Contents lists available at SciVerse ScienceDirect



Journal of Hazardous Materials



journal homepage: www.elsevier.com/locate/jhazmat

Dynamics of aluminum leaching from water purification sludge

Wen-Po Cheng^{a,*}, Chi-Hua Fu^a, Ping-Hung Chen^b, Ruey-Fang Yu^a

^a Department of Safety, Health and Environmental Engineering, National United University, Miaoli 360, Taiwan
^b Graduate Institute of Natural Resource Management, National Taipei University, San Shia District, New Taipei City 237, Taiwan

ARTICLE INFO

Article history: Received 5 October 2011 Received in revised form 10 February 2012 Accepted 3 March 2012 Available online 10 March 2012

Keywords: Aluminum Recovery Acidic Dynamic Shrinking core model

ABSTRACT

In this investigation, the shrinking core model is used to study the rate of aluminum salt leaching from water purification sludge (WPS). This model, which describes the aluminum leaching rate, can be developed to maximize the Al(III) recovering efficiency. Laboratory results indicate that when the mixing speed exceeds 80 rpm, the effect of film diffusion control on the leaching process is greatly reduced, such that any further increase in the mixing speed does not affect the Al(III) leaching rate. Additionally, increasing the temperature or acid concentration improves Al(III) leaching rate. The laboratory data were verified by using the shrinking core model to confirm that the leaching of Al(III) from WPS is consistent with the inert-layer diffusion control model. This finding reveals that large amounts of SiO₂, Al₂O₃ and other inert constituents will form an inter diffusion layer in the WPS and thus become the major limiting factors that control the Al(III) leaching process. The dynamic equation can be expressed as $1 - 3(1 - x)^{2/3} + 2(1 - x) = (2707.3 \exp(-3887.6/T))t$, in which the apparent activation energy and pre-exponential factors are 32.32 kJ/mol and $2707.3 \min^{-1}$, respectively, as determined by solving the Arrhenius equation.

Crown Copyright © 2012 Published by Elsevier B.V. All rights reserved.

1. Introduction

WPS (water purification sludge) has a similar composition to that of soil. Its main constituents are SiO₂ and Al₂O₃ [1] followed by the aluminum hydroxide precipitates that are formed upon the addition of alum coagulant during the water purification process. At normal temperature, acid cannot dissolve SiO₂, but the conversion of Al₂O₃ into dissolved aluminum ions depends on high temperature and excess acid [2,3]. In Taiwan, surface water is the major source of water for use by the population, and commonly using aluminum sulfate or polyaluminum chloride (PACl) as coagulants. Water-borne suspended particles, colloids and some soluble species will be reacted or adsorbed by aluminum hydroxide to form coagulation flocs [4]. The flocs are then removed in a subsequent sedimentation operation in the form of aluminum hydroxide-containing sludge, in which the aluminum hydroxide is present as amphoteric characteristics; and is highly soluble in both acidic and basic solutions [5]. Therefore, simply adjusting the solution pH achieves two purposes, which are (1) reduction of the amount of sludge, facilitating subsequent treatment and disposal [6,7], and (2) recovery of the coagulants under either acidic or basic conditions for reuse [6–11]. The results of Panswad and Chamnan [12] indicate that when sulfuric acid is used to extract aluminum from sludge, around 70–90% of the aluminum can be recovered by appropriately setting the solution pH between 1 and 3. A lower pH results in the dissolution of more aluminum and therefore a higher aluminum recovery ratio. If the aluminum is recovered under basic conditions by adding NaOH or Ca(OH)₂, then the recovery efficiency is maximized in the pH range of 11.2–11.8 [13]. However, under alkaline conditions, some natural organic substances are also easily dissolved, causing problems in the subsequent aluminum recovery operations. Therefore, acidifying the sludge with sulfuric acid is the current method for leaching and recovering the Al(III) ions from WPS [14]. The major chemical reactions, which occur in both the liquid and the solid phase, are expressed by the following equation:

$$2Al(OH)_{3}(s) + 3H_{2}SO_{4}(aq) \rightarrow Al_{2}(SO_{4})_{3}(aq) + 6H_{2}O$$
(1)

Decades of research have been devoted to determine the optimal conditions for acidifying the sludge. However, most of this research effort has been directed toward the amphoteric characteristics of aluminum salts, emphasizing the adjustment of sludge pH values. The mechanism of aluminum leaching from WPS has not been fully elucidated, and so the lack of a theoretical basis for the aluminum recovering process.

