

## Evaluation of Pyrolysis as a Method of Waste Recovery from the Non-Metallic Fraction of Printed Circuit Board Waste

Songpol Boonsawat\*  
Griffith School of Engineering, Griffith University, Australia

Premrudee Kanchanapiya  
National Metal and Materials Technology Center, Thailand

Erik L.J. Bohez  
Asian Institute of Technology, Thailand

Soon Chia  
Viro Group Inc., USA

\* Corresponding author. E-mail: songpol.boonsawat@griffithuni.edu.au DOI: 10.14416/j.ijast.2017.12.002  
Received: 16 June 2017; Accepted: 14 July 2017; Published online: 6 December 2017  
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### Abstract

Pyrolysis has been examined as an effective alternative waste disposal treatment, compared with land filling and incineration of solid waste. The pyrolysis technology allows minimal energy usage, material utilization and a cleaner resource recovery. This research addresses state of the art Pyrolysis of the Non-Metallic Fraction (NMF) of Printed Circuit Boards (PCBs), after the copper separation process is complete. The aim is reducing emissions and increasing the production quality at optimal conditions. The main objectives of this research are to demonstrate the feasibility of using pyrolysis to recover NMF material and results of the experiments were aimed to make a critical argument to the current waste disposal treatments by comparing the possibilities of using pyrolysis as waste disposal treatment. This research investigates technological developments, production yield, production quality and the environmental impacts, to create an argument against current waste disposal treatments, which are not an ideal solution for NMF of PCBs. Within this research, firstly the characteristics of NMF materials and the elemental composition of the materials are reviewed and investigated. Then, the influence of important operating parameters such as reaction temperature, heating rate and processing time in the reaction zone of the pyrolysis process are reviewed and scrutinized. Next, the pyrolysis production characteristics, yield quality and the main properties of the products are explored and summarized. In the final section, the emissions from the NMF pyrolysis process, such as CO, NO<sub>x</sub>, Sox and other possible formation gases are reviewed and summarized. It can be concluded that the innovative pyrolysis process is an effective waste recovery method, especially for NMF in terms of maximizing material utilization. Based on this research information, the prospect of applying this innovative pyrolysis technology to effectively dispose of the NMF material is evaluated and it is suggested that this is a more suitable and effective waste disposal treatment for NMF material.

**Keywords:** Pyrolysis, PCB, Printed Circuit Board Waste, Non-metallic fraction of PCB

Please cite this article as: S. Boonsawat, P. Kanchanapiya, E. L. J. Bohez, and S. Chia, "Evaluation of pyrolysis as a method of waste recovery from the non-metallic fraction of printed circuit board waste," *KMUTNB Int J Appl Sci Technol*, vol. 10, no. 4, pp. 263–278, Oct.–Dec. 2017.

## 1 Introduction

With the rapid development of technology in the electronic industry and the frequent updating of Electrical and Electronic Equipment (EEE), electronic waste (e-waste) has been increasing extremely over a relatively short period of time [1], [2]. The current waste disposal treatments available for electronic waste material are not coping with the increasing supply of waste materials from various types of EEE products [1]. Printed Circuit Board scraps (PCBs) are a typical e-waste component often found in the waste stream [3]. PCBs are particularly problematic to recycle because of the diversity and the complex mix of materials involved [2].

### 1.1 *Emerging issues of NMF waste generation and the available waste disposal treatment options*

Non-Metallic Fractions (NMF of PCBs) are often found as leftover materials after the copper separation process [4]. The main disadvantages of the NMF materials create challenges due to their diversity and the complex mix of materials involved. This mix consists of brominated epoxy resin (thermoset polymer), glass reinforced fiber and various ceramic materials [2], [4]. Therefore, this increases the difficulty of the recycling or recovery process. The current processes for disposal of waste NMF from PCBs in recycling facilities are both environmentally unfriendly and hazardous. The use of primitive technologies have increased over the past years due to the limitation of any suitable technologies for recycling NMF material [5], [6]. Massive amounts of dumping of NMF waste has taken place along with other primitive disposal treatments that do not aim to close the loop of the product [3], [7]. This practice is increasing and generating secondary pollution for the planet. The open burning of NMF waste is also a common method of waste disposal. Disturbingly, dioxin and furans can be generated due to the combustion of BFR in the NMF material [8], [9].

### 1.2 *Rationale of the pyrolysis technology design for NMF material*

Pyrolysis as a new waste disposal method is a

technology that not only can recover valuable materials but it can also deal with the complex organic matter found in NMF material [10], [11]. The differences of the thermal properties that each material the NMF of PCBs consists of, allows the total separation of each of these materials [12]. The thermoset polymer found in NMF material is a valuable resource and can be used to produce fuel [13]. The innovative design of the pyrolysis process, especially for the pyrolysis of NMF material produces lower emissions and less pollution when compared to current waste disposal methods [14].

### 1.3 *Aims and objectives*

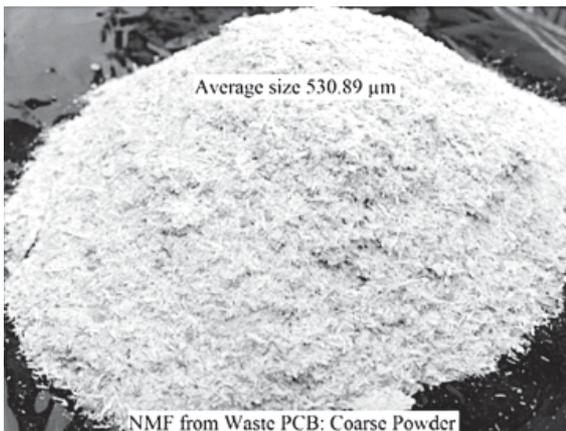
For the benefit of this work, an up-scaled, batch experiment for the pyrolysis of NMF material was designed and used to investigate the optimal conditions for pyrolysis. The results of the experiments were used to make a critical argument by comparing the possibilities of using pyrolysis with current waste disposal treatments. Thermogravimetric analysis was used initially to find the characterization of thermal decomposition of the NMF material, but the main body of work was carried out using a 300 cm × 21 cm diameter tubular reactor that was heated to various target temperatures. All the experiments with multiple target temperatures contained different heating rates and therefore gave insights into the effects the different temperature and heating rates had on the pyrolysis reaction. The by-products from the pyrolysis of the NMF material, including oil, gas and solid residues, were characterized in detail; in particular, the potential of the oil by-product as a potential feedstock or petroleum-substitute fuel was investigated.

## 2 Materials and Methods

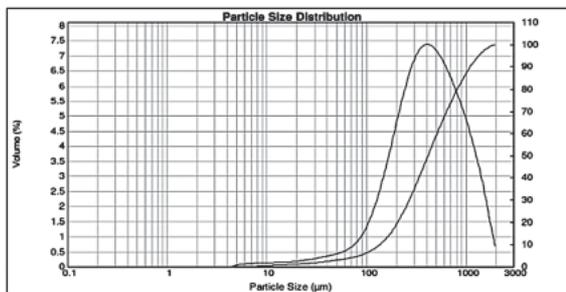
### 2.1 *Sample preparation*

The samples for this experiment were obtained from Thailand's National Metal and Material Technology Center (MTEC) and consisted of 2 types of NMF material: coarse powder and fine powder. (Figure 1 and Figure 2)

Particle size was analyzed to investigate physical



(a)

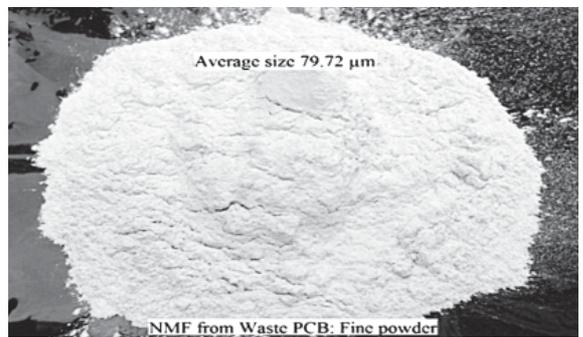


(b)

