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TEM and EELS characterization of carbon dust and co-deposited layers from the TEXTOR tokamak

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Abstract

Co-deposited layers and dust agglomerates formed at TEXTOR tokamak were collected and examined in detail by means of transmission electron microscopy and electron energy-loss spectroscopy. The analysis has shown that thick deposits and dust contain mainly carbon with an admixture of boron precipitates and small amounts of other elements. The carbon deposit was mostly amorphous, while strips were graphite crystallites embedded in dust agglomerates. In thin co-deposits on the collector probe very small graphite crystallites were piled up with a preferential orientation. The results are discussed in terms of processes decisive for the erosion of plasma facing components and for the fuel retention.

1. Introduction

The formation of co-deposited layers on plasma facing materials in a D-T burning machine derives from plasma–material interactions, such as erosion, ionization, transport, charge-exchange and co- or re-deposition. Therefore, the layers 'bear' the information on these complex processes. As a result, the structure of co-deposits depends on the temperature of the substrates and their position relative to the plasma configuration in a torus, and on energy, flux and angle of incidence of incoming ions. In addition, the wall conditions in the machine are particularly important for the dust formation. It is known that hydrogen retention or absorption in graphite is highly related to the structure of carbon materials [1–3]. In general, the more the structure is disordered, the higher hydrogen retention occurs. These facts motivate us to conduct a detailed structural analysis of co-deposited layers. In our previous preliminary work we reported the analyses of layers formed on a graphite tile of the toroidal belt limiter (ALT-II) of TEXTOR [4]. Studies carried out by means of transmission electron microscopy (TEM) and electron energy-loss spectroscopy (EELS) revealed that the layer consisted of amorphous graphitic carbon containing a high density of crystalline boron precipitates and a detectable amount of oxygen. The structure is very similar to the one observed for B-doped graphite irradiated by H^+. The considerable amount of boron comes from routinely employed boronization, and oxygen from the plasma itself, which contains about 3% oxygen as an impurity.

In the present study we have extended the previous analysis to other carbon residues from several plasma facing components in the TEXTOR tokamak, such as the deposited layer from other limiter tiles, dust agglomerates collected from the device floor and thin co-deposits grown on a surface of the collector probe exposed to the scrape-off layer plasma. Based on the detailed structural analysis,

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the effect of deposition temperature and deuterium (tritium) retention will be discussed.

2. Experimental

A sample of a thick co-deposit was taken from an ALT-II graphite tile (#20). The construction of the limiter tiles is shown in [4]. The layer was removed with an adhesive tape. One of the fragmented pieces was pasted on a single-hole copper disc and then thinned by argon ion-milling until the sample was perforated. A sample of carbon dust was collected with a vacuum cleaner after about 14 000 plasma shots and then it was crashed into fine powder in an agate mortar with a pestle. The powder was dispersed in ethanol and skimmed off by a copper mesh with carbon-coated plastic microholes. The formation of thin co-deposits on the collector probe surface was recognized by a pattern of interference fringes. A part of the deposit was removed from the substrate using a copper grid covered with glue.

The samples were examined with a JEOL-JEM200CX TEM (operated at 160 kV) equipped with a parallel EELS, a Gatan DigiPEELS model 766. High-resolution imaging was also conducted with a Hitachi H-1250ST high-voltage electron microscope, operated at 1 MV. Energy-filtering TEM was also applied to obtain elemental distributions with a Gatan Energy-Filter (GIF) attached to a JEOL-3000F TEM.

3. Results

3.1. Characterization by TEM and electron diffraction

The internal structure of the co-deposited layers on the ALT-II tile consists of amorphous graphitic carbon and crystalline boron precipitates. Observations carried out in several areas of the sample confirm that the density of boron precipitates largely varies from place to place. This result is in agreement with previous observations for another tile (#22) from the same limiter [4].

