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MULTI-SCALE MODELLING OF RADIATION EFFECTS IN NUCLEAR MATERIALS

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1. Introduction

Even though they appear continuous, materials are discrete and made of atoms or, more precisely, of atomic nuclei (ions), surrounded by electrons. The description of many physical phenomena mandatorily requires that materials should be described as a set of discrete atoms. For example, the ballistic redistribution of the atoms of a solid when many of them are displaced by incoming particles involves mechanisms that strictly occur at the nuclear and atomic levels. Likewise, the identification of mechanisms whereby atoms of different species are rearranged by diffusion in solids involves the atomic structure of materials. Most physical properties of materials — e.g. optical, magnetic or thermal, as well as chemical — depend on the states of the electrons that surround the atomic nuclei. In theory, the behaviour of a material described as a set of atoms under given conditions could be predicted exactly by solving the time dependent Schrödinger equation for all the ions and electrons that compose it. This is, however, practically impossible, except in simple cases that involve a limited amount of ions and electrons. Thus, even if the atomic and discrete nature of materials is taken into account, approximate models need to be devised.

Despite the discrete nature of materials, for most applications the approximation of describing them as a continuum with uniform properties is appropriate. This is typically done for liquids in fluid mechanics and for solids in continuum mechanics, in the latter case to calculate the stress and strain field, or the temperature field, in all parts of a material subjected to a given mechanical and/or thermal external load. However, by looking at the structure of solids with microscopes of increasing magnification, intermediate structures can be identified that reveal discontinuities even without going down to the atomic level. In solids, grains and sub-grains can be recognised and the mechanical properties of, for example, metals depend enormously on how these grains are structured and change under the conditions to which the material is subjected. Each grain is a crystal in which atoms are distributed according to a repeating pattern, oriented in a certain way, but they are never perfect: defects — i.e. discontinuities in the perfect crystal structure — ranging from point defects — i.e. vacancies (missing atoms) or interstitials (atoms in excess) — to dislocations (lines resulting from the displacement of a block of material with respect to another block), determine, for example in metals, the properties of ductility or brittleness. In fact, materials, especially metals, would be in most cases useless without defects. These intermediate structures (grain boundaries, dislocations, point defects, etc.) can be taken as the unit building blocks to describe the behaviour of the material at that specific scale for a specific phenomenon. Accordingly, discrete modelling tools that describe atomic-level processes in solids (molecular dynamics, either based on quantum-mechanical electronic structure calculations — first-principles or *ab initio* — or, more frequently, on the use of classical empirical interatomic potentials), or the evolution of defects in them making abstraction from the underlying atomic structure (kinetic Monte Carlo, rate theory, dislocation dynamics, etc.) have been progressively developed in the last 60 years [1]. These tools, which rely heavily on the use of computers for simulations, have become increasingly widespread since the early 1990s, because the growing power of computers has allowed ever-more detailed and massive calculations of this type to be performed, and are now commonly used in such complex materials as multi-component iron alloys or actinide compounds.

Radiation damage in solids represents an extreme case in materials science, because the almost instantaneous displacement of atoms localised in a region of a few nanometres by an energetic impinging particle (e.g. neutron or fission product), repeated several times at different points of the material over time, is at the origin of changes in the performance of whole reactor components. For example, in the case of metals used as structural materials this leads to reduction of the amount of deformation experienced before breaking (reduction of *fracture toughness*), as is typical of a brittle material: a behaviour certainly not desired in a structural component. In nuclear fuel, the accumulation of defects and fission products, especially gases, produces cavities and bubbles, which induce in particular microstructure changes and macroscopic swelling, which in turn lead to cracking. So, in radiation damage, atomic- or even nuclear-level processes lead to changes in the way large components, such as a reactor vessel or a fuel pin, respond to loads. In order to be able to describe the physical behaviour of materials under irradiation beyond empiricism, therefore, it is necessary that one develops models for nuclear reactions (activation, transmutation, etc.), atomic-level processes (cascades of atomic displacements, diffusion of chemical species and of defects, etc.), nano- and micro-scale processes (development of nanostructural features such as dislocation loops, segregation of chemical species that change locally the property of the material leading to, for example, localised corrosion, etc.), meso-scale processes (movement of dislocations that govern plastic deformation, or changes in grain boundary behaviour, etc.), and finally macro-scale processes (crystal plasticity applied to aggregates of grains, or continuum mechanical calculations of stress and strain distributions in a component, etc.). In other words, the physical description of the behaviour of a material requires the use of models that address the correct scale for the phenomenon at hand. It also requires that the information about the behaviour of the material at a certain scale is somehow transferred to the higher scale, so that the model at this higher level can be more accurate. This is the principle of the multi-scale modelling approach [1].

2. Multi-scale modelling, experiments and technology

The development of multi-scale models belongs to fundamental science. The problems that need to be addressed and solved for their development encompass different fields of physics and materials science, from quantum mechanics to the theory of plasticity, including often the need to solve purely numerical problems for the implementation of a specific model or the development of a new modelling tool. It is also, inherently, a long-term research activity, which in some cases requires many years before getting to the point that a specific goal of practical relevance is reached: progress can only be incremental. However, this type of research has a strong interaction with, on the one side, experiments and, on the other, technology.

2.1. Modelling and experiments

No model with the ambition of helping in the interpretation of experimental data on a given phenomenon can be conceptually designed and built without a deep knowledge of the experimental phenomenology. One should know which problems should be studied, have as much experimental evidence as possible of the mechanisms involved, and dispose of sufficient data to elaborate a *conceptual* model to guide the development of actual *mathematical* models and calibrate them. This is very different from 'experimental validation'. Models, even multi-scale models that start from so-called *ab initio*, or more generally electronic structure calculations (i.e. calculations performed from first physical principles, without parameters, based on quantum mechanical theories and approximations, see for example Martin (2004) [2]), are not developed in a separate world of platonic ideas and then brought down to earth so as to be compared with experiments, to be 'tested' and to 'see if they work' (i.e. 'validated'). Models need to be informed on

experimental facts and these facts need to be explored at many different scales, using a combination of the most advanced materials examination techniques. In addition to mechanical testing for macroscopic properties (tensile, hardness, Charpy, fracture toughness, etc., as well as nanohardness or small punch tests [3]) and electron microscopy in all of its forms and varieties for microscopic properties [4], analytical techniques such as atom probe tomography [5], small-angle neutron scattering [6], positron annihilation spectroscopy [7], X-ray absorption spectroscopy [8], internal friction [9] and magnetic after-effect [10] are, by now, all combined, irreplaceable tools in the development of physical models that describe radiation effects, because of the detailed information they provide down to the least visible scales (see for example Meslin et al. (2010) [11] and Lambrecht et al. (2010) [,12]). The assumptions made in models must either be suggested by experiments, or deduced from more fundamental models. The first objective in the process of model development is not to make predictions to be verified experimentally, but to make sure that the model reproduces satisfactorily a set of available experiments, providing a key for their rationalisation and interpretation. These experiments must be as complete and detailed as possible, so that the model can be elaborated and verified in all or most of its aspects. In other words, **what a physical model is chiefly expected to do is give an explanation of why the experimental data are as they are**. The fact that experimental data are correctly reproduced by a physical model is likely to mean that the dominant mechanisms have been properly identified and quantified and the identification of the mechanisms can trigger a series of considerations on what can be done to keep a given phenomenon under control or to exploit it for given purposes. Only at an advanced level of development may one attempt *predictions* with the model and plan experiments to verify them. In practice, this does not occur very often. This is why the expression 'experimental validation' of models, as if models and experiments were two separate worlds compared only at the end of a process, is misleading and **one should rather talk of the 'combination of models and experiments in order to better understand reality'.**

One should also consider that advanced physical models and modelling tools are divided in two categories: those developed to investigate phenomena that *cannot* be observed experimentally and those developed to reproduce experimental results, with which they can be compared directly to help in their interpretation. The former type simply cannot be 'experimentally validated'. The obvious example of models that cannot be directly validated are atomistic simulation techniques used to study the evolution of displacement cascades in a material — i.e. the almost instantaneous displacement of thousands of atoms triggered by the collision of an impinging energetic particle (e.g. a neutron or a fission product) with an atomic nucleus. Most of what is known on displacement cascades comes from more or less advanced atomistic simulations and, to date, no experimental device or technique has been developed that would be able to follow the evolution of the cascade in space (a few nanometres) and in time (a few picoseconds), because of the smallness of the scales involved. Another example will help to understand the difference between the two types of models or modelling tools: let us imagine that we want to develop an accurate physical model that describes the formation of cavities in irradiated materials. The development of such a model will require knowledge of the diffusion coefficients of clusters of vacancies of all mobile sizes and there is no experimental technique in the world that is capable of following the motion of a vacancy cluster to provide such numbers. Atomistic simulation techniques (atomistic models) are the only hope to obtain an assessment of such quantity. They are therefore a tool, on the same footing as an experimental measurement, to obtain a quantity that we need to know so as to understand reality better. The value we obtain from the atomistic simulation might not be fully accurate, just as an experimental measurement is affected by uncertainties often not fully quantifiable. However, it is 'the best we can do' to produce data that we believe we need to better rationalise what we observe happening in reality. In some cases, the use of these fundamental models might trigger the experimental search of expected phenomena. For example, one might discover, using *ab initio* calculations, that a chemical element B has very strong affinity for self-interstitial atoms (SIAs) in a crystal of element A — i.e. that there is a strong binding in A

between the SIA and B atoms. Based on this fact, one may deduce, or at least suspect, that clusters of SIA, that form in A under irradiation and grow to become dislocation loops, visible in an electronic microscope, should contain element B: this is a 'prediction' that might be experimentally verifiable. This is, for example, what happened in iron–chromium (Fe–Cr) alloys, leading all the way to finding the likely reason for, among other things, reduced swelling in iron alloys that contain chromium [13] or the possible origin of higher radiation hardening in Fe–Cr as compared to Fe, as well as the minimum in the radiation embrittlement for 9 % Cr steels [14]. However, even if no B was found experimentally in dislocation loops in A, it would not necessarily mean that the SIA-B affinity does not exist: it might be limited to a few SIA or it might not be easily detected with the available experimental techniques. Verifying this requires, on the one hand, the development of proper interatomic potentials to go beyond the size limitation of *ab initio* calculations, as well as, on the other, the design of ever-more–refined experiments. The latter is unfortunately not always practically feasible, or might be extremely expensive, so in many instances one must be content with the indications coming from interatomic potentials, which become therefore very useful tools that compensate for the impossibility of doing corresponding experiments. Moreover, an interatomic potential can, in turn, be used to parameterise the second type of models.

