A PWR Thorium Pin Cell Burnup Benchmark

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ABSTRACT

As part of work to evaluate the potential benefits of using thorium in LWR fuel, a thorium fueled benchmark comparison was made in this study between state-of-the-art codes, MOCUP (MCNP4B + ORIGEN2), and CASMO-4 for burnup calculations. The MOCUP runs were done individually at MIT and INEEL, using the same model but with some differences in techniques and cross section libraries. Eigenvalue and isotope concentrations were compared on a PWR pin-cell model up to high burnup. The eigenvalue comparison as a function of burnup is good: the maximum difference is within 2% and the average absolute difference less than 1%. The isotope concentration comparisons are better than a set of MOX fuel benchmarks and comparable to a set of uranium fuel benchmarks reported in the literature. The actinide and fission product data sources used in the MOCUP burnup calculations for a typical thorium fuel are documented. Reasons for code vs code differences are analyzed and discussed.

1 INTRODUCTION

There has been a revival of interest in the use of thorium in light water reactors because it can facilitate realization of longer intra-refueling intervals and high burnup while reducing spent fuel weapons usability and increasing in-repository durability. These prospects motivated the present work under the strategic nuclear research consortium between MIT and INEEL, and a subsequent separate DOE NERI award, focusing on the use of thorium-based fuel in otherwise-conventional, retrofittable, PWR fuel assemblies.

In 1960-1980, fueling of LWRs with thorium was actively explored, including whole-core demonstrations in Indian Point I, Elk River, and the Shippingport Breeder[1]. Thorium use was also extensively studied in the NASAP and INFCE programs but work was focused on recycle mode fuel cycles, using highly enriched U-235 for startup, and burnup ~ 30 MWd/Kg. However, circumstances have changed since then: once-through fueling is assumed; a <20wt% 235U anti-proliferation limitation has been imposed; and a discharge burnup approaching 60 MWd/Kg has been achieved in uranium-fueled LWRs, with further increases in prospect. In addition, an entirely new generation of codes and cross section sets has become available. Thus, we have initiated a new round of computational benchmarks reflecting these new realities.

The zero leakage and poison-free pin cells were burned to in excess of 70 MWd/kg at constant power using two code packages: CASMO-4 and MOCUP (which combines MCNP4B and ORIGEN2). The subject programs are well known and state-of-the-art.

2 METHODS

CASMO-4[2] is a multigroup transport code for burnup calculations on LWR assemblies or simple pin cells. The cross-section production is co-ordinated with the requirements of the reactor analysis code SIMULATE-3, but the cross-sections can also be used in other codes. CASMO-4 and its predecessor CASMO-3 can handle all known LWR fuel designs from commercial fuel vendors. The calculation can be done with default burnup steps, or specified steps. In addition to the standard library, several other libraries exist for CASMO-4. In this study, the 70-group library provided with the code by Studsvik of America has been used. It is based on the evaluated data files JEF-2.2 and ENDF/B-6, which were developed at the OECD/NEA Data Bank and Brookhaven National Nuclear Data Center respectively.

MOCUP[3] is the MCNP-ORIGEN2 Coupled Utility Program. It employs the MCNP (here version 4B) generalized-geometry Monte Carlo transport code to give the neutronics solution and the ORIGEN2 code to compute the time-dependent compositions of the individually selected MCNP cells. All data communication between the two codes is accomplished through the MCNP and ORIGEN2 input/output files. This allows a general material (target, fuel, control, etc.) to be depleted in a neutral particle field, with the accuracy of a transport neutronics solution. Since the MCNP version 4B library does not contain temperature-dependent neutron cross sections of most actinides, a number of libraries from the UTXS compilation were imported. Also for some fission products, the evaluated data files produced at Los Alamos National Laboratory were imported via INEEL.

3 Pin Cell Model

The work reported here involves analysis of a PWR pin cell excised from a standard 17x17 pin assembly typical of large Westinghouse PWRs. The usual all-UO_2 fuel pellets were replaced by a ThO_2-UO_2 mixture at 94% of theoretical density consisting of 75w/o Th, 25 w/o U on a heavy metal basis, with the latter enriched to 19.5 w/o U-235, to give an overall enrichment of 4.869 w/o U-235 in total heavy metal.

