INTERNATIONAL PERSPECTIVE ON THE ROLE OF ACID GENERATION IN SELECTING DECOMMISSIONING TECHNIQUES FOR URANIUM MINING SITES IN EASTERN GERMANY¹

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<u>Abstract</u>: The control of acidic drainage from uranium mine wastes in Germany is a significant component of mine site rehabilitation that began in 1992. Uranium was mined on a very large scale in the States of Thüringen and Sachsen from 1946 to 1991. Sulfide oxidation creates acidic drainage from waste rock piles at several mine sites and may occur in the future at two tailings impoundments. The extent of the problem of acidic drainage and methods to control acid production are reviewed from an international perspective. A central component of the rehabilitation plans in Germany is a proposal to fill a large open pit with acid-producing waste rock, allowing it to flood, and treating the overflow. This plan is assessed using a regional geological model in a probabilistic framework. This modeling has proven to be a useful tool in assessing management alternatives.

Background

Starting in 1946 and ending in 1991, uranium was mined on a large scale in the States of Thüringen and Sachsen in southeastern Germany. The scale of uranium mining in this area made it one of the world's largest uranium producers. The mining and uranium recovery was undertaken in the former East Germany by U.S.S.R.-German Democratic Republic company, Wismut. In addition to the recent mining for uranium, mining of these areas had been carried out since the Middle Ages for iron, silver, and other metals including bismuth (Wismut 1992). Following the reunification of Germany and the closure of the mines, major work and environmental assessment have been performed to decommission both the Wismut and historic sites. The authors have participated in assisting the Bundes Ministerium für Umwelt Naturschutz und Reaktorsicherheit (BMU) in their ongoing review of the Wismut decommissioning programs.

Over 1,600 sites in the mining region have been identified as having greater than background levels of radioactivity. Most of these sites have been linked to previous mining activity. The major mine waste sites of interest are those that resulted from the uranium mining, where approximately 200 million mt of mine tailings and 500 million mt of mine waste rock have been deposited on surface. The major uranium mining and mineral processing sites were those of the Wismut company; their locations are shown in figure 1.

An inventory of the major wastes remaining on surface is summarized in table 1. Acid generation resulting from the oxidation of sulfide minerals has been identified as a major concern in the mines and waste rock heaps on surface and underground at Ronneburg and is a potential concern at Königstein. In addition, there is a potential problem in two of the tailings basins at the Seelingstädt site, where ores were processed by acid and alkaline leaching to recover uranium.

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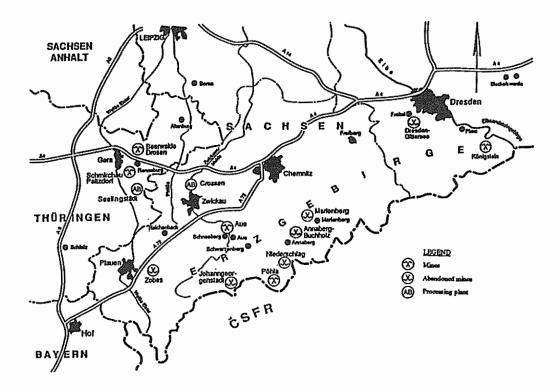


Figure 1. Location of Wismut mining works

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TABLE 1 SUMMARY CHARACTERISTICS OF URANIUM MINE WASTES

Location	Waste type	Volume million m ³	Surface area, ha	Radium Bq/g	Pyrite, %
Ronneburg	Waste rock	170	409	0.3-1.5	3.3-4.8
Seelingstädt	Tailings	104	335	6.7-12	Variable*
Seelingstädt	Waste rock	59	519	0.25-1.65	NA
Crossen	Tailings	45	213	6.0	NA
Crossen	Waste rock	2	22	2.0-2.3	NA
Aue-Schlema	Waste rock	47	343	0.2	NA
Königstein	Waste rock, treatment sludges	3.3	25	0.34-5.6	1-2