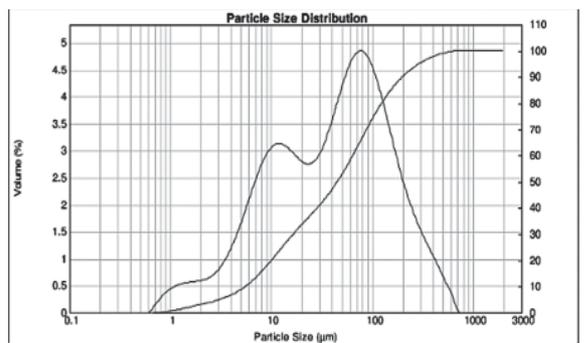
**Figure 1:** (a) Image of “NMF Coarse powder”: Non-Metallic Fraction (NMF) extracted during the PCB’s copper separation process. (b) Average particle size distribution of coarse powder NMF material. This type of NMF material is usually sent back for copper separation and becomes a fine powder.

changes (size) during the process by using Malvern, Mastersizer 2000 Ver 5.60, Serial Number: MAL 1021434 with air dispersion at Medium (dry dispersion) and a pressure of 2 bar at a 50% feed rate. (Instrument settings: Beam Length 10 mm, Particle RI 1.600 and Absorption 0.1). These fractions were expected to contain approximately 50% polymer with a higher density of additives, including BFRs, 50% of mixed Glass Reinforced Fiber (GRF) and Ceramics.

These materials were obtained in powder form and the average is based on a distribution of particles ranging from 5.48 μm (fine powder)– 1115.6 μm (coarse powder). For these distributions, the following results were calculated and presented in the form of D Values.



(a)



(b)

**Figure 2:** (a) Image of “NMF Fine powder”: Non-Metallic Fraction (NMF) extracted during the PCB’s copper separation process. (b) Average particle size distribution of fine powder type of NMF material. This type of NMF material is usually found as a left over product from the copper separation process of PCBs.

*Fine NMF sample powders.*

D(0.1): 5.48 μm/ D(0,5): 44.89 μm/ D(0.9): 198.29 μm/ D(4,3): 79.72 μm/ D(3,2): 12.47 μm/ Span 4.295 μm/ Uniformity: 1.4

*Coarse NMF sample powder*

D(0.1): 137.32 μm/ D(0,5): 418.07 μm/ D(0.9): 1115.91 μm/ D(4,3): 530.89 μm/ D(3,2): 208.6 μm/ Span: 2.339 μm/ Uniformity: 0.709

The D value is the most common used metric to describe particle size distributions where;

D(0.1) is the intercept for 10% of cumulative population.  
 D(3,2) is volume/surface mean.  
 D(4,3) is the mean diameter over volume.  
 D(0.5) is the intercept for 50% of cumulative population.  
 D(0.9) is the intercept for 90% of cumulative population.

These results are fairly typical where the D (4.3) values from both samples is 79.72  $\mu\text{m}$  for fine NMF and 530.89  $\mu\text{m}$  for coarse NMF respectively. They are also larger than their D (0.5) based on the volume basis median value for both samples. Therefore, the NMF particle size distribution can be estimated as ranging between 79.72  $\mu\text{m}$ – 530.89  $\mu\text{m}$ .

The NMF material was screened for material characteristics before processing in the pyrolysis reactor using both Scanning by Electron Microscopy (SEM) and Energy Dispersive X-Ray Spectroscopy (EDS). SEM-EDS analyses of material was performed to demonstrate the characteristics and the chemical composition of the NMF samples [15], [16]. The samples were tested via SEM and images were produced of the sample by scanning with a focused beam of electrons.

The electrons interacted with atoms in the NMF samples and produced various signals that contain information of the sample's surface topography and composition (as shown in Figure 9). These are in 3 modes x45, x500 and x1000 respectively.

SEM observation was used to carry out analysis of the NMF 's characteristics A Hitachi SU5000 Schottky FE-SEM system with the resolution of 3.0 nm at 15kV variable pressure mode, low vacuum system mode, a magnification sensitive area of 50 mm<sup>2</sup> 10~600000x, and the energy resolution of < 145 eV at 300000 cps (count rate Cu > 400000 cps) was used to acquire a set of X-ray maps at 1 ms dwell time per pixel to get approximately 1 million counts. For the elemental analysis of NMF material, EDS was used, in which elemental mapping data was gathered at an accelerating voltage of 15.0 kV.

The temperature of the NMF material was monitored to ensure it had reached the target temperature before processing. The ground fine NMF powder was selected for the experiment because it is more typically the material found

leftover after the PCB separation process, while the coarse powder will normally be sent back for a further copper separation process [4], [5].

The elemental composition of the material was determined and the results are shown in Table 1. The Carbon, Hydrogen, Nitrogen and Oxygen contents of the NMF material were determined by using a Leco TruSpec® CHN (micro) Analyser. The Leco TruSpec® CHN (micro) Analyser was also used to analyse the CHN and O composition of the NMF material under conditions of 1100°C in an oxygen atmosphere. Helium was used as the conductor. The results obtained through CHNS elemental analyser are mentioned in Table 1.

**Table 1:** Elemental composition of NMF material

NMF Sample	Elemental Composition of NMF (%Wt)			
	C	H	N	S
PCB1	28.656	3.267	0.5409	0.0530
PCB2	27.693	3.298	0.6255	0.0520
PCB3	29.434	4.032	0.5552	0.0510
Sum PCB	28.594	3.532	0.5740	0.0520

The Elemental composition of NMF material was tested with 3 replications. Carbon and hydrogen content are the main elements in the NMF sample, as expected. The average ratio between carbon and hydrogen is approximately 8:1 where average carbon is 28.594% and hydrogen is 3.532%. Nitrogen and Sulphur content are very low compared to those of carbon and hydrogen in the NMF material, they are approximately 0.574% and 0.052% respectively.

The thermal decomposition characterization of the NMF material was tested by TGA and DTA in order to establish operating temperature guidelines for the design of pyrolysis experiments. Further design criteria was gathered from preliminary testing, material properties, material characteristics, influencing factors, important parameters, operating conditions etc. to determine the possibility and feasibility of producing liquid fuel from the NMF material [17].

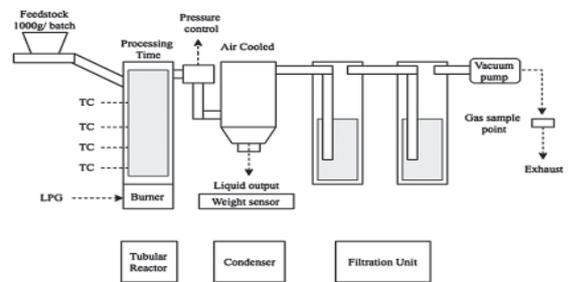
The preliminary material testing was conducted using Thermo Gravimetric Analysis (TGA), which analyses the effective temperature range and processing time of the NMF material that was selected for the design of this experiment [18]. The

TGA (Universal, V4.7A TA instruments Model SDT Q600) was used to observe the phase transition of material decomposition, effective key temperatures, endothermic heat requirements and the effective degradation temperature of the NMF material. TGA testing was performed with 12.5°C/min heating rate. Temperature was raised from room temperature to 1000°C.

## 2.2 Equipment and conditions

The NMF samples were pyrolyzed inside a tubular reactor in batch experiments. Approximately 1000 grams per batch was used for the NMF pyrolysis experiments. The reactor was externally heated using a commercial LPG burner. The physical material characteristics, chemical properties of NMF materials, role in the kinetic reaction of the thermal process and the size were all confirmed through material testing and analysis before the planning and design the experiment. The experimental setup for NMF pyrolysis was supported by the Viro Group Inc. A 1kg pyrolysis system was adopted for the present investigation. The set up consisted of a tubular type stainless-steel reactor, a furnace, an industrial burner, condenser units, flares, pipe, a gas filtration system, a Liquid Petroleum Gas (LPG) tank, a gas flow meter, a pressure transmitter, pressure valves and a regulator. The main intent of this experimental part was to examine the effects of the key pyrolysis parameters, which are reactor temperatures, heating rates, pressures and residence times. It was also to examine their interdependency on the output production by conducting experiments at 4 heating rates (12.5°C/min, 20°C/min, 30°C/min and 40°C/min).

