Predictive geochemical modeling of long-term environmental impacts from waste rocks

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ABSTRACT

Contaminated mine drainage presents a potential hazard for aquatic ecosystems and groundwater resources. Reliable predictions of long-term developments in seepage quality are necessary to select reasonable decommissioning options and remediation measures for waste-rock heaps during mine closure and/or mining site rehabilitation to minimize future risks and environmental hazards. Extensive geochemical modeling work was used to assess the consequences of various remediation options for waste rock heaps at a former uranium mine in Eastern Germany. The predicted contaminant discharges are used for an assessment of long-term benefits of decommissioning alternatives to derive conclusions for sustainable remediation measures using risk-based cost/benefit analyses.

INTRODUCTION

Contaminated mine drainage is recognized as serious environmental issue at many mining sites world wide and is one of the major hydrogeochemical problems resulting form the anthropogenic intrusion into the geosphere. Especially acid mine drainage, resulting from chemical and biochemical oxidation of sulfide minerals like pyrite, marcasite, and/or pyrrhotite, can contain aquatic pollutants, such as sulfate, heavy metals, and naturally-occurring radionuclides in exceptionally high levels. Thus acid mine drainage presents a potential hazard for aquatic ecosystems and groundwater resources at mining sites where sulfide bearing ores, rocks or coals are mined (e.g. Allan 1995; Salomons 1995). In contrast, neutral mine drainage has not yet received as wide spread attention, although sulfate, metals, and especially uranium and radionuclides can also be of concern in neutral drainage (e.g. in heap seepage sufficiently buffered by carbonate minerals, or seepage from alkaline-leach tailings). The discharge of untreated mine waters can have serious detrimental effects on aquatic biota and groundwater quality on a large scale often far exceeding the area of the mining activities themselves. The prevention of contaminated acid or neutral mine drainage is therefore an important task not only during the lifetime of a mine but also a significant issue during decommissioning and/or remediation of abandoned mining sites to reduce environmental impacts and risks in the future. A sustainable close-out of a mining site has to take into account that the contaminant discharge from already acidic waste heaps can continue for several hundred years on a high level, and that currently neutral waste rock heaps may turn acidic at some point in the future, which leads to elevated contaminant discharges. Therefore reliable predictions of long-term developments in heap seepage quality using sophisticated geochemical and physical models are essential to choose appropriate remediation measures and to assess the long-term effects and remaining contaminant discharges after remediation (e.g. after cover placement) as well as potential long-term costs (e.g. for seepage collection and treatment, cover maintenance, etc.).

The former German Democratic Republic was one of the largest uranium producers of the world (cumulative uranium production approx. 220.000 t U) excelled only by the USA and Canada (Barthel, 1993). The close-out and remediation of the various mining and milling sites of the former Soviet-German mining company WISMUT - some of them situated in densely populated areas - is one of the largest remediation projects in the world at present. The mining site near Ronneburg in Thuringia was the most extensive uranium mining site in Germany. Between 1952 and 1990 approximately 125 million m^3 uranium ore with

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an average content of 0.085 wt.% U_3O_8 were produced from underground and surface mines at Ronneburg. The uranium production from this site (approx. 113,000 t) contributed about 50% of the total uranium production of the GDR between 1945 and 1990. During mining activities, approx. 125 million m^3 waste rocks from surface and underground mining were accumulated on various heaps outside the open pit. Further 80 million m^3 of waste rocks were placed in the worked out part of the open pit before 1990. In the following chapters the use and the significance of predictive geochemical modeling work in the ongoing decision making process to achieve a sustainable remediation for the waste rocks at the Ronneburg uranium mining site is outlined.

