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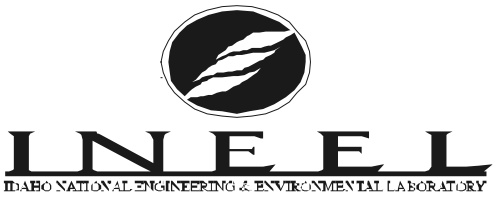

Characteristics of Mixed Thorium – Uranium Dioxide High Burnup Fuel

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June 6, 1999 – June 10, 1999

ANS 1999 Annual Meeting

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CHARACTERISTICS OF A MIXED THORIUM - URANIUM DIOXIDE HIGH-BURNUP FUEL

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November 13, 1998

Abstract

Future nuclear fuel must satisfy three sets of requirements: longer times between refueling; concerns for weapons proliferation; and development of a spent fuel form more suitable for direct geologic disposal. This project has investigated a fuel consisting of mixed thorium and uranium dioxide to satisfy these requirements.

Fuels consisting of mixed thorium and uranium dioxide have been analyzed to determine their performance during extended burnup cycles in pressurized water reactors. The uranium was enriched to 19.5 % U-235. Results using the SCALE 4.3 code system indicated that the mixed Th-U fuel could be burned to 72 MWD/kg or 100 MWD/kg using 25% and 35% UO_2 respectively. Economic analyses indicate that the Th-U fuel required less separative work and less total heavy metal (Th+U) feedstock. Even if the cost of fabricating the mixed Th-U fuel is $100/kg greater, the cost of the Th-U fuel is 13% to 25% less than that of the fuels using uranium only.

The uranium remained below 20 % total fissile fraction throughout the cycle, making it unusable for weapons. Total plutonium production per MWD was a factor of 4.5 less in the Th-U fuel than in the conventional fuel. Pu-239 production per MWD was a factor of 6.5 less in the Th-U fuel than in the conventional fuel. The plutonium produced was high in Pu-238, leading to a decay heat 5 times greater than that from plutonium derived from conventional fuel and 40 times greater than weapons grade plutonium. High decay heat would require active cooling of any crude weapon, lest the components surrounding the plutonium be melted. Spontaneous neutron production for plutonium from Th-U fuel was 2.3 times greater than that from conventional fuel and 15 times greater than that from weapons grade plutonium. High spontaneous neutron production drastically limits the probable yield of a crude weapon.

Because ThO_2 is the highest oxide of thorium, while UO_2 can be oxidized further to U_3O_8, ThO_2- UO_2 fuel may be a superior wasteform if the spent fuel is ever to be exposed to oxygenated water.

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1 Introduction

Three sets of requirements will face nuclear fuels used in Light Water Reactors (LWR) in the foreseeable future:

First, economic competition will demand longer cycles between refueling outages. The extended cycles will allow an increase in the plant capacity factor of 5 to 10%.

Second, concerns for weapons proliferation must be resolved. If nuclear energy is to make a contribution to the growing energy demands of the developing world, then the uranium (U) and plutonium (Pu) produced in that fuel must be as difficult as possible to fashion into a crude nuclear weapon.

Third, spent fuel and waste management considerations require that as much energy as possible be produced by each fuel assembly and that the fuel form be suitable for direct disposal. If reprocessing becomes the spent fuel management technique of choice at some time in the future, the fuel cycle must produce as little total plutonium and as little Pu-239 as possible.

This report has investigated a fuel consisting of mixed thorium and uranium dioxide in a quest to satisfy the above three sets of requirements.

The development of extended burnup fuel using the conventional uranium dioxide fuel requires the use of significant quantities of burnable poison for reactivity control. An alternative approach to achieving extended burnup may be the use of a mixed thorium dioxide – uranium dioxide (Th-U) fuel. In the course of this work the proportions of uranium and thorium dioxide were varied from 25 wt% UO_2 – 75 wt % ThO_2 to 35 wt% UO_2- 65 wt % ThO_2. The uranium is initially 19.5 % U-235 and 80.5 % U-238, while the thorium is 100% Th-232. Burnup ranged from about 72 megawatt days thermal per kilogram of initial heavy metal (MWD/kg ihm) to 100 MWD/kg ihm. Throughout this report, initial heavy metal is thorium and uranium. For purposes of comparison, a conventional 4.5 % enriched UO_2 fuel irradiated to 45 MWD/kg ihm and a high burnup 8.0% UO_2 fuel irradiated to 72 MWD/kg ihm are also discussed. In all cases, cladding was Zircalloy and the pin and assembly dimensions were those of a 17x17 assembly.

A large body of work on the thorium cycles was performed in support of the Light Water Breeder Reactor (LWBR) prototype at Shippingport, Pennsylvania (Belle 84). The LWBR was designed to produce U-233 from the Th-232. After discharge from the reactor, the fuel would be reprocessed to recover the bred U-233 for use in conventional LWRs or for reuse in the LWBR.

Further work on a thorium cycle reactor has been pursed by Radkowsky and co-workers. Both the Shippingport reactor experiment and the Radkowsky design relied on a highly enriched uranium (93%) seed and a surrounding thorium blanket (Galperin 95). As will be discussed on the section on plutonium production pathways, the seed and blanket arrangement results in production of U-233 in a different location from the initial uranium and thus a chemically separable quantity of U-233.

This alternative fuel concept does not rely on the reprocessing and recovery of U-233. Rather, the bred U-233 is used *in situ* to replace the initial U-235 and thus extend the life of the

fuel. This fuel would be configured into fuel pins and assemblies having the same dimensions now used in operating LWRs. The goal of the fuel would be to extend burnup with a high conversion ratio, not to be a Light Water Breeder Reactor (LWBR) as was tested at Shippingport. The fuel would be designed for a once-through fuel cycle, with no recovery of the residual U-233 or U-235 contemplated.

Preliminary scoping calculations have indicated that the reactivity of the mixed thorium-uranium dioxide fuel remains more constant during a long irradiation than does UO_2. Thorium and uranium dioxides have the same crystal structure and can be substituted in a continuous range of proportions. Calculations of the raw material needs and separative work required for the mixed Th-U fuel, described in Section 6, Economic Comparisons, indicate a 13% advantage over the present fuel cycles and a 25% advantage over an extended burnup (72 MWD/kg ihm) UO_2 fuel cycle.

A mixed Th-U fuel also has significantly greater proliferation resistance than does the conventional uranium cycle. The initial uranium in the fuel has an enrichment of 19.5%, below the threshold for proliferation concern. The reactor does not have the blanket and seed configuration used in the Shippingport and Radkowsky thorium reactor designs. Thus, the U-233 bred from Th-232 is always intimately mixed with the initial U-238. The enrichments of both the initial UO_2 (19.5% U-235) and the end-of-life UO_2 (about 7 % U-233, and about 2 % U-235) are well below the thresholds for proliferation concern. The uranium in the fuel is never weapons-usable.

Increasing the proliferation resistance of plutonium within the fuel involves increasing the fraction of Pu-238 and decreasing the fraction of Pu-239. Pu-238 is a copious source of both decay heat and of spontaneous neutrons. With a half-life of 87.7 years, Pu-238 undergoes an alpha decay to produce about 560 W/kg Pu-238. Thus, handling of the plutonium and fabrication into a weapon is extremely difficult. Pu-238 is the isotope used as a heat source on space probes such as the Cassini mission to Jupiter. Spontaneous neutrons cause predetonation of a weapon, drastically reducing the probable yield. Pu-239, on the other hand, produces 300 times less heat and 120,000 times fewer spontaneous neutrons than Pu-238.

It is important to recognize that Pu-238 and Pu-239 are produced from two fundamentally different sources. During extended fuel cycles, some of the U-235 is transmuted to U-236, then Np-237 and finally into Pu-238. Therefore, the amount of Pu-238 increases with nearly the third power of the burnup. On the other hand, Pu-239 is produced with a single neutron capture from U-238. Since the weight fraction of U-238 in the Th-U fuel has been reduced to 20-28% of the total heavy metal inventory, as opposed to the 95% U-238 fraction in conventional fuel, the production rate for Pu-239 is significantly reduced. Furthermore, Pu-239 is fissioned throughout the cycle and the inventory of Pu-239 reaches equilibrium at high burnups. Thus, the amount of Pu-239 in the mixed Th-U fuel is only about 30% that in the conventional fuel and the amount of Pu-239 per MWD is only about 15% that in conventional fuel.

Thus, the plutonium produced in Th-U fuel is high in Pu-238 (about 14 %) a strong source of spontaneous neutrons and of decay heat. The decay heat would melt the internal components of a crude nuclear weapons, while the spontaneous neutrons would cause the crude weapon to pre-detonate, reducing the yield about 20-fold. These two characteristics make the plutonium produced by the thorium – uranium fuel very undesirable for use in a clandestine weapon.

Finally thorium dioxide, the majority constituent of the fuel, has advantage in a once-through fuel cycle in that ThO_2 is the highest oxide of thorium, while UO_2 converts to U_3O_8 in oxygenated water. Thus long-term oxidation of the UO_2 in a fuel repository has the potential for release of fission products and transuranics into the groundwater, whereas there is much less long-term oxidation of $ThO_2 – UO_2$ fuels.

2 Method of Calculation

In order to estimate the feasibility of a mixed Th-U fuel we have done preliminary comparisons with conventional UO_2 fuel irradiated to 45 MWD/kg and an extended burnup UO_2 fuel irradiated to 72 MWD/kg. Calculations were performed using the SCALE 4.3 suite of codes, including BONAMI, NITAWL-II, XSDRNPM, COUPLE and ORIGEN-S. SCALE 4.3 is a modular code system for performing standardized computer analyses for NRC licensing evaluation of LWR fuel. BONAMI performs resonance self-shielding calculations for nuclides that have Bondarenko data associated with their cross-sections. NITAWL-II applies a Nordheim resonance self-shielding correction to nuclides having resolved resonance parameters. XSDRNPM is a general 1-D, discrete-ordinates code for zone-weighting of cross section, eigenvalue calculations for neutron multiplication (k-effective), and adjoint calculations for determining importance functions. COUPLE is the interface module for preparation of cross-section and spectral data for ORIGEN-S. ORIGEN-S is the version of ORIGEN used with SCALE. ORIGEN-S is a general purpose point-depletion and decay code to calculate isotopic, decay heat, radiation source terms and radioactivity levels. All calculations used the 44-group library, which was collapsed from the 238 group Evaluated Nuclear Data File/ version B-number V (ENDF/B-V) library using a spectrum for a pressurized-water-reactor fuel pin lattice. For further details on any of these codes, see NUREG/CR-0200.

3 Fuel Assembly and Fuel Cycle Parameters

The parameters of the mixed Th-U fuel cycles analyzed are shown in the following tables. Three different cycles using mixed Th-U fuel were used. The unique aspects of each of the cycles are shown in Table 1. The common characteristics of the fuel assembly and of the fuel cycles are shown in Table 2 and Table 3, respectively.

Table 1 Parameters of Fuel Cycles Evaluated

Thorium-Uranium Fuel Cycles							
Case	Uranium Enrichment	UO2 fraction	Capacity Factor	Cycle (days)	Refueling Outage (d)	Total Cycle (years)	EOL Burnup (MWD/kg ihm)
Base	4.5%	100%	76.0%	520	27.88	4.5	45
UO2	8.0%	100%	90.1%	703	28	6	72
1	19.5%	25%	90.0%	703	28	6	72
2	19.5%	35%	82.3%	1068	28	10	100

ihm = initial heavy metal (Th +U)

3.1 Fuel Assembly

The parameters of the LWR fuel assembly assumed for all of these calculations are shown in Table 2. The dimensions used are those of a 17 x 17 pressurized water reactor assembly. The same geometry, including pellet diameter and pin pitch, was used in all the cases.

It is likely that some improvement in reactivity and burnup could be achieved in the Th-U cases by using a matrix containing more fuel and less water, but such variations were beyond the scope of this investigation.

Table 2 Fuel Assembly Parameters

Fuel Assembly Parameters		
Pins	17 x 17	
Pitch	1.27	cm
Pellet Diameter	0.823	cm
Clad Outer Diameter	0.9424	cm
Fuel Pins per Assembly	264	
Active Fuel Length	363	cm
Fuel inventory	464.5	kg ihm/assembly
Cladding	Zircalloy	
Cladding thickness	0.0597	cm
Assembly dimension	21.7 x 21.7	cm
Assembly pitch	21.8	cm
non-fuel positions	25	
Non-fuel inner diameter	1.2243	cm
Non-fuel outer diameter	1.1430	cm

ihm = initial heavy metal (Th +U)

3.2 Fuel Cycle

The fuel characteristics common to all the fuel cycles are shown in Table 3. Again, the parameters are those of 17 x 17 assembly. The maximum enrichment of the uranium in the mixed Th-U fuel cases was limited to 19.5% in order to reliably remain below the 20% limit at which restrictions come into force due to weapons proliferation considerations. The specific power was constant among all the cases, except for a very long mixed Th-U fuel cycle shown in Appendix A.

