On the effect of Resonance dependent Scattering-kernel on Fuel cycle and inventory

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Abstract

The original double differential scattering kernel developed by Rothenstein and Dagan for heavy isotopes differs from the current MCNP treatment by the inclusion of the strong scattering resonances for the secondary energy distribution. In particular the scattering kernel of U238 is strongly dependent on the cross section profile in the vicinity of resonances, which leads to noticeable changes in its absorption rate. Consequently the Plutonium production as well as its consumption during the fuel cycle is expected to be changed. In return the burn-up dependent criticality level will be changed too. In addition, the inventory at the end of the cycle of other isotopes, and in particular of the minor actinides is expected to be different.
The new mathematical formalism by Rothenstein enabled the implementation of the modified kernel within the THERMR module of NJOY. After additional modifications a set of energy probability ($S(a,b)$ **)** tables was prepared for U238 in the same manner as it is done for light isotopes. The replacement of the approximated existing scattering kernel treatment in MCNP by the new tables allows practically to improve the assessment of the fuel cycle behaviour and the burned up dependent fuel inventory. Such a quantified evaluation in criticality and in material compositions, using the new probability tables for U238, is demonstrated by means of two selected PWR fuel pin benchmarks.

KEYWORDS: Double Differential Scattering, $S(a,b)$**, resonant absorption**

1. Introduction

The correct solution for slowing down of neutrons is essential for thermal core calculations. At the lower epithermal range, the scattering collision between the neutron and its target is elastic, as the first excited level of the light and heavy scattering isotopes is several KeV. For light isotopes with nearly constant scattering cross section two options are commonly used. The free gas model and the bound isotope approach which takes into account the vibrations of the atom in its molecule in the thermal energy range. For the free gas kernel the double differential scattering kernel including the energy after scattering as well as the cosine of the scattering angle is well known. This equation was obtained directly in a classical way by Rothenstein [1] on the basis of momentum and energy considerations. It was also derived as a simplified case of a quantum mechanical treatment (M.M.R. Williams [5]) where the excited target levels are degenerated and the nuclei are reacting as an ideal gas.
The treatment of bound light isotopes like Hydrogen in water is based on a different approach dealing with the motion of atom its mutual energy transfer with its molecule. This data is then

processed by a module of the NJOY code [2], which forms the scattering kernel into probability tables, called also $S(a,b)$ tables. Those tables are then read by the MCNP[4] code.

The treatment of heavy isotopes, in particular U238 should include the fact that the cross section is **definitely** not constant in the vicinity of the resonances. Yet, the NJOY [2] procedure in its THERMR module is based on light isotopes $S(a,b)$ treatment for all materials. The cumulative kernel over all angles is then normalized according to the scattering cross-section based on the BROADR module in NJOY. For heavy isotopes with pronounced resonances this is not quite accurate as: "The secondary energy distribution will be still incorrect" (THERMR manual [2]).

The full double differential kernel developed by Rothenstein & Dagan [1] includes the influence of the pronounced resonances on the scattering and subsequently on the absorption rate and it is consistent with the broadened scattering cross section calculated by BROADR. The new mathematical approach by Rothenstein [3] reduced the calculation time considerably and provided the way in which NJOY can be improved making use of the developments [1] which was not available when the original models were included in NJOY. This new "Rothenstein" scattering kernel treatment enabled finally the generation of probability tables for heavy isotopes in the same manner as it is done for light isotopes. Consequently the approximated treatment of MCNP is replaced.

As the improved kernel enhances the resonant absorption in U238, the Pu239 concentration is expected to grow proportionally along during the first stage of the fuel cycle. Moreover the simulated inventory of other actinides is going to be affected by the introduction of the new kernel.

In the next sections the new double differential scattering kernel for heavy isotopes is introduced.

Followed are then two types of PWR Benchmarks: a PWR fuel pin at 1200K and a PWR subassembly at 800K. The quantified changes in the heavy isotopes composition due to the implementation of the resonant dependent cross sections are presented.

2. The energy dependent double differential scattering kernel

The inconsistency between the "classical" scattering kernel in THERMR module of NJOY and its "integral value" namely the broadened scattering cross sections in the vicinity of the resonances as given by the BROADR module strengthened the idea of finding an equivalent solution for the scattering kernel which includes the pronounced resonances. Consequently the underlying assumptions exclude the chemical binding effects in the mathematical considerations. This assumption is physically well founded as long as the fuel temperature is high enough above the Debye temperature, which is a measure of the maximal free vibration frequency of the atom in its solid structure. The Debye temperature for oxide uranium is about 500^0 K and for metallic uranium is less than 200^0 K.

