



PII: S0149-1970(02)00043-4

ATW NEUTRONICS DESIGN STUDIES

D. C. WADE, W. S. YANG, H. KHALIL

Argonne National Laboratory

9700 S. Cass Avenue, Building 208

Argonne, IL 60439

ABSTRACT

The Accelerator Transmutation of Waste (ATW) concept has been proposed as a transuranics (TRU) (and long-lived fission product) incinerator for processing the 87,000 metric tonnes of Light Water Reactor (LWR) used fuel which will have been generated by the time the currently deployed fleet of commercial reactors in the US reach the end of their licensed lifetime. The ATW is proposed to separate the uranium from the transuranics and fission products in the LWR used fuel, to fission the transuranics, to send the LWR and ATW generated fission products to the geologic repository and to send the uranium to either a low level waste disposal site or to save it for future use. The heat liberated in fissioning the transuranics would be converted to electricity and sold to partially offset the cost of ATW construction and operations. Options for incineration of long-lived fission products are under evaluation.

A six-year science-based program of ATW trade and system studies was initiated in the US FY 2000 to achieve two main purposes: (1) "to evaluate ATW within the framework of nonproliferation, waste management, and economic considerations," and (2) "to evaluate the efficacy of the numerous technical options for ATW system configuration."

This paper summarizes the results from neutronics and thermal/hydraulics trade studies which were completed at Argonne National Laboratory during the first year of the program. Core designs were developed for Pb-Bi eutectic (LBE) cooled and Na cooled 840 MW_{th} fast spectrum transmuter designs employing recycle. Additionally, neutronics analyses were performed at Argonne for a He cooled 600 MW_{th} hybrid thermal and fast core design proposed by General Atomics Co. which runs critical for ¾ and subcritical for ¼ of its four year once-thru burn cycle.

The mass flows and the ultimate loss of transuranic isotopes to the waste stream per unit of heat generated during transmutation have been calculated on a consistent basis and are compared. (Long-lived fission product incineration has not been considered in the studies reported here.) © 2002 Published by Elsevier Science Ltd.

INTRODUCTION

The function of the proposed ATW is to reduce the amount of – and the long term toxicity contained in – the waste consigned to the US proposed geologic repository.[1] ATW systems are proposed to be interposed between the commercial LWR once through fuel cycle and the repository and would be used to incinerate the TRU contained in the LWR spent fuel by fission (and optionally to transmute selected long-lived fission products). Heat released by the TRU fissioning would be used to generate electricity or other energy intensive products and sold to partially offset the costs of ATW and recycle construction and operation. Two broad strategies are being evaluated: multi recycle and once-through deep burn. For recycle, an electrometallurgical process is considered and specialized waste forms are developed for the fission products; for deep burn, part of the recycle infrastructure is not required and the spent fuel triso particle fuel is considered as a waste form.

Two aspects of the transuranic content in the waste stream sent to the repository from the ATW are relevant. First is its overall mass – the fraction of TRU from the LWR spent fuel which has escaped being transmuted to fission products. For the deep burn strategy, this aspect is controlled by the achievable discharge burnup. For the multi recycle strategy, it is controlled by the loss per recycle pass and the number of passes. The second relevant aspect is the toxicity per unit mass and its time decay behavior. This aspect depends on the isotopic spectrum of the transuranic mass sent to the repository (based on differing toxicity and half-life by isotope). For the deep burn strategy this aspect is affected primarily by the neutron spectrum and secondarily by discharge burnup. For the multi recycle strategy it is affected by neutron spectrum and by conversion ratio (i.e., blend ratio of recycle and feed).[2]

The focus of this paper is a comparison of isotopic mass flows to the repository from three ATW point designs established during the first year of the 6-year science-based ATW program.

