NuFuel 2023

Research into Nuclear Fuel in Europe

Marseille (France), November 7-9

Book of abstracts

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ORAL TALKS

A digital twin for a separate effect experiment about inert gas behavior

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Digital twins are a cutting-edge methodology in the field of modelling and simulation. Compared to classical digital models, they can adapt to a specific physical system and to provide online feedback to the physical system itself. When applied to separate effect experiments concerning fission gas behaviour in nuclear fuel, the digital twin methodology requires specific software developments and careful evaluation of assumptions concerning data availability and definition of the system digital state. In this work, a showcase of the methodology is demonstrated, with the goal of highlighting benefits and limitations compared with standard modelling approaches. The physical systems considered are the annealing experiments by Talip and co-authors, which present online measurements of helium release rate. The digital twin is the SCIANTIX code, which includes a physics-based model for helium behaviuor. The use of a past experiments for this application is done as if the experiments are happening right now, which in turn showcases the viability of using available experimental information to test the performance of new simulation methodologies.

Preliminary Extension of OFFBEAT to TRISO Fuel

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To simulate the behaviours of TRISO during reactor operation, the multi-dimensional fuel performance code OFFBEAT (OpenFOAM Fuel Behaviour Analysis Tool) has been extended for the TRISO case. The material properties of porous carbon buffer, PyC (Pyrolytic Carbon) and SiC (Silicon Carbide) from PARFUME have been implemented into the code, as well as their behaviour models, including anisotropic irradiation dimension change, thermal expansion and creep. The original UO2 models already present in OFFBEAT were used to simulate the material properties, densification and swelling of Kernels. Additionally, a dedicated gap plenum model was used to account for the gap formed by the shrinkage of buffer and PyC, as well as buffer's porosity. The Proksch model was utilized to compute CO(Carbon Monoxide) produced by the reaction of oxygen and carbon. By incorporating the gap plenum model, CO production model and mechanism model of fission gas release, the gap behaviour, internal pressure and heat transfer between buffer and inner PyC can be simulated.

After the extension of new models, a 1-D model of a TRISO particle has been built with wedge boundary conditions along transverse directions and it has been used to verify the code against the IAEA CPR-6 benchmark cases. The results are in line with the predictions of other codes, suggesting that OFFBEAT could be a promising tool to accurately simulating the performance of TIRSO under normal conditions. Currently, efforts are focused on 3-D simulations of TRISO. Future developments will include studying the diffusion of fission products, debonding between different layers, as well as the multi-dimensional and multi-scale coupling between TRISO particles and the pebble bed.

Thermo-mechanical and fluid simulation of granular media: developments and applications to nuclear fuels

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During its life cycle, from manufacturing to recycling, the nuclear fuel of PWR reactors goes through a large variety of steps. It might be in a divided shape and considered as a granular media during hypothetical accidental conditions like LOCA or RIA or during the manufacturing process of fuel pellets. Numerical simulations allows more detailed study than it is possible using experiments. It allows to investigate phenomenon or to make pre-test of the process. The Discrete Element method (DEM) is well-known as an efficient tool to simulate the mechanical behaviour of granular media. In relation with the PLEIADES framework, we present a set of scientific computing tools based on DEM to analyse the thermo-mechanical behaviour of immersed granular material applied to the nuclear fuel by means of a chained coupling between DEM method (using Rockable code) and a Fast Fourier Transform method (TMFFT) using voxelization technique (Merope code) [1,2]. This approach gives access to effective properties of fragment beds like the thermal conductivity. In addition, we coupled the Discrete Element Method with the Lattice Boltzmann to compute the fluid-grain interactions and their thermal behaviour for relocation and dispersal in accidental conditions [3].

1. https://github.com/MarcJos/Merope

2. A DEM/FFT approach to simulate the effective thermal conductivity of granular media. Tristan Calvet, Jean-Mathieu Vanson, Renaud Masson. 2022, International Journal of Thermal Sciences, Vol. 172, p. 107339.

3. LBM/DEM simulation of heat transfer in granular suspensions. Lhassan Amarsid, Jean-Yves Delenne, Jean-Mathieu Vanson, Bruno Collard, Philippe Sornay, Farhang Radjai. [ed.] Powders and Granular Materials Challenges and Future Trends. 2019.

Thermodynamic-based modelling of fuel melting in the fuel performance code ALCYONE

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In recent years, with the development of open-source thermochemical solvers such as OpenCalphad, fuel thermochemistry has been included in fuel performance simulations and in particular in ALCYONE [1], the code co-developed by the CEA, EDF and FRAMATOME within the PLEIADES computational environment. While fuel thermochemistry was initially used to assess the minor and chemically reactive fission gases (mostly iodides) released from the fuel pellet during slow power transients [2], it has recently been used to analyse fuel melting during the severe accident simulated sequences of the VERCORS-VERDON test series [3]. Irradiated fuel melting in oxidizing or reducing conditions with or without strong interactions with the Zircaloy cladding was assessed using the Thermodynamics of Advanced Fuels International Database (TAF-ID [4]), developed within an international OECD project.

In this paper, thermochemical calculations using the TAF-ID are applied to irradiated UO_2 fuels of varying fission product contents to assess the dependency of the liquidus temperature on the fuel burnup. The definition of the solidus temperature from calculations performed considering all the fission products or the fluorite phase only (including soluble rare earth) is then discussed. The thermodynamic-based solidus temperatures are shown to be in reasonable agreement with the recent burnup-dependent correlation proposed in the ESNI++ catalogue [5] from a selection of measurements on irradiated UO_2 and MOX fuels.

The usefulness of a thermodynamic-based modelling of fuel melting is then demonstrated by simulations of two power transients studied during the Power to Melt and Manoeuvrability (P2M) Simulation Exercise [6] of the Framework for IrraDiation ExperimentS (FIDES) international project, managed by the OECD. The extent of partial or total fuel melting is calculated with ALCYONE fuel performance code using the precise assessment of the solidus and liquidus temperatures provided by the embedded fuel thermochemical calculations. Simulation results are discussed and compared to available post irradiation examinations (ceramography, EPMA, SEM).

[1] Introïni, C. et al., (2020), Nuclear Engineering and Design, 369.

[2] Baurens, B. et al. (2014), Journal of Nuclear Materials, 452.

[3] Germain, A. et al. (2022), Journal of Nuclear Materials, 561.

[4] Guéneau, C. et al. (2021), Calphad, 72.

[5] Magni, A. et al. (2020), Tech. Rep. D6.2, v2, INSPYRE.

[6] D'Ambrosi, V. et al. (2023), ANS Nuclear Technology, In Press.

SFR fuel cracks modelling using phase field approach

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During the first power increase of a Sodium-cooled Fast Reactor (SFR), primary radial cracks form in the fuel pellets, induced by the thermal gradient. A phenomenon of healing of these cracks then occurs, depending upon the thermal conditions. Thus, the pellet alternates between cracking and healing during the history of loading cycles.

The goal is to predict the crack network of a cylindrical pellet under SFR irradiation conditions. This network impacts at first order the free volume distribution measured after irradiation.

In the present paper, a local damage-healing model is developed to describe this chemo-mechanical process. In addition to the classical damage variable, a variable associated with a "healing reserve" is introduced. The latter allows the recovery of the damage variable while ensuring a positive dissipation of the system. The results are 2D simulations on the basis of a cohesive zone model. Results illustrate the interest of such a model in the treatment of healing of damaged materials. In perspective, a model calibration could be proposed on the basis of image analysis.

Reduced order modelling of fission gas diffusion in fuel performance codes

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During the first power period of operation, fast reactors experience a radical alteration of fuel micro-structure driven by high temperature and steep temperature gradient, with consequent formation of columnar grains. It is common in fuel performance codes to model fission gas diffusion in the columnar grain zone as if fuel grains were spherical, considering a representative radius of cylindrical geometry and assuming a uniform temperature neglecting the temperature gradient. The different kinetics of diffusion due to the temperature gradient makes the classical approach unsuitable, resulting in inaccurate estimation of gas release. To incorporate temperature gradient effects in fission gas modules of conventional fuel performance codes, in a recent work (Pizzocri et al., submitted to NET, 2023) we proposed an algorithm based on reduced order model to solve the diffusion problem in both spherical and cylindrical geometries. We implemented this algorithm in the grain-scale code SCIANTIX and verified it against the state-of-the-art solutions. In this work, we present the applicability of the proposed model to integral fuel rod analysis by using the TRANSURANUS fuel performance code coupled with SCIANTIX. The effectiveness of the model is evaluated by comparing the simulation results with two irradiation experiments from the IFPE (International Fuel Performance Experiments) database. Specifically, the simulation of the GE7 fuel rod irradiation experiment is used to assess the model for spherical grains, whereas the validation database from the SUPERFACT-1 fast reactor irradiation experiment is employed to assess the model for cylindrical grains. The evolution during irradiation of quantities of engineering interest (e.g., the concentration of gas in fuel grains and fission gas release) is considered for the assessment of simulation results. The outcome represents a significant advancement in the description of fission gas release under fast reactor irradiation conditions.

Mesh Free Methods: Uses and application to Nuclear Fuel

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Nuclear fuel within a reactor represents one of the most complex physical environments known to man. The creation of fission products combined with the high energy environment causes material properties to change and gas to migrate and form micro bubbles within the fuel. This leads to large scale material cracking and deformation of the fuel causes Pellet Cladding Mechanical Interaction (PCMI) which can cause cladding materials to fail. It is therefore important to consider these phenomenon when simulating fuel for commercial licencing. Finite Element Methods (FEM) are currently widely used within fuel performance modelling. Over recent years alternative methods based instead on integral formulations have increased in popularity for industrial problems due to there ability to handle cracking and large scale deformations. This talk aims to outline how one such method, Smooth Particle Applied Mechanics (SPAM), works and show results of SPAM thermal fuel and cladding simulations. This work was completed as a PhD at University of Sheffield with Dr K. Travis, with Dr M. Bankhead from NNL.

Implementation of multi-dimensional transport solvers in OFFBEAT

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Several transport and diffusion mechanisms can play an important role in determining the behaviour of the nuclear fuel. For example, the redistribution of minor actinides towards the center of the pellets, which typically occurs in MOX fuel used in Fast Reactors (FRs), can significantly affect the thermal properties of the pellet, notably its conductivity. In similar conditions, an evaporation-condensation mechanism at the pore surfaces drives the movement of pores towards the pellet center which can lead to the formation of a central void. In the cladding, the diffusion and precipitation of hydrogen atoms in the cladding, produced as a by-product of the water-side corrosion, can lead to excessive embrittlement, endangering the integrity of the rod.

Investigating these transport phenomena in a multidimensional environment offers valuable insights into various aspects of fuel behaviour. For instance, understanding the effects of axisymmetric irradiation conditions, power distribution and heat transfer on these processes is crucial for optimising fuel performance in complex scenarios. As a first effort toward this aim, we present in this work the implementation of three transport solvers in OFFBEAT, the open-source fuel performance code developed at the École Polytechnique Fédérale de Lausanne (EPFL) in collaboration with the Paul Scherrer Institut (PSI) in Switzerland. OFFBEAT is a 3D finite-volume code based on the open source C++ library OpenFOAM developed since 2017 with the objective of enhancing the understanding of poorly-known multidimensional mechanisms underlying local effects.

The first transport solver based on the work of reference [1] treats the redistribution of Plutonium and Americium isotopes within MOX fuel pellets. The hydrogen transport solver is based on the same methodology, and (based on the work of [2]) follows the diffusion of hydrogen atoms in solid solution within the cladding under a concentration or temperature gradient. The precipitation and dissolution of solid hydrides are also accounted for, based on the latest experimental results, and employing models available in the literature [3,4] for the kinetics of hydride nucleation, growth, and dissolution. Finally, the pore transport solver implemented in OFFBEAT solves an advection-diffusion equation to model the pore migration over a temperature gradient [5].

As the physics modelled by the transport solvers might require small time steps for obtaining accurate results, dedicated time-stepping algorithms have been implemented, allowing for flexible control of the simulations based on the specific requirements of each modelled physics. Also, to assess their performance, all transport solvers have been tested against verification cases and, whenever possible, validation cases. While the obtained results demonstrate the accuracy and reliability of the implemented solvers, future work will focus on verifying and validating the transport solvers within multi-dimensional test cases.

