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CONVERSION OF PLASTIC WASTE INTO FUEL USING MICROWAVE-PYROLYZER WITH THE ADDITION OF ALUMINIUM DROSS AS A CATALYST

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CONVERSION OF PLASTIC WASTE INTO FUEL USING MICROWAVE-PYROLYZER WITH THE ADDITION OF ALUMINIUM DROSS AS A CATALYST

HAZELIANA BINTI HENRY LITONG

A dissertation submitted in partial fulfilment

of the requirement for the degree of

Bachelor of Engineering with Honours

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I would like to dedicate my thesis to my beloved family and friends.

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ABSTRACT

Aluminium dross is a by-products produced from the smelting process of aluminium. It has the potential of risking the environment as it will release hazardous gaseous when it is in contact with moisture such as ammonia, methane and hydrogen. In order to reduce the harmful effect of aluminium dross to the environment, it can be treated to become catalyst by undergoing physical as well as chemical processes by recovering aluminium hydroxide ($Al(OH)_3$). Aluminium dross undergoes leaching process in 500 ml of sodium hydroxide (NaOH) at 10% concentration. The second stage was precipitation using hydrogen peroxide (H_2O_2) followed by calcination in a furnace at 600 ˚C for 3 hours. The characteristics of the untreated aluminium dross and treated aluminium dross have been studied using Brunauer-Emmett-Teller (BET) analysis. BET result showed an increase on the surface area of aluminium dross from 10.06 m^2/g to 74.21 m^2/g after treatment. The treated aluminium dross was used as a catalyst in converting plastic into liquid hydrocarbon in a modified microwave pyrolyzer with the condition of 1000W and 800W at 18 minutes and 15 minutes. The result is 1% of liquid obtained from the pyrolysis of plastic waste at the condition of 1000W for 15 minutes and 3% liquid at the condition 800W for 18 minutes. Microwave pyrolysis was also conducted without the presence of catalyst. The result is 1% liquid at the condition 1000W for 20 minutes and 3% liquid at the condition of 800W for 20 minutes.

ABSTRAK

Keladak aluminium adalah hasil sampingan yang terhasil daripada proses peleburan aluminium. Ia mempunyai potensi merisikokan alam sekitar kerana ia akan membebaskan gas berbahaya apabila ia bersentuhan dengan lembapan seperti ammonia, metana dan hidrogen. Bagi mengurangkan kesan bahaya keladak aluminium terhadap alam sekitar, ia boleh dirawat untuk menjadi pemangkin dengan menjalani proses kimia serta fizikal bagi memperolehi aluminium hidroksida ($Al(OH)_3$) Proses kimia tersebut bermula melarutkan keladak aluminium dengan 500 ml natrium hidroksida (NaOH) pada kepekatan 10%. Peringkat kedua adalah pengurasan menggunakan hidrogen peroksida (H_2O_2) diikuti oleh pengkalsinan dalam relau pada 600˚C selama 3 jam. Pencirian keladak aluminium sebelum dan selepas dirawat telah dikaji menggunakan Brunauer-Emmett-Teller (BET) analisis. Keputusan BET menunjukkan peningkatan kawasan permukaan daripada 10.06 m^2/g ke 74.21 m^2/g selepas rawatan. Keladak aluminium terawat telah digunakan sebagai pemangkin dalam menukarkan bahan plastik kepada cecair hidrokarbon dalam pyrolyzer gelombang-mikro yang diubahsuai dengan keadaan 1000W dan 800W selama 18 minit dan 15 minit. Keputusan yang didapati ialah 1% cecair dalam keadaan 1000W selama 15 minit dan 3% cecair dalam keadaan 800W selama 18 minit. Pirolisis gelombang-mikro telah dijalankan tanpa kehadiran pemangkin. Keputysan yang didapati ialah 1% cecair dalam keadaan 1000W selama 20 minit dan 3% cecair dalam keadaan 800W selama 20 minit.

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LIST OF ABBREVIATIONS

AD	-	Aluminium Dross
AlO(OH)	-	Aluminium Oxyhydroxide
$Al(OH)_3$	-	Aluminium Hydroxide
Al_2O_3	-	Aluminium Oxide
$Al_2O_3 \cdot 3H_2O$	-	Trihydrate Aluminium Hydroxide
Au	-	Gold
BET	-	Brunauer-Emmett-Teller
CAD	-	Calcined Aluminium Dross
DBKU	-	Dewan Bandaraya Kuching Utara
Fe_2O_3	-	Iron Oxide
FTIR	-	Fourier Transform Infrared
GCMS	-	Gas Chromatography-Mass Spectrometry
HDHPE	-	High Density Homopolymer Polyethylene
HDPE	-	High Density Polyethylene
H_2O_2	-	Hydrogen Peroxide
LDPE	-	Low Density Polyethylene
LLDPE	-	Linear Low Density Polyethylene
MBKS	-	Majlis Bandaraya Kuching Selatan
MDPE	-	Medium Density Polyethylene
MgO	-	Magnesium Oxide

