

Article

## Modelling and Simulation of Acid Extraction of Lanthanum from Indonesian Low Grade Bauxite using Fixed-Bed Extractor

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**Abstract.** Demand on lanthanides is growing rapidly due to significant growth of the utilization of advanced materials for industries. Efficient and large scale extraction process of lanthanide from their sources is studied. In this sense, extraction of lanthanum from Indonesian low grade bauxite using a fixed-bed cylindrical extractor with dimension of  $30 \times 3$  cm was modelled and simulated. This simulation model would provide the optimum conditions, extraction time, and cost of extraction, useful for industrial application. A mathematical modelling and simulation was also used to investigate the effects of several parameters, such as particle size, fluid velocity, and acid concentration, on extraction yield of lanthanum. The simulation suggested that the maximum amount of lanthanum sulfate extracted from the low grade bauxite was as high as 6.6 mg after an extraction time of 300 min. The extraction yield can be increased when particle size was decreased, sulfuric acid flow rate was decreased, and concentration of sulfuric acid was increased. This finding can potentially lead to the development of new factory for production of lanthanide using new minor resource.

**Keywords:** Acid leaching, fixed-bed extractor, Indonesian low grade bauxite, lanthanide, modelling, simulation.

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## 1. Introduction

Demand on lanthanides which belong to rare earth elements (REEs) is growing rapidly because of their wide applications in advanced information technology, medical industry, and electronic appliances due mainly to their luminescence properties. Various kinds of biological tests and sensors have also been developed based on the luminescence properties and relatively long-lived emission of the REEs.

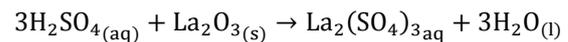
Separation of REEs is a challenging effort in the metallurgy extraction [1]. This is because of the chemical similarity of REEs, and thus separation process requires a series of treatments, including hydrometallurgy method. Extraction of REEs from ores is very complex and lengthy process, such as beneficiation, decomposition of the mineral concentrate to extract REEs, and chemical processing which incorporate the removal of impurity and separation of individual REE to produce a purified saleable product [2]. Hydrometallurgical method involves the series of processes, namely (i) baking or cracking the REEs concentrate; (ii) leaching, neutralization and precipitation methods; (iii) separation and purification processes incorporating solvent extraction and ion exchange [3].

Recovery of REEs from the low grade bauxite has been studied in our laboratory using combination of pyrometallurgy and hydrometallurgy techniques, where acid leaching process is one of the required steps [4-6]. Indonesian low grade bauxite (bauxite tailings) is one of mining wastes with alumina ( $\text{Al}_2\text{O}_3$ ) content about 44.11% [5, 6]. The major components in the Indonesian low grade bauxite are Al of 23.34%, Si of 11.26 % and Fe of 6.82%. With bauxite exploration in Indonesia in total being 1,282,487,720 tons, the low grade bauxite is very abundant in Indonesia. The content of yttrium (Y) and lanthanum (La) metals was found of 0.0041 and 0.0035%, respectively [5, 6]. Separation and extraction of REEs from ores and others using sulfuric acid as an acid leaching solvent have been reported [7-16]. Extraction of REEs from Indonesian low grade bauxite was done in small scale, thus it needs to be developed when the extraction is done in larger scales which is important to fulfil the high demand on REEs in catalyst, optical, and biomedical industries, which will grow rapidly in the future [17, 18]. To this purpose, a larger extractor design has to be considered and a model for extraction of REEs need to be designed, modelled, and simulated to optimize the extractor size, extraction time, and cost of extraction. In this study, a fixed-bed extractor with sulfuric acid as a leaching agent to extract REEs from Indonesian low grade bauxite was used as a model, based on experimental conditions reported previously by Kusri and coworkers [19]. The efficiency of phosphate precipitation for selective extraction of REEs were investigated in details.

## 2. Experimental

### 2.1. Mechanism of Extraction

Solid-liquid extraction is dissolution of one or more compounds from solids by contacting them with liquid solution [20]. The solid-liquid extraction of lanthanum from Indonesian low grade bauxite occurred by several steps as follows: (i) penetration of solvent from bulk through film around particle into particle surface, (ii) solvent diffusion from particle surface in particle pores into surface solid core, (iii) solvent reacts with lanthanum by the reaction of sulfuric acid as solvent and lanthanum as rare earth element in this sample [18], as given by the following reaction scheme,



(ii) REEs will be carried by solvent from the pores to surface of particle, and (iv) REEs will diffuse from particle surface through the film to the bulk phase [21].

