A CRITICAL ESSAY

INCENTIVES AND RECENT PROPOSALS FOR PARTITIONING AND TRANSMUTATION IN THE UNITED STATES

BY

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DISCLAIMER

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1.0 INTRODUCTION

Thirteen years after the passage of the National Waste Policy Act (NWPA) by Congress, after a series of political and technical problems, a permanent solution to the disposal of high level waste and spent nuclear fuel still eludes the United States. Although this solution may be forthcoming by the year 2010 in the form of a geological repository at Yucca Mountain, Nevada, there remain quite a few obstacles to be overcome. At the present time work is being done to characterize the site in order to determine its suitability while at the same time efforts are being made to site and construct a Monitored Retrievable Storage Facility. This MRS facility would serve as an intermediate point for the storage of spent fuel and other high level waste awaiting permanent disposal in the repository, and becomes more necessary every year as more utilities find their own storage capabilities exhausted.

Since the 1970s it has been established that disposal in deep geological repositories is the most acceptable fate for high level waste. Following this policy the NWPA's original objective in 1982 was to establish two national repositories, with one in the western U.S. and the second in the east. By 1987 three sites in the west had been identified as good candidates for further study. However efforts to find suitable sites in the east were blocked by intense local opposition and as a result the NWPA was amended in 1987 and the search for a second repository location was suspended.

The three western sites initially chosen for consideration were in Nevada, Texas, and Washington. Both Yucca Mountain in Nevada, and Hanford in Washington were in federal lands while the Texas site was in Deaf Smith County, in panhandle country. All three possessed very different geologies and after preliminary evaluations it was

concluded that the Yucca Mountain site in Nevada was the most favorable. Yucca Mountain was then designated the first candidate repository in the amendment of 1987 and a full scale characterization was undertaken by several of DOE's national laboratories. At that time the schedule was in place for a repository to begin operation and acceptance of spent fuel and defense high level waste by 2003. This date assumed that the Yucca mountain site would prove to be an acceptable environment for such a repository, which is likely but not assured by any means. The lack of a secondary candidate site makes the prospect of Yucca failing the characterization a worrisome one for all nuclear utilities.

The revised starting date of 2003 has since been further revised to 2010. This is in part due to the huge amount of preliminary work which must be done in order to complete the site characterization and obtain reliable assessments of, among other things, the hydrology, volcanology, geology, and ecology of the site. In addition to the technical hurdles which must be overcome there is also the profound opposition of the government and general populace of Nevada to the project. The state's major criticisms are based on Congress' decision in 1987 to characterize only one site and on various technical issues regarding the suitability of the site itself. Nevada has pursued every available option in order to stall or prevent the characterization process. The state government has even passed legislation preemptively refusing to host a repository although this can be overruled by the president should the need arise.

The attempt to site a monitored retrievable storage (MRS) facility has also run into problems and delays. Many of these are tied to concerns of local authorities that the MRS could become a de facto high level waste repository itself if the repository

project is finally defeated. The MRS is essential if the DOE is to be capable of receiving spent fuel in 1998, as the NWPA specifies.

In the late 1980s and early 90s it began to appear as though the Yucca Mountain Site Characterization Project was plagued with more troubles then it could reasonably hope to overcome. Utilities, growing anxious over the possibility that no solution would be found, began pushing for new ideas. As a result some questions were raised as to whether or not it might be possible to implement some new technological policy which might help to eliminate some of the waste disposal uncertainties and therefore expedite the whole process. This challenge was met by several of the national laboratories in independent studies and the results were interesting. What resulted was the re-emergence of the concept of partitioning and transmutation as a means of separating key problem radionuclides from the waste stream and transmuting them, thereby eliminating them as a problem in waste disposal.

Partitioning and transmutation is perhaps the most elegant means of high level waste disposal. Currently, the cost of fuel obtained from reprocessing spent fuel exceeds the cost of fuel obtained by mining. This has resulted in the once through fuel cycle dominating the U.S. nuclear industry. Despite this fact P-T continues to be examined and debated by the U.S. as well as abroad. The United States first seriously considered P-T between approximately 1976 and 1982 but rejected the concept in favor of reprocessing. More recently, since about 1989, as a result of the once through fuel cycle and the growing problems of waste disposal, studies concerning P-T have resumed. This essay will seek to outline the incentives and goals of partitioning and transmutation as it would apply to the disposal of spent fuel in the U.S.. Recent proposals by various U.S. national laboratories for implementing partitioning and

transmutation as a high level waste management and disposal device will also be discussed. The review will seek to examine the technical concepts utilized in each of the proposals and their feasibility. The major focus of this essay will be the transmutation methods themselves, while the partitioning methods will be discussed only briefly. This is because of the fact that partitioning methods fall under reprocessing as an already fairly well established and accepted technology while feasible methods for transmutation are still being advanced.

2.0 INCENTIVES FOR PARTITIONING AND TRANSMUTATION AND THE CURE CONCEPT

The National Waste Policy Act as amended in 1987 defined the course and requirements for disposal of spent fuel and high level nuclear waste in the United States. The Department of Energy holds responsibility for final disposal of these wastes in a geological repository, tentatively chosen at the Yucca Mountain in Nevada. This site is currently undergoing an extensive characterization process to assess its suitability as a high level waste repository. The primary goal of the characterization is to predict how radionuclides placed as waste within the repository will migrate away from the site over a period of 10,000 years under a variety of possible conditions. This time period is specified by the NWPA and a prediction of this magnitude can only be made based on an extensive knowledge of the site's hydrology, volcanology, and geology.

The current fuel cycle for the United States Nuclear industry is called a once through cycle. This cycle does not utilize reprocessing to salvage usable fuel from its spent fuel. Instead the spent fuel is stored temporarily and then disposed of as shown in Figure 1. The second cycle is referred to as a closed cycle and is also denoted in

Figure 1. The closed cycle uses reprocessing technology to recover uranium and plutonium from spent fuel in order to recycle it back to the reactor as fuel. At present, due to the economic reasons just mentioned and fears associated with plutonium diversion and proliferation there is no commercial reprocessing being done in the U.S. However it's use is not forbidden. Therefore, the current characterization is based primarily on spent fuel from both pressurized water reactors (PWR) and from boiling water reactors (BWR) as its reference waste form. It is entirely possible that preliminary treatment of spent fuel, in the form of reprocessing and partitioning and transmutation could serve to reduce difficulties associated with the 10,000 year performance assessment and lead to a more rapid solution of the problem of permanent disposal. This idea was presented in the late 1980s and since then there have been several studies in the U.S. and abroad which sought to examine the feasibility of P-T as a high level waste disposal method. A 1990 report by Westinghouse Hanford - Pacific Northwest Laboratory entitled CURE - Clean Use of Reactor Energy (Ref. 1) discusses in depth the goals of P-T and outlines many of the basic requirements for success.

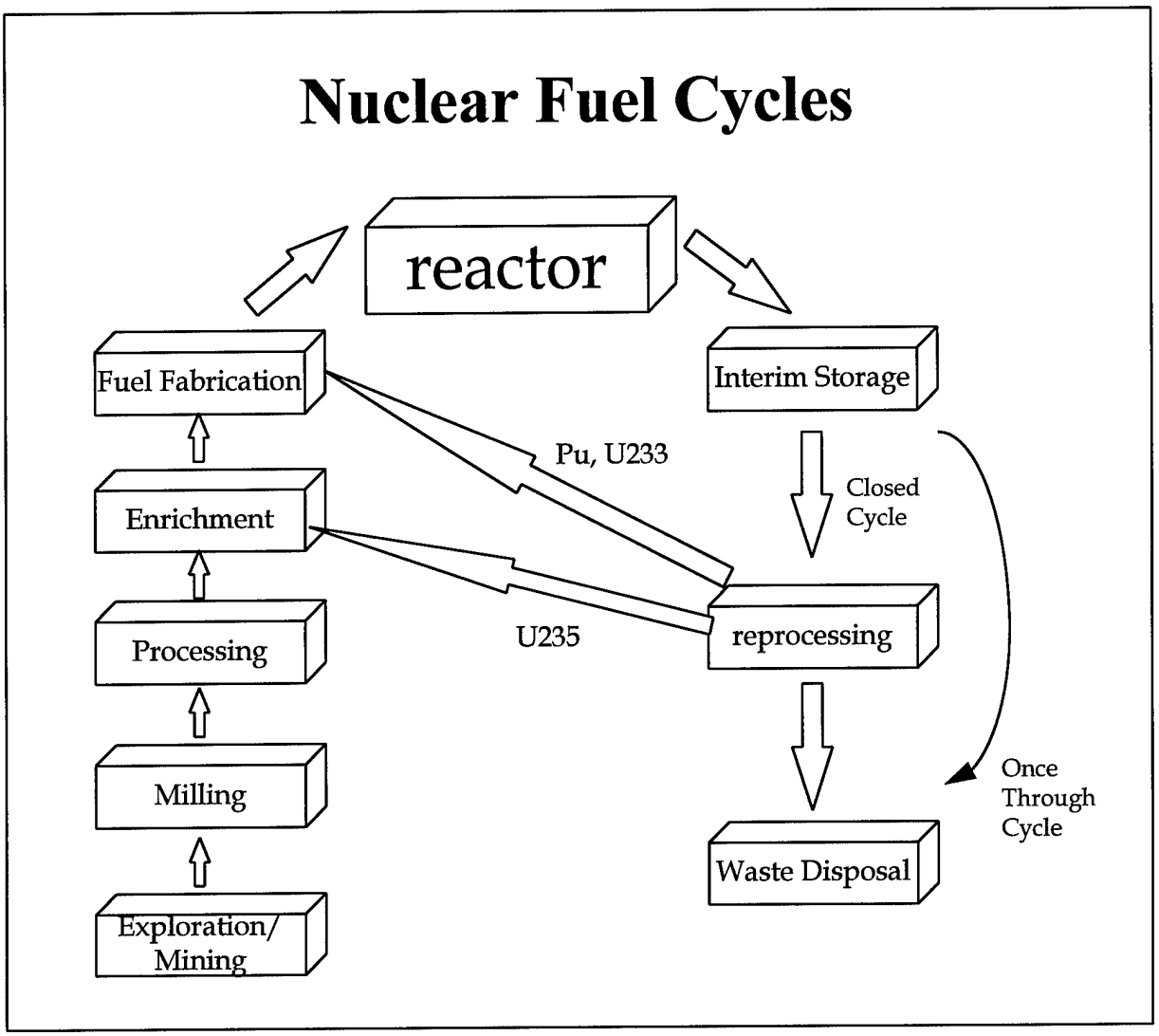

Nuclear Fuel Cycles
reactor
Fuel Fabrication
Enrichment
Processing
Milling
Exploration/
Mining
Interim Storage
Pu, U233
Closed
Cycle
U235
reprocessing
Once
Through
Cycle
Waste Disposal

Figure 1

The CURE report of 1990 was not the first time that a concerted effort was put forward in the U.S. to analyze P-T as a high level waste management/disposal device. During the period from approximately 1976 to 1982 there were several major programs undertaken by the U.S. and by several European countries to develop P-T technology and make evaluations of its feasibility. The U.S. study (Ref. 2,3) performed its assessment based upon two reference scenarios. The first scenario represented the intent of the nuclear industry to reprocess all of its spent fuel for recycling. It included reprocessing facilities, plutonium fuel fabrication, and plutonium recycle and made no accommodations for P-T. The second reference scenario was essentially identical to the first but included facilities for advanced partitioning and transmutation. It was further assumed that a geological repository would be available, without difficulty, to accept the high level waste resulting from both scenarios. Based on these criteria a comparitive risk assessment was performed. This assessment showed that although partitioning and transmutation slightly reduced the long term radiological risks, the additional treatment of the spent fuel to partition and transmute would result in elevated short term risks, both radiological and non-radiological. The 1980 report therefore concluded that no incentives existed for the implementation of P-T technology into the existing nuclear fuel cycle.

