

Transuranic Transmutation and Criticality Calculation Sensitivity to Heterogeneous Lattice Effects - 12391

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ABSTRACT

Using Mixed Oxide (MOX) fuel in traditional Pressurized Water Reactor (PWR) assemblies has been researched at length and has shown to provide the benefit of transmutation and targets the amount and toxicity of high level waste needed to be managed. Advanced MOX concepts using enriched Uranium Dioxide (UO_2) are required for multiple recycling of plutonium. The use of MOX and ordinary UO_2 fuel in the same assembly as well as unfueled rods and assembly edge effects contrasts with the unit cell computational assumption of a uniform infinite array of rods. While a deterministic method of calculating the Dancoff factor has traditionally been employed in fuel assembly analysis due to the lighter computational and modeling requirements, this research seeks to determine the validity of the uniform, infinite lattice assumption with respect to Dancoff factor and determine the magnitude of the impact of non-uniform lattice effects on fuel assembly criticality calculations as well as transuranic isotope production and transmutation.

INTRODUCTION

To use Mixed Oxide (MOX) fuel in a traditional Pressurized Water Reactor (PWR) without major modifications, Uranium Dioxide (UO_2) must also be used along with a core loading of 20-50% MOX fuel assemblies [1]. The use of MOX and UO_2 fuel in the same assembly affects the unit cell computational assumption of an infinite array of rods. While the uniform, infinite lattice assumption was used in the past due to the lighter computational requirement as well as the uniformity of UO_2 fuel assemblies, advanced heterogeneous fuel assemblies call these assumptions into question.

This research explores the pin-to-pin interaction between the MOX and UO_2 fuel pins in a non-uniform lattice fuel assembly. The impact on criticality and isotope composition calculations by the Dancoff factor calculation method is researched to validate the existing methodology of core analysis in PWRs with respect to Dancoff factor. The deterministic method of Dancoff factor calculation assumes a uniform, infinite lattice whereas a Monte Carlo method can model a non-uniform lattice for the Dancoff factor calculation.

Dancoff Factor

The Dancoff factor is the probability that a neutron emitted isotropically from the surface of one fuel lump will pass through and enter a nearby fuel lump. This correction factor is used to determine the flux reduction in resonance integral calculations. The Dancoff factor can be calculated using Equation 1 [2].

$$C = \frac{\int da \int d\Omega \underline{n} \cdot \underline{\Omega} \exp[-\Sigma_t^M l_M(\underline{a}, \underline{\Omega}) - \sum_{i=1}^{n_c} \Sigma_t^{C,i} l_{C,i}(\underline{a}, \underline{\Omega})]}{\int da \int d\Omega \underline{n} \cdot \underline{\Omega}} \quad (\text{Eq. 1})$$

where $l_M + \sum_{i=1}^{n_c} l_{C,i} + \sum_{j=1}^{n_g} l_{G,j} = l =$ length of a vector (l) leading from the surface point \underline{a} of the fuel region of the fuel element under consideration through the moderator and possibly clads in the direction $\underline{\Omega}$ to the surface of another fuel region

M = moderator

C = clad

G = gap

n_c = number of the clad-crossing segments along l (segments of l being inside a clad region)

$l_{C,i}$ = length of the i -th clad-crossing segment

$\Sigma_t^{C,i}$ = total macroscopic cross section of the clad at the i -th clad-crossing segment

n_g = number of gap-crossing segments along l

$l_{G,j}$ = length of the j -th gap-crossing segment

\underline{n} = normal vector of the surface at point \underline{a}

Σ_t^M = total macroscopic cross section of moderator.

The integrals extend in $\underline{\Omega}$ over all angles $\underline{n} \cdot \underline{\Omega} > 0$ and in \underline{a} over the fuel region surface of the fuel element under consideration.

