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HARDENING NEUTRON SPECTRUM FOR ADVANCED ACTINIDE TRANSMUTATION EXPERIMENTS IN THE ATR

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The most effective method for transmuting long-lived isotopes contained in spent nuclear fuel into shorter-lived fission products is in a fast neutron spectrum reactor. In the absence of a fast test reactor in the United States, initial irradiation testing of candidate fuels can be performed in a thermal test reactor that has been modified to produce a test region with a hardened neutron spectrum. Such a test facility, with a spectrum similar but somewhat softer than that of the liquid-metal fast breeder reactor (LMFBR), has been constructed in the INEEL's Advanced Test Reactor (ATR). The radial fission power distribution of the actinide fuel pin, which is an important parameter in fission gas release modelling, needs to be accurately predicted and the hardened neutron spectrum in the ATR and the LMFBR fast neutron spectrum is compared. The comparison analyses in this study are performed using MCWO, a well-developed tool that couples the Monte Carlo transport code MCNP with the isotope depletion and build-up code ORIGEN-2. MCWO analysis yields time-dependent and neutron-spectrum-dependent minor actinide and Pu concentrations and detailed radial fission power profile calculations for a typical fast reactor (LMFBR) neutron spectrum and the hardened neutron spectrum test region in the ATR. The MCWO-calculated results indicate that the cadmium basket used in the advanced fuel test assembly in the ATR can effectively depress the linear heat generation rate in the experimental fuels and harden the neutron spectrum in the test region.

INTRODUCTION

In nuclear power generation, the treatment of spent fuel produced during the operation of commercial power plants is one of the most important issues not only to the nuclear community but also to the general public. One of the viable options of long-term geological disposal of spent fuel is to extract plutonium, the minor actinides (MAs) and potentially long-lived fission products from the spent fuel and transmute them into short-lived or stable radionuclides in a reactor appropriate for the reduction of the radiological toxicity of the nuclear waste stream. An important component of that technology will be a non-fertile actinide transmutation fuel form containing the plutonium, neptunium, americium and (possibly) curium isotopes to be transmuted. The advanced fuel forms, especially ones enriched in the long-life MA (LLMA) elements (i.e. Np, Am and Cm), have few irradiation performance data available from which to establish a transmutation fuel form design. Thus, preliminary irradiation tests on a variety of candidate fuel forms are needed. Recognising these needs, an Advanced Fuel Cycle test series-1 (AFC-1) irradiation test on a variety of candidate fuel forms is now being conducted at the Advanced Test Reactor (ATR), Idaho National Engineering and Environmental Laboratory (INEEL) with the advanced fuel supplied by Los Alamos National Laboratory (LANL) and Argonne National Laboratory-West (ANL-W).

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There is no domestic fast spectrum test reactor and the complications associated with international shipments of nuclear materials are costly and formidable. Furthermore, many fuel performance issues are primarily functions of temperature and/or power and dependent upon neutron spectrum only as a lower order effect. Testing of transmutation fuels is expected to produce useful data regarding such fuel performance issues as irradiation growth and swelling, helium production, gas release, fission product and fuel constituent migration, and fuel phase equilibria.

Currently, two low-fertile candidate fuel forms are under consideration by the AFCI for use in fast-spectrum applications: nitride and metallic fuels. A series of low-fertile fuel tests are being conducted in the ATR. For simplicity in comparing the detailed radial fission power profiles, actinide depletion and build-up and important neutron cross sections vs. effective full power days (EFPDs), an AFC-1F rodlet-4 (U-29Pu-4Am-2Np-30Zr) with density = 11.42 g cm^{-3}, was chosen as the reference advanced fuel specimen in this paper. Note that the metallic alloy compositions are expressed in wt%. The ^{235}U enrichment in the fuel test rodlet is 78 wt%.

