

Leaching of gold ores with high cyanicides: a physico-chemical modeling approach

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Keywords	Abstract
<i>Leaching</i>	Processing of gold ores with high sulfide minerals is problematic as they consume cyanide and reduce gold leaching. Optimization of gold leaching and cyanide consumption requires a methodology to estimate the amount of exposed cyanicides, their leaching kinetics, and speciation of cyanide complexes that consume the free cyanide and compete with gold. In this paper, a physico-chemical approach is presented to estimate the liberation and exposure of cyanicides to the leaching solution, and then prediction of the speciation of all possible related species in the solution. The results obtained show that this methodology not only could successfully estimate the gold leaching and cyanide consumption based on the mineralogical data with a lower number of parameters compared to existing empirical models, but also offers the prediction of formation of all the possible complexes that could be used for optimization purposes.
<i>Liberation</i>	
<i>Cyanide Consumption</i>	
<i>Kinetics</i>	
<i>Speciation</i>	

1. Introduction

In the processing of gold ores with high sulfide minerals, the consumption of cyanide is high, and therefore, could make the conventional cyanidation unprofitable [1, 2]. Moreover, environmental concerns over the cyanide compounds in gold mining wastewater is serious due to the severe toxic properties of different cyanide compounds [3]. In typical cyanide solutions, gold, pyrite, and chalcopyrite dissolve to form the soluble gold, iron, and copper cyanide complexes [4, 5]. The solution pH and the cyanide-to-copper ratio play an important role in the formation of cyanide complexes [6]. When free CN^- is too low, $FeOOH(s)$ and $Cu(OH)_2$ may be precipitated [7]. It is difficult to develop a rigorous phenomenological model of leaching due to the impressive variety of complexes that can be formed with cyanide and the complexity in assessment of the exposure of cyanicides to the solution. As a result, only empirical models have been presented in the literature, while their

applicability is limited to the specific ore and laboratory conditions used. De Andrade Lima and Hodouin (2005) have proposed a pseudo-homogenous empirical model to describe the leaching process [8]. Their model considers the ore particle size (as an indication of liberation of gold and cyanicides), cyanide concentration, and concentration of copper- and sulfur-bearing minerals. Khalesi et al. (2009) have developed an integrated grinding and liberation model for the size reduction of gold ores and a simulator of gold grain exposure of ground ore to add the liberation of the gold ore to the leaching model [9, 10]. Bellec et al. (2009) [11] have proposed an empirical model that is a function of the ore particle size distribution, and cyanide, copper, and sulfur concentrations. In their leaching model, four classes of different zones for position of gold grains have been postulated, as shown in Figure 1. The position influences the degree of surface accessibility to reagents (exposure).

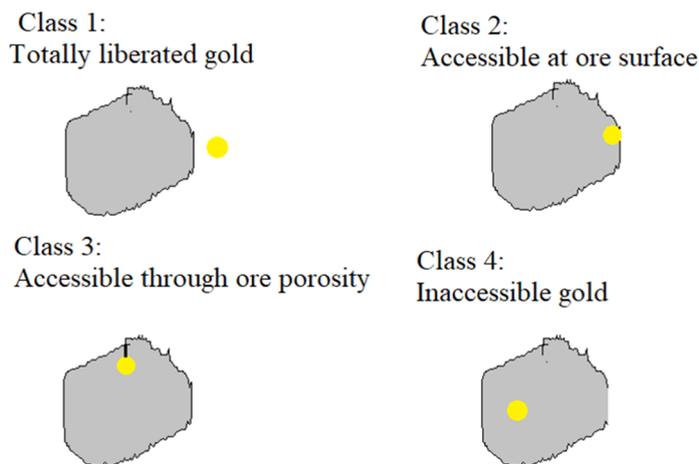


Figure 1. Four classes of gold accessibility for leaching [11].

