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for Waste Handling and Disposal

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214

QUALITATIVE COMPARISONS OF FUSION REACTOR MATERIALS FOR WASTE HANDLING AND DISPOSAL*

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ABSTRACT

The activation of five structural materials and seven coolant/breeder/multiplier materials in a common reference neutron environment was calculated with the FORIG activation code. The reference environment was the neutron flux and spectrum at the first wall of the mirror advanced reactor study (MARS) reactor. Qualitative comparison of these activated materials were made with respect to worker protection requirements for gamma radiation in handling the materials and with respect to their classifications for near-surface disposal of radioactive waste.

INTRODUCTION

The activation of the materials in fusion reactors is the key to remote handling requirements for waste, to processing and disposal methods for waste, and to accident severity in waste management operations.¹⁻⁴ In order to realize the desirable environmental potentials of fusion power systems, there are at least three major goals for waste management. These goals are (a) near-surface burial; (b) on-site disposal of the fusion reactor; (c) acceptable radiation doses at the least cost during and after waste management operations.

Although meeting the above goals will depend finally on the specific reactor designs and on the specific materials chosen for the blankets, early design guidance can be provided by qualitative comparisons of blanket materials under a standard set of conditions. In Ref. 5, the conclusion states that "...all fusion reactors, regardless of geometric shape and first wall radius, having unprotected first walls made of similar material and operated for the same amount of time, will have roughly equal total radioactivities,

adjusted only in proportion to the product of their thermal power and their first wall thickness." This conclusion allows the use of a standard fusion-reactor reference configuration to investigate qualitative comparisons of materials with respect to properties dependent on radioactivity from activation in the first wall zone. Extensions of results to specific reactor designs is then only a matter of scaling based on thermal power and first wall thickness for blanket/first wall materials.

For this study, the reference neutron environment (flux and spectrum) is the first wall zone of MARS⁶ with a 4.1 MW/m² wall loading, a first wall thickness of 0.003 m, and a two year exposure time. The neutron flux and spectrum were the same for all the materials. They were calculated with TART specifically for the first wall of the MARS blanket. This blanket consists of HT-9 for structural material and 17Li83Pb for the coolant/breeder material. The spectra for other combinations of blanket materials would be different, but the integrated values of cross sections would be roughly the same for all combinations. Thus, qualitative comparisons of materials, as opposed to evaluations of detailed design differences among blankets, will be roughly independent of materials combinations in the first wall where 14 MeV neutrons dominate. The reference compositions of materials for this study are shown in Table I.

The comparisons of these materials are based on the Remote Maintenance Rating (RMR) and the Waste Disposal Rating (WDR).^{7,8} The RMR is an index that gives a measure of the gamma-ray contact dose at the surface of a thick infinite slab of activated material. The slab is considered thick when the thickness dimension is several mean free paths of the gamma rays. As described in Ref. 7, the RMR can be converted to contact doses for small spheres or thin sheets by multiplying the RMR by a factor containing the ratio of the radius of the sphere or thickness of the

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sheet to the mean free path of the gamma rays in the materials. However, such conversions will not change the relative ordering of qualitative comparison of materials. The units of the RMR are mrem/hr. If the RMR is less than 2.5 mrem/hr, workers can be in contact with the material without any special protection for gamma radiation. However, because the RMR is defined here only for gamma radiation, there is still the possibility that protection may be needed for inhalation and ingestion of alpha and beta emitters. This is in accordance with the NRC 10CFR20 regulations on worker safety.⁹ As the RMR becomes greater and greater than 2.5 mrem/hr, more and more protection against contact and exposure is needed in the form of gamma shielding and/or use of remote handling equipment.

The WDR is an index that gives a measure for classifying wastes in accordance with the NRC 10CFR61 regulation on licensing requirements for land disposal of radioactive wastes.¹⁰ The WDR is defined as follows:

$$WDR = \sum_i C(i)/L(i),$$

where $C(i)$ is the concentration of the i th radionuclide in the waste material (Ci/m^3) and $L(i)$ is the corresponding allowable concentration limit for a given waste class as provided in 10CFR61. The intent of 10CFR61 is to protect inadvertent intruders at disposal sites from radiation doses greater than 500 mrem/year.^{10,11} The radionuclides explicitly covered by limits in 10CFR61 (shown in Table II-A) are appropriate for waste classification of fission products, fission reactor activation products, and transuranics. However, in fusion there are other nuclides (especially long-lived gamma emitters) that must be considered explicitly to ensure limiting doses to inadvertent intruders to 500 mrem/year or less. Additional nuclides of this type are shown in Table II-B.¹²

For alloys and mixtures of materials, near-surface land disposal can be used when $WDR < 1$. If $WDR > 1$, the materials are not suitable for near-surface disposal without some kind of processing to reduce the WDR to 1 or less. This processing may be simple dilution with inert materials in the waste package so that the WDR of the radionuclides averaged over the total volume of the package is less than 1.

