SYNTHESIS OF ALUMINA FROM ALUMINIUM CAN USING DISSOLUTION PROCESS

BY

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A dissertation submitted in fulfilment of the requirement for the degree of Master of Science (Materials Engineering)

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ABSTRACT

The increasing utilization of aluminum in daily life has produced a rise in aluminium scrap. At present, the method used to process aluminium scrap is smelting process which consumes high levels of energy and causes environmental problem. This research focuses on the development of an alternative technology to process waste aluminium can into high added value of products without releasing any waste materials. In this study, aluminium can was treated using an alkaline solution in a closed reactor. The aluminum dissolved easily in the solution and gas was spontaneously released. The results revealed that the process was mainly controlled by temperature and concentration. The higher the temperature and concentration, the faster the dissolution process and hydrogen generation. At the end of the process, the solution was filtrated and calcined at 800°C and 1000°C based on temperature that obtained from TG/DTA result. X-ray diffraction analysis confirmed that the filtrate sample was aluminium hydroxide, and the calcined filtrate contained of delta and theta alumina. Subsequently, particle size distribution showed that higher temperatures of calcination led to the production of finer alumina particles where the original filtrated sample had a particle size within the range of 30µm to 32µm while the calcined filtrate had a finer particle size within the range of 22µm to 23µm. On the other hand, the existence of hydrogen which was released during the dissolution process was verified through gas chromatography analysis. Energy analysis showed that there was no waste released throughout the process. Instead, the waste was utilized as the main source and converted into added value of products which was alumina powder and hydrogen gas by-products. Based on the results, the proposed process is efficient as it is environmentally safe, produces a green energy source and converts waste into valuable feedstock materials.

خلاصة البحث

أنتجت زيادة استخدام الألمنيوم في الحياة اليومية ارتفاعا في خردة الألومنيوم. في الوقت الحاضر، التكنولوجيا المستخدمة لمعالجة الخردة هي عملية تقليدية التي تستهلك مستويات عالية من الطاقة، وتسبب مشاكل بيئية. هذا البحث يركز على تطوير التكنولوجيا البديلة لمعالجة نفايات الألومنيوم التي تمكن من ارتفاع القيمة المضافة للمنتجات من دون الإفراج عن أي مواد للنفايات. في هذه الدراسة، عولج الألومنيوم باستخدام محلول قلوي في مفاعل مغلق. الألومنيوم تحلل بسوهلة في المحل و ادا الى اطلاق غاز .وكشفت النتائج التي كانت أن العملية كانت تسيطر بشكل رئيسي من قبل درجة الحرارة والتركيز. ارتفاع درجة الحرارة والتركيز يؤدي إلى أسراع عملية الانحلال وتوليد الهيدروجين. في نهاية هذه العملية، كان الحل مرشح ومكلس في 800^o درجة مئوية و 1000^o درجة مئوية. تحليل الحيود من الأشعة السينية أكدت أن نموذج الترشيح هو هيدروكسيد الألومنيوم، والترشيح الوارد الألومينا المكلس نقية من الألومينا دلتا وثيتا. في وقت لاحق، حجم الجسيمات أظهرت أن ارتفاع درجات الحرارة للتكليس أدى إلى إنتاج الجسيمات الدقيقة الألومينا. وتم التحقق من ذلك من خلال الملاحظة البصرية بالمجهر لعينة الرشاحة الأصلية التي تحتوي على حجم الجسيمات داخل نطاق 30µm إلى 32µm بينما في رشاحة المكلس كان حجم الجسيمات الدقيقة ضمن نطاق من 22µm إلى 23µm . من جهة أخرى، تم التحقق من وجود الهيدروجين الذي صدر أثناء عملية حل من خلال التحليل اللوني للغاز. وأظهر تحليل الطاقة أنه لا يوجد أفرج للنفايات طوال هذه العملية. بدلا من ذلك، تم استخدام النفايات كمصدر رئيسي وتحويلها إلى القيمة المضافة للمنتجات من مسحوق الألومينا وغاز الهيدروجين النقي من المنتجات. بناء على النتائج، هذه العملية المقترحة فعالة من حيث أنها آمنة بيئيا، تنتج طاقة نقية، وتحول النفايات إلى مواد وسيطة قيمة.

APPROVAL PAGE

I certify that I have supervised and read this study and in my opinion, it conforms to acceptable standards of scholarly presentation and is fully adequate, in scope and quality, as a thesis for the degree of Master of Science (Materials Engineering).

