Irradiation Damage in Nuclear Power Plants

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Abstract

The chapter gives an introduction into the main processes occurring in metals and alloys under neutron irradiation. Displacement damage, phase reactions, swelling, irradiation creep, and transmutation are the main physical effects changing mechanical properties and microstructure of materials used in nuclear power plants. As results radiation hardening/embrittlement, enhanced stress corrosion cracking, changes in geometry, degradation of creep properties, and other damage can occur. Consequences of such material degradation are discussed for current nuclear plants where a 50-year operation experience exists. Advanced, future nuclear plants are expected to consist also almost exclusively of metals and alloys, and they are expected to undergo in principle the same types of damage. However, changes in other operational parameters (higher temperatures, fast neutrons, other coolants) might change also the degree of radiation damage (e.g., thermal creep in addition to radiation creep). Advanced modeling and testing techniques can be considered as a tool to balance the missing long-term experience with next-generation nuclear plants.

Introductory Remarks

Central components of nuclear plants are usually exposed to the coolant, radiation, and elevated temperatures. These conditions lead to degradation of components during service and limit therefore the lifetime of plants. In current light water reactors (LWRs), embrittlement of the reactor pressure vessel, irradiation-assisted stress corrosion cracking of reactor internals, and irradiation creep of claddings are typical degradation mechanisms caused by neutrons. Advanced reactors, like Generation IV plants, are expected to be exposed to more damaging fast neutrons (higher energy), higher temperatures, and coolants different from water. Although the physics behind radiation damage is expected to remain the same, some differences in damage development between LWRs and advanced reactors can be expected.

In the first part of this chapter, the basic phenomena of irradiation damage of reactor materials will be discussed. The focus will be on metals and alloys which are not only the key materials in current reactors but which will stay also key materials for future reactors. Considerations for ceramics will remain limited to graphite (as moderator for British advanced gas reactors and future high-temperature gas-cooled reactors) and to SiC/SiC composites which are considered for control rod parts or claddings in advanced plants.

In the second part of this chapter, radiation damage occurring in current nuclear plants will be highlighted with examples from reactor pressure vessels, reactor internals, and fuel claddings. Expectations for future plants will be briefly touched upon. Advanced methods of materials science will be introduced as a tool for better understanding of irradiation-related risks in future reactors.

Phenomenology of Radiation Damage

Introduction

Radiation damage is basically the result of the interaction of energetic particles with matter. The consequences of these interactions which depend primarily on energy can be manifold. The considerations of this chapter will be limited mainly to neutrons and ions on the particle side and to metals and alloys and selected ceramics on the matter side. Only the most important effects will be discussed; for more details, I would like to refer the reader to the numerous very good textbooks in this field, e.g., Schilling and Ullmaier (1994), Ullmaier and Schilling (1980), and Was (2007). The types of damage are (Schilling and Ullmaier 1994):

- Elastic collisions where bombarding particles (neutrons, ions, electrons) transfer recoil energy T to the lattice atoms. If T exceeds the threshold energy T_{th} for displacement, a vacancy-interstitial pair (Frenkel defect) is created.
- Nuclear reactions where fast particles produce considerable concentrations of foreign elements within the material. In particular, the inert gas helium which is produced by (n,α) reactions plays an important role for the behavior of metals and alloys under fast-neutron irradiation.
- Electronic excitations are of only very limited importance for metals and the irradiation damage process considered here.

The results of these damage events have an impact on radiation-exposed components like reactor pressure vessels (RPVs) or reactor internals (including fuel claddings). A summary of the damage events in the material and its consequences for components is given in Table 1. The different effects and its influence on the materials will be discussed more in detail in the following.

Types of Radiation Damage

Displacement Damage

Displacement damage starts usually with a bombarding particle that transfers a recoil energy T by elastic collision to a lattice atom. If the recoil energy exceeds a material-dependent threshold energy for displacement, E_{th}, the atom jumps from its original site to an interstitial position creating a vacancy-interstitial pair which is called "Frenkel pair." If the recoil energy is significantly higher than E_{th} (e.g., in the case of fast neutrons), the atom firstly hit by the neutron, the "primary knock-on atom" (PKA) or "primary recoil atom" (PRA) is able to transfer energy by moving further into the crystal creating further Frenkel pairs and a so-called displacement cascade (see Fig. 1). When the energetic particle is heavy and energetic enough, and the material is dense, the collisions between the atoms may occur so near to each other that they cannot be considered independent of each other. In this case the

Table 1 Different types of radiation damage and resulting technical consequences (replotted from Hoffelner (2012))

Effect	Consequence in material	Kind of degradation in component
Displacement damage	Formation of point defect clusters and dislocation loops	Hardening, embrittlement
Irradiation-induced segregation	Diffusion of detrimental elements to grain boundaries	Embrittlement, grain boundary cracking
Irradiation-induced phase transitions	Formation of phases not expected according to phase diagram, phase dissolution	Embrittlement, softening
Swelling	Volume increase due to defect clusters and voids	Local deformation, eventually residual stresses
Irradiation creep	Irreversible deformation	Deformation, reduction of creep life
Helium formation and diffusion	Void formation (inter- and intracrystalline)	Embrittlement, loss of stress rupture life, and creep ductility

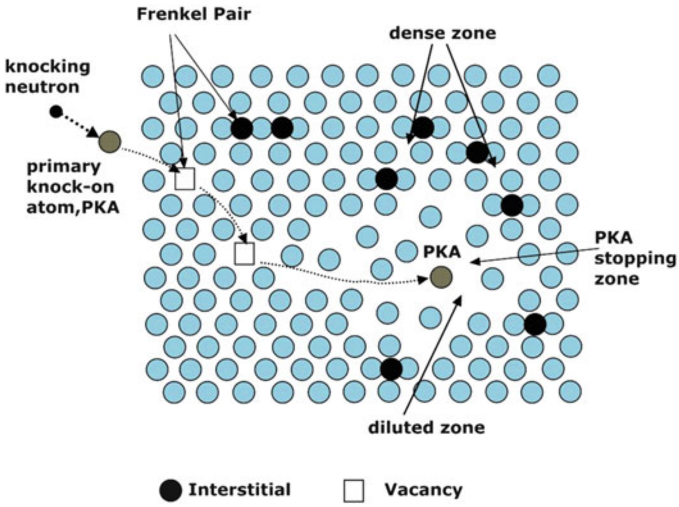


Fig. 1 Development of a collision cascade. The primary knock-on atom starts to move as a result of the energy transfer from the neutron. It creates Frenkel pairs, and it finally ends in a damaged zone with a diluted part where many vacancies exist and a dense part where many interstitials exist (Replotted from Hoffelner (2012), source Seeger (1962))

process becomes a very complicated many-body interaction between very many atoms which can only be treated with molecular dynamics modeling. A heat spike is created which is characterized by the formation of a transient diluted region in the center of the cascade and densified region around it. After the cascade, the densified

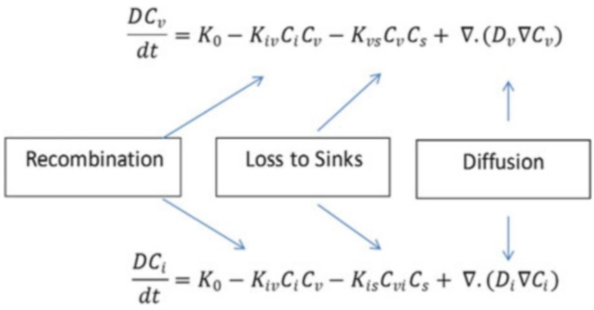


Fig. 2 Point defect rate equations

region becomes a region of interstitial defects, and the diluted region typically becomes a region of vacancy defects.

Elastic collisions produce radiation damage in three different process regimes at different timescales :

- The initial stage of the radiation damage process ($t < 10^{-8}$ s)
- Physical effects of radiation damage ($t > 10^{-8}$)
- Mechanical response of the material to radiation-induced effects

The initial stage of the radiation damage process has just been discussed. The phase of development of physical effects is mainly determined by diffusion and reactions of the created point defects. They can recombine, form agglomerations, or diffuse to sinks. The development of the concentrations of vacancies C_v and of interstitials C_i can be described by rate equations as shown schematically in Fig. 2. These rate equations can be solved for different boundary condition which allows a prediction of the development of the radiation-induced microstructure (Wiedersich 1991a, b). The supersaturation of point defects in irradiated matter leads to high diffusion coefficients already at lower temperatures. With increasing temperature the influence of irradiation on diffusion diminishes, and for temperatures higher than about 600 °C, thermal diffusion becomes the relevant diffusion process in steels. This can be seen from Fig. 3 (Zinkle et al. 1993) where the development of displacement-induced defects is shown as a function of temperature taking an austenitic steel as an example. Although there are some differences for different classes of metals and steels, the main message that point defects can agglomerate to different defects being obstacles for dislocation movement and that displacement damage vanishes with increasing temperature remains valid for several metals and alloys.

