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Review article

Conversion of low-grade coals in sub-and supercritical water: A review

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ABSTRACT

Low-rank coals are very abundant in several regions throughout the world. However, it is difficult to use the low-rank coals directly due to their undesirable characteristics such as high moisture, high ash and high oxygen content. The utilization of low-rank coals has received more and more attention with the increase of consumption of other fossil fuels. Besides the pyrolysis and alcoholysis, the conversion of low-lank coals under hydrothermal conditions (including sub-and supercritical water) for high-quality solid fuel, coal liquefied oil and gaseous fuels has also been widely investigated. In this review, the development of low-rank coal hydrothermal upgrading, the liquid fuel production from coal in sub-and supercritical water and supercritical water gasification of coal for hydrogen production, including partial oxidative gasification, homogeneous/heterogeneous catalytic gasification and related kinetic investigations are discussed. In addition, the element transformation such as N, S and metals during the hydrothermal process are also presented.

1. Introduction

Low-rank coals include lignite and sub-bituminous coals. These coals have low calorific value because of its undesirable characteristics, e.g., high moisture content, high ash and high oxygen content. Lignite is abundant coal resource, its reserve is estimated to be more than 4 trillion tons over the world [1], total low-rank coals constitutes about 45% of the total global coal reserves [2]. However, the commercial application of lignite is very limited due to its undesirable characteristics as mentioned above. On the other hand, the improper utilization of coal could lead to severe environmental problems, coal burning cannot get rid of the emission of NO_x, SO_x and heavy metal [3], taking Hg emission as an example, which is estimated about 1 Gg of Hg was released in the year 2010 (Asia was responsible for 69% of the total releases of Hg from coal combustion by 2010) and the annual releases of Hg from coal combustion continue to increase today [4]. Therefore, the increasing consumption of other fossil fuels and environmental problems have stimulated exploration of a high-efficiency technology to utilize the low-grade coals.

Several coal conversion technologies have been developed for use low-grade coals cleanly and efficiently, including drying and dewatering [5,6], pyrolysis gasification [7,8] and liquefaction by alcoholysis methods [1,9,10]. Among those potential technologies, conversion of coal in hydrothermal environment, especially in sub-and supercritical water (SCW, temperature 374.3 °C and pressure 22.1 MPa) have received more and more attention. One of the advantages by hydrothermal method is that high moisture content materials can be fed directly without any drying process [11]. Moreover, in the sub-and supercritical states, the water can serve as both reaction medium and reactant participating in reactions, resulting in a very high reaction efficiency. Similar to the supercritical water gasification of biomass [12], based on the desired products, the hydrothermal processes can be summarized as hydrothermal carbonization for high-quality coke, hydrothermal liquefaction for liquid fuel and hydrothermal gasification for hydrogen and methane production.

In this review, the essential properties of the hydrothermal water and low-rank coals, mainly on lignite are first introduced. And then, the development of low-rank coal carbonization, the liquid fuel production from coal in sub-and supercritical water and the SCW gasification of coal, including partial oxidative gasification, homogeneous/heterogeneous catalytic gasification and related kinetic investigations are comprehensively discussed. The element transformation such as N, S and metals during the hydrothermal process are also presented.

2. Low-grade coals and hydrothermal water

In fact, there is no single universally accepted definition of low-grade coal, however a coal can be termed as a low-grade coal if it has

