

ENHANCED OXYGEN TRANSPORT BY CONVECTION IN SULFIDIC ROCK PILES: THE INFLUENCE OF THE FORM OF THE SULFIDE OXIDATION RATE

Eugenia Y. Kuo and A. Ian M. Ritchie

Managing Mine Wastes Project, Australian Nuclear Science and Technology Organisation
Private Mail Bag 1
Menai, NSW 2234, Australia
Phone: +61 2 9717 3391, Fax: +61 2 9717 9129
e-mail: Eugenia.kuo@anto.gov.au

ABSTRACT

The overall sulfide sulfur oxidation rate in sulfidic rock piles is an important quantity when considering the management of waste rock piles, or the performance of biooxidation heaps and leach dumps. The transport of oxygen into the pile is a controlling factor in the overall oxidation rate, and diffusion is often the dominant mode of oxygen transport in field situations. However, when bulk convection arises in a rock pile, advection can enhance the transport of oxygen. A potential consequence of this enhanced oxygen transport is higher overall oxidation rates. One of the factors that determines the actual increase in overall oxidation rate is the form of the intrinsic sulfide oxidation rate. Using the 2-D code FIDHELM, we investigate the effect this form has on the enhancement of overall oxidation rates.

INTRODUCTION

The driving mechanism in the generation of polluted drainage from sulfidic mine wastes is the oxidation of sulfides, primarily iron sulfides (Brierley, 1978). While the main oxidant is ferric ion (Rossi, 1990), this is generated in most waste rock dumps and tailings dams, and in some leach dumps and biooxidation heaps, by oxidation of ferrous ion within the pile. The oxidant is gaseous oxygen in the pore space of the pile. Oxygen transport through the pore space then becomes a rate-limiting step in sulfide oxidation.

The dominant oxygen transport processes are diffusion and advection (Cathles and Schlitt, 1980). Advection can be driven: by pressure gradients induced, say, by wind over the pile; by temperature gradients induced by heat released in sulfide oxidation; and by density gradients induced by removal of oxygen in the oxidation of sulfides. Advection caused by temperature and density gradients is often called convection.

While it was shown that the pollutant load from the waste rock dumps at the Rum Jungle mine site could be accounted for by diffusive transport alone (Ritchie, 1977), it was later shown that some oxygen profiles measured in these waste rock dumps could only be explained if convection was contributing to the gas transport process (Harries and Ritchie, 1985). Since that time a number of models have been developed which take into account the physical transport processes involved in sulfide oxidation in waste rock dumps and heap leach piles. Generally simulations performed using these models show, that in a pile with a large ratio of width to height, diffusion dominates gas transport in the central regions of the pile, with convection occurring at the edges of the pile (Pantelis and Ritchie, 1991; Pantelis and Ritchie, 1992; Gelinis et al., 1994; Guo and Parizek, 1994; Casas et al., 1995). An example is shown in Figure 1. Typically, this convection causes elevated temperatures within the pile near the edges. Such elevated temperatures were reported by Harries and Ritchie in the waste rock dumps at the Rum Jungle mine site (Harries and Ritchie,

1981). Gelinas et al. (1994) have also reported elevated temperatures ascribed to convection in a waste rock dump at the Mine Doyon site.

In another study (Kuo and Ritchie, 1999), we showed that convection can occur in the central regions of an oxidising sulfidic rock pile, and called this type of convection, bulk convection, in contrast to convection confined to the edges of a pile. We also demonstrated that when bulk convection arises, it can significantly increase the overall oxidation rate of a rock pile relative to systems where diffusion is the dominant mode of oxygen transport. This should be of concern to the management of sulfidic waste rock dumps, and of interest to the performance of leach dumps and biooxidation heaps. The reason behind the increase in overall oxidation rates is enhanced oxygen transport via advection arising from convection throughout the rock pile.