Deterministic Dancoff Factor Calculation

The deterministic code SCALE (**S**tandardized **C**omputer **A**nalyses for **L**icensing **E**valuation) is a modular code system used to evaluate problem dependent cross-section processing and analysis of criticality safety, and reactor physics problems [3]. SCALE uses the deterministic method of the SUPERDAN algorithm to calculate the Dancoff factor through the above double numerical integration to analytically determine the factor, assuming the fuel assembly is a uniform, infinite lattice of fuel rods. SCALE then calculates the escape probability for an interior region consisting of multiple bodies of the same composition separated by an exterior region with Equation 2.

$$P_I^{(esc)}(\underline{u}_n) = \frac{P_0^{(esc)}(\underline{u}_n)[1-C_1]}{[1-C_1] + C_1 \bar{l}_1 \Sigma_I(\underline{u}_n) P_0^{(esc)}(\underline{u}_n)} \quad (\text{Eq. 2})$$

where $P_0^{(esc)}$ is the escape probability from a single, isolated body in the interior region; \bar{l}_1 is the average chord length of bodies in the interior region; and C_1 is the Dancoff factor [3].

The SUPERDAN algorithm used by SCALE calculates the Dancoff factor for fully absorbing “black” rods. Fuel lumps are considered ‘black’ if they are infinite absorbers such that every neutron that enters into it will undergo absorption. SCALE then uses the Nordheim method of correcting the escape probability for the partial transmission through the absorber regions and taking into account “grey” rods. For ‘grey’ absorbers, there is a finite chance for neutrons to cross a fuel lump without undergoing absorption. This Nordheim correction is

$$P_o^*(E) = P_o(E)(1 - C) / \{1 - [1 - \Sigma_T(E)\bar{r}P_o(E)]C\} \quad (\text{Eq. 3})$$

where $P_o(E)$ is the escape probability, C is the Dancoff factor, $\Sigma_T(E)$ is the total cross section of the absorber medium and \bar{r} is the mean chord length of the absorber region [3].

Monte Carlo Dancoff Factor Calculation

In contrast, DANCOFF-MC is a program that uses a Monte Carlo method to calculate the Dancoff factor in which the assembly architecture and fuel rod composition are explicitly modeled. A neutron is emitted by randomly selecting the position and the direction in which it travels. The lengths travelled in different material regions and the transport probabilities along any given path are calculated according to analytical formulae based on its collision probability definition [2].

For the ‘black’ condition Dancoff factor calculation, the neutron point of emission is selected uniformly at the surface of the fuel rod with a flight path direction based on the probability density of the cosinecurrent. As the flight path goes through regions of fuel and clad, the path lengths in the clad are also evaluated. The exponential attenuation factors determine the probability of a neutron passing through the moderator and clad without a collision using the macroscopic total cross sections of the moderator and clad. This probability is averaged over the different flight directions and points of the surface [2].

To account for the transparency of the fuel lumps for ‘grey’ absorbers, the following expression replaces the exponential in Equation 1:

$$e^{-a_i} \{ (1 - e^{-b_i}) + e^{-a_2} [(1 - e^{-b_2}) + \dots] \}$$

where

$$a_1 = \Sigma_t^{C,1} l_{C,1} + \Sigma_t^M l_{M,1} + \Sigma_t^{C,2} \frac{l_{C,2}}{2} ,$$

$$a_i = \Sigma_t^{F,i-1} l_{F,i-1} + \Sigma_t^{C,i-1} \frac{l_{C,i-1}}{2} + \Sigma_t^M l_{M,i} + \Sigma_t^{C,i} \frac{l_{C,i}}{2} , \quad i=2, \dots, n$$

and

$$b_i = \Sigma_t^{F,i} l_{F,i} , \quad i=1, \dots, n, \quad (\text{Eq. 4})$$

where n is the number of fuel elements crossed. Indexing such as C,i , M,i , and F,i refer to segments in coolant, moderator, and fuel respectively, where i counts the fuel elements crossed, with $i = 0$ referring to the element of origin, $i = 1$ to the first element crossed [2].

