#### Fuel xxx (2012) xxx-xxx



# Fuel



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

Shayan Karimipour<sup>a</sup>, Regan Gerspacher<sup>b</sup>, Rajender Gupta<sup>a</sup>, Raymond J. Spiteri<sup>c,\*</sup>

<sup>a</sup> Department of Chemical & Materials Engineering, University of Alberta, Edmonton, AB, Canada T6G 2V4

<sup>b</sup> Department of Chemical Engineering, University of Saskatchewan, Saskatoon, SK, Canada S7N 5C9

<sup>c</sup> Department of Computer Science, University of Saskatchewan, Saskatoon, SK, Canada S7N 5C9

### 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<sub>2</sub>/CO, CH<sub>4</sub>/H<sub>2</sub>, yield, and gasifier efficiency.
- ▶ Low coal feedrate, average particle size and high steam/O<sub>2</sub> 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.

### ARTICLE INFO

Article history: Received 3 March 2012 Received in revised form 7 June 2012 Accepted 7 June 2012 Available online 27 June 2012

Keywords: Lignite coal Gasification Fluidized bed Design of experiments

# 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<sub>2</sub> 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  $\mu$ m, and 0.5–1.0, for coal feedrate, coal particle size, and steam/O<sub>2</sub> ratio, respectively. The carbon conversion, H<sub>2</sub>/CO ratio, CH<sub>4</sub>/H<sub>2</sub> 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<sup>3</sup> 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.

© 2012 Elsevier Ltd. All rights reserved.

### 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<sub>2</sub> 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<sub>2</sub> 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-



<sup>\*</sup> Corresponding author. Tel.: +1 306 966 2909; fax: +1 306 966 4884. *E-mail address:* spiteri@cs.usask.ca (R.J. Spiteri).

<sup>0016-2361/\$ -</sup> see front matter @ 2012 Elsevier Ltd. All rights reserved. http://dx.doi.org/10.1016/j.fuel.2012.06.052

2

S. Karimipour et al./Fuel xxx (2012) xxx-xxx

bo b <sub>i</sub> b <sub>ij</sub> b <sub>ii</sub> GE GY HHV <sub>c</sub> HHV <sub>s</sub> k m <sub>c</sub>	intercept of the response surface polynomial linear coefficients in the response surface polynomial interaction coefficients in the response surface polyno- mial quadratic coefficients in the response surface polyno- mial gas efficiency (%) gas yield (m <sup>3</sup> gas/kg coal) coal higher heating value (MJ/m <sup>3</sup> ) syngas higher heating value (MJ/m <sup>3</sup> ) number of factors coal mass flow to the gasifier (g/s)	M <sub>CS</sub> n <sub>c</sub> N Q <sub>s</sub> U <sub>c</sub> X <sub>c</sub> X <sub>i</sub> , X <sub>j</sub> X <sub>orig</sub> X <sub>norm</sub> Y	summation of mole flow of carbon in all carbon-bearing components in syngas (mol/s) number of center runs in central composite design number of designed experiments syngas volumetric flowrate (m <sup>3</sup> /s) fraction of carbon in coal from ultimate analysis carbon conversion normalized values of the response variables original version of the operating variables normalized version of the operating variables 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<sup>3</sup>. 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<sup>3</sup> using air and 4.1–4.5 MJ/m<sup>3</sup> 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<sup>3</sup>. 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<sub>2</sub>/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<sup>3</sup> due to the reduction of combustible gas and an increase of H<sub>2</sub>/CO ratio and decrease of CH<sub>4</sub>/H<sub>2</sub> ratio due to moving the water-gas shift equilibrium towards H<sub>2</sub> 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<sub>2</sub>/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<sub>2</sub> 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<sub>2</sub> 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 et al. [27] reported an increase in the residue materials left at the

end of the combustion process with decreasing the particle size that also caused lower carbon conversion. Yu et al. [28] examined a bituminous coal with 20.4-177.1 µm particle diameters and found that the fraction of fixed carbon decreases with decreasing particle size, a result that is consistent with the results of Kök et al. [24,27]. Zhang et al. [29] reported the decrease of the maximum rate of volatile matters release and mass loss with increasing the particle size. Hanson et al. [30] found that moisture does not change with size, but ash content decreases with decreasing particle size. However, within the range of particle sizes they investigated, pyrolysis and gasification were seen to be relatively insensitive to particle size. Generally, combustion reactions are expected to be less catalytic than gasification reactions, and the increase of reactivity due to size reduction and availability of more surface area is reported to be more significant than the decrease in ash content for combustion reactions. Thus, an increase in rate of combustion with size reduction can be always observed. In gasification reactions, these effects plus the change in volatile matter compete with each other and present a more complicated trend that is difficult to predict a priori.

The desired quality of syngas from the gasification process is different for different applications. Accordingly, the operating conditions needed to produce syngas with these qualities may or may not be close to each other. The presence of complicated multi-level interactions between effective factors makes it more difficult to envision the best conditions to get the desired syngas quality for a specific application. The design of experiment tools can be used to determine the operating conditions in favor of one or all of these various applications together with the least number of experiments. The objective of the present work is to study the effect of different operating conditions and their interactions on the syngas quality and find the optimum operating conditions in the operating range studied here to produce syngas with a desired quality. The quality of syngas is defined based on parameters of carbon conversion (X<sub>C</sub>), H<sub>2</sub>/CO ratio, CH<sub>4</sub>/H<sub>2</sub> ratio, gas yield (GY), and gasification efficiency (GE). A complementary objective of this work is to generate a series of reliable experimental data for CFD model tuning and validation. These data can also be very helpful as the starting point for scale-up of the laboratory results to pilot-scale or fullscale gasifier units.

#### 2. Experimental

#### 2.1. Fluidized bed gasifier

The fluidized bed gasifier was made of a cylindrical stainless steel tube with a height of 1.5 m and in which the fluidized bed has an inner diameter of 7 cm and height of 0.5 m, and the freeboard section has 15 cm diameter and 1 m height (Fig. 1). The setup is operable at atmospheric pressure and temperatures up to 850 °C with heating by means of an electric furnace which encapsulates the 7 cm tube. The column was equipped with an external condenser and cyclone. A porous stainless steel plate was used as the distributor. The fluidization air was supplied from building air and controlled with a high-accuracy rotameter. The coal was charged in a pressurized hopper and fed continuously into the reactor 3 cm above the distributor with a screw feeder. A bottom-feed coal injection would give more residence time to char and volatile matter to react at higher temperatures at the bottom and help to reduce the molecular weight of the de-volatilized tar by in-bed reactions and lower the required operating temperatures. The coal feedrate was determined by calibrating the rotation speed of the screw feeder with coal before each experiment and verified before and after measurements of coal mass in the hopper.



Fig. 1. Schematic diagram of the fluidized bed gasifier used in the present work.

The bed temperature and pressure were continuously monitored to achieve steady-state conditions.

#### 2.2. Test material

The bed material was silica sand with a particle density of  $2600 \text{ kg/m}^3$  and Sauter mean diameter of  $250 \text{ }\mu\text{m}$ . The minimum fluidization velocity of the sand particles was calculated to be 0.04 m/s. The height of sand bed was kept constant at 11 cm in all experiments. The coal used for the experiments was lignite coal provided from Boundary Dam mine in Saskatchewan, Canada. The ultimate and proximate analyses and ash composition of the coal are provided in Table 1. The coal particles were ground to three sizes of 70, 285, and 500  $\mu$ m to study the effect of particle size. The moisture of the coal particles was measured before each experiment, and variations were compensated for by adjusting the steam flow rate.

