Jurnal Kejuruteraan 34(6) 2022: 1199-1207
https://doi.org/10.17576/jkukm-2022-34(6)-20

Comparison of Physical and Chemical Method for Removing Copper from Non-Metallic Printed Circuit Board Scrap

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Received 6 January 2022, Received in revised form 12 May 2022
Accepted 17 June 2022, Available online 30 November 2022

ABSTRACT

Printed circuit boards (PCBs) are the basic components of electrical and electronic devices. E-waste management is challenging to implement due to accompanying difficulties and danger. Valuable metals and copper (Cu) are primarily recycled through various methods in the treatment of waste PCB. A large number of non-metals materials in PCBs are disposed of through combustion or in landfill, resulting in secondary pollution and resource waste. To reduce the amount of waste non-metallic PCB (NMPCB) and the influences toward environment, recent studies focused on the usage of NMPCB as filler to replace raw material. NMPCB as filler seems to have a good interaction with the raw materials and thus can enhance the strength of the newly formed product. In our study, we focused on developing new NMPCB added with waste NMPCB. Commercial NMPCBs are often a flat laminated composite comprising non-conductive substrate materials. Hence, pre-treatment methods to remove metals, especially Cu, must be investigated. The present study attempted to remove the Cu layer on PCB by using chemical and physical methods. The untreated and Cu removed PCB residues were characterized using X-ray diffraction (XRD), scanning electron microscopy, and infrared spectroscopy (FTIR) for the determination of structural and functional groups and hydrophobicity test. XRD analysis indicated that the Cu in untreated PCB was successfully removed using physical and chemical methods.

Keywords: E-Waste; Recycling; Printed circuit board; Non-metallic; Copper etching

INTRODUCTION

The rapid advancement of electrical and electronic equipment (EEE) technology has led to the production of e-waste as the world's fastest-growing waste sources and the expansion of the printed circuit board (PCB) manufacturing industry (Hadi et al. 2014). According to Balde et al. (2017), EEE production has shown a steady growth pattern and contributed to the annual increase in waste. Approximately 44.7 million metric tons of e-waste are generated annually, but only 20% is collected and recycled. The United Nations (UN) reported that the global e-waste production recently increased by a 10% annual rate and exceeded 50 million tons (Kaya, 2016). However, according to Qiu et al. (2020), the amount of e-waste produced is increasing at a rate three times that of municipal wastes.

The PCB is a vital part of every electronic and electrical equipment and holds a variety of precious metals and minerals and other compounds that are hazardous to human and environmental health (Karwowska et al. 2014; Ilyas and Lee, 2014; Willner et al. 2015). PCB contains a number of hazardous materials, such as brominated flame retardants and other heavy metals, which are highly dangerous to the environment if not properly handled. Waste (WPCB) accounts for a huge percentage of more than 40% value of metal from e-waste (Golev et al. 2016). PCB, which includes valuable metals, account for around 6 wt% of total e-waste (Evangelopoulos et al. 2015). Metal's enormous economic value attracts attention. As a result, chemical leaching and physical separation procedures are utilized in succession to recycle metals from WPCB. Compared with that the 0.6% copper, 215 ppm silver, 1 ppm gold, and 2.7 ppm palladium (by mass) in an average mine, WPCB's metal concentration is 20% copper, 1000 ppm silver, 250 ppm gold, and 110 ppm palladium (Namias, 2013; Desjardins, 2014; Kumar et al. 2017). Table 1 shows the content of the metals in WPCB reported by several researchers.

TABLE 1. Metal content in waste printed circuit board (WPCB)

Metal (%)	Goosey and Kellner (2002)	Sum (2005)	Park and Fray (2009)	Yang et al. (2009)	Guo et al. (2009)	Akcil et al. (2015)	Akcil et al. (2015)
Copper	16	20	16.0	25.06	26.8	34.49	20.19
Aluminium	-	-	5.0	-	4.7	0.26	0.16
Gold	0.03	0.1	0.025	-	80gt^{-1}	-	0.13
Iron	3	8	5.0	0.66	1.5.3	10.57	7.33
Lead	-	2	2.0	0.8	-	1.87	5.53
Nickel	2	2	1.0	0.0024	0.47	2.63	0.43
Palladium	0.01	0.005	0.01	-	-	-	-
Silver	0.05	0.2	0.1	-	3300gt^{-1}	0.21	0.16
Tin	-	4	3.0	-	1.0	3.39	8.83
Zinc	-	1	1.0	0.04	1.5	5.92	4.48

