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CADMIUM DEPLETION IMPACTS ON HARDENING NEUTRON SPECTRUM FOR ADVANCED FUEL TESTING IN ATR

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ABSTRACT

For transmuting long-lived isotopes contained in spent nuclear fuel into shorter-lived fission products effectively is in a fast neutron spectrum reactor. In the absence of a fast spectrum test reactor in the United States of America (USA), 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. A test region is achieved with a Cadmium (Cd) filter which can harden the neutron spectrum to a spectrum similar (although still somewhat softer) to that of the liquid metal fast breeder reactor (LMFBR). A fuel test loop with a Cd-filter has been installed within the East Flux Trap (EFT) of the Advanced Test Reactor (ATR) at the Idaho National Laboratory (INL).

A detailed comparison analyses between the cadmium (Cd) filter hardened neutron spectrum in the ATR and the LMFBR fast neutron spectrum have been performed using MCWO. MCWO is a set of scripting tools that are used to couple the Monte Carlo transport code MCNP with the isotope depletion and buildup code ORIGEN-2.2. The MCWO-calculated results indicate that the Cd-filter can effectively flatten the Rim-Effect and reduce the linear heat rate (LHGR) to meet the advanced fuel testing project requirements at the beginning of irradiation (BOI). However, the filtering characteristics of Cd as a strong absorber quickly depletes over time, and the Cd-filter must be replaced for every two typical operating cycles within the EFT of the ATR. The designed Cd-filter can effectively depress the LHGR in experimental fuels and harden the neutron spectrum enough to adequately flatten the Rim-Effect in the test region.

Key Words: AFCI, MCWO, ORIGEN-2.2, Cd-filter, Neutron Spectrum Hardening, Advanced Test Reactor.

1. INTRODUCTION

To support the U.S. Advanced Fuel Cycle Initiative (AFCI) program for disposition of plutonium and minor actinides from spent fuel, both non-fertile and uranium-bearing metal and oxide fuels containing minor actinides are under consideration for use as recycled transmutation fuels. However, little irradiation performance data exists for any fuel form containing a significant fraction of minor actinides. An experiment containing metal and oxide actinide fuels has been fabricated and irradiated in the Advanced Test Reactor (ATR) at the Idaho National Laboratory (INL). The first irradiation tests of high-actinide-content fuels, the AFC-1 series tests, within the ATR began in 2003.

The most effective method for transmuting long-lived isotopes contained in spent nuclear fuel into shorter-lived fission products would be to irradiate in a fast neutron spectrum reactor. However, there are no domestic fast spectrum test reactors, and for international fast spectrum test reactors 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 the advanced recycled fuels is expected to produce useful data regarding such fuel performance issues as irradiation growth and swelling, helium production, fission gas release, fission product and fuel constituent migration, and fuel phase equilibrium. Those experiment objectives can be achieved by using a Cadmium (Cd) filter around a fuel test.

For a light water reactor (LWR), the conversion of ^{239}Pu from ^{238}U occurs around the outer rim of a fuel pin, and the self-shielding of the thermal neutrons causes a steep fission power profile in the radial local to average ratio (L2AR) of about 2.5 at the outer rim – "Rim-Effect" [1]. The radial fission power profile of the actinide fuel pin, which is an important parameter in fission gas release modeling, needs to be accurately predicted and compared between the cadmium (Cd) filter hardened neutron spectrum in the ATR and the LMFBR fast neutron spectrum. The comparison analyses have been performed using MCWO. MCWO is a set of scripting tools that couples the Monte Carlo transport code MCNP with the isotope depletion and buildup code ORIGEN-2.2. The results in Ref. 2, indicate that the Cd-filter effectively flattens the Rim-Effect and reduces the linear heat generation rate (LHGR) to meet the AFCI project testing requirements at the beginning of irradiation (BOI). However, the filtering characteristics of Cd as a strong absorber quickly depletes over time and the Cd-filter needs to be replaced for every two typical operating cycles within the East Flux Trap (EFT) of the ATR. A typical ATR operating cycle has 50 effective full power days (EFPDs) of irradiation. In this work, a detailed fuel target and Cd-depletion analysis has been performed to demonstrate that a Cd-filter replaced every other operating cycle can effectively depress the linear heat generation rate (LHGR) in the experimental fuels and harden the neutron spectrum to flatten the Rim-Effect in the test region during the entire irradiation life of ~300 EFPDs.

