# ARC CURRENT CONTROL FOR A CARBON NANOPARTICLE SYNTHESIS STATION

## L. Hernández-Tabares, E. Carrillo-Barroso<sup>a</sup>, J.G. Darias-González<sup>b</sup>, L.F. Desdín-García<sup>b</sup>, R.J. Castillo-Torres<sup>a</sup>, J. Arteche-Díaz<sup>b</sup> y M. Ramos-Aruca<sup>b</sup>

Centro de Aplicaciones Tecnológicas y Desarrollo Nuclear (CEADEN), Cuba. lorenzo@ceaden.edu.cu† a) Instituto de Cibernética, Matemática y Física, Ciudad de La Habana, ernesto@icmf.inf.cu

b) Centro de Aplicaciones Tecnológicas y Desarrollo Nuclear, C. de la Habana, darias@ceaden.edu.cu †autor para la correspondencia

Se desarrolló un sistema, basado en un microcontrolador, para controlar la corriente de arco en una estación de síntesis de nanopartículas. La estación funciona bajo el principio de descarga de arco entre dos electrodos de grafito sumergidos en agua. El sistema mide la corriente en los electrodos y, por medio de un mecanismo de microposicionado, utiliza su valor para controlar la separación entre ellos. Se probaron diferentes algoritmos de control para mantener una corriente constante y óptima para la síntesis. A microcontroller based system has been developed to control the arc current in a carbon nanoparticle synthesis station. The station works under the principle of in-water arc discharge between two graphite electrodes. The system measures the current on the electrodes and, by means of a micropositioning mechanism, uses its value to control the gap between them. Several control algorithms were tested in order to maintain an optimal synthesis constant current.

Palabras claves. carbon/carbon-based materials 81.05.U-, methods of nanofabrication 81.16.-c, fullerenes, 81.05.ub

### INTRODUCTION

The properties of the carbon nanoonions (CNO) and carbon nanotubes (CNT) open many potential applications, such as energy storage materials, high-performance and high-temperature wear-resistance materials, superconductive materials and biomaterials.1

The CNO can be synthesized using several tech-niques: irradiation of amorphous carbon up to 700 °C using the electron beam of an electronic microscope, annealing of nanodiamonds at temperatures between 1100 – 1500 °C, a 120 KeV carbon ion implantation in copper and silver targets, chemical deposition of vapors with reinforced plasma by means of radio frequencies and treatment of carbon soot with shock waves.2-6However, all these methods can only produce minus-cule amounts of CNO.

The possibility of producing CNO in significant quantities was reported for the first time using an electric arc discharge in distilled water. This eliminated the use of vacuum and complex gas valve systems or catalysts.<sup>7</sup>

In an in-water arc discharge synthesis process, the bubble generated by plasma acts as a reaction chamber where carbon atoms, evaporated from the cathode surface, regroup themselves close to liquidvapor surface to form carbon nanostructures.[8] The plasma parameters determine the formed structure type and its defects.

One of the most important parameters in this synthesis process is the stability and the value of the arc current. It is the first one to be controlled and monitored in present and futures works. The literature has reported an optimal arc current value of 30A or 30-40A.8,9

In this work several control algorithms were tested in order to maintain an optimal synthesis constant current. The experimental setup description, two methods for measuring the arc current, the functioning of the control algorithms and their results are presented.

#### METHODOLOGY

Experimental setup. The experimental setup is composed of three major blocks: synthesis station, power supply and control system.

The scheme of the synthesis station is shown in Fig. 1. A micropositioning mechanism (1), driven by a step motor (2), is supported by an aluminum frame (3). The electrode holder (4) of the cathode (8) is fixed to this structure. However, the electrode holder (5) of the anode (6), is mounted on the micropositioning mechanism car allowing a relative electrode coaxial movement.

The electrodes were submerged in  $1.2M\Omega$  resistivity distilled water contained on a 15L pyrex glass vessel (7). The vessel's transparency allowed the electric arc light surveillance.

