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# Conversion of Municipal Solid Wastes into Biochar through Hydrothermal Carbonization

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## Abstract

In this study, the hydrothermal treatment of municipal solid wastes (MSWs) for the production of biochar as a renewable solid fuel was investigated. The properties of surrogate MSWs and mixtures of newspaper and vegetables were greatly improved by hydrothermal treatment and were similar to those of coal-like fuel substances. Hydrothermal treatment increased the calorific value, the fixed carbon, and carbon contents. The composition of the major biomass components of MSW was found to affect the alternation of their physical and chemical properties significantly. These characteristic changes in pure cellulose, hemicellulose, and lignin were similar to those of coalification at the hydrothermal reaction temperature range of 150–280°C. The treated products became a solid fuel substance, the characteristics of which corresponded with fuel between lignite and sub-bituminous coal. The results of this study indicate that hydrothermal treatment can be used as an effective means to generate highly energy-efficient renewable fuel resources using MSWs.

**Keywords:** hydrothermal treatment, biochar production, biomass, municipal solid waste, renewable energy

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## 1. Introduction

Recently, to address increasing energy consumption and the steady depletion of fossil fuel reserve, many investigations have been conducted to develop alternative and renewable energy resources using biomass and waste [1, 2]. Because rapid urbanization is occurring almost everywhere in the world, substantially increasing municipal solid waste (MSW), which is of great global concern. As an effective means for treating MSW, the mechanical

biological treatment (MBT) system has received interest in Korea. MBT consists of two stages: mechanical treatment (MT) and biological treatment (BT). The MBT system was developed in Germany for waste processing [3]. In the first stage of this process (MT), MT is a size-based process that functions to remove all individual elements (regardless of size) that can be used for the production of refuse-derived fuel (RDF) (e.g., metal, plastic, paper, glass, and biodegradable materials). The larger MSW components are collected separately as combustible matter and then separated further to harvest materials for the production of refuse-derived fuel (RDF), which is used in energy generation. After the MT stage, the residual MSW is used for the production of organic fertilizers and biogas ( $\text{CH}_4$ ) in the BT stage. However, the BT stage has many problems, such as a long treatment period requirement, (e.g., more than 1 week or 1 month) and the emission of unpleasant odors [1, 3, 4].

Thermo-treatment concept is the main conversion technology to develop the BT stage, such as carbonization (400–500°C), pyrolysis (500–600°C), gasification (600–1000°C), and combustion (800–1000°C) to produce carbon-neutral energy from several kinds of biomass wastes. Hydrothermal treatment, which is a thermo-chemical conversion process that employs subcritical water (water heated to any temperature less than its critical temperature of 373°C under sufficient pressure to maintain the liquid state), functions by hydrolyzing biomass components that contribute greatly to the decomposition of structural biomass compound, the major constituents of biomass contained in MSW [5–9]. Therefore, the properties and drying performance of biomass as an energy resource can be improved significantly in a short time [7, 10, 11].

In the current research, we employed a pilot-scale hydrothermal treatment system to generate alternative solid fuel products from MSW using subcritical water (200°C, 1.6 MPa). The MSW samples tested in this study were collected from an MSW treatment facility in Korea. The samples were at the MT stage of the MBT system and were mainly composed of food residue (40–50%) and paper (30–40%)—Chung et al. [12]. They had a high moisture content of ~50–60% due to the food residue. The physical and chemical characteristics of the MT residue needed to be altered (dehydrated, compacted, and upgraded) for it to be used as a solid fuel, such as RDF. Thus, cellulose, hemicellulose, and lignin were used as surrogate MSWs for food as well as paper wastes. Then, the effects of hydrothermal treatment on the conversion of the biomass comprising the MSW samples were examined by varying the reaction temperatures in the range of 150–280°C, and the changes in the biomass characteristics were investigated.

## 2. Materials and methods

### 2.1. Materials

Food residue and paper content, which comprised the highest proportions in the composition of the MT residue obtained from an MBT system in Mokpo city, Korea, were evaluated because the composition of the MSW varies according to each season (e.g., food waste and

paper components can fluctuate between 70 and 80% of total MSW) [12], surrogate MSW (SM) residues, which were prepared using newspaper and Korean Kimchi instead of paper and food waste, were mixed at two different ratios (SM 1 and SM 2) after the crushing process: SM 1 = 5:5 and SM 2 = 3:7 (waste paper:Kimchi (wet, w/w)). **Table 1** shows the properties of these surrogate MSWs, including the results of proximate and ultimate analysis and the calorific values. In addition, pure cellulose ( $\alpha$ -cellulose-fiberform, Nacalai Tesque Inc., Kyoto, Japan), xylan (Beechwood, SIGMA) which are the main components of hemicellulose [13], and lignin (Kanto Chemical Co., Inc., Japan) were also tested to investigate the effects of hydrothermal conversion on these materials.

	Paper	Kimchi	Raw surrogate MSW		Hydrothermally treated MSW	
			SM 1	SM 2	SM 1	SM 2
Moisture (a.r.)	2.3	92.4	47.4	65.4	66.3	79.6
Moisture <sup>1</sup>	–	–	36.8	54.2	12.6	17.3
Proximate analysis (wt.%, d.b.)						
Volatile matter	87.0	67.1	87.1	83.1	75.3	74.2
Fixed carbon	5.3	22.6	6.0	8.3	15.0	13.3
Ash	7.7	10.3	6.9	8.7	9.7	12.5
Ultimate analysis (wt.%, d.b.)						
C	40.3	33.6	37.0	35.6	43.5	41.7
H	5.6	5.3	5.4	5.4	5.7	5.3
N	0.2	3.5	1.9	2.6	1.1	0.4
O	46.4	47.3	48.8	47.7	40.0	40.1
Calorific value (MJ/kg, d.b.)	15.5	14.7	15.1	14.9	17.2	16.1

After natural drying for 24 h.  
a.r., as received.  
d.b., dry basis.

