ACTINIDE BURNING IN A STANDARD PRESSURIZED WATER REACTOR

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ABSTRACT

This study investigates actinide burning in a standard lattice PWR. It assumes that after the first loading of depleted uranium, plutonium and minor actinides, only the fission products are removed and only depleted uranium and the plutonium/minor actinide mixture are added. The relative concentrations of plutonium and minor actinide feed are always maintained the same as PWR discharge fuel after 33,000 MWD/MTU burnup. It is found that the minor actinides act as a burnable absorber. After seven reloading, a burnup of 64,000 MWD/MTU yields a reactivity change of less than 3% $\Delta k/k$. This property of the minor actinides suggests that the minor actinides should not be separated from the plutonium. In equilibrium, one actinide burner would be required for every two standard PWRs.

I. INTRODUCTION

Without reprocessing the toxicity of spent fuel remains higher than that of the ore for nearly 1,000,000 years.[1] Actinides dominate the waste toxicity after a couple of hundred years. If the fuel is reprocessed and only the plutonium actinides are removed, the isolation period needed to yield the same toxicity as the original ore is from 1000 to 10,000 years. If all the actinides could be removed it would be reasonable to only require confinement of the waste for about 600 years. If one were to assume that plutonium is a fuel then the objective of actinide burning is to remove the non-uranium/non-plutonium (minor) actinides. If, on the other hand, one is interested in lowering the waste toxicity from the current U.S.A. waste then burning of the plutonium along with the minor actinides is the objective.

To remove an actinide it must be fissioned. Neutron capture only creates another actinide. The minor actinide fission cross section is generally higher in a fast reactor so the fast reactor actinide burner has dominated the research. The plutonium fission cross section, however, is higher in the thermal reactor. The energy per fission is about the same in a fast or thermal reactor so a 600 MWe fast or thermal actinide burner would burn about the same amount of material. The fast reactor would selectively burn more of the minor actinides and less of the plutonium.

Fast reactors generally are perceived to have a higher capital cost than PWRs. This higher cost was initially supportable by expectations of high uranium ore costs and hence a desire to make plutonium. Since ore Costs have remained low and are not expected to dramatically rise in the near future, fast reactors are not under demand. If plutonium is perceived to have a negative value, it is not clear that fast reactors would have a cheaper fuel cycle cost than a mixed oxide fueled PWR. Along with a lower capital cost, the PWR clearly has much more operating experience. It may not be worth the expense of a typical learning curve if the fast reactor was just to burn actinides. This study investigates burning actinides in PWRS

II. METHOD

Since actinide burning requires more than ten cycles to approach equilibrium, spatial studies are prohibitively expensive. Although spatial studies have been attempted for the LMR, no such studies have been performed for thermal reactors. This implies that any actinide burning study must make significant assumptions. Two key assumptions found in the literature have been: 1) Constant one group cross sections, and 2) Constant beginning of life (BOL) k. The constant one group cross section assumption is normally made with the ORIGEN[2] code. Since the isotopics are changing greatly from the starting condition, the spectrum is changing as is the one group

cross section. For this study, burnup is performed with one group cross sections; but they are updated each cycle using COMBINE[3] (4 times during the fuel lifetime in the core). Since the reactivity change as a function of burnup decreases with increasing actinides, the BOL k used should change. This is done for this study by iterating on the BOL k in order to match the desired cycle length.

The PWR modeled for this work was the AP-600. However, the actual model was a pin cell model consisting only of a fuel pellet surrounded by a zirconium clad and then water. All the cases were done without soluble boron. Since it was determined that the lack of soluble boron made the flux on average too thermal, it was decided not to include the water holes or the assembly gap in the model. Thus the model area was just the pin pitch squared. Full power material temperatures and densities are used. This model was compared to a LEOPARD[4] model for the fresh 3.2 w/o U-235 starting case and good agreement was found. This same model is used for all loadings of the actinide fuel. No changes in the fuel pin diameter or pitch were made for any case. This implies all cases would seem to be fully backfitable in the current PWR plants. (Safety criteria with regard to control rod worths, MTC, and Doppler were not checked so some modifications would be likely.)

