PHYSICS CONSIDERATIONS IN THE DESIGN OF LIQUID METAL REACTORS FOR TRANSURANIUM ELEMENT CONSUMPTION

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ABSTRACT

The management of transuranic nuclides in liquid metal reactors (LMR's) is considered based on the use of the Integral Fast Reactor (IFR) concept- Unique features of the IFR fuel cycle with respect to transuranic management are identified. These features are exploited *together with the* hard spectrum of LMR's to demonstrate the neutronic feasibility of a wide range of transuranic management options ranging from efficient breeding to pure consumption. Core physics aspects of the development of a low sodium void worth transuranic burner concept are described. Neutronics performance parameters and reactivity feedback characteristics estimated for this core concept are presented.

1. INTRODUCTION

The desire to reduce the long term radiological hazard associated with the disposal of spent nuclear fuel has motivated numerous studies in several countries (e.g. Refs. 1-6) of ways to destroy the transuranium products of fuel irradiation. Recent efforts in the U.S. at Argonne National Laboratory [6-8] have focused on evaluating the potential for accomplishing this with liquid metal reactors (LMR's), particularly metal fuel systems based on the Integral Fast Reactor (IFR) concept. These evaluations have addressed both the recycle of self-generated actinides and the consumption of externally produced inventories of transuranics, e.g. the transuranic species in spent LWR fuel. Core physics aspects of these evaluations will be discussed in this paper.

2. THE IFR CONCEPT

Key features of the IFR concept [9] include the metallic fuel form (U-Pu-Zr) and the compact and inexpensive techniques being developed for fuel reprocessing (pyrometallurgical process) and refabrication (injection -casting).- Neutronic feedback 'mechanism 'and other 'inherent physical phenomena provide for neutronic shutdown and decay heat removal in accident sequences, resulting in a high degree of passive safety. Fuel burnup levels in excess of 15 at% are being demonstrated for the U-Pu-Zr alloy in EBR-II experiments. Current designs [10] are self-sufficient with respect to fissile mass, providing for favorable fuel cycle economics, and can be modified to yield a large net excess fissile production if desired. Conversely, favorable neutron economy can be sacrificed by reduction of fertile content and core geometric "spoiling" to permit the net consumption of transuranic species instead of their net creation through breeding.

The IFR fuel cycle has a number of attractive features in connection with the management of man-made actinides. They stem from the following:

1. The main reprocessing step (electrorefining) directly provides for the separation of the bulk of the transuranics from uranium and fission products. The small portion (-1%) of transuranics that is not automatically recycled can, with techniques being developed, be stripped from the initial waste and returned to the reactor at the expense of simultaneous y recycling an increased proportion of the rare-earth fission products.

2. The higher radioactivity levels associated with the increased fission product carryover to the transuranic product stream does not significantly complicate fuel refabrication, which is done remotely because of the presence of higher actinides in the repeatedly recycled fuel and the incomplete fission product separation inherent in the basic reprocessing method.

3. The hard neutron spectrum favors destruction of transuranics by fission over creation of higher transuranics via neutron capture. Thus, the relative concentrations of minor actinides do not build up to levels at which their radioactivity becomes intolerable. The spectrum hardness also mitigates the adverse poisoning effect of the recycled portion of the rare-earth fission products.

3. RECYCLE OF' LWR SPENT FUEL

In addition to the main IFR reprocessing and waste treatment steps (i.e. those concerned with recycle of LMR discharge fuel and blanket assemblies), pyrochemical processes are also being developed and demonstrated for the purpose of extracting the transuranic species from LWR spent oxide fuel and for concentrating these transuranics in a metallic form suitable for introduction into the IFR fuel cycle. The goal of the process development is to recover at least 99.9% of the LWR discharge transuranics for use in LMR's. The processes being studied involve the decladding of the spent LWR fuel pins by suitable mechanical and chemical means, and the subsequent pyrochemical decomposition of the spent fuel in. (a) a product stream containing the transuranics and the major portion of rare-earth fission products, (b) a uranium-rich component suitable for storage and potential future use as the source of LWR or LMR fuel, and (c) waste streams that can be processed to recover residual actinides and then converted and packaged into forms acceptable for geologic disposal.

