STATUS OF THE EURAD RESEARCH PROGRAM ACTIVITIES ON IMPROVING SOURCE TERM PREDICTIONS FOR SPENT NUCLEAR FUEL

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ABSTRACT

The European Joint Programme on Radioactive Waste Management (EURAD) is a European Commission sponsored research collaboration and Spent fuel Characterization (SFC) is one of its projects. The status of the efforts concerning the theoretical investigations to improve the understanding of SNF source terms is described, followed by the research to develop, improve and demonstrate NDA methods/systems for SNF characterization, and finally the progress about determining the inventory of activation and fission products in cladding materials.

INTRODUCTION

The European Joint Programme on Radioactive Waste Management (EURAD) is a European Commission sponsored research collaboration towards safe radioactive waste management and disposal [1]. The project consists of 13 work packages and Spent fuel Characterization (SFC) is one of them. SFC in turn is made up of 4 tasks. They focus on fuel property characterization and related uncertainty analysis, behavior of spent-nuclear fuel (SNF) pellets under interim storage conditions and finally accident scenario and consequence analysis.

In the following sections the status of research on SNF fuel property characterization in the EURAD project is described. The focus is on source terms relevant for safe and secure handling, transport, intermediate storage and final disposal [2,3]. In particular neutron emission rates, γ-ray emission spectra, decay heat rate and the inventory of specific nuclides are considered. Nuclides of interest are 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) for example.

The inventory of fissile nuclides is needed for nuclear safeguards requirements and reactivity calculations to prevent criticality [4,5]. The inventory of strong neutron absorbing fission products and actinides allows to credit burnup [6] to avoid overly conservative criticality margins. The inventory of activation products and long-lived FP are important to study the impact to the biosphere during final disposal [7]. The complete list of nuclides in SNF is hard to be measured directly for every single fuel assembly. Therefore, they are estimated based on a combination of calculations with validated codes [8,9] and non-destructive analysis (NDA) measurements [10]. The calculations involve neutron transport solutions during irradiation and nuclide creation and depletion chains both during irradiation and storage and disposal.

In the following sections the theoretical investigations of the project to improve the understanding of SNF source terms is described, followed by the research to develop, improve and demonstrate NDA methods/systems for SNF characterization, and finally the progress for determining the inventory of activation and fission products in cladding material is described.

POTENTIAL TO IMPROVE CALCULATIONS FOR SNF SOURCE TERM DETERMINATION

The results of source term calculations depend on many factors: nuclear data (e.g. microscopic cross sections, fission product yields, neutron emission probabilities and spectra, decay data), 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".

The research activities are divided into two parts. In the first part several PIE (post irradiation examination) samples were selected from the SFCOMPO database [11] and from proprietary experiments available to the project to compare measured and calculated nuclide vectors and decay heat, if possible. The 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 (UO2 or MOX), see Table 1. The aim is to determine well-justified uncertainties due to fuel history, microscopic data and fuel models and to give recommendations for best-practice source-term determination. In total, 17 PIE samples were analyzed, some of them in parallel by different participants. For decay heat, the calorimetric measurements from CLAB, GE-Morris and HEDL were considered, details see for example in [12,13].

Direct observables like decay heat are determined by the sum of contributions from many individual nuclides. The rank of contribution depends on the point in time the decay heat is observed. If fission products are mainly contributing, then uncertainties in fission yields are important followed by their neutron capture cross sections and decay data. For actinides uncertainties in neutron absorption cross sections are first in rank. These dependencies are in turn influenced by fuel history. As a result, a complex network of factors determines a fuel assembly's nuclide vector at the end of irradiation. The consequences for decay heat and other observables are different for different points in time. While codes can be calibrated during time scales accessible to observations (e.g. decay heat standards like in [14] or [15]) the large periods relevant for final disposal mean that nuclide vector uncertainties are more important to a guide conservative predictions.

In Fig.1 the aggregate C/E (code vs. experiment) results for several PIE samples are shown for selected actinides and fission products. Even though the impact of some systematic effects due to the fuel history will average out over many samples there remain noticeable differences for some nuclides. Also, the range of the experimental uncertainties has a visible nuclide dependency. Part of the objective of the project is to develop guidance how these results can be interpreted: the role of biases coming from nuclear data, the role of fuel history conditions and the role of measurement uncertainties.

In the second part of these research activities source terms of a set of samples (with measurements at the same facility and under similar circumstances) will be calculated and compared against experimental results, primarily decay heat and neutron- and γ-ray data. Focus will primarily be on the SKB-50 dataset which consists of recent measurements from Swedish BWR and PWR fuel assemblies [16] and is a follow up to an earlier campaign [17]. Burnup covers values between 10 and 60 MWd/kgU and cooling time between 2000 and 14000 days. The main objective is to obtain statistically relevant statements by applying a standard, recommended calculation procedure to many samples of comparable quality. Calculations will be performed both with an established code like SCALE6 [18] and a more sophisticated code like EVOLCODE2 [19]. The spread of results (C/E values) will be analyzed in the light of those expected from PIE evaluations described above. The role of fuel assembly averaged quantities compared to small size PIE sample quantities will be considered. Recommendations for best-practice predictability of fuel assembly source terms in the current time and for future time frames are going be formulated.