Gold can be accessible at the ore surface, totally liberated or accessible through the pores. The fourth class contains inaccessible gold that cannot be leached. The difference in leaching behavior of totally liberated gold and exposed gold grains at the ore surface is due to the fact that the first one suggests more accessible surface to cyanide. Due to the complexity of the process, their model applies 24 parameters for prediction of gold leaching and cyanide consumption [11]. They showed that the rate of leaching reaction for these three classes could be shown as below:

$$v_l = \frac{4RD_{CN}[CN(t)]D[O_2(t)]}{\sigma_0(D_{CN}[CN(t)]+8RD_{O_2}[O_2(t)])} \quad (1)$$

$$v_e = \frac{4RD_{CN}[CN(t)]D[O_2(t)]}{(\sigma_0+d_g(0)\left(\frac{1-me(t)}{me(0)}\right))(D_{CN}[CN(t)]+8RD_{O_2}[O_2(t)])} \quad (2)$$

$$v_p = \frac{4RKD_{CN}[CN(t)]D[O_2(t)]}{(K(\sigma_0+\sigma_l(0))+\sigma_p)(D_{CN}[CN(t)]+8RD_{O_2}[O_2(t)])} \quad (3)$$

where D_{CN} , D , σ_0 , R , K , and σ_p are the cyanide diffusivity, oxygen diffusivity, distance between gold surface and bulk solution, oxygen efficiency, constant factor used to approximate hindered transport through pores, and threshold distance from the ore surface above which gold/minerals grains are not accessible to chemical attack by cyanide, respectively; v_l , v_e , and v_p are the molar rates of liberated, partially exposed, and accessible through ore porosity gold dissolution per unit of gold surface available to reaction. Equations 1 to 3 could be used in mass balance equations as follow:

$$\frac{dm_l(t)}{dt} = -\frac{6M m_l(0)^{1/3}}{\rho d(0)} v_l m_l(t)^{2/3} \quad (4)$$

$$\frac{dm_e(t)}{dt} = -\frac{Mm_e(0)^{1/3}}{\rho d(0)} v_e m_e(t)^{2/3} \quad (5)$$

$$\frac{dm_p(t)}{dt} = -\frac{Mm_p(0)^{1/3}}{\rho d(0)} v_p m_p(t)^{2/3} \quad (6)$$

where $m_l(t)$, $m_e(t)$, and $m_p(t)$ are the mass of totally liberated, partially liberated, and accessible through ore porosity gold, respectively; M , ρ , and d are the gold molecular weight, density, and length of presumed cubic grain sides; $t = 0$ shows the variable at the beginning of the leaching [11]. Lotz et al. (2009) have investigated the gold leaching kinetics, and assessed the arsenic and sulfur speciation by experimental analysis [12]. By modeling the mineral leaching kinetics and speciation using databases of thermodynamic values, available from computer programs such as MINTeq [13], PHREEQC [14], PHREEQCI [15], WATEQ4F [16], and PHREEQ [17], the amount and type of each complex can be calculated. Leaching of major elements and heavy metals [18] and leaching of Pb, Cd, As, and Cr from cementitious waste [19] are modeled by PHREEQC but incomplete database or lack of flexibility in kinetic modeling or calibration of models limit their applicability in estimation of speciation during leaching of complex systems. Kianinia et al. (2018) [20] have proposed a methodology to simulate the leaching of gold and sulfide minerals, and predict the cyanide consumption [20]. However, they used pure and liberated minerals, and therefore, their work was limited. The aim of the present work was to develop a physico-chemical model to simulate the leaching of a real gold ores and to estimate the cyanide consumption based on the amount of

“exposed” soluble gold and sulfide minerals in the ore (physics) and the solution conditions such as pH and free cyanide concentration (chemistry). The phenomenological nature of such a model makes it possible to apply it for different ores, and passes the limited applicability of empirical models. Besides, such a model tries to give an understanding on the mechanisms and contributions of cyanicides on cyanide consumption and gold leaching behavior in the real ores.

2. Methodology

This work includes modeling the gold and minerals leaching kinetics, which considers their degree of liberation and modeling the speciation based on the amount of leached minerals. The experimental data from Bellec et al. (2009) [11] was used to validate the method. To the knowledge of the authors, the Bellec’s work is one of the few research works integrating the exposure of gold to leaching kinetics. Therefore, his dataset was chosen so that the results of this paper could be comparable to his, and show that bringing thermodynamic calculations to the whole model reduce the number of parameters drastically, and convert the empirical approach of Bellec et al. (2009) [11] in prediction of cyanide consumption to a phenomenological model with less limitations.

