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Research activities on radioactive waste management and on the back-end of the nuclear fuel cycle performed by the Joint Research Centre of the European Commission

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Summary. — The Euratom Research and Training Programme contributes, within its portfolio of activities, to establish and improve the scientific basis of knowledge for the safe management of spent nuclear fuel and radioactive waste. This includes research and innovation activities undertaken by the Joint Research Centre (or JRC, the European Commission's science and knowledge service) in its laboratories. This paper provides an overview and some highlights of the Joint Research Centre (JRC) activities which are dedicated to the safety of spent fuel and high level radioactive waste forms. The fields of experimental and modelling research address various stages of spent fuel management after discharge from the reactor core: cooling in the spent fuel pool; handling, transport, extended interim storage and retrieval thereafter; disposal in a deep geological repository and long term behaviour of the spent fuel/waste form after disposal. The safety of the "back-end" of nuclear fuel cycles which include U-Pu recycling and/or a "fully closed" cycle with minor actinides separation and transmutation is also a major area of research. Both normal operation and accident scenarios, which cause fuel degradation/melting, are investigated. Possible applications for legacy waste management, decommissioning, and safeguards are considered. The relevance of the research is linked to the possibility of investigating "real" spent fuel and highly radioactive compounds using JRC's research infrastructure, which includes hot cells and shielded facilities, and state of the art experimental methods that are (in some cases) rare or even unique. The activities are performed in collaboration with partners and/or in the context of international initiatives. Opportunities and perspectives for enhanced cooperation, including access and sharing of infrastructure are being developed.

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1. – Introduction

Among the specific objectives of the Euratom Research and Training (R&T) programme 2021-2025 [1] to which Joint Research Centre (JRC) contributes, "*safe spent fuel and radioactive waste management and decommissioning*" occupies a prominent role. Within the field of responsible and safe management of spent fuel and radioactive waste, JRC is performing R&D together with the Member States and is facilitating international networking. JRC contributes also to maintain and further develop expertise and competence in the nuclear field and to support the policy of the EU and its Member States. JRC activities are embedded in the European Joint Programme on Radioactive Waste Management (EURAD1 and the upcoming EURAD2), which defines the needs for and promotes integrated research/cooperation among Member States, and are aligned with the strategic vision of Implementing Geological Disposal (IGDTP) and Sustainable Nuclear Energy Technology Platforms (SNETP). The aim of the scientific-technical investigations is to increase confidence in safety of waste management and disposal solutions. The experimental data and knowledge generated by the JRC feeds the modelling of the long-term processes and reduces uncertainties in the associated Safety Assessment. The focus is on *non-destructive assay, long-term storage and disposal of spent fuel*. The research activities encompass *conventional and innovative fuels* (*e.g.*, accident tolerant fuel - ATF) for existing and future reactor systems, and *degraded/molten fuel (corium)* from severe accident scenarios. Radioactive waste forms associated with fuel cycles including recycling of actinides and/or for which technical solutions are still not fully defined (*e.g.*, *legacy waste*) are also investigated.

The following sections provide schematic descriptions and highlights from the main active fields of JRC activity.

2. – Spent fuel aging studies

After storage in the cooling pool at reactor sites for initial heat decay of spent fuel elements, and if no reprocessing is foreseen, the spent fuel will be transported to wet or dry interim storage facilities and then stored for several decades or even centuries prior to retrieval and repackaging for disposal in the geological repository. The safety assessment of extended storage requires predicting the behaviour of the spent fuel assemblies and of the spent fuel itself over a correspondingly long timescale, to ensure that the mechanical integrity and the required level of functionality of all components of the containment system are retained. Accelerated ageing and basic studies supported by modelling are necessary to predict the long term behaviour of these materials. At JRC, experimental studies are performed addressing extreme conditions related to handling/transportation and also long-term ageing issues affecting spent fuel [2]. The potential consequences of accidents causing spent fuel rod failure involve fuel fragments release and dispersion. Impact tests using a hammer drop device, as pictured in fig. 1, and bending tests in hot cell are performed on spent fuel rod segments. Analysis of the particle size distribution of the fuel released from the rod breakage, and determination of dynamic loading parameters from the high speed camera video recording of the impact are performed [3].

