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THE APPLICATION OF SECONDARY ION EMISSION TO IMPURITY CONTROL IN TOKAMAKS*

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The extent to which high Z impurities enter the plasma of a magnetic confinement fusion device depends on the kinetic energy, angle of emission, and very importantly, the charge state of the ejected material. We have been studying both the fundamental process of secondary ion emission and possible techniques for producing surfaces which give rise to high ion fractions during sputtering, with a view to assessing the potential of this approach to impurity control in tokamaks. By carefully choosing materials exposed to fusion plasmas and by properly modifying the surface it may be possible to insure that nearly all the impurities are ejected as ions. As long as certain gas blanket configurations are avoided and especially if a divertor is used, it should then be possible to remove the impurities before they reach the plasma. The relative merits of a variety of materials are considered with regard to this application.

1. INTRODUCTION

It is generally recognized that plasma impurities can seriously limit the performance of magnetic confinement fusion devices. The solution to the problem however, is not so generally agreed upon. Discharge cleaning has been successfully used on a number of devices to remove oxygen and carbon [1]. Titanium gettering has been used on PLT to reduce re-admission of cold hydrogen to the plasma. This technique has been partly responsible for the improved performance of PLT but tritium retention needs to be examined before Ti gettering can be used with a D-T plasma in TFTR.

As reactor conditions are approached, sputtering becomes a more dominant impurity release mechanism and will result in the introduction of wall and limiter material into the plasma. It has already been observed that the successful reduction of oxygen impurities has resulted in an increased concentration of tungsten in the plasma [1]. Because these atoms have high atomic number, they produce much more serious energy losses than comparable levels of oxygen and carbon.

Proposals for reducing the high Z impurity influx fall into two categories: Modifying the plasma edge and modifying the wall surfaces. Assuming that most of the influx is the result of sputtering by charge-exchange neutrals originating in the plasma edge, the sputtering yield would be reduced by lowering edge temperatures by techniques such as cold gas puffing. Alternatively, a magnetic divertor creates a "scrape off" region in which ions at the plasma edge are prevented from colliding with the wall and ions ejected from the wall are removed before they enter the plasma. For divertor action to be effective, neutral sputtered particles must be ionized, requiring the use of a gas layer

[2] to effect charge exchange ionization. The risk inherent in this approach is that the gas layer would most likely increase the charge exchange neutral flux to the wall and thus be self-defeating.

Although current tokamak first walls are made of stainless steel, it is likely that reactor first walls will require the use of other materials. Coatings of low Z materials such as Li, Be, B and C have been proposed [3]. For carbon however, the enhanced chemical sputtering yield associated with methane formation at elevated temperatures may be a serious problem.

A novel approach to impurity control first suggested by Krauss and Gruen [4], is to select wall materials in such a way as to maximize the fraction of atoms which are ejected from the wall as ions. Three possible fates of particles ejected from a tokamak surface facing the plasma are shown in Fig. 1. If the particle ejected from the surface is neutral, it will penetrate the scrape-off region (in a divertor operated tokamak) to a depth determined by the kinetic energy and trajectory of the particle, and density of the gas in this region. If the particle becomes ionized in the scrape-off region, it will be swept into the divertor. Otherwise, it may penetrate the plasma (a). If, on the other hand, the particle is ejected as a low energy ion, it may simply return to the wall (b). This mechanism of impurity removal would be operative in tokamaks with and without divertors. If the ion has high kinetic energy and a small ejection angle, it will penetrate farther before being deflected by the magnetic field. Depending on the velocity, the magnetic field gradient, and the absence of a high particle density close to the wall, the ion may still be guided by the field lines into the limiter or into the divertor (c). The existence of a sheath potential would further

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suppress secondary ions.

