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PARTICLE RELEASE FROM NIOBIUM UNDER 14-MeV NEUTRON IMPACT

by

M. Kaminsky and S. K. Das

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PARTICLE RELEASE FROM NIOBIUM UNDER 14-MeV NEUTRO

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Summary

The particle release from cold-rolled and annealed polycrystalline niobium surfaces under 14-MeV neutron impact to a total dose of 4.6×10^{15} neutrons/cm^2 was investigated in two separate runs at ambient temperature and under ultrahigh and high vacuum conditions, respectively. The type and amount of material released and deposited on a substrate surface was determined independently by four analytic techniques. Surprisingly, there were two types of deposits - one in the form of large chunks, the other a more even layer covering the surface. For the cold-rolled niobium sample with a surface finish of 5 μm the estimated particle release value is $S = 0.25 \pm 0.10$ niobium atoms per incident 14-MeV neutron. A model for the chunk emission is suggested.

Introduction

During the operation of controlled thermonuclear reactors the surfaces of such components as container walls, beam limiters and beam dump walls will be exposed to MeV neutron bombardment. It has been suggested that such neutron bombardment may lead to particle emission and cause both serious wall erosion and plasma contamination (for reviews see Ref. 1—3). Unfortunately, the experimental information available on particle release by MeV-neutron impact on solids is very scarce and contradictory. For example, for monocrystalline gold irradiated with 14-MeV neutrons Garber et al.[4] reported a sputtering yield. $S = 3 \times 10^{-3}$ atoms/neutron, which is five times the upper limit value reported by Keller[5] for polycrystalline gold. However, for iron irradiated with neutrons of a softer energy spectrum (from a fission reactor), Baer et al.[6] reported an even larger yield $S = (5.7 \pm 0.8) \times 10^{-3}$ atoms/neutron. Based on predictions[1,3,7] made for the yield dependence on neutron energy (using rather simple theoretical models) however, one should have expected the yield values to increase with increasing neutron energy (for neutrons in the MeV energy range).

A theoretical estimate of S for niobium irradiated with 14-MeV neutrons by one of the authors (M. K.) yielded $S \approx 6 \times 10^{-5}$ atoms/neutron - a value which was approximately 2 orders of magnitude smaller than the above mentioned ex-

MeV neutron impact in an environment, respectively been reported elsewhere.[8]

Experimenta

The Lawrence Live rotating-target source[10,1] with an ICT accelerator. rate over the entire 4π so source target was about 3 × irradiation run was monito detectors.[12] The drift in recoil detector is thought t was calibrated independent for example,[11,13] by use reaction. The absolute ac of the average dose of the mated to be ±7.5%. The ta train assembly containing mounted in a st. steel hold and were placed inside a v tance between the neutron face of the first target, a approximately 2.5 cm in d was ~1.75 cm. A space of disk from the second targe polycrystalline niobium foi imately2.5 cm and a thick third target, again a mono dimensions similar to thos spaced ~0.28 cm from the of the neutron-source disk the three target disks were

The undoped, optica Si(111) disks (obtained from Corporation) had a purity o first mechanically and then had a surface microfinish o polycrystalline niobium san Materials Research Corpor mechanically polished and had an average surface mic mirror finish with some sc For the second irradiation niobium foil was replaced b foil had been annealed at 1 of approximately 2×10^{-7} polished to an average mic procedure described earlie

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MeV neutron impact in an ultrahigh and a high vacuum environment, respectively. Part of this material has been reported elsewhere.[8, 9]

Experimental Procedures

The Lawrence Livermore Laboratory's rotating-target source[10, 11] was used in conjunction with an ICT accelerator. The total neutron emission rate over the entire 4π solid angle of a fresh neutron source target was about 3×10^{12} neutrons/sec. The irradiation run was monitored with two proton-recoil detectors.[12] The drift in counting efficiency of each recoil detector is thought to be <5%.[13] The total dose was calibrated independently by activation analyses, for example,[11, 13] by use of the $^{93}Nb(n, 2n)^{92}Nb$ reaction. The absolute accuracy in the determination of the average dose of the sample received was estimated to be ±7.5%. The targets were mounted in a train assembly containing 17 targets (each target was mounted in a st. steel holder, held at ground potential) and were placed inside a vacuum chamber. The distance between the neutron source disk and the front face of the first target, a monocrystalline Si(111) disk approximately 2.5 cm in diameter and 0.5 cm thick, was ~1.75 cm. A space of 0.28 cm separated this disk from the second target, a circular, cold-rolled polycrystalline niobium foil with a diameter of approximately 2.5 cm and a thickness of 0.013 cm. The third target, again a monocrystalline Si(111) disk with dimensions similar to those of target No. 1, was spaced ~0.28 cm from the second target. The center of the neutron-source disk and the center of each of the three target disks were on the same axis.