**Table 1.** Properties of raw and treated surrogate MSWs by hydrothermal treatment.

## 2.2. Pilot-scale hydrothermal treatment system

Hydrothermal treatment experiments were performed using a 200 L pilot-scale reactor (**Figure 1(a)**). The reactor consists of a steam boiler and a steam condenser. For all of the experiments, 20 kg of surrogate MSW was supplied to the reactor. The operating temperature of the hydrothermal treatment was set at 200°C, with a pressure of 1.6 MPa, and the reaction was carried out for 60 min. After the hydrothermal reaction was completed, the residual steam was discharged, and the products were collected from the reactor.

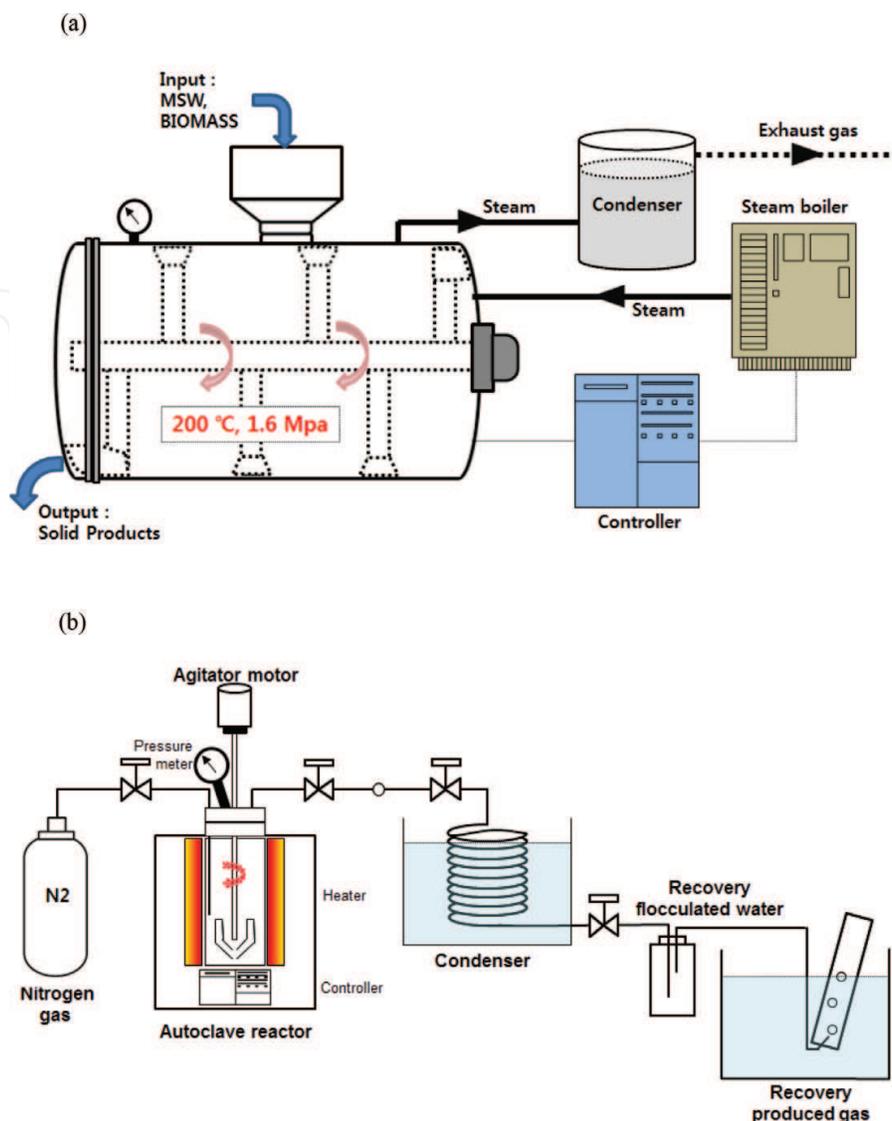


Figure 1. Schematics of (a) pilot- and (b) lab-scale hydrothermal treatment systems.

### 2.3. Lab-scale hydrothermal treatment reactor

A laboratory-scale hydrothermal treatment reactor was used to investigate the effects of hydrothermal treatment on the characteristic changes in pure cellulose, hemicellulose, and lignin. The experiments were performed using a 500 mL autoclave reactor (Figure 1(b)) consisting of a reactor body, heater, and steam condenser and operated under  $N_2$  gas. A compound sample (20 g) was mixed with an equal amount of water and loaded into the reactor. The operating temperatures and pressures ranged from 150 to 280°C and 1.3 to 5.5 MPa, respectively, and the reaction time was 30 min. The components in the reactor were mixed vigorously using an agitator rotating at 200 rpm.

### 2.4. Analytical procedures

Elemental composition analysis of surrogate MSWs, cellulose, hemicellulose, lignin, and their solid products were carried out using a PerkinElmer 2400 Series II CHN organic elemental

analyzer (PerkinElmer, Waltham, MA, USA). Proximate analysis was conducted using a SHIMADZU D-50 simultaneous TGA/DTA analyzer. Calorific values were determined using a bomb calorimetric standard method according to JIS M-8814 (JPN ISO1928:1995). The biomass compositions of the newspaper and Kimchi were analyzed by Nihon Hakko Shiryo Company (Kawasaki, Japan). The biomass compositions of the cellulose, hemicellulose, and lignin are defined in Eqs. (1)–(3) [14, 15].

$$\text{Hemicellulose(\%)} = \text{NDF} - \text{ADF} \quad (1)$$

$$\text{Cellulose(\%)} = \text{ADF} - \text{ADL} \quad (2)$$

$$\text{Lignin(\%)} = \text{ADL} \quad (3)$$

where NDF is neutral detergent fiber, ADF is acid detergent fiber, and ADL is acid detergent lignin.

