Institut national de physique nucléaire et de physique des particules

COLLABORATION AGREEMENT

IN2P3 - COPIN

I. Identification of the laboratories	
Partner	COPIN
IN2P3 laboratories	**IJCLab**
Partner laboratories	NCBJ Varsaw

II. Identification of the collaboration	
Title of the collaboration	Simulation of the behavior of the spent nuclear fuel using energetic ion beams
Number of the collaboration	05-118
IN2P3 spokesperson	F. GARRIDO
COPIN spokesperson	A. TUROS
Scientific Domain	Nuclear Physics

Status of the collaboration	
Status	The renewal of the collaboration is requested for the period January 1st - December 31st, 2023

III. Status report for the period January 1st to December 31st, 2022	
III.1 IN2P3 scientists in COPIN	
Total time approved for 2022	21
Total time used for 2022	7
List of scientists	1. F. GARRIDO (7 days)
III.2 COPIN scientists in France	
Total time approved for 2022	30
Total time used for 2022	26
List of scientists	1. P. JOZWIK (13 days) 2. C. MIESZCZYNSKI (13 days)
III.3 Scientific results of the above-mentioned collaboration	
Description	

In the section below we summarise the aim of this research project and the developments performed in 2022.

This research project aims to investigate the stability of nuclear fuel by using both experimental and computational approaches. Uranium dioxide (UO

$_2$)

single crystals are used as a simplified model of spent nuclear fuel. Radiation damage and incorporation of foreign elements are studied using ion accelerators, in particular, ion implanters and Van de Graaff / Tandem accelerators providing a wide variety of analysis techniques. Such an approach is fruitful to imitate and investigate the conditions in a nuclear reactor by the high-temperature bombardment of the materials of interest using high-energetic particles. Further analysis using, e.g., Rutherford Backscattering Spectrometry in Channeling mode (RBS/C) technique allows getting clues on the role played by various chemical and physical parameters involved in fuel destabilisation. The applied methodology can also be used for the investigation of a large class of irradiated solids beyond nuclear materials (e.g. materials for accelerators and detectors).

UO_2 and mixed oxides (MOX: a solid solution of $(U,Pu)O_2$) are the main nuclear fuels used in today's power plants. Despite decades of investigation the radiation behaviour of UO_2 is still not fully understood, mostly because of the complexity of the spent nuclear fuel in terms of composition and microstructure. The structural stability of the nuclear fuel can be affected by several parameters including (i) irradiation effects due to fission fragments, alpha decay, neutrons, beta decay of fission products, (ii) the presence of incorporated impurities, fission products and transuranium nuclei (actinides), (iii) temperature gradient during in-reactor operation.

Energetic ion beams delivered by accelerators and the use of a simplified system – UO_2 single crystals – provide new ways of modelling the behaviour of the fuel during or after in-reactor operations. Indeed, ion beams allow addressing three major issues: (i) irradiation-induced damage on the various regimes of slowing-down of bombarding ions (electronic and nuclear energy loss); (ii) incorporation of selected impurities by ion implantation; (iii) investigation of the structural and microstructural evolution of solid using nuclear microanalysis techniques (Ion Beam Analysis). Let us stress that the choice of the beam nature and energy allows us to reproduce pure irradiation conditions, as well as pure doping ones, but also to reproduce a wide range of situations where both the physical and chemical processes are mixed. The main idea is to reproduce the physical and chemical features of a solid exposed to given irradiation conditions.

The main advantage of using these experimental techniques coupled to model systems is: (i) the absence of added radioactivity; (ii) the possibility to develop parametric studies, since all the parameters can be modified (e.g. on nature, energy, fluence, flux, the temperature under irradiation). Most of our experiments are carried out at the JANNuS-Orsay at IJCLab and GANIL facilities. The JANNuS platform provides a unique setup coupling with a goniometer on which a single crystal is mounted and two beam lines: one is connected to the ion implanter to incorporate foreign elements of interest, and the second can be used to bombard in situ the single crystal with probing ions (typically He ions) to perform RBS/C analysis. The McChasy simulation code, developed during years at NCBJ under the framework of this collaboration, can calculate the trajectories of probing projectiles in crystalline structures that contain many different types of defects and provide a unique advanced way to interpret channelling data. It is one of the very few Monte Carlo codes in the world that calculates trajectories of probing ions in single crystals to investigate the structure of a solid, essentially providing a crystallographic investigation of the crystal structure in direct space. Two stable variants of the McChasy code provide a wide spectrum of RBS/C data analysis with a different approach.

