

Development of Radiation Resistant Reactor Core Structural Materials

A. Introduction

1. The core of a nuclear reactor is where the fuel is located and where nuclear fission reactions take place. The materials used to encase the fuel in fuel rods, to hold fuel rods together in fuel assemblies, and to hold fuel assemblies in place are all considered ‘core structural materials’, as are the materials used in control rods and core monitoring instruments and their supporting structures. For fusion reactors the core structural materials are the materials of the first wall, blanket and divertor.
2. The economics of current nuclear power plants is improved through increasing fuel burnups, i.e. the effective time that fuel remains in the reactor core and the amount of energy it generates. Increasing the consumption of fissile material in the fuel element before it is discharged from the reactor means less fuel is required over the reactor’s life cycle, which results in lower fuel costs, lower spent fuel storage costs, and less waste for ultimate disposal. There has been a continuous historical increase in fuel burnup from 20–25 GWd/tU in Generation I reactors to 50–60 GWd/tU in today’s Generation II and III light water reactors. Design parameters for Generation IV fast reactors call for more than a doubling to ~100–200 GWd/tU (U.S. DOE 2002). Higher burnups place severe performance demands on materials used in reactor fuels, reactor core components, and reactor vessels. A detailed discussion of very high fuel burnups is provided in ‘Very High Burn-ups in Light Water Reactors’ (NEA 2006) and in the additional documentation on ‘Trends in Nuclear Fuel for Power Reactors’ associated with the *Nuclear Technology Review 2007*.
3. The core structural materials have to retain their functionality to maintain integrity of the fuel rods and fuel assemblies, preventing release of radioactive materials from the fuel to the coolant. To support higher burnups, improved radiation resistant materials need to be developed that can withstand harsher irradiation environments and higher temperatures. A measure of the effect of irradiation on materials is the number of times an atom is displaced from its normal lattice site by atomic collision processes. This is quantified as displacements per atom (dpa) (ANSI 2007). Figure 1 illustrates the required in-service operating environments for core structural materials in various types of reactors. A typical LWR fuel cladding, at a burnup of 40 GWd/tU, will have experienced about 20 dpa, meaning that, on average, each atom is displaced from its site in the crystal lattice 20 times. In future fast reactor systems it is expected that atomic displacement can reach 150–200 dpa, depending on the characteristics of any alloying elements and the neutron spectrum. Precise estimates of radiation damage require complicated computations.
4. Materials behaviour under irradiation has been studied for more than 50 years, with most experience in the ‘thermal reactor materials’ area depicted in Figure 1, where core structural materials are subject to temperatures up to 400°C and damages up to 20 dpa. Some experience has been acquired in the area of ‘fast reactor materials’, but the high temperature reactor (HTR) area is much less explored.
5. Materials to be used for Generation IV reactors and future fusion reactors will operate at even higher temperatures 500°C–1000°C and experience damage up of ~30–100 dpa. The International Thermonuclear Experimental Reactor (ITER) which will shortly begin construction in Cadarache (France) will create new opportunities to experimentally investigate and better understand the factors that may limit the use of structural materials currently used in fusion facilities (‘fusion materials’ in

Figure 1). ITER will have operating temperatures of 100°C–300°C and damages of up to 3 dpa from 14 MeV neutrons (ITER 2005). In DEMO (Demonstration Power Plant), the prototype fusion power reactor that will be constructed after ITER, the operating temperatures are expected to be in the range of 500°C–1000°C and damage will reach ~150 dpa at the end of five years of full power operation (Maisonnier et al. 2006). As indicated in Figure 1, commercial fusion power reactors are expected to reach even higher damages.

