MODELING OF ACID MINE DRAINAGE PHYSICAL PROCESSES IN THE NORDHALDE OF THE RONNENBURG MINING DISTRICT, GERMANY

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ABSTRACT

The Nordhalde is one of the largest waste rock piles in the Ronnenberg mining district, consisting of approximately 27,000,000 m^3 of material. The pile contains significant amounts of pyrite which is currently oxidizing, resulting in the generation of Acid Mine Drainage (AMD). The oxidation of pyrite is directly related to the consumption of oxygen and the production of heat. To better understand the rate and physical processes which govern pyrite oxidation in the Nordhalde, the pile was subject to extensive geochemical characterization and borehole instrumentation. A numerical model which integrates the physical parameters available and represents the coupled physical processes was developed for the Nordhalde using the program TOUGH AMD. This program has an equation of state for water, nitrogen and oxygen and a module representing the kinetics of the pyrite oxidation process. This module calculates the sinks of oxygen and the sources of heat related to pyrite oxidation. A vertical cross-section through the site which intersects the major waste rock types was used as the basis for a two-dimensional model. The relatively low permeability of the material makes diffusion an important oxygen transport mechanism. The global oxidation rate is thus relatively lower than in other waste rock piles where convection is dominant. However, the low heat conductivity of the material is responsible for a significant rise in temperature within the Nordhalde. This creates a temperature gradient within the pile causing advective gas flow despite the low permeability materials. Cyclic variations in surface boundary conditions can be modeled with TOUGH AMD. This feature was used to study the effect of yearly temperature variations and weekly barometric pressure changes on oxygen supply within the Nordhalde. Overall, the model allows a coherent representation of the conditions monitored within the waste rock pile and the confirmation of its physical properties. By being able to predict the longer term potential for AMD production at the site, the model contributes to the choice of the most appropriate approach to rehabilitation.

INTRODUCTION

The Ronnenberg mining district is located in the state of Thuringia in Germany (former German Democratic Republic). Extensive mining of Uranium, from open pit and underground workings, occurred there from 1950 through to 1990. This district is presently the object of a major environmental rehabilitation effort.

The Nordhalde is one of the largest waste rock piles in the area, consisting of approximately 27 million m^3 of material. The pile contains significant amounts of pyrite which is currently oxidizing, resulting in the generation of Acid Mine Drainage (AMD). The oxidation of pyrite is directly related to the consumption of oxygen and the production of heat. To better understand the rate and physical processes which govern pyrite oxidation in the Nordhalde, the pile was subject to extensive geochemical characterization and borehole instrumentation. The boreholes allow the monitoring of pore gas oxygen concentration, temperature and pressure through time.

The development of a numerical model for the Nordhalde was intended 1) to verify previous pyrite oxidation rate estimates from analytical solutions applied to temperature and oxygen profiles, 2) to get a better understanding of the physical mechanisms responsible for oxygen supply within the Nordhalde, and 3) to integrate and validate available information on physical properties of the material. The numerical model used was TOUGH AMD. That model represents the physical processes involved in AMD production in waste rock dumps (Lefebvre, 1994 and 1995). These capabilities were developed starting from the TOUGH2 simulator (Pruess, 1987 and 1991).

ACID MINE DRAINAGE (AMD) PROCESSES

Even though pyrite oxidation is a biochemical process, the generation of AMD in waste rock dumps involves much more than chemistry. Waste rock dumps comprise coarse mineral materials and are partially saturated media. A wide range of coupled physical processes are involved in these systems.

Within waste rock dumps, at the heart of all other processes is the oxidation of the pyrite present in the rocks. Oxidation involves oxygen consumption, even though the direct oxidant is often ferric iron (Fe^{3+}). A supply of oxygen is thus required to sustain pyrite oxidation. Heat production also occurs since pyrite (Py) oxidation is strongly exothermic (11,7 MJ/kg Py oxidized). The release of heat drives temperature up, as high as 70 °C in some places (Gélinas et al., 1994). This increase in temperature is important since it totally modifies the mechanisms responsible for oxygen transport to oxidation sites. Initially, in all systems, diffusion is the main process providing oxygen within waste rock accumulations. In materials of low permeability, such as in mine tailings, diffusion is believed to remain the only means of oxygen transport. However, in higher permeability materials, following an initial increase in temperature, temperature-driven convection currents are generated. The resulting advection is a much more efficient oxygen transport process than diffusion and sustains higher global oxidation rates. Finally, water infiltration occurs through the unsaturated porous material and picks up oxidation components to form an acidic leachate containing high concentrations of sulfate and metals.