Liquid Metal Cooled Multi-Recycle ATW Point Designs

For the liquid metal cooled ATW concepts, a fertile-free fuel was selected so as to maximize the “support ratio” defined as number of LWRs that a given ATW can service (MW_{th} from LWRs/ MW_{th} from ATW). A fuel form comprised of TRU-Zr alloy particles dispersed in Zr matrix in a pin geometry and clad in ferritic stainless steel has been proposed; the average discharge burnup capability for this fuel is postulated to be 30 a/o.[3] In the case of a multi recycle strategy, the goal of minimizing loss of LWR TRU feedstock to the waste stream motivates the design for maximum achievable discharge burnup to minimize number of recycle passes (to reduce opportunity for losses during recycle and refab processing). On the other hand, for fertile free fuel, the source multiplication in the subcritical blanket decreases with increasing burnup due to the reactivity loss and in order to minimize the resulting needs for increasing accelerator power and/or introducing an excess reactivity and active reactivity control, it is desirable to minimize the burnup reactivity loss. The LBE and Na cooled design optimizations were, therefore, focused on trading off two contradictory performance objectives: achieving 30 a/o discharge burnup to minimize number of successive recycle stages while minimizing burnup reactivity loss over an operating cycle – and to do so within the constraints of heat removal under acceptable temperature and coolant velocity limits, and of discharge fluence level, reactivity coefficient values, vessel size, and etc. Given each choice of coolant, a wide range of potential transmuted designs was evaluated at a fission-power level of 840 MW_{th} , driven by a 1 GeV, 11.25 mA proton beam on a LBE spallation target and

operating at a neutron multiplication level at beginning of equilibrium cycle (BOEC) of 0.97.[4,5] The resulting parameters of the optimized liquid metal cooled transmuter core designs are shown in Table I. In both cases, multi batching was used to reduce burnup reactivity loss and radial power peaking; a 7 batch core/42 month fuel residence time for LBE and 8 batch core /48 month fuel residence time for sodium was necessary to hold reactivity loss to ~5% $\Delta k/k$ over a 6 month burn cycle. A reduced power density/high coolant volume fraction design was used for the LBE coolant while conventional values were used for Na. Peak discharge fluence ($40 \cdot 10^{22}$ fast nvt) controlled the fuel residence time, but in both cases discharge burnup nearly attained 30% burnup as was targeted for the inert matrix dispersion fuel. The details of the optimizations are reported elsewhere.[4,5] peaking limited the residence time to 6 batches; a similar notation applies for the sodium system. See Fig. 1 for the gas system fuel loading logistics.

Table I. Main Integral Parameters of LBE, Sodium, and Gas Cooled System Point Designs

Parameter		LBE	Sodium	Gas	
				Critical	Subcritical
Reactor power (MWt)		840.0	840.0	600.0	
Cycle length (days)		137.0	135.0	270.0	
Number of fuel assemblies	Inner	36	42	6	6
	Outer	168	90	102	102
Number of batches*		7/6	8/7	3	1
Multiplication factor	BOEC	0.9703	0.9696	1.0775	0.9634
	EOEC	0.9180	0.9202	1.0062	0.7323
Burnup reactivity loss (% Δk)		5.23	4.94	7.13	23.11
Core-average total flux ($n/cm^2 \cdot s$)	BOEC	4.59E+15	4.41E+15	8.02E+13	1.01E+14
	EOEC	4.99E+15	4.75E+15	8.99E+13	1.63E+14
Core power peaking factor	BOEC	1.46	1.50	1.97	4.79
	EOEC	1.51	1.51	1.64	23.79
Core-average power density (W/cc)	BOEC	156.46	241.36	6.21	6.21
	EOEC	156.26	241.98	6.21	6.21
Coolant Volume Fraction	%	68.2	36.6	18.6	
	atom %	26.79	29.51	47.54	16.44
Average discharge burnup	MWD/kg	250.8	275.2	445.1	153.9
Effective cycle burnup of charged fuel (%)		3.93	3.84	15.85	16.44
% Δk / atom % burnup		1.33	1.29	0.45	1.41
Core ht/diam**	M	1/2.5	1/2.1	7.9/4.9	

*7/6 for the LBE system indicates a 7 batch core – except for the inner most fuel zone where power

**Equivalent fuel region outer diameter of annular core around central spallation target and buffer.

Gas Cooled Hybrid Once Thru ATW Point Design

General Atomics (GA) has proposed an ATW concept[6] based on a variant of the Gas Turbine-Modular Helium Cooled Reactor (GT-MHR). Four transmuters share one accelerator, and each transmuter is comprised of an outer thermal zone and inner fast zone. Each transmuter operates in a three batch-loaded critical mode for three years and in the source driven mode for the fourth year. In the three-years-long