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Dean, Kulliyyah of
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DECLARATION

I hereby declare that this dissertation is the result of my own investigation, except where otherwise stated. I also declare that it has not been previously or concurrently submitted as a whole for any other degrees at IIUM or other institutions.

Wan Roslina Binti Wan Mohd

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LIST OF ABBREVIATIONS

XRD	X-Ray Diffraction
GC	Gas Chromatography
TGA	Thermogravimetric Analysis
DTA	Differential Thermal Analysis
GHG	Greenhouse gas
SMR	Steam Methane Reforming
WGS	Water Gas Shift
HTS	High Temperature Shift
LTS	Low Temperature Shift
TG/DTA	Thermogravimetric Differential Analysis
GJ	Gigajoule
μm	Micrometer
μV	Microvolt
mA	Milliampere
kV	Kilovolt
kg	Kilogram
rpm	Revolution per minute
mm	Millimeter
ml	Milliliter
g	Gram
L	Liter
M	Molarity
°C	Degree celcius
K	Kelvin
kJ/mol	Kilojoule/mol
ΔH	Change in Enthalpy
ΔG	Change in Gibb's free energy
ΔT	Change in temperature
Cp	Specific heat
P	Pressure
V	Volume
M	Molarity
n	Mole
R	Ideal gas constant
T	Temperature
Q	Energy
m	Mass
c	Specific heat capacity
H_2SO_4	Sulfuric acid
CO	Carbon monoxide
CO_2	Carbon dioxide
Al_2O_3	Alumina
NaOH	Sodium hydroxide

$Al(OH)_3$	Aluminium hydroxide
H_2O	Water
Al	Aluminium
AlOOH	Boehmite
$NaAl(OH)_4$	Sodium aluminum hydroxide
$Na_4P_2O_7.10.H_2O$	Tetrasodium pyrophosphate

CHAPTER 1

INTRODUCTION

1.1 OVERVIEW

During the past 100 years, aluminium has developed into staple of the manufacturing industry. Moreover, aluminium usage has increased rapidly since its usefulness was discovered, and the presence of aluminium can now be seen in almost every sector in the industry. This material has unique qualities and an almost endless range of applications, especially for transportation. This includes the rail industry (subway coaches, sleeping cars), the shipping industry (civil and military hydroplanes, motorboats) and many more (Mazzolani, 1995). Year after year, the increasing demand for aluminium has led to the production of a lot of waste or scraps from the automotive, airplane, household and packaging industries.

At present, aluminium scrap is conventionally processed via smelting. This method has been used to process aluminium scrap in many aluminium manufacturing countries such as India, China and Philippines. In the smelting process, a reverberatory or rotary furnace can be used to melt the scrap in the presence of chloride-based slag (Hwang et al., 2006). Although the end of this process will produce non-metallic by-product residue, it is still waste that will be disposed at a landfill which can indirectly cause severe pollution to the environment (Kulik & Daley, 1990).

Alternatively, there is another relatively effective method where aluminium scrap is smelted in a rotary drum or hard smelting furnace in the presence of salt slag. However, its enormous disadvantage lies in the fact that the gases generated from the process are subjected to serious pollutants. The salt slag itself is extremely dangerous. Therefore, the operation must be carried out under stringent safety precautions (Gottschol, 1977).

Smelting aluminium scrap creates several limitations such as high energy consumption, high operating costs and large numbers of operations. Based on estimations, nearly 16 to 19 GJ/t of energy is consumed during this process (Gronostajski & Matuszak, 1999). Furthermore, smelting byproducts are highlighted by the industry as a disposal problem that causes manufacturers to spend millions of dollars in landfill costs, and consequently leads to environmental issues. The amount of waste that is released from this process also needs further processing to create added value to the product and to enhance the bottom line of the aluminium industry economically (Mazzolani, 1995).