### 2.3. Experimental conditions

The operating variables investigated in the present study and their ranges can be seen in Table 2. During gasification, the particles are continuously fed into the bottom of the reactor 3 cm above the distributor. Steam and oxygen were mixed, preheated to 750 °C, and injected to the bed through the distributor. The rate of air, steam, coal feedrate, and coal particle size were specified for each run by the experimental design. The reactor temperature was manually controlled to 800 °C by changing the temperature of the encapsulating furnace. Two thermocouples in the dense

#### S. Karimipour et al. / Fuel xxx (2012) xxx-xxx

#### Table 1

Characterization of the Boundary Dam lignite coal used in the experiments.

Ultimate analysis	%
Fixed carbon	44.43
Volatile matter	39.02
Moisture	1.3
Ash	15.25
Proximate analysis	
Carbon	58.58
Hydrogen	4.25
Nitrogen	1.26
Oxygen	19.89
Sulfur	0.77
Ash	15.25
Calorific value	
23.23 MJ/kg	
Ash analysis	
SiO <sub>2</sub>	33.3
Al <sub>2</sub> O <sub>3</sub>	18.4
CaO	25.6
MgO	3.0
Fe <sub>2</sub> O <sub>3</sub>	8.1
K <sub>2</sub> O	0.7
Na <sub>2</sub> O	0.5
TiO <sub>2</sub>	0.2
SO <sub>3</sub>	10.2

bed and one immediately above were installed to obtain a profile of bed temperature. The temperatures were recorded every 10 s. Sampling of the outlet gas was performed 20, 40, 60, 70, and 80 min into each run, and the samples were analyzed using an Agilent 7890A Gas Chromatograph (GC). All of the experiments were conducted at atmospheric pressure.

### 3. Design of experiments

Three effective operating variables, namely, coal feedrate (CF), coal particle size (PS), and mass ratio of steam to  $O_2$  (S/O), which

would likely be manipulated in a gasifier, were chosen to be experimentally studied. The variables were varied in the ranges of 0.036–0.063 g/s (dry basis), 70–500 µm, and 0.5–1.0, for CF, PS, and S/O, respectively. These operating ranges were chosen by a series of initial experiments to provide a smooth gasification process in the operable range of our experimental set up. The O<sub>2</sub>/coal mass ratio was kept constant at 0.75 during the experiments to reduce the effect of combustion reactions in different experiments. Furthermore, the fluidizing gas velocity is mostly controlled by air  $(N_2 + O_2)$  flowrate; thus, the variances in gas and solids residence times were kept small with the superficial gas velocity ranging from 4 to 7 times the minimum fluidization velocity of the bed. This helps to decrease the unexpected effects of hydrodynamics changes on the experimental results. By a constant O<sub>2</sub>/coal ratio of 0.75, the tested steam/O<sub>2</sub> ratios of 0.5, 0.75, and 1.0 are equal to steam/coal ratios of 0.375, 0.563, and 0.75, respectively.

The experimental results were used to calculate five gasification process parameters and syngas quality indices including gasification efficiency, gas yield, carbon conversion, and  $H_2/CO$  and  $CH_4/H_2$  in syngas, as the response for the experimental design.

The design of experiment (DOE) method is used to design the experiments in such a way to analyze the effect of parameters while using a minimum number of experiments and also to evaluate the interaction between the effective operating parameters. The response surface methodology (RSM) is a technique accompanied by DOE methods used for modeling and analysis of problems where a desired output variable (response) is influenced by several independent variables. The RSM was developed initially by Box and Wilson in 1951 to support the improvement of manufacturing processes in the chemical industry [33].

The first step in the RSM practice is to find the functional relationship between the response variables and the independent variables to generate the response surface for analysis purposes. This response surface can be maximized or minimized to find the optimum experimental conditions for a process even if these optimum conditions are not located in the range of variables experimented. Usually, first- or second-order polynomials are used to estimate this relationship, and the coefficients of the model are found using

#### Table 2

List of designed experiments to study the effects of three operating factors (coal feedrate, coal particle size and steam/O<sub>2</sub> ratio on syngas quality.

Run no.	Coal flow rate (g/s)	Particle size (µm)	Steam/O <sub>2</sub> ratio in feed	Carbon conversion	H <sub>2</sub> /CO ratio in syngas	CH <sub>4</sub> /H <sub>2</sub> ratio in syngas	Gasification eff. (%)	Gas yield (m <sup>3</sup> /kg coal)
1	0.0495	285	0.75	95.48	1.006	0.0587	62.12	3.57
2	0.0495	285	0.75	96	1.011	0.0574	62.24	3.61
3	0.0495	285	0.75	96	0.987	0.0582	60.79	3.6
4	0.036	70	1	93.61	1.23	0.0519	61.17	3.54
5	0.036	285	0.75	95.68	1.013	0.0574	61.37	3.59
6	0.0495	285	1	95.41	1.215	0.0528	59.5	3.63
7	0.0495	285	0.75	96	0.975	0.057	66.88	3.65
8	0.0495	285	0.5	93.79	0.81	0.0654	63.93	3.5
9	0.063	70	1	93.35	1.249	0.0517	57.92	3.57
10	0.063	285	0.75	96.59	1.037	0.0579	62.24	3.63
11	0.063	500	0.5	91.1	0.812	0.0674	56.23	3.44
12	0.063	500	1	94.33	1.268	0.0537	58.98	3.6
13	0.063	70	0.5	91.57	0.81	0.0646	56.5	3.45
14	0.0495	500	0.75	93.83	1.017	0.0611	58.84	3.56
15	0.036	500	0.5	93.58	0.776	0.0702	59.42	3.47
16	0.0495	285	0.75	96.89	1.003	0.0587	64.07	3.66
17	0.0495	285	0.75	96.42	1.01	0.0586	63.66	3.65
18	0.0495	70	0.75	92.56	1.048	0.0573	59.03	3.47
19	0.036	500	1	96.41	1.186	0.0553	62.94	3.63
20	0.036	70	0.5	92.26	0.816	0.0662	59.83	3.42
Valida	tion experiments							
V1	0.063	500	0.75	92.81	1.065	0.057	57.66	3.52
V2	0.036	500	0.75	96.49	1.013	0.059	62.73	3.59
V3	0.063	70	0.75	94.45	1.009	0.058	59.96	3.55
V4	0.036	70	0.75	95.85	1.003	0.06	62	3.58