The final goal of these investigations is to determine parameters and conditions governing the response of spent fuel rods (fuel and cladding with different configurations and histories) to impact loads and other thermo-mechanical solicitations corresponding to normal and off-normal conditions that may be experienced by the rod during handling, transportation, storage, and after extended storage. In addition to mechanical loading

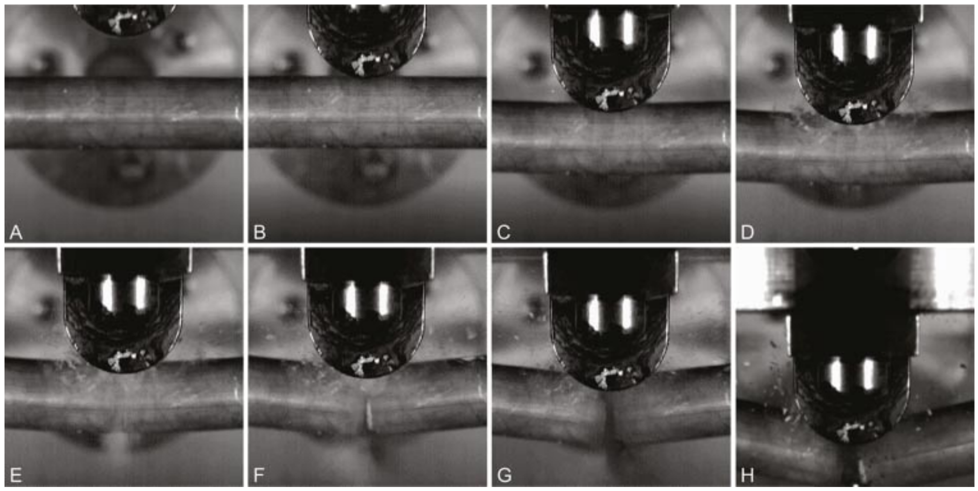


Fig. 1.: Sequence recorded during an impact test on a MOX spent fuel rod segment (BU = 61 GWd/t)

tests, property measurements are performed on spent fuel and analogues to determine the long term evolution and the potential effects of ageing processes on the mechanical integrity of the spent fuel rod. Spent fuel rod alterations as a function of time and cumulative decay damage, and alteration kinetics are monitored at microstructural level (defects and lattice parameter swelling [4] and at macroscopic property level (*e.g.*, hardness [5] and thermal conductivity [6]). The results obtained so far show that saturation of macroscopic hardening and thermal conductivity decrease are to be expected after decades or centuries of storage, depending on the burnup and composition of spent fuel. In order to reproduce cumulative decay damage effects expected after extended storage time within acceptable laboratory timescales, accelerated damage build-up conditions are applied by testing unirradiated (U, Pu) oxide with high specific alpha-activity [7]. Alpha-damage effects have also been studied in aged SUPERFACT fast reactor fuel from partitioning and transmutation safety studies, after having cumulated decay damage and radiogenic helium equivalent to extremely old conventional spent fuels. This irradiated fuel has high minor actinide content, hence high specific activity. The examination of the microstructure of such fuels has evidenced their very high radiation damage resistance [8]. Figure 2 shows typical fracture surface morphology characterized by the presence of large bubbles containing fission gases and radiogenic helium. Helium thermal desorption studies allowed determining a new correlation and diffusion coefficient that could be used for reactor and storage conditions [9].

3. – Modelling

The simulation of spent nuclear fuel under dry storage by means of the TRANSURANUS fuel rod performance code of the JRC started several years ago [10,11]. Specific models for cladding creep were tested and implemented [12]. More detailed models have been developed since then. This was implemented in the TRANSURANUS code, for example as part of the first mechanistic model for fission gas behaviour [13,14], but later

