Module 4

RADIATION DAMAGE TO MATERIALS

OBJECTIVES:

After completing this module you will be able to:

Page 3 ⇔ 4.1 Explain why ionizing radiation has little effect on metals and their mechanical properties, but creates significant damage in non–metals.

Page 3 ⇔ 4.2 a) Describe the damage created in metals by fast neutrons.

Page 5 ⇔ b) Explain how this damage affects: strength, hardness and ductility.

Page 4 ⇔ 4.3 Explain why elevated operating temperatures reduce the effects of fast neutron damage.

Page 6 ⇔ 4.4 State the affect of thermal neutrons on the properties of metals.

4.5 Describe the effect of radiation on:

Page 7 ⇔ (a) oils and greases,

Pages 8, 9 ⇔ (b) plastics and rubbers,

Page 10 ⇔ 4.6 a) Explain how radiation affects the structural integrity and shielding properties of concrete,

Page 10 ⇔ b) State how damage to concrete biological shielding is avoided.

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INSTRUCTIONAL TEXT

INTRODUCTION

Materials in nuclear service are subjected to various types of radiation, e.g. neutrons, gamma, beta and alpha. Some of these can cause significant damage to the crystalline structure of materials. Nuclear radiation focuses large amounts of energy into highly localized areas. Damage is caused by the interaction of this energy with the nuclei and/or orbiting electrons.

The crystal structure may be modified through these energy interactions. We can, therefore, expect changes in the bulk material's mechanical properties.

It is important to remember that materials in our reactors, especially those in core, experience bombardment by all types of radiation simultaneously. Consequently, the damage sustained can be quite complex. In order to more easily understand this damage, we will separately consider each type of radiation and its effects on a material, realizing that the overall effect will be some combination.

EFFECTS OF IONIZING RADIATION ON TYPES OF ATOMIC BOND

Gamma, beta and alpha are classed as ionizing radiation because they interact only with electrons surrounding nuclei. Ionizing radiation strips electrons from the outermost orbitals of the atoms, creating nuclei which are charged ions. This ionization disrupts the atomic bonding of materials. The stripped electrons may continue to create subsequent damage. To explain further, let us consider the different types of atomic bonds.

Covalent bonds occur when atoms are held together by the sharing of electron pairs. Ionizing radiation, by stripping electrons from their atoms, can break up bond pairs of electrons. This causes disintegration of the original molecules and the formation of new and different ones, ie, a chemical change. Organic compounds such as oils, plastics or rubber (natural and synthetic) contain almost exclusively covalent bonding and, therefore, suffer significant damage by ionizing radiation. Further details of this radiation damage are given later in this module.

Ionic bonds form when oppositely charged ions are held together in a crystal lattice by electrostatic attraction only; all electrons in the compound are held in their orbits around particular atoms. Ionizing radiation is not nearly so destructive to an ionic bond as to a covalent one. The electrical charge on individual ions may be altered by the stripping of electrons from an atom, but the oppositely charged pairs will still experience electrostatic attraction.

A displaced electron may eventually find its way to another electron deficient site resulting in no net damage. However, should the electron become trapped in a crystal lattice imperfection, we get what is called an F–centre*. This can cause changes in colour, but has little effect on the mechanical properties of the material. For example, glass soon turns black when exposed to ionizing radiation, though its structural properties are virtually unaffected.

* *This stands for Farbe–centre, named after the scientist who discovered the phenomenon.*

NOTES & REFERENCES

The majority of inorganic materials, other than metals, exhibit ionic bonding and are normally good insulators because all available electron sites are filled and there is very little electron movement from site to site. Since ionizing radiation causes electron migration, the resistivity of these materials is reduced during irradiation, making them more conductive.

Obj. 4.1 ⇔

In **metallic** bonding, a simple view of the bonding arrangement is most practical for considering the effects of ionizing radiation. Metals consist of positive ions held in fixed positions (a crystal lattice) surrounded by a sea of electrons. The electrons are very mobile and not attached to any particular atoms; they move from atom to atom under a variety of influences. Ionizing radiation will have only a transient effect on metals because the electrons stripped from any particular atoms are readily replaced through the free movement of other electrons.

