Co-gasification of petroleum coke with lignite coal using fluidized bed gasifier

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A B S T R A C T

Co-gasification of waste material with coal is a potentially effective method to utilize industrial wastes for power/ heat and/or syngas generation and to solve their disposal problems. Canada with its large-scale oil sand industry is a major producer of unconventional oil. Petroleum coke is one of the main waste streams from oil sand refining processes. This study is an investigation of the effects of feed ratio, equivalence ratio and average particle size on the syngas production by means of co-gasification of petroleum coke with lignite coal in a stainless steel pilot scale fluidized bed gasifier with an inside diameter of 7 cm at 800 °C. Central composite design methodology was used for designing the experiments. Runs for 20 wt.% of petroleum coke in the feed at different equivalence ratios and particle sizes showed positive synergetic effects for syngas yield (up to 61.0 mol/kg of feed), $H_2 + CO$ yield (up to 33.2 mol/kg of feed), carbon efficiency (up to 70.2%), and lower heating value (up to 9020 kJ/m^3). Increasing the equivalence ratio improved gasification performance, but gas heating value and H_2/CO decreased. The effect of particle size (70–500 μm) was insignificant. The use of petroleum coke (20 wt.% in the feed) has resulted in an improvement in the lignite coal gasification performance and can be considered as an effective utilization method for petroleum coke.

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1. Introduction

Due to its larger overall carbon conversion efficiency for synthesis gas (syngas) production in comparison with other thermo-chemical process-es, gasification is one of the most attractive and commercially viable methods for production of power, heat, and fuels at medium and large scales. In addition to the traditional use of syngas for power generation through a combined cycle, syngas can be either converted to liquid hydrocarbon fuels by means of Fischer–Tropsch reaction or used as feedstock for other value-added products such as methanol and ammonia.

Saskatchewan contributes 14–17% of the coal mined in Canada [1]. This coal is mostly lignite (low-grade) and has low heating value (~18,600 kJ/kg), low sulfur content (<1 wt.%) and large ash content (~16 wt.%) [2]. Different types of petroleum coke are produced from the bitumen upgrading process [3]. Compared with lignite coal, petroleum coke has greater carbon content, greater heating value (~33,500 kJ/kg), greater sulfur content (>5 wt.%) and lower ash content (<4 wt.%) [4]. Petroleum coke is generally used either in steel, aluminum and cement industries or in petroleum refineries [5]. Increasing processing of heavy crude oil has resulted in more petroleum coke production [6]. Although an undesirable by-product, petroleum coke, owing to its high carbon content, can be suitably utilized by means of appropriate thermochemical methods.

Co-gasification is an attractive method in the field of energy and environment for industrial and agricultural waste utilization. Co-gasification for binary or tertiary systems including coal, biomass and industrial wastes can solve the disposal problem of wastes, decrease CO_2 emission, bring about positive synergetic effects and finally help the sustainable development of natural resources. In the last ten years, several research groups have worked on the co-gasification of petroleum coke with coal. Cousins et al. [7] studied the co-gasification of oil sand coke with sub-bituminous-lignite coal in an entrained-flow gasifier at 1200–1400 °C. The results for 50% blending of oil sand coke with each coal and 75% of coke with sub-bituminous coal showed better carbon conversion and gas efficiency compared with those for the coke mono-gasification run. Fermoso et al. [8] observed positive synergetic effects for a blend of bituminous coal and petroleum coke (range of 40–60 wt.% of petroleum coke) for gasification at 1000 °C, 1.5 MPa pressure in a tubular reactor using O_2 and steam as the gasifying agent. This synergetic effect was observed for product gas yield, carbon conversion, cold gas efficiency and higher heating values. Lee et al. [9] studied co-gasification of a slurry mixture of lignite coal and petroleum coke in a 1 tonne/day entrained-flow gasifier for the temperature range of 1400–1600 °C. They observed ~5 and 3 vol.% increase in CO and H_2 concentrations, respectively, for co-gasification runs compared with a mono-gasification run for petroleum coke. Vejahati et al. [10] studied the effects of gasification operating conditions and blending ratio for a mixture of sub-bituminous coal and fluid coke on the gasification efficiency for the gasification temperature range of 1000–1400 °C and atmospheric pressure in an entrained-flow gasifier. They observed no

synergetic effect for co-gasification. In addition, they showed that hydrogen production at higher blending ratios of fluid coke was more sensitive to gasification temperature rather than steam flow rate.

This study is focused on the application of coal gasification technology for utilization of petroleum coke (PC) as a waste material from oil sand derived fuel process. The main objectives are a) to study the effects of PC to lignite coal (LC) mass ratio in feed, equivalence ratio and particle size on the quality and quantity of syngas produced in a pilot scale FBG and b) to investigate the possible positive synergetic effect for co-gasification of PC with LC in FBG systems. The impact of wide ranges of feed ratio, equivalence ratio, and particle size were investigated which provides comprehensive information on co-gasification of PC with LC. In addition, there are only few studies on the co-gasification of PC with LC. These research works have used high temperatures for co-gasification (1,200–1,600 °C). The current study has tried to investigate the co-gasification of PC with LC at low temperature of 800 °C. At this temperature, synergetic effects have not been reported earlier for this co-gasification. These effects at lower gasification temperature make co-gasification of PC with LC a cost-effective method for PC utilization and syngas production. In this study, a bubbling fluidized bed gasifier (FBG) is used. The main advantages of a fluidized bed system for gasification are the uniform temperature profile, running variable load [11] and their suitability for large production [12].

