Title
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Publication Date
1985-09-01

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

Submitted to Journal of Materials Research

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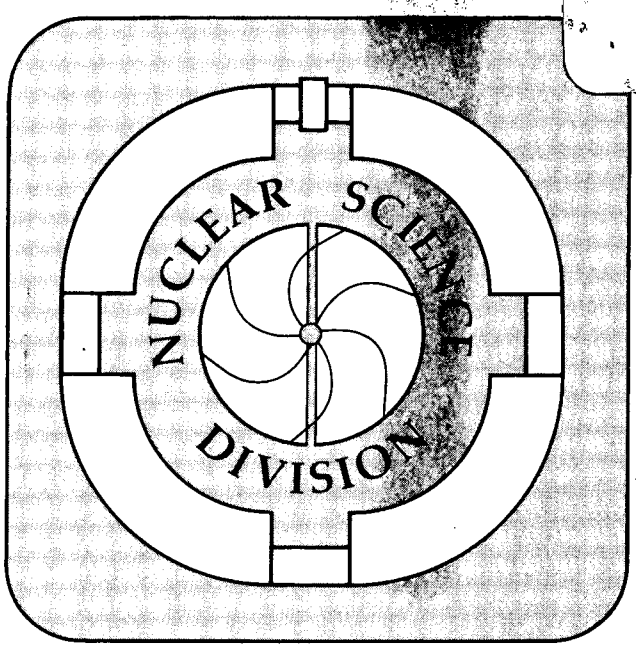
September 1985



Prepared for the U.S. Department of Energy under Contract DE-AC03-76SF00098

DISCLAIMER

LBL-20302

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*This work was supported in part by the Director, Office of Energy Research, Division of Nuclear Physics of the Office of High Energy and Nuclear Physics of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

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Abstract

The ability of heavy ion beams to enhance the adhesion of thin metallic films to substrates has been studied as a function of projectile species. Measurements of the adhesion enhancement of a thin gold film to substrates of tantalum and silicon (with native oxides) have been made for beams of ^{12}C, ^{16}O, ^{28}Si, ^{35}Cl and ^{58}Ni at 2.85 MeV/nucleon. The threshold dose required to pass the Scotch-tape peel test was found for the Au-Ta system to be D^{th} (cm^{-2}) = 10^{17} $(dE/dx)^{-3.0\pm0.2}$ where dE/dx is the electronic stopping power (MeV $mg^{-1}cm^2$) of the ion in Au. For the Au-Si system, $D^{th} = 6 \times 10^{18}$ $(dE/dx)^{-4.1\pm0.3}$. The steep dependence of D^{th} on dE/dx found here is in contrast with an earlier measurement for the Au-Ta system by Tombrello, et al. The adhesion enhancement was observed to decrease with time after the bombardment in a manner suggesting that diffusion of atoms through the gold film is important. The possible importance of small concentrations of extraneous atoms at the interface is discussed.

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I. Introduction

The adhesion of thin metallic films to substrates can be improved by the passage of ionizing radiation through the interface. The resulting adhesion can be quite strong, even for insulating substrates such as polymers and oxides. High velocity, heavily ionizing charged particles such as fluorine and chlorine have been shown to be particularly effective in enhancing adhesion.[1] The number of film-substrate combinations investigated has increased rapidly in the last two years because of the potential applications of this phenomenon and because of interest in the underlying mechanisms.[2-7]

The threshold dose D^{th}(ions/cm^2) required for a thin film to adhere to its substrate rather than to a strip of Scotch tape peeled at 90° is a frequently used indicator of an ion's effectiveness in enhancing adhesion. The variation of D^{th} with projectile species correlates well with the stopping power dE/dx (MeV mg^{-1}cm^2), which is the average energy lost per unit distance of the projectile in the film. A previous measurement of this correlation for a range of projectiles varying from helium to holmium (Z = 67) showed that $D^{th} = 4 \times 10^{14}\ (dE/dx)^{-1.6\pm0.2}$ for a film of 500 Å of gold thermally deposited on a tantalum substrate.[4] The variation of the threshold dose with the stopping power is an important feature to be explained by any model for the enhancement mechanism. However, in developing such models it is necessary to know whether the above result is a general characteristic of the adhesion enhancement process and therefore typical for other film-substrate combinations.

