Energy Recycling from Sewage Sludge by Producing Solid Biofuel with Hydrothermal Carbonization

Peitao ZHAO^{1,2}, Yafei SHEN², Shifu GE¹, Kunio YOSHIKAWA²

¹Key Laboratory of Energy Thermal Conversion and Control of Ministry of Education, School of Energy
and Environment, Southeast University

NO.2 Sipailou, XuanWu District, Nanjing 210096, P.R. China

²Department of Environmental Science and Technology, Tokyo Institute of Technology

G5-8 4259 Nagatsuta-cho, Midori-ku, Yokohama, Kanagawa 226-8503, Japan

E-mail address: pt.zhaoseu@gmail.com (ZHAO) yafeisjtu@gmail.com (Shen)

TEL: +81-45-924-5586 FAX: +81-45-924-5586

ge1962@126.com (GE)

TEL: +86-25-83792379 FAX: +86-25-5771-4489

Abstract: The hydrothermal (HT) conversion has been proposed to produce nitrogen, chlorine free solid biofuel or liquid fertilizer from high moisture and nitrogen content bio-wastes, such as municipal solid waste (MSW), mycelial waste, sewage sludge and paper sludge. However, the energy and economic efficiency of this process has not been fully investigated yet. This work focuses on energy recycling from sewage sludge by producing solid biofuel with HT carbonization, in order to optimize the operating parameters and evaluate the energy efficiency of this fuel production process. The effect of the HT temperature and holding time on the biofuel recovering ratio, calorific value and energy recovery rate was investigated. This evaluation fully considered the effect of the HT conditions, mechanical dewatering, thermal drying, and biofuel recovery ratio. Moreover, the energy consumption of sludge thermal drying was introduced to illustrate the economic efficiency of the HT biofuel production process more intuitively. The results show that the HT biofuel production process was more cost-effective than the conventional thermal drying. The HT temperature was the most important parameter to affect the biofuel properties. The carbon content of solid biofuel kept increasing both with HT temperature and holding time, resulting in an increase in the calorific value of biofuel; whereas, the biofuel recovering ratio α , defined as the mass ratio of solid biofuel to raw sludge, also dropped causing a reduction in the energy recovery rate. After the HT temperature was above 200 °C, the energy recovery rate was around 40%. A moderate condition–HT temperature of 200 °C and holding time of 30 min was suggested to produce solid biofuel from

sewage sludge with an energy recovery rate of 50%. Practically, it is better to improve the intensity of mechanical dewatering to remove more water from the HT products in order to improve thermal efficiency.

Keywords: sewage sludge, hydrothermal carbonization, solid biofuel, biofuel recovery, energy efficiency

1. Introduction

Municipal wastewater treatment results in generating huge amount of sewage sludge, which requires proper and environmentally acceptable management before final disposal. The management of municipal sewage sludge (SS) is a difficult and expensive problem to solve for many countries. In China, about 20.76 million tons of SS with a water content of 80% has been produced in 2010 [1]. Moreover, this amount will be much huger at the end of 2015 according to the China's 12th Five-Year Construction Plan for National Urban Wastewater Treatment and Recycling Facilities [2], indicating that another sludge treatment facilities with a capacity of at least 5.18 million tons dried sludge has to be built to meet the environmental requirements in the following years. In a global context, it is organizationally, technically, and economically hardly possible to prevent or strongly reduce the amount of municipal wastewater because of the rapid urbanization, industrialization, and growth in population [3]. Besides that, the presence of toxic pollutants in municipal wastewater cannot be avoided because a large part of these toxics originates from diffuse sources. Accordingly, it is believed that the sludge output would remain increasing gradually in the coming decades [4], and its quality will not change significantly in the future. SS will therefore remain a permanent waste treatment problem requiring an appropriate solution [5].

Currently, bio-energy is an excellent energy recycling technology with bright prospect, giving its ability to recover energy from waste biomass [1, 6, 7]. The hydrothermal (HT) conversion, which involves the application of heat and pressure to treat biomass in an aqueous medium, was verified as an effective way to densify the energy content of moist biomass without thermal drying [8]. For sludge, it aims to disintegrate the sludge and result in the formation and accumulation of dissolved products [9]. During the reaction, solid decomposition was significantly affected by reaction temperature rather than reaction time. Higher temperature resulted in higher solids conversion to solid biofuel [10].

