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# Study on the combined sewage sludge pyrolysis and gasification process: mass and energy balance

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## Abstract

A combined pyrolysis and gasification process for sewage sludge was studied in this paper for the purpose of its safe disposal with energy self-balance. Three sewage sludge samples with different dry basis lower heat values (LHV(db)) were used to evaluate the constraints on this combined process. Those samples were pre-dried and then pyrolysed within the temperature range of 400–550 °C. Afterwards, the char obtained from pyrolysis was gasified to produce fuel gas. The experimental results showed that the char yield ranged between 37.28 and 53.75 wt% of the dry sludge and it changed with ash content, pyrolysis temperature and LHV(db) of the sewage sludge. The gas from char gasification had a LHV around 5.31–5.65 MJ/Nm<sup>3</sup>, suggesting it can be utilized to supply energy in the sewage sludge drying and pyrolysis process. It was also found that energy balance in the combined process was affected by the LHV(db) of sewage sludge, moisture content and pyrolysis temperature. Higher LHV(db), lower moisture content and higher pyrolysis temperature benefit energy self-balance. For sewage sludge with a moisture content of 80 wt%, LHV(db) of sewage sludge should be higher than 18 MJ/kg and the pyrolysis temperature should be higher than 450 °C to maintain energy self-sufficiency when volatile from the pyrolysis process is the only energy supplier; when the LHV(db) was in the range of 14.65–18 MJ/kg, energy self-balance could be

maintained in this combined process with fuel gas from char gasification as a supplementary fuel; auxiliary fuel was always needed if the LHV(db) was lower than 14.65 MJ/kg.

**Keywords:** sewage sludge; pyrolysis; gasification; char; energy self-balance

## 1. Introduction

Disposal of sewage sludge has become a pressing problem due to the huge amount generated, stricter legislation and shortage of land space for landfilling. Sewage sludge discharged from wastewater treatment plants in China is characterized by high moisture content (about 78–80 wt%) and relatively lower heat values (LHVs; dry basis LHVs often fall below 15 MJ/kg), which make its disposal highly energy consuming. On the other hand, traditional disposal methods, such as land application, landfilling and incineration, are facing stricter controls and requirements, for example, the moisture content of sewage sludge should be lower than 60 wt% for landfilling according to the relevant regulation [1]. Compared to landfilling or land application, thermal treatments, such as incineration and gasification, have the advantage of reducing the sludge volume dramatically. However, for incineration disposal, in addition to expensive investment costs, auxiliary fuel is needed in most of the cases due to the high moisture content of the sewage sludge. Pyrolysis of sewage sludge could be an alternative, considering its lower cost in air pollution control and flexibility in product choice [2].

There are three products from the pyrolysis of sewage sludge: oil, gas and char; both oil and gas leave the pyrolysis furnace as volatile. Sanchez *et al.* [3] studied the pyrolysis oil composition with gas chromatography-mass spectroscopy (GC-MS) and found that the pyrolysis oil was a complex mixture of organic compounds. The volatile oil and gas produced in the pyrolysis process showed a high heat value, comparable to some fossil fuel [4]. In addition the char, which has been a concern in recent years because of its porous structure, can be utilized as a supporting matrix for soil modification [5]. It was reported that some small beneficial soil organisms can be kept

away from their natural competitors by penetrating into the pore space of bio-char [6]. Experimental results also showed that the addition of bio-char improved the production of plants due to the combined effect of increased nutrient availability and improved soil chemical conditions [7].

Gasification of sewage sludge is an alternative of treatment that produces fuel gas which can be used as a fuel [8]. However, the tar content in the gas leaving the gasifier inhibits its possible usage in many applications [9,10]. When the pyrolysis char is gasified, a tar-free fuel gas can be produced [11,12].

In a conventional pyrolysis process the oil is the target product; however, for sewage sludge pyrolysis, the main purpose is its disposal without secondary pollution. Therefore, to make the process economical, energy self-sufficiency is important. Considering most sewage sludge in China is of low LHVs, to make up the energy balance in this work the sewage sludge samples will be pyrolysed first and the volatile oil and gas produced is immediately combusted to provide the energy demanded in the process; then, the char obtained from pyrolysis is gasified to produce fuel gas as auxiliary fuel if necessary; otherwise, the char will be remained as final product if the volatile is enough to balance the energy consumption. Three samples with different dry basis lower heat values (LHV(db)) were used to check the validity of the process and the constraints on moisture content and the LHV(db) of sewage sludge.

