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**Radiation Damage of Structural Materials for Fast
Reactor Fuel Assembly (1)**

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**RADIATION DAMAGE OF STRUCTURAL MATERIALS FOR FAST
REACTOR FUEL ASSEMBLY**

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PREFACE

PREFACE

Nuclear power can be generated using two types of nuclear reactions (Figure): the fission and the fusion. In the fission reaction, the nucleus of a heavy atom like uranium (U^{235} or U^{233}) or plutonium (Pu^{239}) is split into lighter fragments, releasing a large amount of energy ($\sim 200\text{MeV}$ per fission event). In fusion, on the other hand, nuclei of smaller atoms like isotopes of hydrogen are fused into larger nucleus like that of helium, producing energy (${}_1D^2 + {}_1T^3 \rightarrow {}_2He^4 + {}_0n^1 + 17.6 \text{ MeV}$).

Nuclear power offers the advantages of environmental friendly and sustainability. The high heat value of fuel, relative high energy density and possibility of high conversion efficiency are some of the additional advantages of nuclear power. Currently, there are 443 commercial fission based nuclear reactors, which account for about 15% of global electric power generation. The widespread use of nuclear energy and its public acceptance revolves around a few key issues such as improved safety, management of nuclear waste, cost-competitiveness and proliferation resistance.

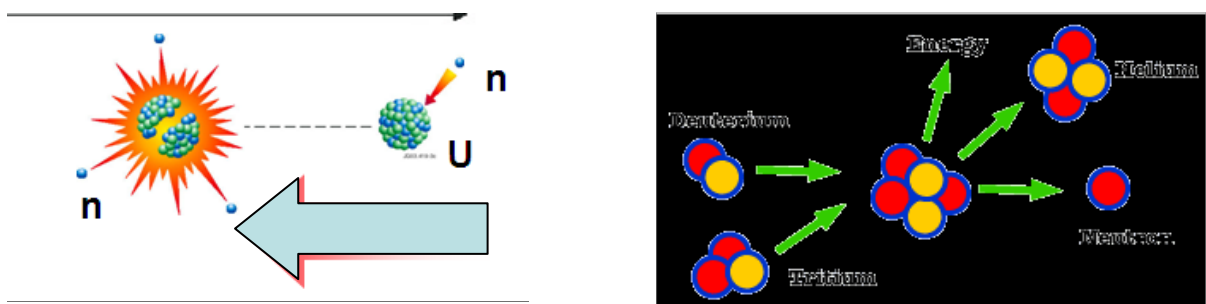


Figure: A typical fission reaction inside a heavy nucleus and fusion reaction between two light nuclei.

Based on energy of the neutrons used for fission of the nuclear fuel, reactors can be broadly classified into two types: 'thermal' and 'fast' reactors. Materials used in thermal reactors (where fission is caused by neutrons with energies < 1 eV) experience relatively less aggressive conditions. The materials used in fast reactors (where fission is caused by "fast" neutrons with energy of the order of 1 MeV) are exposed to more hostile environment, such as high neutron flux (peak flux: $\sim 8 \times 10^{15}$ n/cm²/s against 10^{15} n/cm²/s in thermal reactors) and high temperatures (up to 823 K as compared to 573 K in thermal reactors). In addition, fuels used in fast reactors are irradiated to much higher levels of burn-up (target burn up* of 200GWd/t; present level of burn-up 100 GWd/t) as compared to the fuels in thermal reactors (up to 80GWd/t). The above features suggest that development of radiation resistant materials are absolutely essential to develop cost-competitive fast reactors.

Presently, there is a global attempt to introduce advanced reactor design concepts, to ensure better thermal efficiency, cost, safety, waste management and proliferation resistance. The nuclear materials play a pivotal role to ensure successful implementation of these concepts. Hence, understanding the behaviour of materials under extreme service conditions of a nuclear reactor, is of paramount importance to achieve required "green" energy option, through nuclear source.

INTRODUCTION

STRUCTURAL MATERIALS AND THEIR BEHAVIOUR IN A FAST REACTOR FUEL

ASSEMBLY

INTRODUCTION

STRUCTURAL MATERIALS AND THEIR BEHAVIOUR IN A FAST REACTOR FUEL ASSEMBLY

One of the main future targets of a nuclear industry is cost-competitiveness. The engineering structural materials in a nuclear reactor contribute to roughly around 15% of the cost of the reactor. Hence, there is a large scope for reducing the cost of the reactor by developing the cost-efficient structural materials in a fast reactor fuel assembly. Towards this goal, certain directions are identified:

- (1) develop high burn-up materials;
- (2) increase in lifetime from 60 years to 100 years and
- (3) materials compatible with coolants operating at higher temperature.