The HT conversion has bright perspective with three main merits: 1) largely improving the dewaterability; 2) dramatically reduction in volume, especially for the municipal solid waste (MSW), sawdust, SS and leaves; 3) energy densification. Energy contents of the solid biofuel from primary SS carbonized at 140–200 °C for 4 hours ranged from 21.5 to 23.31 MJ/kg, and kept increasing with carbonization [10]. Liu *et al.* [11] also stated that the energy densification was increased from 1.34 to 1.66 and 1.33 to 1.55 for coconut fiber and eucalyptus

leaves, respectively. The highest energy yield was achieved at the lowest temperature adopted because of the relative high biofuel yield. Lu *et al.* [12] applied HT to produce powder-like solid biofuel from Japanese MSW, Indian MSW and Chinese MSW, at 220 °C and 2.4 MPa for 30 min with a lab-scale autoclave facility. It is reported that the volume based heating value (HV) of MSW was improved at around 6.4–9.0 times. Similar results were also obtained by deriving the data presented by Prawisudha *et al.*, [13] indicating that this volume based energy density was improved around 4–5 times after the HT pretreatment in a pilot plant. The product was easily dryable to powdery fuel with a moisture content of 10% and an average dry basis (d.b.) HV of 20 MJ/kg, as high as that of the low-grade sub-bituminous coal. Moreover, they also observed that the chlorine content of MSW could be reduced from 10,000 ppm (d.b.) to approximately 2,000 ppm (d.b.) resulting from the transformation to water-soluble inorganic chlorine. For the combustion characteristics, it was stated that the blending of HT pretreated MSW improved devolatilization properties of coal and lowers the ignition temperature of coal [12, 14, 15]. He *et al.* [16] also state that the combustion of solid fuel produced from SS by HT is expected to be easier and more stable than raw sludge because of lower activation energy and pre-exponential factor. The solid fuel is supposed to be clean as about 60% of nitrogen and sulfur within SS can be removed during the HT conversion. Moisture content was found to affect the HT carbonization process; feedstocks with higher initial moisture content resulted in lower hydrochar yield [10].

Most of these works have focused on the feasibility of applying HT to produce solid biofuel [11, 13, 17-19], the kinetics and mechanism of HT solid fuel formation [10, 20], the biofuel' characterization [16, 21, 22], and the biofuels' combustion characteristics [7, 12, 14-16]. They have distinctly verified that the HT was an effective pretreatment process to produce clean solid biofuel from waste biomass by controlling the water content without prior drying. However, few of them focused on the energy and economic efficiency of this process by fully considering the HT operating conditions, dehydration performance, consequent thermal drying and biofuel recovery ratio, etc., which are very crucial for process optimization and commercialization of this technology. Therefore, this study mainly investigated on the mass and energy balance of solid biofuel production from SS by HT. The effect of HT temperature and holding time on the biofuel recovering ratio, calorific value of solid biofuel, water removal performance, and energy recovery rate were fully taken into account to evaluate the energy efficiency.

2. Material and Methods

2.1 Experimental Procedures

In this study, the dewatered activated sludge with a moisture content of (85.94±0.22)% was taken from a

wastewater treatment plant in Japan . Its proximate and ultimate analysis results were shown in Table 1.

Fig.1 shows a diagram of the HT solid biofuel production process and an elementary diagram of the experimental facilities. During the HT treatment, the sludge pre-mixed with pure water (Wako Pure Chemical Industries, Ltd. Japan) as the mass ratio (wet basis, w.b.) of 2:1 (90 g sludge and 45 g pure water) was loaded into a glass tube with a volume of 500 mL. Subsequently, the glass tube was put into the reactor, which was heated by an electronic heat jacket. After that, the reactor was sealed and the argon with a purity of 99.999% was supplied from a cylinder to the reactor in order to create an oxygen free circumstance. The reactor was then heated up to the target temperature and kept constant for a predetermined period. In this study, the HT temperature and holding time ranged from 180 to 240 °C and from 15 to 45 minutes, respectively. During the reaction, the mixer was kept stirring with its direction switching every 5 minutes to ensure the uniformity of reaction temperature in the samples. After finishing the reaction, the heater was turned off and the residual steam was discharged and cooled down with a water condenser. When the pressure and temperature fell down to the atmospheric and room temperature, the products were taken out from the glass tube and then kept in a bottle for further use. The condensed liquid was collected and mixed with the products from the glass tube. Each condition was repeated at least 3 times as the same procedure and the final products were mixed to reduce the experimental error.

Fig.1 Biofuel production process and an elementary diagram of experimental facilities

To calculate the recovery rate α of the solid fuel, about 60 g (dividing into three group) of the pre-mixed products was directly dried in an oven at 105 °C. And another 140 g was taken out to conduct the mechanical dewatering in order to evaluate the water removal efficiency of sludge. An elementary diagram of the dewatering device was also shown in Fig.1 (details about this device can be found in Zhao *et al.*, [1]). Differing from the previous one [1], the dewatering pressure and time were 0.6 MPa and 12 minutes respectively. During the dewatering process, the filter was collected by a beaker and its weight was measured by an electronic balance connecting to a computer. After that, the re-dewatered sludge was dried by a forced convective dryer, whose operating condition was air temperature of 30.4±0.4 °C and velocity of 1.34±0.04 m/s all the time. During the drying process, the sample weight was continuously measured by an electronic balance ASP-4100 (AS-one, Japan) and was recorded in a computer with .XLS format at 6 minutes interval for further analysis. The drying process stopped after the sample weight was kept constant for 2 h.

