

# NUMERICAL MODELING OF AMD PRODUCTION IN WASTE ROCK DUMPS<sup>1</sup>

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**Abstract:** A numerical model was developed to represent the main processes involved in acid mine drainage (AMD) production in waste rock dumps: hydrology, gas transfer, heat transfer, geochemistry and mass transport. The model is based on the multicomponent multiphase non isothermal general numerical simulator TOUGH2 and is thus termed TOUGH AMD. Brief descriptions are presented of the reaction core model developed for the model as well as the numerical model formulation and characteristics. Model capabilities are illustrated by an application using the physical properties of the South Dump at La Mine Doyon. Modeling results show that air convection controls the AMD production rate. The model also allows a better understanding of the coupled physical processes involved in AMD in waste rocks.

**Key Words:** Acid mine drainage, waste rocks, numerical modeling, physical processes, reaction core model, air convection, unsaturated flow, heat transfer, mass transfer.

## Introduction

Our objectives for AMD numerical modeling are 1) to provide a better understanding of the physical processes involved in AMD, 2) to allow the integration of available waste rock characterization data, 3) to indicate which new data or studies are required to fill the gaps in our quantitative understanding of AMD processes, and 4) to supply a tool for the prediction of the evolution of AMD production with time taking into account the impact of control methods. These objectives can only be met following sustained research efforts. This study is part of a larger research effort which has been on-going at La Mine Doyon since 1991 (Gélinas et al. (2)).

## Reaction core model

### General concepts

In waste rock dumps, pyrite is contained in rock blocks and surrounded by other minerals, contrary to mine tailings in which pyrite grains are mostly free. Pyrite oxidation proceeds from the surface of waste rock blocks. As pyrite near the surface is oxidized, the oxidant must penetrate within the blocks to reach unreacted pyrite. A zonation appears within waste rock blocks with an external zone in which pyrite is completely oxidized and an internal core where pyrite is unreacted. The whole of pyrite within a block is not oxidized simultaneously because the rate of oxygen consumption generally exceeds the rate of oxygen diffusion in the blocks. Cathles and Schlitt (1) mention that the general validity of reaction core models is supported by field observations of waste rock blocks in which pyrite has disappeared in their external portion but is unreacted within their core. The same observation is made at La Mine Doyon where cubic holes at the surface of waste rock blocks indicate that pyrite has been oxidized whereas pyrite is still present inside the blocks and shows no signs of oxidation.

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Reaction core models provide a link between the pyrite oxidation surface reaction kinetics and the volumetric oxidation rate 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. That relationship allows a determination of the time required to oxidize a given mass fraction of the pyrite initially present within the waste rocks.

Like Pantelis and Ritchie (9), our reaction core model uses only oxygen as the effective pyrite oxidant thus making a direct link between oxygen concentration in the gas phase and the oxidation rate. This also avoids the need for leachate speciation which would have to be made if ferric iron were also directly considered as an oxidant. The model considers the effects of both the surface reaction kinetics and the rate of oxygen diffusion as controls on the volumetric oxidation rate as was used by Cathles and Schlitt (2), Jaynes (3) and Levenspiel (5). Pantelis and Ritchie (9), who used only diffusion as a control on volumetric oxidation, obtained a formulation with a mathematical singularity at initial conditions. The volumetric oxidation rate is also supposed independent of the geochemical conditions within the leachate, such as pH, Eh and ions concentrations. This same supposition was made by Cathles and Schlitt (1) as well as Pantelis and Ritchie (9) to avoid having to perform speciation calculations as in the model of Jaynes (3).

Certain parameters limiting the oxidation rate were included in kinetic factors formulated in a way similar to Jaynes (3). High temperatures were supposed limiting bacterial activity and the oxidation rate. Otwinowski (8) mentions that this effect could be accounted for by the reduction in oxygen solubility at high temperatures. So, even though oxygen solubility is supposed constant in the numerical model, the temperature kinetic factor indirectly accounts for this effect. A minimal oxygen concentration was also prescribed as done by Jaynes (3) to account for the minimum oxygen content required for bacterial activity.

