Modelling and Analysis of a (Th,Pu)O_2 Fuel Bundle Experiment in ZED-2 Using SCALE6 and TSUNAMI

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Abstract

TSUNAMI, a sensitivity and uncertainty analysis tool, was applied to study the intrinsic physics parameters of a (Th,Pu)O_2 fuel bundle experiment in ZED-2. This paper focuses on the investigation of moderator purity reactivity (MPR) by modelling the (Th,Pu)O_2 experiment in two cases with slightly different moderator purity. TSUNAMI analysis derived reaction-specific sensitivity coefficients since they must be taken into account in the calculation of k_{eff} and MPR uncertainties. Isotopes in the (Th,Pu)O_2 bundle were found to be the major uncertainty contributors in MPR and they have greater k_{eff} sensitivity responses in the less pure ZED-2 model.

1. Introduction

Between mid-1950s and 1970s, there were experiments conducted on thorium-based fuels in several types of reactors, part of the international endeavour in developing thorium-based fuel cycle [1]. In Canada, headed by the Atomic Energy of Canada Limited (AECL), there have been similar efforts in utilizing heavy water-moderated reactors to investigate the physics parameters of thorium-based fuels. Given the greater abundance of thorium (Th) in nature than uranium (U), thorium combined with plutonium (Pu) in particular has been found to show significant promises in many aspects. The (Th,Pu)O_2 fuel provides an attractive solution to incinerate weapon or civilian-grade plutonium. In contrast to (Th,Pu)O_2, there is no further production of plutonium associated with (Th,Pu)O_2. Instead, fertile ^{232}Th breeds fissile ^{233}U whose neutron yield is higher than that of ^{239}Pu/U type fuel in thermal reactors. (Th,Pu)O_2 fuel also provides some degree of proliferation-resistance due to the formation of ^{232}U which has strong gamma radiation daughter products [1].

2. Experiment description

The ZED-2 (Zero Energy Deuterium) critical facility operated by AECL put forward a number of experiments on a 36-element (Th,Pu)O_2 fuel bundle cooled by either air or light water. In this paper, only the experiment with a light water coolant will be discussed. Figure 1 shows the bundle cross section, where the 36 fuel pins surround a hollow central support tube with a slightly larger diameter.

Characterized by the critical heights of the heavy water moderator in the reactor, these critical experiments can be simulated using the KENO Monte Carlo code [2] from SCALE6 (Standardized Computer Analyses for Licensing Evaluation V.6). The full core ZED-2 models for these experiments include two types of fuel. First and foremost is the (Th,Pu)O_2 fuel bundle, located in the centre of the ZED-2 core. It is surrounded by 63 natural uranium metal ZEEP (Zero Energy Experimental Pile) rods, distributed in a non-uniform hexagonal array.

Five $(Th,Pu)O_2$ fuel bundles are stacked and fitted inside a single channel with aluminum pressure and calandria tubes, separated by an air gap. Both types of fuel bundles are elevated by 15 cm from the bottom of the calandria tank, which is also the zero reference point for the moderator height. The control of reactivity in ZED-2 is achieved by adjusting the vertical height of the moderator. Therefore, fuel bundles are partially covered by heavy water at criticality. Similarly, the shutdown of ZED-2 relies on fast dumping of the moderator from the dump lines situated below the calandria tank. ZED-2 is also equipped with a graphite reflector, which should be included in the full core model to better represent the true configuration of the reactor.

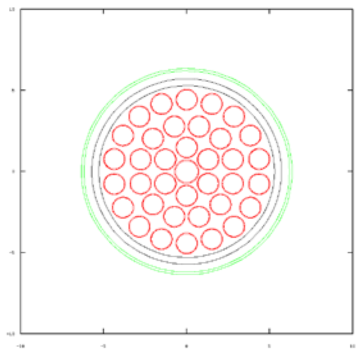

Figure 1 Cross section of the experimental $(Th,Pu)O_2$ fuel bundle.

