

THE SYMBIOTIC RELATIONSHIP BETWEEN WASTE BURNING AND SAFETY IN LIQUID METAL REACTORS

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ABSTRACT

The relationship between the transmutation of minor actinides and fission products, and safety related reactivity feedbacks in liquid metal reactors (LMR) was explored. Several design features appear promising for performing waste transmutation while retaining the desirable safety characteristics. Innovative variations of conventional LMR configurations and compositions establish symbiotic relationships between plutonium fuel, minor actinides, and fission products. These relationships enhance safety characteristics of the core and provide acceptable fuel and burnup performance. Although a specific design has not been developed, an LMR capable of transmuting the minor actinides and fission products from up to 10 comparable light water reactors while retaining desirable safety features, appears to be feasible.

L INTRODUCTION

A fully developed nuclear economy would include nuclear power plants, processing facilities, near-surface storage, and high-level waste repositories. One concept of the reprocessing facilities is represented schematically in Figure 1. Components illustrated in this figure include not only isotope separation and fuel fabrication plants, but also advanced reactors or accelerators. The function of the advanced reactors or accelerators in this concept is to transmute long-lived fission products and minor actinides into stable isotopes, or useful by-products.

The focus of this paper is the potential of the LMR to serve as a transmuter. The basic premise was to answer the question, Can an LMR be designed to destroy the minor actinides and long-lived fission products (^{99}Tc and ^{129}I) produced by itself and as many as 10 conventional LWRS? If such a reactor could be shown to be feasible, then partitioning and transmutation as a way to enhance the performance of deep geologic repositories becomes a much more attractive alternative.

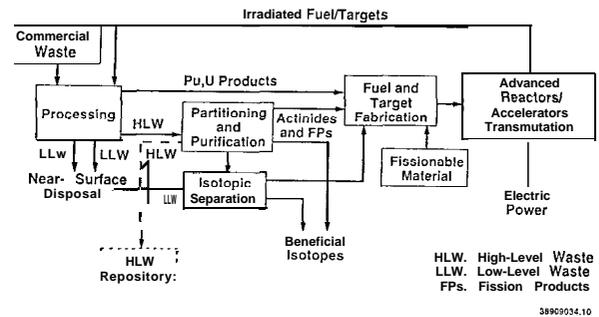


Fig. 1. Schematic of a Reprocessing Concept.

The ability of sodium cooled fast reactors to transmute minor actinides and fission products has been widely investigated. The current focus of LMR designs is on modular units with enhanced safety characteristics. Adapting these designs to perform waste transmutation generally is detrimental to safety characteristics. However, results of recent studies^{2,3,4,5,6} have been examined to determine the feasibility of an LMR waste burner using innovative ways to retain most of the desirable safety characteristics.

Smaller, modular cores have better (lower) sodium void reactivity worth than large cores, and potentially better passive safety characteristics.⁷ Consequently, analyses centered on designs in the 400-to-500-MWth range. Innovative core geometries that dramatically reduced the sodium void reactivity were considered. Included were concepts featuring pancake, parfait, annular, and split core designs. These designs have high neutron leakage rates from the active core into regions containing target material. However, only pancake and parfait designs were actually analyzed. Moderated target assemblies on the core periphery were included to improve fission product transmutation rates.

This study was concerned primarily with the physics aspects of the problem. Questions concerning material

compatibility, fabricability, and operational issues were not fully addressed. Physics parameters evaluated in the course of the study were as follows:

- ^{99}Tc , ^{129}I , and minor actinide destruction rates
- The contribution of ^{99}Tc to the Doppler effect
- Sodium void reactivity worth.

II. CHARACTERISTICS OF LMR WASTE BURNERS

The basic concept examined was that of a single facility, which **could** be composed of smaller modules, that would be capable of transmuted all the minor actinides (neptunium, americium, and curium) and the long-lived mobile fission products (^{99}Tc and ^{129}I) produced annually by its own operation and by ten 3,400-MWth uranium fueled LWRs. The scope was limited to sodium cooled fast reactor concepts using plutonium based fuel.