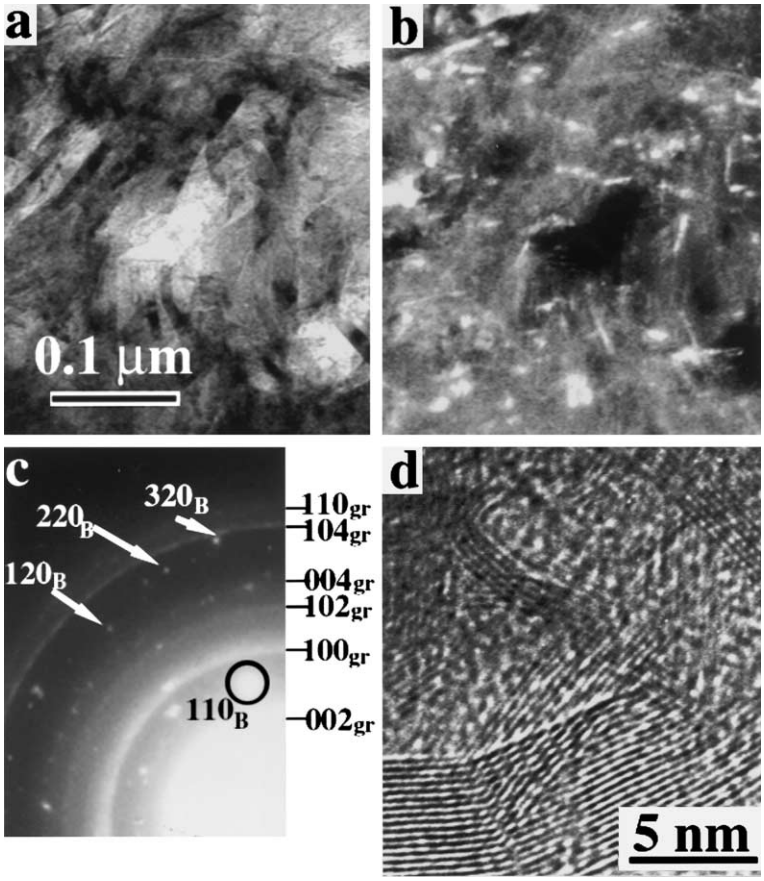

Fig. 1. TEM images of carbon dust: (a) bright-field image, (b) dark field image taken from the spot encircled in (c), (c) electron diffraction pattern in which the Debye rings and discrete spots are indexed. The subscript 'gr' is referred to graphite and 'B' to crystalline boron. (d) High-resolution image of a portion of the dust.

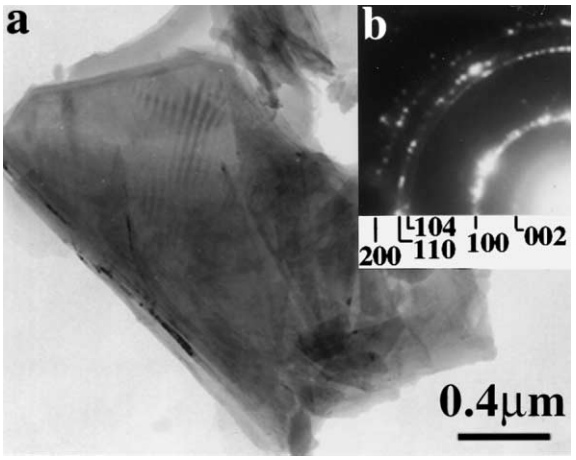

Fig. 2. Bright field TEM image of deposited layer scraped off from the collector probe surface (a), and electron diffraction pattern (b).

TEM images and an electron diffraction pattern from the dust agglomerates are shown in Fig. 1(a)–(d). The dark field image (b) was taken from the spot encircled in (c). It is seen in the images (a) and (b) that the substructure consists of tangled thin strips and small precipitates. The diffraction pattern can be interpreted as a mixture of randomly oriented crystallites of boron and graphite. Narrow strips seen in the high-resolution image (d) are attributed to bent and tangled layers of graphite.