Thereafter, for most practical core applications and for the Benchmarks investigated in this work the new kernel is relevant.

The detailed derivation of the following equation (2.1) can be found in Rothenstein [3].
The scattering kernel for isotopes with strong scattering resonances is:

$$
\begin{aligned}
s_s^T(E \to E', \vec{\Omega}, \vec{\Omega}') = \frac{1}{4p\,E}\sqrt{\frac{A+1}{Ap}} \int_{e_{\max}}^{\infty} dx \int_{t_0(x)}^{t_1(x)} dt \left(\frac{x+t}{2}\right) \\
\times\left(s_s^{tab}\left[\left(\frac{A+1}{A^2}\right)\frac{(x+t)^2}{4}k_B T, 0\right]\right) \\
\times\exp\left(v^2 - \left[\frac{(x+t)^2}{4A} + \frac{(x-t)^2}{4}\right]\right) \\
\times\left(\frac{e_{\max} e_{\min}(x-t)^2}{B_0 \sin\hat{j}}\right)
\end{aligned}
\qquad 2.1
$$

where the integration variables (x , t) are introduced by rotating the velocity plane of integration of the original variables (t , x), where: $t = u\sqrt{(A+1)}$; $x = c\sqrt{(A+1)}$. 'u' and 'c' are the velocity of the neutron in the centre of mass (C.O.M.) system and the velocity of the C.O.M. respectively.

$e_{\max}$, $e_{\min}$ are the values given to the minimal and maximal following velocity terms $e = v\sqrt{(A+1)}$; $e = v'\sqrt{(A+1)}$ (see also [3]). $\vec{n}$,$\vec{n}'$ are the neutron velocity before and after scattering in the laboratory frame of reference.

This kind of manipulation of the velocities vectors into new velocity variables being used over new reduced integration plane enabled to speed up the generation of the scattering probabilities for the out going Energy and angular direction using high resolution energy point solver- up to 16384 energy intervals for the outer integration in Eq. 2.1. For this study 8192 points were used for each calculation of Eq. 2.1 which gives also very good consistency with the BROADR Doppler broadened scattering cross section. For each temperature 950 probability tables were generated in the range up to 210 eV covering the 8 main S resonances of U238. Those tables were then added to the MCNP data libraries. By introducing "mt" cards for U238 within the MCNP input file the scattering probabilities are being unbiased sampled from the $S(a,b)$ tables similar to the procedure done for light isotopes. In this way the solver of the COLIDN subroutine within the MCNP code, which uses the Rejection Technique (Ross [9]) for heavy isotopes is bypassed. This Technique might cause biasing in the vicinity of the resonances. From a physical point of view, this approach is "equivalent" to the delta function velocity constraints which is the basic for development of the classical free gas, constant cross section, scattering kernel model mentioned above.

3. Fuel pin and S/A burn up benchmark calculations

The changes in the burned up dependent fuel cycle criticality and isotopes inventory, due to the implementation of the "Rothenstein" scattering kernel, were evaluated for the two following benchmark problems

3.1 **"Rowlands" Benchmark for PWR fuel pin**

The geometry and material composition of the PWR fuel pin defined by Rowlands [6] is shown in table 1 and table 2 respectively. The enrichment is 3.1 % of U235. A suitable MCNP input was prepared. Two type of burn up calculations were compared. The first with the normal treatment of MCNP and in the second input the "mt" card for U238 was introduced, making use of the new scattering kernel and bypassing the "sampling of the target velocity" approach (MCNP/Chapter 2) in MCNP. The MCNP runs were integrated in the Burn up code System MONTEBURNS [7] using ENDF6.5 data for the Uranium vector, ENDF6.0 for the Plutonium vector and ENDF5 data for the other isotopes. The temperature of the Uranium vector for the current scattering kernel (by using "tmp" cards in the MCNP input) and the improved treatment was at 1200K.

