NUCLEAR ENERGY MATERIALS PREDICTION: APPLICATION OF THE MULTI-SCALE MODELLING PARADIGM

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The safe and reliable performance of fusion and fission plants depends on the choice of suitable materials and an assessment of long-term materials degradation. These materials are degraded by their exposure to extreme conditions; it is necessary, therefore, to address the issue of long-term damage evolution of materials under service exposure in advanced plants. The empirical approach to the study of structural materials and fuels is reaching its limit when used to define and extrapolate new materials, new environments, or new operating conditions due to a lack of knowledge of the basic principles and mechanisms present. Materials designed for future Gen IV systems require significant innovation for the new environments that the materials will be exposed to. Thus, it is a challenge to understand the materials more precisely and to go far beyond the current empirical design methodology. Breakthrough technology is being achieved with the incorporation in design codes of a fundamental understanding of the properties of materials. This paper discusses the multi-scale, multi-code computations and multi-dimensional modelling undertaken to understand the mechanical properties of these materials. Such an approach is envisaged to probe beyond currently possible approaches to become a predictive tool in estimating the mechanical properties and lifetimes of materials.

KEYWORDS : Multi-Scale Modelling, Structural Materials, ODS Ferritic Steels

1. INTRODUCTION

The issue of understanding materials exposed to extreme conditions such as fission and fusion reactors and their respective aging and lifetimes is a complex challenge due to the many and varied phenomena which influence and affect the properties of these materials [1]. It is necessary to predict many constituents that determine and influence the aging processes at work under such environments. Within the Generation IV International Forum (Gen IV) [2], six reactors with very different conditions and quite different materials have been defined. These are the Gas-Cooled Fast Reactor System (GFR), Lead-Cooled Fast Reactor System (LFR), Molten Salt Reactor System (MSR), Sodium-Cooled Fast Reactor System (SFR) and the Supercritical-Water-Cooled Reactor System. A summary of the specific characteristics and present candidate materials is shown in table 1.

The operating temperatures of Gen IV reactors are above those of the current Gen II fission reactors (see table 1 for details depending on the Gen IV system) and in some cases go into temperature regions where phenomena such as creep become important. Gen IV reactors are designed to operate for several years (up to 60) under severe exposure conditions such as neutron irradiation, high temperatures, corrosive environments, and monotonic and cyclic loading. Thus, the determination of the material's lifetime is a complex problem that includes the synergy of these conditions.

Over the years various attempts have been made to obtain physically based lifetime assessments of materials, both structural materials and fuels; however, these design methodologies still follow simple concepts such as the linear life fraction rule. For the development of new materials, a better knowledge of their expected service behaviour would be extremely advantageous. The propagation in materials of strength, fracture, fatigue and creep have been studied as macroscopic events; however, for a more in-depth analysis and understanding of nucleation processes, it is necessary to obtain and incorporate in such studies the microscopic events that occur in materials. Often these microscopic events cannot be investigated experimentally due to the short time scales involved. At the other extreme, long-term aging (of 50 plus years) cannot be investigated experimentally either. Lifetimes are currently computed with the aid of lab-based experimental data, constitutive laws, design rules, and standards based on safety margins. Acquiring long-term statistically secured design data for complex loading conditions, including damage interactions from

experiments, is currently impossible. Furthermore, the task of judging long-term properties during the phase of materials development or optimization is particularly difficult. Such uncertainties also have an impact on design codes such as the ASME code [3] or the French RCC-MR [4]. This modelling combination has been successful; however, to go beyond current materials design, the nucleation and propagation of events, which includes the physics that occurs at the microscopic level, must be addressed.

Although the materials society generally accepts that damage starts at atomistic levels, it is not easy to bridge the gap between the atomic and continuum level. The material issues under these conditions indicate the need to include an atomistic modelling interface to bring forth a predictive design model. As such, the multi-scale modelling paradigm, which uses a range of techniques spanning the nano to macro-scale, has emerged as a next step in materials research and design [5-7]. Such a scheme has the advantage of providing a more in-depth analysis of a material; it also overcomes the time and cost issues associated with traditional macroscopic experimental programs—see figures 1 and 2.

Based on the above description, two really parallel strategies can be developed. On the one hand, the final product may be thought of as a set of key-in-hand codes that parametrically model the behaviour of the material [8]; on the other hand, the models at each stage can be used to learn about the physics of the problem [9]. The second option really assists in the integration of the first because the understanding of the physics leads to a better solution of the modelling of the interfaces between the scales.

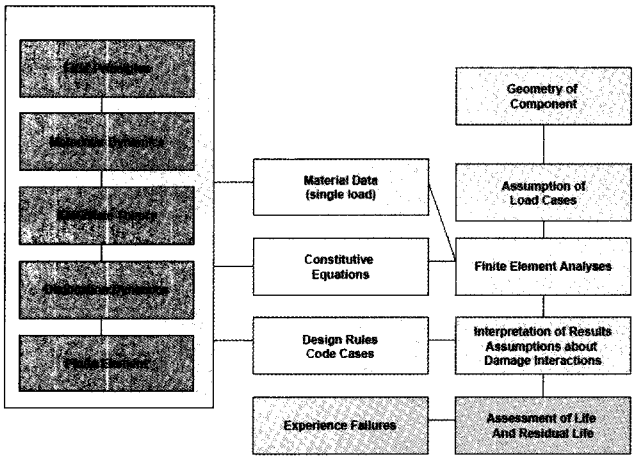

Fig. 1. Possible role of Modelling in the Future Design Process where Fundamental Methodologies are Included in the More General Design Codes

