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SYNTHESIS OF GRAPHENE LAYERS IN FLAME

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Abstract

The paper presents the results on the synthesis of graphene films in premixed hydrocarbon flames. The parameters of combustion, catalytic substrate type were selected and the vertical orientation of the flame, providing education of the smallest number of layers with the greatest structural regularity. Found that in a hydrocarbon flame on a nickel substrate is formed mainly 5-10 graphene layers. It is shown that when the angle of inclination of the substrate relative to the vertical axis of the flame from 0 ° to 30 ° can obtain the minimum number of layers - 2-3 layers. The research on the effect of the ratio C/O and argon flow on the formation of layered graphene films, depending on the type of fuel. The formation of graphene layers are investigated in a wide range of values of C/O = 0.75 to 1.05. It is shown that with increasing values of C/O and the amount of argon introduced into the flame, forming layers of graphene disorder increases.

Keywords: fullerene, graphene, sphere, ellipsoid, tube

Intoduction

Graphene is a single layer two-dimensional carbon structure, the surface of which is laid out in regular hexagons with sides of 0.142 nm and the carbon atoms at the vertices. In its properties, graphene is a semimetal with a small overlap of the conduction and valence bands.

Currently, much attention is paid to graphene because of its unusual and potentially useful properties such as high electrical and thermal conductivity, the dependence of electronic properties on the presence of attached radicals of different nature on the surface of graphene, an adjustable band gap, quantum Hall effect, extremely high mobility of charge carriers (at room temperature it reaches 104 cm2/(V·s), which is significantly higher than mobility in silicon), high elasticity and good electromechanical characteristics. These properties allow using it in future as the basis for new nanomaterials with improved mechanical, electrical and thermal physical characteristics, as well as an element of nanoelectronic devices. However, widespread use of graphene will be possible after detailed studies of its characteristics, as well as by the development of reliable and relatively inexpensive methods for synthesis of these structures.

Synthesis of graphene layers in a hydrocarbon flame

Since the discovery of the first method of isolating graphene, based on mechanical cleavage of graphite layers [1, 4-7], the efforts of many research laboratories focused on developing new and more effective ways to solve the low volume production.

Many methods, such as liquid-phase oxidation of graphite, chemical vapor deposition, epitaxial growth on the metal surface, thermal decomposition of silicon carbide, the creation of graphene in an electric arc have all been used to obtain graphene [7]. However, these methods are very complicated and time-consuming and predominantly result in low yields of graphene. The search for new, less labor intensive and more efficient methods of graphene synthesis is an urgent task.

One such possibility may be production of graphene layers in the flame. The process of flame formation of nanostructures is a fast reaction usually 10-5 to 10-3 seconds. Flame production is a continuous method, simple, inexpensive, and has the ability to develop into a mass production technique.

Comparative studies with a number of authors [2-3] confirm that the preliminary process of obtaining graphene in the flame can successfully compete with its synthesis by chemical vapor deposition (CVD).

This paper presents the results on the synthesis of graphene films in a flame: the parameters of combustion, the type of catalytic substrate and its orientation in the flame, providing the smallest number of layers with the greatest structural regularity.

Synthesis of graphene layers in the double flame

In paper [8] the authors present results on synthesis of graphene layers in a double flame at atmospheric pressure a schematic representation of which is shown in Fig. 1.

A nickel plate which was used as a catalist was placed vertically in an alcohol flame (burner 1), and it had a protective function preventing oxidation of the substrate by atmospheric oxygen. The alcohol flame was the source carbon atoms and the flame from the second burner was a source of heat for carbonization of carbon and growth of graphene layers on the substrate.

The process took place at the temperature above 800 ° C for 10-40 seconds, then after turning off the burners, the substrate was covered and cooled rapidly to room temperature. Upon cooling, solubility of carbon decreases and there takes place formation of surface layers of graphene. Varying the temperature conditions of the experiment, graphene layers with different amounts of impurities were produced [8]. After purification and separation of graphene layers from the substrate they were examined using an electron microscope. The results are shown in Fig. 2 [8].

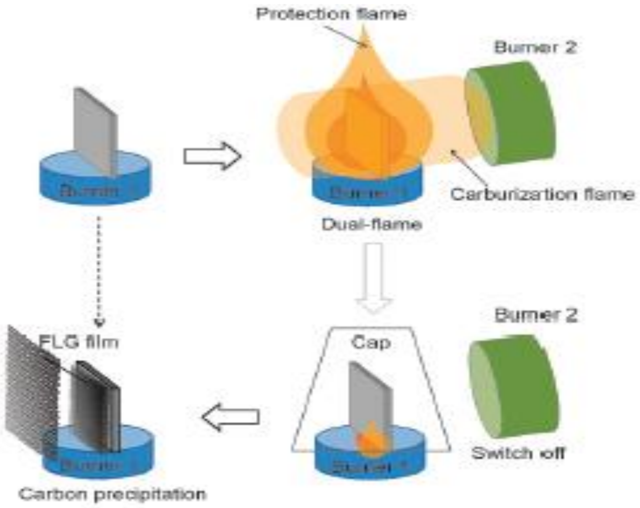


Fig. 1 – Schematic representation of the dual-flame for the synthesis of graphene layers on a nickel substrate [8]

The presence of graphene layers (Figure 2, a and b) was confirmed by electron diffraction pattern with characteristic hexagonal packing of carbon atoms (Fig. 2, c) with the help of TEM of high resolution it was determined that the number of graphene layers being formed was in the range of 2-5 (Fig. 2, d). It is found that pre-annealing at 1000 °C for two hours and the thickness of the nickel substrate are key conditions for formation of high quality graphene layers. Annealing results in the removed of the oxide layer and the surface contaminants on the nickel substrate this leading to the synthesis of a single layer graphene, Raman spectrum of which is shown in Fig. 3 (a) (upper spectrum) [8].

The studies on synthesis of graphene layers, where xylene, hexane, acetone, ethanol, methanol were used as fuel for protective flame, showed that the fuel with a high carbon content results in formation of graphene layers with a large amount of an amorphous structure (Fig. 3) [8]. The best quality of graphene layers was observed, when using ethanol and methanol (Fig. 10) [8].

Synthesis of graphene layers in the diffusion back flame

Of considerable interest are the results on production of graphene layers in the diffusion back flame back on nickel and copper substrates with the thickness of 25 micrometers at atmospheric conditions [9]. For synthesis, a multi-burner flame with a separate combustion process was used, which is schematically presented in Fig. 4 [9].

As a fuel, hydrogen is used with the introduction of CH4, which is a source of carbon for synthesis of graphene layers. The fuel came out on the edges of the burner and the oxidizer was in the center. The reverse flame is characterized by lower soot formation and the presence of unburned fuel under going pyrolysis on substrate.

