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OBTAINING CARBON NANOPARTICLES WITH THE USE OF ELECTRIC-ARC DISCHARGE BETWEEN COAXIAL ELECTRODES

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The structure of a setup and the results of experiments on obtaining carbon nanostructures with the use of a d.c. arc discharge between coaxial graphite electrodes have been presented. The discharge was stabilized by a magnetic field. The condensed carbon has been investigated by the method of atomic-force microscopy.

Introduction. One widespread method of obtaining carbon nanostructures is the electric-arc method, which assumes heating of carbon to the sublimation temperature in the arc spot or in a high-temperature flow in an inert medium, transition of carbon to the gaseous phase, and its subsequent condensation on the cold surfaces of the discharge chamber and the outlet of the setup. Certain conditions for obtaining one carbon nanostructure or another are required in both evaporation of carbon and condensation of it. Since a considerable amount of experience has been accumulated in the field of development of electric-arc heaters of gas [1, 2], we have made an attempt to use it with the aim of creating a device for obtaining carbon nanostructures. The coaxial scheme of organization of an arc discharge for low rates of flow of the working gas enables one to stabilize the discharge, to adjust the position of the discharge zone relative to the electrodes, and to control the rotational velocity of the discharge and hence the thermal state of the electrodes and their erosion by changing the induction of a magnetic field whose influence on the process of formation of carbon nanostructures is not understood.

Experimental Setup. Figure 1 shows a diagram of a coaxial plasmatron with magnetic stabilization of an arc discharge which has been used for obtaining carbon nanostructures. The plasmatron incorporates: a copper support of the central electrode 1, on which a graphite anode 2 is fixed with the use of a threaded joint; solenoid 3 consisting of five series-connected sections each of which is manufactured from an 8×1 mm copper tube; a copper case of the cathode 4 with a graphite cathode 5 molded into it; a copper outlet nozzle 6; a copper cylindrical head 7; a copper spiral 8 (manufactured from a 6×1 mm tube) set into the head 7; insulator case 10; insulator 11 between the central anode electrode and the external cathode electrode. The working gas is fed without swirling via orifice 12 in the insulator. The anode support 1, the solenoid 3, the cathode case 4, the nozzle 6, and the spiral 8 are cooled with water 13. In applying voltage to the electrodes, an electric-discharge 9 is initiated by explosion of a steel wire of diameter 1 mm, fixed in the gap between the anode and the cathode. The graphite anode electrode has an outside diameter of 55 mm, an inside diameter of 36 mm, and a length of 70 mm. The graphite cathode electrode of length 100 mm represents a cylinder with an outside diameter of 92 mm and an inside diameter of 75 mm. Thus, the initial gap between the electrodes is 10 mm. To decrease heat removal the exterior surface of the cathode electrode has contact with the cathode case over three annular surfaces of width 15 mm. The structure of the discharge-chamber units allows experiments at atmospheric and higher-than-average pressures.

The setup is supplied with a d.c. current from two rectifiers with a controlled voltage to 1000 V; one is used for power supply of the arc discharge, whereas the other is used for supply of the solenoid, which enables us to change the current through its coils and hence the induction of the magnetic field. A cooled ballast resistor required for stable arcing and control of the current is incorporated in the power-supply circuit of the arc discharge. The nozzle 6, the head 7, and the case 10 are not insulated from the cathode and are at its potential when voltage is applied. The

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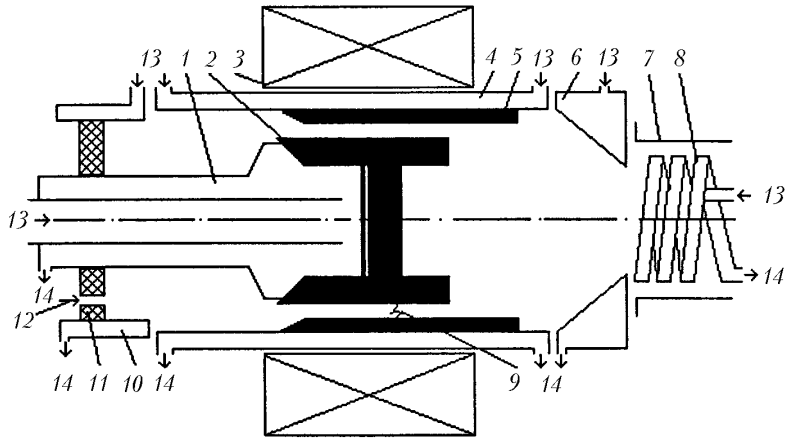
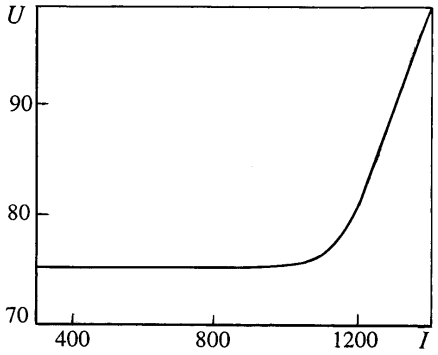


