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Physicochemical Processes of Electroarc Synthesis of Carbon Nanomaterials

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The paper proposes an explanation of the physicochemical processes occurring during the electric-arc plasma-chemical synthesis of carbon nanomaterials. A diagram of the action of forces on charged particles and a diagram of forces acting on the motion of ions in an arc in the presence of a magnetic field for the plasma-chemical synthesis of carbon nanomaterials are presented and considered. The levels of organization of matter in an arc discharge during the formation of carbon vapour with an increase in temperature are considered. A comparative characterization and a conditional scheme for the formation of various carbon nanostructures during plasma-chemical synthesis are proposed. The sequence of carbon transformations during the formation of carbon nanomaterials and the conditional levels of organization of matter with the types of processes occurring on each of them during structure formation, as the classification of carbon structures by size levels under conditions of temperature decrease from plasma temperature to room one are established. Carbon nanomaterials are synthesized by the plasma-chemical method, and the analysis of the obtained and processed carbon nanostructures is carried out by the following methods: scanning electron microscopy, transmission electron microscopy, Raman spectral analysis, UV–VIS spectroscopy, spectrophotometric analysis. The presence of fullerenes in the products of plasma-chemical synthesis is established, and fullerenes are extracted from the synthesized wall soot.

В даній роботі запропоновано пояснення фізико-хемічних процесів, що відбуваються під час електродугової плазмохемічної синтези вуглецевих наноматеріялів. Наведено та розглянуто схему дії сил на заряджені частинки та схему дії сил на рух йонів у дузі за наявности магнетного поля плазмохемічної синтези вуглецевих наноматеріялів. Розглянуто рівні організації матерії у дуговому розряді під час утворення Карбонової пари з підвищенням температури. Наведено схему розташування зон робочого простору реактора від осі дуги (за тиску > 20 ГПа й температури > 12 000°C) до стінки реактора (за температури > 600°C). Запропоновано порівняльну характеризацію й умовну схему утворення різних вуглецевих наноструктур під час плазмохемічної синтези. Встановлено послідовність перетворень вуглецю під час формування вуглецевих наноматеріялів та умовні рівні організації матерії за типами процесів, які відбуваються на кожному з них під час структуроутворення, класифікацію вуглецевих структур за розмірними рівнями в умовах пониження температури від плазмової до кімнатної. Також синтезовано вуглецеві наноматеріяли плазмохемічним методом і проведено аналізу одержаних й оброблених вуглецевих наноструктур методами: сканувальної електронної мікроскопії, просвітлювальної електронної мікроскопії, спектральної аналізи комбінаційного розсіяння світла, спектроскопії в оптичному (видимому) діяпазоні довжин хвиль із прилеглим до нього ультрафіолетовим діяпазоном, спектрофотометричної аналізи. Було встановлено присутність фуллеренів у продуктах плазмохемічної синтези, а також проведено екстракцію фуллеренів із синтезованої пристінної сажі.

Key words: carbon nanomaterials, fullerenes, fullerites, endofullerenes, carbon nanotubes, synthesis, self-organization, plasma, electric arc.

Ключові слова: вуглецеві наноматеріяли, фуллерени, фуллерити, ендофуллерени, вуглецеві нанотрубки, синтеза, самоорганізація, плазма, електрична дуга.

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1. INTRODUCTION

To date, it is very important to understand the physicochemical processes of the synthesis of carbon nanomaterials (CNM), in order to be able to predict the formation of their modifications and create new structures and nanomaterials.

At this level of development of science, a wide range of methods for the synthesis of carbon nanostructures (CNS) is known (laser, detonation, pyrolysis, plasma-chemical in a gaseous medium, and plasma-chemical in a liquid medium) [1–10]. Equipment for the synthesis of CNS can have its own modifications that allow synthesis with the maximum yield of the product of any particular CNS, or allows you to create their modifications. CNM are divided into soluble and insoluble CNS in hydrocarbon solvents. Soluble CNS include fullerenes [11–15], endofullerenes [16–18], exofullerenes, and fullerites [19, 20]. Insoluble CNS include nanotubes, nanofibers, graphenes, and graphene packets [21–25].

Due to such a large number of methods for the synthesis of CNM and a wide range of types of CNS, it has been possible to use them as fillers and reinforcing materials in the automotive and aircraft industries, as well as in 3*D*-printing technologies to increase the tribological characteristics of materials [26–29]. Particular attention is paid to CNM in hydrogen energy, where the developed surface of CNS is used to create new fuel cells on their basis, and soluble CNM are considered as future hydrogen adsorbers [30–36], which can compete with already known sorbents [37–62].

That is why it is important to understand the processes of CNM synthesis in order to increase the range of use of CNS and for the more intensive development of nanotechnology today.

2. PLASMA CHEMICAL SYNTHESIS EQUIPMENT FOR CARBON NANOMATERIALS

The plasma chemical discharge is based on the action of an electric arc that occurs between graphite electrodes in an inert atmosphere (helium or argon). The installation was developed by Krätschmer *et al.* [63] specifically for the synthesis of fullerenes. After modernization (increasing the working temperature of the plasma formed between two graphite rods), the method allows not only for the production of fullerenes, but also other carbon nanomaterials (CNM). This was first seriously considered when obtaining and investigating fullerene-like materials.

In our work, plasma chemical vacuum equipment (Fig. 1) was used for the synthesis CNM.

Fig. 1. Plasma-chemical vacuum equipment for the synthesis of carbon nanostructures.