COMBINE did not have the capability to do burnup so a computer code was written that took the COMBINE output and performed burnup based on the method in 2DB.[5] This computer code also automated the reloading and enrichment search. Further details of the method used in this work were previously presented by Stone.[6,7] Although there is considerable uncertainty remaining, it is believed that the approach taken in this study is the best done to date.

For Stone's thesis he compared his results to that of a study by Gorrell.[8] This comparison gave a general confidence in the method but the level of agreement expected was fairly low since different methods were used. In order to look for finer details, a simpler problem was selected. It was decided to burn 3.2 w/o PWR fuel for 33,000 MWD/MTU, then let it decay 3 years, and compare the isotopics to two references. The first reference comes out of ANL. The paper was presented at the topical meeting in Marseilles in 1990.[9] In 1992 Downar[10] is still using the same isotopics The second paper is out of JAERI.[11] This paper was presented at a topical meeting in Kyoto in 1991. It is a disappointment that for such a seemingly simple problem that is so basic to actinide burning (and normal once through waste storage) such large discrepancies exist. This study agrees best with the JABRI work. Table 1 shows the comparisons that can easily be made.

Since fairly good agreement seems to exist between this work and the JAERI work it is believed that the method was sufficiently accurate to continue the study. It was shocking that the two major laboratories used as a reference were so different on this test (a 14% disagreement in the main minor actinide, Np-237, and close to a factor of two for Am-243). Np-237 and Am243 are very important in any of the waste disposal work so it was believed that better agreement would exist.

The loading scheme assumes that there is a large amount of spent PWR fuel with a burnup of 33,000 MWD/MTU. This fuel is reprocessed but the all transuranic isotopes are kept together. The fuel pellets are a mixture of all transuranic actinides as fuel and depleted uranium as a diluent. It was assumed that the transuranic actinide mixture was stored ten years after being discharged from the PWR. The isotopic content of this fuel is given in Table 2. Notice that this fuel has about five percent Np-237 and due to the significant decay of Pu-241 it has about five percent Am-241. The Pu-241 content is down to seven percent. This mixture contains 88.7% Pu and 11.3% minor actinides. At the end of each fuel lifetime the fission products are removed and a mixture of depleted uranium and the 33,000 MWD/MTU mixture of Table 1 is used to add the appropriate reactivity.

The fuel cycle scheme selected for the actinide burning assumed a four batch core with an average discharge burnup of 64,000 MWD/MTU. In an AP600 type core this would mean each cycle was about two years. It was assumed that the effect of each batch on the keff was the same. At the end of any cycle there would be assemblies with 1, 2, 3 and 4 cycles of burnup. This means the average burnup in cycles is 2.5. The beginning of life (BOL) k needed was calculated as 2.5/4 times the delta k for 64,000 MWD/MTU. Since the change in k with burnup was dependent on the isotopics, the BOL k was determined through iteration.

TABLE 1: **Isotopics** After 33,000 **MWD/MTU** Burning in a PWR

Isotope	Fraction of Minor **Actinides** This Study	JAERI[11]	ANL[9]
Np-237	.569	.562	.491
Am-241	.263	.264	.227
Am-243	.128	.120	.225
Cm-243	.0004	.0003	.0007
Cm-244	.0361	.0511	.050
Cm-245	.0021	.0028	.0046
Fraction of Each Isotope Among the **Pu** Isotopes			
Pu-238	.0169		.0114
Pu-239	.5615		.571
Pu-240	.256		.224
Pu-241	.111		.151
Pu-242	.0546		.0436
Ratio of Minor **Actinides** to **Pu** Isotopes			
	.0899		.124

Table 2: Isotopic Fractions **of Transuranics** in PWR Discharged Fuel **(33,000 MWD/MTU)**

Isotope	Fraction
Np-237	.04737
Pu-238	.01468
Pu-239	.51511
Pu-240	.23587
Pu-241	.07097
Pu-242	.05020
Am-241	.05277
Am-243	.01060
Cm-244	.00226
Cm-245	.00017

III. RESULTS

Table 3 shows the beginning and end of life k for each fuel loading. This is the delta k for 64,000 MWD/MTU burnup. Notice that the delta k with burnup is extremely low. For a standard uranium fueled PWR the delta k would be about 0.6. The first loading shows a factor of over 5 savings in delta k. By the seventh loading the delta k is about 3%. This is a factor of 20 less than that for a normal uranium fueled PWR. This implies that the actinides work as excellent burnable absorbers. There could be significant advantages due to this effect. For example, the moderator temperature coefficient could be more negative since less soluble boron would be required. Power distribution control could be easier for these long cycles since the k of all the fuel would be similar. Criticality accidents would also be less likely due to the lower excess reactivity. The down side of this is that any uncertainty in the cross sections could have a major effect on the results. Since there is a large amount of rarely used isotopes in these cores this uncertainty concern is serious.