The Al(III) ions were formed by adding acid ions to dissolve $Al(OH)_3$ from the WPS and then the dissolved Al(III) ions were removed from the WPS by a dispersion mechanism. Therefore, the reaction rate was controlled by the speed of acid ion flow, the dispersion of the Al(III) ions and the reaction mechanism, and therefore the shrinking core model can be used to study the dynamics of

^{*} Corresponding author. Tel.: +886 3 7381764, fax: +886 3 7333187. *E-mail address:* cwp@nuu.edu.tw (W.-P. Cheng).

^{0304-3894/\$ –} see front matter. Crown Copyright © 2012 Published by Elsevier B.V. All rights reserved. doi:10.1016/j.jhazmat.2012.03.007

aluminum that leaches from WPS, to provide understanding of the leaching process. The shrinking core model comprises the following three reaction mechanisms [15,16].

a. Film diffusion control

$$1 - (1 - x) = \frac{t}{t_{\rm f}}$$
(2)

b. Inert-layer diffusion control

$$1 - 3(1 - x)^{2/3} + 2(1 - x) = \frac{t}{t_i}$$
(3)

c. Surface chemical reaction control

$$1 - (1 - x)^{1/3} = \frac{t}{t_c} \tag{4}$$

In Eqs. (2)–(4), x denotes the aluminum leaching rate, and t is the reaction time. The time for complete dissolution by liquid film control (t_f) , the time for complete dissolution by inert-layer diffusion control (t_i) and time for complete dissolution by surface chemical reaction control (t_c) are given by the following equation.

$$t_{\rm f} = \frac{\rho_{\rm p} R_{\rm S}}{3b M_{\rm P} K_g C_{\rm a}} \tag{5}$$

$$t_{\rm i} = \frac{\rho_{\rm p} R_{\rm S}^2}{6b M_{\rm P} D_{\rm c} C_{\rm a}} \tag{6}$$

$$t_{\rm c} = \frac{\rho_{\rm p} R_{\rm S}}{b M_{\rm P} K_{\rm g} C_{\rm a}} \tag{7}$$

where ρ_p is density of the solid reactant; R_s is the initial radius of the particle; *b* is the molar ratio of solid reactants; M_p is the molecular weight of solid reactant (g/mol); K_g is the mass transfer factor; C_a is the concentration of sulfuric acid; and D_c is the inert-layer diffusion coefficient.

When the sludge is mixed at high speed, the solid–liquid interface is completely mixed, minimizing the thickness of the interface; the resistance of alum to dissolve that is caused by the transportation of liquid (Eq. (2)) can then be ignored [17,18]. Therefore, the leaching rate is determined by only the rate of inert-layer diffusion or the rate of the chemical reaction. Under such conditions, the shrinking core model can be expressed as the combination of Eqs. (3) and (4), as in Eq. (8) [19]:

$$t = \left[1 - (1 - x)^{1/3}\right] t_{c} + \left[1 - 3(1 - x)^{2/3} + 2(1 - x)\right] t_{i}$$
(8)

According to research from Olanipekun [20], Baba and Adekola [21], if the aluminum leaching process is dominated by inert-layer diffusion control mechanism, the chemical reaction rate is fast; then, t_c is near zero. Hence, Eq. (8) can be simplified and manipulated as following kinetic equation:

$$\left[1 - 3(1 - x)^{2/3} + 2(1 - x)\right] = \frac{t}{t_i} = K_i t$$
(9)

 K_i is the reaction rate constant of inert-layer diffusion control (min⁻¹).

Similarly, if the leaching reaction is dominated by the chemical control mechanism, the value of inert-layer diffusion coefficient (D_c) is high; then, Eq. (8) can be simplified and manipulated as following kinetic equation:

$$\left[1 - (1 - x)^{1/3}\right] = \frac{t}{t_c} = K_c t \tag{10}$$

 K_c is the reaction rate constant of the surface chemical reaction control (min⁻¹).