## **2. Materials and methods**

### ***2.1. Sewage sludge samples***

Three samples of sewage sludge were taken from Quyang municipal wastewater treatment plant in Shanghai at different periods of the year and they were named SS1, SS2 and SS3. The proximate analysis and ultimate analysis of these sewage sludge samples are shown in Table 1. Their LHV(db) were tested with an XRY-1 Bomb-Calorimeter and their ultimate analysis was determined with a Vario EL III Elemental Analyser. The proximate analysis was carried out according to Chinese standard GB/T 212-2008 [13].

## 2.2. Sewage sludge pyrolysis

In this work, the sewage sludge samples were firstly dried in a ventilated drier until the moisture content was reduced from its initial value to 3 wt%. Then, the dried sewage sludge samples were put into a horizontal quartz reactor, which was placed in an electrical oven under the pre-set temperature range of 400–550°C, as shown in Figure 1. This temperature range was selected because bio-char formation generally occurs in this range [5]. About 50g of dried sewage sludge was used in each run. In order to ensure inert atmosphere argon flow was passing through the reactor, the flow rate of argon was set at 40 ml/min, ensuring the pyrolysis gas leaves the pyrolysis oven once it is generated, to prevent the pyrolysis oil vapour from secondary cracking [14]. The char was collected and measured after the oven was cooled down. The oil was condensed in a two-stage ice-cooled condenser, and the non-condensable gas was collected for analysis with a GC-9160 type gas chromatograph.

## 2.3. Char gasification

Gasification of the char from pyrolysis was carried out in a small tube furnace, as shown in Figure 2. About 20 g of bio-char was used and the oxygen supply was arranged to control the equivalence air ratio (ER) in the range of 0.35–0.4; the range of ER proved to be practical and suitable for a biomass char, under which the gasification temperature up to 900°C can be maintained by exothermal reactions [12]. In this study the gasification temperature was modified between 800 and 850°C [15] with help of electricity heating regulation. The fuel gas produced passed the ice-cooled condenser before being introduced to a gas chromatograph.

An alternative method was applied to maintain the target ER conditions during the batch tests carried out. Three steps were included in the method and they are as follows.

- (1) The theoretical oxygen demand ( $O_2^0$ ) is estimated according to the LHV of char with help of an empirical formula [16]:

$$O_2^0 = (239Q_{char} + 450)/990, \text{ Nm}^3/\text{kg}_{char} \quad (1)$$

where  $Q_{char}$  represents the LHV of the char, in the unit of MJ/kg.

- (2) The actual oxygen supply ( $O_2^a$ ) can be calculated by the following equation:

$$O_2^a = ER \times O_2^0 \times m_{char}, \text{ Nm}^3 \quad (2)$$

where  $m_{char}$  is the mass of char for gasification, which is 20 g (0.02 kg) in these experiments.

- (3) A reaction period of around 15–20 min was adopted to complete the gasification. The oxygen flow rate was finally determined by supplying  $O_2^a$  evenly during the whole reaction period.

## **2.4. Energy balance**

The energy balance of the process was set up by burning the pyrolysis volatile immediately to supply energy for pyrolysis; if the pyrolysis volatile could not satisfy the energy demand, the pyrolysis char was gasified and the gas was used as supplementary fuel, as shown in Figure 3.

## **3. Results and discussion**

### **3.1. Mass distribution of pyrolysis products**

Figure 4 shows the change of the char yields and volatile fractions with the pyrolysis temperature. As reported by other researchers [3], higher temperature increases the volatile fraction produced during pyrolysis. Therefore, the char production decreases at the same time. As the temperature was higher than 500 °C, the yields of the volatile fraction were higher than 50 wt% for the three testing samples. It verifies that the mass transfer from char to volatile fraction is mainly influenced by pyrolysis temperature. In Figure 4 it is also shown that the char production from SS1 pyrolysis is greater than that from SS2. This is due to the higher ash content and lower LHV(db) associated with SS1.

Both volatile and char yield curves of SS1 were almost unchanged after the temperature

reached 500 °C, which is different from the other two samples. This phenomenon might be caused by its high ash content, which mainly consists of inorganic salts, and could accelerate the pyrolysis process [17]. For the three samples, the char yields fall within the range of 37.28–53.75 wt% of dry sludge; they are mainly affected by the ash content and pyrolysis temperature of sewage sludge, as shown in Figure 4. The higher ash content of SS1 may not necessarily result in its higher char yield, since it corresponds to a lower fixed carbon content at the same time, as shown in Table 1. However, the proximate analysis data in Table 1 were obtained at 920 °C, under which some inorganic salts would decompose to increase the volatile proportion. However, these salts will remain in the char under temperatures in pyrolysis tests, which contributes to the higher char yields of SS1 together with its ash content. In general, the relative level of LHV(db) can reflect the existence of such inorganic salts when the fixed carbon content is low and lower LHV(db) will be associated with higher char yield.