3. SCALE6 simulation

Developed at ORNL (Oak Ridge National Laboratories), the SCALE6 code package contains a criticality safety tool called KENO [2]. It uses Monte Carlo method to determine the value of k_{eff} for complex systems. The ZED-2 reactor was modelled fully in three dimensions and to scale. But a few simplifications in certain bundle components were made. The nuclear cross-section library v7-238 was used. It contains all ENDF/B-VII.0 nuclide data in a 238-group structure from 10^{-5} eV to 2×10^{7} eV [3]. In order to create problem-dependent, group-wise cross section data for KENO, SCALE6 uses material processing modules on user-defined geometry and unit cell specification data. The method of *MULTIREGION* [4] was used throughout all models for resonance self-shielding calculations. KENO also performs the calculation of forward and adjoint neutron flux moments, crucial for sensitivity coefficient computation and uncertainty analysis.

4. TSUNAMI sequence

The TSUNAMI sequence starts with TSUNAMI-3D. It uses KENO's calculations on neutron fluxes, k_{eff} values and feeds them into SAMS (Sensitivity Analysis Module for SCALE). Sensitivity coefficients and the uncertainty in k_{eff} are computed by SAMS using the adjoint-based first-order linear perturbation theory [5]. Sensitivities from TSUNAMI-3D can be used in three different modules: TSAR, TSUNAMI-IP and TSURFER. Figure 2 illustrates typical workflow of the TSUNAMI analysis.

TSAR (Tools for Sensitivity Analysis of Reactivity) studies reactivity responses such as coolant void, fuel temperature or moderator purity change. TSUNAMI-IP generates similarity indices for neutronic similarity of two or more systems. TSURFER (Tool for Sensitivity and Uncertainty analysis of Response Functions using Experimental Results) applies the Generalized Linear Least Squares Method (GLLSM) for nuclear data adjustment [6].

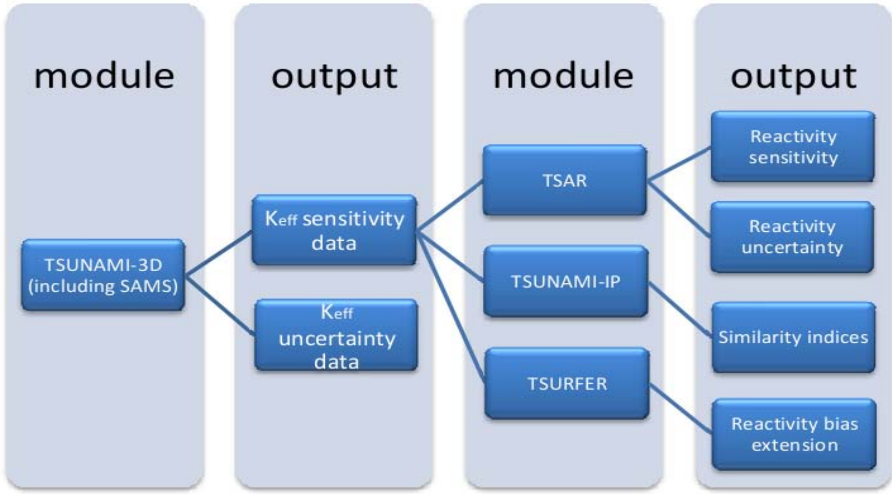

Figure 2 TSUNAMI workflow sequence shows the relevant modules and their outputs.