METHODOLOGY: MCWO

Important neutronics parameters, such as, detailed radial fission power profile, linear heat generation rate (LHGR) and burnup are needed for the fuel performance and fission gas release analyses. The major source of uncertainty in the fuel burnup calculation comes from burnup-dependent cross sections (XS)[1], resonance treatment of neutron spectrum vs. fuel enrichment and minor long-life actinide XS. A UNIX BASH (Bourne Again Shell) script MCWO has been developed at INEEL to couple the Monte Carlo transport code MCNP[2] with the depletion and buildup code ORIGEN2[3]. MCWO[4] is a fully automated tool that links the Monte Carlo transport code MCNP with the radioactive decay and burnup code ORIGEN2. MCWO can handle a large number of fuel burnup and material-loading specifications, ATR powers and irradiation time intervals. The program processes input from the user that specifies the system geometry, initial material compositions, feed/removal specifications and other code-specific parameters. Calculated results from the MCNP, ORIGEN2 and data process module calculations are then output successively as the code runs. The principal function of MCWO is to transfer one-group cross section and flux values from MCNP to ORIGEN2 and then transfer the resulting material compositions (after irradiation and/or decay) from ORIGEN2 back to the MCNP in a repeated, cyclic fashion. Verification of MCWO was made by comparing the MCWO-calculated concentration profiles with post-irradiation examination data[5,6].

MCWO was also used to calculate cadmium (Cd) isotopic concentrations and compositions, and effective cross section vs. burnup. At the beginning of irradiation, the peak LHGR of the metal fuel rodlet-4 (U-29Pu-4Am-2Np-30Zr) with and without the absorber filter are 237 and 2174 W cm^{-1}, respectively[7]. Owing to the depletion of ^{113}Cd during irradiation, to hold down the linear heat and to maintain a hardened neutron spectrum, we have to replace the Cd and Al shroud every cycle (~48 EFPDs) in this study.

ATR AND LMF LATTICE MODELS

The ATR core consists of a serpentine and rotationally symmetric fuel zone about the z-axis of the core centre, with the core divided into five major power lobes—NW, NE, Centre, SW and SE. The major components of the model are the fuel elements, the outer shim control cylinders, neck shims and major experiment facilities. The MCNP ATR full core model cross-sectional view is shown in Figure 5 of Ref. (4).

All fast reactors have a similar neutron-flux spectrum. For convenience and availability of data to the authors, we chose the liquid-metal fast breeder reactor (LMFBR) lattice model as our reference model to generate the fast neutron-flux spectrum. LMFBR has a hexagonal shape fuel assembly arrangement. The pitch (centre-to-centre) of the hexagonal fuel channel is 13.78 cm. Fuel element outer diameter (OD) and pitch is 0.65 and 0.795 cm, respectively. For the detailed description of the LMFBR model see Ref. (8). The LMFBR lattice model was used to generate the neutron spectrum for the actinide burnup analysis.

We have chosen the following three cases for the advanced fuel neutronics burnup characteristics comparison study.

Case 1: ATR neutron spectrum—the fuel test assembly with Cd-filter (see the next subsection for the Cd-filter description).
Case 2: ATR neutron spectrum—the fuel test assembly with Al-basket (no Cd-filter).
Case 3: LMFBR neutron spectrum—the fuel test assembly with stainless steel (SST)-basket.

For the ATR full core model and the LMFBR lattice model, MCNP was used to generate the needed neutron-flux spectra. The comparison of the MCNP-calculated Cd-filter, Al-basket in the ATR and the LMFBR unit lattice neutron-flux spectra with SST-basket is plotted in Figure 1. It clearly shows that Case 2 with Al-basket has the softest neutron spectrum, and the LMFBR has the hardest neutron spectrum.