S. Karimipour et al. / Fuel xxx (2012) xxx-xxx

least-squares fit with the experimental data. Because the interactions between variables are important for this study, the central composite design (CCD) method of experimental design, which is the most common design to fit second-order polynomials, is used here to be able to predict the non-linear interactions between parameters. In this method, three types of experimental runs including factorial runs  $(2^k)$ , axial runs (2k), and center runs  $(n_c)$ should be performed, where *k* is the number of variables [34,35]. Two factors in the design are the number of replication of the center point and the distance of the axial runs from the center ( $\alpha$ ). In the face-centered CCD design,  $\alpha$  is equal to 1 and locates the axial points on the centers of the faces of a cube (located at  $(\pm 1, 0, 0)$ ,  $(0,\pm 1,0)$  and  $(0,0,\pm 1)$ ). A value of  $n_c = 2$  is often sufficient to give a good variance across the experimental range, but more can be used to increase the accuracy of the results [35]. Six replications of the central run, suggested by the Design Expert<sup>®</sup> software, are performed at the midpoints of all the operating ranges to estimate the residual error. Considering three effective parameters and six replications of the center points, the number of experiments required for this study can be calculated as:

$$N = 2^{k} + 2k + n_{c} = 2^{3} + 2 \times 3 + 6 = 20 \tag{1}$$

The list of experimental points calculated for this study using the Design Expert<sup>®</sup> and their corresponding response parameters are shown in Table 2. Each response is used to develop an empirical model that correlates the response to the three operating variables using a second-order polynomial given by:

$$Y = b_0 + \sum_{i=1}^k b_i x_i + \sum_{i=1}^k b_{ii} x_{ii}^2 + \sum_{i< j}^k \sum_{i=1}^k b_{ij} x_i x_j$$
(2)

where *Y* is the predicted response,  $b_0$  the intercept,  $b_i$  the linear coefficients,  $b_{ij}$  the interaction coefficients,  $b_{ii}$  the quadratic coefficients, and  $x_i, x_j$  are the normalized values of the response variables.

Statistical tests are performed to evaluate the precision of the empirical second-order polynomial correlation. Although these correlations are only valid for the range of operating conditions and the experimental setup tested here, they are useful for studying the relative influence of the effective variables and making rough predictions of the systems performance. Because the operating variables have different scales, the variables are normalized to the interval [-1, 1] before the polynomial regression is applied. The normalization is performed according to the following equation:

$$X_{norm,i} = \frac{X_{orig,i} - (X_{orig,max} + X_{orig,min})/2}{(X_{orig,max} - X_{orig,min})/2}$$
(3)

where  $X_{orig}$  and  $X_{norm}$  are the original and normalized versions of the operating variable. Different terms of the model found with normalized variables would be on similar scales and comparable. It is worth mentioning that these correlations are not recommended for use as pure predictors; more elaborate models are needed for this purpose. In the present work, they are used for evaluating the effective variables and their interactions and comparing different cases as well as finding the optimized conditions only for the system under study.

#### 4. Results and discussion

The results of all 20 experiments are provided in Table 2. In this table, the gasification efficiency (GE) is defined as the ratio of syngas heat value to the coal heat value and is calculated as:

$$GE = \frac{HHV_s \times Q_s}{HHV_c \times m_c} \times 100$$
(4)

where  $HHV_s$  is the syngas higher heating value,  $Q_s$  is the syngas volumetric flowrate calculated from N<sub>2</sub> balance assuming 60% of fuel-N

is converted to NH<sub>3</sub> [36], *HHV<sub>c</sub>* is the coal higher heating value, and  $m_c$  is the coal mass feedrate to the gasifier. The *HHV<sub>s</sub>* is calculated as:

$$HHV_s = \sum x_i HHV_i \tag{5}$$

where  $x_i$  is the fraction of each combustible gases in syngas such as CH<sub>4</sub>, CO, C<sub>2</sub>H<sub>4</sub>, C<sub>2</sub>H<sub>2</sub>, and *HHV<sub>i</sub>* is its corresponding heat of combustion.

The gas yield (GY) is defined as the volume of syngas produced per unit mass of coal consumed in gasifier in dry basis (water excluded):

$$GY = \frac{Q_s}{m_c}$$
(6)

The carbon conversion  $(X_c)$  determines the fraction of carbon from coal converted to carbon in syngas constituents:

$$X_{\rm C} = \frac{12 \times M_{\rm CS}}{U_{\rm c} \times m_{\rm c}} \tag{7}$$

where  $M_{CS}$  is the total mole flow of carbon in all carbon-bearing components in syngas,  $U_c$  is the fraction of carbon in coal from ultimate analysis, and  $m_c$  is the coal mass flow to the gasifier.

From the experimental results of Table 2, in the range of the operating variables tested here, the GE, GY,  $X_C$ ,  $H_2/CO$ , and  $CH_4/CO$  in syngas range from 56 to 67%, 3.4 to 3.7 m<sup>3</sup> gas/kg coal, 91 to 97%, 0.776 to 1.268, and 0.0517 to 0.0702, respectively. The syngas HHV was calculated to be between 3.77 and 4.21 MJ/m<sup>3</sup>. These results are in the range of data reported by other researchers for gasification of different rank coals [11–18]. The  $H_2/CO$  and  $CH_4/CO$  values have been directly calculated from experimental data, but the coal ultimate analysis has been used to calculate other parameters. Because ultimate analysis is calculated for samples of the coal, it might not be exactly the same for the coal used in the reactions. Thus,  $H_2/CO$  and  $CH_4/CO$  values are more reliable than the others.

By introducing the coal particles into the gasifier, both pyrolysis (de-volatilization that leads to the release of the volatile matters) and gasification reactions begin with increasing the coal temperature. The volatile matters contain a high fraction of water, but fractions of other gases such as CO2, CO, H2, and CH4 are also noticeable [37]. Besides the de-volatilization reaction, CO is generated by heterogeneous CO<sub>2</sub> and H<sub>2</sub>O gasification. The steam gasification produces the same number of moles of CO and H2. Under the conditions of the experiments, steam gasification is expected to be about 100 times faster than CO<sub>2</sub> gasification [38]. CH4 is generated mainly during coal pyrolysis in the heating phase [39,40]. Production of CH<sub>4</sub> by H<sub>2</sub> gasification (hydrogasification) is considered to be significant only at high temperatures and pressures [41]. The water/gas shift reaction  $(CO + H_2O \rightarrow CO_2 + H_2)$  governs the  $H_2/CO$  ratio in syngas. At high steam partial pressures, the reaction favors CO consumption and H<sub>2</sub> production.

#### 4.1. Statistical analysis of the experimental results

The ANOVA analysis is used to study the effect of operating parameters and their interactions on the gasification process and syngas quality. The ANOVA method has been developed to interpret the results of the systems where several factors are effective and can vary simultaneously. In ANOVA analysis, information from all the experiments is used in the analysis of the results, making ANOVA a more powerful tool than varying only one factor at a time. The effects of experimental errors are determined by repetition of the experiment that resides at the center of design points. The results of ANOVA analysis are provided in Tables 3 and 4.

6

Statistically, three tests are required to evaluate the reliability of the model: the test of significance of terms, the *R*-squared test, and the lack-of-fit test. The test of significance determines which variables must be included in or excluded from the model. Table 3 shows the result of the test of significance of factors and interactions for five responses. The *p*-value determines the probability of a case that the coefficient for a specific term is zero (i.e., that term has no significant effect). For example, if p-value has a value of 0.01 then there is 1% chance that the regression coefficient for that term is zero. The *p*-value for the whole model determines the significance of the whole model. When *p*-value is small (usually less than 0.05), the individual terms in the model have a significant effect on the response, and when *p*-value is larger than 0.1, the term is insignificant. In Table 3, the *p*-values have only been shown for terms with *p*-values of smaller than 0.1. Removing the insignificant terms can simplify the correlation usually without decreasing its accuracy, as represented by its *R*-squared value. As Table 3 shows, according to the test of significance, the effective variables and terms are not similar for different responses.