To support 10 LWRs in addition to itself, the burner would need to destroy approximately 320 kg of ^{99}Tc and 80 kg of ^{129}I per full power year.¹ This includes the fission products generated by the burner itself. The destruction of approximately 400 kg of minor actinides per year would support the 10 LWRs. The quantity of minor actinides generated by the burner varies with design and fuel composition. Whatever is generated by the burner must be added to the 400 kg. Obviously, LMR burner designs that minimize the production of minor actinides are more desirable. For this study, the plutonium generated by the LWRs is considered a fuel resource. However, recycle of this plutonium back into the LWRs would increase the assumed minor actinide generation rate and change the support ratio.

Because this was only a feasibility study, a safety analysis of various concepts was not done. The parameters used to judge safety performance were the sodium void and Doppler reactivity worths. A sodium void reactivity close to zero, or negative is desirable. The target sodium void worth in this study was one dollar ($\$$) or less for the active core region and zero or negative for the total core. Specific criteria for an acceptable Doppler feedback were harder to define. Of course, it should be negative, but the optimum value depends on the accident scenario and the effect of other feedbacks and control requirements.

III. CALCULATIONAL METHODS

Results were based on both the three-dimensional diffusion theory and Monte Carlo analyses. The diffusion calculations were performed using the 3DB⁸ code. The 53 and 12 energy group nuclear cross sections used in the 3DB calculations were generated by the NJOY⁹ and 1DX¹⁰ programs from ENDF/B-V data. These programs employ the shielding factor method¹¹ to account for resonance self-shielding.

The Monte Carlo computer code for neutron photon transport (MCNP¹²) was used because of its versatility, comprehensive geometry features, and its overall physics capabilities, including continuous energy treatment. MCNP

calculations were relied on primarily for analyses involving moderated peripheral target assemblies, where diffusion theory could not be accurately employed.

IV. CALCULATIONAL MODELS

Two basic reactor models were analyzed in these studies. The first, shown in Figure 2, was based on the 400-MWth Fast Flux Test Facility (FFTF) core with mixed oxide fuel. This model contained three safety rods in assembly row 3, and six control rods in row 5. The other 82 assembly locations in the first six rows contained fuel or target assemblies. The inconel reflector assemblies normally in FFTF rows 7, 8, and 9 were replaced. All row 7 locations then contained silicon carbide (SiC) filter assemblies. Twenty-four locations in row 8 (4 on each flat) contained moderated target assemblies. The other row 8, and all row 9 locations contained SiC shield assemblies.

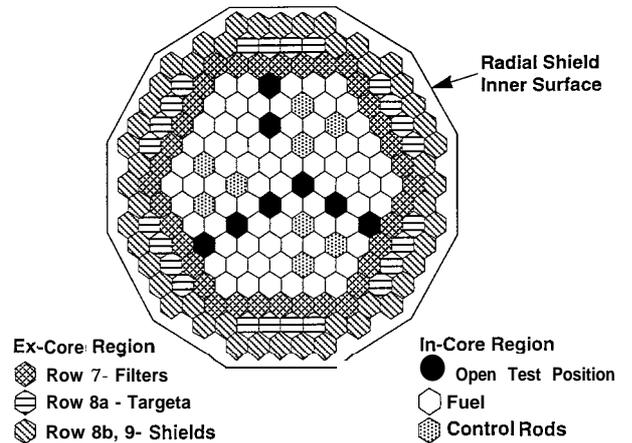


Fig. 2. Core Map of the FFTF-Based Calculational Models.

Each moderated target assembly consisted of 19 large pins containing yttrium hydride (YH 1.7), and 36 small target pins distributed between the moderator pins, as illustrated in Figure 3.

