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Hydrogen and syngas production from catalytic steam gasification of char derived from ion-exchangeable Na- and Ca-loaded coal

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ABSTRACT

Catalytic steam gasification of char derived from low-rank coal possesses substantial potential as a source of hydrogen energy and syngas feedstocks, and its performances are largely associated with the employed catalysts. Therein, ion-exchangeable Na or Ca species are always regarded as excellent in-situ catalysts in low-rank coal. In this paper, gasification of Na-Char, Ca-Char and a Na/Ca-Char mixture with different partial pressures of steam was performed within a temperature range of 700–900 °C using a micro fluidized bed reaction analyzer. The results indicate that Na and Ca species could accelerate the gas release rate during gasification and even significantly increase H₂ production, in sharp contrast to non-catalytic gasification. Variations in the product gases during Na-Char and Ca-Char gasification were completely different, which associated with the different deactivation pathways and catalytic reaction mechanisms of Na and Ca catalysts. With an increasing gasification temperature, the decreasing trend of H₂ production for Na-Char gasification was mainly due to the loss of Na during gasification. Conversely, the enhancement of Ca activity promoted the H₂ production. The H₂/CO ratio of Ca-Char gasification at 700 °C approximately ranged from 1.0 to 2.0 as a function of the partial pressure of steam, which suggested catalytic gasification can be suitable for hydrogen-rich production and subsequent synthesis reactions. In addition, gasification of Na/Ca-Char mixture produced a higher hydrogen content in the product gases than that of Na-Char or Ca-Char gasification alone, particularly for the 30%Na/70%Ca-Char mixture. It implies that the high H₂ production of 70%Ca30%Na-Char mixture was attributed to the cooperative effects of the Na and Ca species on the catalytic activity. This study provides comprehensive information regarding the effects of ion-exchangeable Na, Ca and a Na/Ca mixture on the hydrogen production and syngas composition during steam gasification, which provides new insight into the utilization of low-rank coal.

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Introduction

Gasification is the main thermochemical method for the utilization of various carbonaceous solid fuels in power generation, hydrogen production and synthesis of various liquid fuels and chemicals [1,2]. In particular, hydrogen is being expected the potential energy carrier in sustainable energy system [3–6]. Comparatively, coal is still the essential raw material for gasification nowadays, at least until renewable energy source is developed on certain large scale in future [7–9]. Therein, low-rank coal is particularly suitable for the gasification, owing to its high gasification reactivity and large reserves over the globe [10,11]. The catalytic effect of alkali and alkaline metallic (AAEM) species inherently contained in low-rank coal is one of the major factors promoting the gasification reactivity [12–15]. Among the potential catalysts, those AAEM species existed in ion-exchangeable mode are specially promising due to their superior catalytic activity [16–18]. Therefore, some of the AAEM species remained in char are acted as excellent catalysts for subsequent char gasification. Ion-exchangeable Na and/or Ca species are always the main portions of these catalysts in low-rank coal [19]. The catalytic behavior of ion-exchangeable Na and Ca species on the gasification is greatly important for the utilization of low rank coal.

For ion-exchangeable Na and/or Ca catalysts, the superior catalytic activity on the gasification reactivity has been documented intensively. Ohtsuka et al. [18] found that the rate of Loy Yang coal with only 0.56 or 0.84 mmol/g Ca loading was 40–60 times that of the original coal in steam gasification. Likewise, Takarada et al. [20] reported that the rate of Na-exchanged coal with 5.2% Na loading in weight could achieve 20–30 times enhancement for brown coal. The ion-exchangeable catalysts potentially contribute to the char reactivity with a low loading during gasification. We have also previously demonstrated that the char reactivity of Na-Char and Ca-Char derived from ion-exchangeable Na and Ca coal was higher than that of H-Char (non-catalytic) by 1–2 orders of magnitude [21]. Apart from the reactivity of char, how the ion-exchangeable Na and Ca species affect the product gases from steam gasification is also essential for the good understanding of low-rank coal gasification. The gaseous products mainly consist of hydrogen and other syngas, i.e., carbon monoxide. To date, many researches have reported the productions of hydrogen and/or syngas from steam gasification catalyzed by various catalysts. Wang et al. [22] found that the hydrogen yield was substantially enhanced in the steam gasification of char with K_2CO_3 catalysis in the temperature range of 700–750 °C. Mudge et al. [23] studied the catalytic steam gasification of wood using K_2CO_3 , and reported that this catalytic gasification could yield methanol-quality synthesis gas. Onwudili et al. [24,25] investigated the hydrothermal gasification in the presence of NaOH, and concluded that the alkali could act as a promoter for hydrogen production. The use of calcium-based catalysts in the gasification process for hydrogen and/or syngas quality has also received the bulk of research attention. Shuai et al. [26] reported that a $Ca(OH)_2$ addition in lignite steam gasification had a synergistic effect, in terms of

the catalysis of the water-gas shift reaction and CO_2 absorption, resulting in the production of hydrogen-rich gas. By contrast, Chen et al. [27] observed that using the $Ca(OH)_2$ catalyst in catalytic coal gasification could improve the H_2/CO ratio and produce CH_4 -rich syngas. Moreover, CaO has been regarded as a widely used catalyst for producing hydrogen-rich gas [28–30]. Additionally, Jiang et al. [31] found that the inherent AAEM species in biomass gasification could enhance the production of H_2 and CO_2 by promoting the char-steam reaction, hydrocarbons reformation and the water-gas shift reaction. The previous literature almost emphasized on the final results of the product gases from the catalytic gasification, little attention was paid to the variation in the product gases during the course of gasification. It is well known that the changes in catalytic activity of catalysts always occur during gasification, almost owing to the loss and/or transformation of catalysts [32–34]. Meanwhile, the different profiles of char reactivity indicate the changes in gasification rate during the overall gasification, which is crucial for designing and optimizing the gasification process. As the hydrogen and carbon monoxide are the main products of gasification and resources for power generation and chemical feedstocks, the variations in these product gases are also very important for developing the gasification process. When high production of hydrogen and/or high-quality of syngas is achieved in the certain range of carbon conversion, the more-efficient and cost-competitive production of hydrogen or high-quality of syngas would be provided using the method of char staged conversion. Several multi-stage gasification systems have been developed to achieve the good performances, e.g. high efficiency, desired product gases and low level of impurity, by controlling the operating conditions and decoupling the conversion process [35–39]. The design of multi-stage gasification is essentially on the basis of the reaction mechanisms over the overall gasification. Therefore, detailed information regarding the variation in the product gas during catalytic gasification should be gain for the development of coal staged conversion technology.

Additionally, many studies have focused on the production of hydrogen and other syngas from gasification by catalytic additives, e.g. carbonates, halides and other species, but little work has been published regarding the product gases from catalytic gasification using ion-exchangeable Na and Ca species, although the superior catalytic activity of this approach in gasification has been intensively reported. The early work by Clemens et al. [40] has performed a preliminary study on the effects of exchangeable Ca on the product gases of steam gasification. They found that Ca could influence the product gases of steam gasification through promoting the water-gas shift towards equilibrium. However, the instantaneous data on the gaseous production at 20% carbon conversion level could not represent the true value of the overall gasification because the catalytic behavior of catalyst and the reaction profile always change during gasification. In addition, the variations in the product gases during the gasification, in terms of the yield rates and gas compositions, were not analyzed. In our previous study, the ion-exchangeable Na behaved differently from the ion-exchangeable Ca catalyst on the char reactivity [21]. Therefore, it is necessary to conduct a

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