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Steam gasification of solid recovered fuel char derived from landfill waste: A kinetic study

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Abstract

A kinetic study of steam gasification of char derived from solid recovered fuel (SRF) was performed. The char was prepared by using pyrolysis and using a nitrogen atmosphere at a temperature of 900 °C for 1 h. Gasification in a steam atmosphere was conducted for the char product using isothermal thermogravimetry analysis (TGA). The gasification was carried out at three different temperatures: 800, 850, and 900 °C for at least 1 h. The calculation of the kinetic parameters were done by considering the volumetric model, shrinking core model, and random pore model. The results show that the increase of the gasification temperature was linearly correlated to the rise of the reaction rate of the carbon conversion. Moreover, the activation energy of the char reaction is known to range between 58-83 kJ/mol. The kinetic parameters of char gasification were affected by their corresponding SRF component and the char particle size. Also, there were no significant different in the kinetic parameters calculated using the three different models as the difference between the results was less than 3.5 %.

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Keywords: steam gasification; solid waste; thermogravimetry analysis; char

1. Introduction

It is estimated that currently there are approximately 150,000-500,000 of closed and active landfill sites in the EU-28 countries [1]. Unfortunately, nearly 90% of those landfill sites are non-sanitary landfills [2]. This might lead to severe environmental problems such as fire hazard, pollution of rivers and underground facilities, and other health-

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related problems [3]. Hence, the mining of landfills and the remediation of polluted land has been recognized as essential measures for the protection of air, land, and water resources in Europe [4].

The Enhanced Landfill Mining (ELFM) concept has been proposed as an improvement to the conventional landfill mining [5]. In ELFM, efforts are given to maximize the resource recovery by implementing integrated valorization of landfill waste as both materials (waste-to-material) and energy (waste-to-energy) by using innovative transformation technologies [5]. In ELFM, the plasma-based gasification has an important role in achieving a complete resource recovery by converting the unrecovered fraction of the excavated Municipal Solid Waste (MSW) into syngas and valuable solid raw materials [5]. Moreover, when compared to the other thermal treatments, gasification is found to give better results in terms of increases in material decompositions and chemical energies [6]. In contrast to the use of other gasifying agents, it is believed that gasification using steam is an effective thermochemical method of producing greater yields of H₂; hence the syngas generated by steam gasification is found to have a relatively high lower-heating value (LHV) [7].

In the case of MSW gasification, a fundamental understanding of its char gasification reactivity and kinetics is essential to enable an efficient thermochemical conversion. Understanding the mechanism of char gasification is crucial for the design and simulation of gasifiers [8]. Previously, kinetics studies of MSW conversion using thermogravimetry analysis (TGA) have been done by various researchers [9,10]. However, they mainly focused on the pyrolysis process of MSW using an inert atmosphere. Meanwhile, studies regarding the reactivity and kinetics of char derived from MSW, especially landfill waste, in a steam atmosphere are still limited. Hence, the objective of this study is to investigate the kinetics of the char gasification derived from MSW under steam atmosphere by using TGA. Two different commercial SRFs were used in this study to represent the unrecovered fraction of the MSW from the landfill.

2. Materials and methods

2.1. SRF samples

Table 1. Fuel characteristic and composition of raw SRF samples.

	SRF-1	SRF-2	SRF from landfill waste [11]
Component, wt%			
Plastics	34.92	23.55	NA
Wood	15.83	3.03	NA
Paper and cardboard	40.29	21.60	NA
Textiles and fibers	8.96	13.02	NA
Polyester sponge	-	38.80	NA
Proximate composition, wt%			
Moisture content (ar)	3.5	3.8	14.4
Volatile matter (db)	75.0	NA	80.4
Ash (db)	13.0	6.47	27.1
Elemental composition, wt% (db)			
Carbon – C (db)	50.0	NA	54.9
Hydrogen – H (db)	7.0	NA	7.38
Nitrogen – N (db)	0.8	NA	2.03
Oxygen – O (db)	28.0	NA	NA
Sulfur – S (db)	0.2	3.37	0.36
LHV, MJ/kg (db)	21.00	23.52	22.0

NA = Not available

In this study, SRF fraction from landfill was simulated by using SRF produced from fresh MSW. The SRF samples were supplied by a commercial SRF producer (Shanks, Belgium). The SRFs were produced through a series of operations including shredding, screening, sorting, and drying of commercial and industrial waste. Table 1 shows the fuel characteristic and composition of both raw SRF samples. The SRF-1 samples were obtained in a pelletized

condition that consist of plastics, paper, wood, and a small fraction of textiles. Moreover, SRF-2 samples consisted of a fluffy material that mainly consists of shredded polyester sponge, paper, plastics, and textiles. Data of SRF processed from excavated MSW from a landfill site in Belgium, as found in another study [11], is also provided as a comparison. It can be seen from the table SRF samples used in this study, especially SRF-1 sample, have a similar fuel characteristic compared to the literature data. Both samples have a relatively high ash content and less than 10 % in the difference between the LHV, volatile matter, carbon, hydrogen, and sulfur content.

