Journal of Nuclear Materials 406 (2010) 2–6

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

Editorial

PERFECT – Prediction of Irradiation Damage Effects on Reactor Components: A summary

In nuclear power reactors, materials undergo degradation due to severe irradiation conditions that may limit their operational life. Utilities that operate these reactors need to quantify the ageing and the potential degradations of Reactor Pressure Vessels (RPVs) and also of RPV Internals structures to ensure safe and reliable plant operation.

RPV issues: The RPV is the critical component for plant life management, due to the unacceptable consequences of its failure and due to the difficulty of its replacement. The RP Vessel is subjected to neutron irradiation in the beltline region, which results in irradiation-induced embrittlement that may lead to a shift of the ductile-to-brittle transition temperature. To demonstrate the absence of risk of failure by brittle fracture, it is necessary to assess the admissible ductile–brittle transition temperature for the most severe mechanical and thermal loadings applied to postulated defects. For this purpose, codified integrity assessment methodologies and criteria have been defined (ASME Code, French RCC-M Code). Safety margins are computed, e.g. for representative Pressurized Thermal Shock (PTS) transients, as the ratio between the stress intensity factor at the tip of hypothetical under-clad defects, and the fracture toughness of the RPV steel. Then, the prediction of the fracture toughness curve versus temperature and the prediction of the irradiation-induced shift are major inputs.

RPV Internals issues: The stainless steel Internal Structures of LWR vessels are closest to the core and are thus subjected to intense neutron irradiation; the most irradiated areas of some of these components (PWR internals) may be exposed to doses reaching around 80 dpa after 40 years of operation (that is more than a thousand times higher than the vessel). Due to neutron irradiation, their microstructure changes and consequently so does their mechanical properties: Hardening, strong decrease in ductility and toughness, irradiation creep, possibly void swelling, etc. In addition, these changes seem to be the basis of an increased sensitivity to stress corrosion (said to be assisted by irradiation – IASCC). In service intergranular cracking of some baffle-former bolts has been observed in several PWRs and some of them needed to be repaired. The effects of neutron irradiation on materials for the core internals are then potentially a significant economic and regulatory concern for utilities. Then predictions of irradiation hardening, loss of ductility, void swelling and IASCC sensitivity coupled with in service inspections and safety analysis are necessary to ensure safe and reliable plant operation. At present, predictive models of irradiation-induced hardening are available, but the predictions of void swelling and IASCC sensitivity are non-existent or not reliable.

Continuous progress in the physical understanding of the phenomena involved in irradiation damage and continuous progress in computer sciences have made possible the development of multi-scale numerical tools able to simulate the effects of irradiation on materials microstructure.

The consequences of irradiation on mechanical and corrosion properties of materials are also tentatively modelled using such multi-scale modelling. But it is necessary to develop different mechanistic models at different levels of physics and engineering and to extend the state of knowledge in several scientific fields. The links between these different kinds of model are particularly difficult to deal with and need special techniques, which have yet to be fully realised.

An example of the different scales needed to investigate neutron irradiation embrittlement in the case of the RPV steels is given in Fig. 1.

1. Nature and scope of the PERFECT project

The PERFECT project was initiated in January 2004, under the overall co-ordination of Electricité de France (EDF), as part of the Sixth Framework of the European Atomic Energy Community (EURATOM), for 54 months.

The PERFECT consortium comprised 12 organizations involved in nuclear R&D, which include 1 manufacturer, 1 electric utility and 8 nuclear research centres operating hot cell facilities. Specialised skills required by the project in physics, irradiation effects, numerical simulation and advanced techniques of material characterization were brought by 16 universities and research centres.

The project PERFECT (Prediction of Irradiation Damage Effects in Reactor Components) aimed at developing and building predictive tools for quantifying the effects of neutron irradiation damage on Reactor Pressure Vessels and Internal Structures. In particular, the project focused on the development of predictive tools to model the fracture toughness versus temperature dependence of irradiated RPV steels and also, as a first attempt, development of tools to predict the irradiation hardening, plastic flow channeling, void swelling and IASCC sensitivity of Internals components.

For the successful achievement of these multi-scale models, an integration process was required. For that the numerical couplings of the different modules was realised on a software platform.

The main objective of PERFECT was to build 2 'Virtual Reactors' simulating the effect of neutron irradiation respectively on RPV fracture toughness and on RVP Internal Structures behaviour.

