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## Viewpoint Set

## Sputtering in a high-flux plasma environment

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## ABSTRACT

Sputtering yield measurements of metals using ion beam devices and high-flux plasma facilities are compared. During light inert gas bombardment, a general decrease in the sputtering yield is observed as the flux of energetic particles to the target increases. However, during heavy inert gas bombardment, the decrease is not observed and measurements agree with ion beams and binary collision approximation calculations. A potential cause, accumulation of gas atoms within the implantation zone, is hypothesized to be the culprit and various in-situ surface diagnostic techniques to measure the gas composition in the near surface of the metal targets are discussed.

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## 1. Introduction

One of the most fundamental properties of plasma-material interactions is sputtering [1]. The term sputtering is used to describe a variety of events; physical sputtering [2], chemical sputtering [3], chemically enhanced physical sputtering [4] and temperature enhanced sputtering [5]. In this paper we will focus in on aspects of physical sputtering.

The physical sputtering mechanism involves the transfer of momentum from incoming energetic particles to atoms residing near the surface of the wall material. If the transfer of energy is such that a lattice atom receives sufficient energy normal to the surface to overcome the surface binding energy, the lattice atom can be released from the surface and a sputtering event is said to have occurred. Sputtering is to a large part responsible for the lifetime of plasma-facing components, the impurity contamination of the impinging plasma and is the primary source term for material migration throughout the plasma generating facility. On the more positive side, the sputtering process has been utilized as a surface diagnostic tool (i.e. Secondary Ion Mass Spectroscopy (SIMS)) and industrial processes, such as thin film deposition, have their roots in the sputtering process.

Single particle sputtering events have been successfully modeled using binary collision approximation (BCA) methods, such as TRIM [6] and its many offshoots. Ion beam experiments have been used to benchmark the models and large collections of sputtering data involving a variety of projectiles and surface materials are available [2,7]. By and large the physical sputtering process is believed to be a well understood phenomenon.

However, like most descriptions of physical processes in the dilute-solution approximation, the simple picture of the sputtering process also changes as the flux of energetic particles to a surface increases. As the surface composition changes due to the incoming flux of particles, some of which remain in the surface, the sputtering rate of the original lattice atoms can change. In the case of high temperature plasma used in controlled thermonuclear fusion research, where the flux of particles to surfaces can become extremely large; the sputtering rates can become altered by significant amounts. In this paper we will examine the physical sputtering behavior of metallic surfaces exposed the large fluxes of energetic particles.

One of the conceptually simplest measurement techniques for sputtering is to monitor a change in mass of either the target being sputtered, or a collector plate onto which sputtered particles deposit. The simplicity of the technique has allowed it to be used for decades [8] and improvements in technology have made for more precise measurements. However, one difficulty with the interpretation of mass-loss measurement is the influence of gas adsorption on the surface [9]. During low energy and/or low fluence experiments, where the mass change is small, gas adsorption can cause a significant change in the overall mass of either the target or the collector. One obvious solution for this difficulty is to go to higher flux experiments, where the mass change becomes larger and hence the influence of gas adsorption less significant.

As the local dynamic surface concentration is a balance between the incoming flux and the diffusion into depth and backwards to the surface with increasing flux the concentration increases. The accumulation of the incident species in the material can significantly alter the surface composition and lead to changes in sputtering. Also, the probability for interactions of the sputtered particle with the incident flux of energetic particles is increased and all these effects must be taken into account during the interpretation of the results. This article will first

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review the general trend of a reduction in the sputtering yield of metallic samples exposed to high flux plasma observed in a variety of plasma facilities and will then present a new series of measurements where the incident flux of plasma was systematically varied while measuring the sputtering yield.

## 2. Motivation

The majority of the sputtering results discussed here are from measurements using the PISCES linear plasma facilities [10,11], although similar reductions in sputtering yield observed in other high flux plasma devices will also be mentioned where appropriate. The majority of measurements taken in PISCES-B are related to erosion of beryllium and first those will be briefly summarized.

Beryllium is a chemically active element that readily forms a stable oxide. In early low-flux ion-beam sputtering measurements using beryllium targets, it was speculated that the measured low erosion rate was related to the presence of the surface oxide. Clever experiments [12] were devised where the temperature of the target was elevated (to about 900 K) such that bulk Be atoms were able to migrate through the surface oxide layer, thus presenting a 'clean' Be surface for interaction with the incoming ion beam. These elevated temperature experiments resulted in sputtering yield measurements much closer to BCA calculations.

Initial weight-loss sputtering yield measurements in PISCES-B also showed a yield consistent with sputtering of BeO (a factor of 5–10 less than that expected from clean Be) [13]. However, spectroscopic measurements showed a quick removal of oxide from the sample surface during the initial part of the plasma exposure [14]. In addition, Auger Electron Spectroscopy measurements of the Be sample surface showed very little oxide residing on the surface after the termination of the plasma discharge. Also measurements of the sputtering yield at elevated temperature (920 K) did not show an increase, as was observed in the ion-beam measurements. Having ruled out the influence of a surface oxide, other possible explanations for the low yield measurements were investigated. It should also be noted that during previous studies of codeposition of eroded Be with D in the TPE linear plasma device [15], the erosion rate was well below that expected from BCA calculations.