2.1. Leaching model

In order to include the liberation of the minerals in the method, the leaching model developed by Bellec et al. (2009) [11] (Equations 1-6) was used, which takes into account the exposure degree of leached species. Pyrite and chalcopyrite were determined as the main cyanicides of the ore based on the mineralogical data [21]. The mentioned equations were used for cyanicides and gold, while in the work carried out by Bellec et al. (2009) [11], this model has been applied only for gold. Based on the stoichiometry, it was assumed that from one mole of chalcopyrite, one mole of Cu, one mole of Fe, and two moles of S would be released to the solution.

2.2. Speciation model

The equilibrium position of a set of simultaneous reactions subjected to the constraints of mass balance and mass action were solved in *MATLAB* by a non-linear least squares optimization method. Based on the mineralogy of the ore, the possible species that could be formed in cyanide solution were determined through the database of

PHREEQC software and the related papers [7, 22, 23], which are all shown in Table 1. According to the equations in Table 1, the master species were chosen so that they represented all the chemical elements in each system, and then Table 2 was written for the Cu-CN-Fe-Au-S-O-H system. The entries in the columns of Table 2 are the stoichiometric coefficients of species formation, and the last column is the equilibrium constant of the reactions (log K). For example, one mole of $\text{Fe}(\text{CN})_6^{4-}$ is formed with one mole of Fe, six moles of CN^- , and zero mole of other elements. By multiplying across rows, it is possible to determine the species concentration, and by summing down the columns, the total amount of each element can be recovered. Free cyanide for leaching of gold can be extracted as [20]:

$$C(\text{CN}^-) = C_{\text{CN,T}} - (C(\text{HCN}) + 2C(\text{Cu}(\text{CN})_2^-) + 3C(\text{Cu}(\text{CN})_3^{2-}) + 4C(\text{Cu}(\text{CN})_4^{3-}) + 6C(\text{Fe}(\text{CN})_6^{4-}) + \dots) \quad (7)$$

Leaching of minerals/gold and formation of complexes in the solution was coded in two *MATLAB* m-files that interact with each other, as shown in Figure 2. The leaching model simulates the kinetics of minerals/gold leaching and estimates the concentration of total elements in the solution during the leaching process. The inputs of the kinetic model are the reaction rate constants K, D, and D_{CN} for each mineral/gold and σ_0 for all minerals and gold. The outputs of the leaching model are the total concentrations of Cu, S, Fe, CN, and Au in the solution. The speciation model uses the output of the leaching model for simulating the speciation based on the total concentration of the elements, temperature, and pH. The kinetic model only uses the free cyanide concentration from output of the speciation model. Parameters of the leaching model were estimated by a non-linear least squares optimization method in *MATLAB* with minimizing the sum of squared differences between the experimental and simulation results of the Au, Fe, Cu, and free cyanide concentrations.

2.3. Input data

The data from twelve batch leaching tests performed by Bellec et al. (2009) [11] on samples from the *Sleeping Giant* gold mine ore, Quebec, Canada was used [11]. Those tests were executed on a ground ore and pre-aerated on 50% passing 38 micrometer particles at pH 11, 50% solid, and saturated oxygen. The initial cyanide concentration was 600 ppm. In order to analyze the cyanide and gold concentrations, liquid

samples were regularly collected. After each liquid sampling, cyanide was added to the leaching tank to maintain the cyanide concentration near its initial value. The detailed

experimental procedure can be found in [11]. Table 3 summarizes the twelve tests performed at four different agitation speeds, and their initial conditions.

Table 1. Main reactions of Cu-CN-Fe-Au-S-O-H system (25°C, 1 atm, and I = 0).