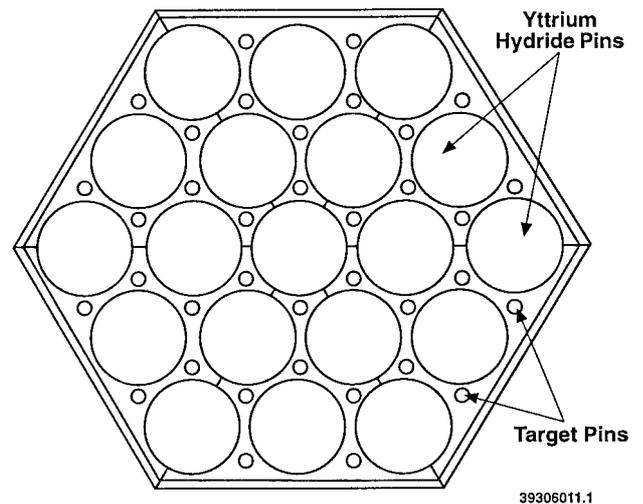


Fig. 3. Moderated Target Assembly.

The MCNP model contained pin detail in the filter, target, and shield assemblies. Thus, self-shielding effects, spectral changes, and flux gradients could be accurately computed. The 3DB models in triangular-Z geometry represented all assemblies homogeneously in the X-Y plane.

The second reactor model (Figure 4) was based on the United States advanced LMR (ALMR) design¹³ that features a 471-MWth modular core with a metal fuel matrix. Both the FFTF and the ALMR design contain 199 lattice positions. However, the lattice pitch in the ALMR design is larger (16 cm versus 12 cm in FFTF), and the ALMR core is taller (135 cm versus 91 cm).

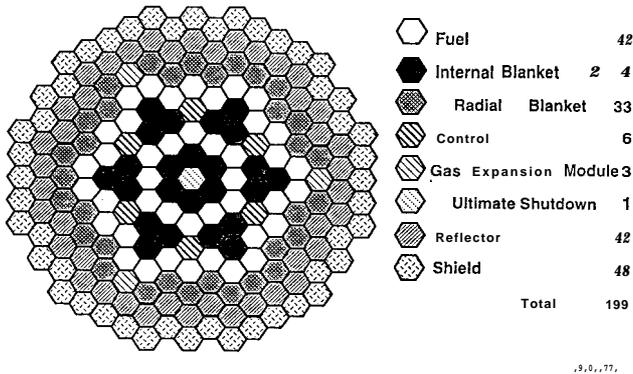


Fig. 4. Core Map for the U.S. Advanced LMR.

Because ⁹⁹Tc is compatible with fuel, and provides Doppler and sodium void benefits described later, models including this isotope in a variety of configurations and concentrations have

been analyzed.^{3,5} Some of these configurations included ⁹⁹Tc mixed in fuel pins, a ⁹⁹Tc material layer in a parafit fuel pin design, separate in-core target assemblies, and peripheral moderated target assemblies.

On the other hand, ¹²⁹I is not compatible with fuel and provides no safety related enhancements. In most reactor models, this isotope, in the form of cerium iodide (CeI₃), was located in separate pins within peripheral moderated target assemblies. The iodine composition was assumed to be 75% ¹²⁹I and 25% ¹²⁷I.

V. FISSION PRODUCT TRANSMUTATION RATES

Analyses reported in References 3 and 5 indicate that a good arrangement for transmuted ⁹⁹Tc and ¹²⁹I in an LMR is to load ⁹⁹Tc into the core, and ¹²⁹I into moderated peripheral target assemblies. The in-core ⁹⁹Tc can be included in the fuel matrix, or loaded into separate target assemblies. Key results from those analyses are summarized in Table 1. The reactor model analyzed was the FFTF-based design. To achieve a ten-to-one (10: 1) support ratio, such a 400-MWth module must eliminate about 38 kg of ⁹⁹Tc and 9 kg of ¹²⁹I per full-power year. From Table 1, it is estimated that the goal amount of ⁹⁹Tc can be transmuted by replacing 30% of the fuel volume with technetium metal. However, this volume fraction may not be practical. A combination of separate in-core targets, peripheral targets, and ⁹⁹Tc in the fuel matrix maybe a more practical means of meeting the stated goal in the unoptimized core design analyzed.

Table 1. Fission Product Transmutation Rates in an LMR.