Units for Irradiation Damage

Before discussing further radiation-induced defects, a few aspects about the measure for damage relevant doses shall be mentioned. Very often particle flux which refers to the number of particles passing through an area in a certain interval of time (commonly measured in neutrons/cm^2·s) or particle fluence (neutron flux integrated over a certain time period, measured in neutrons/cm^2) are used. However,

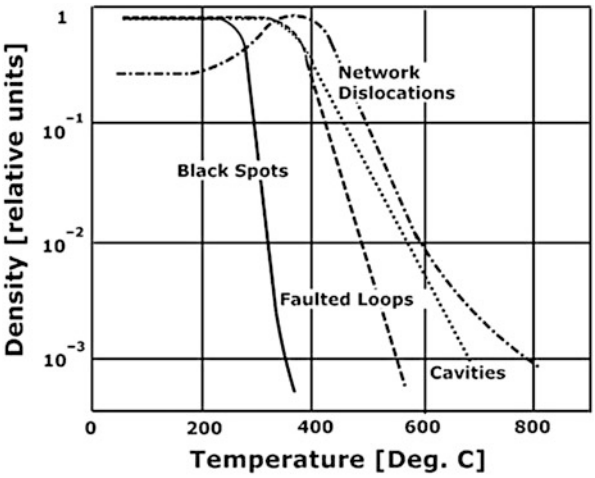


Fig. 3 Influence of irradiation temperature on the formation of different obstacles taking an austenitic steel as an example (Source Zinkle et al. (1993))

investigations of radiation hardening of annealed 316 stainless steel showed that even for the same type of material, different results were found when correlating radiation hardening (change in yield stress) with neutron fluence (Greenwood 1994). Therefore, another measure for radiation exposure or dose is frequently used which is based on the total number of displacements that the PKA will create in the solid. An important quantity is the number of displacements per unit time and per unit volume produced by a flux, $\Phi(E_i)$, of incoming particles of energy E_i. The displacement rate or number of displacements per atom (dpa) per unit time (dpa/s) describes in good approximation the energy-dependent response of an irradiated material. Typical displacement rates in reactors are 10^{-9}–10^{-7} dpa/s. Using this unit hardening for 316 stainless steel, mentioned before, could be very well correlated (Greenwood 1994).

Point Defect-Related Irradiation Damage Other Than Displacement Damage

In the previous section damage due to production, diffusion, and agglomeration of different point defects was considered. The high point defect supersaturation can also lead to other phenomena known from thermal diffusion processes like:

- Radiation-induced segregation
- Radiation-induced precipitation
  - Incoherent precipitate nucleation
  - Coherent precipitate nucleation
- Radiation-induced dissolution
- Radiation-induced phase reactions
  - Radiation disordering
  - Metastable phases
  - Amorphization

Radiation-Induced Segregation (RIS)

Thermal-induced segregation is a temperature-dependent redistribution of alloy constituents at point defect sinks such as grain boundaries. Temper embrittlement of steels is a very well-known example for segregation-related deterioration of toughness. Elements like phosphorus, sulfur, or manganese diffuse to grain boundaries. The cohesion along the grain boundaries is weakened leading to a reduction of toughness (reduction of fracture toughness or increase of ductile-to-brittle fracture appearance transition temperature). Such grain boundaries can also act as preferential corrosion sites leading to stress corrosion cracking as discussed later.

Radiation-induced segregation describes a similar effect, however, driven by radiation-induced point defects. A flow of vacancies into one direction is equivalent to material flow into the opposite direction. It can be understood in terms of the so-called inverse Kirkendall effect (Marwick 1978). This inverse Kirkendall effect refers to cases where an existing flux of point defects affects the interdiffusion of atoms of type A and atoms of type B. Irradiation segregation in a homogeneous AB alloy occurs because the irradiation has produced excess point defects which lead to a point defect flux. Figure 4 explains the mechanism for a binary alloy more in detail. The ordinates represent the concentrations of vacancies and interstitials, respectively, in arbitrary units. The x-axis gives the distance from the grain boundary. Movement of a vacancy into one direction is equivalent with the movement of an atom into the other direction. Therefore, the arrow of the vacancy flow J_v points in another direction than the arrows of the material flows J_A and J_B. In case of the movement of interstitial atoms, the directions of J_i and J_A and J_B are the same. The differences in the diffusion coefficients of A and B lead to a dilution of the

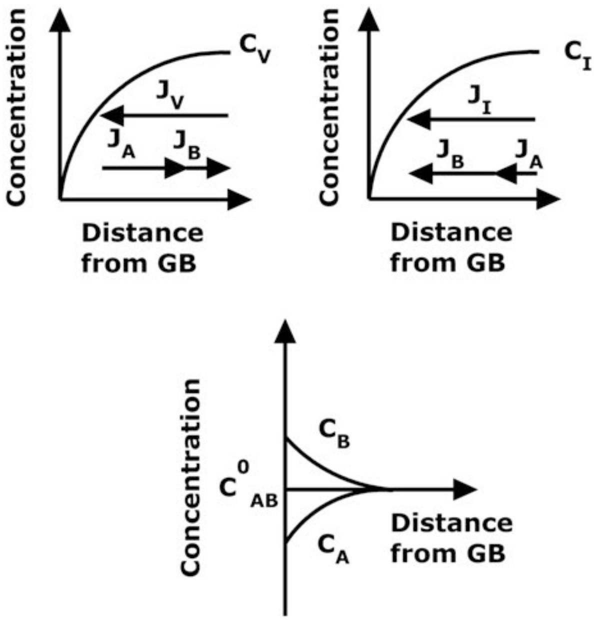


Fig. 4 Principle of radiation-induced segregation for a binary alloy (After Was (2007))

concentration of atoms A and to an increase of the concentration of atoms of type B towards the grain boundaries. As an example a diminishing chromium concentration combined with an increasing nickel concentration at the grain boundaries was found for proton-irradiated austenitic steel (304 SS) (Bruemmer et al. 1999a). Radiation-induced segregation depends on the temperature as well as on dose rate as a diffusion-driven effect. Once the temperature is too low, vacancies can move only slowly, and recombination will become the predominant mechanism. At temperatures where thermal effects become important, radiation effects become negligible. Radiation-induced segregation can therefore only happen in a temperature window between these two conditions (Was et al. 2006). Radiation-induced segregation plays an important role for irradiation-assisted stress corrosion cracking in light water reactors as will be discussed later.

Radiation-Induced Phase Transformations

Other diffusion-controlled irradiation phenomena are radiation-induced phase transformations or phase reactions which can result in precipitation of phases not expected at operation temperatures, dissolution of phases, and amorphization of phases. The driving force behind these microstructural changes is – like for RIS – the presence of large supersaturation of point defects, especially at temperatures between 250 and 550 °C or the inverse Kirkendall effect. Irradiation-induced point defect sinks like interstitial loops, helium bubbles, and voids can also give raise to precipitation.

Radiation-induced precipitation is one phenomenon belonging to this class of radiation damage. Coherent and incoherent precipitates can be formed. Coherent particles fully or partially match the lattice structure of the matrix, and incoherent particles don't. Coherent particles act as sinks for solute atoms, whereas incoherent particles allow solute atoms to be trapped and also to be released (Was 2007). Figure 5a shows an example of radiation-induced precipitates. In this figure also radiation-induced voids are visible. This phenomenon will be discussed in the next section under "void swelling." Radiation-induced nanosized precipitates

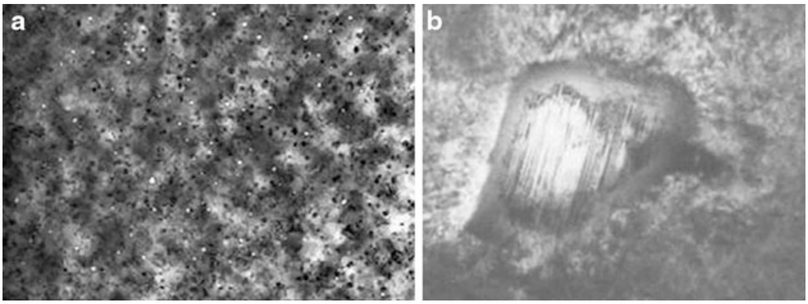


Fig. 5 (**a**) Concurrent void formation and $M_{23}C_6$ precipitation after irradiation at a PWR-relevant dpa rate of 1.8×10^{-7} dpa/s in the experimental breeder reactor II EBR-II in Argonne at 379 °C, after Isobe et al. (2008). (**b**): $Zr(Cr, Fe)_2$ precipitate neutron irradiated at a temperature of 510 K to a fluence of 8 dpa, showing formation of amorphous layer Motta et al. (1991)

(e.g., nanoclusters) are responsible for an increase in strength acting as obstacles for dislocation movement. This increase in strength reduces in turn ductility and toughness of the material which can be considered as life-limiting materials degradation as shown later for reactor pressure vessels.

Radiation-induced dissolution means that due to the presence of a high density of point defects, particles start to dissolve. This is rather similar to processes happening without irradiation during a solution treatment of alloys at high temperatures.

Amorphization: Amorphous metals do not have an ordered atomic-scale structure. They can be produced by very rapid cooling, and they are often referred to as metallic glasses. Amorphization can also occur during mechanical alloying or physical vapor deposition. Radiation-induced amorphization is a result of the radiation-induced high point defect density. Amorphization under irradiation not only happens for metals and alloys, it can also be found in intermetallics and ceramics like graphite or silicon carbide. Figure 5b (Motta et al. 1991) shows a partially amorphized second phase particle in a zirconium-based LWR cladding material (Zircaloy). Amorphization occurs often together with decomposition or dissolution of second phase particles in Zircaloys. The related change in matrix composition can improve the oxidation behavior of these materials in reactor environments.

Radiation-induced disordering can happen in an ordered lattice like an intermetallic phase or alloy when radiation-induced diffusion processes support lattice disordering already below temperatures where it would occur due to thermal reasons.

Metastable phases can form when thermal and radiation-induced phase formations are in competition and that phases appear at conditions where they should be thermally not stable or they disappear when they should remain thermally stable.

The Production of Foreign Atoms

Radiation-induced microstructural changes discussed until now happen at lower temperatures, and they disappear once the temperature exceeds about 600 °C. Radiation-induced production of foreign atoms (see, e.g., Schilling and Ullmaier (1994)) is another important type of radiation damage. Particularly interesting are reactions where gases are generated (e.g., alpha particles or protons) which can further react with the material. This is very important because gaseous atoms, especially helium (i.e., α), can seriously degrade the long-term mechanical integrity of some reactor components. This has already been recognized in the mid-1960s of the last century during the development of alloys for core components of fast breeder reactors (Barnes 1965; Harries 1966). Nuclear reactions where helium can be produced in metals (M) by fast neutrons (n^f) can be the following:

$$ {}^{A}_{Z}M + {}^{1}_{0}n^{\mathrm{f}} \rightarrow {}^{\mathrm{A}-3}_{\mathrm{Z}-2}M' + {}^{4}_{2}He \quad \text{(some MeV)} $$

$$ {}^{A}_{Z}M + {}^{1}_{0}n^{\mathrm{f}} \rightarrow {}^{\mathrm{A}-4}_{\mathrm{Z}-2}M'' + {}^{1}_{0}n^{\mathrm{f}} + {}^{4}_{2}He \quad \text{(some MeV)} $$

Nickel has the highest cross sections for such reactions, and the problems for fast reactors even increase for fusion reactors which will be exposed to 14 MeV neutrons.