NA Not available

Bq/g Becquerel/gram

* Acid leach tailings 0.8% to 5%, alkaline <1%,

Because of the very large quantities of these sulfide-bearing wastes, minimizing acid generation is a critical element in the decommissioning of the sites. The concerns are not only the production of acid waters and their effect on the environment per se, but also the leaching of radioactive species and heavy metals into ground and surface water and their subsequent dispersal in the environment. In addition, if significant acid generation continues to occur, there will be a need to collect and treat water for possibly hundreds of years. Ongoing water treatment also requires ongoing management of water treatment sludges, which contain radioactive and other heavy metals.

This paper describes the acid generation sources and magnitude of the problem and implications for decommissioning these former uranium mining areas.

The Impact Of Acid Generation in Uranium Mine Wastes

Experience in Canada and elsewhere has led to the conclusion that sulfuric acid generation is one of the major concerns associated with the decommissioning of sulfide-containing tailings and waste rock areas. Acid-leached uranium tailings are often significant acid producers even though the sulfide mineral content in the ore may originally have been very low. Strong acid generation can occur because most or all of any natural mineral alkalinity has been removed in the uranium extraction process by a hot sulfuric acid leach.

The chemistry of acid generation has been reviewed in detail (Nordstrum 1982) and can be summarized by the following reactions:

$$2 \text{ FeS}_{2} + 7 \text{ O}_{2} + 2 \text{ H}_{2} \text{ O} \rightarrow 2 \text{ Fe}^{2+} + 4 \text{ SO}_{4}^{2-} + 4 \text{ H}^{+}, \tag{1}$$

$$2 \text{ Fe}^{2+} \rightarrow 2 \text{ Fe}^{3+} + 2e^{-},$$
 (2)

$$2 H^{+} + \frac{1}{2} O_{2} + 2e^{-} \rightarrow H_{2}O, \qquad (3)$$

Although these reactions can occur in a biologically sterile environment, chemolithotrophic bacteria, particularly <u>Thiobacillus ferrooxidans</u>, can accelerate the iron oxidation rate. Ferric iron in acidic solution can also oxidize metal sulfides (MS) according to

$$MS + 2 Fe^{3+} \rightarrow M^{2+} + S^{\circ} + 2 Fe^{2+}$$
(4)

Low pH (<4.0) seepage waters are often associated with mine wastes where sulfide oxidation is occurring, but frequently alkaline minerals such as calcite or dolomite present in the wastes can neutralize the acidity produced and near neutral conditions (pH ~ 7) can result *until the alkaline minerals are consumed or rendered unreactive by surface coatings*. Acidic drainage from mine wastes will solubilize many metals including aluminum, copper, zinc, nickel, manganese, and lead. For uranium-rich deposits, uranium and thorium are also dissolved. Blair et al. (1980) have identified metal contaminants mobilized by acid production as the most important concern in the management of pyritic uranium tailings. Also, acidic drainage has been identified at uranium mine waste rock sites in Australia by Ryan and Joyce (1991). Waste rock piles are very porous to water and oxygen, essential ingredients for sulfhide oxidation; because the piles are usual constructed on porous foundations, this "acid rock drainage" has been very difficult to prevent and control at sites around the world.

Acidic drainage is a significant environmental concern at the former uranium mining area in the States of Sachsen and Thüringen, Germany. High levels of acidity and dissolved metals are found in drainages at several locations. At one location, Ronneburg, autogenous combustion caused by rapid oxidation of sulfide sulfur and carbon (graphitic carbon) had caused additional problems in mining and waste disposal. Significant quantities of sulfide minerals (notably pyrite) are also present in the waste rock at Crossen, Aue, and Königstein. Although most drainages sampled to date indicate neutral drainage, elevated levels of dissolved calcium and magnesium sulfates indicate that the possibility of future acid generation in these wastes cannot be excluded.

