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Sputtering of graphite in pulsed and continuous arc and spark discharges

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Abstract

The environment that leads to the sputtering of graphite electrodes and formation of carbonaceous discharge has been studied with emission spectroscopy. Population level densities, excitation & vibrational temperatures and electron densities have been obtained from a set of three ion sources. The sources operate in continuous and pulsed discharge modes. The sputtered species include monatomic, diatomic and higher carbon clusters. The main sputtered species are excited and ionized C_1 (CI, CII, respectively) and C_2. In the continuous arc discharge the vibrational temperature derived from the Swan band of C_2 is ∼10,000 K, whereas, in the pulsed arc the excitation temperature of Neon is ∼11,000 K. The spark discharge yields an average excitation temperature of CI and NI ∼ 5500 K.

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1. Introduction

In this work sputtering has been studied in different ion source geometries using graphite as electrodes in the presence of a discharge support gas Ne and air. The kind of sources studied here can be operated in two modes, i.e. continuous and pulsed discharge. These sources have been operated in non-regenerative sooting discharge environment. Regenerative sooting discharges, which have been thoroughly reviewed elsewhere [1], are specified by a glow discharge at $V_{dis} \gtrsim 300$ V in graphite hollow cathodes with He, Ne, Ar as the discharge support gases. Recent results from our laboratory have shown that the mechanism of cluster formation in such regenerative sooting environments results in C_3 being the dominant stable species among the carbon clusters. The relatively higher discharge voltage $\geqslant$300–500 V is responsible for the sputtering of the graphite cathode. The sputtered species redeposit on the walls of the cylindrical hollow cathode and may be re-sputtered or desorbed into the discharge [2].

The continuous arc discharge has certain similarities with the soot production by arc discharge of Krätschmer et al. [3] in which gram quantities of carbon clusters, especially fullerenes and nanotubes embedded in the carbon soot, can be produced. Those arc discharges that lead to the formation of soot rich in fullerenes and nanotubes [3,4] are generally produced at low voltages of ∼20 V and large currents of 100–150 A. Such arc discharge conditions in high pressures of noble gases have been suggested as providing ideal conditions for self-assembly of the sublimed C_1 and C_2 on the cathode, which may lead to the

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formation of closed cage carbon nanostructures [5–7]. The role that C_2 plays in cage closure is itself a subject of intensive studies [8–10]. Therefore, there is a subtle difference in the production of carbon clusters in a regenerative environment and the one in non-regenerative mode.

As a result of bombardment of energetic ions of a background inert gas, carbon atoms are sputtered from the surface of a graphite electrode. The emitted atoms may be C_1 or its clusters C_n where $n > 1$. Emission spectroscopy has been used to study the state of excitation of emitted atoms and clusters. The level density N_u of the upper excited levels (electronic or vibrational) can be obtained from the emission spectrum using the relation $I_{ul} = N_u A_{ul} h\nu_{ul}$ [11] where I_{ul} is the calibrated intensity (μW/cm^2-nm), $h\nu_{ul}$ is the energy difference between upper (u) and lower (l) levels and A_{ul} is Einstein transition probability for spontaneous emission. The upper level density N_u is used to calculate the vibrational (or excitation) temperature by the Boltzmann relation

$$N_u = \frac{g_u N}{U(T)} \exp\left(-\frac{E_u}{k_B T_{vib}}\right),$$

where E_u is the energy of the upper level relative to the ground state, N and N_u are total density of particles and density of particles in the u state, respectively, g_u is the statistical weight of the upper state, $U(T)$ is the atomic or molecular internal partition function and k_B is the Boltzmann constant. If a range of upper level densities can be calculated from the emission spectra, then by plotting $\ln(\frac{N_u}{g_u})$ as a function of E_u one can determine the vibrational (or excitation) temperature (T_{vib}) from the slope $(-1/k_B T_{vib})$ [12].

