NENE
International Conference
Nuclear Energy for New Europe

Calculation Method for determining Neutron-induced Nuclide Activities in nuclear Facilities

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ABSTRACT

Reliable knowledge of the distribution of nuclide activities in a nuclear facility at the time of decommissioning forms the basis for decommissioning scenarios, dismantling and disposal studies and corresponding safety analyses. As an alternative to fine-meshed radiological sampling, the distribution of neutron-induced nuclide activities can be determined cost-effectively by simulating the neutron flux distribution and its effect on the structural materials of the reactor building. This quasi-continuous and realistic information offers the opportunity to reduce conservatism, which not only leads to cost savings through computer-aided optimization of segmentation and packaging, but also to an optimized workload (minimization through exposure times). In this paper, we present modern calculation methods for corresponding simulations and discuss calculation methods and modeling decisions. In addition, we report on our experiences with the necessary preparations for high-quality modeling, i.e., the procurement of data such as geometry or material data of the structural materials. Furthermore, we show the procedure practiced at TÜV NORD EnSys for validating the calculation methods and models, with which the specified requirements for the quality of calculated values can be shown.

1 INTRODUCTION

The radiological characterization of a nuclear facility after the end of its lifetime [1], i.e., the determination of the amount and type of radioactive inventory and thus the amount and type of waste, is of high importance for decommissioning. It forms the basis for dismantling and disposal strategies, packaging planning, decontamination, radiation protection, identification of potential risks to humans and the environment, safety analyses, cost estimates, and estimation of the time required for decommissioning. While the boundary conditions to be considered and the procedures may vary in different countries, the focus is often on sampling and measurement procedures for irradiated components. In many cases, attempts are made to establish correlations between radionuclide activities based on the measurement results by the so-called scale factor method [2]. For this purpose, in simplified terms, the ratios of radionuclides to a key nuclide (often Co-60 and Cs-137) are formed. This approach, however, requires a large number of samples in order to make reliable statistical statements about the activities and their distribution. However, this is associated with high costs as well as long times and thus high total dose rates for personnel. In addition, there are areas in nuclear facilities that are difficult to access or where the working time must be kept very short due to excessive radiation exposure in order not to exceed the dose limits. For this reason, remote-controlled equipment often has to be used, which additionally causes a high expenditure of time and money.

TÜV NORD EnSys (TNE) is a German Technical Support Organization (TSO) that usually works on behalf of regulatory authorities and as such is involved, for example, in the decommissioning and dismantling processes of several German nuclear power plants. TNE work includes licensing, concept assessments, radiological measurements, on-site inspections, training and consulting.

Instead of performing radioactive characterization solely based on a large number of measurements, as mentioned in the approach above, one way to reduce costs and dose is to determine the nuclide activities of the components of a nuclear facility by ab initio calculations based on a significantly reduced number of measurements. This approach also offers advantages in the determination of nuclide correlations, since some radionuclides whose activities must be determined due to regulatory requirements can only be measured with great effort, but relatively easily be calculated with the aid of the computational codes. Together with a smaller number of samples and measurements, an accurate and fine-mesh determination of the total nuclide inventory and nuclide correlations is thus possible. In this article, we present the methodology and report on our experience with its use.

2 CALCULATION PROCEDURE

2.1 General Aspects

The calculation chain for determining the nuclide distribution is divided into several steps. The sequence of calculations is shown in Figure 1 and basically corresponds to the recommendation of ISO standard 16966 [3]. It starts with the elicitation of data that are important for the modeling of the nuclear installation. Gathering the necessary information for modelling is not a purely technical challenge. Rather, organizational issues play an essential role in making such a project a success. Even in the preliminary stages of the entire work it is necessary to know exactly what information is being sought, where it can be found at all and how it can be efficiently gathered from the often unstructured offer of information, interpreted and, if necessary, appropriately summarized. The organizational aspect would go beyond the scope of this article, therefore, the following presentation focuses on modeling-relevant activities and data.

