RADIATION SPUTTERING OF SINGLE-CRYSTAL AND POLYCRYSTALLINE COPPER IN THE RADIATION FIELD OF A NUCLEAR REACTOR

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Sputtering of single-crystal and polycrystalline copper targets under bombardment in a reactor has been studied with a fast-neutron flux of 2×10^{12} neutrons/cm^2-sec up to a dose of 10^{19} neutrons/cm^2. The angular distributions of atoms ejected from single crystals show a preferential sputtering in the directions of close packing of atoms in the crystals. The sputtering intensity I and the ejected atom energy E from single crystals at the beginning of irradiation are twice I and E in the case of polycrystals. With increasing dose the difference in the measured values of I and E for single crystals and polycrystals decreases. The results obtained are discussed in the light of the mechanisms of collision focusing and channeling of the displaced atoms in the crystals.

1. It is well known that bombardment of metals by fast neutrons leads to appearance of high-energy displaced atoms which dissipate their energy in atom-atom collision cascades (see, for example, the review by Garber and Fedorenko[1]). When these cascades intersect the boundary between the metal and a vacuum or any other rarefied medium, the atoms which are in the surface layer, having received as the result of a collision an energy above their binding energy in the surface, are ejected from it. This process is essentially similar to the process of sputtering in ion bombardment of metals[2].

Recently it has been shown[3] that in bombardment of a gold single crystal by a collimated, monoenergetic beam of neutrons of energy 14 MeV from a neutron generator, a preferential sputtering of gold atoms is observed in the directions of the closest packing of atoms, which is explained by the focusing of atomic collisions in these directions.

However, the inadequate intensity of the beam from a neutron generator (10^8 neutrons/cm^2-sec) does not permit investigation of the properties of the bombarded target as a function of the dose of radiation. A nuclear reactor is a more powerful source of fast neutrons. However, in experiments with a nuclear reactor it is necessary to reckon with the fact that in a reactor, in addition to fast neutrons, there are present also thermal neutrons and intense γ radiation. In addition, bombardment of materials in the reactor occurs with an isotropic flux.

In the present work we have studied the sputtering of single-crystal and polycrystalline copper in the radiation field of a nuclear reactor as a function of the dose of reactor radiation.

2. The experiment was performed in the vertical channel of the reflector of the VVR-M reactor at the Physics Institute of the Ukrainian Academy of Sciences. The fast-neutron flux at the location of the experiment was 2×10^{12} neutrons/cm^2-sec.

The target being studied was placed in special evacuated glass ampoules (pressure 10^{-6} torr) in such a way that good thermal contact existed between the targets and the walls of the ampoules. Cooling of the ampoules to 60–70°C during the bombardment was accomplished by a flow of air through the channel. The temperature of the targets was measured by a potentiometer method with a copper-constantin thermocouple.

The single-crystal and polycrystalline copper targets studied were disks of area 1 cm^2 and thickness 1 mm. The single-crystal targets were cut by a spark cutter from a single-crystal bar obtained by electron-beam melting followed by repeated zone purification at the E. O. Paton Electric Welding Institute of the Ukrainian Academy of Sciences. The crystallographic orientation of the targets was determined from a back-reflection Laue pattern. The polycrystalline targets were cut from copper of special purity ($< 10^{-4}$ for each of 11 elements). The target surfaces were metallographically finished and then electrochemically polished and vacuum annealed at a temperature close to the recrystallization temperature, for an hour.

The particles ejected from the target during bombardment in the reactor hit a sectionalized hemispherical collector located on one side of the target at a distance of 5 mm, and a screen—on the other side of the target. The collector was made of molybdenum, and the screen of aluminum. Both materials have sputtering coefficients one hundred times smaller than copper and are not activated by thermal neutrons.

During the experiments the collector current was measured by an electrometer circuit with a noise level of 10^{-13} A. The design of the ampoules permitted measurement of the "background" current, which amounts to 10^{-9}–10^{-10} A and apparently arises from ions knocked out of the walls and other parts of the ampoules, which are made of molybdenum glass and molybdenum, which are characterized by low sputtering coefficients. The total collector current was 10^{-8}–10^{-6} A. Both single-crystal and polycrystalline targets were placed simultaneously in each ampoule. The vacuum in the ampoules was maintained during the experiment by means of getters of powdered titanium baked with U^{235} oxide, which were shielded by a mica plate from the measuring elements of the ampoules.

After the experiment the screens with the deposits of sputtered atoms from the bombarded targets were removed from the ampoules. The number of radioactive Cu^{67} atoms in a deposit was measured by means of a PP-8 radiometric apparatus with a scintillation counter. By exposing an x-ray film applied to the screen with the active deposits, autographic pictures were made of the deposits on the screens. The distribution of atoms in the deposits on the screens was determined by photometry of the autographs in an MF-2 microscope.

3. Measurements of the angular distributions of the atoms ejected from the surface of single-crystal and polycrystalline copper targets under the action of fast neutrons showed that atoms are ejected from the (100) surface of a copper single crystal preferentially in the four ⟨110⟩ directions (at an angle of 45° to the normal to the surface of the crystal) and in the central ⟨100⟩ direction (normal to the surface of the crystal) (see Fig. 1, curve 1). The density distribution of particles sputtered from the copper single crystal in the deposit on the screen is similar to the angular distribution of emitted particles measured by means of the sectionalized collector. The autoradiograph of the deposit of sputtered atoms from the copper single crystal (Fig. 2a) reveals a pattern with discrete spots, whose distribution corresponds to the orientation of the low-index directions of the (100) face of the copper crystal bombarded by neutrons. The particles emitted from the polycrystalline targets have an isotopic distribution (Fig. 1, curve 2; Fig. 2b).