The causes for the resurgence of inquiry into P-T should be apparent. There are large fundamental differences which exist between the criteria used for evaluation in 1980 and the criteria which must be used at the present to make the same evaluation. The first difference is that after the Carter administration the U.S. nuclear industry has not pursued reprocessing as part of the fuel cycle. Today's reference scenario must be a

once through fuel cycle. The second major difference has to due with the assumption that a repository would be available without delay or difficulty. Recent problems with the repository has caused some to doubt whether it can be considered as a given in the evaluation of P-T. In fact, in the more recent evaluations, one of the prime selling points of P-T is its potential for making the process of characterizing, licensing, and constructing a repository smoother and quicker. If this capability could be shown concisely then there would be a strong argument for a reconsideration of P-T.

2.1 SPENT FUEL RADIONUCLIDE INVENTORY

2.1.1 TRANSURANICS

For United States thermal neutron reactors, the initial fuel consists of uranium oxide (UO_2) pellets encased in zircaloy tubes, or cladding. The initial uranium isotopic composition of the of the pellets varies slightly between reactors from 96% to 97% U238 and 4% to 3% U235. Nuclear reactors generate power through the release of energy from a sustained fission chain reaction within the core. The U235 is the primary fissioning isotope. This isotope absorbs a neutron and forms the compound nucleus U236, which then fissions at a high probability. However it is also possible, though much less likely, that the compound nucleus will not fission but will instead emit a gamma ray and rermain as U236. U238 may fission but will most likely capture a neutron and form U239. U239 decays by beta emission to form Np239, which itself decays to Pu239. This chain causes a significant buildup of Pu239 as the reactor operates. Pu239 has a large fission cross section and contributes a large amount of energy to the core. The high neutron flux associated with the core of the reactor results

in multiple neutron capture reactions within the fuel and forms a heavy metal isotope chain. Figure 2 represents this chain and shows how the transuranic elements plutonium, neptunium, americium, and curium all form as the reactor operates.

The actual amounts of transuranic elements present in spent fuel is only a small fraction of the total heavy metal mass. Plutonium is the most prevalent transuranic, making up almost one percent of the total. The remaining transuranics neptunium, americium, and curium only constitute approximately one tenth of one percent of the total. Despite their small masses these transuranics represent a tremendous toxicity and long term radiological hazard. This means that by removing only a small fraction of the total spent fuel inventory a much larger fraction of the hazard would also be eliminated.

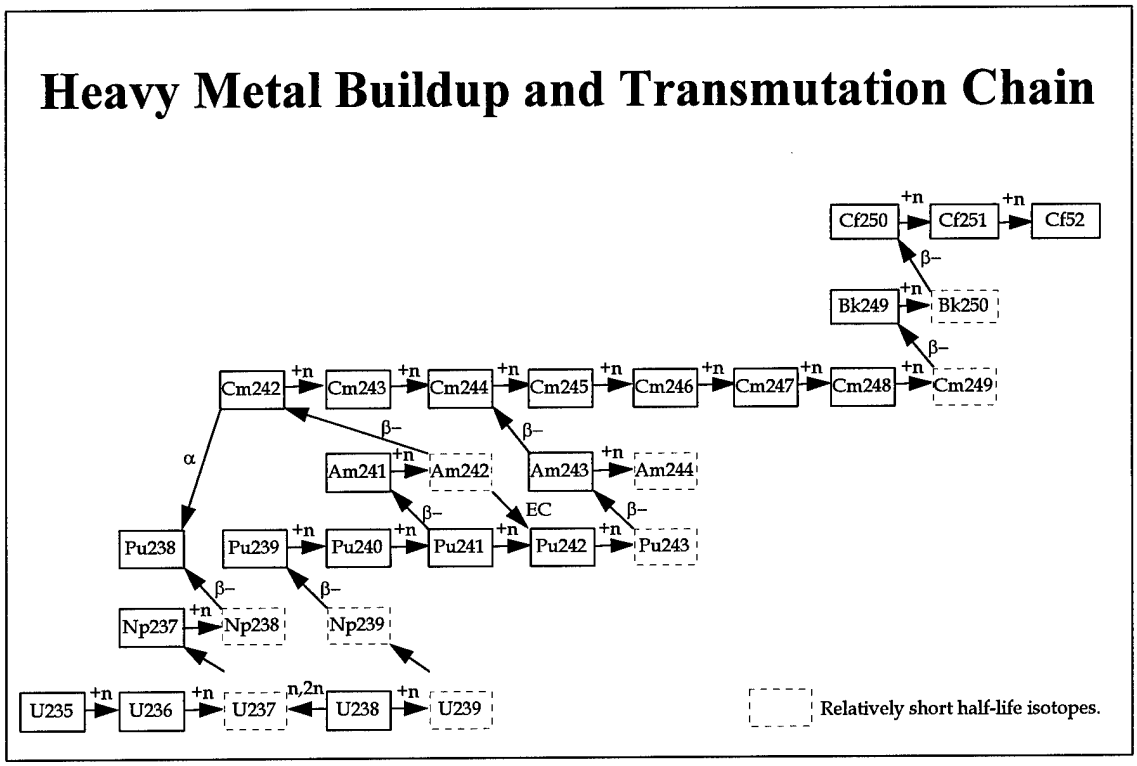


Figure 2 **S.E. Binney et al. Ref. 1.**

2.1.2 FISSION PRODUCTS

In addition to the transuranics which build up as a result of neutron capture within the fuel pellets many radioactive and stable fission products are also present. Because of the extremely dynamic nature of many of these isotopes the inventory of fission products depends very strongly on the amount of time the fuel was present within the reactor as well as the amount of time the spent fuel has been allowed to cool. The first parameter is referred to as the burnup and is expressed in megawatt days per metric ton of uranium (MWD/MTU). Typical burnup for nuclear fuel is approximately 33,000 MWD/MTU and due to the large amount of spent fuel which is presently in storage throughout the U.S., a decay time of ten years is a good assumption. Using these values, computer codes can be used to estimate the total radionuclide inventory as well as other values such as the activity and heat generation. Table I shows a listing of actinides (transuranics) and fission products per metric ton of spent PWR fuel, assuming the above stated parameters.

The total amount of fission products in spent fuel represents approximately 3.3 percent of the total mass. Yet in terms of short term radioactivity and heat load these fission products represent the majority of the hazard associated with the spent fuel. Therefore, between the fission products and the transuranics essentially the entire hazard of spent fuel is due to only four percent of the mass. This fact is what makes partitioning and transmutation so appealing.

Table I: (S.E. Binney et al. Ref 1): PWR Spent Fuel Radionuclide Inventory at 10 year decay and 33,000 MWd/Mt

Element/ Isotope	gm/MTU	Half-life (yr)	Watts/MTU	Curies/ MTU	Ingestion Toxicity $m^3 H_20$
Uranium	956,300	--	0.022	2.63	3.96E4
234U	12.6	2.45e5	2.26e-3	0.079	2.62e3
235U	7373	7.04e8	4.18e-4	0.016	5.32e2
236U	3803	2.34e7	6.67e3	0.246	8.21e3
238U	945100	4.47e9	8.06e-3	0.318	7.95e3
Neptunium	440	--	0.053	18.4	2.85e5
237Np	440	2.14e6	9.49e-3	0.310	1.04e5
Plutonium	8745	--	107	82420	4.71e8
238Pu	137	87.7	78	2.35e3	6.27e7
239Pu	5044	2.41e4	9.67	314	1.06e8
240Pu	2324	6.56e3	16.5	530	3.96e8
241Pu	769	14.4	2.46	7.9e4	3.60e5
242Pu	471	3.76e5	0.0532	1.8	3.6e5
Americium	594	--	58	1.76e3	4.38e8
241Am	503	432	57.3	1.73e3	4.32e8
242mAm	0.660	141	--	6.92	1.73e6
243Am	90.6	7.37e3	.581	18.1	4.52e6
Curium	18.87	--	50.9	1453	2.08e8
243Cm	0.337	28.5	0.637	17.4	3.47e6
244Cm	17.6	18.1	50	1.43e3	2.04e8
245Cm	0.932	8.5e3	5.33e-3	0.160	4.01e4
Total Actinides	9.66e5	--	216	8.57e4	1.68e9
Seleinium	55.4	--	--	1.0e-4	1.35e5
79Se	5.8	6.5e4	1.-e-4	--	1.35e5
Bromine	21.3	stable	--	7.1	--
Krypton	350	--	7.1	4.76e3	
85Kr	12.1	10.72	7.1	4.76e3	
Rubidium	345	--	--	2.1e[illegible]-5	--
Strontium	751	--	64.8	--	1.86e11
88Sr	341	stable	--	--	--
90Sr	409	29	64.8	5.59e4	1.86e11
Yttrium	445	--	310	55880	2.79e9
89Y	445	stable	--	--	--
$90Y_m$		short	310	5.59e4	2.79e9
Zirconium	3555	--	2.1e-4	1.77	2.22e3
93Zr	705	1.5e6	2.1e-4	1.77	2.22e3
Niobium	0.0045	--	1.3e-4	0.75	1.92e3
93mNb	0.0029	13.6	1.3e-4	0.75	1.92e3
Molybdium	3295	stable	--	--	--
Technetium	761	--	6.5e-3	12.9	6.45e4

Element/ Isotope	gm/MTU	Half-life (yr)	Watts/MTU	Curies/ MTU	Ingestion Toxicity m^3 H_2O
99Tc	761	2.13e5	6.5e-3	12.9	6.45e4
Ruthenium	2171	--	3.4e-2	569	5.69e7
106Ru	0.17	373d	3.4e-2	569	5.69e7
Rhodium	463	--	5.5	569	
106Rh		--	5.5	569	
Palladium	1396	--	6.8e-6	0.115	3.82e4
107Pd	223	6.5e6	6.8e-6	0.115	3.82e4
Silver	78		2.9e-3	0.177	5.84e3
110mAg		250d	2.9e-3	0.175	5.84e3
Cadmium	111	--	6.0e-2	35.5	
113mCd	0.154	13.7	6.0e-2	35.5	
Tin	90.7	--	1.3e-3	0.971	3.24e5
126Sn	27.6	1e5	9.8e-4	0.782	2.61e5
Antimony	19.7	--	3.7	1185	1.19e7
125Sb	1.15	2.76	3.7	1.18e3	1.18e7
126Sb	--	12.4d	2.0e-3	0.11	2.61e5
126mSb	--	19m	1.0e-2	0.78	3.65e4
Tellurium	482	--	2.4e-1	289	2.89e6
125mTe		58d	2.4e-1	289	2.89e6
Iodine	234	--	1.5e-5	0.0315	5.25e5
129I	178	1.6e7	1.5e-5	0.0315	5.25e5
Xenon	5293	stable	--	--	
Cesium	2339	--	1.44e2	86680	4.67e9
134Cs	4.06	2.07	54	5260	5.84e8
135Cs	287	3e6	1.1e-4	0.33	3.3e3
137Cs	936	30.17	90	81420	4.07e9
Barium	1710	--	3.02e2	77020	7.7e4
137mBa		2.5m	3.02e2	7.7e4	--
Lanthanum	1200	stable	--	--	--
Cerium	2324	--	9.9e-2	149	1.49e7
144Ce	0.047	284.4d	9.9e-2	149	1.49e7
Praseodymium	1101	--	1.1	151	1.51e2
144Pr		17.3m	1.1	149	
144mPr-		7.2m	6.1e-4	1.8	
Neodmymium	3974	Stable	--	--	--
Promethium	10	--	3.3	9308	4.65e7
147Pm	10	2.63	3.3	9.31e3	4.65e7
Samarium	785	--	3.5e-2	299	7.49e5
151Sm	11.4	90	3.5e-2	299	7.49e5
Europium	128	--	43	6279	2.42e8
154Eu	17.3	8.5	41.8	4.67e3	2.34e8
155Eu	3.45	4.73	1.2	1.6e3	8.0e6
Total Fission Products	3.36e4			2.99e5	1.94e11

