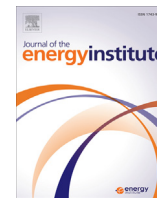




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Process and product characteristics for carbonization of liquid–solid mixtures from coal liquefaction

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ABSTRACT

In this paper, a carbonization process for valid liquid–solid separation and upgrading of coal liquefaction residues was proposed and practiced using batch-type devices. The effect of carbonization times and temperatures on the efficiency of liquid–solid separation and product distributions were investigated using self-prepared liquid–solid mixtures (CSMs) from direct coal liquefaction. It was suggested that the carbonization temperature and blowing gases were essential to the efficient liquid–solid separation in this process, and the carbonization process was characterized by the efficient liquid–solid separation. The *n*-hexane soluble fractions (HS) can be largely separated from CLMs. It was remarkable that over 0.06–1.50% of HSs can be additionally produced at the higher temperatures (430 °C–500 °C). On the other hand, the obtained semi-cokes (SCs) were mainly characterized as a rich carbon-containing material with high heating values. Some of the SCs were characterized by the better caking property with the caking index (*G*) from 68.5 to 75.2. In addition, it was proved that the caking property of SCs is directly affected by the asphaltenes (ASs) instead of pre-asphaltenes (PAs).

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1. Introduction

Direct coal liquefaction (DCL) technology, regarded as one of the potential approaches to convert coal to clean transportation fuels and valuable chemicals [14,47], has been deeply developed exactly as its successful industrialization in China this century (constructed by Shenhua Group in Inner Mongolia). However, there are still many technical challenges confronted at present [41]. A number of recent studies have been involved in various aspects of DCL technology, such as catalyst improvement [22,26], optimized operation conditions [10,24,37], feasible pretreatment of coal [9,39], utilization of heavy ends [5,18,30], demonstrating that the deep comprehension of DCL mechanism and novel technology for product upgrading are much required in the near future. Particularly, the liquid–solid separation and utilization of liquefaction residues become two practical issues which essentially impact on the efficiency, cost of investment and environmental problems in DCL processes, and still remain to be further explored [16,28,51].

The liquid–solid separations of DCL process have been accomplished by a series of approaches. Typical methods, including sedimentation, filtration, hydrocyclones and solvent extraction, were adopted to separate the solids (unconverted coal particles, catalysts and minerals) from liquefaction products for different objectives [3,7,36,38]. For instance, solvent de-ashing process was applied in the BCL process after the first stage hydrogenation to obtain de-ashed oil, preventing the deactivation of secondary hydrogenation caused by ashes [35]. In the Dow Liquefaction Process, the particle solids were removed using hydrocyclones to recycle the catalysts [32]. Filtration and sedimentation methods are considered to be efficient on the separation of smaller particles (<10 μm) along with the issues of high capital

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cost and residue disposal [19]. The complex natures of direct coal liquefaction residues (DCLR), which are inevitably produced by 20–30% of the amount of raw coal in DCL process [15,48], bring a number of difficulties on the separation process [1,19]. Specifically, DCLR are characterized by a high concentration of solids with small particle size ($\sim 1\text{--}20\ \mu\text{m}$) and a certain quantity of heavy fractions (heavy oil, pre-asphaltenes and asphaltenes), which directly cause the high viscosity and the similar density of solid–liquid phases [16,23]. Therefore, DCLR are commonly directly used as the raw materials for gasification or combustion to produce hydrogen and heat [2,6,13]. On the other hand, heavy liquid hydrocarbons and solid carbonaceous matters are abundant in DCLR [27], thus offering a great potential for economical utilizations. Based on the economic and environmental considerations, DCLR are also favorable for preparation of highly value-added products using various methods (such as extraction of valuable components, hydrogenation of heavy liquids and asphaltene modification, preparation of carbon fibers, etc.), which are rational to be considered for applications [5,20,34,45,52,54]. Nevertheless, more innovations are still much required for the development of separation and utilization technologies of DCLR.

On the basis of our previous work [49], an integrated mild liquefaction and carbonization process (named as ECOC process) was proposed for an efficient utilization of coal (especially the low rank coals) [42]. It has been suggested that the carbonization process is favorable to achieve a valid separation of liquid–solid mixtures and produce value-added carbonaceous products [55]. However, the process characteristics (such as the effect of process conditions and the properties of solid products) still need an individual study for more comprehensive understanding of the process. In the present work, the proposed carbonization process in ‘ECOC process’ was studied separately. This paper provided the detailed investigation results of those that are indispensable for the process development.