Also thermal neutrons nth can lead to helium formation although to a lesser extent (lower cross sections). A typical reaction (2 steps) for thermal neutrons is the following:

$$^{58}_{28}Ni + ^{1}_{0}n^{\mathrm{th}} \rightarrow ^{59}_{28}Ni + \gamma$$

$$^{59}_{28}Ni + ^{1}_{0}n^{\mathrm{th}} \rightarrow ^{56}_{26}Fe + ^{4}_{2}He \quad (4.67\ \mathrm{MeV})$$

The problem with helium gas in the metal is that it can form intragranular bubbles as well as intergranular bubbles. Intergranular bubbles lead to strong reduction of creep ductility and sometimes also of creep rupture time. This is the reason why nickel-base superalloys, which are basically the high temperature materials of choice, cannot (or only limited) be used for in-core applications at high temperatures in fast reactors.

Influence of Radiation on Mechanical Properties

In the previous sections the basic principles of radiation damage were discussed. The microstructural changes have an effect on the macroscopic behavior of materials and consequently also on the performance of components.

Strength and Toughness

The presence of radiation-induced obstacles for dislocation movement (point defect clusters, dislocation loops, stacking fault tetrahedra, helium-filled pores) has an influence on the mechanical properties. Radiation hardening is generally accompanied by a reduction in uniform elongation under tensile test conditions due to highly localized plastic flow. A second consequence of radiation hardening that is particularly important for body-centered cubic (BCC) alloys is reduction in fracture toughness and a potential shift in the ductile–brittle transition temperature to values that are above the operating temperature. Figures 6, 7, and 8 show examples for irradiation hardening and embrittlement. Stress–strain curves after irradiation at different temperatures are shown in Fig. 6. The curves were shifted along the strain axis to make the results better visible. In comparison with the yield stress of the un-irradiated material, a significant increase (up to more than a factor of 2) was found. Impact tests (shown in Fig. 7) reveal a very pronounced shift in the brittle-to-ductile transition temperature, and also the upper shelf energy is significantly reduced. At temperatures above 400 °C hardening starts to disappear as a result of annealing. Operation of structural materials in the “lower shelf” fracture toughness regime is usually not feasible based on safety considerations, because this could lead to premature shutdown of the reactor before the design operating lifetime is achieved as discussed later. Embrittlement can also be seen from the temperature dependence of the fracture toughness as shown in Fig. 8.

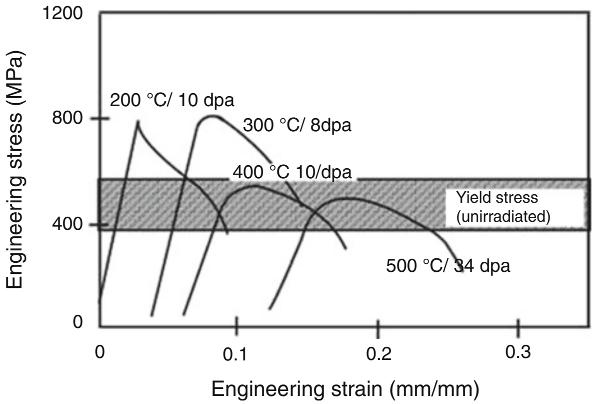


Fig. 6 Irradiation hardening in a ferritic–martensitic steel. At temperatures above 400 °C, hardening starts to disappear as a result of annealing (Replotted from Hoffelner (2012), source Robertson et al. (1997)

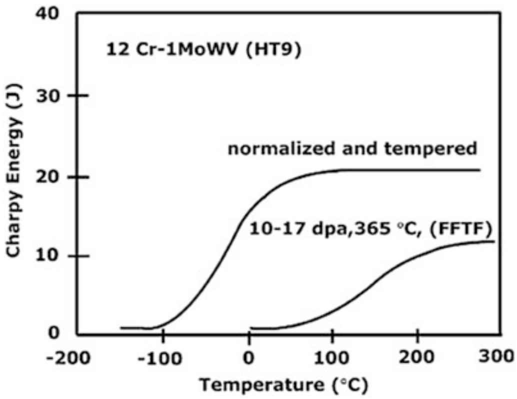


Fig. 7 Shift in the fracture appearance transition temperature as a result of irradiation embrittlement. FFTF: Fast Flux Test Facility, Hanford (Replotted from Hoffelner (2012), source Klueh and Alexander 1992)

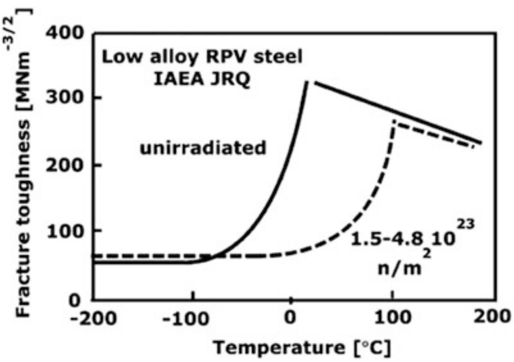


Fig. 8 Influence of neutron irradiation on the fracture toughness (Replotted from Hoffelner (2012). Source (Havel et al. 1993))

Radiation effects in ferritic–martensitic steels for temperatures where irradiation hardening/embrittlement occurs (T <= 450 °C) are well investigated. Analyses of embrittlement of steels which can be attributed to irradiation-enhanced precipitation are only scarcely available. An extended analysis of embrittlement in the absence of radiation hardening for different steels was reported in Klueh et al. (2008). In this investigation nine different irradiated steels (ferritic–martensitic, ferritic, low activation) were analyzed that were embrittled in the absence of irradiation hardening at temperatures exceeding 450 °C. The embrittlement was attributed to irradiation-enhanced precipitation. Precipitates that were concluded to cause the observed behavior varied for the different steels and included $M_{23}C_6$, α', χ, and Laves phase. The observed effects were explained by postulating irradiation-enhanced or irradiation-induced precipitation and/or irradiation-enhanced precipitate coarsening that produced large precipitates acting as crack nuclei for fracture initiation.

Influence of Irradiation on Fatigue and Fatigue Crack Growth

Irradiation increases the yield strength, and it decreases the ductility of metallic materials. This leads to an increase of the fatigue limit (as a result of higher strength) at high number of cycles. However, under low cycle fatigue conditions, the number of cycles to failure decreases as a result of radiation-induced loss of ductility as shown in Fig. 9 and as discussed, e.g., in Hoffelner (2012) in more detail.

Fatigue crack growth rates as a function of the cyclic stress intensity range ΔK remain usually up to temperatures where the environment has only a negligible effect more or less independent from the temperature. Also microstructure has no very pronounced effect, and therefore, no significant effect of irradiation on fatigue crack growth rates is expected. This could be confirmed for an austenitic steel as shown in Fig. 10 (Lloyd et al. 1982). Similar insignificant influence of irradiation on fatigue crack growth rates also reported low-alloy reactor pressure vessel steels (James and Williams 1973).

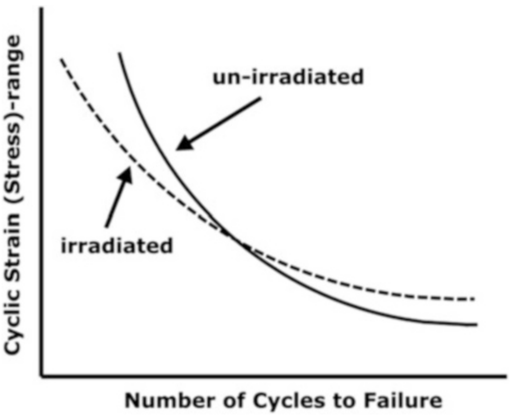
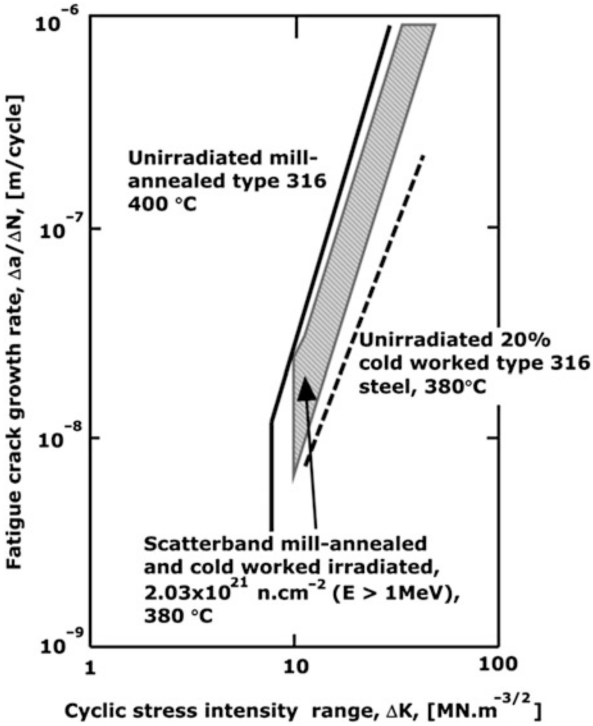


Fig. 9 The influence of irradiation on the fatigue curve (schematically). See also Hoffelner (2012)

Fig. 10 Fatigue crack growth rates for different qualities of type 316 austenitic steel in un-irradiated and irradiated conditions (Replotted from Lloyd et al. (1982))



Influence of Irradiation on Thermal Creep

Application of load at elevated temperatures under radiation can lead to two types of creep: thermal creep and radiation-induced creep which will be discussed later. Degradation of stress rupture life as a result of pre-irradiation has been reported, e.g., in Bloom and Stiegler (1972). The technical relevance of such data is – as far as only displacement damage is concerned – questionable because usually in a nuclear plant irradiation and thermal creep happen synchronously. Specific attention must be paid to helium at high temperatures. The presence of helium bubbles at grain boundaries is expected to contribute synergistically to creep damage forming also voids along the same sites. Helium bubbles at grain boundaries can therefore deteriorate stress rupture ductility as well as creep rupture strength. Some in-pile creep data for an austenitic steel are shown in Fig. 11 from which the influence of irradiation creep becomes clearly visible (Puigh and Hamilton 1987). An exhaustive treatment of creep-irradiation interactions for an austenitic steel can be found in the literature (Wassiliew et al. 1986). This temperature-dependent damage pattern is also reflected in creep–fatigue interactions.

