Effect of Neutron Bombardment on Order in the Alloy Cu_3Au

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Samples of the alloy Cu_3Au in the ordered and disordered states have been exposed in a nuclear reactor, subject to an integrated fast neutron flux up to 3.3×10^{-19} neutron/cm^2. The electrical resistivity, and presumably the disorder, in the initially ordered samples increases, while little change is observed in the initially disordered samples.

IN a nuclear chain reactor under operating conditions, there are present nuclear radiations of several kinds, neutrons, γ-rays, β-particles. In a reactor designed to operate on fissions produced by neutrons of thermal energy, the energy spectrum of the neutrons extends from fission neutron energies, of the order of several Mev, down to thermal energy, about 0.03 ev. The more energetic neutrons, in elastic collisions with nuclei of substances within the reactor, impart to these a considerable fraction of their kinetic energy, on the average the amount ΔE, where

$$\Delta E = E_0(2A/(A+1)^2), \quad (1)$$

E_0 being the initial neutron energy, A the atomic weight of the struck nucleus. For an initial energy $E_0=2$ Mev, on the average a copper atom receives 67,000 ev, a gold atom 20,000 ev of kinetic energy as a result of such an elastic collision.

This energy is sufficient to knock the atom out of its normal lattice site, leaving behind it a lattice vacancy. The recoil atom loses its energy primarily by excitation and ionization of the other atoms it encounters along its trajectory through the solid, and to a smaller extent by displacing in turn other atoms from the lattice, or by exciting localized lattice vibrations of a mean energy corresponding to a temperature higher than the average temperature of the lattice.

While the principal process involved for fast neutrons is elastic scattering, for slow neutrons capture may be the most probable process. Thus, the cross-section σ_c for capture of thermal neutrons by gold is 95×10^{-24} cm^2, while the scattering cross-section σ_s is 6.5×10^{-24} cm^2. Capture of a neutron produces a radio-isotope, here Au^{198} which decays with a half-life of 2.7 days to Hg^{198}, leaving a foreign atom in the lattice.

Thus, the irradiation of a solid by the neutron flux present in a reactor can produce displaced atoms, lattice vacancies, localized thermal lattice vibration, and foreign atoms. These products can affect the macroscopic properties of the solid.

In a simple metal the displaced atoms will not all remain interstitially stuck in the lattice, for vacancies already existing, or produced by the neutron bombardment, will diffuse into the neighborhood of a stuck atom, and the latter will fall into a lattice position equivalent to its original one. The equilibrium number of displaced atoms will depend on the neutron flux, on the rate of vacancy diffusion in the solid, and may be too small to produce significant changes in macroscopic properties. In an order-disorder alloy, however, the possibility exists for observing effects produced by the displacement of atoms, and of lattice vibrations of excessive mean energy, even after the displaced atoms have gone back to lattice sites, for the degree of order of the lattice may have been changed. This change in order will be substantially permanent if the neutron exposure is carried out at a temperature at which no change in order occurs normally, due to conventional thermal annealing. An example of such an alloy is Cu_3Au, which can be permanently kept in any state of order by appropriate heat treatment if the temperature is not subsequently raised above about 200°C.

Samples of the alloy Cu_3Au were prepared in the form of rods 0.3 cm in diameter, 7 cm long. Ordered samples were prepared by slow cooling

TABLE I. Resistivity of samples before and after irradiation.

Original resistivity			Final resistivity	
Ordered samples	Disordered samples	nvt above 50 kev	Initially ordered samples	Initially disordered samples
4.60×10^{-6} ohm cm	11.20×10^{-6} ohm cm	0.4×10^{19} n/cm^2	5.71×10^{-6} ohm cm	11.25
4.60×10^{-6} ohm cm	11.20×10^{-6} ohm cm	0.6	6.25	11.25
4.60×10^{-6} ohm cm	11.20×10^{-6} ohm cm	1.0	7.54	11.17
4.60×10^{-6} ohm cm	11.20×10^{-6} ohm cm	1.5	8.36	11.21
4.60×10^{-6} ohm cm	11.20×10^{-6} ohm cm	3.3	10.10	11.30

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from 400°C during about 50 hours. These samples have an electrical resistivity at 25°C of 4.6×10^{-6} cm. The disordered samples were prepared by quenching in water from 550°C. These have an electrical resistivity of 11.2×10^{-6} cm. The electrical resistivity was used as a measure of the degree of order in these experiments.

The samples were exposed in a chain reactor for various periods in a position where the integrated flux of neutrons of energy higher than 50 kev, times the time of exposure, had the values nvt tabulated below. In each case an initially well-ordered and initially disordered sample were exposed.

During the exposure the temperature of the samples was near 40°C. After exposure the samples are strongly radioactive, and the measurements of electrical resistance must be carried out with remotely controlled devices behind suitable radiation shields. The sample is inserted into a fixture in which current and potential leads are clamped to it by hydraulic pressure acting through a sylphon bellows. The entire fixture is lowered into an oil bath thermostatically controlled at 25°C. The potential drop across a known length of the sample is measured with a Leeds & Northrup Type K2 potentiometer, the current through the sample being determined with the same instrument from the potential drop across a standard 0.001-ohm resistance. The accuracy of the resistance measurements is ±0.2 percent.

The values of the resistivity of these samples before exposure, the integrated neutron flux, and the final resistivity values are given in Table I.

It is evident that the electrical resistance, and presumably the degree of disorder, of the initially well-ordered samples increases with neutron exposure. The electrical resistance of the initially disordered samples increases by less than 1 percent, and this increase of resistance can be accounted for by the formation, through neutron capture, of Hg^{198}, an impurity which raises the resistance of gold.[1]

The samples are so strongly radioactive shortly after exposure that conventional x-ray diffraction study of degree of order through superlattice lines is difficult. However, after the samples had decayed for many months, x-ray evidence confirmed the fact that the initially ordered samples had become disordered.

The calculations of Seitz[2] permit an estimate to be made of the fraction of displaced atoms produced in the integrated flux given in Table I. This fraction is only a few percent, and appears too small to account for the nearly complete disordering of the last sample. However, in addition to atomic displacements, elastic collisions can produce lattice vibrations of greater than thermal energy along the track of the recoil atoms, and reasonable estimates show that the excessive temperature along the recoil tracks may reach values of the order of 1500°K for short periods of the order of 10^{-10} second. These temperature spikes thus produce the effect of highly localized quenches from high temperature, and disorder the lattice in this way. The energy dissipated in exciting elastic vibrations of this type during the exposure appears to be sufficient to account for the disordering observed.

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[1] J. O. Linde, Ann. d. Physik **10**, 52 (1931).
[2] Frederick Seitz, Trans. Faraday Soc. April, 1949 (in press).