### 3.2. Energy consumption of sewage sludge pyrolysis

Energy consumption associated with the pyrolysis process is made up of three parts: the first part is for moisture evaporation (here notated as  $Q_1$ ); the second part is to heat up dry sewage sludge from its initial temperature to the pyrolysis temperature (notated as  $Q_2$ ); and the third part is reaction energy (endothermic or exothermic, notated as  $Q_3$ ), which was measured with a CRY-2P type differential thermal analyser (DTA). An example of a DTA curve for SS2 is shown in Figure 5.

$Q_1$  can be calculated as

$$Q_1 = (\gamma + c_w \Delta T_1) \times M, \text{ MJ/kg} \quad (3)$$

where  $\gamma$  is the enthalpy of vaporization of water during the drying process (2.2 MJ/kg),  $c_w$  is the specific heat capacity of water (4.187 kJ/kg °C),  $\Delta T_1$  is the difference between the initial temperature (which is assumed as 20°C) and 100°C and  $M$  is the moisture content of wet sewage sludge.

$Q_2$  can be calculated as

$$Q_2 = c_s \Delta T_2 \times (1 - M), \text{ MJ/kg} \quad (4)$$

where  $c_s$  is the specific heat capacity of dry sludge (1.95 kJ/kg °C) [18] and  $\Delta T_2$  is the difference between the initial temperature and the pyrolysis temperature.

$Q_3$  is obtained by integrating the DTA curve [19], as shown in Figure 5. Firstly, the DTA curve is divided into several stages by the occurrences of endothermic peaks. For example, the DTA curve in Figure 5 for the SS2 pyrolysis process was classified into three stages. Then a base line of each stage was drawn that represents the energy consumption of heating up dried matters. The enthalpy requirement in  $i$  stage can be calculated by integrating the area between the DTA curve and the base line made up. Finally,  $Q_3$  is calculated as

$$Q_3 = \sum q_i \times (1 - M), \text{ MJ/kg} \quad (5)$$

where  $q_i$  represents the enthalpy requirement in  $i$  stage, in MJ/kg.

The total energy consumption for pyrolysis is calculated by  $Q_1 + Q_2 + Q_3$ . All of the results of those energy consumptions are summarized in Table 2. From Table 2 it can be found that  $Q_1$  is the most crucial part, which accounts for about 90% of total energy consumption.  $Q_2$  and  $Q_3$  for each sample are of the same order of magnitude as reported [20] and they are relatively small.

### 3.3. Energy balance and product choice

The volatile fraction produced during the pyrolysis process is a mixture of oil vapour and non-condensable gas at the pyrolysis temperatures. To avoid energy loss it is unnecessary to condense and separate the former from the latter; the pyrolysis system is simplified simultaneously. A better choice on a large-scale operation is to burn the hot volatile fraction immediately to supply energy for the pyrolysis process. To meet the energy demand for the pyrolysis process, the following equation should be satisfied:

$$Q_s \times \eta \geq Q_1 + Q_2 + Q_3 \quad (6)$$



where  $Q_s$  is the energy supplied to the pyrolysis process and  $\eta$  is the energy utilization efficiency. In the system where the volatile fraction is burnt to supply the energy demand, as shown in Figure 3,  $\eta$  can be assigned the number of 85% based on experiences [16], which accounts for heat losses of radiation, enthalpy in hot exhaust flue gas and incomplete combustion.

To evaluate whether the energy contained in the volatile fraction and in the char can satisfy  $Q_s$ , the LHVs of both the volatile fraction and the char were measured. The LHVs of char and condensed oil are tested by a bomb-calorimeter. The components of non-condensable gas are analysed with the help of a gas chromatograph and its LHV is obtained by summing up the LHVs of each component and multiplying its proportion [21]. The LHVs of char and the volatile fraction and their energy proportions are summarized in Table 3.

Based on data in Table 3 and Equation (6), the average required value of  $Q_s$  and its difference between the LHV of the volatile fraction is shown in Figure 6.

From Table 3 and Figure 6 it can be seen that the volatile fraction alone in most cases cannot provide enough energy for pyrolysis. It can be also noticed that energy demand increases as pyrolysis temperature rises, but the sum of energy carried by both the volatile fraction and char remains constant due to the transference of energy from char to the volatile fraction. The proportion of energy in the volatile fraction rose with temperature and was depending on the LHV of the dried sewage sludge. It was higher than 70% of the total energy for all three sludge samples when the pyrolysis temperature was higher than 500 °C. Theoretically, the sum of volatile energy and the char energy can meet the energy requirement for the pyrolysis process for all the pyrolysis tests presented here.