McChasy-1 performs Monte Carlo simulations in small (~10^2 atoms) virtual crystalline structures created using a selected crystallographic database, e.g., [1]. During ongoing simulations, the structures are repeated to reproduce a full depth of the crystal while the realistic effects (e.g., thermal vibrations and the presence of defects) are applied based on previously developed models. The models of defects already available in the McChasy-1 code include the most common crystallographic defects, e.g., substitutional impurity atoms, randomly displaced atoms, edge dislocations, dislocation loops, and xenon (Xe) precipitations (bubbles) in UO_2. Other models of defects and more details about the McChasy-1 code's features can be found in Ref. [2]. The McChasy-1 code will be developed as part of this project, as described in Milestone 1(i) for 2003.

McChasy-2 is capable to perform Monte Carlo simulations in large (~10^8 atoms) virtual crystalline structures that can be created using a selected crystallographic database by custom build-in McChasy procedures or selected external apps (ATOMSK [3] and LAMMPS [4] codes). More detailes about the McChasy-2 software can be found in Ref. [5]. A powerful feature of the McChasy-2 code is the possibility to perform simulations for structures containing realistic defects, as created by MD simulations. The feature will be further developed as part of this project, as described in Milestone 1(ii) for 2003.

In this general scheme some specific questions are investigated in the framework of this collaboration:

1 – The respective roles played by the chemical impurities and by irradiation-induced damage on the matrix destabilisation. This problem is tackled at JANNuS-Orsay by implantation of selected impurities on a urania single crystal at high temperature (500 °C to reproduce the temperature of the outer part of the fuel during in-reactor operations) and in-situ characterization by RBS/C. The main idea is to compare the behaviour of fully soluble and insoluble atoms in the urania lattice to decouple both contributions to radiation-induced destabilisation. More precisely, we compared the behaviour of

the insoluble noble gas Xe with fully soluble species La (that exist mostly as a 3^+ valence state) and Ce (4^+ valence state) – all three atoms having essentially the same A and Z numbers – by performing 500 keV ion irradiation (therefore, in the ballistic regime). Results show that in the low fluence range, all ions create the same radiation-induced damage, irrespectively of their chemical nature, but at large fluence, the formation of Xe bubbles (thus a chemical effect) plays the major role in the fuel destabilization due to the mechanical stress induced by over-pressurisation.

2 – New experiments were started in the framework of the development of the new Accident Tolerant Fuels (ATF) that will be used in PWR reactors in the nearest future. Chromium was added as an impurity in the fuel (typically in the 1% range) but its impact on the radiation tolerance of the nuclear fuel is not. Implantation of Cr in UO_2 single crystals at the JANNuS-Orsay facility was performed and the lattice location of Cr was designed by ion channelling using RBS to sense the lattice and PIXE to probe the energy state of Cr impurities. The McChasy-1 code was used to fit angular scans and test the various possible locations in different materials including the fluorite-type structure.

