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Impurity release from low-Z materials under light particle bombardment

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Abstract

Recent progress in the understanding of the erosion of low-Z materials under bombardment conditions characteristic of magnetic fusion experiments is reviewed. The role of physical sputtering, chemical sputtering and radiation-enhanced sublimation in tokamaks is considered, and observations are related to laboratory measurements. The role of physical sputtering is largely understood, and tokamak measurements, under conditions where physical sputtering is expected to dominate, can be well predicted, except in the energy range near the sputtering threshold. Chemical erosion and radiation-enhanced sublimation are less-well understood, and predictions of erosion yields under tokamak conditions require assumptions (primarily related to energy and flux density dependence) which do not have a solid experimental basis. Also, only a few quantitative results from tokamaks are available to confirm predictions, and those which are available are not always consistent.

Keywords: Impurity source; Physical erosion; Chemical erosion; Low Z wall material; Radiation-enhanced sublimation

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1. Introduction

The successful operation of next-generation fusion reactors will rely on the ability of plasma-facing components to handle enormous particle and power loading. Contamination of the plasma and component lifetimes will depend on the mechanism and rate of material erosion, as well as the transport and redeposition of eroded particles. Materials at different locations of the reactor first wall will be subjected to significantly different conditions. The lining of the main plasma chamber will be subjected primarily to a flux of charge-exchange neutral hydrogen atoms with energies varying from a few eV to keV's. The divertor target plates and sidewalls will experience much higher fluxes of hydrogen with energies of order 10 eV. In the divertor region, power fluxes will be much larger than on the main wall, and thus power handling capabilities are of primary importance [1]. It has been found that impurities produced at the main wall have a greater chance of penetrating the main plasma than impurities from the divertor region [2], and thus erosion characteristics take on added importance for main wall surfaces. Off-normal conditions (e.g., disruptions) may well cause more serious problem as far as component lifetimes are concerned. Large transient heat loads may be received at almost any location in the torus, possibly leading to the melting of metal components (e.g., Be and W).

Components made from low-Z materials, such as beryllium and carbon, have the advantage that higher concentrations of eroded particles may be tolerated in the main plasma; however, they have the disadvantage of being easier to erode. Carbon-based materials also have excellent thermomechanical properties and will not melt during off-normal conditions (e.g., disruptions), but they are subject to chemical attack and radiation-enhanced sublimation. Beryllium, while not susceptible to chemical attack, has a relatively low melting point, which may limit its usefulness. Other low-Z elements, Li, B and Si, have been shown to have beneficial properties when used in combination with carbon, as dopants or coatings. In light of these considerations, Be and C are considered primary candidate materials for plasma-facing surfaces in ITER.

The focus of this review is on the release of impurities from low-Z materials under plasma conditions expected in

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large tokamaks. Physical sputtering, chemical processes and radiation-enhanced sublimation are all expected to play some role in the reactor environment. Specific questions we wish to address are: (i) What happens to the erosion of beryllium, boron and carbon for bombarding energies of 10–20 eV? In the case of carbon, chemical erosion will dominate in this energy range and many factors can influence reaction rates. (ii) Do erosion processes saturate at high flux densities? A strong reduction in both the chemical erosion and RES of carbon with increasing hydrogen flux density has been observed, however, contrary results also exist. (iii) Can dopants help the erosion behaviour of carbon? At present, both the need for, and the role of, dopants is unclear. Some dopants, such as boron have the ability to suppress H and O chemistry, but generally at a cost of reducing thermal conductivity. (iv) Can we relate current laboratory measurements to tokamak observations? The recent emphasis placed on studying plasma-surface interaction, PSI, issues in large tokamaks has produced a wealth of observations which allow comparisons to be made with laboratory studies, and eventually will aid in making projections to larger reactors.

1.1. Erosion mechanisms

The mechanisms for light-ion induced erosion of low-Z materials have been well established in laboratory experiments over the last 10–20 years. Three primary types of erosion have been identified: (i) physical sputtering, (ii) chemical sputtering/erosion and (iii) radiation-enhanced sublimation; the last one applies to carbon-based materials. Each of these will be described briefly in the present section, and will be dealt with individually in later chapters. Extensive reviews of these erosion mechanisms have recently been the topic of two book chapters [3,4].

Physical sputtering involves the direct transfer of momentum from an incident particle to particles in the target substrate. Any incident species is capable of removing atoms from any target, provided there is sufficient energy available, and the fraction of energy which may be transferred in a collision is sufficient to overcome the surface binding energy of the target material. In a fusion reactor, there will be physical sputtering due to hydrogenic species bombardment, but also self-sputtering due to eroded ions returning to the walls. Reactors with multi-component first wall materials will also experience multi-species impurity bombardment.

Chemical erosion, as the name implies involves chemical reactions between the incident particles and substrate atoms, producing volatile molecules. When the momentum of the incident particles also plays a role in the reaction mechanism, the term chemical sputtering is also used. Primary examples of chemical sputtering relevant to fusion are the production of hydrocarbons during hydrogenic bombardment of carbon, CO/CO_2 production during oxygen (a major plasma impurity, the source of which may not be the primary first wall material) impact on carbon, and the production of tungsten oxide during oxygen bombardment of tungsten.

Radiation-enhanced sublimation (RES) is an erosion mechanism peculiar to carbon-based materials. It is similar to physical sputtering in that it does not involve chemical reactions, but requires incident particles to have sufficient energy to dislodge carbon atoms from their lattice sites. Once in interstitial spaces, the carbon atoms may diffuse to the surface where they are weakly bound and may sublimate with an activation energy significantly lower than that required for thermal sublimation. RES is characterized by an exponential increase in erosion yield for temperatures > 1200 K, with an activation energy much less than that for thermal sublimation [4].

1.2. Measurement techniques

Erosion measurements in this review are composed of three primary types. These are laboratory measurements utilizing accelerators as the source of bombarding particles, laboratory measurements with plasma sources and measurements in tokamak devices. Each of these methods has its advantages and disadvantages and in general a combination of all three types of measurements is the best way to get at the whole picture.

Accelerator measurements have the advantage of providing great control over the bombarding species in terms of energy and flux, and also relative ease in measuring erosion yields. They cannot in general, however, reproduce the conditions expected in a reactor environment, such as the high flux density and the multi-species bombardment. Laboratory plasma sources are used to extend the range of accelerator measurements, primarily at high flux densities and low energies. Their disadvantage is that some of the control is lost over the exact nature of the bombarding particles. With well characterized plasmas, however, this is becoming less of an issue.

Tokamaks are, of course, the ideal place to measure the erosion expected in real fusion devices. Unfortunately, it is often a very difficult task. Indirect means must be employed for measuring both incident and eroded particles. Incident flux densities may be derived from recycled H_α emissions, incident particle energies from wall probes and more recently from spectroscopic measurements. The flux of sputtered particles is evaluated from electromagnetic emissions from excited, ionized or dissociated atoms or molecules. The last few years have seen a dramatic increase in tokamak erosion measurements, allowing a broad range of comparisons to be made with laboratory results.