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[2] O. Courty, A.T. Motta, J. D. Hales, Modeling and simulation of hydrogen behavior in Zircaloy-4 fuel cladding, J. Nucl. Mater. 452, 2014, 311

[3] E. Lacroix, A.T. Motta, J.D. Almer, Experimental determination of zirconium hydride precipitation and dissolution in zirconium alloy, J. Nucl. Mater. 509, 2018, 162

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Peridynamic Modelling Of Cracking In Triso Coated Particle Fuel For High Temperature Reactor (HTRs) – The Importance Of Residual Stresses To The Crack Patterns Formed

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TRISO coated particle fuel is proposed as the fuel concept for high temperature reactor fuels. They consist of a fissile kernel of material surrounded by a porous graphite buffer; a dense inner-PyC layer; a SiC layer; and, a dense outer-PyC layer. During the initial rise to power and subsequent rise to power, a complicated thermal-mechanical load is applied to these materials. This is complicated further by residual stresses generated during particle manufacture and further again by differences in the creep and swelling rates between the materials. In the absence of cracking, finite-element based multiphysics models are able to predict the loads in these materials. They are not however able to capture the initiation and subsequent behaviour of cracks due to these loads. Peridynamics is a developing structural mechanics implementation which moves away from the limitations of finite element analysis. It is able to model crack initiation, propagation, branching and coalescence without prior knowledge of the crack patterns to be expected. In this work, we apply the residual strains predicted by a finite element model created in Abaqus to a peridynamics model for a TRISO fuel particle by applying swelling strains to the bonds in bond-based peridynamic simulation.

Calculating the safety margins of the AGR-like FHR

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This ongoing project proposes to adopt the Advanced Gas cooled Reactor (AGR) geometry and its pin-type fuel in combination with the molten Fluoride salt-Cooled High Temperature Reactor (FHR) concept to create a novel “AGR-like FHR” concept to achieve better economic performance and speed up the development of AGR-like FHRs [1].

This work uses the AGR fuel assembly geometry and parameters (power and coolant outlet temperature) as baseline and FLiBe molten salt as coolant. Using salt as coolant results in a higher average temperature with lower temperature gradient across the core. The chosen material for the cladding is Hastelloy-N due to its demonstrated favourable performance at high temperatures and in corrosive environments. Previous work performed on the design of the AGR-like FHR concept have studied different configurations based on a reference AGR fuel assembly that could potentially achieve a better performance than AGRs while complying with the required safety limits. However, none of these studies included fuel performance over a burnup cycle as a consideration in the analysis and exploration of the design space. Consideration of fuel behaviour suggests that the most limiting parameter is cladding creep given the low pressure of the salt coolant. For this reason, in this work, a series of tests have been performed to estimate the safety margins to rupture and excessive deformation of the cladding using the LMP (Larson Miller Parameter) criterion. Since the cladding material is not validated for nuclear purposes, the LMP limiting curve is obtained from existing Hastelloy-N data. By calculating the safety margins, the feasibility of the AGR-like FHR using the same AGR geometry will be tested and the necessary modifications to the total power and coolant temperature will be performed.

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Flash sintering of $(U,Ce)O_2$ pellets as a surrogate for MOX fuel

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The UK retains a keen interest in the manufacture of mixed uranium-plutonium dioxide (MOx) materials as a reactor fuel for either thermal or fast neutron systems, and additionally is being considered for future disposal concepts (Disposal-MOx). Recently, there has been interest in studying the suitability of advanced, low-temperature sintering techniques for the manufacture of mixed uranium-plutonium dioxide (MOx) fuel within the UK. Part of the challenge the UK faces is utilizing the unique elemental composition of the UK civil Pu stockpile. The progressive formation of highly volatile and long-lived isotopes, such as americium, means that conventional sintering techniques, in which cold pressed MOx greens are sintered in reducing atmospheres at 1700°C with long hold times, cannot effectively retain Am species during manufacture. This presents issues for plant design and potential safety issues that could impact cost effective utilization of historic UK plutonium stocks.

Flash sintering is a technique in which an electric field is passed through a green body during sintering, leading to densification at reduced furnace temperatures and hold times compared to conventional sintering. As a potential manufacturing route, flash sintering has undergone significant development since its inception in the last decade [1] and has demonstrated an ability to greatly reduce energy and time costs for pellet manufacture. Currently there is little understanding about how the rapid pellet densifications achieved by flash sintering will affect UO_2 and MOx fuel pellet quality, and how critical fuel properties encompassing in-reactor behaviour to backend fuel cycle considerations will be impacted. Furthermore, vast challenges still exist to prove that such a manufacturing route can be commercially implemented, and whether this can be done whilst maintaining repeatability.

The author will be presenting the current summary of work comparing flash sintered (FS) surrogate $(U,Ce)O_2$ pellets, manufactured using the MIMAS and SBR powder processing routes, against conventionally sintered (CS) material. The work studies the differences in pellet properties between FS and CS materials using XRD, BSE imaging, Raman Spectroscopy, TGA and EDS mapping. Results from the aforementioned techniques will be presented to give an understanding of critical pellet qualities including density, grain size and stoichiometry, granting an early appreciation of how sintering kinetics are influenced by the varying techniques. Particular attention will be payed to elemental and crystallographic mapping techniques to demonstrate how Ce distribution is influenced by sintering and power processing techniques in finished pellets, and whether the resultant heterogeneity observed is significant enough to influence local redox chemistry & stoichiometry.

[1] Cologna et al. 2010

Fabrication of MOX disks with controlled porosity

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In the framework of the European project ESFR-Simple, several series of porous MOX disks $(U,Pu)O_{2-x}$ with Pu/(U+Pu) = 28.4%) were performed at CEA Marcoule in the Atalante facility with relative densities ranging from 95% (for the reference, without porogen) to about 70% for the most porous disks. The porosity was obtained by incorporating an organic porogen into the milled MOX powder.

Geometrical specifications on the sintered objects (diameter, height) had to be respected to allow thermal property analyses without any rectification step. These measurements of thermal properties (melting temperature, thermal diffusivity) are planned at the JRC Karlsruhe from 2024. The results will allow us to refine the MOX behaviour laws to improve the modelling of thermal properties. Moreover, various characterizations were performed in Atalante (density, O/M ratio, SEM, microprobe, Raman spectroscopy).

Oxidation studies of UN/UB_2

Megan Pritchard, Tim Abram, Joel Turner

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Accident tolerant fuels (ATF) may have the potential to enhance safety and lower costs in nuclear power stations by removing the need for safety systems by providing inherent safety with improved cladding materials. It may be possible to offset increased costs with a fuel material that has a higher uranium density, reducing the required enrichment or extending fuel life. Currently there are challenges facing promising fuel materials for light water reactors (LWR) due to their reaction with high temperature steam.

This work is focused on improving the behaviour of a key long-term UO_2 replacement, uranium mononitride (UN). It explores the effect of light element concentration on UN hydrolysis. A composite UN fuel containing UB_2 has been manufactured - this appears to improve the hydrolysis behaviour of UN when co-sintered. The UN and UB_2 powders have been milled using three different milling methods before sintering and characterised to understand the effects of UB_2 on UN sintering behaviour and oxidation performance. Differences in microstructure have been observed for the three milling methods, oxidation studies will confirm if microstructure has an effect on the reaction with high temperature steam.

Investigating the mechanisms of grain growth additives incorporation in UO_2 fuels

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Large-grain Cr-doped UO_2 fuels are regarded as an innovative pellet design for the United Kingdom next-generation reactors because they offer improved thermochemical and thermomechanical properties when compared to standard UO_2 fuels. One of our previous studies on the solubility of additives in UO_2 was based on the observation that Cr_2O_3 is more soluble in UO_{2+x} than in stoichiometric UO_2. In this study, we are focussed on understanding the mechanisms by which the Cr-dopant incorporates into the hyperstoichiometric UO_{2+x} lattice structure and impacts sintering and operational behaviour.

One proposed mechanism, but with limited data in the literature, is that in a mixture of Cr_2O_3 and UO_{2+x}, the Cr tends to form $CrUO_4$ complex oxide, which then acts as a grain growth initiator. To validate such mechanisms, a separate effect study was carried out to first synthesise $CrUO_4$ and other known grain growth precursors such as $AlUO_4$ and a mixed (Al, Cr)UO_4 in laboratory settings using co-precipitation method. The local structure and surface morphology of these complex oxides were quantified using a suite of experimental tools in the Bangor University Fuel Fabrication Facility (BUFFF) a National Nuclear User Facility (NNUF). The oxide precursors were then doped at different concentration in stoichiometric UO_2, and a variety of advanced, including in situ characterisations were carried out to understand the doped fuel sintering behaviour as well as the changes that occur along UO_2 grain boundary structures.

Synthesis of Uranium nitride from citrate sol-gel method

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It was recently shown that the calcination at 1000°C in Ar of a gel formed from the reaction of uranyl nitrate and citric acid yields nanometric UO_2 clusters, completely embedded and well-dispersed in a carbon matrix. Uranium carbide powders with high sinterability are then easily obtained from the carbothermal reduction of such precursor by a further heat treatment in vacuum at temperatures of 1200-1600°C for few minutes. [1, 2]

Here we report the first results of our investigation on the possibility to obtain the nitride form from the same precursor, by conducting the last step in a nitrogen-containing atmosphere, and on the granulation of such powders by a novel electro-dropping method. The work is being performed within the EU-funded FREDMANS Project.

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Irradiation test design for actinide-bearing chloride salts in the HFR

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Under Task 4.5 of the European MIMOSA (MultI-recycling strategies of LWR SNF focusing on MOlten SAlt technology) project, NRG, together with the EU Joint Research Centre, Orano and Thorizon, is developing a conceptual design of a capsule to irradiate actinide-bearing chloride salts in the High Flux Reactor (HFR) in Petten, The Netherlands.

Four fuel salt capsules are foreseen to be irradiated in the 500 °C - 800 °C temperature region, in a $6E^{13}$ n/cm^{2}s thermal fluence rate (E<0.625eV). The general scope of the irradiation test will be to assess fission product migration (fission gas release, fission product evaporation-condensation) and temperature gradient driven corrosion. An additional goal is to collect irradiated salt for post-irradiation property measurements.

The foreseen fission product and corrosion behaviour evaluations will yield unique data that are vital in advancing the technology readiness level of chloride salt-fuelled reactor technology for the multi-recycling of actinides in spent fuel.

In this contribution the concept design of the salt capsule irradiation carried out within MIMOSA will be discussed, including salt power and fluid flow calculations.

The design will benefit from the previous experience with the irradiation of actinide-bearing fluoride salts obtained by NRG and the EU Joint Research Centre under the SALIENT-01 irradiation.

Peridynamics Modelling of TRISO Coated Particle Fuel, a Comparison between 2D and 3D Models

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TRISO coated particle fuels are an advanced technology fuel developed as a solution for conventional and high temperature gas reactors, with the latter offering the possibility of nuclear co-generation powered industrial facilities and hydrogen production, both technologies that could provide important contributions towards net-zero.

Over the last few decades, an increasing number of fuel performance codes has been developed to simulate the stresses arising in TRISO particles during the thermal transients occurring inside nuclear reactors. However, most of the models available in literature are based either on analytical solutions in spherical symmetry or finite element modelling, both of which are not by design suited to deal with the most typical failure mechanism occurring in TRISO particles, that is the initiation and propagation of cracks in the silicon carbide layer due to the harsh conditions arising in the particle during operation.

A bond-based Peridynamics model has been developed and implemented in the finite element code “Abaqus” in order to simulate the behaviour of particles during thermo-mechanical transients. Thanks to the non-local formulation of Peridynamics, this model can autonomously predict crack formation and propagation without the need for fundamental modifications to the solution strategy, usually required by most of the available codes to deal with fracture mechanics.

Due to the generally higher computational requirements for Peridynamics, the development of this model historically focused on 2D implementations, which present some limitations, such as the approximation used to model the elastic response of the materials in TRISO (plane strain or plane stress). The work proposed in this project aims at verifying that the approximations used in the 2D models adopted in the past are not significantly affecting the capability of the code to predict fracture initiation and propagation in TRISO particles.