2. AFC-1/2 FUEL ASSEMBLY MODEL AT EFT POSITION

The ATR full core was modeled (Fig. 1.) to represent the power splits and operating conditions projected for a typical ATR operating cycle with an East lobe source power of 23 MW. The AFC-1 model consists of six variants of transuranics (U, Pu, Am and Np) containing zirconium-based alloy fuels (in each of the E-1 to E-4 capsule positions). Each fuel pin is 6 inches in length and contains one fuel rodlet. A nominal fuel rodlet has a fuel slug diameter of 0.4013 cm and length of 3.81 cm, with all rodlets similar in size and appearance. Each test assembly contains 6 fuel pins (rodlets), axially designated as rodlet 1 through rodlet 6. The axial and radial views of the fuel test assembly within the EFT of the ATR are shown in Fig. 2. The axial view for the capsule arrangement in the E-1 position is on the left side.

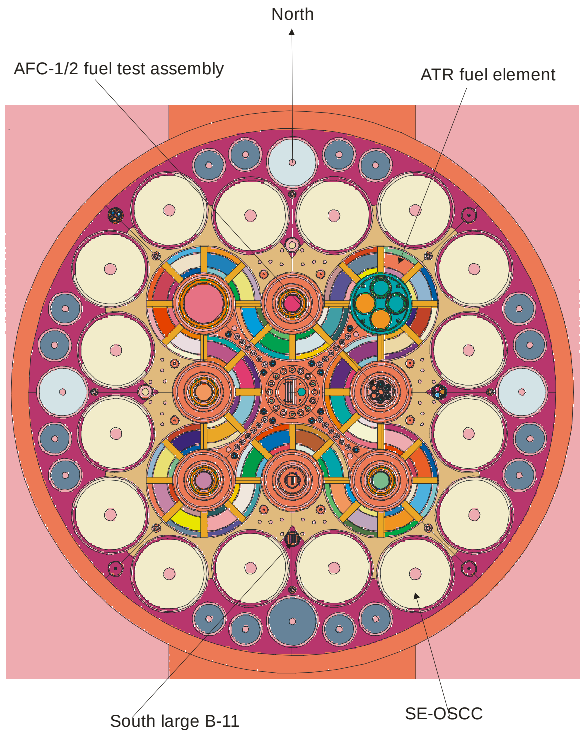

Figure 1. MCNP full core radial cross-section view and the AFC-1/2 series fuel test assembly at the ATR EFT position.

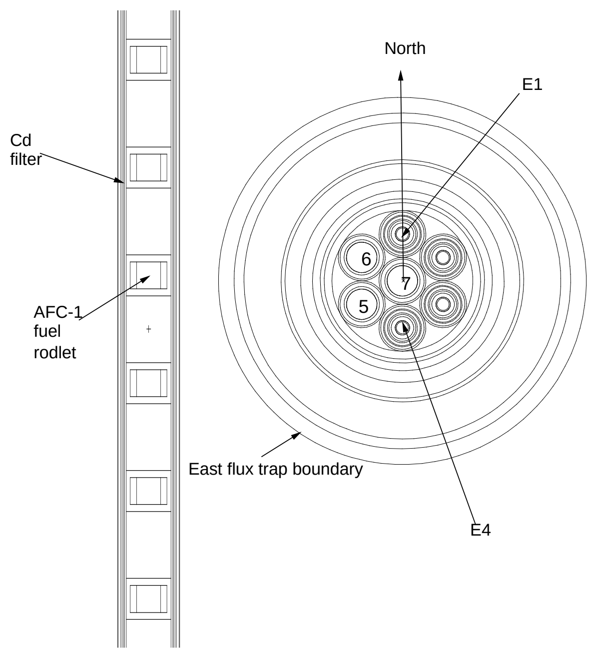

Figure 2. Axial and radial views of AFC-1 test assembly arrangement in EFT position of the ATR.

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

3. FUEL AND ABSORBER BURNUP ANALYSIS CODE – MCWO

Important neutronics parameters, such as detailed radial fission power profile, 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) [3], resonance treatment of neutron spectrum versus fuel enrichment, and minor long-life actinide XS. A UNIX BASH (Bourne Again Shell) script named MCWO has been developed at INL to couple the Monte Carlo transport code MCNP [4,5] with the depletion and buildup code ORIGEN2.2. [6,7] MCWO [8] is a set of scripting tools linking the Monte Carlo with ORIGEN. MCWO can handle a large number of fuel burnup and material loading specifications, ATR powers, and irradiation time intervals. The scripts process input from the user that specifies the system geometry, initial material compositions, feed/removal specifications, and other code-specific parameters. Calculated results from MCNP, ORIGEN2.2, and data process module calculations are then output as the code runs. The principal function of MCWO is to transfer one-group cross-section and flux values from MCNP to ORIGEN2.2, and then transfer the resulting material compositions (after irradiation and/or decay) from ORIGEN2.2 back to MCNP in a repeated, cyclic fashion. Verification of MCWO was made by comparing the MCWO-calculated concentration profiles with post-irradiation examination (PIE) data [8,9].