Element/ Isotope	gm/MTU	Half-life (yr)	Watts/MTU	Curies/ MTU	Ingestion Toxicity m^3 H_2O
Activation Products					
14C	0.126	5730	1.7e-4	0.56	7.1e2
54Mn	1.1e-4	312d	4.4e-3	0.88	8.8e3
55Fe	0.51	2.73	4.1e-2	1210	1.5e6
60Co	2.5	5.27	44	2870	9.6e7
59Ni	63	7.5e4	2.0e-4	5.09	2.5e4
63Ni	11.4	100	2.6e-1	645	2.2e7
93Zr	56	1.53e6	1.7e-5	0.14	1.8e2
93mNb	2.1e-4	13.6	1.1e-5	0.059	1.5e2
94Nb	2.6	20000	5.0e-3	0.49	1.6e5
93Mo	0.020	3500	2.0e-3	0.022	7.2e3
119mSn	5.1e-5	293d	1.0e-4	0.19	6.5e4
121nSn	9.4e-3	55	1.0e-3	0.51	1.7e5
125Sb	0.12	2.73	4.0e-1	127	1.3e6
125mTe	1.7e-3	58d	2.6e-2	30.9	3.1e5
Total Activation Products	136	--	45	4891	1.2e8

2.2 Identifying Key Radionuclides

Simply knowing the radionuclide inventory is not enough. Each fraction's half life, initial inventory, and chemistry will dictate the extent to which it will effect the 10,000 year performance of the geological repository. Therefore, each radionuclide can be assigned a priority based on the relative hazard each presents. The first step in implementing partitioning and transmutation is to identify which radionuclides are to be partitioned followed by a determination of what treatment process is best suited for transmuting each fraction. In order to do this the spent fuel has to be examined from two perspectives, first in terms of near term hazard (present to several hundred years), and secondly from the long term hazard (up to 10,000 years). NRC requires that waste canisters placed in the repository provide containment for a three hundred year period. Therefore it is not expected that any significant release will occur during the short term. Short term considerations are mostly concerned with the thermal heat load placed on the repository by the waste. This heat load is a result of the radioactive decay of the waste, which can cause very high temperature elevations. Removal of the abundant short lived fission product radionuclides cesium-137 and strontium-90 will eliminate the greatest fraction of heat load while removing the actinides also will remove almost all the remaining heat load (see table 1).

The characterization of Yucca Mountain has enabled scientists to establish which radionuclides present in the spent fuel inventory pose the greatest risk. The NRC requires that release rates from the engineered barrier for individual radionuclides, following the required containment period of 300 years, shall not exceed one part in 100,000 per year of the inventory of each radionuclide calculated to be present within

the repository at 1,000 years (Ref 4.). After 1,000 years the short lived fractions, including cesium-137 and strontium-90, will have decayed away to leave only the long lived nuclides remaining. Table II shows the calculated inventory for PWR spent fuel at 1,000 years of decay. Part of the characterization process at Yucca Mountain has examined which of these radionuclides may exhibit dissolution rates exceeding the NRC regulations. These calculations assume completely failed containment and saturation in water. It has been concluded that the radionuclides iodine-129, cesium-135, technetium-99, and carbon-14 may exceed regulations for these assumed conditions. The actinides are generally considered to possess extremely low dissolution rates in the unsaturated, volcanic tuff environment of Yucca Mountain. However uncertainties arise if the assumption of a saturated, oxidizing environment are used in the determination. This uncertainty combined with other considerations of long term hazard such as the extreme ingestion toxicity of the actinides have led to the decision that the long term risk would be most effectively reduced by reprocessing spent fuel to remove Pu and U, followed by advanced partitioning of the minor actinides Am, Np, and Cm, the fission products Cs, Tc and I, and the activation product C. Reduction of short term heat loadings have led to the inclusion of strontium as well.

Table II(S.E. Binney et al. Ref 1): PWR spent fuel radionuclide inventory at 1,000 years.

Radionuclide	Half-Life Years	Ci/1000 MTHM	1,000 year Activity %	Cumulative % of total Activity
241Am	432	8.95e5	51.33	51.33
243Am	7.37e3	3.11e4	1.78	53.11
240Pu	6.56e3	4.77e5	27.37	80.48
239Pu	2.41e4	3.05e5	17.45	97.76
242Pu	3.76e5	1.76e3	0.1	98.07
238Pu	88	967	0.06	98.12
99Tc	2.13e5	1.3e4	0.75	98.87
59Ni	7.5e4	5.15e3	0.295	
63Ni	100	354	0.020	
93Zr	1.53e6	1.93e3	0.111	
93mNb	13.7	1.84e3	0.105	
94Nb	2.03e4	1.24e3	0.071	
14C	5.73e3	1.37e3	0.079	
234U	2.45e5	1.98e3	0.114	
238U	4.47e9	317	0.016	
236U	2.34e7	271	0.018	
237Np	2.14e6	1.00e3	0.057	
126Sn	1e5	772	0.044	
79Se	6.5e6	405	0.023	
135Cs	3e6	345	0.020	
151Sm	90	163	0.009	
107Pd	6.5e6	112	0.006	
129I	1.57e7	32	0.0018	

2.3 PARTITIONING REVIEW

Once determination has been made as to which elemental fraction must be removed from the spent fuel, the next step is to develop methods to accomplish this. For purposes of waste management, partitioning is defined as a treatment which is designed to separate key components of a waste stream and isolate them. These key components are generally elements possessing long lived isotopes and/or extreme toxicity and whose removal is deemed desirable in order to reduce the hazard associated with the remainder of the waste. An important factor to any partitioning process is that the partitioned wastes be isolated and contained in a form suitable for permanent disposal by other means. This should point out the fact that partitioning is not in itself a method of waste disposal, but rather is a method of waste management that may serve as a precursor to waste disposal. As was previously stated, this paper focuses primarily on transmutation methods and is not intended to be an in-depth discussion of the technologies of partitioning. However, it is necessary that a brief introduction be given regarding the state of the art with respect to partitioning.

Although the philosophies differ, partitioning is in many ways an extension of the concept of reprocessing. Reprocessing is a well defined technology in which spent fuel is treated in order to separate the useful amounts of uranium and plutonium and recycle them back into the fuel cycle. Because both concepts seek to treat spent fuel with the intention of removing specific elements the technology employed would be similar. This similarity has caused almost all P-T designs to envision partitioning as merely an extension of a reprocessing system. Although reprocessing is performed commercially in other countries such as France and England, there is at present no commercial reprocessing in the United States. This is due to the unfavorable economics of

reprocessing as compared to the cost of mined uranium, the inherent dangers of proliferation and diversion which exist in a reprocessing fuel cycle, and strong public opinion against reprocessing and nuclear power in general. The United States' experience with reprocessing is extensive, however, in the area of weapons production.

2.4 PARTITIONING TECHNOLOGY

The existing methods for recovery and recycle of actinides are referred to as pyroprocesses and aqueous processes. Aqueous processes are well established and are currently being utilized by all commercial reprocessing facilities worldwide. For this reason they are considered to be the most likely option for partitioning and transmutation. The aqueous processes work by first chopping the spent fuel and dissolving the fuel elements in a nitric acid solution. From this solution the actinides are selectively removed through the extensive use of chemical reagents. The process called PUREX was developed to extract plutonium and uranium whereas TRUEX was developed to extract the minor actinides neptunium, americium, and curium. Figure 3 represents the simplified flow diagram as presented in the CURE report (Ref. 1) for partitioning via a combined PUREX and TRUEX process. Aqueous processes have proven the ability to adequately partition actinides from the spent fuel waste stream. In addition to this there have been several studies done to develop means for further separation of strontium and cesium. (Ref. 5,6). As the figure shows, iodine and carbon can be rather easily separated from the off gases produced in the initial chopping and dissolution of the spent fuel.

