Tetrapak waste treatment using microwave pyrolysis to produce alternative gas fuels

Ilham Ayu Putri Pratiwi*, Harwin Saptoadi¹, Jayan Sentanuhady¹, Chandra Wahyu Purnomo², Tri Agung Rohmat¹

¹ Mechanical and Industrial Engineering Department, Faculty of Engineering, Universitas Gadjah Mada, Jln Grafika No 2, Yogyakarta, Indonesia
² Chemical Engineering Department, Faculty of Engineering, Universitas Gadjah Mada, Jln. Grafika No 2, Yogyakarta Indonesia

ABSTRACT

Tetrapak waste is one of the most abundant wastes in the world. It consists of layers of paper, LDPE, and aluminium foil. It is difficult to recycle, however it can be converted into valuable gaseous fuel if processed by microwave heat using a dual reactor catalytic process, SiC as microwave absorber, and natural zeolite as catalyst. The sample was pyrolyzed at different Microwave Output Power (MOP) levels (300, 450, 600, and 800 W) and Catalytic Temperatures (CT) levels (350, 400, 450, and 500°C) for 30 minutes. The results showed that the MOP and CT levels positively affect the increase of the yield and improve the heating value of the gas product. For instance, at 800 W, the gas yield increased from 73 wt.% in the process without catalyst to 78 wt.% in the catalytic temperature of 500°C. The same effect occurred when various MOP levels were used. The gas yield increased when the MOP level raised. H₂, CH₄, CO, CO₂, and other hydrocarbons with low molecular weights (C₂⁺) are the major components of gas products. The presence of catalyst, an increase in CT, and a rise in MOP all boosted the generation of syngas (H₂ + CO) up to 54.55 wt.%. The gas yields from this experiment have a LHV of 21.97–23.46 MJ/m³ and total energy of 34.59–171.24 kJ. The high-quality gaseous products can be used as alternative fuels or feed gas for chemical synthesis.

Keywords: Microwave pyrolysis, Microwave output power, Natural zeolite, Silicon carbide (SiC), Tetrapak waste.

1. INTRODUCTION

Aseptic packaging is produced mostly by Tetra Pak. This packaging is used for many foods and beverages. Tetrapak packaging keeps nutrients and long shelf lives of foods and beverages without the use of preservatives and coolers. This feature can be obtained from the combination of several material layers to form Tetra Pak, i.e. paper (74%), low-density polyethylene (21%), and aluminium foil (5%).

According to the Tetra Pak Sustainability Report 2021, more than 183 billion units of aseptic packaging (tetrapak) were sold worldwide in 2020. However, only 49 billion tons of used Tetra Pak were effectively recycled (Tetra Pak, 2021). Thus, there are still many opportunities in utilizing this material into various products, such as fuel.

Pyrolysis is a promising waste to energy (WTE) method that can be used to reuse tetrapak waste (TW) (Ma, 2018). The material is heated in the absence of air during the pyrolysis process. The products of pyrolysis are liquid oil, char, and gases (Miandad et al., 2016). Several factors including temperature, size of the particle, feedstock, heating rate, residence time, type of catalyst, temperature of catalyst, and atmospheric gas will have impacts on the quality and quantity of product yields (Atreya et al., 2017; Gómez et al., 2018; Klingler et al., 2018; Safdari et al., 2019). Recently, microwaves are widely used as a heat generator in the pyrolysis process (namely, microwave pyrolysis).
(Shepherd et al., 2018; Zhang et al., 2017). Microwave pyrolysis (MWP) is pyrolysis that involves microwave dielectric heating in the absence of oxygen. MWP is used because of its ability to create fast, selective, and uniform heating (Motasemi and Afzal, 2013). Furthermore, MWP will increase product yields while requiring less reaction time and lower temperature (Rex et al., 2020). Zhao et al. (2011) found that microwave pyrolysis of corn straw has gas yield with the composition of H₂ (35.2 vol.%), CO (18.4 vol.%), CH₄ (23.0 vol.%), CO₂ (18.4 vol.%), C₂H₆ (0.83 vol.%), and C₃H₈ (1.1 vol.%). Many pyrolysis researchers have studied microwave as a heating source for feedstocks, such as coffee hulls (Domínguez et al., 2007), polystyrene and polypropylene mixtures (Rex et al., 2020), sewage sludge (Zuo et al., 2011), Tallow kernel oil (Yu et al., 2018), and oil palm shell (Mushtaq et al., 2015). Domínguez et al. (2007) compared the pyrolysis process using microwave and conventional heating for coffee hulls and found that MWP generated more gas and less oil. However, the gas obtained had a higher H₂ concentration and lower CO₂ concentration. Microwave heating, as compared to conventional heating, directly transfers the microwave energy necessary for reactions to the reacting atoms or molecules via dielectric properties within the material (Li et al., 2019a). In MWP processes, dielectric properties play a significant role in determining the heating characteristics of materials (Li et al., 2019b).

The TW can be treated by microwave-assisted pyrolysis. However, it has weak dielectric properties. Thus, it is necessary to have microwave absorber (MWA) that can absorb microwave energy properly to increase the heating process (Zhang et al., 2017). The presence of MWA improves the process heating rate as well as product selectivity (Mushtaq et al., 2015). Significant effects of the MWA on the pyrolysis products have been found in the literature by Zuo et al. (2011).