2.2 Analytic Methods

The variation of the dry basis moisture content (d.b. kg/kg), which represents the amount of water contained in 1 kg dry solid, was used to characterize the water removal ability. This value can be easily solved according to the weight loss of the sample.

Fig.2 shows the boundary of the mass and energy balance in the current study. The solid recovering rate α was expressed as the ratio of the HT solid product to the raw sludge (d.b.). The proximate and ultimate analysis was used to characterize the solid biofuel property. The heat consumption in the HT was evaluated according to a batch-type HT equipment composing of a pressurized vessel, a boiler, a control unit, piping and incidental components. The vessel was made of SM400B low-carbon steel with a volume of around 1.5 m³. The heat capacity C_{bulk} and the heat loss of the reactor have been theoretically and experimentally evaluated by Namioka *et al* [23]. Ignoring the weight loss and assuming the circumstance temperature is 25 °C, the energy input of the HT can be calculated as:

$$m_w \cdot (H_{l,HT} - H_{l,298}) + (m_s \cdot c_p + C_{bulk} + 3600 \times h_{bulk} \cdot A \cdot \tau) \cdot (T_H - 298) = X_{in} \cdot (H_{g,HT} - H_{l,HT}) \quad (1)$$

$$E_{HT,in} = X_{in} \cdot (H_{g,HT} - H_{l,298}) \quad (2)$$

where $E_{HT,in}$ is the energy input in the HT process; m_w , m_s is the water and solid content in the sludge; H is the enthalpy of the liquid, approximately equaling to that of water; c_p is the specific heat capacity of sludge, 1.7 kJ/(kg·K) [17]; τ is the holding time, hour; C_{bulk} is the heat capacity of the reactor, 1550 kJ/K; h_{bulk} is the convective heat transfer coefficient; A is the surface area of the reactor, $h_{bulk} \cdot A = 0.032$ kW/K [23].

After HT, the steam in the reactor was released from saturated pressure to atmospheric pressure, following with a temperature decrease inside the reactor. Assuming that the temperature inside the reactor was 373 K when the pressure inside the reactor reached to atmospheric pressure, the amount of exhaust steam can be then calculated by Eq.3:

$$(m_w + X_{in}) \cdot (H_{l,HT} - H_{l,373}) + m_s \cdot c_p \cdot (T_H - 373) = X_{out} \cdot (H_{g,HT} - H_{l,373}) \quad (3)$$

The energy output $E_{HT,out}$ after HT can be estimate as:

$$E_{HT,out} = X_{out} \cdot (H_{g,TH} - H_{l,298}) \quad (4)$$

The energy consumption of mechanical dewatering was derived from a 24 t/d pilot scale plant, which has been continually operated for 2 years.

Fig.2 Boundary of mass and energy balance calculation in current study

As for the thermal efficiency of dryer, the energy consumption in the commercial plants was about 1.2 kg(steam)/kg(water) not including the cost of capital, labor, maintenance, and operating [24]. At a lab scale (capacity, 100 kg/h), it was around 70–75% for gas-agitated double screw type dryer [25] and about 25–62% for screw conveyor dryer, a typical values obtained from the falling-rate drying period [26]. Fully taking all the above mentioned data into account, the thermal efficiency of the conventional drying process was assumed to be 70% in this study.

3. Results and Discussion

3.1 Carbonization of sewage sludge

Table 1 presents the proximate and ultimate analysis of the raw sludge and the HT products. The carbon content was increased from 51.2% to 68.0% and the corresponding HV was also improved from 4497 to 4821 kcal/kg. He *et al.* [16] also reported the higher heating values of SS was increased by at least 11% with the HT conversion. The HV of the solid fuel produced at 180 °C and 15 minutes was lower than that of the raw sludge. This could be explained by that the volatile dissolution but not the SS carbonization played the most important role resulting in HV decrease. After lengthening the holding time, the SS carbonization became more and more important, the HV of the fuels were then improved by the HT conversion. The HV was increased with both the HT temperature and holding time. However, the energetic retention efficiency was decreased because of the reduction of biofuel yields, which were decreased both with increasing the HT temperature and holding time. Moreover, high HT temperature and long holding time means high energy consumption in HT conversion. Therefore, the process optimization should consider the energy efficiency of each separated process, including the HT conversion, mechanically dewatering, and thermal drying, in order to maximize the energy recovery rate.