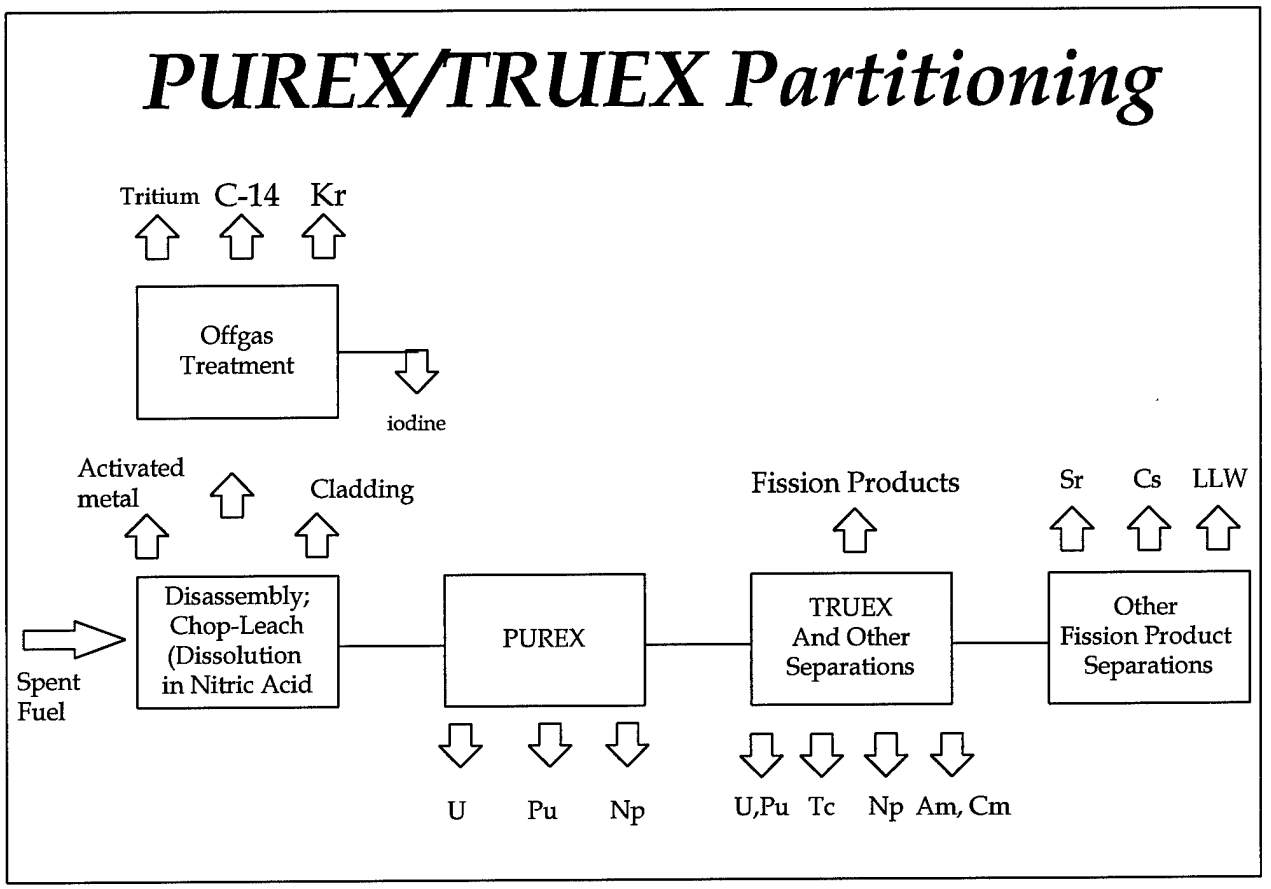


Figure 3 **S.E. Binney et al. (Ref. 1)**

Pyroprocessing is a younger, less developed technology first utilized at the Experimental Breeder Reactor EBR-2 (Ref. 7) for processing and fabricating a metal U-Pu fuel. It is currently being developed further for use in ANL/GE's Integral Fast Reactor Program (Ref. 8). This processing method has advantages due to its comparative simplicity over aqueous methods and its potential to reduce diversion and proliferation risks by avoiding highly purified fractions of partitioned elements such as plutonium. At this point pyroprocessing techniques can not provide the same level of decontamination as aqueous methods can. This may be a disadvantage if pyroprocesses are not be able to meet high decontamination factors necessary to reduce the waste stream from high level to low level. Pyrochemical processing functions by using an electrorefiner which would sequentially deposit onto anodes the bulk of the uranium, plutonium, and minor transuranics. These elements flow through a molten salt in their metallic states so prior to processing spent fuel would have to be treated in order to reduce the oxides to metal form. This process has been proposed as part of the Liquid Metal Actinide Burning concept, which will be discussed in further detail later.

2.5 TRANSMUTATION REVIEW

Transmutation is the second component of the P-T concept. For purposes of this discussion, transmutation is defined as any process which is designed to treat partitioned fractions of spent fuel in order to transmute them into stable, or short-lived nuclides. The efficiency and speed with which a process can meet this goal are vital parameters for choosing the best transmutation system.

Under certain physical and probabilistic conditions, a nuclide can undergo a nuclear reaction and change its elemental properties. When this occurs, it is referred to

as a transmutation. The conditions which induce transmutation vary widely between individual nuclides. For example, radioactive nuclides will transmute spontaneously by emitting a particle or particles from their nucleus. The event itself is a probabilistic one and is characteristic of the unstable nucleus itself. Transmutation does not have to be a spontaneous process. Rather, if the target nuclide is well understood, means can be developed with which to induce a transmutation to occur. This notion of inducing transmutation is the basis for the whole concept of P-T.

There are a wide range of nuclear reactions which cause a target nuclide to transmute. There are also a wide variety of particles available today with which to bombard target nuclei. However, for purposes of transmutation in the United States, only neutron induced transmutations are presently being considered as reasonable options. The reasons for this decision are rather simple. First of all, due to defense and nuclear power related research, there exists in the literature a tremendous amount of information regarding neutron physics. This data describes neutron capture and fission cross sections of many different nuclides under varying neutron energies. In order to obtain an equal amount of such information on other types of particles a tremendous amount of additional research would be required. Due to the large amount of experience and information already available using neutrons and the rush to develop a P-T concept which does not involve radically new technology, it has been decided that neutron based concepts offer the best possibility of success. Another strong reason is that neutron based concepts for P-T can be easily introduced into existing reactors or into designs for advanced reactors. Some view this as advantageous because it might signify the resurgence in nuclear power as reactors become more capable of handling their own waste.

It has been established that in order to minimize the long term risk and short term heat loading on a repository partitioning could be done on the spent fuel to remove actinides Pu, Am, Np, and Cm, plus iodine, cesium, technetium, strontium, and carbon. The next step is to analyze each fraction to determine the best route of transmutation. If transmutation by neutron flux is not practical then alternate methods must be decided upon.

Slightly more then ninety five percent of the heavy metal weight in spent fuel is uranium. This enormous fraction is approximately 98% U238, while about 0.8% is U235, 0.4% is U236, and the remainder is U234. The U235 is at a slightly higher isotopic concentration then in natural uranium and could be separated from the rest of the fraction and sent back to the fuel cycle. However, U236 would act as a strong neutron poison and would have to be separated for storage or disposal. Compared to the large volume available only about one percent of the U238 would be sent back to the fuel cycle as breeding material. The remainder would have to be stored or disposed of as a low or intermediate level waste, very similar in composition and radiological risk to mill tailings.

Approximately 1% of the heavy metal weight in spent fuel is plutonium. Of this 1%, two thirds of the plutonium is fissile Pu-239. This material is a valuable fuel and can be reintroduced to the fuel cycle of existing LWRs and transmuted by fission rather easily. The fraction of plutonium as a whole could be transmuted via thermal or epithermal neutron capture or fission to short lived and/or stable nuclides. Plutonium is also fissionable in a hard neutron spectrum and so can also be used as fuel in liquid metal fast reactor systems.

The minor actinides Np, Am, and Cm comprise about 0.1 % of PWR spent fuel and are not fissile in the soft neutron spectrum of a LWR. Although the minor actinides can fission in a soft spectrum the probability is far outweighed by the probability of neutron absorption. Therefore, transmutation would not be effective for the minor actinides in an LWR. However it is conceivable that they might be transmuted via thermal neutrons to some higher actinide which might be considered more useful. If this were attempted the minor actinides would act as strong poisons and the problems this would present in terms of reactivity and control make transmutation of minor actinides in a thermal neutron flux difficult. However all of the minor actinides are predominantly fissionable when placed in a hard neutron spectrum. Therefore the most likely transmutation option for the minor actinides would be in a liquid metal fast reactor or an accelerator system employing fast neutrons.

For purposes of this discussion, radioiodine consists of I-129 and I-127, and the technetium fraction is entirely Tc-99. These two fractions can be transmuted by thermal neutron absorption without any need to separate individual isotopes. Below are the neutron absorption chains for both isotopes of iodine as well as technetium 99. It can be seen that all neutron capture products are short lived radioisotopes that decay to stable isotopes.

Iodine Transmutation Chain:

129I + n -->	130I -->	130Xe +n -->	131Xe + n -->	132Xe
1.6e7 yr	12.4 hr	(stable)	stable	stable

127I + n -->	128I -->	128Xe + n -->	129Xe + n -->	130Xe
stable	25 min	stable	stable	stable

Technetium Transmutation Chain

99Tc + n -->	100Tc -->	100Ru + n -->	101Ru + n -->	102Ru
2.1e5 yr	16 s	stable	stable	stable

The absorption cross sections for I-129 and Tc-99 are highest for neutrons in the thermal to epithermal range. Therefore it should be possible to transmute these fractions using leaking neutrons from an LWR, or thermalized neutrons from a FMR or an accelerator system.

Cesium-137 and Strontium-90 are much more difficult to deal with then iodine and technetium. As table one shows the strontium fraction consists of both Sr-88 and Sr-90 and the cesium fraction contains the isotopes Cs-133(stable), Cs-134, Cs-135, and Cs-137. Below are the transmutation and decay chains for both cesium and strontium fractions.

Strontium Isotope Transmutation Chain:

90Sr + n --> 91Sr --> Y91 + n --> Y92 --> Zr92
29 yr 9.5 hr 59 d 3.5 hr stable

91Y --> 91Zr + n --> 92Zr
stable stable

88Sr + n --> 89Sr + n --> 90Sr...
stable 50.5 d

89Sr --> 89Y + n --> 90Y --> 90Zr + n --> 91Zr
stable 64 hr stable stable

Cesium Isotope Transmutation Chain:

137Cs+n -> 138Cs -> 138Ba+n -> 139Ba -> 139La+n -> . ... 140La -> 140Ce
30 yr 32 m stable 83 m stable 40 hr stable...

137Cs -> 137Ba
stable

135Cs + n -> 136Cs + n -> 137Cs...
3e6 yr 13 d 30 yr

136Cs -> 136Ba + n -> 137Ba + n -> 138Ba
stable stable stable

133Cs + n -> 134Cs + n -> 135Cs
stable 2.1 yr 3e6 yr

134Cs -> 134 Ba + n -> 135Ba + n -> 136Ba
2.1 yr stable stable stable

Unlike the iodine and technetium fractions it is not possible to effectively transmute the cesium and strontium fractions with neutron absorption. This is because even as the key isotopes Cs-137 and Sr-90 are transmuted, neutron absorption by the other isotopes present cause a buildup of those same isotopes. The conclusion is that the cesium and strontium fractions cannot be transmuted and should instead be stored after being partitioned from the spent fuel. Since these isotopes are relatively short lived the containment might only need be several decades, after which the heat load will have reduced and the fractions can be placed into a repository for permanent disposal. It should be noted that both cesium-137 and strontium-90 have applications in research and industry and it has been suggested that the partitioned fractions could perhaps be economically separated and used for these purposes instead of being disposed of.

None of the proposals which will be discussed mention intentions to transmute the carbon-14 fraction. Assuming that the entire inventory of spent LWR fuel in the United States at the year 2011 is 63,000 tons of heavy metal and that C-14 represents 0.126gm/MTU (see table 1) this corresponds to a total C-14 inventory of 7.9 kilograms. Although it is deemed necessary to partition C-14 from the waste stream in order to prevent it from exceeding NRC's leaching requirement, the amount of material in question is so small that it is not worthwhile to transmute this fraction. Instead the C-14

can be specially contained in a unique, single container and then stored or placed to the repository separately.

3.0 RECENT TRANSMUTATION CONCEPTS PROPOSED IN THE U.S.

As a result of the resurgence of interest in partitioning and transmutation in the U.S. since the late 1980's there has been three important concepts brought forth. These concepts can be categorized as either accelerator based concepts or liquid metal reactor based concepts.

3.1 ACCELERATOR BASED CONCEPTS

3.1.1 THE PHOENIX CONCEPT (Ref. 9): BROOKHAVEN NATIONAL LABORATORY

The Phoenix concept proposed by Brookhaven National Lab offers a means by which to transmute the key minor actinides neptunium, americium, and curium as well as the long lived fission products iodine-129 and technetium-99. Phoenix would employ a hybrid proton accelerator which induces fission in a subcritical lattice. It is calculated that one such accelerator device running at 3,600 MWth would transmute the Np, Am, Cm, and most of the iodine produced by about 75 light water reactors while at the same time produce 850 MWe. Phoenix offers a hard neutron spectrum ideal for transmuting the minor actinides and holds the advantage of operating at a subcritical level. This is made possible because the accelerator can provide the additional neutrons necessary to maintain the fission reaction. The ability to operate at a subcritical level gives the Phoenix a safety feature which cannot be offered by advanced liquid metal reactors.