Table 3 Fuel Cycle Characteristics

Fuel Cycle Characteristics		
Uranium Enrichment	4.5%	base case
(wt % of contained U)	8.0%	high burnup UO2
	19.5%	ThO2-UO2
Theoretical Density (ThO2)	10.00	g/cm3
(UO2)	10.96	g/cm3
Actual Density	94.5%	of T.D.
Specific Power	37.935	kWth/kg ihm
Fuel Temperature (average)	1611	K
	2441	F
Cladding Temperature (ave.)	750	K
	891	F
Coolant Density	0.644	g/cm3
Coolant Temperature (bulk)	605	K
	630	F
Thermal Efficiency	33.7%	

4 Reactivity Changes During Irradiation Cycle

The changes in the assembly reactivity are shown in Table 4 and Table 5. The neutron multiplication factor for a large array of assemblies, the assembly k infinity values, were computed using SCALE 4.3 assuming an infinite array of fuel assemblies. In these calculations there was no burnable poison in the fuel and no boron in the coolant. The fuel, clad and coolant are at the temperatures shown in Table 3, while the assemblies include the absorbing materials indicated in Table 2. Each of the batches is assumed to be at the same average power level, as indicated in Table 1. As stated earlier, the pellet diameter, cladding thickness, pin pitch and overall assembly dimensions were those use in present 17 x 17 LWR fuel. An optimization of these parameters for the mixed Th-U fuel would probably use a lattice with more fuel and less water. Such an optimization was outside the scope of these calculations, but would result in higher neutron multiplication at high burnups.

The core-loading was divided into three equal batches in all the cases analyzed. One third of the core, that is one batch, was replaced at each refueling outage.

It is important to note that fuels containing thorium produce U-233 via the Pa-233 intermediate, which has a 27.0-day half-life. The intermediate for Pu-239 breeding is Np-239, with a 2.355 day half-life. Thus one could expect a small increase in reactivity after a refueling outage, due to the buildup of U-233 from the equilibrium Pa-233 inventory.

Table 4 Reactivity in the Thorium -Uranium Cycle, 6-yr, 72 MWD/kg

Fuel Assembly k infinity						
		6-year, 3-batch cycle to 72 MWD/kg, 25 wt% initial uranium (days since restart)				
Batch	Specific Power (MWth/kg HM)	0.0	176	351	527	703
first	37.94	1.2428	1.1709	1.1649	1.1291	1.0933
second	37.94	1.1563	1.0859	1.0805	1.0388	1.0003
third	37.94	1.0603	0.9887	0.9769	0.9397	0.9094
core-average	37.94	1.1531	1.0819	1.0741	1.0359	1.0010

Note: SCALE recalculated the 44-group flux at the beginning and midpoint of each cycle, resulting in a small adjustment in k infinity.

Table 5 Reactivity in the Thorium -Uranium Cycle, 10-yr, 100 MWD/kg

Fuel Assembly k infinity								
		10-year, 3-batch cycle to 100 MWD/kg, 35 wt% initial uranium (days since restart)						
Batch	Specific Power (MWth/kg HM)	0.0	178	356	534	712	890	1068
first	37.94	1.3088	1.2458	1.2312	1.2000	1.1911	1.1584	1.1261
second	37.94	1.1772	1.1179	1.1120	1.0758	1.0693	1.0321	0.9972
third	37.94	1.0493	0.9872	0.9779	0.9422	0.9281	0.9074	0.8786
core-average	37.94	1.1784	1.1170	1.1070	1.0727	1.0629	1.0327	1.0006

Note: SCALE recalculated the 44-group flux at the beginning, one third and two thirds points of each cycle, resulting in a small adjustment in k infinity.

5 Change in Fuel Composition during Cycle

The isotopic compositions of the uranium and plutonium are shown in the following tables and figures. Note that the Pu-238 content is increasing at somewhat less than the cube of the burnup. This is to be expected since the source of the Pu-238, i.e. the U-235, is being fissioned and replaced with U-233. Nevertheless, Pu-238 is about 14 percent of the total plutonium at the end of life (EOL) in the 100 MWD/kg ihm (Th +U) cycle, compared to 2 percent in the conventional uranium dioxide fuel.

5.1 Base case, 4.5 yr, 45 MWD/kg ihm

In order to evaluate the mixed thorium-uranium fuels, the uranium and plutonium contents were calculated for the same fuel assembly using the same codes. The fuel was irradiated to a burnup of 45 MWD/kg ihm at a power 37.935 kWth/kg ihm for three 520-day cycles followed by a 28-day refueling outage. The uranium content of the fuel is shown in Table 6 and each is shown graphically in Figure 1. In these figures of the changing isotopic composition, each isotope's fraction is shown as the distance between the lines. Therefore, at 20 MWD/kg, the uranium is 97% U-238, 0.3% U-236 and 2.7 % U-235. This type of graphical representation will be used throughout this report.

Table 6 Uranium Content in 45 MWD/kg Base Case

Uranium Content during 4.5 yr, 45 MWD/kg cycle				
(kg/tonne initial heavy metal)				
	Burnup, MWD/kg ihm			
Isotope	0	15	30	45
U 233	0.000	0.000	0.000	0.000
U 234	0.000	0.001	0.002	0.004
U 235	44.94	29.80	18.96	11.29
U 236	0.00	2.84	4.66	5.74
U 238	955.05	944.89	934.73	924.57
U	1000.0	977.5	958.4	941.6
(weight percent of total uranium)				
	Burnup, MWD/kg ihm			
Isotope	0	15	30	45
U 233	0.00%	0.00%	0.00%	0.00%
U 234	0.00%	0.00%	0.00%	0.00%
U 235	4.49%	3.05%	1.98%	1.20%
U 236	0.00%	0.29%	0.49%	0.61%
U 238	95.51%	96.66%	97.53%	98.19%
fissile	4.49%	3.05%	1.98%	1.20%

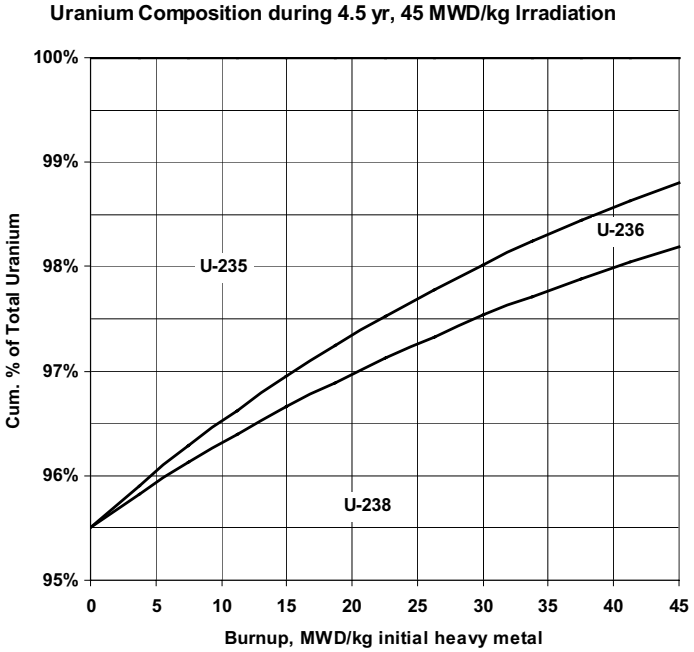

Figure 1 Uranium Composition during 45 MWD/kg Irradiation

The plutonium content of the fuel for the base case is shown in Table 7 and Figure 2. Note that the Pu-238 fraction is about 1 percent at 30 MWD/kg, increasing to 2.3 % at 45 MWD/kg. This is to be expected because of the increasing inventory of the U-236 and Np-237 targets for the three step transmutation. A further explanation of the plutonium production pathways is contained in a later section of this report.

Table 7 Plutonium content during Base Case 45 MWD/kg Irradiation

Plutonium Content during 4.5 yr, 45 MWD/kg Irradiation (kg/tonne initial heavy metal) Burnup, MWD/kg ihm				
Isotope	**0**	**15**	**30**	**45**
Pu 238	0.000	0.017	0.102	0.276
Pu 239	0.000	4.601	6.224	6.632
Pu 240	0.000	0.789	1.742	2.520
Pu 241	0.000	0.438	1.219	1.770
Pu 242	0.000	0.043	0.277	0.692
Pu	0.000	5.889	9.564	11.890
(weight percent of total uranium) Burnup, MWD/kg ihm				
Isotope	**0**	**15**	**30**	**45**
Pu 238	0.0%	0.3%	1.1%	2.3%
Pu 239	100.0%	78.1%	65.1%	55.8%
Pu 240	0.0%	13.4%	18.2%	21.2%
Pu 241	0.0%	7.4%	12.7%	14.9%
Pu 242	0.0%	0.7%	2.9%	5.8%
fissile	100.0%	85.6%	77.8%	70.7%

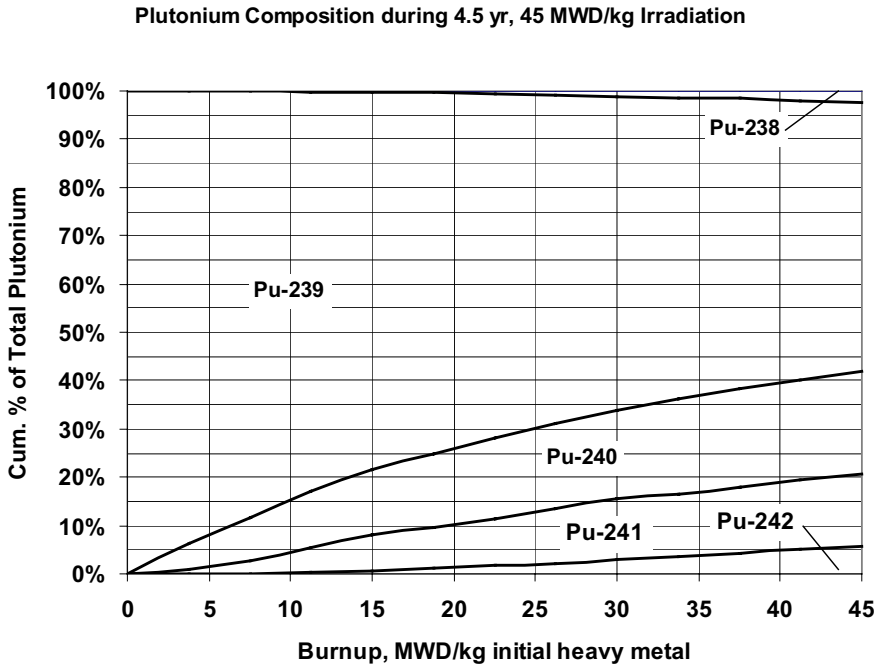

Figure 2 Change in Plutonium Composition during Base Case

5.2 Extended Burnup, UO_2 Fuel Cycle

As a second comparison, a fuel cycle using 8 % enriched uranium dioxide and irradiated to 72 MWD/kg as three batches in a 6-year cycle was investigated. Details of the cycle are shown in Table 1. While this burnup would not be permitted today because of NRC burnup limitations, such an extended fuel cycle is being evaluated as a means of making nuclear power more economically competitive.

As seen in Table 8 and Figure 3, the U-235 is consumed mostly through fission, but about 25 % of the initial U-235 is converted to U-236 by neutron capture.