critical operating mode, the fission process is maintained by the critical thermal region driving the subcritical fast region and discrete burnable poison limits reactivity loss. After three years, the thermal region, loaded with three year burned assemblies, itself becomes subcritical and the transmuter is driven during the fourth year by the spallation source. The overall plant is comprised of four 600 MW_{th} transmuters, sharing one 15 MW accelerator with beam shifting from core to core at one year intervals. The transmuter thermal zone is fueled in TRISO coated particles with (fertile free) TRU recovered from LWR used fuel. The fast zone is fueled with four-year-burned TRISO particles which have been discharged from the thermal zone, separated from the graphite compacts and moderator, and reconfigured into fuel rods. Figure 1 illustrates the loading sequence of fuel as it progresses through its four-year burn cycle. Burnup reactivity loss is mitigated by use of discrete burnable poison (erbium) rods distributed in the thermal zone and benefits from the in situ conversion of Pu²⁴⁰ in the feedstock to fissile Pu²⁴¹. The absorption resonance in erbium shields the principal Pu²³⁹ fission resonance and produces a negative moderator temperature coefficient.

Preliminary analyses by GA have indicated that this design will achieve deep levels of transmutation without requiring reprocessing so that it can be operated once thru, and that it can employ the spent TRISO particles as a waste form thanks to encapsulation of the transmuted materials within the ceramic coated microspheres. Argonne collaborated with General Atomics staff to perform independent core performance and mass flow calculations for the gas cooled hybrid design.[7] Table I summarizes the main parameters of all three ATW point designs studied in the first year of the program.

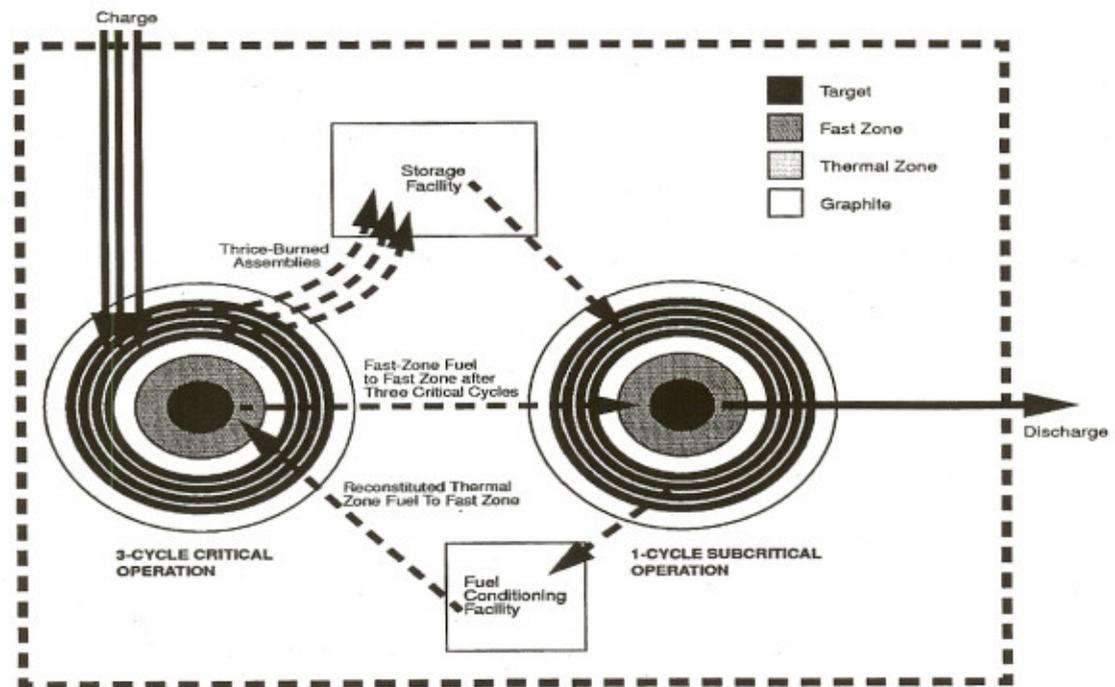


Fig. 1. 3-Batch GT-MHR Coupled to an AD-MHR

Comparison per Unit of Fission Energy of Isotopic Losses into the Waste Stream

Detailed comparisons of equilibrium-cycle mass-flows and discharges to the waste stream destined for the repository were made for the LBE, sodium, and gas cooled system point designs. Because the LE and sodium system point designs are each 840 MW_{th} whereas the gas system point design was for

cluster of four 600 MW_{th} systems, all comparisons were made consistent by basing mass flows on a normalization to the same MW_{th} of fission power.¹