METHOD

A CORAIL Assembly

To evaluate the impact of non-uniform lattice effects on Dancoff factor calculations, a French designed CORAIL assembly was used [4]. A CORAIL fuel assembly employs a standard 17x17 PWR assembly but contains 180 UO₂ fuel rods and 84 MOX fuel rods (Figure 1). In order to maintain reactivity coefficients similar to those in a typical UO₂ fuel assembly, the MOX fuel rods are positioned in the peripheral region of an assembly and the fraction of MOX pins in the assembly is limited to roughly one third of all fuel pin locations [4]. This assembly design was used because even with multiple recycling, the CORAIL assembly has been found to have reactivity coefficients for a full core loading of this assembly type that are comparable to a regular UO₂ fueled core [5].

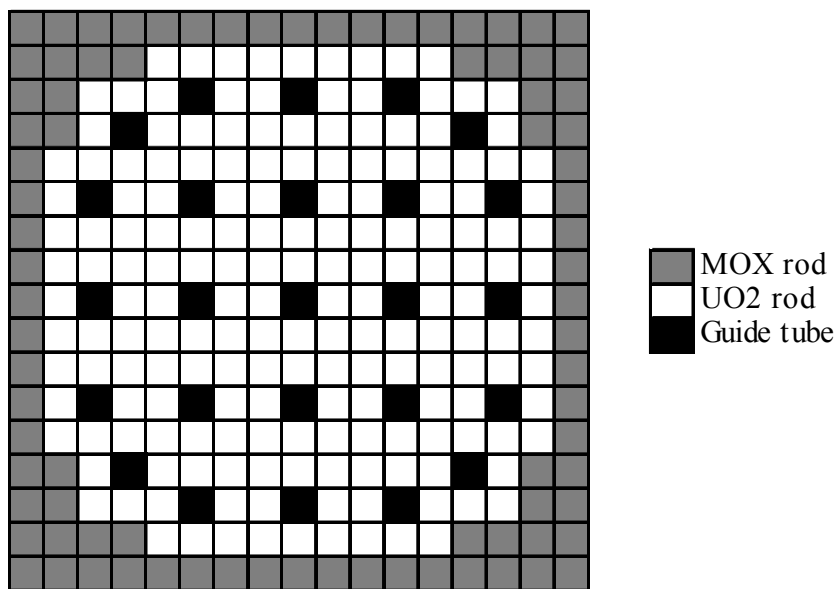


Figure 1. CORAIL Assembly

First, the SCALE program was used to reproduce the benchmark work already performed by researchers to establish that the quarter of an assembly modeled in SCALE to be used for further investigation could be verified [6]. This was done by using the NEW Transport algorithm for two-dimensional discrete ordinates analysis in non-orthogonal geometries (NEWT) sequence of SCALE to calculate the solution to the two-dimensional transport equation using the 238 ENDF6 energy groups. While this type of an assembly would traditionally be modeled under the uniform, infinite lattice assumption and require only one unit cell to represent each fuel type, this research required a 39 unit cell calculation to capture the non-uniform lattice effects. Only the unit cells that would make up an eighth of an assembly had to be modeled due to symmetry. Modeling 39 different unit cells allowed for the manual input of 39 different Dancoff factors to capture the effects of the pin-to-pin interactions in the non-uniform lattice assembly. Table I shows the fuel composition data for the CORAIL assembly modeled [6].

