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Simultaneous Drying and Torrefaction Pretreatment of Organic Waste for Upgrading Calorific Value

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ABSTRACT

Nowadays, renewable energy is of great interest to the world because it reduces greenhouse gas emission (GHG). Torrefaction is a thermochemical pretreatment process that converts biomass into biocoal. Recently, this technology was used during a study where organic waste, a type of biomass, was the focus of the experiment for upgrading biomass. The aim of this research is to evaluate the possibility of whether or not such technology can effectively be used to modify organic waste. Applying systematic composite sampling, the researcher discovered that the top four varieties of high contents were mostly vegetables: cabbage, nappa cabbage, morning glory and kale. The experiment was conducted using a universal oven with a heating rate of around 7.22 °C/min and a controlled air flow rate of 40%. The temperature was set at a controlled range of 80-300 °C and time was set in the range of 0-15 hours. A product called biocoal had a high heating value (HHV) of around 20.08 MJ/kg. Pretreated cabbage samples had a heating value 2.2 times higher than that of an untreated sample. The second test samples: nappa cabbage, kale, and morning glory had higher HHV values than untreated samples at 1.8, 1.8 and 1.4 times, respectively.

1. INTRODUCTION

Today, the world is faced with many challenges but nothing more pressing than the need to reduce if not eliminate the use of fossil fuels and develop more renewable and sustainable energy resources. Biomass renewable energy accounts for a large fraction of renewable energies [1]. Increasing biomass usage for energy reduces greenhouse gas (GHG) emission and meets the targets established in the Kyoto Protocol [2]. In European Union, 30% or more of the total transportation fuel consumption has to be covered by biofuels by 2040 [3]; In the USA, Biomass and waste fuels generated 71.4 billion kilowatt hours of electricity in 2016 or 2% of the total generated in the United States [4]; And in China, the plan establishes targets of biomass power up to 3,104MW in 2020. [5].

The Alternative Energy Development Plan 2018-2037 (AEDP 2018-2037) of Thailand goal is to increase the proportion of renewable energy in the form of electricity, heat, and biofuels by 30 percent in 2037. The initiative is to promote renewable energy production within the full potential of domestic renewable energy

resources to benefit the social and environmental dimensions of the society [6]. In addition, Thailand has currently conducted municipal solid waste (MSW) management in various provinces. Nonthaburi Province, being one of them, is one of the largest and crowded cities in Thailand. Nonthaburi Provincial Administrative Organization has had a well-organized sanitation landfill method since 2006. In 2018, the information of municipal solid waste (MSW) increased 11.1 percent from the previous year (around 1,500 ton per day) and had around 0.73 million tons in 2019 [7]. Organic waste, especially vegetables around 1.5 ton per day, is collected from Nonthaburi city municipality fresh market due to limitation of landfill areas; therefore, waste management is necessary [7].

Large amounts of organic waste from fresh markets, which are now referred to as MSW, can be utilized as biomass for producing refuse-derived fuel (RDF). Biomass in this case (*i.e.*, food waste) can be raw material for renewable energy; however, dumping waste in landfills without oxygen continuously produces methane, a potent greenhouse gas [8]. Because of the inherent drawbacks of utilizing biomass many researchers have focused on improving biomass properties for renewable energy. Nowadays, biological and thermochemical conversion technologies have been used for re-utilized or value-added biomass [9]-[10]. However, biological techniques also give rise to several operational challenges. Mixed wastes, for instance, present a critical issue with these techniques [11]. Drying, bio-drying, hydrothermal, torrefaction and pyrolysis are well-known for their thermochemical conversion techniques; that is, they upgrade biomass in

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forms of energy efficiency, generation of value-added products, and improve pollution control. High contents of moisture in organic waste makes low calorific value and contributes to the wet material for RDF especially organic waste. Therefore, it is necessary to remove moisture and increase calorific value by employing torrefaction technology such as similar work had done [11]-[17]. Because of increasing of torrefied temperature enhanced the degradation of hemicellulose, lignocellulose, cellulose and lignin. The obtained product is called torrefied biomass or biocoal. Surprisingly, little discussion has been devoted to the issue of organic waste as a solid fuel. Hitherto, no data has been published regarding the torrefaction of organic waste. Vegetable is one of organic waste, might be alternative biomass as will see in this research. Moreover, no particular study was observed so far which considered the heat treatment experiment of organic waste. Most information regarding the torrefaction of organic waste remains to be studied.

