ELSEVIER

Journal of Nuclear Materials 307–311 (2002) 1294–1299

journal of nuclear materials

www.elsevier.com/locate/jnucmat

Microstructure and deuterium content of tokamak T-10 carbon erosion products

P.V. Romanov [a], B.N. Kolbasov [a,*], V.Kh. Alimov [b], V.M. Gureev [a], A.G. Domantovskij [a], L.N. Khimchenko [a], P.N. Orlov [a]

[a] *Nuclear Fusion Institute, Russian Research Center 'Kurchatov Institute', Kurchatov Sq. 1, 123182 Moscow, Russia*
[b] *Institute of Physical Chemistry, Russian Academy of Sciences, Leninskij Prospect 31, 117915 Moscow, Russia*

Abstract

An analysis of erosion products deposited inside the T-10 tokamak vacuum chamber revealed the presence of two groups of substances: carbon films and dust microparticles. Specimens of the carbon films were investigated using scanning and transmission electron microscopy. In terms of microstructure, the films appeared to be either smooth (homogeneous) or globular, that is, formed by globe-shaped microparticles. Dust specimens were also collected and analysed. Particles with count median diameter of 10–15 nm were detected. The deuterium concentration in the sampled erosion products was measured using the SIMS/RGA technique.

1. Introduction

The object of this study was to characterise the hydrogenated carbon erosion products (films and dust) formed in the T-10 tokamak operating in the Kurchatov Institute. The content of hydrogen isotopes in the carbon erosion products and their mobilisation also were under study.

Table 1 presents the key parameters of the T-10 tokamak which has a stainless steel vacuum chamber and is equipped with a movable limiter and a fixed ring-shaped diaphragm. Both of these plasma facing components are made of fine-grade pyrolitic graphite MPG-8.

The operational availability and safety of the International Thermonuclear Experimental Reactor (ITER) are affected by the erosion of plasma facing materials, which requires the divertor replacement and the in-vessel tritium inventory control. The hydrogen-to-carbon (H/C) atomic ratio in soft co-deposited carbon films can exceed unity [1]. It is important to determine the rates of hydrogen and carbon isotope removal from hydrogenated carbon films resulting from temperature changes. Another issue is dust: its detrimental effects as well as mobilisation and catalytic properties depend on the size of dust particles.

Some of these problems are studied using both operating tokamaks and laboratory facilities simulating ITER conditions. The characteristics of co-deposited layers are reviewed in [1–7]. A series of dust characterisation studies were recently performed at INEEL (USA) by Carmack et al. using the DIII-D and TFTR tokamaks and the SIRENS accelerator [8–10]. Studies conducted in Russian laboratories focused on the simulation of ITER plasma disruptions [11,12]. In the latter studies, dust particle size distribution histograms relating to plasma disruption conditions showed two distinct peaks – at 10–30 nm and 2–4 μm.

2. Dust and film collection

The previous work has shown that a reliable dust sampling technique is required to obtain representative dust specimens suitable for the analysis.

* Corresponding author. Tel.: +7-095 196 7583; fax: +7-095 947 0073/196 7909.
E-mail address: kolbasov@nfi.kiae.ru (B.N. Kolbasov).

0022-3115/02/$ - see front matter
PII: S0022-3115(02)01064-4

Table 1
T-10 tokamak major parameters

Minor vacuum chamber radius, m	0.39
Major vacuum chamber radius, m	1.5
Magnetic field, T	2.0–2.4
Plasma current during 2000 campaign, kA	100–330
Plasma discharge duration, s	<0.5
Electron temperature in plasma core, keV	1.0–3.0
Electron temperature near the wall, eV	15–20
Ohmic heating power, kW	100–400
ECR heating power, MW	1.0
Plasma density in plasma core, cm^{-3}	$(8–30) \times 10^{12}$
Plasma density near the wall, cm^{-3}	1×10^{12}

Like in the DIII-D and TFTR, we collected dust specimens from the vacuum chamber surfaces by vacuum suction onto filter substrates following the vacuum chamber vents after the completion of the experimental campaigns. Erosion products were vacuumed through 1–3 μm pore size ash-free nitrocellulose filters or through basalt-fiber or glass-fiber filters with fiber diameters ranging from ≃0.3 to ≃1 μm.