The above-mentioned model that describes cavity formation is an example of this second type of models: it will be built based on information from experiments and from more fundamental models, in this specific case atomistic models used to assess the mobility of vacancy clusters. Such a type of model, by being in principle directly comparable with experiments, can be later used to 'validate' indirectly the numbers that come from 'invalidable' models. On the other hand, a model of this type, however accurate and physically justified, will always also contain assumptions that are not fully proven, or coefficients and parameters that are not fully known, no matter how much effort is made to obtain them *ab initio* or from experimental measurements. The model will therefore unavoidably require some 'calibration' that goes together with its validation. The process of calibration/validation requires that one disposes of data coming from a wide range of experimental techniques, because advanced multi-scale models provide information about virtually any detail of the changes undergone by materials under certain conditions, but there is never one single experimental technique that can provide all the required information. In this sense, experiments in support of the development of multi-scale modelling can be very expensive, delicate and time consuming, because they will require that specimens of the same materials (better if *model* materials, in which a certain phenomenon is better observed, or the complexity is reduced, so that the model can be better developed and tested) are analysed using various techniques, often in different laboratories [11, 12]. Eventually, the results must be consistently combined and this is of course done, in turn, with the help of models. But it is important to stress that it is hardly ever possible to obtain experimental information on all the numbers that come out of a physical model. For example, while it is possible to obtain experimental information on all types of defects created by irradiation that involve missing atoms (i.e. vacancies) in the whole range of sizes, from very small clusters (using positron annihilation spectroscopy) to large cavities (using electron microscopy or small-angle neutron scattering), the same cannot be said for interstitial-type defects, which are only visible to the electron microscope but invisible below a certain size. This is why more fundamental, invalidable models based on solid physical theories (e.g. quantum mechanics or approximations of it in electronic structure, or *ab initio*, calculations) are useful and necessary, as otherwise there would be no possibility of getting any kind of information on those invisible processes and relevant numbers. This is also why some calibration is generally unavoidable: not everything can be calculated, even though efforts are made, using more fundamental modelling tools, to reduce the need for calibration to the very minimum (and it should not be 'fitting', and so on). Once calibrated, models of this second type can also be used to attempt predictions within a given range of validity that, since they are physical models, is generally fairly well defined. Eventually, these models can indeed be used to explore a spectrum of conditions that experimentally could not be explored entirely,

providing hints for the interpretation and rationalisation of experimental results, but also anticipating experimental results for yet unexplored conditions. In this sense, models, if appropriate, will help better focus new experiments, both modelling-oriented and technology-oriented. It is in this sense that they achieve some level of predictive capability. The overall relationship between physical models and experiments is schematically illustrated in Fig. 2.1.

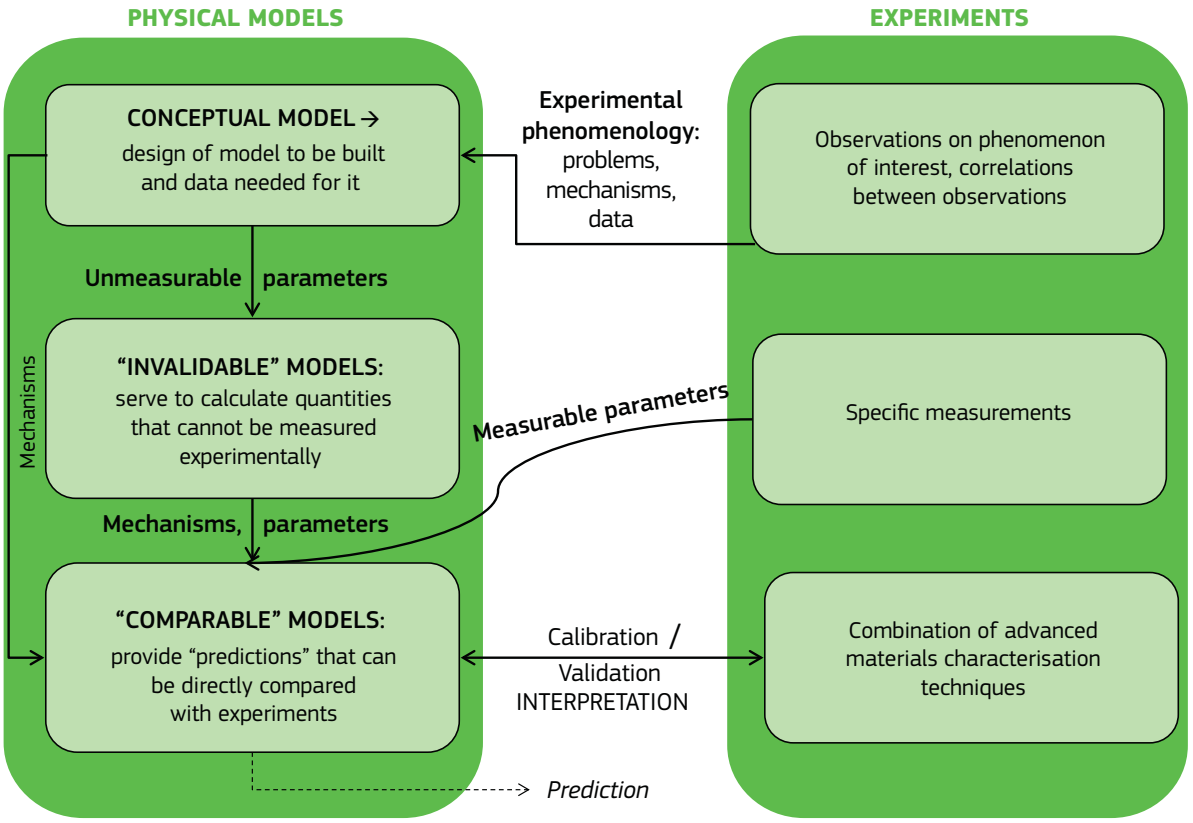

Figure 2.1: Schematic description of the interrelation between experiments and modelling when building a physical model that has the ambition of being predictive

It should be emphasised that, differently from the commonplace misconception, the goal of scientists who develop multi-scale materials models is *not* to 'chain codes to predict materials behaviour replacing experiments'. Instead, the main goal is to understand the physical mechanisms that lead to the effects observed experimentally, a desire that is also intimately connected with the human hunger for knowledge and the fascination of science. Ultimately, it is not impossible that good computer scientists can cleverly combine different established models to provide a chain of codes that is capable of giving answers about materials behaviour under given conditions. However, this should not be regarded as the main, and certainly not the only, goal of the development of multi-scale models. More generally, one should consider that the multi-scale modelling approach includes many tools and is altogether one more tool at our disposal with a view to understanding reality and trying to predict its evolutionary trends. This is illustrated graphically in Fig. 2.2. Figure 2.3 compares a rigid interpretation of the multi-scale modelling approach, in practice disconnected from reality, with the interconnection between tools that is often actually applied.

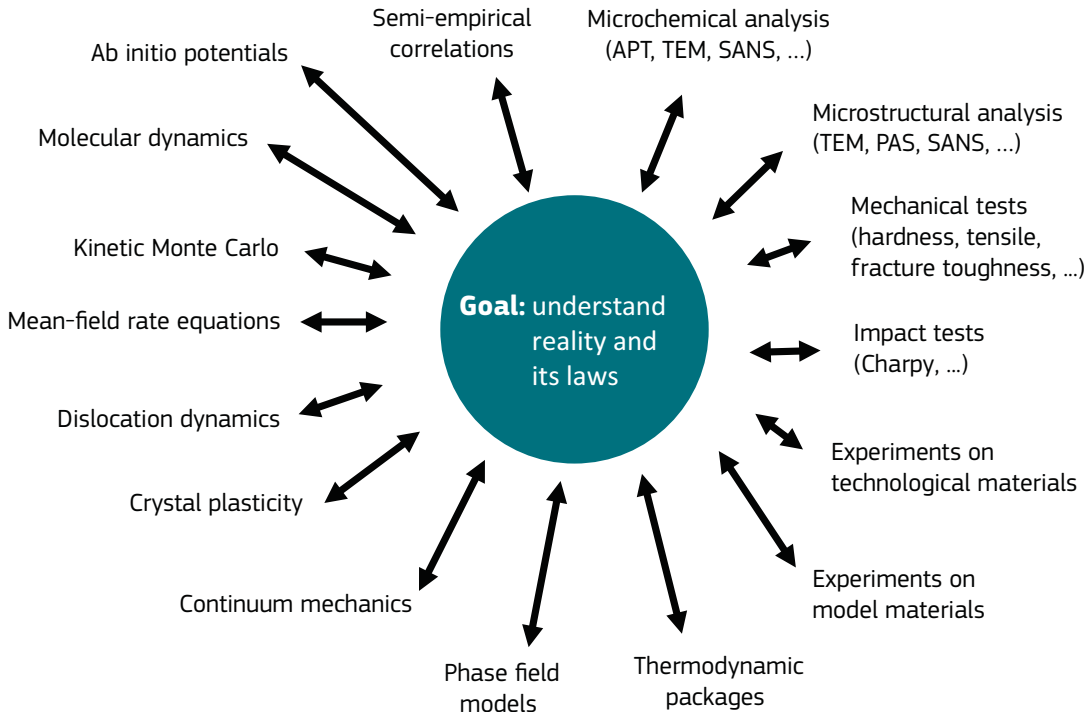

Figure 2.2: Multi-scale modelling tools, experimental techniques and other tools are not rigidly organised in a hierarchy — instead they can all be combined to increase our level of understanding and capability of predicting the behaviour of materials

2.2. Modelling and technology

The connection of multi-scale modelling with technology might appear for some a bit loose, but it definitely exists. First of all, the phenomena that are investigated always correspond to technological concerns in the case of which purely empirical approaches have not led to fully satisfactory solutions, or for which there is a strong belief that a deeper understanding of the physics will lead to an improved mastering or exploitation of the phenomenon at hand (a good example is semiconductor doping for the electronic industry). Secondly, the insight gained from a physical understanding of the phenomena of interest, based on the combination of models and modelling-oriented experiments, will suggest ways of proceeding at the technological level to mitigate the problem or to better exploit the physical process of interest. The connection between physical models and technology is schematically illustrated in Fig. 2.4. These models, as illustrated in Fig. 2.2, should not be separated from the numerous other tools in the hands of scientists and engineers (including the human brain capability of making conclusions and decisions out of limited amounts of data and evidence) for the identification of new technological processes or mitigation solutions. In some cases they can also give indications for the development of semi-empirical correlations that, by relying on a mechanistic approach, will eventually prove to be more reliable.