2. Physical sputtering

2.1. Laboratory measurements and calculations

Physical sputtering values are relatively well known, and details of the mechanism have been well established

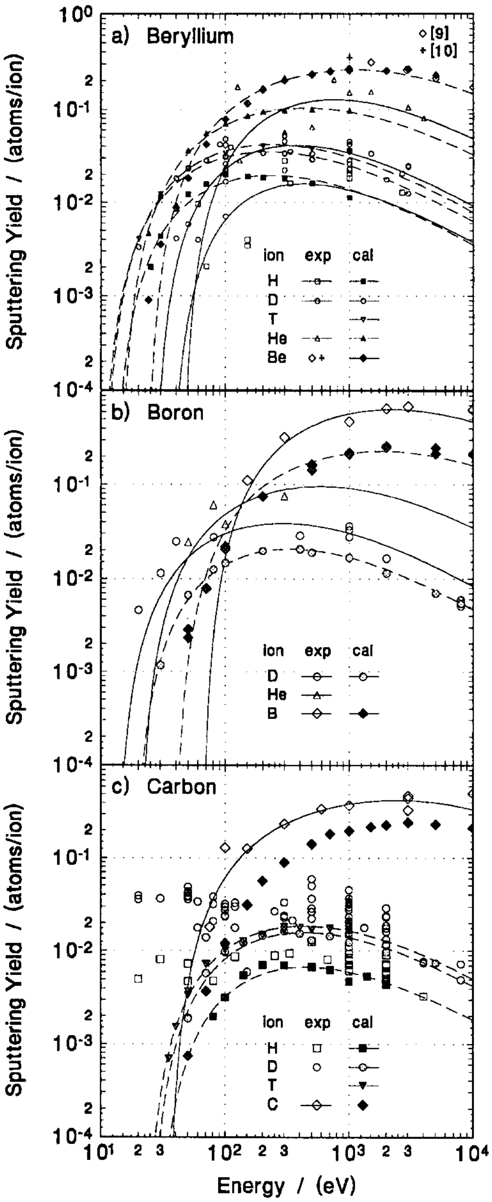

Fig. 1. Physical sputtering yields of Be, B and C for various incident ions. Data from Ref. [8]; recent data on Be self-sputtering from Refs. [9,10].

for many years [4]. Consistent results have been reported from many different laboratories for a great range of ion species and targets. A comprehensive compilation of the results is provided in Refs. [5-7], and only a brief survey of relevant results is presented here. In Fig. 1(a), (b) and (c) we present light ion physical sputtering results for Be, B and C, showing both measured and calculated values [8-10]. Because of the dynamical nature of the processes involved, calculations of physical sputtering have been highly successful [11], and have recently been extended to the low energies relevant to fusion [4].

Several features of the physical sputtering data are observed, which have a large impact on impurity production in tokamaks. For the lightest ions, the threshold for sputtering is near the bombarding energies associated with low temperature divertor plasmas. Physical sputtering could be greatly reduced by operating in this plasma temperature range; observations under detached plasma conditions show a large reduction in carbon influx due to physical sputtering [12]. On a more critical note, there is the possibility of unity self-sputtering yields at moderate energies and angles of incidence [13], a process which could lead to catastrophic impurity buildup. The question of a flux dependence for sputtering has been raised, and there is evidence for a slight decrease in yield with increasing flux density [14], and this is something to look for in tokamak results.

It is noted that our absolute knowledge of physical sputtering yields is limited to about a factor of two. Deviations between calculated and experimental results, or between different experiments is thought to originate from effects such as surface roughness, the existence of oxide layers, and the accumulation of projectile atoms in the target [8]. Furthermore, near the threshold for sputtering, errors could be as large as several orders of magnitude. The lines drawn on Fig. 1 are based on the revised Bohdansky formula [8], which uses threshold energy as a fitting parameter. It is clear from the fact that significantly different values of threshold energy have been used to fit the same species combinations, that large errors could be involved in this energy range. Such errors could make modelling of divertor plate erosion under low temperature plasma conditions subject to equally large uncertainty.

The main questions about physical sputtering at this time are related to the actual observation of physical sputtering in tokamaks and attempts to relate the observed yields to laboratory measurements.

2.2. *Physical sputtering measurements in tokamaks*

The measurement of sputtering yields in an experiment as complex as a tokamak is a difficult task, and only recently have the required diagnostics been available. Difficulty is encountered when more than one material is used for plasma-facing components in the machine, and the exact elemental nature of various surfaces is not known. Measurements near net erosion surfaces, such as divertor plates, often can avoid this problem. Even in a single material machine (which is likely to be all carbon), complications can arise when different erosion mechanisms may be in operation.

In order to make sputtering measurements, it is necessary to have information on the impacting particle flux densities, energies and species, and a measure of the released particle fluxes. Edge diagnostics, such as Langmuir probes (most often available in the divertor region), provide information on the impacting particle flux densities and energies. H_α emissions and heat deposition measurements are often also available to confirm the probe data. However, the Langmuir probe data are subject to concerns over the correct interpretation, especially due to non-Maxwellian energy distributions [15]. Fluxes in regions outside the divertor are often much less well defined, due to the absence of diagnostics, a large variation of fluxes around the torus and the fact that most of the incident flux is composed of neutral atoms.

The erosion and redeposition of materials in tokamaks has been measured through the use of specimens inserted into the reactor and exposed to one or more discharges; the specimens are then extracted for ex-situ analysis. Examples are the DIMES facility on DIII-D [16–18], and similar experiments performed on JET [19], TEXTOR [20] and ASDEX-U [21]. Understanding of the processes leading to the observed erosion/redeposition patterns is gained from models (e.g., Refs. [18–23]) based on measured plasma parameters and available erosion yields from the literature. The transport of eroded atoms is a critical issue in such analyses.

The most common way of measuring the impurity influx, as opposed to material erosion, is through absolutely calibrated spectroscopy in the edge plasma, as described in the review by Behringer et al. [24]. For the technique to be successful, it is necessary to know the local electron temperature in order to evaluate the photon efficiencies. Again, such information is open to interpretation and results are not so widely available outside of the divertor.

The use of interpretive computer codes, such as DIVIMP [25], IMPMC [26] and BBQ [27], allows one to check the consistency of various erosion mechanisms with experimentally observed quantities. These codes allow the incorporation of D^+ physical sputtering, self-consistent self-sputtering, angle of incidence-enhanced effects, as well as other impurity production mechanisms (if required). Comparisons of calculated and measured impurity concentrations near the divertor strike points generally give good agreement, both in spatial distribution and absolute magnitude. A sputtering yield of ~ $0.04C/D^+$ in the ASDEX-U divertor [28] is entirely consistent with D^+ physical sputtering with appropriate corrections for angle of incidence and C^+ self-sputtering. In Fig. 2, we show the calculated CII emission distribution compared with the observed one in JT-60U [26,29]. The calculated levels are in good agreement at the strike points, but disagree in the private plasma region. Similar results were obtained with the DIVIMP code for JET [30] and ASDEX-U [28,31] plasmas. All of these results indicate that either erosion mechanisms other than physical sputtering are involved (see Section 3.5), or that the model is inadequate at describing all aspects of impurity transport. To eliminate chemical effects, discharges have been run with He as the working gas [12]. Here, observations both at the strike points and away give good agreement with modelling based on physical sputtering alone; other impurity sources are not required to explain the experimental measurements (see Section 3.5). Similarly, boron influx measurements in ASDEX-U [32], made after boronization, also match calculated profiles somewhat better than the D → C cases (see Fig. 3). There remains, however, a BII signal away from the strike point which is not matched by DIVIMP modelling. This is possibly due to a diffuse wall source of boron hydrate molecules [32], or again higher order phenomena in the transport which are not included in the code. In an older study of Be sputtering in JET limiter discharges, LIM computer code calculations produced reasonable agreement with observed BeI emissions based on physical sputtering only [33].

