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Overview of tracer techniques in studies of material erosion, re-deposition and fuel inventory in tokamaks

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Abstract

C-13 labeled methane and rhenium–boron coated plates were used at the JET tokamak as tracers for studies of the material transport, its erosion and re-deposition. Experimental procedures are described. The results are discussed in terms of processes underlying the material transport and the change of morphology of targets exposed to the plasma: physical sputtering, chemical erosion, prompt re-deposition. The influence of wall materials on fuel inventory is also addressed. C-14 measurements in the TEXTOR tokamak are presented and possibilities of using ^{14}C in carbon migration studies are considered.

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1. Introduction

Erosion of first wall materials by particle fluxes followed by the re-deposition of eroded and transported species leads to drastic modification of plasma facing components (PFC). The processes have a critical effect on material lifetime and fuel inventory [1]. Therefore, the assessment of such effects is of primary importance for the steady-state operation of a reactor-class device fuelled with a deuterium–tritium mixture. The experimental approach to studies of material transport involves spectroscopy methods, surface probes and tracer techniques. The term 'tracer' denotes an alien agent introduced deliberately to a studied system in minute quantities. The agent must be reasonably harmless to the system under examination and the source of the tracer must be localized. Tracers are applied in many fields of science and technology in order to accomplish a conclusive determination of a reaction rate and mechanism, distribution of components, flow direction, velocity of fluids, etc. An important point in using tracers in controlled fusion devices is the assessment of erosion of a long-range (i.e. m) and a short-range (local, i.e. cm) material transport in the plasma. Tracers are introduced to the plasma edge either by injecting exotic gas (e.g. C-13 labeled methane $^{13}CH_4$, silane SiH_4, *tri*-methylborine $B(CH_3)_3$), by ablating high-*Z* metals with laser beams, or by exposing wall probes and PFC tiles coated with well defined sandwich-type layers of high-*Z* (e.g. W, Re) and low-*Z* (e.g. C, B) elements. The distribution of the tracer on plasma facing surfaces at the conclusion of the tracer experiment gives valuable information on the material transport and re-deposition pattern.

Experimental programmes based on the application of tracers have been developed both at the TEXTOR

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and JET tokamaks. The studies of carbon migration in TEXTOR with a C-13 tracer gave vital information on the high re-erosion rate of carbon from co-deposits [2–4]. Moreover, analytical procedures for the quantification of C-13 in the C-12 environment have been developed [5]. Studies of high-Z erosion from test limiters proved a significant role of prompt local re-deposition of eroded species [4,6].

The intention of this paper is to summarize experimental methods at JET and to give an overview of results obtained in campaigns aiming at the determination of carbon transport (C-13 tracer) and the erosion of high-Z metals. Possibilities of using C-14 labeled tracer will also be addressed. The reason to assess the behaviour of carbon and high-Z metals is that both materials are foreseen for PFC of the ITER divertor [1]. Graphite-based materials are used in most of present day devices [7–12] and, therefore, physically and chemically eroded carbon (^{12}C) is the major plasma impurity species. The formation of volatile C_xH_y erosion products leads to the carbon re-deposition together with vast quantities of hydrogen isotopes. This results in unacceptably high tritium inventory as observed following the full D–T campaign in JET [13]. The majority of long-term tritium retention was found in the inner corner of the Mk-IIA divertor, especially in remote areas shadowed from the direct plasma line-of-sight. Thick, hydrogenated flaking carbon co-deposits were formed on shaded parts of tiles and on water-cooled louvers [10]. A similar scenario in ITER would obstruct its physics programme and the economy of operation if the safety limit for in-vessel inventory (350 g T) was reached within a small number of pulses [1]. The determination of the amount of carbon transported and the location of deposition zones in present day machines is then crucial for next-step devices. This was the major motivation for using material transport tracers in JET.