1000 grams of cleaned and dried NMF feedstock of 79.72  $\mu\text{m}$  in size of fine NMF powder was used as input material in all the experiments. The material size reduction was performed during the separation process and this will improve the packing density inside the reactor and improve the uniform heat and flow rates around the particle surfaces of the NMF samples. Fast pyrolysis was adopted during all 4 targeted temperatures and with 4 different heating rates. The heating rate was controlled by the LPG gas flow meter, the pressure



**Figure 3:** A schematic diagram of the pyrolysis reactor and system.

regulator and the adjustable industrial burner.

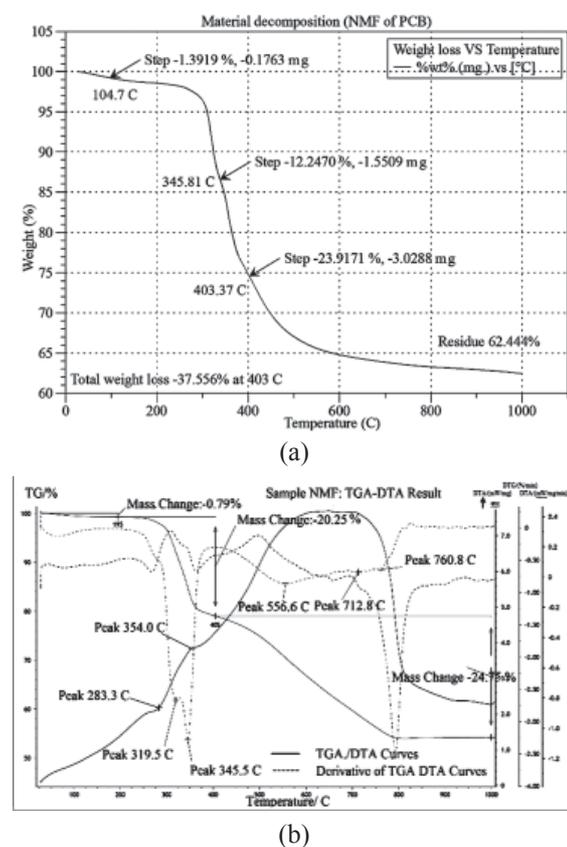
The experiments started with the investigation of an effective cracking temperature. The temperature was set at 300°C, 403°C, 500°C and 600°C. This was observed along with the quality, production yield and the emission release.

By controlling the LPG flow rate at 1.25, 2.0, 3.0 and 4.0 m<sup>3</sup>/h allowed the achievement of constant heating rates of 12.5, 20, 30 and 40°C/min respectively. The temperatures were measured by using k type thermocouples which are located within the inner space of the reactor [19]. In all experiments, an identical pyrolysis environment inside the reactor was created by vacuum system at -125 mbar. The schematic diagram of the pyrolysis system is shown in Figure 3.

## 3 Results and Discussions

### 3.1 Thermal decomposition characterization of NMF via TGA

The NMF material was heated in (TGA), shown in Figure 4 (a) and then tested with Differential Thermal Analysis (DTA) shown in Figure 4 (b) to confirm the range of effective decomposition temperatures for the NMF material. These techniques, in which a physical property of a substance is measured as a function of temperature, are conducted while the substance is subjected to a controlled temperature program. In DTA, the temperature difference that occurs between the sample and the inert reference material is measured while both are subjected to identical heat treatment conditions.



**Figure 4:** Thermal decomposition of the NMF material via (a) TGA Thermo gravimetric analysis and (b) DTA differential thermal analysis of the NMF material for the confirmation of heating and temperature range.

In Figure 4, the TGA curve shows that the NMF sample has an average maximum weight loss at approximately 403°C. It reflects the possible effective decomposition temperature for the NMF material and could be considered for use in the actual NMF pyrolysis experiment to produce liquid fuels. From Figure 4(a), it could be summarized that NMF thermal decomposition of the NMF material is influenced by 3 different temperatures. The TGA curve shows that firstly between 104.7°C–345.81°C the NMF starts its cracking process to break its chemical bonds and it starts to decompose once the temperature is higher than their thermal resistance properties. Secondly, in the temperature range between 345.81°C–403.37°C the NMF material

starts to change state and become gas. It is obvious that the most effective decomposition temperature is in this range, as shown in the steep curve in Figure 4(a).

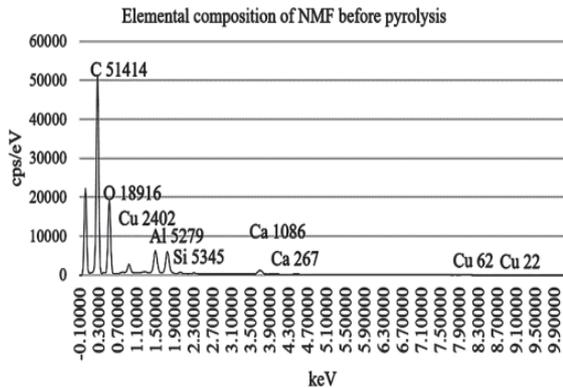
Meanwhile, the DTA data curves shows the average of 20.25% NMF mass change is at approximately 405°C, which matches the TGA data obtained. After the material starts to change state from solid to gas at 405°C it is assumed there is a high possibility that the volatile gases can be condensed to liquid fuel at the condensing unit during the actual experiment of NMF pyrolysis.

The preliminary results from the TGA testing show that the effective temperature range is between 403°C–405°C for critical mass change. This could be used as a key temperature range in the NMF pyrolysis experiment design, to investigate and observe for maximum yields, quality and optimal processing time. The sample test results for average optimal liquid production (with highest yield) was obtained in the range of 400°C–405°C at 12.5°C/min heating rate. The corresponding time to complete the process is longer than with other higher heating rates. The optimal liquid yield processing time was found to be 33 minute. The liquid production obtained at optimum liquid yield conditions were considered for further testing and analysis. The liquid and wax product yield and residue in the reactor was weighed. The gaseous product mass was calculated by using the mass balance. A number of experiments were conducted at each heating rate and each targeted temperature, to check the consistency of the experiments results. The experiment results and values were found to be within a 4% variation.

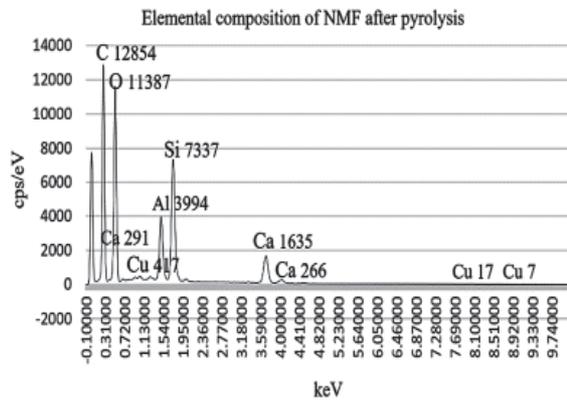
### 3.2 EDS-SEM material and output analysis

A stereo-microscope was used in the first step of the NMF material examination before the NMF pyrolysis experiment. It provides useful information for the initial screening and discrimination of the NMF material [20]. The statistics of the quantitative elemental data found from the EDS spectrum map, such as carbon, aluminium, silicon, calcium, copper and oxygen are shown in Figures 5 and 6.