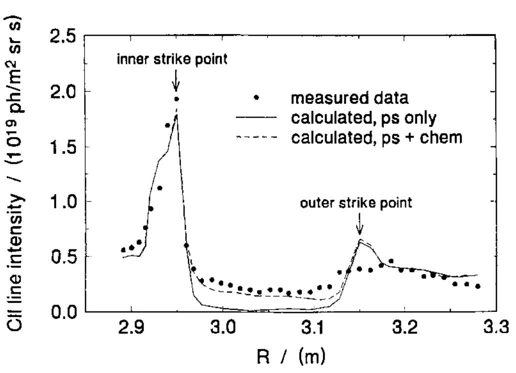

Fig. 2. Experimental and calculated values for CII emissions at the JT-60U divertor [26,29]; 'ps' represents physical sputtering and 'chem' chemical erosion. The chemical erosion yield was 0.05, with 90% redeposition near the strike points.

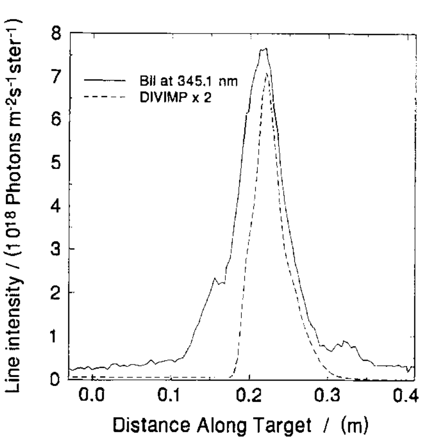

Fig. 3. Experimental and calculated values for BII emissions at the ASDEX-U divertor after boronization [32].

Other information may also be found from tokamak measurements. In TEXTOR [34,35], the penetration of neutral carbon into the plasma from a probe limiter surface has been measured, allowing an evaluation of impurity particle energies. Penetration distributions which clearly

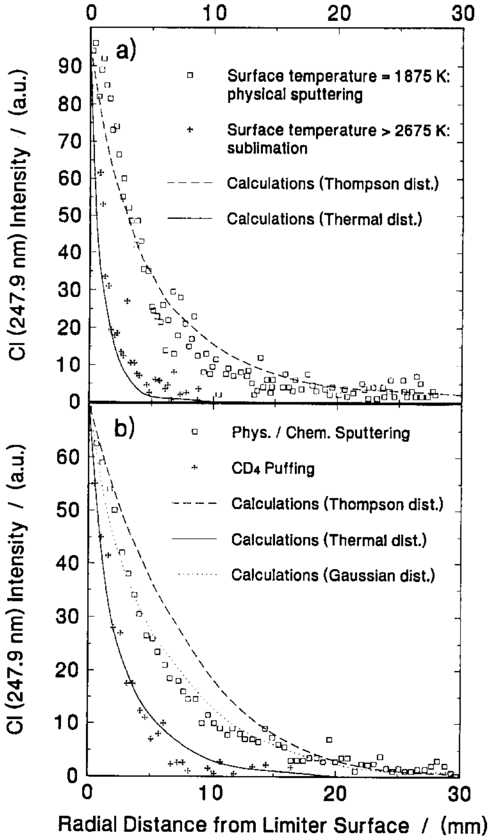

Fig. 4. Radial intensity distribution of CI emissions across the TEXTOR test limiter for different plasma conditions and release mechanisms [34]: (a) low density discharge (high edge temperature) calculations based on the Thompson distribution for physically sputtered particles and a thermal Maxwellian velocity distribution for evaporation at 2675 K; (b) high density discharge (low edge temperature) calculations based on a Gaussian velocity distribution corresponding to a dissociation energy of 1 eV (chemical sputtering).

resemble the Thompson distribution characteristic of physical sputtering are found for conditions of low density (high edge temperature and therefore high impact energy), Fig. 4(a), while at higher density, the observed less energetic impurities may have some chemical sputtering origin, Fig. 4(b).

At present, it appears that our understanding of physical sputtering is sufficient to make reasonable predictions of erosion in tokamaks under circumstances where it is the dominant erosion mechanism.

3. Chemical erosion

Chemical erosion, unlike physical sputtering, is largely an unknown as far as impurity levels in tokamaks are concerned. There are four main reasons for this difficulty. First, wall component surface temperatures are difficult to predict and may vary significantly during the course of a discharge. Since the chemical reactions may be strongly temperature dependent, yields could vary by an order of magnitude. Second, there will be a flux dependence to the erosion yield (assuming any finite reaction time, it is a physical necessity that the reaction process saturates at a sufficiently high flux density), however, it is not yet clear where this saturation will be reached. It should occur at lower fluxes for lower ion energies. Third, the energy dependence of erosion yields for energies less than 50 eV is not yet well established, especially for hydrocarbon products other than methane. And lastly, in a machine with multi-element first wall materials, as ITER is presently envisioned, metal deposition on carbon surfaces may greatly alter the erosion process, or may result in synergisms due to multi-species bombardment. A further complication with regard to the importance of chemical erosion arises from the possibility of prompt redeposition of molecular products or fragments. At lower ion impact energies, there will be a greater production of C_2H_x and C_3H_y hydrocarbons relative to CH_4 [36,37], and these heavier molecules may have a greater redeposition probability, e.g., they have a lower ionization potential and a larger Larmor radius.

3.1. Low energy hydrogenic erosion of carbon

With the high density or gaseous divertor concept, very low edge temperatures, on the order of a few eV, have been predicted for the next generation of tokamaks, like ITER. Evidence for the possibility of achieving such low plasma temperatures is available from current tokamaks; e.g., plasma temperatures as low as $T_e = 3$ eV have been reported in the divertor of ALCATOR C-mod [38] and spectroscopic measurements of CIII Doppler broadening in ASDEX-U have produced T_i values in the 2–10 eV range [39]. As a consequence, hydrogenic bombarding energies ($\sim 2kT_i + 3kT_e$ [40]) in the 10–20 eV range are possible.

New erosion results in this energy range [41,42] appear to show a transition in erosion mechanisms for incident ion energies < 50 eV; see Fig. 5 for methane yields. As the ion energy decreases below 200 eV, there is a broadening of the temperature profiles, a decrease in the peak erosion yield, and a shift in the temperature of the peak erosion yield, T_m, to lower temperatures. Another important observation is that below 50 eV H^+ energy, both the methane and total chemical erosion yields decrease with decreasing ion energy; see Fig. 6. Also seen in Fig. 6, is the increasing contribution of heavier hydrocarbons to the total chemical erosion yield with decreasing energy.

The level of the erosion yield found in these experiments is similar to the latest results reported by Garcia-Rosales and Roth [43], which is about a factor of 2 lower than previous measurements by Roth and Bohdansky [44].