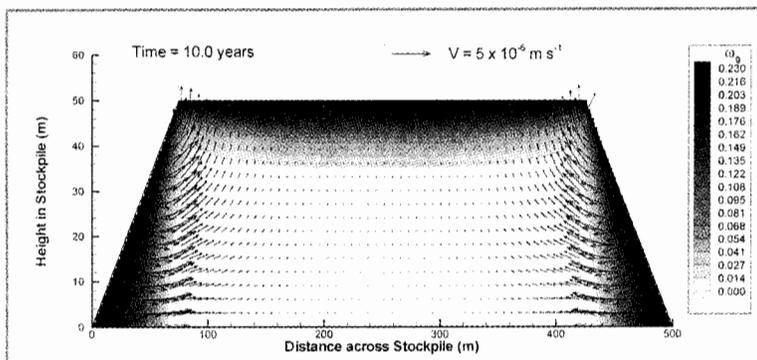


Figure 1. Oxygen mass fraction (ω_o) and gas velocity vectors (V) in an oxidising stockpile after 10 years. $S_0 = 1 \times 10^{-8} \text{ kg(O}_2\text{)} \text{ m}^{-3} \text{ s}^{-1}$, $K_a = 1 \times 10^{-10} \text{ m}^2$, $V_{\text{wind}} = 0 \text{ km/hr}$, $\rho_s = 1600 \text{ kg m}^{-3}$, $\omega_{s0} = 0.01$, $T_{\text{amb}} = 10 \text{ deg C}$; $R_p = 22$, $l = 3.1$. (Variables are defined in Table 1 and later).
Monod form of the IOR.

The simulations and analyses in Kuo and Ritchie (1999) were based on the monod form for the intrinsic oxidation rate (IOR) of sulfur. As the details of the form of the IOR are not usually well known, the sensitivity of the phenomena to this aspect of the modelling is investigated. Of particular interest is the impact upon the increase in overall oxidation rates.

MODEL OF SULFIDIC ROCK PILES

The system is modelled in two dimensions, with the rock pile modelled as a trapezoid of base width W , height L , and sloping sides angled at about 34 degrees from the base. The numerical code used to simulate the system is the 2-D finite-difference code FIDHELM (Pantelis, 1993). Processes essentially describing mass and energy conservation, incorporating transport, generation and consumption, form the backbone of the model as a set of coupled partial differential equations. The processes included are gas transport and consumption (pressure equation), gaseous oxygen transport and consumption, heat transport and

generation, and the oxidation of sulfide sulfur. Water transport is included in the form of a constant infiltration. Other models of the physical processes involved are Darcy's Law for the movement of the gas in the porous waste rock material, and the ideal gas law for the equation of state of the gas. The oxidation rate of sulfur is modelled using the monod and linear forms (Ritchie, 1994).

The boundary conditions reflect those of a dump, the base of which is sitting on an impermeable ground surface, while the other surfaces are exposed to the atmosphere. Gas and oxygen fluxes are set to zero at the base boundary. At the surfaces exposed to the atmosphere, the oxygen concentration is fixed at that of the ambient air, whilst the gas pressure is either fixed at atmospheric pressure, or is the pressure field resulting from wind blowing at a constant rate. The temperature is constant at all boundaries, and is that of the ambient air.

A number of physical properties of the waste rock dump were varied in the simulations. They are listed in Table 1, together with the values used in the simulations. The values reflect the range met in typical field situations. Also listed in Table 1, are the values used for the oxygen diffusion coefficient, and the monod parameters used in the monod form of the IOR.

In the simulations, the initial conditions are constant temperature and oxygen concentration – equal to that of the ambient air – throughout the dump, with the pressure set at that appropriate for the wind condition at the exposed surfaces.

