

THE KINETICS OF INDONESIAN GALENA CONCENTRATE LEACHING IN FLUOROSILICIC ACID AND HYDROGEN PEROXIDE AS ITS OXIDANTS

KINETIKA PELINDIAN KONSENTRAT GALENA INDONESIA DALAM LARUTAN ASAM FLUOROSILIKAT DAN HIDROGEN PEROKSIDA SEBAGAI OKSIDATOR

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ABSTRACT

Lead smelting is known as a lead extraction technique which has negative impact to environment because it produces emissions of dust, SO₂ gas and lead. The leaching is conducted to reduce the smelting's weaknesses by extracting the lead without damaging the environment. This technique, often called hydrometallurgical lead extraction, uses liquid media to extract the lead and separate it from the elemental sulphur (S⁰) by filtration. Particle size, slurry density (solid percentage) and acid concentration affect the dissolution of Indonesian galena concentrate with fluorosilicic acid and hydrogen peroxide as oxidants. By means of these reagents, it is expected that the Pb extraction process from galena concentrate resulted from flotation of sulfide ore can be carried out at low temperatures and lead recovery can be accomplished from the leach solution by electrowinning. Studying galena leaching kinetics is required to perform galena leaching with high Pb extraction percentages. Two kinetics models are commonly used to study leaching kinetics, namely shrinking core model (SCM) and shrinking particle model (SPM). These models are used to determine the mechanism which controls leach reaction and reaction rate constant or diffusion constant of the reacting species. Total plotting of sulfide fraction dissolves with time adjusted to the existing mathematical model, and fitting the curve from the dummy model to the experimental data, are two techniques that are utilized to obtain the best kinetics model which is important in controlling the leaching reaction rate. Applying the Arrhenius equation, the relation between the reaction rate constant or the diffusion constant and the temperature is determined to calculate the leaching activation energy. The result from kinetics analysis showed that the leaching of Indonesian galena concentrate in fluorosilicic acid with hydrogen peroxide as oxidants followed the shrinking core model with diffusion through porous solid product layer as the reaction controller with activation energy of 45.81 kJ/mol or 10.91 kcal/mol.

Keywords: galena concentrate, leaching, fluorosilicic acid, kinetics, activation energy.

ABSTRAK

Peleburan timah dikenal sebagai teknik ekstraksi timah yang memiliki dampak negatif terhadap lingkungan karena menghasilkan emisi debu, gas SO₂ dan timah. Pelindian hadir untuk mengurangi sejumlah kelemahan peleburan, dengan mengekstraksi timbal tanpa merusak lingkungan. Teknik ini, yang sering disebut ekstraksi timbal secara hidrometalurgi, menggunakan media cair untuk mengekstrak timbal dan memisahkannya dari unsur sulfur (S⁰) melalui penyaringan. Ukuran partikel, berat jenis bubur (persentase padatan) dan konsentrasi asam memengaruhi kelarutan konsentrat galena Indonesia dalam asam fluorosilikat dan hidrogen peroksida sebagai oksidator. Dengan menggunakan reagen-reagen

ini, diharapkan proses ekstraksi Pb dari konsentrat galena yang dihasilkan dari flotasi bijih sulfida dapat dilakukan pada suhu rendah dan perolehan timbal dapat dicapai dari larutan kaya dengan electrowinning. Mempelajari kinetika pelindian galena diperlukan untuk melakukan pelindian galena dengan persen ekstraksi Pb yang tinggi. Terdapat dua model kinetika yang biasa digunakan untuk mempelajari kinetika pelindian, yaitu model inti menyusut (SCM) dan model partikel menyusut (SPM). Kedua model ini digunakan untuk menentukan mekanisme yang mengendalikan reaksi pelindian dan konstanta laju reaksi atau konstanta difusi dari spesi-spesi yang bereaksi. Pengaluran fraksi sulfida total yang larut terhadap waktu yang disesuaikan dengan model matematika yang ada dan pencocokan kurva dari kurva dummy terhadap data percobaan adalah dua teknik yang digunakan untuk mendapatkan model kinetika terbaik yang memiliki peran terpenting dalam mengendalikan laju reaksi pelindian. Melalui penerapan persamaan Arrhenius, ditentukan hubungan konstanta laju reaksi atau konstanta difusi dengan suhu untuk menghitung energi aktivasi pelindian. Hasil analisis kinetika menunjukkan bahwa pelindian konsentrat galena Indonesia dalam asam fluorosilikat dan hidrogen peroksida sebagai oksidator mengikuti model inti menyusut dengan difusi melalui lapisan produk padat berpori sebagai pengendali reaksi yang memiliki energi aktivasi sebesar 45,81 kJ/mol atau 10,91 kkal/mol.

Kata kunci: konsentrat galena, pelindian, asam fluorosilikat, kinetika, energi aktivasi.

INTRODUCTION

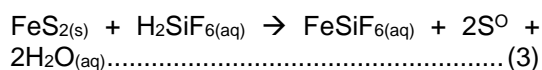
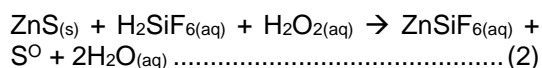
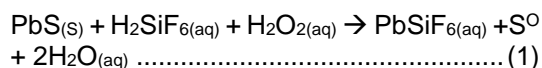
Lead smelting is believed to have a number of impacts on the environment because it produces emissions of dust, SO₂ gas and lead. To overcome such smelting's weaknesses, leaching is a promising process to extract lead without damaging the environment.