Currently, a valuable product that can be generated from aluminium is alumina. Alumina and aluminum-based compounds have a wide range of applications in many areas. Particularly as special hydrates, their application covers a variety of products such as chemicals, pharmaceuticals, catalysts, plastic and pigments, synthetics substitutes, papers and alumina ceramics, refractories, insulators, abrasives, porcelain, electronics and many more. Furthermore, alumina provides some of the special properties such as high hardness, high mechanical strength, high thermal conductivity and good thermal shock resistance. Many attempts has been made to produce alumina either by utilizing waste aluminium or others commercial sources of aluminium as a main raw material. Lòpes et al. (2011) has been utilized a hazardous waste generated in slag milling process by the aluminium industry as a main raw material for the synthesis of alumina. This process consisted of two steps: in the first one, nearly 90% of aluminium present in the waste is recovered as a nanocrystalline boehmite, γ-AlOOH by hydrothermal treatment of the waste. In the second step, the boehmite was calcined at temperature of 1400ºC to convert it into alumina. In other work, Pratima (2002) reported that alumina also can be produced by using concentrated solutions of aluminum salt and urea. In addition, different grades alumina is produced at varying temperature of calcination ranging from 600°C to 1400°C and suitable used for polishing and grinding application. In this work, alumina is obtained through synthesis it from waste aluminium can using dissolution process.

The entire process is schematically shown in Figure 1.1. In this process, waste aluminium can was fed into a reaction reactor containing solution of sodium hydroxide (NaOH) which was heated at desired temperature. Aluminium reacted with NaOH and produce hydrogen gas and solution. The solution was then cooled down to get precipitation of aluminium hydroxide ($Al(OH)_3$). Hydrogen gas would be transferred to a gas tank while the precipitated $Al(OH)_3$ would undergo filtration. After filtration, water (H_2O) and NaOH would be recycled back into the reactor. The precipitated $Al(OH)_3$ would undergo calcination for thermal decomposition and the final product produced is alumina powder (Al_2O_3).

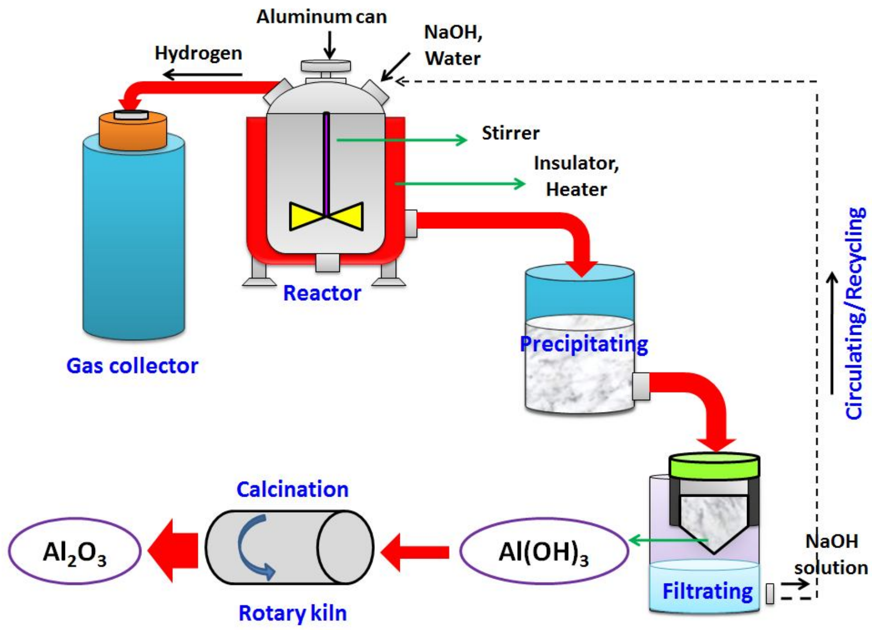

Figure 1.1: Flow diagram of synthesis of alumina from aluminium can

1.2 PROBLEM STATEMENT AND ITS SIGNIFICANCE

Increasing amount of waste generation in Malaysia is forcing local authorities to continually seek new management strategies to overcome this major problem, especially regarding aluminium waste. One conventional method used to process aluminium waste such as can, scrap and foil is the smelting process. However, this process does have its limitations such as high energy consumption where the main source to melt aluminium waste in smelting process is electricity. Furthermore, high temperature required to melt aluminium waste is between 800°C to 1000°C. Other than that, smelting process will produce aluminium ingot and slag as a final product. Aluminium ingot has a low added value of the product because it needs further processing to produce aluminium product. Moreover smelting process generates slag which is characterized as a by-product of smelting process that will be disposed to the landfill.