The TRU consumption per unit of energy production is of course ~1.0 g/MW_{th} day for all systems, because the energy released per fission is approximately constant across all TRU isotopes. It is the evolution of the isotopic mix and the ultimate loss to waste which is of interest. Incore isotopic inventories and consumption rates, normalized to one MW_{th} of fission power, are compared in Table II. First comparing the two liquid metal options, the slightly lower TRU inventory of the sodium system compared to the LBE system is perhaps surprising. For a fixed blanket size, a sodium system would require a higher TRU inventory than a LBE system because of greater neutron leakage, but the sodium system design exploits a higher coolant velocity (and much lower coolant volume fraction) producing a more compact, higher power density, and less leaky core with slightly lower TRU inventory. As shown in Table I, when designed to the same limit on discharge fast fluence, the sodium system attains a slightly higher discharge burnup than the LBE system because the neutron energy spectrum is not as hard so the fast fluence to burnup ratio is slightly lower.

Table II. Isotopic Inventories at BOEC, Consumption Rates, and Annual Burnup at 75% Capacity Factor

Isotope	BOEC inventory (g/MWt)			Average consumption per day (g/MWd)			Atom % burnup per year @ 75% CF		
	LBE	Sodium	Gas	LBE	Sodium	Gas	LBE	Sodium	Gas
U-234	16.81	15.91		0.0023	0.0021		3.55	3.34	
U-235	4.07	3.97	0.03	0.0001	0.0001	0.0000	0.48	0.46	-*
U-236	5.56	5.28	0.13	0.0002	0.0002	-0.0001	0.97	0.91	-
U-238	36.69	34.64	0.00	0.0049	0.0049	0.0000	3.49	3.64	-
Np237	81.28	78.58	95.12	0.0523	0.0519	0.0437	13.74	13.60	16.60
Pu238	171.65	171.14	126.03	0.0242	0.0225	-0.0622	3.63	3.39	-80.77
Pu239	770.49	758.56	630.39	0.5486	0.5460	0.8119	14.79	14.41	24.55
Pu240	1057.65	1038.64	506.42	0.2279	0.2262	0.2903	5.44	5.45	18.95
Pu241	185.13	189.25	339.48	0.0248	0.0258	0.0614	3.50	3.54	11.98
Pu242	363.14	359.49	278.48	0.0483	0.0481	-0.0710	3.46	3.46	-22.12
Am241	191.72	185.21	85.55	0.1062	0.1047	0.0611	12.21	12.06	19.04
Am242	13.68	13.32	1.99	0.0000	0.0001	0.0012	0.09	0.13	19.05
Am243	122.33	118.70	86.81	0.0095	0.0095	-0.0502	2.07	2.11	-78.24
Cm242	12.38	12.62	18.77	-0.0136	-0.0117	-0.0046	-328.96	-296.51	-
Cm243	1.27	1.38	0.50	0.0000	0.0000	-0.0003	-0.72	-0.64	-
Cm244	91.33	91.47	47.21	-0.0050	-0.0044	-0.0343	-1.54	-1.35	-267.07
Cm245	25.10	25.40	3.51	0.0001	0.0001	-0.0095	0.10	0.10	-
Cm246	17.06	15.92		0.0000	0.0000		0.01	0.01	
Pu	2548.06	2517.07	1880.79	0.8738	0.8686	1.0305	8.21	8.11	17.95
MA	556.13	542.60	339.44	0.1495	0.1501	0.0071	6.71	6.78	1.31
TRU	3167.33	3119.46	2220.39	1.0308	1.0259	1.0375	7.86	7.80	16.22

*Isotopic burnup is not defined for these isotopes, since they are not included in the feed stream but are later produced by transmutation.

¹ Even though LWR-discharge TRU (33,000 MWd/t discharge and 25y cooling) was used for all three designs, slightly different feed compositions and depletion chain models were employed for the gas-cooled system as compared with the fast spectrum cases because of peculiarities of the differing neutronics codes used to analyze the thermal system.

The gas system BOEC inventory in Table II is the initial inventory for critical mode operation. Since the thermal region is dominant, its critical mass TRU inventory is smaller than for fast systems; the TRU inventory per unit fission power is ~70% of that of liquid metal systems. Correspondingly the atom % burnup per MW_{th} of the gas system is nearly twice that of the liquid metal systems because the TRU inventory of the gas system is significantly lower and the subcritical cycle is operated only with previously burned fuels.