In hydrometallurgical lead extraction, sulphur is separated from the Pb as the elemental sulphur (S⁰) and removed by filtration. The produced elemental sulfur is a porous solid and does not inhibit the dissolution of galena and becomes a sell-able by-product (Aydoğan *et al.*, 2007). The dissolution of Indonesian galena concentrate in fluorosilicic acid and hydrogen peroxide as oxidants is affected by its particle size, slurry density (solid percentage) and acid concentration (Anugrah, Mubarak and Amalia, 2018). With fluorosilicic acid and hydrogen peroxide as oxidants it is expected that the Pb extraction process from galena concentrate resulting from flotation of sulfide ore can be carried out at low temperatures, and lead recovery can be carried out from the leach solution by electrowinning. To perform galena leaching with high Pb extraction percentages, it is necessary to study its kinetics. Leaching kinetics were studied using a shrinking core model (SCM) and a shrinking particle model (SPM) as determinant mechanism which controls leach reaction and the reaction rate constant or diffusion constant of the reacting species. In this research, through the Arrhenius equation, the relation of the

reaction rate constant or the diffusion constant is determined by temperature to calculate the leaching activation energy.

METHOD

Reaction kinetics provides information about the reaction rate until equilibrium is reached. Dissolution of Pb from galena with H₂SiF₆ in the presence of H₂O₂ may be performed through Eq.(1) while parts of sphalerite and pyrite in the concentrate will be also dissolved through Eq. (2) and (3) (Anugrah, Mubarak and Amalia, 2017).



The chemical stability of hydrogen peroxide is very sensitive to temperature (Pecina *et al.*, 2008) therefore the oxidant should be poured into the reactor in a short time interval. According to Pecina *et al.* (2008), the leaching of sulfide minerals in an acidic atmosphere follows the shrinking core model (SCM) which assume the particle size does not change and the particles are spherical.

Mathematical models for each type of reaction rate controller that follows the SCM are formulated as follows (Levenspiel, 1999).

1. Diffusion through a fluid film on a mineral surface

$$\frac{t}{\tau} = 1 - (1 - X_T) = X_T \dots \dots \dots (4)$$

$$\tau = \frac{\rho_T R}{3bk_l C} \dots \dots \dots (5)$$

2. Diffusion through layer of the porous solid product (ash)

$$\frac{t}{\tau} = 1 - 3(1 - X_T)^{\frac{2}{3}} + 2(1 - X_T) \dots \dots \dots (6)$$

$$\tau = \frac{\rho_T R^2}{6bD_e C_{H_2SiF_6}} \dots \dots \dots (7)$$

3. Interface reaction

$$\frac{t}{\tau} = 1 - (1 - X_T)^{\frac{1}{3}} \dots \dots \dots (8)$$

$$\tau = \frac{\rho_T R}{bk C} \dots \dots \dots (9)$$

Meanwhile the mathematical models of the controller following the SPM are formulated as follows (Levenspiel, 1999).

1. Diffusion through a fluid film on a mineral surface

$$\frac{t}{\tau} = 1 - (1 - X_T)^{\frac{2}{3}} \dots \dots \dots (10)$$

$$\tau = \frac{\rho_T R^2}{2bDC} \dots \dots \dots (11)$$

2. Interface reaction

$$\frac{t}{\tau} = 1 - (1 - X_T)^{\frac{1}{3}} \dots \dots \dots (12)$$

$$\tau = \frac{\rho_T R}{bk C} \dots \dots \dots (13)$$

where t is the leaching time, τ is the residence time of leaching until the metal sulfides in the galena concentrate has totally dissolved and X_T is the total sulfides fraction that dissolves at the time t . For leaching of galena concentrate, it is assumed that the core is shrinking due to the dissolution of PbS, ZnS and FeS₂ and leaving sulfur behind so that X_T is calculated from the total of the moles metallic sulfides dissolved and then it is divided by the moles of the metallic sulfides in the feed (galena concentrate), where R is the average of particles radius, ρ_T are the specific molar of the ore, b is the ratio of the stoichiometric coefficient to the acid. $C_{H_2SiF_6}$ is a concentration of the fluorosilicic acid reagent, k_l is the diffusion coefficient in the diffusion film, D_e is the coefficient in the porous solid (ash) layer and k'' is the reaction rate constant.

The experiments were performed for 150 minutes with a feed weight of 10.66 g, stirrer speed of 300 rpm, H₂SiF₆ volume of 295.38 ml and H₂O₂ volume of 103.88 ml.

The procedure for conducting a kinetics study of leaching galena concentrate can be seen in the following flow chart (Figure 1).