The undoped, optical-grade monocrystalline Si(111) disks (obtained from the Materials Research Corporation) had a purity of 99.999%. They were first mechanically and then chemically polished and had a surface microfinish of ~0.03μm. The cold-rolled polycrystalline niobium sample was obtained from Materials Research Corporation(Marz grade). It was mechanically polished and lightly electropolished[14] and had an average surface microfinish of ~5 μm(a coarse mirror finish with some scratch traces still present). For the second irradiation run this cold-rolled niobium foil was replaced by an annealed one. This foil had been annealed at 1200°C for ~2 h in a vacuum of approximately 2×10^{-7} Torr before it was electropolished to an average microfinish of ~2.0 μm by the procedure described earlier.[14]

The target was evacuated with sorption pumps

...surface was determined independently by four analytic techniques. Surprisingly, there were two types of deposits - one in the form of large chunks, the other a more even layer covering the surface. For the cold-rolled niobium sample with a surface finish of 5 μm the estimated particle release value is S = 0.25 ± 0.10 niobium atoms per incident 14-MeV neutron. A model for the chunk emission is suggested.

Introduction

During the operation of controlled thermonuclear reactors the surfaces of such components as container walls, beam limiters and beam dump walls will be exposed to MeV neutron bombardment. It has been suggested that such neutron bombardment may lead to particle emission and cause both serious wall erosion and plasma contamination (for reviews see Ref. 1—3). Unfortunately, the experimental information available on particle release by MeV-neutron impact on solids is very scarce and contradictory. For example, for monocrystalline gold irradiated with 14-MeV neutrons Garber et al.[4] reported a sputtering yield. S = 3×10^{-3} atoms/neutron, which is five times the upper limit value reported by Keller[5] for polycrystalline gold. However, for iron irradiated with neutrons of a softer energy spectrum (from a fission reactor), Baer et al.[6] reported an even larger yield S = $(5.7 \pm 0.8) \times 10^{-3}$ atoms/neutron. Based on predictions[1,3,7] made for the yield dependence on neutron energy (using rather simple theoretical models) however, one should have expected the yield values to increase with increasing neutron energy (for neutrons in the MeV energy range).

A theoretical estimate of S for niobium irradiated with 14-MeV neutrons by one of the authors (M. K.) yielded S ≈ 6×10^{-5} atoms/neutron - a value which was approximately 2 orders of magnitude smaller than the above mentioned experimental value for gold. More recently Behrisch[7] estimated a yield value for 14-MeV neutron bombarded niobium that was only about a quarter of that of Kaminsky. On the basis of such an estimated value, this author concluded that neutron sputtering would have a negligible effect on wall erosion and plasma contamination during the operation of a fusion reactor. In view of the great discrepancy between the theoretical estimates and the available experimental results, it seemed imperative to conduct new experiments under controlled conditions.

The present experiments were undertaken to provide information on the erosion of surface of cold-rolled and annealed polycrystalline niobium under 14-

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The target was evacuated with sorption pumps to a pressure of 1×10^{-3} Torr. Subsequently, a combination titanium-sublimation-ion pump was used for pumping. At the beginning of the first neutron-irradiation run, the pressure in the chamber was ~1.5×10^{-9} Torr, and it dropped to ~4×10^{-10} Torr towards the end of the ~54 h irradiation run. The pressure at the start of the second ~53.5 h irradiation run was 5×10^{-7} Torr and dropped to 1.5×10^{-7} Torr towards the end of the run. The total 14-MeV neutron dose on both types of niobium foils in the two runs was estimated from the total number of counts of the proton recoil counters (with the appropriate corrections for the relative positions of source, counter, and target, for the effective neutron

