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REVIEW OF BURN-UP CALCULATION VALIDATION FOR SPENT PWR URANIUM OXIDE FUEL INCLUDING ACTINIDES AND FISSION PRODUCTS

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ABSTRACT

With the increasing amount of spent fuel assemblies with high initial ^{235}U enrichment stored at nuclear facilities the need to account for the burn-up of the fuel in criticality safety assessments for transport and storage casks becomes more important. In the past, this burn-up credit was mostly limited to considering the changes in the concentration of the main actinides only, as those isotopes have been investigated thoroughly as concerns burn-up credit. But recently a method for applying burn-up credit based on nine actinides and six fission products to a package for spent PWR uranium oxide fuel (TN 24 E) by TN International has been approved for the first time by the German competent authority BfS.

The paper focuses on the review of the validation of the spent fuel nuclide inventory calculation. In order to enable the use of burn-up credit for fission products, experimental data from French post-irradiation examination experiments have been provided by the applicant and reviewed by the German competent authority BfS. The review additionally included post-irradiation examination results from other international sources and calculations with the TRITON module of the modeling and simulation code system for nuclear safety analysis SCALE. Procedures and limitations for the validation of nuclide inventory calculations are discussed. The results also emphasize the need for evaluated high quality PIE data applicable to actinides and fission products.

INTRODUCTION

Spent modern fuel assemblies with high ^{235}U initial enrichment can be transported more efficiently if their irradiation in the reactor is taken into account in the criticality safety assessment for the package, as this will allow for more fuel assemblies to be stored and transported in a single cask. This method is called Burn-up Credit (BUC) and considers the change in neutron multiplication due to the change in nuclide composition of the fuel. It consists of calculating the nuclide inventory after the irradiation (burn-up calculation) followed by calculating the neutron multiplication factor of the package loaded with spent fuel of this calculated nuclide composition (criticality calculation). The use of BUC is restricted by the requirements in computational hard- and software and validation of the method. Within the BUC method an increasing number of nuclides taken into account in the calculations leads to a further increase in the cask loading capacity, but also to an increase of the validation effort. For this reason BUC in the past was mostly limited to accounting for changes in the concentration of

actinide isotopes only, for which a significant amount of validation data is available.
Recently BfS finished the review of the validation of BUC for the package TN 24 E for the transport and storage of spent PWR uranium oxide fuel according to an application by TN International (TNI). This application took into account nine actinides and six fission products, posing a real challenge regarding the validation of the method.
In the paper the basis for the validation of the burn-up calculations for this package and the result of the review of the validation by BfS are described.

NEED FOR VALIDATION OF THE CALCULATIONAL METHOD

Any criticality safety assessment requires a validation of the method and computer codes used.
The use of BUC increases the validation effort as follows:

- The method for calculating the nuclide content in the spent fuel (burn-up calculation) has to be validated against results of radiochemical post irradiation examination (PIE) of samples from irradiated fuel assemblies. To be suitable for validation the PIE samples should be similar to the fuel assemblies to which BUC is applied regarding type (PWR/BWR, UOX/MOX), initial isotopic composition and burn-up. The data publicly available are based on a handful of PIE from commercial light water reactors from Europe, Japan and the USA. PIE data are collected and analyzed by the OECD/NEA WPNCS Expert Group on Assay Data of Spent Nuclear Fuel (ADSNF) [1].
- The method for calculating the neutron multiplication factor (criticality calculation) has to be validated against critical experiments taking into account all nuclides included in the calculations. Methods for such validation and derivation of an upper subcritical limit (USL) are described e.g. in [2]. The critical experiments used for validation should be carefully selected regarding similarity to the application system concerning parameters like fuel composition, moderator and moderation ratio and neutron absorbers. A large database of evaluated critical experiments can be found in [3], some of them are applicable to BUC. Details of the validation of the criticality calculation are not subject of this paper.

The need for a sufficient experimental base for validation of both the burn-up calculation as well as the criticality calculation limits the applicability of BUC and the selection of nuclides taken into account significantly. More information about BUC in available e.g. in publications of the OECD/NEA WPNCS Expert Group on Burn-up Credit Criticality Safety (BUC) [4].

