# The Comparison of Red Mud Modification by Acid and Heat for Phosphate Removal from Aqueous Solution

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**Abstract:** *This paper highlights the result obtained from red mud modification by chemical and heat methods. These results pointed that at the same conditions of pH 7.0, modified red mud dose 10g/L, contact time 4 hours, shaking speed at 150rpm in the batch way. The phosphate adsorption capacity of red mud modified by heat at 400°C within 1 hour was found to be over 82%, higher three and twice times compared to raw red mud and red mud modified by 1.5M HCl solution at 80°C in 2 hours. The results also pointed that the isotherm and kinetic of phosphate adsorption onto the surface of both modified red mud were best described by Freundlich and Pseudo second order with the correlation coefficient $R^2 > 0.99$. Besides, heat modification is also considered as a lower cost and environmentally friendly method than the other methods such as acid or heat-acid modification due to without generating waste solution and acid gases.*

**Keywords:** *Red mud modification, phosphate removal, heat modification of red mud.*

## 1. INTRODUCTION

Vietnam is considered as one of the richest bauxite ore resource countries in the world, the total of bauxite ore capacity estimated about 5.5 billion tons and concentrates mostly in the Central Highland (91.4%). In Dak Nong and Lam Dong provinces, the total of bauxite ore is investigated to 4.3 billion tons [1]. At the moment, the biggest alumina plants in Vietnam are Nhan Co alumina (Dak Nong province) and Tan Rai alumina (Lam Dong province) emit approximately 1.2 million tons of dry red mud annually. According to the mass balance, the production of two tons of alumina creates from 2 to 4 tons of red mud or bauxite residue. Therefore, in the next 50 years, about 1.15 billion tons of red mud will be generated from alumina production in the Central Highland.

In the Bayer process, red mud is known formally as bauxite residue and considered a harmful factor to environment due to the chemical composition of red mud. It found that red mud contains various oxides such as $Fe_2O_3$, $Al_2O_3$, $SiO_2$, $TiO_2$, $Na_2O$, $CaO$, $SO_3$, $P_2O_5$, $Cr_2O_3$, *etc* (the percentage of $Fe_2O_3$ and $Al_2O_3$ are estimated about 78.5%). However, the research conducted recently have showed that red mud can be used to remove contaminants in an aqueous solution such as $Zn^{2+}$ [2], $PO_4^{3-}$ [3], $Pb^{2+}$ [4], $Cd^{2+}$ [5], $Co^{2+}$ [6], $As^{5+}$ [7]., etc. With the large amount of red mud generated above, the use of this waste to remove the contaminants in wastewater in general, phosphate in particular can be seen as the effective solution for environmental protection in Vietnam. However, the chemical compositions of red mud is strongly dependents on the characteristics of precursor bauxite ore, as well as on applied technological process, and consequently fluctuates strongly among samples generated in the different parts of the world. Therefore, the results reported previously could not be applied for red mud in Vietnam. Thus, it needs to be conducted the detail research on the red mud modification before using this material for removing of phosphate from an aqueous solution.

**Table 1:** *Chemical compositions of red mud*

| Nations / Elements (%Wt) | Vietnam [1] | China [8] | Turkey [9] | Iraq [10] |
|---|---|---|---|---|
| $SiO_2$ | 2 | 19.14 | 15.95 | 8.52 |
| $Fe_2O_3$ | 15.9 | 12.76 | 36.47 | 26.41 |
| $Al_2O_3$ | - | 6.93 | 19.88 | 18.94 |
| $Al(OH)_3$ | 13.6 | - | - | - |
| $TiO_2$ | - | 3.43 | 4.97 | 7.4 |
| $CaTiSiO_5$ | 9 | - | - | - |
| $CaO$ | - | - | 2.33 | 21.84 |
| $MgO$ | | 1.15 | - | - |
| $FeO(OH)$ | 50.8 | - | - | - |
| $\gamma\text{-}FeO(OH)$ | 3.8 | - | - | - |
| $K_2O$ | - | 1.2 | - | 0.068 |
| $Na_2O$ | - | 2.37 | 10.03 | - |

At the moment, chemical and heat are the most popular methods for red mud modification due to these methods improve the adsorption capacity of red mud

for contaminants removal compared to raw red mud. This study not only concentrated in the evaluation the effect of these methods for red mud modification for phosphate removal from an aqueous solution, but also compared the modification cost between chemical and heat methods to find out which method is really advantage.