A conceptual schematic of the Phoenix is presented in Figure 4. The major component of the system is a large linear proton accelerator which produces 1.6 GeV protons at a maximum current of 104 mA. Such an accelerator is larger then any currently in operation. However in a recent study (Ref. 10) an accelerator was designed to produce 204 mA and 1.6 GeV as part of a system to produce tritium. This design was evaluated by DOE and determined to be technically feasible. Therefore the Phoenix accelerator is envisioned by Brookhaven as simply a scaled down version of the tritium producing accelerator and should not present any insurmountable technical difficulties.

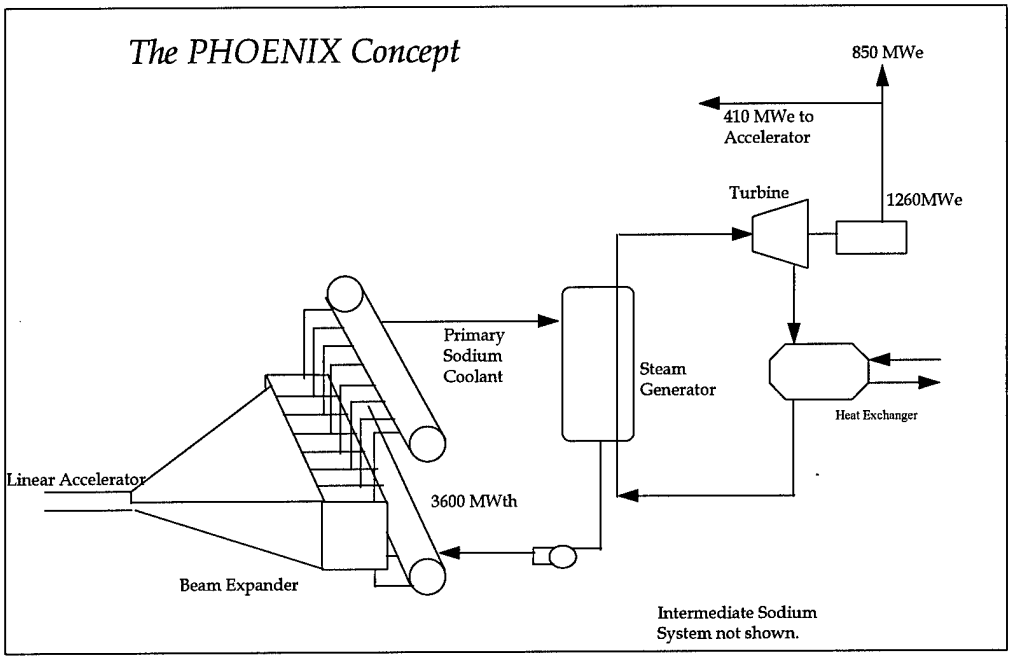


Figure 4.A G.J. Van Tuyle et al. (Ref. 9))

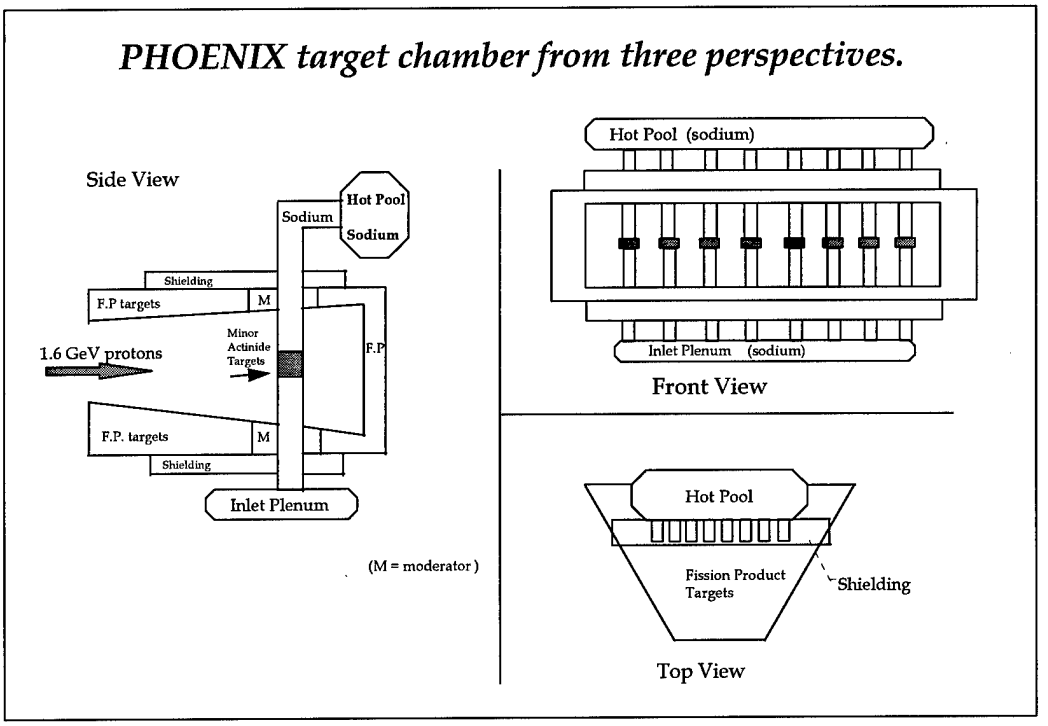


Figure 4.B G.J. Van Tuyle et al. (Ref. 9)

Spallation and Evaporation

The accelerator provides the additional neutrons necessary to initiate the fission reaction within the lattices and to maintain the reaction despite its subcriticality. It does this by means of a process known as spallation and evaporation. As a very high energy proton encounters the minor actinide matrix it will interact with several nuclei, in sequence, causing their destruction by spallation or evaporation. Spallation is essentially a form of very high energy fission. In spallation the interaction raises the target nucleus to an extremely excited state which rapidly fissions, releasing energy and several neutrons. Evaporation is slightly different then spallation in that the excited nucleus does not fission. Instead it rapidly spits off a series of particles, including neutrons, protons, deuterons, tritons, alphas, and helium-3. Figure 5 illustrates this interaction process.

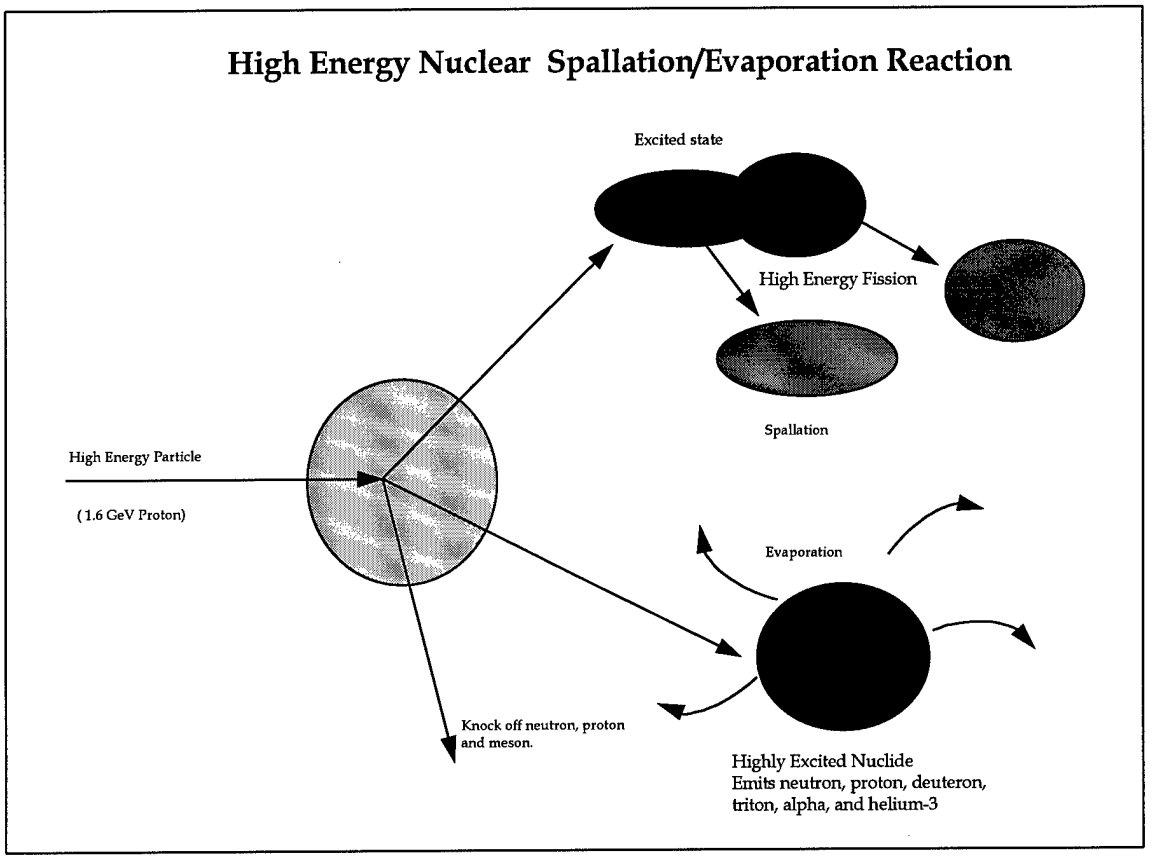


Figure 5 G.J. Van Tuyle et al. (Ref. 9)

The interactions of highly energetic particles with target nuclei are fairly well understood. Using empirical data and formulae it is possible to predict the number of neutrons released per proton due to spallation and evaporation within the target lattice. It is estimated that each proton will cause the destruction of 5-6 nuclei and cause the release of 50 neutrons. These neutrons provide the starting point for neutron multiplication within the subcritical lattice. The term subcritical means the effective neutron multiplication factor k_{eff} is less then one. In other words the fission reaction will not be self sustaining as long as $k_{eff} < 1$. However,if k_{eff} is known then the total number of neutrons produced per proton, via spallation and fission, can be calculated and the total number of transmutations per proton can be approximated by:

$$T = N * [\, k_{eff}/(1-k_{eff}) \,] / v + Tp$$

where

N = number of neutrons released as the proton penetrates into the lattice.
k_{eff} = effective neutron multiplier for the subcritical lattice.
v = average number of neutrons released per fission.
Tp = number of transmutations caused directly by the proton.

In a subcritical lattice of $k_{eff} = 0.9$ each proton will cause the transmutation of 172 target nuclides. If $k_{eff} = 0.95$ there will be 357 transmutations per proton. For $k_{eff} = 0.9$, if we estimate that 180 Mev are released by per transmutation, then approximately 31 GeV are released per 1.6 Gev proton. This approximation demonstrates how an accelerator can produce impressive amounts of power from a subcritical lattice.

