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# CHARACTERIZATION AND RESOURCE RECOVERY OF MUNICIPAL SOLID WASTES EXCAVATED FROM OLD LANDFILL SITES IN INDONESIA

DOCTORAL DISSERTATION



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# **Chapter 1**

## **Introduction**

### **1. Municipal Solid Waste and Landfill Mining**

The landfill has played an important role as a part of municipal solid waste (MSW) management in urban societies since ancient times. As civilization changes, the way of people disposing of their daily activity residue also changes. The landfill is not just an unmanageable pile of garbage, but people start to realize to take care of landfills in a safe and sound approach.

Despite the high development of alternatives disposal means, the landfill is still the most popular disposal means in the form of open dumping and sanitary landfill. UNEP counted that in Asia, 51% of disposal process is open dumping while 31% is sanitary landfill. Incineration and Recycling only take 5% and 8% of the total. In Africa, 47% is open dumping, and 29% is sanitary landfill. On the other hand, in North America, sanitary landfill takes 91% of waste disposal method (UNEP, 2015). It shows that most of developing countries rely on a landfill as disposal means for their MSW because it cost less than other methods (Tchobanoglous and Kreith 2002). Operation of landfill site not only lead to decreasing environmental quality of surrounding area from smells but also at a global scale, the methane released will bring worse effect than CO<sub>2</sub> to global warming (Cherubini, Bargigli, & Ulgiati, 2009), thus, a method to minimize the impact while bringing the benefit is necessary.

Methane produced in the landfill was coming from the decomposition of degradable organic content (DOC) of inputted waste material (Rees 1980). Capturing methane gas is one of the solutions to reduce greenhouse gas emissions. The methane can be used for power generation by coupling with a gas engine or steam engine. Once the methane depleted, another method is needed to treat DOC left, not decomposed, and other inert material. Landfill mining was proposed as one of breakthrough to recycle the residue left (Dickinson 1995). Krook, et al. (2012) address that, despite the idea of landfill mining was started since the 1950s, the research about this topic increase rapidly in the 1990s because of stricter new environmental regulation. The development of more sophisticated waste treatment and recycling

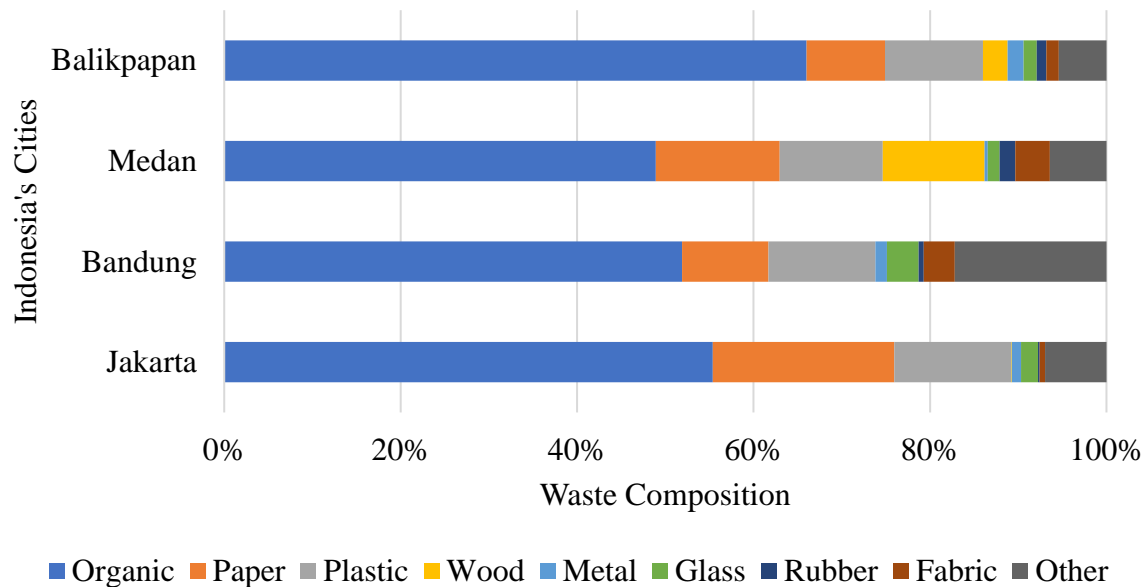
programs in early 2000 drastically change the attention of this issue, until 2007 the idea about the extended concept of landfill mining arise in Europe (Hogland et al. 2010).

A consortium in the EU initiated Enhanced Landfill Mining (ELFM) studies to build an integrated strategy for material and energy recovery. The so-called project Closing the Circle was launched to build a pilot project in Remo Landfill (Hogland et al. 2010). Hull et. Al (2005) questioned the cost feasibility of landfill mining and argue that it will only work on particular condition such as; availability of special funds for remediation, availability of feedstock to make sure waste to energy plant running in its full capacity and the presence of cement company which will buy solid fuel from processed material. Recyclable recovered from landfill mining is low-quality material, and the feasibility to utilize it is deficient. Several landfill mining projects in Europe acquire waste with a calorific value of up to 20 MJ/kg, which means high feasibility to build a waste to energy plants for processing excavated waste (Hogland et al. 2004). While European countries have a constant value for its waste composition, developing countries such as Indonesia have gone to a rapid change of development, resulting in an active change in waste composition and make it harder to predict the characteristic of its landfill waste (Damanhuri and Padmi 2016).

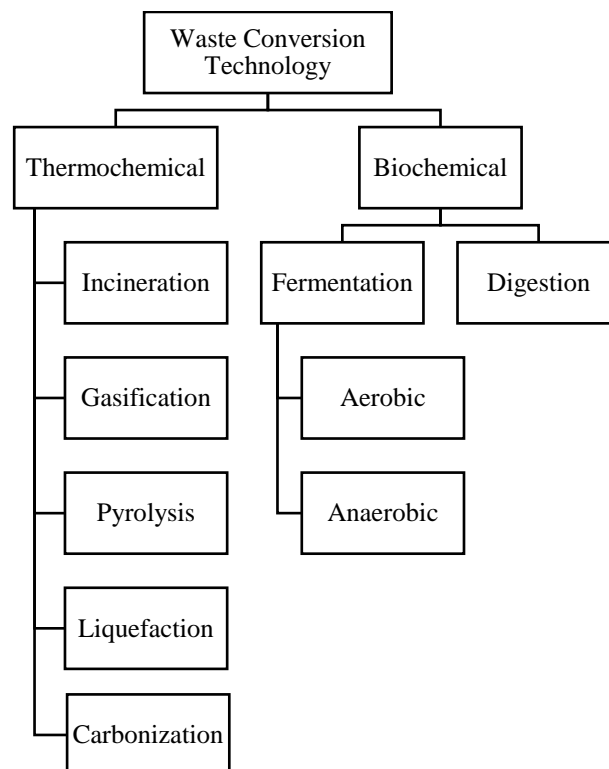
Indonesia, with a population of 257 million in 2015, shows exponential growth in its GDP and also its waste generation. In 2012, Indonesia generated solid waste up to 151,921 tons per day (tpd), and 7,896 tpd of it was from Jakarta alone (2015). Many people attracted to move to big cities to work because of low employment in rural areas. This urbanization phenomenon puts a massive burden on waste management since their landfill capacity is minimal (Damanhuri 2008). Since the enactment of Indonesian Law number 18/2008 on Waste Management, the amount of landfill sites in Indonesia is increasing rapidly. In 2014, the Ministry of Public Works was built 110 landfill sites around Indonesia. Currently, local government in Indonesia operates 521 landfills with a total area of 2098 ha, most of them are open dumping landfill (2015). Even though the basic design was a sanitary landfill, low commitment from the government for consistent operation turn the landfill into open dumping or at least a controlled landfill. In developing countries, such as Indonesia, wet organic wastes from food residue and parks dominate the composition of MSW (Figure 1.1) (Damanhuri and Padmi 2009, 2016;



Arjuna 2012; Purwaningrum et al. 2014). Various waste conversion technologies have been developed to effectively recover the energy and synthesize the material from highly contaminated waste. The overall scheme of waste conversion technology is shown in Figure 1.2.



**Figure 1. 1 Waste Fraction from the Four Highest Populated Cities in Indonesia**



**Figure 1. 2 Waste Conversion Technology for Energy and Material Recovery**

The appropriate conversion route depends on the composition and characteristics of waste. Recently, the thermochemical route was favored because it has a faster processing time and yields higher energy

**Table 1.1. Various Operating Condition of Thermochemical Technology**

Technology	Temperature (°C)	Feedstock Condition	Product
Incineration/Combustion	800-1400	Dry	Heat
Gasification/Partial Combustion	500-1300	Dry	Gas
Pyrolysis	300-600	Dry	Liquid
Liquefaction	180-400	Dry	Liquid
Carbonization	180-300	Dry/Wet	Solid

density compared to the biochemical route. Based on the targeted product and operating conditions, the thermochemical process can be classified into several processes, as shown in Table 1.1.

Incineration is the most common thermochemical technology to recover energy from waste. In principle, incineration is the oxidation of combustible waste into non-combustible gas and ash (Kokalj and Samec 2013). The incineration plant consists of a combustion chamber, heat recovery system, and gas cleaning process. There are several stages during the incineration processes, such as drying and degassing, pyrolysis and gasification, combustion, and post-combustion (Fruergaard and Astrup 2011). Those processes occupy almost the same space and time in the reactor and react almost instantly. The design of the reactor, feed characteristic, and air supply influence the performance of the incineration process in regards to energy efficiency and pollutant emission (Ruth 1998).

The main challenge of incineration is that most of the unprocessed contaminated waste contains a high percentage of plastic. Developed countries such as Japan and Northern Europe account for 20% and 13% of plastic fraction in their waste, respectively. Meanwhile, the developing countries in South America and East Asia have about 10% of plastic in their waste stream (2006). Organic chlorine in plastic waste is the main contributor to the formation of hydrochloric acid, dioxin, and furan in the flue gas. Europe was known to implement strict standards about the chlorine content in the flue gas, which is ranging between 0.1 to 1.0% (2015).

With the increase of environmental concern, gasification is favorable technology to produce synthetic gas (syngas) for power generation or chemical feedstock. Gasification converts feedstocks into syngas by partial oxidation (Arena 2012). Gasifying agents, such as air, oxygen, and steam, were used as an oxygen carrier. Recently, recycled flue gas with a high amount of carbon dioxide was mixed with oxygen to prevent NO<sub>x</sub> formation during combustion (Leckner 2015). Another similar system using a solid oxygen carrier, called chemical looping gasification, has also been developed recently. In this case, metal oxide as an oxygen carrier performs a solid-solid reaction with waste feedstock to produce a high yield of hydrogen (Cuadrat et al. 2012).

Pyrolysis also has a lower environmental impact compared to incineration. Pyrolysis is a thermal degradation process in the absence of oxygen with an operating temperature of 400–600 °C. Pyrolysis yields gaseous, liquid, and solid products. By modifying the temperature, pressure, and catalyst, different product compositions could be arranged. Typical product energy yield from pyrolyzed waste is between 5 to 15 MJ/m<sup>3</sup> (Bosmans et al. 2013). Secondary treatment of pyrolysis gas and char have been done through condensation of the gases into usable oil mixtures or incineration of gas and char to generate heat and destroy the organic matter. The advantages of pyrolysis processes are to allow multiple product generations in one process. The organic fraction can be recovered for material or fuel (e.g., methanol and Fischer-Tropsch fuels) (Dayton 2002). Char can be utilized for various feedstock, such as solid fuel, activated carbon, and carbon nanotubes (Cha et al. 2016). Moreover, pyrolysis gas could be used in gas engines or gas turbines for power generation (Zhou et al. 2014).

Carbonization is a process which increases the content of carbon element from organic material by removing volatile matter, reducing moisture content, removing inorganics, and reducing the atomic ratios O/C and H/C, usually without the presence of oxygen (Marsh and Rodríguez-Reinoso 2006). Torrefaction and hydrothermal are commonly used in the carbonization process. Torrefaction prefers dry material since inert gas is used as a medium in temperature between 240 to 320 °C, while hydrothermal uses highly pressurized steam as a medium to breakdown the long carbon chain in temperature between 180 to 250 °C (Libra et al. 2011; Rudolfsson et al. 2015). The steam will also wash

away the inorganics, such as chlorine, potassium, and heavy metals (Prawisudha et al. 2012; Novianti et al. 2015; Huang and Yuan 2016).

**Table 1.2. Various Hydrothermal Treatment Process**

Process	Temperature	Pressure (bar)	Medium	Product
HTC	180-250	1-20	Subcritical Water	Solid
HTL	200-400	50-221	Subcritical or Supercritical Water	Liquid
HTG	300-700	90-410	Subcritical or Supercritical Water	Gas

Some inherent properties of MSW, such as high moisture content, low energy density, hygroscopicity, low heating value, high alkali content, heterogeneity, and high ash content, have become challenges in its thermochemical conversion. MSWs, especially in developing countries, were usually mixed with improper separation at the source. The composition is dominated by food waste, causing the moisture content becomes very high. It also has a low bulk density because of the high amount of plastic bag waste. The condition becomes worse in the rainy season, which usually could last for more than six months in tropical countries (Helmy et al. 2014). It leads to difficulties in collecting, handling, and transporting the MSW to the landfill site. Moisture in MSW also decreases its heating value in general. Organics from food and garden wastes have fibrous and tenacious characteristics, while plastic waste has high elasticity. Due to those characteristics, MSW is difficult to be ground into small homogenous particles.

Poor grindability can cause lower reactor performance and other serious problems (Ramos Casado et al. 2016). High ash content with high alkali and chlorine content can cause slagging and fouling in the furnace and promotes the production of corrosive and toxic flue gases (Demirbas 2004). The main contributor to fouling is from the content of the inorganic of the feedstock. Sodium and potassium lower the melting point of ash, thus increase ash deposition and fouling of boiler tubes. The alkalis are more available and reactive in the bio-based organics rather than in coal deposits (Vassilev et al. 2013). Accumulation of ash reduces heat transfer and causes severe corrosion at high temperatures. Ash deposition from biomass fuel is denser and harder to remove compared to coal. Ash content also directly affecting the heating value. Higher ash content means lower heating value (Brunner and Rechberger

2015). Therefore, MSWs in developing and tropical countries become more difficult to be burnt or gasified with high efficiency and low emission.

Alternative pretreatments are necessary to effectively and efficiently process MSW and recover the material and energy. This paper focuses on reviewing and discussing a hydrothermal treatment, a process of breaking a long carbon chain while removing inorganics using heated and pressurized water or steam, for MSW. Current issues and development of utilization of hydrothermal to process post-consumer goods and MSW will be the focus of this work. Several experimental data of hydrothermal experiments are compiled and reviewed in order to compare each experiment. Reaction time was expressed in hours (h) and was log10 transformed. The ratio of solid feedstock and the liquid medium was expressed as weight percentage on a dry basis of feedstock divided by the total of liquid and solid in the reactor. In addition, the data were interpreted through principal component analysis.

## **2. Hydrothermal Treatment**

Hydrothermal treatment is a process involving steam/water and elevated temperature. There are three types of hydrothermal treatments: hydrothermal carbonization (HTC), hydrothermal liquefaction (HTL), and hydrothermal gasification (HTG). Wet feedstock, such as MSW, which can retain up to 60% of moisture in its weight, can be processed without pre-drying, thus allows energy saving (Kruse et al. 2013). There are similarities in the targeted product and working temperature between HTC and torrefaction, HTL and pyrolysis and torrefaction, and HTG and gasification. However, hydrothermal treatment could run in the lower temperature because the water acts as a solvent, catalyst, and reactant in the process. Table 1.2 shows the distinction between each hydrothermal process in detail.

Usually, a hydrothermal reactor consists of feeder, reaction, and product discharge units. In the feeder unit, the feedstock is mixed with the water, steam, or solvent and catalyst if necessary. The mixture is then moved into a reaction unit using a slurry pump or screw. In the reaction unit, the temperature is held according to the designated retention time. After the reaction finished and the reactor has been cooled down, the slurry was moved to the product discharge unit and pressed to remove the water. The final product could be dried using natural drying or wind blower.

The water plays a vital role during the reaction process. Water characteristics and properties will change depending on the temperature and pressure. In the temperature of 200 to 280 °C, the acidic and basic properties will change depending on the ion presence. The dielectric constant decrease and make it act as a non-polar solvent. The number of hydrogen bonds is also lower and weaken. The isothermal compressibility is higher, which helps in the destruction process of the material. The solubility of organic compounds increases while the solubility of inorganic salts decreases (Funke and Ziegler 2010). At 250–350 °C, the feature of water as a solvent will be similar to the feature of organic solvents at room temperature. In a subcritical temperature (100–374 °C), the ionization constant of water increases with temperature. Nevertheless, getting closer to a critical point (374 °C, 22.1 atm), ionization constant, dielectric constant, and the concentration of ionization products decrease sharply. Usually, reactions in liquid are controlled by diffusion, and thus, the rate of reaction depends on the viscosity of the liquid (Falco et al. 2011).

During the hydrothermal process, added water enters the subcritical phase, where the liquid water behaves as a non-polar solvent. High ionization degree of water at high temperature and pressure followed by dissociation of water into  $\text{OH}^-$  and  $\text{H}_3\text{O}^+$  has acidic and basic characteristics at the same time (Marcus 1999). The subcritical condition promotes hydrolysis reaction, which reduces the activation energy of cellulose and hemicellulose. Thus, the feedstock will be depolymerized and degraded into water-soluble products (Bobleter 1994). The hydrolysis of cellulose and the decomposition of hemicellulose convert 40 to 60% of the initial biomass into a dissolved state.

Temperature is also a critical element that influences product characteristics after hydrothermal treatment. The temperature has a positive effect on the increase of carbon content but decreases the hydrochar yield (Novianti et al. 2014). The rate of hydrolysis and depolymerization of biomass depends on temperature. Hemicellulose was hydrolyzed between 180 to 200 °C, lignin was degraded between 180 to 220 °C, while cellulose is degraded above 220 °C (Hardi et al. 2017).

The span of reaction time up to 2.5 h has a significant effect on hydrochar yield, ash content, carbon content, O/C-ratio, energy densification, and energy yield (Mäkelä et al. 2015). Retention time promotes the changes in morphology, which is related to the higher released volatiles, and more char

carbonization occurs. Reaction time longer than that tends to increase the ash content and lower the yield. Some ash formed during carbonization stages could seep into the hydrochar pores in longer retention time (Hoekman et al. 2013; Lokahita et al. 2017).

The hydrothermal treatment does not necessarily need a drying system since the particle size reduction is performed in wet conditions inside the reactor. It does, however, need steam generation energy to supply steam to the reactor.

### Research Objective and Scope

This research aims to investigate the potential of hydrothermal treatment to excavated waste from an old landfill in the perspective of energy and material recovery. Aseptic packaging was used to model the composite material from landfills and was subjected to hydrothermal treatment before using actual waste excavated from landfills. Landfill excavation was done to understand the actual waste composition and understand the material flow analysis. Excavated waste was treated using the hydrothermal treatment, and the potential utilization was investigated.

### Dissertation Outline

#### Chapter 1. Introduction

In this chapter, the essential background and objectives of the research are presented. The situation on waste management and landfill mining process were reviewed. An overview of the latest development of hydrothermal treatment in the field of post-consumer waste and MSW treatment, with a particular focus on operating conditions and physicochemical characteristics of the hydrochar, will be presented. Then, the research objective is stated. Finally, the outline and scope of this study are presented.

#### Chapter 2. Hydrothermal Treatment of Aseptic Packaging Waste

This chapter will discuss the result from hydrothermally treated Aseptic Packaging. Aseptic Packaging waste was chosen to model the behavior of composite waste during hydrothermal treatment. Aseptic Packaging contains a layer of cellulose, plastic, and aluminum with a composition of 75%, 20%, and 5%, respectively. The experiment was done at the three different holding times between 0 and 60 min

and temperatures between 200 and 240 °C. A total of nine experiments were conducted to understand the effects of holding time and temperature on the quality of solid fuel and composites.

### Chapter 3. Landfill Mining and Excavated Waste Characteristics

The objective of this chapter is to examine two landfill sites in Indonesia, Jatibarang landfill (JL) and Piyungan landfill (PL). The potential uses for waste-to-energy (WtE) and recycling were observed by a combination of process modeling and material flow analysis.

### Chapter 4. Energy Recovery from Hydrochar

In this chapter, a method to enhance the quality of combustible excavated waste in an energy recovery point of view using hydrothermal treatment was proposed. Two-level factorial screening design was used to determine the effect of temperature, solid load, and holding time towards the properties of the solid products.

### Chapter 5. Hydrochar utilization as activated carbon for landfill leachate treatment

This chapter presents the alternative utilization of hydrothermally treated excavated waste. Excavated waste from landfill mining project is hidden resources which potentially be utilized for many applications after treatment. The hydrothermal treatment has proved to alter the properties of excavated waste by increasing its carbon content and calorific value, though it is still possessed high ash content. Utilization of hydrochar as activated carbon could be an alternative uses of hydrochar besides solid fuel. Leachate emitted from landfill sites contains high chemical oxygen demand form active organic compounds. The hydrochar was activated using steam and CO<sub>2</sub> to modify the surface properties and improve the adsorption capacities.

### Chapter 6. Conclusion and Recommendations

The main findings from each chapter are re-summarized in this chapter. The experimental results are linked together between each study to articulate and explain the important mechanism. Finally, the recommendation for future work or implementation of this research is suggested.



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## Chapter 2

# Hydrothermal Treatment of Aseptic Carton Waste

### 1. Introduction

The most popular product for Aseptic Packaging is Tetra Brik ® by Tetra Pac ©. Tetra Brik is used for milk packaging after ultra-pasteurization, juice, and even wine. The aseptic packaging is mainly composed of three materials, which are organized into six layers: paper (75%), LDPE (20%), and aluminum (5%). A layer of an ethylene/methacrylic acid copolymer [poly (ethylene-co-methacrylic acid (EMAA))] is used as an adhesive agent of the aluminum film to the plastic layer because of its behavior as an inert polymer and its similar properties with LDPE. Having a nonionized structure, this random ethylene copolymer has comonomers that scattered along the sequence. These copolymers also present proper thermal sealing in the presence of aqueous fluids (Phichai et al. 2013).

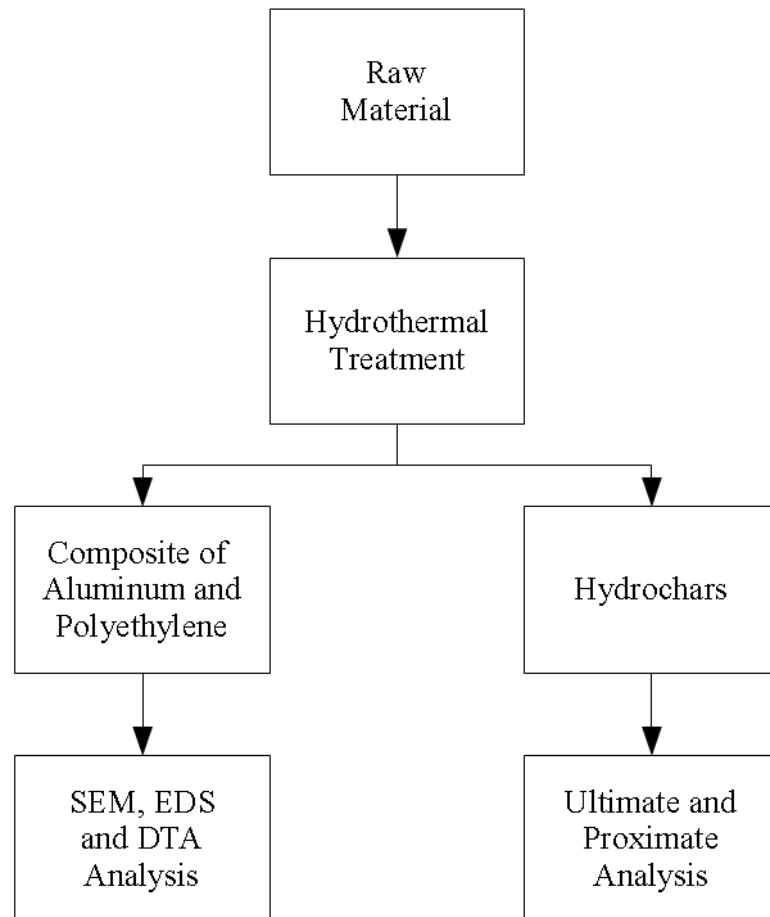


**Figure 2.1 Various type of Aseptic Packaging Product**

Hydrothermal treatment is gaining popularity to process organic material or waste from the municipality and also the industry in recent decades. The hydrothermal treatment is a thermal process by utilizing water in the subcritical condition to convert waste into upgraded products such as coal-like solid fuel or solid fertilizer. The degradation of the material under the subcritical water condition is caused mainly by hydrolysis, dehydration, decarboxylation, condensation, and aromatization reactions. Various studies have shown that hydrothermal treatment can increase the heating value, improve the natural drying performance, and reduce organic chlorine content of the feedstock resulting in a safe and clean alternative solid fuel product (Novianti et al. 2016b).

## 2. Methodology

In this study, Aseptic Packaging was processed using the hydrothermal treatment in order to produce solid fuel and recover the aluminum part. Figure 2.2 shows the schematic diagram of the experiment and analysis.



**Figure 2.2 Research diagram of the hydrothermal treatment on aseptic packaging waste**

Aseptic Packaging sample employed in this work was obtained from Tetra Pak Japan. Tetra Pak Japan collects Aseptic Packaging waste from municipalities to be recycled in their plant. The sample was received in an open and clean condition. The sample then chipped into about 1cm<sup>2</sup> using scissors and then dried overnight.

## 2.1. Experimental

The hydrothermal treatment was carried out using a lab-scale reactor series MMJ-500 made by OM Lab-tech Co., Ltd., Japan. The reactor vessel is made of glass with 500 ml of the volume capacity. A motor stirrer, a pressure gauge, and a PID temperature controller were equipped in the reactor.

Approximately 9 grams of solid samples mixed with 81 grams of distilled water were used continuously for all experiments. The reactor was sealed and purged with argon gas to create an oxygen-free environment inside the reactor. Then the reactor was heated up to the targeted temperature of 200, 220, and 240 °C by the electric heater before retained for 0-60 minutes. The pressure was not controlled but monitored by the pressure gauge. The motor stirrer was set at 400 rpm to help the separation process by creating a vortex inside the reactor.

**Table 2.1. Experimental parameter**

Experiment no.	Temperature (°C)	Time (minutes)
1	200	0
2	240	0
3	200	60
4	240	60
5	200	30
6	240	30
7	220	0
8	220	60
9	220	30
10	220	30
11	220	30

The experiments were standardized, corresponding to the central composite design (Areeprasert 2015). A total of eleven experiments, including three repetitions in the center point was conducted to understand the effect of time and temperature on the quality of the produced solid fuel. Table 2.1 shows the variation of time and temperature used in this experiment.



After the reaction is finished, the reactor was cooled down to 80 °C. Then the sample was discharged from the reactor to be dried at 105 °C overnight. Dried samples contained two materials; the hydrochar and the composite of aluminum and polyethylene. Those materials could be separated easily because they are in different phases. After the separation, the sample was stored in a sealed bag before the characterization.

