Sputtering of Nb by 12-MeV Protons*

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Sputtering yields for niobium bombarded by 12-MeV protons have been measured using a technique involving prior activation of the target material. Values obtained for backward and forward sputtering are $S_B = (1.6 \pm 0.1) \times 10^{-4}$ and $S_F = (4.7 \pm 0.3) \times 10^{-4}$, respectively. These values, expected to be similar to fast-neutron sputtering yields, are consistent with most recently published fast-neutron data.

The phenomenon of sputtering has received increased attention recently, largely because of its significance in the choice of wall materials for thermonuclear reactors. In particular, accurate values are required for sputtering rates of possible wall materials by fast (14-MeV) neutrons, produced in the D-T fusion reaction. Recent measurements[1-4] of sputtering yields of niobium by fast neutrons have differed by four orders of magnitude. Kaminsky, Peavey, and Das[1,2] have reported sputtering yields as high as $S = 0.25 \pm 0.10$ Nb atom per neutron, including a deposit of micron-sized "chunks." Harling *et al.*[3] obtained values of S ranging from 1.1×10^{-5} to 1.3×10^{-3} Nb atom per neutron, and argued in favor of the lower values due to possible contamination of their collector surfaces. Though they found no evidence for chunk emission, this phenomenon is strongly dependent on the surface finish of the Nb samples. Thus, its absence in their experiment may not be in disagreement with the results of Kaminsky, Peavey, and Das.[1,2] The null results of Jenkins *et al.*[4] imply sputtering yields $< 10^{-4}$ atom per neutron. Reference 3 lists references to earlier measurements of neutron sputtering yields.

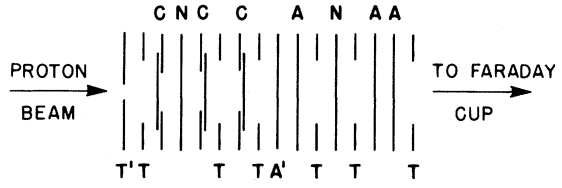
The expected flux of fast neutrons in a thermonuclear reaction is much greater ($\sim 4 \times 10^{14}$ neutrons/cm^2 sec) than the highest flux of 14-MeV neutrons currently available ($\sim 1 \times 10^{12}$ neutrons/cm^2 sec).[5] It has been suggested[5] that radiation damage by fast neutrons might be more conveniently studied in simulation experiments using high-energy protons, for which much higher beam intensities are routinely available. Experiments by Mitchell, Logan, and Echer[6] have indicated that radiation damage by 16-MeV protons is indeed very similar to that produced by 14-MeV neutrons. Other experiments[7,8] have sputtered niobium with 16-MeV protons, but observed neither sputtered material nor chunks, and deduced an upper limit of 10^{-4} Nb atom per proton.

In an attempt to resolve these differences, we have measured the sputtering of niobium by 12-MeV protons using a new experimental technique.[9] The material to be sputtered is first activated by nuclear reactions induced by light-ion beams from an accelerator. The bombarding energy is chosen so that the reaction cross section remains approximately constant as the beam energy is degraded within the target, producing uniform activation throughout the region which contributes to sputtering. The activated target is then sputtered, and the sputtered material is collected on surrounding foils. The collector foils are analyzed by a γ-ray spectrometer, and the quantity of sputtered material is determined from a measurement of the yield of γ rays having energies characteristic of the original target activity. This method has two significant advantages: It is totally insensitive to traces of the target material in the collector foils, since only the previously activated material is detected; and the total sputtering yield can be extracted in a single measurement, without scanning, since the detector is sensitive to all the active material simultaneously. Measured values of sputtering yields obtained with this technique have been compared[9] to values obtained by other techniques, using both activated and unactivated targets, for sputtering by low-energy heavy ions, where the sputtering yields are much higher. The values obtained in all cases were consistent, indicating that the sputtering rate is not greatly affected by the displacement of atoms during activation, and that the active atoms are sputtered at effectively the same rate as the host material.

In the present experiment, the target used during activation consisted of five Nb foils pressed together in a single stack. The target material was cold-rolled Nb foil (supplied by Hamilton Metal Science, Lancaster, Pa.), having a thickness of 2.5 μm and a surface roughness of 0.15 μm. The target stack was irradiated by a beam

of 17-MeV α particles, producing ^{96}Tc, with a half-life of 4.3 days, by the reaction ^{93}Nb$(\alpha,n)^{96}$Tc. The beam was swept magnetically across the targets in a two-dimensional pattern in order to produce uniform activation over a 1.3-cm-diam area. Activation by α particles was used in the present experiment in order to avoid confusion with activation caused by the 12-MeV protons during sputtering. The irradiation time was 21 h, for an integrated flux of 2×10^{16} α particles, producing a concentration of about 10^{-8} active atom per target atom in the active area of the Nb foils. Identifying the foils by numbers from 1 to 5, in the order seen by the beam, foils 1 and 5 were discarded in order to eliminate nonuniformities which might be produced at the entrance and exit surfaces. A 252-μg section about 3×3 mm^2 was cut from the center region of foil 2 to act as a calibration standard for the ^{96}Tc activity of the target foils. Foils 3 and 4 were sputtered.

