Investigation of a high voltage AC plasma torch operating on mixtures of methane and other gases

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Abstract. The article considers the high-voltage AC plasma torch working on a mixture of gaseous hydrocarbon (methane, propane-butane) and inert gases (argon). During the burning of the electric arc, methane is transformed into soot containing fullerenes. The composition of the soot was analyzed by optical UV-spectroscopy, IR-spectroscopy and X-ray phase analysis.

1. Introduction

Plasma technologies are actively investigated for a wide range of processing substances: plasma gasification of solid fuels [1], reforming of natural gas [2], decomposition of toxic substances [3], and the production of oxide systems. In addition, plasma pyrolysis of organic substances in an inert atmosphere is studied for a long time. In this case, soot, hydrogen and a small amount of acetylene are formed. There are a number of studies devoted to the plasma pyrolysis of methane under the action of a DC arc [4]. This paper describes the production of fullerene soot in the helium atmosphere (up to 30 kPa) during the evaporation of graphite electrodes. With the change in the interelectrode space, the total yield of fullerences was 4.7-10% at relatively low productivity. The ratio of fullerences is the following: C60 - 65—85; C70 - 11—22; C76-86 - 2—9.3 wt%. The processing time was 4 hours.

In MINES ParisTech using AC arc system [5], soot is produced in the continuous process. At the same time, finely dispersed graphite is used as the precursor of fullerene soot. It is fed into the flow of helium electric arc plasma. The flow rate of helium was changed from 10 to 35 Nm³/h, and the carbon flow rate was 20-600 g/h. The power of the low-voltage three-phase AC plasma torch was 20 kW.

In all these cases, a low voltage was applied, which led to a rapid erosion of the electrodes (due to high electric current). Before, high-voltage plasma torches working on oxidizing media (low-voltage with rail electrodes [6], high-voltage with hollow electrodes [7], multigas plasma torches [8]) were investigated. The erosion of the electrodes of such plasma torches is comparable to the erosion of electrodes of DC plasma torches (at the same currents), which leads to a decrease in the erosion products.
2. Experimental part
A new three-phase AC plasma torch operating on methane and other gases is being investigated (figure 1). A protective gas (argon) is supplied to the near-electrode zone, and methane is supplied to the arc zone. At the same time, the arc ignition is facilitated, and the electrode insulators are prevented from coming into contact with the electrically conductive soot. The power source of the plasma torch consists of a high-voltage transformer, current-limiting inductors, reactive power compensators and a system for measuring electrical parameters. The action principle of such the plasma torch is described in [9].

![Figure 1. The AC Plasma Torch.](image)

The soot was collected from the surface of a water-cooled refrigerator made of stainless steel. The composition and properties of the produced soot were investigated by scanning electron microscopy, IR-spectroscopy and X-ray phase analysis.

3. Results and discussion
Table 1 presents the parameters of plasma synthesis experiments for fullerene soot. Methane and a propane-butane mixture were used as a carbon carrier, and argon was used as the carrier gas.

| Sample | Conditions for the synthesis | Place of sampling | Power, kW |
|--------|-------------------------------|-------------------|-----------|
| 1      | argon - 3.5 g/s, methane - 0.1 g/s | Discharge chamber | 10        |
| 2.1    | argon - 3.5 g/s, methane - 0.05 g/s | Discharge chamber | 9         |
| 2.2    | argon - 3.5 g/s, methane - 0.05 g/s | Cooler           |           |
| 3      | argon - 3.5 g/s, propane-butane mixture - 0.1 g/s | Cooler           | 10        |

The elemental composition of the samples (table 2), with the help of X-ray spectral analysis, practically does not change.

| Spectrum | C     | O     | Ti    | Cr    | Fe    | Cu    |
|----------|-------|-------|-------|-------|-------|-------|
| Spectrum 1 | 90.77 | 2.43  | 0.00  | 0.77  | 2.29  | 3.74  |
| Spectrum 2 | 88.71 | 1.88  | 0.21  | 0.43  | 1.27  | 7.50  |
| Spectrum 3 | 88.80 | 2.82  | 0.36  | 0.87  | 2.23  | 4.92  |
| Spectrum 4 | 89.53 | 3.67  | 0.18  | 0.24  | 0.78  | 5.59  |
On the X-ray diffractogram, reflexes are noted: $30.1^\circ; 50.2^\circ; 59.1^\circ; 64^\circ; 89^\circ$ (figure 2). Its correspond to copper from electrodes and carbon nanotubes.

**Figure 2.** Diffraction pattern of sample 2.1.

From the IR spectrogram (figure 3), a number of bands are supposed to correspond to the following oscillations: $\nu, \text{sm}^{-1}$: 2958 (CH$_3$); 2920 CH$_2$ (or CH$_3$ from benzene); 2874 (CH$_3$); 2840 (CH$_2$)$_n$; 1640 (C=C, RHC=CH$_2$); 1460 (CH$_3$); 1380 (CH$_3$); 1160 (CH$_3$); 1105; 1062; 998;

**Figure 3.** IR spectrum of sample 1.
975 (C-H, RHC=CHR’); 840 (C-H, RR’C=CHR’)). Some peaks are typical for fullerene C60 (1450, 1180).

Figure 4 shows the photomicrographs of sample 1, which show that soot particles are large and highly amorphous. This type of particles is characteristic for soot, obtained by electric arc methods.

![Figure 4. SEM photomicrographs of sample 1.](image)

In the extract of carbon black, treated with ortho-xylene, the content of fullerenes is determined. Figure 5 shows the spectrum of this extract, which shows the presence of a peak of 336±1 nm corresponding to fullerenes.

![Figure 5. Ultra-violet/visible-light spectrum of the o-xylene extract of sample 3.](image)

For all samples, the amount of fullerenes C60 and C70 is determined by the procedure [10]:

\[
\begin{align*}
C(\text{C60}) &= 13.1(D_{335.7} - 1.81D_{472}) \\
C(\text{C70}) &= 42.5(D_{472} - 0.0081D_{335.7})
\end{align*}
\]

Table 3 shows the results of calculating the content of fullerenes.
### Table 3. Estimation of the content of fullerenes.

| Sample | ω(C60),% | ω(C70),% |
|--------|----------|----------|
| 1      | 1,2      | 0,3      |
| 2.2    | 1,3      | 0,2      |
| 3      | 2,2      | 0,8      |

As can be seen, the mass ratio C60 / C70 = 3:1 is typical for fullerene synthesis, which indicates the coincidence of the mechanism of their production with other methods.

### 4. Conclusions

The fullerene-containing carbon black from hydrocarbons was obtained by plasma-arc. The presence of fullerenes is proved by characteristic bands in the UV spectrum (336 ± 1 nm) and absorption bands in the IR spectrum corresponding to the fullerene nucleus (1428, 1182, 1160 cm⁻¹).

According to the optical density of solutions, the content of lower fullerenes in soot samples was estimated. The highest is determined in sample 3 (of the order of 3%, the mass ratio C60 / C70 is typical for fullerene synthesis – 3:1).

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