As shown in Figure 6, the energy of the volatile fraction produced under 400 °C for all samples was not enough to meet the energy demand for the pyrolysis process. To increase the proportion of energy in the volatile fraction, a higher pyrolysis temperature is required, for example, 450 °C or even higher. However, to make energy balance, char gasification is an alternative to an increased pyrolysis temperature. Part or the whole char was gasified to generate fuel gas as a complementary fuel to the

volatile fraction when the pyrolysis temperature was below 450 °C. The char obtained under 400 °C was used for gasification experiments. Table 4 shows gas production and the LHV of the produced gas.

Combining data in Tables 3 and 4, it can be seen that when the char was gasified to produce fuel gas, the sum of energy from the pyrolysis volatile fraction and gas could meet the energy demand for the pyrolysis process, theoretically. According to the results presented, when the moisture content of sewage sludge is around 80 wt%, the LHV of dried sludge should be higher than 14.65 MJ/kg and all the char produced from pyrolysis should be gasified in order to ensure energy balance of this combined process.

If the char is to be used in other applications, auxiliary energy should be provided when the pyrolysis temperature is 400 °C. The amount of auxiliary energy will be reduced by increasing the pyrolysis temperature, since the proportion of energy in the volatile fraction will rise. To use the char in other applications and the volatile fraction as an energy supplier without auxiliary fuel, the critical LHV of dried sludge with different initial moisture content should satisfy the following equation:

$$Q_c = (Q_1 + Q_2 + Q_3)/[\eta \times \eta_v \times (1 - M)] \quad (7)$$

where  $Q_c$  is the critical LHV(db) of sewage sludge;  $\eta_v$  is the proportion of energy in the volatile fraction, as listed in Table 3;  $\eta$  is the energy utilization efficiency, as defined above; and  $M$  is the moisture content of sewage sludge. Figure 7 shows some results of the critical LHV(db) requirement for different initial moisture contents based on Equation (7). It can be seen that, as initial moisture content decreased and pyrolysis temperature increased, the required critical LHV(db) of sludge decreased.

In general, the LHV(db) of sewage sludge is around 15 MJ/kg [22]. From data shown in Figure 7, energy balance can be expected for the pyrolysis process by burning the volatile fraction alone when the initial moisture content is lower than 75% and the pyrolysis temperature is higher than 450 °C. While part of the char can be gasified to serve as supplemental energy if the initial moisture is 80%. Since in most cases the sewage sludge has an initial moisture content of 80%, the LHV(db) should be higher than 18 MJ/kg and

the pyrolysis temperature higher than 450 °C if the volatile fraction alone is used as an energy supply without auxiliary fuel. On the other hand, when the initial moisture content decreased to 65wt%, the critical LHV (db) of sewage sludge accordingly decreased to 10 MJ/kg or below.

#### **4. Conclusion**

Combined pyrolysis and gasification was investigated to treat high moisture content sewage sludge with the target of energy self-balance. Three samples, SS1, SS2 and SS3, were subjected to pyrolysis, and then the mass distribution and energy balance were discussed. Char produced from sewage sludge pyrolysis under 400 °C was gasified to produce fuel gas. The fuel gas can be used as auxiliary fuel to the pyrolysis process. Based on the above investigation, for sewage sludge with initial moisture content around 80 wt%, the following conclusions can be generally drawn.

- (1) When the LHV(db) is lower than 14.65 MJ/kg, auxiliary fuel or energy is needed for the combined process.
- (2) The pyrolysis process can reach energy self-sufficiency when the LHV(db) is higher than 18 MJ/kg; all the char produced can be used in other applications when the pyrolysis temperature is higher than 450 °C.
- (3) Part or all of the char should be gasified to produce additional energy to pyrolysis process if the LHV(db) ranges between 14.65 and 18MJ/kg. The char consumption needed for this purpose will be lower if the pyrolysis temperature increases.
- (4) As the initial moisture content decreases from 80 to 65 wt%, the critical LHV(db) of sludge decreases accordingly from above 18 MJ/kg to below 10 MJ/kg.

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Table 1

Table 1. Proximate analysis and ultimate analysis of sewage sludge samples.