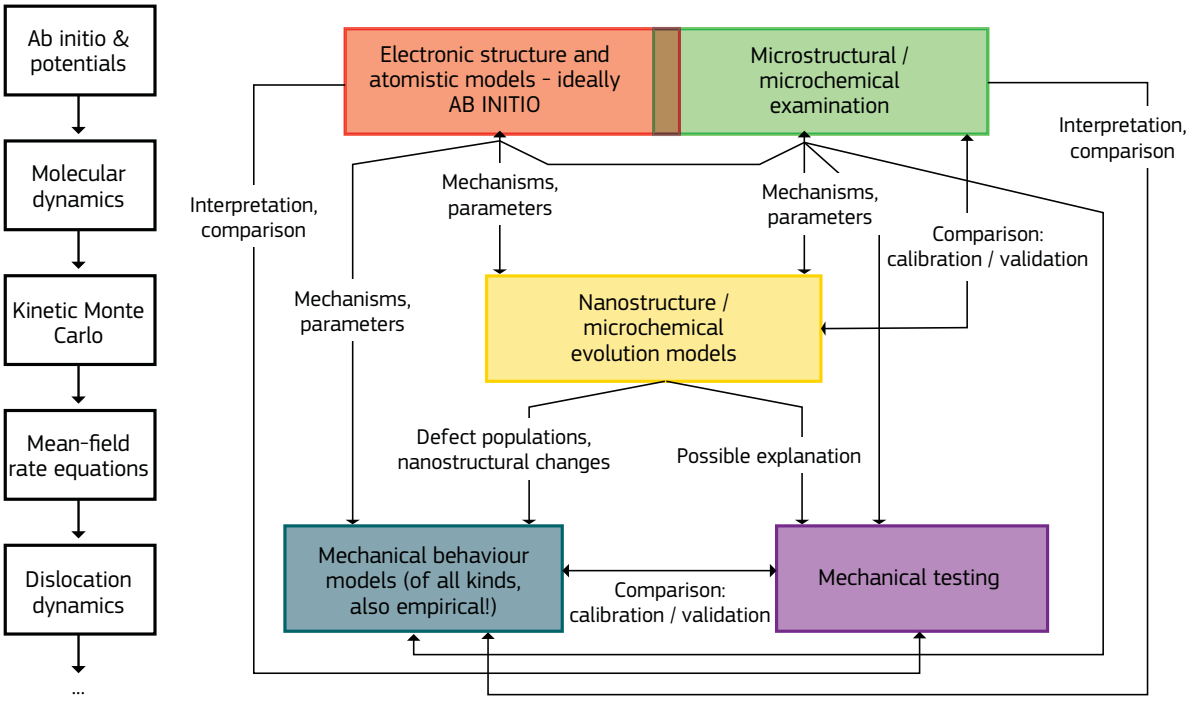

Figure 2.3: Left: rigid and idealistic schematisation of a multi-scale modelling approach; Right: multiple connections between multi-scale modelling tools, as used in reality

A first example of the last point, of relevance for nuclear energy, is the development of trend curves that describe the embrittlement (increase of the ductile-brittle transition temperature) of reactor pressure vessel (RPV) steels as a function of neutron dose (and many other variables, especially temperature, flux and chemical composition). Pressure vessels are irreplaceable components of nuclear power plants (NPPs), the lifetime of which essentially decides the lifetime of the whole reactor unit. The development of physical models (especially atomistic models) of increasing complexity, combined with advanced experimental examination techniques (especially atom probe), led to the identification of the fact that not only copper precipitation and phosphorus segregation were responsible for the embrittlement, but also nickel, manganese and silicon, thereby pushing to the elaboration of new semi-empirical correlations of mechanistic derivation [15, 16]. These physical models suggested — and further progress is being made in this respect — that the mechanisms involved are much more complex than initially thought and led to predictions of facts that might have been, and indeed have been, observed at high enough neutron dose (fluence), namely the formation of manganese-nickel–rich clusters. As a matter of fact, right now one of the concerns for utilities that intend to extend the lifetime of existing NPPs is the lack of high fluence data for pressure vessel steels and the uncertainty in their interpretation, because of two important questions in the background: (1) Can we rely on embrittlement data coming from materials test reactors where the neutron fluence reached is higher but also the flux is much higher than in power reactors?, (2) Is it true that, above a certain incubation dose, a sudden increase in the embrittlement will appear, related with the appearance inside the material of what are called 'late blooming phases'? Both these questions are difficult to answer experimentally because of the cost of experiments (in terms of resources but also time, if low flux/high fluence data are to be collected) and the large spread that is encountered in the relatively limited amount of high fluence data obtained at both low and high flux. However, recent advances in multi-scale modelling, mostly achieved in the framework of the Seventh Framework Programme (FP7) PERFORM 60 project, led to a high level of understanding of the mechanisms of formation of the so-called

late blooming phases and, based on this, to the development of physical models that can, up to a certain extent, under appropriate approximations, describe the nanostructural evolution under irradiation of iron alloys that model RPV steels [17, 18]. These models, although they still require elaboration and improvements, will allow studies of the effect of flux to be systematically performed. These models suggest that what have been named late blooming phases might actually be a misleading concept. By understanding their mechanisms of formation, which appears to be intimately related with the production of radiation defect clusters, one can preliminarily deduce that these features do not appear 'late' as they start in fact forming very early (even though experimentally they cannot be detected: but atomic-scale models can 'see' them even when experimental examination cannot), and that they follow a continuous evolution, such that no sudden 'blooming' will probably ever occur [19]. Their effect on embrittlement will probably be steadily growing according to a law that is close to being deduced from physical considerations, which most probably does not lead to any catastrophic effect (thereby appeasing the fears of utilities, and so on). These models will help to focus future expensive experiments, as well as the analysis of the data, on the investigation of more precise and relevant questions than has been done so far.

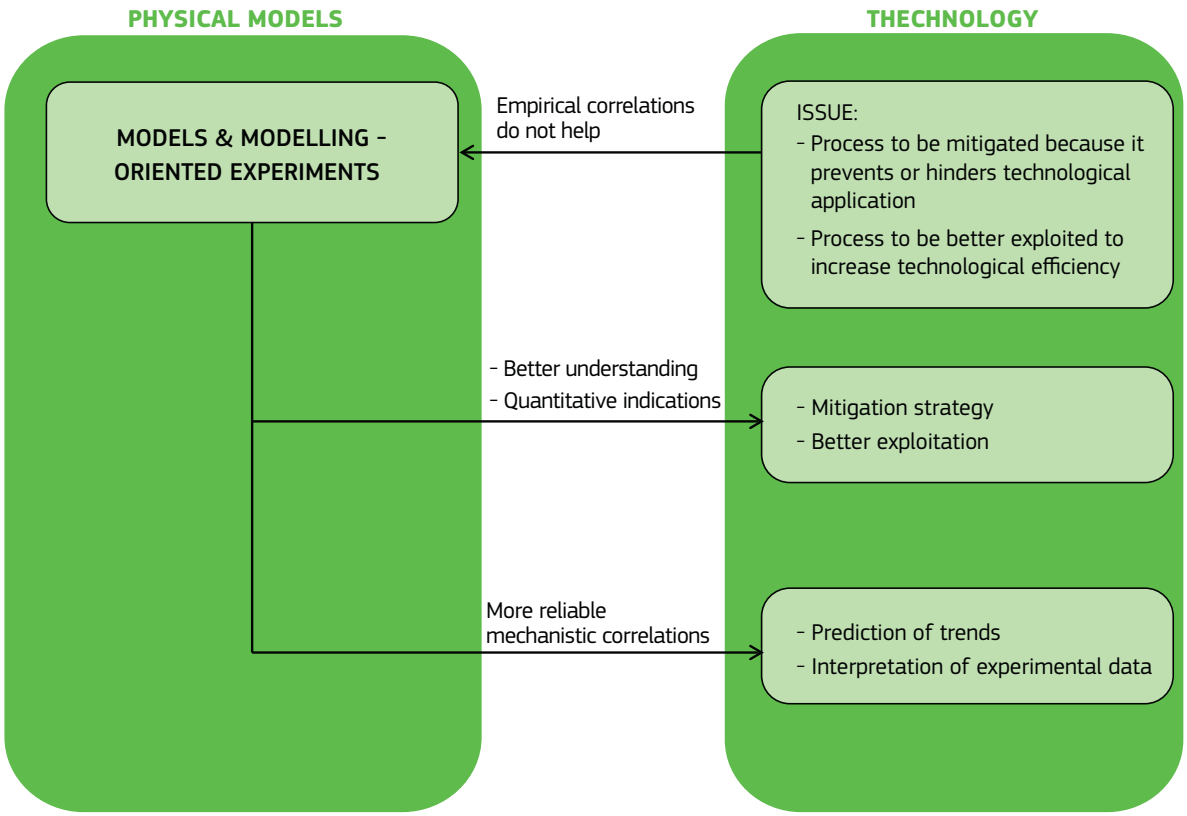

Figure 2.4: Schematic description of the interrelation between technology and physical modelling

A second example concerns nuclear fuels. The evolution of fission gases in fuels is closely coupled to their performance and limits the maximal burn-up they can be submitted to. These fission gases, because they are rare gases chemically inert and particularly insoluble, tend either to form small nanometre-size clusters by interaction with vacancies or to be released from the fuel. This induces:

- over-pressurisation of the rod, which increases the pressure on the clad walls;
- extensive fuel swelling, which leads to mechanical interaction with the clad and increases the probability for clad breach [20].

Retained fission gas bubbles also decrease the thermal conductivity of the fuel and consequently contribute to limiting the operating temperature and the degree of burn-up. Understanding the elementary mechanisms of the solubility of gas in fuel lattice and in bubble is paramount to predict the quantities of gas staying in the fuel and those released [21] and will help in developing solutions to mitigate the consequences of the formation and evolution of these large quantities of gas.

