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PROCESSES OF TRANSFER IN A LOW-TEMPERATURE PLASMA

COMPARISON OF THE CHARACTERISTICS OF A PLASMA GENERATED BY A D.C. ARC PLASMATRON AND A HIGH-VOLTAGE A.C. PLASMATRON WITH RAIL-SHAPED ELECTRODES (GLIDING ARC) AS APPLIED TO THE SYNTHESIS OF CARBON NANOMATERIALS

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Comparative results of investigating the parameters of plasmas generated by atmospheric-pressure electric-arc plasmatrons of two types operating in the regime of simultaneous partial oxidation of hydrocarbons and their pyrolysis, have been given. It has been established by spectral and thermophysical measurements that the component composition of the plasma and its thermal characteristics at exit from a d.c. plasmatron significantly differ from the parameters of a plasmatron of the second type — an a.c. plasmatron with rail-shaped electrodes of the gliding-arc type. The temperature nonequilibrium and the presence of the carbon dimers C_2 *in the plasma (in the absence of the monomer C) generated by the rail-shaped-electrode plasmatron point to the fact that realization of the synthesis of fullerene-containing particles and, probably, particles containing nanotubes is, in principle, possible in reactors based on it, just as in more energy-intensive reactors with low-voltage d.c. arc plasmatrons.*

Low-temperature plasmas of C–N, C–H–O, and C–H–O–N systems, which are generated based on hydrocarbons, have actively been studied in the recent decade in developing technologies for such fields as hydrogen power engineering (production of H_2, synthesis gas, and C_2H_2) and the synthesis of carbon nanomaterials in reactors implementing pyrolysis or partial oxidation of alkanes [1–3]. In creating hydrocarbon-gas plasmatrons and technological apparatuses (reactors, furnaces) based on them, one must have a prescribed chemical (component) composition of the plasma. In this work, we give certain results of spectral and thermophysical measurements of determination of the component composition of plasma jets generated, in different ways, from air-propane-butane mixtures in the regime yielding carbon oxide. The prime objective of investigation is to extend the applicability range for a plasma of this kind in devices intended for synthesis of carbon nanomaterials.

We used two types of atmospheric-pressure arc plasmatrons as plasma generators: 1) a d.c. plasmatron of a linear circuit, tested for pyrolytic synthesis of carbon nanotubes and fullerenes [4, 5] (we selected a version with sleeve copper electrodes of the PD-1M type developed at the Heat and Mass Transfer Institute [6]) and 2) a high-voltage a.c. plasmatron with rail-shaped electrodes of the gliding-arc type (sometimes called a railtron in the literature) [7–9].

From physicochemical considerations, the processes of plasma synthesis of carbon nanomaterials from a hydrocarbon raw material are subdivided into two trends [1, 3] under the conditions of: a) a purely pyrolytic process with the endothermic dissociation of alkanes in an oxygen-free plasma based in inert gases or nitrogen; b) partial oxidation of the raw material by reactions of the type

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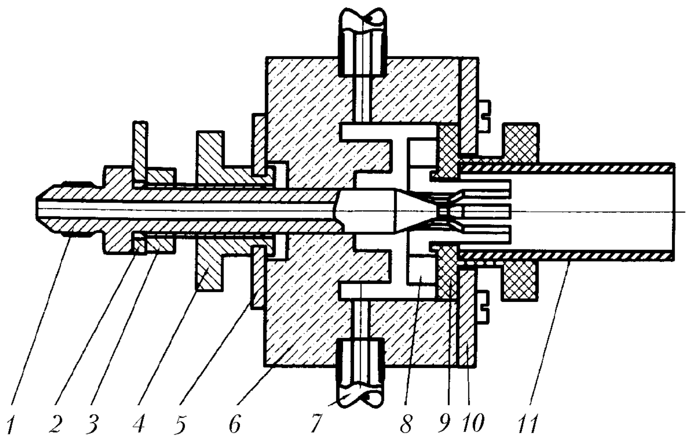

Fig. 1. High-voltage a.c. arc plasmatron with rail-shaped electrodes (gliding arc) of the S-1 type (version), developed at the Heat and Mass Transfer Institute: 1) unit for injection of a plasma-generating gas; 2, 5, 9) insulating pads; 3) fixing nut; 4) fastening nut; 6) casing; 7) union for injection of a stabilizing gas; 8) electrodes; 10) flange; 11) tubular nozzle.

$$C_3H_8 + 1.5O_2 \rightarrow 3CO + 4H_2 \quad (\Delta H^0 = -227.7 \text{ kJ}) \tag{1}$$

or in a combined pyrolytic-oxidation process with the contribution of exothermic reactions in an oxygen-containing plasma. The first of these cases is characterized by the formation of such intermediate products, precursors to multiwalled carbon nanotubes and fullerenes, as unsaturated hydrocarbons of the C_2H_2 type. In the second case we will have CO molecules — precursors to the catalytic formation of single- and multiwalled carbon nanotubes — in the reaction zone (the disproportion (Boudoir) reaction $2CO \leftrightarrow CO_2 + C$ is known to be essential to the chemism of formation of carbon nanotubes).