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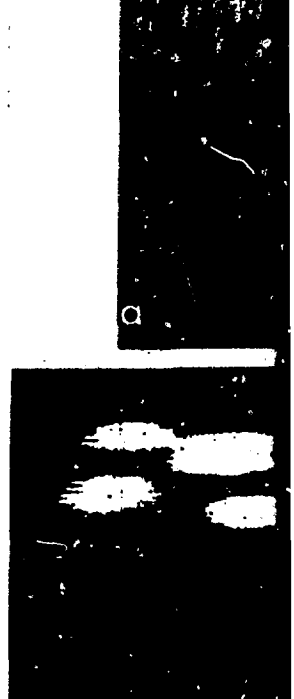
absorption length, and for the neutron flux peaking in the forward direction). The estimated dose was $\sim 4.6 \times 10^{15}$ neutrons/cm^2, and the average dose rate was $\sim 2.4 \times 10^{10}$ neutron cm^{-2} sec^{-1}. During the irradiation the targets were at ambient temperature (estimated to be near room temperature). The irradiated targets were transported under ultrahigh vacuum conditions ($\sim 1 \times 10^{-9}$ Torr) from Lawrence Livermore Laboratory to Argonne National Laboratory for analysis.

To determine the type and amount of the materials released from the surface of one sample (e.g., niobium target No. 2) and deposited on the surface of the one facing it [e.g. Si(111) target No. 1] the analytical techniques used were Rutherford backscattering (using a 750-keV $^4He^+$ ion beam from a 2-MeV Van de Graaff), Auger spectroscopy, ion microprobe microanalysis (an ARL ion microprobe), and scanning electron microscopy (a Cambridge Stereoscan Mark IIA) in conjunction with an energy dispersive x-ray spectrometer. The amount of deposited material could be estimated quantitatively by using calibration standards prepared by vapor deposition. For the niobium deposits, the detection sensitivities [expressed as fractions of a monolayer (ML) of niobium deposited on silicon substrate] of three of the analytical techniques (in the order of decreasing values) were Rutherford backscattering (~0.0005ML), ion microprobe (~0.001 ML), and Auger spectroscopy (~0.01 ML). The scanning electron microscope together with an energy dispersive x-ray spectrometer was also used to identify the deposits. A metallograph was used for visual inspection of the irradiated targets. To test contamination build-up during the period of storage, unirradiated targets were kept in their irradiation train assembly in the ultrahigh vacuum-storage chamber. No significant contamination buildup could be detected.

Results

An examination of the surface of Si(111) target No. 1 which faced the cold-rolled polycrystalline niobium target revealed the surprising result that the niobium deposits appeared in two forms. One form covered the substrate surface as a fractional atomic layer with an estimated "average" coverage of ~0.026 ML. The other form appeared as chunks of various irregular shapes as illustrated in Fig. 1. The optical micrograph in Fig. 1(a) shows some of the chunks. The secondary-ion ($^{93}Nb^+$) micrograph shown in Fig. 1(b) was obtained for the same area as in Fig. 1(a); it confirmed that the



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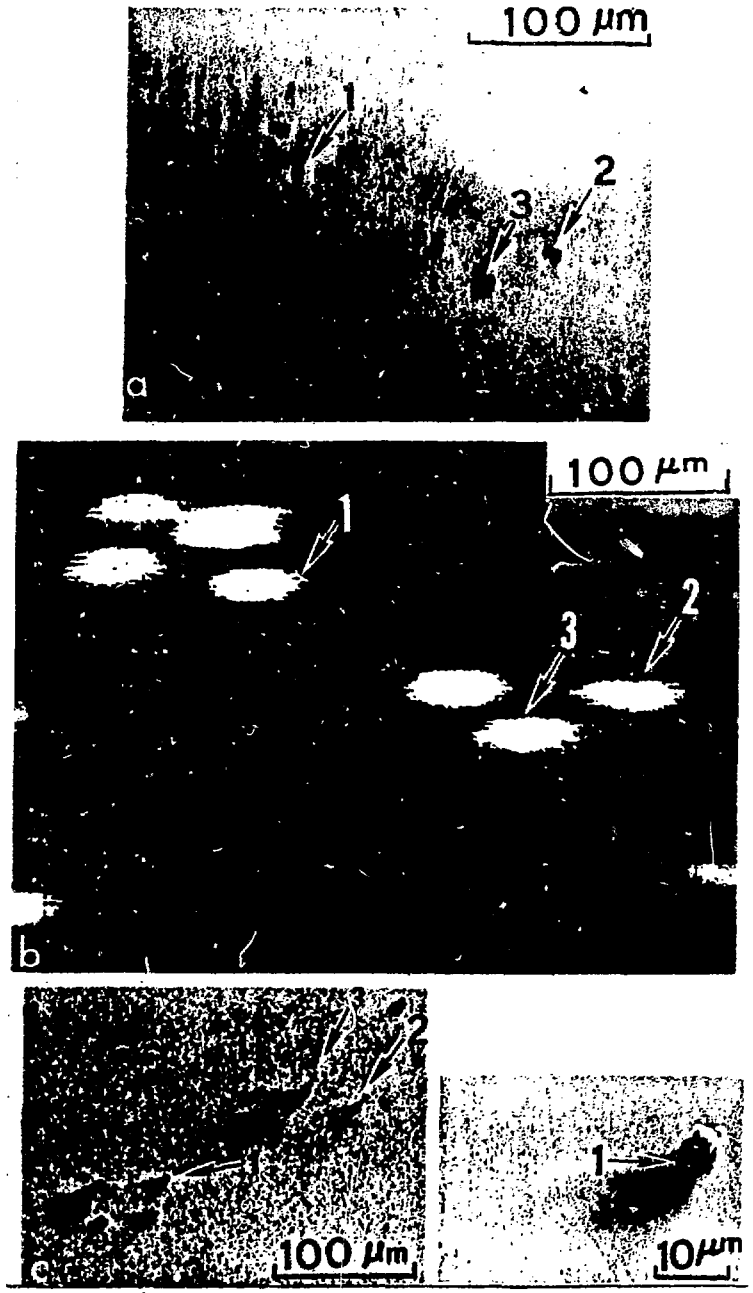