As electrodes, spectroscopic pure graphite cylinders ( $\emptyset$ cathode = 12mm and  $\emptyset$ anode = 5mm) were used.

To power the synthesis station a dedicated direct cur-rent power supply was developed. It was able to provide currents of up to 100A and variable voltages in the range of 15-25VDC (  $\emptyset$ 0.5VDC). The power supply was connected to the electrode holders polarizing them. An am-meter was placed in the circuit to show the electrode current (Fig. 2).

Two methods were evaluated to measure the arc cur-rent: the optoelectronic method and the shunt method.

With the optoelectronic method the arc current could be estimated by measuring the arc emitted light with an optoelectronic sensor. No direct correlation was observed between the arc emitted light intensity and the ammeter's current indication. This light was not possible to observe from the same point because it was constantly emitted in different directions from the electrodes. Due to these problems this method was discarded.

The arc current could also be determined by placing a shunt in the circuit (Fig. 2) and measuring the voltage drop on it. This voltage drop is linearly proportional to the arc current. This method was more simple and direct to use than the optoelectronic one.

The control system was formed by a current shunt, a fixed-gain low-pass filter, a PIC16F4550 microcontroller and a four-phase step motor driver (Fig. 3).

The  $330\mu\Omega$  shunt generated a voltage drop proportional to the electrode current. This voltage was amplified and filtered by a 4th order Butterworth low-pass analog active filter with a cutoff frequency of 20Hz. The filtered signal was digitalized by the microcontroller's 10bit ADC.



Figure 1. Synthesis station.



Figure 2. Electric diagram of the experimental setup.



Figure 3. Block diagram of the control system.

Depending on the current value, the microcontroller was acting over the step motor to force the anode to move forwards or backwards to the cathode.

In this work a half-stepping control of the step motor was used.

Control system description. Three control algorithms were used during the experiments: the so called by us "forward-backward" algorithm, the "only forward" algorithm and the "constant speed" algorithm.

The first two algorithms worked in a similar way. In both cases two current set points (maximum and minimum) were prefixed. If the electrode current was smaller than the minimum set point, the microcontroller forced the step motor to rotate one step forwards. Consequently the gap between the electrodes got smaller increasing the current. On the contrary, when the electrode current was higher than the maximum set point, the step motor rotated one step backwards, thus the gap between the electrodes got bigger and the current got smaller. No movement of the step motor was ever made when the electrode current was within the maximum and minimum set points.

When the system was turned on the electrodes were separated and there was no arc discharge between them. As the electrode current was zero it was smaller than the minimum set point and the microcontroller started to move the anode until it reached the cathode (short cir-cuit). In that moment the arc discharge started, the electrode current became higher than the maximum set point and the microcontroller started to separate the electrodes until the arc current was within the minimum and maximum set point. In the "forward-backward" algorithm the minimum and maximum set point values were fixed very close one to each other, usually  $30A \pm 3A$  (or  $\pm 5A$ ).

In the "only forward" algorithm the minimum set point was prefixed around 25A but the maximum set point was prefixed far from the minimum, usually 60A.

The third control algorithm was tested based on the anode average displacement speed, calculated from the "only forward" algorithm. In this algorithm the anode displacement speed was constant and did not depend on the arc current value (open loop).

#### RESULTS

The "forward-backward" algorithm showed certain in-stability managing the arc discharge; even some times it caused the arc to turn off. That happened because of small mechanical imperfections any mechanical system has, increased by drastic changes of the step motor direction of rotation, inertia of our micropositioning system and mechanical oscillations the arc discharge causes on the electrodes. Testing this algorithm it was realized that, as the electric arc was constantly eroding the anode, it was also permanently increasing the gap between electrodes; consequently, to make backwards movements to increase the gap could be not necessary.