The reason to choose carbon, aluminium, silicon, calcium, copper and oxygen as the target elements for analysis or screening is that; firstly, these elements are the basic components of the NMF



**Figure 5:** Quantitative elemental data for the NMF material before the pyrolysis process.

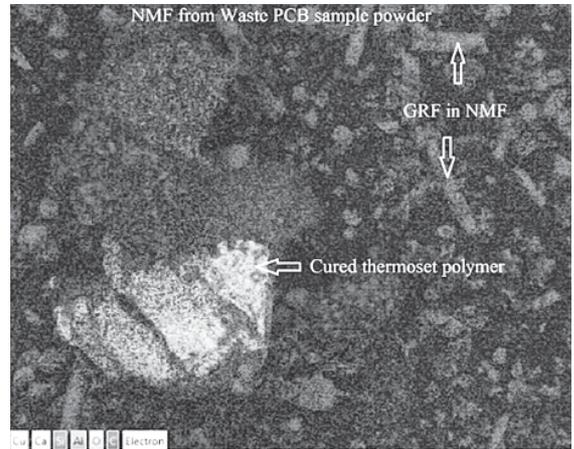


**Figure 6:** Quantitative elemental data for the NMF material after the pyrolysis process.

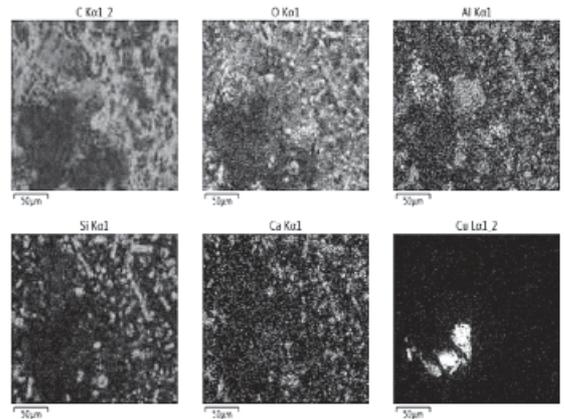
material, and secondly, they can possibly be added or mixed with the environment and create pollution if found in high percentages [21].

The main disadvantage of the EDS analysis is the small amount of NMF material in each trial. Therefore the results established using this technique do not represent the exact quantitative values of those targeted elements in the NMF material [22]. However as the material analysis is a comparative guide to the material composition after the pyrolysis process and the ratio of all the peaks that were obtained in different runs of particular samples remains undisturbed, the data serves at least as an expressive pattern of the NMF discrimination [5].

It's obvious to see from the quantitative data that the carbon was significantly reduced by 74.99% from 51414 before the process to 12854 after the



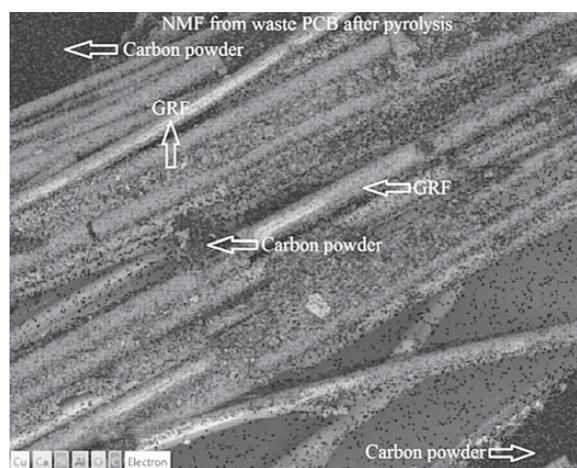
(a)



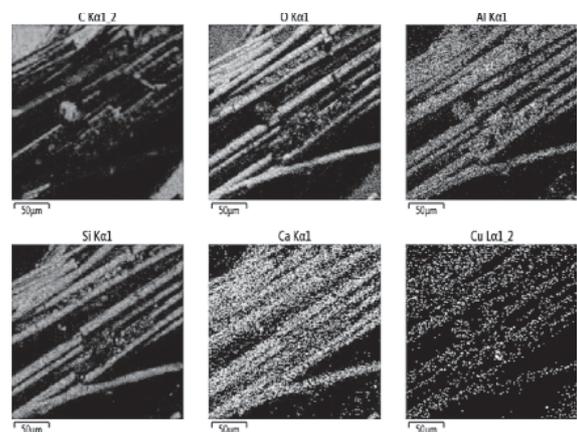
(b)

**Figure 7:** SEM micrograph of non-metallic materials from waste PCB (a) overall SEM image of NMF from waste PCB (b) Image of material's surface before the pyrolysis process.

process. More over the oxygen is was reduced 39.80% whereas Cl, Cu, Al and Si were slightly reduced. Figure 7(a) shows images of the NMF sample's surface before being processed in the pyrolysis experiment. In Figure 7(b), the material is containing high percentage of carbon content, the oxygen created surface of the NMF material in Figure 7(b) could have been formed from the thermos chemical reaction during the copper separation process. These images mostly represent the powder characteristics of the NMF material as shown in Figure 9 (a).



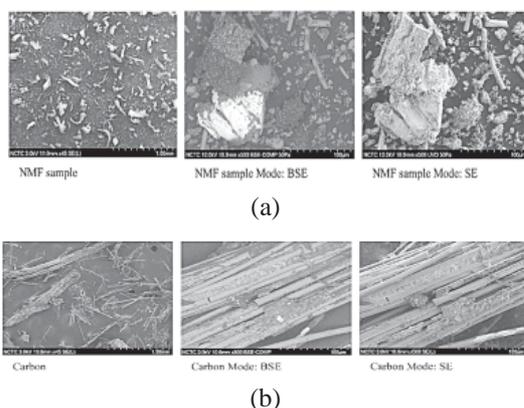
(a)



(b)

**Figure 8:** SEM micrograph of residue after pyrolysis. (a) Overall SEM image of residue (b) Image of residue's surface after the pyrolysis process.

Figure 8 (a), shows images of the material after the process where recoverable material with lower thermal property was converted into byproduct. It is obvious to see that the NMF sample after the process has changed in material structure and only left a residue of short glass reinforced fiber mixed with carbon. The material surfaces and structures have been changed from powder to dried straight cuttube and it is more like a glass reinforce fiber. The images show that the surface of the material is coated with a lower elemental contents than the input sample from before pyrolysis processing.



(a)

(b)

**Figure 9:** A typical image of NMF material before and after the pyrolysis process: (a) An NMF sample before the pyrolysis process with three steps of image resolution captured from x45, x100 and x500 respectively, (b) An NMF sample after the pyrolysis process.

### 3.3 NMF pyrolysis yield

The weight of liquid oil and gas yield are summarized in Table 2. The amount of oil condensed by the system is estimated at 45.007% of the sample input weight. The weight of gases (Including volatiles) was calculated as the difference between the input weight and the total weight of the residues and condensed oils. These weights were then converted to percentages of the original weight of input. The results are shown in Table 2.

**Table 2:** Analysis results of the pyrolysis process of scrap NMF powder

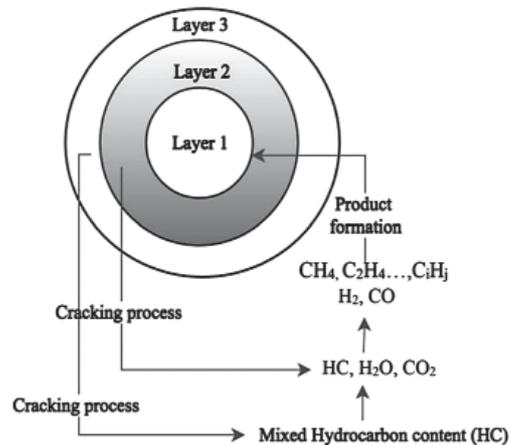
Liquid Yield	Yield (g)				Reaction time (min)
	T 300°C	T 403°C	T 500°C	T 600°C	
20°C/min	383.21	450.07	235.01	191.9	33
30°C/min	327.21	392.71	192.21	180.51	27
40°C/min	277.64	347.85	182.63	169.11	18
Non-condensable gaseous yield	Yield (g)				Reaction time (min)
	T 300°C	T 403°C	T 500°C	T 600°C	
20°C/min	109.327	42.467	257.527	300.637	33
30°C/min	165.327	99.827	300.327	312.027	27
40°C/min	214.897	144.687	309.907	323.427	18

Table 2 shows a summary of the liquid output yield from NMF pyrolysis. The results show that the main influencing factors for NMF pyrolysis in this study are the heating rate and the reactor temperature. The maximum output liquid yield was obtained at a heating rate of  $12.5^{\circ}\text{C}/\text{min}$  but it produced a waxier by-product. We did not take that into the account for this study.