## 2. CHEMICALS AND METHODS

### 1. Red mud

Red mud was collected from Tan Rai Alumina Company, is located in Lam Dong province, Central Highland of Vietnam. The pH and surface area of red mud were measured to be 12.8 and 86 $m^2$/g, respectively. 92.3% particle sizes of red mud less than 600 μm. XRD analysis detected that red mud contains the following mineral components: $FeO(OH)$ (50.8% Wt), $Fe_2O_3$ (15.9% Wt), $Al(OH)_3$ (13.6% Wt), $CaTiSiO_5$(9.0% Wt), $CaCO_3$ (4.9% Wt), $\gamma$-$FeO(OH)$ (3.8% Wt), and $SiO_2$ (2% Wt). After pH neutralization by deionized water, red mud was modified by heat at 400ᵒC within 1 hour by using Lenton furnace (1.9KW). Besides, red mud was also modified with 1.5M HCl solution at 80ᵒC in 2 hours. After modification, 400ᵒC modified red mud was used directly for phosphate adsorption, 1.5M HCl modified red mud was continue washed and neutralized by deionized water and dried at 105ᵒC.

### 2. Chemicals

All chemicals used in this study (HCl, $HNO_3$, NaOH, $KH_2PO_4$) were of analytical grade and produced by Merck (Germany). The phosphate working solution with the concentration range of 25.0 - 200 mg/L was prepared from $KH_2PO_4$ salt and diluted by deionized water. The initial pH of phosphate solution was adjusted by using NaOH and $HNO_3$ solutions

### 3. Experimental procedure

The phosphate adsorption experiments were conducted by batch technique at the room temperature. A fixed amount of the modified red mud (except the adsorbent dose experiments) was added into a series of the 100 mL flask containing 25 mL of phosphate solution with the different of concentrations, initial pH, and shaken at 150 rpm. After finishing each adsorption experiments and phases separation by Universal 320R centrifuge (Hettich Zentrifugen) at 6000rpm in 5 minutes, the pH of aqueous solution was measured again to determine the equilibrium pH by using Orion 4 star pH meter

(Thermo Scientific). The residual phosphate concentration in the adsorbed solution was analyzed by SMWW 4500 – E method [11] and Lambda 35 UV/VIS Spectrometer (Perkin Elmer). The phosphate removal efficiency is calculated based on the mass balance equation as follows.

$$H(\%) = \frac{c_o - c_e}{c_o} x 100 \qquad (1)$$

Where:

$C_o$: Initial phosphate concentration (mg/L)

$C_e$: phosphate concentration in the adsorbed solution at the equilibrium (mg/L)

Besides the adsorption experiments, the blank sample was also carried out to ensure that no adsorption was taking place on the wall of the flask used.

### 4. Analytical methods

The change in chemical compositions and surface of the heat and acid modified red mud before and after adsorption will be determined by X-ray Powder Diffraction (Siemens D5005, Germany) and SEM analysis methods (Nova Namo Scaning Electron Microscopy NEP 199). The weight reduction of red mud before and after heat and acid modification was determined by GH 252 analytical balance (A&D Instrument Company, England).