### Formulation of the reaction core model

The first order kinetic constant  $k_{Ox}$  (m/s) represents the rate of oxygen consumption during the surface oxidation reaction of pyrite and has a relatively constant and well determined value (Otwinowski, (8)). However, one has to distinguish between the surface oxidation rate and the volumetric oxidation rate  $Q_{Ox}$  (kg/m<sup>3</sup> s) observed in a unit volume of waste rocks. This last rate is related not only to surface oxidation but is also controlled by the oxygen diffusion rate in waste rock blocks. The reaction core model provides a relationship to determine this rate:

$$Q_{Ox} = -K_{Ox} \cdot X_T \cdot X_{WO} \cdot \rho_{Ox}^{air} \cdot f(X) , \quad (1)$$

where  $K_{Ox}$  is the global volumetric kinetic constant (s<sup>-1</sup>),  $X_T$  is the temperature kinetic factor (0 to 1),  $X_{WO}$  is the oxygen partial density in the liquid phase kinetic factor (0 to 1),  $\rho_{Ox}^{air}$  is the oxygen partial density in the air (kg/m<sup>3</sup>), and  $f(X)$  is the geometric factor which is a function of the proportion of pyrite remaining  $X$  (0 to 1).

The kinetic constant  $K_{Ox}$  is here a global parameter related to the volumetric oxidation rate in a unit volume of waste rocks having a porosity  $n$ . This parameter takes into account the surface kinetic constant  $k_{Ox}$  (m/s), the surface area of pyrite per unit volume of rock  $a_{Py}^{rock}$  (m<sup>-1</sup>), the conversion factor between the partial densities of oxygen in the gas and liquid phases  $\gamma$  (-), the thickness of the layer in which pyrite oxidation takes place  $\delta$  (m) and the radius of waste rock blocks  $R$  (m):

$$K_{Ox} = \frac{3 (1-n) k_{Ox} a_{Py}^{rock} \gamma \delta}{R} , \quad (2)$$

The geometric factor  $f(X)$  takes into account the kinetic effect of the reduction in the proportion of unreacted pyrite  $X$  in waste rock blocks. The larger the proportion of reacted pyrite, the further has oxygen to diffuse within the blocks to reach the reaction layer, thus causing a reduction in the volumetric oxidation rate. This factor thus takes into account the relative effects of surface oxidation and oxygen diffusion on the reaction kinetics. These effects are represented respectively by the total time required to fully oxidize the pyrite in waste rock blocks 1) if only surface oxidation is considered  $\tau_c$  (s) or 2) if only oxygen diffusion is considered  $\tau_d$  (s):

$$f(X) = \frac{X^{2/3}}{6 \tau_d/\tau_c X^{1/3} (1-X^{1/3}) + 1} , \quad (3)$$

where

$$\tau_c = \frac{\rho_{Py}^{rock} R}{b k_{Ox} a_{Py}^{rock} \delta \gamma \rho_{Ox}^{air}} \quad \text{and} \quad \tau_d = \frac{\rho_{Py}^{rock} R^2}{6 b D_e \gamma \rho_{Ox}^{air}} , \quad (4)$$

where  $\rho_{Py}^{rock}$  is the pyrite partial density in rocks ( $\text{kg/m}^3$ ),  $b$  is a conversion factor between the masses of oxygen and pyrite consumed (-), and  $D_e$  is the effective diffusion coefficient of oxygen in the blocks ( $\text{m}^2/\text{s}$ ).

### Numerical methods used in TOUGH AMD

#### Physical processes and equation system

This theoretical development is based on the ones by Pruess (11, 12) describing the formulation used in TOUGH and TOUGH2. However, the modifications required to develop TOUGH AMD are included here, mainly the addition of a new component (oxygen) and a new primary thermodynamic variable (oxygen mass fraction in air).