Nonmetals, which account for 70% of the total WPCB, are also manufactured in large amounts (Huang et al. 2014); however, only about 10% is recycled. The Environmental Protection Agency (2012) reported that the recovered non-metallic PCB (NMPCB) from waste PCB is unlikely recycled and up to 94% of waste is thrown away in landfills and causes secondary pollution. Owing to their poor value and hazardous compounds, NMPCBs are generally impractical to be properly handled and recovered (Kaya, 2019). Incineration and landfilling are not the best ways to get rid of waste NMPCB because of the release of a large number of carcinogens (Kumar et al. 2018). To reduce the impact of NMPCB toward environment, most researchers proposed the direct use of NMPCB as secondary replacement substances, such as filler for building materials, paint, decorating agent, adhesives, insulating materials, or non-metallic plate for the production of composite boards (Sohaili et al. 2012).

Thermosetting epoxy resin and fiberglass in NMPCB can be removed from their solvents and cured to be used as fillers in new inorganic and composite materials, such as wood, concrete, asphalt, plastic, and other similar materials (Xie et al. 2014; Guo et al. 2009; Sun et al. 2014; Zheng et al. 2009; Sun et al. 2015). Considering that NMPCB addition as asphalted fillers enhances the composite's elasticity and stiffness, Xin et al. (2017) investigated the rheological properties, thermal sensitivity, fracture toughness, and structure of PCBs and showed that the addition of NMPCB may improve the performance of asphalt at different temperatures by increasing the asphalt modified's fatigue resistance. Gao et al. (2017) investigated epoxy resins and fiber/epoxy composites as alternative NPRP fillers. Mechanical testing revealed a considerable improvement in the composites' flexural and impact properties when modified NPRPs were used. Kakria et al. (2020) demonstrated the uses of NMF to replace fly ash and metakaolin in a geopolymer mortar and found that NMPCB enhances the compressive strength of the geopolymer mortar. Yang et al. (2020) discussed the manufacturing of wood composites reinforced with NMPCB and found that the addition increased the thermal stability of the wood composite and provided an attractive structure that increased the composite's tensile strength. A NMPCB is recognized as acceptable for recovery as a polyvinyl chloride reinforcement filler for cable isolation due to the high content of glass fibers and the robust epoxy matrix. The homogeneity of composites at low filler doses increases or remains constant compared with the polymer matrix.

Considering the rapid growth of EEE production, we proposed the usage of waste NMPCB as filler in new non-metallic PCBs. PCBs are often a flat laminated composite comprising non-conductive substrate materials with layers of copper circuitry concealed within or on the exterior surfaces. The removal of metals, mainly copper, must be conducted before waste NMPCB can be used as filler and develop a new non-metallic PCB that is insulated toward electricity. A Imthiyas et al. (2020) also supported this statement and reported that the PCB base should have a non-conductive substrate. In this work, the copper in non-metallic PCB was removed so that it can be used as filler in new NMPCB. Recycling is generally performed using two methods: physical and chemical. Physical techniques, such as density-based separation, electrostatic separation (Suponik et al. 2021; Franke et al. 2020; Hamerski et al. 2018), and magnetic and gravity separation (Zhu et al. 2020), have been well studied. Numerous investigations were conducted on copper leaching from waste PCBs by chemical treatment using a variety of lixiviants, such as inorganic acids (Yazici and Deveci. 2013), organic acids (Lisinska et al. 2022; Krishnamoorthy et al. 2021), and ionic liquids (Chen et al. 2015; Zhang et al. 2018; Barrueto et al. 2021). The removal of copper using microorganisms such as bacteria and fungi has also been reported (Xia et al. 2018; Awasthi et al. 2019; Dave et al. 2020; Arshadi et al. 2020), particularly in removing Cu from PCBs. According to Veloso et al. (2016) and Nikoloski et al. (2017), Fe(III) salts are an effective leachant of Cu from a variety of minerals (Sethurajan and van Hullebusch. 2019). The differences between physical and chemical recycling methods are discussed in Table 2..