In summary, waste rock dumps are complex systems posing an interesting problem of coupled physical processes: multiphase flow, heat transfer, and mass transfer in the liquid phase (advection) and in the gas phase (advection and diffusion). Numerical simulation is needed to handle all those processes and understand their interactions.

TOUGH AMD DESCRIPTION

The TOUGH AMD model is described elsewhere by Lefebvre (1994 and 1995). The model was developed in the context of a detailed study of a waste rock dump at Mine Doyon (Gélinas et al., 1994). Only the main features of the program are summarized here.

TOUGH AMD evolved from TOUGH2 (Pruess, 1987 and 1991) with the equation of state model for water and air (EOS3). Only the main changes made to TOUGH2 are mentionned. Three components are considered in TOUGH AMD: water and air subdivided in two components (oxygen and the other air gases). Oxygen has to be considered as a separate component because it is consumed by the oxidation reaction and its concentration affects the first-order kinetics. To define a system with three components and heat, four primary variables are required. The mass fraction of oxygen in air was thus added to the primary variables. As in TOUGH2, those variables differ depending on the number of phases present.

In waste rock dumps, pyrite is contained in rocks in which the pyrite grains are surrounded by other minerals. Pyrite oxidation proceeds from the surface of waste rock fragments. As pyrite near the surface is oxidized, the oxidant must penetrate within the blocks to reach unreacted pyrite. A zonation appears within waste rock fragments, showing an external zone in which pyrite is completely oxidized and an internal core where pyrite is unreacted.

Reaction core models provide a link between the pyrite oxidation surface reaction kinetics and the volumetric oxidation rate observed in waste rocks. They must account for the concentration and surface of pyrite exposed within waste rocks and consider the supply of oxidant within the blocks by diffusion. These models also establish a relationship between the reduction of pyrite fraction and its impact on the volumetric oxidation rate. A new reaction core model was developed and implemented in TOUGH AMD. The reaction core model computes the oxygen loss and heat production resulting from pyrite oxidation as a function of temperature, oxygen concentration and pyrite mass fraction.

New modifications were made to TOUGH AMD for the present study. The program was first migrated to Intel-based personal computers. New capabilities for representing sinusoidal variations in temperature and pressure in boundary elements were developed. This feature includes the internal calculation of static pressure profiles for boundary elements located at different elevations. Finally, output files were made compatible with the LBNL program EXT to allow the use of Tecplot for graphics.

PHYSICAL CONDITIONS AND PROPERTIES

The extensive monitoring and characterization program carried at the Nordhalde provides a unique opportunity to better understand physical processes in waste rock systems, especially the oxygen transport mechanisms.

The Nordhalde waste rock dump results from the extensive uranium mining operations which occurred in the area over a 40 year period. It contains pyritic slates covering an area of 100 ha at a height of more than 70 m. The dump consists primarily of Zone A, or strongly reactive, waste rock. Overlying this in part is an unintentional cover of Zone C material that is not producing AMD (Hockley et al., 199*).

Eight boreholes reaching depths between 8 and 75 m, were instrumented with transducers and sampling tubes at 2 m intervals. Downhole pressure and temperature were measured every hour. Oxygen concentration measurements daily. Monitoring began in July, 1996. This study considers a full year of data from the site. Figure 1 shows a monthly sampling of profiles measured in borehole 38. Temperature (Fig. 1a) is seen to increase well above the average surface temperature of 9 °C, indicating active heat production by pyrite oxidation. The oxygen profiles (Fig. 1b) are more telling of the processes active in the pile throughout the year. During summer months, oxygen patterns are typical of a diffusion process, showing an exponential decrease with depth. Short term barometric changes affect this pattern slightly, moving the profiles vertically upwards and downwards. During winter months, oxygen profiles become erratic, showing an overall increase in oxygen concentration at depth, especially near the edge of the pile. However, O_2 depletion occurs near the surface. This is indicative of thermal convection from the edge to the summit.