MCWO was also used to calculate absorber Cd isotopic concentrations and compositions, and effective cross section versus burnup. At the beginning of irradiation, the peak LHGR of the metal fuel rodlet, AFC-1F rodlet-4 (U-29Pu-4Am-2Np-30Zr) with and without the absorber filter are 305 W/cm and 2,745 W/cm, respectively [10]. Due to the depletion of ^{113}Cd during irradiation, the Cd-filter and Al basket are replaced every two cycles (~100 EFPD) in order to hold down the linear heat and maintain a hardened neutron spectrum.

4. RESULTS AND DISCUSSION

To meet the current defined AFCI testing requirements for the AFC-1/2 drop-in experiments, a Cd-filter with Cd thickness of 0.114 cm (0.045") effectively holds down the linear heat rate and hardens the neutron spectrum. The physics analysis was performed using the computer code MCWO currently being used for evaluation of experimental programs in the ATR. The fuel and absorber depletion tool, MCWO, is used to calculate burnup, fission heat rate distributions, and Cd-filter axial depletion profile versus EFPDs.

The typical ATR axial power profile is a chopped cosine shape with a core center peak–to-average ratio of 1.414. Rodlet-4 within each capsule is located near the ATR core mid-plane. For simplicity in comparing the detailed radial fission power profiles, actinide depletion and build-up, and important neutron cross sections versus effective full power days (EFPD), an AFC-1F rodlet-4 (U-29Pu-4Am-2Np-30Zr) with density = 11.42 g/cm3, was chosen as the reference advanced fuel specimen in this study. Note that the metallic alloy compositions are expressed in weight percent. This report describes the results of the MCWO calculations performed to provide the isotopes depleted and built-up at the 1st beginning of cycle (BOC), 1st end of cycle (EOC), 2nd EOC, 3rd BOC, 3rd EOC, 4th EOC, 5th BOC, 5th EOC, and 6th EOC for a total of 300 EFPDs.

The analysis was performed using a burnup time interval of 10 EFPD and an East lobe power of 23 MW. In the study case run, 30 time intervals were modeled for a total of 300 EFPD. For each time step, an MCNP fixed-source calculation with 6.0×10^8 source neutrons was performed. The fission tally calculation for each fuel node achieves a 1σ standard deviation of 0.8% or less.

Because the oxide fuel has lower fuel density and oxygen (O^2) neutron scattering cross section than the metal (Zr) fuel, the oxide fuel is more transparent for the source neutrons. For the comparison of the radial fission power profile versus Cd-filter depletion between metal and oxide fuel, an AFC-2D oxide rodlet-3 (U17Pu-2.4Am-1.3Np-12.3O) with density of 9.64 g/cc was chosen in this study. The MCWO-calculated oxide fuel rodlet results are presented and discussed in Section 4.4.

4.1 Comparison of the Neutron Flux Spectrum, Fission Power L2AR Profile at BOL

The following three cases for the neutronics burnup characteristics comparison study were chosen.

Case-1: ATR intermediate neutron spectrum - the fuel test assembly with Cd-filter. Case-2: ATR thermal neutron spectrum - the fuel test assembly with aluminum (Al)-basket (no Cd-filter). Case-3: LMFBR fast neutron spectrum - the fuel test assembly with stainless steel (SST)-basket.

For the ATR full core model and LMFBR lattice model,2 MCNP was used to generate the neutron flux spectra. The comparison of the MCNP-calculated Cd-filter, Al-basket in ATR and the LMFBR unit lattice with SST-basket neutron flux spectra is plotted in Fig. 3. The results clearly show that Case-2 with the Al-basket has the softest neutron spectrum, and the LMFBR has the hardest neutron spectrum. The fractions of the fast neutron flux (E > 0.1 MeV) of Cases-1, -2. and -3 are 0.43, 0.58, and 0.25 respectively. This shows that the Cd-filter can adequately harden the neutron spectrum in ATR EFT position.

