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# Evaluation of shrinking core model in leaching process of Pomalaa nickel laterite using citric acid as leachant at atmospheric conditions

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**Abstract.** Most of kinetics studies related to leaching process used shrinking core model to describe physical phenomena of the process. Generally, the model was developed in connection with transport and/or reaction of reactant components. In this study, commonly used internal diffusion controlled shrinking core model was evaluated for leaching process of Pomalaa nickel laterite using citric acid as leachant. Particle size was varied at 60-70, 100-120, -200 meshes, while the operating temperature was kept constant at 358 K, citric acid concentration at 0.1 M, pulp density at 20% w/v and the leaching time was for 120 minutes. Simulation results showed that the shrinking core model was inadequate to closely approach the experimental data. Meanwhile, the experimental data indicated that the leaching process was determined by the mobility of product molecules in the ash layer pores. In case of leaching resulting large product molecules, a mathematical model involving steps of reaction and product diffusion might be appropriate to develop.

## 1. Introduction

In this era, the use of nickel laterite in mineral processing has to be done, though the nickel content in laterite ore is small. It needs to be done as a result of depletion of nickel sulphide resources in the world [1-5]. During this time, smelting method dominates in mineral processing which the ore has low nickel content [2, 3]. However, high energy consumption and high operational cost in smelting process make the other process, like leaching process at atmospheric condition should be done [6,7]. The utilization of this process is expected to be useful and applicable in industry.

One of the important factor in the application of leaching process at atmospheric conditions in industrial scale is information about process mechanism, mathematical model, and constants' value related on that process. This information will be useful in scale-up process. In the study about leaching process, shrinking core model is the most popular model among researchers. Several studies, like the studies conducted by Astuti, et.al., Thubakgale et.al., Agacayak and Zedef, used this model and were able to show that shrinking core model were the most suitable to describe the physical phenomenon of leaching process [7-9].

Shrinking core model was introduced at the first time by Yagi and Kunii (1955) and is described completely in the text book of Chemical Reaction Engineering written by Octave Levenspiel. This model describes that the reaction occurs first at the outer skin of particle and then the reaction zone moves into the deeper unreacted zone. The zone where reaction has occurred will become ash layer and the layer is assumed to be inert [10].

Leaching process of nickel laterite is a heterogeneous process which involves more than one phase i.e. the solid-liquid phase. Thus, this process will be controlled by several steps, such as diffusion through liquid film layer, diffusion through ash layer, and chemical reaction. The overall rate of the



process is determined by the slowest step [7]. Types of the shrinking core models in leaching process can be summarized in the following equations [7,10] :

$$\text{Diffusion through liquid film layer control} : k_f \cdot t = x \quad (1)$$

$$\text{Diffusion through ash layer control} : k_d \cdot t = 1 - 3(1-x)^{0.67} + 2(1-x) \quad (2)$$

$$\text{Chemical reaction control} : k_r \cdot t = 1 - (1-x)^{0.33} \quad (3)$$

where  $x$  is the reacted fraction;  $t$  is time;  $k_f$ ,  $k_d$ , and  $k_r$  are reaction rate constants. Most of models used for leaching process are of diffusion through ash layer control model.

In this present study, the use of ash layer diffusion controlled shrinking core model was evaluated against the experimental results from leaching process of Pomalaa nickel laterite using citric acid as leachant.

## 2. Materials and Methods

### 2.1. Materials

In this study, nickel laterite was limonite type and was mined from Pomalaa, Southeast Sulawesi Province, Indonesia. Based on the result of x-ray fluorescence (XRF) test, this sample contained 2.73% wt of nickel. As leachant, citric acid (Merck) was used in the form of solution with a concentration of 0.1 M.

### 2.2. Experimental design

300 ml of 0.1 M citric acid solution was poured into the three-neck flask and was heated until 85°C. After the temperature was reached, 60 grams of nickel laterite sample was added into the reactor. The leaching process was done for 2 hours isothermally. In this experiment, the particle size was varying at fractions of 60-70 mesh, 100-120 mesh, and >200 mesh. Periodically, samples were taken and then the solid phase was separated from the liquid phase using centrifuge at 1.000 rpm for 10 minutes. The filtrate (liquid phase) was diluted with distilled water as much as 20 times. Then, the elements in the sample was analyzed using Inductively Coupled Plasma-Optical Emission Spectroscopy (ICP-OES).