## 2.2. Product Characterization

### 2.2.1. Ultimate Analysis

The most important fuel elements are carbon and hydrogen. The fuel also contains some oxygen and small quantities of incombustibles (water vapor, nitrogen, and ash). The ultimate chemical analysis determines the mass percentage of carbon (C), hydrogen (H), oxygen (O), nitrogen (N), and ash in the fuel. By understanding data from the ultimate analysis, the high heating value and the atomic ratio could be predicted. The atomic ratios of O:C and H:C in the product will be used to build Van Krevelen diagram. Van Krevelen diagram classifies the quality of fuel by the type of kerogen in the material. Coal like material will be positioned in the lower respective ratios as it contains richer carbon and higher energy content of the material. By comparing with other fuels, the operational condition which brings coal-like product was found.

In this research, the ultimate analysis was performed using the Vario Micro Cube Elemental Analyzer (Elementary, Germany). The high heating value of the treated sample was calculated using the formula by Phichai with the dry ash-free base (Phichai et al. 2013), which is shown in Equation 2.1.

$$\text{HHV} = 343.08 \text{ C} + 424.92 \text{ H} + 261.98 \text{ N} + 27.76 \text{ O} \quad (2.1)$$

### 2.2.2. Proximate Analysis

The proximate analysis determines the mass percentage of volatiles, ash, and fixed carbon in the material. The proximate analysis of hydrochar was performed using the Shimadzu D60 TGA/DTA Analyzer. Approximately 10 mg of samples were loaded in a crucible and then placed in the analyzer. Nitrogen was purged for 5 minutes in order to create an inert atmosphere inside the analysis chamber. Then the samples were rapidly heated to 40 °C before the measurement began. After measurement

started, samples were heated at a constant rate of 10 °C/min until 105 °C and held for 20 minutes to remove the moisture. The thermal decomposition of the volatile matter was observed when the samples were heated at a constant rate of 50 °C/min until 950 °C and held for 7 minutes. Lastly, the combustion of the final residue was done to measure the amount of fixed carbon by holding it for 15 minutes at 950 °C under the air atmosphere.

### 2.2.3. Density of Aluminum

The density of the composite of aluminum and polyethylene was calculated using a pycnometer and an analytical balance. Pycnometer can be used to determine the density of solid by comparing the mass difference between the liquid with and without the target product. First, the weight of the pycnometer was measured together with the product inside. Then, distilled water was added. Lastly, by using another empty pycnometer, it was filled with the same liquid, and the mass was measured. The volume of the measured object was the difference between the volume of water that filled the empty pycnometer and the volume of water that filled the pycnometer with the solid object. The density of the solid object was calculated by dividing the measured object mass with the volume of the measured object.

### 2.2.4. Aluminum Content

DTA analysis was used to determine the appearance of aluminum in the composite by observing the thermal behavior of the material. Aluminum was known to melt at the temperature of 660 °C (Wu and Chang 2001), while polyethylene melts at 115–135 °C (Peterson et al. 2001). From a DTA curve, the melting point of the substance can be determined as the point of intersection of the leading edge of the melting peak with the extrapolated baseline.

The appearance of aluminum in the surface composite was analyzed by scanning the cross-section of the composite using the Scanning Electron Microscopy (SEM) coupled with the Energy Dispersive Spectroscopy (EDS) by JEOL.

SEM is a characterization method that is used for various types of samples, from hard metals to soft polymers and biological tissues. An electron beam is focused and scanned over the targeted area, and signals are produced from the interactions of electron-matter on the surface of the specimen. Secondary

and backscattered electrons for the image formation and the characteristic x-ray from the elements present in the sample for composition studies also included in the main signal. SEM gives information regarding the morphology of the surface of the samples at the sub-micrometric. The final pictures will look like monochrome photographs.

EDS is a spectroscopic method to determine the presence and relative abundance of the elements that compose the surface of the specimen under study. The X-ray photons are produced when an electron beam reaching the surface is detected, and their energy depends on the atom from which they come. Only X-rays coming from atoms with atomic numbers higher than four are detected. Thus, H, He, and Ba are not detected by EDS, while this technique can study all other elements of the periodic table. EDS is also a powerful tool for qualitative x-ray microanalysis. Since the x-ray is always obtained in all parts of the specimen, the information needed is always available. Qualitative analysis was done by analyzing the statistically significant peaks. The minimum size of the peak after the background subtraction must be three times the standard deviation of the background at the peak position.

From the mass balance, the actual yield of aluminum could be predicted. After removing the mass of contaminants by burning the samples at 950°C, the leftover ash minus the ash of contaminants was predicted as the actual aluminum content.

### **3. Result and Discussion**

Hydrochar and PEAL (composite of polyethylene and aluminum) were formed during the process of the hydrothermal treatment of Aseptic Packaging. In this chapter, hydrochar properties as a solid fuel and PEAL properties as sources of recyclables are discussed.

#### **3.1. Hydrochar properties as a solid fuel**

The ultimate and proximate analysis were used as tools to evaluate the characteristics of the produced hydrochar. The raw material was unable to undergo the ultimate analysis because of aluminum content in the sample resulting in high ash content. Thus, the analysis becomes unreliable.

The ultimate analysis of treated Aseptic Packaging will present the effect of time and temperature in the percentage of carbon, hydrogen, oxygen, and nitrogen. Table 2.2 shows the result of the ultimate analysis and comparison with other fuel.

The elemental composition and the heating value of hydrochar were changed after being treated by the hydrothermal process. Both time and temperature have a positive effect on raising the value of the carbon content, the nitrogen content and the high heating value (HHV) which confirms to the previous research (Muthuraman et al. 2010; Lu et al. 2011; Prawisudha et al. 2012; Areeprasert 2015; Novianti et al. 2016b). The carbon content was ranged from 42.91% to 65.72%; the nitrogen content was ranged from 0.1% to 0.24%, and HHV was ranged from 18.59 MJ/kg to 25.2 MJ/kg.

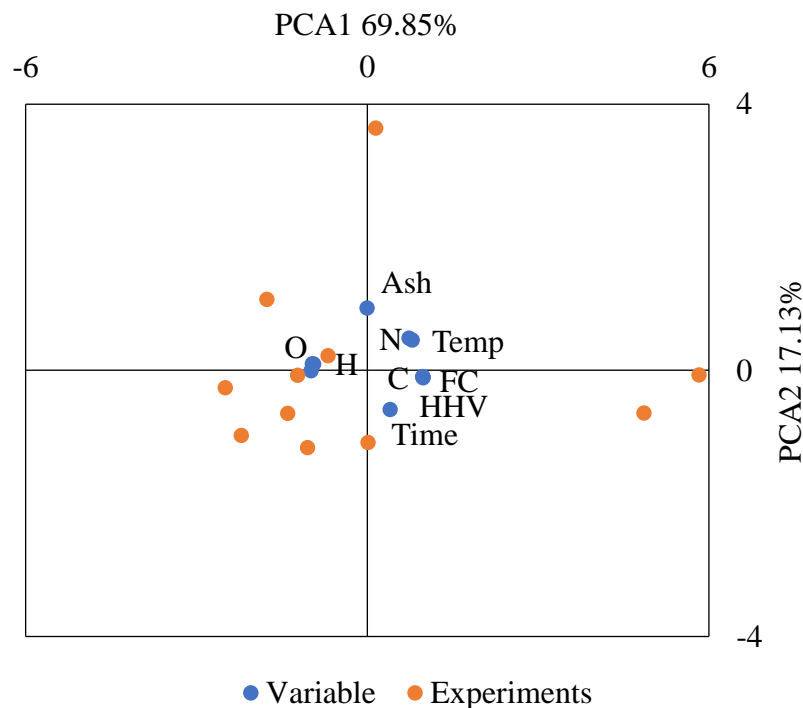
**Table 2.2 Ultimate Analysis of the hydrochar**

Temperature °C	Time (minutes)	C	H	N	O	HHV (MJ/kg)
200	0	43.67%	6.15%	0.10%	42.91%	18.81
240	0	45.18%	5.66%	0.23%	44.85%	19.20
200	60	43.55%	5.93%	0.18%	44.82%	18.75
240	60	65.72%	4.34%	0.24%	27.48%	25.20
200	30	43.15%	5.68%	0.12%	48.95%	18.59
240	30	59.94%	4.27%	0.19%	27.82%	23.18
220	0	42.91%	5.92%	0.15%	47.81%	18.59
220	60	46.01%	5.49%	0.16%	45.10%	19.39
220	30	44.99%	5.85%	0.18%	44.82%	19.21
220	30	44.86%	5.88%	0.14%	46.45%	19.20
220	30	44.02%	5.96%	0.13%	45.70%	18.93

The highest carbon content and HHV were achieved at the temperature of 240 °C and the holding time of 60 minutes, which are closed to the value of high-rank coal (Demirbas 2004). On the other hand, hydrogen and oxygen were decreased with the increase of temperature and time. The hydrogen content

was ranged from 4.27% to 6.15%, while the oxygen content was ranged from 27.48% to 48.95%. Those behaviors happened because of the hydrolysis reaction in the hydrothermal process cut down the long chain of hydrocarbons in the paper part of Aseptic Packaging into smaller carbon chain. Research by Srikandi, 2016, shows the same behavior towards the mechanism of hydrolysis in the hydrothermal treatment of an empty fruit bunch (Novianti et al. 2016a). The principal component analysis shown in Figure 2.3 demonstrates a better view of the effect of the reaction time and temperature on the composition and the heating value of hydrochar.

Figure 2.3 shows the principal component of the ultimate analysis results evaluating the correlation of experiment variables and product properties. The first principal component mainly describes the carbon content, the high heating value, nitrogen, oxygen, and hydrogen. The second principal component describes the holding time and the operating temperature. Time, temperature, carbon, nitrogen, and HHV is located in the same region, which means that they have the same positive interaction between each other. Since the temperature has a higher correlation value than time, the temperature had higher effects in the experiment.

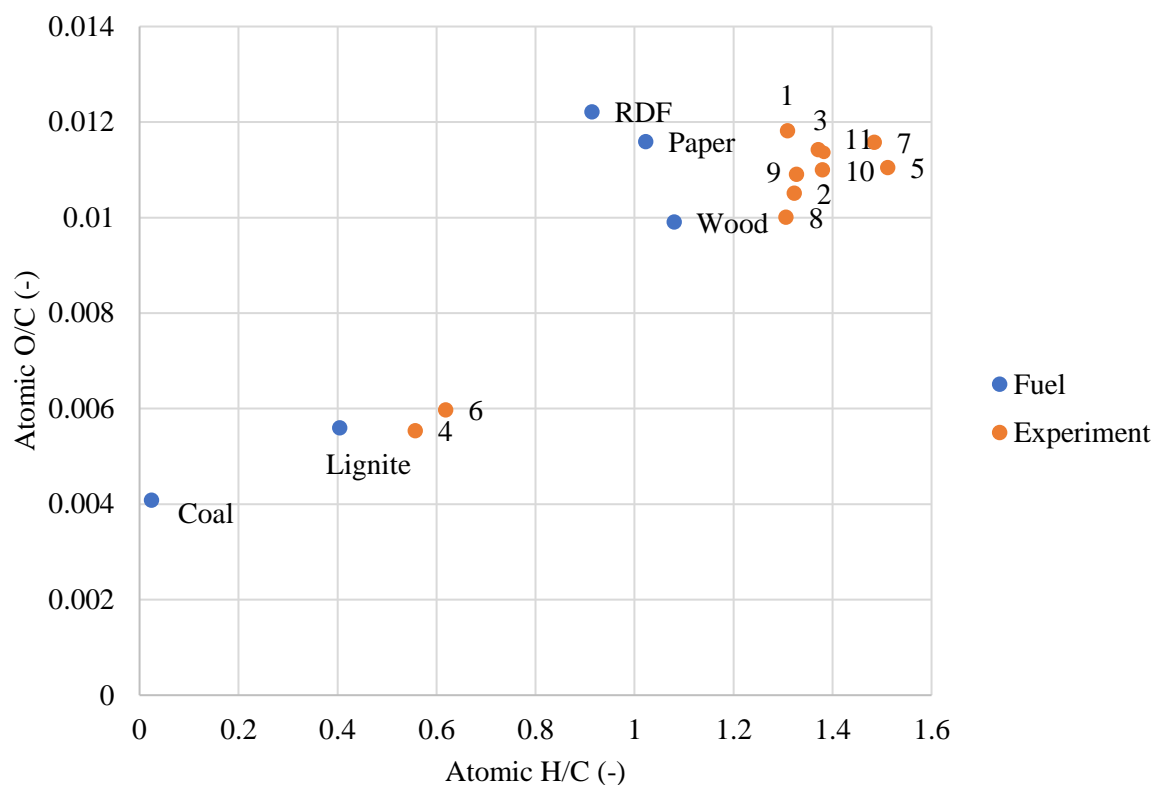


**Figure 2.3 Principal component analysis of the experiment and variables**

Most of the experiments also sit together in the same region, meaning that they have similar behavior. Three points of experiments outliers are the experiment at a temperature of 240 °C. Since they sit in the same quadrants with carbon, nitrogen, and HHV, they have the highest value of those variables. It also means that at the temperature of 240 °C, the samples changed dramatically compared with other experiment temperature. 240 °C could be the tipping point to yield higher heating value. In general, the first and the second principal components explained 87.08% of the variation in the data matrix. Similar behavior of carbon, oxygen, and calorific value in PCA analysis results were also shown by Mäkelä and Yoshikawa (2016).

The hydrothermal treatment is mimicking the process of coalification in nature by utilizing the temperature and the holding time. The effect of the hydrothermal treatment on the atomic composition could be understood by plotting the atomic H/C and O/C ratios of biomass, and the corresponding hydrochar were plotted in a Van Krevelen diagram. As mentioned before, the hydrothermal treatment altered the chemical characteristic of the hydrochar. Figure 2.4 shows the Van Krevelen diagram to compare produced hydrochar with conventional fuels.

Experiments at the temperature of 200 °C and 220 °C showed high atomic H/C and O/C ratios. These ratios decreased as the temperature and the time increased. The products from the experiments of No. 4 and 6 showed similar characteristics with lignite. The hydrochar obtained in the experiments No. 4 and 6 were denser compared to others due to the degradation of hemicellulose and other extractives at the temperature above 200 °C. There was no significant change in hydrochar of the experiments Nos. 1,2,3,5,7,8,9,10 and 11. They have a similar composition with paper and wood. On that point, the cellulose component in the samples already started to decompose, but the amount is still low, resulting in low carbon yields. The trend in the diagram also shows that dehydration is the major reaction during the process as oxygen content decreased in proportion to the hydrogen content. In summary, low H/C and O/C ratios within the fuel are favorable because of the reduced energy loss, smoke, and water vapor during the combustion process.



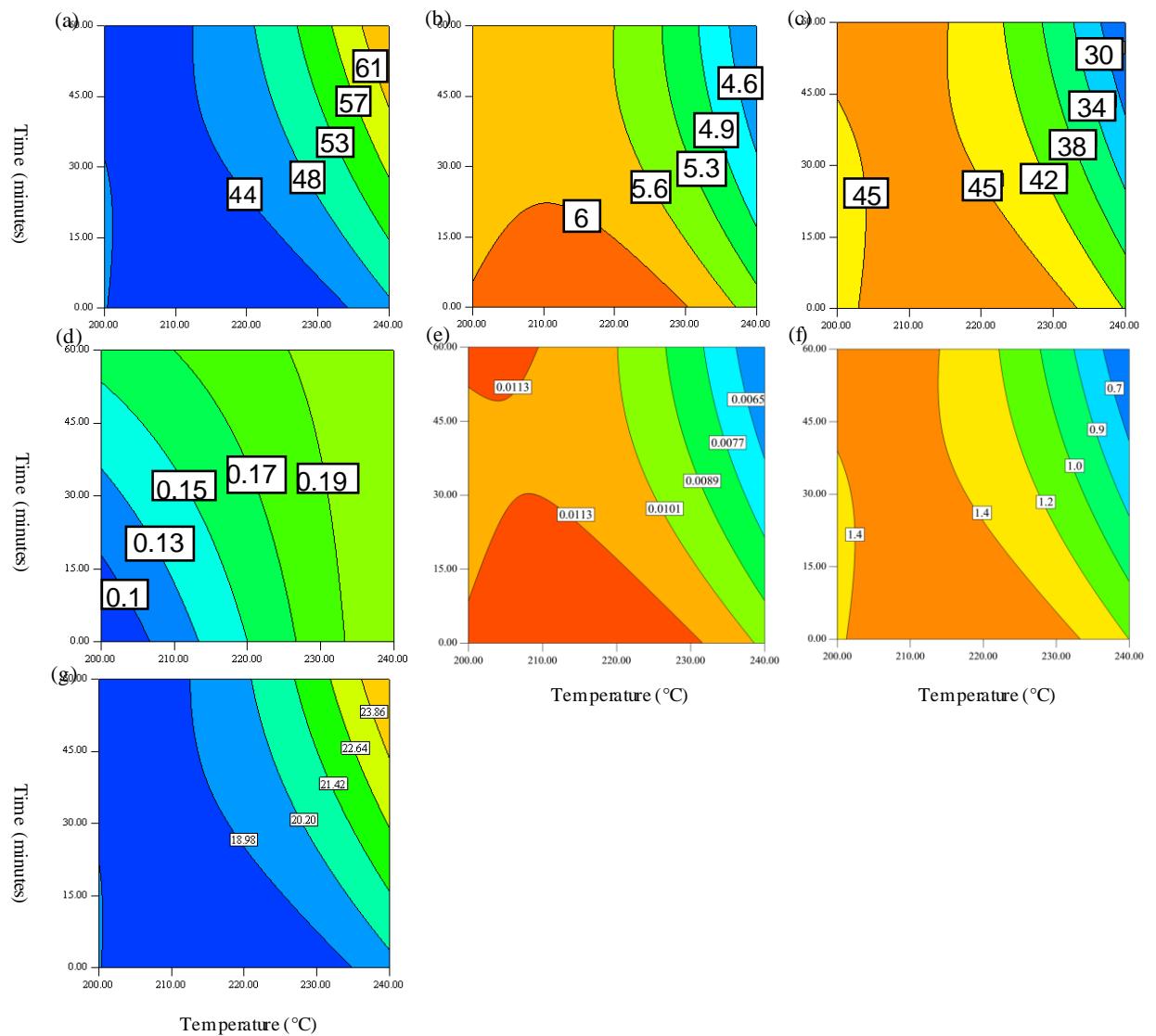
**Figure 2.4 Van Krevelen Diagram**

Experimental models were used to predict the hydrochar behavior. The response contour based on the reactor temperature and the holding time will be the best way to illustrate the models. Figure 2.5 shows the response contour of the carbon content, the nitrogen content, and the calorific value increased by increasing the reactor temperature and the holding time. The hydrogen content, the oxygen content, the atomic H/C ratio, and the atomic O/C ratio decreased by increasing the reactor temperature and the holding time. The increase in HHV and the carbon content are mainly due to dehydration, decarboxylation, and the partial dissolution of organic components. In the hydrothermal process, the material was mostly in contact with saturated vapor resulting in the minimum carbon dissolution into the liquid phase and an increase of the energy yield of sludge hydrochar.

**Table 2.3 R-Squared and Standard Deviation value of the model**

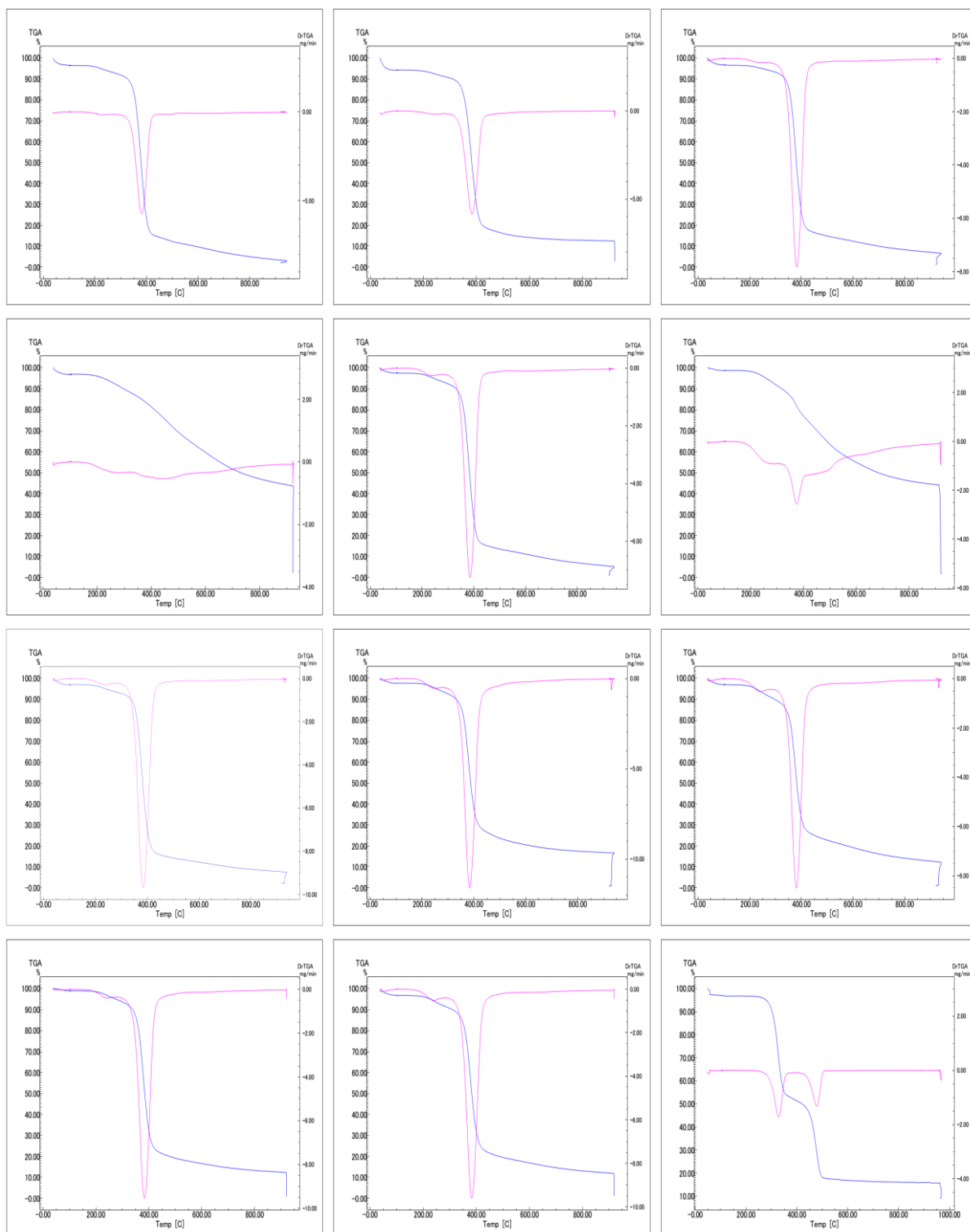
	C	H	N	O	Atomic O/C	Atomic H/C	HHV
R-Squared	95.30%	89.54%	73.46%	87.45%	91.18%	94.09%	95.54%
Std. Dev.	2.35%	0.29%	0.03%	3.75%	13.71%	0.08%	65.04%

Table 2.3 described the R-Squared and Standard Deviation value of the model. The coefficient of determination (R-Squared) value is ranging from 95.54% to 73.46%, which indicated a good fit between predicted values and the experimental data points. The highest R-Squared observed in the measurement of HHV which implies that the independent variables explain 95.54% of the variations for HHV, and this also means that the model does not explain only 4.46% of the variation. On the other hand, the standard deviation value is ranging from 0.03% to 65.04%.



**Figure 2.5** Response contours of hydrochars as functions of temperature and time: (a) % carbon content, (b) % hydrogen content, (c) % oxygen content, (d) % nitrogen content, (e) atomic H/C ratio, (f) atomic O/C ratio, (g) HHV (MJ/kg)





**Figure 2.6 Mass fraction (blue) and derivatives (purple) profile of hydrochars in experimental standard order from top left to right and Aseptic Packaging (bottom right)**

A series of thermogravimetric analyses (TGA) was performed to investigate the effect of hydrothermal treatment on the thermal decomposition of hydrochars from Aseptic Packaging. Figure 2.6 presented a comparative analysis of the TG and DTG curves of hydrochars and Aseptic Packaging. The curvature

in the experiments Nos. 1, 2, 3, 5, 7, 8, 9, 10, and 11 showed similar decomposition behavior. Their significant mass loss occurred in the temperature range of 200-600 °C with only one maximum rate of decomposition observed at around 350 °C to 400 °C. The mass reduction in that area was due to the hemicellulose degradation, lignin decomposition, and cellulose breakdown since the hydrochar was mostly made of the paper fraction of Aseptic Packaging.

As reported by Zaini et al. (2017), hemicellulose starts to degrade before the cellulose in biomass will be combusted. Hemicellulose is more easily oxidized because it has an amorphous structure with a lower molecular weight in its chemical structure. Celluloses itself has been known to have the highest thermal stability compared to other lignocellulose components owe to its crystalline structure. The crystalline structure of celluloses is constructed by the intramolecular and intermolecular hydrogen bonds between hydroxyl groups within the same cellulose chain. The chains then are arranged parallel and form a crystalline supramolecular structure. However, it has been investigated that the thermal treatment utilizing hot compressed water could change the crystalline structure of cellulose (Lynam et al. 2011).

The experiment Nos. 4 and 6 showed different thermal decomposition curves compare to other experiments. The single trend of curvature means that the decarboxylation of cellulose in hydrochar almost turned all material into carbon. This result aligns with a previous explanation about high carbon content in that experiment. The experiment Nos. 4 and 6 also showed a similar thermogravimetric curve of hydrochar produced at the temperature above 230°C and Malaysian coal which have a smooth degradation curve up to 800 °C and around 60% mass loss (Sevilla and Fuertes 2009; Idris et al. 2010). Moreover, the experiment No. 4 showed complete breakup of cellulose since the peak in the degradation rate was not found. Another researcher also observed the total disappearance of cellulose after the hydrothermal treatment at 320 °C and 25 MPa (Peterson et al. 2008). The decomposition rate peak was observed in experiment No. 6, meaning that a little fraction of cellulose still existed in the hydrochar.

The bottom right of Figure 2.6 shows the thermogravimetric curve of Aseptic Packaging before the hydrothermally treated. The DTG curve presents two peaks indicating the presence of virgin paper

material and polyethylene. By comparing with other graphs, the lower mass loss indicates the high content of aluminum in the residue.

The proximate analysis was done to analyze the volatile, fixed carbon, and ash contents in the hydrochar. Table 2.4 reported the result of the proximate analysis after several experiments using the Shimadzu TG-60 TGA/DTA analyzer.