2.2 Relevant Data for a Transport Model

Obtaining the correct data is essential to generate a good transport model. In our experience, the most successful way to obtain the most appropriate data to describe the real plant situation is to contact the relevant plant personnel directly. In this way, changes to the plant from the generic design or old approval status can be easily captured.

The modeling of the source is a crucial part of the generation of the transport model, since it determines the neutron flux densities to which, for example, the components surrounding the reactor core will be exposed. The first step is to determine how detailed the analyses need to be. In most cases, a steady state source is modeled for simplicity. The thermal power of a reactor determines the total number of neutrons produced per second in the core. The next step in modeling the source is to determine the spatial and energetic distribution of the neutrons over the spatial domain of the reactor core. This requires operational records such as burn-up profiles and power density distributions. The level of detail of a source model depends on the problem to be solved. For example, if the total neutron flux and the associated spectra along the core center plane in the reactor pressure vessel (RPV) are to be determined, a one-dimensional calculation with a cylindrical source, whose equivalent radius corresponds to the real core volume, will already give good results. However, for the calculation of neutron fluxes and spectra in the entire reactor building or even beyond, three-dimensional modeling is unavoidable.

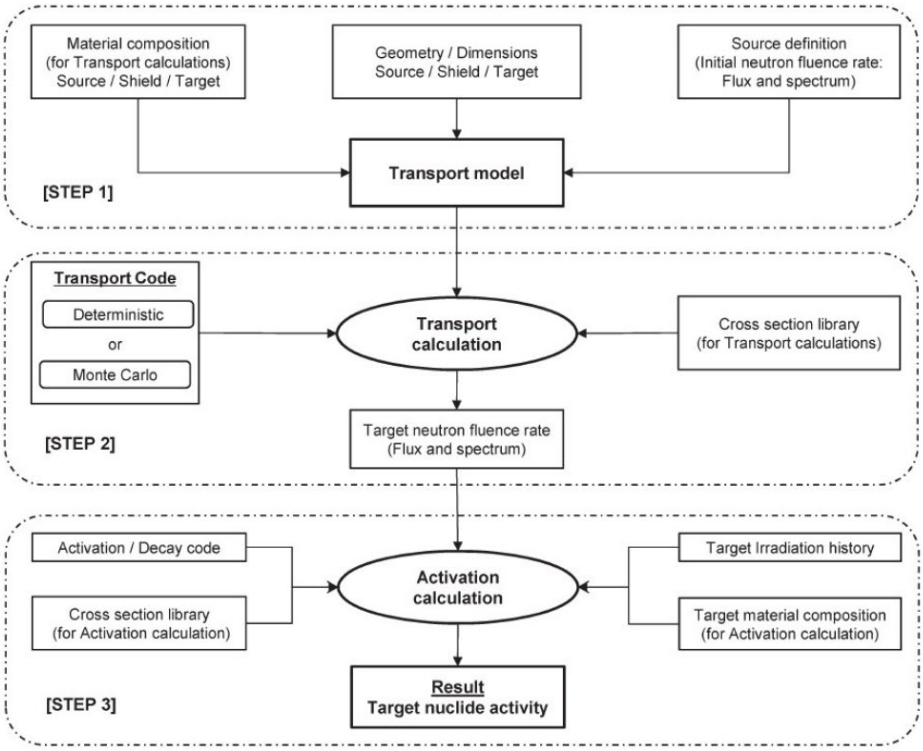

Figure 1: Flowchart of the calculation procedure (referred to [3]).

