

HEAT TRANSFER IN COMBUSTION PROCESSES

INFLUENCE OF THE ELECTRIC FIELD ON THE SOOT FORMATION IN THE FLAME AT A LOW PRESSURE

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UDC 661.666.14

We have investigated the influence of an electric field in the form of a gas discharge on the flame pattern, the radiographic parameters of soot particles, and the yield of fullerenes and polycyclic aromatic hydrocarbons. It has been shown that an external electric field permits controlling the processes proceeding in the flame, as well as the soot formation, and even increasing the soot yield under certain conditions. It has been established that the height of the soot packet L_c and the interplanar spacing d_{002} remain unchanged with increasing voltage, which is confirmed by the absence of graphitization under the action of the electric field. It has been revealed that negative polarity has a stronger effect on the increase in the yield of fullerenes with increasing voltage applied compared to positive polarity.

Keywords: flames, combustion, soot, electric field, pressure, hydrocarbon fuel, fullerenes.

Introduction. Investigation of processes preceding soot formation has not lost its importance and urgency till now. This is due to the complex and still not clearly understood phenomena taking place in the flame in a very short time measured in microseconds. Investigation of the mechanisms of soot formation and the structure of soot particles is important from the point of view of both environment protection against products of incomplete combustion of gasoline and diesel engines and the physics and chemistry of various forms of solid carbon.

An important aspect in investigating soot formation is the question on the influence of an electric field on the processes proceeding in the flame. Combustion is accompanied by the appearance in the flame of ions of positive and negative charges, as well as of electrons [1, 2]. Charged particles in the flame arise in chemical reactions due to the thermal ionization, ionization by electron impact, and photoionization [3]. The stationary homogeneous flame is a system having generally a neutral charge [4]. However, many authors note that in the process of hydrocarbon fuel combustion the ion concentration in the flames reaches values of 10^{12} cm^{-3} , which exceeds considerably the equilibrium thermal ionization equal to 10^6 cm^{-3} [5]. This phenomenon is inherent only in hydrocarbon fuels and is absent, for example, from H_2 or CO flames [5]. In the flame itself the charged particles are distributed nonuniformly: the reaction zone and the outer cone are characterized mainly by a positive charge, and the inner cone is characterized mainly by a negative charge [4]. Such a distribution of opposite charges is due to the different mobility of positive ions and negative particles — electrons. The specificity of the electrical phenomena and the character of the structure of the flame front in the process of combustion are such that even a weak external electric field produces an effect on all processes proceeding in the flame [3, 4]. Since the flame is initially a weakly ionized plasma, the application of an electric field considerably shifts the processes of soot formation, changes the structure and form of the flame, increases its temperature and the density of charged particles, and, as a result, influences the process of nucleation of particles and their growth, the yield of combustion products, and their structural characteristics and composition [3, 4, 6, 7]. It is also known that soot particles in the flame acquire mainly a positive charge [3, 4].

The kinetics of the process of soot formation under the action of an external electric field is largely determined by C_nH_n^+ -type ions, which are active centers for the growth of soot particles [1–3]. Soot particles acquire a charge at the earliest stages of soot formation; therefore, their size and mass are determined by their residence time in the combustion zone, which can be changed by applying an electric field [3, 8].

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TABLE 1. Voltage and Current Ranges Determining the Type of Gas Discharge

Parameters	Type of discharge		
	dark	corona	normal glow
U , kV	$0.5 \leq U < 1$	$1 \leq U < 3$	$3 \leq U < 20$
I , A	$10^{-7} \leq I < 10^{-6}$	$10^{-6} \leq I < 10^{-5}$	$10^{-5} \leq I < 10^{-2}$