2.3. Elaboration of a multi-scale materials modelling plan

When elaborating a plan for the development of (multi-scale) physical models to investigate a given phenomenon, it is extremely important to keep in mind the feasibility of the plan, based on the knowledge of the state of the art, as well as, as much as possible, the expected timeframe. Modern modelling tools are not all-powerful, irrespective of the power of available computers. One could fantasise over *ab initio* simulations of the interfaces between a steel and a flowing liquid metal to investigate liquid-metal embrittlement, or between irradiated fuel and its cladding leading to mechanical-chemical interactions between them, but such plans, although *in principle* possible with a science-fictionally powerful computer, are unfeasible in practice. More to the point, it is not by attempting the simulation of a very complex phenomenon, including all the complexity of the system, that one understands and quantifies the underlying physical mechanisms. A modelling plan should address problems that are sufficiently well-understood, based on experimental data, so that a reasonable conceptual model exists already that can be used to actually build a quantitative model. In this case, the physical model that includes contributions from invalidable calculation techniques can actually provide an added value to cast some light on still unclear issues. Often, the problem is so complicated that empirical correlations are the only way to find out which variables play a role and the determination of these variables corresponds to the very first step to try to rationalise experimental observations: without this knowledge, no further step can be taken and it would be delusive to expect that all aspects of the problem can be elucidated by trying to simulate it in the computer. The development of physical models and the application of computer simulation should be undertaken with precise questions in mind that it seems possible to answer with the available tools. Often, atomistic models allow different possible mechanisms to be explored and might sometimes reveal unexpected mechanisms. However, in general one should never assume that everything can be obtained *ab initio*, even though *ab initio* calculations are very powerful and by now essential tools in our hands; nor should one think that, by simply reproducing certain conditions in the computer simulations, answers to questions will come out spontaneously. The use of the brain to 'Cartesianly' split the problem into parts that are simpler to address is not an obsolete practice: it remains key when designing a modelling plan. Consistently with what has been said above, the plan should also always include feasible experiments that involve the use of a combination of characterisation techniques so that physical mechanisms can be deduced also from the experimental work: the final model will have to include these mechanisms one way or another and the data will be usable for the calibration of the model (even though the objective is not that the model should be *fitted*, but rather that the *calibration* should occur on parameters physically well-defined, though not fully known either from experiments or calculations). The tools to be used to develop the model should be identified depending on the scale and one should be able to select the most suitable one out of a, by now, fairly wide choice: sometimes the difference between tools lies only in very technical and subtle details, but they can be important. Sometimes it turns out that the development of new modelling tools is necessary. Developing tools that allow more 'realistic' simulations to be performed and the complexity of real systems to be better caught is certainly stimulating because of the challenge it poses and is part of the path to be followed towards steady progress. However, experience shows that the development of new tools or approaches is time consuming, it is never as easy as one might think at the beginning, it always requires proceeding step-by-step and it often leads to the need to solve numerical or mathematical problems that will probably look to the layman as totally academic and of hardly

any use, but might require years to be solved. Thus, in the case of an especially complex phenomenon, one should not rely too much on the development of revolutionary modelling tools, nor attempt a 360 simulation. Instead, key questions amenable to modelling with existing or foreseeable tools should be identified. It is of course possible to think of an ambitious plan that relies on new developments at all scales, but in the short-to-medium term the investigation should rely mainly on existing modelling tools that only need to be adapted, following an incremental approach.

3. Multi-scale modelling activities in EERA's Joint Programme Nuclear Materials (JPNM)

From what has been said above, it is clear that the development of multi-scale models, which rely enormously not only on computer simulation but also on refined experiments, especially in the nuclear materials field, is at the interface between fundamental science, applied science and technology. It is a sort of torch that casts lights here and there and helps decide where to search further in the attempt to take our bearings in the dark room that is often the behaviour of materials. It provides a set of tools that enable the assessment of quantities that cannot be experimentally measured, or the exploration of conditions experimentally problematic to reproduce and that, in combination with appropriate experiments, can determine a significant leap forward in our understanding. It is a necessary activity in view of the optimisation of materials for nuclear reactors, of a better mastering of nuclear reactor safety issues that unavoidably involve materials, and in general to provide the scientific background that is always needed in order for innovation to emerge from a sea of fundamental research. Moreover, because of the extreme complexity of the processes that occur under irradiation, multi-scale models developed for nuclear materials offer as spin-offs valuable contributions to the development of similar models for simpler processes, for example, thermal ageing, which is an issue common to all high-temperature energy technologies. It is, however, an activity that, at the moment at least, cannot enjoy direct support from industry (besides in part by very large utilities that can afford large research, development and innovation departments) and that unavoidably competes with other, more technology-oriented (even though generally more costly), applied research activities, not only in connection with existing reactors but even more with future (Generation IV (Gen IV) and fusion) systems. In this respect, it does need the support of public money (both at national and European levels) to keep flourishing and to maintain the level of advancement and maturity reached in the last decade in Europe, right now superior to other 'nuclear continents' such as the Japan and the United States, or emerging Asian countries (China, India, etc.). In particular, it is widely recognised that in order to move to systems that, like Gen IV and fusion reactors, will subject materials to extreme conditions, thereby requiring that the properties of materials are progressively improved, a strong basis of physical and mechanistic understanding is mandatory. For this reason, in the framework of the Joint Programme Nuclear Materials (JPNM) of the European Energy Research Alliance (EERA), two sub-programmes (SPs) exist that are devoted to the development of multi-scale physical models for structural materials (SP4) and fuel (SP6).

3.1. Multi-scale modelling applied to nuclear structural materials

The multi-scale modelling activities in the SP4 (modelling of structural nuclear materials) of the JPNM are organised in work packages (WPs) that match the other SPs devoted to structural materials. Namely, WP1 is devoted to modelling alloys that correspond to commercially available nuclear steels — i.e. on the one hand ferritic/martensitic (F/M) steels and, on the other, austenitic steels, mirroring the SP1. WP2 is meant to model oxide-dispersion strengthened (ODS) F/M alloys (the subject of SP2), and WP3 was foreseen to address the modelling of ceramic composites and refractory alloys (the subject of SP3). For what concerns WP2 and

WP3, task forces are currently busy with the elaboration of a JPNM coordinated modelling plan. There are a number of European research centres interested in modelling ODS alloys and already progressing in that direction, but in a still not fully coordinated way. The activity on ceramic composites and refractory alloys, on the other hand, is actually currently very limited. On the contrary, WP1 includes a large amount of ongoing coordinated activities in Europe, within defined pilot projects, many of which have been or are part of the work performed in FP7 projects and, hopefully, to be performed in Horizon 2020.

3.1.1. Issues addressed in WP1 — F/M and austenitic steels

The physical modelling activities in WP1 of the SP4 of the JPNM address mainly the following two issues, having in mind mostly ferritic and partly also austenitic iron alloys:

(1) nanostructure and microchemical evolution under irradiation and thermal ageing;
(2) radiation-induced hardening and embrittlement.

In addition, some activities start to try to address the following issues:

(3) radiation-induced embrittlement without hardening;
(4) loss of elongation due to plastic flow localisation;
(5) thermal creep and creep-fatigue;
(6) swelling and irradiation creep;
(7) liquid-metal corrosion and embrittlement.

There are several reasons why the most established activities concern nanostructural and microchemical evolution under irradiation and subsequent hardening (which in turn causes embrittlement).

- These issues correspond to well-posed problems for the available modelling tools: the scales are adequate for atomic-level and nano-scale tools, the responsible physical mechanisms are qualitatively well known (displacement cascades, defect and solute diffusion, dislocation-defect interaction, etc.) and it is therefore feasible to devise a *sequence* of tools that altogether can potentially provide a complete model that spans several scales (ab initio → molecular dynamics → kinetic Monte Carlo and/or rate theory → dislocation dynamics → crystal plasticity, and so on).
- The problem of describing the nanostructural and microchemical evolution is key almost irrespective of the radiation effect addressed, because these changes are always at their origin; thus, it is quite obvious that large efforts in this direction have been made using atomic-level and nano-scale simulation tools.
- Since hardening and embrittlement is a problem that affects RPV steels of current NPPs, it has been addressed already for a long time and by many laboratories worldwide, so the incremental advances that have been made brought the community very close to obtaining very relevant results and an increasingly large amount of researchers have been willing to contribute to this effort. The fact that the problem remains of relevance for Gen IV and fusion reactors obviously causes the community to be even more prone to putting efforts on these issues.

Radiation hardening and embrittlement

In FP7 projects such as PERFORM 60 [22] and GETMAT [23], significant advances have been made towards the development of models at different scales that manage to explain the processes leading to radiation hardening and subsequent embrittlement. Tools and models have been developed that allow one to be ever-more quantitative and possibly eventually predictive in some respects. Wide knowledge has been accumulated concerning the effect of the most important alloying element in iron on the nanostructural evolution under irradiation, both interstitial (C and N) and substitutional (Cu, Ni, Mn, Si, P, Cr, etc.), thanks both to extensive

ab initio calculations [24] and to the development of multi-component interatomic potentials [18]. The latter rely on the solid methodology that has been established for their development, based both on *ab initio* calculations and thermodynamic information from phase diagrams, now explicitly accounted for in the fitting of the potentials [25]. Atomistic kinetic Monte Carlo techniques have been pushed to high levels of physical and chemical complexity and computational efficiency: they can be used to simulate the first stages of irradiation processes in complex alloys (almost all important alloying elements are included) [26], as well as the annealing (e.g. in resistivity recovery experiments) [27], and processes such as radiation-induced segregation [28], or to study systematically the mobility of vacancy clusters in alloys, including the possibility of forming complexes with solute atoms [29]. All this information can be used to refine nanostructure evolution models. In particular, a reasonable kinetic Monte Carlo model has been developed that reliably describes, as compared to experiments, the build-up of populations of defects (vacancy and SIA clusters, evolving to voids and dislocation loops) in iron-carbon (Fe–C) alloys, in a range of temperatures from room to ~ 300 °C, *including post-irradiation annealing experiments* [30, 31]. From this basis, first approximation models for more complex alloys have been calibrated [32]. Effective methods have been developed to transfer information about dislocation/defect interactions simulated atomistically with molecular dynamics to mesoscopic dislocation dynamics models [33], thereby being able to reproduce the irradiation hardening measured in experiments, based on physical grounds [34]. Dislocation dynamics-based constitutive equations for crystal plasticity simulation have been elaborated [35]. In this way, a true connection between scales is now in place. All these advances, although not yet enough to make multi-scale modelling tools of direct use for industry or for reactor design, are very encouraging.

The still open issues, on which effort needs to be focused now, are:

- the development of models capable of describing *at the same time*, and up to sufficiently high doses/temperatures, the nanostructural and the microchemical evolution under irradiation, especially in concentrated alloys, including at least all the most important alloying elements found in nuclear steels in connection with radiation damage effects and ageing in general;
- the further development of methods to describe, in atomistic kinetic Monte Carlo models, the effect of the presence of defects that produce large strain fields (dislocation lines and loops, grain boundaries, etc.), as well as the influence of vibrational and magnetic entropy on migration barriers at high temperature;
- the consolidation of the link between molecular dynamics and dislocation dynamics, on the one hand, and of the latter with crystal plasticity, on the other, in order to be able to obtain from physical models the stress–strain curve of irradiated steels;
- the identification of physics-grounded effective methods to obtain the fracture toughness of metals, beyond the current semi-empirical approaches, probably starting from a re-visitation of the local approach.