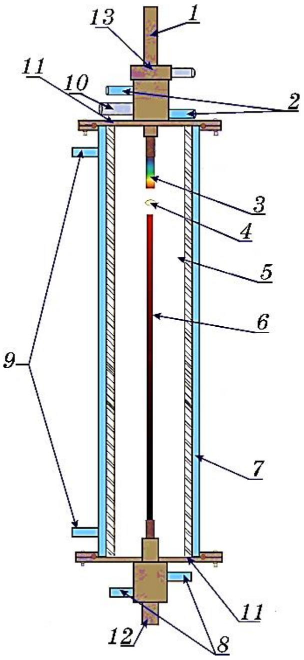


Fig. 2. The reactor of the equipment for the synthesis of nanostructured carbon samples. *1*—cathode; *2*—cathode coolers; *3*—a movable current-carrying non-consumable graphite rod; *4*—plasma zone; *5*—helium atmosphere; *6*—working graphite rod; *7*—thermostatic device; *8*—anode cooling; *9*—reactor cooler; *10*—device for effective pumping of the reactor to a deep vacuum; *11*—reactor sealing flanges; *12*—anode; *13*—replaceable catalytic sleeves.

Experiments to obtain working samples of CNS were conducted in a reactor (Fig. 2) with an internal diameter of 150 mm, which has a water-cooled jacket and flanges with circular rubber seals. The jacket temperature is 25–30°C, and the inner wall of the reactor has a temperature of 500–600°C.

Teflon insulators for current leads are attached to the flanges (cathode 1 and anode 6, respectively). A graphite rod is fixed on the cathode using a copper clamp, which does not wear out, while a high-quality graphite rod (FPG-7 grade) measuring 800 mm is fixed on the anode, which wears out during the experiment. The cathode is connected (through a ceramic insulator) to a movable polished stainless-steel stem that slides in Wilson seals and transmits translational motion from a non-consumable cathode to a stepper motor. A scale is applied to the stem to measure electrode consumption rates and prevent the melting of stems and clamps. Graphite washers are placed between copper holders and graphite rods.

The reactor has a device for pumping the reactor to medium vacuum (MV) from 10^2 to 10^{-1} Pa and filling the internal space with inert gases, as well as pressure control, which fully ensures the maintenance of a stable vacuum in the reactor.

Graphite was evaporated in a vacuum under a helium pressure of 0.02–0.09 MPa and a voltage on the electrodes of 22–30 V and a current of 250–300 A.

3. PHYSICOCHEMICAL PROCESSES OF PLASMA CHEMICAL SYNTHESIS OF CARBON NANOMATERIALS

The thermodynamic stability region of gaseous carbon lies in the plasma temperature zone ($\cong$ 8 000 K), so, the transition of carbon atoms to an excited state is only possible at temperatures significantly higher than this value. In the interelectrode gap (Fig. 3), conditions arise that allow carbon atoms to be transferred from graphite electrodes to both a free (atomic) and plasma state.

As seen in Fig. 3, the area with the highest temperature in the arc discharge is the cathode ($\cong$ 12 000 K). The surface of the anode is heated less ($\cong$ 10 000 K), but the temperature on its surface reaches values when carbon atomization is already possible. Thus, ‘carbon vapour’ is generated near the surface of both electrodes.

To understand the mechanisms of obtaining carbon nanomaterials in the interelectrode space, it is necessary to consider the processes that occur there. They inevitably affect the process of formation and chemical composition of the products that are formed during the destruction of the anodic carbon precursor.

The distribution of charged particles in different sections of the electric arc is non-uniform: the particles concentration will be high-

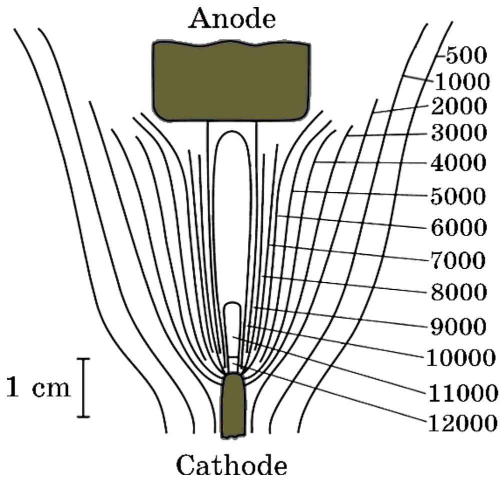


Fig. 3. Distribution of temperature (in [K]) zones along the axis of the electric arc between graphite electrodes at a current of 200 A [64].

est along its axis. At the same time, some electrons and positive ions will move from the arc centre to the periphery along the radius, *i.e.*, perpendicular to the magnetic field (the simultaneous action of magnetic and electric fields). The force F acting on an electric charge in a magnetic field is perpendicular to the velocity and trajectory of motion and will have the greatest impact. Its value depends on the charge (e), average particle velocity (v), magnetic induction (B), and angle between the directions of the magnetic induction and particle velocity, *i.e.*, $F = evB\sin\alpha$. This force (Fig. 4) will be maximal at an angle of $\alpha = 90°$ (then, $F = evB$), and smallest at $\alpha = 0$ (in this case, particles move only under the influence of the electric field). The direction of the force depends on the magnetic field formed by the electrodes.

Hence, particles in the arc column under the influence of a magnetic field will rotate around the axis of the arc.

The rotations of ions and electrons occur (in accordance with their opposite charges) in opposite directions; they can also capture neutral particles that are in the arc column.