### 2.2. Modelling

Model will be created in macro scale and micro scale. Macro scale will represent the bulk phase and micro scale will represent bed particle. Model is created in COMSOL *Multiphysics* 5.3

#### 2.2.1. Bulk phase

In the liquid-bulk phase, a stream of sulfuric acid convection was flowed along the bed. In this stream, axial dispersion occurs as result of velocity gradient during the fluid flow along the bed. In this bulk phase, there is also product accumulation from reaction inside the pellet. In this section, the modelling for the mass balance of the liquid bulk phase will be explained. The liquid bulk phase are considered as convection-diffusion phenomena, and governing equation can be expressed in Eq. (1) as follows:

$$\frac{\partial C_i}{\partial t} = - \underbrace{\frac{\partial}{\partial z} (\varepsilon_b D_i \frac{\partial C_i}{\partial z})}_{(a)} - \underbrace{u \cdot \frac{\partial C_i}{\partial z}}_{(b)} + \underbrace{R_i}_{(c)} \quad (1)$$

where (a), (b), and (c) terms represent dispersion phenomenon, convection, and generation or consumption in the bulk phase, respectively,  $D_i$  is axial dispersion coefficient for fluid in bulk phase,  $C_i$  is substance concentration in bulk phase,  $u$  is interstitial velocity in bulk phase, and  $\varepsilon_b$  is bed porosity. The value of  $R_i$  was calculated using Eq. (2).

$$R_i = h_D (C_i - C_{pei}) S_b \quad (2)$$

where  $h_D$  is interphase mass transfer coefficient calculated using Sherwood number,  $C_{pei}$  is substance concentration in particle and  $S_b$  is bed specific surface area (1/m).

## 2.2.2. Liquid and solid phase in particle

In bed particle, dominant mass transfer is diffusion, thus convection mass transfer can be neglected. In order to calculate concentration of each species, common convection-diffusion inside bed particle takes place, and mass balance for the liquid bulk phase was calculated using Eq. (3).

$$4\pi N \left\{ r^2 r_{pe}^2 \varepsilon_{pe} \frac{\partial C_{pe,i}}{\partial t} + \frac{\partial}{\partial r} \left( -r^2 D_{pe,i} \frac{\partial C_{pe,i}}{\partial r} \right) \right\} = r^2 r_{pe}^2 R_{pe,i} \quad (3)$$

where  $N$  is number of particles per bed volume,  $r$  is a dimensionless radial coordinate that goes from 0 (center) to 1 (particle surface),  $r_{pe}$  is the particle radius,  $\varepsilon_{pe}$  is particle porosity,  $C_{pe,i}$  is species concentration inside particle,  $D_{pe,i}$  is effective diffusion coefficient inside bed particle and  $R_{pe,i}$  is the reaction source term and taken per unit volume of porous particle material.

## 2.3. Economic Analysis

To study the feasibility of the extraction system of lanthanum from Indonesian low grade bauxite whether it can be implemented or not, there are several aspects that must be considered, including cash flow, net present value (NPV), internal rate of return (IRR), and break-even point (BEP).

### 2.3.1. Cash flow

Cash flow shows the amount of profit or loss that will be felt by a project. Cash flows are calculated before taxes are levied and also after taxes are levied. Cash after tax is taken into account, including depreciation and residual value of goods. The cash inflows from the project generally are the selling value and the residual value of the goods and the outflow flow of initial investment costs, operating costs, and loans.

### 2.3.2. Net present value

The net present value shows the benefits gained during project time with a certain interest rate. NPV also shows the current cash flow value obtained from a project. To calculate the NPV, it is important to determine the exact interest rate value. A project is said to be feasible if the value of  $NPV > 0$ , which means that there is a current asset on the project. If the value of  $NPV < 0$ , this means the project does not generate profits or no current assets on the project. To calculate the NPV, cash flows up to the  $n$ -th year are withdrawn to the present time in order to provide current value with reasonable interest. The calculations followed the Eq. (4)-(6).

$$CF_{n,0} = \frac{CF_n}{(1+i)^n} \quad (4)$$

where  $CF_{n,0}$  is the cash flow with the current value,  $CF_n$  is the cash flow in year  $n$ , and  $i$  is the interest rate.

### 2.3.3. Internal rate of return

Internal rate of return (IRR) calculates the maximum interest rate value of a project. IRR is also the value of interest rate when the NPV value is 0. The calculation of IRR using Eq. (5).

$$NPV = \sum_{n=1}^{n=T} \frac{CF_n}{(1+IRR)^n} = 0 \quad (5)$$

By deducing the value of NPV, the IRR value will be obtained.

### 2.3.4. Break even period

Break-even point is an analysis to determine how many goods or services should be sold to cover the overall investment cost. The BEP was calculated using Eq. (6).

$$BEP = \frac{\text{total investment cost}}{(\text{unit price} - \text{operational cost per unit})} \quad (6)$$

## 3. Results and Discussion

### 3.1. Model Validation

The mathematic model for extraction of lanthanum was validated by experimental results in the same operation conditions is as shown in Table 1. There are only different in the raw material treatments between experimental which Indonesian low grade bauxite was firstly roasted and that in the kinetics experimental where the bauxite was untreated. Validation has been conducted by aligning the model curve with the experiment curve. The model and experiment results were calculated into cumulative mass data. As shown in Fig. 1, it can be concluded that the mathematic model is valid.