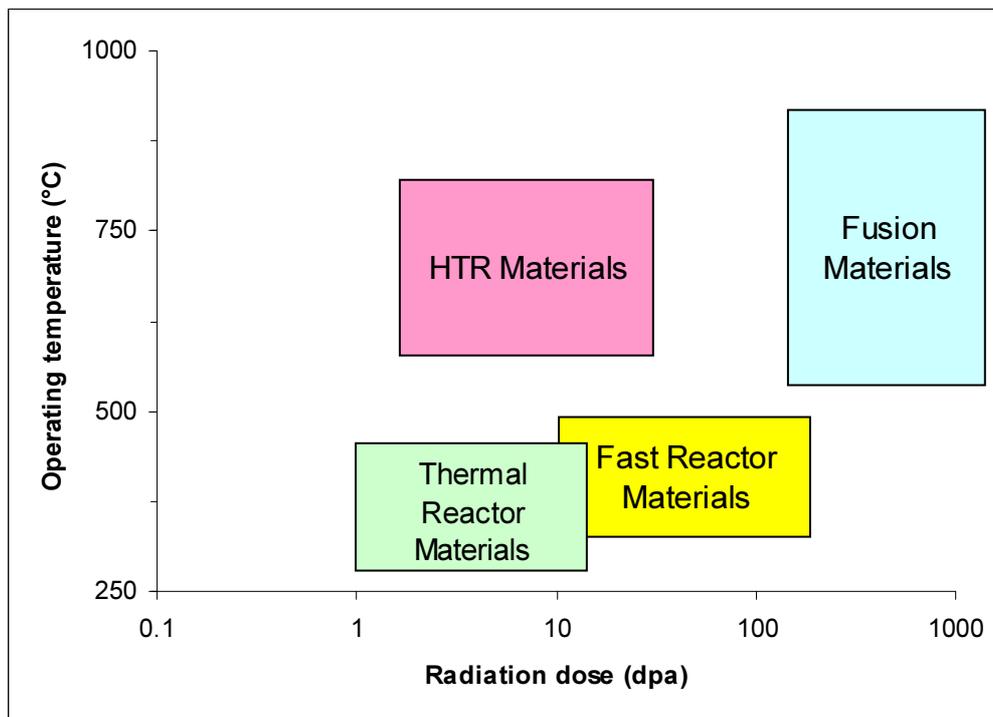


Figure 1. Operating conditions for core structural materials in different power reactors

6. Significant experimental and theoretical progress will be needed to solve the challenges facing the development of advanced reactors and their core structural materials. Progress will require long-term, coordinated, multidisciplinary efforts, with intensive research needed in the study of radiation damage of materials and especially changes of their mechanical and physical properties at high temperature and radiation dose. The following sections describe briefly radiation damage in materials, technological developments, experimental and computational tools, and future challenges. Special emphasis is given to core structural materials exposed to extreme operating conditions.

B. Radiation Damage and Radiation Effects

7. Given the tight specifications within which a nuclear reactor must operate, it is critical that, throughout the working life of the reactor, structural materials maintain their mechanical properties and dimensional stability within specified tolerances. Incremental changes in materials during steady-state reactor operations must stay within specifications, and all materials must be able throughout the reactor's life to perform as required under all postulated accident conditions.

B.1. The Basic Nature of Radiation Damage

8. Radiation induced changes in material properties are the result of microstructural defects. An energetic particle (e.g. neutron or fission fragment) collides with an atom in a material, transferring to it some energy and knocking it out of its lattice position. This primary knock-on atom and the recoiling particle cause additional collisions with other atoms generating a cascade of displaced atoms. Given that the average energy of a fission neutron is ~ 2 MeV and the threshold energy to displace an atom from its lattice position in metals is ~ 20 – 40 eV, a typical number of displaced atoms in a displacement cascade is $\sim 50\,000$. In most metals, 90–99% of these displaced atoms eventually recombine to vacated lattice positions. It is the remaining non-correctly but stably sited radiation defects and microstructural re-arrangements that constitute the radiation damage that changes the material's microscopic and macroscopic properties. Various types of radiation-induced defects are illustrated in Figure 2.

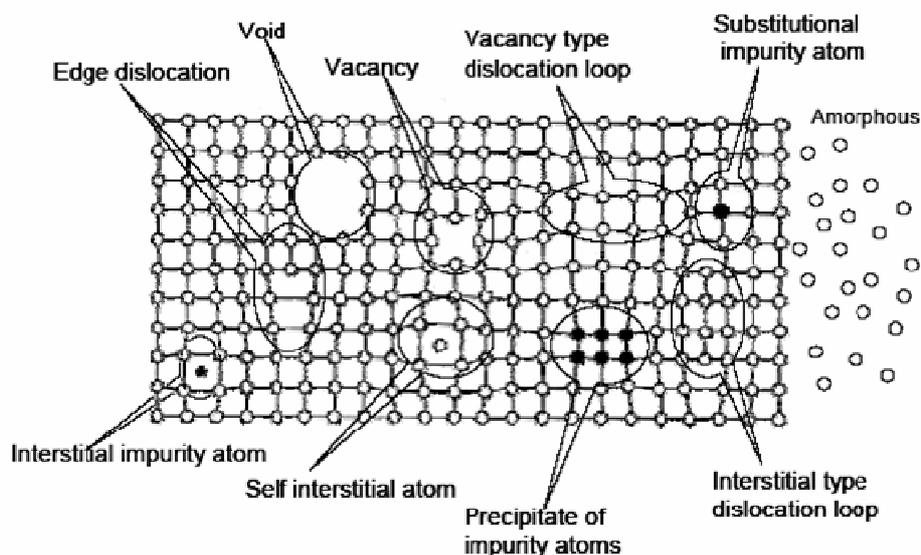


Figure 2. Defects in the lattice structure of materials that can change their material properties.