EFFECTS OF FAST NEUTRON RADIATION

Neutrons with sufficient energy can disrupt the atomic arrangement or crystalline structure of materials. Fast neutrons* entering a material displace atoms, resulting in structural damage and possibly temporary localized zones of high temperature. The influence of structural damage is most significant for metals because of their relative immunity to damage by ionizing radiation.

Thus in the following discussion, although we concentrate on the effects of fast neutrons on metals, we must realize they can have similar effects on all materials.

INFLUENCE OF FAST NEUTRON DAMAGE ON MECHANICAL PROPERTIES

Obj. 4.2 a) ⇔

Room temperature tensile tests show that **fast neutron radiation causes significant increase in yield and tensile strength, along with a decreased ductility in metals.** Figure 4.1 illustrates this. Accompanying the increase in strength is an increase in hardness. These changes are due to the fact that deformation is more difficult in the damaged metal than it is in the undamaged metal. The defects** created in the damaged metal introduce irregularity to the crystal lattice and hinder deformation processes.

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* *Neutrons with energies of about 1 MeV, or greater. This energy represents the vast majority of fission neutrons. Lower energy fission neutrons will also cause damage, but they are less numerous.*

** *These defects are called vacancies and interstitials. A vacancy is the space left behind by a displaced atom. An interstitial is an atom forced into a position that it would not naturally assume between other atoms.*

NOTES & REFERENCES

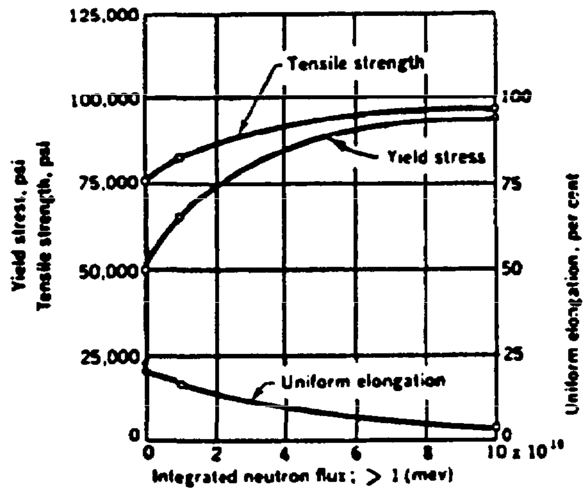

Figure 4.1: Fast Neutron Damage Influence on Mechanical Properties

Obj. 4.3 ⇔

If the metal is heated to elevated temperatures after irradiation (a form of annealing), it is found that the strength and ductility return to the same values as before irradiation. This means that radiation damage can be annealed out of a metal*. Therefore we can expect little change in the strength and hardness of a metal operating at a high temperature while being irradiated. The action of heat, as you may recall from Module 1, expands the crystal structure and makes it less rigid. The defects (vacancies and interstitials) created by neutron bombardment of the metal become mobile in this expanded structure. Some move to locations where they are less hindrance to deformation. Others move together and recombine, resulting in the elimination of these defects.

In nuclear reactors, many metal components are subjected simultaneously to radiation fields, elevated temperatures and stress. Metal under stress at elevated temperature exhibits the phenomenon of **creep**, ie, the gradual increase in strain with time. Radiation damage, provided it is at ambient temperatures, should reduce creep. However, as noted in the previous paragraph, radiation damage can be annealed out at elevated temperatures, thus assisting deformation processes. **Creep of metal components at reactor operating temperatures becomes faster when they are exposed to a radiation field.** This is illustrated in Figure 4.2. As will be discussed in Module 5, the effect of radiation on creep is of great importance in determining the life of pressure tubes**.