SITE DESCRIPTION

The uranium ores at the Ronneburg mining site are hosted mainly in Paleozoic (black)-shales, carbonates, and basic volcanic rocks (diabase) with ages ranging from Ordovician to Devonian. These rocks were intensely folded during the Variscan orogeny and faulted (Lange and Freyhoff 1991; Lange et al. 1991). The southern part of the mining area is located in the Variscan basement of the Ronneburg Horst. This area is drained by several small streams; some of them dried up due to the mining activity but will probably be water-bearing again after the flooding of the underground mine. The north-east boundary of the Ronneburg Horst is formed by an extensive NW-SE-striking fault zone (Crimmitschauer fault). North of this fault zone, the Variscan basement hosting the ore deposit is overlain by platform sediments of Permian (Zechstein) and Triassic ages (Bunter). These platform sediments contain several important groundwater aquifers; the uppermost is used for drinking water supply. The uranium ore at Ronneburg, which is thought to have formed by a combination of hydrothermal and supergene processes, consists mainly of pitchblende and coffinite and is accompanied by sulfide mineralization (mainly pyrite) and minor cobalt and nickel arsenides. The mine workings comprised several independent underground mines, which reached a maximum depth of approx. 950 m below surface and covered an area of approx. 50 km^2, and an open pit, which had an original volume of approx. 160 million m^3. Table 1 summarizes the characteristics of the Ronneburg mining site before the beginning of the close-out and the start of remediation measures.

Table 1 Characteristics of the Ronneburg uranium mining site before remediation

Feature	Dimension
Underground mining system	
depth	up to 950 m
area	approx. 50 km^2
number of levels	21
number of shafts	38
length of underground drifts	approx. 3.000 km
amount of excavated material	approx. 45 million m^3
Open pit	
area	1.6 km^2
original volume	approx. 160 million m^3
original depth	240 m
open volume	approx. 80 million m^3
Waste rock heaps	
number of heaps	14
total volume	125 million m^3
total area	4.7 km^2

During the mining activities, approx. 125 million m^3 of waste rocks from underground and surface mining were placed on various heaps outside the open pit covering an area of approx. 4.7 km^2. The size of the individual heaps ranges between some 100.000 m^3 up to 65 million m^3. In addition, some 80 million m^3 of waste rocks were placed in the mined out part of the open pit during mining operations. Table 2 presents an overview on the waste rock heaps at the Ronneburg mining site and their current seepage characteristics.

Several waste rock heaps at the Ronneburg mining site are currently generating acid seepage with high levels of uranium, heavy metals, and sulfate, due to the rapid oxidation of sulfide minerals, enriched especially in black shales containing high amounts of organic matter. Neutral seepage containing elevated uranium and nickel concentrations and very high sulfate levels occurs in heaps, where the sulfuric acid generated by the sulfide oxidation process is at present sufficiently buffered by magnesium-bearing carbonates (e.g. dolomite). In the effluents from these heaps, the formation of aqueous magnesium-sulfate complexes enhances the sulfate solubility far above the equilibrium concentration expected from gypsum saturation (approx. 2 g/L). The high sulfate discharges and the elevated temperatures in these heaps indicate high sulfide oxidation rates even at an overall neutral pH. In Table 3 the seepage qualities of an acid and a neutral heap from the Ronneburg mining site are given as an example.

Table 2 Dimensions and seepage characteristics of waste rock heaps at the Ronneburg uranium mining site before remediation

Heap	Volume [million m^3]	Area [10,000 m^2]	Seepage
Absetzerhalde	65.8	224.7	acid
Innenkippe [1]	64.0	-	acid / neutral
Nordhalde	27.2	83.9	acid
heap Paitzdorf	7.6	24.9	neutral [3]
Gessenhalde [2]	6.9	28.7	acid
heap Reust	6.3	20.5	neutral
heap Beerwalde	4.5	23.9	neutral [3]
heap Drosen	3.5	22.8	neutral
heap 4	0.9	7.8	- [4]
heap 370	0.8	6.0	neutral
heap Korbussen	0.4	4.1	neutral
heap 377	0.3	2.9	neutral / slightly acid
heap 381	0.1	1.0	- [4]
Diabashalde	0.1	1.0	neutral

[1] waste rocks placed in the open pit during production

[2] acid leach heap

[3] currently neutral seepage, but very high sulfate discharge indicating high sulfide oxidation rates

