Proposed Reactor Radiation Damage Monitor

One of the perplexing problems in reactor radiation damage studies is that of measuring the fast neutron exposure in such a way that exposures of the same type of material to different reactor neutron spectra can be readily and meaningfully intercompared. The most commonly used method is to measure the reactor neutron spectrum with various threshold detectors (*1*). This method has at least three deficiencies. It is cumbersome. It neglects neutron energies in the most important region, i.e., below an effective threshold energy of about 3.5 Mev (S^{22}), unless measurements are made with fission foils (U^{238}, Np^{237}, Pu^{239}) which are not readily available and are difficult to use. Finally, it is difficult to relate the measured spectrum to the observed radiation damage. Other techniques have been suggested which are based on measurement of the change in property of a reference solid that varies in a known way with irradiation (*2*, *3*). These techniques, however, yield a parameter that is only indirectly related to the radiation damage process in materials other than that of which the monitor is composed. In addition, the response of such monitors is very apt to be temperature dependent. The author believes that the method proposed herein eliminates or reduces the aforementioned difficulties. The method also provides a measurement that is directly related to the atomic displacement process on which radiation damage in solids so critically depends, and shows promise of providing an *in situ* monitor of radiation damage in the components of operating reactors.

The method is based on the following arguments. Since collisions between fast neutrons and the atoms in a solid result in transfer of energy to the struck atoms, it may be expected that some of the atoms residing near the surface of the irradiated material will recoil with sufficient energy to be ejected from the surface. The process is in fact analogous to the phenomenon of sputtering by low energy ions. In both ion sputtering and fast neutron sputtering (which has not yet been experimentally demonstrated) the emission of atoms from the surface results from the displacement of atoms from their normal positions in the solid near the surface (*4*), and is thus closely related to atomic displacement processes that are produced deeper in the bulk solid by fast neutrons or high energy ions. Consequently, if the atoms sputtered by fast neutrons during a reactor exposure could be collected and counted, the sputtering process could provide a radiation damage monitor more closely related to the primary radiation damage process than anything we now have available.

The success of such a monitor will depend on the value of the sputtering ratio, defined here as the number of atoms sputtered per neutron incident on the surface. To obtain an estimate of this ratio, we use the theory of sputtering by high energy ions (>50 kev) developed by Goldman and Simon (*5*). The only modifications needed to adapt this theory to a fission spectrum are the substitution of suitable values for the cross section σ_d for production of a primary displacement, and for the average number $\bar{\nu}$ of secondary displacements per primary. The resulting expression for the sputtering ratio is

$$r = \frac{0.17\bar{\nu}\sigma_d}{\pi R^2 \cos\psi} = \frac{0.17\bar{\nu}}{\pi R^2 \cos\psi}\bar{\sigma}_s\left(1 - \frac{AE_d}{4E_n}\right) \qquad (1)$$

where $\bar{\sigma}_s$ is the average fast neutron scattering cross section for the incident neutron spectrum, E_n is the average fission neutron energy, A is the atomic weight of the irradiated material, E_d is the threshold energy for an atomic displacement, ψ is the angle between the surface normal and the incident flux direction, and R is the distance of closest approach in hard sphere collisions of the moving displaced atoms with like atoms at rest. R is determined by the expression

$$E = \frac{Ze^2 \exp(-R/a')}{R} \qquad (2)$$

where E is the energy of the moving atom, Z is the atomic number, e is the electronic charge, $a' = \alpha a_0/\sqrt{2}\,Z^{1/3}$, a_0 is the Bohr radius = 5.3×10^{-9} cm, and α is an experimentally determined parameter (*6*).

The sputtering ratio of copper will be calculated as an example. In Eq. (2) we choose E to be the average energy, $\bar{E}_p$, of the primary recoils produced by a fission neutron spectrum. Thus

$$\bar{E}_p = \tfrac{1}{2}E_p(\max)/\ln\frac{E_p(\max)}{E_d}$$

$$= 5.6 \times 10^3 \text{ ev}$$

where $E_p(\max) = 92$ kev and $E_d = 25$ ev. van Lint *et al.* (*7*) have found that for copper the best value of α is 1.7. Substituting in Eq. (2) we find $R = 3.7 \times 10^{-9}$ cm. For our purposes, a satisfactory value of $\bar{\nu}$ is given by

$$\bar{\nu} = \frac{\bar{E}_p}{2E_d} = 1.1 \times 10^2$$

To a sufficiently good approximation, $\bar{\sigma}_s = 3 \times 10^{-24}$ cm².

Choosing normal incidence ($\cos\psi = 1$) and substituting the above values in Eq. (1), we find the sputtering ratio to be $r = 1.4 \times 10^{-6}$. Thus for an integrated neutron flux of 10^{18} neutrons/cm², the yield of sputtered copper atoms is 1.4×10^{12} per cm² of Cu surface. If the sputtered atoms are collected on a foil adjacent to or in contact with the emitting surface, their number can be determined by thermal neutron activation. In practice, the monitoring device can consist of a sandwich of alternate layers of target and collector foils. With such an arrangement, an emitting surface area of 100 cm² is not difficult to obtain in a compact device. Thus, in the case considered here, we may expect a total yield of sputtered copper atoms of about 1.4×10^{14}. The saturation activity of Cu^{64} in a thermal flux of 10^{13} neutrons/cm² sec is then about 4×10^3 disintegrations/sec. This activity is quite sufficient for accurate measurement.

The feasibility of the foil sandwich technique for collecting atoms ejected from a surface by recoil has been nicely demonstrated by van Lint *et al.* (*7*), in their measurements of the range of low energy recoil atoms. The sputtering of atoms from a gold foil by 300-kev protons has been used by Thompson (*8*) in an elegant experiment to demonstrate the existence of focussing collisions predicted by Silsbee (*9*). It is also noteworthy that Thompson used thermal neutron activation analysis to detect the sputtered gold atoms. In addition, Thompson's measurements provide a valuable check on the validity of the Goldman-Simon sputtering theory (*5*). Thompson (*8*) reports that the number of gold atoms collected during a total exposure of 3.4×10^{16} protons of 300-kev energy was approximately 10^{13}. This gives a sputtering ratio of 3.1×10^{-4} which is a factor of 3 smaller than the theoretical ratio of 10^{-3}. For

our purpose, the agreement between theory and experiment is adequate. Thus, there seem to be no theoretical or experimental reasons why an exposure monitor based on sputtering should not be successful.

The monitor should be applicable to a wide variety of materials. The important criteria are that the sputtered atoms be detectable with adequate sensitivity by activation analysis and that the collector foils produce no activity that will interfere with detection of the collected atoms. Separate neutron activation will often be unnecessary since the reactor thermal neutron flux at the position of the monitor will provide activation. In addition, for long exposures ($>10^{20}$ neutrons/cm^2) of reactor structural materials, it may be possible to measure the number of sputtered atoms by microchemical or microphysical analysis.

One other possible use of the sputtering technique should be noted. Since sputtering is related to the displacement process, the measurement of sputtering ratios for fast neutrons and energetic charged particles may provide an accessible route to the determination of the number of atoms displaced in the bulk solid. Further theoretical and experimental studies are required to demonstrate the utility of this approach.

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S. I. TAIMUTY

Stanford Research Institute
Menlo Park, California
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