As the distance from the axis of the electric arc increases, the temperature and concentration of ions, as well as the speed of gas particle rotation, decrease. The rotational movement of gas particles in the arc column, caused by the magnetic field action, further reduces the diffusion rate and 'pulls' the ionized hot gas towards the axis of the plasma column. Because of the mutual influence of these factors, charged particles will move in spirals that narrow (Fig. 5).

The centripetal force that arises when particles rotate in a circle

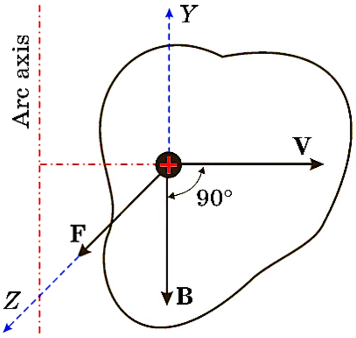


Fig. 4. Scheme of the forces' action on charged particles in a magnetic field.

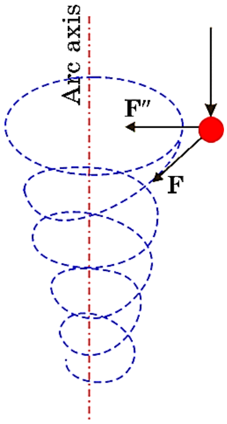


Fig. 5. Scheme of the forces' action on the ions' movement within the arc in the presence of a longitudinal magnetic field.

also contributes to the increase in gas concentration gradient near the axis of the electric arc.

The influence of electric and magnetic fields on the resulting carbon particles is crucial in the formation of carbon nanostructures. The interelectrode arc can be considered as a conductor of electric current with its own magnetic field, and the force lines of the arc cord form closed circles.

It is known that the passage of electric current in metals is due to the movement of electrons, which react to the influence of positively charged ions fixed in the crystal lattice. Unlike the metal matrix, cations in the arc column, like electrons, move, forming their own magnetic field.

In the plasma that arises between graphite electrodes, there are two opposing streams of charged particles (electrons and ions with a positive charge); the latter consist of carbon cations, carbon clusters, and graphene fragments.

The speed of movement of positively charged particles depends on the action of the electromagnetic field created between the electrodes.

The opposing movement of oppositely charged particles promotes their collision, and the energy of interaction depends on their momentum. This can lead to the destruction of existing structures, as well as ionization of atoms.

The considered processes are consistent with the provisions of the kinetic theory of electromagnetic processes by Yu. L. Klimontovich [65] and the results of other studies [66–85].

3.1. Physicochemical Processes at the Anode

During the transition of carbon from a solid state to a gaseous state, its volume increases significantly. It should be taken into account that when such a volume is heated from 300 K to 12 000 K, the pressure increases $\cong 40$ times. Thus, it can be concluded that during the arc evaporation of graphite, local pressures in the microvolumes of the arc discharge can exceed 20 GPa, *i.e.*, the formation of nuclei of carbon structures during an arc discharge occurs under conditions of high pressure.

3.2. Organization of Matter in Plasma Chemical Synthesis with the Formation of Carbon Vapour

According to our research [1–25] and the works of other scientists [63, 64, 66–69, 73, 80, 85], we propose an illustrative scheme of one of the variants of the carbon vapour formation process (Fig. 6) with an increase in temperature (from room one to 12 000 K).

The entire process can be divided into five main energy stages. In the first stage (stage I), compact graphite is under normal conditions, where graphene sheets are bound together by Van der Waals forces. Upon heating the graphite above 4 500 K (stage II), one-, two-, and three-layered fragments of graphene sheets begin to separate from its surface under the influence of the arc discharge.

At the next stage (III), the increase in temperature and collision of graphene sheets with the electron stream leads to their destruction into cyclic and chain fragments, where atoms are bounded by covalent bonds. Further temperature increase (stage IV) and collision of cyclic and chain formations with the electron stream leads to the formation of multiatomic particles. In the final stage (V),

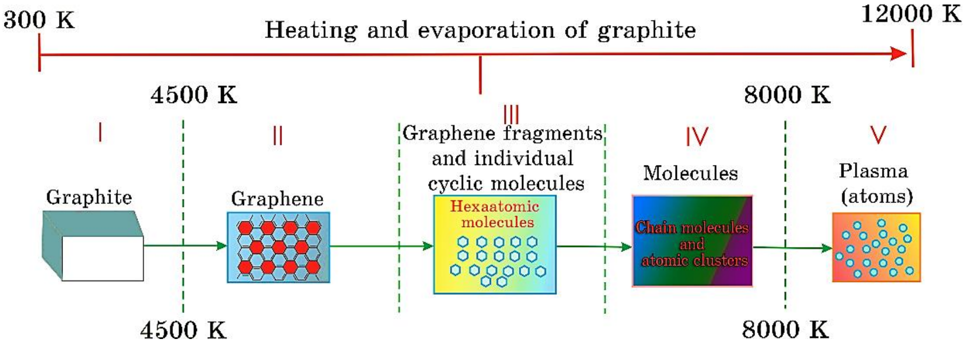


Fig. 6. Levels of organization of matter in an arc discharge during the formation of carbon vapour with an increase in temperature.

there is plasma interaction between cations, electrons, and other charged particles. These transformations occur sequentially between graphite electrodes in the plasma of an arc discharge.

3.3. Processes Occurring in the Gas Phase and on the Reactor Walls during Plasma Chemical Synthesis

Next, we focus on the processes, which occur in the internal electrode space, as well as on the reactor wall.

