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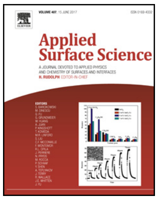

Fluence-dependent sputtering yield of micro-architectured materials

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ABSTRACT

We present an experimental examination of the relationship between the surface morphology of Mo and its instantaneous sputtering rate as function of low-energy plasma ion fluence. We quantify the *dynamic evolution* of nano/micro features of surfaces with built-in architecture, and the corresponding variation in the sputtering yield. Ballistic deposition of sputtered atoms as a result of geometric re-trapping is observed, and re-growth of surface layers is confirmed. This provides a *self-healing* mechanism of micro-architectured surfaces during plasma exposure. A variety of material characterization techniques are used to show that the sputtering yield is not a fundamental property, but that it is quantitatively related to the initial surface architecture and to its subsequent evolution. The sputtering yield of textured molybdenum samples exposed to 300 eV Ar plasma is roughly 1/2 of the corresponding value for flat samples, and increases with ion fluence. Mo samples exhibited a sputtering yield initially as low as 0.22 ± 5%, converging to 0.4 ± 5% at high fluence. The sputtering yield exhibits a transient behavior as function of the integrated ion fluence, reaching a steady-state value that is independent of initial surface conditions. A phenomenological model is proposed to explain the observed transient sputtering phenomenon, and to show that the saturation fluence is solely determined by the initial surface roughness.

1. Introduction

The physics of plasma–material interactions (PMI) has a wide range of applications across various technologies, including electric propulsion, plasma processing, microelectronic fabrication, and fusion energy devices. It is now appreciated that in PMI, the uniformity of plasma interaction with surfaces is the exception rather than the norm. Spatially non-uniform interaction leads to two potentially interesting areas of study. The first one is the possibility of fabricating surfaces with self-organized nano structures, and the second intriguing aspect is the potential for the surface to heal itself as a result of immediate deposition of some of the sputtered atoms. The two areas are complementary, since spatial non-uniformity of PMI can destabilize even flat surfaces, and lead to self-organization, while an initially structured surface may tend to become more uniform via ballistic atom deposition. The present study focuses on the second facet of PMI non-uniformity, in an attempt to explore how an initially structured surface can be designed to partially heal itself. Additionally, we wish to quantify the dynamic relationship between surface morphology evolution and the sputtering erosion rate as function of ion fluence. Understanding how surface structure and roughness affect the rate of atomic sputtering, and vice-versa, is a key consideration in engendering longevity and resilience to materials used in plasma devices, for example in electric space propulsion applications.

Utilization of energetic ion beams to produce surface nano-patterns has been the subject of intense interest for a couple of decades. Experimental and theoretical efforts have focused on understanding the influence of such parameters as the ion energy, flux, fluence, angle of incidence, and sample surface temperature on the formation of specific surface patterns. Makeev et al. [1] and Behrisch and Eckstein [2] have provided in-depth reviews of such studies that have focused on explaining the physics of plasma sputtering. Early insights into the relationship between surface structure and the sputtering rate were first presented by Sigmund [3], who formulated the theory of atomic sputtering from a flat surface. The theory was later expanded to consider the influence of surface roughening [4]. The basic idea is as follows: a collision cascade is formed downstream of an obliquely incident ion just beneath the surface, and the energy from the cascade causes ejection of surface atoms when the energy exceeds a critical value (binding energy). If the surface is not locally flat (rough), surface peaks will tend to eject less atoms (because the cascade is somewhat farther away), while valleys will tend to eject more atoms. This

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fundamental idea was further advanced by Bradley and Harper (BH) [5], who showed that surface instability ensues, and that roughening and patterning of surfaces bombarded with energetic ions may indeed be feasible.

The theoretical assumptions of these sputtering models have inspired the examination of the influence of surface roughness on the sputtering rate through experimental efforts. Sputtering yield, defined as the ratio of ejected atoms from the surface to incident ions, has been recently shown to be dependent on the surface nano- and micro-structure [6]. Surface atoms that receive sufficient energy from the ion collision cascade that exceeds the binding energy will be removed, and will either escape or interact with an adjacent surface feature. In the latter case, the ejected atom may have a chance to be deposited on a nearby surface feature, and become part of the evolving surface itself. Thus, the surface structure is a dynamically evolving system, where atoms can be removed or deposited in a way that depends on the surface evolution history. Such geometric re-trapping of atoms offers an opportunity to design sputter-resistant surfaces that have self-healing properties.

The relationship between the surface structure and the rate of sputtering erosion has been experimentally explored. Rosenberg and Wehner first examined the influence of surface roughness on a material's sputtering yield. They compared the sputtering yield of a smooth nickel rod bombarded by Ar^+ ions from 70 to 600 eV to that of a threaded rod with a pitch of 0.45 mm [7]. The experiment showed that surface roughness produces an overall decrease in the measured sputtering yield as a result of increased geometric trapping of sputtered atoms. This finding inspired future explorations to understand how varying roughness parameters quantitatively affect the sputtering yield. Huerta et al. [8] have used a view factor model to further examine the sputter deposition behavior observed by Rosenberg and Wehner. Their model found significant decreases in net sputtering yield for surface pitch angles beyond approximately 45°, due to forward-biased sputtering that favors deposition of sputterants into surrounding surfaces.