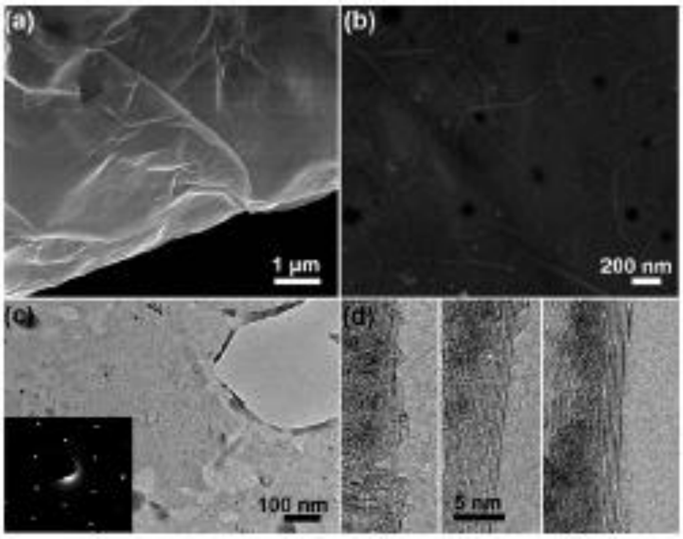


Fig. 2 – Graphene layers with their identification by means of SEM (a, b) and TEM (d) [8]

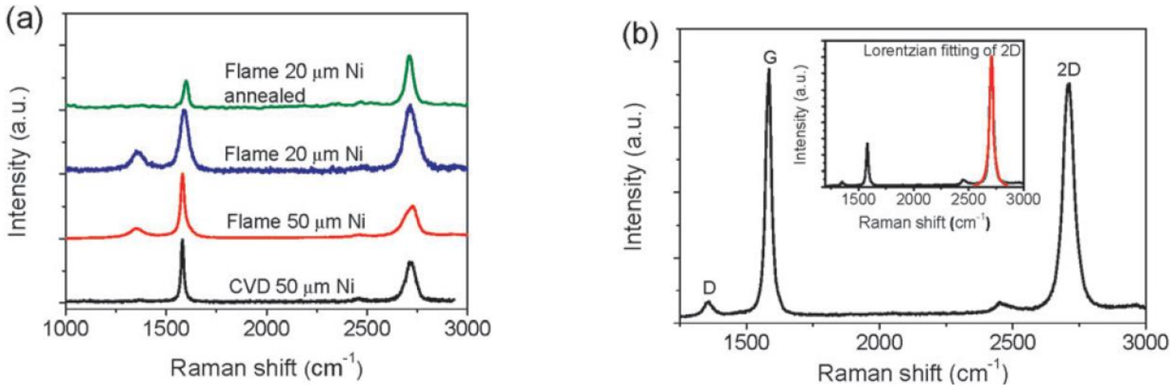


Fig. 3 – Raman spectra of graphene layers obtained in the flame by CVD method (a), and with different fuels of the protective flame (b) [8]

The experiments were performed in the following order: the substrate was preliminarily subjected to roasting in rich hydrogen flame for 10 minutes, then the graphene layers were synthesized in the process of methane feed (at an equivalent ratio of ~ 3) for 5-10 minutes at 950 °C; after putting out the supply of oxygen and extinction of the flame there occurs cooling of the substrate to room temperature under the effect of inert gases and fuel [9].

After purification and separation of the graphene layers from the substrate with SiO2/Si, they were examined on the Raman spectrometer (λ = 514.5 nm). Raman spectrum of graphene layers synthesized on a copper substrate is shown in Fig. 5 [9].

Graphene layers were transferred to a quartz substrate (Fig. 5, a) and studied on the atomic force microscope. The presence of flakes of graphene with the thickness of about 4 nm, which corresponds to 8-10 layers of graphene was stated (Fig. 5, a, b). Raman spectrum of graphene layers has three characteristic peaks D, G and 2D (Fig. 5, d).

IG/I2D peak intensity ratio is in the range of 1.3 to 1.7 that characterizes the presence of about 3-10 layers of graphene (Fig. 5, c) [9]. The ratio of the peak intensities IG/I2D in this case is 0.35, which is comparable with the dimensions of graphene layers obtained by other methods [10].

The effect of the flame temperature on synthesis of graphene layers on a copper substrate in the form of changes in the ratio of peak intensities I2D/IG is shown in Fig. 6 [9].

It is shown that the temperature is a determining factor in the synthesis of graphene layers. The increase in temperature result in the increase in the intensity of The change in the intensities of 2D-lines of Raman spectra 2D-peak, which characterizes the decrease in the amount of synthesized layers on a copper plate with the increase in their quality [9].

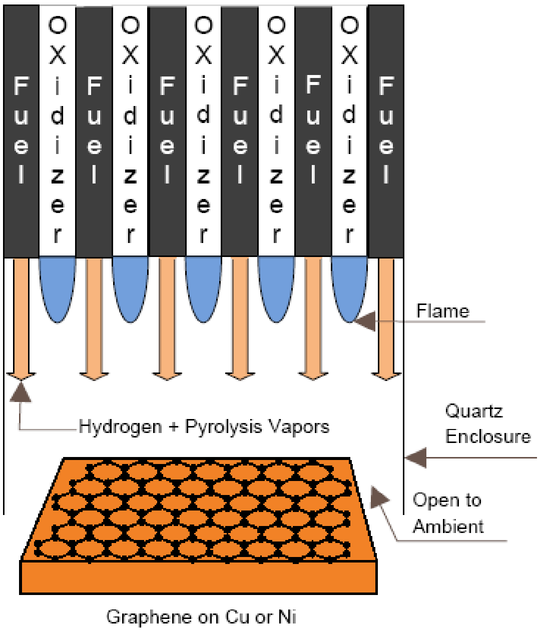
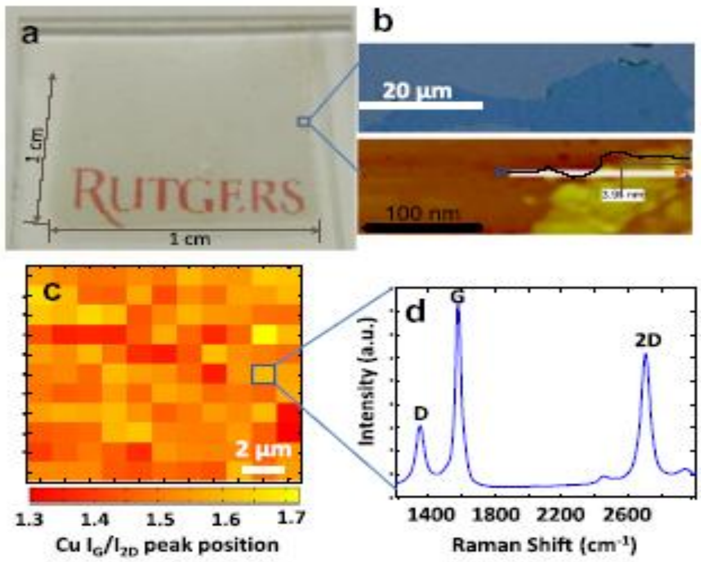


