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# Evaluation of effect of evaporation on supercritical water gasification of black liquor by energy and exergy analysis

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## ABSTRACT

Supercritical water gasification (SCWG) is a promising innovative black liquor handling method. The low concentration of black liquor is a burden for the system scale and investment cost. In this study, we introduced black liquor evaporator to increase the concentration using self-generated steam and power, and studied its influence on the energy and exergy efficiency. The results showed that increasing black liquor concentration from 10 wt% to 20 wt% reduced the energy efficiency but increased the exergy efficiency of the system without evaporator. With the evaporator, an optimal target concentration existed for the exergy efficiency owing to the influence of the target concentration on the balance between the energy consumption and saving brought by the evaporator. With black liquor condensed from 15 wt% to 21.82 wt%, maximum exergy efficiency of the system (41.95%) was obtained. For lower-concentration black liquor, more water needs to be evaporated to get the optimal concentration and evaporator was more desired because it brought greater improvement on the exergy efficiency. The increasing effect number of multi-effect evaporator shifted the optimal concentration to higher values, which increased from 19.93 wt% to 23.13 wt% when the effect number increased from 4 to 7. The exergy efficiency of the system was also improved, and the improvement was more significant at higher target concentration.

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## Introduction

Black liquor (BL) is the byproduct of paper pulping process, which mainly contains lignin, hemicellulose and their degraded products as well as pulping chemicals. It has high COD content, pH value and irritating odors, which pose a hazardous threat to the environment and human health if not properly treated. Nowadays, black liquor is mainly treated with Tomlinson recovery boiler, where the organics are burned in a boiler for energy recovery. And the inorganics are

recovered after combustion and reused in the pulping process after causticization. Black liquor gasification was also regarded as a potential alternative for black liquor treatment. Great progress has been made on this technology in recent decades and the demonstration plant has been built [1]. However, there are still some drawbacks related to Tomlinson recovery boiler and conventional gasification, including the safety and operating risks brought by the molten alkalis at high temperature, the energy consumption of black liquor evaporation and the emission of NO<sub>x</sub>, SO<sub>x</sub> and fine particles [1–3].

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**Nomenclature**

$Ex_{CH}$	Chemical exergy rate (kW)
$Ex$	Exergy rate (kW)
MP	Medium-pressure steam
LP	Low-pressure steam
BL	Black liquor
SCWG	Supercritical water gasification
COD	Chemical oxygen demand
NG	Natural gas
LHV	Lower heating value (kJ/kg)
HHV	Higher heating value (kJ/kg)
G-L	Gas-liquid
PSA	Pressure Swing Adsorption
SCW	Supercritical water
MEE	Multi-effect evaporator

SCWG is an innovative treatment method of biomass [4,5], coal [6–9] and organic wastes [10]. It takes advantage of the unique properties of water above its critical point (22.1 MPa, 374 °C) to gasify the organic wastes and biomass into hydrogen-rich gases [11–14]. Its application on black liquor treatment can overcome the above drawbacks. The unique reaction environment provided several advantages for black liquor treatment: (1) The high solubility of organics and gases makes SCWG a homogeneous reaction, which can reduce the mass transfer resistance and accelerate the reaction rate. (2) High-efficiency gasification can be realized at lower temperature, which can avoid the smelting of the alkali salts in black liquor and the related safety problems. (3) The alkali in black liquor is an effective catalyst for biomass gasification, which can improve the gasification efficiency and water-gas shift reaction [15–18]. Also, the inorganic components, including the alkali can be recovered for its low solubility in SCW [19–21]. (4) The emissions commonly generated in combustion and conventional gasification, including  $NO_x$ ,  $SO_x$  and fine particles are seldom released in SCW reaction environment [22,23]. (5) Wet biomass can be directly gasified in SCW without evaporation, which can save the energy consumption and improve the energy efficiency. Particularly, black liquor always contains high content of water (>80 wt%), so considerable energy consumption for evaporation can be saved. Therefore, SCWG of black liquor attracted much attention around the world and encouraging results were achieved [24–28]. After treatment by SCWG, the COD content was enormously reduced and the pH value was decreased from about 12 to the neutral ranges (6–8). The color of the black liquor was turned to clear after treatment. Meanwhile, the syngas with high fraction of hydrogen (40.26%–61.02%) was produced [24]. De Blasio et al. [26] obtained over 80% hot gas efficiency in gasifying black liquor in the Inconel reactor at 700 °C. Recently, we gasified black liquor (9.5 wt%) in SCW with a batch reactor at high temperature and long reaction times, and the highest carbon gasification efficiency of 94.10% was obtained at 750 °C [25]. These results showed that SCWG was a promising alternative for black liquor treatment. For the

unique reaction conditions, SCWG of black liquor is encouraged to cogenerate heat and power for the pulp mill. In the previous study, we analyzed the system of SCWG of black liquor coupled with pulping process, which revealed its superiority in energy efficiency over the conventional methods [29].

As described above, an important advantage of SCWG of black liquor treatment is that weak black liquor can be directly gasified without evaporation. The concentration of weak black liquor is usually in the range of 10–20 wt% and it needs to be evaporated to the concentration above 70 wt% before combustion or conventional gasification [30,31]. As a result, black liquor evaporation is an energy-intensive process and about 37% of the energy consumed in the conventional pulp mill is used to evaporate the black liquor [32]. Therefore, the absence of black liquor evaporation can improve the energy efficiency significantly. However, employ of black liquor evaporation can reduce the treatment scale and the investment cost. And the energy loss of the system may be also reduced for decreasing the black liquor flow rate. The usage of evaporation with a mild target concentration may reduce the energy requirement to run the system and benefit the energy efficiency. The necessity of the black liquor evaporation and the target concentration can be assessed by balancing the energy consumption in the evaporation and the energy savings for reducing the system scale. Besides, concentration was reported as an important parameter in SCWG, which influenced the gas product composition from both experimental and thermodynamic analysis [10,13,33]. The energy output and input of the system would be affected by the change of gas product composition. As a result, black liquor concentration was supposed to have complex influences on the system performance. Its investigation will provide important information on the design and optimization of the system of SCWG of black liquor. To the best of our knowledge, no literature was reported on the assessment of the black liquor evaporation in SCWG and the influence of the target concentration on the system performance.

In the present study, we evaluated the influence of black liquor evaporation and the target concentration on the energy and exergy efficiency of the system based on the modelling system as we constructed previously [29]. A new integrated system was constructed by introducing evaporator into SCWG system coupled with the energy requirement of the pulp mill. Firstly, the influence of the initial concentration of black liquor on the system performance without evaporator was evaluated. The variation of the overall energy and exergy efficiency as well as the input and output energy of the system with the concentration was calculated. Secondly, the influence of the target BL concentration of the evaporator on the system performance was evaluated. An optimal target concentration was found in the analysis, where the highest exergy efficiency of the system was obtained. Then the influence of the initial concentration on the SCWG system coupled with BL evaporator was evaluated. Its influence on the optimal target concentration and system performance was studied. Finally, the influence of effect number of the multi-effect evaporator on the optimal target concentration and overall exergy efficiency was calculated.

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