Table I. Fuel Composition for Assembly Modeled

	ISOTOPE	Atomic Density Atoms/barn-cm
UO ₂ PIN	U-235	1.1315E-03
	U-238	2.1226E-02
	O-16	4.4716E-02
MOX PIN	U-235	5.2055E-05
	U-238	2.0508E-02
	Pu-238	6.9723E-05
	Pu-239	7.2243E-04
	Pu-240	5.3327E-04
	Pu-241	2.1750E-04
	Pu-242	2.0904E-04
	Am-241	2.1892E-05
	O-16	4.4667E-02

DANCOFF-MC

Once the 39 unit cell SCALE model was verified against published two unit cell benchmarks, the next step was to calculate the Dancoff factor for each of the 39 unit cells to be manually inputted into the SCALE model. The program DANCOFF-MC was used to calculate these Dancoff factors using a Monte Carlo method to determine the impacts of a non-uniform lattice. A MOX and UO₂ quarter assembly geometry was modeled in FORTRAN which allowed the calling of the DANCOFF-MC subroutines to calculate a Dancoff factor for each fuel rod. Because DANCOFF-MC assumes a void boundary condition on the outside of the model, two additional rows were constructed around the quarter assembly model to ensure the correct neutron escape probabilities could be accounted for in the calculation of the Dancoff factors for the outer most rods of the modeled assembly. The dimensions from the CORAIL assembly benchmark were used with a fuel radius of 1.0482 cm, clad radius of 1.2049 cm, and an assembly half pitch of 1.603 cm [6]. The exact model geometry for DANCOFF-MC can be seen in Figure 2.

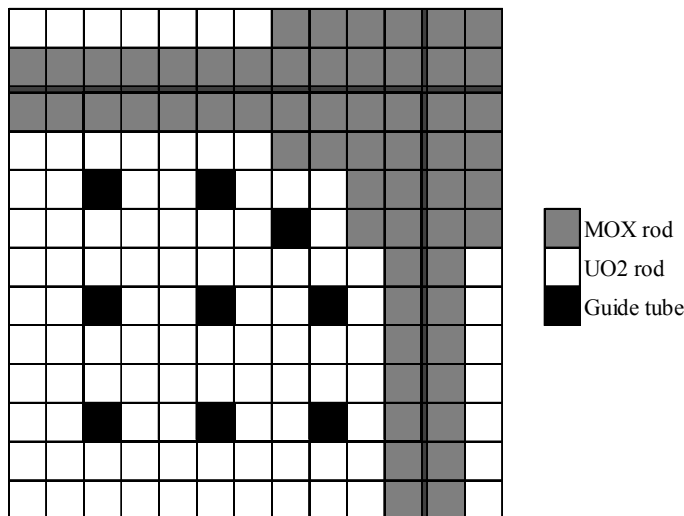


Figure 2. Dancoff-MC Model

DANCOFF-MC requires an input of the total cross-section of the fuel, clad, and moderator for both the UO₂ and MOX fuel pins for the Dancoff factor calculations. TRITON, a two-dimensional transport and depletion module in SCALE, was used to calculate the required cross-sections for the UO₂ and MOX unit cells. The 238 multi-group cross-sections were collapsed into two separate groups and the collapsed cross-sections with energy boundaries from 1 eV to 20 MeV were used as input into DANCOFF-MC. Because the calculation of Dancoff factor is dependent on the escape probabilities for resonance integral calculations, the resonance region was the most important.

DANCOFF-MC calculated the ‘grey’ Dancoff factor for each of the fuel rods in the advanced MOX quarter assembly modeled, each varying due to position in the assembly as well as fuel rod type. These Monte Carlo method derived Dancoff factors were manually inputted in the 39 unit cell SCALE model. Criticality and fuel composition calculations from SCALE were compared between the two methods of Dancoff factor calculation: the manually inputted, Monte Carlo method derived, Dancoff factors and the single SCALE, deterministically calculated, Dancoff factor that was automatically applied to each unit cell. The use of multiple Dancoff factors and unit cells were used to validate the traditional practice of using only one Dancoff factor calculated for a uniform, infinite lattice of one fuel rod type, regardless of position in the assembly.

RESULTS AND DISCUSSION

The DANCOFF-MC calculated Dancoff factors for the ‘grey’ rod condition using the model and methodology described are shown in Figure 3. Figure 4 lists the ‘black’ condition Dancoff factors to show that there is a significant difference in the calculated Dancoff factors when the chance for neutrons to pass through a fuel lump without having a collision is taken into account. SCALE calculated the ‘black’ condition Dancoff factor to be 0.36407 using the deterministic SUPERDAN algorithm.

