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Economic Assessment of Toxic Gases Separation from Hydrolysis of Ssecond Aluminum Dross

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Abstract

Second aluminium dross from the recycling process is regarded as a hazardous waste. This work investigates the methods to convert the second aluminum dross into the useful product (i.e., sodium aluminate) and eliminate the toxic gases. Three different processes to treat the second aluminum dross are considered and compared. The first process involves the hydrolysis of aluminum dross in basic solution coupled with gas stripping using water. The second one is to hydrolyze aluminum dross in basic solution coupled with gas stripping using hydrochloric acid. Aluminum hydroxide in aluminum dross is hydrolyzed in basic solution to obtain sodium aluminate. The third one is to hydrolyze aluminum dross in acid solution coupled with gas stripping using potassium permanganate followed by ethylene glycol. The suitable concentration of hydrochloric acid is 0.01 mol dm^{-3} that can trap ammonia gas in the second process. For the third process, 1 mol dm^{-3} of potassium permanganate with the flow rate of 100 L min^{-1} can completely react hydrogen sulfide. From the economic analysis, the total investment cost of the third process is the lowest due to the less number of unit operations. However, the second process is preferable regarding the net present value and payback period.

Keywords: Second aluminum dross, Hydrolysis, Gas stripping, Process design, Economic assessment

1. Introduction

Aluminium metal is used for many chemical industries and should be recycled for efficient utilization of its resource. Aluminum casting and aluminium recycling cause secondary aluminium dross that is a hazardous waste. It is usually disposed to landfill because the elimination of this waste is an expensive cost. However, this treatment leads to the generation of toxic gases such as ammonia and hydrogen sulfide (Calder and Stark, 2010). Many researchers reported that the hydrolysis of aluminium dross could minimize this waste but an increase in ammonia gas (Tsakiridis, 2012). As a consequence, it leads to an odor pollution problem and affects the respiratory system. In general, the separation process, such as water stripping, in chemical industries has low. Few effective separation process can reduce toxic gases from aluminium recycling on an industrial scale (Singh et al., 2019). Thus, the treatment and separation process should be explored for the elimination of secondary aluminium dross and toxic gases. Chemical scrubbing process can separate toxic gases from the air for desulfurization of the natural gas product (El-Melih et al., 2016). It can be implemented for aluminium dross hydrolysis process. Thus, the design of the separation process is necessary to reduce the effect of the gas emission from aluminium dross treatment. Therefore, this research aims to propose alternative processes for aluminium dross reduction and determine optimal process parameters via simulation study using Aspen Plus. Aluminium dross from CHJ metal company in Thailand is used as raw material. The operation in a laboratory scale is performed to find optimal parameter such as concentration for process design. The suitable process in an industrial scale is proposed by preforming an economic analysis.

2. Experimental Study

Firstly, 20 g of waste aluminium dross from CHJ metal company in Thailand with an average particle size of 75-250 μm was used as raw material and characterized by using X-ray diffraction (XRD). Then, an experiment of aluminium dross treatment by hydrolysis in basic solution and the acid solution was set. Chemical composition that was not founded by XRD was determined by the gas probe. Next, the waste gas separation process in a laboratory scale was carried out as shown in Fig. 1. For the first experimental set, the aluminium dross of 20 g was hydrolysed in a flask (1) in the presence of sodium hydroxide solution with the different concentrations of 0.01 M, 1 M and 2 M. The temperature was kept at 25 ^{o}C for 2 h. Ammonia gas was generated and trapped in four bottles (2) using the solvents such as water and HCl of 0.01 M. Phenolphthalein was used as an indicator to identify trapped gas. For the second experimental set, the hydrolysis in acid solution (1) was performed. Hydrogen sulphide (H_2S) and phosphine (PH_3) were generated during hydrolysis. $KMnO_4$ solution with various concentration was used as an agent for H_2S stripping (2). Ethylene glycol is used for PH_3 stripping. The amount of all gases were determined by replacing water in cylinder (3).

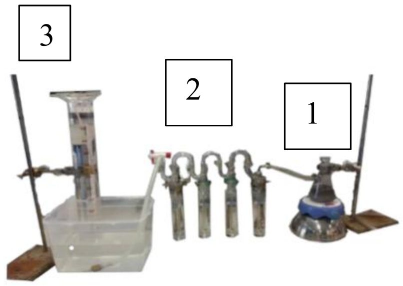

Fig. 1 Experimental apparatus for aluminium dross hydrolysis.

