Activation of Target Coatings/Hohlraums and Influence on Waste Management of IFE Chambers

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Abstract

The choice of target coating and hohlraum wall materials is among the most critical decisions to be made for inertial fusion energy (IFE) designs. Gold and gold/gadolinium have long been considered to be the coating and hohlraum wall materials of choice for direct drive (DD) and indirect drive (ID) targets, respectively, offering high target performance and low beam energy losses. More recently, a variety of other materials have been considered, including W, Pb, Pt, Pd, and Ag for the DD target coating and Au, W, Pb, Hg, Ta, Cs, and Hf for the hohlraum wall of the ID target. The choice of the coating/hohlraum material is a tradeoff between the target design elements. We identified the key safety issues and have investigated the neutron-induced irradiation effects of the candidate coating/hohlraum materials using the radiation chamber conditions of the ARIES-IFE dry wall concept. The safety requirements have specific impacts in terms of the coating/hohlraum materials choice.

1. Introduction and General Overview

Recent developments have led to viable approaches for IFE power plants. Laser or heavy ion beams can be used directly or indirectly to drive a DT target that must be repetitively injected into the chamber. In June 2000, the ARIES team established an assessment approach for IFE and investigated three types of chamber concepts, which are the dry wall, wetted wall, and thick liquid wall. Between 1975-1995, several concepts of each of these types have been proposed, but an integrated assessment by the multi-institution ARIES team was envisioned necessary at the present time. Many leading universities, national laboratories, and industries are participating in this study bringing with them their current understanding of the IFE system elements. The ARIES approach is not to develop point designs, but instead establish credible design windows and examine critical issues in concert with potential laser and heavy ion beam (HIB) drivers and targets.

The first phase of the dry wall study is now completed[1] with particular emphasis on demonstrating the tradeoff between the various design parameters, defining the design windows in key design areas, and developing the data needs and R&D priorities for IFE fusion research. During the first phase, a range of 160-400 MJ target yield served as the source of irradiation for the dry wall chamber. The radius of the solid first wall (FW) is in the 4-7 m range. The laser spherical shell targets[2] have a radius of 1.95 mm, containing the frozen DT fuel interior layer and are covered with a 300 Å thick coating. For the HIB driver,[3,4] a 15 μm thick hohlraum wall, having a 0.0085 cm^3 volume, surrounds the capsule. It is estimated that for a rep-rate of 6 Hz, approximately 190 million targets will be needed per year. The more massive HIB hohlraums produce much heavy metal debris in the chamber, 20 tonnes of Au/Gd per year as compared to 5 kg/y for Au laser target coating.

The motivation of this assessment is to develop a list of recommended coating/hohlraum materials that would offer outstanding safety features under the ARIES-IFE operating conditions. In Section 2, the main issues and concerns are addressed. Sections 3 and 4 describe the chamber configuration, calculational model, and assumptions. Major results, recommendations, and conclusions from the target activation study are presented in Sections 5 and 6.

2. Main Issues and Concerns

The sequence of the activation process would begin with the insertion of the target at the center of a spherical or cylindrical chamber that is either empty or filled with a low-pressure buffer gas. The multiple laser (or HI) beams focus on the target and initiate the DT fusion process, generating highly energetic ion debris, x and gamma rays, and neutrons. The coating/hohlraum debris interact with the source neutrons during burn and become radioactive, then travel through the cavity, and eventually condense on the solid wall in the absence or presence of a low pressure chamber buffer gas (< 0.5 torr). For high yield targets, the gas is essential to slow down the debris and mitigate the effect of the shock waves on the structure of the chamber. During the subsequent shots, the condensed materials on the wall get re-irradiated for several years, and then are disposed of with the FW and blanket at the end of their service lifetime. The accumulation of the radioactive target materials on the FW has prompted an interest in the issues regarding the waste management of the chamber structure plated with radioactive target debris. We have begun the process of evaluating the activity and the subsequent radiological inventory to assess the hazard of the target materials and their ultimate disposition as radwaste. The candidate coatings[5] and hohlraums[4] are listed in Table 1. Based on target physics, Au and Au/Gd are the materials of choice for laser and HIB targets, respectively. However, the activation of materials and target physics are not the only issues that play an essential role in the choice of target materials for IFE power plants. Other considerations include:

- Target performance (instability, gain, etc.)
- Target fabrication (cryo-layering, T fill time, etc.)
- Target injection
- Target heating (emmissivity, reflectivity, etc.)
- Tritium retention/inventory/permeation
- Safety:
  - Radioactive waste:
    - Waste disposal rating (high- or low-level waste)
    - Volume of target waste
  - Offsite dose during in-chamber accident

The technical issues and concerns related to the radiological response of the dry wall plated with radioactive target materials have not been addressed in past IFE designs. Specific work on the thick liquid wall concept with an HIB driver has focused on the activation issues of the hohlraums irradiated with target flux.[6] After burn, the hohlraum debris penetrate into a thick liquid FLiBe blanket and the authors addressed the safety issues of the recovered materials from the breeder at the end of the 30 years plant life.[6] A direct comparison between the two studies is not feasible due to the quite different nature of the problem.