Table 1 the ultimate and proximate analysis of the samples

Fig.3 illustrated the coalification band of the upgraded SS. This coalification was mainly attributed to the hydrolysis reaction resulting from the heat, pressure, and time [19]. The quality of solid biofuels produced by the HT could be compared to the lignite or even sub-bituminous, depending on the reaction severity. Mass balance analyses indicated that the feedstock carbonization retained a significant fraction of carbon in the solid parts [27]. Both the atomic ratio of H/C and O/C kept decreasing with the HT temperature; yet, the degree was relevant to both the HT conditions and the feedstock. All the HT products exhibited lower H/C and O/C ratios than the initial materials, owing to the evolution of H₂O and CO₂ due to the dehydration and decarboxylation reactions [28]. Dehydration is generally explained by elimination of hydroxyl groups, while decarboxylation is

the thermal cracking of the long chain carboxylic acids. Chemical dehydration significantly carbonizes biomass by lowering the H/C and O/C ratios [20]. According to the composition of the hydrochar produced from municipal wastes, Berge *et al.* [27] concluded that both the dehydration and decarboxylation occurred during carbonization, resulting in structures with high aromaticity. Longer HT holding time seemed to decrease oxygen containing functional groups while carbon aromaticity structure increased, thereby rendering hydrochars highly hydrophobic [16]. The evolution of the H/C and O/C atomic ratios follows essentially the path of a dehydration process, similar to that previously observed for the HT transformation of saccharides such as glucose, sucrose or starch [22]. Significant decarboxylation only appeared after plenty of water has been generated [20]. The dehydroxylation and condensation reaction are regarded as possible paths for water sources. Above 150 °C, the carboxyl and carbonyl groups were rapidly degraded to release CO₂ and CO respectively. Early observations concluded that the HT decomposition of the formic acid, which is formed in significant amounts during the cellulose degradation, yielding primarily CO₂ and H₂O, was also one likely source for CO₂ formation [29].

Fig.3 The coalification band of sewage sludge with HT treatment

Many reaction mechanisms would contribute to the coalification in HT conversion, whereas more detailed reaction pathways and kinetics are largely unknown except for the HT degradation of glucose. According to previous studies on hydrochar formation mechanisms [16, 18, 21, 30-35], the hydrocarbon can be obtained via three main reaction pathways: (1) direct solid-solid conversion of biomass materials (termed “char”) undergoing devolatilization, condensation, dehydration and decarboxylation; (2) polymerization of dissolved intermediates degraded from the original biomass towards polyfurans; (3) carbonization *via* further intermolecular.

3.2 Mass balance of HT solid biofuel production

Fig.4 shows a typical mass balance of the HT solid biofuel production process run under a temperature of 200 °C and holding time of 30 min. The initial ratio of sludge to water was 2:1 with about 90.3 g SS and 45.2 g water in the parent materials. Accordingly, the solid content of the parent materials was 9.4%, about 12.7 g. After the HT, about 132.4 g (97.8% of the initial weight) product was obtained and the other 3.0 g was lost resulting from the possible weight loss and uncondensed gases. Following with a mechanically dewatering, most of the liquid (about 104.6 g, accounting for 77.2%) within the HT product was removed and 27.8 g dewatered residues (occupying 20.5%) was obtained. If not accounting the water added in the HT stage, about 60 g water (4.7 kg water/kg solid) was removed by the combination of HT treatment and mechanical dewatering. It indicates the HT treatment was an effective solution to improve sludge dewaterability. After the thermal drying,

about 11.58 g HT solid biofuel was acquired, representing 8.6% of the total sample weight. The biofuel recovery rate α was solved as 91.3% at this operating condition. Due to the avoidance of the forced evaporation of water, the energy consumption in HT was expected to be largely reduced comparing with the traditional SS thermal drying.

Fig.4 Evaluation of biofuel recovery from sludge by employing HT in Lab-scale

Table 2 listed a mass balance analysis of all the operating conditions mentioned in this study. It indicated that the weight loss of all these samples was lower than 3.5%. The sludge dewatering performance kept improving both with the HT temperature and holding time as indicated by the decrease in “dewatered residuals”. For the sample treated under 240 °C, the amount of “dewatered residuals” did not always decrease with the holding time. One possible reason was the sludge particles were too fine. Although the hydrophobic was improved [16], no effective porosity was formed during the mechanical dewatering, resulting in an increase in the resistance to filtration [36]. The other one was the O-ring placed in the connection increased the resistance to filtration and reduced the pressure acting on the sample surface due to the small volume of sample.

In virtue of the improvement of dewaterability, the amount of “evaporated liquid” was largely reduced for HT pretreated sample. The energy density of the SS was enhanced by the HT with the exception of the sample treated at 180 °C and 15 min. Its reason has been discussed in section 3.1. The energy density was improved continuously; whereas the fuel recovery ratio kept decreasing both with increasing the HT temperature and holding time. This result implies that there existed an optimal operating condition to maximize the energy output of HT solid biofuel production process. Moreover, the energy density and corresponding fuel recovery ratio must be taken into account when evaluating the economic efficiency.

All these data show that most of the liquid could be removed by combining the HT pretreatment with mechanical dewatering prior to thermal drying. After the HT pretreatment and dewatering, the wet basis HV of the products was largely improved to achieve the self-sustaining combustion.