The *R*-squared test is a statistical measure of how well a model approximates the data. *R*-squared values range from 0 to 1, where 1 represents the ideal model. The *R*-squared values for different correlations have also been provided in Table 3. Because *R*-squared always increases by adding new terms, large models even with weak predictability always have better *R*-squared values. Thus, the adjusted *R*-squared is calculated to compensate for this drawback. As can bee seen, the *R*-squared values are close to 1 for all correlations tested, indicating that the models are of good quality. As Table 3 indicates, the regression model for GE produces a poor *R*-squared value of 0.59. Thus, the results for GE discussed based on this model are expected to be less accurate compared to other syngas quality parameters.

The lack-of-fit test is used to determine whether discrepancies between measured and expected values can be attributed to random or systematic error. The lack-of-fit test compares the residual error to the pure error based on the replicated design points. If the *p*-value for lack-of-fit is less than 0.05, there is a statistically significant lack-of-fit at the 95% confidence level. According to Table 3, the *p*-values for all the models are more than 0.05. Table 4 provides the correlations derived for all five responses as a function of normalized and actual variables after discarding the insignificant terms. In order to test the models, four additional experiments were conducted in conditions different from design points. The predictions of the models without the insignificant terms for all the design data plus the validation data points are provided in Fig. 2. As can be seen, there is good agreement between actual and predicted data. This is in conformity with the ANOVA analysis of Table 3. The model is able to predict the validation data points with an error of less than 5%.

### 4.2. Effects of operating variables on syngas quality

Table 4 demonstrates the functionality of different parameters to the three operating variables and their interactions based on the ANOVA analysis. Figs. 3–7 each contain three types of plots for various parameters. The perturbation plots facilitate the comparison of the effect of all factors in a single plot at a particular point in the design space; this is more convenient than the common one-factor-at-a-time approach [42]. A steep slope or curvature for a factor indicates that the response is sensitive to that factor, whereas a relatively flat line shows insensitivity to change in that factor. In order to plot the perturbation lines in a single graph, the normalized variables are used.

The interaction plots reveal the presence of interaction between experimental variables. Mutual interaction plots are generated by fixing the third variable at its middle value. The nonparallel and crossed lines confirm the presence of various levels of interaction between those variables, indicating that the effect of one variable is not uniform across the range of other variable and depends on the level of the other variable. The mutual interactions are plotted at the middle point of the third variable. If the value of the third variable is found to be effective on the trend of interactions, the interaction is plotted for different levels of that variable. Only effective interactions are plotted and discussed. The illustrated 3D versions of the interaction between different variables have also been supplied.

#### 4.2.1. Effect of operating variables on carbon conversion $(X_c)$

Fig. 3a–e illustrates the effect of different variables and their interactions on the carbon conversion. The two signs at the end of the lines in the interaction plots only dilineate the end points of the experimented range for each line and do not show the points underlying the plot. The perturbation plot shows the effect of each variable when the other variables are at the middle of their variation range. As can be seen in Fig. 3a and supported by *p*-value data of less than 0.1 in Table 3, all three variables are effective on the carbon conversion. The carbon conversion seems to decrease with increasing the coal feedrate and then levels off. Although the oxygen/coal and steam/O<sub>2</sub> is kept constant, the mixing of gas/particles may not increase with the same rate by increasing the solid fraction in the system. Thus, the observed decrease of carbon conversion can be attributed to the slight decrease of the mixing. The

Table 3

Ana	lysis of	f variance	(ANOVA)	for the	e models	generated	based	l on t	he exper	imental	data.
-----	----------	------------	---------	---------	----------	-----------	-------	--------	----------	---------	-------

Source	Carbon conversion		H <sub>2</sub> /CO ratio	H <sub>2</sub> /CO ratio		CH <sub>4</sub> /H <sub>2</sub> ratio			Gasification Efficiency	
	SS	p-value	SS	p-value	SS	p-value	SS	p-value	SS	p-value
Model	53.59	< 0.0001	0.46	< 0.0001	5.14E-04	<0.0001	9.80E-02	< 0.0001	8.16E+01	0.0022
A-Coal flowrate	2.12	0.0627	2.35E-03	0.0089	3.13E-06	0.0304		0.0201	1.66E+01	0.0474
B-Particle size	3.49	0.0215	8.85E-04	0.0836	2.57E-05	< 0.0001	6.38E-03	< 0.0001	3.90E-01	0.7457
C-Steam/O <sub>2</sub>	1.17E+01	0.0004	4.50E-01	< 0.0001	4.67E-04	< 0.0001	4.70E-02			
AB	1.65E+00	0.0964	1.38E-03	0.0357						
AC										
BC										
A2										
B2	1.45E+01	0.0001	1.06E-03	0.0604	4.15E-06	0.015	4.40E-02	< 0.0001	6.46E+01	0.0006
C2	1.71E+00	0.0912			3.23E-06	0.0283				
Residual	6.66E+00		3.57E-03		7.56E-06		1.50E-02		5.74E+01	
Lack of fit	5.53E+00		2.50E-03	0.4051	4.82E-06	0.5423	8.17E-03	0.8307	3.51E+01	0.7025
Pure error	1.13E+00		1.07E-03		2.74E-06		7.16E-03		2.23E+01	
R-Squared (R2)	0.8894		0.9923		0.9855		0.8645		0.5869	
Adjusted R2	0.8384		0.9895		0.9803		0.8391		0.5095	
Predicted R2	0.7013		0.9836		0.9709		0.8053		0.3938	

### S. Karimipour et al./Fuel xxx (2012) xxx-xxx

### Table 4

The empirical correlations developed for various syngas quality index parameters based on the experimental data as a function of normalized and actual operating factors.

Index	Туре	Correlation
Xc	Norm.	$X_{C} = 96.0 - 0.46 \times CF + 5.9 \times PS + 1.08 \times C - 0.45 \times CF \times PS - 2.13 \times PS^{2} - 0.73 \times S/O^{2}$
	Actual	$X_{C} = 81.12 + 10.4 \times CF + 0.0367 \times PS + 21.84 \times S/O - 0.156 \times CF \times PS - 4.61E - 5 \times PS^{2} - 11.68 \times S/O^{2} \times PS - 10.0367 \times PS + 21.84 \times S/O^{2} \times S^{2} - 10.0367 \times PS + 21.84 \times S^{2} - 10.0367 \times S^{2} - $
$H_2/CO$	Norm.	$H_2/CO = 1.01 + 0.015 \times CF - 9.41E - 03 \times PS, + 0.21 \times S/O + 0.013 \times CF \times PS + 0.015 \times PS^2$
	Actual	$H_2/CO = 0.415 - 0.152 \times CF - 4.47E - 4 \times PS + 0.85 \times S/O + 4.52E - 3 \times CF \times PS + 3.16E - 7 \times PS^2 \times PS + 1.16E - 1.16E + 1.$
$CH_4/H_2$	Norm.	$CH_4/H_2 = 0.058 - 5.59E - 4 \times CF + 1.6E - 3 \times PS - 6.83E - 3 \times S/O + 1.14E - 3 \times PS^2 + 1.0E - 3 \times S/O^2 + 1.0E $
	Actual	$CH_4/H_2 = 0.089 - 0.041 \times CF - 6.58E - 6 \times PS - 0.051 \times S/O + 2.46E - 8 \times PS^2 + 0.016 \times S/O^2 \times S/$
GY	Norm.	$GY = 3.61 + 0.025 \times \textit{PS} + 0.069 \times \textit{S}/\textit{O} - 0.094 \times \textit{PS}^2$
	Actual	$GY = 3.2 + 1.28E + 3 \times PS + 0.274 \times S/O - 2.04E - 6 \times PS^2$
GE	Norm.	$GY = 62.68 - 1.29 \times CF + 0.2 \times PS - 3.6 \times PS^2$
	Actual	$GY = 60.82 - 95.3 \times CF + 0.045 \times PS - 7.78E - 5 \times PS^2$



Fig. 2. Predicted versus experimental data, the filled points are experimental data used for models regression and the hollow points are validation experimental data.