Fig. 1 shows the pin cell model representing the unit lattice cell of a Westinghouse PWR fuel bundle. The burnup calculations described in this study are based on this model. Since all actinides in the thorium and uranium chains will be produced during burnup, this calculation is a challenge to all the actinide neutron libraries used by MCNP, and also a challenge to CASMO's treatment of each actinide's group constants.

Table 1 and Table 2 show detailed parameters of the pin-cell model for a Westinghouse PWR assembly. Parameters at hot full power were used in our benchmark calculations.

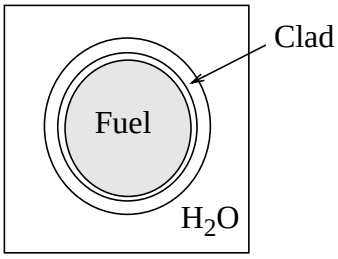

Fig.1 Pin-cell Model

Table 1: Pin-cell Model Parameters

Parameter	Cold Zero Power	Hot Full Power
Fuel Temperature (˚K)	300	900
Power Density (KW/KgHM)	0.0	38.1347
Power Density (KW/liter cell)	0.0	107.284
Fuel Density (g/cm3)	9.614	9.424
Cladding Temperature (˚K)	300.0	621.1
Cladding Density (g/cm3)	6.550	6.505
Coolant Pressure (bars)	155.13	155.13
Coolant Temperature (˚K)	300.0	583.1
Coolant Density (g/cm3)	1.003	0.705
Fuel Pellet Radius (mm)	4.096	4.1274
Cladding Inner Radius (mm)	4.178	4.1896
Cladding Outer Radius (mm)	4.750	4.7609
Pin Pitch (mm)	12.6	12.626

Table 2: Initial Compositions (at Hot Full Power Conditions)

	Nuclide	Weight Percent (%)	Number Density($1/cm^3$)
Fuel	Th-232	65.909	1.61215E+22
	U-234	0.034	8.24518E+18
	U-235	4.291	1.03615E+21
	U-238	17.740	4.22957E+21
	O-16	12.026	4.26835E+22
Cladding	Zr-4 (Zircaloy-4)	100	4.31438E+22
Coolant	H-1	11.19	4.71053E+22
	O-16	88.81	2.35662E+22

4 Results and Analyses

Fig. 2 shows the comparison of eigenvalue history between the two codes and Fig. 3 shows MOCUP's difference from CASMO. Table 3 shows values of this comparison at selected time steps. Considering that the point of major concern is the burnup value where reactivity reaches 0.03, (which is representative of an n-batch core-average end of cycle value, with allowance of 3% reactivity loss for leakage), this eigenvalue comparison shows almost no difference at that point. This is encouraging because one must achieve better accuracy for thorium fueled cores than for all-uranium fueling (since the slope of k vs burnup is less steep) to achieve equal accuracy in cycle length estimates. At 60 MWd/kg (corresponding to projected end-of-life core average burnup), the eigenvalue difference increased to approximately 0.015. INEEL's MOCUP result had about 0.02 lower K-inf at the first point but agrees well with MIT's results later because the different cladding compositions used in the INEEL model acted as a small amount of "burnable poison". Notice that there is a weak oscillation in the MOCUP curve. This is typical of the MOCUP system. Reference[4] discusses how to decrease this oscillation and how to improve the MOCUP and CASMO comparison. Some other differences between these two MOCUP runs and techniques adopted to improve the comparison will be discussed in section 5 of this paper.

Table 4 shows the isotope concentration comparison at 60.749MWd/Kg which is at the upper limit of discharge burnup if a 3-batch core refueling scheme is considered. Uncertainties in number densities of a recent extensive MOX fuel pincell benchmark[5] and a uranium fuel pincell benchmark[6] are shown in the last two columns. The MOX and all-U benchmark "uncertainties" are the largest spread for a dozen or so contributions, while our results are single code vs code differences. All-in-all, it appears that our results agree as well or better than these other contemporary comparisons. One observation is that the concentrations of thorium chain actinides calculated by MOCUP are almost all larger than those of CASMO-4 by about 4%, whereas the concentrations of uranium chain actinides are on average smaller by 4%. Further calculations show that increasing the initial Th-232 concentration by 2% and at the same time decreasing U-238 concentration by the same amount in CASMO eliminated roughly half of the differences. This suggests that refinement in the thorium and uranium cross section sets (or in CASMO the equivalence relation used for self-shielding) should be looked into. Total end-of-life heavy metal destruction was about 1% higher in MOCUP, which supports the conclusion that overall average energy per fission plus capture differs slightly between the two codes. This is a well-known dilemma, since few codes disaggregate the capture contribution: use of a constant overall approximation of ~8(υ-1) MeV is common. This means that MOCUP will grind through more of the nuclide chains for the same EFPD. Another point of interest is the large difference in U-234 concentrations, which merits further attention, even though this nuclide has a small effect on k.

