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Energy-efficient recovery of black liquor through gasification and syngas chemical looping *

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HIGHLIGHTS

- Cogeneration system from black liquor is proposed.
- Integrated system covers drying, gasification, syngas chemical looping and power generation.
- High energy efficiency in H₂ and power coproduction can be realized.
- The proposed system can achieve high efficiency about 70%.

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ABSTRACT

One of the strategies to improve environmentally friendly energy harvesting can be realized by using biomass as a primary energy source for generating electricity and H_2 . In addition, high energy efficiency can be achieved by minimizing exergy loss through process integration and exergy recovery. As an implementation, this study proposes a cogeneration system for black liquor (BL) to co-produce electricity and H_2 . The system primarily comprises BL drying, circulating fluidized bed gasification, syngas chemical looping (SCL), and power generation. The Aspen Plus V8.8 software package is used for modeling and performing calculations of the proposed integrated system. Furthermore, thermodynamic analysis of gasification is performed by employing Gibbs energy minimization. The effects of target solid content on the required total work and compressor outlet pressure during drying and gasification-scL processes are significantly cleaner and more energy efficient. The proposed integrated system can achieve a net energy efficiency of about 70% with almost 100% carbon capture.

1. Introduction

Hydrogen (H₂) has been accepted as an important clean energy carrier in the future owing to its characteristics, such as high efficiency, various possible production and utilization technologies, and cleanliness [1]. Therefore, both production and utilization technologies, at large and small scales, are being developed actively. Similar to electricity, as an efficient energy carrier (secondary energy source), H₂ potentially leads to zero carbon emissions at the point of use [2]. Unfortunately, despite its high specific energy content, hydrogen is mostly available on earth in the oxidized state (water). Therefore, H_2 should be generated using a suitable conversion technology, such as gas reforming, oil reforming, coal and biomass gasification, and splitting of water by electrolysis. Compared to pyrolysis, gasification is considered a well-established technology that can give better results in terms of the increases in material decomposition and chemical energy [3].

Among the available types of biomass, including wastes, black liquor (BL) from the pulp mill industry is considered to have very high potential as an energy source. BL conversion through gasification is considered as an alternative technology with a faster reaction rate and

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Nomeno	clature	V L	velocity (m s ⁻¹) length (m)	
MC	moisture content (wt% db)	d	diameter (m)	
Т	temperature (°C)	g	gravitational acceleration (m s ^{-2})	
Р	pressure (kPa)	μ	viscosity (Pas)	
f	fanning friction factor (-)	W	work (MW)	
ρ	density (kg m ^{-3})	η	Efficiency	

excellent conversion efficiency. This approach can replace the inefficient and uneconomic conventional recovery cycle owing to the high moisture content of BL, which ranges from 83 to 87 wt% on wet basis (wb) [4,5]. In terms of electricity production, conventional recovery boilers show very low energy efficiency of about 9–14% [6]. Naqvi et al. [7,8] reported on BL performance gasification by employing a circulating fluidized bed (CFB) with direct causticization for synthetic natural gas production. Gasification with direct causticization can replace the lime kiln process; therefore, significant reduction of the energy required in this stage can be achieved. In addition, they found that O_2 -blown gasification resulted in higher conversion efficiency (cold gas efficiency of 58%) than air-blown gasification. By employing O_2 as the gasifying agent, syngas dilution with nitrogen can be avoided as well. Unfortunately, their study did not focus on the effort involved to sufficiently circulate and recover energy throughout the system.

Ferreira et al. [9] studied the combination of BL gasification and combined cycle (BLGCC) system with and without CO_2 capture. However, they focused only on the exergetic and economic analyses, and did not attempt to develop an innovative system to improve the total energy efficiency. Recently, Darmawan et al. [4] proposed an integrated system of electricity production from BL. Although they achieved relatively high energy efficiency, the adopted entrained flow gasifier was costly and rather difficult to use in terms of operation and material handling owing to its high operating temperature.

Regarding H_2 production from BL, Cao et al. [10] investigated the application of supercritical water gasification (SCWG), pressure swing adsorption (PSA)-based H_2 separation, and steam turbine. Although SCWG is considered energy efficient, a large amount of energy is required to elevate BL to the supercritical condition. In addition, PSA-based H_2 separation is considered an energy-intensive process, leading to low total energy efficiency. Furthermore, Andersson and Harvey [11] studied the conventional BL gasification system for H_2 production, especially from the CO₂ emission viewpoint. Unfortunately, there was no significant effort to improve the energy efficiency of the system.

To the best of the authors' knowledge, there is almost no study clearly dealing with improving the energy efficiency of H_2 and power cogeneration from BL. In this study, an integrated system consisting of drying, CFB gasification, syngas chemical looping (SCL), and power generation is proposed. After primary conversion through gasification, SCL is adopted to effectively cogenerate H_2 and electricity from the produced syngas. Because the process produces pure H_2 and a CO₂ streams in two discrete reactors, the additional CO₂ separation step can be avoided [12]. In addition, to improve energy efficiency throughout the integrated system, process integration and exergy recovery are adopted.

2. Integrated system

2.1. Conventional power and H_2 production system from BL

In a single mill, BL has a large amount of energy of about 250–500 MW [13]. Since BL is available at the boundaries of a pulp mill, it can easily be processed as biomass fuel. To understand BL recovery performance, in this section, we discuss the technologies available for electricity and H_2 production from BL. The first conventional system, illustrated in Fig. 1(a), mainly uses a recovery boiler to produce

steam. This method is commonly adopted in modern pulp mills. First, weak BL with high moisture content (about 85 wt% wb) is made to flow through multiple-effect evaporators (MEE) to increase its solid content (up to 70–75 wt% wb) to make it suitable for combustion. In the combustion stage, heat is recovered, and high-temperature steam is produced for generating power. To increase efficiency, a superheater and economizer are adopted as well. The remaining steam is then sent to the pulp mill for other processes.

Figs. 1(b) and (c) show the BLGCC without and with CO_2 capture, respectively. These BLGCC schemes are based on the data of the business-as-usual scheme in integrated pulp and paper mills [9,14]. The BLGCC system consists mainly of an air separation unit (ASU), gasifier, gas cleaning unit, gas turbine, heat recovery steam generator, steam turbine, condenser, and biomass boiler. Compared to BL recovery through combustion, gasification can significantly increase the system efficiency (> 10%) and shift the pulp mill from electricity importer to





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