A TEM image and a corresponding electron diffraction pattern of the thin deposits scraped off from the collector probe surface are shown in Fig. 2. The Debye rings are mainly indexed by ($hk0$)-type Miller indices of graphite, though the (0 0 2) and (1 0 4) rings also faintly occur, as shown in Fig. 2(b). This suggests that the co-deposit consists of preferentially oriented graphite, i.e. the graphite layers are stacked with the basal plane nearly parallel to the collector probe surface but randomly rotated around their c-axis. High-resolution observation confirmed that the coherent thickness of the graphite layer is in the range from 3 to 10 nm. The basal planes are not curved or bent unlike those in the samples of dust.

3.2. EELS measurements and mapping of elemental distribution

EEL spectroscopy is well suited for light-element analysis, in particular for the determination of local chemical states of constituents. The detection limit of the technique is, however, not better than 1 at.%. The co-deposited layers on the limiter and the samples of dust consist mainly of carbon, boron and a detectable amount of oxygen. The boron content significantly varies, from 20 at.% to about 70 at.%, in different parts of the analyzed materials. In co-deposits on the collector probe only carbon is detected by EELS, though significant amounts (≃10–500 ppm) of deuterium, tungsten and other elements were detected by ion beam analysis techniques which are more sensitive than EELS but do not allow any information on the chemical state of the constituents. Therefore, in the following section we focus on the chemical state of carbon and boron as determined by EELS.

The K-shell absorption edge spectra of boron from the present samples are shown in Fig. 3. For comparison, the spectra for crystalline boron and B_4C are inserted in the figure. Comparing the obtained spectra with those recorded for known materials, boron in the carbon dust is considered to be in a crystalline form. It is consistent with the electron diffraction pattern shown in Fig. 1(c). In contrast, the electron energy loss near edge fine structure (ELNES) spectrum for the co-deposit on the limiter tile resembles to some extent the one for B_4C better than that from a pure boron crystal, accompanying by a small but meaningful chemical shift compared to the onset of pure boron absorption edge. This suggests that a considerable fraction of boron in the co-deposited layers occurs in a form different from the pure crystalline state.

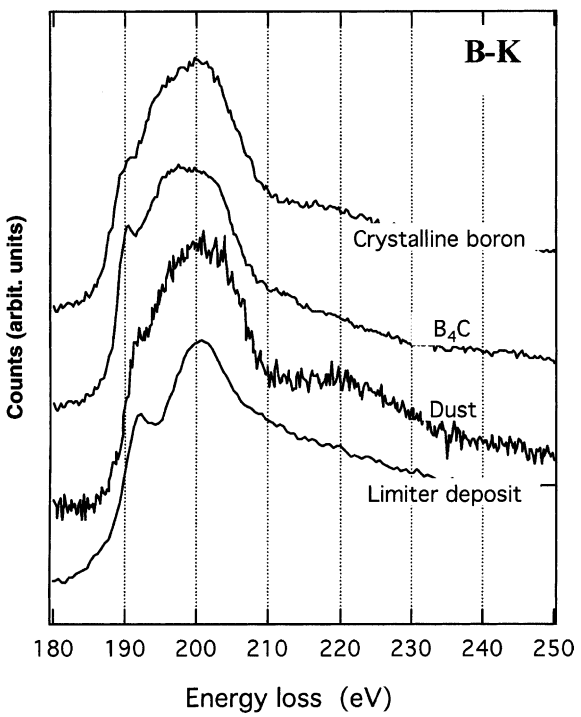

Fig. 3. ELNES of B-K edges acquired from limiter deposits and dust in comparison with those from boron and B_4C crystals.