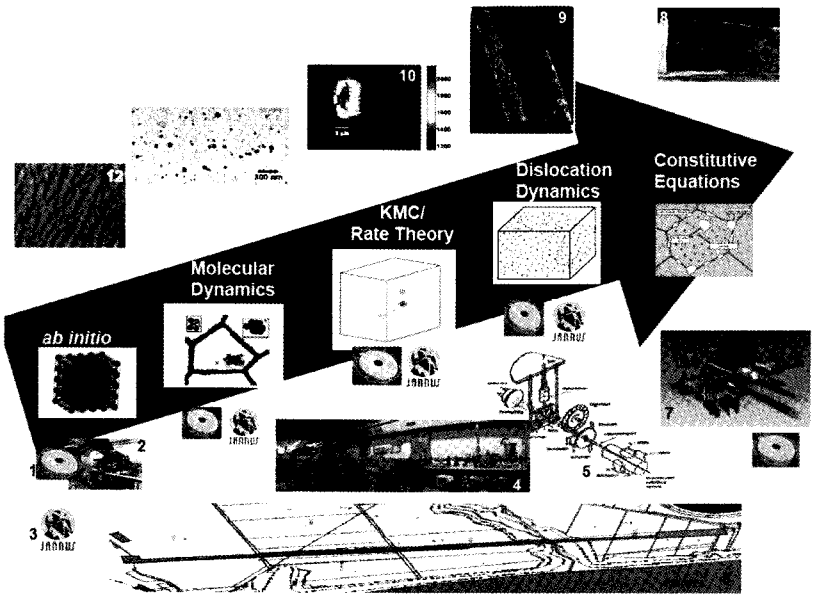

Fig. 2. Multi-Scale Modelling and Experimental Validation Methodologies. The Arrow across the Diagonal Contains the Modelling Methodologies and Refers to Larger Length and Time Scales when Moving from the Bottom-Left Corner to the Top-Right Corner. Below the Diagonal are Types of Experimental Equipment that can be Used at the Different Modelling Scales: 1) Synchrotron Irradiation Facility, Swiss Light Source (SLS), Switzerland; 2) MicroXAS Beamline, SLS; 3) Jannus Facility, France; 4) Irradiation Facility, Tandem Accelerator, ETHZ; 5) *in Situ* Irradiation Creep Facility; 6) Future XFEL Facility, Paul Scherrer Institute, Switzerland; 7) Future Gen IV Reactor. The Figures above the Diagonal Show Types of Results that can be Compared to the Modelling Method in that Column, Such as the Following: 8) Fatigue [96]; 9) Channelling [97]; 10) Chemical Composition of Clusters [98,6]; 11) TEM Samples [57]; and 12) Magnetic Domain Patterning [27]

2. MATERIALS

Materials are usually separated into structural components, claddings and fuels because their matrices are often very different. However, there are overlaps, and materials are often considered for dual applications. Table 1 (based on [10] and [11]) summarises the range of candidate materials under consideration and investigation for the various Gen IV reactors that have to deal with the phenomena summarised in table 2. One of the candidate materials is the oxide dispersoid strengthened (ODS) ferritic/martensitic steel, which is the focus of this paper.

Oxide dispersoids are included in materials to improve their high temperature resistance properties, particularly because of their ability to pin dislocations and defects such as helium and therefore repress void swelling [12,13]. Indeed, ODS ferritic/martensitic steels have the advantage of high tensile, creep and fatigue strength. The microstructure resulting from an accumulation of defects leads to a hardening process. As such, an understanding of the role of ODS-dislocation interactions and estimates of the yield strength are important. A recent comprehensive review describes the state-of-the-art oxide dispersoids in ferritic steels [14].

Claddings encapsulate the fuel and are therefore exposed to microstructural and chemical degradation as a consequence of neutron irradiation, fission product interaction and their diffusion within the material's matrix, fuel-cladding interactions, and compatibility on the coolant side. Depending on the temperature, various materials are candidates—see table 1.

3. MULTI-SCALE MODELLING OF MATERIALS ISSUES

Candidate materials are faced with higher temperatures, fatigue, crack growth, stress rupture, creep, failure, creep-fatigue, thermal aging, corrosion, impurities such as helium and oxygen, and diffusion which leads to voids

Table 1. Comparison of Gen IV Reactors in Terms of Temperature, Pressure, Dose rate, and the Damage Mechanisms they are Subjected to, as well as the Critical Components, Types of Materials Envisaged, and the Types of Design Rules Usually Used to Study Them. Adapted from [10] and [11]

	PWR	SCWR	VHTR	SFR	LFR	GFR	MSR
Coolant inlet temp (°C)	290	290	400-600	370	600	450	700
Coolant outlet temp (°C)	320	500	950	550	800	850	1.000
Pressure (MPa)	16	25	7	0.1	0.1	7	0.1
Max. rad.dose (dpa)	100	10-70	1-10	200	200	200	200
coolant	water	water	helium	Liqu. sodium	Liqu. Pb/PbBi	He/CO2 supercooled	Molten salt
Critical components	RPV, internals, cladding	RPV, internals, cladding	RPV, core, IHX, heat coupling	cladding	cladding	Fuel/core	core
metals	Ferritic steels, Zircaloy	Ferritic steels, Ni-base,ODS	F-M steels, Ni-base, ODS	F-M steels, ODS	F-M steels ODS	F-M steels (RPV)	Ni-Base
ceramics			Graphite, C/C, SiCf/SiC, SiC			SiC, TiC Other ceramics	graphite
Main damage mechanisms	corrosion, embrittl. LCF	corrosion, embrittl. LCF	HT-corr. creep, LCF	corrosion, creep (th/irrad), LCF, irrad.	corrosion, creep (th/irrad), LCF, irrad	corrosion, creep (th/irrad), LCF, irrad.	corrosion, creep (th/irrad), LCF, irrad.
Design rules	RCC-MR ASME	RCC-MR ASME to be mod.	RCC-MR ASME (modif. in progress)	RCC-MR, ASME (to be modified/ developed)	to be developed	to be developed	to be developed

Table 2. Comparison of Technical Relevant Problems with Related Physical Effects

Physical Phenomena	Technical Relevance
Condensation and diffusion	Phase Diagram, time-temperature-phase diagram, microstructural stability,
Dislocation – obstacle interactions	Effects of precipitates, dispersoids and point defect clusters on yield strength, stress rupture stress, and creep strength
Dislocation-dislocation interactions	Dislocation arrangements, Yield stress (shear stress), fatigue, creep-fatigue
Point defect – defect and boundary interactions	Effects of irradiation on existing voids at boundaries (void growth, void shrinkage)
(Grain) boundary diffusion	creep damage, segregation, toughness/embrittlement
Decohesion of lattice	Crack formation and rupture
Surface phase formation	Oxidation and Corrosion
Effect of He	Bubble formation, swelling cracking, embrittlement
Fission gas release	Fission gas transport, swelling, fatigue, cracking
Actinide and oxygen redistribution	Thermo-chemical stability
Effect of Minor actinide	Stability, phase diagram, waste disposal

and bubble nucleation in grain boundaries (GBs) with the growth of cracks and irradiation. These issues all require a multi-scale modelling approach that encompasses atomistic investigation of a single defect movement, dislocation interactions and GB sliding, meso-scale modelling for dislocation and grain-based issues, and macro-scale modelling for the purpose of inclusion into constitutive equations (see figure 2). These issues are discussed next in terms of modelling methodologies that can and are being performed.