In this work, we give results of investigations with the use of a mixture of compressed air and propane-butane of the SPBT grade according to the All-Union State Standard 20448-90 with the following composition (vol.%): CH_4 — 0.6, C_2H_6 — 5.6, C_3H_8 — 72.5, *n*-butane — 12.2, and isobutane — 9.0; the mean molecular weight was 47.2 g/mole. This mixture was fed to the plasmatron via an RS-5 flowmeter or an RM rotameter corresponding to the All-Union State Standard 13045-67).

The experiments were performed at atmospheric pressure (P = 0.1 MPa) on two plasma units (on the pilot benches of the Heat and Mass Transfer Institute): the diagram of the first unit (with a d.c. arc plasmatron) has been given in [4], whereas the diagram of the second one (with an a.c. plasmatron with rail-shaped electrodes) was coincident with the standard for such devices [9, 10]. In preparation for the experiment, we developed and manufactured laboratory plasmatrons ("railtrons") of two modifications — single-phase and three-phase ones (see Fig. 1 for the general view of the latter) — with a power of up to 1 kW. The main volume of the experiments was carried out with the single-phase structure without an external coolant loop; the structure operated off an industrial a.c. supply line via a special high-voltage transformer.

The optical-emission-spectroscopy method widely used for diagnostics of a low-temperature plasma, including a carbon-containing one, was employed for determination of the component composition of the plasma and its electron temperature [11–17]. Spectral measurements were carried out using an SL40 spectrometer enabling us to record the radiation of plasma jets in the range of wavelengths λ = 210–900 nm with the use of diffraction gratings with a density of up to 1200 bars/mm. Also, we performed differential calorimetric measurement of the plasmatrons on the basis of the stationary-heat-flux method. We measured the electrical parameters of the plasmatrons: the voltage and current on the arc, the mass flow rate and temperature of water fed for cooling the elements of the unit, and the flow rates of plasma-generating gases. The mass-mean enthalpy of the plasma at exit from the plasmatron nozzle and its mean "frozen" (without allowance for the thermal effects of chemical reactions) temperature were determined from these measurements [18]. In the case of the a.c. plasmatron with rail-shaped electrodes we evaluated the dynamics of change in

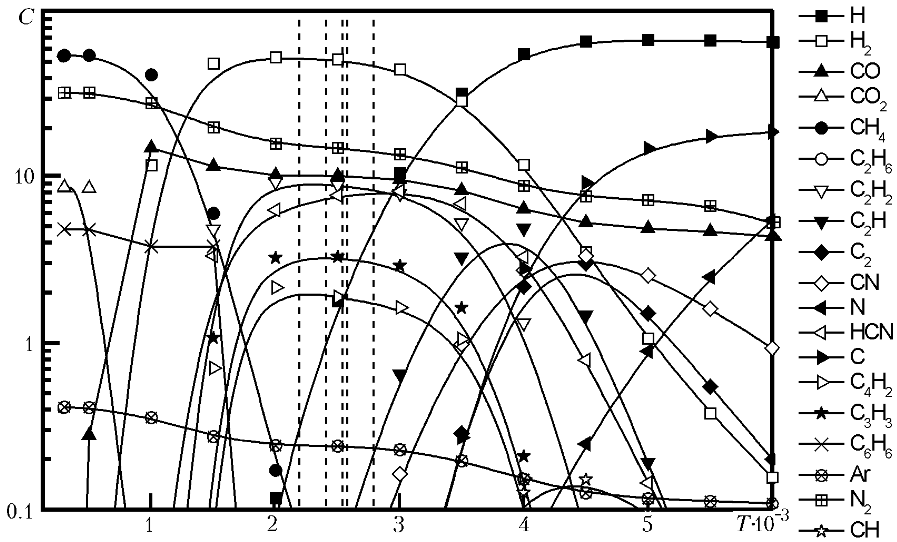

Fig. 2. Quasiequilibrium composition of the C–N–O–H system (without allowance for the formation of condensed carbon) for the component relation that corresponds to the air-propane mixture with the flow-rate ratio corresponding to regime Nos. 26–30, Table 1 (calculation); dashed lines, mass-mean temperatures in regime Nos. 26–30 (experiment).

the electrical parameters of the arc with time by their oscillography (in small-load regimes with level of power consumed by the arc lower than 5% of the transformer power) on the low-voltage side of the transformer with monitoring of the running value of voltage on its high-voltage side by an S169 kilovoltmeter.

Selection of propane-butane as the raw material was presubstantiated both in the context of thermodynamics [19] and from the viewpoint of its commercial availability. We note that the relation of the mass flow rate of propane-butane to the flow rate of air in the experiments was $G_{pr-b}/G_a > 1.0$ in both kinds of plasmatrons, which corresponds to the mixed mechanism region: to the simultaneous partial oxidation of hydrocarbons and their pyrolysis (realization of the mechanism of "pure" partial oxidation of a propane-butane raw material according to the stoichiometry of reactions of the (1) type requires the relation $G_{pr-b}/G_a \approx 0.2$, whereas the relation $G_{pr-b}/G_a \approx 0.06$ is needed to completely oxidize the raw material to CO_2 and H_2O). The reason was that, in most existing plasma apparatuses for synthesis of carbon nanomaterials (with chromatographic monitoring of the composition of the gas [1, 19]), the G_{pr-b}/G_a relation necessary for partial oxidation in actual fact is much higher than 0.2 because of the axial and radial nonisothermicity in the plasmatron channel and mass-exchange constraints. Another reason was that in the specific case of the high-voltage a.c. plasmatron with rail-shaped electrodes in use its operation with a mixture of propane-butane and air is the most stable precisely in the region of the relation $G_{pr-b}/G_a \geq 1.0$.

