Simulation of Bioleaching Heat Effects for Enhancement of Copper Recovery from Sarcheshmeh Chalcopyrite

ALI REZA MAHMOUDIAN, S.K. SADRNEZHAAD, and ZAHRA MANAFI

A heat-transfer model was formulated to determine the distribution of temperature within a bioheap of chalcopyrite of Sarcheshmeh copper mine. Bioleaching employs mixed mesophilic and thermophilic microbes for Cu extraction. Thermophiles are better than mesophiles to dissolve $CuFeS_2$. The solution irrigation and aeration rates were taken into account as the main operational factors. The model was validated by comparing the temperature profiles of test columns with those of bioheap. The model was used to find the optimal ratio of irrigation to aeration. It was found that when the solution was fed at a flow rate of 5 kg/m^2 h and air was blown at a flow rate of 7.5 kg/m^2 h, the transition from a mesophilic to thermophilic state inside the heap was possible. In this situation, the maximum temperature rise inside the heap was about 332 K (59 °C) after 60 days.

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I. INTRODUCTION

As worldwide reserves of high grade ore are rapidly diminishing because of heavy exploitation, traditional techniques of chemical processing have become less economically attractive. Microorganisms bear, however, a clear advantage over the traditional techniques by offering clean and economically viable alternatives. Microorganisms such as bacteria and fungi convert metal compounds into water-soluble forms. They act, hence, as biocatalyst of the leaching processes. Bioleaching occurs naturally while harnessing metal recovery from low-grade ores.[1]

Among the most significant applications of microorganisms to extract precious metals is heap bioleaching. Heap bioleaching is a hydrometallurgical process which can take place over a period of months to years. It involves the application of acid solution and bacteria (either natural or inoculated). To keep the aerobic bacteria alive, air is injected (sparged) into the heap using pipes laid before heap stacking. The solution infiltrates the heap from the top, soaking the ore, and leaching the metals into solution. Finally, the solutions are collected at the base of the heap for subsequent electrowinning of the metal.[2]

The need for efficient heap bioleaching operations has led to scientific research and development of mathematical models. A number of heap bioleaching models developed before have been reviewed.[3]

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Among the significant processes that occur in a bioheap are the heat generation and conservation. An analysis of heat conservation for copper sulfide heap leaching has been accomplished with the aid of a computer model.[4] Such analysis is carried out for several reasons. First, the bacteria which catalyze the oxidation reactions are sensitive to temperature. Second, many oxidation reactions influencing the copper recovery are strongly temperature dependent.[3,4]

Chalcopyrite is the most important source of copper in the world; however, its sources are increasingly declining in grade.[5] It has been observed that chalcopyrite passivation is not observed in thermophile leaching.[6,7] Therefore, the aim of the current study is to find the conditions by simulation under which thermophilic leaching is established within the heap of Sarcheshmeh copper mine.

II. MATERIALS AND METHODS

Heap bioleaching involves loading of material onto an impervious base, irrigating the top of the heap with a suitable lixiviate, aeration from bottom, and treatment of the pregnant solution draining from the heap for extraction of the dissolved metal values. The leaching solution flows downward under influence of gravity, and air is supplied upward from the pipe lines that are located in the bottom of the heap. In some cases, the leaching solution is recirculated to reach a proper concentration of leached metals. However, in the current study, we used fresh solutions.

The heat loss from the lateral walls because of advection can be neglected in comparison with the heat transfer within the heap in the direction of the leaching solution and flowing air. The leaching solution and air are flowing in opposite directions. The heat analysis will take place, therefore, in the axial direction.[4]

The model formulation is based on the assumptions similar to those of Dixon[4] and Vilcaez *et al.*,[8] but the high night-time heat losses were not considered. A heat balance equation has been proposed by Dixon,[4] which includes the effect of heat losses due to water vaporization, the air, and vapor flows, and the heat generation within the heap. Some types of heat flux, such as convection and evaporation, are taken into consideration.

The partial differential equations were semi-discretized using the method of lines, by applying second-order finite-difference discretization in space. The enthalpy balance is a second-order, nonlinear partial differential equation with mixed nonlinear boundary conditions. This equation is solved numerically using a fully implicit finite-difference formulation, including central differences for the accumulation term. The calculation procedure was coded in Fortran. The time step used was 0.1 hour, and calculations showed that the reduction of time steps did not affect on the results obtained.

A. *Apparatus and Procedure*

Chemical and mineralogical compositions of the mineral are reported in Table I. The column–leach experiments were conducted in columns having 30-cm inner diameter and 4-m height. Each column was thermally insulated with merely its top open. There were several thermocouples positioned at a distance of 40 cm from each other and were arranged from the wall of column to the center. The columns were charged with sulfide ore from Sarcheshmeh mine. The sizes of minerals were between 0.5 and 2 cm in diameter ($0.5 \le \phi < 2$ cm).