3.1 Fundamentals

For ferritic steels, the first step is to understand the main components of these materials: namely Fe, Cr, C and dispersoids. A limited amount of research has been performed on the Fe-C system [15,16] with an empirical potential developed to study diffusion with low C concentration [17]. The lack of research performed in C is due to the difficulty of simulating *ab initio* supercell sizes that are large enough to depict the low C concentration present in ferritic steels. Nevertheless, an Fe-C potential has just been developed [18], and the first results of larger scale simulations should soon be available. Note also the *ab initio* work performed on Fe-C to date can also facilitate the study of steel-reinforced concrete used in the storage of spent fuels.

Ferritic steels are made up of magnetic elementary components: these components affect the stability configuration and defect mobility of the steel [19] and lead to differences in the final defect state of the material, thereby altering its mechanical properties. Indeed, magnetism has been found to be the reason for the minimum energy configuration of self-interstitial Fe, which contrasts all other bcc non-magnetic transition metals [20]. The inclusion of anti-ferromagnetic Cr into the ferromagnetic Fe matrix further complicates the magnetic properties of the material. Experimental research has shown that Cr in Fe affects the point defect and interstitial cluster mobilities, as well as the dislocation propagation and multiplication mechanisms [21-23]. Projector augmented wave (PAW) calculations show that the sign of the mixing enthalpy changes at 6%Cr, predicting a phase separation at higher Cr concentrations and a solid solution for lower concentrations [24-26]. The inclusion of local effects and spin-orbital coupling due to the anisotropic behaviour of Fe-Cr, implemented with the Full-potential Linearised Augmented Plane Wave (FLAPW) method (e.g. [27,28]) reveal that the internal positions of the Cr atoms at the different Cr concentrations are of the utmost importance due to the changes that occur in the lattice parameter and Cr moments and provide an explanation of the large fluctuations seen in [25] due to the different local structural environment of Cr. Those studies show that the coordination shell distances are dependent on the orbital and spin moments and reveal the importance of including the spin-orbital moments in such calculations.

Likewise, nickel and uranium, which are also magnetic [29], may need to be revisited to investigate the role of magnetism in their structure. With regard to plutonium, such investigations are underway and, although plutonium is non-magnetic as such, the spin-orbital coupling plays a major role in the structure of the material [30,31]. Clearly, unless the important mechanisms are properly understood and included at the quantum mechanical level, any modelling predictions of fuel behaviour will be erroneous.

3.2 Clustering, Segregation and Evolution of Mechanical Properties

The high temperature expected in future Gen IV reactors is likely to instigate deformation processes, including microstructural or nanostructural changes such as dislocation movement and arrangement, and interface or GB sliding. These processes are affected by the density of point defects or the point defect clusters introduced by irradiation. Thus, it is important to understand the kinetics of vacancies, interstitials, clustering and cascade ageing.

The defects produced in an irradiation environment affect the deformation processes that depend on these defects and their evolution in time. The defects are formed when high energy particles displace atoms in the material and induce an area of disorder. Initial modelling investigations of irradiation were performed on the basis of Rate Theory, where sinks and their strengths were used to study the evolution of defects [32]. To obtain a detailed quantitative understanding of irradiation, it was realized that input from smaller time and length scale simulations are necessary. This necessity led to the study of irradiation by means of Molecular Dynamics (MD) simulations for ballistic and spike phases and Kinetic Monte Carlo (KMC) simulations for longer term evolution. In MD simulations the initial damage state in the first 100ps has been investigated [33]. GBs have also been included in such irradiation simulations, revealing the irradiation resistance of GBs and nanocrystal sized grains due to their sink strength [34-37].

Clustering, whether irradiation induced or not, and the long term evolution of defects, activation energies of their migrations, diffusion of impurities which lead to the formation of precipitates [38,39] can be studied using a combination of *ab initio* and KMC calculations (and sometimes go further afield by including MD simulations). Such DFT-KMC studies have yielded a description of the low-temperature annealing stages of the resistivity of electron irradiated Fe [40] and the desorption mechanisms in He-implanted Fe at different temperatures [41]. Investigations of the state of the Fe-Cr alloy have also been studied due to the different states possible as a consequence of Cr concentration (below 6%Cr, ordered material; above there is an inhomogeneous distribution which leads to segregation and clustering). How and why clustering occurs is important in establishing defect interaction and mobility in the background medium. Such combined *ab initio* and KMC investigations have shown that in the 8-11% Cr range a temperature dependent decrease of the short range order at low Cr concentrations and a minimum occurs [42] and that the inversion of the short range order parameter is due to the formation of stable α' precipitates.

One of the main causes of irradiation-assisted stress corrosion cracking (IASCC) comes from radiation-induced segregation (RIS) at GBs. The use of Rate Theory models in studies of RIS at GBs has a long history (see [43]). In one investigation [44], the GB sink strength was incorporated as a function of the misorientation angle of the GB taken from MD and MS simulations [45]. The simulations reproduced experimental results obtained by electron and proton irradiation. Furthermore, the Solute Drag method within Rate Theory has also been used to study shifts in the DBTT [46]; this method has successfully predicted the radiation-induced impurity segregation in 9Cr ferritic steel, showing that free carbon plays a significant role in impurity segregation.