A very good and detailed approach is to model the source in fuel rod resolution. This approach is followed by TNE as far as possible. This was made possible by the fact that TNE's tasks included the evaluation of the core design calculations of several German reactors. Based on these calculations, which were performed with the core design codes CASMO/SIMULATE (Studsvik Scandpower (SSP)), it was possible to calculate the power density distributions of the individual fuel assemblies and fuel rods for any cycle points. An in-house software tool was used to determine the individual source strengths for 32 axial nodes of each fuel pin. The source strengths (neutrons per second) were averaged over one cycle. Test calculations have shown that the differences between using one cycle and considering individual cycles and multiple cycle states are small. However, the representativeness of the selected cycle(s) needs to be verified. Depending on the material composition of the fuel (proportions of U-235, Pu-239 and Pu-241), an average fission spectrum per node (superimposed Watt spectra) can be generated. For questions of activation outside the core, the first two fuel assembly rows usually play a role, since these are largely decisive for the activation of the peripheral structures. The remaining fuel assemblies have only a minor influence on the neutron flux densities. For this reason, the modeling of the core can also be limited to the first two to three radial fuel assembly rows and the first two to three lower and upper fuel assembly nodes. The material composition of the fuel does not play a major role. The proportion of fissile material can be set such that a neutron multiplication factor of approximately 1 is achieved in the criticality calculations.

The extent of the modeling depends on the calculation goals. For instance, if only the activities of components near the core (such as the core enclosure or the upper and lower core grid) need to be calculated, it is unnecessary to incorporate the structures outside the RPV. If, conversely, the activation of the RPV is of interest, including at least the nearby building structures, like the biological shield and the reactor pit, the accurate representation of the reflection of neutrons from the walls and the axial neutron flow in the interstices in the modeling is essential. For modeling the geometry, it is highly recommended to acquire data from construction drawings and specifications from the construction phase.

The level of detail in geometric modeling should always be assessed for potential impacts on the local neutron flux. Generally, the components within the RPV should be modeled with maximum detail, as they represent the greatest activities. Also, relevant components for radiological characterization should be modeled in enough detail to aid in subsequent dismantling and packaging planning. Inside and outside the RPV, it is essential to fully depict all flow paths in the model. This encompasses steam separators, pipelines, and corresponding wall openings, including insulation configuration, RPV insulation, the annular gap between the storage pool and reactor pit, and ceiling openings. In addition, material density changes should be appropriately addressed in the geometric model sections. This is particularly pertinent for moderator densities in a Boiling Water Reactor (BWR) and varied concrete compositions. In principle, the model should include strongly absorbing components such as the control assemblies. However, as the control assemblies are mostly inactive in a Pressurized Water Reactor (PWR) during operation, their consideration in the fully withdrawn state is enough. A similar logic applies to the BWR as well.

The consideration of trace elements or material impurities of the components is not relevant for the transport model, unlike for the activation calculations. It suffices to consider the main element of the material as the interactions during neutron transport are dominated by scattering processes. Trace elements, in this case, have a negligible influence. For concrete structures, it is crucial to consider the water content and air inclusions, commonly known as porosity, as they can significantly impact the moderation behavior of concrete. This, in turn, affects the neutron flux densities, spectra, and reflective behavior.

2.3 Transport Calculations

The neutron transport problem is a complex physical problem because of the strong heterogeneity that results from the combination of optically thin insulation layers with massive reinforced concrete shields and complex geometric structures. The problem is solved using transport codes. The solution methods utilized differentiate between two basic approaches to solving the transport problem: the deterministic and the stochastic (Monte Carlo methods) approach. We limit ourselves here to a small number of aspects for both methods, as this would otherwise exceed the scope of this article.

With a deterministic neutron transport code, the transport problem is solved for the average neutron behavior in a discretized model phase space. This space is defined by the position and angle coordinates as well as the energy range. By doing this, the transport problem can be mapped onto a set of algebraic equations, allowing for numerical calculation of neutron fluxes for discrete points, directions, and energy intervals. The solution obtained is accurate for the discretized model phase space. However, when deterministic solution methods are employed, an unfavorable choice of discretization can result in inaccurate neutron flux densities and spectra because of inadequate descriptions of effective cross-sections and geometry. Physically, the transition from optically dense to thin media (such as the cavity gap between the RPV and biological shield) is crucial for neutron transport. If the description of the angular variable is too low, it can lead to an inaccurate description of neutron streaming due to the ray effect. One advantage of deterministic transport codes is that they offer a global solution and are less computationally intensive compared to Monte Carlo codes. Examples of codes that use deterministic calculation methods include PARTISN (LANL), Denevo, SCALE/NEWT (ORNL), ATILLA (VAREX), and CASMO (SSP).