Neutrals are ionized in a very short distance once they enter the plasma edge, but they are then subject to a variety of complicated plasma transport processes which have been observed to allow impurities to reach the plasma center. At the wall, the particle density is much lower and the mean free path of a sputtered ion far exceeds both the gyro-radius and the distance at which an ion would be turned back by a sheath of 100-200 V/cm. However, the extent to which ions ejected with a given energy and angular distribution will actually be blocked from entering the plasma has not yet been determined. From Fig. 1 it may be seen that at least qualitatively, the impurity influx could be strongly affected by the charge state, kinetic energy, and angle of emission of the sputtered particles.

Fig. 1. Schematic cross-section of a divertor-equipped tokamak showing some of the possible fates of a particle ejected from the wall as: a neutral (a), a low energy ion (b), and a high energy ion (c).

2. EXPERIMENTAL

In our laboratory, we have, for some time, been studying a variety of techniques leading to increased ion fractions in sputtering with a view to assessing the potential of this approach for impurity control in Tokamaks [4-7]. The ion beams, used for the sputtering experiments are produced by either a Varian 3 keV sputter ion gun or a Danfysik 80 keV isotope separator. The ion analyzer consists of an electrostatic energy analyzer followed by a quadrupole mass analyzer. A more detailed discussion has been presented elsewhere [5]. By summing counts during the acquisition of secondary ion energy distributions, relative secondary ion yields may be obtained, corrected for the transmission versus energy characteristics of the analyzer

but not for transmission versus mass or the angular variation of the secondary ion emission.

3. DISCUSSION

3.1 Theories of secondary ion emission.

In order to fully exploit the ion fraction approach to impurity control, the processes which determine the ion fraction yields have to be much better understood than they are at present. A number of theories which account for some aspects of secondary ion emission have been advanced, but none are completely satisfactory. Recent analysis of the secondary ion emission of Be [7,8] and Al [9] indicates that several mechanisms may be operating simultaneously. Some of these processes will lead to ionization of neutrals while others will neutralize previously formed ions. Still another process destroys the excitation which could produce auto-ionization. The probability of producing a secondary ion may be written [8,10]:

$$R^+ = \sum_{n,m} \int_0^\infty p_n^{\text{ion}} \cdot p_m^{\text{sur}} dt \quad (1)$$

where p_n^{ion} is the rate at which ions are produced by process n and

$$1 - p_m^{\text{sur}}$$

is the probability that a secondary ion will be neutralized by process m .

3.2 Charge fraction

The charge fraction is defined as:

$$\beta \equiv S^+/S = \int_0^\infty R^+(E) dE \quad (2)$$

where S^+ and S are the secondary ion and total sputtering yields respectively. Figure 2 shows the variation of β for various elements as compiled from a number of literature references [11-15]. For those references which list S^+ rather than the charge fractions, β has been determined by Eq. 2 where S has been calculated from the semi-empirical sputtering model of Smith [16]. It can be seen that there is substantial discrepancy between published values. Many of these values have been challenged as erroneous as a result of surface contamination even under high current density conditions [17]. There is however, substantial variation from element to element and from the clean to the oxidized state of a given element. Since oxygen forms an ionic bond with most metals, a metal atom associated with such a bond frequently escapes as an ion when ejected from the surface. Several authors have suggested that this effect be exploited for impurity control in Tokamaks [4,18-20].

Instead of introducing oxygen, another approach would be to choose materials which have a high ion fraction in the unoxidized state. It can be seen from Fig. 2 that of all the elements the alkali metals produce the highest ion fraction of sputtered products. The alkali metals and possibly the alkaline earths are also the

only elements to form an ionic bond with hydrogen [21], and one might therefore expect that the inevitable presence of surface hydrogen in a reactor environment would further increase the already high ion fraction of these elements. However, not only the ionic nature but also the bond strength determine the ion fraction of the sputtered products. Thus for titanium, the positive ion fraction for an oxygen covered surface lies between ~20 and 100% [13,6,22] and for hydrogen coverage may go as high as ~40% [4]. On the other hand for beryllium which may have an ionic molecular bond with hydrogen but is a very weak hydride former, hydrogen has no effect on the secondary ion yield [7].