Table 2 Evaluation of mass balance of solid biofuel production by HT

3.3 Energy balance

Fig.5 illustrates an energy balance of the proposed process (capacity 1 t/batch) producing solid biofuel under a temperature of 200 °C and holding time of 30 mins. It clearly shows that the HT pretreatment could be used to produce solid biofuel to recover energy from the high moist SS. Under the current condition, about 47.6% of the energy generated from the solid biofuel combustion could be recovered in the form of heat or power; the other

52.4% could be utilized to run the fuel production process, including the HT treatment, mechanical dewatering (press filter) and thermal drying (forced evaporation). Moreover, it is noteworthy to mention that this calculation assumed that the water residing in the solid after mechanical dewatering was removed by forced evaporation and the reactor was preheated every time. However, the reactor was required pretreating only once in a sequence operating process. Besides, the residing water was typically removed to around 20% by natural drying, which has been demonstrated to be improved by the HT pretreatment [37-39]. Therefore, the energy consumption could be reduced in practical so that more energy could be recycled. As indicated in Table 3, about 59.6% of the energy generated from the biofuel combustion could be recycled when ignoring the reactor preheating process.

Fig.5 Energy balance of solid biofuel production by HT pretreatment

In order to illustrate the energy efficiency more intuitively, Fig.5 also compares the energy consumption of the conventional thermal drying with that of the HT biofuel production process. Obviously, the heat generated from combustion of the dried SS could not fulfill the energy demand for water evaporation in conventional drying. Additional fuel with energy of 5.3 MJ should be added in order to treat 1 ton sludge with a moisture content of 85.94%. However, in HT process, the water within sludge was removed mainly by mechanical dewatering, not forced evaporation. Only the sensitive heat was required to improve the temperature of SS and water under a certain pressure. About 47.6% of the heat generated from the biofuel combustion could be recovered. This result indicates that the biofuel production from the high moist SS by employing the HT treatment was not only feasible in the view of technique, but also cost effective in terms of energy efficiency. In a similar process employing the steam explosion to produce solid biofuel from SS and leaves, Zhao *et al.* [1] reported that the energy consumption of the thermal pretreatment was only about 22% of that of consumed in the conventional thermal drying, which was consistent with that reported by Namioka *et al* [17].

Table 3 shows the energy consumption and recovery under various operating conditions. After the HT temperature was above 200 °C, the energy recovery rate was around 40%. If not taking the reactor preheating into account, the energy recovery rate was close to 60%. Before the temperature reaching 240 °C, it kept increasing with the HT temperature and holding time. The thermal drying process is the most energy-intensive one during the biofuel production, accounting for about 70% of the total energy consumption. Thus, one of the efficient ways to save energy was to improve the intensity of mechanical dewatering to remove more water from HT products. The other one is to lengthen the dewatering time. Practically, the pressure for press filter and time were about 1.2 MPa and 2 hours, respectively. In this case, the moisture content of the products could be reduced to about 40% for SS [17, 37], much lower than that reported in Table 2. Thus, the energy consumption

could be further reduced in practical. Considering the energy consumption of HT treatment, about half of the energy was consumed in the reactor preheating. Therefore, it would be better to shorten the interval between two batches to save energy.

Table 3 Evaluation of energy recovery from solid biofuel production by HT

Conclusions

This work focused on the mass and energy balance of solid biofuel production from SS by hydrothermal (HT) carbonization in order to optimize the operating parameters. The results show that the HT temperature was the most important parameter to affect the biofuel property. The sludge dewaterability, the carbon content and heating value of the biofuel were improved with increase of the HT temperature and hold time; whereas the ratio of the solid biofuel to raw sludge (biofuel recovering rate, α) drops causing a reduction in the energy recovery ratio. Thus, the process optimization should fully take into account the dehydration performance, drying performance, biofuel recovering rate α and calorific value to improve the thermal efficiency and lower the cost of biofuel production. According to the energy balance, the energy recovery rate was around 40% after the HT temperature was over 200 °C. To save energy, a moderate condition—HT temperature of 200 °C and holding time of 30 min was suggested to produce solid biofuel from SS with an energy recovery rate of 50%.

Acknowledgement

This work was financially supported by the State Scholarship Fund of China under Grant No.2011609050, No. 201206230168, and National Science and Technology Major Project of China (Water Program) (2010ZX07319-002). This research work was also supported by Strategic China-Japan Cooperative Program on “Science and Technology (S&T) for Environmental Conservation and Construction of a Society with Less Environmental Burden” of the National Nature Science Foundation of China and Japan Science and Technology Agency (No. 21161140329).

References

- [1] P. Zhao, S. Ge, K. Yoshikawa. An orthogonal experimental study on Solid fuel production from sewage sludge by employing steam explosion. *Appl Energy*. 112 (2013) 1213–21.
- [2] General Office of the State Council of the People's Republic of China. The China's 12th Five-Year Construction Plan for National Urban Wastewater Treatment and Recycling Facilities. April 5th, 2012.
- [3] W. Rulkens. Sewage sludge as a biomass resource for the production of energy: Overview and assessment of

the various options†. *Energy Fuel*. 22 (2007) 9-15.