This research aims to examine simultaneous drying and torrefying pretreatment for increasing calorific value of organic waste by using universal oven. To provide a deep insight into the technique, this study is intended to investigate the effects of torrefaction temperature and residence time upon the properties of various types of organic waste as a solid fuel.

2. METHODOLOGY

2.1 Raw Material and Preparation

One ton and a half of organic waste was obtained from Nonthaburi city municipality fresh market per day that was presented as raw materials. Organic waste composition was determined by systematic sampling from Nonthaburi city municipality fresh market, Nonthaburi province, Thailand. Nonthaburi fresh market has large amounts of fresh organic waste, about 1.5 ton/day. Figure 1 shows organic waste composition.



Fig. 1. Organic waste composition by systematic sampling from Nonthaburi city municipality fresh market, Thailand.

2.2 Experimental Procedure

The procedure used fresh cut samples of size $1 \times 1 \text{ cm} \pm 0.17 \times 0.24 \text{ cm}$ for size control and then weighed 5 g per each sample and placed them in aluminium bowls. Drying for moisture removal and torrefaction process were conducted by using Universal oven (UF55, Memmert, Germany). Effect of material size was done at size of 1×1 , 2×2 and $3 \times 3 \text{ cm} \times \text{cm}$. Time and temperature were controlled at 1, 3, 5, 10 and 15 hours and 80, 105, 150, 200 and 300°C , respectively. Electric heat was used as energy source with heating rate around $7.22^\circ\text{C}/\text{min}$. Forced-air circulation in the oven was controlled at 40% air flow rate by air turbine without air flow in or out of the oven (0% flap).

2.3 Product Characterization

To observe the effect of torrefaction conditions on fuel properties of torrefied biomass, proximate analysis, calorific value, surface morphology and chemical functional groups were analyzed. Moisture was analyzed

according to American Society for Testing and Materials Methods (ASTM E 871-82) [18] by using Universal ovens (UF55, Memmert, Germany). Volatile component was analyzed according to American Society for Testing and Materials Methods (ASTM E-872) [19]. Ash was analyzed according to American Society for Testing and Materials Methods (ASTM D 1102) [20]. Fixed carbon (FC) was calculated with the following equation:

$$\text{FC} = 1 - \text{M} - \text{VM} - \text{ASH} \quad (1)$$

where M, VM, and ASH stand for moisture, volatile matter, and ash content, respectively. Fiber analysis, fiber contents, cellulose, hemicellulose and lignin contents were analyzed according to AOAC (AOAC Official Method 2002.04, 2016 [21], AOAC Official Method 973.18, 2016 [22] and AOAC Official Method 973.18, 2016 [22]), respectively. C H O N S analysis were analyzed according to methods of ISO 16948:2015 with CHNS/O Analyzer with FlashSmart model, Thermo Scientific (Thermo Quest). Calorific value (high heating value, HHV) of torrefied biomass was analyzed

using 1341 plain jacket bomb calorimeter, Parr®, USA in accordance with ASTM E 711 Method [23]. Surface morphology of torrefied biomass was analyzed by scanning electron microscopy (SEM), Quanta450W, FEI, USA and processed with xT microscope control software. Surface chemical functional groups were determined by FTIT Spectrometer, (Spectrum Two, UATR Two, Perkin Elmer®, USA) with The Spectrum 10™ software. Thermogravimetric Analyzer (TGA) was analyzed by Pyris 1, Perkin Elmer®, USA.

2.4 Calculation

Removed moisture, mass yield (Y_{mass}), energy yield (Y_{energy}) and enhancement (E_{energy}) were calculated according to Equations (2)-(5). Dimension of removed moisture percentage were based on fresh weight or called wet basis and calculated according to equation;

$$\% \text{Removed moisture} = \left(\frac{\text{weight before drying} - \text{weight after drying}}{\text{weight before drying}} \right) \quad (2)$$

$$Y_{mass} = \frac{m_{\text{torrefied char}}}{m_{\text{raw biomass}}} \quad (3)$$

$$Y_{energy} = Y_{mass} \left(\frac{HHV_{\text{torrefied char}}}{HHV_{\text{raw biomass}}} \right) \quad (4)$$