3.3.1. Schematic Model of Reactor Space Zones

As the distance from the axis of the arc discharge increases (Fig. 7), working parameters (temperature, pressure) of the environment and reactant composition [66–69, 80, 85] change radially. Additionally, substance concentration in different cross-sections of the electric arc will be uneven. Electrons are displaced towards the axis of the arc, and a greater number of positively charged particles are trapped in the magnetic field.

The temperature of the central part of the arc significantly decreases (by radius) towards the periphery. Neutral particles move

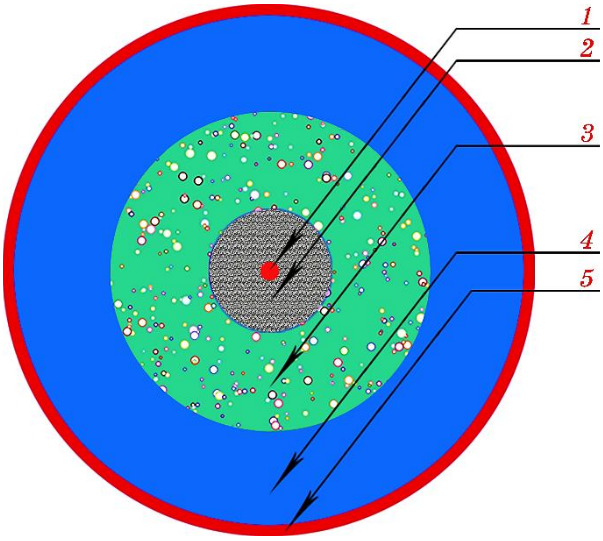


Fig. 7. Diagram of the location of the zones of the working space of the reactor. Zone '*1*'—arc axis ($P > 20$ GPa, $T > 12\,000$°C); zone '*2*' ($P \cong 0.5$–0.6 MPa, $T \cong 4\,000$–$13\,000$°C); zone '*3*' ($P \approx 0.1$–0.2 MPa, $T \cong 200$–300°C); zone '*4*' ($P \approx 0.09$ MPa, $T > 600$°C); zone '*5*'—reactor wall ($T > 600$°C).

away from the axis of the arc due to temperature and pressure gradients, entering the reactor ash filled with helium and forming ring-shaped clusters, which can be considered as coaxially arranged reaction zones [86].

In zone 1, due to high temperature, the precursor graphite undergoes destruction. The transition of carbon from a solid state to a gaseous state and high temperature contribute to the emergence of a sufficiently high pressure in the mixture, which leads to the formation of compounds that participate in the formation of various nanostructures. Zone 2 has lower pressure due to the condensation of gaseous carbon (carbon atoms, chain formations, cyclic molecules, as well as fullerene and graphene as microstructure nuclei). Zone 3 is the coldest. The temperature of zone 4 rises again to $\cong 600°C$ due to radiative heating and bombardment of the reactor wall by electrons. Thus, zone 4 has conditions favourable for pyrolytic processes and plays a significant role as a catalyst in this zone (see Fig. 2).

The formed particles that reach the reactor wall are made up of carbon atoms generated in zone 1. Then, under the influence of temperature gradients (up to 12 000 K) and pressure ($>$ 20 GPa), they enter to zone 4, where conditions sufficient for pyrolysis processes are achieved, and the presence of a metal phase in the reactor wall (zone 5) or a variable sleeve (Fig. 2) can catalyse structural transformation processes. The distance from the centre to the wall of the reactor shell (zone 5) plays a special role in the synthesis of carbon nanoparticles.

3.3.2. Formation of Carbon Nanomaterials during Plasma Chemical Synthesis

As mentioned above, charged carbon particles are held by an electromagnetic field [65, 67–69] and cannot practically escape into the interelectrode space in large quantities. Therefore, in transformations, which occur in the gas phase (outside the arc zone and on the walls of the reactor), their insignificant leakage can be disregarded, focusing only on the behaviour of neutral particles.

The ratio of the number of neutral and charged particles generated during graphite evaporation depends on the conditions of the process. Based on the results of studies of the composition and morphology of products formed on the reactor walls, a conditional scheme can be compiled (Fig. 8) [4].

Some amount of carbon vapour, consisting of neutral particles of the plasma flow, under experimental conditions, moves under the influence of the T and P gradients and leaves the interelectrode space at a speed of over 20–25 m/s, reaching the reactor wall in

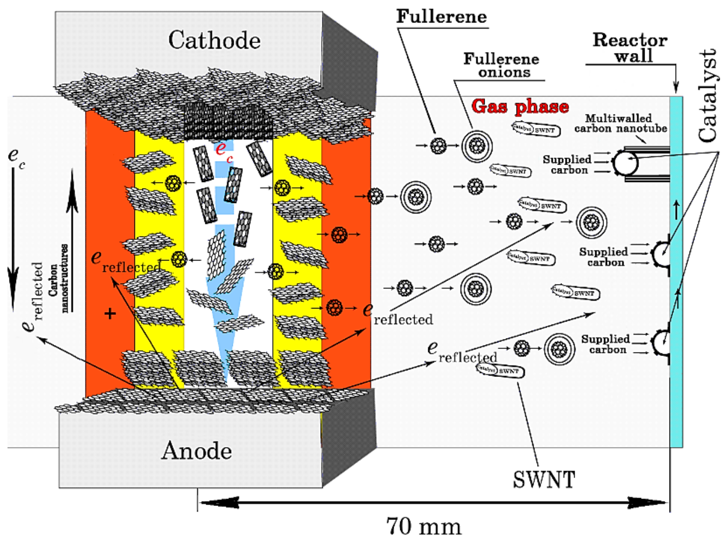


Fig. 8. Conventional scheme of the formation of various carbon nanostructures (CNS) during the arc process: SWNT—single-walled carbon nanotubes (SWCNT); e_c—the flow of electrons moving from the cathode to the anode; $e_{\text{reflected}}$—reflected electrons; carbon nanostructures with a positive charge $[\text{CNS}]^+$; carbon nanotubes (CNT).

