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NEUTRON SPUTTERING YIELDS FROM Nb, Au AND Co[†]

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ABSTRACT

High energy neutron sputtering yields have been determined for Nb, Au and Co. Two experiments have been conducted. In the first, graphite catcher foils were used, and in the second, polished Si wafers. From data collected in the first experiment the neutron sputtering yields of Nb, Au and Co were determined to be in the range of 10^{-5} to 10^{-4} particles per incident neutron. Examination of the collector foils from the second experiment did not show any evidence of large particle ejection from the Nb targets.

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The value of the sputtering yield (atoms/incident neutron $\equiv S_n$) of Nb due to high energy neutron impact under ultra-high vacuum conditions has been a subject of some controversy over the past two years. While neutron sputtering ratios have been measured by various research groups over a much longer period of time, the results agree rather poorly. However, attention here will be drawn only to the more recently available data since they will illustrate the disagreement which still persists.

Kaminsky, Peavy and Das[1], hereafter referred to as KPD, using a rotating target 14 MeV neutron source, reported sputtering of large particles of material containing 10^{11} - 10^{12} atoms/particle from cold-rolled Nb specimens with a 3-10 μm grain size and an average surface microfinish of about 5 μm . From their data they estimate $S_n = 0.25 \pm 0.10$, a value which included the fractional "atomic" layer yield value of $S_n = (8.7 \pm 3.0) \times 10^{-3}$. However, these initial crude estimates of the total yield have recently been revised and corrected values of $S_n \lesssim (2-15) \times 10^{-4}$ reported[2]. Using the same neutron source, Harling et al.[3], hereafter referred to as HTBR, found no evidence for neutron-induced large particle emission from cold-worked Nb specimens (one of which was prepared and furnished by M. Kaminsky) with a surface roughness of 1-5 μm , and reported total forward sputtering yield values of $\sim (1-60) \times 10^{-5}$. Employing a neutron source based on the $^9\text{Be}(d,n)$ reaction, which produces neutrons with energies broadly peaked about 15 MeV and characterized by a deposited-energy/neutron comparable to that of a monoenergetic 14 MeV source[4], Jenkins, et al.[5] determined yield values of $S_n \lesssim 10^{-4}$ for both single crystalline and discharge machined polycrystalline Nb, and a search for large particle ejection from the targets produced negative results.

In an attempt to understand the reasons for these differences in experimental data, we have conducted two additional neutron irradiation experiments. In each we have used the same neutron source as in our previous experiment[5], a complete description of which will soon be available[6]. Also, during these two separate irradiation experiments the conditions were the same as those reported previously[5], except for the greater neutron fluences reported here. The sample stacks consisted of a series of 2.5 cm diameter discs separated by 0.25 mm-thick annular spacers. Monitor foils of (2.5 cm diameter) ^{59}Co and Ni (natural), to be used as means of estimating neutron fluence, were placed periodically in each of the two sample arrays. In both experiments the targets and collector samples were 0.25 mm-thick and the detector foils 0.12 mm. Each target had two collector samples, one in front and one in back with respect to the neutron beam origin, to serve as collectors for sputtered materials.

In one experiment graphite was used as the collector material because discs of high purity could be obtained and because of the lack of activation products which would interfere with analysis. The purpose of this experiment was to establish reliable numbers for sputtering yields. The relatively rough surface structure of the graphite collector samples precluded microscopic detection of large particles, but had multi-atom particles of the size and number (revised) reported by Kaminsky and associates[2] been sputtered, the yields would have been much higher than those observed. The results obtained are shown in Table 1. The amount of sputtered material was determined by neutron activation analysis. The method is quite sensitive for Au, samples of which were included to

serve as a comparison standard. Nb was determined using a Ge(Li) x-ray detector to count ^{94m}Nb decay after the sensitivity of the method was established using Nb-doped standards which had been activated and counted for predetermined periods of time. Also, the sputtering data from (n,2n) recoil processes are shown in Table I since the information could be useful to others, particularly in attempts to compare data obtained using different neutron sources.

The sample array in the other irradiation contained six cold-worked Nb samples, three of which were supplied by Kaminsky's group at Argonne National Laboratory (ANL) and the remaining three by Harling and associates at Batelle Pacific Northwest Laboratories (PNL). Details of the methods of sample preparation will be presented at this Conference[7,8]. Since this particular experiment was designed primarily to investigate large particle ejection, polished Si wafers identical to those used in our previously reported work[5] with a (111) surface orientation were used as collector foils in order to facilitate examination with the SEM. Dimensions of all target, collector and monitoring foils were the same as those listed above. Prior to mounting in the sample stack, the Nb targets which had been received sealed under high-purity conditions were cleaned by ultra-sonic washing in acetone. This was done in order to remove any loosely adhering material and because it was the standard final cleaning process for targets which we had prepared ourselves. This was the only treatment given these samples at this laboratory prior to irradiation. Following neutron irradiation to fluences $\sim 10^{17}/\text{target}$, the twelve Si collector foils were examined for large particles of Nb using a SEM. Each collector was examined within a 1 cm^2 area which corresponded to that portion of the target subjected to the greatest neutron fluence. A total

of twenty 1 mm^2 sections were selected at random within this area on each of the twelve collectors and scanned with the SEM at 1000X magnification. A few large particles were observed, but x-ray fluorescence spectroscopy revealed their composition not to be Nb, but rather other common dust particulate contaminants which blank specimens showed to be present in the laboratory air. In summary, a total of 240 regions, 1 mm^2 in area, were examined on the twelve Si collectors associated with the six Nb targets supplied by ANL and PNL, and in no case could micron-size particles of Nb be detected. At present, attempts are being made to determine sputtering yields from these targets, but the data are incomplete.