2. Experimental

2.1. C-13 injection

C-13 labeled methane (1.3×10^{23} molecules) and silane (3.2×10^{21} as 1.5% of SiH_4 mixed with D_2) were injected to the plasma edge of JET through gas inlet modules (GIM) located in the top of the vessel and in the outer divertor, respectively. Details of the gas injection system at JET have been shown previously [14]. Puffing was done during fifteen L-mode discharges, plasma current $I_p = 2.4$ MA and charge $Z_{eff} = 1.5$. The major aim was to determine the distribution of tracer re-deposited on PFC in the main chamber and divertor. Therefore, the experiment was carried out on the last operation session before the JET shut-down for restructuring of the divertor. Several tiles from the main chamber wall and the divertor (a full poloidal set) were then retrieved for ex situ examination. In Section 3.1 only results for C-13 will be presented because the amount of puffed silane was too small to be conclusively detected.

2.2. Marker tiles

Fig. 1 shows an example of a marker-coated tile installed in JET as a replacement of regular wall component. Marker films of a sandwich structure were prepared by Plansee AG (Austria) on carbon fibre composite tiles: 650 nm rhenium layer obtained by electron-beam evaporation and coated with a 2.5 μm carbon–boron overlayer (10 at.% B) deposited from separate C and B sources. The presence of boron admixture in the homogeneous C–B layer was to ensure the conclusive identifications of carbon erosion or re-deposition on the tile. The film composition and uniformity of its distribution were pre-characterised before

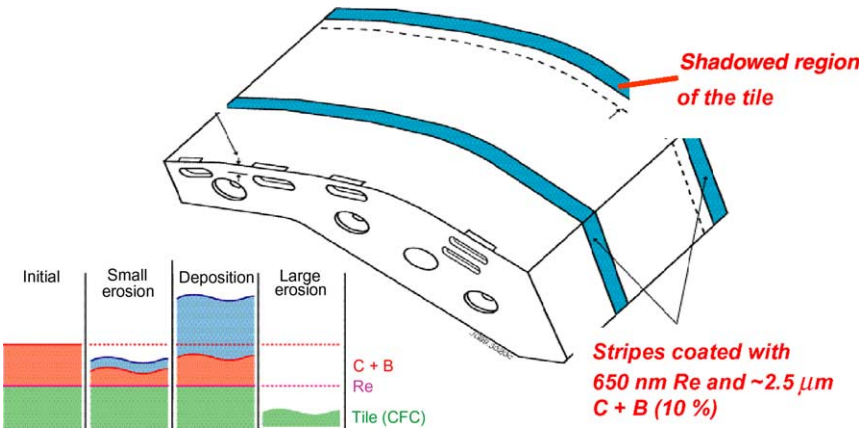

Fig. 1. Marker tiles used for studies of material migration in JET.

placing the tiles inside the torus. A number of such tiles were mounted in various locations (divertor, inner guard and outer poloidal limiters) and they were facing the plasma during the 1999–2001 operation campaign with the Mk-II Gas Box divertor.

2.3. Surface analysis

The exposed tiles removed from the torus were examined by means of ion beam analysis methods. The amount and distribution of C-13, deuterium, beryllium (regularly evaporated on the main chamber wall of JET), boron and rhenium tracers from the marker tiles were studied. The C-13 content was measured with nuclear reaction analysis (NRA) $^{13}C(^3He,p)^{15}N$ a 2.5 MeV $^3He^+$ beam and with enhanced proton scattering (EPS) $^{13}C(p,p)^{13}C$ at resonance and off-resonance energies. $^3He^+$ was also used for tracing deuterium $d(^3He,p)^4He$ and beryllium $^9Be(^3He,p)^{11}B$, whereas a H^+ beam at 0.7 MeV was used to determine the boron content $^{11}B(p,\alpha)^8Be$. Rhenium was measured with Rutherford backsattering spectroscopy (RBS) and particle induced X-ray emission (PIXE) using protons at 2.5 MeV. Depth profiling of surface layers (down to 80 μm) was done by means of secondary ion mass spectrometry (SIMS) and, for the quantification, the SIMS results were calibrated according to results of the time-of-flight elastic recoil detection analysis (TOF-ERDA). The amount of carbon-14 in co-deposits on PFC in TEXTOR was determined with accelerator mass spectrometry (AMS).