In Figure 1 the criticality level of the reference case with the normal treatment of MCNP is shown for the 15 burn up steps each of which is about 3.8 MWD/Kg. The standard deviation of the criticality value for each calculation in this figure and in the results shown up to figure 4 was 1×10^{-4}. Figure 2 shows the criticality differences due to the introduction of the new kernel in comparison to the reference solution shown in figure 1. At the beginning of the fuel cycle the criticality is about 440 pcm lower and after about 51 MWD/Kg is 200 pcm higher than the reference case which implies a change of 660 pcm over the whole cycle. This result is a corollary of the change in the isotopic density which can be seen in Figure 3. The fact that the new scattering kernel "allows" for more neutrons to be absorbed in the resonances of U238 means lower K_{eff} at the early stage of the fuel cycle. However, in return more PU239 is produced. Through the burn up cycle the increase in the PU 239, and to some extent the PU241 and other fissionable materials, the pin cell is actually more efficiently "breeding" so that the criticality level is steadily increasing up to about 51 MWD/kg burn up.

Figure 4 presents the relative inventory change in grams for a 3.9 meter long fuel pin between the reference and the calculation with the mt card for U238. It can be seen that at about 45 MWD/kg the calculated change in the amount of Pu239 is steady while the U235 calculated differences is decreasing. This is a consequence to the growing importance of the PU239 as the main fissile material at this high burn up stage compared to the reduced importance and amount of U235, which its total weight is only a third of the PU239 .

Pin cell Benchmark	PWR UOX
Fuel radius	0.4 cm
Cladding outer radius	0.45 cm
Equivalent cell radius	0.67703 cm

Table 1: Geometrical dimensions of the pin cell

Isotope	Zone	Density atoms/b-cm
U_{235}	**Fuel**	**0.00070803**
U_{238}	**Fuel**	**0.022604**
O_{16}	**Fuel**	**0.046624**
Zr	**Clad**	**0.043241**
H	**Moderator**	**0.066988**
O	**Moderator**	**0.033414**

Table 2: particle densities of the PWR unit cell benchmark

3.2 GRS fuel Sub-assembly Benchmark

The GRS Sub-assembly - S/A [8] is an 18 by 18 matrix with 4% w/o U235 enrichment including 24 water holes. The geometry of the each cell and the isotopes densities are defined in table 3 and 4 respectively. As a preliminary test a pin cell calculation was performed in a similar manner as the Rowlands cell was analyzed and at the same temperature (1200K). However the calculation were done with the burn-up version of MCNPX [10] using data based upon [1]. The overall trend was similar to the above, namely the change in the criticality level was between -500 pcm and +170 pcm after 50 MWD/kg. Based on this result a fuel subassembly based on this fuel pin composition was analyzed. The procedure is similar to the pin cell calculation. All fuel pins are considered as one fuel material in the MCNP input so that the additional differences in the fuel consumption due to heterogeneity effects in the vicinity of the water holes "smeared". The fuel temperature was changed to 800K through out the whole calculated fuel cycle. The boron concentration is kept constant, so the differences shown are directly attributed to the different scattering kernel approach for U238.
Thereafter the results presented below at relative lower temperature and including "smoothed" water holes heterogeneity effects provides the minimal deviations caused by the current approximated MCNP solver and it is expected that in reality for pin by pin calculations the shown differences in figures 6-8 would be larger.
Figure 5-8 are similar to figure 1-4 respectively. The figures have also the same range so the changes due to the different temperature can be more visible. The overall change in criticality 410 pcm is as expected lower at 800K. The inventory for the main isotopes is also in accordance with the pin cell calculation, namely all changes are mitigated as the importance of the correct scattering kernel is growing with the fuel temperature. On the average the growth in the PU239 inventory is 44.3 grams for the GRS Benchmark and for such a S/A (figure 9) based upon the Rowlands pin cell benchmark would be about 63 grams. The accuracy of the MCNP calculations in this case was 2.2×10^{-4} which could be partially the explanation for the swings in Fig. 6. This type of local uncertainties was beyond of the scope of the current study.

S/A Benchmark	PWR UOX
Fuel radius	0.411 cm
Cladding outer radius	0.475 cm
cell pitch	1.1448 cm

Table 1: Geometrical dimensions of a fuel pin in the S/A

Isotope	Zone	Density atoms/b-cm
U_{235}	**Fuel**	**0.00088802**
U_{238}	**Fuel**	**0.0210349**
O	**Fuel**	**0.043864**
Zr	**Clad**	**0.05104**
H	**Moderator**	**0.05000**
O	**Moderator**	**0.025002**
B	**Moderator**	**0.0000208**

Table 2: particle densities of the pin cell for the PWR S/A benchmark

5. Conclusions and outlook

The success of introducing the full double differential scattering kernel for Heavy isotopes with pronounced resonances within the MCNP code enables to evaluate better the criticality as well as the isotopes consumption during the burn up cycle. In particular for the 8 first resonances of U238 it was shown that for LWR fuel type the change over a typical subassembly is of importance concerning the optimization of the fuel cycle and the fuel inventory. More detailed calculation should be performed to analyze peak powers in pins in the vicinity of the water holes and the influence of other heterogeneous effects on core parameters, which could concern the safety margins of the fuel elements design concept. Furthermore the additional production of PU and other actinides increases to some extent the helium production and the *a* ray release leading to deviation in the assumed real integrity stage of the fuel pins. As at higher burn-up more resonant isotopes are present, it is needed to include the new scattering kernel treatment for other isotopes like PU240 etc and to analyze their influence on criticality and fuel inventory.

