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Editorial

Introducing the nuclear material challenges

Nuclear power currently accounts for about 20% of the worldwide electricity. And the demand should increase steadily. Since this is based on a relatively restricted number of production plants, the enhancement of production will require more performing materials utilised as structural components or as fuels. In addition the waste stock pile increases and ecological solutions are required.

This Symposium compares and contrasts the material performance requirements. The Symposium also discusses candidate materials in various existing and proposed nuclear power plant components. A common theme for all of these proposed future nuclear power systems is the aiming toward higher operation temperatures and burn-up. An additional key challenge to the successful development of materials for fission and fusion systems is the harsh neutron irradiation environment. Several examples are given to illustrate how multiscale modelling and advanced experimental testing techniques are used to investigate and to resolve key material issues.

Through the last 20 years of material science, Europe is a leader in nuclear material science with strong cooperation's with American, Asiatic and other organisations. The Symposium has been structured on the production unit structural materials, the fuels and the waste forms. These materials are component materials for future generation units such as advanced fission/fusion systems, followed by structural materials for thermal reactor units. The fuel materials including fuel matrices and targets for transmutation are considered prior, during and after irradiation as well as the waste form materials for intermediate and geological disposal.

These nuclear materials are studied for their high thermal stress capacity, good resistance to radiation damage, compatibility with coolant or fluid phase, compatibility with other materials (cladding, backfield, fluids, ...), long lifetime in the system, high reliability, adequate resources and easy fabrication, and good safety and environmental behavior.

1. Component materials for advanced fission or fusion systems

Looking at the future advanced fusion and 'Generation IV' fission reactors, systems are proposed such as the International Thermonuclear Experimental Reactor (ITER) or the (Very) High Temperature Reactor ((V)HTR) or the Gas Fast Reactor (GFR) as next nuclear generation units.

For thermonuclear units that could be operational in some decades such as ITER, the reactor would run with fusion power of 500 MW, with a yield (fusion power/auxiliary heating power) of the order of 10. Under these conditions the temperature of the plasma and the flux of neutrons (simulated by light ions) through the confinement components are extremely high. However the pressure around the vessel is better than 10^{-5} Pa. The materials considered or the unit components are listed in Fig. 1(a). They need to be assessed for safety and environmental considerations [1,2]. They range from refractory metals, steels to carbons and silicon carbides on one side to composites such as oxide dispersion straightens steel (ODS) materials. Tungsten is a promising armour material for plasma facing components and ferritic or martensitic steels are presently considered as promising structural materials for the first wall and breeding blanket.

On the other hand, for the fission reactors, the Generation IV Forum has selected the (V)HTR as one of the most versatile units. Those units shall