[4] Y. Cao, A. Pawłowski. Sewage sludge-to-energy approaches based on anaerobic digestion and pyrolysis: Brief overview and energy efficiency assessment. *Renew Sust Energy Rev*. 16 (2012) 1657-65.

[5] Ronald J. Leblanc, Peter Matthews, R.P. Richard. Global atlas of excreta, wastewater sludge, and biosolids management: Moving forward the sustainable and welcome uses of a global resource. UN-HABITAT2009.

[6] A. Demirbaş. Biomass resource facilities and biomass conversion processing for fuels and chemicals. *Energy Convers Manage*. 42 (2001) 1357-78.

[7] A. Demirbas. Biofuels sources, biofuel policy, biofuel economy and global biofuel projections. *Energy Convers Manage*. 49 (2008) 2106-16.

[8] Y. Yu, J. Liu, R. Wang, J. Zhou, K. Cen. Effect of hydrothermal dewatering on the slurriability of brown coals. *Energy Convers Manage*. 57 (2012) 8-12.

[9] E. Neyens, J. Baeyens. A review of thermal sludge pre-treatment processes to improve dewaterability. *J Hazard Mater*. 98 (2003) 51-67.

[10] E. Danso-Boateng, R. Holdich, G. Shama, A. Wheatley, M. Sohail, S. Martin. Kinetics of faecal biomass hydrothermal carbonisation for hydrochar production. *Appl Energy*. 111 (2013) 351-7.

[11] Z. Liu, A. Quek, S.K. Hoekman, R. Balasubramanian. Production of solid biochar fuel from waste biomass by hydrothermal carbonization. *Fuel*. 103 (2013) 943-9.

[12] L. Lu, T. Namioka, K. Yoshikawa. Effects of hydrothermal treatment on characteristics and combustion behaviors of municipal solid wastes. *Appl Energy*. 88 (2011) 3659-64.

[13] P. Prawisudha, T. Namioka, K. Yoshikawa. Coal alternative fuel production from municipal solid wastes employing hydrothermal treatment. *Appl Energy*. 90 (2012) 298-304.

[14] M. Muthuraman, T. Namioka, K. Yoshikawa. Characteristics of co-combustion and kinetic study on hydrothermally treated municipal solid waste with different rank coals: A thermogravimetric analysis. *Appl Energy*. 87 (2010) 141-8.

[15] M. Muthuraman, T. Namioka, K. Yoshikawa. A comparison of co-combustion characteristics of coal with wood and hydrothermally treated municipal solid waste. *Bioresour Technol*. 101 (2010) 2477-82.

[16] C. He, A. Giannis, J.-Y. Wang. Conversion of sewage sludge to clean solid fuel using hydrothermal carbonization: Hydrochar fuel characteristics and combustion behavior. *Appl Energy*. 111 (2013) 257-66.

[17] T. Namioka, Y. Morohashi, R. Yamane, K. Yoshikawa. Hydrothermal treatment of dewatered sewage sludge cake for solid fuel production. *J Environ Eng*. 4 (2009) 68-77.

319 [18] T. Karayıldırım, A. Sinağ, A. Kruse. Char and coke formation as unwanted side reaction of the
 320 hydrothermal biomass gasification. *Chem Eng Technol.* 31 (2008) 1561-8.

321 [19] M. Nonaka, T. Hirajima, K. Sasaki. Upgrading of low rank coal and woody biomass mixture by
 322 hydrothermal treatment. *Fuel.* 90 (2011) 2578-84.

323 [20] A. Funke, F. Ziegler. Hydrothermal carbonization of biomass: a summary and discussion of chemical
 324 mechanisms for process engineering. *Biofuel Bioprod Bior.* 4 (2010) 160-77.

325 [21] S. Kang, X. Li, J. Fan, J. Chang. Characterization of Hydrochars Produced by Hydrothermal Carbonization
 326 of Lignin, Cellulose, d-Xylose, and Wood Meal. *Ind Eng Chem Res.* 51 (2012) 9023-31.

327 [22] M. Sevilla, A.B. Fuertes. Chemical and structural properties of carbonaceous products obtained by
 328 hydrothermal carbonization of saccharides. *Chem-eur J.* 15 (2009) 4195-203.

329 [23] T. Namioka, M. Miyazaki, Y. Morohashi, K. Umeki, Y. Kunio. Modeling and analysis of batch-type thermal
 330 sludge pretreatment for optimal design. *J Environ Eng.* 3 (2008) 170-81.

331 [24] J. Werther. Potentials of biomass co-combustion in coal-fired boilers. *Proceedings of the 20th International
 332 Conference on Fluidized Bed Combustion.* Springer, Xi'an, China, 2010. pp. 27-42.