The sputtering yields for Nb shown in Table 1 are an order of magnitude lower than those we previously reported[5]. This is due to the greater neutron fluence achieved here as well as to lower background contamination levels in the graphite collector foils. These values are within the lower range reported by HTBR[3] and about an order of magnitude less than the revised estimates of Kaminsky's group[2]. Because of differences in the neutron spectra employed, the difficulty with contamination, background, and the extremely low levels of material being analyzed, etc., the sputtering yields reported here probably can be considered to be in reasonable agreement with the data of HTBR[3]. Since the yields determined from the graphite collectors for Nb, Au and Co agree well with theoretical predictions, and since the same yield values were obtained from Nb single-crystalline and polycrystalline materials, it is reasonable to assume that in this experiment no large particles were ejected from the Nb targets; therefore, the lack of agreement with yield values reported by the ANL group[2] is understandable. Also, our failure to detect large particles

in the experiment using Si collectors with the Nb samples furnished by PNL and ANL is consistent with the data of HTBR but not with those of KPD. From our experience and the data of other it would appear that large particle emission must be related to surface preparative techniques. Figure 1 illustrates the vast difference in surface structure of typical specimens prepared at ANL and PNL. The ANL specimen surfaces consisted of rather long scratches and grooves with material pile-up frequently observed at points along the grooves. The piled-up material was identified as Nb by x-ray fluorescence analysis in the SEM. Most piled-up matter appeared to be attached to the substrate more firmly than in the example shown in Fig. 1. Since the ANL group have now reported revised estimates of large particle sizes and densities[2] which are generally smaller than those they originally estimated[1], it appears reasonable that quite rough surfaces possibly contain some loosely attached particulates which could detach during the course of the experiment. This supposition is supported by the data of Kaminsky and Das[9] who observed "chunks" of silicon ($\approx 3-5 \text{ cm}^{-2}$) and aluminum ($\approx 10-20 \text{ cm}^{-2}$) on Nb targets facing collector discs of these materials. Since the Si collector was a flat, highly-polished single crystal and the Al collector an annealed specimen, neither would be expected to possess the necessary residual internal stresses required by Guinan's[10] model for large particle emission; and loosely attached particulates on the collector surfaces would appear to be more plausible sources for these large particles of Si and Al. It is quite possible that the controversy concerning neutron sputtering yields and large particle emission is a natural consequence of comparing data which have been gathered from ill-defined, non-reproducible

surfaces. When one attempts to apply concepts from disciplines concerned with the physics of particle-solid interactions or of surfaces in interpreting data gathered from specimens which have been characterized only in terms of their metallurgical properties, conflicting results should be anticipated. At any rate, the data collected at ORNL, PNL, and the revised data from ANL establish that erosion rates of a fusion reactor wall, due to neutron impact, are not of technological significance.

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Table 1. Neutron Sputtering Yields

Target	Yield (atoms/neutrons) $\times 10^{-5}$		Fluence (neutron/ foil) $\times 10^{17}$	(n,2n) Yield (atoms/foil) $\times 10^9$ Forward
	Forward	Back		
Nb-1 ^a	≤ 2.8	≤ 4.2	1.00	5.05
Nb-2 ^b		≤ 3.4	0.92	4.50
Nb-3 ^b	≤ 5.0	≤ 4.7	0.72	3.90
Nb-4 ^c	≤ 5.6	≤ 4.0	0.67	3.25
Nb (average)	≤ 4.5	≤ 4.1		
Nb (theory) ^g	1.2			
Au-2 ^d	2.0	2.8	1.17	7.03
Au-3 ^e	3.5	1.4	0.79	4.53
Au (average)	2.8	2.1		
Au (theory) ^g	2.2			
Co-1 ^f		2.4	1.09	
Co-2 ^f		0.9	1.37	
Co (average)		1.6		
Co (theory) ^g	≈ 2.7			

^a"Single-crystal", 3 large grains comprise 90% of target area, slight HNO₃-HF etch.

^bCold-rolled, spark discharge machined, HNO₃-HF etch to ~ 5 μ m surface roughness.

^cCold-rolled, HNO₃-HF etch to ~ 5 μ m surface roughness.

^dCold-rolled.

^eCold-rolled, electropolished.

^fCold-rolled, acid etch, surface roughness not determined.

Table 1. Neutron Sputtering Yields (Cont.)

^aCalculated using $S = \alpha \Lambda N \langle \sigma \hat{E} \rangle$; see R. Behrisch, Proc. 5th International Conf. on Atomic Collisions in Solids, Amsterdam, 1975; to be published in Nucl Inst. Methods. Co estimation based on extrapolation from Cu damage energies using total cross-section ratio $\sigma(\text{Co})/\sigma(\text{Cu})$. For values of $\langle \sigma \hat{E} \rangle$ see Ref. 4.

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FIGURE CAPTIONS

Fig. 1 Scanning electron photomicrograph of typical sample surface structures supplied by ANL and PNL groups. The 25 μm reference mark is on the micrograph of a PNL surface, and the magnification is the same for both specimens.

Fig. 1