0022-3115/$ - see front matter © 2010 Published by Elsevier B.V.
doi:10.1016/j.jnucmat.2010.09.029

Specific modules and databases – dealing with damage production, microstructure evolution, mechanical and corrosion phenomena induced by irradiation – complement each of these Virtual Reactors. The resulting numerical tools were integrated in a Software Integration Platform.

2. Project organization

To reach its objectives, PERFECT was organized into four technical sub-projects as follows:

- Physics Modelling of RPV and internals materials, to predict the evolution of radiation-induced microstructures from first principles.
- Mechanics Modelling of the RPV, to predict the fracture behaviour of Reactor Pressure Vessel components as a function of material type and characteristics, loading and irradiation conditions.
- Mechanics and corrosion modelling of RPV internals, to formulate predictive models for initiation and propagation of radiation-induced stress corrosion cracks in irradiated stainless steels.
- Integration. The main activities of this sub-project are the numerical coupling of the calculation modules. The results of this coupling was realised on the Integration software platform, which provides a common computer architecture.

3. Main achievements

3.1. "Physics Modelling of RPV and Internals" main achievements

The overall objective of the sub-project, Physics Modelling, was to develop advanced numerical tools based on the physics at the atom scale where radiation effects can be reliably described. These tools were calibrated versus experimental data that either exist or were complemented within PERFECT.

The different scales and tools necessary for multi-scale modelling of radiation-induced microstructures are summarized in Fig. 2.

Atom-based multi-scale modelling has been developed essentially either for pure metals under irradiation or for simple alloys under thermal ageing. The major alloying elements of RPV steels (interstitials like C, and substitution such as Cu, Ni, Si, Mn) are known to be determinant in the evolution of the microstructure, and, the hardening and loss of fracture toughness of irradiated RPV steels. The originality of PERFECT is the objective to introduce a physical description at the atomic scale of the effect of these alloying elements. Concerning the behaviour of austenitic steels, the effect of He on the prediction of microstructure evolution and possible occurrence of swelling, as well as the reaction between dislocation and point defects at the origin of the localisation of the plastic deformation, is also based on the physics at the atom scale. This strategy implies a careful assessment of the properties of point defects and their interaction with the alloying elements and impurities in the actual materials.

3.1.1. Ab initio calculations and empirical potentials

A large set of DFT data on configuration energies of point-defects and their clusters, as well as migration energies in pure α-Fe, including the effect of solute–defect interaction, both for substitutional (Cu, Ni; Mn; Si, P) and interstitial (C, N) solute atoms has been produced. Data relevant for austenitic alloys are still limited in number, but some of them exist and more are being produced. The work that the DFT community should mainly face, now, is that of mutual validation of results. Generally VASP and SIESTA data are consistent and when this happens the data themselves become more trustworthy. However, the choice of different DFT approaches may produce somewhat different results and it is important to establish the range of variability when these differences appear, particularly in that case where not only quantitative, but also qualitative differences may arise. The case where this discrepancy is most worrying and important at the same time is that of the non-parallel SIA cluster configurations.

The knowledge acquired from DFT studies is being transferred, using one approach or another, to empirical interatomic potentials and is also being directly used to fit kinetic Monte Carlo models. The EAM-like advanced potentials for pure Fe can be divided in two classes: Mendelev-type and 'magnetic'. At the moment, although both represent a clear improvement compared to earlier potentials, the former class is found to perform better than the second one when compared to *ab initio* data. The reason of the difference is likely to be ascribable purely to the care put in the

