2. Radiation Damage

2.1 Primary Damage Processes

2.1.1 The Basic Characteristics of Radiation Damage

In the irradiation of metallic materials by particles with sufficiently high energy, interactions of the incident particles with the atoms of the irradiated material lead to dynamic disturbance and reconstruction of the regular atomic ordering in the crystal lattice. These processes are accompanied by the formation of characteristic lattice defects which affect both the microstructure and the macroscopic structurally sensitive properties of the irradiated material. The unfavourable property changes that occur spontaneously during the interaction of the radiation and the material are a result of radiation damage and can have dramatic consequences, for example, for the safety and economy of nuclear equipment. However, in some cases, these processes can have beneficial consequences and can be utilized, for example in the preparation of special surface layers of new materials. Thus, the term "radiation effects" is now preferred. Radiation effects are a wide range of phenomena that occur spontaneously in solid substances as a result of interaction of the incident particles (with sufficiently high energy) and the atoms of the solid substance.

On passage through the crystal lattice, the bombarding species interact with the lattice atoms and lose some of their energy to these atoms. These processes can be accompanied by damage to the crystal lattice, in general of three types:

(1) production of lattice atoms shifted out of their regular lattice positions, i.e. displacement damage;

(2) changes in the chemical composition by stopping of the bombarding particles (called ion implantation) or capture of particles in the atomic nucleus with consequent transmutation;

(3) excitation of electrons and ionization of atoms (which does not produce permanent damage in metals).

The elementary interaction between the incident particles and the lattice atoms is their collision. This binary process occurs in times of $<10^{-17}$ s and the surrounding lattice can be considered at rest during this time period (the frequency of the thermal oscillation of the atoms is $\sim 10^{13}\ \mathrm{s}^{-1}$). The collisions with the lattice atoms lead to changes in the original direction of the incident particles, called scattering.

These collisions can be elastic or inelastic. A collision is considered elastic when the internal state of all particles involved remains unchanged or the overall kinetic energy of both species remains constant. In inelastic collisions, part of the original energy is lost in some manner; thus

$$E_1 = T_1 + T_2 + Q \tag{2.1}$$

where E_1 is the initial kinetic energy of the particle prior to the collision, T_1 and T_2 are the kinetic energies of the two particles and Q is the inelastic loss.

Kinetic energy T transferred to the lattice atom during elastic collision can be described by the following equation in the classical hard sphere approximation, which can be used for metals in the centre-of-mass coordinate system [1]:

$$T = \frac{4M_1M_2}{(M_1 + M_2)^2} E \sin^2 \vartheta/2 \tag{2.2}$$

where M_1 and M_2 are the masses of the incident particle and the lattice atom at rest, respectively, E is the energy of the incident particle, and ϑ is the scattering angle. The maximal value of T_m in a head-on collision ($\vartheta = 180°$) is

$$T = \frac{4M_1M_2}{(M_1 + M_2)^2} E\,. \tag{2.3}$$

Atoms knocked out of the lattice positions by the impinging particles, called primary knock-on atoms — PKA — can have various energies from zero to T_m even for monochromatic incident radiation. In addition, the energy spectrum of the PKA depends on the type (mass, charge) and energy of the incident particles. Differences in the PKA energy spectrum lead to different damage development and extent.

The formation of PKA is equivalent to the formation of a vacancy-interstitial defect pair (or Frenkel pair).

The minimal energy required for displacement is called the displacement threshold energy, T_d, and its mean value (independent of the crystallographic direction) is between 20 and 50 eV for all metals. The lattice atoms can attain this energy in collisions with electrons with an energy of >1 MeV, neutrons with an energy of $>$ 1keV, or ions with an energy of >100 eV.

The transfer of energy smaller than T_d leads to a local increase in the atomic oscillation, i.e. a local temperature increase.

Part of the energy of the impinging particles is consumed in inelastic collisions and interactions with the electron subsystems and the atomic nuclei of the crystal and does not affect the kinetic energy of the atoms.

