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Integration of pyrolysis and entrained-bed gasification for the production of chemicals from Victorian brown coal — Process simulation and exergy analysis

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ABSTRACT

This paper introduces a novel process for the integration of pyrolysis and entrained-flow gasification for the as-mined Victorian brown coal containing 65 wt.% moisture. An initial mild pyrolysis of coal is proposed to reduce moisture content, and produce multiple products including char, tar and hydrogen-rich coal gas. Subsequently, the resulting water-free char is subjected to an entrained-flow gasifier, upon the blending of silica additive or high-ash bituminous coal. The resultant syngas passes through a cleaning unit and water gas shift reactor before mixing with the pyrolysis-derived gas to reach a target H_2/CO molar ratio of 2.0 that is essential for the synthesis of a number of chemicals. In this paper, process simulation and exergy analysis were conducted to prove the advantages of the proposed process against the conventional drying–gasification combination. The results show that, the proposed pyrolysis–gasification integration process for Victorian brown coal possesses an exergy efficiency 4.5% higher than the drying–gasification process, and 1.5% higher than the drying case for another lignite containing 25 wt.% moisture. A prior removal of 20% of the inherent moisture can further improve the exergy efficiency by 4% for Victorian brown coal. The addition of 20% of black coal is the optimum ratio to improve the ash slagging propensity, as well as improve exergy efficiency compared to the conventional drying–gasification process.

1. Introduction

With the depletion of high-rank coals and petroleum, the use of low-rank subbituminous coal and lignite has been receiving increased attentions in the energy and mining industries. In Victoria, Australia, brown coal is the single largest source contributing to over 85% of the electricity supply in the State. The Victorian brown coal (VBC) has a number of advantages such as large reserve, low-cost, high reactivity, low ash (less than 3 wt.% on dry basis) [1]. However, the high moisture content up to 70 wt.% (as received) is the largest barrier to restrict its utilisation, which entails high transportation costs, potential safety hazards in transportation and storage, and the low thermal efficiency in combustion of such coals [2,3].

Gasification is the reaction of solid fuels with oxidant, including air, oxygen, steam, carbon dioxide, or a mixture of these gases at a high temperature to yield a gaseous product namely syngas, which is rich in CO/H_2 and is suitable for use either as a source of energy or as a raw material for the synthesis of chemicals, liquid fuels or other gaseous fuels [4,5]. In the practical gasification processes, a broad range of reactor types has been examined, including moving-bed gasifier, fluidised-bed gasifier, and entrained-flow gasifier [4,6]. The temperature for moving bed and fluidised bed gasifier is relatively low, less than 1100 °C, which restricts their use to the coals with high reactivity, and high ash fusion temperature (e.g. above 1100 °C) to avoid slagging. However, the cold gas efficiency from low temperature gasification is relatively low, and the carbon conversion rate is also low [7]. Instead, the entrained-flow gasifier can achieve higher cold gas efficiency, and close to 100% carbon conversion rate. The temperature of entrained-bed gasification is usually up to 1600 °C, and the pressure maximises at 3 MPa with a large throughput [8].

Entrained-flow gasifiers are usually applicable to coals with low ash content for both economic and technical reasons [9]. The favourite ash content range for entrained-flow bed gasifier is 10–40 wt.% [8,10], to ensure the formation of a slag coating layer to protect the refractory gasifier wall and minimise heat loss through the wall. In addition, the ash slagging propensity is critical. In an entrained flow gasifier, coals selected for slagging gasifier should have an ash fusion temperature (AFT) below the operating temperature of the gasifier (1400–1600 °C) [4], to maintain the molten slag which can flow down the gasifier walls and drains out from the gasifier. In this sense, the black coals with high fusion temperatures are not suitable to gasification directly in an entrained flow gasifier [10]. Instead, the flux such as limestone is normally added into a coal to lower its AFT [11]. Moreover, the blending of different coals is another common strategy used for the entrained-flow gasification process. To date, the blending of black coal with

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biomass or petroleum coke in fluidised bed gasifier has been examined [12]. For entrained-flow gasification, the blending of biomass with coal has been tested [13]. However, there is still no study focusing on the blending of black coal, particularly high-ash black coal with low-rank coal char. Their blending is supposedly advantageous to exert a number of benefits. On the one hand, the overall ash content can be adjusted to the required ash content level, since the low-rank coal is generally lean in ash. On the other hand, the abundance of alkaline earth metals (Ca, Mg) and/or iron (Fe) in a brown coal char is good flux to promote the melting of black coal ash. More importantly, the brown coal char generally has a high reactivity compared to black coal. Their blending will thus increase the overall reaction rate, and even alter the syngas composition as well.

This study aims to prove the above-mentioned benefits related to brown coal char – black coal blends, through process simulation and exergy analysis. Process simulation has proven to shed insight into the conceptual design of a new chemical process by determining the most efficient process through a sensitivity analysis [14]. Emun [15] conducted a flow sheeting for integrated gasification combined cycle (IGCC) using Aspen Plus, and optimised the operating conditions to achieve a 45% thermal efficiency. The gasification of different fuels, including bituminous coal, lignite and biomass has been modelled in entrained flow bed and fluidised bed reactor [16–20]. Exergy is defined as the maximum theoretical work obtainable from a system compared with the ambient environment. It is based on both the first and second laws of thermodynamics and assumes that work can be derived from reversible process [21]. It provides insights that elude a purely first-law energy conservation approach by focusing on the quality of energy, rather than its quantity. It is used to assess inefficiencies in the system by considering the magnitudes, locations, and types of exergy losses. The exergy analysis methodology has been applied extensively in the processes of coal-fired steam power plants [22], combustion and gasification processes [23,24], combined cycle power plants, and also CO_2 emission control processes, such as integrated coal gasification combined cycle (IGCC) [25], and chemical looping combustion (CLC) processes [26]. However, there is still scarce research on the use of exergy analysis for brown coal entrained-flow gasification.