Target Type	Target Location (Row)	Target Isotope	Target Isotope Mass		Destruction Rate	
			Kg/Assy	Total Kg	%/yr	Kg/yr
Moderated	8	⁹⁹ Tc	2.5	60.5	11.6	7.0
Moderated	8	⁹⁹ Tc	15.6	373.4	4.8	18.0
Moderated	8	⁹⁹ Tc	31.0	746.8	3.2	24.0
Unmoderated	2,4,6	⁹⁹ Tc	46.7	373.4	4.1	15.3
1% of fuel	1 - 6	⁹⁹ Tc	0.47	38.5	7.0	2.7
10% of fuel	1 - 6	⁹⁹ Tc	4.6	373.4	5.2	19.6
50% of fuel	1 - 6	⁹⁹ Tc	23.3	1914.0	2.7	52.2
Moderated	8	¹²⁹ I	0.72	17.3	14.0	2.4
Moderated	8	¹²⁹ I	4.2	101.0	9.3	9.4
Moderated	8	¹²⁹ I	8.4	202.0	6.2	12.5

Table 1 also shows that the ^{129}I goal destruction rate of 9 kg/year can be met by irradiating 24 peripheral moderated target assemblies containing 4.2 kg of ^{129}I each. If higher assembly loadings are used, fewer targets would be required. Unneeded ^{129}I targets could then be replaced by ^{99}Tc targets.

VI. TECHNETIUM-99 DOPPLER EFFECT

The Doppler reactivity effect of ^{99}Tc relative to ^{238}U was computed in 3DB for one loading in the FFTF model. This case had 82 fuel assemblies in which 10% of the fuel volume was replaced with ^{99}Tc metal, and had ^{99}Tc in 24 moderated row 8 target assemblies. Only the temperature of the ^{99}Tc in the fuel was varied. Temperature dependent, 53-group cross-section data used in the 3DB calculations were generated in IDX, as described in Section III.

Results of the 3DB eigenvalue calculations indicated that the Doppler reactivity coefficient for ^{99}Tc is 60% of that for ^{238}U on a per atom basis. This result suggests that ^{99}Tc may provide a significant Doppler feedback when included in an LMR fuel matrix.

VII. MINOR ACTINIDE DESTRUCTION RATES

Numerous physics studies have shown that actinide waste discharged from LWRS could be burned in LMRs as well as other systems. Relationships between safety/economic characteristics and minor actinide burn rates usually are established. In a recent study,⁴ sodium void reactivity worth was compared to minor actinide support ratio for a selection of core configurations based on the ALMR design illustrated in Figure 4. The ALMR was fueled with a combination of plutonium, uranium, and minor actinides using the fuel pin and assembly dimensions of Reference 13. However, nitride fuel was used, rather than metal fuel.

Two core heights (76 cm and 135 cm) were included to evaluate the effect of core height on sodium void worth. Void worths were computed for cores containing LWR discharge, minor actinide compositions, and for cores containing minor actinides recycled through an LMR.

The results of that study indicate that an LMR burner with a minor actinide support ratio of 10:1 and sodium void coefficient no larger than for present conventional LMR designs is feasible. Depending on where the actinides are located in the core, 20% to 30% of the fuel assemblies would need to be minor actinide targets (considering these targets as a fuel resource). The addition of these actinides increases the sodium void reactivity by an estimated 3\$.

Minor actinides discharged from LWRS increase in reactivity worth with exposure in an LMR. Consequently, they can be used to decrease the burnup reactivity swing, and thus decrease control requirements, with benefits discussed in Section X.

VIII. REDUCTION OF SODIUM VOID REACTIVITY

There are a number of features that can be incorporated into an LMR design to reduce the coolant void coefficient of reactivity. However, in a dedicated burner, the idea of inserting a ^{99}Tc layer at the core midplane resting in a parfait core, is appealing. Reference 6 explored this concept for a core design based on the one shown in Figure 4. However, all blanket assemblies were replaced by either fuel assemblies, or parfait assemblies. The parfait assemblies consisted of a ^{99}Tc target layer sandwiched between an upper and lower fuel layer. The target layer was centered at the core midplane.