The electron excitation or atomic ionization (or stripping of electrons from the impinging particles) that occur in the crystal lattice is rapidly compensated by the free electrons in the lattice of the metal. This type of interaction does not lead to atom displacement, i.e. to the formation of permanent defects*).

Non-elastic interactions with the nuclei, i.e. particle capture in the nuclei, lead to transmutation or fission**). In general, these processes can be described by the scheme:

$$a(M_1) + X(M_2) \rightleftharpoons Y(M_1 + M_2) \rightleftharpoons Z(M_3) + b(M_4) + Q\,, \tag{2.4}$$

or, in short, $X(a, b)Z$ or (a, b). Small letters a and b designate the incident and emitted particles, respectively, capital letters X, Y and Z are the nuclei, $M_1 \dots M_4$ are the masses of the corresponding reaction components and Q is the reaction energy. Thus, $Y(M_1 + M_2)$ designates the unstable nucleus formed with mass $M_1 + M_2$, which immediately decomposes. In all these processes, the basic laws of conservation are fulfilled (conservation of energy, of momentum, of the overall charge and of the number of nucleons.). Atoms with nuclei with mass M_4 can obtain sufficient kinetic energy to leave their lattice positions. In contrast to PKA formed in elastic collisions, these inelastic collision atoms are termed recoil atoms. Their energy spectrum is very different from that of PKA.

In contrast to atom displacement as a result of elastic collisions (a physical process), transmutation processes, also termed chemical processes, occur continuously over the whole irradiation period and their frequency depends on the energy spectrum of the particles and on the composition of the irradiated material, and very little on the irradiation temperature.

New isotopes ((n, γ) reactions) or other chemical elements ((n, α), (n, p) or (n, β)) reactions are formed in transmutations. In technical practice, the formation of atoms of the inert gases is most important, especially of He (and H), which has been observed in a number of materials, e.g. steel. The presence of these gases greatly affects defect interaction during irradiation at elevated temperatures, as a result of their easy diffusion at elevated temperatures, negligible solubility in the lattice and chemical inertness; they thus greatly affect the development of the microstructure of the material during the irradiation.

Binary elastic collisions between the moving particle/atom and the stationary lattice atom are the dominant mechanism of formation of primary defects in metal crystals and a natural elementary step in all models and computer simulations of damage processes.

---

*) However, these interactions can produce displaced atoms through a non-collision mechanism in ionic crystals and semicondutors.

**) It should be pointed out for the sake of completeness that, under special conditions, loss of energy of the impinging particles can also be connected with nuclear excitation, through the formation of bremsstrahlung or Cherenkov radiation.

2.1.2 Characteristic Parameters

2.1.2.1 Flux, Fluence

The quantities characterizing a given type of particle are:

(a) the energy E or energy spectrum $\varphi(E)$ (flux density spectrum) of the species for monoenergetic or continuous radiation;

(b) the flux ϕ (or flux density), characterizing the rate at which the radiation acts and

(c) the fluence (dose, exposure) Φ describing the time effect of the radiation. It often happens that material can be exposed to several types of radiation at once.

The flux of a given type of particle (e.g. neutrons, protons, electrons) incident on the material is given by the number of the particles of the energy E that pass per second through an area of 1 cm^2 perpendicular to the radiation direction. It holds that

$$\phi = \int_0^\infty \varphi(E)\,\mathrm{d}E \tag{2.5}$$

where ϕ is the integral particle flux m^{-2}s^{-1}, $\varphi(E)$ is the differential flux of particles with an energy E (flux density, m^{-2}s^{-1}MeV^{-1}). The effect is often studied in a particular energy interval, so that

$$\phi(E_a, E_b) = \int_{E_a}^{E_b} \varphi(E)\,\mathrm{d}E \tag{2.6}$$

where E_a and E_b are the limits of the energy interval considered. Quantity ϕ_d — the displacement flux — is important in the study of radiation damage, determining the flux of particles that can produce atomic displacement

$$\phi_\mathrm{d} = \int_{T_\mathrm{d}}^\infty \varphi(E)\,\mathrm{d}E \tag{2.7}$$

where T_d is the threshold energy for displacement. In general, the differential flux is a function of time, $\varphi(E, t)$.