S. Karimipour et al./Fuel xxx (2012) xxx-xxx

opposite trend can be seen for the steam/ $O_2$  ratio as the carbon conversion increases and then levels off with increasing steam/ $O_2$  ratio. For the three levels of steam/ $O_2$  ratio of 0.5, 0.75, and 1.0, the carbon conversion for 0.5 was less than 0.75 and 1.0 and was almost the same for 0.75 and 1.0. This can be attributed to the inhibitive effect of  $H_2$  dissociative chemisorption on steam gasification in reducing the rate of gasification and preventing the progress of steam gasification of the char beyond a level of conversion, as reported by many researchers [43–47]. The graph presents a maximum at middle sizes with increasing the coal particle size. As reported by Hanson et al. [30], ash content decreases with

decreasing the particle size. The volatile matter may also de-volatilize in the grinding process and thus be less present in small coal particles [29]. This can have a direct influence on the catalytic effect of ash in the gasification reactions and decrease of coal reactivity by reducing ash content and volatile matter. On the other hand, smaller particles offer more reacting surface. The interaction of these two competing effects might explain the trend of variation of carbon conversion with particle size. Initially, increased surface enhances the conversion, but gradually other variables associated with size reduction dominate and decrease the conversion.



**Fig. 3.** The effect of different factors and their interactions on the carbon conversion, (a) perturbation plot, (b) 3D interaction plot between coal feedrate and particle size at steam/O<sub>2</sub> ratio of 0.75, (c)–(e) mutual interaction between coal feedrate and particle size at steam/O<sub>2</sub> ratio of (c) 0.5, (d) 0.75, (e) 1.0.

S. Karimipour et al./Fuel xxx (2012) xxx-xxx



**Fig. 4.** The effect of different factors and their interactions on the  $H_2/CO$  ratio in syngas, (a) perturbation plot, (b) 3D interaction plot between coal feedrate and particle size at steam/O<sub>2</sub> ratio of 0.75, (c) Mutual interaction between coal feedrate and particle size at steam/O<sub>2</sub> ratio of 0.75.

The interaction plots between different variables and Table 4 results indicate that only the interaction of coal feedrate and particle size is effective on carbon conversion, as shown by the non-parallel lines in Fig. 3c-e. The figures show that the level of interactions differs for different levels of steam/O<sub>2</sub> ratio. For high values of steam/ O<sub>2</sub> ratio, large coal particle sizes are favored for higher carbon conversions. However, the two lines cross for a steam/O<sub>2</sub> ratio of 0.5 at a coal feedrate of around 0.056 g/s. Thus, at low steam/ $O_2$  ratios and high coal feedrates, small coal particles are preferred. When particles are larger, their residence time is longer, but the effective surface area is lower. A higher steam/O<sub>2</sub> ratio helps to decrease the problem of lower effective surface area for larger particles by decreasing the diffusion resistance. The increased steam/O<sub>2</sub> ratio along with higher residence time can counteract lower effective surface area in case of large particles, thus providing better conversion for larger particles at high steam/O2 as compared to small particles. Generally, for higher steam/O<sub>2</sub> ratios, the increase in conversion with increase in the particle size suggests that the effect of increase in residence time counterbalances the decrease in effective surface area. However, for lower steam/O2 ratio, a decrease in conversion with increase in particle size suggests that the effect of decrease in effective surface area is more dominant compared to the effect of increase in residence time. Fig. 3c-e indicates that low coal feedrates, particle sizes, and high steam/O<sub>2</sub> ratios are favorable to obtain high rates of carbon conversion.

# 4.2.2. Effect of operating variables on H<sub>2</sub>/CO ratio in syngas

Fig. 4a–c illustrates the effect of the three variables and their interactions on the  $H_2/CO$  ratio. The relative importance of these

variables can be seen from Table 3 and 4. As can be seen in Fig. 4a, the steam/ $O_2$  ratio has the greatest effect on the  $H_2/CO$  ratio. An increase of steam/O<sub>2</sub> ratio from 0.5 to 0.75 leads to a 25% increase in the H<sub>2</sub>/CO ratio. The other two variables seem to have minor effects on the  $H_2/CO$  ratio. The  $H_2/CO$  ratio slightly increases with increasing the coal feedrate and decreases with increasing the particle size. As mentioned before, the water/gas shift reaction governs the H<sub>2</sub>/CO ratio in syngas. By increasing the steam partial pressure, the reaction favors CO consumption and H<sub>2</sub> production. In fact, the extra steam available has also been claimed to alleviate the bed temperature, preventing the formation of carbon monoxide and thus increasing the H<sub>2</sub>/CO ratio [20]. Fig. 4b magnifies the effect of coal feedrate and particle size on various parameters at the middle value of the steam/O<sub>2</sub> ratio. The trend of the plots is similar across the range of steam/O<sub>2</sub> ratios. As can be seen in Fig. 3b, the H<sub>2</sub>/CO ratio slightly increases with increasing the coal feedrate. Because the  $H_2/CO$  ratio in volatile matter is fixed, the increase of  $H_2/$ CO ratio should come from the reactions in the gasifier and not from any extra volatile matter released. Because the supply of  $O_2$  to the system was fixed by an  $O_2$ /coal ratio of 0.75, more coal means more CO<sub>2</sub> and CO production. The produced CO triggers more H<sub>2</sub> production and CO consumption with the water/gas shift reaction [20].

Fig. 4a–c. The effect of different factors and their interactions on the  $H_2/CO$  ratio in syngas, (a) perturbation plot, (b) 3D interaction plot between coal feedrate and particle size at steam/O<sub>2</sub> ratio of 0.75, (c) Mutual interaction between coal feedrate and particle size at steam/O<sub>2</sub> ratio of 0.75.

Fig. 4b reveals that the increase of  $H_2/CO$  ratio by increasing the coal feedrate is noticeable mostly at higher coal particle sizes. This

S. Karimipour et al. / Fuel xxx (2012) xxx-xxx



**Fig. 5.** The effect of different factors and their interactions on the CH<sub>4</sub>/H<sub>2</sub> ratio in syngas, (a) perturbation plot, (b) 3D interaction plot between coal feedrate and particle size at steam/O<sub>2</sub> ratio of 0.75, (c) Mutual interaction between coal feedrate and particle size at steam/O<sub>2</sub> ratio of 0.75.