An observed difference between typical ion beam sputtering measurements and high-fluence plasma erosion measurements is the development of surface morphology on the plasma-exposed targets. Surface morphology had been observed on Be samples exposed in PISCES-B following the experiment. Be line emission from directly in front of the target during the plasma discharge was used to determine the temporal evolution of the sputtering rate. It was determined that the erosion rate of Be dropped concurrently with the formation of the surface morphology. However, the reduction could only account for a reduction factor of 2–3 and not the entire 5–10 that was needed to explain the sputtering yield discrepancy with BCA calculations [16]. The factor of 2–3 reduction in erosion due to surface morphology formation is consistent with high-fluence ion-beam sputtering measurements reported in the literature [17]. It is also important to note that at elevated surface temperature, morphology does not develop on the surface [18] yet the measured sputtering yield is still well below BCA estimates.

In order to avoid possible complications involved with deuterium plasma experiments [19], such as chemically assisted physical sputtering (of BeD molecules) and the influence of molecular ions (i.e.  $D_2^+$  and  $D_3^+$ ) measurements were made with noble gas (He and Ar) plasmas. While the sputtering yield reduction, and morphology formation for He, were similar to those of D, the erosion behavior in Ar plasma exposures was measured to agree with BCA calculations [18]. In addition, the temporal behavior of the erosion rate, measured spectroscopically during Ar plasma, was constant throughout the plasma exposure. Investigation of the surface after the Ar plasma exposure also revealed a smooth Be surface. These measurements tended to corroborate the

influence of the role of surface morphology in changes in erosion, but again the effect was too small to explain the entire discrepancy.

The plasma environment also presents differences to ion-beam sputtering measurements in the probability of eroded particles becoming ionized in the plasma and transported back to the sample surface where they are redeposited, effectively reducing the weight loss measurement. The 3D Monte-Carlo impurity transport and plasma-surface interaction modeling code, ERO, was adapted to the linear geometry of PISCES-B to determine the redeposition probability of sputtered Be atoms. ERO predicted only about a 20% correction to the weight loss data due to redeposition effects, again insufficient to explain the measured discrepancies [20].

Finally, the large flux of energetic particles to the plasma-exposed surface can result in a large fraction of the incoming particles residing near the surface of the target. The presence of gas atoms in the lattice can have multiple effects. First, it can reduce the concentration of the target species in the implantation region and thereby reduce sputtering due to collisions of the incoming particles with the concentration of gas atoms. On the other hand, it can also reduce the binding energy of the lattice atoms and act to increase the sputtering rate of the target atoms. Molecular Dynamics (MD) simulation of the bombarded sample surface was employed to simulate the magnitude of this effect. While the results of the simulations showed a small decrease in the erosion rate of target atoms [18], performing the simulations at increasing concentrations of gas atoms in the surface proved difficult with problems stemming from the likelihood that the near surface region is far from equilibrium. The MD simulations were used to generate an upper and lower bound to the Be erosion rates [21] based on the observations in PISCES-B experiments.

All this research eventually led to systematic erosion measurements of Be using D plasma in the JET-ILW confinement facility [22]. For simplicity limiter plasma operation was used and the results modeled using the ERO code adapted to the JET-ILW geometry. In these experiments the modeling showed agreement with the experimental data only when a larger reduction in erosion, as compared to PISCES-B data, was used. These results confirmed the fact that the erosion reduction was not an artifact of the linear geometry and verified the deviation from BCA calculations during high flux plasma interactions with surfaces.

Although all the detailed measurements discussed above involved using beryllium as the target material, the effect is not limited to beryllium. A variety of other metals have been investigated and similar discrepancies exist for light inert gas (LIG) plasma bombardment. As is the case with Be, better agreement to BCA estimates of sputtering are obtained when heavy inert gas (HIG) plasma is used to bombard the metallic targets. Fig. 1 is a plot of the results obtained using several different metals as targets in PISCES A&B. The plot compares the experimentally determined sputtering yield from weight loss measurements to the yield obtained by Eckstein using a version of the TRIM code [2]. As can be seen the results using either He or D plasma fall far below the calculated yield, whereas the yield measured using Ar, or Xe, plasma agree with, or are slightly larger than, the calculated values. As a simple crosscheck that there was no systematic error in the experimental data used to interpret the sputtering yield, the erosion of graphite by He plasma was measured and found to agree with the yield predicted by BCA codes.

Erosion measurements in Pilot-PSI have also shown a large reduction in the measured sputtering yield of helium plasma bombardment of Al and Ti compared to BCA calculations [23]. Fig. 2 is reproduced from [23]. Interestingly, in the Pilot-PSI no reduction in the sputtering yield of Cu by He was observed, although the ion energy was extremely close to the threshold value which may make the comparison to BCA estimates challenging. Surface morphology was believed to play a minor role in the erosion measurements, in agreement with the earlier PISCES erosion measurements. However, in these Al experiments the deviation from TRIM calculations was about three orders of magnitude. A big

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