Fig. 4 – Schematic representation of the synthesis of graphene layers with the use of the burner with the inverse diffusion flame [9]



(a) - layers of graphene deposited on a quartz substrate 1 x 1 cm;
(b) - Pictures of graphene layers on the substrate SiO2/Si, obtained in an optical microscope and the height of the graphene layer profile obtained with an atomic force microscope,
(c) - two-dimensional image of the change in the ratio of Raman spectra peaks IG/I2D on the surface with the size more than 12 x 12 μm;
(d) - Raman spectrum of graphene layers on the SiO2/Si substrate with the thinkness of, an oxide film of 300 nm [9]

Fig. 5 – Graphene layers synthesized in flames on a copper substrate

The decrease in temperature result in the decrease in the intensity of 2D-peak and the increase in the intensity of D-peak, which characterizes the increase of the defect of graphene layers being synthesized. Typical Raman spectrum of graphene layers produced at low temperature is shown in the inset of Fig. 6 [9]. The effect of the ratio of CH4: H2 on the growth of graphene layers on a copper substrate is stated: at values of 1:5 to 1:20, there occurs the growth of graphene layers, and at the value of 1:40 the growth of graphene layers is not observed.

The research results on synthesis of graphene layers on a nickel substrate have shown that the ratio of the intensities of I2D/IG of typical Raman spectrum shown in Fig. 7 corresponds to 5-10 layers of graphene [9]. TEM image of graphene layers synthesized on a nickel substrate and the diffraction pattern are shown in Fig. 7 (b) [9].

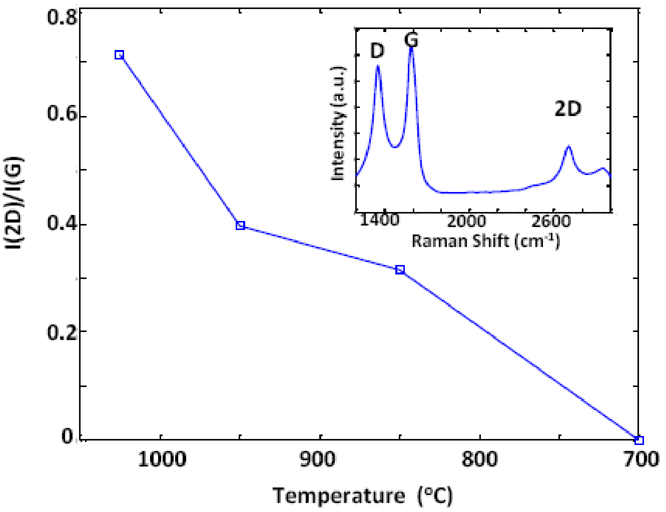


Fig. 6 – The effect of temperature on the growth of graphene layers on a copper substrate by changing the ratio IG/I2D and a typical Raman spectrum of a graphitized structure observed at low temperatures (shown in the inset) [9]

The defects of graphene layers formed on the nickel substrate (ID/IG = 0.1, Fig. 7, a) were stated to be than those of smaller graphene layers synthesized on a copper substrate (ID/IG = 0.35, Fig. 5, d), which is due to different mechanisms of growth of graphene layers on nickel and copper substrates [9].

Different solubility of carbon in copper and nickel determines the different mechanism of the growth of graphene layers. The extremely low level of carbon dissolution in copper forms the surface mechanism of growth of graphene layers on the substrate.

Surface growth of graphene on a copper substrate creates the preconditions for controlling the number of layers on the surface of the copper substrate.

However, it creates certain requirements to the parameters of the flame. As the synthesis of graphene on a copper substrate process on the surface, excess soot formation leads to a rapid coating of the surface with soot and ceases formation of graphene layers.

On a nickel substrate at the temperature above 800 °C, there takes place formation of a metastable solid phase and, upon cooling, carbon diffuses from nickel onto the surface to form graphene.

This process in some extent limits the number of layers on a nickel substrate. The growth of graphene layers on a nickel substrate can last longer than that on the copper substrate.

Fig. 8 shows the comparative picture of the growth of graphene layers with the ratio of CH_4: H_2 = 1:10 on Cu and Ni substrates by peak 2D of Raman spectrum [9].

For Cu, methane was introduced during 10 minutes at the temperature of 950 °C, and for Ni, methane was injected within 5 minutes at the temperature of 850 ^{o}C.

It is shown that the values of 2D peak of graphene layers for a copper substrate does not exceed 2700 cm-1, which characterizes the limit and the formation of several layers of graphene (see Fig. 7, a). The values of 2D peak of graphene layers on a nickel substrate reaches 2727 cm-1, indicating the formation of more than 10 layers (Fig. 7, b) [9].

Synthesis of graphene layers in premixed flames

The investigation on formation of layered graphene films were carried out in the flame comprising a premixed propane-oxygen mixture at atmospheric pressure under the following conditions: flow rate of propane - 219 cm3/min, oxygen consumption - 381 cm3/min, corresponding to the ratio of C/O=0.86.

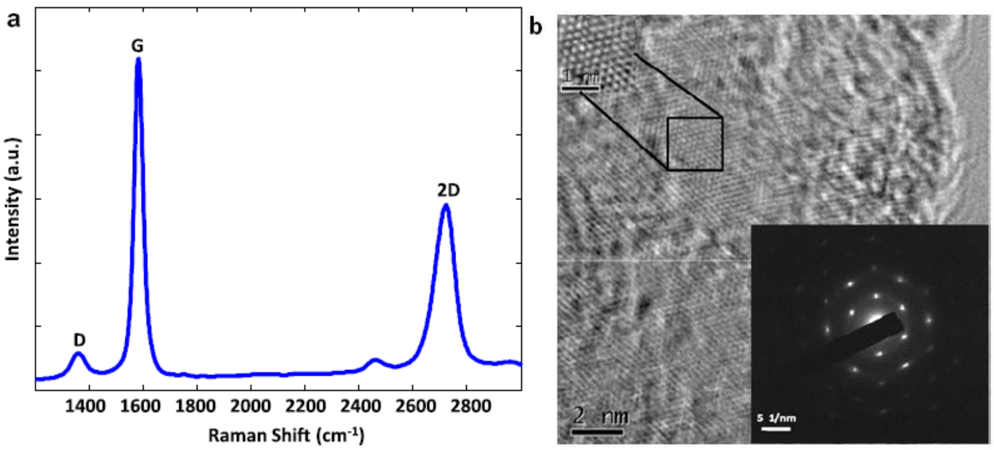
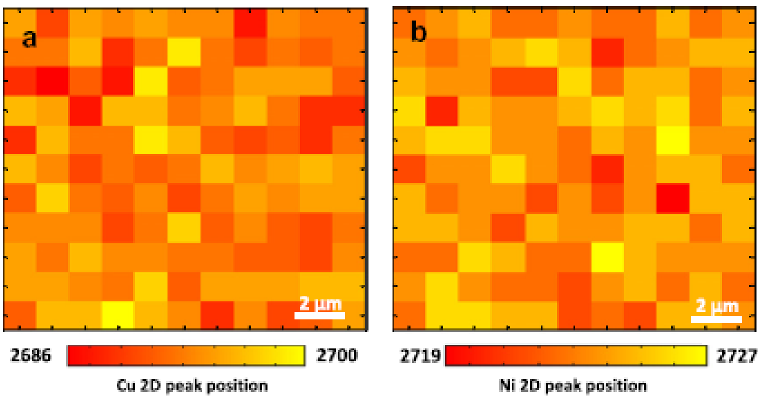


