

Investigation of Changes in Europium Isotopic Ratios Following Irradiation

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INTRODUCTION

Studies are planned to evaluate extending the cycle length of the High Flux Isotope Reactor (HFIR) by increasing the ^{235}U content of the fuel elements. One part of these studies will be the evaluation of the regulating and safety element worths for modified fuel elements.

The HFIR reactor is composed of two, concentric annuli of highly enriched uranium (HEU) fuel, surrounded radially by a beryllium reflector. The regulating and safety elements for the reactor are located in an annulus between the HFIR core and reflector and contain three regions: a “black” strong neutron-absorber region containing Eu_2O_3 dispersed in an aluminum matrix; a “gray” moderate neutron-absorber region with tantalum particles in an aluminum matrix; and a “white” region (or follower) of perforated aluminum.

RESULTS

Reference 1 provides isotopic data for the europium region of the control element after an irradiation interval of 48,615 MWd. These data are presented in Table 1 in the column identified as “measured”. The gadolinium content and europium isotopics present at the measured location (0.5 in. from the Eu/Ta interface) were calculated by using a two dimensional, r-z computational model of the reactor core (the DORT program, a discrete ordinates method) and cross sections derived from the ENDF/B-V data library³. The thermal flux (energy <0.5 eV) at the centerline of the europium region, 0.5 in. from the Eu/Ta interface was taken from the DORT calculation for a beginning-

of-life configuration and had a magnitude of $1.50(10^{12})$ neutrons/(cm²·s) for 85 MW. Note that while the position of the control elements varies over the lifetime of a fuel cycle, this irradiation level corresponds to 24 fuel cycles. Consequently the assumption of an “average constant” flux seems reasonable. Table 1 shows the calculated europium/gadolinium fractions, europium isotopics, and “neutrons absorbed per initial Eu atom” after an irradiation period of 48,615 MWD. Also shown in Table 1 are two parametric studies in which the thermal flux was varied to first match the reported “neutrons absorbed per initial Eu atom” and then varied to match the measured Eu/Gd fraction.

No information regarding the size of the sample analyzed is known. The variation in flux to match “measured” quantities is within the spatial variation that occurs, radially and axially, across small distances in the control elements. Nevertheless, these variations in flux do not explain the differences among measured and calculated isotopics.

Though calculation-to-experiment agreement shown in Table 1 is poor, experience at the reactor has shown that the worth of the europium region is generally independent of burnup. This is because even though the distribution among isotopes and even elements seems not to be well-known, isotopes of both elements are strong absorbers. Furthermore, those portions of the europium that are 2 in. or more from the Eu/Ta interface receive relatively little neutron fluence, and the europium isotopics are essentially unchanged at EOL (ref. 2). Consequently the shutdown margin of the

reactor is essentially unaffected by irradiation.

CONCLUSIONS

Differences between measured and calculated control plate isotopics are significant and variations in assumed, constant, average flux input to the depletion calculation do not explain the differences among measured and calculated isotopics. Potential future modifications to the HFIR fuel element design to increase the ^{235}U loading, with consequent changes in element reactivity, coupled with the poor ability to predict Eu consumption would seem to provide justification for further review of the europium cross section data. A new evaluation and/or additional experimental measurements seem justifiable.

REFERENCES

1. R. W. Knight and A. E. Richt, "Evaluation of Absorber Materials Performance in HFIR Control Cylinders," *Nuclear Technology*, **15**, 384–390 (September 1972).
2. R. T. Primm III, *Reactor Physics Input to the Safety Analysis Report for the High Flux Isotope Reactor*, ORNL/TM-11956, Oak Ridge National Laboratory, March 1992
3. N. M. Greene, J. W. Arwood, R. Q. Wright, and C. V. Parks, *The LAW Library—A Multigroup Cross-Section Library for Use in Radioactive Waste Analysis Calculations*, ORNL/TM-12370, Oak Ridge National Laboratory, August 1994

