Publ. Astron. Obs. Belgrade No. 76 (2003), 233-236 *Contributed paper*

THE EFFECT OF CARBON CONCENTRATION ON ROTATIONAL PLASMA TEMPERATURE OF CARBON ARC IN HELIUM

B. TODOROVIĆ-MARKOVIĆ[1], Z. MARKOVIĆ[1], Z. NIKOLIĆ[2], T. NENADOVIĆ[1]

[1] *"Vinča" Institute of Nuclear Sciences, P.O.B. 522, 11001 Belgrade, Yugoslavia*

[2] *Faculty of Physics, University of Belgrade, P.O.B. 368, 11001 Belgrade, Yugoslavia*
E-mail:biljatod@vin.bg.ac.yu

Abstract. In this work, we present the results of a spectroscopic investigation of the emission from C_2 radicals of carbon arc in inert gas-helium. The rotational temperature of C_2 radicals was determined in dependence of electrode-anode- diameter. It was found that the rotational temperature of diatomic carbon molecules decreased when electrode diameter decreased. The pressure dependence on rotational temperature has shown that the rotational temperature of C_2 decreased with the increase of gas pressure.

1. INTRODUCTION

Since the fullerenes were discovered by Kroto and co-workers in 1985, they attracted a lot of interest of researchers because of their excellent properties (Kroto *et al.,* 1985). There are a few methods for fullerene synthesis: they are obtained by burning hydrocarbons, by resistive and rf heating of graphite in atmospheres of inert gases and in the so-called contact-arc (Withers *et al*., 1997). In order to understand the phenomena related to fullerene formation and to increase the fullerene yield, it is essential to study the role of parameters of carbon plasma. Kinetic model of fullerene formation in plasma arc reactor was developed previously (Marković *et al*., 1998). The fullerenes were formed in a hot jet of an inert gas that expanded from the interelectroede space. Based on the kinetic model, it was found that fullerene yield depended on four physical parameters simultaneously: carbon concentration, the velocity of carbon-helium jet from the interelectrode space, axial temperature and temperature gradient between the arc channel and chamber walls (Marković *et al*., 2002).

In this article, the results of spectroscopic measurement of emission molecular spectra of C_2 radicals are presented. We focus our attention on the determination of rotational temperature of C_2 radicals as a function of electrode diameter and the influence of gas pressure on the rotational temperature as well.

2. EXPERIMENTAL

The experimental used in this study was the plasma reactor for fullerene synthesis with minor modifications including two additional quenched glass windows for spectroscopy. All experiments were carried out under helium atmosphere, using electrodes with different diameters (D_{el}=12, 9, 7, 6, 5, 4 mm). The soot produced during the discharge was collected and the fullerene content was measured by the method described before (Marković *et al*., 2002). The gas pressure was in the range of 200-600 mbar. The current intensity had values of 50, 70 and 100 A.

The emission molecular spectra were recorded using the plane grating spectrograph of Ebert design (RSV). The light emitted from the carbon arc gap and the entrance slit of the spectrograph was fixed on 20 cm. The emission spectra of the carbon arc in helium were registered by CCD digital camera (Camedia-Olympus C700 ultra zoom). The spectra of Swan molecular bands of C2 radicals ((0,0) at 516.5 nm) were recorded with resolution of 150000 and reciprocal dispersion of 0.45 nm/mm which is enough to resolve rotational structure of emission spectra. The apparatus function was 0.02 nm. The spectrograph entrance slit was 15 μm. During the experiment, the expansion of carbon vapor from the interelectrode space was monitored on calibrated screen.

3. RESULTS AND DISCUSSION

The carbon vapor was formed in C/He arc plasmas under conditions favoring formation of fullerenes. In the spectra, intensive lines of CI, CII, HeI and Swan bands of C_2 radicals were detected.

The determination of gas temperature in the arc is performed by measurements of the rotational temperature of C_2 radicals. Owing to a small energy separation between the rotational levels of the molecules, the populations of the rotational states correspond more closely to the translational gas temperature in the arc column. Inelastic electron-molecule collisions excite C_2 radicals without altering their angular momentum. Hence the excited states have the same rotational distribution as the ground state. The C_2 ($d^3\Pi_g$) lifetime of the order of 1μs is much longer than the mean time between collisions at the atmospheric pressure which can be evaluated as less than 1 ns. Since rotational equilibrium typically requires ≈10 collisions, rotation-translation equilibrium does prevail for all emitting species.

Typical emission spectrum of C_2 ($d^3\Pi_g \rightarrow a^3\Pi_u$) recorded at the center of arc discharge is presented in Fig. 1. The rotational temperature of C_2 molecules was determined by Boltzman plot method. The obtained values should be considered to be average values over the spatial region. We assumed the plasma is optically thin.

The rotational temperature of C_2 radicals was measured as a function of carbon concentration on and around the arc axis. The carbon concentration was changed in the wide range by varying the electrode-anode-diameter and the gas pressure. In Fig. 2, the rotational temperature of C_2 radicals is presented as a function of electrode diameter. As can be seen from the diagram, the temperature decreases with the decrease of electrode diameter. For electrode diameter of 12 mm (carbon concentration is small), the rotational temperature is in the range of

5500-7000 K. For electrode diameter of 5 mm (carbon concentration is high), the rotational temperature of diatomic carbon molecules is varied from 2500-3000 K.