Table 1. Summary of data used in experimental.

| Operation Parameter         | Value    |
|-----------------------------|----------|
| Extractor Length            | 30 cm    |
| Extractor Diameter          | 3 cm     |
| Bed Porosity                | 0.40516  |
| Temperature                 | 25°C     |
| Pressure                    | 1 atm    |
| Sulfuric Acid Flow Rate     | 1 mL/min |
| Sulfuric Acid Concentration | 3 M      |
| Particle Diameter           | 2 mm     |
| Particle Porosity           | 0.06116  |

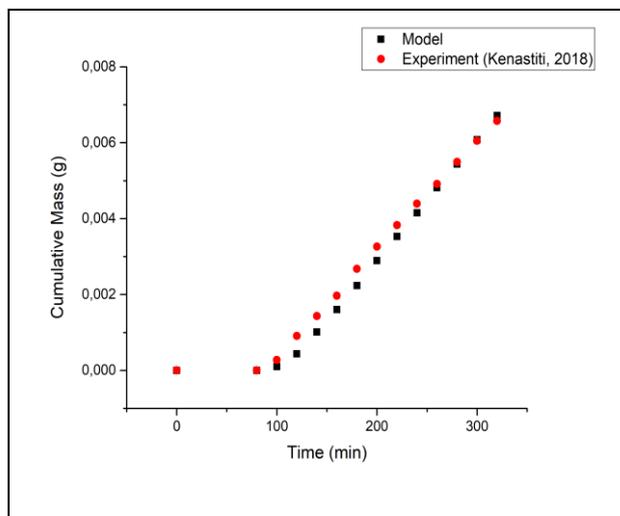


Fig. 1. Validation curve of model based on Experimental.

### 3.2. Simulation Studies

The simulation results including some parameters such as concentration of main extraction product, namely lanthanum sulfate, is shown in Fig. 2. Concentration was calculated as the total lanthanum sulfate irrespective of their forms, either in bulk phase, porous particles, or solid particles. Figure 2 shows the concentration profile of lanthanum sulfate as a function of time at the outlet of extractor bulk phase. As shown in Fig. 2, the time of 80 min of extraction time, there is no lanthanum sulfate yielded. Lanthanum sulfate was yielded at 100 min and increasing gradually until it was saturated at 180 min. At this saturation condition, the concentration of lanthanum sulfate was constant at  $0.23 \text{ mol/m}^3$ .

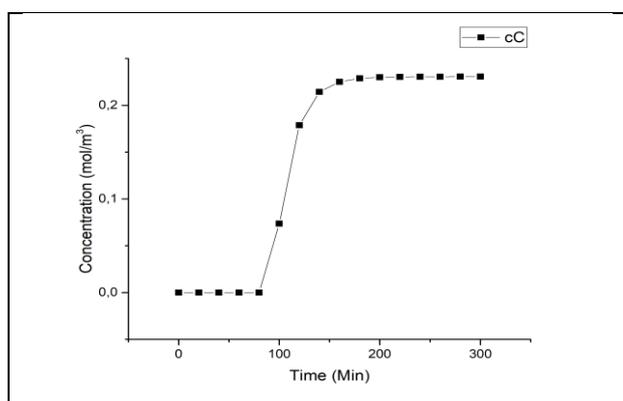


Fig. 2. The concentration profile of lanthanum sulfate at extractor outlet as a function of extraction time at 298 K and 1 atm.

We note that for the first 80 min, there is no lanthanum sulfate at the outlet because fluid flow from inlet to outlet is yet not completed. With the fluid flow rate of  $1 \text{ mL/min}$ , it takes more than 80 min to fully wetting the bed column with 30 cm height. In Fig. 3 we show that the concentration of lanthanum sulfate is slowly moving from the inlet to outlet until 120 min when it reaches the

outlet. The concentration of lanthanum sulfate increased until one point (180 min) then it will be at its equilibrium or saturation, as no more lanthanum sulfate was produced. Lanthanum sulfate was not completely converted because of several factors such as incomplete diffusion due to mass transfer hindrance. Figure 3 shows the concentration of lanthanum sulfate along the extractor at different extraction times from 0 to 300 min. It is clearly observed that the concentration gradient of lanthanum sulfate increase with time, due to fluid convection carrying the lanthanum salt. The concentration gradient of lanthanum sulfate at an equilibrium condition is given in Fig. 3(h).

Concentration profiles of lanthanum sulfate in the form of particle pores along the extractor at different extraction times from 0 to 100 min are shown in Fig. 4, while those in the form of solid phase are shown in Fig. 5. It was observed that both the formations of lanthanum sulfate were dissolved in liquid phase increase slowly along the fluid flow direction from inlet to outlet.

