



## Interaction and its induced inhibiting or synergistic effects during co-gasification of coal char and biomass char



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### HIGHLIGHTS

- Both inhibiting and synergistic effects were observed during co-gasification.
- The deactivation of K was caused by the formation of  $\text{KAlSiO}_4$ .
- The gasification rate discrepancy determines what kind of interaction will happen.

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### ABSTRACT

Co-gasification of coal char and biomass char was conducted to investigate the interactions between them. And random pore model (RPM) and modified random pore model (MRPM) were applied to describe the gasification behaviors of the samples. The results show that inhibiting effect was observed during co-gasification of corn stalk char with Hulunbeier lignite coal char, while synergistic effects were observed during co-gasification of corn stalk char with Shenmu bituminous coal char and Jincheng anthracite coal char. The inhibiting effect was attributed to the intimate contact and comparable gasification rate between biomass char and coal char, and the loss of the active form of potassium caused by the formation of  $\text{KAlSiO}_4$ , which was proved to be inactive during gasification. While the synergistic effect was caused by the high potassium content of biomass char and the significant difference of reaction rate between coal char and biomass char during gasification.

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### 1. Introduction

In the new century, the great demands of energy have caused huge depletion of fossil fuels and simultaneously led to severe environmental problems, including air pollution, global warming, acid rains, etc (Wall et al., 2002). These issues force us to seek a renewable and clean resource to partially substitute fossil fuels. Fortunately, biomass such as agriculture residue, forest residue and municipal solid waste, is considered as the fourth largest fuel following oil, coal and natural gas and can be used as renewable and clean energy resources. Due to the remarkable advantages including fast growing, wide distribution and  $\text{CO}_2$  neutral, biomass becomes a promising energy resource to partially replace fossil fuels (Di Blasi, 2009).

There are many thermal processes which can be used to deal with biomass resources, like gasification, combustion and liquefaction. Among these technologies, gasification is an effective way for

producing syngas from biomass (Fermoso et al., 2009; Zhang et al., 2008). Besides the abovementioned advantages, biomass also has other merits such as high gasification reactivity, low ash content, low sulfur and nitrogen content. However, biomass individual gasification does have some obstacles when operated in large scale: (1) It will be a great cost for collecting, transporting and drying original biomass materials due to its low energy density and high water content, (2) The supply of biomass resource varies with region and climate and (3) The low gasification temperature will lead to relatively high content of tar (Krerkkaiwan et al., 2013). While it is believed that co-gasification of coal and biomass provides a better way to utilize both coal and biomass, due to the following factors: (1) Most biomass materials are rich in alkali and alkaline earth metal, thus biomass can be used as a cheap and effective catalyst to improve the efficiency of coal gasification when co-gasified with coal (Jeong et al., 2014; Yuan et al., 2012), (2) Stable supply of gasification materials can be guaranteed, which overcomes the shortage of biomass induced by the seasonal influence (Howaniec et al., 2011), (3) Compared with biomass gasification, the higher temperature of coal gasification could reduce

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the formation of tars (Krerkkaiwan et al., 2013), (4) The emission of CO<sub>2</sub> and other pollutant gases (SO<sub>x</sub>, NO<sub>x</sub> etc.) can be reduced (Spliethoff and Hein, 1998).

Gasification mainly consists of two steps: (1) pyrolysis of the raw samples and (2) gasification of the residual chars. Compared with devolatilization, the gasification rate of char is much slower, and thus the latter becomes the rate-limiting step during gasification and studies on the kinetics of co-gasification behaviors of char are of great importance in designing a gasification reactor, while the interactions between coal char and biomass char during co-gasification can influence the gasification behaviors. As a result, many studies have focused on the interactions between coal char and biomass char during co-gasification, and synergistic effect, inhibiting effect and no influence have been observed (McLendon et al., 2004). Kajitani et al. (2009) carried out co-gasification of two bituminous coal with cedar bark in CO<sub>2</sub> atmosphere and synergistic effect was observed at 850 and 950 °C in the thermogravimetric analyzer (TGA) and 1200 °C in the drop tube furnace. However, no distinguished synergy was found at 1400 °C. Yuan et al. (2012) prepared mixed chars from a bituminous coal and rice straw in a drop style high-frequency furnace, and the gasification results carried out in TGA showed that synergy only occurred when the mass ratio of coal and biomass was 4:1, but high biomass ratios (1:1 and 1:4) not only weakened the synergies but also decreased the gasification reactivity of the residual char. Moreover, both inhibiting and synergistic effects were observed in the research of Habibi et al. (2012), where the biomass char was mixed with coal char or fluid coke char, and obvious inhibiting effect was observed during co-gasification of biomass char and coal char, in contrast, enhancement was found during co-gasification of biomass char and fluid coke char.