3.3 Swelling

When a certain number of stable voids are created in a material, vacancies will congregate at voids and interstitials at dislocations, such that swelling becomes a segregation process. Iron and ferritic-martensitic alloys have low swelling or large incubation periods, so the question of void formation in these materials became the early subject of modelling. Swelling studies, including flux and temperature dependent transient delays, recognise that the material properties are dependent on the initial microstructural evolution. Swelling has been successfully modelled using Rate Theory [32] with the inclusion of sink strength to study the microstructural evolution of the material [47,48]. To study nucleation, the master equation/ Monte Carlo hybrid method, a rate theory approach, which includes defect trapping [49] as well as Fockker-Planck methods [50,51] have improved the original rate theory investigations.

Swelling due to impurities such as He (transmutation products) and phase reactions (e.g. amorphization) is an effect of neutron irradiation. The nucleation of voids and bubbles and their diffusion through the material differs as a consequence of temperature. The agglomeration of bubbles finally leads to crack nucleation and propagation. Cracking also occurs as a consequence of cyclic stresses and strains which were not anticipated at the design stage. This cracking determines to some extent the fatigue life of the material. Modelling of such phenomena on the multiscale needs *ab initio* calculations [52] and MD simulations [53-55] of He in Fe/Cr in order to define the energies and state of He- vacancy clusters which can then be used as input into KMC or Dislocation Dynamics (DD). Within the KMC framework, this has lead to He desorption experiments [56]. On a longer length scale, He and dispersoids interactions with dislocations in an ODS ferritic steels have been studied using DD [57-60] simulations where the inclusion of He induces an increase the critical resolve shear stress by a factor of two relative to He free samples (unirradiated samples [57,58]). Figure 3 encompasses the multi-scale modelling of He in Fe described above.

3.4 Creep

Stress rupture at high temperature can be studied by understanding creep. Stress rupture assumes dislocation

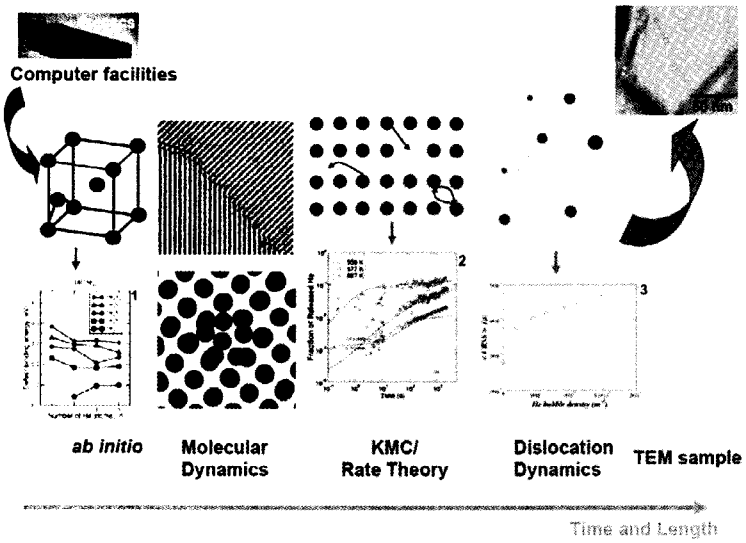

Fig. 3. Study of Helium in an Iron Matrix on the Multi-scale. Various Computing Facilities are Used for the Modelling (e.g. [99]). At the ab Initio Level, the Types of Results that can be Obtained are the Binding Energies as a Function of He [52]. MD Simulations can be Used to Study Larger Helium Clusters and Helium that Goes to GBs [94]. For Longer Atomistic Time and Length Scales, Rate Theory Yields Results Such as the Fraction of Helium Released [41,56]. For the Meso-Scale, DD Calculations Provide Information on the Critical Shear Stress of a Dislocation that Moves through a Field of ODS and He Particles and Reveals the Role of Helium in the Sample [60]. The Ultimate Goal being to Reproduce what is Seen Experimentally: e.g. TEM Samples [95]

movement and climb GB sliding, GB void formation and void growth as the main damaging mechanisms. The presence of irradiation-damage complicates the situation because there are a certain number of point defects (such as vacancies and interstitials) per unit of creep time, which may interfere with the moving dislocations and therefore can enhance the growth rate of the GB voids. Both point defect clusters and small helium bubbles can act as pinning points for dislocations, thereby reducing the creep rates. These questions need to be addressed when lifetime assessments under creep and irradiation are performed. After the introduction of quantitative descriptions of creep and creep damage in metals in the middle of the last century (such as the Norton equation and the Monkman-Grant rule), it took about 25 years to develop an understanding of creep-fatigue interactions [61]. The introduction of damage mechanics in terms of sub-critical crack growth and the introduction of constitutive laws for creep-fatigue interactions [62] lead to further improvements in lifetime assessments of structures. Finite element codes are still being used to study creep [63] by including, for example, a theta projection creep algorithm [64]. For a more quantitative understanding in the future, combinations of *ab initio* MD and DDD simulations can provide information on stress-strain curves; that information can then be incorporated into quantitative descriptions to go beyond the current constitutive equation approach.

3.5 Corrosion

Corrosion normally remains confined to surface or near surface areas (GBs) and can be considered as an additional effect, which can be coupled to irradiation damage in irradiation assisted stress corrosion cracking (IASCC). IASCC would need an atomistic approach to understand the movement and effect of oxygen in iron at and near the surface. One aspect of corrosion is the sink strength of surfaces and GBs. Stress rupture is also dependent on GB and nucleation and growth of voids therein. MD studies of water moving to Fe and Fe-Cr surfaces [65] show that the charged oxygen atoms break the metal-metal bond by bonding themselves with the metallic atom, indicating the formation of a surface oxide layer. These MD studies also show the important role that the oxide layer plays in the nucleation of such phenomena as stress corrosion cracking [65].

The IASCC of internals is being modelled in the PERFECT program [8] by using a combined multi-pronged approach, in which previous experimental data is used to parametrize the modelling. This approach has identified various mechanisms and critical factors, such as the effects of dislocation channeling and RIS. Together with the modelling of the local electrochemistry in cracks and crevices and the post-irradiation mechanical behaviour, the information is used to produce a Finite Element Method (FEM) model of the growth rate of stress corrosion cracking. Particular attention has been paid to the process of stress/strain concentrations associated with dislocation channeling. Finally, a model for crack initiation has been developed on the basis of statistical nucleation and deterministic growth and coalescence.