[4] no seepage observed

Table 3 Seepage quality for selected Ronneburg heaps

Parameter	unit	Nordhalde	heap Beerwalde
pH	[-]	2.0 – 3.5	7.5 – 8.0
Ca	[mg/L]	200 – 650	270 – 430
Mg	[mg/L]	800 – 2,800	4,200 – 8,000
SO_4	[mg/L]	5,000 – 20,000	19,000 – 29,000
HCO_3	[mg/L]	<1	200 – 600
Fe	[mg/L]	300 – 3,200	0.1 – 2.0
Al	[mg/L]	200 – 400	0.1 – 1.5
Mn	[mg/L]	20 – 180	2.5 – 22.0
U	[mg/L]	0.4 – 6.5	3.5 – 5.5
Ra-226	[mBq/L]	10 – 200	10 – 300
Ni	[mg/L]	24.0 – 38.5	0.1 – 1.2
Co	[mg/L]	6.5 – 13.5	0.02 – 0.25
Cu	[mg/L]	2.5 – 5.0	0.02 – 0.05
Cd	[mg/L]	0.2 – 0.5	<0.05
Zn	[mg/L]	16.5 – 26.0	0.1 – 0.25

REMEDIATION STRATEGY

Key issues of the decommissioning of the Ronneburg mining site comprise (i) the flooding of the extensive underground mining system, (ii) the remediation of the - previously partly backfilled - open pit, and (iii) the remediation of waste rock heaps from underground and surface mining. Further measures deal with the demolition of radioactive contaminated buildings as well as the construction of a water treatment plant, etc.

At present, environmental impacts from the waste rock heaps at the Ronneburg mining site result from:

- the discharge of contaminated seepage into ground- and surface waters,
- radon exhalation from waste rocks, and
- dispersion of radioactive contaminated dusts from the heaps.

Possible remediation measures for the waste rocks include the *in situ* remediation of waste-rock heaps using various types of engineered covers or the relocation of waste rocks into the open pit and cover placement on the backfilled pit. The backfilling of parts of the open pit and the use of a cover is necessary to enhance the slope stability of the pit and to reduce radiological hazards. Even after remediation, the heaps and the backfilled open pit, which is hydraulically connected to the underground mine, will serve as long-term sources for contaminants having potentially detrimental effects on ground and surface water qualities.

Appropriate and sustainable remediation measures should reduce the contaminant discharge and radon exhalation from the waste rocks to acceptable levels (e.g. below existing guide lines), and should have the best cost/benefit ratio (i.e. the "lowest overall costs"; cf. Goldammer and Mager 1995; Goldammer et al. 1999). These "overall costs" comprise direct short-term costs for remediation measures (e.g. placement of engineered covers; backfilling waste rocks in underground workings or open pits), long-term costs (e.g. costs for seepage collection and - if necessary - water treatment; monitoring; maintenance; etc.) as well as the monetary equivalents of remaining risks for human life and health and impacts on ecosystems. Due to the requirements of the regulating authorities dealing with radiation protection during remediation of uranium mining and milling sites in the former GDR, the optimization of costs, benefits and remaining risks of remediation measures has to take into account a time frame of at least 200 years.

PREDICTIVE GEOCHEMICAL MODELLING OF REMEDIATION OPTIONS

The use of risk-based cost/benefit analyses in the ongoing decision-making process for a sustainable remediation of the Ronneburg mining site requires the assessment of long-term environmental impacts of remediation measures and associated long-term costs. In this context, reliable predictions of the future heap seepage quality and the contaminant discharge from the back-filled open pit are indispensable to assess water treatment costs, radiological and toxic risks resulting from use of ground and surface waters, as well as environmental detriments, and to apply reasonable remediation measures. The geochemical modeling work performed during the assessment of the benefits and environmental impacts associated with various remediation options for waste rocks from surface and underground mining at the Ronneburg site comprised:

- the modeling of long-term contaminant release from waste rocks heaps left in their current condition;
- the modeling of the consequences of an *in situ* remediation of waste rock heaps by placing different types of engineered covers for the limitation of oxygen supply and reduction of infiltration; and
- simulations of selected scenarios for backfilling the open pit.