The proposed plant consists of two 6-m-high heaps of approximately 15,000 tons each, stacked on a prepared base on which the aeration pipes are overlaid. Drainage pipes were located at the bottom of the heaps, beside the aeration pipes. The site consists of PLS ponds, raffinate ponds, inoculum ponds, and auxiliary ponds. The solution drainage gravitates to the PLS ponds and from there to the main PLS pond of hydrometallurgy unit. Return raffinate is pumped to the raffinate ponds from where the heap is irrigated. Inoculum solution ponds are situated above the auxiliary ponds, for the purpose of storage of prepared inoculum. The PLS and raffinate ponds overflow to the auxiliary ponds, and provision is made for solution transfer between auxiliary and inoculum ponds. There are several thermocouples positioned at a space of 40-cm from each other at the center of heaps. The heaps are irrigated with a series of drippers and sprinklers. Figure 1 illustrates an implementation of a heap bioleaching process.

Three experiments were carried out by downward percolation of dilute sulfuric acid in the test columns (pH = 1.3 to 2.0) at different G_l/G_a ratios: 1/6, 1/3, and 2/3. The initial temperature was 293 K (20 °C), and fresh air was sparged from the bottom of the columns. Each column was thermally insulated by means of some calcium silicate insulation boards and with pipe

Table I. Chemical and Mineralogical Compositions of the Mineral Used in this Research

Element/Mineral	Cu (total)	Cu*	Fe	FeS_2	CuS	Cu_2S	$CuFeS_2$	Fe_2O_3	Fe_3O_4
Weight (pct)	0.37	0.02	3.92	8.12	0.06	0.14	0.58	0.03	0.06

*Acid soluble copper.

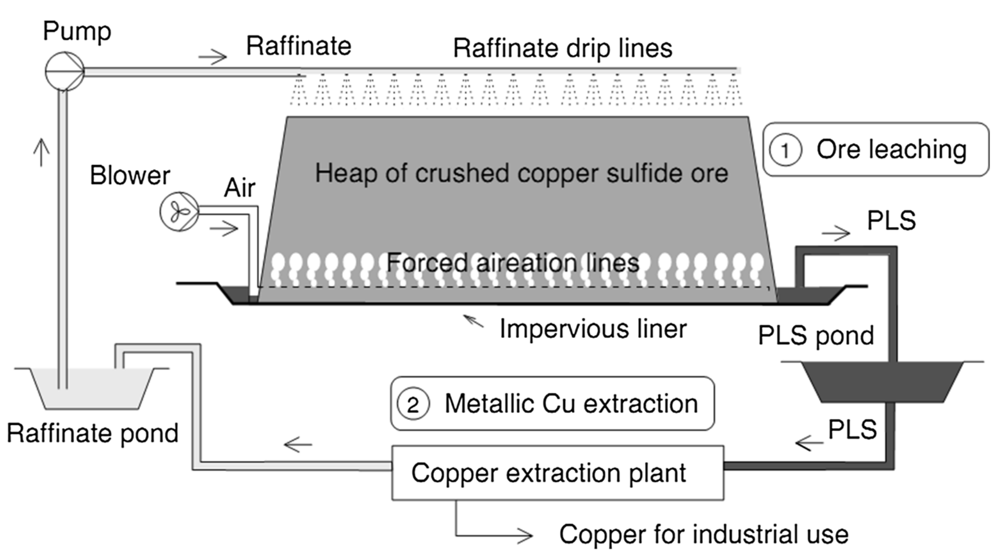

Fig. 1—Simplified copper heap leaching process.

insulation features. Two experiments were carried out in heaps at two different G_l/G_a ratios: 1/6 and 1/3. Leaching tests were carried out for a duration of up to 100 days.

At the beginning of the experiments, the columns were rinsed with approximately 25 L of sulfuric acid (2.5 g/L). Subsequently, a mixture of mesophilic and moderately thermophilic bacteria consisting of *Acidithiobacilluc Caldus*, *Leptospirillum Ferriphilum*, *Sulfubacillus, and Ferroplasma* were fed to the top of the columns at a rate of approximately 2 L/m^2 h until the bacteria appearing at the bottom of each column reached 10^7 cells/mL. During the experiments, the population of the bacteria in the collected solution at the bottom of columns was counted. More bacteria were added from the top, whenever the population of bacteria would fall to less than 10^7 cells/mL. The population of free bacteria in solution was determined by direct microscopic counting.

Solution was fed to the top of the column with a peristaltic pump at approximately 5 kg/m^2 h and collected at the base into a separate container. Air was blown at the base of the column at different rates. In column experiments, a fresh sulfuric acid solution at pH = 1.3 was used.

In the heap experiments, the pH of the feed was adjusted to 1.3 with the addition of concentrated sulfuric acid, and the collected solution was circulated as long as the dissolved copper and iron concentrations remained lower than 3 and 4 g/L, respectively. If the levels were greater than the expected extents, 50 pct of leach solution was replaced by fresh solution. The pH was then adjusted to 1.3. Drip emitters were used for heap irrigation, to keep the surface heat-transfer coefficient constant. The temperature was monitored and recorded for a few days.

Table II contains the parameters that were used in the modeling.

For simulation, the rate of heat generation S was assumed to be uniform and constant (equal to the initial rate S_0). This assumption was similar to Dixon's assumption.[4] The surface heat-transfer coefficient, h, was also assumed to be constant. For modeling the effect of ratio of the solution flow rate to the aeration flow rate, the flow rate of the solution was kept constant (5 kg/m^2 h), and the experiments were carried out at different G_l/G_a ratios. The standard conditions were assumed in all the cases. The column experiments showed that within 80 days, steady-state temperature profile was established, and no significant changes in the temperature profiles occurred.