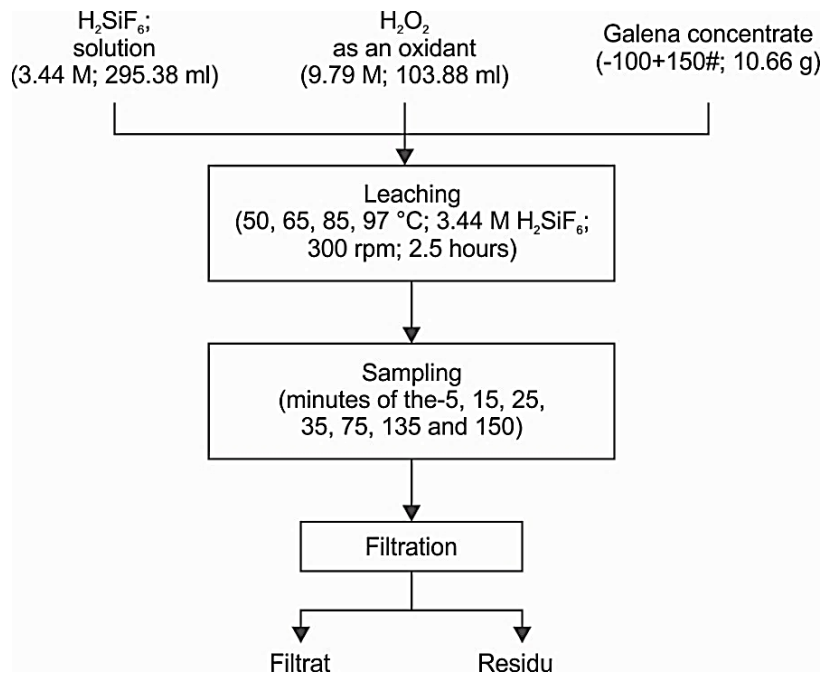


Figure 1. Experimental procedure of kinetics study of leaching galena concentrate

RESULTS AND DISCUSSIONS

According to the previous research, Anugrah, Mubarak and Amalia (2018) has determined the chemical composition of the galena concentrate as the Table 1 shows.

Table 1. Chemical composition of the galena concentrate sample (as-received sample) (Anugrah, Amalia and Mubarak, 2018)

| Element | Content (%) |
|---------|-------------|
| Pb | 66.6 |
| Zn | 7.38 |
| Cu | 0.84 |
| Fe | 2.79 |
| S Total | 17.62 |
| Sb | <0.001 |
| Mo | 0.12 |
| Co | <0.001 |

It is assumed that the dissolved metallic sulfide minerals are PbS (galena), ZnS (sphalerite) and pyrite (FeS₂). By inputting the data of the Pb, Zn and Fe metals dissolved in fluorosilicic acid and those of the Table 1, then the mole of the metallic sulfides dissolved and X_T can be calculated as shown by Table 2.

According to Astuti *et al.* (2016), the kinetics of the leaching reaction were examined over a specific temperature range. Leaching kinetics analysis was carried out from leaching experiment data at 50, 65, 85 and 97°C. Profiles of the fraction of the total mole of the converted sulfide (X_T) against time (t) for each leaching temperature are presented in Figures 1 to 4.

Table 2. Calculation of Pb, Zn and Fe sulfides dissolved mole and the fraction of the total dissolved sulfides (X_T)