The recovery and reuse of resources from WPS has been a major field research. However, the key to the successful reuse of WPS is the development of a cost-efficient method for dissolving the metal



Fig. 1. EDS elemental composition analysis.

from WPS. In this investigation, sulfuric acid is added to acidify the sludge to leach out the aluminum ions, with a view to elucidating the leaching kinetics by varying some control parameters, including mixing intensity, concentration of sulfuric acid applied, temperature and reaction time [3,17]. Laboratory results are obtained to calibrate the model so that an accurate kinetic equation can be developed to elucidate the correlation between reaction rate and important parameters, including sludge characteristics, temperature and concentration. Simulation studies can then be performed using the leaching model to optimize the aluminum ion leaching efficiency.

2. Experimental

2.1. Materials

WPS samples were collected from the sludge drying bed of Ming-Der Water Treatment Plant (Miaoli, Taiwan), dried at 105 °C. After drying and grinding the sludge, the sludge particles were separated by sieving into 12–32 mesh for use in the laboratory. The median value of the theoretical particle diameter is 950 μ m.

The prepared sludge sample was initially observed under a scanning electron microscope (SCM, JSM5600, JEOL Co, Japan) with an energy dispersive spectrometer (EDS, Oxford Link ISIS, JEOL Co, Japan) to identify the constituent elements. As shown in Fig. 1, Si, Al, and Fe are the main elements in the WPS sludge, followed by K and Mg. The crystalline structure of the sludge was further observed using an X-ray diffraction (XRD, D/Max 2550PC, Rigaku Co. Japan). The results in Fig. 2 indicate that Si and Al are in the form of SiO₂ and Al₂O₃, respectively; they are derived mostly from the sand,



Fig. 2. Results of XRD analyses of prepared WPS samples.



Fig. 3. Experimental setup for mixing.

clay and silt particles that were originally present in the raw water. Additionally, the sludge contains a small amount of Fe_2O_3 .

The results of chemical analyses indicate that aluminum is a major constituent of the WPS sample. Al_2O_3 cannot react with acid at normal temperature; only the solid-phase amorphous $Al(OH)_3$ that is in the sludge precipitate can be leached out by acidifying the sludge. Therefore, the total quantity of aluminum that can leach out in the acidifying process was determined by the standard method that is promulgated by the Taiwan Environmental Protection Administration (NIEA R353.00C). The WPS sample was dissolved in nitric acid (1:1) and the resulting solution was analyzed using atomic absorption spectrometry (Z-5000 Hitachi Co. Japan) to determine the total quantity of aluminum, or Al(III), that can leach out of the sludge under acidic conditions. The result of repeated tests indicate that around 40.03 mg aluminum ions can leach from 1 g WPS, and the Al(III) leaching ratio (x) can be expressed as:

$$x = \frac{C}{C_0} \tag{11}$$

where C_0 denotes the total quantity of Al(III) that can be leached out by the acid washing process (40.03 mg/g), and *C* is the amount quantity of Al(III) that leaches out under various conditions (mg/g).

2.2. WPS leaching procedure

Fig. 3 presents the experimental setup. A 1000 mL reactor that was equipped with a variable-speed mixer and submerged in a constant-temperature water bath was used. Acidic leaching of Al(III) from the WPS sample was performed in the reactor using sulfuric acid solutions at various concentrations, mixing intensities and temperatures. After the leaching study was complete, the solution was filtered, and the filtrate was subjected to atomic absorption analyses (HITACHI Z-5000 Flame Atomic Absorption Spectrophotometer) to determine the Al(III) concentration. Each experiment was repeated two times and represented with average values.

2.2.1. Effect of mixing intensities on leaching rate

All studies were performed at a constant temperature of $25 \,^{\circ}$ C. A mixture of $30 \,\text{g}$ of the prepared WPS sample and $500 \,\text{mL}$ of $0.5 \,\text{N}$ sulfuric acid was placed in the 1000 mL reactor. Six sets of above solutions were prepared. These solutions were individually mixed with paddles at 20, 40, 60, 80, 100 and 120 rpm for 20 min. The samples were taken at the end of time for each stirring speed.