Table 3: Beginning and End of Life K_{eff} For Each Fuel Loading For **PWR Actinide** Burning

Fuel Loading	k_{eff} BOL	k_{eff} EOL
1	1.072	.9613
2	1.034	.9809
3	1.029	.9835
4	1.025	.9857
5	1.022	.9867
6	1.021	.9885
7	1.019	.9889
8	1.018	.9894
9	1.017	.9897
10	1.017	.9905
11	1.017	.9912

Table 4 shows the actinide contents at the end of each fuel lifetime. As can be seen from this table, the reactor is actually an actinide storage device. The quantities of the minor actinides are still small. After ten reloading there would only be 1.2 MT, 0.8 MT and 1 kg of Am, Cm and Cf respectively in the core. This is out of 66 MT of heavy metal in the core. Each loading represents 8 years so Table 4 represents 80 years of operation of actinide burners. In these 80 years few of the minor actinides have reached equilibrium. However, none of the actinides are changing more than 21% between loadings.

Table 5 shows the quantities of the feed actinides in each cycle. The first loading shows the total mass in the core. The following loadings are feed material used to replace the fission products. It is assumed that the feed isotopes are blended with the fuel from the previous loading that did not fission. PWRS produce about 1 gm actinides/MWD or about 25,000 kg/yr with our current operating PWRs. Almost all of the fissioning is in transuranic actinides. Since the energy per fission is nearly a constant one could calculate that the

actinide burning PWRs burn about 575 kg per year. This implies that the 25,000 kg/yr produced would be roughly what a 600 MWe actinide burner could burn in its lifetime. Therefore, it would require a new actinide burner for each year of operation of the current reactors. One would also need on the order of ten of these reactors to reduce the current backlog. Since no region in the U.S.A. can use an addition of 600 MWe each year, it can be concluded that several sites would have actinide burners. It can also be concluded that the addition rate of actinide burners would require less that a 1% growth rate per year of nuclear electricity which is less than the current electrical growth rate.

Table 6 normalizes the data by the energy produced. From this table it can be observed that for about every two new reactors ordered an actinide burner would be required. The actinides represent an attractive fuel and since the minor actinides act as burnable absorbers the advantage of removing the minor actinides from the Pu is not significant.

IV. CONCLUSIONS

Actinide burning in a PWR was found not only to be possible but would have significant benefits. The minor actinides were found to act as excellent burnable absorbers for the PWR core. The actinide burner would start with 90% depleted uranium and 10% transuranic actinides. This enrichment would allow a 64,000 MWD/MTU burnup. All of the following cycles could be done by removing only the fission products and adding the transuranic mixture and some depleted uranium. This concept never requires separation of the plutonium from the other transuranic actinides.

If the objective of actinide burning is to lower the toxicity of the once through fuel cycle, then burning plutonium as well as the minor actinides is desirable. Since the energy per fission is about the same for a fast reactor as for a thermal reactor, the PWR will burn actinides as well as the fast reactor. The advantage of the PWR is the lower capital cost and the many years of operating experience. Both the fast and thermal actinide burners use only waste products as fuel. In equilibrium, a 600 MWe PWR actinide burner would burn about 575 kilograms of transuranic actinides a year. In equilibrium about one third of the reactors would have to be actinide burners.

The actinide burning PWR has a very low reactivity change with burnup. This implies very low soluble boron concentrations are needed. Also due to the low excess reactivity it would seem that criticality accidents are less likely. There may be safety advantages to the PWR actinide burner over a standard PWR.