Samples	Proximate analysis					Ultimate analysis (dry basis)				
	Moisture content %	Volatile content % (dry basis)	Fix carbon content % (dry basis)	Ash content % (dry basis)	LHV (MJ/kg) (dry basis)	C %	H %	N %	O %	S %
SS1	77.8	58.9	10.86	30.14	14.92	30.9	5.06	5.42	19.7	0.67
SS2	80.2	68.5	16.40	23.27	18.45	40.5	5.84	6.84	14.1	1.28
SS3	78.4	55.3	19.29	25.38	15.65	37.5	5.85	5.36	12.3	0.61

Table 2

Sample	$Q_1$ (MJ/kg-ss)*	$Q_2$ (MJ/kg-ss)	$Q_3$ (MJ/kg-ss)	Total energy consumption (MJ/kg-ss)
SS1-400 °C**	1.94	0.14	0.046	2.146
SS1-450 °C		0.15	0.071	2.161
SS1-500 °C		0.17	0.096	2.206
SS1-550 °C		0.19	0.106	2.236
SS2-400 °C	2.02	0.12	0.091	2.231
SS2-450 °C		0.13	0.099	2.249
SS2-500 °C		0.15	0.124	2.294
SS2-550 °C		0.16	0.153	2.333
SS3-400 °C	1.98	0.16	0.069	2.209
SS3-450 °C		0.18	0.073	2.233
SS3-500 °C		0.20	0.094	2.274
SS3-550 °C		0.22	0.151	2.351

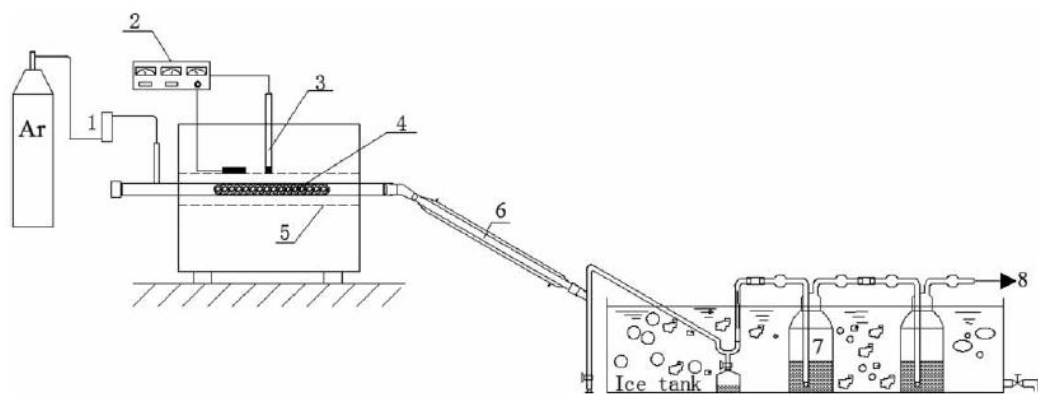
\*The unit of kg-ss means per kg of wet sewage sludge; \*\*the pyrolysis temperature for the sample SS1 was 400 °C.

Table 3

Sample	LHV of char (MJ/kg)	Energy in char by LHV (MJ/kg-ss)	Energy in volatile by LHV (MJ/kg-ss)	Sum of energy (MJ/kg-ss)	Proportion of energy in volatile (%)	Energy demanded for pyrolysis process ( $Q_s$ , MJ/kg-ss)
SS1-400 °C	11.96	1.427	1.885	3.312	56.91	2.525
SS1-450 °C	9.43	1.061	2.251	3.312	67.97	2.542
SS1-500 °C	9.00	0.969	2.343	3.312	70.73	2.595
SS1-550 °C	8.05	0.873	2.439	3.312	73.65	2.631
SS2-400 °C	11.04	1.187	2.500	3.686	67.81	2.625
SS2-450 °C	10.96	0.978	2.709	3.686	73.48	2.646
SS2-500 °C	10.53	0.905	2.781	3.686	75.44	2.699
SS2-550 °C	10.14	0.821	2.866	3.686	77.74	2.745
SS3-400 °C	13.96	1.105	2.276	3.380	67.32	2.599
SS3-450 °C	12.01	1.072	2.309	3.380	68.30	2.627
SS3-500 °C	11.77	1.009	2.372	3.380	70.16	2.675
SS3-550 °C	11.02	0.958	2.422	3.380	71.66	2.766

Table 4

Sample	LHV of char (MJ/kg-char)	Fuel gas yield (m <sup>3</sup> /kg-char)	LHV of fuel gas (MJ/m <sup>3</sup> -gas)	Sum of energy supplied from volatile and fuel gas
SS1-400 °C	11.89	1.612	5.31	2.92(MJ/kg-ss)
SS2-400 °C	13.96	1.844	5.65	3.36(MJ/kg-ss)
SS3-400 °C	11.04	1.608	5.01	3.08(MJ/kg-ss)



1, flow meter; 2, temperature control unit; 3, thermal couple; 4, sewage sludge; 5, heating rod; 6, water cooler; 7, oil collector; 8, gas to gas chromatographer.