Currently, a few processes have been proposed to utilize waste aluminium into valuable product especially aluminium oxide (Al_2O_3) which is commonly called as alumina or hydrogen gas through dissolution process in chemical reaction. The main principal idea of these processes using dissolution as a main method. For example, Seung & Ho (2007) had utilized aluminium can by dissolving it in sulfuric acid solution in order to produce aluminium ethoxide powder. There is some limitation from this process where sulfuric acid itself is dangerous and need extra precaution during handling it. Moreover, the solution cannot be repeatedly recycled and it will release as a waste materials. Other previous research had done by Martines et al. (2005) where aluminium can is dissolved in alkaline solution to produce one product which was hydrogen gas only. The solution that containing precipitate of $Al(OH)_3$ did not bring to the further processing to produce alumina. Instead, the solution was released as a waste. Furthermore, this work used concentrations of alkaline solution which were about 1.10M, 1.5M and 2M where these concentrations were higher compared with proposed process. Soler et al. (2005) done a research which was focused on hydrogen generation only via dissolving commercial aluminium foil in alkaline solution at concentrations of 0.1M, 1.0M, 5.0M and at temperature between 70°C to 80°C. These experimental conditions for dissolution process also were higher than proposed process. Other than that, they used commercial aluminium as a main source instead of utilizing waste aluminium.

Based on these previous researches, it can be observed that those processes provide some limitations such as release waste, only one product is produce either alumina powder or hydrogen gas, using higher experimental condition for dissolution (temperature and concentration) and there is no energy analysis has made to analysis how much energy had been utilized throughout the process. Therefore, this research has been proposed to utilize waste aluminium can into valuable feedstock materials which were alumina powder as main product and hydrogen gas as a byproduct without release any waste materials at low temperatures and concentrations. Instead of that, energy analysis of the overall process also was evaluated. Furthermore, this process will give benefit to industry and promote a green technology without releasing any waste materials.

1.3 RESEARCH OBJECTIVES

The main objectives of this work are to synthesis alumina from waste aluminium can through dissolution process in alkaline solution without releasing any waste. Other than that, hydrogen gas also is aimed produced as a byproduct during the reaction. In order to achieve this objective, several aims need to be addressed:

i. To investigate the effect of experimental condition of the dissolution process. The parameters involved in this process are:
   a. Temperature
   b. Concentration of solution
ii. To evaluate the effect of calcination temperature on the properties of alumina product
iii. To evaluate the energy balance of overall process

1.4 RESEARCH METHODOLOGY

In order to achieve all the objectives specified, a well-planned methodology is required which is outlined below.

1. Planning and experimenting to produce alumina powder (Al_2O_3) and evaluation of their properties which include

i. Preparation of raw materials

Waste aluminium can was cleaned and dried in room temperature. After that, aluminium can was burnt using gas burner to remove polymer coating on the surface. Aluminium can was measured based on desired dimension and then it was cut into the pieces.

ii. Dissolution

This section involved the preparation of aqueous solution of sodium hydroxide (NaOH) which has different concentrations (0.19M, 0.25M, 0.38M and 0.5M). Other than concentration, the main parameters that also involved in this section are temperatures (35ºC, 40ºC, 45ºC and 50ºC). These two important parameters controlled the experimental condition during the dissolution process.

iii. Precipitation and filtration

After the reaction was stopped, the solution was cooled down to allow precipitation occured. The precipitate product was then subjected to filtration.

iv. Calcination of filtrated sample

The filtrate product will undergo to calcination. In this section, the control parameter is temperature. The effect of temperature to the properties of the filtrate product was investigated.

v. Characterization of filtrate sample

Characterizations of filtrate product are performed by X-Ray Diffraction (XRD), Thermogravimetric-Differential Analysis (TG/DTA), Particle Size Distribution, and Gas Chromatography (GC). The aims of these characterizations are to determine the phase identification that presence in filtrate product, measuring the particle size, and to verify the existence of hydrogen from the gas which are released during the dissolution.

2. Energy analysis

   Internal energy, work and heat involved during the reaction and throughout the process were evaluated based on the first law of thermodynamic. The research methodology throughout the process is simplified in a flow chart as shown in Figure 1.2.