The maximum yields for both condensable and non-condensable gases were obtained from different operating temperatures and heating rates. This means that the cracking process, gas forming and the condensing reaction of gases are all optimal for each particular reaction of the material at selected temperatures. In other words, a high heating rate could rush the cracking process, and a high temperature could cause a forming phenomenon and create non-condensable gases. The optimal temperature in this study was performed at  $403^{\circ}\text{C}$  and with a  $20^{\circ}\text{C}/\text{min}$  heating rate for the NMF pyrolysis experiment. The liquid yield was 450.07 g, and the non-condensable gases were calculated to be 42.467 g. The total reaction time was 33 mins.

The residue left after the process was expected to be carbon mixed with short length fiberglass [23]. However, there is high quality carbon powder found in the reactor which can be further separated. The original NMF material had been entirely converted and separated into organic elements and produced a fine carbon powder. The carbon content of the NMF powder was reduced by 31.428% after the pyrolysis process, whilst there is an increment of oxygen content at an average 57.143%. During the process, there is a possibility of some additional oxygen being incorporated onto the newly created surface inside the reactor once heat is introduced, because the reactor temperature was set to increase from a room temperature of  $30^{\circ}\text{C}$  before being heated [24]. The oxygen content in the reactor was not a sufficient factor in the pyrolysis process according to Figures 5 and 6.

From Table 2 it can be estimated that the potential fuel was produced at a rate of approximately 45.007% of the original feed stock weight (material composition ratio) (Polymer: GRF: Other) equivalent to (49.2537: 49.2537:1.4925). Or a 91.378% yield of polymer content from the NMF material was converted to pyrolysis fuel, which is made up of oil and flammable



**Figure 10:** The principle of pyrolysis for a single NMF particle where (Layer 1) Dry zone (Layer 2) Pyrolysis zone (Layer 3) Cracking zone.

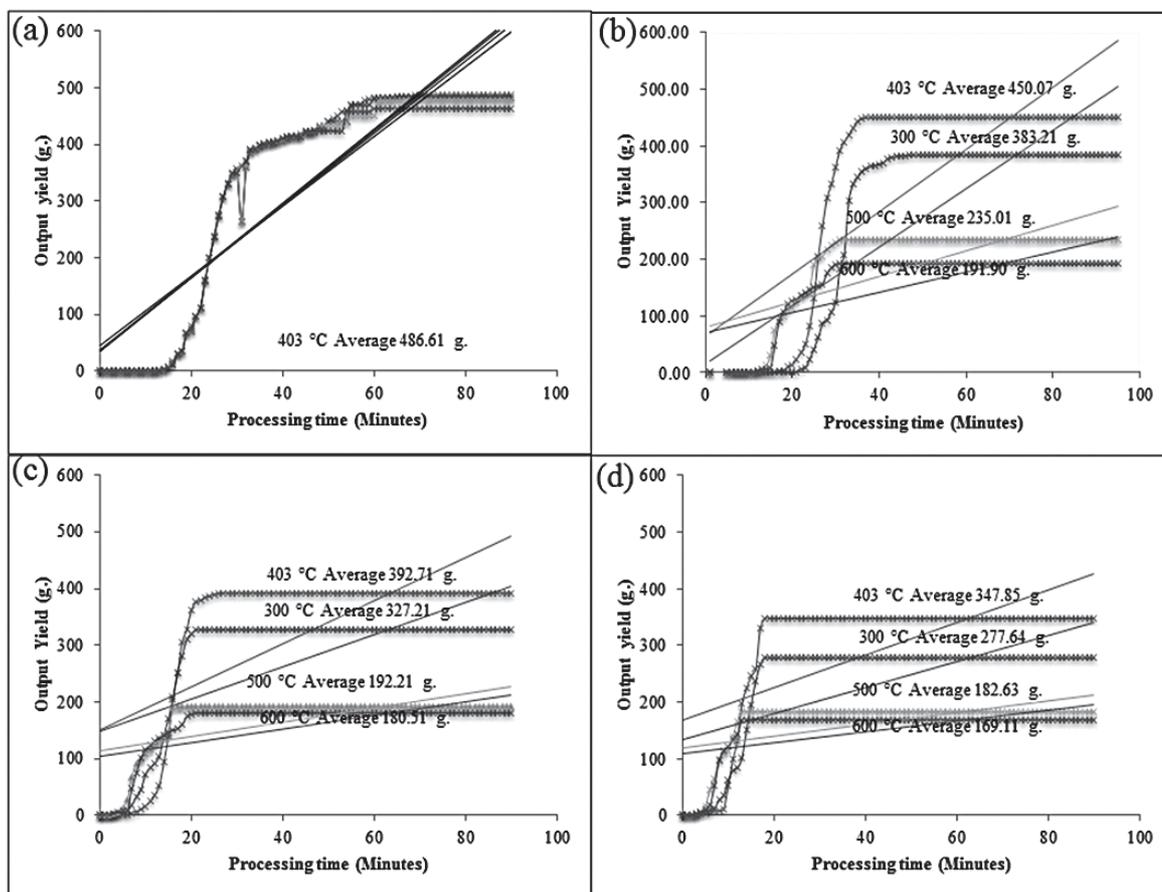
gases [6], [25]. However, the solid residues require additional treatments and further separation processes [26].

Figure 10, shows the principles of NMF particle pyrolysis in this study. At a targeted temperature (High temperature between  $300^{\circ}\text{C}$ – $600^{\circ}\text{C}$ ), the volatile parts such as hydrocarbons and gases are released during the cracking process of hydrocarbon and leave a solid residue by-product behind. [27]. The reaction of the hydrocarbon product during the cracking process is able to form carbon and hydrogen byproducts consisting of a range of gases in the secondary and ternary reactions [10], [28].

### 3.4 Liquid phase output analysis

The experimental runs for pyrolysis of NMF were carried out at 4 different temperatures of  $300^{\circ}\text{C}$ ,  $403^{\circ}\text{C}$ ,  $500^{\circ}\text{C}$  and  $600^{\circ}\text{C}$  and with different heating rates [29]. Figure 11 shows the degree of the yield change versus time at different temperatures.

The yield sharply increases at  $403^{\circ}\text{C}$  with the maximum yield among the compared temperatures at various heating rates. Since the TGA results from the test of NMF material has matched the pyrolysis testing results, the temperature of  $403^{\circ}\text{C}$  can be considered the optimal temperature for pyrolysis of NMF material. Figure 11 shows that different heating rates gave different results and different operating



**Figure 11:** Pyrolysis yield within ranges of temperature and heating rates (a) NMF pyrolysis at 12.5°C/min heating rate (b) NMF pyrolysis at 20°C/min heating rate (c) NMF pyrolysis at 30°C/min heating rate (d) NMF pyrolysis at 40°C/min heating rate.

temperatures affect the production yield. Secondly, the experiments were carried out using various heating rates. The results from Figure 11(a), (b), (c) and (d) show that the yields are affected by the heating rate in the same way as by the target temperatures. The lower heating rate requires extra time for the cracking process to be complete while also allows time for the product formation to be completed [14].

However, the lower heating rate also creates a slow cracking process therefore the hydrocarbons may not be at enough high temperature to chemical formation and condense as liquid oil. As a result, the lower heating rate gives a higher yield but produces a waxy form of material output at some point. From the 4 different heating rates at targeted temperatures

ranging from 12.5°C/min, 20°C/min, 30°C/min and 40°C/min. At 30°C/min and 40°C/min, produced a significant lower amount of liquid yield with a lighter liquid fuel in dark colour, whilst the 20°C/min gave a higher yield with a better quality of liquid fuel in term of appearance, when comparing the quality with an acceptable quantity yield from these experiments. The acceptable level heating rate which is higher than 12.5°C/min and it was apparent that 20°C/min is recommended as optimal condition for the pyrolysis of NMF material in this experiment. In experimental parts, the operation temperature ranged from 300°C to 600°C and heating rates varied from 12.5°C/min<sup>-1</sup> to 40°C/min<sup>-1</sup> respectively with a fixed condensing temperature of 30°C. It was

assumed that the weight loss of the NMF material was induced by the decomposition of brominated epoxy resin under the pyrolysis reaction conditions. The decomposition rate of resin was determined by investigating the efficiency of the pyrolysis reaction. The following formula (Equation 1) was used to calculate the decomposition rate of the polymer (brominated epoxy resin):

$$\text{Pyrolysis Efficiency (\%)} = \frac{(W_{input} - W_{Solid\ residue})}{W_{polymer\ in\ input}} \times 100 \quad (1)$$

$W_{input}$  : is the weight of initial material.