It should be noted that the elimination of fission products from the transuranic output of the above processes is not required, because this output is designed for introduction into the IFR electrorefining step, which accomplishes the requisite degree of fission product removal. The high radioactivity and low purity of the transuranic product limit both the risk of its diversion and its attractiveness for weapons applications. *On* the *other* hand, the effected separation of the bulk of the uranium is important because it helps preserve the compactness of the electrorefining process by allowing this process to deal with only about 1 to 2% of the total heavy metal in the spent fuel.

Currently, three different pyrochemical separation concepts, referred to as the "salt transport", "magnesium extraction", and "zinc-magnesium" processes, are being investigated with the objective of identifying and further developing the most promising concept-

4. TRANSURANIC MANAGEMENT IN SELF-SUFFICIENT LMR's

In a self-sufficient LMR, transuranic losses by fission are compensated by transuranic (Pu-239) breeding, allowing for sustained power production with only fertile material (e.g. depleted U) supplied as makeup. Table 1 compares the transuranium isotopic mix of discharged fuel from a LWR (once-through) and a 1200 MWt LMR (equilibrium/recycle, based on the IFR concept). The relative concentrations of the vast majority of minor acti nides are substantial! y smaller in the LMR discharge,

while the Pu-239 proportion therein is greater. Thus, a self-sufficient LMR using the recycled transuranics from LWR discharge as its startup fissile source would, in addition to producing power over its operating life, result in an equilibrium fuel composition with reduced long-term radiological toxicity owing to the reduced proportion of higher actinides. Of particular importance is the reduced fraction of Pu-241, whose decay product (Np-237) dominates the long-term hazard. The energy production and long-term waste radiotoxicity reduction benefits are realized at the costs associated with the reprocessing of LWR fuel and the increased amount of fission product waste.

5. TRANSURANIC CONSUMPTION IN PURE BURNER CONCEPTS

Characteristics of LMR concepts designed for net consumption of pre-existing transuranic inventories have also been investigated. Taken to an extreme, no fertile U-238 is utilized in these concepts, and only transuranics are used for the startup inventory and as the source of makeup material added to the actinides recovered from self-discharged fuel. The neutronic feasibility of such "pure burner" concepts, which maximize the transuranic consumption per unit of energy production, has been verified in 1200 MWt LMR's - both for the case in which all transuranic species in the LWR discharge are used as fuel and for the case in which only the minor actinide species (no Pu) are used. Illustrative design and performance characteristics for both cases are summarized in Table IL It should be emphasized that the the fuel compositions being employed in such cases are quite exotic in containing essentially no uranium, and that the assembly/core configurations using such compositions (and constrained by a beginning of cycle excess reactivity requirement) are nonconventional. Thus while these concepts are feasible neutronically, the establishment of their overall viability requires extensive technological effort% a discussion of which is beyond the scope of this paper.

The consumption of transuranics in LWR's is considerably less attractive. Not only does the thermal spectrum favor the buildup (in proportion) of the higher actinides, but it also renders most of the minor actinide species as poisons whose accommodation requires greater fissile enrichment a thermal system fueled entirely with the LWR-discharge isotopic mix of minor actinides is not feasible neutronically based on the fundamental requirement of criticality. Moreover, the technological difficulties of adapting current aqueous reprocessing methods to impositions rich in higher actinides are likely to be severe.

6. CONCEPTUAL DESIGN FOR TRANSURANIC CONSUMPTION

The focus of recent conceptual design efforts at ANL [7] has been the development of core concepts for net transuranic consumption whose design parameters, unlike those of the pure burner concepts, remain within the bounds of the in-place development program. For example, limits on transuranic enrichment (28 wt% in heavy metal), linear power (50 kW/m), discharge bumup (15 at%), and peak fast fluence (3.5×10^{2} n.cm^{-2}) were observed. A key additional objective of the core development was to achieve a low (near-zero) sodium void worth. Finally, the core concepts were developed so that net fissile breeding, if desired, could be readily achieved as an alternative to net transuranic consumption. Such flexibility for increased breeding was achieved by use of external blankets to minimize the effecton reactor design and to mitigate the inevitable increase in sodium void worth that would occur with enhanced internal conversion.

