connected mainly with the activity and permeability of the multibarrier and with the level of pollution of the waters being treated.

Table 2. Data about the acid mine drainage and the effluents from the permeable reactive multibarrier

Parameters	Acid mine drainage	Multibarrier effluents	Permissible levels for waters intended for use in agriculture and industry
Temperature °C	(+1.2) - (+25.1)	(+1.4) - (+27.5)	-
pH	2.42 – 4.25	6.22 – 7.83	6 – 9
Eh, mV	(+290)-(+597)	(-140)-(-280)	-
Dissolved O ₂ , mg/l	1.7 – 6.0	0.2 – 0.4	2
TDS, mg/l	930 – 2972	545 – 1827	1500
Solids, mg/l	41 – 159	32 – 104	100
DOC, mg/l	0.5 – 2.1	51 – 159	20
SO ₄ ²⁻ , mg/l	532 – 2057	275 – 1225	400
U, mg/l	0.10 – 2.75	< 0.05	0.6
Ra, Bq/l	0.05 – 0.50	< 0.03	0.15
Cu, mg/l	0.79 – 5.04	< 0.20	0.5
Zn, mg/l	0.59 – 59.8	< 0.20	10
Cd, mg/l	<0.01 – 0.10	< 0.004	0.02
Pb, mg/l	0.08 – 0.55	< 0.02	0.2
Ni, mg/l	0.17 – 1.49	< 0.03 – 0.10	0.5
Co, mg/l	0.12 – 1.22	< 0.03 – 0.10	0.5
Fe, mg/l	37 – 671	0.5 – 9.5	5
Mn, mg/l	2.8 – 79.4	0.5 – 5.2	0.8
As, mg/l	0.05 – 0.32	< 0.01	0.2

The activity of the multibarrier was based on three main mechanisms participating in the removal of pollutants: chemical neutralization, biosorption and microbial dissimilatory sulphate reduction. The chemical neutralization was performed by the crushed limestone mainly in the alkalizing drain but also in the second, rich-in-organics section. In the alkalizing drain the pH of the polluted waters was increased to values near the neutral point and as a result of this most of the dissolved iron (present as Fe³⁺ ions) was precipitated as ferric hydroxides. Portions of the non-ferrous metals and aluminum (usually about 20 – 40 % and about 70 – 80 %, respectively) were also removed in the drain as a result of hydrolysis and subsequent precipitation as well as by sorption by the gelatinous ferric hydroxides. Portions of uranium and arsenic were also removed in this way (Table 3). The increase in pH in the alkalizing drain facilitated the growth of microorganisms in the second section of the multibarrier. The role of chemical neutralization was essential during the cold winter days when the growth and activity of the microbial community in the multibarrier was inhibited or

completely ceased. The chemical generation of alkalinity steadily decreased in the course of time because the limestone was armored due to the iron precipitates deposited on its surface.

Table 3. A typical example for the changes in the composition of the polluted waters during their treatment in the multibarrier

Parameters	AMD before treatment	Alkalizing drain	Zones in the section for MDSR		
			2 m from the inlet	4 m from the inlet	6 m from the inlet
pH	2.96	5.06	5.63	6.10	6.28
Eh, mV	+ 467	+ 320	+145	- 45	- 214
Acidity, mmol/l	3.8	2.2	0.9	-	-
DOC, mg/l	-	-	135	140	98
Cu, mg/l	50.4	3.73	0.50	0.21	0.05
Zn, mg/l	59.8	13.02	2.62	0.57	0.50
Cd, mg/l	0.04	0.02	0.02	< 0.005	< 0.005
Pb, mg/l	0.16	0.14	< 0.03	< 0.03	< 0.03
Ni, mg/l	1.43	1.35	1.05	0.70	0.008
Co, mg/l	1.10	0.88	1.03	0.24	< 0.005
Fe, mg/l	128	1.81	0.27	0.34	0.55
Mn, mg/l	19.8	20.8	21.5	15.4	5.24
U, mg/l	1.23	0.34	0.07	0.05	0.03
SO ₄ ²⁻ , mg/l	1990	1870	1810	1650	680

Table 4. Content of pollutants in the dead solid plant biomass in the permeable reactive multibarrier

Pollutants	Content, mg/kg dry biomass				
	November 2004	March 2005	August 2005	March 2006	September 2006
Uranium	10 – 32	17 – 71	23 - 88	32 – 114	32 – 122
Radium	5 – 14	10 – 32	15 - 41	21 – 53	23 – 57
Copper	28 – 73	37 – 134	44 - 181	60 – 225	62 – 221
Zinc	14 – 51	28 – 82	37 - 190	51 – 230	59 – 242
Cadmium	2 – 12	6 – 19	6 - 27	8 – 41	8 – 44
Lead	8 – 30	7 – 59	10 - 64	15 – 73	15 – 77
Nickel	8 – 35	9 – 62	9 - 77	15 – 90	19 – 95
Cobalt	5 – 30	11 - 51	14 - 70	17 – 82	16 – 90
Manganese	32 – 109	37 - 135	44 - 190	51 – 210	55 – 233
Arsenic	2 - 14	6 - 23	10 - 31	12 – 44	12 – 51

The biosorption of pollutants by the dead plant biomass present in the second section of the multibarrier was also an essential mechanism in the water clean up during the warmer but mainly during the cold months of the year (Table 4).