We have recently completed a systematic study of adhesion enhancement induced by different heavy-ion projectiles bombarding thin (500 Å) films of gold on substrates of tantalum and silicon (both having native oxides). Two of the observations made in this work appear to be of particular significance and are reported in this Rapid Communication. A complete description of the experiments and results will be published separately.

II. Experiments and Results

The substrates of cold-formed tantalum sheet were cleaned in hot detergent, rinsed in de-ionized water, etched in a 1:1 solution of nitric acid and water, and rinsed again in water. A rinse in ethanol preceded loading the samples into the evaporator. Before depositing the gold at a pressure of $\lesssim 8 \times 10^{-6}$ Torr, the samples were subjected to a glow discharge cleaning. An Auger analysis[8] indicated that the actual substrate onto which the gold was deposited was a native tantalum oxide of about 40 Å thickness. No elements at the interface other than Au, Ta, and O were indicated by this analysis. The silicon substrates were 1000 Ω cm n-type wafers with a highly polished surface. The cleaning procedure was the same except for the omission of the nitric-acid etch.

The samples were irradiated by beams of ^{12}C, ^{16}O, ^{28}Si, ^{35}Cl and ^{58}Ni at 2.85 MeV/nucleon from the Pelletron accelerator of the Weizmann Institute.[+] Our results are shown in Fig. 1 and may be expressed by $D^{th} = 10^{17}(dE/dx)^{-3.0\pm0.2}$ and $D^{th} = 6 \times 10^{18}\ (dE/dx)^{4.1\pm0.3}$ cm^{-2} for tantalum[++] and silicon, respectively. While our results also are well described by a power law, the values of the exponents are quite different for silicon and tantalum substrates, and the exponent for the Au-Ta system is different from that reported in refs. 2 and 4.

The second observation we report concerns the time dependence of the adhesion enhancement. By repeating the peel test over a period of time on samples irradiated with different doses, we found that the level of adhesion decreased with time (a point of concern for practical applications). Figure 2 shows the dose required to pass a peel test performed a given number of days after the bombardment. Thus, a Au-Ta sample irradiated with $5 \times 10^{14} cm^{-2}$ chlorine ions passed the test for two days after bombardment, but by the end of five days the level of adhesion had deteriorated beyond the failure point. We found that the rate of decrease in adhesion was more rapid the thinner the Au film, and that the level of adhesion could be restored by a

[+] The irradiations were made at angles of 70°-80° with respect to normal incidence. In a separate series of experiments we determined that D^{th} varies as cos (θ). The values of D^{th} given in this article are the doses that would be required for normal incidence.

[++] A tantalum substrate with a 6000 Å oxide layer gave identical results.

subsequent bombardment. The Au-Si system also showed a time dependence, although not as pronounced as for Au-Ta.

III. Discussion

The threshold dose varies dramatically with stopping power: doubling the energy deposited per ion at the interface reduces the required number of ions by factors of 8 and 16 for our Au-Ta and Au-Si samples, respectively. Thus, the number and (or) strength of adhesive bonds induced by an ion's passage through the interface varies in a highly non-linear way with energy deposition. Most puzzling are the different results obtained here and in ref. 4. A critical examination of the procedures described here and in refs. 2 and 4 suggests that neither dose determination nor the application of the peel test is responsible. Rather, we suspect that the different results arise from the preparation of the surface of the sample, and could depend on the level of other elements such as hydrogen or oxygen (or their compounds) adsorbed on the surface of the substrate. While this inference is speculative, it is clear from Fig. 1 that the relative effectiveness of different ions in producing adhesion is quite sensitive to the particular interface.

