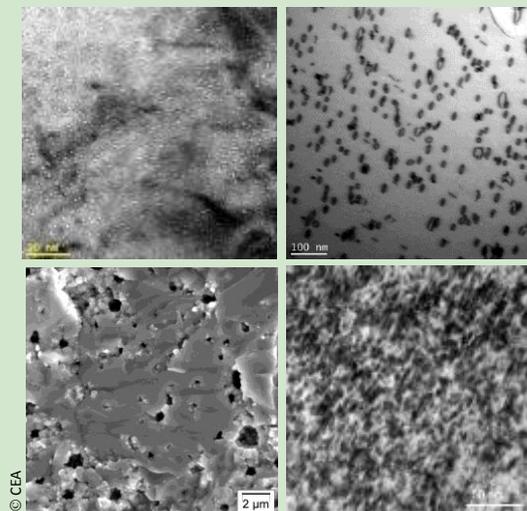




Centre of Excellence for Nuclear Materials

2nd International Workshop

Irradiation of Nuclear Materials: Flux and Dose Effects



November 4-6, 2015
CEA – INSTN Cadarache, France



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RJH Reactor under construction at the CEA Cadarache



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Front area of the LECA hot facility (Fuel Research Department, CEA Cadarache)

Content

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- List of speakers and lists of local scientific committee members, organizers and sponsors

Scope and objective of the workshop

Created in 2011 by the Nuclear Energy Division of CEA, the French leader for research on materials for nuclear applications, the **MINOS Centre of Excellence for Nuclear Materials** sustains fundamental and applied research related to the behavior of nuclear materials and structures in severe environments. The main objectives of MINOS research are increasing the safety level and lifespan of current reactors, optimizing waste management, and developing new materials for generation IV systems, in particular fast neutron reactors. To accomplish these goals, it is essential to identify and understand the mechanisms of irradiation damage and to develop predictive models. These models are not only necessary to describe irradiation-induced microstructure evolution but also to predict the role of this microstructure in the evolution of the mechanical behavior and the material properties. In this domain, neutron irradiation is an important source of atomic-scale microstructural disorders (cascades) that generate a large number of structural defects (vacancies, loops, solute clusters, segregation, precipitates...). These defects could lead to various phenomena that affect the macroscopic physical and mechanical properties, such as hardening, embrittlement, swelling, creep, and cracking. These detrimental phenomena can reduce the efficiency and operability of nuclear power plant parts and components (i.e. fuel, claddings, internals, pressure vessels for PWR's).

MINOS intends to promote research on nuclear materials, to reinforce the visibility of CEA in this field, to identify new topics of research, and to contribute to training activities in materials science. To help achieve these objectives, MINOS organized an international workshop on “Materials Innovation for Nuclear Optimized Systems” in December 2012 and an international school on the “Modeling of Corrosion for Nuclear Reactors” in April 2014.

The 2015 MINOS international workshop on “Irradiation of Nuclear Material: Flux and Dose Effects” will provide the opportunity to address specific topics related to state-of-the-art research on the effects of irradiation on materials used in existing reactors and materials intended for future nuclear systems for fission or fusion. A large part of the workshop will be dedicated to structural materials (mainly metallic alloys) and fuels materials, but other materials, such as composites, nuclear glass, and polymers, will also be considered. Emphasis will be given to flux and dose effects. The workshop will consist of four sessions, and each one will combine keynotes by leading scientists and lectures by experts in materials science and nuclear engineering.

The workshop will be held at the National Institute for Nuclear Science & Technology (INSTN) in Cadarache, France. It is organized by the CEA Nuclear Energy Division with the support of the INSTN.

2nd MINOS workshop: program

Wednesday, November 4, 2015

- 8:15 • Registration
- 9:00 • Welcome, **C. Gallé (MINOS)**, CEA Saclay (France)
- 9:05 • Opening Talk, **J.-P. Bouyer (DEN/DANS/Dir)**, CEA Saclay (France)

SESSION 1: IRRADIATION TOOLS AND FACILITIES, NOV. 4TH, (a.m.)