It is necessary to develop an in-depth understanding of the materials behaviour to achieve the above goals.

It is necessary to define what we mean by 'structural materials' in a fast reactor fuel assembly, before we proceed to radiation damage in these materials. The first part of the chapter deals with definition of 'structural materials' in a fast reactor fuel assembly.

CLASSIFICATION OF ENGINEERING MATERIALS IN A REACTOR:

The term 'nuclear materials' refers to a host of engineering materials which are used in the reactor, other than fuel, coolant and moderator. Some examples of nuclear materials are the zirconium based alloys used in water reactors and the high temperature radiation resistant steels used in the sodium cooled fast reactors. The nuclear materials can broadly be grouped (Figure.1.) into two categories: core and out-of-core component materials.

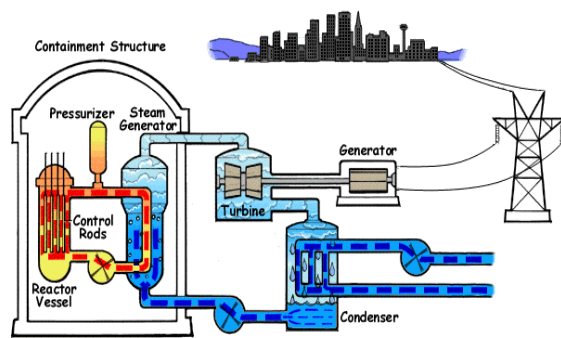


Figure.1. Schematic to represent components in core (red), out-of-core(blue) and balance of plant (black) circuits in a reactor.

The core component materials include the following: fuel assemblies and the neighboring coolant channels for water reactors; and clad (cylindrical tubes which houses the fuel pellets) for the fuel and wrapper (a container which houses fuel elements, in between which the coolant flows) for sub-assemblies of fast reactors.

The structural materials include those which are within the core other than clad and wrapper and hence, do not belong to the ‘conventional’ part of the power generation unit. The out-of-core materials, made of different forms of steels form the remaining part of nuclear steam supply system. The balance of plant components are similar to the conventional systems of any power plant.

CORE COMPONENT MATERIALS:

Conventionally, in a fast reactor systems, the materials that constitute the clad and the wrapper are the major components of the fuel sub assembly or the core. These are referred to as ‘core component materials’. The necessity to develop these materials arises from the fact that the “burn-up” of the fuel, depends on the performance of the clad materials. The development of robust materials technology to achieve high burn-up (energy production from unit quantity of the fuel) of the fuel is an important step

towards reducing the cost of nuclear energy production. The higher burn up of the fuel, increases the ‘residence time’ of the sub assembly in the core, thus lowering the cost. Figure. 2 shows the dependence of the burn up on the residence time, based on Russian experience.

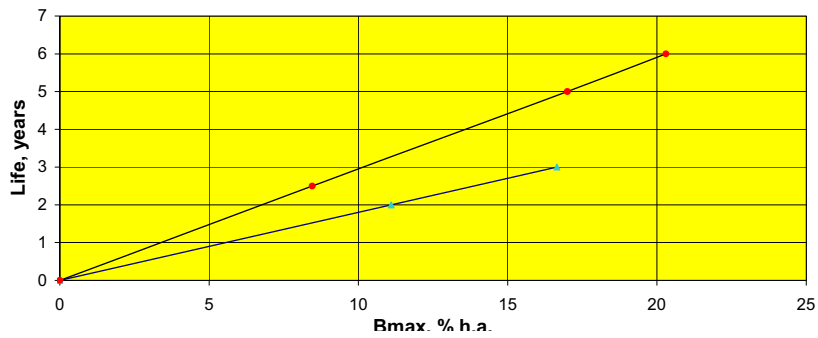


Figure.2. Dependence of residence time on burn up of fuel, for different fuel element diameter. Blue dots : BNH 600 fuel element diameter-6.9 mm & maximum linear heat rate – 47.5 kW/m; Red dots: BN-K fuel element diameter- 9.5 mm & maximum linear heat rate – 45 kW/m.