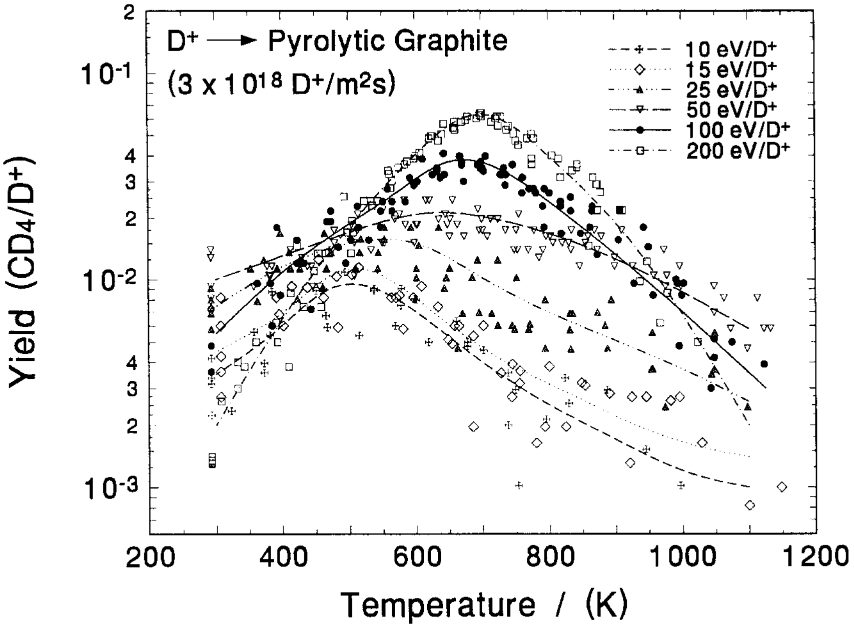

Fig. 5. Temperature dependence of methane yield for low energy D^+ impact on pyrolytic graphite [41].

Similarly, these measurements are up to an order of magnitude lower than the model projections of Garcia-Rosales and Roth [45], and show a decreasing trend with decreasing ion energy, rather than remaining constant as the model predicts.

3.2. Flux dependence of chemical erosion

The hydrogenic flux to the main plasma chamber walls of a machine such as ITER will be in the 10^{20} to 10^{22} H/m^2 s range. Such fluxes are within the range of what is achievable by conventional accelerator sources at the low end, and plasma simulators at the high end. The erosion yields on these surfaces should not be subject to significant uncertainty on the basis of a flux-dependent erosion yield; hydrocarbon production will not have saturated. High flux surfaces, such as limiters and divertor plates, however, may receive fluxes as much as two orders of magnitude higher. Saturation of the erosion process is a distinct possibility on these surfaces.

Various carbon erosion yields, obtained from many sources, including tokamak measurements are plotted in Fig. 7 [36,37,41,43,44,46-57]. There is a lot of scatter in the data due to the different types of measurements and different temperatures and energies. Over a flux range of five orders of magnitude, there is no clear trend indicating a decrease in erosion yield with flux. Two recent tokamak studies, however, do give rise to some optimism. In JT-60U [58], mass spectrometer measurements of CD_4 and C_2H_4 pressures in the divertor region were found to rise as $P_{CD4,C2D4} \propto P_{D2}^{0.7}$, or $Y \propto \phi_{D2}^{-0.3}$, indicating a strong drop in the erosion yield with increasing flux density. Caution is of course advised in the interpretation of such indirect measurements of erosion, as surfaces not directly in contact with the plasma may be affecting the results [12]. More direct spectroscopic measurements in TEXTOR [59] have produced an even steeper drop in erosion yield, $Y \propto \phi^{-0.6}$, for CD_4 production at the graphite test limiter surface, Fig. 8. In these measurements, the incident flux was varied by moving the probe into the plasma, and thus conditions are not quite the same in all measurements (e.g., probe temperature and ion impact energy), making the interpretation of the CD signals somewhat uncertain. Nevertheless, the clear implication is that chemical erosion is saturating.

3.3. Chemical erosion of doped graphites

It has long been known (e.g., Ref. [60]) that non-reactive elements added to carbon (doping) could reduce the level of chemical erosion. Many such studies have since been carried out [3], with varying degrees of success, depending on the dopant(s) and other properties of the graphite. Very little information, however, is available for ion energies < 100 eV. At higher energies (> 100 eV) [61], boron was found to be more effective at reducing methane production than Ti, Si or W. The reduction, however, is greatest for temperatures $> T_m$, with less reduction for $T < T_m$. For 50 eV D^+ impact on USB15 (B-doped graphite manufactured by NII Graphite, Moscow), no reduction in erosion yield as compared to pyrolytic graphite was observed below 600 K [43]. Also, the erosion of a Si-doped CFC due to ~ 30 eV H^+ at high

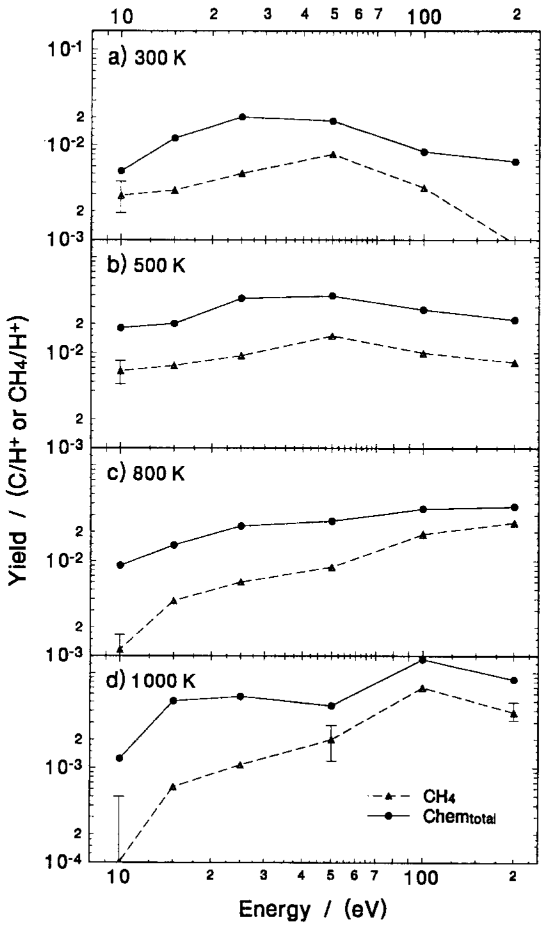

Fig. 6. Incident energy dependence of the methane yield and the total chemical erosion (i.e., $\mathrm{Chem}_{\mathrm{total}} = CH_4 + 2(C_2H_2 + C_2H_4 + C_2H_6) + 3(C_3H_6 + C_3H_8)$) of graphite due to H^+ impact [42]. The legend in (d) also applies to (a), (b) and (c).

flux density (1.2×10^{22} H^+/m^2 s) was similar to that of the undoped CFC for temperatures < 600 K [56]. However, other experiments on Si-doped graphite, for 10 eV D^+ bombardment at room temperature, may indicate that some suppression of chemical reactions is occurring [62].

It has been proposed [3] that two reaction channels contribute to the formation of methane during D^+ bombardment of graphite. For ion energies greater than ~ 100 eV, the high temperature reaction channel, which has a maximum near 800 K and leads predominantly to the formation of CH_3 radicals, dominates. The low-temperature reaction channel, which becomes more important at lower ion energies, has a maximum at temperatures possibly as low as 200 K [63] and leads primarily to CH_4 production. The addition of B to carbon materials acts to suppress the high temperature erosion branch through the creation of a more carbidic structure [64]. Such changes, however, do not affect the low temperature branch [3]. These combined effects lead to the characteristic shift to lower T_m observed for boron-doped graphites, but do not lead to significant yield reductions at low temperatures.