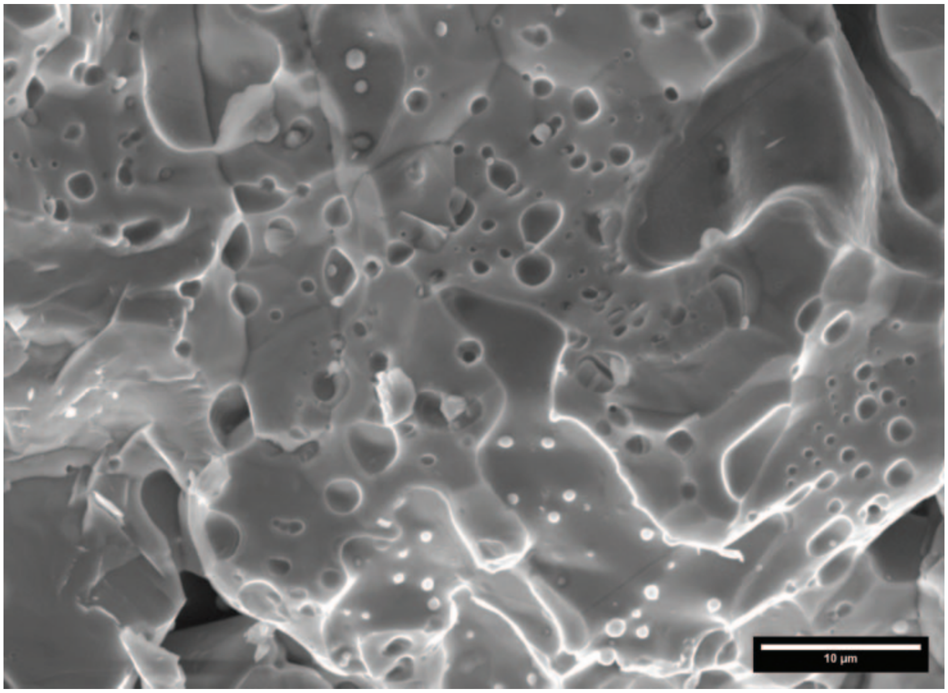


Fig. 2.: SEM micrograph of an aged irradiated fast reactor fuel sample with high specific activitiy (SUPERFACT) showing large fission gas/helium bubbles and metallic fission products precipitates (white dots).

also in standalone codes that can be coupled with TRANSURANUS. First there was the SCIANTIX code [15]. In a second step, a coupling was made with the MFPR-F code [16]. This code started from the most elementary level of point defects and enabled to deal with other fission products and their chemical interactions with oxygen and actinides in the MFPR code [17]. The coupling with both codes has now been demonstrated to simulate irradiated fuel rods during a loss of coolant accident, but also paves the way to account for the resulting accumulation of radiation damage and radiogenic He. In parallel to the experimental work, the MFPR-F code will be extended to include the capability to simulate the effects of α-self-irradiation in UO_2 at an atomic and mesoscopic scale. Following the implementation of the phenomena and variables in the code, a first comparison with available experimental data obtained at the JRC-Karlsruhe will be applied. In a second step, the code will applied for the analysis of the self-irradiation effects on the mechanical and thermal properties. In a third and last step, the MFPR-F code will be tested when coupled with the TRANSURANUS code for the simulation of a complete fuel rod. As far as the cladding behaviour of high burnup rods is concerned, it is important to take into consideration the hydrogen uptake and subsequent redistribution, precipitation of hydrides and their re-dissolution. For this purpose, the JRC has implemented and tested the new TRANSURANUS model for hydrogen uptake under stationary [18] and transient conditions. In the frame of a collaboration with CIEMAT, a coupling of the CIEMAT HYDCLAD model is underway [19,20]. The model will consider

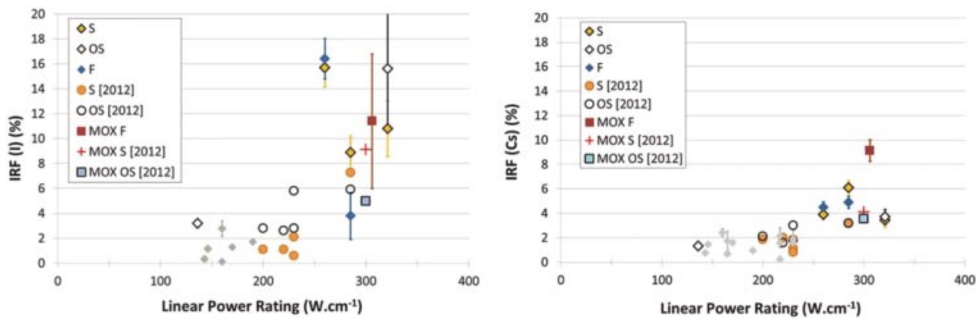


Fig. 3.: Correlation between Instant Release Fraction (IRF) and irradiation history. The linear power rating is the main factor affecting the IRF [24].

the nucleation and growth of hydrides as a function of the local hydrogen concentration and temperature as in the BISON code [21, 22].

