



Formation behaviors of gas and liquid products during the two-stage hydrogasification of low-rank coal



Jie Zhang, Xuantao Wu, Yangyang Qian, Jie Wang*

Department of Chemical Engineering for Energy, Key Laboratory of Coal Gasification and Energy Chemical Engineering of Ministry of Education, East China University of Science and Technology, 130 # Meilong Road, Shanghai 200237, PR China

HIGHLIGHTS

- The study is focused on the effects of post hydrocracking on the gas and liquid formations.
- The intermingled effects of temperature, pressure and vapor residence time are separated.
- The yields of water by hydrogasification of various coal and biomass samples are determined.
- CH₄ and benzene substantially increase in yield with aggravating hydrocracking.
- CO₂, toluene, xylene, phenol decrease in yield with aggravating hydrocracking.

ARTICLE INFO

Article history:

Received 15 December 2014
 Received in revised form 5 February 2015
 Accepted 9 February 2015
 Available online 19 February 2015

Keywords:

Coal hydrogasification
 Two-stage reactor
 Methane
 Benzene
 Water formation

ABSTRACT

The hydrogasification of a low-rank coal was carried out in a two-stage fixed bed reactor with a focus on the separate effects of hydrocracking temperature, hydrogen pressure and vapor residence time on the formations of gaseous and liquid products, particularly the oxygen-containing products. While the yields of CH₄ increased with the increase of hydrocracking temperature (500–750 °C) and hydrogen pressure (0.1–5.0 MPa), the yields of CO₂ showed a downtrend. Both the yield of CH₄ and that of CO₂ changed insignificantly with the vapor residence time (20–100 s). The yield of CO varied somewhat complicatedly with the hydrocracking conditions. The gas generation was influenced not only by the hydrocracking of volatile matter but also by the gas-phase reactions. The increased severity of hydrocracking substantially enhanced the yield of BTX (benzene, toluene and xylene), even with the strengthened decomposition of toluene and xylene. Phenol was hydrocracked to an increased extent, with increasing hydrocracking temperature and hydrogen pressure, whereas naphthalene was barely hydrocracked till the hydrogen pressure was elevated to 5.0 MPa. The pressurized hydrogen intensified the hydrogenation of oxygen atoms in coal forming more water. The yields of water arising from the hydrogenation reactions were found to have a good correlation with the oxygen contents in various coal and biomass samples.

© 2015 Elsevier Ltd. All rights reserved.

1. Introduction

The coal-to-SNG (substitute or synthetic natural gas) technology has found in application for a period of history. Recently, it attracts a renewed interest since a huge demand for SNG arises worldwide, especially in developing countries [1–6]. Coal hydrogasification or hydrolysis is known to be a direct and effective way to produce a methane-rich gas, chemical products such as BTX (benzene, toluene and xylene), and a cleaner char. However, no coal hydrogasification process has commercially put into production so far. Kopyscinski et al. [1] made an updated review

on several early and recent coal-to-SNG processes which are under development of various stages. In China, increasingly pressing energy and environmental problems has driven the researches on coal hydrogasification. A pilot-scale coal hydrogasification process is being developed in ENN Group Co., Ltd., which is fund by a national key R&D project. In the interim of this research plan, an entrain refraction flow reactor with the handling capacity of 10 t/d coal has been built and operated successfully in 2014 year.

Coal hydrogasification has been extensively studied in the past decades [7–23]. The coal hydrogasification involves in a number of hydrogenation reactions, which can broadly be divided into two classes. One is a fast hydrocracking reaction of volatile matter; another is a slow reaction between char and hydrogen. The reactor types used in laboratories include autoclave [10], Curie-point

* Corresponding author. Tel./fax: +86 21 64252462 (office).
 E-mail address: jwang2006@ecust.edu.cn (J. Wang).

pyrolyzer [12], entrained flow reactor [18,21], fixed bed or packed reactor [15,22], fluidized bed reactor [17], free-fall reactor [13,19], hot rod reactor [14], wire mesh reactor [7,14], and thermo-gravimetric balance [9,23]. Among them, fluidized bed reactor, free-fall reactor and entrained flow reactor are usually equipped with a continuous feeding system. These reactors furnish a rapid heating rate and a longer residence time of volatile matter moving through a hot zone, while the residence time of char in the hot zone varies from case to case. Meanwhile, autoclave, fixed bed reactor and thermo-gravimetric balance are characterized as the captive sample technique. These reactors has a common feature of slow heating and prolonged char stay in the hot zone, so that they are advantageous to get the kinetic profiles of gas release or weight loss during heat-up and the reactions of char with reactive gas in an isothermal stage. Curie-point pyrolyzer, hot rod reactor and wire mesh reactor can be also classified into the captive sample technique. However, these reactors serve as a fast or flash heating apparatus with a shorter residence time of volatile matter in the hot zone. Consequently, these reactors are suitable to shed light on the behaviors of primary-stage hydrogasification or devolatilization.