Considerable portions of all heavy metals, arsenic and uranium as well as most of the radium were removed in this way. The biosorption, together with the chemical neutralization, was the prevalent mechanism during the first 5 – 6 months since the start of the operation and even later was prevalent in the first 3 – 4 m from the inlet of the anoxic section, especially in the top layers (down to 50 – 70 cm from the surface). The sorption capacity of the dead biomass steadily decreased in the course of time.

The microbial dissimilatory sulphate reduction played an essential role in the water clean up during the warmer months of the year. The anaerobic sulphate-reducing bacteria were a quite numerous and diverse population in the multibarrier (Table 5). The prevalent and most active strains of these bacteria were related to the genera *Desulfovibrio* (mainly *D. desulfuricans*) and *Desulfobulbus* (mainly *D. elongatus*) but representatives of the genera *Desulfococcus*, *Desulfobacter* and *Desulfosarcina* were also well present. As a result of their activity the pH of the waters was stabilized around the neutral point due to generation of hydrocarbonates ions during the microbial sulphate reduction (the role of limestone was also essential as it was mentioned earlier). The non-ferrous metals, iron and arsenic were precipitated mainly as the relevant insoluble sulphides. Uranium was precipitated mainly as uraninite (UO₂) as a result of the prior reduction of the hexavalent uranium to the tetravalent form.

Table 5. Sulphate-reducing bacteria in the effluents from the permeable reactive multibarrier

Sulphate-reducing bacteria	Cells/ml
<i>Desulfovibrio</i> (mainly <i>D. desulfuricans</i>)	10 ⁴ - 10 ⁷
<i>Desulfobulbus</i> (mainly <i>D. elongatus</i>)	10 ² - 10 ⁷
<i>Desulfococcus</i> (<i>D. postgatei</i>)	10 ² - 10 ⁶
<i>Desulfobacter</i> (<i>D. multivorans</i>)	10 ² - 10 ⁵
<i>Desulfotomaculum</i> (mainly <i>D. nigrificans</i>)	10 ¹ - 10 ⁴
<i>Desulfosarcina</i> (<i>D. variabilis</i>)	10 ² - 10 ⁵
<i>Desulfomonas</i> (non-identified species)	10 ¹ - 10 ⁴

A relatively long period of time (of about 3 – 4 months) was needed for the sulphate-reducing bacteria to establish a numerous and very active population in the multibarrier. The microbial sulphate reduction was the prevalent mechanisms in the deeply located layers and in the back zones in the multibarrier (at distances longer than 4 – 5 m from its inlet). The microbial sulphate reduction was a function of the concentration of organic monomers dissolved in the waters. These monomers were generated as a result of the biodegradation of the solid biopolymers by the different heterotrophs possessing hydrolytic enzymatic activity. This process resulted in a steady decrease in the concentration of the easily degradable solid biopolymers (cellulose and hemicellulose) present in the multibarrier (Table 6). At the same time, precipitation of pollutants caused a negative effect on the permeability of the multibarrier (Table 7). It must be noted that the precipitation of the different pollutants

was not homogenous and the different sites in the multibarrier differed considerably from each other with respect to their chemical and mineralogical composition (Table 8).

Table 6. Exhaustion of the solid organic substrates in the multibarrier after 28 months of treatment

Organic fraction	Initial mixture	Sampling points, distance from the inlet			
		1 m top	1m down	3m top	3 m down
g organic carbon/ 100g solid sample					
Crystalline polysaccharides	8.2	0.5	0.5	0.4	0.8
Non-crystalline polysaccharides	37.4	1.4	1.9	1.7	2.8
Lignin	5.1	4.6	4.8	4.3	4.1

The microbial activity in the multibarrier markedly depended on the temperature and during the cold winter days was negligible. However, at air temperatures about 0 °C and water temperatures close to the freezing point, the temperatures inside the multibarrier, within the deeply located layers, usually were in the range of about 2 – 5 °C. Under such conditions the microbial sulphate reduction still proceeded, although at much lower rates. In any case, the water clean up efficiency of the multibarrier was much higher during the warmer months of the year (Table 9).

