STUDIECENTRUM VOOR KERNENERGIE
CENTRE D'ETUDE DE L'ENERGIE NUCLEAIRE

BOOK OF ABSTRACTS
SCK•CEN-BA-27

Topical day

Fission Gas Release Modelling

Mol, 14 September 2009

SCK•CEN
Boeretang 200
BE-2400 MOL
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http://www.sckcen.be

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This topical day is sponsored by

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Context and objective

Fission gas release is one of the most limiting phenomena in view of the present and near-coming fuel's performances (in the framework of Gen II and Gen III). Its effects are potentially enhanced by the tendency for higher burn-ups and for higher Pu enrichments. Fission gas release will also be an important issue for some reactor concepts considered for the future Gen IV nuclear reactors.

This Topical Day aims at bringing together modellers working at different scales, from ab-initio calculations to integral fuel performance modelling. The objective is to link the efforts made at the different scales (atomic-scale modelling, meso-scale modelling and experiments, integral FGR), by discussing the progress and the shortcomings at the different levels, and by reflecting on their interactions.

The ultimate goal of this Topical Day is to identify the type of data that are still expected by the different groups of modellers, and to discover potential contributions to the improvement of models at other scales.

Programme

	Chairman: D. Parfitt (IC London) – K. Govers (SCK•CEN)
09.15 – 9.40	*Registration and coffee*
09.40 – 09.55	**L. Sannen** - **SCK•CEN** Welcome
09.55 – 10.25	**M. Freyss – CEA Cadarache** Ab initio study of the stability of point defects and volatile elements in uranium carbide
10.25 – 10.55	**L. Van Brutzel – CEA Saclay** Molecular Dynamics Study of Small Xenon Clusters in Urania
10.55– 11.15	*Coffee break*
11.15 – 11.45	**C. Bishop – Imperial College London** Fission gas and radiation damage in uranium dioxide
11.45 – 12.15	**K. Govers – SCK•CEN** MD simulation of thermal spike interactions with fission gas bubbles
12.15 – 13.30	*Sandwich lunch*
	Chairman: S. Lemehov (SCK•CEN) – F. Jutier (SCK•CEN)
13.30 – 14.00	**N. Nakae – JNES** Kinetics of Gaseous Atoms in Particular Field
14.00 – 14.30	**M. Verwerft – SCK•CEN** Local analysis of retained fission gas in nuclear fuel: onset of clustering and release to the free volume
14.30 – 15.00	*Coffee break*
15.00 – 15.30	**S. Lemehov – SCK•CEN** Modelling radial power and burnup distributions in high burnup LWR fuel and MOX
15.30 – 16.00	**F. Jutier – SCK•CEN** Post-Irradiation Examination of high-enriched MIMAS MOX: confrontation with modelling results
16.00 – 18.00	*Closure and drink*

Ab initio study of the stability of point defects and volatile elements in uranium carbide

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Abstract

The scope of this study is to shed light on the behaviour of point defects and volatile impurities in uranium carbide, as a first step in the study of the potential Gen IV nuclear fuel (U,Pu)C. The study of point defects and volatile impurities is central in order to get some comprehension in the behaviour of the materials under irradiation. We calculate and compare the formation energies of the various types of point defects that can be found in UC. Vacancies, interstitials, dumbbells, Frenkel pairs, anti-site defects and small vacancy clusters are considered. Then, the behaviour in UC of the volatile fission products Kr, Xe and I, as well as O which could be incorporated by oxidation, is studied. The most stable location of these elements in the lattice is determined and their incorporation and migration energies are compared.

All calculations are done using the *ab initio* PAW method (Projector Augmented Waves) based on the DFT as implemented in the VASP code. Exchange-correlation interactions are taken into account within the GGA approximation. The use of the GGA+U approximation will be discussed.

A preliminary study on the incorporation of rare gas atoms in small molecules has been performed to assess the accuracy of the DFT for the description of bonds formed by rare gases. Results obtained using various functionals and a post-Hartree method are compared.

Keywords: point defects, impurities, carbides

Molecular Dynamics Study of Small Xenon Clusters in Urania

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Abstract

Molecular dynamics simulations using empirical potential have been used to study the stability and the formation of xenon clusters in several extended defects such as grain boundaries and nanocavities up to 30 Shottky defects in urania.

New interatomic potentials have been fitted for this study. The potentials reproduce the xenon insertion energies in the fluorite interstitial sites and in the oxygen and uranium vacancies calculated by DFT *ab-initio* simulations.

Static calculations show that xenon atoms are more likely to aggregate than staying homogeneously distributed. Moreover, the lowest insertion energy is found for xenon atoms in grain boundary ($\Sigma 5$). The binding energy of different nanocavity shapes changes with the presence or not of xenon atoms. With xenon atoms, the spherical nanocavities are energetically more stable than any other nanocavities investigated in this study.

