

X-RAY RAPID ANALYSIS OF FULLERENE CONTENT IN ARC SOOT

Kirillov A.I.*, Karnatsevich V.L., Rasnetsov L.D. ⁽¹⁾

Institute of Organo-metallic Chemistry of RAS,
49 Tropinin St., Nizhny Novgorod, Russia 603095
ZAO "Fullerene-Center" ⁽¹⁾ 4 Kostina St., Nizhny Novgorod 603000

*fax: (8312) 127-497 e-mail: kir@imoc.simm.ru

The first step of the production process of fullerene products by arc method comprises the production of fullerene-containing soot and the content of fullerenes in the soot serves as the index of its quality. This content should be invariable at constant parameters of arc process optimized by the maximum yield of fullerenes in the used set-up. However, the content of fullerenes in soot may be substantially changed due to variation in not adequately controlled parameters, i.e. the purity of buffer gas, the external pressure in case the pressure gages are used, the presence of leakage, etc. This variation may be determined at the subsequent steps of extraction and separation processes but the reason to it may be the changes in parameters of these processes. That is why it is necessary to monitor the content of fullerenes in soot after its unloading from the set-up of arc synthesis.

The generally accepted methods for determination of fullerene content are as follows:

1. Weighting of the unloaded soot (or a part of it as a sample) from the set-up, extraction of fullerene in Sokslet container and its weighting.
2. A complete dissolution of preliminarily weighted sample and measurement of its density in spectrophotometer.

While using not all soot but only its part for determination of the content it should be taken into account that the content of fullerenes in the volume of set-up can change by an order of magnitude. It is hardly possible to provide a uniform mixing of the soot containing particles with size from several nanometers to parts of a millimeter. That is why for reliable determination of the content it is necessary either to pass (or dissolve) in Sokslet all soot unloaded from the set-up or to carry out sampling from a certain part of the set-up. In the latter case the ratio of concentrations in the complete volume and in the sample should be preliminarily found. The change in parameters leads to the change in concentration of fullerenes in any part of the set-up but non-uniformity of this change may be considered to be negligible. However, these methods are time-consuming.

We have attempted to develop a technique for quantitative x-ray analysis of the fullerene-containing soot. The essence of the technique is as follows.

Three phases, i.e. crystalline carbon and fullerene plus amorphous phase, are distinctly observed on diffraction pattern of fullerene-containing soot. The amorphous phase mainly consists of the soot, i.e. of the non-structured carbon (Fig. 1). While changing the ratio of integral intensities of peaks for carbon and fullerene in different samples and determining the content of fullerene during washing in Sokslet container it is necessary to plot a calibration chart in the coordinates of relation between intensities and relative concentration. To remove the non-uniformity in distribution of fullerene inside samples several samples should be taken with subsequent averaging of the result.

To plot the calibration chart the samples were taken from different parts of the reactor with various content of fullerene. The concentration was determined by weighting the residue from Sokslet container.

The measurement was carried out on (computerized) DRON-3M diffractometer with Cu-K α radiation in step mode at narrow angle intervals under the peaks of fullerene and carbon. This chart is given in Fig. 2 for four samples.

It is seen from the chart that deviation in intensity values does not exceed 10%. The chart is used in all subsequent measurements.

The merits of the method are as follows:

1. No need to weigh a sample since only the total intensity of diffracted beam depends upon quantity of substance in the sample (density of cell packing) as the ratio of intensities, being proportional to phase concentrations, remains constant.
2. The time of analysis with subsequent processing of the result is about one hour which is by an order of magnitude less than in the above-mentioned techniques.

We believe that the method of x-ray analysis of soot is the optimum one in fullerene technology combining the required accuracy and rapidness.

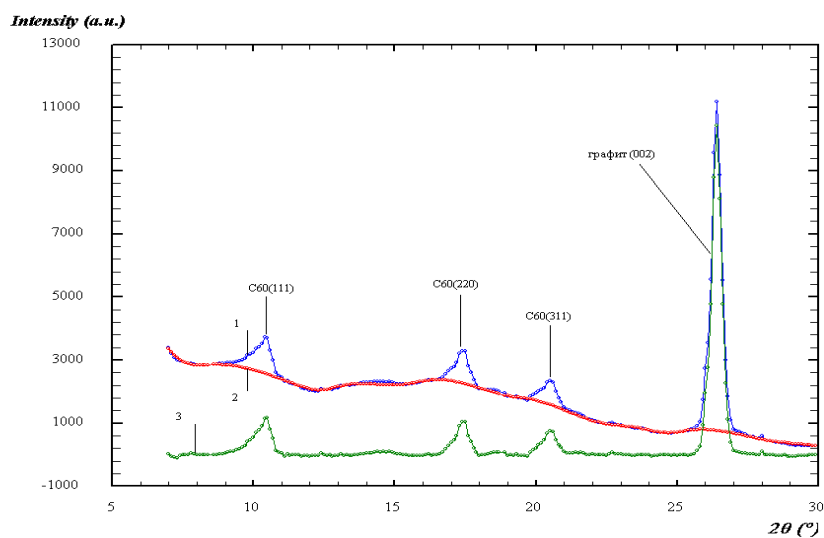


Fig. 1. Diffraction pattern of fullerene-containing soot:
1-the initial diffraction pattern; 2-amorphous constituent; 3-crystalline constituent

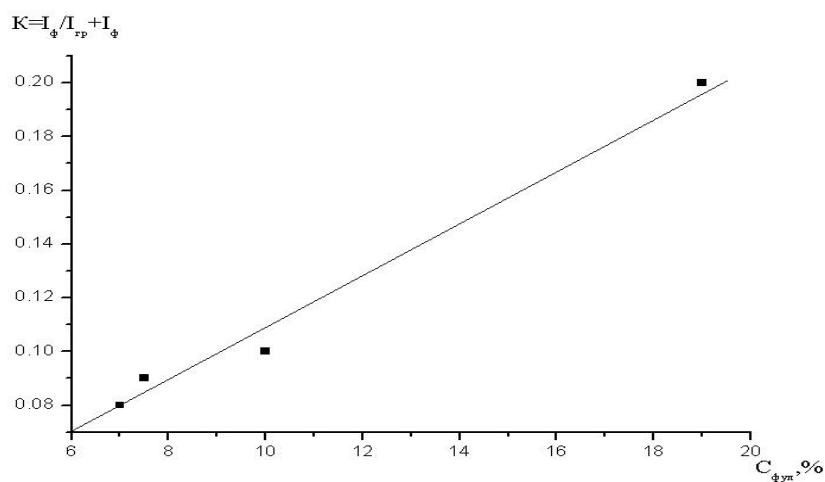


Fig. 2. Calibration plot for determination of the fullerene content in soot

The work is partially granted by ISTC 2511 project.