The minor actinides will fission most effectively in a very hard neutron spectrum. It was necessary to design the Phoenix to produce as little neutron moderation as possible. This is most effectively done by employing a liquid metal core. Because of the rather large amount of experience in nuclear engineering with liquid

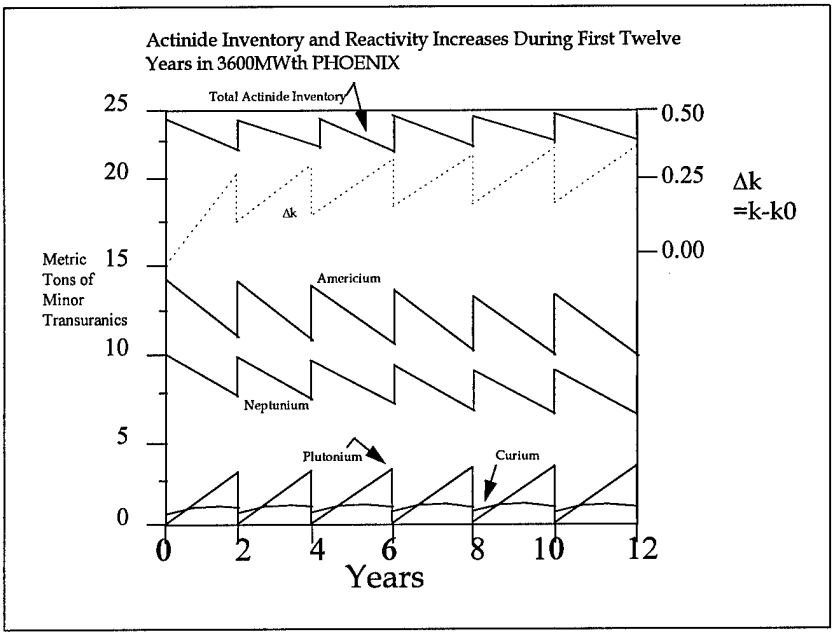
sodium cooled fast reactors it was decided that sodium would be the best choice for coolant. Other liquid metals such as a lead based coolant could show significant advantages by further hardening the neutron spectrum. However a lead based coolant requires a great deal of additional development. In order to make the Phoenix immediately feasible the sodium coolant is preferred.

The fuel lattices for the Phoenix are modeled directly after the fuel lattices of the Fast Flux Test Facility (FFTF) (Ref. 11). The FFTF is a liquid sodium cooled fast neutron test reactor and its modular fuel lattices are tested and well understood. Essentially the Phoenix lattice would simply substitute the uranium and plutonium of the FFTF lattice with minor actinides. The subcritical lattice is composed of up to eight individual target modules. Each module produces 450 MWth and contains 124 assemblies of 217 pins.

Minor Actinide Burnup

For the preliminary design presented, Brookhaven performed calculations to assess the rate of actinide removal, assuming a 75% power factor, and their results are shown in Figure 6. It was assumed that the entire lattice was initially loaded with minor actinides partitioned from light water reactor spent fuel. Although not necessary, it was decided that Phoenix would operate on a two year burnup period for fuel. Furthermore, a two year cool and decay period was to be allowed for the spent fuel. As the results indicate, the minor actinides are transmuted at a rate of about 2.6 metric tons per year. Americium and neptunium are the most significantly reduced, whereas curium, the smallest fraction, actually increases by a very small amount during each cycle due to the heavy metal buildup chain. This excess curium would have to be partitioned out during reprocessing and stored or disposed of. Plutonium is created in

the Phoenix system at a rate of approximately 1.55 metric tons, or 6.35% of minor actinide initial inventory, per year. The plutonium created is predominantly Pu-238 (85-87%), with the remaining fraction composed of Pu-239 and Pu-242. This creation is due to the heavy metal buildup chain, as the minor actinides absorb neutrons.



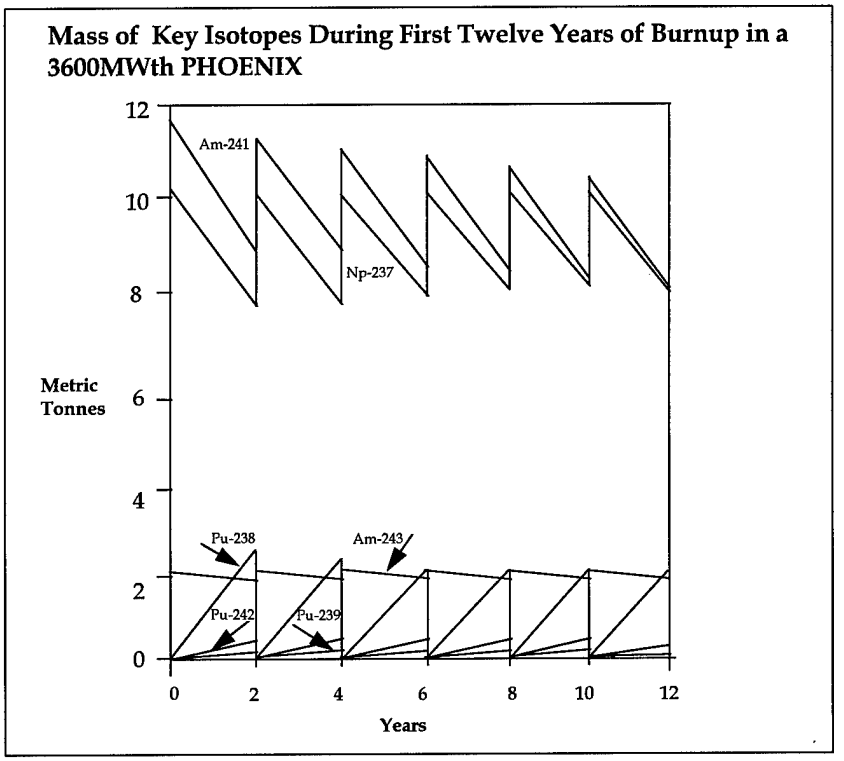


Figure 6 G.J. Van Tuyle et al. (Ref. 9)

Iodine Burnup

Figure 4 shows that the Phoenix is designed to accommodate fission products as well as minor actinides. This is possible because of the necessity of keeping the lattice subcritical. To accomplish this, the modular core is designed to allow a large fraction of neutrons to leak from the lattice system. These neutrons can be moderated and used to transmute fission product targets. In the Phoenix proposal only iodine was considered for transmutation because it was assumed that technetium could be easily transmuted via existing LMRs; however the accommodation of Technetium-99 could be made if necessary. Preliminary estimates state that the Phoenix can transmute 300 kg/year of iodine. At this rate a single PHOENIX could transmute all of the iodine produced annually by approximately 75 light water reactors. This estimate assumes that a reliable target system can be developed for iodine that will contain both volatile iodine and its gaseous transmutation product xenon. A report by Brager, Blackburn, and Wootan describes progress made by Westinghouse-Hanford in this area (Ref. 12).

Reactivity Swings

One important aspect of the Phoenix system can be seen on Figure 6 regarding the reactivity of the system over its two year operational period. Although the system starts out subcritical, as fissile material builds up the effective neutron multiplier increases. The predictions which Brookhaven calculated are only approximations since they do not account for geometrical variations. However, accommodations must be made in order to maintain a condition of subcriticality despite the tendency for k_{eff} to increase. Control can be achieved by changing the Phoenix geometry between cycles to

allow greater neutron escape or by adding poisons to fresh modules. A great deal of control can be maintained over the power output of the assemblies by varying the beam current as long as the system remains subcritical.

CONCLUSIONS

A full scale engineering feasibility study has not been performed regarding the Phoenix project. However, the fundamental technology called for is regarded as feasible. The accelerator is based on a scaled down version of an existing, approved design. The lattice system is modeled after the lattice core of the Fast Flux Test Facility, a sodium cooled fast reactor which operates at 400 MWth. It is assumed that fuel damage will be similar between the Phoenix and the FFTF. The fact that the Phoenix lattice will sustain additional damage due to high energy spallation will require additional research. However, since each proton produces only about 5 spallations as compared to 180 fissions (at $k_{eff} = 0.9$), the assumption that spallation damage is negligible is likely to be a good one. Brookhaven asserts that with additional research the modular design, fuel form, and coolant type can all be optimized to further improve Phoenix performance.

The primary advantage of Phoenix is its ability to run at a subcritical level, which improves its inherent safety potential. Also, due to its modular design, the Phoenix is capable of running at reduced loadings while still producing usable energy. This can be done by running the accelerator at a reduced duty factor. Although the accelerator concept may appear at first to be more complex technologically than typical reactor systems, in fact the technologies are not very far removed. One disadvantage is that the estimated cost of the Phoenix is about 1.5 times the estimated cost of a liquid

metal cooled reactor of equal rating. However, the Phoenix has a higher ability to transmute minor actinides and iodine then would an LMR of equal power output.

3.1.2 ACCELERATOR TRANSMUTATION OF NUCLEAR WASTE (ATW) (Ref. 13) LOS ALAMOS NATIONAL LABORATORY

Introduction

The second major U.S. program which would utilize an accelerator for transmutation has been proposed by LANL. Similar to Phoenix, this concept employs high current, medium energy protons to produce a large number of neutrons which are then used to transmute various radionuclides. However apart from the basic similarities between the two concepts the ATW is in fact quite different in many respects.

Figure 7 is a conceptual schematic of the ATW system. The accelerator provides 1.6 GeV protons at a current of 50 mA. The beam is directed into a target which is a cylinder of molten lead and bismuth. Through the spallation interaction described previously each proton produces roughly 55 high energy neutrons. At the outer surface of a lead-bismuth target 50 cm in diameter and 150 cm in height a maximum neutron flux of 10^{16} cm^{-2} sec^{-1} will be produced. Even in the outside regions of the moderator fluxes are only expected to drop by a factor of 3 or 4 times.

