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RESEARCH DURING 1982

(1) In a set of companion papers LePoire, et al. and Melcher, et al. have reported measurements of sputtering yields from solid SO_2 impacted by high energy ions as part of a study of a sputtering mechanism which may be important on Io. The high energy sputtering process investigated is distinct from low energy sputtering phenomenon which can be understood in terms of a billiard ball-like model. At high (MeV) energies, magnetospheric particles can erode condensed gases, such as SO_2, at rates orders of magnitude faster than expected on the basis of our knowledge of low energy sputtering. Yields as high as ~7000 SO_2 molecules per incident ion were observed. ["Sputtering of SO_2 by High Energy Ions" by D. J. LePoire, B. H. Cooper, C. L. Melcher, and T. A. Tombrello, Radiation Effects, in press; and "Erosion of Frozen Sulfur Dioxide by Ion Bombardment: Applications to Io" by C. L. Melcher, D. J. LePoire, B. H. Cooper, and T. A. Tombrello, Geophys. Res. Letters 9 (1982) 1151-1154.]

(2) Cooper and Tombrello have recently concluded a study of the erosion of frozen water ice by fast atomic particles, for a variety of ions and incident energies. These results bear on the understanding of the sputtering of icy satellite surfaces by energetic magnetospheric particles. ["Enhanced Erosion of Frozen H_2O Films by High Energy ^{19}F Ions" by B. H. Cooper and T. A. Tombrello, submitted to Radiation Effects.]

(3) In response to the above experiments, and to the general need for describing sputtering yields for dielectric materials (including frozen volatiles) bombarded by fast ions, Watson and Tombrello have developed a model which predicts both sputtering yields and the mean energy of ejected particles,

for arbitrary fast (MeV) incident beams. The model contains no adjustable parameters, and agrees well with both the H_2O and SO_2 sputtering yield data, as well as with similar data on a wide variety of other dielectric targets. It is anticipated that the approach of Watson and Tombrello will find utility in addressing questions of high energy sputtering in both the Jovian and Saturnian systems. ["A Modified Lattice Potential Model of Enhanced Ion Erosion" by C. C. Watson and T. A. Tombrello, submitted to Radiation Effects.]

(4) We have investigated the diffusion of Xe in olivine, a major mineral in meteorites and lunar samples. Xe ions were implanted into single-crystal synthetic-forsterite targets; depth profiles were measured by alpha particle backscattering before and after annealing the samples. The fraction of Xe retained following annealing was strongly dependent on the implantation dose. Retention was 100% at a dose of 3×10^{15} Xe ions/cm^2 and was less at lower doses, for example there was a > 50% loss at 1×10^{14} Xe ions/cm^2. Taking the diffusion coefficient at this dose as a lower limit the maximum activation energy necessary for Xe retention for 10^7 y in a 10 mm layer was calculated as a function of temperature.

In addition to the above work, a number of other investigations of technological interest arose partly as by-products of our work on planetary applications of sputtering. These included:

(a) "Sputtering of Silicon and Its Compounds in the Electronic Stopping Region" by Y. Qiu, J. E. Griffith, W. J. Meng, and T. A. Tombrello, submitted to Radiation Effects. Fast ion sputtering yields of the dielectrics silicon dioxide and silicon nitride were determined. The Watson and Tombrello model, which was successful in correlating sputtering data on frozen volatiles is also able to predict yields in these non-volatile materials.

(b) "Sputtering of UF_4 by High Energy Heavy Ions" by C. K. Meins, J. E. Griffith, Y. Qiu, M. H. Mendenhall, L. E. Seiberling, and T. A. Tombrello, submitted to Radiation Effects. The Watson-Tombrello model is also successful here. The UF_4 experiments are important because use of special radiation techniques involving uranium allowed an energy spectrum measurement of the sputtered particles; the mean energy of a sputtered atom is very low (< 1 eV) in this material, a fact which has important implications for sputtering of satellites, where a typical atomic escape energy may exceed 1 eV.

(c) "A New Technique for Measuring Sputtering Yields at High Energies" by Y. Qiu, J. E. Griffith, and T. A. Tombrello, submitted to Nuclear Instruments and Methods. It was discovered in the course of trying to measure very small amounts of sputtered material that high-sensitivity analysis of a broad range of materials was made possible by the detection of forward Rutherford-scattered ions. We have called this new technique Rutherford Forward Scattering (RFS).

(d) "Heavy Ion Rutherford Backscattering as a Thin Film Analysis Technique" by M. R. Weller and T. A. Tombrello, submitted to Nuclear Instruments and Methods. This is another analysis technique developed in our studies of sputtering.