The fuel sub-assembly consists of hexagonal array of wrappers which contains the clad tubes, as shown in figure.3.

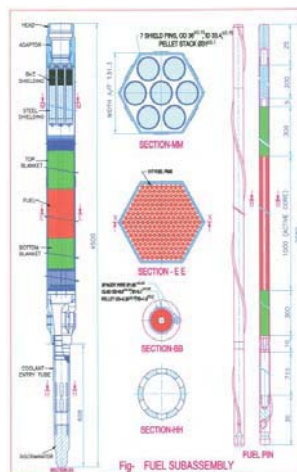


Figure.3. Schematic of the clad, wrapper and fuel sub-assembly in the core of a fast reactor.

The clad materials in fast breeder reactors are exposed to severe environmental

Criterion	Clad Tube	Wrapper Tube
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service conditions, as shown in Table.1.

- ★ Operation in hot sodium
 - Corrosion/erosion of structural materials
 - Appropriate control of sodium chemistry
- ★ Fission gas release in fuel pins
 - Increase in internal pressure
 - Provide plenum space of sufficient volume
- ★ Internal corrosion of cladding due to volatile fission products
 - Lanthanides series fission products
- ★ Severe radiation environment leads to material problems
 - Fast neutron flux: 10^{20} n/m²/s
 - Fluence: 2×10^{27} n/m²
 - Achievable burnup limited by material degradation
- ★ Irradiation induced changes in structural material properties
 - Significant dimensional & mechanical property changes
 - Severe radiation environment leads to material problems like
 - Void swelling
 - Irradiation creep
 - Helium embrittlement

Table.1. Service exposure conditions for clad materials in a fast reactor core.

Given the above environmental or operating conditions of a fast reactor, it is necessary to evaluate the severity of these conditions component by component. For example, the clad tube which contains the fuel, experiences more aggressive environment than the wrapper which houses a large number of clad tubes. Table.2. provides the difference in the exposure condition of the clad and wrapper in a fast reactor core.

Irradiation effects	Temp.,Dose rate and dose to be written Void swelling Irradiation creep Irradiation embrittlement	Temp.,Dose rate and dose to be written Void swelling Irradiation creep Irradiation embrittlement
Mechanical properties	Temp to be written Tensile strength Tensile ductility Creep strength Creep ductility	Temp to be written Tensile strength Tensile ductility
Corrosion	Compatibility with sodium Compatibility with fuel Compatibility with fission products	Compatibility with sodium
Good workability		
International irradiation experience as driver or experimental fuel subassembly		
Availability		

Table.2. Selection criteria for clad and wrapper in core of fast reactors:

The accumulated experience in fast reactor technology is mainly from sodium cooled reactors, in a few countries like France, Japan, UK, USA, Russia and India. Two types of materials, the Stainless Steels (SS) and the nickel-iron based alloys, were evaluated in 1970s for core applications, based on their excellent high temperature mechanical properties. Subsequent efforts focused on stainless steels, since nickel based alloys under irradiation exhibited high tendency for helium embrittlement. Three generations of steels have been developed: variants of austenitic Stainless Steel (SS) like 316 SS and D9 (Ti modified 15Ni-15Cr austenitic steels), the 9-12 Cr based ferritic steels and the Oxide Dispersion Strengthened (ODS) advanced ferritic steels.

STRUCTURAL MATERIALS:

Apart from the above crucial components like the fuel sub-assemblies, there are many components in the core. These include core support structures, main vessel, inner vessel, intermediate heat exchangers, near core components like the grid plate, to mention a few. The operating conditions of these components are not as severe as the fuel sub-assemblies, clad and wrapper. The operating temperatures, temperature gradients, irradiation levels and sodium interaction are all less. However, these are permanent structures of the reactors, expected to serve for about the life of the reactor, which nowadays is expected to reach 100 years. The materials of these components called as 'structural materials' are normally different types of special steels. The major selection criteria of structural steels are shown in Table.3.

