Nuclear Reacter Materials & Applications B. M. Ma, Van Nostrand Rembold 1983

Chapter 4 Fundamental Radiation Effects on Materials

This chapter introduces the fundamental radiation effects, or damages, on crystal lattice defects, or imperfections, of nuclear materials. The various crystal lattice defects induced by fast neutrons or heavy particles in intense radiation environments are defined and described. Crystal lattice defects are the vacancies, interstitial atoms, ionization and electronic excitation (in the temporal state), temperature spike, displacement spike, and replacement collisions. Some important theories, based on experimental results, proposed to interpret the radiation effects are the atomic displacement, temperature or thermal spike, displacement spike, and replacement collision models for metals. The fundamental radiation effects on the change of the crystal structure and mechanical properties, electronic and physical properties, and thermal and diffusion-controlled rate process properties are discussed. The radiation effect on these properties is generally proportional to the radiation flux, particle energy (or energy spectrum), irradiation time, and irradiation temperature. The change of the material properties can have a great effect on nuclear (power) reactor design, operation, performance, and safety.

4.1. INTRODUCTION

The unique characteristic of the nuclear reactor environment is the presence of intense radiation accompanied by high temperature in the core. The intense radiation, or irradiation, and high temperature can change the nuclear, physical, thermal, chemical, and mechanical properties of materials, especially the nuclear materials fabricated and operated through a long period of service in the reactor core. Changes or alterations of these properties with operating time can be drastic and must be considered in the design of reactors and associated equipment.

Nuclear radiation in a reactor consists of α , β , and γ rays, neutrons, fission fragments, and possibly protons. The nuclear radiation effects on crystalline solids vary with the crystal structures and the nature of the radiation. Ioniza-

61

62 NUCLEAR REACTOR MATERIALS AND APPLICATIONS

tion and electron excitation, for example, produced by β and γ rays cause little permanent change in metals. Heavier particles, such as neutrons, protons, α particles, and fission fragments, however, can produce significant changes in the properties of metals. Based on the crystallography, these heavy particles can cause imperfections or defects in the regular array of atoms in crystalline solids, both metallic and nonmetallic.

4.2. CLASSIFICATION OF CRYSTAL IMPERFECTIONS OR DEFECTS

From the diffraction of x rays by crystals and the consequent elucidation of the structures of various solids, understanding of the properties of solids has been advanced. An ideally perfect crystal can be defined as the lattice arrays of atoms persist without defects in all directions in the crystal. In other words, an ideally perfect crystal consists of a periodically orderly array of atoms whose arrangement fulfills the condition of a space group of the pattern theory or geometrical crystallography. In the case of mechanical (cold or strain-hardening) work or nuclear radiation (or irradiation) due to heavy particles, however, imperfection, or defect, is often used to describe any deviation from such an orderly array of atoms in crystalline solids (1, 2). If the deviation from an orderly array is localized to the vicinity of only a few atoms, it is called a point imperfection or defect. On the other hand, if the deviation extends through spread regions in the crystal, it is called a lattice imperfection, because it produces discontinuity in the lattice. Crystals have two types of lattice imperfections: (1) line defects, which propagate along lines in a crystal, and (2) plane defects, which have an areal extent in the lattice plane.

4.2.1. Point Defects

Solid solutions of metal provide typical examples of point imperfections: (a) the interstitial, where extra atoms are present in interstitial solid solutions; (b) the Schottky defect, in which atoms are missing from their regular sites; and (c) the Frenkel defect, in which atoms are displaced to interstitials and thus create nearby vacancies (3, 4). Figure 4.1 illustrates the three types of point imperfections or defects occurring at a point in the crystal.

4.2.2. Line Defects

In connection with lattice imperfections, dislocation or linear deviation from true periodic array occurs when the periodicity of the atomic lattice array is interrupted along certain directions in a crystal. There are two kinds of line defects related to the dislocations: (a) edge dislocation, or Taylor dislocation, in which the dislocation line appears to mark the edge of a plane of atoms that

FUNDAMENTAL RADIATION EFFECTS ON MATERIALS 63



Fig. 4.1. Point imperfections or defects in crystals

has been inserted, part way, into the crystal; and (b) screw dislocation, or Burgers dislocation, in which a row of atoms about a normal crystallographic plane appears to spiral in the manner of a screw. The screw dislocation consists usually of a line of atoms, each of which has the correct number of coordinating atoms. The coordination, however, is distorted due to dislocation movement. The direction and magnitude of a screw dislocation are sometimes represented by the Burgers vector of the Burgers dislocation. The Burgers vector is normal to the distorted plane of the crystal. The genesis of screw dislocation is most clearly seen in a three-dimensional view of the crystal.