The RMR and WDR are calculated from activation product concentrations obtained with the Lawrence Livermore National Laboratory (LLNL) computer codes TARTNP,¹³ ORLIB,¹⁴ ACTL,¹⁵ and FORIG.¹⁶ The data inputs are the reference neutron environment

(MARS) and the materials compositions listed in Table I.

QUALITATIVE COMPARISONS OF REMOTE MAINTENANCE RATINGS

The relative rankings by RMR for blanket materials considered in this study are shown in Table III for various times after removal from the neutron flux. Inspection of Table III reveals that there are spreads among the materials of four to six orders of magnitude in the RMR at a given time after shutdown with the greatest spreads at the later times. Also, the relative rankings of materials change from one time to another. Depending upon the material, the RMR drops four to nine orders of magnitude from the 1-hour to 100-year time for that material. Only the RMR for Li_2O , FLIBE, and lithium drop enough for the materials to be handled without worker protection at 100 years. However, even these materials require worker protection at 10 years or less for large bodies of the materials or for close contact with components made with the materials. The degree of protection needed will depend on specific dimensions of components as well as on the material, but the RMR can be helpful in assessing the consequences of materials selection early in the design phase of a reactor and in planning waste disposal.

The major radionuclides contributing to the RMR for the materials at various times are shown in Table IV. Many of these are activation products of elements that are impurities in the materials. Thus, it is important to consider carefully the purity of materials to control costs of remote handling and shielding. The task of detailed identification of the sources (essential or impurity elements) is beyond the scope of this study. In general, Co60 is an undesirable component in all of the materials at 10 years. It originates mainly from cobalt, nickel, and copper. Therefore, if these elements are impurities, they should be carefully controlled if significant materials handling is expected at 10 years (say for waste processing). For 1-hour and 1-day times, Na22 and Na24 are prominent nuclides in the breeding/multiplier materials and must be carefully considered if they come from impurities like elemental sodium or magnesium. (Nitrate salt has a high sodium content as an essential element.) Both breeding/multiplier and structural materials contain Mn54 and Mn56 as major contributors to RMR at early times. In both cases, these two nuclides come from natural elemental manganese and iron.

At 100 years, the prominent radionuclides are Al26, Co60, Nb94, Ag108m, Bi207, and Bi208. Except for LiAlO₂, which has aluminum

as an essential element, the Al26 content of the materials can perhaps be controlled to reduce RMR. The Nb94 comes from molybdenum and niobium and can be controlled only to the extent that it is possible to control these two elements. The Ag108m comes from elemental silver, which appears to be an impurity and as such should be reduced as much as possible consistent with costs for shielding and handling. The bismuth nuclides come from bismuth and lead. Thus, Bi207 and Bi208 will always be generated in the LiPb breeding/multiplier; control of bismuth as an impurity can reduce but not eliminate the bismuth gamma emitters.

QUALITATIVE COMPARISONS OF WASTE DISPOSAL RATINGS

The WDR at 10 years after shutdown for the materials of interest to the blanket comparison and selection study (BCSS) are displayed in Table V. The ratings for individual nuclides are shown as well as for subtotals of nuclides grouped according to Table II. Note the contributions of the supplemental nuclides in Table II-B to the total WDR for LiAlO₂, LiPb, TENELON, and ModHT9. The supplemental nuclides increase the WDR to values greater than 1 for each of these materials. Thus, they are significant factors in determining suitability for near-surface disposal of LiAlO₂, LiPb, TENELON, and ModHT9.

Upon consideration of the WDR in Table V and the 10CFR61 regulations, the suitability of materials for near-surface disposal can be summarized as shown in Table VI. Column 1 indicates that beryllium, Li₂O, lithium, and FLIBE are well within the limits for near-surface disposal. However, at 10 years or less after removal from a reactor, all of these materials in intact packages would require worker protection during handling. At 100 years, the last three would not require protection, but beryllium would still be too hazardous to handle without protection even though it can be disposed near-surface burial. All of the materials in column 1 are breeding/multiplier materials.

The alloys in column 2 are structural materials whose WDR are greater than 1, but it is expected that, when they are packaged with other inert and less active materials from a reactor, the WDR integrated over the volume of the waste package will be less than 1 and near-surface disposal is permissible. Again, however, worker protection will be required even at 100 years because of the high RMR of intact waste packages.