Of all, the last issue is the most challenging one, in view of reaching the level of maturity that is necessary to meet the expectations from the multi-scale modelling approach in connection with technological applications. It is also of paramount importance that adequate and wide-range modelling-oriented irradiation and post-irradiation examination experimental programmes on model alloys and steels are set up and performed, to provide the experimental insight required to advance further in the model development and, eventually, validation, along the lines already discussed in Section 2. This will in fact necessarily absorb a significant amount of resources.

Concerning the other issues addressed in WP1, points (3) to (7) above, they all correspond to complex phenomena, on which the level of understanding of the fundamental mechanisms is not as high as for nanostructural and microchemical evolution, on the one side, and hardening and embrittlement, on the

other. They are briefly described in what follows. These descriptions show that on each of them further work is needed to progress towards models that fully connect with the technological applications.

Radiation-induced embrittlement without hardening

Radiation-induced embrittlement without hardening is related with segregation of chemical elements at grain boundaries capable of promoting intergranular fracture [36]. A special version of this phenomenon corresponds to helium embrittlement, which is expected especially under fusion spectra at high temperature and is also connected with void swelling [37, 38]. Most physical studies focus on the evaluation, with atomistic calculation techniques, of the change in the grain boundary energy due to the segregation of specific elements [39, 40] according to different mechanisms and under different forms, including He bubbles [41]. However, to be useful in connection with structural integrity applications, this knowledge should in principle be transformed into information introduced in crystal plasticity models, such that the occurrence of intergranular versus transgranular fracture is predicted. This remains a far goal.

Plastic flow localisation

Loss of elongation due to plastic flow localisation within clear bands (dislocation channels) is a phenomenon observed in all irradiated metals, above a certain dose, that tends to be fairly low for F/M steels (less than 1 dpa), though it can be relatively high in austenitic steels (up to 10 dpa) [42]. When plastic localisation happens, the metal, after yield, deforms plastically under lower engineering stress than the yield strength (softening), with consequent drastic reduction of elongation. The formation of dislocation channels, especially their crossing and convergence at grain boundaries, is also suspected to promote intergranular fracture and to make metals more susceptible to stress corrosion cracking [43]. While qualitative and partially also quantitative understanding of dislocation channel formation exists [44, 45], the detailed mechanisms are still elusive. For example, it is unclear whether grain boundaries are the regions from where clear banding starts, or the region where clear banding ends or through which clear bands propagate to other grains. The possibility of triggering this effect at crystal plasticity-level is a goal not yet reached and plasticity models generally assume that the soft channel has already appeared, thereby limiting the actual representativeness of continuum mechanical models that include this type of ingredient. Work on these subjects is foreseen in the DOMOPLEX pilot project of the JPNM, which, however, is currently lacking appropriate funding.

Thermal creep and creep-fatigue

Thermal creep and creep-fatigue are amongst the main factors that limit the lifetime of components, in nuclear reactors and not only [46]. Thermal creep limits the possibility of increasing the operational temperature so as to boost the energy efficiency of NPPs, especially Gen IV concepts. Creep-fatigue has often appeared in nuclear reactors as an unexpected limiting factor that obliged expensive components or parts of them to be replaced. The physical mechanisms of thermal creep at the atomic level are understood in terms of dislocation climb, but the translation of this mechanism into creep deformation constitutive laws of application in continuum mechanics is still lacking. The combination of creep and fatigue is still addressed in a very empirical fashion at design code-level, but the complexity of the phenomenon, that might involve grain coarsening during cycling load, is such that no full understanding exists.

Swelling and irradiation creep

Swelling and irradiation creep are interrelated phenomena very much connected with the nanostructural evolution of irradiated materials [47, 48]. Attempts at modelling swelling date back to the early 1970s, when the phenomenon was identified. Existing models are mainly based on mean-field approximations and allowed some level of understanding to be reached, but in fact no model is capable of describing the incubation phase followed by the steady-state unlimited swelling typical of steels [49]. The mechanisms that trigger the transition from the incubation phase to the steady-state have not been clearly identified,

yet. One serious problem in connection with swelling for nanostructure evolution models informed by atomistic mechanisms is that swelling occurs at high doses and temperatures, both currently out of reach for, for example, kinetic Monte Carlo models, because of computational limitations. Mean-field rate theory models remain, therefore, in practice the only usable approach, but they are in turn limited in the possibility of including detailed physical mechanisms, besides sometimes running into numerical problems to solve the equations. Irradiation creep mechanisms have been proposed in large amount already 20–30 years ago [50], but it is still not clear which ones actually operate and are dominant in a given material. It is believed that a re-visitation using modern atomistic techniques of these mechanisms might help cast some light on the problem and this type of work is about to start in the FP7 MatISSE project.

Liquid-metal corrosion and embrittlement

Finally, liquid-metal corrosion and embrittlement are extremely complex phenomena on which no full consensus exists as to physical mechanisms and factors that determine susceptibility to them [51]. These are processes that occur or not depending on the solid metal and liquid metal involved. For example, liquid metal embrittlement is not detected in F/M steels in contact with liquid sodium, but it is potentially a problem in F/M steels in contact with liquid lead, or lead-bismuth eutectic. On the other hand, austenitic steels are not affected by liquid metal embrittlement even when in contact with heavy liquid metals. The existence of this phenomenon may prevent the use of F/M steels in a lead fast reactor (LFR) and oblige the choice of austenitic steels, even though these are penalised by higher and earlier swelling. A full understanding of the mechanisms at the origin of liquid metal embrittlement might help mitigate this problem, but experimental investigations aimed at understanding the physical processes leading to this manifestation are limited and have started only recently to be performed somewhat systematically, having in mind nuclear applications, within the FP7 GETMAT project. Work is expected to continue in MatISSE. In the absence of a clear phenomenological understanding, it is difficult at the moment to identify the modelling path to be followed to provide an added value, besides estimating wetting and interface energies between specific liquid metals and solid metal substrates, trying to predict the phases involved and their kinetics of formation.

3.1.2. Issues addressed in WP2 — ODS steels — and WP3 — Ceramic composites

Concerning WP2, the plan that is being elaborated by the task force can be expected to be devoted to the following issues:

- Clarify processes that occur during the fabrication of the ODS alloys, since complete control over the final properties of the alloy depends enormously on how the fabrication process is conducted and mastered.
- Devote attention to the mechanisms whereby ODS alloys deform and fracture: given the hardness of the ODS matrix, processes such as grain boundary sliding might be expected; the role of temperature is especially crucial, as these alloys are supposed to be operating up to very high temperature (700–800 °*C), but* exhibit low fracture toughness even in that range.
- Investigate the stability of the particles, their role as absorbers of point-defects and He atoms, the changes that occur in the oxide/matrix interface while defects and He are absorbed, and the possible impact of all this on mechanical properties.

As to WP3, it is hoped that a limited activity devoted to the mechanisms of deformation in composite ceramics (essentially SiC fibres in SiC matrix (SiC_f/SiC)) will be set up.

3.2. Multi-scale modelling applied to nuclear fuel materials

The research SP6 on the multi-scale modelling and separate effect experiments of nuclear fuel proposed for the JPNM concerns the knowledge improvement of the fundamental properties of the advanced fuel materials and basic processes that govern the mechanical, physical and chemical behaviour, aiming at the multi-scale (time and length) modelling of nuclear fuel behaviour. European research in this field is already well developed and is at the forefront of worldwide developments, in particular thanks to the work performed in the FP7 F-BRIDGE project [52].

In-depth knowledge of the behaviour of nuclear fuels at high temperatures — i.e. under accidental and normal operating conditions — is essential for the reactor safety assessment and the assessment of potential new fuel forms, such as those that aim to incorporate minor actinides (MAs) for transmutation.

The SP aims at extending the basic research approach developed in particular in the F-BRIDGE project to more complex systems, properties or conditions, in particular those of interest for the development and safety analysis of fuels for Gen IV reactors. It will therefore contribute to solving a selection of critical applied issues relating to nuclear fuel behaviour under irradiation under normal and off-normal operating conditions, getting complementary visions of the essential phenomena and improving our fuel behaviour modelling capabilities.

Melting temperature, fission product chemistry and release, microstructure evolution under irradiation and mechanical integrity are the key issues that have been identified from a first top-down analysis done in interaction with industry representatives involved in the user group of the F-BRIDGE project.

The SP aims at performing cross-cutting activities that will cover various types of future and sustainable reactors fuels, aligned with the European Sustainable Nuclear Industrial Initiative (ESNII) goals, and concentrating on sodium-cooled fast reactor, LFR, gas-cooled fast reactor and accelerator-driven system reactor types, namely UO_2 as the reference, and mixed oxides (MOX): (U,Pu) O_2,(U, Am)O_2, etc., but also carbides and nitrides.

It will achieve its goals thanks to the strong interaction between modelling and experimental tasks carried out at relevant scales and through complementary studies relating to in-pile irradiated fuels as well as separate effects experiments on non-active materials for which irradiation and fission products are simulated with ion beam irradiations, implantations and a-emitting dopants.

The SP is therefore organised in four WPs, one dedicated to data inventory and gap assessment and three research WPs each focusing on one of the key issues cited above.

3.2.1. WP1: Data inventory and gap assessment

This WP will examine in detail the achievements made in the basic research investigation of nuclear fuels in previous European initiatives or independently and the knowledge acquired, and perform a gap analysis to determine the needs to be fulfilled in future European programmes.

3.2.2. WP2: Assess and increase margins to fuel melting

Margins to melting depend strongly on the properties of the fuel material so understanding material properties at various time and space scales is essential. In this way, the operational limits of a fuel can

be determined on a scientific basis, rather than on an empirical basis, as is currently the case. Moreover, research performed in the framework of the F-BRIDGE project has shown that not all properties are known with sufficient accuracy, as a new estimate of the melting temperature for pure PuO_2, almost 300 K higher than the previous reference value, clearly demonstrated [53].

The scientific studies in WP2 will focus on three main topics: (1) melting temperature determination of MOX phases that are foreseen as fuels or as MA-bearing blankets for nuclear reactors; (2) thermal conductivity measurements of these phases, that will be performed, also looking closely at the compositional variations of the samples; (3) high-temperature thermochemistry of these materials, that will be studied using mass spectrometry to analyse the solid-liquid-gas equilibria of these oxide systems. For all these studies, new samples will be manufactured that enable measurements. The experimental work will be closely coupled to modelling studies. The main modelling tool will be CALPHAD that deals with the optimisation of the phase diagrams based on equilibrium data from the new experiments and literature data from the past. In the F-BRIDGE project databases for the U-Pu-O-C and U-Pu-Np-Am-O systems have been developed [54]. The latter will be improved and extended by the proposed research, resulting in more accurate prediction of melting temperature and vaporisation behaviour of MOX fuels containing MAs, as well as associated uncertainties.