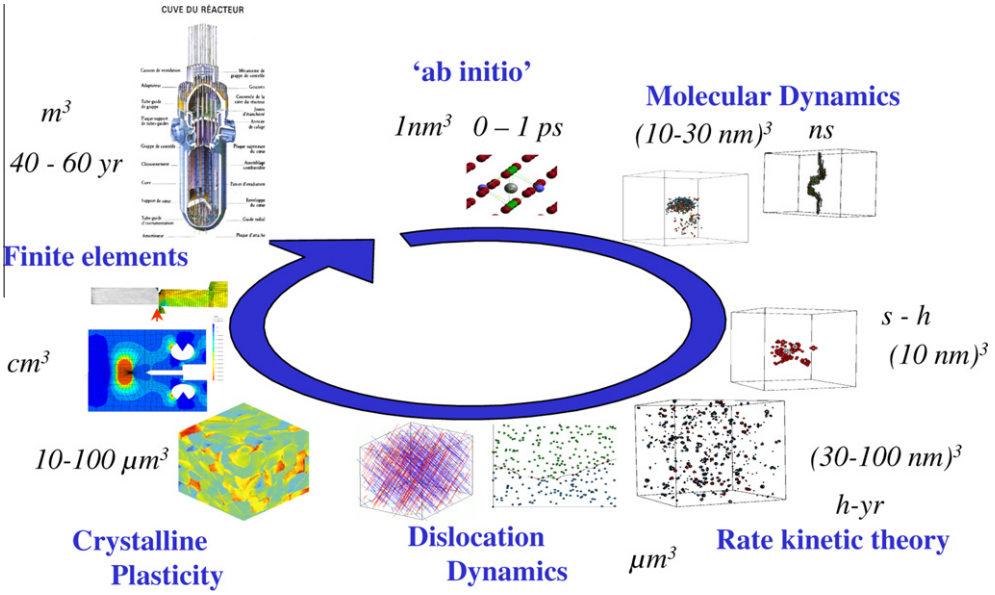

Fig. 1. Multi-scale modelling for RPV steels.

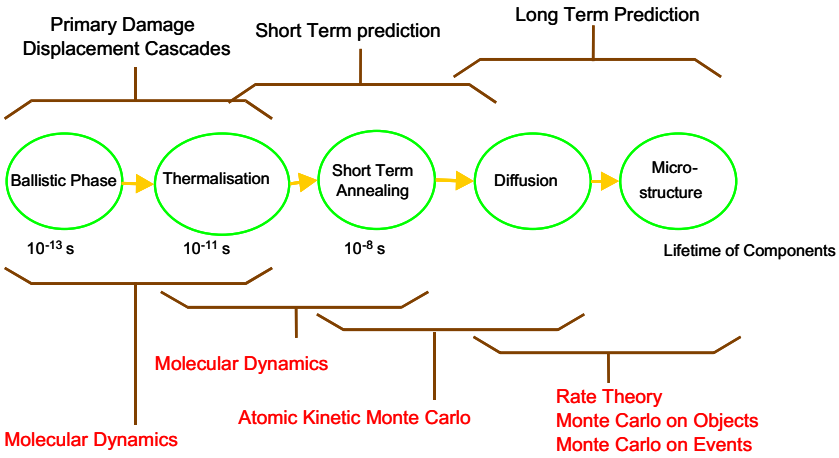

Fig. 2. Scales and tools for multi-scale modelling of radiation-induced microstructures.

fitting. In itself, the formalism of the 'magnetic' potential, by including explicitly an assessment of the magnetic contribution to the total energy, is interesting and promising. However, more work on its parameterisation is needed. The Mendelev-type potentials already allow, to some extent, DFT studies to be extrapolated to ranges inaccessible to DFT. However, on many instances differences exist between potentials of this same class and whenever a clear *ab initio* or experimental reference is lacking, the results must be taken with care and considered as possibilities, rather than certainties.

On the Fe alloy potential side, within the framework of reliability of Mendelev-type potentials for Fe, advances have been also made and these were determined not only by the systematic use of *ab initio* data for the fitting, but also by the attention paid to a correct reproduction of the thermodynamic properties of the alloys, especially in the case of substitutional alloys, such as FeCu and FeNi. The problem of FeC, as expected, was a particularly tough one, especially if the existence of covalent CC bonding has to be allowed for. Nonetheless, two FeC potentials are at least available for studies concerning the interaction of point-defect clusters and dislocations with a single C atom, one of them being able to reproduce correctly most *ab initio* configuration energies, including repulsion between a C atom and close-by dumbbells.

The available potentials do not cover, yet, the full range of elements that appear in RPV steels However, an FeCuNi ternary potential – the first of this kind to our knowledge – is now available and complex substitutional element effects can be now studied at the atomic level at the scale accessible to Molecular Dynamics, as well as, separately, the effects of the presence of C. In the case of austenitic steels, FeNi is the best model alloy immediately at reach, but the possibility of fabricating a ternary potential for an FeNiCr ternary alloy may be now possible.

