

NuFuel-MMSNF Programme and Abstracts

NuFuel & MMSNF 2015

First Workshop on Research into Nuclear Fuel in Europe
and Materials Modeling and Simulation for Nuclear Fuels Workshop
Karlsruhe, Germany, November 16th to 18th, 2015

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NuFuel-MMSNF Programme and Abstracts

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Programme

Monday, November 16th

9.00 Welcome by JRC-ITU

9.10 Welcome by meeting chairs. Why Nufuel and aim of combined MMSNF-Nufuel

9.20 Presentation of the Joint Program of Nuclear Materials of the European Energy Research Alliance

9.30–12.10 First oral session: fuel synthesis and characterization (Session chairs: Marco Cologna and Elio D'Agata)

- 9.30 Talk 1.1, Vaclav. **Tyrpekl**, ITU (Germany),
[Preparation and characterization of nanostructured UO₂ for LWR fuel safety assessment](#)
- 9.50 Talk 1.2, Kyle **Johnson**, KTH (Sweden),
[Ternary Phase Characterisation within the U-N-Si system](#)

- 10.10 Talk 1.3, Elizabeth **Sooby**, Los Alamos National Laboratory (USA),
Synthesis and Oxidation Testing of U-Si-Al Ternary Compounds
- 10.30 Break
- 11.00 Talk 1.4, Laura **Martel**, ITU (Germany),
High Resolution Solid-State NMR of Actinides-bearing compounds
- 11.20 Talk 1.5, Damien **Prieur**, ITU (Germany),
(U,M)O_{2±x} solid solution: a RT XANES and HT XANES study
- 11.40 Session wrap-up

12.10 Lunch

14.00–16.10 Second oral session: irradiations and post-irradiation examinations.
(Session chairs: Fabienne Delage, Joe Somers)

- 14.00 Talk 2.1, Mélanie **Chollet**, PSI (Switzerland),
Micro X-ray diffraction analysis of a fully recrystallized irradiated UO₂ fuel
- 14.20 Talk 2.2, Fabiola **Cappia**, ITU (Germany),
Investigation of mechanical and local properties of high burnup (>60 GWd/tHM) UO₂ fuel with focus on the pellet rim
- 14.40 Talk 2.3, Elio **D’Agata**, JRC (Netherlands),
Outcomes from the implementation of irradiation tests on minor actinides bearing oxide fuels
- 15.00 Talk 2.4, Sander van **Til**, NRG (Netherlands),
The SPHERE irradiation experiment: comparison of irradiation performance of pellets and spherepac MA-bearing driver fuel
- 15.20 Talk 2.5, D. **Staicu**, ITU (Germany),
New Catalog on (U,Pu)O₂ Properties For Fast Reactors: Work Plan Of The ESNII+ Project and Perspectives
- 15.40 Session wrap up
- 16.10 Break

16.30–19.00 Poster session

Tuesday, November 17th

9.00–13.00 Third oral session: Separate effect experiments. (Session chairs: Dragos Staicu and Ken McClellan)

- 9.00 Talk 3.1, Claire **Onofri**, CEA-Cadarache (France),
Separated effect studies: a necessary experimental approach to understand the fuel behavior under in-pile irradiation

- 9.20 Talk 3.2, Mohamed **Naji**, ITU (Germany),
On the Raman Spectrum of Plutonium dioxide: vibrational and crystal electric field modes
- 9.40 Talk 3.3, Yue **Ma**, CEA-Cadarache (France),
The study of the $\text{UO}_{(2+x)}$ crystalline structure with high-resolution neutron diffraction experiment
- 10.00 Talk 3.4, Eric **Gilbert**, CENBG/Université de Bordeaux (France),
Air storage effect for Kr and Xe transport in UO_2
- 10.20 Talk 3.5, Oliver **Dieste**, ITU (Germany),
Long term structural stability of severely alpha-damaged fuels
- 10.40 Break
- 11.10 Talk 3.6, René **Bès**, Aalto University (Finland),
Valence states and local structure of Gd doped actinide oxides ($\text{U}_{1-x}\text{Gd}_x$) O_2 and ($\text{Th}_{1-x}\text{Gd}_x$) O_2 measured by X-ray Absorption Spectrometry on the MARS beamline.
- 11.30 Talk 3.7, Alice **Seibert**, ITU (Germany),
Thermal Conductivity of the Uranium-Zirconium Oxide
- 11.50 Talk 3.8, Emtethal **Kassim**, ITU (Germany),
Assessment of the Phase Equilibrium in the ThO_2 - ZrO_2 System
- 12.10 Talk 3.9, Fidelma Giulia Di **Lemma**, JAEA (Japan),
Fission products chemisorption mechanism following severe accidents: a separate effect study on CsOH reaction with stainless steel
- 12.30 Session wrap-up

13.00 Lunch

14.30–17.30 Fourth oral session: Thermodynamic modelling. (Session chair: Christine Guéneau, Rudy Konings)

- 14.30 Talk 4.1, Theodore M. **Besmann**, University of South Carolina (USA),
Thermodynamic Evaluation of a Matrix of High Density, Composite, and Accident Tolerant Fuels
- 14.50 Talk 4.2, Tsvetoslav **Pavlov**, ITU (Germany),
Inverse method for calculating thermo- physical properties of nuclear materials up to their melting point
- 15.10 Talk 4.3, Enrica **Epifano**, CEA-Marcoule (France),
In-situ high temperature X-ray diffraction study of Americium Dioxide
- 15.30 Talk 4.4, Anna L. **Smith**, Delft University of Technology (Netherlands),
Thermodynamic assessment of the neptunium-oxygen system: mass spectrometric studies and thermodynamic modelling
- 15.50 Break
- 16.20 Talk 4.5, Emily **Corcoran**, Royal Military College (Canada),

Thermodynamic Investigations of the Uranium-Molybdenum-Oxygen System by a Coupling of Density Functional Theory and Calphad Methodologies

- 16.40 Talk 4.6, Carl-Magnus **Arvhult**, KTH (Sweden),
Thermodynamic modeling of the Ni-Te and Fe-Te intermetallic phases

16.40–17.10 Session wrap-up

19.00 [Workshop Dinner](#)

Wednesday, November 18th

9.00–13.00 Fifth oral session: Modelling from the atomic to the mesoscopic scales. (Session chairs: Michael Rushton and Marjorie Bertolus)

- 9.00 Talk 5.1, Michael **Cooper**, Los Alamos National Laboratory (USA),
[Simulation and Measurement of Thermal Conductivity in UO₂](#)
- 9.20 Talk 5.2, Laurent Van **Brutzel**, CEA-Saclay (France),
[Mechanical behaviour of UO₂ under irradiation: a molecular dynamics study](#)
- 9.20 Talk 5.3, Paul C. M. **Fossati**, Imperial College London (UK),
[Xe diffusion and bubble nucleation around edge dislocations in UO₂](#)
- 9.40 Talk 5.4, David **Andersson**, Los Alamos National Laboratory (USA),
[Simulation of fission gas diffusion in UO₂ nuclear fuel](#)
- 10.00 Talk 5.5, Zhexi **Guo**, PSI (Switzerland),
[Atomic-scale effects of chromium-doping on defect behaviour in uranium dioxide fuel](#)
- 10.20 Break
- 10.50 Talk 5.6, Conor **Galvin**, Imperial College London (UK),
[Modelling the thermomechanical properties of actinide oxides](#)
- 11.10 Talk 5.7, Michel **Freyss**, CEA-Cadarache (France),
[First-principles modelling of the structural and electronic modifications induced by point defects in actinide mixed oxides](#)
- 11.30 Talk 5.8, Daniel **Griffin**, NNL (UK),
[Calculation of diffusion coefficients in ceramic oxide fuels using adaptive kinetic Monte Carlo modelling](#)
- 11.50 Talk 5.9, Antoine **Claisse**, KTH (Sweden),
[Ab initio determination of gas atoms migration energy in UN](#)

12.10–12.40 Session wrap-up

12.30 Lunch

14.00–17.00 Sixth oral session: Fuel performance codes. (Session chairs: Paul Van Uffelen and Lelio Luzzi)

- 14.00 Talk 6.1, Thomas **Helfer**, CEA-Cadarache (France),
Recent improvements of the fuel thermomechanical modelling in the PLEIADES Platform to better simulate accidental transients conditions using the Alcyone fuel performance code
- 14.20 Talk 6.2, Mark **Wenman**, Imperial College London (UK),
Explicit fracture models of nuclear fuel pellets using peridynamics
- 14.40 Talk 6.3, Davide **Pizzocri**, ITU (Germany),
A semi-empirical model for the formation of the high burnup structure
- 15.00 Talk 6.4, Thomas **Haynes**, Imperial College London (UK),
Modelling Pellet Relocation in PELICAN, a Finite Element Model for Pellet-Clad Interaction in AGR & PWR Fuel
- 15.20 Talk 6.5, Michael J. **Welland**, Canadian Nuclear Laboratories (Canada),
Simulations and Experiments of U-Mo/Al and U-Mo/Mg Research Reactor Fuels
- 15.40 Coffee break
- 16.10 Talk 6.6, Sergei **Lemehov**, SCK • CEN (Belgium),
Modeling and simulation of fast reactor minor actinide bearing oxide fuels in support of MABB and MADF concepts
- 16.30 Session wrap up

17.00 Conclusions of the workshop

- 17.00 Meeting Chairs, General Conclusions

17.15 End of the meeting

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- Carl-Magnus **Arvhult**, KTH (Sweden) *Thermodynamic modeling of the Ni-Te and Fe-Te intermetallic phases* (Tuesday, November 17th, 16.40)
- Theodore M. **Besmann**, University of South Carolina (USA) *Thermodynamic Evaluation of a Matrix of High Density, Composite, and Accident Tolerant Fuels* (Tuesday, November 17th, 14.30)
- Laurent Van **Brutzel**, CEA-Saclay (France) *Mechanical behaviour of UO₂ under irradiation: a molecular dynamics study* (Wednesday, November 18th, 9.20)

- René **Bès**, Aalto University (Finland) Valence states and local structure of Gd doped actinide oxides ($U_{1-x}Gd_x$)O₂ and (Th_{1-x}Gd_x)O₂ measured by X-ray Absorption Spectrometry on the MARS beamline. (Tuesday, November 17th ,11.10)
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- Elizabeth **Sooby**, Los Alamos National Laboratory (USA) Synthesis and Oxidation Testing of U-Si-Al Ternary Compounds (Monday, November 16th, 10.10)
- D. **Staicu**, ITU (Germany) New Catalog on (U,Pu)O₂ Properties For Fast Reactors: Work Plan Of The ESNII+ Project and Perspectives (Monday, November 16th, 15.20)
- Sander van **Til**, NRG (Netherlands) The SPHERE irradiation experiment: comparison of irradiation performance of pellets and spherepac MA-bearing driver fuel (Monday, November 16th, 15.00)
- Vaclav. **Tyrpekl**, ITU (Germany) Preparation and characterization of

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- Michael J. **Welland**, Canadian Nuclear Laboratories (Canada) Simulations and Experiments of U-Mo/Al and U-Mo/Mg Research Reactor Fuels (Wednesday, November 18th, 15.20)
 - Mark **Wenman**, Imperial College London (UK) Explicit fracture models of nuclear fuel pellets using peridynamics (Wednesday, November 18th, 14.20)
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- Poster 5.2: Marjorie **Bertolus**, CEA-Cadarache (France), [Positron annihilation spectroscopy study in UO₂: coupled theoretical and experimental approach](#)
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Talk Abstracts

Talk 1.1: Preparation and characterization of nanostructured UO₂ for LWR fuel safety assessment

Vaclav. Tyrpekl, M. Cologna, J. Somers

- European Commission, Joint Research Centre, Institute for Transuranium Elements (ITU), Hermann-von-Helmholtz Platz 1, PO Box 2340, DE-76125 Karlsruhe, Germany

Abstract

During its lifetime in a nuclear power plant the fuel (UO_2) undergoes significant restructuring at the periphery of the pellet, with the grains (10 μm or more) of the fresh fuel converting to a ‘cauliflower’ like structure with grain size of 100–300 nm (20% porosity). Despite the investigations made thus far on this structure, their scope is severely limited due to the high radioactivity of the irradiated fuel. To circumvent this limitation we have developed a process to produce UO_2 pellets with nano-crystalline structure to enable a series of separate effect investigations on surrogate material (mechanical properties, gas retention, etc.) and a deeper understanding of this material and its safety assessment. This paper concentrates on the preparation of the nanostructured UO_2 material, starting from ex oxalate and sol gel powders, consolidation using spark plasma sintering (SPS), characterisation and mechanical property determination.

Talk 1.2: Ternary Phase Characterisation within the U-N-Si system

Kyle Johnson¹, Alicia Marie Raftery¹

- ¹: KTH Royal Institute of Technology, Stockholm, Sweden

Abstract

Research into Accident Tolerant Fuels (ATF) in the wake of the Fukushima Dai-ichi Nuclear Disaster has precipitated interest in a diverse array of fuels and fuel combinations with favorable properties for accident tolerance. One such combination is the UN- U_3Si_2 system, which is purported to have favourable characteristics regarding thermal conductivity and water tolerance. In this paper, a ternary phase constituted by U-N-Si formed during SPS sintering is identified and characterized.