As mentioned above, the methodology of sensitivity and uncertainty analysis is derived from the adjoint-based first-order linear perturbation theory. First the cross section nuclear data (Σ) are perturbed such that the system's k_{eff} deviates from its original value. The sensitivity coefficients associated with k_{eff} ($S_{k,\Sigma}$) can be expressed in equation (1) as the most simplified form:

$$S_{k,\Sigma} = \frac{\Sigma}{k}\frac{\partial k}{\partial \Sigma} \tag{1}$$

where ∂k and $\partial\Sigma$ are deviations from the unperturbed k_{eff} and Σ respectively. It should be noted that $S_{k,\Sigma}$ is a sum of explicit and implicit effects due to the perturbations. The meaning of $S_{k,\Sigma}$ essentially reflects how k_{eff} changes in response to an increase or decrease in nuclide contributions in the system.

Sensitivity coefficients are also used to determine the propagated effect of nuclear cross-section uncertainties into k_{eff} uncertainty. Equation (2) shows that the k_{eff} variance (σ_k^2) is defined as a product of sensitivity coefficient vectors (S_k and S_k^T) and the covariance matrix $C_{\alpha\alpha}$ containing nuclear data uncertainties:

$$\sigma_k^2 = S_k C_{\alpha\alpha} S_k^T \tag{2}$$

where S_k^T is the transpose of sensitivity coefficient vector S_k. The significance of the covariance matrix is present throughout the TSUNAMI analysis in that, when two or more systems are compared, they must still share the same nuclear data uncertainties, even though their isotopic sensitivities can be drastically different.

5. Preliminary TSUNAMI results

To illustrate how TSUNAMI can be applied to the (Th,Pu)O_2 fuel bundle experiment in ZED-2, two models were constructed and their TSUNAMI results will be described in the following sections. The first model (case 1) is based on an AECL internal report on the (Th,Pu)O_2 fuel bundle experiment, which achieved criticality with a moderator purity of 99.495 wt%. The second model (case 2) shares the same geometry, materials and moderator critical height of the first model, however having an idealized moderator purity of 100 wt%. These two cases can shed light on the isotopic sensitivity responses of the (Th,Pu)O_2 bundle at different moderator purity levels and help us understand moderator purity reactivity (MPR) of the bundle.

5.1 k_{eff} sensitivity and uncertainty

In order to investigate the sensitivities of particular isotopes in different moderator purity conditions beyond the global system behaviour characterized by k_{eff}, the following k_{eff} sensitivity coefficient profiles were examined.

First of all, it is obvious that the ability of ^{2}H (elastic) scattering is dependent on the moderator purity. Hence, as shown in Figure 3, ^{2}H (elastic) has a positive sensitivity response which increases with the moderator purity level. It is also a plausible explanation for why the case of higher moderator purity (case 2) has a more negative sensitivity of ^{238}U (n,γ) in the thermal range in Figure 4. It is resulted from a larger number of thermalized neutrons in case 2 and more neutrons are lost in the ^{238}U thermal absorption.