Table 3: Comparison of Eigenvalues for the Pin-cell Model

Burnup (MWd/kg)	CASMO-4	MOCUP	INEEL MOCUP
0.000	1.23782	1.23354	1.22347
0.114	1.20071	1.19708	1.18051
5.835	1.14828	1.14466	1.13563
10.411	1.12108	1.11662	1.11325
19.563	1.07245	1.07154	1.06648
31.004	1.02014	1.02168	1.01906
40.156	0.98190	0.98453	0.98514
49.308	0.94636	0.95383	0.95035
51.596	0.93817	0.94477	0.94063
60.749	0.90701	0.91851	0.91447
72.189	0.87348	0.88449	0.87942

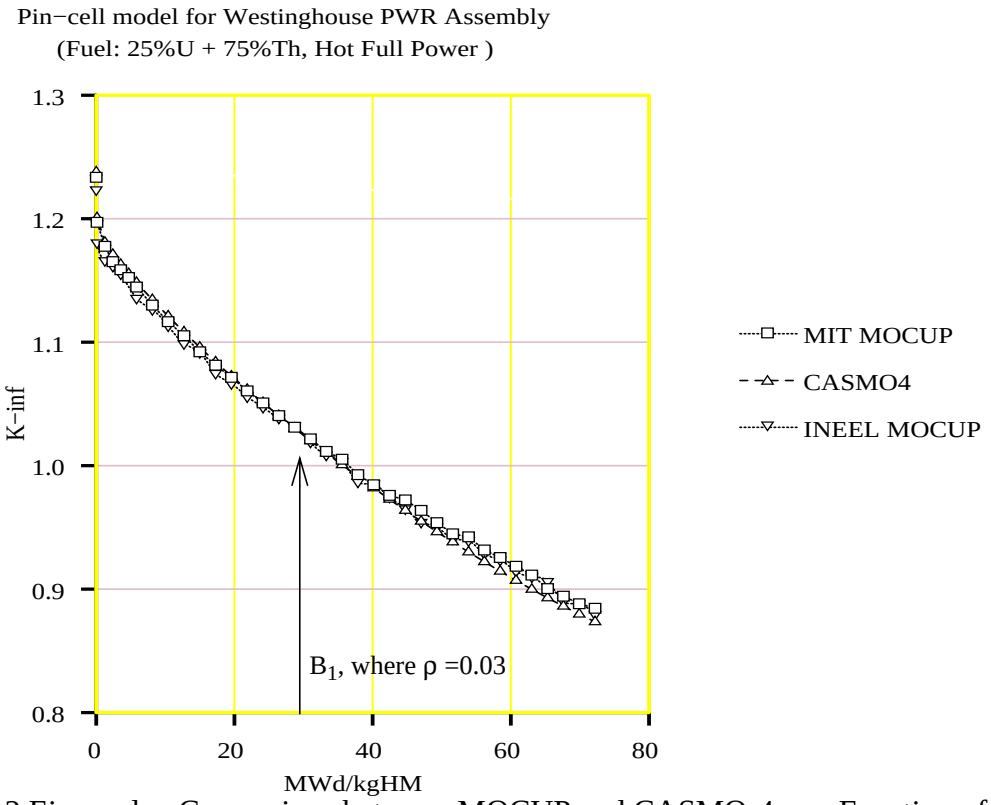

Fig. 2 Eigenvalue Comparison between MOCUP and CASMO-4 as a Function of Burnup

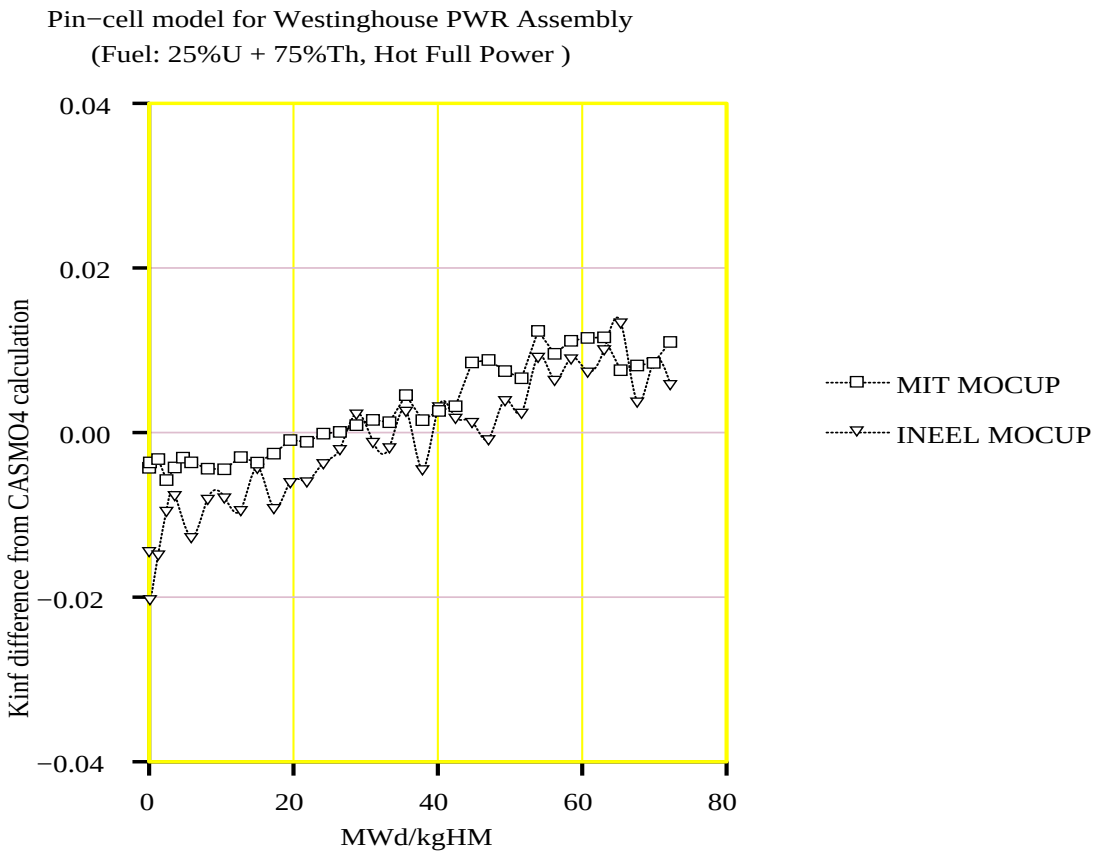

Fig.3 Eigenvalue Difference from CASMO-4 as a Function of Burnup

Table 4: Fractional Difference(1) in Isotope Concentration @ 60.749MWd/Kg

Isotopes	CASMO-4	MIT MOCUP	INEEL MOCUP	Uncertainties for MOX fuel[5]	Uncertainties for U fuel[6]
Th-232	1.53769e+22	-0.003	-0.003	NA	NA