Assuming the neutron energy spectrum constant, the overall effect of radiation on material over the irradiation period t_r is given by its fluence Φ (m^{-2}):

$$\Phi = \int_0^{t_\mathrm{r}} \phi(t)\,\mathrm{d}t = \int_0^{t_\mathrm{r}}\int_0^\infty \varphi(E, t)\,\mathrm{d}E\,\mathrm{d}t. \tag{2.8}$$

The neutron energy spectrum $\varphi(E)$ for reactor radiation [2] is very wide (0–20 MeV) and is usually separated into three components:

$$\varphi(E) = \varphi_t(E) + \varphi_{int}(E) + \varphi_r(E) \tag{2.9}$$

where $\varphi_t(E)$ is the thermal, $\varphi_{int}(E)$ is the intermediate (or epithermal) and $\varphi_r(E)$ is the fast part of the spectrum, Fig. 2.1. However, the boundaries of the individual energy intervals are not unambiguously defined; thermal neutrons

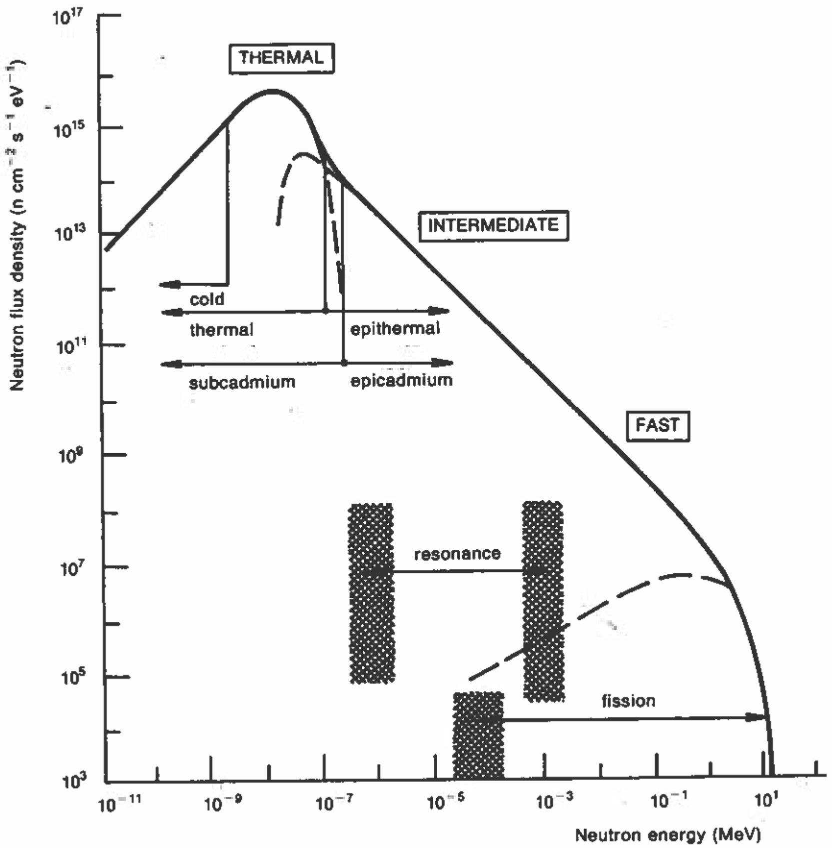

Fig. 2.1. Neutron spectrum for a moderated thermal reactor. From [2].

have an energy in the interval (0–1 eV), epithermal neutrons are usually considered to lie in the interval (1 eV, 0.1 MeV), while fast neutrons have an energy of (0.1 to 20 MeV). Fast and thermal neutrons participate variously in the number of defects produced by the radiation.