The materials in column 3 have values for WDR too high to be reduced by simple dilution. Therefore, they must be processed in some way to remove the highly active components for near-surface burial of the bulk of the materials. The high activity components would then need to be disposed of at high level waste as suggested in footnote (c) of Table VI.

The major nuclides preventing near-surface disposal of materials in column 3 are listed in Table VII. The limiting source elements for LiAlO₂ and nitrate salt are essential to the composition of the materials and cannot be reduced or eliminated. The WDR for HT9, PCA, and LiPb can be reduced by reducing the amounts of the indicated source nuclides.

Activation and subsequent decay chains in lead and in lead with bismuth impurities produce several alpha-emitting nuclides (see Table VIII). These may cause safety problems in accident and materials handling scenarios for inhalation and ingestion. For waste management under the current 10CFR61, none of these nuclides would be limited for near-surface burial because only transuranic nuclides are explicitly covered. If 10CFR61 is modified to cover any alpha-emitting nuclide with an effective half life greater than five years, then Pb210, Bi210m, and Po210 may have limits. However, if the limit is 100 nCi/g (the current level for transuranic alpha emitters), there would still be no problem for waste disposal because the concentrations predicted for MARS are much less than 100 nCi/g for all the alpha emitters at 10 years after shutdown as shown in Table VIII.

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Table 1-A. Elemental compositions.

Z-#	Element	Structural materials				
		PCA (wt%)	V15Cr5Ti (wt%)	HT-9 (wt%)	Modified HT-9 (wt%)	TENELON Fe 1518 (wt%)
3	Li					
4	Be					
5	B	0.005		0.01	0.001	0.001
6	C	0.005	0.005	0.2	0.15	0.15
7	N	0.01	0.01	0.05	0.001	0.005
8	O		0.01	0.01	0.007	0.007
9	F					
11	Na					
12	Mg					
13	Al	0.03	0.02	0.01	0.008	0.008
14	Si	0.5	0.03	0.15	0.2	0.2
15	P	0.01	0.003	0.02	0.013	0.013
16	S	0.005	0.001	0.02	0.004	0.004
17	Cl		0.0001			
19	K	0.0003	0.00001	0.0003	0.0003	0.0003
20	Ca					
22	Ti	0.30	5.0	0.09	0.1	0.003
23	V	0.10	79.80	0.3	0.3	0.002
24	Cr	14.0	15.0	12.0	11.0	15.0
25	Mn	2.0		0.55	0.53	15.0
26	Fe	64.88	0.004	85.0	Balance	Balance
27	Co	0.03		0.02	0.005	0.005
28	Ni	16.0	0.0004	0.5	0.006	0.006
29	Cu	0.02	0.0002	0.09	0.003	0.003
30	Zn					
33	As	0.02	0.0002			
38	Sr					
40	Zr	0.005		0.001	0.001	0.001
41	Nb	0.03	0.0004	0.11	0.00011	0.00011
42	Mo	2.0	0.001	1.0	0.00027	0.00027
47	Ag	0.0001		0.0001	0.00009	0.00009
48	Cd	0.0002		0.0001	0.0001	0.0001
50	Sn	0.005		0.003	0.003	0.003
51	Sb	0.001		0.001	0.0005	0.0005
56	Ba	0.001		0.001	0.0002	0.0002
65	Tb	0.001		0.0005	0.0002	0.0002
73	Ta	0.01	0.001	0.001	0.0004	0.0004
74	W	0.05	0.0002	0.5	2.50	0.01
77	Ir	0.001		0.0005	0.0002	0.0002
82	Pb	0.001		0.001	0.0005	0.0005
83	Bi	0.001		0.001	0.0002	0.0002

Table I-B. Elemental compositions.