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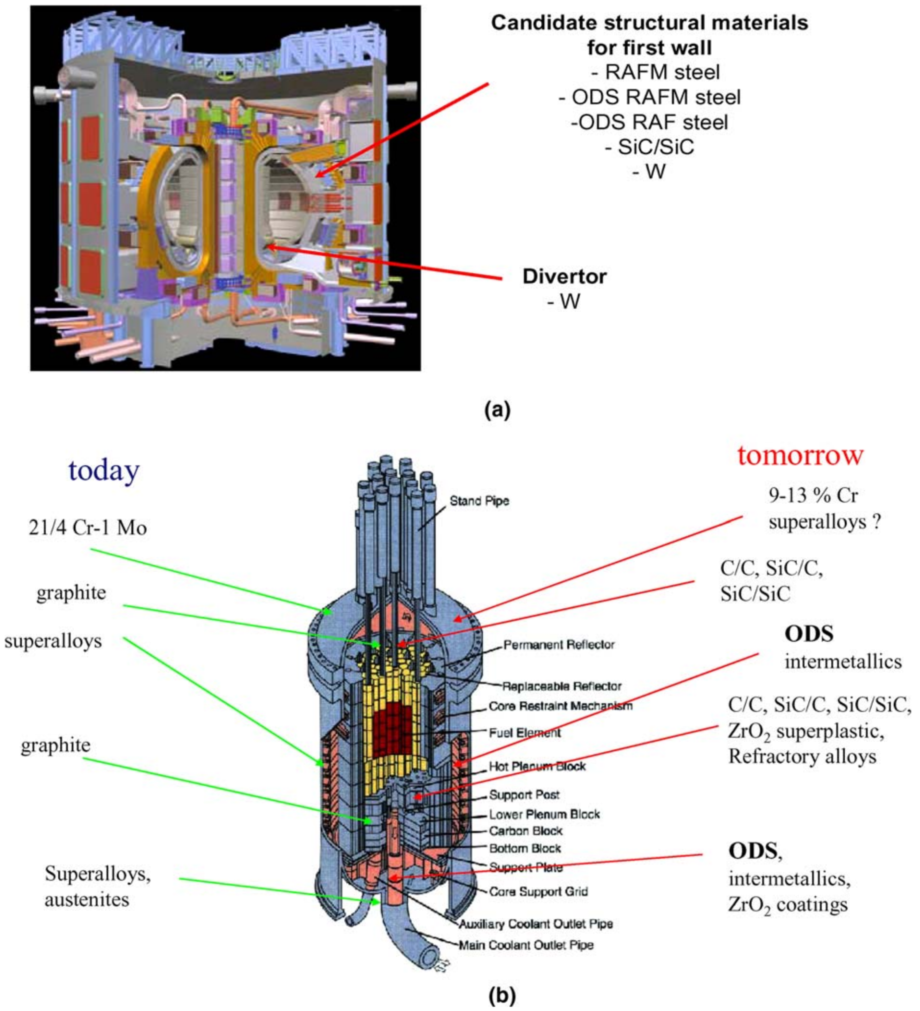

Fig. 1. (a) Fusion unit. (b) (Very) High Temperature Reactor core.

operate with an output of 500 MW at temperature of the order of 1300 K and with pressures of the order of 5 MPa working with He as coolant. The neutron flux in the core of such units should be of the order of 10^{14} cm^{-2}. Here the materials are Ni-based alloys, titanium–aluminium intermetallics, oxide dispersed strengthened steel (ODS) such as depicted in Fig. 1(b). These materials need to be characterised by advanced techniques [3].

With component materials ranging from elements (C, W), alloys (austenites, high chromium ferritic martensitic), oxides dispersed strengthened alloys to ceramics such as zirconia coating. RAF/M stands for Reduced Activation Ferritic/Martensitic.

In the material, neutrons (simulated by ions such as protons or hellions) absorb and yield impurities by nuclear reactions. In the tested samples, damages are consequent with atomic displacement cascades producing point structure defects such as vacancies and interstitials. The final microstructure of the irradiated material results from interactions between the various irradiation-induced defects including defect clusters, dislocation loops, stacking fault tetrahedral, precipitates, voids and/or gas bubbles. This affects also the porosity of the material and consequently several physical and mechanical properties need to be revisited, such as: decrease of electrical conductivity (low temperatures), decrease of thermal conductivity (ceramic materials), dimensional and mechanical stability (swelling, hardening, loss of ductility, loss of fracture toughness and loss of creep strength). Studies include experimental characterisation of these parameters under thermal program and under electron, neutron, proton, helium irradiations as well as damage simulations.

2. Component materials for thermal reactor units

With the increase of operational life of nuclear power plants, and the enhancement of burn-up require specific studies on components such as core vessel steels and cladding materials.

Steels from the core vessel and from piping need analysis of their mechanical properties. This becomes more difficult since reactor irradiated samples are increasingly scares. Thus, in these conditions, micro-scale specimen techniques or non-destructive tests are becoming very attractive to characterize the mechanical properties and the in-pile degradation of the material e.g. [4].

Stress corrosion cracking is a life-limiting factor in many components of nuclear power plant in which failure of structural components presents a substantial hazard to both safety and economic performance [5]. Uncertainties in the kinetics of short crack behaviour can have a strong influence on lifetime prediction, and arise from the complexity of the mechanisms and from the difficulties of making experimental observations. Three dimensional observations and modelling of inter-granular stress corrosion cracking in austenitic stainless steel has to be performed by advanced techniques such as micro-tomography. The understanding of radiation damage in reactor pressure vessel steels is necessary for long term evolution modelling and possible retarding and healing measures.