Dynamics calculations confirm these results. When randomly distributed, the xenon atoms aggregate rapidly in small cluster along the (111) plane creating several point defects in the urania. Spherical xenon bubbles (5-6 nm) with different xenon densities remain stable with no diffusion of xenon atoms into the surrounding urania matrix. For the highest xenon density studied (0.12 mol/cm^3), the xenon crystallizes into fcc structure and small dislocation loops appear in the urania matrix.

Fission gas and radiation damage in uranium dioxide

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Abstract

Cascade events have been simulated in uranium dioxide. As the energy of the PKA increases, the resulting damage becomes more isotropic with respect to the initial direction of the PKA and more disruption to the uranium lattice is observed. The disordered region may act as a nucleation site for fission gas products. Bubble nucleation is investigated using molecular dynamics techniques.

MD simulation of thermal spike interactions with fission gas bubbles

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Abstract

The fission process results in the emission of generally two energetic "fission fragments". These fragments progressively slow down through collisions within the matrix, creating a heavily disturbed zone, often referred as the "thermal spike". A simplified description of thermal spikes using molecular dynamics techniques has already illustrated different phenomena accompanying thermal spikes: the emission of shock-waves at the early beginning of the event, the progression of the molten zone and its re-solidification and the formation of dislocation loops.

In the present study, focus has been given on the interactions of thermal spikes with fission gas bubbles in order to investigate their possible destruction/nucleation. Preliminary results show that the simplified description of thermal spikes used, that do not model the initial ballistic phase, is not sufficient to re-solve all the gas present in the bubble into the matrix.

This observation emphasizes the role of the initial ballistic phase in the re-solution process. Further investigations will enable to better characterize the re-solution as well as the bubble nucleation process.

Kinetics of Gaseous Atoms in Particular Field

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Abstract

Kinetic of gaseous atoms is generally treated in the field of academics such as statistical mechanics and/or statistical thermodynamics established by Ludwig Boltzmann. The movement of only gaseous atoms is considered and heat capacity, entropy and enthalpy of gaseous atom are derived and discussed. The kinetics of fission gas atoms in nuclear fuel is, however, much different from those discussed in these academics fields. The movement of gaseous atoms in solids shall be basically considered also the field of movement should have unique microstructure such as pore and grain boundary, also especially plutonium enriched zone in the case of Plutonium and Uranium Mixed Oxide (MOX). When the fission gas behavior is discussed, the movement in the particular field shall be taken into account.

The mechanism of fission gas release has been studied by many researchers for these nearly fifty years since Booth first set down the simple approximations to fission gas release from irradiated UO2. However, there is still no general agreement what mechanisms are responsible for observed coalescence of intergranular bubbles during their growth. It might be due to the complexity of the kinetics of gaseous atoms in particular field which corresponds to nuclear fuel pellet having unique microstructure, temperature gradient and irradiation damage.

The objectives of this paper are to introduce the brief review of fundamental process of the kinetics in particular field and fission gas release mechanism in general, and to introduce the bond percolation model for the coalescence and growth of intergranular bubbles and effective fission gas release. The fission gas release mechanism for MOX fuel which has heterogeneous microstructure including plutonium enriched zone (Pu spot) is also introduced.

Though it is indeed extraordinary to develop mesoscopic and/or atomic scale modelling for kinetics of fission gas atom and fission gas release, we shall challenge to do so and find out the mechanisms which are responsible for observed coalescence of intergranular bubbles during their growth.

Local analysis of retained fission gas in nuclear fuel: onset of clusteringand release to the free volume

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Abstract

The accurate analysis of locally retained fission gas in nuclear fuel is inherently difficult since the physical form under which it is stored varies from atomically dispersed to bubbles with a diameter of several hundreds of nanometers. One of the techniques that has been applied since more than twenty years is Electron Probe Microanalysis (EPMA). This technique however is difficult to apply in a quantitative manner if the studied materials are inhomogeneous at the scale of the electron-solid interaction volume. In this presentation, a method is presented to analyse a system of gas bubbles distributed in a solid matrix. It is based on the geometric modelling of a gas bubble dispersion and the assessment of its influence on the emitted X-ray intensity. The resulting, more accurate analysis of gas retained in nuclear fuel may lead to a better insight in the gas release mechanisms.

Modelling radial power and burnup distributions in high burnup LWR fuel and MOX

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Abstract

This presentation discuss an approach to model radial distributions in high burnup LWR fuels and in mixed oxides with elevated content of plutonium. In such fuels highly absorbing thermal and resonance neutrons isotopes of plutonium (238 to 243) play an important role in forming and evolution of non-uniform power and burnup profiles. Energy deposition and energy release per fission event are also sensitive to actual fuel composition.