Chairman: **R. P. Webb**, University of Surrey (UK) – Co-Chairman: **J.-L. Béchade**, CEA Saclay (France)

- 9:20 • **EMIR Federation: Network of Accelerators Dedicated to Material Irradiation**, **S. Bouffard et al.**, CEA Saclay (France)
- 10:00 • Surrey Ion Beam Centre: the EPSRC “MRF” for Ion Beam Applications, **R. P. Webb**, University of Surrey (Surrey, UK)
- 10:25 • The Jules Horowitz Reactor Research Project : a New High Performance Material Testing Reactor Working as an International User Facility – First Developments to Address R&D on Material, **G. Bignan et al.**, CEA Cadarache (France)
- 10:50 Coffee Break
- 11:15 • The Characteristics and Irradiation Capabilities of MARIA Research Reactor in NCBJ Świerk, **G. Krzysztoszek**, NCBJ (Świerk, Poland)
- 11:40 • Plasma Material Interaction in Tokamak: the Contribution of WEST and of Laboratory studies, **C. Grisolia et al.**, CEA Cadarache (France)
- 12:05 • Discussion
- 12:35 Lunch Break

SESSION 2: PRIMARY DAMAGE AND STRUCTURAL DEFECTS, NOV. 4TH, (p.m.)

Chairman: **F. Gao**, University of Michigan (USA) – Co-Chairman: **J.-P. Crocombette**, CEA Saclay (France)

- 14:00 • **Modeling and Simulation of Primary Damage and Structure Evolution in Ceramics and Metals**, **F. Gao**, University of Michigan (Ann Arbor, USA)
- 14:40 • DART, a BCA Code to Assess and Compare Primary Irradiation Damage in Nuclear Materials Submitted to Neutron and Ion Flux, **L. Lunéville et al.**, CEA Saclay (France)
- 15:05 • Polymers under Ionizing Radiations: the Specificity of Swift Heavy Ions, **Y. Ngoni-Ravache et al.**, CEA Saclay (France)
- 15:30 Coffee Break
- 15:55 • Structure of Defects and Microstructure Evolution in Oxide Ceramics – Role of Electronic Excitation and Selective Displacement Damage, **K. Yasuda**, Kyushu University (Fukuoka, Japan)
- 16:20 • Irradiation Induced Phenomena in Nuclear Glass, **J.-M. Delaie et al.**, CEA Marcoule (France)
- 16:45 • Simulation of C-Component Dislocation Loops by Ions and Protons Irradiation. A Tool to Understand the Breakaway Growth of Recrystallized Zr-Based Alloys, **J.-P. Mardon et al.**, AREVA NP (Lyon, France)
- 17:10 Discussion
- 17:40 End of the First Day

18:30 Welcome Cocktail (CEA Cadarache guest house, 18h30 – 20h30)

2nd MINOS workshop program (continued)

Thursday, November 5, 2015

SESSION 3: EVOLUTION & CHARACTERIZATION OF THE MICROSTRUCTURE, NOV. 5TH, (a.m.)

Chairman: M. J. Caturla, University of Alicante (Spain) – Co-Chairman: G. Jomard, CEA Cadarache (France)

- 9:00 • **Microstructure Evolution in Fe and Fe-Cr Alloys with OKMC Methods, M. J. Caturla et al., University of Alicante (Alicante, Spain)**
- 9:40 • Atomistic Modeling of Segregation and Precipitation in Fe-Cr Alloys under Irradiation, **F. Soisson et al.**, CEA Saclay (France)
- 10:05 • Study of Rare Gases Behavior in Uranium Dioxide, **G. Carlot et al.**, CEA Cadarache (France)
- 10:30 Coffee Break
- 10:55 • Positron Annihilation Spectroscopy to Characterize Irradiation Induced Vacancy Type Defects in Materials for Nuclear Fission & Fusion, **M-F. Barthe**, CNRS-CEMHTI (Orléans, France)
- 11:20 • α Self-Irradiation Effects on Structural Properties of (U,Am) $O_{2\pm\delta}$ Materials, **F. Lebreton et al.**, CEA Marcoule (France)
- 11:45 • Discussion
- 12:20 Lunch Break

SESSION 4: IMPACT ON PHYSICAL, CHEMICAL, MECHANICAL CHARACTERISTICS & PROPERTIES, NOV. 5TH, (p.m.)