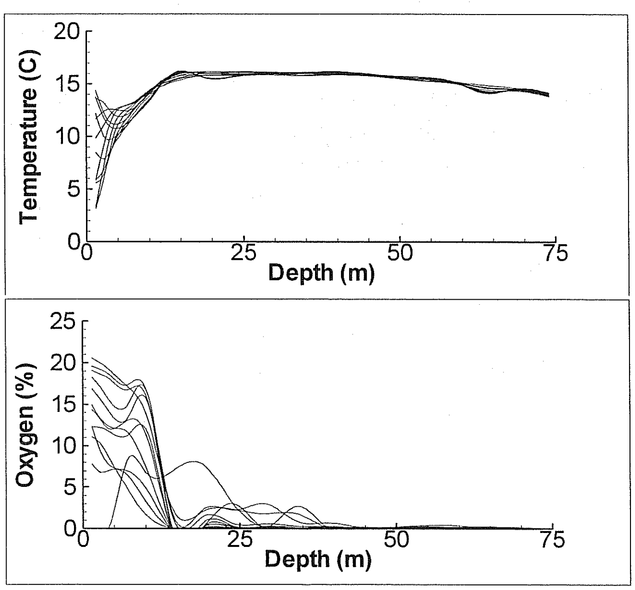

Figure 1. a) Temperature and b) oxygen profiles in Nordhalde borehole 38 in 1997

The monitoring program and numerous physical measurements in the field and laboratory allow a relatively precise evaluation of the global physical properties of materials in the Nordhalde. Table 1 summarizes the parameters required for the numerical model. These properties were determined from an hydrological evaluation of the infiltration rate, hydraulic conductivity tests, and measurements of grain sizes, global density and water content. Vertical air permeability was determined by matching pressure changes within the Nordhalde after barometric pressure variations with a SRK internal numerical model. Likewise, heat diffusivity was obtained by matching cyclic near-surface temperature variations with an internal SRK model. Initial values of the oxidation rate were derived from a diffusion and heat conduction analytical model of pyrite oxidation matched to temperature profiles.

Table 1. Physical properties of the Nordhalde

Property	Symbol and values
Global oxidation constant	$K_{ox} = 10^{-8}$ s^{-1}
Diffusive/Chemical times	$\tau_d/\tau_c = 0$
Pyrite mass fraction	$w_{py} = 0{,}031$
Horizontal permeability	$k_h = 8\text{x}10^{-11}$ m^2
Vertical permeability	$k_v = 8\text{x}10^{-12}$ m^2
Porosity	$n = 0{,}30$
Solids density	$\rho_s = 2751$ kg/m^3
Dry thermal conductivity	$K_{th,d} = 0{,}25$ W/m·°C
Wet thermal conductivity	$K_{th,w} = 1{,}2$ W/m·°C
Heat capacity of solids	$c_{ps} = 710$ J/kg·°C
Thermal conduct. of base	$K_{th} = 1{,}2$ W/m·°C
Global density of base	$\rho_b = 2106$ kg/m^3
Heat capacity of base	$c_p = 1021$ J/kg·°C
Standard diffusion coef.	$D_o = 4\text{x}10^{-5}$ m^2/s
Temperature diffusion coef.	$\theta = 1{,}8$
Tortuosity factor	$\tau = 0{,}85$
van Genuchten *m* factor	$m = 0{,}256$
van Genuchten α factor	$\alpha = 0{,}00036$ Pa^{-1}
Residual water saturation	$S_{wr} = 0{,}284$

THE NORDHALDE NUMERICAL MODEL

Figure 2 shows the numerical model grid used to represent the Nordhalde. TOUGH AMD considers gas convection and diffusion that affect oxygen supply, heat transfer by conduction, fluid advection and gas diffusion, and water infiltration from the surface by an imposed value of saturation. Infiltration affects heat transfer and leachate production – which is not discussed here. Pyrite oxidation is represented by a reaction core model but no limiting effect of diffusion is considered. Since the material is relatively fine grained, pyrite is supposed mostly free in the wastes and exposed to oxygen.

A one-dimensional vertical model was used to verify initial model parameter values and boundary conditions. Since such a model cannot represent convection properly, the temperature reached in this model was below the observed values at the site. However, the results of the one-dimensional model was useful in developing the two-dimensional model.