Catalyst has an important role in the pyrolysis process, either using conventional or microwave heating. The main objective of catalytic pyrolysis is to remove oxygenated molecules while also to break high molecular weight products into lower chain compounds (Pattiya, 2018). On the other hand, the catalyst enhances the quality and yield of pyrolysis products while also increasing process efficiency by optimizing the targeted reactions, lowering temperature and reaction time (Zaker et al., 2019). The acidity, pore size, pre-treatment, and calcination of the catalyst are the primary factors that impact the pyrolysis process (Pattiya, 2018). A large surface area and acidity catalyst, or microporous catalyst, may produce more gases and decrease the liquid oil yield. Meanwhile, macroporous catalyst with a large volume has little effect on the quantity of the liquid yield because it generates long carbon chain molecules (Miandad et al., 2016). Zeolites, particularly natural zeolites (NZ), are one of the most utilized catalysts in the pyrolysis process. Natural zeolite was employed in the study by Miandad et al. (2019). In this study natural zeolite used as catalyst for the pyrolysis of different types of plastic waste (PS, PE, PP, and PET) as single or combined in various ratios, in a batch reactor at 450°C. It was found that the majority of the liquid oil produced a high aromatic content, along with some aliphatic and other hydrocarbon components. The pyrolysis of PS produced the maximum production of pyrolysis oil (54 wt.%), whereas PP produced the lowest yield of pyrolysis oil (14 wt.%). The mixture produced 22 wt.% pyrolysis oil. Furthermore, the high heating value was in the range of 41.7–44.1 MJ/kg. All of the results in this study suggested that natural zeolite has the ability to increase the pyrolysis rate and enhance the characteristics of the pyrolysis product. NZ might be utilized to replace the commercial catalyst (e.g. commercial ZSM-5 zeolite used in the pyrolysis of plastic waste using a semi-batch reactor at 440°C for 30 min (López et al., 2011). NZ is available all around the world, especially in Indonesia. Indonesian NZ deposits are extensive and of excellent quality, with a silica concentration of approximately 60%. Several zeolite mines areas are Bayah, Cikembar (West Java), Nangapanda, Ende (NTT), Malang (East Java), Gunung Kidul (Yogyakarta), and Klaten (Central Java) (Wahono et al., 2014).

The novelties in the present study are utilizations of double reactor catalytic process of TW, NZ as catalyst, and SiC as absorber. There have been no investigations on microwave-assisted pyrolysis at constant temperatures utilizing natural zeolite as catalyst in the secondary reactor, which uses SiC as absorber. The effect of microwave output power (MOP) and catalytic temperature (CT) on the MWP of TW are investigated to determine the quality and quantity of pyrolysis products.

2. MATERIALS AND METHODS

2.1 Materials

2.1.1 Tetrapak Waste

The feedstock used was aseptic packaging waste, produced by the Tetra Pak company. TW samples were collected from household garbage dumps. From the inside out, they are arranged in six ordered layers: LDPE, LDPE, aluminium foil, LDPE, paper, and LDPE. Based on the proximate analysis, the composition of TW consists of moisture (10.13 wt.%), fixed carbon (3.7 wt.%), volatile matter (73.47 wt.%), ash (7.38 wt.%), and aluminium foil (5.3 wt.%). The TW samples were washed to remove impurities, the cut about 5 mm × 5 mm. The samples were dried using a conventional oven at 105°C for 2 h. Subsequently, the samples were put on the glass reactor in the microwave oven as shown in Fig. 1(a) number 5.

2.1.2 Microwave Absorber

This research utilized silicon carbide (SiC), which is available in the market, as absorber. The SiC were purchased from TAKA Engineering, Indonesia. The purity, melting point, bulk density, and molar mass of SiC is 99%,
2600°C, 1.38 g/cm³, and 40.1 g/mol, respectively. Before being placed in the MWP reactor, SiC was crushed in a rotary speed mill and sieved to get samples in the 40–80 mesh range. Afterward, the SiC was oven-dried for 2 h at 105°C.

2.1.3 Catalyst
In this study, NZ was used as catalyst in the MWP process. NZ was obtained from the Klaten District of Central Java, Indonesia. Based on the SEM-EDX analysis, the composition of NZ consist of Si (26.19 wt.%), Al (8.73 wt.%), and O (65.08 wt.%). It has a BET surface area of 17.93 m²/g. Each experiment utilized 50 g of NZ. Before usage, the catalyst was pulverized into particles ranging in size from 40 to 80 mesh. The catalyst was then washed and dried in an oven for 2 h at 105°C for activation.

2.2 Experimental Apparatus and Procedures
2.2.1 Microwave Pyrolysis
In the MWP experiments, an Electrolux microwave oven EMM2308X model with 1300 W electrical input power, and a frequency 2.45 GHz was utilized. The MOP variations utilized were determined by the microwave oven settings (300, 450, 600, and 800 W). The CT variations used were based on researches conducted by Onwudili et al. (2019), Sonawane YB, (2015), Syamsiro et al. (2014a), and Williams et al. (2002). They showed that the ideal temperature for NZ catalyst were 350–500°C. Fifty grams of TW were combined with 25 g of SiC. The sample was placed in a 1000 mL glass reactor. Catalyst particles are placed in a tube made from stainless steel with a diameter of 25 cm, and heat is generated using electric resistance heater. The schematic system diagram is displayed in Fig. 1(a). To eliminate the air before the pyrolysis process began, nitrogen gas was injected into the main reactor at a rate of 100 mL/min.