Chairman: I. M. Robertson, University of Wisconsin-Madison – Co-Chairman: D. Féron, CEA Saclay (France)

- 14:00 • **Dislocation Grain Boundary Interactions in Irradiated Metals, I. M. Robertson et al., University of Wisconsin-Madison (Madison, USA)**
- 14:40 • Effects of Clear Bands on Intergranular Stresses and IASCC Early Damage, **M. Sauzay et al.**, CEA Saclay (France)
- 15:05 • Evolution under Irradiation of Optimized Austenitic Steel for Gen-IV Reactors, **A. Courcelle et al.**, CEA Saclay (France)
- 15:30 • Multiscale Modeling of Radiation Hardening in RPV and Austenitic Stainless steels: from the Atomic to the Continuum Scale, **G. Monnet**, EDF R&D (Moret-sur-Loing, France)
- 15:55 Coffee Break
- 16:20 • High Burn-up Structure in Nuclear Fuel: Impact on Fuel Behavior, **J. Noirot et al.**, CEA Cadarache, (France)
- 16:45 • Influence of Irradiation on Stainless Steel Corrosion in PWR Primary Conditions, **S. Perrin et al.**, CEA Saclay, (France)
- 17:10 • Discussion
- 17:40 Break
- 17:50 • Closing Statements, **F. Carré (DEN/DS)**, CEA Saclay (France)

18:00 End of the Second Day

2nd MINOS workshop program (continued)

Friday, November 6, 2015

JULES HOROWITZ REACTOR AND LECA-STAR LABORATORY VISITS*, NOV. 6TH, (a.m.)

(*): limited participation

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|-------|--|
| 8:00 | • Meeting at the guest house parking and departure (by bus) to the main gate of CEA Cadarache for entrance procedure |
| 8:45 | • First group to visit RJH reactor and LECA-STAR facility |
| 10:45 | • Transfer |
| 11:00 | • Second group to visit RJH reactor and LECA-STAR facility |
| 13:00 | • End of visits, back to the guest house |
| 13:15 | Lunch Break (guest house) |
| 14:00 | • End of MINOS Workshop |

Keynote and lecture speakers

- Serge Bouffard, CEA Saclay (France)
 - Maria José Caturla, University of Alicante (Spain)
 - Fei Gao, University of Michigan (USA)
 - Ian M. Robertson, University of Wisconsin-Madison (USA)
-
- Marie-France Barthe, CNRS-CEMHTI (France)
 - Gilles Bignan, CEA Cadarache, JHR (France)
 - Jean-Marc Delaye, CEA Marcoule (France)
 - Gaelle Carlot, CEA Cadarache (France)
 - Arnaud Courcelle, CEA Saclay (France)
 - Christian Grisolia, CEA Cadarache (France)
 - Grzegorz Krzysztozek, NCBJ (Poland)
 - Florent Lebreton, CEA Marcoule (France)
 - Laurence Lunéville, CEA Saclay (France)
 - Jean-Paul Mardon, AREVA NP (France)
 - Ghiath Monnet, EDF R&D (France)
 - Yvette Ngono, CEA Saclay (France)
 - Jean Noirot, CEA Cadarache (France)
 - Stéphane Perrin, CEA Saclay (France)
 - Maxime Sauzay, (CEA Saclay (France)
 - Frédéric Soisson, CEA Saclay (France)
 - Roger P. Webb, University of Surrey (UK)
 - Kazuhiro Yasuda, Kyushu University (Japan)