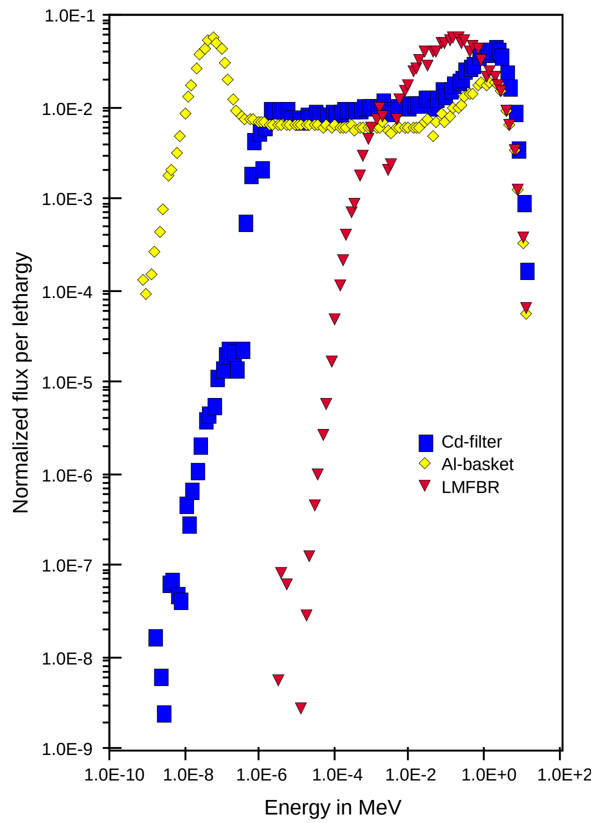

Figure 3. Comparison of the Cd-filter, Al-basket in ATR and the LMFBR unit lattice neutron flux spectra.

For the assumed fission power density, 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 stainless steel alloy traditionally used in fast reactors, its irradiation performance is already well established and need not be demonstrated by the AFCI irradiation tests.

Rodlet-generated fission heat will transfer radically. The radial profiles are needed for tuning the important fission gas release modeling. The detailed radial mesh in this study contained 25 equally subdivided sub-cells (by volume) in the fuel rodlet. The neutronics analysis of the detailed relative radial fission power profiles at the beginning of life (BOL) in ATR for Cases-1 and -2, and LMFBR Case-3 were calculated and are compared in Fig. 4. The MCWO-calculated results are normalized to an ATR E-lobe power of 23 MW.

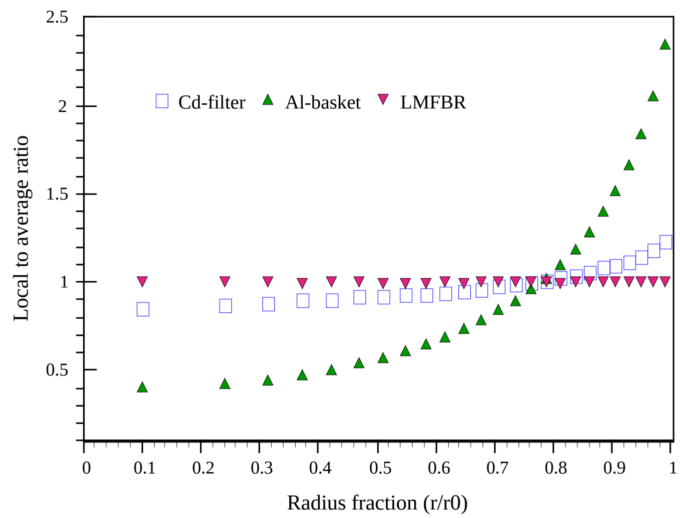

Figure 4. Comparison of the radial fission power profiles at the beginning of irradiation life.

One of the AFC-1 testing requirements is to demonstrate that the peak LHGR will be less than 350 W/cm. The LHGR is assumed to be 303.6 W/cm for Case-1 with Cd-filter at the 1st BOC in this study. The corresponding LHGR for Case-2 without Cd-filter is 2757.3 W/cm, which is much higher than the 350 W/cm limit.

4.2. Cadmium Physics Property, Neutron Cross Sections, and Isotopes Concentration versus EFPDs

A summary of the Cd isotopes abundance and decay half-life times is tabulated in Table I. The table I indicates that ^{112}Cd has a rather high abundance of 24.13%, which can transmute to ^{113}Cd by capturing a neutron. Because of this transmutation to ^{113}Cd, which has the highest neutron absorption XS, will slow down the decrease of the effectiveness of neutron absorption versus irradiation time.

MCWO-calculated 8 major Cd isotopes (^{106}Cd, ^{108}Cd, ^{110}Cd, ^{111}Cd, ^{112}Cd, ^{113}Cd, ^{114}Cd, and ^{116}Cd) atomic density (AD), and the neutron spectrum weighted micro- and macro-scopic (n,γ) capture cross sections at the beginning of irradiation are tabulated in Table II. Table II indicates that ^{113}Cd has the highest σ(n,γ) cross section (XS) of 197.8 b and the highest Σ(n,γ) XS of 1.06 cm-1, which represents a 97.8% of the total Cd Σ(n,γ) XS.