The proportion of minor actinides in the gas system BOEC inventory is lower (and that of Pu-239 higher) compared to the LBE and sodium systems because the gas system is fueled with 100% LWR discharge TRU whereas the recycle fuel charged to the liquid metal systems has a significant self-recycle blending component. Furthermore, due to the large capture cross section of Pu-240 in the thermal energy range, the gas system end of equilibrium cycle (EOEC) inventory has significantly greater Pu-241 fraction than the liquid metal systems.

The gas system burns plutonium Pu-239, 40 and 41 isotopes more effectively, but minor actinides and Pu 238 and 242 less effectively than the LBE and sodium systems. In the LBE and sodium systems, all isotopes except for Cm-242 and Cm-244 are net consumed, whereas in the gas system, *net production* of all minor actinides except for Am-241, Am-242, and Np-237 occurs. It was observed that even Pu-241 and Pu-242 are net produced during the critical mode segments of operation of the gas system, so that in the accelerator-driven segment of the cycle, the minor actinide inventory further increases (negative net consumption) owing to neutron capture on Pu-241 and Pu-242.

Table III shows the annual isotopic feed of LWR-discharge isotopes compared to the annual isotopic waste stream (i.e., the "leakage loss" to the repository) for the three systems.² In the estimation of the isotopic losses from the ATW to the repository, all fuel discharged from the gas system was assumed to go to the waste stream. For the LBE and sodium systems, a fraction of the discharge fuel was assumed lost to the waste stream on each recycle step. The overall (per recycle step) loss factor (recycle plus refab) was arbitrarily assumed³ here to be 0.1%. (For other recycle loss fractions, the isotopic losses are closely scaleable to this fraction because the evaluation has been done for an equilibrium cycle).

The external LWR feed per MW_{th} for the LBE and sodium system are much smaller than that of the gas system because external feed is used only to makeup the TRU consumed by fission – whereas the gas system operates once through and LWR discharge feed must therefore make up for both fissioned and discharged fuel. The LBE system releases 2.7% of the LWR TRU feed (3.3% of MA) to the waste stream, and the sodium system releases 2.4% of the TRU feed (2.8% of MA). The gas system releases only 3% of the Pu²³⁹ charged but releases altogether 36% of the TRU feed to the waste stream; i.e., much of its fuel has been transmuted to higher mass isotopes, incompletely burned and sent to waste. The amount of minor actinides discharged from the gas cooled system to the waste stream is comparable to the amount of LWR-discharge minor actinides initially loaded.

² The loss to waste reported here neglects any that would occur in processing the LWR fuel for TRU recovery and fabrication into the initial ATW transmuter loading. It also neglects losses in reconfiguring the gas system thermal zone discharge fuel into the fuel for the fast zone. Moreover, spallation and activation products in the core and accelerator are not accounted for.

³ A design target for the loss per cycle from the ATW has been set much smaller – at 0.1% cumulative loss over multi recycle steps.[1]

Table III. External (LWR-Discharge) Isotopic Feeds and Isotopic Losses from ATW System per Year (based on 75% Capacity Factor)

Isotope	External feed per year (g/MWt)			Mass loss per year (g/MWt)		
	LBE	Sodium	Gas	LBE	Sodium	Gas
U-234	0.000	0.000		0.0046	0.0039	
U-235	0.011	0.011		0.0012	0.0010	0.0038
U-236	0.006	0.006		0.0016	0.0014	0.0300
U-238	1.349	1.343		0.0100	0.0084	0.0004
Np237	14.180	14.112	18.252	0.0162	0.0134	6.2971
Pu238	3.590	3.573	5.342	0.0469	0.0419	22.3651
Pu239	150.167	149.447	229.481	0.1472	0.1244	7.2168
Pu240	60.787	60.495	106.304	0.2731	0.2382	26.8271
Pu241	10.677	10.625	35.568	0.0500	0.0456	18.7595
Pu242	13.228	13.165	22.258	0.0987	0.0874	41.6827
Am241	25.313	25.191	22.258	0.0407	0.0341	5.5363
Am242	0.040	0.039	0.445	0.0037	0.0032	0.1107
Am243	2.615	2.602	4.451	0.0343	0.0298	18.1921
Cm242	0.000	0.000		0.0041	0.0035	1.2703
Cm243	0.006	0.006		0.0004	0.0004	0.0748
Cm244	0.293	0.292	0.890	0.0275	0.0248	10.2686
Cm245	0.026	0.026		0.0074	0.0068	2.6103
Cm246	0.003	0.003		0.0050	0.0042	
Pu	238.449	237.305	398.953	0.6159	0.5375	116.8511
MA	42.476	42.272	46.297	0.1393	0.1201	44.3602
TRU	282.291	280.937	445.250	0.7725	0.6723	161.2459

CONCLUSIONS

ATW transmuter core point designs have been developed for LBE and Na cooled concepts based on a multi-recycle strategy; a gas cooled hybrid concept based on a deep burn once thru strategy which has been proposed by General Atomics and has been independently analyzed.