From the XRD result, the percentage of chemical composition aluminium dross are 52.6% of aluminium nitrile (AlN), 22.4% of magnesium aluminium oxide ($MgAl_2O_4$), 16.1 % of brownmillerite (Ca_2FeAlO_5) and 8.9% of aluminium oxide (Al_2O_3). In the laboratory scale, hydrolysis in basic condition generates NH_3 as toxic gas of 260 ppm. One bottle distilled water with 25 ml can trap NH_3 as in NH_4OH form. However, NH_3 can evaporate within 1 day. Thus, HCl solution is preferred to distilled water. The effect of concentration and amount of HCl are studied. For example, 0.01 M of HCl in a solution 100 ml of HCl can react with NH_3 to form NH_4Cl. For hydrolysis in acid solution, NH_3 is not detected but hydrogen sulfide and phosphine are formed. Other solvent, such as toluene, $CuSO_4$, $Al(NO_3)_3$, $KMnO_4$, and ethylene glycol, are examined. The experimental results show that only $KMnO_4$ solution can trap H_2S and ethylene glycol can be used as solvent for phosphine gas stripping because it dissolves in polyhydric alcohol such as ethylene glycol. The toxic gases are completely trapped in the solvents under the suitable condition in the laboratory scale as shown in Table 1.

Table 1. Suitable conditions for gas treatment.

Hydrolysis condition	Final gas generation (ml)	Type of toxic gas	Toxic gas concentration (ppm)	Trapping solution	Trapping volume (mL)
Basic	540 ± 15	NH_3	260 ± 5	HCl 0.01M	100
Acid	810 ± 17	H_2S	300 ± 10	$KMnO_4$	200
		PH_3	280 ± 5	$C_2H_6O_2$	560

3. Design of Aluminum Dross Process

In this section, the aluminium dross at the flow rate of 1 ton/day is used as a basis for process design. In an industrial scale, three processes are performed based on types of gas formation during the hydrolysis of aluminium dross in basic or acid solution. All processes consist of aluminium dross reduction, gas separation and production from residue waste. In the simulation, the electrolyte NRTL model is employed to predict thermodynamic properties and phase equilibrium of weak electrolyte. The first process as shown in Figure 2a is the hydrolysis in the presence of NaOH solution integrated with water absorption tower. Aluminium dross and NaOH solution is fed into a hydrolysis reactor represented by the RSTOIC reactor model in Aspen Plus. The hydrolysis reaction completely generates ammonia gas as shown in eq (1). The effluent stream from the reactor is sent to a filtration for salt cake removal (i.e., aluminium oxide). Afterward, liquid product, i.e., sodium aluminium hydroxide, enters a flash separator for ammonia removal. It can be converted to sodium aluminate ($Na[Al(OH_4)]$) by calcination using a heater. The ammonia gas is passed at the bottom of an absorber. The two stages of absorber are required. The stream 6 is the solvent. Water is used for the first process, so ammonia can dissolve in water to produce ammonium ion and hydroxide ion. This reaction is ammonia dissociation. Thus, pure air is obtained at the top of column.

$$\mathrm{AlN} + 3\mathrm{H_2O} \rightarrow \mathrm{Al(OH)_3} + \mathrm{NH_3} \tag{1}$$

The second process is similar to the first one. However, HCl solution is used as a solvent instead of converting ammonia to ammonium chloride (NH_4Cl) at room temperature. The reaction in eq (2) is performed in an absorber.

$$\mathrm{NH_3} + \mathrm{HCl} + \mathrm{H_2O} \leftrightarrow \mathrm{NH_4Cl} + \mathrm{H_3^+O} \tag{2}$$

The third process involves the aluminium dross hydrolysis in acid condition as shown in Figure 2b. Hydrogen sulphide and phosphine are generated during the reaction as shown in eq (3)–(4). Then, all of the substances are moved to a filter for salt cake separation. The liquid product is separated from gas by using a flash separator. Afterward, NaOH is added for neutralization and aluminium oxide is generated and sent to furnace, resulting in a solid waste. Hydrogen sulfide elimination is carried out in the reactor by using $KMnO_4$ solution as shown in eq (5). Sulfur is formed and separated by the separator. The last step is phosphine elimination that is trapped by ethylene glycol solution at room temperature. Thus pure air is obtained at the top of column.