The simulations of the backfilling of the open pit focused on the optimization of the pit filling process to minimize the long-term release of contaminants from the backfilled pit, which will discharge together with parts of the mine water from the underground mine into a small stream in the future. These simulations addressed (i) the use of various criteria to determine the best placement order for different types of waste rocks (e.g. acidic, potential net-acid-generating, non-acid generating) depending on their contamination potential, (ii) the consequences of varying the amount and types of waste rocks filled in the pit (i.e. if some heaps will be left *in situ*), as well as (iii) technical measures during backfilling, such as addition of lime to already acidic waste rocks, compaction of backfilled material using heavy trucks, as well as requirements for the quality of the final pit cover.

The geochemical models employed for the prediction of the long-term contaminant discharge from the waste rocks and the assessment of associated environmental impacts especially take into account the following processes (amongst others):

- acid generation by chemical and microbiological oxidation of sulfide minerals (e.g. pyrite, marcasite, and pyrrhotite);
- formation and dissolution of acid and contaminant storing minerals (e.g. jarosites, melanterite, schwertmannite, ferric hydroxide etc.);
- buffering of acid by carbonates, silicates, and hydroxides;
- chemical weathering processes (e.g. silicate weathering);
- formation of aqueous complexes (e.g. uranium-carbonate-complexes, magnesium-sulfate-complexes) that can enhance the solubility and mobility of uranium and other contaminants;
- the mobilization of heavy metals, uranium and other radionuclides, and their retardation (e.g. by ion-exchange, sorption and/or co-precipitation).

The geochemical models allow predictions of heap seepage and ground water qualities up to several hundred years. In addition, the convective and diffusive transport of oxygen in the waste rocks, which is essential regarding the sulfide oxidation rates, heat and vapor transport due to exothermal sulfide oxidation, and changes in particle size due to physical and chemical weathering processes are addressed. In contrast to hydrogeochemical models making extensive use of equilibrium thermodynamics, the applied models make use of kinetic descriptions to evaluated the important effects of reaction kinetics (e.g. regarding the rates of important processes, like sulfide oxidation, buffering by carbonates and silicates, dissolution of stored acidity, etc.) on acid generation and consumption in waste rocks or tailings. Sulfide oxidation rates are addressed using a shrinking reactive front for large particles or sulfide minerals embedded in the rock matrix and a shrinking radius concept for fines. The kinetics of non-oxidative dissolution of silicate and carbonate minerals is described by surface area and/or transport controlled reaction mechanisms (cf. Stumm, 1992). Buffering minerals, which were taken into account in the reactive modeling comprise carbonates (calcite, dolomite, siderite) and silicates (sericite/illite, chlorite) at neutral resp. slightly acidic pH, as well as hydroxides of aluminum, manganese and iron and minerals of the jarosite group at lower pH.

Based on the construction history (e.g. source of waste rocks and their lithology), the construction methods (e.g. by truck, end dumping, conveyer belt, etc.), and the results of drill core analyses and trench samples, the modeled waste rock heaps were divided into up to 10 modeling compartments with different geochemical and hydrogeological characteristics. For the assessment of the relocation of waste rocks into the open pit, the unsaturated and the in future saturated part of the backfilled pit, the surrounding rocks, and parts of the underground mine were simulated using up to 20 modeling compartments. Each compartment was further subdivided into horizontal layers, in which the geochemical processes are allowed to run independently to simulate the development of reaction fronts (e.g. due to oxygen diffusion and convection, or percolation of oxygen rich water). Background water flows and qualities in the underground mine and the quality of mine water inflowing in the saturated part of the open pit where taken from previously performed modeling runs of the flooding process and the long-term contaminant discharge from the flooded underground mining system (Kistinger et al., 1998). The geochemical models were calibrated using time series of data for heap seepage quality and the results of leaching experiments and column test work performed on waste rocks from the Ronneburg site by WISMUT. In addition to sensitivity analyses for some important input parameters (e.g. infiltration rates, oxygen diffusion rates, availability of sulfide and carbonate minerals), probabilistic calculations employing Monte-Carlo-simulations using probability distributions for various input parameters were performed, to address and keep track of uncertainties in the input parameters for the geochemical modeling runs.

