EPJ Nuclear Sci. Technol. **9**, 13 (2023)

https://doi.org/10.1051/epjn/2022029

EPJ Nuclear Sciences & Technologies

Euratom Research and Training in 2022: challenges, achievements and future perspectives, Roger Garbil, Seif Ben Hadj Hassine, Patrick Blaise and Cécile Ferry (Guest editors)

Available online at: https://www.epj-n.org

REVIEW ARTICLE

OPEN ACCESS

Spent nuclear fuel management, characterisation, and dissolution behaviour: progress and achievement from SFC and DisCo

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Received: 22 April 2022 / Received in final form: 12 July 2022 / Accepted: 22 September 2022

Abstract. SFC is a work package in Eurad that investigates issues related to the properties of the spent nuclear fuel in the back-end of the nuclear fuel cycle. Decay heat, nuclide inventory, and fuel integrity (mechanical and otherwise), and not least the related uncertainties, are among the primary focal points of SFC. These have very significant importance for the safety and operational aspect of the back-end. One consequence is the operation economy of the back-end, where deeper understanding and quantification allow for significant optimization, meaning that significant parts of the costs can be reduced. In this paper, SFC is described, and examples of results are presented at about half-time of the work package, which will finish in 2024. The DisCo project started in 2017 and finished in November 2021 and was funded under the Horizon 2020 Euratom program. It investigated if the properties of modern fuel types, namely doped fuel, and MOX, cause any significant difference in the dissolution behavior of the fuel matrix compared with standard fuels. Spent nuclear fuel experiments were complemented with studies on model materials as well as the development of models describing the solid state, the dissolution process, and reactive transport in the near field. This research has improved the understanding of processes occurring at the interface between spent nuclear fuel and aqueous solution, such as redox reactions. Overall, the results show that from a long-term fuel matrix dissolution point of view, there is no significant difference between MOX fuel, Cr+Al-doped fuel, and standard fuels.

1 SFC

Eurad is a large European Commission project aiming to joint programming of nuclear waste management in the European Union. The project consists of many work packages, of which Spent fuel Characterization (SFC) is the largest. SFC, in turn, consists of four tasks: Task 1 – S/T coordination, State-of-the-art and training material, Task 2 – Fuel properties characterization and related uncertainty analysis, Task 3 – Behavior of SNF pellets under interim storage conditions, Task 4 – Accident scenario and consequence analysis.

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SFC investigates issues related to the properties of the spent nuclear fuel (SNF) in the back end of the nuclear fuel cycle. Decay heat, nuclide inventory, and fuel integrity (mechanical and otherwise), and not least the related uncertainties, are among the primary focal points of SFC. These have very significant importance for the safety and operational aspect of the back-end. For example, the thermal situation is most often the limiting factor in the various parts of the back-end. Important parts of the back-end include cooling, transportation, intermediate storage (wet and dry), encapsulation, reprocessing (for some countries), and final disposal. One consequence is the operation economy of the back-end, where deeper understanding and quantification allow for significant optimization, meaning that significant parts of the costs can be reduced. The results are relevant for countries with small as well as large and advanced nuclear waste management programs. Great care has been put into doing research complementary to research already done or done elsewhere, not repeating previous efforts.

In this paper, SFC is described, and examples of results are presented at this point in time, which is about half-time of the work package; it will finish in 2024.

1.1 SFC Task 2: fuel properties characterization and related uncertainty analysis

The main objective of this task is to produce experimentally verified and validated procedures to determine reliable source terms of SNF, including realistic uncertainties. The focus is on source terms which are of primary importance for safe, secure, ecological, and economical handling, transport, intermediate storage, and final disposal of SNF [1]. The main source terms of interest are neutron emission rates, γ-ray emission spectra, decay heat rate, and the inventory of specific nuclides, i.e., activation products (e.g., ^{14}C and ^{36}Cl), long-lived fission products (FP), fissile nuclides (^{235}U, ^{239}Pu) and minor actinides (^{241,243}Am, ^{242,244}Cm). The inventory of fissile nuclides is needed for nuclear safeguards requirements and reactivity calculations to prevent criticality. To avoid overly conservative criticality margins, a burnup credit approach can be applied. Such an approach requires the inventory of strong neutron-absorbing fission products and actinides. The inventory of activation products and long-lived FP is important to study the impact on the biosphere. Some of the FP (e.g., ^{148}Nd) are used for burnup determination.

The complete list of source terms is hard to be measured directly, in particular during industrial operation. Therefore, they are estimated based on a combination of calculations and results of non-destructive analysis (NDA) measurements to verify the calculations. It requires the calculation of a complex inventory of nuclides with strongly varying characteristics. The calculations involve neutron transport and nuclide creation and depletion codes. The results of such calculations strongly depend on nuclear data (including cross sections, FP yields, neutron emission probabilities and spectra, decay data etc.), fuel fabrication data (design, composition), and reactor operation and irradiation conditions (burnup, neutron spectrum). The design, composition, production, operation, and irradiation conditions will be referred to as "fuel history".

1.1.1 SFC Subtask 2.1: theoretical study of SNF source terms

The goal of subtask 2.1 is to provide recommendations in terms of neutronics transport and decay calculations for estimating the SNF composition (nuclide inventory) and its decay heat. Such recommendations should lead to well-justified best estimate values, with uncertainties and biases, later used in subtask 2.4. To reach this ambitious goal, a number of PIE samples were selected either from the SFCOMPO database [2] or from proprietary descriptions available within the EURAD SFC project. These samples were selected to cover the characteristics of a large number of existing SNF in terms of assembly types (for BWR or PWR), burnup values, initial enrichment, and fuel type (UO_2 or MOX). They help compare calculated nuclide concentrations with measured ones within the context of specific calculation tools, nuclear data libraries, and modeling assumptions. Such comparisons lead to the estimation of biases for the nuclide concentrations and finally to recommendations. Additionally, if uncertainties were considered for the model description (e.g., fuel temperature, void fraction, but also nuclear data), it would also be possible to provide a calculated uncertainty for the nuclide concentrations based on the input variations. Another source of uncertainty can be estimated if different partners independently analyze the same PIE sample. Such overlap will most likely lead to different calculated concentrations, thus indicating the user effect, which is also considered an uncertainty.