Fig. 1. Diagram of the setup for obtaining carbon nanostructures: 1) anode support; 2) anode; 3) solenoid; 4) cathode case; 5) cathode; 6) outlet nozzle; 7) head; 8) cooling spiral; 9) electric-arc discharge; 10) insulator case; 11) insulator; 12) feed of neutral gas; 13) supply of cooling water; 14) removal of cooling water.



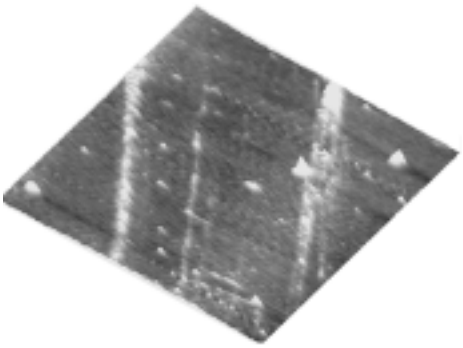


Fig. 2. Volt-ampere characteristic of the arc discharge. The argon flow rate is G = 0.5 g/sec and B = 0.2 T. I, A; U, V.

Fig. 3. Three-dimensional AFM image of carbon nanostructures deposited on the substrate. The scanning field is 4.6 × 4.6 μm, the maximum height is 5.5 nm, and the standard deviation of the height is 0.3 nm.

rate of flow of the inert gas argon can also be changed, but it was constant in our experiments and was 0.5 g/sec. The axial velocity of the gas in the electrode gap in an unperturbed flow is equal to 0.15 m/sec. The direction of the solenoid's axial magnetic field coincides with the direction of the gas flow. In interaction of the arc and the radial component of the magnetic field, at the periphery of the solenoid, the discharge is pulled in into the zone with a maximum magnetic field. The calculation has shown that the maximum value of the axial component of the induction of the magnetic field B on the section and at the center of the solenoid is equal to 0.31 and 0.36 T respectively for a current of 1000 A. The displacement of the magnetic field from the electrodes and the value of the magnetic field can be changed by disconnecting individual sections of the solenoid.

Results of the Experiments. Figure 2 gives the volt-ampere characteristic of the arc discharge. In the discharge-current range 300–1100 A, the characteristic has a horizontal portion for a voltage of ~75 V. The voltage increases to 100 V with increase in the current to 1400 A. The rotational velocity of the arc, calculated from the dependence of [3], was 100–500 m/sec in the discharge-current range 300–1400 A. The zone of attachment of the arc discharge on the anode is determined from the traces of the reference arc spots and represents a gray trail of width 12–15 mm, which is located nearly below the center of the solenoid.

Before the series of experiments and after it, we weighed the anode electrode and determined the specific erosion. Thus, in the range of discharge currents 300–1400 A, the average value of the specific erosion was $7.5 \cdot 10^{-5}$ g/C