Parameter	Symbol	Values
Max. Intrinsic oxidation rate	S_0	5×10^{-10} , 5×10^{-9} , 1×10^{-8} , 5×10^{-8} , $1 \times 10^{-7} \text{ kg (O}_2\text{)} \text{ m}^{-3} \text{ s}^{-1}$
Gas permeability	K_g	1×10^{-12} , 1×10^{-10} , 1×10^{-9} , $1 \times 10^{-8} \text{ m}^2$
Pile height	L	20, 30, 50, 70 m
Pile width	W	500, 700 m
Wind speed at a distance	V_{wind}	0, 30 km/hr
Bulk density	ρ_s	1600, 2000 kg m^{-3}
Initial sulfur mass fraction	Ω_{s0}^s	0.001, 0.01
Ambient temperature	T_{amb}	-20, 10 deg C
Oxygen diffusion coefficient in pile material	D_{ox}	$4.52 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$
Monod parameters	σ_1, σ_2	0.05, 0.05

Table 1: Physical parameters and values.

FORMS OF THE INTRINSIC OXIDATION RATE (IOR)

We investigate the impact the form of the IOR has on the phenomena of bulk convection using two distinct functional forms at opposing ends of the spectrum: the monod form and

the linear form. For the purposes here, the monod form of the IOR of sulfur is:

$$S = \epsilon^{-1} S_0 \cdot \frac{(1+\sigma_1)(\omega^g / \omega_0^g)}{(\omega^g / \omega_0^g) + \sigma_1} \cdot \frac{(1+\sigma_2)(\omega^s / \omega_0^s)}{(\omega^s / \omega_0^s) + \sigma_2}$$

and the linear form is:

$$S = \epsilon^{-1} S_0 \frac{\omega^g}{\omega_0^g} \frac{\omega^s}{\omega_0^s}$$

where ω^g is the oxygen mass fraction, ω_0^g is the ambient oxygen mass fraction, ω^s is the sulfur mass fraction, ω_0^s is the initial sulfur mass fraction, and $\epsilon=1.75$ is the ratio of the mass of oxygen (O_2) to that of sulfur (S) consumed in the oxidation reaction.

The spatially varying variable of interest where bulk convection is concerned is the oxygen mass fraction. For the linear form, the relative IOR is linearly dependent upon the oxygen mass fraction (hence the name 'linear'), whilst for the monod form, the relative IOR is nearly independent of the oxygen mass fraction when ω^g/ω_0^g is much greater than σ_1 .

BULK CONVECTION

Bulk convection can arise in an oxidising sulfidic rock pile due to an instability in the diffusion-dominated system. In a diffusion-dominated system, a vertical oxygen concentration gradient exists due to the consumption of oxygen in the oxidation processes, leading to a higher oxygen concentration on top. Consequently a vertical density gradient - with a higher density on top - exists, which has the potential to give rise to buoyancy-driven instabilities leading to convection (Kuo and Ritchie, 1999).

Figure 2 shows the oxygen mass fraction and the gas velocities at 8 years for one of the simulations in which bulk convection occurs. This particular figure shows a steady convection state; the convection states observed in the simulations take many forms, some of which are dynamical, with gas trajectories varying in space and time.

The phenomenon of bulk convection in oxidising sulfidic rock piles can be captured by a 2-parameter phase diagram (Kuo and Ritchie, 1999). Figure 3(a) shows the phase diagram for the monod form: there are two regions - one in which simulations show bulk convection occurring, and one in which simulations do not. The two regions are separated by an approxima-

te theoretically calculated curve of where the transition is expected to occur. There is good agreement.

The two dimensionless parameters used to describe the systems are: the Rayleigh number, defined to be:

$$R_\beta = \frac{\rho_{g0} \beta g L K_g}{D_{ox} \mu_g}$$

and a scaled rock pile height, defined as the pile height divided by the oxygen diffusion length:

$$l = \frac{L}{\sqrt{\frac{2C_0 D_{ox}}{S_0}}}$$

ρ_{g0} is the density of ambient air, β (=0.029 under the ideal gas law) is a dimensionless coefficient measuring a relative change in gas density due to changes in oxygen concentration under constant pressure conditions, μ_g is the gas dynamic viscosity, g is the gravitational acceleration, and C_0 is the oxygen concentration in the ambient air. The curves in Figures 3 (a) and (b), show the critical Rayleigh number, $R_{\beta c}$, as a function of l , calculated for the monod form. For a given l , if R_β for the system is greater than $R_{\beta c}(l)$, then bulk convection would be expected in a sufficiently wide dump, at least one governed by the monod form of the IOR.