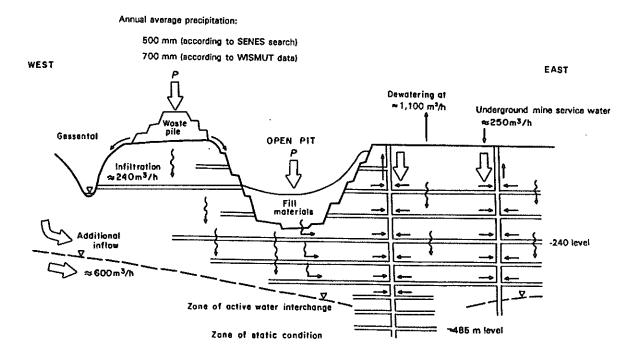
Site Considerations

Ronneburg-Drosen

The Ronneburg-Drosen mines are in east Thüringen, Wismut's most important uranium mining area. In total, these mines produced over 170 million m³ of waste rock, all of which is potentially acid generating. At present, over 100 million m³ of waste rock are stored on surface (in 14 waste piles). One pile was subjected to in situ uranium leaching using mine water supplemented with sulfuric acid. The remainder had been placed in a large open pit. Like most of the waste piles in southeastern Germany, the waste piles are located close to villages and individual homes (within several hundred meters). In addition to the understandable concerns about environmental radioactivity, particularly radon emissions, there are concerns about contamination of surface water and ground water and physical access to the materials. A cross section of the most environmentally significant part of the mining area at Ronneburg is shown in figure 2. Mining ceased at Ronneburg in December 1991, and the lower levels of the mine are now being allowed to flood.

Current plans (Wismut 1992) are to fill the open pit with as much waste rock as possible. The most severe acid producers will be placed on the pit bottom, but about 30 million m³ will remain above the water table. The final contouring and cover applied to these materials will be important factors in determining long-term acid production. Excess alkalinity, for example, lime and/or limestone, is being considered to help control acid generation, and studies were recently completed to determine the effect of submerging most of the waste rock under the water table in the open pit.

Until the mine workings are flooded, acid will continue to be produced from sulfide minerals in open spaces and fractured zones underground. In addition, in the long term after the mines are flooded, acid will continue to be produced in the fractured ground above the water table.





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Seelingstädt

Seelingstädt was a central ore processing facility for the Wismut mines. Uranium was extracted from the ore in parallel circuits, one with alkaline sodium carbonate, and the other with sulfuric acid. The sulfuric acid circuit was used to leach uranium from high sulfide ores and pyrite concentrates. Tailings from the acid circuit are potential acid producers if dewatered and exposed to oxygen. The tailings from the alkaline circuit represent no acid generation potential because the pyrite was converted to soluble sulfate by the following reaction:

 $4FeS_2 + 15O_2 + 16Na_2CO_3 + 14H_2O \rightarrow 4Fe(OH)_3 + 8Na_2SO_4 + 16NaHCO_3.$ (5)

Available data (Wismut 1992) indicate potential for acid generation in two large tailings basins at this site. At present, the basins are water saturated, and much of the formerly exposed tailings surface is covered with soil. The conceptual plan (Wismut 1992) is to dewater the tailings pond and to cover the balance of the dewatered tailings with a soil cover. Dewatering has the potential to result in increased oxidation and acidification of sulfide minerals, even though the pore fluids are currently alkaline from the mixing in of alkaline chemicals used in the milling process.

With respect to tailings and acid generation, the situation at Seelingstädt is unique; acid leached tailings are covered with alkaline water (pH 7.4 to 9.9) with high levels of dissolved sulphates (10 G/L). However, any residual mineral alkalinity has been removed in the acid leaching process, and if the tailings are dewatered as planned, infiltration of rainwater could flush out these buffering salts and permit acid generation to start. Based on the appearance of ferric iron salt coloration on exposed tailings beaches, this acidic generation potential appeared to be confirmed.

