Kinetics of electron-beam dispersion of fullerite C60

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Features of Electron-Beam Dispersion of Fullerite C₆₀

1. Introduction

Electron beam irradiation of fullerite C₆₀ can lead to various physical-chemical effects ranging from molecular structure modification to bulk explosive sublimation. The effect depends on the energy of the electrons, the current density of the electron flux, the exposure dose and the conditions of the energy dissipation.

Polymerization of thin C₆₀ films was observed upon irradiation by electrons emitted from a scanning tunneling microscope (STM) tip with energy of 3-4 eV [1-4]. Fullerene fragmentation and formation of coalescent spheroid structures was detected with increase of the energy of the electrons emitted from a STM tip to 20-45 eV [5] and to 120-310 eV [6]. The authors suggested that heating effect is negligible under the irradiation conditions used (the exposure dose of up to 7200 nC and the electron flux of $\sim 10^{-3}$ - 10^{-1} nA/nm²) and therefore, it is electron excitation that causes fragmentation and coalescence of C₆₀ molecules.

Polymerization, hydrogenation and consequent fragmentation and graphitization of ultrathin C₆₀ films grown on hydrogen passivated Si (100) substrates was observed after irradiation with 0.5 keV and 3.3 keV electrons. The electron beams were produced by an electron gun with a nominal maximum of electron flux density of ≈ 64 mA/cm² [7]. The films were exposed to

the dose of up to 3×10^{-2} C/cm² for 0.5 keV beam and 9×10^{-1} C/cm² for 3.3 keV beam. No measurable desorption of the film material was detected under the irradiation conditions used. Thermally reversible polymerization was reported for ultrathin C₆₀ films on GaAs (110) substrate exposed to a defocused 1.5 keV electron beam with the flux density of ≈ 1.6 mA/cm² and the exposure dose of up to ≈ 18 C/cm² [1]. The polymerization was described as a nucleation-and-growth process influenced by the irradiation-induced heating. Formation of peanut-shaped C₆₀ polymers in a 70-nm-thick C₆₀ film grown on CsI substrate was observed upon irradiation by a scanning 3 keV electron beam [8]. The electron flux density used was ≈ 0.1 - 0.3 mA/cm². The nonlinear dependence of the reaction rate on the electron dose rate was found. The authors have concluded that a complex reaction between secondary electrons and intermediate products leads to coalesced C₁₂₀ dimer formation.

A normalized modification parameter P was proposed on the basis of the relative change of the low-energy part of the electron energy loss (EEL) spectrum of C₆₀ films under 150-1500 eV electron irradiation. It was found that the modification parameter is a monotonously raising function of the irradiation dose and incident electron energy. Fast change of the P under small irradiation doses corresponds to C₆₀ polymerization. Subsequent saturation of the P under high irradiation doses corresponds to C₆₀ fragmentation and amorphization [9, 10]. It should be noted that a low-intensity electron beam of the electron spectrometer was used for irradiation. Therefore, the irradiation-induced heating can be considered negligible under the conditions given.

A 20 keV tubular electron beam with a current of 50 mA focused into a spot of 3 mm in diameter (the current density of $\approx 700 \text{ mA/cm}^2$) was used for explosive sublimation of a fullerene target within less than 1 s and simultaneous deposition of a nonpolymerized fullerene film onto a substrate [11]. In another report [12], 20 keV electron irradiation was used to decrease significantly the dissolution rate of a C_{60} film owing to fullerene polymerization and subsequent graphitization. The film was exposed to the irradiation dose of up to 0.024 C/cm^2 . As no detectable film desorption was observed, we can suppose that the electron beam used had a considerably lower current density than in the previous case.

We have introduced the method of electron-beam dispersion (EBD) for synthesis of thin films on the basis of fullerene C_{60} [13]. In the EBD method, an electron beam of 1-1.5 keV energy and 30-50 mA current is used to heat and sublime the target made of pristine material in high vacuum. The coating is deposited onto a substrate from the volatile sublimation products typically containing neutral and excited molecules, ions and radicals. In addition, the coating is deposited under the irradiation of scattered primary electrons and true secondary electrons from the target and the target holder. We have shown that the EBD of a fullerite C_{60} target produces thin C_{60} coating containing different C_{60} polymeric forms [14]. In this report, we will concentrate on the analysis of kinetics of fullerite C_{60} EBD. On the basis of the obtained experimental data and

simple physical estimates, the physical-chemical processes in the fullerite target during its EBD will be speculated about.

