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Obtaining of Gas, Liquid, and Upgraded Solid Fuel from Brown Coals in Supercritical Water

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Abstract—Two new conversion methods of brown coals in water steam and supercritical water (SCW) are proposed and investigated. In the first method, water steam or SCW is supplied periodically into the array of coal particles and then is ejected from the reactor along with dissolved conversion products. The second method includes the continuous supply of water–coal suspension (WCS) into the vertically arranged reactor from above. When using the proposed methods, agglomeration of coal particles is excluded and a high degree of conversion of coal into liquid and gaseous products is provided. Due to the removal of the main mass of oxygen during conversion in the composition of CO_2, the high heating value of fuels obtained from liquid substantially exceeds this characteristic of starting coal. More than half of the sulfur atoms transfer into H_2S during the SCW conversion already at a temperature lower than 450°C.

Keywords: brown coal, supercritical water, upgrading, liquefaction, gasification, desulfurization

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It is known that water at supercritical parameters ($t > 374$°C, $p > 22.1$ MPa) becomes a nonpolar solvent and the source of $H^{\bullet}$ and $HO^{\bullet}$ radicals, which ensures liquefaction and gasification of a considerable organic mass of coals (OMC); oxygen is removed in the composition of CO_2; and the solid residue is enriched with carbon [1–5]. For example, in [4] hydrothermal treatment of brown coal in an autoclave at 350°C and 18 MPa ensures an increase in high heating value from 25.0 to 33.1 MJ/kg only due to a decrease in the oxygen content in the organic mass. Authors of [5], based on the results of analyzing the products of hydrothermal treatment of coal in an autoclave at 150–350°C, concluded that reactions of pyrolysis and hydrolysis of the OMC lead to destruction of ether bonds in it, to detachment of aliphatic substituents, and redistribution of nonvalent interactions. They also found that at t = 250–300°C, due to quenching the radical fragments with radicals $H^{\bullet}$ and $OH^{\bullet}$, the sources of which are the H_2O molecules, the OMC is saturated with hydrogen, which leads to an increase in the yield of liquid hydrocarbons [5].

We proposed and investigated two new conversion methods for brown coals. It was already noted above that the first method [6] includes the periodic input of supercritical water (SCW) into the array of coal particles and ejection of SCW with dissolved conversion products from the reactor in a mode of a stepped increase in temperature. The second conversion method [7] consists in the continuous supply of the water–coal suspension (WCS) into the vertically arranged reactor.

COAL CONVERSION IN A MODE OF PERIODIC FILLING THE REACTOR WITH SCW AND EJECTION OF REACTANTS

As the object of investigations, we selected brown coal of the Yakutsk coal basin (Zhiganskoe deposit) with humidity $W = 8.4\%$, ash content in dry coal A_0^d = 12.5%, and the following elemental composition of the organic mass: C 70.2%, H 4.7%, N 0.8%, S 0.4%, and O 23.9%. The gross formula of the OMC was calculated in a form of atomic ratios of H, N, S, and O to carbon: $CH_{0.80}N_{0.01}S_{0.002}O_{0.25}$.

The experimental order was as follows. A certain amount of coal was poured into a cylindrical sleeve with an inner diameter of 18 mm. The open sleeve end was closed by a partition (Fig. 1) made of porous stainless steel. The sleeve was placed into a vertically arranged reactor with the partition downwards. The partition prevented the carryover of coal from the reactor when discharging the reactants. After sealing the reactor, all installation systems were evacuated.

Table 1 represents conditions of experiments 1 and 2, namely, the input time of steam or SCW into the reactor τ_{in}, soaking time τ_r, ejection time of reactants from reactor τ_{ej}, temperature t, pressure p and density ρ of steam or SCW, and the number of input–ejection cycles N. The passage to a new temperature level was performed by uniform heating of the reactor (2°C/min). Liquid reactants of all ejections, which correspond to the temperature and the pressure, were collected in dismountable samplers. The analysis of gaseous products and evacuation of the collector were