Continuity equations express the principle of conservation of mass and energy. In TOUGH AMD, these equations are written in integral form for an arbitrary flow domain  $V_n$  in which mass and energy must be balanced for the four components  $\kappa$  of the system. There are three mass components ( $\kappa=1$ : water,  $w$ ;  $\kappa=2$ : gases other than oxygen in air,  $a$ ;  $\kappa=3$ : oxygen,  $o$ ) and one energy component ( $\kappa=4$ : heat,  $h$ ). The following relationship expresses that the changes in time of the accumulation  $M^\kappa$  of component  $\kappa$  in the volume  $v$  considered has to be equal to the sum of fluxes of the component  $F^\kappa$  in the direction  $n$  perpendicular to the domain surface  $\Gamma$  and the sources of that component  $q^\kappa$  in the volume considered:

$$\frac{d}{dt} \int_{V_n} M^\kappa dv = \int_{\Gamma_n} F^\kappa \cdot n d\Gamma + \int_{V_n} q^\kappa dv . \quad (5)$$

Four equations, one for each component, thus have to be satisfied simultaneously to solve the system. The accumulation terms  $M^\kappa$  represent the storage capacity for mass ( $\text{kg/m}^3$ ) or energy ( $\text{J/m}^3$ ) in the fluids and solids of the system. The mass flux of fluid components and of energy

depends on the advection and diffusion transfer processes. The advective mass flux of phase  $\beta$  (liquid or gas) is determined by the general form of Darcy's law for multiphase flow multiplied by the phase density. Mass transfer of a component in the gas phase by diffusion is determined by Fick's law corrected for unsaturated porous media.

The pyrite oxidation reaction produces sinks and sources of certain components. It produces heat and consumes oxygen which are components of the system. Furthermore, that reaction consumes pyrite and produces sulfate, iron and acidity. The pyrite remaining has to be taken into account since it affects the reaction rate. The production, accumulation and transport of sulfate is also followed to represent AMD production outside the dump. The reaction core model is used to calculate the global volumetric oxygen consumption rate  $Q_{O_x}$  (kg/m<sup>3</sup> s) in a unit volume of waste rocks, to which is related the heat production rate through the reaction enthalpy.

The relationship (5) presented applies to continuous systems and needs to be discretized to allow a numerical solution. In TOUGH2, these equations are discretized in space by the method of integral finite differences (Narasimhan and Witherspoon (7); de Marsily (6)).

### **TOUGH AMD general capabilities**

TOUGH AMD is adapted to the modeling of AMD and allows the representation of the main processes involved in waste rocks:

- **Hydrology**: Takes into account variable infiltration with time from the surface and represents unsaturated and saturated liquid flow within the dump.
- **Gas transfer**: Represents gas convection under pressure or temperature gradients as well as the diffusion of gas components under concentration gradients.
- **Heat transfer**: Includes the processes of conduction, fluid advection and gas diffusion. Takes into account the effect of phase changes on heat transfer. Allows the calculation of heat losses to confining beds.
- **Geochemistry**: Uses a reaction core model including the effects of surface reaction and diffusion on the volumetric oxidation rate. Computes the rates of consumption of pyrite and oxygen and the rates of heat and sulfate production.
- **Mass transport**: Uses sulfate to represent AMD production. Computes the production and the advective transport of sulfate. Computes the rates of mass accumulation within the dump as well as its release outside the dump.

### **Application of AMD numerical modeling**

The first objective of numerical modeling is here to better understand the physical processes involved in AMD production in waste rocks. A general case is modeled to study the interaction of processes and identify the parameters having the greatest impact on the behavior of the system. The parameters and conditions used in the base case are representative of the South Dump at La Mine Doyon so that results may be compared to observations at this site (Gélinas et al. (2)). We are presenting here some of the findings of the modeling work for a base case. The application of the model to the evaluation of a control method, a border membrane, as well as a more detailed account of the modeling work is presented by Lefebvre (4).

### **Model parameters and conditions**

The base case is modeled for 15 years and as such includes the period during which the dump has been in place (about 9 years) and provides a look at the potential future evolution of AMD production. It would be unrealistic to model the behavior of the dump for a longer period given the uncertainty in our knowledge of the site and in particular the absence of data on the evolution of physical properties in time which could affect significantly the behavior of the dump. This model actually supposes that physical properties remain constant even though we know from

field observations that the material evolves and new minerals are formed and could alter the physical properties. Further studies are required to quantify the impact of this change in physical properties. The extensive characterization and monitoring program at La Mine Doyon allowed an evaluation of many physical properties of the waste rocks (Gélinas et al. (2)). Table I summarizes the physical parameters used in the model.