In light of this, it seems that the primary advantage of using B-doped graphite in a reactor would be for its oxygen gettering abilities and the subsequent reduction in chemical erosion associated with reduced oxygen levels. Another advantage would be the reduced reactivity to oxygen during an accidental vacuum leak [61]. On the other hand, there is the reduction in thermal conductivity [65] and increase in D retention [64,66] to consider.

3.4. Oxygen erosion

Oxygen is one of the primary intrinsic impurities in current fusion reactors, and can lead to high levels of erosion unless measures are taken to reduce its concentration in the plasma. The addition of oxygen gettering elements (Be, B, Li) to the reactor walls is a common practice in many tokamaks. In carbon machines without such precautions, C and O influxes are seen to be strongly coupled [35,67,68], supporting oxygen's dominant role in wall erosion.

Due to multiple charged states, oxygen ions are likely to possess an energy of several tens of eV, and possibly hundreds eV, when bombarding wall structures. Under these circumstances, erosion yields close to unity are expected [3]. Ion beam measurements of the erosion yield are well established [3,69]; CO is the primary volatile product, with lesser amounts of CO_2. In O^+ erosion of B-doped graphites, BO, BO_2, B_2O_2 and B_2O_3 were all observed as reaction products, in addition to CO and CO_2 [70,71], while for Si-doped graphite only SiO, CO and CO_2 were observed [72].

The observation of CO emission bands has been reported in TEXTOR [73], however, they were not recognized as such until later experiments on PISCES-A [74,75]. The situation is complicated by the overlapping of CO bands with hydrogen molecular bands, making it difficult to obtain absolute data. It has been possible to derive CO fluxes from such measurements [35], confirming that a major portion of the oxygen was recycling as CO. Hopefully, in the future we will see O^+-induced erosion measurements in tokamaks similar to the hydrocarbon measurements which are now starting to appear.

In a reactor, there will naturally be a much larger flux of hydrogenic species to the walls, in addition to any oxygen flux. Such multi-species bombardment leads to the possibility of synergistic reactions resulting, in this case, in a reduction of the erosion yield. The production of D_2O has been observed in the laboratory under conditions of D^+/O^+ simultaneous bombardment of carbon [3,76]. Depending on the carbon temperature and the bombardment conditions, O + H reactions lead to ~ 15% of the oxygen being recycled as H_2O, with the remainder recycling as CO or CO_2 [76]; see Fig. 9. No O_2 is re-emitted. The

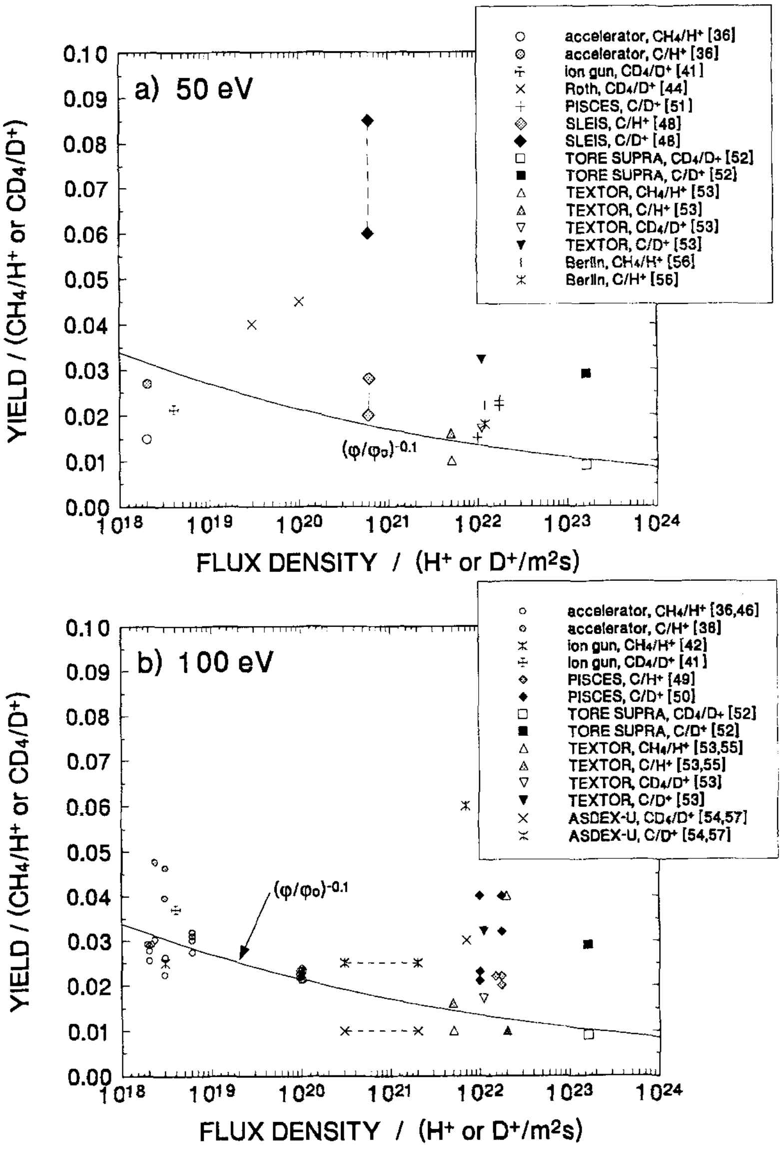

Fig. 7. Flux dependence data of the chemical erosion of graphite.

production of H_2O is strongly temperature dependent, with a maximum about 800 K, near the maximum for hydrocarbon production. So far, as for CO and CO_2, there have been few measurements of OH or OD band emissions in fusion reactors.

3.5. Chemical erosion in tokamaks

The formation of molecular impurities in tokamak discharges has a history as long as the devices themselves; for an excellent review of this topic see Ref. [12]. The actual measurement of erosion yields is a far more recent development. Several types of tokamak measurements have already been mentioned in the flux dependence section and presented in Fig. 7. The early results from TEXTOR were derived from 'sniffer probe' measurements [53], where the tokamak plasma was used as a high flux particle source for RGA-type erosion measurements. Mass spectroscopic observations of methane and heavier hydrocarbons in the exhaust gases of various tokamaks (recent examples: Refs. [54,58]) clearly indicate an active C/H chemistry. What is not clear is whether these hydrocarbons were produced by plasma/surface contact or by atomic hydrogen acting in the vicinity of the pump ducts [12]. Furthermore, hydrocarbons produced by plasma contact are likely to be dissociated very quickly and are unlikely to escape the plasma.

Evidence for the chemical erosion of carbon on tokamak wall surfaces can be obtained from observations of

CD and C_2 molecular band emissions in the edge plasma. Other chemical reaction products, such as BeD lines in JET [77] or BD lines in ASDEX-U [12], have also been observed. Further evidence for chemical erosion comes from the level of ionic carbon (or boron [32]) observed in the vicinity of divertor strike points [26,28,30,31,78]. With the great advances made in the quantification of spectroscopic information, it is now possible to have reasonable certainty (within a factor of two) on erosion yield measurements in tokamaks.