Fig. 6. The effect of different factors and their interactions on the gas yield (GY), (a) perturbation plot, (b) 3D interaction plot between coal feedrate and particle size at steam/ O<sub>2</sub> ratio of 0.75.

can also be seen by the presence of the positive effect of the squared particle size term in the fitted model. This can be attributed to the relatively higher carbon conversion ( $X_C$ ) for larger particles as previously discussed. The H<sub>2</sub>/CO ratio monotonically

decreases with increasing particle size at low coal feedrates but presents a minimum at high coal feedrates. This minimum corresponds to the crossover point of coal particle size effect on carbon conversion at high coal feedrates, as discussed before. Similar to

S. Karimipour et al./Fuel xxx (2012) xxx-xxx



**Fig. 7.** The effect of different factors and their interactions on the gas efficiency (GE), (a) perturbation plot, (b) 3D interaction plot between coal feedrate and particle size at steam/O<sub>2</sub> ratio of 0.75.

carbon conversion, the only noticeable interaction is between coal feedrate and particle size, as shown in Fig. 4c. Contrary to the carbon conversion, the trend was found to be independent of the steam/O<sub>2</sub> ratio. Processing the results reveals that for a high  $H_2/$  CO ratio having high steam/O<sub>2</sub> ratio is the major requirement; higher coal feedrate and larger particle sizes are also marginally preferential.

#### 4.2.3. Effect of operating variables on $CH_4/H_2$ ratio in syngas

Fig. 5a-b exhibits the effect of the three studied operating conditions on the syngas quality parameters. The perturbation plot of Fig. 5a shows that the steam/ $O_2$  ratio presents the major influence on the CH<sub>4</sub>/H<sub>2</sub> ratio and particle size is next. As Fig. 5a shows, the CH<sub>4</sub>/H<sub>2</sub> ratio dramatically decreases by increasing the steam/O<sub>2</sub> ratio across its whole tested range. The CH<sub>4</sub>/H<sub>2</sub> ratio seems to be relatively less dependent on the coal feedrate. A minor sensitivity appears at high coal feedrates. The coal particle size effect is essentially negligible for below-average sizes (285 µm), but the effect increases for more than average coal sizes. The major source of CH<sub>4</sub> is the volatile matter. The hydrogasification reaction  $(2H_2 + C \rightarrow CH_4)$ also produces methane, but its contribution is usually minor, especially at low pressures, compared to pyrolysis [39]. In contrast, the steam gasification reaction, which is the major source of H<sub>2</sub>, is significant at all pressures and is highly affected by the steam concentration. Thus, the increase in H<sub>2</sub> production caused by increasing the steam/O<sub>2</sub> ratio is much more than the increase of CH<sub>4</sub> production with higher  $H_2$  concentration. This leads to a decrease in the  $CH_4/H_2$  ratio as the steam/O<sub>2</sub> ratio increases.

Fig. 5b magnifies the effect of coal feedrate and particle size on  $CH_4/H_2$  ratio at steam/O<sub>2</sub> of 0.75. As can be seen, the  $CH_4/H_2$  ratio slightly increases with increasing coal particle size. This can be attributed to the higher volatile matter content in larger particles [29].

The  $CH_4/H_2$  ratio seems to be insensitive to the coal feedrate at small coal sizes and slightly decreases with increasing the coal feedrate for larger particles. Higher coal feedrates mean more volatile matter, which is believed to be the major source of  $CH_4$ . However, these higher feedrates were also found to increase  $H_2$  production. The decrease of  $CH_4/H_2$  ratio with increasing coal feedrates shows that the rate of  $H_2$  production is greater than that of  $CH_4$  in this situation. Fig. 5c shows no significant interaction between variables but reveals the absolute preference of large coal particles to get higher  $CH_4/H_2$  ratios in syngas.

### 4.2.4. Effect of operating variables on gas yield (GY)

Fig. 6a–b shows the effect of variables on GY. This figure and Table 3 indicate that only particle size and steam/ $O_2$  are comparatively effective. The steam/ $O_2$  ratio is the most effective variable. Because GY is calculated on a dry basis, the direct increase of water due to increasing the steam/ $O_2$  ratio has been excluded in GY. However, steam participates in steam and hydrogasification and water/gas shift reactions that produce more gases in the

#### Table 5

The results of the optimization of operating conditions to achieve desirable syngas quality for different applications.

Target var./opt no.	1	2	3	4	5	6	7	8	9	10
Carbon conversion H <sub>2</sub> /CO ratio CH <sub>4</sub> /H <sub>2</sub> ratio	Max	Max	Max			Max Max Max		Max Max	Max	Max
Gas yield Gasification efficiency				Max	Max	Max Max	Max Max		Max	Max Max
<i>Operating conditions</i> Coal flowrate (g/s) Particle size (μm) Steam/O <sub>2</sub> ratio	0.036 367 0.97	0.063 500 1	0.036 500 0.5	0.063 380 1	0.0495 290 0.5	0.036 434 0.86	0.063 288 1	0.0495 500 0.77	0.063 376 1	0.063 290 1
<i>Target values</i> Carbon conversion H <sub>2</sub> /CO ratio CH <sub>4</sub> /H <sub>2</sub> ratio	97.2	1.263	0.0699			97 1.074 0.0573		1.041 0.06	1.249	1.244
Gas yield Gasification efficiency Desirability	1	0.991	0.986	3.66 1	65 0.824	3.63 62 0.687	3.66.8 63 0.843	0.491	3.66 0.980	3.66 63.8 0.878

calculation of GY. Increasing the coal particle size first increases and then decreases GY, with somewhat higher GY at larger particle sizes. The same argument presented for the trend of carbon conversion with particle size can be applied for the particle size effect on GY. The presence of similar trends for GY and carbon conversion has also been reported by others [20]. The GY does not show noticeable sensitivity to coal feedrate except for a slight increase at the highest feedrates. No strong mutual interaction between operating variables was observed regarding GY of the produced syngas.

#### 4.2.5. Effect of operating variables on gasification efficiency (GE)

Fig. 7a-b illustrates the effects of operating variables on gasification efficiency (GE). The GE is calculated based on the heating value and concentration of combustible gases in the syngas. In many of the previous works, the effect of operating conditions on GE could not be accurately studied due to having a variable O<sub>2</sub>/coal ratio during the experiments [15,19]. Because  $O_2$ /coal is constant during the current experiments conducted here, the influence of consumption of carbon and gases by combustion can be assumed to be equal for all of the experiments. A study of the syngas composition reveals that the major contributors to GE are H<sub>2</sub> and CO. Thus, a combination of operating conditions that increases the production of both of these gases, not the H<sub>2</sub>/CO ratio, would increase GE. As Fig 7a shows, only coal feedrate influences the gasification efficiency, where, GE uniformly decreases by increasing the coal feedrate. As discussed earlier, increasing the coal feedrate decreases the carbon conversion. Thus, it negatively affects GE by decreasing the production of combustible gases due to lower conversions. No noticeable mutual interaction between variables was detected regarding the gasification efficiency.