## 3. RESULTS AND DISCUSIONS

### 1. Effect of initial pH on phosphate removal

The effect of initial pH on phosphate removal by 400ᵒC and 1.5M HCl modified red mud is presented in Fig 1. It is easy to find the same trend for both modified red mud, when initial pH was 1; 3; 5; 6, the phosphate removal efficiency increased and reached maximum 82% and 52% with 400ᵒC and 1.5M HCl modified red mud at pH 6.0. After that phosphate decreased with the increase of pH. This trend similar behavior was also reported for phosphate from aqueous solution by Pradhan [9], Liu Chang-jun [12], and Genc [13].
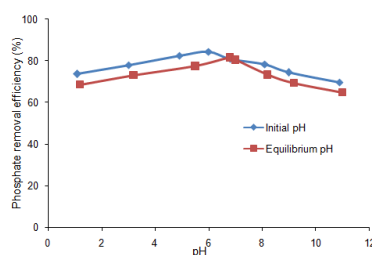


**Fig 1:** *Effect of initial pH on phosphate removal*

*(Modified red mud dose 10g/L, temperature 25ᵒC, shaking 150 rpm, initial phosphate concentration 50 mg/L, contact time 4 hours)*

This trend occurred due to the competition for binding sites among cations and protons at low pH values. While at higher pH, solubility of complexes decreases sufficiently allowing precipitation which may complicate the sorption process and do not bind to the sorption sites on the surface of the modified red mud [10].

As Kamiyango calculated and reported [14], the pH effects on both fraction compositions of phosphate species and charge development on the red mud surface. In the solution, phosphate exists under various forms such as $H_2PO_4^-$, $H_3PO_4$, $HPO_4^{2-}$, and $PO_4^{3-}$ (Fig -2), in the range of pH from 4.0 to 6.0, phosphate is mainly exists in $H_2PO_4^-$, which form easy to metal oxides containing in the red mud with the binding energy is 135.2eV [15].
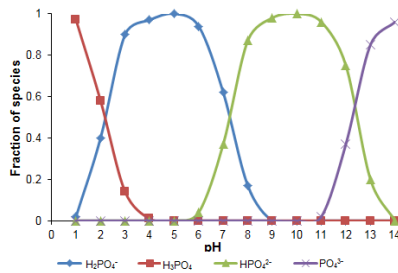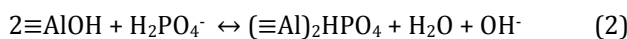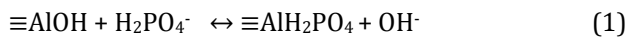


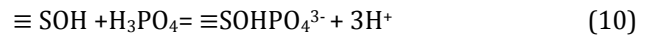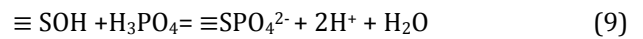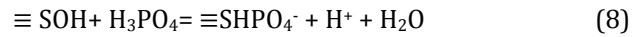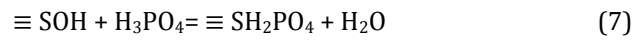**Fig 2:** *Fraction composition of phosphate species in the solution at different pH [14]*

Kamiyanggo also indicated that the adsorption of phosphate on $Al(OH)_3$, $FeO(OH)$ is guided by the $\equiv AlOH$ and $\equiv FeOH$ group, located on crystal edges. When phosphate ion contact with $Al(OH)_3$ and $FeO(OH)$ will create the inner sphere complexes between phosphate and singly coordinated hydroxyl group. The ligand exchange reaction mechanism is presented as equations following:

$$\equiv AlOH + H_2PO_4^- \ \leftrightarrow \equiv AlH_2PO_4 + OH^- \tag{1}$$

$$2\equiv AlOH + H_2PO_4^- \ \leftrightarrow (\equiv Al)_2HPO_4 + H_2O + OH^- \tag{2}$$

$$\equiv FeOH + H_2PO_4^- \ \leftrightarrow \equiv FeOPO_2OH + H_2O \tag{3}$$

$$2\equiv FeOH + H_2PO_4^- \leftrightarrow \equiv (FeO)_2PO_2 + 2H_2O \tag{4}$$

The same point with Kamiyango, Liu Chang-Ju [12] also indicated that a ligand exchange mechanism took place in the sorption of phosphate onto the samples, and the sorption reaction may be written with the following ligand exchange reactions. When the red mud was added into a solution, there exists such surface hydrolysis reactions depending on pH [12]:

