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Current Status of Hydrothermal Treatment for Energy and Material Recovery Toward a Sustainable Post-consumer Material Cycle

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Abstract

The demand for energy-efficient and environmentally friendly municipal solid waste (MSW) processing has increased in developing countries. The thermochemical process offers a fast and reliable solution to reutilize or reduce the volume of MSW. Hydrothermal treatment is a novel MSW treatment technology that is compatible with high-moisture-content feedstock. It involves the thermal degradation of MSW in pressurized water or steam, which promotes the disintegration of cellulosic and polymer materials. Recent advances have shown effective MSW conversion into homogenous solid hydrochar with higher energy density. Alkali and chlorine content, which causes issues in combustors, was successfully removed due to the washing effect of hydrothermal treatment. The possibility of activated carbon production also exists because the surface area is significantly increased after the treatment. This paper presents an overview of the latest development of hydrothermal treatment in the field of post-consumer waste and MSW treatment, with particular focus on the operating conditions and physicochemical characteristics of the hydrochar. Several experimental results from post-consumer waste feedstock were compiled and interpreted using principal component analysis to observe the effect of different operating conditions and feedstock during the hydrothermal process.

1. Introduction

Municipal solid waste (MSW) management in many countries has focused on the promotion of the “reduce, reuse, and recycle” principle in the past few decades [1]. Urban mining and landfill mining are the most popular methods of recovering and reusing secondary and tertiary resources [2],[3]. A significant amount of accumulated post-consumer waste is present in anthropogenic stocks and landfills. Several layers of separation technique are used to maximize the recycling potential, but in some cases, the contamination remains high, thus potentially
increasing recycling costs. These materials will mostly end up in landfills or processed overseas [4]. In developing countries such as Indonesia, wet organic wastes from food residue and parks dominate the composition of MSW (Figure 1) [5]–[8]. Various waste conversion technologies have been developed to effectively recover the energy and/or synthesize the material from highly contaminated waste. The overall scheme of waste conversion technology is shown in Figure 2.

The appropriate conversion route depends on the composition and characteristics of waste. Recently, the thermochemical route has been favored because it has a faster processing time and yields higher energy density compared with the biochemical route. On the basis of the targeted product and operating conditions, the thermochemical process can be classified into several processes, as shown in Table 1.

![Figure 1. Waste Fraction from the Four Most Populated Cities in Indonesia](image1)

![Figure 2. Waste Conversion Technology for Energy and Material Recovery](image2)
Incineration is the most common thermochemical technology that is used to recover energy from waste. In principle, incineration is the oxidation of combustible waste into non-combustible gas and ash [9]. An incineration plant consists of a combustion chamber and heat recovery system and involves a gas cleaning process. The incineration process includes several stages, such as drying and degassing, pyrolysis and gasification, combustion, and post-combustion [10]. Those processes occupy almost the same space and time in the reactor and react almost instantly. The design of the reactor, feed characteristics, and air supply influence the performance of the incineration process in terms of energy efficiency and pollutant emission [11].

The main challenge of incineration is that most of the unprocessed contaminated waste contains a high percentage of plastic. Developed countries, such as Japan and those in northern Europe, account for 20% and 13% of plastic fraction in their waste, respectively. Developing countries in South America and East Asia have about 10% of plastic in their waste stream [12]. Organic chlorine in plastic waste is the main contributor to the formation of hydrochloric acid, dioxin, and furan in flue gas. Europe is known to implement strict standards for the chlorine content in flue gas, which ranges between 0.1 to 1.0% [13].

As a result of the increasing concern for the environment, gasification has become a favorable technology for producing synthetic gas (syngas) for power generation or chemical feedstock. Gasification converts feedstocks into syngas by partial oxidation [14]. Gasifying agents, such as air, oxygen, and steam, are used as an oxygen carrier. Recently, recycled flue gas with a high amount of carbon dioxide was mixed with oxygen to prevent NOx formation during combustion [15]. Another similar system that uses a solid oxygen carrier, called chemical looping gasification, has also been developed recently. In this case, metal oxide as an oxygen carrier performs a solid–solid reaction with waste feedstock to produce a high yield of hydrogen [16].