Table.3. Criteria for Selection (Reactor Structural)

- Good high temperature mechanical properties at plant operating conditions
 - creep
 - low cycle fatigue
 - creep-fatigue interaction
 - high cycle fatigue
- Compatibility with liquid sodium
- Structural stability
- Availability of design data
(RCC-MR / ASME design codes)
- Weldability
- Workability
- International experience
- Cost

The materials for out-of-core and balance of plant circuits will not be discussed in this chapter. The core component materials, clad and wrapper, alone will be discussed.

With the above introduction to the definition of different types of engineering materials in a fast reactor fuel assembly, the next part of the chapter would focus on introducing the behaviour of core component materials..

BEHAVIOUR OF CORE COMPONENT MATERIAL:

In a fast reactor core, the materials can change its shape and volume by tens of percentages, increase hardness by factors of five or more, reduce ductility and increase embrittlement. To give an example, it is possible for a cube of nickel to increase its side from 1 cm to 1.06 cm, after an exposure to irradiation of 10^{22} n/cm². This is nearly 20 % isotropic increase in volume due to ‘void swelling’ !!

Another feature is the ‘irradiation growth’. A cylindrical rod of uranium, say 10 cm long 1 cm in diameter, (volume - 7.85 cm³) irradiated to a fluence of 10^{20} n/cm², can increase its length three times, 30 cm and reduce its diameter to 0.58 cm, retaining the volume same as 7.85 cm³. The irradiation growth, anisotropic in nature, introduces severe distortion in core components.

The third feature is the irradiation hardening. Exposure of metals under irradiation at temperatures $< 0.3 T_m$, changes the stress- strain behaviour significantly. The yield strength of the material increases, concomittant with the significant reduction in ductility. These changes are introduced by the steady state defect microstructure of the material under irradiation.

Irradiation creep, the most important parameter for design consideration is the augmentation of thermal creep of the material, under irradiation. This leads to premature failure of the material and restricts the service life. The mechanisms that contribute to

reduction in creep life are related to the interaction of point defects with other defects in the material.

Irradiation embrittlement, another frequent observation in materials exposed to irradiation refers to the increase in the 'Ductile to Brittle Transition Temperature (DBTT)' during irradiation. In certain materials, this phenomenon makes the material so brittle at room temperature that the post-irradiation handling of core component materials becomes difficult. Some of the engineering materials contain nickel, an element which undergoes (n,α) reaction, producing high concentration of helium, which also causes embrittlement called the 'helium embrittlement'.

The damage mechanisms operating in a material under irradiation in a fast reactor fuel assembly can be summarized as follows:

- (1) Void Swelling
- (2) Irradiation Growth
- (3) Irradiation Hardening
- (4) Irradiation Creep
- (5) Irradiation Embrittlement and
- (6) Helium Embrittlement.

All the above degradation mechanisms are related directly to the production, diffusion and interaction of point defects in the material. Hence it is essential to understand the key features of point defects before proceeding to the degradation mechanisms.

BASICS OF POINT DEFECTS:

We know that the atoms in a crystalline lattice occupy their equilibrium positions, under normal circumstances, as shown in Figure.4.

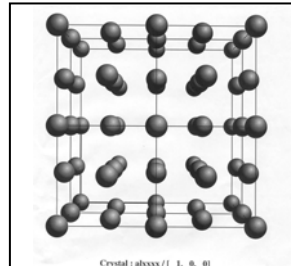


Figure.4. Periodic arrangement of atoms in a crystalline 'host' lattice.

When a stream of high energy neutrons 'bombard' this lattice, the atoms are 'knocked off' from their equilibrium locations. For the sake of simplicity, let us assume that only one high energy neutron, or in a general case, a particle is impinging on one lattice atom. The amount of energy transferred by the incident particle to the host lattice atom is related to the masses of the incident particle (m_1), the host lattice atom (m_2) and the incident energy (E_0). Based on conservation of kinetic energy before and after collision, the energy transferred to the host lattice atom can be shown to be

$$E = E_0 \left\{ \frac{4(m_1 \times m_2)}{[m_1 + m_2]^2} \right\} \quad \dots\dots 1.1$$

Usually in crystalline lattices, the energy required to 'knock off' an atom from its equilibrium position is of the order of few tens of eV's. For example, the 'displacement energy' in a lattice of copper is around 35-40 eV. The remaining energy which is transferred by the incident neutron manifests as kinetic energy of the host lattice atom. Thus, the collision of the neutron with a first lattice atom (Primary Knockon Atom- PKA) leaves behind another host lattice atom, with high kinetic energy to 'knock off' another lattice atom, by the same process of collision. The only difference is that the energy transferred by the projectile of host atom to another static lattice atom is far higher,

since mass of an atom is order of magnitude more than neutron. This process repeats till the mobile atoms have transferred all their kinetic energy and have come to rest or ‘relax’ in available locations, which need not be lattice site. The process is called the ‘displacement cascade’. The first atom which gets knocked off is called the primary knock on atom and the subsequent displaced atoms are called the secondary, tertiary knock-on atoms respectively (Figure.5.).