Local scientific committee members

- Jean-Luc Béchade, CEA Saclay (France)
- Lucile Beck, CEA Saclay (France)
- Philippe Bossis, CEA Saclay (France)
- Damien Féron, CEA Saclay (France)
- Muriel Ferry, CEA Saclay (France)
- Christian Grisolia, CEA Cadarache (France)
- Gérald Jomard, CEA Cadarache (France)
- Bernard Marini, CEA Saclay (France)
- Maylise Nastar, CEA Saclay (France)
- Jean Noirot, CEA Cadarache (France)
- Sylvain Peugeot, CEA Marcoule (France)
- Sylvie Pillon, CEA Marcoule (France)
- Benoît Tanguy, CEA Saclay (France)
- Carole Valot, CEA Cadarache (France)

Organization

- Christophe Gallé, CEA/MINOS and Corine Farnaud-Pasquier, CEA/Cadarache
- Constantin Meis and Mireille Cestrières, CEA/INSTN, Saclay

Sponsors





MINOS

2nd workshop international
4-6 nov. 2015, (CEA- INSTN, Cadarache)

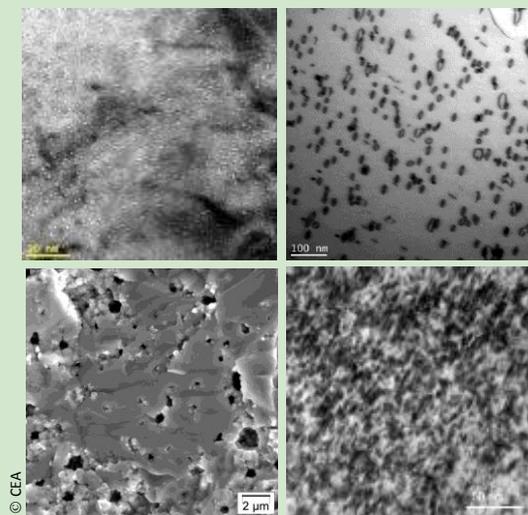




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Main comments and outcomes of session 1: Irradiation Tools & Facilities

Chair: R. P. WEBB, Surrey Ion Beam Centre, University of Surrey (Guilford, UK)

Co-chair: J.-L. BÉCHADE, CEA-DEN-DMN, Service de Recherches de Métallurgie Physique, SRMP (Saclay, France)

From the presentation of S. Bouffard, it arises that thanks to the EMIR platform (six facilities), a large range of irradiation conditions are available in France. The Surrey Ion Beam Centre (R. P. Webb talk) provides complementary facilities mainly devoted to support micro-electronics irradiation damage studies for industry (with an intensive quality insurance and standards program). On the other side, it was underlined that Materials Testing Reactors are also able to provide useful complementary data about irradiation effects with neutrons: MARIA reactor in Poland (NCBJ) till 2030 (G. Krzysztoszek presentation) and RJH (C. Colin and G. Bignan presentation) which is under construction in CEA Cadarache and should be in operation at the end of 2020. For RJH, impressive devices will be proposed to study in service but also incidental and accidental conditions for RPV, internals and claddings of PWR. Finally a good illustration of the need to use modelling approaches and experimental irradiation was illustrated by the west project (C. Grisolia) where tungsten-based material is submitted to high thermal gradient and high flux of hydrogen isotopes.

Finally, main concerns shared by the participants have shown that:

- Access to charged particles irradiation facilities could be complex, Ease of access to European Facilities could be improved. The mixed funding model, even within CEA for example, causes some confusion - some provide free (at the point of use) access, some cost real money. There are also scheduling issues for some when there is only a single call for users each year.;
- It seems that facilities in Europe are enough for what research teams want to investigate although there are higher energy facilities in Japan and USA but it is essential to keep and develop irradiation tools and to reinforce the complementarity between accelerators and research reactors ;
- The question of the representativeness of neutron irradiation using charged particles irradiation is of main concern (especially concerning the flux effect and defects generation, for which different mechanisms can be considered) ;
- Predictive numerical simulation have to integrate more accurately the information related to primary damage ;
- Online characterization and in situ deformation under irradiation considering a corrosive environment is essential (solutions already exist but have to be improved sometimes).