The Monte Carlo method is a numerical technique used to solve physical and mathematical problems through modeling random variables. Unlike deterministic methods, the method simulates the interactions that a particle experiences during transport processes. This enables computer calculations of the transport of nuclear radiation, including neutron and/or gamma radiation, which is produced during the operation of a nuclear facility. The procedure enables the modeling of large, intricate geometries with actual material compositions of reactor

and building components. Consequently, the mapping of gaps and penetrations in shielding building walls is ensured. In Monte Carlo codes, the necessary nuclear physics data are considered by the coupling with the nuclear libraries. In addition, it is not necessary to discretize the directional and energetic parameters of the simulated particle into groups, as is the case with deterministic methods. Instead, they can be kept as continuous state variables. Codes employing Monte Carlo methods are e.g., MCNP (LANL), KENO, MONACO, Shift (ORNL), TRIPOLI (CEA), GEANT (CERN), Serpent (VTT), and OpenMC (freeware). One disadvantage of Monte Carlo codes is their high computational time requirement, particularly for larger and more complex geometries with heterogeneous ray transport paths. It is important to note that Monte Carlo simulations provide expected values within a precisely quantified variance, rather than sharp physical values offered by deterministic methods. To reduce the variance by half (i.e., statistical error), the computation time must usually be increased by a factor of four.

To accelerate the process of variance reduction, hybrid methods have been developed, for example the ORNL code system SCALE/MAVRIC. Since TNE uses the MCNP and ADVANTG (ORNL) programs in tandem, we restrict ourselves to these options. The geometric models featured in MCNP, which are implemented to evaluate neutron flux densities, can be tremendously extensive. Therefore, neutron flux densities experience significant attenuation in outer regions, including the biological shield. To obtain low-uncertainty neutron flux densities in these areas, variance reduction methods are employed. Multiple variance reduction techniques are integrated into MCNP. Weight Windows (WW) is one such method utilized in MCNP, although its simplicity comes at the expense of inefficiency when dealing with intricate geometries and depends on user expertise.