Talk 1.3: Synthesis and Oxidation Testing of U-Si-Al Ternary Compounds

Elizabeth Sooby, J. T. White, D. D. Byler, A. T. Nelson, and K. J. McClellan

- Materials Science and Technology Division, Los Alamos National Laboratory, Los Alamos, NM USA

Abstract

Uranium silicide intermetallic compounds are currently under investigation by the DOE-NE Advanced Fuels Campaign as candidate accident tolerant nuclear fuel concepts. While some compounds in this system offer benefits compared to reference UO_2 , they have been shown to display poor oxidation resistance and pellet cracking. It is hypothesized that introducing an additional compositional degree of freedom by exploring U-Si-X (X=Al or Ti) ternary systems may provide the means to design a candidate nuclear fuel which optimizes the benefits of each binary. Reported here are the fuel synthesis techniques and metallurgical processing of these U-Si-Al ternaries. In addition the preliminary oxidation results of a series of ternary compounds, beginning with the known $\text{U}_3\text{Al}_2\text{Si}_3$ system will be presented.

Talk 1.4: High Resolution Solid-State NMR of Actinides-bearing compounds

Laura Martel, Chris Selfslag, Joseph Somers

- European Commission, Joint Research Centre, Institute for Transuranium Elements (ITU), Hermann-von-Helmholtz Platz 1, PO Box 2340, DE-76125 Karlsruhe, Germany

Abstract

Magic angle spinning nuclear magnetic resonance (MAS-NMR) is a very powerful analytical technique for the study of the local structure in materials. However, the analysis of actinide-bearing compounds and their $5f$ -unpaired electrons remained for a long time elusive mainly because of their well-known high radiotoxicity. Recently, the Joint Research Centre Institute for Transuranium Elements (JRC-ITU) in Karlsruhe (Germany) acquired a 9.4T commercial NMR spectrometer dedicated for the study of actinides-bearing compounds. It has been integrated with a radioactive glovebox and a 1.3 mm commercial probe (rotation rates up to 70 kHz). It has been used on a broad range of materials including fuels, glasses and ceramics for nuclear waste disposal. The presentation will concentrate on applications in the fuel domain.

Talk 1.5: (U,M)O_{2±x} solid solution: a RT XANES and HT XANES study

Damien Prieur¹, Philippe Martin², Enrica Epihano², René Bès^{2,3}, Laura Martel¹, Jean-francois Vigier¹, Daniel Neuville⁴, Tim Prussmann⁵, Tonya Vitova⁵, Kristina Kvalsina⁶, Andreas C. Scheinost⁷, Joseph Somers¹

- ¹: European Commission, Joint Research Centre, Institute for Transuranium Elements (ITU), Hermann-von-Helmholtz Platz 1, PO Box 2340, DE-76125 Karlsruhe, Germany
- ²: CEA, DEN, DEC, SPUA, LMPC, Centre de Cadarache, F-13108 Saint Paul Lez Durance Cedex, France
- ³: Synchrotron SOLEIL, 91190 Saint-Aubin, France,
- ⁴: CNRS-IPGP, 75005 Paris, France
- ⁵: Karlsruhe Institute of Technology, Institute for Nuclear Waste Management, D-76125 Karlsruhe, Germany
- ⁶: European Synchrotron Radiation Facility, 6 rue Jules Horowitz, BP 220, 38043 Grenoble, France
- ⁷: Helmholtz-Zentrum Dresden-Rossendorf, Institute of Resource Ecology, P.O. Box 10119, 01314 Dresden, Germany

Abstract

The introduction of new fast-neutron reactors like ASTRID, MYRRHA, ALLEGRO and AFFRED, is an important step towards a closed nuclear fuel cycle. The first cores will be fueled with (U,Pu)O₂ enabling Pu recycling. At a later stage, dedicated (U,MA)O_{2±x} targets for the transmutation of minor actinides (MA = Np, Am, Cm) will be deployed in dedicated assemblies at the reactor core periphery. In these cases, the substitution of tetravalent uranium by aliovalent atoms can lead, under specific conditions, to a modification of the uranium oxidation state leading ultimately to a change of the overall oxygen to metal ratio (O/M). Fresh fuels (e.g. U_{1-y}Pu_yO_{2±x} and U_{1-y}Am_yO_{2±x}) must have an O/M less than 2.00, to ensure that no reaction occurs with the cladding or with the coolant. Nevertheless, an oxidation of the fuel can occur in incidental conditions leading to an increase of the O/M ratio and ultimately affecting phase stability, defect chemistry and oxygen chemical potential. Therefore, an in depth knowledge of the impact of stoichiometry variation is essential for the safety assessment of these materials.

In the present work, the influence of three cations (i.e. La, Pu and Am) has been investigated, their selection being motivated by fundamental interests. The electronic structure and local environments of these materials have been studied combining several short- and long-range order techniques, including X-ray absorption near edge structure (XANES) and extended X-ray absorption fine

structure (EXAFS) spectroscopy. These room temperature methods have shown that ideal fluorite solid solutions are obtained for similar synthesis conditions. However, significant discrepancies have been observed regarding the charge distribution. Indeed, U(V) co exists with La(III) and Am(III), but not with Pu(III). In that latter case, U(V) is observed only when Pu is tetravalent. In addition, in situ XANES measurements have been performed on $(\text{U,Lu})\text{O}_{2\pm x}$ to study the charge distribution as a function of the temperature and the atmosphere. Thus, the formation mechanism of the solid solution and the accommodation of a aliovalent cation by the UO_2 structure can be understood.

Talk 2.1: Micro X-ray diffraction analysis of a fully recrystallized irradiated UO_2 fuel

Mélanie Chollet, Johannes Bertsch, Daniel Grolimund, Vallerie Ann Samson and Matthias Martin

- Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland

Abstract

During irradiation, the UO_2 nuclear fuel undergoes significant chemical and microstructural changes that could have significant performance and safety issues: changes in thermal conductivity, fission gas release, loss of mechanical integrity. In particular, above a certain burn-up (~ 60 MWd/kgU), the so-called high-burn structure (HBS) develops at the rim of the pellet. It is characterized by a high intra- and inter-granular porosity as well a reduction of grain sizes down to the sub-micrometric level. At the Paul Scherrer Institute (PSI), we have characterized the microstructure of a very high-burn-up UO_2 pellet (average 80 MWd/kgU) by synchrotron micro-X-ray diffraction (XRD). Particles from the different locations of the pellet radius from the center to the periphery were collected, with sizes according to the activity limit at the micro-XAS beamline of the Swiss Light Source (PSI). After X-ray fluorescence mapping at the U-LIII edge of the samples to identify the position of the particles on the kapton sample holder, XRD patterns were recorded in transmission by a MarCCD camera using a microbeam of 1×1.8 micrometer². To complete the analysis, a fresh non-irradiated UO_2 sample was also characterized as a reference.

Analysis of the 2D XRD patterns show a complete re-crystallisation of the pellet even in the center which was not expected since HBS is known to preferentially develop at the edge of the pellet and higher temperatures prevailing in the center should anneal any defects. Full rings indicating very fine grains (nanometric), smaller than the beamsize, were observed in each analyzed particle. On the opposite, 2D patterns of the fresh UO_2 show well defined spots in agreement

with 10 micrometer grain size reported for this sample. Evolution of the full width at half maximum (FWHM) of the diffraction peaks and d-spacings as a function of pellet radius will be discussed in regards to the fresh sample and in terms of released strain and accommodation of fission gas products in the lattice. Ultimately, a comparison with the few comparable data available in the literature on lower and/or different UO_2 fuel chemistry will be commented.

Talk 2.2: Investigation of mechanical and local properties of high burnup (>60 GWd/tHM) UO_2 fuel with focus on the pellet rim

Fabiola Cappia^{1,2}, D. Pizzocri³, P. Van Uffelen¹, A. Schubert¹, R. Macián-Juan², V.V. Rondinella¹

- ¹: European Commission, Joint Research Centre, Institute for Transuranium Elements (ITU), Hermann-von-Helmholtz Platz 1, PO Box 2340, DE-76125 Karlsruhe, Germany
- ²: Technische Universität München, Boltzmannstraße 15, 85747 Garching bei München Germany
- ³: Politecnico di Milano, Nuclear Engineering Division, Via La Masa 34, 20156 Milan Italy

Abstract

During in-pile operation, the pellet-cladding mechanical interaction (PCMI) is strongly affected by the fuel mechanical properties and its microstructural changes. Upon reaching a certain burnup and temperature level, fission gas accumulation into bubbles causes fuel swelling and gap closure. At the pellet periphery, the steep increase of gas-filled porosity typical of the high burnup structure (HBS) formation affects the PCMI at high burnup. It is important to understand the consequences of such porosity levels under accident conditions.

Mechanical testing on irradiated fuel is very difficult due to extensive cracking and radial heterogeneities. Micro-indentation allows local micro-hardness measurements with radial resolution sufficient to tackle some fuel heterogeneities. In this work, we present Vickers micro-hardness indentation results obtained from light water reactor (LWR) UO_2 fuel in the local burnup range 60–200 GWd/tHM. The micro-hardness profiles are related to porosity distribution profiles measured by scanning electron microscopy image analysis, with particular focus on the pellet rim, where the HBS is present. At the pellet periphery, a noticeable decrease of the micro-hardness is seen, primarily related to porosity build-up, which decreases the load-bearing area.

The current data provides the first step for an assessment of micro-hardness and other mechanical properties in relation with local fuel microstructure and supports the development of predictive models in fuel performance codes like TRANSURANUS, with which the consequences of such material changes can be evaluated.

Keywords: Vickers micro-hardness, porosity, high burnup UO₂ fuel, PCMI, HBS.

Talk 2.3: Outcomes from the implementation of irradiation tests on minor actinides bearing oxide fuels

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Abstract

The PELGRIMM European project (“PELlets versus GRanulates: Irradiation, Manufacturing, Modelling”) deals with modelling and experimental studies on Minor Actinides (MA) bearing oxide fuels devoted to both homogeneous recycling in driver fuels and heterogeneous recycling in radial core blankets, in Sodium-cooled Fast Reactor (SFR). Significant effort is directed towards the development of irradiation tests. Behaviour under irradiation of Americium Bearing Blanket (AmBB) fuel is addressed through the MARIOS separate-effect irradiation aiming at studying gaseous release and swelling of small fuel discs as a function of temperature thanks to an innovative experiment design. Study of AmBB behaviour is also the purpose of the semi-integral irradiation experiment MARINE in which are tested both pelletized and spherepacked fuel under irradiation. A specific feature of MARINE is the use of pressure transducers allowing direct measurement of gaseous release from the fuels during the experiment. SPHERE experiment addresses the behaviour under irradiation of both pelletized and spherepacked fuel for homogenous MA recycling. The description and the status of these irradiation tests will be provided in the presentation. Through this description, the different realisation steps of an irradiation experiment will be illustrated:

- design phase studies and irradiation preparation with MARINE experiment,

- performing of the irradiation with SPHERE experiment,
- interpretation of the irradiation and obtaining of the experimental results with MARIOS post-irradiation examinations.

Acknowledgments

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Talk 2.4: The SPHERE irradiation experiment: comparison of irradiation performance of pellets and spherepac MA-bearing driver fuel

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Abstract

The SPHERE irradiation experiment is the latest in a series of irradiations studying the performance of minor actinide (MA) bearing driver fuels for future sodium fast cooled reactors (SFR). SPHERE has been designed to compare conventional pellet-type fuels with so-called sphere-pac fuels under similar conditions. The pellet and sphere-pac fuels have nearly identical compositions of $U_{0.76}Pu_{0.2}Am_{0.03}O_{2-x}$.

The SPHERE irradiation was conducted in the High Flux Reactor in Petten, within the FP7 project FAIRFUELS, and has run for 11 cycles between August 2013 and April 2015 up to an estimated burn-up of ~5% FIMA. Linear power in both fuel stacks was ~300 W/cm at the start of irradiation. A post-irradiation neutron radiogram has revealed central hole formation in the sphere-pac fuel only.

Post irradiation analyses are underway in FP7 project PELGRIMM and consist of non-destructive testing (NDT), i.e. profilometry, gammaspectrometry and puncturing of the pins for analysis of fission gas release. A summary of the irradiation history and preliminary results on the NDT campaign are presented.

Talk 2.5: New Catalog on (U,Pu)O₂ Properties For Fast Reactors: Work Plan Of The ESNII+ Project and Perspectives

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Abstract

In order to develop the fast neutron systems, three prototypes of the Sodium Fast Reactor, the Gas-cooled Fast Reactor and a heavy liquid metal cooled Accelerator Driven System are studied in Europe: ASTRID (SFR prototype), ALLEGRO (GFR prototype) and MYRRHA (an LBE-cooled ADS system related to the ALFRED LFR-demonstrator). The ESNII+ project with its work package 7-FUEL SAFETY aims to provide a set of oxide fuel properties needed for the fuel element design of each prototype. The improvement of fuel properties will also reduce uncertainties in safety behaviour evaluations, in nominal conditions as well as during transients and will be achieved by the update of the European catalogue on the MOX fuel properties, used for the fuel design of the ESNII prototypes. While the properties of fresh fuel will be updated with new measurements, properties on irradiated mixed oxide fuel will be integrated and a tentative correlation with burn-up will be proposed.