APPLICATION CASE AND SELECTION OF NUCLIDES FOR BUC

This paper deals with BUC as applied by TNI to the package TN 24 E for transport and storage of spent PWR fuel assemblies from German NPP. BUC is necessary for this package due to the high initial enrichment of some of the fuel assembly types. For the first time in Germany, the BUC approach is based not just on nine actinides (^{235}U, ^{236}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am) but also on six of the major fission products, namely ^{103}Rh, ^{133}Cs, ^{143}Nd, ^{149}Sm, ^{152}Sm and ^{155}Gd. As usual in Germany, the BUC is limited to a burn-up of 12 GWd/tHM, mainly accounting for the irradiation during one normal operational cycle in the reactor. The set of fission products taken into account has been selected mostly based on the availability of data suitable for validation. Publicly available PIE data are focused on actinides, most fission product have been measured only in a small number of experiments. To fill this gap, TNI has acquired proprietary data suitable for the validation of the depletion calculation. On this basis TNI has produced in cooperation with CEA and IRSN a validation report for the French APOLLO2 / DARWIN depletion codes used for the TN 24 E application. The validation is limited to the fuel assemblies and the burn-up range necessary for the application.
The PIE data were provided by CEA, EDF and AREVA in the form of measurements of samples

from the French reactors Bugey3 and Gravelines. For the analysis, 14 samples from fuel rods from several fuel assemblies have been analysed after varying times in the reactor core. The radiochemical analyses were performed on cuts from the extracted rods which were taken from the axial area with the highest burn-up. The data include measurements from a burn-up range of about 20 GWd/tHM to 61 GWd/tHM and enrichments in ^{235}U between 3.1% and 4.5%. They provide nuclide concentrations for the nine actinides as well as the six fission products, but not all nuclides were measured for all samples. Thus, 14 measurements were available for all U and Pu isotopes, but only 9 measurements for ^{143}Nd, 8 for ^{133}Cs and ^{241}Am and 5 for ^{103}Rh, ^{149}Sm, ^{152}Sm and ^{155}Gd. As the history of the investigated fuel rods was known, the chemical composition was then calculated by the burn-up code. The results of these calculations were used for a comparison with the chemical assay data, and from this comparison a conservative correction factor for each nuclide was calculated.

The validation report submitted by TNI includes a thorough investigation of the uncertainties involved in all of the measurements and calculations. For the PIE data evaluation, the report covers all aspects of the irradiation history, like the measurement of the burn-up of the samples, the moderator temperature to within 2 degrees, the fuel temperature to within 50 degrees, the uncertainty of the chemical analysis as well as the approximations involved in the modelling of the experiments. All of these uncertainties are included in determining the correction factors for the depletion calculations. An additional conservatism is included, as a correction factor is only applied if it is conservative on its own, i.e. a correction factor is only applied if this leads to a reduction in the concentration of an absorber nuclide or an increase of the concentration of a fissionable nuclide. This way, only correction factors leading to the largest neutron multiplication factor get applied.

For validation of the criticality calculation the “HTC” and “Fission Product” experiments (see description in [5, 6]) including the same set of nuclides were taken as a basis, supported by oscillation experiments in the MINERVE reactor [7]. The validation of the criticality calculation will not be discussed here.

REVIEW BY THE GERMAN COMPETENT AUTHORITY

During the review of the criticality safety analysis for approval of a package design the German competent authority BfS assesses the calculations done by the applicant, usually including own, independent calculations. For this purpose, the SCALE code package has been used in Germany since 1998 in varying versions.

Regarding the application of BUC to the TN 24 E package BfS decided that the experimental basis taken by TNI for the validation was not sufficient. At first, the number of measurements was too small for some of the nuclides. Additionally, due to common methods used in the assessment of the irradiation conditions and the radiochemical measurement for the PIE, uncertainties may be underestimated by taking into account only these data. BfS therefore introduced into the validation additional independent experiments as described below.