333 [25] H.S. Kim, M.S. Shin, D.S. Jang, E.S. Na. A study for the thermal treatment of dehydrated sewage sludge
 334 with gas-agitated double screw type dryer. *J Environ Sci Heal A.* 40 (2005) 203-13.

335 [26] S. Waje, B. Thorat, A. Mujumdar. An experimental study of the thermal performance of a screw conveyor
 336 dryer. *Dry Technol.* 24 (2006) 293-301.

337 [27] N.D. Berge, K.S. Ro, J. Mao, J.R. Flora, M.A. Chappell, S. Bae. Hydrothermal carbonization of municipal
 338 waste streams. *Environ Sci Technol.* 45 (2011) 5696-703.

339 [28] J. Akhtar, N.A.S. Amin. A review on process conditions for optimum bio-oil yield in hydrothermal
 340 liquefaction of biomass. *Renew Sust Energ Rev.* 15 (2011) 1615-24.

341 [29] J. Yu, P.E. Savage. Decomposition of formic acid under hydrothermal conditions. *Ind Eng Chem Res.* 37
 342 (1998) 2-10.

343 [30] A. Kruse, P. Maniam, F. Spieler. Influence of proteins on the hydrothermal gasification and liquefaction of
 344 biomass. 2. Model compounds. *Ind Eng Chem Res.* 46 (2007) 87-96.

345 [31] E. Dinjus, A. Kruse, N. Tröger. Hydrothermal carbonization–1. Influence of lignin in lignocelluloses. *Chem
 346 Eng Technol.* 34 (2011) 2037-43.

347 [32] C. Falco, N. Baccile, M.-M. Titirici. Morphological and structural differences between glucose, cellulose
 348 and lignocellulosic biomass derived hydrothermal carbons. *Green Chem.* 13 (2011) 3273-81.

- [33] M. Zhang, H. Yang, Y. Liu, X. Sun, D. Zhang, D. Xue. Hydrophobic precipitation of carbonaceous spheres from fructose by a hydrothermal process. *Carbon*. 50 (2012) 2155-61.
- [34] B. Hu, K. Wang, L. Wu, S.H. Yu, M. Antonietti, M.M. Titirici. Engineering carbon materials from the hydrothermal carbonization process of biomass. *Adv Mater*. 22 (2010) 813-28.
- [35] A. Kruse, A. Funke, M.-M. Titirici. Hydrothermal conversion of biomass to fuels and energetic materials. *Curr Opin Chem Biol*. (2013).
- [36] P. Zhao, S. Ge, Z. Chen, X. Li. Study on Pore Characteristics of Flocs and Sludge Dewaterability Based on Fractal Methods (Pore Characteristics of Flocs and Sludge Dewatering). *Appl Therm Eng*. 58 (2013) 217-23.
- [37] Z.L. Jiang, D.W. Meng, H.Y. Mu, K. Yoshikawa. Study on the hydrothermal drying technology of sewage sludge. *Sci China Technol Sci*. 53 (2010) 160-3.
- [38] D. Meng, Z. Jiang, K. Yoshikawa, H. Mu. The effect of operation parameters on the hydrothermal drying treatment. *Renew Energ*. 42 (2012) 90-4.
- [39] P. Zhao, S. Ge, C. Areeprasert, D. Ma, K. Yoshikawa. Effect of the hydrothermal pretreatment on the convective drying of paper sludge. *Sci China Technol Sci*. (2013) submitted to SCIENCE CHINA Technological Sciences.

Figures' Captions

Fig.1 Fuel production process and an elementary diagram of experimental facilities

Fig.2 Boundary of mass and energy balance calculation in current study

Fig.3 The coalification band of sewage sludge with HT treatment

Fig.4 Evaluation of fuel recovery from sludge by employing HT in Lab-scale

Fig.5 Energy balance of solid fuel production by HT pretreatment

Note: all figures are color on the Web (free of charge) and in black-and-white in print