Table 8 Uranium Composition during 6 yr, 72 MWD/kg cycle using only UO_2

Uranium Content during 6-yr, 72 MWD/kg Irradiation (all uranium fuel)							
(kg/tonne initial heavy metal)							
	Burnup, MWD/kg ihm						
Isotope	**0**	**12**	**24**	**36**	**48**	**60**	**72**
U 233	0.00	0.00	0.00	0.00	0.00	0.00	0.00
U 234	0.00	0.00	0.00	0.00	0.01	0.01	0.01
U 235	79.81	66.26	54.21	43.92	34.74	26.81	20.08
U 236	0.00	2.97	5.34	7.26	8.82	10.03	10.89
U 238	920.2	910.0	904.9	899.8	889.7	884.6	874.4
U	1000.0	979.2	964.5	951.0	933.2	921.4	905.4
(weight percent of total uranium) Burnup, MWD/kg ihm							
Isotope	**0**	**12**	**24**	**36**	**48**	**60**	**72**
U 233	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%
U 234	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%
U 235	8.0%	6.8%	5.6%	4.6%	3.7%	2.9%	2.2%
U 236	0.0%	0.3%	0.6%	0.8%	0.9%	1.1%	1.2%
U 238	92.0%	92.9%	93.8%	94.6%	95.3%	96.0%	96.6%
fissile	8.0%	6.8%	5.6%	4.6%	3.7%	2.9%	2.2%

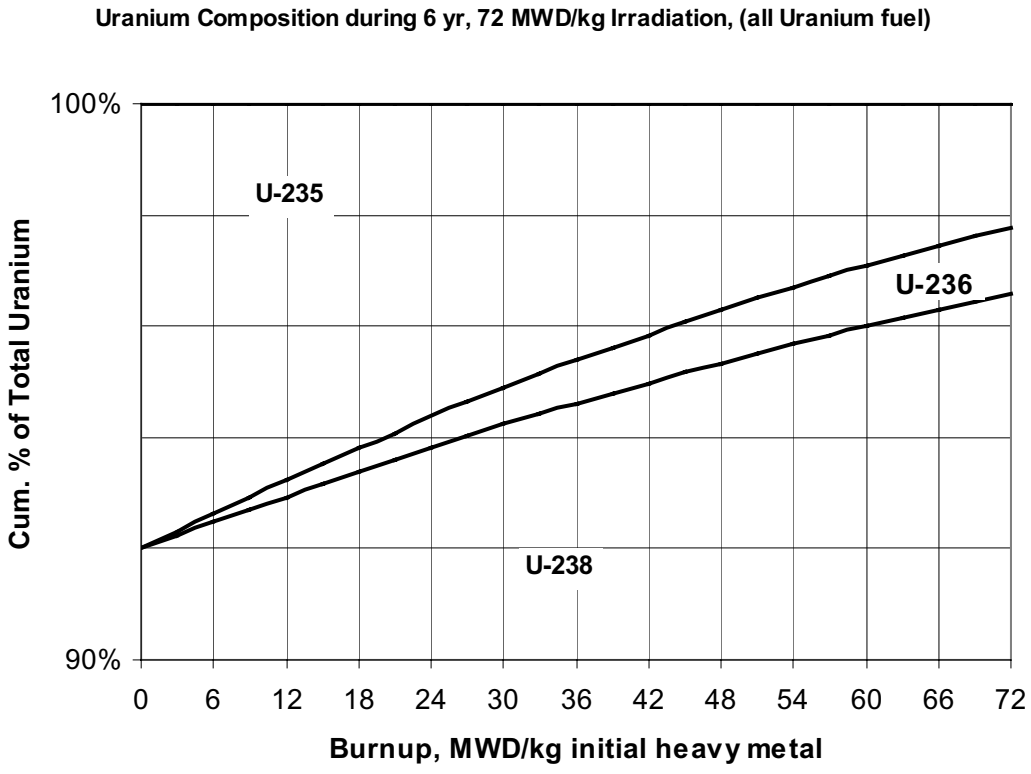

Figure 3 Change in Uranium Composition during 6 yr, 72 MWD/kg Irradiation, UO_2 fuel

The plutonium content and composition in the extended burnup UO_2 fuel are shown in Table 9 and Figure 4. Note that the Pu-238 content is 4.4%, nearly double the 2.3% in the 45 MWD/kg base case. However, the total plutonium content is 16 grams per kg of initial heavy metal in the fuel, versus 11 grams for the 45 MWD/kg base case. This comparison demonstrates that the source of the Pu-238 is primarily the U-235, whereas the Pu-239 and higher isotopes originate from the U-238.

Table 9 Plutonium Composition during 6 yr, 72 MWD/kg cycle, UO_2 fuel

Plutonium Content during 6-yr, 72 MWD/kg cycle (all uranium fuel) (kg/tonne initial heavy metal)							
	Burnup, MWD/kg IHM						
Isotope	0	12	24	36	48	60	72
Pu 238	0.0	0.009	0.049	0.133	0.271	0.467	0.712
Pu 239	0.0	4.523	6.740	7.975	8.644	8.850	8.798
Pu 240	0.0	0.402	1.018	1.633	2.211	2.728	3.162
Pu 241	0.0	0.189	0.669	1.235	1.759	2.179	2.485
Pu 242	0.0	0.008	0.063	0.189	0.383	0.641	0.943
Pu	0.0	5.131	8.539	11.165	13.269	14.865	16.101
(weight percent of total plutonium)							
	Burnup, MWD/kg IHM						
Isotope	0	12	24	36	48	60	72
Pu 238	0.0%	0.2%	0.6%	1.2%	2.0%	3.1%	4.4%
Pu 239	100.0%	88.2%	78.9%	71.4%	65.1%	59.5%	54.6%
Pu 240	0.0%	7.8%	11.9%	14.6%	16.7%	18.4%	19.6%
Pu 241	0.0%	3.7%	7.8%	11.1%	13.3%	14.7%	15.4%
Pu 242	0.0%	0.2%	0.7%	1.7%	2.9%	4.3%	5.9%
fissile	100.0%	91.8%	86.8%	82.5%	78.4%	74.2%	70.1%

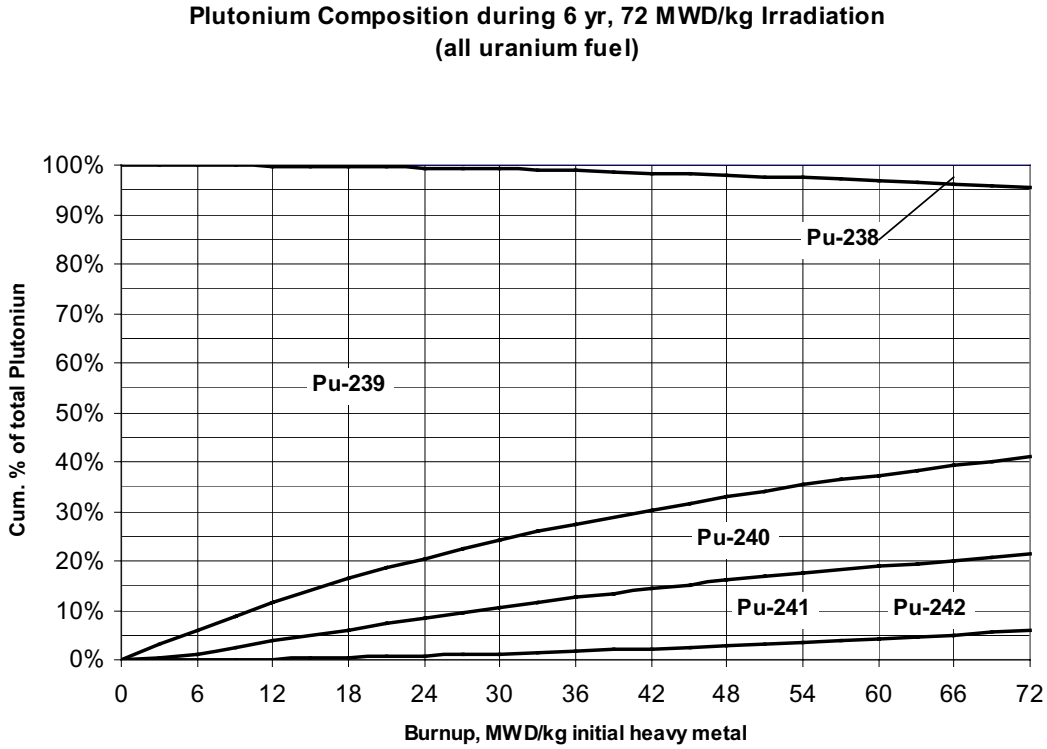

Figure 4 Plutonium Composition during Extended UO_2 Fuel Cycle

5.3 6 year, 72 MWD/kg ihm ThO_2-UO_2 Cycle

The uranium content of the Th-U fuel is shown in Table 10. Note that the U-233 increases while the U-235 is being consumed. At discharge, after 72 MWD/kg, the fissile content is still 7.7 weight percent of the total uranium and 1.6 weight percent of the initial heavy metal in the fuel. A graph showing the changing composition of the uranium is shown in Figure 5.

Table 10 Uranium Content and Composition in 6 yr, 72 MWD/kg ThO_2-UO_2 Fuel

Uranium Content during 6-yr, 72 MWD/kg Irradiation							
(kg/tonne initial heavy metal)							
	Burnup, MWD/kg ihm						
Isotope	**0**	**12**	**24**	**36**	**48**	**60**	**72**
U 233	0.00	4.45	7.78	9.83	11.09	11.64	11.79
U 234	0.00	0.22	0.61	1.10	1.64	2.20	2.77
U 235	48.92	35.47	25.25	17.35	11.43	7.18	4.37
U 236	0.00	2.56	4.35	5.59	6.40	6.91	7.11
U 238	201.88	198.80	196.24	193.68	190.60	188.04	184.97
U	250.80	241.50	234.23	227.55	221.17	215.98	211.01
(weight percent of total uranium)							
	Burnup, MWD/kg ihm						
Isotope	**0**	**12**	**24**	**36**	**48**	**60**	**72**
U 233	0.0%	1.8%	3.3%	4.3%	5.0%	5.4%	5.6%
U 234	0.0%	0.1%	0.3%	0.5%	0.7%	1.0%	1.3%
U 235	19.5%	14.7%	10.8%	7.6%	5.2%	3.3%	2.1%
U 236	0.0%	1.1%	1.9%	2.5%	2.9%	3.2%	3.4%
U 238	80.5%	82.3%	83.8%	85.1%	86.2%	87.1%	87.7%
fissile	19.5%	16.5%	14.1%	11.9%	10.2%	8.7%	7.7%

Note also that the U-236 content is 3.4 %, compared with 0.61 % in the case for conventional fuel at45 MWD/kg. U-236, which has a half-life of 23 million years, is the limiting step in the production of Pu-238. Thus, if this uranium were to be separated from the Th-U fuel, any further plutonium produced would be high in Pu-238, severely limiting its weapons-usability.

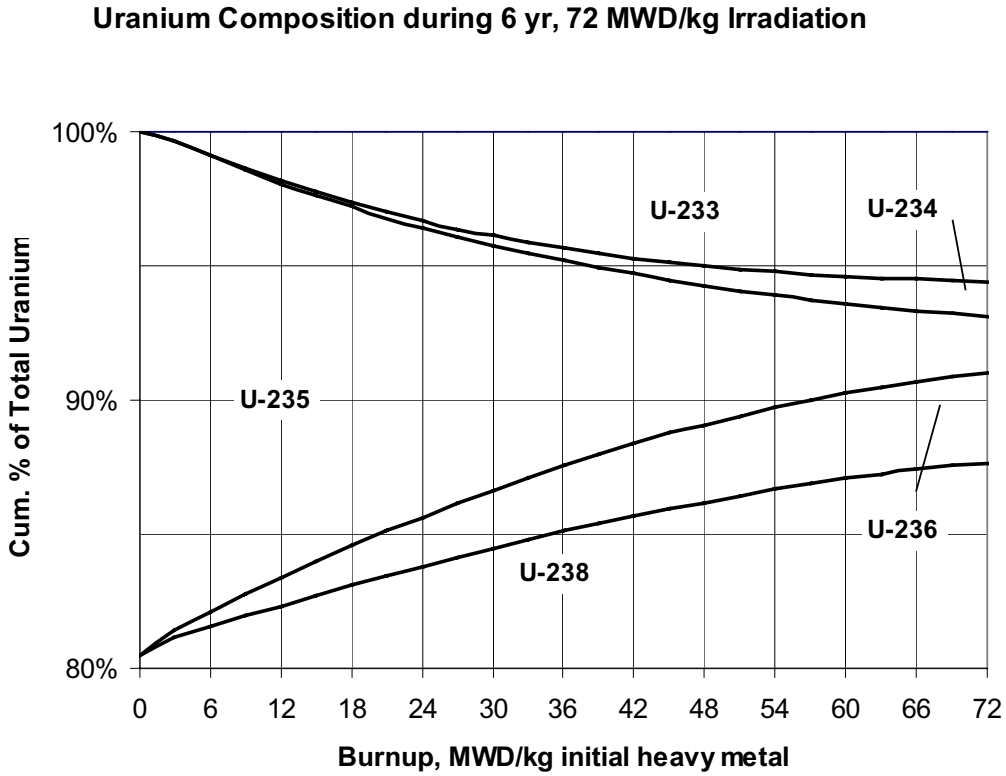

Figure 5 Uranium Composition during 6-yr, 72 MWD/kg Irradiation

The plutonium content of the mixed thorium-uranium fuel during the fuel cycle is shown in Table 11. The changing composition of the plutonium during the entire cycle is shown in Figure 6.