The simulation was evaluated by applying the main operational variables and the ratio of solution to the aeration mass flow rates (G_l/G_a). The irrigation concentrations of mesophiles and thermophiles were kept at the

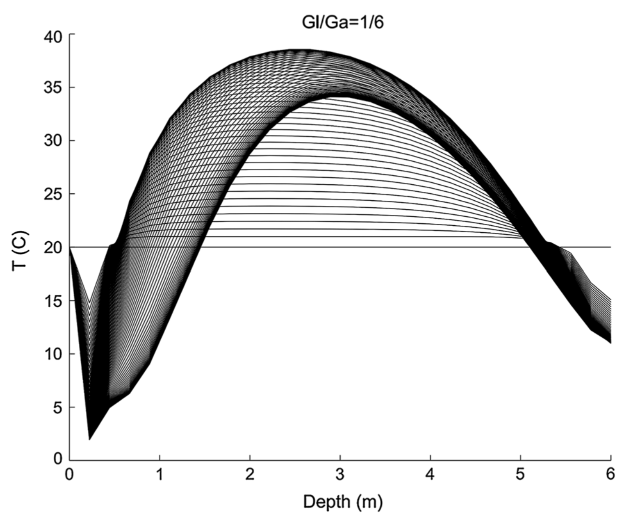

Fig. 2—Simulated time histories for temperature profiles carried out up to 80 days at G_l/G_a = 1/6. Each curve represents 1 day of leaching.

Table II. Parameters for the Base Case

Parameter	Value	References
Height of the heap	L = 6 m	adjusted
Temperature of the leaching solution	T_l = 283 K (10 °C)	adjusted
Temperature of the air stream	T_a = 283 K (10 °C)	adjusted
Initial temperature in the heap	T_i = 293 K (20 °C)	adjusted
Temperature of the environment	T_{inf} = 293 K (20 °C)	assumed
Liquid fraction in the heap	θ = 0.2	[8]
Average thermal conductivity of the heap	k = 3600 J/m K h	[4]
Average density of the heap	ρ = 1700 kg/m^3	calculated
Average density of the sulfide minerals	ρ_{min} = 2700 kg/m^3	[8]
Air density	ρ_a = 1.208 kg/m^3	[8]
Average heat capacity of the heap	C_p = 1000 J/kg K	[4]
Average heat capacity of the leach solution	C_{pl} = 4184 J/kg K	[8]
Average heat capacity of the dry air	C_{pa} = 1000 J/kg K	[4]
Average heat capacity of the vapor phase	C_{pv} = 1840 J/kg K	[4]
Surface heat-transfer coefficient	h = 72000 J/m^2 h K	[4]
Latent heat of water vaporization	λ = 2258 × 10^3 J/kg	[4]
Air relative humidity	w_a = 30 pct	[4]
Solution irrigation rate	G_l = 5 kg/m^2 h	[4]
Aeration rate	G_a = variable	

maximum amount reachable (about 10^{11} cells/mL). Other parameters such as change of the size of particles and the accumulation of precipitates on the mineral surface were not taken into consideration. The mathematical model developed was also tested by means of the case study parameters given in the literature (Table II).

B. *Considerations*

One of the major heat sources during heap bioleaching is pyrite, and this is necessary for thermophilic operation. As it is seen from Table II, there is sufficient amount of pyrite within the heap. Chalcopyrite ores cannot be leached at low temperatures, and oxidation of some sulfides like pyrite increases the temperature; hence, using thermophilic microorganisms becomes possible.[9]

This modeling is done for thermal behavior of bioleaching, and in this case, the heat source, S, has been considered to be constant. However, in fact, during the leach process, the amount of S changes as the composition of solid phase changes. However, in this research, the amount of S is considered to be constant till the steady-state is established.

The other assumption in the current study is that the heat loss from column walls is negligible. For this purpose, the columns were thermally insolated, and no attempt was made to calculate the heat loss from the walls.

In Dixon's study, the radiative heat flux has been taken into account, which includes a term for the absorption of solar radiation, and for gray body radiative exchange with the sky; however, in the current study, this term is no longer valid, especially in laboratory column tests.

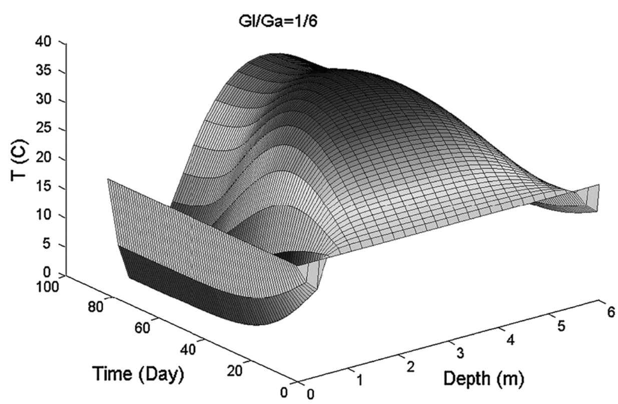

Fig. 3—Simulated 3D shape of temperature profile inside the heap at $G_l/G_a = 1/6$.