3 – The further development of the McChasy-2 software to simulate the channelling spectra measured during RBS/C experiments and to study RBS/C response for large defects in crystals. Up to recently, the available version of the code (McChasy-1) was designed to implement a specific type of structural defects (e.g., atoms displaced randomly or at a given location in the structure, dislocations, mosaic structure in the single crystal) in the otherwise perfect monocrystalline structure and it calculates spectra and angular scans that are compared to experimental data to improve step by step the model of a disordered crystal. A new version of the McChasy code (McChasy-2) was developed and tested. It differs from McChasy-1 by the fact, that it can be used to study defects whose sizes are in the range of 0.1–20 nm, especially dislocations and dislocation loops. Moreover, the code can model ion-channelling in structures obtained with the MD simulation technique. This fact opens a new gate. Such a modification models in a realistic way the presence of dislocations in the solid (dislocations essentially break the lattice translation symmetry, thus large crystal cells are needed to fully reproduce the structure of a real crystal), one very common radiation-induced defect type. This transformation implies the full transformation of the code to run it on PC workstations to define crystal samples involving 10^6-10^8 atoms. A novelty, compared to previous years, is the extension of the Monte Carlo code with a part that allows the introduction of virtual samples of materials obtained by MD into the simulation in the McChasy-2 code. Such an extension has been already tested on monoelemental samples (of Si and Ni).

IV. Renewal of the collaboration for 2023

IV.1 Proposed scientific program

Description

For the future years (2023 and beyond), we propose to extend the program of the collaboration to a larger class of nuclear materials keeping the same experimental and computational methods since it can be applied to most systems of interest. Two main nuclear materials are selected here:

1. uranium dioxide, the nuclear fuel, since we already collected a lot of experimental data on this system whose strong radiation tolerance still needs to be elucidated concerning mechanisms of the structural reorganisation at the atomic scale (for instance, the formation of the High Burnup Structure in UO_2 nuclear fuel still deserves detailed investigations as well as the role played by chromium in accident tolerant fuels);

2. nickel (and Ni-based superalloys) as a key material for the vessel and internal parts of the nuclear reactor (fuel subassemblies and structures). The proposed strategy to achieve a description as complete as possible of the behaviour of a solid exposed to irradiation requires the use of a very simplified model system (here we select intentionally monocrystalline solids) irradiated in well-defined conditions provided by energetic ion beams delivered by accelerators and submitted to the most advanced characterization techniques (ion channelling and transmission electron microscopy – both techniques can be performed in-situ or ex-situ at the JANNuS facility) to identify the radiation defects and to follow their evolution. One of the key points is the capability to model extended defects (e.g., dislocation loops and lines) realistically by creating virtual crystals defined through MD simulations and by comparing them with experimental data. Such an approach requires a new Monte Carlo simulation code (McChasy 2), which is capable to work with large (nm-size) structures. The development of such code will be the cornerstone of the project.

3. Various metallic alloys used as structural materials in nuclear engineering (ferritic-martensitic steels, ODS, HEAs, ceramics) irradiated with heavy ions under controlled stress conditions. The stress will be applied by a tensometric stage adapted to work in a vacuum environment (ordered by NCBJ in CNTech company UK). Main objective of this study is to identify the role of the stress on kind of radiation defects created in the material and thresholds of defect transformations occurring with the increasing radiation dose. Research methodology will consist in irradiation of the samples

loaded with a given stress in a tensometric stage, extraction of FIB-made lamellaes using SEM/FIB device and observation of the defects ina Transmission Electron Microscopy. This part of the collaboration will be partially done jointly with the CEA Saclay (Jean-Luc Bechade) in a dedicated irradiation chamber that will be able to host the tensometric stage.

In 2023 the proposed program focuses on following **Milestones**:

1. The further development of the McChasy computation code and its validation against experimental data recorded on UO_2, Ni, and Ni-alloys single crystals.

1. Further development of the McChasy-1 code: the models of dislocation, dislocation loops and precipitations of gases in crystalline materials available in the McChasy-1 code will be developed as part of this project. In particular, geometrical parameters of dislocations and loops will be obtained from Molecular Dynamics (MD) simulations as well as Transmission Electron Microscopy (TEM) providing also a unique comparison of the computational and experimental approaches as a tool to assess lattice distortion in the vicinity of dislocations/loops. The model of Xe precipitations in UO_2, a defect commonly observed in materials exposed to high-energetic radiation, will be extended to precipitations of other gases in a wider variety of nuclear materials (with special attention paid to Ni and its alloys).