2. Experimental details

Experimental setup for studying the kinetics of fullerite EBD is shown in Fig. 1. 80 mg of fullerite C₆₀ powder (purity > 99,5 wt.%) was pressed into a disc of 5 mm in diameter and of ≈ 2.5 mm thickness under pressure of ≈ 25 MPa for 30 s. The target was placed on a copper heating table in a stainless steel vacuum chamber. The vacuum chamber was evacuated by a turbomolecular pump and kept under the pressure of 1×10^{-2} Pa. The fullerite target was then heated up to 473 K and annealed at this temperature for 1 hour. Vacuum annealing of the fullerite target was introduced to decrease the amount of oxygen absorbed by fullerite.

Electron-beam dispersion of the target was enabled by an electron gun with a directly heated wire cathode of tungsten. Electron beams with energy of 1-1.5 keV and current of up to 50 mA were used for EBD. However, we have found that the electron beam with energy of 1.2 keV and current of 30 mA provides the most stable EBD of the fullerite target under the given experimental conditions. Hereafter, we will describe and analyze EBD of fullerite C₆₀ by a 1.2 keV, 30 mA electron beam. The electron beam spot was larger than the target thus providing its uniform irradiation. The current density of the electron beam irradiating the surface of the target was about 10 mA/cm² according to the measurement by a Faraday cup. It should be noted that this

measurement was performed by replacing the heating table with the target by the Faraday cup and thus, it does not consider possible target charging upon irradiation.

The temperature of the target surface was registered by an IMPAC IPE 140 radiation pyrometer with a PbSe infrared sensor (spectral range of 3-5 μm) through a vacuum window made of a KRS-5 crystal. The pyrometer enabled measurement of the surface temperature of the target in a spot of 0.7 mm in diameter. The emissivity coefficient $\varepsilon \approx 0.85$ characteristic of graphite in the spectral range of 3-5 μm was used for temperature measurement. The transmission characteristics of the KRS-5 window were also taken into account.

The emissivity coefficient depends on the surface condition of the material and the measuring temperature. During the EBD, the temperature of the target surface changes in the range of several hundred kelvins. The surface of the target is modified by the electron irradiation and irradiation-induced heating. It will be shown later that the structure of the target surface changes from pristine C_{60} to disordered carbon upon EBD. Therefore, the emissivity of the target surface changes significantly during the EBD process affecting the accuracy of the temperature measurement as well. Hereafter, we will refer to the temperature measured by the pyrometer as the radiation temperature.

For fullerene C_{60} below 1000 K, the heat radiation is mainly emitted by infrared-active vibrations [15]. As C_{60} does not have infrared-active absorption

bands in the spectral range used by the pyrometer, its emissivity is thought to be significantly lower than that of graphite. Hence, in the beginning of EBD process the real temperature of the target surface is higher than the radiation temperature registered by the pyrometer. The difference between the real temperature and the radiation temperature is thought to decrease with time of electron-beam irradiation upon formation of the disordered carbon layer on the surface of the target.

The effective mass thickness of the coating deposited on the substrate was monitored by a quartz crystal microbalance (QCM). The mass density of the deposited material was chosen to be equal to that of fullerite (1720 kg/m^3) to recalculate the obtained data into the geometrical thickness. No micro-size pores or defects were found in the deposited films using the atomic-force microscopy measurement [14]. However, the mass density of fullerite increases upon polymerization and therefore, the obtained values of geometrical thickness are still somewhat overestimated depending on the film molecular structure.

A Faraday cup was placed into the substrate holder together with the QCM sensor. The Faraday cup was registering the current of scattered and true secondary electrons and ions from the target and the copper heating table used as the target holder. To summarize, kinetics of the radiation temperature of the target surface, thickness of the coating on the substrate and current irradiating the substrate during the EBD process was registered. The first-order derivatives of these three parameters representing growth rate of the coating, heating rate

of the target surface and the rate of the substrate irradiation change were also analyzed.

A Jasco NR 1800 spectrometer with a cooled CCD detector was used for Raman spectroscopy measurement of the fullerite target before and after the EBD process. The 488 nm line of an argon laser was used as the excitation source.

3. Results and Discussion

The whole process of fullerite C₆₀ EBD can be divided into an initial induction stage, a relatively short transitional stage and a stationary coating deposition stage (Fig. 2). During the induction stage, the electron beam irradiates the target yet the sublimation of the target material is negligible. The induction stage of fullerite C₆₀ EBD is characterized by target negative charging, target heating and irradiation-induced modification of the target surface layer.