Table I - Physical properties of the base case.

Property	Symbol, value and units
Volumetric oxidation constant	$K_{ox} = 0.75 \times 10^{-6} \text{ s}^{-1}$
Diffusive / Chemical total times	$t_d/t_c = 2.5$
Pyrite mass fraction in solids	$w_{py} = 0.07$
Horizontal permeability	$k_h = 2.5 \times 10^{-9} \text{ m}^2$
Vertical permeability	$k_v = 1.0 \times 10^{-9} \text{ m}^2$
Porosity	$n = 0.33$
Solids density	$\rho_s = 2740 \text{ kg/m}^3$
Dry thermal conductivity	$\lambda_d = 0.9 \text{ W/m } ^\circ\text{C}$
Saturated thermal conductivity	$\lambda_w = 3.7 \text{ W/m } ^\circ\text{C}$
Heat capacity of solids	$c_{ps} = 837 \text{ J/kg } ^\circ\text{C}$
Thermal conductivity of the base	$\lambda = 1.55 \text{ W/m } ^\circ\text{C}$
Global density of the base	$\rho_b = 2008.6 \text{ kg/m}^3$
Heat capacity of the base	$c_p = 1504 \text{ J/kg } ^\circ\text{C}$
Standard diffusion coefficient	$D_o = 2.13 \times 10^{-5} \text{ m}^2/\text{s}$
Temperature diffusion coef.	$\theta = 1.80$
Tortuosity factor	$\tau = 0.7$
van Genuchten "m" factor	$m = 0.23$
van Genuchten "α" factor	$\alpha = 0.504 \text{ Pa}^{-1}$
Residual water saturation	$S_{wr} = 0.14$

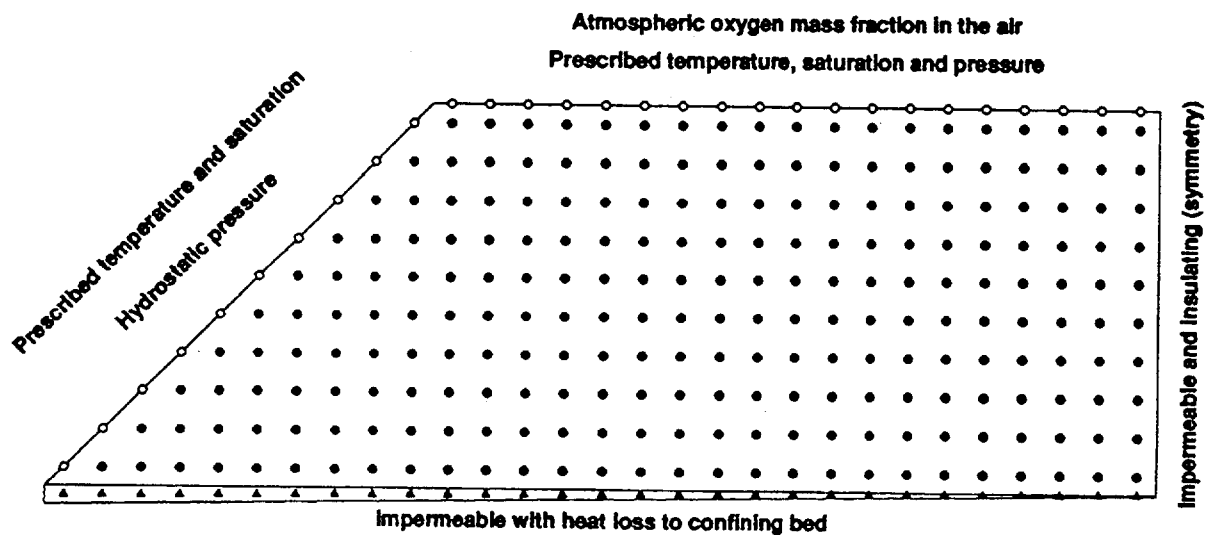


Figure 1 - Computation grid and limit conditions for the base case.

