Analysis of BWR Lattices to Recycle Americium

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Abstract

This study was carried out to assess the ability to eliminate meaningful quantities of americium in a primarily thermal neutron flux by "spiking" modern BWR fuel with this minor actinide (MA). The studies carried out so far include the simulation of modern 10x10 BWR lattices employing the Westinghouse lattice physics code PHOENIX-4 alongside validation studies using MCNP5 models of the same lattices that were spatially depleted via the MONTEBURNS code coupling to ORIGEN.

When considering the total inventory of minor actinides in Am-spiked pins, excluding isotopes of uranium and plutonium, the results indicate that a reduction of approximately 50% or more in the total mass inventory of these minor actinides is viable within the selected pins. Therefore, these preliminary results have encouraged the extension of this work to the development of improved lattice designs to help optimize the transmutation rates as well as absolute MA inventory reductions. The ultimate goal being to design batches of these advanced BWR bundles alongside multi-cycle core reload strategies.

KEYWORDS: Minor Actinides, Transmutation, BWR, Lattice, Thermal Flux

1. Introduction

Among the minor actinides found in LWR spent nuclear fuel, americium-241 (half-life of 432 years) is among the most radiotoxic during the first two thousand years of storage. Another minor actinide, neptunium-237 (half-life of 2.14 million years) takes over in the longer term, becoming a major contributor to the total dose by ten thousand years, and is predicted to account for about 67% of the total dose by 75 thousand years in a high level waste repository such as Yucca Mountain. Consequently, it is postulated that selective removal of some minor actinides and certain long-lived fission products from nuclear waste, prior to long term storage, can significantly reduce the requirements for storage capacity and storage time at facilities such as Yucca Mountain. The success of this approach, however, relies on the ability to "incinerate" (in a nuclear transmutation context) these highly radiotoxic isotopes extracted from the waste.

This study is intended to help assess the ability to meaningfully eliminate quantities of americium in a primarily thermal neutron flux by "spiking" modern BWR fuel with this minor actinide. The first studies so far include the simulation of modern 10x10 BWR lattices with the Westinghouse lattice physics code PHOENIX-4 alongside studies carried out using MCNP5 models of the same lattices depleted via the MONTEBURNS code coupling to ORIGEN.

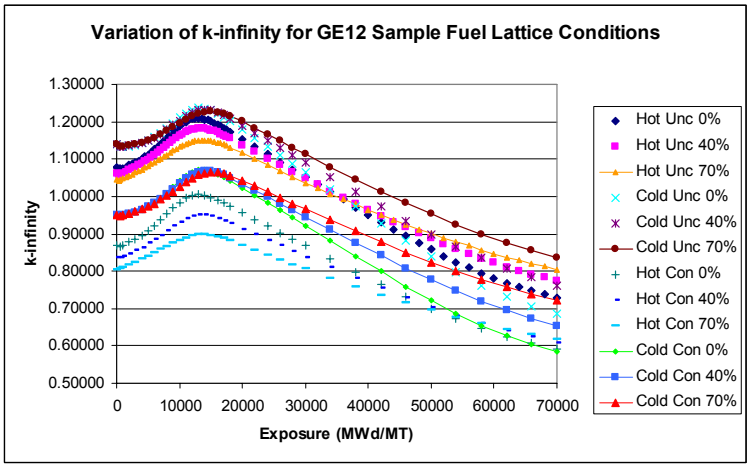
Future studies plan to extend this work to designing bundles (stacked lattices), loading cores with batches of these bundles, and performing multi-cycle calculations employing the POLCA-7 Westinghouse code within CoreMaster2 reload engineering code environment.

1.1 Preliminary Validations

The first key tasks carried out for this project involved the validation of MCNP5 models depleted with MONTEBURNS/ORIGEN, a process still considered to be an emerging technology. Accordingly, comparisons were carried out against the well-seasoned Westinghouse PHOENIX-4 industrial-grade code package by creating inputs for typical 10x10 modern BWR lattices, such as the GE12 model described in Reference [1].