Direct measurement of hydrocarbon production in tokamaks by spectroscopic means is now available on at least four tokamaks. The probe limiter experiments on TEXTOR [34,59] are probably the most advanced, with the added advantage of having control over the surface involved. Calibrations, which include the processes involved in reaching the CH molecular state, as well as photon efficiencies, are performed by controlled gas puffing through the limiter itself. The results from TEXTOR, some of which are shown on Fig. 7, have provided us with data on RES [79], chemical sputtering (methane production) [59] and impurity transport [34] at very high flux densities and low (~ 100 eV) ion energies. At high temperatures, the CD band emissions decrease as expected, leading to a factor of 2 drop in CII and CIII line intensity in front of the limiter [55]. This indicates that about half of the carbon in the higher ionization states originated from chemical sputtering. TEXTOR, being a limiter machine, however, cannot simulate the very low energies and high fluxes of a high-recycling divertor, as expected to be the case in ITER. A further indication that chemical sputtering occurs at high flux densities (low ion energies) is the existence of negative velocities (i.e., velocities directed toward the limiter), which must originate from the dissociation of molecules [35].

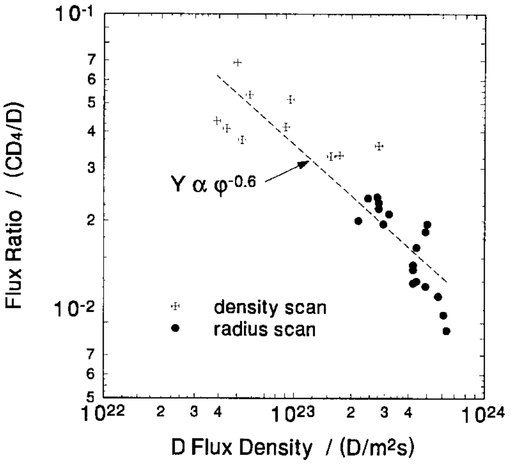

Fig. 8. CD_4/D flux ratio at the TEXTOR graphite test limiter ($T \sim 825$ K) measured by optical means [59]. Changes in flux density were achieved through variations of the plasma density (density scan) and variations in the radial position of the test limiter (radius scan).

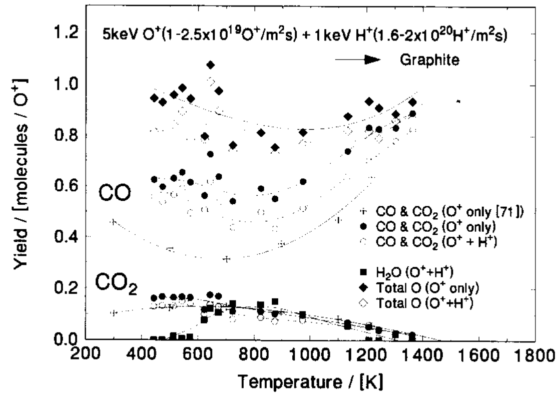

Fig. 9. Temperature dependence of the erosion of graphite due to combined H^+ and O^+ bombardment [76].

Measurements in Tore Supra [52] have been used to deduce heavier hydrocarbon production on the basis of C_2 band emissions, as well as methane production. The methane yield was 0.009 CD_4/D^+, and the total chemical yield was estimated to be 0.029 C/D^+. The relative size of the two yields is in reasonable agreement with laboratory measurements of chemical erosion at these energies [36,37] (100–200 eV). In later experiments [80,81], a temperature dependence for CD_4 production was found, but the C_2 signal was too weak to infer any yields.

In ASDEX-U, spectroscopic measurements of hydrocarbon influxes have been made at the main walls of the reactor. Under these circumstances, charge-exchange neutrals and recycling hydrogen atoms are dominating the incident hydrogen flux, and as a consequence, the flux and energy of the incident particles is not so well known. Naturally, there is no control over the surface temperature, which generally does not vary significantly from room temperature. By varying the edge temperature of the plasma, the effective bombarding energy of the escaping neutral flux can be varied, thus allowing a qualitative energy dependence for chemical erosion to be obtained. Under these circumstances, C_2 band emissions were observed to increase relative to CD band emissions as the edge plasma temperature decreased [82]. This can be explained by an enhanced production of heavier hydrocarbons (C_2H_x and C_3H_y) relative to methane, as observed in ion beam experiments [36,37], but also by an increasing transition probability with decreasing plasma temperature. At present, it is uncertain which process dominates [82]. Similar increases in C_2 band emissions were observed in detached TEXTOR discharges [59], with the same difficulty in interpretation.

While it is clear from the observation of CD [12,77,78], BeD [77] and BD [12] molecular spectra in the divertor regions of tokamaks that chemical erosion is occurring, the

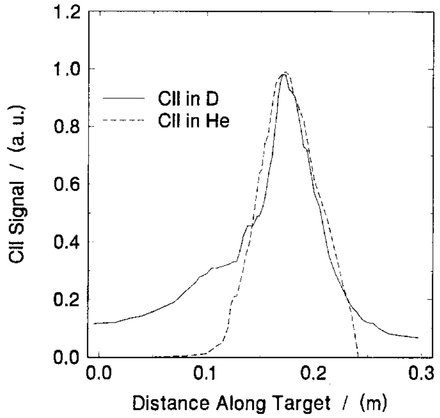

Fig. 10. Normalized radial CII intensity distribution across the outer target plate of ASDEX-U [12].

source of these impurities is not so clear. As in physical sputtering, interpretive computer codes are used to determine the extent to which erosion mechanisms other than physical sputtering are contributing to the carbon or boron impurity levels. As discussed in Section 2.2, spectroscopic observations of carbon or boron impurity levels in front of divertor target plates are well explained by physical sputtering alone [26,30–32]. Away from the strike points, either in the private plasma region, or towards the main plasma chamber, however, physical sputtering is not sufficient to explain the C impurity levels. Chemical effects are thought to be the cause of this discrepancy, and this is confirmed by results with helium discharges [12] (Section 2.2). In Fig. 10, we show a comparison of CII emissions in ASDEX-U discharges in He and D [12]. In the He discharges, the shape of the radial emission profile is very similar to that calculated by DIVIMP with physical sputtering only [31]. The source of the chemical erosion in the deuterium discharges, however, has not been clearly identified.

In the case of boron wall sputtering [32], again only physical sputtering is expected, and reasonable agreement is obtained between the observed and calculated (by DIVIMP) B influx; see Fig. 3. There is, however, a component of BII signal extending away from the divertor strike point which is not observed in the He discharges (Fig. 10) or predicted by DIVIMP. It is possible that boron/hydrogen chemistry (supported by BD band emissions [12,32]) might be leading to some of the discrepancy. A diffuse, low level chemical erosion wall source [32] may be the best explanation for this. However, laboratory measurements of B erosion generally give very low yields.

Chemical erosion sources have been employed to try to obtain agreement between the observed impurity distributions and impurity influx model calculations. In Ref. [29], the addition of chemical erosion to the calculation leads to much better agreement with the observed CII profiles than with physical sputtering only. This is demonstrated in Fig. 2, where the calculated results, including chemical sputtering, are also shown in comparison with those calculated by physical sputtering only. In Tore Supra [27,81], similar modelling with the BBQ computer code required the inclusion of chemical sputtering to match observed CII emission distributions. Even the general shape of the profiles could not be reproduced by the inclusion of physical sputtering only. In JET [30], however, the addition of chemical sputtering to the calculations did not in general lead to better agreement with measured profiles. In order to match the erosion yields in the private plasma region, the erosion yields at the strike points were much too high, unless a strong flux dependence was applied.