In addition, the fuel swelling induced by irradiation can lead to chemical and mechanical interaction with the cladding. Possible chemical reactions need to be verified in carefully designed diffusion couple tests. Zirconium and iron will be added to the database to also allow the accurate prediction of eutectic formation between fuel and cladding during pellet–cladding interaction.

In the event of a rupture of the pin, fuel can also interact with coolant. The thermophysical properties of fuel coolant compounds will be investigated to provide information on their role in exacerbating or, on the contrary, limiting the effect of such irregular incursions of coolant into a fuel pin. Sodium, lead and helium are the main coolants of interest.

The knowledge generated in WP2 will be used in other WPs, particularly in WP3, in which a coupling with the fission product chemistry will be made.

3.2.3. WP3: Improve control of fission product release and reduce impact

During in-reactor irradiation, actinide fission generates large quantities of fission products, volatile and non-volatile, which have a significant influence on the chemical composition, as well as on the structural and mechanical properties of nuclear fuels. In parallel, nuclear interactions with neutrons and fission products induce atomic displacements and disturb the crystalline structure, electronic excitations disturb the chemical bonds, and transport phenomena assisted by temperature and irradiation modify the microstructure. The interaction between fission products and the various defects created govern a large number of operational properties and processes: cracking, gas bubble nucleation, radial migration or corrosive fission product availability at the fuel–cladding interface.

WP3 will focus on investigating fission products properties and their impact upon nuclear fuel behaviour. Three areas of research have been identified following the work done during the F-BRIDGE project:

- The transport properties of defects, which influence many basic material properties such as oxidation, actinide or fission product redistribution, fuel swelling and creep [23]. These defects interact significantly with fission products. The studies performed in the F-BRIDGE project have brought an improved atomic-scale description of UO_2 and its defects using electronic structure calculations

and empirical potentials [55, 56, 57]. The combined electronic structure, classical molecular dynamics and separate effects experimental studies of O self-diffusion has yielded information on the elementary mechanisms involved in this migration [58, 59]. The recombination of defects has also been investigated [60]. To get further insight into the properties of defects, experiments and dynamical modelling investigations on the uranium sub-lattice are needed to complement electronic structure investigations [61] and bring the same insight as for O migration. Larger vacancy clusters, which act as traps for fission gases, as well as interstitial clusters, with various charged states must also be investigated in detail. Experiments for more controlled stoichiometries must then be performed and of course the methods developed or validated in F-BRIDGE must be applied to the investigation of defects and non-stoichiometries in MOX.

- The behaviour of fission gases, which, since they are chemically inert and particularly insoluble, tend either to form small nanometre-size clusters by interaction with vacancies or to be released from the fuel. This induces over-pressurisation of the rod or extensive fuel swelling. Bubbles are liable to grow and become in turn trapping sites for migrating defects or other insoluble atoms. As mentioned in Sub-section 2.2, further insight into the elementary mechanisms of the solubility of gas in fuel lattice and in bubble is paramount to predict the quantities of gas staying in the fuel and the quantities released.

In and outside F-BRIDGE progress was made on the mechanisms of atomic migration [62, 63, 64] and early stage of bubble formation [65, 66, 67, 68] using electronic structure methods, empirical potential calculations and experimental characterisation at the nano scale after ion implantation. It has also been shown that atomic-scale studies yield relevant results on the elementary migration mechanisms of gases that can be put in meso-scale models [69]. Then, the combination of cluster dynamics modelling and desorption experiments yielded information on the migrating species and on the mechanisms of gas release [70]. The multi-scale approach must be developed further by extending the meso-scale models and complementing the calculations of the input parameters of these models at the atomic scale. More controlled experiments with detailed characterisation must also be performed to obtain improved basic data on gas behaviour.

- The behaviour of non-gaseous fission products and their compounds, which governs fuel chemistry under irradiation under normal and accidental operating conditions. The study of transport properties of iodine in UO_2 was started during F-BRIDGE [71, 72, 73]. It should be continued and extended to other non-gaseous fission products, such as Cs, Te, Mo, Ru, Rh, Sr or Pd, and to various fuel compositions particularly relevant to Gen IV. The thermodynamics of the numerous compounds formed between various fission products and the original fuel constituents should also be determined.

The materials studied will include, in addition to non-stoichiometric pure and doped uranium dioxide and carbide, plutonium dioxide and MOX fuels, as well as fuels containing americium and surrogates. The investigations will consist of the combination of innovative characterisation techniques and multi-scale modelling from the atomic to the mesoscopic scale, such as those described in Sub-section 2.1 and Fig. 2.2. Results will be transferred to fuel performance codes, as initiated in F-BRIDGE for the TRANSURANUS code [74].

3.2.4. WP4: Obtain data on microstructural changes and mechanical integrity of fuel under irradiation

The microstructural changes and mechanical behaviour of nuclear fuels under irradiation are of course central to predicting the structural integrity of fuel elements. And although 3D thermo-mechanical modelling is now becoming widely available, modelling relies on mechanical behaviour laws that have been obtained from macroscopic testing and integral in-pile experiments [75]. Extrapolation of these laws to materials or irradiation conditions different from those that were the object of the original experiments is

often tenuous. One aim of this WP is therefore to draw on recent experimental and modelling advances to develop further elastic, plastic, creep and damage models.

Research will focus on three main topics. First, the mechanical properties will be investigated since they are critical and most of the basic data on processes at the nanoscopic and microscopic scales are still to be determined. First, modelling studies of dislocations [76] and fracture mechanisms [77] were performed in UO_2 and these must be continued and extended to other fuels, virgin and irradiated. Experimental and modelling methods will be improved or specifically developed to gain further insight into the underlying mechanisms of fuel creep, fracture and pellet–cladding interaction. A specific emphasis will be put on local characterisation techniques on the scale of a few grains, within one grain or at the grain boundaries.

Second, the investigation of elementary mechanisms involved in radiation damage, which has already made progress in the case of UO_2 in the F-BRIDGE project using both modelling [78] and experimental techniques [79], will be continued. In the modelling studies, simulation of displacement cascades will be carried out. This will provide a picture of the primary damage arising from collision sequences, as well as thermal spikes that can to a certain extent simulate the transfer of electronic energy deposited into non-radiative atomic motion. Experimentally, in order to decorrelate effects, irradiation will be simulated using a range of projectile masses and energies comprising variable energy electrons to a-doped samples, for which well-defined damage is created by the decay of an a-emitter homogeneously distributed in the fuel matrix.

Finally, the role of grain boundaries, which are effective sinks for radiation-induced defects and influence transport and mechanical properties, as well as radiation tolerance, will be studied using atomic-scale modelling. First, empirical studies of grain boundary structures in UO_2 [80, as well as of their influence on defect [81] and gas diffusion [82, 83] and on the irradiation damage produced [84] were performed in the F-BRIDGE project. Given the variety and complexity of structures, these studies must continue. The mobility of grain boundaries, which plays a key role in many microstructure transformations, will also be studied.

The contributions in these separate fields will be coupled and integrated to assess their combined effects on fuel behaviour and safety by feeding the results into models in fuel performance codes, thus strongly contributing to improved safety analysis. In addition, the results will be an important input for an assessment of advanced MA fuels designs and will help to optimise the fabrication process.

4. Closing remarks

As a closing remark and recommendation, it is important to stress the following. The multi-scale modelling approach relies on the idea that the proper tool should be used to model a given phenomenon at the appropriate scale and that information has to be transferred from the lower to the upper scale to make the upper scale model more accurate. Conversely, one can start from a large-scale model and try to obtain the missing information from the lower-scale model, in order to increase the accuracy of the large-scale model, or focus the lower-scale studies on some particularly important processes. Whatever the perspective, in this scheme each scale has its own right of existence and is as important as the others. However, it is currently a fact that the majority of researchers involved in multi-scale modelling activities are actually using mainly *atomistic* calculation and simulation techniques. In particular, lots of people perform *ab initio* calculations, and many use classical molecular dynamics for a variety of purposes and studies. *Ab initio* techniques are absolutely essential to inform multi-scale models; however, they should not absorb all the effort. On the opposite side, continuum mechanics is a widespread modelling tool used at all levels, including the industrial level. The *in-between scales*, on the other hand, are much more

'deserted', and even fewer researchers address systematically the problem of transferring information from scale to scale. But stepping to higher scales, even with limited *ab initio* information, with a view to approaching levels where it becomes easier to calibrate/validate models on experiments, is extremely important. Even the development of interatomic potentials for complex systems is addressed by only relatively few experts worldwide. Monte Carlo tools are widely used, but they are mainly atomistic ones. Not so many researchers deal with the development of approximate models, such as object kinetic Monte Carlo, where sometimes very strong and not fully justified approximations must be adopted, the identification of which requires actually the accumulation of a large amount of experimental and atomistic data, to obtain a semi-quantitative picture of what is going on in a specific material or system: these are not models from which mechanisms come out spontaneously, but rather models that imply that the fundamental mechanisms have been understood. Maybe for this reason they are less 'appealing' to the physicist, while being of not-yet immediate use for the engineer. Even more importantly, the expertise in mean-field rate theory models, once upon a time the models 'par excellence' used in radiation damage, is dramatically decreasing, whereas more than ever models of this type, which include information from more fundamental studies, are necessary in order to address the problem of high doses and high temperatures. While many researchers deal with dislocation dynamics in metals, only a handful are busy with addressing the issue of radiation damage and similar considerations can be made for what concerns crystal plasticity. In short, without diminishing the importance of *ab initio* techniques on the one hand and continuum mechanics on the other hand, effort should also be put into developing models that address the scales that are too large to be addressed with atomistic models, but still too small to be able to use fully continuum models.

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References

[1] Malerba, L., 'Multi-scale modelling of irradiation effects in nuclear power plant materials', in *Understanding and mitigating ageing in nuclear power plants: Materials and operational aspects of plant life management (PLiM)*, Tipping, P.G. (Ed.), Woodhead Publishing Ltd., 2010, p. 456 (Chapter 15).

[2] Martin, R.M., *Electronic Structure: Basic Theory and Practical Methods*, Cambridge University Press, Cambridge, 2004.

[3] Hoffelner, W., 'Advanced Mechanical Testing and Analysis Methods', in *Materials for Nuclear Plants*, Springer London, 2013, p. 359 (Chapter 7).

[4] Jenkins, M., Kirk, M., *Characterization of Radiation-Damage by Transmission Electron Microscopy*, Taylor Francis Ltd, London, 2000.

[5] Gault, B., Moody, M.P., Cairney, J.M., Ringer, S.P., 'Atom Probe Microscopy', Springer Series in *Materials Science*, Vol. 160, 2011.