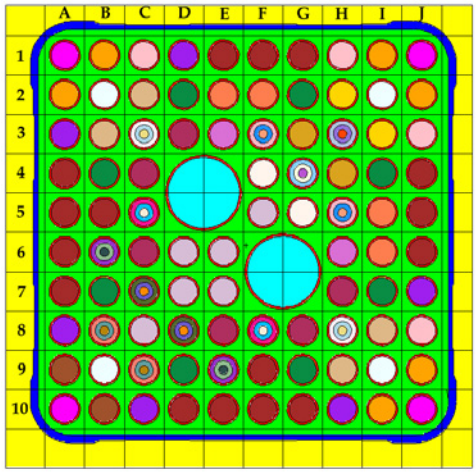
PHOENIX-4 simulations were compared favorably against a few available standards, whereby simulations were carried out for 0%, 40%, and 70% void, at hot and cold operational conditions, and for combinations of controlled and uncontrolled conditions (with and without control blades), as shown in Figure 1. These results confirmed that the trends observed were reasonable.

Figure 1: PHOENIX-4 Results for a 10x10 BWR Lattice [2].



Subsequently, as illustrated in Figure 2 via an MCNP5 visual editor rendition of the "dominant zone" (lower axial region) of a BWR GE12 10x10 fuel bundle, MCNP5 [3] models of these lattices were developed. Labels have been added to help identify individual pin locations.

Figure 2: MCNP5 Model of a 10x10 BWR Lattice with MONTEBURNS Depletion Regions.



With regard to Figure 2, it should be noted that each different color shown denotes an individual "material region" that can be independently depleted via the MONTEBURNS [4] interface to ORIGEN. Due to a built-in limitation on the total number of "material regions" allowed by MONTEBURNS (MB), we chose to maintain symmetry across the diagonal between the two large water holes in blue.

An important highlight of the MCNP5/MB model shown in Figure 2 includes the modeling of independent concentric depletion "rings" in pins with gadolinium loadings. This technique is used to account for the high level of self shielding that occurs in highly absorbing materials, whereby these pins modeled in this manner can deplete in a manner much similar to how an "onion peels," layer by layer. This is best illustrated within Figure 2, in which the density of Gd-155 and Gd-157 in the outer, middle, and inner rings is presented as a function of time. This figure also highlights an important feature of carrying out calculations with the MCNP5 and MB coupling, that is, the ability to extract isotopic "tallies" for any given independent region that can be pre-defined by the user.

Figure 3: Gadolinium Self-Shielding Effect Illustrated from Pins B8 and C9.

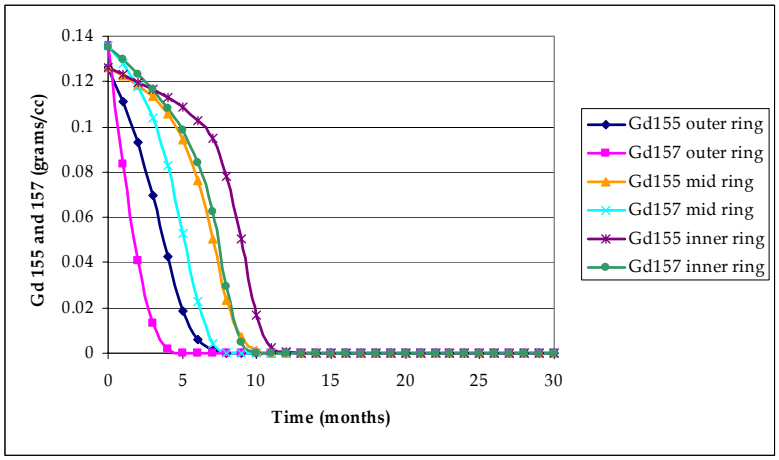
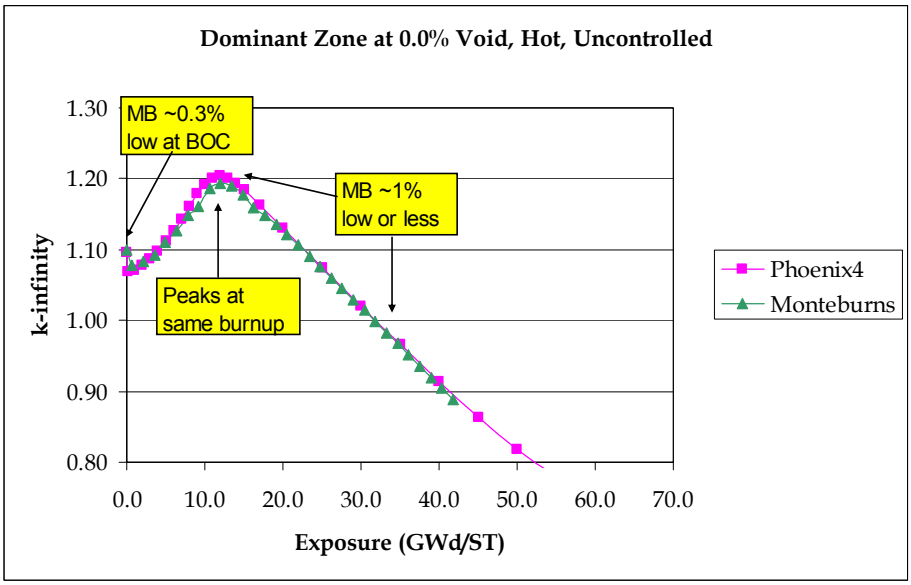