The second calculated quantity, the SNF decay heat, can be analyzed in a similar manner. This will also lead to best estimate values, uncertainties, and biases. Naturally, these two quantities (nuclide concentrations and decay heat) are connected, at least from the calculation aspect. One additional constraint on the decay heat, at least from the user's point of view, can be the necessity to use standards such as the ANS (see `www.ans.org` for details) or the DIN (see `www.din.de` for details) standards. A deviation between the results from dedicated simulations and standards will necessitate understanding and justification, which can be backed up by the present project. To illustrate such possibility, an example for a specific PWR assembly is presented in Figure 1. In this example, differences between measured and calculated decay heat can be observed, but also differences between standard values (ANS and DIN) and both CASMO5 and SNF calculations. If successful, explanations will be provided by the present project.

In total, 17 PIE samples were analyzed, some of them by different participants. For decay heat, the calorimetric measurements from CLAB (Swedish Central Intermediate Storage Facility), GE-Morris, and HEDL (Hanford Engineering Development Laboratory) were considered, leading to more than 250 cases. To support the recommendations of this subtask, some studies were already published, see references [3–15]. A global review of the

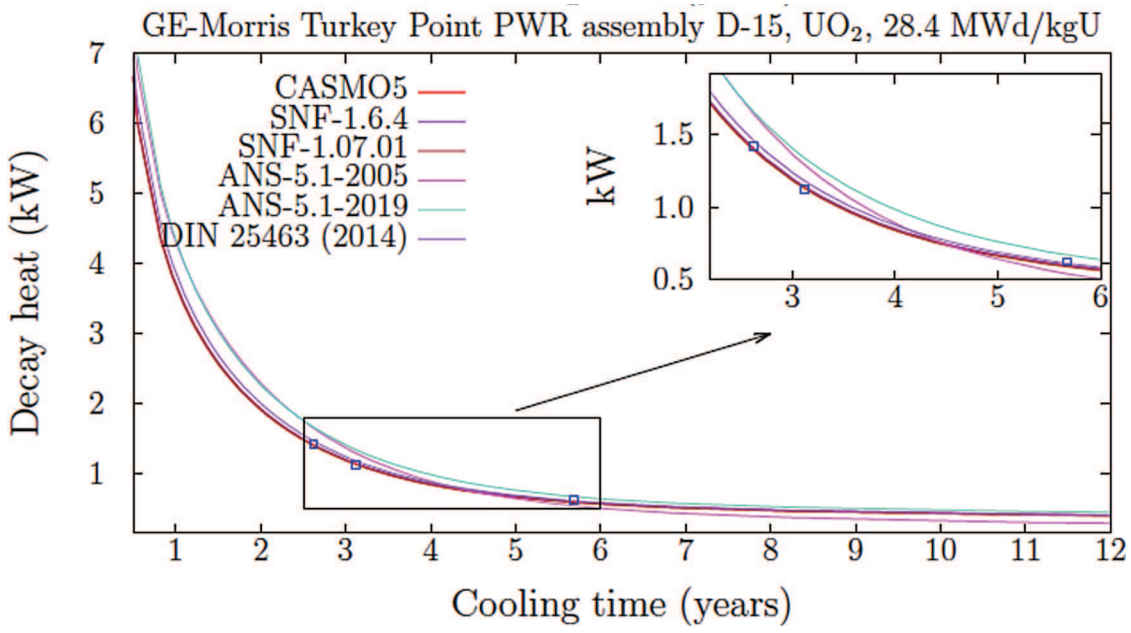

Fig. 1. Comparison of the measured and calculated decay heat for a specific PWR assembly from the GE-Morris facility.

work performed in this subtask is expected for the end of 2022.

1.1.2 SFC Subtask 2.2: develop, improve and demonstrate NDA methods/system for SNF characterization

The aim is to produce NDA systems that can be used to provide experimental data to validate the performance of the codes (including nuclear data) and to verify the declared design and operational history of the assemblies. Within this task, the performance of current NDA methods used to characterize SNF is assessed and improved. In addition, innovative neutron and gamma-ray detectors and NDA systems are developed. Two types of measurement systems are investigated: systems to characterize small SNF samples (including pellets) and systems to characterize entire SNF assemblies. The focus is on NDA methods based on the detection of gamma-rays and neutrons and calorimetric systems determining the decay heat rate.

A procedure to measure the neutron emission rate of a spent nuclear fuel segment sample by an NDA method without any calibration using a representative sample was developed, validated, and demonstrated. A standard neutron well-counter designed for routine nuclear safeguards applications was applied. The method relies on a transfer procedure adapted to the hot-cell facilities at the Laboratory for High and Medium level Activity of the SCK CEN. The sample was a segment taken from an SNF rod that was irradiated in the Tihange 1 PWR reactor to a burnup of 50 GWd/t. The composition and design specifications of this fuel rod and the irradiation conditions are fully documented. The method and procedures are described in detail in [16]. The results of these measurements will be used in subtask 2.4 to validate codes.

A large effort is being made to perform a full uncertainty evaluation of the NDA systems that were used at the CLAB facility as part of the SKB-50 measurement campaign. This includes the gamma-ray spectroscopic scanning system and calorimeter, which are installed at CLAB for routine use, and the Differential Die Away Self-interrogation system (5DDSI) that is developed at the Los Alamos National Laboratory. For the gamma-ray measurements, a study of systematic effects due to the determination of the net peak area and the positioning of the assembly with respect to the detector was carried out and reported in reference [17]. In addition, a model was developed to determine the ratio of the inventory of some key nuclides absolutely without the need for additional calibration measurements or transport calculations. A paper describing the use of this method on the SKB-50 data is in preparation.

