Model for Predictive Analysis of Residue Lead Content During Leaching of Galena by the Oxidative Action of Acidithiobacillus Ferrooxidans

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A model for predictive analysis of the lead content of residue during bioleaching of galena has been derived. The leaching solution was inoculated with acidithiobacillus ferrooxidans which oxidatively catalyzed the process. The established model:

$$\varphi_R = \left(\frac{\beta_R}{0.0449\ \beta_i} \right)$$

indicates that the mass of lead in the residue resulting from the bioleaching process is directly proportional to the ratio of percent concentration of lead in residue to percent concentration of lead in the as-beneficiated concentrate (β_R/β_i), the associated correlation being precisely unity. The validity of the model is rooted in the expression $\varphi_i \beta_R \approx \beta_i \varphi_{R;}$ both sides of the expression being correspondingly approximately almost equal. The mass of lead per unit time sipping into the reaction residue during the bioleaching process as obtained from experiment and derived model are 0.0036 g/hr and 0.0038 g/hr. This is an indication of proximate agreement between experimental and model-predicted results. The deviation of the model-predicted mass of lead (sipping into the residue) from the corresponding experimental values is approximately 4.5% all through the leaching process.

*Keywords***:** *Model, Residue Lead Content, Acidithiobacillus Ferrooxidans, Leaching, Galena*.

1. Introduction

The usefulness of lead in metallurgical and allied industries has led to concerted efforts to extract lead from its ore by applying various techniques which mainly includes acid and microbial leaching.

Experiments [1] on the quantitative leaching of lead from galena in hydrochloric acid solutions have been reported. The effects of contact time, acid concentration, number of leaching stages, solid to liquid ratio, particle size, temperature, and stirring speed, on the leaching reaction are reported. About 96% of lead was dissolved within 120 min by 8.42 M hydrochloric acid solution at 95 ^{o}C. Multi-stage leaching was not so advantageous, but the extent of leaching increased greatly with temperature. The optimum solid to liquid phase ratio and particle size were found to be 1/30 kg/L and -88+53 m, respectively. Dissolution was enhanced by stirring speed over the range 0 to 400 min-1 and reached a steady rate thereafter.

It was also found [2] during the leaching of zinc and copper out from their respective sulphide ore that the concentrations of zinc and copper formed reduced as particle size decreased while silica, sulphur, iron and lead contents increased. Also leaching rate of copper was found to be lower than zinc.

Studies [3, 4] have been carried out to evaluate the possibility of extracting metals from their respective ores. It was shown [3] that in addition to thiobacilli, some other bacteria exist, which are effective in solubilizing sulphidic minerals. In hot biotopes containing sulphur or oxidizable sulphur compounds, such as hydrothermal vents and self heating brown coal dumps, one can find an archaebacterium

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named Sulpholobus. Like thiobacilli, it is acidophilic, chemolithoautotroph and derives its energy from oxidation of sulphur and sulphur compounds and from oxidation of ferrous ion like Thiobacillus Ferrooxidans. Often, one can see in acid metal leaching biotopes spirilloid bacteria. They belong to the species Leptospirillum Ferrooxidans, a gram negative spirillum, facultatively chemolithoautotroph, deriving its energy from oxidizing ferrous ion like Thiobacillus Ferrooxidans. But in contrast to this latter bacterium, it cannot oxidize sulphur or sulphur compounds and is incapable of utilizing the ion of sulphidic minerals. Leptospirillum Ferrooxidans alone cannot solubilize sulphidic ferrous ion containing minerals. But in co-operation with Thiobacillus Thiooxidans, which on its part cannot solubilize sulphidic minerals, both bacterial together disintegrate sulphidic ferrous iron containing minerals by oxidation and bring them to solution. Also when Thiobacillus Thiooxidans is used together with Thiobacillus Ferrooxidans, the concentration of leached metal is far greater than that obtained when only T. Ferrooxidans is used. The enhancement is due to the oxidation of elemental sulphur by T. Thiooxidans culminating in the formation of hydrogen sulphide when ferric ions are used as the oxidizing agent.

Past research [4] has evaluated two species of mesophilic acidophilic bacteria; Thiobacillus Ferrooxidans and Leptospirillum Ferrooxidans, as the most significant microorganisms involved in sulfide mineral oxidation. The study also shows that moderately thermophilic (or thermotolerant) bacteria and extremely themophilic archaea are also important in certain situations, such as self heating coal spoils and bioleaching operations in which temperature exceed 40^0C. Both T. Ferrooxidans and L. Ferrooxidans are generally regarded as obligate chemolithotrophs and synthesize cell carbon via enzymic fixation of CO_2, although it has been shown that T. Ferrooxidans has a limited capacity to utilize organic-carbon.