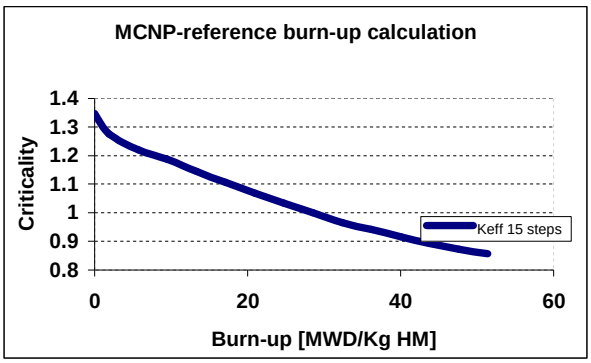

Fig. 1: Keff as function of the burn-up for the Rowlands pin cell benchmark.

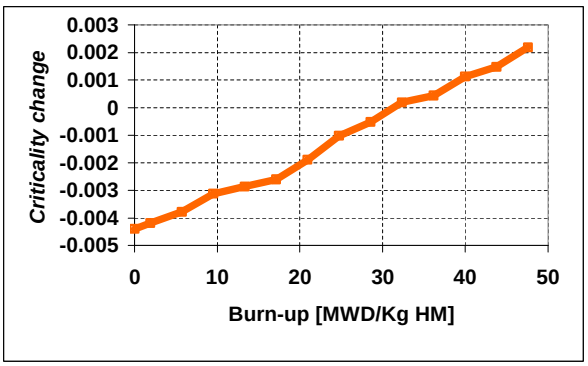

Fig. 2: Change of criticality for a pin cell followed by the introduction of the new scattering kernel.

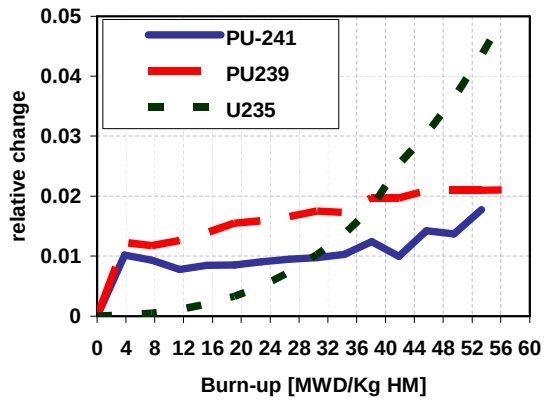

Fig. 3: relative change during the fuel cycle at 1200^0 K due to the usage of the new kernel

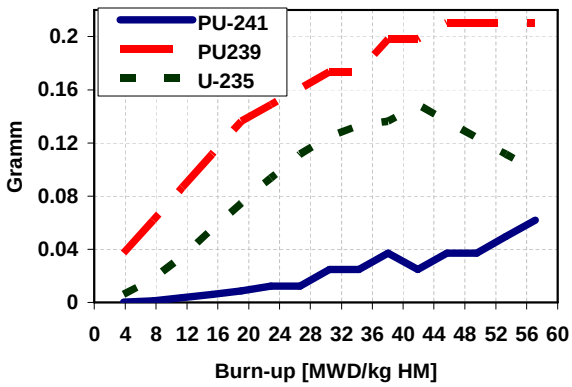

Fig. 4: change in the calculated inventory of several isotopes in gram at 1200 K for a 3.8 meter long pin, due to the use of the new kernel.