Corrosion of zirconium alloy cladding is a crucial process that limits the use of the fuel element. The inner dense layer requires full characterisation because it contributes to passivation. The oxygen distribution across the metal–oxide interface is also of great interest and the corrosion interfaces of various zirconium alloys, irradiated for several cycles require inter-comparison studies. The oxide films are currently studied by advanced methods such as micro-X-ray diffraction and synchrotron radiation analysis. And the sub-microscopic observations have to be utilised to be compared with corrosion layer build up models [6].

On the other hand, irradiation effects [7], fission gas burst and pin fracture may develop during operation and handling. This requires failure analysis with primary defect characterization and characterization of hydride phases that reorientate by stress.

The effect of irradiation on embrittlement using fracture toughness tests on cladding samples is carried out. Friction has to be taken into account in the evaluation of the test results. Limited friction helps to avoid stress concentrations in the cladding that might delay crack propagation.

3. Fuel materials, fuel matrix and target for transmutation

The fuel material remains the first safety barrier that retains the fission products. However, since safe energy transfer is the first concern, associated parameters such as thermal diffusivity, thermal conductivity, ... are crucial to assess this process. The fuel is a metallic or ceramic form including the fissile, or the fertile material. The design of the fuel materials includes spherical or cylindrical shape elements i.e. kernels or pellets, with homogeneous, heterogeneous set up (see Fig. 2). Metals, carbides, nitrides or oxides are usually used in research, fast or thermal reactors. For the later, uranium, plutonium or thorium dioxides ... are the fissile or fertile (according to isotope) components and zirconia or magnesia are possible inert matrices, see Ref. [8]. For safety reason the melting point is compared to thermal conductibility and their change with burn-up require measurements. On the other hand, evaluation of thermal properties of zirconia based inert matrix fuel can be estimated by molecular dynamic simulation as it does for uranium and plutonium dioxides. The central fuel element temperature is important to be calculated or measured because it affects the retention of fission products.

In light water reactors, fuel utilisation at high burn-up has required enhanced performance of the uranium dioxide or uranium–plutonium mixed oxide fuel materials. Studies concern highly pressurised intra-granular bubbles and their impact on the mobility and release of rare gases. Calculations and measurements of xenon and krypton phases in fuels as a function of burn-up are required. Ab initio modelling of the behaviour of xenon and helium in oxide nuclear fuel is performed in specific studies.

Kernel solid solutions such as thoria–urania or zirconia–plutonia need to be investigated prior and after irradiation. For example, characterisation of 'triso' fuel particle cross-section is required as well as understanding the fuel-coating material interactions from a thermodynamic point of view.

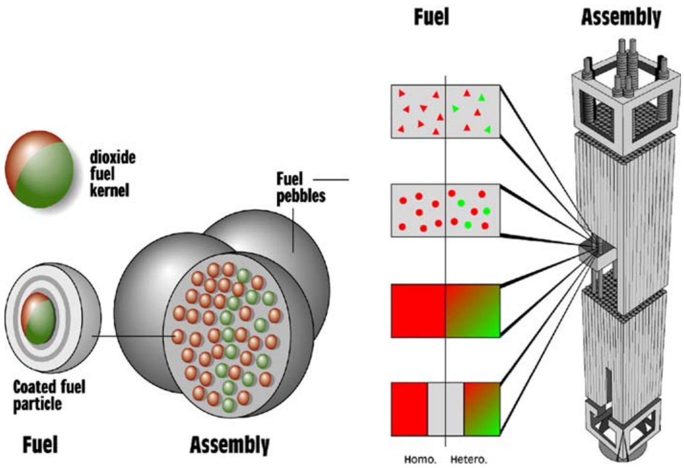

Fig. 2. Fuel design and structure including homogeneous e.g. solid solution or heterogeneous e.g., cercer, cermet as applied for pebble bed power reactor or thermal reactor. Candidates: advanced MOX, UN, MN, UC, MC, ThO*x*, to be compared with UO_2 as reference material; homo and hetero refer to presence or absence of U (green) as fissile component, with Pu in red. (For interpretation of the references in colour in this figure legend, the reader is referred to the web version of this article.)