# 4.3. Finding the preferred operating conditions to produce syngas for different applications

The desired quality of the syngas from the gasification process varies according to its application. In order to find the desired quality, a desirability function is defined as the objective function for a numerical optimization problem. The desirability is a function that ranges from zero to one, where zero corresponds to a situation in which all of the targets are far off the limits and one corresponds to a situation in which all of the target parameters are at their maximum. In other words, desirability is a measure of the overall closeness of the combination of targets from their maximum. The numerical optimization finds a point that maximizes the desirability function. Desirability is a relative variable and the value desirability in optimization problems with different targets cannot be compared. The Nelder–Mead technique is used to solve the optimization problem [48].

Three main applications of the gasification-derived syngas are considered to be fuel cell (high H<sub>2</sub>/CO ratio), domestic natural gas (high CH<sub>4</sub>/H<sub>2</sub> ratio), and IGCC (high GY and GE). Different optimization problems can be defined based on these applications, which can come separately or altogether. Table 5 shows ten combinations of targets for optimization of the operating conditions. The operating variables have been changed only in the range of variables tested in the present work where the models are believed to be most valid. As can bee seen in Table 5, the desirability is highest when single targets are defined and lowest when contradictory targets are combined or multiple targets are requested. The results also indicate that low coal feedrates favor carbon conversion and  $CH_4/H_2$  ratio (cases 1 and 3), whereas  $H_2/CO$  and GY prefer high feedrates (cases 2 and 4). Coal particles of above-average size are preferred for all targets. Achieving high rates of carbon conversion,  $H_2/CO$  ratio, and GY need high steam/ $O_2$  ratios, whereas  $CH_4/H_2$ and GE need minimal steam content. Producing multi-purpose

syngas that satisfies all targets is less achievable based on the low value of desirability calculated (case 6). However, the conditions can be adjusted to satisfy a combination of two targets with satisfactory values of overall desirabilities, as can be seen for cases 7 through 9. As can be observed, these findings can be qualitatively extracted by direct evaluation of the experimental data, but quantitative finding of the best conditions for various purposes is greatly facilitated using experimental design tools.

### 5. Conclusions

The design of experiment tool based on response surface methodology (RSM), which is believed to be much more accurate than the common one-factor-at-a-time approach, is used to study the effect of three operating variables, namely, coal feedrate, coal particle size, and steam/O<sub>2</sub> ratio, and their interactions on the quality of syngas produced from fluidized bed gasification of lignite coal. Five indices including carbon conversion,  $H_2/CO$  ratio,  $CH_4/H_2$  ratio, gas yield, and gasification efficiency are used as measures of syngas quality. The effects of individual operating variables and their interactions on each syngas quality index were determined to be as follows:

#### 5.1. Carbon conversion

- The carbon conversion seems to decrease with increasing the coal feedrate and then levels off.
- The carbon conversion increases and then levels off with increasing steam/O<sub>2</sub> ratio.
- The carbon conversion graph presents a maximum at middle points with increasing the particle size.
- A study of interaction plots between different variables and Table 4 results indicates that only the interaction of coal feedrate and particle size is effective on carbon conversion. The level of interactions differs for different levels of steam/O<sub>2</sub> ratio: for high values of steam/O<sub>2</sub> ratio, large coal particle sizes are strictly favored for higher carbon conversions. However, small coal particles are preferred for low steam/O<sub>2</sub> ratios and high coal feedrates.
- In general, low coal feedrates, average particle sizes, and high steam/O<sub>2</sub> ratios are favorable to obtain high rates of carbon conversion.

#### 5.2. H<sub>2</sub>/CO ratio

- $\bullet$  The steam/O2 ratio has the greatest effect on the  $H_2/CO$  ratio, as expected.
- The effect of other two variables is minor. The H<sub>2</sub>/CO ratio slightly increases with increasing the coal feedrate and decreases with increasing the particle size.
- Similar to carbon conversion, the only noticeable interaction is between coal feedrate and particle size. But, contrary to the carbon conversion, the trend was found to be independent of the steam/O<sub>2</sub> ratio.
- In general, a high steam/O<sub>2</sub> ratio is the major requirement for a high H<sub>2</sub>/CO ratio, and higher coal feedrates and larger particle sizes are marginally preferential.

### 5.3. $CH_4/H_2$ ratio

- $\bullet$  The steam/O\_2 ratio presents the major influence on the  $CH_4/H_2$  ratio, and particle size comes next.
- The CH<sub>4</sub>/H<sub>2</sub> ratio dramatically decreases by increasing the steam/O2 ratio across the entire tested range.
- The CH<sub>4</sub>/H<sub>2</sub> ratio is relatively independent of the coal feedrate.

#### S. Karimipour et al./Fuel xxx (2012) xxx-xxx

- The coal particle size effect is negligible for less-than-average particle sizes (285 μm), but the effect increases for aboveaverage sizes.
- No significant interaction between variables was found, but large coal particle sizes are preferable to get higher CH<sub>4</sub>/H<sub>2</sub> ratios in syngas.

#### 5.4. Gas yield (GY)

- All three variables affect GY. The steam/O<sub>2</sub> ratio is the most effective variable.
- The GY does not show noticeable sensitivity to coal feedrate until a slight increase at the highest feedrates of coal.
- No significant interaction between variables was found.

### 5.5. Gasification efficiency (GE)

- The major contributors in GE are  $H_2$  and CO. Thus, a combination of operating conditions that increases the production of both of these gases, not the  $H_2/CO$  ratio, increases GE.
- GE uniformly decreases by increasing the coal feedrate.
- No significant interaction between variables was found.

#### 5.6. Optimized conditions for different purposes

- Low coal feedrates favor carbon conversion and  $CH_4/H_2$  ratio whereas  $H_2/CO$  and GY prefer high feedrates.
- Coal particles of above-average size are preferred for all targets.
- Achieving high rates of carbon conversion, H<sub>2</sub>/CO ratio, and GY need high steam/O<sub>2</sub> ratios whereas CH<sub>4</sub>/H<sub>2</sub> and GE need minimal steam content.
- Producing syngas with such quality as to satisfy different applications is less achievable based on the low value of desirability calculated. Desirability is a measure of distance between the current value of each syngas quality index and its optimum value.
- The conditions can be adjusted to satisfy a combination of two targets with satisfactory values of overall desirabilities.

#### References

- Chui EH, Majeski AJ, Lu DY, Hughes R, Gao H, McCalden DJ, et al. Simulation of entrained flow coal gasification. Energy Procedia 2008;1:503–9.
- [2] Natural Resources Canada. Canada's clean coal technology roadmap. <a href="http://www.cleancoaltrm.gc.ca">http://www.cleancoaltrm.gc.ca</a>.
- [3] Natural Resources Canada. Canada's CO<sub>2</sub> capture & storage technology roadmap. <a href="http://www.co2trm.gc.ca">http://www.co2trm.gc.ca</a>.
- [4] Stiegel GJ, Maxwell RC. Gasification technologies: the path to clean, affordable energy in the 21st century. Fuel Process Technol 2001;71:79–97.
- [5] Basu P. Biomass gasification and pyrolysis: practical design and theory. Burlington, USA: Academic Press; 2010.
- [6] Beenackers AACM. Biomass gasification in moving beds, a review of European technologies. Renewable Energy 1999;16:1180–6.
- [7] Babu P. Thermal gasification of biomass technology development: end of task report for 1992 to 1994. Biomass Bioenergy 1995;9:5–15.
- [8] Keeler CG. Wabash river in its fourth year of commercial operation. San Francisco: EPRI/Gasification Technologies Council Technology Conference; 1999. p. 17–20.
- [9] Hirsch RL, Gallagh JE, Lessard RR, Wesselhoft RD. Catalytic coal gasification: an emerging technology. Science 1982;215:121–7.
- [10] Vleeskens JM, Ross M. Attrition of chars in fluidized bed combustion. Fuel 1989;68:825–8.
- [11] Watkinson AP, Cheng G, Prakash CB. Comparison of coal gasification in fluidised and spouted beds. Can J Chem Eng 1983;61:468–77.
- [12] Kawabata J, Yumiyama M, Tazaki Y, Honma S, Takeda S, Yamaguchi H, et al. Performance of a pressurised two-stage fluidised gasification process for production of low-BTU gas from coal char. Chem Eng Commun 1981;11:335–45.
- [13] Saffer M, Ocampo A, Laguerie C. Gasification of coal in a fluidised bed in the presence of water vapor and oxygen: an experimental study and a first attempt at modeling the reactor. Int Chem Eng 1988;28:46–61.