The effect of an electric field on the process of combustion at atmospheric pressure was considered by many researchers from the point of view of the influence on the structure and yield of soot particles. It was determined conclusively that the application of a dc electric field to a diffusion acetylene flame at atmospheric pressure markedly decreased the yield of soot particles and at an intensity of 200 kV/m the decrease was 90% at negative polarity (the electrode over the flame) and 70% in the case of positive polarity [9]. It was concluded that the decrease in the quantity of soot under the action of the electric field was due to the oxidation of soot particles. In so doing, an increase in the voltage applied and the polarity change led to a significant change in the form of the flame. The temperature on the flame end at negative polarity was about 773 K higher than in the absence of electric field and equalled 2073 K on the flame end (throughout the flame the temperature was above 2073 K), which was not observed at positive polarity [9]. The influence of an applied electric field on the formation of nuclei of soot particles and their growth in the pyrolysis zone and deposition was investigated in [3]. The rate of formation of soot particles depends thereby on both the polarity and the ion flow through the pyrolysis zone. Positive ions act as nuclei for the formation of soot particles, which obviously cannot be said about negative charges that have a tendency to decrease soot formation by neutralizing positive ion nuclei [3]. From a detailed analysis of the soot particle sizes performed with the help of electron photomicrography, it was established that the sizes of particles with a diameter of 50 nm obtained in experiments without applying an electric field decreased to 10 nm in diameter upon the application of an electric field of a few kilovolts [3]. In so doing, as was shown in [6], with increasing voltage the soot particles become more homogeneous. Of interest is the use of a low-temperature soot-forming flame for obtaining various polycyclic aromatic hydrocarbons (PCHAHs) with a number of valuable properties [10], soot with given structural parameters [6], and fullerenes [11, 12]. The application of an electric field to the soot-forming flame makes it possible to carry out directional synthesis, influence the process of soot formation, and create a high-temperature level and a high electron concentration for the fullerene yield. At an electric field intensity of 200 kV/m and higher the released Joule heat becomes comparable to the heat release due to the chemical reaction, which increases the flame temperature to 2073 K and higher [9] and creates more advantageous conditions for the formation of fullerenes [13, 14].

Investigation of the effect of an electric field in the form of a gas discharge on the form of the flame, the yield of soot, fullerenes, and PCHAHs, and the structural characteristics of the soot formed at a low pressure is another important aspect in investigating the processes of soot formation.

Experimental. The investigations were carried out using a premixed benzene/oxygen/argon mixture at parameters corresponding to the maximum yield of fullerenes [15]: C/O ratio = 1.0, pressure $P = 5.33$ kPa, and argon content 10% by volume. The burning device was made of quartz glass, and inside of it a burner having the form of a cylinder filled with balls from an inert material was located. Steady-state combustion ensued at an ejection velocity of the combustible mixture from the burner of 16.9–18.4 cm/sec and was provided by a perforated stabilizer set at the exit from the cylinder. The discharge was initiated between two electrodes by applying to the flame a longitudinal dc electric field. The upper electrode was made from tungsten wire and its end had the form of a needle. The role of the lower level was played by the burner. The interelectrode gap was $H = 18$ cm. Voltage was fed to the electrodes from dc high-voltage sources in the range of initial values $U = 0.5$ –20 kV. Depending on the voltage applied and the interelectrode gap, different types of gas discharge arose. The kind of the discharge was determined from the average value of the current in the interelectrode gap (Table 1).

Experiments were performed at negative and positive polarities, i.e., when the upper electrode was a cathode (\ominus) or an anode (\oplus). The duration of one experiment was 20 min. Upon completion of the experiment, soot was collected from the inner surface of the burning device and the filter–soot collector. Soot samples were investigated with the use of a Jem-100CX electron microscope at an accelerating voltage of 100 kV and a DRON-3M diffractometer (CuK_{α} -radiation, $\lambda = 1.54051$ Å, intensity of 1000 pulses/sec, $U = 30$ kV, $I = 20$ mA). To identify fullerenes and