Pyrolysis has a lower environmental impact compared with incineration. Pyrolysis is a thermal degradation process in the absence of oxygen with an operating temperature of 400–600 °C. Pyrolysis yields gaseous, liquid and solid products. Different product compositions could be arranged by modifying the temperature, pressure, and catalyst. The typical product energy yield from pyrolyzed waste is between 5 to 15 MJ/m³ [17]. Secondary treatment of pyrolysis gas and char has been performed through condensation of the gases into usable oil mixtures or incineration of gas and char to generate heat and destroy the organic matter. One advantage of the pyrolysis process is that it allows multiple product generations in one process. The organic fraction can be recovered for material or fuel (e.g., methanol and Fischer–Tropsch fuels) [18]. Char can be utilized for various feedstocks, such as solid fuel, activated carbon, and carbon nanotubes [19]. Moreover, pyrolysis gas could be used in gas engines or gas turbines for power generation [20].

Carbonization is a process that increases the content of carbon element from organic material by removing volatile matter, reducing moisture content, removing inorganics, and reducing the atomic ratios O/C and H/C, usually without the presence of oxygen [21]. Torrefaction and hydrothermal are commonly used in the carbonization process. Torrefaction prefers dry material because inert gas is used as a medium in temperature between 240 to 320 °C, while hydrothermal uses highly pressurized steam as a medium to break down the long carbon chain at temperatures between 180 to 250 °C [22],[23]. The steam will also wash away inorganics, such as chlorine, potassium, and heavy metals [24]–[26].

Some inherent properties of MSW, such as high moisture content, low energy density, hygroscopicity, low heating value, high alkali content, heterogeneity, and high ash content, have become challenges in its thermochemical conversion. MSWs, especially in developing countries, are usually mixed through improper separation at the source. The composition is dominated by food waste, thus causing very high moisture content. It also has low bulk density because of the high amount of plastic bag waste. The condition worsens in the rainy season, which could usually last for more than six months in tropical countries [27]. This condition leads to difficulties in collecting, handling, and transporting the MSW to the landfill site. Moisture in MSW also decreases its heating value in general.
Organics from food and garden wastes have fibrous and tenacious characteristics, while plastic waste has high elasticity. As a result of those characteristics, MSW is difficult to grind into small homogenous particles. Poor grindability can cause low reactor performance and other serious problems [28]. High ash content with high alkali and chlorine content can cause slagging and fouling in the furnace and promote the production of corrosive and toxic flue gases [29]. The main contributor to fouling is the content of the inorganic of the feedstock. Sodium and potassium lower the melting point of ash, thus increasing ash deposition and fouling of boiler tubes. The alkanes are more available and reactive in the bio-based organics rather than in coal deposits [30]. Accumulation of ash reduces heat transfer and causes severe corrosion at high temperature. Ash deposition from biomass fuel is denser and harder to remove compared with coal. Ash content also directly affects the heating value. A high ash content means a low heating value [31]. Therefore, MSWs in developing and tropical countries are more difficult to burn or gasify with high efficiency and low emission.

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

2. Hydrothermal Treatment

Hydrothermal treatment is a process involving steam/water and elevated temperatures. Three types of hydrothermal treatments exist: hydrothermal carbonization (HTC), hydrothermal liquefaction (HTL), and hydrothermal gasification (HTG). Wet feedstock such as MSW, which can retain up to 60% of moisture in its weight, can be processed without pre-drying, thus allowing energy saving [32]. Similarities exist between the targeted product and the working temperature between HTC and torrefaction, HTL and pyrolysis and torrefaction, and HTG and gasification. However, hydrothermal treatment could run at a lower temperature because the water acts as a solvent, catalyst, and reactant in the process. Table 2 shows the distinction between each hydrothermal process in detail.