Several models have been developed for the full scale fuel performance code MACROS to cope with non-uniform energy release and deposit as for prediction fission rate density across the fuel radius and radial distributions of fission products. Developed models start with considerations of neutronic effects in heterogeneous fuel lattices using one-cell approach to model absorption of resonance neutrons and build-up/burning of plutonium isotopes. Neutronic models provide MACROS code with inputs that then are used to model evolution of gaseous and volatile fission products due to processes of diffusion and irradiation-induced transport. Combined consideration of neutronic, depletion, transport and diffusion problems ultimately gives output distributions of Xe, Cs and other key fission products that can be directly compared with EPMA and SIMS measurements. However, direct comparison with PIE is not conclusive because of limitations in measurement technique.

Neutronic code SCALE is being used since recently to provide independent source of validation data that MACROS needs for better prediction of radial effects.

Examples of MACROS and SCALE applications for analysis of MOX and high burnup UO2 fuels are shown and discussed in context of its importance for better understanding and prediction of local and rod integral Fission Gas Release.

Post-Irradiation Examination of high-enriched MIMAS MOX: confrontation with modelling results

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Abstract

In the frame of the Characterisation of HIgh enriched Plutonium rodS (CHIPS) programme performed between the Japanese Nuclear Energy Safety organization (JNES) and the StudieCentrum voor Kernergie / Centre d'Etude Nucléaire SCK•CEN, the irradiation behaviour of MOX fuel rods having a high initial Pu enrichement of nearly 14% is investigated.
The MOX fuel rods have been fabricated following the MIcronised MAsterblend (MIMAS) route by Belgonucléaire S.A. in 1985. They were then irradiated in the last irradiation cycle of the Belgian Reactor-3 (BR-3) and in the CALLISTO loop of the Belgian Reactor-2 (BR-2) which reproduces the irradiaton conditions of a Pressurized Water Reactor.
In this presentation, Post-Irradiation Examination results from Electron Probe MicroAnalysis (EPMA), radiochemical analyses and Fission Gas measurement will be confronted to the modelling results of the codes SCALE (Standardized Computer Analyses for Licensing Evaluation) package 6 and the MACROS code developped by S. Lemehov.

Biographies

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Since 2001 : researcher at CEA Cadarache, in the Fuel Study Department, in charge of first-principles modeling of nuclear fuels, in particular of the behaviour of point defects and fission products.

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I did my Msci degree at University College London in Chemistry with Mathematics during which time I spent a year in the Davy Faraday Research Laboratory at the Royal Institution of Great Britain where I modelled ice using molecular dynamics. I undertook my PhD studies at University College London where I modelled the filling of carbon nanotubes with molten salts using molecular dynamics, under the supervision of Mark Wilson in the Chemistry department. In February this year I joined the Grimes group at Imperial College London as part of the Fbridge project where I have been modelling uranium dioxide using atomistic simulation techniques.

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Kevin Govers did a M. Sc. in Physical Engineering at the Free University of Brussels. Then he had the opportunity to start a PhD at SCK•CEN (in collaboration with ULB) on atomic-scale simulations of the nuclear fuel, using molecular dynamics techniques. The objective was to investigate the behaviour of noble gases in the UO_2 matrix. Now he is still at SCK•CEN, working in the fuel material group. A part of his work is devoted to atomic-scale modeling, through participation to the EFP-7 F-Bridge project. Other activities are related to general fuel behaviour at larger scales.

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Dr. Nakae currently works for the Japan Nuclear Energy Safety Organization (JNES), an Incorporated Administrative Agency, where he holds the position of Senior Officer in the Reactor Core and Fuel Reliability Evaluation Group of the Safety Standard Division.

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Dr. Marc Verwerft works at SCK•CEN since 1994. Marc graduated in Physics from the University of Antwerp in 1987; obtained his PhD degree on "Modulated Phases in Ceramics" in 1991. In 1992-1994 he worked as post-doctoral researcher and lecturer at the Applied Physics Department of the University of Groningen. At SCK•CEN, he heads today the nuclear fuel materials research group.

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Obtained my engineer diploma at Moscow Engineering Physics Institute (Technical University, MIPhI) with a specialty "Theoretical Nuclear Physics" in 1981. Since then I worked in the Russian Nuclear Research Centre "Kurchatov Institute" till 1995 where I modelled civil and military nuclear fuels. I undertook my PhD studies at MIPhI (1991) where I modelled the processes of in-pile densification and sintering of different ceramic compounds. Since 1995 I worked in international centers, in particular, in the OECD Halden Reactor Project, the University of Tokyo (associate professor), Japan Atomic Energy Research Institute and since 2001 at SCK•CEN where perform full scale modeling high burnup LWR fuels and mixed oxide fuels with plutonium and higher actinides.

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