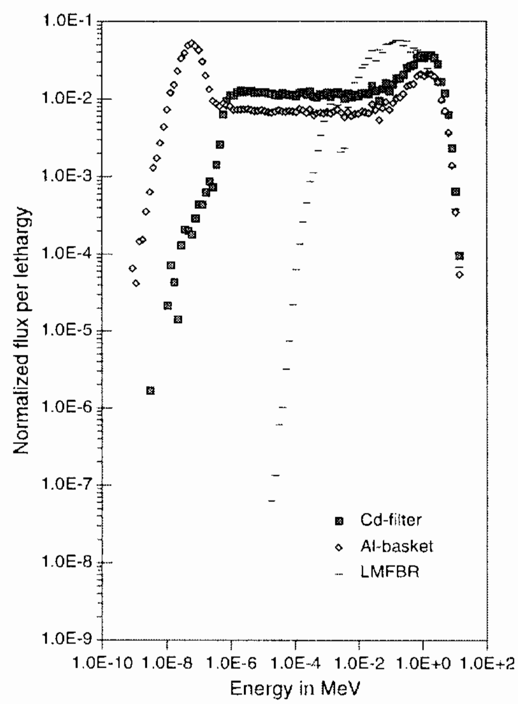

Figure 1. Comparison of the Cd-filter, Al-basket in ATR and the LMFBR unit lattice with SST-basket neutron-flux spectra.

Table 1. AFC-1F: low-fertile, metallic-fuelled experiment fuel column constituent densities (g cm^{-3}).

AFC-1F	^{237}Np	^{235}U	^{238}U	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	Zr	Total density
Rodlet 1	0.27	3.16	0.89	0.00	2.83	0.56	0.02	0.01	0.43	3.46	11.62
Rodlet 2	0.27	1.84	3.42	0.00	3.71	0.73	0.02	0.02	0.54	2.56	13.10
Rodlet 3	0.20	3.04	0.23	0.00	2.21	0.44	0.01	0.01	0.25	4.29	10.68
Rodlet 4	0.24	3.10	0.88	0.00	2.68	0.53	0.02	0.01	0.41	3.57	11.42
Rodlet 5	0.02	3.72	0.28	0.00	2.70	0.54	0.02	0.01	0.61	3.48	11.38
Rodlet 6	0.22	2.84	0.21	0.00	2.16	0.43	0.01	0.01	0.29	4.26	10.43

Advanced fuel test assembly model description

The current test train for AFC-1 fuel testing in the ATR consists of six variants of transuranic-based fuels containing Am and Np. Each miniature fuel pin is 6 inch in total length and all pins are externally identical.

The fundamental component, which contains the fuel specimen, is termed a rodlet. The irradiation vehicle will contain six rodlets, designated as rodlet 1 to rodlet 6. The radial and axial views of the fuel test assembly at the ATR East flux trap position[9] (Figure 1). The rodlet-4 of the metallic fuelled AFC-1F experiment evaluated in this paper has a fuel slug diameter of 0.4013 cm and 3.81 cm length. Table 1 provides the detailed fuel composition for the chosen reference rodlet-4 in the AFC-1F test assembly.

The passive absorber-type filter, in the form of a Cd-filter basket with a 0.114 cm (0.045 inch) Cd thickness and 121.92 cm (48 inch) length, is currently used in the actinide-fuel capsule design for the East flux trap position in the ATR to depress the LHGR in the experimental fuels and to harden the neutron spectrum.

Isolated fuel test assembly model description

To improve the efficiency in the MCNP when tallying over a very small rodlet cell and detailed radially divided rodlet sub-cell, we developed an MCNP isolated fuel pin model with a spherical neutron source surrounding the fuel test assembly to compute the detailed power profile within the fuel pins in the ATR. First, a detailed MCNP ATR core model was used to generate the neutron-flux spectrum in the test assembly with the Al–Cd-filter basket. Then, the generated spectrum was used as the isolated fuel pin model's outer spherical surface source to calculate the detailed radial fission power profiles in the fuel pins. The outer spherical surface boundary is set to be 230 cm, which is large enough to contain the whole-advanced fuel test assembly. An inward-directed neutron source with the MCNP-calculated neutron spectrum will bombard the fuel rodlet model. The inward-directed source with the unbiased cosine distribution will generate a spectrum-dependent uniform neutron-flux field[10]. As a result, MCWO can be used to calculate the detailed fluxes and neutron reaction rates in the fuel rodlet sub-divided cells.