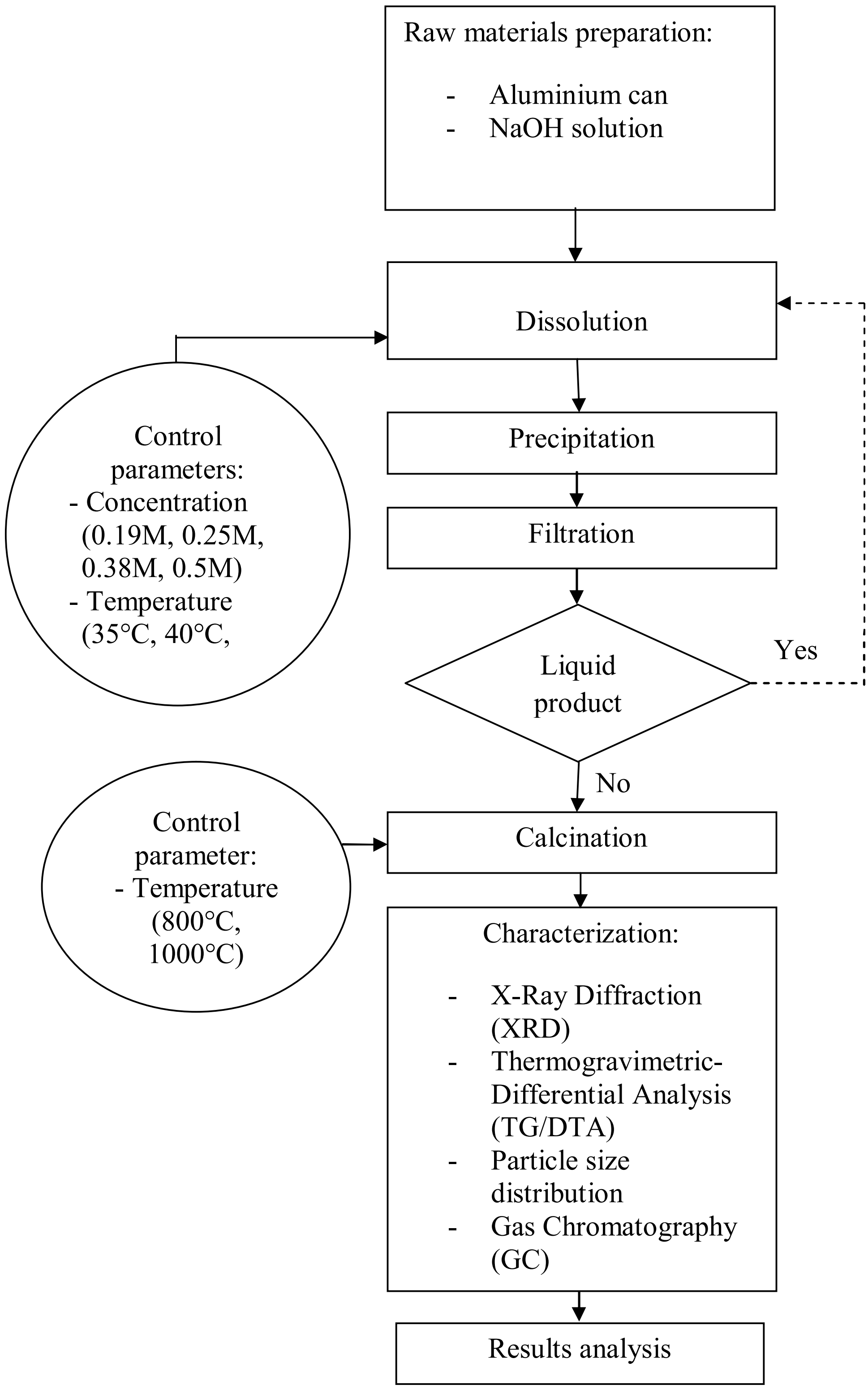

Figure 1.2: Research methodology in a flow chart

1.5 SCOPE OF RESEARCH

This research focused on the synthesis of alumina powder from aluminium can waste. Instead of that, generation hydrogen gas as a byproduct also was expected. The effect of experimental condition of the dissolution process for aluminium was investigated as well as the effects of calcination temperature on the properties of alumina powder. An energy balance of the overall process was also be evaluated.

1.6 DISSERTATION ORGANIZATION

This dissertation is organized into five chapters. Chapter One gives an overview of the research. Chapter Two describes a brief review of recent literature related to aluminium scrap, benefits of recycling aluminium, production alumina from dross, thermodynamic of the dissolution of aluminium and hydrogen production. Chapter Three highlights the experimental details conducted throughout this work including the preparation of raw materials, dissolution, precipitation and filtration, calcination of filtrate sample, characterizations of filtrate sample and energy analysis. Chapter Four presented the results and discussion of findings throughout this work. Chapter Five provides a general conclusion and recommendations for future research in this area.