The dust specimens of the second type were produced during the tokamak plasma discharges and deposited in situ onto substrates fixed on a movable rod within the vacuum chamber nearby the limiter and the diaphragm, 225 mm away from the plasma column. The location of the rod carrying a dust collector is shown in Fig. 1. The rod could be removed from the chamber between chamber vents. Basalt-fiber, glass-fiber, quartz-fiber filters, and copper meshes covered with amorphous carbon films served as dust collectors.

Specimens of carbon films were collected within the vacuum chamber in cross-sections A and D, that are mutually perpendicular in the toroidal direction (see Fig. 2). Due to the proximity of cross-section A to the limiter and the diaphragm, the temperature there is much higher than at cross-section D, which is about 2.4 m away from these components. Access to the vacuum

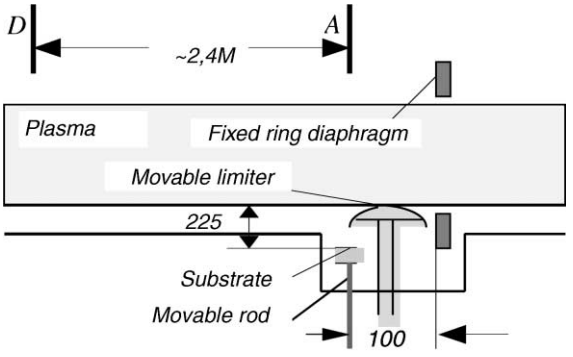

Fig. 1. Arrangement of the movable limiter and the ring diaphragm in the T-10 tokamak vacuum chamber.

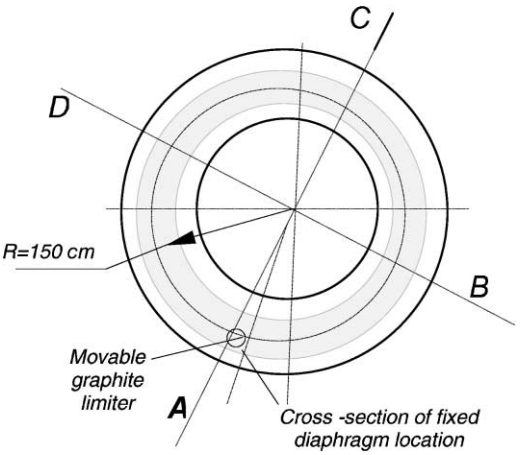

Fig. 2. Cross-sections of the T-10 vacuum chamber.

chamber bottom was provided by removing top flanges at cross-sections A and D. There is also a bottom flange at cross-section A which was removed during vents. It contained erosion products on its plasma facing surface.

3. Film and dust microstructure

The microstructure of films collected in the T-10 vacuum chamber was studied using an optical microscope, a scanning electron microscope (SEM) and a transmission electron microscope (TEM). The study revealed the existence of smooth, or homogeneous, and globular film types [3,13,14].

The globular films were collected by vacuum suction from the lower surfaces of the vacuum chamber interior (particularly the diaphragm bottom), at cross-section A. They have probably flaked off from their initial locations during the venting, e.g. at air ingress into the chamber. Such films were also found on the bottom flange at the same cross-section and were extracted from oil used in the T-10 vacuum pumping system. One can see from Fig. 3(a) that some of the film on the diaphragm is peeling off and has cracks.

A relatively low-magnification SEM microphotograph of the globular film surface (Fig. 4) shows a rather uniform cauliflower pattern over the specimen surface. The globule diameter (up to 30 μm) is commensurable with the thickness of the film. This suggests that the film is, in fact, a monolayer made up of globules.

A diffraction laser microanalyzer with the resolution of 0.22–1100 μm was used to determine the size distribution of microparticles making up the globular films. A histogram showing such a microparticle size distribution is given in Fig. 5.