$$E_{energy} = \frac{HHV_{\text{torrefied char}}}{HHV_{\text{raw biomass}}} \quad (5)$$

where $m_{\text{torrefied char}}$ and $m_{\text{raw biomass}}$ in Equation (3) represented the mass of the torrefied char and the mass of raw biomass at the initial time (g), respectively. $HHV_{\text{torrefied char}}$ and $HHV_{\text{raw biomass}}$ in Equations (4) and (5) signified the higher heating value of the torrefied biomass and raw biomass (MJ/kg), respectively. Remark here: raw biomass is referred as biomass after drying at 105°C 1 h. For each condition was repeated three times.

3. RESULTS AND DISCUSSION

3.1 Composite Sampling

Organic waste composition was done by systematic sampling method as shown in Figure 1. Composition of organic waste was done systematically by composite sampling [24] from 1.5 ton of organic waste from Nonthaburi city municipality fresh market. Firstly, unit boundary was set and then divided into six sections. In each section divided into 4 sections, A, B, C and D and more dividend to twelve subsections. Proportion of each

type of organic under subsections was evaluated and calculated in from of volumetric percentage. Top four varieties, cabbage, nappa cabbage, morning glory and kale were found in high contents and selected as raw materials. Cabbage has the highest content in organic waste (56.25% by volume). Nappa cabbage was second in row (22.40%). Morning glory (6.42%) and kale (4.17%) were the third and fourth, respectively. The rest was around 10.76% consisting of false pak choi, basil, pumpkin and banana blossom.

3.2 Characteristics of Organic Waste

Nappa cabbage, morning glory and kale had portion of stalk and leaf. The ratio of stalk and leaf of Nappa cabbage, morning glory and kale by weight were 59:41, 71:29 and 65:35, respectively. Properties of each proportion are provided in Table 1. Radish was also selected and investigated because of difference in morphology. All varieties had moisture more than 90%. Stalk had higher moisture content than leaf. The HHV of cabbage was higher value than morning glory around 18.64% and higher than kale and radish around 11.41%. Leaf of all four varieties, kale, morning glory and nappa cabbage had higher heating value than stalk about 21%, 38% and 17%, respectively. This basic information is useful for certain heating value requirement. The HHV of organic waste can be calculated by using heating value of each portion according to equation:

$$HHV = \left(HHV \times \left(\frac{\% \text{weight of stalk}}{100} \right) \right) + \left(HHV \times \left(\frac{\% \text{weight of leaf}}{100} \right) \right) \quad (6)$$

This equation calculated heating value of nappa cabbage, kale and morning glory at treating temperature at 105°C and found the error data from this equation compared to exacted value around 4.12%. TGA analysis of organic waste was shown in Figure 2. Weight loss was sharply decreased at lower temperature around 200°C and gradually decreased to around temperature 450°C.

These vegetable samples contain high moisture contents as seen in Table 1. It implied that the process must input large energy for moisture removal. The ratio of energy input to energy output from torrefied biomass was low and inefficient in the biomass energy production. Such energy ratios of cabbage, nappa cabbage, morning glory, kale and radish were -0.021, -0.082, -0.090, -0.057 and -0.034, respectively.

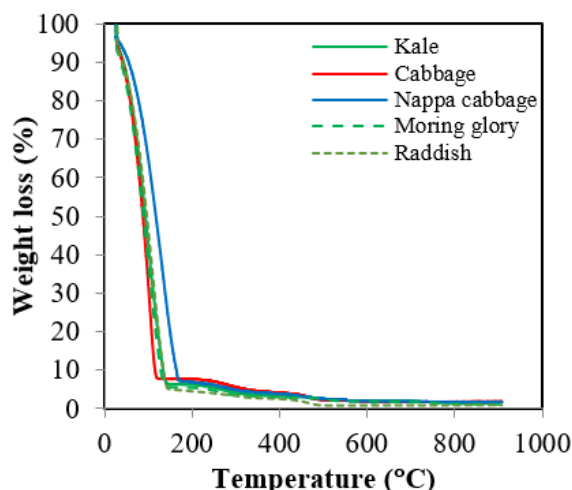


Fig. 2. TGA of organic waste.

