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Full Length Article

Ex-situ experimental simulation of hard coal underground gasification at elevated pressure

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HIGHLIGHTS

- An ex-situ experiment of coal gasification under pressurized regime was performed.
- An analysis of pressure influence on gasification process was conducted.
- A process gas efficiency, composition and calorific value were calculated.
- A significant influence of gasification pressure on process gas composition was observed.

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ABSTRACT

The experiment of ex-situ hard coal gasification under the 0.5 MPa pressure was carried out by using oxygen, air and oxygen enriched air as gasification reagents. The main process objectives were to investigate the influence of increased gasification pressure and type of gasification reagents on the gasification efficiency. The results of 168 h experiment demonstrated that the most advantageous gasification conditions were obtained with pure oxygen and then with oxygen enriched air. The increase in the content of combustible gas components was proportional to the oxygen content in the gasification medium. It was proved that for each gasification stage there is an optimal flow rate of gasification medium which allows producing gas with the highest energy efficiency. The comparison between the process gas composition at elevated pressure UCG with the literature data on atmospheric pressure UCG operations revealed that under pressurized conditions the methane and carbon dioxide contents were significantly higher contrary to lower concentration of hydrogen and carbon monoxide.

1. Introduction

Underground coal gasification (UCG) is a technology that can be applied to convert the coal resources into synthetic gas. The process involves the injection of oxygen or air into an ignited coal seam and conversion of coal in situ to a combustible gas that can be used for energy production or as a chemical feedstock. UCG can be applied to deep coal seams that are not economical for mining.

The UCG process prediction is crucial to correctly interpret pilot and field data, bring on to a better understanding and design of the gasification process.

The experimental data and research based on UCG trials as well as mathematical models and numerical simulations, give better insights to the process and provides information about gasification conditions. There are comprehensive literature studies on the geological conditions and location of UCG reactors, contamination migration and UCG process progress as well as UCG experiments [1–4].

One of the most important factors affecting the productivity and composition of gaseous products of underground coal gasification is the process pressure. In the available experimental results and studies related to underground coal gasification is little experimental data for relatively high pressure processes [5]. Some of numerical models considered coal gasification up to the pressure of 0.5 MPa [6–9]. Recent UCG gasification trial in Alberta (Canada) was conducted under 12 MPa operational pressure [10]. The operational conditions of UCG pilot plant in Alberta were determined by the great coal seam depth, i.e. 1400 m.

The role of elevated pressure in the UCG process is usually considered in a context of its influence on gasification kinetics and gasification equilibrium.

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The chemical reactions occurring during underground coal gasification can be described by the following equations:

$$C_{(s)} + O_{2(g)} \rightarrow CO_{2(g)} \quad \Delta H = -394.9 \text{ MJ/kmol} \tag{1}$$

$$2C_{(s)} + O_{2(g)} \rightarrow 2CO_{(g)} \quad \Delta H = -226.0 \text{ MJ /kmol} \tag{2}$$

$$2CO_{(g)} + O_{2(g)} \rightarrow 2CO_{2(g)} \quad \Delta H = -563.8 \text{ MJ/kmol} \tag{3}$$

$$C_{(s)} + CO_{2(g)} \rightarrow 2CO_{(g)} \quad \Delta H = +168.9 \text{ MJ/kmol} \tag{4}$$

$$C_{(s)} + H_2O_{(g)} \rightarrow CO_{(g)} + H_{2(g)} \quad \Delta H = +135.7 \text{ MJ/kmol} \tag{5}$$

$$C_{(s)} + 2H_{2(g)} \rightarrow CH_{4(g)} \quad \Delta H = -87.5 \text{ MJ/kmol} \tag{6}$$

$$CO_{(g)} + H_2O_{(g)} \rightarrow CO_{2(g)} + H_{2(g)} \quad \Delta H = -33.2 \text{ MJ/kmol} \tag{7}$$

$$CO_{(g)} + 3H_{2(g)} \rightarrow CH_{4(g)} + H_2O_{(g)} \quad \Delta H = -206.2 \text{ MJ/kmol} \tag{8}$$

$$CO_{2(g)} + 4H_{2(g)} \rightarrow CH_{4(g)} + 2H_2O_{(g)} \quad \Delta H = -165.0 \text{ MJ/kmol} \tag{9}$$

Significance of subscripts: (s) – solid phase, (g) – gas phase.

The main source of heat energy necessary for the endothermic gasification process are exothermic reactions of coal combustion (1–3). According to thermodynamic data [11] all equilibrium constants in the reactions (1–3) decrease with temperature hence, these reactions are favoured at low temperature.

The effect of pressure on the reactions described in Eqs. (1–3) is more difficult to predict because of the homo and heterogeneous reactions, which influence each other. Reaction in Eq. (1) is favoured with increasing of oxygen partial pressure at a fixed total pressure (increase in the oxygen diffusion rates). As total pressure increases, the reaction rates for Eqs. (2) and (3) increase initially and then decrease [12,13].