Details of the target assembly used during sputtering are shown in Fig. 1. There were two independent stacks, separated by a thin aluminum foil, each stack containing one activated Nb foil. The collector foils in the first stack were carbon; those in the second stack were 99.999% pure aluminum foil (supplied by Ventron Corporation). In each case, collector foils were placed both upstream and downstream of the Nb target, and a third identical foil was included to act as a control sample. The target assembly was mounted in an all-metal vacuum system, which operated at $\sim 10^{-8}$ Torr during the sputtering. It was isolated from the accelerator vacuum system by a 30-cm-long, in-line liquid-nitrogen cold trap.



	COMPONENT	MATERIAL	THICKNESS	HOLE DIAM
T′	Aperture	Tantalum	0.5 mm	3 mm
T	Spacer	Tantalum	0.25 mm	13 mm
C	Collector	C foil on Al frame	23 μg/cm^2 0.5 mm	—— 8 mm
N	Target	Niobium	2.5 μm	——
A′	Shield	Aluminum	6 μm	——
A	Collector	Aluminum	25 μm	——

FIG. 1. Schematic diagram of target arrangement used for the sputtering. The entire stack was clamped together, with a total thickness of about 4 mm.

The target assembly was bombarded by a beam of 12-MeV protons, the maximum energy available from the U. S. Office of Naval Research–California Institute of Technology tandem accelerator. This energy is somewhat lower than the suggested optimum energy of 16 MeV,[5] but the damage-energy spectrum in Nb does not change greatly in this range of incident proton energies.[5] A significant fraction of primary interactions proceeds through nonelastic processes whose cross sections vary only slowly for incident beam energies above 10 MeV. The energy lost by the beam prior to striking the second Nb foil was 300 keV, so that the sputtering yield on the aluminum collector foils was expected to be virtually the same as that on the carbon foils.

The beam current was integrated at a Faraday cup 35 cm beyond the target. Both the target and Faraday cup were biased at +600 V, to suppress electron emission, and magnetic suppression was used between the target and Faraday cup. Transmitted beam currents were 350–400 nA, target stack currents were less than 0.1 nA, and the beam spot area was estimated to be 1–2 mm^2 by visual inspection of the radiation damage in the carbon control sample. The integrated beam was 2.5×10^{-2} C, or 1.6×10^{17} protons, corresponding to a fluence of $(0.8-1.6)\times10^{19}$ protons/cm^2. The sputtered material was collected within an area of a few square millimeters in the center regions of the collectors; thus the geometry was well defined for γ-ray analysis.

The activity on the foils was analyzed using two shielded 73-cm^3 Ge(Li) detectors, each having a resolution of about 2.2 keV at the γ-ray energies of interest. The samples were mounted on the front surface of the detector housing in order to maximize counting efficiency and provide a reproducible detection geometry.

Figure 2 shows portions of representative γ-ray energy spectra, where the principal ^{96}Tc transition γ-ray peaks at 778, 812, and 850 keV are labeled. The upper spectrum was obtained in 300 sec, using the calibration source taken from Nb foil 2, 102 h after the end of the sputtering run. The three peaks are prominent, and were used to establish the γ-ray yield per microgram of Nb, with the same geometry and detector efficiency used in analyzing the collector foils. The lower spectrum was taken with the downstream carbon collector foil as source, for a counting time of 4.71×10^4 sec (about 13 h), starting 88 h after the sputtering was terminated. In addition to ^{96}Tc γ rays, peaks present at 803

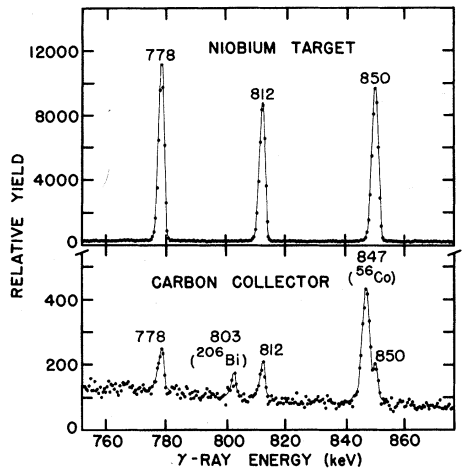


FIG. 2. Portions of γ-ray energy spectra obtained from a section of activated Nb foil (top) and from the downstream carbon collector foil (bottom).

