

Contents lists available at ScienceDirect

Bioresource Technology

journal homepage: www.elsevier.com/locate/biortech



Short Communication

Potential method for gas production: High temperature co-pyrolysis of lignite and sewage sludge with vacuum reactor and long contact time



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HIGHLIGHTS

- Gas production method based on lignite/sewage sludge co-pyrolysis was reported.
- Beneficial synergetic effect on gas yield was clearly observed.
- Mechanism of gas production during co-pyrolysis was discussed.

ARTICLE INFO

Article history: Received 21 October 2014 Received in revised form 24 November 2014 Accepted 25 November 2014 Available online 3 December 2014

Keywords: Co-pyrolysis Gas production Lignite Sewage sludge Synergetic effect

ABSTRACT

Lignite and sewage sludge were co-pyrolyzed in a vacuum reactor with high temperature (900 °C) and long contact time (more than 2 h). Beneficial synergetic effect on gas yield was clearly observed. Gas yield of blend fuel was evidently higher than that of both parent fuels. The gas volume yield, gas lower heating value (LHV), fixed carbon conversion and H_2/CO ratio were 1.42 $Nm^3/kg_{(blend\ fuel)}$, 10.57 MJ/Nm^3 , 96.64% and 0.88% respectively, which indicated this new method a feasible one for gas production. It was possible that sewage sludge acted as gasification agents (CO_2 and H_2O) and catalyst (alkali and alkaline earth metals) provider during co-pyrolysis, promoting CO_2 -char and H_2O -char gasification which, as a result, invited the improvement of gas volume yield, gas lower heating value and fixed carbon conversion.

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1. Introduction

Lignite is a kind of low rank coal which cannot be utilized directly due to its low energy quality (high moisture content and low heating value) and negative environmental contributions (CO₂ and pollutant emissions). Co-utilization of lignite and other renewable fuels can solve this problem to a large extent. And sewage sludge is one of the fuels that can be co-processed with lignite. Co-pyrolysis and co-gasification, deemed as promising methods, were investigated by many researchers (Idris et al., 2010; Park et al., 2010; Saw and Pang, 2013; Kern et al., 2013). Generally, gas production is the main purpose of gasification, while pyrolysis, usually, is applied to produce solid and liquid fuels (char and tar).

In our previous work (Yang et al., 2014), lignite and biomass (rice husk) were co-pyrolyzed in a vacuum reactor with high temperature (900 °C) and long contact time (more than 2 h). We surprisingly found that the gas yield of blend fuel was evidently higher than that of parent fuels. This beneficial synergetic effect

on gas yield indicated a potential new method for gas production based on co-pyrolysis under the special pyrolysis conditions.

In this work, co-pyrolysis experiments were carried out for two purposes. One is to observe synergetic effects during co-pyrolysis of lignite and sewage sludge. Product yields and compositions of char and gas were studied to analyze synergetic effects. The other is to investigate the feasibility and mechanism of this new gas production method. High yields of volatiles (CO₂ and H₂O) from pyrolysis of sewage sludge were expected to facilitate H₂O-char and CO₂-char gasification during co-pyrolysis. And AAEM (alkali and alkaline earth metals) contained in sewage sludge was also anticipated to act as catalyst and promote gasification process in further (Rizkiana et al., 2014; Veraa and Bell, 1978). Gas yield, gas lower heating value (LHV), fixed carbon conversion and H₂/CO ratio were used to evaluate this method for gas production.

2. Methods

2.1. Experimental samples

Anaerobically digested sewage sludge from a wastewater treatment plant of Tianjin city and lignite from Neimeng Province,

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China were used in this study. The samples were dried in a drying oven at 105 °C for 24 h, and then ground into small particles to make sample granularity arrive at 80 meshes. Sewage sludge and lignite particles were mixed and blended complying with the ratio of 1:1 in weight. Proximate and ultimate analyses of samples are listed in Table 1.

2.2. Experimental apparatus and procedure

All pyrolysis experiments were carried out on the fixed bed pyrolysis system shown in Fig. 1. The inner size of the fixed bed reactor is $0.3 \times 0.3 \times 0.4$ m (height). Air in the fixed bed reactor was replaced by N₂ after samples were loaded. A vacuum pump then pumped N₂ out, providing a vacuum space for pyrolysis experiments. The initial total pressure in the reactor was around 5 kPa (-95 kPa). Final total pressures were below 30 kPa (-70 kPa) when pyrolysis processes ended according to actual experiments. The reactor was kept closed during the whole pyrolysis process. Approximately 10 g samples were used in each experiment. Each sample was heated, under the control of heating controller, from ambient temperature to 900 °C at a rate of 10 °C/ min and kept at 900 °C for 2 h. Volatile products (tar and gas) were pumped out. Tar was eliminated by condensing (empty traps in ice bath), washing (traps with hexane, dichloromethane and isopropanol), and filter (activated carbon filter). Non-condensable gas was measured by a cumulative volume flow meter and collected by gas bags. The char was collected and weighed when the temperature in the reactor was lower than 100 °C. Each experiment in this study was repeated three times.