Investigation Results. Figure 2 exemplifies the thermodynamically quasiequilibrium composition of a C–N–O–H system (calculation by the method of minimization of the total isobaric-isothermal potential using the CHEMKIN program with the constant base Chem05VERS) for the component relation corresponding to an air-propane mixture with a ratio of $G_{pr-b}/G_a = 1.15$; this ratio corresponds to experimental regimes of operation of the plasmatrons (Nos. 26–30, Table 1).

The operating parameters in the experiments with a d.c. plasmatron and a high-voltage plasmatron are given in Tables 1 and 2. Tables 3 and 4 show the typical spectra of the plasma based on the data of [11–17].

A comparison of the spectral composition of the plasma of a mixture of propane-butane with air on the d.c. plasmatron investigated (see spectra in Fig. 3) and the thermodynamic calculations modeling the regimes of experiment Nos. 15, 25, and 30 (heating of C_3H_8 + 19 wt.% or 45–47 wt.% of air to 4000 K respectively) has shown that hydrogen, nitrogen, carbon (similarly to the composition of nitrogen-propane plasma jets [19]), and partially nitrogen-hydrogen-carbon compounds and CO are the most probable atomic-molecular components of the plasma in both cases. From the thermodynamic data for the regime of experiment Nos. 15, 25, and 30 at T = 4000 K, the mixture is enriched to the greatest extent with the components H (60.2 and 54.8–55.3 vol.%), H_2 (14.2 and 11.8–12 vol.%), N_2

TABLE 1. Experimental Parameters of Operation of a PD-1M D.C. Arc Plasmatron by a Mixture of Propane-Butane with Air

Regime No.	I, A	U, V	$U{\cdot}I$, kW	G_a, g/sec	$G_{pr\text{-}b}$, g/sec	$G_{pr\text{-}b}/G_a$	H, MJ/kg	T_g, K
11	130	290	37.70	1.80	3.58	1.99	3.62	1490
12	130	310	40.30	1.68	2.86	1.70	4.81	1850
13	140	270	40.18	1.40	3.43	2.45	4.26	1620
14	145	240	34.80	1.24	4.44	3.58	2.90	1210
15	150	230	34.5	1.16	4.86	4.19	2.70	1140
16	165	170	28.05	1.00	5.15	5.15	1.61	1000
17	135	295	39.83	1.80	3.58	1.99	4.01	1600
18	140	290	40.60	1.80	3.58	1.99	4.20	1650
19	160	280	44.80	1.80	3.58	1.99	5.00	1860
20	165	280	46.2	1.80	3.58	1.99	5.20	1910
21	150	230	34.50	1.24	4.44	3.58	2.90	1210
22	160	225	36.00	1.24	4.44	3.58	3.20	1290
23	180	190	34.20	1.24	4.44	3.58	2.84	1190
24	120	400	48.00	2.21	2.86	1.29	5.87	2240
25	105	405	42.52	2.37	2.86	1.21	4.66	1930
26	115	400	46.00	2.37	2.72	1.15	5.50	2190
27	130	385	50.05	2.37	2.72	1.15	6.29	2410
28	140	375	52.5	2.37	2.72	1.15	6.77	2540
29	150	355	53.32	2.37	2.72	1.15	6.91	2580
30	165	350	57.75	2.37	2.72	1.15	7.75	2790

TABLE 2. Experimental Parameters of Operation of a High-Voltage Single-Phase Arc Plasmatron with Rail-Shaped Electrodes ("Railtron") by Air and by a Mixture of Propane-Butane with Air

Regime No.	U, kW	I, A	$U{\cdot}I$, W	G_a, g/sec	$G_{pr\text{-}b}$, g/sec	$G_{pr\text{-}b}/G_a$
1	1.8	0.045	81.0	3.25	0	0
2	1.5	0.047	70.5	2.03	0	0
9	1.8	0.018	32.4	3.56	0	0
14	2.3	0.042	96.6	5.08	0	0
15	1.0	0.028	28.0	6.60	0	0
16	2.0	0.036	72.0	6.60	0	0
17	1.7	0.016	27.2	1.48	0	0
28	2.0	0.050	100.0	1.73	2.6	1.5
29	2.3	0.050	115.0	1.73	2.1	1.2