The time dependence of the adhesion enhancement is not likely to arise from an intrinsic time dependence of the induced adhesive bond (e.g. a meta-stable bond) since it depends on the thickness of the Au film and otherwise resembles a diffusion-driven process. (The dashed line is of the form $D^{th} = a + b\sqrt{t}$ where a and b are adjusted constants and t is the time after bombardment.) It appears likely that ambient gases such as oxygen or water vapor may diffuse to the interface and thereby weaken the bonds induced as a result of bombardment. Thus, the presence or subsequent introduction of relatively small numbers of extraneous atoms at the interface may provide the explanation both for the large variation in the power-law dependence observed here and in ref. 4. for Au-Ta, and for the observed time dependence. The important influence of small amounts of adsorbed gases on the adhesive properties of a metal-oxide interface has been demonstrated by Pepper in measurements of shear strength performed in UHV conditions,[9] and Baglin has suggested the importance of interface contaminants for ion-induced

adhesion.[10)]

In summary, the dependence of ion-induced adhesion enhancement on the projectile species is shown to be well described by a power-law variation with stopping power, $D^{th} \propto (dE/dx)^{-n}$. The value of the exponent, however, is observed to vary widely from one film-substrate combination to the next, and it appears to depend on the specific manner in which the substrate is prepared. Our observation of an enhancement that deteriorates with time indicates that atoms diffusing to the interface after bombardment affect the ion-induced adhesion. Both observations are consistent with the conjecture that the mechanism for adhesion enhancement in these systems is influenced by relatively small concentrations of extraneous or "contaminant" atoms at the interface.

Acknowledgements

R. G. Stokstad is pleased to acknowledge the support of an Erna and Jacob Michael visiting Professorship during his stay at the Weizmann Institute. We wish to thank Dr. R. Brener for performing the Auger analyses, and Drs. Thomas Tombrello and J. E. E. Baglin for several helpful comments and stimulating discussions.

This work was supported in part by the Director, Office of Energy Research, Division of Nuclear Physics of the Office of High Energy and Nuclear Physics of the U. S. Department of Energy under Contract DE-AC03-76SF00098.

References

1. J. E. Griffith, Y. Qiu, and T. Tombrello, Nucl. Inst. and Methods, *198* (1982) 607.

2. M. Mendenhall, Ph.D. thesis, Calif. Inst. Technology (1983), unpublished.

3. S. Jacobson, B. Jonsson, and B. Sundeqvist, Thin Solid Films *107* (1983) 89.

4. T. Tombrello, Mat. Res. Soc. Symp. Proc. *25* (1984) 173.

5. I. V. Mitchell, J. S. Williams, P. Smith, and R. G. Elliman, Appl. Phys. Lett. *44* (1984) 193.

6. J. E. E. Baglin, G. J. Clark, and J. Bøttiger, Mat. Res. Soc. Symp. Proc. *25* (1984) 179.

7. A. E. Berkowitz, R. E. Benenson, R. L. Fleischer, L. Wielunski, and W. A. Lanford, Nucl. Inst. Meth. *B7/8* (1985) 877.

8. R. Brener, private communication.

9. S. V. Pepper, J. Appl. Phys. *50* (1979) 8062; J. Appl. Phys. *47* (1976) 801.

10. J. E. E. Baglin, Proc. Mat. Res. Soc. *47* (1985).

11. L. C. Northcliffe and R. F. Schilling, Nucl. Data Tables 7, Academic Press, N.Y. (1970).

Figure Captions

Fig. 1. The threshold dose for the Scotch-tape test for different bombarding ions (at normal incidence) for a 500 Å gold film on a tantalum oxide substrate and for a 250 Å gold film on a silicon (plus native oxide) substrate. In order of increasing dE/dx values,[11] the open squares correspond to beams of ^{12}C, ^{16}O, ^{28}Si, ^{35}Cl and ^{58}Ni, all at 2.85 MeV/nucleon. Note the difference in both magnitude and slope of the results obtained here and in ref. 4 for Au-Ta systems. The straight lines correspond to $(dE/dx)^{-1.6}$ (the dashed line), $(dE/dx)^{-3.0}$ for Au-Ta and $(dE/dx)^{-4.1}$ for Au-Si (solid lines).