Still another approach makes use of a technique analogous to high energy discharge cleaning to alter the chemical composition of the near-surface region, to a depth of several tens of microns [23]. Such in-situ discharge-induced ion implantation techniques could be used to modify surfaces in such a way as to lead to increased ion fractions.

Fig. 2. Plot of the secondary ion fraction (β) for forty two elements. Open symbols are for nominally clean surfaces and filled symbols are for oxygen-covered surfaces. Refs. are: Δ Andersen (14), $\circ$ Benninghoven (13), $\triangle$ Reske (15), $\times$ Blaise (33), $\square$ Krauss and Gruen (6,7), and ∇ Schroerer (12).

3.3 Energy distribution of secondary ions.

The energy distribution of atoms sputtered from a random solid by long collision cascades is expected to be similar for all elements, [24] the main difference being the position of

a low energy peak which varies from ~0.5 to 4.5 eV. Because of the velocity dependence of the ionization and survival probabilities, the secondary ion energy distribution may look quite different from that of the neutrals. In particular, the high energy tail falls off more slowly and the peak may be shifted to much higher energies. Figure 3 shows some representative secondary ion energy distributions. It should be noticed that the alkali metal distributions peak at very low energy (<1 eV) and then drop very rapidly with energy. Tungsten on the other hand displays a very broad maximum with a substantial number of secondary ions having kinetic energies of several hundred eV.

Long random collision cascades are not the only mechanisms by which atoms may be sputtered. Short cascades and direct and deflected surface recoils account for a relatively small proportion of the total sputtering yield but since they tend to produce high energy particles, they may penetrate the plasma more effectively. However the ion fraction associated with these

Fig. 3. Representative secondary ion energy distributions for 3 keV Ar^+ on seven elements. The vertical scales have been shifted for display purposes and do not represent relative yields.

mechanisms is high. Surface recoil emission depends on the exposed crystal face. Preferential sputtering during a long bombardment of a polycrystalline sample will produce faceting and the exposed low sputtering yield planes are more likely to eject atoms by surface recoil. Such behavior is observed in the Ti^+ energy distribution of Fig. 4 where a second peak consistent with deflected recoils is observed and the bulk of the secondary ions have kinetic energies between 100 and 1500 eV. Similar behavior has

been observed for Al^+ [25] and Au neutral [26] sputtering after prolonged argon ion bombardment.

Not only the ion yields, but also the secondary ion energy distributions are sensitive to the chemical state of the surface. The quantities P_{ion} and P_{sur} are sensitive to such quantities as the conduction electron concentration [27] and density of states [28], the work function [12,27], screening time [29], and the local surface dipole moment [30]. All of these quantities are strongly influenced by strong and especially by ionic bonds.

Ions escaping from a clean metal surface usually have a very high probability of neutralization, unless they escape with a very high

Fig. 4. Secondary ion energy distributions illustrating E^{m-n} behavior for K^+ and Al^+ . The double-peaked distribution for Ti^+ indicates a substantial contribution of recoil emission to the secondary ion yield.

velocity. Consequently the ion distribution is shifted toward higher energies. We may write:

$$N^+(E) = R^+(E) N(E) \quad (3)$$

where $R^+(E)$ is defined in Eq. 1 and $N^+(E)$, $N(E)$ are the ion and total energy distributions, respectively. If certain properties of $N(E)$ are either known or assumed, it is possible to derive $R^+(E)$ from the experimental secondary ion energy distribution. The information may then be used to elucidate the mechanisms of secondary ion emission. This has been done for beryllium [7,8].

3.4 Angular distribution of secondary ions.

The random collision cascade model of sputtering [24], predicts that for kinetic energy of the sputtered species $\ll$ than that of the pri-

mary projectile,

$$N(E) = \frac{CE}{(E+E_b)^3} \frac{\cos \phi_e}{\cos \phi_i} \quad (4)$$

where ϕ_e is the angle of emission and ϕ_i is the angle of incidence, both measured relative to the surface normal, E_b is the surface binding energy, usually taken as the heat of sublimation, and C is a materials-dependent parameter. The shape of $N(E)$ is predicted by Eq. 4 to be independent of both ϕ_e and ϕ_i . However, it has been observed that the peak of the Al^+ energy distribution depends on the angle of emission [6,31](Fig. 5.) The reason for this behavior is found in the angular dependence of the ion neutralization process.