OVERALL OR GLOBAL SULFUR OXIDATION RATES

In order to quantify, and hence compare, the impact of bulk convection on overall or global oxidation rates, we first define certain parameters. We define $G_A(S)$ to be the global (sulfur) oxidation rate (GOR(S)) of the rock pile divided by the horizontal area of the pile. $G_A(S)$ has units of $kg(S) m^{-2} s^{-1}$. It is the (sulfur) oxidation rate per unit area of the pile. The overall or global oxidation rate, one of the quantities of interest in the design of sulfidic rock piles, is thus

$$GOR = G_A(L) \cdot A$$

noting that G_A depends on L , the height of the pile, in a *nonlinear* fashion, and A is the horizontal area of the pile.

A second parameter of interest is Γ ; Γ is defined to be $G_A(S)$ divided by $G_A(S)_{diff}$, where $G_A(S)_{diff}$ is the value of $G_A(S)$ expected at early times for a one-dimensional diffusion-dominated system of the same height. The normalising factors, $G_A(S)_{diff}$, are different for the monod and linear forms of the IOR. For the monod form, it is (Gibson et al., 1994):

$$G_A(S)_{mon,diff} \approx \begin{cases} \epsilon^{-1} S_0 \frac{L}{l} & , \text{ if } l \geq 1 \\ \epsilon^{-1} S_0 L & , \text{ if } l < 1 \end{cases}$$

and for the linear form, it is

$$G_A(S)_{lin,diff} \approx \epsilon^{-1} \frac{S_0 L}{\sqrt{2l}} \tanh(\sqrt{2l})$$

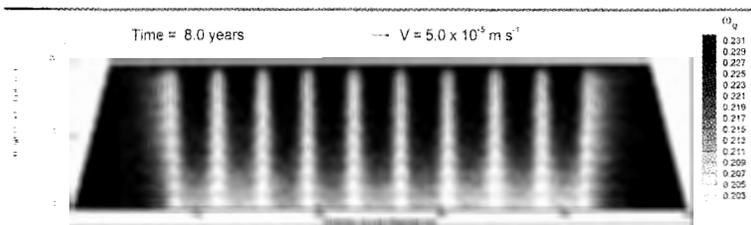


Figure 2. Steady bulk convection state: oxygen mass fraction (ω_g) and gas velocity vectors (V) in an oxidising stockpile after 8 years. $S_0 = 5 \times 10^{-9} kg(O_2) m^{-3} s^{-1}$, $K_g = 1 \times 10^{-8} m^2 s^{-1}$, $\rho_s = 0 km/hr$, $\rho_s = 1600 kg m^{-3}$, $\omega_{s0} = 0.01$, $T_{amb} = 10 deg C$; $R_\beta = 8.9 \times 10^2$, $l = 0.88$. Monod form of the IOR.

It was found in Kuo and Ritchie (1999) that Γ could increase significantly when bulk convection occurred, demonstrating the impact bulk convection can have on oxygen transport through advection and consequently overall oxidation rates.

SIMULATIONS AND RESULTS

To compare the impact the form of the IOR has on global oxidation rates, in particular when bulk convection occurs, a series of parallel simulations were run. That is, for a given system with identifying parameters S_0 , K_g , L , W , etc, a pair of simulations were run, the only difference between the two being the form of the IOR – either monod or linear. Both $G_A(S)$ and Γ were then evaluated for each simulation at 2 years. At that time transients have in general decayed away, and in most cases only a small fraction of the sulfide has been oxidised.