When the carbon concentration is small (D_{el}=12 mm), the carbon vapor is mainly located between the electrodes. The dominant mechanism for the excitation of C_2 is the collisions among electrons and C_2 radicals. In the case of high carbon concentration (D_{el}=5 mm), carbon vapor was located in the wide region: between and around the electrodes. Then, chemiluminiscent excitation of C_2 is dominant excitation mechanism. Particularly, this effect is very strong in the case of carbon arc discharge under conditions favoring the fullerene formations.

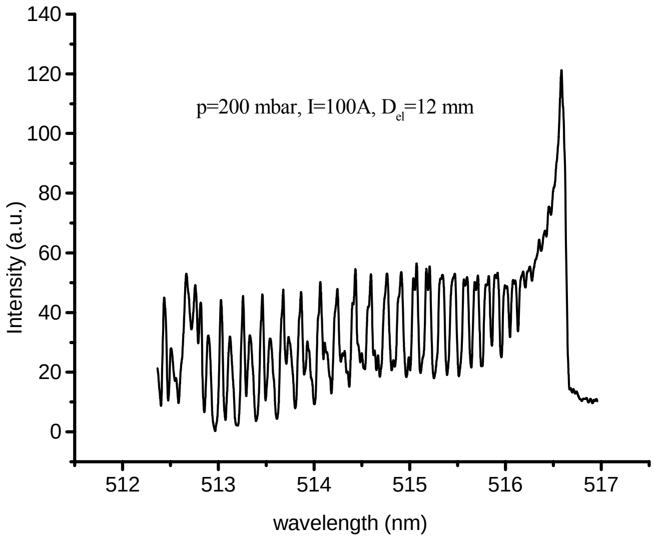

Fig.1: Emission molecular spectrum of C_2 radicals.

By changing the gas pressure (200-600 mbar), the rotational temperature of C_2 decreases with the increase the pressure of buffer gas. It was found that T_{rot}=7000 K was nearly constant when the anode diameter of 12 mm was applied. In the case of anode of 6 mm, significant changes of T_{rot} were detected. Rotational temperature was in the range of 4500-3500 K as the gas pressure increased. By increasing of buffer gas pressure, jet velocity from the interelectrode space decreases (Ramakrishnan *et al.*, 1978). Then, the concentration of C_2 increases. Therefore, T_{rot} will decrease.

The obtained values of T_{rot} have different dependence on carbon concentration than vibrational temperature. While T_{rot} decreases with electrode diameter decreases, T_{vib} increases with carbon concentration increases (Marković *et al.*, 2003).

It is worth mentioning that the highest yield of fullerene produced under He atmosphere was determined to be about 13 %.

4. CONCLUSION

In this work, the influence of carbon concentration and gas pressure on rotational temperature of C_2 radicals was studied. The rotational temperature was determined from the emission molecular spectra of the 0-0 band of $d^3\Pi_g \rightarrow a^3\Pi_u$ electronic transition of C_2. It was found that the rotational temperature of C_2 decreased as carbon concentration increased.

The difference between the rotational and vibrational temperatures of C_2 radicals indicates that there is no thermal equilibrium between rotational and vibrational population.

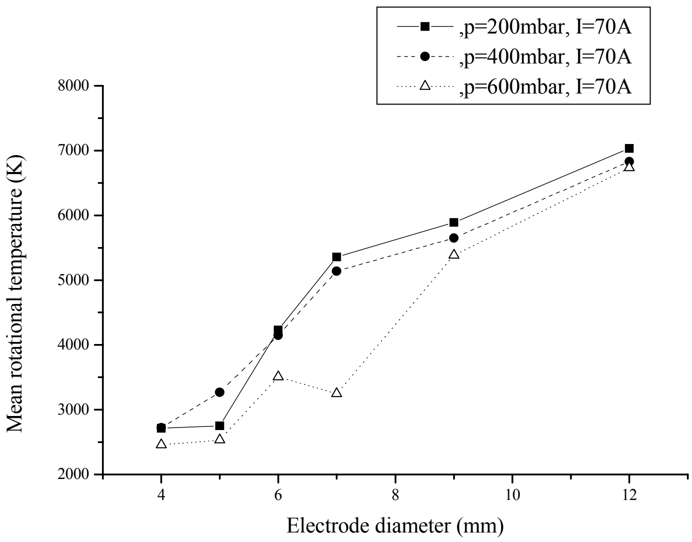

Fig. 2: Rotational temperature of C_2 radicals as a function of electrode diameter.

References

Kroto, H.W., Heath, R., O'Brien, S.C., Curl, R.F., Smalley, R.E.: 1985, *Nature*, **318,** 162.

Withers, J.C., Loutfy, R.O., Lowe ,T.P.: 1997, *Full. Sci.and Technol*., **5**, 1.

Marković, Z., Todorović-Marković, B., Jokić, T., Pavlović, P., Stefanović, P., Blanuša, J., Nenadović, T.: 1998, *Full. Sci. and Technol*. **6**, 1057.

Marković, Z., Todorović-Marković, B., Nenadović, T.: 2002, *Full.Nanotubes and Carbon Nanostructures*., **10,** 81.

Ramakrishan, S., Stokes, A., Lowke, J.: 1978, *J. Phys. D:Appl. Phys*., **11**, 2267.

Marković, Z., Todorović-Marković, B., Marinković, M., Nenadović, T.: 2003, *Carbon*, **41**, 369.