In the "only forward" algorithm we took advantage of the previous argument. Once the gap was reduced and the current was over the minimum set point, the anode electrode was kept in a fixed position while the erosion process was making the gap to increase again, thus decreasing the current. That caused the current to oscillate close to the minimum set point, without making back-wards movements (Fig.4). For a minimum set point of 25A an arc current of  $30A \pm 5A$  with some spikes suitable for the synthesis of CNO was observed (Fig.4).



Figure 4. Arc current in the "only forward" algorithm.



Figure 5. Arc start current.



Figure 6. Self-extinction process of the arc current.

Actually, the "only forward" algorithm can make backward movements but, as we moved away the maximum set point (60A) from the minimum, it acts only as a protection against high currents (e.g. at the moment the arc starts).

A current data acquisition for this algorithm was made using the microcontroller's USB interface and a program developed in LabVIEW 8.6 for Windows based computer.

In order to get more information on the arc discharge processes two more data acquisitions were made when using the "only forward" algorithm. The first one registered the current behavior at the moment the arc starts (Fig. 5) and was noticeable the absence of a peak at the very beginning due to the filter's action. The second one captured the selfextinction process of the arc current after the control system was stopped, keeping the electrodes in a fixed position (Fig. 6). It was expected the current to slowly decrease to a certain value and then to turn off, but a series of irregular form steps with decreasing tendency were observed. This showed the complexity of the current's behavior in the system.

In the "constant speed" algorithm, the arc current, in-stead of being constant, it was slowly increasing until the current was too high, or decreasing, until the arc was turned off. As the electrode erosion speed is not uniform, there is a difference between the calculated speed and the real one. This leads to an accumulative error on the displacement over the time. If the calculated speed was smaller than the real one the gap between the electrodes was growing bigger until the arc turns off. On the con-trary, when the calculated speed was bigger than the real one, the gap was every time smaller and the arc current was increasing beyond the optimal for the synthesis value.

#### CONCLUSIONS

To measure the arc current using a resistor shunt is a reliable, simple, economic and direct method.

The "only forward" control algorithm showed to be appropriate to guarantee a stable arc current suitable for the synthesis on CNO.

With the current under control the system can be improved further and extended to monitor other parameters also important for the synthesis process and for research purposes. The optical method needs to be better evaluated as it was reported in the literature and the monitoring of more parameters can be added to the control system.[10]

The less the size of the step is, the smaller the increment in current will be. Reducing the step size the control could be more accurate but we have then to rotate faster the motor to comply with the erosion speed of the process. Doing this we have to take care in not to exceed the motor's speed limit.

beam irradiation", Nature 359, 707-709 (1992).

[3] V.L. Kuznetsov, "Onion-like carbon from ultra-disperse diamond", Chem. Phys. Lett. 222(4), 343-348 (1994).

[4] T. Cabioch, "Structure and properties of carbon onions layers deposited onto various substrates", J. Appl. Phys. 91(3), 1560-1567 (2002).

[5] H. Chen, "New method of carbon onion growth by ra-dio-frequency plasma-enhanced chemical vapour deposition", Chem. Phys. Lett. 336(3-4), 201-204 (2001).

[6] K. Yamada, "Electron microscope study of carbon par-ticles developed using a conically converging shock-wave technique", Carbon 26(6), 867-871 (1988).

[7] N. Sano, "Synthesis of carbon onions in water", Nature 414, 506-507 (2001).

[8] N. Sano, "Study on reaction field in arc-in-water to produce carbon nanomaterials", Thin Solid Film 516(19), 6694-6698 (2008).

[9] H. Lange, "Nanocarbon production by arc discharge in water", Carbon 41, 1617-1623 (2003).

[10] D. Bera, "Optoelectronically automated system for carbon nanotubes synthesis via arc-discharge in solution", Rev. Sci. Instrum. 76(3), 033903-1-033903-6 (2005).

<sup>[1]</sup> X. Bing-she, "Prospects and research progress in nano onion-like fullerenes", New Carbon Materials 23(4), 289-301 (2008).

<sup>[2]</sup> D. Ugarte, "Curling and closure of graphitic networks under electron-