Fig. 1. Niobium deposited on a silicon (111) substrate when a niobium target at ambient temperature (estimated to be near room temperature) was irradiated with 14-MeV neutrons to a total dose of 4.6×10^{15} neutrons/cm^2. (a) Optical micrograph of niobium chunks deposited on a Si(111) surface. (b) Secondary ion ($^{93}Nb^+$) micrograph of the same area as in fig. 1(a). (c) Scanning electron micrograph (backscattered electron image) of the same area as in fig. 1(a), showing the size and shape of the deposited chunks of Nb. (d) An enlarged view of a roughly spherical chunk No. 1 shown in fig. 1(c).

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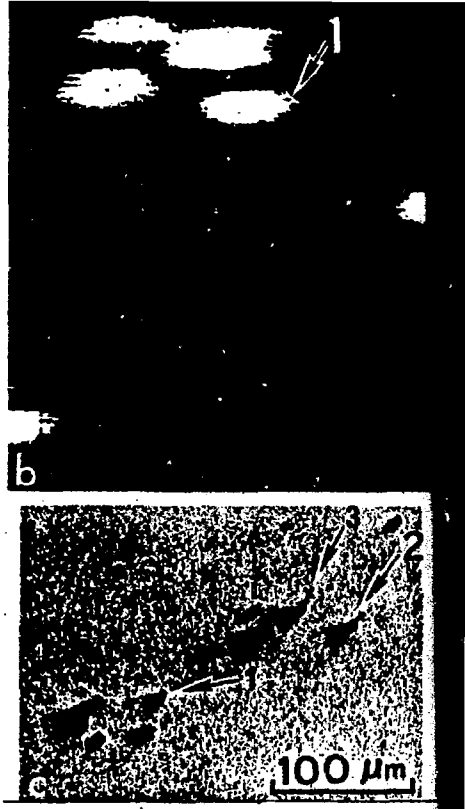
has been only crudely estimated from the enlarged scanning electron micrographs. Since the chunks were irregular in shape, the "average" diameter of

ple (e.g., niobium target No. 2) and deposited on the surface of the one facing it [e.g. Si(111) target No. 1] the analytical techniques used were Rutherford backscattering (using a 750-keV $^4He^+$ ion beam from a 2-MeV Van de Graaff), Auger spectroscopy, ion microprobe microanalysis (an ARL ion microprobe), and scanning electron microscopy (a Cambridge Stereoscan Mark IIA) in conjunction with an energy dispersive x-ray spectrometer. The amount of deposited material could be estimated quantitatively by using calibration standards prepared by vapor deposition. For the niobium deposits, the detection sensitivities [expressed as fractions of a monolayer (ML) of niobium deposited on silicon substrate] of three of the analytical techniques (in the order of decreasing values) were Rutherford backscattering (~0.0005ML), ion microprobe (~0.001 ML), and Auger spectroscopy (~0.01 ML). The scanning electron microscope together with an energy dispersive x-ray spectrometer was also used to identify the deposits. A metallograph was used for visual inspection of the irradiated targets. To test contamination build-up during the period of storage, unirradiated targets were kept in their irradiation train assembly in the ultrahigh vacuum-storage chamber. No significant contamination buildup could be detected.