Equilibrium	log K	Ref.
$H_2O = H^+ + OH^-$	-13.98	[14]
$Au^+ + CN^- = AuCN(s)$	38.9	[14]
$\frac{1}{2}O_2 + CN^- = OCN^-$	3.5	[28]
$Cu^+ + 2CN^- = Cu(CN)_2^-$	23.7	[29]
$Fe^{2+} + 6CN^- = Fe(CN)_6^{4-}$	35.4	[14]
$S + CN^- = SCN^-$	0.9	[28]
$Cu^+ + 3CN^- = Cu(CN)_3^{2-}$	28.5	[29]
$Cu^{2+} + 2H_2O = Cu(OH)_2 + 2H^+$	-16.24	[14]
$Cu^+ + 4CN^- = Cu(CN)_4^{3-}$	30.6	[29]
$Fe^{2+} + 3H_2O = Fe(OH)_3^- + 3H^+$	-32	[14]
$Fe^{3+} + 2H_2O = FeOOH + 3H^+$	-0.5	[14]
$Cu(CN)_2^- = CuCN(s) + CN^-$	-4.91	[14]
$CN^- + H^+ = HCN$	9.21	[14,19]
$Au^+ + 2CN^- = Au(CN)_2^-$	39.3	[2]

Table 2. Main components of Cu-CN-Fe-Au-S-O-H system.

Species	H ⁺	Cu	CN	Fe	Au	S	O ₂	log k
H ⁺	1	0	0	0	0	0	0	0
S	0	0	0	0	0	1	0	0
CN ⁻	0	0	1	0	0	0	0	0
Au	0	0	0	0	1	0	0	0
Fe	0	0	0	1	0	0	0	0
OH ⁻	-1	0	0	0	0	0	0	-14
Cu(CN) ₂ ⁻	0	1	2	0	0	0	0	23.7
Cu(CN) ₃ ²⁻	0	1	3	0	0	0	0	28.5
Cu(CN) ₄ ³⁻	0	1	4	0	0	0	0	30.6
HCN	1	0	1	0	0	0	0	9.21
Fe(CN) ₆ ⁴⁻	0	0	6	1	0	0	0	35.4
SCN ⁻	0	0	1	0	0	1	0	0.9
OCN ⁻	0	0	1	0	0	0	0	3.5
Au(CN) ₂ ⁻	0	0	2	0	1	0	0	39.3
CuCN	0	1	1	0	0	0	0	-20
Fe(OH) ₃ ⁻	-3	0	0	1	0	0	0	-32
FeOOH	-1	0	0	1	0	0	1	-0.5
Cu(OH) ₂	-2	1	0	0	0	0	0	-16.24
Total	Total H	Total Cu	Total CN	Total Fe	Total Au	Total S	Total O ₂	

Table 3. Initial conditions of leaching tests [11].

Experiment	Time (h)	RPM	S (%)	Cu (%)	Au (ppm)	O ₂ (ppm)
1	1	1200	2.86	0.147	10.81	33.5
2	1	1350	3	0.148	9.72	33.4
3	1	1500	2.9	0.151	9.43	35.1
4	1	1650	2.97	0.158	8.48	36.9
5	2	1200	2.98	0.139	8.34	37.3
6	2	1350	2.87	0.157	8.53	38.1
7	2	1500	2.89	0.164	7.53	41
8	2	1650	2.86	0.149	11.18	37.9
9	4	1200	2.74	0.18	9.05	35.3
10	4	1350	2.92	0.142	9.56	33.9
11	4	1350	2.77	0.148	7.2	37.1
12	4	1650	2.78	0.146	9.5	32.8