The associated uncertainties have to be rigorously determined; the two main driver criteria for fuel element evaluation are the margin to melt for the fuel and the risk of clad failure. It is commonly assumed that the properties of the fuel yield the dominant contribution to the uncertainties in thermal and mechanical evaluation. The measurements of the main thermal and mechanical properties on fresh and irradiated fuel should enhance the reliability of fuel behaviour calculations and contribute to the design of safer fuel elements for ALLEGRO, ASTRID and MYRRHA. Indeed this work starts with property measurements on existing fresh and irradiated fuel samples, identified to cover the fuel characteristics for ESNII prototypes. The choice of the different samples

was the result of an analysis during the project preparation and lead to identify fresh and irradiated existing samples with suitable characteristics.

Finally, this work will be a guidance to focus experimental works on features of interest. Indeed this work plan will start with property measurements on existing fresh and irradiated fuel samples, identified to cover the fuel characteristics for the 3 prototypes.

Talk 3.1: Separated effect studies: a necessary experimental approach to understand the fuel behavior under in-pile irradiation

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Abstract

Uranium dioxide (UO₂) pellets are worldwide used as a nuclear fuel for power plants. During in-pile irradiation, extended defects (dislocations) are produced in the fuel (mainly because of the energy loss of fission products), as well as rare gases, like Krypton or Xenon. After more than 60 years old of studies on the behavior of UO₂, it remains a lack in the formation and release of fission gases knowledge. Release of these fission gases is an important nuclear safety issue, since the integrity of the first barrier of containment has to be warranted. A previous study [1] has shown that the presence of the extended defects induce a preferential growth of fission gas bubble on these defects. Hence, the study of extended defects created under irradiation is a significant step to better understand the behavior of fission gas bubbles.

The evolution of extended defects under irradiation is the result of the simultaneous influence of several parameters: temperature, burn-up, radiation damages... However the understanding of the contribution of each parameter on the evolution of these defects remains yet limited in in-pile conditions. Using a separated effects approach, the current study attempts to clarify the effect of two parameters: temperature and radiation damage on the extended defects evolution. One promising way to understand the behavior under irradiation of the UO₂ nuclear fuel is to use ion implantations and further analyze the microstructure. So, this experimental study is based on energetic ion implantations on polycrystalline

UO₂ at high temperature (600°C), and at the liquid nitrogen temperature, followed by thermal annealing. In-situ observations at the nanometer scale using Transmission Electron Microscopy enable a description of extended defects evolution with the combined effect of temperature and irradiation, without any temperature contribution, and with only the temperature effect.

Complementary techniques, which provide characterizations at the macroscopic scale, are also coupled with TEM observations, and show a good correlation as highlighted in our previous study [2].

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Talk 3.2: On the Raman Spectrum of Plutonium dioxide: vibrational and crystal electric field modes

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Abstract

In the present work, the Raman spectrum of plutonium dioxide is studied both experimentally and theoretically. Particular attention has been devoted to the identification of high-energy modes at 2120 cm⁻¹ and 2625 cm⁻¹, the attribution of which has so far been controversial. Crystal Electric Field calculations reported in this work show that these two modes can be respectively assigned to the $\Gamma_1 \rightarrow \Gamma_5$ and $\Gamma_1 \rightarrow \Gamma_3$ CEF transitions within the 5I4 manifold. These two modes, together with the only vibrational line foreseen by the group theory for the Fm⁻³m PuO₂ symmetry, the T2g Pu-O stretching vibrational peak observed at 478 cm⁻¹, can thus be retained as the Raman fingerprint of fcc plutonium dioxide. In addition to the Stokes spectrum, a parallel study of the T2g anti-Stokes lines, carried out in this research for the very first time, allowed us to reasonably estimate the temperature dependence of the observed Raman modes. The definition of a consistent Raman fingerprint for plutonium dioxide

can have important applications in nuclear waste management, nuclear forensic and safeguard activities.

Talk 3.3: The study of the $\text{UO}_{(2+x)}$ crystalline structure with high-resolution neutron diffraction experiment

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Abstract

The basic properties of UO_2 are essential to understanding and predicting the in-reactor behavior of fuels. In particular, UO_2 oxidation has been studied for many years in relation to both fuel fabrication and spent fuel storage issues. Phase transition properties of oxide fuels are fundamentally controlled by the nature and behavior of the atomic-scale defects on the anion sublattice.

In this study we aim to characterize the local crystalline structure incorporated with oxygen rearrangement has occurred in the $\text{UO}_{(2+x)}$ ($0 < x < 0.25$) in order to better describe the formation of defects and atomic thermal motions as a function of temperature. Neutron diffraction is one of the rare analytical techniques which enables the bulk properties of the anion sublattice to be studied.

We present here a neutron diffraction experiment in ILL Grenoble on several samples with different deviations-from-stoichiometry in a temperature range covering the phase transition between the two-phase ($\text{UO}_{(2+x)}$ and U_4O_9) and single-phase ($\text{UO}_{(2+x)}$) zones. The PDF (Partial Distribution Function) analysis and the Rietveld refinement on both $\text{UO}_{(2+x)}$ and UO_2 fresh fuel specimens will also be performed to understand how and to what extent the temperatures distort the long-ranging ordering of the dispersed oxygen defects in the hyperstoichiometric uranium dioxide. What is more, we focus particularly on the dependence upon temperature and deviation-from-stoichiometry of lattice parameter and the Debye-Waller factors of both uranium and oxygen sublattice between 296K and 1273K. A classic Debye behavior has been observed in UO_2 which compares favorably to the previous research.

Talk 3.4: Air storage effect for Kr and Xe transport in UO₂

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Abstract

Under separate effect study of UO₂ fuel, the behavior of noble gases Kr and Xe were studied in collaboration with the LLCC / Cadarache and CENBG. The objective is to understand the transport of volatile elements depending on different manufacturing parameters (stoichiometry, sintering, polishing, irradiation).

For this, the samples are prepared at LLCC, and characterized by PAS, RBS and TEM. Then, they are irradiated by Kr or Xe ions at low fluences in order to limit radiation damage and the precipitation phenomena in the material. After that, these samples are analyzed by thermal desorption coupled with high-sensitivity mass spectrometry, which allows to trace back the different transport processes and to determine the diffusion coefficients. A first series of experiments (A. Michel, University of Caen, 2011) was to implant Kr and Xe ions at low fluence ($\sim 10^{11}$ at/cm²) and at energies of 250 and 400keV. At these doses, the precipitation of the rare gas in the bubbles is low, and the release is dominated by vacancy diffusion. The measured isotherms clearly show a quick process (burst) superimposed on a slower process linked to the diffusion of noble gases in stoichiometric UO₂. An interpretation of this burst is a surface oxidation layer which allows faster diffusion of atoms.

To demonstrate the influence of the air storage time and therefore oxidation, we will present the study of samples that were stored in contact with the air over periods ranging from seconds to year. It appears that the burst is visible when the sample is in contact with air for a minimum period. Diffusion coefficients and the thickness of the oxidized layer could be determined with a macroscopic model and Fick's law.

Talk 3.5: Long term structural stability of severely alpha-damaged fuels

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Abstract

During the irradiation of the most commonly used nuclear fuels, minor actinides like americium or neptunium are produced. Together with plutonium they will be responsible for the long term radiotoxicity of spent nuclear fuel, and for the medium term heat loading of the fuel in open nuclear fuel cycles. Increasing amounts of alpha-damage and helium during spent fuel storage and in the repository will be generated, as these transuranic elements are mainly alpha-emitters. In the SUPERFACT irradiation in the mid-80's, a UO_2 fuel containing 20 at. % Np and 20 at. % Am has been irradiated in the Phenix fast reactor to demonstrate the transmutation feasibility of such fuels. It was shown that as a consequence of the actinide loading and irradiation very large amounts of helium were generated corresponding to standard irradiated LWR fuels aged for a few hundred thousand years. This aspect was investigated as a contribution to assessing properties and behavior of very aged spent fuels. Additionally, UO_2 samples doped with ^{238}Pu have been studied to characterize the effects of alpha-damage and helium production under accelerated conditions corresponding to spent fuel after storage/disposal time of centuries/thousands of years. In the present work we report on experimental observations of alpha-damage effects as well as helium release behavior in $(\text{U}_x, ^{238}\text{Pu}_{1-x})\text{O}_2$ and in the irradiated $(\text{U}_{0.6}, \text{Am}_{0.2}, \text{Np}_{0.2})\text{O}_2$ SUPERFACT samples by Transmission Electron Microscopy (TEM), and Electron Energy Loss Spectroscopy (EELS).

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Talk 3.6: Valence states and local structure of Gd doped actinide oxides $(\text{U}_{1-x}\text{Gd}_x)\text{O}_2$ and $(\text{Th}_{1-x}\text{Gd}_x)\text{O}_2$ measured by X-ray Absorption Spectrometry on the MARS beamline.

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Abstract

To improve the performance of nuclear fuels, one has to extend the capabilities of UO_2 fuels and develop alternatives, such as thoria based fuels [1,2]. To this aim, doped fuels such as $(\text{U}_{1-x}\text{Gd}_x)\text{O}_2$ and $(\text{Th}_{1-x}\text{Gd}_x)\text{O}_2$ allow controlled properties along fuel life-time, are of interest.

Gadolinia doped UO_2 is widely used to reduce the power peaking and excess reactivity during the first reactor cycle of fresh fuel assemblies as ^{155}Gd and ^{157}Gd isotope act as burnable neutron absorber. According to the fuel assembly design and the core management constraints, the concentration of Gd varies between 2 wt.% and 10 wt.% (weight fraction of Gd_2O_3). In such concentrations, Gd and UO_2 form a solid solution $(\text{U}_{1-x}\text{Gd}_x)\text{O}_2$ which keeps the fluorite structure of UO_2 by substituting U^{4+} ions by Gd^{3+} . The charge balance is usually considered as only modification of the valence state of U^{4+} to U^{5+} in good agreement with the fact that several phases are only observed for $x > 0.5$ [3]. Nevertheless, theoretical studies have recently revealed that it involves not only $\text{U}^{4+}/\text{U}^{5+}$ valence state mixing but also oxygen vacancies.

Thorium based nuclear fuels represent potential alternatives to uranium based fuels as the fission of thorium isotopes can reduce the long-lived radioactive nuclides. The influence of gadolinium content on doped thoria is of great interest to evaluate performance of this alternative fuel. The charge compensation is here awaited to be due to the presence of oxygen vacancies only, as recently demonstrated by molecular dynamics calculations [4]. Indeed, for thorium atoms, valence state modification is not allowed and Th^{4+} remains the only possible valence state in the fluorite structure.

The challenging question of the complex interplay between atomic defects and valence state mixing in $(\text{U}_{1-x}\text{Gd}_x)\text{O}_2$ and $(\text{Th}_{1-x}\text{Gd}_x)\text{O}_2$ will be discussed on the basis of X-ray Absorption Spectroscopy and theoretical calculations.

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Talk 3.7: Thermal Conductivity of the Uranium-Zirconium Oxide

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Abstract

A good knowledge of the thermophysical properties of a molten core is required in order to determine or analyse the scenario of a severe accident. For a Light Water Reactor (LWR), in a severe accident the Zircaloy cladding is oxidised by steam and corium forms by melting of the core material, with a composition depending on the reactor type. The typical core inventories range from U/Zr molar ratios of about 40/60 for a PWR to 60/40 for a BWR. A first series of simulated LWR corium was prepared taking into account the requirement of having samples representative of a molten material. Discs of Uranium-Zirconium oxide from a sol-gel assisted preparation method combined with Spark plasma sintering (SPS) were synthesised with U/Zr molar ratios of 0/100, 20/80, 40/60, 50/50, 60/40, 80/20 and 100/0.

The samples were characterised with X-ray Diffraction and SEM. The thermal diffusivity has been measured between 500 and 1600 K by the laser-flash technique. The thermal conductivity was calculated from the measurements of the thermal diffusivity, using literature data for the specific heat and thermal dilatation coefficients. The thermal conductivity of SPS UO_2 is in good agreement with the literature values available for sintered samples. The results show a fast degradation of the thermal diffusivity when the fraction of Zr increases. The extrapolation of the observed trend to pure ZrO_2 is not straightforward, as ZrO_2 is not stable over the whole temperature range, but shows a phase transition in the experimental temperature range and as it is a semi-transparent material requiring coating for the determination of its thermal diffusivity by the laser flash method. The results are compared to literature data and a recommendation is made.

Talk 3.8: Assessment of the Phase Equilibrium in the ThO_2 - ZrO_2 System

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Abstract

Nuclear Fuel oxides such as UO_2 , PuO_2 and ThO_2 are known to form solid solutions with ZrO_2 with monoclinic, tetragonal and cubic (fluorite) structures [1–3]. Studies of the phase equilibrium of these oxides with the ZrO_2 witnessed a vast amount of research due to its importance for nuclear fuel disposition. Thermodynamic modelling has been demonstrated for the majority of the oxide systems with experimental data widely existing for the UO_2 system [4–7]. However, very limited experimental data is present for the ThO_2 oxide. This may be related to the lack of interest in thorium oxide as a nuclear fuel due to it being not fissile. However, Thorium can still be used as a fuel in a nuclear reactor as a fertile material for the production of U-233, or as an inert matrix in the presence of a fissile material (e.g. U-233, U-235 or Pu-239) which acts as a driver to initiate a chain reaction. The development of such thorium based fuels is currently under investigation e.g. in Norway and India.