9. Conceptually, these defects can be visualized as regions where there is either a deficiency of lattice atoms (voids, vacancies, edge dislocations, vacancy type dislocation loops) or an excess of lattice atoms (self interstitial atoms, interstitial type dislocation loops). These deficiencies or excesses produce geometrical distortions in the lattice structure. Similarly, impurity atoms (interstitial impurity atoms, precipitates of impurity atoms, substitutional impurity atoms) also distort the lattice structure. All these distortions lead to changes in the mechanical and geometrical properties of the material. For example, vacancies (voids) lead to macroscopic swelling and distortion of the lattice structure, which alters the material's strength and ductility.

10. The microstructural evolution of a material under irradiation depends on its crystallographic structure. Most metals used in nuclear structural materials have one of three crystallographic structures, two cubic and one hexagonal. Examples of cubic materials are tungsten, iron, vanadium and ferritic steels, which are body centred cubic (BCC), as well as copper, austenitic steels and nickel alloys, which are face centred cubic (FCC). Zirconium and its alloys are the most important 'hexagonal close packed' metals (HCP). A material's crystallographic structure plays an important role in its behaviour under irradiation. Each structure has its advantages and disadvantages and

examples of materials used in different core environments and with differing requirements are described in the following sections.

B.2. Macroscopic Effects of Radiation Damage

11. The most important observable physical changes in material properties are embrittlement, radiation induced growth and swelling, creep, and phase transitions.

- Embrittlement can seriously affect the performance of many reactor components. In materials like BCC ferritic steels, for example, point defects make the material less plastic.
- Radiation induced growth and swelling influence the geometry of core components and can complicate, for example, coolant flow and control rod movements. Swelling is common for FCC austenitic steels and nickel alloys where vacancies tend to form volume clusters. Radiation induced growth in materials such as zirconium, uranium and graphite can lead to different dimensional changes in different directions, depending on the metallographic condition of the material. This is due to an uneven distribution of radiation induced defects forming on different crystallographic planes in their HCP lattice.
- Irradiation creep is a permanent deformation caused by the evolution of different irradiation induced defects, depending on their orientation relative to an applied stress. The material grows in a particular direction and does not return to its original dimensions when the applied stress is removed.
- Phase transitions can be stimulated by irradiation in different ways, leading to negative (and sometimes positive) changes in radiation resistance. Radiation stimulated diffusion of non-equilibrium defects and redistribution of alloy elements may trigger a local transformation of the material's lattice into a more energetically favoured shape. In other cases, irradiation can lead to a homogenization of a multi-phase structure due to atom mixing effects in the displacement cascades.

12. Fast neutrons can create helium and hydrogen atoms via neutron- α particle (n, α) and neutron-proton (n, p) reactions. These atoms can coalesce into gas bubbles that grow and produce voids and swelling in the material. Additionally, neutron induced transmutations (the change of a natural element due to neutron absorption) can produce significant changes in the elemental composition of materials that adversely affect their properties. Helium embrittlement and transmutations in fusion materials and fast reactors thus exacerbate the degradation of materials from radiation.

B.3. Means to Enhance Radiation Resistance

13. Given the origins and effects of radiation damage as outlined above, a major focus of current research is on finding ways to stabilize the displaced atoms, vacancies, and lattice distortions. This can be done, for example, by creating features in the material such as grain boundaries or other vacancy sinks to capture and hold migrating radiation defects. Engineering and nucleonic considerations largely determine the major elemental constituents and phase composition of core structural materials, but there remains scope for the adjustment of the micro- and nanostructures of the material to improve radiation resistance. These options include items such as grain size and orientation, trace elements, introduced sinks, and dispersed strengthening precipitates.

C. Nuclear Reactor Materials Development

14. Resistance to radiation damage, such as dimensional change, embrittlement and creep, is one of the most important considerations in selecting structural materials for a reactor core. More than other components, fuel cladding is exposed to extreme temperatures, pressures, and radiation levels. Consequently cladding material is the most critical in terms of performance and specification design windows. This section thus begins with cladding materials.