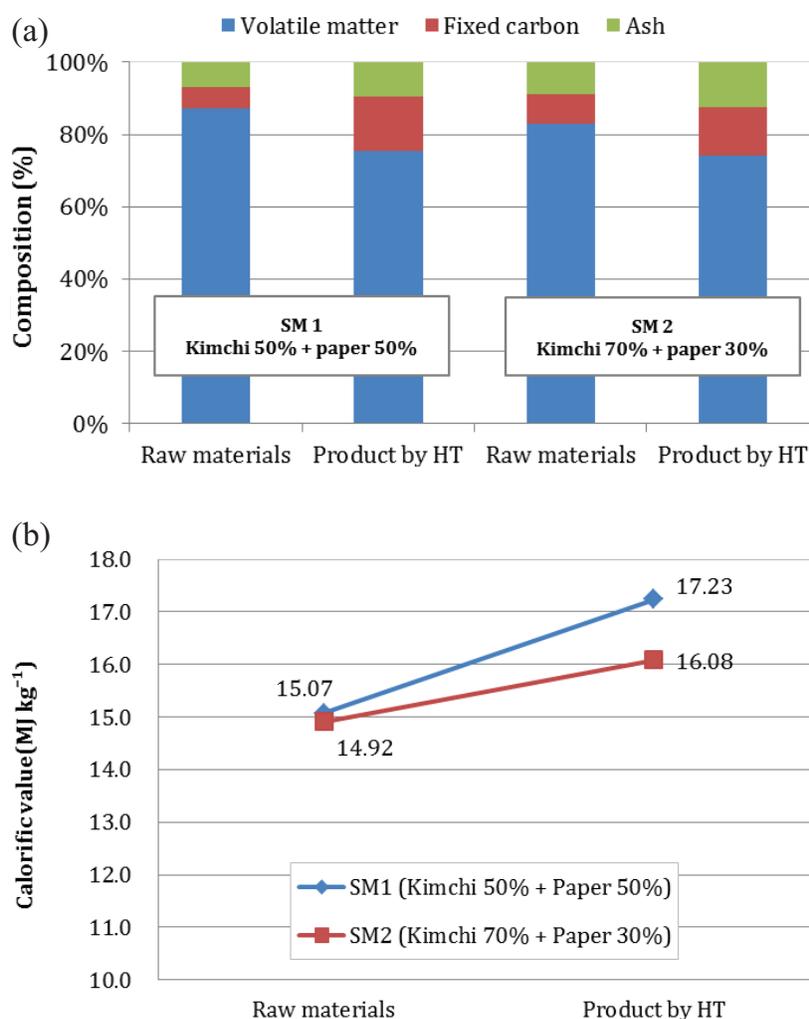
### 3. Results and discussion

#### 3.1. Properties of surrogate MSWs after hydrothermal treatment

**Figure 2** shows the biochar products from MSW. Furthermore, **Table 1** and **Figure 3(a)** show the properties of raw and treated SM 1 and SM 2 using the pilot-scale hydrothermal treatment reactor operated at 200°C.



**Figure 2.** Biochar products process from waste by hydrothermal carbonization; (a) surrogated municipal solid waste, (b) pilot-scale hydrothermal treatment systems, and (c) biochar products.



**Figure 3.** Enhancement of properties of surrogate MSWs by hydrothermal treatment; (a) the proximate analysis and (b) the calorific values.

The moisture content in the raw SM 1 and SM 2 samples, decreased significantly after 24 h of natural drying. However, the moisture content of the treated SM 1 and SM 2 samples decreased from 66.3 to 12.6% and 79.6 to 17.3%, respectively, after 24 h of natural drying. Hydrothermal treatment is known to dissociate physical and chemical structures of biomass in surrogate MSWs [5, 16]; therefore, the biomass in MSWs is decomposed into small and simple molecules. Our results demonstrated that hydrothermal treatment improved the drying performance of the surrogate MSWs. Additionally, the chemical properties of the surrogate MSWs were changed using the hydrothermal treatment. The raw SM 1 and SM 2 samples contained high volatile matter (87.1 and 83.1%, respectively) and oxygen (48.8 and 47.7%, respectively) contents, which are similar to the volatile matter and oxygen contents of other biomass materials [13, 17, 18]. After the hydrothermal treatment, the volatile matter and oxygen contents decreased, whereas the fixed carbon content increased from 6.0 to 15.0% for SM 1 and from 8.9 to 13.3% for SM 2 via hydrolysis reactions (**Figure 3a** and **b**) shows the calorific values of the surrogate MSWs (SM 1 and SM 2) before and after the hydrothermal treatment. After the hydrothermal treatment, the calorific values of SM 1 and SM 2 samples increased

from 15.1 to 17.2 MJ/kg and 14.9 to 16.1 MJ/kg, respectively, which resulted in an increase of fixed carbon content. The increase in the calorific value of SM 1 was greater than that of SM 2 due to the higher paper content in the MSWs. This result suggests that the hydrothermal treatment is more effective at producing an upgraded solid fuel from MSW with high paper content.