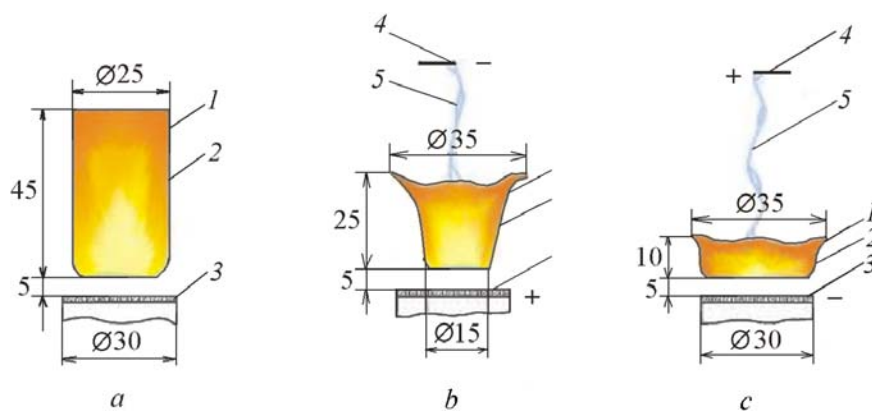


Fig. 1. Form and sizes of the flame at an interelectrode spacing $H = 18$ cm (the flame sizes are given in millimeters): a) without an electric field; b) with an electric field of negative polarity ($U = 10$ kV); c) same, of positive polarity ($U = 10$ kV); 1) flame front; 2) middle luminous part; 3) burner; 4) upper electrode-needle; 5) luminous filament of the glow discharge.

PCAHs, the soot sample in the amount $M = 100$ mg was subjected to cold extraction in benzene for 72 h, and then the obtained extract in the form of a dry residual was investigated on a "Perkin Elmer" IR-Fourier spectrometer.

Change in the Form of the Flame. Investigation of the influence of an electric field on the form of the flame is interesting from the viewpoint of both practical use of the obtained effects and explanation of the mechanism of action of an electric field on flame propagation. Since the flame is a certain electric system with a distributed space charge, upon the application of an electric field a change in the form of the flame is observed: in a transverse electric field it is deflected towards the cathode and in a longitudinal field its height decreases [4]. Experimental investigation of the influence of a dc longitudinal electric field on the acetylene diffusion flame in the process of combustion in the atmosphere [9] showed that in the case of application of negative voltage the end of the flame expanded and its height shortened with increasing voltage. A positive potential of voltage $U \geq 10$ kV smothered the flame.

Investigation of the effect of a longitudinal dc electric field acting on a soot-forming benzene oxygen flame with the addition of 10% of argon at a pressure of 5.33 kPa showed that the effect of the electric field on the flame became visual beginning with $U \geq 1.35$ kV for negative polarity and $U \geq 3.0$ kV for positive polarity [16]. The influence of the electric field showed up as compression, oscillation, stretching, and flattening of the flame as it is transformed into a tulip-shaped form, and other phenomena. In the absence of electric field the flame was steady, without oscillations, and had the form of a cylinder (as the form of the burner) of diameter 2.5 cm and height of the luminous zone 4.5 cm, and with a 0.5 cm lift-off from the burner (Fig. 1a).

Upon the application of an electric field of voltage $U = 10$ kV and higher the upper part of the flame front acquired the form of a wave (see Fig. 1b and c) following the modulatory action on it of the luminous winding filament of the glow discharge, but the lift-off of the flame from the burner remained unaltered. The flame height at negative polarity decreased by 45%, and at positive polarity by 80%. Consequently, the flame height decreases upon the application of an electric field independent of its direction, which agrees with the conclusions given in [4].

For the positive polarity, beginning with $U \geq 10$ kV the effect of the electric field on the flame became strong enough to extinguish the flame, which is analogous to the results obtained under diffusion combustion of acetylene at atmospheric pressure [9], i.e., under the action of an electric field positive ions and neutral particles entrained by them were carried out of the flame front into the preflame zone and further to the negative electrode-burner, which extinguished the flame. At the reversed polarity there was no flame failure, which confirms indirectly the predominant role of positive ions in the organization of the combustion process [3, 4].

In the course of the experiment the voltage dropped, which was followed by an increase in the conduction current (Fig. 2) and the appearance of a large number of hovering soot particles emitted from the flame and depositing on the walls around the burner and then drawn again into the flame front under the action of the electric field [16].