Fig. 2. The threshold dose required for an irradiated spot on a 600 Å Au-Ta sample to pass the Scotch-tape test if the test is applied a period of time after the bombardment. The increased dose required as a function of time after the bombardment corresponds to a loss of adhesion with time.

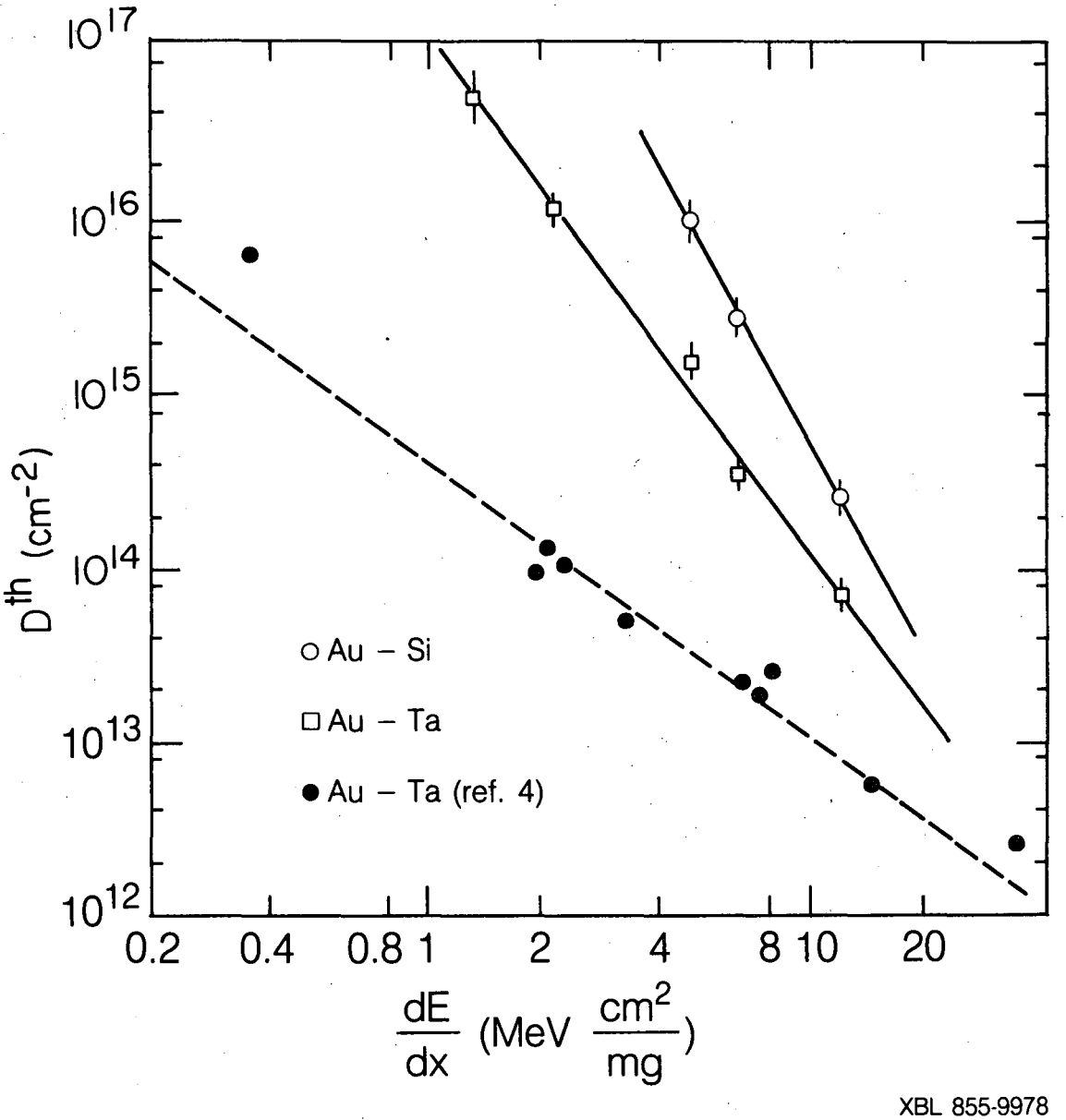

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Fig. 1.