Table 1. Candidate Coating/Hohlraum Materials

Laser		**HIB**	
Gold	$_{79}Au$	Gold/Gadolinium	$_{79}Au/_{64}Gd$
Tungsten	$_{74}W$	Gold	$_{79}Au$
Lead	$_{82}Pb$	Tungsten	$_{74}W$
Platinum	$_{78}Pt$	Lead	$_{82}Pb$
Palladium	$_{46}Pd$	Mercury	$_{80}Hg$
Silver	$_{47}Ag$	Tantalum	$_{73}Ta$
		$Pb/Ta/_{55}Cs$	
		Hg/W/Cs	
		$Pb/_{72}Hf$	

3. ARIES-IFE Chamber Configuration

The ARIES dry wall chamber employs the low activation SiC/SiC composites as the main structure and the $Li_{17}Pb_{83}$ eutectic as the coolant and breeder. The heat extraction and breeding zones are essentially those of the ARIES-AT advanced tokamak.[7] The design allows the flexibility of installing a thin armor on the FW to protect it against ablation by target x-rays and enhance its survivability. Currently, two materials are being considered for the armor: carbon and tungsten.[1] A schematic of the ARIES-IFE chamber radial build is shown in Figure 1. Based on neutronics calculations, the thickness of the blanket is sufficient to provide an overall tritium-breeding ratio of 1.1. A burnup limit of 3% for the SiC/SiC structure has been adopted in the study. This translates into an end-of-life fluence of 21 MWy/m^2 meaning a neutron wall loading of 3.5 MW/m^2 would correspond to a FW lifetime of 6 full power years (FPY). Figure 2 displays the influence of the FW location on the neutron wall loading for both 160 and 400 MJ target yields with 6 Hz rep-rate.

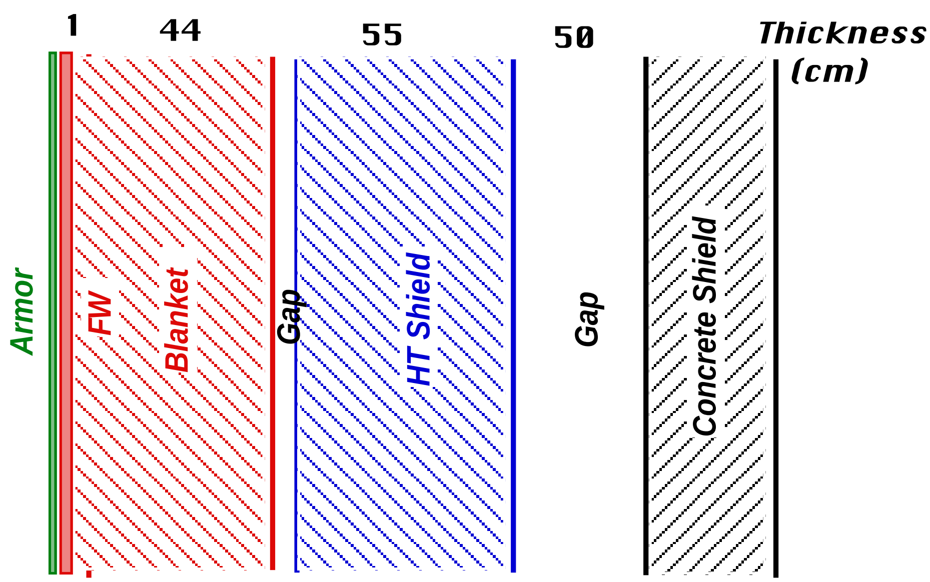

Figure 1. Radial build of ARIES-IFE dry wall chamber.

