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Inventory and Relative Biological Hazard of LMFBF Core Materials

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M. L. Tobias

ABSTRACT

Information on the composition of an LMFBR core by element has been collected on the basis of published design data and ORIGEN calculations. The mass quantities of structural materials, actinides, and fission products are in the ratio of 1:1.4:0.1 with actinides, particularly plutonium and neptunium, representing the major inhalation and ingestion hazard. Some of the fission products are present in quantities large enough to suggest that they warrant specific attention in source term analysis.

1. INTRODUCTION

This report discusses information collected for the Aerosol Release and Transport (ART) Program¹ at Oak Ridge National Laboratory. It is intended to be of use in estimating the potential composition of the material that may be released as a result of a hypothetical core-disruptive accident (HCDA) in a liquid-metal-cooled fast breeder reactor (LMFBR).

Two types of information are presented. First, the nominal inventories of fuel, sodium, and structural materials for various LMFBR concepts are given. Next, results obtained from existing ORIGEN² code calculations of isotope inventories for the case of a commercial-size LMFBR during full-power operation are listed. Gram quantities by element of fission products, actinides, and structural materials are tabulated together with comments on relative biological hazards. Particular objectives of the listings are (1) the identification of those constituents which have a high biological hazard, and yet make up only a small percentage by weight of a possible effluent, and (2) the identification of fission product elements which are generated in mass quantities sufficient to have a possible effect on aerosol behavior.

2. TYPICAL NORMAL INVENTORIES OF CORE MATERIALS FOR VARIOUS LMFBRs

Table 1, taken from various sources,³⁻⁶ gives the power, core volume fractions, and fissionable inventory for experimental fast reactors, prototype reactors, and commercial-size fast reactors.

More recent designs have been prepared as part of the Prototype Large Breeder Reactor (PLBR) studies,⁷ details of which are not yet generally available. Advanced designs⁸ involving concepts such as spoiled geometry and heterogeneous ("parfait") cores would appear to require plutonium enrichments of about 16%. Other design changes have been made, but at this writing it is not clear what bearing they may have on aerosol source term analysis.

The average and standard deviations of all the values of volume fractions given in Table 1 are: fuel, 0.37 ± 0.06 ; sodium, 0.37 ± 0.09 ; and other, 0.26 ± 0.03 . Table 1 shows that fuel inventory is a function of reactor power, with reactors of the same size having more or less the same amount of fuel. Plutonium/uranium ratios in the commercial reactors vary in the table from 0.14 to 0.25. These figures indicate that it is not difficult to typify LMFBRs from the standpoint of core volume fractions, fuel inventory, or plutonium fraction.

3. ELEMENT INVENTORIES FROM ORIGEN CALCULATIONS

The assessment of the aerosol source term requires knowledge of the types, gram quantities, and relative biological hazards of the materials which might be released in an HCDA.

ORIGEN code calculations for the Atomic International follow-on LMFBR are available which are useful in such an evaluation.^{2,5,9,10} As will be seen, the relative biological hazards of the nuclides are not necessarily in the same order as their gram quantities.

The bases of the calculations have been reconstructed as follows. Reference 11 states that the core inventory is 19.09 metric tons (1 metric ton = 1000 kg) of heavy metal. The total power is 2400 MW(t) and the core power fraction is given as 0.917. Using an 80% power

Table 1. Fuel and volume fraction data
for various fast reactor types

Reactor	Country	Power [MW(t)]	Core volume fraction			Core loading (10 ³ kg)	
			Fuel	Sodium	Other	UO ₂	PuO ₂
Experimental fast reactors ³							
Rapsodie F	France	40	0.42	0.40	0.18	0.106	0.045
PEC	Italy	118	0.35	0.37	0.28	0.76	0.31
JOYO	Japan	50	0.36	0.40	0.24	0.7	0.15
FFTF	USA	400	0.35	0.41	0.24	2.13	0.723
EBR-II	USA	62.5	0.32	0.19	0.49	0.24	0.01
Fermi	USA	200	0.27	0.46	0.27	1.88	
BOR-60	USSR	60	0.40	0.28	0.32	0.24	
Prototype reactors ^{3,6}							
Phenix	France	568	0.36	0.36	0.28	3.8	0.8
Phenix 450	France	1050	0.36	0.36	0.28	8.0	1.8
SNR-300	Germany	762	0.31	0.50	0.19	4.2	1.65
Monju	Japan	714	0.34	0.40	0.26	4.4	1.5
PFR	UK	612	0.36	0.42	0.22	3.1	0.9
CRBR	USA	975	0.33	0.42	0.25 ^a	5.3	1.85
BN-350	USSR	1100	0.46	0.32	0.22	7.3	
BN-600	USSR	1470	0.45	0.33	0.22	8.5	
Commercial reactors [power in MW(e)] ³⁻⁵							
Superphenix	France	1200	0.34	0.34	0.32	30.6	6.3
CFR	UK	1250				16	4
B&W ^b	USA	1000	0.44	0.17	0.39	^c	3.8
CE ^b	USA	1000	0.39	0.40 ^d	0.21 ^d	^e	^c
AI ^b	USA	1000	0.33	0.44	0.24	16.6 ^f	2.5 ^g
CE ^b	USA	1000	0.47	0.38	0.15	15.1 ^g	2.1 ^g
Westinghouse ^b	USA	1000	0.29	0.52	0.19		

^aStructure, 0.234; gap, 0.019.