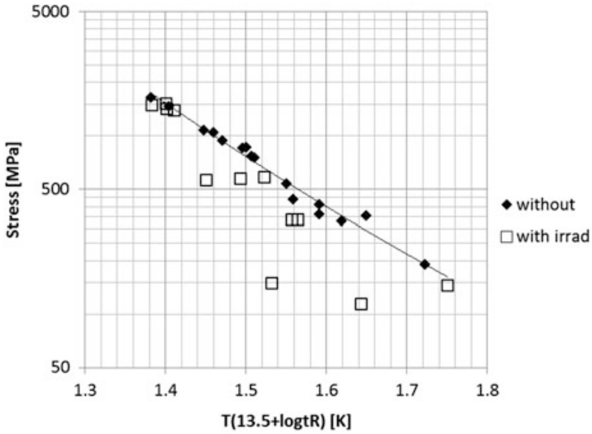


Fig. 11 Influence of irradiation on stress rupture performance of an austenitic steel. The influence of irradiation has been attributed to helium effects (Replotted from Hoffelner (2012), source Puigh and Hamilton (1987)

Radiation-Induced Dimensional Changes

Void Swelling

Voids or bubbles containing either vacuum (vacancy clusters) or gas (helium) can develop under irradiation in a crystal (see Fig. 5a as an example). According to Garner (2010a) one defining feature for discrimination between void or bubble is that bubbles tend to grow slowly by gas accumulation, while voids are either totally or partially vacuum filled, but which are free to grow rapidly via vacancy accumulation without further gas addition. Without going further into detail concerning the growth mechanisms, it is obvious that holes in a body usually increase its volume. Void swelling is the effect which leads to a three-dimensional change of the material during irradiation in a temperature interval of $0.3\ T_m <= T <= 0.5\ T_m$. Two phases must be considered for void formation: void nucleation and void growth (Russel 1971; Katz and Wiedersich 1971). The fact that voids form although its formation is energetically not very favorable is attributed to the fact that additional heterogeneities like very small helium gas bubbles are present during irradiation which promote clustering of vacancies. Void growth is quantitatively better understood than nucleation. In contrast to interstitials which tend to migrate to dislocations, the vacancies are rather attracted by voids. This net flux of vacancies to voids causes them to grow which leads macroscopically to swelling. Three stages can be discriminated: transient period, steady state swelling, and saturation. During the first period nucleated voids start to grow until a steady state is reached during which an almost linear relation between dose and volume swelling exists. With further increasing void size, the relative contribution of radiation-induced defects to macroscopic swelling decreases leading to saturation. The duration of the transient regime of swelling in austenitic and high-nickel steels is exceptionally sensitive to irradiation parameters, composition, heat treatment, and mechanical processing (Garner 2010a).

Irradiation Creep

Void swelling is a three-dimensional change of the volume which occurs without mechanical load. Superposition of radiation and mechanical load leads to deformation of the material at stresses far below the yield stress and at temperatures where thermal creep cannot be observed. An extensive review on irradiation creep phenomena is given in Garner (1994). For cold-worked and recrystallized austenitic steel, in-beam fatigue tests with hold times at 300 and 400 °C were performed, and a clear influence of the radiation was found which was attributed to irradiation creep–fatigue interaction (Scholz and Mueller 1996).

Figure 12 shows irradiation creep during helium implantation of the ferritic oxide dispersion strengthened (ODS) steel PM 2000 at 500 °C as an example. The response to thermal creep is also shown for comparison, and no indications for thermal creep can be seen.

Swelling and irradiation creep are not really separate processes. Both phenomena are caused by the presence of point defects as a result of radiation. While swelling attempts to be isotropic, irradiation creep redirects mass flow anisotropically. Irradiation creep can operate before the onset of swelling but is accelerated when swelling begins. Radiation creep is traditionally described in terms of transient contributions saturating at 1 dpa, stress-enhanced creep (proportional to void swelling), and the creep compliance B_0 in the absence of swelling. For many high exposure applications, the transient can be ignored (Garner 1994). Neglecting also possible effects related to the void swelling rate, the irradiation creep compliance B_0 remains the most important contribution. It can be written as:

$$\dot{\varepsilon} = B_0 \sigma K$$

which says that the irradiation creep rate $\dot{\varepsilon}$ is proportional to irradiation displacement damage rate K and to stress σ (at least for moderate stresses). It is interesting

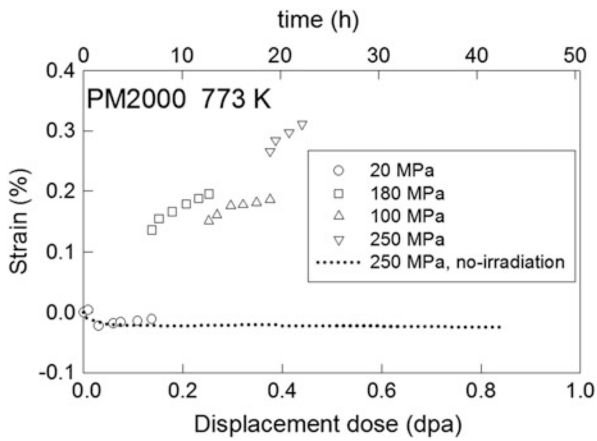


Fig. 12 Irradiation creep under helium implantation of a commercial ODS alloy Chen et al. (2008)

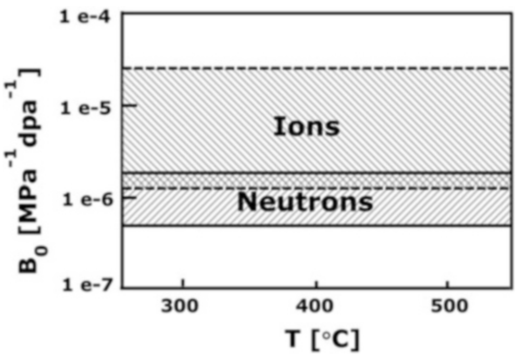


Fig. 13 Comparison of irradiation creep compliances B_0 as a function of irradiation temperature T. Ions refer to light ion irradiations (protons/helium) and represent the materials: ODS PM2000, 19Cr-ODS, ODS Ni-20Cr-1ThO2, and martensitic steel. Neutrons refer to neutron irradiations to doses below 25 dpa and above 25 dpa and represent the materials: ODS MA957, HT9, F82H, and Fe-16Cr (Sources Hoffelner (2012), Chen et al. 2010))

to notice that in this creep law, the stress exponent is 1 which is also the case for diffusion-controlled thermal creep. This is compatible with the fact that radiation creep is also a diffusion-controlled process. Irradiation creep is important for temperatures below which thermal point defects become predominant. This has been shown for austenitic and ferritic steels, and it was also found for advanced nuclear materials like ODS alloys or titanium aluminides. Some discussion is ongoing concerning the influence of type of the energetic particle on irradiation creep. Figure 13 compares irradiation creep compliances of several types of alloys. A typical value for the irradiation creep compliance of alloys under neutron irradiation is about 7.10^{-7} MPa^{-1}. dpa^{-1}. For light ions qualitatively a similar behavior was found; however, the average value was about five times higher. Possible reasons for this difference could be:

- Real influence of the type of radiation
- Radiation rate effect (because light ion irradiation is usually performed typically with 0.1 dpa/h compared with 0.003–0.004 dpa/h in a fast reactor)
- Dependence on total dose (ion irradiation tests go usually up to 1–2 dpa only)
- Influence of state of stress (multiaxiality)

Even if a quantitative explanation is still missing, it should be pointed out that the qualitative results are the same. This means that creep tests under ion irradiation allows a relative comparison between different materials which is very important for material development. Although the phenomenology of irradiation creep is quite consistent, a thorough physical understanding is still missing.

Radiation Damage in Nonmetallic Structural Materials

Graphite

Graphite is of concern for some reactor types like the British AGR or the high-temperature gas reactor. Therefore, graphite has been frequently investigated, and the mechanisms of the damage which graphite undergoes on neutron irradiation are quite well understood (IAEA 2000). However, many processes have not been correlated with the properties of the pristine graphites. In other words, the behavior of a new graphite cannot be quantitatively predicted. Certain behaviors may be anticipated, but this is an insufficient basis for a designer. This is the reason why worldwide projects on irradiation damage of graphite are underway. The basic radiation damage mechanisms for graphite are comparable with metals. A displacement cascade creates vacancies and interstitials which rearrange in the graphite lattice forming interstitial loops and vacancy loops (Ball 2008; Burchell 1999). The essential processes which happen in graphite under irradiation are the following (Fig. 14). As a result of vacancy creation and formation of vacancy clusters, the crystal undergoes an a-axis shrinkage. In contrast to this shrinkage, agglomeration of interstitials leads to an expansion along the c-axis. At irradiation temperatures $T_{irr} < 400\ ^{\circ}C$, damage accumulates rapidly (lack of vacancy mobility), and the crystal changes start to interact with the porosity. At high temperatures ($T_{irr} > 300\ ^{\circ}C$) shrinkage with turnaround to swelling at higher doses is observed. This turnaround into volume swelling due to incompatibility of crystal strains causes new pore generation. The radiation-induced microstructural changes lead not only to swelling and shrinking, but they also affect the physical properties of graphite. Thermal creep in graphites is negligible at temperatures up to ~2,000 °C. Irradiation creep is significant at all temperatures. Application of external load leads to irradiation creep of graphite similar to metals Fig. 15. Without external stress the graphite follows the "unstressed" line which shows shrinkage converting to