Table I: Cadmium isotopes physics properties.

Isotopes	Abundance	half-life
^{106}Cd	1.25%	>9.5×10^{17} y
^{107}Cd		6.5 h
^{108}Cd	0.89%	>6.7×10^{17} y
^{109}Cd		462.6 d
^{110}Cd	12.49%	^{110}Cd is stable with 62 neutrons
^{111}Cd	12.80%	^{111}Cd is stable with 63 neutrons
^{112}Cd	24.13%	^{112}Cd is stable with 64 neutrons
^{113}Cd	12.22%	7.7×10^{15} y
^{113m}Cd		14.1 y
^{114}Cd	28.73%	>9.3×10^{17} y
^{115}Cd		53.46 h
^{116}Cd	7.49%	2.9×10^{19} y

Table II. Summary of the Cd isotopes AD and cross-section data at the BOC for 1st irradiation cycle (0 EFPD).

Cd-isotopes	Day-00 AD	Microscopic	Macroscopic
	(atoms/b-cm)	σ(n,γ) (barn)	Σ(n,γ) (cm^{-1})
Cd-106	5.73E-04	0.5149	2.950E-04
Cd-108	3.92E-04	1.143	4.484E-04
Cd-110	5.51E-03	0.7871	4.337E-03
Cd-111	5.64E-03	1.878	1.060E-02
Cd-112	1.06E-02	0.4155	4.416E-03
Cd-113	5.38E-03	197.8	1.064E+00
Cd-114	1.27E-02	0.2999	3.795E-03
Cd-116	3.31E-03	0.08302	2.744E-04
XS Σ(n,γ)			1.088E+00

The MCWO-calculated Cd isotopes AD, and the neutron spectrum weighted (n,γ) and Σ(n,γ) capture cross sections at the 1st EOC and at the end of the 2nd EOC were calculated. The Cd-filter is discharged at the 2nd EOC with 100 EFPDs of irradiation. The neutron-spectrum-averaged one-group neutron cross-sections for the Cd-filter at the 1st EOC and at the end of 2nd EOC are tabulated in Tables III and IV, respectively.

Table III. Summary of the Cd isotopes AD and cross-sections data at the 1st EOC (50 EFPD).

Cd-isotopes	Day-50 AD	Microscopic	Macroscopic
	(atoms/b-cm)	σ(n,γ) (barn)	Σ(n,γ) (cm^{-1})
Cd-106	5.73E-04	0.5134	2.939E-04
Cd-108	3.92E-04	1.135	4.444E-04
Cd-110	5.50E-03	0.816	4.490E-03
Cd-111	5.63E-03	1.93	1.087E-02
Cd-112	1.06E-02	0.4191	4.458E-03
Cd-113	3.62E-03	277.6	1.005E+00
Cd-114	1.44E-02	0.2884	4.155E-03
Cd-116	3.31E-03	0.08288	2.739E-04
XS Σ(n,γ)			1.030E+00

Table IV. Summary of the Cd AD and cross-section data at 2nd EOC (100 EFPD).

Cd-isotopes	Day-100 AD	Microscopic	Macroscopic
	(atoms/b-cm)	σ(n,γ) (barn)	Σ(n,γ) (cm^{-1})
Cd-106	5.72E-04	0.5103	2.919E-04
Cd-108	3.91E-04	1.124	4.395E-04
Cd-110	5.50E-03	0.8619	4.737E-03
Cd-111	5.62E-03	2.03	1.142E-02
Cd-112	1.06E-02	0.4227	4.50E-03
Cd-113	2.28E-03	407.6	9.290E-01
Cd-114	1.57E-02	0.2774	4.368E-03
Cd-116	3.31E-03	0.08256	2.729E-04
XS Σ(n,γ)			9.550E-01

Due to the neutron self-shielding effect, the neutron-spectrum-averaged neutron cross sections are functions of the irradiation EFPD. The ^{113}Cd represents about 97.8 % of neutron absorption in the Cd-filter. The ^{113}Cd depletion percent at the 1st EOC and at the end of 2nd are 32.65% and 57.61%, respectively. However, the depletion of the Cd-filter, particularly ^{113}Cd, its σ(n,γ) XS will increase from 1st BOC toward the 2nd EOC (Cd-filter discharged) as 197.8 b, 277.6 b, and 407.6 b, respectively. As a result, the effectiveness of the Cd-filter in capturing the thermal neutrons is reduced to about 87.8% at the 2nd EOC (100 EFPD).