In a broader perspective, i.e. including work done outside the project and looking forward to a continuation of the Project, the landscape is encouraging. It is easy to see that future projects, for both internals and RPV steels, may take seriously the possibility of developing potentials for multi-component systems.

3.1.2. Cascades and short term evolution in RPV

Most of AKMC codes usually used to study solute and point-defects clustering or precipitation kinetics are based on simple "broken bonds model" to compute the point defects migration barriers and jump frequencies. During the PERFECT program a key improvement has been the generalisation of *ab initio* calculations based on the density functional theory to fit the corresponding AKMC parameters. This gives access to atomic diffusion properties quite difficult to get from experimental data and more reliable than the ones obtained from empirical potentials. Such an approach has been used by the groups of EDF and the University of Lille and by the group of CEA Saclay (the resulting AKMC codes differs in the details of the migration barrier models and in the use of different *ab initio* approximations).

The other essential development observed in the framework of the PERFECT program has been the introduction of the self-interstitial diffusion mechanism, with the same level of description developed these last years for the vacancy mechanism. First simulations of precipitation and segregation phenomena under irradiation, using these recent developments have been performed.

Ab initio calculations are nevertheless too time consuming to compute more than a few representative migration barriers (not to speak of on-the-fly calculations): The Hybrid KMC method developed at CEN•SCK, with ANN methods using an EAM potential and MD techniques to compute a much larger number of migration barriers, has proven to be quite efficient in the case of vacancy jumps. The application to the dumbbell diffusion (with specific technical difficulties) is under progress and it should soon possible to assess its application to precipitation kinetics in particular alloys.

It is worth to notice however, that even with the previous progress, key improvements remains necessary to get quantitative predictive tools. In all the previous studies for example, the only interstitial diffusion mechanism taken into account is the rotation–translation jump of the 110 dumbbells, while *ab initio* calculations suggest that in copper rich configurations (and in most of other bcc transition metals), the 111 dumbbells could be more stable.

Even with such an approximation and using simple rigid lattice model with only pair interactions, AKMC simulations are still very time consuming and then limited to short evolution times. This is especially true for Fe–Cu dilute alloys, because of strong trapping effects of point defects in copper rich clusters and strong correlations effects.

3.1.3. Long term evolution in RPV

This task deals with the improvement of simulation tools to reproduce the long-term evolution of irradiation-induced hardening defects and segregations in RPV steels. It is also in charge of the quantitative characterization of the microstructure of irradiated steels to validate simulation results.

The experimental programme consists in measurements of irradiation damage and segregations with a large panel of techniques on pure Fe and different types of Fe alloys (FeC, FeCu, FeMnNi and

RPV steels). The main conclusions that could be drawn from these characterizations of the microstructure of irradiated materials are the following:

For all the materials: Irradiation hardening is observed and the increase of the yield stress is approximately linear with the square root of the dose.

For Pure Iron and Fe C: Neutrons induced the formation of interstitial loops (mostly of ⟨1 0 0⟩ type), vacancy clusters and nanovoids at high dose. A lower dose rate introduces less int-clusters but more V-type. C decreases the size and increase the density of all defects.

For FeCu binary alloy: Cu decreases the size of both V-clusters and I-clusters. Nucleation of Cu clusters already achieved at low dose. These clusters growth with dose by co-clustering of vacancies and Cu. Kinetic of precipitation is faster at low dose rate.

For FeMnNi and FeCuMnNi alloys: The presence of Mn and/or Ni hinder the clustering of vacancies. Interstitial clusters are very small. MnNi(P) clusters are observed at high dose rate/high dose and at low dose rate. Kinetic of precipitation is faster at low dose rate.

CuMnNi(P) clusters nucleate even at low dose but do not grow with dose. Nucleation of Cu free clusters is observed at high dose/high dose rate, and at low dose rate.

For RPV steels: Irradiation hardening is measured but no defects have been observed by TEM. However nano-clusters of one or two tens of atoms were detected by TAP (Mn, Ni, Cu, etc.).

For the simulation tools, two types of methods are used: Monte Carlo on object or on events on one hand and rate theory on the other. All these methods are based on the same set of parameters: Diffusion coefficients capture radii, concentrations of point-defect clusters, binding energy of point defects or of solute atoms in clusters. The implementation of relevant parameters and mechanisms to simulation e-irradiation of Fe and FeC has been compared to Takaki experimental results using the SIESTA-calculations that are in good agreement with VASP. The KMCE simulations allowed the elucidation of some of the open question related to the defect recovery stages in Fe and FeC.