A model for athermal fission gas release in SCIANTIX

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The rising interest in low temperature applications of nuclear fuel, e.g., in small modular reactors, calls for the description of athermal fission gas behaviour. In this work, a physics-based modelling of the athermal fission gas behaviour is presented, extending mechanistic models available in the open literature. Athermal release is described as the fraction of gas vented from fuel through its open porosity. This fraction is accounted for via a semi-analytical description of the gas concentration gradient in the proximity of grain edges. Such semi-analytical results are included in SCIANTIX thanks to a dedicated neural network. This athermal release model is complemented with a semi-empirical model for solid swelling, which is also relevant in low temperature conditions compared to gaseous swelling. The behaviour of the model is assessed in the SCIANTIX code through the comparison with experimental results from Baker and Kashibe.

Chromia doped UO_2 fuel: Modelling of chromium solubility and fission gas diffusivity in SCIANTIX

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Among the current developments of improved nuclear fuel is the modification of the microstructure of the fuel pellet by increasing average grain size. This aims at improving the performance of the fuel at high burn-up, where it is mainly limited by the behaviour of gaseous fission products. In this work, we review models for the solubility of chromium in UO_2, and the evolution of the chromium phases in the fuel matrix during irradiation. These models were implemented in SCIANTIX, an open-source code to describe inert gas behaviour in nuclear fuel. We adjusted the chromium solubility model keeping each parameter within its range of compatibility with experimental data, targeting a better representation of available EPMA data of chromium content in fuel after irradiation. As for fission gas behaviour, we considered a physics-based description of the chromium impact on the fission gas diffusivity in fuel grains. The expression for the fission gas diffusivity in standard non-doped uranium oxide was corrected by adding the impact of the concentration of defects introduced by interstitial oxygen excess representing the effect of chromium content in the fuel itself.

Plastic deformation of uranium dioxide at high temperature: modelling of the single crystal plastic anisotropy

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Uranium dioxide (UO_2) is the primary fuel material of nuclear power plants. The understanding of its mechanical behaviour is of prime importance, especially in the context of accidental operating conditions of the reactor, where the cladding tube can have a mechanical interaction with the pellet. This study aims to build a dislocation-based crystal plasticity model that can reproduce specific features of UO_2 single crystal including the plastic anisotropy observed around 1600 K [1]. First, various hypotheses about the slip system activation are considered including dislocation glide in the ½<110>{100}, ½<110>{110}, and possibly ½<110>{111} slip systems. In this model, the constitutive law parameters are determined by adjusting the temperature dependency of the experimental critical resolved shear stress for the various slip modes. At first order, results show significant discrepancies with the compression stresses measured in the experiments, and more particularly for orientations where ½<110>{111} slip is observed highlighting the complexity of UO_2 plasticity processes. Secondly, integrating a detailed description of the dislocation-dislocation interactions computed using discrete dislocation dynamics only partially improves the model's reliability. Finally, we shall see that the composite slip hypothesis that relies on dislocation cross-slip between slip modes allows for the complete description of UO_2 single crystal plastic anisotropy [2].

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Modelling stress concentrations induced by zirconium hydrides in fuel cladding by neural networks

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Delayed Hydride Cracking (DHC) is a phenomenon of interest in the maintenance of Zirconium based fuel cladding. Modelling DHC requires knowledge of the internal stresses within the Zr polycrystal structure as tension is applied by fuel swelling, as this dictates where hydrogen will diffuse within the structure. Calculating these stress states requires significant computation with traditional numerical methods due to the complex geometry of polycrystals so constructing statistical models covering the possibility space is currently intractable. The use of a neural network as a surrogate model is proposed, as neural networks can be much faster to execute compared to traditional numerical methods. Models based on UNet and Fourier Neural Operator architectures has been developed and applied to linear elastic calculations and can be used for a 6000x speedup and 6500x memory reduction compared to finite element analysis using ABAQUS and is currently being adapted for application to CPFE calculations for greater acceleration. The results of these models may then be fed into a fast simulation of DHC allowing for statistical analysis of coarse grain structure properties on the progression of cracking. As models accounting for plasticity take significantly longer to run, speedups in the 100,000x range may be possible.

Dislocation core and mobility of the screw dislocation in uranium dioxide

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Uranium dioxide (UO_2) is the standard fuel in 2nd and 3rd generation pressurized water reactors (PWR). During operation, especially under transient or accidental conditions, the fuel pellets reach temperature and stress conditions under which UO_2 exhibits a viscoplastic behaviour. Plasticity is a crucial phenomena for nuclear safety since it allows the fuel pellets to accommodate the stresses in the rod. The understanding of UO_2 viscoplastic behaviour, including irradiation-induced hardening, is paramount in providing physically based mechanical constitutive laws for use in fuel performance codes.

The plastic deformation of UO_2 single crystal is governed by the motion of dislocations that glide in the 1/2<110>{001}, 1/2<110>{110} and possibly the 1/2<110>{111} slip systems, the first one being known to be activated at lower stress than the other two. Recently, a dislocation mobility law was derived from atomistic simulations to describe the glide of the 1/2<110>{001} edge dislocation, known to be the rate-limiting character in {001} [1]. However, it is the screw dislocation that becomes of interest with regard to irradiation hardening as it is more prone to form locks with {110} irradiation loops [2]. In order to study the hardening induced by irradiation defects, it is thus important to investigate further the screw dislocation properties, including dislocation core and mobility properties, in the various slip systems of UO_2.

We use molecular dynamics with empirical interatomic potentials to study the motion of the screw dislocation in UO_2 in the various glide planes available. The microscopic mechanisms of motion are examined in the 1400 K to 2200 K temperature range. Special attention will be paid to the cross-slip mechanism, which is very favourable in this temperature regime. Based on this, mobility laws are proposed for use in discrete dislocation dynamics simulations of UO_2 single crystal plasticity.

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PLEIADES/Mérope: A microstructure generator for simulation of nuclear materials

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In this talk, we present the computational library Mérope, which is part of the nuclear fuel software platform PLEIADES, which is developed at CEA. This library is devoted to microstructure generation, using the Representative Volume Element method, a numerical technique used to derive the effective properties of a random heterogeneous material. Mérope performs 3 steps: define a geometric microstructure, discretize it and calls a solver (FFT-based or a classical Finite Element ones) in order to extract the effective physical behaviour at the mesoscale. (The physics under concern is mainly thermomechanics.)

The core functionalities of Mérope are devoted to defining a random microstructure and discretizing it. Mérope conceptualizes the microstructure as a combination of inclusions, polycrystals, and random fields. Its discretization is either a periodic tetrahedral mesh, or a periodic voxellation (a regular Cartesian grid); composite voxels are available. An emphasis is put on the efficiency (speed and ease of use) of the tool; we will exemplify it on a recent study [1], where the authors propose a model for the porous network of a nuclear fuel material, and optimize its parameters taking into account experimental images of the real microstructures, in an automated procedure.

Mérope is available for research in the open-source repository [2].

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First principles study of volatile fission products trapping properties and chromium doping in actinide oxides

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UO_2 and $(U,Pu)O_2$ are the most used fuels in the current nuclear reactor fleet. Under irradiation in a reactor, new chemical elements are created in these materials by the fission of uranium and plutonium nuclei. Some of these fission products, especially iodine, caesium and tellurium may chemically react with one another to form compounds that are potentially corrosive for the cladding.

One way to avoid the cladding corrosion consists of preventing the formation and migration of such corrosive species within the fuel. To that end, the determination of the most stable chemical forms of iodine, caesium and tellurium in the fuel as well as the trapping and diffusion mechanisms of these elements in UO_2 and $(U,Pu)O_2$ is of first interest.

The gain of Cr-doped UO_2 fuel on the fission gas retention has already been demonstrated experimentally [1]. However, no data are yet available on the release of corrosive fission products in Cr-doped UO_2. With a view to simulating the behaviour of these species within Cr-doped UO_2, one has to prior identify the most favourable oxidation state and location site of Cr in UO_2, which is a much debated topic in the literature [2].

We perform electronic structure calculations, using the Hubbard-corrected density functional theory (DFT+U) to evaluate the preferred trapping site of I, I_2, Cs and Te in UO_2 and $(U,Pu)O_2$ crystals, as well as the preferred oxidation state and location of Cr in UO_2. We first determine the stability of I, I_2, Cs, Te and Cr in various point defects and then compute their XANES spectra in each considered site, using the FDMNES code [3] and the DFT+U atomic configurations. The comparison of the computed spectra with the experimental ones contributes to the identification of the chemical forms and the trapping sites of iodine, caesium, tellurium and chromium in the actinide oxides.

The use of a Hubbard term (GGA+U) allows us to take into account the strong correlations of the actinide 5f electrons. To avoid the metastable states inherent to this method, we use the occupation matrix control (OMC) procedure [4], which also allows us to control the valences of each species in the simulation. This particular point makes our approach reliable with respect to the determination of fission products incorporation energies in the various studied defects.

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Latest developments in the MARGARET fission gas and microstructure model, based on recent experimental findings and atomic scale modelling

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We present the most recent developments and improvements to the advanced gas model MARGARET [1] in relation to the latest experimental discoveries and atomic scale simulations.

Recently, new classical Molecular Dynamics (MD) simulations have been performed [2] to study the phenomenon of dynamic resolution of gas from intra-granular bubbles back into the matrix, which is of utmost importance for the description of fission gas during irradiation. After a critical review on the subject and comparisons to a detailed and dedicated numerical modelling for trapping and resolution, we have built a new resolution model that can be adopted easily by fuel performance codes, based on resolution profiles provided by M. Cooper and coming from the MD simulations from reference [2].

On the other hand, 3D experimental images of base-irradiated fuel [3] seem to indicate that inter-granular bubbles, below a certain threshold temperature, do not form an interconnected network, as previously thought. Therefore, since we observe fission gas release also in the absence of bubble interconnection, we introduced in MARGARET a new escape pathway for the fission gas, which is the long-range diffusion through inter-granular grain-boundaries.

Other recent experimental findings [4] [5] indicate that, at high burn-up and in the central region of the pellet, grains divide into slightly disoriented sub-domains. The appearance of this phenomenon is in concomitance with the formation of a second, bigger, population of intra-granular bubbles and with a higher fission gas release. Given the apparent relevance of this phenomenon in the behaviour of fission gas, we added in MARGARET a description of these sub-domains.

The final objective of our work is to calibrate the new version of MARGARET including these features and compare it with experimental data. The results of the first tests are presented.

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Inter- and transgranular fracture properties of UO_2 with and without the presence of fission products are studied using electronic structure theory

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Inter- and transgranular fracture properties of UO_2 with and without the presence of fission products are studied using electronic structure theory. An excess energy assessment approach is employed to calculate the model version of stress-strain (traction-separation) curves from density functional theory. The symmetric tilt Σ5(310)/[001] and twist Σ5(001)/[001] grain boundaries were selected. The results show that the lattice has higher toughness than the studied grain boundaries in UO_2, consistent with experimental observations. The impact of two different relevant fission products (FPs) with different chemical bonding, Xe and Mo, on the fracture of UO_2 was also studied using the same methodology. The presence of FPs in the cleavage plane weakens both lattice and grain boundaries, and it is noted that Xe has a more profound effect than Mo.

Micromechanical behaviour of UO_2: modelling of dislocation climbing in the recovery creep stage

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A UO_2 crystal plasticity model [1] has been improved to take into account the thermally activated plasticity and the hardening/recovery creep at high temperature. The proposed improvements allow a physically based modelling of a double-kink mechanism for the computation of the thermally activated slip strain rate. Concerning the hardening/recovery creep, an effective strain rate is introduced in link with the travelling time of dislocations during their interactions. This latter includes the dynamic/static annihilation of stored dislocations with cross-slip and climb respectively. The dislocation climbing velocity is computed with a vacancy diffusion model in a statistically stored dislocations population, submitted to an external stress. Simulations with a finite element tool are undertaken in order to adjust the vacancy diffusion model depending on the dislocation density and their orientation vis-à-vis the applied external stress. The two inelastic strains, thermally activated plasticity and hardening/recovery creep, are coupled with an assumption of addition of travel times [2]. The resulting micromechanical model has been implemented with a finite transformation formalism and integrated by an implicit Newton-Raphson method, using the MFront tool.