The catalytic temperature is switched to the desired temperature level. After the temperature has reached the required value and the primary reactor has reached an inert condition, the MOP is switched to the required MOP level and held for 30 min. The MWP was carried out from ambient temperature to 450°C in the microwave reactor.

The secondary reactor (Fig. 1(a) number 6) is the place of catalytic degradation of volatile matter from primary reactor. The volatile matter which has interacted with the catalyst flows into the condenser (Fig. 1(a) number 9). In the pyrex reactor, char products and aluminum foil were left. In a sample container (Fig. 1(a) number 10) the condensed product, namely liquid and wax, were collected. Meanwhile, non-condensable gas products were collected in Tedlar bags (Fig. 1(a) number 11). To ensure data accuracy, each MWP set of tests was carried out at least twice. The results presented are the average of each variation. The findings are fairly consistent between batches, and the relative error is less than 5%.
2.2.2 Analysis Method of Gas Products

The gas products were analyzed using a Shimadzu model QP2010 SE Gas Chromatography-Mass Spectrometry (GC-MS). This tool uses FI detectors. Meanwhile, the columns used were Rtx-5MS (30 m in length and 0.25 mm in inner diameter), column coated with a 0.25 μm thick film of 5% diphenyl-95% dimethyl polysiloxane. Helium (He) is used as the carrier gas, and its pressure is kept constant at 73 kPa. The oven was preheated to 50°C for 2 min before being raised to 290°C at a rate of 5°C/min. The GCMS analysis was repeated once or twice to ensure data accuracy. The transfer line temperatures were held at 230 °C and 300°C, raised to 290°C at a rate of 5°C/min. The ion source and the gas yields:

\[(vi) (\text{Dai et al., 2000})\]:

\[\text{CO, H}_2, \text{CH}_4, \text{and HC}_x, \text{respectively.}\]

The products of pyrolysis were determined using Equation (i)-(v) (Onwudili et al., 2019):

\[\text{Aq. phase yield (wt. %)} = \frac{\text{mass of aq-phase produced}}{\text{mass of TW}} \times 100\% \quad (i)\]

\[\text{Wax yield (wt. %)} = \frac{\text{mass of wax produced}}{\text{mass of TW}} \times 100\% \quad (ii)\]

\[\text{Wax yield (wt. %)} = \frac{\text{mass of wax produced}}{\text{mass of TW}} \times 100\% \quad (iii)\]

\[\text{Al. foil yield (wt. %)} = \frac{\text{mass of al. foil produced}}{\text{mass of TW}} \times 100\% \quad (iv)\]

\[\text{Gas yield (wt. %)} = 100\% - \text{Aq. phase yield} - \text{Wax yield} - \text{Char yield} - \text{Al. foil yield} \quad (v)\]

The LHV of gas yields can be determined using Equation (vi) (Dai et al., 2000):

\[\text{LHV (kJ/m}^3\) = [(30.0 \times \text{CO}) + (25.7 \times \text{H}_2) + (85.4 \times \text{CH}_4) + (151.2 \times \text{HC}_x)] \times 4.2 \quad (vi)\]

The volume percentages of CO, H₂, CH₄, and other hydrocarbons (C₂+) in the total gas product are denoted by CO, H₂, CH₄, and HCₙ, respectively.

Equation (vii) was used to determine the total energy of the gas yields:

\[\text{Total Energy (kJ)} = \text{LHV} \times \text{volume of the gas products} \quad (vii)\]

Gases have an indefinite shape and volume. On the other hand, the gas generated in the MWP of TW is dominated by gases that have relatively small molecules (H₂ and CO). As a result, the volume of gas is estimated using a mass balance of the gas from the ideal gas law (Onwudili et al., 2019). In thermodynamics theory, the ideal gas law can be used at high temperature processes with good accuracy regardless of pressure. In the current study, the process temperature reached 500°C.

3. RESULTS AND DISCUSSION

3.1 Temperature and Mass Profile

Microwave-assisted heating of feedstock mostly consists of two stages: drying and pyrolysis (Zhang et al., 2017). Based on the analysis of Fig. 2, once the temperature of feedstock rose to 100°C or even 200°C, most of the moisture content evaporated and some volatile materials were released. After that, the temperature increased to the maximum level during the pyrolysis process (e.g., 450°C), and the dried feedstock decomposed into oil, gas, and solid char.

The heating profile of the TW is shown in Fig. 2. From the temperature profile graph for all MOP variations, at the beginning of the run, the temperature of TW increased rapidly until it reached the maximum temperature of each MOP, and then decreased until it reached the equilibrium temperature point between the system and its surroundings. The increase of temperature up to 105°C was used to remove fixed water content inside TW (Caroko et al., 2020b; Korkmaz et al., 2009). After that, the cellulose inside the paper layer decomposed at a temperature of 200°C to 400°C (Korkmaz et al., 2009). Meanwhile, the LDPE content of TW began to decompose at 400°C (Siddiqui et al., 2020). Furthermore, the temperature decreased because the capacity of the material used to increase the temperature had decreased (Motasemi and Afzal, 2013).