### 3.2. Effect of hydrothermal treatment on upgrading of biomass in MSW

The surrogate MSW was prepared by blending newspaper and Kimchi. The content of the three major biomass components (cellulose, hemicellulose, and lignin) in the MSW samples was considered to be an important factor affecting the performance of the hydrothermal treatment. The characteristic changes of these components were analyzed. Additionally, the effects of hydrothermal treatment on the calorific values of cellulose, hemicellulose, and lignin were also examined.

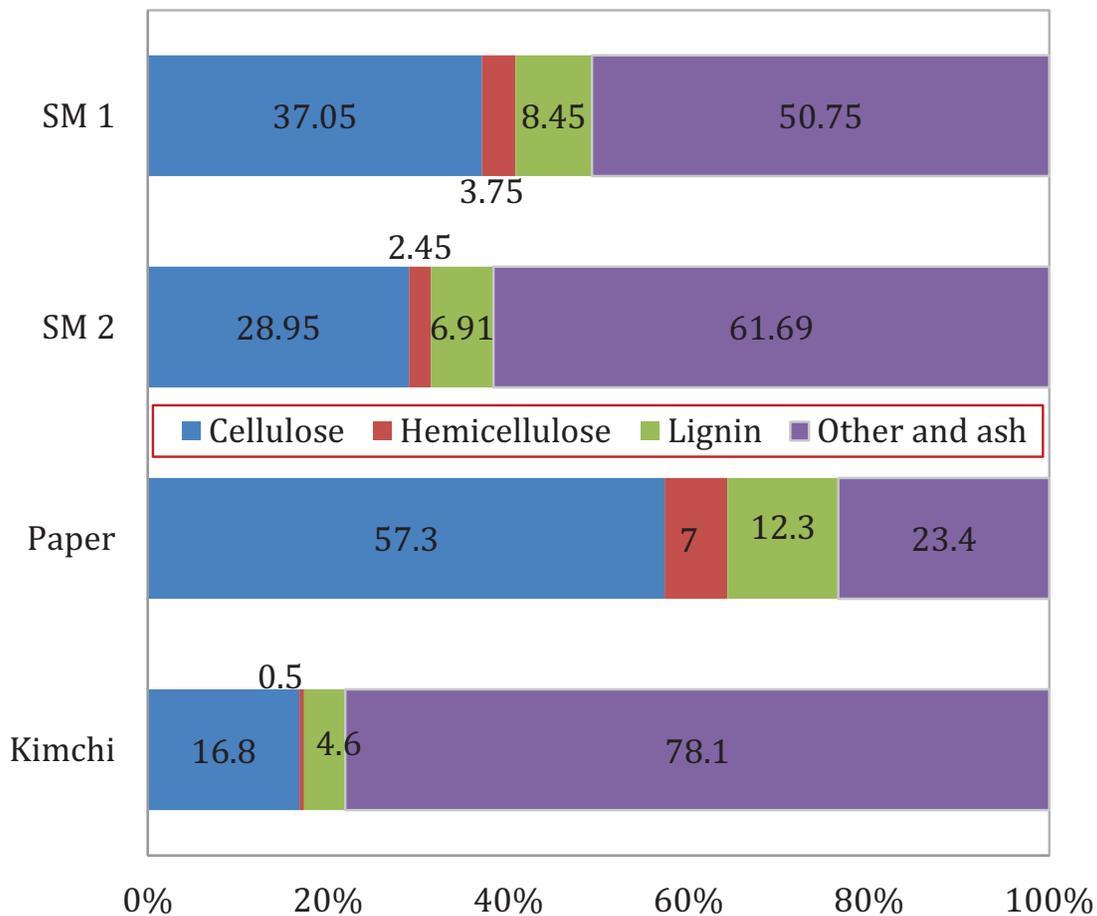


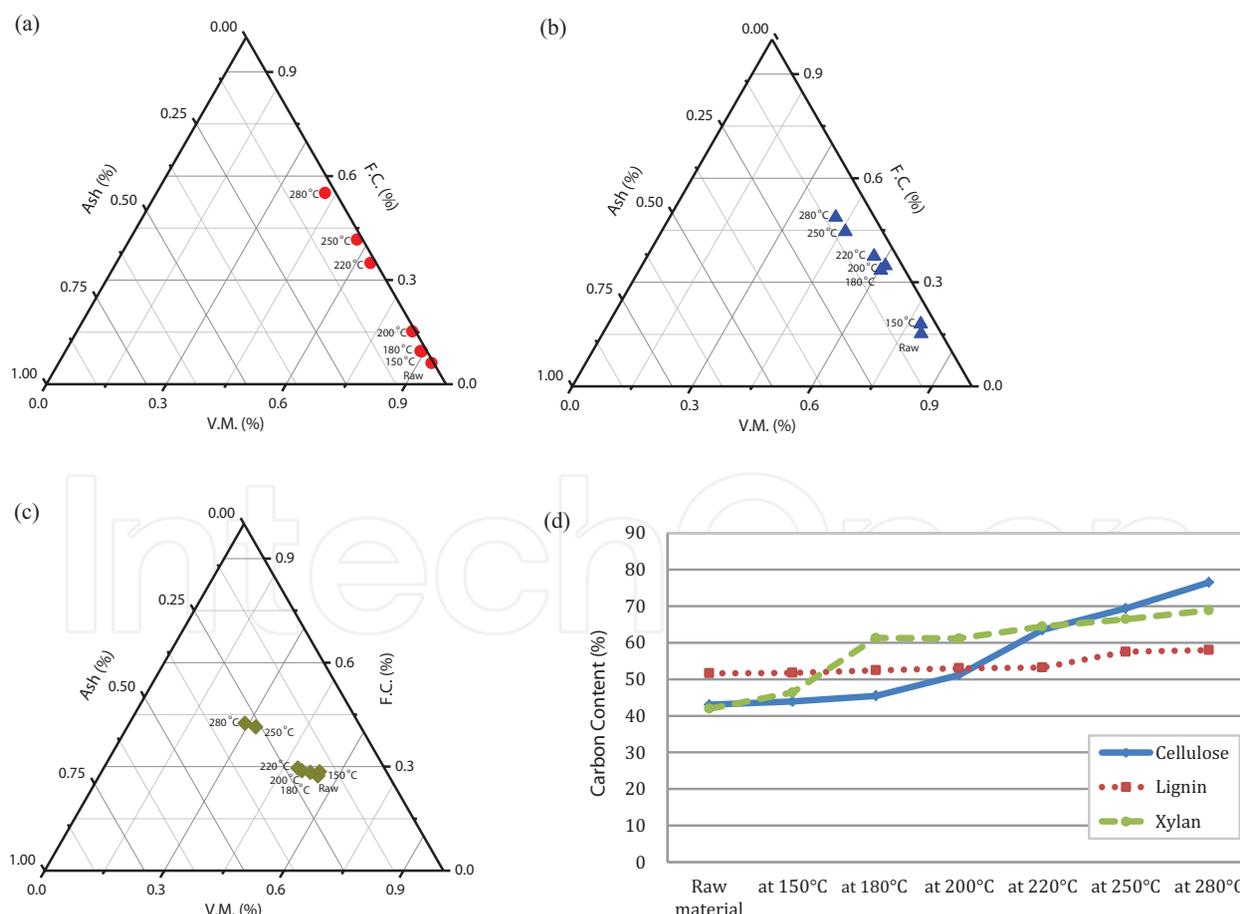
Figure 4. Biomass composition in the surrogate MSWs and their comparison with those of newspaper and Kimchi.