While part of the difficulty in fitting chemical erosion yields to tokamak divertor observations may be due to a lack of laboratory data at low energy, these low energy yields may not be relevant. Due to the simultaneous flux of more energetic carbon ions, a synergism may result in D erosion yields which are more characteristic of D^+ ions with much higher energy [83,84]. Reductions in divertor plasma temperatures below 10–20 eV may not have as large an effect as expected, as long as there is a few per cent carbon impurity as part of the flux. Similarly, charge exchange fluxes of D° or He° with several hundred eV could also have an enormous influence [36,85], even when they only comprise a small fraction of the total flux. The temperature dependence of the synergistic erosion yield will also be altered, making it very difficult to predict chemical yields under tokamak divertor conditions.

The fact that chemical sputtering does not appear to be important under the conditions at the divertor strike points, i.e., high flux density and low ion energy, suggests that there is indeed a flux density dependence to the erosion yield. Alternately, it has been postulated [30] that the plasma conditions lead to the prompt redeposition of hydrocarbon fragments, greatly reducing the effective erosion yield. Either mechanism could be supported by the observation of CD band emission across the strike zone [78].

4. Radiation-enhanced sublimation

A basic understanding of the processes involved in radiation-enhanced sublimation (RES) has been established for many years [4]. While the existing RES models are reasonably successful [86–90], there are still several questions which have not been dealt with sufficiently to ensure confident extensions to tokamaks. As in the case of chemical sputtering, predictions of the yield in a fusion reactor environment requires understanding of the energy, temperature and flux density dependence of the erosion yield. The effect of dopants, or surface coverage by other elements further complicates the picture. The temperature dependence is generally well represented by an exponential increase in yield with increasing temperature. Activation energies of ~ 0.8 eV [4,89,91] are characteristic of the

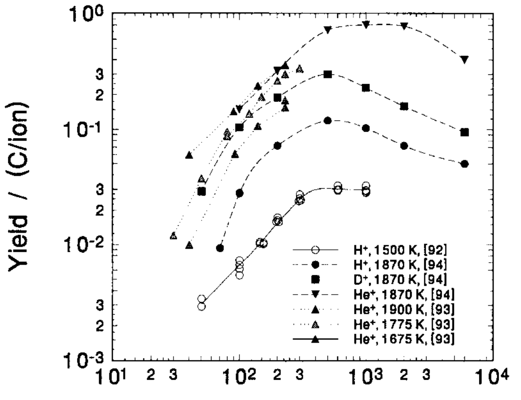

Fig. 11. Energy dependence of radiation-enhanced sublimation.

process, and clearly distinguish it from thermal sublimation.

Knowledge of the energy dependence of RES, for low impact energies of the incident particles, is necessary for the prediction of yields at surfaces exposed to high-density, low-temperature plasmas. Since RES involves the creation of interstitial/vacancy pairs, requiring ~ 20 eV dislocation energy, one would expect a threshold incident ion energy as is observed for physical sputtering. In Fig. 11, the energy dependence of the erosion yield is plotted [92–94]. The data confirm the expected threshold, indicating that exposure of carbon to low-temperature divertor plasmas would not lead to $H^+/D^+/He^+$-induced RES. Self-ion-induced RES may continue to be important at low plasma temperatures, due to the lower threshold energy, and the higher incident energy. Divertor target plates, which are the most likely surfaces to reach high enough temperatures for RES to occur, may be exposed to low energy plasmas such that the incident particles will be below the RES threshold.

The possibility of RES occurring in a fusion reactor due to high energy neutron bombardment has been considered by Franzen [95]. Calculations indicate that, while the RES process could occur due to neutron irradiation, and the levels could be an order of magnitude higher than neutron-induced physical sputtering, the levels are still much smaller than that due to charge-exchange neutrals.

The properties of RES which are current topics for investigation are the flux-density dependence and the influence of dopants.

4.1. RES as a function of flux density

The models for RES have consistently predicted a strong dependence of the erosion yield on the incident flux density, of the form $Y \propto \phi^{-\epsilon}$, where $\epsilon \sim 0.2$–0.3 [4,86–89]. Laboratory experiments with ion beams, however, have been equally consistent at demonstrating a flux dependence of $Y \propto \phi^{-0.1}$ [87,91,92,96]. The reason for this discrepancy is not clear, but new results at high flux density, using plasma sources, may help in our understanding. Results by Ueda et al. [90] and Ohtsuka et al. [97], with a high-flux, low-energy plasma ion source, display the type of flux dependence predicted by the models. The new results of Ueda et al. are shown on Fig. 12, along with the results of ion beam studies. The implication of this result is that the ion beam studies are at too low a flux density to see the stronger flux dependence. The damage done by 5 keV Ar^+ is about 50 times larger than due to 500 eV D^+ ions [90]. Thus a similar drop in yield due to D^+ would not be expected until fluxes of ~ 10^{22} D/m^2 s are reached. Because this is much higher than the fluxes achievable with ion beams, and is the upper limit of fluxes from H^+/D^+ plasma simulators, the effect would not have been observable in laboratory experiments with H^+ and D^+.

High heat flux surfaces in tokamaks should be at a sufficiently high flux density to be in the steeper-slope RES regime. If such a flux dependence holds (i.e., $Y \propto \phi^{-0.26}$), it would imply a factor of ~ 2 drop in yield for an order of magnitude increase in flux density. Therefore, on tokamak components which receive flux densities > 10^{24} D/m^2 s, RES may be difficult to see in comparison with physical sputtering.

4.2. Tokamak measurements

There are also other sources for high-flux RES measurements. Large increases in carbon influx (carbon blooms) were observed in JET and TFTR (and other machines) due to local overheating of carbon tiles. In both of these machines, two regions of enhanced carbon emission were identified: ~ 2100 to 2600 K and above 2600 K in JET [98] and ~ 1900 to 2600 K and above 2600 K in

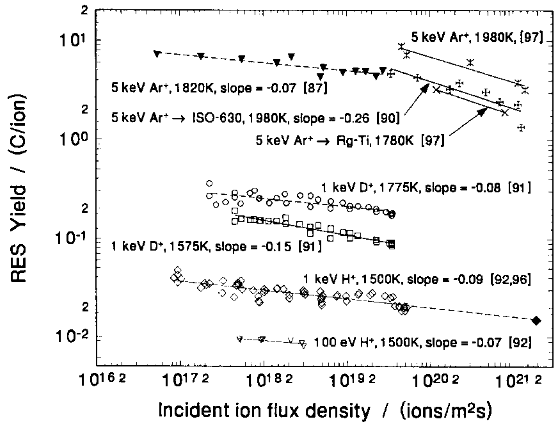

Fig. 12. Flux dependence of RES. Except where indicated, specimens are pyrolytic graphite. The low-flux data for 5 keV Ar^+ [87] has been scaled to match the data from Ref. [90].

TFTR [99]. The first region was interpreted as being due to RES, in particular due to incident carbon ions with yields near unity [99], and the latter region due to thermal sublimation. However, the temperature range, and the activation energy are higher than observed in beam experiments. In JET, Be contamination of the tile surfaces was thought to explain the discrepancy, or it could be a consequence of the high flux density. Because of these uncertainties, and the fact that it was a transient effect, the results cannot be thought of as conclusive evidence for RES in Tokamaks.