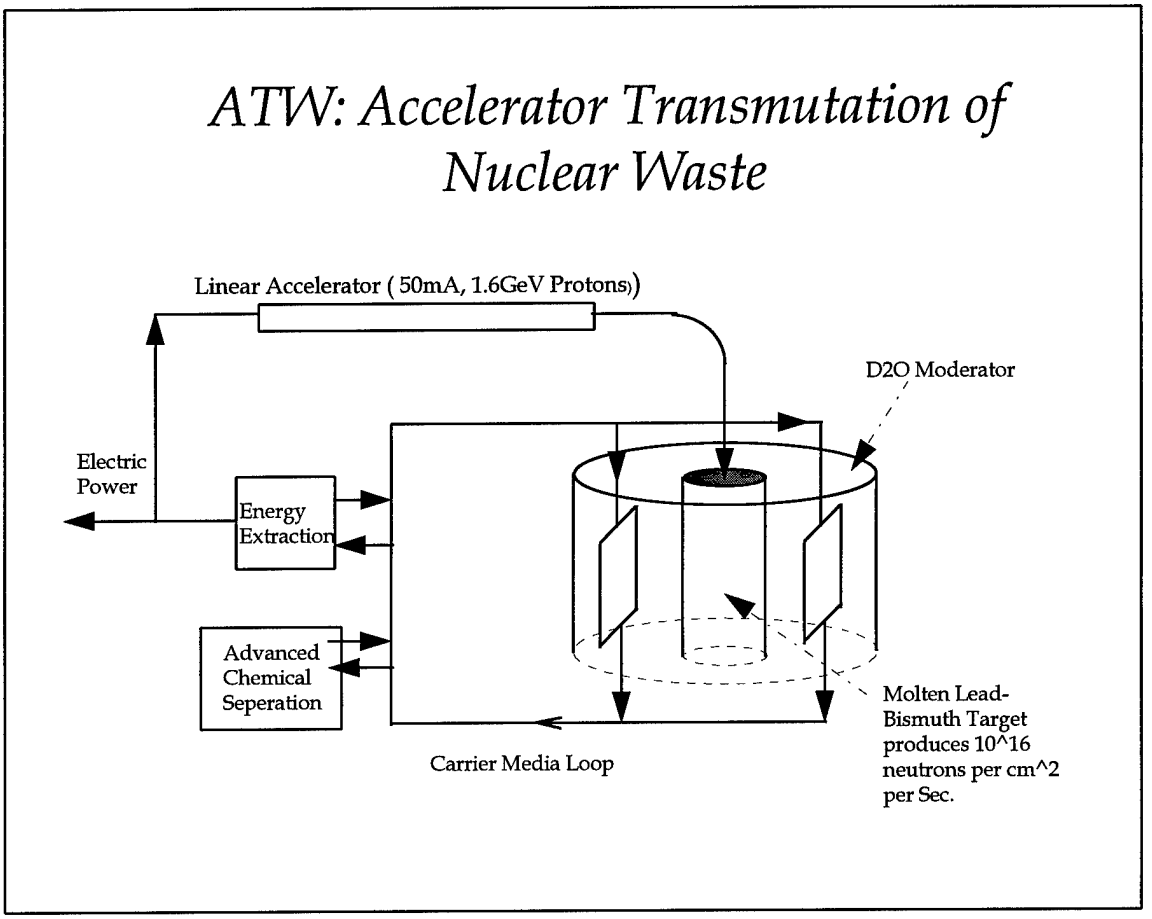


Figure 7 **E.D. Arthur (Ref. 13)**

The intense flux of neutrons would be thermalized by a deuterium oxide (D_20) blanket which surrounds the lead-bismuth target. It has already been established that the higher actinides fission much more efficiently under a hard neutron spectrum and are more likely to undergo neutron capture at thermal energies. For this reason previous studies have rejected the use of thermal neutrons for transmuting the actinides. However, LANL asserts that due to the extremely high intensity flux it is indeed possible to efficiently fission the higher actinides such as neptunium-237. Apparently, for a system of high flux thermal neutrons, the effective fission cross section is much greater than it would be at low flux. LANL describes a two step fission process in which two neutrons are required to induce fission, which itself produces 2.7 neutrons. Therefore the fission process for high flux thermal neutrons produces a net gain of neutrons and the effective fission cross sections become as much as 100 times greater then thermal fission cross sections at lower fluxes.

The fact that LANL's ATW can achieve both higher flux and higher cross sections leads to another important advantage. The rate of transmutation is described by the following relationship:

$$R = \Phi * \sigma * N$$

where
R = transmutation rate (transmutation/second)
N = number of target nuclides.
Φ = neutron flux (n/cm^2/s)
σ = cross section (cm^2)

It has been estimated that the high flux and fission cross section which the ATW can achieve results in an transmutation rate almost 1,000 times greater then rates achievable by other systems at a given inventory. Therefore, in order to achieve the

same rate of transmutation between the two systems the ATW inventory N can be reduced by the same factor of 1,000. In other words the ATW can achieve the same transmutation rate with an inventory of 100kg that might require an inventory of 10,000kg or more in a normal system employing normal fluxes and fast neutrons.

Carrier Media

The ATW system would require only a very small inventory to achieve a high rate of burnup. LANL has proposed that a continuous material flow system such as illustrated in Figure 7 would be the most appropriate for these conditions. The choice for carrier media has not been specified but possibilities include aqueous streams, slurries, and molten salts. Chemical processing and removal of fission and activation products would probably be easiest if an aqueous stream was used. This is due to the large, existing amount of experience in aqueous processing and the advanced state of the technology compared to processing slurries or molten salts. On the other hand molten salts hold a great deal of promise because their very high temperatures would allow highly efficient heat removal and conversion to electricity.

Transmutation Rates

The ATW would contain an inventory of about 100kg for the described 50mA system. Therefore the actual mass of actinides and fission products which would be transmuted per year is also rather small, approximately 70kg of Tc-99 and 10kg of actinides. Rates for iodine-129 have not been established. This is due to the problem of developing proper containment of the volatile iodine and its gaseous transmutation product xenon within a high temperature liquid carrier media.

Because of its small capacity the ATW would not be suitable for the transmutation of the relatively large volume of light water reactor wastes. Instead the ATW is perceived as an ideal system for the transmutation of actinides and technetium from DOE production facilities defense wastes. At the rates stated above one ATW system could transmute all of the actinides (mostly Np-137) and all of the technetium in the high level wastes at Hanford, Washington, in thirty years of operation.

In terms of feasibility the technology required to make the ATW a reality is reasonable. Similar to the PHOENIX, the accelerator is advanced but by simply downscaling the existing approved design for a 250mA, 1.6GeV accelerator there should be no major difficulties. Primary challenges would lie in the chemical processing systems associated with the ATW. Certain improvements may need to be made on these systems to assure adequate removal of fission and transmutation products from the carrier stream. These improvements will not require any major new inventions in processing however, and should be possible.

3.2 REACTOR BASED CONCEPTS:

3.2.1 ADVANCED LIQUID METAL RECTORS (ALMR) ARGONNE NATIONAL LABORATORY/GENERAL ELECTRIC

Recent reactor based proposals which have been studied most actively in the U.S. focus on the utilization of the liquid metal reactor for transmutation. LMRs provide the fast neutron spectrum which is most suited for the minor actinides and studies have been done to include fission products for transmutation as well. The technology of liquid metal reactors is well established throughout the world. Although the U.S. does not currently operate any commercial LMRs it was the first to begin development of this

reactor type and it was the U.S. Experimental Breeder Reactor EBR-1 that produced the world's first nuclear electric power in 1952. Currently the U.S. possesses two LMRs for research and development purposes, although funding for these research reactors is currently in danger due to large defense budget cuts. These are the Fast Flux Test Reactor (FFTF) in Hanford Washington, and the Experimental Breeder Reactor EBR-2 operated by Argonne in Idaho. In addition to these existing reactors there have been various designs proposed for advanced liquid metal reactors which are designed to provide increased simplicity, reliability, and enhanced passive safety characteristics.

The promise of increased safety combined with the potential for energy self sufficiency through breeding has kept the interest of the U.S. as well as many other countries. Although recently halted by the Clinton administration, the Department of Energy had been actively funding a national ALMR program. This program sought to develop General Electric's designs for a commercially viable integral fast reactor (IFR) system. Argonne National Laboratory was also involved in the development of the necessary supporting technology. This development mostly involved improving pyroprocessing methods for processing the ALMR metal fuel.

The ALMR fuel cycle is by definition closed loop since its startup fuel is procured from the reprocessing of LWR spent fuel. In addition to that, the ALMR must then reprocess its own spent fuel to attain self sufficiency. The design concept for the Integral Fast Reactor involves utilizing nine small reactor modules arranged in three power blocks of 465 MWe each, for a total of 1395MWth. Each reactor core is cooled with liquid sodium and burns fuel in a metal form, as opposed to the oxide fuels used in today's U.S. LWRs and FFTF. The metal fuel is an alloy of uranium, plutonium, and fission products (fissium) which is clad in stainless steel. EBR-2 was the first to use a

metal fuel in the 1960's and several advantages were noted, the most noteworthy being that the metal fuel form reduced moderation of neutrons and hardened the spectrum, causing the burning efficiency to rise. Metal fuels also have the potential to reduce proliferation risks due to the pyroprocessing methods used in fabrication which avoid highly purified plutonium or uranium fractions.

The original intent of the ALMR was to operate as a breeder reactor. Spent fuel from LWRs was to be reprocessed and the actinides fabricated into metal fuels. In the breeding mode the actinides from LWR spent fuel would only be necessary as startup fuel. Assemblies would be fabricated for the initial ALMR core and two reload cores. After startup the ALMR would operate at a breeding ratio greater then one. This would allow the ALMR to produce all the additional fuel it would require for its operating life. What actinides remained in the cores at the end of reactor life could then be refabricated to fuel and used as startup material for a new generation of ALMRs. This self sufficiency is the major reason why many countries continue to develop LMR technology despite the present high cost of reprocessing and the low cost of mining uranium in a once through fuel cycle.

It was recognized early on that the ALMR did not need to supply its own fuel by operating as a breeder. Instead, by operating at a breeding ratio of less than one, the ALMR would be called a "Burner" reactor (Ref. 14). This implies that the reactor consumes more fuel than it can produce. It was proposed that if ALMRs were utilized as burners then they could be used to consume the actinide inventory of today's LWRs. Although not the original intent of the IFR-ALMR program, studies have concluded that it would indeed be possible to operate at a breeding ratio as low as 0.75 while still maintaining the inherent safety characteristics which make the ALMR attractive. This

breeding ratio could be established by removing blanket assemblies and using a homogeneous core. A burner ALMR would transmute its core actinides at a rate of about two to three percent per year. Studies have also determined that although the metal fuel was originally designed to contain only Pu and U it is possible to substitute the higher actinides in the metal matrix as well. It was realized that the ALMR could function effectively as a transmutation device while also providing a new source of nuclear power.

Minor Actinide Transmutation

Several studies have been performed in order to estimate the rate of transmutation of transuranics by ALMRs. The use of computer codes such as 1DX and CITATION (Ref. 15, 16) allows calculations for both metal and oxide fueled LMRs. Results are similar for both cases but the metal fueled LMRs produce slightly higher transmutation rates due to a harder neutron spectrum.

Studies by Choi and Pigford (Ref. 17, 18) included calculations of burnup assuming minor actinides were placed with uranium in separate oxide targets in the core of an LMR. The remaining core space was filled with normal Pu/U oxide assemblies. Results showed that minor actinides neptunium and americium would be reduced by approximately two orders of magnitude over the lifetime of the reactor. The curium fraction as a whole was decreased by about a factor of ten. However Cm-246, Cm-247, and Cm-248 inventories were all increased due to chain absorption. The average transmutation rate for the minor actinides was calculated to be twelve percent per full power year, or eight percent per year at a power factor of seventy five percent. At this rate the total minor actinides inventory would be reduced by a factor of about

120 over the estimated thirty-seven year life of the reactor. Another study by Inoue, Sakata, et al. (Ref. 19) performed similar calculations with the assumption that the minor actinides were simply incorporated into the fuel of an LMR. This study concluded that transmutation rates would be eleven to twelve percent per year for oxide fueled LMRs and fourteen to fifteen percent for metal fueled LMRs. If metal fuels were fabricated to contain fifteen percent by weight of minor actinides, then a 1000MWe LMR with a 47 ton inventory would transmute approximately one ton per year.

Another paper by Choi (Ref. 20) examines the total transuranics transmutation potential if LMRs were brought on line according to the ALMR proposed schedule. This schedule planned for the 63,000 metric tons of heavy metal (MTHM) of LWR spent fuel to be reprocessed and the recovered transuranics fabricated to start up fuel for a series of ALMRs operating as burner reactors at a power capacity of 1395 MWe. Operation as burners would necessitate makeup fuel after each cycle throughout the lives of the ALMRs. It has been determined that the lowest breeding ratio possible while still maintaining the inherent safety characteristics of the ALMR is approximately 0.75. At this ratio the total spent fuel inventory of all US LWRs contained enough transuranics to fuel sixteen ALMRs throughout their 40 year operating lifetimes. The proposed schedule called for the introduction of the first ALMR in the year 2013, with the addition of one per year until the year 2028. If this was done then the total inventory of the LWR spent fuel would be reprocessed and utilized by the year 2068.