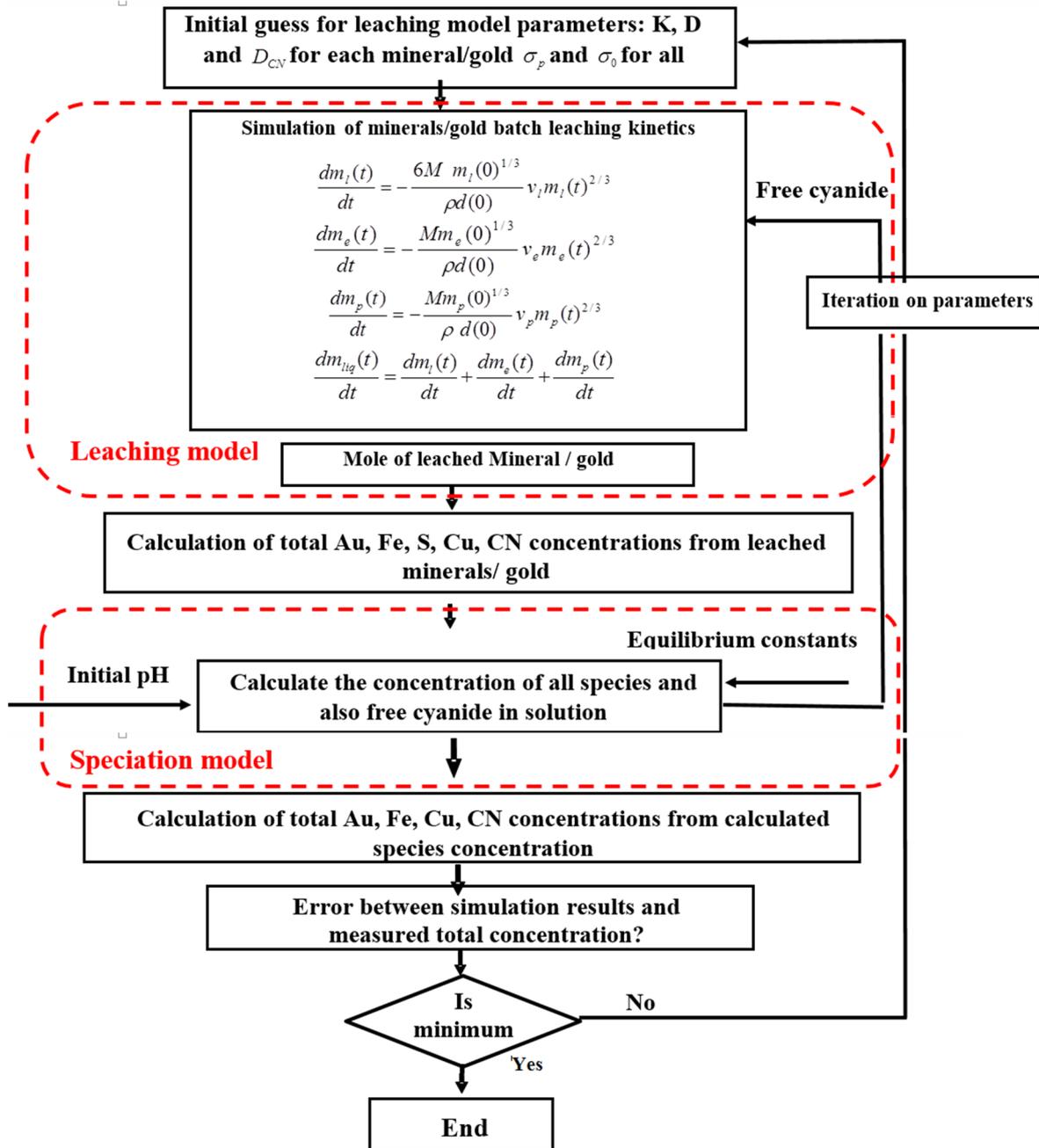


Figure 2. Methodology used for calibration of the model used for estimation of gold/minerals leaching, speciation, and cyanide consumption.

3. Results and discussion

Like the work of Bellec et al. (2009) [11], the results of nine tests were used for model calibration, and the remained ones were used for validation. These were the two leaching tests at 1500 rpm as well as the 1-hour and 4-hour tests at the three other agitation speeds. Table 4 shows the calibrated parameters for gold and other cyanide leaching kinetic model and also the numerical values of estimated parameters for gold leaching

kinetic model from Bellec et al. (2009) [11]. Comparisons between the simulated and experimental gold and cyanide concentrations in solution are shown in Figures 3 and 4, respectively.

Although liberation and exposure of other minerals and formation of complexes are included in this paper, the estimated parameters for gold leaching kinetic are not very different from the work of Bellec et al. (2009) [11].

Table 4. Numerical values for estimated parameters.

Mineral	K	D_{CN} $\mu\text{m}^2 / \text{min}$	D $\mu\text{m}^2 / \text{min}$	σ_{0-1200} μm	σ_{0-1350} μm	σ_{0-1500} μm	σ_{0-1650} μm	σ_p μm
Pyrite	0.04	14300	94286	25.51	16.44	7.01	4.75	9.95
Chalcopyrite	0.045	14300	94286	25.51	16.44	7.01	4.75	9.95
Gold	0.047	14300	94286	25.51	16.44	7.01	4.75	9.95
Gold in the reference	0.067	14300	94200	20.51	8.48	6.02	3.77	9.95