In this work, the phase equilibrium of ThO_2 - ZrO_2 is investigated. A series of different molar ratio (80:20, 60:40, 40:60, and 20:80 ThO_2 : ZrO_2) compositions of ThO_2 and ZrO_2 were synthesised using external sol-gel method utilised at JRC-ITU. The XRD data of samples sintered at 1650°C shows bi-phasic behaviour throughout the range compositions with small solubility (< 5%) of ZrO_2 into ThO_2 and limited solubility of ThO_2 into ZrO_2 . These results were supported with RAMAN spectroscopy and SEM microscopy to have a better understanding of the distribution of the components within each other. Furthermore, solid/liquid equilibria in the ThO_2 - ZrO_2 system are investigated here for the first time by laser heating coupled with fast optical thermometry. These results contribute to assessing the thermal stability of the system in a ThO_2 fuel based reactor.

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Talk 3.9: Fission products chemisorption mechanism following severe accidents: a separate effect study on CsOH reaction with stainless steel

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Abstract

Fission products chemisorption on reactor structural materials is an important phenomenon to be considered following an accident, as it can lead to retention of fission products in the containment, affecting the radioactive release to the environment and reactor decommissioning. The knowledge acquired from this study could help improve the models for cesium chemisorption and could be applied to evaluate the distribution and properties of Cs deposits in the Fukushima-Daiichi Nuclear Power Plant (1F-NPP), providing guidance for its decommissioning.

These separate effect studies were performed vaporizing CsOH over stainless steel samples, varying experimental parameters, such as temperature (800–1000°C) and atmosphere (H_2/Ar , $H_2/H_2O/Ar$). Sample surfaces were analyzed by SEM/EDX, XRD to evaluate the deposit microstructure and chemical composition. Cross sectional analyses were also applied to investigate the corrosion mechanism. Finally leaching tests were performed to evaluate the behavior of such deposit, showing that Cs is retained more effectively at low temperature and in reducing conditions. Our studies demonstrate a congruent distribution between Cs and Si, in agreement with the existing models[1] and the performed thermodynamics calculations, which predict the formation of cesium silicates under the experimental conditions. The XRD patterns permitted the chemical identification of the formed Cs compound as $CsFeSiO_4$. These results show the importance of the presence of impurities in stainless steel, as they can affect the formation of stable cesium deposits following a severe accident. More tests are however needed to extend our observations to a possible model (such as tests at 600°C, with high oxygen potentials, and using pre-oxidized samples). Finally it would be interesting to evaluate the effect of other chemicals on Cs adsorption, such as boron, which has been shown to be important during accident progression in a BWR.

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Talk 4.1: Thermodynamic Evaluation of a Matrix of High Density, Composite, and Accident Tolerant Fuels

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Abstract

Alternative compositions of light water reactor (LWR) fuels are being considered to replace solely UO_2 pellet fuel to either improve performance or to be more accident tolerant. Any alteration in fuel behavior will have a cascade effect on reactor operation such as expected thermal performance, choice of cladding, and fission product behavior. An important consideration is the thermodynamic stability of these fuel forms, whether inter-compatibility of composite fuel components, potential interactions with cladding, and behavior during generation of fission products. The addition of secondary phases to either UO_2 or high density fuel forms may result in potentially problematic property changes such as lower melting temperatures than the primary phase, which can result in reactor safety concerns, as well as lead to enhanced diffusion of fission products. Thus, an understanding of the phase stability as a function of both fuel phases and their relative ratios is important.

The fuel phases of current interest are urania, uranium borides, uranium mononitride, and uranium silicides. The phase equilibria of various combinations of these phases are expected to be designed to meet improved fuel performance and safety, and these systems have been preliminarily evaluated. As a result, phase regions of concern have been identified, and areas of insufficient knowledge and understanding delineated. This report reviews the matrix of high density and composite fuel options under consideration and provides a thermochemical understanding based on newly developed as well as available fundamental information.

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Talk 4.2: Inverse method for calculating thermo-physical properties of nuclear materials up to their melting point

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Abstract

The proposed method uses experimental thermograms obtained via laser- flash heating of a disc-shaped sample in combination with finite element analysis and parameter optimization. The experimental part involves heating samples to a steady state temperature via two lasers (on the back and front sides) and subsequently subjecting the front sample surface to a short laser pulse, resulting in a temperature transient (thermogram). A thermal camera records the temperature transients at equally spaced points along the radius of the rear surface of the sample. The noisy thermograms are filtered via Fast Fourier Transforms and a total variation minimization algorithm.

Subsequently, an optimization technique known as the Levenberg-Marquardt method is applied, whereby multiple parameters (emissivity, heat transfer coefficient, heat capacity and thermal conductivity) can be optimized and used as inputs in a finite element model. The parameters are changed until the least square difference between the numerical and experimental thermograms reaches a minimum. The calculated properties at each of the radial points correspond to different temperatures along the surface of the sample and thus temperature dependent expressions of these properties are obtained as well.

The method has been tested using both simulated and experimental data. Both heat capacity and thermal conductivity are in good agreement with literature data for POCO AXM-5 graphite.

Talk 4.3: In-situ high temperature X-ray diffraction study of Americium Dioxide

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Abstract

Partitioning and transmutation of Minor Actinides (MA) are currently studied to reduce radiotoxicity and heat generation in nuclear wastes. In the frame of minor actinides recycling in sodium fast reactors, (U,Am)O₂ mixed oxides are promising transmutation targets. To assess the thermodynamic properties of the U-Am-O system, it is essential to have a thorough knowledge of the binary phase diagrams, which is difficult due to the lack of thermodynamic data on the Am-O system[1]. Nevertheless, a thermodynamic modelling of the Am-O system has been recently proposed by Gotcu-Freiss et al.[2] AmO_{2-x} (fcc), AmO_{1.62} (bcc) and Am₂O₃ (hcp) phases are reported for an Oxygen/Americium (O/M) ratio ranging from 2 to 1.5.

Here, we investigated the Am-O system using in situ high temperature X-ray diffraction. First, we analyzed the evolution of the lattice parameter of the AmO_{2-x} (fcc) phase under air in temperature up to 1873 K. Below 1200K, the lattice parameter change is associated only to the temperature effect and the thermal expansion coefficient was calculated. Above this temperature, we observed a steeper increase of the lattice parameter, which is due to a reduction of the phase, i.e. a lower O/M, as predicted by our thermodynamic calculations based on the Gotcu-Freiss model.

Second, we explored the AmO_{2-x} region (1.6<O/M< 2) under controlled atmosphere as a function of the temperature and of the oxygen potential and we monitored the evolution of the lattice parameter induced by hypo-stoichiometry. By coupling our results with the thermodynamic calculations, we were able to correlate the lattice parameter and the O/M. Future thermogravimetric analyzes in the same temperature-atmosphere conditions are envisaged to further confirm this relationship.

Finally, in the presentation, we will discuss the existence of a miscibility gap[3] and the general agreement of our experimental results with the thermodynamic model.

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Talk 4.4: Thermodynamic assessment of the neptunium-oxygen system: mass spectrometric studies and thermodynamic modelling

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Abstract

A thorough knowledge of the inherent characteristics and behavior under normal and accidental conditions of advanced nuclear fuels to which minor actinides have been incorporated, i.e. (U,Pu,Np,Am,Cm)O₂ fuel, is essential for the safe use of future Generation IV nuclear reactors. Temperatures can reach up to 2273 K in normal operating conditions at the centre of the fuel pin of Sodium-cooled Fast Reactors, and about 893–923 K on the pellet edge [1]. The prediction of the nature of the oxide phases formed and their compositions under specific temperature and oxygen potential conditions is crucial. Moreover, the determination of their liquidus temperatures and evaporation processes is also needed in the scenario of an accident, with uncontrolled temperature increase.

The binary U-O, Pu-O and ternary U-Pu-O systems have been investigated extensively already, and thermodynamic models have been developed for these systems using the CALPHAD method [2,3]. The data available on the Np-O system are much more limited [4], however, and there is no satisfactory overall description using CALPHAD. In the context of heterogeneous in-pile recycling where a high concentration of minor actinides is added to UO₂ fuel assemblies, the knowledge of this system is essential, and a sound description via models is needed.

The experimental data available on the neptunium-oxygen system, encompassing phase equilibria, thermodynamic data, oxygen chemical potential, and vaporization studies of NpO₂, have been critically reviewed in the present work. Knudsen effusion cell mass spectrometry measurements have also been performed on neptunium dioxide to improve the understanding of its vaporization behavior. A

thermodynamic model for the neptunium-oxygen system has furthermore been developed using the CALPHAD method. The non stoichiometric NpO_{2-x} phase is described herein using the compound energy formalism with ionic constituents, while the liquid phase is represented with the ionic two-sublattice model. The reliability and consistency of all optimized parameters has been verified by comparison of the calculated and experimental data.

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Talk 4.5: Thermodynamic Investigations of the Uranium-Molybdenum-Oxygen System by a Coupling of Density Functional Theory and Calphad Methodologies

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Abstract

The process of fissioning uranium oxide or mixed oxides of uranium plutonium nuclear fuels results in the production of a large number of fission and activation products (e.g., Mo, Ce, Ru, Sr, Ba, La, Nd, Pd, Rh, Cs, Pu, Xe, Np, Y, Tc, Pr, Rb, Te, I, etc.). These elements distribute themselves into phases (e.g., gas, liquid, or solid) depending on burnup (composition), temperature, and pressure. The chemical state of the fuel has a direct influence on the physical properties and consequently on fuel behaviour (e.g., melting point, swelling, fission gas release, thermal conductivity, etc.). Thermodynamic modelling to predict the chemical state can be beneficial for safety analysis, understanding fuel oxidation, ascertaining potential interaction with other reactor materials, interpretation of experimental data of discharged fuel, corrosion, waste disposal,

and waste reprocessing. As Molybdenum (Mo) is one of the more abundant fission products in nuclear fuel and has a large influence on the fuel oxidation; a thermodynamic model for high burnup oxide fuels must include a careful assessment of the uranium (U) – molybdenum (Mo) –oxygen (O) system.

The U-Mo-O system has been reviewed extensively. Numerous ternary oxides have been identified by crystallographic means, however only a few compounds have been thermodynamically studied. Therefore, when no thermodynamic data was available in the literature, further experiments were completed or (where possible) an ab-initio paired quasi-harmonic model was used to generate these data.

This presentation reviews:

1. the available thermodynamic data in the literature for the U-Mo-O system,
2. experiments completed to gather missing data,
3. a review of the ab-initio paired quasi-harmonic model,
4. the thermodynamic equations used to assess these data, and
5. the thermochemical model developed for the U-Mo-O system.

Talk 4.6: Thermodynamic modeling of the Ni-Te and Fe-Te intermetallic phases

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Abstract

For the purpose of evaluating the possibility of premature stainless steel cladding failure due to internal corrosion induced by volatile fission products in Gen IV Sodium cooled Fast Reactors, CEA is in cooperation with KTH performing thermodynamic assessments of Metal-Fission Product systems, using the Calphad method, as a contribution to the TAF ID database. The thermodynamic descriptions are the fundamental basis for understanding and modeling the corrosion process, which may possibly be a life-limiting factor of the ASTRID Gen IV prototype reactor design.

In this study, both Fe Te and Ni Te binary systems have been under investigation. In the Fe Te system there are two known intermetallic phases stable

at room temperature, the tetragonal β -phase ($\sim\text{Fe}_{1.11}\text{Te}$) and the orthorhombic γ -phase (FeTe_2). A high-temperature rhombohedral δ -phase ($\sim\text{Fe}_{1.125}\text{Te}$) has been analysed by high Temperature XRD and has earlier been found as a ternary $\text{Fe}_{1.5}\text{Ni}_{1.5}\text{Te}_2$ phase. This is not stable in the Ni-Te system, where instead another high temperature phase resides with an orthorhombic structure. The NiAs type structure is present in both system as the monoclinic ϵ -phase ($\text{Fe}_{0.75}\text{Te}$), and in the Fe-Te system undergoes a first-order transition into the CdI_2 type hexagonal ζ -phase ($\text{Fe}_{0.67}\text{Te}$) at increasing Te content; in contrast, this is a second-order transition in the Ni-Te system. In the Ni-Te system this phase extends in composition up to the CdI_2 prototype composition whereas in the Fe-Te system the ζ -phase becomes stable.

The intermetallic compounds are described using sublattice models representative for the physical ordering of atoms in the crystal structure lattices; vacancy defect sublattice models are used for any phase with published evidence that the heterogeneity in composition is due to partly occupied interstitial metal atom sites in a matrix of Te atoms. There are published indications regarding a possible liquid miscibility gap in the Fe-Te composition range, whereas this is not evident in the Ni-Te system. An ionic associate liquid model ensures the possibility to model this in the Fe-Te system while experiments to attempt the confirmation of this are planned. Results on the thermodynamic modelling of the two binary systems will be presented.

Talk 5.1: Simulation and Measurement of Thermal Conductivity in UO_2

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Abstract

To fully utilise nuclear fuel its material properties must be maintained under extreme conditions. Thermal conductivity is particularly important owing to its role in mitigating centreline fuel melting. Firstly, the role of UO_2 magnetism in phonon scattering has been investigated experimentally. Furthermore, through the Callaway Model magnetism sufficiently accounts for the difference between molecular dynamics and experiment at low temperatures. By combining defect-phonon scattering (from molecular dynamics) with the magnetic contribution

(from experiment), the Callaway Model can be parameterized to predict the degradation of UO_2 thermal conductivity due to different fission products. Zr, Xe, La and Pu are all predicted to influence thermal conductivity to different degrees depending on their valence state and atomic size relative to the host lattice.