Table 1. Comparison of calculated-to-measured physics parameters

Parameter		Measured (ref. 1, 0.5 in. from Eu/Ta interface)	Calculated [assumed thermal flux, $\text{n}/(\text{cm}^2 \cdot \text{s})$ at 0.5 in. from Eu/Ta interface]		
			$1.50 \times 10^{12}^a$	$5.40 \times 10^{12}^b$	$3.00 \times 10^{12}^c$
Fraction (wt %)	Eu	86.5	93.4	74.7	86.4
	Gd	13.5	6.6	25.3	13.6
Europium isotopics (at. %)	^{151}Eu	5.20	33.28	12.95	23.07
	^{152}Eu	8.16	8.44	9.91	10.92
	^{153}Eu	66.92	47.85	48.71	47.58
	^{154}Eu	13.77	9.74	25.40	16.85
	^{155}Eu	5.94	0.59	1.87	1.18
Neutrons absorbed per initial europium atom		1.07	0.29	0.998	0.56

^aFlux from DORT calculation for beginning-of-life configuration.

^bFlux to yield reported neutrons absorbed per initial europium atom.

^cFlux that yields measured Gd/Eu ratio.

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*Presented at the Winter Meeting of the
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This study is an outgrowth of work presented at the PHYSOR 2004 conference.



- High Flux Isotope Reactor (HFIR)
 - Neutron scattering research
 - Isotope production
 - Neutron activation analyses
 - Uses Ta/Eu-based control elements

The Physics of Fuel Cycles and Advanced Nuclear Systems: Global Developments



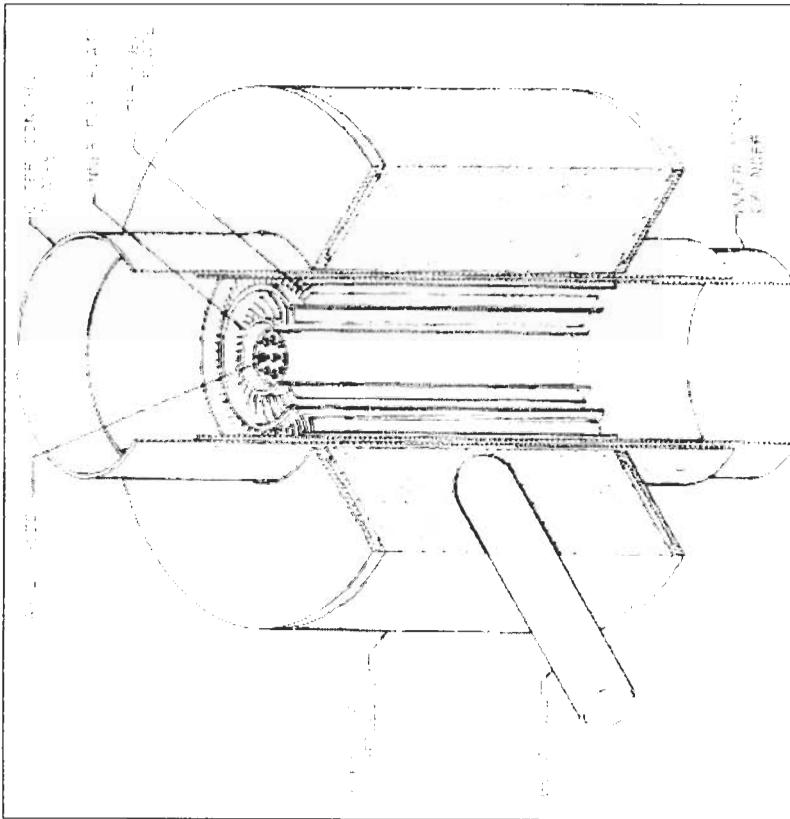
April 25-29, 2004, Chicago, IL USA

➤ Have made first change to control element material density since operation began in 1965 – reduced Ta content