Fig. 7 – Raman spectrum of graphene layers synthesized in flames on a nickel substrate (a) TEM image (b) and the diffraction pattern showing the crystal structure of graphene (b, inset) [9]



a - Raman mapping for Cu, illustrating the self-limiting growth of several graphene layers
b - Raman mapping for Ni, indicating the growth of 10 layers of graphene [9]

Fig. 8 – Raman peak 2D mapping of 12 um x 12 um with a ratio CH4: H2 = 1:10

The studies were also carried out with the addition of argon in the propane-oxygen mixture in an amount of 300-650 cm3/min. The substrates were used to support the growth of graphene consisting of either copper or nickel plates, which were placed in the central part of the flame [11].

The residence time of the substrate in the flame was varied: 5, 10, 20, 30, 40, 60 seconds, 5 and 10 minutes, and the angle of the substrate relative to the vertical axis of the flame was altered: α = 0, 30, 45, 60, 85°. The flame temperature in the experiments was in the range 900-950 °C. Measurements of the temperature profile in the central part of the flame were conducted using a chromel-alumel thermocouple in steps each measurement point being at the distance of 5 mm, and a reference point was taken at the base of burner. The results of measurements of the temperature profile at the height in the middle of the propane-oxygen flame by adding argon are shown in Table 1.

Table 1 – The temperatures at the height of the central part of the propane-oxygen-argon flame

The value of C/O	0.75	0.85	0.95	1.05
Consumption of O2 for C3H8, cm3/min	438.2	386.6	345.9	313.0
Consumption of O2 for C4H10, cm3/min	584.3	515.5	461.3	417.3

At the second stage, the process of formation of layered graphene films in the flame of premixed propane-oxygen and butane-oxygen mixtures at atmospheric pressure depending on to the ratio C/O with addition of argon was studied. Argon was fed in the amount of 150, 250, 350, 450 and 500 cm3/min. The residence time of the plate in the flame was 5 minutes. Since the flame temperature is largely dependent on the amount of argon and the ratio C/O, and with placing of the substrate in the flame its temperature decreases by 30-50 °C due to the heat dissipation by the plate and the holder, the initial flame temperature was within 950 - 970 oC. The flame temperature in the process of synthesis of graphene on a catalytic substrate was maintained within 900 -920 °C.

The study of formation of layered graphene films was carried out at a constant rate of propane or butane consumption in amount of 219.1 cm3/min. In order to obtain the desired ratio C/O, oxygen consumption was varied the values of which are given in Table 2.

Table 2 – The values of oxygen consumption depending on the type of fuel to obtain the designed ratio C/O

The flame height, mm	Voltage of the thermocouple, mV	Temperature, oC
5	36	867
10	37	892
15	36	867
20	39	943
25	41	993
30	40	968

In the experimental study of the influence of the ratio C/O on synthesis of graphene layers in the combustion of premixed propane-oxygen or butane-oxygen mixture, the consumption of argon was constant and was equal to 250 cm3/min.

The flame temperature largely depends on the amount of argon in the given point of the flame, which explains the variation of the temperature traversing the height of the flame. Samples of the obtained soot on the substrate were investigated by Raman spectroscopy (NTEGRA spectra at an excitation wavelength λ= 473 nm).

Analysis of the samples showed that the formation of layered graphene films is preferred when using a nickel substrate. A copper substrate, which has been used elsewhere to support the growth of single layers of carbon, is exposed to strong oxidation conditions in the flame to form an oxide coverage of the surface, which flakes on cooling.

It is stated that the formation of graphene layers on the substrate occurs when it is placed vertically in the flame. The formation of graphene occurs on both sides of the substrate.

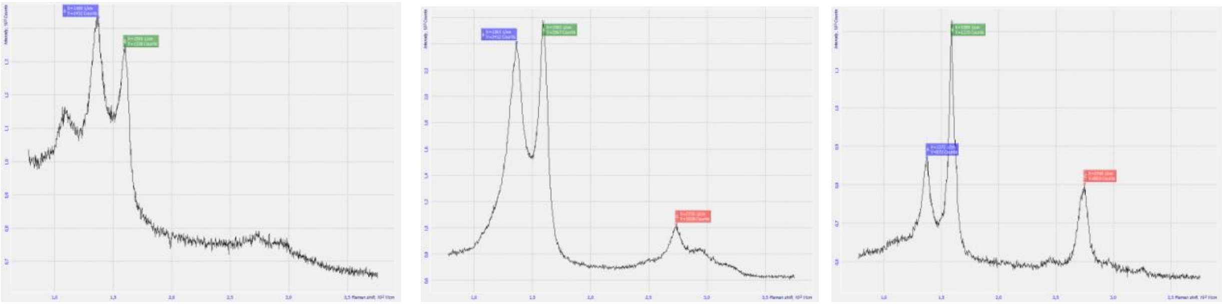
When tilting the substrate at an angle relative to the vertical axis on the side closer to the stabilizer, the graphene formation region increases. It is determined that the zone of formation of graphene layers precedes the border of soot particles. When tilting the substrate at an angle relative to the vertical axis on the side closer to the stabilizer, the graphene formation region is further increased.

In the typical spectrum of graphene there are three peaks: the first peak corresponds to the D-band at 1351 cm-1, the second peak is the G-band at 1580 cm-1 and the third peak is the 2D band at 2700 cm-1 [9, 9]. The rratio of the intensity of the G peak (IG) and 2D peak (I2D) provides an estimate of the number of graphene layers (IG/I2D) [9]. For single-layer graphene, this ratio is less than unity.

The studies reveal that the increase the residence time of the substrate in the flame from 5 to 60 seconds correlates to the formation of graphene layers exhibiting the decrease in the degree of disorder of their graphitized structure, characterized by the appearance of 2D peak at 2700 cm-1 and the decrease in the intensity of D-peak at 1351 cm-1, Fig. 9.