2. Experimental

2.1. Materials

Lignite coal (LC) and delayed petroleum coke (PC) were provided by SaskPower Shand power station (SK, Canada) and Suncor Energy Inc. (AB, Canada), respectively. PC was first crushed with a jaw crusher while the LC had already been crushed at the Shand power station. Both were then ground in a Black and Decker blade type coffee grinder and sieved to three particle size ranges with Sauter Mean Diameter of 70, 285 and 500 μm. Table 1 presents the results for elemental analysis, proximate analysis, and inductively coupled plasma-mass spectrometry (ICP-MS) results of the materials with average particle size of 285 μm.

2.2. Process

The schematic for the fluidized bed gasifier (FBG) setup including 7 cm (ID) stainless steel gasifier with a height of 50 cm, freeboard with a 15 cm diameter and 1 m height, screw feeder, external cyclone, water cooled condenser, boiler and Masterflex peristaltic pump is shown in Fig. 1. To provide flowability in the feeding system, this study used commercially available LC and PC which were air dried to a moisture content of about 5 and 1%, respectively. However, dry LC and PC mixture feed rate was maintained at 0.0384 g/s for all runs. This

target feed rate as well as the injection time of 77 min are chosen based on the previous work (coal gasification study) performed in this research group with the same equipment [13]. A screw feeder was used to continuously charge feed to the FBG (above the stainless steel distributor) from a pressurized hopper. To consider the effect of particle size and density on the function of screw feeder, for each run, rotation speed of auger was calibrated for the nominal feedrate. The deviation of actual feedrate from the calibrated value was determined by the measurement of the feed added to the hopper before each run as well as remaining in the hopper after each run. The deviation was in the range of −2.7 to 2.6% of the target feedrate of 0.0384 g/s. Total gas flow rate was kept constant at 8.6 standard liter per minute (slpm) including air as the source of oxygen, helium as trace gas (0.4 slpm) and nitrogen as the carrier gas. Steam flow rate for each run was calculated based on the equivalence ratio (ER) used for each experiment by keeping the molar ratio of steam to O_2 (in air) constant at 1.8 for all runs. For each run, the required steam flow rate was adjusted to account for the moisture in the feed. Silica sand with particle density of 2600 kg/m^3 and Sauter Mean Diameter of 250 μm was used as bed material.

Each run was started by heating up the gasifier containing silica sand to the gasification temperature (800 °C) under air and nitrogen flow. Then, at this temperature, continuous feed injection and steam flow were started. Feed and steam injection time for each experiment was 77 min. At the end of each run, the gasifier was cooled down under nitrogen flow. Ash discharging was discontinuous, with it being emptied from the gasifier with the silica sand once cooled. After unloading, a sample mixture consisting of unreacted feed, ash and sand (totally ~20 g) was placed in a muffle furnace for heat treatment at 650 °C under air atmosphere. Amounts of ash and unreacted feed were measured this way. Gas product flow rate was estimated using mass balance for helium as trace gas and nitrogen as carrier gas. The flow rates of these gases into the gasifier were known and their concentrations in the outlet gas were measured by means of a gas chromatograph (GC). Unreacted water and tar produced by process were collected in the condenser. The unreacted water and tar were separated by evaporating the water and their amounts were measured. Gas samples were collected every 10 min from the FBG using Tedlar bags and were analyzed by means of GC.

2.3. Characterization techniques

Moisture and ash contents of LC and PC were determined according to ASTM D 2866-94 (Approved 1999). The volatile matter in the feedstock was determined by the procedure given in ASTM D 3175-07. The feedstock sample (1.0 g) was taken and placed in a muffle furnace maintained at 950 ± 10 °C for 7 min. Then, the crucible was removed from the furnace and placed in the desiccators. The loss of weight was given as the volatile matter in the feedstock. Particle size distribution (PSD) for feedstock was determined based on the laser beam diffraction pattern of particles, by means of a particle size analyzer (Mastersizer S Long Bench, Malvern Instruments Ltd., Malvern, UK) using the dry method. Higher heating values (HHV) of LC and PC were determined in an oxygen bomb calorimeter (Parr 6400, PA, USA) using ~1.0 g of sample. The inductively coupled plasma-mass spectrometry (ICP-MS) analysis of the materials was performed using a Sciex Elan 5000 provided by Perkin Elmer, USA. Tedlar bags were used to collect the gas from the FBG and then injected into the gas chromatograph (Agilent 7890A) equipped with two TCDs (thermal conductivity detector) and one FID (flame ionization detector). The concentrations of hydrogen sulfide and sulfur dioxide in the product gas were measured using a gas chromatograph (Model CP-3800, Varian Inc.) equipped with a pulse flame photometric detector (PFPD).

2.4. Design of experiments

Central composite design (CCD) was used to design the experiments. Three parameters including feed ratio (mass ratio of PC to LC),

Table 1
Elemental analysis, proximate analysis, and ICP results of precursors with average particle size of 285 μm (on dry-basis).

Feedstock	C (wt.%)	H (wt.%)	N (wt.%)	S (wt.%)	O (wt.%)	Ash (wt.%)
Petroleum coke	83.9	3.4	1.5	5.5	1.9	3.8
Lignite coal	57.1	3.4	1.0	0.6	21.6	16.3

Feedstock	Fixed carbon (wt.%)		Volatile matter (wt.%)		Moisture (wt.%)		Ash (wt.%)
Petroleum coke	80.9		14.8 ± 1.3		0.5 ± 0.1		3.8 ± 0.1
Lignite coal [13]	44.4		39.0		1.3		15.3

Feedstock	Li[a]	Na	K	Mg	Ca	Al	Fe	Ti	P
Petroleum coke	7	274	357	803	3088	3693	2104	956	109
Lignite coal	10	1653	967	240	548	9772	2665	1065	162

[a] Elements' concentration unit: ppm.