≅ 0.003 seconds and cooling almost to room temperature. During this time, a series of processes occur, the duration of which varies within nanoseconds, and the resulting product (wall soot) is located on the reactor walls.

As the distance from the axis of the electric arc increases, the temperature and concentration of particles, carbon atoms, and radicals decrease, while their geometric dimensions increase. The diffusion rate and number of collisions with other particles per unit time also decrease. As a result, different structures can form in the gas phase. If fullerenes are formed, they can transform into onions, and in the presence of a catalyst, into nanotubes or other structures.

Experimental studies [1–25, 4, 70–72, 74-75, 77, 79, 81, 84, 87, 88, 89, 90] have shown that metals catalyse both the processes of fullerene destruction and synthesis of other graphite-like structures.

During the operation of the electric arc, a lot of energy is released in the form of radiation. This raises the temperature of the inner surface of the reactor to over 600°C. Electromagnetic radiation and electron beams generated by carbon plasma promote the transition of metal atoms from the surface layers of the metal cartridge case to the gas phase, which affects the formation of new carbon nanomaterials.

4. FEATURES OF THE FORMATION OF CARBON NANOMATERIALS

Previous studies [1–25] and our experiments [4, 87, 70–72, 77, 79, 81, 84] indicate that the duration of the carbon nanoparticle synthesis process (especially the time that the reactants spend in the reaction zone) varies, although the morphology of the nuclei formed at early stages may be identical. For example, the residence time of reactants in the reaction zone during pyrolysis is of $\cong 1{\cdot}10^4$ s, while, during arc synthesis in the gas phase, it is of $\cong 3{\cdot}10^{-3}$ s. Moreover, our studies of arc synthesis in liquid environment show that the residence time of reactants in the reaction zone is $< 1{\cdot}10^{-9}$ s. [1, 24, 25, 88, 90–93] (Fig. 9). Thus, the difference in the duration of the process between these synthesis methods is $\cong 13$ orders of magnitude. Therefore, the transition from one method to another and the reduction of interaction time do not significantly affect the morphology of the formed nuclei but mainly affect the yield of products and depend primarily on changes in the geometry of the formed products.

Thus, during the synthesis of carbon nanomaterials by any method, nanoscale objects are first formed, which become nuclei of new structural units. Therefore, special attention should be paid to nanosecond-duration processes that determine the morphology and properties of the final products.

4.1. Transformation of Carbon during the Formation of Carbon Nanomaterials

Based on the results of our studies on the formation processes of

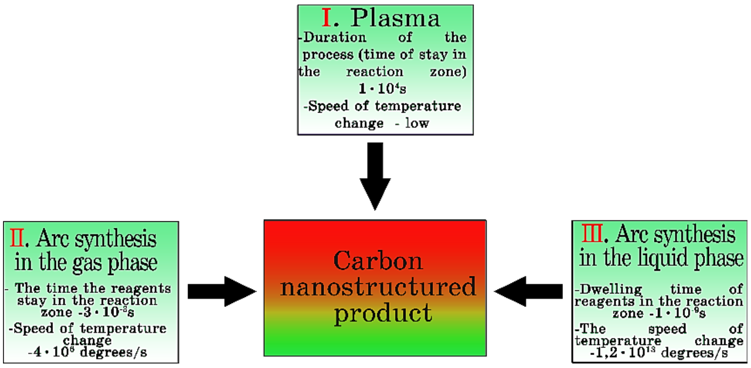


Fig. 9. Comparative characteristics of the processes used in the synthesis of carbon structures.

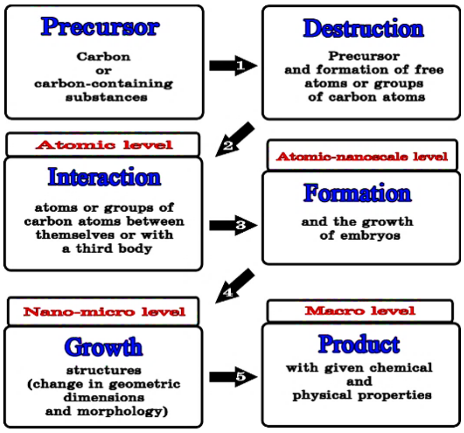


Fig. 10. The sequence of carbon transformations during the formation of carbon nanomaterials.

carbon nanomaterials [1–25, 87, 91, 92, 93], we have determined the technological chain of transformations that the starting carbon-containing reagents undergo (Fig. 10). Thus, for the synthesis of new structures, carbon or carbon-containing precursors are chosen, from which nuclei of a certain structure are formed. Depending on the synthesis conditions, these can be carbon, graphite, diamond, fullerenes, nanotubes, and other allotropic (or polymorphic) modifications of carbon.

The method of synthesis of carbon nanomaterials determines the energetic state of the starting reagents [67, 68, 80, 85], and therefore, the features of the formation and structure of the synthesized materials.