Table 4 summarizes the predicted long-term contaminant discharges from the 27.2 million m^3 sized, acid generating Nordhalde for two different types of engineered covers as an example.

Table 4 Predicted contaminant discharges from Nordhalde waste rock heap for different engineered covers (mean values of probabilistic simulations)

		simple cover		complex cover	
		100 years	**200 years**	**100 years**	**200 years**
SO_4	[t]	157,000	265,000	47,500	74,100
Fe	[t]	21,900	39,400	4,950	7,100
Al	[t]	4.030	4,540	2,500	3,120
Mn	[t]	809	1.380	366	597
U	[t]	35.2	71.5	7.3	12.2
Ra-226	[MBq]	734	1.720	153	291
Ni	[t]	171.2	268.7	45.7	68.8
Co	[t]	44.3	60.5	18.4	26.9
Cu	[t]	61.5	103.9	13.1	19.3
Cd	[t]	2.1	3.2	0.5	0.8
Zn	[t]	105.8	168.2	25.3	37.6

The simple cover scenario refers to the completion of the partly existing thin soil cover on this heap (assumed mean thickness approx. 0.5 m). The complex cover refers to an engineered cover containing a water-saturated compacted clay layer (0.5 m), which is overlain by a water retaining soil layer (1.5 m) that is capable of sustaining the assumed future vegetation that would be typical for the humid climate at the site (i.e. mainly trees and bushes), without root penetration into the clay barrier. The long-term hydraulic properties of the covers as well as oxygen ingress by diffusive and/or convective transport - the latter relevant mainly for the simple cover - were estimated using the models HELP 3 (Schroeder et al., 1994) and SoilCover 4.01 (SoilCover, 1997), various in-house models, as well as experiences from other sites. It was assumed that the simple soil cover would allow percolation rates through the heap between approximately 20 and 30% of the mean annual precipitation (681 mm/a); oxygen diffusion rates were estimated in the order of 1×10^{-6} m²/s. Percolation rates for the complex cover were estimated at 10±5% of the mean annual precipitation in the long term. Furthermore, a reduction of diffusive air flow of about 2 orders of magnitude compared to the simple cover was assumed in the modeling calculations. Time dependent changes in the physical parameters of the waste rocks themselves, such as changes in moisture content and oxygen flow due to the decrease in grain size resulting from weathering were not taken into account and may therefore add to the uncertainties evaluated in the stochastic simulations. In figure 1 results of a stochastic simulation of the effluent quality of the Nordhalde waste rock heap for the two cover scenarios are shown for selected parameters.