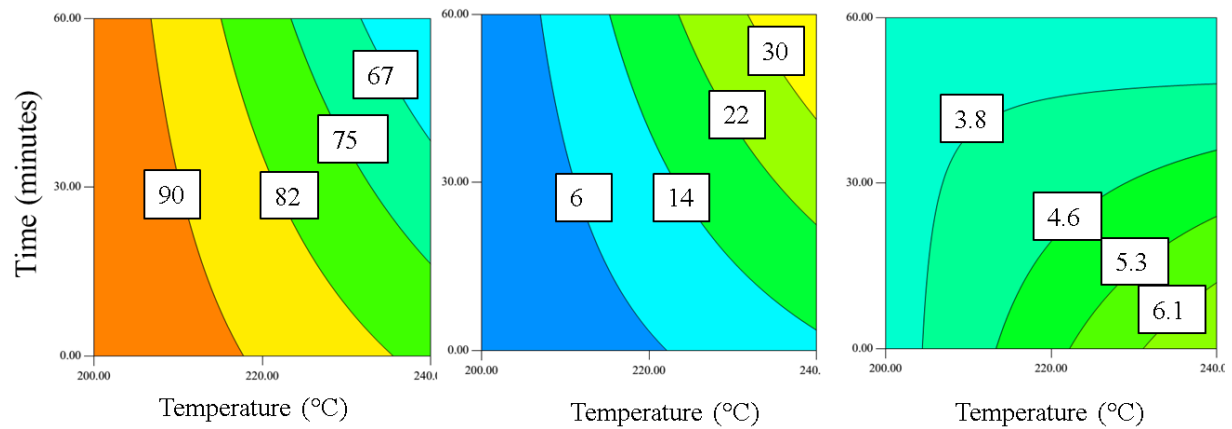
**Table 2.4 Proximate analysis of hydrochar (dry base)**

Experiment no.	Parameter		Volatile	Fixed Carbon	Ash
	Temperature (°C)	Time (minutes)			
1	200	0	95%	1%	4%
2	240	0	82%	9%	8%
3	200	60	92%	3%	4%
4	240	60	60%	35%	5%
5	200	30	95%	2%	4%
6	240	30	58%	39%	3%
7	220	0	91%	4%	5%
8	220	60	82%	15%	3%
9	220	30	87%	8%	4%
10	220	30	88%	10%	2%
11	220	30	87%	8%	4%

The hydrothermal treatment with various operating temperatures and holding times was proved to alter the composition of hydrochar. The volatile content was ranging from 58% to 95%. The fixed carbon content was ranging from 1% to 39%, while the ash content was ranging from 2% to 5%. The response surface methodology was used to build a prediction model of the proximate analysis. Figure 2.7 shows the response surface contour of the proximate analysis of hydrochar.

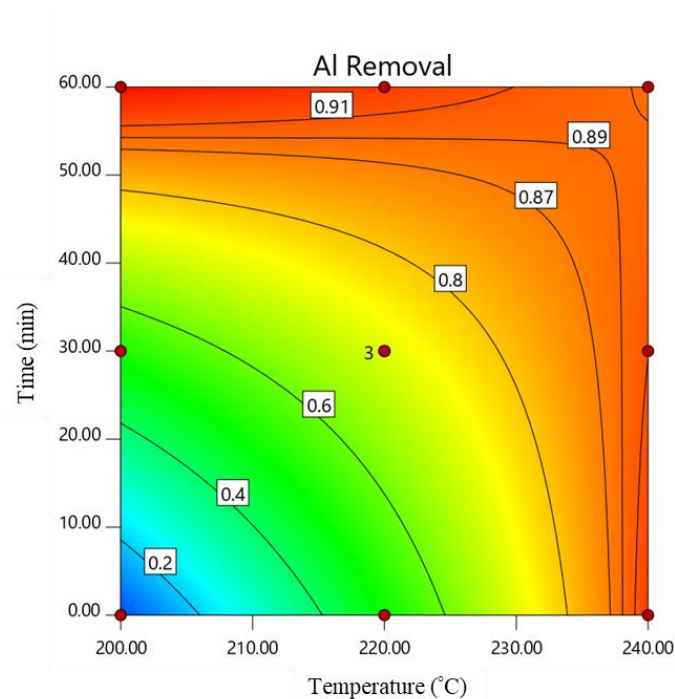
In general, volatile matter and ash in hydrochar decreased as the operating temperature and the holding time increased. On the other hand, fixed carbon increased with an increase in these experimental conditions. The paper part of Aseptic Packaging, which composed of cellulose, will undergo several thermochemistry reactions such as decarboxylation, the volatilization, the hydrolysis, and the breakdown of cellulose (Peterson et al. 2008). The hydrothermal treatment also turns volatile organic compounds into carbon which increases the percentage of fixed carbon (Zaini et al. 2017). This behavior

was expected, as was mentioned in previous researches (Prawisudha et al. 2012; Areeprasert 2015; Novianti et al. 2016a).



**Figure 2.7** From left to right, response surface contour of volatile, fixed carbon and ash content (% dry base)

The purity of hydrochar is essential in terms of solid fuel utilization. Especially when contaminant such as aluminum is one of the contributors in slagging and fouling inside the reactor. Figure 2.8 shows the aluminum removal performance which shows that, up to 99% of aluminum is removed and formed a composite with polyethylene. The composite was easily formed in higher temperature and longer reaction time.



**Figure 2.8** Aluminum removal performance

### 3.2. Composite of Aluminum and Polyethylene Properties

The experiment of the hydrothermal treatment of Aseptic Packaging with the operating condition of 200°C and 0 minutes (No. 1 in the standard order) gave poor results since the composite was not formed and the plastic and paper fractions were only partially deteriorated. Figure 2.9 shows the comparison of the experiment results of the experiments No. 1,2,3, and 4.

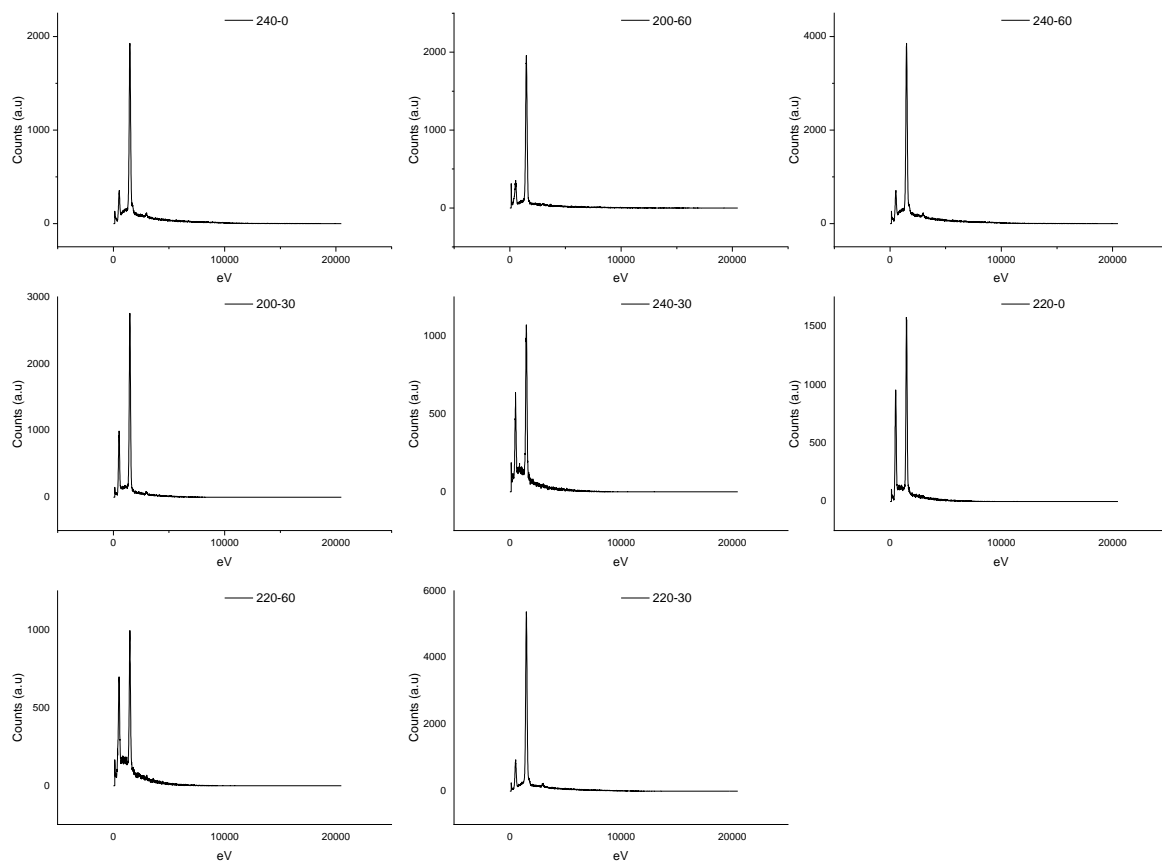


**Figure 2.9 Appearance of the composite formed after the hydrothermal treatment (From left to right, 1-4)**

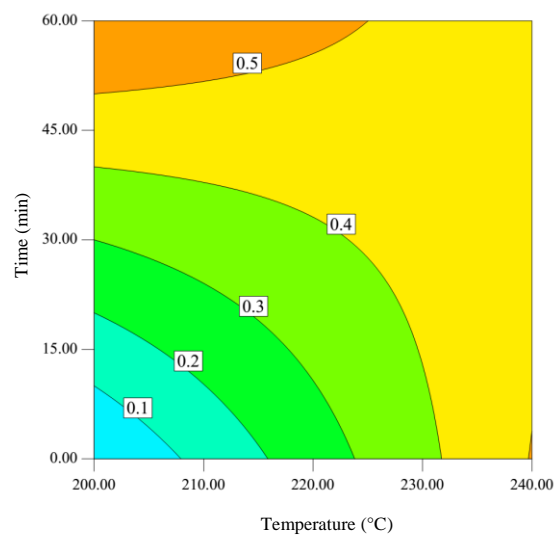
The ineffectiveness of the first experiment was caused by insufficient reaction temperature and time to break cellulose, melt polyethylene, and gather the polyethylene and aluminum into one floc. The minimum requirement of the hydrothermal treatment for composite forming was 200 °C and 30 minutes. Several tests, such as EDS and DTA, were conducted to predict the aluminum content. The composite was also weighed and burnt at 900 °C to determine the actual aluminum content.

In general, the amount of aluminum will increase when the operating temperature and time increase. At 240 °C and 60 minutes, the concentration slightly decreased due to hydrochar, which attached to the material surface. All EDS test confirm the presence of aluminum in the composite (figure 2.10). The aluminum concentration is presented in figure 2.11

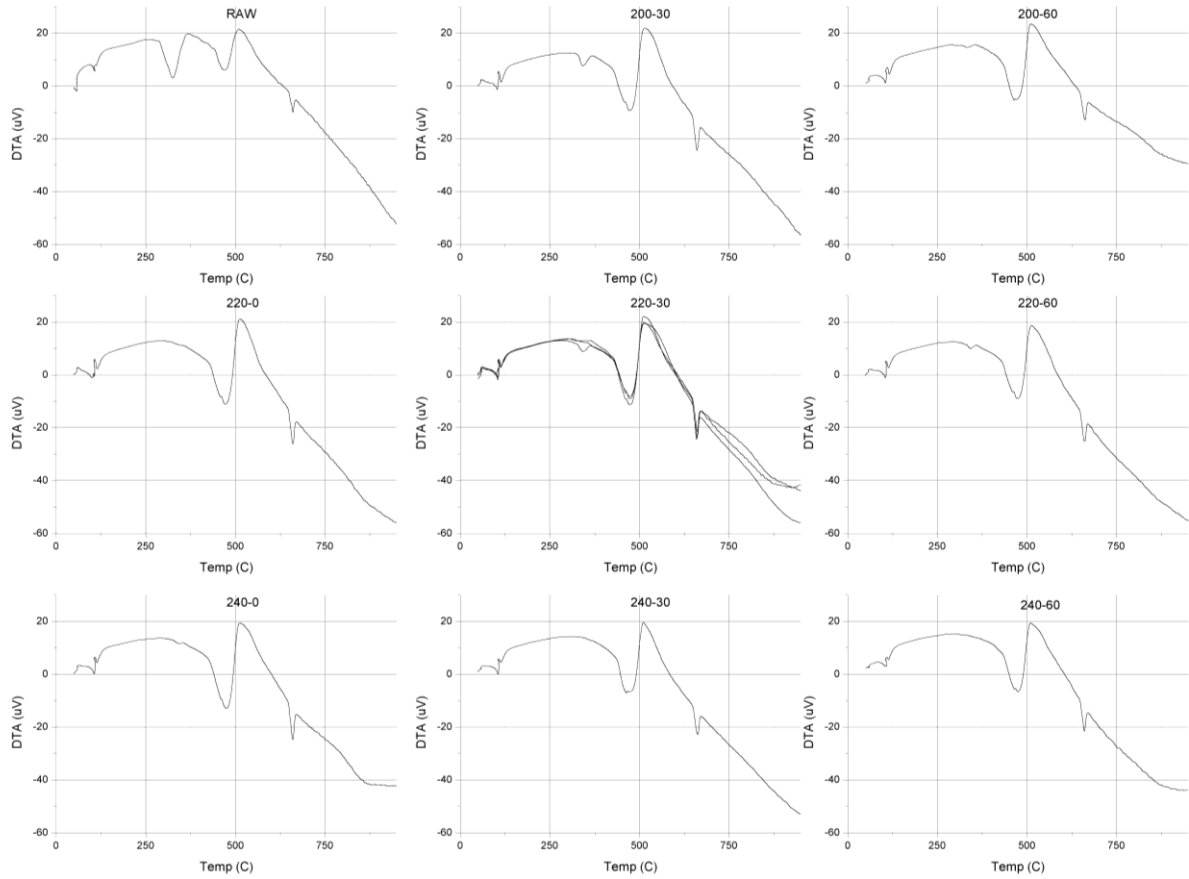
DTA spectra (Figure 2.12) of raw material indicates that few materials were detected as four endothermic peaks. The first peak shows the evaporation of water and the melting of polyethylene (Peterson et al. 2001). The second peak indicates the presence of cellulose corresponding to the melting point of cellulose (Zaini et al. 2017). The third peak is the decomposition peak of polyethylene (Peterson et al. 2001). The last peak is the melting point of aluminum at 660 °C (Schmitz 2006).



**Figure 2.10 EDS spectra of the surface of the composite**



**Figure 2.11 Aluminum content on the surface of the composite by the fraction**

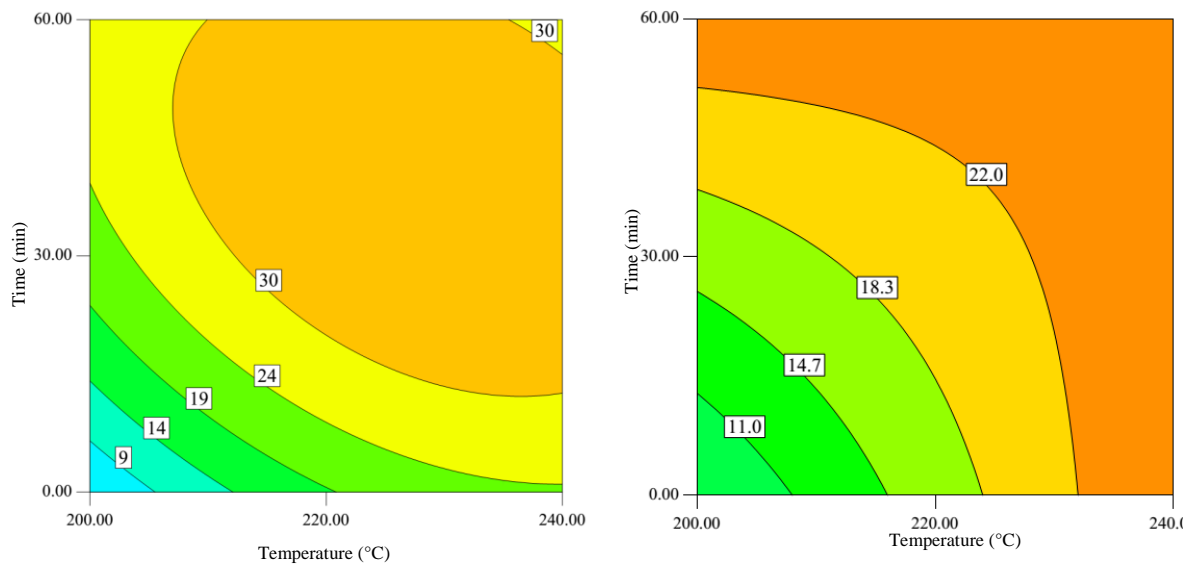


**Figure 2.12 DTA spectra of the composite**

DTA spectra in Figure 2.12 show the similarity of thermal properties products among all experimental conditions. The peaks below 200 °C are polyethylene melting conditions. The downward bend occurring between 400 °C and 600 °C is caused by the endothermic reaction, such as the dehydration, the phase transformation, or the decomposition. In this case, it was the decomposition temperature of polyethylene. The endothermic peaks at 660 °C are caused by aluminum melting. Those spectra confirm the presence of aluminum and polyethylene in the composite.

As mentioned before that the presence of aluminum in the composite is absolute. After removing the mass of contaminants by burning the samples at 950 °C, the leftover ash minus the ash of contaminants was predicted as the actual aluminum content. Figure 2.12 shows the response surface plot of the ash content and the actual aluminum yield of the composite. Temperature and time have a positive effect on the rise of the aluminum yield and the ash content. This behavior is aligned with the behavior of the

aluminum content on the surface of the composite, as mentioned before. The highest yield was 37%, at a temperature of 220 °C with 30 minutes holding time.



**Figure 2.13 Response surface plot of actual aluminum yield (%) and ash content (%) of the composite.**

Compared to the composite of aluminum and polyethylene produced in a paper mill, the hydrothermal treatment could produce a composite with a higher aluminum concentration. The composite of LDPE produced in Brazilian paper mill yielded 15% of aluminum (Lopes and Felisberti 2006).

**Table 2.5 Density of the composite**

Temp	Time	Density(g/cm <sup>3</sup> )
200	0	0.582804
240	0	0.59642
200	60	0.682171
240	60	0.757373
200	30	0.821962
240	30	0.819213
220	0	0.930962
220	60	0.832299
220	30	0.772879
220	30	0.836541
220	30	0.71981



Table 2.5 shows the density of the composite, measured by a pycnometer. The trend indicates that the increase in the operating temperature will increase the density of the composite. The highest density of 0.93 g/cm<sup>3</sup> was obtained at a temperature of 220 °C with 0 minutes of holding time. The composite produced in Brazilian paper mill also has a similar density of 0.9 g/cm<sup>3</sup>. The response surface of composite density is shown in figure 2.13. Figure 2.14 shows roughly the behavior of the composite in water column.



**Figure 2.14 Different of material density shown by floating aluminum and submerging chars in the water column**

#### **4. Conclusion**

The hydrothermal treatment of Aseptic Packaging was useful in separating the hydrochar and composite of aluminum and polyethylene. The analysis was conducted, focusing on understanding the quality of solid fuel and aluminum recovery after the hydrothermal treatment. It was observed that the reaction temperature and time have a positive effect on the improvement of the quality of the solid fuel and the aluminum yield. The hydrothermal treatment could increase the HHV of biomass from Aseptic Packaging up to 25.22 MJ/kg, which is comparable to lignite and coal. EDS and DTA spectra confirm the presence of aluminum. The aluminum yield in the present process was up to 37%.

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## Chapter 3

### Landfill Mining and Excavated Waste Characteristics

#### 1. Introduction

Despite the high development of alternatives disposal means, currently, a landfill still becomes the most popular option for waste disposal in the form of open dumping and sanitary landfill. According to UNEP, in Asia, open dumping and sanitary landfill have a very high share of 51% and 31%, respectively, as waste disposal means. On the other hand, incineration contributes to only about 5%, while recycling contributes to about 8% of the total. Also, in Africa, about 47% of waste disposal is adopting open dumping, and about 29% is employing sanitary landfill.

Furthermore, in North America, about 91% of the waste disposal method is conducted by using sanitary landfills (UNEP (United Nations Environment Programme) 2015). It shows that most developing countries rely on the landfill as a disposal facility for their municipal solid waste (MSW) as it costs less than another method (Tchobanoglous and Kreith 2002). The operation of landfill sites leads to the decrease of environmental quality of the surrounding area from smells and leachate, which is a major source of heavy metals contamination to the environment (Dhaliwal et al. 2019). On a global scale, landfill sites released methane, which has a worse effect than CO<sub>2</sub> to global warming (Cherubini et al. 2009). Thus, a method and technology which can reduce the effects and bring an optimal benefit significantly are urgently demanded.

Methane produced in a landfill is coming from the decomposition of degradable organic content (DOC) of inputted waste material (Rees 1980). Capturing methane gas is one of the solutions to reduce greenhouse gas emissions. The methane is recovered and used for power generation by using a gas engine. Once the methane is depleted, undecomposed organic carbon residue and inert material remain. Therefore, in order to recycle the left residue, landfill mining has been proposed and developed (Dickinson 1995). Although the idea of landfill mining initially appeared in the 1950s, its research and development became relatively intensive since the 1990s due to new environmental regulation, which became stricter than the older one (Krook et al. 2012). The development of more advanced technologies

for waste treatment and recycling in early 2000 has promoted the extended concept of landfill mining in Europe (Hogland et al. 2010).

In Europe, Closing the Circle project had been launched to build a pilot project in the REMO landfill site, Belgium (Hogland et al. 2010). The cost feasibility of landfill mining will only work on particular condition, such as availability of special funds for remediation, availability of feedstock to make sure that the waste-to-energy (WtE) plant is running in its full capacity and the presence of cement company which will buy the solid fuel from the processed material (Hull et al. 2005).

The excavated waste (EW) is highly heterogenic and mainly contains fine particles such as soils and degraded organics (Hogland 2002). Wet jigging could classify EW based on the different densities of the waste constituents and remove the finest particles from the surface of the material (Wanka et al. 2017). Recyclable materials recovered from landfill mining are low-quality; therefore, more efforts to recover and utilize are urgently required. Many landfills in Europe contain waste with a calorific value of up to 20 MJ/kg, leading to high feasibility to build a WtE plant for EW (Hogland et al. 2004). The excavated plastic waste could be pyrolyzed into the oil with characteristics close to petroleum diesel oil (Canopoli et al. 2018). Zaini et al. (2019) suggest a smaller sieve opening size for landfill waste separation processes to increase the quality of EW derived fuel. High organic matter in fine fraction and soil-like material should be removed by physical or biological treatment before utilization. Anaerobic treatment could stabilize the soil-like material as well as generate methane. Washing the material could remove the organics faster but could overburden the leachate treatment (Mönkäre et al. 2019). While European countries generally have a constant waste composition, the developing countries, such as Indonesia, have gone to a rapid change of development, resulting in a dynamic change on waste composition and make it harder to predict the characteristic of its landfill wastes (Damanhuri and Padmi 2016).

Industrial landfill site containing lime milk has been mined and utilized for cattle feed ingredient. In Brazil, a missed proper pretreatment from the recovery process leads to dioxin contamination in cattle products (Torres et al. 2013). Korea has attempted to convert landfilled WtE through landfill mining and reclamation (LFMR) projects, which is converting the landfills into reusable landfills in five cities (KECO (Korea Environment Corporation)). Japan uses a bioreactor to stabilize the EW from illegal

dumping sites and recover the energy. Higuchi (2005) has developed a system to properly reclaim waste from old landfills from survey mechanisms, technology selection, safety measures during excavation, waste recovery options until the re-utilization of landfill sites.

Furthermore, waste is a product of human activity. Every region has different activities and cultures, resulting in different waste compositions. Assessments concerning spatial aspects, infrastructure readiness, and regional policy are necessary for landfill optimization projects.

Indonesia has a vast population (about 260 million in 2017) and represents exponential economic growth (5.07%), which is followed by the significant increase of waste generation (67 Mt) (Damanhuri 2017; BPS 2018). The urbanization has given a considerable load in waste treatment and management because of the limited capacity of a landfill in the big cities. As the most densely populated island in Indonesia, Java Island has 61 active landfills in operation, which are receiving wastes from 119 cities. However, the number is far from being sufficient, and those landfills are facing several problems (PT. Arkonin Engineering MP 2015). Land demands for residential, commercial, and industry are increasing every year, make it hard to find a new landfill site. Indonesia's government has ratified aggressive policy to escape from dependency to landfill by providing incentives and tipping fees for the WtE plant (Government 2018). Unfortunately, many cities received a rejection from their citizens for building the plant in their neighborhood. They also have a problem to provide stable waste input since their low efficiency in the collection system (Purwaningsih 2012; Dodi and Raharjo 2015; Thohiroh et al. 2017). The old landfill could be used as an alternative supply for the WtE plant by excavating, segregating, and processing the wastes (Anex 1996). The organic material from landfills is readily fermented to form organic carbon chain material with a higher energy density. The plastic is already fragmented, and the chlorine was leached out (Rees 1980; Wall and Zeiss 1995). Both are suitable for energy recovery.

Material flow analysis (MFA) is one of the assessment methods to provide a holistic review of the landfill mining process by considering waste composition, flows, treatment processes, and regional infrastructure (Islam and Huda 2019). The analysis could be done before, during, or after the project involving different assumptions (Wittmaier et al. 2009). For analysis before the existing project, MFA was included for feasibility studies. Process modeling is necessary since no actual data is available yet.

By integrating the process modeling in MFA, reliable inventory data could be obtained (Wang et al. 2015).

However, previous studies on landfill mining project lacked an investigation on chemical characterization of the landfilled waste and environmental impacts of the landfill waste utilization (Hölzle 2019; Yi 2019). The feasibility of EW utilization depends on the quality of the recovered waste and technologies which are available for utilization. Hence, this paper focuses on the characterization and utilization potential of the waste from the Jatibarang site in Semarang City (Central Java) and the Piyungan site in the Bantul Regency (Special Administrative Region of Yogyakarta). The records from authorities provide limited information about sites condition and waste characteristics. The exploratory field tests have been carried out in Jatibarang and Piyungan sites. Both sites receive waste from the residential area, commercial area, and industries (Lokahita et al. 2018). Chemical characterization, such as proximate and ultimate analyses of EW, was used as input in process modeling to calculate the energy and emission potential of WtE utilization. Furthermore, the material and energy flows of the LFM operations, such as excavation, sorting, transportation, processing, and restoration was done to provide better insight related to factors influencing the execution of landfill mining.

## **2. Site description and methods**

### **2.1. Jatibarang landfill in Semarang City, Central Java Province**

Semarang is the capital of Central Java Province, which is located on the north coast of Java (about 540 km east of Jakarta). Semarang City Environment Agency is the stakeholder who has the full responsibility of solid waste management in the Semarang City area. The total volume of waste that was produced from this city was about 5,080 m<sup>3</sup>, and the total weight was about 1,270.13 Mg/d, according to the survey conducted in 2016. About 77% of the wastes were collected and then transported to the landfill, while the rest of 23% was either scavenged, collected by the waste bank, and unmanaged [10,11].

Jatibarang landfill has been in operation since 1992 and is located 13 km from the downtown of Semarang City. The total area is 460,183 m<sup>2</sup> and surrounded by a forest green belt as a buffer zone. 850 Mg/d of wastes come to the landfill, and most of them are organic wastes. This landfill also has a fertilizer factory that can produce about 250 Mg/d of granule fertilizer (Lokahita and Takahashi 2017; Lokahita et al. 2019). Also, the leachate generated from the landfill is collected in the leachate pond and treated initially before it is dischargeable to the river. The landfill gas is also collected and utilized as kitchen fuel by surrounding residents

In order to observe the characteristics of wastes coming to the landfill, a preliminary survey has been conducted. In the seven days of the monitoring campaign, the waste truck was randomly chosen, and one bucket (~1 m<sup>3</sup>) of waste was sampled for composition analysis. Municipal wastes coming to the Jatibarang landfill consist of kitchen and garden waste (50%), paper (8%), textile (9%), plastic (26%), and metal (1%).