2. Experimental

2.1. Preparation of raw material

In this study, the liquefied coal-solvent mixtures (CSMs) prepared by the liquefaction using coal and solvent were used as the raw material. The coal was provided by Shaanxi Coal and Chemical Industry Group Co., Ltd, containing 3.98% of moisture, 37.19% of volatile matter, 62.81% of fixed carbon and 6.82% of ash. The ultimate analysis showed that the raw coal (daf basis) consists of 84.77% carbon, 5.40% hydrogen, 1.12% nitrogen, 0.25% total sulfur (dry basis) and 8.46% oxygen (by difference). The tetralin and $\gamma\text{-FeOOH}$ were used as donating-solvent and catalyst for liquefaction. Coal liquefaction was achieved in a 500 mL stirring autoclave [12]. The liquefaction temperature, resident time and solvent/coal ratio were designed as $410\ \text{C}$, 60 min and 1.5:1. The detailed liquefaction operations were illustrated in our previous work [49]. In order to guarantee the homogeneity of chemical compounds, the CSMs were abundantly prepared and gathered after fully mixing, then preserved under low temperatures ($0\text{--}4\ \text{C}$). The compositions of CSMs determined by solvent extractions were mentioned in Section 2.3.

2.2. Procedures of carbonization

The carbonization of CSMs were carried out using a self-built apparatus. The schematic diagram of carbonization apparatus was illustrated in Fig. 1. For a typical run, a 75 g CSMs was loaded at the bottom of a batch type reactor. After sealing the reactor, the tubes connected to the reactor were heated to $200 \pm 10\ \text{C}$ and the blowing gas was entrained at the prescribed rate justified by a mass flow meter. Then, the reactor was placed into a salt bath, which had been preheated to the prescribed temperatures ($320\text{--}500\ \text{C}$). In order to detect the carbonization temperature, a thermocouple was deeply insert into the reactor to insure a complete contact to CSMs. The carbonization was hold for a predetermined time (10–120 min) after CSMs were rapidly heated to designated temperatures (within 3–5 min). The liquids were purged out and condensed in cold traps, which were cooled by an ice-water bath loaded with salt water and approximately hold at -4 to $0\ \text{C}$. A small amount of organic vapor was adsorbed by an adsorption flask filled with acetone. To terminate the carbonization process, the

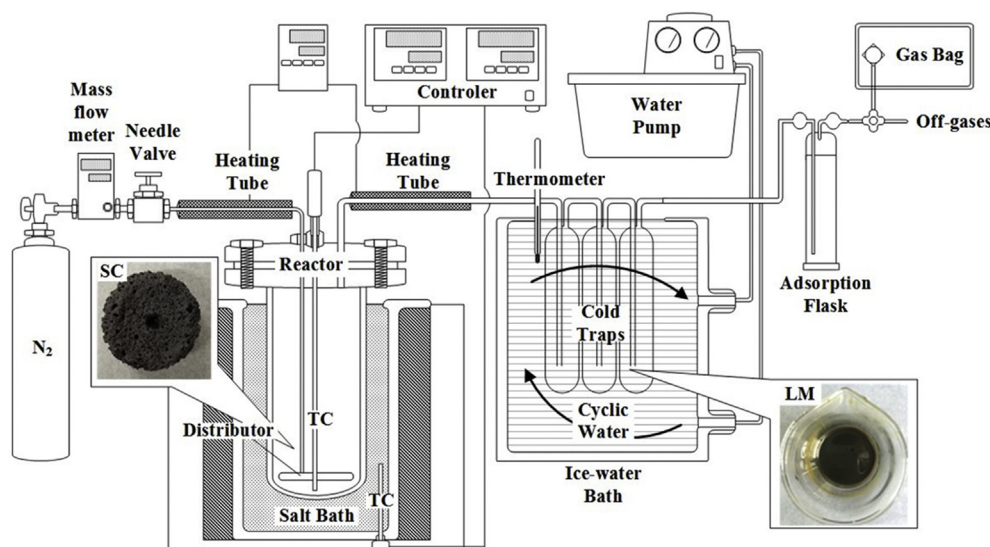


Fig. 1. Schematic diagram of self-built carbonization apparatus.

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