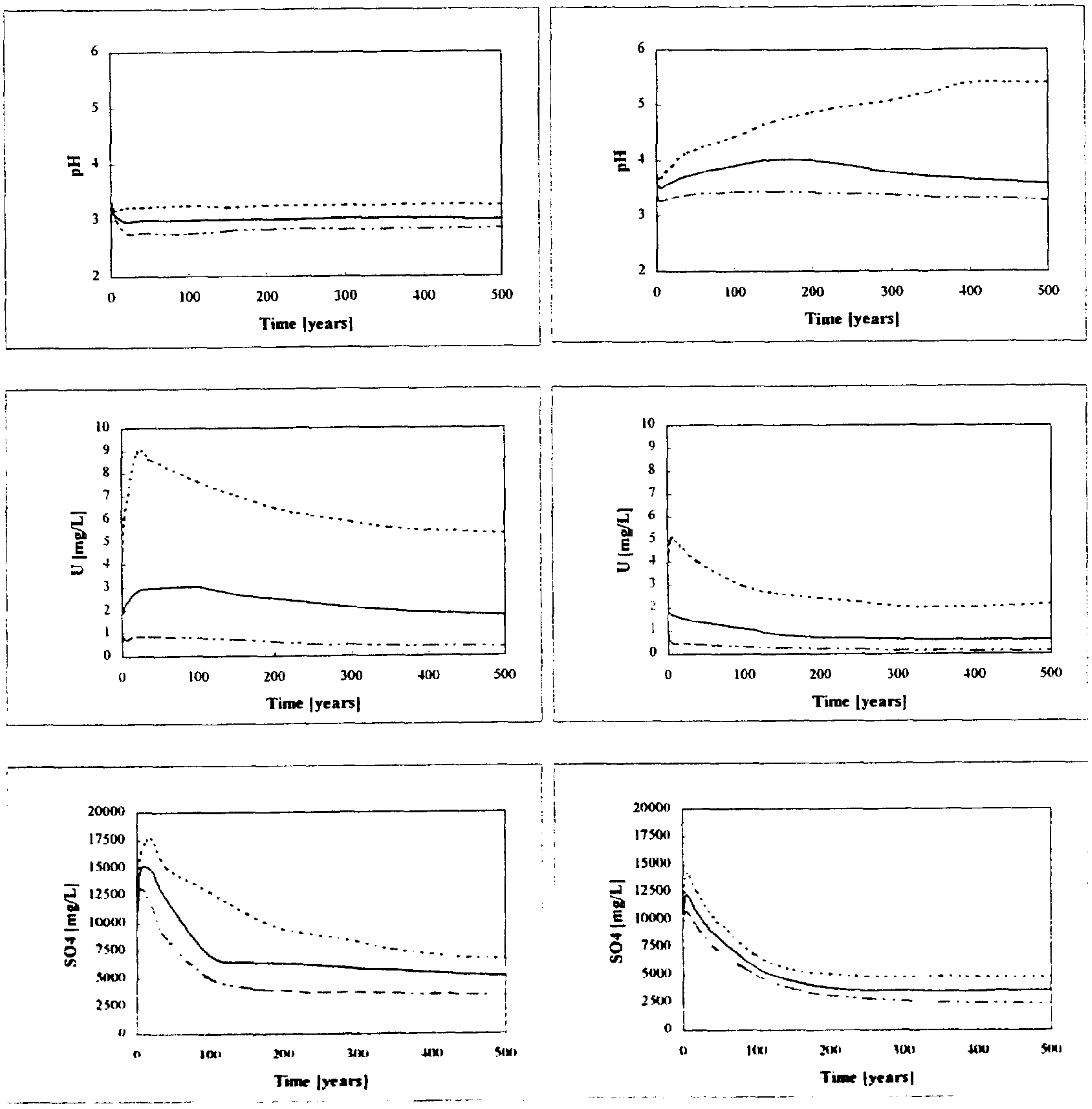

Figure 1 Predicted seepage quality for Nordhalde waste-rock heap for different covers. Shown are the 5th (stroke-dashed line), 50th (solid line), and 95th percentiles (dashed line) of a stochastic simulation for a simple (left column) and a complex cover (right column).

The placement of a simple cover does not lead to a significant reduction of contaminant loads from this heap compared to the situation when left in the current condition, due to only slight reductions in infiltration rates and oxygen ingress. In contrast, the long-term contaminant discharge can be significantly lowered by an engineered cover containing a water saturated clay liner to reduce infiltration and oxygen supply, although the seepage remains acidic throughout the simulation time. The long-term effluent quality from this heap is strongly effected by the amount of contaminants stored in acid pore waters and secondary minerals like jarosites, as well as the inhibition of still present buffering minerals (carbonates) by gypsum and ferric hydroxide coatings. Therefore even after placement of a complex cover that strongly reduces the oxygen ingress and thus the sulfide oxidation process, acidic conditions prevail in this heap in the long term. Simulation runs for the backfilled open pit suggest that the relocation of the complete Nordhalde into the open pit does not increase the long-term contaminant discharge from the backfilled pit via the underground mine into surface waters, if the acidic and potential acid-generating waste rocks from this heap are mixed with lime and placed below the future groundwater table in the pit.