The negative charging of the target is caused the following reasons. The coefficient of secondary electron emission (SEE) is below 1 for carbon-only materials in the primary electron energy range of 1000-1500 eV [16]. Moreover, fullerite C₆₀ has low conductivity at room temperature thus causing the negative charging of the target upon the beginning of the electron-beam irradiation. Whereas the SEE coefficient changes negligibly with temperature, we will discuss later how the increase of the fullerite conductivity upon heating can significantly influence the target charging.

Owing to the negative charging of the target surface at the beginning of the electron-beam irradiation, the density of the current of negative charges onto the substrate decreases from initial $\approx 1.1 \text{ mA/cm}^2$ to a value of $\approx 0.7 \text{ mA/cm}^2$ within few seconds (Fig. 3a). During the induction stage, the substrate is irradiated only by a flow of secondary and scattered primary electrons from the target and the target holder. A slight change of the geometry of the wire cathode of the electron gun upon its heating causes relatively small decrease of the primary electron beam current and hence, it can also cause the decrease of the current density on the substrate. However, in our opinion, the observed decrease is mainly caused by the charging of the target. The electric field of the negatively charged surface of the fullerite target is thought to suppress and scatter secondary electrons emitted from the target holder surface around the target.

First, we have measured the energy distribution of the electron flow on the substrate under the experimental conditions given without the fullerite target. The measurement was conducted using a Faraday cup with a copper mesh in front of it. A decelerating negative electrostatic potential from 0 to 300 V was applied to the mesh. The measurement showed that secondary electrons with energy of less than 50 eV constitute $\approx 37\%$ of the whole electron flow under the experimental conditions given. The value obtained is quite close to the decrease of the current density on the substrate observed during the EBD of fullerite.

The analysis of the kinetics of the current density on the substrate during the EBD of fullerite showed that its decrease upon the beginning of the electron-beam irradiation indeed can be fitted by a sum of two exponential decay functions (Fig. 3a). A function with the time constant of about 8 s and the amplitude of about 0.15 mA/cm², which we connect to the particularities of the electron gun operation, and a function with smaller time constant of about 1 s and larger amplitude of about 0.42 mA/cm². As explained above, we connect the second decay function with the suppression and scattering of SEE from the target holder.

During the induction stage, the primary electron beam is decelerated and scattered by the electrostatic potential of the target surface. Consequently, the power density of the electron beam on the target and its heating rate also decrease during the first few seconds of fullerite EBD (Fig. 3b). Fullerite C₆₀ has relatively low thermal conductivity (0.4 W/(m·K) for a C₆₀ crystal [17]). Thus, the fullerite target can be considered as a semi-infinite body during the first seconds of electron irradiation until the heating front reaches the bottom of the target. For a semi-infinite body, the heating rate of the surface is proportional to $q/\sqrt{t}$, where q is the power density of the heat flux and t is the time. However, the negative electrostatic potential of the target is thought to decelerate and defocus the primary electron beam. Therefore, we have used a $f(t) = (C_1 \cdot \exp(-t/\tau) + C_2)/\sqrt{t}$ formula with an exponential decay function instead of the constant q for fitting the experimental data (Fig. 3b).

The time constant $\tau \approx 0.88$ s, obtained by the fitting, is quite close to the value, obtained for the decay of the current density on the substrate ($\tau \approx 0.98$ s). This agreement proves that both decay processes are caused by the same reason, negative charging of the target surface. The obtained ratio of constants $C_1/C_2 \approx 30$ is determined by the ratio of the power density of the electron beam on the target before and after charging under the stipulation that the thermal physical properties of the target did not change during this short period. Based on this result, we expect the target negative potential of several hundred volts and significant scattering of the primary electrons by the electric field of the target.

After the negative charging of the target is completed, the rest of the induction period is characterized by further slow growth of the target temperature and irradiation-induced modification of a thin surface layer of the target. The penetration depth of the primary electrons into the fullerite target can be estimated by using a formula proposed by Kanaya and Okayama [18]. For 1.2 keV electrons it is ≈ 53 nm. It should be noted that the primary electrons are decelerated by the target negative charge and thus, the calculated value is an upper estimate.