The review of the burn-up code validation by the German competent authority BfS was based on the following concept:

- TNI presented for validation as well as for the application case calculations with the burn-up code DARWIN. Directly redoing DARWIN calculations by BfS was considered to be not reliable due to the lack of experience with this package at BfS.
- BfS performed own calculations for validation and application with the TRITON module of SCALE 6.0 (and SCALE 5.1, see below). The calculations by BfS aimed at assessing the value of the Bugey3 and Gravelines PIE data by comparing them to other experiments and at validating TRITON including derivation of nuclide correction factors from the combined set of PIE experiments. Finally the application calculations were repeated with TRITON

for comparison of the corrected nuclide composition with that calculated by TNI.
According to this concept, a benchmark study of the submitted data relevant to the validation was performed with SCALE 6. To this end BfS created a problem specific burn-up library for the PWR fuel assemblies with an enrichment level between four and five percent ^{235}U (as necessary for the application case) and a burn-up level of up to 90 GWd/tHM using the SCALE module TRITON, a two-dimensional transport and depletion code. The burn-up level has been chosen this high because the library files could also be employed in the source term evaluation for the shielding report. This library was then used to recalculate the irradiation of the Bugey and Gravelines samples. By comparing the results of these calculations with the experimental PIE data and taking into account all experimental and calculation uncertainties correction factors for the specific nuclides were determined.
In the next step the Bugey and Gravelines results were compared with the publically available PIE data. To simplify this process comprehensive studies of PIE data from the ARIANE and REBUS programmes [8] and the Calvert Cliffs, Takahama 3 and Three Mile Island reactors [9] were taken into account. Since [8, 9] use SCALE 5.1 calculations the Bugey and Gravelines samples as well as some of the other samples were recalculated applying SCALE 5.1. The results of these calculations were then completed by the results from the papers [8, 9] to estimate the uncertainty associated with the experiments. From this process new nuclide correction factors were calculated, now taking into account all experiments. These new nuclide correction factors then were the basis for the criticality safety assessment of the package.
In Figure 1 the differences between neutron multiplication factors calculated with SCALE for the TN 24 E package using different sets of nuclides and correction factors are presented. The burn-up calculations were done with the TRITON module for a burn-up of 12 GWd/tHM achieved in one reactor cycle, with a cooling time of two years. For the criticality calculations the module CSAS6 was used with a model of the cask for accident conditions of transport.