| Temp. (°C) | Minutes | Hours | Metals dissolved (g/L) | | | Mole of the dissolved metals | | | Mole of the metallic sulfides dissolved | | | | X_T |
|------------|---------|-------|------------------------|------|------|------------------------------|--------|--------|---|--------|------------------|--------|--------|
| | | | Pb | Zn | Fe | Pb | Zn | Fe | PbS | ZnS | FeS ₂ | Total | |
| 50 | 0 | 0.00 | 0.00 | 0.00 | 0.00 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 |
| | 5 | 0.08 | 1.87 | 0.05 | 0.06 | 0.0036 | 0.0003 | 0.0004 | 0.0036 | 0.0003 | 0.0004 | 0.0043 | 0.0841 |
| | 15 | 0.25 | 2.89 | 0.13 | 0.11 | 0.0056 | 0.0008 | 0.0008 | 0.0056 | 0.0008 | 0.0008 | 0.0071 | 0.1385 |
| | 25 | 0.42 | 3.79 | 0.30 | 0.15 | 0.0073 | 0.0018 | 0.0011 | 0.0073 | 0.0018 | 0.0011 | 0.0102 | 0.1978 |
| | 35 | 0.58 | 4.71 | 0.33 | 0.18 | 0.0091 | 0.0020 | 0.0013 | 0.0091 | 0.0020 | 0.0013 | 0.0124 | 0.2399 |
| | 75 | 1.25 | 7.33 | 0.58 | 0.26 | 0.0141 | 0.0035 | 0.0019 | 0.0141 | 0.0035 | 0.0019 | 0.0195 | 0.3784 |
| | 135 | 2.25 | 7.58 | 0.70 | 0.30 | 0.0146 | 0.0043 | 0.0021 | 0.0146 | 0.0043 | 0.0021 | 0.0210 | 0.4075 |
| | 150 | 2.50 | 7.94 | 0.72 | 0.30 | 0.0153 | 0.0044 | 0.0021 | 0.0153 | 0.0044 | 0.0021 | 0.0218 | 0.4233 |
| 65 | 0 | 0.00 | 0.00 | 0.00 | 0.00 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 |
| | 5 | 0.08 | 7.26 | 0.63 | 0.20 | 0.0140 | 0.0038 | 0.0014 | 0.0140 | 0.0038 | 0.0014 | 0.0193 | 0.3734 |
| | 15 | 0.25 | 7.83 | 0.64 | 0.20 | 0.0151 | 0.0039 | 0.0014 | 0.0151 | 0.0039 | 0.0014 | 0.0204 | 0.3958 |
| | 25 | 0.42 | 10.15 | 0.64 | 0.23 | 0.0196 | 0.0039 | 0.0016 | 0.0196 | 0.0039 | 0.0016 | 0.0251 | 0.4866 |
| | 35 | 0.58 | 10.54 | 0.67 | 0.26 | 0.0203 | 0.0041 | 0.0019 | 0.0203 | 0.0041 | 0.0019 | 0.0263 | 0.5089 |
| | 75 | 1.25 | 11.19 | 0.75 | 0.26 | 0.0216 | 0.0046 | 0.0019 | 0.0216 | 0.0046 | 0.0019 | 0.0280 | 0.5426 |
| | 135 | 2.25 | 11.38 | 0.85 | 0.28 | 0.0219 | 0.0052 | 0.0020 | 0.0219 | 0.0052 | 0.0020 | 0.0291 | 0.5643 |
| | 150 | 2.50 | 11.35 | 0.88 | 0.29 | 0.0219 | 0.0054 | 0.0021 | 0.0219 | 0.0054 | 0.0021 | 0.0293 | 0.5682 |
| 85 | 0 | 0.00 | 0.00 | 0.00 | 0.00 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 |
| | 5 | 0.08 | 12.40 | 0.50 | 0.18 | 0.0239 | 0.0031 | 0.0013 | 0.0239 | 0.0031 | 0.0013 | 0.0282 | 0.5472 |
| | 15 | 0.25 | 10.40 | 0.74 | 0.23 | 0.0200 | 0.0045 | 0.0016 | 0.0200 | 0.0045 | 0.0016 | 0.0262 | 0.5078 |
| | 25 | 0.42 | 15.37 | 0.79 | 0.24 | 0.0296 | 0.0048 | 0.0017 | 0.0296 | 0.0048 | 0.0017 | 0.0362 | 0.7007 |
| | 35 | 0.58 | 14.75 | 0.84 | 0.25 | 0.0284 | 0.0051 | 0.0018 | 0.0284 | 0.0051 | 0.0018 | 0.0353 | 0.6848 |
| | 75 | 1.25 | 14.35 | 1.02 | 0.28 | 0.0277 | 0.0062 | 0.0020 | 0.0277 | 0.0062 | 0.0020 | 0.0359 | 0.6954 |
| | 135 | 2.25 | 13.04 | 1.52 | 0.59 | 0.0251 | 0.0093 | 0.0042 | 0.0251 | 0.0093 | 0.0042 | 0.0386 | 0.7486 |
| | 150 | 2.50 | 14.96 | 1.77 | 0.72 | 0.0288 | 0.0108 | 0.0051 | 0.0288 | 0.0108 | 0.0051 | 0.0448 | 0.8678 |
| 97 | 0 | 0.00 | 0.00 | 0.00 | 0.00 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 |
| | 5 | 0.08 | 11.54 | 1.18 | 0.22 | 0.0222 | 0.0072 | 0.0016 | 0.0222 | 0.0072 | 0.0016 | 0.0310 | 0.6010 |
| | 15 | 0.25 | 12.56 | 1.22 | 0.29 | 0.0242 | 0.0074 | 0.0021 | 0.0242 | 0.0074 | 0.0021 | 0.0337 | 0.6536 |
| | 25 | 0.42 | 13.53 | 1.08 | 0.22 | 0.0261 | 0.0066 | 0.0016 | 0.0261 | 0.0066 | 0.0016 | 0.0342 | 0.6635 |
| | 35 | 0.58 | 15.62 | 0.96 | 0.18 | 0.0301 | 0.0059 | 0.0013 | 0.0301 | 0.0059 | 0.0013 | 0.0372 | 0.7218 |
| | 75 | 1.25 | 16.41 | 0.93 | 0.21 | 0.0316 | 0.0057 | 0.0015 | 0.0316 | 0.0057 | 0.0015 | 0.0388 | 0.7519 |
| | 135 | 2.25 | 16.20 | 1.50 | 0.16 | 0.0312 | 0.0092 | 0.0011 | 0.0312 | 0.0092 | 0.0011 | 0.0415 | 0.8046 |
| | 150 | 2.50 | 17.08 | 2.67 | 0.16 | 0.0329 | 0.0163 | 0.0011 | 0.0329 | 0.0163 | 0.0011 | 0.0504 | 0.9759 |