(1.88 and 8.11–8.81 vol.%), C_2H (7.9 and 4.94–5.18 vol.%), HCN (2.03 and 3.27–3.31 vol.%), CN (1.53 and 2.69–2.75 vol.%), C (3.39 and 2.81–2.87 vol.%), and C_2 (3.21 and 2.2–2.29 vol.%). From the spectral data, a band characteristic of H_2, C_2, CN, and Cu with a center at 516.4 nm has the maximum relative intensity (more than 4100 rel. units). Also, there are two strong lines of H atoms (with an intensity of up to 4020 rel. units at 656.1 nm). In comparing to the thermodynamically expected composition, this suggests that temperatures no lower than 3500–4000 K, i.e., 1000 to 2000 K higher than the mass-mean temperatures of the "frozen" mixture of air with propane, which have been calculated by calorimetric measurement of the plasmatron, are attained in the regimes of generation of air-propane-butane plasmas (see Table 1). Also, we observe no less than two pronounced C_2 bands (with an intensity of up to 940 rel. units; an additional contribution of C monomers to the band at 467–473.7 nm is possible, whereas there is, apparently, a contribution of CO molecules to the band with a center at 563.5 nm) and an explicit C-monomer band at a wavelength of 248 nm (in the spectrum of regime No. 25 (Fig. 3b)). It is important that Anazava et al. [11] observed the presence of this C line precisely in a CO-containing plasma, where soot with a content of 95% of multi-walled carbon nanotubes in it was synthesized (with application of a magnetic field to the plasmatron arc).

TABLE 3. Spectral Characteristics of an Arc Air Plasma on the Section of a High-Voltage Plasmatron ("Railtron") in Regime No. 2 (see Table 2)

Characteristic peaks λ, nm	Intensity i, rel. units	Identification of lines and bands according to [11–17, 20] (λ, nm)
222.1	41.4	Cu (222.6 nm)
226.4	45.0	NO, γ-system
236.3	47.6	(226.9; 236.3; 247.1 nm)
247.0	43.4	—
296.6	39.9	—
314.1	54.7	—
316.2	59.0	N_2 (316.0 nm)
325.2	39.6	Cu (324.8 nm) or N_2^+ 1^- system
327.6	37.8	Cu (327.4 nm) or N_2^+ 1^- system
337.3	88.7	N_2 (337.1 nm)
353.8	45.3	—
357.8	58.0	N_2 (357.7 nm)
375.8	39.6	—
380.5	39.1	N_2 (380.5 nm)
391.7	41.2	N_2^+ (391.4 nm)
776.7	38.6	O (777.1 nm)

TABLE 4. Spectral Characteristics of the Arc Plasma of a Mixture of Propane-Butane with Air on the Section of a High-Voltage Plasmatron ("Railtron") in Regime Nos. 28 and 29 (see Table 2)

Characteristic peaks λ, nm		Intensity, rel. units		Identification of lines and bands according to [11–17, 20] (λ, nm)
regime No. 28	regime No. 29	regime No. 28	regime No. 29	
307.4	—	98.04	—	OH (309.0 nm) in the range 306–312 nm or CO (310 nm)
309.7	309.7	132.46	108.38	
337.3	—	78.44	—	N_2 (337.1 nm) or O_2 (337.0 nm)
429.3	429.3	99.98	90.54	The radical CH (431.4 nm) is possible
468.1	468.1	92.12	97.9	The Swan C_2 band in the range 468–473.7 nm; a contribution of the C line (467.9 nm) is possible
469.3	469.3	91.62	91.28	
471.0	471.0	90.08	90.92	
473.3	473.3	93.78	93.16	
512.6	512.6	281.18	237.3	The C_2 band with peaks at 510.0, 513.0, and 516.5 nm; a contribution of the CN band (515.6 nm) is possible
516.0	516.0	499.24	408.7	
553.9	—	82.6	—	The C_2 band with peaks at 546.5 and 563.5 nm; a contribution of the OH (553.4 nm), CO (553.2 nm), and NO_2 (553.4 nm) bands is possible
558.4	558.4	104.14	102.3	
563.4	563.4	109.54	105.58	
589.1	589.1	144.86	129.74	—
	612.6		82.7	—
766.3	766.3	203.54	234.28	Cu^+ (766.5 nm)
769.8	769.8	119.58	140.66	—

In the regimes of generation of the plasma of the mixture of propane-butane with air for a nearly 1.0 relation of their flow rates, we also note the presence of oxygen atoms, i.e., a line with intensity up to 369 rel. units in the region of λ at 616 nm and 777 nm in the spectra, just as in the case of a purely air plasma. Atomic oxygen is usually

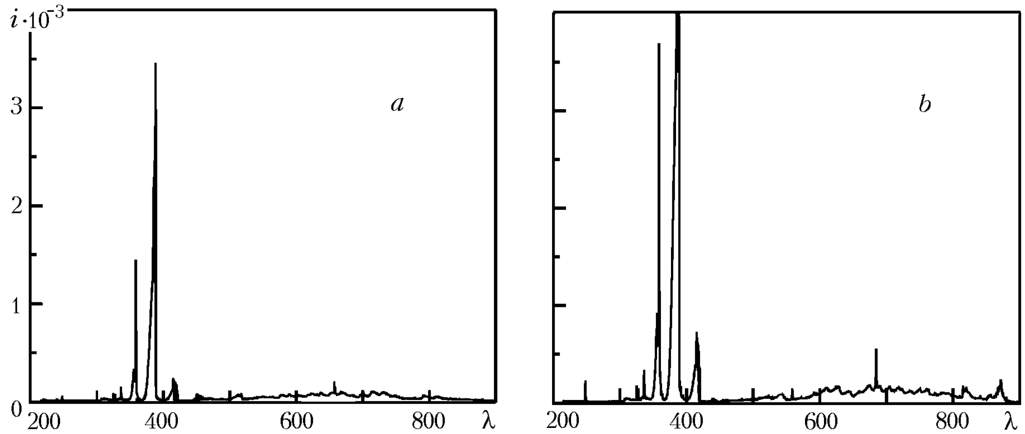

Fig. 3. Emission spectrum of a plasma jet generated from a mixture of propane with air by a PD-1M d.c. arc plasmatron in regime Nos. 11 (a) and 25 (b) according to Table 1; averaged over a period of 1.4 sec.