$$\equiv SOH + H^+ = \ \equiv SOH_2^+ \tag{5}$$

$$\equiv SOH = SO^- + H^+ \tag{6}$$
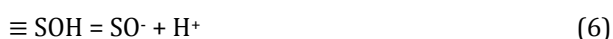
When phosphate exists in the solution, then these sorption reactions take place depending on pH:

$$\equiv SOH + H_3PO_4 = \ \equiv SH_2PO_4 + H_2O \tag{7}$$

$$\equiv SOH + H_3PO_4 = \ \equiv SHPO_4^- + H^+ + H_2O \tag{8}$$

$$\equiv SOH + H_3PO_4 = \ \equiv SPO_4^{2-} + 2H^+ + H_2O \tag{9}$$

$$\equiv SOH + H_3PO_4 = \ \equiv SOHPO_4^{3-} + 3H^+ \tag{10}$$

Where: $\equiv SOH$ represent sample surface of red mud.

When the pH increase, the amount of $H_3PO_4$ decreased and $H_2PO_4^-$ and $HPO_4^{2-}$ were the predominant aqueous species, in the study of Shengjiong Yang [16], phosphate - metal precipitation reaction seems to be an alternative explanation for phosphate adsorption efficiency, which could be presented as:

$$xPO_4^{3-} + 3M^{x+} = M_3(PO_4)x \tag{11}$$
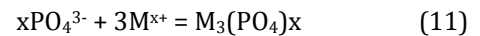
Where: M is the metal component, such as Al, Fe, Ca, and Si. Shengjiong Yang also pointed that monovalent dihydrogen phosphate ($H_2PO_4^-$) was well adsorbed by red mud, this idea is the same with Kamiyanggo.

## 2. Effect of contact time

Contact time is one of the most effective factors in batch adsorption process. Adsorption rate initially increased rapidly, and the optimal removal efficiency 82% and 52% was reached after 4 hours for 400°C and 1.5M HCl modified red mud, respectively. Further increase in contact time did not show significant change in equilibrium concentration; that is, the adsorption phase reached equilibrium due to the nature of adsorbent and the available adsorption sites affect the rate of adsorption of phosphate. The mechanism of solute transfer to the solid includes diffusion through the fluid film around the adsorbent particle and diffusion through the pores to the internal adsorption sites. In the initial stages of adsorption of phosphate, the concentration gradient between the film and the available pore sites is large, and hence the rate of adsorption is faster. The rate of adsorption decreases in the later stages of the adsorption probably due to the slow pore diffusion of the solute ion into the bulk of the adsorbent [17].
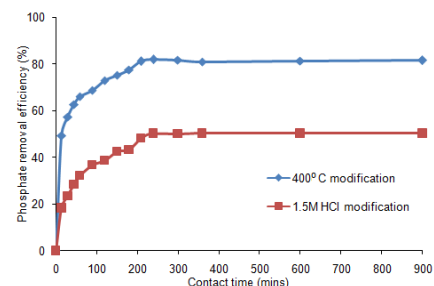


**Fig 3:** *Effect of contact time*

*(Initial phosphate concentration 50mg/L, temperature 25oC, red mud dose 10g/L, initial pH 6.0, shaking 150rpm)*