## 2.2. Piyungan landfill in Bantul Regency, Special Administrative Region of Yogyakarta

Piyungan landfill acts as a regional landfill receiving waste from Yogyakarta City, Bantul Regency, and Sleman Regency. It is located in the northeast area of Bantul Regency, near the border of Yogyakarta City. About 350 Mg/d of wastes come to this landfill, covering a total area of 125,000 m<sup>2</sup>. The landfill has been operated since 1995 and already filled up its capacities. Yogyakarta City takes 50% of Piyungan landfill capacity, followed by the Sleman Regency for 35% and Bantul Regency for 15% (Sari 2015). Previous studies about the incoming waste to Piyungan landfill shows that the organic waste dominates up to 77%, followed by plastic (10%), paper (6%), textile (2%), metal (1%), and others (4%) (Syamsiro 2015).

## 2.3. Sampling Method

For the Jatibarang case, nine different points spreading throughout the landfill area were randomly selected and excavated using a mid-size excavation machine for about 3–4 m. On the other hand, the sampling process in Piyungan utilized drilling rig to drill for every 1 m up to 12 m in four different locations.



The EW was sieved in a 10 mm mesh to remove soil-like (SL) material. Then, the bulky material was manually sorted for composition analysis into organic, wood, paper, plastic, textile, metal, and glass. The samples were stored in a plastic vacuum bag for further analysis.

Moreover, organic, plastic, textile, and paper were categorized as combustible waste (CW), while metal and glass were categorized as non-combustible waste (NC). The CW will be analyzed for proximate analysis, ultimate analysis, and high heating value (HHV). The proximate analysis was adopted to understand the percentage of moisture, volatile, fixed carbon, and ash. It was performed using Shimadzu D60 TGA/DTA Analyzer based on ASTM D3172. In addition, the ultimate analysis determines the percentage of carbon, hydrogen, oxygen, nitrogen, sulfur, and ash in the material. The analysis was conducted on a dry basis after removing the moisture, using Vario MICRO Cube elemental analyzer (Elementar, Germany). Moreover, the HHV of CW was determined using Autobomb Calorimeter (Gallenkamp, England).

#### 2.4. WtE Process Modelling

Investigated WtE plant consists of a combustion chamber, heat recovery system, and gas cleaning process. There are several stages during the incineration process, such as drying and degassing, pyrolysis and gasification, combustion, and post-combustion. Those processes occupy almost the same space and time in the reactor and react almost instantly. The design of the reactor, feed characteristic, and air supply influence the performance of the incineration process in regards to energy efficiency and pollutant emission.

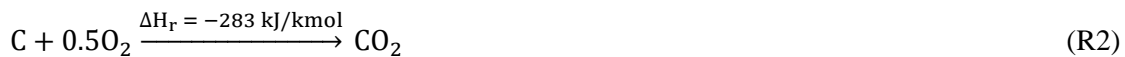
The thermochemical model flowsheet of the incinerator is shown in Figure 3.1. The model was built and calculated using Aspen Plus V8.8® (Aspen Technology, Inc). Unit operation and the chemical reaction were modeled by interconnecting the block using the material, work, and energy streams. A steady-state simulation was modeled in where all of the solid and gaseous phases are thermodynamically equilibrium, and the considered reactions can achieve chemical equilibrium. By adjusting parameters like excess oxygen rate and combustion temperature, performance analysis of the incineration process could be studied.

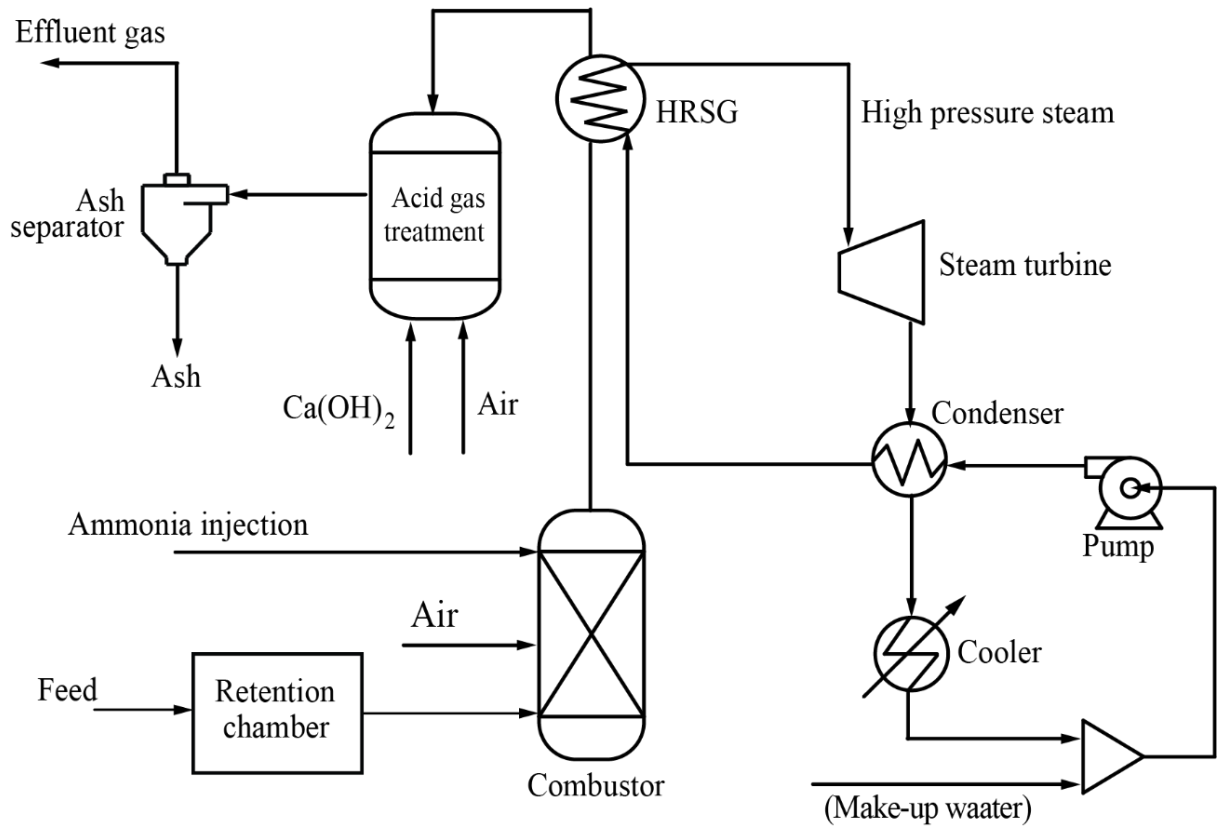
The combustion reactor model was divided into three parts: decomposition, combustion, and post-combustion. In the decomposition zone, each material was decomposed using RYIELD block into its constituent elements, covering carbon, hydrogen, oxygen, nitrogen, sulfur, chlorine, ash, and moisture in STP conditions. RYIELD block is used to model the reactor when the information related to reaction stoichiometry and kinetics are unavailable. The yield composition was determined using a calculator block with Fortran script from the ultimate and proximate analyses. The decomposition heat is transferred into the burner operating unit.

RGIBBS block used in the combustion zone is a rigorous reactor that could calculate the chemical and phase equilibrium through the minimization of Gibbs free energy. It can be used to predict possible product composition. The equation for the RGIBBS reactor is shown as:

$$\text{Min} G, G = \sum_{j=1}^S n_j^c G_j^0 + \sum_{j=S+1}^M \sum_{l=1}^P n_{jl} G_{jl} \quad (1)$$

G is the system total Gibbs free energy, S is a number of single phases, c is component number of single phases, M is the total number of phases, and P is the number of components. In the RGIBBS reactor, the constituent elements of EW are mixed with air to produces H<sub>2</sub>O, N<sub>2</sub>, NO, NO<sub>2</sub>, O<sub>2</sub>, H<sub>2</sub>, C (solid), CO, CO<sub>2</sub>, Cl<sub>2</sub>, HCl, S, SO<sub>2</sub>, and SO<sub>3</sub>. Below are the assumed main reactions in the process.



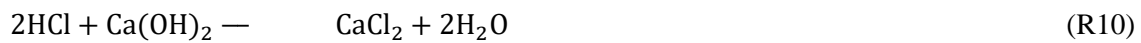


**Figure 3.1 Process flow diagram of the incineration model**

In the post-combustion zone, the removal of  $\text{NO}_x$  is performed by ammonia injection in the process, which is called a selective non-catalytic reduction (SNCR). SNCR typically achieves  $\text{NO}_x$  removal efficiency between 20% to 60% with stoichiometric ratios of 1-2 (Falcone Miller and Miller 2010). The reaction in the process is shown as follows.



Acid gases such as  $\text{SO}_x$  and  $\text{HCl}$  are removed in dry scrubber employed with an alkaline reagent such as  $\text{Ca}(\text{OH})_2$ . Dry scrubber reactor was modeled using the RSTOIC block based on the fractional conversion of the reaction by 0.9 (Ting et al. 2008).



The excess heat from the hot gas stream could be used for cogeneration or rotating the steam turbine. In the developed model, hot gases after the combustion process are moved to the heat recovery steam generator (HRSG) unit. In this unit, the hot gas stream is recovered using a heat exchanger to transform water into steam. The steam is used to generate electricity via a steam turbine. The proposed model used the HEATER block for the HRSG process and COMP block for the steam turbine process. HEATER block can calculate the amount of transferred heat to reach a designated temperature of outlet gas. COMP block will calculate the amount of work produced or needed for changing the pressure. In this study, the isentropic method is adopted with an efficiency of 80% for COMP block calculation (Kehlhofer et al. 2009).

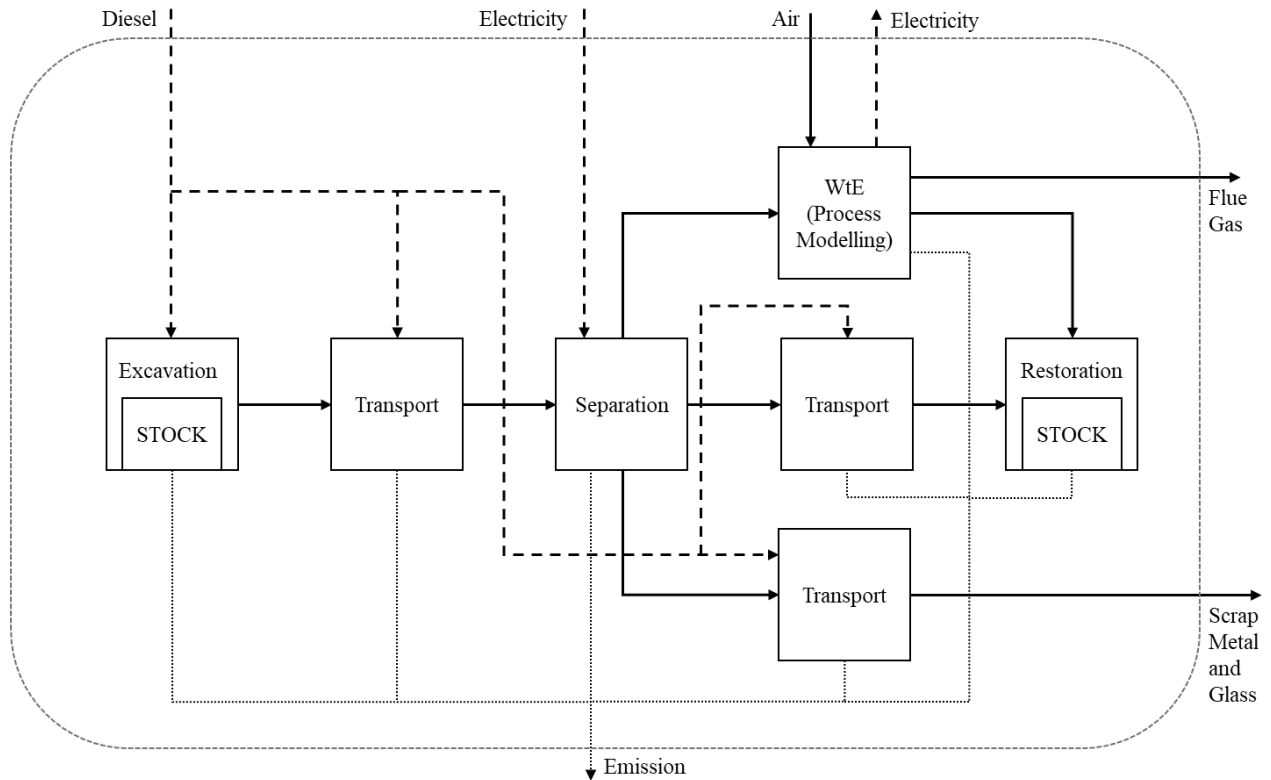
After passing the steam turbine, the steam will expand, leading to the lower pressure and temperature. The steam is going to the condenser, which is simulated using HEATER block to even lower temperature and turns back into the water, and then, it is transferred to the reservoir to be used again. This water cycle process could reduce water usage during the process.

## 2.5. Material Flow Analysis (MFA)

The Indonesia Presidential Decree No. 35/2018 demands the WtE power plant to operate with a minimum capacity of 100 Mg/d. MFA is used to analyze the quantity of necessary excavated material to comply with the government standard. STAN 2.0© (TU Wien, Institute for Water Quality, Resource and Waste Management) was used to conduct a material flow analysis on the landfill case study to understand the potential of landfill resource recovery and recirculation. The process modeling resulted from process modeling simulation is integrated into the MFA under the WtE process.

The system boundary begins at the excavation process, where the stock material is gradually extracted. The landfill process consists of excavation, transportation, waste separation, WtE, and residual restoration, as shown in Figure 3.2. Firstly, the landfill site was excavated using a mid-size excavator with a 133 hp engine and 1 m<sup>3</sup> bucket size. 100 hp dump truck with the 4 m<sup>3</sup> container was used for transporting the samples between the excavation site to the processing site. The processing site consists of separation and WtE units. The separation unit is equipped with star-screen to remove the soil-like fraction and conveyor belt for manual sorting of CW and NC. Bulldozer and roller vibro were used,

restoring the soil-like fraction from the separation process and ash from the WtE plant. NC, such as metal scrap and glass, were transported to the recycling facility outside the landfill site.



**Figure 3.2 Process of landfill mining and the input/output flow of mass, energy, and emission**

### 3. Excavation Results

The excavation process in both landfills can be seen in Figures 3.3 and 3.4. During the excavation in the Jatibarang landfill, 397 kg of waste was sampled and analyzed. Because of the poor infrastructure in the landfill area and intense heat from the weather, it took about one week to finish the excavation. The average humidity and ambient temperature during the excavation were 63% and 28 °C, respectively. The benefit of the excavation method lies in its capability to procure a large number of samples, but the depth of the excavation is limited to the length of the excavator.

Figure 3.3 summarized the results of excavation in the Jatibarang landfill. The soil fraction (<10mm) exhibits 52% in the Jatibarang landfill. In addition, the CW exhibits 45% of the EW, which consists of 18% organic and 27% plastic, while the rest is an NC waste (metal and glass).

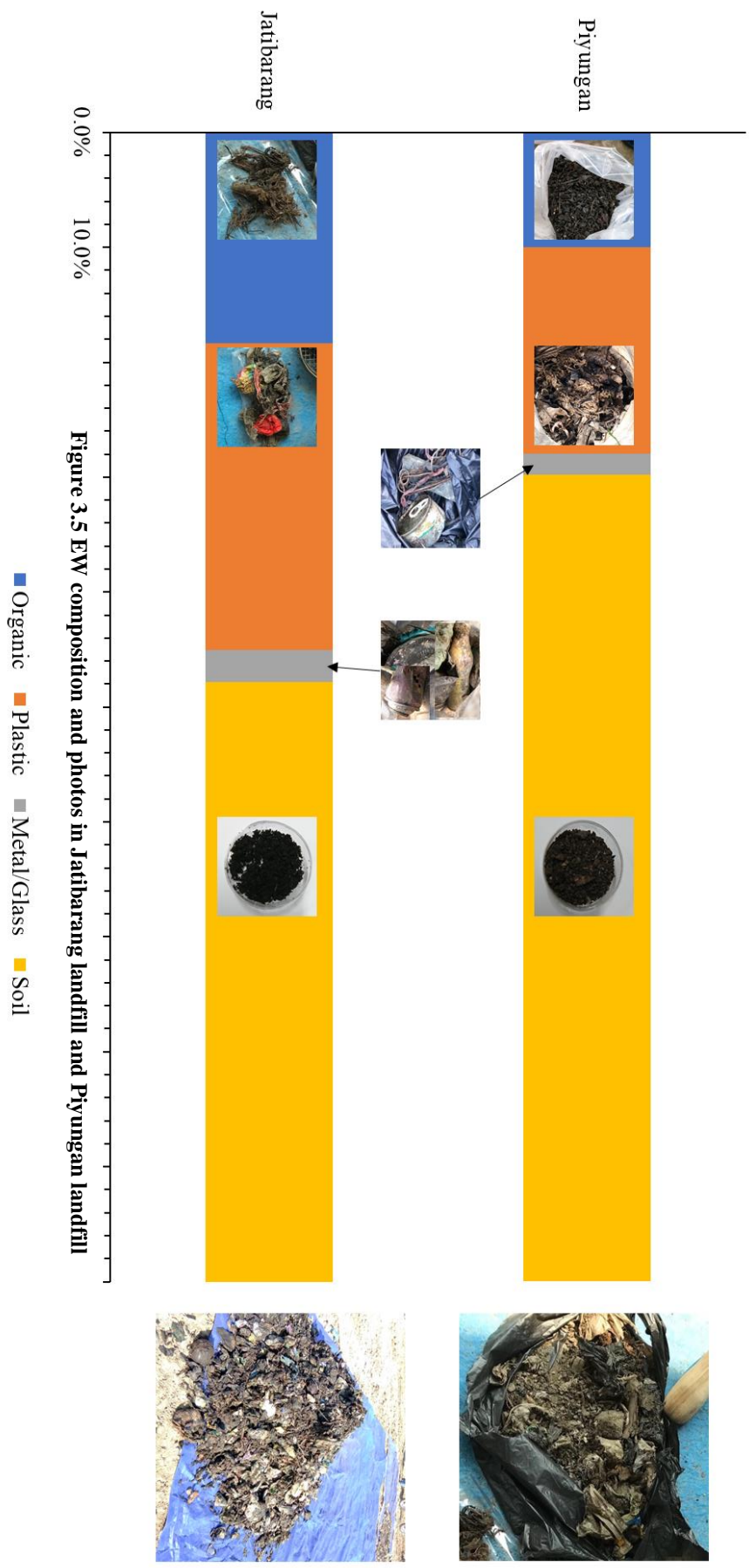
On the other hand, the Piyungan landfill was not designed for sanitary landfills, and the bottom of the landfill is a natural bedrock from the landscape. Drilling was done every one meter until reaching the bedrock layer in 12 m. The total of 40 kg of the sample with the density varied from 115 to 618 kg/m<sup>3</sup> was obtained. The average ambient temperature and humidity during the sampling process were 29 °C and 78%, respectively. The excavation results in the Piyungan landfill are also presented in Figure 3.3. The soil fraction (<10mm) exhibits 70% in the Piyungan landfill. The CW exhibits 28% of the EW, consisting of 10% organic and 18% plastic, while the rest is NC wastes (metal and glass).



**Figure 3.3 Excavation process in Jatibarang Landfill**



**Figure 3.4 Excavation process in Piyungan Landfill**



### 3.1. Characteristic of CW

**Table 3.3 Characteristic of EW**

Parameter	Jatibarang landfill	Piyungan landfill
Proximate Analysis		
Moisture (wt%)	32.20	33.63
Fixed Carbon (wt%, dry)	7.58	17.99
Volatile (wt%, dry)	61.01	33.46
Ash (wt%, dry)	31.41	48.55
Ultimate Analysis (wt%, dry)		
Carbon	37.03	25.46
Hydrogen	5.16	3.32
Nitrogen	1.30	1.12
Chlorine	0.86	0.55
Sulfur	0.67	0.55
Oxygen	23.57	20.45
HHV (MJ/kg)	24.28	12.39

The CW sample was tested for proximate analysis, ultimate analysis, and HHV. The analysis results are summarized in Table 3.3. The moisture, volatile, fixed carbon, and ash in the proximate analysis of Jatibarang landfill CW were about 32.2%, 61.01 dry%, 7.58 dry%, and 31.41 dry%. On the other hand, Piyungan landfill proximate analysis shows different results with 33.63% of moisture, 33.46 dry% of volatile, 17.99 dry% of fixed carbon, and 48.55% of ash. The ultimate analysis shows that the Jatibarang landfill has a higher value in all parameters than the Piyungan landfill. Bomb calorimeter shows higher HHV for the Jatibarang landfill at 24.28 MJ/kg. Waste composition of CW and ash percentage are essential in the quality of EW as solid fuel utilization. Jatibarang landfill has a higher percentage of plastic in the CW composition resulting in higher calorific value and chlorine content. The ash content



also might be influenced by dirt from soil-like particles, which are hard to remove. Additional pretreatment such as washing could remove the contaminant, but produces wastewater and more energy for drying. More advanced pretreatment techniques such as hydrothermal treatment not only could reduce the ash content but also homogenized the feedstock, increase calorific value, upgrade the carbon content, and improves drying performance (Lokahita et al. 2017; Triyono et al. 2019).

#### **4. Process Simulation Results**

The process modeling simulation was performed by using Aspen Plus ® by Aspen Tech to estimate the power output and emission generated from EW thermal conversion. The excess oxygen in the combustion chamber was set to 20% to ensure the complete combustion of EW. The concentration of calculated emission compounds from the incineration flue gas is shown in Table 2. Jatibarang landfill case has a lower CO<sub>2</sub> concentration, which is 7672.4 mg/Nm<sup>3</sup>, compared to the Piyungan case, which is 84759.06 mg/Nm<sup>3</sup>. SNCR for NO<sub>x</sub> removal effectively reduces NO<sub>2</sub> concentration of both the Jatibarang case and Piyungan cases into 1.27 mg/Nm<sup>3</sup> and 1.28 mg/Nm<sup>3</sup>, respectively.

The acid gas removal system was also successfully reducing the HCl and SO<sub>2</sub> concentration of Jatibarang and Piyungan cases to meet the Indonesian government standard (HCl = 10 mg/Nm<sup>3</sup>, SO<sub>2</sub> = 210 mg/Nm<sup>3</sup> (Ministry of the Environment and Forestry of The Republic of Indonesia 2019). The chlorine content of the feedstock is affecting the combustion process. Not only from the high percentage of plastic waste but also from organic waste, which absorbs the leachate and carries out the minerals (Rees 1980).

In terms of electricity production, EW from Jatibarang landfill could produce higher power compared to the Piyungan landfill, but the emission is also higher. Although the power output is lower in the Piyungan landfill, the plant efficiency is higher than the Jatibarang landfill by 20% for the Piyungan landfill and 14% for the Jatibarang landfill. In response to the higher efficiency, the greenhouse gas emission from the Piyungan case proves to be lower than the Jatibarang case. The summary for power output and emission from both cases is also shown in Table 3.4.

**Table 3.4 WtE Simulation Result**

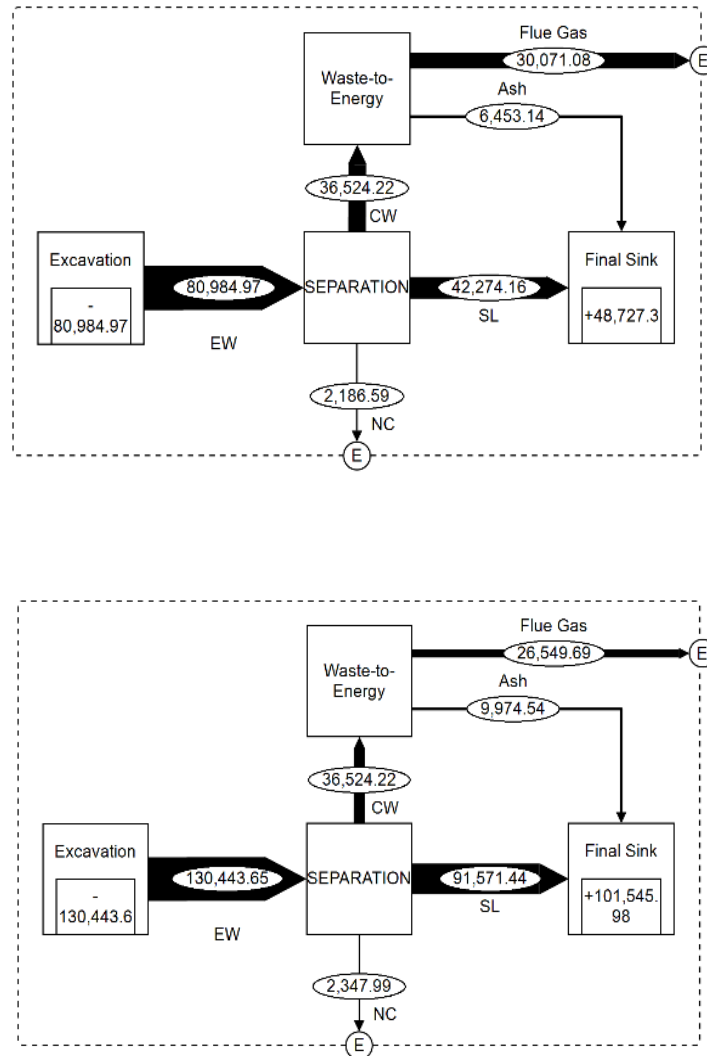
Flue Gas	Jatibarang Case	Piyungan Case
CO <sub>2</sub> (mg/Nm <sup>3</sup> )	76472	84759
Cl <sub>2</sub> (mg/Nm <sup>3</sup> )	12.16	9.62
HCl (mg/Nm <sup>3</sup> )	2.85	2.59
SO <sub>2</sub> (mg/Nm <sup>3</sup> )	71.93	100.23
NO <sub>2</sub> (mg/Nm <sup>3</sup> )	1.27	1.28
Power Output (MW)	3.73	2.83
CO <sub>2</sub> -e (Mg/d)	108.46	84.61

## 5. MFA Results

### 5.1. Mass Balance

MFA was conducted using STAN 2.0 with an initial target to obtain 100 Mg/d of CW as a feedstock for the WtE plant. After inputting the result of excavation and process modeling simulation, the material flow analyses revealed that there is a large number of resources that are potentially recoverable from the landfills in order to lengthen the landfill lifespans.