A first application of the new model has been performed to simulate compression tests on single crystals at high temperature. After calibrating the physical parameters, the results are compared with experiments to validate the model with the prediction of the effect of crystal orientation. With the climbing velocity model, we can also predict the impact of temperature, strain rate and deviation from stoichiometry, and add it to the comparison to experimental results for validation. The limits of the validation are discussed with in particular the strong dispersion of the experimental data and the question of the structural effects related to the representation of the compression displacement loading in the simulation.

This work is supported financially in the context of the PLEIADES platform co-developed by the French Alternative Energies and Atomic Energy Commission (CEA), Électricité de France (EDF) and Framatome.

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Understanding the Oxidation Mechanisms of Zircaloy using Peridynamic Modelling

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During operation in water-cooled reactors, zircaloy cladding is subject to oxidation. The oxide layer formed is initially passivating, before undergoing a transition, where the protective properties breakdown, causing accelerated oxidation. This is also associated with the formation of lateral cracks, which can be observed periodically though an oxide layer after multiple transitions. The metal-oxide interface can also change from a planar shape to a rough, undulating one. While these effects have been observed experimentally, the underlying mechanisms have not yet been established. Therefore, the aim of this work is to develop a microscale model capturing the metal to oxide transformation with physical phenomena, to explain the experimentally observed behaviour.

This work used the bond-based peridynamics modelling technique, implemented in the finite element method software Abaqus, via subroutines. Peridynamics is a non-local continuum modelling method, suited to modelling the fracture of brittle materials, such as oxides. This method is integral based, to capture discontinuities such as cracks, and allows cracks to form naturally within the model, without a pre-determined initiation point. The material is represented by points connected pairwise by bonds, which are represented by nodes and truss elements respectively in Abaqus. This implementation allowed the anisotropic volumetric expansion of metal to oxide, according to the Pilling-Bedworth ratio of 1.56, be incorporated. Furthermore, an experimentally determined Weibull distribution of fracture strain that was scaled by volume was applied to the trusses, to represent a random distribution of defects and grain structures in the oxide layer, which are too small to be modelled explicitly. A finite element mesh has also been overlaid onto the peridynamics mesh, sharing the nodes in the peridynamics mesh, to incorporate an oxygen diffusion model and observe how this affects the shape of the metal-oxide interface.

Smooth Particle Hydrodynamics for Fuel Performance Modelling

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Research efforts around the work are focused on reducing some of the large uncertainties in the prediction of fuel behaviour that underpins fuel performance models. Smooth particle hydrodynamics is a Lagrangian-based, mesh-free, continuum modelling method. Here this method and its application to the simulation of uranium dioxide fuel microstructures is described. Progress in the development of a tool for the mesoscale modelling of fuel microstructure will be reported and plans to extend its use to allow the behaviour of monolithic and novel kernel-based fuels, for which there is limited empirical data, will be outlined.

Fission gas bubble population in a High burn-up irradiated UO_2 fuel until their internal pressure determination using STEM-EELS

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The behaviour of fission gas (FG) in high burn-up (BU) fuel during steady-state and transient conditions is of special interest for safety reasons. One of the key issues for the fuel behaviour under irradiation is to improve model prediction and it is necessary to know the concentration and the size of fission gas (FG) bubbles in the normal conditions of irradiation.

In this study nano-sized FG bubbles are characterized using TEM (Transmission Electron Microscopy) at different radial positions in the high BU irradiated UO_2 fuel. This work also emphasizes the aspects to be considered for the TEM characterizations, i.e., the representativeness of a lamella in relation to analyses carried out at higher scales, the impact of a defocus value of the objective lens [1], and the impact of the absolute thickness measurement using EELS [2]. After these considerations, a distribution in size and in number of FG bubbles is obtained as a function of the radial position in UO_2 fuel; itself is dependent of the irradiation temperature [3].

To go further in the description of these gas bubbles, it is also necessary to obtain the number of gas atoms contained in these bubbles. In the past, experimental works were performed using EDX method [4,5], on FG bubbles with sizes ranging from a few nanometres to hundreds of nanometres. Thanks to the TEM characterizations done in the LECA-STAR hot laboratory at the CEA Cadarache in France using a TALOS F-200X G2 and a Continuum GATAN Imaging Filter system, it has been possible to acquire EELS spectra centred on the Xe peaks of bubble with sizes of a few nanometres into a high BU UO_2 fuel. Values of xenon atom densities are measured and with choosing an appropriated Equation of State (EOS), the pressures are reached.

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Characterisation by μXRD/XRF of the interlayer between the fuel and its cladding in a high burnup fuel rod

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During irradiation of fuel rods in light water reactors (LWR), the UO_2 pellets directly interact with the enclosing Zircaloy cladding when the fuel-cladding gap tends to close due to pellet swelling and cladding creep-down onto the fuel. After a physical contact between the pellet and cladding is established in this way, the inner surface of the Zircaloy side at the pellet-cladding interface is oxidized, which in turn leads to the formation of a bonding layer at the region of fuel-cladding interface. As the presence of fuel-cladding bonding may affect the thermo-mechanical behaviour of a fuel rod, especially for high-burnup fuel, experimental analyses of the formation, stability, stress-strain states and microstructure of this bonding layer are important for ascertaining validity and reliability in predicting the thermal as well as mechanical behaviour of fuel rods, especially for long term storage.

The focus of this study lies in the pellet-clad interface region of a high burnup spent fuel pellet irradiated in a commercial Swiss nuclear power plant. Structural characterisation of the prepared sample has been carried out using a combined micro-beam X-ray fluorescence (μXRF) and X-ray diffraction (μXRD) experiment in a scanning mode. All measurements have been performed at the microXAS beamline of the Swiss Light Source (SLS) synchrotron facility. In order to comply with the radioprotection aspects, as well as with legislation issues for investigation of radioactive materials at the SLS, a small-size sample at the micrometre scale (about 34 μm x 34 μm x 18 μm) from the targeted region of the objective UO_2 pellet has been prepared by focused ion beam (FIB) technique and subsequently analysed. μXRF maps obtained with high spatial resolution were used to localize the pellet-clad interface region and crystalline phases were identified using XRD with a correlation map representing the spatial variation of crystalline phases. The results show that the tetragonal zirconia (t-ZrO_2) is the main phase present in the bonding layer (although it is a metastable form under LWR temperature and pressure conditions). In addition, the cubic phase of UO_2 (in contact with zirconia) has a larger lattice parameter than that of unirradiated UO_2. The presence of implanted fission products could be the reason for these differences.

Experimental investigation on the stability of corium metallic phases

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During a severe accident in a nuclear reactor, the interaction at high temperature between the UO_2 fuel and the structural materials (Zr alloy cladding and Fe-Cr-Ni steel vessel) leads to the formation of “corium”, a mixture of partially or fully molten phases. The so-called “in-vessel corium” forms a molten pool which is expected to relocate at the bottom of the reactor vessel. The pool may be constituted of stratified layers of oxide and metallic liquid phases. If we consider the metallic region of the corium, the multi-component system U-Zr-Fe-Cr-Ni should be investigated. Due to their high stability, U-containing Laves phases [1] of composition AB2 – where A=Fe,Cr,Ni and B=U,Zr, respectively – are likely to form [2–5] during the solidification of corium (Figure 1). These phases may have a strong impact on the long-term behaviour of corium because of their peculiar mechanical properties [1]. However, crystallographic data as well as composition information on these multi-components phases are rare.

In order to improve their thermodynamic descriptions in the TAF-ID database [6], we investigated the stability of six uranium-containing Laves phases. The samples were fabricated by arc furnace and then annealed at 700°C for 28 days. After quenching, the samples were prepared for metallographic analyses and high-resolution structural analyses on the Mars beamline of the SOLEIL synchrotron [7,8]. This new set of data helps us to redefine the solubility limits and the phase stability domains in the U-Zr-Fe-Cr-Ni multi-component system as well as to get information on the crystalline structure of the Laves phases.

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Thermal conductivity of UN as a function of solid fission product concentration: Separate-effect testing coupled with first principles models

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Thermal conductivity is a crucial property in the design and operation of nuclear reactors as it determines the heat transfer rate between the fuel and coolant. Uranium mononitride (UN) is an advanced nuclear fuel suitable for several high-performance reactor designs, including fast neutron and breeder reactors, due to its superior thermophysical, chemical and mechanical properties. For thermal conductivity, limited data is available in the literature for most advanced nuclear fuel concepts. In this work, we present a comprehensive study by coupling separate-effect tests and first-principles electronic structure theory to calculate the thermal conductivity as a function of temperature and solid fission product concentrations.

The thermal conductivity of UN and $(U_{1-x}Zr_x)N$ pellets was estimated using laser flash analysis (LFA) thermal diffusivity measurements between 298 and 1400 K. The fuel samples were sintered using spark plasma sintering (SPS) of powder fabricated following the hydriding-nitriding method of uranium metal. The ZrN powder used for the composite pellets was fabricated by carbothermal nitridation of ZrO_2. Several compositions were tested with x ranging from 0-50 at%. In addition to the experimental measurements, we also performed first principle atomistic calculations using density functional theory (DFT) combined with Boltzmann transport theory (BTT) in order to predict the thermal conductivity of materials not available in the laboratory, such as plutonium, at the atomic level, providing a detailed understanding of the underlying physics. By comparing the experimental results with the theoretical calculations, we can provide insights into mechanisms and the physics determining the transport properties of complex materials. The results are discussed in detail, including the dampening effect of fission products or transmutation impurities on the relatively high thermal conductivity of pure UN, with respect to other nuclear fuel candidates. Overall, this study provides valuable insights into the thermal conductivity of UN and SIMFUELs and highlights the importance of experimental and theoretical methods in understanding their behaviour. The methodology developed can be extended to multi-composite SIMFUELs, providing data of advanced fuel thermal conductivity degradation during irradiation, not available before. This effort is a step towards an Accelerated Fuel Qualification methodology where separate-effect tests and modelling can reduce the time needed to develop and qualify new fuel systems.

Steady-State Secondary Creep Rate of Uranium Mononitride using Spark Plasma Sintering

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As a part of the qualification and licensing campaign of uranium mononitride (UN) to be used in future lead-cooled fast reactors, the steady-state secondary creep rate of UN has been measured using a unique design of the spark plasma sintering heating element. The experimental configuration consists of the standard graphite die and punches in addition to Al_2O_3 and Si_3N_4 disks, which act as insulators and support materials. Tests at temperatures of 1300 °C and 1400°C were conducted under sample pressures of 10, 20, and 30 MPa. The results show that the steady-state secondary creep rate is in good agreement with the limited data available in the literature. The values of the strain rate at different temperatures and pressures were used to calculate the activation energy and the stress exponent.

Interdiffusion behaviour of UN with Zircaloy-4 via diffusion couple studies

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Uranium nitride (UN) has been of particular focus recently as an advanced fuel candidate for light water reactors (LWRs) having both a high uranium density and thermal conductivity. The interaction between UN and zirconium alloys has not been studied in great detail. To determine the suitability of these fuel-clad combinations, diffusion couples between UN and zircalloy-4 have been performed up to 1000°C for durations up to 100 hours. Minimal interaction was observed at low temperatures (400°C). At temperatures of 700°C and above, an interaction was observed between the materials resulting in bonding of the UN and Zircalloy-4. Investigation via electron microscopy has revealed significant migration of uranium into the Zircalloy-4, up to depths of up to 500 µm within the 1000°C experiments. In addition to the movement of uranium, other elements within the fuel-clad combination also undergo relocation and form additional phases at the contact interface. This study applies up-date-techniques to enhance the limited data currently available.