DEVELOPMENT, IMPROVEMENT AND DEMONSTRATION OF NEW NDA METHODS

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 that are based on the detection of gamma-rays and neutrons and on 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 the need of any calibration using a representative sample was developed, validated and demonstrated [20]. A standard neutron well-counter designed for routine nuclear safeguards applications was applied in this case. The detection efficiency of the device was derived by combining measurements with a ^{252}Cf(sf) point source and results of Monte Carlo simulations. The detection efficiency for the ^{252}Cf(sf) point source was obtained from the total and real coincident count rates based on the point model proposed by Hage and Cifarelli [21]-[24] and Böhnel [25]. The results of the Monte Carlo simulations were used to account for the difference in geometry of the SNF segment sample and for the differences in energy distribution with respect to the ^{252}Cf(sf) point source. These simulations were also used to estimate additional effects such as neutron leakage and fission probabilities for different neutron energy distributions.

The sample was a segment taken from a 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 and therefore the uncertainty of the fuel history is minimal. Fig. 2 shows a schematic representation of the transfer and GT-75 transport container to move the SNF segment sample from the hot cell to the detection device for the measurements under standard controlled area conditions. The influence of the following components on the neutron production rate S_{sf} and α-ratio uncertainty were taken into account: total and real coincidence rate, the fraction of delayed neutrons, the detection efficiency for prompt spontaneous fission neutrons, gate fraction and first and second order normalized factorial moments of the multiplicity distribution for spontaneous fission. The results suggest that the neutron production rate of a SNF sample due to spontaneous fission and related ^{244}Cm inventory can be derived with an uncertainty in the order of 1.5 %.

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 operation and the Differential Die Away Self-interrogation system (DDSI) that was developed at the Los Alamos National Laboratory [26]. A study of systematic effects related to gamma-ray spectroscopic measurements, such as the determination of the net peak area and the positioning of the assembly with respect to the detector, was performed was carried out [28]. In addition, a model was developed to determine the inventory of ^{134}Cs and ^{154}Eu relative to the ^{137}Cs inventory without the need of additional calibration measurements or gamma-ray transport calculations.

DETERMINATION OF THE INVENTORY OF ACTIVATION AND FISSION PRODUCTS IN CLADDING MATERIALS

Firstly, impurities are introduced into the cladding during fabrication. Secondly, the insertion of pellets into the cladding leads to traces of fuel being adherent on the inner cladding surface. The existence of uranium on the inner surface obviously means 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. Thirdly, during operation fission fragments are sputtered off the pellets and impact the cladding. Fourthly, due to cladding creep-down and pellet swelling there is direct contact between cladding and pellet at higher burnup during operation and surface deposition of nuclides onto the cladding occurs.

Currently fuel assemblies are about to be stored for much longer periods than originally foreseen, and the feasible failure of the cladding within dry storage casks could become an obstacle in handling the fuel rods later for final disposal. Evidently, the ability of quantifying those radioactive nuclides which cause radioactive defects in the cladding via high-energy alpha particles is of importance. Furthermore, the exact material composition of the cladding itself is a key issue concerning the sustainability of the thermal and mechanical loads on the irradiated material [28].

In view of the above, the objective is to analyze the inventory of fission and activation products present in irradiated Zircaloy. The importance of this analysis lies in the assessment of the integrity of the cladding within the dry storage period, which could reach up to 100 years or more. Dedicated simulations are ongoing to compare and validate codes and their burn up procedures to estimate the fission product concentration present in the cladding material.

For the experimental studies, two kinds of samples were prepared:(a) cladded fuel pellets of UO2 (50 GWd/tHM) irradiated in a pressurized water reactor (PWR); (b) a plenum cladding obtained from a UO2 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 [29] and evaluation of results is ongoing.