The Boudouard reaction, which is Eq. (4), is the reaction where CO_2 reacts with carbon to produce CO. This reaction is reversible and endothermic. Therefore, higher temperatures favour the production of carbon monoxide. Lower pressures promote the production of CO, while higher pressures favour the production of CO_2, thereby providing some control over the syngas composition. The reaction temperature should be at least 700 °C (under ambient pressure) [14–16].

The reversible and endothermic water gas reaction (5) produces both carbon monoxide and hydrogen, thus it is one of the most important gasification reactions. This reaction required thermal energy and is favoured by high temperature and low pressure. This reaction is not so heavily affected by the temperature as the Boudouard reaction. Above 800 °C the equilibrium concentrations of gases (steam, carbon monoxide and hydrogen) do not change significantly [14,15].

The hydrogenation reaction (6) contributes to methane content in the UCG product. Methane has higher heating value than carbon monoxide and hydrogen so it is a desired product in the gasification process. The reaction (6) is exothermic and favoured by low temperature and high pressure [15,17]. The influence of elevated process pressures on the methane yields is not as pronounced as lower process temperatures. The reaction is often accelerated by a catalyst (coal ash and metallic oxides).

Reaction in Eq. (7) called shift conversion or water-gas shift reaction is reversible, mildly exothermic that has the tendency to increase the amount of hydrogen compared to carbon monoxide in the product gas [14,17]. The direction of the reaction strongly depends on the conditions in the reactor. If the temperature is high enough, the reverse reaction takes place and the amount of carbon monoxide increases in the expense of hydrogen. Pressure does not affect on the hydrogen yield through this reaction.

Methane can also be produced [18,19] by the two reversible methanation reactions (8) and (9). For these reactions, the ratio of hydrogen to carbon monoxide/dioxide should be at least as 3:1 or 4:1. Both reactions are exothermic and they are favoured by low temperature and high pressure. For these reactions, the catalyst is needed (ruthenium, cobalt, nickel and iron). The methane obtained in methanation retains the high heat of combustion and the exothermic release of this heat reduces the quantity of additional heat required to gasify the coal. The presented data show that methane production (6,8,9) is favoured by high pressures. This is a result of the hydrogenation and methanation reactions, which converts C, CO or CO_2 and H_2 into CH_4 at high pressures, leading to a reduction in the number of molecules in the reacting system [20].

The presented dependences shows, that pressure influence for the process gas yield and its composition is more complex because during underground gasification many factors influence each other.

The UCG process has a zonal character. The following zones may be distinguished in the underground cavity: oxidation, reduction, pyrolysis and drying [1,21–23]. During the gasification process in the UCG cavity, while pressure is largely constant throughout the reactor, the temperature changes from the oxidation to reduction zones, making the modeling of the cavity more difficult.

There are some studies on the kinetic rates of the coal combustion measured at 0.1–0.7 MPa [24,25] and the effect of pressure (up to 1.5 MPa) on the oxidation of very fine bituminous coal at elevated pressure [25,26]. From these studies it can be concluded that the surface reaction rates increase with pressure up to a certain level and then decrease or become constant. The other studies describe the experiments carried out on Australian coal [27] in order to measure the reaction rates with oxygen, carbon dioxide, and water at increased pressures (up to 3 MPa). The results demonstrate that the pressure increased the rate of the coal reactions.

The experiments of reaction in the German coal with carbon dioxide and hydrogen [28] demonstrated that the reaction rates increase in the 1.5–2.0 MPa range. The similar experiments [29] with coal reactivity under the pressure of 0.1–3.0 MPa of CO_2 and, separately H_2O, demonstrated that reaction rates increase with increasing pressure, but the rate of increase is reduced at higher pressures or may become constant. There are also limited data for coal pyrolysis process at high pressures. The studies on the pressure effect on the pyrolysis demonstrated [30] that the pressure decreases the extent of pyrolysis. The studies of the effect of pressure on the gas evolution in pyrolysis [31] showed weak pressure effect on the gaseous products. Both studies conclude that the effect of pressure on coal pyrolysis is not significant, compared to its effect on the chemical reactions.

According to studies [13,21,22] the increase of the hydrostatic pressure with the coal seam depth forces the use of higher pressures during the gasification process. Deeper coals offer the opportunity to achieve much higher pressures in the reactor resulting in higher methane content and resultant higher heating value of gas [23].

The present paper describes the results of hard coal gasification under higher pressure (0.5 MPa). The experiment was conducted in ex-situ reactor specially designed to simulate the underground conditions at elevated pressures. The obtained results we compared with other experimental and literature data.

2. Experimental

2.1. Description of the installation

The study was conducted in an experimental high pressure installation designed to simulate the process of UCG in the surface