* *Operations are not conducted in a specific attempt to achieve this result.*

** *Due to the effect of radiation on creep any change in power of the reactor, either up or down, will result in an initial sharp increase in the creep rate.*

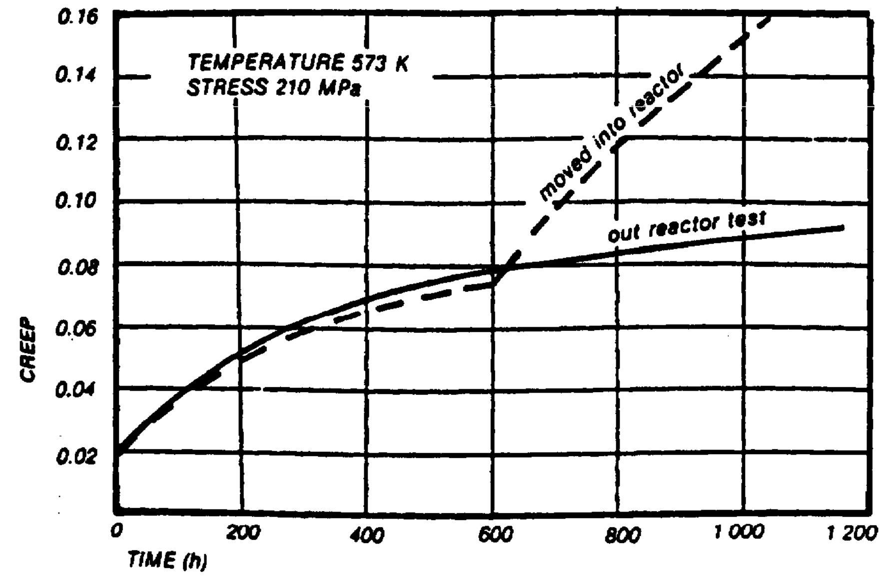

Figure 4.2: Influence of Neutron Radiation on the Creep of Zircaloy–2

Before leaving this section, let us consider some specific effects of radiation damage on two important metals: steels and zirconium alloys.

Neutron irradiation of plain carbon steel produces a marked increase in yield and ultimate tensile strength accompanied by a decrease in the elongation to fracture. Hardness increases, but less so in steels that have been intentionally hardened, eg. the end fittings. The increase in strength after irradiation may be beneficial; however, the accompanying decrease in ductility means increased susceptibility to brittle fracture. Impact tests on irradiated steels indicate that the energy required to cause fracture is decreased and the ductile brittle transition temperature is raised. In addition, it has been found for many pressure vessel steels that operation at 260° C *cannot* anneal out radiation induced brittleness. Even stainless steels, which are not normally notch sensitive, have reduced impact resistance when irradiated.

Neutron radiation damage sustained by Heat Transport System carbon steel piping over its lengthy service life causes gradual raising of its NDTT*. This requires operational restrictions to be put in place. The temperature at which the HTS can first be pressurized during a unit start–up may have to be increased over time.

Obj. 4.2 b) ⇔

Irradiation of Zircaloy considerably increases the yield strength, and to a lesser extent, the ultimate tensile strength. Ductility is reduced, particularly the uniform elongation. Cold working the material prior to irradiation (such as is the condition for pressure tubes) reduces the amount of change seen in mechanical properties.

* *The concept of Nil Ductility Transition Temperature was introduced in Module 2.*

NOTES & REFERENCES

Also, irradiation at elevated temperatures does not produce as severe a change in mechanical properties as irradiation at room temperature. Fuel sheathing becomes embrittled by neutron bombardment. This does not substantially reduce the life expectancy of fuel sheathing, but does restrict the number of positions to which fuel can be shifted to avoid it becoming too brittle to handle on discharge from the reactor.

EFFECT OF THERMAL NEUTRONS

Obj. 4.4 ⇔

From Nuclear Theory, recall that thermal neutrons may be captured by nuclei in the irradiated material, possibly changing its identity. The addition of a neutron to a nucleus changes the atomic mass number and produces an isotope of that nucleus. This isotope may be stable like the original nucleus or may be radioactive, decaying in time to a stable nucleus which will be a different element. For example, Cobalt 59 becomes radioactive Cobalt 60 when it captures a neutron. However, neutron capture by aluminum produces a radioactive isotope which decays to a stable isotope of silicon*. Most neutron captures by iron atoms result in a heavier stable isotope of iron.