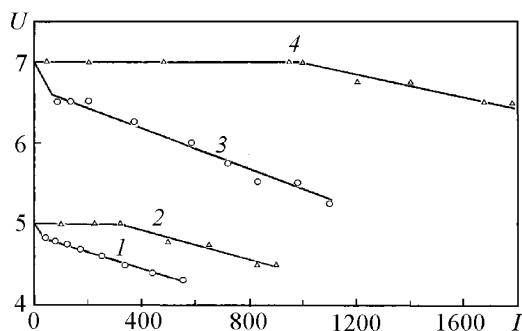


Fig. 2. Applied voltage versus the conduction current in the flame and the electrode polarity during 20 min: 1 and 3) negative polarity; 2 and 4) positive polarity; 1 and 2) at $U = 5$ kV; 3 and 4) at $U = 7$ kV. I , μA .

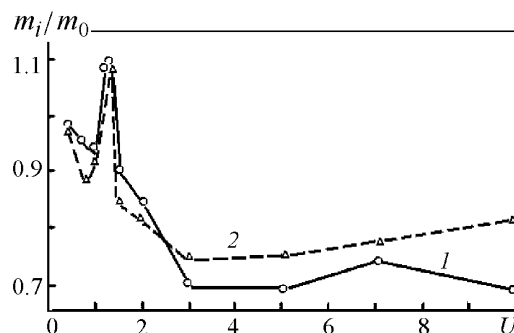


Fig. 3. Experimental dependence of m_i/m_0 on the applied voltage and polarity of the electric field at $H = 18$ cm: 1) negative polarity; 2) positive polarity. U , kV.

The voltage-current curve given in Fig. 2 is, as a rule, descending throughout the range of investigated voltages but is different for different voltage values in both quantitative data and the rate of increase in the discharge current. As is known, this phenomenon is characteristic of gas gaps not obeying Ohm's law [3, 4, 17]. It has been determined that at positive polarity the increase in the conduction current occurred at a smaller voltage drop (curves 2 and 4) than at negative polarity (curves 1 and 3), which agrees with the glow discharge theory [17]. In the glow discharge, which belongs to independent discharges, the electric field is essentially inhomogeneous. The highest intensity of the field and the largest potential drop take place near the cathode, which was just observed at negative polarity in the present work. When the electrode-needle was an anode (positive polarity), the anode region was filled with a positive luminous column in which the process of intensive recombination of electrons with positive ions was proceeding, which resulted in high conductivity of the gas gap in this region [17]. In the analyzed experiments, more complex phenomena appeared, connected with the interaction of ions and electrons formed in the combustion process with the electric field applied. In particular, at the positive polarity the arising positive ions stayed in the combustion zone for a longer time, which was not observed at negative polarity. As a result, this produced a strong effect on the soot formation.

Yield and Structural Characteristics of Soot Particles. As is known, by applying an electric field to the flame, one can control, up to in certain limits, the soot yield. Investigation of the influence of an electric field on this process has shown that in the voltage range from 1.2 to 1.35 kV, whatever the polarity, the soot yield was 10% higher than that without an electric field (Fig. 3) [16].

This phenomenon can be explained on the basis of fundamental investigations [3, 4]. Since the application of an external electric field leads to the appearance of a competing interaction between the process of soot formation and the electric forces tending to remove or hold the active centers in the pyrolysis zone, naturally, a moment most advantageous for soot formation kinetics may occur [3, 4]. In this case, the pyrolysis zone will be crossed by a large flow of positive $C_nH_n^+$ ions. They are active centers of the growth of soot particles according to the soot formation theory [1, 2] and at a certain critical value of the applied voltage can remain almost stationary with respect to the axial component of the gas flow. In so doing, there is a multiple increase in the rate of soot particle formation compared to the case of the absence of an electric field, and in the pyrolysis zone a large number of usually observed soot particles and filaments appear. Analogous arguments apply, according to [3, 4], to the flow of negative charges as well. The maximum decrease in the soot yield compared to the yield without the field was 40% at negative polarity and 25% at positive polarity.