It is to be noticed in Fig. 4 that at high energies the Al^+ distribution is given by $N^+(E) \sim E^{-1.4}$. This may be written as:

$$N^+(E) \sim E^{m-n}. \quad (5)$$

This type of behavior has been predicted by several authors [11,12,14], as the form expected for pions although the models used appear to be on somewhat uncertain ground. Adding the ion survival probability we expect [10]:

$$R_{\text{Al}}^+(E) \sim E^{m-n} \exp(-A/v_l). \quad (6)$$

The dependence on v_l occurs because the neutralization probability is determined the the amount of time the secondary ion spends in the near-surface region. Using the parameters $m = .6$ and $n = 2$ the peak shift observed in Fig. 5 can be reproduced (arrows) with a value of $A = 2.5 \times 10^3$ m/sec. This value is essentially in agreement with the value obtained from the doppler broadening of the lines associated with bombardment-induced photon emission [32].

Fig. 5. Variation of secondary ion kinetic energy with angle of emission. The data is identified by the corresponding value of ϕ_e . The arrows indicate the peak position calculated from Eq. 6 using the parameters $m = .6$, $n = 2$, $A = 2.5 \times 10^3$ m/sec.

4. SUMMARY

The extent to which impurities may be controlled by the use of surfaces with a high secondary ion fraction depends on the details of the transport processes for a particle originating as an ion. Such an assessment has not yet been made. The process of secondary ion emission

is a complicated and poorly understood one, and a large number of questions must be answered before it can be determined whether the technique will be useful. Qualitatively, the efficiency with which sputtered impurities enter the plasma is determined by the charge state, kinetic energy and angle of emission from tokamak surfaces. Crystallographic effects appear even in polycrystalline material and give rise to high energy ejection at oblique angles to the surface. The intensity of emission however, is much reduced compared with emission along the surface normal, and so the severity of this problem is uncertain.

By carefully choosing materials exposed to fusion plasmas and by properly modifying their surfaces, it may become possible to insure that nearly all the impurities ejected from such surfaces by sputtering are in the form of ions. As long as certain gas blanket configurations are avoided and especially if a divertor is used, it should then be possible to remove the impurities before they reach the plasma.

The refractory metals such as W and Mo generally favored for limiter design unfortunately produce very high energy sputtering products. In the clean state, sputtering is overwhelmingly in the form of neutrals. By oxygen treatment the charge fraction may be increased but still remains below ten percent. Common structural materials such as Fe and Cr have lower kinetic energies when sputtered and Cr in particular responds well to oxygen exposure [13]. However, the ion fraction of Fe remains below 10% and neither metal shows any marked sensitivity to hydrogen exposure. The hydride-forming metals such as Ti and V undergo a desirable increase in charge fraction upon exposure to hydrogen. They may however, present a tritium retention problem.

The alkali metals give the highest charge fraction and lowest kinetic energy of all the elements surveyed and are therefore good candidate materials for tokamak surfaces exposed to plasmas. However, they possess unfavorable chemical and thermodynamic properties. Lithium presents the fewest problems from this viewpoint, but still is not suitable for use as an elemental coating in a reactor. A number of alkali metal alloys are known to be much less reactive and have thermodynamic properties similar to those of the major alloying element [34]. Because of its low surface binding energy and large atomic radius segregation of alkali metal is expected to occur at the surface [35,36]. Since this is the region from which sputtering originates, the impurity emission could therefore be expected to be quite similar to that of the bulk alkali metal. It should be noted that not only the elevated operating temperature but also the radiation environment could promote surface segregation [37]. Lithium may be especially attractive because of its low atomic number and relatively high solubility in a variety of less volatile metals. Secondary ion emitting properties of such lithium containing alloys are currently being studied in our laboratory.

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