[6] Grillo, I., 'Small-Angle Neutron Scattering and Applications in Soft Condensed Matter', in *Soft matter characterization*, Borsali, R., Pecora, R. (Eds), Springer Verlag, 2008 (Chapter 13).

[7] Hautojärvi, P., Corbel, C., 'Positron spectroscopy of defects in metals and semiconductors', in *Positron Spectroscopy of Solids*, Dupasquier, A., Mills, A.P. (Eds), IOS Press, 1995.

[8] Bunker, G., Introduction to XAFS, Cambridge University Press, Cambridge, 2010.
[9] Schaller, R., Fantozzi, G., Gremaud, R. (Eds), *Mechanical Spectroscopy Q^{-1} 2001 with Applications to Materials Science*, Trans Tech Publications Limited, Switzerland, 2001.
[10] Minov, B., Konstantinovic, M., Dupré, L., ' Development of Magnetic After-Effect Setup and Application in the Study of Relaxation Processes in Fe-C, Fe-Cu and Fe-Cr Alloys', *Electr. Rev.*, Vol. 87, 2011, p. 120.
[11] Meslin, E., Lambrecht, M., Hernández Mayoral, M., Bergner, F., Malerba, L., Pareige, P., Radiguet, B., Barbu, A., Gómez Briceño, D., Ulbricht, A., Almazouzi, A., ' Characterization of neutron-irradiated ferritic model alloys and a RPV steel from combined APT, SANS, TEM and PAS analyses', *J. Nucl. Mater.*, Vol. 406, 2010, p. 73.
[12] Lambrecht, M., Meslin, E., Malerba, L., Hernandez-Mayoral, M., Bergner, F., Pareige, P., Radiguet, B., Almazouzi, A., ' On the correlation between irradiation-induced microstructural features and the hardening of reactor pressure vessel steels', *J. Nucl. Mater.*, Vol. 406, 2010, p. 84.
[13] Malerba, L., Caro, A., Wallenius, J.,' Multiscale modelling of radiation damage and phase transformations: The challenge of FeCr alloys', *J. Nucl. Mater.*, Vol. 382, 2008, p. 112.
[14] Terentyev, D., Bonny, G., Domain, C., Monnet, G., Malerba, L., ' Mechanisms of radiation strengthening in Fe-Cr alloys as revealed by atomistic studies', *J. Nucl. Mater.*, Vol. 442, 2013, p. 470.
[15] Odette, G.R., Lucas, G.E., 'Recent progress in understanding reactor pressure vessel steel embrittlement', *Radiation Effects and Defects in Solids*, Vol. 144, 1998, pp. 189.
[16] Odette, G.R., Nanstad, R.K., 'Predictive reactor pressure vessel steel irradiation embrittlement models: Issues and opportunities', *JOM*, Vol. 61, 2009, p. 17.
[17] Ngayam-Happy, R., Becquart, C.S., Domain, C., Malerba, L., ' Formation and evolution of MnNi clusters in neutron irradiated dilute Fe alloys modelled by a first principle-based AKMC method', *J. Nucl. Mater.*, Vol. 426, 2012, p. 198.
[18] Bonny, G., Terentyev, D., Bakaev, A., Zhurkin, E.E., Hou, M., Van Neck, D., Malerba, L., ' On the thermal stability of late blooming phases in reactor pressure vessel steels: An atomistic study', *J. Nucl. Mater.*, Vol. 442, 2013, p. 282.
[19] Malerba, L., presentation given at IAEA TM on 'Degradation of primary components of pressurized water cooled nuclear power plants: current issues and future challenges', 6–8 November 2013, Vienna.
[20] Olander, D.R., 'Fundamental Aspects of Nuclear Reactor Elements', NTIS, ERDA, 1975.
[21] Garcia, P., Martin, G., Sabathier, C., Carlot, G., Michel, A., Martin, P., Dorado, B., Freyss, M., Bertolus, M., Skorek, R., Noirot, J., Noirot, L., Kaitasov, O., Maillard, S., ' Nucleation and growth of intragranular defect and insoluble atom clusters in nuclear oxide fuels ', *Nucl. Instrum. Meth. Res. B*, Vol. 277, 2012, p. 98.
[22] Almazouzi, A., Alamo, A., Lidbury, D., Moinereau, D., Van Dyck, S., ' PERFORM 60: Prediction of the effects of radiation for reactor pressure vessel and in-core materials using multi-scale modelling ects of radiation for reactor pres', *Nucl. Eng. Des.*, Vol. 241, 2011, p. 3403.
[23] Fazio, C., Gómez Briceño, D., Rieth, M., Gessi, A., Henry, J., Malerba, L., 'Innovative materials for Gen IV systems and transmutation facilities: The cross-cutting research project GETMAT', *Nucl. Eng. Des.*, Vol. 241, 2011, p. 3514.
[24] Olsson, P., Klaver, T.P.C., Domain, C., '*Ab initio* study of solute transition-metal interactions with point defects in bcc Fe', *Phys. Rev. B*, Vol. 81, 2010, p. 054102.
[25] Bonny, G., Pasianot, R.C., Malerba, L., 'Fitting interatomic potentials consistent with thermodynamics:Fe, Cu, Ni and their alloys', Phil. Mag., Vol. 89, 2009, p. 3451.
[26] Ngayam-Happy, R., Becquart, C.S., Domain, C., 'First principle-based AKMC modelling of the formation and medium-term evolution of point defect and solute-rich clusters in a neutron irradiated complex Fe–CuMnNiSiP alloy representative of reactor pressure vessel steels', *J. Nucl. Mater.*, Vol. 440, 2013, p. 143.

[27] Ngayam-Happy, R., Olsson, P., Becquart, C.S., Domain, C., 'Isochronal annealing of electron-irradiated dilute Fe alloys modelled by an ab initio based AKMC method: Influence of solute–interstitial cluster properties', *J. Nucl. Mater.*, Vol. 407, 2010, p. 16.

[28] Martinez, E., Senninger, O., Fu, C-C., Soisson, F., 'Decomposition kinetics *of* Fe-Cr solid solutions during thermal aging', *Phys. Rev. B*, Vol. 86, 2012, p. 224109.

[29] Castin, N., Pascuet, M.I., Malerba, L., 'Mobility and stability of large vacancy and vacancy-copper clusters in iron: An atomistic kinetic Monte Carlo study', *J. Nucl. Mater.*, Vol. 429, 2012, p. 315.

[30] Jansson, V., Malerba, L., 'Simulation of the nanostructure evolution under irradiation in Fe–C alloys', *J. Nucl. Mater.*, Vol. 443, 2013, p. 274.

[31] Jansson, V., Chiapetto, M., Malerba, L., 'The nanostructure evolution in Fe–C systems under irradiation at 560 K', *J. Nucl. Mater.*, Vol. 442, 2013, p. 341.

[32] Chiapetto, M., Malerba, L., 'Nanostructure evolution under irradiation in FeMnNi alloys: a 'grey alloy' object kinetic Monte Carlo model', to be published.

[33] Monnet, G., Osetsky, Y.N., Bacon, D.J., ' Mesoscale thermodynamic analysis of atomic-scale dislocation-obstacle interactions simulated by molecular dynamics', *Phil. Mag.*, Vol. 90, 2010, p. 1001.

[34] Terentyev, D., Monnet, G., Grigorev, P., 'Transfer of molecular dynamics data to dislocation dynamics to assess dislocation–dislocation loop interaction in iron', *Scripta Materialia*, Vol. 69, 2013, p. 578.

[35] Monnet, G., Vincent, L., Devincre, B., ' Dislocation-dynamics based crystal plasticity law for the low- and high-temperature deformation regimes of bcc crystal ', *Acta Materialia*, Vol. 61, 2013, p. 6178.

[36] Bruemmer, S.M., Simonen, E.P., Scott, P.M., Andresen, P.L., Was, G.S., ' Radiation-induced materials changes and susceptibility to intergranular failure of light-water-reactor core internals', *J. Nucl. Mater.*, Vol. 274, 1999, p. 299.

[37] Trinkaus, H., Singh, B.N., 'Helium accumulation in metals during irradiation - where do we stand?', *J. Nucl. Mater.*, Vol. 323, 2003, p. 229-242.

[38] Garner, F., Gelles, D., Greenwood, L., Okita, T., Sekimura, N., Wolfer, W., 'Synergistic influence of displacement rate and helium/dpa ratio on swelling of Fe-(9,12) Cr binary alloys in FFTF at ~400°C', *J. Nucl. Mater.*, Vol. 329–333, 2004, p. 1008-1012.

[39] Yamaguchi, M., 'First-Principles Study on the Grain Boundary Embrittlement of Metals by Solute Segregation: Part I. Iron (Fe)-Solute (B, C, P, and S) Systems', *Metall. Trans. A*, Vol. 42, 2011, p. 319.

[40] Ebihara K., Yamaguchi M., Nishiyama Y., Onizawa K, Matsuzawa H., 'Numerical Simulation of Irradiation-Induced Grain-Boundary Phosphorous Segregation in Reactor Pressure Vessel Steels Using Rate Theory Model With First-Principles Calculations', *ASME 2009 Pressure Vessels and Piping Conference*, Vol. 1: Codes and Standards (Prague, Czech Republic, July 26–30, 2009), p. 395.

[41] Zhang, L., Fu, C-C., Lu, G-H., 'Energetic landscape and diffusion of He in α-Fe grain boundaries from first principles', *Phys. Rev. B*, Vol. 87, 2013, p. 134107.

[42] Byun, T., Farrell, K., ' Plastic instability in polycrystalline metals after low temperature irradiation', *Acta Materialia*, Vol. 52, 2004, p. 1597.

[43] Sauzay, M., Evrard, P., ' Modelling of the effect of dislocation channel on intergranular microcrack nucleation in pre-irradiated austenitic stainless steels during low strain rate tensile loading', *J. Nucl. Mater.*, Vol. 405, 2010, p. 83.

[44] Nogaret, T., Rodney, D., Fivel, M., Robertson, C., ' Clear band formation simulated by dislocation dynamics: Role of helical turns and pile-ups', *J. Nucl. Mater.*, Vol. 380, 2008, p. 22.

[45] Arsenlis, A., Rhee, M., Hommes, G., Cook, R., Marian, J., 'A dislocation dynamics study of the transition from homogeneous to heterogeneous deformation in irradiated body-centered cubic iron', *Acta Materialia*, Vol. 60, 2012, p. 3748.

[46] Tomkins, B., 'Structural integrity issues in the nuclear industry', *Comprehensive Structural Integrity*, Vol. I, 2003, p. 173.

