Criticality Safety in the Waste Management of Spent Fuel from NPPs

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Abstract:
During irradiation in the reactor core, fissile material from nuclear fuel is depleted for power generation. However, significant amounts of fissile nuclides, mainly ^{235}U and ^{239}Pu, are still present in spent fuel unloaded from the core, with the inherent potential for maintaining an inadvertent nuclear chain reaction under unfavorable conditions. Due to this reason, criticality safety has to be demonstrated for all stages of waste management of spent nuclear fuel. This pertains e.g. to wet storage in cooling ponds, dry storage and transport of the fuel in casks, dissolver stages in reprocessing plants, as well as the direct final disposition of the spent fuel. Usually, for system design the reactivity of the spent fuel has been considered as being equal to fresh fuel. This can lead to large safety margins and thus an over-conservative design of corresponding waste management systems. In the recent past, the reduction in reactivty due to irradiation of the fuel is considered to a greater extent in the design of new systems or the modification of existing systems, e.g. due to the increase in initial enrichment. However, this so-called "burn-up credit" imposes additional efforts on the validation of calculation systems and nuclear cross-section data bases.

1 INTRODUCTION

In the criticality safety analysis for systems including spent nuclear fuel, usually the reactivity of the fuel has been considered as being equal to fresh fuel. This facilitates the analysis as no burn-up calculation for inventory determination is necessary. Only few isotopes, mainly ^{235}U and ^{238}U as well as moderator and structure materials, need to be considered in the criticality calculation. However, in the recent past, in modern spent fuel waste management systems (WMS), the benefits of the *de facto* reduction of reactivity in spent fuel have been considered. Higher initial enrichments on the one hand or more dense packing of assemblies within storage racks or dry casks on the other hand become feasible by considering the fuel burn-up in the criticality safety design. While in the time frame of interim storage, as long as full control over the WMS is given, the “classical” deterministic criticality safety analysis is mandatory, for the long term evolution of a final repository for spent fuel assemblies, additional probabilistic analyses have to be performed to demonstrate sub-criticality for up to 1 million years.

2 CRITICALITY AND BURN-UP OF SPENT NUCLEAR FUEL

The term criticality denotes a self-sustaining nuclear fission chain reaction. Due to its high energy release, it has to be avoided outside dedicated facilities, e.g. power reactors or research facilities. The state of criticality is described by the neutron multiplication factor k_{eff} which equals one in case of a self-sustaining, stable chain reaction. Due to the fact that in spent fuel, still between 1 and 1.5 % fissile material as ^{235}U and ^{239}Pu is present, criticality safety has to be demonstrated for the management of spent nuclear fuel.

The detailed amount of fissile material as well as neutron absorbing species as e.g. ^{240}Pu depends among others on the burn-up of the spent fuel. This is the energy produced per kg initial heavy metal and is the sum of the energy released by fissions of various nuclides and from other nuclear reactions as e.g. from (n,γ)-reactions. Burn-up is an integral measure and does not denote in detail the way the specified energy has been produced. However, the latter depends on irradiation details as neutron flux and height, coolant and fuel temperature, neutron leakage, and others. Hence the numerical value of burn-up is not a direct measure for the detailed nuclide composition of spent nuclear fuel. Moreover, as the features mentioned above typically vary horizontally and vertically inside the core, burn-up also varies locally within single fuel assemblies. But the neutron multiplication properties for a given system containing spent fuel are determined mainly by its nuclide inventory, so, in consequence, burn-up and k_{eff} do not correlate directy to each other.

2.1 Burn-up Credit and Loading Curve

A spent fuel WMS designed with the application of burn-up credit requires quantification of a conservative minimum burn-up of the spent fuel assembly. This in principle applies for every single storage cell or position but may be covered by a single bounding burn-up value. Here, conservative bounding assumptions have to be applied especially to the determination of the nuclide inventory and, if applicable, on the shape of a bounding axial burn-up profile, cf. also chapter 2.3 below. As the minimum required burn-up, amongst others, depends on the initial enrichment, this has to be accounted for dedicatedly in the criticality safety analysis. The so-called *loading curve* denotes the relationship between the initial enrichment of the fuel and its minimum required burn-up which is required to allow for placement of the fuel into the WMS. The loading curve is a feature based on the characteristics of the fuel itself, the power plant and its typical operation features, as well as the WMS itself and defines the criterion if an assembly may be loaded into the respective loading position or not. This relationship belongs to this unique environment and is no more valid if any of these conditions change. It includes well defined bounding conditions for e.g. the irradiation history, to ensure that an assembly of given burn-up does not exceed the reactivity on which the determination of the loading curve was based.