TABLE 2. Differences between physical and chemical recycling methods

Recycling method	Physical	Chemical
Advantages	1. It is feasible to recycle a large volume of e-waste in a short period of time. 2. Minimizing waste for final disposal and minimizing landfill costs.	1. Reducing the overall quantity of garbage that must be disposed of. 2. Extracting precious and valuable metals from WPCB is simple.
Disadvantages	1. Expensive due to the requirement for expenditure and the problems associated with metal separation (impurities). 2. Pollution caused by dust, excessive metal exposure to the surrounding environment, and a risk to human health.	1. Chemical reagents in large quantities are required. 2. Difficult to operate and dangerous to handle. 3. Probability of secondary pollution is highest.
Impacts	1. Pollutant from metal exposure that is either direct or indirect can harm the environment and people's health. 2. Environmental contamination was a significant impediment to industrial progress.	1. Most of non-metallic components are disposed of in landfills, incinerators, or open dumping, posing a potential environmental threat and resulting in significant resource loss 2. Significant source of environmental pollution, particularly for soil and water.

On the basis of previous studies on the highest copper content in PCB, two different techniques of simple physical and chemical removal of copper were selected as the focus of the present study.

METHODOLOGY

MATERIALS

NMPCB was collected from local waste electrical and electronic equipment disposal company in Melaka, Victory Recovery Resources Sdn Bhd, Malaysia and then used for the removal of copper using chemical and physical processes.

REMOVAL OF COPPER LAYER CHEMICAL PROCESS

Leaching was conducted in an etching machine, and ferric chloride was used as the lixiviant. The treatment was performed at 40 °C for about 10 minutes to remove the copper layer. The samples were rinsed with distilled water to ensure no copper residue was left on the NMPCB and dried at room temperature. The samples were labelled as chemical recycling.

PHYSICAL PROCESS

Sanding using a 150–180 grit sand paper was conducted to remove the copper layer. Once the copper layer was removed, the samples were rinsed with distilled water to ensure no copper residue was left on the NMPCB and dried at room temperature. The samples were labelled as physical recycling. Figure 1 shows the image of NMPCB before and after the removal of copper.

SEM-EDX ANALYSIS

Scanning electron microscopy (SEM) was performed using ZEISS SIGMA VP to study the morphology of the surface. XRD analysisX-ray diffraction (XRD) scattering pattern of NMPCB was investigated using an X-ray diffractometer Bruker X-Ray Diffractometer model D8 ADVANCE with high-intensity Cu-Kα radiation ($\lambda = 0.754$ nm) in the 2θ range from 0° to 90°.

FT-IR ANALYSIS

Fourier transform infrared (FTIR) spectrum in the range 4000–400 cm^{-1} was analyzed using a Nicolet 6700 USA FTIR spectrometer. A small piece of NMPCB was analyzed in the FTIR spectrometer to determine the functional groups present in the samples.

HYDROPHOBICITY TEST

Drop shape analysis was performed using Drop Shape Analyzer-DSA100. The contact angle obtained indicates whether the surface is hydrophobic or hydrophilic. When the contact angle between two surfaces reaches 90, the surface is hydrophobic.

RESULTS AND DISCUSSION

SEM-EDX ANALYSIS FOR NMPCB

The morphology study of NMPCB was carried out to find out the dispersion of copper on the surface. Figure 2 (a) denotes that the untreated NMPCB had a smooth and even surface with copper scattered on the surface. Figure 2 (b) and (c) shows the image of NMPCB after the removal of copper. The smooth surface of NMPCB changed to a rough surface with formed void spaces. This structural changed confirm that copper was removed from the NMPCB. Zhao et al. (2008) also reported structural changes in NMPCB after bioleaching. The copper content in the untreated NMPCB was 90 wt%. On the basis of the EDX results, copper was successfully removed using chemical and physical techniques.

XRD ANALYSIS FOR NMPCB

The XRD pattern of the NMPCB samples is shown in Figure 3. XRD primarily aims to identify the crystallographic characteristics of samples. Polymers often have a semi-crystalline crystallographic structure, which results in a broad peak in the spectra. Yao et al. (2015) detected Ca elements in PCB boards in the range of 10°–30° and noticed two peaks across the XRD patterns. In the present research, a broad diffraction peak was found between 10 ° and 25° and was associated with the presence of a polymer matrix. The present findings of were similar to the reported of Das et al. (2019). Meanwhile, the sharp and strong peaks in XRD spectra corresponded to metals. Three diffraction peaks at 42°, 50°, and 75° were attributed to the presence of copper in the untreated PCB. After physical and chemical removal treatments, copper was totally removed. The absence of sharp and intense peaks indicated that the metallic particles were totally eliminated from the untreated PCB (Luo et al. 2015).