(e) "Adhesion Enhancement from High Energy Ion Irradiation" by M. H. Mendenhall, Y. Qiu, and T. A. Tombrello, submitted to Science, and "Enhanced Adhesion from High Energy Ion Irradiation" by B. T. Werner, T. Vreeland, Jr., M. H. Mendenhall, Y. Qiu, and T. A. Tombrello, presented at the Materials Research Society Annual Meeting, Boston, November 1-4, 1982. In the case of our thin film SO_2 sputtering experiments it was noticed that a sulfur residue remained firmly stuck to the backing material after high energy (MeV) irradiation. Further investigation revealed that this ion beam enhanced adhesion effect was a general result of bombarding thin films with energetic particles. It has proven possible in this way to produce strong mechanical and ohmic contacts between materials which are otherwise notoriously difficult to bond (such as Au and Ag on Si). The technological applications of this technique are potentially substantial, and accordingly a patent application has been submitted on behalf of NASA, NSF, and Caltech.

APPENDIX

The following pages contain the abstracts of work completed during the current grant period.

SPUTTERING OF SO_2 BY HIGH ENERGY IONS†

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ABSTRACT

Sputtering yields of solid SO_2 by high energy ions were measured in order to study the mechanism for sputtering dielectrics with ions in the electronic stopping power region. The incident ions were helium and fluorine with energies ranging from 1.5 MeV to 25 MeV. Yields as high as 7000 SO_2 molecules/incident F ion were measured; the 1.5 MeV ^4He beam had a sputtering yield of 50. The data are compared to yield measurements made on UF_4 and H_2O targets. There is a striking similarity in the yield as a function of the incident F energy for all three targets. The data compare favorably with theoretical yield curves based on a new model for the sputtering which considers the electronic excitations induced in the target by the incident beam. Measurements and calculations of this sort are also useful in understanding processes which occur on the surface of Jupiter's satellite Io, which is covered with SO_2 frost and bombarded by energetic ions trapped in the Jovian magnetosphere.

†Supported in part by the National Aeronautics and Space Administration [NAGW-148 and -202], and the National Science Foundation [PHY79-23638 and CHE-13273].

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EROSION OF FROZEN SULFUR DIOXIDE BY ION BOMBARDMENT: APPLICATIONS TO IO

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Abstract. The erosion of frozen SO_2 due to bombardment by both light and heavy ions (He and F) was measured for bombarding energies of 0.08 to 1.3 MeV/amu. The number of SO_2 molecules ejected from the target per incident ion (i.e., the sputtering yield) was 50 for 1.5 MeV He ions and 7300 for 6 MeV F ions. Ion bombardment followed by heating produced an oxygen/sulfur residue which was much more stable against subsequent ion bombardment than the initial frozen SO_2. The erosion rate of SO_2 frost on Jupiter's moon Io depends strongly on the elemental composition and energy spectra of the magnetospheric ion flux which bombards the surface. The combined effects of ion bombardment and heating which produced residues on our target substrates may also occur on Io from magnetospheric ion bombardment and heating by volcanism. Our experimental results compare favorably with a new model of the sputtering process which considers the energy loss of the incident ion to electronic excitation in the target.

I. INTRODUCTION

The bombardment of planetary surfaces by the solar wind, solar flare particles, and magnetospheric particles has been suggested as a possible mechanism for modification of these surfaces as well as for the generation of coronae (see, e.g., Matson et al., 1974; Haff et al., 1981; Johnson et al., 1981; Watson, 1981). Measurements of erosion rates of frozen H_2O due to charged particle bombardment have previously been carried out (Brown et al., 1980; Cooper, 1982). The innermost Galilean moon Io, however, shows evidence of a different kind of ice on its surface, frozen sulfur dioxide. Volcanic gases appear to condense on the cold surface to form an SO_2 frost or adsorbate (see e.g., Nelson et al., 1980). This paper describes measurements of the erosion (sputtering) of frozen SO_2 due to bombardment by both light and heavy ions (He and F) with energies ranging from 0.08 to 1.3 MeV/amu. Comparison of these data with the theoretical work of Watson and Tombrello (1982a,b) should also provide a reliable basis for calculation of the erosion rate of SO_2 by other ions with other energies.