A bioleaching process has been carried out [5] on a Nigerian ore consisting of siderite, sphalerite, galena and quartz, with traces of pyrite and chalcopyrite using mixed cultures of Acidithiobacillus ferrooxidans, Acidithiobacillus thiooxidans and Leptospirillum ferrooxidans in mechanically stirred glass reactors. The process was embarked on to ascertain the effect of ore mineralogy on the microbial leaching of low grade complex sulphide ore. This was carried out by considering the variations in the mineral and phase distribution within particle sizes of -53, 53, 75 and 106μm. The results from the process show that the highest bioleaching recoveries were obtained at a particle size of 75μm, while particle sizes of 106μm gave the least recoveries. The research shows that higher silica content which reduced acidity, iron mobility and oxidation led to lower recoveries at particle sizes of 53 and -53 μm.
The aim of this work is to derive a model for predictive analysis of the lead content of residue during bioleaching of galena using acidithiobacillus ferrooxidans.

2. Model

During the bioleaching process, the ore was assumed to be stationary in the reaction vessel and contains the un-leached lead as part of reaction remnants. The ore was attacked by Fe (iii) ions from the leaching solution within the liquid phase, and in the presence of oxygen.

2.1 Model Formulation

Results from experimental work [6] carried out at SynchroWell Research Laboratory, Enugu were used for the model derivation. These results as presented in Tables 1 and 2 show that:

$$\beta_i = \beta_S + \beta_R \tag{6}$$

Dividing equation (6) by β_i to get mass fraction

$$\left(\frac{\beta_S}{Bi}\right) + \left(\frac{\beta_R}{\beta_i}\right) = 1 \tag{7}$$

$$\text{Furthermore, } \beta_R\varphi_T = 100\varphi_R \tag{8}$$

$$\varphi_R + \varphi_S = \varphi_i \tag{9}$$

$$\varphi_i \ \beta_R \approx \beta_i\varphi_R \tag{10}$$

From equation (6),

$$\beta_R = \beta_i - \beta_S \quad (11)$$

Substituting equation (11) into equation (10) and then re-arranging gives;

$$\left(\frac{\varphi i\,(\beta_i - \beta_S)}{\varphi_R}\right) = \beta_i \quad (12)$$

$$\varphi_i\,(\beta_i - \beta_S) = \varphi_R\,\beta_i \quad (13)$$

$$\varphi_i\,(\beta_i - \beta_S) - \varphi_R\,\beta_i = 0 \quad (14)$$

$$\varphi_i\,\beta_i - \varphi_i\,\beta_S - \varphi_R\,\beta_i = 0 \quad (15)$$

Collecting like terms reduces equation (15) to ;

$$\varphi_i\,\beta_i - \varphi_R\,\beta_i - \varphi_i\,\beta_S = 0 \quad (16)$$

Further collection of like terms reduces equation (16) to:

$$\beta_i\,(\varphi_i - \varphi_R) = \varphi_i\,\beta_S \quad (17)$$

$$\varphi_i - \varphi_R = \left(\frac{\varphi i\,\beta_S}{\beta_i}\right) \quad (18)$$

From equation (8)

$$\varphi_R = \left(\frac{\beta_R \varphi_T}{100}\right) \quad (19)$$

Substituting equation (19) into equation (18) reduces it to:

$$\varphi_i - \left(\frac{\beta_R \varphi_T}{100}\right) = \left(\frac{\varphi i\,\beta_S}{\beta_i}\right) \quad (20)$$

$$\varphi_i - \left(\frac{\beta_R \varphi_T}{100}\right) - \left(\frac{\varphi i\,\beta_S}{\beta_i}\right) = 0 \quad (21)$$

Collecting like terms in equation (21)

$$\varphi_i\left(1 - \left(\frac{\beta_S}{\beta_i}\right)\right) - \varphi_T\left(\frac{\beta_R}{100}\right) = 0 \quad (22)$$

Substituting equation (19) into equation (22) reduces it to:

$$\varphi_i\left(1 - \left(\frac{\beta_S}{\beta_i}\right)\right) - \varphi_R = 0 \quad (23)$$

$$\beta_S = \beta_i\left(\frac{\varphi_i - \varphi_R}{\varphi_i}\right) \quad (24)$$