### 3.2.1. Biomass composition of MSW

**Figure 4** shows the composition of the biomass components comprising the surrogate MSW. Dried newspaper was composed of 57.2 cellulose, 12.3 lignin, and 6.9% hemicellulose. Dried Kimchi was composed of 16.8 cellulose, 4.6 lignin, and 0.5% hemicellulose. Therefore, the total cellulose, lignin, and hemicellulose content of the newspaper and Kimchi were 76.5 and 29.1%, respectively. The total cellulose, lignin, and hemicellulose content in the biomass of the SM 1 and SM 2 samples were 49.4 and 38.4%, respectively. Specifically, the SM 1 and SM 2 samples contained 37.1 and 29.0% cellulose, 8.5 and 6.9% lignin, and 3.8 and 2.5% hemicellulose, respectively (**Figure 4**). The composition of biomass components (cellulose, hemicellulose, and lignin) of the MSW was found to influence the properties of the hydrothermal products.

### 3.2.2. Changes in the properties of biomass components

Hydrothermal treatment changed the properties of cellulose, hemicellulose, and lignin.



**Figure 5.** The changing characteristics of biomass components by hydrothermal treatment; (a) cellulose, (b) hemicellulose, (c) lignin, and (d) carbon content.

<i>Cellulose</i>							
	Raw cellulose	At 150°C	At 180°C	At 200°C	At 220°C	At 250°C	At 280°C
Proximate analysis (wt.%, d.b.)							
Volatile matter (d.b.)	93.4	88.9	89.3	84.0	63.7	56.9	42.2
Fixed carbon (d.b.)	6.1	9.6	9.5	15.2	35.0	41.7	55.1
Ash (d.b.)	0.5	1.6	1.3	0.8	1.4	1.4	2.7
Ultimate analysis (wt.%, d.b.)							
C	43.0	43.9	45.5	51.2	63.5	69.4	76.5
H	6.4	6.5	6.0	5.7	4.7	4.6	4.5
N	0.0	0.0	0.0	0.0	0.0	0.0	0.0
O	50.1	48.0	47.2	42.3	30.4	24.6	16.3
Calorific value (MJ/kg, d.b.)	16.5	16.6	18.9	23.0	26.5	26.8	27.7
<i>Hemicellulose</i>							
	Raw xylan	at 150°C	at 180°C	at 200°C	at 220°C	at 250°C	at 280°C
Proximate analysis (wt.%, d.b.)							
Volatile matter (d.b.)	79.8	78.3	60.5	61.0	56.8	46.1	41.6
Fixed carbon (d.b.)	15.2	17.9	33.5	34.8	37.5	44.6	48.7
Ash (d.b.)	5.1	3.8	6.0	4.3	5.7	9.4	9.7
Ultimate analysis (wt.%, d.b.)							
C	41.9	46.4	61.3	61.2	64.5	66.5	68.9
H	6.0	5.3	5.2	5.0	5.0	5.0	4.9
N	0.0	0.0	0.0	0.0	0.0	0.0	0.0
O	47.0	44.5	27.5	29.5	24.8	19.1	16.5
Calorific value (MJ/kg, d.b.)	13.9	14.9	21.5	22.6	24.9	25.6	26.5
<i>Lignin</i>							
	Raw lignin	at 150°C	at 180°C	at 200°C	at 220°C	at 250°C	at 280°C
Proximate analysis (wt.%, d.b.)							
Volatile matter (d.b.)	54.8	54.6	52.4	50.1	48.7	32.2	28.9
Fixed carbon (d.b.)	27.4	28.6	28.4	28.8	29.6	41.5	42.5

<i>Cellulose</i>							
Ash (d.b.)	17.8	16.8	19.2	21.1	21.7	26.3	28.6
Ultimate analysis (wt.%, d.b.)							
C	51.6	51.8	52.5	53.0	53.2	57.5	58.0
H	4.3	4.4	4.2	4.1	3.9	3.5	3.4
N	0.0	0.0	0.0	0.0	0.0	0.0	0.0
O	26.3	26.0	24.1	21.8	21.2	12.7	10.0
Calorific value (MJ/kg, d.b.)	20.4	20.1	21.8	22.8	23.1	25.2	26.0

d.b., dry basis.