The electron-induced modification of the surface target layer goes through polymerization to fragmentation and amorphization upon the increase of the exposure dose [9, 10]. However, fullerene polymers are metastable in

comparison with the monomer phase and undergo spontaneous dissociation to monomer in the temperature range of 400-600 K [19-22]. Therefore, heating and polymerization/fragmentation of the target material are two interconnected processes in the surface layer of the target whereas in the deeper layers, heat transfer prevails.

The duration of the induction period and scenario of the sublimation start strongly depend on the thermal physical parameters of the target and the thermal contact between the target and the target holder. Under the experimental conditions used, the target sublimation starts after approximately 3 minutes of electron-beam irradiation and goes through the pores in the modified surface layer. The porous surface layer can be seen as a thin flaky film having metallic luster on fullerite targets after the EBD process (Fig. 4b). On the other hand, preparation of thinner and denser targets of higher thermal conductance results in longer induction times and start of the sublimation through an explosive take-off of the flakes of the dense modified barrier layer by the overheated subsurface fullerenes. In the areas, where the surface film has been taken away, the direct surface sublimation prevents the recurring formation of the irradiation-induced barrier layer. These areas can be seen as the pristine-black eroded spots on the fullerite targets after the EBD (Fig. 4c,d).

The material of the thin modified surface layer represents amorphous carbon with inclusions of fullerene polymers. It is insoluble in toluene. It was purified by the repeated toluene extraction of unreacted C₆₀. The Raman

spectrum of the material drastically differs from the spectrum of the pristine C₆₀ (Fig. 5). Two broad and intensive peaks at around 1357 and 1585 cm⁻¹ are characteristic of disordered carbon structures with a mixture of sp² and sp³ carbon sites and known as D and G peak, respectively [23]. Among the less intensive peaks, the peak at 1465 cm⁻¹ can be assigned to C₆₀ polymers.

The non-stationary coating deposition stage of the fullerite C₆₀ EBD is triggered by the release of the accumulated negative charge with the first sublimation products. Consequently, the current density of negative charges onto the substrate increases from ≈ 0.7 to ≈ 1.0 mA/cm² (Fig. 6b). The observed increase corresponds to the electron flow density of approximately 1.9×10^{15} s⁻¹ cm⁻² whereas the maximum flow density of depositing fullerene molecules was $\approx 1 \times 10^{14}$ s⁻¹ cm⁻² (≈ 0.7 nm/s) under the experimental conditions given and the stipulation of 100% accommodation of the depositing molecules. Hence, the possible contribution of ion current is negligible. The recovered value of the current density is close to its value before the negative charging of the target surface (≈ 1.1 mA/cm²). Thus, the target may be only slightly charged during the following stages of the EBD process.

The increase of the power density of the electron beam on the target owing to its discharging enables a fast raise of the target surface temperature. The peak of the heating rate during the first 10 seconds of the sublimation (Fig. 6c) coincides well with the peak of the target discharging rate. The latter can be seen as a peak of the first derivative of the current density on the substrate (Fig.

6b). The fast raise of the target temperature has two main consequences on the EBD process: increase of the target conductivity and start of the intensive thermal sublimation.

The dominating mechanism of conductivity of fullerite C_{60} changes with heating. Upon reaching the sublimation temperature, it switches from activated hopping via band tail states to band conduction via delocalized states [24]. The corresponding increase of the conductivity is thought to enable a leakage current of the target surface charges into the target holder, significant enough to prevent charging of the target during the stationary deposition stage of EBD. Indeed, we have shown that the content of negative fullerene ions in the sublimation products is negligible during the stationary stage [14]. Therefore, the sublimation enables the release of the surface charges only during the transitional non-stationary deposition stage whereas the leakage current through the target comes into play after the target got sufficiently heated.

The start of the intensive thermal sublimation of the target material (Fig. 6a) owing to increase of the target temperature introduces a new factor into the heat balance of the target. In addition to the heat transfer through the target, the convection heat extraction by the sublimation also starts cooling the target surface. These factors slow down the heating of the target by the electron beam, the power density of which increases owing to the decrease of the target electrostatic potential. As a result, the temperature of the target stabilizes by the end of the transitional stage.