		Breeding/multiplier materials						
Z-#	Element	Li (wt%)	FLIBE (wt%)	17Li83Pb (wt%)	Nitrate salt (wt%)	Li ₂ O (wt%)	LiAlO ₂ (wt%)	Be (wt%)
1	Li	99.9	8.60	0.68			10.3	0.0003
4	Be		13.0	0.001			0.001	99.0
5	B			0.001			0.001	0.0002
6	C	0.001						0.10
7	N	0.005		0.001	15.0			0.03
8	O			0.026	51.6		48.6	0.7
9	F		78.4					
11	Na	0.03		0.01	12.3	0.005	0.005	
12	Mg			0.001				0.08
13	Al			0.005		0.002	40.9	0.09
14	Si	0.008		0.001		0.001		0.06
15	P			0.05			0.05	
16	S		0.0005	0.03				
17	Cl	0.004		0.002		0.01	0.01	
19	K	0.02		0.03	21.0	0.037	0.05	
20	Ca	0.03		0.01		0.021	0.003	0.02
22	Ti			0.003			0.003	
23	V	0.0003		0.003			0.003	
24	Cr	0.0002	0.0019	0.002	0.08		0.003	0.01
25	Mn			0.001		0.002	0.001	0.015
26	Fe	0.001	0.0166	0.01	0.005	0.010	0.003	0.06
27	Co			0.003	0.00001		0.003	0.0005
28	Ni		0.0026	0.001	0.001	0.002	0.002	0.03
29	Cu			0.001		0.0006	0.001	0.01
30	Zn			0.05			0.05	0.02
33	As			0.03			0.05	
38	Sr			0.10			0.10	
40	Zr			0.03			0.01	
41	Nb				0.0001			
42	Mo			0.003	0.001		0.003	0.002
47	Ag			0.001			0.001	
48	Cd			0.005			0.01	0.0002
50	Sn			0.001			0.03	
51	Sb			0.003			0.01	
56	Ba			0.01			0.01	
73	Ta							
74	W							
82	Pb			99.32		0.008	0.001	0.002
83	Bi			0.01			0.001	

Table II-A. Limits for class C wastes appropriate for activation wastes in fusion and defined in 10CFR61.

Nuclide	Limit (Ci/m ³)
All nuclides with less than 5-year half life	No limit
H3	No limit
Co60	No limit
C14 in activated metal	80
C14	8
Ni63 in activated metal	7000
Ni63	700
Ni59 in activated metal	220
Nb94 in activated metal	0.2
Tc99	3
Sr90	7000
Ce137	4600
Alpha emitting transuranic nuclides with half lives greater than five years	100 (nCi/g)

Table II-B. Estimated limits for class C long half-life gamma emitters in fusion activation wastes not listed in 10CFR61.^a

Nuclide	Estimated limit (Ci/m ³)
A126	0.1
Nb92	0.3
Ag108m	3
Bi207	17000
Bi208	0.1
Bi210m	2

^aBased on adaptations of INVERSI and INVERSW codes in Ref. 10.

Table III. Relative rankings of materials by RMR.

Time after shutdown							
1 Hour		1 Day		10 Years		100 Years	
Material	RMR(mrem/hr)	Material	RMR(mrem/hr)	Material	RMR(mrem/hr)	Material	RMR(mrem/hr)
TENELON	9.14x10 ¹⁰	LiAlO ₂	2.83x10 ¹⁰	PCA	2.30x10 ⁸	LiAlO ₂	2.09x10 ⁴
LiAlO ₂	8.17x10 ¹⁰	VCrTi	1.36x10 ¹⁰	Nitrate salt	4.61x10 ⁷	HT9	2.07x10 ⁴
PCA	3.69x10 ¹⁰	TENELON	1.33x10 ¹⁰	HT9	4.41x10 ⁷	PCA	9.44x10 ³
ModHT9	2.93x10 ¹⁰	PCA	1.16x10 ¹⁰	TENELON	1.39x10 ⁷	LiPb	9.24x10 ³
HT9	2.89x10 ¹⁰	ModHT9	4.24x10 ⁹	ModHT9	1.11x10 ⁷	ModHT9	4.26x10 ²
VCrTi	1.93x10 ¹⁰	HT9	4.17x10 ⁹	LiAlO ₂	5.36x10 ⁶	TENELON	4.22x10 ²
Nitrate salt	8.55x10 ⁹	Nitrate salt	3.25x10 ⁹	LiPb	4.16x10 ⁶	VCrTi	1.83x10 ²
Be	4.92x10 ⁸	Be	1.67x10 ⁸	Be	1.26x10 ⁶	Be	4.99x10 ¹
LiPb	1.42x10 ⁸	LiPb	8.85x10 ⁷	Lithium	9.13x10 ⁴	Nitrate salt	2.33x10 ¹
Li ₂ O	2.17x10 ⁷	Lithium	5.86x10 ⁶	Li ₂ O	4.44x10 ⁴	Li ₂ O	1.72x10 ⁰
Lithium	1.48x10 ⁷	Li ₂ O	5.53x10 ⁶	FLIRE	2.79x10 ⁴	FLIBE	2.02x10 ⁻¹
FLIBE	6.06x10 ⁶	FLIBE	1.73x10 ⁶	VCrTi	6.53x10 ³	Lithium	1.87x10 ⁻²

Table IV. Major nuclides for high RMR.