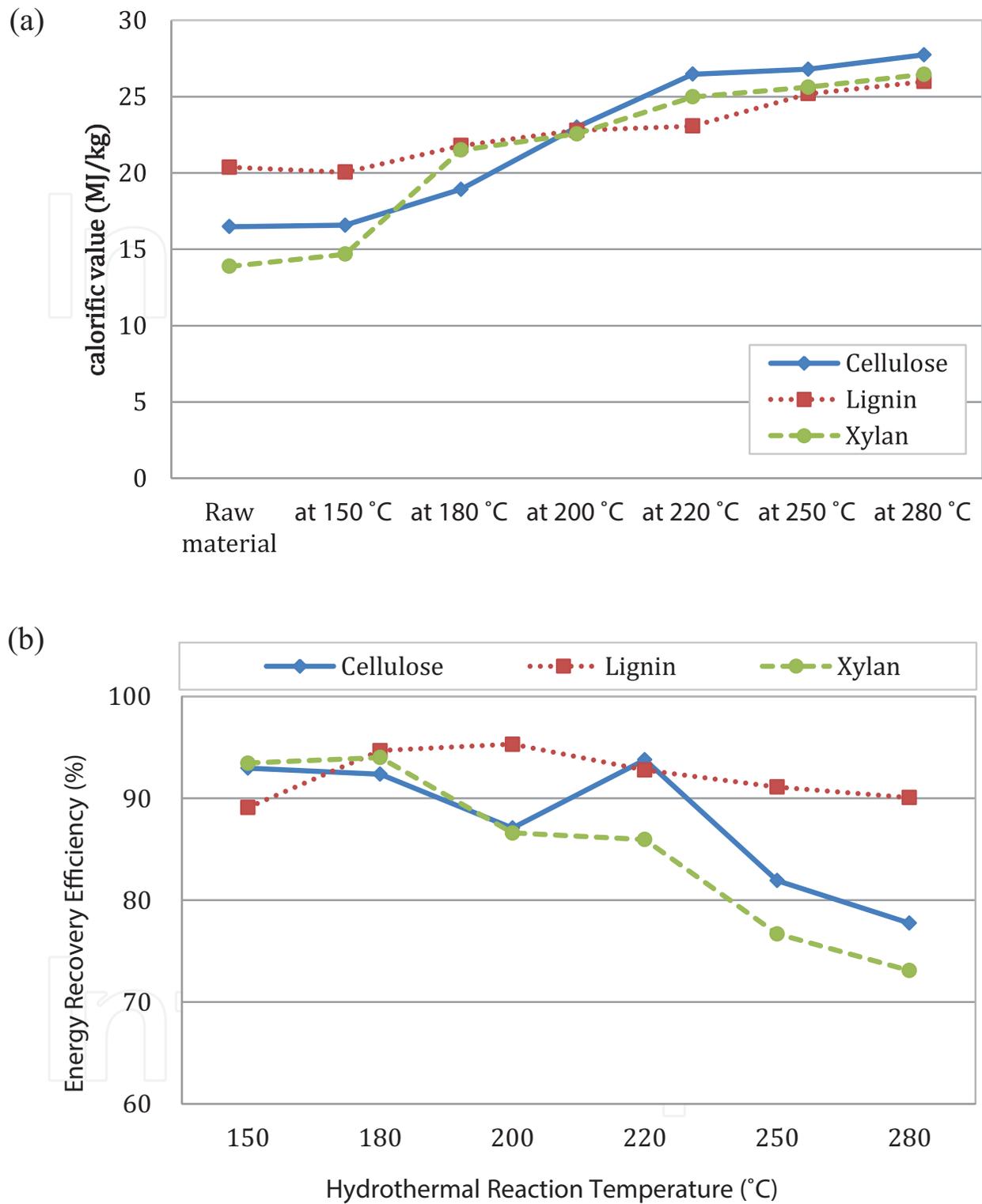
**Table 2.** Effect of hydrothermal treatment on changes in the properties of cellulose, hemicellulose, and lignin.

Along with the results of the ultimate analysis, **Figure 5** and **Table 2** show the results of the proximate analysis of the biomass components by varying the hydrothermal reaction temperature. The fixed carbon content of cellulose increased from 6.1 to 35.0% in response to hydrothermal treatment at 220°C (**Table 2** and **Figure 5(a)**). This result suggests that the cellulose began to decompose at 220°C. When xylan was used as a hemicellulose, the fixed carbon and carbon contents increased from 15.2 to 33.5% and from 41.9 to 61.3%, respectively, at 180°C (**Figure 5(b)**). Below 180°C, the compositions of these products were not different from those of the raw material. This result is not surprising because the hydrolysis of hemicellulose occurs at 180°C [6, 11]. As the fixed carbon content and carbon contents increased due to the hydrothermal treatment, the calorific value increased. However, the results for lignin were different from those for cellulose and hemicellulose. Lignin started to decompose at temperatures exceeding 250°C (**Figure 5(c)**). This can most likely be attributed to the decomposition or pyrolysis of lignin at temperatures slightly below 250°C.

As a result, the increase in the fixed carbon content of the surrogate MSWs (SM 1 and SM 2) was influenced by the increase in the fixed carbon content of cellulose and hemicellulose (**Figure 5(d)**). Additionally, the ash content of lignin was higher than that of cellulose, indicating that lignin possesses higher ash content.