Although the UO_2 fuel is not a metal, it should be mentioned that it can experience a production of gases during the process of neutron capture and radioactive decay. Fission, which is initiated by thermal neutrons, produces several gaseous fission products (eg, krypton, xenon and iodine). If the fuel is operated at temperatures higher than those for which it is rated, the gas atoms (normally held at the grain boundaries) migrate and coalesce to form gas bubbles. As these bubbles accumulate, the pressure within the fuel sheath increases.

Thermal neutrons, when compared to fast neutrons, do little structural damage to a metal's crystal lattice. Generally, there is only displacement of the nucleus which captured the neutron. This nucleus is in an excited or high energy state and excess energy** is released by emitting high energy gamma rays with the result that the emitting nucleus recoils and is displaced.

It is important to note that, regardless of any physical damage caused by neutron exposure, metal components within the reactor, or in nuclear process systems, will be activated and must be dealt with as radioactive material when removed.

* *Transformation of materials by neutron capture is a slow process. About 0.4% of the aluminum at the centre of AECL's Chalk River NRX reactor was changed to silicon over a 10 year period.*

** *This energy is the composite of gamma radiation and the kinetic energy of the recoil nucleus.*

NOTES & REFERENCES

SUMMARY OF THE KEY CONCEPTS

- Metals are reasonably unaffected by ionizing radiation because their atoms are bound by a sea of free–moving electrons. Removal of an electron from a particular atom only affects the bond momentarily, as the electron sea quickly provides a replacement.
- Atomic bonding in non–metals involves the sharing or transfer of electrons between atoms. Ionizing radiation strips an electron from the bond, thus altering the bond.
- Fast neutrons cause structural damage to the crystal lattice of metals by displacing atoms.
- Fast neutron damage affects the mechanical properties of metals. It increases strength and hardness but decreases ductility.
- Elevated operating temperatures cause a metal's crystal lattice to be more open (less rigid) allowing fast neutron induced defects to move more easily. This reduces their restriction of deformation allowing strength and ductility to return to their pre–exposure values.
- Thermal neutrons have a less significant affect on the mechanical properties of metals. They can be captured by nuclei of the irradiated material, which will become radioactive.

EFFECT OF RADIATION ON SPECIFIC MATERIALS

Oil and Grease

Obj. 4.5 a) ⇔

Oils suffer an **increase in viscosity** as a result of cross–linking which takes place after bonds have been ruptured. The damage is caused by both neutrons and ionizing radiation. The amount of viscosity increase depends on the composition of the particular oil; the more vulnerable types can actually solidify. In general, oils of high viscosity are more sensitive to damage. In any event, the tendency is to form gummy, tar–like polymers which can restrict lubricant flow.

Grease of the soap–oil type, for example, is susceptible to radiation damage. Radiation causes the breakdown of the soap–based binder (used to hold the relatively liquid lubricant in place in the bearing), making the grease **become more fluid**. Exposure of the grease to air during irradiation, accelerates its oxidation and reduces the amount of radiation required to cause significant damage.

NOTES & REFERENCES

Special additives in the grease, introduced during its manufacture, can be very helpful in limiting radiation induced oxidation and other damage.

In our stations, the lubricants used in the fueling machines are changed on a more frequent basis than would be the case if the lubricant was used in equipment not exposed to radiation.

Plastic

Obj. 4.5 b) ⇔

Plastics (also referred to as *polymers*) are comprised of long chains of molecules. Plastics undergo drastic changes under the influence of radiation, since the bond arrangement can be extensively changed. Bonds are broken by radiation and reform in a different arrangement. The types of chemical changes that occur include cross–linking (bonding between chains), polymerization, de–polymerization, oxidative degeneration and gas evolution. This leads to changes in properties such as viscosity, solubility, conductivity, tensile strength, hardness and flexibility.