Water plays a vital role during the reaction process. The water characteristics and properties will change depending on the temperature and pressure. In the temperature range of 200–280 °C, the acidic and basic properties will change depending on the ion presence. The dielectric constant decreases and make it act as a nonpolar solvent. The hydrogen bonds are also few and weak. The isothermal compressibility is higher, which helps in the destruction process of the material. The solubility of organic compounds increases, whereas that of inorganic salts decreases [33]. At 250–350 °C, the feature of water as a solvent will be similar to that of organic solvents at a room temperature. At a subcritical temperature (100–374 °C), the ionization constant of water increases with temperature. Nevertheless, upon approaching the critical point (374 °C, 22.1 MPa), the ionization constant, the dielectric constant, and the concentration of ionization products decrease sharply. Usually, reactions in liquid are controlled by diffusion; thus, the rate of reaction depends on the viscosity of the liquid [34].

During the hydrothermal process, added water enters the subcritical phase, where the liquid water behaves as a nonpolar solvent. The high ionization degree of water at high temperature and pressure is followed by the dissociation of water into OH− and H3O+, which has acidic and basic characteristics at the same time [35]. The subcritical condition promotes hydrolysis reaction,

<table>
<thead>
<tr>
<th>Table 2. Various Hydrothermal Treatment Process</th>
</tr>
</thead>
<tbody>
<tr>
<td>Process</td>
</tr>
<tr>
<td>---------</td>
</tr>
<tr>
<td>HTC</td>
</tr>
<tr>
<td>HTL</td>
</tr>
<tr>
<td>HTG</td>
</tr>
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</table>

Makara J. Technol.  April 2020 | Vol. 24 | No. 1
which reduces the activation energy of cellulose and hemicellulose. Thus, the feedstock will be depolymerized and degraded into water-soluble products [36]. The hydrolysis of cellulose and the decomposition of hemicellulose convert 40%–60% of the initial biomass into a dissolved state.

Temperature is also a critical element that influences product characteristics after hydrothermal treatment. The temperature has a positive effect on the increase of carbon content but decreases the hydrochar yield [37]. The rate of hydrolysis and depolymerization of biomass depends on temperature. During hydrothermal treatment, hemicellulose was hydrolyzed in the hydrothermal temperature between 180 to 200 °C, lignin was degraded in the hydrothermal temperature between 180 to 220 °C, and cellulose was degraded in the hydrothermal temperature above 220 °C [38].

The span of reaction time up to 2.5 h has a significant effect on hydrochar yield, ash content, carbon content, O/C ratio, energy densification, and energy yield [39]. Retention time promotes changes in morphology, which is related to the higher released volatiles, and more char carbonization occurs. A reaction time longer than 2.5 h tends to increase the ash content and lower the yield. Some ash formed during the carbonization stages could seep into the hydrochar pores at a longer retention time [40],[41].

3. Methods

Experimental data on the effects of hydrothermal treatment temperature, retention time, and solid load on carbon content, hydrogen content, oxygen content, and high heating value were compiled from relevant literature reports. The feedstock, which correlates with post-consumer activity, such as mixed MSW or separated recyclables, were examined thoroughly. The final dataset included 29 individual experiments on 10 different feedstocks (Table 3) [42]–[48]. The dataset provides an overview of different post-consumer goods treated by hydrothermal treatment. The compiled data were reduced to enable comparison between different experiments. Reaction time was expressed in minute (min) and was log10 transformed. The reactor solid load was expressed as a weight percentage (%) of the combined mass of added liquid and the feed on a dry basis (db).

<table>
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<tr>
<th>Sample</th>
<th>Temperature (°C)</th>
<th>Time (min)</th>
<th>Solid Load</th>
<th>Volatile Matter (%)</th>
<th>Fixed Carbon (%)</th>
<th>Ash (%)</th>
<th>C (%)</th>
<th>H (%)</th>
<th>N (%)</th>
<th>S (%)</th>
<th>O (%)</th>
<th>HHV (MJ/kg)</th>
<th>Product</th>
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<td>30</td>
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<td>63.8</td>
<td>8.8</td>
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<td>0.1</td>
<td>26.6</td>
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<td>50.36</td>
<td>47.43</td>
<td>2.21</td>
<td>73.0</td>
<td>7.0</td>
<td>5.2</td>
<td>0.3</td>
<td>17.1</td>
<td>31.0</td>
<td>Solid</td>
</tr>
</tbody>
</table>
**Tabel 4. Overview of the Dataset**