For the fuel test assembly at the East flux trap position, MCNP (Fixed-Source mode with four tasks, nps $= 5 \times 10^8$) calculations were performed, where each time step run requires 160 min of DELL-650 XEON-2-CPU 3.06 GHz workstation computer time to achieve 1 SD (1σ) $< 0.6\%$ in the sub-cell fission tallies.

RESULTS AND DISCUSSION

MCWO was used to calculate neutron-flux spectra, isotopic concentrations and compositions, and fission power vs. burnup. For a typical ATR cycle, the lobe power splits are: 18 MW (NW), 18 MW (NE), 23 MW (C), 25 MW (SW) and 25 MW (SE), which represent an E-lobe power-(NE + C + SE)/3 of 22 MW. MCWO-calculated results are normalised to an ATR E-lobe power of 22 MW. Owing to the depletion of ^{113}Cd during irradiation, we have to replace the Cd and Al shroud every cycle (~48 EFPDs) to hold down the linear heat and maintain the neutron hard spectrum.

The average power density in the fuel rodlet is assumed to be 2400 W cm^{-3} in this study, which represents an LHGR of 303.6 W cm^{-1}. The MCWO calculations have each depletion time interval set to be 16 EFPDs. The MCWO-calculated results, for U, Np, Am and Pu isotope concentrations, $^{240}Pu/Pu$ ratio vs. EFPDs and detailed radial fuel kernel fission power profile vs. fuel compact's fraction of radius (r/r_0) at different burnups are presented and discussed.

Comparison of the actinide neutron cross sections at the beginning of life

The neutron-spectrum-averaged one-group neutron cross sections for Cases 1, 2 and 3 at the beginning of life (BOL) irradiation are tabulated in Table 2. For

Table 2. MCNP-calculated one group cross sections of AFC-1F advanced fuel rodlet with Cd-filter at the beginning of irradiation life in ATR East-flux trap position.

		(n,γ) barns	(n-2n) barns	(n-3n) barns	(n,f) barns
^{232}U	922320	1.684E+01	2.223E-03	0.00E+00	1.917E+01
^{233}U	922330	6.872E+00	1.334E-03	0.00E+00	3.399E+01
^{234}U	922340	2.30E+01	4.276E-04	0.00E+00	4.815E-01
^{235}U	922350	4.456E+00	2.967E-03	3.633E-010	1.031E+01
^{236}U	922360	1.129E+01	2.282E-03	8.361E-06	3.517E-01
^{237}U	922370	1.228E+01	5.932E-03	3.986E-05	5.352E-01
^{238}U	922380	4.799E+00	3.773E-03	4.811E-06	8.958E-02
^{237}Np	932370	2.658E+01	8.345E-04	0.00E+00	4.871E-01
^{238}Np	932380	8.210E+00	2.228E-03	0.00E+00	3.659E+01
^{238}Pu	942380	8.041E+00	1.232E-03	0.00E+00	1.732E+00
^{239}Pu	942390	5.545E+00	1.555E-03	0.00E+00	1.102E+01
^{240}Pu	942400	4.764E+01	6.779E-04	0.00E+00	5.465E-01
^{241}Pu	942410	7.835E+00	5.421E-03	3.457E-06	2.518E+01
^{242}Pu	942420	4.812E+01	1.730E-03	6.893E-07	4.112E-01
^{241}Am	952410	3.836E+01	1.902E-04	0.00E+00	6.821E-01
^{242}Am	952421	6.946E+00	5.287E-03	1.315E-05	5.773E+01
^{243}Am	952430	7.284E+01	2.042E-04	0.00E+00	4.602E-01
^{242}Cm	962420	4.293E+00	5.227E-05	0.00E+00	3.298E-01
^{243}Cm	962430	1.115E+01	2.794E-03	0.00E+00	8.964E+01
^{244}Cm	962440	2.731E+01	1.085E-03	0.00E+00	1.089E+00