$$Al_2S_3 + 6HCl \rightarrow 3H_2S + 2AlCl_3 \tag{3}$$

$$AlP_3 + 3HCl \rightarrow 3PH_3 + 2AlCl_3 \tag{4}$$

$$3H_2S + 2KMnO_4 \rightarrow 2S + 2H_2O + KOH + 2MnO_4 \tag{5}$$

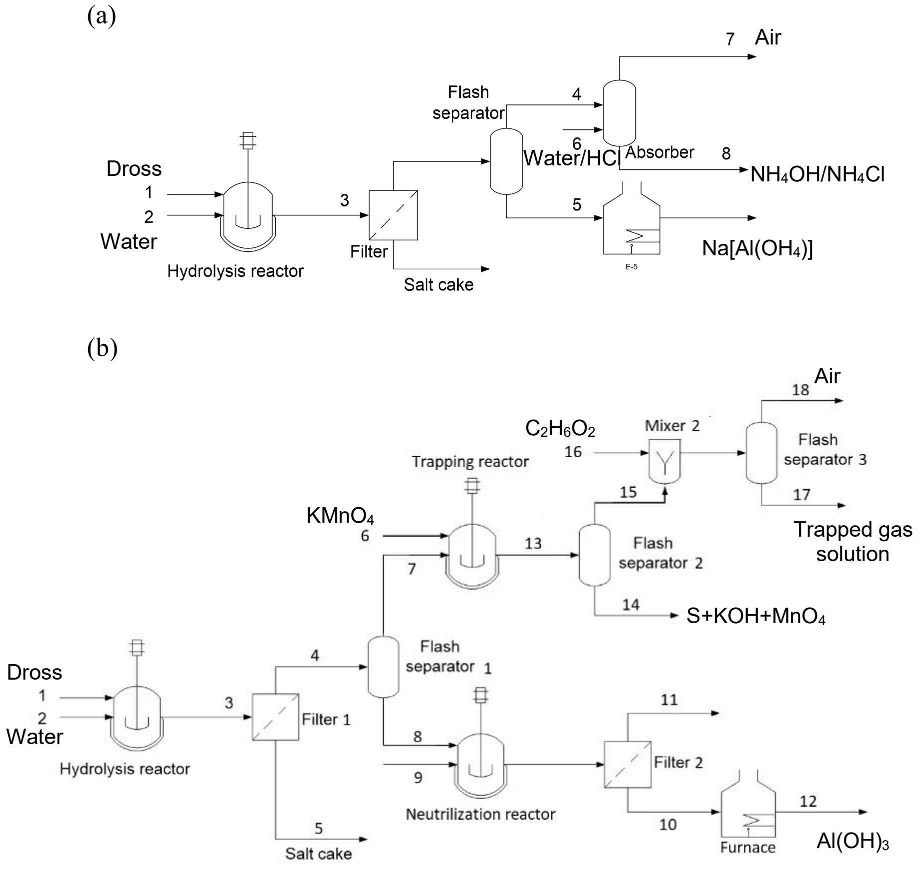

Fig. 2 (a) Hydrolysis aluminium dross in NaOH solution integrated with (1) water stripping of the first process (2) HCl stripping of the second process and (b) hydrolysis aluminium dross in NaOH solution integrated with H_2S and PH_3 stripping of the third process.

4. Results and Discussions

4.1. Parametric Analysis

Fig. 3a shows that NH_3 can be eliminated with an increase in water flow rate. NH_3 with a high concentration in the gas phase can dissolve in water. Ammonia which is the weak base reacts with water to form ammonium hydroxide. The simulation results show that ammonium ion and hydroxide ion are increased with an increase in water flow rate. The water flow rate of 2.2 l/min leads to complete NH_3 removal. Fig 3b shows the NH_3 removal using HCl stripping. An increase in HCl flow rate and concentration has a positive effect on NH_3 removal. Ammonium ion and chloride ion are found in the solution. Concentration of HCl 0.01 mol/l is selected because it can react with NH_3 easily at the room temperature to form NH_4Cl. Fig 3c shows H_2S removal by using $KMnO_4$ with the concentration of 1 mol/l. It is eliminated with an increase in $KMnO_4$ flow rate. This is because increasing $KMnO_4$ flow rate results in high reaction rate. However, increasing the flow rate of $KMnO_4$ higher than 100 l/min shows an opposite trend. As the reaction occurs in the liquid phase, a low resident time of H_2S solutes in the liquid phase is found at high $KMnO_4$ flow rate. Fig 3d presents the PH_3 removal by using ethylene glycol as a solvent; it is eliminated with an increase in ethylene glycol flow rate. It is noted that PH_3 can solute in the liquid phase at the mole fraction of 0.025 under the atmospheric pressure according to Henry's law (Gerrard, 1980). PH_3 is removed completely when the flow rate of ethylene glycol is 4.2 l/min. The results imply that the toxic gas separation process can be scaled- up in an industrial process.