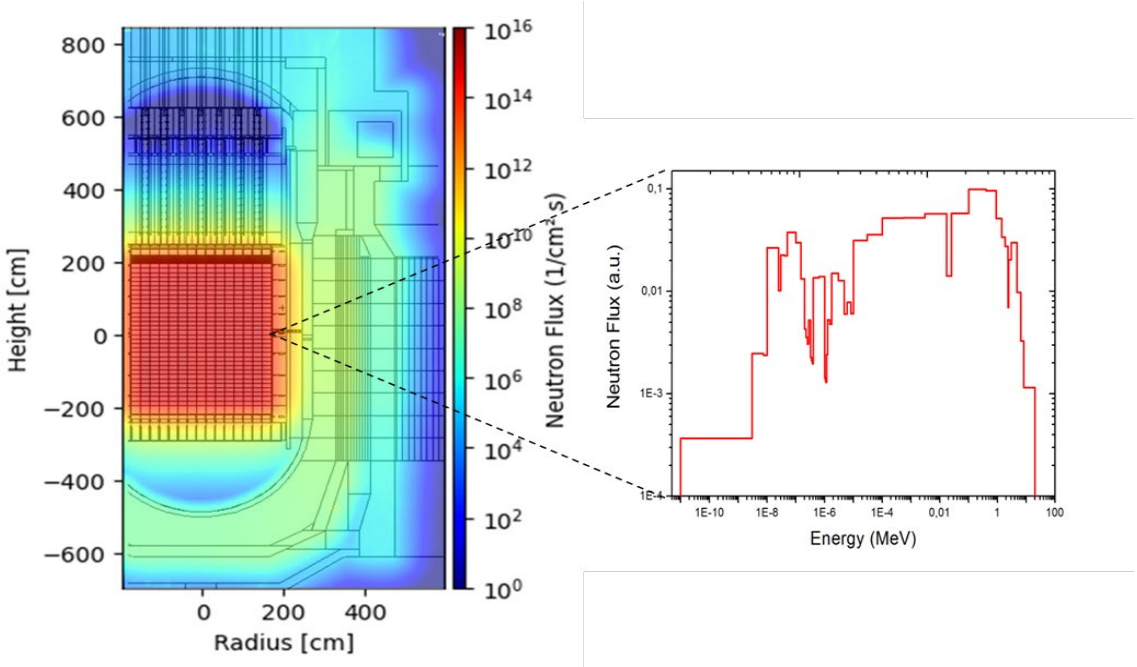

Figure 2: Mesh-Plot of the total neutron flux in the core center plane of a PWR and the corresponding neutron flux spectrum of the core shroud in a 56-energy group structure.

The software ADVANTG offers the capability to automatically produce an optimized weight window file. Essentially, weight windows enhance the simulation and assessment of neutrons in significant zones by increasing their numbers and decreasing them in non-critical zones while maintaining particle weight. ADVANTG was created to generate variance reduction parameters automatically for fixed-source MCNP simulations. ADVANTG generates a grid of location- and energy-dependent weight-window boundaries. The individual material volume fractions in each grid cell are determined by ray tracing.

Subsequently, the transport equations for each grid cell are solved using this approximation using deterministic methods. The result is a weight-window file for the actual MCNP problem. After achieving good convergence through the procedure, solutions for the total neutron flux density and spectra can be obtained. Figure 2 presents a mesh plot depicting the total neutron flux and the corresponding neutron flux spectrum of the core shroud in a 56-energy group structure for a PWR.

2.4 Activity Calculations

The neutron flux spectra determined in the transport calculation are needed to calculate the neutron activation activities. With these, it is possible to determine a spatially resolved inventory of nuclides for the spatial regions and components of interest for the respective irradiation time. A nuclide inventory code is generally used to calculate the nuclide compositions. The temporal evolution of the nuclide composition of a material can be described by the following system of linear ordinary differential equations:

$$\frac{dN_i}{dt} = \sum_j A_{ij} N_j \qquad \text{with} \qquad A_{ij} = \begin{cases} -\lambda_i - \sigma_i \phi, & i=j \\ b_{ij}\lambda_i + \sigma_{ij}\phi, & i \neq j \end{cases} \tag{1}$$

where A is the transition matrix. Here, λ_i is the decay constant of nuclide i, σ_{ij} is the energy-integrated microscopic cross section for a neutron-induced reaction of nuclide i to nuclide j and Φ is the integral neutron flux. The branching ratio b_{ij} denotes the fraction of the decay of nuclide i that leads into nuclide j. σ_i here means the sum of the cross sections of all neutron-induced reactions that convert one nuclide i into another nuclide j. In a multigroup representation, the energy-averaged neutron flux cross sections are given by

$$\phi(r,t) = \sum_g \phi_g(r,t)\,, \quad \text{and} \quad \sigma_{ij}(r,t) = \frac{1}{\phi(r,t)} \sum_g \sigma_{ij}^g(r,t)\phi_g(r,t)\,. \tag{2}$$

When calculating the nuclide inventory, it is standard practice to integrate over a specific spatial volume and assume that both the neutron flux and effective cross sections remain piecewise constant in time. This results in a linear system of equations with constant coefficients for determining the nuclide inventories. To conduct this calculation, we utilize the SCALE/ORIGEN (ORNL) program code. The following statements apply identically to other codes such as FISPACT II (UKAEA). The first step is to determine a 1-group cross section library for the spatial areas under consideration. For this purpose, the neutron flux spectra calculated for these areas are used. Therefore, the neutron flux spectra must be collapsed with the microscopic cross sections of all possible reactions which results in an approximation. This leads to one cross section value for the possible reactions. At TNE, we use the code SCALE/COUPLE (ORNL) for this purpose, to which the neutron flux spectrum must be transferred in a corresponding energy group structure. With these data the differential equations (1) for the nuclide concentrations can be solved numerically.