Na_3AlF_6	-	Sodium Aluminium Fluoride
NaOH	-	Sodium Hydroxide
PAHs	-	Polycyclic Aromatic Hydrocarbons
PE	-	Polyethylene
PP	-	Polypropylene
RAD	-	Raw Aluminium Dross
SUD	-	Sustainable Urban Development
SEM	-	Scanning Electron Microscopy
SiO_2	-	Silicon Dioxide
TAD	-	Treated Aluminium Dross
TGA	-	Thermogravimetric Analyser
UAD	-	Untreated Aluminium Dross
XRD	-	X-Ray Fluorescence

CHAPTER 1

BACKGROUND OF STUDY

1.1 Overview

Aluminium dross is a by-products produced from the smelting process of aluminium (Hwang, Huang, & Xu, 2006). It is a major by-product of all processes which comprises molten aluminium (Drouet, Leroy, & Tsantrizos, 2000). The chemical compositions of dross are aluminium oxides, metallic aluminium, nitride, salts and several alloy (Dai, 2012). It can be divided into white dross and black dross in which black dross contain salt whereas white dross does not contain any salt (Dai, 2012). Aluminium dross has the potential of risking the environment as it will release hazardous gases when it is in contact with moisture such as ammonia, methane and hydrogen (Miskufova, Petranikova, & Kovacs, 2009). One of the alternatives in reducing this waste is by processing it in a rotary kiln to retrieve aluminium which result in salt cake that will be sent for landfilling (Dai, 2012). This in turn, could harm the environment due to the potential leaching of the salt cake (Dai, 2012).

Aluminium dross can be used as a filler in concrete and asphalt products to enhance stiffness abrasion resistance as well as controlling micro-cracking (Wastes & Study, 2007). Another use of dross is it can be a substitute of sand or cement to produce concrete blocks (Ozerkan et al., 2014). Manufacturing of aluminate cement is also an application of dross as it contain salt cake which is a source of calcium oxide (CaO) and aluminium oxide (Al_2O_3) (Ozerkan et al., 2014).

Aluminium dross has the potential in becoming catalyst. It can undergo physical as well as chemical processes to be used in catalysis and filtration process (Yusoff, Muslim, & Paulus, n.d.). The dross catalyst can assist in microwave pyrolysis process of oil from plastic wastes (Lam & Chase, 2012). Meanwhile, plastic is a cheap and lightweight item which does not rot or rust with a reusable application (Panda, Singh, & Mishra, 2010). Thus, it is used widely nowadays. Panda et al., (2010) stated that plastic will be used continuously in the future. As the use of plastic increase, the amount of plastic as a waste will also increase which demands solution to decrease the waste. Therefore, plastic waste has been used in the plastic cracking process to be recycled into petrochemical materials (Buekens & Huang, 1998).

Accordingly, aluminium dross will be used in the catalyst cracking of plastic waste using microwave pyrolyzer to obtain the product of liquid hydrocarbons which is known as biodiesel.

1.2 Problem Statement

The by-product aluminium dross generation amount in Malaysia is within the range of 5000 to 10000 tonnes per month which is a huge amount. Problems arise as 95% of the waste by-product is land filled (Hong et al., 2010). Without proper treatment, the land filling activity of aluminium dross affects surface and groundwater (Hong et al., 2010). However, the recycling process through refining of the waste consumes a large amount of electricity which also require large amount of money (Hiraki et al., 2005).

Another recycling process of aluminium dross would be the resmelting. The by-product of smelting is white dross meanwhile resmelting produce a by-product of black dross along with salt cake (Gómez, Lima, & Tenório, 2008). Malaysia oblige disposal activities, storage and transportation of aluminium dross is carried out by contractors with license (Yusoff et al., n.d.). Yet, the disposal cost RM2000 per tonne as per licensed company (Yusoff et al., n.d.). This causes certain company to take cheaper route which is to landfill the waste. Hence, the need to treat or finding an alternative in managing the by-product is high. Furthermore, the recycling of dross is attractive from the energy and economic perspective because aluminium production uses large amount of energy to transform the material (Drouet et al., 2000).