1.1.3 SFC Subtask 2.3: determine the inventory of activation and fission products in cladding material

Although the cladding seems free of fissionable nuclides, impurities are introduced into the cladding during the fabrication. Moreover, by inserting the pellets within the cladding, traces of fuel are adherent on the inner surface of the cladding. The existence of uranium on the inner surface means the generation of fission products and actinides on and within the cladding during reactor operation, which has a direct impact on the long-term disposal characteristic of the cladding. In view of the current situation, where fuel assemblies are about to be stored for much longer periods than foreseen, the feasible, but not necessary, failure of the cladding within the dry storage cask could become a large obstacle in handling the fuel rods later on for the final disposal. Evidently, the ability to quantify the radioactive nuclides, particularly those that

cause radioactive defects in the cladding via high-energy alpha particles, is of high importance. Furthermore, the exact material composition of the cladding itself is a key issue for the sustainability of the irradiated mechanical and thermal loaded cladding. One of the goals of this subtask is to combine the confirmed simulations with the measured activated materials and by a reverse process to identify the original "real" material composition, which will allow estimating the failure time of the cladding (if at all) in the future, evidently determining the technical allowed temporary storage period.

In view of the above, the objective is to analyze the inventory of fission and activation products present in irradiated Zircaloy. The inventory of these products comprises the fraction that is formed during irradiation within Zircaloy and the fraction that is deposited on the inner surface of the cladding. The importance of this analysis lies in the assessment of the integrity of the cladding, in particular within the dry storage period, which could reach, as mentioned above, 100 years or more. Dedicated simulations are ongoing to compare and validate codes and their burnup procedures. The next step after code-to-code validation is the comparison with experimental results to depict the manufacturing effects (defects) on the cladding material composition as explained above. In that sense, primary calculations have shown that for the same Monte Carlo codes, the inventory of the main nuclides is very similar, which is, for an amount of roughly seven orders of magnitude lesser than the values within the fuel pellets, very encouraging.

For the experimental studies, two kinds of samples were prepared:

- cladded fuel pellets of UO_2 (50 GWd/tHM) irradiated in a pressurized water reactor (PWR).
- A plenum cladding obtained from a UO_2 fuel rod segment irradiated in a PWR.

Subsamples of these highly active claddings were analyzed by means of different (radio-) chemical analytical methods e.g., gamma-spectrometry, alpha spectrometry, LSC, and ICP-mass spectrometry.

The comparison between the experimental data and the simulation results will allow for the estimation of the adherent fuel on the inner surface of the cladding and, as said above, a reliable determination of the cladding sustainability under changing radiation conditions, as well as heat and pressure changes within the storage containers.

1.1.4 SFC Subtask 2.4: define and verify procedures to determine the source terms of SNF with realistic uncertainty limits

Within this task, validated state-of-the-art procedures will be defined to determine the SNF source terms with realistic uncertainty limits. This task combines the results of the other tasks. The final procedures will be validated based on the SKB-50 data. The main objective is to obtain statistically relevant statements (about the bias and uncertainty of source term calculations) by applying the standard, recommended calculation procedures to a large set of data. Calculations will be performed both with an established code like SCALE6 and a more sophisticated code like EVOLCODE2.

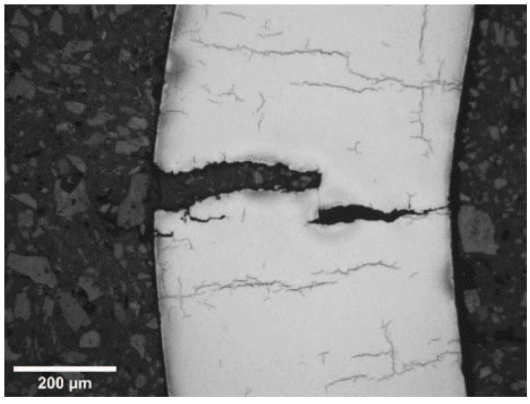

Fig. 2. Brittle failure of a pre-hydrided unirradiated ZIRLO(R) cladding sample with radial hydrides subjected to a ring compression test.

Source term calculations are influenced by many factors whose uncertainties are often insufficiently comprehended: initial material and geometry conditions, irradiation power history, spectrum influences from neighboring fuel assemblies, control rod history, and measurement procedures, for example. By evaluating many samples, the influence of these factors can be minimized. On the other hand, uncertainties from microscopic nuclear data are the same for all samples, and they manifest themselves as a bias in all calculations. Hence the determination of the latter allows conclusions about the improvement potential of cross sections and fission yields.

Finally, comparing results from different codes will allow assessing the impact of different methodological approaches for source term determination.

1.2 SFC Task 3: behaviour of nuclear fuel and cladding after discharge

The main aim of SFC Task 3 is progress regarding spent nuclear fuel (SNF) characterization and performance with respect to pre-disposal activities. In particular, the behavior of irradiated cladding, the phenomena ruling the potential SNF degradation, the fuel/cladding chemical interaction (FCCI), and aging effects under conditions of extended interim storage, transport, and emplacement in a final disposal system are investigated through experimental as well as modeling studies.

The understanding of spent fuel performance during dry interim storage is indispensable both for analyzing the failure probability and to characterize the state of the cladding, so that fuel management can be conducted with accurate knowledge of the fuel conditions. The main interest is in cladding degradation mechanisms, as it is the first physical barrier between fission products and the environment (Fig. 2). A description of the key phenomena governing the fuel rod state during dry storage can be found in the gap analysis carried out by Hanson et al. [18].

1.2.1 SFC Subtask 3.1: thermo-mechanical-chemical properties of the SNF rods and cladding

Subtask 3.1 investigates the thermo-mechanical-chemical properties of the spent nuclear fuel rods and cladding. In this subtask, experimental and simulation activities are taking place. Some highlights are given below.

At JRC-Karlsruhe, a ring compression test (RCT) was performed in an irradiated duplex cladding sample (from the upper plenum, average burnup 67 GWd/tHM) [19]. This test was modeled by BAM using finite element analysis and an inverse method with an iterative approach.

At the Hungarian Centre for Energy Research, creep tests on as-received and hydrogenated E110 cladding samples were performed. Samples with 1100 ppm hydrogen content showed a significantly lower thermo-mechanical creep rate than the as-received samples.

At UPM, a newly developed device was used to perform a hydride reorientation treatment in pre-hydrided unirradiated ZIRLO(R) cladding samples. RCTs were performed in samples with a significant fraction of radial hydrides at different temperatures. Failure was brittle at room temperature and 135 °C, with cracks initiating at radial hydrides (Fig. 2).

NAGRA (National Cooperative for the Disposal of Radioactive Waste in Switzerland) validated its three-dimensional static finite element analysis of 3-point bending tests at JRC-Karlsruhe, a sensitivity analysis on different beam implementations in finite element modeling, and a derivation of appropriate rod failure criteria.