The next step is to specify the material composition of the irradiated material. The material composition should be known in detail. Our experience shows that in the construction phase, for example, only the material components with the largest proportions have been determined precisely via melt samples. In the case of trace elements, the information is often vague and can differ significantly from plant to plant [4]. Deviations in the proportions of trace elements can lead to a significant increase in the specific activities. Therefore, either new samples should be taken and analyzed during the transition phase when operation has ceased or conservative assumptions should be made regarding the trace elements that are decisive

for the activity inventory. The greatest uncertainties of the activation activities stem from the lack of knowledge of an exact material composition.

The last step is specifying the irradiation history of the activated material. Our experience shows that modeling every cycle according to its cycle and outage lengths is sufficient. A possible detailed modeling of the flux evolution during each cycle is laborious, but has practically no impact on the results, because the half-lives of the most important radionuclides are usually longer than the cycle lengths. The calculations can be performed for arbitrary reference times. Finally, in Figure 3 we give a scheme of the calculation procedure, the software codes, and libraries we use. Figure 4 shows an exemplary time course of some nuclide activities calculated with ORIGEN for steel of a core shroud in a PWR which was irradiated for approximately 37 years.

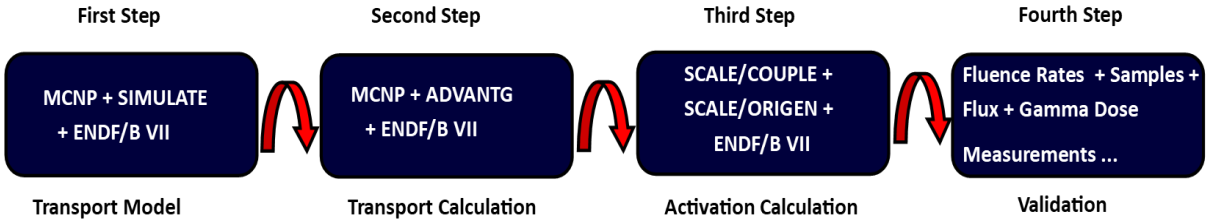

Figure 3: Scheme of the calculation procedure used by TNE to calculate activation activities.