The Boltzmann relation is valid only if the plasma is in local thermodynamic equilibrium (LTE). The population inversion parameter (PI) is defined by $[N_u g_l/N_l g_u]$ where, N_u and N_l are upper and lower level number densities, g_u and g_l are the statistical weights of the upper and lower states, respectively, [13]. For LTE plasma PI < 1 for all the levels involved in the transition. For non-local thermodynamic equilibrium (NLTE) plasma, defined by PI > 1, one cannot define any temperature using the Boltzmann relation. The excitation temperature T_{exc} can be calculated for those pair of levels N_u and N_l of a species which falls on a common level r and also PI < 1 for these levels. The ratio of the relative level densities N_u/N_l can be derived from Boltzmann relation [11,13] given by

$$\frac{N_u}{N_l} = \left(\frac{g_u}{g_l}\right) \exp\left[-\frac{(E_u - E_l)}{kT_{exc}}\right].$$

2. Experimental

Three experimental setups are shown in Fig. 1 representing three types of discharges being studied. These setups are used to study the sputtering of graphite in continuous and pulsed discharge modes

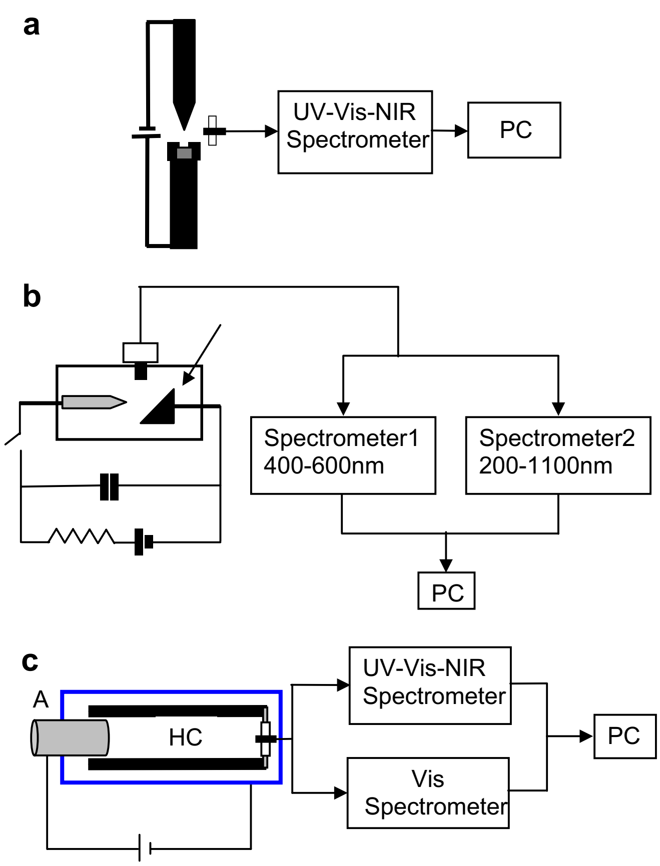

Fig. 1. Schematic diagram of experimental setups for the (a) spark discharge, (b) pulsed arc discharge and (c) continuous arc discharge.

2.1. Continuous spark discharge

Fig. 1(a) shows the experimental setup used for continuous spark discharge. A pointed graphite cathode is about 3 mm distant from a graphite anode having a 5 mm deep cavity filled with graphite powder. Both electrodes are 40 mm long with a 6 mm diameter. Spark discharge is operated at atmospheric pressure in air with a voltage of 19 kV and current 6 A.

2.2. Pulsed arc discharge

The experimental arrangement for a pulsed arc discharge is shown in Fig. 1(b). It occurs between a pointed iron anode with a wedge-shaped graphite cathode, separated by 10 mm, in the presence of support gas Ne. It is generated by applying high voltage ~kV. The pulsed voltage is applied with the help of a 12 μF capacitor. The capacitor is charged with a power supply in series with a resistance of 1.43 MΩ . The pulsed discharge mode ensures the passage of high electrical energy through the gas in a short interval of time ~μs. The auto discharge voltage is about 0.4 kV with a repetition rate of 0.12 s^{-1}. However, in this way one cannot deliver energy above a certain level due to geometrical constraints for a specific gas. To increase the pulse energy, the capacitor is discharged