At CIEMAT, the code FRAPCON-xt has been used to validate the initial conditions for fuel rod behavior assessment at dry storage, applied to a specific PWR fuel rods dataset with burnups ranging from 45 to 65 GWd/tHM. The results show that practically 90% of deviations might be explained by uncertainties in the estimate, as shown in Figure 3.

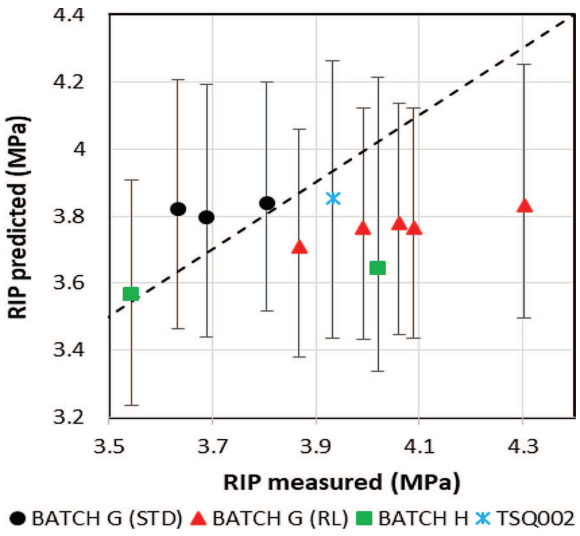

Fig. 3. Best estimate plus uncertainties of rod internal pressure (RIP) at EOL for different rod designs (distinguished by symbols and colors).

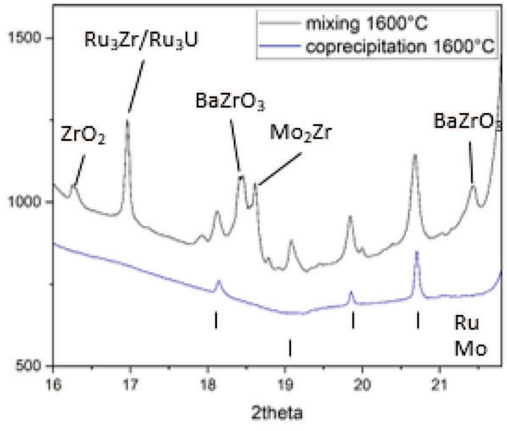

Fig. 4. Synchrotron XRD of SIMFUEL manufacture by solid (mixing) and liquid (co-precipitation) routes.

1.2.2 SFC Subtask 3.2: behavior of SNF pellets under interim storage conditions

In a strategy of long-term interim storage, it is required to evaluate the SNF evolution over long periods and to quantify the consequences of non-standard conditions such as aging or defective cladding. An accurate understanding of SNF performance during dry interim storage is indispensable for ensuring the safety of all relevant operations. If the fuel pellet is exposed to an oxidizing environment during dry storage due to fuel management operation issues and an undetected defect in the cladding, UO_2 oxidation can occur. The main concern is that if oxidation develops up to the formation of U_3O_8, it results in a volume expansion, which may involve a fission gas release and fuel grain de-cohesion.

The objective of subtask 3.2 focuses on the performance of irradiated fuels and simulated SNF pellets both under (ab-)normal dry interim storage conditions and under conditions of water contact, i.e., container or cladding failure scenarios, respectively, and extended storage prior to their transport, during the transport and emplacement in a disposal system. The main goal is to provide new insights into the aging and degradation mechanisms of the SNF (e.g., He build-up, oxidation of the SNF, influence of the various fission products, etc.). The potential evolution of the SNF in case of mal operating conditions (e.g., moisture, aqueous dissolution of the UO_2 matrix, corrosion of the SNF, etc.) is also considered. Various compositions of simulated SNF are used as lanthanide Ln (La, Nd, Ce, Gd, Eu)-doped UO_2, and SIMFUEL pellets doped with metallic particles (Ln–PGM–Mo–UO_2 ceramics) to be tested under conditions relevant in the back-end of the fuel cycle (dry and wet). Morphological and chemical/spectroscopic analyses of the fuel behavior are performed by means of conventional techniques (i.e., ICP-MS, SEM-EDX, ESEM, TEM, XRD, Raman spectroscopy). Cutting-edge synchrotron methods at the Rossendorf beamline (i.e., high-resolution diffraction, x-ray absorption spectroscopy, and x-ray emission spectroscopies) are also applied. The results provide degradation performance and mechanisms at a multi-scale level ranging from the atomic scale to the macroscopic scale (Fig. 4).

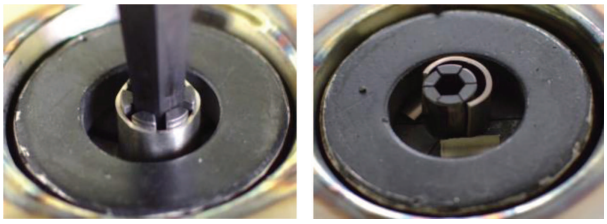

Fig. 5. The mandrel test performed with a ductile (left) and a brittle sample (right).

1.2.3 Subtask 3.3: pellet-cladding interaction under conditions of extended storage, transport, and handling of SNF rods

During normal operation, the gap between the fuel pellet and the cladding closes. In case of sudden changes in operating conditions or in accidents, the thermal expansion of the pellet can rupture the cladding wall. Air ingress during high-temperature dry storage can oxidize the fuel pellet resulting in volumetric expansion; this also can result in cladding rupture. Hence, the ductility of the cladding is an important parameter under transport and storage conditions.

In subtask 3.3, the mandrel test simulates the pellet-cladding mechanical interaction (PCMI) by expanding segmented dies (mandrels) inside cladding samples (Fig. 5). This setup represents the mechanical conditions of the cladding under internal pressure. The result is the maximum force, the force-displacement curve integral, the maximum diameter, and the mode of failure, which all give important information about the ductility of the sample.

The tests were performed with samples from E110 and sponge-based E110M cladding tubes. Some rings were pretreated with hydrogen and heat treated at high temperatures. The hydrogen content was between 300 ppm and 3000 ppm. The tests were performed at 20 °C and 300 °C in air. The cladding samples behaved gradually less ductile as the hydrogen content in the samples increased. The minimum ductility was reached at 2000 ppm absorbed hydrogen. Above 2000 ppm hydrogen content, the ring samples failed in brittle mode after minimal plastic deformation at higher force compared to the as-received samples.