Understanding thermal property behaviour as a function of radiation damage defects in advanced nuclear fuels

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The search for accident tolerant fuels (ATF) has received a large international effort since the Fukushima accident in 2011. Novel fuel materials, such as uranium nitride (UN) are of interest due to its much higher thermal conductivity compared with UO_2 which reduces operating centreline temperatures and could delay the impact of a loss of coolant accident allowing operators more time to regain control of the reactor. Furthermore, silicon carbide (SiC) has been examined extensively for replacement of the Zr cladding due to its much higher melting temperature and good thermal conductivity, which may provide a passive nuclear fuel assembly and mitigate the effects of a loss of coolant with and avoid meltdown of the fuel assembly completely. Beyond Gen III systems, novel fuel assemblies which have much improved thermophysical properties are needed due to the higher fuel operating temperatures with one candidate, TRISO using microsphere of UO_2, coated with layers of pyrolytic carbon and SiC. One of the main advantages of these novel nuclear fuel and cladding materials are their excellent thermophysical properties such as thermal conductivity. Neutron irradiation and post irradiation examination of these materials are limited due to the challenges working with spent nuclear fuel and irradiated materials which require specialised facilities. This programme of work looks to examine ion beam irradiation as a surrogate for neutron/fission product damage and perform thin film measurements of the materials thermophysical properties. Coupled with extensive micro and nano-structural characterisation of the radiation damage defects to develop a model of the effect of radiation damage defects on the thermal conductivity of advanced nuclear fuel and clad materials.

UN and SiC fabricated at the Nuclear Fuels Centre of Excellence (NFCE) at the University of Manchester have been irradiated with heavy ions, Ar, to replicate fission gas in UN and Si ions to mimic neutron damage in SiC at temperatures of 300°C to reactor relevant DPA levels. Post irradiation examine results will be presented, detailing X-ray diffraction (XRD), Transmission Electron Microscope (TEM) and Raman results of the defect type as a function of radiation damage dose. Transient grating spectroscopy (TGS) has been used then to probe the first ~μm of ion irradiated material and measure the thermal diffusivity. For SiC, this has shown a significant decrease in thermal conductivity with low radiation damage dose, coupled to lattice parameter measurements and Raman results this suggests point defects in the materials have the most significant impact before extended defects such as dislocation loops and voids become visible in the TEM. Compared with neutron-irradiated bulk property measurement data this has also validated the techniques of ion irradiation and TGS as techniques to probe the thermal property behaviours in nuclear fuel materials which can be a very attractive tool to pre-screen candidate materials prior to a neutron irradiation campaign.

Chromium speciation in Molten Salt Reactor fuels

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Fast neutron spectrum Molten Salt Reactors are one of the leading options for the future of the nuclear power generation. Considered primarily in the context of the small modular reactor, they are also an attractive option for a new generation of larger industrial designs. One of the main concerns on the safety of this technology, is the structural resistance of the reactor vessel to the strongly corrosive environment of the molten salt fuel. It has been demonstrated that the chromium is the element in the structural nickel-based alloy (reference material for MSR), with the highest probability of depletion [1]. Furthermore, studies of the formation of CrF_3 in molten fluoride salt conditions (FLiNaK), have shown that in halogen solution, the presence of the chromium ions could enhance the mass loss of the pure chromium exposed [2].

Understanding the conditions of formation of chromium corrosion products (e.g., CrF_2, CrF_3, $CrCl_2$, $CrCl_3$) in the fuel salt is therefore critical to the safety assessment of the MSR [3]. In this work, various fluoride and chloride salt mixtures, considered as potential fuel options, have been exposed to an excess of pure chromium metal for 100 hours under operating conditions (973 K). The samples have been thereafter analysed to identify the chemical state of chromium and intermediate compounds eventually formed, using techniques such X-Ray Diffraction.

This preliminary study aims to improve our understanding of the chromium behaviour in different molten salt fuel environments, and to help the design of corrosion management strategies.

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Analysis of Future Fuel/Cladding Systems for Light Water Reactor Use

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Advanced technology fuels (ATFs) have gained significant interest in recent years, driven by a desire to replace zirconium cladding and remove the possibility of the zirconium-steam reaction. Fuel components of ATF are typically higher density and offer improved thermal conductivity when compared to UO_2. There is therefore the potential for ATFs to provide greater safety margins within the core design to allow better outcomes during a loss of coolant accident. Furthermore, due to several ATFs having a higher uranic density than uranium dioxide there is also potential for greater fuel economy.

Several combined fuel-cladding systems are of interest for near-, medium- and long-term light water reactor use. These can include uranium nitride or uranium boride with FeCrAl; refractory materials such as molybdenum; zircaloy-4; other zirconium alloys. Although studies have been carried out previously on fuel-clad interaction, these are generally isolated investigations into specific systems, rather than an examination and down-selection of potential fuel-cladding options. This project aims to manufacture several fuel materials using the Nuclear Fuel Centre of Excellence facility at the Royce Institute, and systematically investigate their interaction with potential cladding options via diffusion bonding experiments achieved either through conventional methods or using Spark Plasma Sintering.

Solid-state electrochemical methods for characterising thermodynamic and diffusion properties of non-stoichiometric fuels

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Solid-state electrochemical methods have been used for many years for characterising thermodynamic properties of oxides in general and non-stoichiometric both fresh and irradiated fuels in particular [1,2,3]. Recently, we have developed a unique tool, dedicated to high temperature studies, that has helped us build on these applications. It transpires that such tools have much more far-reaching applications than originally reported [4].

In this comprehensive talk, we start by describing the device we have developed for applications relating to mixed uranium oxides. We identify the physical meaning of the quantities that are monitored during any given experiment, based on electrochemical diffusion theory and electrical conduction properties of the studied materials [4].

We then demonstrate the versatility of this tool through several examples relating to non-stoichiometric uranium and cerium oxides. Steady state regimes provide the basic thermodynamic and phase transition data so crucial to the in-service use of such materials. In addition, we show that through a careful analysis of the data using appropriate point defect models [5], the nature of oxygen point defects in oxygen deficient or excess oxides may also be determined. In a novel development and with an appropriate model, the transient data pertaining to solid-state or gas-phase redox experiments is quantitatively related to oxygen chemical diffusion coefficients or oxygen solid-gas exchange coefficients. Additional theoretical analyses reveal that under certain conditions, the method may provide first-hand estimates of oxygen point defect diffusion coefficients, making this a unique source of data for assessing first principles approaches. An example is provided of our device's performance in relation to uranium fuels and how their oxygen activity can be buffered using appropriate additives, in a reduced and oxidized form.

Finally, we draw on examples from past studies [3] to illustrate the potential usefulness of such solid-state methods for characterizing the chemical state of irradiated fuels. We dwell upon the significance of O/M or O/U ratios in relation to these studies and the practical significance of such quantities whilst pointing out the experimental complications such measurements entail. Throughout this presentation, we go to lengths to point out the practical limitations of the method and the caveats one should be aware of when implementing it.

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Fission chemistry and high temperature behaviour of irradiated MOX fuels with Pu/(U+Pu)=0.45 by thermodynamic calculations

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In the frame of the PUMMA European project, the aim of the present work is to contribute to the understanding of the high temperature behaviour of MOX fuels with Pu/(U+Pu)=0.45 for SFR (Sodium cooled Fast Reactors) reactors [1]. The influence of the burnup on the thermodynamic properties is studied through the fission product thermochemistry. Thermodynamic calculations using the CALPHAD method are performed using the Open Calphad software [2] and the TAF-ID database version 14 [3]. The thermodynamic properties of the irradiated MOX fuels in CAPRIX (in PHENIX reactor, France) [4] and TRABANT (in HFR reactor, the Netherlands) experiments [5] are studied. In a first step, the input data for the thermodynamic calculations: average and local fuel compositions with fission product inventory, local temperature at the end of the irradiation, are estimated using the GERMINAL V2 fuel performance code [6].

In addition to actinides and oxygen, 15 fission products are taken into account in the thermodynamic calculations. The calculated data are: the phase mole fraction and composition as a function of temperature, the fuel melting temperature (solidus), the oxygen to metal ratio of the fuel matrix, the oxygen potential, the heat capacity as a function of temperature. To study the impact of the burnup, the calculated data are compared between the un-irradiated and irradiated fuels. For CAPRIX fuel, the calculations are compared with PIE characterizations. A good agreement is obtained. The results for CAPRIX and TRABANT fuels are also compared. This study shows that such a thermodynamic approach highly contributes to the understanding and prediction of the behaviour of high plutonium irradiated MOX fuels.

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Coupled modelling of structural, thermodynamic and physicochemical properties of NaF-KF-UF_4 fuel salt

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Molten Salt Reactors (MSRs) are one of the Generation IV nuclear reactor designs that hold the promise of generating cheaper, stable and carbon-neutral energy in a modular and fast scalable fashion [1]. MSRs contain the fuel in the liquid phase - a molten salt mixture of the fissile material, e.g., uranium fluoride. It is of the utmost importance to know the accurate thermo-physical and transport properties of the liquid fuel such as solidification temperature, density, viscosity, heat capacity, thermal conductivity and vapour pressure, at the design stage, during reactor operation, and to perform risk analysis of off-normal conditions. The hazardous and challenging nature of the actinide-containing salts that are radioactive, hygroscopic and corrosive, as well as the advances in the accuracy of in-silico research make atomistic simulations and thermodynamic modelling attractive in the study of ions within molten salt systems, reducing the experimental input. The reference fuel salt for the MSR design at Seaborg is a NaF-KF-UF_4 eutectic mixture, known under the acronym FUNaK, for which a comprehensive thermodynamic assessment and key physicochemical properties are still incomplete [2].

In this work we have focused on the development of coupled model of the structural and thermodynamic properties of the NaF-KF-UF_4 system from a microscopic (local structure) to macroscopic scale, combining experimental measurements, molecular dynamics (MD) simulations and a CALPHAD modelling approach. We have investigated the local structure of selected relevant compositions in the NaF-UF_4 and KF-UF_4 binary sub-systems and ternary system NaF-KF-UF_4 using high temperature X-ray absorption spectroscopy (XAS) measurements at the KARA synchrotron facility [3]. Moreover, we have coupled XAS with MD simulations using the Polarizable Ion Model (PIM) [4]. We have determined the coordination number of uranium, nature of neighbouring atoms and identified the formation of molecular complexes in the melt as a function of temperature, composition and change of the solvent. In overall, we validated molecular dynamics simulations of the binary and ternary systems and improved the predictive capabilities of the MD models.

The molecular simulations moreover allowed us to calculate relevant thermo-physical properties (density, thermal expansion, mixing enthalpies, heat capacity, viscosity, thermal conductivity), which are validated against the existing experimental data.

Finally, the experimental and simulation results served subsequently as input to build a comprehensive model of the NaF-KF-UF_4 system, linking structure and thermodynamic properties based on the CALPHAD methodology and modified quasi-chemical model in the quadruplet approximation (MQMQA) [5].

The authors would like to acknowledge Seaborg Technologies ApS for fully funding this project.

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Study of the Pu-Np-O system: an approach coupling experimental and CALPHAD modelling

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Partitioning and transmutation (P&T) of minor actinides (MAs) is a nuclear fuel cycle strategy aimed at reducing the long-term radiotoxicity and volume of nuclear waste. This route consists of the separation of MAs from spent nuclear fuel, followed by their conversion into less hazardous isotopes through transmutation processes. This approach is studied in the framework of the European project PATRICIA. The concept developed in this project is based on the use of dedicated inert-matrix fuels, MA-bearing MOX or MA-bearing blankets in fast neutron reactors. Having a comprehensive understanding of the thermophysical properties of this new type of fuel (such as heat capacity, melting temperature or thermal conductivity) is crucial for the design of reactor cores and the understanding of their behaviour throughout reactor operations and off-normal conditions. Unfortunately, there is a lack of data in the literature, especially for fuels containing Np. In fact, some studies have focused on the system U-Np-O ([1], [2], [3]) and U-Pu-Np-O ([4]). However, the Pu-Np-O system was not assessed to this date. In this context, the acquisition of experimental data enabling the development of thermodynamic models is essential.

In the present study, the system Pu-Np-O system is investigated by various experimental techniques (drop calorimetry, laser heating techniques, Knudsen Effusion Mass Spectrometry, X-Ray Diffraction, Raman spectroscopy and microscopy). Based on these data, a thermodynamic model of this system is developed using the CALPHAD method with the Thermocalc software.

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Experimental insight and modelling of the NaCl-$ThCl_4$-$PuCl_3$ fuel salt properties

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Molten salt fuels offer immense potential for advanced GEN IV nuclear reactors due to their unique safety and operational characteristics. Furthermore, in a time of heightened environmental concerns and resource scarcity, the closure of the nuclear fuel cycle – which reduces the radioactive waste disposal footprint and the need for natural resources – reinforces a sustainable nuclear energy strategy. This involves reprocessing of spent nuclear fuels coming from the current generation of Light Water Reactors, and the extraction of valuable elements that can be (re)used. Significant improvement in converting Pu isotopes and minor actinides (MA) into fission products with shorter half-lives can be achieved with the introduction of advanced fast reactor systems, among which the Molten Salt Reactor based on chloride salt is a particularly promising option.