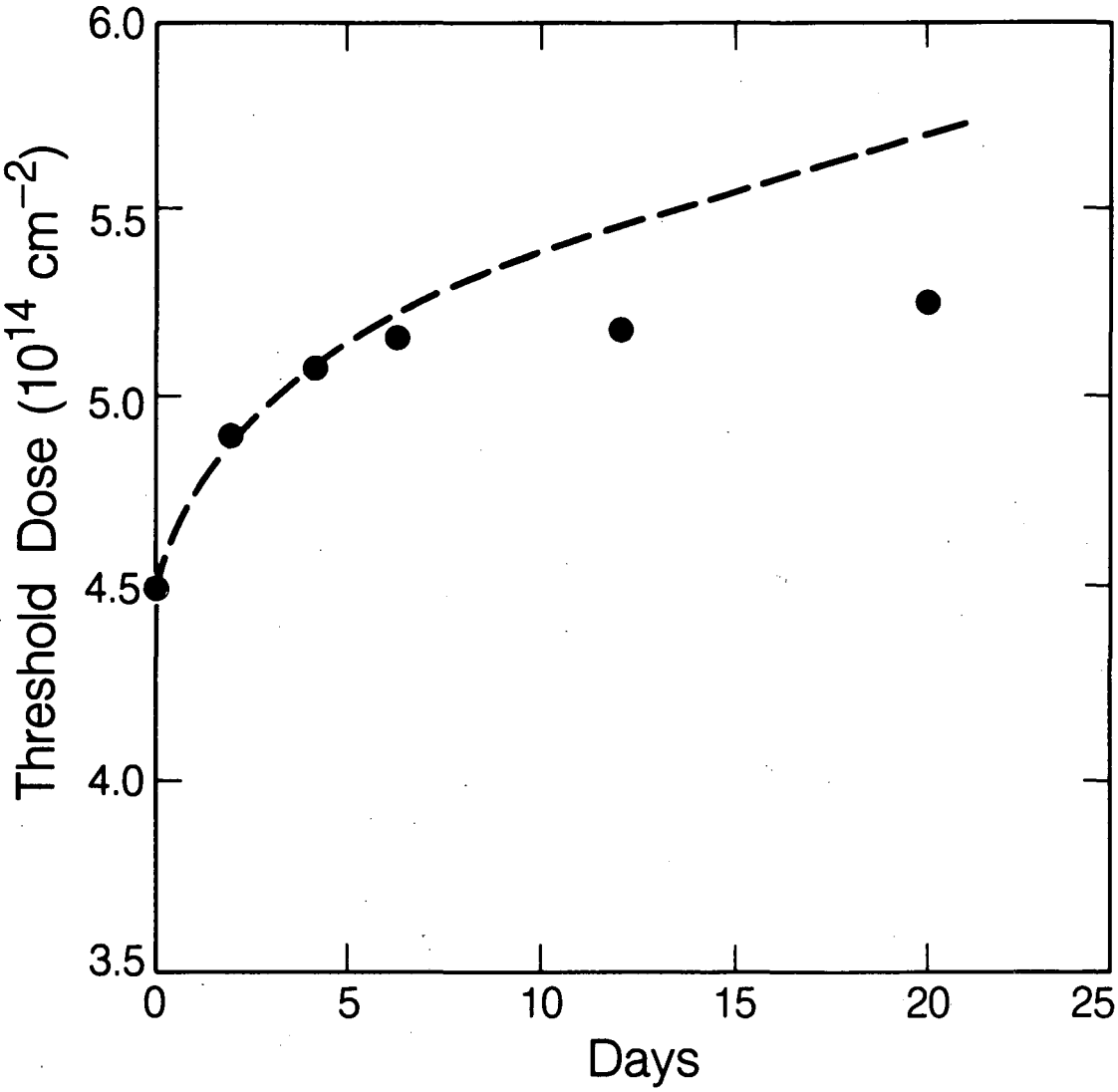

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Fig. 2.

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