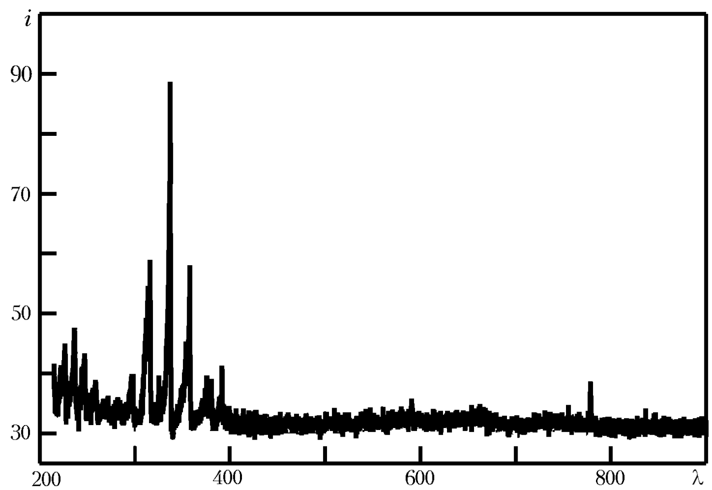

Fig. 4. Emission spectrum of an air-plasma set generated by a rail-shaped-electrode plasmatron (gliding arc) in regime No. 2 according to Table 2; averaged over a period of 1.4 sec.

formed by the Zel'dovich mechanism [12, 14]: $N + O_2 \leftrightarrow (NO + O) \leftrightarrow NO_2 + h\nu$ due to the beginning of the mixing of the jet with a cold ambient air. Such a situation was spectrally recorded by Gallimore et al. [14] in mixing of the jets of a nitrogen plasma with air. It is noteworthy that the thermodynamically equilibrium composition of an air plasma at 4000 K is as follows: O — 26.2 vol.%, O_2 — 3.09 vol.%, N — 0.14 vol.%, NO — 4.13 vol.%, and N_2 — 65.6 vol.%. Furthermore, a certain decrease in the intensity of C and C_2 lines with increase from 105 to 165 A in the current strength and from 42.5 to 58 kW in the power (all other parameters being equal) has been noted in the regimes of this series with a PD-1M plasmatron, which may suggest the reduction in the concentration of these components in the plasma-jet cross section under recording.

The typical spectra of the plasma (i.e., its component composition) and its thermal characteristics at exit from a plasmatron of the second type, an a.c. plasmatron with rail-shaped electrodes of the gliding-arc type (see Tables 2–4 and Figs. 4 and 5), significantly differ from the above-described parameters of the plasma generated by a d.c. plasmatron. Examples of the types of "railtron" in the device and the oscillograms of its current and voltage are shown in Figs. 6 and 7 and demonstrate its strongly nonstationary operation (the pause of the current attains 55% in a number of regimes, which is indirectly related to the temperature nonequilibrium of the plasma). It is important that, from the data of calorimetric measurement, the mass-mean temperature at exit from the "railtron" is within 330–700 K (see Table 2 for low powers and high flow rates of plasma-generating gases). Interpretation (performed with allowance for

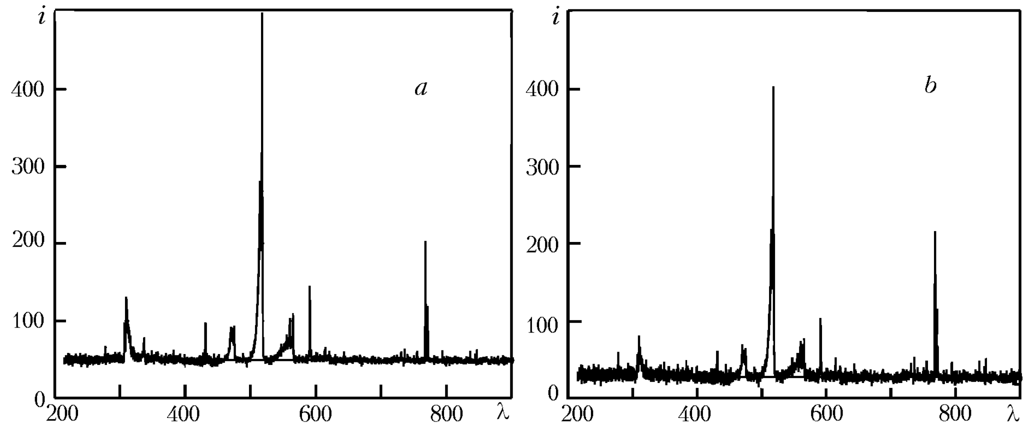

Fig. 5. Emission spectrum of a plasma generated from a mixture of propane with air by a rail-shaped-electrode plasmatron (gliding arc) in regime Nos. 28 (a) and 29 (b) according to Table 2; averaged over periods of 9.8 and 14.7 sec respectively.

