



## Full Length Article

## Co-gasification reactivity of petcoke and coal at high temperature

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## HIGHLIGHTS

- Co-gasification reactivity of petcoke with coal was studied at high temperature.
- The reactivity of petcoke can be improved greatly by co-gasifying with coal.
- The synergistic effect is closely related to the minerals in coal but not coal rank.
- The Ca-/Fe-components is beneficial for gasification of petcoke even at 1400 °C.

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## ABSTRACT

Petcoke, as a byproduct of petroleum refinery process, can be used as feedstock of gasifiers due to its high carbon content and high calorific value. However, its low gasification reactivity and low ash content restrict its application to some gasifiers. In this work, experimental study on co-gasification reactivity of petcoke with coal under CO<sub>2</sub> atmosphere was investigated in a drop-in-fixed-bed reactor in the temperature range of 1200–1400 °C. Produced CO was monitored continually using an online mass spectrometry to calculate the carbon conversion and gasification rate. The experimental results reveal that the gasification reactivity of petcoke can be improved greatly by co-gasifying with coals or by loading their ashes, even at temperatures higher than the ash fusion temperature. Both the coal and its ash can improve the gasification reactivity of petcoke. The synergistic effect is closely related to the composition of minerals in coal but not coal rank. The high content of active components such as Ca- and Fe- in coal or ash is beneficial for co-gasification. Some coals, however, are lack of these active components and with high Si- and Al-components content which may retard the gasification reaction. During co-gasification, the oldhamite (CaS), anhydrite (CaSO<sub>4</sub>) and magnetite (Fe<sub>3</sub>O<sub>4</sub>) have positive catalytic effect, while mulite (3Al<sub>2</sub>O<sub>3</sub>·2SiO<sub>2</sub>) has negative effect on gasification reaction at high temperature.

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## 1. Introduction

With the increasing demand of petroleum and the installation of more petroleum deep conversion process units, the output of petcoke has increased rapidly [1]. Petcoke has a high carbon content and calorific value which usually be used as fuel to combust, inevitably with low efficiency and high pollution especially high-sulfur petcoke. Fortunately, it is increasingly considered as an attractive feedstock for gasification to produce hydrogen, power, and steam in the petroleum industry [2,3]. However, the low reactivity and ash content of petcoke has greatly restricted its applicability for gasification [4–6].

Increasing the gasification temperature is one of the most effective methods to improve the gasification reactivity of petcoke. In our previous work [7], it was found that the CO<sub>2</sub> gasification rate of petcoke increases 25 times as the temperature increases from 1200 °C to 1600 °C. However, the gasification rate of petcoke was still several times lower than that of coals even at 1600 °C in both CO<sub>2</sub> and steam atmosphere. In order to achieve a high carbon conversion, a higher temperature or a larger residence time is necessary. But this will reduce the gasification efficiency and economics. Moreover, the low ash content of petcoke is not suitable for entrained-flow slagging gasifiers with a cooled membrane wall.

On the other hand, it is known that coal is the most common feedstock used for gasification. The reactivity of coal often varies with its rank, which decreases from low rank coal to high rank coal. The low rank coal usually has a high reactivity as its high content of

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volatile and mineral matter [8]. However, it is not often used as feedstock in most entrained-flow gasifiers, which is because of its high moisture, low calorific value and low concentration in coal-water slurry [9,10]. How to utilize the large deposits of low rank coal remains a serious problem, especially in China.

Considering the characteristics of petcoke and low rank coal, co-gasification will be a better method to improve the low reactivity of petcoke and also offer a suitable way to utilize the large amount of low rank coal. Understanding the co-gasification behavior and reaction mechanism of petcoke and coal is essential for the development of co-gasification technology. Up to now, there are a number of studies concerning on co-gasification of coal and petcoke. It is important to note that some synergetic effects may occur during co-gasification of petcoke with coal or other materials [11–13]. Due to the synergy effect, the gasification reactivity, carbon conversion, syngas heating value and cold gas efficiency of petcoke can be improved by co-gasifying with coal or other materials [14–16].

However, it should be noted that although the studies in the literatures are valuable for understanding the co-gasification behavior of petcoke with coal, there is still some insufficiency especially for co-gasification reaction mechanism at high temperature. As the complicated properties of petcoke and coal, the detail reasons of synergetic effect during co-gasification is not very clear. It is worth to verify which components in coal has positive effect on co-gasification reaction at high temperature. Moreover, it is necessary to investigate whether the synergetic effect is existent at temperatures higher than the coal ash fusion temperature.