One special feature of burn-up credit is the need for redundant controls and measures against inadvertant misload of a fresh assembly or an assembly which burn-up is not sufficient to allow for placement into the corresponding position in the WMS. A robust burn-up credit design may allow for the erroneous placement of one fresh assembly into the WMS without reaching criticality.

2.2 Nuclide Inventory

In contrast to fresh fuel, spent nuclear fuel comprises a high number of nuclides which have been generated by fission, neutron capture reactions and radioactive decay. These reactions are mainly determined by neutron flux spectrum and height. Consecutively, the nuclide inventory, both higher actinides and fission products, vary for each spent fuel assembly. Moreover, the composition varies with the irradiation conditions and is inhomogeneous axially and horizontally for each fuel assembly. Figure 1 depicts the build-up and decay reactions for the higher actinides in the fuel during irradiation and later on during storage.

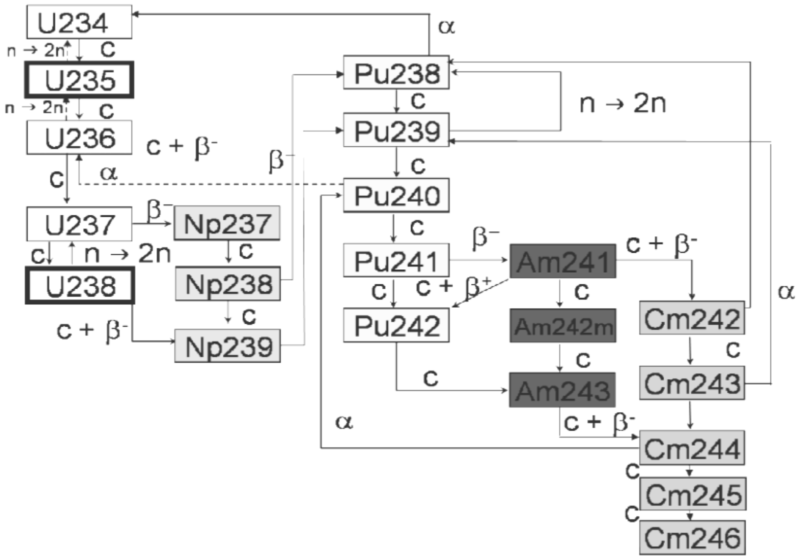

Figure 1: Build-up of major actinides during fuel irradiation in the reactor core

In a burn-up credit approach, all nuclides increasing reactivity have to be included in the analysis. For spent uranium dioxide fuel, these are mainly ^{235}U, ^{239}Pu and ^{241}Pu. For very long term periods, i.e. final disposition, ^{233}U originating from ^{237}Np decay also might have to be included. Absorbing fission nuclides—the beneficial constituents for burn-up credit—which have the potential to be included in the criticality safety analysis need to be present during the whole license period (preferrably they are stable) and shall not be volatile even under accident conditions. They need to comprise a high neutron capture cross section or have to be present in a sufficiently high amount. The most important absorbing nuclide in spent fuel is ^{240}Pu, but also ^{236}U and ^{241}Am may be included. Typical fission product candidates for inclusion in a burn-up credit approach are ^{95}Mo, ^{99}Tc, ^{101}Rh, ^{103}Ru, ^{109}Ag, ^{133}Cs, ^{143}Nd, ^{145}Nd, ^{147}Sm, ^{149}Sm, ^{150}Sm, ^{151}Sm (decaying to ^{151}Eu), ^{152}Sm, ^{151}Eu, ^{153}Eu and ^{155}Gd [1]. In the inventory determination for the criticality safety analysis, conservative isotopic correction factors coming from benchmark calculations for the implemented nuclides, may have to be included for both fissile and absorbing nuclides.

2.3 Axial Burn-up Profiles

As the neutron flux varies along the axis of a fuel assembly, the burn-up and its consecutive nuclide inventory varies and results in an inhomogeneous burn-up profile. The shape of this profile is determined by neutron leakage at top and bottom of the core, the temperature gradient in the core from inlet to outlet accompanied by a moderator gradient, the total average burn-up, and other local features given by the plant and its usual operation mode. It is basically unique for each fuel assembly. Figure 2 shows a typical axial burn-up profile for a German PWR. It is normalized to the average burn-up value of the whole assembly and averaged over 850 profiles taken from in-core measurements [2].