Pa-231	1.70440e+18	0.048	0.018	NA	NA
Pa-233	1.95229e+19	0.035	0.045	NA	NA
U-232	1.56006e+18	0.034	-0.003	NA	NA
U-233	2.74202e+20	0.040	0.044	NA	NA
U-234	5.15172e+19	0.176	0.174	0.67	0.0899
U-235	1.78104e+20	-0.021	-0.033	0.03	0.0812
U-236	1.39420e+20	0.054	0.057	0.09	0.026
U-238	3.88419e+21	0.004	0.003	0.001	0.0021
Np-237	1.82660e+19	-0.058	-0.035	0.34	0.0942
Np-238	5.46097e+16	-0.037	-0.096	NA	NA
Np-239	7.61806e+17	-0.043	-0.019	NA	NA
Pu-238	8.90932e+18	-0.026	-0.099	0.36	0.1386
Pu-239	5.37090e+19	-0.071	-0.050	0.08	0.0712
Pu-240	1.82233e+19	-0.032	0.021	0.10	0.0527
Pu-241	1.90707e+19	-0.024	-0.041	0.04	0.0686
Pu-242	9.96772e+18	-0.036	0.027	0.22	0.0839
Total Fissile	7.54683e+20	0.024	0.026	--	--
Total Actinide Depleted(2)	0.062601217	0.010	0.013	--	--
Fertile Transformed(3)	0.0535	0.029	0.034	--	--
Ratio of Th232 to U238 Depletion(4)	2.15589	0.107	0.095	--	--