4. – Spent Nuclear Fuel corrosion

The JRC has over 25 years of R&D experience in corrosion studies of spent fuel in groundwater under conditions relevant for geological disposal. The scope of this research has evolved over time to respond to the needs of Member State stakeholders. The studies address the following areas:

1. Geological disposal: The main objective is to provide experimental data on the effect of the environment on the fuel matrix corrosion and the associated instant release fraction [23]. Governing parameters are related to both the spent nuclear fuel conditions (*e.g.*, irradiation history) and the repository conditions (fig. 3).

2. The verification/confirmation that additive-containing and evolutionary LWR fuels will behave similarly to standard UO_2 fuel under repository conditions. Other types of spent fuel (ATF, fast reactor fuel not reprocessed) are also considered.

3. The comparative study of corium and spent fuel corrosion mechanisms. Research on how to deal with the aftermath of an occurrence or accident which caused release of radionuclides and contamination has been quite scarce. In particular, there have been relatively few investigations on the decontamination, remediation and waste management of a site after a severe accident.

Collaboration between the JRC and end-users (waste management organizations) is essential to address their specific needs. The scientific data and knowledge generated by the JRC feeds into the modelling of very long-term processes and reduces uncertainties in safety assessments.

5. – R&D programs for novel characterisation methods for difficult to measure isotopes

JRC performs also research and development activities in the field of *difficult to measure* radioisotopes determination [25]. A Saturated-Absorption Cavity Ring-down Spectroscopy (SCAR) high sensitivity, laser based spectroscopy technique has been adapted to operate in a nuclear facility for the precise determination of radiocarbon in nuclear and decommissioning waste. Results obtained in a preliminary proof of principle setup in collaboration with the Italian National Institute for Optics of the CNR demonstrated that the laser based SCAR technique is an accurate and relatively simple alternative to Accelerator Mass Spectroscopy (AMS) and Liquid Scintillation Counting (LSC) for the determination of ^{14}C content at mole fraction levels ranging from below the natural abundance to enriched [26]. The radiocarbon content values are provided in around 5 min, after a careful sample combustion has been done avoiding spectroscopic interference. SCAR represents a new class of relatively small footprint instruments potentially applicable to process many samples at the place of origin. The SCAR technique can also be adapted to measure other radiologically relevant nuclides, such as ^{36}Cl.

6. – Spent nuclear fuel and waste characterisation

JRC contributes as Task leader to the Spent Fuel Characterisation Work-package of EURAD. The objective is to produce experimentally validated procedures to determine reliable source terms of Spent Nuclear Fuel (SNF), including realistic uncertainties [27, 28].

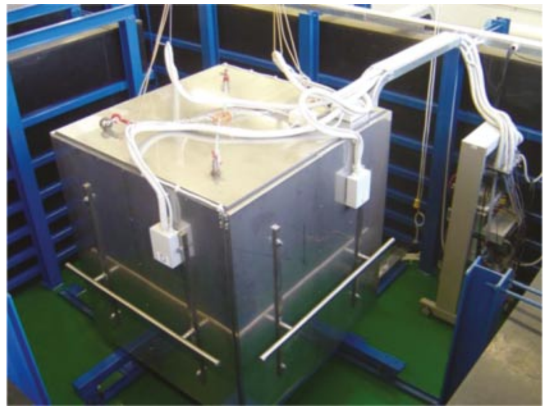
The main contribution of JRC is to develop Non-Destructive Assay (NDA) methods to validate the performance of depletion codes and to verify the declared fuel history. The focus is on NDA methods based on the detection of γ-rays and neutrons and on calorimetric systems determining the decay power. In addition, nuclear data needs to improve the accuracy of depletion calculations are identified and routes to improve them are defined. The nuclear data work is in collaboration with IJS, PSI (EPFL) and SCK CEN.

JRC in collaboration with Swedish Nuclear Fuel and Waste Management Company (SKB) performs a detailed study of the measurement principles of the calorimeter installed at Central Interim Storage Facility for Spent Nuclear Fuel (CLAB) to measure the decay power of SNF assemblies. The study includes an analytical model to describe the heat transport and Monte Carlo simulations to describe the γ-ray transport from the assembly to the pool. A variance analysis is applied to quantify uncertainties due to random and systematic effects. In addition, JRC in collaboration with PreussenElektra (PE) co-ordinates a blind test to assess the performance of depletion codes based on results obtained with this calorimeter.