## 3. Effect of modified red mud dose

The effect of modified red mud dose was studied in the range of 2.0 - 20 g/L. It was observed that the adsorption percentage of phosphate ion onto the modified red mud increased rapidly with the increasing of adsorbent dose (Fig -4). This result is expected because the increase of adsorbent dose leads to greater surface area. When the adsorbent concentration was increased from 2 to 10 g/L, the percentage of phosphate ion adsorption increased from 42.2 and 25.9% to 82 and 52% for 400oC and 1.5M HCl modified red mud, respectively. At higher dosage, the equilibrium uptake of phosphate ion did not increase significantly with increasing modified red mud dosage. Such behavior is expected due to the saturation level attained during an adsorption process. This finding agrees with Koumanova.B [18]. For subsequent studies, a dose of 10 g/L of modified red mud was selected.
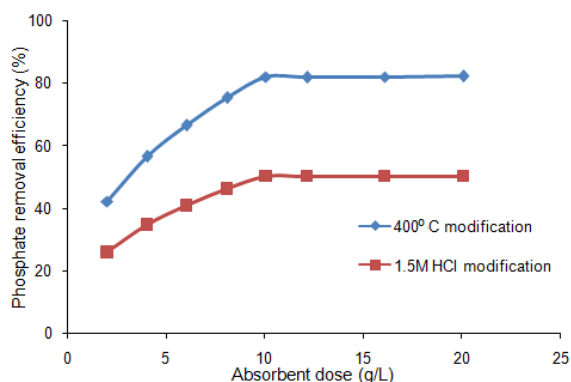


**Fig 4:** *Effect of modified red mud dose*

*(Initial pH 6.0, contact time 4h, temperature 25oC, shaking 150rpm, initial phosphate concentration 50mg/L)*

## 4. Effect of initial phosphate concentration

The effect of initial adsorbate concentration on the adsorption was investigated by varying the initial concentration of phosphate from 25 to 200 mg/L. The experimental data were illustrated in Fig -5. The results indicated that the increase in the initial phosphate concentration led the removal efficiency of heat and acid modified red mud decrease from 90.2%, 55.33% (when phosphate was 25mg/L) to 46.32% and 28.42% (when phosphate increased to 200mg/L). The decrease can be explained by the competition of amount of phosphate to contact with the limited of adsorption sites on the surface of red mud.
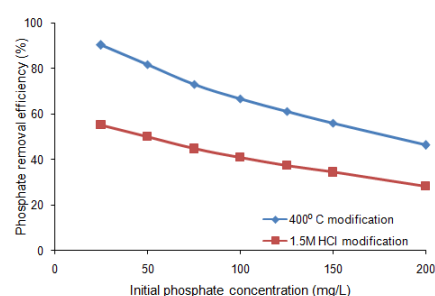


**Fig 5:** *Effect of initial phosphate concentration (modified red mud dose 10g/L, initial pH 6.0, temperature 25oC, shaking 150rpm, contact time 4 hours)*

## 5. Adsorption isotherm

**The linearized Langmuir adsorption isotherm** equation which is valid for monolayer sorption onto a surface with finite number of identical sites is as follows:

$$\frac{t}{q_e} = \frac{1}{q_m} bC_e + \frac{t}{q_m} \tag{12}$$

Where:

$C_e$: the equilibrium concentration of phosphate in solution (mg.L$^{-1}$),

$q_e$: the amount phosphate adsorbed at equilibrium (mg/g),

$q_m$: the theoretical maximum adsorption capacity (mg/g)

b: the Langmuir constant (L/mg).

The linear plot of $\frac{1}{C_e}$ versus $\frac{t}{q_e}$ with correlation coefficient R$^2$ over 0.98 for both 400oC and 1.5M HCl modified red mud indicates the applicability of Langmuir adsorption isotherm (Figure 6). This indicates a monolayer sorption of lead onto the adsorbent surface. The maximum adsorption capacity 'q$_m$' and binding energy constant 'b' of heat and acid modified red mud for phosphate was 10.2 mg/g and 7.63mg/mg, respectively according to Langmuir model.
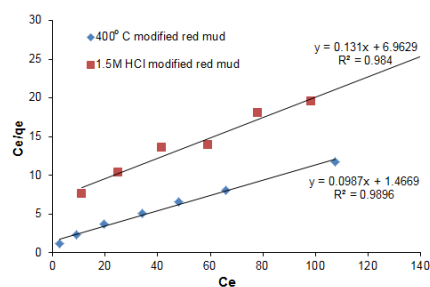