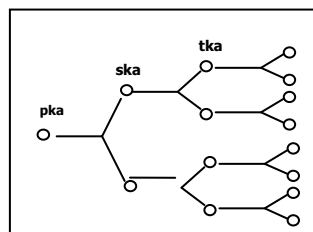


Figure5. Generation of primary knock on atom by the impingement of energetic incident neutron, followed by creation of secondary and tertiary knock on atoms in subsequent collisions.

How many lattice atom displacements will occur if a neutron of energy E_i strikes a lattice atom ?

The answer to this question will enable us to quantify the displacement damage. It is basically required to find out the no of atoms displaced by a PKA of energy E . It is done as follows:

To quantify displacement damage, it is necessary to set up the damage rate equation and solve. A brief approach is given below:

The displacement rate can be written as

$$R_d = N \int \phi (E_i) \sigma_D (E_i) dE_i \quad \dots\dots 1.2$$

Wherein N is the density of lattice atoms, ϕ the energy dependent particle flux and σ_D the displacement cross-section.

$$\sigma_D (E_i) = \int \sigma (E_i, T) \gamma (T) dT \quad \dots\dots 1.3$$

Where $\sigma(E_i, T)$ is the probability that particle of energy E_i imparts recoil energy to a struck lattice atom and γ is the parameter to be evaluated, namely number of displaced atoms during collision. While solving these equations using appropriate models, it is possible to show that

$$\gamma(T) = 0 \text{ if } E_t < E_d \quad \dots\dots 1.4$$

$$1 \text{ if } E_d < E_t < 2E_d,$$

$$E_t/2E_d \text{ if } 2E_d < E_t < E_c$$

$$E_c/2E_d \text{ for } E_t \gg E_c.$$

The resulting behaviour can be summarised as shown in figure.6.

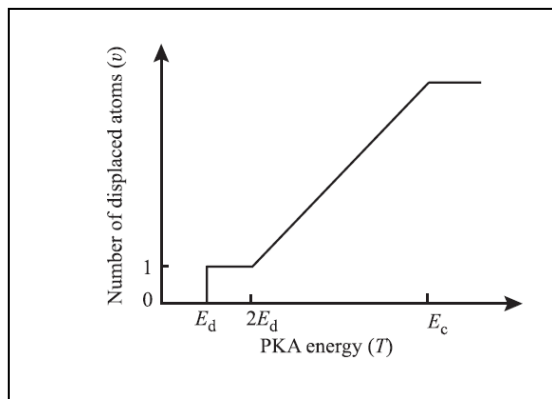


Figure.6. Variation of number of displacements in a lattice with the energy of the primary knock on atom(PKA).

If the energy of the PKA is less than the displacement energy of around 20-40 eV in most metals, no atom is displaced. The small energy that is transferred manifests as increased vibration of the lattice atom about its mean equilibrium position and hence as ‘heat’. When the energy transferred is between E_d and $2E_d$, two possibilities emerge: the struck atom gets displaced and the PKA with energy $< E_d$, jumps back into the lattice site. Or if the original PKA does not transfer E_d , there is no displacement. Either way, there can only be one displacement possible. The number of displacements can range from 0 to

$E_f/E_d - 1$, and for large E_f , no of displacements reach E_f/E_d . The displacement function is given by the above function and the behaviour is depicted in figure.6. For most metals, the value of E_d is around 20 or 40 or 90 eV.

There are four major stages in which the cascade evolves during irradiation:

1. Collisional
2. Thermal Spike
3. Quenching
4. Annealing.

In the collisional stage, the primary recoil atom initiates a series of displacive collisions that continues till no atom has enough energy to displace any more atom. This stage lasts for about 1 ps. At the end of this stage, the lattice ends up with energetic displaced atoms and vacant lattice sites. This time is not sufficient for the formation of stable lattice defects.