### 3.3. Parametric Variation

In Fig. 6, it can be seen that larger the particle size yields lower concentration of lanthanum sulfate. This is caused by the increasing particle size gives more resistance for substance to get pass through, so the reactant will become more difficult to enter into the particle and the product also become more difficult to get out of particle. Therefore, the concentration of lanthanum sulfate extracted into the phase bulk becomes lower. This applies vice versa for smaller particles radius. However, for smaller particle sizes it is important to note that it will affect the porosity of the bed so that the porosity will be smaller as the fluid becomes harder to flow in the bed and may cause puddles on the bed.

Figure 7 shows that for greater flow rate, the saturation condition is reached faster with the flow rate of sulfuric acid, but the concentration of lanthanum sulfate lower. This can be rationalized by considering that faster flow rate of sulfuric acid causes shorter residence time of sulfuric acid in the bed, so sulfuric acid will quickly wet the entire bed but the length of contact with the bed becomes shorter. In this condition, the substance diffuses into the particles, making possibility of the reaction between lanthanum and the leaching agent. The opposite condition for the slower flow rates. The dependence of the concentration of lanthanum sulfate on the concentration of sulfuric acid is shown in Fig. 8. It was clearly observed that higher concentrations of sulfuric acid produced higher concentration of extracted lanthanum sulfate. This can be simply understood as the extraction process involves chemical reactions, thus when the concentration of reactants increased, the concentration of the product also increased. Extraction of lanthanum from Belitung silica sand by the processes series of heap leaching using sulfuric acid, precipitation and complexation methods has been also reported [22]. It is noted that sulfuric acid is very useful for extraction of lanthanide from minor resource.

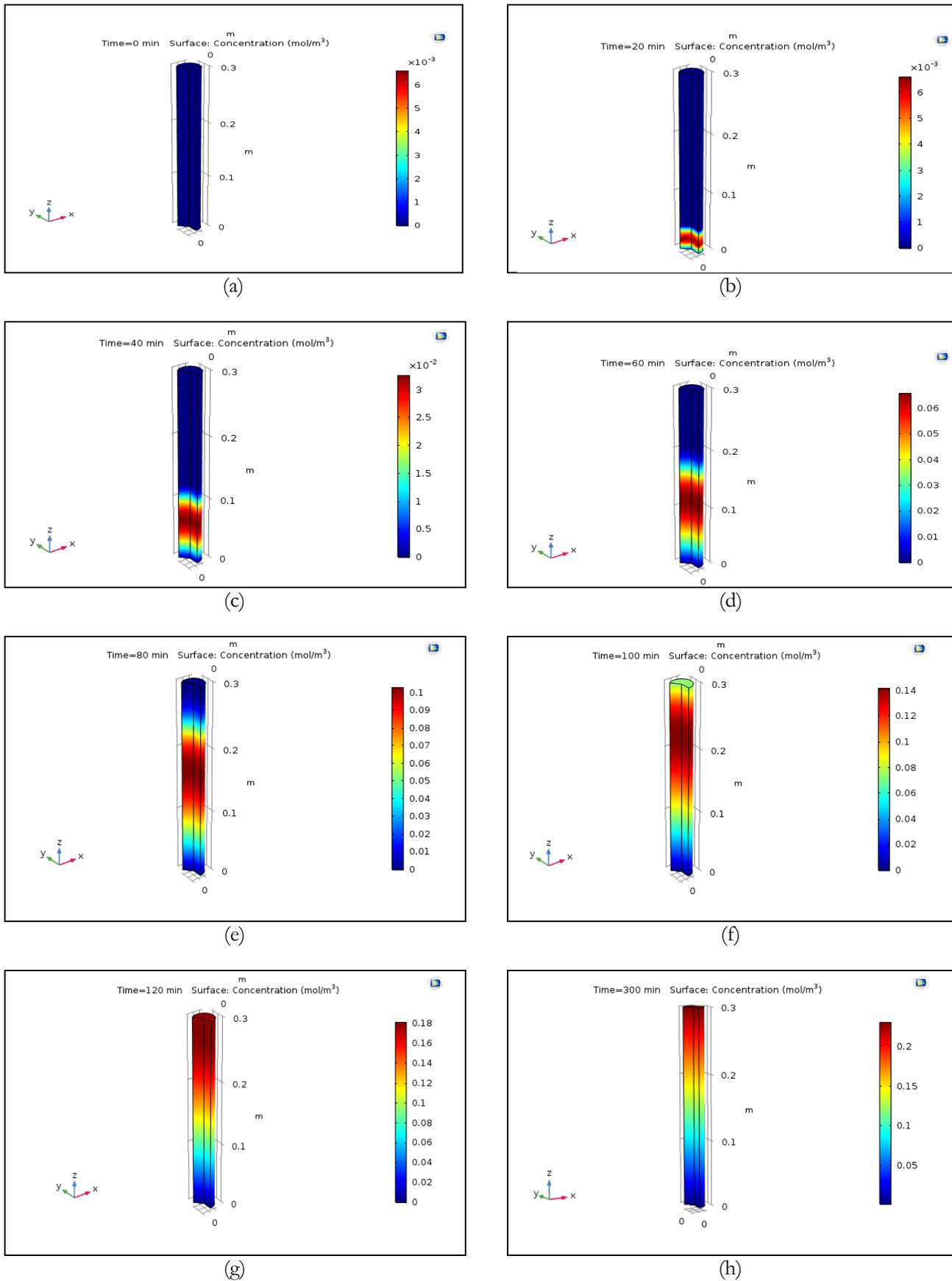


Fig 3. The concentration profile of lanthanum sulfate in bulk phase along the extractor at (a) 0, (b) 20, (c) 40, (d) 60, (e) 80, (f) 100, (g) 120, and (h) 300 min at 298 K and 1 atm.