Table 7. Data about the changes in the permeability of the main sections of the multibarrier during the treatment of the polluted waters

Section of the multibarrier	Time since the start of the operation, months				
	0 (start)	9	16	22	28
Filtration coefficient, m/h					
Alkalizing drain	110.4	34.6	10.8	4.7	0.08
Zones in the section for MDSR:					
2 m from the inlet	7.2	3.8	1.5	1.4	0.06
4 m from the inlet	7.2	5.7	3.1	2.6	0.07
6 m from the inlet	7.2	6.6	5.6	5.1	0.08

Note: MDSR = microbial dissimilatory sulphate reduction

The effluents from the multibarrier were enriched in dissolved organic compounds and in some cases still contained manganese in concentrations higher than the relevant permissible levels (Table 2). These effluents were treated in a natural wetland where the Mn^{2+} ions were oxidized to Mn^{4+} by some heterotrophic bacteria producing peroxide compounds and the enzyme catalase, which degraded the excess of peroxides to molecular oxygen and water. The Mn^{4+} ions precipitated as MnO_2 . The dissolved organic compounds were efficiently degraded by the different heterotrophs inhabiting the wetland.

The data from the recent monitoring of the multibarrier operation clearly reveal that after a period of twenty eight months since its start it is necessary to replace the armored limestone and the exhausted solid biodegradable organic substrates by fresh batches of these materials.

Table 8. Data about the composition of solid samples from the permeable multibarrier

Parameters	Zones in the section for microbial sulphate reduction					
	2 m from the inlet		4 m from the inlet		6 m from the inlet	
	Time since the start of the operation, months					
	16	28	16	28	16	28
pH (KCl)	5.68	7.06	6.35	7.23	7.43	7.45
Ash content, %	71.0	81.9	62.0	70.7	64.0	72.4
Organic content, %	29.0	18.1	38.0	29.3	36.0	27.6
Cu, mg/kg	1211	1950	482	1466	76.4	212
Zn, mg/kg	587	1488	363	921	135	183
Cd, mg/kg	4.9	15.2	2.9	5.2	0.9	1.0
Pb, mg/kg	46.6	41.9	74.9	68.0	36.9	70.7
Ni, mg/kg	269	1565	249	906	20.2	118
Co, mg/kg	298	890	98.4	495	8.8	59
Fe, mg/kg	6204	6632	6512	6804	6468	7120
Mn, mg/kg	473	895	810	2161	459	783
U, mg/kg	60.3	215	45.3	190	11.1	59.6
As, mg/kg	4.96	0.8	0.4	1.1	1.1	1.4
S total, mg/kg	6612	10087	6239	9111	5520	7050

Table 9. Removal of pollutants from the acid mine drainage by means of the permeable reactive multibarrier during different climatic seasons

Pollutants	Pollutant removed, g/24 h	
	During the warmer months	During the cold winter months (at 0 – 5 °C)
Uranium	2.42 – 19.2	0.35 – 2.27
Copper	9.74 – 80.2	1.52 – 7.20
Zinc	6.44 – 114.4	1.40 – 9.72
Cadmium	0.14 – 1.20	0.03 – 0.19
Lead	1.24 – 5.05	0.28 – 1.34
Nickel	2.71 – 11.35	0.60 – 2.84
Cobalt	1.80 – 7.81	0.41 – 2.08
Manganese	23.5 – 194	4.73 – 25.9
Arsenic	0.95 – 2.71	0.27 – 1.04
Iron	594 – 5481	88.4 – 712

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Groudev S., Spasova I., Nicolova M., Georgiev P., *Oczyszczanie kwaśnych odcieków ze złoża uranu za pomocą układu pasywnych barier*, Physicochemical Problems of Mineral Processing, 41 (2007) 265-274 (w jęz. ang.).

Kwaśne odcieki, „wytwarzane” przez złożo rudy uranowej zlokalizowane w Curilo (Bułgaria) były oczyszczane w instalacji o skali pilotowej, która składała się z wielu naturalnych barier. Całkowita objętość instalacji wynosiła 23 m³ i składała się ona z drenażowej sekcji alkalinizującej, z sekcji bakteryjnej redukcji siarczków w warunkach beztlenowych oraz z sekcji biosorpcji i dodatkowo, z sekcji chemicznej neutralizacji. Ścieki wykazywały pH w zakresie od 2.5 do 4.0 i zawierały: radionuklidy (U, Ra), metale ciężkie (Cu, Zn, Cd, Pb, Ni, Co, Fe, Mn) oraz arsen i siarczki. Stężenia jonów w ściekach było znacznie

większe niż przewidują to normy zezwalające na wykorzystanie ścieków w rolnictwie i przemyśle. Przepływ cieczy przez układ składający się z wielu barier był zmienny w granicach od 1 do 17 m³/24 godz. Odpowiada to czasowi przebywania roztworu w całej instalacji od 300 do 18 godzin. Wysoka efektywność usuwania zanieczyszczeń przez wielobarierową instalację była niezależna od pory roku. Nawet podczas chłodnej zimy temperatura powietrza i ścieku wahała się w okolicy 0°C. Jednak, pewne istotne zmiany w składzie cieczy i we właściwościach wielobarierowej instalacji miały miejsce podczas pracy instalacji w okresie 28 miesięcy.