3.6 Fracture

Fracture has been studied most effectively with multi-scale modelling in the PERFECT program [8], which has already achieved a high degree of integration. Atomistic simulations such as those described above, are implemented and provide a description of the critical resolve shear stress, together with a model for microcrack behaviour. To study the mechanics of the material a first meso integration at the level of grain aggregate has been produced. In the case of RPV steels, a cleavage criteria at the bainite larh level has been developed, together with a model for P-induced brittle IGF under irradiation is included together with other models for brittle behaviour and macro-scale models as a function of the loading and irradiation conditions. In that work, the local fracture criterion is used to predict the fracture toughness [8]. A local probability of fracture was derived from experimentally determined defect size distributions and a continuous DD model was used to study the brittle-ductile fracture toughness transition. Furthermore, the brittle-to-ductile transition and fracture toughness models were developed with the Local Approach and continuous dislocation dynamics DD, respectively. With the incorporation of Charpy-V impact test data, the Local Approach was also used to predict the cleavage fracture toughness behaviour.

3.7 Lifetime Predictions

Lifetime predictions under a creep-fatigue loading are a very well known problem in materials science. The yield stress, creep rate, and crack-growth, to mention only a few of the many materials properties, heavily depend on the microstructure. However, a conversion of micro- and nanostructural features into stresses and/or strains is still absent. Constitutive relations connect applied stresses or forces to strains or deformations. They form the basis of finite element calculations, design rules and codes. Thus, curve fitting techniques are used as a basis for design. During the past 40 years several methods were developed which are mainly based on ideas which were already summarized in the early 1970s [66]. None of these models, however, managed to become part of a design code. Design is still based on a simple linear life fraction rule. Recent developments within ASME try to improve lifetime prediction under creep-fatigue loading. An example is the investigation of ferritic martensitic modified 9Cr 1Mo steels (grade 91) [66]—see figure 4. Decomposition of an inelastic creep-fatigue strain range ($\Delta\varepsilon_{in}$) into a plastic strain range ($\Delta\varepsilon_{pl}$) and a creep strain range ($\Delta\varepsilon_c$) allows the determination of the number of cycles to failure by the simple relation:

$$N = 1/A(\Delta\varepsilon_{pl})^{\alpha} + 1/B(\Delta\varepsilon_c)^{\beta},$$

where A, B, α, and ß are fitting constants and $\Delta\varepsilon_{in} = \Delta\varepsilon_{pl}$

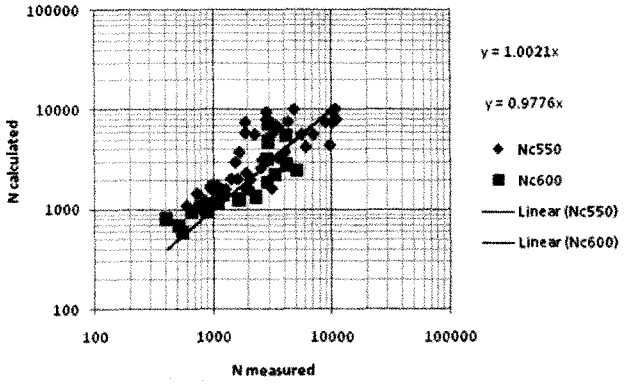

Fig. 4. Demonstration of the Validity of this Vpproach. Although the Prediction is Quite Accurate (within the Range of Scatter of Pure Fatigue Data) the Physical Interpretation of the Data is Missing

$+(\Delta\varepsilon_c)$. Such results, however, must be validated experimentally with long lifetimes. The validation is based on relatively short-term laboratory data (with hold-times mainly in the range of minutes to hours) under a uni-axial state of stress. The influence of microstructure cannot be predicted and the stage of early crack-growth is not included. The question as to whether this approach is generally valid for ferritic and martensitic materials cannot be easily answered. It is hoped that at least predictions about trends can be obtained from advanced microstructural modelling techniques and incorporated to obtain a more robust prediction, as was the case for the example in [67].

3.8 Fuels

New and advanced fuels for Gen IV reactors exposed to extreme conditions need to be designed with a high burn-up capacity, high density, high melting temperature, high conductivity, modest linear expansion coefficients, they should not interact with cladding in a detrimental manner, they should be safe, have proliferation resistant manufacturing and minimal (or optimally zero) radioactive waste. Methods of recycling fuel rods and reducing their radioactivity are now being conducted and investigated and must go hand in hand in any future renaissance of nuclear energy reactors. Although less developed than its structural materials modelling counterpart, a surge in modelling fuels on the multi-scale has recently emerged, such as the co-ordinated Framework 7 project F-Bridge [68], as well as the study of heat transport phenomena [67]. Important issues that have already been studied include: how the material deforms under irradiation [69]; the high burn-up structure (HBS); and the fundamental processes that lead to nucleation of HBS and the role of GBs [70,71]. At the MD level, an empirical potential that properly predicts defect configurations needs to be implemented, with the current potentials each having their pros and cons [74,75]. Most recently charge transfer

has been included in such empirical potential [76]. MD studies have focused on the mobility and diffusion of interstitials [72,73] as well as the effects of fission products, such as Xe [77] and He [78,79]. At the continuum, pore formation in the HBS has been modelled with a one-dimensional mass balance model [80,81]. In regard to waste, issues such as corrosion and actinide release need to be understand in order to eradicate them [82,83].

4. VALIDATION EXPERIMENTS

To best use computational tools it is also necessary to assess existing data and optimise new experiments, the results of which should be incorporated into the simulation code in order to develop realistic models which will allow extrapolation beyond the existing databases for lifetime prediction. It is therefore important to have an iterative program with complementary experiments for the purpose of verifying, validating and further developing the computational models. This interchange between computational models and experimentation is critical for developing accurate predictive models (as exemplified in figure 2).

A wide range of experimental procedures can now be used to gain a better understanding of the different properties of materials. In the scope of approaching materials with a modelling scheme, experiments are slowly emerging that can be used to validate the simulations performed, provide vital information to allow the improvement of the modelling schemes, and to develop important experiments that should be performed through the guidance of modelling results.