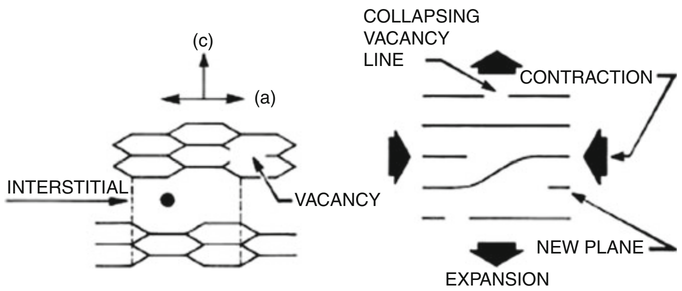


Fig. 14 Dimensional changes of graphite as a result of point defect reactions (Courtesy Burchell TD ORNL, Ball (2008), Burchell (1999))

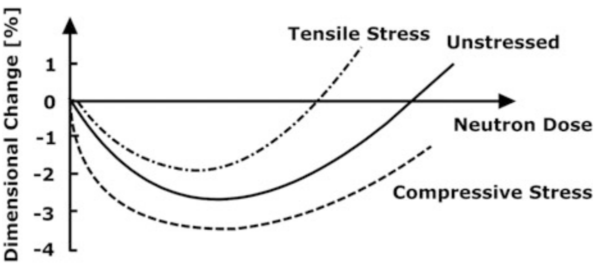


Fig. 15 Irradiation-induced dimensional change in stresses and in unstressed graphite (Courtesy Burchell TD ORNL, Ball (2008), Burchell (1999))

swelling with increasing radiation. Addition of a tensile load enhances swelling, whereas addition of a compressive load diminishes swelling.

Silicon Carbide

Fiber-reinforced materials like SiC/C or SiC/SiC are candidates for structural applications in fusion as well as advanced fission plants. They were mainly investigated with respect to fusion (Ozawa et al. 2010). Silicon carbide shows different types of radiation damage depending on temperature:

- Amorphization (up to about 200 °C)
- Point defect swelling (between 200 and 1,000 °C)
- Void swelling (above 1,000 °C)

Significant improvements with respect to resistance against irradiation could be made for SiC fibers. Also strong improvements of the matrix could be achieved with advanced compaction techniques. Indications exist that the strength of irradiated advanced fiber material could remain unchanged up to at least 10 dpa and perhaps higher. Further advances will likely require tailoring the interface swelling characteristics to compensate for differential swelling between the fiber and matrix. An exhaustive review of state of the art in ceramics for nuclear applications can be found in Katoh et al. (2007). Although this report is entitled as “Assessment of Silicon Carbide Composites for Advanced Salt-Cooled Reactors,” it is a broad review of literature and results on radiation damage of SiC/SiC covering particularly fusion developments. For some advanced reactor applications like control rod or structural parts of a VHTR, the radiation damage of commercially available (German MAN today MT Aerospace AG, German DLR) ceramic composites (SiC/SiC, SiC/C) was investigated. Irradiation was performed in the SINQ neutron spallation source of PSI (2013) (up to 27 dpa, 2,300 appm He, up to 550 °C) (Pouchon et al. 2011). Under these conditions the chemical vapor-infiltrated (CVI) SiC with amorphous carbon fibers showed the best radiation resistance (almost no loss of strength). The surprisingly inferior behavior of SiC/SiC might be attributed to the fact that in the material investigated no radiation optimized SiC fibers were used (Pouchon et al. 2011).

Irradiation Damage of Components

Light Water Reactors

Pressure Vessels

Light water reactor pressure vessels are made of low-alloy steel with the inside covered with austenitic steel cladding (against corrosion) and weldments of flanges and penetrations. The aging behavior of the RPV is particularly important because of its enormous safety relevance. Low-alloy steels exhibit a brittle–ductile temperature transition. Above a characteristic temperature RPV steels are tough which means that they have a relatively high fracture toughness. Below this characteristic temperature the fracture toughness is low and fracture is dominated by cleavage. Embrittlement is characterized by an increase in the ductile-to-brittle transition temperature as well as a reduction in the fracture toughness in the ductile fracture regime. The lower fracture toughness for the embrittled material reduces the allowable (critical) crack length and therefore reduces the safety margin as shown in Fig. 16. The lower line refers to the real crack length and how it develops with time. The upper line refers to the critical crack length at which the component fails. The critical crack length is not a constant because effects like thermal embrittlement or thermal aging can reduce the fracture toughness and therefore also reduce the critical crack length. Nondestructive evaluation performed in intervals determined by the expected subcritical crack growth rates (ISI) and condition-based monitoring are therefore extremely important safety measures. Main parameters for irradiation damage of RPVs are material and its chemical composition, temperature, neutron flux, energy spectrum of neutrons, irradiation time, and neutron fluence.

Radiation damage of light water reactor pressure vessels has been summarized and thoroughly reviewed in Odette and Lucas (2001), Hashmi et al. (2005), and

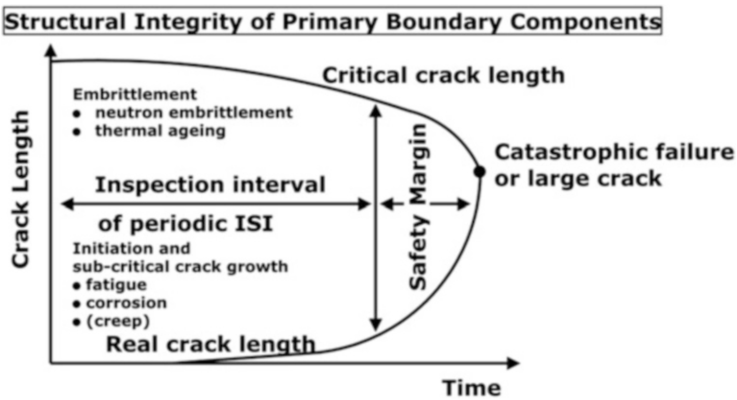


Fig. 16 Schematic of damage development in nuclear plants; ISI means "in-service inspection" (Source Bakirov (2010))

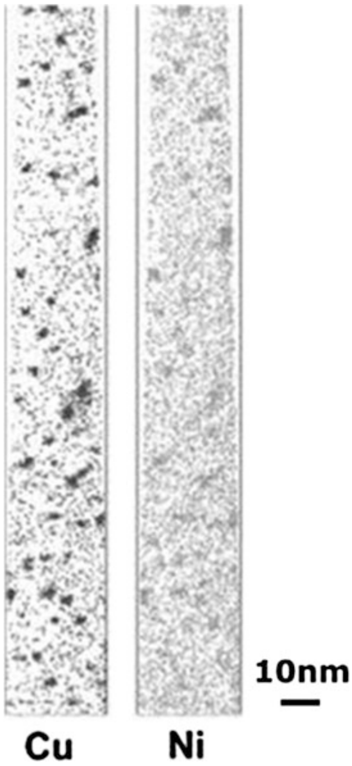


Fig. 17 Atom probe tomography (APT) image of Cu–Ni nanocluster in irradiated RPV-steel (Source (Miller and Russel 2007))

Steele (1993). It is typically caused by displacement damage and irradiation-induced nano-precipitates:

- Displacement damage: Point defect clusters and loops acting as pinning points for dislocations increasing the strength and decreasing ductility
- Radiation-induced phase transformations: Precipitation of Cu nanoclusters (see Fig. 17) or "manganese–nickel-rich precipitates" (MNPs) or "late blooming phases" (LBPs) which additionally contribute to hardening and embrittlement

Copper impurity has long been recognized as the dominant detrimental element in reactor pressure vessel (RPV) steels at copper levels in excess of about 0.1 wt%. The formation of copper-rich precipitates gives rise to considerable hardening and embrittlement at levels of neutron fluence well below the design end-of-life (EoL) of RPVs of operating nuclear power plants. The role of copper content on the embrittlement of RPV steels was thoroughly investigated with small angle neutron scattering (SANS) and tensile tests (Bergner et al. 2009). From the 1990s there has been increasing evidence of clusters enriched with manganese and nickel appearing in low-Cu steels (Cu 0.1 wt%).

The terms "manganese–nickel-rich precipitate" (MNP) or "late blooming phase" (LBP) (Odette and Wirth 1997) emphasize different aspects of this phenomenon. MNPs were first predicted by thermodynamic arguments (see, e.g., Odette and

Wirth (1997, Odette and Lucas (1998)) and then confirmed by means of several experimental techniques including atom probe tomography (APT) and positron annihilation spectroscopy. The vessel walls in the reactor beltline regions are subjected to the highest fluences and degradation due to irradiation embrittlement. Therefore, the welds within that region become possibly the weakest link since the welds are likely to contain defects that can become cracks. Additionally, the higher copper (and nickel) content in many of the older vessel welds has led to much higher radiation damage sensitivity. The base metals should not be ignored, since the copper content in older plates and forgings was not controlled to a minimum level; however, there appears to be less irradiation embrittlement in base materials as compared to welds with the same copper/nickel concentrations. A quantitative understanding of irradiation embrittlement of the reactor pressure vessels is extremely important for assessments of residual safe life of a nuclear plant. Condition-based monitoring of degree of damage belongs therefore to key tasks in several plant life extension concepts.

Surveillance specimen can help to assess the degree of radiation damage of an RPV. Those samples were built into the vessel at locations where they were exposed to neutron damage. After removal the samples could be further analyzed (Charpy tests, metallographic studies, etc.) for an assessment of the condition of the RPV. Fracture toughness can be assessed from shifts of the ductile-to-brittle transition temperature using the "master-curve""approach." It is based on the observation that at least for low-alloy steels, a good relationship between a "reference temperature" and the fracture toughness, K_{JC}, exists:

$$K_{JC} = 30 + 70.\exp\left[0.019\left(T - T_0\right)\right]\ MN\ m^{-3/2}$$

where T is the temperature of interest and T_0 is the ductile-to-brittle transition temperature (Wallin 1991; IAEA 2009). To improve the accuracy of residual life assessments, also more detailed analyses with a combination of mechanical testing of miniaturized samples, extended microstructural analyses, and advanced material modeling tools are considered (see, e.g., Hoffelner (2012)).