The computation grid and limit conditions used in the base case are presented in figure 1. That grid represents a vertical half section within the dump and uses cartesian coordinates. The model has an unsaturated zone 30 m thick and a thin wedge-shaped saturated zone from zero to 1.5 m thick. Lateral dimensions are 87 m at the base and 57 m at the surface. The slope at the border is necessary to obtain representative air convection patterns as shown by Pantelis and Ritchie (10). Since the internal limit is a symmetry limit, the grid represents a system twice as wide. Those dimensions are representative of the south part of the main zone at the South Dump.

The limit conditions used are a water saturation of 0.42 generating an infiltration of about 35 cm per year, a temperature of 5 °C and atmospheric oxygen mass fraction (0.2315). Atmospheric pressure is set at 100 kPa at the surface whereas a hydrostatic pressure profile is used at the sloping border. The system is supposed to be homogeneous. These conditions are also used as initial conditions within the dump at time zero. The following sections summarize the main conclusions of the modeling runs using these conditions.

### **Physical conditions within the dump**

Figure 2 shows the conditions prevailing in the dump after 9 years of AMD production: a) gas velocity and temperature, b) oxygen mass flux and relative concentration, and c) oxidation rate and remaining pyrite mass fraction. The key process in AMD production is oxygen supply to oxidation sites. Convection can provide oxygen in a much more efficient way than diffusion.

Figure 2a shows that air convection is controlled by temperature gradients. An increase in temperature reduces density and causes gas movement. Since temperature is higher within the dump than outside, the gas column is lighter in the dump and the gas pressure will be smaller thus forcing air entry inside the dump at the border. Within the dump, this light gas has a tendency to move upward and emerges at the top surface of the dump. A convection movement is thus initiated which is responsible for the supply of oxygen within the dump. Gas convection at the border generates an auxiliary convection pattern from the inner top surface of the dump to the base and then upward through the zone of highest temperature.

Figure 2b illustrates the oxygen mass flux and the relative oxygen concentration in the dump (with respect to atmospheric oxygen concentration) after 9 years of AMD production. The most important source of oxygen supply is through the border and is related to air convection which provides oxygen from the outside of the dump. The oxygen flux is reduced along a given convection path within the dump because it is gradually consumed by the oxidation reaction. The oxygen concentration is thus maximum near air entry points and is gradually reduced within the dump. The slower convection path from the inner top surface of the dump does not supply oxygen fast enough so it is totally consumed before reaching the base of the dump. The central core of the dump is thus deprived of oxygen and does not produce much AMD. The effect of diffusion is only apparent at the central surface of the dump where gas flow is horizontal and contains little oxygen provided from the surface of the dump by diffusion.

Since pyrite oxidation is supposed to follow first order kinetics with respect to oxygen, its distribution has a direct impact on the oxidation rate. Figure 2c shows, after 9 years of AMD production, the remaining pyrite mass fraction (contours) and the oxidation rate (circles). Of course, the highest oxidation rates are observed in areas of maximum oxygen concentration at the border of the dump and near its surface. The remaining pyrite mass fraction follows similar patterns which are generally perpendicular to the direction of oxygen supply. There is a sharp decrease in pyrite at the border of the dump whereas the reduction is not as important at the surface of the dump. In the central part of the dump, very little pyrite is oxidized and a large potential for further AMD production remains.