Note: Because the occurrences of (n-3n) reaction are very small, the tallies of (n-3n) are not reliable. The (n-3n) cross sections tabulated here are only for reference purpose

the constant source power density, the total neutron fluxes of the Cases 1–3 are 4.56×10^{14}, 1.15×10^{14} and 2.92×10^{15} n cm^{-2} s^{-1}, respectively. Case 3 has the lowest U and Pu fission cross sections, which means, for a constant power density, the total neutron flux should be higher than the thermalised cases. The total neutron fluxes are very different between the thermal and fast spectrum environments; however, since the cladding material to be employed in the fast spectrum transmutation system will be a SST alloy traditionally used in the fast reactors, its irradiation performance is already well established and need not be demonstrated by the AFCI.

Comparison of the fuel isotopes concentration and actinides isotopic ratio vs. EFPD

The MCWO-calculated sum of ^{235}U and ^{239}Pu isotopes average atom density (10^{24} atom per cm^3 = atom per b-cm) vs. EFPDs have about the same ($^{235}U + {}^{239}Pu$) depletion at the end of 304 EFPDs for the same power density in three study cases. The higher transmutation of the ^{237}Np through ^{238}Np beta decay (half-life 2.117 d) to ^{238}Pu, ^{239}Pu and ^{241}Pu in Case 1, makes a little less depletion in the sum of ^{235}U and ^{239}Pu.

The MCWO-calculated ^{237}Np depletion vs. EFPDs is plotted in Figure 2. The ^{237}Np depletion rate is a function of the ^{237}Np absorption-XS ($\sigma_a = \sigma_\gamma + \sigma_f$) and target fission power. In our three case studies with a constant neutron-flux field, the fission power is proportional to ^{239}Pu-σ_f. The ratio of ^{237}Np-σ_a to ^{239}Pu-σ_f, which is an index of ^{237}Np depletion rate, for Cases 1–3 are 2.45, 0.45 and 1.14, respectively. Note that ^{237}Np XS has a high resonance over the neutron energy range 0.7–100 eV. The Cd-filter modifies the neutron spectrum such that all the thermal neutrons are eliminated, while still keeping the intermediate neutron spectrum. For this reason, Case 1 has the highest

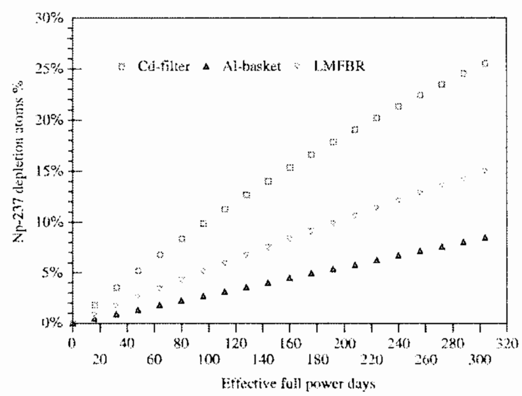

Figure 2. Comparison of the ^{237}Np fraction depletion (atom%) vs. EFPDs.

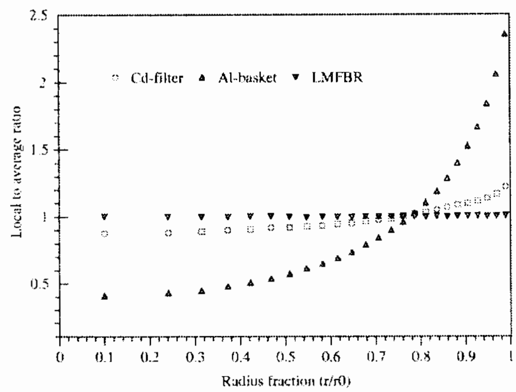

Figure 3. Comparison of the radial fission power profiles at the BOL.