Introducing the value of φ_i into equation (24) reduces it to:

$$\beta_S = \beta_i\,[\,1 - 0.0449\,\varphi_R\,] \quad (25)$$

From equation (11),

$$\beta_S = \beta_i - \beta_R \quad (26)$$

Substituting equation (26) into equation (25)

$$\beta_i - \beta_R = \beta_i\,[\,1 - 0.0449\,\varphi_R\,] \quad (27)$$

Dividing both sides of equation (27) by β_i

$$\varphi_R = \left(\frac{\beta_R}{0.0449\,\beta_i}\right) \quad (28)$$

where

φ_i = 22.27g (Average total mass of lead in the as-beneficiated galena concentrate as determined in the experiment [6])

φ_S = Mass of lead in solution (g).

φ_R = Mass of lead in residue (g).

φ_T = Total mass of residue (g).

β_R = Concentration of lead in residue (%)

β_S = Concentration of lead in solution (%)

β_i = 50.53% (Concentration of lead in the as-beneficiated galena concentrate as determined in the experiment [6]).

Equation (28) is the derived model**.**

Table 1: Variation of the leaching time with percent lead content of residue and solution [6]

Time (hrs)	β_i (%)	β_R (%)	β_S (%)
70	50.53	46.40	4.13
140	50.53	40.01	10.52
210	50.53	43.08	7.45
280	50.53	42.37	8.16
350	50.53	41.42	9.11
420	50.53	42.37	8.16
490	50.53	43.08	7.45

Table 2: Variation of the leaching time with mass of lead in residue and solution [6]

Time (hrs)	φ_i (g)	φ_R (g)	φ_S (g)
70	22.27	19.57	2.70
140	22.27	16.87	5.40
210	22.27	18.17	4.10
280	22.27	17.87	4.40
350	22.27	17.47	4.80
420	22.27	17.87	4.40
490	22.27	18.17	4.10

3. Boundary and Initial Condition

The lead sulphide ore was placed in cylindrical flask 30 cm high containing leaching solution (2.0 g/dm^3 Fe (II) liquid solution) inoculated with acidithiobacillus ferrooxidans. The leaching solution is non flowing (stationary). Before the start of the leaching process, the flask was assumed to be initially free of attached bacteria and other micro organism. Initially, the effect of oxygen on the process was assumed to be atmospheric. Mass of galena used; 60g. Initial pH of leaching solutions used: 2 and range of leaching time: 70 - 490hrs were used for all samples. A constant leaching temperature; 25°C and average ore grain size: 150 µm were also used. Details of the experimental technique are as presented in the report [6].

The leaching process boundary conditions include: atmospheric levels of oxygen (considering that the cylinder was open at the top) at both the top and bottom of the ore particles in the gas and liquid phases respectively. A zero gradient was assumed for the liquid scalar at the bottom of the particles and for the gas phase at the top of the particles. The sides of the particles were assumed to be symmetries.

4. Model Validation

The formulated model was validated by calculating the deviation (from the corresponding experimental values) of the model-predicted lead content of the residue.

The deviation recorded is believed to be due to the fact that the surface properties of the ore and the physiochemical interactions between the ore and bioleaching solution which were found to play vital roles during the actual leaching process [6] were not considered during the model formulation. It is expected that introduction of correction factor to the predicted mass of lead in the reaction residue, gives exactly the experimental mass of lead.

Deviation (Dv) (%) of model-predicted mass of lead (in residue) from those of the experiment is given by:

$$D_v = \left(\frac{P_m - E_m}{E_m}\right) \times 100 \qquad (29)$$

where P_m *= Mass of lead in residue as predicted by model*

E_m *= Mass of lead in residue as obtained from experiment [9]*

Since correction factor (Cr) is the negative of the deviation,

$$Cr = - Dv \qquad (30)$$

Substituting equation (29) into equation (30) for Dv,

$$C_r = - 100\left(\frac{P_m - E_m}{E_m}\right) \qquad (31)$$

It was observed that addition of the corresponding values of Cr from equation (31) to the model-predicted mass of lead (in residue) gave exactly the corresponding experimental values [6].

5. Results and Discussion

Equation (28) is the derived model**.** Computational analysis of results from the experiment [6] as presented in Tables 1and 2 gave rise to Table 3.