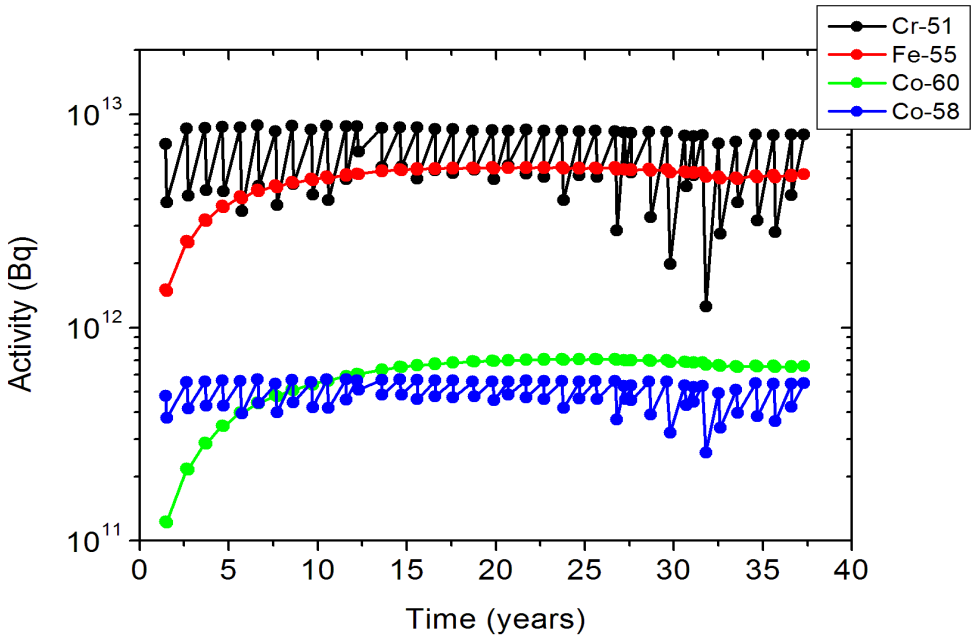

Figure 4: Time course of some nuclide activities calculated with ORIGEN for the steel of a core shroud in a PWR which was irradiated for approximately 37 years.

2.5 Validation

The activation calculations' results can be validated in various ways, including comparing the calculated activities and neutron fluxes with the measured ones. For instance, the activation of Fe-54 and Nb-93 samples placed in the RPV can be calculated and compared to the analysis results for RPV embrittlement monitoring. In Figure 4 the results of a deterministic calculation of the fast neutron flux performed by the manufacturer (reference) are shown. This was carried out in this context for a PWR. The reference results showed a maximum deviation of 3 % from the measured activities of the Fe-54 and Nb-93 samples. We compared these

results with those obtained from deterministic calculations and MCNP (figure 4). All results show a quite good agreement. This indicates a reasonable modelling of the PWR with MCNP.

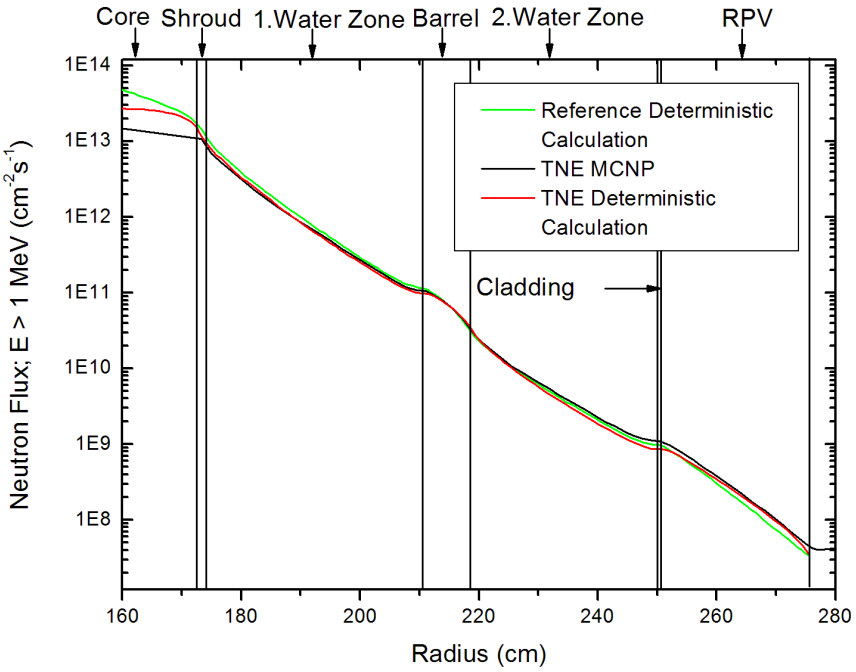

Figure 4: Comparison of fast neutron fluxes (E > 1 MeV) calculated with a deterministic code and MCNP.

Another option for validating calculated neutron fluxes involves comparing neutron fluxes measured via inner and outer power distribution detectors throughout operation. Once a nuclear facility has reached the end of its service life, samples can be obtained from the reactor pressure vessel internals or concrete structures for analysis. The resulting radionuclide activities can then be used to validate the calculated fluxes. In this process, TNE's task is to evaluate whether enough and representative samples have been analyzed and to oversee the collection of samples.

It is crucial to note that this is an iterative process where the measurements' findings and validation are incorporated into the calculation results for ongoing updates.

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