It is known [4] that the sizes of soot particles vary continuously depending on the flame height: in the lower part the particles grow and in the upper part they burn up. Under the action of an electric field the soot particles acquire an additional charge, which may promote their more intensive coalescence and growth. Simultaneously, there is an increase in the temperature of the flame and in the rate of combustion and a change in the character of the soot formation and in the residence time of particles in the combustion zone. All this may change the law of size distribu-

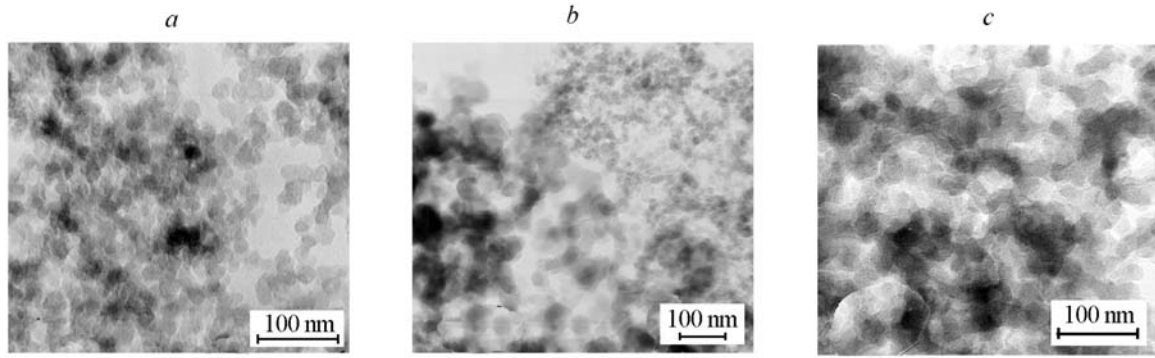


Fig. 4. Photomicrographs of soot samples at $P = 5.33$ kPa: a) $U = 0$ kV; b) 1 kV (\ominus); c) 10 kV (\ominus).

TABLE 2. Parameters of the Size Distribution of Soot Particles

U , kV	Electrode polarity	Soot particle sizes			Standard deviations δ , nm	Variation coefficient $V = \frac{\delta}{d_m} \cdot 100$, %	Normalized index of	
		d_m	d_{max}	d_{min}			asymmetry β_1	excess β_2
0	—	17.2	22.5	11.2	3.36	19.5	0.019	1.9
1	\ominus	18.2	31.4	7.84	6.38	35.1	0.28	2.3
5	\ominus	17.5	26.2	9.84	4.29	24.5	0.053	2.42
10	\ominus	18.9	26.7	11.1	4.01	21.6	0.0004	2.37
20	\ominus	18.8	30.6	14.1	4.19	22.3	2.36	3.5
1	\oplus	22.8	33.3	11.5	4.76	20.9	0.016	2.86
5	\oplus	19.1	34.5	6.9	5.12	26.8	0.95	3.81
10	\oplus	20.9	36.4	12.1	4.68	22.4	0.55	3.54
20	\oplus	16.4	28.1	10.8	4.64	28.3	0.93	3.16

tion of soot particles. Experimental studies have made it possible to elucidate the character of the change in the soot particle sizes under the action of electric fields of different polarities on the benzene/oxygen flame at a pressure of 5.33 kPa [16, 18]. As is known, soot is a polydispersed material. The shape of particles of most kinds of soot is close to spherical. Figure 4 shows photomicrographs of samples of soot particles obtained in the process of combustion of a benzene/oxygen/argon flame at a low pressure.

Usually the degree of dispersion of soot is characterized by the arithmetic mean value of the diameter of soot particles. However, this parameter does not reflect the character of the size spread of particles and does not enable one to draw a conclusion about the degree of their homogeneity and about the law of the size distribution of particles. To this end, we used statistical processing of the results of calculating the weight-averaged sizes of particles by photomicrographs. The results of the investigations of the numerical characteristics of the size distributions of soot particles are presented in Table 2 [18].