Another issue related to waste disposal is plastic waste which is a problem faced by consumers globally. The increase of plastic usage is caused by the fact that it is much cheaper and have good physical properties as compared to other material such as glass and metal (Sarker et al., 2012). It is a waste which cannot biodegrade naturally (Buekens & Huang, 1998). Consequently, alternatives in managing plastic waste need to be done instead of land filled. Another problem faced with plastic waste is littering because plastic is available everywhere from being used in food packaging, to being used around the house. Littering then will cause drain clogging and also health problem towards animals (Sarker et al., 2012). Plastic waste management come with a cost and it demands an efficient way to make full use of the waste (Sarker et al., 2012).

In order to minimize the amount of plastic waste, solutions are required. A number of studies on the degradation of different type of plastic waste into liquid fuel using different catalysts have been carried out. However, many of them are impracticable due to cost of manufacturing (Panda & Singh, 2013). Thus, the potential of aluminium dross as a catalyst will be studied. The aluminium dross as a catalyst will be used in converting plastic waste into fuel. The main purpose of this study is to monitor the effect of time and power of microwave pyrolyzer in converting plastics into hydrocarbon oil.

1.3 Research Objectives

The main objectives of this study are as shown below:

i. To treat aluminium dross by undergoing the process of leaching, precipitation and calcination to be used as catalyst in converting plastic waste into liquid hydrocarbons
ii. To evaluate the properties of treated aluminium dross using Brunauer-Emmett-Teller (BET) and Scanning Electron Microscopy (SEM) analysis
iii. To assess the performance of treated aluminium dross as a catalyst in the conversion of plastic waste into fuel using microwave pyrolyzer

1.4 Research Scope

In this study, aluminium dross will be treated as a catalyst. The main component from dross to be used as a catalyst is aluminium hydroxide ($Al(OH)_3$). The step of treating aluminium dross into catalyst can be divided into three stages. The first stage is a leaching process of the dross in 500 ml of sodium hydroxide (NaOH) at 10% concentration. The second stage is precipitation using hydrogen peroxide (H_2O_2) followed by calcination in a furnace at 600 ˚C for 3 hours. The characteristics of the untreated aluminium dross and treated aluminium dross will be studied. Treated aluminium dross will be used as a catalyst in converting plastic waste high density polyethylene (HDPE) into hydrocarbon oil in a modified microwave pyrolyzer. The presence of catalyst in the pyrolysis process will increase the rate of chemical reaction without undergoing any changes on the catalyst itself. Thus, the yield of liquid hydrocarbons will increase.

CHAPTER 2

LITERATURE REVIEW

2.1 Aluminium Dross

Aluminium dross is categorised as schedule waste in Malaysia (Yusoff et al., n.d.). It is a by-product of the production of aluminium in which the formation is inevitable (Ünlü & Drouet, 2002). The melting of metal in a conservative purification or recycling method creates aluminium dross (AD) (Li, Zhang, & Yang, 2014). Electrolysis of alumina into aluminium is known as primary smelters and result in the formation of dross (Drouet et al., 2000). Meanwhile, the recycling method that produce aluminium dross occur at remelt plants which recycle aluminium scrap and beverage cans (Drouet et al., 2000).

The dross by-product can be obtained from primary aluminium production which will undergo three steps. The first step is bauxite mining, followed by aluminium production and finally the electrolysis (Tsakiridis, 2012). This first step is known as Bayer Process in which the standard process is to extract alumina from bauxite using caustic soda within elevated pressures and temperatures (Berdowski et al., 2009). The normal yield of aluminium oxide (Al_2O_3) is 35% to 55% from a tropical monohydrate bauxite grades (Baux & America, n.d.). Aluminium oxyhydroxide ($AlO(OH)$) can be found in caustic bauxite whereas aluminium hydroxide ($Al(OH)_3$) is a component in lateric bauxite (Baux & America, n.d.). Mining includes washing of the raw material bauxite to remove any solid impurities (Fitzgerald & French, 1971).

Next step is the aluminium production. This is where the process of aluminium oxide (Al_2O_3) extraction from the bauxite occur (Pahladsingh, 2004). Bauxite is a mixture of trihydrate aluminium hydroxide ($Al_2O_3·3H_2O$), iron oxide (Fe_2O_3) and silicon dioxide (SiO_2) (Fitzgerald & French, 1971). Reactions which involves in the extraction of Al_2O_3 is shown in Eq 1, Eq 2, Eq 3 and Eq 4 (Fitzgerald & French, 1971).

$$Al_2O_3 + 6NaOH + 3H_2O \rightarrow 2Na_3Al(OH)_6 \dots\dots\dots\dots\dots\dots (Eq\ 1)$$

$$SiO_2 + 4NaOH \rightarrow Na_4SiO_4 + 2H_2O \dots\dots\dots\dots\dots\dots\dots (Eq\ 2)$$