Talk 5.2: Mechanical behaviour of UO_2 under irradiation: a molecular dynamics study

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Abstract

Mechanical properties of nuclear fuel are a complex problem, involving many coupled mechanisms occurring at different length scales. Most of our knowledge regarding this mechanical behaviour is obtained by experiments on unirradiated fuel, or post-mortem analysis on spent fuel. Atomistic models can alternatively provide insight on the behaviour of the fuel in conditions inaccessible to current experiments, which are used as input parameter for models at larger length-scales. The present investigation considers recent studies on the evolution of the mechanical properties of UO_2 after irradiation damage by means of atomistic simulations with molecular dynamics method using empirical potentials.

Firstly, we will describe the mechanical behaviour under tensile and compressive load in pristine UO_2 single-crystal. Secondly, mechanical properties and crack propagation mode will be investigated in large UO_2 single-crystal including point defects distribution arising from irradiation events. The influence of the crystallographic orientations, temperature, and dose will be discussed.

Finally, we will present the influence of gas fission nanobubble distribution in the UO_2 matrix on the mechanical properties.

Talk 5.3: Xe diffusion and bubble nucleation around edge dislocations in UO_2

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Abstract

As the primary phenomenon involved in plasticity, dislocation behaviour is of highest importance to describe creep mechanisms in UO_2 nuclear fuel.

In particular, in high burnup fuels, dislocations interact with various radiation-induced defects such as voids, bubbles and intrinsic point defects.

In this study, we focus on the interaction of dislocations with fission gas bubbles. Whilst previous work have shown the role of dislocations in the formation of xenon bubbles[1], little is known about the effect of those bubbles on dislocation mobility once they have formed.

In UO_2 the most active slip systems are $\{100\}$, $\{110\}$ and $\{111\}$. The three dislocation types have been simulated interacting with xenon bubbles. The critical stresses of such systems have been calculated depending on bubble size and density. We also describe the effect of dislocations on bubble properties.

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Talk 5.4: Simulation of fission gas diffusion in UO_2 nuclear fuel

David Andersson, Xiang-Yang Liu, Romain Perriot, Giovanni Pastore, Michael R. Tonks, Blas P. Uberuaga and Christopher R. Stanek

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Abstract

In UO_2 nuclear fuel, the retention and release of fission gas atoms such as xenon (Xe) are important for nuclear fuel performance. We use multiscale simulations to determine fission gas diffusion mechanisms as well as the corresponding rates in UO_2 under both intrinsic and irradiation conditions [1,2]. Density functional theory (DFT) calculations are used to study formation, binding and migration energies of small clusters of Xe and vacancies. Empirical potential calculations enable us to determine the corresponding entropies and attempt frequencies for migration as well as investigate the properties of large clusters or small fission gas bubbles. A continuum reaction-diffusion model is developed for Xe and point defects based on the mechanisms and rates obtained from atomistic simulations. Effective fission gas diffusivities are then obtained by solving this set of equations

for different chemical and irradiation conditions using the MARMOT phase field code[2]. The predictions are compared to available experimental data. The importance of the large XeU_3O cluster (a Xe atom in a uranium + oxygen vacancy trap site with two bound uranium vacancies) is emphasized, which is a consequence of its high mobility and high binding energy. We find that the XeU_3O cluster gives Xe diffusivities that are higher for intrinsic conditions than under irradiation over a wide range of temperatures. Under irradiation the fast-moving XeU_3O cluster recombines quickly with irradiation induced interstitial U ions, while this mechanism is less important for intrinsic conditions. The net result is higher concentration of the XeU_3O cluster for intrinsic conditions than under irradiation. We speculate that differences in the irradiation conditions and their impact on the XeU_3O cluster can explain the wide range of diffusivities reported in experimental studies. However, all vacancy-mediated mechanisms underestimate the Xe diffusivity compared to the empirical radiation-enhanced model used in most fission gas release models. We investigate the possibility that diffusion of small fission gas bubbles or extended Xe-vacancy clusters may give rise to the observed radiation-enhanced diffusivity. These studies highlight the importance of U divacancies, surface termination and non-stoichiometry for the cluster migration properties. Finally, diffusion of Xe is strongly impacted by the temperature distribution in the fuel rod and, in this context, studies of the contribution from phonon-spin scattering [3] as well as other mechanisms to the reduction of UO_2 thermal conductivity will be briefly discussed.

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Talk 5.5: Atomic-scale effects of chromium-doping on defect behaviour in uranium dioxide fuel

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Abstract

The effects of doping conventional UO_2 fuel with chromium are studied through atomistic simulations using empirical force field methods. We first analyse the stable structures of unirradiated doped fuel by determining the preferred lattice configuration of chromium ions and oxygen vacancies within the matrix. In order to understand the physical effects of the dopants, we investigate formation energies of isolated defects and Frenkel pairs in the vicinity of chromium. The behaviour of point defects is then studied with collision cascade simulations and relaxation of doped simulation cells containing Frenkel pairs. The defective structures are scanned in minute detail using a powerful and versatile analysing tool, called ASTRAM, developed in-house for the purpose. Results indicate definite effects of chromium-doping on the ease with which defects are formed and also on the residual damage in the fuel matrix, which can be extrapolated to phenomena following irradiation under real operating conditions. We also show that the extent of these effects is related to the dopant concentration.

Talk 5.6: Modelling the thermomechanical properties of actinide oxides

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Abstract

Mixed actinide oxides have been used as nuclear fuel material, furthermore UO_2 fuel effectively becomes a mixed oxide during reactor operation due to transmutation and decay. As a result, understanding the behaviour of mixed oxides is of considerable importance. Atomic scale simulations are helpful in understanding the fundamental features governing fuel performance. In the current work, molecular dynamics simulations have been carried out using a new many-bodied potential model to investigate the thermophysical and diffusion properties of actinide oxides at high temperatures. Lattice parameter, enthalpy and specific heat have been calculated for a range of $(\text{Th}_x, \text{Pu}_{1-x})\text{O}_2$ compositions, over a temperature range 300–3200 K. It is difficult to obtain experimental data at higher temperatures for mixed oxides; therefore these simulations provide important insight for systems that are still not sufficiently well understood. In addition to the thermophysical properties, the oxygen diffusivity for $(\text{Th}_x, \text{Pu}_{1-x})\text{O}_2$ was also

calculated. The creation of oxygen defects and subsequently a superionic transition causes a ‘bump’ in the lattice parameter, enthalpy and specific heat plots. Enhanced oxygen diffusivity was also observed below this superionic transition.

Talk 5.7: First-principles modelling of the structural and electronic modifications induced by point defects in actinide mixed oxides

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Abstract

The first-principles DFT+U method is used to study structural, electronic and thermodynamic properties of actinide mixed oxides and their point defects. The objective is to determine the effect of the chemical composition and of the deviation from stoichiometry on the properties of these materials. First-principles electronic structure calculations enable one to get insight into some aspects of these phenomena at the atomic scale, in particular to identify the point defects that accommodate deviations from stoichiometry and to determine the structural and electronic modifications induced by the presence of the defects.

This approach is applied to the study of the actinide mixed oxides (U,Ce)O₂ and (U,Pu)O₂, the former being of interest as a weakly radioactive mixed oxide model material for separate effect experiments. We focus on oxygen defects with various chemical environments in mixed oxides of various compositions.

The results are confronted to experimental data from the literature, obtained by X-ray absorption spectroscopy and X-ray diffraction, and to the results obtained in pure UO₂.

Talk 5.8: Calculation of diffusion coefficients in ceramic oxide fuels using adaptive kinetic Monte Carlo modelling

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Abstract

Fuel performance modelling codes are reliant upon knowledge of material properties of the fuels, such as thermal conductivity or fission product diffusion coefficients. Diffusion coefficients can be difficult to measure experimentally and therefore it is often desirable to use modelling techniques, such as molecular dynamics, to support, or in place of, experimental measurements. However, the timescales and computational effort required to reliably calculate diffusion coefficients, particularly for cationic and fission product species, can be prohibitive. This work investigates the use of adaptive kinetic Monte Carlo (AKMC) modelling [1] as an alternative to classical molecular dynamics (MD) for the calculation of diffusion coefficients in ceramic oxide fuels. Whereas classical MD simulates molecular motion in ‘real time’, thus limiting practical simulations to timescales in the region of 10^{-9} – 10^{-6} s, AKMC is able to target the ‘rare-event’ atom translations associated with species diffusion and hence enables access simulations upwards of 10^{-3} s in length. In theory, the longer timescales accessible with AKMC enable statistically reliable calculation of diffusion coefficients for species present in irradiated nuclear fuels that have previously been challenging to obtain via atomistic modelling approaches.

Since oxygen diffusion in oxide fuels is understood to be several orders of magnitude faster than most other cationic or fission product species present, and is therefore readily accessible through classical MD, a comparison of oxygen diffusion coefficients in UO_2 as calculated using AKMC [2] and MD is presented to demonstrate this novel application of the AKMC modelling approach.

References:

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Talk 5.9: Ab initio determination of gas atoms migration energy in UN

Antoine Claisse, Luca Messina and Pär Olsson

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Abstract

Nitride fuels are being considered as an accident-tolerant option for both thermal and fast reactors, since their thermo-mechanical properties could allow for higher safety margins. However, R&D is still greatly needed, and in particular, the thermal diffusion coefficients of Xe and Kr reported in the literature differ by several (tens of) orders of magnitude.

This work proposes to refine this value by determining migration barriers in the DFT framework. To avoid meta-stable states, the occupation matrix control minimization scheme is used. The diffusion coefficient of Xe is then calculated by applying a self consistent mean field method [1] to the FCC antiferromagnetic sublattice of this alloy. The activation energy that is found is well within the experimental range.

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Talk 6.1: Recent improvements of the fuel thermomechanical modelling in the PLEIADES Platform to better simulate accidental transients conditions using the Alcyone fuel performance code

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Abstract

The French Alternative Energies and Atomic Energy Commission (CEA) and its industrial partners Électricité de France (EDF) and AREVA have been developing the collaborative platform PLEIADES for more than 10 years. This platform now supports the development of several state-of-the-art fuel performance codes: Alcyone and Cyrano for Pressurized Water Reactor (PWR), Germinal V2 for Sodium Fast Reactors (SFR), Licos for innovative fuel elements and experimental irradiation devices, MAIA for Material Testing Reactors (MTR), etc.

This paper first provides an up-to-date overview of the PLEIADES platform and then focuses on recent improvements of the thermomechanical modelling abilities recently introduced in PLEIADES's fuel performance codes. Various topics will be discussed and illustrated using Alcyone simulations of the PWR fuel rod in normal and off-normal situations, including LOCA and RIA:

- PWR fuel pellet cracking during the reactor start-up;
- description of grain boundary decohesion in the fuel oxide during a RIA transient;
- multi-fragments modelling of the PWR in 2D(r;);
- various numerical improvements ;
- finite strain modelling in 1D;

Talk 6.2: Explicit fracture models of nuclear fuel pellets using peridynamics

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Abstract

Three dimensional models of explicit cracking of nuclear fuel pellets for a variety of power ratings have been explored with peridynamics, a non-local, mesh free, fracture mechanics method. These models were implemented in the explicitly integrated molecular dynamics code LAMMPS, which was modified to include thermal strains in solid bodies. The models of fuel fracture, during initial power transients, are shown to correlate with the mean number of cracks observed on the inner and outer edges of the pellet, by experimental post irradiation examination of fuel, for power ratings of 10 and 15 W g⁻¹ UO₂. The models of the pellet show the ability to predict expected features such as the mid-height pellet crack, the correct number of radial cracks and branching of radial cracks as they reach the outer edge of the pellet. This work presents a modelling alternative to empirical fracture data found in many fuel performance codes, which requires just one parameter of fracture strain. Weibull distributions of crack numbers were fitted to both numerical and experimental data using maximum likelihood estimation so that statistical comparison could be made. The findings show P-values of less than 0.5% suggesting an excellent agreement between model and experimental distributions.

Talk 6.3: A semi-empirical model for the formation of the high burnup structure

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Abstract

In the rim zone of UO₂ nuclear fuel pellets, the combination of high burnup (>60 GWd/tU – i.e., high radiation damage and fission product concentration) and low temperature (<1000°C – i.e., limited thermal recovery of the radiation damage) drives a microstructural change, leading to the formation of the high burnup structure (HBS). In this work, we propose a preliminary model to describe the formation of the HBS. This process always includes four characteristic phenomena: (i) first, dislocations pile-up forming an entangled network, followed by (ii) the polygonization/recrystallization of smaller grains, together with (iii) the decrease of the intra-granular fission gas concentration (depletion), and (iv) the formation of a novel population of spherical bubbles. These processes are not strictly sequential, but may be thought as (partially) concomitant. The present model embraces phenomena (i-iii), describing them as inherently related. Based on experimental observations, we assume an exponential reduction of the average grain size (i-ii), paired with a simultaneous depletion of intra-granular fission gas driven by diffusion (iii). The assumption that a reduction in the average grain size can represent both the dislocation pile-up and the actual polygonization/recrystallization is based on the experimental observation of dislocations acting as sinks for fission gas. As regards the diffusion process, the determination of the intra-granular diffusion coefficient in the HBS is based on the combination of EPMA measurements of fission gas concentration and SEM measurements of the grain size. The underlying physical interpretation is that HBS formation can be seen as a phase transition from a structure with low surface to volume ratio (i.e., big grain size 10 μm) to a structure with much higher surface to volume ratio (i.e., small grain size 300 nm), the second being able to endure more radiation damage. In this picture, the depletion of intra-granular fission gas (iii) and the formation of new bubbles (iv) can be related to the increased surface to volume ratio, which enhances diffusion and promotes HBS pore growth.