$W_{solid\ residue}$  : is the weight of solid residue.

$W_{polymer\ in\ input}$  : is the theoretical weight of resin in the initial material.

Experimental errors caused by operation and the loss of product was regarded as system error and could be ignored by an acceptable level of ability to repeat the experimental procedure. The yield analysis was summarized as shown in Figure 12.

To summarize, the results from the experiments in this study demonstrate that the maximum yield [shown in Figure 11(a)] results from a heating rate of 12.5°C/min, producing a higher yield in percentage by weight of the input material. However, this does not take into the account that the output characteristic was not formed in liquid but was formed a flammable waxy material. Figure 12 shows the summary of the yield efficiency of NMF pyrolysis. Maximum pyrolysis efficiency was calculated, it is obviously seen that 20°C/min heating rate at 403°C was the most effective condition. The pyrolysis efficiency was approximately 87.3217% for liquid fuel production. The minimum efficiency of pyrolysis of NMF was approximately 61.3466% at the condition with 40°C/min heating rate at 600 °C. The yield effect on the NMF material during the pyrolysis process are defined in Equation 2.

$$\text{Yield efficiency (\%)} = \frac{W_{yield}}{W_{polymer\ in\ input}} \times 100 \quad (2)$$

Pyrolysis Efficiency: rate of decomposition (%wt)

where;

$W_{yield}$ : is weight of output fuel.

$W_{polymer\ in\ input}$ : is theoretical weight of resin in the initial material.

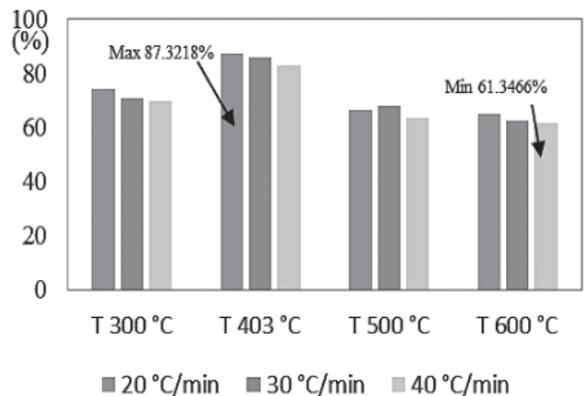


Figure 12: Summary of NMF pyrolysis yield efficiency.

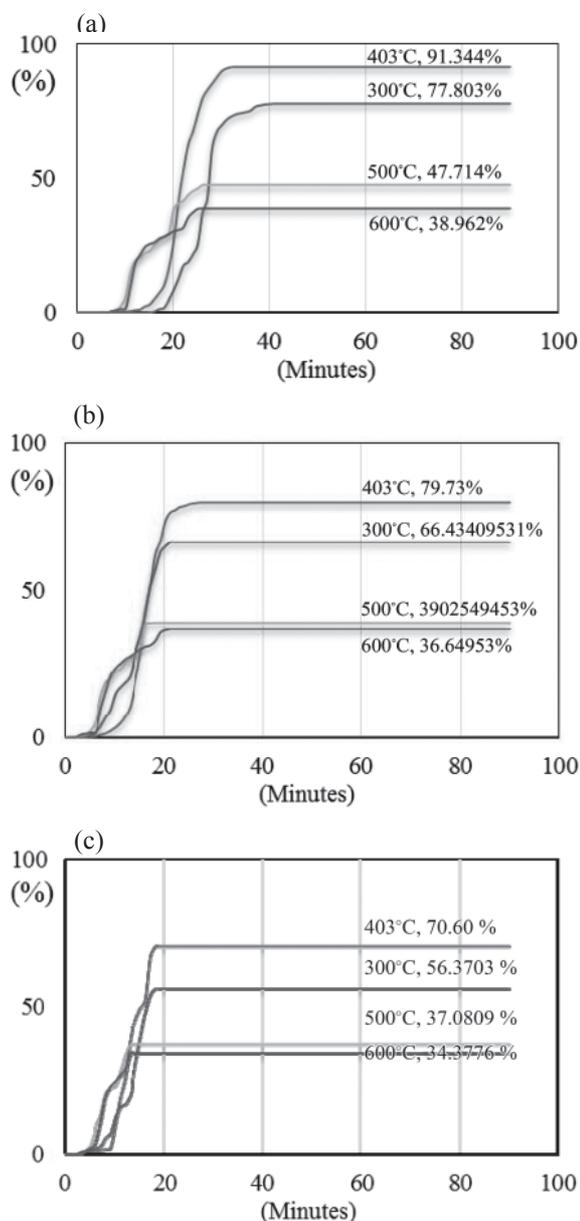
From Figure 13, The yield efficiency sharply increases after the process started at different time. The maximum yield efficiency approximately 91.344% was obtained from the operating temperature at 403°C. Among the different heating temperature, the higher temperature allows quicker cracking process with higher temperature but reduce the ability for the liquid to be form properly. Therefore, it can be concluded that non-condensable gases are increased and produced at high temperature than produced at the lower temperature.

The comparison data in Table 3 shows the elemental composition of CHNOS of raw material versus the pyrolysis oil and diesel. It is seen that the NMF pyrolysis oil contains 86.1836% carbon content which is the same as diesel.

Table 3: The composition of CHNOS of raw NMF material was compared to the pyrolysis oil and diesel

Composition %wt	NMF %wt	Pyrolysis Oil %wt	Diesel %wt
Carbon (C)	87.3045	86.1836	86.4
Hydrogen (H)	10.7840	9.9562	14.01
Nitrogen (N)	1.7525	0	-
Sulphur (S)	0.1587	0	-
Oxygen (O)	-	0.22	-

The carbon content of NMF material ensures the final liquid product contains rich hydrocarbons. The hydrogen content among those three samples, NMF, NMF pyrolysis oil and diesel are slightly



**Figure 13:** Yield efficiency rate of NMF pyrolysis at different heating rates (a) 20°C /min (b) 30°C/min (c) 40°C/min.

different, approximately 10.7840%, 9.9562% and 14.01% respectively. NMF contains 1.7525% nitrogen where NMF pyrolysis oil and diesel contain none. There is a quantity of oxygen found in pyrolysis

oil which may occur during NMF pyrolysis process.

The output liquid was collected and tested according to ASTM fuel standards, the results are shown in Table 4. There are 4 testing components that are not available in diesel, these are ash, specific gravity, water content and sediment. The flash point of NMF pyrolysis oil is approximately 40°C while diesel standard requires 52°C as a minimum. The combustion heat of gross diesel is approximately 32.40%, less than diesel standard. The pure point of diesel is approximately -16°C as minimum while the NMF pyrolysis oil is around 27°C on average.

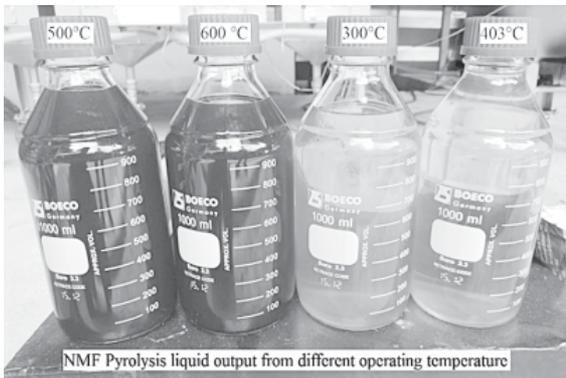
However, the sulphur content from pyrolysis oil is significantly better than diesel whereas the rest of the test methods are not much different.