While Al_2O_3 and SiO_2 are soluble, Fe_2O_3 is insoluble in the solution due to its basic oxide and can be filtered out (Fitzgerald & French, 1971). The product from Eq 1 will then decomposed as shown in Eq 3.

$$Na_3Al(OH)_6 + 2H_2O \rightarrow 3NaOH + Al(OH)_3 \cdot 3H_2O \dots\dots\dots (Eq\ 3)$$

Heating of $Al_2O_3·3H_2O$ up to temperature above 1000˚C will extract Al_2O_3 as shown in Eq 4 (Fitzgerald & French, 1971).

$$2Al(OH)_3 \cdot 3H_2O \rightarrow Al_2O_3 + 9H_2O \dots\dots\dots\dots\dots (Eq\ 4)$$

The third step in primary aluminium production is electrolysis process, the electrolytic reduction of Al_2O_3. This process is also known as the Hall Heroult process (Pahladsingh, 2004). At the temperature of around 960˚C, electrolytic reduction of Al_2O_3 dissolved in molten cryolite, sodium aluminium fluoride (Na_3AlF_6) (Berdowski et al., 2009). This form an electrically conductive solution which result in Eq 5 and Eq 6 when undergoes reaction using consumable carbon anode with two concurrent reactions (Fitzgerald & French, 1971). This finally results in the formation of aluminium along with aluminium dross.

$$Al_2O_3 + 3C \rightarrow 2Al + 3CO \dots\dots\dots\dots\dots\dots\dots\dots\dots. (Eq\ 5)$$

$$2Al_2O_3 + 3C \rightarrow 4Al + 3CO_2 \dots\dots\dots\dots\dots\dots\dots\dots\dots (Eq\ 6)$$

Aluminium dross has the potential in affecting health. The dust, fumes and powder of aluminium dross can causes irritation towards the eyes, nose, throat and respiratory tract (“Material Safety Data Sheet,” 2008). Wet aluminium dross releases ammonia and cause moderate to severe eye irritation. The released ammonia can also cause moderate irritation towards the respiratory tract when inhaled (“Material Safety Data Sheet,” 2008). The presence of nickel in aluminium dross can cause respiratory tract and skin sensitization ("Material Safety Data Sheet," 2008). Chromium is also presence in aluminium dross. It can cause skin ulcers, dermatitis, perforation of the nasal septum, lung cancers, nasal cavity and as well as paranasal sinuses ("Material Safety Data Sheet," 2008).

2.1.1 Chemical Composition

The chemical composition of aluminium dross (AD) can be obtained using the X-Ray Fluorescence (XRF) analysis. The main composition of the dross is shown in **Table 2.1**. The highest component in AD is aluminium oxide with the mass fraction of 64.40% followed by silicon dioxide with the mass fraction of 7.40% which is comparable with mass fraction of other component such as magnesium oxide and sodium oxide.

Table 2.1 Chemical Composition of Aluminium Dross (Hong et al., 2010)

Component	Mass fraction (%)
Al_2O_3	64.40
SiO_2	7.40
MgO	6.40
Na_2O	6.20
Cl	5.80
CaO	2.70
Fe_2O_3	2.50
TiO_2	1.50
Others	3.10

Dai (2012) carried out calcination of dross at 1450˚C for an hour in air in his study to evaluate the equivalent oxide content of metallic elements. The result of the comparison is shown in **Table 2.2**. The major difference between raw aluminium dross (RAD) and calcined aluminium dross (CAD) is the composition. The major constituent of the CAD is aluminium oxide of 84% whereas the second major constituent is magnesium oxide (MgO).

Table 2.2 Composition Comparisons of Raw Dross and Calcined Dross (Lorber, 2012)

Raw Aluminium Dross (wt%)		Calcined Aluminium Dross (wt%)	
$MgAl_2O_4$	48	**Al_2O_3**	84
AIN	28	**MgO**	11
a- Al_2O_3	7	**SiO_2**	2
$(NO)_2Al_{22}O_{34}$	6	**CaO**	1
$NaAl_{11}O_{17}$	6	**Na_2O**	0.7
CaF_2	3	**K_2O**	0.4
Al	2	**Fe_2O_3**	0.3
		TiO_2	0.3

2.1.2 Physical Characterisation

The physical characteristic of aluminium dross can be classified into four classes which are the volume shrinkage, apparent porosity, cold crush strength and also permeability. It can be investigated by identifying the percentage of absorbent aluminium phyllosilicate, bentonite in the dross (Adeosun et al., 2014).