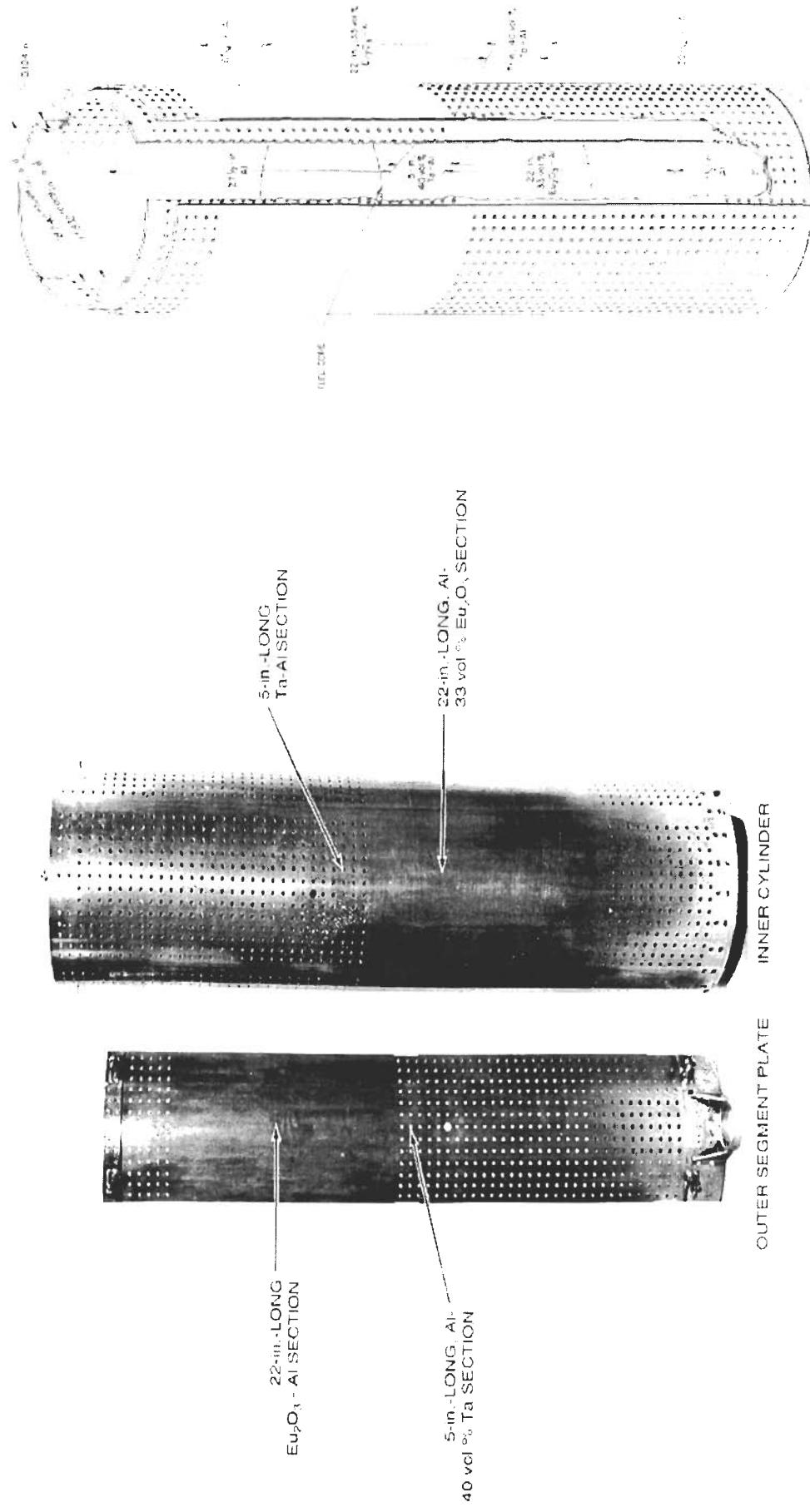
➤ During Ta studies, discovered Eu-related anomalies when comparing measurements to calcs

Reactor control elements are located between the exterior of the fuel and the beryllium reflector.