Fragmentation into shorter chains, as a result of bonds broken by radiation, will result in the formation of a new polymer that is **softer and weaker** than the original material, perhaps even liquid. Teflon, plexiglas, lucite and butyl rubber are most susceptible to this process. New bonds may be formed to make a new polymer with longer chains than before. When radiation damage causes cross–linking to occur, a **more rigid and brittle** polymer is produced. This form of damage predominantly occurs in polyethylene, polystyrene, silicone, neoprene and natural rubber.

Polymers vary in their resistance to radiation. This is illustrated in Figure 4.3, which shows radiation damage versus absorbed dose for several polymers.

Problems have been discovered in some of our units involving radiation damage to the insulation on power and control cables, especially those within the reactor vaults. The outer jacket on some ion chamber instrument cables has been affected so severely that it has become brittle, cracked and fallen off. Insulators in the associated connectors have also become brittle and failed.

There has also been evidence of terminal blocks decomposing inside junction boxes that are exposed to high radiation fields. A program for cable assessment and replacement has been instituted.

Ion–exchange resins are polymers based on polystyrene. They are also subjected to radiation damage. The radionuclides removed from active water systems deposit much of their energy within the resin bed, leading to its destruction. At present, the active resins are not regenerated, but are discarded. The extent of radiation damage is an unknown, thus discard is a more practical procedure than regeneration.

NOTES & REFERENCES

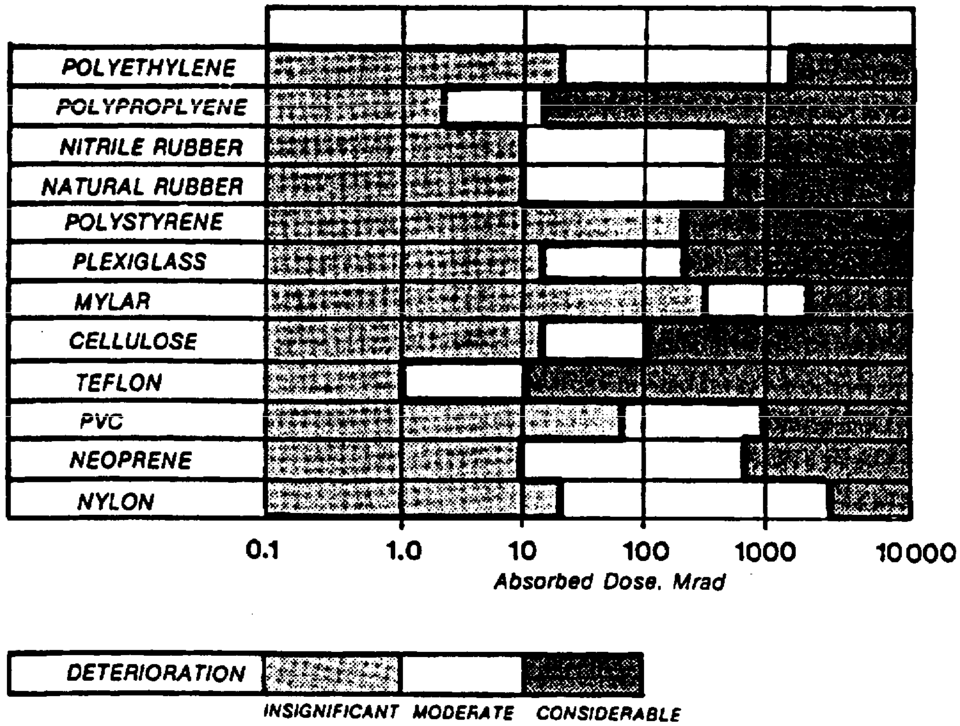

Figure 4.3: Effect of Radiation on Various Polymers

Rubber

Obj. 4.5 b) ⇔

The molecules in elastomers (the generic name for rubber–like substances) are long strings of atoms. Their properties depend on a fine balance between inherent freedom of motion of the chain and the degree of cross–linking between the chains. Natural rubber is more resistant to radiation damage than synthetic rubbers, but the **tensile strength of all is reduced**. The effect on hardness depends on the material– for example, natural rubber gets harder, butyl rubber softens.