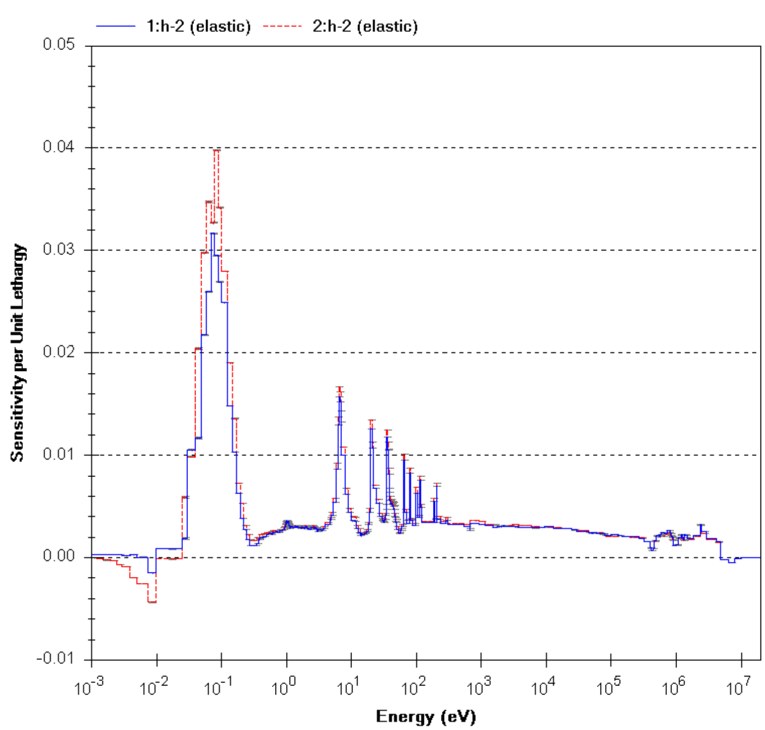

Figure 3 Sensitivity profiles of ^{2}H (elastic) where 1 (blue) corresponds to moderator purity 99.495 wt% and 2 (red) corresponds to moderator purity 100 wt%.

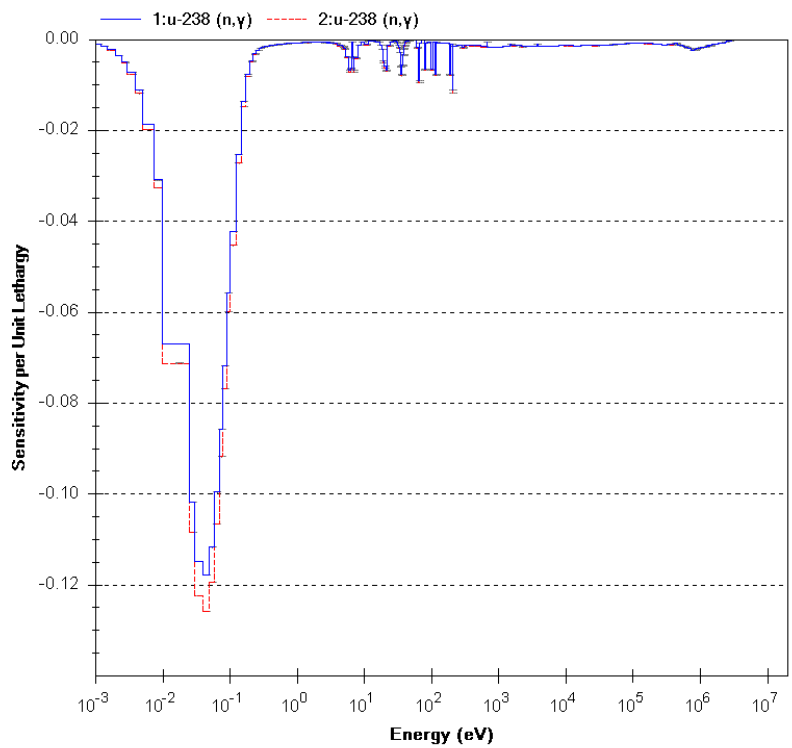

Figure 4 Sensitivity profiles of ^{238}U (n,γ) where 1 (blue) corresponds to moderator purity 99.495wt% and 2 (red) corresponds to moderator purity 100 wt%.

For this sensitivity analysis, the focus is fixed upon the isotopes in the $(Th,Pu)O_2$ bundle. Figure 5 and Figure 6 are the sensitivity profiles for ^{239}Pu (fission) and ^{232}Th (n,γ) respectively, revealing the case of 99.495 wt% moderator purity (case 1) has larger sensitivity responses in magnitude for both isotopes. After examining the fission density by locations, it is found that when the moderator purity is improved (from case 1 to case 2), fission density in the $(Th,Pu)O_2$ bundle is reduced, manifesting in the decrease of ^{239}Pu (fission) sensitivities. ^{232}Th (n,γ) sensitivities are also reduced when the moderator purity is increased. These phenomena might be explained by the more significant neutron thermalization in the large volume where ZEEP rods reside (case 2), which gives less opportunity for neutrons to reach the $(Th,Pu)O_2$ bundle imbedded in the centre of the reactor core. This is analogous to the self-shielding effect on the scale of fuel channels, and a likely reason for experiencing lower neutron fluxes and consequently smaller sensitivities in the $(Th,Pu)O_2$ bundle. In contrast, ZEEP rods have higher fission densities in the case of better moderator purity. Therefore the system k_{eff} was able to increase with moderator purity as expected.