Smooth carbon films are formed at some distance from the limiter and the diaphragm, in particular at

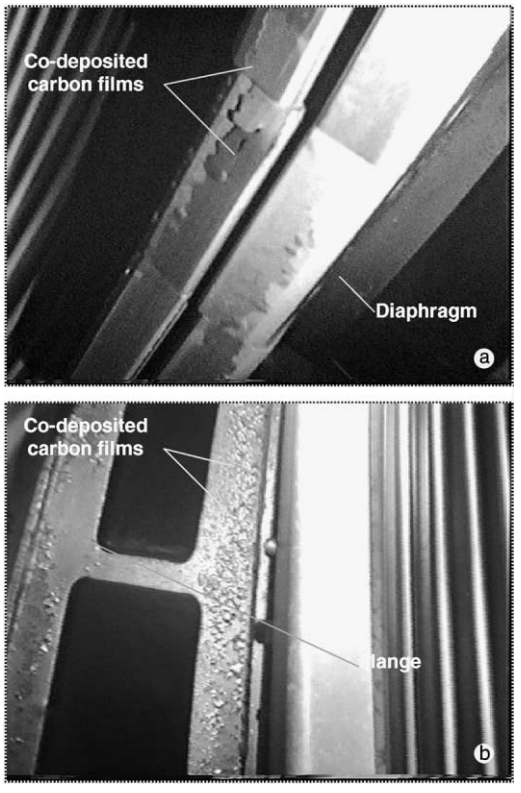

Fig. 3. (a) Segment of the ring diaphragm nearby cross-section A, covered with globular films; (b) bottom flange at cross-section D, covered with smooth films.

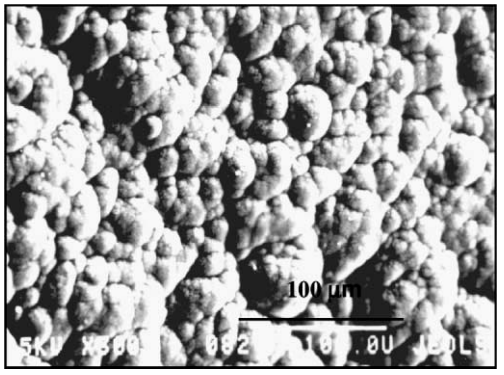

Fig. 4. Microstructure of a globular film formed close to the T-10 limiter and diaphragm.

cross-section D. The surface of these films has cracks and other defects (Fig. 3(b)) causing their flaking and separation. They were easy to remove mechanically (by scraper) or by vacuuming.

In terms of microstructure, the surface of smooth carbon films is very different from that of the globular

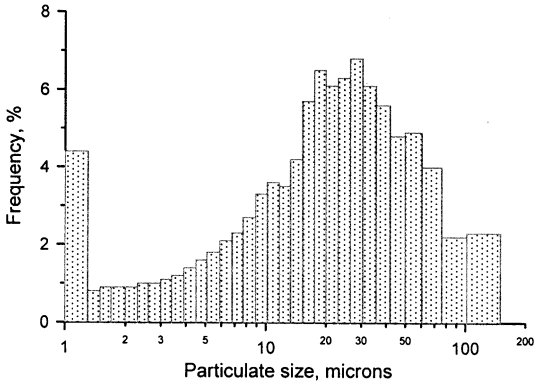

Fig. 5. Histogram of particle size distribution in a globular film deposited on the T-10 diaphragm surface.

films discussed above. The relief of films vacuumed from surfaces facing both the chamber wall and the plasma (see Fig. 6) was virtually the same. The only difference was the amount of impurities. No microparticles forming the smooth films are observed. This means that the smooth films have probably been formed from microparticles smaller than 100 nm. The thickness of the smooth films is from 3 to 20 μm.

The smooth carbon films collected in the T-10 vacuum chamber were of two types: black, with soot particles on the plasma facing surface (No. 1); and reddish-gold semitransparent films (No. 2). The difference in colouring of carbon films usually reflects different temperatures of the film formation: the black films are formed at higher temperatures than the reddish-gold ones. Both film types have practically the same microstructure.

