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Study of factors affecting syngas quality and their interactions in fluidized bed gasification of lignite coal

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HIGHLIGHTS

- ▶ The effect of three operating factors on syngas quality in fluidized bed lignite gasifier is studied.
- ▶ The syngas quality was defined based on conversion, H_2/CO, CH_4/H_2, yield, and gasifier efficiency.
- ▶ Low coal feedrate, average particle size and high steam/O_2 are favorable to high conversion rates.
- ▶ The steam/O_2 ratio has the greatest effect on the H_2/CO and CH_4/H_2 ratio.

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ABSTRACT

A series of experiments has been designed and conducted to study the effect of three operating factors, namely, coal feedrate, coal particle size, and steam/O_2 ratio, and their interactions on the quality of syngas produced from fluidized bed gasification of lignite coal. The quality of syngas is evaluated based on five indices including carbon conversion, H_2/CO ratio, CH_4/H_2 ratio, gas yield, and gasification efficiency. The design of experiment tool based on the response surface methodology (RSM), which is believed to be more accurate than the common one-factor-at-a-time approach, is used to facilitate the comparison of the effect of all factors. The factors are tested in the ranges of 0.036–0.063 g/s, 70–500 μm, and 0.5–1.0, for coal feedrate, coal particle size, and steam/O_2 ratio, respectively. The carbon conversion, H_2/CO ratio, CH_4/H_2 ratio, gas yield, and gasification efficiency are found to range from 91% to 97%, 0.776 to 1.268, 0.0517 to 0.0702, 3.4 to 3.7 m^3 gas/kg coal, and 56% to 67%, respectively. The effects of individual operating factors and their interactions on each syngas quality index are discussed using RSM tools. A set of operating conditions to achieve syngas with a desired quality for different applications is also proposed by optimization of the response surface of each index.

1. Introduction

Declining supplies of crude oil in combination with increased environmental pressure to reduce greenhouse gas emissions from coal-fired power plants has led to renewed interest in gasification as a clean-coal technology. Currently, about 75% of power generation in China, more than 50% in the US, and nearly 40% of the world production of power relies on coal [1]. Canada presently has 51 coal-fired power plant units producing 19% of the country's electricity and 13% of its greenhouse gas emissions. However, 33 of those plants are expected to reach the end of their economic lives by 2025. According to more strict new environmental regulations announced by the Canadian government, these coal-fired power generators must either reduce their carbon emissions to the equivalent of a natural gas plant or be retired. In accordance with the Canada's Clean Coal Technology Roadmap [2] and CO_2 Capture and Storage Technology Roadmap [3], clean coal research is ongoing throughout Canada, but the focus is not currently on the utilization of low-rank sub-bituminous and lignite coals. The focus of the current study is the gasification of lignite coal, which exists in significant quantities in certain regions of Canada and the world.

Using gasifiers instead of combustors has many advantages, including producing syngas with sufficient quality to be used in specialized downstream units such as clean fuel combustion, production of Fischer–Tropsch liquids, and fuel cells, plus a low-cost and concentrated CO_2 ready for underground sequestration. Moreover, it provides a hot gas that can be used in integrated gasification combined cycles (IGCCs) for power generation. However, high degree of reliability required for commercial use of gasifica-

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Nomenclature

b_0	intercept of the response surface polynomial
b_i	linear coefficients in the response surface polynomial
b_{ij}	interaction coefficients in the response surface polynomial
b_{ii}	quadratic coefficients in the response surface polynomial
GE	gas efficiency (%)
GY	gas yield (m^3 gas/kg coal)
HHV_c	coal higher heating value (MJ/m^3)
HHV_s	syngas higher heating value (MJ/m^3)
k	number of factors
m_c	coal mass flow to the gasifier (g/s)
M_{CS}	summation of mole flow of carbon in all carbon-bearing components in syngas (mol/s)
n_c	number of center runs in central composite design
N	number of designed experiments
Q_s	syngas volumetric flowrate (m^3/s)
U_c	fraction of carbon in coal from ultimate analysis
X_C	carbon conversion
x_i, x_j	normalized values of the response variables
X_{orig}	original version of the operating variables
X_{norm}	normalized version of the operating variables
Y	predicted response