A quantitative analysis of the microstructure is also needed for a better understanding of damage with time. Improvements in microstructural and nanostructural investigation methods (quantitative electron microscopy and microanalysis, high resolution transmission microscopy, atomic force microscopy, and synchrotron beamline technologies [84]) together with sophisticated specimen preparation (such as the use of the focused ion beam method) enable the local mechanical properties to be directly coupled with the local microstructure. Facilities such as the Joint Accelerators for Nano-science and Nuclear Simulation (JANNUS) facility [85], which perform controlled experiments, are also emerging as a validation tool for modelling. Ion (He) and proton irradiation in linear accelerators is a multi-scale approach that speeds up the formation of damage, provides a powerful tool for the production of irradiation damage, and has been taken up by fusion and fission material science programs [86,87]. The disadvantage, that only small areas in the material are damaged, can be counterbalanced by using local analysis techniques. Thus neutron irradiations can be confined to benchmark testing and to qualifying assumptions and modelling based on ion/proton damage. Bulk material behaviour of small volumes can be understood using thin strip tensile/creep tests, miniaturised three-point tests and small punch tests [88]. A correlation with larger volumes can then be established by performing finite element calculations [89]. Investigations performed in this way are important for understanding which mechanism is present in the material depending on the temperature that the material is subjected to.

Often it is necessary to study damage in even smaller volumes (due to issues such as high activity of samples, lower energy of ions, local microstructure of interest). The local mechanical properties can be determined with micro-hardness, nano-indentation and nano-pillar-compression tests. Effects such as irradiation hardening, temperature effects and hardening or softening as a result of irradiation-induced phase transformations or segregations can be monitored. Indentation can be used to study irradiation hardening in order to study stress-strain curve expectations [90]. Using a combination of advanced microstructural investigations of TEM and focused ion beam (FIB), the interactions of dislocations with point defect clusters around the indent are then visible. Similar results can be obtained with the investigation of irradiation damage in ceramics [91].

The future X-ray free electron laser (XFEL) [92,93] will enable a direct comparison of more fundamental issues to bring forth an understanding in the local mechanical properties and the local microstructure. Investigations of local properties of small samples are expected to act as fingerprints of the more general properties of the material [84].

5. CONCLUSION AND FUTURE DIRECTIONS

A new wave of peaceful interest in nuclear energy has emerged as one of the possible energy sources to help meet the future world energy supply needs which is environmentally friendly by maintaining a low emission of green house gases. Lifetime assessments of components for Gen II, III and IV and fusion reactors require an understanding of the single damage effects and their interactions. In this framework, modelling emerges as a very effective tool to complement experiments, and even to design future experiments. The potential of a multi-scale approach for a quantitative multi-scale assessment of damage and damage evolution in the materials of future fusion and fission reactors (such as Gen IV) has been discussed in the context of materials issues, showing implementation of the multi-scale modelling paradigm and complementary model validation. The strategy entails simulations from the micro-scale to the macro-scale and encompasses *ab initio*, MD, KMC, rate theory, DDD, FEM, and continuum models to study the fundamentals behind the problem: combining microscopic techniques from synchrotron irradiation

facilities and *ab initio* calculations to understand the structure and magnetic properties of the material; irradiation, nanoindentation, using MD and KMC to understand the microstructural mechanisms present; hardness, and strain using DDD to understand the mesoscopic mechanisms will produce an overall understanding of the material's behaviour which will help to predict its lifetime behaviour and is expected to help to in the a predictive methodology for material's development. Incorporation of such data into existing design codes will enable tremendous improvement in material property predictions.

The modelling paradigm presented above is still in its infancy. Computing facilities in one way or another limit the length and time scales of each of the codes discussed, with the consequence that, until now, only simple binary and tertiary alloy systems have been tackled. Research on carbon, the first step towards a real steel, has only just begun. The quality of empirical potentials used in MD simulations are improving, so that databases of simulations have to be re-evaluated. Inclusion of temperature, whether low, or high, is not always so simple, with *ab initio* and DDD still calculated at zero Kelvin and MD limited to the temperature range where the empirical potential is valid. Model validation then becomes even more vital to such modelling schemes and at the same time more complex to perform to deal with similar temperatures, purity of the materials, and time and length scales which are on par. Regardless of these drawbacks, using a multi-scale modelling approach to understand a material brings forth much information on the mechanisms which influence the material at various scales and play a role in determining the aging of the material under various conditions.

Coupling computational multi-scale modelling with experimental validation will produce a robust research activity to understand materials' behaviour, which probes beyond current experimental approaches, as well as allowing the set-up of better tuned experiments. The resultant conglomeration of information with the incorporation of important competing mechanisms will yield a more robust modelling scheme. Thus, for a better understanding of a material's mechanical properties and its lifetime, it is necessary to tackle the material with a multi-scale multi-disciplinary experimental and modelling approach. In the long term, modelling activities are expected to become an effective toolbox for finding optimal materials. Indeed, the know-how that is expected to be built up in the coming years will enable modellers to extend their research to more complex structures and a broader range of materials.

REFERENCES

[1] T. Allen, H. Burlet, R.K. Nanstad, M. Samaras, S. Ukai, *MRS Bulletin* **34**, 20 (2009).

[2] http://www.gen-4.org/PDFs/GenIVRoadmap.pdf

[3] ASME Boiler & Pressure Vessel Code, Section III, ASME BPVC (2007).

[4] RCC-MR code, AFCEN (2007).

[5] M. Samaras, W. Hoffelner, M. Victoria, *J. of Nucl. Mater.* **371**, 28 (2007); M. Samaras, W. Hoffelner, C.-C. Fu, M. Guttmann, R. E. Stoller, *ICAPP 2007* proceedings; *Revue Générale Nucléaire*, SFEN, France.

[6] M. Samaras, M. Victoria, *Mat. Today* **11**, 54 (2008).

[7] M. Samaras, M. Victoria, W. Hoffelner, 'E.U. Advanced Materials Modeling Perspectives', accepted *J. Nucl. Mater.* (2008).

[8] www.fp6perfect.net/perfect

[9] M. Victoria, *et al.*, O1B-I-28, SOFT, *Fusion Engineering and Design*, **82**, 2413 (2007).

