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Kinetics of steam gasification of bituminous coals in terms of their use for underground coal gasification

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ABSTRACT

The kinetics of steam gasification was examined for bituminous coals of a low coal rank. The examined coals can be the raw material for underground coal gasification. Measurements were carried out under isothermal conditions at a high pressure of 4 MPa and temperatures of 800, 900, 950, and 1000 °C. Yields of gasification products such as carbon monoxide and carbon dioxide, hydrogen and methane were calculated based on the kinetic curves of formation reactions of these products. Also carbon conversion degrees are presented. Moreover, calculations were made of the kinetic parameters of carbon monoxide and hydrogen formation reaction in the coal gasification process. The parameters obtained during the examinations enable a preliminary assessment of coal for the process of underground coal gasification.

1. Introduction

Underground coal gasification (UCG) is an alternative technical option for using coal without the necessity of excavating it [1]. This is an environmentally friendly way of utilizing coal [2], technically feasible, and its application is also economically justifiable [3,4]. Coal conversion directly in the seam into a suitable fuel gas occurs with the aid of gasification agents like air, oxygen, steam or their mixture. The most important components of the gas are: carbon monoxide, hydrogen, methane, and carbon dioxide, and its calorific value ranges from 4.1 to 10.7 MJ/Nm^3. The shares of these components depend on many factors, e.g. the composition of the gasification agent mixture, coal properties, as well as geological and hydrological characteristics of the coal seam [5]. The optimum operating conditions should be used in order to achieve desired conversion coal to syngas [6]. UCG is inherently an unsteady process since a number of parameters, such as the growth of the cavity, inherent variation in the properties of the coal along the seam, quantity of water influx, ash layer build-up, affect the rates of the homogeneous and heterogeneous reactions occurring therein [7–9]. The gas may be used for energy production or the synthesis of chemicals, liquid fuels, or other gaseous fuels. The underground coal gasification may also be successfully used to produce hydrogen from coal [10].

In a coal seam, where the process of underground coal gasification is conducted, three zones may be distinguished: oxidation zone, gasification zone and pyrolysis zone. In the oxidation zone, exothermic combustion reactions occur between the oxygen contained in the gasification mixture and carbon contained in the coal seam (Eqs. (1)–(3)). The resulting heat causes the warming of the seam up to a high temperature.

$$C + O_2 \rightarrow CO_2 \qquad \Delta H = -393,51 \ \ kJ/mol \tag{1}$$

$$C + 0,5O_2 \rightarrow CO \qquad \Delta H = -110,53 \ \ kJ/mol \tag{2}$$

$$CO + 0,5O_2 \rightarrow CO_2 \quad \Delta H = -282,98 \ kJ/mol \tag{3}$$

In the gasification zone endothermic reactions (Eqs. (4)–(5)) occur between steam and carbon dioxide, which results in the formation of hydrogen and carbon monoxide.

$$C + H_2O \leftrightarrow H_2 + CO \qquad \Delta H = +131,28 \ kJ/mol \tag{4}$$

$$C + CO_2 \leftrightarrow 2CO \qquad \Delta H = +172,45 \ \ kJ/mol \tag{5}$$

Additionally, due to the catalytic influence of ash [11] and the high pressure connected with the depth at which the process occurs, apart from the above-mentioned reactions, the methanation reaction also takes place (6).

$$C + 2H_2 \leftrightarrow CH_4 \qquad \Delta H = -74,81 \ \ kJ/mol \tag{6}$$

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In the pyrolysis zone, thermal destruction of coal occurs, accompanied by the release of such components as H_2O, CO_2, CO, C_2H_6, CH_4, H_2 and tar.

Also products of gasification can react especially CO according to the equation:

$$CO + H_2O \leftrightarrow CO_2 + H_2 \qquad \Delta H = -41,2 \ \ kJ/kmol \tag{7}$$

As it can be concluded from the above-mentioned reactions, the most important products, i.e. carbon monoxide (CO) and hydrogen (H_2) are formed in the coal gasification zone as a result of the occurrence of the water gas reaction (Eq. (4)) and the Boudouard reaction ((Eq. (5)) there. Therefore, it is very important to learn about this stage of the process.

The complexity of the gasification process results in the fact that it is still the subject of numerous examinations and analyses [12]. The impact of particular factors on the gasification process, and, especially, the carbon conversion degree as well as the yields of gaseous products of the process are assessed most frequently with the aid of thermovolumetric and thermogravimetric methods. In the vast majority of cases, the examinations of gasification kinetics are conducted for coal chars, with the temperature at which they are obtained being very important. The impact of temperature and pressure at which the process is conducted on the rate of coal and lignite gasification as well as the yields of gaseous product still remains a valid problem [13-16]. The presented examinations confirm that temperature is one of the more important factors influencing the carbon conversion degree and coal char gasification rate. Also, the process pressure has an effect on both, the course of the coal pyrolysis stage and coal char gasification. As far as the pyrolysis stage is concerned, pressure influences it in two ways: firstly, it affects the amount of the obtained coal char and, secondly, its structure. The growth of pressure at which the pyrolysis process is conducted facilitates a reduction in the loss of coal mass, that is, the formation of greater amounts of coal char, and the most significant changes are observed within the pressure range of 0.01–1 MPa [17]. The examinations also focus on establishing the impact of the composition and structure of the organic and mineral matter on the gasification rate [18,19]. Investigations were also conducted aiming at determining the activation energy, during which it was found that the energy does not depend on grain size distribution of coal sample and it increases along with the coal rank [20–22]. The results of examinations contained in the work [15] have confirmed the impact of partial pressure of steam on gasification rate. As regards gasification with steam, the H_2O/C ratio is also important and according to [23], together with the increase of this parameter, the total gas yield as well as the CO and H_2 yields grow, while the CO_2 concentration and the carbon conversion degree decrease. In turn, the presence of gases released during pyrolysis, as well as hydrogen, in the course of gasification with steam results in slowing down the process [20].