Thus synergistic and inhibiting effects were all observed in the co-gasification experiments, and some researchers have been trying to explain the underlying reasons and mechanisms of the interactions. Lahijani et al. (2013c) co-gasified both biomass char and acid-treated biomass char with tire char, the results showed that the reactivity of blended samples was lowered when acid-treated biomass chars were co-gasified with tire char, such reduction became more severe when empty fruit bunch was used, since the fruit bunch contained a higher level of K, which signified the profound catalytic effect of K during gasification. Habibi et al. (2012) observed that when the ratio of K/Al in the mixed char sample of biomass and coal (or fluid coke) was less than 1, K may bound to Al and Si to form KAlSi<sub>3</sub>O<sub>8</sub> and inhibit gasification. But when the K/Al ratio was higher than 1, it will allow excess potassium to activate the catalytic effects and improve the gasification efficiency. However, due to the complexity of coal and biomass, the underlying reasons and mechanisms/pathways of these effects are still unclear. However, this is very important to the industrial catalytic gasification, where the catalysts must keep high activity and be easily recycled by hot water leaching or other simple methods. But the catalysts usually react with the mineral matters in coal to form inactive or water insoluble compounds, which deactivate the catalysts and make the recycle of potassium catalyst difficult (Bruno et al., 1988; Kühn and Plogmann, 1983). Thus, deeper insights about the interactions between mineral/mineral or char/mineral and the deactivation of catalyst during co-gasification of coal char and biomass char should be obtained before a successful catalytic gasification process can be developed. In addition, these are also attractive academic areas need to be explored.

In this paper, the coal char and biomass char samples were prepared in a fixed bed reactor, the co-gasification of coal char and biomass char was conducted, and the interactions between biomass char and coal char and its induced inhibiting or synergistic effects during co-gasification were investigated. In addition, the chars were characterized and analyzed in detail to interpret the

underlying mechanisms for the interactions observed during co-gasification.

## 2. Methods

### 2.1. Raw materials

Corn stalk (CS), collected from the southeast of Shanxi province, is the most abundant agriculture residue in China and rich in alkali metal, thus it was chosen as a representative biomass sample. Three different ranks of coals, Hulunbeier lignite coal (HL), collected from the east of Inner Mongolia, Shenmu bituminous coal (SM), collected from Shaanxi province and Jincheng anthracite coal (JC), collected from the southeast of Shanxi province, were selected as coal samples.

CS was crushed and sieved to a particle size of less than 600 μm, and coal samples were ground and sieved into a particle size of 100–120 μm. All the samples were dried in an oven at 105 °C for 12 h, and then stored in a desiccator for further use.

### 2.2. Preparation of char, ash and KAlSiO<sub>4</sub> samples

Char samples were prepared in a fixed bed reactor, which was modified from a high pressure thermogravimetric analyzer (PTGA, Cahn TG-151, Thermal Cahn, USA). The schematic of the reactor is shown in Fig. 1. The main part of the reactor is a pressure vessel (furnace vessel) and an inside quartz tube (34 mm internal diameter, 325 mm length), which is isolated from the furnace vessel by two fluororubber (FKM) O-rings. The furnace vessel can withstand temperatures of 1100 °C (0.1 MPa) and 1000 °C (6.9 MPa). In each run, the alumina crucible charged with samples (biomass about 500 mg, coal about 1500 mg) was hung on a platinum wire and put into the sample cell (cooled zone), and then the screw nut was tightened. The furnace was heated at a ramp of 10 °C/min to the pre-set temperature under a flow of N<sub>2</sub> (200 ml/min for purge gas and reactant gas, 400 ml/min for furnace gas). When the temperature of the isothermal zone reached 900 °C and stabilized for a while, the ball valve was opened, and then the alumina crucible was fast lowered to the isothermal zone (about 50 mm long) by lowering the magnet, meanwhile, the furnace temperature was suddenly dropped but would back to normal within 30 s. The sample was held in the isothermal zone for 30 min till most of the volatiles evaporated. After that the crucible was lifted up to the sample cell and kept there until the sample temperature fell to room level. Then the screw nut was loosened and the crucible was taken out. The residual char was ground to a diameter of less than 100 μm and stored as char sample.

Ash samples were prepared in a horizontal quartz tube reactor, under steam atmosphere (10 vol.%, diluted by N<sub>2</sub>), the same condition as the gasification experiments performed on TGA (details will be illustrated in Section 2.5). Char samples, either biomass char, coal char or blended char were loaded in a ceramic boat and pulled into the middle of the quartz tube, then all the connected parts was sealed, and the quartz tube was heated at a ramp of 10 °C/min to the pre-set temperature under N<sub>2</sub> (purity ≥ 99.999%). When the desired temperature reached, steam was introduced into the tube by the steam generator to start gasification. The experiment was ended until no product gas (mainly H<sub>2</sub> and CO) was detected in the vent of the system. Then pure N<sub>2</sub> was purged into the tube until the temperature dropped to room level. Ash samples were collected and stored in a desiccator for further analysis.

### 2.3. Preparation of acid-treated char

In order to exclude the influence of inherent mineral matter in gasification experiments, biomass char was demineralized by acid

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