tion is not yet supported by common types of gasifier reactors. Thus, gasification is not yet economically and operationally attractive for the power industry [4] and more research is needed to facilitate the process and improve the desirability of the gasification process. Various types of gasifiers such as moving bed, entrained flow, and fluidized beds have been employed by industry. All of these technologies were invented in Germany prior to World War II. Lurgi invented the moving bed, Kopper-Totzek (K-T) invented the entrained flow, and Winkler invented the fluidized bed gasifier [5]. Problems such as high tar yields in the product gas [6], the inability to maintain uniform radial temperature, and slagging in large installations [7] make moving bed gasifiers relatively less desirable. In entrained flow gasifiers, the mixture of air and solids (biomass or coal) is blown into the reaction chamber. Entrained-flow gasifiers overcome some of the deficiencies of moving-bed gasifiers but do not provide the flexibility of fluidized bed gasifiers. Because fluidized bed reactors operate at lower temperatures (800–1000 °C) and have less slag handling and ash fusion problems [8], the above-mentioned objectives can be met by using fluidized beds. Compared to other types of reactors, relatively large fluidized bed reactor vessels can be built and operated, so that comparatively fewer reactors would be required in a commercial plant [9]. Furthermore, the possibility of using sorbents for sulfur removal in the fluidized bed gasifier also lowers or eliminates downstream use of the expensive desulfurization units. The maximum bed temperature of a fluidized bed gasifier is limited by the ash softening temperature, at which ash begins to stick to other particles and solid surfaces. However, superior mixing and heat transfer make it possible to operate at lower temperatures.

Coal gasification is a two-stage process where rapid initial pyrolysis both de-volatilizes releasing volatiles with high reactivity and produces a char that reacts more slowly [10]. The pyrolysis is believed to occur on the order of seconds after injection of pulverized coals into the bed. The gasification step that comes next includes heterogeneous reactions between char and gases and homogeneous reactions between gas components. Different studies have been performed on coal gasification in fluidized beds.

Watkinson et al. [11] carried out gasification experiments with different coals in a fluidized bed with steam and air and found that gas heating values were between 1.6 and 4.2 MJ/m^3. Similar results were found by Kawabata et al. [12] and Saffer et al. [13]. Tomeczek et al. [14] reported gas heating values between of 2.9–3.5 MJ/m^3 using air and 4.1–4.5 MJ/m^3 using steam–air mixtures. Ocampo et al. [15] experimented with Colombian lignite coals for steam/coal ratios of 0.58 and 0.71 and found gas heating values of 2.7 and 3.3 MJ/m^3. The air/coal ratios were respectively 2.4 and 2.6 for these experiments. They attributed their low gas heating values to the high rate of particle entrainment as a consequence of the short freeboard section in their fluidized bed setup.

In three consecutive works, Purdy et al. [16,17] and Rhinehart et al. [18] studied the effect of bed temperature, coal feedrate, and steam/carbon ratio on the gasification of coal with different ranks in a 15.2 cm diameter fluidized bed under around 8 bar pressure. For a 0.5 mm size de-volatilized bituminous coal gasified at 925 and 1025 °C, Purdy et al. [16] found the bed temperature and steam/carbon ratio to be the most important factors in determining the gas yield. However, they adjusted the bed temperature by regulating the oxygen flowrate, which consequently changes the rate of combustion reactions and gas and solid residence times. Thus, the operating conditions were not precisely controlled in their experiments. Rhinehart et al. [18] used lignite coal of 0.17–0.91 mm size and achieved an H_2/CO molar ratio of 1.5–4.5 and a carbon conversion of from 70% to nearly 100%. Kim et al. [19] studied the gasification of a sub-bituminous coal in a down-flow reactor (downer). By increasing the steam/coal ratio from 0.23 to 0.86, they observed a drop in the calorific value of syngas from 9.0 to 6.4 MJ m^3 due to the reduction of combustible gas and an increase of H_2/CO ratio and decrease of CH_4/H_2 ratio due to moving the water–gas shift equilibrium towards H_2 production. A similar trend was reported for bituminous and anthracite coals by Zhou [20].

In almost all of these experiments, the oxygen content or the O_2/coal ratio was varied along with the operating variables due to changing coal feedrate (when the steam/O_2 was constant) or changing steam/coal ratio (when the coal feedrate was constant). Due to the changing oxygen content of the system, the effect of carbon or gas combustion was not isolated from gasification reactions. For example in Kim et al. [19], with an increasing coal feedrate of 5.0–9.3 kg/h, the volume percentage of H_2 and non-methane hydrocarbons increased due to an increase in supply of volatile matter, whereas CO and CO_2 concentrations decreased due to the decrease of O_2/coal ratio and availability of oxygen for combustion. This leads to the increase of calorific value of the product gas and decrease of gas yield. As can be seen, the changing syngas quality is mostly due to the change in O_2 availability to consume combustible gases and not due to the production of combustible gases by reactions. These uncontrolled effects make it difficult to interpret the experimental data and extract the true effects of the operating variables. In the present work, the O_2/coal is kept constant to preclude the effect of more oxygen availability on changing the gas composition.

Although the combustion and gasification of pulverized coals in fluidized beds have been widely investigated in the past years, few data are available on the effect of particle size on coal properties and reactivity. It has been reported that the volatile matter measured by the ASTM standard depends on particle size [21,22], and some studies suggest that the content of ash and fixed carbon are also significantly dependent on the particle size [23–36]. Kök