As the results of the investigations have shown, the size distribution of soot particles is characterized by the obvious absence of pronounced asymmetry without displacement of the distribution mode from their median values. It has also been established that with increasing initial voltage, independent of the upper electrode polarity, the variation coefficient of distributions is practically in the same range of values (Table 2). The lowest variation coefficient is observed thereby without the application of an electric field. The estimation of the calculation results with the help of Pearson curves [19] by the values of normalized indices of asymmetry $\beta_1 = \mu_3^2/\mu_2^3$ and excess $\beta_2 = \mu_4/\mu_2^2$ has shown (Table 2) that for soot particles formed by combustion of a premixed benzene/oxygen/argon mixture at a pressure of 5.33 kPa both without the field and independent of the voltage applied and the upper electrode polarity, the normal distribution law prevails. However, for the negative polarity of the upper electrode at the initial voltage $U = 20$ kV there is a tendency for the displacement of the soot particle distribution into the region of the uniform distribution law, which confirms the previous conclusion [6] that the structure of soot particles becomes ordered with increasing voltage.

TABLE 3. Distribution Parameters of the X-ray Structural Characteristics of Soot for the Electrode Needle-Plane System

Polarity	Before extraction			After extraction			Before extraction			After extraction			Before extraction			After extraction		
	L_a , Å	δ , Å	V , %	L_a , Å	δ , Å	V , %	L_c , Å	δ , Å	V , %	L_c , Å	δ , Å	V , %	d_{002} , Å	δ , Å	V , %	d_{002} , Å	δ , Å	V , %
⊖	61.7	13.1	21.1	45.3	5.5	12.2	10.93	0.3	2.9	11.1	0.6	5.3	3.7	0.06	1.5	3.71	0.01	0.27
⊕	58.8	6.8	11.5	46.1	4.4	9.5	11.01	0.4	4.0	11.42	0.7	6.5	3.69	0.03	0.82	3.72	0.02	0.54

Note. L_a , L_c , d_{002} are average sizes of the soot crystallite and the interplanar spacing in the voltage range from 0.5 to 20 kV.

We have investigated the influence of the electric field voltage and polarity on the radiographic characteristics of soot samples. As is known, the structural organization of soot particles can be given in the form of three levels: microcrystallites, particles consisting of microcrystallites bound by "amorphous" oxygen, and fractal clusters composed of particles. All these levels are reflected on the diffraction spectra of soot from which the interplanar spacing d_{002} and the microcrystallite width L_a and height L_c were determined. The soot obtained under low-temperature combustion contains also sorbed intermediates representing a mixture of carbon-containing compounds of different classes such as polycyclic aromatic hydrocarbons. Because of their small content, the latter are not registered radiographically but are well diagnosed by IR spectroscopy [16]. The results of the statistical processing of the radiographic characteristics of soot samples obtained with the use of an electrode needle-plane system at different voltage values and polarities of the electric field are presented in Table 3 [18].

It has been determined by the mean statistical indices (see Table 3, values of δ and V) that whatever the applied voltage and the upper electrode polarity, the height of the soot packet L_c and the interplanar spacing d_{002} change insignificantly, which points to the absence of graphitization with increasing voltage of the electric field. The width of the soot crystallite L_a undergoes a greater change towards increasing values when an electric field is applied. The L_a values of soot samples increase due to the addition of polycyclic structures with side chains [20]. When PCAHs are produced as a result of extraction in benzene, the L_a value decreases (Table 3), which is confirmed by the instability of the soot structures formed, and this is more characteristic of amorphous carbon than of graphite. The process of extraction is also accompanied by recrystallization of soot samples with decreasing imperfection, which shows up as an increase in the mean value of L_c and as an increase in the d_{002} values to 3.71–3.72 Å (Table 3). As is known [21], on the flame height there occurs a regular change in the L_c and d_{002} values, and the absence of a law describing their change with increasing applied voltage is explained by the predominant yield of soot formed at a particular level of the flame height. This is a multifactor and stochastic process, and its character did not change at the investigated voltages.

Yield of Fullerenes and PCAHs. Analysis of the IR absorption spectra has made it possible to identify fullerenes C_{60} and C_{70} . The wavelengths corresponding to fullerene C_{60} , 528, 577, 1183, and 1429 cm^{-1} , showed up fairly vividly in the spectra, while for C_{70} they were less vivid. Apart from fullerenes, the extracts also contained a mixture of polycyclic aromatic hydrocarbons. Such compounds as pyrene, fluoranthene, coronene, anthanthrene, and 1,12-benzperylene were identified. The agreement between the experimental and standard wavelength of the absorption spectra of the above compounds is shown in Table 4 [16].

