FORMATION OF CARBON NANOMATERIALS UNDER THE CONDITIONS FORMED BY ATMOSPHERIC PRESSURE HIGH-VOLTAGE PLASMA

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The methods of producing carbon nanomaterials (CNM) are diverse [1-3]. The authors of work [4] proposed a new method of synthesizing CNM. It was based on forming carbon nanoobjects from carbon-containing gases processed by atmospheric-pressure high-voltage discharge plasma. The main mechanism of CNM formation under these conditions was taken as the reaction of disproportionation, in which carbon oxide was a reagent.

However, further studies enable one to assume another mechanism of formation of nanostructures, according to which the growth of nanotubes and nanofibers occurs from some surfaces of the reactor working zone.

It is known [5,6] that the pyrolysis of hydrocarbons on a metal catalyst results in forming the carbon structures, whose morphology depends on the reaction conditions. In this case, the mechanism of the CNM growth is such [5]: 1) absorption of hydrocarbon molecules on the metal surface and their catalytic decomposition, followed by the carbon formation; 2) carbon dissolving in metal and diffusion of carbon atoms inside metal; 3) saturation of the near-surface layer of metal with carbon; 4) formation of carbon nanoparticles and a further growth of nanotubes.

Taking into account the above scheme of CNM formation, it is possible to assume that the rate of formation of carbon nanotubes will be determined by temperature and material of a deposition surface, surrounding gas composition, temperature field and configuration of the gas flow behind the discharge zone. This work presents the experimental verification of some of these propositions.

The work was carried out on the detailed in [4] device with a modified reactor, which allowed for the change of a CNM deposition (Fig. 1). The mixture flowrate was 656 l/hr (methane 200 l/hr + air 456 l/hr), discharge power – 420 Wt.





Fig. 1. Scheme of the reactor. 1 – quartz tube; 2 – cathode; 3 – anode; 4 – tube for admission of a gas mixture; 5 – deposition surface (foil); 6 – thermocouple; 7 – heat insulation

Fig. 2. Specific amount of deposited CNM vs. discharge power. The mixture composition at the reactor outlet is: 18%CO+37%H₂ + 42% N₂+ 3% CH₄

The influence of the surface material on the CNM yield was determined in the following way. The deposition surface was made of two semi-cylinders – nickel and molybdenum foil. The specific quantity of deposited CNM from the nickel foil surface m_{Ni} was equal to $m_{Ni} = 16 \cdot 10^{-5}$ g/mm²hr, the material was not observed on the molybdenum foil surface. On the copper foil $m_{Cu} = 0.41 \cdot 10^{-5}$ g/mm²hr.

As mentioned above, the important factor in the process of CNM formation is the temperature of the deposition surface, which determines the rates of hydrocarbon decay on the surface and carbon diffusion in metal. It has been established that under our experimental conditions CNM is not formed at the surface temperature less than 500° C.

In experiments it has been revealed that the presence of the temperature gradient between the gas flow and the deposition surface plays an important role. If the surface temperature is close or equal to the flow temperature, then CNM is not formed on the surface. Probably, this is bound up with thermodiffusion processes that determine the rate of supply of gaseous hydrocarbon to the metal surface.

Thus, the performed experimental studies show that under the conditions of synthesis of carbon nanomaterials in the plasma of the atmospheric-pressure high-voltage discharge the processes of catalytic decomposition of methane occur with great probability on the metal surface. This determines, how the CNM yield depends on the material of the deposition surface, its temperature and the entire configuration of a temperature field in the deposition zone.

It should be noted that under the nonequilibrium conditions of the high-voltage discharge there is a probability to conduct the reaction of disproportionation, but it is in prospect to determine the particular importance of this process in the overall picture.

References

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