[10] http://www.nea.fr/html/science/struct_mater/Presentations/ZINKLE.pdf

[11] http://www.nea.fr/html/science/struct_mater/Presentations/CARRE.pdf

[12] S. Ukai, M. Fujiwara, *J. Nucl. Mater.* **307-311**, 749 (2002).

[13] J. Wunder, PhD thesis 'Mikrostrukturelle Beschreibung der Warmfestigkeit ferritischer Superlegierungen', Friedrich-Alexander Universität, Erlangen-Nürnberg, Germany, (1994).

[14] G.R. Odette, M.J. Alinger, B.D. Wirth, *Annu. Rev. Mater. Res.* **38**, 471 (2008).

[15] C. C . Fu, F. Willaime, *C. R. Physique* **9**, 335 (2008).

[16] C. Domain, C. S. Becquart, J. Foct, *Phys. Rev. B* **69**, 144112 (2004).

[17] C.S. Becquart, J.M. Raulot, G. Bencteux, C. Domain, M. Perez, S. Garruchet, H. Nguyen, *Comp. Mater. Sci.* **40**,119 (2007).

[18] http://homepages.ed.ac.uk/graeme/moldy/moldy.html

[19] C. C. Fu, F. Willaime, P. Ordejon, *Phys. Rev. Lett.* **92**, 175503 (2004).

[20] D. Nguyen, et al., *Phys. Rev. B* (2006) **73**, 020101 (2006).

[21] Garner F. A., et al., *J. Nucl. Mat.* **123**, 276, (2000).

[22] A. Kohyamaet. al., *J. Nucl. Mat.* **138**, 233, (1996).

[23] Abe H., and Kuramoto E., *J. Nucl. Mater.* **209**, 271 (1999).

[24] P. Olsson, I. A. Abrikosov, L. Vitos, J. Wallenius, *J. Nucl. Mat.* **321**, 84 (2003).

[25] T.P.C. Klaver, R. Drautz, M. W. Finnis, *Phys. Rev. B* **74**, 094435 (2006)

[26] T. P. C. Klaver, P. Olsson, M. W. Finnis, *Phys. Rev. B* **76**, 214110 (2007).

[27] A. Froideval, R. Iglesias, M. Samaras, S. Schuppler, P. Nagel, D. Grolimund, M. Victoria, W. Hoffelner, *Phys Rev Lett.* **99**, 237201 (2007).

[28] R. Iglesias, M. Samaras, A. C. Uldry, M. Victoria, W. Hoffelner, work in preparation.

[29] U. Köbler, A. Hoser, R.M. Mueller, K. Fischer, *Journal of Magnetism and Magnetic Materials* **315** 12(2007)

[30] R. H. Heffner, E. D. Bauer, B. Chung, M. J. Fluss, W. Higemoto, T. U. Ito, D. E. MacLaughlin, L. A. Morales, G. D. Morris, K. Ohishi, J. L. Sarrao, L.Shu, Proc. of 5th Int. Symposium on ASR-WYP-2005-Advances in the Physics and Chemistry of Actinide Compounds-*J. Phys. Soc. Jpn.* **75**, 14 (2006).

[31] M. Fluss, S. Mccall' Talk presented at Fall MRS 2008, Symposium R, 'Experimental Determination of Metal Fuel Point Defect Parameters', www.mrs.org/s_mrs/doc.asp?CID=16988&DID=217327

[32] G. Martin, P. Bellon, *R. Physique* **9**, 323 (2008).

[33] N. Soneda, T. Diaz de la Rubia, *Phil. Mag. A* **78**, 995 (1998).