A 1575 MWt (600 MWe) design concept satisfying these objectives and constraints was developed [7]. Key features of this concept, in its transuranic consumption mode, are (a) elimination of internal and external blankets to minimize transuranic production by U-238 capture, (b) adoption of a pancaked core shape to reduce breeding and void worth, and (c) incorporation of a non-fueled

central region in an annular core configuration. Detailed evaluations of this core configuration were carried out based on the use of LWR-discharge transuranium isotopics as the reactive fuel feed Component limited analyses of this core with its equilibrium (infinite-recycle) fuel composition have also been performed but are not described here in detail. The use of a flat, annular core shape enabled the achievement of a low void worth ($0.16 for EOC voiding of the entire core and upper plenum region) while simultaneously satisfying the assumed 28% transuranic enrichment limit. In particular, the annular geometry mitigates the central power peaking tendency without recourse to enrichment zoning, thereby enabling the enrichment and linear power limits to be satisfied concurrently with meeting the criteria of transuranic consumption and near-zero void worth.

The breeding ratio of 0.53 permits the net consumption of 234 kg of LWR transuranics on an annual basis. The fissile Pu component of this net loss (218 kg/y) would, with repeated recycle, decrease somewhat as the nuclides that are less likely to fission increase in proportion. Thus some changes in core performance would be observed with repeated recycle as the transuranic isotopic mix gradually shifts to an equilibrium recycle distribution characteristic of the care geometry and fuel cycle parameters. Preliminary calculations suggest that the coolant void worth of the equilibrium core is nearly $1.0 greater.

Additional performance results are summarized in Tables III and IV. Of particular note are the burnup reactivity loss of 4.2% Δk (5.7% Δk without midcycle replacement of central absorber assemblies), which is significantly greater than the nominally zero reactivity swing achievable in fissile self-sufficient designs. The small Doppler coefficient can be attributed to the hard spectrum and the low U-238 Concentration contributions to the Doppler effect by the transuranics are minimal because of their presence as dilute species in the reprocessed LWR discharge composition. Finally, the flat core shape results in the radial expansion reactivity coefficient being significantly more negative, and the axial expansion coefficient being less negative, than in conventional designs. For the same reason, and because of the higher control rod worth dictated by the greater burnup reactivity loss, the control rod driveline expansion coefficient is much more negative than for conventional cores. Dynamics analyses [11] utilizing the computed feedback parameters have demonstrated significant passive safety margins for unprotected loss-of-flow and reactivity insertion accidents.

7. SUMMARY

The IFR fuel cycle has a number of attractive features for the management of transuranics. These features, when successfully demonstrated, can be exploited along with the hard neutron spectrum characteristic of LMR's to achieve substantial flexibility in transuranic management options, ranging from efficient breeding to destruction in burner concepts.

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Table I. COMPARISON OF LWR AND LMR TRANSURANIC ISOTOPICS.

	LWR Discharge	LWR (3.2 *V* Decay)	1200 MWt LMR Discharge	1200 Mwt LMR (2.0 y Decay)	Ratio of LWR (3.2 y Decay) to LMR (2.0 v Decay)
Fraction of Transuranic Mass					
Np-237	5.35-2	5.40-2	6.16-3	6.19-3	8.7
Pu-236	2.41-7	1.12-7	6.12-8	4.08-8	2.7
Pu-238	9.08-3	1.01-2	6.27-3	6.19-3	1.6
Pu-239	0.503	0.508	0.747	0.747	0.68
Pu-240	0.201	0.199	0.200	0.200	1.0
Pu-241	0.157	0.134	2.13-2	1.96-2	6.8
Pu-242	3.93-2	3.88-2	8.49-3	8.49-3	4.6
Am-241	3.44-3	2.51-2	6.94-3	8.63-3	2.9
Am-242m	1.14-4	1.114	5.11-4	5.07-4	0.22
Am-243	2.51-2	2.48-2	1.74-3	1.74-3	14.0
Cm-242	1.31-3	9.73-6	3.56-4	2.36-5	0.41
Cm-243.	8.55-5	7.86-5	1.93-5	1.85-5	4.2
Cm-244	6.32-3	5.52-3	7.96-4	7.444	7.4
Cm-245	5.15-4 , ,	5.08-4	1.63-4	1.63-4	3.1
Cm-246	6.42-5	6.31-5	4.72-5	4.72-5	1.3
MA/Fissile Pu	0.137	'O. 172	0.022	0.024	7.2
MA/Total Pu	0.099	0.124	0.017	0.018	6.9
Np-237/MA	0.591	0.490	0.368	0.343	1.4
Am-241/MA	0.038	0.228	0.415	0.478	0.48
Am-243/MA	0.278	0.225	0.104	0.096	2.3
Np chain	0.214	0.213	0.034	0.034	6.3