2.1.2.2 Interaction Cross-sections

The cross-sections are material parameters characterizing the interaction of the radiation particle and the material. These crosssections depend on the type of irradiated atoms and on the type of radiation and are a function of its energy (this dependence need not be monotonous or continuous). In general, to every interaction (for example elastic or inelastic collision, excitation of atomic electrons, ionization, nuclear reactions, etc.) corresponds a cross-section.

The probability of the interaction between the impinging particle and an atom of the irradiated material is determined unambiguously by its differential cross-section for any type of interaction, for example $\sigma(\vartheta)$ for scattering to the angle ϑ. This cross-section and kinematic relations give the distribution of the scattering angles for scattering of a beam of particles in the given interaction. In addition to the cross-section $\sigma(\vartheta)$ with dimension of area/solid angle, the cross-sections of equivalent quantities such as energy can be used, for example $\sigma(E)$ (with dimension of area/energy) or the double differential cross-section $\sigma(E, \vartheta)$ (with dimension area/solid angle, energy).

Integration over the whole range of space angles yields the total scattering cross-section, $\sigma_s(E)$ (resp. σ_{tot} [3]*)), which is, in general, a function of the energy E_1 of the incident particles. Integrations across selected parts of the integration interval are termed partial or incomplete cross-sections [3].

Processes that lead to displacement of the lattice atoms are important in the study of radiation damage. The displacement cross-section $\sigma_d(E)$ – see Section 2.2.2.2 – is a cross-section which includes only those interactions that can involve

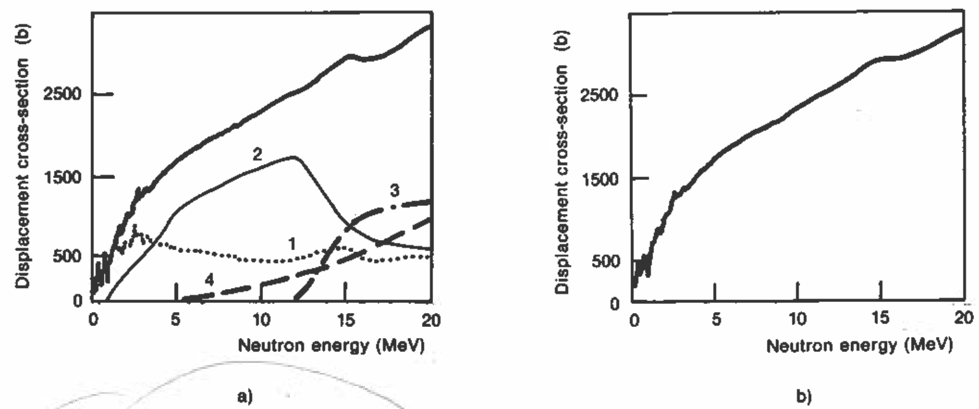

Fig. 2.2. Displacement cross-sections for iron a) and 18/10 stainless steel b). Key to reactions: 1 (n, n), 2 (n, n'), 3 (n, 2n), 4 (n, c). After [4].

*) *Note:* σ_{tot} is usually reserved for the sum of cross-sections of interactions taking part in the collision.

elastic collisions
inelastic collisions (recoil, chemical)

the transfer of energy greater then the threshold displacement energy. In the simplest approximation, it includes only the contribution of elastic scattering; in the case of real reactor radiation, the contributions from inelastic collisions with electrons and nuclei must also be considered (see Section 2.2.2), Fig. 2.2 [4].

The above cross-sections are a measure of the probability of a given type of interaction and are equal to the mean number of reactions per impinging particle per target atom per unit area. This quantity has the dimensions of m^2 and is given in units 10^{-28} m^2 (barns). Geometrically, it can be considered to be the microscopic cross-section corresponding to the area occupied by the targed particles perpendicular to the path of the impinging particles. (For example, for hard sphere collision, it corresponds to $\pi(R + r)^2$, where R and r are the radii of the targed and incident particles.)