Talk 6.4: Modelling Pellet Relocation in PEL-ICAN, a Finite Element Model for Pellet-Clad Interaction in AGR & PWR Fuel

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Abstract

Pellet fragment relocation is of importance in fuel as it affects the effective gap between the clad and fuel, impairs heat transfer, enhances fission gas release and leads to increased pellet-clad interaction (PCI). When fuel has been operating at low power for extended periods, return to power is limited by PCI restrictions. These reduce the ability of nuclear power to load-follow grid demands. We present a r- finite element model for pellet relocation and PCI. It is able to model the opening and closing of cracks between fuel fragments at the pellet waist as well as pellet-bore closure in the case of an annular fuel pellet. The model is able to predict stress concentration ahead of a radial fuel pellet crack as well as the additional radial displacement caused by pellet relocation during periods of reduced power operation.

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Talk 6.5: Simulations and Experiments of U-Mo/Al and U-Mo/Mg Research Reactor Fuels

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Abstract

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This work presents numerical predictions of the thermal behaviour of U-Mo/Mg pin-type research reactor fuels to support fuel development, performance and safety analyses at the Canadian Nuclear Laboratories. To circumvent fuel failures experienced with U-Mo/Al fuels resulting from the formation of a UAl_x interaction layer in-reactor, U-Mo/Mg fuels have been proposed, since Mg is for all practical purposes chemically inert with U and Mo. However, a separate interaction layer has been experimentally observed between the Mg meat and Al cladding, which has not resulted in any failures in the laboratory, but may be problematic in-reactor. Meso-scale simulations have been performed to provide best estimates of the thermal material properties. Thermodynamic modelling provides predictions of secondary phase formation, such as the beta and gamma phases represented by the Mg/Al interaction layers. Finally, continuum scale simulations are informed by meso- and thermodynamic-scales to provide thermal predictions under a variety of conditions. This multi-scale multi-physics framework continues to provide guidance for an on-going experimental campaign of U-Mo/Mg fuels with irradiation testing planned in the near future.

Talk 6.6: Modeling and simulation of fast reactor minor actinide bearing oxide fuels in support of MABB and MADF concepts

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Abstract

The European FP-7 project PELGRIMM addresses the development of Minor-Actinide (MA) bearing oxide fuels for Sodium-cooled Fast Reactors. MA homogeneous and heterogeneous recycling options are investigated in this project. Work-Package 3 (WP3) addresses modelling of minor actinide enriched fuels under irradiation regarding both MADF and MABB compositions - spherepacked or pelletized – in order to establish reliable predictive capabilities of existing fuel performance codes with respect to specific issues of MADF/MABB fuels.

The concept of MABB – Minor Actinide Bearing Blanket fuels – stands for use of depleted UO_2 with incorporated Am in elevated quantities (up to 10–15 wt.%) in radial blanket on the periphery of the outer core in fast reactors. Such (U,MA) O_2 fuels can also be doped with Np to accelerate the rate of plutonium build-up and to keep (U,MA) O_2 fuel longer in the reactor for deeper transmutation of Americium. In the complementary concept MADF – Minor Actinide Driver Fuel – compositions are manufactured with high initial content of PuO_2 for use in the core of a fast reactor to generate power and neutron flux.

Most of fuel performance codes have been originally developed, verified and validated to model standard LWR or SFR types of fuels made of enriched uranium dioxide or a mixture of uranium and plutonium oxides (MOX). Implementation of new compositions and fuel forms (pellets or spherepac) is naturally made by correction of existing models and correlations for standard fuels. Such approach is practical; however, validity of it still shall be tested. Many in-pile properties of fuels depend on temperature – the higher centre-line temperature the higher scale/rate of all temperature dependent processes and properties. Thus, thermal properties of MADF and MABB fuels under consideration are to be reviewed and analyzed first. The objective of the study performed in the FP7-PELGRIMM project and its WP3 is an assessment and a comparative evaluation of the fuel performance codes and their ability to model the behavior of fuel pins with either sphere-pac or pellets.

Several fuel performance codes (MACROS – SCK•CEN, TRANSURANUS – ENEA, NRG, JRC-ITU, GERMINAL -CEA) have been used by participants to cover the WP3 scope of work and particular tasks.

The PELGRIMM WP3 work started with the general model evaluations of material properties of MADF and MABB dedicated compositions and reference irradiation experiments – SUPERFACT and SPHERE. The efforts that participants of the WP3 made so far cover aspects of modeling such as helium production and release under a fast neutron spectrum; plutonium, americium and oxygen redistribution; influence of MA and fission products (i.e. burnup effects) on melting temperature; evolution rate of fuel restructuring, columnar grain growth and central void formation; and, finally, some predictive behavior simulations have been made for the SPHERE irradiation test.

There is a good understanding based on carried out work that concepts of MADF

and MABB fuels open promising prospective for safety assessments and design efforts underway in the WP4 which addresses problematics of core physics.

Poster Abstracts

Poster 1.1: Outcomes from investigations of oxide spherepacked fuels synthesis

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Abstract

The European FP-7 project PELGRIMM addresses the development of Minor-Actinide (MA) bearing oxide fuels consolidated as pellets and spherepac for Sodium-cooled Fast Reactors. Two fuel compositions: MABB and MADF, are under investigation. In the Minor Actinide Bearing Blanket (MABB), MA-oxide is concentrated in UO_2 and sub-assemblies are located in the radial blanket of the core. In MADF – Minor Actinide Driver Fuel – MA oxide is diluted in the (U,Pu)O₂ driver fuel.

Even if powder metallurgy, implemented to supply (U,Pu)O₂ standard fuels at industrial scale, can be used at lab-scale to prepare Am-bearing fuel samples, dust-free routes and simplified methods, are essential to scale-up the Am-bearing fuel fabrication processes. Three options have been investigated within the project: two are based on the sol-gel processes that lead to dense or porous spherical particles and the other relies on the adaptation to oxide fuels of the Weak Acid Resin technology.

For the MARINE irradiation test (implemented within PELGRIMM) that aims at comparing a sphere-packed and pelletized (U,Am)O_{2-x} fuel: the preparation of pellets includes a stage of UO_2 porous beads infiltration by a low acid Am nitrate solution, followed by heat treatments, pressing and sintering. The preparation of sphere-pac particles (50 and 800 μ m) was performed using direct co-precipitation of mixed (U,Am) nitrate solution by external gelation before heat treatments and vibro-packing.

In parallel, a variant of the internal gelation route, for the gelation of the drops using a microwave cavity instead of a silicon oil hot bath, has been investigated. Finally, the Weak Acid Resin flowchart has been revisited and adapted to oxide fuels for the actual synthesis of (U,Am)O₂ beads and pellets.

The experiences gained on fuel synthesis during these campaigns are presented and conclusions for future approaches are drawn.

Acknowledgments:

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Poster 2.1: High frequency acoustic microscopy as post irradiation examination technique

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Abstract

The non-uniform variation of porosity along the radius of the irradiated uranium dioxide pellets can contribute, in addition to other effects, to affect the safety of the nuclear fuel during the in-pile operations. Acoustic Microscopy is a non destructive technique that allows the determination of the mechanical properties of the irradiated nuclear fuel by measuring the Rayleigh surface wave velocity; in particular it enables the determination of the Young's modulus and the porosity on local scale. Due to the fact that matter is not opaque to acoustic waves, using high frequency acoustic microscopy is also possible to obtain information regarding the cracks below the surface of the sample. A study performed on a 67 GWd/tU uranium dioxide pellet has demonstrated the good comparability between the porosity calculated in different points of the radius by ceramographic image analysis and acoustic microscopy. The consistency of the results obtained has confirmed the validity of the acoustic microscopy as post irradiation examination technique.

Poster 2.2: TRABANT: Studies on a high power MOX irradiation

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Abstract

Between 2001 and 2005, several fast reactor MOX fuel pins were irradiated in the HFR Petten in the TRABANT programme. The PIE on one of these pins is currently being performed at NRG's Hot Cell Laboratories within the FP7 project ESNII+.

The pins (Phénix-cladding (15–15Ti SS) contain high content Pu annular MOX pellets that were originally produced in the CAPRA programme. The pins are submersed in a sodium-filled capsule and produce a linear power of 460–520 W/cm in the HFR. These powers correlate to the fast flux linear power in an SFR, which is achieved by placing the pins in a Cd-shielded rig that absorbs thermal neutrons. The designed central fuel temperature and cladding temperature were 2100°C and 600°C, respectively. The irradiation was stopped after three irradiation cycles after observations of unexpectedly high powers and an apparent mobility of the central hole in the pellets in neutron radiographs.

In this contribution the fuel characteristics, the irradiation history and the causes that lead to the unexpected behaviour are presented. Both non-destructive testing (gamma scanning, profilometry and fission gas release), as well as ceramography and element analysis (EDS, WDS) on different regions of interest will be reported.

Poster 2.3: Irradiation of Fuel Salts in the High Flux Reactor Petten

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Poster 3.1: Incorporation and migration mechanisms of Molybdenum in the stoichiometric and hyper-stoichiometric UO_2 .

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Abstract

Since the Fukushima accident, an increased attention has been paid to models used for evaluating the radiological impact of radionuclides (RN) potentially released to the environment. During an accident in a Pressurized Water Reactor (PWR), oxidation of the nuclear fuel likely occurs, causing an increase in the oxygen potential of UO_2 . It leads to the formation of UO_{2+x} which impacts the local microstructure of the fuel and the RN transport properties [1]. A better understanding of the chemical and physical mechanisms responsible for the RN release in accidental conditions is then necessary.

Among fission products (FP), Molybdenum (Mo) is one of the most abundant with a fission yield equivalent to that of Xe. Moreover, it is poorly soluble into UO_2 and can precipitate either as an oxide or as a metallic phase. Especially Mo is known to have an indirect impact on the radiotoxic species speciation as it can form complexes with other elements, like the CsMoO_4 complex, thus changing the mechanisms and amounts of released FP [2,3]. In addition, Mo has a very important role in the fuel chemistry and is known to influence the oxygen potential of UO_2 [4,5].

This work studies the incorporation mechanisms of Mo and its diffusion in a hyper-stoichiometric UO_2 matrix, when subjected to high temperatures and/or

irradiation. The aim is to determine the thermal (and athermal if possible) diffusion coefficient of Mo by coupling an experimental approach with DFT and molecular dynamics calculations. Depleted UO_2 pellets are oxidized up to a O/U ratio equal to 2.1, using thermal annealing under a controlled oxidizing atmosphere. They are implanted with a stable isotope of Molybdenum (^{95}Mo), and annealed and/or irradiated. The ^{95}Mo migration in the $\text{UO}_{2.1}$ matrix is followed by SIMS. Matrix microstructural modifications are characterized by XRD and Raman spectroscopy, and the superficial UO_2 stoichiometry is determined by Nuclear Reaction Analysis. A comparison with first DFT results is proposed.

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Poster 3.2: Thermal Diffusivity and Conductivity of Thorium- Uranium Mixed Oxides

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Abstract

Thorium-Uranium oxide pellets with high densities were prepared at CNL by co-milling, pressing, and sintering at 1700°C, with UO_2 mass contents of 0, 1.5, 3, 8, 13, 30, 60 and 100%. At ITU, $(\text{Th,U})\text{O}_2$ solid solution pellets were fabricated using the spark plasma sintering (SPS) technique with 79 and 93 % wt.% UO_2 . The thermal diffusivity was measured at CNL and at ITU using the laser flash technique and the thermal conductivity was deduced using the measured density and literature data for the specific heat.

The thermal conductivity for ThO_2 is significantly higher than for UO_2 . The thermal conductivity of $(\text{Th,U})\text{O}_2$ decreases rapidly with increasing U content,

and for UO_2 contents of 60 % and higher, the conductivity of $(\text{Th,U})\text{O}_2$ is close to UO_2 . The new results are compared to the data and models available in the literature.

As the mass difference between the Th and U atoms is small, the thermal conductivity decrease is attributed to the phonon scattering enhanced by lattice strain due to the introduction of uranium in ThO_2 lattice. However, this interpretation does not take into account the semi-transparent nature of ThO_2 , where heat transfer takes place by a coupled conductive and radiative mechanism. The effect of the semitransparency is clearly visible when the thermal diffusivity is measured by the laser pulse technique, with an almost instantaneous temperature rise of the sample surface opposite to the surface receiving the laser pulse. This effect is tempered by coating the front and rear surfaces of the samples with carbon, which ensures the laser energy deposits at the sample surface only, and not in the volume. However, a contribution of the radiative heat transfer remains present on the thermograms, and therefore also on the determined apparent thermal diffusivity. This semitransparency decreases progressively with the addition of UO_2 and is almost lost at about 30 % of UO_2 in ThO_2 .