### 3.2.3. Changes in calorific value and energy recovery efficiency

**Figure 6(a)** shows the calorific values of cellulose, hemicellulose, and lignin. Pure lignin had a higher calorific value (20.4 MJ/kg) than pure cellulose (16.5 MJ/kg) and hemicellulose (13.9 MJ/kg). When cellulose was treated by hydrothermal treatment, the calorific values increased to 18.9, 23.1, 26.5, and 27.7 MJ/kg at 180, 200, 220, and 280°C, respectively. When xylan was used as a hemicellulose, the calorific value increased from 13.9 to 21.5, 22.6, 25.0, and 26.5 MJ/kg at 180, 200, 220, and 280°C, respectively. However, the calorific value of lignin started to increase from 20.4 to 25.2 MJ/kg at approximately 250°C. The increase in the calorific values for SM 1 and SM 2 was influenced by their cellulose and hemicellulose content. As reaction temperatures increase, the calorific value of solid products increases, but the amount of solid product decreases due to



**Figure 6.** Alternation of properties in biomass components by hydrothermal treatment; (a) the calorific values and (b) energy recovery efficiency (ERE).

chemical dehydration and decarboxylation (i.e., removal of  $\text{CO}_2$ ) [7, 18–21]. Therefore, in hydrothermal reactions, an optimum temperature should be maintained to maximize the energy recovery efficiency (ERE) [22–24]. The ERE is an important parameter for evaluating the effect of hydrothermal treatment on solid fuel production (**Figure 6(b)**). The ERE is defined by Eq. (4):

$$\text{ERE} = \frac{(\text{calorific value of solid product} \times \text{mass of dried solid product})}{(\text{calorific value of raw materials} \times \text{mass of dried raw material})} \quad (4)$$

The highest ERE of each material indicates the optimum reaction temperature for hydrothermal treatment. Cellulose decomposition began at 220°C, and the optimum reaction temperature used to maximize the ERE was 220°C (ERE = 93.8%). When xylan was used as a hemicellulose, it began to decompose at 180°C, and the optimum reaction temperature was 180°C (ERE = 94.0%). Lignin began to decompose at 250°C, and the maximum ERE was 95.3% at a reaction temperature of 200°C.

These results suggest that the optimum temperature of hydrothermal treatment to produce more energy-rich solid fuel is approximately 200°C. Fundamentally, pure lignin has a high calorific value (20.4 MJ/kg) and constitutes approximately 4–12% of solid fuel sources. When treated at 200°C, the calorific values of cellulose and hemicellulose were improved and the total remaining solid mounts were increased. Consequently, the optimum ERE of the MSWs and biomass products after hydrothermal treatment was obtained.

### 3.3. Mechanism of hydrothermal treatment for upgrading solid products

The results indicated that the hydrothermal treatment induce hydrolysis, chemical dehydration, and decarboxylation reactions (Figure 7). Hydrolytic reactions caused by 1 mol of water cleave cellulose and hemicellulose at ester and ether bonds of cellulose and hemicellulose into smaller molecules [6, 7, 11, 25]. This hydrolysis reaction can complete the conversion of biomass within a few reaction cycles. Furthermore, the fuel properties of the products generated

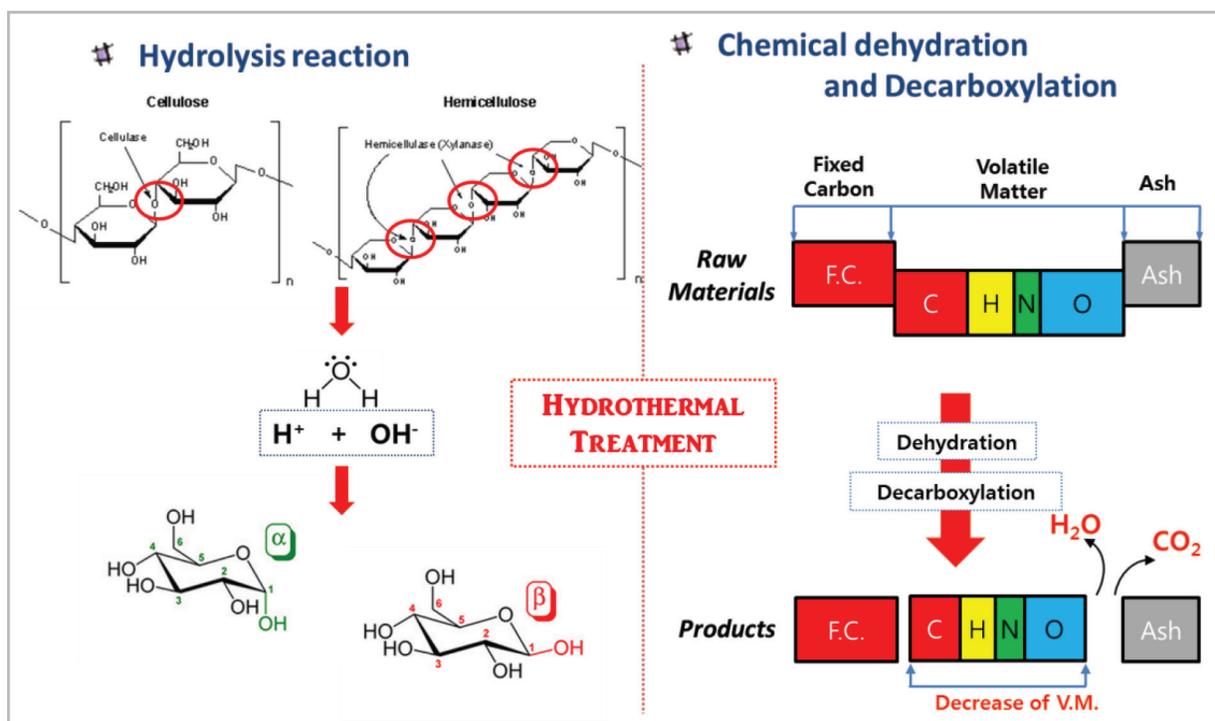


Figure 7. Mechanism of hydrothermal treatment to upgrade solid products.