^bSo-called "follow-on" study reactor designs proposed about 1968.

^cAverage fissile enrichment of feed, 11.6 wt % ²³⁹Pu and ²⁴¹Pu.

^d0.0461 BeO, 0.0155 shim poison, 0.0213 safety poison. Sodium occupies shim position site unless tantalum rods are inserted.

^eSpecific inventory, including plutonium in blankets and out of reactors, 3.6 kg of fissile material/MW(e); seed region enrichment 10 to 12% ²³⁹Pu and ²⁴¹Pu.

^fEstimate from data in Ref. 5.

^gEstimate from data in Ref. 5.

factor, we find

Core power/metric ton heavy metal

$$= (2400)(0.917)(0.8)/19.09 = 92.23 .$$

(The actual input number was 92.17 MW.)

No attempt was made to mock up the three-year cycle postulated in the follow-on study; not only is it awkward to do so in the version of ORIGEN used, but the simplification of using an average burnup time of two years is also quite justifiable. (At the end of an equilibrium cycle, one-third of the core will have been irradiated for one year, one-third for two years, and one-third for three years, resulting in an average exposure of two years.) The burnup per metric ton of heavy metal is therefore

$$(92.17 \text{ MW/metric ton})(2 \text{ years})(365 \text{ days/year})$$

$$= 67,284 \text{ Mwd/metric ton} .$$

The actual input number used was 67,099.* The principal characteristics of the Atomic International follow-on reactor are outlined in Table 2.

Tables 3 to 5 list the most significant elements in three classes[†] from the standpoints of mass inventory and relative inhalation and ingestion hazard. (The melting and boiling points of the elements have been included as incidental information; obviously, in many cases the nuclide will be present as a compound.) Assuming that all the elements are released in an accident, the masses and hazards are in the ratios listed in Table 6.

These tables permit the ordering of concerns with particular elements from the standpoint of aerosol source generation. Thus, the actinides appear far more significant than fission products, which in turn are much more hazardous than the structural materials. The most

*The calculation was performed in 1972, making it impractical to determine the reasons for these minor differences.

[†]The ORIGEN code deals with reactor materials in three classes: (1) structural materials (e.g., coolant and cladding materials); (2) actinides; and (3) fission products.

Table 2. "Task III" Atomics International final
reference design reactor characteristics⁶

Thermal power, MW(t)	2,400		
Gross electrical power, MW(t)	1,040		
Refueling cycle time, years	1		
Reactor power fractions			
Core	0.917		
Axial blanket	0.031		
Radial blanket	0.052		
Volume fractions	<u>Fuel</u>	<u>Sodium</u>	<u>Metal</u>
Core and axial blanket	0.439	0.326	0.235
Inner radial blanket	0.556	0.281	0.163
Outer radial blanket	0.584	0.269	0.147
Core fissile enrichments, at. %			
Inner zone	10.5		
Outer zone	13.1		
Fissile masses (Pu + ²³⁵ U), kg			
Core	2,191		
Axial blanket	176		
Radial blanket	377		
Initial loading (Pu + U), kg			
Core	19,090		
Axial blanket	13,132		
Radial blanket	15,688		

hazardous nuclides among the actinides appear to be plutonium and neptunium followed by iodine among the fission products. However, this ordering is only an initial guide, for it assumes that all the elements are released equally, an unlikely assumption. Further analysis will certainly produce changes in their relative importance.

Table 3. Inventory of structural materials and their
postirradiation hazards

Basis: 1000 kg (1 metric ton) of U + Pu as charged

Power = 92.17 MW; burnup = 67,099 MWd

Element	Melting point (°C)	Boiling point (°C)	Loading (g)		Hazard	
			Initial	Discharge	Inhalation ^a	Ingestion ^a
H	-259	-253	0	5.53		
C	3550	4827	4.21E02	4.38E02		
O	-218	-183	1.34E05	1.34E05		
Na	98	883	1.31E03	1.31E03	2.E3	0.01
Si	1410	2355	3.94E03	3.94E03	2.E4	1.E3
P	44	280	2.10E02	2.10E02	2.E3	0.01
S	113	445	1.58E02	1.57E02		
Cr	1857	2672	8.94E04	8.94E04	1.E3	2.E3
Mn	1244	1962	1.05E04	1.05E04	0.33	0.30
Fe	1535	2750	3.42E05	3.43E05	0.02	0.02
Co	1495	2870	5.27E02	5.56E02	0.63	0.52
Ni	1453	2732	6.57E04	6.56E04	4.E4	2.2E3
Nb	2468	4742	0.0	2.90E-2	4.E5	5.E5
Mo	2617	4612	1.31E04	1.31E04	0.02	0.13
Tc	2172	4877	0.0	3.20E01	2.E4	2.E3
Totals			6.62E05	6.62E05	6.56E14	1.74E10