In the second stage, the 'thermal spike', lasting for another ps., the collisional energy of the displaced atoms is shared amongst the neighbouring atoms. This leads to formation of a region of high energy density leading to a spike of about 1ps., and the position of atoms in the spike region would resemble that of molten metal.

As there is a continuous, simultaneous transfer of energy to the adjacent atoms, the spiked region gets quenched or relaxed, in about 10 ps., leading to a stage close to thermodynamic equilibrium. Stable lattice defects like point defects or the defect clusters like loops form during this stage.

The annealing stage, as the name suggests is a thermally activated process. Further rearrangement of defects occur by thermal diffusion of mobile lattice defects. This is a stage which depends on the irradiation condition, temperature, and defect kinetics with the matrix.

The above processes leads to the creation of large number of vacant lattice sites (since many atoms are ‘knocked off’ from their equilibrium sites), called the ‘vacancies’ and host atoms coming to ‘halt’ at sites, which are not equilibrium lattice sites, called the ‘interstitials’. Normally, the concentration of vacancies produced during irradiation at room temperature is $\sim 10^{-2}$, which is two orders of magnitude higher than thermal equilibrium concentration of vacancies near melting point of say, copper $\sim 10^{-4}$. The core of a displacement cascade is rich with vacancies and peripheral area with interstitials. A typical disorder introduced in a perfect lattice in a displacement cascade is shown in figure.7. These defects, the vacancies and the interstitials, are called the point defects.

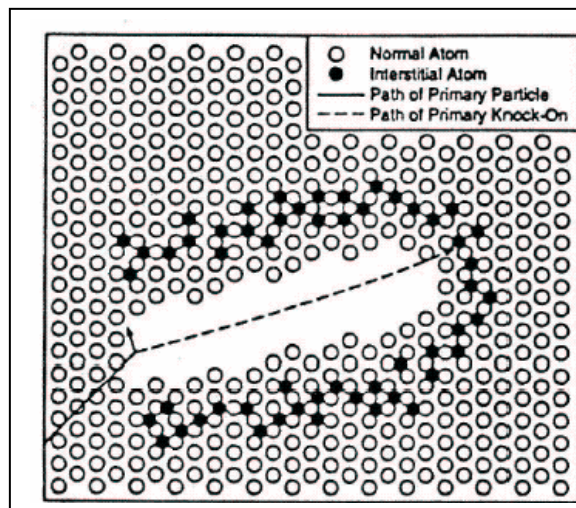


Figure.7. Atom position at the end of a single ‘displacement cascade’ showing the vacancy rich core and the interstitial rich peripheral regions.

The defects that are generated in a crystalline lattice after irradiation are

- Point Defects (0 D) : vacancies and interstitials
- Line Defects – (1D) : dislocation lines
- Planar Defects – (2D) : dislocation loops
- Volume Defects (3D) : voids, bubbles,

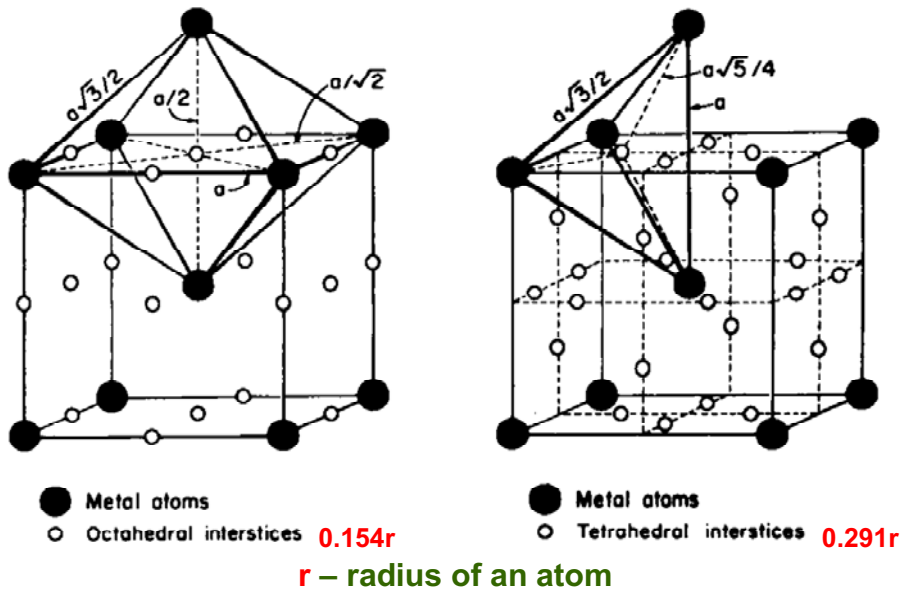
Point defects are the basic entity.