Main comments and outcomes of session 2: Primary Damage & Structural defects

Chair: F. GAO, Dept. of Nucl. Eng. and Radiological Sciences, College of Engineering, University of Michigan (Ann Arbor, Michigan, USA)

Co-chair : J.-P. CROCOMBETTE, CEA-DEN-DMN, Service de Recherches de Métallurgie Physique, SRMP (Saclay, France)

Method to model irradiation damage

Different approaches exist to simulate primary damage state:

- Molecular dynamic, as presented in F. Gao's talk, which is good qualitatively and quantitatively to describe the primary state of damage in terms of structure and repartition of defects but quite heavy and quite limited in energy. It works either with empirical potentials with the weaknesses of such

potentials or, more recently with the use of ab initio simulations, addressing charge transfer and charge-modified migration energy of defects. This might be important for ceramics and glass, but ab initio MD limits nowadays to 1000 atoms.

- The Binary Collision Approximation (BCA) approach, as in L. Lunéville talk. This method is very generic and very good at producing number for the amount of damage; but does not describe the defects created.

The work presented by J.-M. Delaye, exemplified an ad hoc method which relies on completely different principles. Indeed this approach does not model at all the irradiation itself but focuses on its effect on the structure of the material. As it appeared during the discussion, such methodology can work only in specific situations and in cases where experimental results are available to valid the defective, port-irradiation structure.

Effect of electronic losses, their importance and the difficulty to model them

Electronic losses appeared clearly in the talks of K. Yasuda on oxides and Y. Ngonon-Ravache on polymers. They have shown that such electronic effects are of great importance for their materials. There is no surprise these were experimental talks! But in a sense, electronic losses were also somehow there, as a ghost, in F. Gao's talk, as these electronic losses are known to be of importance in SiC material, but we have little way to model them, beyond the charge transfer effects mentioned at the beginning. There was an agreement that we have a lot of work ahead of us on these matters, both experimentally and theoretically, even if the situation is less dramatic than thought by many. Indeed Monte-Carlo codes for the transport of particles (such as GEANT-4) exist which allow a description of the travelling of excited electrons in matter. Moreover two-temperature models are an emerging tool to describe the transfer of energy from the atomic and electronic systems. Nevertheless no formalism exists to evaluate to creation of defects by excited electrons. The description of swift heavy ion damage or fission track damage in ceramics also involves a lot of works ahead of us. Of course, Molecular Dynamics simulations of heat spikes provide an ad hoc method to simulate heavy ion damage or fission track damage in ceramics. The effectiveness and correctness of this approach remains an open question.

Modeling of neutron with ion

This topic was tackled from two different perspectives by L. Lunéville and J.-P. Mardon. This is a very complex topic we had no time to discuss in details. Questions pertaining to this subject include: in which way, the ion irradiation is similar to neutron irradiation. Do we understand the differences between neutron and ion irradiations? How can we model neutron irradiation and ion irradiation?

J.-P. Mardon was very satisfied with his ion irradiation experiments and he said he has gained understanding on what happens with real neutron irradiations.

A few other points were tackled during the discussion:

The importance of a correct description of primary damage was debated as well as the possibility to effectively measure it. Some thought it is impossible to measure while model laser experiments were suggested by others, as well as experiments with advanced proton sources. The accuracy and utility of the NRT dpa was also discussed in connection with the on-going thinking of the EAN and IAEA on that subject. The fact the dpa was a measure of damage not defects and that the number of defects is roughly one third of what is predicted by NRT was stressed. Finally the importance of measuring and calculating the sputtering due to fission fragments in nuclear fuels was stressed. This sputtering comes mainly from electronic losses of high energy fission fragments and is due to the heating along the fission track.