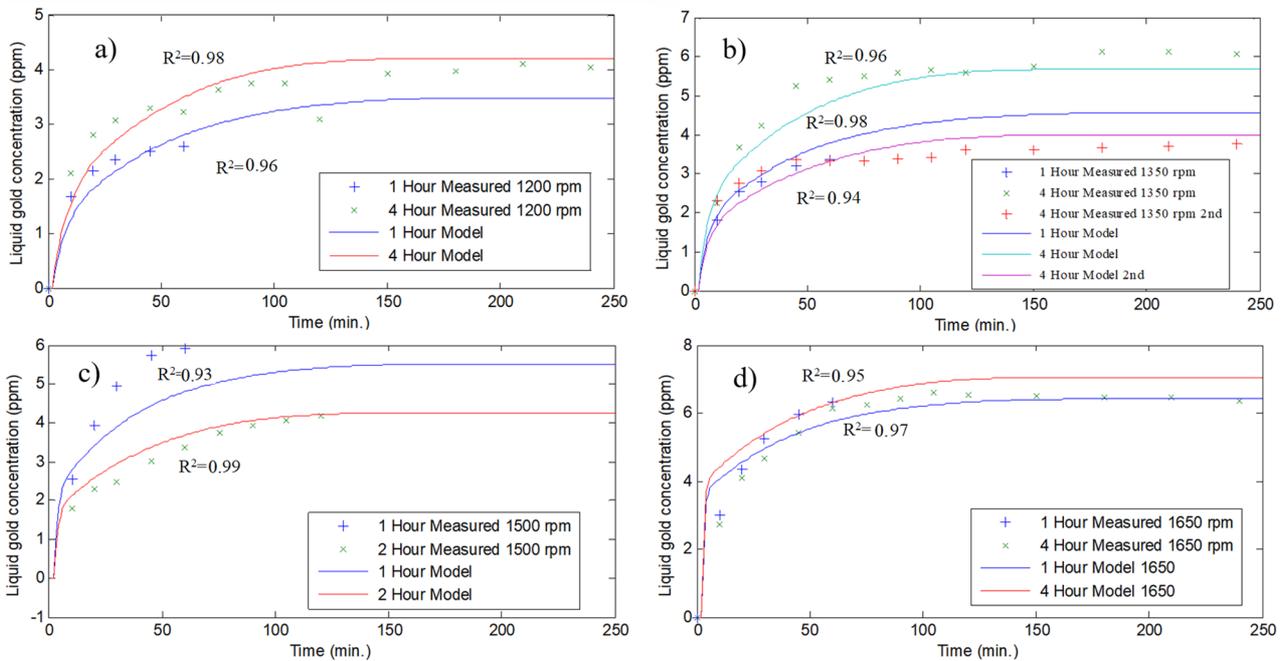


Figure 3. Comparison between experimental and simulated gold liquid concentrations (a) Results at 1200 rpm, (b) Results at 1350 rpm, (c) Results at 1500 rpm, (d) Results at 1650 rpm.