III. RESULTS AND DISCUSSION

The first experiment was done by choosing $G_l/G_a = 1/6$. The experimental results for the case of $G_l/G_a = 1/6$ (column and heap tests) showed that, after about 45 days, temperature rose to around 313 K (40 °C), and the maximum temperature changed the position and the amount–increasing the distance from the heap surface, and then falling to around 307 K (34 °C) nearer to the other exterior. However, this case showed a confusing behavior in practical experiments. This case was not used to calibrate the model. The experimental results are not described in this article completely, because too many data are available. Figure 2 shows a simulated 2D history of the temperature profile inside the heap for the $G_l/G_a = 1/6$, and Figure 3 shows a simulated 3D of the same. The results of days 35 and 75 are drawn in the Figures 4(a) and (b), respectively, to compare between the simulation and the experimental results. As is clearly seen, there is a close

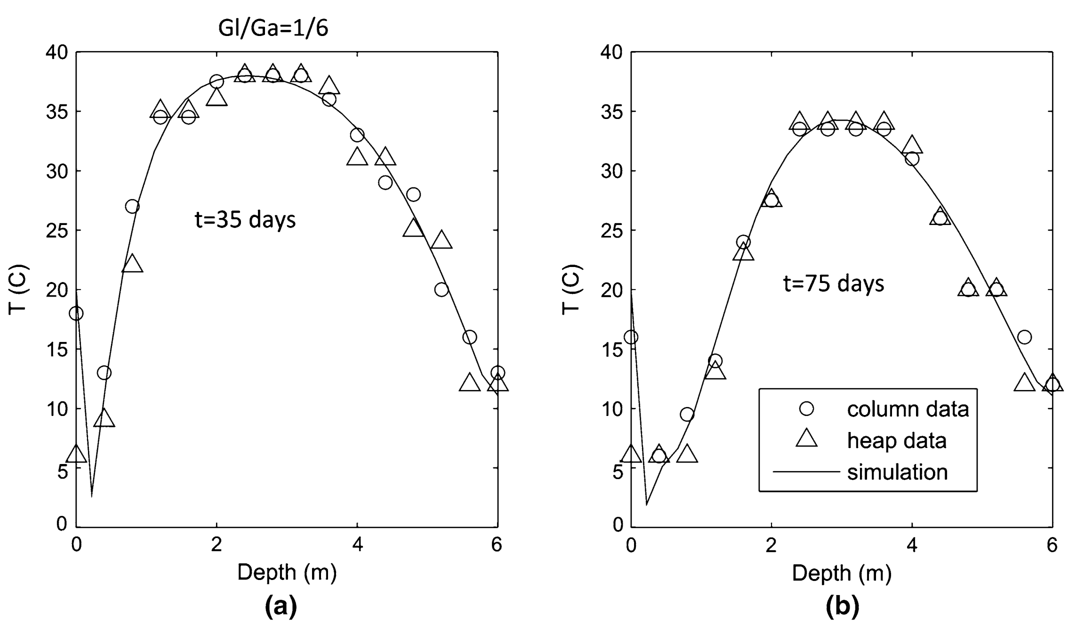

Fig. 4—Temperature profiles at $G_l/G_a = 1/6$ for (*a*) day 35 and (*b*) day 75. Each case consists of column test, heap, and simulation results.

correspondence between the simulation results and the heap and column tests results. Although this temperature was not suitable for thermophilic transition, it is not economic to establish this condition because of very high air flow rate and the need for powerful blowers.

Subsequent experiments were conducted by choosing $G_l/G_a = 1/3$. The results of column test were used to calibrate the model. Experimental data for days 35 and 75 are compared with simulation results. Result of simulation is shown in Figure 5, which shows the temperature rising to 316 K (43 °C), which first slightly decreases to 314 K (41 °C) and then remains constant. Figures 6(a) and (b) shows the results of column test, heap, and simulation at days 35 and 75, respectively. As can be seen, decreasing the aeration rate caused an increase in temperature, because the high rate of aeration in the pervious case had a cooling effect within the heap. Although the internal temperature has increased with the decrease in the aeration rate, it was, however, not enough for causing the transition from the mesophilic to the thermophilic state. The study on bacteria showed no thermophilic activity.

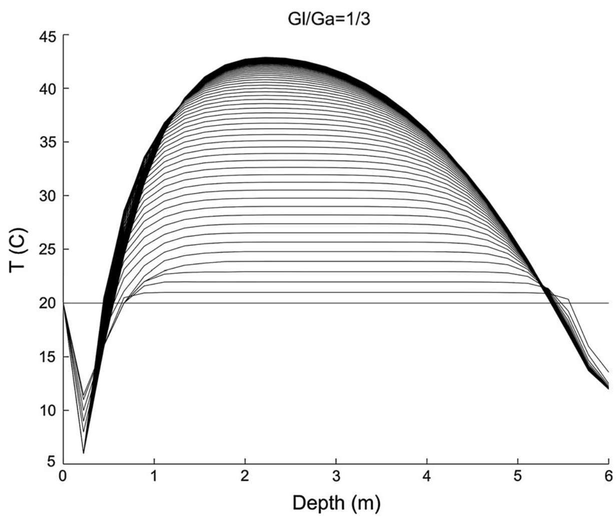

Fig. 5—Time histories for temperature profiles carried out up to 80 days at $G_l/G_a = 1/3$. Each curve represents 1 day of leaching.