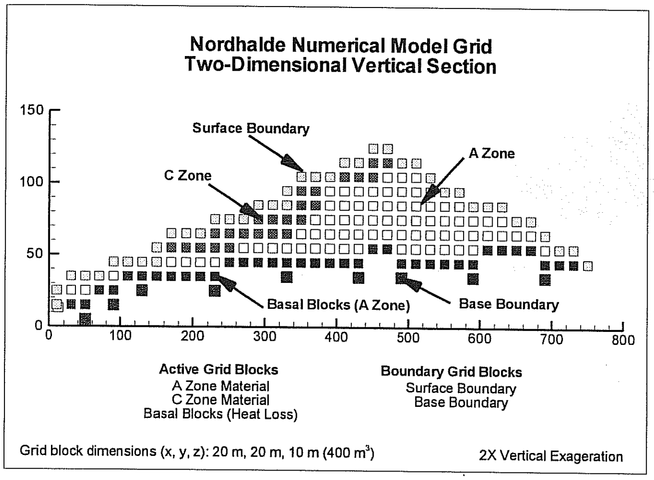

Figure 2. Numerical model grid located in a transverse cross section through the Nordhalde

Figure 2 shows the transverse vertical section used in the two-dimensional model. The grid blocks are uniform in dimensions: 10 m high with a 20x20 m base for a total of 135 active elements. A section is necessary to represent gas convection and diffusion processes. Since gas exchanges should be limited in the longitudinal direction of the heap, a three-dimensional model was not judged necessary to properly represent the most important oxygen transport features of the Nordhalde.

The surface boundary (41 non active elements) had imposed pressure, water saturation (sets infiltration rate), oxygen concentration and temperature. Yearly cyclic surface temperature were used for all runs. Variable pressure conditions were used only in special short term runs to study their general effect.

Active elements represented material with Zone A (reactive) and Zone C (low reactivity) properties. The bottom layer of the waste rock materials allowed heat loss to the base computed with a semi-analytical module adapted from TOUGH2. The base boundary is only made up of a few (9) saturated inactive elements allowing the drainage of the infiltrating water. These elements have a very low heat conductivity so that no heat is lost to them.

CONDITIONS AND GAS FLUX WITHIN THE NORDHALDE

Figure 3 shows the results obtained from the model after 30 years, corresponding to the period of operation of the Nordhalde. The model shows a significant increase in temperature, reaching nearly 20 °C, very similar to observed values. To get such an increase, some of the oxygen has to be supplied by advection. This advection occurs under the temperature gradient established between atmospheric and internal gas. However, convection is triggered only if permeability is sufficiently high. Initial runs with slightly lower values of permeability would not allow advection to start and temperatures remained well under observed values (at about 12 °C). The Nordhalde is thus at the limit between conditions that would imply a totally diffusive system and a convective system. This conclusion is supported by field indications of increased convection under winter conditions.

Even though convection occurs, diffusion is still an important oxygen supply mechanism. In Figure 3 (middle), oxygen fluxes are seen to be mostly perpendicular to oxygen concentration profiles, as would be expected from a diffusive process. The combination of oxygen transport processes cannot counteract oxygen depletion by pyrite oxidation. Oxygen concentrations are thus only important near the surface of the heap whereas the core is totally depleted in oxygen. Oxygen fluxes are seen to be higher on the edges of the heap than at the surface. This is explained by the upward advection of gas with a reduced oxygen content that counteracts downward diffusion.

The Zone C material has some impact on the global oxidation rate. This is seen on the lower part of Figure 3. Lower temperatures and lower oxidation rates are encountered where Zone C material is present. Low reaction kinetics were used to model this material. This is supported by observed oxygen and temperature profiles. In boreholes going through Zone C material, temperature profiles show straight line increases and oxygen remains high, indicating that oxidation is not significant. Uncertainties remain however about the exact reactivity of this material.