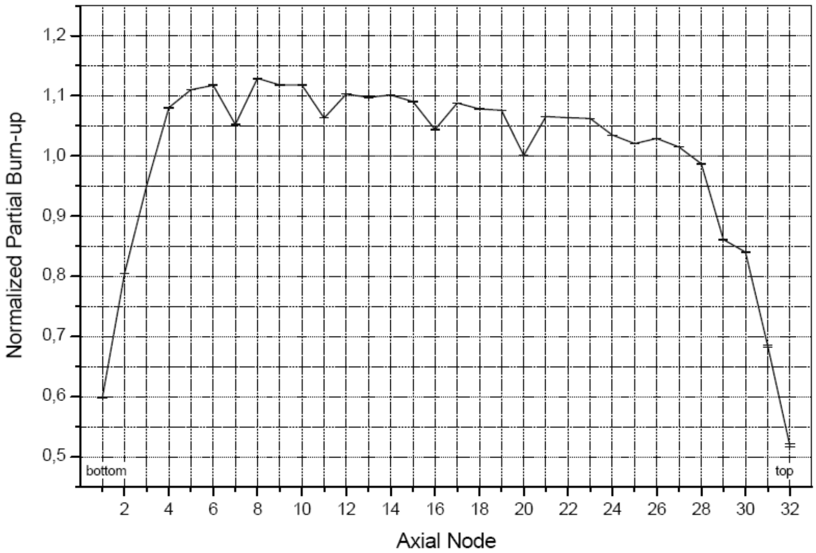

Figure 2: Typical normalized axial burn-up profile from a German PWR reactor

Here the lower burn-up at top and bottom due to leakage effects can be clearly figured out. The middle part comprises a plateau-like shape, slightly falling towards the top end. The latter is caused by the in-core temperature and thus moderator gradient. As the neutron spectrum also varies along the assembly axis, zones with equal numerical burn-up value not necessarily comprise identical local nuclide inventories. In a criticality analysis, consideration of the axial burn-up profile on the one hand and a uniform, homogenized inventory based on the average burn-up on the other hand typically do not yield the same k_{eff}. The difference is known as "end effect" as the lower burnt ends of the assembly drive the reactivity of higher burnt fuel. It is calculated by the formula $\Delta k = k_{axial} - k_{uniform}$ and is typically positive for burn-ups higher than about 15 to 20 GWd/tHM, depending on various parameters. It increases further at increasing average burn-ups. This means that neglection of the profile at higher burn-ups is non-conservative, and the profile has to be accounted for in a criticality safety analysis. As each assembly shows a different profile, typically bounding assumptions are applied.

2.4 Code Validation

2.4.1 Inventory Determination

For the validation of the inventory determination code, radiochemical assay data from post-irradiation examination (PIE) experiments have to be recalculated. From these calculations, isotopic correction factors for the mass of each nuclide can be derived, to implement the calculated inventories conservatively bounding into the criticality safety analysis. However, publically available PIE data are limited and often lack information on nuclides potentially of interest to burn-up credit applications, e.g. certain neutron absorbing stable fission products. Recently published data from the ARIANE programme or data gathered in the frame of the former Yucca Mountain Project in the United States, e.g. TMI-1 or Calvert Cliffs, have alleviated the situation.

2.4.2 Criticality Code

To estimate bias and uncertainty of a criticality calculation code system, critical benchmark experiments representative for the burn-up credit application under scope have to be evaluated. This implies that nuclides which shall be implemented in the burn-up credit approach also require benchmark systems including those nuclides. Today, for plutonium isotopes, a high number of good quality benchmarks is available. In contrast, only few experiments are publically available which have burn-up fission products. For some of the most interesting fission products, no benchmarks are available at all in free literature. Hence validation of these nuclides becomes unfeasible, and they cannot be included in the analysis. This comprises one major drawback on the application of burn-up credit.