4.2. Economic Assessment

Mass and energy balances of the optimally designed aluminium dross process are used for economic analysis. The project life time is assumed to be 10 years, and interest rate of 7% and operating hours of 8 hours/day are also assumed. The total cost and economic indicators such as net present value and payback year are shown in Table 2. The third process provides the highest total production cost that depends on the raw material cost of 77% while the first and second processes depends on the raw material cost of 45%. This is because the toxic gases, such as H_2S and PH_3, are formed in the third process. So two solvents for gas trapping are required. Furthermore, energy consumption for hydrolysis in acid solution and dry neutralized product are high. The residue waste from the third process need to be eliminated whereas the first process and second process produce $Na[Al(OH_4)]$ that can be used in widespread applications. The positive net present value of the first and second processes indicates project earnings. Although the first process is the most economical process in terms of net present value and payback period, the second process is preferred because NH_4Cl is formed and ammonia gas cannot release to the environment.

5. Conclusions

In this study, three processes for treatment of second aluminium dross coupled with toxic gas separation were proposed. Toxic gases are completely eliminated by stripping process using chemical solvents. Although hydrolysed aluminium dross coupled with water stripping performs the highest present value and payback period. The hydrolysed aluminium dross coupled with gas stripping using hydrochloric acid offers more the benefits due to no toxic gas emission. The results show that the concentration of hydrochloric acid of 0.01 mol/l can trap ammonia gas in an absorber. In addition, it can produce high value-added products such as sodium aluminate and ammonium chloride.

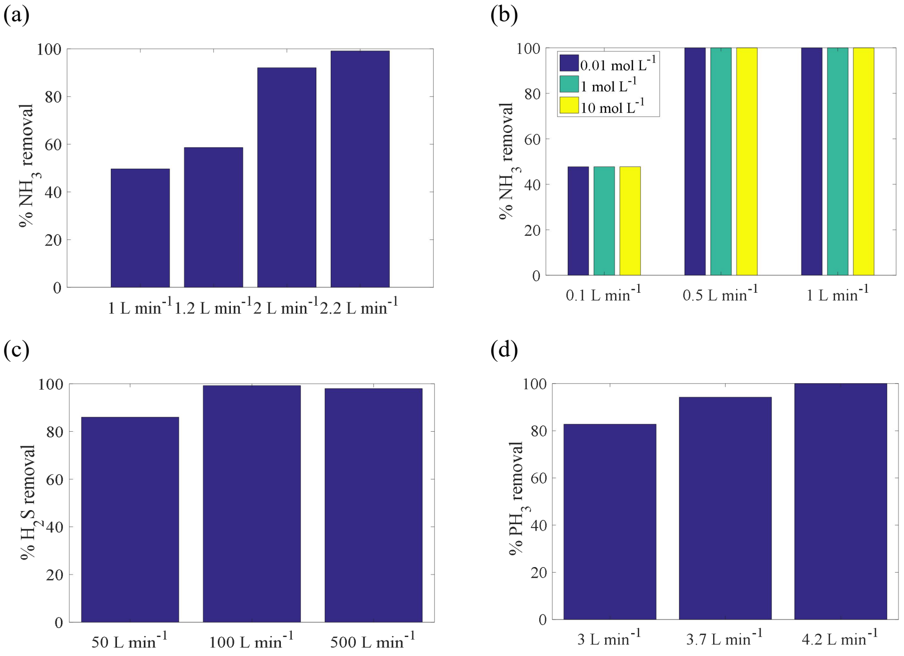

Fig. 3 Effect of solvent on percentage of toxic gas removal (a) NH_3 removal using water stripping (b) NH_3 removal using HCl stripping (c) H_2S removal (d) PH_3 removal.

Table 2 Total cost and economic indicator.

	Process 1	Process 2	Process 3
Total investment cost ($)	3.84x10^5	4.19x10^5	4.68x10^5
Total production cost ($)	1.86x10^5	1.91x10^5	7.41x10^5
Net present value ($)	6.19x10^5	5.45x10^5	-5.6x10^6
Payback period (year)	2.69	3.04	N.A.

6. References

Calder G.V., Stark T.D. (2010). Aluminum reactions and problems in municipal solid waste landfill. *Practice periodical of hazardous, toxic, and radioactive waste management*, 258 – 265.

El-Melih, A.M., Shoaibi, A. Al., and Gupta, A. K. (2016). Hydrogen sulfide reformation in the presence of methane. *Applied Energy*, 178, 609- 615.

Gerrard, W. (1980). Gas solubilities widespread applications. Pergamon Press 1st edition.

Tsakiridis, P. (2012) Aluminium salt slug characterization and utilization-A review. *Journal of Hazardous materials*, 217, 1-10.

Singh K.K., Meshram A., Gautam D., and Jain A. (2019). Hydrogen production using waste aluminium dross: from industrial waste to next-generation fuel. *Agronomy research*, 17, 1199-1206.