Concerning RT codes in RPV-1, the RT code MFVIC by CEA is now working with multi-sink strengths, improved capture radii, and, a stable numerical scheme implemented by EDF. FZD has introduced, for the case of precipitation, interfacial energy effects and evaluate the sensitivity of radiation-induced microstructure to variations of these parameters. Due to the assumption of homogenization, the parameters describing point defect and atom diffusion and binding energies are effective ones that have to be adjusted on the observed microstructure. Also, some effort has been done to update the rate theory code that exist already in RPV-1, and the code has been delivered to be used as complementary to MFVIC in the RPV-2 version.

3.1.4. Discrete Dislocation Dynamics in RPV

Molecular Dynamics (MD) simulation was used to investigate dislocation proprieties at the atomic level and to provide a database of pinning forces of irradiation-induced defects.

Dislocation Dynamics (DD) simulations are expected to give a comprehensive and quantitative description of dislocation microstructures and mesoscopic mechanical behaviour. The database of pinning forces is used to account, on the mesoscopic scale, for the irradiation-induced strengthening. MD results are thus integrated in the mesoscopic scale according to a physically based model. This allows the comparison and validation with experimental results on the irradiation-induced hardening in RPV steels.

Plasticity of the un-irradiated material: DD simulations have been used to predict strengthening due to carbides in RPV steel. The mobility law already constructed for dislocations is found to provide a good description of dislocation behaviour. They are therefore used to investigate the effect of carbide on the flow stress. The interaction of infinite dislocations with infinite periodic row of carbides was studied. The study characterizes the temperature and rate effects on the interaction and the influence of the carbide distribution on the flow stress. The increase in critical resolved shear stress induced by carbides is found to be of the order of 40 MPa. This is two times smaller than the forest hardening.

Analysis of micro-cleavage stability: The temperature dependence of the critical cleavage stress has been investigated. The proposed treatment is adapted to micro-defects characteristic size a much smaller than the characteristic lath dimensions. Stress distributions in the three {1 0 0} cleavage planes are obtained at two different temperatures. The results reveal that the dislocation microstructure preferentially screens out the stress acting in the [0 1 0] and [0 0 1] cleavage planes, where the (projected) applied stress and the temperature are larger. This could be a track to follow to explain the apparent increase critical cleavage stress with temperature.

Plasticity of the irradiated material: Using the Mendelev potential as the only input data, MD simulations provided the following results: The interaction with copper cluster and vacancy cluster seems to lead to only a shear of the clusters. The interaction with vacancies is stronger than with copper atoms for an identical cluster size. The interaction with interstitial loops lead to the formation of an helicoidal turn, leading to the strongest interaction. DD simulation of dislocation interaction with voids have been performed and are in fair agreement with macroscopic results: The predicted relative increase of critical resolved shear stress is close to the measured macroscopic increase of yield stress.

3.1.5. Mechanics Modelling of RPV

The objective of the "Mechanics Modelling" sub-project is to provide an accurate, physically based description of the fracture behaviour of Reactor Pressure Vessel components as a function of material type, composition and characteristics, loading, temperature and irradiation conditions. The sub-project provided a verified "fracture toughness" module on the platform of PERFECT for the purpose of simulating the behaviour of cracks in irradiated RPV components. The fracture toughness module is based on sub-modules that describe the flow and fracture behaviour of irradiated materials at length scales starting with discrete dislocations (output of DDD model from Physics SP), to single crystals, to polycrystalline aggregates (test specimens, components).

The interaction of dislocations with hardening defects was studied by Molecular Dynamics or Discrete Dislocations Dynamics (DDD) in the sub-project Physics Modelling. Finally, the critical resolved shear stress is computed on each slip system, which is the critical input to determine the macroscopic yield strength. However, output from MD, KMC or even DDD models cannot directly be converted at the macroscopic scale into a yield strength or hardening modulus. To do this, an intermediate level of modelling is required at the granular scale (Fig. 3). The overall strategy adopted for the achievement of multi-scale fracture modelling is to use crystal plasticity and local approach methods to derive macroscopic properties (stress–strain curves, toughness) from DDD simulations.