V. REFERENCES

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Table 4: Inventory (Kg) of Actinides in a 600 MWe PWR Actinide Burner

End of Load	U234	u235	U236	u238	Np237	Pu23 8	PU23 9	Pu240	Pu241
1	24.	46.9	17.1	5.69E+4	108.	411.	1660.	1500.	496.
2	57.	26.6	19.4	5.42E+4	131.	715.	2320.	2330.	660.
3	90.	19.5	19.2	5.26E+4	128.	924.	2550.	2810.	730.
4	120.	16.7	18.7	5.14E+4	126.	1080.	2730.	3180.	777.
5	148.	15.6	18.5	5.05E+4	124.	1190.	2850.	3470.	809.
6	172.	15.2	18.5	4.97E+4	125.	1280.	2960.	3720.	837.
7	193.	15.3	18.8	4.90E+4	125.	1350.	3030.	3930.	857.
8	212.	15.6	19.3	4.85E+4	125.	1410.	3090.	4100.	873.
9	228.	16.0	19.8	4.80E+4	126.	1460.	3130.	4250.	887.
10	242.	16.4	20.4	4.76E+4	128.	1500.	3190.	4390.	900.

End of Load	Pu242	Am241	Am242	Am243	Cm242	Cm243	Cm244	Cm245	Cm246
1	476.	249.	2.56	148.	.0355	1.50	124.	25.9	3.42
2	822.	394.	4.66	231.	.0511	2.10	231.	56.3	11.5
3	1090.	469.	5.77	286.	.0586	2.39	311.	79.5	22.3
4	1310.	524.	6.59	329.	.0637	2.58	372.	96.8	34.2
5	1500.	566.	7.20	364.	.0672	2.70	419.	110.	46.1
6	1680.	603.	7.74	394.	.0703	2.80	455.	120.	57.5
7	1820.	630.	8.13	420.	.0724	2.88	485.	128.	68.3
8	1960.	654.	8.47	442.	.0743	2.94	510.	135.	78.2
9	2070.	674.	8.75	462.	.0758	2.99	530.	141.	87.3
10	2180.	694.	9.03	480.	.0773	3.03	548.	146.	95.5

End of Load	Cm247	Cm248	Bk249	cf249	Cf250	cf251	Cf252
1	.122	.019	1.77E-5	8.06E-4	4.47E-5	3.33E-5	5.49E-6
2	.608	.185	2.13E-4	1.17E-2	1.26E-3	1.04E-3	2.47E-4
3	1.40	.616	7.58E-4	4.56E-2	6.25E-3	5.34E-3	1.52E-3
4	2.35	1.33	1.69E-3	1.07E-1	1.69E-2	1.46E-2	4.53E-3
5	3.33	2.26	2.94E-3	1.93E-1	3.32E-2	2.90E-2	9.40E-3
6	4.29	3.34	4.40E-3	2.96E-1	5.44E-2	4.76E-2	1.58E-2
7	5.18	4.50	5.98E-3	4.10E-1	7.86E-2	6.90E-2	2.32E-2
8	6.01	5.68	7.59E-3	5.28E-1	1.05E-1	9.20E-2	3.12E-2
9	6.77	6.84	9.19E-3	6.45E-1	1.32E-1	1.16E-1	3.94E-2
10	7.45	7.96	1.07E-2	7.60E-1	1.59E-1	1.39E-1	4.75E-2

Table 5: Kilograms Inserted Each Loading in a 600 MWe PWR Actinide Burner

Loading	Depleted Uranium	Plutonium Mixture	Minor Actinides	Total Non U Actinidee
1	59949	5907	754	6661
2	24	4013	512	4525
3	1088	3081	393	3475
4	1355	2863	365	3228
5	1563	2694	344	3037
6	1621	2655	339	2994
7	1770	2532	323	2855
8	1817	2499	319	2818
9	1876	2454	313	2767
10	1870	2465	315	2779
11	1887	2454	313	2768

Table 6: Grams Inserted Per MWD Electricity Produced in a PWR Actinide Burner

Loading	Depleted Uranium	Plutonium Mixture	Minor Actinidea	Total Non U Actinides
1	43.95	4.33	0.55	4.88
2	0.02	2.94	0.38	3.32
3	0.80	2.26	0.29	2.55
4	0.99	2.10	0.27	2.37
5	1.15	1.97	0.25	2.23
6	1.19	1.95	0.25	2.19
7	1.30	1.86	0.24	2.09
8	1.33	1.83	0.23	2.07
9	1.38	1.80	0.23	2.03
10	1.37	1.81	0.23	2.04
11	1.38	1.80	0.23	2.03