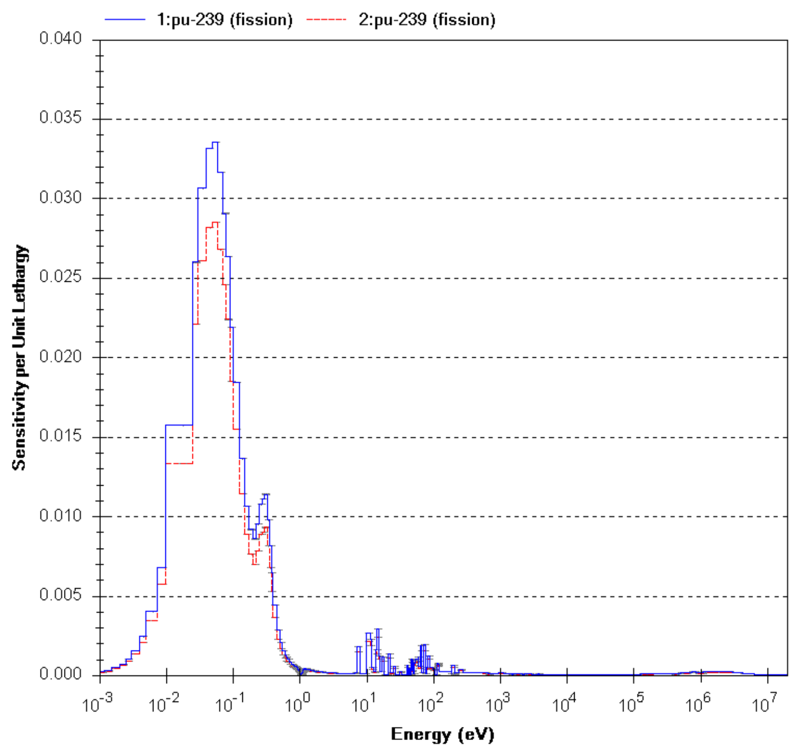

Figure 5 Sensitivity profiles of ^{239}Pu (fission) where 1 (blue) corresponds to moderator purity 99.495wt% and 2 (red) corresponds to moderator purity 100 wt%.