The concentration and diffusion characteristics of point defects determine the changes in physical and mechanical properties of materials.

Point Defects:

Interstitial: An interstitial is an atom which occupies a position in a crystal lattice, which does not belong to regular lattice site. There are two types of interstitial sites: Octahedral and tetrahedral. Octahedral sites are interstitial positions that are surrounded by an octahedron, whose six vertices are occupied by lattice atoms. Similarly, the tetrahedral sites are interstitial positions that are surrounded by a tetrahedron, whose four vertices are occupied by the lattice atoms. Figure...8 shows the octahedral and tetrahedral sites in a fcc and bcc lattice.

Vacant site - BCC



Vacant site - FCC

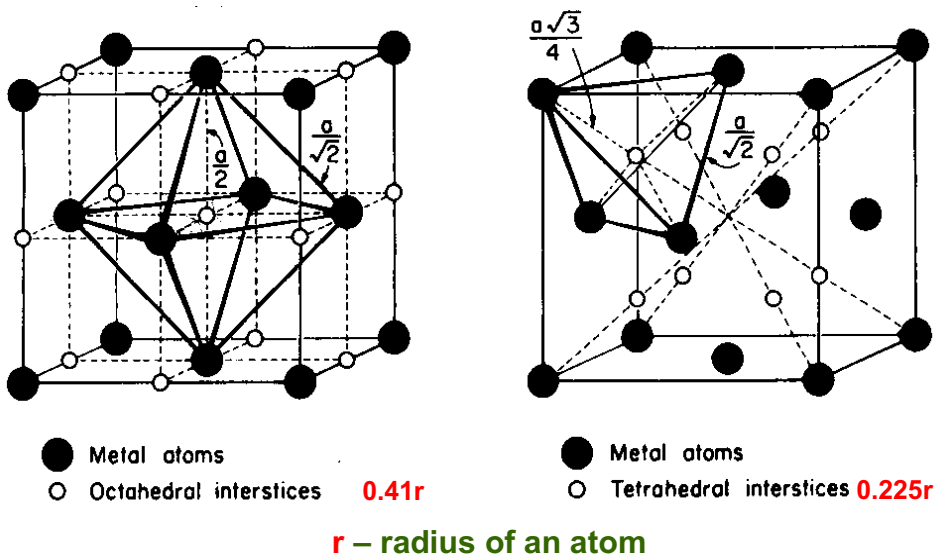


Figure.8 Octahedral and tetrahedral sites in fcc and bcc lattices.

This is too simple a picture of an interstitials. Most often, two Self Interstitial Atoms (SIA's) share a single interstitial site, forming a dumb-bell. The formation of such dumb-bells displace the atoms around them from their equilibrium lattice site, resulting in an

elastic displacement field around the interstitial. The total amount of distortion of the original lattice due to the formation of such interstitials or dumb-bells is called the 'relaxation volume'. The value of the relaxation volume for interstitials in metal lattices range from 1 to 2 atomic volume. The high distortion or the elastic interaction around interstitials is responsible for the interstitials to attract mobile SIAs. It is difficult to form interstitials, as clear from the high formation energy of around 2 to 3 eV in most metals. This leads to very low equilibrium concentration of interstitials near melting point, $\sim 10^{-18}$ to 10^{-6} . Interstitials are highly mobile, with the migration energy of around < 0.1 eV. The interstitials have high binding energy of around 1 eV. Hence, it is difficult to dissociate large interstitial clusters, at low temperatures.

Vacancies:

The vacancy or a missing lattice atom is the simplest defect in a lattice. Vacancies, unlike interstitials, have low formation energy ($\sim 1-2$ eV), low relaxation volume (0.1 to 0.5 atomic volume) and high migration energy (> 0.5 eV). Hence, vacancies are less mobile than SIA's. Multiple vacancies have less binding energy (0.1eV) and hence vacancy clusters are less stable than clusters of SIA's. Vacancies can bind to oversized solute or an oversized impurity atom (0.2 to 1 eV), to lower the overall free energy of the system.