4.2. Classification of Carbon Nanomaterials by Levels of Matter Organization

The duration of the synthesis of carbon nanomaterials determines the change in their geometric dimensions. The physicochemical nature of CNM is determined by the thermodynamics of the nucleation process. Interaction at the atomic level occurs relatively quickly (fraction of nanosecond). To obtain a product with a certain dispersion, namely, a material consisting of particles of certain geometric dimensions and structure, as well as having defined properties, it is necessary to control the interaction time at each level of organization of structure formation.

The nucleus can consist of chains of different lengths and branching, cycles and polyhedra. Its skeleton can be frame-like or a combination of these structural elements. As the number of atoms in the cluster skeleton increases, the diversity of ways they can be connected grows. At nuclearity above 20, a spherical spatial structure becomes thermodynamically and geometrically most favourable, that is characteristic for fullerene carbon clusters.

Based on an analysis of available information, the conditions and mechanisms of processes, which occur during the synthesis of carbon nanomaterials, have been considered [67, 68, 85]. A classification of carbon structures by dimensional levels has also been carried out, and the types of processes occurring at each level have been considered. The main results of the studies are presented in Fig. 11 (bottom part), from which it follows that the processes that occur during the formation of carbon nanoscale materials have a nanosecond duration, occur at the atomic-molecular level, and can be technologically controlled. Figure 11 (bottom part) shows the classification of carbon structures and the processes of their formation using the ‘primary table’ of S. P. Gubin.

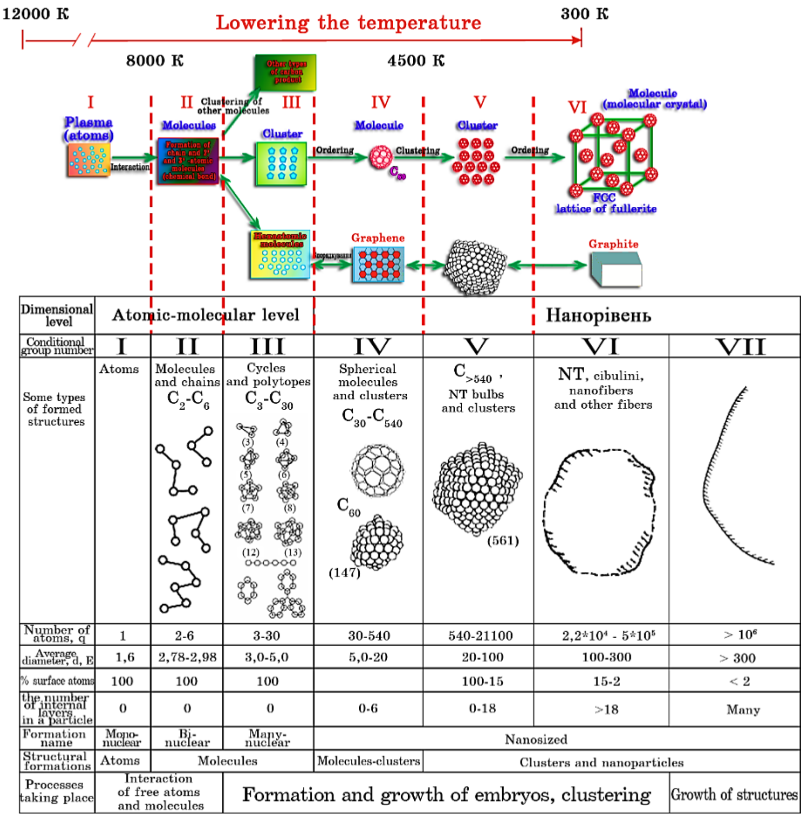
5. PROCESSES OF FORMATION OF CARBON MOLECULES

As mentioned above, the processes that lead to the formation of carbon vapour and the formation of nanomaterials have nanosecond duration and occur under highly non-equilibrium conditions. The explanation of the facts of nanomaterials synthesis, which lead to the self-organization of the discussed systems into new spatial-temporal structures, is usually considered within the framework of the thermodynamics of non-equilibrium processes [94, 95]. However, creating a sequence of transformation processes that occur during the transformation of matter at all levels of organization of CNS from carbon vapour causes some difficulties.

The synthesis of particles and their transformation into substances at a reduced temperature of the system is fundamentally possible due to changes in their thermodynamic characteristics. Figure 11 (top part) shows the proposed mechanism for the synthesis process of fullerite from carbon vapour at a temperature decrease from plasma to room temperature.

During the cooling of carbon vapour, covalent structures arise. At the first stage, structures are formed, which use p-electrons of a carbon atom to form a chemical bond. The bond energy in this chain is the highest (720 kJ/mol).