The total amount of spent fuel projected to exist in the U.S. in the year 2011 is 63,000 MTHM. Within this amount is contained approximately 612 MTHM of transuranics. Sixteen ALMRs would require 441 MTHM of transuranics as startup fuel while the remaining 171 MT would be required as make up fuel. After the 40 year

lifetime of all reactors there would still be 441 MTHM remaining in the cores while 170 MTHM would have been transmuted. This analysis shows that the total transuranic reduction factor achieved by implementing the Actinide Burning concept is 1.4, calculated by the ratio of the 612 tons available at the beginning and the 441 tons remaining at the end. In addition to this the remaining transuranics could be used as startup fuel for the next generation of ALMRs. This concept would make the ALMRs something of a cross between a monitored retrievable storage facility (MRSF) and a partitioning and transmutation device. As long as ALMRs continued to be deployed there would be no need to send any spent fuel transuranics to a repository because it can all be stored in ALMR cores or refabricated as startup fuel for new ALMRs.

Fission Product Transmutation

The concept of actinide burning using ALMRs does not extensively examine the transmutation of the long lived fission products iodine 129 or technetium 99. These fractions are transmuted more effectively in a thermal neutron flux which is not automatically provided by in the LMR core. However specific design modifications could be made to thermalize leaked neutrons and use them to transmute these fractions. Some research has been done to study this at the FFTF (Ref. 21). Target assemblies were made with iodine or technetium and included the compound yttrium hydride to act as a moderator. These assemblies were placed outside the fuel region of the core and relied on leaked neutrons. Calculations predict that the 400MWth FFTF could transmute 12kg/yr of Tc-99 or 5kg/yr of I-129 if most of the FFTF radial reflectors were utilized. The FFTF also produces about 3.2kg/yr Tc-99 and 1.1kg/yr of I-129. These initial

results show promise that LMRs could be utilized for transmutation of iodine and technetium as well as the transuranics.

Reactivity

The addition of minor actinides to the metal fuel of the IFR-ALMR causes a reactivity buildup over time as neutron absorption reactions occur with Np-237, Am-241, and Am-242 and cause the production of more fissionable heavy metals. Although with the frequent adjustment of rod stops in the core the reactivity can be controlled, the fractional limit for the amount of minor transuranics which should be added to the metal fuel is about 15 percent by mass. This weight fraction limit in turn restricts the total minor actinides which could be transmuted per year in a ALMR. Depending on the priority which may be placed on transmuting the minor actinides, it is probably best to simply add them in the same fraction they occur in spent fuel, or about ten percent of the plutonium mass. This would most likely be the easiest for fabrication and would keep the reactivity swings low.

4.0 SUMMARY

LWRs operating in the United States generate approximately 2700 MTHM of spent fuel every year. This spent fuel consists of 96% uranium, 1% plutonium, 0.1% minor actinides, and about 3% fission products. At the current rate of production approximately 63,000 MTHM of spent fuel will have accumulated in the U.S. by the year 2011. This amount far exceeds the capacity of today's nuclear utilities to temporarily store spent fuel. With this problem in mind the DOE has been seeking to characterize and construct a geological repository in which to dispose of this waste since 1982.

The repository has met with trouble both politically and technically. Nevada's government and populace have shown enormous resistance to the Yucca Mountain characterization and will make every effort to refuse to host a repository, however safe, in their state. From a technical standpoint the characterization must show with high confidence that the repository will perform within the strict limits set forth by the NRC. Among these limits the most demanding is that the repository's spent fuel inventory after 1,000 years must not exceed dissolution rates of 1 part per 100,000 per year under the worst foreseeable conditions. Tests have shown that radionuclides I-129, Tc-99, Cs-135, and C-14 may all exceed this limit. Under the worst case conditions of an oxidizing, highly saturated repository environment; even the actinides may present dissolution problems. These factors plus others such as waste heat loading and toxicity have led to the identification of the actinides Pu, Am, Np, and Cm, the fission products Cs, I, and Sr, and the activation product C-14 as the key problematic factors in the successful characterization of Yucca Mountain.

The urgency for a resolution of this problem resulted in the re-emergence of partitioning and transmutation as a possible solution. P-T involves the reprocessing of spent fuel to retrieve uranium and plutonium. This is followed by advanced partitioning techniques to remove the minor actinides and key fission and activation products. Partitioned fractions are then either transmuted by various systems employing neutrons or, if transmutation proved impossible, the fractions would be placed in a monitored retrievable storage facility or packaged separately for permanent disposal.

Recent concepts advanced in the United States for partitioning and transmutation have been based on liquid metal reactor systems or accelerator systems.

They all rely on neutrons, fast or thermal, to induce transmutation. The two accelerator based concepts are known as Phoenix (Ref. 9) and ATW (Ref. 13). Phoenix was proposed by Brookhaven National Laboratory. It uses a 1.6 GeV, 104mA proton accelerator to drive a subcritical lattice of minor actinides. The minor actinides are fissioned in a hard neutron spectrum which is unmoderated due to liquid sodium cooling. Operating at 850MWe the Phoenix would transmute 2.6 metric tons of minor actinides per year, while producing 1.5 metric tons of plutonium and 1.05 metric tons of fission products. To maintain subcriticality a large number of neutrons must be leaked from the main fuel area. These neutrons are thermalized and used to transmute approximately 300kg/yr of Iodine-129.

Using similar technology Las Alamos National Laboratory proposed the second accelerator concept known as Accelerator Transmutation of Nuclear Waste, or ATW. The ATW accelerator produces an intense neutron flux by spallating a molten lead-bismuth target with 1.6GeV protons at a current of 30mA. The neutrons are moderated in a deuterium oxide (D_2O)blanket which surrounds the target and they are then used to transmute higher actinides and fission product technetium. With an advantage of high flux and high cross section the ATW can utilize small inventories to achieve high transmutation rates; therefore the system is quite small. Designed primarily for the transmutation of actinides and technetium in defense high level waste the ATW could transmute approximately 70kg/yr of Tc-99 and 10kg/yr of actinides. The small capacity of the ATW makes it unsuitable for the LWR spent fuel but it is nevertheless an innovative and impressive idea. It would also be possible to use a higher current accelerator to increase the capacity of the ATW.

The major concept in the U.S. for LMR based partitioning and transmutation is known as Actinide Burning. Actinide Burning was a change in direction for the Advanced Liquid Metal Reactor Program in the US which was funded by the Department of Energy. Integral Fast Reactors, instead of operating on a breeding cycle, would operate as burners with a breeding ratio of less then one and use the existing actinides in LWR waste as fuel. The actinides in LWR waste by the year 2011 could provide startup and makeup fuel for sixteen advanced liquid metal reactors. These reactors could transmute 170 metric tons of actinides in their forty year lifetimes while providing a sort of storage for the remaining 441 metric tons. In addition, the 441 metric tons could then be used as startup and makeup for a new generation of ALMRs, eliminating the need to ever permanently dispose of these actinides in a repository.

Table III is a breakdown of some of the advantages and disadvantages of each of the three concepts presented.

Table III: List of Transmutation Rates, Inventories, and Advantages/Disadvantages for each P-T concept.

P-T Concept	Total Inventory	Rate of Transmutation	Rate of Production	Advantages	Disadvantages
PHOENIX (Accelerator) Brookhaven National Lab.	24 tons HM of Minor Actinides	2.6 tons per year Minor Actinides and 300 Kg per year Iodine-129	1.55 tons per year of Plutonium, mostly ^{238}Pu, and 1.05 tons per year Fission Products.	Subcritical Lattice High Trans. rate	Criticality increase High Cost Plutonium Production Relatively new technology.
Accelerator Transmutation of Nuclear Waste (ATW) (Accelerator) Los Alamos National Lab.	100 Kg Total: 70 kg ^{99}Tc and 30 kg minor actinides	70 kg per year ^{99}Tc and 10 kg per year minor actinides	Very little actinide waste. 10kg/yr Fission Products.	Subcritical Lattice Very high flux of 10^{16} neutrons per cm^2 per sec, allows high rate of trans. with small inventory.	Low capacity, better suited for defense high level waste transmutation. Relatively new technology
Advanced Liquid Metal Reactors Program: Actinide Burning	27 tons HM, includes Uranium, Plutonium, and the minor actinides.	16 ALMRs transmute 170 tons of Actinides in 40 years, rate is 0.27 tons per year per ALMR Studies show that a 400MWth LMR can transmute 12kg/yr ^{99}Tc or 5kg/yr ^{129}I	Reduction of all Actinide groups when operating as a burner. Some Curium isotopes build up with time. Fission products accumulate and are partitioned during reprocessing. LMRs could burn 99Tc and 129I faster then it is created.	Inherent Safety features of ALMR. Transmutation as well as storage of total actinide inventory from LWRs as of 2011.	Criticality increases with addition of minor actinides to the metal fuel. Development needed to perfect pyroprocessing and metal fuel fabrication.

5.0 CONCLUSIONS

Considering the difficult state that the nuclear industry finds itself in with respect to waste disposal, it is easy to understand why partitioning and transmutation might arouse interest. The benefits which a policy of P-T might provide are substantial, including the reduction of the long term heat load and toxicity, the reduction of the short term heat load, and the resulting increase in repository capacity. It has also been suggested that, given these benefits, the implementation of P-T would actually speed the process of characterization and make licensing and construction of the repository easier. However, as in all new proposals, the risks and costs must be considered as well as the benefits in the evaluation of P-T. Implementation of P-T would be much more expensive then simply burying the waste. This is due to the very high price of reprocessing as well as the high projected costs of the new transmutation devices such as the accelerators and the liquid metal reactors. Another drawback is the negative reaction which would almost certainly result from the public if reprocessing was implemented in the U.S.

The characterization at Yucca Mountain, despite facing opposition and technical difficulties, has acquired a momentum which is difficult to change. The recent re-examination of P-T, although promising, simply does not provide enough concrete incentive to justify its immediate implementation. Therefore, at the present time the characterization will forge ahead based on the once through fuel cycle. This decision is a risky one and failure to obtain a repository will be a very serious problem indeed.

The future of P-T is uncertain in the United States, as is the future of the nuclear industry in general. If, as some predict, there is a resurgence in nuclear power in the U.S., followed by an increase in uranium prices, commercial reprocessing will almost

surely be revived in the U.S.. At such a time it would be rather easy to include advanced partitioning while the new generation of reactors could act as transmutation devices. It seems probable that, should the future bring a resurgence of nuclear power, P-T will be implemented simply as a means to keep the volume of high level radioactive waste to a minimum.

One of the most exciting results of the recent studies into P-T was the idea that the new breed of powerful accelerators could be used to produce power from fission in a subcritical system. Although this area requires much further development it could very well open up a whole new realm of nuclear power generation. If costs could be made competitive, systems such as the PHOENIX and ATW could become viable not simply for their P-T capabilities but for their inherently safe ability to generate power.

In conclusion, the author would point out that future energy demands and the lack of suitable large scale energy alternatives will almost surely bring a revival of nuclear power generation in this country. In preparation of this revival, steps are already being taken to design the new generation of reactors, both liquid metal and light water, to be both safer and simpler than today's reactors. A large scale revival of nuclear power will also be likely to require the use of reprocessing technology in order to conserve limited uranium resources. Perhaps most importantly of all, some technology or policy will have to be established in order to avoid the nagging and destructive problems presently associated with high level waste disposal. This technology or policy will almost surely have to include partitioning and transmutation to some extent. Therefore the author concludes that, while P-T may not be the solution to the problems at Yucca Mountain, its potential benefits are such that it will almost certainly have a place in the future of nuclear power.

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