The MOP affects the heating rate and the maximum temperature reached (Caroko et al., 2020a). This experiment reached a maximum temperature of 486.2°C in 540 s (300 W), 555.4°C in 360 s (450 W), 657.8°C in 220 s (600 W), and 723.2°C in 160 s (800 W). This showed that the higher the MOP, the higher the heating rate and the maximum temperature reached.

In all MOP level variations, mass loss reached 61 wt.% for 800 W, 53.5 wt.% at 600 W, 40.4 wt.% at 450 W, and 40.5 wt.% at 300 W. Based on the analysis of Fig. 2(a), the TW decomposition process started with the evaporation of the moisture-content (represented by the first gradient at temperatures less than 105°C) then continued to the decomposition of paper and ended with the decomposition of LDPE (indicated by the second gradient at temperatures above 105°C). The mass loss graph shows a different trend because of the usage of MOP levels affecting the heating rate of the material used.

According to the analysis, in all variations of MOP, TW can reach the pyrolysis temperature. As a result, MOP 300–800 W is suitable for the MWP process on a mixture of TW...
3.2 Microwave Pyrolysis of TW

The equation used to determine the DTG refers to the calculation performed by Caroko et al. (2020b):

\[
\frac{dTG}{dt} = \frac{dm}{dt} = \frac{m_t - m_{t-1}}{t_t - t_{t-1}} \tag{viii}
\]

DTG (Differential Thermogravimetry) curve is generated as the first derivative of the weight with respect to temperature or time. In the current study, the weight loss rate increased with increasing temperature. The negative value of dTG/dt indicates that the sample mass decreases.

DTG curve from microwave pyrolysis of TW can also be seen in Fig. 2(b). Pyrolysis of all layers from TW occurred in one stage. The first decomposition occurred on the paper layer, whereas the second was decomposition of the LDPE layer. This is similar to study conducted by Korkmaz et al., (2009). Thermal decomposition of TW at 300 W occurred at 400–800 s, 450 W at 250–700 s, 600 W at 50–550 s, and 800 W at 50–400 s. The lower the MOP levels, the longer the decomposition process takes. When compared to other MOP levels, the dTG/dt values in both variations of MOP levels, 600 and 800 W, were the best. This finding indicates that MOP levels have a significant effect on dTG/dt reduction.

The dTG/dt was also positively affected by the presence of SiC as MWA. Using SiC as MWA resulted in the highest final temperature, highest heating rate, and highest gas yield (Zuo et al., 2011).

3.3 The Effect of MOP on the Product Distribution of MWP

The products of pyrolysis varied depending on the type of feedstock (Tröger et al., 2013). In this case, the MWP of TW produced the aqueous phase and organic compounds (wax) in the sample container. The solid residue left in the microwave reactor consisted of char and aluminium foil. The aluminium foil was separated from the char by sinking it in water. The MWP product from TW is divided into five categories: char, aluminium foil, aqueous phase, wax, and gas.

The aqueous phase is a mixture of oxygenated components in water solution, with water being the most essential component (López et al., 2011). It is formed from the degradation of cellulose and lignocellulose (Korkmaz et al., 2009). Light waxes (hydrocarbons C_{21}–C_{40}) are products of polyolefin pyrolysis (low- and high-density polyethylene, and polypropylene) (Jerzak et al., 2021).

Fig. 3 demonstrates the distribution of TW products utilizing microwave oven with SiC as MWA under different MOP levels. It was revealed that at all catalytic temperature, the pyrolysis gas results increased with increasing MOP level, with an increase from 300 W to 800 W. In contrast, the results of pyrolysis wax and solid residue decreased with increasing microwave power from 300 to 800 W. These results were in line with another investigation (Huang et al., 2013). The highest results of gas were 800 W. This might be due to TW cracking reactions and volatilization to volatiles which left the reactor and were stored as non-condensable gases. The stronger organic bonds (e.g. lignin) in the TW were broken when the MOP levels increased, resulting in more non-condensable gas (Kefli et al., 2017). This might suggest that when product vapors are generated as a result of TW pyrolysis devolatilization, more hydrocarbons in the vapors can be converted into permanent gases at higher power.

It should be noticed that the MOP level is an essential element to properly assure TW pyrolysis throughout the MWP process. This is mostly because the heat generated by TW absorbing microwave radiation energy will not only be used for pyrolysis, but a significant portion of the heat will be dispersed to the low-temperature surroundings (Bing et al., 2021).

https://doi.org/10.6703/IJASE.202212_19(4).008
Fig. 3. The product distribution of TW pyrolysis utilizing microwave oven with SiC as MWA and NZ as catalyst under different MOP levels and CTs (a) without catalyst, (b) 350°C, (c) 400°C, (d) 450°C, and (e) 500°C.
Fig. 4. The product distribution of TW utilizing microwave oven with SiC as MWA and NZ as catalyst under different CTs and MOP levels (a) 300 W, (b) 450 W, (c) 600 W, and (d) 800 W