OUTLOOK

The research activities cover a wide spectrum of parameters relevant for interim storage and final disposal: nuclide vectors, source terms (decay heat, neutron and γ-ray sources), NDA measurement techniques and fuel cladding characterization for long-term storage. The interdisciplinary effort and the necessity of collaboration among many European partners is self-evident. The project brings the opportunity to condense the spread-out expertise about these fuel assembly parameters into a more coherent understanding,

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TABLES AND FIGURES

Table 1: PIE sample overview used in the project

Sample	Burnup MWd/kgHM	Enrichment wt%	Cooling Years	Fuel Type	Reactor Type
BM1	47	2.36	$5-7$	MOX	PWR
BM3	47	2.36	$3-7$	MOX	PWR
GU1	59	3.5	$3-6$	UO_2	PWR
GU3	52	4.1	$1.6-2.3$	UO_2	PWR
U1	34	4.1	$7-9$	UO_2	PWR
SF95-5	30	4.11	4	UO_2	PWR
SF95-4	37	4.11	4	UO_2	PWR
S1.PWR				UO_2	PWR
A1-B23-I2680	27	2.53	3	UO_2	BWR
C5-B23-K2680	25	2.53	3	UO_2	BWR
ENRESA-1	50	3.95	4	UO_2	BWR
ENRESA-2	51	3.95	4	UO_2	BWR
ENRESA-3	50	3.95	4	UO_2	BWR
ENRESA-4	51	3.95	4	UO_2	BWR
ENRESA-5	44	3.95	4	UO_2	BWR
ENRESA-6	43	3.95	4	UO_2	BWR
ENRESA-7	49	3.95	4	UO_2	BWR
ENRESA-8	38	3.95	4	UO_2	BWR

Figure 1: Differences between the calculated (C) and experimental (E) nuclide inventory. The uncertainties are from experimental, fuel history and microscopic contributions.

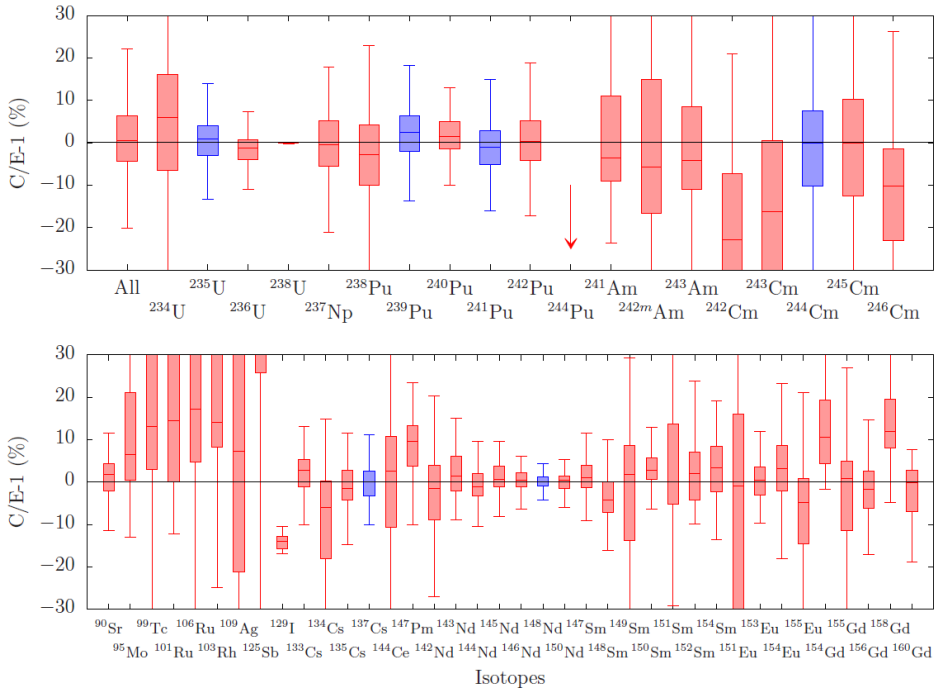
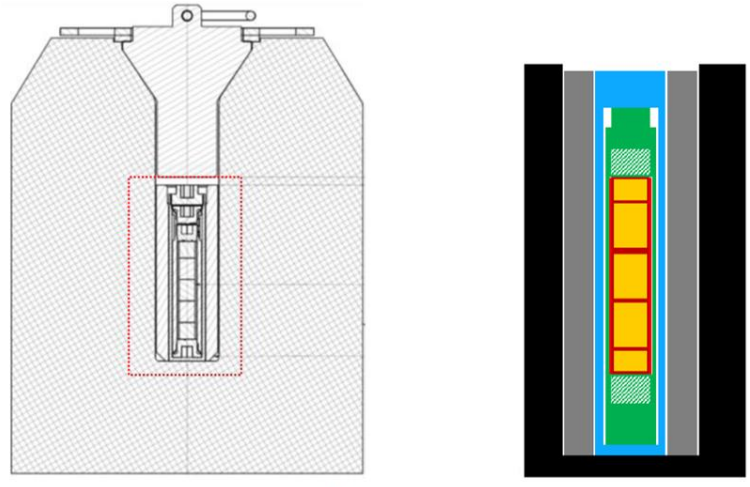


Figure 2: Drawing of the transfer and GT-75 transport container of SCK CEN loaded with the SNF sample. The color coding on the right is as follows: orange/red for the SNF sample; green for the stainless-steel capsule; blue for the aluminum capsule; grey for the sleeve and black for the lead shielding.



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