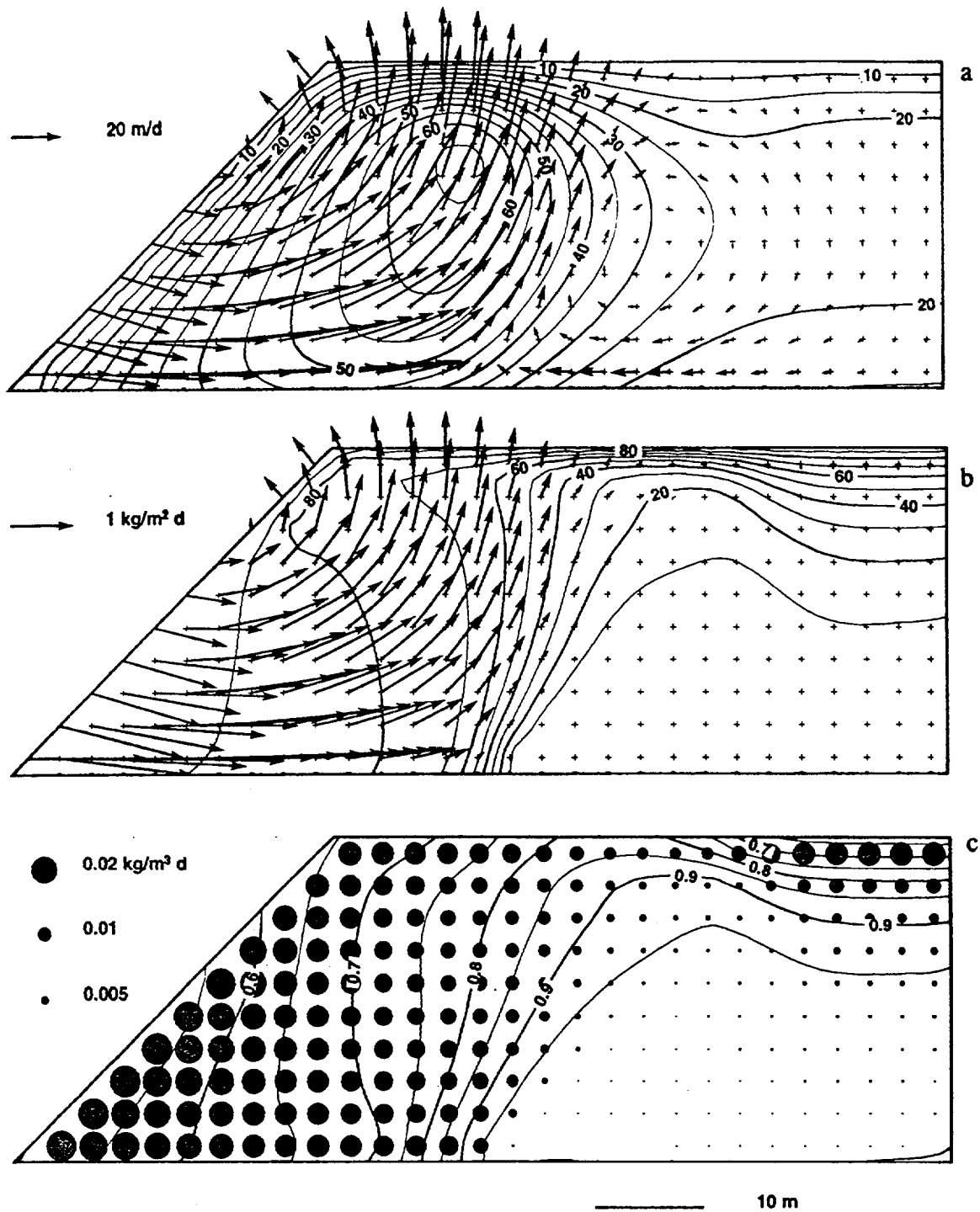


Figure 2 - Conditions for the base case after 9 years of AMD production. a) Gas velocity (m/d) and temperature ( $^{\circ}\text{C}$ ). b) Oxygen mass flux ( $\text{kg}/\text{m}^2 \text{ d}$ ) and oxygen mass fraction in air with respect to atmospheric value (%). c) Unreacted pyrite mass fraction and oxidation rate ( $\text{kg O}_2/\text{m}^3 \text{ d}$ ).

## Physical transfer processes

Transfer processes play an important role in AMD production. We will discuss here numerical modeling results providing more insights on the processes of heat transfer, water circulation and sulfate transport in waste rock dumps.