Poster 3.3: Interaction study between MOX pellets and lead-bismuth eutectic coolant

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Abstract

The MYRRHA reactor is an accelerator driven system (ADS) in development. The sub-critical core of this reactor contains uranium-plutonium mixed oxide (MOX) fuel with 30% of PuO_2 . The coolant is liquid lead-bismuth eutectic (LBE). In this context, interactions tests between MOX and LBE have been performed. The three parameters investigated in this study is the O/M ($M=\text{U}+\text{Pu}$) ratio in the fuel with tests on stoichiometric $\text{U}_{0.7}\text{Pu}_{0.3}\text{O}_{2.00}$ and substoichiometric $\text{U}_{0.7}\text{Pu}_{0.3}\text{O}_{2-x}$, the temperature (500°C and 800°C), and the oxygen content in the LBE. These tests took place in sealed containers for 50 hours. Thereafter pellet integrity was checked by optical microscope. The presence of potential interaction compounds was investigated with XRD of the pellet surface and SEM analyses of pellet cross sections. Finally, LBE samples were dissolved

in acidic solutions to perform ICP-MS analysis and determine the actinide release to the LBE during the tests. In general, the results of these tests show a good compatibility between MOX and LBE without significant reaction detected. Nevertheless, significant actinide release from the pellets was detected for specific conditions.

Poster 3.4: Further insight into the chemistry of the Bi-U-O system

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Abstract

Safety assessment for a lead-bismuth eutectic (LBE) cooled reactor requires detailed knowledge of potential lead and bismuth uranate phases that could form in the event of a pin breach. Thus, we studied the phase relations in the Pb-U-O and Bi-U-O systems to identify possible phases that could be formed, and in a second step to determine their properties. Cubic fluorite-type phases have been reported in the U(IV) system, $\text{UO}_{(2+x)}\text{-Bi}_2\text{O}_3$, over the entire compositional range. However, a non-linear evolution of the lattice parameter with relative stoichiometry has been observed, in contrast to other M(III)-U(IV)-O systems. This unusual behaviour was explained previously based on the fact that the substitution increases the amount of anionic vacancies than going towards higher Bi_2O_3 content. In these extensive investigations on the $\text{UO}_{(2+x)}\text{-Bi}_2\text{O}_3$ system, we have confirmed the unusual behaviour of the lattice parameter with composition, but have found that, even under inert atmosphere at 800°C, U(IV) is oxidised to U(V)/U(VI) as a function of the substitution degree. Thus, using a combination of three methods (XRD, XANES and Raman) we have identified the formation of the BiU(V)O_4 and $\text{Bi}_2\text{U(VI)O}_6$ compounds, within this series.

Poster 3.5: Features of Zr-rich corner of Zr-N-O ternary system by controlled low-pressure oxidation

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Abstract

It is well known that in accidents involving LWR fuels with air ingress, the oxidation of zirconium alloys are much faster with larger heat generation. This vigorous oxidation is related to the formation of zirconium nitride and the instability of the ZrO₂ layer in the presence of nitrogen [1, 2]. On the other hand, even the reliable ternary phase diagram of Zr-N-O system nor relevant thermodynamic data are still lacking. Due to the existence of the oxynitride phases, the N-rich part of the Zr-N-O ternary system has attracted much attention. But the information on the Zr-rich corner is scarce. We investigated the feature of the Zr rich corner of the Zr-N-O ternary system by oxidation experiments in low oxygen partial pressure conditions. The experiments included both the separated reaction sequence (oxidation of the nitrated Zr) and also the simultaneous reaction sequence (simultaneous nitridation and oxidation of fresh Zr). Low oxygen partial pressure conditions were realized by utilizing REDOX couples of Mo/MoO₂ and CuO/Cu₂O. After heating, the samples were analysed by XRD, EPMA and laser raman spectroscopy. We found that at a low partial pressure of oxygen the oxidation proceeds both at the surface of ZrN layer and at the interface of Zr(N,O) and -Zr(O,N). The latter proved to be much faster than the former. Based on the results, the homogeneity ranges of nitride Zr(N,O)_{1-x} and metal -Zr(O,N) phases will be discussed.

Key words: Zr oxidation, ZrN, Zr-N-O phase diagram.

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Poster 3.6: Kinematic hardening in creep of Zircaloy 2

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Abstract

Kinematic hardening is an increase in flow stress in the course of plastic deformation. In contrast to isotropic hardening, the flow stress decreases upon load reversal. The latter phenomenon is also known as the Bauschinger effect. It is well known that kinematic hardening occurs during plastic deformation of Zircaloy. There is a limited number of slip systems in the hexagonal crystallographic structure of alpha-Zirconium which results in intergranular deformation incompatibilities and buildup of back stress. The back stress opposes the deformation during prestrain, and favours it upon load reversal.

The situation is less clear for creep: here, the kinematic hardening would manifest itself by strain rate decrease followed by its increase (in magnitude) upon stress reversal. In theory, it could be possible that due to long times at high temperatures, the back stress would relax to a large extent, e.g. by some diffusional process, and the kinematic hardening in creep would be less pronounced than in plastic deformation. On the other hand, creep experiments with stress change reported in the literature indicate that isotropic hardening alone cannot explain the observed transient creep behavior. However, a systematic investigation of kinematic hardening in creep of Zircaloy is not available in the literature so far.

The present work describes a dedicated creep experiment with stress reversal on Zircaloy 2 to investigate possible kinematic hardening. Gas filled cladding tube samples were deformed in furnace (internal pressure, resulting hoop stress of 75 MPa at 380°C) and in autoclave (outer overpressure, resulting hoop stress of -75 MPa at 380°C). The samples were periodically removed after each 7 days in furnace/autoclave to measure the outer diameter. Two of the samples were removed from the furnace and put into the autoclave after 35 days, two were moved from the autoclave to the furnace. For comparison, two other samples were continuously deformed in the furnace, and two others in the autoclave.

The creep rate measured after the stress reversal is in both cases (furnace to autoclave as well as autoclave to furnace) even higher than the primary creep rate of the fresh material, indicating a strong kinematic hardening. The experimental results will be presented in detail and an interpretation in terms of a mixed isotropic and kinematic hardening model will be proposed.

Poster 4.1: Experimental investigation and thermodynamic modelling of the in-vessel corium for severe accident studies in PWR reactors

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Abstract

During a severe accident in a PWR reactor, the oxide nuclear fuel (UO₂ or MOX) reacts at high temperature with the zirconium alloy clad and the steel vessel to form a partially or fully molten mixture so-called “in-vessel corium”. In such a case, the corium forms a pool in the bottom of the vessel, constituted of two liquid phases, metallic and oxide. The formation of such a configuration of the corium pool is due to the existence of a miscibility gap in the liquid state. The fractions and compositions of these liquid phases have to be well known in order to model the thermal hydraulic properties of the pool. The aim of the present work is to investigate experimentally the thermodynamic properties of the U-Zr-O-Fe system representative for the in-vessel corium to improve the thermodynamic modelling using the Calphad method. Laser heating techniques as well as heat treatments are used to measure solid/liquid transition temperatures and to highlight the miscibility gap in the liquid state. Experimental data are used to assess the thermodynamic properties of this key system. Both experimental and theoretical results will be presented.

Poster 4.2: Fuel chemistry of irradiated FBR mixed oxide fuel – Comparison between thermodynamic calculations and post-irradiation experimental results.

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Abstract

The mixed oxide fuel used in the Sodium-cooled Fast Reactors (SFR) undergoes high temperature level in the fuel pin with a large temperature gradient between the center and the periphery of the pellet. These extreme conditions are the cause of many thermal, mechanical and chemical transformations in the fuel. Moreover, the behavior of the fission products (FP) generated by the irradiation strongly depends on their chemical nature as well as conditions in the fuel (temperature, oxygen potential, ...).

Experimental observations show the formation of a layer located between the fuel and the cladding material for a burn-up of 6 to 8 ha%. This layer, called JOG for “Joint Oxyde-Gaine” [1] is due to the release, migration and association of caesium, iodine, and tellurium mainly with oxygen and molybdenum created in the fuel pellet. At high burn ups, these fission products (FP) compounds can react with the cladding components (Fe,Cr,Ni) corroding the inner part of the cladding material. The fuel and cladding chemical interaction is nowadays recognized as one of the major factors determining integrity and lifetime of pins in the SFR [2].

For all these reasons, it is important to model the physico-chemical behavior of the fuel pin under irradiation conditions typical of FBR mixed oxide fuel. The modelling of mixed oxide fuel pins has been recently improved by CEA with the development of the GERMINAL V2 fuel performance code [3] where the nuclear FP database of the neutronics module has been chained to a thermodynamic code, ANGE, advanced version of the SOLGASMIX software. In addition, in conjunction with the OECD/NEA project [4/5], thermodynamic modelling of fission product systems such as Cs-Te, Cs-I and Cs-Mo-O ... have been performed with the CALPHAD method and incorporated into the TAF-ID database.

Thermodynamic calculations on irradiated MOX fuels using ANGE code + TBASE on one side and by using Thermo-Calc and Open Calphad software [6] + TAF-ID database on the other side will be presented and compared to post-irradiation observations performed on $(U_{0.78}Pu_{0.22})O_{1.975}$ fuel samples irradiated to 13.6 ha% into the PHÉNIX reactor.

Next steps for the improvement of the thermodynamic description of the irradiated MOX fuel as well as the coupling between thermodynamics and the fuel performance code GERMINAL will be proposed and discussed.

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Poster 4.3: Characterisation of Essential Corium Subsystems Up To Melting

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Abstract

In the unfortunate event of a Nuclear Power Plant (NPP) core meltdown accident (cf. Fukushima, Chernobyl, Three Mile Island) a liquid radioactive mass containing nuclear fuel, cladding and containment materials, called “corium”, is formed. In-depth investigation of such phenomena is paramount for the a priori determination of the fuel’s safe performance limits and the prevention of such mishaps both in current reactors and in future even safer ones. In addition, the occurrence of segregation in the re-crystallised corium can lead to the formation of zones richer in fissile materials. This would result in additional safety and safeguard issues.

The present research project is focussed on the experimental study of solid/liquid equilibria and phase segregation in crucial material systems simulating the real corium. In particular, the system $\text{UO}_2\text{-ZrO}_2$ is revisited in this work by laser heating under controlled (reducing, oxidising) atmospheres coupled with fast optical thermometry. Newly measured phase transition points measured in an inert or reducing atmosphere are in fair agreement with the early measurements performed by Masset in 1953, the only study available in the literature on the

whole pseudo-binary system. However, a minimum melting point is more clearly identified here for the composition $(\text{U}_{0.6}\text{Zr}_{0.4})\text{O}_2$, at 2815 K. The solidus line is rather flat on a broad range of compositions around the minimum. It increases for compositions closer to the end members, up to the melting point of pure UO_2 (3130 K) on one side and pure ZrO_2 (2970 K) on the other. UO_2 -rich compositions are the most challenging because of the complex chemistry of uranium dioxide. In fact, uranium can assume different oxidation states (+3, +4, +5, +6) in its dioxide form, this feature allowing the formation of both hypo-stoichiometric and hyper-stoichiometric dioxides. The behaviour of uranium dioxide is therefore strongly atmosphere-dependent and differences can be noticed between the thermograms recorded when the sample is melted in an inert or reducing atmosphere (argon or $\text{Ar}+\text{H}_2$) compared to those recorded in an oxidising atmosphere (compressed air). In the last case, solid/liquid phase boundaries can decrease by several hundreds of K.

Solid state phase transitions (cubic-tetragonal-monoclinic) have also been observed in ZrO_2 -rich compositions with the help of XRD and Raman spectroscopy. The well-known Raman spectra of ZrO_2 are used to identify the different crystal lattice structures. The U and Zr valence states of samples melted in different atmospheres have been assessed with the help of X-ray absorption spectroscopy (XAS).

Additional systems have been explored, for example by substituting Th to U in the mixed dioxide, or by adding small amounts of PuO_2 to the pseudo-binary UO_2 - ZrO_2 in order to better simulate the reaction of a real MOX fuel with its oxidised cladding. Raman spectroscopy has been employed, in these particular cases, also for the identification of the hypothetically segregated PuO_2 hotspots upon fast cooling.

The present results are important for assessing the thermal stability of the system fuel – cladding in an oxide based nuclear reactor, and for simulating the system behaviour during a hypothetical severe accident.

Poster 4.4: Thermodynamic study on the speciation of CsI in nuclear fuel

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Abstract

The existence of CsI in nuclear fuel pellets has been one of debate. Cs and I and formed in a ratio of 10 to 1 during irradiation. According to thermodynamic studies CsI should be the most stable form of I present under the conditions that prevail during irradiation. However, up to date there is no clear experimental evidence to support the existence of CsI in nuclear fuel.

It is generally accepted that I does not exist in its elemental form in nuclear fuel. If Iodine exist in its elemental form, then it would exhibit similar behaviour to that of the noble gasses. Two arguments prove this not to be true. First, Iodine shows a different release during Knudsen cell experiment compared to noble gasses. Secondly, it does not migrate into the plenum region like the noble gasses do [1]. In addition, several arguments speak in favour of CsI a.o.: Thermodynamic studies show CsI to be the most stable form. X-ray fluorescences of Cs and I deposits on the internal cladding surfaces of commercial irradiated fuel rods has been found. And lastly, Cs and I show the same behaviour during iodine spike phenomena[2].