Table 11 Plutonium Content and Composition in 6 yr, 72 MWD/kg ThO_2-UO_2 Fuel

Plutonium Content during 6-yr, 72 MWD/kg cycle (kg/tonne initial heavy metal) Burnup, MWD/kg IHM							
Isotope	0	12	24	36	48	60	72
Pu 238	0	0.0077	0.04601	0.1209	0.2265	0.3464	0.4611
Pu 239	0	1.6002	2.07356	2.1044	1.9758	1.806	1.6568
Pu 240	0	0.232	0.49447	0.6717	0.775	0.8215	0.8422
Pu 241	0	0.123	0.38861	0.5863	0.6693	0.6693	0.633
Pu 242	0	0.0094	0.07033	0.1891	0.3439	0.5074	0.6617
Pu	0	1.9723	3.07298	3.6725	3.9905	4.1506	4.2548
(weight percent of total plutonium) Burnup, MWD/kg IHM							
Isotope	0	12	24	36	48	60	72
Pu 238	0.0%	0.4%	1.5%	3.3%	5.7%	8.3%	10.8%
Pu 239	100.0%	81.1%	67.5%	57.3%	49.5%	43.5%	38.9%
Pu 240	0.0%	11.8%	16.1%	18.3%	19.4%	19.8%	19.8%
Pu 241	0.0%	6.2%	12.6%	16.0%	16.8%	16.1%	14.9%
Pu 242	0.0%	0.5%	2.3%	5.1%	8.6%	12.2%	15.6%
fissile	100.0%	87.4%	80.1%	73.3%	66.3%	59.6%	53.8%

Plutonium Composition during 6 yr, 72 MWD/kg Irradiation

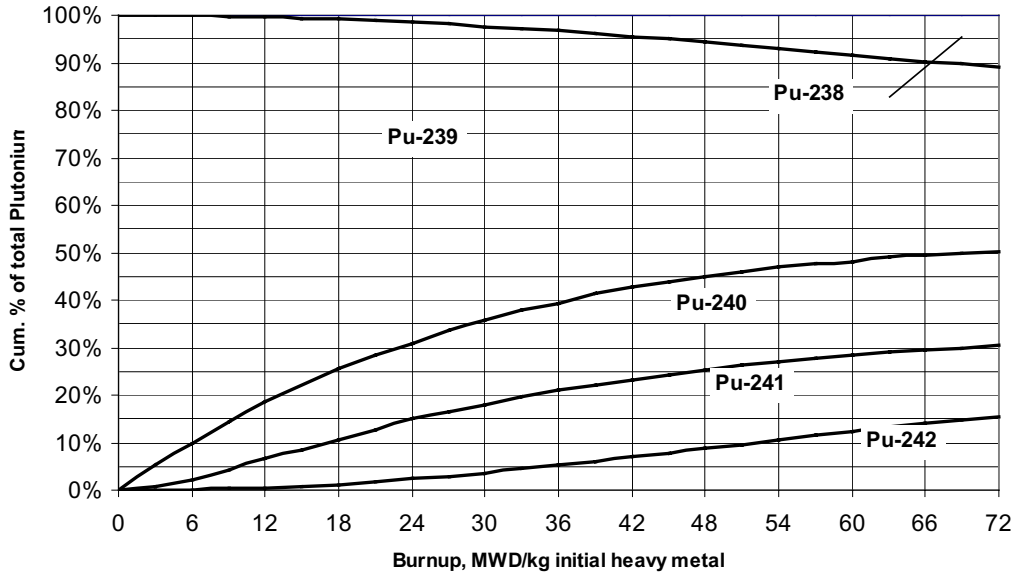

Figure 6 Changes in Plutonium Composition during 6 yr, 72 MWD/kg Cycle

5.4 10 year, 100 MWD/kg ihm ThO_2-UO_2 Cycle

A further investigation of the potential use of the Th-U fuel considered an irradiation to 100 MWD/kg of initial heavy metal. The fuel cycle was a three-batch operation, burning the fuel for 1068 days between refueling outages, as shown in Table 1. The refueling outages were 28 days. In order to have sufficient reactivity for this very extended burnup, the uranium dioxide fraction in the initial fuel was increased to 35 % from the 25% used in the 6 yr, 72 MWD/kg ThO_2-UO_2 fuel. The enrichment of the uranium was again 19.5 %, to remain reliably below thresholds for fuel use limitations due to proliferation considerations.

Note that the uranium at discharge is still 5.5 % fissile, but that more than two thirds of the fissile content is U-233. The U-236 content is now 3.6 %, leading to high Pu-238 production in any subsequent use of the uranium, for the reasons cited earlier.

Table 12 Uranium Content and Composition during 10 yr, 100 MWD/kg Irradiation

Uranium Content during 10-yr, 100 MWD/kg cycle					
(kg/tonne initial heavy metal) Burnup, MWD/kg IHM					
Isotope	**0**	**25**	**50**	**75**	**100**
U 233	0.00	6.57	9.93	11.09	11.04
U 234	0.00	0.44	1.21	2.13	3.04
U 235	68.81	41.89	23.42	11.28	4.57
U 236	0.00	5.18	8.23	9.81	10.16
U 238	283.9	277.2	270.5	263.9	256.7
U	352.7	331.3	313.3	298.2	285.5
(weight percent of total uranium) Burnup, MWD/kg ihm					
Isotope	**0**	**25**	**50**	**75**	**100**
U 233	0.00%	1.98%	3.17%	3.72%	3.87%
U 234	0.00%	0.13%	0.39%	0.71%	1.07%
U 235	19.51%	12.65%	7.48%	3.78%	1.60%
U 236	0.00%	1.56%	2.63%	3.29%	3.56%
U 238	80.49%	83.67%	86.34%	88.50%	89.91%
fissile	19.51%	14.63%	10.65%	7.50%	5.47%

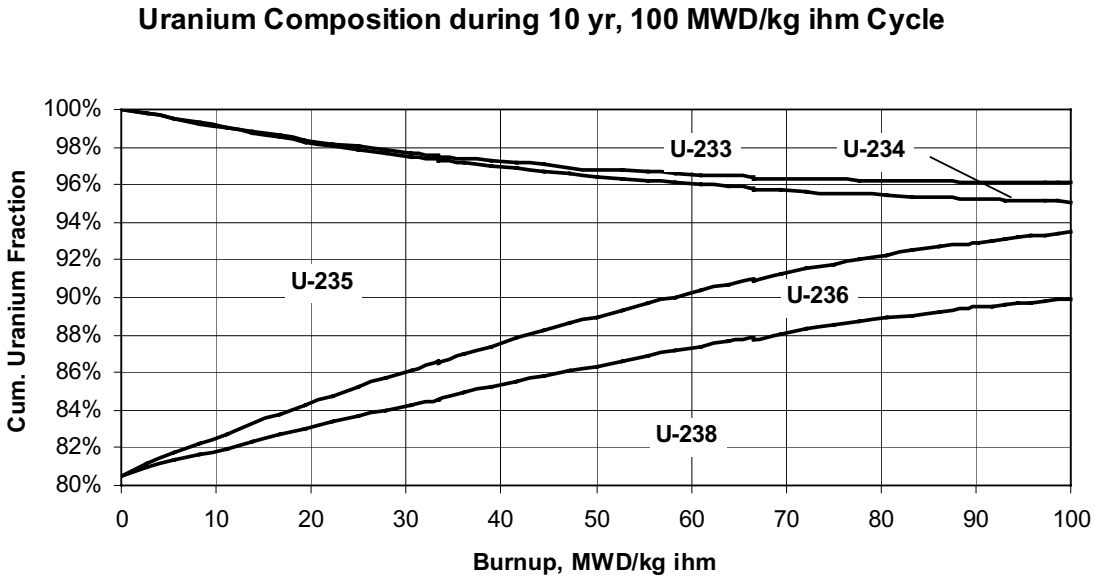

Figure 7 Change in Uranium Composition during 10 yr, 100 MWD/kg Irradiation

The plutonium content of the Th-U fuel after an irradiation to 100 MWD/kg is shown in Table 13 and Figure 8. These further amplify an important feature of the Th-U fuel. Note that the Pu-238 content has become nearly 14 %. The high decay heat and spontaneous neutron production of the Pu-238 have strong ramifications for the handling of the plutonium, should it ever be separated from the fuel. These ramifications are discussed later in this report.

Table 13 Plutonium Content and Composition during 10 yr, 100 MWD/kg Irradiation

Plutonium Content during 10-yr, 100 MWD/kg cycle					
(kg/tonne initial heavy metal) Burnup, MWD/kg ihm					
Isotope	**0**	**25**	**50**	**75**	**100**
Pu 238	0.000	0.044	0.242	0.569	0.871
Pu 239	0.000	2.851	3.092	2.691	2.274
Pu 240	0.000	0.537	0.982	1.168	1.214
Pu 241	0.000	0.391	0.877	0.981	0.872
Pu 242	0.000	0.051	0.295	0.662	1.016
Pu	0.000	3.874	5.489	6.070	6.247
(weight percent of total plutonium) Burnup, MWD/kg ihm					
Isotope	**0**	**25**	**50**	**75**	**100**
Pu 238	0.00%	1.14%	4.42%	9.37%	13.94%
Pu 239	100.00%	73.59%	56.34%	44.33%	36.40%
Pu 240	0.00%	13.87%	17.89%	19.24%	19.44%
Pu 241	0.00%	10.09%	15.98%	16.16%	13.95%
Pu 242	0.00%	1.31%	5.38%	10.90%	16.26%
fissile	100.00%	83.67%	72.32%	60.49%	50.36%

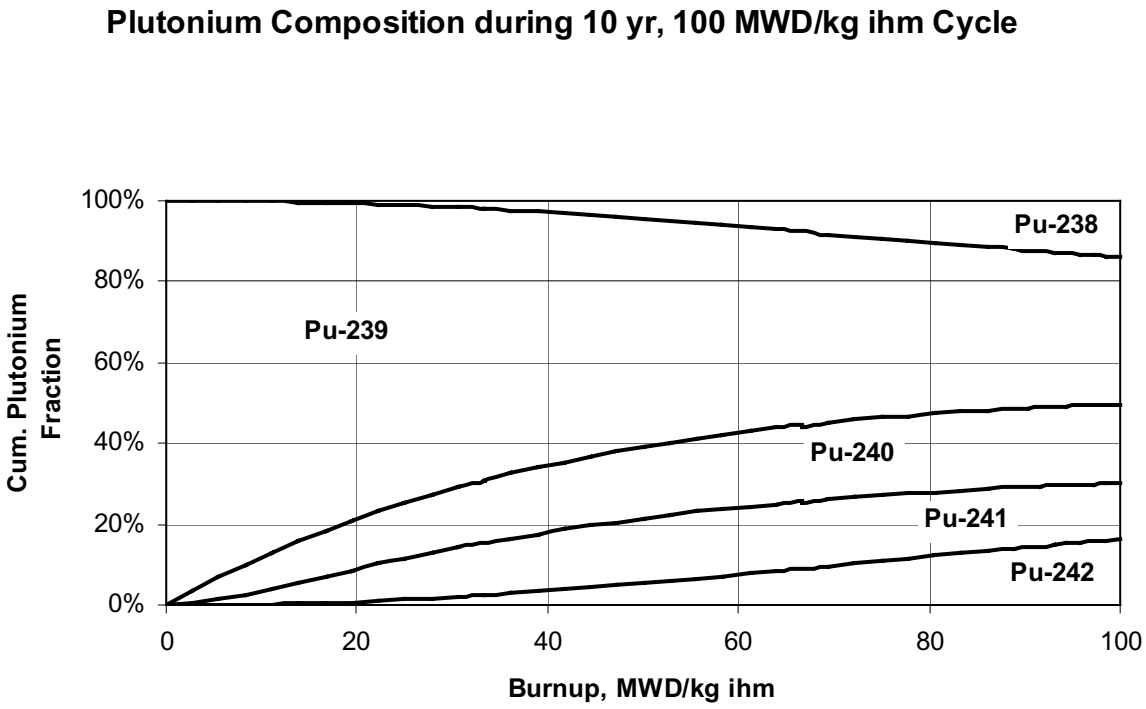

Figure 8 Change in Plutonium Composition during 10 yr, 100 MWD/kg Irradiation

6 Economic Comparisons

The results of the economic comparisons among the two uranium dioxide fuels and the two mixed Th-U fuels are shown in Table 14. These calculations show a 10-20% advantage for the mixed fuel compared with the two UO_2 fuels.

Note that we have assumed that the fabrication cost of the Mixed Th-U fuel is $300/kg, 50% higher than that of the current LWR fuel. Also, the tails assay is 0.3% U-235, reflecting the low current prices of yellow cake. The use of 0.2 % tails assay would result in a greater advantage for the mixed Th-U fuel because of the somewhat lower enrichment per MWD required.