Fig. 4. Effect of stirring intensity on alum leaching rate.

2.2.2. Effect of sulfuric acid concentrations on leaching rate

The effect of sulfuric acid concentration on the Al(III) leaching rate was studied using a set of samples that were prepared by adding 500 mL of 1, 0.7, 0.5, or 0.3 N sulfuric acid to 30 g WPS samples. The leaching study was performed at $25 \,^{\circ}$ C with the paddle speed set to 100 rpm. Samples were collected at regular intervals for analysis to determine the amount of soluble Al(III).

2.2.3. Effect of temperature on leaching rate

The leaching studies involved adding 30 g WPS samples to 500 mL of 0.5 N sulfuric acid solution. Experiments were performed at temperature of 10 °C, 40 °C, and 70 °C with the paddle speed maintained at 100 rpm and 60 min leaching time. Samples were collected at regular intervals to determine the amount of soluble Al(III). Due to the fix particle diameter requirement for using shrinking core model, an image analysis for observing sludge particle diameter change during the leaching process was conducted at the leaching temperature of 40 °C. In this experiment, samples were taken by a pipet for a captured image analysis of the particle diameter [22,23]. The average diameters were found to be 707, 683, 725 and 731 μ m at 1, 20, 40 and 60 min. These results proved that the sludge particle diameters did not significantly change.

3. Results and discussion

3.1. Influence of mixing intensity on leaching of Al(III)

The effect of the stirring speed (20–120 rpm) on Al extraction efficiency can be observed in Fig. 4. As presented in Fig. 4, the leaching ratio of Al(III) increases with the mixing speed below 80 rpm; beyond 80 rpm, the leaching ratio does not increase with mixing speed. This phenomenon reveals that when the mixing speed reaches a threshold, the thickness of the diffusion layer can no longer be effectively reduced. Therefore, the film diffusion control, described by Eq. (2) does not affect the Al(III) leaching rate, so the mixing speed exceeds 80 rpm, the leaching process is influenced only by the chemical reaction (Eq. (4)), the inert-layer diffusion (Eq. (3)), or the combination of both (Eq. (8)).

3.2. Influence of sulfuric acid concentration on leaching process

The Al(III) leaching ratios at various reaction times and a paddle speed of 100 rpm for various sulfuric acid concentrations are presented in Fig. 5. At a particular leaching time, the Al(III) leaching ratio increases with sulfuric acid concentration, and at a fixed



Fig. 5. Al(III) leaching ratio at various times for different concentrations of sulfuric acid.

sulfuric acid concentration, it increases reaction time. However, when the reaction time exceeds 20 min, the leaching reaction reaches a steady state that is not influenced by sulfuric concentration. Thereafter, very little change in the Al(III) leaching ratio is observed. Hence, the Al(III) leaching rate data for a reaction time of less than 20 min will be used to examine the kinetics of Al(III) leaching.

3.2.1. Evaluating Al(III) leaching using surface chemical reaction control model

The surface chemical reaction control model in Eq. (10) is utilized to evaluate the Al(III) leaching process. A linear relationship between $1 - (1 - x)^{1/3}$ and *t* reveals that the surface reaction mechanism controls the Al(III) leaching process, and the slope of the linear plot is the reaction rate constant K_c . In Fig. 6, the plots of $1 - (1 - x)^{1/3}$ against reaction time *t* (min) do not demonstrate a strong linear relationship for all four sulfuric acid concentrations, indicating that the leaching process does not proceed by the surface reaction mechanism. Restated, the acidic ions react rapidly with Al(OH)₃ but do not necessarily cause the Al(III) to leach out of the WPS sludge.



Fig. 6. Relationship between $1 - (1 - x)^{1/3}$ and leaching time for different sulfuric acid concentrations.



Fig. 7. Linear relationship between $1 - 3(1 - x)^{2/3} + 2(1 - x)$ and leaching time different sulfuric acid concentrations.