Mass flows have been calculated and fractions of LWR feedstock lost to the waste stream were compared for the three concepts on a consistent per MW_{th} basis. The once thru deep burn strategy employed in the Na cooled thermal/fast spectrum hybrid concept avoids the costs of some of the recycle/refab equipment and reduces TRU in the repository as compared with an LWR once through strategy by ~60%. On the other hand, the discharged isotopic spectrum is unfavorable – with essentially unchanged MA mass flow to the repository as compared with LWR spent fuel. The presence of Pu²⁴¹ in large amounts in the discharge is especially undesirable because its ultimate decay daughter is Np237 which, with a 2.14 million year half-life and high toxicity factor, constitutes the dominant long term (i.e., subsequent to 10,000 years) toxicity hazard in the Yucca Mountain repository source term[1]. The substantial burnup in a soft neutronic spectrum coupled with the small burnup increment attainable in the fast zone of this particular point design is ill suited to minor actinide consumption.

The fast spectrum multi recycle concepts which were examined bear the extra cost of recycle, but they achieve greater overall reduction in mass sent to the repository. Based on an assumed 1% loss fraction per recycle pass, used in these calculations, the multi recycle strategy using either of the two fast spectrum liquid metal cooled transmuter designs and a metallurgical recycle technology achieves a factor of about 400 reduction in TRU as compared with LWR once thru and achieves a reduction of about 325 in MA mass. The design goal for ATW recycle is 0.1% cumulative for multi recycle; if achieved, the reduction factors would reach a value of 1000.

The desired degree of reduction in waste mass and toxicity and the cost-to-benefit ratio of achieving any specified level of reduction are issues of public debate and are not yet resolved. The tradeoff analyses reported here are part of the technology program intended to inform that ongoing debate.[1] The point designs completed thus far focus on core neutronics, mass flows, and heat removal. Future work will address further optimization and will add dynamics, safety evaluations, and cost considerations.

ACKNOWLEDGMENTS

The detailed core design optimizations and analyses which provide the basis for this comparison were performed by Yang, Hill, Taiwo and Khalil and are reported in References 4, 5, and 7.

REFERENCES

1. US Dept. of Energy (Oct. 1999), *A Roadmap for Developing Accelerator Transmutation of Waste (ATW) Technology; A Report to Congress*, DOE/RW-0519.
2. R. N. Hill et al (September 1995), Physics Studies of Weapons Plutonium Disposition in the Integral Fast Reactor Closed Fuel Cycle. *Nucl. Sci. Eng.* **121**, No. 1.
3. D. Crawford and M. Meyer (Nov. 28 - Dec. 1, 2000 to be published), Current Plans for US ATW Blanket Fuel Development, *Proceedings of the IAEA Technical Committee Meeting on Core Physics and Engineering Aspects of Emerging Nuclear Energy Systems for Energy Generation and Transmutation*, Argonne National Laboratory.
4. W. Yang, H. Khalil (to be published), *Blanket Design Studies of an LBE Cooled Accelerator Transmutation of Waste System*, *Nuclear Technology*.
5. R. N. Hill, et al (Nov. 28 - Dec. 1, 2000 to be published), *Physics Studies of a Sodium Cooled ATW Design*, *Proceedings of the IAEA Technical Committee Meeting on Core Physics and Engineering Aspects of Emerging Nuclear Energy Systems for Energy Generation and Transmutation*, Argonne National Laboratory.
6. A. Baxter and Carmelo Rodriguez March 2000, *Private Communication*, GA.
7. T. Taiwo, et al (Nov. 28 - Dec. 1, 2000 to be published), Core Physics Performance of Recycled LWR Discharge TRU Oxide Fuel in a GT/AD-MHR, *Proceedings of the IAEA Technical Committee Meeting on Core Physics and Engineering Aspects of Emerging Nuclear Energy Systems for Energy Generation and Transmutation*, Argonne National Laboratory.