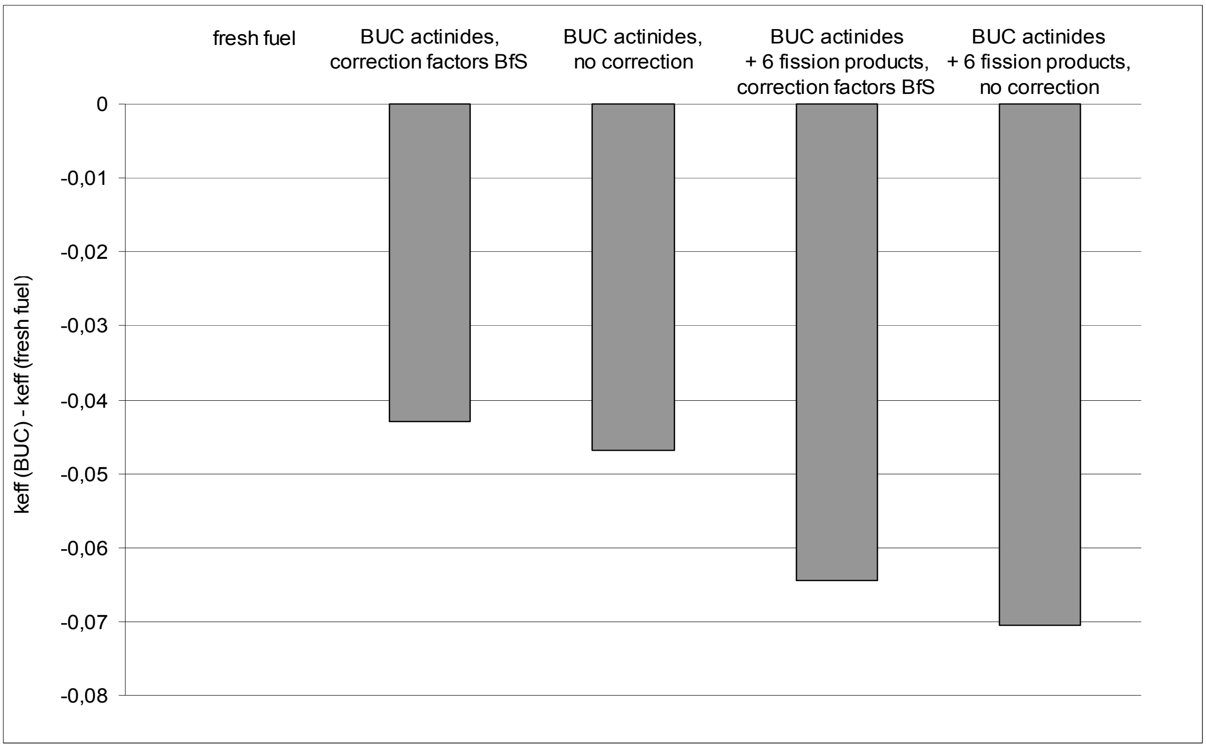

Figure 1. Differences in the TN 24 E neutron multiplication factors calculated for different sets of nuclides and nuclide correction factors

The Figure 1 shows that the main decrease in k_{eff} arises by including the changes in the actinide concentrations in the calculations. Nevertheless, adding fission products leads to an additional decrease of about 2% in k_{eff} for this package.

SUMMARY AND CONCLUSIONS

A validation of burn-up calculations for BUC including nine actinides and six fission products for the package TN 24 E by TNI has been approved by the German competent authority BfS. The assessment took into account proprietary PIE data from the French Bugey and Gravelines reactors as well as publicly available PIE data. BfS states that currently a validation of burn-up calculations for fission products based exclusively on the public data or exclusively on the French proprietary data is not possible.

Unfortunately, the use of public PIE data is limited by the still small number of such data including measurements of fission products and by incomplete descriptions of some experiments. The NEA WPNCS Expert Group on Assay Data of Spent Nuclear Fuel (ADSNF) has already realized the need to collect new PIE data and to get the PIE data sets reviewed by experts, and the group has already taken steps to prepare such review and to issue a new database containing more and reviewed PIE data. This is considered to be a very important development for improved use of BUC.

REFERENCES

[1] http://www.oecd-nea.org/science/wpncs/ADSNF/

[2] J. J. Lichtenwalter, S. M. Bowman, M. D. DeHart, C. M. Hopper, Criticality Benchmark Guide for Light-Water-Reactor Fuel in Transportation and Storage Packages, NUREG/CR-6361 ORNL/TM-13211, Oak Ridge National Laboratory, 1997

[3] International Handbook of Evaluated Criticality Safety Benchmark Experiments, NEA Nuclear Science Committee

[4] http://www.oecd-nea.org/science/wpncs/buc/

[5] Nicolas Leclaire, Tatiana Ivanova, Eric Létang, Emmanuel Girault, and Jean-François Throc, Fission Product Experimental Program: Validation and Computational Analysis, NUCLEAR SCIENCE AND ENGINEERING: 161, 188–215 (2009)

[6] F. Fernex, T. Ivanova, F. Bernard, and E. Létang, HTC Experimental Program: Validation and Calculational Analysis, NUCLEAR SCIENCE AND ENGINEERING: 162, 1–24 (2009)

[7] A. Santamarina et al., Experimental Validation of Burnup Credit Calculations by Reactivity Worth Measurements in the MINERVE Reactor, Proc. of the Int. Conf. on Nuclear Criticality ICNC '95, Albuquerque (USA), September 17-21, 1995

[8] G. Ilas, I. C. Gauld, and B. D. Murphy, Analysis of Experimental Data for High Burnup PWR Spent Fuel Isotopic Validation - ARIANE and REBUS Programs (UO2 Fuel), NUREG/CR-6969 (ORNL/TM-2008/072), prepared for the U.S. Nuclear Regulatory Commission by Oak Ridge National Laboratory, Oak Ridge, Tenn., February 2010

[9] G. Ilas, I. C. Gauld, F. C. Difilippo, and M. B. Emmett, Analysis of Experimental Data for High Burnup PWR Spent Fuel Isotopic Validation - Calvert Cliffs, Takahama, and Three Mile Island Reactors, NUREG/CR-6968 (ORNL/TM-2008/071), prepared for the U.S. Nuclear Regulatory Commission by Oak Ridge National Laboratory, Oak Ridge, Tenn., February 2010