4.3. Cd-filter depletion Impacts on the detailed METAL fuel radial Fission Power Profiles versus EFPDs

Fuel rodlet-generated fission heat will transfer radically. The radial profiles are needed for tuning the important fission gas release modeling. The detailed radial mesh in this study contained 25 equally subdivided sub-cells (by volume) in the fuel rodlet. Due to the depletion of ^{113}Cd during irradiation, we have to replace the Cd-filter every two cycles (~ 100 EFPD) to hold down the LHGR and maintain the hardened neutron spectrum. The MCWO-calculated results are normalized to an ATR E-lobe power of 23 MW. The results of the fuel rodlet burnup analysis performed provide the detailed relative radial fission power profiles at the 1st beginning of cycle (BOC), 1st end of cycle (EOC), 2nd EOC, 3rd BOC, 3rd EOC, 4th EOC, 5th BOC, 5th EOC, and 6th EOC for a total of 300 EFPDs. The MCWO-calculated relative radial fission power profiles of the fuel rodlet are shown in Figs. 5, 6, and 7.

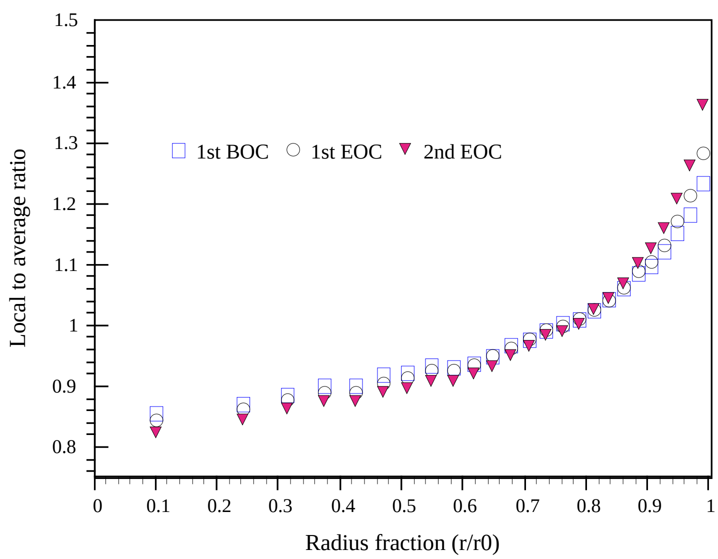

Figure 5. MCWO-calculated detailed radial fission power density profiles at the 1st beginning of cycle (BOC), 1st end of cycle (EOC), 2nd EOC.

Figs. 5, 6, and 7, clearly show that the metal radial fission power profiles did not show large variations over irradiation time. Their radial fission power density profiles are very similar to Case-2 in Figure 4. Due to the ^{113}Cd depletion impact on the metal radial fission power density profiles, the peak L2AR increases from 1.235 (1st BOC) to 1.359 (6th EOC).

The MCWO-calculated fuel rodlet LHGR for 30 time steps is shown in Fig. 8. Because the Cd-filter depleted faster than the fissile nuclides in the fuel rodlet, the LHGR increased from 305.0 W/cm at 1st BOC to 334.6 W/cm at 2nd EOC. After the Cd-filter is replaced, the LHGR increases from 304.5 W/cm at 3rd BOC to 334.2 W/cm at 5th EOC. Finally, after the Cd-filter is replaced again, the LHGR increased from 304.0 W/cm at 5th BOC to 331.2 W/cm at 6th EOC. These results strongly indicate that the Cd-filter effectively maintains the LHGR below 350 W/cm, and flattens the radial fission power density profile during the whole fuel irradiation duration of ~300 EFPD.

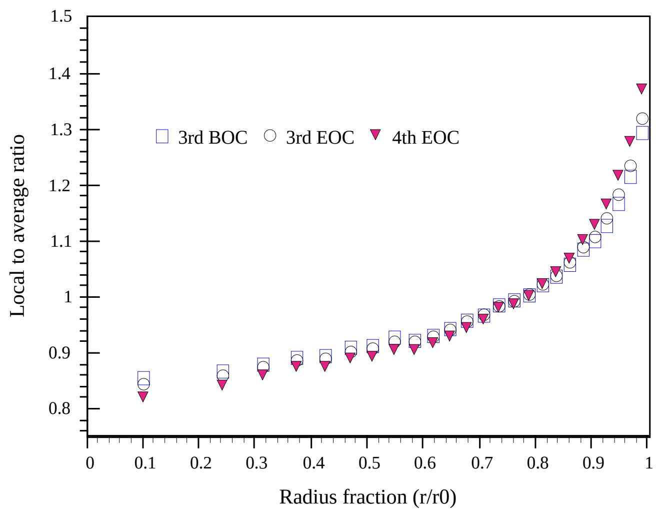

Figure 6. MCWO-calculated detailed radial fission power density profiles at the 3rd BOC, 3rd EOC, and 4th EOC.