A crystal plasticity model has been calibrated for bainitic steel, with an explicit introduction of hardening effect due to carbides. Macroscopic response, stress levels in the ferrite matrix and stress heterogeneities in the representative volume are consistent with experimental data. This provided a fine micro-mechanical description of a typical RPV steel, with an evaluation of the local stress fields involved in the cleavage nucleation criterion. The development of this formulation built also a strong link between simulations proposed by the Physics Modelling sub-project, at a lower

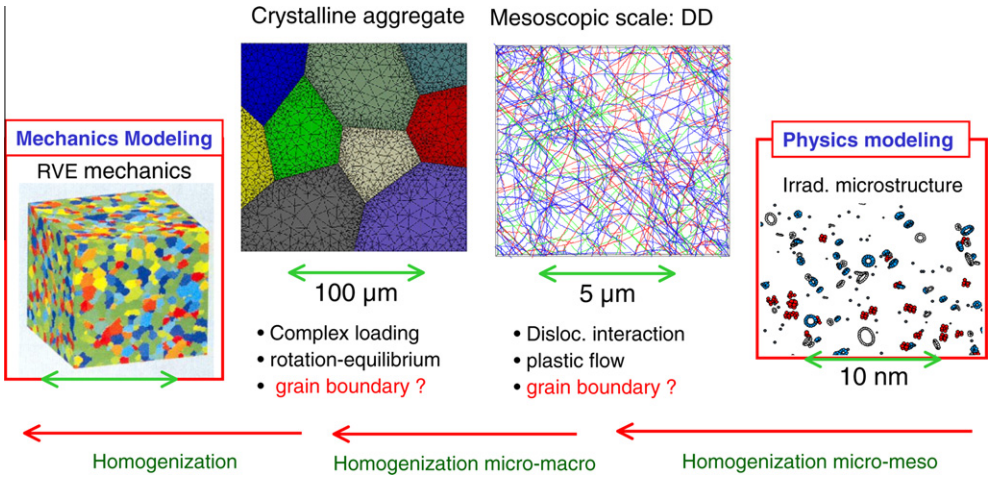

Fig. 3. Necessary transitions between Physics Modelling and Mechanics Modelling.

scale, and classical continuum mechanics and Finite Element analyses.

3.2. Mechanics and corrosion of internals and "IASCC module"

The overall objective of the sub-project was to develop the IASCC code module, describing the stress corrosion cracking behaviour of irradiated stainless steels. The IASCC module consists of a crack initiation and a crack propagation module.

3.2.1. Crack propagation modelling

The simulation uses the approximation of the slip dissolution model. The crack propagation rate was determined by the crack tip strain rate (linked to the load, the geometry and the material mechanical properties (yield stress and plastic strain hardening behaviour)) and the electrochemical repassivation behaviour (determined by the local environment and the local material composition). The electrochemical repassivation behaviour was determined by the local electrochemistry, given by the equilibrium between the inner crack environment and the outer crack environment (conservation of charge).

3.2.2. Crack initiation modelling

The final tool has been specified as a crack nucleation, coalescence and growth model, based on empirical rules for crack nucleation, interaction and growth. The first prototype of this module was parameterised using literature data for sensitised stainless steels in oxygenated water (BWR relevant conditions).

In the second version of the initiation module, experimental data were used for parameterisation, taken from crack initiation tests, performed on model materials in primary water.

In parallel, work has been done on the development of supporting – complementary modules:

- *Environmental chemistry module*: This module is supporting the crack propagation module of the final product.
- *Mesoscopic crack propagation modelling*: This module allows the quantification of the effect of flow localisation on the stress and strain distribution in stressed irradiated stainless steels. It is developed as a complement to the analytic approximation of the slip dissolution model of the CGR module of the end product.
- *Microscopic crack propagation modelling*: This module describes the strain distribution around a crack, based on Discrete Dislocation Dynamics.
- *Oxide film behaviour modelling*: Based on the point defect model, the transport phenomena across an oxide film can be modelled. But more work is needed to link the transport phenomena to the vacancy transport in the metal substrate and the nucleation of cracks at the grain boundaries.

Jean-Paul Massoud
EDF

Stéphane Bugat
EDF

Bernard Marini
CEA

David Lidbury
SERCO

Steven Van Dyck
SCK.CEN