**Table 4:** Fuel testing analysis and report *Standard value of Diesel*

Test Item	Test Method	Unit	Results
Ash	ASTM D 482-13	%wt	-
Flash Point	ASTM D 93-16	°C	52 Minimum
Heat of CombustionGross	ASTM D 240-14	cal/g	10038.24–10755.26
Pour Point	ASTM D 97-16	°C	-16 Minimum
Sulphur	ASTM D 4294-16e1	%wt	0.5
Viscosity at 50 °C	ASTM D 445-15a	mm <sup>2</sup> /s	1.9–4.1
Specific Gravity at 15.6/15.36 °C	ASTM D 4052-15	-	-
Water and Sediment	ASTM D 1796-11(R16)	%vol	-

*Physical, thermal and chemical property results of NMF pyrolysis oil*

Test Item	Test Method	Unit	Results
Ash	ASTM D 482-13	%wt	0.764
Flash Point	ASTM D 93-16	°C	40
Heat of CombustionGross	ASTM D 240-14	cal/g	7270
Pour Point	ASTM D 97-16	°C	27
Sulphur	ASTM D 4294-16e1	%wt	Less than 0.01
Viscosity at 50°C	ASTM D 445-15a	mm <sup>2</sup> /s	4.545
Specific Gravity at 15.6/15.36°C	ASTM D 4052-15	-	1.0768
Water and Sediment	ASTM D 1796-11(R16)	%vol	36



**Figure 14:** Liquid output from NMF pyrolysis.

In Figure 14, It can be seen that at higher temperatures above 403°C (500°C and 600°C), the reaction temperature at the reactor unit causes differences of liquid colour and liquid yield. The lower reaction temperature provides light clear colour of output liquid due to the time allowance for gas to form liquid phase and therefore the less excess carbon were formed in the output liquid.

**3.5 NMF pyrolysis emission testing and analysis**

The emission testing details for air quality measurements during NMF pyrolysis is shown in Table 5. The testing parameters are particulate, sulphur dioxide, nitrogen oxide and carbon monoxide of which we are able to compare with the incineration of NMF material. Results are shown in Table 6.

**Table 5:** Details of the work plan for the environmental quality measurement by viro solutions group PTY LTD [2], [11], [18], [30]

Measure Items	Parameter	Analytical Technique
Quality of air contaminants emitted from stack (LPG-HC combined close system)	Particulate	Isokinetic, Gravimetric Method
	Sulphur Dioxide SO <sub>2</sub>	Absorption, Barium-Thorin Titrimetric Method
	Oxide of Nitrogen NO <sub>x</sub>	Absorption, Phenoldisulfonic Acid Method
	Carbon Monoxide	Electrochemical Sensor Method

Table 5 shows the measure items of quality of air and the emission from NMF pyrolysis and the details of the air quality measurement are as follow:

- Measure date: 4/11/2016
- Used fuel: LPG + Hydrocarbon gas combined closed system
- Material: Non-Metallic Fraction of PCB (PCB fine powder)
- Height of Stack: 2 Meters
- Diameter of stack: 0.15 meters
- Temperature within stack: 391°C
- Atmospheric pressure within stack: 754.01 mm. Hg
- Velocity of air within stack: 10.55 m/s
- Flow rate of air within stack: 0.19 m<sup>3</sup>/s
- Percentage of Oxygen: 11.20
- Percentage of Carbon dioxide: 6.58
- Percentage of Moisture: 3.44

**Table 6:** Results of quality measurement of air contaminants emitted from NMF pyrolysis [5], [13], [30]

Parameter	Unit	Result	Standard1	Evaluation
Particulate	mg/m <sup>3</sup>	4.37	<320	Excellent
Sulphur Dioxide	ppm	<1.3	<60	Excellent
Oxide of Nitrogen	ppm	131	<200	Good
Carbon Monoxide	ppm	178	<690	Excellent

Standard1: American standard testing method for emission from stack oven

The analytical technique used to measure are shown in Table 5. Where the basic parameters are particulate, SO<sub>2</sub>, Nitrogen Oxide and Carbon Monoxide. The results of the emission testing are shown in Table 6.

Overall results in Table 6 are excellent where Particulate was measured at approximately 4.37 mg/m<sup>3</sup>, SO<sub>2</sub> was approximately less than 1.3 ppm from 60 ppm standard, oxide of nitrogen was 131 ppm from the standard of 200 ppm and the carbon monoxide was measured at 178 ppm which also less than the emission standard.

Moreover, the open flame combustion testing was conducted 4 times and the results showed that there was consistency of the experiment. In this

study 20 ml of each fuel was used, such as Unleaded Petrol (ULP), Diesel and Kerosene to be examined and compared with the Pyrolysis fuel named (Viro fuel) via an open flame combustion method. The results showed that the Viro fuel was burnt longer than ULP, Diesel and Kerosene in all trials. It was measured that the Viro fuel has a slightly lower heating value when compared to Diesel and ULP, however the NMF pyrolysis fuel burnt longer than the ULP and Diesel by 253% and 184%, respectively due to the presence of alkane, alkenes, alcohol and constituents with a higher molecular weight of hydrocarbon content. The leftover carbon product in the testing crucible and smoke was significantly less than that of the ULP and Diesel.

The micro graphical image in Figure 9 that was obtained from the SEM of NMF material, revealed the presence of large particles that appeared to be formed by several flaky particles stacked together in the form of a material group of tubular agglomerates. These impurities were not supremely observed. EDS analysis of the NMF powder was conducted to ascertain the constitutions and changes in hydrocarbon content (ratio between C and H) due to the effect of the thermal process, these are indicators of the hydrocarbon products and their qualities. From the results shown in Table 2. It can be concluded that the optimal conditions for the pyrolysis of NMF material obtained from this study is at 403°C, with a heating rate of 20°C per minute. The output was totally converted to a liquid flammable product as shown in Figure 14.

#### **4 Conclusions**

This study has provided a concise summary of the plastic materials in the NMF of PCB pyrolysis for each type and a discussion of the main affecting parameters to optimize liquid oil yield. The emission results from Table 6 show that the innovative design of pyrolysis with a gas cleaning process to treat the by-product gases (condensable gases/ non-condensable gases) will enable and ensure lower emissions than the current standard comparing to incineration. Particulate is approximately 73 times lower than the current standard where sulphur dioxide and others are confirmed to be lower than the accepted standard for testing in an emission

stack oven. Based on the study of literature, the pyrolysis process was chosen by most researchers because of its potential to convert the most energy from plastic waste to valuable liquid oil, gases and char. Therefore, it is a proper alternative waste disposal treatment for plastic and plastic-like waste in terms of emissions when compared to those of land filling and incineration. The flexibility that it provides in terms of by-product preference could be achieved by adjusting the parameters accordingly. The pyrolysis could be performed in both thermal and catalytic processes. However, the catalytic process provided a lower operating temperature with a greater yield of liquid oil for most plastics with the right catalyst selection but on the other hand it is also required a chemical additive to be used to eliminate within the final products. The sustainability of the process is unquestionable, since the amount of plastic-like waste such as NMF material and laminated packaging waste, which is available in every country, is reaching millions of tons each year. With the pyrolysis method, the waste management becomes more efficient, less capacity of landfill is needed, less pollution is created and it's very cost effective. Moreover, with the existence of the pyrolysis method as an alternative waste disposal treatment to recover plastic and other waste materials, including NMF material, into valuable energy fuel, the dependence on fossil fuel as a non-renewable energy source can be reduced and this option contributes to solving the rise in energy demand.

#### **Acknowledgments**

I would like to thank the Griffith University team, Dr Jimmy Yu, Dr Sunil Herat for the support and guidance through the experiments and study. I thank to Prof. Erik Bohez from Asian Institute of technology for supports. Also thank Viro Group Inc. for the building of the pyrolysis experimental unit and for financial assistance of this research, Clarence Valley Council for supporting the project in Grafton NSW and I am very much appreciated the helps and supports from Dr. Premrudee Kanchanapiya of Thailand National Metal and Material Technology Center (MTEC) for the supplying NMF material and laboratory testing.