The increase of the residence time of the substrate in the flame to five minutes results in the stable formation of graphene layers with a decreasing degree of disorder. The further increase in the residence time does not significantly change the resulting graphene structures on the substrate. Further studies were conducted on a nickel substrate during the exposure time of 5 minutes. Estimation of the influence of the substrate tilt angle relative to the vertical axis of the flame on formation of graphene layers elucidated the fact that formation of graphene layers occurs best at the inclination of less than 90o.

When the substrate is at an angle of 90o (perpendicular to the axis of the flame), the formation of an amorphous carbon black structure was resulted.



a b c

a – 5 seconds, b - 30 seconds, c – 60 seconds

Fig. 9 – Raman spectra of samples revealing a graphitized carbon structure on a nickel substrate at different exposure times (C/O=0.86)

The changing ratio of IG/I2D, characterized by the number of graphene layers on the substrate, depends on the angle of the substrate with respect to the vertical axis of the flame and is shown in Table 3.

Table 3 – The ratio values of IG/I2D depending on the angle of the nickel substrate relative to the vertical axis of the flame

The angle of inclination, degrees	The minimum value, IG/I2D	The maximum value, IG/I2D
0	1.3	2.0
30	1.1	1.9
45	1.7	2.4
60	2.0	2.3
85	2.0	2.2

It was determined that the minimum number of graphene layers (two or three layers, IG/I2D=1.1-1.3) occurred in the range of the angle of inclination of the substrate between 0° and 30°, Fig. 10.

Placing the substrate at an angle of 30o to the vertical axis of the flame yielded two layers of graphene (IG/I2D = 1.1), as depicted in the Raman spectrum shown in Fig. 2, b. The further increase in the angle of inclination of the substrate relative to the vertical axis of the flame (over 30 °) leads to the increase in the minimum number of graphene layers formed on a substrate, i.e. from 5 to 10 layers (IG/I2D = 1.7-2.4), (Fig. 10, c).

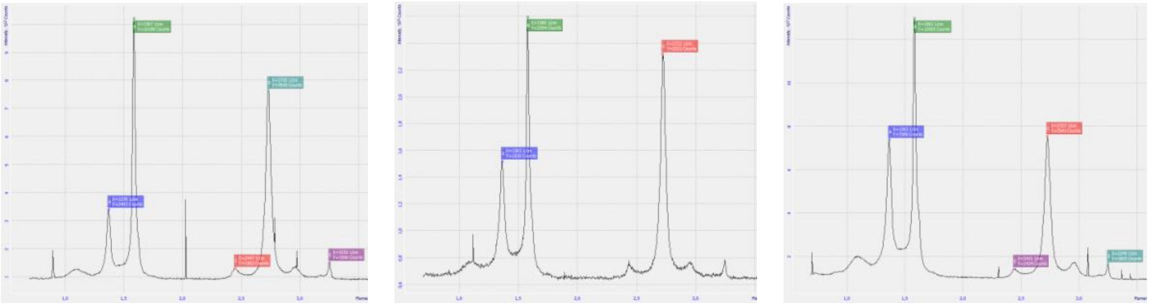
The studies on the formation of graphene layers in the propane-oxygen flame without the addition of argon on a nickel substrate during the exposure time of 5 minutes and the inclination of 30o showed that the substrate contained a minimum of 5 to 10 layers (IG/I2D = 1.6-2.5), as revealed in the Raman spectra in Fig. 11.

It is also evident that the zone of formation of graphene layers precedes the border of the formation of soot particles. To confirm this, a series of experiments were conducted where the flame on a nickel substrate did not reach the area of that soot formation, but was part of it. After, five minutes, the substrate was removed from the flame (Fig. 12).

Preceding the area of particulate structures is a zone of graphenes, shown by the arrow in Fig. 12. It is confirmed that only the formation of particulate structures on the surface of the substrate contributes to the formation of graphene layers. Fig. 13 shows the Raman spectra of the resulting structures as they approach the particulate layer.

The data obtained from Raman spectroscopy of graphene formation on a nickel substrate, allowed elucidation of the size of graphene layers formed, both in the vertical and horizontal directions. It was discovered, that the formation of a continuous layer of graphenes, as a rule, takes place in the vertical direction.

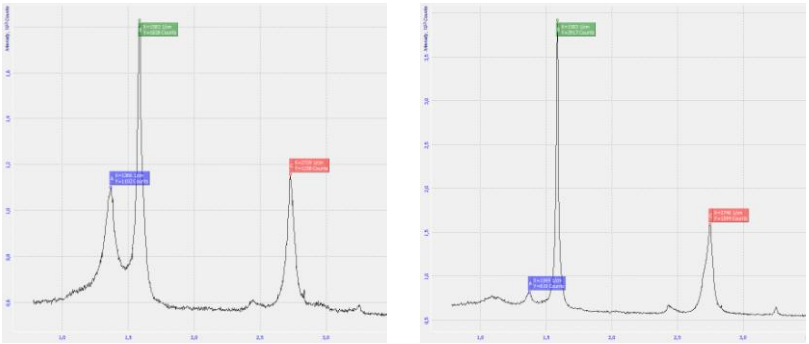
The maximum size of the graphene layer achieved in the vertical direction in these studies was 2 mm, and the horizontal - one to five micrometers (Fig. 13). However, there is scope to transform these early results into rapid graphene formation over wider areas using a flame combustion approach.



a b c

a - three layers (α = 0o, IG/I2D = 1.3),
b - two layers (α = 30o, IG/I2D = 1.1),
c – five layers (α = 45o, IG/I2D = 1.7)

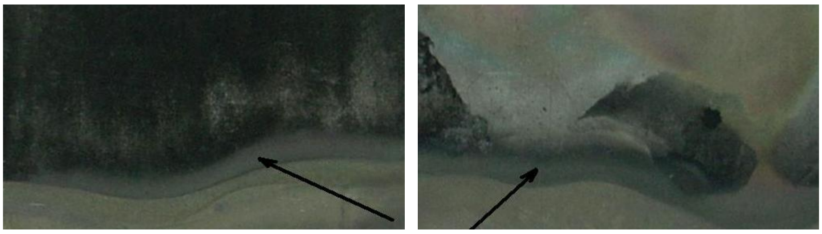
Fig. 10 – Raman spectra of graphene on a nickel substrate (C/O = 0.86, t = 917oC, τ = 5 min)



a b

a – five layers (IG/I2D = 1.6), b – ten layers (IG/I2D = 2.5)

Fig. 11 – Raman spectra of graphene layers synthesized on a nickel substrate in propane-oxygen flame without addition of argon (C/O=0.86, t = 917oC, τ = 5 min)