The aim of the current study was to find a condition at which the thermophilic state would be established inside the heap. For this purpose, the temperature of heap should raise to at least 333 K (60 °C). When the heat-transfer model, according to Dixon's model,[4] was calibrated for the column results in the case $G_l/G_a = 1/3$, there was a close correspondence of these results with the pervious practical tests ($G_l/G_a = 1/6$), and the other cases were simulated to find the optimal condition.

Other simulations were conducted without real-time tests. The chosen amounts and ratios were approximately similar to those of Dixon's study.[4] The model simulation results for the cases: $G_l/G_a = 1/6$, 1/3, 1/2, 2/3, 3/4, and 1 are shown in Figures 7 and 8. It was seen that the decrease in the aeration, for G_l/G_a ratios from 1/6 to 2/3, causes a rise of temperature inside the heap. However, any further decrease in this ratio causes a drop in the temperature. Because, at the lower aeration rates, increasing the aeration rate should be as effective as the addition of fuel sources such as sulfide sulfur to increase the heat generation rate.[4] Hence, there is an optimal G_l/G_a ratio between 1/6 and 1. In the case of $G_l/G_a = 2/3$, the maximum temperature inside the heap will reach up to 332 K (59 °C). This temperature is sufficient to cause transition from a mesophilic to a thermophilic state. At lower and larger ratios of G_l/G_a, the maximum temperature is around 314 K (41 °C)

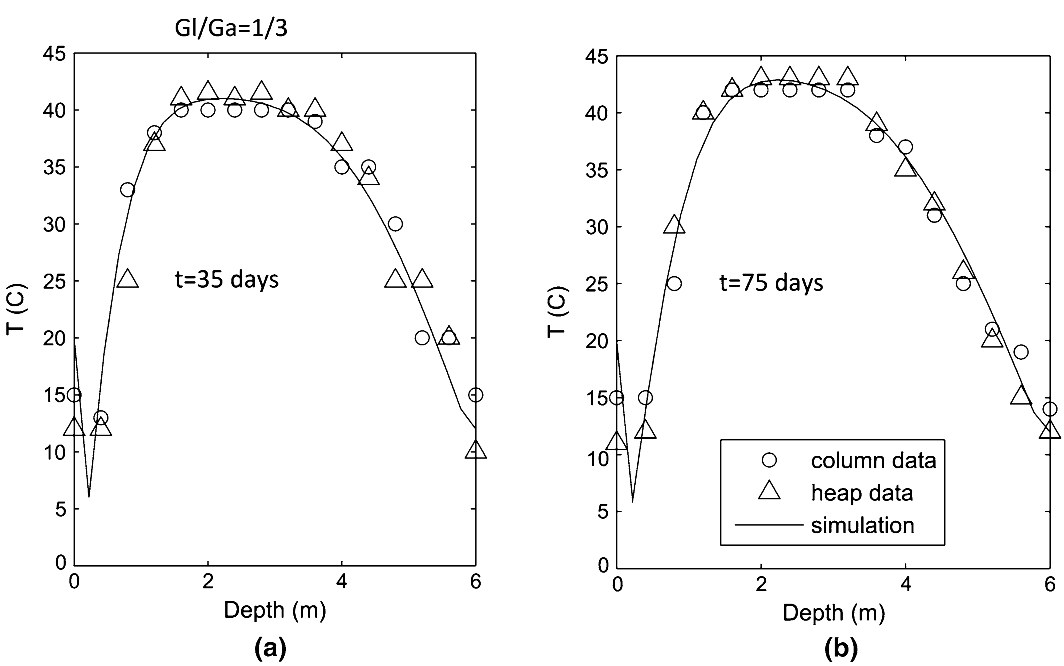

Fig. 6—Temperature profiles at $G_l/G_a = 1/3$ for (a) day 35 and (b) day 75. Each case consists of column test, heap, and simulation results.