Figures 3 (a) and (b) show the phase diagrams for the monod and linear forms of the IOR respectively. The value of Γ (at 2 years) for each simulation is shown through the size (diameter) of the circles. It can be seen that there is little qualitative difference between the two phase diagrams, even though on average, Γ appears to be slightly larger for the linear cases. Furthermore, even though the theoretical critical Rayleigh number curve was calculated for the monod form, the Rayleigh numbers for the transition to bulk convection for the linear form are clearly very close to those for the monod form.

More quantitative comparisons can be done by comparing Γ and $G_A(S)$ (evaluated at 2 years) for each monod and linear pair of simulations. These are shown in Figures 4 and 5 respectively. Though there is scatter, it is found from Figure 4 that the relationship between Γ_{lin} and Γ_{mon} for the pairs where bulk convection occurred is approximately

$$\Gamma_{lin} = 1.1 \cdot \Gamma_{mon} + 0.1,$$

and from Figure 5 that the relationship between $G_A(S)_{lin}$ and $G_A(S)_{mon}$ does not change significantly when bulk convection arises.

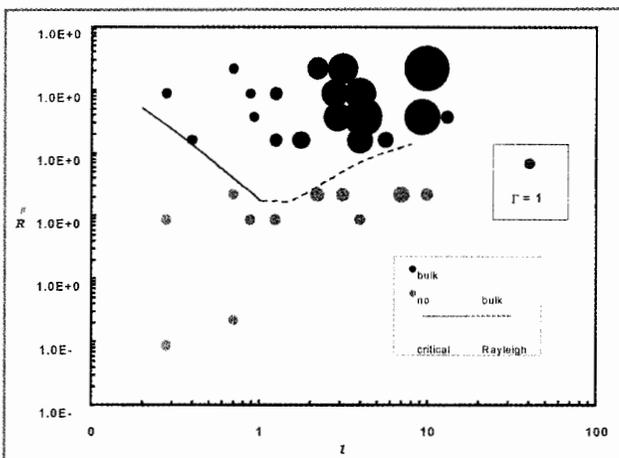


Figure 3 (a). Phase diagram and Γ_{mon} (at 2 years) for the monod form of the intrinsic oxidation rate. The diameter of the circles are proportional to Γ . The critical Rayleigh number curve comes from an approximate theoretical calculation, with the dashed portion being more uncertain.

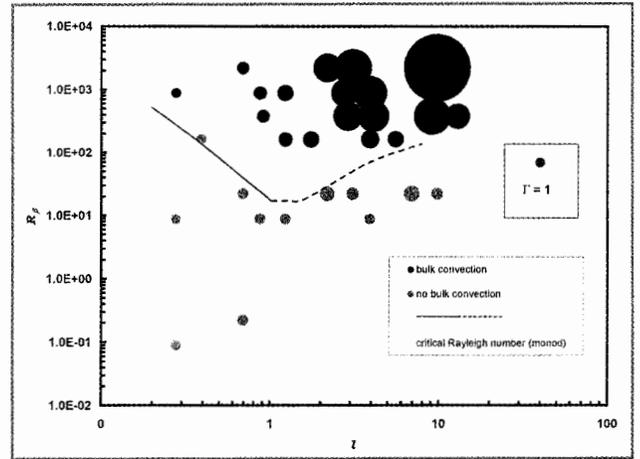


Figure 3 (b). Phase diagram and Γ_{lin} (at 2 years) for the linear form of the intrinsic oxidation rate. The diameter of the circles are proportional to Γ . The critical Rayleigh number curve comes from an approximate theoretical calculation based on the monod form, with the dashed portion being more uncertain.