0.1171	0.1190	0.1189	0.1202	0.1186	0.1188	0.1179	0.1162	0.1102
0.1326	0.1279	0.1202	0.1352	0.1290	0.1268	0.1247	0.1247	0.1164
	0.1385	0.1391		0.1406	0.1388	0.1295	0.1247	0.1182
0.1370	0.1318	0.1322	0.1413	0.1419		0.1384	0.1270	0.1182
0.1366	0.1315	0.1322	0.1402	0.1357	0.1419	0.1406	0.1290	0.1186
	0.1382	0.1395		0.1402	0.1404		0.1357	0.1202
0.1360	0.1312	0.1313	0.1393	0.1323	0.1321	0.1385	0.1286	0.1192
0.1362	0.1311	0.1313	0.1388	0.1312	0.1310	0.1382	0.1284	0.1186
	0.1359	0.1368		0.1367	0.1363		0.1327	0.1176

MOX rod

UO₂ rod

Guide tube

Figure 3. DANCOFF-MC Calculated ‘Grey’ Dancoff Factors

0.3174	0.3174	0.3174	0.3176	0.3174	0.3175	0.3175	0.3171	0.3042
0.3257	0.3290	0.3176	0.3261	0.3290	0.3306	0.3307	0.3307	0.3170
	0.3265	0.3265		0.3248	0.3265	0.3297	0.3304	0.3174
0.3262	0.3293	0.3293	0.3264	0.3246		0.3265	0.3306	0.3175
0.3262	0.3292	0.3292	0.3263	0.3275	0.3246	0.3248	0.3290	0.3175
	0.3264	0.3265		0.3263	0.3266		0.3260	0.3176
0.3264	0.3293	0.3292	0.3265	0.3294	0.3293	0.3265	0.3290	0.3174
0.3264	0.3292	0.3293	0.3265	0.3293	0.3292	0.3265	0.3289	0.3174
	0.3264	0.3263		0.3263	0.3263		0.3258	0.3175

	MOX rod
	UO2 rod
	Guide tube

Figure 4. DANCOFF-MC Calculated 'Black' Dancoff Factors

The 'grey' calculated Dancoff factors from DANCOFF-MC were manually inputted into the 39 unit cell assembly SCALE model that was used to evaluate the eigenvalue (k_{inf}) as a function of burnup. The eigenvalue was also calculated as a function of burnup with the same 39 unit cell assembly model but without a user defined Dancoff factor in the input. Without a user defined input, SCALE used the deterministically calculated 'black' condition Dancoff factor that was corrected to the 'grey' condition as described in the introduction. The same deterministically calculated 'grey' condition Dancoff factor was automatically applied to each of the 39 unit cells. The percent differences of the k_{inf} calculation as a function of burnup between these two methods can be seen in Figure 5. Percent difference is shown as k_{inf} calculated with Monte Carlo Dancoff factors minus k_{inf} calculated with SCALE Dancoff factors divided by k_{inf} calculated with Monte Carlo Dancoff factors times one hundred.