3 APPLICATION

3.1 Wet Storage of Spent Nuclear Fuel

On-site wet storage cooling ponds at nuclear power plants are often divided into two regions: One region which design allows for placement and storage of a reactivity equal to fresh fuel of the maximum enrichment. This requires large dimensions and the use of absorber plates for each single storage cell. A second region may use burn-up credit for its design. This allows for a more dense placement of fuel assemblies, which now have to fulfill a minimum burn-up criterion to be placed into this region. On the other hand, redundant controls of the assembly burn-up and measures against misplacements are mandatory. Moreover, a robust design allows for single inadvertant assembly misplacements without reaching criticality. At certain facilities, burn-up credit was a possibility to overcome storage limitations in terms of increase of initial enrichment: While the design of the wet storage rack had been made on basis of the fresh fuel assumption for the former maximum enrichment, now with increased enrichment a—mostly small—minimum burn-up of the assembly is required to placed into the rack; with other words: Burn-up credit is used.

3.2 Transport and Dry Storage of Spent Nuclear Fuel

For the use of burn-up credit in dry casks for storage and transportation, in principle the same argumentation is valid as for the wet storage pools discussed in chapter 3.1 above. Instead of increased storage capacity now a reduced number of casks for a given amount of spent fuel is necessary. In terms of transportation, an additional benefit by reducing the number of transport procedures for the same number of spent fuel assemblies comes into play. In Germany, the GNS CASTOR© V/19 now is licensed for assemblies of increased initial enrichment on basis of a small amount of burn-up and on basis of actinides only without inclusion of absorbing fission products. Moreover, the Federal Office for Radiation Protection expects the licensing of a transport and storage cask, the Transnuclear International TN24E, in 2010; here a minimum burn-up of 12 GWd/tHM at 4.05 % initial enrichment is required, including actinides and—for the first time for a dry cask in Germany—six fission products in the criticality safety analysis [3].

3.3 Final Disposition of Spent Nuclear Fuel

During the operational phase of the repository, disposal casks are emplaced within the disposition area. Until closure of the repository, criticality safety is essentially given by design of the repository, the casks, handling procedures etc. So for the operational phase, the same argumentation applies as for other transport and storage dry cask systems. After the final closure of a repository for spent nuclear fuel, the so-called post-closure phase, typically the control over the system is (intentionally) lost. Due to the lack of knowledge on the long term evolution of the repository, criticality safety analysis cannot be based on deterministic methodologies alone. Moreover, probabilistic aspects become more and more important. Additionally, the radioactive decay of important fissile and absorbing nuclides shows significant effects on the nuclide inventory and thus the reactivity of the spent fuel on geological time frames.

3.3.1 Neutron Multiplication Factor

Due to the radioactive decay and different half lives of its main contributors, k_{eff} varies significantly over time and especially during geological time scales. Figure 3 depicts the evolution of k_{eff} for a generic disposal system from 5 up to 10^6 years after discharge from reactor [4].

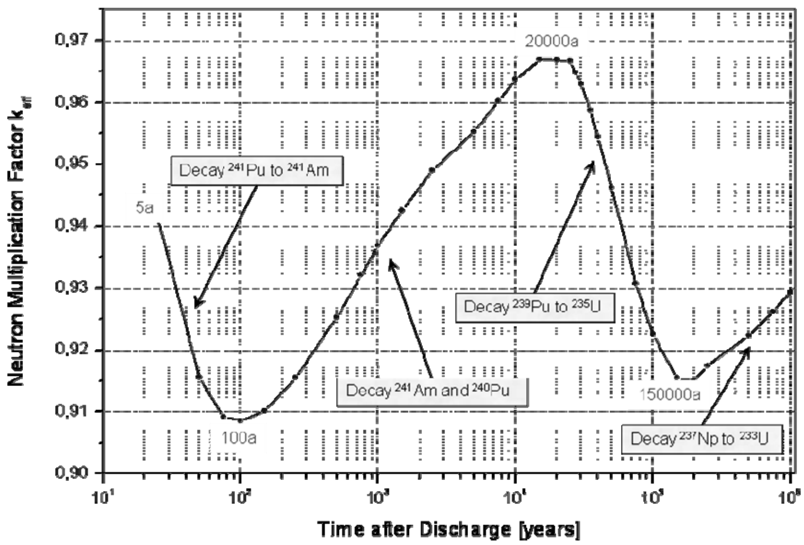

Figure 3: Evolution of the neutron multiplication factor k_{eff} fora generic spent nuclear fuel system over time

The decrease of k_{eff} between 5 and 100 years after discharge is due to the decay of the fissile ^{241}Pu to the absorbing ^{241}Am with a half-life of 14.35 years. The increase in k_{eff} up to 20000 years after discharge is due to the decay of the absorbers ^{241}Am (432.2 years) and ^{240}Pu (6563 years) mainly. The subsequent decrease is because of the decay of ^{239}Pu to ^{235}U (24110 years), and finally the increase after 150000 years is due to the generation of the fissile ^{233}U by the decay of ^{237}Np. These changes in inventory have duly to be taken into account when analysing credible evolution scenarios of the repository.