For the Jatibarang case, 221.73 Mg/d of landfill waste was excavated to maintain 100 Mg/d of CW as WtE supply, resulting in an annual excavation of 80,894.97 Mg. The EW was brought to the separation area to be sorted into CW, SL, and NC. 6,453.14 and 42,274.16 Mg/yr of ash and SL are stored in ex-landfill site for the final sink. 2,186.59 Mg/yr of NC, which consists of scrap metals and glass, was transported to the off-site processing facility. The Sankey diagram for the Jatibarang case MFA is presented in Figure 3.4 (top). As for the Piyungan case, from 130,443.6 Mg/yr of EW, 36,524.22 of CW



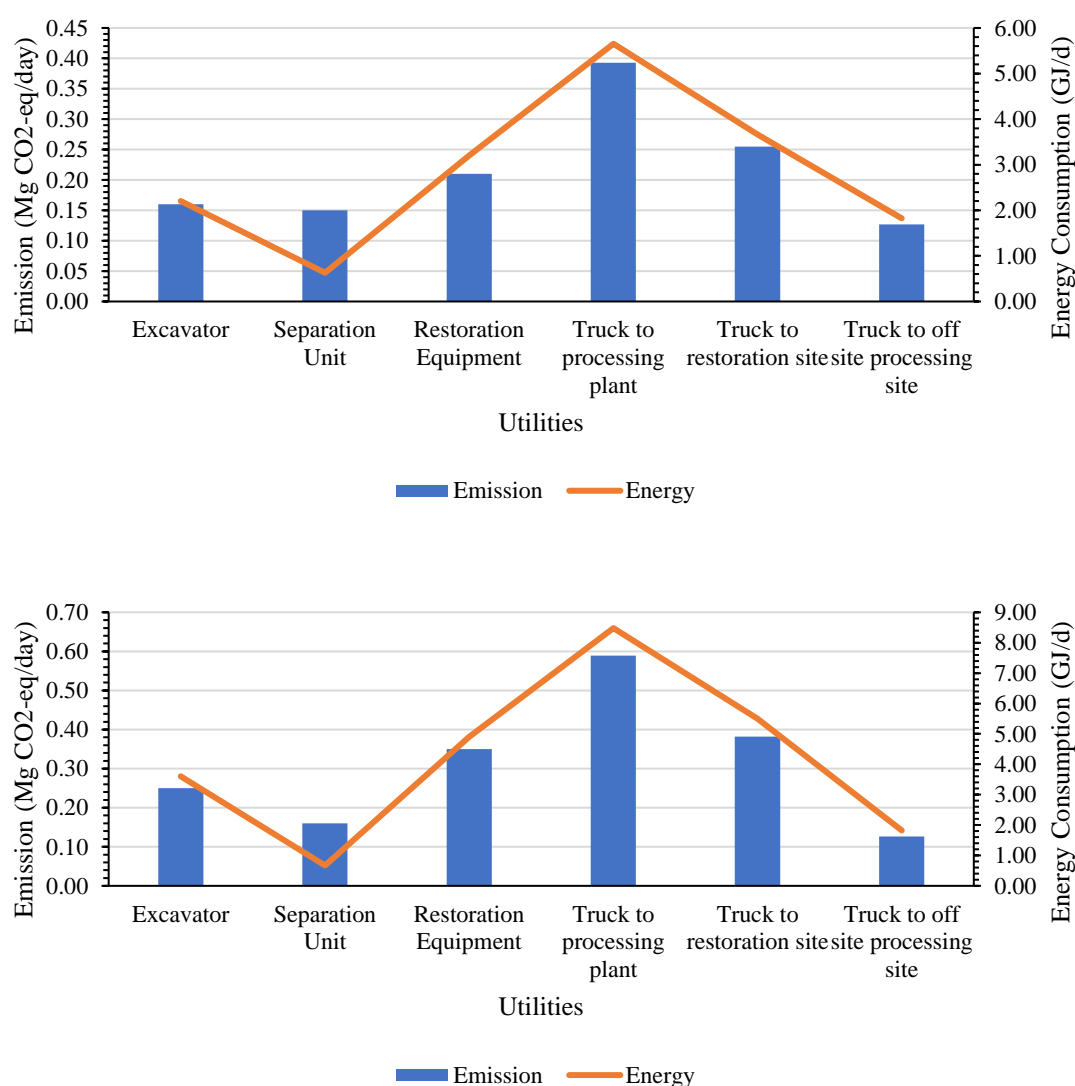
**Figure 3.6 Sankey Diagram of Jatibarang Case (top) and Piyungan Case (bottom).  
Mass unit in Mg/yr (ton per annum)**

is going to WtE plant and 91,571.44 of SL and will be stored in the final sink together with the ash from WtE plant (9,974.54 Mg/yr). The complete MFA Sankey diagram is shown in Figure 3.4 (bottom).

Similar to an investigation by Hogland et al. (2019), Jatibarang and Piyungan landfills also contain a high proportion of soil-like material. The soil-like fraction is the mixture of soil cover, which is divided into each cell and decomposed as an organic fraction. In the landfill, an organic compound such as food waste or wood reacts faster than others in the anaerobic environment because they consist of cellulose which is readily fermented to form simpler organic carbon chain material with a higher calorific value and is suitable for energy recovery (Wall and Zeiss 1995; Anex 1996).

## 5.2. Energy consumption and emission

Utilities that are involved in the landfill mining process consist of the dump trucks, excavator, bulldozer, roller vibro, star screen, and belt conveyor. The utility specification was based on the existing equipment in the Jatibarang landfill. The dump truck has a 100 hp engine with a diesel consumption of 0.46 GJ/h and CO<sub>2</sub> emission of 32 kg CO<sub>2</sub>-e/h. Excavator has a 133 hp engine with a diesel consumption of 0.60 GJ/h and CO<sub>2</sub> emission of 42 kg CO<sub>2</sub>-e/h. Bull Dozer has an 82 hp engine with a diesel consumption of 0.37 GJ/h and CO<sub>2</sub> emission of 26 kg CO<sub>2</sub>-e/h. Roller Vibro has a 100 hp engine with a diesel consumption of 0.46 GJ/h and CO<sub>2</sub> emission of 32 kg CO<sub>2</sub>-e/h. Star screen and belt conveyor were



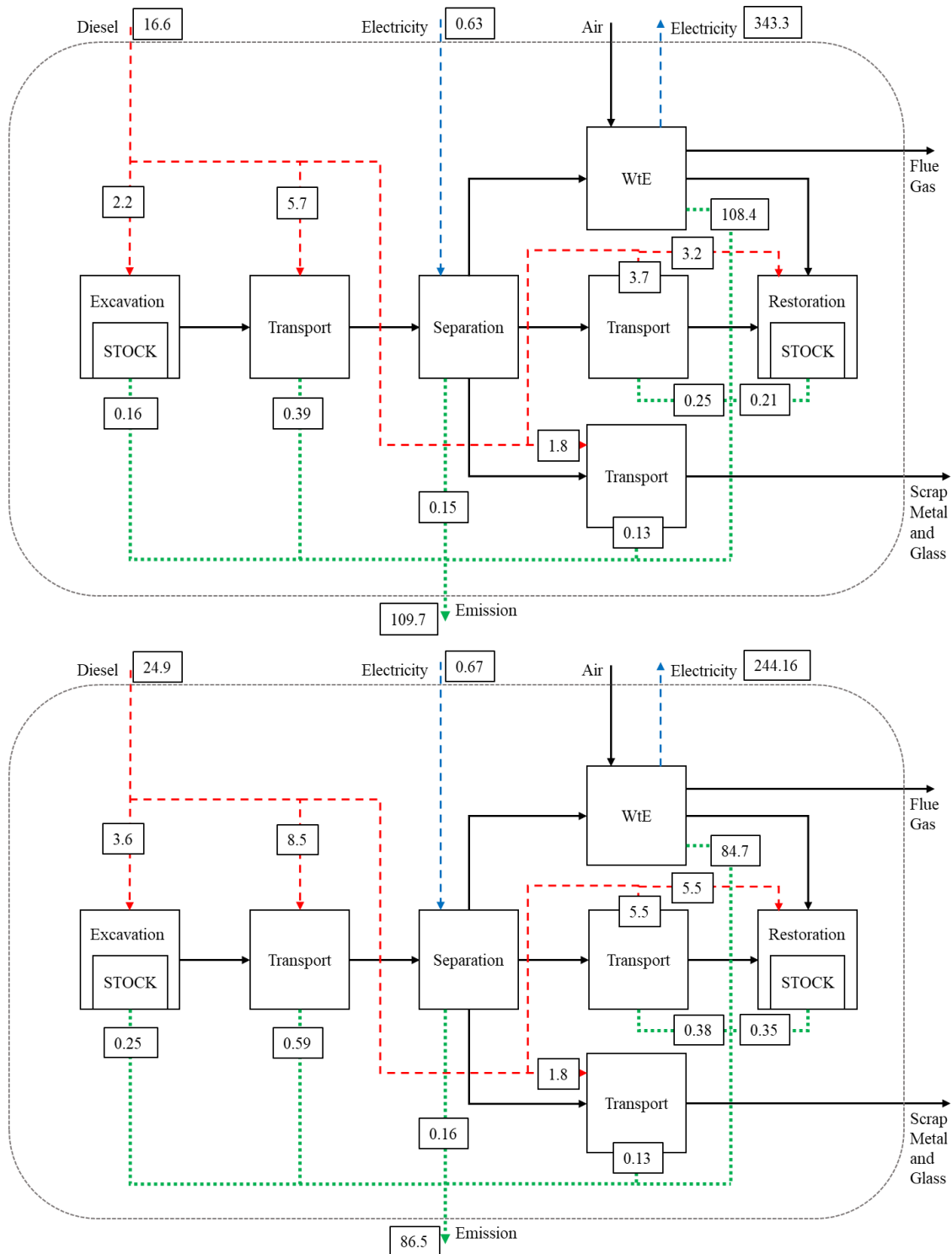
**Figure 3.7 Energy Consumption and Emission of landfill mining utilities and equipment for Jatibarang (top) and Piyungan (bottom) landfills case**

equipped with 25 kW electric motor having electricity consumption of 0.09 GJ/h and CO<sub>2</sub> emission of 22 kg CO<sub>2</sub>-e/h. In general, transportation dump truck consumes the most energy and emit CO<sub>2</sub> highest for both landfill utility cases. The overview of energy consumption and CO<sub>2</sub> emission for project utilities is presented in Fig 5.

For the Jatibarang case, the daily import was 16.6 GJ of diesel and 0.63 GJ of electricity, and the daily production was 343.3 GJ of electricity and 109.7 Mg of CO<sub>2</sub>-eq (Figure 3.6, top). For each ton of EW, 74.8 MJ diesel and 2.8 MJ electricity are utilized while producing 1548.3 MJ of electricity and 494.7 kg CO<sub>2</sub>-e. Most of the diesel was consumed for transportation activity (11.2 GJ/d), and transportation from excavation sites to separation facility is the highest (5.7 GJ/d). The separation facility is located in a landfill site complex with a distance of about 1 km. It required two dump truck units with a total round trip of 46 times for 6h working time. Transportation of SL material to restoration site required 3.7 GJ/d with a total round trip of 11 times, while transportation of NC to off-site processing facility required 1.8 GJ/d for one round trip. The emission generation of transportation to separation facility, restoration site, and off-site processing facility was 0.39, 0.25, and 0.13 t of CO<sub>2</sub>-e/d. In general, the system has a positive energy balance with 326 GJ daily surplus from the WtE process. WtE unit also emits 108.4 of CO<sub>2</sub>-e/d.

Comparable to the Jatibarang case, in the Piyungan case, 24.9 GJ of diesel and 0.67 GJ of electricity were imported, and 244.16 GJ of electricity and 86.5 t of CO<sub>2</sub>-e were produced every day (Figure 3.6, bottom). It means that to process 1 Mg of EW, it consumed 69.7 MJ diesel and 1.9 MJ electricity while emitting 242.2 kg of CO<sub>2</sub>-e. Electricity generated from the WtE process is up to 683.7 MJ per ton of waste, almost half of the Jatibarang case. Similar to the Jatibarang case, transportation consumes most of the energy import in the form of diesel (15.8 GJ). Fuel for transporting EW and SL in the Piyungan case (8.5 GJ and 5.5 GJ) is higher than the Jatibarang case because the amount of round trip is higher by 74 and 16 for EW and SL. Diesel needed to transport NC to the off-site processing facility is the same between both cases since the facility located in the industrial complex located within 2h driving. The emission generation of transportation to the separation facility, restoration site, and the off-site processing facility were 0.59, 0.38, and 0.13 Mg of CO<sub>2</sub>-e/d, respectively. The WtE unit generates less

energy compared to the Jatibarang case but produces lower emission (244.16 GJ and 84.7 Mg of CO<sub>2</sub>-e).



**Figure 3.8 Energy and emission flow of Jatibarang (top) and Piyungan (bottom) case. Diesel and Electricity in GJ/day. Emission in Mg CO<sub>2</sub>-eq/day.**

A similar project was done in Germany and consumed 103 MJ diesel and 1.9 MJ of electricity. It also produces 12 kg of CO<sub>2</sub>-e without considering the energy recovery option (Hölzle 2019). Landfill mining

study was done by Laner et al. (2016) counting the thermal recovery process calculates the emission, producing up to 640 kg of CO<sub>2</sub>-e/Mg but saved up to 1550 kg of CO<sub>2</sub>-e/Mg when including the system in the regional energy mix (Laner et al. 2016). Energy and emission of the landfill mining project will be depending on the recoverable material characteristic, landfill site properties, and supporting infrastructure availability. Jatibarang and Piyungan landfill sites have similar properties but very distinct waste characteristics. Both landfills located near the city within two hours, driving from the industrial complex. The differences lie in the socio-culture of the regions.

### 5.3. Impact of Landfill Mining towards current waste management

Finding an appropriate site to keep municipal waste safely has become harder over time. The landfill mining project will influence the current waste management by providing an additional lifespan in the form of a final sink. The final sink is designed to only keep inert waste, such as fly ash and bottom ash. The capacity of the final sink could be calculated from the potential of recovered space after excavation and the amount of ash and SL that will be delivered to the landfill (Yi 2019). Assuming that the project will excavate 100% of waste, the potential recovered capacities of Jatibarang and Piyungan landfills are 1,245,000 m<sup>3</sup> and 709,250 m<sup>3</sup>, respectively (Mawarni 2016; Purnama Putra et al. 2018). The landfill mining project can add the lifespan of the landfill site by 15 years for the Jatibarang landfill and 9 years for the Piyungan landfill until the project finished. The lifespan could be further extended until it reaches the original design capacity. The new landfill sites will be converted from single used landfill into multiple zones landfill to keep the material cycle inside the landfill. There are four zones consisting of reception and stabilization, excavation, energy and material recovery, and final sink zones.

Indonesia's government has pushed the development of the waste-to-energy facilities after the sign of Presidential Decree No. 35/2018. The decree mentioned the feed-in tariff for the WtE power plant in Indonesia is 13.35 cents USD/kWh and an additional subsidy for waste processing for about 35.5 USD/ton. With the assumption that the MFA scenario was done very well, landfill mining projects in the Jatibarang landfill and Piyungan landfill is potentially providing a positive cash flow of 5,249,970 USD and 5,598,517 USD respectively.

## **6. Conclusion**

Assessment of landfill mining as an alternative solution to solve waste management problems in Indonesia is presented in this paper. Indonesian government encourages its municipality to build the WtE power plant not only to reduce the burden of the landfill but also to contribute to the country's energy mix. To maintain the sustainability of the power plant, a constant supply of feedstock is needed. Accumulated waste in the landfill has potential as a feedstock for the WtE power plant. By excavating the landfill and recovering the material, the environmental burden in the surrounding landfill area can be reduced. This study demonstrates that with proper landfill mining operations, the lifespans of the existing landfill could be extended.

There are potentially 45% of CW from the Jatibarang landfill and 28 % of CW from the Piyungan landfill to be recovered. The potentials of inert recyclables such as metal and glass are 3% from the Jatibarang landfill and 2% from the Piyungan landfill. They could be recycled after removing the contamination.

Analyzing the material flow in Jatibarang and Piyungan landfills helps to identify the required material, energy yield, and emission-related to the process (i.e., excavation, transportation, separation, energy recovery, and restoration). The integration of MFA and process modeling produces reliable input and output data for mass balance. The excavated waste shows similarities in terms of composition and characteristics. The most significant emission was coming from a WtE plant with the amount of HCl in the flue gas exceeding the required standard by the government. The energy balance was considered profitable, with an efficiency of 20% for the Piyungan case and 14% for the Jatibarang case.

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## **Chapter 4**

# **Energy Recovery from Hydrothermally treated Excavated Waste**

### **1. Introduction**

The rapid growth of the economy and development in Indonesia lead to an increase in waste generation. The landfill is the most used method for waste disposal in Indonesia. More than 60% of 65 million tons of waste was dumped on the landfill, and 57% of it going to open dumping landfills. 19.62% of waste was unmanageable, and the rest was managed by the informal sector (Directorate of Waste Management 2017).

There are resources that are piled up inside the landfill, and landfill mining is developed and conducted to recover these materials. They can be utilized either to be recycled or as an energy source via thermal conversion (Hogland et al. 2010). Several landfill mining research, which has been done recently either in developed or developing countries, shows positive energy balance results and proved to bring environment benefit (Lokahita et al. 2017a; Hogland et al. 2018; Lokahita et al. 2018).

In the landfill, an organic compound such as food waste or wood reacts faster than others in the anaerobic environment because they consist of cellulose, which is readily fermented to form simpler organic carbon chain material with a higher calorific value which is suitable for energy recovery. Since plastic degradation will take hundreds of years, it will remain as it is, but contaminated with dirt. Contaminated plastic cannot be reused and very costly to recycle (Rees, 1980). The excavated material needs a pre-treatment before appropriately used (Lokahita et al. 2019).

The hydrothermal treatment has proved to increase the quality of waste and biomass for solid fuel utilization (Darmawan et al. 2017; Lokahita et al. 2017b). The hydrothermal treatment uses hot compressed steam at low temperature for thermochemical conversion and promotes dehydration and decarboxylation efficiently (Novianti et al. 2016).

In this study, we proposed the utilization of hydrothermal treatment to increase the quality of combustibles extracted from landfills. We use the material from the Jatibarang Landfill, which located on the southwest side of Semarang City, Central Java Province, Indonesia. The experimental design was two-level factorials with three replication points in the middle. We use this design as a screening purpose for the factors affecting the hydrothermal treatment. Holding temperature, solid load, and holding time were varied according to the design. Volatile matter, fixed carbon, and ash content were observed. We also noted the possibility of washing effect on the lower solid load experiment condition. Analysis of variance was used to determine the coefficient estimate of each factor.

## 2. Material and Method

A mid-size excavator was used to obtaining samples from nine different points in Jatibarang Landfill. The excavated waste (EW) was sorted by hand into combustible, incombustible, and soil. The combustible fraction was stored in a plastic vacuum bag then stored in the freezer before further analysis and treatment. The characteristic of EW is presented in table 4.1.

**Table 4. 1 EW Characteristic by proximate and ultimate analysis**

Parameter	Value	
Moisture	32.20	wt%
Fixed carbon	7.58	wt% dry
Volatile	61.01	wt% dry
Ash	31.41	wt% dry
Carbon	37.03	wt% dry
Hydrogen	5.16	wt% dry
Nitrogen	1.30	wt% dry
Chlorine	0.86	wt% dry
Sulfur	0.67	wt% dry
Oxygen	23.57	wt% dry

The combustible fraction of excavated wastes was hydrothermally treated using a lab-scale reactor, series MMJ-500, made by OM Lab-tech Co., Ltd., Japan. The reactor vessel was a glass tube with a volume capacity of 500 ml. A stirrer with an electric motor provided a centrifugation effect during the holding time. The 0.95kW electric heater was employed to raise the temperature. The temperature was controlled using a proportional–integral–derivative (PID) controller. The reactor was also equipped with a pressure gauge to monitor pressure changes during the reaction.

Five grams of samples and 5-45 gr of Mili-Q ultra-pure water were mixed in the reactor tube. The reactor was sealed, purged with argon, then heated to 200-240 °C. The holding time was set from 0 to 60 minutes. After the reaction finished and the reactor cooled down to 60 °C, the slurry was discharged from the reactor to be dried at 70 °C overnight. For experiment condition in lower solid load, vacuum filtration using Whatman filter paper were performed before drying. The dried sample was stored in a tightly sealed bag before further analysis.

**Table 4. 2 Individual Experiments**

Experiment	Holding Temperature (°C)	Solid load (-)	Holding Time (h)
1	200	0.1	0
2	240	0.1	0
3	200	0.5	0
4	240	0.5	0
5	200	0.1	1
6	240	0.1	1
7	200	0.5	1
8	240	0.5	1
9	220	0.3	0.5
10	220	0.3	0.5
11	220	0.3	0.5



Proximate analysis (VC, FC, and ash) was measured according to ISO standard. The sample was heated to 900 °C for 7 minutes, and the mass loss was measured as VM. For ash determination, the sample was heated until 815 °C and maintained this temperature for 60 minutes. The ultimate analysis was performed using Vario Micro Cube Elemental Analyzer. The carbon, hydrogen, and nitrogen ratio were used to build Van Krevelen Diagram. The morphological structure of the hydrochar particle was observed using Scanning Electron Microscope KEYENCE VE-8800.

A total of 11 individual experiments was done, and three variables, volatile matter (VM), fixed carbon (FC), and ash, were observed. The two factorial screening experimental design was used to understand the influence of temperature, holding time, and solid load on the product characteristic.

### 3. Data Interpretation

The statistical significance of a model can be tested by comparing the variation explained by the model and the variation of model residuals through the analysis of variance (ANOVA). The idea of ANOVA is to compare the variable changes using a statistical test to prove the significance of the model and error.

$$\sum_{i=1}^a \sum_{j=1}^n (y_{ij} - \bar{y}_{..})^2 = n \sum_{i=1}^a (\bar{y}_{i.} - \bar{y}_{..})^2 + \sum_{i=1}^a \sum_{j=1}^n (y_{ij} - \bar{y}_{i.})^2 \quad (2)$$

Equation 2.1 is the fundamentals of the ANOVA test.  $y_i$  represents the total of the observations under the  $i^{\text{th}}$  treatment.  $\bar{y}_{i.}$  represent the average of the observations under the  $i^{\text{th}}$  treatment.  $\bar{y}_{..}$  represent the total of all the observations and  $\bar{y}_{..}$  represent the grand average of all the observations.

It states that the total variability in the data, as measured by the total corrected sum of squares, can be partitioned into a sum of squares of the differences between the treatment averages and the grand average plus a sum of squares of the differences of observations within treatments from the treatment average. The difference between the observed treatment averages and the grand average is a measure of the differences between treatment means, whereas the differences of observations within a treatment from the treatment average can be due to only random error. Thus, we may write Equation 2.1 symbolically as;

$$SS_{Tot} = SS_{Mod} + SS_{Res} \quad (3)$$

$SS_{Mod}$  is the sum of the square of the model, describing the interaction of factors and response.  $SS_{Res}$  is the sum of the square due to error within treatments. The numbers of degrees of freedom (d.f.) divide the sum of the square for each source of variation (total, regression, residual, lack of fit, and pure error). Thus, the media of the square was obtained by dividing the sum of the square with degrees of freedom. Table 4.1 shows the formula for all variation sources [13].

**Table 4. 3 Analysis of variance for the fitted mathematical model to ab experimental data set using multiple regression**

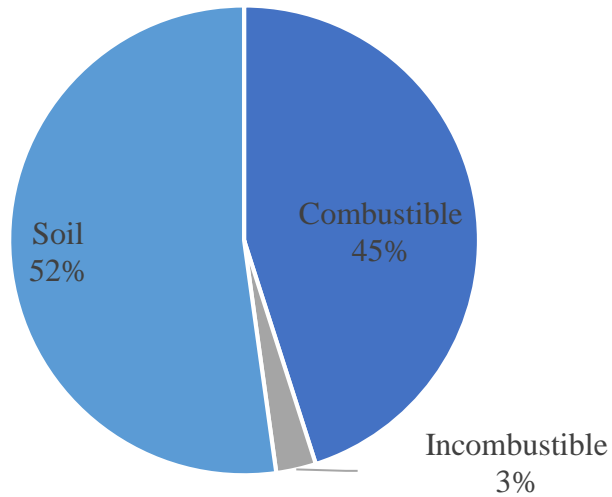
Variation Source	Sum of the Square	d.f.	Media of the square
Regression	$SS_{reg} = \sum_i^m \sum_j^{n_1} (\hat{y}_i - \bar{y})^2$	$p - 1$	$MS_{reg} = \frac{SS_{reg}}{p - 1}$
Residual	$SS_{res} = \sum_i^m \sum_j^{n_1} (y_{ij} - \hat{y}_i)^2$	$n - p$	$MS_{res} = \frac{SS_{res}}{n - p}$
Lack of fit	$SS_{lof} = \sum_i^m \sum_j^{n_1} (\hat{y}_i - \bar{y}_i)^2$	$m - p$	$MS_{lof} = \frac{SS_{lof}}{m - p}$
Pure error	$SS_{pe} = \sum_i^m \sum_j^{n_1} (y_{ij} - \bar{y}_i)^2$	$n - m$	$MS_{pe} = \frac{SS_{pe}}{n - m}$
Total	$SS_{tot} = \sum_i^m \sum_j^{n_1} (y_{ij} - \bar{y})^2$	$n - 1$	

$n_i$ , number of observations;  $m$ , the total number of levels in the design;  $p$ , number of parameters of the model;  $\hat{y}_i$  estimated value by the model for the level  $i$ ;  $\bar{y}$ , overall media;  $y_{ij}$ , replicates performed in each level;  $\bar{y}_i$ , media of replicates performed in the same set of experimental conditions. The significant results by 95% confidence level ( $p < 0.05$ ) were selected and will be displayed as a response surface curve. Meanwhile, the non-significant result will be only presented the raw data.

#### 4. Results

Soil like material dominates the composition of waste in Jatibarang Landfill. The combustible part made up 45% of the total composition mainly consist of organic and plastic. Incombustible such as metal and glass take 3% part of the composition. More than 50% of domestic waste in Semarang City was organic.

Because of oxidation and fermentation in the landfill, most of the organic wastes were decomposed and left out hard cellulose waste. The average composition of excavated waste is shown in Figure 4.1.