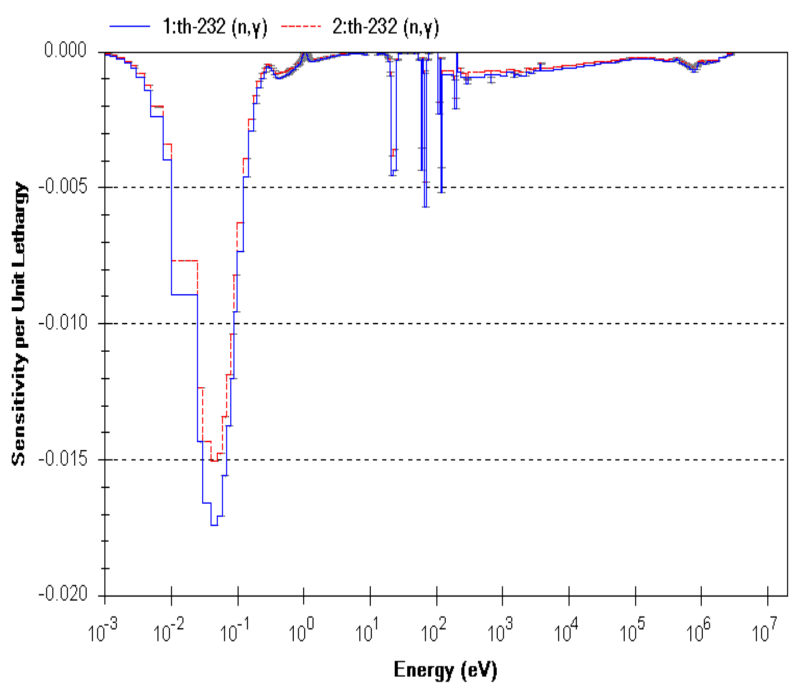

Figure 6 Sensitivity profiles of ^{232}Th (n,γ) where 1 (blue) corresponds to moderator purity 99.495wt% and 2 (red) corresponds to moderator purity 100 wt%.

In addition to generating reaction-specific group-wise sensitivity coefficients, the SAMS module in TSUNAMI-3D is also responsible for computing k_{eff} variance(σ_k^2) due to nuclear data covariances using the sensitivity data as shown in equation (2). k_{eff} standard deviation (σ_k) can be further broken up into reaction-specific components (σ_i), which are called the uncertainty contributors. For example, Tables 1 and 2 show the k_{eff} standard deviation and the top three uncertainty contributors for the two ZED-2 models respectively. However, it is important to realize because the uncertainty takes into account the nuclear data covariances, reactions with large uncertainties may not correspond to high sensitivities, and vice versa.

$\sigma_k = 6.10$ mk		
Nuclide reaction	σ_i (mk)	σ_i^2/σ_k^2
^{238}U(n,γ)	4.41	52.16%
^{235}U - $\bar{\nu}$	2.38	15.16%
^{2}H (n,2n)	2.09	11.71%

Table 1 Top k_{eff} uncertainty contributors in the case of 99.495 wt% moderator purity.

$\sigma_k = 6.71$ mk		
Nuclide reaction	σ_i (mk)	σ_i^2/σ_k^2
^{238}U(n,γ)	4.98	55.19%
^{235}U - $\bar{\nu}$	2.60	15.03%
^{2}H (n,2n)	2.27	11.51%

Table 2 Top k_{eff} uncertainty contributors in the case of 100 wt% moderator purity.

5.2 Reactivity response

The effect of moderator purity was manifested in the sensitivity responses of several isotopes discussed above. To quantify such an effect, we can define the moderator purity coefficient of reactivity (MPR) in equation (3) where Δwt% D_2O is the difference in moderator purity percentage, $k_{eff,1}$ and $k_{eff,2}$ represent the k_{eff} values for 99.495 wt% (case 1) and 100 wt% (case 2) moderator purity respectively. The reactivity difference ($\rho_{1\to2}$) between the two models is 61.3 mk. Knowing that the value of Δwt% D_2O is 0.505%, we obtain a MPR value of 121.4mk per wt% purity.

$$\mathrm{MPR} = \frac{1/\mathrm{k}_{\mathrm{eff},1} - 1/\mathrm{k}_{\mathrm{eff},2}}{\Delta\mathrm{wt\%D_2O}} = \frac{\rho_{1\to2}}{\Delta\mathrm{wt\%D_2O}} \tag{3}$$

The TSAR module performs sensitivity calculation for MPR using a modified sensitivity definition. Compared to the k_{eff} sensitivity, MPR sensitivity coefficients ($S_{\rho,\Sigma}$) take into account the reactivity change:

$$S_{\rho,\Sigma} = \frac{\Sigma}{\rho_{1\rightarrow 2}}\left[\frac{\partial\lambda_1}{\partial\Sigma} - \frac{\partial\lambda_2}{\partial\Sigma}\right] \tag{4}$$

where λ is equal to 1/k. With the MPR sensitivity coefficients, the MPR variance due to nuclear data uncertainties can be computed the same way as k_{eff} variance. The MPR uncertainty (σ_ρ) and several important top uncertainty (σ_i) contributors are summarized in Table 3. It is clear that the uncertainty contribution from ^{1}H (n,γ) is a direct result of the moderator impurity in case 1. Isotopes ^{239}Pu and ^{232}Th from the (Th,Pu)O_2 bundle also play important roles in the MPR uncertainty contribution because of their sensitivity variations, discussed in the previous section.