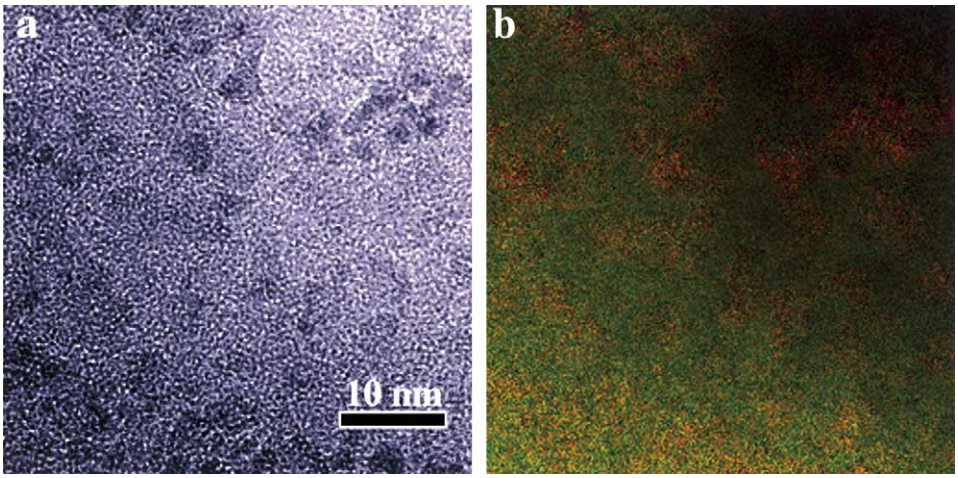

Fig. 4. Energy-filtered images of the limiter co-deposit: (a) zero-loss image, (b) elemental distribution map of boron (red) and carbon (green).

In order to know the spatial distribution of boron in co-deposits from the limiter tile layers, energy-filtering imaging was applied. In Fig. 4(a) and (b) an image taken with elastically scattered electrons only and an elemental distribution map (boron and carbon) are taken. In Fig. 4(b) boron and carbon distributions were separately imaged by selecting the respective K-edge spectrum portions with the energy-selecting slit on the EELS spectrum, and then the two images were superposed in order to illustrate the distribution of boron (red) and carbon (green).

One notices dark agglomerates, a few nanometers in size, scattered in image 4(a). These are most likely boron nanocrystals because these areas correspond to boron-enriched regions as shown in image 4(b), and the dark contrasts should be interpreted by the diffraction contrast, considering the similar electron scattering power of boron and carbon. Boron is widely distributed over the whole area under investigation, also in regions other than those with distinct B segregation. This fact supports the statement that a specific B-ELNES feature of the co-deposits (as shown in Fig. 3) is attributed to boron embedded in an amorphous carbon matrix.

The carbon K-edge spectra are shown in Fig. 5. For highly oriented pyrolytic graphite (HOPG) in a reference spectrum, the σ^*-ELNES of graphite should reveal a 4-peak fine structure because of the sixfold symmetry [5]. The spectra for the dust and the collector probe both show the same feature as that from HOPG, consistent with the TEM observations above. The σ^*-ELNES for the co-deposit from the limiter tile is different: it shows a broad and structureless peak, because the local sixfold symmetry is lost in the amorphous state, similar to that from amorphous carbon (a-C) produced by prolonged electron irradiation in TEM. It should be noted that the