An additional motive behind this work is to investigate the physical processes involved in sputtering. The widely used sputtering theory of Sigmund (1969) predicts negligible erosion due to incident particles with energies exceeding several keV/amu. In recent years, however, a number of measurements on dielectric materials have shown the existence of an enhanced sputtering effect when insulating targets are bombarded with ions having energies in the 0.1-1.0 MeV/amu range (Biersack and Santner, 1976; Brown et al., 1980; Griffith et al., 1980; Qiu et al., 1982; Dück et al., 1980; Ollerhead et al., 1980). The effect has been observed in diverse materials including CsI, ergosterol, UF_4, KCl, H_2O, Al_2O_3, $LiNbO_3$, Xe, SiO_2, and CaF_2. Frozen SO_2 also shows this effect and the data presented here are of interest for comparison with the previously studied materials to help in deciding which of several proposed models best describes the enhanced high energy sputtering process. Fluorine was chosen for the heavy ion bombardment in the present study to enable direct comparison with sputtering data on other materials, especially UF_4 and H_2O. In addition, fluorine is close in mass and nuclear charge to oxygen which is a major component of the Jovian magnetospheric ion flux.

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0094-8276/82/002L-1328$3.00

II. EXPERIMENTAL PROCEDURE

The sputtering yields (number of SO_2 molecules removed per incident ion) for He and F ions incident on solid SO_2 targets were measured for F bombarding energies ranging from 1.6 to 20 MeV and 1.5 MeV He ions. The frozen SO_2 targets (2000-4000 Å thick) were formed by vapor deposition onto a cold (10°K) substrate. This temperature was maintained by flowing liquid helium through an open cycle transfer line on which the substrate was mounted. The substrate consisted of a thick Be disk onto which a 50 Å Au film had been evaporated. The substrate was located in an ultra-high vacuum chamber with a base pressure of 10^{-9} torr. The apparatus is described in more detail elsewhere (Cooper, 1982; LePoire et al., 1982).

Sputtering yields were obtained by measuring the initial target thickness, bombarding the target with a known dose of energetic ions, and then measuring the final target thickness. Target thicknesses were obtained by backscattering spectrometry of 1.5 MeV $^4He^+$ ions using a silicon surface barrier detector; the F ion dose was calculated from the number of ions scattered by the Au marker. Three methods of calculating the SO_2 thickness from the backscattering spectra were used: the energy shift of the substrate Au peak, the number of S counts, and the number of O counts. The sputtering yields calculated using these methods always agreed to within ± 15%.

Targets deposited at 10°K were non-stoichiometric, with 10-25% more oxygen than expected for pure SO_2. The excess O was reduced to 5-10% by raising the temperature of the target substrate to 30°K during target formation. The backscattering spectra indicated that the targets were free of any other contaminants.

ENHANCED EROSION OF FROZEN H_2O FILMS BY HIGH ENERGY ^{19}F IONS†

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ABSTRACT

We have measured sputtering yields of H_2O films with ^{19}F ions at bombarding energies from 1.6 to 25 MeV. The sputtering yield was found to be very sensitive to the incident charge state of the beam, and insensitive to the thickness of the ice film for thicknesses ranging from approximately 20 to 80 $\times$ 10^{16} H_2O/cm^2. The yield was independent of the target substrate temperature from 10 to 60°K and independent of the F beam current density from < 1 to approximately 5.5 particle nanoamps/mm^2. The modified lattice potential model is in good agreement with the observed sputtering yields.

†Supported in part by grants from NASA [NAGW-202 and -148], the NSF [PHY79-23638 and CHE81-13272], and the Caltech President's Fund.

RETENTION OF ION-IMPLANTED-XENON IN OLIVINE:

DEPENDENCE ON IMPLANTATION DOSE

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ABSTRACT

The diffusion of Xe in olivine, a major mineral in both meteorites and lunar samples, was studied. Xe ions were implanted at 200 keV into single-crystal synthetic-forsterite targets and the depth profiles were measured by alpha particle backscattering before and after annealing for 1 hour at temperatures up to 1500°C. The fraction of implanted Xe retained following annealing was strongly dependent on the implantation dose. Maximum retention of 100% occurred for an implantation dose of 3×10^{15} Xe ions/cm^2. Retention was less at lower doses, with $\geq 50\%$ loss at 1×10^{14} Xe ions/cm^2. Taking the diffusion coefficient at this dose as a lower limit, the minimum activation energy necessary for Xe retention in a 10 μm layer for 10^7 years was calculated as a function of metamorphic temperature.