(1) Fractional Diff. = $(N - N_{Casmo4})/N_{Casmo4}$, where N is nuclide concentration

(2) Total Actinide Depleted = $(N_{Actinide,t} - N_{Actinide,0}) / N_{Actinide,0}$, where $N_{Actinide,t}$ is the total amount of actinides at time t; $N_{Actinide,0}$ is the total amount of actinides at time 0.

(3) Amount of Fertile Transformed = $(N_0-N_t)/N_0$

N_0 = Initial Fertile amount N_t = Fertile amount at time t.

(4) Ratio = (Th-232 depleted) / (U-238 depleted)

5 Discussion

Some techniques for increasing the accuracy of MOCUP such as increasing the number of histories in MCNP are described in Ref.[4]. Two additional techniques were employed in this study to improve the benchmark comparison.

Based on ORIGEN output, the actinides and fission products were sorted according to absorption fraction at around EOL burnup, and the actinides and fission products which account for the majority of absorption in fuel were then designated as tracked nuclides in MOCUP. Experience shows that this is important in improving MOCUP's accuracy at high burnup.

As shown in Table 5, the chosen 17 actinides account for 99.9% of the neutron absorption of all actinides, and from Table 6, the chosen 41 fission products account for 97.3% of the absorption of all fission products. The absorption of all fission products is 15% of all absorptions, so the part that is excluded in this study is: (1-99.9%)85%+(1-97.3%)15% = 0.49%. This can explain approximately one-third of the reactivity discrepancy seen at the end of the curves in Fig. 2 and Fig. 3. The difference when collapsing the one group constants for burnup may cause the difference in slope, because CASMO uses a buckling adjustment to get the constants under critical conditions but MOCUP uses the constants determined under non-critical conditions. A detailed discussion can be found in Ref.[8].

The numbers under column "zaid" are the identification numbers that MCNP uses to get the corresponding library during execution, and the "library name" is the file name used by MCNP to store those libraries. In the "source" column, MCNP means that the library comes with MCNP, and can ultimately be traced back to ENDF/B-V, or ENDF/B-VI, or LLNL, or T2. ENDF/B-V and ENDF/B-VI are the Evaluated Nuclear Data Files, an effort coordinated by the National Nuclear Data Center at Brookhaven National Laboratory. LLNL represents the evaluated nuclear data libraries compiled by the Nuclear Data Group at Lawrence Livermore National Laboratory and T-2 represents the libraries from the Nuclear Theory and Applications group T-2 at Los Alamos National Laboratory. UTXS indicates that the library comes from UTXS compilation which can be found at http://radon.me.utexas.edu/. The UTXS library was prepared at The University of Texas at Austin using the NJOY processing code. At UTXS, all evaluated nuclear data files used were obtained from National Nuclear Data Center (NNDC), Brookhaven National Laboratory (BNL) and Los Alamos National Laboratory, and when available, ENDF/B-6 was used. INEEL represents the libraries evaluated at Los Alamos National Laboratory or Oak Ridge National Laboratory but which came through INEEL. INEEL MOCUP runs used the same actinide and fission product list with some exceptions on library sources as shown in table 7. For minor actinides and all the fission products, neutron cross section libraries at room temperature were used.