Heat transfer in waste rock dumps occurs through the mechanisms of 1) conduction in the bulk of the material resulting from temperature gradients, 2) advection of fluids (liquid and gas) carrying heat and 3) diffusion of components in the gas phase. Figure 3a shows, after 9 years of AMD production, the temperature distribution and the components of heat transfer. Three families of arrows represent respectively 1) the total heat flux, 2) the conductive heat flux (perpendicular to temperature contours) and 3) the diffusive heat flux. The difference between the total flux and the other components is the advective heat flux. At the outer limits of the dump, heat conduction is generally the dominant heat transfer mechanism. Conduction is also responsible for heating the center of the dump by carrying heat away from the zone of maximum temperature. Heat transfer by gas advection is dominant in the central hot zone where the gas phase contains more water vapor and carries a lot of heat. Water advection and its effect on heat transfer is very small in this case because of the low infiltration rate. However, this process can be significant in other circumstances. Heat transfer by diffusion in the gas phase is negligible because it is a binary process with diffusion and counterdiffusion of components with nearly equal heat capacity.

It is important to understand the modes of water circulation in a waste rock dump since the mass produced during AMD (sulfate, acidity, metals) is carried in solution. Figure 3b shows, after 9 years of AMD production, the distribution of water saturation and the mass fluxes of water in the liquid (downward pointing gray arrows) and vapor phase (black arrows). Liquid water moves downward from the surface during infiltration in the unsaturated zone. Water is also transferred in the vapor phase by gas advection. Water transfer in the vapor phase is actually responsible for the redistribution of water within the dump. Relatively dry cold air entering the dump, mainly at the toe of the slope, acquires more and more water as it first moves horizontally and heats up. The gas phase carries the most water, as much as the liquid phase, as it moves upward through the hottest zone. Beyond that zone, in cooler areas near the surface, water vapor condenses and contributes to more infiltration. That process induces water saturation variations within the dump from 37.5% to 41.5%. The zone most influenced is near the toe of the slope at the base where water is withdrawn. Although important, the process of water vapor transfer by advection does not lead to important water losses from the dump because most of the water withdrawn from the base recondenses near the top surface before the gas phase leaves the dump.

AMD production is followed in the model by sulfate production, transport and release. Figure 3c shows the distribution of sulfate concentration (contours) and the net sulfate flux in the grid elements. These net fluxes represent the difference in mass of sulfate entering and leaving an element. Net negative fluxes (dark circles) indicate that more mass exits the element than enters it whereas positive net fluxes (light circles) have an opposite meaning. Negative net fluxes occur in zones of high oxidation rate and consequently of high sulfate production. In these areas, more mass leaves the elements because internal production adds to the sulfate mass flux. Positive fluxes occur mainly at the core of the dump where sulfate mass production is low: in these areas, leachate of high concentration mixes with the leachate contained in the elements and comes out with less mass. This type of mass transport representation in the unsaturated zone supposes mixing of fluids instead of piston displacement and accounts for the limited leaching caused by small infiltration rates compared with the large volume of leachate stored in the unsaturated zone. This results in an increase in concentration and sulfate mass storage within the dump as more mass is produced than may be leached during the early years of AMD production. This representation of mass transport is simplistic and requires more work. Also, no account is taken in the model of mass loss due to new minerals precipitation (gypsum, jarosite). It is clear that the very high concentrations indicated by the model could not be reached in a real system because oversaturation and precipitation would occur to limit the concentration.