The corresponding macroscopic cross-section Σ is related to a given group of atoms, usually to the number of atoms per unit volume, and is given by the relationship

$$\Sigma = N\sigma \tag{2.10}$$

where N is the number of target atoms per unit volume. It has dimensions of m^{-1} and is inversely proportional to the mean free path of the particle in the material. In a multicomponent material, the macroscopic cross-section is given by the sum of the contributions of the individual components.

The quantities characterizing the interactions of a beam of particles with flux density spectrum $\varphi(E)$ and the material are termed reaction rates. They give the number of reactions per second per target atom. They are defined by expressions such as $\int\sigma(E)\,\varphi(E)\,dE$ and integration is carried out over the whole energy range. These relationships can be used for any cross-section.

2.1.3 Primary Events

2.1.3.1 Neutrons

Neutrons have no electric charge and thus need not overcome repulsive coulombic forces and can interact directly with the nucleus. This interaction depends on the energy of the impinging neutrons.

Atoms are shifted out of their normal lattice positions as a result of interactions with neutrons by two basically different mechanisms:

(1) Collision of the neutrons with the atomic nucleus, during which energy greater than T_d is transferred to the nucleus, which is, however, not changed. These collisions can be either elastic (and can be considered to be classical collisions of hard spheres) or inelastic. The incident neutron must have an energy of $\gtrsim 10^2$ eV for such a collision to occur.

thermal neutrons
fast neutrons

(2) Neutron capture in the nucleus producing an excited compound nucleus with a mass of $(M_2 + 1)$, which is unstable and decays over a very short time period ($\sim 10^{-16}$ s) with emission of neutrons, protons, and γ radiation or fission

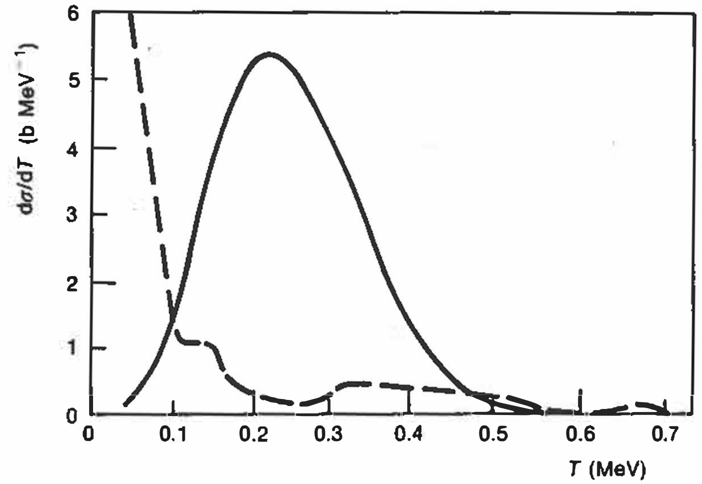

Fig. 2.3. The energy spectra of PKA (dashed line) and recoil atoms (solid line) produced in nickel by 14 MeV neutron irradiation. From [5].

products. The recoil connected with emission of the decay products, similar to the previous case, can lead to atom displacement. In contrast to PKA, these atoms are termed recoil atoms [5].

The energy spectrum of the displaced atoms — PKA in the first case and recoil atoms in the second case — can thus differ. Recoil atoms have higher energy, see Fig. 2.3. The contribution of the recoil atoms to the radiation damage increases with increasing energy of the incident neutrons.

Neutron capture can occur with both thermal neutrons (neutrons in thermal equilibrium with their environment, with very low energy $\approx 10^{-2}$ eV) and fast neutrons with energy above 2MeV.

PKA and recoil atoms behave identically on their path through the crystal lattice: they produce further displaced atoms until their kinetic energy is exhausted.

A considerable part of the interactions of thermal neutrons with the crystal atoms are inelastic (for example, the elastic scattering cross-section for Fe is only about four times larger than that for capture). Nuclear decay is then accompanied by emission of one or more γ quanta, and the reaction is designated as (n, γ). Energy T_γ transferred to the recoil atom is then given by the equation

$$T_\gamma = \frac{E^2}{2M_2c^2} \tag{2.11}$$