Figure 4 illustrates what could be regarded as a reasonable agreement between the deterministic PHOENIX-4 result and the MCNP5/MB depletion approach to the simulation of a Hot, Uncontrolled, 0% Void case for the same lattice shown above, whereby this simulation reasonably predicts the absolute BOC reactivity, matches well the exposure at the mid-cycle reactivity peak (though not the magnitude), and the reactivity profile aligns well beyond mid-cycle and beyond. Other conditions yielded similar results, and the MCNP5/MB calculations generally seemed to under-predict the results provided by PHOENIX-4. At BOC, the Monte Carlo based results are about 0.3% lower, where as at the reactivity peak through MOC and beyond, the MCNP5/MB results remain approximately 1% or less below the PHOENIX-4 results. These differences were deemed acceptable for side-by-side comparisons of two completely different approaches to BWR lattice depletion. Also these results follow similar trends to those reported by other researchers comparing Monte Carlo based depletions to CASMO lattice physics [5].

Figure 4: MCNP5/MONTEBURNS Depletion Contrasted Against PHOENIX-4.



2. Spiking BWR Fuel with Americium

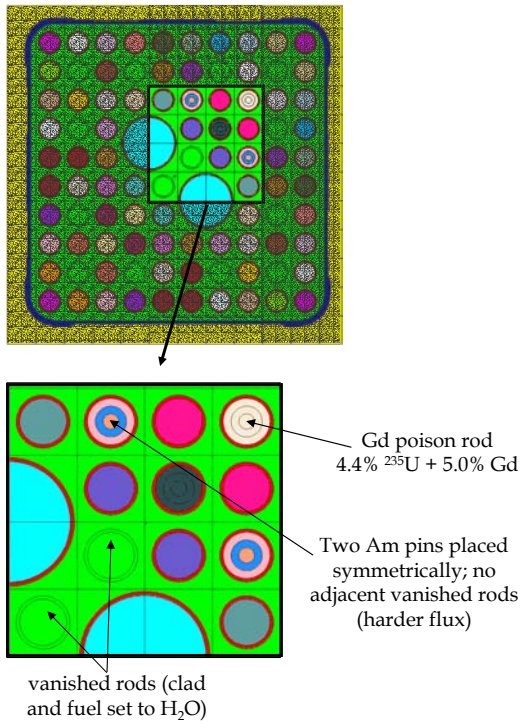
2.1 Proof of Principle Cases

Once various conditions of these 10x10 BWR lattices were modeled with reasonable fidelity, the MCNP5/MB models could then be adjusted to test out a few non-traditional "proof of principle" lattice designs. For instance, by focusing upon the upper axial region or "vanishing zone" of a BWR bundle (the region in which partial length rods are not present), in which a harder neutron spectrum and higher steam void is normally expected, one interesting case herein studied attempted to approximately "match the reactivity" of a couple of selected gadolinium pins by blending AmO_2 with the fuel and gad.