**Fig 6:** *Langmuir isotherm of phosphate adsorption by modified red mud*

*Freundlich adsorption isotherm* adopts multilayer adsorption on heterogeneous surfaces. Linearized form of the Freundlich equation is given by the following equation:

$$\ln q_e = \ln K_f + \frac{1}{n} C_e \qquad (13)$$

Where:

$q_e$: the amount of lead ions adsorbed at equilibrium time (mg/g),

$C_e$: the equilibrium concentration of lead ions in the solution (mg/L),

$K_f$: the adsorption capacity (mg/g),
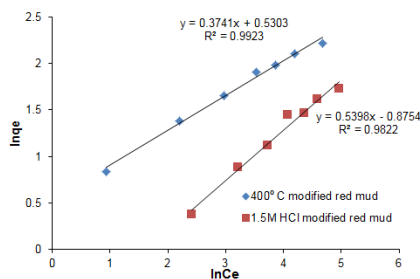
n: an empirical parameter,



**Fig 7:** *Freundlich isotherm of phosphate adsorption by modified red mud*

The isotherm parameter of both Langmuir and Freundlich are shown in Table 2. According to $R^2$ coefficient, it is easy to release that the phosphate adsorption followed both single and multi-layer on the surface of red mud. However, the Freundlich isotherm provides a better fitting to isotherm than Langmuir isotherm due to the higher $R^2$. Therefore, it can be suggested that some heterogeneity in the surface of modified red mud play a role in the phosphate adsorption.

**Table 2:** *Isotherm parameters of Langmuir and Freundlich for phosphate adsorption*

| Langmuir isotherm | | |
|---|---|---|
| 400°C modified red mud | $Q_{max}$ (mg/g) | 10.20 |
| | K | 0.68 |
| | $R^2$ | 0.986 |
| 1.5 M HCl modified red mud | $Q_{max}$ (mg/g) | 7.63 |
| | K | 0.14 |
| | $R^2$ | 0.984 |
| Freundlich isotherm | | |
| 400°C modified red mud | n | 2.67 |
| | $K_f$ | 1.45 |
| | $R^2$ | 0.992 |
| 1.5 M HCl modified red mud | n | 1.68 |
| | $K_f$ | 1.71 |
| | $R^2$ | 0.982 |

## 6. Adsorption kinetics

The rate constant $K_1$ for the adsorption of phosphate was studied by Lagergren rate equation [19], for initial phosphate concentration of 50 mg/L.

*Pseudo-first-order rate expression of Lagergren equation:*

$$\log(q_e - q_t) = \log q_e - \frac{K_1 t}{2.203} \qquad (14)$$

Where:

$q_e$: The amount of phosphate adsorbed at the equilibrium (mg/g)

$q_t$: The amount of phosphate in the adsorbed solution at time 't' (min), respectively (mg/g)

$K_1$: The Pseudo-first-order rate constant (min$^{-1}$).

The $K_1$ and correlation coefficient $R^2$ were calculated from the slope of the linear plot of $\log(q_e - q_t)$ versus 't' at different time intervals. The $K_1$ and $R^2$ of both 400°C and 1.5M HCl modified red mud are shown in Table 3

*The Pseudo-second-order rate expression is:*

$$\frac{t}{q_t} = \frac{1}{K_2 * q_e^2} + \frac{t}{q_e} \qquad (15)$$

Where: $K_2$ is the Pseudo-second-order rate constant (g/mg.min).

The calculation from the slope and intercept of the plot $\frac{t}{q_t}$ versus time 't' was almost linear shown in Fig -9.