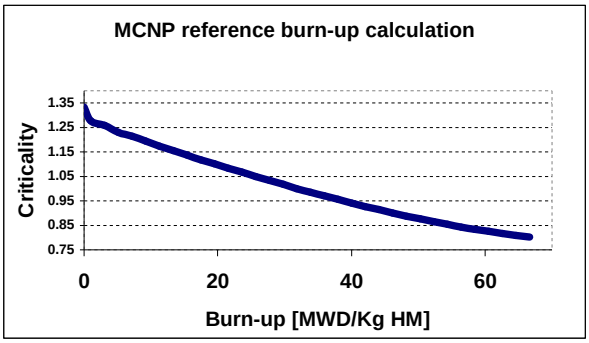

Fig. 5: Keff as function of the burn-up for the GRS S/A Benchmark

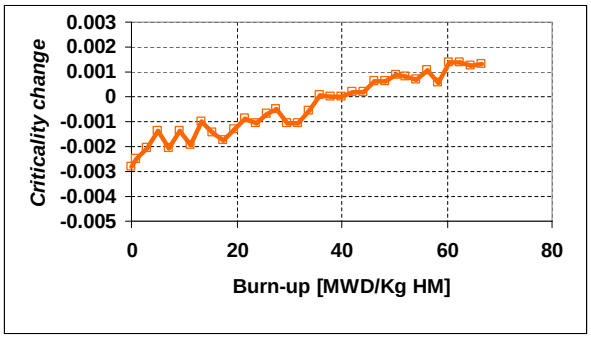

Fig. 6: Change of criticality for a S/A followed by the introduction of the new scattering kernel.

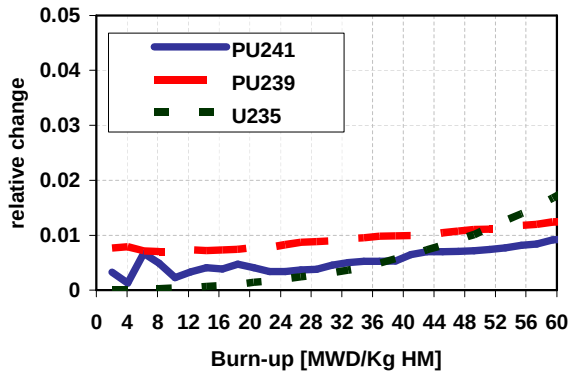

Fig. 7: relative change of averaged fuel pin in S/A during the fuel cycle at 800^0 K due to the usage of the new kernel

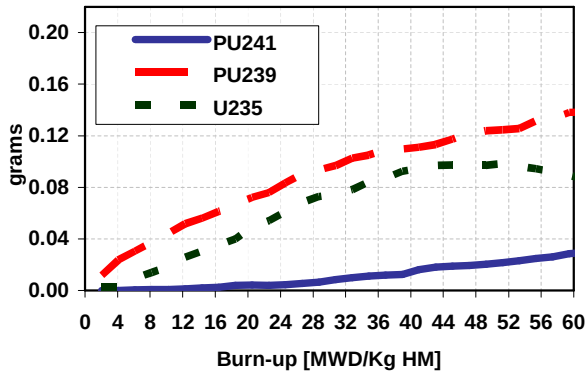

Fig. 8: change in the calculated inventory of several isotopes in gram pro averaged fuel pin 3.8 meter long in the S/A at 800 K due to the use of the new kernel.

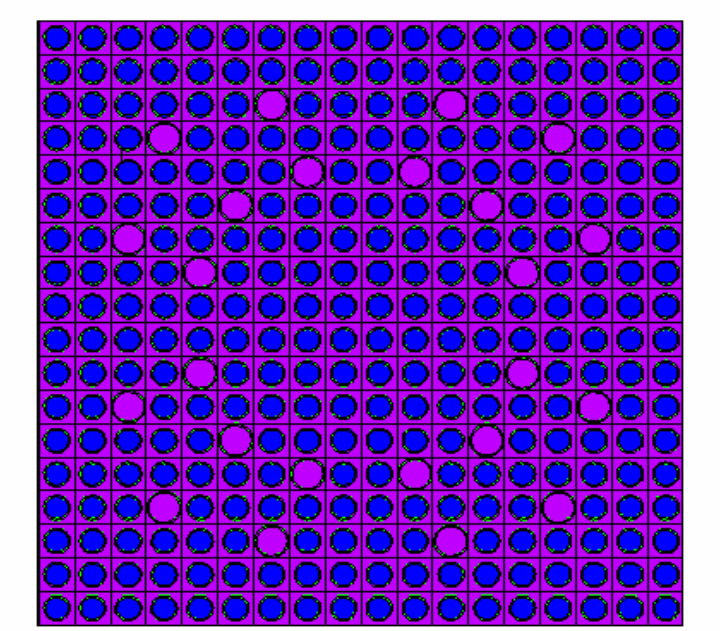

Fig. 9: GRS S/A configuration including 24 water holes.

6. References

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