3. Results and discussion

3.1. Re-deposition pattern in the divertor: C-13, deuterium and beryllium

Fig. 2(a) shows the poloidal cross-section of the Mk-II GB divertor, whereas in Fig. 2(b) the distribution of C-13 on tile 1 is plotted. Tiles 1 and 3 were the only areas where significant concentration of C-13 was found in the very surface layer of co-deposits. Minute quantities (though above the natural background level of ^{13}C in carbon) were also detected on tile 4 and, only by SIMS, on components of the main chamber wall. No tracer was found both on the outer divertor tiles and on the septum plates of the gas box. Assuming the toroidal symmetry of carbon transport and re-deposition, the integrated amount of C-13 in the inner divertor was assessed at the level of 5.8×10^{22} atoms, corresponding to 45% of the total tracer input to the torus. This result is reported with a high degree of confidence, because the same value has been obtained in two independent measurements: SIMS calibrated versus TOF-ERDA [14] and $^{13}C(p,p)^{13}C$ enhanced proton scattering with a 2.5 MeV H^+ beam. The tracer experiment has decidedly proven

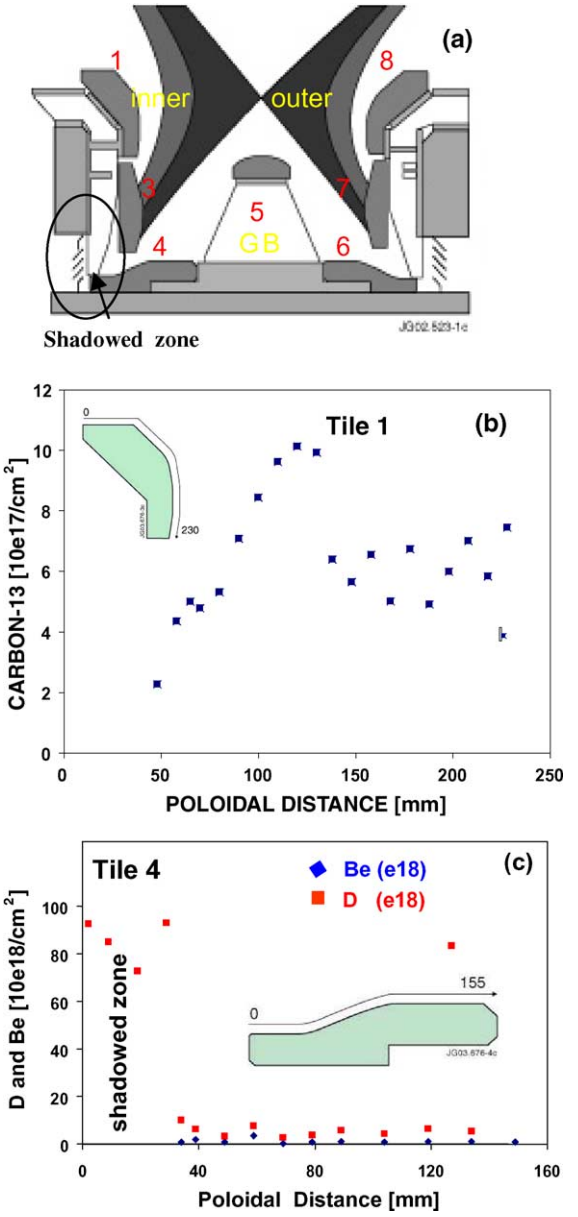

Fig. 2. Cross-section of the Mk-IIGB divertor of JET with marked region of the shadowed zone (a) and distribution of species on tiles of the inner divertor: distribution of C-13 on Tile 1 (b) and distribution of deuterium and beryllium on Tile 4 (c). Inserts in frames (b) and (c) show the tile geometry.

the preferential flow of the background plasma and impurity species towards the inner divertor leg. Moreover, it has also been noted that no C-13 was found in the shadowed region on tile 4, where thick carbon (^{12}C) have been formed. From the distribution of deuterium and beryllium shown in Fig. 2(c) one notices distinct differences in the distribution of Be and D along the tile.