Figure 1

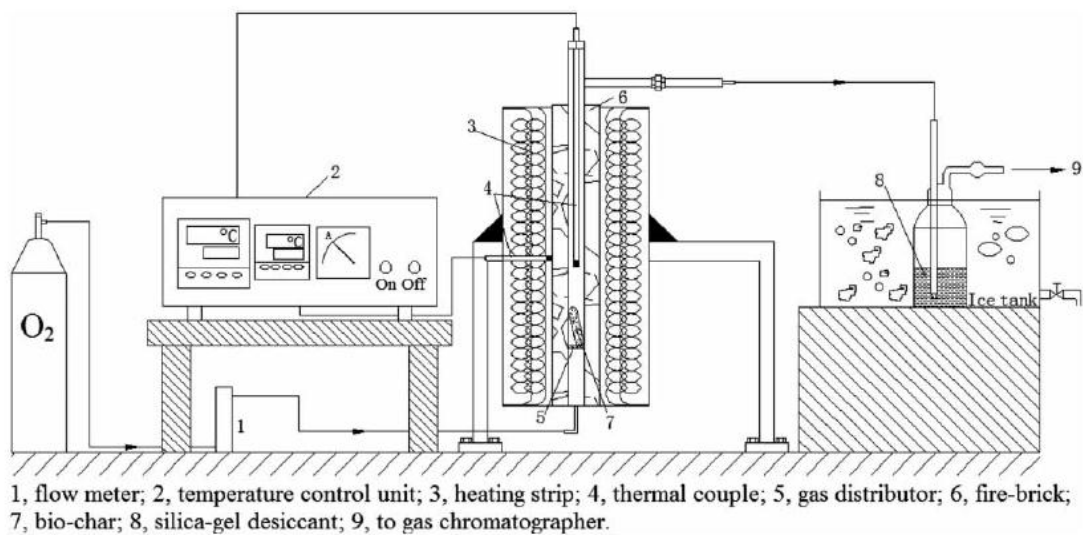


Figure 2

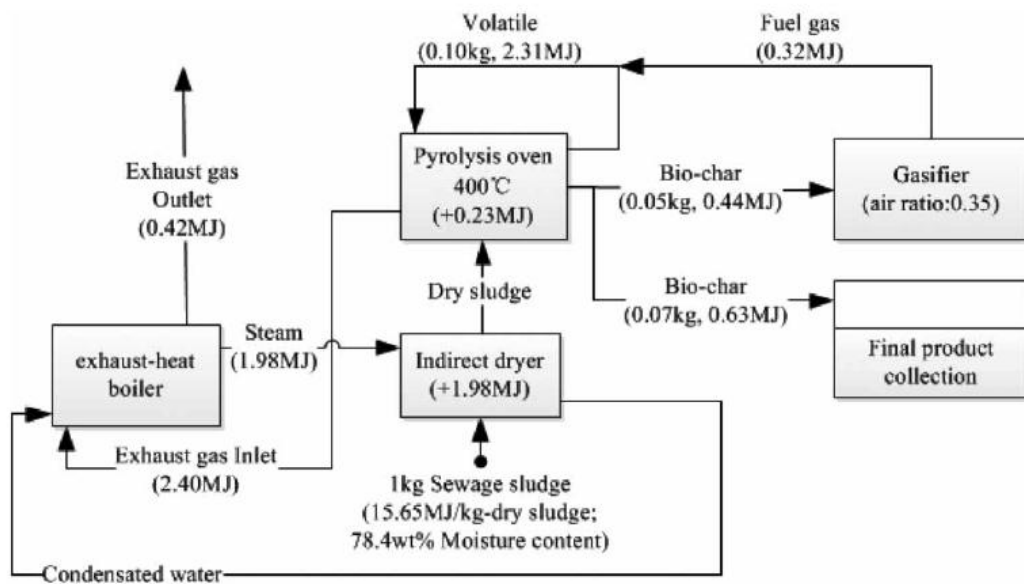