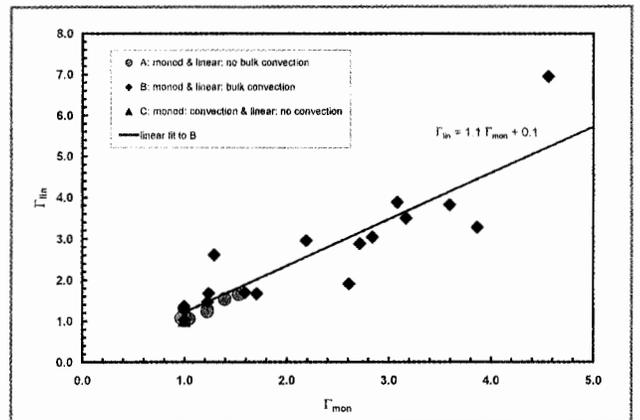


Figure 4. Γ_{lin} vs Γ_{mon} (evaluated at 2 years) for pairs of corresponding simulations.

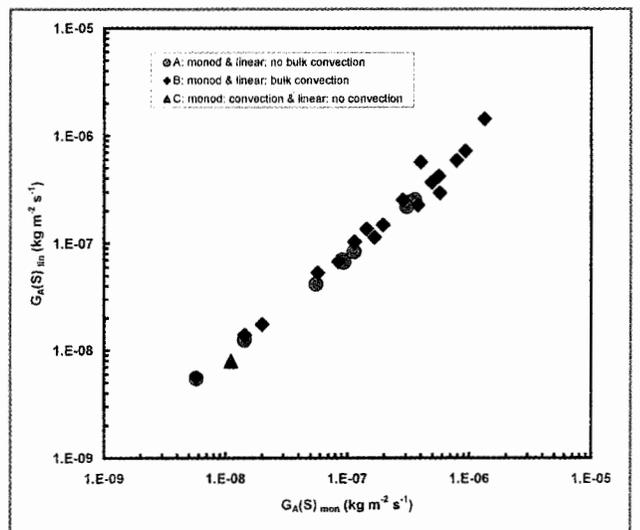


Figure 5. $G_A(S)_{lin}$ vs $G_A(S)_{mon}$ (evaluated at 2 years) for pairs of corresponding simulations.

It is expected that the enhanced oxygen transport when bulk convection occurs has little effect on the global oxidation rate when I is less than unity for the monod form, and that this would be less so for other forms of the IOR (Kuo and Ritchie, 1999). This is seen in the phase diagrams. However, as expected, the increase in global oxidation rates is more pronounced when I is greater than unity, as convection can then allow significant quantities of oxygen to reach much more of the pile relative to a diffusion-dominated system.

The somewhat larger than unity values for Γ for some systems in which bulk convection did not occur can be explained by the fact that all the simulated systems were of finite, not infinite width, with sloping sides. Convection confined to the sides of the pile can occur (without bulk convection occurring), contributing to an increase in the global oxidation rate of the pile, e.g., see Figure 1. It can be seen though, that bulk convection has the potential to increase the global oxidation rate in a much more significant way.

Figures 3, 4 and 5 thus indicate that the form of the IOR does not have a significant qualitative influence on either the occurrence of bulk convection or the consequent potential increase in global oxidation rates, at least at early times in the evolution of an oxidising sulfidic rock pile system. This implies that exact detailed knowledge of the form of the IOR is not necessary in order to estimate the effects of bulk convection on overall oxidation rates.

Figures 6 (a) and (b) show the time evolutions of Γ_{lin} and Γ_{mon} , and $G_A(S)_{lin}$ and $G_A(S)_{mon}$, over 10 years for a particular pair of simulations where bulk convection arose. We see for this particular case, that bulk convection gives rise to a substantial increase in the global oxidation rate at early times over that expected for a diffusion-dominated system. The global oxidation rate for the linear form diminishes more rapidly with time; this can be attributed to the rapid decrease in the sulfide reactant remaining in the pile, on which the oxidation rate of the linear form is linearly dependent, and on which the oxidation rate of the monod form is not.