A number of efforts have been made to observe the reduced sputtering yield of rough or structured surfaces. Ziegler et al. used Chemical Vapor Deposition (CVD) to cover tungsten surfaces with tungsten whiskers, which were up to 80 μm high. They found that the whiskers dramatically reduced the sputtering coefficient for various ion energies, using 2–3 keV He^+ ions to a dose of $2.8 \times 10^{23}\,m^{-2}$ [9]. CVD has also been used by Hirooka et al. to produce textured molybdenum surfaces characterized by dome-like and faceted features on the order of 1 μm in diameter. The textured surfaces showed little damage compared to a polished sample, each under 40 keV He^+ ion irradiation dose of $3 \times 10^{22}\,m^{-2}$ [10]. Exposure of beryllium surfaces to 1000 eV H^+ ions has shown to produce high-angle, closely packed nano-cones at irradiation doses near $7.3 \times 10^{21}\,m^{-2}$ [11]. As the cones develop, the sputtering yield of the surface was found to be reduced by 30–40%. More recent efforts confirm these findings [12,13], in which beryllium exposed to 100 eV heavy hydrogen plasma at a fluence dose of $3 \times 10^{25}\,m^{-2}$ causes a needle-like morphology to emerge at higher ion fluence, resulting in a measured reduction in the sputtering yield by a factor of ~2. These investigations provide a basis for the effect of surface roughness and texturing on a material's sputtering yield. However, little is known about the evolution behavior of material surfaces that have nano- and micro-features and the corresponding sputtering rate during prolonged plasma exposure. We aim to extend this body of knowledge by quantifying the relationship between *surface evolution* and the sputtering yield of surfaces with purposefully-designed surface architecture. The current research broadens our recent findings [6] that nano- and micro-architectures result in reduced sputtering yield, with a focus on the transient nature of *both* surface structure and sputtering yield.

The objective of the present work is to observe the *dynamics* of surface nano- and micro-structure as function of ion fluence, and to examine its corresponding influence on the sputtering yield under prolonged low-energy Ar ion bombardment. We thus focus on the time (ion fluence) dependence of *concurrent* surface morphology and sputtering yield to reveal the interconnection between them. We employ a CVD process to fabricate refractory metal surfaces with unique surface architectures, and present observations of their temporal evolution as function of plasma ion fluence. These changes are the result of sputtering erosion, deposition of ejected surface atoms onto adjacent structures, and atom transport by surface diffusion. A Quartz Crystal Microbalance (QCM) is used to provide quantitative measurements of the sputtering rate at different ion fluence. In the next section, we present a description of the experimental facility and diagnostic tools used to characterize samples as function of plasma exposure fluence. We give an explanation of the CVD process used to fabricate surfaces with uniform microarchitecture. Section 3 presents observations of erosion and deposition of dendritic surfaces that have micropillar architecture under low fluence irradiation. We also examine the self-healing qualities of the surface, and report on the fluence dependent nature of sputtering yield in view of an erosion-ballistic deposition mechanism. In Section 4, the behavior of the observed sputtering yield according to the cumulative fluence exposure is mathematically modeled in a way that characterizes rough surfaces. Lastly, the results are discussed and summarized in Section 5.

2. Experiments

2.1. Plasma source and diagnostics

The UCLA Plasma–materials interaction (UCLA Pi) test facility uses a hollow cathode to generate a plasma that is confined by a series of solenoidal magnets. The plasma terminates approximately 2 m downstream of the cathode on a negatively biased material sample at a spot size of approximately 1 cm diameter. For the result reported herein, an argon plasma was used. The target surface can be differentially biased to provide normally incident ion bombardment at a desired ion energy. Tests were performed at a working pressure of 8×10^{-5} Torr, calibrated for argon. This results in a mean free path of the ions on the order of meters, which exceeds the scale of the plasma sheath, and ensures normal incidence of the plasma relative to the sample surface. Matlock et al. present a detailed description of the Pi facility and the plasma operating conditions in references [14,15]. The Pi facility is equipped with multiple diagnostics for measuring the properties of the plasma and sputtered atoms. An extensive description of the Pi facility in-situ and ex-situ diagnostics is provided in reference [16], so only a brief description of the QCM measurement system and techniques used in this study is given here.

The QCM (setup shown in Fig. 1) provides time-resolved sputtering rate of the target sample. As incoming sputtered atoms get deposited on the surface of the sensor, the resonance frequency of the quartz crystal decreases. In turn, this correlates with the collected mass from the target sample, and hence the number of sputtered atoms from the target area. This measurement allows the differential sputtering rate at a given location to be measured over a period of collection time. Polar measurements are obtained by mounting the QCM to a rotary stage with a moment arm of 25.4 ± 0.2 cm. The sputtering distribution is assumed to be axisymmetric because of the cylindrical nature of the incident plasma column, which allows the differential sputtering rate, measured in $[ng\,cm^{-2}\,s^{-1}]$, to be considered a function of the polar angle only. The measurements in this study concern the *overall* sputtering rate of the entire surface. Therefore, the orientation of surface features