At the second stage, s-covalent structures arise, which have a lower bond energy between molecules, which form high-molecular compounds (711 kJ/mol—energy of graphene molecule destruction, 714 kJ/mol—energy of C_{60} molecule destruction). The bond between molecules is formed by s-electrons in an excited state (675 kJ/mol—



Dimensional level	Atomic-molecular level			Нанорівень			
Conditional group number	I	II	III	IV	V	VI	VII
Some types of formed structures	Atoms	Molecules and chains C_2-C_6	Cycles and polytopes C_3-C_{30}	Spherical molecules and clusters C_{30}-C_{540}	$C_{>540}$, NT bulbs and clusters	NT, cibulini, nanofibers and other fibers	
Number of atoms, q	1	2-6	3-30	30-540	540-21100	$2{,}2*10^{4}$ - $5*10^{5}$	$> 10^{6}$
Average diameter, d, E	1,6	2,78-2,98	3,0-5,0	5,0-20	20-100	100-300	> 300
% surface atoms	100	100	100		100-15	15-2	< 2
the number of internal layers in a particle	0	0	0	0-6	0-18	>18	Many
Formation name	Mono-nuclear	Bi-nuclear	Many-nuclear	Nanosized			
Structural formations	Atoms	Molecules		Molecules-clusters	Clusters and nanoparticles		
Processes taking place	Interaction of free atoms and molecules		Formation and growth of embryos, clustering				Growth of structures

Fig. 11. Conventional levels of the matter organization, the types of processes, which occur in each of them at structure formation, and the classification of carbon structures by dimensional levels under the conditions of a decrease in temperature from plasma to normal.

energy of carbon atom transition to an excited state $2s \to 2p$) (σ-bond).

Figure 11 (top part) shows the levels of material organization from carbon vapour to the formation of a spherical carbon molecule up to the fullerite lattice with a temperature change from 12 000 K to room temperature.

During clustering and ordering of carbon vapour, Van der Waals structures are formed, which are crystalline substances. The bond in them is formed using *s*-electrons (π-bond). Taking into account the above, the bond energy varies from 17 kJ/mol (bond energy between graphite layers) to 180 kJ/mol (energy of destruction of the fullerite structure).

The process of forming CNS can be conditionally divided into six main stages: I—plasma interaction of atoms; II—formation of 2-

and 3-atomic particles due to chemical bonding; III—formation of chain and cyclic molecules and their clustering; IV—ordering of five-atom molecules to form a spherical C_{60} structure using covalent bonds; V—clustering of spherical carbon molecule C_{60}. Stage VI is carried out after the extraction of C_{60} molecules from soot and their crystallization from solutions—molecular crystals of fullerite with f.c.c. lattices are formed.

Figure 11 provides information on the structural transformations of carbon at different levels of matter organization, the types of processes that occur at each of them during structure formation, and the classification of carbon structures at different dimensional levels. The proposed scheme makes it possible to trace the transformations that the system of carbon atoms undergoes due to energetically stimulated structural transformations at different levels of matter organization.

6. EXPERIMENTAL DATA OF PRODUCTS OF PLASMOCHEMICAL SYNTHESIS OF CARBON NANOMATERIALS

There are many methods of analysing synthesized soluble [11–20] and insoluble carbon nanomaterials [21–25, 96].

In a vertical reactor, the anode (consumable electrode) remains stationary, and the cathode (non-consumable electrode) moves along the axis of the reactor. Thus, the conditions of uniform distribution of sediment over the entire inner surface of the reactor walls are met and relatively the same thermodynamic conditions for condensation of synthesis products are achieved (Fig. 2).

Working graphite electrodes doped with catalytic additives evaporate in a helium environment at a pressure of 0.02–0.09 MPa.

On the walls of the reactor, a soot-like sediment is formed [90], which contains soluble (fullerenes and fullerene-like structures) and insoluble nanomaterials (nanocomposites, carbon nanotubes, graphene). The removal of wall soot is carried out by opening the upper and lower reactor flanges (Fig. 2), followed by the displacement of the sediment with a rubber piston from top to bottom. The wall soot is collected in a storage container.

The structures of carbon products were studied using scanning (JSM-T20) and transmission (JEM 100 CXII) electron microscopes (SEM and TEM). The results of electron microscopy indicate that the nanoscale components have different geometric shapes and structures (Figs. 12–16).

The soluble components of wall soot are fullerenes and fullerene-like structures obtained by extraction (Fig. 14).

The results of the analysis of wall soot by Raman spectroscopy

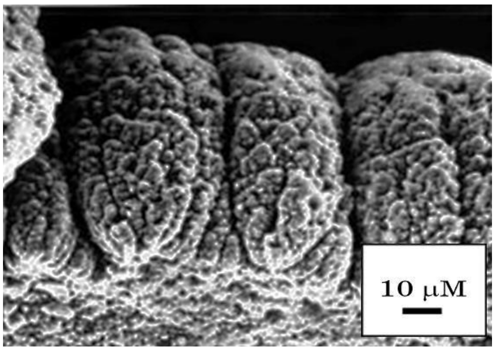


Fig. 12. SEM-image of insoluble nanostructures of wall soot treated with ultrasound in an alcohol environment.

Fig. 13. TEM-image of the morphology of nanosized particles that are a component of wall soot.

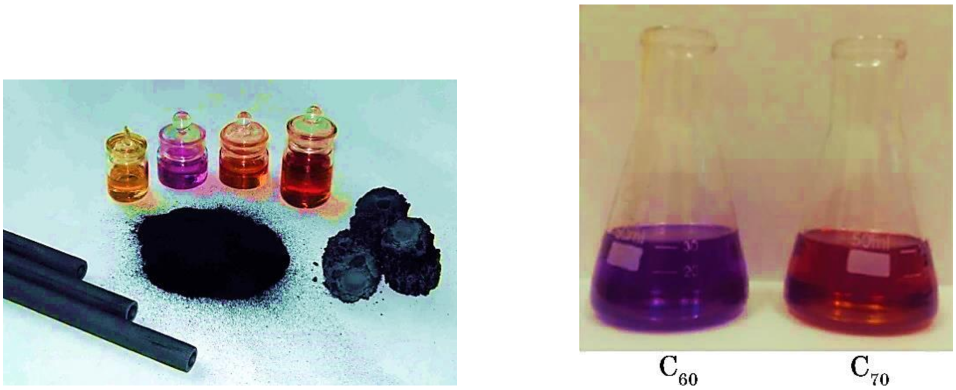
are shown in Fig. 15, where the two-peak structure of the *G*-maximum indicates the presence of two types of nanotubes in the samples; the maximum near the frequency of 1570 cm^{-1} is associated with 'armchair' type nanotubes (conductor), and the maximum near the frequency of 1590 cm^{-1} is associated with 'zigzag' nanotubes (semiconductor).

