



## Research article

## Experimental study and modeling of heavy tar steam reforming

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## ABSTRACT

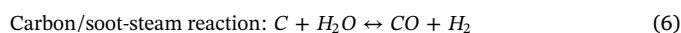
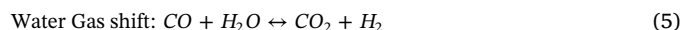
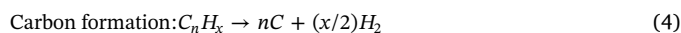
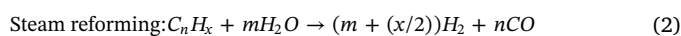
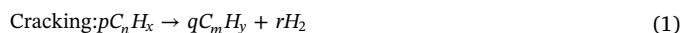
In this paper, the elimination of heavy tar by steam reforming at high temperature was studied with experiments and numerical simulation. The experiments were conducted in a tube reactor with five model compounds at temperatures of 1273–1673 K, steam of 0–40 vol% and sample weight of 3–20 mg with residence time of 2 s. The simulation was performed with the plug flow model in CHEMKIN program based on a kinetic model that consists of > 200 chemical species and 2000 elementary step-like reactions. The results of experiments indicate that increasing temperature will accelerate the decomposition of heavy tar; moisture could significantly prompt the decomposition of heavy tars, but not so efficient above 20 vol%; less sample could help the decomposition of heavy tar; longer residence time can slightly increase the decomposition of heavy tar. At high temperature (1573 K or above), the heavy tar is mainly cracked into two parts: the one carbon molecules (CO and CO<sub>2</sub>) and the soot at the beginning of reactor. Then the soot decomposes into CO, CO<sub>2</sub> and H<sub>2</sub> by steam reforming. The conversion and kinetic data of heavy tar can be divided by two temperature sections. The carbon conversion degrees of different model compounds from 1173 K to 1473 K are close with similar trends, which may be represented by a universal kinetic model. Our results provide an insight in the understanding of the heavy tar decomposition and give the necessary information for the designation and operation of the tar cracker to eliminate heavy tar.

## 1. Introduction

At present, the circulating and bubbling fluidized bed technology has been successfully used in the gasification of coal and biomass with a high production capacity [1]. However, the major problem of biomass and coal gasification is the presence of tar in the produced that blocks the pipeline and causes corrosion, erosion and abrasion of equipment. Therefore, it is necessary to remove tar contents in the product gas to an acceptable value or transform tar into other valuable gas, either in the gasifier or in the downstream.

Tar is a complex mixture of single-ring to 5-ring aromatic compounds with other oxygen-containing hydrocarbons and complex poly-aromatic hydrocarbons (PAH) [2,3]. There are many techniques to remove or transform tar [4,5]. Many experiments have been conducted on the thermal and catalytic cracking of primary tars and secondary tars, including naphthalene a tertiary tar, based on model components, such as phenol [6], toluene [7–11], naphthalene [12–14]. Also, several numerical simulations on tar reforming with light model compounds, such as benzene and toluene, have been performed with reaction kinetic models, and some simple reaction schemes have been built up [1,2,4,9].

The main reactions involved in these processes on the model compounds are as follows:



The syngas from fluidized bed gasifier contains both heavy tar and steam [15–17]. If adding a tar cracker at the outlet of the gasifier to increase the temperature of syngas, the steam reforming will occur and eliminate heavy tar into H<sub>2</sub>, CO and CO<sub>2</sub>. To design the tar cracker and reduce the heavy tar in the syngas, it is essential to study the process and mechanism of heavy tar steam reforming at high temperature. Though many researchers have studied the steam reforming of light tar, less data and studies are found on the heavy tar and at high temperature (> 1273 K), especially for the compounds more than two benzene

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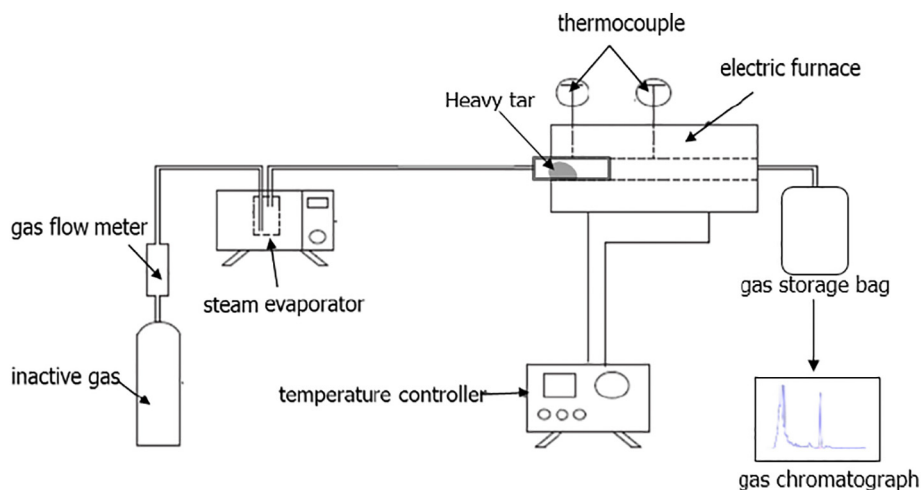


Fig. 1. Schematic graph of experimental setup (solid feeding of PAHs).

rings.