The values of $K_1$, $K_2$ and $R^2$ of both 400°C and 1.5M HCl modified red mud are shown in Table 4
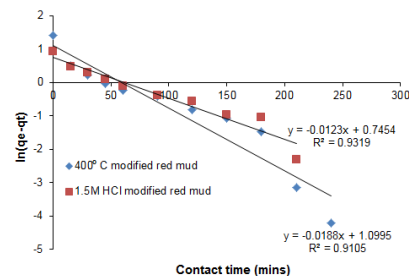


**Fig 8:** *Pseudo –first – order*

**Table 3:** *Kinetic parameter of Pseudo –first – order*

| | Pseudo first order | | | |
|---|---|---|---|---|
| | $K_1$ (min$^{-1}$) | $q_e$ (mg/g) | $R^2$ | $q_e$* (mg/g) |
| 400°C modified red mud | 0,018 | 3 | 0,91 | 4.09 |
| 1.5M HCl modified red mud | 0.012 | 2.13 | 0.931 | 2.51 |
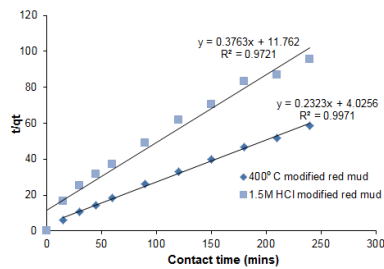
**Fig 9:** *Pseudo –second – order*

**Table 4:** *Kinetic parameter of Pseudo –second – order*

| | Pseudo second order | | | |
|---|---|---|---|---|
| | $K_2$, g/mg.min | $q_e$, mg/g | $R^2$ | $q_e^*$ |
| $400^oC$ modidied red mud | 0,014 | 4,31 | 0,997 | 4.09 |
| 1.5M HCl modidied red mud | 0.012 | 2.66 | 0.972 | 2.51 |

*Note: $q_e^*$ is experimental value*

The fitted curve and parameters of kinetic models are shown in **Fig -8, Fig -9** and Table 3, Table 4. It is clear that the phosphate adsorption process on modified red mud is well described by the Pseudo second-order model because of the high correlation coefficient $R^2$ was 0.997 and 0.972 for heat and acid modified red mud compared to $R^2$ was 0.91 and 0.93 by Pseudo first-order.

According to the results introduced above, it can be suggest that the phosphate adsorption by red mud modified by heat at $400^oC$ is more effective than acid method with 1.5M HCl solution due to the increase in area surface. After heat modification the area surface of red mud was measured about $100.68 m^2/g$ higher than 11.85% and 13.73% compared to red mud modified by acid ($88.83 m^2/g$) and raw red mud ($86.95 m^2/g$), respectively. Besides, some chemical compositions of modified red mud were found to decrease in content. In the detail, some elements content in red mud modified with 1.5M HCl solution were found to decrease such as $FeO(OH)$, $Fe_2O_3$. $CaTiSiO_5$, $CaCO_3$ then these elements content in red mud modified by heat at $400^oC$ were found to increase, especially $CaTiSiO_5$ and $CaCO_3$ content were higher twice time. It is considered as a cause of phosphate adsorption capacity of heat modified red higher than acid modified red mud (**Fig -8**)
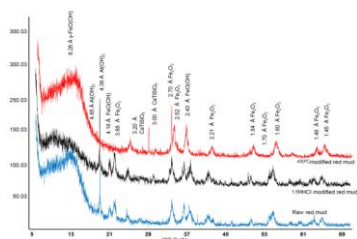


**Fig-8:** *XRD analysis results of red mud after heat and acid modification*

The increase in surface are of red mud can be seen easily through SEM images (Fig -9), the particle size red mud modified by heat at $400^oC$ is finest compared with raw red mud as well as red mud modified with 1.5M HCl solution.
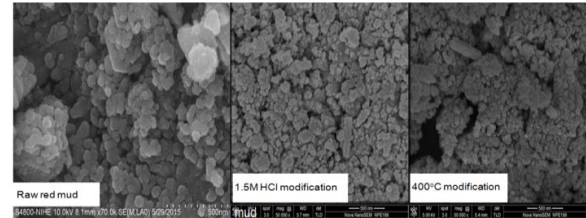