In this work, an attempt was made to understand the co-gasification behavior of petcoke with coal with respect to CO<sub>2</sub> in the temperature range of 1200–1400 °C. A drop-in-fixed-bed reactor coupled with a mass spectrum (MS) was used to analyze the co-gasification reactivity. Four different rank coals, including two low rank coal, one middle rank coal and one high rank coal were chosen to seek the key factor that influence the co-gasification reactivity. The effect of coal ash on gasification of petcoke was investigated. XRD method was used to analysis the major mineral matters in the residual char.

## 2. Experimental section

### 2.1. Samples

Four coals from different rank (XLT and SL, low rank coal; XCG, middle rank coal and GP, high rank coal), and two petcokes (YS and JS) were selected. The samples were pulverized to pass through a 100-mesh screen (0.149 mm) and dried in vacuum oven at 110 °C for 24 h prior to use as the raw sample. The results of the proximate and ultimate analyses of the raw samples are listed in Table 1.

The powder petcoke and coal or ash sample were thoroughly mixed with a certain ratio in a mortar for 5 min. Then, about 10% of distilled water was added to the blends. Finally, the blends were

compressed into pellets of about 3 mm in diameter and 60 mg in weight. The choice of the pellet was to facilitate the feeding of the samples into the drop-in-fixed-bed reactor. The cylindrical pellets were formed by placing the wetted samples into a mould and applying a pressure of 0.5 MPa. Under the same preparation conditions, the thickness of the pellets is more or less the same.

### 2.2. Gasification experiments

The experiments were performed in a drop-in-fixed-bed reactor which was described in detail in our previous work (Fig. 1) [7]. The alumina reaction tube is placed vertically with a constant-high-temperature of about 100 mm. The gas is feed in the reaction tube from the bottom. The sample is charged from the top by the sample feeder.

In the experiment, CO<sub>2</sub> with atmospheric pressure was used as gasifying agent. The flow rate of CO<sub>2</sub> is 500 mL/min which was selected to avoid the effect of flow rate on gasification reaction. A single pellet was placed on the sample feeder. When the reactor reached to the reaction temperature, the pellet was fed and being placed on the reaction bed within 0.51 s (estimated by considering the gravity and gas drag force). The gaseous products were immediately swept into an online Mass Spectrometry by the gas flow and monitored continuously.

Here it should be mentioned is that the pellet mentioned above may burst into powder after it dropped into the reaction bed. Fig. 2 shows the change of the particle size for typical test samples before and after fed into the reaction bed, in which the sample is devolatilized in Ar under atmospheric pressure at 1400 °C for 1 min. It shows that the coal/petcoke pellets are likely burst into powder.

### 2.3. Data processing

The carbon conversion ratio was calculated based on the profile of CO obtained by transforming the data from the online MS. It can be assumed that the peak area of the CO profile is proportional to the amount of CO produced during the reaction. The carbon conversion ratio can be calculated by Eq. (1).

$$X = \frac{n_{t,CO}}{n_{total,CO}} = \frac{k_{CO} \cdot S_{t,CO}}{k_{CO} \cdot S_{total,CO}} = \frac{S_{t,CO}}{S_{total,CO}} \quad (1)$$

where  $X$  is carbon conversion ratio;  $n_{total,CO}$  is the total amount of CO produced;  $n_{t,CO}$  is the amount of CO produced from time 0 to time  $t$ ;  $S_{total,CO}$  is the total peak area of CO profile;  $S_{t,CO}$  is the peak area of CO profile from time 0 to time  $t$ ; and  $k_{CO}$  is the corresponding coefficients for the peak area of CO. The gasification rate,  $r$ , is defined as the differential of the carbon conversion ratio with respect to time and can be derived from Eq. (1) as:

$$r = \frac{dX}{dt} = \frac{1}{S_{total,CO}} \cdot \frac{dS_{t,CO}}{dt} \quad (2)$$

The data process procedure can be seen in our previous work [7], where the repeatability of the method was also confirmed.

**Table 1**  
Proximate and ultimate analyses of the samples.

Samples	Proximate (wt%)			Ultimate (wt%, daf)				
	A <sub>d</sub>	V <sub>d</sub>	FC <sub>d</sub>	C	H	O <sup>a</sup>	N	S <sub>t</sub>
XLT	10.34	42.86	46.80	77.20	4.81	15.51	1.26	1.22
SL	43.16	27.98	28.86	75.40	4.75	17.43	1.22	1.20
XCG	4.11	34.84	61.05	81.73	4.73	12.06	1.23	0.25
GP	22.84	9.14	68.02	88.06	3.81	6.30	1.36	0.47
YS	0.88	7.99	91.13	88.97	3.12	5.54	1.01	1.36
JS	1.54	9.83	88.63	84.98	1.90	5.79	0.93	6.40

A: ash; V: volatile matter; FC: fixed carbon; d: dry basis; daf: dry ash-free basis.

<sup>a</sup> By difference.

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