The process to be developed by us is the integration of brown coal pyrolysis and entrained bed gasification. Through the initial pyrolysis, the brown coal, bearing an inherent moisture content up to 65 wt.%, is dried to release steam, and devolatilised into dried char, liquid tar and gas that is rich in reducing components including CO and H_2. The resultant moisture-free char and steam, blended with either high-ash black coal or additive are subsequently fed into an entrained-bed gasifier or the downstream water–gas shift unit. In the meanwhile, the tarry oil is expected to be sold or refined into high-value liquid fuels and chemicals, whereas the pyrolysis gas is combined with the gasification-derived syngas to ultimately produce chemicals downstream. Such a process is supposedly efficient, since the “easily gasified” moieties (e.g. volatiles) in a brown coal are firstly converted into gas under mild conditions, whereas only the remaining ‘difficultly gasified” components are subjected to entrained-bed gasifier employing a harsh condition. This is different from the conventional entrained-bed gasification system where the whole coal is subjected to the harsh conditions.

2. Methodology

2.1. Description of the proposed pyrolysis–gasification process

The schematic of the proposed new process is shown in Fig. 1. As-mined VBC is initially pyrolysed at a low temperature (e.g.600 °C) to remove moisture and a portion of volatiles. The resulting char is subsequently fed into an entrained-flow gasifier. The volatiles removed from pyrolysis process partially convert into gas, which is rich in hydrogen and carbon monoxide [27]. The condensable tar derived from volatiles is condensed and separated at 200 °C [28]. The large amount of moisture derived from the pyrolysis step is used for gasification and water gas shift (WGS) to minimise the water consumption for the system. The resulting gas is combined with the gasification-derived syngas, whereas the tar can be sold as a value-added by-product, or further refined into value-added chemicals such as Benzene Toluene Xylene (BTX). The syngas will then be used for downstream chemicals production.

Fig. 2 shows the details of the flowsheet which was built in Aspen Plus. The pyrolysis–gasification integration process (Fig. 2(a)) is composed of an air separation unit (ASU), coal pyrolysis and gas separation unit, gasification unit, WGS unit, and gas cooling and cleaning unit. All these units were rigorously modelled using Aspen Plus v8.4. The traditional treatment for high moisture brown coal is steam drying, which was simulated in this paper (Fig. 2(b)) (namely *drying case*) to compare with the pyrolysis–gasification integration process (namely *pyrolysis case*). A boiler is used to generate super-heated steam to 350 °C for coal drying, dropping to 135 °C after mixing with wet coal in the dryer. The moisture content drops to <10 wt.%, which is suitable for entrained flow gasifier feeding [8]. The simulation target here is fixed at 50 kmol/h CO and 100 kmol/h H_2 corresponding to an H_2/CO molar ratio of 2 that is used for the synthesis of most of the chemicals.

2.2. Simulation approaches

Two typical brown coals, VBC and a Chinese lignite (CHL), were selected for this simulation. The silica based additive and a black coal (BLK) was used to promote the brown coal slagging propensity, and to generate sufficient slag to protect the gasifier refractory wall. The proximate and ultimate analyses of coal samples are shown in Table 1, and their ash compositions and fusion temperatures are tabulated in Table 2. The CHL ash has very low fusion temperatures, as expected. However, the VBC coal ash is very difficult to melt. This is due to the abundance of iron oxide accounting for 34.9% within its ash. Since iron is a good flux additive, the blending of VBC char and the black coal BLK would remarkably enhance the ash slagging propensity. It will be detailed by thermodynamic calculation later.

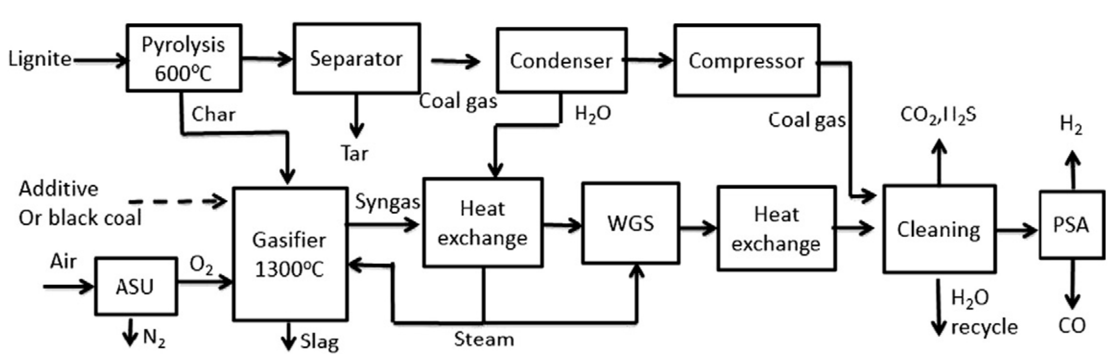

Fig. 1. Schematic of the proposed brown coal pyrolysis–gasification process.