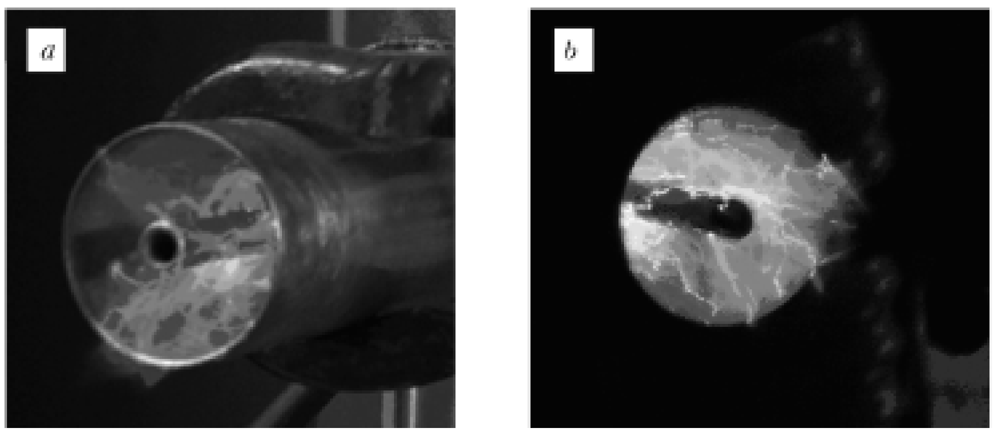

Fig. 6. Illustration of the operation of a single-phase railtron by an air plasma (a) and by an air-propane-butane mixture (b).

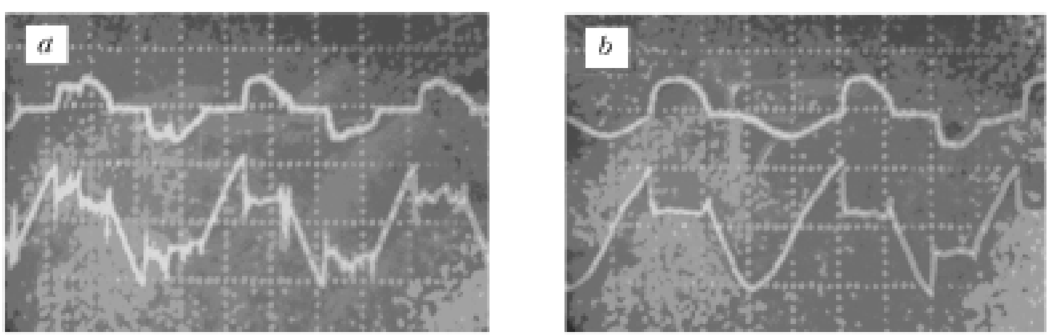

Fig. 7. Oscillograms of the current strength (upper curves) and voltage (lower curves) in operation of a single-phase railtron by an air plasma in regime (according to Table 2) Nos. 9 (a) and 17 (b). The scale along the ordinate axis (with allowance for the transformation ratio k = 100) is equal to 0.10 A/square and 5000 V/square; the (time) scale along the abscissa axis is 0.005 sec/square.

TABLE 5. Experimental Electron T_e (from the Spectral Data) and Mass-Mean T_g Temperatures of the Air and Nitrogen Plasmas Generated by a D.C. Arc Plasmatron

No. of operating regime of the PD-1M plasmatron	T_g from the thermal balance of the plasmatron, K	T_e from the pair of 510.6 and 515.3 nm CuI lines, K	T_e from the pair of 510.6 and 521.8 nm CuI lines, K	Average spectral T_e, K
5 (air)	6250	6630	6450	6540
31 (nitrogen)	5770	6960	6840	6900
34 (nitrogen)	5410	6760	6620	6690

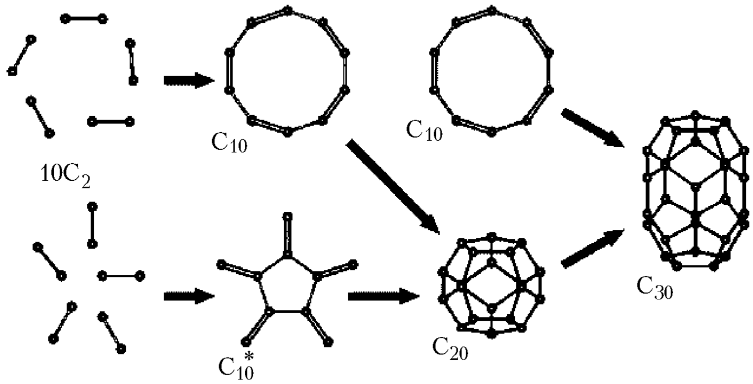

Fig. 8. Probable mechanism of formation of fullerenes in an arc plasma according to the scheme of [22, 23].