1.3 SFC Task 4: accident scenario and consequence analysis

Task 4, coordinated by CIEMAT, was set up to study SNF performance under potential accident conditions, which might lead to a loss of confinement during storage, transport, and predisposal activities. The investigation to be carried out was split into two subtasks. The first one (4.1), coordinated by NAGRA, is specifically focused on thermo-mechanical aspects of fuel behavior under dry interim storage. The second one (4.2), coordinated by the Chernobyl Research and Development Institute (ChRDI), pays attention to the consequences of potential accidents. In the next paragraphs, a synthesis of the progress made in each of them since the inception of EURAD is to be described.

As for Task 4 as a whole, coordination has managed three major achievements:

- communication with Task 3. Any accident analysis requires an accurate description of the SNF at the time of the accident. This, in turn, needs a thorough characterization of SNF properties evolution during irradiation, which is the main purpose of Task 3. After exploring different ways of communication with Task 3, the one that has resulted in more efficient under the circumstances has been the bridge built by the participation of some partners in both Tasks 3 and 4. This is now well settled through CIEMAT and NAGRA.
- Consolidation of the program of work. The research activities initially identified were diverse and have been consistently distributed so that each subtask addressed a specific aspect of interest in fuel performance, as described above. Deliverables to be released have been better accommodated as for their scope and, no less important, due dates. Additionally, several technical reports will be issued concerning accident scenarios and methodologies, Source Term in Vault systems, and criticality studies in accidental configuration.

1.3.1 SFC Subtask 4.1: fuel thermo-mechanics

Subtask 4.1 aims to provide a synthetic analysis of identified accident scenarios in different stages of the back-end spent nuclear fuel cycle (i.e., storage, transportation, and handling). The core topics include the identification and analysis of potential accident scenarios in interim storage and/or packaging facilities and, consequently, the assessment of the fuel performance. Therefore, the work related to this subtask is closely linked to the experimental and numerical activities performed in the other tasks with regard to the fuel properties characterizations and related uncertainty analysis, as well as to the behavior of nuclear fuel and cladding after discharge. The development of a methodology trying to link degradation mechanisms to fuel performance and possible accident scenarios would be beneficial to support future specific National Programs in relation to the safe management of SNF. The work in this subtask is a synthesis of individual studies performed by five contributing participants and will be summarized in deliverable 8.11.

In particular, the development of a methodology for the evaluation of the SNF performance under accident scenarios is being done by Nagra (National Cooperative for the Disposal of Radioactive Waste in Switzerland) and CIEMAT (Center for Energy, Environmental and Technological Research in Spain). For this purpose, two different approaches are being followed: Nagra is working on a methodology based on numerical investigations with Finite Element Analysis; and CIEMAT is relying on an engineering methodology capable of assessing the fuel performance with considerably less computational cost and by utilizing fuel performance code (i.e., FRAPCON-xt). These developments, though, are not fully independent from each other, as CIEMAT plans to use some of the findings from NAGRA's research.

The identification and analysis of possible accident scenarios will be performed by IDOM (Spain) and TUS

(Technical University in Sofia). IDOM already summarized a list of all accident scenarios, including potential initiating event and their associated risks (probability and consequences). In addition, they are reviewing multiple studies to identify the types and magnitudes of the different loads that SNF assemblies might undergo in different scenarios. By doing so, they foresee identifying the main fuel parameters and related degradation mechanisms involved in every accident scenario. In the future, TUS plans to support IDOM by studying and reviewing information sources related to the possible phenomena and the consequences that could appear during SNF long-term storage in case of an accident.

Finally, CEA (French Alternative Energies and Atomic Energy Commission) has already started the detailed mechanical characterization of SNF that underwent fuel matrix oxidation due to defective rods in dry storage and off-normal conditions, i.e., air ingress inside a defective rod. The work includes annealing spent fuel rodlets (with Zr4 cladding) until fuel crack failure, followed by a series of experimental investigations (3D FIB/SEM).

1.3.2 SFC Subtask 4.2: accident management and consequences

The general objective of subtask 4.2 is to study SNF behavior under accident conditions with the intention to assess the current capability of consequence analysis and to support national programs for safe management during back-end activities. The main focus of the subtask is the writing of Deliverable 8.13, "Analysis of the conditions of the state-of-emergency radioactive wastes packages containing SNF, FCM (Fuel-Containing Materials) or HLW/LLW generated due to ChNPP accident", as it is considered that the experience gained in the aftermath of the accident might be instrumental in building accident management guides elsewhere. Presently, the report is being drafted, and a final version will be released soon to partners other than ChRDI. Additionally, two studies are planned to explore the analytical capabilities of the Source Term in a vault system undergoing a postulated leak from an SNF and the criticality values that might be reached in accident arrangements within a storage cask. The former has already been started; the latter sets a bond between Tasks 2 and 4, in this case through CIEMAT participation in both tasks.

2 DisCo

The disposal of new and unconventional fuel types was identified as a high-priority topic in the Strategic Research Agenda of the IGD–TP (Implementing Geological Disposal–Technology Platform). The DisCo project was initiated to test the hypothesis that modern fuel types, such as doped fuel, will dissolve in a way that does not significantly differ from standard conventional nuclear fuels. Specifically, the DisCo project focused on Cr-, Cr+Al, and Gd-fuels. The project aimed to provide data from leaching experiments of spent nuclear fuel, model materials (analogs), and models are describing the chemical system expected in the failed waste container. Two main objectives motivated the project: (1) to improve understanding of spent fuel matrix dissolution under relevant repository conditions and (2) to assess the dissolution behavior of modern types of fuel (MOX, doped) compared to standard fuel.