Unfortunately, due to the specificity of underground coal gasification, examinations of gasification kinetics inside the coal seam are practically impossible. Matching a proper coal to the gasification process is very important [24]. At the stage of assessment of particular coals for underground coal gasification, laboratory methods can be applied. In this work, examinations focused on the formation kinetics of gaseous products of coal gasification with steam under conditions similar to those of the coal seam. For the purposes of the examinations, a unique laboratory equipment was used, which enables examining the kinetics of gasification with steam under a high pressure and for coal particles of several millimeters. The investigations were carried out for coals which may be a potential raw material for underground coal gasification. A rarely used approach during gasification kinetics investigations, i.e. examining coals and not the chars obtained from them, enables getting practical information about phenomena occurring in a coal seam, among others, concerning the composition of gaseous products released at the initial stage of the process, when the reactions of pyrolysis dominate. The kinetic parameters of the examined coals were determined; moreover, on the basis of the course of the kinetic curves, the yields of the most important gasification products were calculated, i.e. hydrogen, carbon monoxide, carbon dioxide as well as methane, and the curves of carbon conversion degree were produced. On the basis of the results of the examinations a preliminary assessment of coals can be made, among others, for the process of underground coal gasification [25].

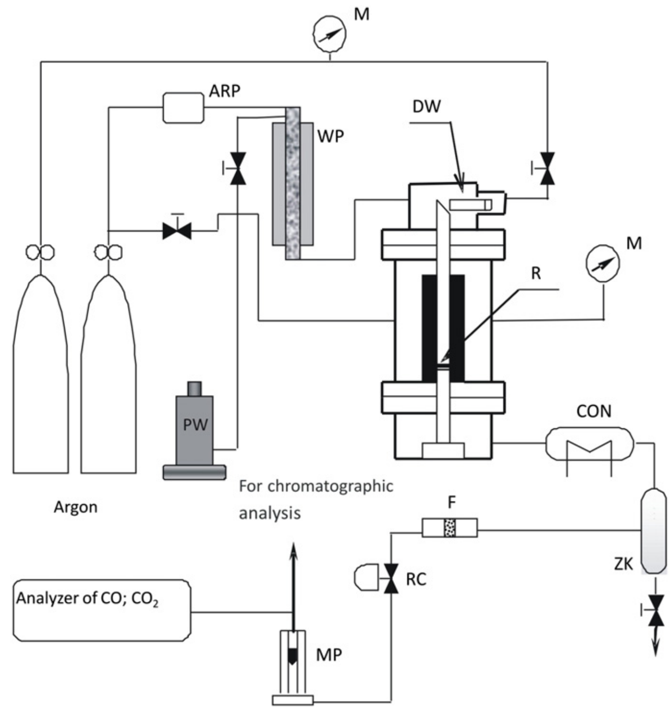

Fig. 1. The laboratory equipment for kinetic examinations of coal gasification: R — reactor; WP — steam generator; PW — water pump; CON — condenser; ZK — tar separator; MP — rotameter; DW — coal feeder; ARP — mass flowmeter; M — pressure gauge; F — gas filter; and RC — backpressure regulator.

Table 1
Characteristics of examined coals.

Parameter	Ziemowit coal	Bobrek coal	Bogdanka coal	Wieczorek coal
Proximate analysis (%)				
Moisture — M^a	6.6	4.0	2.2	3.3
Ash — A^a	5.6	2.6	4.3	13.0
Volatile matter — VM^{daf}	40.8	37.5	41.3	38.2
Higher heating value — HHV (MJ/kg)	28.3	34.6	32.8	29.6
Ultimate analysis (%)				
C^{daf}	79.3	87.1	83.3	83.8
H^{daf}	5.3	5.7	5.7	5.8
S_t^{daf}	0.78	1.03	1.04	0.31
N^{daf}	1.2	1.3	1.5	1.4
O^{daf} (difference)	13.4	4.9	8.5	8.7
O/C ratio	0.17	0.06	0.10	0.10
Ash composition (%)				
Fe_2O_3	9.3	7.1	8.4	9.3
CaO	6.5	7.1	4.8	4.9
MgO	4.7	5.5	0.4	3.5
Na_2O	4.8	0.6	0.6	1.1
K_2O	0.6	2.8	0.3	1.8
SiO_2	28.7	43.8	32.2	39.3
Al_2O_3	28.8	23.9	31.6	15.5
Alkali index AI (−)	2.52	0.89	0.98	4.89