- Compact core— high-power density
- Flux-trap design
- Concentric cylinders
 - Target
 - Fuel
 - Control
 - Reflector
- Upper (or outer or safety) “cylinder” actually has four, independently moving quadrants
- Lower (or inner or control or regulating) “cylinder” maintained in symmetric configuration to upper but not required



Control Plate and Cylinder



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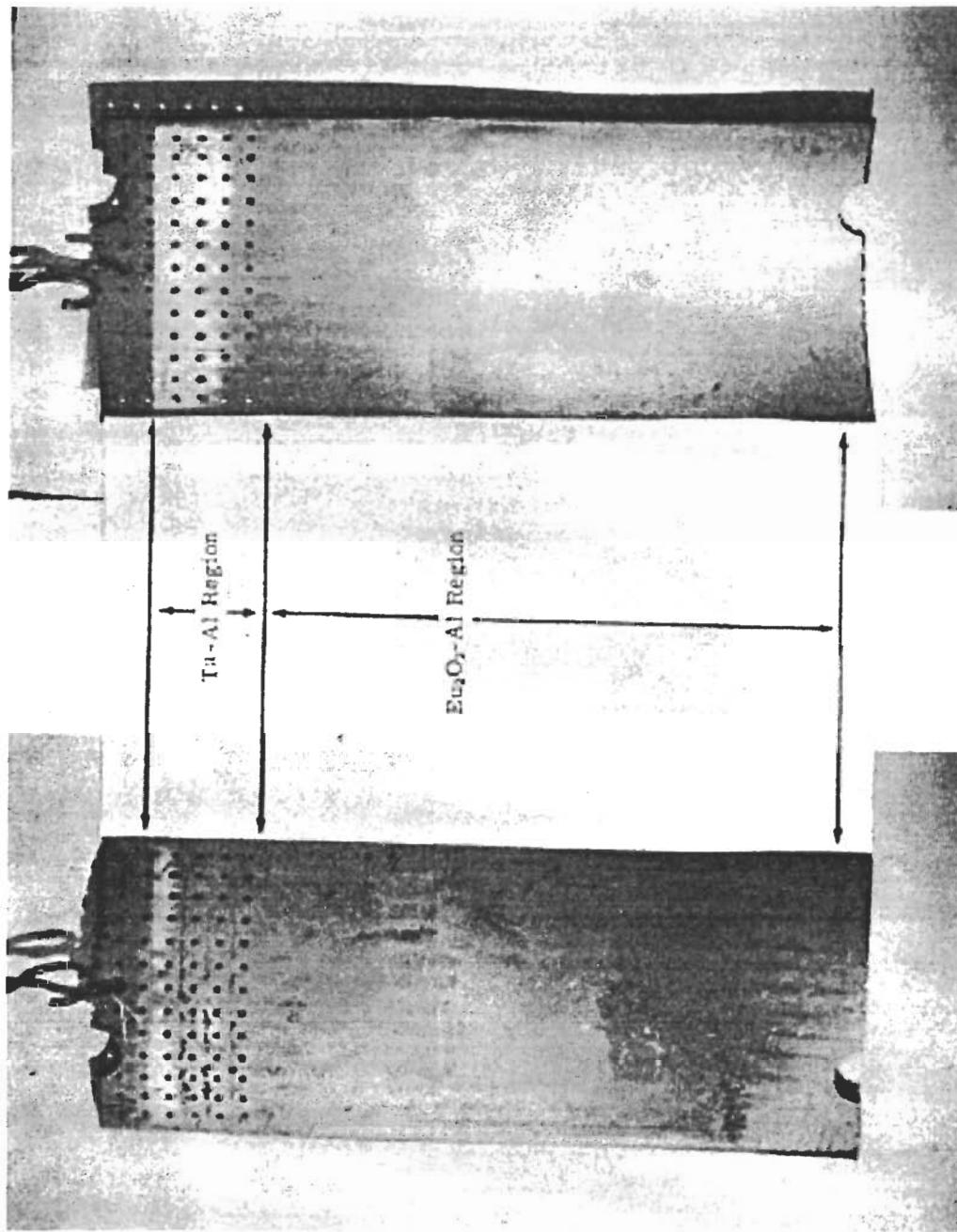
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PHYSOR 2004 study used destructive evaluation as benchmark

- R. W. Knight and A. E. Richt, “Evaluation of Absorber Materials Performance in HFIR Control Cylinders,” *Nuclear Technology*, 15, 384–390 (September 1972).
- Control plate destructively assayed
- 48,615 MWd; about $\frac{1}{2}$ of lifetime; about 2 years of irradiation
- Gd and Eu isotopics measured at $\frac{1}{2}$ inch from interface; size of sample not stated
- Single Ta measurement agreed well with calc