The results of the detailed simulations of the discharge of various contaminants (e.g. radionuclides, heavy metals, sulfate, acidity, etc.) from the mine wastes served as an integral part of a comprehensive assessment of the effects of heap remediation on the long-term quality of ground and surface waters in the Ronneburg area. To compare the potential long-term significance and benefits of various decommissioning alternatives regarding long-term water treatment costs, remaining risks for human life and health resulting from radiological and conventional contaminants, remaining environmental detriments, etc., an integrated approach for the assessment of radiological and non-radiological risks employing stochastic methods (Monte-Carlo simulations) was used (Goldammer et al., 1999). In case of the Nordhalde, direct costs for cover placement and long-term costs (net-present values) for water treatment and cover maintenance as well as monetary equivalents of the remaining risks and environmental detriments for an *in situ* remediation with an engineered cover exceed the rather high short term costs for the relocation of the large volume of waste rocks into the open pit. Therefore the relocation of the waste rocks from this heap was recommended.

CONSEQUENCES FOR MINING SITE REMEDIATION AND WASTE ROCK MANAGEMENT

The predictive geochemical modeling of the contaminant discharge from the waste rocks, the assessment of the long-term benefits and costs of remediation measures, as well as detailed analyses and modeling of the hydrogeological regime after flooding of the underground mine in Ronneburg (c.f. Kistinger, 1997), lead to the following conclusions regarding a sustainable remediation of the waste-rock heaps at the Ronneburg uranium mining site:

- the *in situ* remediation of waste rocks employing engineered covers containing a water saturated clay liner to minimize oxygen penetration (and reduce thus the sulfide oxidation rates) and radon exhalation will prevent currently neutral waste rock heaps characterized by high sulfide oxidation rates and high sulfate and uranium discharges from turning acidic in the future;
- placement of such covers on already acid waste rock heaps will slow down the acid generating process, but due to the amount of acidity stored in porewater and secondary minerals, the seepage quality does not improve significantly even in the long-term, although the discharged contaminant loads are significantly reduced due to lower infiltration rates;
- the relocation of waste rock material from the heaps with the highest contamination potential, i.e. the acid leached Gessenhalde (6.5 million m^3), the Absetzerhalde (65 million m^3), and the Nordhalde (27 million m^3) into the open pit is the remediation measure associated with the lowest long-term detrimental impacts on ground and surface water and the lowest overall costs. Those already completed (Gessenhalde) and ongoing measures (Absetzerhalde and Nordhalde) will result in the remediation of approx. 100 million m^3 (i.e. 80%) of the waste rocks at the Ronneburg mining site;
- the relocation of further, currently non-acid-generating waste rocks into the open pit will not increase the cumulative contaminant discharge from the backfilled pit into the environment within 500 years.

The strategy developed for the relocation of waste rocks into the open pit comprises the following features to minimize the contaminant discharge from the backfilled pit into the ground and surface water:

- the waste rocks are placed in the open pit according to the amount of stored acidity and net-acid-generation potential determined by field tests; acid waste rocks will be placed in the part of the open pit that will be water-saturated after flooding the underground mine.

- lime is added to acidic waste rocks placed below the future groundwater table to reduce the contaminant release by buffering of stored acidity; the amount of lime to be added depends on the amount of stored acidity and is determined by field tests.
- the waste rocks are placed in thin layers and compacted by heavy trucks to reduce the hydraulic conductivity and the oxygen transport in the unsaturated material as well as to avoid cover failures in the future due to settling.

The recommended remediation measures for the waste rocks at the Ronneburg mining site, which were derived from an integrated assessment of risks, cost, and benefits of different remediation options, are in compliance with the relevant guidelines regarding radiation protection and surface water quality.

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