**Figure 4. 1 Waste Composition in Jatibarang Landfill**

**Table 4. 4 Proximate Analysis Results**

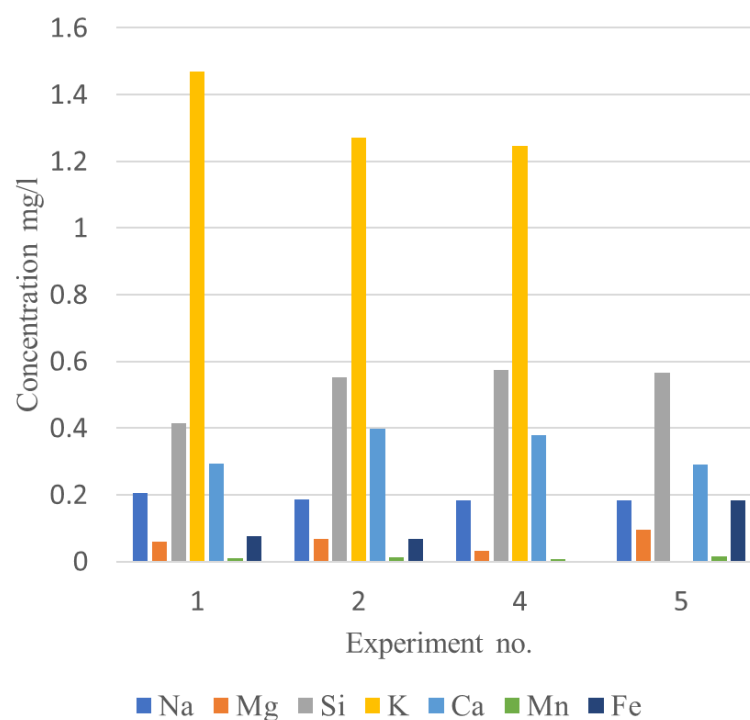
Experiment	Steam Quality (%)	Fixed Carbon (dry wt%)	Volatile (dry wt%)	Ash (dry wt%)
1	7.90	7.3	63.9	28.8
2	16.90	30.7	50.0	19.2
3	78.41	5.1	77.5	17.3
4	100.00	23.6	57.0	19.4
5	7.90	15.4	63.7	20.9
6	16.90	7.1	60.4	30.5
7	78.41	11.4	64.6	24.0
8	100.00	10.6	58.8	29.5
9	49.08	17.9	65.9	16.2
10	49.08	13.5	69.9	16.7
11	49.08	15.1	67.1	17.8

The results from the proximate analysis are shown in table 2 above. The steam quality was calculated using the steam table (IAPWS 95) with various temperature and specific volume of each experiment. Only experiments 4 and 8 achieved 100% steam quality, which means all of the ultra-pure water was converted into steam.

In experiments 1, 2, 5, and 6, we expect some minerals were leached out due to low steam quality. The liquid products were analyzed using ICP-AES while the solid products were burned at 900°C in a muffle furnace, and the ash was analyzed using XRF. The results show that the raw product and solid product consisted mainly of Ca, Si, Mn, Na, Fe, Mg, Cl, and K. Most elements were also found in the liquid product (Figure 4.2).

**Table 4. 5 Ultimate Analysis Result**

Experiment	C (dry wt%)	H (dry wt%)	N (dry wt%)	S (dry wt%)	O (dry wt%)	Cl (dry wt%)	HHV (MJ/kg)
1	50.1	7.8	1.0	0.1	12.5	0.0	25.28
2	41.5	6.3	0.6	0.1	32.3	0.0	22.90
3	54.4	8.8	0.9	0.1	18.1	0.4	27.12
4	45.4	6.6	1.3	0.1	26.7	0.6	27.15
5	54.6	8.4	0.7	0.1	13.6	1.7	21.13
6	47.9	6.9	0.8	0.0	11.8	0.1	20.46
7	44.9	6.7	1.3	0.0	22.4	0.7	24.73
8	45.3	6.5	1.3	0.1	15.6	0.7	18.10
9	45.9	6.6	1.5	0.1	29.0	0.8	20.74
10	57.0	9.0	1.1	0.0	15.5	0.7	28.48
11	52.7	7.9	1.2	0.1	19.8	0.6	20.47



**Figure 4. 2 Leached minerals in the liquid product**

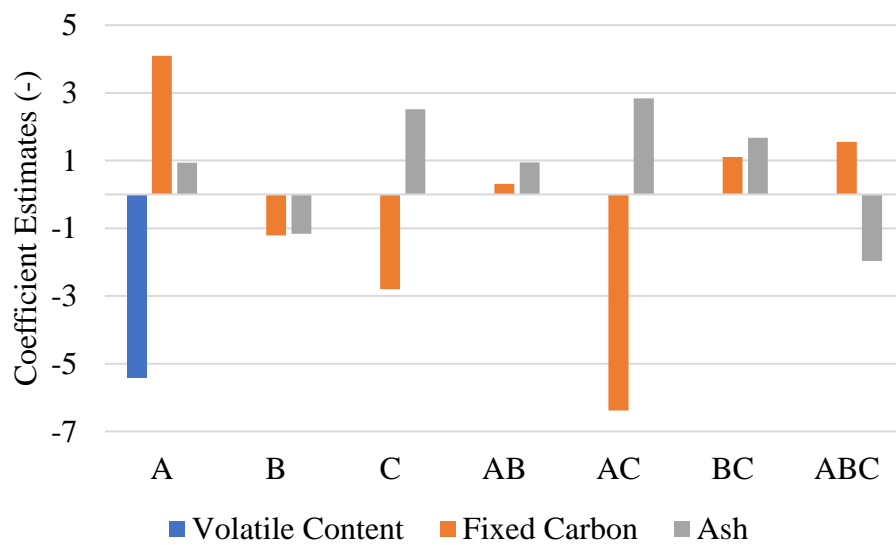
## 5. Discussion

ANOVA analysis is used to determine the model vector to estimate the statistical significance of model coefficients. Coefficients that are found statistically insignificant should be removed from the model unless they are included in significant interactions or higher order coefficients. The factors were coded as follows; A for temperature, B for solid load, C for holding time, AB for temperature-solid load interaction, AC for temperature-holding time interaction, BC for solid load-holding time interaction, and ABC for temperature-solid load-holding time interaction. The initial model included all coefficients for the full quadratic model. The model with full quadratic significance only found for chlorine content, fixed carbon content, and ash content. P-values less than 0.0500 indicate model terms are significant. Values higher than 0.0500 indicate the model terms are not significant. Conversely, for the lack of fit, P-values higher than 0.0500 is wanted to make sure that the model is fit. The other model will be further screened out to remove the insignificant factors.

The model for volatile content became significant after leaving interaction terms A with a model p-value of 0.0217 and a Lack of Fit P-value of 0.1. The model for Sulfur content became significant after

leaving interaction terms C and AB with a model p-value of 0.01 and a Lack of Fit P-value of 0.99. The model for Hydrogen content became significant after leaving interaction terms A and BC with a model p-value of 0.045 and a Lack of Fit P-value of 0.926. Model for Nitrogen content became significant after leaving interaction terms B with a model p-value of 0.029 and Lack of Fit P-value of 0.57. The model of carbon and HHV is still insignificant even after model reduction. The complete ANOVA table for is presented in Appendix B.

The coefficient estimate represents the expected change in response per unit change in factor value when all remaining factors are held constant. The temperature has a negative impact on VM but positive on FC. The solid load has a negative effect on the ash content. On the other hand, increasing holding time could heavily increase the ash content and reduce fixed carbon. Coefficient estimates of significant terms of proximate analysis are shown in Figure 4.3 below.



**Figure 4. 3 Coefficient Estimates of Significant Terms in Proximate Analysis Model**

Interaction terms AB, AC, and BC show a positive effect on ash content. Interaction AB and BC have a slight effect of increasing the fixed carbon, but interaction AC significantly reduces the fixed carbon. On the other hand, ABC interaction terms could reduce the ash content while increasing the fixed carbon. Interaction AB involves temperature and solid load behavior during hydrothermal process. In high temperature, more volatile matter will be converted to fixed carbon. High solid load also promotes the

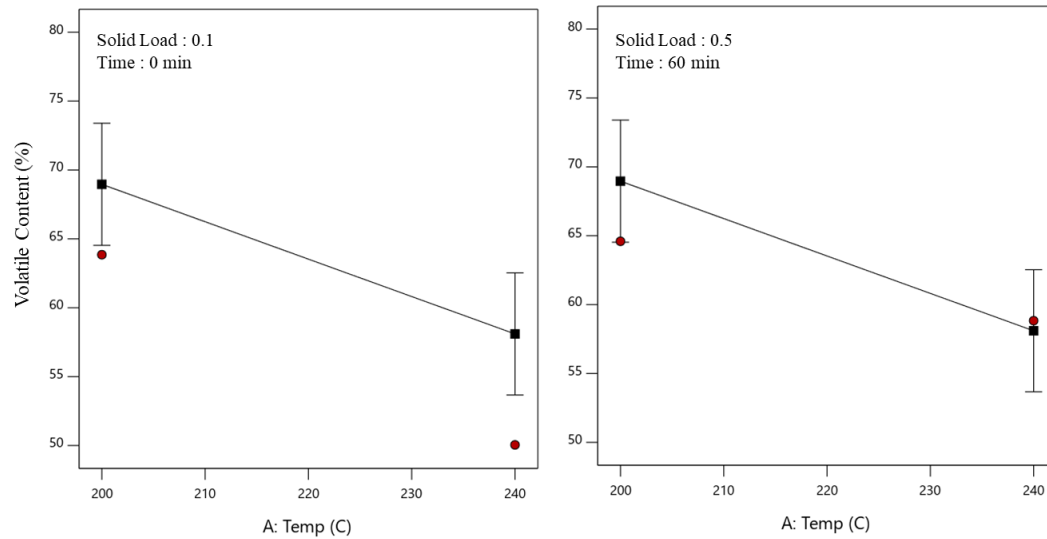
fixed carbon formation because there are more solid to react with steam in the reactor. Some fixed carbon will also be converted to ash from the reaction with steam. On the other hand, AC interaction which involves temperature and holding time could reduce the fixed carbon content. In high temperature, the longer reaction held, the more fixed carbon is converted to ash. In correspond to that, ash content will also increase. BC interaction which involves solid load and holding time behaves similar to AB interaction. Higher solid load and holding time will have high amount of mass react with steam in a longer time which promote the conversion of volatile matter to fixed carbon and fixed carbon to ash. When the micropores start to forming in the material surface, and the holding time is still going, there are chance of minerals that have been leach out to be reabsorbed. Diffusion of minerals to the char will increase the ash content. As the coefficient estimates of ash in AB and BC interaction is higher, there are possibility of higher conversion rate from fixed carbon to ash rather than the formation of fixed carbon from volatile matter.

ABC interaction will create a curvature in which the formation of fixed carbon is increase as temperature, solid load and holding time, but at some point it will going down. Similar to that, ash content will decrease in parallel to the operating condition, but it will go down at some point.

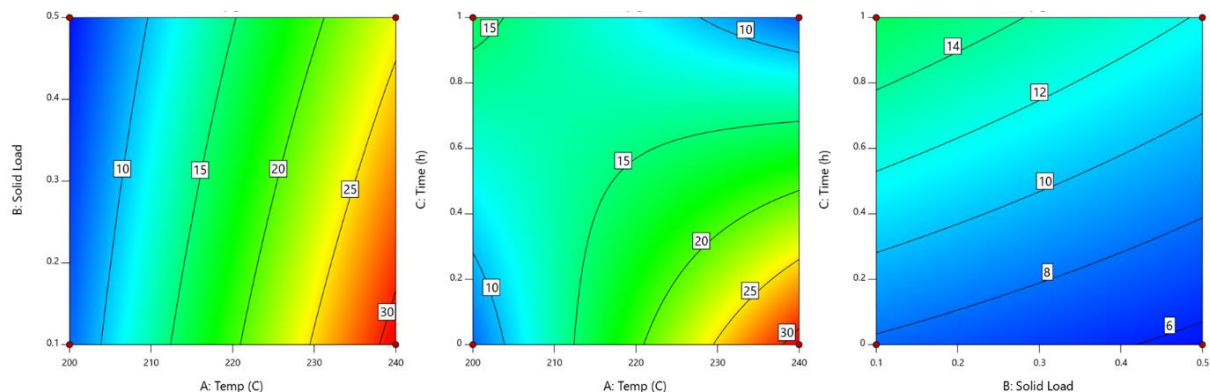
During the hydrothermal process, VM will be converted to FC through decarboxylation. The higher the temperature, the more VM will be converted. Hydrocracking also happened and increasing the surface area by increasing the porosity. When the sample was held for a long time, more pores formed but, the minerals leached out during the process could be re-absorbed thus, increasing the ash content. In general, the temperature was the dominant parameter for altering the characteristic of EW during the hydrothermal process.

The VM is decreasing as a function of temperature and time due to the decomposition of organic components during the reaction (Figure 4.4). Decomposition of biomass from decarboxylation and dehydration produces a simple carbon chain, organic acid, and releases the minerals in the form of ash (Mäkelä and Yoshikawa 2016). Even though the VM is decreasing, it is still a major constituent in the product, with a value ranging from 50%-77%. High VM means the product is easy to ignite. VM, this high is coming from the plastic parts in the EW. The plastic has not breakdown so much during

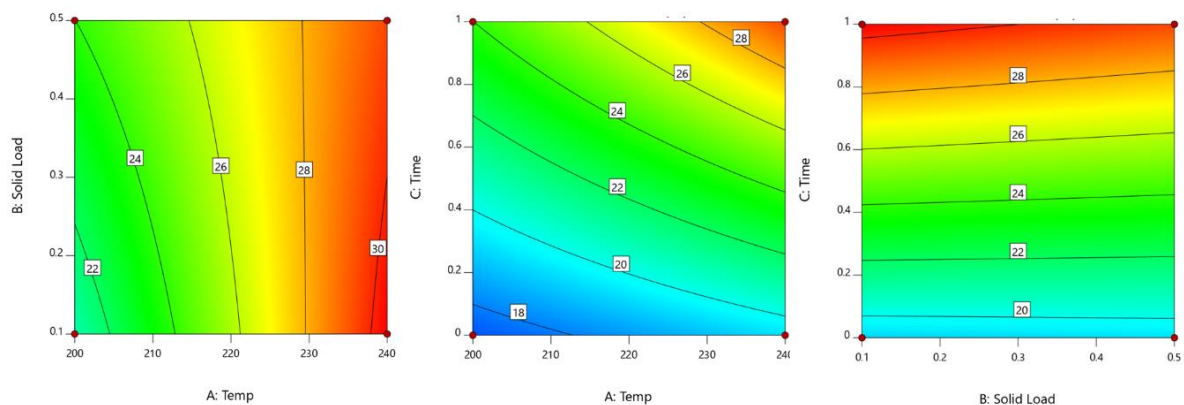
hydrothermal treatment. The plastic deformed into a smaller build and increase its plasticity. FC is increasing as the function of temperature and solid load (Figure 4.5). As the most crucial aspect of calorific value it is important to pay attention to this alteration.



**Figure 4. 4 One factor graph of volatile content**



**Figure 4. 5 Response surface graph of fixed carbon**

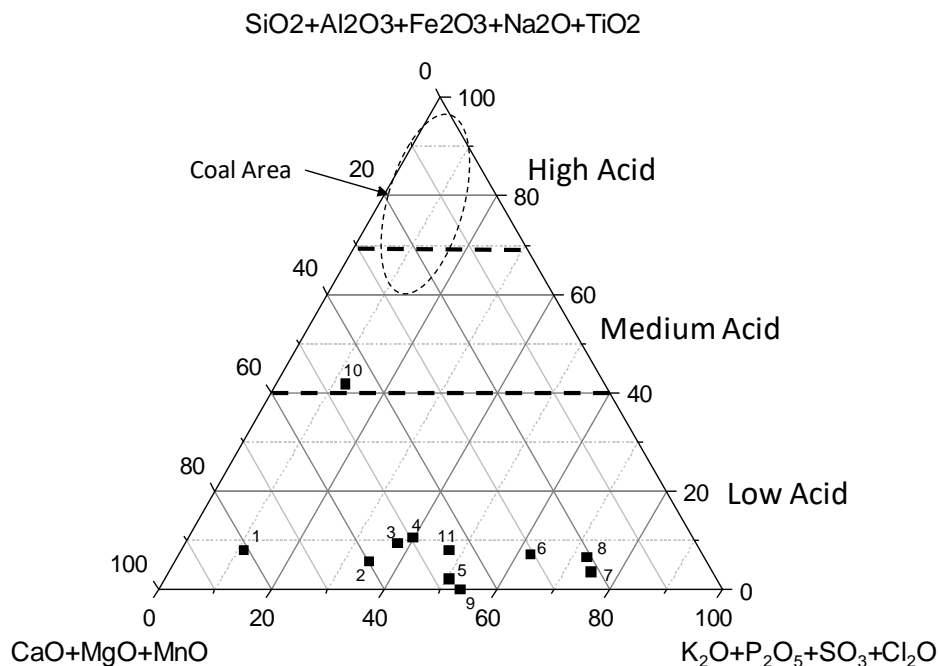


**Figure 4. 6 Response surface graph of ash**



Ash content mainly affected by solid load and holding time. The higher the solid load less ash content material was obtained. On the contrary, holding temperature and reaction time increase the ash content significantly. Similar findings were found where kimchi and paper waste was used. Temperature from 180 °C to 220 °C was applied and the sample ash content increases (Yoshikawa and Prawisudha 2014). This phenomenon caused when the surface of the material starts to breakdown, forming a higher surface area and adsorbing minerals in the hydrothermal liquid.

Higher ash content is not suitable for waste to energy since it promotes slagging and fouling. Numerical optimization method was done using Design Expert <sup>TM</sup> v11 to find the best solution from our model by selecting the desired goals for each factor and response. Numerical optimization process involves combining the goals into an overall desirability function. A desirability closer to 1.00 means the goals were easier to reach and better results may be available. The numerical optimization feature in the design expert package finds one point or more in the factors domain that would maximize this objective function. Parameter with full significant terms, fixed carbon and were chosen as a basis. The optimum criteria were to maximized the fixed carbon and minimized the ash. Thus, the recommended optimum operating condition for high FC and lower ash is 240 °C, 0.1 solid loads and 0 hour holding time resulting on 31.17% fixed carbon and 19.21% ash content with 0.88 desirability.

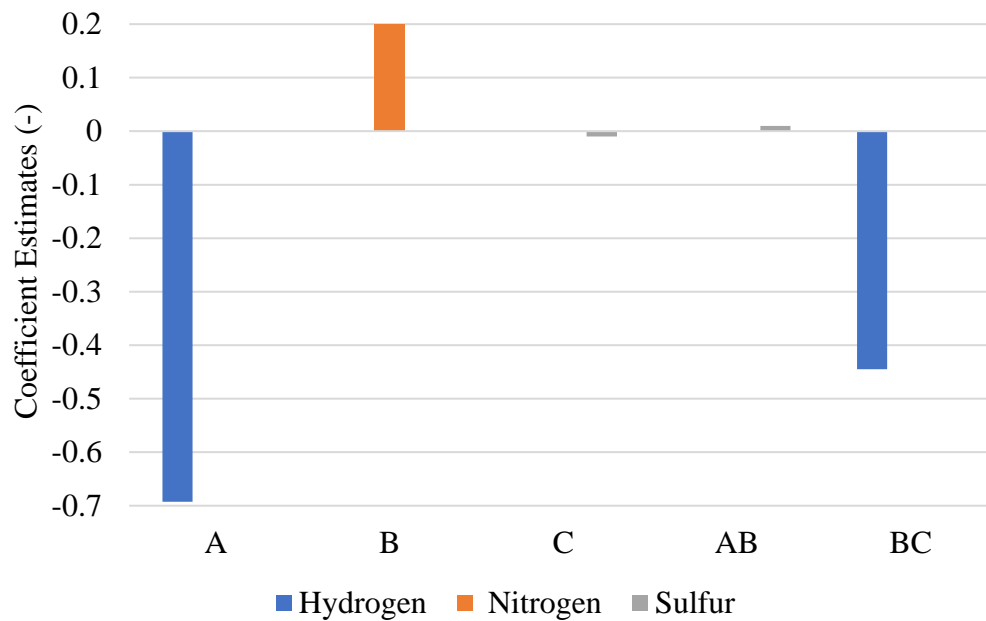


**Figure 4. 7** Ash composition of char samples produced from different feedstock based on the classification system of Vassilev et al

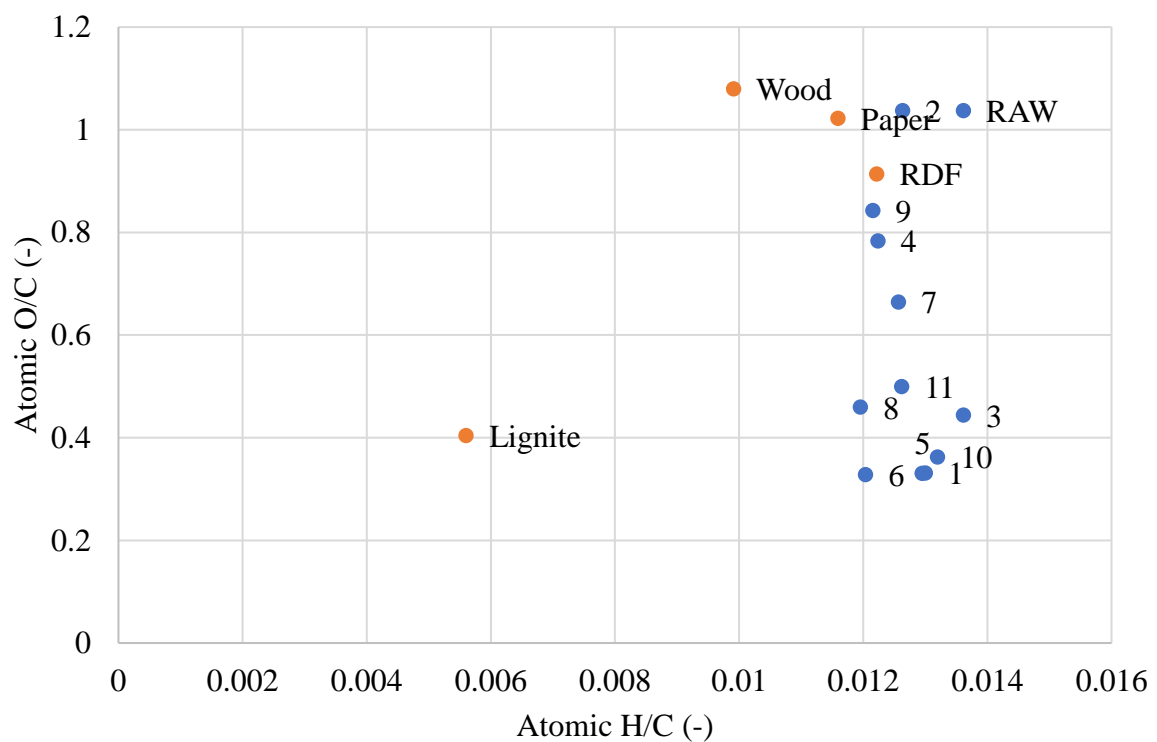
Hydrochar combustion behavior was affecting the ash composition. To estimate the potential ash behavior, various parameters based on the ratio of acidic and basic were used. These parameters have been implemented for biomass (Jenkins et al. 1998). It is known that an increase of Ca, Fe, Mg, Na, and K minerals gives lower ash melting temperatures and can cause slagging and fouling during coal combustion. Vassilev et al. (Vassilev et al. 2013) found that biomass ash mixture behavior has more variable than ash from coal combustion. Ca, Al, and Ti increased ash deformation temperatures and mainly medium to high contents of K, and Si governed the formation of mixtures in low temperature. Vassilev et al. (Vassilev et al. 2014) also proposed a new classification system to enhance preliminary predictions on ash behavior for solid biomass fuels. Ash behaviors on the hydrochar based on this classification are illustrated in Figure 4.7. The effect of being in the low acid region is that the minerals tend to diffuse to the sample which makes it hard for the leaching effect of hydrothermal treatment to develop.

For ultimate analysis, the model for carbon and hydrogen was not significant enough to be built. The hydrogen model was built based on A and BC terms. The nitrogen model was built based on the B term. The sulfur model was built based on C and AB terms. The coefficient estimates for hydrogen, nitrogen and sulfur is available in figure 4.8. The hydrothermal treatment is mimicking the process of coalification in nature by utilizing the temperature and the holding time. Hydrogen content was expected to decrease, correspond to dehydration and decarboxylation reaction in the hydrothermal reaction. Nitrogen and Sulfur behave differently according to the original species but, the sulfur is easily soluble in the water and could also get carried by residual steam in the form of  $H_2S$ . As the nitrogen content was still high even after HTT, there is the possibility of  $NO_x$  emission reduction during the combustion process in the WTE facility.

The effect of the hydrothermal treatment on the atomic composition could be understood by plotting the atomic H/C and O/C ratios of biomass, and the corresponding hydrochar were plotted in a Van Krevelen diagram. As mentioned before, the hydrothermal treatment altered the chemical characteristic of the hydrochar. Figure 4.9 shows the Van Krevelen diagram to compare produced hydrochar with conventional fuels.



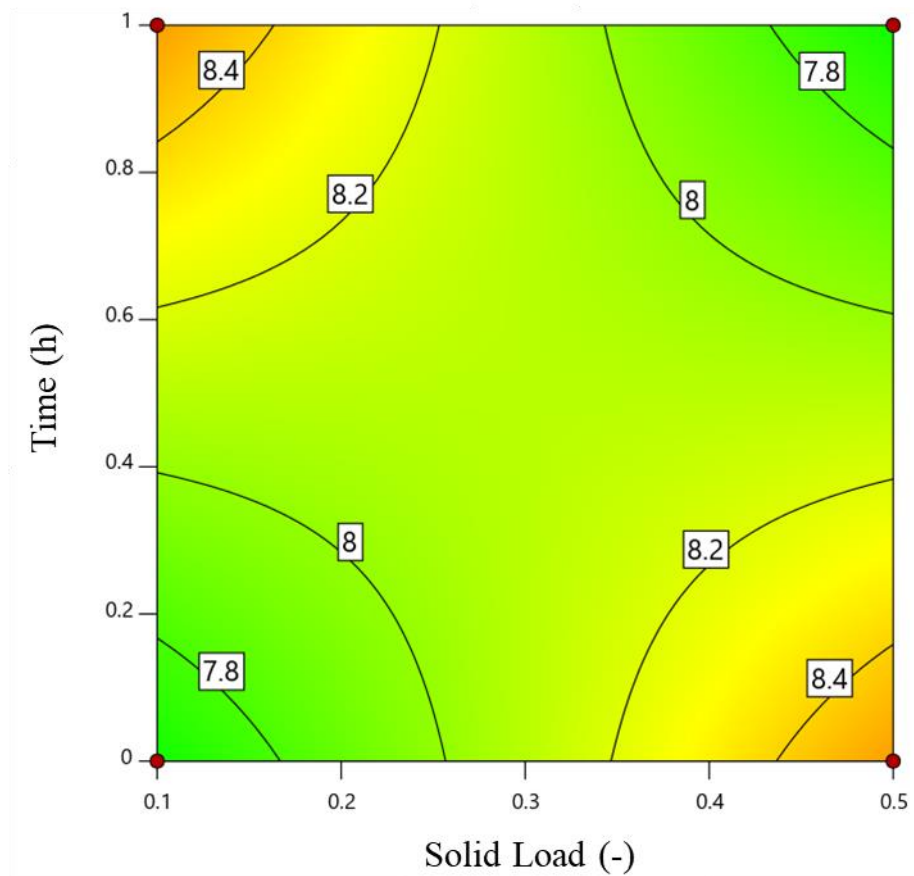
**Figure 4. 8 Coefficient Estimates of hydrogen, nitrogen, and sulfur model**



**Figure 4. 9 Van Krevelen Diagram of hydrothermally Treated Excavated Waste and other biomass for comparison**

The best atomic ratio was found for the result of experiment no. 9. The properties of the material have a similarity with commercial RDF. Experiment 1, 3, 5, 6, 8, 10, and 11 shows similar O/C reduction, which is similar to lignite but not in the H/C ratio. It means that the dehydration process has failed to occur during reaction time. The response surface for hydrogen content is shown in figure 4.10.