More specifically, for example, a pin with an original BOC loading of "4.4% ^{235}U + 5.0% Gd_2O_3" loading, was iteratively tested (using a single cell model) to try to approximate an equivalent loading of americium that would approximate the reactivity behavior of the original pin without adding this minor actinide. In this particular case, and for this specific pin, of which there were two of them in the assembly, it was determined that a pin with "5% ^{235}U + 3% Gd_2O_3 + 7% AmO_2" would yield a similar reactivity behavior. Figure 5 illustrates the two symmetric pins spiked with americium in the manner described within the vanishing zone of a BWR bundle.

As previously noted, despite the fact that a typical lattice depletion using MCNP5/MB could require approximately 10 CPU days on a modern PC, the ability to "tally" a variety of results anywhere within the defined geometry is quite valuable in this case to help understand what is happening exclusively within these two pins. For example, in the Am-spiked pins shown in Figure 5, the inventory of americium isotopes could be tracked over time for ^{241}Am, ^{242}Am, and ^{243}Am.

Figure 5: MCNP5/MONTEBURNS BWR Pins "Spiked" with Americium.



In fact, Figure 6 shows graphically the depletion of these americium isotopes, whereas Table 1 shows their actual mass (grams) as a function of time extending up to approximately 36 months, in which it appears evident from the data collected that a significant reduction in the inventory of the key americium isotopes is achievable.

Figure 6: Americium isotopic inventory in Am-Spiked pins.

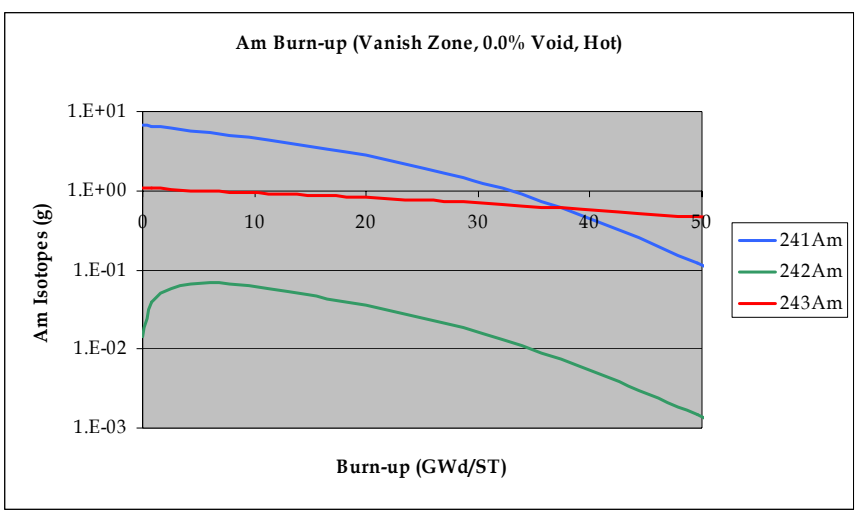
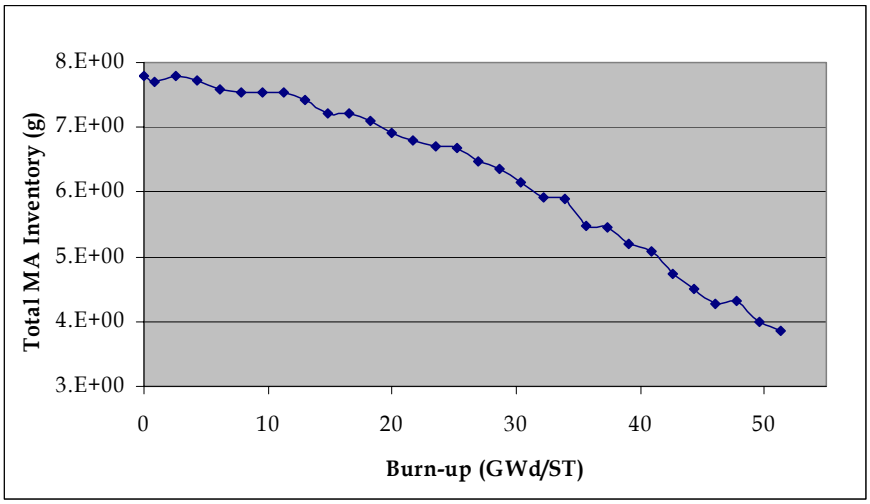


Table 1: Americium isotopic inventory (grams) in Am-spiked pins versus time.