Calculations were performed using the 3DB program and a one-sixth core model in triangular-Z geometry. The thickness of the target layer was varied from approximately 17 to 68 cm. The parfait assemblies were sequentially loaded in rows 2 through 5. In all, The sodium void reactivity was calculated for 16 core configurations. The fuel enrichment was adjusted, as necessary, to keep k_{eff} close to unity.

The void coefficient was calculated by removing the coolant from the fuel and parfait assemblies. The analysis showed that it was advantageous to load the parfait assemblies in all available locations in rows 2 through 5. Results for various ^{99}Tc layer thicknesses are summarized in Table 2. The case with the 34-cm layer of ^{99}Tc in the parfait assemblies provides a near-zero sodium-void coefficient. This is a sharp contrast to the case with no ^{99}Tc layer, which had a positive void worth of about 9.8\$. There were slight reductions in the sodium void worth, and modest improvements in the ^{99}Tc destruction rates for thicker layers. But, increasing the ^{99}Tc layer thickness beyond 34 cm is probably not warranted.

Table 2. Effect of a Technetium Layer in Rows 2-5 Fuel Assemblies. "

Tc Layer Thickness (cm)	Total Tc Mass (kg)	Sodium Void Reactivity (\$)	Tc Destruction Rate	
			%/yr	Kg/yr
0.0	0	9.8	-	0.0
16.94	659	8.3	1.8	12.0
33.88	1317	-0.1	1.5	19.8
50.82	1976	-0.6	1.0	21.6

IX. OPTIONS, TRADEOFFS, AND SAFETY CONSIDERATIONS

To achieve a high-support ratio, it was necessary to reduce parasitic captures in the core by reducing the amount of ^{238}U in the fuel composition. But, this impacts at least two safety parameters. First, the Doppler coefficient, a function of ^{238}U concentration, is reduced. However, adding ^{99}Tc to the fuel matrix in place of ^{238}U provides a significant Doppler feedback.

Second, with less ^{238}U available to breed ^{239}Pu , the reactivity swing caused by fuel burnout increases. As a result, control requirements correspondingly increase. Loading minor actinides in the core reduces control requirements, but also increases the sodium void reactivity.¹⁴ This tradeoff was not investigated.

The increased sodium void reactivity associated with the addition of minor actinides to the core can be more than offset by including a ^{99}Tc layer at the center of the fuel assemblies. Calculations that lead to this conclusion were discussed in Sections VII and VIII.

Calculations described in Sections V through VIII used metal, oxide, and nitride fuel. However, no calculations provide direct comparisons of fuel types. Higher fissile densities can be obtained with metal fuel. As a result, potentially more ^{99}Tc can be included in the fuel matrix. But, the sodium void characteristics of metal fuel are not as good as oxide or nitride fuel. Sodium void reactivity worths are nominally 3% higher in a metal fueled LMR than in either an oxide or nitride fueled LMR. Nitride fuel appears to be a good compromise between metal and oxide fuel. Sodium void worths are comparable to oxide, but fissile densities are significantly higher than in oxide fuel. There is, however, not as much experience with nitride fuel as with metal or oxide fuel.

X. CONCLUSIONS

The purpose of the work described in this paper was not to arrive at a single, thoroughly analyzed design that provides the goal of a 10:1 support ratio. It was, however, to complete or draw on parametric studies that would indicate whether this goal is obtainable, and that could serve as a basis for future detailed design calculations.

One set of calculations indicates that a 10:1 support ratio for fission product (^{99}Tc and ^{129}I) destruction is achievable. Another set of calculations indicates that a 10:1 support ratio for minor actinide destruction is achievable. The question is, Can an LMR that has a 10:1 support ratio for both fission product and minor actinide destruction be designed? The conclusion is that the stated goals can possibly be realized using innovative variations of conventional LMR configurations and compositions that optimize the support ratio. These variations can also establish symbiotic relationships between plutonium fuel, minor actinides, and fission products that enhance safety characteristics of the core and provide acceptable fuel and burnup performance. Designs with these qualities are likely to have the following features:

- A modular core
- Conventional plutonium-uranium fuel with minor actinides included in some fuel assemblies
- ^{99}Tc also incorporated in the fuel mix replacing ^{238}U , and possibly in separate in-core targets and peripheral moderated target assemblies

• ^{99}Tc concentrated in other in-core regions mainly to improve the sodium void reactivity

• All ^{129}I irradiated in moderated peripheral target assemblies.