The SEM and TEM images (Fig. 7(a) and (b), respectively) of dust microparticles collected from the T-10

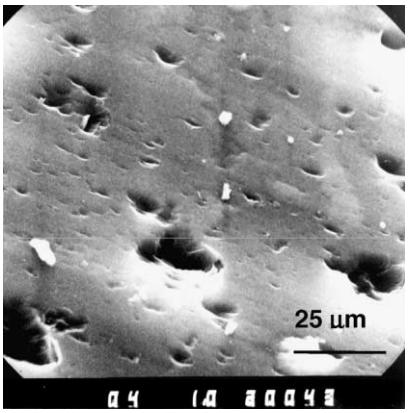

Fig. 6. Top view (at an angle of 20°) of the plasma facing side of a smooth film deposited inside the T-10 vacuum chamber at cross-section D.

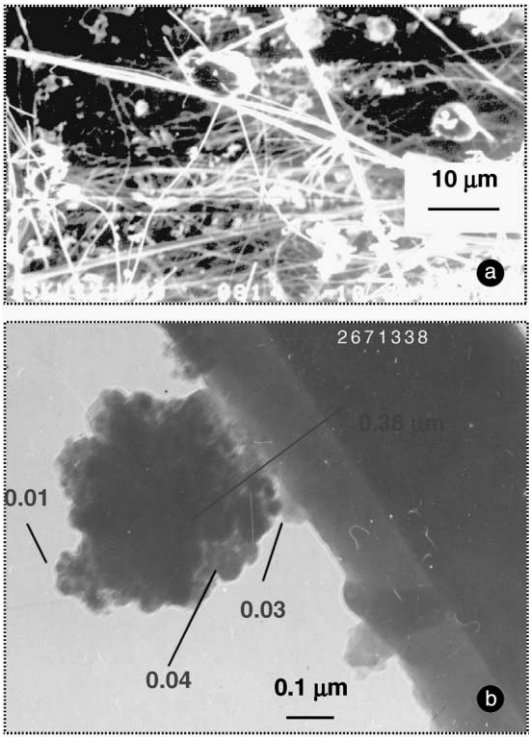

Fig. 7. Glass-fiber filter with dust microparticles vacuumed from inside the T-10 vacuum chamber: (a) SEM; (b) TEM.

vacuum chamber onto a glass-fiber filter reveal a considerable size variation: from submicron particles with diameters of only 10–40 nm to relatively large (several tens of microns across) agglomerates of globe-shaped microparticles with diameters of 10–40 nm each.

4. Deuterium concentration measurements

The concentration of deuterium in hydrogenated carbon films was determined using a combination of secondary ion mass spectroscopy (SIMS) and residual gas analysis (RGA) techniques. Measurements were performed while the film surface was sputtered with Ar ions to the depth of 0.4–1.0 μm.

Various forms of hydrocarbon films can be distinguished by the content of (sp^2)C, (sp^3)C and hydrogen [15]. Previous studies involving the analysis of carbon–deuterium substances [16] showed that SIMS detects only D atoms bound to (sp^2)C atoms. The other part of deuterium, trapped by sp^3 orbitals of C atoms can be detected as D_2 molecules by RGA.

We assume that the sputtering rate of hydrogenated carbon films is roughly equal to that of pure graphite. The sputtering rate of a highly ordered pyrolytic graphite was estimated with an accuracy of 20% as a ratio of the depth of a crater produced on the graphite surface by sputtering to the sputtering time [16].

The SIMS D^- and CD^- signals are well correlated and reflect D atoms bound up with (sp^2)C atoms, as mentioned above. The intensity of the C^- reference signal was used for standardization of the SIMS signals. In this approach, neither the changes in the primary argon ion current nor the position of the hydrocarbon film being analysed influenced the D concentration measurements. The SIMS signal was calibrated by measuring the $D^-$and CD- signals in graphite implanted with 3 keV D ions at 300 K to a fluence of 5×10^{22} D/m^2, so that the maximum atomic ratio D/C for D atoms associated with (sp^2)C atoms was 0.12. The uncertainty of SIMS calibration was about 20%.