C.1. Zirconium Alloys

15. Zirconium is the most extensively used material for fuel cladding and assembly structure in both light and heavy water-cooled reactors. Its low neutron capture cross-section combined with relatively good corrosion and mechanical properties are among its superior advantages and led to its early use in nuclear reactors in preference to stainless steels. Although high purity zirconium has very good corrosion resistance in water, it has low strength at high temperatures, so alloying is required. Four major alloying elements are used — tin, niobium, iron and chromium — with concentrations of not more than 1.5%. These four elements have very low neutron absorption, so they do not affect the neutron economy of the reactor, but allow the zirconium alloy to meet the required engineering criteria for cladding and assembly components.

16. The first zirconium alloys developed as structural materials for fuel rods and assemblies in light water reactors were, for U.S. designed BWRs and PWRs, Zircaloy-2 and Zircaloy-4 (with ~1.5% Sn as the main alloying element and no Nb). Russian designed PWRs (WWER) and graphite-moderated pressurized tube type reactors (RBMK) used the E-110 alloy (with ~1 % Nb as the main alloying element and no Sn). The need for suitable materials for different irradiation environments and water chemistries in different reactor designs led to evolutionary developments of such materials in France, Japan, Russia and the USA. These included variations involving small amounts of oxygen (~0.1%), iron (~0.01–0.4%) and chromium (~0.1%), which yielded the current generation of zirconium alloy fuel rod claddings known as M5, MDA, ZIRLO and E-635. In addition, duplex alloys, where an enhanced corrosion resistant outer layer of different alloy composition is applied to a Zircaloy-4 base material, are used in some high burnup applications.

17. Zirconium and its alloys have a built-in anisotropy (directional dependence) due to their hexagonal close packed (HCP) lattice structure. Such lattice structures can be adversely aligned during material fabrication, resulting in direction dependent irradiation growth leading to cracking. Special thermo-mechanical treatment during tube fabrication aligns the closed packed planes of the lattice parallel to the tube surface, which prevents the radial growth of hydrides from the radiolysis of water and reduces the possibility of radial crack formation and growth and, ultimately, clad cracking. Nonetheless, current understanding of the complex connection between atomistic effects caused by radiation and alterations in observable macroscopic properties is still very limited, and the ability to design, engineer and predict the performance of new materials in high damage radiation environments is still well beyond the present knowledge base.

C.2. Stainless Steels

18. Although stainless steels are currently used for many core components, the future challenge concerns applications for fuel rod cladding and fuel assembly components in fast reactors that use liquid metals as coolants. Fast reactor components operate at high temperatures (up to 750°C) and neutron irradiations (≥ 100 dpa), and only special stainless steels can comply with these requirements.

19. The task of developing fuel cladding materials for a commercial fast reactor able to operate up to burnups of ~ 200 GWd/tU has not yet been solved. In the extremely demanding design conditions for a commercial fast reactor, both austenitic and ferritic-martensitic steels have limitations — the former due to swelling and the latter due to insufficient high-temperature strength. However, both austenitic and ferritic-martensitic steels allow safe fast reactor operations in power regimes that are at the lower limit of commercial efficiency. Their use in research applications will make it possible to accumulate experience on materials behaviour, and to develop and test new advanced fuels. Currently, the only fast reactor in commercial power operation is the Russian Federation's BN-600, commissioned in 1981. Its fuel cladding is made of austenitic stainless steel (ChS-68) with a ferritic-martensitic stainless steel fuel (EP-450) assembly casing. Increased burnup to ~ 150 GWd/tU is planned using improved versions of these materials. Related R&D to develop structural materials for fast reactors is also underway in China, France, India, Japan and the USA.

20. During the past few years, interest has been growing in the development of non-swelling ferritic-martensitic materials with the matrix strengthened by yttrium oxides. This approach is a continuation of an established technique of material strengthening by dispersed fine precipitates. These artificially implanted oxides act as stable obstacles to dislocation movements and reduce creep. Despite the conceptual simplicity of the idea, its technological implementation is difficult.

C.3. Fusion Materials

21. Fusion power plants will require materials that can withstand high operating temperatures (for efficient thermodynamic cycles), tolerate extremely high displacement damage (for long service lives), and produce low activity by-products (for safe disposal at the end of their useful working lives). Entirely new structural materials will have to be developed for fusion to meet the demanding high-performance requirements, and three major material groups, all of which can fulfil the 'low-activation' requirement, are under investigation: reduced activation ferritic-martensitic steels, vanadium alloys, and silicon carbide composites. Reduced activation ferritic-martensitic steels have so far demonstrated the most potential and are the most advanced in terms of research and development. These steels are the reference structural material for DEMO and will be tested and evaluated in ITER. The requirements for fusion reactors are close to those for Generation IV fission reactors, and materials research activities in the two areas are mutually supportive.