Results

An examination of the surface of Si(111) target No. 1 which faced the cold-rolled polycrystalline niobium target revealed the surprising result that the niobium deposits appeared in two forms. One form covered the substrate surface as a fractional atomic layer with an estimated "average" coverage of ~0.026 ML. The other form appeared as chunks of various irregular shapes as illustrated in Fig. 1. The optical micrograph in Fig. 1(a) shows some of the chunks. The secondary-ion ($^{93}Nb^+$) micrograph shown in Fig. 1(b) was obtained for the same area as in Fig. 1(a); it confirmed that the chunks were niobium. The larger fraction of the chunks (approximately 4/5ths of the total deposits) appears to be roughly "spherical" in shape (e.g., chunk No. 1 in Figs. 1c and 1d), while the smaller fraction (approximately 1/5th of the total deposits) appears to be more "cylindrical" in shape (e.g., chunk No. 3 in Fig. 1c). Some of the more "cylindrically" shaped chunks have their long axes nearly normal to the surface of the Si(111) substrate. Many of the chunks show microprotrusions in certain regions. So far the size distribution of the chunks



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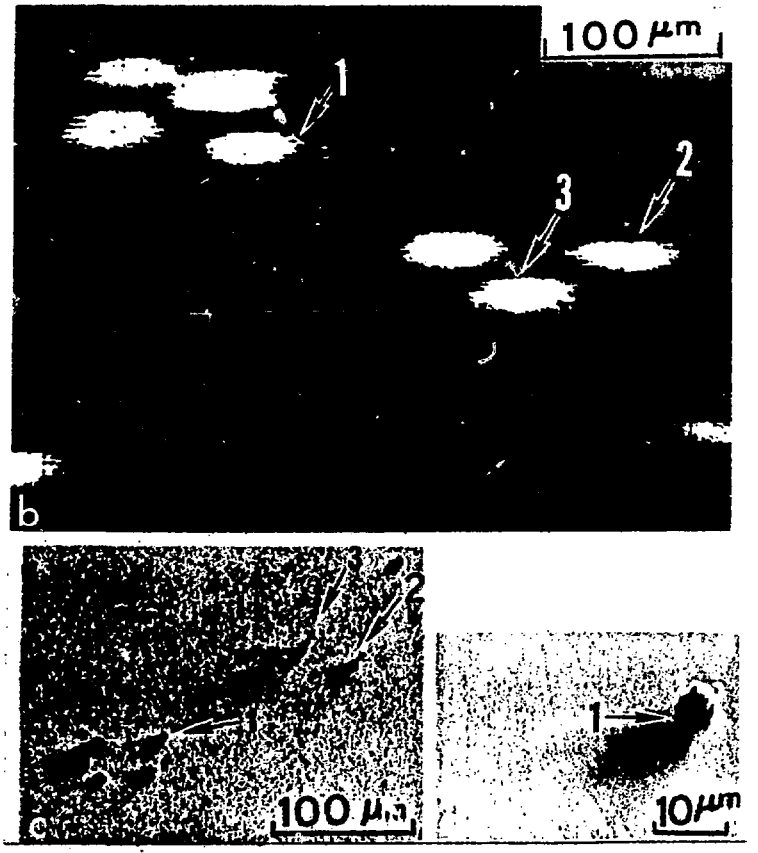

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has been only crudely estimated from the enlarged scanning electron micrographs. Since the chunks were irregular in shape, the "average" diameter of either a roughly "spherical" or a "cylindrically"-shaped chunk was defined as the diameter of a circle whose area is equal to that of the projection of the chunk onto the surface plane.

For the "cylindrically"-shaped chunks, the "average" diameter ranged from about 0.5 to 5 μm, while the lengths of the "cylinders" ranged from about 3 to 15 μm. A majority of these chunks have a volume of $\sim 4 \times 10^{11}$ cm^3, which corresponds to $\sim 2 \times 10^{12}$ atoms/chunk.