keV (^{206}Bi, $\tau_{1/2}=6.2$ d) and 847 keV (^{56}Co, $\tau_{1/2}=77$ d) were produced by (p,n) reactions on ^{206}Pb and ^{56}Fe impurities, respectively, in the collector foils during sputtering. The yield of sputtered material was determined from the yield of 778- and 812-keV γ rays, since the 850-keV peak could not be reliably separated from the 847-keV ^{56}Co peak. The observed activity was not produced by the reaction ^{96}Mo$(p,n)^{96}$Tc during the 12-MeV proton bombardment, which might have occurred if the collector foils contained traces of Mo. This was established by the absence of activity from other naturally occurring isotopes of Mo and the absence of ^{96}Tc activity on the control foils.

A summary of the results is presented in Table I. Spectra were obtained at several different times, as indicated in the table. The ^{96}Tc activity was most clearly seen in spectra taken 3 or 4 days after the sputtering run. Earlier spectra, especially from the downstream collector foils, contained a very high background of 7-h activity from ^{93m}Mo, produced during sputtering by the reaction ^{93}Nb(p,n); later spectra suffered from the natural decay of ^{96}Tc activity. Values obtained using the two separate peaks at 778 and 812 keV, and from spectra taken at different times, are generally in good agreement. The consistency of values obtained at different times indicates that sputtered material was not lost in handling. The assumption that the presence of activated material on the collector foils was due to sputtering by the beam, rather than by some other process, is supported by the absence of activity on the control foils, and by the correlation of yield with beam direction, i.e., the upstream and downstream yields differ by a factor of 3, but the values obtained using different collector materials are in good agreement. The mean values are $S_B=(1.6\pm0.1)\times10^{-4}$ for backward sputtering, and $S_F=(4.7\pm0.3)\times10^{-4}$ for forward sputtering.

The present values are the most precise reported to date. No evidence for chunk emission was observed in an examination of one of the aluminum collector foils with a scanning electron microscope. However, prior irradiation by α par-

TABLE I. Sputtering yields of niobium by 12-MeV protons.

Collector	E_γ (keV)	Measured values of S (in units of 10^{-4} atoms per proton)[a] 26 h	36 h	61 h	88 h	105 h	Weighted mean	
Upstream C	778	1.6 ± 0.3		1.8 ± 0.3		1.0 ± 0.3	1.6 ± 0.1	$S_B=1.6\pm0.1$
	812	1.9 ± 0.4		1.8 ± 0.4		2.0 ± 0.3		
Upstream Al	778			1.2 ± 0.5		0.8 ± 0.5	1.5 ± 0.3	
	812			1.4 ± 0.6		2.6 ± 0.6		
Downstream C	778		5.1 ± 0.9		4.6 ± 0.5		5.3 ± 0.3	$S_F=4.7\pm0.3$
	812		8.8 ± 1.0		5.3 ± 0.6			
Downstream Al	778				2.9 ± 0.6		3.4 ± 0.5	
	812				4.3 ± 0.8			

[a]Column headings indicate elapsed time (in hours) from end of sputtering run to start of spectrum accumulation.

ticles may affect the probability of chunk emission during proton bombardment. Moreover, in view of the thin targets and relatively smooth surface finish, the present values are not inconsistent with later results of Kaminsky,[10] who reported that the yield of chunks decreases greatly as the surface finish is improved. The sputtering yields for high-energy protons measured in the present experiment are more than an order of magnitude lower than the atomic deposit measured by Kaminsky, Peavey, and Das,[1,2] and are consistent with the range of values obtained in recent fast-neutron measurements by Harling *et al.*[3]

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COMMENTS

Photon-Photon Scattering Contribution to the Anomalous Magnetic Moment of the Muon*

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A careful and systematic recomputation of the photon-photon scattering contribution to the muon magnetic-moment anomaly is made. The result is $\Delta a_{\mathrm{ph-ph}} = (21.32 \pm 0.05)(\alpha/\pi)^3$ leading to the theoretical value $a_\mu{}^{\mathrm{th}} = (1165\,918 \pm 10) \times 10^{-9}$.

It has been known[1] for several years that the photon-photon scattering contribution dominates the sixth-order muon magnetic-moment anomaly. The computed results[1-4] obtained, however, disagree with each other well outside of their assigned 91% confidence levels:

$$\frac{\Delta a_{\mathrm{ph-ph}}}{(\alpha/\pi)^3} = \begin{cases} 18.4 \pm 1.1, & \text{Ref. 1,} \\ 20.77 \pm 0.43, & \text{Ref. 2,} \\ 19.76 \pm 0.16, & \text{Ref. 3,} \\ 19.79 \pm 0.16, & \text{Ref. 4.} \end{cases} \tag{1}$$