3.2.2. Evaluating Al(III) leaching using inert-layer diffusion control mechanism

The leaching process is evaluated using the inert-layer diffusion control mechanism, given by Eq. (9). The results in Fig. 7 reveal a linear relationship (slope is the reaction rate constant K_i) between $1 - 3(1 - x)^{2/3} + 2(1 - x)$ and reaction time *t* for all four acid concentrations, indicating that the Al(III) leaching reaction is consistent with the inner-layer diffusion control model.

The main constituents in WPS are SiO₂ and Al₂O₃, which do not react with sulfuric acid at pH values of the solution herein. As shown in Fig. 8, when the Al(OH)₃ species on the outmost layer of the sludge reacts with H⁺ to produce Al(OH)_mⁿ⁺ ions, which leach out of the sludge, the core of the Al(OH)₃-containing sludge gradually decreases while the SiO₂ and Al₂O₃ combine with other inert constituents to form an inert diffusion layer [20,21,24]. This layer gradually becomes thicker as the reaction time increases (Fig. 8(b)), forming a barrier, through which external H⁺ ions must diffuse before they react with the Al(OH)₃ species in the core of the sludge. Likewise, the resulting Al(OH)_mⁿ⁺ ions must also pass through this layer to diffuse into the bulk of the solution. Therefore, this mechanism is the basis of the reactions that are involved in the inert-layer diffusion control model.

3.3. Effect of temperature on leaching process

The extent of the leaching of Al(III) ions at various reaction times for various solution temperatures was presented in Fig. 9. At a particular reaction time, the Al(III) leaching rate increases with temperature. At a particular reaction temperature, the Al(III) leaching ratio increases with time, but the time for the leaching process to reach steady state varies differently. When the temperatures are 70, 40, and $10 \,^{\circ}$ C, the times to reach the steady state are 12 min, 30 min and 40 min, respectively. The laboratory data are further analyzed using the inert-layer diffusion control model (Eq. (9)) and the surface chemical reaction control model (Eq. (10)) to evaluate the various parameters that control the Al(III) leaching process.

A simulation using the surface chemical reaction control model (Eq. (10)) yields a non-linear relationship between $1 - (1 - x)^{1/3}$ and t (Fig. 10). In contrast, a simulation using the inert-layer diffusion control model (Eq. (9)) to simulate the results yields a strong linear relationship between $1 - 3(1 - x)^{2/3} + 2(1 - x)$ and t at the various temperatures of interest (Fig. 11). Therefore, the results obtained using various sulfuric acid concentrations and temperatures indicate that the inert-layer diffusion control model is more suitable for simulating the leaching of Al(III) from WPS.



Fig. 8. Acidic leaching from WPS: (a) sludge surface in initial leaching period, and (b) the core of the Al(OH)₃-containing sludge reduction and diffusion of Al(III) ions out of sludge become not easy.



Fig. 9. Al(III) leaching ratio at various times for various temperatures.



Fig. 10. The relationship between $1 - (1 - x)^{1/3}$ and leaching time, *t*, for various temperatures.



Fig. 11. The relationship between $1 - 3(1 - x)^{2/3} + 2(1 - x)$ and leaching time, *t*, for different temperatures.

If the Arrhenius equation [25] is used to describe the kinetics of Al(III) leaching, the relationship equation can be expressed as:

$$K = A e^{-E_{a}/RT} \tag{12}$$

Eq. (13) is obtained by taking the logarithm of Eq. (12):

$$\ln K = \ln A - \frac{E_a}{RT} \tag{13}$$

where A is the pre-exponential factor; E_a is the apparent activation energy in J/mol; *R* is the universal gas constant in 8.314 K⁻¹ mol⁻¹, and *T* is the absolute temperature in K. Table 1 presents the leaching reaction rate constants obtained at the various temperatures from the slope of the linear plot in Figs. 10 and 11. According to Table 1,

Table 1

Temperature	Apparent rate constants (min ⁻¹)		Correlation coefficient (R^2)	
	Ki	Kc	Ki	Kc
10°C	0.0336	0.0456	0.9924	0.9233
40 ° C	0.0101	0.0143	0.9789	0.7323
70°C	0.0029	0.0064	0.9145	0.5769



Fig. 12. Linear Arrhenius plot of $\ln K$ against T^{-1} for leaching of alum from WPS.

the correlation coefficient value (R^2) of K_i is higher than the value of K_c . Therefore, in this study, the K value of Arrhenius formula represents the K_i value.