Table 1. The composition and characteristics of gas product from MWP process of TW with/without catalyst at different MOP levels

| MOP Level | CT Level | H₂ (vol.%) | CH₄ (vol.%) | CO (vol.%) | CO₂ (vol.%) | HC₅ (vol.%) | LHV (MJ/m³) | Total Energy (kJ) |
|-----------|----------|------------|------------|-----------|------------|------------|-------------|------------------|
| NA        | 350°C    | 21.60      | 17.10      | 27.95     | 15.55      | 17.80      | 23.30       | 56.82            |
| 300 W     | 400°C    | 21.70      | 16.80      | 28.10     | 15.30      | 18.10      | 23.41       | 59.58            |
|           | 450°C    | 21.75      | 16.75      | 28.35     | 15.00      | 18.15      | 23.46       | 64.68            |
|           | 500°C    | 22.10      | 16.45      | 28.45     | 14.80      | 18.20      | 23.44       | 72.07            |
|           | NA       | 21.95      | 16.95      | 28.85     | 15.30      | 16.95      | 22.86       | 79.97            |
|           | 350°C    | 22.00      | 16.70      | 29.15     | 15.00      | 17.15      | 22.94       | 89.98            |
| 450 W     | 400°C    | 22.45      | 16.65      | 29.55     | 15.25      | 16.10      | 22.35       | 94.79            |
|           | 450°C    | 22.60      | 16.45      | 30.00     | 15.15      | 15.80      | 22.16       | 101.04           |
|           | 500°C    | 22.80      | 16.55      | 30.05     | 15.05      | 15.55      | 22.06       | 112.30           |
|           | NA       | 22.45      | 16.60      | 29.00     | 15.25      | 16.70      | 22.63       | 122.36           |
|           | 350°C    | 22.60      | 16.35      | 29.20     | 15.10      | 16.75      | 22.24       | 132.06           |
| 600 W     | 400°C    | 23.45      | 15.90      | 29.45     | 15.00      | 16.20      | 22.09       | 145.23           |
|           | 450°C    | 23.75      | 15.75      | 29.70     | 14.85      | 15.95      | 21.97       | 165.39           |
|           | 500°C    | 23.95      | 15.65      | 29.85     | 14.80      | 15.75      | 23.02       | 146.47           |
|           | NA       | 22.60      | 16.00      | 29.30     | 14.55      | 17.55      | 23.02       | 178.21           |
|           | 350°C    | 22.80      | 15.65      | 29.65     | 14.60      | 17.30      | 22.80       | 183.77           |
| 800 W     | 400°C    | 23.10      | 15.25      | 30.15     | 14.45      | 17.05      | 22.60       | 184.49           |
|           | 450°C    | 23.95      | 14.95      | 30.55     | 14.30      | 16.25      | 22.12       | 182.97           |
|           | 500°C    | 23.80      | 14.80      | 30.75     | 14.25      | 16.40      | 22.17       | 183.39           |
3.4 The Effect of CT on the Product Distribution of MWP

The product distribution of TW pyrolysis utilizing a microwave oven with SiC as MWA and NZ as catalyst under different CT (without catalyst, 350°C, 400°C, 450°C, and 500°C) are shown in Fig. 4. The MOP levels were 300, 450, 600, 800 W, and the catalyst to feed ratio was 1:1.

The difference in product yields demonstrated that the catalytic temperature had a major impact on product distribution. For example, at 800 W there was a wax yield production of about 1 wt.% in the absence of catalyst, but no wax was generated when catalyst was added. This is similar to Siddiqui’s suggestion that catalyst was added to decrease wax formation (Siddiqui et al., 2020). Furthermore, the gas yield increased from 73 wt.% in the absence of catalyst to 78 wt.% at 500°C catalyst temperature. This occurred because high catalytic temperature conditions supported the breaking of long carbon chains waxes and phenolic compounds. It resulted in the reduction of huge molecules and the production of light chemical compounds (Duan et al., 2017). The cracking reaction, as an endothermic process, was intensified at high temperatures which increased the amount of gas yield. However, when the temperature was raised slightly higher, the synthesis of lighter compounds occurs more rapidly (Dai et al., 2017).

NZ has BET surface area that allows a lot of contact between the catalyst and the pyrolysis gas, which means more gas will be cracked in order to generate shorter chain hydrocarbons (Syamsiro et al., 2014b). On the other hand, NZ has a high acidity which can decrease the liquid yield but increase the gas yield (Miandad et al., 2017). NZ must be pre-treated (activated) because its catalytic characteristics are poorer than commercial catalyst (e.g. surface area of NZ is smaller than commercial catalyst). The effect of the activation type has been studied by Miandad et al. (2019). NZ catalyst was modified via thermal activation and acid activation. Thermal activation of the catalyst produces higher gas yields than acid catalyst activation (Miandad et al., 2019).

Pyrolysis of TW using catalyst has been carried out by Haydary et al. (2013), two types of catalysts used are dolomite and red clay. The results show that at the pyrolysis temperature of 650°C, the red clay catalyst produces a gas yield of 62 wt%, while the dolomite catalyst produces 64 wt%. These results show a significant difference with the MWP of TW using NZ as catalyst in this study. This is due to NZ having a larger surface area than dolomite and red clay. This makes volatile matter streaming from the primary reactor more accessible at the catalytic site and reduces coking-induced pore plugging.

3.5 Composition and Characteristics of Gas Products

During the MWP pyrolysis of TW, gaseous products were produced. The main composition of gas products such as hydrogen (H2), methane (CH4), carbon monoxide (CO), carbon dioxide (CO2), and the other light hydrocarbons (C2Hx) were observed by GC-MS analysis.