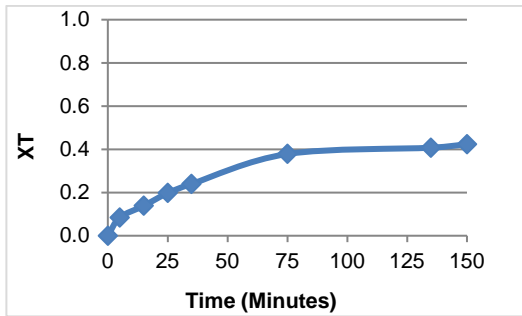


Figure 1. X_T profile against t from 50°C experiment

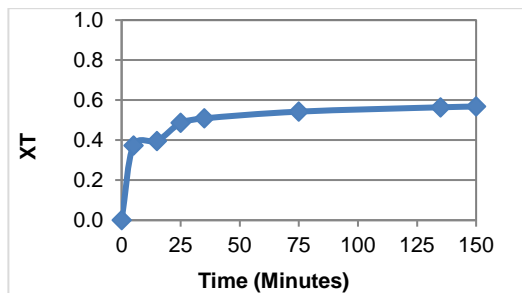


Figure 2. X_T profile against t from 65°C experiment

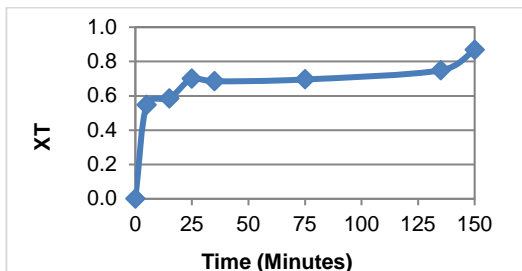


Figure 3. X_T profile against t from 85°C experiment

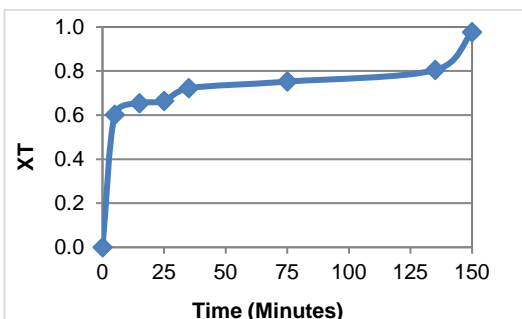


Figure 4. X_T profile against t from 97°C experiment

Figure 1 shows the total mole of the converted sulfide increased as the leaching times increased. Figure 2 to 4 show the similar tendency compared to Figure 1 but after 15 minutes of leaching, the X_T slightly declined then rose following the increase in temperature. The X_T declined at minutes of 15 because there was a probably porous solid product formation which will be examined in the next stage.

Based on the SCM, the reaction kinetics will be controlled by (Şahin and Erdem, 2015):

1. Diffusion through fluid film if the plot of X_T and t was linear
2. Diffusion through the porous solid product (ash) layer if plot of $1-3(1-X_T)^{2/3}+2(1-X_T)$ and t was linear
3. Interface reaction if the plot of $1-(1-X_T)^{1/3}$ and t was linear.

Meanwhile, based on the SPM, the reaction kinetics will be controlled by:

1. Diffusion through fluid film if the plot of $1-(1-X_T)^{2/3}$ and t was linear
2. Chemical reaction on the interface if the plot of $1-(1-X_T)^{1/3}$ and t was linear.

All of those plots, both SCM and SPM are presented at these following Figures 5 to 9:

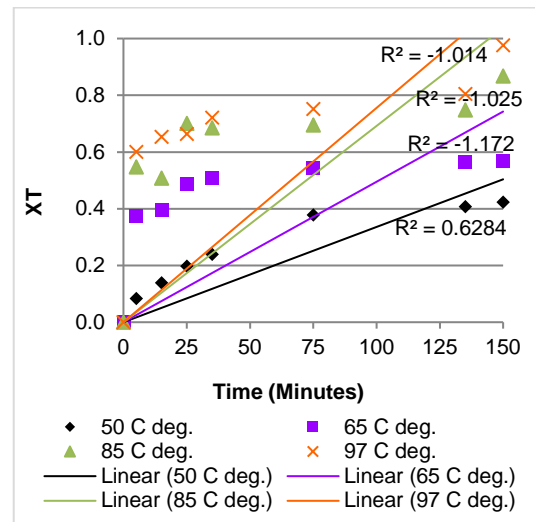


Figure 5. Plot and linear regression of X_T to t (times) for SCM

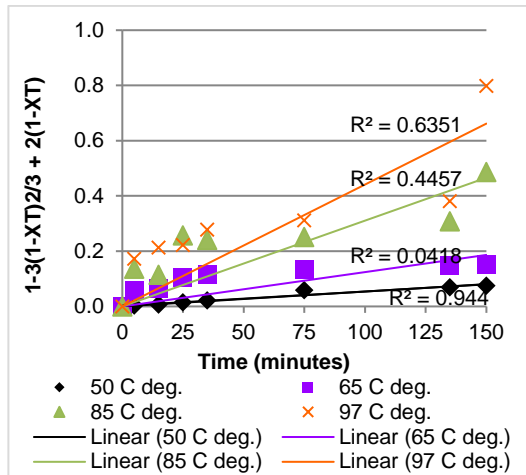


Figure 6. Plot and linear regression of $1-3(1-X_T)^{2/3} + 2(1-X_T)$ to t (times) for SCM