Main comments and outcomes of session 3: Evolution & Characterization of the Microstructure

Chair: M. J. CATURLA, Dept. Física Aplicada, Facultad de Ciencias, Fase II, University of Alicante (Alicante, Spain)

Co-chair: G. JOMARD, CEA-DEN-DEC, Service d'Etudes et de Simulation du Comportement des Combustibles, SESC (Cadarache, France)

The main topic that has been addressed was related to the link between modelling and experiments. It is clear that both approaches must take benefit from a strong coupling (not only comparison) between experimentalists and modelers. Validation of models is not always straightforward since there are only rare cases where calculations results and experimental observations are directly comparable: Positron lifetime in defected materials and XAS spectra are two of these. Usually experiments and simulations are not performed at the same size or time scales which complicates the comparison.

Often the interpretation of the experiments requires some modeling. This is the case of positron annihilation experiments where accurate DFT calculations of lifetimes are very useful in the interpretation of the results. In the case of TEM, quantification of the results is complex since it depends on the resolution limit, on the families of loops that can or cannot be observed for a given orientation or even the distance of defects to the surface. In this case maybe TEM image simulations could help in the quantification of the results. Some other methods were also pointed out such as XAFS or XDR which would need data from simulations. It was pointed out that both in the US and in Europe there are some developments towards sharing raw data between scientists, although it is not clear how this will be done in practice. Each community however agrees that the interaction between experiments and simulations must be strengthened.

The issue of the quantification of damage induced by irradiation has been addressed. It seems reasonable to treat dpa for what it is: a measure of the energy deposited in a sample during irradiation instead of a measure of an amount of defects. However, there is still a debate about if this value is being used properly in different contexts.

Among quantities that can be obtained from experiments and which will be very valuable for modelers, the threshold energy for U and O displacement in UO₂ has been identified as a key parameter that needs to be measured.

Main comments and outcomes of session 4: Impact on Physical, Chemical, Mechanical Characteristics & Properties

Chair: I. M. ROBERTSON, Dept. of Materials Science and Engineering, University of Wisconsin-Madison, (Madison, Wisconsin, USA)

Co-chair: D. FÉRON, CEA-DEN-DPC, Service de la Corrosion et du Comportement des Matériaux dans leur Environnement, SCCME (Saclay, France)

The first talk in this session by I. Robertson et al., described experimental observations of dislocations interacting with grain boundaries in un-irradiated and ion-irradiated FCC metals. It was demonstrated that the slip system activated to transfer slip across a grain boundary, at least at low level of strains, was determined by the magnitude of the Burgers vector of the dislocations left in the grain boundary as a result of the transmission event. It was claimed that the local resolved shear stress on the selected slip system needs to be sufficient to move dislocations from the grain

boundary source into the grain. This makes the magnitude of the local resolved shear stress important in irradiated alloys as it must be sufficient to move dislocations through the field of irradiation-produced obstacles. An outstanding question to be answered is does the slip transfer process across grain boundaries change as the local strain is increased.

The second talk by M. Sauzay et al., provided state-of-art simulations and modeling of the impact of the slip bands, cleared channels, impacting a grain boundary and used a critical normal stress and fracture energy criteria to deduce the conditions under which the grain boundary would fail. Important advances in the formulation of the problem are the consideration of the width of the channel, which relates to having dislocations available on not one but multiple parallel and closely spaced slip systems. The cohesive strength of the grain boundary was assumed to change in the near surface region due to oxidation. An interesting question to consider is can this method be used to explain why only select slip band intersections of the grain boundary result in cracking of the grain boundary.

The third talk by A. Courcelle et al., demonstrated that a cold-worked Ti-stabilized austenitic stainless steel exhibits a high resistance to void swelling even at high doses. A comparison of the precipitate interactions with vacancy clusters showed differences between neutron and ion irradiated materials. This work demonstrated the need for further understanding the precipitation so that it may be accounted for appropriately in models.