Reactor Internals

Radiation-induced material changes and susceptibility to intergranular failure of light water reactor core internals were summarized in Bruemmer et al. (1999b). Such failures have occurred after many years of service in boiling water reactor (BWR) core components and, to a lesser extent, in pressurized water reactor (PWR) core components. These failures occur in stainless iron- and nickel-base alloys exposed to a significant flux of neutron radiation in the reactor coolant environment.

Stress corrosion cracking (SCC) without radiation is an unexpected sudden failure of ductile metals subjected to a tensile stress in a corrosive environment. The stresses can be the result of the crevice loads due to stress concentration, or they can be caused by the type of assembly or residual stresses from fabrication (e.g., cold working). SCC cracks are predominantly intergranular in nature. Reactor

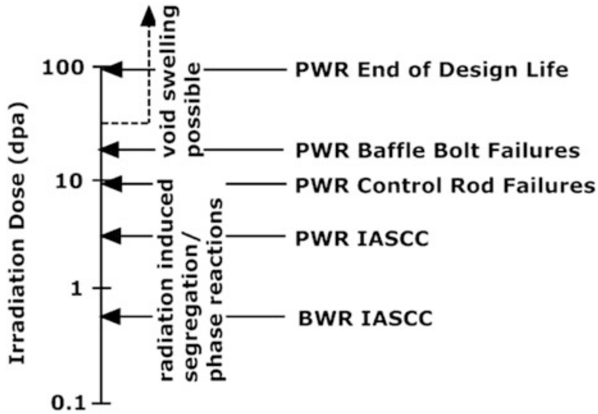


Fig. 18 Possible damage of LWR components due to irradiation-assisted stress corrosion cracking dependent on time of exposure, given as irradiation dose (Source Bruemmer et al. (1999b))

internals do not belong to the group of pressure boundaries, and therefore, the mechanical loads are lower than in RPV and pressure piping. Internals are mainly exposed to the coolant, and therefore, corrosion resistance is more important than strength. Under these conditions austenitic steels are much better suited than the low-alloyed ferritic/bainitic steels used for RPVs. During service cracking susceptibility often results as a combination of radiation, stress, and a corrosive environment. The resulting failure mechanism has therefore been termed irradiation-assisted stress corrosion cracking (IASCC). Intergranular(IG) SCC is promoted in austenitic stainless steels when a critical threshold fluence is reached (see, e.g., Was (2007), Hoffelner (2012)). This time dependence leads to occurrence of cracking after some time of operation as shown in Fig. 18 (Bruemmer et al. 1999b). Therefore, it took quite a while until this type of damage became obvious in nuclear plants. As in classical stress corrosion cracking (SCC), the aqueous environment chemistry and component stress/strain conditions also strongly influence observed cracking.

Several important metallurgical, mechanical, and environmental aspects that are believed to play a role in the cracking process (Bruemmer et al. 1999b; Andresen et al. 1990; Was and Andresen 1992; Scott 1994; Ford and Andresen 1994) are shown schematically in Fig. 19.

The damage happens most probably in the following form:

- Irradiation damage leads to hardening of the matrix making basically the grain boundaries more attractive as path for growing cracks. This is what often happens as a result of hardening also without irradiation.
- Irradiation is also responsible for changes in grain boundary compositions by radiation-induced segregation (primarily chromium depletion) which can further weaken the cohesion between them.
- The surface of the cracks (particularly at the crack tip) is exposed to the radiolysis products which lead to chemical corrosion attack.
- Additionally, the crack can act as crevice supporting crevice corrosion.

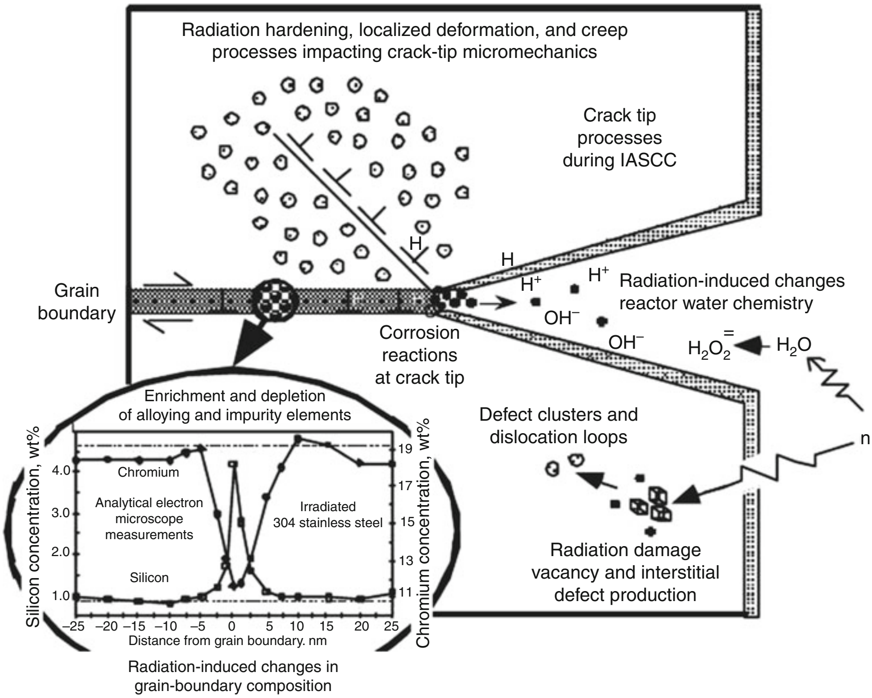


Fig. 19 Main mechanisms of irradiation-assisted stress corrosion cracking; reprinted with permission of ASM International. All rights reserved. www.asminternational.org (Source see also Was (2007))

All these facts together contribute to enhanced crack growth along the grain boundaries. As cracks in the reactor internals do not have the same damage potential as cracking of primary boundary components, measures were developed to stop or to slow down crack growth by chemistry (MacDonald et al. 1995; Hettiarachchi et al. 1995). A successful concept is the technique of noble metal chemical addition (NMCA) which has been commercially applied since 1996 (Hettiarachchi et al. 1997).

With respect to life extension and residual life assessments of reactors, discussions are ongoing if void swelling and eventually even helium effects might become relevant for very long times of operations (see Fig. 18). Void swelling was of concerns and even application limiting for earlier fast reactors. But void swelling has not yet been of real concern for LWR internals. Due to the very low dose (2–3 dpa maximum) expected in the shrouds of BWRs, void swelling per se is not considered to be a license extension issue for BWRs. However, as recently discussed (Garner et al. 2005; Garner 2010b), there is a growing body of evidence that swelling and irradiation creep might become important for life extensions of LWRs to 60 years and beyond. The most swelling-vulnerable locations (>5 %) are expected to be concentrated in small volumes of the reentrant corners of PWR baffle-former assemblies constructed from AISI 304 stainless steel. Even at lower swelling levels, however, differential swelling of annealed 304 baffle-former plates and cold-worked 316 baffle bolts is being considered as a possible contributor to corrosion and cracking of bolts.

Zircaloy Claddings

Claddings are the structural parts which are mostly exposed to irradiation and therefore to irradiation damage. The effects of neutron irradiation on microstructure and properties of Zircaloy were summarized by Adamson (Adamson 2000). For Zircaloy with a hexagonal crystal structure black dots, dislocation loops (often related to the basal c-plane) and microstructural changes leading to swelling and irradiation creep are the most significant types of damage. After service exposure in LWRs, a high density of black spots is present (see Fig. 20). The spots are very small, and a further analysis of its nature is not easily possible.

Irradiated Zircaloy undergoes also swelling and irradiation creep both being important for design because they are responsible for structural changes during service which always bears some risk for failure. Swelling of Zircaloy is of high importance for CANDU reactors where also the pressure pipes are made of Zircaloy. Swelling phenomena of Zircaloy are today reasonably well understood which is a result of the long experience with light and heavy water reactors. However, increasing burnup and possible effects of service exposure on fuel rods after its active life during transport or at final storage trigger active research in this field still today. Swelling is a function of fluence, microstructure, and temperature but also on hydrogen content and other parameters.

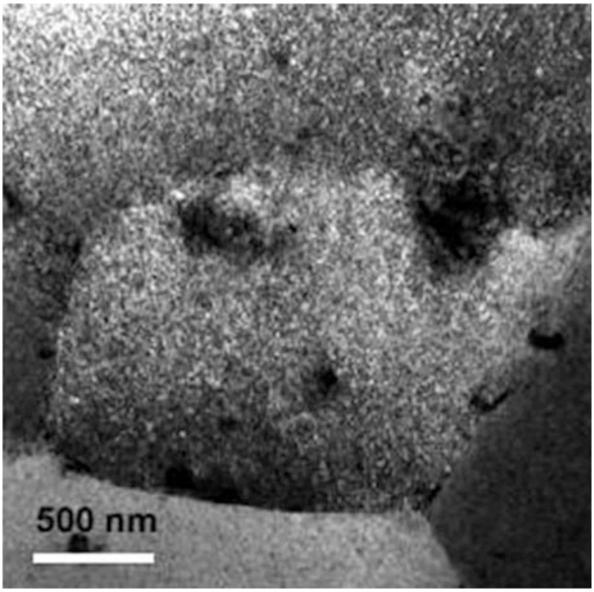
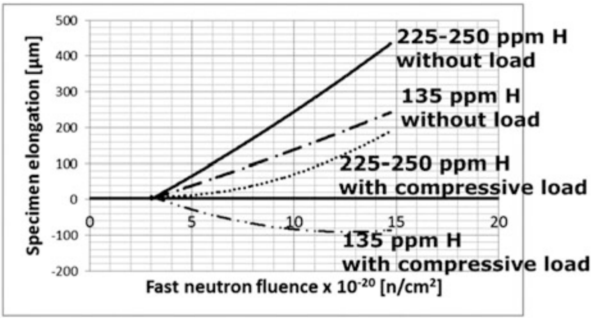


Fig. 20 Irradiation damage in service-exposed Zircaloy cladding. Black dots are clearly visible (TEM bright field micrograph replotted from Hoffelner (2012))

Fig. 21 Irradiation creep of Zircaloy guide tubes tested in the Halden test reactor (Source McGrath et al. (2010))



Swelling and creep of samples from several sections of pre-exposed, recrystallized Zircaloy-4 guide tubes from 2 commercial PWRs were studied in the Halden test reactor (McGrath et al. 2010). The load for the creep test was applied by the loop pressure squeezing a sealed bellow which applied a compressive axial force on guide tubes. The results are shown in Fig. 21. The radiation creep effect due to compressive load is clearly visible. It also looks like increasing hydrogen content would increase swelling and radiation creep. According to the authors of this study, a quantitative explanation needs to consider several factors including the prehistory of the claddings.