3.3 Effect of Size

Fresh organic waste cannot directly be used as biocoal or RDF in furnace because they cannot be burnt; therefore, they have to remove moisture through drying. Material size has affected drying rates. The removed moisture rate was calculated at slope initial of the curve which presents the highest difference of percentage removed moisture versus time or called the removed moisture rate. Effect of material size on removed moisture at 80°C is demonstrated in Figure 3; (a) cabbage (b) nappa cabbage (stalk) and nappa cabbage (leaf) with dimensions Number×Number = width × length (cm. ×cm.) at 80°C and (c) cabbage at 200°C. Cabbage with smaller material size has higher percentage removed moisture rate than bigger size as the

same trend as nappa cabbage (salk) because of higher surface area. The effect of material size on the percentage removed moisture rate was not found in nappa cabbage (leaf); it might be because of thickness. At 200°C, torrefaction zone, material size did not affect to the percentage of removed moisture rate. However, at higher operating temperature, the percentage removed moisture rate was higher than lower operating temperature (higher value of slope at 200°C). Smaller size had higher HHV; however, HHVs showed no significant difference (19.15-19.10 kJ/kg) that contradicted Peng et al., 2012’s research [25] with smaller material size in dimension of micrometer can make product with high value of heating value. It might because of too big material size in this research

Table 1. Characteristics of organic waste.

Compositions (%wb)	Types							
	Cabbage	Nappa cabbage (stalk)	Nappa cabbage (leaf)	Moring glory (stalk)	Moring glory (leaf)	Kale (stalk)	Kale (leaf)	Radish
Moisture	93.33±0.14	96.46±0.37	94.43±0.86	96.22±0.01	92.03±0.67	93.56±0.55	90.04±0.96	96.01±0.11
Ash	0.82±0.04	0.47±0.02	0.67±0.04	0.85±0.06	1.02±0.04	1.17±0.08	0.76±0.17	0.44±0.05
Fixed carbon	2.40±0.23	2.30±0.21	4.30±0.55	2.20±0.70	5.09±0.21	5.19±0.63	8.57±0.96	3.07±0.28
Volatile component	1.10±0.05	0.37	1.71±0.21	1.41	0.85±0.17	1.39	0.38±0.00	0.48±0.11
Ultimate analysis (% as received)								
C	38.89±0.80	37.03±1.44		38.73±2.79		37.52±0.67		39.05±0.25
H	5.50±0.13	5.47±0.41		5.31±0.49		5.40±0.41		6.03±0.16
N	3.49±0.01	4.11±0.88		5.23±0.63		3.61±0.50		1.44±0.16
O	37.61±1.41	42.76±1.96		35.98±0.01		42.70±2.55		43.37±0.40
S	1.26±0.19	1.16±0.04		1.04±0.15		0.88±0.01		0.81±0.08
Fiber analysis (%)								
Cellulose	2.42±0.06	0.75±0.1		5.92±0.14		1.63±0.04		0.56±0.01
Hemicellulose	2.02±0.03	0.66±0.01		7.69±0.16		0.70±0.01		0.19±0.02
Lignin	0.70±0.01	0.39±0.00		4.31±0.06		0.38±0.01		0.15±0.00
HHV (MJ/kg) (as-dried)	16.85	13.53	16.37	11.64	18.78	13.71	17.35	14.87