The "spherically" shaped chunks had "average" diameters ranging from approximately 1 to 5 μm. A majority of these chunks have a volume of about 1×10^{-11} cm^3, which corresponds to about 5×10^{11} atoms per chunk. For both types of chunks, the distribution over the irradiated substrate surface area is nonuniform. Sometimes both types of deposits (fractional atomic layer and chunks) appear clustered on certain substrate areas. For example, they appear as wide streaks (width >40μm, length >400μm) which are not associated with the microstructure of the substrate surface (e.g. microscratches). They also appear in irregularly shaped patches. The estimates given above are very crude and work is in progress to improve the accuracy of these estimates.

An investigation of the Si(111) surface of target No. 3 which faced the backside of the polycrystalline niobium target No. 2 revealed niobium chunk deposits similar to those reported above for target No. 1. The surface of Si(111) target No. 1 which faced the stainless steel flange revealed chunk deposits of stainless steel. In the second irradiation run target No. 2 was the annealed polycrystalline niobium foil mentioned earlier, and target No. 3 was an annealed, polycrystalline aluminum foil with a microfinish of ~6 μm. Niobium chunks could be observed on the aluminum target surface facing target No. 2, and aluminum chunk deposits could be observed on the niobium surface (target No. 2) facing the aluminum target No. 3. In the second irradiation run target No. 1 was a highly polished Si(111) target of the same type used in the first run. A preliminary examination of the niobium deposits on the Si target surface facing the annealed polycrystalline niobium target (No. 2) revealed again niobium chunks but their number was smaller by approximately a factor of five than observed for the cold-rolled foil. Again, in addition to the chunk deposits one observes also deposits which cover the substrate surface as a fractional atomic layer for each of the target surfaces mentioned above.

By ion milling the niobium chunks deposited on the Si(111) surface (target No. 1) facing the cold rolled polycrystalline niobium target (No. 2) [first irradiation run] to about 1/10th of their original size by using the ion microprobe with 20-keV O^- ions, it was determined that the chunks consisted of niobium and that they did not contain contaminants to any significant degree. Figures 2(a) - (c) illustrates this ion milling of a silicon surface area which has been most heavily deposited with niobium chunks. The binding of the niobium chunks to the Si(111) substrate appeared to be rather strong since the chunks could not be readily scrapped off mechanically, or