Tables' Captions

Table 1 the ultimate and proximate analysis of the samples

Table 2 Evaluation of mass balance of solid fuel production by HT

Table 3 Evaluation of energy recovery from solid fuel production by HT

Table 1 the ultimate and proximate analysis of the samples

Items	Raw	180 °C			200 °C			220 °C			240 °C		
	sludge	15 min	30 min	45 min	15 min	30 min	45 min	15 min	30 min	45 min	15 min	30 min	45 min
Ultimate analysis (dry ashes free basis, <i>d.a.f.</i> , %)													
C _{ad}	51.20	52.19	53.20	55.31	55.07	57.44	59.23	59.31	60.00	62.31	61.29	62.62	67.96
H _{ad}	6.64	6.57	6.52	6.69	6.72	6.65	6.68	6.68	6.45	6.29	6.26	5.96	5.71
N _{ad}	8.85	9.08	8.98	9.08	9.29	9.04	7.98	9.03	8.24	8.73	8.95	8.19	8.51
S _{ad}	1.37	1.36	1.39	1.37	1.30	1.26	1.26	1.18	1.19	1.20	1.33	1.29	1.56
O _{ad} ^a	31.94	30.81	29.91	27.55	27.62	25.61	24.84	23.80	24.13	21.47	22.17	21.93	16.25
Proximate analysis (air dry basis, %)													
M _{ad}	0.99	1.45	1.54	1.61	1.11	1.07	0.99	0.81	1.24	0.29	1.02	0.53	0.48
V _{ad}	78.49	74.47	72.52	69.63	71.16	70.37	67.56	68.94	67.36	66.59	64.63	62.40	57.42
FC _{ad}	2.01	2.06	2.77	3.20	3.06	3.19	3.60	3.02	3.79	3.97	5.12	6.66	7.42
A _{ad}	18.51	22.02	23.17	25.56	24.67	25.37	27.85	27.23	26.61	29.23	29.83	30.41	34.68
HV ^b	4497	4374	4546	4645	4518	4725	4747	4717	4786	4748	4746	4816	4821

^a Calculated from the elements mass balance: O=100-C-H-N-S; ^b HV: heating value (kcal/kg).

Table 2 Evaluation of mass balance of solid fuel production by HT

samples	Input (%)	Hydrothermal (%)		Dewatering (%)		Thermal drying (%)		HV (kcal/kg)	α (%)	
		Gas+loss	HT products	Separated liquid	Dewatered residuals	Evaporated liquid	Solid fuel			
Raw sludge	100	—	—	29.70	70.30	56.24	14.06	4497	100	
180 °C	15 min	100	1.36	98.64	68.08	30.56	21.48	9.09	4374	96.94
	30 min	100	1.89	98.11	72.89	25.23	16.45	8.77	4546	93.91
	45 min	100	1.92	98.08	77.35	20.74	12.26	8.48	4645	90.59
	15 min	100	3.06	96.94	74.23	22.72	14.00	8.72	4518	93.26
200 °C	30 min	100	2.22	97.78	77.24	20.54	11.99	8.55	4725	91.27
	45 min	100	1.29	98.71	80.11	18.60	10.31	8.29	4747	88.40
	15 min	100	2.42	97.58	76.68	20.89	12.38	8.52	4717	90.75
	30 min	100	3.47	96.53	77.81	18.72	10.60	8.11	4786	86.60
220 °C	45 min	100	2.65	97.35	81.03	16.32	8.41	7.91	4748	84.43
	15 min	100	2.12	97.88	80.73	17.15	9.18	7.97	4746	85.16
	30 min	100	1.98	98.02	79.40	18.62	10.94	7.68	4816	82.07
	45 min	100	3.24	96.76	78.51	18.25	10.93	7.33	4821	78.14

Table 3 Evaluation of energy recovery from solid fuel production by HT

Samples		Input in HT ^a (MJ)	Input in HT (MJ)	Dewatering (MJ)	Thermal drying(MJ)	^a Total input (×10 ³ MJ)	Total input (×10 ³ MJ)	Total output (×10 ³ MJ)	β^a (%)	β (%)
Raw sludge		0	0	0	3 181.93	3.18	—	2 655.57	-19.82	—
180 °C	15 min	603.73	331.47	6.43	1 208.00	1.82	1.55	2 503.90	27.39	38.26
	30 min	608.79	336.52	6.43	933.41	1.57	1.28	2 574.71	38.57	49.37
	45 min	613.85	341.58	6.43	699.43	1.34	1.05	2 539.71	46.89	57.85
200 °C	15 min	639.04	331.88	6.43	797.07	1.46	1.14	2 541.51	42.02	54.37
	30 min	644.75	337.58	6.43	683.92	1.35	1.03	2 602.43	47.57	59.64
	45 min	650.46	343.29	6.43	590.81	1.26	0.94	2 534.09	49.65	62.05
220 °C	15 min	674.51	332.39	6.43	704.32	1.40	1.04	2 583.54	45.20	58.73
	30 min	680.87	338.74	6.43	606.56	1.32	0.95	2 560.56	47.14	61.11
	45 min	687.23	345.10	6.43	485.31	1.20	0.84	2 479.39	50.20	64.65
240 °C	15 min	710.20	333.01	6.43	528.37	1.28	0.87	2 526.83	47.84	63.64
	30 min	717.21	340.02	6.43	624.35	1.39	0.97	2 504.66	42.25	58.41
	45 min	724.21	347.03	6.43	621.74	1.40	0.98	2 395.38	39.21	56.16

a: based on the assumption that the reactor was preheated every time.

Fig.1 (color on the Web (free of charge) and in black-and-white in print)

Fig.2

Fig.3 (color on the Web (free of charge) and in black-and-white in print)

Fig.4 (color on the Web (free of charge) and in black-and-white in print)

Fig.5 (color on the Web (free of charge) and in black-and-white)