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Characteristics comparison of tar from lignite pyrolysis with inherent and simulated moisture for adopting a two-stage gasification process

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ABSTRACT

To utilize low rank coal cleanly and high efficiently, a new fluidized bed two-stage gasification (FBTSG) process, mainly consisting of a FB pyrolyzer and a transport bed gasifier, has been proposed to treat pressurized coal (below 10 mm) with relatively high moisture for the production of clean fuel gas. In this study, a laboratory FB pyrolysis apparatus was adopted to examine the influence of moisture mode on pyrolysis behavior from 600 °C to 900 °C, especially in the yield and property of pyrolysis tar. Two common experimental methods in literatures, namely wet coal with inherent moisture and dried coal with the simulated moisture by introducing steam, were systematically analyzed. Compared to the simulated moisture, inherent moisture in coal can facilitate effectively the generation of tar at 700 °C and 800 °C, and had strong reforming effect on tar conversion to light fraction, leading to more production of light fractions and micro-molecule compounds. All of these differences were much related to the difference in interaction pathway between volatile matter and moisture.

1. Introduction

Worldwide, coal represents one of the most important fossil resources in primary energy. Among the numerous kinds, low rank coal, including lignite, peat and sub-bituminous coal, will play an increasingly important role in industrial applications, because of large reserve (430 billion tons, nearly 50% of the total reserve), low mining cost, high reaction reactivity, rich in volatiles, and low pollution-forming impurities, such as sulfur, nitrogen and heavy metals [1–3]. However, the high moisture of low rank coal (40%–60%) makes it a great challenge for high-efficient utilization [4–6]. Generally, for the thermal conversion processes, including pyrolysis, gasification and combustion, the requirement in moisture for the adopted coal is generally about 10% [7,8]. To control the moisture content, the complex dehydration or drying processes have to be employed, requiring large amount of energy and power [9–11]. On the other hand, the existing studies have demonstrated that the inherent moisture in coal has desirable effect on the reactions of pyrolysis, gasification, combustion, and tar reforming by improving the reaction activity [12,13]. Perhaps the utilization of low rank coal with inherent moisture has higher efficiency than that treated by drying process. So, for the clean, efficient utilization of low rank coal, whether the moisture content in the adopted coal can be improved further and what is the suitable moisture content to be maintained for thermal-chemical conversion [14] process? This makes it very necessary and essential to understand the role of moisture in process characteristics and improve the feasibility in feedstock with high moisture by innovating reactor structure and thermal conversion technology.

According to the experimental method adopted, the existing researches in literatures on the role of moisture in pyrolysis and gasification can be divided into two categories, namely using the wet coal with inherent moisture and the pre-drying coal with simulated moisture by introducing steam [15–18]. Yip et al. [19] found that the influence of inherent moisture in Collie coal during pyrolysis was much related to reactor structure, residence time, heating rate and reaction temperature. For example, in the fluidized bed reactor, the significant interactions between in-situ steam from moisture and coal particles, can be observed only at the temperature above 800 °C, leading to much lower

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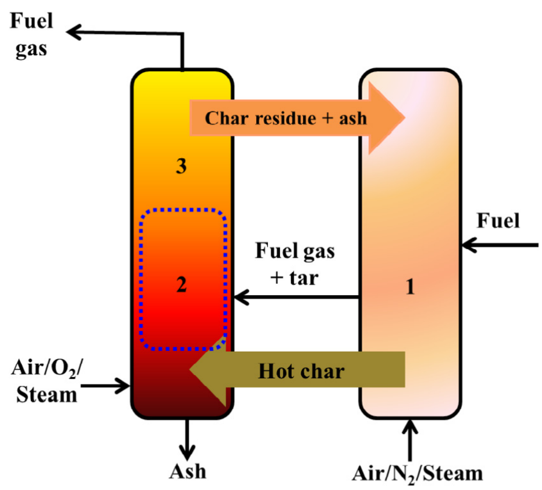

Fig. 1. Conceptual diagram of the proposed two-stage gasification process.

char yield. Hu et al. [20] found that drying and pyrolysis occurred in a relatively shorter time, and compared to the dried coal, the interactions between the auto-generated steam from inherent moisture and the intermediate products, such as tar and char, can be enhanced evidently, leading to more hydrogen produced. Xiong et al. [21] reported that at 1000 °C, with the increase of moisture, the relative content of PAHs with 2 rings was significantly increased, while the PAHs with 6–7 rings presented a major decrease, decreasing the tar yield. Demirbas et al. [22] reported that compared to the dried coal, the inherent moisture content (34.7%) in wet coal can not only increase the tar yield from 22% to 34%, but also decrease the viscosity of tar sample. On the other hand, Pütün et al. [23] found that, the yield of tar from steam atmosphere was much higher nearly about 20% than that in inert atmosphere. However, for the similar experiment conducted by Tyler et al. [24], the results indicated that adsorbing water vapor in coal had little effect on coal pyrolysis. Furthermore, Gray et al. [25] found that the presence of moisture or steam increased the yield of char, suppressed the formation of tar for the demineralized coal sample, but enhanced it for the ash-containing coal. The analysis mentioned above indicated that the conclusions about the effect of moisture/steam in pyrolysis were very different and confusing. Moreover, there were little researches on the comparison of different moisture modes directly. So, in the author's opinion, there are still some important points to clarify, such as the role of inherent moisture in pyrolysis and the different effect between inherent moisture and simulated moisture on coal pyrolysis.