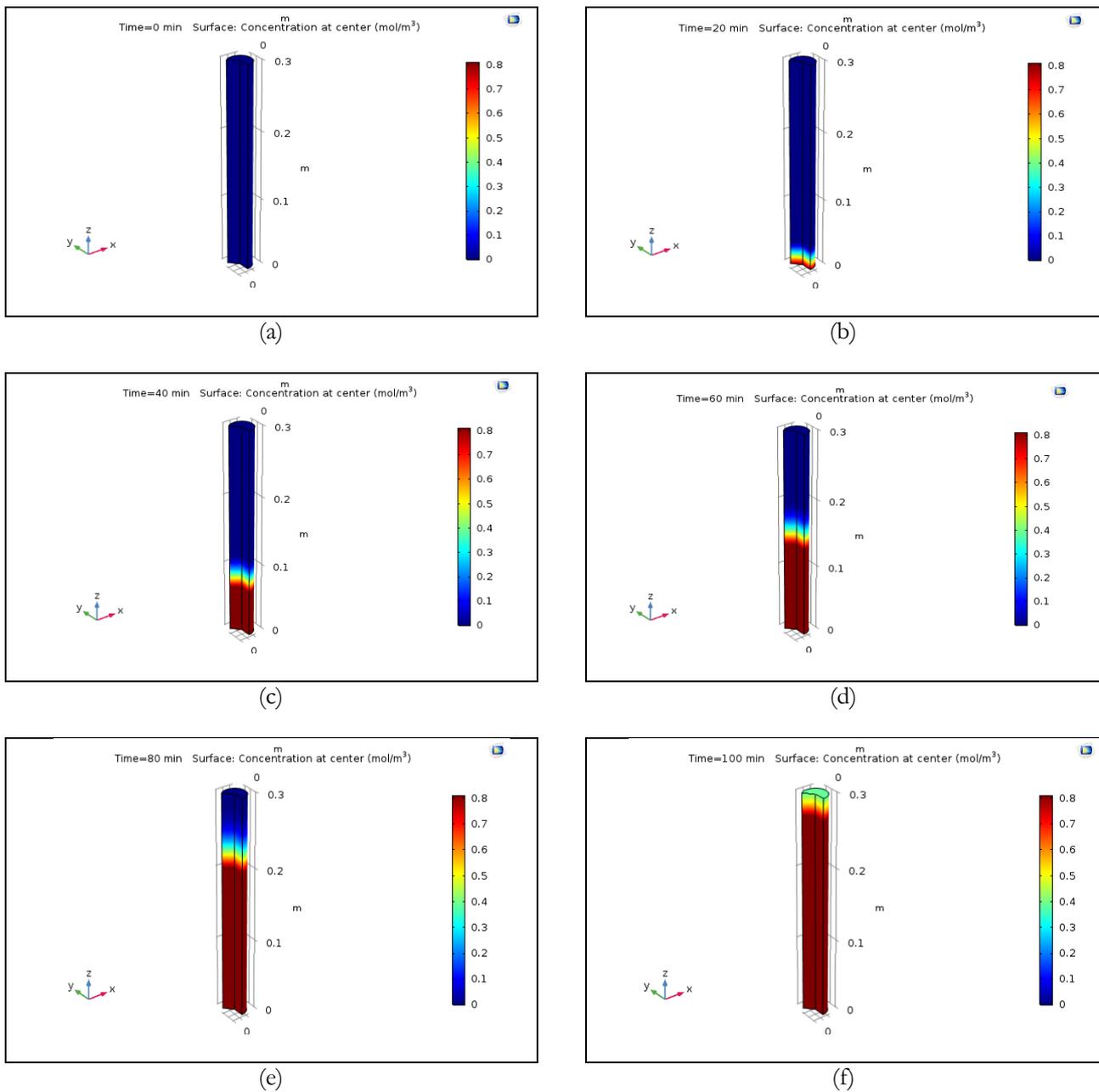


Fig 4. The concentration profile of lanthanum sulfate in particle pores along the extractor at (a) 0, (b) 20, (c) 40, (d) 60, (e) 80, and (f) 100 min at 298 K and 1 atm.