[11–17, 20]) of the spectra of the plasmas of air (see Table 3) and a propane-butane + air mixture for the "railtron" shows the presence of such components as N_2^+ (lines at 380.5 and 391.4 nm in the air plasma) and Cu^+ (766.5 nm line in the air-propane-butane plasma) which are nonrepresentative for an ordinary arc plasma (see above data for the PD-1M plasmatron). The most important (including the practical aspect) difference of the "railtron" plasma from an equilibrium one is the presence of the carbon dimer C_2 (identified by three explicit Swan bands in the regions 468–473.7 nm, 510–516.5 nm, and 546.5–563.5 nm) in it as one basic component with a simultaneous high probability that the gaseous monomer C is absent and with no pronounced lines of atomic or molecular hydrogen (it is most likely present in bound form as OH radicals). The nonequilibrium of this kind of plasma manifests itself as both a variation of the chemical composition from that thermodynamically expected at these mass-mean temperatures (T_g = 330–700 K) (see Fig. 2) and a substantial excess of the electron temperature T_e (being at a level no lower than 1000–1500 K, from the data for analogous high-voltage plasmatrons [7, 9, 20] and according to our spectral measurements) over the indicated level of mass-mean temperature.

Table 5 compares, as an example, the experimental values of the electron T_e (calculated from the data of spectral measurements by the standard procedure [21]) and mass-mean T_g (determined from the thermodynamic data from the mass-mean calorimetric enthalpy) temperatures of air and nitrogen plasma jets generated by a d.c. arc plasmatron. The difference $\Delta T = T_e - T_g$ is within 290–1200 K, which confirms the well-known fact that this kind of plasma, unlike the nonequilibrium plasma of a high-voltage plasmatron (gliding arc), belongs to the class of quasiequilibrium plasmas satisfying the LTR approximation [9, 18].

Investigation of the parameters of the C–N–O–H-system's plasma generated by atmospheric-pressure electric-arc plasmatrons of two types operating in the regime of simultaneous partial oxidation and pyrolysis of propane-butane has enabled us to reveal the following fact. The composition of the carbon-containing plasma and its thermal characteristics at exit from a d.c. plasmatron significantly differ from the parameters of a plasmatron of the second type — an a.c. single-phase one with rail-shaped electrodes (gliding arc).

In accordance with the model of formation of fullerenes after Heath [22] and with the scheme of Poklonskii et al. [23], there is a relationship between the presence of carbon dimers in the plasma and a probable growth of fullerenes in the reaction zone. According to this model, carbon clusters become fullerenes with 30 to 40 atoms in a cluster and they grow further due to the embedding of C_2 dimers. The scheme of [23] explains the formation of the intermediate cluster C_{20} (key in the model of [22]) and its subsequent growth to higher fullerenes by coalescence of C_{10} rings (see Fig. 8). The formation of the metastable cluster C_{10}^{*} in the plasma is a limiting stage since, once C_{20} has been formed, its growth due to the addition of C_{10} is very rapid in the arc plasma. We note that if there is no isolated direction in the plasma, a fullerene ceases to grow on C_{60} and C_{70}; if the direction is created (due to the special electrode shape or to the application of electromagnetic fields), carbon nanotubes with a diameter no less than that in C_{20} particles may subsequently be formed.

Thus, when we use a plasmatron of the second (of those investigated) type, i.e., the so-called "railtron," characterized by a lower mass-mean temperature, a small energy contribution to the raw material compared to ordinary low-voltage arc plasmatrons, and the thermal nonequilibrium of the plasma, the synthesis of fullerene-containing particles and nanotubes can, in principle, be realized because of the presence of a high concentration of C_2 dimers in the flow. This, in particular, is possible for such parameters as a pressure of 10^5 Pa and a rate of quenching of a carbon-containing gas of 10^5–10^6 K/sec in condensation. Another evidence in favor of this fact is that we carried out earlier [19] the synthesis of carbon nanostructures (with a content of fullerenes of up to 15 wt.%) in the plasma of a mixture of nitrogen and propane-butane; this plasma was generated by a PD-1M low-voltage plasmatron and the set of lines of the carbon dimer C_2 in its emission spectra was characterized by the highest intensity, too.

NOTATION

C, concentration, vol.%; *G*, flow rate of the gas, g/sec; *h*, Planck constant; *H*, specific enthalpy, kJ/kg; *i*, intensity, rel. units; *I*, current strength, A; *k*, transformation ratio of the NOM-10-66 high-voltage transformer; *P*, pressure, Pa; *T*, temperature, K; *U*, voltage, V; $U \cdot I$, plasmatron power, kW; ΔH^0, thermal effect of the reaction under standard conditions; λ, plasma-radiation wavelength, nm; ν, electromagnetic-radiation frequency, Hz. Subscripts: e, electron; g, gas; a, air; pr-b, propane-butane.