There is considerable international experience in predicting the rate of acid generation in pyritic mine tailings, and estimates of the rate of acid production by sulfide-bearing uranium tailings have been given by Halbert et al. (1983) and Scharer et al. (1991). Acid-base accounting on field samples (Wismut 1992) has shown that the acid-leached tailings are potentially acid generating; precise predictions cannot be made until the decommissioning scenario (eg., dewatering and covering) is established.

<u>Königstein</u>

The Königstein site in Sachsen has extensive underground workings, which were initially developed by conventional mining, and more recently by underground in situ leaching with sulfuric acid addition to leach solutions recirculated from surface. The key environmental concern at this site is the potential for contamination of an aquifer which overlies the mining zone and which is used for drinking water. About 1 million m³ of acid leach solution has been lost from circulation and is presently in the pore space in the fractured rock. In addition, pyrite is present in most of the uranium-bearing zones, and these strata represent significant (but currently unknown) acid generation potential.

In the near future, the mine will be flooded, and acid production will cease, but until such time acid will continue to be produced in the open mine volumes. Also there will remain considerable residual acidity from the underground leaching operation, and plans are being developed to neutralize this acidity on surface by recirculation of flood waters. Since the underground workings cover 5 km², the effect of the natural acidification is potentially significant.

Also at Königstein, there exists a 3.2 million m³ surface waste pile, which is composed of 2.0 million m³ of heap leach waste rock and other wastes, principally uranium-barren waste rock and water treatment sludges. The main concern with respect to acid generation is the heap leach pile. Although it has been depleted in sulfide sulfur content by bacteria-catalyzed oxidation, oxidation of the estimated remaining 1% to 2% pyrite content could accelerate in the near future. Currently, the waste pile produces an acidic effluent, as shown in table 2

3 to 10 m³/h		
2 to 4		
30 mg/L		
0.5 Bq/L		
200 mg/L		
3000 mg/L		

TABLE 2 ACIDIC DRAINAGE FROM KÖNIGSTEIN WASTE ROCK

Technical Considerations for the Prevention and Control of Acid Production

Various interdependent geochemical, physical, and biological factors control the rate and quantity of sulfide oxidation in the waste rock and tailings. These factors include temperature, bacteria activity, oxygen profile, pore water composition, effective grain size of sulfide minerals, and acid neutralization potential.

As an example of currently available predictive techniques, the authors have recently completed an extensive review of the environmental impact of depositing acid-producing waste rock into the bottom of the open pit at Ronneburg. To develop a reference point for environmental assessment, a waste rock pile that had formerly been a heap leach for uranium recovery was selected for evaluation. A computer model (ACIDROCK) was used to simulate the placement and leaching history of the waste rock pile (SENES 1993a). The model was used to predict the long-term seepage quality for various management options (e.g., covered with an engineered soil cover, and uncovered). The results of modeling the 27-ha pile showed that -

- Acidity will be produced for more than 100 years, with or without cover;
- A state-of-the art earth cover will reduce acid production and water treatment requirements by an order of magnitude;
- Continuous care and maintenance of the site would be required.

A much more complex predictive modeling evaluation was completed on the option of placing the same waste rock pile into the bottom of the pit. Geochemical measures, such as the addition of lime to neutralize acidity contained in the waste rock, were considered as part of the evaluation. The first stage of the modeling was the development and application of a hydrological model that simulated the flooding process, the establishment of steady-state flows with all of the mine cavities filled with water, and the contaminated ground water emerging at a nearby stream valley (Brenk Systemplanung 1993). A filled pit in the flooded condition is shown in figure 3.