	Time steps (1 time step = 1.2 months)								
	1	4	7	10	13	17	22	27	30
^{241}Am	6.401	5.423	4.386	3.391	2.486	1.462	0.6069	0.1984	0.0937
^{242}Am	0.039	0.069	0.057	0.044	0.032	0.018	0.0074	0.0024	0.0011
^{243}Am	1.061	0.974	0.913	0.851	0.797	0.714	0.6007	0.4979	0.4531

In addition, when considering the total inventory of Minor Actinides in the Am-spiked pins, excluding isotopes of uranium and plutonium, Figure 7 reveals an approximate 50% decrease in the total mass inventory of these MA's within the selected pins.

Figure 7: Total MA inventory in Am-Spiked pins (excluding Pu and U).



2.2 More Practical Cases

To design more practical and realistic lattices, we looked to work being carried out at Oak Ridge National Laboratory on the development of fuel fabrication techniques for a surrogate "Sphere-Pac Rodlet" [6] so to employ compositions of minor actinide (MA) loadings that would be more consistent with feasible manufacturing technologies. For example, we looked at cases in which the initial ^{235}U enrichment for MA pins was 5% and the Am content was 5% and 10%, respectively. Likewise, we also considered cases in which the initial ^{235}U enrichment was 4%, with the density of the MA pins at 8.61 g/cm^3.

As a starting point, we substituted gadolinium pins in the original (reference) lattice with the MA pins in the arrangement shown in Figure 8 to test their performance as possible burnable absorbers. The number in each box denotes the level of U-235 enrichment in each pin, the label "MA" denotes the americium-spiked pins, where as "WR" are the water rods.

Accordingly, Figure 9 illustrates the reactivity performance of such lattice as a function of exposure. In fact, review of Figures 1 and 4 for the reference lattice in contrast to Figure 9 highlights a couple of important observations. First, the reactivity "hold-down" impact at BOC

for the 5% and 10% americium loadings appears to be sufficient to maintain the peak reactivity at or below that for the reference lattice, while still achieving a comparable EOC reactivity. Second, it is evident that the burnout rate of the MA pins is quite different than that of gadolinium pins, whereas Figures 1 and 4 display the classic burnable absorber (BA) behavior in which the burnout rate of the BA early in the cycle edges out the depletion rate of the fuel, therefore, overall reactivity increases early in the cycle until the BA burns out and the fuel depletion rate takes over. In contrast, the MA pins deplete at a more uniform rate throughout the cycle and, thus, do not exhibit the reactivity swing traditional to BA loaded lattices.

Figure 8: Test lattice with "MA" pins substituted for burnable absorbers.

	A	B	C	D	E	F	G	H	I	J
1	2.00	2.80	3.60	4.40	4.40	4.40	3.95	3.60	2.80	2.00
2	2.80	4.40	3.95	4.90	3.95	3.95	4.90	3.95	4.40	2.80
3	3.60	3.95	MA	MA	MA	4.90	4.40	MA	3.95	3.95
4	4.40	4.90	MA	MA	4.90	WR		4.40	4.90	4.40
5	4.40	3.95	MA	4.90	4.40			MA	4.40	4.40
6	4.40	3.95	4.90	WR		4.40	4.40	4.40	MA	4.40
7	3.95	4.90	4.40			4.40	4.40	MA	4.90	4.40
8	3.60	3.95	MA	4.40	MA	4.40	MA	4.40	MA	3.95
9	2.80	4.40	3.95	4.90	4.40	MA	4.90	MA	4.40	3.60
10	2.00	2.80	3.95	4.40	4.40	4.40	4.40	3.95	3.60	2.00

Figure 9: Reactivity behavior for MA loaded lattices.