The DisCo project was based on three main parts: experiments with real spent nuclear fuel, experiments with model systems, and chemical modeling. To allow comparisons between the parts and to ensure the modelers received the required data in the right format, a specific work package was dedicated to synchronizing sample synthesis, characterization, and experimental systems. The idea was to provide a well-defined initial state of the system before the dissolution experiments. Synthesized model UO_2 included additions with Cr, Al, Gd, Th, and Pu, either as sole dopants or in a combination of two dopants. Studied spent fuel samples included UOX fuel doped with Cr and, Cr+Al, and spent MOX fuel, as well as standard UOX fuels. In addition, failed fuel recovered from fuel ponds was characterized, and data was used as input to the modelers. Most of the spent fuel experiments were performed using simplified, synthetic granitic groundwater with bicarbonate, while some used synthetic cement water with Ca – this water has a high pH and more complex chemistry. In the model materials experiments, four different water types were used: simple bicarbonate water and young cement water to mimic the experiments performed with real spent fuel; simulated Callovo-Oxfordian groundwater relevant for clay-based geology, and real granitic groundwater from the Olkiluoto site in Finland. Most spent fuel experiments were performed in anoxic and/or reducing environments (with hydrogen in the gas phase), and this was also the aim for most model systems experiments. In some model systems experiments, however, the oxidative dissolution of doped UO_2 was investigated by using hydrogen peroxide as a simulant for the expected radiolytically produced oxidants. Samples were taken from the systems at various intervals, and the fluids were analyzed with mass spectrometry to provide dissolution data. When available, these data were used in the modeling activities, including models developed using different tools and approaches. The presence of near-field materials, such as iron components, was taken into account in the models.

2.1 DisCo results

The main outcome from the different work packages providing results from SNF, model systems, and modeling are provided below.

2.1.1 Experiments using spent nuclear fuel

Samples from the following spent nuclear fuels were prepared and characterized before starting the dissolution experiments: standard UOX with average burnup of 50–59 MWd/kgU, doped fuel (Cr, Cr+Al) with average burnup of 57–58 MWd/kgU, and MOX fuel with average burnup of 38–56 MWd/kgHM. By also using different chemical conditions, the data produced was used to assess the effect of various parameters:

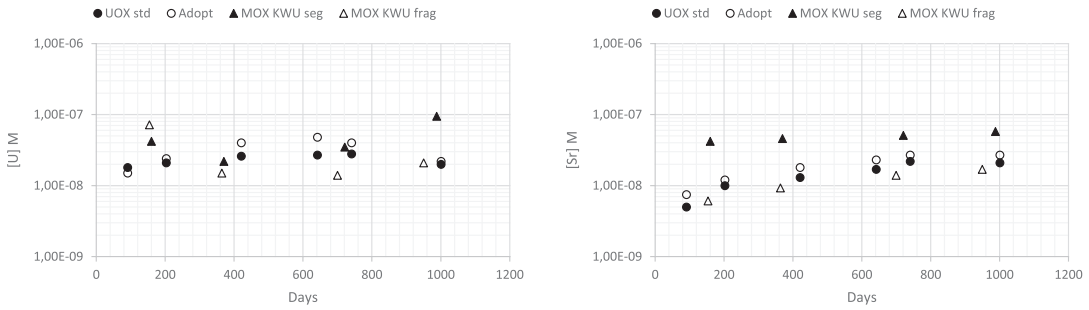

Fig. 6. Concentration in aqueous solution vs. time for standard fuel, doped fuel (Adopt), and MOX. Experiments with hydrogen overpressure (Studsvik: initially 50 bar hydrogen. KIT: initially 40 bar with 3,2 bar hydrogen). NB Only data after 90 days are shown on this plot. Data produced by Studsvik and KIT in the DisCo project.

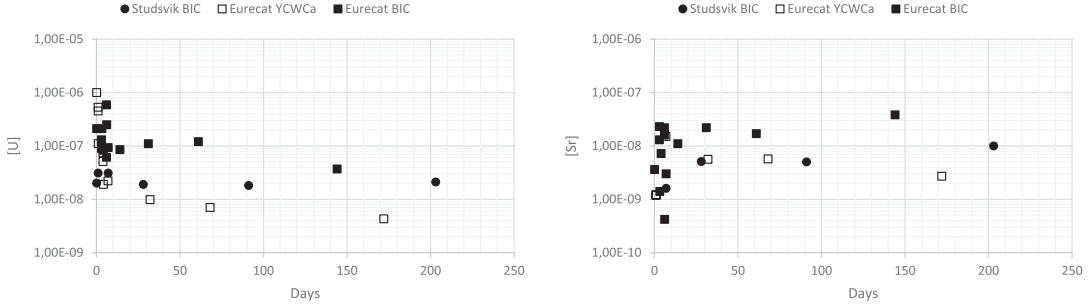

Fig. 7. Concentration in aqueous solution vs. time for standard fuel leaching in bicarbonate (BIC) water and Young cement water with Ca (YCWCa), both with hydrogen overpressure (Studsvik: initially 50 bar hydrogen. Eurecat: initially 35 bar with 1,7 bar hydrogen). NB for the Studsvik dataset, only data up to ∼200 days are shown. Data produced by Studsvik and Eurecat in the DisCo project.

(1) fuel type;
(2) hydrogen overpressure;
(3) aqueous solution;
(4) sample preparation.

The release of actinides (uranium plutonium americium) and other radionuclides commonly show some scatter in all experiments during the first year of dissolution [20–22]. This makes it hard to draw clear conclusions from those data with regard to matrix dissolution and the effects of different parameters. To assess the effects on matrix dissolution, data from experiments that were run longer than one year are presented in Figure 6 ([22]; data from Studsvik and KIT)). In experiments using bicarbonate water and hydrogen overpressure, the U and Sr release is quite similar. Release data from other radionuclides (not shown) are also similar, indicating that neither doped fuel nor MOX exhibit any significantly different dissolution behavior compared with standard UOX. For the three fuel types studied here, the hydrogen clearly inhibits the oxidative dissolution and the release rate approach but have not yet reached, zero. Experiments without hydrogen display much higher uranium concentrations due to the oxidation of uranium in the spent fuel matrix. To assess effects of chemical conditions, datasets using the same fuel type, but different aqueous solutions are compared in Figure 7 ([22]; data from Studsvik and Eurecat). When writing[1], data from experiments using young cement water (high pH) under hydrogen are provided for up to ca 200 days. Therefore, interpretations with respect to the effect on matrix dissolution should be made cautiously. One may observe, however, that using the young cement water appears to lower the concentrations of some radionuclides (Pu, Am).