In the framework of the European project MIMOSA [1], our group is currently investigating the thermochemical and thermophysical properties of the NaCl-$ThCl_4$-$PuCl_3$ system, one of the candidate fuels [2] for this type of application. In this presentation, we will delve into the experimental insight and modelling of the fuel salt, with first a focus on the synthesis of high purity $ThCl_4$ using a chemical vapour transport reaction (CVTR). Renewed investigations of the phase equilibria in the NaCl-$ThCl_4$ system by Differential Scanning Calorimetry (DSC) will also be presented. Moreover, molecular dynamics (MD) simulations based on the Polarizable Ion Model (PIM) will be reported that provide key thermo-physical properties such as density, viscosity, thermal conductivity and diffusion coefficients. Comparison with the available experimental data will be discussed. The ultimate objective, as will be presented, is to develop a coupled model that accurately represents the structural, thermodynamic and thermo-physical properties of the NaCl-$ThCl_4$-$PuCl_3$ fuel salt, i.e. from the atomic to the macroscopic scale.

[1] MIMOSA (MultI-recycling strategies of LWR SNF focusing on Molten Salt technology) EURATOM-2021-NRT-01 project, https://www.mimosa-euratom.eu/

[2] T. Dumaire, J.A. Ocadiz-Flores, R.J.M. Konings, A.L. Smith, Calphad 79 (2022) 102496

POSTERS

P1 Advancing Fuel Performance Analysis: Integration of Finite Element Model with TRANSURANUS, Methodology and Validation

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This work introduces an innovative approach to enhance the capabilities of the TRANSURANUS fuel performance code by integrating it with a reliable Finite Element Model (FEM) using ANSYS. TRANSURANUS, a widely utilized 1.5-D code, conducts thermomechanical analysis with radial discretization for each axial slide, assuming cylindrical symmetry during simulations. This study focuses on incorporating the material properties of 15-15Ti and investigating nonlinearities, including thermal and irradiation-induced swelling and creep.

The implementation of 15-15Ti material properties is explored, with particular emphasis on addressing nonlinearities. The methodology and material property implementation are validated through comparisons with literature data. Additionally, validation based on in-pile irradiation experiment data is performed to establish the reliability of the approach.

The findings demonstrate a promising and reliable approach for investigating complex three-dimensional phenomena. By leveraging the extensively validated TRANSURANUS fuel performance code, this study provides an advanced framework for exploring fuel performance and behaviour in Gen-IV reactor systems.

P2 Estimates of fragmented fuel thermal conductivity using the DEM-FFT simulation method

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The issues of Fuel Fragmentation, Relocation and Dispersal (FFRD) of oxide fuel are critical to understand and predict the behaviour of PWR fuel rods in the event of a loss-of-coolant hypothetical scenario. In order to introduce a homogenized representation of pulverized fuel that could be used in fuel performance codes, the thermal conductivity of UO_2 granular beds in noble gas mixes was studied numerically. The Discrete Element Method was first used to compute the arrangement of particles only submitted to gravity with a representative size distribution and a homogeneous spatial distribution. These generated geometries were then transformed into a 3D image (voxellized) in order to compute their homogenized properties using a Fast Fourier Transform solver(1). Different hypotheses could be made in the voxellation process about how to deal with gas-solid and solid-solid interfaces, which in turn provided different numerical approximations of the effective properties. Due to the finite spatial discretization and high polydispersity, a two-scale scheme was introduced and subjected to numerical validation. The DEM computed packing fraction and FFT computed effective thermal conductivity were compared to usual models for granular materials. Typical heat transfer properties of fragmented fuel at several burn-ups were then computed, taking into account approximations of the Knudsen and radiation size effects specific to packed beds.

(1) A DEM/FFT approach to simulate the effective thermal conductivity of granular media. T. Calvet, J-M. Vanson, R. Masson. 2022, International Journal of Thermal Sciences, Vol. 172, p. 107339

P3 Fuel centreline temperature estimation using machine learning surrogate models

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Nuclear fuel design optimization and safety analysis rely on the ability to accurately predict the thermomechanical behaviour of fuel rods in all operating and accidental conditions. Fuel behaviour comprises multiple complex, interdependent phenomena driven by heat generation and transfer under irradiation.

State-of-the-art fuel performance codes (FPCs) employ explicit physical models of these phenomena to predict key metrics of fuel behaviour, such as pellet centreline temperature, radial cladding displacement and amount of gaseous fission products. FPC predictions are inevitably affected by uncertainties due to choice of alternative models, simplifying assumptions and semi-empirical correlations. Fuel thermomechanical quantities are thus often under- or over-estimated in comparison to integral tests, potentially compromising the fuel safety margins. In this contribution, we develop surrogate machine-learning based integral models of fuel behaviour, trained directly on in-pile experimental data.

The training dataset is built from selected Halden reactor experiments equipped with fuel centreline temperature measurements (e.g., IFA-677.1) with linear power and burnup upper limits of ~40 kW/m and ~45 MWd/kg$_{UO2}$ respectively. The pellet centreline temperature is chosen as output metric due to both its relevance as safety limit for reactor design and operation, and the availability of experimental datasets.

Machine learning algorithms allow us to model the dependence of fuel behaviour metrics on fuel operating conditions without explicitly modelling the separate phenomena and their interaction. In our work, we choose Kernel Ridge Regression (KRR) and Long Short-Term Memory (LSTM) neural networks as both algorithms are suitable to capture non-linearity among input features evolving in time. KRR and LSTM models are trained separately on different sets of features, subsets of the training dataset and kernel functions. Hyperparameter optimization is performed via randomized search in KRR and manual tuning in LSTM. Model predictions are evaluated against test datasets and compared to best estimate FPCs. The results of our work aim to contribute to the application of surrogate modelling in the nuclear field.

P4 PuMMA european project on Pu management in GENIV systems

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The European project PUMMA (Plutonium Management for More Agility) is dedicated to the different Pu management options in 4th generation systems to assess the impact on the entire fuel cycle. Fast neutron reactors with associated fuel cycle strategies have been chosen to cope with these options because they are flexible: they offer the possibility of isogeneration, burning or breeding of plutonium.

The fuel cycle scenarios associated with the different strategies will be evaluated. The behaviour of MOX fuel with Pu contents of 45% will be studied experimentally through the characterizations of fuels from three irradiations carried out under nominal conditions (in MTR and SFR) and incidental (in MTR). PUMMA will provide additional results on the thermo-mechanical properties of this fuel covering the full range of composition and effect of irradiation. These studies will be supplemented by dissolution tests on spent fuels with high Pu contents because to date, the studies have been limited to concentrations below 30%.

The construction of this project was carried out with complementarity between the disciplines of the fuel cycle and with a close exchange between simulation and experimental verification for each of the fields: fuel behaviour under irradiation, material properties, spent fuel dissolution and partitioning. PUMMA will be the link between Europe and other international organizations: fuel cycle studies at IAEA and OECD, GEN-IV systems at ESNII and GIF, studies on fuel materials at OECD.

Another objective is to maintain the expertise and skills on the management of plutonium in Europe involving the young generation of researchers with experts who have contributed to these projects for over 20 years.

Twenty-two participants contribute to this project with a total budget of around 7 M €. PUMMA started in October 2020.

P5 Uncertainty Quantification of Thermal Performances in Nuclear Fuels

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Currently Nuclear Fuel Performance Codes approved by national regulator do not account for bimodal grain size distributions in nuclear fuels and instead assume that eg fission gas release kinetics can be adequately described by a single effective grain size value. A more realistic approach is to account for a bimodal grain size distribution and quantify the effect on fission gas release which affects the centreline temperature by changing the conductivity of the fuel-clad gap in the fuel. Here, we focus on quantifying accompanying uncertainties in fission gas release due to grain size distribution, and its overall effect on thermal performance.

The work is part of WP3 of the “Enhanced Methodologies for Advanced Nuclear System Safety (EMEANSS)” Project funded by EPSRC.

P6 Modelling of the fragmentation of a UO_2 pellet in a LOCA situation using the discrete element method (DEM) coupled with a cohesive zone model (CZM)

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The conditions of a loss of coolant accident (LOCA) cause a drop of the pressure surrounding the fuel rods while the temperature increases in the fuel element, especially in the cladding. These loading conditions together with the fuel-cladding differential expansion and the presence of fission gases trapped inside microscopic bubbles in the fuel, can lead to overfragmentation of the UO_2 fuel pellets. The confinement pressure threshold at fragmentation depends on fuel behaviour parameters such as the fracture toughness or the surface fraction of bubbles at the grain boundaries. In order to adjust these parameters, which are poorly quantified to date, and to describe the behaviour of the fuel at fracture, a model combining the discrete element method (DEM) with a cohesive zone model (CZM) is used. The evaluation of the failure threshold as a function of the material parameters allows us to bound these values and leads to a better prediction and measurement of the fragmentation of the fuel in real conditions.

P7 Effect of pre-sintering temperature of uranium dioxide (UO_2) granules on the morphology of molybdenum channels and thermal conductivity of UO_2-Mo composites synthesized by spark plasma sintering (SPS)

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Although uranium dioxide (UO_2) is widely used as fuel for light water reactors (LWRs). However, it has a low thermal conductivity limits its performance both during normal operation and in case of an accident. Adding a material with high thermal conductivity is a potential approach to enhancing the thermal conductivity of fuel compounds consisting of UO_2. Molybdenum (Mo) has been deployed as an additive material in UO_2 composites and forming an interconnected structure of the reinforcement material can significantly enhance the thermal conductivity of the reinforced composite. Pre-sintered UO_2 granules were used in this study to improve the continuity of Mo channels. UO_2–10 vol% Mo composites were produced using UO_2 granules pre-sintered at 800°C and 1200°C by carbolite tube furnace in Ar–3% H_2 atmosphere for 2 hours and non pre-sintered granules as a control, followed by spark plasma sintering (SPS) of the mixtures at 1200°C for 5 minutes. The composites were characterised on their microstructural properties and the thermal conductivity of UO_2–Mo pellets were measured and compared to those of the UO_2 by laser flash analysis (LFA). At a maximum measurement temperature of 800°C, a 50% increase in thermal conductivity was achieved of the composites containing UO_2 pre-sintered at 800°C and the not pre-sinter granules. While the increase was about ~30% for composites UO_2 pre-sintered at 1200°C. These results suggest that higher temperature pre-sintering may be detrimental to forming the continuous channels. This will be investigated through microstructural characterisation of the composite materials.

P8 FEM simulation of $(U,Pu)O_2$ powder compaction for Nuclear fuel fabrication, elasto-plastic calibration model and sensitivity study

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In the fuel cycle manufacturing process is based on milling, pressing and sintering steps. Many parameters can influence these steps and affect the behaviour of the powder. The pressing step must be mastered, takes into account the history of the powder, to generate a good pellet quality in order to facilitate the sintering step and obtain the best behaviour of the fuel in the reactor. Then, the pressing cycle must be adapted to each project, and using simulation to predict the result before manufacturing and to show the influence of the parameters on the quality of the final product.

To do this, the elasto-plastic model used in the FEM calculation code must be calibrated [1]. This is the purpose of this study. In fact, we propose a sensitivity study of the parameters of the Drucker Prager Cap model on the simulation result. Initially, we started the simulation with the initial model $(U,Pu)O_2$ and we gradually checked the influence of the model parameters on the simulation results. Finally, we studied the model behaviour and the fuel shape impact on the variation of the main parameters.

Keywords: Fem, Macroscopic, Elasto-plastic, Drucker Prager Cap, sensitivity study.

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P9 Multiscale modelling of irradiation-induced strengthening in UO_2

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Uranium dioxide (UO_2) is the standard material used as nuclear fuel in pressurized water reactors. Understanding its mechanical behaviour in nominal, power transient or accidental conditions is of great interest for nuclear safety. In normal operation, fuel pellets reach temperatures of ~1300 K and withstand high thermo-mechanical stresses under creep conditions. Under off-normal operating conditions, temperature drastically increases in the fuel (T>1900K) what promotes creep strains and can impact the fuel integrity. In addition, UO_2 fuel pellets are subjected to microstructural changes due to irradiation-induced defects such as ½<110>{110} prismatic dislocation loops generated from interstitial atoms accumulation that can impede plastic flow and influence the pellet viscoplastic behaviour.