It has been revealed that the negative polarity produces a stronger effect on the increase in the yield of fullerenes with increasing voltage applied compared to the positive polarity.

Analysis of the experimental results for the yield of PCAHs has shown that without an electric field the yield of PCAHs accounted for 13–15% of the mass of the soot obtained. At negative polarity, as the applied voltage is increased, the PCAH yield decreases from 11% at $U = 0.5$ kV to 7% at $U = 20$ kV [16]. At positive polarity the picture is different: first there is a sharp decrease in the PCAH yield to 5% at $U = 0.5$ kV, whose value remains practically unaltered up to $U < 7$ kV, and then at $U \geq 7$ kV the PCAH yield begins to increase and reaches its maximum value of 11% at $U = 20$ kV. The increase in the yield of PCAHs at $U \geq 7$ kV is in accord with the soot yield in this range (see Fig. 3) if we take into account the fact that PCAHs are nuclei for the formation of soot particles.

TABLE 4. Comparison between the Experimental and Standard Wave Numbers of the Spectra of Fullerenes and PCAHs

Substance	Wave number λ , cm^{-1}	
	extract	standard
Fullerene C ₆₀	528, 578, 1183, 1429	528, 577, 1183, 1429
Fullerene C ₇₀	457, 538, 563, 578, 679, 798, 1136, 1414, 1430, 1460	458, 535, 565, 578, 642, 674, 795, 1134, 1414, 1430, 1460
Pyrene	711, 755, 842, 1183	710, 750, 840, 1190
Fluoranthene	618, 755, 775, 827	615, 750, 775, 825
Coronene	543, 842, 1314	545, 850, 1313
Anthanthrene	690, 775, 880	690, 762, 877
1.12-Benzperylene	755, 775, 817, 842	645, 750, 765, 817, 845

In general, it should be noted that the analysis of the influence of an electric field on the flame is a complicated problem in estimating the emerging effects, since the flame itself is already an inhomogeneous weakly ionized plasma. There is no doubt that the application of an electric field to a nonequilibrium flame, which is the flame of a hydrocarbon fuel, causes significant changes in the structure of the flame, increases the space density of charged particles and the flame temperature, and determines the quantitative and qualitative yield of combustion products — soot, PCAHs, and fullerenes.

CONCLUSIONS

1. The flame form changes considerably with increasing applied voltage, and, in so doing, the flame height decreases by a maximum of 45% at negative polarity and by 80% at positive polarity.

2. It has been shown that an internal electric field in the form of a gas discharge at a pressure of 5.33 kPa in the voltage range of 1.2–1.35 kV leads to an increase in the soot yield by 10% compared to the yield without the field.

3. The electric field in the investigated voltage range causes no appreciable graphitization of soot particles, which is reflected by the values of the X-ray structural parameters of soot samples.

4. Negative polarity of the electric field promotes a more effective yield of fullerenes with increasing voltage compared to positive polarity; the action of the electric field on the flame decreases the yield of PCAHs.

NOTATION

d_{\min} , d_{\max} , d_m , minimum, maximum, and mean diameter of soot particles, nm; H , interelectrode spacing, cm; I , current, A; L_a , L_c , d_{002} , width, height, and interplanar spacing of the soot crystallite, Å; M , soot mass, mg; m_0 , specific soot yield without a field, mg/ml; m_i , specific soot yield in the i th experiment with the application of a field, mg/ml; P , pressure, kPa; U , voltage, kV; V , variation coefficient, %; β_1 , β_2 , normalized asymmetry and excess indices; δ , standard deviation, nm; λ , wave number, cm^{-1} ; μ_i , central moments of the i th order of distribution. Subscripts: a, width of the soot crystallite; 0, 1, 2, 3, 4, i , component number; max, maximum value; min, minimum value; c, height of the soot crystallite; m, mean value; 002, reflection index for the hexagonal close-packed lattice of the soot crystallite.

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