Table 3: Variation of $\varphi_i \beta_R$ with $\varphi_R \beta_i$

$\varphi_i \beta_R$	$\varphi_R \beta_i$
1033.33	988.87
891.02	852.44
959.39	918.13
943.58	902.97
922.42	882.76
943.58	902.97
959.39	918.13

where

$\varphi_i \beta_R$ = *Average total mass of lead in the as-beneficiated galena concentrate* x *percent concentration of lead in residue*

$\varphi_R \beta_i$ = *Mass of lead in residue* x *percent concentration of lead in the as-beneficiated galena concentrate*

Figure 1 shows that the masses of lead per unit time sipping into the reaction residue as obtained from the experiment [6], designated by the line ExD and as predicted by the model (line MoD) are in proximate agreement all through the leaching time range (70-490 hrs). The validity of the model is believed to be rooted on equation (10) where both sides of the equation are correspondingly approximately almost equal. Table 3 also agrees with equation (10) following the values of $\varphi_i \beta_R$ and $\beta_i \varphi_R$ evaluated following statistical and computational analysis carried out on the experimental results in Tables 1 and 2.

5.1 Mass of lead per unit time sipping into the reaction residue

The mass of lead per unit time, sipping into the reaction residue as a result of bioleaching of the galena within the time range; 140 – 420 hrs was determined following comparison of the mass of lead per unit time obtained by calculations involving experimental results, and model-predicted results obtained directly from the model.

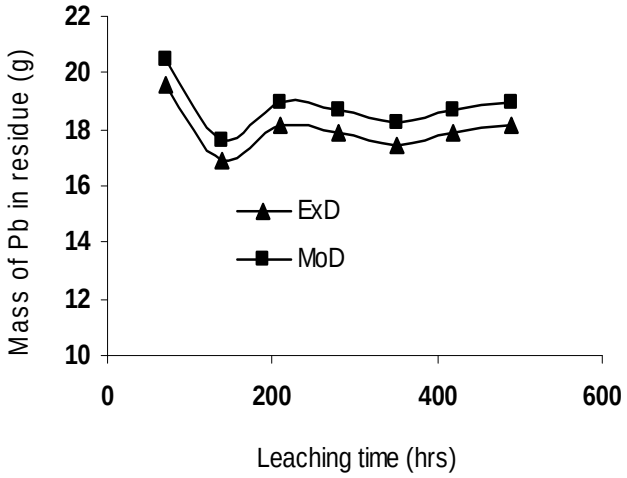

Figure 1: Comparison of the masses of lead (sipping into the reaction residue) as obtained from experiment [6] and derived model.

Mass of lead per unit time (sipping into the reaction residue), M_r (g/hrs) was calculated from the equation:

$$M_r = M / t \qquad (32)$$

Therefore, a plot of concentration of lead content of the residue M against leaching time t, as in Fig. 1 (for curve ExD) using experimental results in Table 2, gives a slope, S at points (16.87, 140) and (17.87, 420) following their substitution into the mathematical expression:

$$S = \Delta M / \Delta t \qquad (33)$$

Equation. (33) is detailed as

$$S = M_2 - M_1 / t_2 - t_1 \qquad (34)$$

where ΔM = *Change in the masses* M_2, M_1 *at two leaching time values* t_2, t_1.

Considering the points (16.87, 140) and (17.87, 420) for (M_1, t_1) and (M_2, t_2) respectively, and substituting them into eqn. (34), gives the slope as 0.0036 g/hrs which is the mass of lead per unit time (sipping into the reaction residue) during the actual experimental bioleaching process. Also similar plot (as in Fig. 1, curve MoD) using model-predicted results gives a slope S = 0.0038 g/hrs on substituting points (17.63, 140) and (18.68, 490) for (M_1, t_1) and (M_2, t_2) respectively into eqn. (34).

5.2 Deviation of model-predicted mass of lead per unit time, sipping into the reaction residue and its associated correction factor

The deviation (from experimental values) of the model-predicted mass of lead per unit time, sipping into the reaction residue is approximately 4.5% all through the leaching time range 70 – 490 hrs.