We have included the cost of burnable poison in the UO_2 fuels. We have also not included any waste disposal adjustment for the smaller volume of spent fuel resulting from an extended irradiation. The extended burnup of the 100 MWD/kg mixed Th-U fuel case would result in about half the spent fuel per unit of electricity generated, when compared with the 45 MWD/kg base case.

Table 14 Cost Comparison Using Current Prices

Fuel Cost Comparison					
	Current PWR Uranium Dioxide Fuel	Ultra High Burnup Uranium Dioxide Fuel	Mixed Uranium/ Thorium Dioxide Fuel 72 MWD/kg	Mixed Uranium/ Thorium Dioxide Fuel 100 MWD/kg	units
Specific Power	37.935	37.935	37.935	37.935	kWth/kg
Total cycle length	4.5	6	6	10	years
Effective Full Power Days	1186	1897	1897	2635	efpd
Burnup	45.0	72.0	72.0	100.0	MWD/kg HM
feed U-235 content	0.72%	0.72%	0.72%	0.72%	atom %
product U-235 enrichment	4.95%	8.00%	19.70%	19.70%	atom %
tails U-235 content	0.30%	0.30%	0.30%	0.30%	atom %
feed U-235 content	0.711%	0.711%	0.711%	0.711%	wt %
product U-235 enrichment	4.891%	7.907%	19.500%	19.500%	wt %
tails U-235 content	0.296%	0.296%	0.296%	0.296%	wt %
fraction uranium in fuel	1.000	1.000	0.250	0.350	
separative work	7.034	13.081	9.367	13.114	kg-SWU/kg fuel
burnable poison	0.00	0.10	0.00	0.00	kg/kg fuel
natural uranium	11.077	18.350	11.575	16.206	kg/kg fuel
natural thorium	0.000	0.000	0.750	0.650	kg/kg fuel
Total heavy metal mined	11.077	18.350	12.325	16.856	kg/kg fuel
	0.246	0.255	0.171	0.169	kg/MWD
Rates					
interest rate	8.0%	8.0%	8.0%	8.0%	per year
natural uranium	$ 25.00	$ 25.00	$ 25.00	$ 25.00	/kg
	$ 13.40	$ 13.40	$ 13.40	$ 13.40	/lb U3O8
natural thorium	$ 88.50	$ 88.50	$ 88.50	$ 88.50	/kg
Gadolinium	$ 115.00	$ 115.00	$ 115.00	$ 115.00	/kg
conversion U3O8 to UF6	$ 5.00	$ 5.00	$ 5.00	$ 5.00	/kg enr U
separative work	$ 75.00	$ 75.00	$ 75.00	$ 75.00	/kg-SWU
Costs					
raw materials	$ 276.93	$ 470.26	$ 355.76	$ 462.66	/kg fuel
separative work	$ 527.54	$ 981.08	$ 702.51	$ 983.51	/kg fuel
conversion	$ 5.00	$ 5.00	$ 1.25	$ 1.75	/kg fuel
fabrication	$ 200.00	$ 250.00	$ 300.00	$ 300.00	/kg fuel
Total cost	$ 1,009.47	$ 1,694.84	$ 1,359.52	$ 1,747.93	/kg fuel
interest during use	$ 181.70	$ 406.76	$ 326.28	$ 699.17	/kg fuel
Total fuel cost	$ 26.48	$ 29.20	$ 23.43	$ 24.48	/MWth-day
	$ 0.323	$ 0.357	$ 0.286	$ 0.299	/million BTU
	$ 3.27	$ 3.61	$ 2.90	$ 3.03	/MWe-hr
Difference from minimum	13.0%	24.7%	0.0%	4.5%	

Note that the interest paid on the initial investment in the fuel is a significant penalty for the extended burn fuels as compared with the conventional fuel. Avoiding the need for refueling outages in the longer cycles may counteract that penalty, but costs of refueling outages are not included in this analysis.

A comparison at higher uranium prices and comparable thorium prices is shown in Table 15. The yellowcake prices are about double today's low levels, while the price of thorium has been reduced to $60 per kg. Because of the higher uranium costs, the tails assays from the enrichment plant has been lowered to 0.2 wt percent U-235.

Thorium is about three times as abundant in nature as uranium, but has a higher price today, primarily because of the smaller amounts mined. On the other hand, about 60,000 t of uranium is mined worldwide annually. Thus one would expect that the economies of scale have already been achieved. The continued use of low enriched uranium in LWRs, where five to ten kilograms of uranium must be mined per kilogram of fuel, can be expected to put upward pressure on the price of uranium in the next century.

Thorium is produced from monazite ore, a rare-earth-thorium-phosphate mineral and as a byproduct of the processing of heavy mineral sands for titanium, zirconium or tin. (USGS-Th-96)

Note that a higher price for uranium and a slight decrease in the thorium price result in a 20 to 30 % cost advantage for the mixed Th-U fuel. While the uranium used in the mixed Th-U fuel is more highly enriched, the total UO_2 content is only 25 % or 35% of the uranium only fuels. Therefore, less total uranium is required, 9.4 kg/kg of fuel for the mixed Th-U fuel versus 15 kg/kg of fuel for U fuel, where the burnup for both is 72 MWD/kg. The cost of the mixed Th-U fuel should be relatively insensitive to the price of thorium, since no enrichment is required (or possible since Th-232 is the only isotope.) Therefore, no enrichment tails are generated and only 0.75 or 0.65 kg of natural thorium is needed per kilogram of fuel.

Table 15 Cost Comparison Using Higher Uranium Prices

Fuel Cost Comparison Using Future Prices					
	Current PWR Uranium Dioxide Fuel	**Ultra High Burnup Uranium Dioxide Fuel**	**Mixed Uranium/ Thorium Dioxide Fuel 72 MWD/kg**	**Mixed Uranium/ Thorium Dioxide Fuel 100 MWD/kg**	units
Specific Power	37.935	37.935	37.935	37.935	kWth/kg
Total cycle length	4.5	6	6	10	years
Effective Full Power Days	1186	1897	1897	2635	efpd
Burnup	45.0	72.0	72.0	100.0	MWD/kg HM
feed U-235 content	0.72%	0.72%	0.72%	0.72%	atom %
product U-235 enrichment	4.95%	8.00%	19.70%	19.70%	atom %
tails U-235 content	0.20%	0.20%	0.20%	0.20%	atom %
feed U-235 content	0.711%	0.711%	0.711%	0.711%	wt %
product U-235 enrichment	4.891%	7.907%	19.500%	19.500%	wt %
tails U-235 content	0.197%	0.197%	0.197%	0.197%	wt %
fraction uranium in fuel	1.000	1.000	0.250	0.350	
separative work	8.649	15.862	11.182	15.654	kg-SWU/kg fuel
burnable poison	0.00	0.10	0.00	0.00	kg/kg fuel
natural uranium	9.139	15.014	9.398	13.157	kg/kg fuel
natural thorium	0.000	0.000	0.750	0.650	kg/kg fuel
Total heavy metal mined	9.139	15.014	10.148	13.807	kg/kg fuel
	0.203	0.209	0.141	0.138	kg/MWD
Rates					
interest rate	8.0%	8.0%	8.0%	8.0%	per year
natural uranium	$ 50.00	$ 50.00	$ 50.00	$ 50.00	/kg
	$ 26.80	$ 26.80	$ 26.80	$ 26.80	/lb U3O8
natural thorium	$ 60.00	$ 60.00	$ 60.00	$ 60.00	/kg
Gadolinium	$ 115.00	$ 115.00	$ 115.00	$ 115.00	/kg
conversion U3O8 to UF6	$ 5.00	$ 5.00	$ 5.00	$ 5.00	/kg enr U
separative work	$ 75.00	$ 75.00	$ 75.00	$ 75.00	/kg-SWU
Costs					
raw materials	$ 456.97	$ 762.19	$ 514.88	$ 696.83	/kg fuel
separative work	$ 648.65	$ 1,189.61	$ 838.63	$ 1,174.08	/kg fuel
conversion	$ 5.00	$ 5.00	$ 1.25	$ 1.75	/kg fuel
fabrication	$ 200.00	$ 250.00	$ 300.00	$ 300.00	/kg fuel
Total cost	$ 1,310.63	$ 2,195.30	$ 1,654.75	$ 2,172.66	/kg fuel
interest during use	$ 235.91	$ 526.87	$ 397.14	$ 869.06	/kg fuel
Total fuel cost	$ 34.37	$ 37.83	$ 28.51	$ 30.43	/MWth-day
	$ 0.420	$ 0.462	$ 0.348	$ 0.372	/million BTU
	$ 4.25	$ 4.68	$ 3.53	$ 3.76	/MWe-hr
Difference from minimum	20.6%	32.7%	0.0%	6.7%	

7 Neutron Sprectra in High-Burnup UO_2 and Mixed ThO_2-UO_2 Fuels

In a comparison of the uranium and thorium-uranium fuels, a single pin model of a full length PWR fuel rod was modeled using MCNP4B. The upper and lower plena and support structures were also modeled. The isotopic content of the pins at various burnups were used to determine the neutron energy sprectra. Figure 9 and Figure 10 show the energy spectra calculated using 0.1 lethargy unit bins. While the source region in energy space for the two fuels is comparable, special attention should be paid to the region near 0.3 eV (0.3E-6 MeV). Pu-239 has a strong resonance for both fission and capture in that region, as shown in Figure 11 and Figure 12. (McLane 88)

The peak of the 0.3 eV resonance for fission is 3500 b while that for absorption is about 2200 b. U-233, on the other hand, has only a weak resonance for absorption (30 b at 0.15 eV). The U-233 fission cross-section has no resonances below 1.0 eV. The fission and capture cross sections for U-233 are shown in Figure 13 and Figure 14. (McLane 88) The fission to capture ratio for U-233 below 1 eV is about 10 while the fission to capture ratio for Pu-239 is about 2. Therefore, U-233 is far more likely to fission and not produce a higher actinide than is Pu-239.