3.3.2 Scenarios and Probabilistic Assessment

As the in-detail long term evolution of a final repository is basically unknown, credible evolutions scenarios have to be developed and analysed [4]. For the assessment, scenarios for a criticality under repository conditions have to be defined, then credible pathways have to be found leading to this scenario, including time frames, reaction rates, branching probabilities etc. On this basis, by a probabilistic assessment the likelyhood of a critical excursion can be estimated. Recent preliminary analyses yield to a conditional probability for such an excursion on basis of a postulated water ingress into the repository of $P_{cond} < 10^{-6}$. However, these studies are not completed up to now [4]. Figure 4 shows an example for such a hypothetical evolution pathway for a generic repository for spent nuclear LWR fuel leading to to a supposed critical excursion.

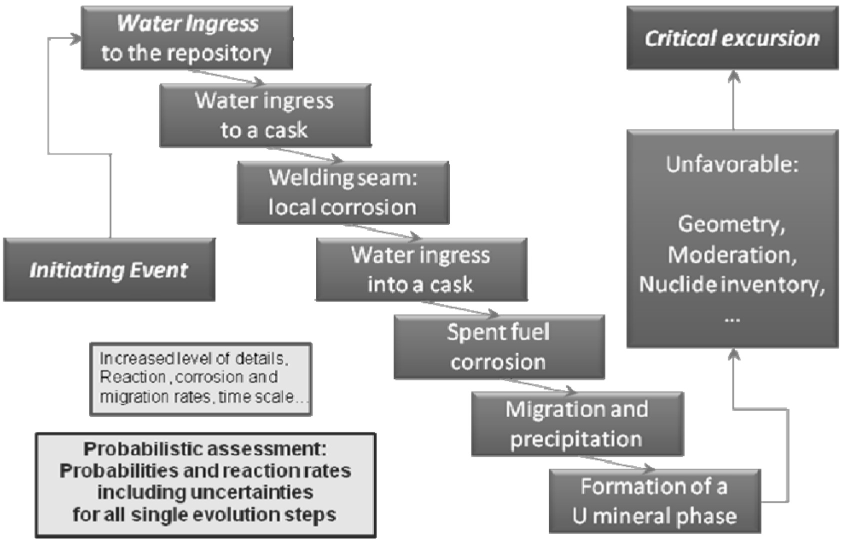

Figure 4: Hypothetical exemplary evolution pathway for a generic repository for spent nuclear fuel leading to a supposed critical excursion

3.3.3 Consequence Analysis

As a critical excursion under repository conditions cannot be completely excluded up to now, such an excursion can be postulated and its consequences on the repository with its technical and natural barriers can be estimated. Therefore, a dedicated tool named PUNKT-V6 is currently under development in GRS [5]. The aim is to calculate the power level, heat and fission product generation as well as the transient behavior of a critical excursion. Other questions are whether the excursion could be self-terminating, or how long it could endure. The work on this code is still ongoing.

4 SUMMARY

Due to the residual amount of fissile nuclides in spent nuclear fuel, for spent fuel waste management systems sub-criticality has to be demonstrated. Rather than the assumption of fresh fuel, nowadays more and more realistic assumptions on the properties of spent fuel are applied. The so-called burn-up credit requires the determination of the nuclide inventory as well as a sophisticated criticality safety analysis. Both inventory determination code and criticality calculation code require increased efforts on code validation. Due to its economical and environmental benefits, burn-up credit is today applied in several nuclear power plant operating countries, amongst them also Germany. Typical applications range from wet storage pools, transport and storage dry casks, over reprocessing dissolver stages up to the design of a final repository and its long term evolution.

5 ACKNOWLEDGEMENTS

The work and results presented in this paper are in major parts been funded by the German Federal Ministry for the Environment, Nature Conservation and Nuclear Safety, and by the German Federal Office for Radiation Protection.

6 LITERATURE

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