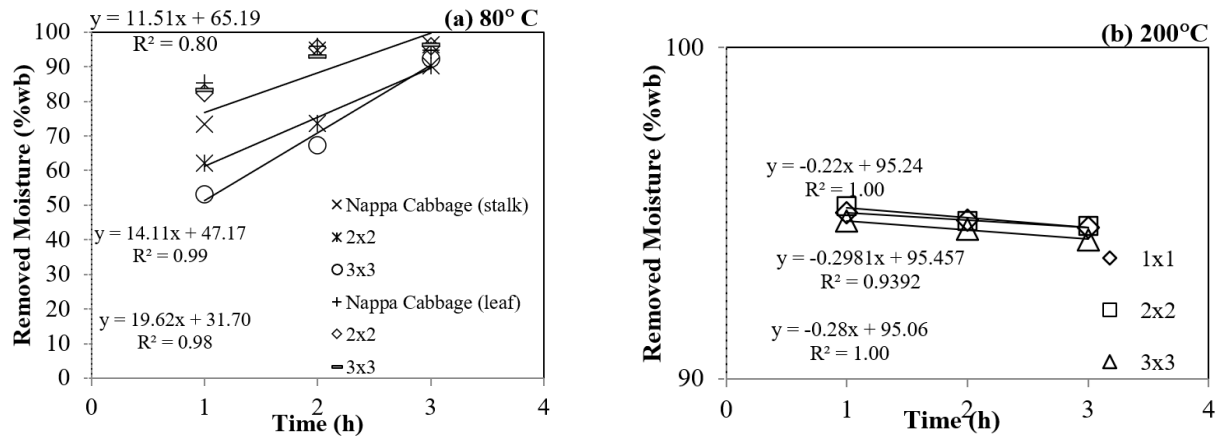


Fig. 3. Effect of material size on removed moisture (a) cabbage at 80°C (b) nappa cabbage (stalk) and nappa cabbage (leaf) with dimensions Number×Number = width×length (cm.×cm.) at 80°C (c) cabbage at 200°C.

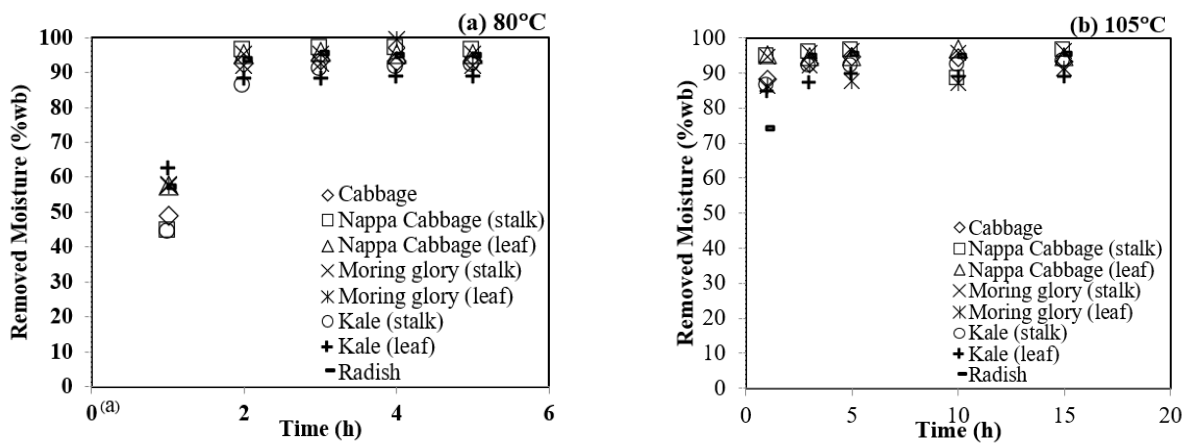


Fig.4. Effect of drying time at (a) 80°C and (b) 105°C of various organic waste varieties.

3.4 Effect of Time

Organic waste composes of biological structure, cellulose, hemicellulose and lignin. When fresh leaf of vegetable was placed at high temperature, it shrank along the vein whereas stalk shrank along the radius and decreased in size. Because unbound water is initially removed, bound water comes out from vegetable cell surface. Longer proceeding times changed the color of the surface to brown because of browning effect and burning. Consequently, increasing the drying time enhances the browning effect. Browning is most noticeable among white radishes. Effect of drying time at 80°C and 105°C demonstrates in Figure 4. The percentage of removed moisture rate is at its highest at the initial stage of the operation; then it flattens out around drying time of 3 hours. At higher drying temperature of 105°C in Figure 4 (b), the highest of removed moisture rate occurs in shorter time (only 1 hour) compared to 80°C in Figure 4 (a). Simultaneous removed moisture process and torrefaction process of cabbage at 150°C from 1 hour to 3 hours can increase HHV products from 13.86 to 17.06 MJ/kg (increasing around 23%). As a result, the moisture was constant

while HHV remained the same. However, the HHVs were not significantly different because the operating condition was proceeded under 40% airflow rate which had contacted to remain oxygen that might be affected to HHVs.