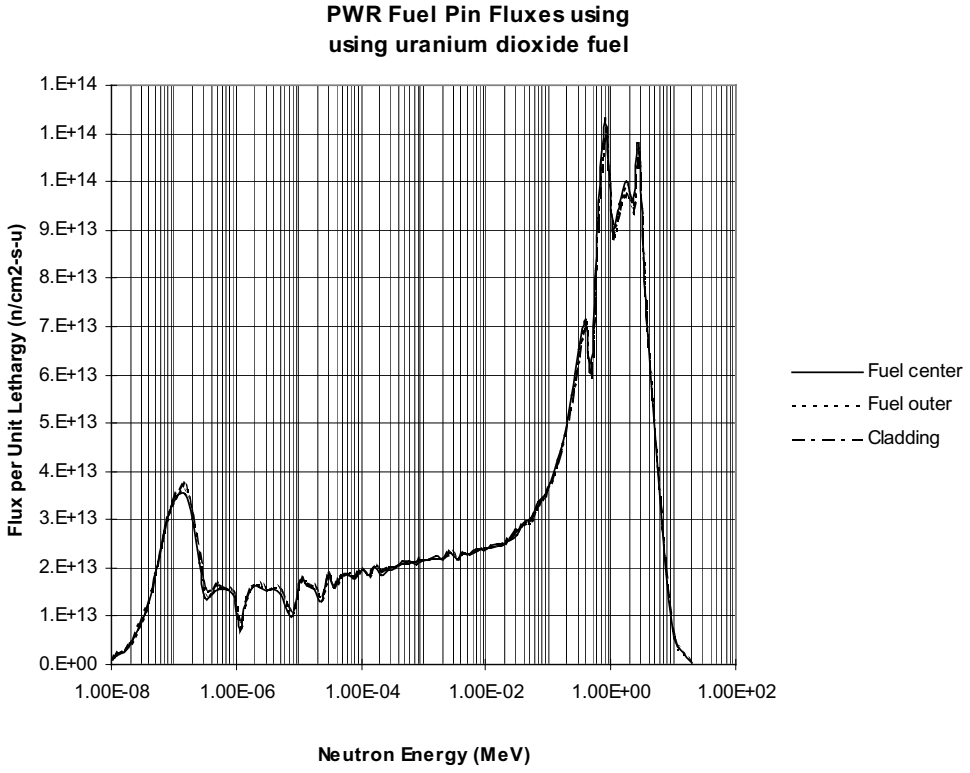

Figure 9 Neutron Spectrum in PWR Fuel Pin Fluxes with UO2

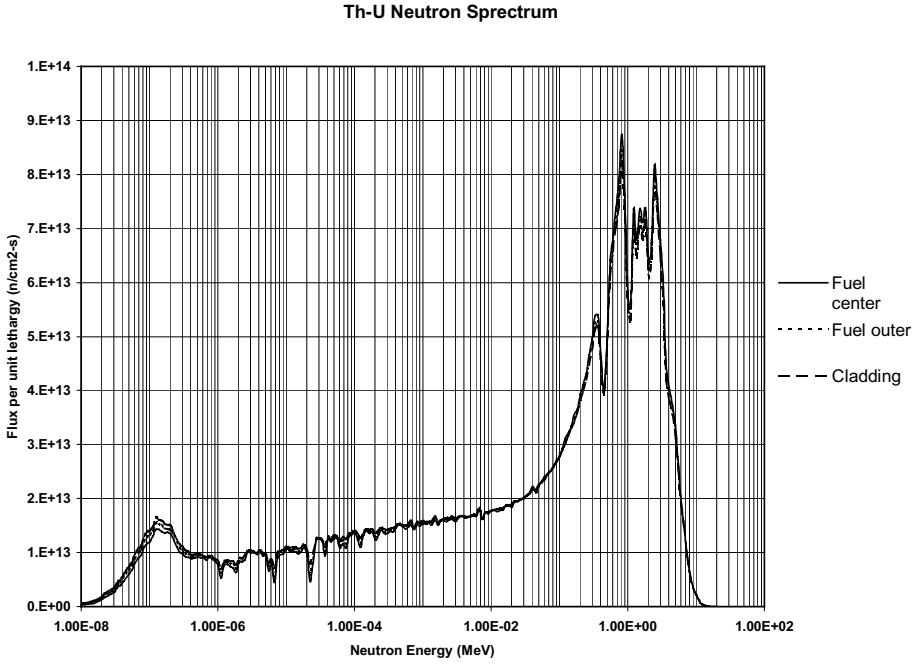

Figure 10 Neutron Spectrum in Mixed Th-U Fuel

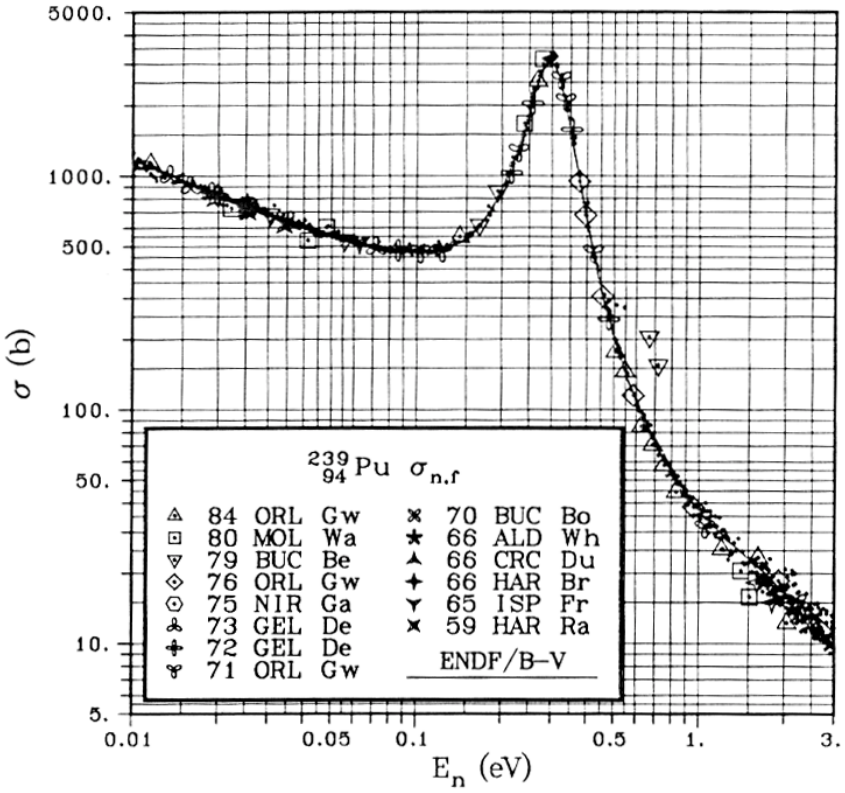

Figure 11 Pu-239 fission cross section in thermal range

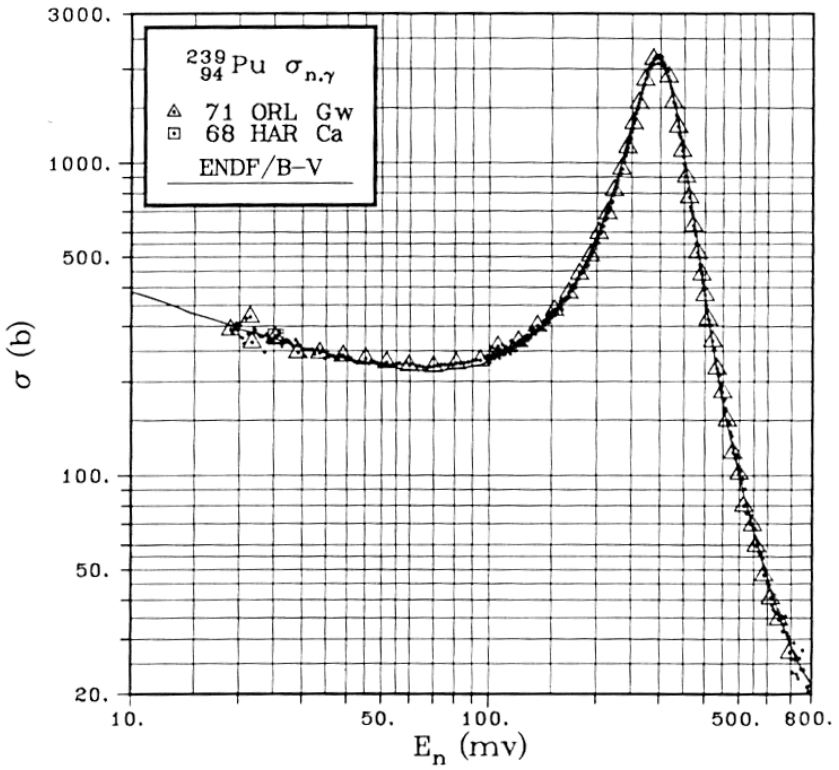

Figure 12 Pu-239 (n,γ) cross section in thermal range

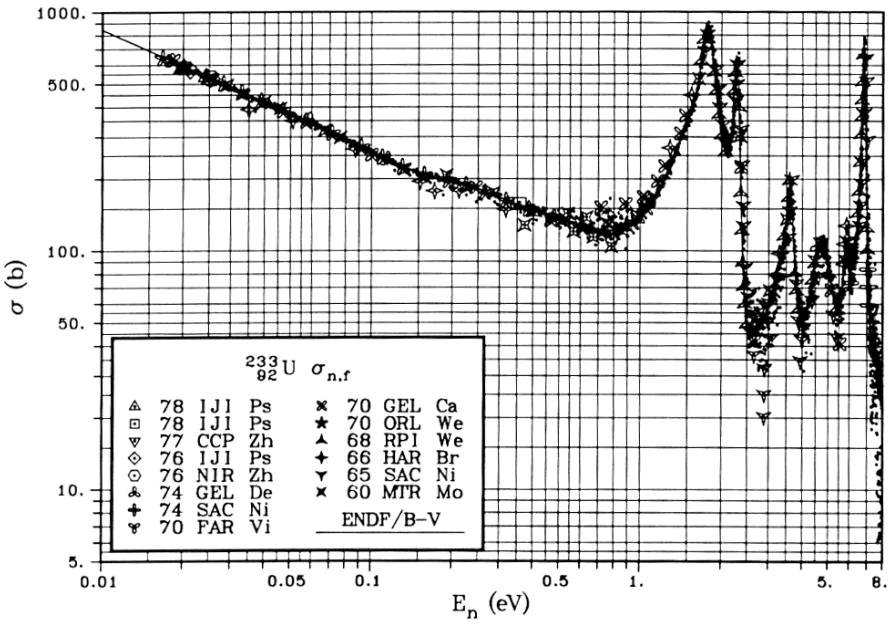

Figure 13 U-233 fission cross section in thermal range

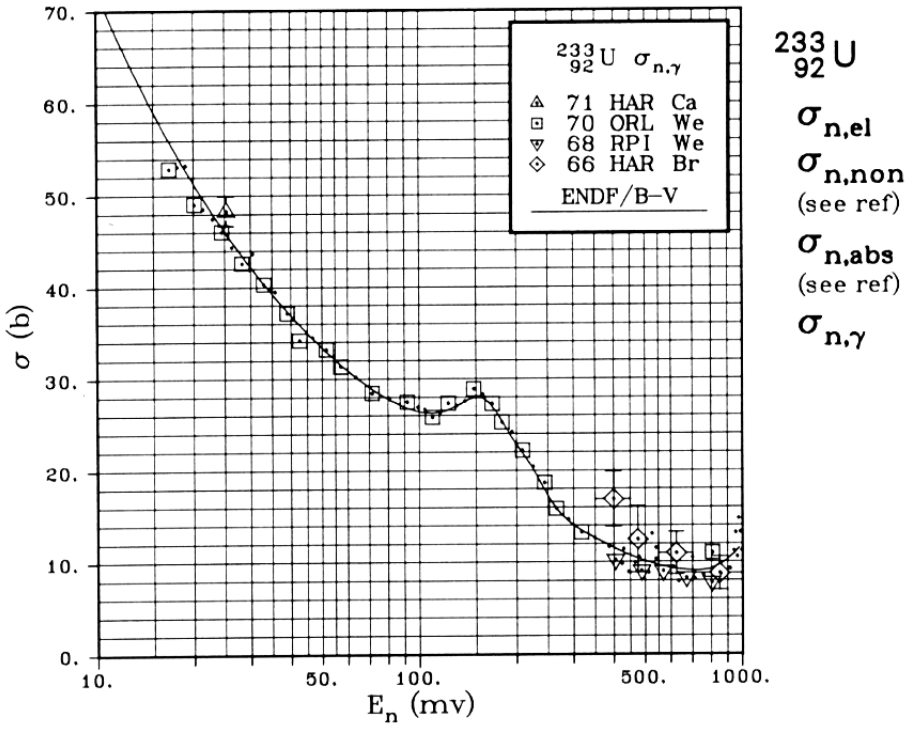

Figure 14 U-233 (n,γ) cross section in thermal range

8 Proliferation Considerations

This section of the report compares the characteristics of plutonium produced in a high burnup thorium-based fuel with that produced in conventional fuel irradiated to 45 megawatt days thermal per kilogram of initial heavy metal (MWD/kg ihm) in a pressurized water reactor.

The clandestine chemical separation of plutonium from spent commercial reactor fuel is a concern with the wider use of uranium fission for nuclear power. While the safeguards and inspections specified in the Nuclear Non-Proliferation Treaty of 1967 (NPT) can reduce the risk that a clandestine weapon will be fashioned by a national or sub-national group, the inherent characteristics of the spent fuel can add another layer of protection.

In the cases analyzed here, we assume the elementally pure plutonium can be separated from the fuel. Surreptitious chemical separation of plutonium from high burnup spent fuel is very difficult, but for the purposes of this analysis we will assume that such separations are successful. Comparisons of the isotopic mixtures of plutonium are made based on the heat and spontaneous neutrons produced by a given mass of the separated plutonium.

8.1 Characteristics of Plutonium from Various Sources

Several standard grades of plutonium have entered the nomenclature, as summarized in Table 16. Obviously, pure Pu-239 would be the most desirable for weapons use. Super grade and weapons-grade plutonium are produced by irradiating natural or depleted uranium targets to

relatively low fluences. Reactor-grade and mixed U-Pu oxide (MOX) grade plutonium is produced at higher fluences in low enrichment fuel. Of course, these grades are not fixed and the particular mixture of isotopes produced in any fuel cycle will depend on the neutron spectrum and flux, the length of the cycle, and the cooling allowed both during refueling outages and after discharge from the reactor.

Table 16 Isotopic Composition of Various Grades of Plutonium

	Isotopes				
Grade	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
Super-grade		98.0%	2.0%		
Weapons-grade	0.012%	93.8%	5.8%	0.35%	0.022%
Reactor-grade	1.3%	60.3%	24.3%	9.1%	5.0%
MOX-grade	1.9%	40.4%	32.1%	17.8%	7.8%

The spontaneous neutron generation rate and the amount of decay heat produced by the various plutonium isotopes determine the usability of a particular mixture of isotopes for use in a clandestine weapon. The properties of the dominant plutonium and americium isotopes are shown in Table 17. Am-241 is the produced of the 14.4 year beta decay of Pu-241 and would grow into separated plutonium over time.