from biomass (i.e., cellulose, hemicellulose, and lignin) have been shown to be upgraded via chemical dehydration and decarboxylation reactions that released H<sub>2</sub>O and CO<sub>2</sub> [6, 7, 11, 26]. Along with a loss in weight, these reactions caused a decrease in volatile matter and an increase in carbon content in the biomass products compared with the raw materials. These effects can be utilized for the drying and carbonization of biomass into an alternative fuel.

### 3.4. Coalification band of hydrothermal products

Hydrothermal treatment can upgrade the properties of the biomass components of MSWs in a manner similar to the coalification process. The coalification bands of raw and treated surrogate MSWs (SM 1 and SM 2) were compared with the coalification band of pure cellulose, hemicellulose, lignin, and various types of coal (**Figure 8**). MSW is known to have high H/C and O/C ratios, which is similar to those of cellulose and other biomass materials [6, 17, 18, 21]. The H/C and O/C ratios of SM 1, SM 2, cellulose, hemicellulose, and lignin decreased with the coalification status between lignite and sub-bituminous coal. This occurs when the biomass components of MSW are converted into carbonaceous products by chemical dehydration reactions during hydrothermal treatment ( $4(C_6H_{10}O_5)_n \Leftrightarrow 2(C_{12}H_{10}O_5)_n + 10H_2O$ ) [6, 10, 11, 18, 20]. Due to dewatering, dehydration, and decarboxylation, hydrothermal reactions can enhance biomass properties by reducing the hydrogen and oxygen contents of reaction products, resulting in increased calorific values of biomass products.

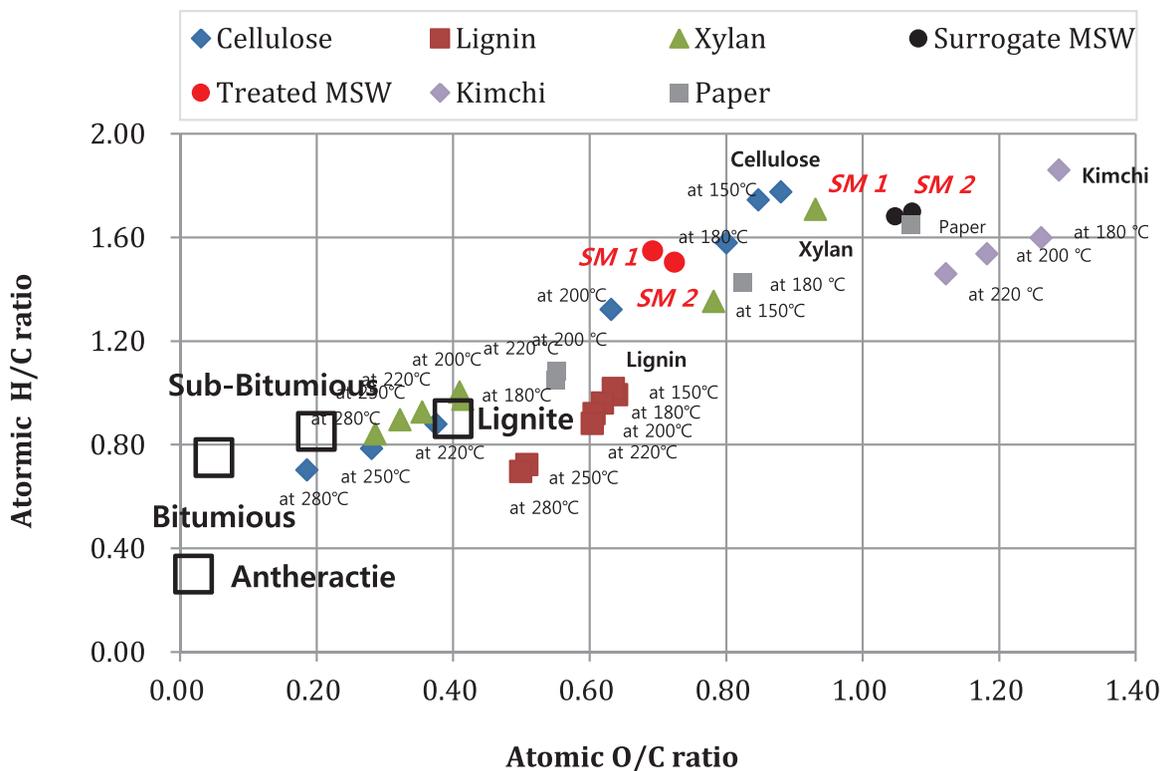


Figure 8. Comparison of coalification bands of raw and treated surrogate MSWs.

## 4. Conclusion

The effects of hydrothermal treatment on the properties of MSWs were investigated for their conversion into fuel products with high-energy efficiencies. After the hydrothermal treatment, the surrogate MSWs containing high paper content demonstrated significant increases in their carbon content and calorific values. Therefore, cellulose, hemicellulose, and lignin that constitute the MSWs as biomass were used to investigate the effects of the reaction temperature. The optimum reaction temperature for a mixture of cellulose, hemicellulose, and lignin was found to be approximately 200°C. As a result, the status of the treated products corresponded with solid fuels between lignite and sub-bituminous coal.

## Author details

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