[34] M. Samaras, P.M. Derlet, H. Van Swygenhoven, M. Victoria, *Phys. Rev. Lett.* **88**, 125505 (2002).
[35] M. Samaras, PM. Derlet, H. Van Swygenhoven, M. Victoria, *J. Nucl. Mater.* **351**, 47 (2006).
[36] M. Samaras, W. Hoffelner and M. Victoria, *J. of Nucl. Mater.* **352**, 50-56, (2006).
[37] M. Samaras, W. Hoffelner and M. Victoria, submitted to *MRS proceedings* (2008).
[38] C.S. Becquart, A. Souidi, C. Domain, M. Hou, L. Malerba, R.E. Stoller, *J. Nucl. Mater.* **351**, 39 (2006).
[39] C.S. Becquart, *Nucl. Inst. Meth. Phys. B* **228**, 111 (2005).
[40] C.C. Fu, J. Dalla Torre, F. Willaime, J.-L. Bocquet, A. Barbu, *Nature Mater.* 4, 68 (2005).
[41] C. Ortiz, M. J. Caturla, C. C. Fu, F. Willaime, *Phys. Rev. B* **75**, 100102(R) (2007).
[42] P. Erhart, et al.,, *Phys Rev B* **77**, 134206 (2008).
[43] R. G. Faulkner, *Encyclopedia of Materials: Science and Technology*, 829 (2008).
[44] N. Sakaguchi, S. Watanabe, H. Takahashi, R. G. Faulkner, *J. Nucl. Mater.* **329-333**, 1166 (2004).
[45] S. Watanabe, Y. Takamatsu, N. Sakaguchi, H. Takahashi, *J. Nucl. Mater.* **283-287** 152 (2000).
[46] Z. Lu, R.G. Faulkner, N. Sakaguchi, H. Kinoshita, H. Takahashi, P.E.J. Flewitt, *J. Nucl. Mater.* **329-333** 1017 (2004).
[47] A.D. Brailsford, R. Bullough, *J. Nucl. Mater.* **44**, 121 (1972).
[48] A.D. Brailsford, *J. Nucl. Mater.* **118** 303 (1983).
[49] M. Michael P. Surh, J.B. Sturgeon , W.G. Wolfer, *J. Nucl. Mater.* **328**, 107 (2004).
[50] A A Semenov, C H Woo, *Appl. Phys. A* **69**, 445 (1999).
[51] A A Semenov, C H Woo, *J. Nucl. Mater.* **323**, 192 (2003).
[52] C. C. Fu, F. Willaime, *Phys. Rev. B* **72**, 064117 (2005).
[53] R. Schäublin, Z.L. Chiu, *J. Nucl. Mater.* **362**, 152 (2007) .
[54] F. Gao, et al.,, *J. of Nucl. Mater.* **133**, 351 (2006).
[55] Osetsky Yu N. ., *Mater. Res. Soc. Symp. Proc.* **59**, 527 (1998).
[56] M. J. Caturla, C. J. Ortiz, *J. of Nucl. Mater.* **141**, 362 (2007).
[57] B. Bako, D. Weygand, M. Samaras, J. Chen, M. Pouchon, P. Gumbsch, W. Hoffelner, *Phil. Mag. A* **87** 3645 (2007).
[58] B. Bakó, D. Weygand, M. Samaras, W. Hoffelner, and M. Zaiser, *Phys. Rev. B.* **78**, 144104 (2008)
[59] B. Bakó, M. Zaiser, D. Weygand, M. Samaras, W. Hoffelner, *J. Nucl. Mater.*, in press (2009).
[60] B. Bakó, M. Samaras, D. Weygand, J. Chen, P. Gumbsch, W. Hoffelner *J. Nucl. Mater.*, in press (2009).
[61] Fatigue at elevated temperatures, A. E. Carden, A. J. McEvJily, C. H. Wells, editors, *ASTM Special Technical Publication, 520, AMERICAN SOCIETY FOR TESTING AND MATERIALS*, (1973).
[62] J. Lemaitre, J.-L. Chaboche Eds., *Mechanics of Solid Materials*, Cambridge University Press ISBN 0521477581, 9780521477581 (1994).
[63] D. Cole et al, *Sic Tech. Wld. Join.* **5**, 81 (2000).
[64] M. Law, W. Payten, K. Snowden, *International Journal of Pressure Vessels and Piping* **79**, 847 (2002).
[65] N. K. Das, K. Suzuki, Y. Takeda, K. Ogawa, T. Shoji, *Corros. Sci.* 50, 1701 (2008).
[66] W. Hoffelner, ASME tasks on New Generation Nuclear Plant (NGNP) 2008, unpublished.
[67] M. Stan, J.C. Ramirez, P. Cristea, S.Y. Hu, C. Deoa, B.P. Uberuaga, S. Srivilliputhur, S.P. Rudin, J.M. Wills, *Journal of Alloys and Compounds*, **444-445**, 415 (2007).
[68] www.f-bridge.eu/; www.f-bridge.eu/index.php/Papers/F-Bridge-Publications.html
[69] L. Van Brutzel, P. Crocombette, *MRS Proceedings* **358**, n°2-3, 209 (2006).
[70] L. Van Brutzel, E. Vincent-Aublant, *J. Nucl. Mater.* **377**, 522 (2008).
[71] Hj. Matzke, M. Kinoshita, *J. Nucl. Mater.* **247**, 108 (1997).
[72] G. Martin, S. Maillard, L. Van Brutzel, P. Garcia, B. Dorado, C. Valot, *J. Nucl. Mater.* (2009).
[73] G.E. Murch, *Diff. Defect Data* **32** 9 (1983).
[74] K. Govers, S. Lemehov, M. Hou, M. Verwerft , *J. Nucl. Mater.* **366** 161 (2007).
[75] K. Govers, S. Lemehov, M. Hou, M. Verwerft, *J. Nucl. Mater.* **376** 66 (2008).
[76] M. Iannuzzi, M. Krack, M. Zimmermann, M. Samaras, unpublished.
[77] K. Govers, S. Lemehov, M. Verwerft, *J. Nucl. Mater.* **374**, 461(2008)
[78] Y. Yun, O. Eriksson, P. M. Oppeneer, *J. Nucl. Mater.* accepted (2008).
[79] David C. Parfitt, Robin W. Grimes, *J. Nucl. Mater.* **381** 216 (2008).
[80] P. Blair, A. Romano, Ch. Hellwig, R. Chawla, *J. Nucl. Mater.*, **350**, 232 (2006).
[81] P. Blair, thesis, http://library.epfl.ch/theses/?nr=4084
[82] L. Shuller, R. C. Ewing, U. Becker, *Mater. Res. Soc. Symp. Proc.* Vol. **985** 0985-NN12-03 (2007).
[83] M. Katayama, J. Adachi, K. Kurosaki, M. Uno, S. Miwa, M. Osaka, K. Tanaka, S. Yamanaka, *Mater. Res. Soc. Symp. Proc.* Vol. **1043** 1043-T09-06 (2008).
[84] W. Hoffelner, A. Froideval, M.A. Pouchon, J. Chen, M. Samaras, *Metall. Mater. Trans. A*, 10.1007/s11661-007-9326-z.
[85] www-ist.cea.fr/publicea/exl-doc/200700001902.doc
[86] A. Kohyama, GFR Steering Committee Meeting, Idaho May 21st, 2003.
[87] B. D. Wirth, M.J. Caturia, T. Diaz de la Rubia, T. Khraishi, H. Zbib, Nucl. *Instru. Meth. Phys. B* **180**, 23 (2001).
[88] W. Hoffelner, M. Pouchon, J. Chen, *TMS Letters Issue* **3**, 81 (2005).
[89] E. N. Campitelli, P. Spätig, R. Bonadé, W. Hoffelner, M Victoria, *J. Nucl. Mater.* **335**, 336 (2004).
[90] M. A. Pouchon, J. Chen, M. Döbeli, W. Hoffelner, *J. Nucl. Mater.* **352**, 57 (2006).
[91] A. Kohyama, Y. Katoh, L.L. Snead, R.H. Jones, www.iaea.org/programmes/ripc/physics/fec2000/pdf/ftp1_06.pdf
[92] B.D. Patterson, et al., *Swiss Physical Society Newsletter*, **23**, 16 (2003).
[93] J. F. van der Veen, *Synchrotron Radiation Instrumentation*, **3-9** 705, (2004).
[94] N. Gao, M. Samaras, P. M. Derlet, H. Van Swygenhoven, M. Victoria, W. Hoffelner, unpublished.
[95] J. Chen, M.A. Pouchon, A. Kimura, P. Jung, W. Hoffelner, *J. Nucl. Mater.* accepted.
[96] J. Chen, unpublished.
[97] P. de Almeida, M. Victoria, *Solid State Commun.* **125** 195 (2003).
[98] C. Borca, et al, unpublished.
[99] http://www.cscs.ch/