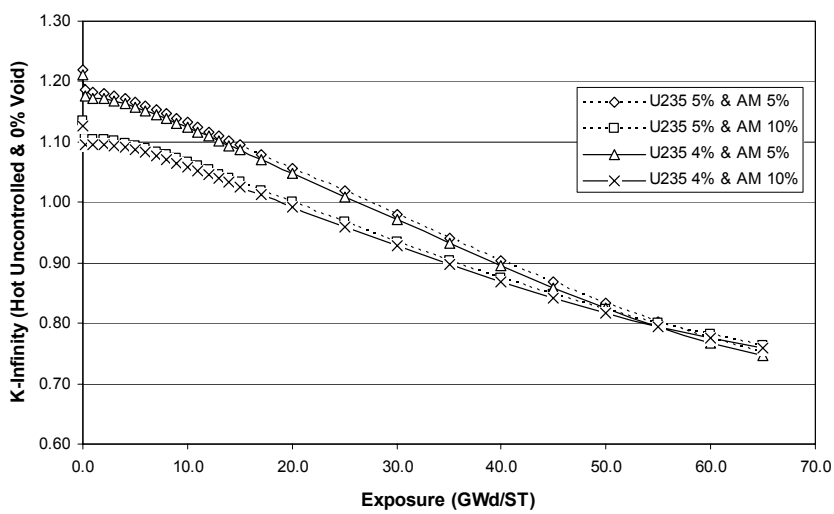
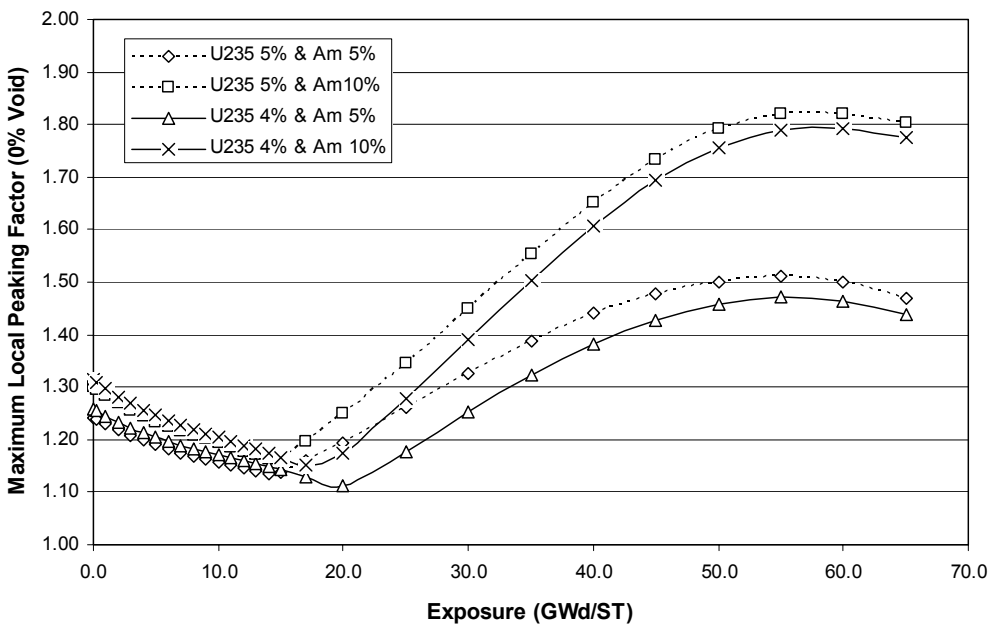


Figure 10 shows the local power peaking factors observed in the MA-loaded lattices, which for the 10% americium loadings, in particular, dramatically increase at the end of cycle to levels rendering these specific designs to not be readily feasible. Therefore, at this point, adjustments and optimization of the loadings and pattern arrangements provide the future direction for this research. Nevertheless, the results herein presented provide some key insights, which are outlined in the next section.

Figure 10: Local Power Peaking in MA loaded lattices.