4.2.3. Plane Defects

When the line defects cluster together in a plane, they can form a plane defect. There are three types of plane defects: (a) lineage boundary defect, where the boundary between two adjacent perfect regions in the same crystal is tilted with respect to each region in the plane; (b) grain boundary defect, where the grain boundary between two crystals in a polycrystalline solid exists; and (c) stacking fault, or defect, in which the boundary between two parts of a close packing has alternate stacking. Most common materials (or nuclear reactor materials) consist of many small interlocking crystals or grains with random orientations. The boundary between adjacent grains, therefore, must be compatible with the structures and orientation of all the grains involved at the common border, or grain boundary, but the grain boundaries can be distorted.

4.2.4. Dislocation, Slip Plane, and Climb Process

Dislocations of the line and screw defects were originally introduced to explain plastic deformation, crystal growth, electric resistivity, and physical properties



64 NUCLEAR REACTOR MATERIALS AND APPLICATIONS

Fig. 4.2. Screw dislocation in crystals

of metallic solids (5, 6). A dislocation can also be described by means of a closed loop, or Burgers circuit, surrounding the dislocation line. This closed loop is formed by proceeding through the undisturbed region surrounding a dislocation in steps of each lattice translation, as shown in Fig. 4.2. The distorted plane that contains the dislocation line is called the *slip plane*. An edge dislocation is free to move in its slip plane because the coordination of the atoms in the edge dislocation, however, can move in any plane parallel to itself because the motion of a screw dislocation is contained along successive, parallel atom rows. When an edge dislocation moves out of its slip plane, the process is called *climb*, and such motion is normally accompanied by the creation of interstitials or vacancies (or point defects) in the crystal during the climb process.

4.2.5. Summary of Crystal Imperfections or Defects

Imperfections, or defects, in crystals are summarized in Table 4.1. These imperfections in crystals or nuclear reactor materials can be produced by mechanical, thermal, and radiation effects. In particular, neutron irradiation in the environment of nuclear (research or power) reactors can produce a variety of crystal defects and radiation effects, or damages, in nuclear reactor materials (7, 8).

4.3. INTERACTION OF NUCLEAR RADIATION WITH MATTER

The interaction of nuclear radiation with matter at high energies is a complex phenomenon that can be resolved into primary and secondary stages, or effects. Table 4.2 gives the primary and secondary effects of the fundamental particles:

FUNDAMENTAL RADIATION EFFECTS ON MATERIALS 65

CLASSIFICATION	TYPE	DESCRIPTION
Point defects	Interstitial	Extra displaced or impurity atom in an interstitial site
	Schottky defect	Atom missing from regular site in an orderly array
	Frenkel defect	Atom displaced in interstitial site and creates a nearby vacancy
Line defects	Edge dislocation	Atom row marks edge of a crystallographic plane inserted partly into the crystal
	Screw dislocation	Atom row about which a normal crystallographic plane appears to spiral along the thread of a screw
Plane defects	Lineage boundary	Boundary between two adjacent perfect regions in the same crystal is tilted with respect to each region
	Grain boundary	Boundary between two crystals in a polycrystalline solid
	Stacking fault	Boundary between two parts of a closest packing has incorrectly alternate stacking procedure

Table 4.1. Imperfections or Defects in Crystals

electron (β ray), photon (γ ray), α -particle (α ray), proton, and neutrons interacting with matter. Nuclear transmutations, such as ${}^{9}\text{Be}(p, \alpha){}^{6}\text{Li}$, ${}^{27}\text{Al}(p, \gamma){}^{28}\text{Si}$, ${}^{6}\text{Li}(n, \alpha){}^{3}\text{T}$, ${}^{27}\text{Al}(n, \gamma){}^{28}\text{Al}$, ${}^{9}\text{Be}(\alpha, n){}^{12}\text{C}$, ${}^{23}\text{Na}(n, \gamma){}^{24}\text{Na}$, etc., can induce a number of interactions of fundamental particles with nuclear reactor materials. At the same time, the fission fragments induced in a nuclear fission reactor can produce heavy damage within the fuel material.

Among the fundamental particles, neutrons, especially fast neutrons, have these features: (a) neutral particle, (b) high penetrating power, (c) relatively heavy mass, and (d) plays the most significant role in a nuclear fission reactor and probably in a deuterium-tritium fusion reactor in the future. The radiation effect on nuclear reactor materials, particularly fuel and structural materials, therefore, will be very important.