The amount of D_2 molecules, released from sputtered layers and reflecting the trapping of D atoms by the sp^3 orbitals of C atoms, was estimated based on the D_2 sensitivity factor for RGA quadrupole mass spectrometry (QMS) and the known pumping speed. The uncertainty of RGA QMS calibration was about 40%. The use of two quadrupole mass spectrometers made it possible to register the SIMS and the RGA signals simultaneously.

In graphite irradiated with D ions at 300 K the maximum atomic D/C ratio for D atoms bound up with (sp^3)C atoms was of 0.2 [16]. Hence, taking into account the concentration of D atoms bound up with (sp^2)C atoms, we found that the maximum value of the total atomic D/C ratio in the D-irradiated graphite measured by the combined SIMS/RGA technique was 0.32. This ratio is very close to the value measured by nuclear reaction analysis [17].

We used also the Rutherford backscattering (RBS) and elastic recoil detection (ERD) methods to verify the data obtained in SIMS/RGA measurements. The component composition was determined by the RBS method. Absolute hydrogen isotope concentrations and depth profiles in the co-deposited carbon films were obtained by the ERD analysis [18].

4.1. Deuterium concentration in carbon films

Depth profiles of deuterium concentration were determined from both sides of deuterated carbon films collected within the T-10. The results are shown in Fig. 8.

Under the combined SIMS/RGA analysis, deuterium in the smooth films was detected only by RGA and was not observed in the SIMS measurements. This suggests that practically all the deuterium atoms are bound to the sp^3 orbitals of C atoms. The atomic D/C ratios in smooth films increase from the surface value to the level of about 0.37 and 0.78 for films No. 1 and No. 2, respectively (Fig. 8(a)). In deep layers (>250–300 nm from the surface) the D/C ratios remain at these levels practically

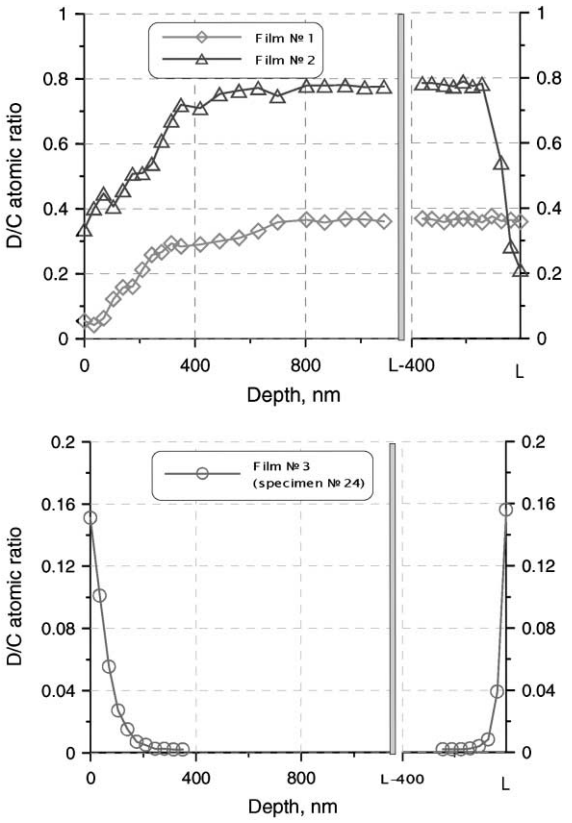

Fig. 8. D/C atomic ratio profiles for specimens of smooth (a) (top) and globular (b) (bottom) co-deposited films collected in the T-10 tokamak vacuum chamber. L – total film thickness.

constant. The average values of steady-state deuterium concentrations determined by SIMS/RGA and ERD/RBS methods are in agreement within the experimental uncertainty of 20% for every type of films [18]. Considering the high concentration of deuterium in smooth films, we infer that the films show properties typical of soft, polymer-like a-C:D layers [3]. The densities of C atoms in such layers are between 4.4 and 4.5×10^{28} m^{-3}.