**Fig 9:** *SEM images of red mud before and after $400^o$ and 1.5M HCl modification*

As shown in Fig 10, $\gamma FeO(OH)$, $Al(OH)_3$, $FeO(OH)$, $Fe_2O_3$, $CaTiSiO_5$ were detected on the surface of modified red mud before phosphate adsorption. After the adsorption process, two strange peaks ($4.25A^o$ and $2.25A^o$) appeared on the spectrum, which demonstrated that phosphate was successfully adsorbed onto the surface of modified red mud. Although the XRD's peaks did not indicate exactly the form of phosphate appeared after adsorption, but based on Barbaux's report, phosphate could react with $FeO^+$ and $Al(OH)_3$ by the precipitation and ion exchange mechanism [20] or the formed phosphate complexes including $[FeOPO_4]_{adsorption\ species}$ and $[Al(OH)_3-PO_4]_{adsorption\ species}$ could also adsorb additional $H_3PO_4$ analog by hydrogen bonding [21].
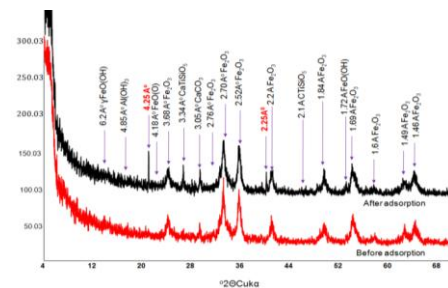


**Fig 10:** *XRD analysis results of $400^oC$ modified red mud before and after adsorption*

## 7. Modification cost

The phosphate removal efficiency of red mud modified by heat and acid was not only investigated on the adsorption capacity, but also compared on the modification cost. The data introduced in Table 5 is the comparison of cost modification by heat and acid.

It is easy to release that to modifying the same amount of raw red mud (1kg), heat method is a very low cost and simple compared to acid method. Besides, heat

method is also friendlier to environment due to without flue gases as well as aqueous solution generation and modification cost was 0.237 USD only, while acid method generated 10 litter of HCl solution and HCl gas during modification and modification cost up to 3.431 USD, higher than 10 times compared to heat modification.

**Table 5:** *Modification cost of red mud*

| Thermal modification method Modification time : 1h Type of furnace: Lenton (England) Capacity         : 1.9KW Max temperature   : 800$^o$C | | |
|---|---|---|
| | Electric consumption (kWh) | Cost (USD) |
| Temperature modification: 400$^o$C | | |
| Electric consumption | 0.95 | |
| Cost/KWh | | 0.25 |
| Waste generation (flue gases, waste water) | None | |
| **Modification cost** | | **0.237** |
| **HCl modification method** Modification time: 2h | | |
| Heater : <br>• Max temperature: 300$^o$C <br>• Capacity: 0.625KW <br>• Temp. modification: 80$^o$C <br>• Electric consumption | 0.333KWh | 0.083 |
| Stirrer: <br>• Max stirring speed: 1200rpm <br>• Stirring speed: 300rpm <br>• Capacity: 0.125KW | 0.063KWh | 0.016 |
| • HCl (industrial quality 4USD/Litter) <br>• HCl consumption: 0.833 litter | | 3.332 |
| Waste generation (flue gases, waste water) | • 10 liters of waste HCl solution <br>• HCl gas <br>• Deionized water for neutralization | |
| **Modification cost** | | **3.431** |

## 4. CONCLUSION

According to the results obtained in this study, some conclusion can be recommended as follow:

• Initial pH, initial phosphate concentration, contact time, modified red mud dose could influence the phosphate adsorption performance of 400$^o$C and 1.5M HCl modified red mud significantly.

• The kinetic and isotherm of phosphate adsorption onto the surface of both 400$^o$C and 1.5M HCl modified red mud were well described by Pseudo second order and Langmuir–Freundlich isotherm, which demonstrated that the phosphate adsorption onto 400$^o$C and 1.5M HCl modified red mud could be governed by multiple mechanisms.

• Heat modification of red mud at 400$^o$C in 1hour is an effective and lower cost compared acid modification method with HCl 1.5 M solution. The adsorption capacity of 400$^o$C modified red mud higher 30% and modification cost 0.237USD/kg of raw red mud only, 10 times lower than acid modification with modification cost 3.431USD/kg of raw red mud.

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