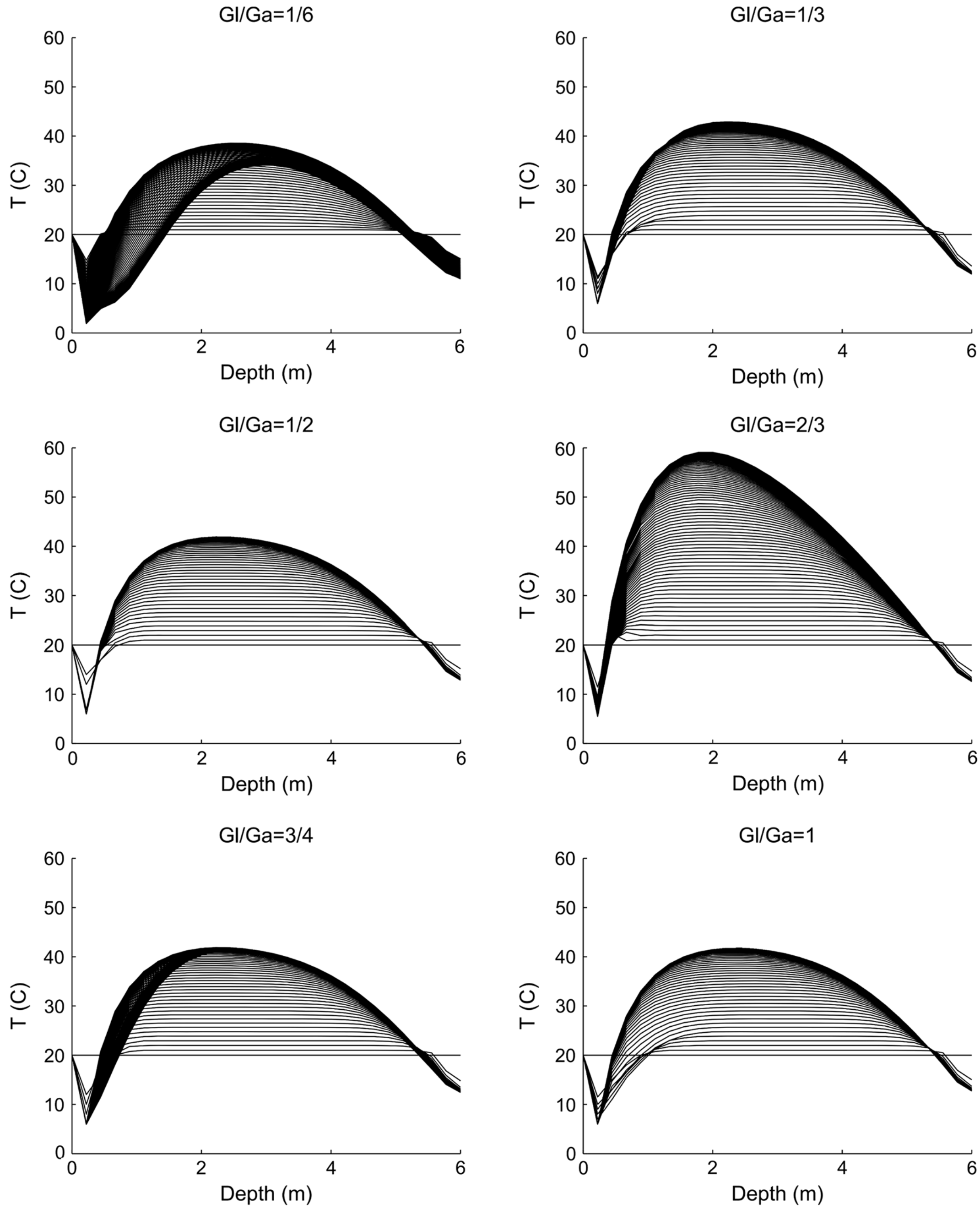

Fig. 7—Time histories for temperature profiles carried out up to 80 days at G_l/G_a = 1/6, 1/3, 1/2, 2/3, ¾, and 1. Each curve represents 1 day of leaching.

which is not high enough to cause the transition from mesophilic to thermophilic state.

The other column test was carried out at $G_l/G_a = 2/3$ to study the optimal conditions which were obtained from simulation. The results of a $G_l/G_a = 2/3$ ratio in the column were in good agreement with the simulation results, and the maximum temperature of 330 K (57 °C) was obtained after 60 days. Figures 9(a) and (b) shows the column test and simulation results for days 35 and 75, respectively.

The average internal temperature of heap is calculated from Eq. [1]:

$$T_{avr} = \frac{1}{L}\int_0^L T(z) \cdot \mathrm{d}z, \qquad [1]$$

where z represents the distance from the heap surface.

Figure 10 shows plots of the simulated average heap temperature *vs* time at six different aeration rates. A notable result from this figure is that there is an optimal