Table 1 shows the composition and characteristics of the gas product from MWP of TW with/without catalyst at different MOP levels. Without catalyst addition at 800 W, the composition of H2, CH4, CO, CO2, and HCx were about 22.60, 16.00, 29.30, 14.55, and 17.55 vol.%, respectively. On the other hand, when the catalyst was added into the process the composition of H2, CH4, CO, CO2, and HCx at 350°C CT were about 22.80, 15.65, 29.65, 14.60, and 17.30 vol.%, respectively. Generally, the addition of catalyst, an increase in CT, and an increase in MOP are increasing the formation or release of H2 and CO. Meanwhile, the composition of CO2, CH4, and HCx decreased. This indicates that for catalytic pyrolysis of TW, decarboxylation to CO is preferable than decarboxylation to CO2 (Wang et al., 2015).

The influence of MOP level on temperature and reaction time profile can optimize the cracking reaction, resulting in H2 and CO production, thus increasing the volume percentage of H2 and CO. This indicates that the MWP process of TW using SiC as MWA and NZ as catalyst can produce the main component of syngas (CO + H2). The presence of significant levels of H2 and CO in the gas product suggests that the pyrolysis gas product might be used as an alternative fuel.

The lower percentages of CH4, CO2, and the greater percentages of CO and H2 found in the gas generated in the MWP process at TW might be explained by reaction [1], [2] and the Boudouard reaction (see reaction [3]).

Water gas reaction:

\[
\text{C} + \text{H}_2\text{O} \rightarrow \text{CO} + \text{H}_2 \quad \Delta H_{298K} = 132 \text{ kJ/mol} \quad [1]
\]

Methane gasification:

\[
\text{CH}_4 + \text{H}_2\text{O} \rightarrow \text{CO} + 3\text{H}_2 \quad \Delta H_{298K} = 206.1 \text{ kJ/mol} \quad [2]
\]

Boudouard reaction:

\[
\text{C} + \text{CO}_2 \rightarrow 2\text{CO} \quad \Delta H_{298K} = 173 \text{ kJ/mol} \quad [3]
\]

It is worth noting that the single gas products of volatile cracking may perform gas-phase interactions with others. Thus, methanation reaction (reaction [4]) and water-gas shift reaction (reaction [5]) may change CH4, CO, and CO2 contents (Menéndez et al., 2004).

Methanation reaction:

\[
\text{CO} + 3\text{H}_2 \rightarrow \text{CH}_4 + \text{H}_2\text{O} \quad \Delta H_{298K} = -206.1 \text{ kJ/mol} \quad [4]
\]

Water-gas shift reaction:

\[
\text{CO} + \text{H}_2\text{O} \rightarrow \text{CO}_2 + \text{H}_2 \quad \Delta H_{298K} = -41.5 \text{ kJ/mol} \quad [5]
\]

The total LHV of the gas products via the MWP process of TW was 21.97–23.46 MJ/m3. These results suggest that the gas product is more suitable to be applied as fuel or feed gas for the synthesis of chemicals.

The total energy of the gaseous fraction from the MWP process of TW is shown in Table 1. The total energy of the gas products was 56.82–184.49 kJ. The MOP level and CT have a significant effect on the total energy. When the MOP level and CT are increased, the total energy is increased as well. The maximum total energy was obtained from the experiment at MOP levels of 800 W and CT of 400°C.

In the previous study (Pratiwi et al., 2021), activated
carbon was utilized as MWA in MWP of TW. The total energy obtained was 34.59–171.24 kJ. This value was lower than the total energy obtained in the current study. It demonstrates that MWA plays a crucial part in the MWP process. It is due to the better physical properties (dielectric properties) of SiC than activated carbon (Zuo et al., 2011).

4. CONCLUSION

The MWP process is suitable to convert TW into a gaseous fuel that is both economically sustainable and environmentally friendly, as well as to solve the problem of TW disposal. This process uses SiC as MWA and NZ as catalyst to increase the results and improve the quality of the gas product. The MWP experiments were implemented at different MOP and CT levels. The gas yield significantly increased, and the wax yield decreased by increasing the MOP and CT levels. NZ was used to decrease the formation of wax and increase the combustible gas of the gas product. The main components of the gas products are H2, CO, CO2, CH4, and the hydrocarbons with low chain molecular (C2+).

The production of H2 and CO was increased by the presence of catalyst, an increase in CT, and an increase in MOP. Meanwhile, CO2, CH4, and HC3 compositions decreased. The main component of syngas (CO + H2) was produced by the MWP process of TW, and this suggests that the method had a significant advantage in the production of syngas. The LHV of the gas products was 21.97–23.46 MJ/m3, and the total energy was 56.82–184.49 kJ. Finally, the MWP process is a promising technology to convert TW into an alternative energy source and as synthesis gas.

ACKNOWLEDGMENT

The authors wish to thank the PMDSU grant no 6/EI/KP/PTNBIH/2021 and 2352/UN1/DITLIT/DIT-LIT/PT/2021 from Ministry of Education and Culture, Directorate General of Higher Education, Indonesia for the financial support.

REFERENCES

Atreya, A., Olszewski, P., Chen, Y., Baum, H.R. 2017. The effect of size, shape and pyrolysis conditions on the thermal decomposition of wood particles and firebrands. International Journal of Heat and Mass Transfer, 107, 319–328.

Bing, W., Hongbin, Z., Zeng, D., Yuefeng, F., Yu, Q., Rui, X. 2021. Microwave fast pyrolysis of waste tires: Effect of microwave power on product composition and quality. Journal of Analytical and Applied Pyrolysis, 155, 104979.