The second technique for improving the eigenvalue comparison involved using ORIGEN to do the power normalization automatically. The flux from MCNP is normalized per source neutron. In order to get the absolute value of flux, we have to calculate the normalization factor in terms of power level, energy per fission, etc. This can be done exogenously and approximately by using an average value of various parameters such as 202Mev/fission for fission energy. Alternatively, this work actually can be done by ORIGEN, since it has a built-in function for calculating flux from power. After a small modification of MOCUP at MIT, this work was transferred to ORIGEN,

where this normalization is computed in a more complex and accurate way[7]. Table 8 shows the energy per fission used by ORIGEN and by CASMO. Notice that the difference between ENDF6 and the codes is that the energy release from capture reactions such as (n,γ) are included in the codes as "fission energy". The maximum difference between ORIGEN and CASMO for any isotope is less than 5%. For U-235, in which the majority of fissions occur, the difference is only 0.25%. The INEEL MOCUP run used an average value of 202Mev/fission, which is also very close to the fission energy of U-235 shown in Table 8. The differences in fission energy used in different codes or methods caused small differences in total actinide depleted shown in Table 4. It should be noted that in the case of multi-zone burnup calculation, e.g. assembly burnup, where pin power distribution keeps changing, it is more convenient to calculate the normalization factor in the conventional way, even though a more accurate calculation of fission energy yield is still feasible using the MCNP-calculated fractional neutron loss to fission for all fissile nuclides in the modeled system[8].

Table 5: The Chosen Actinides in MOCUP

Actinide	Absorption Fraction	Zaid	Library Name	Source	Temperature (˚K)
U235	3.088e-01	92235.54c	endf5mt1	MCNP	881
TH232	2.630e-01	90232.86c	th232.900	UTXS	900
U233	1.409e-01	92233.86c	u233.900	UTXS	900
U238	1.126e-01	92238.86c	u238.900	UTXS	900
PU239	9.489e-02	94239.86c	pu239.900	UTXS	900
PU240	2.813e-02	94240.86c	pu240.900	UTXS	900
PU241	1.714e-02	94241.86c	pu241.900	UTXS	900
U236	1.071e-02	92236.86c	u236.900	UTXS	900
PA233	9.743e-03	91233.50c	endf5u	MCNP	294
U234	7.150e-03	92234.86c	u234.900	UTXS	900
NP237	3.042e-03	93237.55c	rmccsa1	MCNP	294
PA231	1.023e-03	91231.60c	endf60	MCNP	294
PU242	9.635e-04	94242.86c	pu242.900	UTXS	900
PU238	4.911e-04	94238.86c	pu238.900	UTXS	900
U232	2.587e-04	92232.60c	endf60	MCNP	294
NP239	1.984e-04	93239.60c	endf60	MCNP	294
NP238	3.665e-05	93238.35c	endl85	MCNP	0
Sum	0.99907635				

Table 6: The Chosen Fission Products in MOCUP

Fission Product	Absorption Fraction	Zaid	Library Name	Source	Temperature (˚K)
XE135	2.105e-01	54135.50c	endf5mt1	MCNP	294
SM149	8.077e-02	62149.50c	endf5u1	MCNP	294
ND143	8.017e-02	60143.50c	kidman	MCNP	294
RH103	7.560e-02	45103.50c	rmccsa1	MCNP	294
PM147	6.948e-02	61147.50c	kidman	MCNP	294
CS133	6.321e-02	55133.50c	kidman	MCNP	294
XE131	6.258e-02	54131.50c	kidman	MCNP	294
TC99	4.128e-02	43099.50c	kidman	MCNP	294
SM152	3.210e-02	62152.50c	kidman	MCNP	294
SM151	3.170e-02	62151.50c	kidman	MCNP	294
ND145	2.683e-02	60145.50c	kidman	MCNP	294
PM148M	2.062e-02	61148.91c	ornlxsb1	INEEL	294
MO95	2.037e-02	42095.50c	kidman	MCNP	294
EU153	1.824e-02	63153.50c	rmccs	MCNP	294
RU101	1.397e-02	44101.50c	kidman	MCNP	294
SM150	1.062e-02	62150.50c	kidman	MCNP	294
PM148	8.831e-03	61148.50c	kidman	MCNP	294
EU154	8.602e-03	63154.50c	endf5u	MCNP	294
CS134	8.536e-03	55134.60c	endf60	MCNP	294
EU155	7.985e-03	63155.50c	kidman	MCNP	294
PR141	7.683e-03	59141.50c	kidman	MCNP	294
MO98	7.473e-03	42098.50c	mason1	INEEL	294
KR83	7.270e-03	36083.50c	rmccsa	MCNP	294
LA139	6.454e-03	57139.60c	mason1	INEEL	294
ZR93	6.159e-03	40093.50c	kidman	MCNP	294
SM147	5.998e-03	62147.50c	kidman	MCNP	294
RH105	5.912e-03	45105.50c	kidman	MCNP	294
AG109	5.880e-03	47109.50c	rmccsa	MCNP	294
CS135	4.690e-03	55135.50c	kidman	MCNP	294