The $\ln K_i$ values in Table 1 are plotted as a function of T^{-1} for regression analyses. As shown in Fig. 12, the apparent activation energy and pre-exponential factor in the Arrhenius equation are estimated to be $E_a = 32.32 \text{ kJ/mol}$ and $A = 2707.3 \text{ min}^{-1}$, respectively. The activation energy (32.32 kJ/mol) compared with that obtained by Abdel-Aal [26], who used the shrinking core model to study the leaching behavior of nickel oxide in sulfuric acid solution and indicated that the activation energy based on the diffusion control model is between 1 and 6 kcal/mol (4.18-25.10 kJ/mol). In contrast, the activation energy for a chemically controlled process is usually greater than 10 kcal/mol (41.84 kJ/mol) [26-28]. The active energy obtained from this study (32.32 kI/mol) is higher than the value of diffusion control reaction, but is lower than the value of surface chemical reaction control. Perhaps both inert-layer diffusion control and surface reaction control might simultaneously affect the leaching dynamics of the reaction. Hence, the mixed-controlled model is discussed below.

The mixed-controlled model is developed by manipulating Eqs. (8)–(10) to Eq. (14). Where both inert-layer diffusion control and surface chemical reaction control are included in the equation:

$$K_{\rm m}t = \left[1 - 3(1-x)^{2/3} + 2(1-x)\right] + B\left[1 - (1-x)^{1/3}\right]$$
(14)

where $B = K_i/K_c$ and K_m is the rate constant of the mixed-controlled model. Recently, some studies successfully applied the Eq. (14) to describe the leaching of metal ion in acid solution [29–31].

In Eq. (14), the *B* value is set between 0 and 1. Different *B* and respective experimental data are plugged into Eq. (14) to calculate the outcome data. The outcome data are then plotted for finding best regression coefficient. When *B* value is small, the reaction dynamics is dominated by the inert layer diffusion control mechanism. In contrast, if *B* is high, the surface chemical reaction control dominates the reaction. According to Figs. 13–15, under selected reaction temperature (10, 40 and 70 °C), when *B* value is increased, the correlation coefficient (R^2) of the regression line starts to decrease. This result indicates that the surface chemical control mechanism is not the key factor in an aluminum leaching process. Therefore, the experiment to determine the activation energy of the reaction confirms that the leaching of Al(III) from WPS is controlled by inert-layer diffusion.



Fig. 13. A diagram of the equation $1-3(1-x)^{2/3}+2(1-x)+B(1-(1-x)^{1/3})$ as a function of time in a leaching experiment at 10 °C, when *B* value is between 0 and 1.



Fig. 14. A diagram of the equation $1 - 3(1-x)^{2/3} + 2(1-x) + B(1-(1-x)^{1/3})$ as a function of time in a leaching experiment at 40 °C, when *B* value is between 0 and 1.



Fig. 15. A diagram of the equation $1 - 3(1-x)^{2/3} + 2(1-x) + B(1-(1-x)^{1/3})$ as a function of time in a leaching experiment at 70 °C, when *B* value is between 0 and 1.

Accordingly, the leaching of Al(III) from WPS is simply controlled by inert-layer diffusion. Eq. (8) can thus be re-written as the following inert-layer diffusion equation:

$$K_{i}t = 1 - 3(1 - x)^{2/3} + 2(1 - x)$$
(15)

Substituting the activation energy data and other parameters into Eq. (12) yields Ki and Eq. (15) can then be rewritten as:

$$1 - 3(1 - x)^{2/3} + 2(1 - x) = \left(2707.3 \exp\left(-\frac{3887.6}{T}\right)\right)t$$
(16)

Under certain temperature conditions, Eq. (16) can be used to predict the relationship between aluminum recovery rate (x) and reaction time (t) in the process of aluminum leaching from water treatment sludge.