Portion of inner cylinder analyzed

(other samples away from interface measured; not of interest)

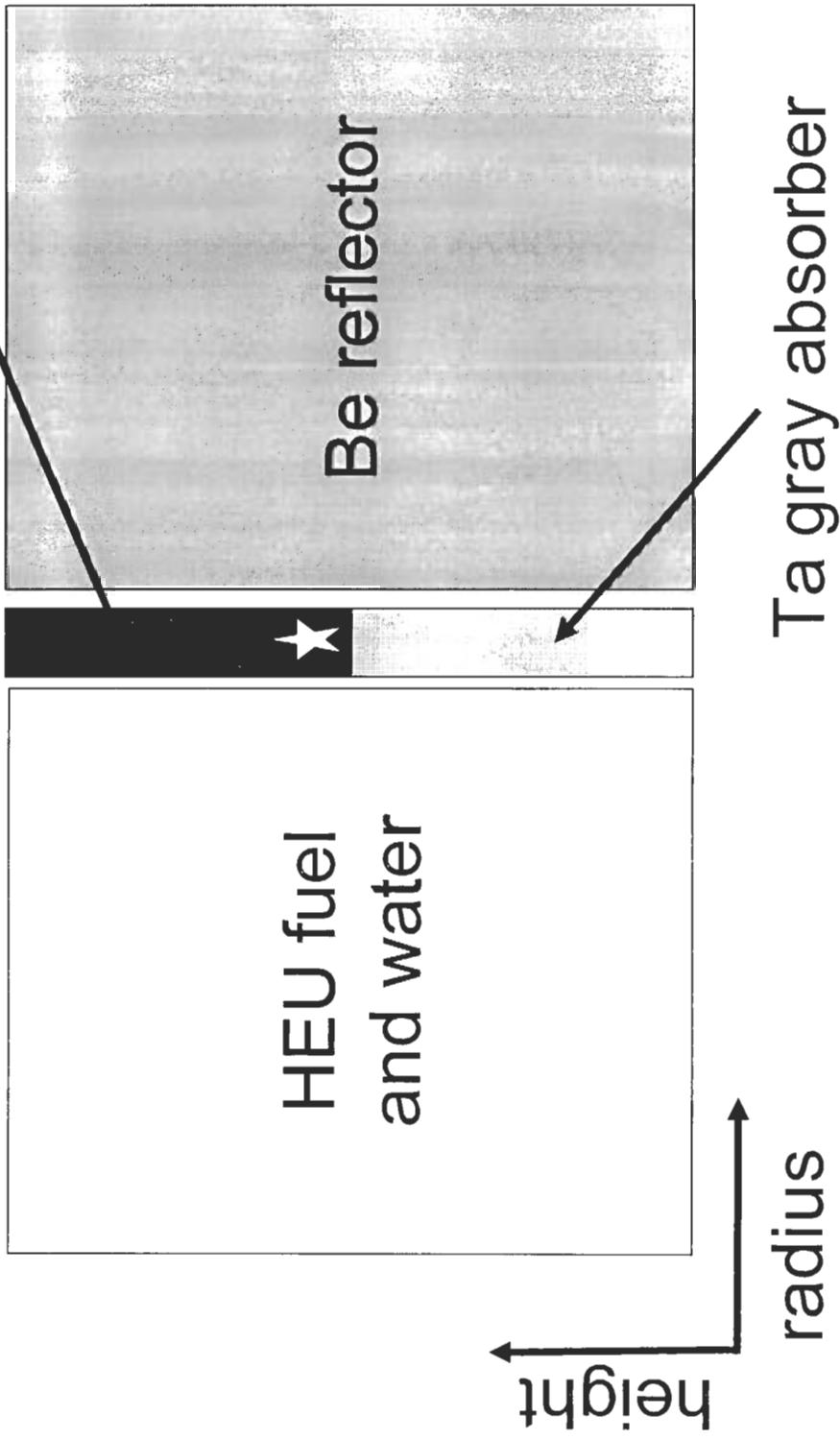


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Spatially complex problem to calculate - 1.3 cm from interface with Ta (★)