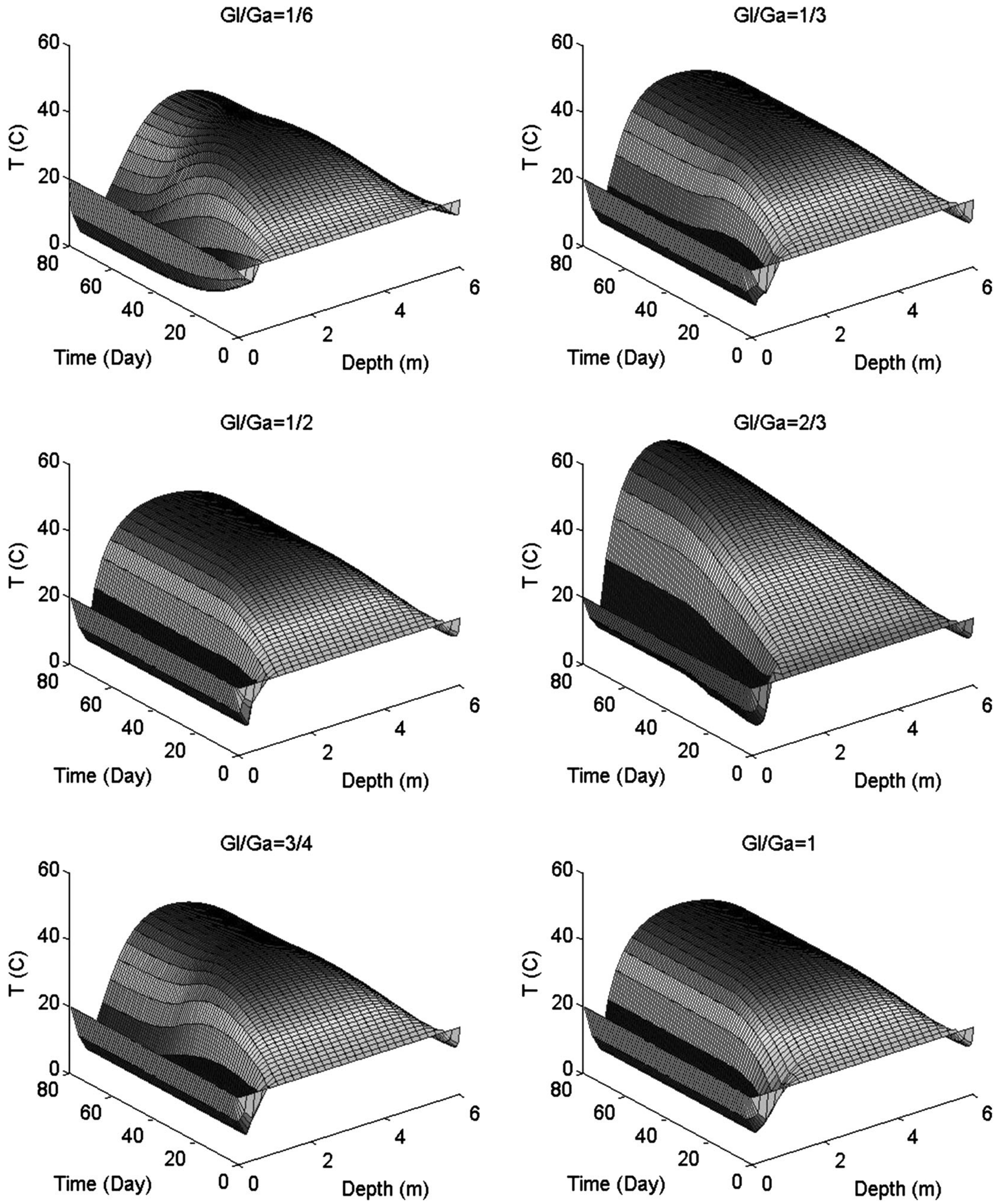

Fig. 8—3D shapes of temperature profiles inside the heap at $G_l/G_a = 1/6$, 1/3, 1/2, 2/3, 3/4, and 1.

amount for aeration rate. It was seen that at aeration rate of more than or less than 7.5 kg/m^2 h ($G_l/G_a = 2/3$), the average temperature of the heap would decrease. This figure also shows that the average temperatures at aeration rates of 10 kg/m^2 h ($G_l/G_a = 1/2$), 15 ($G_l/G_a = 1/3$), and 5 ($G_l/G_a = 1$) are the same, and the maximum temperature of heap is approximately 314 K to 316 K (41 °C to 43 °C). This temperature is not sufficient to activate the thermophiles bacteria.

In column experiments, controlling some parameters, like initial temperature of concentrate, solution, and air, was feasible. However, in actual heap tests, it was not possible to control them. The results of Dixon[4] showed that the initial temperatures of heap, solution, and air have no significant effects on average and maximum temperatures of the heap. Therefore, we decided to choose the aeration rate, as the main model parameter. Decreasing the initial temperatures will, however, increase the time for warming up the heap and has no effect on the amount and the position of maximum temperature inside the heap.

There was no investigation of mass balance in the current study, but investigations of bacteria within the columns and heaps showed that no thermophilic activity was observed except in the case of $G_l/G_a = 2/3$.