The reasons for the desirability of Pu-239 and the corresponding undesirablity of Pu-238, Pu-240, Pu-242 and Am-241 are apparent. The spontaneous neutron generation rate and the decay heat rate for Pu-239 is orders of magnitude lower than for the adjacent isotopes. In order to make any separated plutonium as undesirable as possible for clandestine use in a weapon, the fractional of Pu-238 should be raised as high as possible. Pu-238 decays with a 5.5 MeV alpha particle and a 87.7 year halflife to stable and non-fissile U-234. Pu-238 is used as a heat source for spacecraft such as the Cassini probe to Jupiter.

Table 17 Properties of Dominant Plutonium and Americium Isotopes

	Halflife	**Bare Crit**	**Spontaneous Fission Neutrons**	**Decay Heat**
Isotope	years	kg, α–phase	neutrons/gm-s	Watts/kg
Pu-238	87.7	10	2600	560
Pu-239	24,100	10	0.022	1.9
Pu-240	6,560	40	910	6.8
Pu-241	14.4	10	0.049	4.2
Pu-242	376,000	100	1700	0.1
Am-241	430	100	1.2	114

8.2 Production of Plutonium in Reactor Fuel

It is important to note that the plutonium present in nuclear fuel is produced in by two chains of reactions. About 75% of the Pu-238 present in ultra high burnup fuel is produced in the following three-absorption reaction chain. The cross-sections shown are the thermal cross-sections followed by the resonance integrals.

$$^{235}U(n,\gamma)\xrightarrow[99b,140b]{}{}^{236}U(n,\gamma)\xrightarrow[5.1\,b,360b]{}{}^{237}U\xrightarrow[\beta^-(6.75\,days)]{}{}^{237}Np$$

$$^{237}Np(n,\gamma)\xrightarrow[170b,730b]{}{}^{238}Np\xrightarrow[\beta^-(2.117\,days)]{}{}^{238}Pu$$

Since this chain of reactions requires three neutron absorption reactions, the inventory of Pu-238 increases approximately as the cube of the burnup. The inventory of Pu-238 deviates from the cube at high burnups because of the consumption of U-235. As mentioned earlier the limiting step is the $^{236}U(n,\gamma)^{237}U$ reaction, for which the thermal cross section is 5.1 b.

On the other hand, the Pu-239 is produced in the following familiar reaction.

$$^{238}U(n,\gamma)\xrightarrow[2.68b,277\,b]{}{}^{239}U\xrightarrow[\beta^-(23.5\,\mathrm{min})]{}{}^{239}Np\xrightarrow[\beta^-(2.35\,days)]{}{}^{239}Pu$$

Thus a fuel cycle will produce plutonium with a high fraction of Pu-238 if the initial U-235 is taken to high burnup, to increase Pu-238 production, and if the inventory of U-238 is kept low to prevent the generation of other plutonium isotopes.

The amounts of plutonium produced in the various fuel cycles are compared in Table 18. Note that the total amount of plutonium produced in any of the mixed Th-U fuels is about a factor of 4 to 4.5 less than that produced in the conventional uranium dioxide 45 MWD/kg fuel. This follows from the fact that the U-238 content of the mixed Th-U fuels is only about 20 to 28% of that present in conventional fuels. Furthermore, the amount of Pu-239 is a factor of 6.5 less than that of the conventional fuel, first due to the smaller amount of U-238 present and secondly due to the higher burnup during which the Pu-239 is fissioned.

Table 18 Plutonium Production in U and Th-U Cycles

Plutonium Production in U and Mixed Th-U Cycles				
	U 4.5 yr, 45 MWD/kg	U 8 % enr, 6 yr, 72 MWD/kg	Th-U 6 yr, 72 MWD/kg	Th-U 10 yr, 100 MWD/kg
Burnup (MWD/kg)	**45**	**72**	**72**	**100**
Production		**gram/kg ihm**		
Pu-238	0.276	0.712	0.461	0.871
Pu-239	6.632	8.798	1.657	2.274
Pu-240	2.520	3.162	0.842	1.214
Pu-241	1.770	2.485	0.633	0.872
Pu-242	0.692	0.943	0.662	1.016
Total Pu	11.890	16.101	4.255	6.247
Production per MWD				
grams Pu/MWD	0.264	0.224	0.059	0.062
relative	4.47	3.78	1.00	1.06
grams Pu-239/MWD	0.147	0.122	0.023	0.023
relative	6.48	5.37	1.01	1.00

The spontaneous neutron production and the decay heat production in the various plutonium grades are shown in Table 19. Note that the spontaneous neutron rate from any plutonium separated from the Th-U fuel is more than double that of the conventional fuel and at least 15 times that of either of the weapons grades.

More importantly, the decay heat production is four to five times higher than the conventional fuel and some forty times greater than either of the weapons grades.

In order to estimate the difficulty of handling separated plutonium from any of the cycles, a 6.1-kg mass of plutonium was assumed. The plutonium mass of 5.0 kg was chosen as the approximate size of the Trinity pit (Rhodes 95). The total decay heat for such a mass was calculated and an equilibrium temperature was calculated. This equilibrium temperature assumed that plutonium was in a sphere 88 mm in diameter and that heat removal was via blackbody radiation with a surface emissivity of 1.0. The temperature shown is the surface temperature of the sphere. This heat removal approximates the heat removal from a sphere setting on a table top. While some natural convection may be induced by the heat of the plutonium sphere, free air flow would be impeded by the table itself. Furthermore, the surface of the table would increase in temperature, both insulating the sphere and negating the assumption that the sphere can radiate to a 4π surroundings at 300 K. Thus, a sphere of plutonium derived from the mixed Th-U fuel would likely burn or melt any surface on which it is set. If the sphere is surrounded by a covering for concealment or transport, the temperature would rise still higher. More detailed analysis of the heat transfer from the sphere to naturally circulating air is needed.

Forced convection of air around the sphere would lower the temperatures somewhat, but these cooling methods would require active equipment and/or favorable surrounding airflow paths.

Table 19 Spontaneous Neutron and Decay Heat

Comparison of Spontaneous Neutron and Heat Production						
	Super grade	Weapons grade	U 4.5 Yr, 45 MWD/kg	U 8 % enr, 6 yr, 72 MWD/kg	Th-U 6 yr, 72 MWD/kg	Th-U 10 yr, 100 MWD/kg
Composition						
Pu-238	0.0%	0.012%	2.3%	4.4%	10.8%	13.9%
Pu-239	98.0%	93.80%	55.8%	54.6%	38.9%	36.4%
Pu-240	2.0%	5.80%	21.2%	19.6%	19.8%	19.4%
Pu-241	0.0%	0.35%	14.9%	15.4%	14.9%	14.0%
Pu-242	0.0%	0.022%	5.8%	5.9%	15.6%	16.3%
Spontaneous Neutron Production						
(n/kg-s)	1.82E+04	5.35E+04	3.52E+05	3.93E+05	7.26E+05	8.16E+05
relative	1.0	2.9	19.3	21.6	39.9	44.8
Decay Heat						
(W/kg)	2.0	2.3	16.1	27.8	63.4	80.7
relative	1.0	1.1	8.1	13.9	31.7	40.4
For a 5.0 kg sphere						
Spontaneous Neutrons						
n/s	9.11E+04	2.67E+05	1.76E+06	1.97E+06	3.63E+06	4.08E+06
Decay Heat						
W	10	11	81	139	317	403
Temperature (C)	79	84	236	302	426	468
(F)	174	184	456	576	799	875
(see text for explanation of temperature calculation)						

8.3 Difficulty in Weapons Fabrication

Another consideration is the heating of the high explosive surrounding the separated plutonium. The high Pu-238 content results in a heat generation of about 400 to 500 W for the mixed Th-U fuels, compared with 100 W for the conventional reactor fuel and less than 15 W for the two weapons grades.

While the thermal conductivity for the high explosive (HE) used in U.S. weapons is not available (and may vary), one may surmise that it is about 0.2 to 0.5 W/m-K. This admittedly simplistic analysis assumed a spherical geometry and an HE thickness of 60 mm. Using this range of thermal conductivities, Table 20 shows that peak temperatures at the plutonium-high explosive interface are above the melting/damage point for the HE[1]. For this analysis, the plutonium mixture from the 6 year, 72 MWD/kg thorium-uranium fuels was assumed. Clearly, the use of plutonium containing 10 to 14 percent Pu-238 in a weapon would present a severe thermal management challenge.

Table 20 Peak Temperatures at the Pu-HE Interface

Peak High Explosive Temperatures			
(using Pu from 72 MWD/kg U-Th fuel)			
k HE	T interface		
(W/m-K)	K	C	F
0.2	1714	1441	2626
0.3	1243	970	1778
0.4	1007	734	1354
0.5	866	593	1100
0.75	677	404	760
1	585	312	594
2	441	168	335

8.4 Predetonation due to Spontaneous Neutrons

All plutonium mixtures derived from power reactors substantially reduce the probable yield of a crude weapon because of the neutrons spontaneously emitted during the implosion. The high spontaneous neutron production rate (15 times higher than 'weapons-grade') make detection of the separated plutonium easy, but makes its use in a weapon extremely difficult due to fissioning during the implosion. Both of the Th-U fuels have less than a 7 % probability that the yield will exceed 10 % of the design yield. While 10 % of the design yield is still a terrible blast, these probabilities would make plutonium derived from the thorium-uranium fuel much less desirable to potential terrorists.

9 Wasteform Characteristics

A fuel initially consisting of a solid solution of ThO_2 and UO_2 may be required to serve as the final wasteform after an extended irradiation. To reliably contain the fission products and

[1] High explosives, such as RDX, melt in the 200° C range. The explosive may still function in a liquid state, but the weapon would have lost the necessary precision in its explosive configuration.

actinides within the fuel, the fuel must not undergo significant chemical reactions and resultant changes in crystal structure. Thorium dioxide is the highest oxide of thorium and does not depart significantly from its stoichiometric composition of ThO_2 at temperatures below 2000 K (Belle 84, p 131). Uranium dioxide, on the other hand, is oxidized to O/U ratios exceeding 2.67 (e.g. U_3O_8) in periods of less than a month when exposed to oxygenated water at temperatures greater than 377 K (see Belle 61, p. 398). ThO_2-UO_2 mixtures appear to be susceptible to some corrosive attack in oxygenated water, but much less susceptible than UO_2.

To quote Belle:

> The behavior of ThO_2 and various ThO_2-UO_2 compositions up to 50 weight percent UO_2 was studied in high temperature water (633 K), alkaline (pH 10) flowing water, both degassed and oxygenated (5 and 100 ppm) as part of a study of the behavior in such media of various oxide nuclear fuels including UO_2. The results of the exposure tests confirmed early work (Belle 61) in showing that sintered compacts of UO_2 are extremely resistant to corrosive or erosive attack in high temperature water in the absence of oxygen, but corrode rapidly in the flowing, oxygenated high pH water system. The ThO_2 samples displayed good corrosion resistance even in oxygenated high-temperature water; this result is not surprising since thorium forms no higher oxide that ThO_2. In the case of the ThO_2-UO_2 solid solutions, no corrosion attack was noted for any of the oxide compositions in either the non-oxygenated water or in the water containing 5 ppm O_2.
>
> Exposure of ThO_2-UO_2 solid solution of two compositions (ThO_2-20 weight percent UO_2 and ThO_2-50 weight percent UO_2) to water oxygenated to 100 ± 10 ppm the highest level tested, showed high weight gains. However, all the specimens retained their mechanical integrity throughout the test period of 140 days, forming a surface-oxidized phase. This oxidized phase was found, by x-ray diffraction analysis, to be cubic, probably of the M_4O_9 type. This result suggests that an M_4O_9 type of single-phase region exists in the low temperature ternary phase diagram for ThO_2-UO_2-O_2.
>
> (Belle 84, pp. 231-2)

10 Conclusions

Several important conclusions can be drawn from this analysis.

The mixed Th-U fuel, using a mixture of 25 weight percent UO_2 and 75 weight percent ThO_2, where the uranium is initially enriched to 19.5 % U-235, appears to have sufficient reactivity to be used for extended burn cycles in LWRs. Likewise, a mixture containing 35 weight percent UO2, with the same enrichment, and 65 weight percent ThO2, appears suitable for extended cycles approaching 100 MWD/kg of initial heavy metal. The *in situ* breeding of U-233 maintains a more uniform reactivity during the course of irradiation and eliminates the need for burnable poisons.

The mixed Th-U fuel reduces the amount of total plutonium production by a factor of 4.5 and the Pu-239 production by a factor of 6.5, when compared to conventional UO_2 fuel irradiated to 45 MWD/kg ihm.

The cost per unit energy of the mixed Th-U fuel is 13 to 25 % less than the conventional or extended burn UO2 fuels, if present uranium prices are used. If projected future prices of uranium and thorium are used, the advantage for mixed Th-U fuels is 20 to 33%.

At no time during the fuel cycle can a uranium component be chemically separated from the fuel that could be usable in a nuclear weapon.