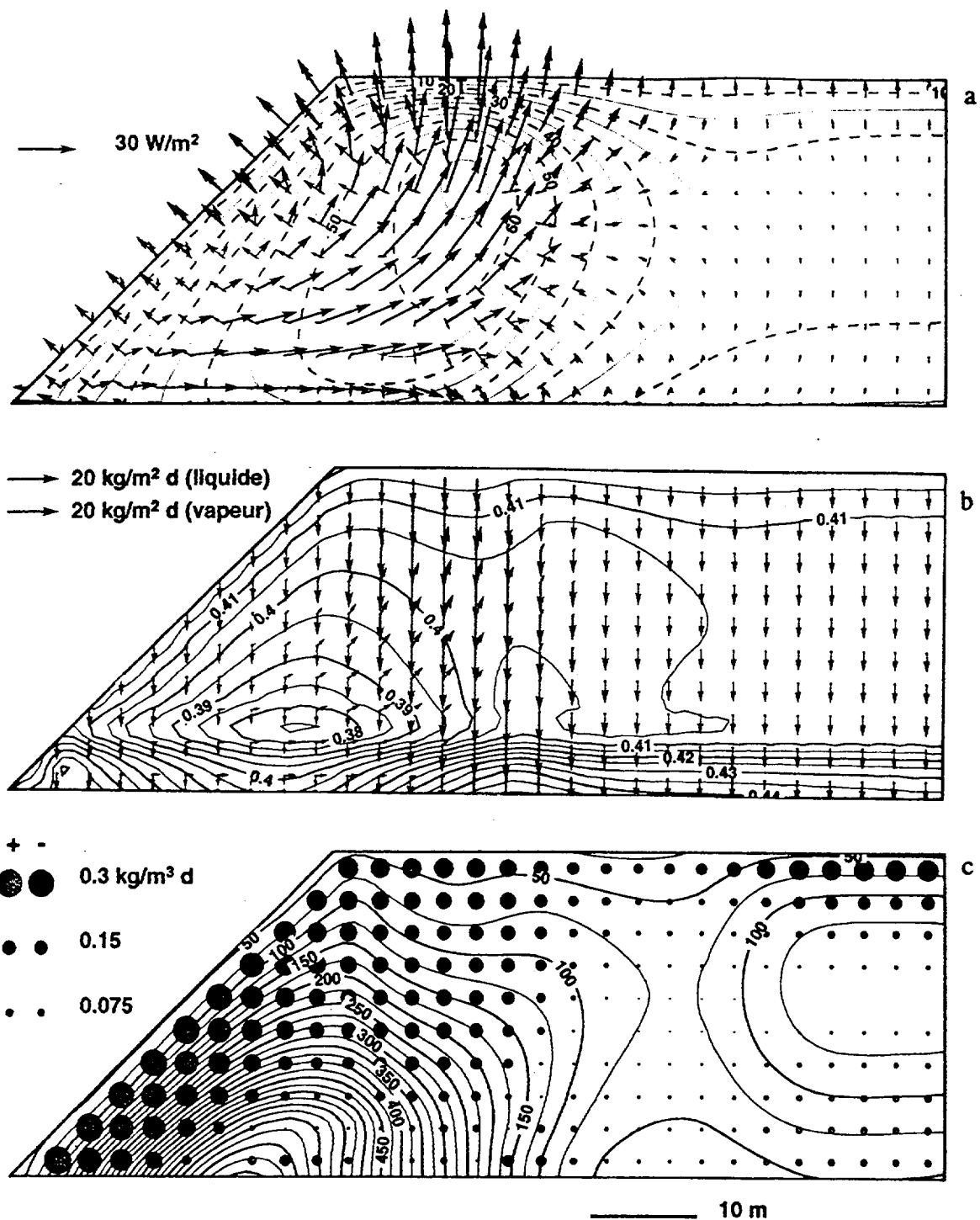


Figure 3 - Physical processes for the base case after 9 years of AMD production. a) Total, conductive and diffusive heat fluxes ( $W/m^2$ ) and temperature ( $^{\circ}C$ ). b) Liquid (gray) and vapor (black) water mass fluxes ( $kg/m^2 d$ ) and water saturation (-). c) Net sulfate flux ( $kg/d$ ) (negative: dark, positive: light) and concentration ( $g/L$ ).

## Conclusion

Some observed differences between the model predictions and field observations point out to limitations in the numerical model and in our general knowledge of some key processes related to AMD production. Further research is needed to develop the firm fundamental knowledge upon which applied numerical modeling must be based. The model is still useful to better understand coupled physical processes involved in AMD production in waste rocks. Also, modeling can be used to evaluate control methods mainly aimed at reducing air convection (Lefebvre (4)).

The processes of water infiltration in a coarse heterogeneous porous media, mass transport in the unsaturated zone, and the leachate geochemical behavior (interaction with rocks and mineral precipitation) need to be studied further both in the lab and in the field. The lack of knowledge on these key processes is the main impediment on further development of AMD modeling and our capability to predict the behavior of waste rock dumps. Further field studies are also required as we need more site characterization with integrated monitoring and modeling programs. For those sites, we have to develop methods to better characterize air permeability and its anisotropy as it affects both gas convection and water infiltration. We also have to be able to characterize, in the field and lab, the changes in physical properties of waste rocks through time and especially the effect of mineral precipitation on permeability.

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