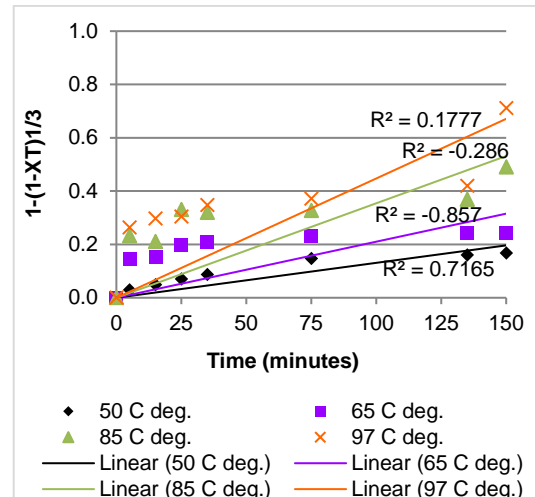


Figure 9. Plot and linear regression of $1-(1-X_T)^{1/3}$ to t (times) for SPM

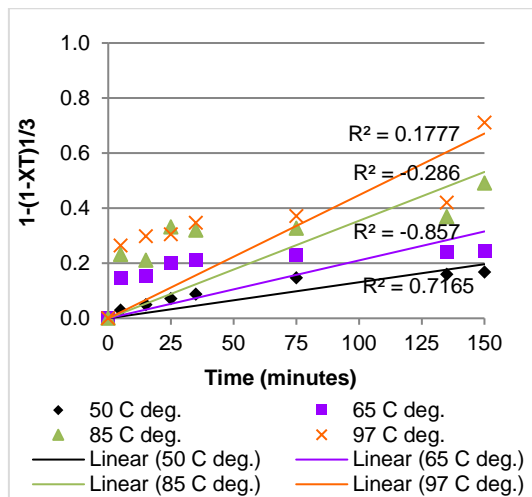


Figure 7. Plot and linear regression of $1-(1-X_T)^{1/3}$ to t (times) for SCM

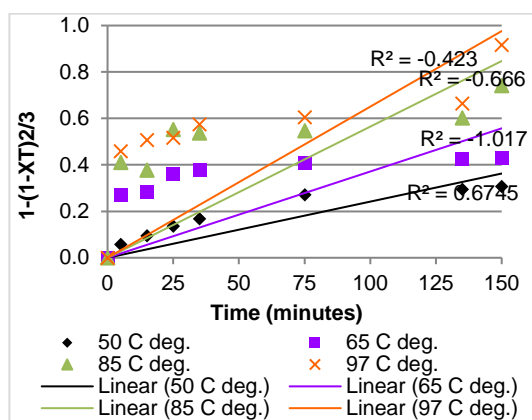


Figure 8. Plot and linear regression of $1-(1-X_T)^{2/3}$ to t (times) for SPM

A summary of R^2 values which was obtained from the linear regression equations of experimental data with all the kinetic models was used for each leach temperature and presented in Table 3.

Based on Table 3, the value of R^2 which close to 1 was obtained from the experimental data of the linear regression of SCM for porous solid layer diffusion. It concludes that the most suitable model for analyzing the leaching kinetics of galena concentrate in fluorosilicic acid solution was a SCM model with diffusion of porous solid product layer as chemical reaction controller.

The selection of reaction rate controller was also done by a method which is fitting the curve from the dummy model to the experimental data. For this purpose, the value of τ (time needed for all minerals to react completely when $X_T = 1$) needs to be calculated first. The value of τ was determined from the slopes of all linear equations following the SCM for porous solid layer diffusion as reaction controller since its R^2 was closer to 1 than other types have. $\tau = 1/\text{slope}$ was determined from linear regression of $1-3(1-X_T)^{2/3} + 2(1-X_T)$ to t . Table 4 presents the values of τ from leaching at 50, 65, 85 and 97°C.

From the values of τ , dummy curve was constructed that was fitting to the experimental data by plotting the values of X_T orderly from 0 to 1 and determined the time that was suitable for the simulated X_T . This

procedure was conducted for all the leaching temperatures. The result for fitting the dummy

curve to the experimental data is presented at Figures 10 to 13.

Table 3. Summary of R^2 values which is obtained from the linear regression equations of experimental data with all the kinetic models used for each leach temperature

| Temperature (°C) | R^2 for each kinetics models of reaction rate controller | | | | |
|---------------------|--|------------------------|--------------------|----------------------|--------------------|
| | SCM | | | SPM | |
| | Fluid film diffusion | Porous layer diffusion | Interface reaction | Fluid film diffusion | Interface reaction |
| 50 | 0.628 | 0.944 | 0.716 | 0.674 | 0.716 |
| 65 | -1.88 | 0.041 | -1.67 | -1.78 | -1.67 |
| 85 | -1.02 | 0.445 | -0.28 | -0.66 | -0.28 |
| 97 | -1.01 | 0.635 | 0.177 | -0.42 | 0.177 |