Often tokamak erosion measurements can be ambiguous and require careful interpretation; however, sometimes they can be very clear. Results from TEXTOR [34,79] with a probe limiter heated to sublimation temperatures showed only a 15% increase in released C flux for temperatures up to ~ 2300 K, while thermal sublimation was clearly indicated at somewhat higher temperatures. The ion impact energies, ~ 250 eV, are high enough to cause RES (see Fig. 11); the primary distinguishing feature of the experiment is the high flux density, ~ 2×10^{23} D/m^2 s. Energy distributions of the desorbing C atoms [34] confirm the energetic nature of the C atoms for temperatures < 2400 K, which is consistent with physical sputtering, while also confirming the thermal nature of the sublimating C atoms above 2400 K (see Fig. 4(a)). A complicating factor in these experiments may be the simultaneous flux of boron or other impurity species to the surface. However, since boron is not known to be a good suppressor of RES (see below), this does not seem a likely possibility.

A very different result has been obtained in Tore Supra, where spectroscopic measurements of C influx above an inertial limiter show the characteristic temperature dependence of RES [80,81], see Fig. 13. Even the activation energy, ~ 0.8 eV, is fully consistent with laboratory RES measurements. It is possible that changes in H$^+$ flux, and local hot spots on the graphite surface are contributing to the increase in C influx, although it is difficult to see how these features can account for the observed behaviour. At the present time, it is not possible to reconcile these results with those from TEXTOR. It is noted that the incident flux density at the leading edge of the Tore Supra limiter, ~ 1.6×10^{22} D/m^2 s, is an order of magnitude smaller than at the TEXTOR test limiter. The difference between the TEXTOR and the Tore Supra results may be an indication of a strong flux dependence; however, more recent results on TEXTOR at lower flux density ($2-3 \times 10^{22}$ H$^+$/m^2 s) [55] appear to contradict this. These results serve to indicate that a full understanding of RES under reactor conditions has not been achieved. The one positive note from the Tore Supra study was that, even though a strong increase in carbon influx was observed for $T > 1500$ K, there was no observed impact on the central plasma impurity density (presumably due to the low penetration velocity of the neutrals into the plasma).

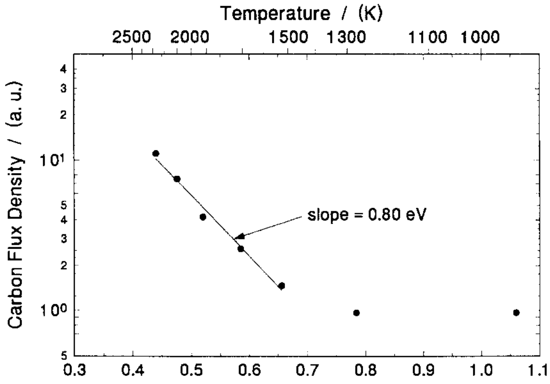

Fig. 13. Temperature dependence of the carbon influx from the ion side leading edge of an inertial limiter in Tore Supra [81].

4.3. RES of doped graphites

The effect of dopants on RES has not been studied to the extent as is the case for chemical erosion, and a consistent picture has not yet emerged. The most dramatic reductions in erosion yield have been observed for titanium-doped graphites. In 1990, Begrambekov [100] reported the complete suppression of RES for a Ti-doped specimen; however, other Ti-doped carbons, e.g., the Rg-Ti materials [101] and boron-doped graphites [9,101–103], did not exhibit such a suppression. More recently, Franzen et al. [104] also saw a large reduction in erosion for a Ti-doped specimen (~ 16 at.% Ti) as part of an extensive study covering several dopants at various concentrations. Other dopants (B, Si) did not lead to such large reductions, nor did specimens with lower Ti concentration, or even the same material in a different orientation. At this point, it is not clear what property of the Ti-doped graphites is responsible for reduction in RES.

Aside from the Ti-doped graphites discussed above, minor reductions in RES have been observed for a variety of doped graphites, but no consistent results point to a material which would satisfy reactor requirements. Small reductions in RES would likely be offset by the increase in temperature associated with decreased thermal conductivity.

Suppression of RES by metals evaporated onto the surface may more closely represent the situations in tokamaks. The evaporation of Ti onto graphite to maintain a surface concentration of ~ 10% led to a complete suppression of RES [105]. It may be that under certain conditions, sufficiently high Ti surface concentrations can be achieved by bulk doping, and lead to the same result. The lack of reproducibility may indicate a complex Ti/C transport. A multi-component machine, like ITER, is likely to result in the gradual coverage of low-Z materials with higher Z ones, e.g., the 'carbon poisoning' observed in Be erosion

[106]. The coverage of carbon surfaces with tungsten may be sufficient to eliminate RES, without the need for doping. Also, the suppression of RES in the TEXTOR probe limiter experiments (after boronization) may already be a consequence of such an effect.

5. Summary and future requirements

With the improvements made to erosion measurements in tokamaks, it is becoming possible to make reasonable comparisons between laboratory measurements and those made under actual fusion conditions. With respect to physical sputtering, the agreement within a factor of 2 is probably equivalent to the error on both the laboratory and tokamak results. The primary need for laboratory information is in the area of very low energy (near threshold) sputtering of Be, C and W, especially under conditions of simultaneous bombardment by several species. The flux dependence of chemical erosion will be a critical factor in determining carbon erosion yields; the evidence so far points to a very minor effect. The measurement of the flux dependence in current tokamaks is likely to provide the best indication of the magnitude of the problem for future machines.

Results from tokamaks will also be required to sort out the flux dependence of RES. The large disagreement between results at TEXTOR and Tore Supra could mean that a strong flux dependence is in operation (TEXTOR result), or alternately that laboratory measurements may be extrapolated to reactor conditions several orders of magnitude higher (Tore Supra result). TEXTOR-like probe limiter experiments in the all-carbon Tore Supra may be an excellent way to shed light on the discrepancy.

The doping of carbon with B has successfully demonstrated a reduction in chemical erosion by up to an order of magnitude. It is unlikely though, that such reductions would be observed in future machines where the particle flux is dominated by low-energy particles, and the component temperatures are kept at < 600 K or > 1000 K. Similarly, Ti-doping has been found to almost entirely suppress RES in some doped carbon materials, but these results are very material specific. It is not clear that doped graphites would possess any advantages over undoped ones in a machine like ITER, but they cannot be ruled out either. It would probably be reasonable to continue testing more doped materials, possibly including materials with multiple dopants. There would be some reduction in thermal conductivity, however, it is not clear that heavily neutron-damaged doped graphite will have a significantly lower thermal conductivity than neutron-damaged undoped graphite.

While in some cases [80,81] the local influx of carbon due to chemical sputtering and RES on limiter surfaces has not been clearly linked to core impurity levels, contrary results again exist [55]. Even if such erosion mechanisms do not lead to core contamination, they will make a contribution to the formation of codeposited layers and hence, tritium inventory. On the other hand, experiments [54] and interpretive computer modelling [2] indicate that wall chemical erosion may dominate core impurity levels in divertor tokamaks.

Lastly, with a multi-material machine, the transport of different materials onto carbon or beryllium surfaces may greatly alter the erosion process. This may make it extremely difficult to predict erosion rates, but then, the erosion yields are likely to be lower due to this coverage.

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