3.5 Effect of Temperature

Drying temperature effect on physical appearance of biocoal is illustrated in Figure 5. Increasing in drying temperature enhanced brown and black color and powdered biocoal. These changes can engender several reactions: the decomposition of cellulose, the crosslinking cooperating with condensation reactions, the enzyme-mediated (Maillard) reactions and the oxidation reaction [26]. Effect of drying temperature on percentages of removed moisture is shown in Figure 6. The increase in drying temperature increases the percentage of removed moisture. At higher temperatures than 105°C for 1 hour, the percentage of removed moisture is constant and can remove moisture more than 74%. The highest percentage value of removed moisture gets around 98.1% (moring glory (stalk)). Increase temperatures from 80 to 105°C increases HHV of biocoal. The highest HHV is found at 300°C (20.08

MJ/kg); however, in the drying process, biocoal might have lower HHV because of the burning effect of oxygen. When compared to raw biomass, an increment in HHV of 85.30% after simultaneous drying and torrefying. Remarking here that organic waste at operating temperature lower than 80°C for 1 hour was

still wet and could not ignite in the explosive calorimeter. Operating higher than 80°C or longer time than 1 hour can make organic waste dry and remove microorganism. The results found were the same in all varieties of organic waste.



Fig. 5. Physical appearance of cabbage at various temperatures; (a) 80°C (b) 105°C (c) 150°C (d) 200°C (e) 300°C at 15 hours drying time.

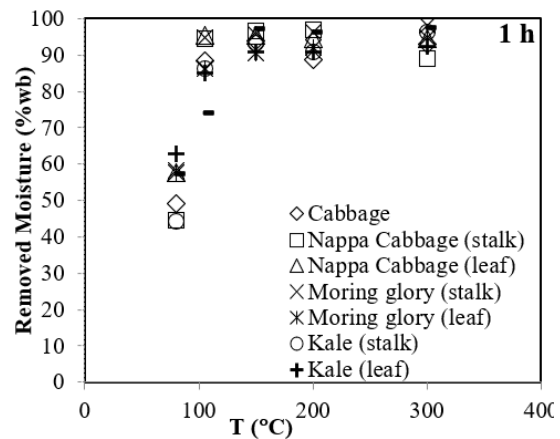


Fig. 6. Effect of drying temperature at 1 hour drying time.

For other important parameters, as in mass yield (Y_{mass}), energy yield (Y_{energy}) and enhancement factor (E_{energy}), which some research called high heating value ratio (HHV ratio) and energy density ($Y_{energy\ density}$) [16-17], were used for pretreatment evaluation. Results of Y_{mass} , Y_{energy} and enhancement factor of cabbage are shown in Figures 7, 8, and 9, respectively. Y_{mass} at various temperature and time of cabbage was constant in range of drying zone (80-150°C) then increased continuously to 200°C (the highest at 1- and 3-hours drying time). In range of torrefaction zone (200- 300°C), mass yield decreased continuously as the same as Nitipong *et al.*, 2020 [16] because of biomass changed to char structure whereas mass yield of nappa cabbage, morning glory and kale were continuously decreasing all along both drying and torrefaction zones. Y_{energy} of cabbage was constant in range of drying zone (80-150°C) (the highest at 200°C 1 hour) and decreased continuously from 200 to 300°C. The results of other organic waste varieties found the same except kale. Energy of cabbage is shown in Figure 9, decreasing in drying zone then increasing in torrefaction zone. The

results found the same in nappa cabbage except morning glory and kale at 10- and 15-hours drying time. After biomass preparation, burnt calorimeter and found hard solid of slag at operating temperature was lower than 200°C. However, at higher temperature than 200°C, there was ash in the crucible. Morning glory had larger amount of slag than other types. The results found the same in previous research that contained inorganic residue [9], [26].

Figure 10 FTIR results of nappa cabbage products, which were treated at various temperatures, was demonstrated at torrefied time of 5 hours. The increasing of torrefied temperature enhanced the degradation of hemicellulose, cellulose and lignin [9], [28]. Moreover, thermal can degraded hemicellulose, cellulose and lignin [29]. For torrefied temperature higher than 150°C, FTIR peaks of hemicellulose and cellulose had obviously disappeared. Moreover, increasing temperature removed moisture – stemming from the depletion of hemicellulose – made cell dense of carbon and became smoother after torrefaction [9], [16], [28] as shown in SEM images, Figure 11.