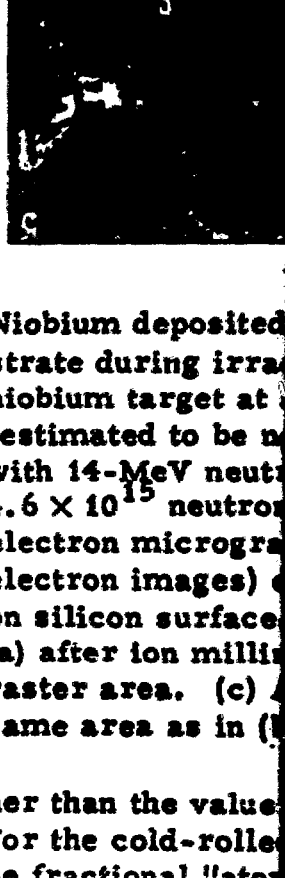

Fig. 2. Niobium deposited
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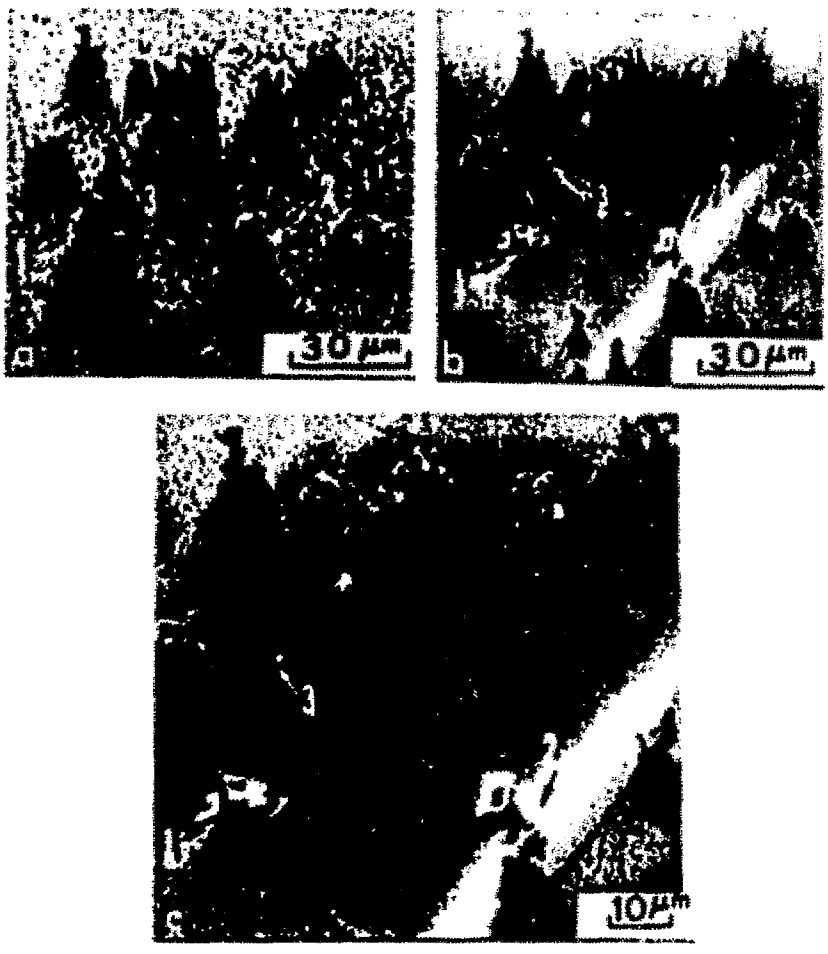

Fig. 2. Niobium deposited on a silicon (111) substrate during irradiation of a cold rolled niobium target at ambient temperature (estimated to be near room temperature) with 14-MeV neutrons to a total dose of 4.6×10^{15} neutrons/cm^2. (a) Scanning electron micrographs (back scattered electron images) of niobium chunks deposited on silicon surface. (b) The same area as in (a) after ion milling with oxygen ions in the raster area. (c) An enlarged view of the same area as in (b).

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~20% higher than the value quoted above for target No. 1. For the cold-rolled polycrystalline niobium sample the fractional "atomic" layer deposit alone gives a yield value of $S=(8.7 \pm 3.0) \times 10^{-3}$ niobium atoms per neutron. Considering both the chunk and the atomic layer deposits a total yield $S=0.25 \pm 0.10$ niobium atoms per neutron is obtained.

Discussion

On the basis of the two types of deposits observed, the authors speculate that target particle emission via momentum transfer from collision cascades as well as via thermal spike "vaporization" will contribute to the fractional "atomic"

patches. The estimates given above are very crude and work is in progress to improve the accuracy of these estimates.

An investigation of the Si(111) surface of target No. 3 which faced the backside of the polycrystalline niobium target No. 2 revealed niobium chunk deposits similar to those reported above for target No. 1. The surface of Si(111) target No. 1 which faced the stainless steel flange revealed chunk deposits of stainless steel. In the second irradiation run target No. 2 was the annealed polycrystalline niobium foil mentioned earlier, and target No. 3 was an annealed, polycrystalline aluminum foil with a microfinish of ~6 μm. Niobium chunks could be observed on the aluminum target surface facing target No. 2, and aluminum chunk deposits could be observed on the niobium surface (target No. 2) facing the aluminum target No. 3. In the second irradiation run target No. 1 was a highly polished Si(111) target of the same type used in the first run. A preliminary examination of the niobium deposits on the Si target surface facing the annealed polycrystalline niobium target (No. 2) revealed again niobium chunks but their number was smaller by approximately a factor of five than observed for the cold-rolled foil. Again, in addition to the chunk deposits one observes also deposits which cover the substrate surface as a fractional atomic layer for each of the target surfaces mentioned above.

By ion milling the niobium chunks deposited on the Si(111) surface (target No. 1) facing the cold rolled polycrystalline niobium target (No. 2) [first irradiation run] to about 1/10th of their original size by using the ion microprobe with 20-keV O^- ions, it was determined that the chunks consisted of niobium and that they did not contain contaminants to any significant degree. Figures 2(a) - (c) illustrates this ion milling of a silicon surface area which has been most heavily deposited with niobium chunks. The binding of the niobium chunks to the Si(111) substrate appeared to be rather strong since the chunks could not be readily scrapped off mechanically, or burned off by the high flux (~3mA/cm^2) of 20-keV oxygen ions.