FT-IR ANALYSIS FOR NMPCB

The FTIR spectra of all samples are represented in Figure 4. The FTIR wavenumber patterns of each sample were quite similar to each other. The broad absorption peak of NMPCB around 3200–3500 cm^{-1} was related to O-H stretching vibration band. The two peaks between 2850 and 28940 cm^{-1} can be associated with the C-H stretching vibration in all NMPCBs. Furthermore, the peaks at 1727, 1716, and 1735 cm^{-1} were attributed to C=O; the peaks at 1478, 1468, and 1481 cm^{-1} were attributed to C=C/CN; the peaks at 1101, 1027, and 1101 cm^{-1} were attributed to RCO–OH stretching vibration; and the peaks at 667, 551, and 555 cm^{-1} were attributed to C–H wagging vibrations. These peaks also complemented the brominated bisphenol in the epoxy resin found in PCB type FR4 (Zhao et al. 2017 & Xiong et al. 2020). Das et al. (2019) and Narayanasamy et al. (2018) also reported similar results.

On the basis of the XRD results, ferric sulphate can leach out Cu from PCB. Isldar et al. (2016) conducted an XRD study of powdered PCBs and discovered that the primary mineral in PCBs is metallic Cu. During chemical removal, ferric chloride can react with copper to generate copper (II) ion, Cu^{2+}, and release it to the liquid phase. The Cu leaching from PCBs by ferric sulphate can be described by Equation (1).

$$Cu + FeCl_3 \rightarrow CuCl_2 + Fe \qquad (1)$$

HYDROPHOBICITY TEST

Hydrophobicity refers to the ability of a solid insulator surface to prevent the creation of a continuous film of water and shows that the isolation fault caused by electrochemical migration on PCBs may be resisted. Numerous studies were conducted on the hydrophobicity and weight change of isolated materials produced by soaking in distilled and saltwater solutions at various temperatures. Motoyama and Hackam (2001) reported that the hydrophobicity of NMPCB is decreased upon the removal of the laminating copper layer. Figure 5 shows the drop shape analysis of NMPCB. The hydrophobicity properties of NMPCB did not change even after undergoing physical and chemical treatments. The purpose of the physical and chemical removal of copper in NMPCB is to use the recycled NMPCB as filler. On the basis of the results, the hydrophobicity of NMPCB seems unchanged. Therefore, waste NMPCB is usable as filler. Further study on its properties must be conducted.

CONCLUSION

This study showed that NMPCB can be used as filler in most insulated component, especially in EEEs. Chemical and physical recycling methods showed similar results toward the removal of copper on NMPCB, but the chemical method is the most effective because it removed other metals in the NMPCB. Findings on the morphological surface structure also confirmed that metals, especially copper, were almost completely removed. Chemical method extracted the copper in the small space, and physical method only removed the metal on the surface. This result proved that most of the metals, especially copper, were removed from NMPCB using both methods. Hence, the treated waste NMPCB can be used as filler. Its performance and interaction with the other raw materials must be further studied. As a recommendation, various concentrations of ferric chloride and NMPCB should be crush to small size for further improvement of this research.

ACKNOWLEDGEMENT

The authors were grateful and appreciate the Ministry of Higher Education for the Fundamental Research Grant Scheme (FRGS/1/2018/TK05/UKM/02/4) for the financial support throughout this study.

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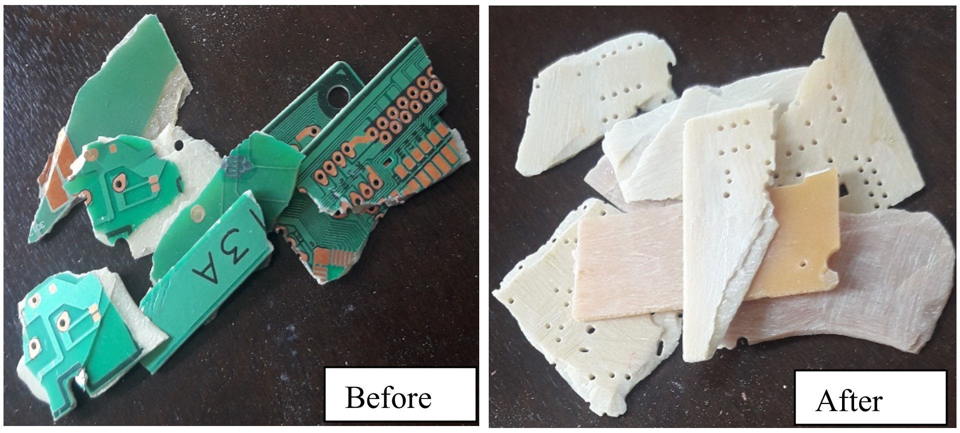