Experiment 2 has a similar composition with paper and wood. On that point, the cellulose component in the samples already started to decompose, but the amount is still low, resulting in low carbon yields. The trend in the diagram also shows that dehydration is the primary reaction during the process as oxygen content decreased in proportion to the hydrogen content. In summary, the treated excavated waste still need to be upgraded before being utilized as solid fuel. It is also possible to find alternative application other than solid fuel. Decarboxylation reaction could be promoted more by adding catalyst or co-treated with other biomass. Decarboxylation will produce higher fixed carbon bring it closer to the coal properties.



**Figure 4. 10 Response surface of hydrogen content**

## 6. Conclusions and Recommendation

We investigate the possible utilization of hydrothermal treatment to increase the quality of excavated waste for an energy recovery application. One landfill in Indonesia, Jatibarang Landfill, has been chosen as case studies to be excavated and treated in a hydrothermal reactor.

The experiment was using two factorial screening experimental design. The primary purpose is to observe the most significant factor which alters the VM, FC, and ash content of the EW. The hydrothermal treatment has successfully decomposed the cellulose and plastic in the EW into smaller carbon chain, as shown in the decrease of VM as we increase the experiment parameter. Correspond to this behavior, the ash content has decreased significantly.

The highest HHV and carbon content is 28.8 MJ/kg and 57 wt%, respectively. It was found in the operating condition of 220 °C, 0.3 solid loads, and 30 min holding time, which also has similar characteristics with RDF. The lowest ash content was obtained in the operating condition of 200°C, 0.5 solid load and 0 min holding time for 17.3%. To maximize the fixed carbon and minimize ash yield, operating condition in 240 °C, 0.1 solid loads, and 0 h holding time was suggested.

Van Krevelen diagram shows that the process are lacking decarboxylation reaction which make it hard to achieve coal-like material properties. Although in terms of HHV and C:H:O ratio it already sufficient for energy recovery by incineration, high ash content is still not meet with the RDF standard. Ash content for RDF should be lower than 16%. High ash content is the leading cause of slagging and fouling in the reactor. It also promotes de novo synthesis of dioxin.

The hydrothermal treatment has shown good potential to be applied as an intermediary treatment for landfill mining and reclamation scheme. The product could be used for refused derived fuel or enhancing the soil quality. During hydrothermal process, the organochlorine was converted into inorganic chlorine in the lower temperatures compared to the conventional dichlorination process. The water-soluble inorganic chlorine content in the product can be easily removed by the water-leaching and dewatering process (Zaini et al. 2017). It is suggested that a combination of the hydrothermal process and multiple water-washing processes could further reduce the chlorine content of hydrothermally treated product from MSW.

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## **Chapter 5**

# **Hydrochar utilization as an adsorbent for landfill leachate treatment**

### **1. Introduction**

Organic contaminant is problematic parameters in landfill leachate treatment. Common leachate treatment facilities are utilizing biological treatment technology to remove organic matter in the early stages when the BOD/COD ratio of the leachate is high (Berrueta and Castrillón 1992). It takes days to reduce the COD, and the ratio decreases with the age of the landfill, and the process is less effective with time due to the presence of refractory organic matter (Alkalay et al. 1998; Timur and Ozturk 1999; Rodriguez Iglesias et al. 2000). Young landfill leachates are usually treated more quickly as compared to the old ones. As a landfill stabilizes over time, the biodegradable organic content of the leachate tends to decrease, and consequently, the effectiveness of the biological process decreases, and physio-chemical processes may become one of the appropriate options. The typical features of stabilized leachate are high strengths of ammoniacal nitrogen (3000–5000 mg/L) and moderately high strengths of COD (5000–20,000 mg/L), as well as a low ratio of BOD/COD (less than 0.1) (Kargi and Pamukoglu 2004).

One of the physio-chemical processes is adsorption using either activated carbon or other adsorbents such as zeolite, activated alumina, or low-cost adsorbents such as limestone, rice husk ash, and peat. Activated carbon provides an attachment surface for micro-organisms to regenerate the activated carbon biologically (Jin et al. 2006). Combinations of organic and inorganic pollutants that exist in landfill leachate need adsorbents that can remove a variety of pollutants, including organic and inorganic species. It is well known that activated carbons are the most effective adsorbents for the removal of organic pollutants from the aqueous or gaseous phase. Therefore, this type of adsorbent is widely applied as a commercial adsorbent in the purification of water and air (Aghamohammadi et al. 2007).



However, general activated carbon does not have enough adsorption capacity because it usually possesses a non-polar surface due to manufacturing conditions at high temperatures, which is a disadvantage for some applications because of weak interaction with some polar adsorbates (Leboda 1993). This is the reason that much research has been focused on modifying the activated carbon surfaces or on producing composite adsorbent that can interact with either polar or non-polar adsorbates. On the other hand, the surface of carbon is hydrophobic with pore sizes in the nanometer range or above, which makes it more suitable for the adsorption of organic substances (Halim et al. 2011).

Various biomass has been investigated as alternative raw materials for carbon-based adsorbent, but rarely have tested on waste material, not to mention excavated waste from an old landfill. Excavated waste containing organics and plastics is potentially used as carbon-based adsorbent because, after the hydrothermal carbonization process, the carbon content will significantly increase. This work will fill on the gap on current carbon-based adsorbent development by testing char produced from hydrothermally treated excavated waste from Jatibarang Landfill in Indonesia. The adsorption capabilities will be tested using methylene blue and real leachate samples.

## **2. Materials and Method**

### **Materials**

The excavated waste as adsorbent material was obtained from Jatibarang Landfill, the same material which was used in Chapter 4. The leachate sample used in this study was collected from the leachate pond of the Piyungan Landfill in Yogyakarta. The collected leachate was stored in the plastic containers, transferred to the lab, and keep in the refrigerator below 4°C. The leachate was filtered and analyzed for its characteristics in the Water and Environment Lab. of the Islamic University of Indonesia (Table 5.1). Wako Chemical Company Japan supplied cationic dye methylene blue (MB) (C<sub>16</sub>H<sub>14</sub>N<sub>3</sub>SCl.3H<sub>2</sub>O) Reagent Grade.

### **Hydrothermal Carbonization**

The same excavated waste used in Chapter 4 was subjected to hydrothermal carbonization using a lab-scale reactor, series MMJ-500, made by OM Lab-tech Co., Ltd., Japan. The reactor vessel was a glass tube with a volume capacity of 500 ml. A stirrer with an electric motor provided a centrifugation effect

during the holding time. The 0.95 kW electric heater was employed to raise the temperature. The temperature was controlled using a proportional–integral–derivative (PID) controller. The reactor was also equipped with a pressure gauge to monitor pressure changes during the reaction.

Five grams of samples and 11-45 gr of Mili-Q ultra-pure water were mixed in the reactor tube correspond to the solid load of 0.3 and 0.1. The reactor was sealed, purged with argon, then heated to 200-240 °C. The holding time was set to 30 minutes. After the reaction finished and the reactor cooled down to 60 °C, the slurry was discharged from the reactor to be dried at 70°C overnight. For experiment condition in lower solid load, vacuum filtration using Whatman filter paper were performed before drying. The dried sample was stored in a tightly sealed bag before further analysis. A total of six experiments was carried out. Table 5.2 shows the experimental condition for hydrothermal treatment.

**Table 5. 1 Leachate characteristics from Piyungan Landfill**

Parameter	unit	value
TDS	mg/L	14,803
TSS	mg/L	113
pH		8.77
BOD	mg/L	184
COD	mg/L	5,071
Nitrate	mg/L	22
Ammonia	mg/L	279
Nitrite	mg/L	0.57
H <sub>2</sub> S	mg/L	921
Mn	mg/L	0.13
Cu	mg/L	0.03
Zn	mg/L	0.83
Cr <sup>6+</sup>	mg/L	0.43
Cd	mg/L	0.03
Pb	mg/L	0.09
Detergent	mg/L	2.14
Phenol	mg/L	0.3
Fat	mg/L	27.9

**Table 5. 2 Hydrothermal Carbonization Experimental Conditions**

Temperature	Solid Load	Holding Time	Pressure	Code
200	0.1	30	1.5	HC200-1
220	0.1	30	2.3	HC220-1
240	0.1	30	3.3	HC240-1
200	0.3	30	1.5	HC200-2
220	0.3	30	2.3	HC220-2
240	0.3	30	3.3	HC240-2

#### Hydrochar Activation

Two activation methods, CO<sub>2</sub> and steam activation, were chosen to increase the adsorption performance of hydrochar. The CO<sub>2</sub> activation was conducted at 600 °C for 30 min inside the horizontal furnace with a 300 ml/min CO<sub>2</sub> flow rate. Dried hydrochars were heated with a heating rate of 10 C/min under the N<sub>2</sub> atmosphere. Once it reaches the target temperature, the gas flow was switched to CO<sub>2</sub>, and thus the activation process happened. After 30 min, the sample was cooled down to room temperature under N<sub>2</sub> stream. The obtained char was stored in tight seal bags before further application, and an additional “A” label will be put in the code name. For example, sample code HC220-1A means CO<sub>2</sub> activated char from hydrothermally treated excavated waste at 220 °C and 0.1 solid load.

For steam activation, the hydrochar was put into the horizontal furnace, purge with N<sub>2</sub>, and heated to 800 C with a heating rate of 50 °C/min. After reaching the target temperature, the steam was injected using a capillary pump within a flowrate of 0.5 gr/min. After 15 min, the sample was cooled down to room temperature under N<sub>2</sub> stream. The obtained char was stored in tight seal bags before further application, and an additional “B” label will be put in the code name. For example, sample code HC220-1B means steam activated char from hydrothermally treated excavated waste at 220°C and 0.1 solid loads.

### Adsorption of Methylene Blue

Adsorption test was carried out with batch mode to understand the total MB adsorption capacity after equilibrium. The adsorption behavior of each sample was observed. A total of 18 experiments was carried out by contacting 10 ml of MB with a concentration of 50 mg/l in a glass bottle with a sealed cap. The sample was agitated at 140 rpm under 37 °C for about 12 hours. Final methylene blue concentration was analyzed using Shimadzu UV-mini in wavelength of 663 nm. The adsorption capacity was calculated using equation (1).

### Adsorption of Landfill Leachate

Batch experiments were conducted at ambient temperature by 20 ml contacting landfill leachate with 10 mg of adsorbent inside tightly closed vessels. The vessels were agitated at 140 rpm with contact time variation of 4 hours, 6 hours, and 10 hours. 10 hours of contact time were considered more than enough to judge that the adsorbent has reach equilibrium based on previous research. The adsorption capacity was calculated using equation (1).

$$q_e = \frac{(C_o - C_{eq})V}{M} \quad (1)$$

Equation 1 was used to calculate the adsorption capacity for MB and COD adsorption tests. The equation denotes that  $q_e$  as material adsorption capacity at equilibrium (mg/g),  $C_o$ , and  $C_{eq}$  as the liquid concentration at initial and equilibrium (mg/L),  $V$  is the solution volume (L) and  $M$  as the mass of adsorbent (g).

## 3. Results and Discussion

From the total of 18 experiments, two-way ANOVA test was done to estimates the significance of HTT temperature (200 °C, 220 °C, 240 °C), HTT solid load (0.1, 0.3) and activation method (no activation, steam activation, CO<sub>2</sub> activation) with 95% confidence level ( $p < 0.05$ ). Only activation method found to be significant enough with  $p$  value of 0.01 in adsorption capacity test and 0.003 in MB removal test. Full ANOVA table is presented in Appendix C.

Methylene blue adsorption capacity and removal performance from 18 experiment is presented in figure 5.1 and 5.2. Without any activation treatment, hydrochar was able to adsorb methylene blue to some

extent. It was decreasing when the temperature reaction and reactor solid load increases. It happened because methylene blue is cationic dye and it response to charged surface. Without any activation, the hydrochar only relies on its surface area and oxygen functional group (OFG) to attract the methylene blue. In higher temperature and solid load, more volatile is converted to fixed carbon and reduce the OFG as well as the methylene blue attraction to hydrochar surface.

On the other hand, CO<sub>2</sub> activation was not effective to increase the adsorption capacity. Worse than that, it reduces the adsorption capacity at some point. CO<sub>2</sub> activation significantly reduce the OFG in the char surface. Gradual increase in HC200-1A, HC220-1A and HC240-1A was observed. Those experiment points were conducted in solid load reactor of 0.1. There is higher possibility of porous formation in those operating condition because in low solid load there are more steam which also gives higher pressure. Although CO<sub>2</sub> activation remove the OFG, it gives enough charges to keep the char from adsorbing the methylene blue.

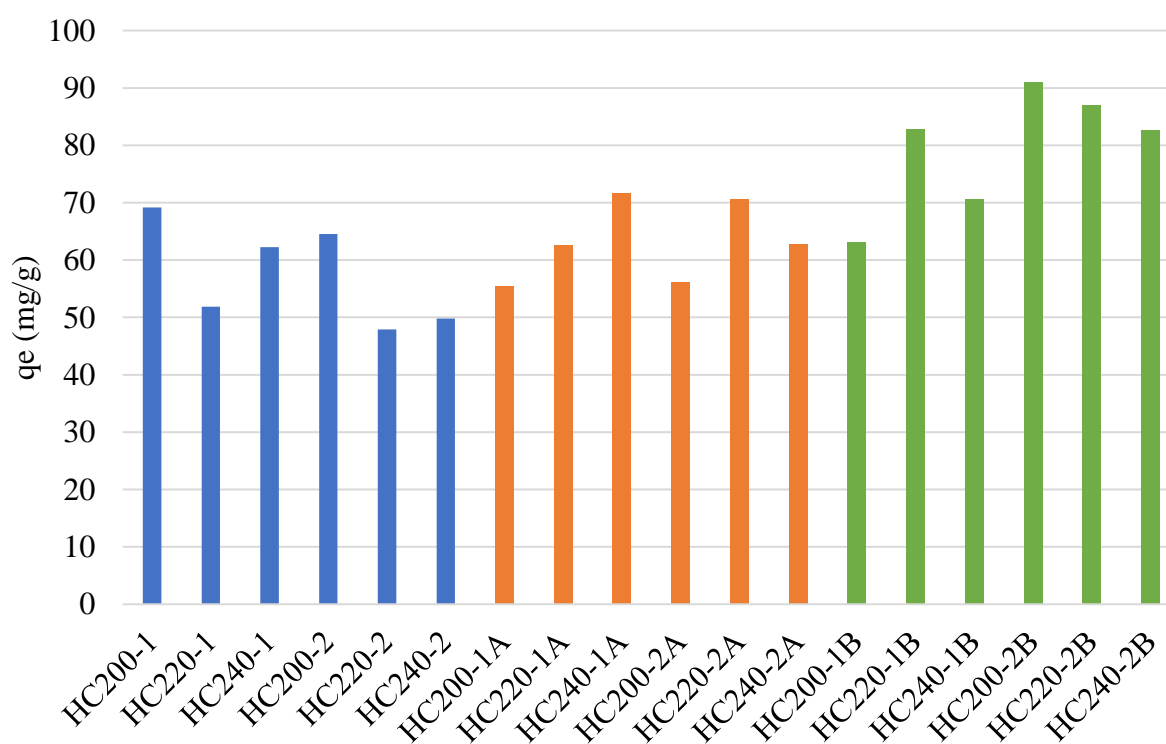
Organics adsorption process heavily depend on the surface area of the sorbent and functional group on the surface. As seen on FTIR analysis in figure 5.3, unexpectedly, CO<sub>2</sub> activation has removed some aliphatic carbon group (-CH<sub>n</sub>) in the material surface. The aliphatic carbon group (highlighted area in figure 3) was formed during the hydrothermal process from the degradation of biomass-based material. It causes less attraction towards organics to compare to hydrochar before activation.

It is seen in figure 5.1 and 5.2 (green) that after steam activation is effective for increasing the adsorption capacity and removal performance of methylene blue. Gradual reduction in HC200-2B, HC220-2B and HC240-2B was observed. Higher temperature tends to encourage the ash formation and thus, reduce the adsorption performance. Methylene blue adsorption capacity increases significantly to 91 mg/g for sample HC200-2B. After 12 hours adsorption, the maximum removal was obtained in steam activated hydrochar HC220-2B for 99.5%. High adsorption performance for steam activated char was mainly affected by its higher surface area. BET surface area were measure for steam activated char to confirm the methylene blue adsorption behavior.

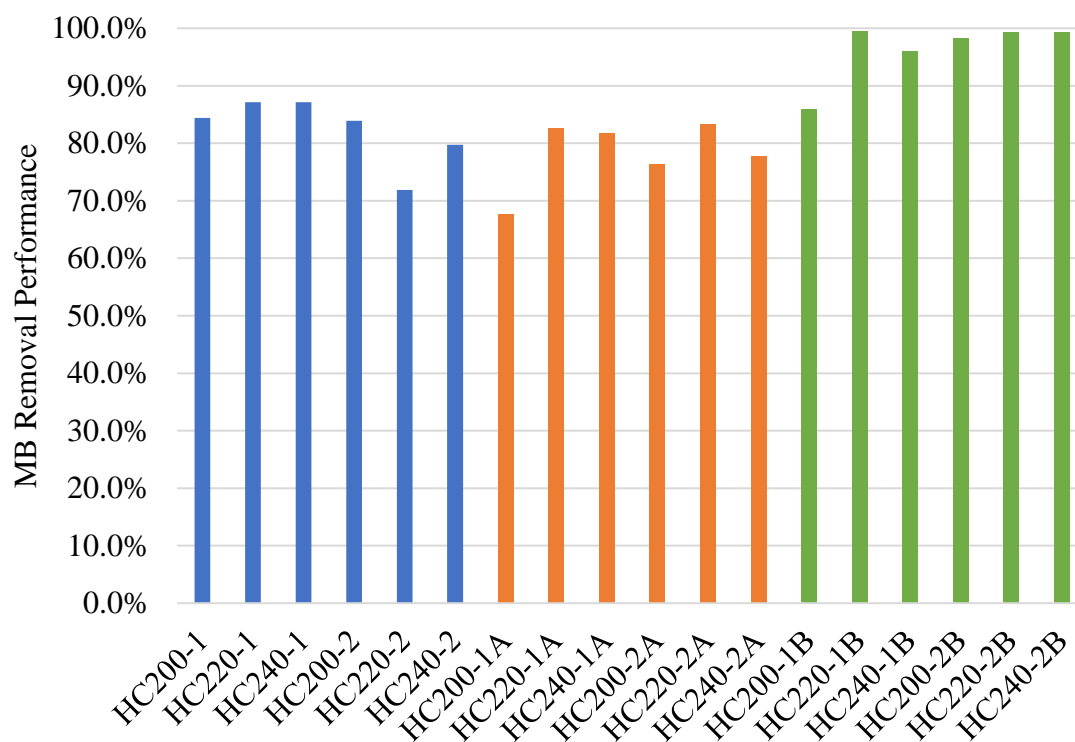
Nitrogen adsorption surface area measurements were performed at 77 K using a MICROMETRIC 3Flex BET Surface Area analyzer (Norcross, USA). The apparent nitrogen surface area was calculated using the BET equation for a pressure range  $P/P_0$  0.03 – 0.3. The highest BET surface area was found at 96.8  $\text{m}^2/\text{g}$  for steam activated hydrochar (Table 5.3).

**Table 5. 3 BET analysis of steam activated char**

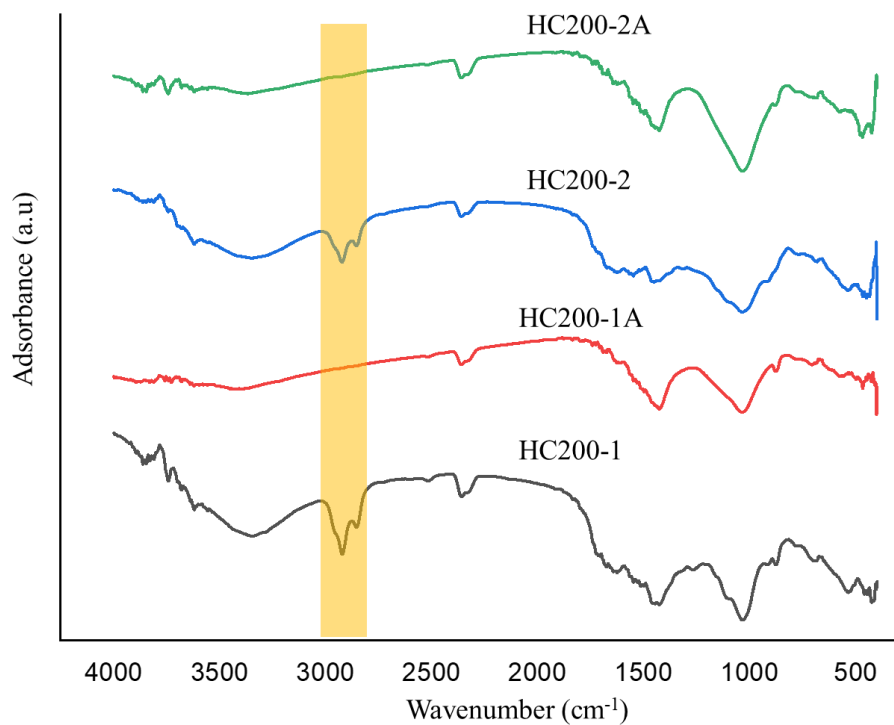
Temperature ( $^{\circ}\text{C}$ )	Solid Load	BET $\text{m}^2/\text{g}$
200	0.1	68.14
220	0.1	56.20
240	0.1	55.95
200	0.3	75.76
220	0.3	72.73
240	0.3	96.82



**Figure 5. 1 Methylene Blue adsorption capacity test**

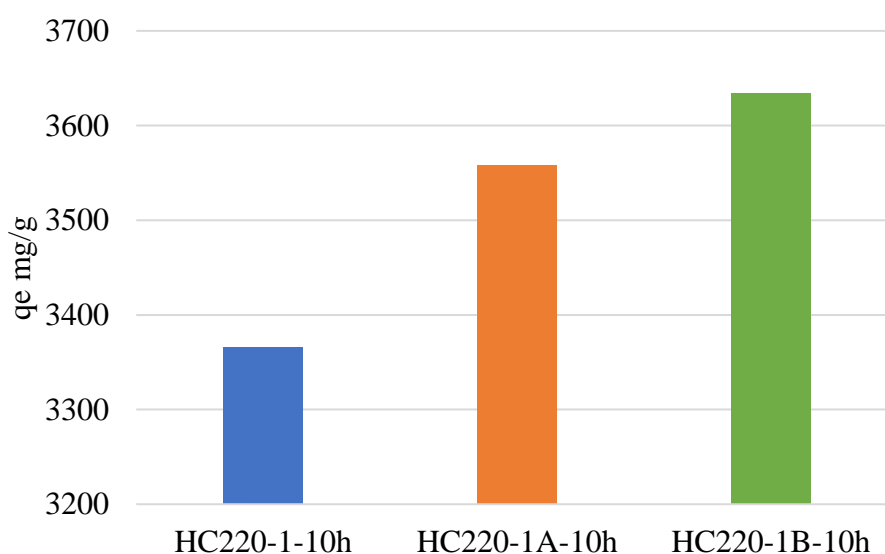


**Figure 5. 2 Methylene Blue removal performance**

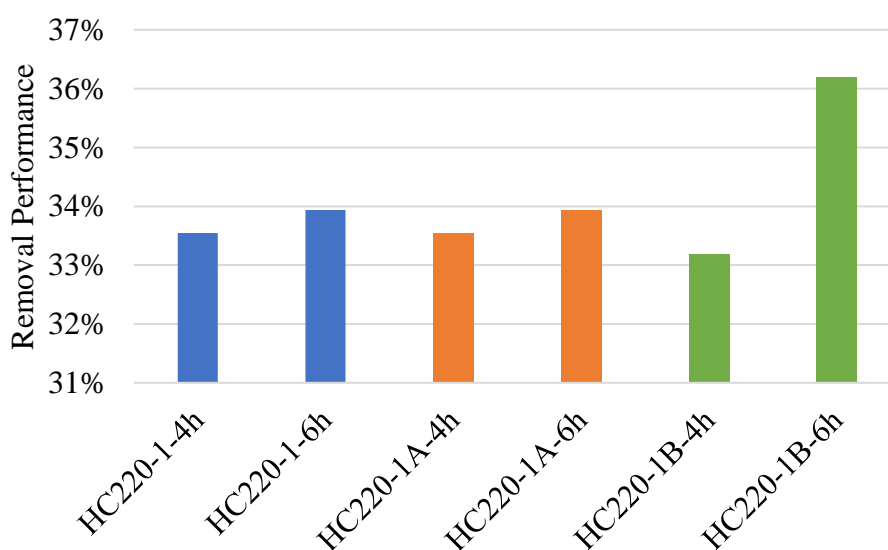


**Figure 5. 3 FTIR analysis of hydrochar and CO<sub>2</sub> activated hydrochar**

The aggregate organic constituent is generally indicated by chemical oxygen demand or COD. Figure 5.4 and 5.5 indicates that steam activation is the most effective way to increase the adsorption performance of hydrochar. The COD adsorption capacity for sample HC220-1B after 10h reaction was 3634 mg/g, increase 8% from before activation. The COD removal performance shows that although the removal in the first 4h reaction was lower for HC220-1B compare to other samples, it rapidly increases in the 6<sup>th</sup> reaction hour. Hydrochar and CO<sub>2</sub> activated hydrochar did not show much difference in COD removal performance.



**Figure 5. 4 COD adsorption capacity test**

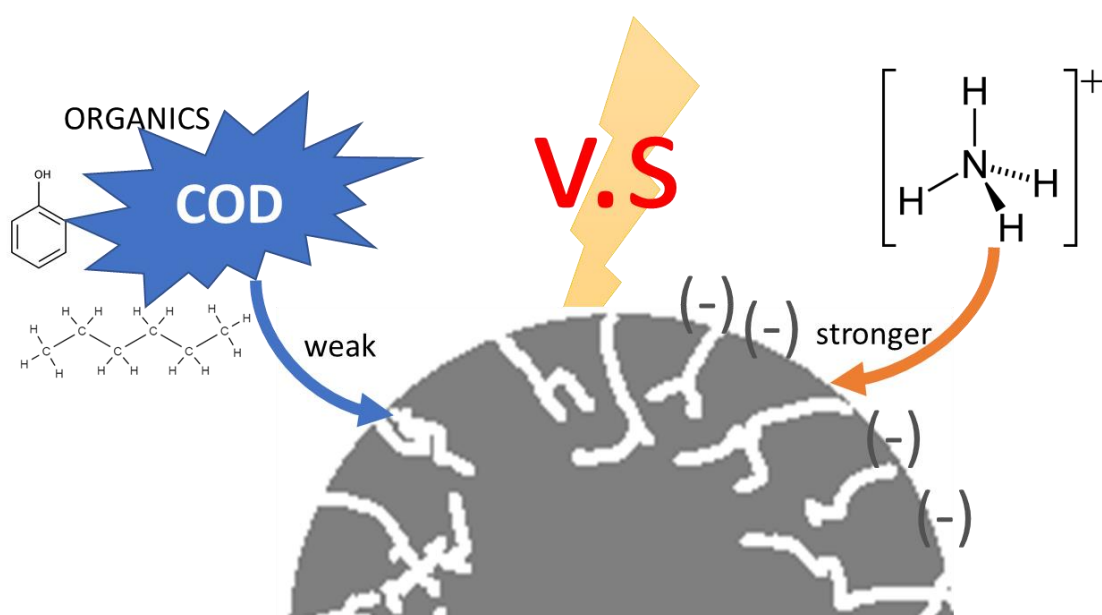


**Figure 5. 5 COD removal performance**



Our materials have effectively reduced the methylene blue concentration but not so practical in reducing COD content from landfill leachate. As we know that methylene blue is cationic substances, while landfill leachate is a complex substance consist of organics and inorganics. Sewwandi et al. (2013) investigate the properties of landfill leachate in Sri Lanka and found that the leachate was dominated by anionic substances, mainly  $\text{Cl}^-$ . Chloride is highly mobile under all conditions and it is not attenuated by soil which can be the reason for high Cl concentration in leachate though it flows through the soil.