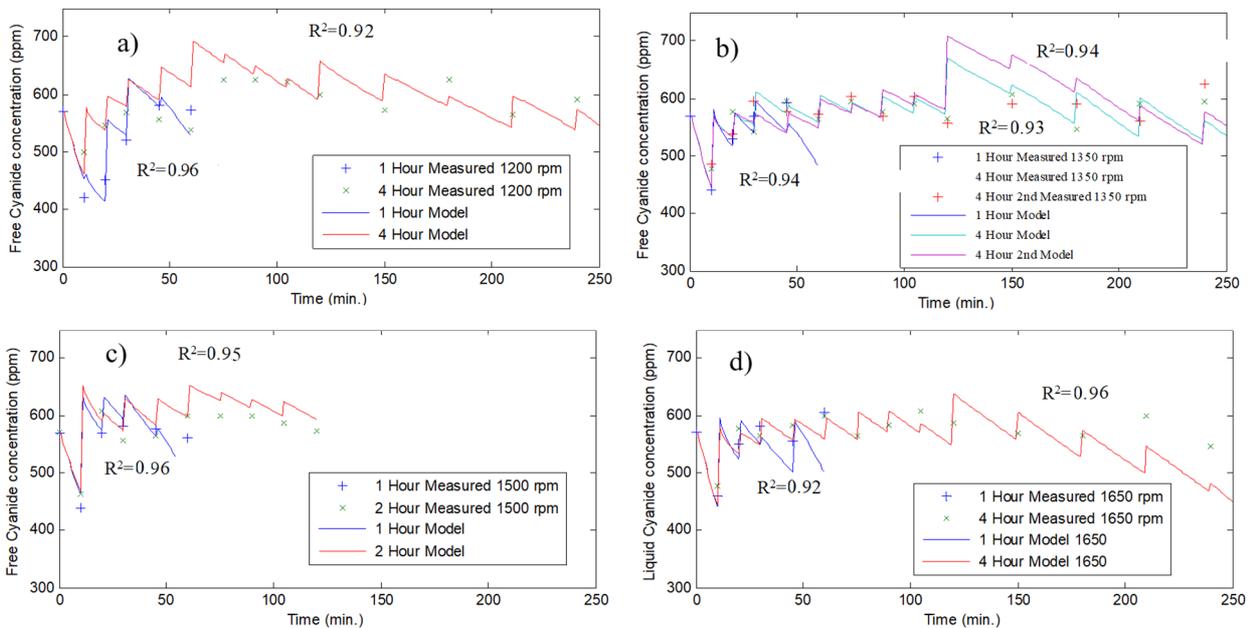


Figure 4. Comparison between experimental and simulated cyanide concentrations (a) Results at 1200 rpm, (b) Results at 1350 rpm, (c) Results at 1500 rpm, (d) Results at 1650 rpm.

4. Cross-validation

In order to validate the proposed methodology, the three 2-hour cyanidation tests at 1200, 1350, and 1650 rpm were used. Figures 5 and 6 show the results predicted by the model with the measured liquid concentrations of gold and cyanide. The results obtained show an acceptable level of

confirmation between the models and measured data. It should be noted that here, all the thermodynamic phenomena of formation of different cyanide complexes are linked to exposure of minerals and gold, while in the work of Bellec et al. (2009) [11], an empirical model estimated the cyanide consumption.

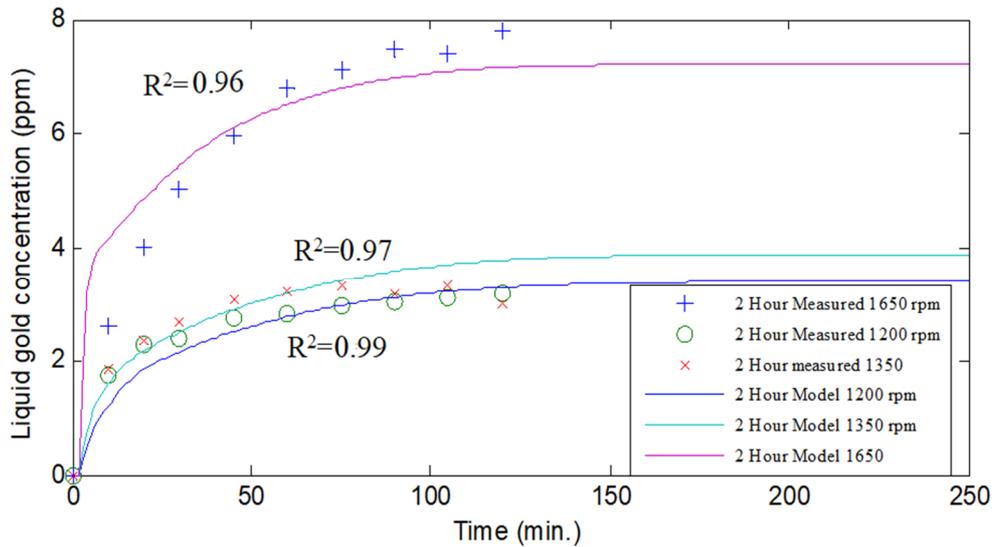


Figure 5. Comparison between experimental and predicted gold liquid concentrations.