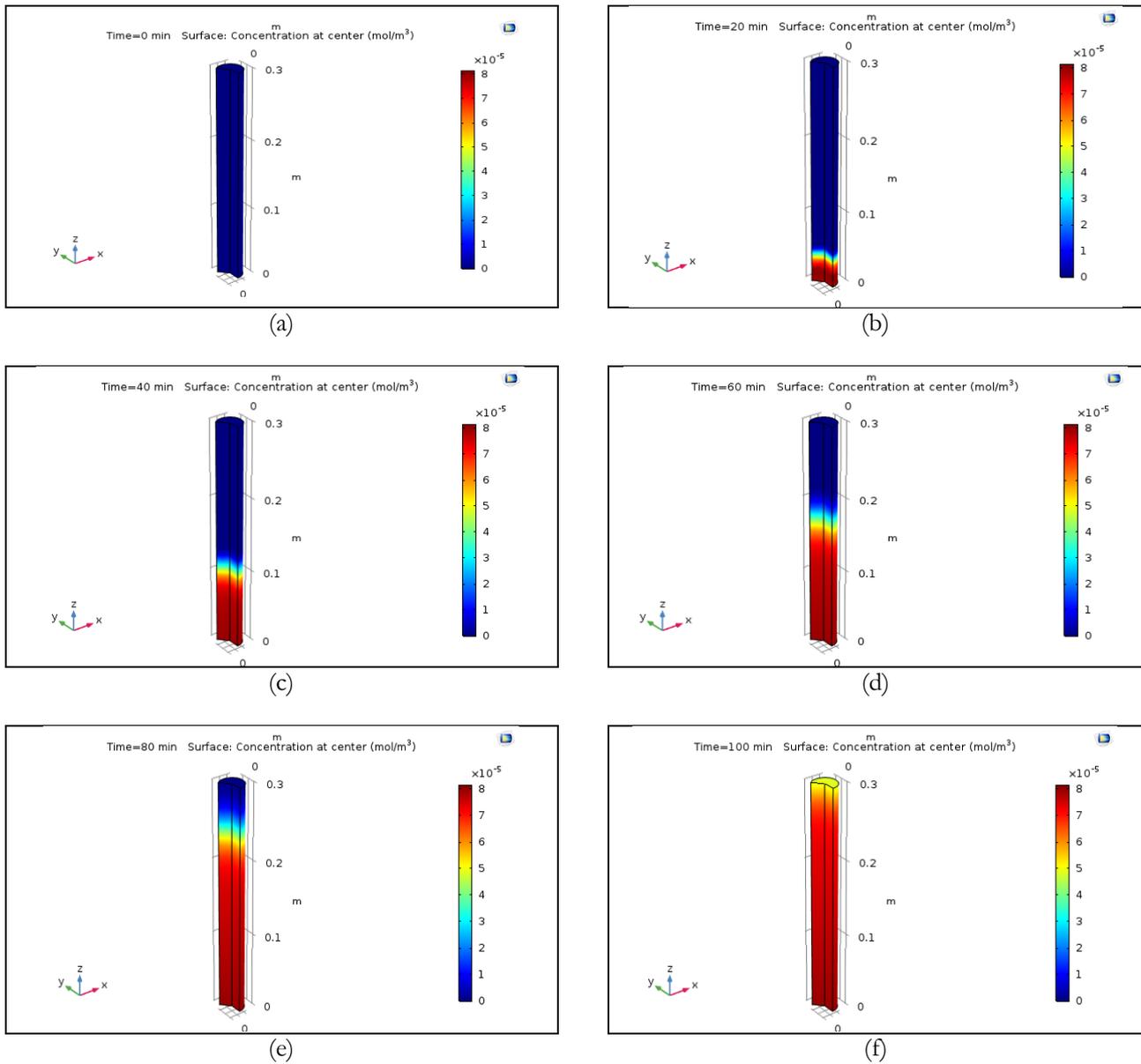


Fig. 5. The concentration profile of lanthanum sulfate in the form of particle solid along the extractor at (a) 0, (b) 20, (c) 40, (d) 60, (e) 80, (f) 100 min at 298 K and 1 atm.

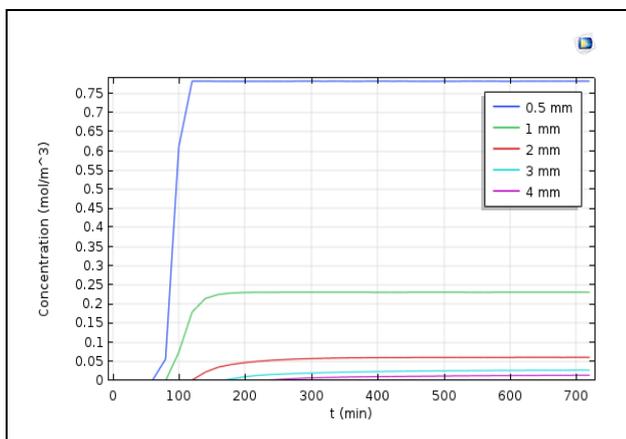


Fig. 6. Relationship between particle size and the concentration of lanthanum sulfate per time at 298 K, 1 atm.