REFERENCES

1. A. Huczhko, H. Lange, and G. Cota-Sanchez, Plasma synthesis of nanocarbons, in: *Proc. 7th Int. Conf. on Thermal Plasma Processing* (TPP-7), France (2003), p. 617.
2. M. Deminsky, V. Jivotov, B. Potapkin, and V. Rusanov, Plasma-assisted production of hydrogen from hydrocarbons, *Pure Appl. Chem.*, **74**, No. 3, 413–418 (2002).
3. É. G. Rakov, Methods for obtaining carbon nanotubes, *Usp. Khim.*, **69**, No. 1, 41–60 (2000).
4. A. V. Gorbunov, A. F. Bublievskii, A. L. Mossé, et al., Synthesis of carbon nanomaterials in pyrolusis of hydrocarbons in an atmospheric-pressure plasma-chemical arc reactor, in: *Proc. III Int. Symp. "Fullerenes and Fulleren-like Structures in Condensed Media"* [in Russian], ITMO, Minsk (2004), pp. 118–120.
5. A. V. Eletskii, A. F. Pal, N. N. Dzbanovsky, and N. V. Suetin, Plasma methods of carbon nanotube synthesis, in: *Proc. 4th Int. Conf. on Plasma Physics and Plasma Technology*, Minsk (2003), pp. 926–929.
6. A. L. Mossé, A. V. Gorbunov, and E. M. Ermolaeva, *Plasma Processing of Toxic Organic and Halogen-Containing Waste. Problems and Possibilities of Contamination* [in Russian], Preprint No. 2 of the A. V. Luikov Heat and Mass Transfer Institute, National Academy of Sciences of Belarus, Minsk (2003).
7. A. Czernichowski, GlidArc assisted preparation of the synthesis gas from natural and waste hydrocarbons gases, *Oil Gas Sci. Technol. — Rev. IFP*, **56**, No. 2, 181–198 (2001).
8. M. F. Zhukov, E. I. Karpenko, V. S. Peregudov, et al., Plasma oil-free firing of boilers and stabilization of combustion of a coal-dust torch, in: *Low-Temperature Plasma* [in Russian], Vol. 16, Nauka, Novosibirsk (1995).
9. A. Czenichowski, Gliding arc. Applications to engineering and environment control, *Pure Appl. Chem.*, **66**, No. 6, 1301–1310 (1994).
10. F. Richard, J. M. Cormier, S. Pellerin, and J. Chapelle, Physical study of a gliding arc discharge, *J. Appl. Phys.*, **79**, No. 5, 2245–2250 (1996).
11. K. Anazava, R. Shimotani, C. Manabe, H. Watanabe, and M. Shimizu, High-purity carbon nanotube synthesis method by an arc discharging in magnetic field, *Appl. Phys. Lett.*, **81**, No. 4, 739–741 (2002).

12. J. L. Prebola, Jr., Performance of a Plasma Torch with Hydrocarbon Feedstocks for Use in Scramjet Combustion, Ms. Sci. Thesis, Virginia Polytechnical Inst., USA (1998).
13. S. Hofmann, B. Kleinsorge, and C. Ducati, Low-temperature plasma enhanced chemical vapor deposition of carbon nanotubes, *Diamond Relat. Mater.*, **12**, 1–6 (2003).
14. S. D. Gallimore, L. S. Jacobsen, and W. F. O'Brien, Spectroscopic investigations of hydrocarbon- and nitrogen-based plasmas for supersonic ignition, AIAA Paper, 2002–5247 (2002).
15. R. Yano, V. Contini, E. Plonjes, et al., Supersonic nonequilibrium plasma wind-tunnel measurements of shock modification and flow visualization, *AIAA J.*, **38**, No. 10, 1–8 (2000).
16. M. A. Elliott, P. W. May, J. Petherbridge, et al., Optical emission spectroscopic studies of microwave enhanced diamond CVD using CH_4/CO_2 plasmas, *Diamond Relat. Mater.*, **9**, 311–316 (2000).
17. J. Petherbridge, P. W. May, S. R. J. Pearce, et al., Molecular beam mass spectrometry investigations of low temperature diamond growth using CO_2/CH_4 plasmas, *Diamond Relat. Mater.,* **10**, 393–398 (2001).
18. A. Ambrazyavichyus, Heat transfer in quenching of gases, in A. Zhukauskas (Ed.), *Thermal Physics* [in Russian], Vol. 15, Mosklas, Vilnius (1983).
19. A. F. Bublievskii, A. A. Galinovskii, A. V. Gorbunov, S. A. Zhdanok, L. I. Sharakhovskii, and A. L. Mossé, Plasma pyrolytic synthesis of carbon nanostructures in a mixture of nitrogen and propane–butane, *Inzh.-Fiz. Zh.*, **79**, No. 2, 3–9 (2006).
20. V. Chernyak, V. Naumov, and V. Yukhimenko, Spectroscopy of atmospheric pressure air jet plasma in transverse arc discharge, in: *Proc. 12th Int. Congress on Plasma Physics*, Nice, France (2004).
21. J. E. Sansonetti and W. C. Martin, *Handbook of Basic Atomic Spectroscopic Data*, London (1995).
22. J. R. Heath, Synthesis of C_{60} from small carbon clusters: a model based on experiment and theory, in: G. S. Hammond and V. J. Kuck (Eds.), *Fullerenes: Synthesis, Properties and Chemistry of Large Carbon Clusters*, ACS, Washington, DC (1991), pp. 1–23.
23. N. A. Poklonskii, O. N. Bubel', E. F. Kislyakov, and S. A. Vyrko, A possible way for formation of fullerenes and carbon nanotubes, in: *Ext. Abstr. of Papers presented at II Int. Symp. "Fullerenes and Fullerene-Like Structures in Condensed Media"* [in Russian], ITMO, Minsk (2002), pp. 190–191.