A crude estimate based on the assumption that the niobium deposits were smeared out evenly over the Si(111) substrate No. 1 (first irradiation run) yielded an equivalent surface-coverage degree of 0.86 ML for all types of niobium deposits. For Si(111) target No. 3 the average surface-coverage degree of the niobium deposit was found to be

Fig. 2. Niobium deposited on a strate during irradiati niobium target at ambi (estimated to be near with 14-MeV neutrons 4.6×10^{15} neutrons/c electron micrographs electron images) of ni on silicon surface. (b (a) after ion milling wi raster area. (c) An e same area as in (b).

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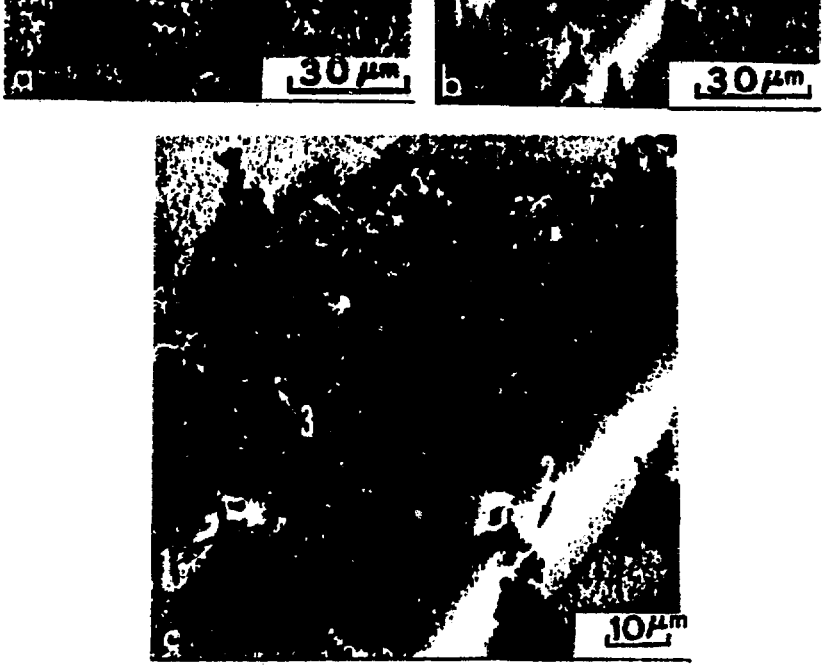

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4 Discussion

On the basis of the two types of deposits observed, the authors speculate that target particle emission via momentum transfer from collision cascades as well as via thermal spike "vaporization" will contribute to the fractional "atomic" layer deposit. (It should be noted that aggregates of smaller than ~200°A diameter [limit of resolution of scanning electron microscope] are counted as being part of the "atomic" layer deposit). The mechanism for the emission of chunks, however, is not clearly understood. The observation that the number of deposited chunks is ~5 times larger for irradiating the cold-rolled niobium surface (microfinish of ~5μm) than the annealed niobium surface suggests

that the amount of stored energy (due to cold working) and the degree of microstructure of the irradiated surface (e.g., microcracks, scratches, hillocks) affects the chunk emission significantly. The energy deposited by a 14-MeV neutron interacting with niobium lattice atoms in the near-surface region via elastic and inelastic collisions (calculated mean and maximum energies for primary knock-on niobium atoms are ~180 and 600 keV, respectively) can lead to localized thermal and ionization (electron) spikes which in turn may cause the generation of shock waves. The interference of such shock waves in a small volume in the near surface region in which the stored energy is very high (due to cold working), may set up stresses large enough to release energy by initiating submicroscopic cracks or by propagating already existing microcracks and cause the emission of chunks from highly stressed and rough surface regions. For example, microprotrusions with only a small fraction of the surface atoms bound to the host lattice atoms may be released by such a process. However, one cannot exclude the possibility that other mechanisms may contribute to or dominate the chunk-ejection process.

The yield $S=0.25 \pm 0.1$ niobium atoms/neutron obtained for cold-rolled polycrystalline niobium with a surface finish of only ~5μm at near room temperature leads to an annual erosion rate of 0.6 ± 0.3 mm/year for a neutron flux of 4×10^{14} neutrons cm^{-2} sec^{-1}. If the erosion rates for a fusion reactor wall at operating temperature are similar to those obtained here, then erosion by neutron impact cannot be neglected, contrary to the conclusion by others.[7]

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