In this study, we investigate how primary ½<110>{100} dislocations interact with ½<110>{110} prismatic loops with the aim to build a crystal plasticity framework of UO_2 deformation and hardening. First, molecular dynamics simulations are used to characterize local contact reactions between regular ½<110>{100} dislocations and various prismatic loop configurations at 2000 K under a constant shear stress. A specific attention is paid to categorize the reaction outcomes for various size of irradiation loops as function of the dislocation character. In parallel, discrete dislocation dynamics (DDD) simulations are performed to verify how much atomistic results can be reproduced using the elastic theory. While parts of the dislocation reactions show similarities with common observations in metals [1], original configurations also occur requiring a specific treatment within the DDD.

Massive DDD simulations are then run to quantify the strengthening induced by a large population of irradiation loops adjusted on the experiments [2]. At this stage, local contact reactions are investigated using a statistical approach and irradiation-induced strengthening is discussed as compared to other strengthening contributions in UO_2 as e.g., forest hardening as computed by Portelette et al. [3]. Finally, a crystal plasticity model including the various contributions to strain hardening in UO_2 is proposed.

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P10 MOX fuel with interconnected porosities for higher flexibility in Fast Neutron reactors

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One of the design requirements for fast reactor fuels is to be able to evacuate as much as possible the fission gases in order to enhance the safety of the fuel element during power transients. In fact, gas retention is at the origin of a gaseous swelling during a power transient that could lead to fuel melting and/or cladding failure. Very high gas release happens in SFR MOX fuel (average of 80% release [1], [2] for a standard fuel pin) at the high fuel temperatures reached in the typical nominal power conditions (1500-2700 K), thanks to the thermally-activated diffusion and high temperature gradient (≈4000 K/cm).

The scope of the microstructure subject of this work is to release the gas also athermally (at low power and temperature conditions), meaning relying on the shape of open porosities as the main driver of the gas release instead. This is necessary since different reactor designs (SFR, LFR, GFR, AMR), as well as requirements on reactor power flexibility (load following) in order to adjust to higher proportions of variable renewable energy in the electricity mix, lead to run at low power regime (50-300 W/cm). A MOX microstructure with interconnected porosities was investigated to reach this objective. This microstructure has already been developed and tested in reactor on other fuel types and provided a proof of concept [3], [4]. A trade-off between the capability of the fuel to evacuate gases (requiring medium-high and very interconnected porosity) and to conduct heat (requiring low porosity) is inevitable. For this reason, an optimization work is underway to identify the optimal fabrication parameters and routes.

We will summarize the work done to identify the relevant geometrical parameters that influence the steady-state behaviour of the microstructure (from the thermal and gas diffusion points of view).The homogenization method has been used to investigate the heat transport and gas transport properties of a SFR MOX fuel on a representative volume element. Sensibility analyses have been carried out on the equivalent heat conductivity to investigate the separate effects of the different geometrical parameters. The size of the pores with respect to that of the grains and the dispersion in size of the grains have been found as two of the most important parameters impacting heat conductivity. An optimization algorithm has then been employed that generates the microstructure, evaluates heat conductivity and gas retention, combines these two outputs together with global porosity in a cost function and minimizes this cost function over successive iterations.

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P11 Self-diffusion coefficients modelling from atomistic-scale calculations in $(U,Pu)O_2$ mixed oxides for fuel behaviour modelling

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Uranium-plutonium mixed oxides (MOX) are a type of nuclear fuel currently operated in pressurized water reactors, in the aim of optimizing uranium resources and moving towards the closing of the fuel cycle. The prediction of their behaviour requires the characterization of its properties by a combined modelling and experimental effort. Multiple phenomena affecting their structural and physical properties occur during the fuel irradiation, such as swelling or fission-gas release. The latter are triggered by the formation and growth of defect clusters and gas bubbles driven by the atomic-scale diffusion of irradiation defects and fission products. Most of the current diffusion models for MOX assume a behaviour similar to that of UO_2, or simplified interpolations for Pu concentrations ranging from 0% to 100% [1]. Further studies are needed to assess the validity of this assumption, but the task is far from trivial due to the chemically disordered nature of the compound.

In this work, we compute the thermal self-diffusion coefficients in MOX fuel by a combination of ART-nouveau [2], KineCluE [3], and atomic-scale calculations of migration energy barriers. ART-nouveau allows us to explore the energy landscape and uncover non-trivial migration mechanisms while atomic-scale calculations compute their corresponding energy barriers. KineCluE uses these mechanisms and energy barriers to determine transport coefficients as thermodynamic averages with the Self-Consistent Mean Field theory. This approach allows for a wide exploration of the possible diffusion trajectories, thus for a more accurate evaluation of self-diffusion coefficients with respect to previous approaches. We first investigate the diffusion properties in the pure oxides (UO_2, PuO_2), and then obtain self-diffusion coefficients in MOX as functions of Pu concentration. This work will serve as a base to study at a later stage the diffusion properties of fission gases.

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P12 Study of chromium doped UO_2 as Accident Tolerant Fuel by the means of a new variable-charge interatomic potential

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Accident-tolerant fuels (ATF) are designed to improve fuel performance under accident conditions, such as a loss-of-coolant accident (LOCA) or reactivity-initiated accident (RIA), while maintaining good operational characteristics under normal conditions. Chromium-doped UO_2 is one of these new concepts.

The purpose of this study is to investigate the impact of chromium on the diffusive behaviour of two fission products (Cs and Mo), known for their influence on fuel chemistry in accidental conditions. This is achieved with a new version of the SMTB-Q(1) potential, named SMTB-QB (Second Moment Tight Binding Qeq Bond). This new potential ensures local electronegativity, rather than global electronegativity as was the case with the SMTB-Q potential.

The new potential was implemented in a molecular dynamics code (LAMMPS) in order to study charged defects in UO_2 or in non-stoichiometric UO_2. Using molecular statics calculations, we investigated the different incorporation sites of chromium in the doped UO_2 matrix and compared our results to DFT calculations. Using molecular dynamics calculations, we investigated the diffusion of oxygen in UO_2 and compared it to chromium-doped UO_2. The next step in this work will be to study, using molecular dynamics, the diffusion of the fission products of interest (molybdenum and caesium) in chromium-doped UO2 by determining their preferential migration paths and the associated diffusion coefficients, and compare these results to those obtained in undoped UO_2.

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P13 Modelling the evolution of point and extended defects in UO_2 under irradiation

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This work summarizes the development of a physically-based model for the evolution of dislocation density in irradiated UO_2. The model was conceived to substitute correlations such as the one used in the CEA MARGARET code for applications that go beyond their calibration range. The model is sufficiently complex to be used in a fuel performance code.

Furthermore, the model test case predicts high values of dislocation densities at low fuel temperature, suggesting a substantial HBS formation that may interest all the fuel pellet in novel SMRs for heat production. The model is also in good agreement with experimental results performed on conventional reactor fuel rods. A surrogate model of the full-scale model, based on ANN, is presented. The ANN takes as input fission rate, temperature, and burnup to calculate eventually the dislocation density variation. The mean square error between training and validation data is around 0.01%, indicating the surrogate model is able to reproduce adequately well the model results.

P14 Atomic scale calculation of thermodynamic properties of Americium-bearing oxides

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Americium is a chemical element produced by neutron capture in nuclear reactors, whose strong radiotoxicity is a major issue for the management of nuclear waste. One solution envisaged to reduce the amount of americium in the waste is to re-irradiate it in fast neutron reactors in order to transform it into a less radiotoxic element thanks to a transmutation reaction [1]. Two main transmutation approaches are currently studied. Homogeneous transmutation consists in including a small Americium content (< 5%) into the nuclear fuel to transmute it during power production. Heterogeneous transmutation aims at including higher percentages (around 10 to 20%) of Americium into either a matrix located in specific pins inside the core, or in minor actinides bearing blankets located on the core periphery. Another possibility would be to use an accelerator driven system, an under-critical reactor core driven by a proton accelerator coupled to a spallation source and used for the transmutation of compounds containing high concentrations of minor actinides (up to 50%) [2].

It is therefore necessary to evaluate the impact of Americium content and deviation from stoichiometry on the behaviour of nuclear fuels, and in particular on their high temperature thermal properties. This requires, among other things, a very good knowledge of the Uranium – Plutonium – Americium – Oxygen thermodynamic system. For this, a reliable description of the ternary systems (U-Pu-O), (U-Am-O) and (Pu-Am-O) is a prerequisite. The (U-Pu-O) system has been revisited extensively in recent years and is relatively well described [3]. Much less data is available on the Americium-bearing oxides, especially at high temperature and for large deviations of the oxygen stoichiometry [4].

Atomic scale modelling methods are now essential tools to complement experimental characterizations of nuclear fuels and get further insight into their properties and behaviour. We will present the results of the investigation of structural and thermodynamic properties of non-stoichiometric Americium bearing oxides using a combination of electronic structure calculations and empirical interatomic potentials. Concerning electronic structure calculations, we used the DFT+U method combined with the orbital control scheme [5]. For empirical atomic potentials, we developed a potential in the Cooper-Rushton-Grimes formalism [6] to allow for the systematic study of Uranium-Americium mixed oxides.

This project has received funding from the Euratom research and training programme 2019-2020 under grant agreement No 945077.

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P15 Defect chemistry and radiation stability of (Gd & Zr) co-doped UO_2 solid solutions

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Nuclear fuels undergo complex microstructural changes as a result of the extreme environments of intense radiation and high temperature during in-reactor operation. Several studies are being performed to look into the effect of crystalline defects and fission products on the lifespan of fuel rod elements. The irradiation behaviour of pure UO_2 is rather well experimentally characterised, but little is known about the irradiation stability of UO_2 doped with fission products such as rare-earth elements. This study is devoted to investigating the effect of heavy-ion irradiation of UO_2 (to simulate radiation damage within a reactor) when doped with both trivalent Gd (a common burnable poison) and tetravalent Zr (a high concentration fission product) at varied concentrations.

Surface characterisations of the synthesised co-doped UO_2 pellets revealed that the dopants incorporation altered the initial UO_2 microstructure. The intrinsic defect chemistry manipulation caused by multivalence dopants, as well as their behaviour as a result of Kr-ionic implantation, were studied using experimental XRD, HERFD-XANES, and Raman spectroscopy coupled with theoretical DFT+U calculations. The key mechanisms that occur near defect-boundary interfaces will be discussed in this talk in order to highlight the precise role of dopants and grain boundary areas in changing the lattice defects mechanisms in fluorite structures relevant to nuclear fuels.

P16 Corrosion behaviour of candidate MSR structural materials exposed to molten chloride salts

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Generation IV nuclear reactors are a promising concept that have gained significant attention in recent years. Research efforts into their design has brought a set of intrinsic benefits compared to traditional Light-Water Reactors (LWRs), especially in terms of safety and sustainability. Among the Generation IV type reactors, the Molten Salt Reactor (MSR) uses molten salts as fuel and coolant. To this date, fluorides and more recently chlorides, are the reference choice for the fuel matrix. However, a key challenge in the design of these reactors is the corrosive nature of the salts, which combined with the high operation temperature, may cause corrosion in an accelerated manner, thus compromising the integrity of the reactors' structural materials.

This study, carried out within the European MIMOSA project [1], focuses on the corrosion behaviour of various structural materials, namely ceramics, stainless steels and Ni-based alloys, exposed to molten NaCl-$MgCl_2$-$CeCl_3$ salt, where $CeCl_3$ is used as a surrogate for $PuCl_3$. The aim of the research is to do a comparative study on the corrosion behaviour of several candidate materials exposed to the molten salt at high temperatures under a controlled argon environment by characterizing the alloys, as well as the salts, before and after the exposure experiments.

The research methodology involves the systematic evaluation of corrosion resistance by analytical techniques such as Optical Microscopy (OM), Scanning Electron Microscopy – Energy Dispersive X-ray Spectroscopy (SEM-EDS), X-ray Diffraction (XRD), Differential Scanning Calorimetry (DSC) and Inductively Coupled Plasma Mass Spectrometry (ICP-MS).