On the other hand, in recent years, the concept of multi-stage or decoupling thermal conversion process has been proposed, such as the processes of two-stage gasification or decoupling gasification [26,27]. By physically separating fuel gasification into fuel drying, fuel pyrolysis and char gasification via different but connected reactors or sections, it is expected to be feasible in coal with high moisture and effective for the production of clean fuel gas with low tar content. A typical biomass two-stage gasification technology was developed by Technical University of Denmark [28,29], consisting of a screw pyrolyzer for drying and pyrolysis of woody chips, and a downstream fixed bed gasifier for char gasification and tar catalytic reforming by hot char bed. Up to now, this process has been verified on a series of pilot (75 kW) and demonstration plants (200 kW, 500 kW), realizing the treatment of woody chips with moisture of about 30%, low tar content in gas (< 15 mg/Nm^3), and high cold gas efficiency (> 95%). Another representative process, namely fluidized bed two-stage gasification (FBTSG) process, has been proposed and developed by the Institute of Process Engineering (IPE), Chinese Academy of Sciences (CAS) [30,31]. As illustrated in Fig. 1, it mainly consists of a FB pyrolyzer and a transport fluidized bed (TFB) gasifier. Firstly, fuel is dried and then pyrolyzed autothermally in the FB pyrolyzer. All the pyrolysis products, namely tar, char, fuel gas, and steam, will be blown into the downstream transport bed gasifier to conduct char gasification and fuel gas upgrading. The produced steam in pyrolyzer will be acted as reactant for char gasification and tar catalytic reforming. According to the results from a demonstration plant with a treating capacity of 10,000 t/a of herb residue, this process realized the treatment of powder feedstock (below 10 mm) with a moisture content below 25%, and lower tar content in fuel gas. However, the effect of moisture on reaction behavior, especially for the tar yield and tar property, is still not very clear in this process. So, to research the FBTS gasification process more successfully and expand its application in coal, it urgently requires a better and clearer understanding of the role of inherent moisture in reaction behavior, production distribution, and product property from the processes of coal pyrolysis and gasification.

In this study, a laboratory fluidized bed reaction apparatus was adopted to examine the influence of moisture on pyrolysis behavior over a temperature range from 600 °C to 900 °C, especially in tar yield and component. Meanwhile, to elucidate the difference between inherent and simulated moisture, a systematical comparison on tar yield, distribution of distilling fraction, tar component, and kinds of functional group, will be conducted by multi-scale analysis from the methods of simulated distillation, gas chromatography-mass spectrometer (GC–MS) and Fourier transform infrared spectroscopy (FTIR). Finally, the interaction pathway between volatile matter and char during the pyrolysis of coal with inherent moisture and simulated moisture was analyzed to probe the difference in tar yield and its property. As a consequence, this study hopes to provide a clear understanding about the role of inherent moisture in pyrolysis and also to facilitate the development of FBTS gasification process for the utilization of low rank coal.

2. Experimental

2.1. Coal sample

A kind of lignite from Shenli coal mine in China was adopted in this study, whose typical property was listed in Table 1. From the proximate, ultimate and ash analysis, one can see that the contents of carbon and hydrogen in coal and the contents of CaO, MgO and Fe_2O_3 in ash were relatively abundant, indicating good gasification reactivity. Before utilization, the collected raw sample from coal mine was crushed by a hammer breaker, sieved to obtain samples with a particle size over the

Table 1
Property of coal sample used in experiments.

Coal analysis	Proximate analysis [wt. %, d]				Ultimate analysis [wt. %, daf]					LHV [MJ/Kg]
	A	V	FC		C	H	S	O^a	N	20.42
	16.7	36.2	47.1		75.1	4.3	1.1	18.7	0.8	
Ash [wt.%]	CaO	SO_3	SiO_2	Al_2O_3	MgO	Fe_2O_3	Na_2O	P_2O_5	TiO_2	
	26.8	22.8	25.8	11.6	2.1	4.7	0.6	1.0	2.5	

①d: dry basis; ② daf: dry and ash-free basis; ③^a By difference.