As the sublimation goes through the pores in the amorphous carbon surface layer, microcavities appear and grow under the pores. Indeed, in the case of the direct sublimation from the surface, the sublimation rate is proportional to the target surface temperature. With the stabilization of the temperature, the sublimation rate is also thought to stabilize and not to change significantly until the target is used up. Such sublimation kinetics is observed when parts of the modified surface layer had been taken off during the sublimation start. In the case of the sublimation only through the pores, the sublimation rate from the whole target is proportional to the vapor pressure inside the microcavities. At the constant temperature of the microcavity walls, the pressure inside it is proportional to the sublimation area of the walls and inversely proportional to the volume of the microcavity. As the volume of the microcavity increases faster than its inner surface, we observe a peak on the graph of the sublimation rate rather than just a step-like increase (Fig. 6a).

The modified surface layer of amorphous carbon plays an important role during the stage of the stationary coating deposition as well. The layer is formed during the induction stage by the primary electrons decelerated in the electrostatic field of the charged target. During the coating deposition stage, the target may be only slightly charged and the primary electrons penetrate through the modified surface layer into the microcavities under it. For electrons, the maximum of the ionization cross section of C_{60} lies at around 80-100 eV energy [25]. Hence, the ionization of C_{60} vapor is facilitated inside the microcavities in

comparison with direct sublimation from the surface. Indeed, we have previously shown that the products of fullerite C₆₀ EBD during the stationary deposition contain few mass percent of positive fullerene ions with the energy of up to ≈ 7 eV [14] that strongly influence the coating structure.

We define the end of the transitional stage and beginning of the stationary coating deposition stage by the stabilization of the coating growth rate and the current density on the substrate. These process parameters determine the conditions of the coating formation on the substrate and therefore, its structure. The coatings used for structural studies described in [14] were formed during the stationary deposition stage. We used a shutter to cover the substrates during the first two stages.

The stationary deposition stage is characterized by the current density on the substrate of about 0.95-1 mA/cm² and the coating deposition rate of 0.01-0.05 nm/s. It should be mentioned that the deposition rate slowly decreases, however within the time limits necessary for depositing 100-200-nm-thick coatings on the substrate, the decrease is negligible. The deposition of the C₆₀ EBD products onto the substrate under the conditions described results in the formation of a thin film containing a mixture of fullerene C₆₀ and its polymeric forms.

4. Conclusions

Experimental data on the target surface temperature, density of the flow of charged particles on the substrate, deposition rate of the coating of C₆₀, structure of the modified surface layer of the target has been analyzed. It was shown that the process of irradiation of a pressed fullerite C₆₀ target with an electron beam of energy of 1.2 keV and current density of $\approx 10 \text{ mA/cm}^2$ can be considered as a process consisting of several stages.

The first, induction stage (duration of approximately 3 minutes) is characterized by the negative charging of the target and decrease of the current density of the electron beam on the target, heating and radiation-induced modification of the surface layer.

The second, transitional stage (duration of approximately 3 minutes) begins with the loss of the target mass with sublimation of fullerene molecules through the modified surface layer leading to the release of the negative charge and increase of the power density of the electron beam on the target. Sublimation through the pores in the surface layer results in the formation of microcavities under the surface. As a result of the increased power density of the electron beam on the target, the target temperature and consequently, target conductance and sublimation rate also increase. Convection cooling by sublimation and decrease of the pressure inside the microcavities leads to the stabilization of the process.

During the third, quasi-stationary stage, the coating is deposited onto the substrate at a constant rate of 0.05-0.1 nm/s under the electron and ion bombardment leading to formation of polymerized fullerene structures.

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Fig. 1. Experimental setup for electron-beam dispersion of fullerite C_{60}

Fig. 2. Kinetics of the coating deposition rate and the current density of negative charges on the substrate during the EBD of fullerite C_{60}

Fig. 3. Kinetics of the current density of negative charges on the substrate (a) and the radiation temperature and heating rate of the target surface (b) with respective fitting functions (dashed lines) during the negative charging of the fullerite target under the electron-beam irradiation

Fig. 4. Photographs of the fullerite C_{60} target before (a) and after the process of EBD (b, c, d); in the case of target (b), the sublimation was going through the pores in the surface layer whereas in the case of targets (c) and (d), parts of the surface layer were explosively taken off during the start of sublimation

Fig. 5. Raman spectra of pristine fullerite C₆₀ powder and the electron-modified surface layer of the fullerite target after the EBD process

Fig. 6. Kinetics of the coating deposition (a), current density of negative charges on the substrate (b) and radiation temperature of the target surface (c) with the respective first derivatives during the transitional stage of non-stationary sublimation (time is set to 0 at the moment of the coating deposition beginning)