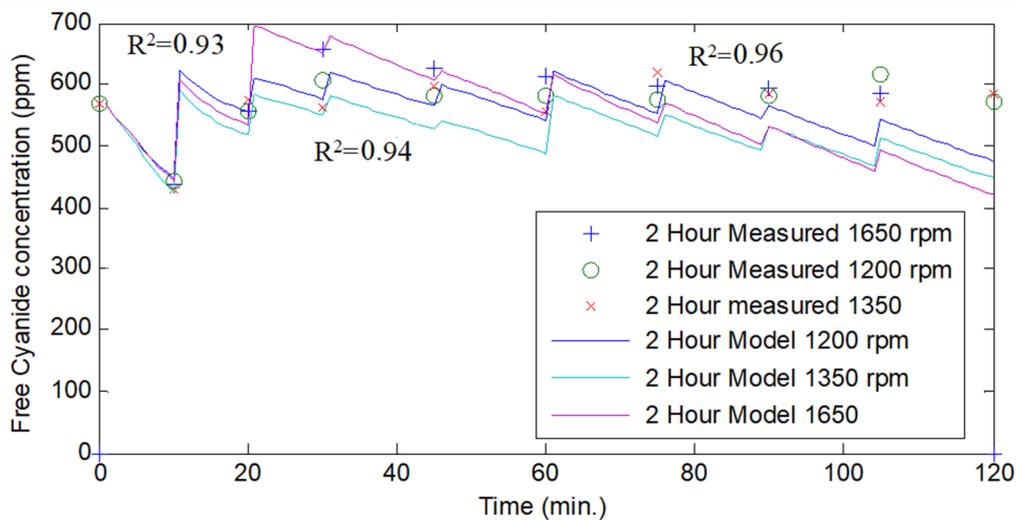


Figure 6. Comparison between experimental and predicted cyanide concentrations.

5. Conclusions

An integrated liberation, leaching, and speciation model for simulation of a real gold ore that includes the exposure data of minerals and gold (physics) and solution conditions (chemistry) has been proposed in this paper. Calculations were developed in *MATLAB* software, and a non-linear least squares optimization method was used to estimate the parameters of the proposed model.

The Bellec et al.'s (2009) [11] leaching data was applied for model calibration, and it was shown that with a lower number of parameters (10 against 24), not only the gold leaching and cyanide consumption could be calculated but also all the complexes formed could be simulated via the thermodynamic calculations implemented inside the simulator. If other minerals capable of leaching and formation of complexes with cyanide

were present in the ore, their reaction indices should be added to Tables 1 and 2 but the methodology remains the same. The model could be utilized to predict the process routes based on the mineralogical data and degree of liberation resulting in geometallurgical flowsheet development, environmental purposes of reducing hazardous species, and optimization of reagent consumption.

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فرآوری کانسنگ طلاي حاوی کانی‌های با مصرف سیانور بالا: رویکرد مدل‌سازی فیزیکی- شیمیایی

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چکیده:

فرآوری کانسنگ طلا با مقادیر بالای کانی‌های سولفیدی به دلیل مصرف بالای سیانور و کاهش انحلال طلا مشکل‌زا است. به منظور بهینه‌سازی انحلال طلا و مصرف سیانور لازم است یک روش برای تخمین مقدار کانی‌های مصرف‌کننده سیانور در معرض قرار گرفته، سینتیک انحلال آن‌ها و شبیه‌سازی کمپلکس‌های سیانوری تشکیل‌شده که سیانور آزاد محلول را مصرف و با طلا رقابت می‌کنند موجود باشد. در این پژوهش، یک رویکرد فیزیکی- شیمیایی برای تخمین آزادشدگی و در معرض قرارگیری کانی‌های مصرف‌کننده سیانور در محلول لیچینگ و سپس پیش‌بینی کل گونه‌های احتمالی تشکیل‌شده در محلول ارائه شده است. نتایج به دست‌آمده نشان داد که روش ارائه شده نه تنها قادر به تخمین انحلال طلا و مصرف سیانور بر اساس داده‌های کانی‌شناسی با تعداد پارامترهای کمتر از مدل‌های تجربی موجود است، بلکه همه گونه‌های احتمالی قابل تشکیل در محلول را پیش‌بینی می‌کند که می‌توان از آن در اهداف بهینه‌سازی استفاده کرد.

کلمات کلیدی: انحلال، آزادشدگی، مصرف سیانور، سینتیک، تشکیل گونه‌ها.
