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OBTAINING CARBON NANOMATERIALS IN A PLANT WITH A PLASMA GENERATOR AND A WORKING ZONE OF RECTANGULAR CROSS SECTION

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Results of experimental investigations of the conditions of formation of carbon nanostructures in a plasma-chemical reactor of rectangular cross-section from the products of decomposition of hydrocarbons in a low-temperature plasma are presented. The influence of the additional flow region on the process was determined. Data on the content of structurized carbon in the material obtained and on the yield of the process are presented.

Keywords: *carbon nanomaterials, nanotubes, nanofibres, decomposition of hydrocarbons, plasma, electric discharge, plasma generator.*

Introduction. The increasing demand for carbon nanomaterials has stimulated the development of new methods and plants for obtaining these materials in industrial quantities. In this connection, investigators should find ways of increasing the production of the indicated materials on the existing equipment, e.g., by direct scale-up of the reactor as aresult of its multiprogramming or through modernization of the reactor design.

Formulation of the Problem. The present investigation has been carried out within the framework of works carried out at the Laboratory of Nonequilibrium Processes and the Laboratory of Physics and Chemistry of Combustion of the A. V. Luikov Heat and Mass Transfer Institute of the National Academy of Sciences of Belarus and devoted to development of high-production plants based on a plasma generator for production of carbon nanomaterials. Experience in setting up the work described in [1] has shown that the design problem of increasing the yield of carbon nanomaterials can be solved by increasing their growth surface. In this case, the power of the heaters should be increased, which can be realized with the use of a higher-power plasma generator or several plasma generators of moderate power.

A plant with several heaters should include a collector serving to combine the gas flows from the plasma generators that are then directed to the working zone of the reactor.

The aim of the present work is to determine the influence of the additional flow region (the collector zone) on the process of formation of structurized carbon in the working zone of a reactor with a rectangular cross section.

Experimental Setup. An experimental setup has been developed on the basis of a low-temperature plasma generator with a power of up to 40 kW. Its main units are as follows: a system of power supply for the discharge, a cathode-anode system and a unit for water cooling of this system, a collector for distribution of the plasma flow, and a plasma-chemical reactor with a rectangular cross section of plane-parallel geometry (Figs. 1 and 2). The outside of the plasma-chemical reactor was covered with a heat-insulating layer for maintenance of high temperatures sufficient for formation of carbon nanomaterials on its inner surface (650–800^{o}C). In the working regime of the setup, a mixture of air with hydrocarbon was fed to the plasma flow, as shown in Fig. 1, where it was decomposed. Then the reaction products passed through the holes in the collector and entered the plasma-chemical reactor where they were deposited on the metal walls and formed carbon nanostructures (nanotubes and nanofibres). The following parameters were controlled during the experiments (Fig. 2): 1–5) the temperature at the outer wall of the plasma-chemical reactor, 6) the temperature at the face of the collector, 7) the temperature at the inner wall of the plasma-chemical reactor, and 8) the temperature of the gas at the output. The rates of the gas flows were varied within the following ranges depending on the experimental conditions: 10–15 m^3 for air (including the plasma-forming air) and 5–8 m^3 for methane. In our ex-

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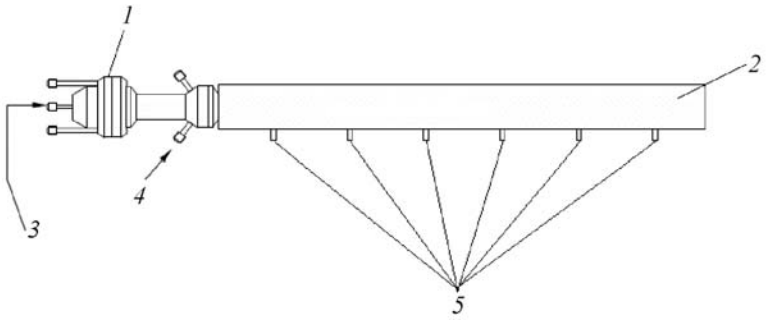

Fig. 1. Diagram of a plasma generator and a collector for distribution of a plasma flow: 1) plasma generator; 2) collector; 3) supply of the plasma-forming gas (air); 4) supply of the working mixture (air + hydrocarbon); 5) holes in the collector leading to the reactor.

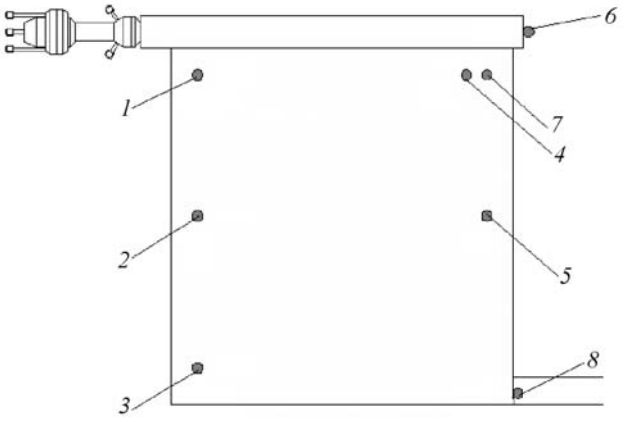

Fig. 2. Diagram of disposition of thermocouples in the plasma-chemical reactor and the collector for distribution of a plasma flow (the numbers of thermocouples correspond to the numbers presented in the figure).