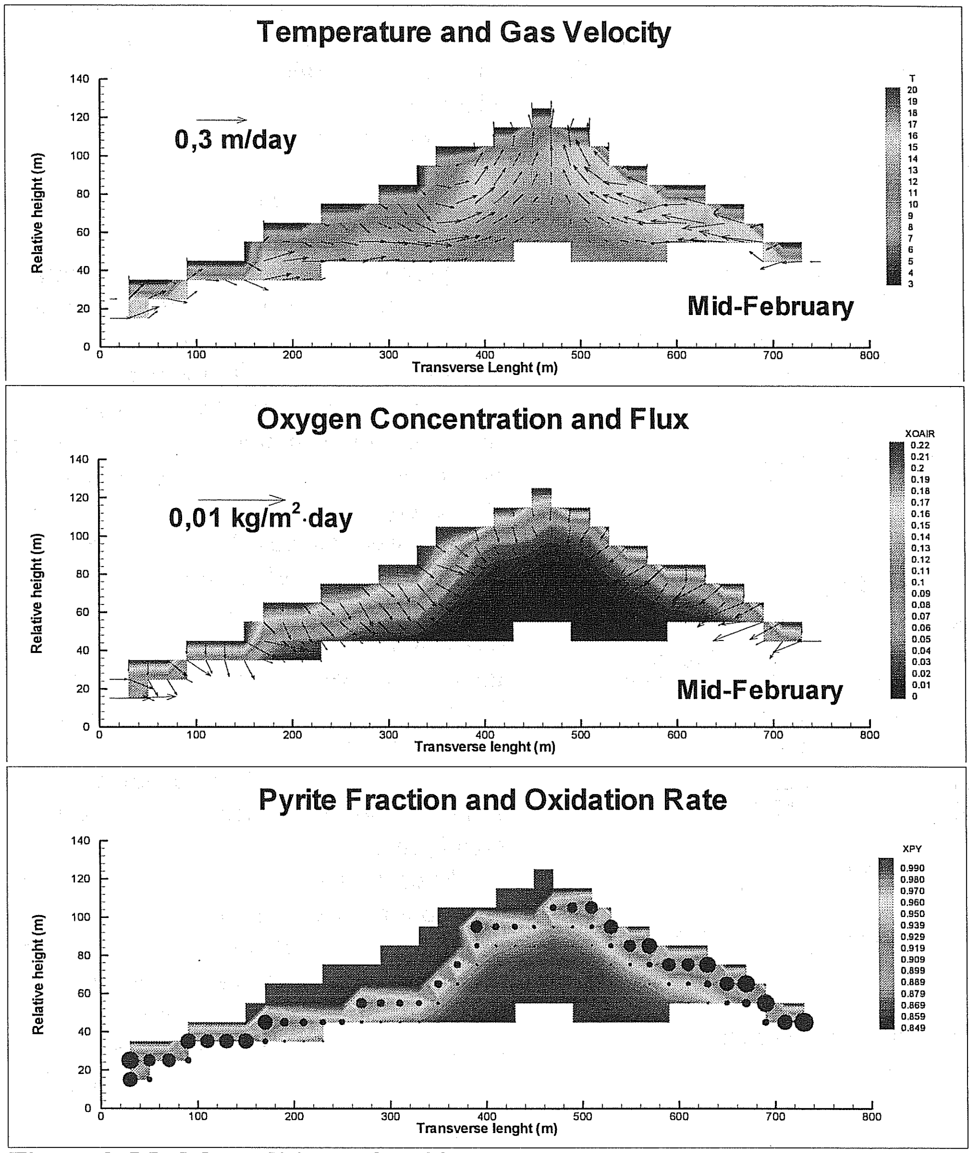

Figure 3. Model conditions after 30 years. Top: Temperature and gas velocity Middle: Oxygen concentration and flux. Bottom: Pyrite remaining and oxidation rate

CYCLIC BOUNDARY CONDITIONS

The fact that physical conditions predicted by the model are representative of observation would indicate that the physical processes operating in that system are globally well described, especially the ones responsible for oxygen supply. Since observations indicate that seasonal changes occur in oxygen concentrations, we were interested in modeling the effect of variable boundary conditions. This prompted the development of such capabilities for the model.

The first effect modeled was the yearly cyclic temperature variations. Since average temperature varies slowly, the use of cyclic surface temperature did not imply a high computational burden. However, barometric pressure changes are much more rapid and more erratic. Thus, variable boundary pressure was not included like temperature in the long term modeling runs. Instead, a short term run was used to study the effect of variable surface pressure. The conclusions from these modeling efforts are not yet definitive.