In this study, naphthalene, phenanthrene, acenaphthylene, fluoranthene and pyrene will be used as model compounds to study the steam reforming of heavy tar at high temperature (as high as 1573 K). Naphthalene represents two-ring compounds; acenaphthylene and phenanthrene represent three-ring compounds; pyrene and fluoranthene represent four-ring and higher compounds, which are the major contents in their class of heavy tars [17–21]. The effects of the reactor temperature profile, operating temperature, steam content, sample weight and residence time on the carbon conversion degree of heavy tar will be investigated in a tube reactor. The steam reforming process will be described and studied by both experiments and simulation. The kinetic data of five model compounds will be achieved.

## 2. Experimental

### 2.1. Experimental setup






The newly designed horizontal tube reactor is shown as the electrical furnace in Fig. 1. This reactor is special modified with large heating area (800 mm in length, 30 mm in diameter) and long isothermal space (250 mm range from 773 to 1673 K). There is a corundum sample injector that can be controlled to put sample in anywhere inside heating area of the tube reactor. The temperature distribution of the tube reactor was measured by a Pt-Rh thermocouple (HT1270S, Beijing Aerospace Oriental, China) with accuracy class of  $\pm 0.25\% \times \text{temperature}$  (e.g.  $\pm 2.5$  K at 1273 K).

Naphthalene (99.6%, Xiya Chemical Industry, China), phenanthrene (97%, Xiya Chemical Industry, China), acenaphthylene (99%, Xiya Chemical Industry, China), fluoranthene (98%, J&K Scientific, China) and pyrene (98%, J&K Scientific, China) were used as model compound representing heavy tars. The reacting gas is a mixture of Ar (> 99.999%) and steam from steam generator.

3–20 mg model compound was put on the sample injector and then was moved to the beginning (at 20 cm) of the isothermal space at certain temperature (1073–1673 K) and flow rate (0.5–10 L/min at s.t.p.). The model compound was vaporized immediately and carried by the gas flow through the high temperature isothermal area. The gas product after condensation and filtration was collected by a gas storage bag. When the gas bag was cooled to room temperature, the composition of the product gas was analyzed by gas chromatograph (490 Micro GC, Agilent, United States). Reaction conditions of the experiments are listed in Table 1.

Table 1

Reaction conditions.<sup>a</sup>

Tar model compounds	Sample weight (mg)	Temperature (K)	Steam (vol%)	Residence time(s)
Naphthalene 	3–20 (10)	1273–1673 (1573)	0–40 (30)	2–10 (2)
Acenaphthylene 	3–20 (10)	1273–1673 (1573)	0–40 (30)	2–10 (2)
Phenanthrene 	3–20 (10)	1273–1673 (1573)	0–40 (30)	2–10 (2)
Fluoranthene 	3–20 (10)	1273–1673 (1573)	0–40 (30)	2–10 (2)
Pyrene 	3–20 (10)	1273–1673 (1573)	0–40 (30)	2–10 (2)

<sup>a</sup> Standard conditions are in brackets; total pressure is around 1.05 atm; Ar is the balance gas.

### 2.2. Evaluation of experimental data

The carbon conversion degree,  $X_c$ , of model compound  $j$  with carbon number  $N_{C,j}$  and hydrogen number  $N_{H,j}$  is the ratio of one carbon gas product (CO, CO<sub>2</sub>, CH<sub>4</sub>) to model compound  $j$ , which describes the amount of carbon in model compound converted to one carbon gas:

$$X_c(\%) = \frac{n_{CO,out} + n_{CO_2,out} + n_{CH_4,out}}{n_{j,in} \cdot N_{C,j}} \times 100 \quad (7)$$

H<sub>2</sub> yield is defined as the percentage of the stoichiometric potential, which describes the amount of hydrogen and carbon in model compound converted to hydrogen gas:

$$Y_{H_2}(\%) = \frac{n_{H_2,out}}{n_{j,in} \cdot (2N_{C,j} + N_{H,j}/2)} \quad (8)$$

The reaction rate  $r_j$  of each model compound can be described by following equation:

$$r_j = k_j \cdot c_j^m \cdot c_{H_2O}^n \quad (9)$$

$$k_j = A_j \cdot \exp\left(-\frac{E_{A,j}}{RT_R}\right) \quad (10)$$

The residence time  $t$  can be calculated by volume or length divided by gas flow rate  $Q$  or gas velocity  $v$  as:

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