Figure 3



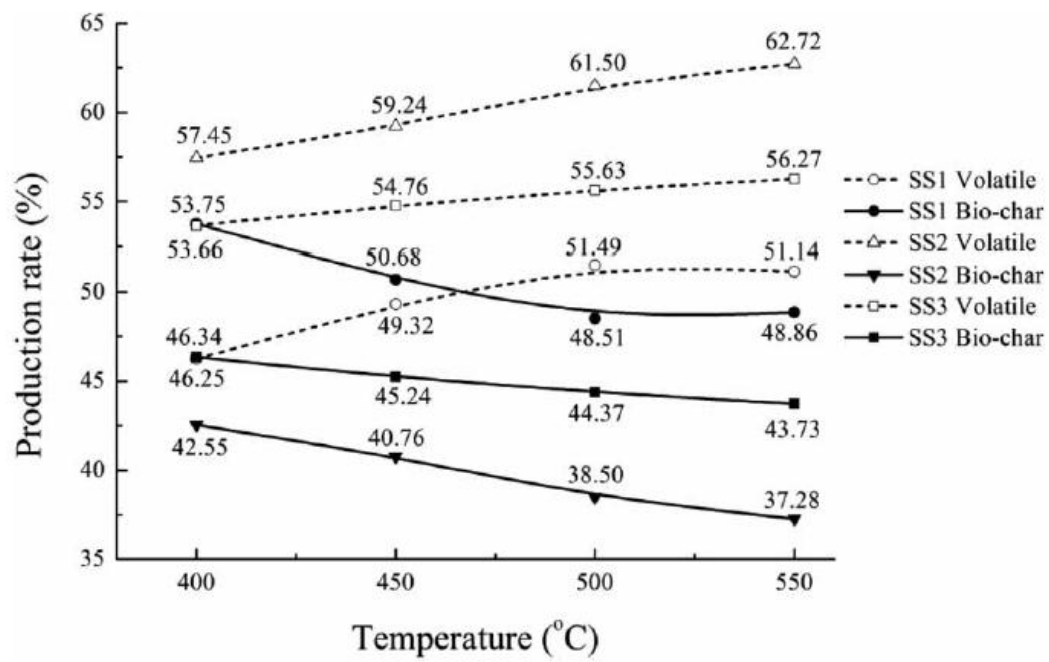


Figure 4

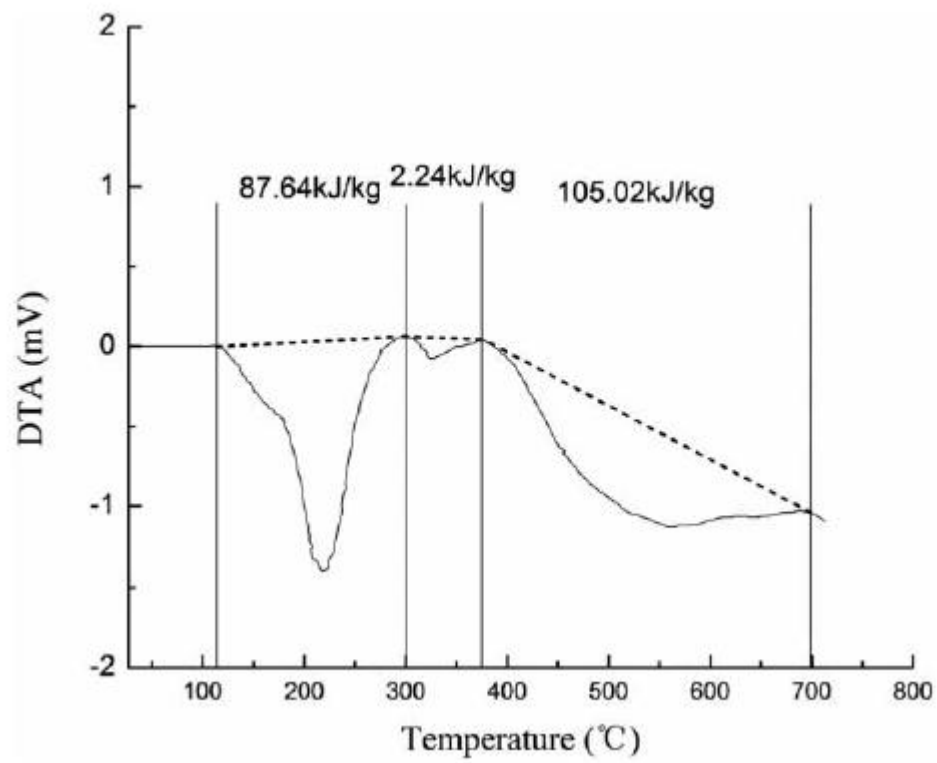