Figures 4 and 5 illustrate the modeled evolution of global average conditions within the Nordhalde using variable surface temperature. During the modeling period, there is a steady increase in temperature with indications of some leveling off at the end. Figure 4 is quite interesting because it indicates the onset of convection. The break in slope of the average oxygen concentration beyond 5 years occurs when the systems goes from being dominated by diffusion to one in which convection plays a more important role, as revealed by graphs like figure 3 for this time. After this time, the more steady oxygen profile is related to well established convection.

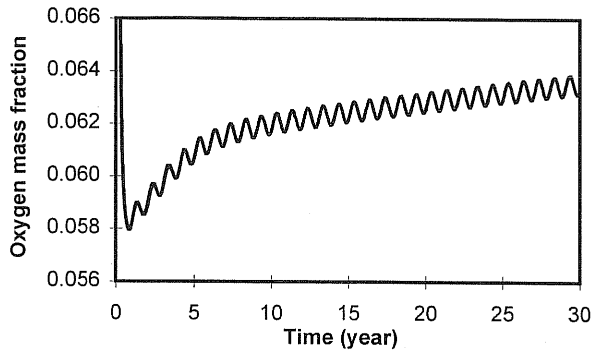

Figure 4. Evolution of average oxygen concentration in gas phase

The relatively steady increase in oxygen concentration shown in figure 4 does not equate to higher average oxidation rates. On the contrary, figure 5 shows that oxidation rates are globally decreasing. This is caused by the depletion of readily accessible pyrite. The reaction core model involves a reduction in the "reactivity" of the material as pyrite gets depleted. This counteracts the increase in oxygen.

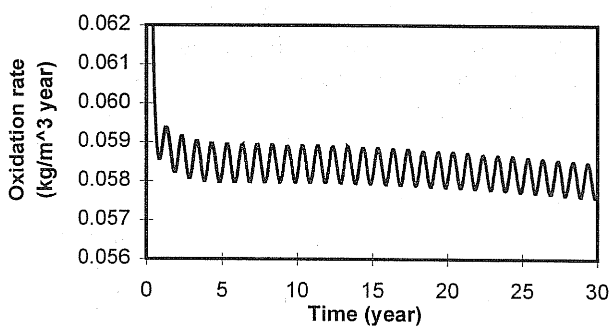

Figure 5. Evolution of the average volumetric oxidation rate (oxygen mass consumption)

Figure 6 shows the result of a short term modeling run with variable barometric pressure. Comparing figures 4 and 6, we see that important atmospheric pressure variations induce short term changes in average oxygen concentration. This effect is thus potentially important as an oxygen supply mechanism. Graphs similar to figure 3 show that convection patterns are actually modified quite significantly by variations in barometric pressure. More work needs to be done to fully understand the interaction between temperature and pressure fluctuations that influence convection patterns within the Nordhalde and its impact on oxygen supply and pyrite oxidation rate.

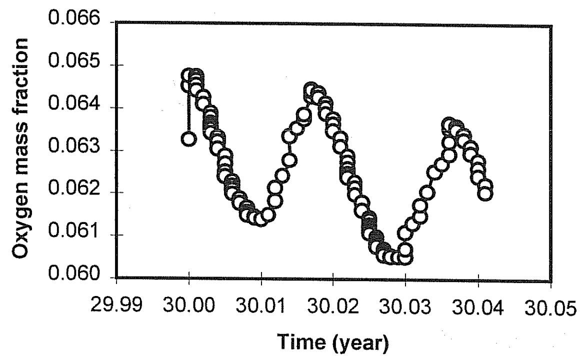

Figure 6. Variations in oxygen concentration in the gas phase during barometric pressure changes

CONCLUSION

The Nordhalde is an important site and can teach us a great deal about the oxygen supply mechanisms in AMD-producing mined materials. This site is exceptional by the quality of the characterization and monitoring program. Also, the low permeability of the material allow us to better understand the conditions necessary to trigger convection. This site actually shows evidence of convection under conditions that were previously believed to be unfavorable.

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AKNOWLEDGEMENTS

This paper is presented by kind permission of WISMUT GmbH.