- [14] Tomeczek J, Kudzia W, Gradon B, Remarczyk L. The influence of geometrical factors and feedstock on gasification in a high temperature fluidised bed. Can J Chem Eng 1987;65:785–90.
- [15] Ocampo A, Arenas E, Chejne F, Espinel J, Londono C, Aguirre J, et al. An experimental study on gasification of Colombian coal in fluidised bed. Fuel 2003;82:161–4.
- [16] Purdy MJ, Felder RM, Ferrell JK. Coal gasification in a pilot scale fluidized bed reactor. 1. Gasification of a devolatilized bituminous coal. Ind Eng Chem Process Des Dev 1981;20:675–82.
- [17] Purdy MJ, Felder RM, Ferrell JK. Coal gasification in a pilot scale fluidized bed reactor. 2. Gasification of a New Mexico subbituminous coal. Ind Eng Chem Process Des Dev 1984;23:207–94.
- [18] Rhinehart RR, Felder RM, Ferrell JK. Coal gasification in a pilot-scale fluidized bed reactor. 3. Gasification of a Texas lignite. Ind Eng Chem Res 1987;26:2048–51.
- [19] Kim YJ, Lee SH, Kim SD. Coal gasification characteristics in a downer reactor. Fuel 2001;80:1915–22.
- [20] Zhou H. Air and steam coal partial gasification in an atmospheric fluidized bed. Energ Fuel 2005;19:1619–23.
- [21] Jones RB, McCourt CB, Morley C, King K. Maceral and rank influences on the morphology of coal char. Fuel 1985;64:1460–7.
- [22] Arnold BJ, Smith DB. Coal blending-meeting specifications. Min Eng 1994;10:1144-9.
- [23] Jayaweera SAA, Moss JH, Thwaites MW. The effect of particle size on the combustion of weardale coal. Thermochim Acta 1989;152:215–25.
- [24] Kök MV, Ozbas E, Karacan O, Hicyilmaz C. Effect of particle size on coal pyrolysis. J Anal Appl Pyrolysis 1998;45:103–10.
- [25] Mathews JP, Hatcher PG, Scaroni AW. Particle size dependence of coal volatile matter: is there a non-maceral-related effect? Fuel 1997;76:359–62.
- [26] Lytle JM, Daniel JL, Tingey GL. Concentration of sulphur and mineral rich components in particle classes during coal comminution. Fuel 1983;62:1299–303.
- [27] Kök MV, Özbas E, Hicyilmaz C, Karacan Ö. Effect of particle size on the thermal and combustion properties of coal. Thermochim Acta 1997;302:125–30.
- [28] Yu D, Xu M, Sui J, Liu X, Yu Y, Cao Q. Effect of coal particle size on the proximate composition and combustion properties. Thermochim Acta 2005;439:103–9.
- [29] Zhang C, Jiang X, Wei L, Wang H. Research on pyrolysis characteristics and kinetics of super fine and conventional pulverized coal. Energ Convers Manage 2007;48:797–802.
- [30] Hanson S, Patrick JW, Walker A. The effect of coal particle size on pyrolysis and steam gasification. Fuel 2002;81:531-7.
- [31] Hower JC, Calder JH. Maceral/microlithotype analysis of the hardgrove grindability of lithotypes from the Phalen coal bed, Cape Breton. Nova Scotia Metall Process 1997;14:49–54.
- [32] Hartgers WA, Sinninghe Damste JS, de Leeuw JW, Ling Y, Dyrkacz GR. Molecular characterization of flash pyrolysates of two carboniferous coals and their constituting maceral fractions. Energ Fuel 1994:8:1055–67.
- [33] Hill WJ, Hunter WG. Review of response surface methodology: a literature survey. Technometrics 1966;8:571–90.
- [34] Lazic ZR. Design of experiments in chemical engineering. 1st ed. Weinheim: Wiley-VCH Verlag GmbH; 2004 [chapter 2].
- [35] Montgomery DC. Design and analysis of experiments. 4th ed. USA: John Wiley & Sons; 1997 [chapter 13].
- [36] Leppdahti J, Koljonen T. Nitrogen evolution from coal, peat and wood during gasification: literature review. Fuel Process Technol 1995;43:1–45.
- [37] Suuberg EM, Peters WA, Howard JB. Product composition and kinetics of lignite pyrolysis. Ind Eng Chem Process Des Dev 1978;17:37–46.
- [38] Ye DP, Agnew JB, Zhang DK. Gasification of a South Australian low-rank coal with carbon dioxide and steam: kinetics and reactivity studies. Fuel 1998;77:1209–19.
- [39] Laurendeau NM. Heterogeneous kinetics of coal char gasification and combustion. Prog Energ Combust 1978;4:221–70.
- [40] Lin S, Wang Y, Suzuki Y. Effect of coal rank on steam gasification of Coal/CaO mixtures. Energ Fuel 2007;21:2763–8.
- [41] Misirlioglu Z, Canel M. Hydrogasification of chars under high pressures. Energ Convers Manage 2007;48:52–8.
- [42] Czitrom V. One-factor-at-a-time versus designed experiments. Am Stat 1999;53:126–31.
- [43] Yang RT, Duan RZ. Kinetics and mechanism of gas-carbon reactions: conformation of etch pits, hydrogen inhibition and anisotropy in reactivity. Carbon 1985;23:325–31.
- [44] Chitsora CT, Mühlen H-J, van Heek KH, Jüntgen H. The influence of pyrolysis conditions on the reactivity of char in H<sub>2</sub>O. Fuel Process Technol 1987;15:17–29.
- [45] Hüttinger KJ. Mechanism of water vapor gasification at high hydrogen levels. Carbon 1988;26:79–87.
- [46] Molina A, Mondragon F. Reactivity of coal gasification with steam and CO<sub>2</sub>. Fuel 1998;77:1831–9.
- [47] Bayarsaikhan B, Sonoyama N, Hosokai S, Shimada T, Hayashi J-I, Li CZ, et al. Inhibition of steam gasification of char by volatiles in a fluidized bed under continuous feeding of a brown coal. Fuel 2006;85:340–9.
- [48] Design-Expert. Version 8.0.5. User's guide. USA: Stat-Ease Inc; 2010