Aluminium dross which contain higher percentage of bentonite have a lower volume shrinkage while dross which contain more aluminium have a higher volume shrinkage (Adeosun et al., 2014). This behaviour can be explained by structural changes that occur due to thermal treatment (Adeosun et al., 2014). Bentonite is hydrophilic and the percentage of bentonite is directly proportional to water content of the dross (Adeosun et al., 2014). High water content means less air-pores space in aluminium dross which will be eliminated during firing and causes shrinkage (Adeosun et al., 2014). This also depicts the level of apparent porosity of the dross. Higher water content which caused by higher percentage of bentonite means lower porosity (Adeosun et al., 2014).

The cold crush strength of aluminium dross depends on the size of the particles. The larger the particle size, the lower the cold crush strength due to low binder content (Adeosun et al., 2014). Moreover, physical characteristic of aluminium dross can be measured by the permeability. Fine particles of dross promote de-agglomeration while large particles form gaseous pores (Adeosun et al., 2014). For both of the cases, the permeability increases.

2.2 Plastic Waste

Plastic wastes are categorized into two categories which are municipal and industrial plastic waste, depending on the origin of the waste, which indicates the difference in properties and qualities (Panda et al., 2010). As plastics are used in our daily lives with various of uses, the generation of this wastes increases significantly (Al-Salem, Lettieri, & Baeyens, 2009). Therefore, finding an alternative or a path in managing the waste is important.

Around 55 million tons of plastic waste generated by the USA, Europe and Japan (Brems et al., 2013). Narrower statistics which is in Kuching region located in Sarawak, Malaysia is obtained from Majlis Bandaraya Kuching Selatan (MBKS), Dewan Bandaraya Kuching Utara (DBKU) and Sustainable Urban Development (SUD). The increase of plastic waste generation is shown in **Table 2.3**. Comparing the data of plastic waste, it is noticed that the amount of plastic waste increased more than double amount within the range of 1 year and 15 years.

Table 2.3 Plastic Waste Generation in Kuching (Huong, Yang, & Larsen, 2003)

Type of waste	**Waste source and amount**			
	MBKS ('86) Kuching, landfill waste	DBKU ('96) Kuching, residential	Trienekens ('97) Kuching, landfill waste	SUD ('01) Kuching residential
Organic waste	48%	40%	50%	50%
Paper & cardboard	24%	21%	15%	14%
Plastics	9%	21%	16%	20%
Metals	6%	6%	4%	3%

Johari et al. (2014) studied on the management of solid waste including plastic waste in Malaysia. The 7 years statistics are shown in **Table 2.4** which shows that plastic waste increased throughout the years up to 25.2% in 2010.

Table 2.4 Solid Waste Statistics in Malaysia (Johari et al., 2014)

Components	2001	2002	2003	2004	2005	2007	2010
Food waste & organics	68.40%	56.30%	37.40%	49.30%	45.00%	42.00%	43.50%
Mix plastic	11.80%	13.10%	18.90%	9.70%	24.00%	24.70%	25.20%
Mix paper	6.30%	8.20%	16.40%	17.10%	7.00%	12.90%	22.70%
Textiles	1.50%	1.30%	3.40%	NA	NA	2.50%	0.90%

Note: NA – Not available

There are issues regarding the disposal of the municipal solid waste. Improper disposal of plastic waste causes in increasing of undesirable species such as soot and polycyclic aromatic hydrocarbons (PAHs) (Lam & Chase, 2012). Sewage effluents and urban runoff contain high concentrations of PAHs due to the contamination of the waste (Lam & Chase, 2012). Another problem caused by plastic waste is that the disposal of it on land makes the land barren (Bhawan & Nagar, 2012).

Toxic emission is also a problem caused by the disposal of plastic waste primarily on burning of the municipal plastic waste (Bhawan & Nagar, 2012). The toxic released are carbon monoxide, chlorine, hydrochloric acid, dioxin, furans, amines, nitrides, styrene, benzene and acetaldehyde (Bhawan & Nagar, 2012). Additives in low density polyethylene (LDPE), high density polyethylene (HDPE) and polypropylene (PP) such as lead and cadmium pigments are toxic and leach out due to plastic waste disposal (Bhawan & Nagar, 2012). Littered plastic waste will also block drains (Bhawan & Nagar, 2012).

The potential of plastic waste to be converted into hydrocarbon oil have been studied. The product of pyrolysis towards different type of plastic waste is shown in **Table 2.5**. Only certain plastics are allowed to be used as a feedstock of liquid fuels such as polyethylene, polypropylene, polystyrene and polymethyl metacrylate (Into,