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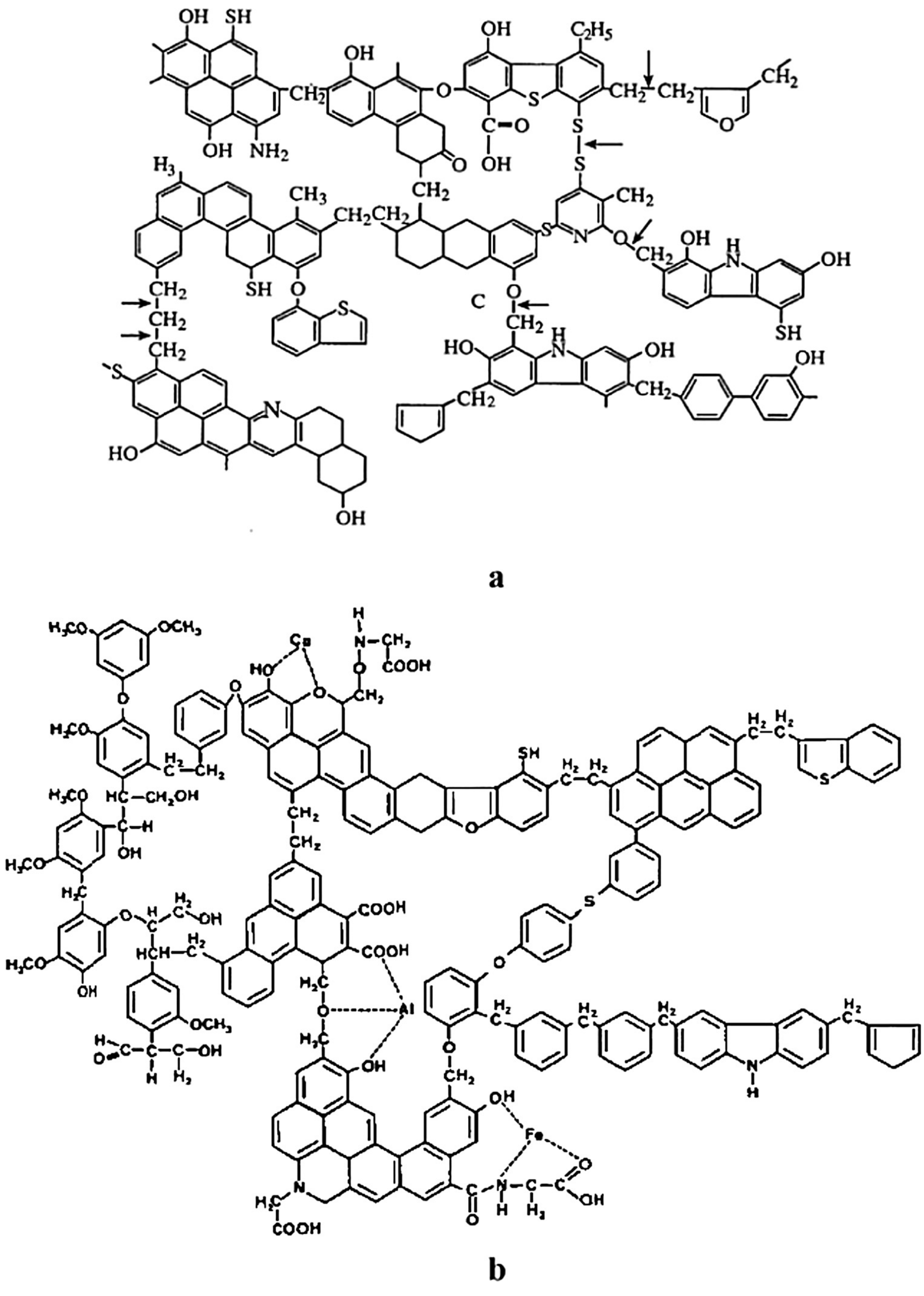

Fig. 1. (a) Wiser model for coal structure and (b) Model of Rhenish brown coal propose by Wolfrum [15].

one or more troublesome properties related to use, the troublesome properties include low heating value, high moisture content, high ash content, high sulfur content and high alkali/alkaline content [13]. Lignite is a representative low-rank coal, Li et al. [1] have summarized the proximate and ultimate analyses of different lignite, their ash and volatile matter content are usually higher than 15% and 45%, respectively. The oxygen content is usually higher than 27%. Compared with high-rank coals, lignite is more reactive due to its higher H/C ratio and oxygen content. And thus, lignite is easily converted into caking coal, liquid and gaseous fuels.

The chemical structures of coals is too complex to be elaborately characterized, but it is significantly important to deeply understand its structural features for efficient utilization of coals. The Wiser model (shown in Fig. 1a) for coal structure is extensively adopted [14], which has been considered as a relatively reasonable model, the chemical properties and liquefaction behavior of coal can be explained with Wiser model. However, in the Wiser model, the oxygen content is very low, which is inconsistent with the elemental analysis of real lignite. Wolfrum [15] showed a chemical structure of Rhenish brown coal (shown in Fig. 1b), the structure revealed the connections of lignin, humic acid and aromatic structural elements. In the model of Rhenish brown coal, the oxygen content is about 25% and the oxygen-containing functional group mainly consists of phenolic hydroxyl group (Ar–OH), alcoholic hydroxyl group (R–OH), carboxyl (–COOH), methoxyl (–O–CH_3) and ether oxygen atom (R–O–R). Note that, lignite retains more macromolecular structures of coal-forming plants and contains larger amounts of oxygen-containing moieties due to its low degree of coalification [9]. Hence, the structure model shown by Wolfrum seems to be more reasonable in description the chemical structure of lignite compared with Wiser model. Li et al. [16] found that