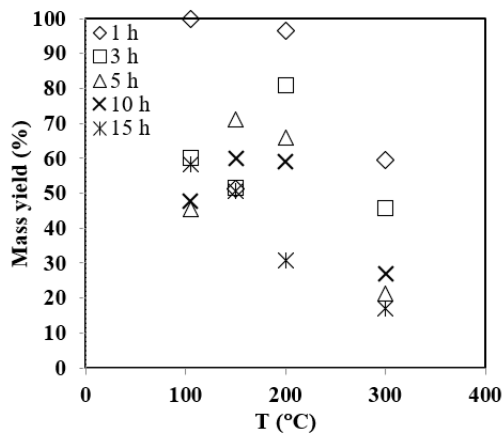


Fig. 7. Effect of torrefaction temperature on the mass yield.

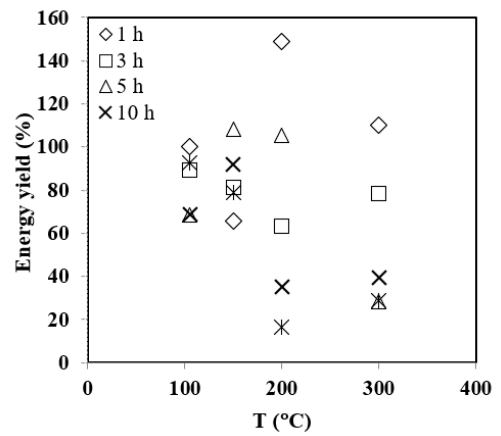


Fig. 8. Effect of torrefaction temperature on energy yield.

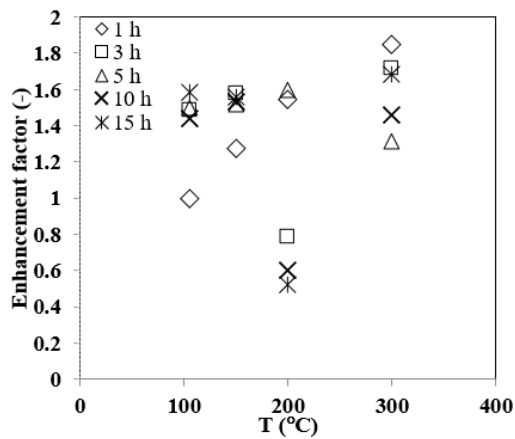


Fig. 9. Effect of torrefaction temperature on enhancement factor at various torrefaction times.

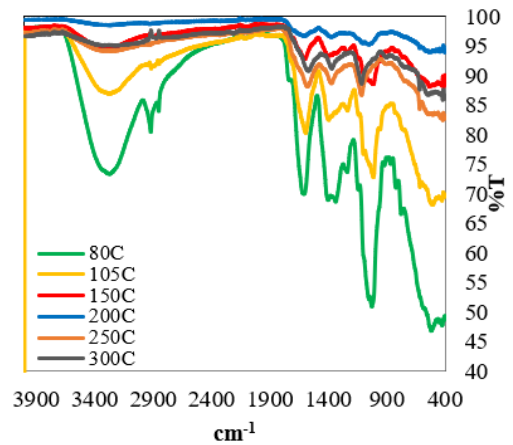


Fig. 10. FTIR of torrefied nappa cabbage at various torrefaction temperatures at 5 hours.

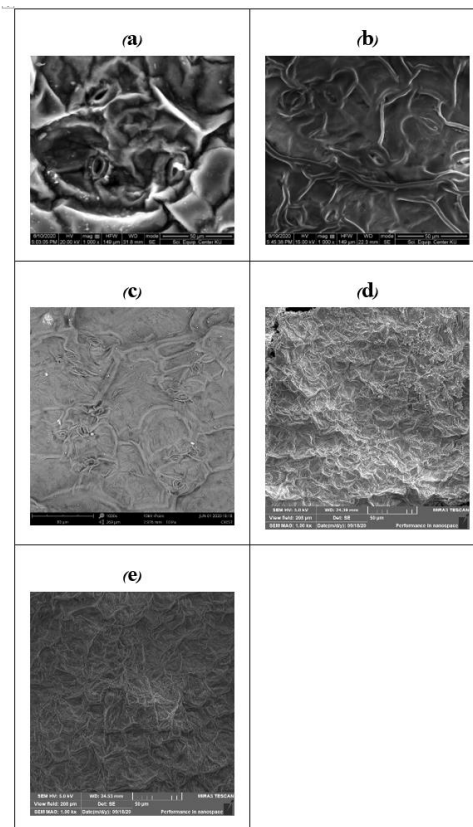


Fig. 11. Effect of temperature on SEM of cabbage at (a) untreated sample (b) 80°C (c) 105°C (d) 200°C (e) 300°C at 5 hours.