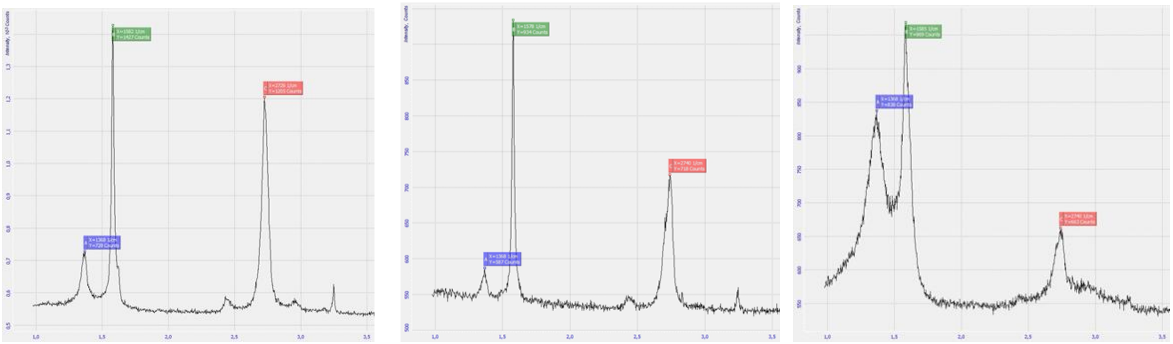
а б

a - the inner side of the cylinder of the nickel substrate,
b - the outer side of the cylinder of nickel substrate

Fig. 12 – Photo of a nickel substrate with synthesized carbon structure

The experimental results on synthesis of graphene layers under the conditions of combustion of premixed propane and butane-oxygen mixtures at various ratios of C/O are shown below. Argon consumption in all the experiments was constant and equal to 250 cm3/min. The results of the obtained minimum ratio IG/I2D, characterizing the number of graphene layers on the substrate depending on the type of initial fuel and the ratio C/O are given in Table 4.

It is preferably in the flame of propane and butane under these conditions that formation of graphene layers in an amount of 5-10 layers was stated to take place. The results of Raman spectroscopic studies of soot samples are shown in Fig. 14 and 15.



a b c

a - two layers (IG/I2D = 1.2);
b - three layers (IG/I2D = 1.3);
c - five layers (IG/I2D = 1.5)

Fig. 13 – Raman spectra of samples with a graphitized carbon structure on a nickel substrate

Table 4 – The minimum ratio IG/I2D depending on the ratio of C/O and type of fuel

The value of the ratio, C/O	0.75	0.85	0.95	1.05
Propane, IG/I2D	2.2	1.8	1.5	1.9
Butane, IG/I2D	1.5	2.0	1.3	1.6

It is found that the graphene layers are formed independently of the type of fuel in the studied range of ratios C/O with a pronounced peak of graphite G. The tendency for increasing the degree of disorder of the graphene layers with increasing ratio of C/O, which is characterized by the increase in the intensity D-peak is observed. For propane-oxygen mixture, the degree of disorder is observed at a higher ratio of C/O than for butane-oxygen mixture. However, butane-oxygen mixture at the ratio of C/O = 0.95, the minimum number of graphene layers was obtained (three layers, IG/I2D = 1.3).

The effect of argon flow on synthesis of graphene layers with its supply to the propane or butane-oxygen flames was studied. It is found that at the ratio C/O = 0.75 for propane-oxygen flame, the increase in the argon content from 150 cm3/min to 500 cm3/min does not activate the process of formation of graphene layers, but leads to the formation of soot of predominantly amorphous structure, which is shown in the Raman spectra presented in Fig. 16.

A similar situation for propane-oxygen flame is observed at a higher ratio of C/O. Raman spectra of graphene layers at C/O = 1.05 with a different consumption of argon are shown in Fig. 17.

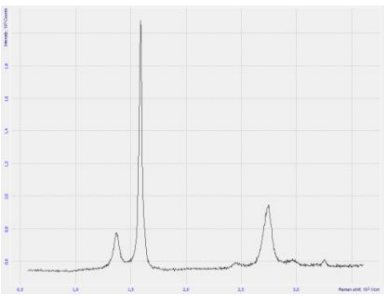
In the propane-oxygen flame at the ratio of C/O = 1.05 with the feed of argon 150 cm3/min on a nickel substrate we observed the formation of a minimum number of graphene layers (Fig. 17, and three layers, IG/I2D = 1.3).

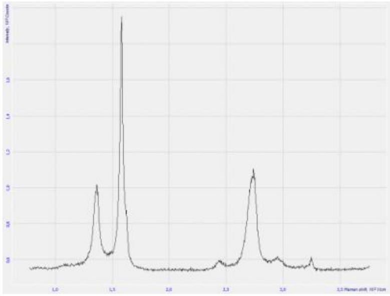
For butane-oxygen flame at C/O = 0.75 the increase in the feed of argon does not lead to the formation of an amorphous soot structure, as observed in propane-oxygen flame (Fig. 16). Raman spectra of the obtained graphene layers are shown in Fig. 18.

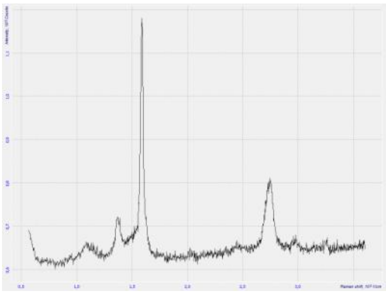
The increase in argon consumption at C/O = 0.75 in butan-oxygen flame result in formation of minimum number of graphene layers (Fig. 18, b: three layers, IG/I2D = 1.3).

At a greater ratio of C/O > 0.75 for butane-oxygen flame the increase of argon consumption leads to a greater disorder of graphene layers being formed as observed in propane-oxygen flame. Raman spectra of the obtained soot structures are shown in Fig. 19.

However, in butane-oxygen flame this process is observed at a higher flow rate of argon (500 cm3/min) than for the oxygen-propane flame (250 cm3/min). Earlier, H. Bockhorn [12] proposed a general reaction scheme for soot formation in a homogeneous mixture of premixed flames, where intermediate products are polycyclic aromatic hydrocarbons (PAHs). After that this scheme was developed and modified by Mansurov Z.A. in [13], where he noted that PAHs can be considered as the basis of graphene formation in flames. Fig. 20 shows the modified scheme for soot and graphene formation process in flames.