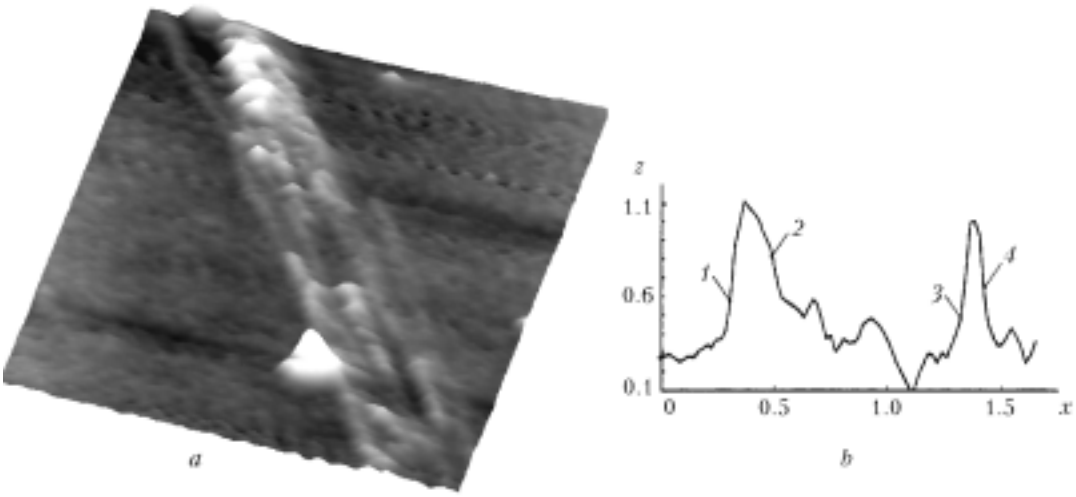


Fig. 4. AFM image of a nanotube beam (scanning field 2.2 × 2.2 μm, maximum height 5.5 nm, standard deviation of the height 0.4 nm): a) three-dimensional representation; b) profile cross section: 1) x = 305.1 and z = 0.5; 2) 488.2 and 0.8; 3) 1322.3 and 0.4; 4) 1424.0 and 0.6. x, μm; z, nm.

(0.07 g/sec per unit time). The carbon, evaporating from the graphite electrodes at the sites of action of the arc spots, condensed to form soot on the cold parts of the discharge chamber and the outlet of the setup. This is attributable to the presence of gas flows resulting from the rotation of the arc discharge in the electrode gap with a large velocity. These flows entrain nearby volumes of the working gas together with gaseous carbon and its solid particles and transfer them throughout the volume of the chamber. In this respect, we can probably compare the rotating discharge to a centrifugal pump.

After the series of experiments, we collected the soot from each structural element separately. From the carbon-based material obtained, we prepared a chloroform suspension in an ultrasonic bath; the suspension was subsequently diluted to a concentration of 0.2 mg/ml and was applied to a 5 × 5 mm polished silicon plate (substrate). The sample was dried for 30 min at room temperature and for 60 min at 150^{o}C. Visualization of the nanostructures obtained was carried out by the method of atomic force microscopy (AFM). The measurements were performed with an NT-206 meter (Mikrotestmashiny) in a contact regime. Use was made of CSS2I silicon probes (Micro Mash) with a point radius of about 10 nm and a rigidity of the cantilever of 0.12 N/m. The scanning fields were 4.6 × 4.6 μm in recording of the data at 256 × 256 image points. Figure 3 gives the AFM image of nanostructures deposited from the suspension on the substrate. Linear structures a few micrometers long, oriented in one direction, are pronounced within the limits of the scanning field. It can be assumed that in this manner we have been able to record carbon nanotubes.

The dissimilar transverse dimensions of the nanostructure are noteworthy in the picture. They are from 40 to 180 nm for a height of a few nanometers. It should be taken into account that in scanning of nanoparticles, we have a convolution of the curvature of the object measured and the curvature of the microprobe point. For the standard probe the result of measurement of the lateral dimension can be five times larger than the true value. On this basis, we can speak of the presence of single-wall carbon nanotubes and multiwall tubes with different number of walls on the substrate under study. A beam of individual nanotubes is recorded at the right-hand corner of the picture. A more detailed measurement of it on the 2.2 × 2.2 μm scanning field (Fig. 4) shows a combination of nanotubes of different diameter. An important feature of the sample of carbon nanotubes under study is the orientation of the nanotubes in one direction, be they combined into a beam or arranged separately.

CONCLUSIONS

Carbon nanostructures, in particular, nanotubes, have been obtained in the arc discharge between coaxial carbon-filled electrodes stabilized by a magnetic field in an atmospheric-pressure argon medium for currents of 300–1400

A. Further investigations will be aimed at obtaining data on the concentration of nanostructures in condensed carbon as a function of different factors, in particular, the strength of the external magnetic field, and at improving the structure and technology to increase productivity.

NOTATION

B, magnetic induction, T; *I*, current, A; *U*, voltage, V.

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