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PARTICLE EMISSION BY FISSION

by

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*Work performed under the auspices of the U. S. Atomic Energy Commission.

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Abstract

We have investigated the sputtering by fission fragments of Cu, Al, and Nb doped with 0.1 at. % U^{235} and bombarded with thermal neutrons. This was primarily a search for micron size particles emitted from the surfaces of the doped sources. Kaminsky et al., have observed large particles (several microns in size) emitted from the surface of samples bombarded by 14 MeV neutrons, with a maximum knock-on energy of 600 keV in Nb. The fission energy deposition of 200 MeV should be more than adequate to yield any observable effects that might be present at 600 keV. The concentration of fission events was 10 times the concentration of knock-ons in the Kaminsky experiments. With scanning electron microprobe and energy dispersive x-ray analyses of our sputtering collectors both before and after irradiation, we found no sputtered particles larger than one detection limit of 0.1 μ .

Introduction

Recently, Kaminsky et al.(1) studied sputtering of silicon, niobium and aluminum by 14 MeV neutrons. In addition to the usual sputtered atoms, large aggregations (up to 10^{11} atoms) of material were found on the collectors. These aggregates were verified by a number of techniques including visual observation. Such a behavior is beyond understanding in terms of current atomic collision concepts(2). As is generally the case, this stimulation has generated scientific inquiry and the experiments described in this paper fall into this category.

Indeed the sputtering yield observed from this particle emission is sufficiently great in terms of erosion and plasma poisoning to question feasibility of proposed fusion reactors. Thus, in addition to a strong scientific motivation, there is a strong technological motivation to further investigate this problem.

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The original motivation for this experimentation was to study the effect of energy on particle emission. The inability of the usual atomic collision theory to explain particle emission led to the suggestion that deposition of a large amount of energy near the surface could possibly eject particles by another mechanism(3). Furthermore, it is clear from the experience of thermal reactors that particle emission of the intensity observed by Kaminsky would result in measurable erosion in fuel cladding and in reactor components. A threshold energy for particle emission greater than that attainable by a fission neutron could explain the absence of this effect. Consequently, in these experiments, samples were doped with 0.1 at. % U^{235} and bombarded with thermal neutrons. This in turn caused fission with the attendant release of 167 MeV divided between the two fission fragments.

Experimental Procedure

High purity Nb, Cu and Al were doped with 0.1 at. % U^{235} . The alloys were prepared by arc melting in an argon atmosphere. The weights of the alloy buttons (about 30 gm.) agreed with the sums of weights of the components to within a milligram, indicating little if any loss of constituent metals during melting. The alloy buttons were flipped over and remelted three times to insure proper mixing of the constituents. The buttons were rolled to a 1/16 in. plate and two disks, 1/2 in. in diameter, were spark machined from each plate. Both sides of these disks were then polished in the standard metallographic way, i.e. through silica carbide papers and alumina polishing wheels. The final mechanical polish was done with 0.5 μ alumina.

It is worth noting that in the case of niobium the polishing resulted in plastic flow of the surface. This flowed surface was periodically removed with a chemical polish. One each of the Al and Cu disks were annealed at 500°C to remove the plastic deformation. All of the samples were then electropolished. Very smooth bright surfaces were obtained for the Al and Cu. The Nb, however, was oxidized by the electropolish. The Nb sources were then mechanically repolished with 1 μ diamond paste. One Nb source was additionally polished with 0.25 μ diamond paste. In the case of the more highly polished source, the flowed material was periodically removed. Whereas in the other sample, the flowed material remained. The grain size in the as rolled plates were determined by etching. The grain size was very large in all these metals; on the order of cms. Following the anneal of the Al and Cu, the grain size was considerably reduced.

The collector plates were high purity (99.999%) Al cylinders, 1/2 in. in diameter and 1/4 in. high. These were mechanically polished and electropolished in the same manner as the Al sources. On each collector surface a cross hairs was scratched, dividing the surface into quadrants for identification purposes. Each collector was then examined in the scanning electron microscope (SEM). The samples were so smooth that focussing would have been difficult without the scratches. However, a few SiC particles were found imbedded in the collector surfaces along with an occasional deep pit. The SiC particles were identified by the energy dispersive micro x-ray analysis attachment to the SEM. A unique pit formation was used in each collector as an identification marker and photographs of these regions were made at magnification of 100, 1000 and 10,000. The surface smoothness of both collectors and sources were judged in this manner to be approximately 0.1 μ .

For bombardment, the sources were assembled in an Al tube with a collector adjacent to each face and separated from the source by a 0.02 in. washer. This assembly, shown schematically in Fig. 1 was then placed in an aluminum can, evacuated to 10^{-5} Torr and sealed off. The bombardment was performed in liquid helium (for cryopumping) in the CP-5 Thermal Flux Cryogenic Facility to a dose of 4×10^{17} thermal neutrons. It is estimated, however, that as result of poor thermal contact and nuclear heating, the source-collector assembly reached a temperature between 50 and 100K.

Results

As might be expected the sources were highly radioactive. The collectors had some radioactivity due to the collected fission fragments, but relatively speaking they were inactive. Following SEM examination the fission fragment content of the collectors was determined by counting the Zr^{25} decay with a lithium drifted germanium solid state detector. All of the collectors had the same level, $1.5 \pm 0.1 \times 10^{11} Zr^{95}$ atoms/cm² which indicated 3×10^{12} fission fragments/cm². These results are consistent with the fission yield determined from the neutron dose the fusion cross section, and the range of fission fragments in the sources. It is thus possible to conclude that there were 2×10^{16} fissions/cm³ (4×10^{16} fragments), and this compared to 3×10^{15} primary knock-ons in the Kaminsky experiment.

The microscopic examination of the collectors in SEM failed to show even one particle larger than our detection limit of 0.1 μ that was emitted from the source. Aside from the collectors adjacent to the Al sources, which received a less rigorous examination, about 75% of the collector areas were examined at

1000X and 25% at 10,000X. A sample of before and after irradiation pictures are shown in Fig. 2. A few new particles were found on the surface (see Fig. 3), but energy dispersive x-ray analysis showed that they were not source material. These small particles were all identified as either dirt or Al (or lower atomic number). The Al particles probably originated from the machined spacer washers. Indeed the appearance of these particles demonstrated the ability to detect small particles.

In summary, it has been demonstrated that in our samples, despite a large number of fission fragments, depositing internally large amounts of energy, there was no emission of particles exceeding 0.1μ in size. It would appear reasonable, considering Kaminsky's results and ours, that surface roughness plays an important role in particle emission. Currently, experiments are under way to investigate this factor. Atomic sputtering of niobium was determined by Rutherford back-scattering techniques. The results showing 1.5×10^4 sputtered niobium atoms/cm² are rather tentative as only one collector was examined. It can thus be concluded that 50 niobium atoms are sputtered per emerging fission fragment.

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References

1. M. Kaminsky and S. K. Das, this conference.
2. M. T. Robinson, this conference.
3. M. Guinan, this conference.

List of Figures

Fig. 1. Schematic of sputtering source and collector assembly.

Fig. 2. Sample SEM photographs of collector adjacent to annealed Cu source at 10,000X before (B) and after (A) irradiation.

Fig. 3. SEM photographs of same collector as in Fig. 2 at 1000X before (B) and after (A) irradiation. Note small particles on A (light spots just above pit) which were identified as Al (or lower atomic number).

NEUTRON SPUTTERING EXPERIMENT