Annual average precipitation:

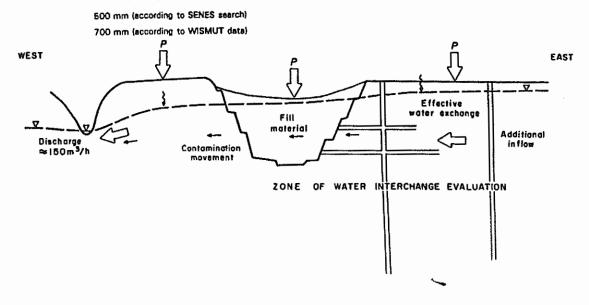


Figure 3. Wismut Decommissioning Ronneburg Site Future Hydrological Situation Typical Conceptual E-W Section

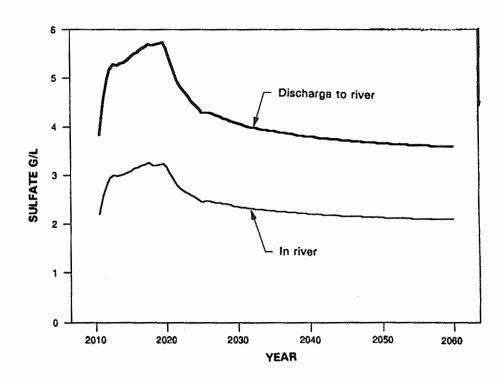


Figure 4. Predicted Sulfate

A computer model (ROCKSTAR) was used to simulate the geochemical processes occurring in the flooded pit materials and the associated underground mining areas (SENES 1993b). The model was used deterministically to predict the water quality within the pit fill and mining areas, as well as the water quality of the surface water body (river) that would receive the discharge from the mining region. Figure 4 shows the predicted quality of the discharge to the river, and the quality in the river.

Probabilistic modeling was also employed. A computer model (RANSIM) was used to randomly select many input values from the specified probability distributions for the key model parameters (SENES 1989). These values are substituted into the geochemical model (ROCKSTAR) to obtain a single value for each output variable. This procedure is repetitive, with each selection being referred to as a trial. Using the probabilistic approach, subjective probability distributions rather than single numbers will be obtained for the calculated variables. An example of the predicted water quality of the discharge to the river for 50 trials of the probabilistic model is shown in figure 5.

The predictive modeling assessment of placing the heap leach waste rock into the pit at Ronneburg can be summarized as follows:

- Nearby mine openings and underground broken rock zones control water flow patterns. (The underground mine excavations included over 1,800 km of tunnels).
- The contained acidity and stored oxidation products of the waste rock had a small impact on the emerging water quality.
- The addition of lime to the waste rock reduced the predicted impact of the waste rock to insignificant levels (reduced sulphate and metal mobility).

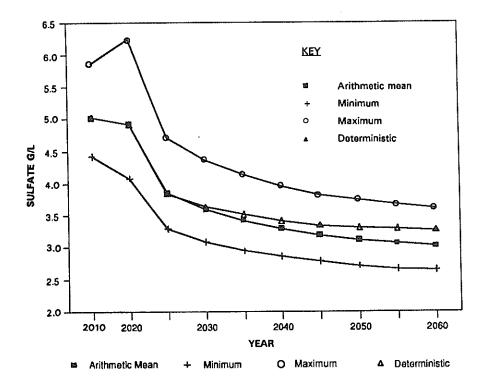


Figure 5. Predicted Water Quality for Discharge to River Probabilistic Approach - 50 Trials

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<u>Summary</u>

Control and treatment of acid rock drainage is a major focus of the decommissioning strategies being developed for the uranium mine sites in south eastern Germany.

Sophisticated mathematical models are being applied in the evaluation of mine decommissioning technology, which includes prevention and control of acidic drainage from waste rock and tailings. A major focus of the control measures for waste rock is depositing the rock into an open pit and then allowing the pit to flood with mine waters. Worldwide, the evaluation of options for dealing with acid-producing waste rock and tailings has led to the selection of the use of water covers and underwater disposal as the best technology.

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