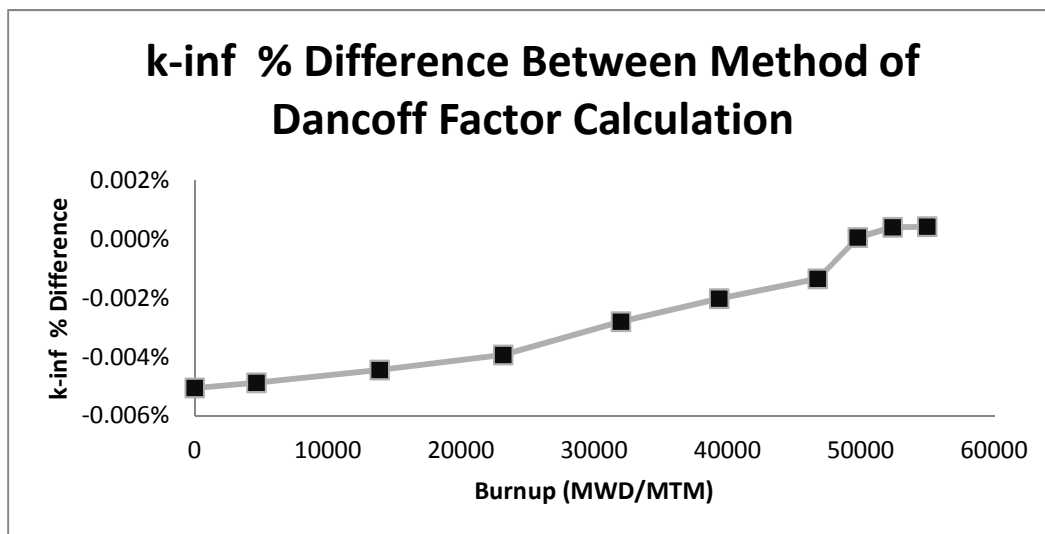


Figure 5. k_{inf} Percent Difference with Non-Uniform Lattice Assumed Dancoff Factors Compared with Uniform, Infinite Lattice Assumed Dancoff Factors

The percent difference is greatest at low burnup and then becomes less significant throughout the cycle. However, when the Dancoff factors calculated by the Monte Carlo method are used, the effects on k-inf calculation as a function of burnup are not significant due to the uncertainty in the ENDF data used by SCALE in these calculations.

In addition to the k-inf differences between the Dancoff factor calculation methods, differences in the transmutation rate of transuranic isotopes in the MOX fuel are also present. The calculated fuel composition at final removal is shown in Figure 6. To calculate the percent differences, the thrice burned final composition in grams prior to cooldown of each of the isotopes initially present in the MOX fuel were compared between the different Dancoff factor calculation methods at 55000 MWD/MTM. Percent difference is shown as grams calculated with Monte Carlo Dancoff factors minus grams calculated with SCALE Dancoff factor divided by grams calculated with Monte Carlo Dancoff factors times one hundred.

