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Conversion of brown coal continuously supplied into the reactor as coal–water slurry in a supercritical water and water–oxygen mixture

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ABSTRACT

The results of a two-stage conversion of brown coal (gross-formula $CH_{0.96}N_{0.01}S_{0.002}O_{0.31}$) in supercritical water (SCW) under isobaric conditions (30 MPa) in the vertical tubular reactor of an original configuration are presented. The first stage provides for a continuous supply of coal–water slurry (CWS) into the reactor and discharging of the products with SCW into the demountable samplers. The CWS composition was as follows (weight portions): coal—100, water—125, and starch—2.25. It was found out that when increasing the temperature of coal particles up to 600 °C during their falling down along the reactor axis, the yields of volatile and liquid products were 25.4 and 27.5%, respectively, relative to the weight of organic matter supplied into the reactor. At the second stage of conversion, the SCW/O_2 fluid was pumped through a layer of coal particles accumulated at the reactor bottom during the first stage. It was shown that as a result of heat emission during partial oxidation of coal residue, the autothermal conditions for conversion were realized. The yields of volatile combustible and liquid products at conversion in the SCW/O_2 fluid were 25.1 and 3.4 wt.%, respectively. The increment of hydrogen in the conversion products caused by water decomposition was equal to 20% relative to the weight of hydrogen in initial coal.

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1. Introduction

Continuous increase in energy consumption contributes to the development of new energy-efficient and environmentally friendly methods for the petrochemical feedstock and energy resources production. Conversion of different low-grade fuels in supercritical water (SCW) can be the basis for these technologies [1,2]. As supercritical water ($T > 374$ °C, $P > 22.1$ MPa) has low viscosity and a low dielectric constant along with high density, it becomes an effective solvent for organic substances and gases [3–5]. When temperature rises, H_2O molecules participate in the redox reactions with fuel's components more actively.

Despite vast stocks and appreciably widespread occurrence of brown coals, their usage in fuel power industry is negligible due to their low heating value (20–25 MJ/kg) and high moisture content (up to 60 wt.%) caused by high oxygen content (up to 30 wt.%) in coal organic matter (COM) [6]. The treatment of brown coals with sub- and supercritical water provides for removal the major portion of chemically bound oxygen as CO_2 and results in coal's wetting ability decrease and heating value increase [7,8]. Moreover, brown coals, due to their appreciably high H/C atomic ratio (0.8–1.1), are considered as the starting material to obtain liquid and gaseous fuels [6,9,10]. By now, SCW conversion of brown coals has been studied in autoclave [11,12] and flow [13–15] modes, at continuous supply of coal–water slurry (CWS) [16–20], and in a cyclic pressurization-depressurization mode [21]. Besides, the problems dealing with coal combustion directly in SCW to obtain a working substance of electro-generating devices have been actively discussed [22–24]. The use of SCW for the coal conversion is also associated with the problems of effective use of watered fuels whose fine-grained grades are hardly subjected to dehydration.

To provide for a continuous supply of CWS into the reactor with SCW, it is necessary to obtain a stable fluid CWS. This is ensured by the use of stabilizing additives, coals with different degrees of crushing and CWS with proper coal concentration. A supply of CWS (16 wt.% coal + 1.5 wt.% sodium carboxymethyl cellulose (CMC) as an additive to prevent the precipitation + 1 wt.% K_2CO_3 as a catalyst) into the reactor is successfully performed followed by CWS continuous gasification (750 °C, 25 MPa) in [17]. The authors [17] revealed that when the concentration of coal increased to 20 wt.%, only part

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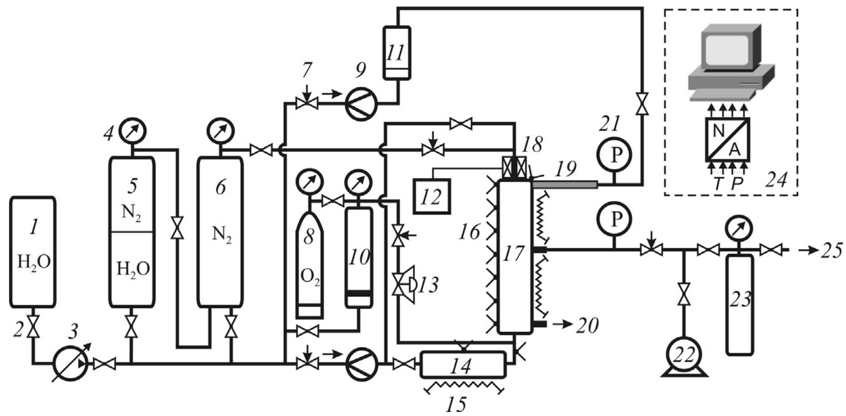

Fig. 1. Schematic diagram of the experimental setup: (*1*) vessel with distilled water; (*2*) valve; (*3*) high-pressure plunger pump; (*4*) strain gauge; (*5*) damping vessel; (*6*) high-pressure vessel with N_2; (*7*) regulating valve; (*8*) balloon with O_2; (*9*) differential pressure gauge; (*10*) cylinder with floating piston; (*11*) vessel with castor oil; (*12*) electronic control system for electromagnet; (*13*) O_2 mass-flow controller; (*14*) thermostat; (*15*) resistance heaters; (*16*) thermocouple; (*17*) reactor; (*18*) electromagnetic system for coal knife motion; (*19*) fuel cylinder; (*20*) output canal for solid conversion residue; (*21*) membrane pressure gages; (*22*) first-stage vacuum pump; (*23*) demountable sampler for collecting products; (*24*) data-collecting system; (*25*) mass spectrometer vacuum unit.