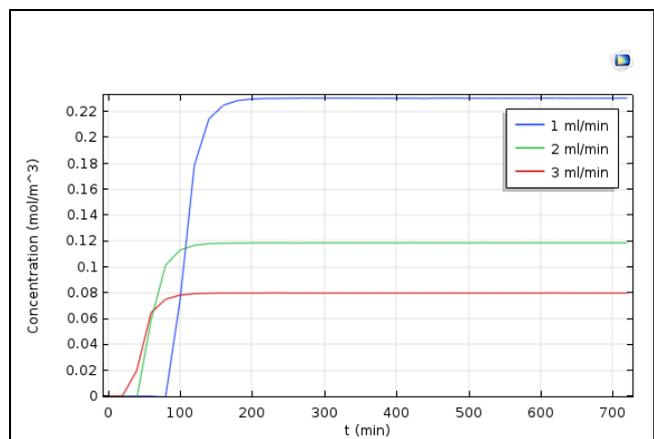


Fig. 7. Relationship between sulfuric acid flow rate and the concentration of lanthanum sulfate per time at 298 K, 1 atm.

### 3.4. Economic Evaluation

Economic evaluation for extraction of lanthanides using a fixed bed extractor was found to be five years with total production of 8.68 g in per year, where extractor operates is 330 days per year and 4 batches per day. By calculating capital expenses (CAPEX) and operational expenses (OPEX) to create a cash flow to determine net present value (NPV) we can found that this extractor is profitable. Table 2 shows the total capital expenses, while Table 3 exhibits the operational expenses and Table 4 presents the cash flow for this project.

Table 2. Capital Expenses.

| No. | Component           | Cost (IDR) | Percentage (%) |
|-----|---------------------|------------|----------------|
| 1.  | Fixed Bed Extractor | 3,800,000  | 100            |
|     | CAPEX               | 3,800,000  | 100            |

Table 3. Operational Expenses.

| No. | Component        | Cost (IDR)  | Percentage (%) |
|-----|------------------|-------------|----------------|
| 1.  | Raw Material     | 24,578,400  | 19.22          |
| 2.  | Labour           | 101,400,000 | 79.29          |
| 3.  | Labour Insurance | 234,000     | 0.18           |
| 4.  | Maintenances     | 570,000     | 0.45           |
| 5.  | Utility          | 1,104,864   | 0.86           |
|     | OPEX             | 127,887,264 | 100            |

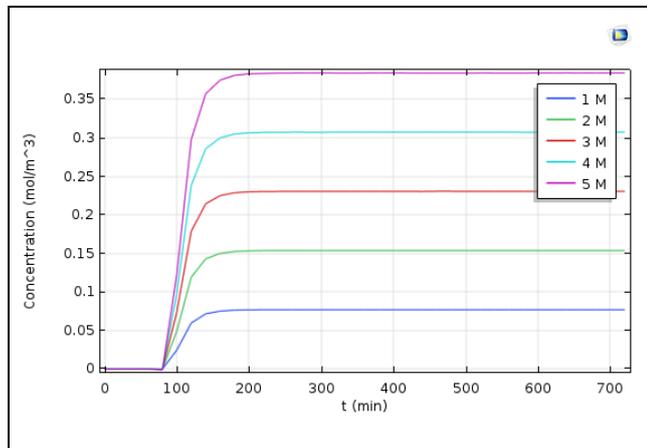


Fig. 8. Relationship between the concentrations of sulfuric acid concentration and lanthanum sulfate per time at 298 K, 1 atm.

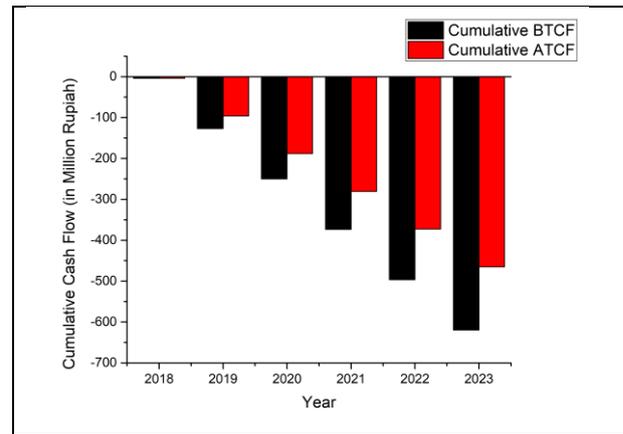


Fig. 9. Cumulative Cash FI.