Eu black absorber - 0.48 cm thick



Under irradiation, natural Eu transmutes to other strong absorbers

Gd152	Gd153 267.0	Gd154 218	Gd155 14.00	Gd156 20.47	Gd157 15.66
0.20 1.1E140	0.2E4 0.00	0.6E1 0.00	0.6E2 0.00	0.254E3 0.00	0.254E3 0.00
0.20 1.1E140	0.2E4 0.00	0.6E1 0.00	0.6E2 0.00	0.254E3 0.00	0.254E3 0.00
0.20 1.1E140	0.2E4 0.00	0.6E1 0.00	0.6E2 0.00	0.254E3 0.00	0.254E3 0.00
0.20 1.1E140	0.2E4 0.00	0.6E1 0.00	0.6E2 0.00	0.254E3 0.00	0.254E3 0.00
Eu151 67.0	Eu152 13.0	Eu153 6.2	Eu154 0.590	Eu155 4.76	Eu156 16.2
0.20 1.1E140	0.2E4 0.00	0.6E1 0.00	0.6E2 0.00	0.254E3 0.00	0.254E3 0.00
0.20 1.1E140	0.2E4 0.00	0.6E1 0.00	0.6E2 0.00	0.254E3 0.00	0.254E3 0.00
0.20 1.1E140	0.2E4 0.00	0.6E1 0.00	0.6E2 0.00	0.254E3 0.00	0.254E3 0.00
0.20 1.1E140	0.2E4 0.00	0.6E1 0.00	0.6E2 0.00	0.254E3 0.00	0.254E3 0.00

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Depletion analysis achieved by using DORT fluxes (ψ_{DORT}) in ORIGEN

- DORT (3.2) 2D discrete ordinates r-Z model of system, 44 group cross-sections (p3) collapsed from 238 group ENDF/B-V, S8 and S16; ~9000 mesh cells; ~20 min/run, 600 MHz DEC alpha
- Time dependence - 24 fuel cycles
- Constant flux modeled in ORIGEN; no delay between cycles

Justification for a constant flux in ORIGEN

- Resurrect VENTURE model from 1992. "Prediction and Measurement of Fuel Depletion Due to Irradiation of HFIR Fuel Elements," *Annu. Meet. Am. Nucl. Soc., Boston, June 7-12, 1992.*
- For BOC, DORT thermal (<0.5 eV) flux = $1.50(10^{12})$, VENTURE = $2.44(10^{12})$ at tip
- VENTURE showed thermal flux increases from BOC value for days 2-14 of cycle
- Eu region exists core at 17 days – 35% of cycle left; thermal flux drops to zero, effectively.
- Best estimate from VENTURE = $3.4 * \text{DORT BOC flux}$

All measured parameters cannot be matched simultaneously.

Parameter	Measured		Calculated [assumed thermal flux, n/(cm ² .s) at 0.5 in. from Eu/Ta interface]	
	Fraction (wt. %)		1.50 (10 ¹²) DORT BOC	5.40 (10 ¹²) VENTURE mod DORT BOC
Eu	86.5	93.4	74.7	86.4
Gd	13.5	6.6	25.3	13.6
¹⁵¹ Eu	5.20	33.28	12.95	23.07
¹⁵² Eu	8.16	8.44	9.91	10.92
¹⁵³ Eu	66.92	47.85	48.71	47.58
¹⁵⁴ Eu	13.77	9.74	25.40	16.85
¹⁵⁵ Eu	5.94	0.59	1.87	1.18
Neutrons abs/ Initial Eu atom	1.07	0.29	0.998	0.56

Why look further?

- Eu is an excellent poison for low maintenance, high-burnup reactor concepts; 40 years of experience at HFIR
- Future possible HFIR fuel modifications (larger fuel loading; alternative fuel compositions) will require assessment of impact on control element worth
- Eu was used in recent irradiation experiments to tailor flux spectrum. Irradiated specimens of the “right size” (not a six foot long control plate) exist in the HFIR storage pool