Submitted to: GEOCHIMICA ET COSMOCHIMICA ACTA

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A MODIFIED LATTICE POTENTIAL MODEL OF ENHANCED ION EROSION*

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ABSTRACT

We develop a new, quantitative model describing the enhanced erosion of dielectric materials by ions in the electronic stopping energy regime. This model is based on the concept that the effective intermolecular lattice potential is transiently but significantly modified in some restricted region about the primary ion's path, due to intense electronic excitation. The acceleration of the nuclei in this modified lattice potential transfers appreciable energy to the molecules. Sputtering yields are calculated without the introduction of adjustable parameters or arbitrary normalization, and the results compare quite favorably with the available data. Sputtering induced by both heavy and light ions, on both refractory dielectric materials and condensed volatiles may be treated. The model calculations reproduce observed yields ranging over three orders of magnitude. There are possible applications of the model to related radiation effects, including track registration and the so-called soft failure of solid state computer devices.

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Submitted to Radiation Effects

BAP-30

ORIGINAL PAGE IS
OF POOR QUALITY

SPUTTERING OF SILICON AND ITS COMPOUNDS IN THE ELECTRONIC STOPPING REGION†

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ABSTRACT

We have sputtered silicon, silicon dioxide, and silicon nitride with chlorine ions at 5 MeV and 20 MeV. While the yield from the silicon target was unmeasurably low, the insulating compounds exhibited the enhanced yields observed in other insulating targets. The yield follows the electronic stopping power and seems to be independent of the target's thermal properties. Some of the data suggest that the enhanced sputtering mechanism may be active in extremely thin films (≥ 3 monolayers).

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SPUTTERING OF UF_4 BY HIGH ENERGY HEAVY IONS*

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ABSTRACT

The sputtering of UF_4 targets by energetic beams of ^{16}O, ^{19}F, and ^{35}Cl ions has been investigated for beam energies in the range 0.12 to 1.5 MeV/amu. The sputtering yields, which follow the same trend as the electronic part of the projectile energy loss in the material, are observed to have a strong dependence on the charge state of the incident ions. Data have been taken both in transmission and reflection (0° and 180° to the incident beam direction, respectively). Energy spectra of the neutral sputtered particles have been obtained for 5 MeV ^{19}F ions and for 13 MeV ^{35}Cl ions; in both cases the spectrum has a Maxwellian form. The data obtained are compared with several models of the high energy sputtering process.

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A NEW TECHNIQUE FOR MEASURING SPUTTERING YIELDS AT HIGH ENERGIES*

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ABSTRACT

The use of thin, self-supporting carbon catcher foils allows one to measure absolute sputtering yields in a broad range of materials with high sensitivity. Analyzing the foils with Rutherford forward scattering, we have measured sputtered Al, Si and P surface densities down to $5 \times 10^{13}/cm^2$ with uncertainties of about 20%.

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Submitted to Nuclear Instruments & Methods

HEAVY ION RUTHERFORD BACKSCATTERING AS A THIN FILM ANALYSIS TECHNIQUE*

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ABSTRACT

Rutherford backscattering with heavy ions is an extremely useful technique for the analysis of thin films. It retains the many advantages of alpha backscattering, but provides greater sensitivity and better mass resolution than this more common technique. In addition, many of the disadvantages of heavy ion use are not important for this particular application. Areal densities of surface atoms on light substrates below 10^{13} atoms/cm^2 have been routinely determined.

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BAP-29

ADHESION ENHANCEMENT FROM HIGH ENERGY ION IRRADIATION*

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ABSTRACT

We have recently discovered that the adhesion of thin films to various substrates can be greatly improved by bombarding the films with high energy heavy ion beams. The optimal beam energies lie near the peak of the electronic stopping power for the incident ion (typically 0.5 MeV/amu). Doses range from $1 \times 10^{13}/\mathrm{cm}^2$ to $1 \times 10^{16}/\mathrm{cm}^2$ with hard materials requiring high doses of heavier ions.

Submitted for publication in Science

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ENHANCED ADHESION FROM HIGH ENERGY ION IRRADIATION*

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ABSTRACT

We have found that irradiation of a variety of thin film substrate combinations by MeV/amu heavy ion beams will produce a remarkable enhancement in the adherence of the film. For example, gold films can be firmly attached to soft materials like teflon using a 1 MeV beam of protons (10^{14} cm^{-2}) or helium ions (10^{13} cm^{-2}) and to harder materials like silicon (10^{15} cm^{-2}), quartz (2×10^{15} cm^{-2}) and tungsten (2×10^{14} cm^{-2}) with 0.5 MeV/amu beams of fluorine or chlorine ions. In the case of metal films on semiconductors a low resistance contact results. The mixed layer at the interface is observed to be quite thin ($\lesssim$ 50 Å); for Ag on Si electron diffraction and imaging studies of the interface region reveal the presence of crystalline Ag compounds.

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