Figure 5

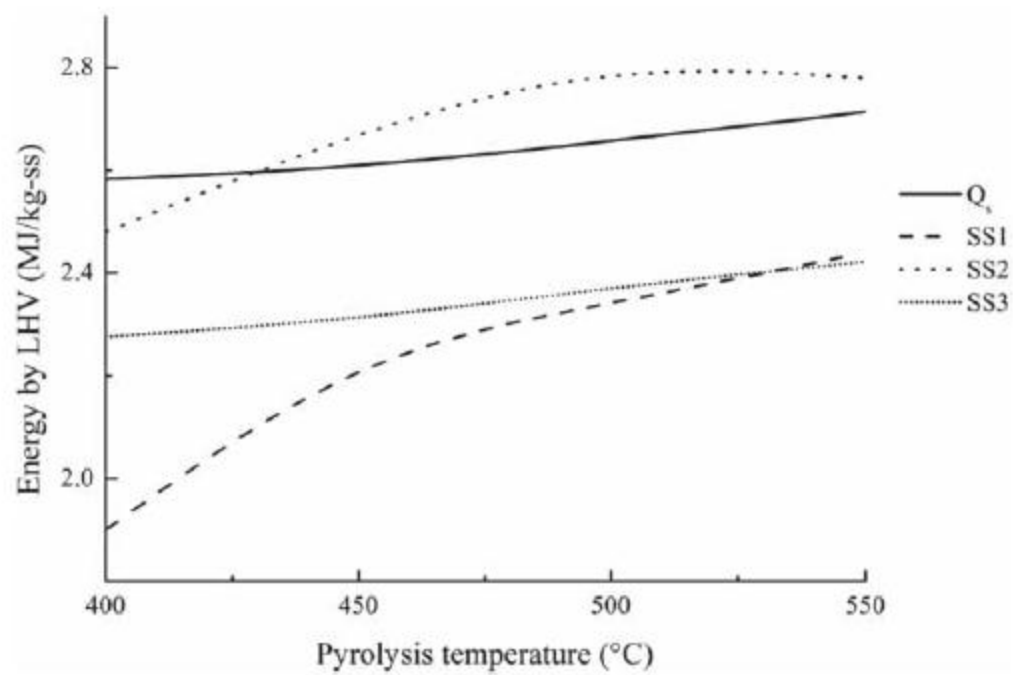


Figure 6

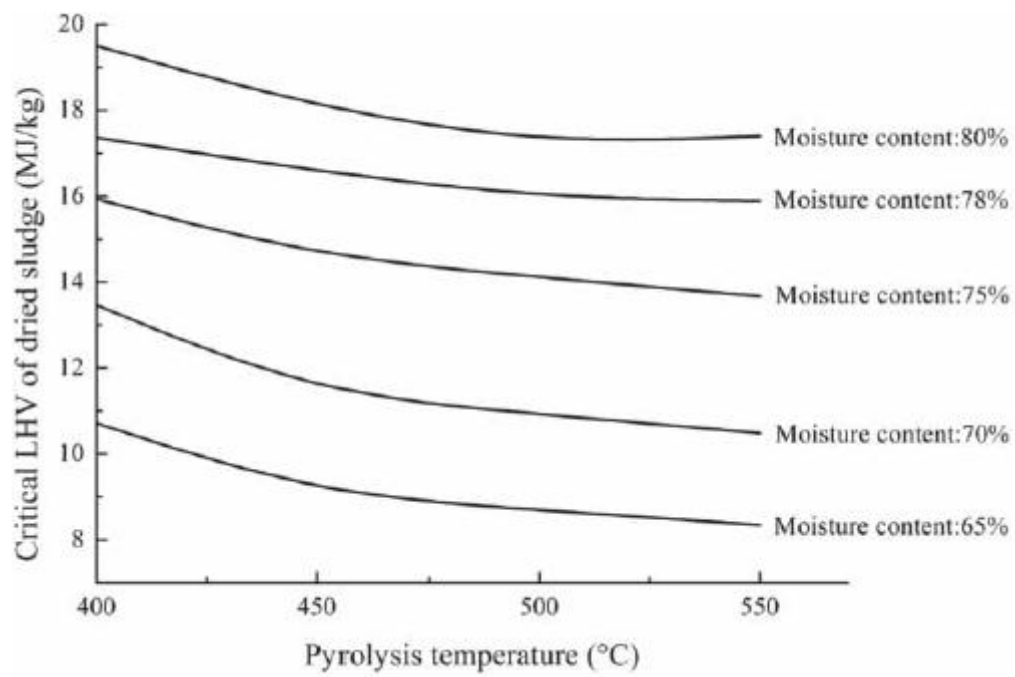


Figure 7