σ_ρ = 0.544 mk		
Nuclide-reaction	σ_i (mk)	σ_i^2/σ_ρ^2
^{239}Pu - $\bar{\nu}$	0.319	55.19%
^1H (n,γ)	0.315	33.5%
^{239}Pu(fission)	0.169	5.25%
^{232}Th(n,γ)	0.098	3.24%
^{235}U(fission)	0.092	2.86%

Table 3 A list of important top MPR uncertainty contributors.

5.3 TSUNAMI-IP

The similarity between the two ZED-2 models can be described by two indices primarily, calculated using the TSUNAMI-IP module. Similarity index c_k is an integral measure based on the shared k_{eff} uncertainties due to cross section covariance data for the two models. The second similarity index G is a combination of the similarity for fission, capture and scattering processes. Both indices can have values from -1 to 1. In our case, TSUNAMI-IP was able to generate c_k and G values which are very close to 1 as shown in Table 4, indicating the two models have high similarity. The index G_{capture} has the lowest similarity value, emphasizing the moderator impurity by ^{1}H (n,γ) is an important factor for criticality.

c_k	G	$G_{fission}$	$G_{capture}$	$G_{scatter}$
0.997	0.9198	0.9698	0.8636	0.9322

Table 4 Similarity indices generated by TSUNAMI-IP for the two ZED-2 models with different moderator purity.

5.4 Future Work on TSURFER

TSURFER, the third module in the TSUNAMI sequence (Figure 2), involves intensive mathematical calculations in order to reduce the bias in the computed $k_{eff.}$ It uses the method of Generalized Linear Least Squares to determine a set of nuclear data which would generate a computed k_{eff} value very close to it experimental value. The accuracy of our models highly depends on what we know about the possible uncertainty sources. They are likely to originate from assumptions made in the KENO geometry and material specifications, as well as numerical errors in computers. KENO model uncertainties can be minimized by acquiring and including more experimental details. However, numerical uncertainties are inevitable and must be tolerated. Perhaps the most important type of uncertainty comes from the nuclear data in the ENDF libraries, because of its inherent presence in all reactor physics models. Therefore, in order to tackle the bias in the computed k_{eff} or other physics parameters (for example, coolant void reactivity), TSURFER will be applied to the ZED-2 models to make intelligent adjustments on the nuclear data, aiming to constrain uncertainties leading to large biases.

6. Conclusion

Sensitivity and uncertainty analysis was performed for an experiment of $(Th,Pu)O_2$ fuel bundle cooled by light water in the ZED-2 reactor. Two KENO models were constructed and they only differ in the moderator purity levels: 99.495 wt% and 100 wt%. The purpose of the latter was to explore moderator purity reactivity effects on important isotopes such as ^{239}Pu and ^{232}Th. The two models were analyzed using the TSUNAMI-3D, TSAR and TSUNAMI-IP modules from TSUNAMI. It was observed that the $(Th,Pu)O_2$ bundle was able to fission more in the model with less pure moderator. While ^{238}U (n,γ) contributes the most uncertainty to the k_{eff} uncertainties in both cases, the top uncertainty contributors to the moderator purity reactivity belong to isotopes in the $(Th,Pu)O_2$ bundle. It was also determined that there was a high degree of similarity between the two models, indicated by similarity indices c_k of 0.997 and G of 0.9198. Forthcoming works include improvement on the models' accuracy using the TSURFER module, which will provide more insights on the nuclear data uncertainties involved in the calculation of k_{eff} or other physics parameters.

7. Acknowledgements

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