Globular films have a D/C atomic ratio of $(2.0 \pm 0.5) \times 10^{-3}$ in the bulk of the specimen. Higher values observed at the surface (up to 0.15) can be attributed to surface saturation with gaseous deuterium available in the vacuum chamber (Fig. 8(b)).

4.2. Deuterium concentration in dust

The deuterium concentration in dust specimens vacuumed from the T-10 diaphragm bottom onto a glass-fiber filter was also measured by the SIMS/RGA technique and is shown in Fig. 9.

Unfortunately, the vacuum suction method does not ensure that all the dust is collected by filters. We believe that dust settling on upper filter layers provides a more

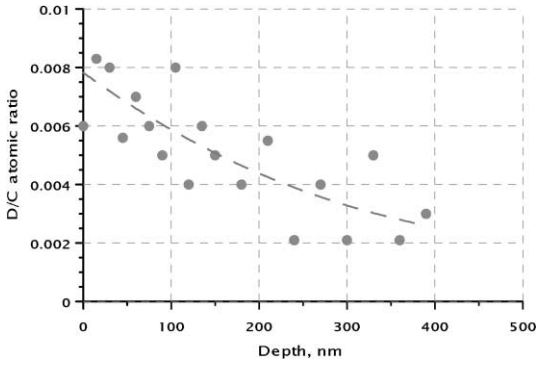

Fig. 9. D/C atomic ratio profile for dust specimens vacuumed from the T-10 diaphragm and deposited onto a glass-fiber filter.

representative picture of the whole spectrum of dust particles occurring in corresponding tokamak areas than the material that had passed through the upper layers and deposited on lower ones. The D/C atomic ratio in dust captured by the upper filter layers is about 0.01, which is far less than what we observed in the smooth films. Apparently, this is due to a much higher temperature nearby the limiter and the diaphragm where dust was formed, than at a distance from these elements, where the smooth films co-deposited.

4.3. Effect of annealing on D/C atomic ratio in smooth carbon films

Fig. 10 shows effect of the annealing in air on the D/C atomic ratio in smooth carbon films. The initial film thickness was 20–25 μm. The annealing duration, at all the temperatures, was 32 h. For every annealing, fresh films were used. Deuterium concentration was measured by the combined SIMS/RGA technique. We present our data along with results obtained by Causey et al. [7,17], who studied hydrogen isotope release from ≃50 μm

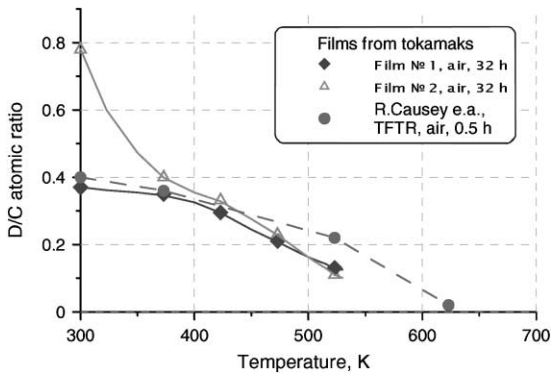

Fig. 10. D/C atomic ratio in co-deposited smooth carbon films formed in T-10 and TFTR vs. temperature of annealing in air.

thick carbon layers deposited on TFTR tiles under their annealing in air for 0.5 h.

The D/C atomic ratios in films deposited on TFTR tiles agree well with those in the smooth film No. 1 practically across the whole range of temperatures, from room temperature to 673 K. The small difference between the D/C concentrations in the range from 423 to 623 K is probably due to a different annealing duration in our experiments and those by Causey et al. The D/C atomic ratios for film No. 2 are the same at temperatures above 370 K.

5. Conclusions

The erosion products deposited on the T-10 in-vessel surfaces consist of co-deposited carbon films and dust particles.

We observed dust particles of submicron size with a few tens of nanometers across. Some of form agglomerates that had apparently been generated in plasma. There are also dust particles of micron-size.