FIGURE 1 Image of NMPCB before and after the removal of copper layer

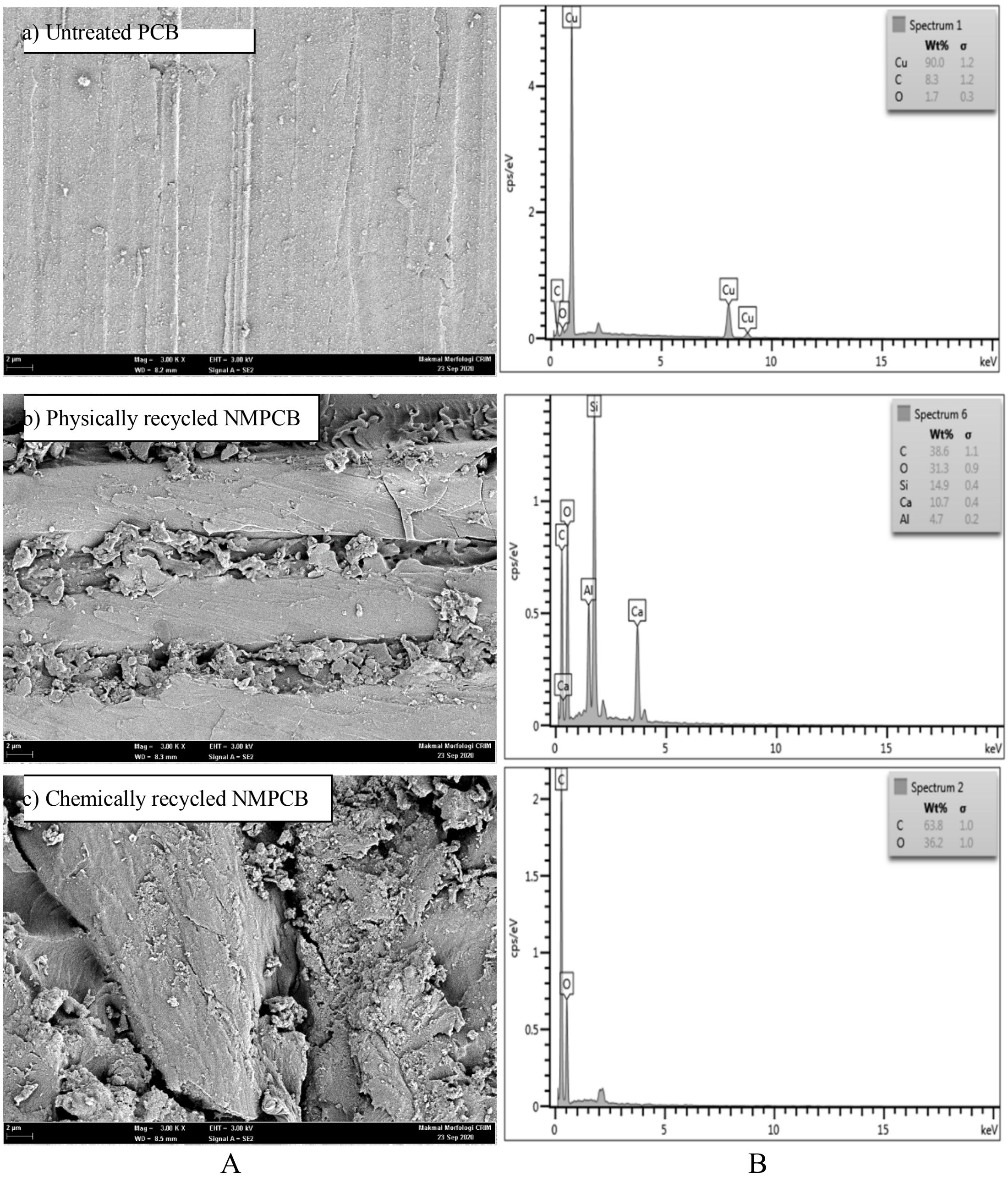

FIGURE 2 Images for A) SEM and B) EDX for a) untreated NMPCB, b) physically recycled NMPCB, and c) chemically recycled NMPCB