In this study, we aim to find a decisive answer to the existence of CsI. We do that by performing Knudsen-cell experiments on pure CsI, spark plasma sintered UO_2 -CsI (SPS-CsI) both stoichiometric and oxidised and compare the data to data from irradiated fuel. Would I be present as CsI in the fuel pellet than its release behaviour and diffraction pattern, would be similar to that of pure CsI and to that of SPS-CsI.

Preliminary results show that CsI might be present in the form of CsI. The ratios between the $(\text{Cs}^+)/(\text{I}^+)$, $(\text{Cs}^+)/(\text{CsI}^+)$ and $(\text{CsI}^+)/(\text{I}^+)$ peaks in the stoichiometric SPS-CsI sample are found to be similar to the ratios for irradiated fuel samples. This gives us the impression that CsI might be present in the form of CsI. In order to get full disclosure more analysis on different irradiated fuel samples will be done.

References:

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Poster 4.5: Thermodynamic Considerations in the Potential Use of Ni-Be Alloy to Braze Zircaloy-4

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Abstract

The construction of fuel bundle elements for CANDU® reactors involves the attachment of numerous small Zircaloy-4 appendages to the thin walled Zircaloy-4 fuel sheathing (cladding). These appendages provide inter-element spacing and improve coolant circulation thereby enhancing heat transfer. The outermost appendages, or bearing pads, provide the spacing between the fuel bundle and the pressure tube and facilitate the safe movement of the fuel bundles within the fuel channels. These appendages have been attached by a specialized brazing process using beryllium as the filler material.

To better control beryllium exposure during the manufacture of CANDU® fuel bundles, a nickel-beryllium brazing alloy to affix the appendages is being considered. A Ni-2wt%Be shows promise in substantially reducing beryllium volatility while minimally perturbing existing fabrication processes.

This work considers Ni-Be foil as a braze filler material. This choice eliminates the vapour deposition coating step, as well as any handling of pure Be in the fuel assembly plant. Avoiding both operations in a high production throughput workplace is a benefit. Furthermore, the use of foil as a means of applying the braze alloy is not a major modification to long established plant operations. Most important is that the alloying of Be with a high concentration of Ni reduces the Be partial pressure substantially during actual brazing (as distinct from the coating operation) without preventing in principle the chemical displacement of the thin ZrO₂ passive layer in spite of the considerable reduction in thermodynamic activity of Be due to nickel dilution.

Guidance on the potential use and benefits of Ni-Be braze foil is to be found in the proposed thermodynamic model of the Zr-Ni-Be ternary alloy system developed by interpolations from adaptations of treatments of the component binary systems. To this end, thermodynamic models of the condensed phases of the binary Zr-Be, Ni-Be and Ni-Zr alloy systems have been developed to provide a basis for the construction of a provisional ternary phase diagram that in particular identifies the domain of the liquid phase. The ternary model not only permits the computation of phase equilibria that bear on such essential

matters as melting but also provides thermodynamic properties that can be used in other computations on oxidation and partial pressure of Be.

Poster 5.1: Assessment of electronic structure methods for the investigation of nuclear fuel under irradiation: example of uranium dioxide

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Abstract

One challenge for the development of Gen IV nuclear reactors is to improve significantly the effectiveness of the design and selection of innovative fuels. To this aim, multiscale modelling approaches are developed to build more physically based kinetic and mechanical mesoscale models to enhance the predictive capability of fuel performance codes. Atomic scale methods, in particular electronic structure calculations, form the basis of this multiscale approach. It is therefore essential to know the accuracy of the results computed at this scale if we want to feed them into higher scale models.

Electronic structure calculation methods, especially density functional theory (DFT), have been used extensively on molecular and solid systems during the last thirty years. Numerous assessments of these methods have been performed, which show that they are powerful tools yielding precise and predictive results for a large number of solid and molecular systems, therefore contributing to the understanding of numerous phenomena. The application to nuclear materials under irradiation and especially to fuels, however, is more delicate and calls for bespoke developments. A specific assessment of the atomic scale methods for the description of nuclear fuel under irradiation is therefore necessary.

We will present the result of the extensive assessment effort of the results of state-of-the-art electronic calculations on uranium dioxide performed in the “Working Party on Multiscale Modelling of Fuels and Structural Materials for Nuclear Systems (WPMM)” of the OECD/NEA.

Poster 5.2: Positron annihilation spectroscopy study in UO_2 : coupled theoretical and experimental approach

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Abstract

One of the experimental methods that can be used to study open volume defects is positron annihilation spectroscopy (PAS). This non-destructive technique consists in recording the radiation emitted at the beginning and the end of life of positrons in a material and deducing the properties of the positrons and the electrons with which they have annihilated. There are two positron annihilation characteristics that allow one to detect defects in materials: The first one is the lifetime of positrons in the material is mostly sensitive to the open volume of the defects. Second, the Doppler broadening of the annihilation radiation gives information on the chemical environment in which positron annihilate. These two characteristics can be measured experimentally. To identify the types of defects present in studied materials, however, comparison with other characterization methods or calculations is necessary.

Positron lifetime and Doppler broadening can be calculated using the two-component density functional theory. We performed self-consistent calculations of positron characteristics for vacancies and vacancy clusters in uranium dioxide using the implementation of the two-component density functional theory in the ABINIT code. The comparison of theoretical results with experimental measurements obtained for irradiated uranium dioxide will be presented along with the proposed identification of the observed signals.

Poster 5.3: Analysis of U L3-edge X-ray absorption spectra for uranium dioxide based on molecular dynamics simulations

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Abstract

Uranium L3-edge X-ray absorption spectroscopy is used to study the atomic structure of uranium dioxide (UO₂). The extended X-ray absorption fine structure (EXAFS) is interpreted within the ab initio multiple-scattering approach combined with classical molecular dynamics (MD) to account for thermal disorder effects. This MD-EXAFS approach allows for a direct comparison of simulated and experimental EXAFS spectra. Several force field models are assessed in this way and the role of multiple-scattering contributions is evaluated.

Poster 5.4: Defect characterisation in atomistic simulations

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Abstract

Atomistic simulations of nuclear materials often involve creation, recombination and transformation of defect structures. Such defects can be of various shapes, sizes and kinds, from isolated vacancies or interstitial atoms to large clusters, voids, dislocations and grains boundaries. To understand the behaviour of these defects and their consequences on other material properties, they need to be properly detected, isolated and characterised. A technique will be shown, based on the statistical analysis of the considered atomistic configuration, which allows the creation of a graph representing the topology of the crystal. This graph can be processed to automatically determine the positions of point defects and clusters, and perform higher-level analysis, like grain boundary or dislocation detection. This technique is suitable to the analysis of complex microstructures like dislocations and poly-crystals in large-scale simulations even at high temperature.

Poster 5.5: Effects of stoichiometry on the defect clustering in uranium dioxide

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Abstract

The study addresses the on-going topic of point defects and point defect clusters in uranium dioxide. Molecular statics simulation using an extended pair potential model that accounts for disproportionation equilibrium as charge compensation has been applied to assess the effect of disproportionation on structural properties and clustering in non-stoichiometric uranium dioxide. The defective structures are scanned in minute detail using a powerful and versatile analysing tool, called ASTRAM, developed in-house for the purpose. Unlike pair potential models ignoring disproportionation effects, our model reproduces volume changes observed experimentally in non-stoichiometric $\text{UO}_{(2-x)}$ and $\text{UO}_{(2+x)}$. The oxygen defect energetics computed is in good agreement with data in the literature. The model is used to assess the clustering that occurs in bulk samples of non-stoichiometric uranium dioxide. This study confirms the generation of split-interstitial clusters as the dominant defect type in non-stoichiometric uranium dioxide. A new key mechanism for defect clustering in hyper-stoichiometric uranium dioxide is proposed that is based on the progressive aggregation of primitive blocks identified as 1-vacancy split-interstitial clusters.

Poster 5.6: Atomistic modeling of Xe adsorption on UO_2 surfaces

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Abstract

Uranium dioxide UO_2 is used as a standard fuel in pressurized water reactors (PWR). During fission reactions in uranium intragranular bubbles of xenon are generated. The presence of these bubbles modifies the thermomechanical properties of the fuel. The need to characterize these effects led to an extensive work both from experimental and theoretical points of view.

It is known from the literature that these bubbles are microfaceted, with (111) and (100) surfaces. We then study here simplified systems of xenon on semi infinite UO_2 surfaces. In a first step, we assess the relative stability of UO_2 surfaces according to their orientation and then to their polarity, by mixing

thermostatistical relaxation and analytic formulations within a simple electrostatic model. The main result is that, whereas the (111) surface appears stable by construction and does not involve major reorganization, the polar (100) one is only stabilized through drastic rearrangement of the surface region.

In a second step, we proceed to xenon adsorption on these relaxed surfaces through Monte Carlo simulations in the grand canonical ensemble within semi-empirical potentials. The most striking feature revealed by the simulation is the existence of a phase transition from a dilute xenon phase towards a dense one. Structure analysis of the dense phase indicates a coexistence of the FCC, BCC and HCP structures. Otherwise, the xenon density is found to increase with the temperature for a given chemical potential.

In a third step, the pressure inside the xenon bubble and in the UO_2 matrix has been investigated. In the former case, we show that whatever the xenon structure may be, the pressure increases with the xenon density, but not with the temperature for a fixed density. Concerning the UO_2 matrix, we present pressure profiles before and after xenon adsorption. The next step will be to introduce these results in a micromechanical model, which will allow to derive a thermomechanical behavior law for the porous UO_2 .

Poster 5.7: Ab initio investigation of thermodynamic properties of corrosive fission product compounds in MOX fuels

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Abstract

The ability to predict the onset and extent of internal corrosion in fuel pins is needed for safe and economical nuclear reactor operation. GERMINAL V2 is a fuel performance code developed by CEA, EDF, and Areva dedicated to simulating the behaviour of mixed oxide (MOX) fuel under irradiation in sodium cooled fast reactors (SFRs). In order to improve the current internal corrosion prediction capabilities, efforts are made to develop and implement a new model based on thermodynamic properties of compounds consisting of certain fission products suspected to play a role in the corrosion process of the cladding material. Using ab initio simulations in the framework of density functional theory, thermodynamic properties and crystal structures of binary compounds containing

Te have been investigated. Special attention has been given to $\text{Fe}_{(1.11)}\text{-Te}$ since more data for this compound is needed in order to better describe the Fe-Te phase diagram in the range surrounding FeTe using the CALPHAD approach.

Poster 6.1: Phase-field model to simulate pore morphology, migration and consequences in MOX fuels

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Abstract

A phase-field model is developed to simulate pore morphology, migration, and consequences in MOX fuels. The model describes pore evolution driven by vaporization of matrix material at the hot side of the pore and condensation on the cold side, propagating the pore up the temperature gradient. Non-congruent vaporisation of the multi-component matrix constituents redistributes U and Pu, resulting in enrichment of Pu towards the fuel centreline in MOX fuels. The local O/M has a strong effect on pore migration due to large variations in UO_3 partial pressure. The gas inside the pore is not in mechanical equilibrium with the surrounding matrix, and the pore is observed to assume a lenticular shape with the minor axis oriented approximately parallel to the temperature gradient.

The model couples multi-component, multi-phase diffusion, structural mechanics, and interfacial energy. The simulation is used to examine classical theories for pore migration velocity as a function of temperature and temperature gradient, which provides information on the extent of the columnar grain zone and central void.

Poster 6.2: Modeling of SPHERE irradiation with TRANSURANUS fuel performance code

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Abstract

The SPHERE irradiation was conducted in the High Flux Reactor in Petten, The Netherlands, and has run for 11 cycles between August 2013 and April 2015, achieving in total 295 Full Power Days (FPDs). The goal of the SPHERE irradiation is to study the performance of MOX-fuel, bearing 3% americium (Am) in two forms: regular pellet type fuel and a sphere-pac fuel. The nominal linear power in two SPHERE pins containing both types of fuel was ~ 300 W/cm, and the burnup was close to 5% FIMA.

In this work the actual irradiation history, together with the temperature readings was used to model the fuel behavior with the TRANSURANUS fuel performance code. The results of the modelling, in terms of the fuel central temperatures, fuel restructuring and central hole formation are compared to the neutron radiography images obtained at the end of the irradiation. The modelled fission products distribution (axial) and fission gas production and release are compared to the results of non-destructive post-irradiation examination: gamma scan, fuel pin puncturing.

Poster 6.3: Numerical simulation of the melt relocation behavior in fuel assembly scale structures

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Abstract

We have been developing the numerical simulation method for the melt relocation for severe accidents in a fuel assembly scale. The purpose is to deeply understand each processes of the core melt progression which has not enough accuracy for numerical analyses and is to contribute the improvement of severe accident analysis codes and debris state analysis in the Fukushima accident. For example, in the process of the fuel degradation of the BWR, since the understanding for the reaction process between molten fuel and core support plate

might not be enough, we are developing the numerical simulation code with experiments in simplified system. The numerical simulation code is based on a unified, phenomenological, and multiphase thermal-hydraulic simulation models. In this paper, the developed code was applied to numerical simulations of the melt behavior of the complicated fuel support structures, e.g., fuel support piece, control rod and channel box, and also applied to the melt relocation experiment as an validation of our code. From the present numerical results, the code stably worked in the complicated structures and showed in good agreement with the results of the experiment. Therefore, it was confirmed that the developed code has potential to accurately understand melt relocation behavior inside an RPV. In the near future, in order to perform the more detail simulation, we will implement the eutectic reaction model to the code and also proceed the experiment analysis.