Profitability for production of REEs from Indonesian low grade bauxite was analyzed using the cash flow (see Table 4). Comparison of before and after tax cash flow is shown in Fig. 9. From the cash flow can be seen that the value of net present value is negative approximately USD 30,000. This indicates that the project is unprofitable and provides great losses, so the IRR value or payback period cannot be calculated. Sensitivity analysis cannot be done because the project value is not profitable. Therefore, this project is impossible to implement economically and still needs further development. This production is not economical because of some reasons such as (i) extraction output value is very low that causes production capacity is really low that affect the income much lower than operational expenses, (ii) in operational expenses, labor force cost is really dominant reaching almost 80 % of total operational expense, it incriminates operational expense. This is caused by the result used for the estimation where it was calculated for industrial scale. Therefore, to make this production of lanthanum from Indonesian low grade bauxite process is profitable, the yield of extraction was needed to be increased, thus the income from sales was also increased. If the same calculation was used, this production can be proposed and it is profitable if the production capacity of lanthanum increased by 27.4 times with the minimum process. It means that the production capacity of 237.8 g lanthanum per year. A benchmark has been done for production cost per mass of REEs with other extraction methods using different raw materials such as clay waste [23]. In this benchmark, the value was only compared with the operational expenses, where the cost mass was reported to be USD 1,000 per gram.

This finding can potentially lead to the development of new factory for production of lanthanide using new minor resource such as Indonesian low grade bauxite. These results can be further utilized by combining in the development of new materials and their derivative compounds based on the lanthanide and low grade bauxite as adsorbents application for advanced technology and industries.

Table 4. Cash Flow for production of lanthanum from Indonesian low grade bauxite.

| Year        | 2018    | 2019      | 2020  | 2021    | 2022  | 2023    |
|-------------|---------|-----------|-------|---------|-------|---------|
| Total       | 0       | 4,709,115 | 4,709 | 4,709,1 | 4,709 | 4,709,1 |
| Income      |         |           | ,115  | 15      | ,115  | 15      |
| CAPEX       | -       | 0         | 0     | 0       | 0     | 0       |
|             | 3,800,0 |           |       |         |       |         |
|             | 00      |           |       |         |       |         |
| OPEX        | 0       | -         | -     | -       | -     | -       |
|             |         | 127,887,2 | 127,8 | 127,887 | 127,8 | 127,887 |
|             |         | 64        | 87,26 | ,264    | 87,26 | ,264    |
|             |         |           | 4     |         | 4     |         |
| BTCF        | -       | -         | -     | -       | -     | -       |
|             | 3,800,0 | 123,178,1 | 123,1 | 123,178 | 123,1 | 123,178 |
|             | 00      | 49        | 78,14 | ,149    | 78,14 | ,149    |
|             |         |           | 9     |         | 9     |         |
| Accumulati  | -       | -         | -     | -       | -     | -       |
| ve BTCF     | 3,800,0 | 126,978,1 | 250,1 | 373,334 | 496,5 | 619,690 |
|             | 00      | 49        | 56,29 | ,448    | 12,59 | ,747    |
|             |         |           | 9     |         | 8     |         |
| Depreciatio | 0       | 760,000   | 972,8 | 396,902 | 192,4 | 170,252 |
| n           |         |           | 00    |         | 18    |         |
| Taxable     | 0       | -         | -     | -       | -     | -       |
| Income      |         | 123,938,1 | 124,1 | 123,575 | 123,3 | 123,348 |
|             |         | 49        | 50,94 | ,052    | 70,56 | ,401    |
|             |         |           | 9     |         | 8     |         |
| Income Tax  | 0       | -         | -     | -       | -     | -       |
|             |         | 30,984,53 | 31,03 | 30,893, | 30,84 | 30,837, |
|             |         | 7         | 7,737 | 763     | 2,642 | 100     |
| ATCF        | -       | -         | -     | -       | -     | -       |
|             | 3,800,0 | 92,193,61 | 92,14 | 92,284, | 92,33 | 92,341, |
|             | 00      | 2         | 0,412 | 387     | 5,508 | 049     |
| Accumulati  | -       | -         | -     | -       | -     | -       |
| ve ATCF     | 3,800,0 | 95,993,61 | 188,1 | 280,418 | 372,7 | 465,094 |
|             | 00      | 2         | 34,02 | ,411    | 53,91 | ,967    |
|             |         |           | 4     |         | 8     |         |

#### 4. Conclusion

Modelling and simulation of extraction of REEs using a fixed-bed extractor with dimension of  $30 \times 3$  cm have been investigated. With the model and sulfuric acid as leaching agent, extraction of lanthanum sulfate from Indonesian low grade bauxite was simulated. The results suggest that the acid leaching of lanthanum sulfate from the low grade bauxite using a fixed-bed extractor can produce the lanthanum salt approximately 6.6 mg for 300 min. The extraction yield of REEs from low grade bauxite increased when particle size was decreased, sulfuric acid flow rate was decreased, and the concentration of sulfuric acid was increased. This finding can potentially lead to the development of new factory for production of lanthanide using new minor resource and it can be further utilized by combining in the development of new materials and their derivative compounds based on the lanthanide for advanced technology and industries.

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