The plutonium that is produced in the mixed Th-U fuel is high in Pu-238, producing copious amounts of decay heat and spontaneous neutrons. The high decay heat would not allow a 6.1 kg sphere of the plutonium to be placed on a tabletop without burning through the table. The decay heat would also melt or burn any surrounding explosive used in fashioning a crude weapon. The spontaneous neutrons drastically limit the probable yield of any such crude weapon.

The fission and capture cross-sections of U-233 and Pu-239 are such that U-233 is much more likely to fission, while Pu-239 has a 30% probability of forming a higher nuclide.

A matrix of ThO_2 and UO_2 is more resistant to long-term corrosion in oxygenated water than is a matrix of only UO_2. Thus $ThO_2 – UO_2$ is a superior wasteform if the spent fuel is slated for direct disposal rather than reprocessing.

References

Belle 61 J. Belle, editor, *Uranium Dioxide: Properties and Nuclear Applications*, Naval Reactors, Division of Reactor Development, United States Atomic Energy Commission, Washington, DC July 1961.

Belle 84 J. Belle and R. M. Berman, editors, *Thorium Dioxide: Properties and Nuclear Applications*, DOE/NE-0060, Naval Reactors Office, U. S. Department of Energy August 1984.

Galperin 95 Alex Galperin and Alvin Radkowsky, "Thorium Fuel for Light Water Reactors – Reducing Proliferation Potential of Nuclear Power Fuel Cycle, INIS-MF-15507.

McLane 88 Victoria McLane, Charles L:. Dunford and Philip F. Rose, *Neutron Cross Sections,* (successor edtion to BNL-325), National Nuclear Data Center, Brookhaven National Laboratory, Upton, New York, Academic Press, 1988.

NUREG/CR-0200 "SCALE: A Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluation," NUREG/CR-0200, Rev. 5 (ORNL/NUREG/CSD-2?R5), Vols I, II, and III (Draft September 1995).

Rhodes 95 Richard Rhodes, *The Making of the Atomic Bomb,* Touchstone Books, 1995.

Serber 43 Robert Serber, *The Los Alamos Primer: The First Lectures on How to Build an Atomic Bomb* (Berkeley, CA; University of California Press, 1992). (Notes on lectures given by Serber in 1943 at the start of the Los Alamos Laboratory, declassified circa 1965.)

USGS-Th-96 James B. Hedrick, "Thorium," U.S. Geological Survey – Minerals Information, 1996, available at http://minerals.er.usgs.gov/minerals/pubs/commodity/thorium/

Appendix A: Very Long Cycle Using Mixed Thorium and Uranium Oxide Fuel

This fuel cycle was analyzed to determine the feasibility of a very long cycle in which the specific power was reduced to 18.97 MWth/kg ihm. The reactor would operate for four years between refueling outages, which would be 180 days in length. This operating cycle is designed for a remotely located power reactor that would be transported to a location off-shore from a developing country, generate 600 MWe for a four-year period and then be transported to a central facility for refueling and maintenance. The reactor may be remotely controlled from either an on-shore facility in the host country or from a central control facility serving many such reactors. The reactor deployment scheme would require a very long operating cycle, relatively low power density and great proliferation resistance. In addition, the corrosion resistance of the ThO_2 fuel matrix in oxygenated water would be very helpful in mitigating accidental releases.

The parameters of the cycle are shown in Table 1, while the parameters of the B&W fuel assembly are shown in Table 2 and the reactor conditions are shown in Table 3. The neutron multiplication factor for a large array of assemblies is shown in Table 21. The neutron multiplication is slightly higher than that in the 6-year, 72 MWD/kg case in the main text) where the specific power was 37.935 kWth/kg ihm because the equilibrium inventory of the intermediate Pa-233 (half-life 27 days) is smaller and thus fewer neutrons are lost to its 460 b resonance integral. The uranium content and composition are shown in Table 22.

Table 21 Neutron Multiplication Factors in the Thorium -Uranium Cycle, 13-yr, 72 MWD/kg

Fuel Assembly k infinity						
		13-year, 3-batch cycle to 72 MWD/kg, 25 wt% initial uranium (days since restart)				
Batch	Specific Power (MWth/kg HM)	0.0	365	731	1096	1461
first	18.97	1.2453	1.1885	1.1833	1.1467	1.1107
second	18.97	1.1688	1.1031	1.0981	1.0550	1.0164
third	18.97	1.0719	1.0045	0.9930	0.9550	0.9248
core-average	18.97	1.1620	1.0987	1.0915	1.0523	1.0173

Note: SCALE recalculated the 44-group flux at the beginning and midpoint of each cycle, resulting in a small adjustment in k infinity.

Table 22 Uranium Content and Composition, Very Long Th-U cycle

Uranium Content during 13-yr, 72 MWD/kg 3-batch cycle (kg/tonne initial heavy metal)								
		Burnup, MWD/kg IHM						
Isotope		0	12	24	36	48	60	72
U	233	0.00	4.70	7.98	10.03	11.24	11.79	11.99
U	234	0.00	0.17	0.51	0.95	1.45	1.96	2.49
U	235	48.92	35.57	25.35	17.45	11.48	7.23	4.37
U	236	0.00	2.54	4.32	5.54	6.40	6.86	7.06
U	238	201.9	199.3	196.2	193.7	191.1	188.0	185.5
U		250.8	242.3	234.4	227.7	221.7	215.9	211.4
(weight percent of total uranium)								
		Burnup, MWD/kg IHM						
Isotope		0	12	24	36	48	60	72
U	233	0.0%	1.9%	3.4%	4.4%	5.1%	5.5%	5.7%
U	234	0.0%	0.1%	0.2%	0.4%	0.7%	0.9%	1.2%
U	235	19.5%	14.7%	10.8%	7.7%	5.2%	3.4%	2.1%
U	236	0.0%	1.0%	1.8%	2.4%	2.9%	3.2%	3.3%
U	238	80.5%	82.3%	83.7%	85.1%	86.2%	87.1%	87.7%
fissile		19.5%	16.6%	14.2%	12.1%	10.2%	8.8%	7.7%

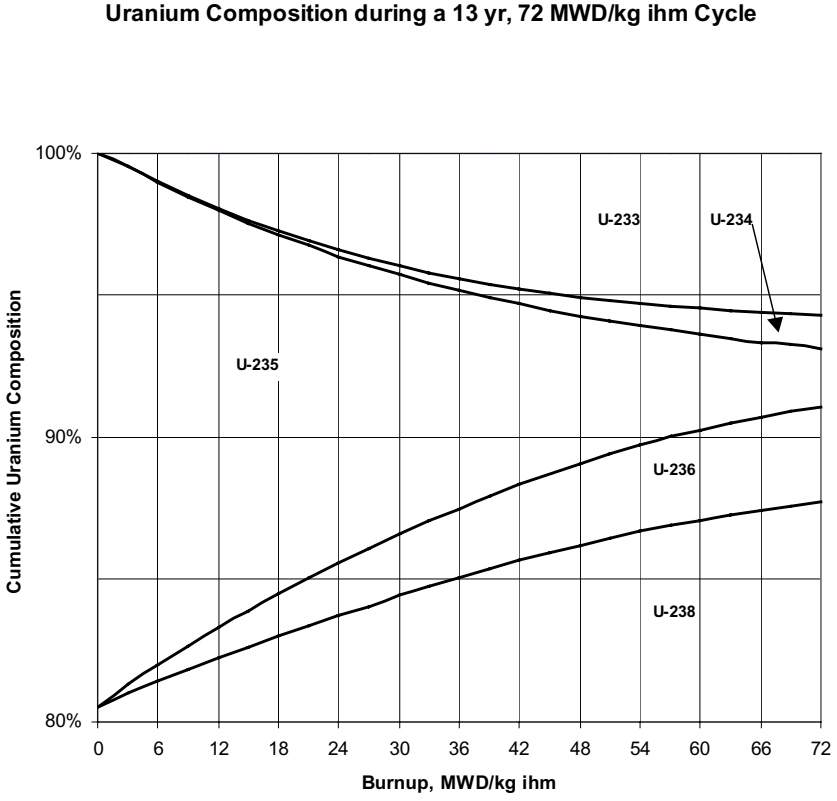

Figure 15 Uranium Composition during a 13 yr, 72 MWD/kg ihm cycle

The plutonium content and composition of the thorium-uranium fuel during the 13-year, 72 MWD/kg ihm irradiation are shown in Table 23 and Figure 16.

Table 23 Plutonium Content and Composition, Very Long Th-U Cycle

Plutonium Content during 13-yr, 72 MWD/kg cycle								
(kg/tonne initial heavy metal)								
		Burnup, MWD/kg IHM						
Isotope		0	12	24	36	48	60	72
Pu	238	0.000	0.008	0.047	0.127	0.241	0.371	0.492
Pu	239	0.000	1.590	2.063	2.099	1.971	1.806	1.662
Pu	240	0.000	0.227	0.489	0.667	0.770	0.822	0.842
Pu	241	0.000	0.117	0.371	0.555	0.638	0.638	0.612
Pu	242	0.000	0.009	0.067	0.181	0.331	0.489	0.641
Pu		0.000	1.951	3.037	3.629	3.950	4.125	4.250
(weight percent of total plutonium)								
		Burnup, MWD/kg IHM						
Isotope		0	12	24	36	48	60	72
Pu	238	0.0%	0.4%	1.5%	3.5%	6.1%	9.0%	11.6%
Pu	239	100.0%	81.5%	67.9%	57.9%	49.9%	43.8%	39.1%
Pu	240	0.0%	11.7%	16.1%	18.4%	19.5%	19.9%	19.8%
Pu	241	0.0%	6.0%	12.2%	15.3%	16.2%	15.5%	14.4%
Pu	242	0.0%	0.5%	2.2%	5.0%	8.4%	11.8%	15.1%
fissile		100.0%	87.5%	80.2%	73.1%	66.0%	59.2%	53.5%

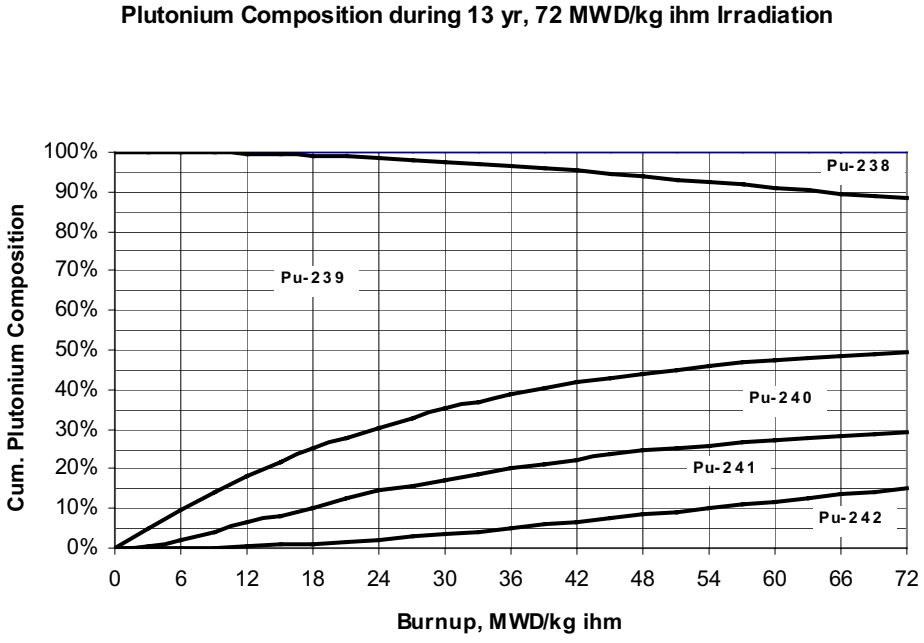

Figure 16 Plutonium Composition during 13 yr, 72 MWD/kg ihm Irradiation

The plutonium production of the cycle is summarized in Table 24. Note that total amount of plutonium produced is about a factor of 4.5 less than that of the UO_2 fuel irradiated in a 4.5-year cycle to 45 MWD/kg ihm. The production of Pu-239 is about a factor of 6.5 less than the base case.

Table 24 Comparison of Plutonium Production in 13 year Cycle with Base Case

Plutonium Production in 13 year Th-U Cycle		
		Th-U 13 yr, 72 MWD/kg
Burnup (MWD/kg)		**72**
Production	**fraction**	**gram/kg ihm**
Pu-238	11.6%	0.492
Pu-239	39.1%	1.662
Pu-240	19.8%	0.842
Pu-241	14.4%	0.612
Pu-242	15.1%	0.641
Total Pu	100%	4.250
Production per MWD		
grams Pu/MWD		0.059
relative to base case		4.48
grams Pu-239/MWD		0.023
relative to base case		6.38