Table 4. The determination of τ from the line equations of the diffusion through porous solid layer

| Temperature (°C) | Equation | Slope | τ (minutes) | τ (hours) |
|------------------|-------------|--------|------------------|----------------|
| 50 | $y=0.0005x$ | 0.0005 | 2000.00 | 33.33 |
| 65 | $y=0.001x$ | 0.001 | 1000.00 | 16.67 |
| 85 | $y=0.003x$ | 0.003 | 333.33 | 5.56 |
| 97 | $y=0.004x$ | 0.004 | 250.00 | 4.17 |

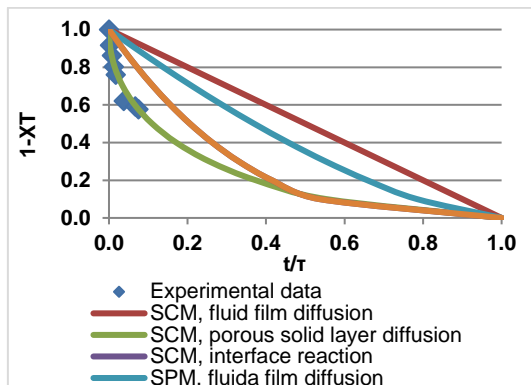


Figure 10. Fitting dummy curve model to experimental data at 50°C leaching temperature

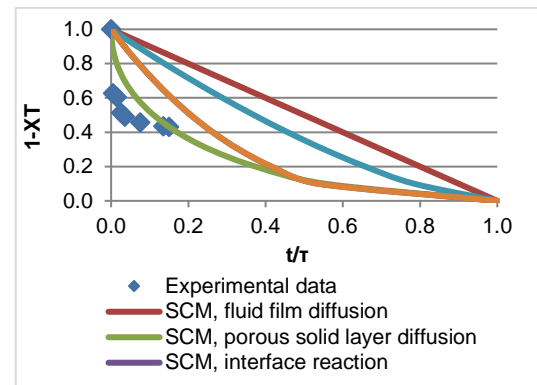


Figure 12. Fitting dummy curve model to experimental data at 85°C leaching temperature

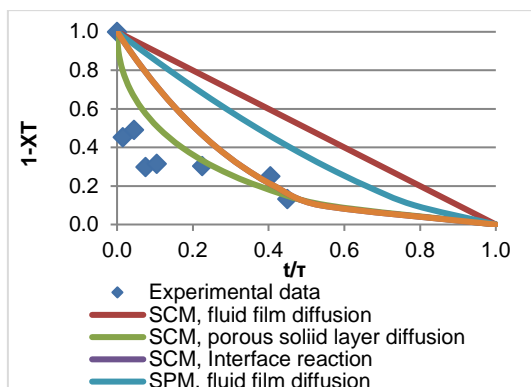


Figure 11. Fitting dummy curve model to experimental data at 65°C leaching temperature

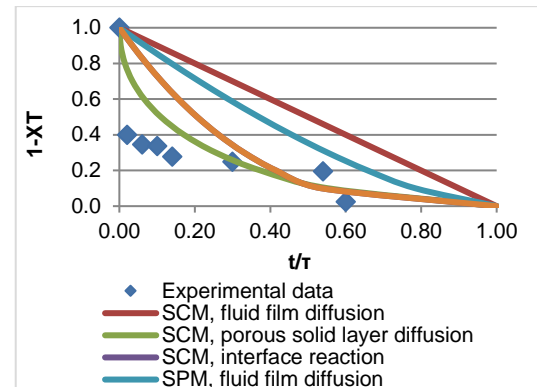


Figure 13. Fitting dummy curve model to experimental data at 97°C leaching temperature

From all the graphs in the dummy models to the experimental data fitting curves, it shows that the dummy curve of the SCM kinetics model was controlled by diffusion through a porous solid layer which was the closest to the experimental data. This curve fitting result convinces the previous conclusion which used kinetics analysis with linear regression. Again, the reaction rate of the galena concentrate leaching in fluorosilicic acid was controlled by the diffusion through the porous solid products (ash) layer.

Activation energy (E_a) is determined from the Arrhenius equation for diffusion controller through the porous solid products (ash) layer (Levenspiel, 1999):

$$D_e = D_0 \exp \left(\frac{-E_a}{RT} \right) \dots \dots \dots (14)$$

or

$$\ln D_e = - \frac{E_a}{RT} + \ln D_0 \dots \dots \dots (15)$$

By plotting $1-3(1-X_T)^{2/3}+2(1-X_T)$ to t (time), the gradient or slope (m) was determined and thus D_e was also calculated by using the following formula.

$$D_e = m \frac{\rho_T R^2}{6bC_{H_2SiF_6}} \dots \dots \dots (16)$$

Where ρ_T is total sulfide molar density, R is particle radius, $C_{H_2SiF_6}$ is fluorosilicic acid concentration and b is stoichiometric ratio of feed to fluorosilicic acid. The concentration of the fluorosilicic acid was 3.44 M or $C_{H_2SiF_6} = 3.44 \text{ mol/litre} = 3,440 \text{ mol/m}^3$. Total sulfide molar density was calculated by the following formula:

$$\rho_T = \frac{\text{total sulfide mol}}{\text{total volume}} \dots \dots \dots (17)$$