Caroko, N., Saptoadi, H., Rohmat, T.A. 2020b. Kinetics of microwave co-pyrolysis of palm oil industry solid waste and polyethylene terephthalate waste. Journal of Advanced Research in Fluid Mechanics and Thermal Sciences, 71, 72–82.

Dai, L., Fan, L., Liu, Y., Ruan, R., Wang, Y., Zhou, Y., Zhao, Y., Yu, Z. 2017. Production of bio-oil and biochar from soapstock via microwave-assisted co-catalytic fast pyrolysis. Bioresource Technology, 225, 1–8.

Dai, X., Wu, C., Li, H., Chen, Y. 2000. The fast pyrolysis of biomass in CFB reactor. Energy and Fuels, 14, 552–557.

Dominguez, A., Menéndez, J.A., Fernández, Y., Pis, J.J., Nabais, J.M.V., Carroll, P.J.M., Carroll, M.M.L.R. 2007. Conventional and microwave induced pyrolysis of coffee hulls for the production of a hydrogen rich fuel gas. Journal of Analytical and Applied Pyrolysis, 79, 128–135.

Duan, D., Wang, Y., Dai, L., Ruan, R., Zhao, Y., Fan, L., Tayier, M., Liu, Y. 2017. Ex-situ catalytic co-pyrolysis of lignin and polypropylene to upgrade bio-oil quality by microwave heating. Bioresource Technology, 241, 207–213.

Gómez, N., Banks, S.W., Nowakowski, D.J., Rosas, J.G., Cara, J., Sánchez, M.E., Bridgwater, A.V. 2018. Effect of temperature on product performance of a high ash biomass during fast pyrolysis and its bio-oil storage evaluation. Fuel Processing Technology, 172, 97–105.

Haydary, J., Susa, D., Dudás, J. 2013. Pyrolysis of aseptic packages (tetrapak) in a laboratory screw type reactor and secondary thermal/catalytic tar decomposition. Waste Management, 33, 1136–1141.

Huang, Y.F., Kuan, W.H., Chang, C.C., Tzou, Y.M. 2013. Catalytic and atmospheric effects on microwave pyrolysis of corn stover. Bioresource Technology, 131, 274–280.

Jerzak, W., Bieniek, A., Magdziarz, A. 2021. Multifaceted analysis of products from the intermediate co-pyrolysis of biomass with Tetra Pak waste. International Journal of Hydrogen Energy.

Kefli, A.A., Khasri, A., Roahazad, N.M., Ahmad, M.A. 2017. Microwave assisted pyrolysis of Imperata Cylindrica with ionic liquids pre-treatment. International Journal of Petrochemistry and Research, 1, 73–75.

Klinger, J.L., Westover, T.L., Emerson, R.M., Williams, C.L., Hernandez, S., Monson, G.D., Ryan, J.C. 2018. Effect of biomass type, heating rate, and sample size on microwave-enhanced fast pyrolysis product yields and qualities. Applied Energy, 228, 535–545.

Korkmaz, A., Yanik, J., Brebu, M., Vasile, C. 2009. Pyrolysis of the tetra pak. Waste Management, 29, 2836–2841.

Li, K., Chen, G., Chen, J., Peng, J., Ruan, R., Srinivasakannan, C. 2019a. Microwave pyrolysis of walnut shell for reduction process of low-grade pyrolusite. Bioresource Technology, 291, 121838.