4. Conclusions

Sulfuric acid is utilized herein investigation to study the kinetics of the leaching of Al(III) from water purification sludge. An experiment was performed in the laboratory in which the mixing intensity, sulfuric concentration, temperature and reaction time varied; the simulation was performed using the shrinking core model to obtain kinetic equations and the values of the various kinetic parameters. The following conclusions are drawn concerning the leaching of Al(III) from WPS, based on laboratory results and theoretical considerations.

The Al(III) leaching rate increases with mixing speed. However, when the mixing speed exceeds 80 rpm, the leaching rate will not further be enhanced indicating that the film diffusion control will not affect the Al(III) leaching rate at higher mixing speed.

The Al(III) leaching rate (x) increases with sulfuric acid concentration for a particular reaction time; the leaching rate also increases with the reaction time for a fixed sulfuric acid concentration. However, when the reaction time increases above 20 min, the Al(III) leaching rate remains relatively constant. The laboratory data reveal a highly linear relationship between $1 - 3(1 - x)^{2/3} + 2(1 - x)$ and the reaction time, revealing that the leaching mechanism is consistent with the inert-layer diffusion control model. The SiO₂ and Al₂O₃ contents in the sludge are considered to react with other inert constituents, to form an inert diffusion layer that becomes the main factor that controls the leashing reaction.

Under the conditions of the laboratory studies, the Al(III) leaching rate increased with temperature, and with reaction time at a constant temperature. Additionally, the time to complete leaching is shorter at higher temperature. The apparent activation energy of the leaching reaction is calculated to be 32.32 kJ/mol by substituting the various reaction rate constants obtained at various temperatures into the Arrhenius equation. Through a mixed-controlled model, experimental data also proved that the leaching of Al(III) from WPS is controlled by inert-layer diffusion. This value of activation energy is consistent with inert-layer diffusion control; it is used to obtain the kinetic equation $1 - 3(1 - x)^{2/3} + 2(1 - x) =$ (2707.3 exp(-3887.6/T))t.

Acknowledgment

The authors acknowledge the financial support of National Science Council, Taiwan, ROC for this work (NSC-98-2221-E-239-006-MY3).