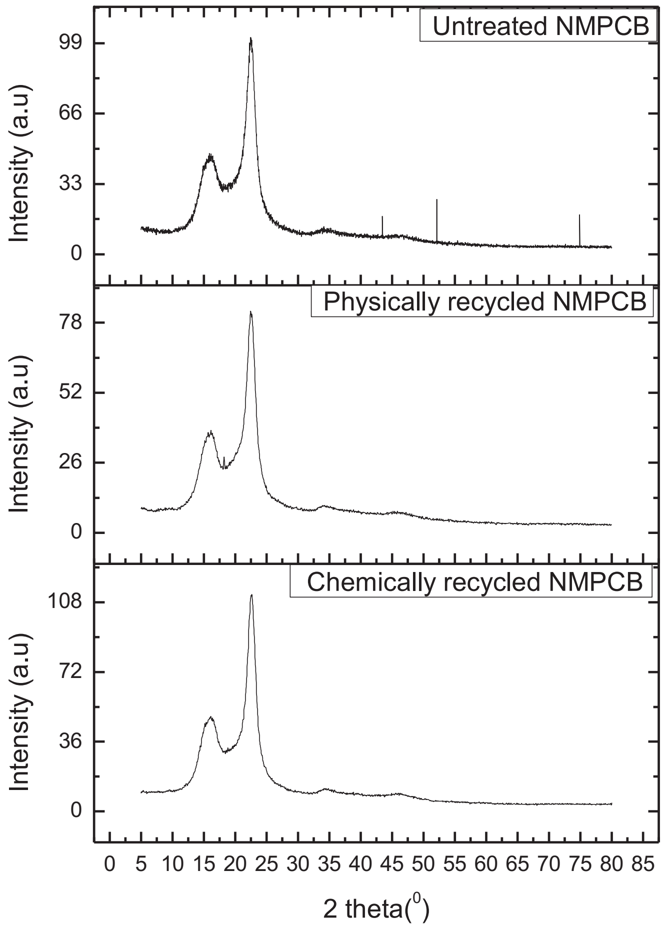

FIGURE 3 XRD pattern of NMPCB

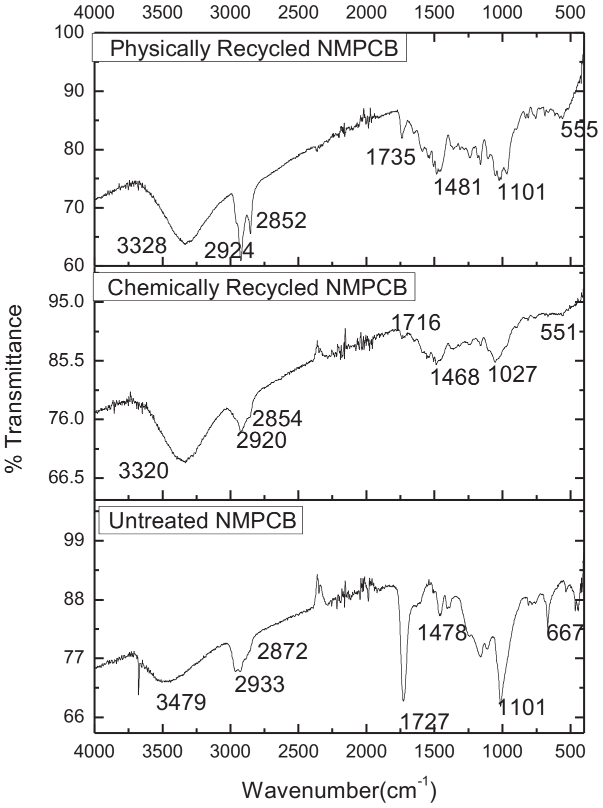

FIGURE 4 FTIR pattern of NMPCB

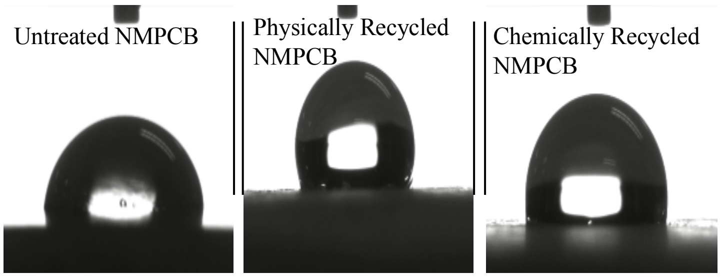

FIGURE 5 Drop shape analysis of untreated, physically recycled, and chemically recycled NMPCBs