The galena specific density was 6.04 g/cm^3 ($6,040 \text{ kg/m}^3$). The kinetics experiment was performed by using 10.66 g feed so that the total sulfide mol of PbS, ZnS and FeS_2 dissolved was 0.0516 mol. These data was put into equation (17)

$$\rho_T = \frac{0.0516 \text{ mol} \times 6,040 \text{ kg/m}^3}{10,66 \text{ gram}}$$

$$\rho_T = 29,236.77 \text{ mol/m}^3$$

The feed particle size (P_{80}) was -100+150 mesh, therefore the average diameter of the particles was

$$D = \frac{(100+150)\text{mesh}}{2}$$

$$D = \frac{(0.149+0.100)\text{mm}}{2} = 0.1245 \text{ mm}$$

or the particles radius (R) was

$$R = \frac{D}{2} = \frac{0.1245 \text{ mm}}{2}$$

$$= 0.0622 \text{ mm}$$

$$R = 0.0622 \times 10^{-3} \text{ m}$$

Based on the previous experiment (Anugrah, Mubarak and Amalia, 2018), the values and the b of the converted mole fractions from the dissolution of galena concentrate's mineral sulfides, are presented in Table 5.

The next step was calculating D_e based on equation (16) from the gradient of the line equation for porous solid layer diffusion at different leaching temperatures. Then $1/T$ and $\ln D_e$ were also calculated according to Table 6.

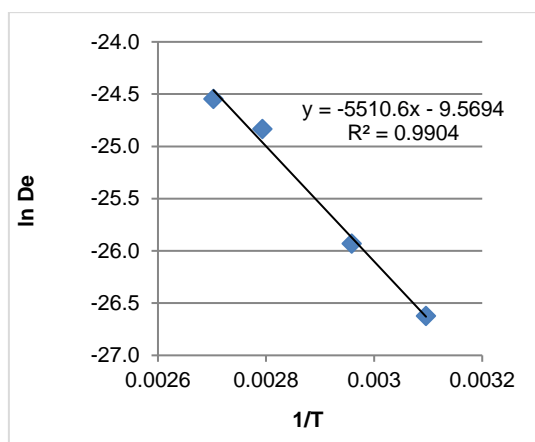
Table 5. The calculation of the sulfide minerals converted fraction and the value of b

| Sulfides | Content | Feed weight (g) | Weight (g) | Mole | Mole fraction (X) | b | bX |
|----------|-----------|-----------------|------------|----------------|-------------------|---|--------|
| PbS | Pb 66.60% | 10.66 | Pb 7.0996 | PbS 0.0343 | 0.6638 | 1 | 0.6638 |
| ZnS | Zn 7.38% | | Zn 0.7867 | ZnS 0.0120 | 0.2331 | 1 | 0.2331 |
| FeS_2 | Fe 2.79% | | Fe 0.2974 | FeS_2 0.0053 | 0.1032 | 1 | 0.1032 |
| Total | | | | 0.0516 | 1.0000 | | 1.0000 |

Table 6. In D_e and $1/T$ calculation

| Temperature | | Slope (m) | D_e (m/s) | $1/T$ | ln D_e |
|-------------|-----|-----------|-------------|----------|----------|
| °C | K | | | | |
| 50 | 323 | 0.0005 | 2.74013E-12 | 0.003096 | -26.6230 |
| 65 | 338 | 0.0010 | 5.48025E-12 | 0.002959 | -25.9299 |
| 85 | 358 | 0.0030 | 1.64408E-11 | 0.002793 | -24.8313 |
| 97 | 370 | 0.0040 | 2.1921E-11 | 0.002703 | -24.5436 |

Plotting ln D_e to $1/T$ results in a graph at Figure 14.

Figure 14. Plot of ln D_e to $1/T$

According to Figure 14, plotting ln D_e to $1/T$ resulted in a linear line equation ($R^2 = 0.99$) with a gradient of -5,510. The activation energy was then calculated by this following calculation.

$$\begin{aligned}
 \text{Activation energy (E}_a\text{)} &= \text{gradient} \times R \\
 &= 5,510 \text{ K} \times 8.3143 \\
 &\quad \text{J/K/mol} \\
 &= 45,811.79 \text{ J/mol} \\
 &= 45.81 \text{ kJ/mol} \\
 &= 10.91 \text{ kcal/mol}
 \end{aligned}$$

Activation energy of 45.81 kJ/mol for diffusion-controlled reactions through porous solid layer can be categorized as high. According to Randhawa, Gharami and Kumar (2016), for the reaction that was controlled by diffusion mechanism, E_a ranged between 1 to 3 kcal/mol and higher if it was controlled by chemical reaction. The high activation energy is was probably affected by Pb existence which undissolved because it reacted with SO_4^{2-} ion forming PbSO_4 precipitate (Zárate-Gutiérrez, Lapidus and Morales, 2012). It can also be affected by a mineral existence which has low solubility in fluorosilicic acid such as

chalcopyrite (CuFeS_2) (Zárate-Gutiérrez, Lapidus and Morales, 2010).

CONCLUSION

Leaching Indonesian galena concentrate in fluosilicic acid and hydrogen peroxide as oxidants followed the shrinking core model with diffusion through porous solid product layer as the reaction controller with activation energy of 45.81 kJ/mol or 10.91 kcal/mol.

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