This work is funded by the European Union. Views and opinions expressed are however those of the author(s) only and do not necessarily reflect those of the European Union or EC-Euratom. Neither the European Union nor EC-Euratom can be held responsible for them.

[1] MIMOSA (MultI-recycling strategies of LWR SNF focusing on Molten Salt technology) EURATOM-2021-NRT-01 project, https://www.mimosa-euratom.eu/

P17 TEM characterizations of dislocation motion and sub-grain boundaries induced by mechanical strain in UO_2 at 1550°C

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The knowledge of the viscoplastic behaviour of the UO_2 fuel pellets is a significant nuclear safety issue, particularly during incidental operating conditions of Pressurized Water reactors (PWR). In particular, in pile irradiation may induce important thermal gradients inside the pellets leading in turn to a local mechanical interaction with the cladding. This pellet-to-cladding interaction can be further accentuated during an incident power transient. This latter is characterized by a high power rise within a time frame of about a few tens of milliseconds to several minutes (according to the incident type), leading to important stresses within the fuel and accelerates the viscoplastic deformation of the pellet centre where the temperature can typically reach 1200 -1700 °C.

However, the fuel viscoplastic behaviour is a complex process due to the heterogeneous fuel microstructure and its evolution under irradiation. A few experimental studies of the microstructure induced by deformation tests in fresh UO_2 exist. Different dislocation substructures were observed using transmission electron microscopy (TEM) [1][2][3] or accurate electron channelling contrast imaging (accurate-ECCI) [4][5][6]. Many hexagonal networks were observed and were attributed to cross slip in {111} type planes when double slip conditions in {100} or {110} type planes are gathered [2]. However, more complex networks were also observed, especially at high temperature possibly involving climb, with sometimes parallel dislocations or square network parts. Unfortunately, neither detailed analysis about elementary dislocation processes of motion nor complete analysis about the complex sub-grain boundaries are available so far in the literature.

In this study, characterizations using TEM were performed on polycrystalline UO_2 pellets after uniaxial compressive tests with a constant strain rate at 1550°C. For the first time, it was shown that ordinary dislocations can move in UO_2 by a mixed mechanism involving climb, called mixed climb [7]. Various sub-grain boundaries were observed, some of them involving Hirth junctions, where Burgers vector stand along the <100> direction, and up to three independent families of dislocations. The detailed characterization of ordinary dislocation motion and of dislocation networks will help us to provide a better knowledge of the plastic deformation mechanisms at the nanoscopic scale.

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P18 Kr and Xe diffusion in UO_2 and UO_{2+x} by Thermo-Desorption Spectrometry: summary of a decade of measurements

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Due to their insoluble nature in nuclear fuels, gaseous fission products (Kr, Xe) cause unfavourable evolutions of fuel properties (microstructure, thermal conductivity) that impair fuel performances and constrain operational safety margins (through the risk of fuel pin bursting in case of gas release from the fuel).

The study of Kr and Xe behaviour in UO_2 and derived compositions has thus generated a considerable amount of experimental and computational research since the dawn of nuclear power; however, there is still some relevant experimental conditions that needs to be better understood. This notably encompass experimental diffusion in very low concentrations (allowing to reach defect-free interstitial lattice diffusion constants) or in overstoichiometric compositions (UO_{2+x}, relevant as the high burn-ups UO_2 fuels can reach overstoichiometry of circa x = 0.001).

During the last decade, we have tried to fill these gaps by investigating the separate effects of several parameters on fission gas diffusion in Kr or Xe ion-implanted UO_2 pellets: Xe vs. Kr, temperature [1], fluence [2], UO_2 bulk and surface microstructure [2], and oxygen overstoichiometry (either from surface oxidation during storage or by oxygen controlled additions during annealings [3]). Xe and Kr diffusion kinetics and behaviours were measured using Thermo-Desorption Spectrometry, a technique that allowed us to work with fluences down to few 10^{10} ions.cm^{-2}.

We thus propose to present at NuFuel a summary of these works and an outlook of what should be done next (application to MOX or irradiated fuels, to alternative fuels etc.).

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P19 Study of the corrosion by fission products of the steel fuel cladding in SFR Reactors

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In the frame of Sodium-cooled Fast Reactors (SFRs), this study aims at understand corrosion mechanism of fuel steel cladding by fission products (FP) formed in the fuel pellet. Indeed, the migration of the volatile fission products (Cs, I, Te, Mo, Ba, Pd) towards the periphery leads to accumulation of these elements between the fuel pellet periphery and the steel cladding, leading to the formation of a layer enriched in fission products, so-called JOG ("Joint Oxyde Gaine" in French). These fission products can then react with the steel cladding to form a corrosion layer, so-called ROG ("Reaction Oxyde Gaine" in French). A detailed understanding of these two phenomena, first the fission product accumulation and then the corrosion reaction with the cladding, is essential for extending the lifetime of fuel cladding. Previous laboratory studies have faced challenges in validating corrosion mechanisms due to the difficulty of reproducing simplified but representative corrosion conditions. This study aims first at reproduce experimentally the composition of the JOG using thermodynamic simulations with the TAF-ID database. The JOG will be synthesized by mixing fission product compounds (Cs_2MoO_4, $BaMoO_4$, Te, Pd, CsI …). Secondly, understanding of corrosion mechanism is planned through corrosion tests between the JOG and the steel cladding. To achieve these objectives, the following steps are considered:

(1) Synthesis of the simulated volatile fission products enriched layer (JOG) coupled with Thermo-Calc simulations to better understand its formation conditions (oxygen pressure and temperature);

(2) Identification of possible compounds to be formed during the interaction between the steel cladding and the corrosive environment (synthetized in step 1);

(3) Corrosion tests in controlled environment (with controlled oxygen potential, temperature and simulated fission products JOG) to achieve the same corrosion facies in a simplified dedicated set-up as the one observed in reactors;

(4) Determination of steel corrosion kinetics;

(5) Identification and understanding of cladding corrosion mechanisms.

P20 Thermo-physical properties of $U_{1-y}Pu_yO_{2-x}$ mixed oxides with high plutonium contents ($y \geq 0.6$)

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$U_{1-y}Pu_yO_{2-x}$ mixed oxides (MOX) are the reference fuels for Sodium-cooled Fast neutron Reactors (SFRs). Their plutonium fraction (y = Pu/U+Pu) and their Oxygen/Metal (U+Pu) ratio must range between 0.20 to 0.35 and $1.94 \leq O/M < 2.00$, respectively. During irradiation, a thermal gradient along the fuel pellets radius (typically ~ 4 mm) occurs, with a temperature reaching 773 K close to the cladding and 2073-2273 K at the center. This thermal gradient induces a redistribution of plutonium and oxygen atoms along the radius. Therefore, the center of the pellet is enriched in plutonium with a local Pu fraction in the 0.30-0.70 range (depending on the location in the core and its initial Pu content and O/M ratio) and depleted in oxygen leading to a reduced O/M ratio (< 1.95). Close to the cladding, Pu fraction is 0.20, and the fuel O/M ratio is close to 2.00. These mass transport phenomena lead to significant heterogeneities in the thermo-physical properties of the fuel along the pellet radius. In particular, an enriched local plutonium content induces a modification in the local melting temperature, which must be known precisely to assess whether the safety margin of the reactor is maintained. The investigation of the thermodynamic and structural properties of U-Pu mixed oxides with high plutonium content is thus mandatory. In a preliminary step, single-phase dense MOX pellets with Pu fraction of 0.60, 0.65 and 0.70 and two initial O/M ratios (2.00 and < 2.00) were manufactured by a powder metallurgy process. Their structural and microstructural properties were fully characterized at ATALANTE facilities by X-ray diffraction, μ-Raman spectroscopy, scanning electron microscopy and electron probe microanalysis.

Measurements of thermo-physical properties were then performed at the JRC Karlsruhe on these samples, including melting temperature and thermal diffusivity by laser flash methods. In order to study of the influence of the deviation from stoichiometry, measurements were performed with two atmospheres in the measurement vessel: under inert (Ar) and reducing (Ar + H_2). The quantification of the chemical composition and oxygen stoichiometry of the samples before and after melting was obtained by X-Ray fluorescence and HERFD-XANES (cationic species) measurements performed at the MARS-SOLEIL synchrotron beamline.

During the conference, the results obtain will be presented and the influence of the Pu content and O/M ratio on these properties will be highlighted. A comparison with the thermodynamic modelling using the CALPHAD method will also be discussed.

This research work is carried out as part of the European PuMMA project (Plutonium Management for More Agility) which has received funding from the European Union's Horizon 2020 (grant agreement No 945022).

P21 Development of predictive nuclear forensic signatures for uranium oxides

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Research and development of nuclear forensics signatures has been an area undergoing rapid expansion in recent years. Such signatures can be vital in assisting law enforcement agencies to identify illicit use of nuclear materials. Typically, this relies on material databases from physical measurements. With the ever growing adaptation of nuclear fuels developing these signatures through physical measurements will be time consuming and costly. Predictive nuclear forensics looks to identify the key signatures of interest through computational means underwritten by physical measurements to reduce the future burden.

P22 Thermochemistry of volatile fission products Cs and I in Lead-cooled Fast Reactors

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Lead-cooled Fast Reactors (LFRs) are one of the Generation IV nuclear reactor designs that are to be cooled with either liquid Pb or a liquid eutectic mixture of Pb and Bi. In Europe, mixed oxide fuel $(U,Pu)O_2$ is currently the reference for such type of reactors. During irradiation, several classes of fission products are generated, making the fuel a multi-component system: metallic precipitates, volatile fission products, oxide products and fission products that dissolve in the fuel matrix [1].

In case of clad breach, the fuel and fission products can come into contact with the coolant, giving rise to a potentially very complex chemistry. An important safety consideration is the chemical interaction between coolant and volatile fission products Cs and I, giving rise to the chemistry of the Pb-Bi-Cs-I system. Fundamental structural and thermodynamic studies into the relevant systems are a pre-requisite for the evidence-based thermodynamic databases that are used in the nuclear community.

Recently, we have investigated the phase equilibria in the CsI-PbI_2-BiI_3 system using differential scanning calorimetry and X-ray diffraction [2], and we have measured the standard entropies of $CsPbI_3$, Cs_4PbI_6 and $Cs_3Bi_2I_9$ [3]. Further thermodynamic studies in the solid and gas phase are planned for the coming months. Using the acquired data, thermodynamic models of the binary sub-systems and ternary system have been developed using the CALPHAD approach.

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P23 Characterisation and modelling of the "Joint Oxyde-Gaine" and the corrosion layer of the inner clad in Sodium Fast cooled Reactor fuels

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Within the PLEIADES simulation platform co-developed by CEA, EDF and FRAMATOME [1], GERMINAL is the fuel performance code dedicated to the simulation of the in-pile behaviour of mixed oxide fuel pins for Sodium-cooled Fast Reactors [2].

The feedback on such fuel elements, based on experimental observations mainly acquired during the French PHÉNIX reactor operation, shows the formation of the JOG (“Joint Oxyde-Gaine”) between the fuel pellet and the cladding from a burn-up of 6 to 8 %FIMA [3], which has an impact on the thermo-mechanical behaviour of the whole fuel element. At higher burn-ups, the volatile fission products (VFPs) compounds can react with the cladding material components (Fe, Ni, Cr), leading to inner clad corrosion through the FCCI (“Fuel Cladding Chemical Interaction”).

A first model has been implemented in the GERMINAL fuel performance code to empirically describe the JOG and FCCI layers [4]. However, to improve our chemical modelling of the chemical behaviour of uranium and plutonium mixed oxide fuel under SFR irradiation conditions, the thermochemical software OpenCalphad (OC) [5] and the TAF-ID database [6] have been integrated into the GERMINAL code [7] as a complementary option of the previous empirical description model.

Destructive examinations on a standard PHÉNIX fuel pin irradiated up to 13 %FIMA have been performed in order to optimize the sample preparation and the measurement methodologies by microanalysis techniques (EPMA, SIMS, SEM-FIB, TEM) in order to accurately characterize these complex and multiphase layers. The aim of this study is to compare the description of JOG and FCCI obtained from the thermochemical model with post-irradiation examinations for different local burn-ups and pin types.

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