2.1.2 Experiments using model systems

In the DisCo project, experiments were performed using the following types of model materials: pure UO_2 and UO_2 with one additive (Cr, Gd, Th, or Pu) and two additives (Cr+Al and Cr+Pu). Adding Pu as ^{238}Pu adds alpha radiation to the system, and the amount is adjusted to mimic spent fuel with an age of ca 10 000 years. The effect of alpha doping was also investigated using UO_2 doped with ^{233}U. These model materials were exposed to both oxidizing and reducing conditions and different aqueous solutions with compositions mimicking those used for spent fuel experiments (see Sect. 2.1.1). In addition, real Finnish (granitic) ground waters were used in a few experiments.

The work performed during the preparation of model materials provided essential information and knowledge concerning the uranium dioxide solid-state characteristics. The preparation method chosen affects the resulting product: the microstructure, as well as the amount of Cr that is soluble in the UO_2 matrix, is controlled both by the oxygen potential and the amount of Cr available during sintering [23,24]. It was shown that the solubility limit of Cr is between ca 700 and 1000 ppm. Above the solubility

[1] January 2022. More data will be available later in the year.

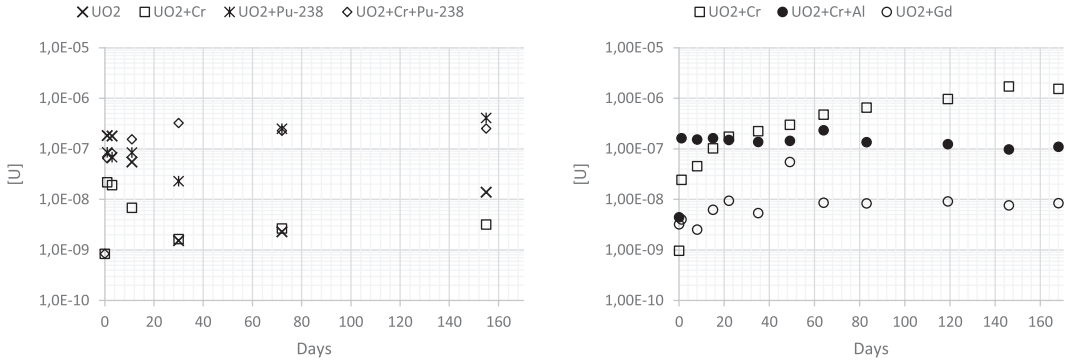

Fig. 8. Uranium concentration in aqueous solution vs. time for undoped and doped UO_2 in experiments with hydrogen in the gas phase. Left: data from SCK CEN (10 bar hydrogen). Right: data from Ciemat (initially 8 bar with 5% hydrogen; NB, some air contamination noted).

limit, Cr, which acts like a sintering agent, will precipitate long grain boundaries. Detailed studies of the defect concentration and Cr oxidation state revealed how the potential formation of oxygen vacancies influences the Cr incorporation into the uranium dioxide lattice [25,26].

Unirradiated Pu-rich MOX was already available at the start of the project, however, two sets of Th-MOX were produced: heterogeneous and homogeneous samples were needed to compare with previously available data from unirradiated Pu-MOX as well as data produced in this project [27].

Dissolution experiments in oxidizing conditions show that Cr-doping may affect the dissolution rate – however, in these systems, it appears to be more related to the microstructural (e.g., grain size, porosity) effects of the doping rather than the chemical effect of Cr. The differences observed are mainly related to the initial stages of dissolution, and in the long run, the differences are smaller, and the Cr-doping appears insignificant. Another important observation is that when the sample is repeatedly exposed to hydrogen peroxide, the dissolution rate of uranium is reduced. It appears a second phase is produced that affects the surface reactivity of the uranium dioxide, even if the aqueous solution contains bicarbonate [28].

Model materials dissolution experiments with hydrogen present aimed to investigate U dissolution in strongly reducing conditions. However, in some of these experiments, there may have been traces of oxygen (from the air). Oxidants were also available in systems with alpha-doped material [29]. In most cases, data indicates no effect of Cr-doping on the U dissolution rate. Only in a few cases an effect of doping with Cr are seen (Fig. 8). It seems Cr may have some anti-oxidation effect, but it is small and only apparent when the conditions are weakly oxidizing. The Gd-doped material produces less oxidized U, however, the combination of Cr and Al may also be inhibiting U oxidation.

Unirradiated MOX systems were investigated using simulated Callovo-Oxfordian water. For these systems, it appears the added Pu and Th have a protective effect regarding uranium oxidation. However, the most obvious is the effect of iron on the systems. Even in a system with relatively high alpha-radiolysis, U is kept at the level of UO_2 (am) solubility, and Fe precipitates are found both on the Pu-MOX-pellet as well as on the corroding iron. This corroborates the results from an alpha-doped sample in natural groundwater and iron – the U concentrations in these systems are very low. Overall, in the experiments performed in the DisCo project, iron appears to have a more pronounced effect on U concentrations than hydrogen for both doped and undoped systems.

2.1.3 Modeling the dissolution of spent fuel in repository conditions

The modeling work considered experimental data produced before the project and data generated along Disco. To this aim, an experimental Excel database was generated, including all experimental details needed by modelers to incorporate into the numerical models. The concept of this database constituted an important outcome of the project.

The main question relating to the potential effect of dopants is how the oxygen potential of the irradiated fuel changes due to the elements added to the uranium dioxide lattice. Thermodynamic modeling using the GEM-Selektor code was employed to explore this. The development of the model went through different stages to account for the solubility of Cr in the lattice and the effect of irradiation, implying modeling a complex system including fission products and Cr. The final results show that above a burnup of ca 20 MWd/kgU, Cr-doping has a negligible effect on the oxygen potentials [30]. The model results also confirmed that zirconium alloy oxidation at the pellet-cladding interface might locally reduce the oxygen potential.

Important improvements were incorporated into the Matrix-Alteration Model (MAM) during the DisCo project. The MAM model has now been implemented with the iCP (Comsol-PhreeqC coupling interface) tool, including modeling the effects of metallic aggregates and hydrogen. The model is able to reproduce data from the experimental systems studied in the DisCo project [31]. For example, the possible effect of combining Cr and Al in the spent fuel has been modeled. The experimental results indicate that the Cr+Al combination may have a more

pronounced catalytic effect with regards to the hydrogen effect than if only Cr is reproduced by the model.