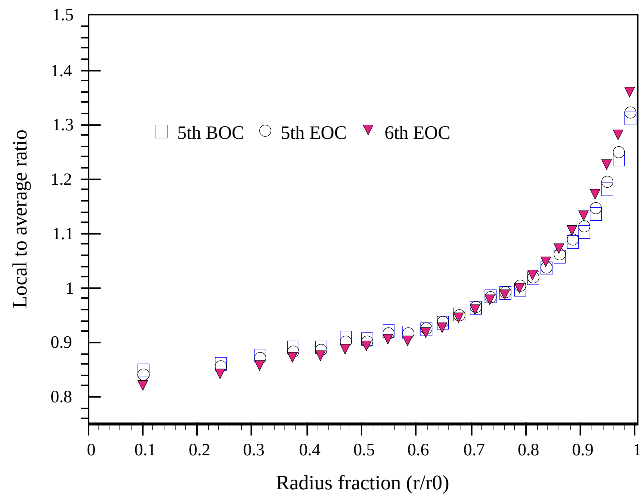

Figure 7. MCWO-calculated detailed radial fission power density profiles at the 5th BOC, 5th EOC, and 6th EOC.

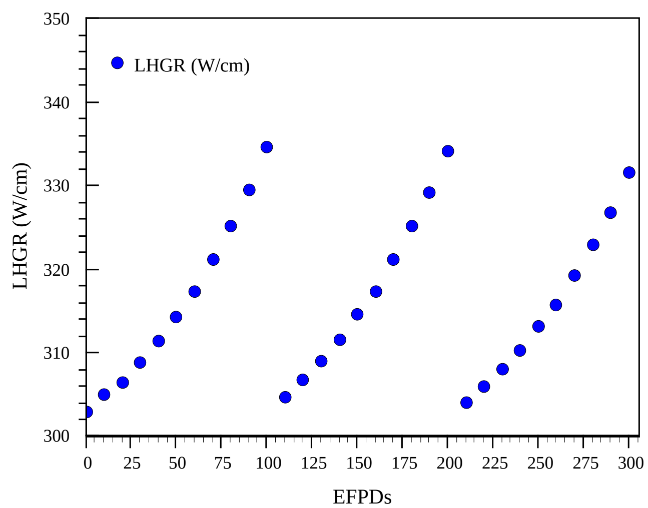

Figure 8. MCWO-calculated LHGR versus EFPDs from the 2nd BOC to the 6th EOC for a total of 300 EFPDs.

4.4. Cd-filter Depletion Impacts on the Detailed OXIDE Fuel Radial Fission Power Profiles versus EFPDs

The MCWO-calculated AFC-2D oxide rodlet-3 results are normalized to an ATR E-lobe power of 23 MW. The results of the oxide fuel rodlet burnup analysis performed provide the detailed relative radial fission power profiles at the 1st beginning of cycle (BOC), 1st end of cycle (EOC), 2nd EOC, 3rd BOC, 3rd EOC, 4th EOC, 5th BOC, 5th EOC, and 6th EOC for a total of 300 EFPDs. The MCWO-calculated relative radial fission power profiles of the oxide fuel rodlet are shown in Figs. 9, 10, and 11.

As expected, since the oxide fuel has lower fuel density and higher oxygen neutron moderation power than the metal fuel, the oxide detailed radial fission power density profiles shows a flatter pattern than the metal fuel. The oxide peak L2AR increases from 1.178 (1st BOC) to 1.265 (6th EOC).

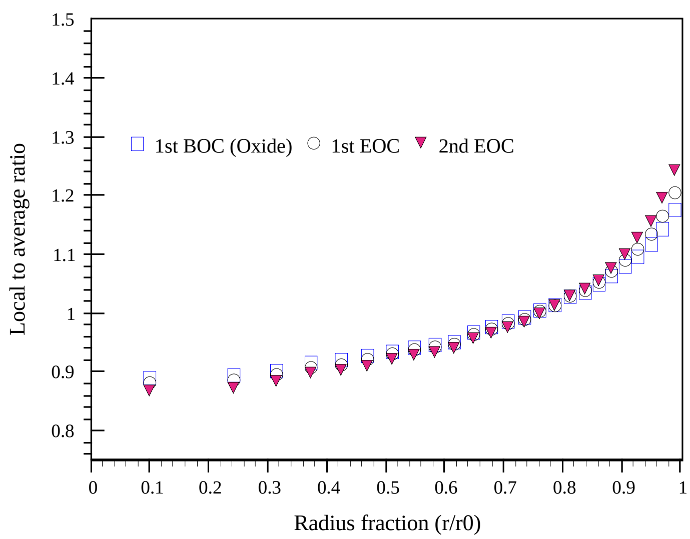

Figure 9. MCWO-calculated AFC-2D detailed radial fission power density profiles at the 1st beginning of cycle (BOC), 1st end of cycle (EOC), 2nd EOC.