2.2. Char preparation

A fixed bed reactor was used to produce the SRF char by using the pyrolysis process. The reactor consists of a metal tube, a thermocouple, an electric heater, a cooling bath, and a gas cleaning system as shown in Fig.1. For each experiment, approximately 10-15 g of sample was placed in the middle of the metal tube. Thereafter, a thermocouple was inserted into the tube until its end was about 1 cm above the sample. Before heating up the reactor, purging with nitrogen with a flow of 100 ml/min was done for about 20 min to make sure the tube was free from oxygen. Then, the reactor was heated up from an ambient temperature to 900 °C, by using an average heating rate of 25 °C/min. The reactor was then maintained at the same temperature for 1 h which followed by a cooling process to an ambient temperature. The nitrogen flow rate was kept constant during the pyrolysis experiment. Prior to the char gasification experiment, the char products were crushed and sieved to a particle size of less than 0.25 mm and 1.40 mm.

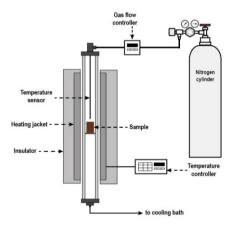


Fig. 1. Schematic diagram of pyrolysis reactor for char preparation.

2.3. Char gasification experiment

An isothermal gasification was carried out by using a thermogravimetry analyzer (STA 449 F1 Jupiter, NETZSCH). For each experiment, approximately 50 mg of char was placed inside a crucible. The experiment was started with a heating step from the ambient temperature to the target temperature under an inert temperature that was achieved by flowing 100 ml/min of nitrogen. Three gasification temperatures were used in the experiment, which were 800, 850, and 900 °C. Once the target temperature was achieved, approximately 40 ml/min of steam was inserted into the furnace. It should be noted that during gasification, 30 ml/min of nitrogen was kept inside the furnace as a protector gas. The gasification was then continued for at least for 1 h. The data of the mass degradation (balance resolution = $0.025 \mu g$), temperature (temperature resolution = 0.001 K), and reaction time were collected during the experiment.

2.4. Kinetics model and calculation

The data of the mass degradation obtained from the TGA experiment are converted to conversion ratio (X) by considering the mass (m), initial mass (m_0) , and final mass (m_f) of the sample during gasification as formulated in the following equation.

$$X = \frac{m_o - m}{m_o - m_f} \tag{1}$$

During the gasification, the apparent reaction rate $\left(\frac{dX}{dt}\right)$ is defined as a differential of the conversion ratio against time (t) that can be expressed as follows:

$$\frac{dX}{dt} = -\frac{dm}{dt} \frac{1}{m_o - m_f} \tag{2}$$

The apparent reaction rate can also be defined as a function of the kinetic rate constant (k) and the changes in the physical and chemical properties of the sample. According to the Arrhenius equation, the kinetic rate constant can be expressed as a function of the temperature written as follows:

$$k = A \exp\left(-\frac{E_a}{RT}\right) \tag{3}$$

where A is the pre-exponential factor, E_a is the activation energy, R is the gas constant (8.314 J K⁻¹ mol⁻¹), and T is the temperature. In this study, three models were adopted to investigate the reactivity of the SRF char: the volumetric model (VM), the shrinking core model (SCM), and the random pore model (RPM).

VM is the simplest model which assumes uniform gas diffusion occurs during the gas-solid reaction within the entire particle in all possible places, both outside and inside the particle surface [8]. The basic equation of this model is [8]:

$$\frac{dX}{dt} = k_{VM} \left(1 - X \right) \tag{4}$$

In SCM, the particles are assumed as a porous particle that consists of uniform nonporous spherical grains which have a non-reacting core at its center. The reaction occurs on the grain surface in which the shrinking core behavior applies to each of these grains [8,12]. The reaction rate of SCM can be expressed as follows [8]:

$$\frac{dX}{dt} = k_{SCM} (1 - X)^{2/3} \tag{5}$$

RPM considers the structural change of the particles during gasification due to coalescence and overlapping of pore surfaces [12]. In RPM, ψ represents the structural parameter of the sample that can be calculated by finding the maximum conversion ratio, X_{max} , for which maximum reaction rate occurs [12]. The reaction rate of RPM is given by the following equation [8,12]:

$$\frac{dX}{dt} = k_{RPM} \left(1 - X \right) \sqrt{1 - \psi \ln(1 - X)} \tag{6}$$

where ψ can be determined by using the following equation,

$$\psi = \frac{2}{2\ln(1 - X_{\text{max}}) + 1} \tag{7}$$

3. Results

3.1. Conversion ratio and reaction rate at different gasification temperatures

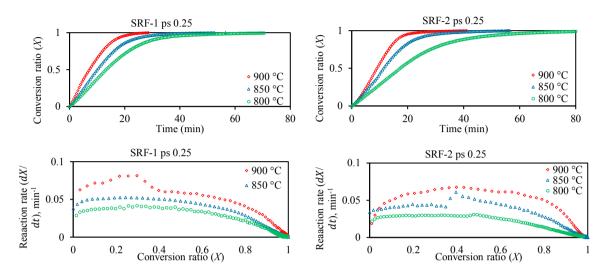


Fig. 2. (a) Conversion ratio and (b) reaction rate of char (particle size 0.25 mm) during gasification at different temperatures.