Phase changes are another effects encountered in Zircaloy under irradiation as previously mentioned already. Zircaloy contains (mainly intermetallic) precipitates, so-called second phase particles which play an important role for oxidation. Common precipitates are $Zr(Fe,Cr)_2$ and $Zr_2(Fe,Ni)$ which can amorphize and decompose during service (Valizadeh et al. 2010; Herring and Northwood 1988). Simultaneously, iron (and at a much slower rate, chromium) can be lost to the matrix. Decomposition of second phase particles changes the local chemical composition of the matrix and plays therefore an important role for the oxidation of Zircaloys.

Advanced Nuclear Plants

Basic Considerations

Although current LWRs undergo permanent improvements, also other reactor types are considered for the future. Six reactor concepts shown in Table 2 were chosen for further consideration within the International Generation IV initiative (Roadmap 2002). Cooling media different from water (except for the supercritical water reactor) as well as the use of a fast-neutron spectrum (in contrast to the thermal spectrum of current light water reactors) are characteristics of these systems. Thermal neutrons have typically energies ranging from about 1 eV to 0.1 MeV. Fast neutrons have energies higher than 0.1 MeV. The coolants allow higher operation temperatures and provide therefore also options for combined cycle plants generating electricity and process steam or heat. The fast spectrum allows

Table 2 Generation IV types of reactors and nuclear plants. Nuclear fusion is often referred to as Generation V

System	Neutron spectrum	Coolant	Outlet temperature °C	Fuel cycle	Typical size (MW_e)
VHTR (very-high-temperature reactor)	Thermal	Helium	900–1,000	Open	250–300
SFR (sodium-cooled fast reactor)	Fast	Sodium	500–550	Closed	50–150 300–1,500 600–1,500
SCWR (supercritical-water-cooled reactor)	Thermal/fast	Water	510–625	Open/closed	300–700 1,000–1,500
GFR (gas-cooled fast reactor)	Fast	Helium	850	Closed	1,200
LFR (lead-cooled fast reactor)	Fast	Lead	480–570	Closed	20–180 300–1,200 600–1,000
MSR (molten salt reactor)	Thermal/fast	Salts fluoride	700–800	Closed	1,000

closed fuel cycles which would be a real step towards waste minimization and resource saving. A fast spectrum is obtained when the reactor operates without moderators like water or graphite. Together with advanced recycling techniques, fast reactors could use spent fuel currently considered as long-life high-level radioactive waste as mixed fuel. The residuals for final storage would be only fission products with much shorter half-life than plutonium and minor actinides which are currently of concern for final repositories. Also uranium could be used far more efficiently as fuel.

Except the SCWR and GFR, several reactor concepts have been already studied earlier (Hoffelner et al. 2011). The German HTR was a high-temperature reactor with direct cycle helium turbine, and the French "Phenix" and "Superphenix" were sodium fast reactors. Experience with lead-cooled reactors exists from Russian submarines. MSR considerations were reported already in the 1960s of the last century by Oak Ridge National Laboratory (ORNL).

Besides the six GenIV concepts, also other advanced nuclear power options are under consideration: Quite some interest found the sodium-cooled traveling wave reactor of Intellectual Ventures (US) in which the active part penetrates slowly (several years) through pipes (similar to a burning cigarette) (Wald 2013) and Accelerator-Driven Systems (ADS) where the neutrons are created by directing protons from an accelerator towards a target, creating neutrons this way (Rubbia et al. 1995). These neutrons are coupled preferentially to a lead-cooled fast reactor. Finally nuclear fusion should be mentioned as advanced nuclear system (2013), however, without further description.

Fast neutrons are (according to higher energy) more damaging to radiation-exposed materials than the predominantly thermal neutrons in current LWRs. In contrast to LWRs only scarce information concerning in-service radiation damage

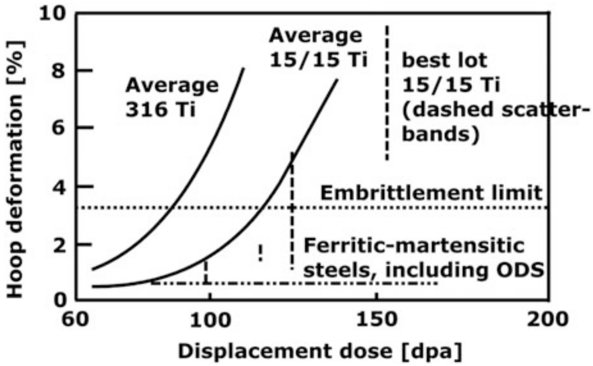


Fig. 22 Swelling of different cladding materials in French fast reactors. Ferritic–martensitic steels show the best behavior, but the titanium-modified austenitic steels could provide an alternative (Source Yvon and Carré (2009))

of components is available. Environments other than water and higher operation temperatures are additional challenges for materials and components.

Vessel embrittlement is expected to stay as an important issue also for advanced reactors. The advantage for SFR, LFR, and MSR types is that they don't need a pressure vessel, only a vessel which is preferentially made of austenitic steel or a nickel-base alloy. Vessel embrittlement is not expected to have the same impact on safety like in case of LWRs. SCWR, GFR, and VHTR vessels are pressurized vessels necessitating ferritic–martensitic steels (low-alloy steels, martensitic 9 % Cr steels) which are expected to show comparable displacement damage like current RPVs. Eventually other long-term degradation mechanisms need to be investigated.

Swelling and irradiation creep could become life-limiting factors for claddings and internals of fast reactors. Ferritic–martensitic steels show a better swelling behavior than austenitic steels (see Fig. 22). Austenitic steels based on 316 with distinct Ti additions (15/15 Ti) show acceptable swelling behavior and are therefore considered as claddings for SFRs at least for moderate burnup.

Development of helium voids and bubbles is considered as a major damage cause particularly with the high temperatures causing additionally thermal creep. Helium produced during service can move to grain boundaries forming bubbles and reducing creep ductility and creep rupture strength (see Fig. 11). Oxide dispersion strengthened (ODS) steels contain nanosized oxide dispersoids which can trap the helium and therefore prevent it from moving to the grain boundaries. Unfortunately the production of ODS materials is rather difficult, and additionally they are extremely expensive which limits its potential for future applications. Interestingly, ODS steels do not show significant improvements in irradiation creep although they show much better thermal creep resistance compared with non-ODS grades as shown in (Toloczko et al. 2004; Chen and Hoffelner 2009).

Advanced Material Characterization Tools

To gain more confidence on possible material degradation in service besides testing and evaluation of traditional samples as methods of advanced material research (materials modeling and model validation) could make an important contribution to safe operation of advanced nuclear plants. Figure 23 summarizes current testing,

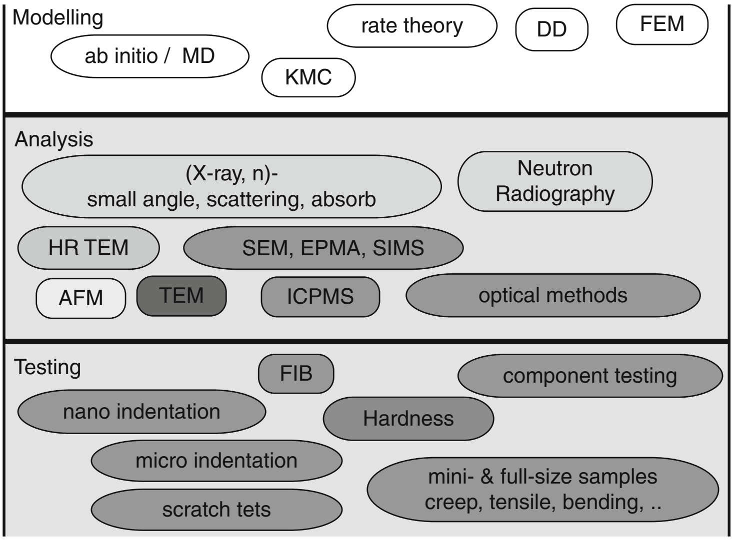


Fig. 23 Testing and analysis techniques used to characterize material behavior on different scales (MD molecular dynamics, DD dislocation dynamics, KMC kinetic Monte Carlo, n neutrons, HR high resolution, TEM transmission electron microscope, SEM scanning electron microscope, AFM atomic force microscope, ICPMS inductively coupled mass spectroscopy, SIMS secondary ion mass spectroscopy, EPMA electron probe micro analysis, FIB focused ion beam, FEM finite element analysis) (Source Hoffelner (2012))

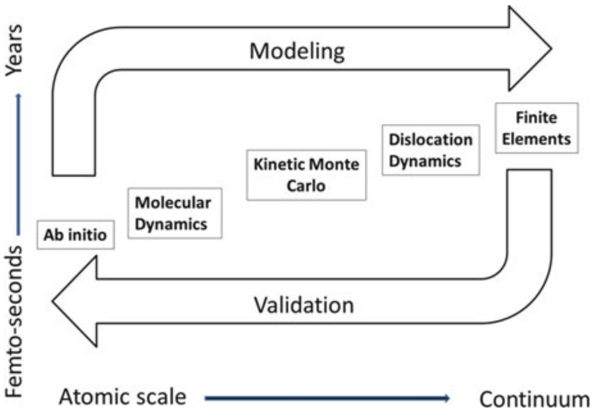


Fig. 24 Scale dependence of different modeling techniques (Source Hoffelner (2012))

analysis, and modeling methods which are used for materials and damage characterization. A detailed description of the different techniques with respect to assessment of irradiation damage in advanced plants goes far beyond the scope of this chapter, and only a few important aspects shall be highlighted. The most important modeling methods are shown in Fig. 24.