a


b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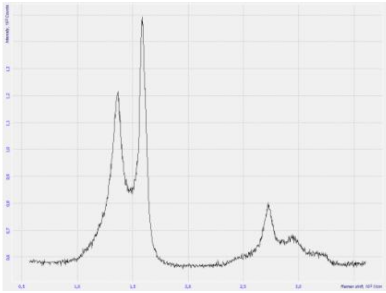
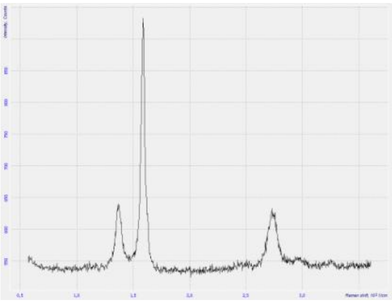
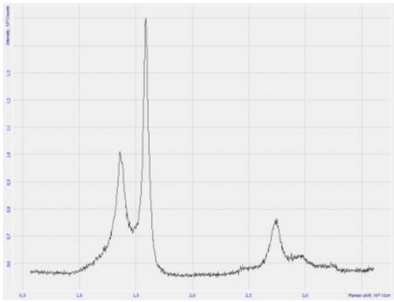
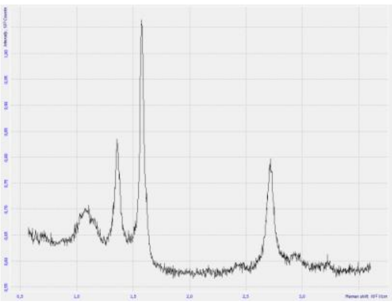

d

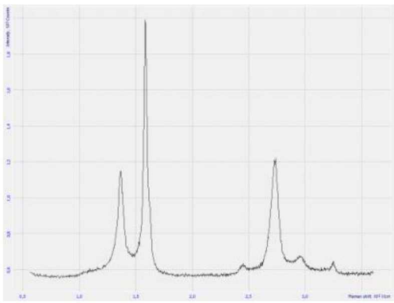
a – 0.75 (IG/I2D = 2.2);
b – 0.85 (IG/I2D = 1.8);
c – 0.95 (IG/I2D = 1.5);
d – 1.05 (IG/I2D = 1.9)

Fig. 14 – Raman spectra of the samples with a graphitized carbon black structure obtained on a nickel substrate in propane-oxygen-argon flames at different C/O


a

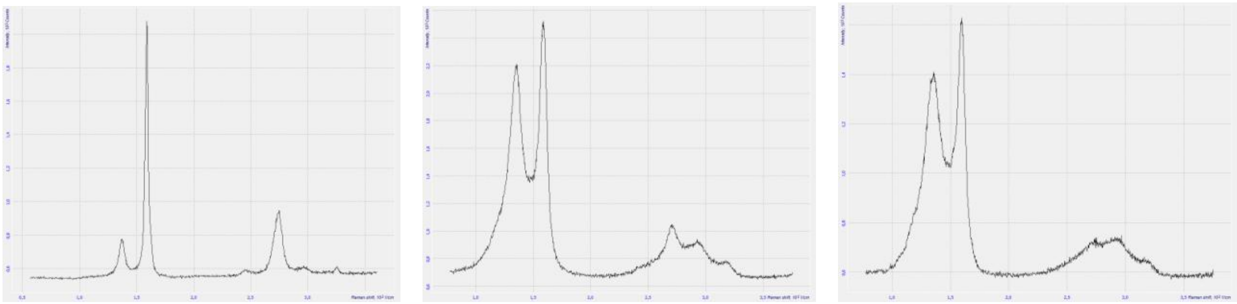

b


c


d

a – 0.75 (IG/I2D = 1.5);
b – 0.85 (IG/I2D = 2.0);
c – 0.95 (IG/I2D = 1.3);
d – 1.05 (IG/I2D = 1.6)

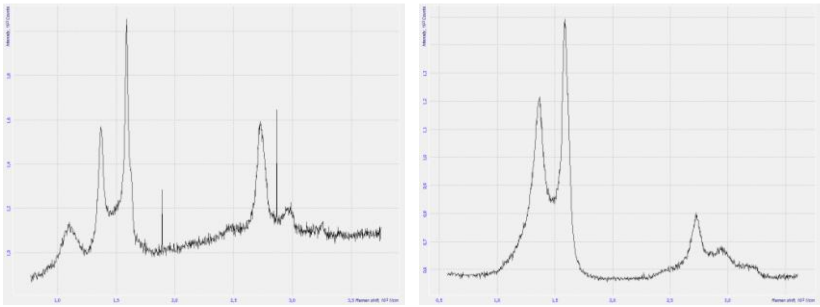
Fig. 15. Raman spectra of the samples with a graphitized carbon black structure obtained on a nickel substrate in the butane-oxygen-argon flames at different C/O



a b c

a – 150 cm3/min (IG/I2D = 2.2);
b – 350 cm3/min (IG/I2D = 2.4);
c – 500 cm3/min (IG/I2D = 2.2)

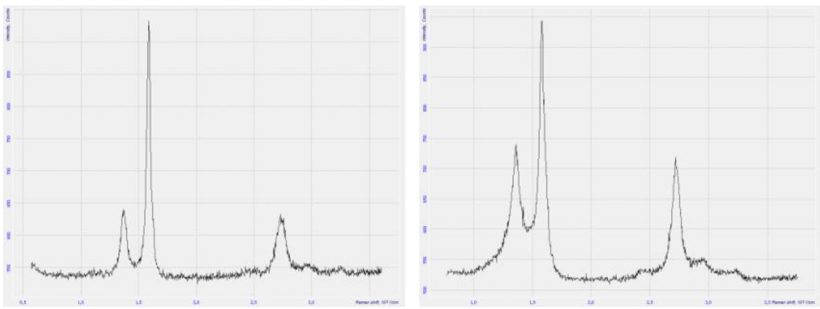
Fig. 16. Raman spectra of the samples with a graphitized soot structure obtained on a nickel substrate in propane-argon-oxygen flame at C/O = 0.75 at various argon flow



a b

a - 150 cm3/min (IG/I2D = 1.3);
b - 250 cm3/min (IG/I2D = 1.9)

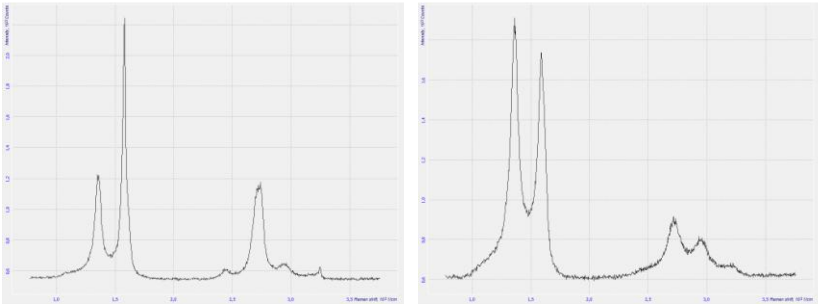
Fig. 17 – Raman spectra of graphene layers produced on a nickel substrate in a propane-oxygen-argon flame with C / O = 1.05 at different flow of argon



a b

a - 250 cm3/min (IG/I2D = 1.5);
b - 500 cm3/min (IG/I2D = 1.3)