The fourth talk by G. Monnet described the complex problem of building a multi-scale model of radiation-induced hardening was presented. It demonstrated the advances that have been made as well as those needed to advance the field further. For example, a challenge is to identify the dominant processes responsible for a particular effect, e.g., the mechanisms of annihilating the radiation-produced damage as it is known that the annihilation event does not produce an easy glide path, which raises the question of the nature of the remnant damage. Also, as indicated by the talk by M. Sauzay, the cleared channels involve activity on closely spaced parallel planes within a confined volume. An interesting question is how to move from isolated behaviors and interactions, which are well understood, to the more complex and important ones involving the collective behavior or summation of all possible interactions and use this information to bridge between length scales.

The fifth talk by J. Noirot et al., described the impact of high burn up on fuel behavior. Significant changes in the microstructure, grain size reduction, larger bubbles and precipitate, were shown to occur in the high burn-up region. It was suggested that the differences that have been reported may be due to the different levels of impurities in each of the systems.

The last talk by S. Perrin et al., was dedicated to the influence of irradiation on stainless steel corrosion in PWR primary conditions. A focus was made on damages due to IASCC in austenitic stainless steel 316L in order to understand the role of defects on oxidation process. Oxide layer characterizations (oxide layer properties, oxide kinetic, oxygen transport) were performed on non-implanted and implanted samples with xenon. This study shown that, the defects created by xenon implantation have an effect on the oxide layer thickness and composition, on the internal layer growth kinetic and on the oxygen diffusion in this layer.

Key points from the discussion:

1 - Multi-scale modelling and simulation efforts that are coupled directly to a multi-scale experimental efforts are needed. The complexity of the irradiation damage produced and the interaction of dislocations either singly or collectively with the radiation-produced damage presents a major challenge in the development of a multi-scale model that links material performance to atomic- and meso-scale events that can occur over disparate timescales. Despite the advances that have been made there remains a significant gap in the spatial and temporal scales accessible to both approaches. Continued efforts to reduce the gap are needed or, alternatively, the problem being considered must be carefully framed to allow for the synergistic coupling of the two approaches.

On the experimental side, what is needed are in-situ in-operandi experimental methods that bridge the appropriate spatial and temporal scales that will provide new insight into the governing processes

and mechanisms. For example, the closed loop system with the in-situ analytical tools that is being construction should provide new insight to the governing mechanisms of corrosion. Coupling this experimental capability with advances in computational methods provides a unique opportunity to address this critical problem.

On the computational side, it is necessary to continue to improve the algorithms to capture the new physics and chemistry that will permit studies of performance of materials in environments that simulate actual operational conditions. Also, needed are advances in the potentials used in simulations such that they more accurately reflect and actual material, e.g., magnetic effects and native oxides.

2 - Data-enabled and –driven science and engineering. The nuclear materials community has over the decades generated a significant amount of data and knowledge, much of which will be lost with the generational change in nuclear material scientists and engineers. There is a unique opportunity to create searchable data bases that will allow new interrogation approaches to be employed to look for hidden synergies and effects. The value of moving in this direction was debated with no consensus being reached. The discussion did raise the issue about gathering and storage of data that would be meaningful to others.

Other challenges were also mentioned:

- From atomic to the continuum scale, modelling and characterization of damages need to be improved on RPV and stainless steels, on new cladding alloys, on fuels... ;
- While modelling and observation scales in terms of geometrical dimensions are quite in accordance, it is not at all the case in terms of time. The accumulation of irradiation or environmental damages is over years while atomistic modelling is often much less than a second. The resolution of this dilemma may lead to the evaluation of incubation and initiation periods of damage phenomena ;
- Irradiation is one of the parameters among other ones coming from the environment. It is not only multiscale phenomena which are studied in geometrical and time scale, but also multi-parametric processes.