Ab initio calculations investigate the physics of matter. They are mainly based on the application of density functional theory (DFT). This allows (with a number of approximations) the determination of the energy of the ground state of a system of interacting particles. Basically this would need a many-body solution of the quantum mechanics Schrödinger equation. DFT provides a reformulation of this problem to a single-body problem. Due to computational restrictions, such calculations are currently limited to a small number of (up to 1,000) atoms. Most calculations are static, thereby neglecting dynamic effects. Methods for dynamic calculations basically exist, but they are extremely computing time-consuming and expensive. Despite these limitations which do not allow the investigations of temperature effects or the behavior of larger systems, the method allows to gain insight into the basic atomistic behavior of a solid.

Details about the arrangement of atoms and similar microstructural issues can be studied with molecular dynamics (MD) simulations. Molecular dynamics implements the potentials to describe the movement of atoms in space and time as results of interatomic and external forces. The equations of motion of classical mechanics in combination with the potentials determined with quantum mechanics are solved for a set on N interacting atoms starting from assigned initial conditions. Controlling variables like temperature or pressure can be conveniently introduced as constraints.

MD simulation can accurately describe the atomistic behavior, but the total simulation time is typically limited to less than 1 ms. On the other hand, the important damage processes in structural materials usually occur on much longer timescales. These processes include reactions between atoms, adsorption-desorption on the surface, occasional transitions from one state to another, and especially diffusion and annihilation of defects after a cascade event in an irradiation experiment. Such effects can be studied using a combination of MD and Kinetic Monte Carlo (KMC). The KMC method is a probabilistic approach that enables the prediction of longer-term damage evolution. The output data of MD is used in KMC in order to determine the probabilistic motion and reaction between defects and atoms (Dalla Torre et al. 2005; Barbu et al. 2005; Domain et al. 2004) where motion and clustering of point defects are the dominant mechanism.

Models based on reaction *rate theory* have been broadly and successfully applied to simulate radiation-induced microstructural evolution radiation damage (Stoller and Greenwood 1999; Stoller et al. 2008). The use of these models involves the simultaneous solution of a modest number of differential equations to predict phenomena such as void swelling, irradiation creep, or embrittlement. The timescale of interest for these processes is determined by atomic diffusion rates and the desired in-service lifetime of irradiated components. Rate theory is well suited to span this time range from seconds to years and a size scale from micrometers to macroscopic dimensions. However, the source term in the rate equations is dictated by atomic displacement cascades, events that occur on a timescale of a few tens of picoseconds and a few tens of nanometers in space.

Discrete dislocation dynamics (DDD) simulation is a mesoscopic tool to study plastic deformation and the interaction between dislocations as well as between

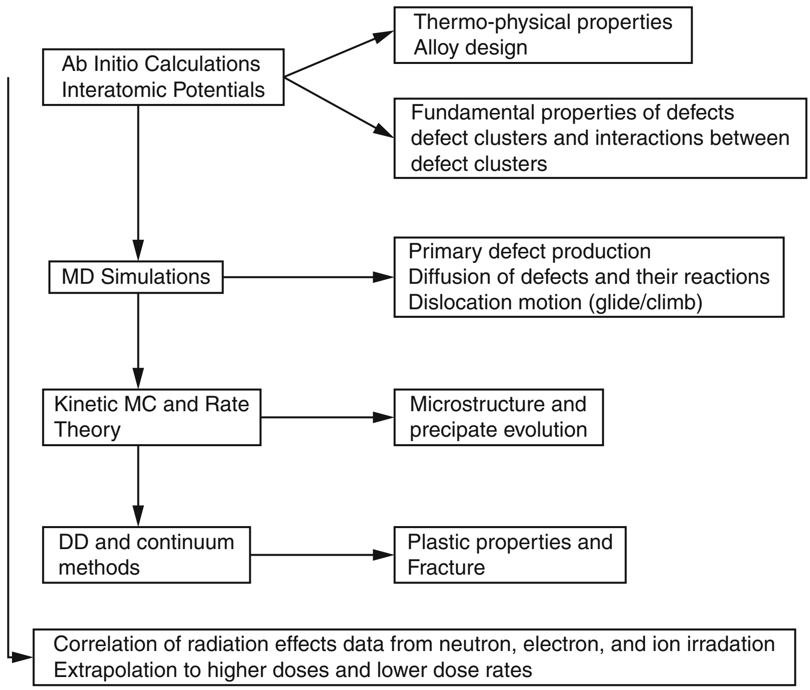


Fig. 25 Elements of a full-scale model description of radiation damage in metals (Source Stoller and Mansur (2005))

dislocations and obstacles. It directly simulates the dynamic, collective behavior of individual dislocations and their interactions. In a numerical implementation, dislocation lines are represented by connected discrete line segments that move according to driving forces including dislocation line tension, dislocation interaction forces, and external loading. Discrete dislocation dynamics operates in single crystals. There are also developments of dislocation field dynamics modeling where dislocations are characterized by stress fields rather than discrete dislocation line elements (Ghoniem et al. 2000; Ortiz 1999; Koslowskia et al. 2002). Such models operating on larger scale than discrete dislocation dynamics are able to go beyond the limitation of single crystals.

Constitutive equations and finite element calculations describe the macroscopic behavior of a material. Figure 25 (Stoller and Mansur 2005) shows schematically how the different modeling techniques can contribute to the understanding of radiation damage.

Model validations can be done with advanced analytical techniques like high-resolution TEM or beamline techniques (X-rays, neutrons) and with advanced mechanical testing. In contrast to testing of large laboratory samples, advanced mechanical testing uses micro- or nanosize testing equipment. This allows the

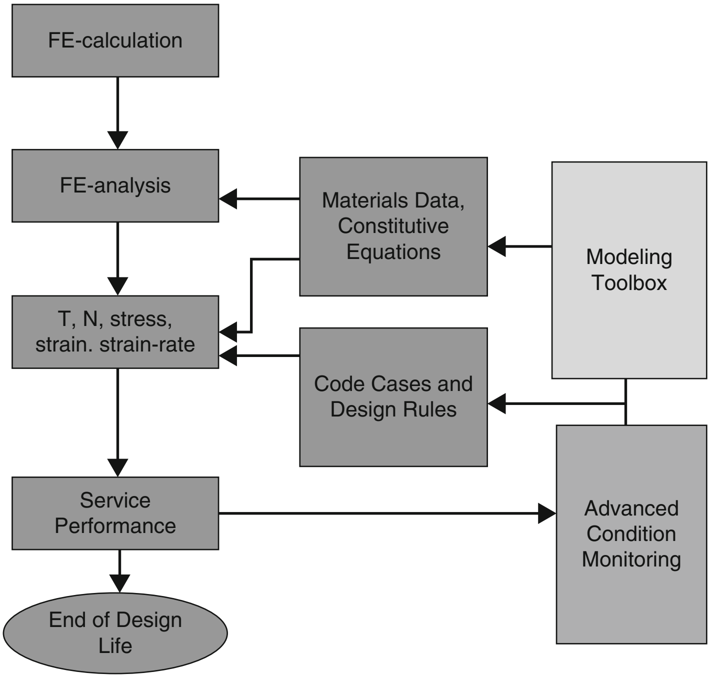


Fig. 26 Possible interaction of advanced modeling methods and advanced condition monitoring techniques with traditional design (Source Hoffelner (2012))

determination of mechanical properties of small volumes which is necessary for a good correlation between microstructure and strength. Experimental investigation of the early stage of radiation damage needs tools with extremely high time resolution and extremely short detection ranges. It can be expected that the new generation of beamline techniques (free electron lasers (Swissfel 2011; Jefferson Lab 2013)) can bring significant contributions to experimental studies of early irradiation damage.

Determination of material response under neutron irradiation requires very time-consuming, difficult, and expensive tests under neutron irradiation. Tests with other energetic particles (ions, electrons) are therefore an important tool to study radiation damage (though quantitative differences to neutrons might punctually exist).

Although all these advanced methods are remarkable improvements for the basic understanding of materials, they can currently not deliver real design data. For those data still testing of large samples and service experience in test beds and demonstrators are necessary. However, it can be expected that traditional design methods together with advanced materials science and service experience can strongly enhance damage and lifetime assessments in future nuclear plants as schematically shown in Fig. 26.

Conclusions

Irradiation damage of structural materials is a complex particle–matter interaction which shows a wide range of effects at different timescales:

- Scattering effects leading to point defect supersaturation
- Point defect reactions leading to obstacles for dislocation movement
- Nuclear reactions leading to phase transitions and phase changes
- Nuclear reactions leading to transmutation of elements

On component level these effects lead to changes in geometry (swelling), changes of mechanical properties (hardening/embrittlement), and changes of chemical behavior (irradiation-assisted stress corrosion cracking, oxidation resistance). Irradiation together with mechanical loads can lead to irradiation creep at temperatures far below which thermal creep occurs. Particularly important are transmutation reactions which can lead to alpha-radiating isotopes which are sources for gas development in the material which can severely degrade mechanical properties.

Irradiation effects and its consequence on material properties and safety are quite well understood for current light water reactors with service experience of about 50 years. New future nuclear plants will in principle undergo the same types of damage. However, the fast-neutron spectrum together with higher temperatures and new working environments need a careful assessment of damage and safety in such plants. Advanced tools and methods of materials science are expected to bring significant contributions there.

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