of CWS could be pumped into the reactor and the experimental system could not be operated continuously. In [18] 24 wt.% coal–water slurry (2 wt.% CMC + 1 wt.% K_2CO_3) was continuously transported and stably gasified without plugging problems. It was found out that when the concentration of CWS increased, the hydrogen fraction decreased while the methane fraction increased [18]. In the study [18] on coal SCW gasification (580 °C, 25 MPa), it was shown that the coal particle size (fractions < 149, <105, and <74 μm) had no significant effect on the gaseous product concentration. In the study [19] on coal SCW gasification (600 °C, 25 MPa), it was also stated that the yields of H_2 and CO_2 decreased by 25%, while that of CH_4 increased by 50% with increase in the CWS concentration (the coal particle size <76 μm) from 20 to 50%. The possibility of a continuous supply of CWS (≈50 wt.% coal + 0.8 wt.% NaOH) was described in [16,20] where it was found that a bimodal size distribution of coal particles (40–50 μm—25 wt.% and 200–315 μm—75 wt.%) maximized stability and fluidity of CWS.

In this paper we present a detailed study of brown coal conversion in a two-stage process: at a continuous supply of CWS (44 wt.% coal + 1 wt.% starch) into the reactor and subsequent pumping of the SCW/O_2 fluid through a layer of coal residue; the autothermal conditions for conversion were realized at partial combustion of coal residue in the SCW/O_2 fluid.

2. Experimental

2.1. Reagents and experimental procedure

The object of our investigation was brown coal from the Kansk-Achinsk coal-field (Kaichakskoe deposit). The characteristics of the coal sample (the particle size ≤315 μm) were as follows: moisture content W = 23.0 wt.%; ash content per dry basis A^d = 5.1 wt.%; the gross-formula of coal organic matter $CH_{0.96}N_{0.01}S_{0.002}O_{0.31}$.

Fig. 1 presents the scheme of the experimental setup. The main parts of the setup are made of stainless steel 12Cr18Ni10Ti, an analogue of AISI 321. The reactor (internal diameter 30 mm; length 155 cm) is placed vertically (Fig. 2). SCW and SCW/O_2 fluids were supplied into the reactor through a nipple located at its bottom (*20*, Fig. 2). Distilled water from the vessel (*1*, Fig. 1) under the pressure created by the plunger pump *3* was supplied into the reactor through the thermostat *14*. Oxygen was supplied from the vessel *10* (V = 2 dm^3) under the pressure of water and regulated with the Bronkhorst El-Flow. The unit for fuel supply consists of two removable cylinders (*8*, Fig. 2) with the internal diameter of 25 mm and the length of 60 cm. These cylinders are located on a horizontal plane, symmetrically and perpendicular to the reactor axis. Even dosing of fuel, supplied from the cylinders into the reactor, was performed by means of two knives *6*. The frame with the knives is installed along the reactor axis in the gap between the ends of fuel cylinders. The frame moves reciprocally under the action of electromagnet *2* whose magnetic coil *3* receives electric pulses with

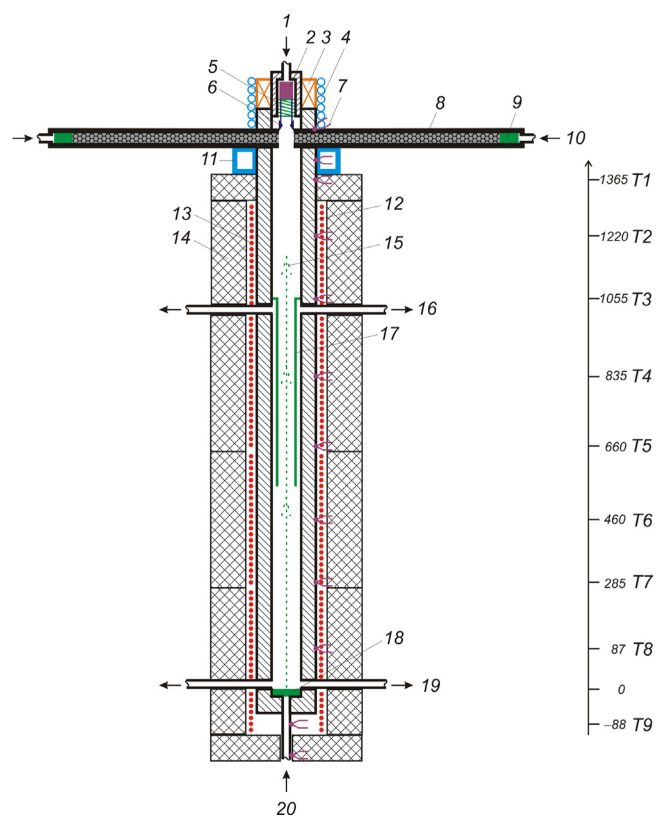

Fig. 2. Reactor's configuration: (*1*) water and/or gas input; (*2*) ferromagnetic core; (*3*) electromagnetic coil; (*4*) copper pipe for water cooling of coil and reactor top; (*5*) spring; (*6*) knife for coal dosing; (*7*) thermocouple; (*8*) fuel cylinder; (*9*) piston; (*10*) hydraulic supply of coal–water slurry; (*11*) water cooling jacket for reactor top; (*12*) reactor's body; (*13*) resistant heaters; (*14*) heat insulation; (*15*) disperser for falling coal; (*16*) reactant output; (*17*) shielding cylinder; (*18*) plate made of porous stainless steel; (*19*) output canal for solid conversion residue; (*20*) oxidizer and SCW input. Thermocouples' Ti (i = 1–9) location (mm) relative to the reactor bottom shown in the vertical axis on the right; Ti (i = 2, 4, 6, 8)—regulating thermocouples.

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