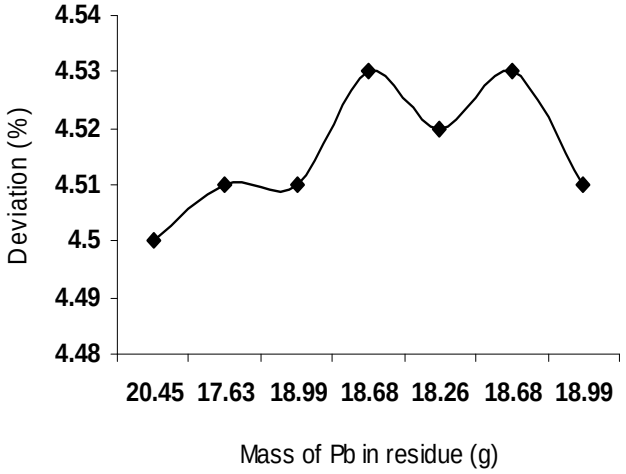

Figure 2: Variation of model-predicted mass of lead (in residue) with its associated deviation

Figure 2 shows that the least and highest magnitudes of deviation of the model-predicted lead content of the reaction residue (from the corresponding experimental values) are + 4.5% and + 4.53% which correspond to leaching times 70 and 280 hrs and lead masses 20.45 and 18.68 g respectively.

Figure 3 similarly shows that the least and highest correction factors to the model-predicted lead content of the residue are - 4.5% and - 4.53% also correspond to leaching times 70 and 280 hrs and lead masses 20.45 and 18.68 g respectively. These are opposite that of deviation values. This is because correction factor is the negative of the deviation as shown in eqns. (30) and (31). It is believed that the correction factor takes care of the effects of the surface properties of the ore and the physiochemical interaction between the ore and the bioleaching solution which (affected experimental results) were not considered during the model formulation.

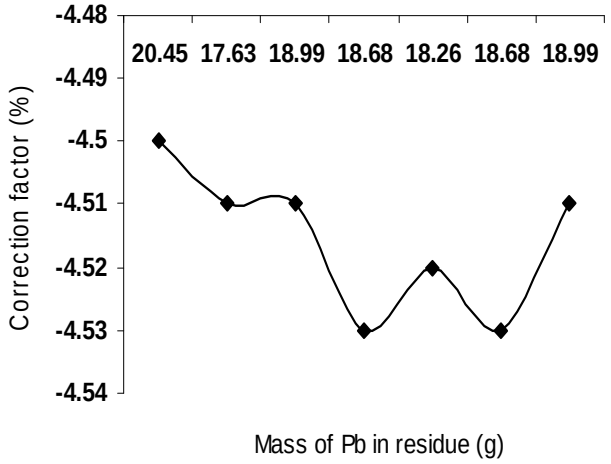

Figure 3: Variation of model-predicted mass of lead (in residue) with its associated correction factor

Comparative analysis between the derived model ($\varphi_R = \beta_R / 0.0449\ \beta_i$) and Fig.4 indicates that the model-predicted mass of lead in residue is directly proportional to the ratio of percent concentration of lead in residue to percent concentration of lead in the as-beneficiated concentrate (β_R / β_i), the associated correlation being unity.

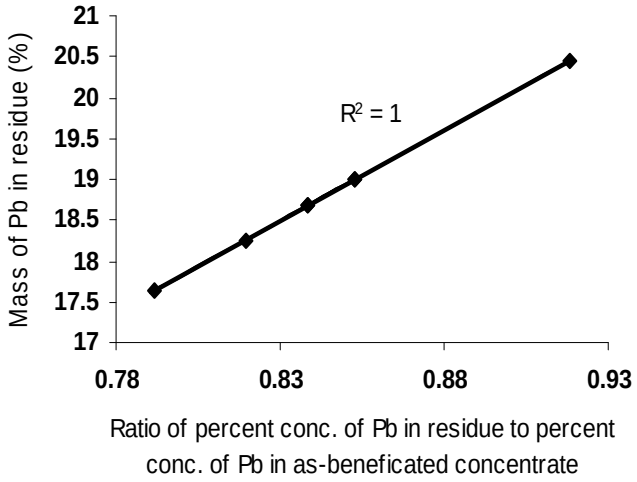

Figure 4: Variation of β_R / β_i ratio with model-predicted mass of lead in residue

6. Conclusion

The model predicts the mass of lead sipping into the residue directly proportional to the ratio of percent concentration of lead present in the residue to percent concentration of lead in the as-beneficiated galena concentrate during leaching of Ishiagu (Nigeria) galena using a culture of acidithiobacillus ferrooxidans. The validity of the model is rooted in the expression $\varphi_i \beta_R \approx \beta_i \varphi_{R;}$ both sides of the expression being correspondingly approximately almost equal. The mass of lead per unit time sipping into the reaction residue during the bioleaching process as obtained from experiment and derived model are 0.0036 g/ hr and 0.0038 g/ hr. This is an indication of proximate agreement between experimental and model-predicted results. The deviation of the model-predicted mass of lead (sipping into the residue) from the corresponding experimental values is approximately 4.5% all through the leaching process.

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