MA = sum of minor actinides
Fissile Pu = Pu-239 + Pu-241
Np chain = Np-237 + Am-241 + Pu-241

Table II. COMPARISON OF EQUILIBRIUM CYCLE CHARACTERISTICS FOR 1200 MWt ACTINIDE RECYCLE CONCEPTS

	Fissile Self-Sufficient LMR	Pu and MA Pure Burner	MA Pure Burner
No. of Batches	4	1	1
Cycle Length, days	365	254	453
Capacity Factor, %	89	80	80
Driver Average Bumup, MWd/kg	104	111	111
Driver Peak Burnup, MWd/kg	140	150	158
Driver Peak Fast Fluence, 10^{23}n/cm^2	3.51	1.30	1.76
Driver Peak Linear Power, kW/ft	13.0	13.6	14.5
EOC-BOC Ak Swing, %	0.27	-12.1	-4.33
Driver HM Loading, kg/y	2770	4040	3040
%U	77.2	0	0
%Pu	22.4	64.6	37.5
%MA	0.4	35.4	62.5
Blanket HM Loading, kg/y	3910		
%U	100		
%Pu	0		
%MA	0		
Makeup Feed, kg/y	466	352	346
%U	100	0	0
%Pu	0	87.6	0
%MA	0	12.4	100

HM = Heavy Metal
MA = Minor Actinides

Table 111. EQUILIBRIUM-CYCLE PERFORMANCE PARAMETERS

Breeding Ratio	0.533
EOC-BOC Reactivity Swing, %ΔK	-4.17
Average Discharge Burnup	
MWd/kg	82.6
Atom %	8.8
Peak Discharge Bumup	
MWd/kg	118.6
Atom %	12.6
Peak Linear Power, W/cm	
BOC	495
EOC	462
Power Peaking Factor	
BOC	1.64
EOC	1.53
Peak Flux, $10^{15}cm^{-2}s^{-1}$	
BOC	4.84
EOC	5.02
Peak Fast Flux, $10^{15}cm^{-2}s^{-1}$	
BOC	3.68
EOC	3.77
Peak Fast Fluence, $10^{23}cm^{-2}$	2.96
Mass Flow, kg/y	
Heavy Metal	5,790
Transuranics	1,492
Fissile Pu	958
Net Loss, kg/y	
Heavy Metal	508
Transuranics	234
Fissile Pu	218

Table IV. LOW VOID WORTH ACTINIDE BURNER REACTIVITY COEFFICIENTS

	BOC[a]	EOC
Sodium Void Worth, $		
Core	1.78	2.85
Plenum	-2.20	-2.70
Total	-0.43	0.16
Doppler Coefficient, - 10^{-3}Tdk/dT		
Flooded Core		
Fuel	0.73	0.88
Structure[b]	0.30	0.35
Voided Core		
Fuel	0.36	0.46
Structure[b]	0.21	0.25
Axial Expansion Coefficient, $/cm		
Fuel	-1.50	-1.19
Fuel and Structure[b]	-1.37	-1.01
Radial Expansion Coefficient, $/cm	-1.43	-1.48
Control Rod Driveline Expansion Coefficient, $/cm	-0.778	-0.414
Effective Delayed Neutron Fraction	3.50E-3	3.47E-3
Prompt Neutron Lifetime, s . .	2.17E-7	2.47E-7

[a] BOC values are calculated for critical configuration, with primary rods inserted 24 cm.

[b] Values reflect perturbation of total structure; clad effects can be estimated as 63% (volume fraction of clad in the total structure) of the total structure effect.