2. Testing and further development of the McChasy-2 code: Indeed, the first versions of McChasy-2 are running at present but extensive testing of the code in various conditions is mandatory. A validation procedure will be performed by comparison of the calculated spectra with the ones obtained in the same conditions on the previous version of the code McChasy-1. Moreover, due to the very large number of RBS/C spectra and angular scans collected on UO_2 single crystals either in a pristine state or irradiated in various irradiation conditions (ballistic damage created by low energy ions, damage generated by electronic processes, swift heavy ions delivered by the GANIL accelerator) or oxidised in well-controlled conditions (such as U_4O_9 crystals), this unique model system provides us with a way to test the McChasy-2 code against well-qualified data. Moreover, extreme conditions in a nuclear reactor will be modelled by MD simulations of interactions of high-energetic particles (mimicking the fission products) with crystalline structures of UO_2, Ni, and Ni alloys. Defects created in this way will be analysed by the McChasy-2 code and compared with the experimental data delivered by TEM for selected depths of the samples.

Study of dislocation and dislocation loop models as well as a parametrisation of the ion irradiation-based extended defects in the Ni and Ni-based single crystal fcc structures. The unique combination of two experimental techniques such as TEM and RBS/C mode as well as MD simulations allows us to track the evolution of dislocation loops in the irradiated material and their parameterisation. We can compare the defects obtained by MD with those in the real material and then use the simulated structures to perform Monte Carlo channelling simulations of experimentally obtained spectra.

The study of the lattice location of Cr in UO_2 single crystals, including both experiments and data analysis using the McChasy code to validate its position in the crystal structure at different concentrations.

Acquisition of the tensometric stage, its adaptation to the existing devices in the NCBJ (ion implanter, XRD, Raman), irradiation with selected ions (He, Ar), extraction of lamellaes via FIB technique, TEM analysis of the defects created.

Large computational developments of the McChasy, Monte Carlo simulation code are performed to allow the defective structure of irradiated solid, as proposed by MD calculations, to be used as an input in the code, allowing a direct calculation of the expected channelling spectra. Such a new feature will open the possibility to use channelling data as a validation tool of the defects proposed by ab-initio computation and opens a new insight for the development of channelling and the modelling of the complex structures of irradiated solids. An illustrative example is the predicted evolution of the behaviour of the nuclear fuel under low-energy ions as calculated by MD calculations, describing the formation of extended defects as dislocations loops, lines and finally dislocation network while increasing the ion fluence (and thus the value of a displacement per atom, DPA). McChasy-2 provides the possibility to generate calculated RBS/C spectra for a direct comparison with spectra recorded on a genuine experiment performed at the

JANNuS-Orsay facility and thus validate (or exclude) the atomistic scenario as predicted by MD. Such an approach, coupling different experimental (IBA, TEM) and modelling (MD, McChasy-2) open a new field in the deep understanding of the evolution of solids under irradiation.

The studies of the role of the stress on kind of radiation defects created will be done using mostly the equipment available in the NCBJ: mainly ion implanter equipped with a new tensometric stage, SEM/FIB/EDS/EBSD system (ThermoFisher Helios 5 UX), new TEM system (Jeol F200) arriving in 2023. These analyses will be completed by in-situ experiments in IJCLab (JANNuS accelerator coupled with FEI TEM). The experiments carried out in NCBJ will involve bulk samples, whereas the experiments in IJCLab will be done on thin TEM specimens.

REFERENCES

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IV.2 Estimated duration for IN2P3 scientists in COPIN	
Total time requested for 2023	30
List of scientists	1. F. GARRIDO (16 days) 2. A. GENTILS (7 days) 3. M. HITIER (7 days)
IV.3 Estimated duration for COPIN scientists in France	
Total time requested for 2023	60
List of scientists	1. J. JAGIELSKI (15 days) 2. I. JOZWIK (15 days) 3. P. JOZWIK (15 days) 4. C. MIESZCZYNSKI (15 days)

Comment Validation	
Unity Director	Fadi IBRAHIM (IJCLab) - 2022-10-13 17:18:56