^aThe totals in the last lines of the inhalation and ingestion columns are the volumes in cubic meters of air and water, respectively, required to dilute the radioactivity in all the nuclides the code considers to levels given in Table II of Title 10 of the Code of Federal Regulations, Part 20. The figures for individual elements are fractions of this quantity and (rounded off) sum to 1.0.

Table 4. Inventory of accinides and their postirradiation hazards

Basis: see Table 3

Element	Melting point (°C)	Boiling point (°C)	Loading (g)		Hazard	
			Initial	Discharge	Inhalation ^a	Ingestion ^a
U	2865 ^b	3487 ^b	8.26E05	7.60E05	2.1E-7	9.6E-5
Np	640	3902	0.0	4.56E02	2.5E-3	0.953
Pu	2390 ^c	~3200 ^d	1.74E05	1.66E05	0.94	0.029
Am	994	2607	0.0	1.70E03	0.02	0.006
Cm	1340		0.0	7.78E01	0.04	0.012
Totals			1.00E06	9.28E05	1.24E18	6.40E11

^aThe totals in the last lines of the inhalation and ingestion columns are the volumes in cubic meters of air and water, respectively, required to dilute the radioactivity in all the nuclides the code considers to levels given in Table II of Title 10 of the Code of Federal Regulations, Part 20. The figures of individual elements are fractions of this quantity and (rounded off) sum to 1.0.

^bAs UO₂.

^cAs PuO₂.

^dAs mixed oxide with UO₂.

Table 5. Inventory of fission products and their postirradiation hazards

Basis: 1000 kg (1 metric ton) of U + Pu as charged

Element	Melting point (°C)	Boiling point (°C)	Loading (g)	Hazard	
			Discharge	Inhalation ^a	Ingestion ^a
Ru	2310	3900	6.39E3	0.147	0.002
I	114	184	6.02E2	0.441	0.828
Ce	798	3257	5.32E3	0.130	0.020
Sr	769	1384	1.06E3	0.086	0.049
Y	1523	3337	5.75E2	0.032	0.012
Zr	1852	4377	5.80E3	0.056	0.015
Nb	2468	4742	1.06E2	0.014	0.002
Ba	725	1640	2.76E3	0.041	0.011
La	920	3454	2.26E3	0.010	0.011
Kr	-157	-152	5.33E2	0.001	
Mo	2617	4612	6.22E3	0.007	0.006
Rh	1966	3727	1.68E3	0.002	0.002
Pd	1552	3140	4.36E3	0.001	0.001
Te	450	990	1.17E3	0.010	0.009
Cs	28	678	7.54E3	0.006	0.001
Pr	931	3212	1.78E3	0.006	0.004
Nd	1010	3127	6.46E3	0.002	0.002
Pm	1080	2460	6.79E2	0.005	0.002
Xe	-112	-107	8.19E3	0.001	
Eu	822	1597	3.01E2	0.001	
Sb	631	1750	1.10E2	0.001	
Totals			6.94E4	9.92E16	1.80E13

^aThe totals in the last lines of the inhalation and ingestion columns are the volumes in cubic meters of air and water, respectively, required to dilute the radioactivity in all the nuclides the code considers to levels given in Table II of Title 10 of the Code of Federal Regulations, Part 20. The figures for individual elements are fractions of this quantity and (rounded off) sum to 1.0.

Table 6. Mass and hazard ratios of the three classes of nuclides in the Atomic International follow-on reactor core^a

Nuclide class	Mass	Inhalation hazard	Ingestion hazard
Structural	1.0	1.0	1.0
Actinides	1.4	1890	1034
Fission products	0.1	151	37

^aInventories and hazards by isotope are also available but have been omitted here for brevity.

CONCLUSIONS

The gram quantities and constituents of material that might be ejected from the core of an LMFBR following a core-disruptive accident have been assessed using published information on reactor designs and ORIGEN calculations. The ratio of mass quantities of structural materials, actinides, and fission products, is 1:1.4:0.1. The inhalation and ingestion hazards are mainly from the actinides, led by plutonium and neptunium. Gram quantities of fission products are not insignificant and should not be neglected in examination of aerosol behavior.

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