The MOX experimental system, corresponding to the French repository system with the Callovo-Oxfordian groundwater, was modeled using the CHESS-HYTEC approach, using geochemistry and reactive transport. The results clearly show that iron in the system is very efficient in reducing oxidants, and the U and Pu concentrations are very low [32]. Thus, the corroding iron in this groundwater system appears to be enough to keep the actinides in their reduced form, keeping the dissolution rate low.

2.1.4 Failed fuel in ponds

As an additional study, the DisCo project also involved characterization and modeling of fuel exposed to pond water for ca 40 years. The data from the characterization of this fuel (gamma spectrometry, visual inspection, and optical microscopy) were used in the efforts to model the fuel behavior using electrochemical modeling. The so-called Mixed-Potential-Model, the electrochemical model, previously used and developed in Canada [33], was further developed and improved in the DisCo project. However, the results were not encouraging due to a large number of unknown parameters, which forces estimations and thus increases the uncertainty level. Therefore, the electrochemical approach is suggested to be abandoned in [34].

2.2 Discussion of DisCo results

The combined results presented in section 3 show that even though grain size and microstructure are affected by synthesis procedures and doping with Cr (or Cr+Al), the effect on irradiated fuel is insignificant with regard to oxygen potential and radionuclide release patterns. In some cases, there is an effect of the doping – especially Gd seems to be able to reduce the redox reactivity of the UO_2 matrix. This corroborates with previous results from [35].

The spent nuclear fuel and model systems data used in modeling has improved the understanding of the systems; results indicate that a three-valent additive may protect the uranium dioxide from oxidation. Even though only Cr may not be so efficient in this respect, combining Cr with Al makes the reactivity towards oxidants in the presence of hydrogen seem to be diminished. This supports the hypothesis that dopants do not affect the dissolution of spent fuel negatively – it rather seems to have some protective effects. The weaker effect of Cr compared with Gd may perhaps be related to the possibility that the valence of Cr is not always (III).

The chemical environment in which UO_2 dissolution takes place has the most pronounced effect. Clearly, if the oxidation of U can be avoided, the dissolution rate is slow and is controlled by the release of U(IV). The results show that hydrogen does inhibit the U oxidative dissolution, but in these experiments, the spent fuel matrix seems to continue to be very slowly dissolved even after ca two years. In at least one case (Studsvik), some answer to this may lie in a strong pre-oxidation of the sample. There are other recently published data where hydrogen overpressure has completely stopped oxidative dissolution [36,37].

Finally, the results from model systems show that the presence of corroding iron in groundwater (both natural granite and simulated clay water) can efficiently remove oxidants and minimize the rate of oxidative dissolution. The systems have very low Eh and U concentrations equal to (or lower) than that expected from equilibrium with UO_2 (am).

Even though much has been learned from the new results produced in the DisCo project, some questions remain. There is a need to further understand the hydrogen effect – e.g., reactions involving hydroxyl radicals at the fuel surface and potential countereffects from groundwater constituents – and the relative effects of iron and hydrogen in a repository setting. The impressive effect of Fe on the MOX system means we need to further explore the effects of various expected Fe concentrations in the different repository settings. With regards to the effects of dopants, there is some remaining uncertainty regarding the valence of Cr in the uranium dioxide matrix that still requires some attention. In general, the influence of dopants with a predominant valence other than +3, for example, Th, is not satisfactorily understood and requires further study.

2.3 Conclusions from the DisCo project

The results from the DisCo project show that the matrix dissolution behavior of MOX, doped fuels (with Cr and Al as dopants), and standard fuel types are similar. This indicates that the modern fuel types studied in this project can be included in the plans and safety assessments for spent nuclear fuel repositories without any detrimental effects. Project results have confirmed that a reducing environment and the presence of corroding iron in the repository can keep the dissolution rate of spent nuclear fuel low. The mineral chemistry and microstructure of the uranium dioxide matrix are better understood due to efforts in producing the model systems, and a tangible outcome is the actual samples available for further studies. The modeling of spent fuel dissolution and radionuclide release and transport performed in the DisCo project has also improved understanding of the repository systems, from the thermodynamics of the spent fuel to the effect of a redox front on the radionuclide transport in the repository. The models developed are ready to be included in the safety assessments. The DisCo project has thereby contributed importantly to the safety case for spent fuel repositories in a range of different settings. Uncertainties in critical parameters are reduced and important for the performance assessments, but the outcomes are also useful for developing waste acceptance criteria for spent fuel repositories, an important issue closely connected to the licensing process.

Conflict of interests

The authors declare that they have no competing interests to report.

Acknowledgements

The SFC project has received funding from the European Union's Horizon 2020 research and innovation program under grant agreement No 847593. The DisCo project has received funding from the Euratom research and training program 2014–2018 under grant agreement No 755443. The DisCo project has also benefited from financial support from project partner organizations as well as Waste Management Organizations involved in the project. We are grateful to all DisCo project participants who have performed the experiments and modeling.

Funding

The SFC-project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 847593. The DisCo project has received funding from the Euratom research and training programme 2014–2018 under grant agreement No 755443.

Data availability statement

Data from experiments performed in DisCo are found in project deliverables available via https://cordis.europa.eu/project/id/755443/results.

Author contribution statement

All the authors were involved in the preparation of the manuscript. All the authors have read and approved the final manuscript.

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Cite this article as: Anders Sjöland, Petra Christensen, Lena Zetterström Evins, Dirk Bosbach, Lara Duro, Ian Farnan, Volker Metz, Uwe Zencker, Jesus Ruiz-Hervias, Nieves Rodríguez-villagra, Márton Király, Peter Schillebeeckx, Dmitri Rochman, Marcus Seidl, Ron Dagan, Marc Verwerft, Luis Enrique Herranz Puebla, Dmitri Hordynskyi, Francisco Feria, and Efstathios Vlassopoulos. Spent nuclear fuel management, characterisation, and dissolution behaviour: progress and achievement from SFC and DisCo, EPJ Nuclear Sci. Technol. **9**, 13 (2023)