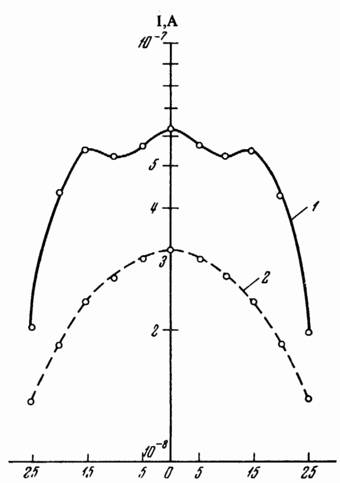

FIG. 1. Angular distributions of atoms ejected from the surface of a single-crystal copper target (curve 1) and a polycrystalline copper target (curve 2) under the action of fast neutrons.

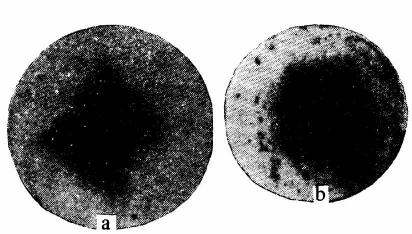

FIG. 2. Autoradiographs of deposits of sputtered atoms from the surface (a) of a single-crystal copper target, and (b) from a polycrystalline copper target.

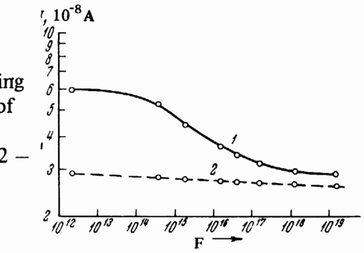

FIG. 3. Intensity of sputtering of copper targets as a function of integrated fast-neutron dose. Curve 1 – single-crystal target, 2 – polycrystalline target.

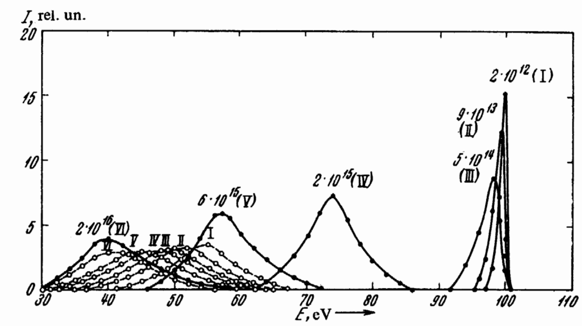

FIG. 4. Spectra of particles emitted from copper targets, as a function of the integrated fast-neutron dose. ● – Single crystal target, ○ – polycrystalline target. I – 2×10^{12} neutrons/cm², II – 9×10^{13} neutrons/cm², III – 5×10^{14} neutrons/cm², IV – 2×10^{15} neutrons/cm², V – 6×10^{15} neutrons/cm², VI – 2×10^{16} neutrons/cm².

The angular distribution of sputtered atoms from single-crystal targets subjected to bombardment with 10^{15}–10^{16} neutrons/cm² reveals no preferential emission of atoms in the directions of close packing and is close to isotropic.

The sputtering intensity of copper single crystals, which at the beginning of the exposure is more than twice the sputtering intensity of polycrystals, decreases with increasing integrated neutron dose up to 10^{16} neutrons/cm² (Fig. 3), remaining, however, above the sputtering intensity of the polycrystalline target.

Measurements of the energy spectra of particles emitted from single-crystal and polycrystalline copper targets also showed that with increasing neutron dose the energy of the particles emitted from the single crystals decreases, reaching the energy of particles emitted from polycrystals (Fig. 4). Furthermore, with increasing bombardment dose the spectrum of emitted particles from single crystals is smeared, while the spectrum of particles emitted from polycrystals is almost unchanged.

4. The measurements made of angular distributions, energy spectra, and intensity of particles sputtered from copper single crystals and polycrystals are direct proof of the existence of collision focusing and channeling[1] of fast displaced atoms in crystals under neutron bombardment in a reactor.

[1]In crystals a portion of the fast particles, both the bombarding particles and those displaced from their lattice sites, falling into open channels bounded by close-packed rows of atoms, move in a space almost free from forces, and for this reason travel anomalously large distances, several times greater than the average range calculated for an isotropic solid body. The term was first introduced by Liebfried [4] in 1963.

The higher yield of sputtered atoms from single crystals in comparison with the yield from polycrystals, the preferential ejection of atoms from single crystals in the directions of close-packed rows of atoms, the high energy value of the emitted particles from an undamaged single crystal (of the order of the energy of focusons in copper under ion bombardment[2]), and also the monoenergetic nature of the emitted particles from undamaged single crystals, are due to collision focusing and channeling of fast displaced atoms in the radiation cascades arising in the copper crystal lattice under neutron bombardment in the reactor. With increasing radiation dosage the scattering and blocking of focusons, crowdions,[2)] and channeled fast displaced atoms by radiation defects produced in the crystal lattice during the bombardment increases, with the result that the difference between the single crystal and the polycrystal is almost imperceptible. In polycrystals, because of the disorientation of crystallites in the surface layer, the presence of significant distortions of the structure, and a number of other factors, the scattering and blocking of focusons, crowdions, and channeled fast displaced atoms are so great as to result in lower sputtering yield of atoms and absence of effects with strong anisotropy which are present in single crystals. Radiation damage of polycrystals with increasing neutron bombardment dose does not have a significant effect on the quantities measured.

[2)]Quasiparticles which, depending on the conditions of their formation, are called focusons (along the chain of atoms in the crystal there occurs a transfer of energy and momentum from the bombarding particle) or crowdions (transport of mass also occurs). The terms were first introduced by Leibfried [5] in 1959.

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