3. Conclusions

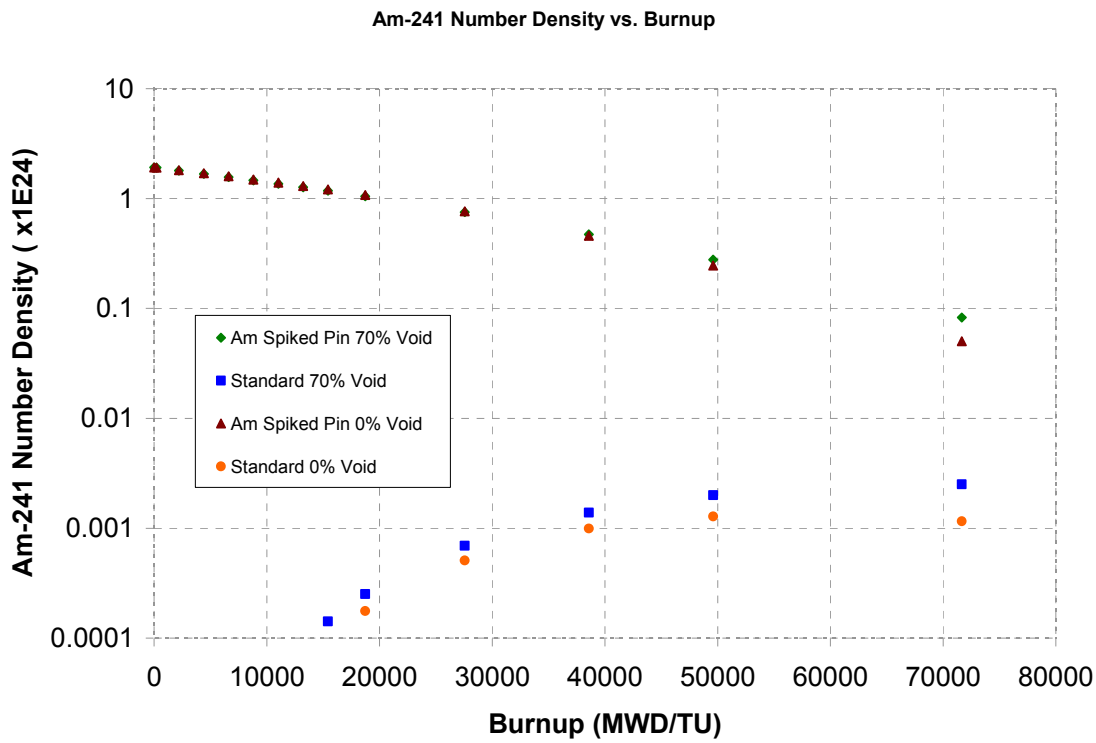
Pre-loaded americium can be transmuted within BWR bundles. For the more realistic MA bundle considered in Section 2.2, the net reduction of TRU (Np, Pu, Am and Cm) is about 40.35% of the initial americium loaded. These results are consistent with the proof of principle "two-pin" case considered in Section 2.1 in which reductions of about 50% appear to be feasible.

These early results also indicate that MA pins could potentially act as burnable absorbers if properly designed, even though considerable "tuning" of these specific lattice designs remains to be done at this time, especially to address the end of cycle power peaking excursions highlighted by Figure 10.

Review of the fissile inventory of U-235, Pu-239, and Pu-241 shows important trends that indicate that the inventory of fissile plutonium dramatically increases in these lattices relative to a standard/reference lattice without MA pins, potentially creating as much as 3 times greater quantities of Pu-239 and 1.5 times greater quantities of Pu-241, thus, indicating a lower initial U-235 enrichment requirement for Am-spiked bundles. In fact, we're currently studying lattices that use U-235 enrichments consistent with tails in the MA-spiked pins as another possibly interesting option to take advantage of the additional breeding taking place.

Another important result has recently emerged from this research that is somewhat counterintuitive. When comparing the Am-241 transmutation rates in higher steam void environments, we've noted that these are not dramatically different than those cases with 0% void content, especially at EOC. In fact, the data shown in Figure 11 actually illustrates that at end of cycle, the transmutation rates of Am-241 in the 70% void environment are actually lower than for the 0% void cases, a behavior which holds true for the reference lattices without MA loaded and the Am-spiked cases, both. This result may be an indication that the spectral effects due to a higher void content (i.e., a harder neutron spectrum and slightly higher fission to absorption cross section) are being overshadowed by other competing effects taking place, such as the production of fissile plutonium isotopes ultimately competing for neutrons, as one possible contributing factor.

Figure 11: Americium destruction with higher steam void content.



In closing, these early results, despite only being carried out at the lattice level so far, provide encouraging conclusions in support of the continuation of this project. Future studies will lead to pin by pin lattice loading optimization, and ultimately to bundle and core design (multi-cycle) studies employing the Westinghouse CoreMaster2 work environment that couples PHOENIX-4 to the three-dimensional simulator POLCA-7.

Acknowledgements

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