In a previous work, we used a one-stage fixed bed reactor to investigate the hydrogasification of three low-rank coals [24]. Unfortunately, this reactor is less useful to clarify the post reactions of volatile matter with different conditions because the volatile matter is allowed to leave from the hot zone soon. In the present study, a two-stage fixed bed reactor is employed to investigate coal hydrogasification. Interest is taken in the post reactions of nascent volatile matter and their act on the product formations. We believe that this information would help in design and operation whatever reactors and processes are considered in practice.

The two-stage reactor has been used as a mean of investigating coal hydrogasification [25–30]. An early work by Finn et al. [25] demonstrated that an increase in the vapor cracking temperature from 500 °C to 950 °C enhanced the yield of methane above 627 °C and resulted in a broad summit of the benzene yield under an extremely high hydrogen pressure of 15 MPa. Cyprés and Furfari [26] examined the influence of post-cracking temperature (600–900 °C) on the yields of products under 1 MPa, and found that an increase in the post-cracking temperature augmented the yield of gas, mainly methane, at the expense of oil yield, but the oil was rich in BTX. In a later study, they reported that elevation in the hydrogen pressure from 1 MPa to 5 MPa promoted the formation of BTX at the post-cracking temperature of 700 °C, whereas the yield of PCX (phenol, cresol and xylenol) was not affected [27]. Later researchers extended to study the catalytic hydrocracking of volatile matter using the two-stage reactor [28–30].

However, few studies have had an insight into the influences of hydrocracking conditions on the formations of oxygen-containing gas and liquid products such as carbon oxides and water. For the oxygen-enriched feedstock such as low-rank coal, the oxygen-involved reactions would have a large impact on the yields and slates of gas and liquid products as well as the hydrogen consumption.

Therefore, it is worthwhile studying on this area. Moreover, although the effects of temperature, pressure and residence time on the yields of methane and BTX are widely documented, insufficient attention is paid to their intermingled effects on the hydrocracking reactions. The present work is intended to make some new observations on the above points.

2. Experimental

2.1. Coal and biomass samples

A Chinese low-rank coal (WJT coal) from Wangjiata district, Inner Mongolia, was used throughout this study. The coal was crushed and pulverized to the particles with diameter sizes of less than 0.15 mm. The proximate and ultimate analyses of WJT are shown in Table 1, where the properties of three other coals (YX lignite, FG subbituminous coal and HB bituminous coal) and two biomass samples (pine wood and rice husk) are also listed. The latter five samples were only used to show the relationship between the water yield and the oxygen content in coal.

2.2. Hydrogasification apparatus and procedures

The schematic diagram of the pressurized reactor system was shown elsewhere [24]. In this work, the electric furnace was remodeled to heat the vertical tubular reactor by two separate temperature controllers. Fig. 1 illustrates the typical longitudinal temperature profiles with the heating time inside the reactor. In this case, the upper zone of reactor was preheated and held at 700 °C, and the sample packed in the lower zone of reactor was

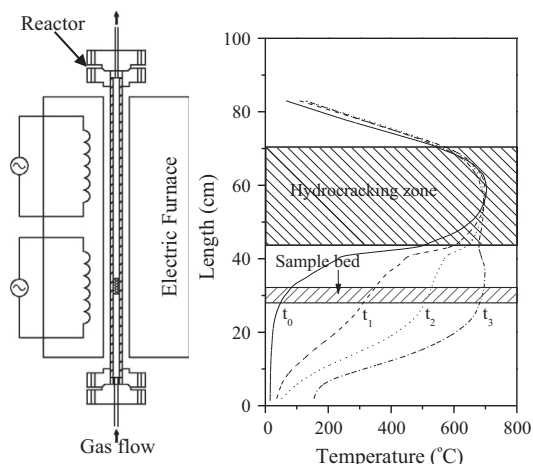


Fig. 1. Schematic diagram of the reactor used for two-stage coal hydrogasification and the change of longitudinal temperature profile within the reactor with the heating time (t_i). —, $t_0 = 0$; - -, $t_1 = 26$ min; ···, $t_2 = 37$ min; - · -, $t_3 = 50$ min.

Table 1
Proximate and ultimate analyses of the coal and biomass samples.

	Proximate analysis (wt.%, ad basis)				Ultimate analysis (wt.%, daf basis)				
	Moisture	Ash	Volatile	Fixed carbon	C	H	N	S	O ^a
Pine wood	14.14	0.52	72.32	13.02	49.96	5.78	0.10	0.00	44.16
Rice husk	1.55	9.86	72.58	16.01	51.20	6.17	0.36	0.10	42.17
YX coal	11.44	22.22	38.60	27.74	65.20	5.90	1.25	0.15	27.50
WJT coal	2.00	10.82	30.00	57.18	81.41	4.44	0.99	0.79	12.37
FG coal	3.15	8.88	30.07	57.90	81.05	5.24	1.17	0.29	12.25
HB coal	1.40	10.74	14.93	72.93	89.17	4.07	1.30	0.32	5.14

^a By difference.

Download English Version:

<https://daneshyari.com/en/article/205704>

Download Persian Version:

<https://daneshyari.com/article/205704>

[Daneshyari.com](https://daneshyari.com)