Material	1 Hour	1 Day	10 Years	100 Years
PCA	Mn54, Mn56 Co58, Co60	Mn56 Co58, Co60	Co60	Co60, Nb94 Ag108m, Bi207
HT9	Mn54, Mn56	Mn54 Co58, Co60	Co60	Nb94
ModHT9	Mn54, Mn56 W187	Mn54 W187, Ta182	Mn54 Co60	Co60, Nb94 Ag108m, Bi207
TENELON	Mn54, Mn56	Mn54, Mn56	Mn54 Co60	Co60 Ag108m, Bi207
V15Cr5Ti	Sc46, Sc48	Sc46, Sc48	Co60 K42	Nb94 K42, A126
LiAlO ₂	Na24	Na24	Co60	A126 Ag108m
17Li83Pb	As76, Co60 Na24, Zn65 Pb203, Sb124 Zr89, Mn56 Y89m	As76, Co60 Zn65, Sb124 Na24, Zr89 Ti202, Y89m	Co60	Ag108m Bi207, Bi208
Be	Na24 Mn54, Mn56	Na24 Mn54, Co58	Co60	A126 Co60
Li ₂ O	Mn54, Mn56 Na24	Mn54 Na24, Co58	Co60 Na22	A126, Co60 Bi207
Lithium	Na22, Na24	Na22, Na24	Na22 Co60	K40 K42
Nitrate salt	Na22, Na24 Ar41	Na22, Na24	Na22	Nb94 C136
FLIBE	Mn54, Mn56 Co58	Co58, Mn54 Co60	Co60	Co60

Table V. Waste disposal rating at 10 years after shutdown.

Material nuclide	WDR (dimensionless ratio)											
	HT9	PCA	LiAlO ₂	Nitrate salt	LiPb	VCrTi	TENELON	Mod HT9	Be	Li ₂ O	FLIBE	Li
C14	1.0	0.21	0.38	70.9	0.03	0.16	0.10	0.02	0.15	0.33	0.14	0.01
Ni59	0.01	2.4										
Ni63	0.40	6.6			0.01		0.01	0.01	0.01			
Nb94	477	176	0.02	0.10	0.10	1.32	0.46	0.46	0.01			
Tc99	5.38	11.0	0.01		0.02							
Subtotal	484	196	0.41	71	0.16	1.48	0.57	0.49	0.17	0.33	0.14	0.01
A126	0.11	0.33	147		0.07	0.17	0.09	0.09	0.23	0.01		
Ag108m	0.85	0.86	2.8		10.6		0.77	0.76				
Bi207					0.02							
Bi208	0.25	0.26	0.08		13.6		0.05	0.05				
Subtotal	1.2	1.45	150		24.3	0.17	0.91	0.90	0.23	0.01		
Total	485	198	150	71	24.5	1.65	1.5	1.4	0.40	0.34	0.14	0.01

Table VI. Suitability of materials for near-surface burial.

WDR < 1	1 < WDR < 5	WDR > 5
Class A ^a	Class C ^b	Not suitable for near-surface burial ^c
Be	V15Cr5Ti	HT9
Li ₂ O	ModHT9	PCA
Lithium	TENELON	LiAlO ₂
FLIBE		17Li83Pb
		Nitrate salt

^aRequires only packaging to provide physical, mechanical, and chemical stability for near-surface burial.

^bWill be diluted to Class C by normal mixing with inert and less active materials from reactor during waste packaging. Requires packaging for physical, mechanical, and chemical stability for near-surface burial.

^cWDR and total quantities of material make reduction to Class C by dilution impractical. Processing is required to remove high activity nuclides to bring the remaining material to Class C for packaging for near-surface burial. The high activity nuclides require at least a "hot" waste facility or perhaps even deep geological burial.

Table VII. Major nuclides preventing near-surface burial.

Material	Major nuclides	Source elements
HT9	Nb94 Tc99	Nb, Mo Mo
PCA	Nb94 Tc99	Nb, Mo Mo
LiAlO ₂	Al26	Al
17Li83Pb	Bi208 Ag108m	Bi, Pb Ag
Nitrate salt	C14	N

Table VIII. Alpha emitter concentrations for LiPb.

Nuclide	Concentration (nCi/g)	
	Shutdown	10 yr after shutdown
Pb210	1.31×10^1	9.56×10^0
Bi210	6.55×10^5	1.82×10^1
Bi210m	3.25×10^{-1}	3.25×10^{-1}
Po210	4.93×10^5	1.83×10^0