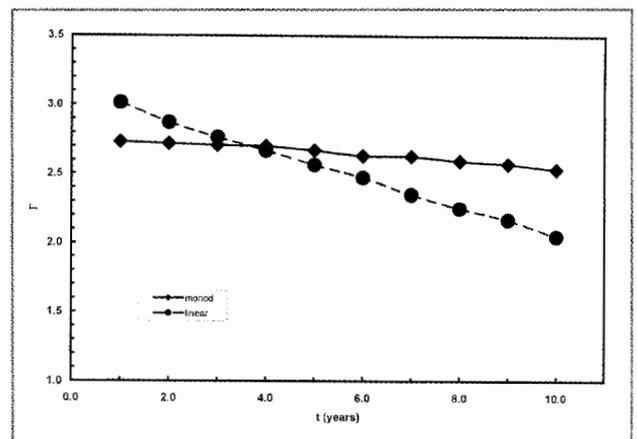
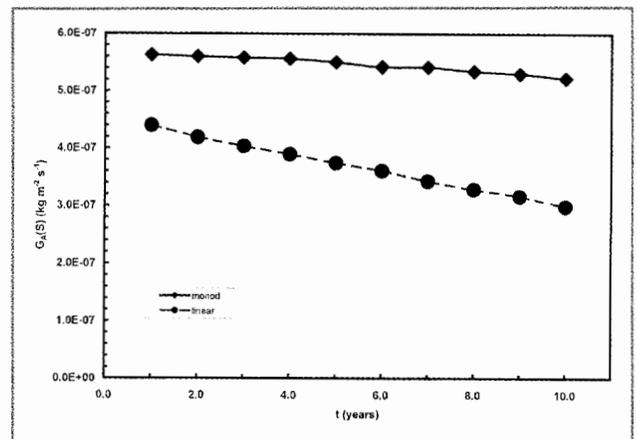
CONCLUSIONS

We have compared the impact the form of the intrinsic oxidation rate has on global oxidation rates in oxidising sulfidic rock piles when bulk convection arises. This was done by simulating systems in pairs, with the only difference being the form of the intrinsic oxidation rate – either monod or linear. Two parameters were then compared: Γ , the ratio of the global oxidation rate to that expected at early times for a diffusion-dominated system (interpretable as the gain in global oxidation rate due to bulk convection arising), and $G_A(S)$, the global oxidation rate per unit area of the rock pile.

It was found for both the linear and monod forms that bulk convection can lead to a significant increase in the global oxidation rate of an oxidising sulfidic rock pile. This is reflected

in a value of Γ substantially larger than 1. Although Γ (at 2 years) is on average slightly larger for the linear form when bulk convection occurred, it is not a significant difference. Furthermore, the relationship between $G_A(S)_{lin}$ and $G_A(S)_{mon}$ does not appear to change substantially when bulk convection sets in.

The form of the intrinsic oxidation rate would thus seem not to be an important factor in determining, at early times, the gain in global oxidation rates with the onset of bulk convection. As far as the management of sulfidic waste rock dumps, or the performance of leach dumps and biooxidation heaps is concerned, the more important factor would appear to be whether bulk convection will occur in the oxidising sulfidic rock pile.



Figures 6(a) and (b). The time evolution of Γ and $G_A(S)$ respectively, for the first 10 years for a monod-linear pair of simulations. The other parameters of the systems are $S_0=5 \times 10^{-8} \text{ kg (O}_2\text{) m}^{-3} \text{ s}^{-1}$, $Kg=1 \times 10^{-6} \text{ m}^2$, $L=20 \text{ m}$, $W=500 \text{ m}$, $V_{wind}=0 \text{ km/hr}$, $\rho_s=1600 \text{ kg m}^{-3}$, $\omega_{s0}=0.01$, $T_{amb}=10 \text{ deg C}$; $R_p=8.9 \times 10^2$, $l=2.8$.