At some stations, valves in air and D_2O systems located in containment have experienced breakdown of diaphragms and "O"rings due to radiation (and temperature) effects. Procedures have been changed in response to this concern. The diaphragm components in the heat transport control valves are changed on a call–up at a frequency proven to prevent extensive radiation damage that could otherwise lead to valve failures. Inspection programs conducted on the Instrument Air system during each outage assure that diaphragm problems are detected and the faulty components replaced immediately.

Concrete

Concrete is a common, and relatively inexpensive, construction material used extensively in nuclear stations for structural and radiation shielding applications. Like all materials, it suffers radiation damage. Gamma rays interact with the atoms and the energy of the rays is converted to heat. Neutrons, on being slowed down by either elastic or inelastic collisions, give up their lost energy directly or indirectly as heat. (We will ignore the effect of the relatively few neutrons which are captured and thereby produce new atoms.)

Obj. 4.6 a) ⇔

Heat drives water out of concrete, resulting in cracking and spalling (pieces of concrete separating from the surface). There are two types of water in concrete: chemically bound water (water of hydration), and physically trapped water. Chemically bound water is part of the chemical matrix of cured concrete. Physically trapped water is in excess over that required chemically. It is added to maintain the workability of the mixture, prior to curing. Thus, it is merely suspended in the concrete, not bound to it.

Heat tends to drive away both types of water, particularly the trapped water. Trapped water is readily heated by radiation, but the bonds holding the chemically bound water must first be broken, before this water can be heated and driven off. The internal stresses resulting from the water vaporizing and trying to escape cause cracking.

There is an important second ramification of this water loss; the neutron shielding efficiency decreases. In concrete, the hydrogen atoms in the contained water slow down neutrons by elastic scatter, thus thermalizing neutrons and providing an effective radiation shield.

Obj. 4.6 b) ⇔

The biological shield is one of the most important barriers which protects station personnel from exposure to radiation. In early CANDU stations, it is only provided by thick walls of concrete. It is essential that the integrity of these walls is maintained. To ensure this, at these stations, a system of pipes is placed throughout the concrete and a flow of cooling water is circulated to remove the heat deposited by radiation.

Other stations employ some type of water–filled thermal/biological shield that is provided with its own cooling equipment. The concrete walls beyond this shielding are not subject to sufficient heating to require internal cooling pipes.

NOTES & REFERENCES

SUMMARY OF THE KEY CONCEPTS

- Radiation (neutron and ionizing) causes the viscosity of oils to increase as gummy, tar–like polymers are formed. Radiation causes soap–oil type greases to become more fluid.
- Plastics undergo drastic changes when exposed to radiation. Polymer chains can be fragmented, cross–linked, lengthened or rearranged. All result in new and different material properties. Rubber may become harder or softer depending on its type.
- Concrete, under radiation exposure, heats up. This drives the water out of its internal structure. Swelling, cracking and spalling result.
- To avoid radiation damage to concrete biological shielding at some stations, heat is removed by cooling water pipes embedded in the concrete shielding. At other stations thermal and biological shielding is combined by using water–filled steel tanks/walls which are separately cooled. Concrete shielding beyond the tanks/walls is not subjected to damaging levels of heat.

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You can now do **assignment questions 1 – 9.**

NOTES & REFERENCES

ASSIGNMENT

1. Explain why ionizing radiation has little effect on metals and their mechanical properties, but creates significant damage in non–metals.
2. Describe the damage created in metals subjected to fast neutron irradiation.
3. Describe how fast neutron damage affects the mechanical properties (ie, strength, hardness and ductility) of metals.
4. Explain how elevated operating temperatures reduces the effects of fast neutron damage.
5. Describe the effects of thermal neutrons on metals.
6. Describe the effects of radiation on oils and greases.
7. Describe the effects of radiation on plastics and rubbers.
8. Explain how radiation affects concrete.
9. Describe two methods used to reduce the effects of radiation on concrete.

Before you move on to the next module, **review the objectives** and make sure that you can meet their requirements.

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