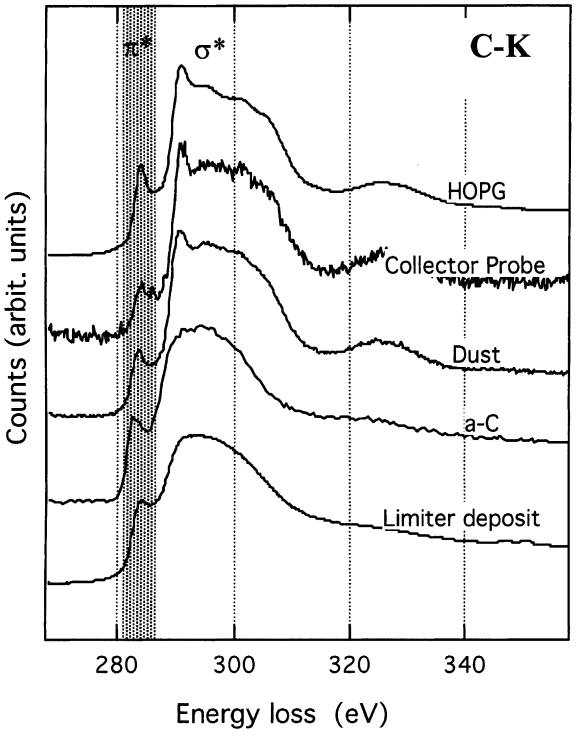

Fig. 5. ELNES of C-K edges from the present samples and other reference samples. HOPG stands for highly oriented pyrolytic graphite.

π^* peak unambiguously occurs with their areal ratio to the σ^*-peaks nearly the same as that of a-C. This indicates that the amorphous carbon matrix of the co-deposit from the limiter consists of nearly 100% sp^2 bonding. That fact and the B-ELNES feature mentioned

above lead us to a conclusion that boron atoms dispersed in the amorphous carbon matrix are incorporated in the graphitic threefold sp^2 network.

We also applied the angle-resolved EELS to record the C-K edge characteristic for thick co-deposits (from ALT-II) to examine the preferential orientation of the structure. No anisotropy was found, unlike in the previous observation [4].

4. Discussion

The results show that the degree of the structural order in the co-deposited layer on the ALT-II limiter is the lowest, whereas the one best orientation of material is found in thin layers on the collector probe.

It should be noted that the surface of the ALT-II limiter crosses the magnetic field lines at the angle of $\approx 2°$. It means that the majority of incoming high-energy carbon ions are reflected, whereas low energy species preferably produce co- or re-deposits. In addition, the base temperature of the ALT-II limiter was about 600 K far below the graphitization temperature of about 1600 K. As a result, the structure of the co-deposits on the limiter is amorphous but still keeps sp^2 bonding and contains fairly homogeneously distributed significant amounts of boron in the rather homogeneous amorphous carbon matrix. In addition, small crystalline boron precipitates originating from the boronization are present in the layer.

In case of the collector probe, the magnetic field lines cross the exposed surface nearly perpendicularly. As a result, only a fairly small fraction of the incident particle flux is reflected. Accordingly, the co-deposits formed consist of quite well crystallized graphite particles retaining layered structure as a consequence of the growth on shot-by-shot basis. In such layers, the deuterium retention is expected to be lower than that of the co-deposits on the limiter.

Assuming that the dust originates from the exfoliation of co-deposited layer on the limiter, its temperature can rise high during the exposure to the plasma. As a result, in dust agglomerates the amorphous structure of the co-deposited layer is re-crystallized. This was indeed observed with TEM. Therefore, the deuterium retention in dust is lower than in co-deposits tightly adhered to the limiter. Very small boron precipitates in the dust are most probably attributed to the segregation of this element during the re-crystallization process. This is in contrast to a fairly homogeneous boron distribution in amorphous thick co-deposited on the limiter. Therefore, the result confirms a significant plasma-stimulated temperature rise in the dust. The observed differences of the structure between dust and thick co-deposits clearly prove that the dust grains cannot simply be treated only exfoliated co-deposits.

5. Summary

Co-deposited layers and dust agglomerates formed at TEXTOR tokamak were examined in detail by means of TEM and EELS. The deposited layer on an ALT-II limiter tile was mostly amorphous including a considerable amount of boron, in forms of crystalline precipitates and B–C composites. The thin co-deposits on a collector probe consisted of very small graphite crystallites piled up with the basal planes nearly parallel to the probe surface. These morphologies can be understood by the energetics of incident ions during the plasma experiments. On the other hand the dust agglomerates, probably formed by the exfoliation of the co-deposited layer from the limiter surface during plasma shots, were admixtures of tangled strips of graphite and crystalline boron particles. This predicts a considerable plasma-assisted temperature increase of the dust during its formation process.

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