ACKNOWLEDGMENTS

We are gratefully indebted to Mr Paul Miskelly for carrying out the many FIDHELM simulations necessary for this work. He also kindly produced Figures 1 and 2 using output from the simulations. We would also like to thank Mr Michel Noël for advice, and Mr Ot Sisoutham for help in converting figures to TIFF format.

REFERENCES

- Brierley, C.L., 1978. Bacterial Leaching, *CRC Critical Rev. Microbiol.* 6, 207-262.
- Casas, J.M., J. Martínez, L. Moreno and T. Vargas, 1995. Two-dimensional model of heat and gas transport and mineral oxidation in copper bioleaching dump, *Biohydrometallurgical Processing 1. Proc. International Biohydrometallurgy Symposium IBS-95*, eds. T. Vargas, C.A. Jerez, J.V. Wiertz and H. Toledo, Viña del Mar, Chile, Nov. 1995, University of Chile, 447-457.
- Cathles, L. and W.J. Schlitt, 1980. A model of the dump leaching process that incorporates oxygen balance, heat balance, and two-dimensional air convection. *Proc. Symp. Leaching and Recovering Copper from As-Mined Materials*, ed. W.J. Schlitt, Las Vegas, USA, Feb. 1980, Society of Mining Engineers of A.I.M.E., 9-27.
- Gelinas, P., R. Lefebvre, M. Choquette, D. Isabel, J. Locat and R. Guay, 1994. Monitoring and modelling of acid mine drainage from waste rock dumps, La Mine Doyon case study. *MEND Report 1.14.2a*.
- Gibson, D.K., G. Pantelis and A.I.M. Ritchie, 1994. The relevance of the intrinsic oxidation rate to the evolution of polluted drainage from a pyritic waste rock dump. *Proc. International Land Reclamation and Mine Drainage Conference and Third International Conference on the Abatement of Acidic Drainage*, Pittsburgh, USA, April 1994, United States Dept. of the Interior, Bureau of Mines Special Publ. SP 06B-94 2, 258-264.
- Guo, W. and R.P. Parizek, 1994. Field Research on Thermal Anomalies Indicating Acid Reactions in Mine Spoil. *Environmental Geochemistry of Sulfide Oxidation*, eds. C.N. Alpers and D.W. Blowes, American Chemical Society Symposium Series 550, 645-659.
- Harries, J.H. and A.I.M. Ritchie, 1981. The use of temperature profiles to estimate the pyritic oxidation rate in a waste rock dump from an openpit mine. *Water, Air and Soil Pollution* 15, 405-423.
- Harries, J.H. and A.I.M. Ritchie, 1985. Pore gas composition in waste rock dumps undergoing pyritic oxidation. *Soil Sci.* 140, 143-152.
- Kuo, E.Y. and A.I.M. Ritchie, 1999. The Impact of Convection on the Overall Oxidation Rate in Sulfidic Waste Rock Dumps. To appear in the *Proceedings of Sudbury '99, "Mining and the Environment II"*, Sudbury, Ontario, Canada, Sept. 1999.
- Pantelis, G., 1993. *FIDHELM: Description of Model and Users Guide*. Australian Nuclear Science and Technology Organisation report, ANSTO/M123.
- Pantelis, G. and A.I.M. Ritchie, 1991. Macroscopic transport mechanisms as a rate-limiting factor in dump leaching of pyritic ores. *Appl. Math. Modelling* 15, 136-143.
- Pantelis, G. and A.I.M. Ritchie, 1992. Rate-limiting factors in dump leaching of pyritic ores. *Appl. Math. Modelling* 16, 553-560.
- Ritchie, A.I.M., 1977. *Heap Leaching: A Gas Diffusion Rate Limited Model*. Australian Atomic Energy Commission report AAEC/E429.
- Ritchie, A.I.M., 1994. *Biooxidation Heaps and AMD from Waste Rock Dumps – The Importance of the Intrinsic Oxidation Rate*. The AusIMM Annual Conference, Darwin, Australia, August 1994, 473-477.