References

- C.H. Wu, Regeneration and reuse of water treatment plant sludge: adsorbent for cations, J. Environ. Sci. Health A 39 (2004) 717–728.
- [2] R.E. Treybal, Mass Transfer Operations, 3rd ed., McGraw-Hill, New York, 1980.
 [3] L. Bao, A.V. Nguyen, Developing a physically consistent model for gibbsite leaching kinetics, Hydrometallurgy 104 (2010) 86–98.
- [4] R.A.R. Boaventura, A.S.D. António, M.F. Almeida, Aluminum recovery from water treatment sludges, in: IV International Conference, Water Supply and Water Quality, Kraków–Poland, September 11–13, 2000, pp. 1–4.
- [5] G. Sposito, The Environmental Chemistry of Aluminum, 2nd ed., CRC Press, Boca Raton, Florida, 1996.
- [6] M.M. Bishop, A.T. R olan, T.L. Bailey, D.A. Cornwell, Testing of alum recovery for solids reduction and reuse, J. Am. Water Works Assoc. 79 (1987) 76–83.
- [7] S.H. Huang, J.L. Chen, K.Y. Chiang, C.C. Wu, Effects of acidification on dewaterability and aluminum concentration of slum sludge, Sep. Sci. Technol. 45 (2010) 1165–1169.
- [8] C.W. Li, J.L. Lin, S.F. Kang, C.L. Liang, Acidification and alkalization of textile chemical sludge, Sep. Purif. Technol. 42 (2005) 31–37.
- [9] B. Jimenez, M. Martinez, M. Vaca, Alum recovery and wastewater sludge stabilization with sulfuric acid, Water Sci. Technol. 56 (2007) 133–141.
- [10] G.R. Xu, Z.C. Yan, Y.C. Wang, N. Wang, Recycle of alum recovered from water treatment sludge in chemically enhanced primary treatment, J. Hazard. Mater. 161 (2009) 663–669.
- [11] S. Petzet, B. Peplinski, S.Y. Bodkhe, P. Cornel, Recovery of phosphorus and aluminium from sewage sludge ash by a new wet chemical elution process (SESAL-Phos-recovery process), Water Sci. Technol. 64 (3) (2011) 693–699.
- [12] T. Panswad, P. Chamnan, Aluminum recovery from industrial aluminum sludge, J. Water Supply Res. Tech.-Aqua 10 (4) (1992) 159–167.
- [13] W.J. Masschelein, R. Devleminck, J. Genot, The feasibility of coagulant recycling by alkaline reaction of aluminum hydroxide sludge, Water Res. 19 (1985) 1363–1368.
- [14] S. Ishikawa, N. Ueda, Y. Okumura, Y. Iida, K. Baba, Recovery of coagulant from water supply plant sludge and its effect on clarification, J. Mater. Cycles Waste Manage. 9 (2) (2007) 167–172.
- [15] J.Y. Park, O. Levenspiel, The crackling core model for the reaction of solid particles, Chem. Eng. Sci. 30 (1975) 1207–1214.
- [16] O. Levenspiel, Chemical Reaction Engineering, 3rd ed., Wiley, New York, 1999.
- [17] B.R. Reddy, S.K. Mishra, G.N. Banerjee, Kinetics of leaching of a gibbsitic bauxite with hydrochloric acid, Hydrometallurgy 51 (1999) 131–138.
- [18] Y.B. Xu, Z.Y. Luo, Q.H. Wang, J.S. Zhou, Z.L. Shi, Leaching kinetics of vanadium pentoxide from ash of stone coal by sulfuric acid, Chin. J. Process Eng. 10 (2010) 60–64.
- [19] I.H. Lee, Y.J. Wang, J.M. Chern, Extraction kinetics of heavy metal-containing sludge, J. Hazard. Mater. 123 (2005) 112–119.
- [20] E.O. Olanipekun, Kinetics of leaching laterite, Int. J. Miner. Process. 60 (2000) 9–14.
- [21] A.A. Baba, F.A. Adekola, Hydrometallurgical processing of a Nigerian sphalerite in hydrochloric acid: characterization and dissolution kinetics, Hydrometallurgy 101 (2010) 69–75.
- [22] W.P. Cheng, Y.J. Hsieh, R.F. Yu, Y.W. Huang, S.Y. Wu, Characterizing polyaluminum chloride (PACI) coagulation floc using an on-line continuous turbidity monitoring system, J. Taiwan Inst. Chem. Eng. 41 (2010) 547–552.
- [23] W.P. Cheng, Y.J. Hsieh, P.H. Chen, R.F. Yu, Y.W. Huang, Comparing floc strength using a turbidimeter, Int. J. Miner. Process. 100 (2011) 142–148.
- [24] V. Safari, G. Arzpeyma, F. Rashchi, N. Mostoufi, A shrinking particle-shrinking core model for leaching of a zinc ore containing silica, Int. J. Miner. Process. 93 (1) (2009) 79–83.
- [25] P.W. Atkins, Physical Chemistry, 4th ed., Oxford University Press, Oxford, 1990.
 [26] E.A. Abdel-Aal, M.M. Rashad, Kinetic study on the leaching of spent nickel oxide
- catalyst with sulfuric acid, Hydrometallurgy 74 (2004) 189–194.
- [27] M.E. Wadsworth, Advances in the leaching of sulphide minerals, Miner. Sci. Eng. 4 (4) (1972) 36–47.
- [28] S. Aydogan, A. Aras, M. Canbazoglu, Dissolution kinetics of sphalerite in acidic ferric chloride leaching, Chem. Eng. J. 114 (2005) 67–72.
- [29] R. Dehghan, M. Noaparast, M. Kolahdoozan, Leaching and kinetic modelling of low-grade calcareous sphalerite in acidic ferric chloride solution, Hydrometallurgy 96 (2009) 275–282.
- [30] A.A. Baba, A.F. Adekola, R.B. Bale, Development of a combined pyro- and hydrometallurgical route to treat spent zinc-carbon batteries, J. Hazard. Mater. 171 (2009) 838–844.
- [31] O.S. Ayanda, F.A. Adekola, A.A. Baba, O.S. Fatoki, B.J. Ximba, Comparative study of the kinetics of dissolution of laterite in some acidic media, J. Miner. Mater. Charact. Eng. 10 (2011) 1457–1472.