## 3. Results and Discussion

The effect of particle size on recovery nickel is shown in Figure 1. Based on that figure, it can be seen that the smaller the particle size was used, the higher the nickel recovery was obtained. For the small particles, the ash layer formed by the reaction is relatively thin so that the molecule transport through the layer is faster. However, Figure 1 also shows that the particle size with a fraction of >200 mesh has no significant effect on the nickel recovery.

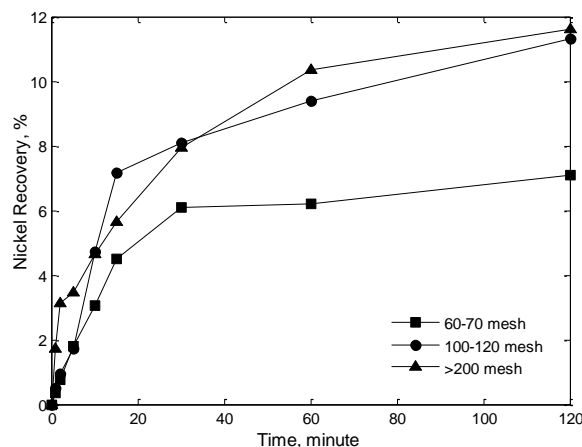
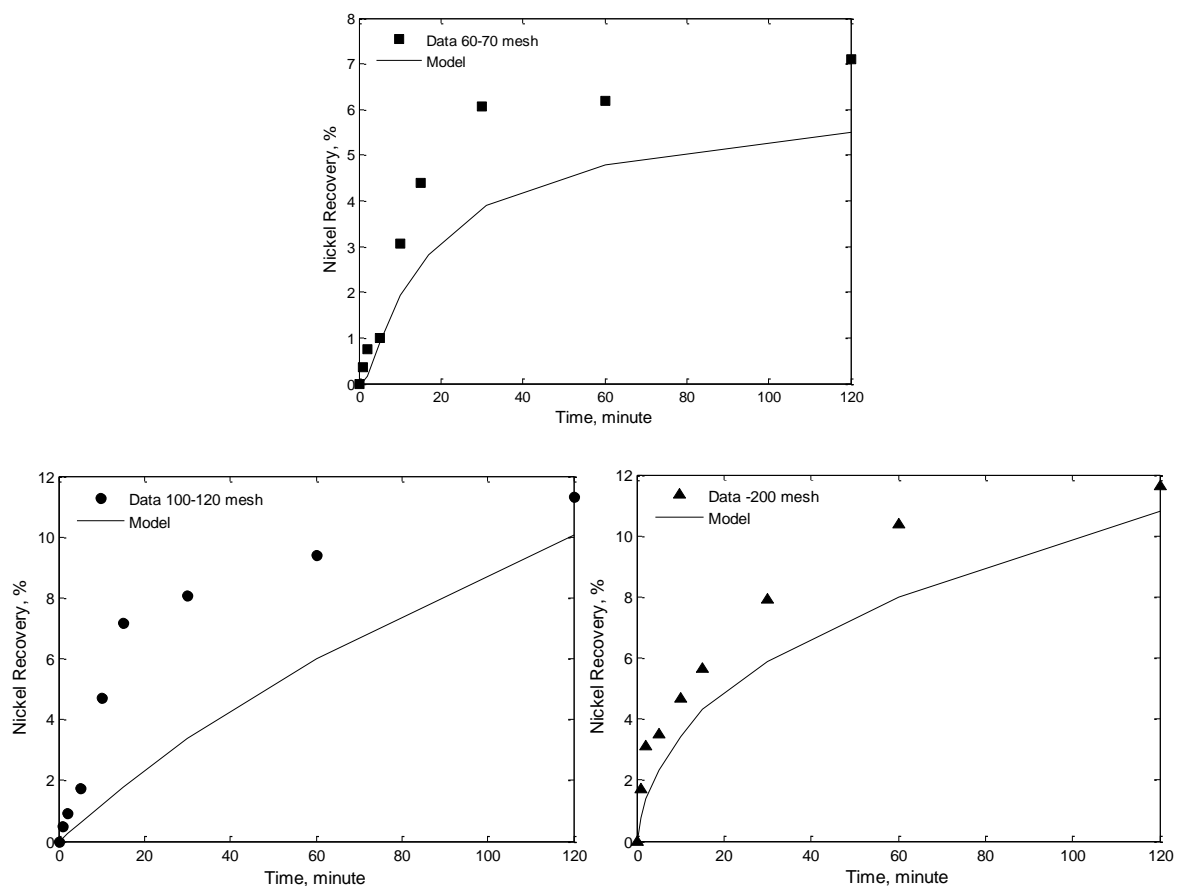