Table 6: The Chosen Fission Products in MOCUP

Fission Product	Absorption Fraction	Zaid	Library Name	Source	Temperature (˚K)
PD105	4.394e-03	46105.50c	kidman	MCNP	294
MO97	4.110e-03	42097.60c	mason1	INEEL	294
I129	3.072e-03	53129.60c	endf60	MCNP	294
CD113	1.883e-03	48113.60c	mason1	INEEL	294
GD157	1.763e-03	64157.50c	endf5u	MCNP	294
XE133	1.692e-03	54133.62c	xe133.300	UTXS	300
ND144	1.650e-03	60144.96c	ornlxsb1	INEEL	294
PD107	1.419e-03	46107.96c	ornlxsb1	INEEL	294
ZR91	1.388e-03	40091.96c	ornlxsb1	INEEL	294
ND148	1.259e-03	60148.50c	kidman	MCNP	294
PM149	6.134e-04	61149.50c	kidman	MCNP	294
EU151	6.024e-05	63151.50c	rmccs	MCNP	294
Sum	0.97281664				

Table 7: The Different Libraries Used in the INEEL MOCUP Run

Nuclide	Zaid	Library Name	Source	Temp(K)
U238	92238.54c	endf5mt1	MCNP	900
Pu239	94239.82c	putemp1	INEEL	900
Pu240	94240.82c	putemp1	INEEL	900
Pu241	94241.82c	putemp1	INEEL	900
Np237	93237.50c	endf5p1	MCNP	294
Pu242	94242.82c	putemp1	INEEL	900
Xe135	54135.54c	endf5mt1	MCNP	900
Cs133	55133.60c	endf60	MCNP	294
Tc99	43099.60c	endf60	MCNP	294
Eu153	63153.60c	endf60	MCNP	294
Ag109	47109.60c	endf60	MCNP	294
Gd157	64157.60c	endf60	MCNP	294
Xe133	54133.60c	mason1	INEEL	294

Table 8: Energy (Mev) per fission for various actinides

Actinide	ENDF6[1]	ORIGEN[2]	CASMO[3]
Th232	188.47	193.41812	NA[4]
Pa233	189.84	197.3529	NA
U233	191.04	200.98224	NA
U234	191.84	201.34208	NA
U235	193.72	201.70114	202.22
U236	194.49	202.05945	202.22
U238	198.06	202.77378	199.73
Np237	196.37	206.11736	202.22
Pu238	197.38	210.23021	199.73
Pu239	199.92	210.6019	209.10
Pu240	199.47	210.97281	209.71
Pu241	201.98	211.34295	210.96
Pu242	201.58	211.71233	210.96
Am241	201.96	215.1551	NA
Am242	202.29	215.53238	210.96

(1) Values provided by Pavel Hejzlar in Ref.[9], based on interpreted ENDF/B-VI files from Nuclear Theory and Application Group T-2 at LANL, for fission only

(2) R_i (Mev/fission) = 1.29927 x $10^{-3}(Z^2A^{0.5})$+33.12; including allowance for capture

(3) from CASMO4 manual, page 9-5, Table 9.5.1.

(4) Not documented in CASMO4 manual

6 Conclusions and Future Work

CASMO-4 and MOCUP excel at different roles in neutronics calculations, but based on the present intercomparisons it appears that they both can do thorium related calculations with an acceptable agreement. It is preferable to perform thorium utilization studies using the simpler, more user-friendly CASMO-4 code to assess assembly and whole-core performance versus reference all-uranium lattices, and to use MOCUP for special output such as fuel pin power distribution in rim effect calculations. Use of 202 MeV/fission for thorium fuel as average fission energy produces results with comparable accuracy to a more disaggregated modelling.

Additional comparisons are planned using HELIOS, SCALE and CPM. Preliminary results show that HELIOS's results agree with MOCUP and CASMO-4 quite well, but SCALE differs by more than an acceptable amount. Tracking down the source of these differences should prove salutary to the overall challenge of modelling thorium-rich cores. It is also hoped that other groups interested in thorium fueling will elect to contribute benchmark results for the subject pin cell using different codes and cross section sets.

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