Two types of co-deposited carbon films were found in the T-10 tokamak. The first type are smooth films, which can be either black, containing soot particles on the plasma facing surface, or semitransparent reddish-gold films. The black and reddish-gold smooth films have practically the same microstructure. The carbon films of the second type have a globular microstructure.

The D/C ratios in the smooth films increase from the surface value to the level depending on the film type – 0.37 for the black films and 0.78 for reddish-gold ones. In deep layers (>250–300 nm from the surface) the D/C ratios remain at these levels practically constant.

In the globular films, the D/C ratio (<0.01) is far less than in the smooth films.

The most reliable value of the D/C atomic ratio in the carbon dust collected from the T-10 diaphragm bottom is about 0.01.

Acknowledgements

The authors would like to thank the staff of the T-10 Tokamak Department (NFI RRC KI), headed by V.F. Bogdanov, for their assistance in collecting erosion products from the tokamak vessel; V.M. Korzhavin of the RF Ministry of Atomic Energy for his support and general supervision; and H.-W. Bartels, C. Gordon and L.N. Topilski of the ITER International Team (Garching, Germany) for their attention and useful discussions of the obtained results.

References

[1] W. Jacob, Thin Solid Films 326 (1998) 1.
[2] J.N. Brooks, D. Cooper, G. Federici, et al., J. Nucl. Mater. 290–293 (2001) 260.
[3] V.Kh. Alimov, V.L. Goncharov, M.I. Guseva, et al., in: Problems of Atomic Science and Engineering, Ser. 'Thermonuclear Fusion', Kurchatov Institute, Moscow, 2000, No. 2, p. 45 (in Russian).
[4] K. Maruyama, W. Jacob, J. Roth, J. Nucl. Mater. 264 (1999) 56.
[5] J. Winter, G. Gebauer, J. Nucl. Mater. 266–269 (1999) 228.
[6] O.I. Buzhinskij et al., J. Nucl. Mater. 266–269 (1999) 793.
[7] G. Federici, R. Anderl, J.N. Brooks, et al., Fus. Eng. Des. 39&40 (1998) 445.
[8] W.J. Carmack et al., DIII-D Dust Particulate Characterisation (June 1998 Vent), Idaho National Engineering and Environmental Laboratory Report INEEL/EXT-1999-00095, 1999.
[9] W.J. Carmack et al., Analysis and Characterisation of TFTR Dust, Idaho National Engineering and Environmental Laboratory Report ITER/US/98/TE/SA-8, 1998.
[10] D. Petti, K. McCarthy, Fus. Technol. 37 (2000) 1.
[11] M.I. Guseva, V.M. Gureev, S.N. Korshunov, et al., J. Nucl. Mater. 220–222 (1995) 957.
[12] Yu.V. Martynenko, M.I. Guseva, V.I. Vasiliev, et al., J. Nucl. Mater. 258–263 (1998) 1120.
[13] B.N. Kolbasov, A.Yu. Biryukov, D.A. Davydov, et al., Fus. Eng. Des. 54 (2001) 451.
[14] M.I. Guseva, V.M. Gureev, A.G. Domantovskij, et al., in: Problems of Atomic Science and Engineering, Ser. 'Thermonuclear Fusion', Kurchatov Institute, Moscow, 2001, No. 1, p. 3 (in Russian).
[15] W. Jacob, W. Möller, Appl. Phys. Lett. 63 (1993) 1771.
[16] V.Kh. Alimov, A.E. Gorodetsky, A.P. Zakharov, J. Nucl. Mater. 186 (1991) 27.
[17] R.A. Causey, W.R. Wampler, D. Walsh, J. Nucl. Mater. 176&177 (1990) 987.
[18] B.N. Kolbasov, P.V. Romanov, V.A. Alimov, et al., Deuterium Mobilization from Co-deposited Carbon Films Produced in T-10 Tokamak, ITER Russian Federation Home Team Report RF 1 F 226 14/06/01 E, 2001.