<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>Time (min)</th>
<th>Solid Load</th>
<th>Volatile Matter (%)</th>
<th>Fixed Carbon (%)</th>
<th>Ash (%)</th>
<th>C (%)</th>
<th>H (%)</th>
<th>N (%)</th>
<th>S (%)</th>
<th>O (%)</th>
<th>HHV MJ/kg</th>
</tr>
</thead>
<tbody>
<tr>
<td>Range</td>
<td>150–380</td>
<td>10–5760</td>
<td>0.047–0.47</td>
<td>33.6–94.7</td>
<td>1.8–47.43</td>
<td>2.21–55.8</td>
<td>27.8–74.2</td>
<td>2.7–8.97</td>
<td>0–5.17</td>
<td>0.05–0.77</td>
<td>7.8–49</td>
</tr>
<tr>
<td>Mean</td>
<td>251</td>
<td>986</td>
<td>0.19</td>
<td>66.49</td>
<td>21.11</td>
<td>11.36</td>
<td>58.64</td>
<td>6.34</td>
<td>1.23</td>
<td>0.18</td>
<td>25.85</td>
</tr>
<tr>
<td>Std</td>
<td>49</td>
<td>1985</td>
<td>0.11</td>
<td>19.07</td>
<td>14.32</td>
<td>15.82</td>
<td>12.55</td>
<td>1.58</td>
<td>1.58</td>
<td>0.19</td>
<td>11.24</td>
</tr>
<tr>
<td>5th–95th</td>
<td>162.5–365</td>
<td>12.5–5760</td>
<td>0.07–0.44</td>
<td>34.32–92.78</td>
<td>2.92–45.814</td>
<td>2.76–47.96</td>
<td>30.65–74</td>
<td>3.3–8.89</td>
<td>0–4.885</td>
<td>0.05–0.441</td>
<td>8.43–47.83</td>
</tr>
</tbody>
</table>

**Figure 3.** Variable Loadings (a and b) and Object Scores (c and d) Based on the First Three Principal Components. In c and d, Scores Were Colored Based on the Materials. In e and f, the Objects Were Classified Into Two Groups Based on the Product (Solid And Liquid), With Ellipses Denoting 95% Probability.
The compiled multivariate data were interpreted through PCA. PCA explains the maximum amount of variance with the fewest principal components. The input variables were changed into principal components that have linear combinations of the original variables. The maximum number of extracted components always equals the number of original variables.

In this research, three factor variables, i.e., temperature, time, and solid load, are compared with four response variables to obtain the principal component, representing $x_0 \cdots x_6$. All variables are independent from each other. The general PCA is described by the following equation:

$$X = \sum_{i=1}^{n} t_i p_i^T + E_n$$

where $X$ expresses a preprocessed data matrix containing an individual sample as row objects and measured variables $(x_0 \cdots x_6)$ as the corresponding columns. The vectors $t_i$ and $p_i$ describe the respective principal component scores and orthonormal variable loadings, respectively, while $E_n$ is the residual matrix after $n$ components.

4. Results and Discussion

Most of the hydrothermal experiments were performed within a temperature range of 162–365 °C, a holding time of 12.5–5760 min, and a solid load of 0.07–0.44. The product characteristics show variation in volatile matter in the range of 33.6%–94.7% on dry basis, and 90% of the observation was within 34.32%–92.78%. Fixed carbon, which plays an essential role in calorific value, takes place in a broader range of 1.8%–47.43%, with 90% of the observation within 2.92%–45.82%. Ash content also occupy a wide range between 2.21% and 55.8%, with major experiments obtaining a result between 2.762% and 47.96%. High heating value (HHV), which directly affects the waste-to-energy combustion performance, shows results within 12.64%–44.95% because 90% of the observation is within 13.17%–44.81%. The presence of outliers was further evaluated based on the determined principal components.

Principal components are useful to describe correlations and groupings in multivariate data and to reduce data dimensions for a more straightforward interpretation [49,50]. Furthermore, principal components can reduce data uncertainty from experimental errors by choosing the only components that could interpret the variation systematically. Three principal components, which have a total variation of 90.9% in the data, were chosen for the first dataset. As expected, the data seemed slightly noisy. Variable loadings and scores based on the first three principal components are presented in Figure 3.