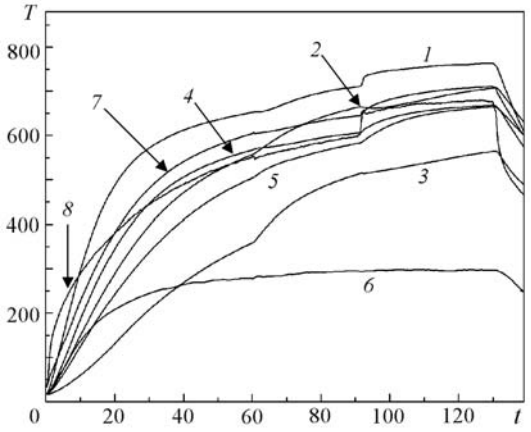

Fig. 3. Typical temperature distributions of the process (the numbers of curves correspond to the numbers of the thermocouples in Fig. 2). T, ^{o}C; t, min.

periments the ratio between the amounts of the reagent and the oxidizer was controlled by the equivalence factor comprised $\gamma \sim$ 4.2–4.3 in the working regime.

Results and Discussion. At the previous stages of the experiments we selected thermal regimes to provide the most optimum conditions for heating of the inner surface of the plasma-chemical reactor to the temperatures at which carbon nanostructures are formed (650–800^{o}C). However, the subsequent experiments have shown that the heating of

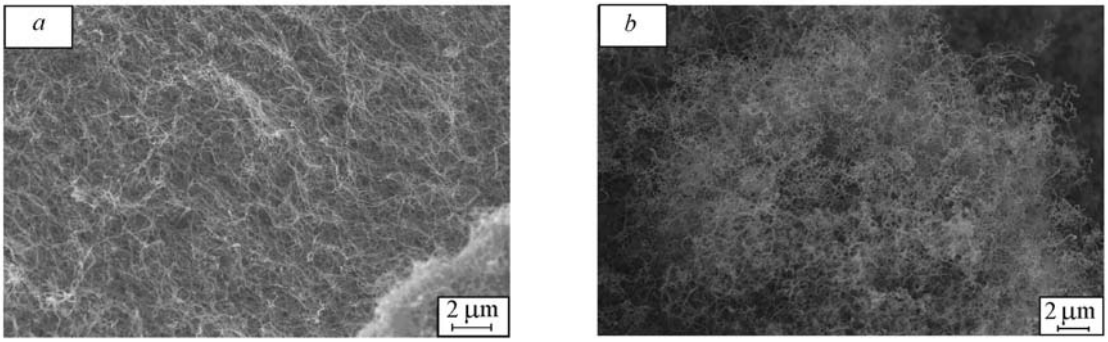

Fig. 4. Microphotographs of the carbon nanomaterials taken from the collector (a) and from the zones positioned near the plasma flow (b).

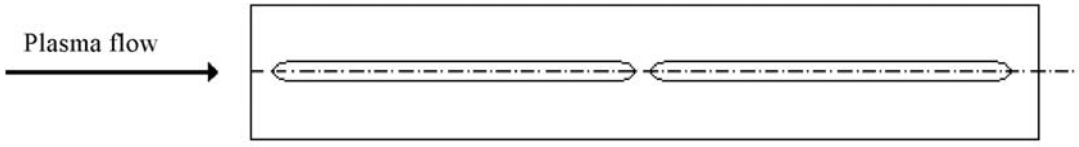

Fig. 5. Diagram of the reconstructed collector (top view to the reactor, width of the slots equal to ~16 mm).

this surface to the indicated temperatures is not a sufficient condition for the growth of nanofibers and nanotubes. Figure 3 presents the typical temperature dependences for experiments with a working regime of duration 40 min. In this case, the electric power of the plasma generator was 33.6 kW. It is seen that the larger part of the reactor had a temperature higher than 600°C; however, the carbon nanomaterial was formed only in certain zones of its walls, near the burners of the collector (i.e., the "torches" of the plasma flow), while the thickness of the deposit layer formed on the inner surface of the collector reached 1.5–2.5 mm. An analysis of the microphotographs made with the use of a scanning electron microscope has shown that the deposit formed in the collector (Fig. 4a) and in the zones positioned near the plasma flow (Fig. 4b) contains carbon nanofibres and nanotubes that have a diameter of 20–100 nm and are different in length and shape; they are intertwined closely together and form cover on the surface of the carbon-black microparticles.

The carbon nanomaterial was not formed on the surface of the reactor, probably because of the small temperature gradient between this surface and the gas flow. Actually, as noted above, the deposit with a high content of carbon nanostructures, accumulated in the collector, was gathered from the wall of the reactor positioned near the plasma generator (near the first burner of the collector). Therefore, the design of the collector was changed such as to provide an increase in the velocity of the gas flowing through the collector–reactor-channel path and, consequently, an increase in the heat exchange between the gas and the wall downstream, i.e., in the reactor channel. The essence of the change in the design of the collector is that, instead of the hollows, two slots were made in it, as shown in Fig. 5.

Further experiments with the reconstructed collector have shown that the carbon nanomaterial was formed not only in the high-temperature zones of the collector near the plasma generator, but also in the plasma-chemical reactor. In this case, the plant produced ~270 g/h of the carbon nanomaterial with a content of carbon nanofibres and nanotubes of ~40%.

Conclusions. In the process of our investigations we have developed a plant for production of carbon nanomaterials with an output of up to 270 g/h; the content of structurized carbon (in the form of nanotubes and nanofibres) was ~40%. These investigations allowed us to broaden the range of change in the experimental parameters determining the conditions of growth of carbon nanomaterials. It has been established that the time of movement of the reaction products from the plasma reactor to the working zone where the carbon nanomaterial is formed negatively influences the processe of formation. This means that, to increase the production of carbon nanomaterials in the designing of plants for obtaining them one must do away with the collector to decrease; the time of passage of the "hot" reaction products from the plasma reactor to the working zone where carbon nanomaterials are formed to a minimum.

NOTATION

t, experimental time, min; T, temperature, ^{o}C; γ, equivalence factor (stoichiometric ratio).

REFERENCE

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