Unlike interstitials, vacancies are thermodynamically stable defects. Even in a perfect crystal, in the absence of irradiation, presence of vacancies is thermodynamically desirable. It is possible to show that the free energy of a crystal, F is reduced with vacancies.

$$F \sim G = U + pV - TS = H - TS \quad \dots\dots\dots 1.5$$

Where F is the free energy of the crystal of volume V at constant pressure, p at temperature T , U the internal energy, S the entropy and H the enthalpy of N atoms constituting the system.

$$\nabla S_{\text{mix}} = k \ln w \quad \dots\dots\dots 1.6$$

Where w is the number of possible configurations.

Consider the introduction of ‘ n ’ defects with N available sites in the crystal. The increase in free energy is

$$\nabla G_f = n \nabla H_f - T \nabla S \quad \dots\dots\dots 1.7$$

∇H_f is the increase in the internal energy due to introduction of defects and ∇S is the corresponding increase in entropy. It can be shown that

$$w = \{ [N (N-1) (N-2) \dots (N-n+1)] / n ! \} \quad \dots\dots\dots 1.8$$

∇S_{mix} is calculated using the expression for w in the equation for ∇S_{mix} . The introduction of defects introduce vibrational disorder, which can be calculated and finally the total ∇S , by adding both contributions.

In equilibrium n will be such that $dG_f/dn = 0$. This condition can be used in the equation for dG_f to arrive finally at the concentration of ‘defects’ as follows:

$$C = n / N = \exp (-\nabla G_f / kT) \quad \dots\dots\dots 1.9$$

The above general equation, can be now re-written for the two types of point defects, to get an expression for concentration of the point defects.

For vacancies,

$$C_v = \exp (-E_{fv} / kT) \times \exp (S_{fv} / k) \quad \dots\dots\dots 1.10$$

$$C_i = \exp (-E_{fi} / kT) \times \exp (S_{fi} / k) \quad \dots\dots\dots 1.11$$

Where the subscript f refers to formation term and v and i refer to vacancies and interstitials. ($E_f = \nabla H_f$,). It requires much less energy to form vacancies than interstitials, ($E_{fv} < 1\text{eV}$ and $E_{fi} = \sim 4\text{eV}$), $C_v \gg C_i$, when the system is at thermal equilibrium.

The diffusion of these defects, through the lattice occurs at different rates by various mechanisms.

Defect Dynamics in a Crystal under Irradiation:

When the crystal is subjected to irradiation, it is already shown that Frenkel pairs (pair of vacancy and interstitial) are produced, whose concentration is two orders of magnitude more than that in thermal equilibrium. These defects are either lost through recombination of vacancies and interstitials or to the sinks like loops, voids. Thus, the local concentration of defects, either vacancies or interstitials, is a balance between (1) local production rate, (2) reaction with other defect species and (3) diffusion into or out of the region of interest. The main point defect reactions considered are



Where, \square is a lattice site. Point defect reactions with sinks, written as



The chemical rate equations representing the above situations can be written as

$$dC_v / dt = K_o - K_{iv} C_i C_v - K_{vs} C_v C_s \dots\dots\dots 1.14$$

$$dC_i / dt = K_o - K_{iv} C_i C_v - K_{is} C_i C_s, \dots\dots\dots 1.15$$

where C represents the concentration of either vacancy, v or interstitial i or sink, s and K's represent the rates of either production K_o or K_{iv} or K_{is} the reaction rate between vacancy and interstitial or interstitial/vacancy and sink.

It is possible to solve the above equations to arrive at the time evolution of each type of defects, namely concentration of vacancy or interstitial, for different combination of temperature and sink densities.

The defects explained above finally rearrange themselves by a combination of all the above mechanisms in the four stages into a lattice containing a high concentration of point defects, dislocation loops, voids and bubbles. The response of such a lattice to external engineering stresses any component is subjected to in a reactor is vastly different from the normal material.

The five damage mechanisms, introduced earlier, void growth, irradiation growth, irradiation hardening, irradiation creep and irradiation embrittlement are mainly caused by the point defects, linear, planar and volume defects introduced during irradiation. Each of the damage mechanism will be dealt with in the second chapter.