Quantitative and qualitative analysis of fullerene solutions was carried out by UV–VIS-spectroscopy (Fig. 16), where fullerenes C_{60} and C_{70} were detected in extraction solutions of wall soot, where the peak maxima $\lambda_{max} = 335.6$ and 407 are characteristic for C_{60} fullerenes and $\lambda_{max} = 334.6$ and 472.8 are characteristic for C_{70} fullerenes.

7. CONCLUSIONS

Based on literature data and our own experimental studies of graphite arc vaporization, a model of the processes of carbon product synthesis formation has been proposed. The model is based on the peculiarities of particle behaviour in an electromagnetic field at extremely high pressure and temperature gradients.

One of the main conclusions can be considered the fact that micro- and macroquantities of carbon nanomaterials are formed at the



a *b*

Fig. 14. Soluble component of wall soot. (*a*) Fullerenes, wall carbon black, graphite and electrodes are an allotropic form of carbon; (*b*) fullerenes obtained after extraction of wall soot.

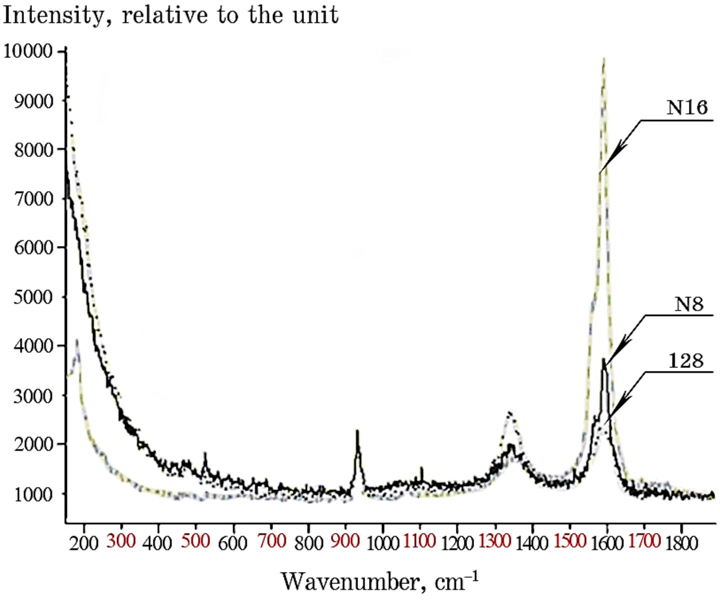


Fig. 15. Raman spectrum of wall soot.

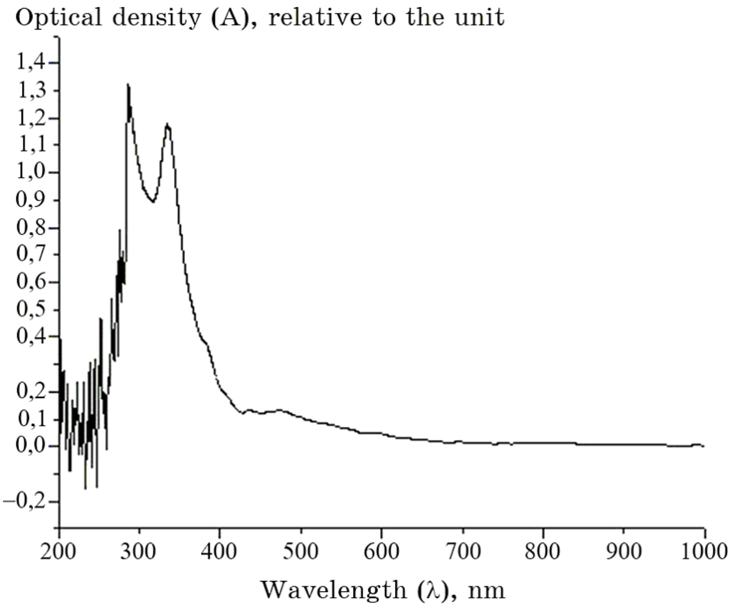


Fig. 16. Optical density UV–VIS spectroscopy. Spectrum of fullerene solution.

stage of nuclei formation, *i.e.*, the nanostructural product consists of nuclei of different structures.

The sequence of transformations, which the initial carbon-containing reagents undergo during the formation of carbon nanostructural modifications, has been justified. A classification of carbon structures and the processes of their formation have been developed. A sequence of levels of material organization has been proposed, which reflects the mechanism of formation of spherical carbon molecules C_{60} from carbon vapour, when the temperature changes from 12 000 K to room temperature. Carbon nanomaterials, including fullerene-like materials, are formed in the vapour phase because of interaction between carbon atoms.

The proposed scheme of carbon structural transformations at different levels of matter organization, types of processes occurring at different dimensional levels allows us to trace the transformations that a system of carbon atoms undergoes due to energetically stimulated structural transformations at different levels of matter organization.

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