As illustrated in Figure 3a, the first principal component shows 38.2% of the variation in the data and the effects of treatment temperature on HHV and carbon content. Increased treatment temperature has a close correlation with the increase of HHV. It also promotes the reduction of oxygen content, resulting in a lower O/C and H/C atomic ratio. Mason et al. also mentioned a strong positive correlation between HHV and carbon content, which revealed a correlation coefficient of 198.11 [51]. The second principal component shows the opposite effect of retention time and solid load toward carbon content and hydrogen content and covered 28.2% of the data variation (Figure 3a). Increasing holding time has a negative effect on carbon and hydrogen contents, while increasing the solid load increases carbon content and hydrogen content. For comparison, Figure 3b with total principal component of 62.7% shows positive correlation between temperature, HHV and carbon content.

By contrast, Funke et al. reported that higher solid loads decreased the carbon losses to the liquid, but produced a solid with a lower final carbon content in the comparison experiment of hydrothermal and vaporthermal [52]. Several correlations describing the effects of treatment temperature and retention time on degradation of organic components have been investigated [36,53]. The hydrothermal treatment reaction rate is exponentially dependent on retention time and directly dependent on on treatment temperature. From the first two principal components, no clear groupings are found in the properties of the solid product between different feedstock. As shown in Figure 3c, two outliers existed, namely, anaerobic digestion waste and mixed MSW from the USA.

The effects of solid load and retention time on oxygen content are presented in the third principal component, which covers 24.5% of data variation (Figure 3b). Both solid load and holding time have an adverse effect on the oxygen content. During the hydrothermal process, the O/C atomic ratio is gradually reduced at a longer reaction time [53]. A lower O/C atomic ratio means the decrease of oxygen content and/or the increase of carbon content. Figure 3d shows two separate groups of feedstocks. The larger group in the lower half includes products from material with a high percentage of hard cellulosic content, such as aseptic packaging waste, coffee ground, furniture waste, and chopstick waste. The smaller group in the upper half is composed of products from higher ash feed with heterogeneous composition, such as Indonesian MSW, USA MSW, paper waste, food waste, and anaerobic digestion waste. Figures 3e and 3f show the verification scores of the first three principal components, which were classified into two different groups on the basis of product type (solid and liquid). With 95% probability, both product groups show similarities, especially in aseptic packaging waste and furniture waste products. Both materials contain virgin and hard cellulose.
In terms of solid product target, an operating condition within 220–240 °C is enough to acquire good chars with a quality close to that of lignite. For targeting the liquid product, a higher temperature is necessary to maintain high pressure and subcritical condition. In this case, temperature above 240 °C is essential. Utilizing a higher temperature also means higher reactor pressure. The cost in real-scale application could increase exponentially as we increase the applicable pressure.

5. Conclusion

The use of hydrothermal treatment to prepare carbonaceous material for various purposes has been an interesting subject for many researchers in the past few decades. Hydrothermal carbonization has been proven to convert post-consumer waste materials into high calorific solid fuel. Reaction temperature, holding time, and reactor solid load are the important factors during hydrothermal treatment. Datasets from 10 different feedstocks within 29 experimental conditions were examined. The ultimate analysis, proximate analysis, and calorific value show a wide range with a deviation from 1.58 to 19.07. Increasing the reaction temperature could significantly increase the calorific value. Holding time and reactor solid load show different effects depending on the feedstock.

Previous hydrothermal research was limited to utilizing synthetic or homogeneous samples. Research on hydrothermal treatment of excavated waste from old landfills by using real extracted material has not been conducted yet. With a highly heterogeneous sample, such as excavated waste, conducting research to understand the behavior of composite material during hydrothermal treatment is necessary.

Given that hydrothermal treatment uses saturated steam as a medium, wastewater will be produced in the form of condensed water and slurry. Some minerals and metals leach out to those side products. The leaching behavior during hydrothermal treatment should be investigated further.

Understanding the leaching or diffusion effect of mineral constituents is necessary because it also directly influences the ash characteristics after the combustion process.

References