An absolute method to determine the neutron production rate of a SNF segment, avoiding any reference to a representative SNF sample to calibrate the device [29], was deployed at the Laboratory for High and Medium level Activity of SCK CEN. The neutron production rate due to spontaneous fission is determined with an uncertainty that is a factor 2 smaller compared to radiochemical analysis. The results were used to study the performance of depletion codes, *i.e.*, ALEPH2, SCALE and Serpent2. An optimised detection system will allow measuring both high neutron emitting samples (*e.g.*, from irradiated MOX) and samples with a low neutron emission rate and relatively high γ-ray emission rate [30]. In collaboration with the Uppsala University, improved analysis procedures for measurements of SNF assemblies by γ-ray spectroscopy and the Differential Die-Away Self-Interrogation (DDSI) system developed at Los Alamos National Lab (LANL) are being developed.

Neutron based NDA are particularly suited for the assay of nuclear materials in waste due to the strong neutron signature arising from fission events and the high penetrability of fast neutrons of most waste matrix materials. Figures 4(a) and 4(b) illustrate two examples of JRC experimental R&D programme to develop very senstivite NDA instru-

(a) JRC Waste Drum Monitor



(b) PUNITA

Fig. 4.: JRC Waste Drum monitor: studies on passive neutron correlation methods aiming at Pu mass determination using spontaneous fission neutrons. PUNITA: studies on active neutron interrogation for mass determination of U and Pu content in waste packages.

mentation needed to verify compliance with waste acceptance criteria and to support verification efforts by safeguards authorities.

The JRC Waste Drum Monitor is a passive neutron counter for the assay of Pu in standard waste packages of the 210 litres. In total 148 ^{3}He neutron detectors surround the waste package. The very high neutron detection efficiency allows application of neutron correlation analysis to determine the Pu mass and two additional parameters of the waste item simultaneously. The Monitor has been deployed by Euratom at various waste repositories in EU for safeguards verification campaigns. In JRC, the instrument is used in decommissioning training courses, and for studies of improvements in data processing by reduction of electrical noise in detector electronics, and neutron detections from background sources. The Pulsed Neutron Interrogation Test Assembly (PUNITA) is a facility for R&D on active neutron methods. It incorporates the Differential Die-Away (DDA) technique for the mass determination of small quantities of U and Pu in waste, and includes a pulsed 14-MeV neutron generator. The pulsing of the external neutron source allows detection of the induced fission signatures in the time intervals when the neutron generator is switched off, a feature that provides exceptionally low detection limits. Currently, the PUNITA facility is used for studies of different fission signatures (prompt and delayed neutrons, delayed gamma rays), as well as neutron detection systems other than ^{3}He, such as liquid scintillation detectors and ^{4}He detectors.

7. – JRC open access to nuclear research infrastructures

JRC provides access to its nuclear research infrastructures via European Union (EU) collaborative research projects and agreements and, in addition, via the JRC open access programmes ActUsLab in Karlsruhe (Actinide User Laboratory) [31, 32], EUFRAT in Geel (European facility for nuclear reaction and decay data measurements) [33] and

EMMA in Petten (Laboratory of the Environmental & Mechanical Materials Assessment). Based on a call for proposals([1]), access is allocated on the basis of feasibility and scientific merit, following the recommendations of an internal feasibility check and an external scientific Review Panel. Users performing experiments at the JRC nuclear research infrastructures receive training on radiation protection and safety rules in the nuclear laboratories, training on the use of state-of-the art instruments and analysis tools and the handling of nuclear material. This contributes to maintaining the nuclear competences in the EU for the safe operation of nuclear facilities in the whole lifecycle of energy and non-energy applications. As an example, in the period 2020–2022, 6 out of 9 proposals specifically related to radioactive waste management have been accepted and implemented in ActUsLab.

8. – Policy support

JRC supports the implementation of the Council Directive 2011/70/Euratom [34] on responsible and safe management of spent fuel and radioactive waste. This activity contributes to the timely and proper implementation of the Directive by the Member States. The Commission reports and staff working documents on the implementation of the Directive, mainly drafted by JRC and based on the JRC technical reviews of national programmes and national reports, provide comprehensive overviews of the radioactive waste and spent fuel management situation in the EU and identify challenges in national policies and programmes. Additionally, JRC supports the development and execution of the International Atomic Energy Agency (IAEA) Integrated Review Service for Radioactive Waste and Spent Fuel Management, Decommissioning and Remediation (ARTEMIS) by participating as EC observer to peer-review missions and provision of feedback to DG ENER and (through DG ENER) to the IAEA; the development of the IAEA ARTEMIS peer-review service is supported by the European Commission.

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