[47] Garner, F.A., Toloczko, M.B., Sencer, B.H., ' Comparison of swelling and irradiation creep behavior of fcc-austenitic and bcc-ferritic/martensitic alloys at high neutron exposure', *J. Nucl. Mater.*, Vol. 276, 2000, p. 123.

[48] Garner, F.A., Toloczko, M.B., 'Irradiation creep and void swelling of austenitic stainless steels at low displacement rates in light water energy systems', *J. Nucl. Mater.*, Vol. 251, 1997, p. 252.

[49] Barashev, A.V., Golubov, S.I., ' Unlimited damage accumulation in metallic materials under cascade-damage conditions', *Phil. Mag.*, Vol. 89, 2009, p. 2833.

[50] Matthews, J.R., Finnis, M.W., ' Irradiation Creep Models - an Overview', *J. Nucl. Mater.*, Vol. 159, 1988, p. 257.

[51] Soler Crespo, L., 'Compatibility of Structural Materials with LBE and Pb: Standardisation of Data, Corrosion Mechanism and Rate', in *Handbook on Lead-bismuth Eutectic Alloy and Lead Properties, Materials Compatibility, Thermal-hydraulics and Technologies*, NEA OECD, 2008 (Chapter 6).

[52] Valot, C., Bertolus, M., Konings, R., Somers, J., de Groot, S., ' Basic research in support of innovative fuels design for the GEn IV systems: The F-BRIDGE project ', *Nucl. Eng. Des.*, Vol. 241, 2011, p. 3521.

[53] De Bruycker, F., Boboridis, K., Manara, D., Pöml, P., Rini, M., Konings, R.J.M., ' Reassessing the melting temperature of PuO_2', *Materials Today*, Vol. 13, 2010, p. 52.

[54] Guéneau, C., Dupin, N., Sundman, B., Martial, C., Dumas, J.-C., Gossé, S., Chatain, S., De Bruycker, F., Manara, D., Konings, R.J.M., ' Thermodynamic modelling of advanced oxide and carbide nuclear fuels: Description of the U–Pu–O–C systems ', *J. Nucl. Mater.*, Vol. 419, 2011, p. 145.

[55] Bertolus, M., Freyss, M., Martin, G., Dorado, B., Van Brutzel, L., Chartier, A., Grimes, R., Parfitt, D., Bishop, C., Krack, M., Devynck, F., Govers, K., 'Development and assessment of atomistic modelling methods for the investigation of fuel materials under operational conditions', F-BRIDGE Deliverable D-221, 2010 (http://f-bridge.eu/).

[56] Dorado, B., Amadon, B., Freyss, M., Bertolus, M., ' DFT+U calculations of the ground state and metastable states of uranium dioxide ', *Phys. Rev. B*, Vol. 79, 2009, p. 235125.

[57] Yakub, E., Ronchi, C., Staicu, D., ' Computer simulation of defects formation and equilibrium in non-stoichiometric uranium dioxide ', *J. Nucl. Mater.*, Vol. 389, 2009, p. 119.

[58] Dorado, B., Garcia, P., Freyss, M., Carlot, G., Fraczkiewicz, M., Pasquet, B., Baldinozzi, G., Simeone, D., Freyss, M., Bertolus, M., ' First-principles calculation and experimental study of oxygen diffusion in uranium dioxide ', *Phys. Rev. B*, Vol. 83, 2011, p. 035126.

[59] Govers, K., Lemehov, S.E., Hou, M., Verwerft, M., ' Molecular dynamics simulation of helium and oxygen diffusion in $UO_{2\pm x}$ ', *J. Nucl. Mater.*, Vol. 395, 2009, p. 131.

[60] Devynck, F., Iannuzzi, M., Krack, M., ' Frenkel pair recombinations in UO_2: Importance of explicit description of polarizability in core-shell molecular dynamics simulations ', *Phys. Rev. B*, Vol. 85, 2012, p. 184103.

[61] Dorado, B., Andersson, D.A., Stanek, C.R., Bertolus, M., Uberuaga, B.P., Martin, G., Freyss, M., Garcia, P., ' First-principles calculations of uranium diffusion in uranium dioxide ', *Phys. Rev. B*, Vol. 86, 2012, p. 035110.

[62] Andersson, D.A., Uberuaga, B.P., Nerikar, P.V., Unal, C., Stanek, C.R., ' U and Xe transport in $UO_{2\pm x}$: Density functional theory calculations ', *Phys. Rev. B*, Vol. 84, 2011, p. 054105.

[63] Thompson, A.E., Wolverton, C., ' Pathway and energetics of xenon migration in uranium dioxide ', *Phys. Rev. B*, Vol. 87, 2013, p. 104105.

[64] Michel, A., ' *Etude du comportement des gaz de fission dans le dioxyde d'uranium : mécanismes de diffusion, nucléation et grossissement de bulles* ', PhD thesis, Université de Caen Basse-Normandie, France, 2011.

[65] Martin, G., Garcia, P., Sabathier, C., Van Brutzel, L., Dorado, B., Garrido, F., Maillard, S., ' Irradiation-induced heterogeneous nucleation in uranium dioxide ', *Phys. Letters A*, Vol. 374, 2010, p. 3038.

[66] Murphy, S., Chartier, A., Van Brutzel, L., Crocombette, J.P., ' Free energy of Xe incorporation at point defects, in nanovoids and bubbles in UO_2 ', *Phys. Rev. B*, Vol. 85, 2012, p. 144102.

[67] Michel, A., Sabathier, C., Carlot, G., Kaitasov, O., Bouffard, S., Garcia, P., Valot, C., ' An in situ TEM study of the evolution of Xe bubble populations in UO_2 ', *Nucl. Instrum. Methods B*, Vol. 272, 2012, p. 218.

[68] Michel, A., Sabathier, C., Carlot, G., Cable, M., Bouffard, S., Garcia, P., ' A TEM study of bubbles growth with temperature in xenon and krypton implanted uranium dioxide ', *Defect Diffus. Forum*, Vol. 323–325, 2012, p. 191.

[69] Bertolus, M., Freyss, M., Dorado, B., Martin, G., Hoang, K., Maillard, S., Skorek, R., Garcia, Ph., Valot, C., Chartier, A., Van Brutzel, L., Fossati, P., Grimes, R.W., Parfitt, D.C., Bishop, C.L., Murphy, S.T., Rushton, M.J.D., Staicu, D., Yakub, E., Nichenko, S., Krack, M., Devynck, F., Ngayam-Happy, R., Govers, K., 'Multi-scale exercise on transport properties in uranium dioxide', F-BRIDGE Deliverable D-226, 2012 (http://f-bridge.eu/).

[70] Skorek, R., Maillard, S., Michel, A., Carlot, G., Gilabert, E., Jourdan, T., ' Modelling Fission Gas Bubble Distribution in UO_2 ', *Defect Diffus. Forum*, Vol. 323–325, 2012, p. 209.

[71] Pizzi, E., Garcia, P., Carlot, G., Palancher, H., Maillard, S., Pasquet, B., Roure, I., Pozo, C., Maurice, C., ' Iodine volume diffusion measurements in uranium dioxide ', *Defect Diffus. Forum*, Vol. 323–325, 2012, p. 197.

[72] Dorado, B., Freyss, M., Martin, G., ' GGA+U study of the incorporation of iodine in uranium dioxide ', *Eur. Phys. J. B*, Vol. 69, 2009, p. 203.

[73] Crocombette, J.-P., ' First-principles study with charge effects of the incorporation of iodine in UO_2 ', *J. Nucl. Mater.*, Vol. 429, 2012, p. 70.

[74] Kotomin, E.A., Mastrikov, Y.A., Rashkeev, S.N., Van Uffelen, P., ' Implementing first principles calculations of defect migration in a fuel performance code for UN simulations ', *J. Nucl. Mater.*, Vol. 393, 2009, p. 292.

[75] Laux, D., Baron, D., Despaux, G., Kellerbauer, A.I., Kinoshita, M., ' Determination of high burn-up nuclear fuel elastic properties with acoustic microscopy ', *J. Nucl. Mater.*, Vol. 420, 2012, p. 94.

[76] Parfitt, D.C., Bishop, C.L., Wenman, M.R., Grimes, R.W., ' Strain field and line energies of dislocations in uranium dioxide ', *J. Phys.: Condens. Matt.*, Vol. 22, 2010, p. 175004.

[77] Van Brutzel, L., Fossati, P., Grimes, R., Parfitt, D., Murphy, S., 'Thermo-mechanical properties of complex microstructures', F-BRIDGE Deliverable D-224, 2011 (http://f-bridge.eu/).

[78] Martin, G., Maillard, S., Van Brutzel, L., Garcia, P., Dorado, B., Valot, C., ' A molecular dynamics study of radiation induced diffusion in uranium dioxide ', *J. Nucl. Mater.*, Vol. 385, 2009, p. 351.

[79] Staicu, D., Wiss, T., Rondinella, V.V., Hiernaut, J.-P., Konings, R.J.M., Ronchi, C., ' Impact of auto-irradiation on the thermophysical properties of oxide nuclear reactor fuels ', *J. Nucl. Mater.*, Vol. 397, 2010, p. 8.

[80] Nerikar, P.V., Rudman, K., Desai, T.G., Byler, D., Unal, C., McClellan, K.J., Phillpot, S.R., Sinnott, S.B., Peralta, P., Uberuaga, B.P., Stanek, C.R., ' Grain Boundaries in Uranium Dioxide: Scanning Electron Microscopy Experiments and Atomistic Simulations', *J. Am. Ceram. Soc.*, Vol. 94, 2011, p. 1893.

[81] Vincent-Aublant, E., Van Brutzel, L., Delaye, J.M., ' Self-diffusion near symmetrical tilt grain boundaries in UO_2 matrix: a molecular dynamics simulation study ', *J. Nucl. Mater.*, Vol. 392, 2009, p. 114.

[82] Nerikar, P.V., Parfitt, D.C., Casillas Trujillo, L.A., Andersson, D.A., Unal, C., Sinnott, S.B., Grimes, R.W., Uberuaga, B.P., Stanek, C.R., ' Segregation of xenon to dislocations and grain boundaries in uranium dioxide ', *Phys. Rev. B*, Vol. 84, 2011, p. 174105.

[83] Govers, K., Lemehov, S.E., Verwerft, M., ' Molecular dynamics study of grain boundary diffusion of fission gas in uranium dioxide ', *Defect Diffus. Forum*, Vol. 323–325, 2012, p. 215.

[84] Van Brutzel, L., Aublant-Vincent, E., ' Large molecular dynamics simulations of collision cascades in mono-crystal, bicrystal, and poly-crystal UO_2', *Nucl. Instr. Methods B*, Vol. 267, 2009, p. 3013.