Li, K., Chen, G., Li, X., Peng, J., Ruan, R., Omran, M., Chen, J. 2019b. High-temperature dielectric properties and pyrolysis reduction characteristics of different biomass-
pyrolusite mixtures in microwave field. Bioresource Technology, 294, 122217.
López, A., de Marco, I., Caballero, B.M., Laresgoiti, M.F., Adrados, A., Torres, A. 2011. Pyrolysis of municipal plastic wastes II: Influence of raw material composition under catalytic conditions. Waste Management, 31, 1973–1983.
Ma, Y. 2018. Changing Tetra Pak: From waste to resource. Science Progress, 101, 161–170.
Menéndez, J.A., Dominguez, A., Ingluano, M., Pis, J.J. 2004. Microwave pyrolysis of sewage sludge: Analysis of the gas fraction. Journal of Analytical and Applied Pyrolysis, 71, 657–667.
Miandad, R., Barakat, M.A., Aburiazaiza, A.S., Rehan, M., Nizami, A.S. 2016. Catalytic pyrolysis of plastic waste: A review. Process Safety and Environmental Protection, 102, 822–838.
Miandad, R., Barakat, M.A., Rehan, M., Aburiazaiza, A.S., Ismail, I.M.I., Nizami, A.S. 2017. Plastic waste to liquid oil through catalytic pyrolysis using natural and synthetic zeolite catalysts. Waste Management, 69, 66–78.
Miandad, Rashid, Rehan, M., Barakat, M.A., Aburiazaiza, A.S., Khan, H., Ismail, I.M.I., Dhavamani, J., Gardy, J., Hassanpour, A., Nizami, A.S. 2019. Catalytic pyrolysis of plastic waste: Moving toward pyrolysis based bio refineries. Frontiers in Energy Research, 7, 1–17.
Motasemi, F., Afzal, M.T. 2013. A review on the microwave-assisted pyrolysis technique. Renewable and Sustainable Energy Reviews, 28, 317–330.
Mushtaq, F., Amran, T., Abdullah, T., Mat, R., Nasir, F. 2015. Optimization and characterization of bio-oil produced by microwave assisted pyrolysis of oil palm shell waste biomass with microwave absorber. Bioresource Technology, 190, 442–450.
Onwudili, J.A., Muhammad, C., Williams, P.T. 2019. Influence of catalyst bed temperature and properties of zeolite catalysts on pyrolysis-catalysis of a simulated mixed plastics sample for the production of upgraded fuels and chemicals. Journal of the Energy Institute, 92, 1337–1347.
Pattiya, A. 2018. Catalytic pyrolysis. Direct Thermochemical Liquefaction for Energy Applications, 29–64.
Pratiwi, I.A.P., Saptoadi, H., Sentanuhady, J., Purnomo, C.W., Rohmat, T.A. 2021. The influence of microwave oven output power and catalytic temperature on products yields of aseptic packaging waste pyrolysis. 11, 1871–1879.
Rex, P., Masilamani, I.P., Miranda, L.R. 2020. Microwave pyrolysis of polystyrene and polypropylene mixtures using different activated carbon from biomass. Journal of the Energy Institute, 93, 1819–1832.
Safdar, M.S., Amini, E., Weise, D.R., Fletcher, T.H. 2019. Heating rate and temperature effects on pyrolysis products from live wildland fuels. Fuel, 242, 295–304.
Shepherd, B.J., Ryan, J., Adam, M., Beneroso Vallejo, D., Castañó, P., Kostas, E.T., & Robinson, J.P. 2018. Microwave pyrolysis of biomass within a liquid medium. Journal of Analytical and Applied Pyrolysis, 134, 381–388.
Siddiqui, M.Z., Han, T.U., Park, Y.K., Kim, Y.M., Kim, S. 2020. Catalytic pyrolysis of tetra pak over acidic catalysts. Catalysts 2020, 10, 602 (article number 602. https://www.mdpi.com/2073-4344/10/6/602).
Sonawane YB. 2015. Use of catalyst in pyrolysis of polypropylene waste into liquid fuel. International Research Journal of Environment Sciences, 4, 24–28.
Syamsiro, M., Cheng, S., Hu, W., Saptoadi, H., Pratama, N., Trisunaryanti, W., Yoshikawa, K. 2014a. Liquid and gaseous fuel from waste plastics by sequential pyrolysis and Catalytic Reforming Processes over Indonesian Natural Zeolite Catalysts. Waste Technology, 2, 44–51.
Syamsiro, M., Saptoadi, H., Norsujianto, T., Noviasri, P., Cheng, S., Alimuddin, Z., Yoshikawa, K. 2014b. Fuel oil production from municipal plastic wastes in sequential pyrolysis and catalytic reforming reactors. Energy Procedia, 47, 180–188.
Tetra Pak. 2021. Food. People. Planet. Tetra Pak Company. Switzerland. Retrieved 2021−12−15 from https://www.tetrapak.com/en-id/sustainability/sustainability-updates.
Tröger, N., Richter, D., Stahl, R. 2013. Effect of feedstock composition on product yields and energy recovery rates of fast pyrolysis products from different straw types. Journal of Analytical and Applied Pyrolysis, 100, 158–165.
Wahono, S.K., Hernawan, Kristiani, A., Tursiloadi, S., Abimanyu, H. 2014. Characterization and utilization of gunungkidul natural zeolite for bioethanol dehydration. Energy Procedia, 47, 263–267.
Wang, K., Johnston, P.A., Brown, R.C. 2015. Comparison of in-situ and ex-situ catalytic pyrolysis in a micro-reactor system. Bioresource Technology, 173, 124–131.
Williams, P.T., Brindle, A.J. 2002. Catalytic pyrolysis of tyres: Influence of catalyst temperature. Fuel, 81, 2425–2434.
Yu, Z., Wang, Y., Jiang, L., Dai, L., Liu, Y., Ruan, R., Duan, D., Zhou, Y., Fan, L., Zhao, Y., Zou, R. 2018. Microwave-assisted catalytic pyrolysis of Chinese tallow kernel oil for aromatic production in a downdraft reactor. Journal of Analytical and Applied Pyrolysis, 133, 16–21.
Zaker, A., Chen, Z., Wang, X., Zhang, Q. 2019. Microwave-assisted pyrolysis of sewage sludge: A review. Fuel Processing Technology, 187, 84–104.
Zhang, Y., Chen, P., Liu, S., Peng, P., Min, M., Cheng, Y., Anderson, E., Zhou, N., Fan, L., Liu, C., Chen, G., Liu, Y., Lei, H., Li, B., Ruan, R. 2017. Effects of feedstock characteristics on microwave-assisted pyrolysis – A review. Bioresource Technology, 230, 143–151.
Zhao, X., Zhang, J., Song, Z., Liu, H., Li, L., Ma, C. 2011. Microwave pyrolysis of straw bale and energy balance analysis. Journal of Analytical and Applied Pyrolysis, 92, 43–49.
Zuo, W., Tian, Y., Ren, N. 2011. The important role of microwave receptors in bio-fuel production by microwave-induced pyrolysis of sewage sludge. Waste Management, 31, 1321–1326.