Significant D content in flaking carbon deposits in the shadowed zone is not accompanied by the presence of beryllium. The result indicates that only chemically eroded carbon migrates to the shadowed zone. However, lack of ^{13}C in the shadow suggests that the carbon migration is a multi-step process, i.e. the carbon arrives into that region not directly but after several steps of re-erosion and re-deposition on the divertor target. The issue may conclusively be solved by injecting the tracer twice: first in the middle and later at the end of an experimental campaign, followed by SIMS depth profiling of co-deposits on tiles.

3.2. Marker tiles

When post-exposure and initial content of boron and rhenium on marker tiles are compared, one finds that the boron was reduced by at least 98% (in some areas B is completely removed) thus indicating also the erosion of carbon from the C–B film. In case of rhenium, the erosion of 97% is measured in the central part of the limiter while less erosion is noted on the sides. This is shown in Fig. 3. Some differences have been observed between the erosion effects on various tiles, but in all cases the central part was most strongly eroded [15]. The erosion from limiter is not unexpected, but from the tracer experiment one concludes a significant contribution of carbon and beryllium impurity fluxes in the erosion mechanism of high-Z. While carbon and boron are easily eroded by deuterium ions and charge exchange neutrals (threshold energy for physical sputtering, E_{th}, below 20 eV), the erosion of rhenium is mainly caused by heavier impurities. As calculated according to the formula given in [16], the threshold for sputtering by deuterium is around 220 eV, whereas it is about 50 and 60 eV for carbon and beryllium, respectively. Basically, the electron temperatures, T_e, at limiters in many medium-density discharges in JET are well below 10 eV, however much higher values may be expected in low-density (low collisionality) plasmas. Moreover, ions are not monoenergetic and their distribution can have a high-energy tail. Taking into account these energy considerations one concludes that the erosion of rhenium by deuterium is negligible in comparison to erosion by multiply charged impurity ions accelerated through the electric sheath potential, $V \approx 3qkT_e$, where q denotes the charge state. Under these conditions effective contribution of D^+ to rhenium erosion would require edge temperatures above 80 eV, but at the same time the erosion by minority species (at least 3% of C^{4+} and Be^{3+} in the edge flux) would still prevail due to sputtering yields being significantly greater than that for deuterium, $Y^D(Re) \approx 3 \times 10^{-3}$.

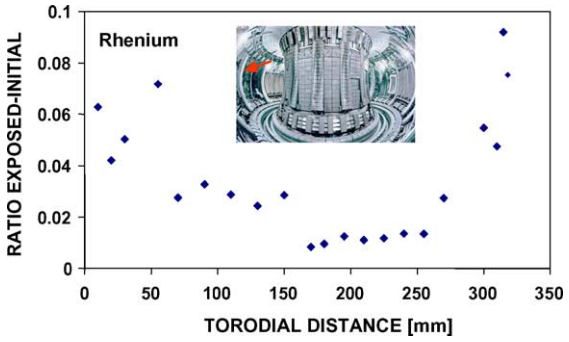

Fig. 3. Erosion of rhenium from the marker tile. Results are presented as a ratio of post-exposure-to-initial value. The insert shows the JET vacuum vessel; location of the marker tile on the wall is indicated with an arrow.