## References

- [1] S. M. Alston and J. C. Arnold, “Environmental impact of pyrolysis of mixed WEEE plastics part 2: Life cycle assessment,” *Environmental Science and Technology*, vol. 45, no. 21, pp. 9386–9392, 2011.
- [2] S. M. Alston, A. D. Clark, J. C. Arnold, and B. K. Stein, “Environmental impact of pyrolysis of mixed WEEE plastics part 1: Experimental pyrolysis data,” *Environmental Science and Technology*, vol. 45, no. 21, pp. 9380–9385, 2011.
- [3] M.-H. Cho, S.-H. Jung, and J.-S. Kim, “Pyrolysis of mixed plastic wastes for the recovery of Benzene, Toluene, and Xylene (BTX) aromatics in a fluidized bed and chlorine removal by applying various additives,” *Energy and Fuels*, vol. 24, no. 2, pp. 1389–1395, 2010.
- [4] T. Fujita, H. Ono, G. Dodbiba, and K. Yamaguchic, “Evaluation of a recycling process for printed circuit board by physical separation and heat treatment,” *Waste Management*, vol. 34, no. 7, pp. 1264–1273, 2014.
- [5] J. Dong, Y. Chi, Y. Tang, M. Ni, A. Nzihou, E. Weiss-Hortala, and Q. Huang, “Partitioning of heavy metals in municipal solid waste pyrolysis, gasification, and incineratio,” *Energy and Fuels*, vol. 29, no. 11, pp. 7516–7525, 2015.
- [6] L. Zhang and Z. Xu, “Separating and recycling plastic, glass, and gallium from waste solar cell modules by nitrogen pyrolysis and vacuum decomposition,” *Environmental Science and Technology*, vol. 50, no. 17, pp. 9242–9250, 2016.
- [7] J. Robinson, C. Dodds, A. Stavrinides, S. Kingman, J. Katrib, Z. Wu, J. Medrano, and R. Overend, “Microwave pyrolysis of biomass: control of process parameters for high pyrolysis oil yields and enhanced oil quality,” *Energy and Fuels*, vol. 29, no. 3, pp. 1701–1709, 2015.
- [8] C. Ma, J. Yu, B. Wang, Z. Song, F. Zhou, J. Xiang, S. Hu, and L. Sun, “Influence of zeolites and mesoporous catalysts on catalytic pyrolysis of Brominated Acrylonitrile-Butadiene-Styrene (Br-ABS),” *Energy and Fuels*, vol. 30, no. 6, pp. 4635–4643, 2016.
- [9] T. V. Le Doan, N. W. Bostrom, A. K. Burnham, R. L. Kleinberg, A. E. Pomerantz, and P. Allix, “Green river oil shale pyrolysis: semi-open conditions,” *Energy and Fuels*, vol. 27, no. 11, pp. 6447–6459, 2013.
- [10] F. A. Agblevor, O. Mante, N. Abdoulmoumine, and R. McClung, “Production of stable biomass pyrolysis oils using fractional catalytic pyrolysis,” *Energy and Fuels*, vol. 24, no. 7, pp. 4087–4089, 2010.
- [11] U. Jena and K.C. Das, Comparative evaluation of thermochemical liquefaction and pyrolysis for bio-oil production from microalgae,” *Energy and Fuels*, vol. 25, no. 11, pp. 5472–5482, 2011.
- [12] W. Kaminsky, “Pyrolysis of polymers,” *Emerging Technologies in Plastics Recycling*, vol. 513, pp. 60–72, 1992.
- [13] C. Dorado, C. A. Mullen, and A. A. Boateng, Coprocessing of agricultural plastic waste and switchgrass via tail gas reactive pyrolysis,” *Industrial and Engineering Chemistry Research*, vol. 54, no. 40, pp. 9887–9893, 2015.
- [14] J. Hao, H. Wang, S. Chen, B. Cai, L. Ge, and W. Xia, “Pyrolysis characteristics of the mixture of printed circuit board scraps and coal powder,” *Waste Management*, vol. 34, no. 10, pp. 1763–1769, 2014.
- [15] M. J. Workman, A. Serov, B. Halevi, P. Atanassov, and K. Artyushkova, “Application of the discrete wavelet transform to sem and afm micrographs for quantitative analysis of complex surfaces,” *Langmuir*, vol. 31, no. 17, pp. 4924–4933, 2015.
- [16] J. Sabo, G. Judd, and S. Ferriss, “Examples of sem analyses in forensic evidence applications,” *Forensic Science*, vol. 13, pp. 75–82, 1975.
- [17] M. Uchimiya, L. H. Wartelle, K. T. Klasson, C. A. Fortier, and I. M. Lima, “Influence of pyrolysis temperature on biochar property and function as a heavy metal sorbent in soil,” *Journal of Agricultural and Food Chemistry*, vol. 59, no. 6, pp. 2501–2510, 2011.
- [18] N. K. Sundararajan and A. R. Bhagavathi, “Experimental investigation on thermocatalytic pyrolysis of HDPE plastic waste and the effects of its liquid yield over the performance, emission, and combustion characteristics of CI engine,” *Energy and Fuels*, vol. 30, no. 7, pp. 5379–5390, 2016.
- [19] S. Du, J. A. Valla, R. S. Parnas, and G. M. Bollas,

- "Conversion of polyethylene terephthalate based waste carpet to benzene-rich oils through thermal, catalytic, and catalytic steam pyrolysis," *ACS Sustainable Chemistry and Engineering*, vol. 4, no. 5, pp. 2852–2860, 2016.
- [20] H. L. Chiang, K. H. Lin, M. H. Lai, C. T. Chen, and s. Y. Ma, "Pyrolysis characteristics of integrated circuit boards at various particle sizes and temperatures," *Journal of Hazardous Materials*, vol. 149, no. 1, pp. 151–159, 2007.
- [21] H. Ben and A. J. Ragauskas, "Pyrolysis of kraft lignin with additives," *Energy and Fuels*, vol. 25, no. 10, pp. 4662–4668, 2011.
- [22] C.-C. Chang, S.-R. Wu, C.-C. Lin, H.-P. Wan, and H.-T. Lee, "Fast pyrolysis of biomass in pyrolysis gas: Fractionation of pyrolysis vapors using a spray of bio-oil," *Energy and Fuels*, vol. 26, no. 5, pp. 2962–2967, 2012.
- [23] E. V. Antonakou, K. G. Kalogiannis, S. D. Stephanidis, K. S. Triantafyllidis, A. A. Lappas, and D. S. Achilias, "Pyrolysis and catalytic pyrolysis as a recycling method of waste CDs originating from polycarbonate and HIPS," *Waste Management*, vol. 34, no. 12, pp. 2487–2493, 2014.
- [24] T. Mani, P. Murugan, and N. Mahinpey, "Pyrolysis of oat straw and the comparison of the product yield to wheat and flax straw pyrolysis," *Energy and Fuels*, vol. 25, no. 7, pp. 2803–2807, 2011.
- [25] M. D. Kass, C. J. Janke, R. M. Connatser, S. A. Lewis Sr., J. R. Keiser, and K. Gaston, "Compatibility assessment of fuel system elastomers with bio-oil and diesel fuel," *Energy and Fuels*, vol. 30, no. 8, pp. 6486–6494, 2016.
- [26] C. Dorado, C. A. Mullen, and A. A. Boateng, "H-ZSM5 catalyzed co-pyrolysis of biomass and plastics," *ACS Sustainable Chemistry and Engineering*, vol. 2, no. 2, pp. 301–311, 2014.
- [27] P. C. Tarves, C. A. Mullen, and A. A. Boateng, "Effects of various reactive gas atmospheres on the properties of bio-oils produced using microwave pyrolysis," *ACS Sustainable Chemistry and Engineering*, vol. 4, no. 3, pp. 930–936, 2016.
- [28] V. Srinivasan, S. Adhikari, S. A. Chattanathan, and S. Park, "Catalytic pyrolysis of torrefied biomass for hydrocarbons production," *Energy and Fuels*, vol. 26, no. 12, pp. 7347–7353, 2012.
- [29] P. A. Jensen, F. J. Frandsen, J. Hansen, K. Dam-Johansen, N. Henriksen, and S. Horlyck, "SEM investigation of superheater deposits from biomass-fired boilers," *Energy and Fuels*, vol. 18, pp. 378–384, 2004.
- [30] U. R. Gracida-Alvarez, L. M. Keenan, J. C. Sacramento-Rivero, and D. R. Shonnard, "Resource and greenhouse gas assessments of the thermochemical conversion of municipal solid waste in Mexico," *ACS Sustainable Chemistry and Engineering*, vol. 4, no. 11, pp. 5972–5978, 2016.