2.3. Characterization

The proximate and ultimate analyses were taken place in a tube furnace and Vario MACRO CHN/CHNS element analyzer respectively according to GB483-87 in China. Inorganic materials in lignite and sewage sludge were analyzed by inductively coupled plasma-optical emission spectroscopy (ICP-OES). The nitrogen adsorption and desorption at 77.35 K using char samples were applied for BET analysis. The pyrolysis gas was analyzed by Agilent

Table 1 Analysis of raw material.

Description	Lignite	Sewage sludge
Ultimate analysis (dry basis)		
Carbon (wt.%)	53.65	41.5
Hydrogen (wt.%)	4.12	4.71
Nitrogen (wt.%)	0.41	4.28
Sulfur (wt.%)	0.46	1.11
Oxygen (wt.%)	35.93	11.34
Proximate analysis (dry basis)		
Moisture (wt.%)	_	9.56
Volatiles (wt.%)	56.98	50.15
Fixed carbon (wt.%)	37.59	3.23
Ash (wt.%)	5.43	37.06
Inorganic composition analysis (ICP-OE	S)	
MgO (wt.%)	0.58	0.96
CaO (wt.%)	1.58	11.73
Na ₂ O (wt.%)	0.05	1.78
K ₂ O (wt.%)	0.06	1.89
SiO ₂ (wt.%)	1.08	6.72
TiO ₂ (wt.%)	0.01	0.22
Al_2O_3 (wt.%)	1.04	5.35
Fe ₂ O ₃ (wt.%)	0.71	2.9
MnO (wt.%)	0.01	0.03
P ₂ O ₅ (wt.%)	0.01	2.52
SO ₃ (wt.%)	0.29	2.07

^{-,} Not detected.

7890A GC/TCD with a Molsieve 5A packed column (6 ft \times 1/8" \times 2 mm).

2.4. Calculations

Average relative deviation between experimental value and calculated value was employed to analyze synergetic effects during co-pyrolysis. In this study, the blend ratio is a fixed value (1:1). The calculated values can be calculated by Eqs. (1)–(3):

$$C_{\text{yield},i} = \frac{Y_{\text{lignite},i} + Y_{\text{ss},i}}{2} \tag{1}$$

$$C_{\text{mfra},m} = \frac{F_{\text{Lgas},m} \times Y_{\text{lignite,gas}} + F_{\text{ssgas},m} \times Y_{\text{ss,gas}}}{Y_{\text{lignite,gas}} + Y_{\text{ss,gas}}}$$
(2)

$$C_{\text{char},n} = \frac{E_{\text{Lchar},n} \times Y_{\text{lignite,char}} + E_{\text{sschar},n} \times Y_{\text{ss,char}}}{Y_{\text{lignite,char}} + Y_{\text{ss,char}}}$$
(3)

where $C_{\rm yield,i}$ is the calculated yield of i, including char and gas volume. $Y_{\rm lignite,i}$ and $Y_{\rm ss,i}$ are lignite and sewage sludge experimental yields of i. $C_{\rm mfra,m}$ is the calculated molar fraction of gas component m. $F_{\rm Lgas,m}$ and $F_{\rm ssgas,m}$ are molar fractions of gas component m in lignite gas and sewage sludge gas. $Y_{\rm lignite,gas}$ and $Y_{\rm ss,gas}$ are experimental volume yields of lignite gas and sewage sludge gas. $C_{\rm char,n}$ is the calculated value of char's characteristic n, including moisture content, volatiles content, fixed carbon content and ash content. $Y_{\rm lignite,char}$ and $Y_{\rm ss,char}$ are experimental yields of lignite char and sewage sludge char. $E_{\rm Lchar,n}$ and $E_{\rm sschar,n}$ are experimental values of lignite char's and sewage sludge char's characteristic n. The average relative deviations can be calculated by Eq. (4):

$$\label{eq:average} \text{Average relative deviation} = \frac{|\text{Blend experimental value} - \text{Calculated value}|}{\text{Calculated value}} \\ \times 100\% \tag{4}$$

Besides gas yield of the blend, gas lower heating value (LHV) and fixed carbon conversion were applied to evaluate this new gas production method. They can be calculated by Eqs. (5) and (6):

$$LHV_{blend gas} = \sum F_m LHV_m \tag{5}$$

$$FCC = \frac{0.5 \times FC_{lignite} + 0.5 \times FC_{ss} - Y_{blend\ char} \times FC_{blend\ char}}{0.5 \times FC_{lignite} + 0.5 \times FC_{ss}} \times 100\%$$

where LHV_{blend gas} is the lower heating value of blend gas. F_m is the molar fraction of combustible gas component m. LHV $_m$ is the lower heating value of combustible gas component m. FCC is the fixed carbon conversion. FC_{lignite}, FC_{ss} and FC_{blend char} are fixed carbon contents of lignite, sewage sludge and blend char. Y_{blend char} is the yield of blend char.

3. Results and discussion

3.1. Synergetic effects analysis

3.1.1. Product yields

The yields of lignite char, sewage sludge char and blend char were 0.22, 0.35 and 0.24 g/g, respectively. The calculated value was 0.32 g/g. The volume yields of gases derived from pyrolysis of lignite, sewage sludge and blend were 1322.55, 660.44 and 1421.37. And the calculated value was 991.49 mL/g. Beneficial synergetic effect on gas volume yield was clearly observed. Gas volume yield of the blend was higher than that of both lignite and sewage sludge. The average relative deviation between the blend gas volume yield and calculated value was 43.4%. The char

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