3.3. Carbon-14 as a material transport tracer?

In a search for new and ultra-high sensitivity tracer techniques in studies of carbon migration in fusion experiments we have considered the use of ^{14}C-labeled compounds. C-14 analysis is a well established technique applied for carbon dating in matter. The greatest sensitivity is reached by means of AMS [17] which is capable of tracing 10^{-17} g (5×10^5 atoms) of the isotope. Fossil fuels (crude oil and natural gas) used as substrates in production of graphite do not contain ^{14}C (β^- emitter 0.156 MeV, $t_{1/2} = 5730$ y, activity of 1 g ^{14}C corresponds to 1.65×10^{11} Bq = 4.5 Ci). Therefore, the presence of the isotope in a graphite-wall tokamak would be mainly a result of carbon activation by neutrons from the n-yielding branch of D–D fusion. The first step was the assessment of the isotope content in TEXTOR operated with a graphite wall. Two samples were studied: thick and flaking co-deposits from the toroidal limiter tiles (after 4 h of plasma operation) [18] and a thin deposit removed from a graphite collector probe exposed for 200 s to particle fluxes in the scrape-off layer plasma. Analysis by means of AMS was preceded by checking the overall activity level of a sample in order to avoid possible contamination of the accelerator ion source, in case the activity would have been above 25 Bq/g. Fig. 4 shows the β^- spectrum recorded with a liquid scintillator (LSC) for a sample of the flaking co- deposit. The measurements confirm the presence of a certain amount of tritium (product of the second branch of D + D fusion) [19,20]. The content of ^{14}C is however definitely below the LSC sensitivity level of 60 Bq/g. The amount of C-14 then determined by AMS was 18 ± 7 mBq/g. The second sample contained significantly less tritium and around 12 mBq/g of carbon. This is significantly less than the ^{14}C activity in presently living material, approximately 250 mBq/g carbon, which results from the natural, cosmic ray induced production of ^{14}C in the atmosphere. To some extent, the study addresses the point of carbon activation under fusion conditions.

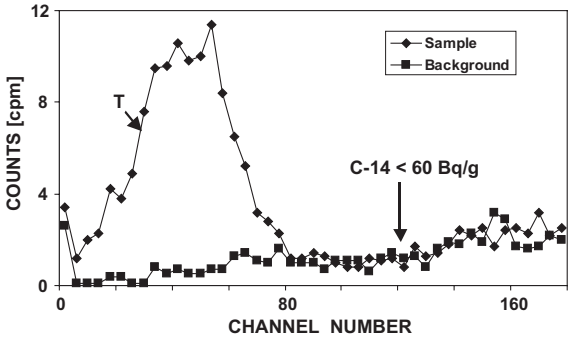

Fig. 4. Spectrum of β^- radiation recorded with a liquid scintillator for a flaking co-deposit formed at TEXTOR during 4 h of plasma operation.

The amount of ^{14}C determined in TEXTOR should be treated as a background level if a tracer experiment would be carried out. Two scenarios may be considered: injection of gas or coating a limiter tile with a ^{14}C-labeled organic compound stable to the temperature of about 300°, i.e. the base temperature of PFC in TEXTOR. Taking into account the sensitivity of AMS and existing isotope background, the amount required would be in the range of 1–2 μCi. This is calculated under the assumption that the migration of C-14 from the introduced agent would result in the uniform coverage of the device wall.

4. Concluding remarks

We have applied various tracer elements for detailed studies of material erosion and migration under the plasma operation. Tracer techniques have then significantly contributed to the overall picture of erosion and re-deposition pattern in JET. Experiments with C-13 marked methane have proven long-range migration of carbon to the divertor, whereas the application of high-Z metal tracers allowed identification of major species involved in the erosion process. The results allow also conclusions regarding the implication for ITER. Analyses of tiles following the operation with the Gas Box divertor show a similar deposition pattern as observed with the previously used Mk-IIA divertor of open geometry [10,21]. The majority of fuel is stored in shadowed zones of the inner divertor and the results also indicate that the migration to the shadowed zone is a multi-step process. Moreover, it has been confirmed that no physically sputtered beryllium reaches that zone in the divertor corner (too low T_e for Be sputtering in the divertor) and it means that the vast fuel inventory is associated with chemically eroded carbon. The use of carbon in next-step devices should then be thoroughly re-assessed. However, as long as carbon-based materials are considered as candidates for the PFC in next-step machines the quantitative aspects of carbon will remain an important issue and new diagnostic tools and sensitive material transport tracers should be used. Preliminary studies of carbon 14 background in TEXTOR show that the use of ^{14}C-labeled tracers can be considered.

Acknowledgements

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