22. A notable difference between material requirements for Generation IV reactors and fusion reactors is in the amounts of transmutation helium produced by (n, α) reactions. In Generation IV reactors it is projected to reach a maximum of ~ 3 – 10 appm (atoms per part per million), whereas in ITER it is ~ 75 appm and in DEMO ~ 1500 appm. As previously mentioned, helium induced embrittlement and swelling are major challenges that need to be overcome for the deployment of commercial advanced reactor concepts.

D. Research Facilities for Materials Testing and Investigation

23. Various micro- and nanoscale experimental and computational tools are required to investigate phenomena in radiation materials science. Experimental techniques are needed to probe elemental compositions, structures, and disordered systems. Nanoscale techniques that have demonstrated their applicability include high resolution transmission electron microscopy, synchrotron radiation techniques and micro-X-ray diffraction, small angle neutron scattering, atom probe tomography,

positron annihilation spectroscopy, and muon spin resonance spectrometry. The major facilities required include synchrotron light sources, high-flux research reactors, spallation neutron sources, and high power accelerators.

24. Research also requires a supply of specimens of suitably damaged materials. Currently there are not enough such specimens because there are not enough facilities where materials can be subjected to sufficiently intense radiation and high damage. For testing potential fusion materials with 14 MeV neutrons, the International Fusion Materials Irradiation Facility (IFMIF) has been proposed (Moeslang et al. 2006). For testing fast reactor materials, there are currently only five operational fast flux reactors available: PHENIX (France), FBTR (India), JOYO (Japan), BOR-60 and BN-600 (both in the Russian Federation) (IAEA 2007). Given the shortage of experimental test facilities, the possibility of using particle accelerators to emulate the neutron damage created by the fission neutron spectrum has been investigated, with encouraging results (Was et al. 2002). However, in-situ experiments done at elevated temperatures and in environments typical of actual operating conditions would still be preferable.

25. Simulation and modelling of radiation damage also provide new essential insights into the microstructural evolutions that occur during the extremely short period between the initiation and final outcome of the radiation damage cascades. The various steps of collision-recombination-relaxation-defect migration-clustering-nucleation-growth etc. are all essentially completed within periods ranging from picoseconds to milliseconds. Experimentally, it is not possible to observe in real-time these many competing processes. Only afterwards can the resultant static microstructure be investigated in a laboratory. Simulations have already demonstrated their value in radiation materials science using a variety of approaches such as molecular dynamics, kinetic Monte-Carlo, rate theory, and dislocation dynamics (ORNL 2004). But there is still substantial potential for improvement.

E. Current and Emerging Challenges

26. In the past, it has been possible to test material performance in research and power reactors since the required accumulated radiation damage was relatively small. But with increasing burnup, this approach will not be realistic due to the prohibitively long residence periods and high costs of the required experiments. Existing methods will have to be modified and new approaches developed. One possibility is simulation and modelling to quantitatively predict alterations in material properties, initially at low irradiation doses, following which the models can be refined and validated via experiments (ORNL 2004). Through continuously improving the theory and models to better match experimental results, and through iterations involving increasing irradiation doses, the objective is sufficient reliability and confidence in the model codes to predict the performance of nuclear structural materials in regimes that cannot be realistically achieved experimentally.

27. Research and power reactors, spallation sources, and accelerators, have all been used to irradiate test materials and study radiation damage. However, despite similarities in radiation damage mechanisms cutting across these different irradiation environments, there are still differences and the same irradiation under different conditions can give different results. Such differences raise as yet unanswered questions about the comparison and evaluation of experimental results in different settings. To help answer such questions, better knowledge of the nature of radiation effects is required through inter-comparison experiments on model materials under controlled irradiation conditions.

Well coordinated research projects can make an important contribution, and the IAEA is fostering such cooperative research and development.

28. Thus while the current level of nuclear materials research and development is adequate to meet the needs of currently operating nuclear facilities, advanced new generation nuclear power plants, both fission and fusion, present major challenges. These are sufficiently substantial in terms of the science, technology, and required resources that no one country or small group of countries will likely be able to maintain the necessary research momentum over an extended period. There is an agreed need for an international mobilisation of resources and internationally coordinated research efforts, probably over many decades. As one immediate contribution, the IAEA facilitates information dissemination and exchange, and promotes collaboration among nuclear institutions, governments, industry, academia, and international organizations.

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