Figure 1. The effect of particle size on nickel recovery

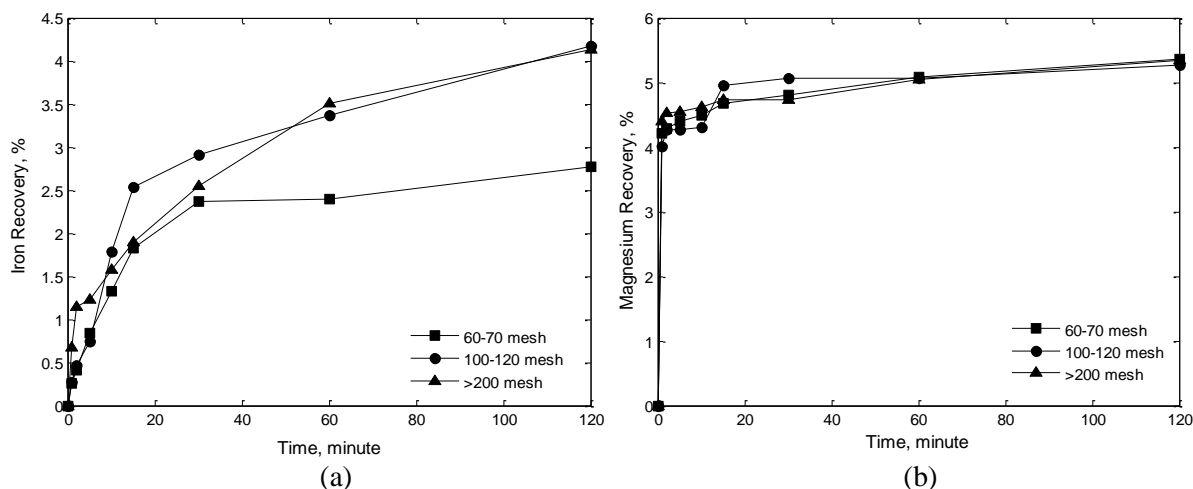
In this study, shrinking core model determined by diffusion through ash layer is evaluated and the simulation results can be seen in Figure 2. Figure 2 shows that shrinking core model is not suitable to describe the phenomenon of the leaching process. The results suggests that the leaching process of Pomalaa nickel laterite using citric acid as leachant is not determined by only one-step controlling process as commonly used in shrinking core models for leaching processes.

Simulation result from this study is not in agreement with the the results reported by some previous researchers. Astuti, et.al. studied leaching process of Pomalaa saprolitic nickel laterite using citric acid as leachant. Their results showed that shrinking core model with diffusion through ash layer controlling process was the most suitable model [7]. Similar conclusion was also reported by other researchers [8,9] that showed that the shrinking core model with diffusion controlling process was much better than other models. However, since the evaluation of the models was merely based on single component data, the conclusion might be misled.



**Figure 2.** Simulation results using diffusion controlled shrinking core model

The experimental data as shown in Figure 1 and 3 indicates that there is another mechanism, besides internal diffusion that might determine the overall leaching process.



**Figure 3.** The effect of particle size on iron and magnesium recovery

As shown in Figure 1 and 3, particle size plays a role in the recovery of nickel and iron. The recovery of nickel and iron was found higher with the decrease of the particle size. However, even though the same leachant was used in all experiments, recovery of magnesium was not influenced by the particle size. Reaction between citric acid and nickel or iron compounds might result in large product molecules that experience steric hindrance of internal diffusion. This result suggests that the leaching process is not controlled by internal diffusion of the reactant molecules. On the other hand, diffusion of product molecules in the ash layer seems to have more important role.

Since the conventional shrinking core model is insufficient to approach the experimental data in this work, development of new mathematical model is necessary. A model that considers steps of chemical reaction and product diffusion through ash layer might be appropriate to describe the physical phenomenon of leaching process of Pomalaa nickel laterite using citric acid as leachant.

#### 4. Conclusion

Shrinking core model was not suitable to describe the phenomenon of leaching process Pomalaa nickel laterite using citric acid as leachant. Experimental data indicated that diffusion of product molecules played a role in the leaching process. Therefore, development of mathematical model involving steps of reaction and product diffusion through ash layer might be necessary.

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