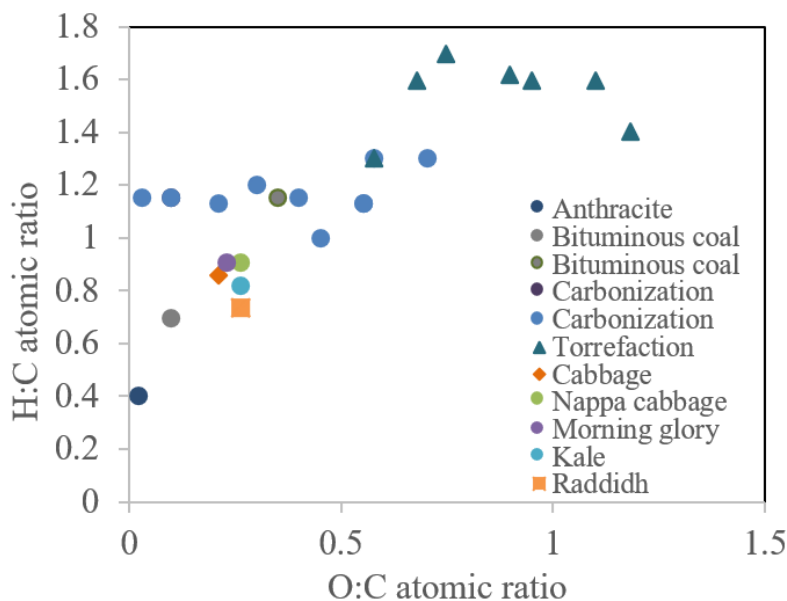


Fig. 12. Van Krevelen diagram for comparison with different fossil fuels [31] and organic wastes [32]-[34].

3.6 Specific Energy Consumption

High operating temperature increased power use and the specific energy consumption (SEC) of organic waste during the process which expressed in Wh/kg of product as revealed in Equation (7): 1 hour 105°C SEC = 3.31 (661.69 Wh) and 1 hour 300°C SEC = 6.43 (1,285.57 Wh). The energy provides to biomass:

$$SEC = \frac{E}{m_{bio\ char}} \quad (7)$$

where E is energy used for torrefaction and m_{bio} coal is weight of biocoal product.

3.7 Calorific Value Comparison

The calorific value of torrefied biocoal product was in the same range of general torrefied biomass by HHV in range 16-29 MJ/kg whereas coal has HHV around 25-35 MJ/kg [9], [26]. Biocoal product has high HHV than biomass, sugarcane, soybean, corn, rice, sorghum and sunflower [30]. However, biocoal product was lower HHV than torrefied bagasse (24.01 MJ/kg) [17] around 19%.

3.8 CHO Analysis

Amount of C H O obtained from this research was in range of fossil as seen in Figure 12. Drying zone can decrease H and O content in organic waste and torrefaction zone, thermal treatment can degrade hemicellulose, cellulose and lignin [29].

4. CONCLUSION

Thermochemical pretreatment, by using drying and torrefaction technology, can upgrade organic waste by increasing high heating value (HHV). Product called coal had HHV at the highest value around 20.08 MJ/kg. Top four varieties as in cabbage, nappa cabbage, morning glory and kale were found in high contents from Nonthaburi city municipality fresh market, Thailand. This research used many parameters: mass

yield (Y_{mass}), energy yield (Y_{energy}) and enhancement factor (E) and chemical structure of biocoal and each operating condition was analyzed by FTIR and SEM. The results linked well to other types of biomass in previous research. The suitable torrefaction condition of cabbage was 300°C for 1 hour; nappa cabbage was around 300°C for 1-5 hours; morning glory was around 300°C for 1-5 hours and kale was 300°C at 3 hours. This research has shown that organic waste can be treated or modified by drying and torrefaction technology.

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