Fig. 2(a) shows the relationship of the conversion ratio against reaction time during steam gasification of the SRF char samples with a particle size of 0.25 mm (SRF-1 ps 0.25 and SRF-2 ps 0.25) at three gasification temperatures. It is clear that the reaction time to complete the char conversion during gasification was affected by the gasification temperature. Rising the temperature cause a shorter reaction time. Fig. 3 shows the time needed to achieve the 0.5 conversion ratio (t_{0.5}) for each temperature range, which is used to characterize the char conversion in this study. As can be seen from the figure, at the same particle size, all of the t_{0.5} value of the SRF-1 sample were at least 1.5 min lower than that of SRF-2 that indicated a higher reactivity of SRF-1 char.

Fig. 2(b) shows the relationship between the reaction rate and the conversion ratio during gasification. The SRF-1 char exhibited the highest reaction rate at a conversion ratio of approximately 0.25 as the reaction rate known to reach 0.081 min⁻¹ at 900 °C, while in the case of SRF-2 the highest reaction rate, 0.067 min⁻¹ at 900 °C, occurred at a higher conversion ratio (0.40). The reactivity of char during gasification is strongly affected by the char physical structure, especially the char porosity [8]. Hence, one of the main reason of the difference between the reactivity of the SRF char might be caused by the different porosity of the char as their raw SRF have different components. A further investigation is needed to confirm this finding.

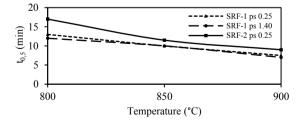


Fig. 3. The time required to complete 50% conversion ratio for various SRF char.

3.2. Effect of char particle size on conversion rate

Gasification of SRF-1 char sample with two different particle sizes was performed to investigate the effect of the char particle size on the char reactivity. The results obtained indicated that the conversion rate of the char showed no significant different in size range tested in term of the reaction time. As can be seen in Fig. 3, the t_{0.5} of both SRF-1 ps 0.25 and SRF-1 ps 1.40 char were quite similar for each gasification temperature as the difference was less than 1 min for each gasification temperature.

3.3. Kinetic parameters

Kinetic models Particle size Char (mm) VM SCM RPM A [min-1] 0.25 46.97 37.48 46.97 SRF-1 SRF-1 1.40 209.33 174.74 209.33 548.29 SRF-2 0.25 337.58 548.29 Ea [kJmol-1] SRF-1 0.25 58.26 58.32 58.26 SRF-1 1 40 72.42. 72.90 72.42 SRF-2 0.25 83.20 80.44 83.20

Table 2. Kinetic parameters of char gasification

Three kinetic models (VM, SCM, and RPM model) were fitted with the TGA results to predict the kinetic behavior of the SRF char samples. The rate constants of the models (k_{VM} , k_{SCM} , and k_{RPM}) were calculated from the data for three different temperature within the conversion ratio range of 0.1 - 0.8. In order to evaluate the kinetic parameters, the Arrhenius law described in Equation (3) was adopted by plotting the reciprocal relationship between the absolute temperature (1/T) and the logarithm of the rate constant (ln k) obtained for each model at each temperature range. The linearization of the relationship curve was verified with a good correlation in which the correlation coefficients were equal to at least 0.95.

Table 2 shows the calculated kinetic parameter for 0.25 mm SRF-1, 1.40 mm SRF-1, and 0.25 mm SRF-2 samples. It can be seen that according to the studied kinetic models, the activation energies of the SRF char were in the range of 58 – 83 kJ/mol. In general, there was no significant different between the kinetic parameters calculated using those models as the difference between the results was about 1.01 - 3.32 % for all samples. At the same particle size of 0.25 mm, the SRF-2 char exhibited a higher activation energy (80.20 - 83.20 kJ/mol) than the SRF-1 char (58.26 - 58.32 kJ/mol). Moreover, larger particle size causes a higher activation energy as shown in the result of 1.40 mm SRF-1 sample.

4. Conclusion

In this study, steam gasification of char derived from SRF was investigated. The result shows that the reaction rate of the char conversion was significantly affected by the gasification temperature as at a higher temperature, the gasification occurred at a shorter time. Moreover, the activation energy of the char reaction is known to range between 58-83 kJ/mol in which there was no significant difference between the VM, SCM, and RPM results. The kinetic parameters of char gasification were significantly affected by their corresponding SRF components. Moreover, the larger particle size of the char causes a higher activation energy although there is no difference in term of the conversion rate during gasification. The results of this study will be an important part of our next studies that will include simulation studies of steam plasma gasification of landfill waste.

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