As mentioned in Table 1, landfill leachate contains high ammonia concentration (279 mg/L). Ammonia has cationic properties, which made it highly attracted to the char because of the polar charge and its interfering with the adsorption of COD related substances which only depend on OFG in char surface and char surface area (figure 5.6). Furthermore, ammonia oxidation occurred during COD measurement which can cause an error measurement especially in the presence of Cl (Kim 1989).



**Figure 5. 6 Competition between ammonia and COD related substance**

The pore size distribution for one of the steam activated 220°C hydrochar was measured using 3Flex-micrometrics. The results show that the average BJH Adsorption average pore diameter (4V/A) is 21.481 Å (2.1481 nm). The particle size of wastewater was also measured using Zetasizer Dynamic Light Scattering. The results show that most of the particle in the wastewater (79.4%) has average diameter of 7.8 nm. We also found larger aggregate with average diameter of 3461 nm. The reason of

low adsorption efficiency is because the pore size is too small for the particle compound in the wastewater to be adsorb. Introducing higher pressure during hydrothermal treatment or longer reaction time during steam activation could possibly increase the pore size.

Compare to commercially available activated carbon, excavated waste char have a much lower surface area. However, in terms of alternative material, the result of this research has shown promising results compared to the previous studies. Halim et al. 2010 utilize composite material from zeolite, activated carbon, rice husk, and limestone bonded with cement. The material only has 61 m<sup>2</sup>/g BET surface area, and they need about 5 g/L dosage to achieve our performance. Steam activated hydrochar has a BET surface area up to 68.143 m<sup>2</sup>/g and could reach 33-36% COD removal with only 0.5 g/L dosage, which is ten times less than previous studies.

#### **4. Conclusion**

Excavated waste was a subject for hydrothermal carbonization and used as material to make adsorbent for COD removal from landfill leachate. Steam activation is the best method to increase the adsorption capacity of hydrothermally treated excavated waste. Sample HC220-1B attained the optimum result. Although the effectiveness is low, it is still better compared to alternative materials from previous studies.

The intrusion of ammonia in the COD adsorption process needs to be solved in the future. The material could be modified by adding zeolite which more attractive to ammonia compared to carbon. Doping the char with the oxidizer is also one option since the organic substance could directly be degraded faster. COD detection method to minimize the interference from ammonia oxidation is necessary to be developed.

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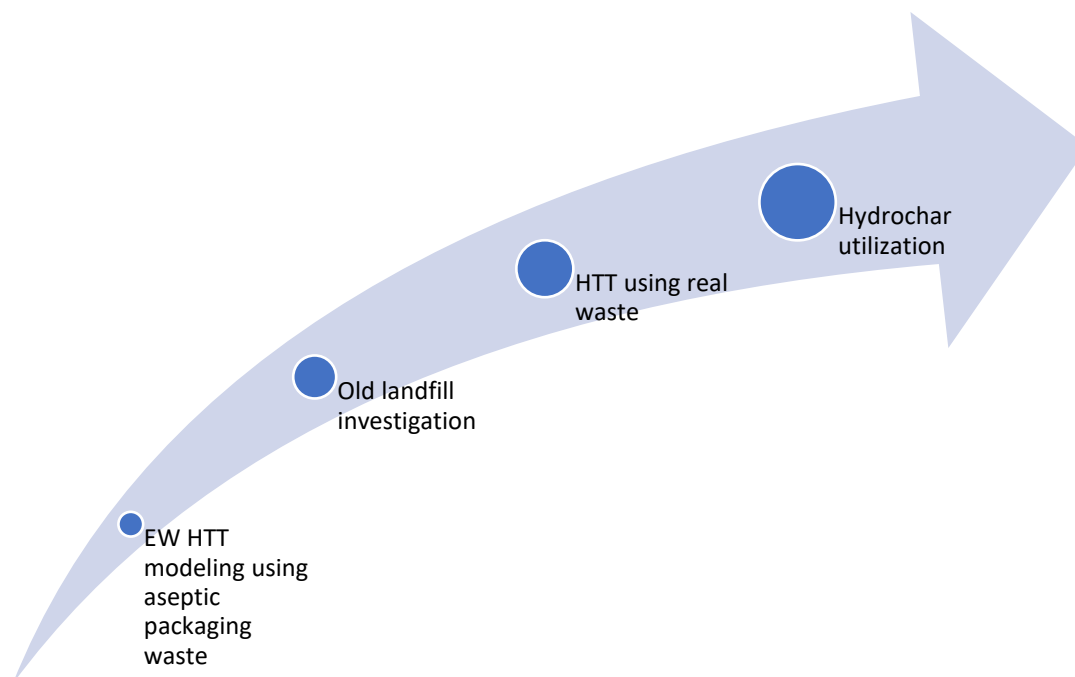
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## Chapter 6

### Conclusion and Recommendation



**Figure 6. 1 Outline of this research**

Outline of this research in Figure 6.1 shows the way to promote the Waste-to-Energy technology landfill mining project by using the hydrothermal treatment (HTT) to upgrade the excavated waste followed by potential examination as solid fuel and leachate adsorbent.

Landfill practice in developing countries has caused many environmental problems. Excavating the landfill and utilizing the material is necessary. Hydrothermal treatment is a promising technology to process the material. Landfill investigation in Indonesia shows that each landfill has distinguishable characteristics that were affecting the mining and recovery performance process. Since direct utilization of excavated waste is difficult, hydrothermal treatment was utilized to improve the quality of excavated waste.

Introducing hydrothermal treatment to process composite material such as aseptic packaging and excavated waste enhances the material by removing impurities to give better performance in energy

recovery facilities. The aluminum fraction in aseptic packaging was removed easily, and the hydrochar has high calorific value and carbon, oxygen, hydrogen ratio similar to lignite. The similar results were expected for the excavated waste cases, but the results show the opposite. It seems that the mineral composition is holding back the carboxylation process in hydrothermally treated excavated waste. Unlike aseptic packaging, which only has aluminum, excavated waste contains a high amount of Ca, Mg, and Mn, which tend to diffuse into the material and produce high ash content material. Alternative utilization as leachate adsorption was proposed. It shows that after activation, the char could reduce a significant amount of COD compare to the similar research done by another researcher.

The general conclusion both in and in-between each study can be summarized as follows:

1. Chapter 2, Hydrothermal treatment on aseptic packaging shows promising results for the use of HTT to separate and purify composite material. The results showed that hydrothermal treatment could effectively produce hydrochar, which is comparable to lignite after the aluminum part is removed. In addition, the aluminum and polyethylene composites were well-formed. The holding time and temperature had a positive influence on the results of the analyses. As the carbon content increased, the high heating value (HHV) also increased, whereas the ash content decreased accordingly. The highest calorific value was found at an operating temperature of 240 °C and a holding time of 60 min. Hydrothermal treatment can increase the calorific value of biomass from an Aseptic Packaging by up to 25.22 MJ/kg, which is comparable to lignite and coal.
2. Chapter 3, After investigating Jatibarang Landfill and Piyungan Landfill in Indonesia, on average, the waste composition on both landfills was dominated with soil-like (SL) material. The SL materials in Jatibarang Landfill and Piyungan Landfill were 52.2% and 70.2%, respectively. The second most found materials are plastics (JL=26.7% and PL=18%), followed by organic materials (JL=18.3% and PL=10%). Material flow analysis (MFA), coupled with process modeling, was conducted to simulate the material cycle and energy balance of landfill mining projects in both landfills. From WtE process modeling, the Jatibarang landfill produces more power and higher CO<sub>2</sub>-e compared to the Piyungan landfill. The MFA estimates the life span extended landfill site operation by 15 years for the Jatibarang landfill and 9 years for the Piyungan landfill. Not only to

recover precious material, but landfill mining could also be used to transform single used dumping into a multi-zone waste processing site to maintain the material cycle.

3. Chapter 4, The hydrothermal treatment on excavated waste shows opposite results from the expected one. The highest HHV and carbon content was found in the operating condition of 220 °C, 0.3 solid load, and 30 min holding time. The lowest ash content was obtained in the operating condition of 200 °C, 0.5 solid load, and 0 min holding time. The washing effect was also found in the lower solid load where some metals were detected in the liquid residue. Though, the hydrochar still retains a high amount of ash, which makes it unsuitable for solid fuel utilization.
4. Chapter 5, Hydrothermally treated excavated waste was seen as a promising adsorbent for organic substances. The preliminary test using Methylene Blue shows that there is a significant increase in adsorption capacity after steam activation from 71 mg MB / g adsorbent to 99.5 mg MB / g. Leachate from the Piyungan landfill was sampled and used in the COD removal experiment. COD in the leachate was reduced from 5071 mg/l to 3634 mg COD / g adsorbent with removal performance up to 36% using steam activated hydrothermally treated excavated waste.

For the recommendation on future work is to build an LCA formulation to extend the MFA analysis. Improvement of the product is also necessary. Chemical looping combustion (CLC) reactor might be one option instead of incineration to recover the energy and material from hydrochar. EW also contains a metal oxide, which could also be as hydrogen generation in the CLC reactor. Hydrothermal treatment is also possible to process soil-like material to become stable fertilizer. The plastic and cellulose residue in the soil fraction will be degraded into simple carbon, which readily accepted by plants. Future research should also find out the alternatives treatment without removing the soil-like material. The treated soil-like material is potentially used for fertilizer.

# APPENDIX



**A. Main operating parameter and block description for process modelling simulation**

<b>Phenomena</b>	<b>Block</b>	<b>Parameter</b>	<b>Description</b>
<b>Decomposition</b>	RYIELD	P = 1 bar; T = 25°C	Convert feedstock into element constituent based on known yield distribution
<b>Combustion</b>	RGIBBS	P = 1 bar	Rigorous reactor. Calculate phase and chemical equilibrium based on Gibbs free energy minimization
<b>SNCR</b>	RSTOIC	P = 1 bar; heat duty = 0; fractional conversion = 0.5	Ammonia injection. Stoichiometric reactor based on known fractional conversions.
<b>HRSG</b>	HEATER 1	Cold stream outlet T = 120°C	Reduce the temperature of hot gasses. Transfer the heat to HEATER 2
	HEATER 2	Cold stream outlet T = 600°C	Convert water into steam.
<b>Steam Turbine</b>	COMP	P = 1 bar Isentropic efficiency = 0.8	Generate electricity. Reduce steam pressure
<b>Condenser</b>	HEATER	Vapor Fraction = 0	Reduce steam temperature. Convert steam into water
<b>Acid gas treatment</b>	RSTOIC	P = 1 bar; heat duty = 0; fractional conversion = 0.9	Stoichiometric reactor based on known fractional conversions
<b>Pump</b>	PUMP	P = 125 bar Init Water = 4.35 kg/sec	Bring necessary amount of water calculated by targeting the 0-residual heat in the turbine unit.

## B. Pre-defined parameter for energy and emission analysis of landfill mining project

### a. Jatibarang Landfill Site Map



b. Piyungan Landfill Site Map



c. Utilities Specification

Equipment	Specification	Activity	Energy (GJ/h)	Emission (kg CO <sub>2</sub> /h)
Dump Truck	100 hp engine, 4m <sup>3</sup>	General Transportation	0.46	32
Excavator	133 hp engine	Excavation	0.61	42
Bull Dozer	82 hp engine	Restoration	0.37	26
Roller Vibro	100 hp engine	Restoration	0.46	32
Star Screen	25 kW electric motor	Separation	0.09	22*
Belt Conveyor	25 kW electric motor	Separation	0.09	22*

d. Transportation Details

<b>Activity</b>	<b>Jatibarang Landfill</b>	<b>Piyungan Landfill</b>
<b>From Excavation Site to Processing Site (Excavated Waste)</b>		
Distance (km)	1	1
Amount of roundtrip (times)	47	75
Working hours (hours)	6	6
Truck Unit (-)	2	3
<b>From Processing Site to Restoration Site (Soil-like material and Ash)</b>		
Distance (km)	1	1
Amount of roundtrip (times)	26	53
Working hours (hours)	3.6	5.2
Truck Unit (-)	3	3
<b>From Processing Site to Off-site treatment plant (Noncombustible)</b>		
Distance (km)	13	30
Amount of roundtrip (times)	1	1
Working hours (hours)	4	4
Truck Unit (-)	1	1

**C. ANOVA Table of Ultimate and Proximate Analysis of Hydrothermally Treated  
Aseptic Packaging**

a. Ultimate Analysis

Carbon

Source	Sum of Squares	df	Mean Square	F-value	p-value	
<b>Model</b>	0.0562	5	0.0112	20.27	0.0025	significant
<b>A-Temp</b>	0.0273	1	0.0273	49.23	0.0009	
<b>B-Time</b>	0.0092	1	0.0092	16.63	0.0096	
<b>AB</b>	0.0107	1	0.0107	19.25	0.0071	
<b>A<sup>2</sup></b>	0.0090	1	0.0090	16.16	0.0101	
<b>B<sup>2</sup></b>	0.0003	1	0.0003	0.5904	0.4770	
<b>Residual</b>	0.0028	5	0.0006			
<b>Lack of Fit</b>	0.0027	3	0.0009	32.66	0.0299	significant
<b>Pure Error</b>	0.0001	2	0.0000			
<b>Cor Total</b>	0.0590	10				

Hydrogen

Source	Sum of Squares	df	Mean Square	F-value	p-value	
<b>Model</b>	0.0004	5	0.0001	8.56	0.0171	significant
<b>A-Temp</b>	0.0002	1	0.0002	23.42	0.0047	
<b>B-Time</b>	0.0001	1	0.0001	7.46	0.0412	
<b>AB</b>	0.0000	1	0.0000	3.49	0.1207	
<b>A<sup>2</sup></b>	0.0001	1	0.0001	8.33	0.0343	
<b>B<sup>2</sup></b>	9.737E-06	1	9.737E-06	1.12	0.3377	
<b>Residual</b>	0.0000	5	8.668E-06			
<b>Lack of Fit</b>	0.0000	3	0.0000	44.01	0.0223	significant
<b>Pure Error</b>	6.467E-07	2	3.233E-07			
<b>Cor Total</b>	0.0004	10				

Nitrogen

Source	Sum of Squares	df	Mean Square	F-value	p-value	
<b>Model</b>	1.416E-06	3	4.719E-07	6.46	0.0200	significant
<b>A-Temp</b>	1.127E-06	1	1.127E-06	15.42	0.0057	
<b>B-Time</b>	1.667E-07	1	1.667E-07	2.28	0.1747	
<b>AB</b>	1.225E-07	1	1.225E-07	1.68	0.2364	
<b>Residual</b>	5.114E-07	7	7.306E-08			

<b>Lack of Fit</b>	3.714E-07	5	7.429E-08	1.06	0.5505	not significant
<b>Pure Error</b>	1.400E-07	2	7.000E-08			
<b>Cor Total</b>	1.927E-06	10				

Oxygen

Source	Sum of Squares	df	Mean Square	F-value	p-value	
<b>Model</b>	0.0490	5	0.0098	6.97	0.0263	significant
<b>A-Temp</b>	0.0222	1	0.0222	15.82	0.0106	
<b>B-Time</b>	0.0055	1	0.0055	3.92	0.1047	
<b>AB</b>	0.0093	1	0.0093	6.61	0.0499	
<b>A<sup>2</sup></b>	0.0118	1	0.0118	8.42	0.0337	
<b>B<sup>2</sup></b>	0.0004	1	0.0004	0.2751	0.6223	
<b>Residual</b>	0.0070	5	0.0014			
<b>Lack of Fit</b>	0.0069	3	0.0023	34.52	0.0283	significant
<b>Pure Error</b>	0.0001	2	0.0001			
<b>Cor Total</b>	0.0560	10				

HHV

Source	Sum of Squares	df	Mean Square	F-value	p-value	
<b>Model</b>	38.53	3	12.84	10.08	0.0062	significant
<b>A-Temp</b>	21.77	1	21.77	17.09	0.0044	
<b>B-Time</b>	7.57	1	7.57	5.94	0.0449	
<b>AB</b>	9.18	1	9.18	7.20	0.0313	
<b>Residual</b>	8.92	7	1.27			
<b>Lack of Fit</b>	8.87	5	1.77	70.30	0.0141	significant
<b>Pure Error</b>	0.0505	2	0.0252			
<b>Cor Total</b>	47.45	10				

b. Proximate Analysis

Volatile

Source	Sum of Squares	df	Mean Square	F-value	p-value	
<b>Model</b>	0.1422	3	0.0474	12.68	0.0032	significant
<b>A-Temp</b>	0.1128	1	0.1128	30.19	0.0009	
<b>B-Time</b>	0.0195	1	0.0195	5.21	0.0565	
<b>AB</b>	0.0099	1	0.0099	2.65	0.1479	
<b>Residual</b>	0.0262	7	0.0037			
<b>Lack of Fit</b>	0.0261	5	0.0052	134.32	0.0074	significant
<b>Pure Error</b>	0.0001	2	0.0000			
<b>Cor Total</b>	0.1683	10				

Fixed Carbon

Source	Sum of Squares	df	Mean Square	F-value	p-value	
<b>Model</b>	0.1420	3	0.0473	11.05	0.0048	significant
<b>A-Temp</b>	0.1021	1	0.1021	23.83	0.0018	
<b>B-Time</b>	0.0257	1	0.0257	6.00	0.0441	
<b>AB</b>	0.0142	1	0.0142	3.32	0.1114	
<b>Residual</b>	0.0300	7	0.0043			
<b>Lack of Fit</b>	0.0299	5	0.0060	98.47	0.0101	significant
<b>Pure Error</b>	0.0001	2	0.0001			
<b>Cor Total</b>	0.1720	10				

Ash

Source	Sum of Squares	df	Mean Square	F-value	p-value	
<b>Model</b>	0.0011	3	0.0004	1.56	0.2813	not significant
<b>A-Temp</b>	0.0003	1	0.0003	1.14	0.3202	
<b>B-Time</b>	0.0004	1	0.0004	1.87	0.2140	
<b>AB</b>	0.0004	1	0.0004	1.68	0.2361	
<b>Residual</b>	0.0016	7	0.0002			
<b>Lack of Fit</b>	0.0012	5	0.0002	1.28	0.4926	not significant
<b>Pure Error</b>	0.0004	2	0.0002			
<b>Cor Total</b>	0.0027	10				

**D. ANOVA Table of Ultimate and Proximate Analysis of Hydrothermally Treated  
Excavated Waste**

a. Ultimate Analysis

Carbon

Source	Sum of Squares	df	Mean Square	F-value	p-value	
<b>Model</b>	71.16	1	71.16	3.59	0.0908	not significant
<b>A-Temp</b>	71.16	1	71.16	3.59	0.0908	
<b>Residual</b>	178.56	9	19.84			
<b>Lack of Fit</b>	115.58	7	16.51	0.5242	0.7819	not significant
<b>Pure Error</b>	62.99	2	31.49			
<b>Cor Total</b>	249.73	10				

Hydrogen

Source	Sum of Squares	df	Mean Square	F-value	p-value	
<b>Model</b>	5.42	2	2.71	4.67	0.0453	significant
<b>A-Temp</b>	3.84	1	3.84	6.61	0.0331	
<b>BC</b>	1.58	1	1.58	2.73	0.1372	
<b>Residual</b>	4.64	8	0.5806			
<b>Lack of Fit</b>	1.94	6	0.3239	0.2399	0.9267	not significant
<b>Pure Error</b>	2.70	2	1.35			
<b>Cor Total</b>	10.07	10				

Nitrogen

Source	Sum of Squares	df	Mean Square	F-value	p-value	
<b>Model</b>	0.3160	1	0.3160	6.68	0.0295	significant
<b>B-Solid Load</b>	0.3160	1	0.3160	6.68	0.0295	
<b>Residual</b>	0.4258	9	0.0473			
<b>Lack of Fit</b>	0.3345	7	0.0478	1.05	0.5702	not significant
<b>Pure Error</b>	0.0913	2	0.0456			
<b>Cor Total</b>	0.7418	10				



# Oxygen

Source	Sum of Squares	df	Mean Square	F-value	p-value	
<b>Model</b>	306.23	3	102.08	3.87	0.0639	not significant
<b>A-Temp</b>	49.55	1	49.55	1.88	0.2129	
<b>C-Time</b>	86.12	1	86.12	3.26	0.1138	
<b>AC</b>	170.56	1	170.56	6.46	0.0385	
<b>Residual</b>	184.73	7	26.39			
<b>Lack of Fit</b>	89.64	5	17.93	0.3771	0.8360	not significant
<b>Pure Error</b>	95.09	2	47.54			
<b>Cor Total</b>	490.96	10				

# Sulfur

Source	Sum of Squares	df	Mean Square	F-value	p-value	
<b>Model</b>	0.0016	2	0.0008	8.38	0.0109	significant
<b>C-Time</b>	0.0008	1	0.0008	8.38	0.0200	
<b>AB</b>	0.0008	1	0.0008	8.38	0.0200	
<b>Residual</b>	0.0008	8	0.0001			
<b>Lack of Fit</b>	0.0002	6	0.0000	0.0909	0.9902	not significant
<b>Pure Error</b>	0.0006	2	0.0003			
<b>Cor Total</b>	0.0024	10				

# Chlorine

Source	Sum of Squares	df	Mean Square	F-value	p-value	
<b>Model</b>	2.18	7	0.3117	10.71	0.0387	significant
<b>A-Temp</b>	0.2520	1	0.2520	8.66	0.0604	
<b>B-Solid Load</b>	0.0512	1	0.0512	1.76	0.2767	
<b>C-Time</b>	0.6384	1	0.6384	21.93	0.0184	
<b>AB</b>	0.4050	1	0.4050	13.91	0.0336	
<b>AC</b>	0.3961	1	0.3961	13.60	0.0346	
<b>BC</b>	0.1800	1	0.1800	6.18	0.0888	
<b>ABC</b>	0.2592	1	0.2592	8.90	0.0584	
<b>Residual</b>	0.0873	3	0.0291			
<b>Lack of Fit</b>	0.0687	1	0.0687	7.39	0.1128	not significant
<b>Pure Error</b>	0.0186	2	0.0093			
<b>Cor Total</b>	2.27	10				

HHV

Source	Sum of Squares	df	Mean Square	F-value	p-value	
<b>Model</b>	40.61	1	40.61	4.81	0.0560	not significant
<b>C-Time</b>	40.61	1	40.61	4.81	0.0560	
<b>Residual</b>	76.04	9	8.45			
<b>Lack of Fit</b>	34.71	7	4.96	0.2400	0.9357	not significant
<b>Pure Error</b>	41.33	2	20.66			
<b>Cor Total</b>	116.66	10				

b. Proximate Analysis

Volatile

Source	Sum of Squares	df	Mean Square	F-value	p-value	
<b>Model</b>	235.86	1	235.86	7.68	0.0217	significant
<b>A-Temp</b>	235.86	1	235.86	7.68	0.0217	
<b>Residual</b>	276.36	9	30.71			
<b>Lack of Fit</b>	268.16	7	38.31	9.35	0.1000	not significant
<b>Pure Error</b>	8.20	2	4.10			
<b>Cor Total</b>	512.22	10				

Fixed Carbon

Source	Sum of Squares	df	Mean Square	F-value	p-value	
<b>Model</b>	564.73	7	80.68	15.65	0.0227	significant
<b>A-Temp</b>	134.38	1	134.38	26.06	0.0145	
<b>B-Solid Load</b>	11.76	1	11.76	2.28	0.2282	
<b>C-Time</b>	62.80	1	62.80	12.18	0.0398	
<b>AB</b>	0.7951	1	0.7951	0.1542	0.7208	
<b>AC</b>	326.00	1	326.00	63.23	0.0041	
<b>BC</b>	9.82	1	9.82	1.91	0.2613	
<b>ABC</b>	19.18	1	19.18	3.72	0.1494	
<b>Residual</b>	15.47	3	5.16			
<b>Lack of Fit</b>	5.34	1	5.34	1.06	0.4123	not significant
<b>Pure Error</b>	10.13	2	5.06			
<b>Cor Total</b>	580.19	10				

Ash

Source	Sum of Squares	df	Mean Square	F-value	p-value	
<b>Model</b>	193.26	7	27.61	38.63	0.0255	significant
<b>A-Temp</b>	7.03	1	7.03	9.83	0.0884	
<b>B-Solid Load</b>	10.73	1	10.73	15.01	0.0606	
<b>C-Time</b>	50.68	1	50.68	70.91	0.0138	
<b>AB</b>	7.22	1	7.22	10.10	0.0864	
<b>AC</b>	64.33	1	64.33	90.02	0.0109	
<b>BC</b>	22.39	1	22.39	31.34	0.0305	
<b>ABC</b>	30.89	1	30.89	43.23	0.0224	
<b>Curvature</b>	101.25	1	101.25	141.69	0.0070	
<b>Pure Error</b>	1.43	2	0.7146			
<b>Cor Total</b>	295.94	10				

### E. ANOVA Table of methylene blue adsorption behavior

#### a. Adsorption Capacity

Source	Sum of Squares	df	Mean Square	F-value	p-value	
<b>A-Temp</b>	0	1	0	0	0.9872	
<b>B-Solid Load</b>	28.4	1	28.4	0.387	0.5566	
<b>C-Activation Method</b>	1563	2	781.5	10.663	0.0106	significant
<b>AB</b>	92.3	1	92.3	1.26	0.3045	
<b>AC</b>	248.2	2	124.1	1.694	0.2611	
<b>BC</b>	367.6	2	183.8	2.508	0.1616	
<b>ABC</b>	8.5	2	4.2	0.058	0.9443	
<b>Residuals</b>	439.7	6	73.3			

#### b. Methylene Blue Removal Performance

Source	Sum of Squares	df	Mean Square	F-value	p-value	
<b>A-Temp</b>	0.005	1	0.005	1.8	0.23422	
<b>B-Solid Load</b>	0.00004	1	0.00004	0.01	0.91462	
<b>C-Activation Method</b>	0.1	2	0.05	17.7	0.00306	significant
<b>AB</b>	0.007	1	0.007	2.3	0.18287	
<b>AC</b>	0.004	2	0.002	0.6	0.56163	
<b>BC</b>	0.01	2	0.007	2.2	0.19666	
<b>ABC</b>	0.0004	2	0.0002	0.07	0.93388	
<b>Residuals</b>	0.02	6	0.003			