The inert matrix fuel will remain a key topic [9]. Behaviour of composite phases e.g. magnesia–zirconia will bring new orientation. Neutronic double heterogeneity effect has to be calculated in particles dispersed type inert matrix fuels. The inert matrix concept is also now applied to minor actinide transmutation.

Irradiations in research reactors are very important, while locally fissile enrichment grade is decreased for non-proliferation reasons and performances may change. In all cases Post Irradiation Examination's (PIE) are required and constitute precious data for model verification.

4. Waste form materials for geological disposal

After utilisation in reactor, the spent fuel may be reprocessed or not. The high level waste form may be either the spent fuel itself or, after reprocessing, the material produced for geological disposal. The waste form may be a crystalline material such as uranium dioxide, uranium–plutonium mixed oxide, a ceramic inert matrix fuel, thorium dioxide, etc. or, a non-crystalline material such as a high level waste glass.

Tests and models are applied for phases formed from leaching of low burn-up fuels. They show rapid release from the gap with time scales of the day and relatively rapid release from the grain boundaries with periods of the year, followed by slow release with the matrix alteration. In high burn-up uranium dioxide and uranium–plutonium mixed oxide fuels, the restructured zone characterized by high porosity and small grains play a role for the 'Instant Release Fraction' and the 'Matrix Alteration' which models are adapted for describing this behaviour [10], while emphasis is also given on solubilities [11].

Releases governed by the Zircalloy corrosion and that corresponding to the labile fraction in fresh spent fuel are affected by hydrogen generation counteracting radiolysis. Fast U(VI) dissolution contrasts with very slow U(IV) dissolution making oxidation of the fuel matrix and other redox sensitive nuclides relevant in the safety analysis.

Other waste form materials investigated concern ceramics such as gadolinium–manganese–titanium oxide, thorium- or thorium–uranium(IV) phosphate–diphosphate in solid solution, or apatites. Their behaviours under irradiation are investigated e.g. [12]. Perlites are considered for caesium immobilisation. The dissolution of the matrix and release of radionuclides must be completed with precipitation of secondary solid phases studies. Adjacent materials to waste form may form intelligent barrier that could be part of canister on which the role of irradiation and environment on corrosion are studied.

5. Main focus and future trends

The challenge of Symposium N at the EMRS 2005 is to bring together scientists from various part of the nuclear domain, because several processes such as irradiation damage, phase segregation, ... take place at various level of the material utilisation. Symposium N contributes to exchange nuclear science experiences and data and may contribute to a renaissance of the nuclear culture.

Research of new materials including metals, carbides, nitrides, oxides, alloys or solid solutions or composites focuses on higher stability and better mechanical performances. Characterisation of these materials is carried out using advanced techniques ex situ such as transmission electron microscopy/electron energy loss spectroscopy, micro-X-ray absorption fine structure spectroscopy, small angle X-ray scattering, micro-X-ray diffraction, neutron diffraction, small angle neutron scattering, muon spin resonnance spectroscopy, atom probe, etc. Studies on irradiated materials and at elevated temperatures are required using accelerator and in-pile tests. Characterisation should be performed in situ by non-invasive techniques e.g. using inert windows for observation and analysis (video, Raman spectroscopy, diffuse reflection spectroscopy, ...).

The final goal will be to propose advanced materials for *components* of fusion and Generation IV fission systems, high reliable materials for *structural* pieces of thermal reactors with excellent behaviour in-pile, economical *fuel* materials and targets for transmutation, and, ecological *waste* form materials for geological disposal. To really see the future, our R&D should make the utilisation of the nuclear materials more sustainable, safe, economical and ecological.

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