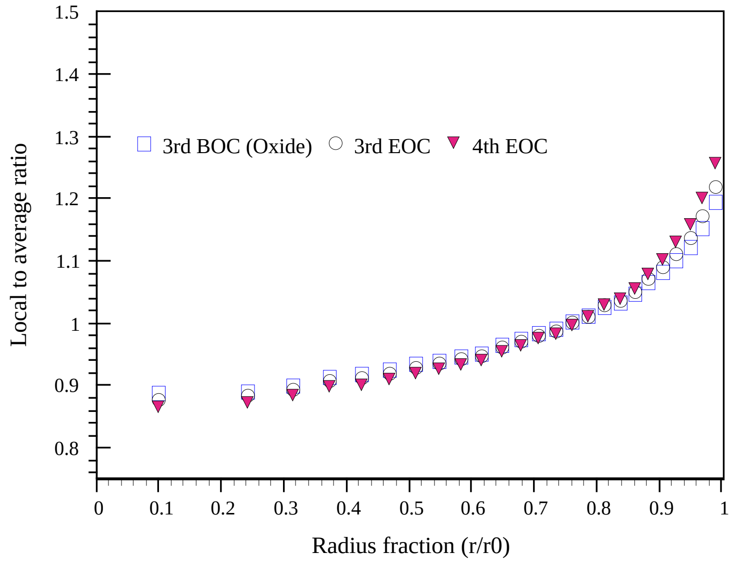

Figure 10. MCWO-calculated AFC-2D detailed radial fission power density profiles at the 3rd BOC, 3rd EOC, and 4th EOC.

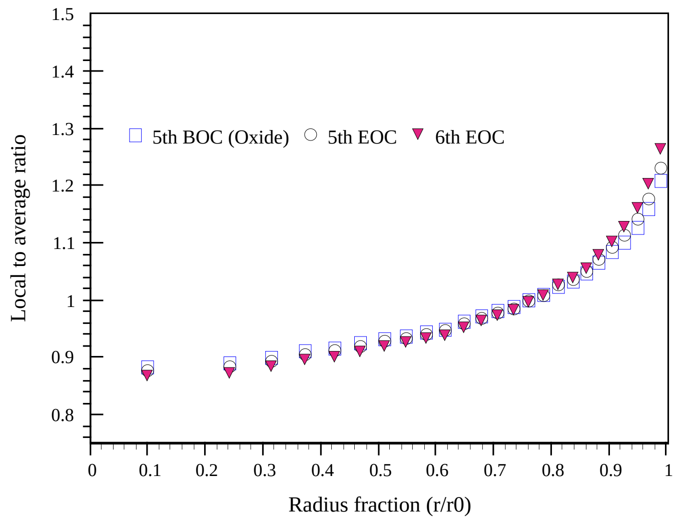

Figure 11. MCWO-calculated AFC-2D detailed radial fission power density profiles at the 5th BOC, 5th EOC, and 6th EOC.

5. 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 demonstrates that the MCWO method could provide the needed accurate neutronics parameters. The results show that the fission power profile differences between the thermal and fast neutron flux are quite significant. The ratios of the metal fission power in the outermost rodlet shell for the thermal and fast neutron flux are 2.34, and 1.01, respectively. However, the installed Cd-filter can harden the neutron flux spectrum and reduce the metal ratio of fission power in the outermost rodlet shell from 2.34 to 1.23, which can satisfy the AFC-1 experiment need. The MCWO-calculated metal and oxide peak L2AR at 1st BOC and 6th EOC are 1.235 and 1.359; 1.178 and 1.265, respectively.

The MCWO-calculated burnup and fission heat rate distributions, and Cd filter depletion of the proposed AFC-1 versus 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 requirements. In addition, the Cd-filter can also reduce and maintain the rim-effect in the radial fission power profile throughout the fuel irradiation life. It is recommended that the developed MCWO and Cd depletion procedures apply to the AFC-2 series experiments to provide the best estimate detailed radial fission power density profile versus EFPDs.

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