Features of formation of fullerene-containing soot during combustion gaseous and liquid hydrocarbons

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Abstract. Features of obtaining fullerene-containing soot during combustion of gaseous and liquid hydrocarbons with the influence of an electric field on the flame are discussed. The characteristic voltage - current characteristics of the discharge created in the flame region and electron microscopic photographs of fullerene-containing soot were obtained.

1. Introduction
The paper considers the features of combustion of hydrocarbon raw materials with an electric field acting on the flame in order to obtain fullerene-containing soot. Synthesis of fullerenes with low cost is relevant due to the great prospects for their use in various technologies. At the moment, the most common way to obtain fullerenes is to obtain them in an arc discharge plasma at pressures of the order of 100 torr. This method is implemented in the presence of an inert gas, such as helium or argon, and the source of carbon atoms is the atomized material of graphite electrodes in an arc discharge [1]. However, the productivity of producing fullerene-containing soot was not high enough.

Another method for producing fullerene-containing soot in flames was considered in [2]. In [3,4], methods producing fullerene-containing soot are proposed, there carbon atoms are obtained through plasma-chemical decomposition of liquid hydrocarbon, such as fuel oil. In [5] a method for producing soot containing fullerenes and nanotubes from gaseous hydrocarbon raw materials is proposed. In [6] the production of carbon nanotubes was considered by decomposing heavy hydrocarbons by the glow discharge creeping along the surface.

Currently a method is promising producing fullerene-containing soot by combustion of hydrocarbon gas or liquid fuels [7-9]. In [7] the process of fullerene formation in flame was considered as the result of a complex sequence of reactions competing with soot formation. Possible paths to fullerenes and soot via aromers are presented on Figure 1. While the formation of fullerenes from fla-
vorings occurs mainly as a result of unimolecular reactions at high temperatures, soot particles are formed in bimolecular reactions of addition of small unsaturated hydrocarbons (for example, \(\text{C}_2\text{H}_2\)) at lower temperatures. Small fullerenes can be formed in very small amounts as a result of oxidative degradation of larger fullerenes. During the process fullerenes and soot particles are formed, and particle degradation reactions occur but relatively large particles are not formed at all. If the temperature is low enough and small growth centers are present in high concentrations, then reactions involving aromers predominate, which do not give enough time to close the cell, which leads to the formation of soot particles. It was suggested that fullerenes are formed in that part of the flame, where the combustion temperature higher.

In [8] it was found that in addition to soot and fullerenes, nanotubes are also formed in flames. In [9], the data was presented on fullerene synthesis in hydrocarbons combustion with the impact of gas discharge. In [10, 11] reviews are given on fullerene synthesis in combined combustion of raw materials under the influence of gas discharge. In [12], data are presented on the production of fullerene-containing soot during gaseous hydrocarbon combustion with the imposition of an additional electric field.

In the presence of electric field, the efficiency of producing fullerene-containing soot significantly depends on the combustion modes and geometry of used electrodes. To increase the efficiency of electric field using it is desirable to maintain stable volume discharge. Some ways of increasing stability of discharge are discussed in [13-15]. In [16] data are presented on the distribution of discharge characteristics in inter-electrode gap, taking into account its inhomogeneity. In [17-19] it is discussed the management of discharge characteristics by implementing various organizations of the glow discharge in inter-electrode gap.

2. Experiment

An installation was created to study the effects of electric fields on combustion processes at pressures of atmospheric, which allowing to combust hydrocarbons both in liquid and gaseous form with the imposition of an external electric field. Kerosene or natural gas was used as fuel. Design feature of the assembled unit is the possibility of rapid changes in inter-electrode distance to create external electric field and control the flow of hydrocarbons in the combustion zone.

Various variants of applying an external electric field were implemented in the experiments. In the first version one electrode in the form of a ring or rod was placed over the burner, which served as the second electrode. In the second version both rod electrodes were located above the burner and created an electric field with the perpendicular orientation to the direction of flame. The schematic diagram of the second installation option is shown in Figure 2, where indicated: 1, 2 - electrodes, 3 – the burner matrix. Combustion control was performed by adjusting the fuel supply and the voltage applied to the electrodes. The supplied electric voltage varied in the range from 0.5 kV to 10 kV.

In the case of natural gas combustion the process of soot formation was carried out on a nozzle burner with a radius of the output channel \(r =0.5 \text{ mm}\). Another burner was used for combustion of kerosene fuel. The base and top of the unit were made of dielectric material. Electrodes were fixed to the upper part of the unit and the distance between them was regulated. The installation also allowed changing the distance between the electrodes and the burner, which made it possible to study the effect of the electric field on different areas of the flame.

The duration of an individual experiment was about 10 min. After the completion of the cycle of combustion, soot is separately gathered from different parts of the installation. Studies of the effect of the electric field were carried out at different inter-electrode distances for the following electrode systems: when burning hydrocarbon natural gas - needle-needle, when burning kerosene fuel - needle-needle and ring-ring. In this case two electrodes were located above the burner matrix.
Experiments both the non-current mode of applying an electric field and the mode with the occurrence and maintenance of an electric discharge were implemented.

The ring-ring electrode system proved to be more effective for the formation of fullerene-containing soot with burning kerosene fuel (the used distance between the electrodes is 8 mm). An electric field was applied to the flame from the burning kerosene fuel. When a voltage of 2.3 kV was applied, soot began to form actively on the anode. When the supplied voltage was increased to 2.6 kV the discharge was contracted, the voltage dropped sharply and a contracted discharge was formed between the cathode and the anode. Figure 3 shows the discharge voltage - current characteristics with the used ring-ring electrode system.

A characteristic electron microscopic photos of fullerene-containing soot particles obtained during natural gas combustion and exposure to an electric field is presented.

In this case the ring serving as the anode and located at the top was made from a copper tube and the burner matrix (serving as the cathode) and through which gaseous hydrocarbons were given, was located at the bottom. The Figure 4 shows the presence of multiple volume formations of fullerene-containing soot with a size of the order of 40 nm or higher.

Figure 3. Ring-to-ring electrode system. The distance between the electrodes is 8 mm.

Figure 4.

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