Displacement (at high energy)
Displacement (at high energy)
Radiation damage (at high energy)
Displacement (subsequent)
Ionization

Table 4.2. Interaction of Fundamental Radiation Particles with Matter

4.4. RADIATION EFFECT, OR DAMAGE, BY NEUTRONS

The change, or damage, in nuclear, physical, thermal, chemical, or mechanical properties of reactor materials under intense radiation is called the *radiation effect*, or damage. The principal radiation effects resulting from the interaction of fundamental particles with nuclear reactor materials are electronic ionization and atomic displacement (see Table 4.2). Electronic ionization (by electron, photon, proton, and α -particles) is a temporary effect. Atomic displacement, for instance, by neutrons, is permanent damage. Because the neutron has no charge, it produces radiation damage only by energetic interaction with nuclei in the reactor materials. A fast neutron can impart both energy and momentum to a nucleus with which it interacts, and the nucleus becomes excited and displaced, or recoiled, from its regular site, or orderly array, in the crystal (see Section 4.2). Therefore, the radiation, as well as irradiation, effect by neutrons can change the properties and reduce the performance or service lifetime of nuclear reactor materials appreciably.

There are a number of radiation defects induced by intense nuclear radiation, particularly, fast neutron irradiation. These radiation defects are described concisely below:

- (1) Vacancies. The creation of vacant sites in the crystal lattice due to collisions between fast neutrons and nuclei or atoms. The energy transferred from the fast neutron to the nuclei is usually so large that each primary collision produces subsequent knock-ons and cascade collisions, resulting in vacancies.
- (2) Interstitials. Nuclei displaced by collisions to interstitials or irregular nonequilibrium positions are not recombined with nearby vacancies.
- (3) Impurity atoms or alloying atoms. Impurity or alloying atoms are produced by nuclear transmutations due to collisions of incident neutrons and regular nuclei of nuclear reactor material. Fission products, such as Mo, Zr, and Ru (or fissium), produced by nuclear fission reactions are the impurity or alloying elements in nuclear fuels.
- (4) Ionization and electronic excitation. Neutrons and γ rays can induce local ionization and electronic excitation in their passages. The local ionization and electronic excitation induced by the neutral particles or rays can introduce further vibrational energies in nuclei or atoms.
- (5) Temperature and thermal spikes. Atoms have high vibrational energies in excess of the normal state. The region in which a large number of atoms are involved in strong vibrational states is called a *temperature spike*. If the vibrational excitation is relatively small, so that only a small number of atoms are involved, or leave their equilibrium sites, such a weak vibrational state is called a *thermal spike*.

- (6) Displacement spike. The displacement spike originated from the atomic displacement model that can be produced by (a) primary knock-ons and subsequent cascade collisions, and (b) sufficient vibrational excitation to allow many atoms to leave their lattice sites and move around the displacement spike. It usually contains a large number of vacancies, interstitials, and other irregular crystal lattices concentrated as a spike.
- (7) Replacement collisions. Scattered interstitial atoms, after collisions between moving interstitial atoms and stationary atoms, fall into their vacant lattice sites and dissipate their excited or kinetic energy through lattice vibrations. As a result, the interchange of moving interstitial atoms with stationary lattice atoms through the collisions and replacement process is known as *replacement collisions*.

The first three defects are the basic crystal defects due to radiation of neutrons (see Section 4.2). The last four are the cluster crystal defects. Experimental results obtained from postirradiation examinations verify the phenomena of crystal defects of the neutron irradiation effect or damage.

4.5. PROPOSED MODELS OF RADIATION EFFECTS, OR DAMAGE

The effect of nuclear radiation, mainly, fast neutron irradiation, on metals in general is the result of localized lattice defects or damage to the crystal structure. Various models or theories have been proposed for the radiation effect of different metals, based on different sets of experimental data. Among them, some primary models proposed to account quantitatively for the radiation effect are as follows:

- (1) Atomic displacement model. In the atomic displacement model, the struck, or knocked-on, atoms are displaced from their equilibrium positions or regular arrays in the crystal lattices, as a result of the primary and secondary collisions between the fast neutrons, or heavy particles, and the atoms. The energies transferred from the neutron to the atom in an elastic collision could far exceed the value required to displace an atom in a nuclear reactor material; e.g., the energy required is about 25 eV to displace a metallic atom or nucleus and about 12-15 eV to displace an atom of semiconductor, germanium or silicon (see Sections 4.6, 4.7). In many cases, the kinetic energies transferred to the knocked-on atoms are so large that they produce secondary knock-ons by elastic collisions with other atoms in the material. Finally, those displaced atoms come to rest in the interstitial positions and vacancies to form a displacement spike.
- (2) Temperature or thermal spike model. A fission fragment usually dissipates all its energy, and a fast neutron normally loses a large proportion of its