To achieve the goal destruction rate for ^{99}Tc , it may be necessary to include this isotope in minor actinide assemblies. This strategy may also be useful in regulating the power in the actinide assemblies.

The parfait core design looks like a promising means of enhancing the safety characteristics in LMR burner designs. By including a layer of ^{99}Tc in the middle of the fuel pins, sodium void reactivity can be greatly reduced, as in pancake core designs. However, the parfait design should have higher radial neutron leakage, and thus, better transmutation rates in the moderated target assemblies.

REFERENCES

1. J. A. RAWLLNS, CURE: Clean Use of Reactor Energy, WHC-EP-0268, Westinghouse Hanford Company, Richland, Washington (May 1990).
2. R. A. KARNESKY, D. P. JORDHEIM, K. D. DOBBIN, and D. W. WOOTAN, "Application of Advanced Liquid Metal Reactors to the Destruction of Radioactive Wastes," Proceeding of International Conference on Fast Reactors and Related Fuel Cycles, Kyoto, Japan (October 1991).
3. D. W. WOOTAN, J. W. DAUGHTRY, S. H. FINFROCK, J. GREENBORG, R. A. KARNESKY, J. V. NELSON, R. P. OMBERG, H. TOFFER, and A. E. WALTAR, "A Comparative Assessment of the Destruction of Selected Fission Products in Fast and Thermal Reactors," Proceeding of International Conference on Design and Safety of Advanced Nuclear Power Plants, Tokyo, Japan (October 1992).
4. K. D. DOBBIN, S. F. KESSLER, J. V. NELSON, D. W. WOOTAN, and R. P. OMBERG, "Evaluating the Efficacy of a Minor Actinide Burner," presented at this conference.
5. D. W. WOOTAN and J. V. NELSON, "Transmutation of Selected Fission Products in a Fast Reactor," presented at this conference.
6. S. F. KESSLER, "Reduction of the Sodium-Void Coefficient of Reactivity by Using a Technetium Layer," presented at this conference.
7. A. E. WALTAR and A. B. REYNOLDS, Fast Breeder Reactors, Pergamon Press, New York, New York (1981).
8. R. W. HARDIE and W. W. LITTLE, Jr., 3DB. A Three-Dimensional Diffusion Theory Burnup Code, BNWL-1264, Pacific Northwest Laboratory, Richland, Washington (March 1970).

9. R. E. MACFARLANE, D. W. MUIR, and R. M. BOICOURT, The NJOY Nuclear Data Processing System. Volume I: User's Manual, LA-9303-M, vol. 1 (ENDF-324), Los Alamos National Laboratory, Los Alamos, New Mexico (1982).
10. F. M. MANN, MIDX. A One-Dimensional Diffusion Code for Generating Effective Nuclear Cross Sections, HEDL-TME 82-12, Westinghouse Hanford Company, Richland, Washington (September 1982).
11. L. P. ABAGYAN, N. O. BAZAZYANTS, I. I. BONDARENKO, and M. N. NIKOLAEV, Group Constants for Nuclear Reactor Calculations, Consultants Bureau, New York, New York (1964).
12. J. S. BREIMEISTER (Editor), MCNP — A General Monte Carlo Code for Neutron and Photon Transport, Version 3A, LA-7396-M, Rev. 2, Los Alamos National Laboratory, Los Alamos, New Mexico (1986).
13. M. L. THOMPSON "The Modular ALMR (PRISM) Fuel Cycle," Proceedings of the American Nuclear Society Special Cluster Session — LMR: Decade of LMR Progress and Promise, Washington, D.C. (November 1990).
14. K. D. DOBBIN, D. P. JORDHEIM, J. V. NELSON, J. A. RAWLINS, and D. W. WOOTAN, "Transmutation of Light Water Reactor High-Level Waste in and Advanced Liquid Metal Reactor," Trans. Am. Nucl. Soc., 64, p. 122, (November 1991).