Fig. 18 – Raman spectra of the samples with a graphitized carbon black structure obtained on a nickel substrate in the butane-oxygen flame with C/O = 0.75 at different flow of argon



a b

a - 250 cm3/min (IG/I2D = 1.9);
b - 500 cm3/min (IG/I2D = 1.9)

Fig. 19 – Raman spectra of the samples with a graphitized carbon black structure obtained on a nickel substrate in the butane-oxygen flame with C/O = 0.95 at different flow of argon

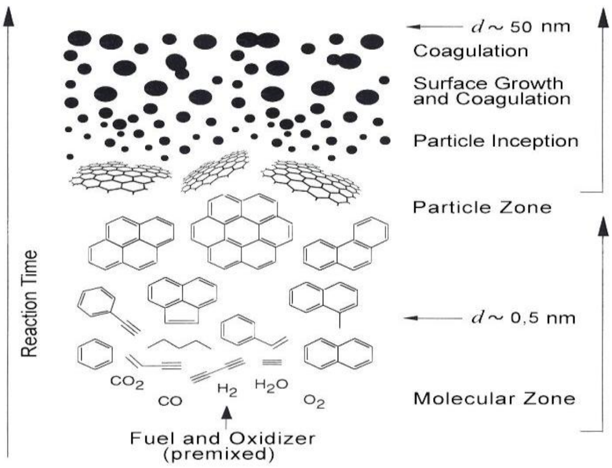


Fig. 20 – Modified scheme for soot and graphene formation process in flames

Conclusions

The results of the study on the formation of layered graphene films by flame combustion allow to making the following conclusions:

- It is possible to synthesize two or three layers of graphene in a premixed propane-oxygen-argon flame on a nickel substrate during the exposure time of 5 minutes;

- Formation of a minimum number of graphene layers (two or three layers, IG/I2D = 1.1-1.3) is possible in the range of the angle of inclination of the substrate from 0° to 30°;

- The further increase in the angle of inclination of the substrate relatively to vertical axis of the flame (over 30°) leads to the increase in the minimum number of graphene layers being formed on the substrate - from 5 to 10 (IG/I2D = 1.7-2.4);

- In the absence of argon, 5-10 graphene layers are usually formed on the substrate (IG/I2D = 1.6-2.5);

- The formation of graphene layers on the substrate occurs vertically in the flame during the transition to a soot structure.

- Formation of graphene layers occurs both in propane-oxygen and butane-oxygen flame in the investigated range of the ratio C/O: 0.75, 0.85, 0.95, 1.05;

- It is shown that mainly in the propane and butane flame under these conditions formation of graphene layers in the amount of 5-10 layers is observed;

- The tendency for increasing the degree of disorder of graphene layers with the increase in the ratio of C/O, which is characterized by the increased in the intensity of D-peak is observed;

- For propane-oxygen mixture, the degree of disorder is observed at a higher ratio of C/O than for butane-oxygen mixture;

- Formation of graphene layers shown to be effected by the consumption of argon supplied to the flame.

References

[1] Geim A.K. Uspehi Fiz. Nauk 181 (12) (2011).

[2] Ruess G., Vogt F. Monatshefte Chem. 78 222 (1948).

[3] Boehm H.P., Clauss A., Fischer G.O., Hofmann U.Z. Anorg. Allgemaine Chem. 316 119 (1962).

[4] Novoselov K.S., Geim A.K., Dubonos S.V., et al. Nature 426 812 (2003).

[5] Novoselov K.S., Geim A.K., Morozov S.V.,et al. Science 306 666 (2004).

[6] Geim A.K., Novoselov K.S. Nature Mater. 6 183 (2007).

[7] Eletsky A.V., Iskandarova I.M., Knizhnik A.A. Uspehi Fiz. Nauk, 181 (3), 233 (2011).

[8] Li Z., Zhu H., Xie D., et al. Chem. Commun., 47, 3520-3522 (2011).

[9] Memon N.K., Tse S.D., Al-Sharab J.F., et all Carbon 9 5064-5070 (2011).

[10] Bhaviripudi S., Jia X., Dresselhaus M.S., Kong J. Nano Lett 10:4128–33 (2010).

[11] Prikhodko N.G., Lesbayev B.T., Auyelkhankyzy M., Mansurov Z.A. et al. VII Intern. Symp. Phys. Chem. of Carbon Materials / Nanoengineering. Almaty (2012).

[12] Bockhorn H. (ed). Soot Formation in Combustion. Springer, Berlin/Heidelberg. 1994. 596.

[13] Mansurov Z.A. Intern. Symp. Modern challenges of higher education and science in the field of chemistry and chemical engineering. Almaty (2013), P. 14-19.

СИНТЕЗ СЛОЕВ ГРАФЕНА В ПЛАМЕНИ

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Аннотация

В статье представлены результаты синтеза графена в углерод содержащих пламенах. Были выбраны параметры горения, каталитический тип субстрата и вертикальная ориентация пламени, обеспечивающая образование самого малого числа слоев с большой структурной упорядоченностью . Установлено, что в углерод содержащем пламени на подложке из никеля формируется 5-10 слоев графена. Показано, что, когда угол наклонения подложки из никеля по отношению к вертикальной оси пламени от 0° до 30°, можно получить минимальное число слоев – 2-3 слоя. Исследовано образование слоев пленок графена в зависимости от соотношения С/О, количества аргона, введенного в пламя, и от типа топлива. Образование слоев графена исследовано в широком интервале значений С/О = от 0.75 до 1.05. Показано, что с увеличением значений С/О и количества аргона введенного в пламя, формирование слоев графена увеличивается.

ЖАЛЫНМЕН БАҒАНДА ГРАФЕНҚАБАТТАРЫНЫҢ СИНТЕЗІ

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Аннотация

Қағаздыдайын қоспасы көмірсутек жалын граффен қабыршақтардың синтездеу нәтижелерін ұсынады. Жану параметрлері, каталитикалық субстрат түрі таңдалған және жалын тік бағдар, ең үлкен құрылымдық тұрақты қабаттарының ең кішкентай санының білуге болды. Никель ұлтанды көмірсутегі жалын негізінен 5-10 бағанында қабаттарын қалыптасады деп табылды. 2-3 қабаттар - Ол 0 ° 30 ° дейін жалын тік осіне субстрат қатысты көлбеу бұрышы қабаттарының санын төменгі алуға болады, бұл кезде көрсетіледі. Отын түріне байланысты қабатты граффен қабыршақтардың қалыптастыру жөніндегі қатынасы С / О және аргон ағынының әсері туралы зерттеулер. Граффен қабаттарының қалыптасуы 1.05 үшін С / О = 0,75 мәндер кең ауқымды зерттелген. Ол граффен бұзылуы артып қабаттарын қалыптастыру, бұл С / О және жалын енгізілген аргон сомасының жылыҚатпарсыз құндылықтармен көрсетіледі.