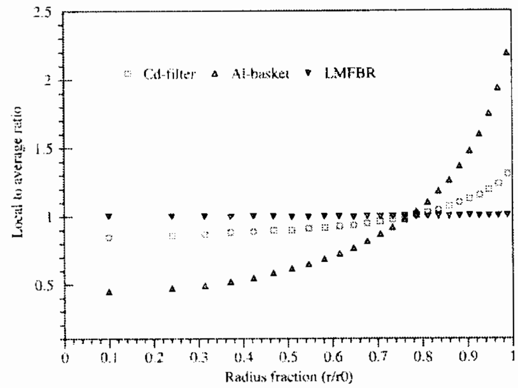

Figure 4. Comparison of the radial fission power profiles at 304 EFPDs.

ratio of ^{237}Np-σ_a to ^{239}Pu-σ_f. As a result, Case 1 also has highest depletion in ^{237}Np atom% at the end of 304 EFPDs.

An important issue of burning actinide fuel is that the discharged fuel shall meet the spent fuel standard[11]. The isotopic compositions of the discharged actinide fuel should be about the same as the LWR UO_2 spent fuel, particularly, the $^{240}Pu/Pu$ ratio should be >25%. The MCWO-calculated $^{240}Pu/Pu$ ratio at the end of 304 EFPDs, for Cases 1–3 are 14.1, 23.6 and 20.9%, respectively.

Comparison of the detailed radial fuel rodlet fission power profiles vs. EFPDs

Rodlet fission heat generated will transfer radially. The radial profiles are needed for tuning the important fission gas release modelling. The detailed radial mesh in this study contained 25 subdivided equal volume sub-cells in the fuel rodlet. The results of the fuel rodlet burnup analysis performed provide the detailed relative radial fission power profile vs. EFPDs. The neutronics analysis of the detailed relative radial fission power profiles at the BOL and 304 EFPDs in ATR Cases 1 and 2, and LMFBR Case 3 were calculated and are compared.

The MCWO-calculated (each depletion time interval was set to 16 EFPDs) relative radial fission power profiles of the fuel rodlet are shown in Figures 3 and 4. Because of the spatial and spectral self-shielding effects, especially in the thermalied neutron field, the fission power density is higher in the fuel outer shell for Cases 1 and 2. The MCWO-calculated radial power profiles indicate that the softest neutron spectrum, Case 2 has the highest rim-effect, which represents a peak local fission power to an average ratio of 2.34. However, for Cd-filter Case 1, the peak local fission power to average ratio at outmost shell only reaches 1.22, which is closer to the Case 3 fission power profile, because the hardest neutron spectrum does not have any significant neutron self-shielding effect. Owing to the depletion of the fissile materials ^{235}U and ^{239}Pu, the fission XS increase for Cases 1 and 2 towards the end of irradiation. For the hard neutron spectrum in Case 3, the ^{235}U and ^{239}Pu fission XS do not show any significant variation over irradiation time. As a result, there is no significant rim-effect observed in the Case 3 radial fission power profiles vs. EFPDs. From Figures 3 and 4, the radial fission power profiles for all three cases did not show large variations over time.

CONCLUSIONS

The ability to accurately predict the advanced fuel pellet radial power profile, burnup, and burnup-dependent XS is essential in the advanced fuel performance evaluation. This paper demonstrated that the MCWO method could provide the needed accurate neutronics parameters. We show that the fission power profile differences between the three cases studied are quite significant. The ratio of fission power in the outermost rodlet shell for Cases 1–3 are 1.22, 2.34 and 1.01, respectively.

The MCWO-calculated burnup and fission heat rate distributions, and Cd-filter depletion of the proposed AFC-1 vs. EFPDs shows that the Cd-filter can harden the neutron spectrum and effectively reduce the LHGR of the advanced fuel rodlet to meet the AFC-1 experiment needs. In addition, the Cd-filter can also reduce the rim-effect in the radial fission power profile.

The developed MCWO method is being used to perform the neutronics analysis for particle fuel testing in the ATR. The MCWO method can also be used in a wide variety of other applications, including advanced high-temperature gas-cooled reactor (both fast and thermal neutron-flux Gen-IV reactors) fuel cycle performance analysis, LLMA transmutation, strong absorber depletion analysis, actinide fuel design and reactor materials test assembly design.

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