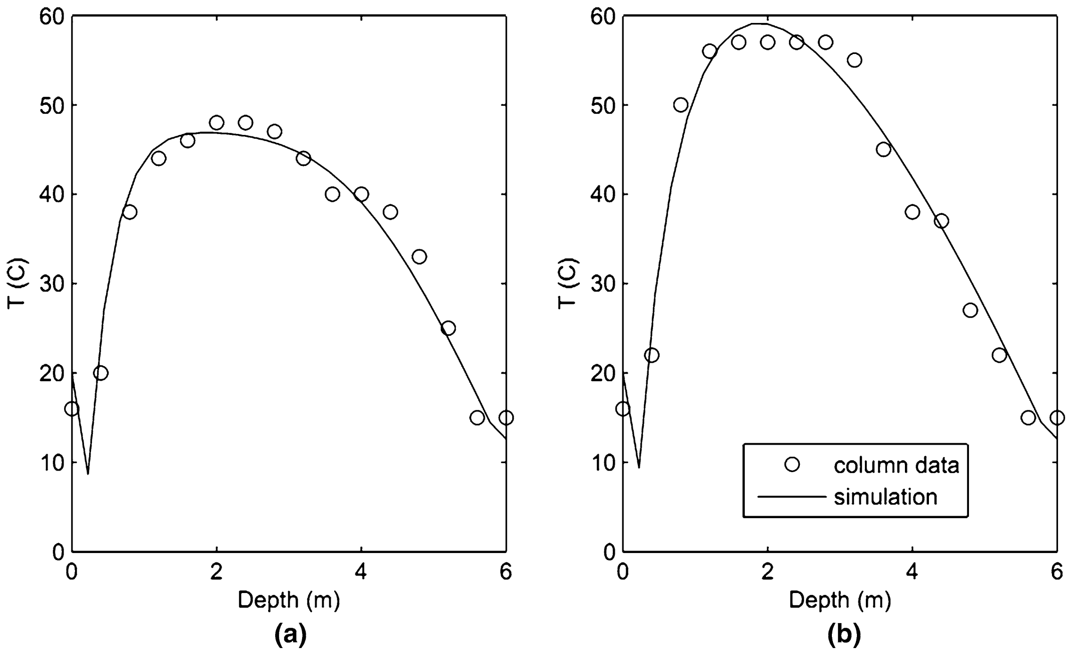

Fig. 9—Temperature profiles at $G_l/G_a = 2/3$ for (*a*) day 35 and (*b*) day 75. Each case consists of column test and simulation results.

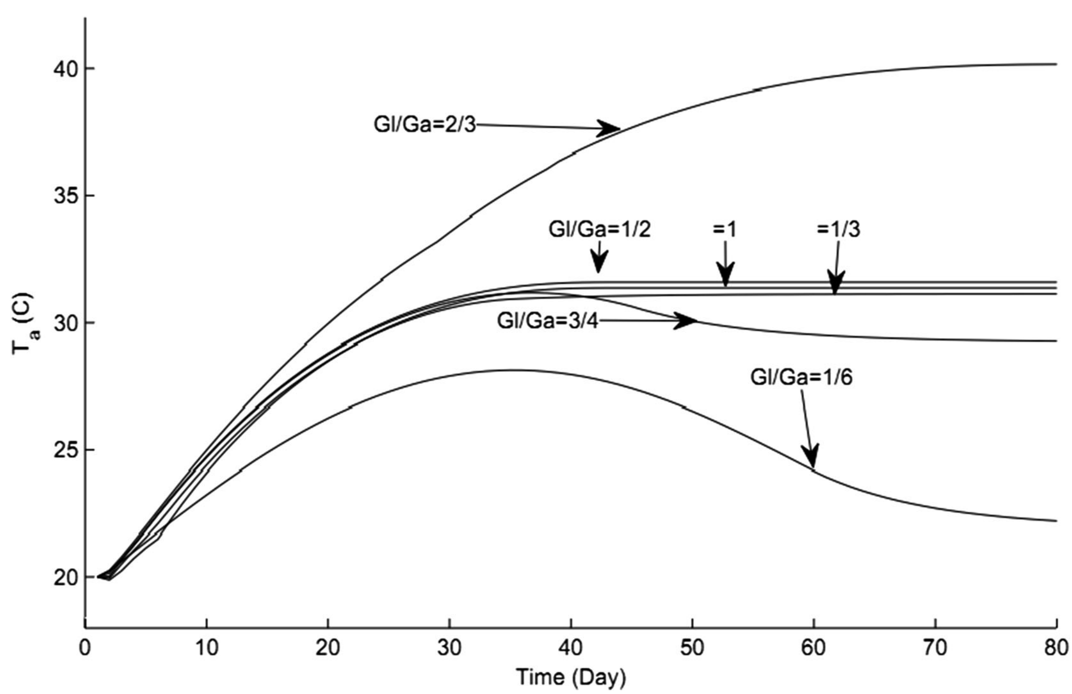

Fig. 10—Integrated average temperatures *vs* time at six different aeration rates.

IV. CONCLUSIONS

Thermal simulation of chalcopyrite leaching of Sarcheshmeh mine was accomplished. It was found from simulation results that, at an irrigation rate of 5 kg/m^2 h and an aeration rate of 7.5 kg/m^2 h, it is possible to realize transition from mesophilic to thermophilic state. Increase of the inner temperature of heap up to around 333 K (60 °C) is sufficient to achieve this transition.

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REFERENCES

1. N. Ogbonna, J. Petersen, and D.G. Dixon: *Proc. 35th Annu. Meet. Hydrometall.*, 2005, pp. 225–40.

2. M.J. Leahy, M.R. Davidson, and M.P. Schwarz: *Miner. Eng.*, 2005, vol. 18, pp. 1239–52.
3. D.G. Dixon, and J. Petersen: in *Copper 2003*: *Hydrometallurgy of Copper (Book 2)*, P.A. Riveros, D. Dixon, D. Dreisinger, and J. Menacho, eds., The Canadian Institute of Mining, Metallurgy and Petroleum (CIM), Montreal, 2003, pp. 493–516.
4. D.G. Dixon: *Hydrometallurgy*, 2000, vol. 58, pp. 27–41.
5. H. Kametani and A. Aoki: *Metall. Trans. B*, 1985, vol. 16B, pp. 695–705.
6. M. Gericke, A. Pinches, and J.V. van Rooyen: *Int. J. Miner. Process.*, 2001, vol. 62, pp. 243–55.
7. K.A. Third, R. Cord-Ruwisch, and H.R. Walting: *Biotechnol. Bioeng.*, 2002, vol. 78 (4), pp. 433–40.
8. J. Vilcaez, K. Suto, and C. Inoue: *Hydrometallurgy*, 2006, vol. 94, pp. 82–92.
9. F.K. Crundwell and A.F. Norton: Patent No. US, 7,575,622 B2, 2009.