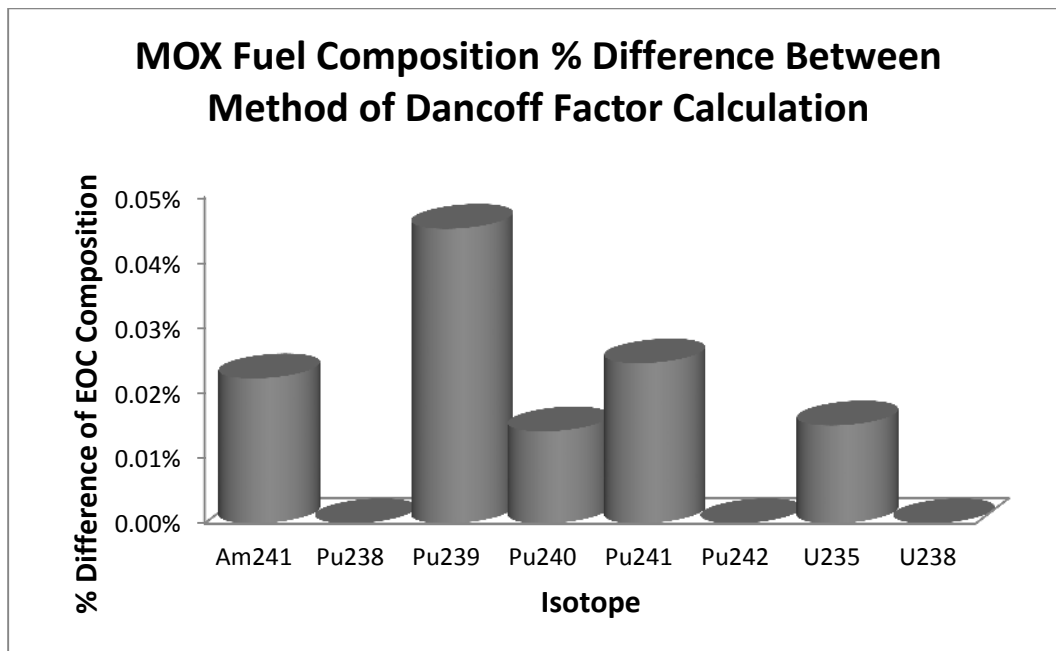


Figure 6. MOX Fuel Composition Percent Difference with Non-Uniform Lattice Calculated Dancoff Factors Compared with Uniform, Infinite Lattice Assumed Dancoff Factor

Differences in the transmutation rate of transuranic isotopes in the MOX fuel are present between the Dancoff factor calculation methods. The largest difference is in Pu-239, Pu-242, and Am-241 composition whereas U-238, Pu-242, and Pu-238 composition was not changed by taking into account the non-homogenous lattice effects. However, these differences are also not great enough to be significant due to the uncertainty in the ENDF data used by SCALE in these calculations.

CONCLUSION

This research explored the pin-to-pin interaction in a non-uniform lattice of MOX fuel rods and UO₂ fuel rods through the impact of the calculated Dancoff factors from the deterministic method used in SCALE versus the Monte Carlo method used in the code DANCOFF-MC. Using the Monte Carlo method takes into account the non-uniform lattice effects of having neighboring fuel rods with different cross-sectional spectra whereas the Dancoff factor calculated by SCALE assumes a uniform, infinite lattice of one fuel rod type.

Differences in eigenvalue calculations as a function of burnup are present between the two methods of Dancoff factor calculation. The percent difference is greatest at low burnup and then becomes smaller throughout the cycle. Differences in the transmutation rate of transuranic isotopes in the MOX fuel are also present between the Dancoff factor calculation methods. The largest difference is in Pu-239, Pu-242, and Am-241 composition whereas U-238, Pu-242, and Pu-238 composition was not changed by taking into account the non-homogenous lattice effects.

Heterogeneous lattice effects do change the calculated eigenvalue and transmutation rate in a non-uniform lattice of MOX fuel rods and UO₂ fuel. However, the uncertainty in the ENDF data used by SCALE in these calculations is large enough that the infinite lattice assumption remains valid.

REFERENCES

1. NUCLEAR ENERGY AGENCY WORKING GROUP, "Plutonium Fuel: An Assessment," *NEA-OECD Report* (1989).
2. S. FEHÉR and P.F.A. DELEEGER, "DANCOFF-MC - A Computer Program for Monte Carlo Calculation of Dancoff Factors in Irregular Geometries," IRI-131-95-003, *Delft University of Technology, Interfaculty Reactor Institute* (June 2007).
3. —, "SCALE 5.1: A Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluation," C00732 MNYCP, Vols. I-III, *Radiation Safety Information Computational Center* (November 2006).
4. G. YOUINOU, "Heterogenous Assembly for Plutonium Multi-recycling in PWRs: the CORAIL Concept," *GLOBAL*, Paris, France (2001).
5. T. K. KIM, J. A. STILLMAN, T. A. TAIWO, R. N. HILL, P. J. FINCK, and M. SALVATORES, "Assessment of Transuranics Stabilization in PWRs," Edited by Argonne National Laboratory, *PHYSOR*, Seoul, Korea (October 7-10, 2002).
6. F. GANDA, E. GREENSPAN, M. MILOSEVIC, and T. TAIWO, "TRITON/NEWT Calculation of the CORAIL Assembly for Plutonium Recycling in PWR." *Proceedings of M&C+ SNA 2007, Joint International Topical Meeting on Mathematics & Computation and Supercomputing in Nuclear Applications*, Monterey, CA (April 15-19, 2007).