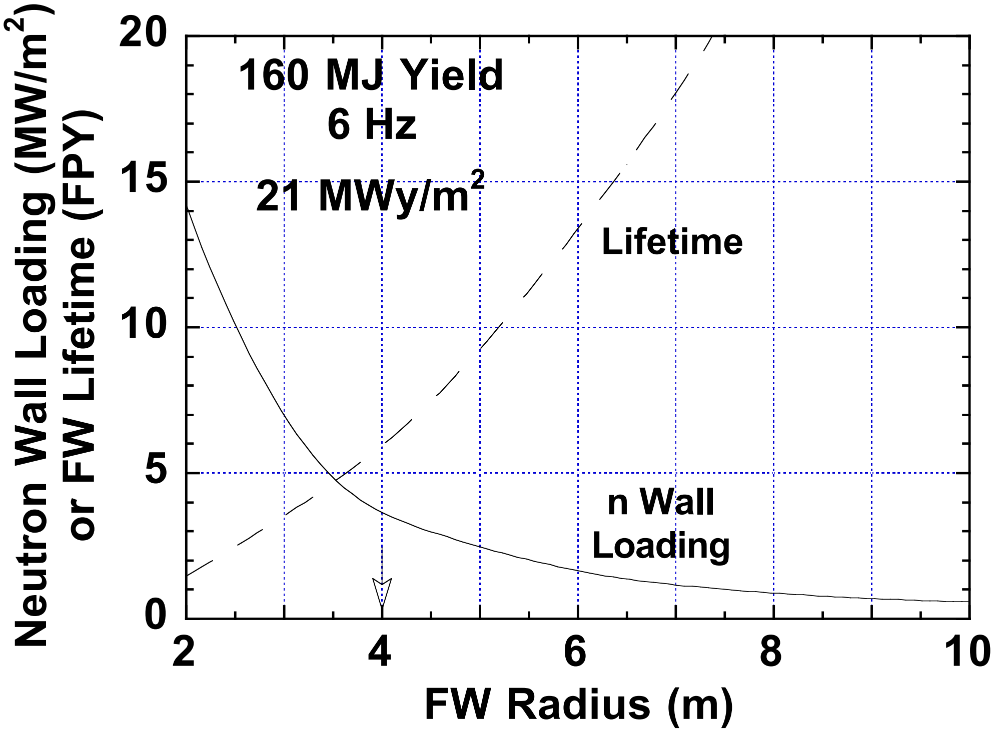

Figure 2a. Variation of neutron wall loading and lifetime with FW radius for 160 MJ target yield.

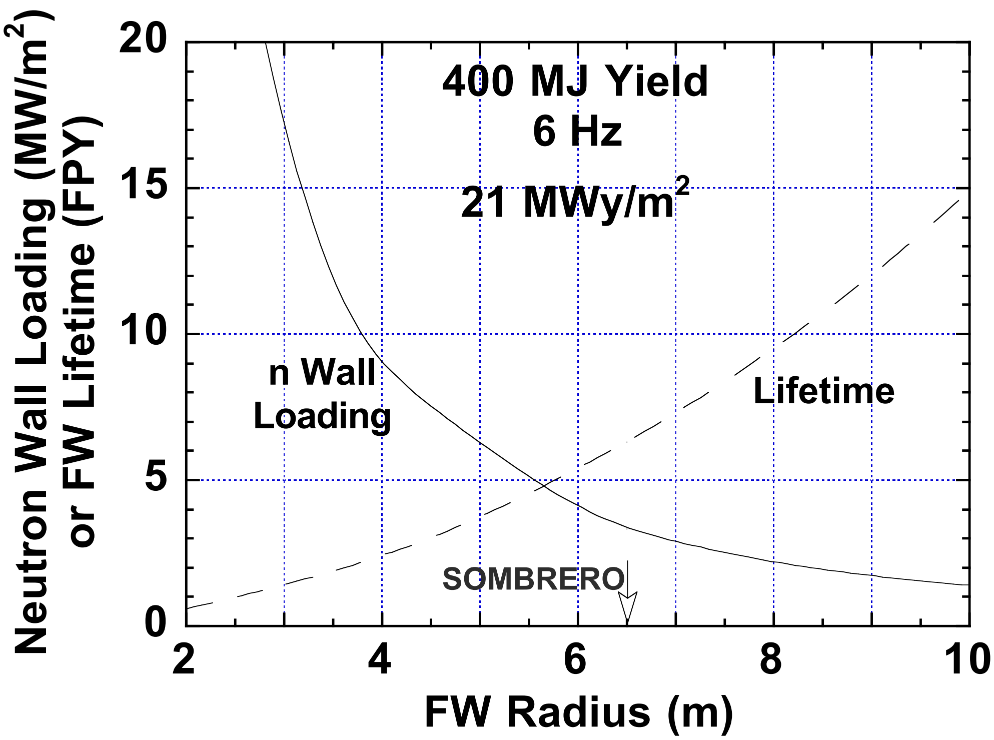

Figure 2b. Variation of neutron wall loading and lifetime with FW radius for 400 MJ target yield.

4. Model Description and Assumptions

The activation problem of the target coating and hohlraum materials is more complex than that of the chamber components. The energetic source neutrons irradiate the target materials immediately after the shot. The target materials reach the chamber wall in a few microseconds, and then get re-irradiated by the softer FW neutron flux for up to 10^9 subsequent shots before the replacement of the FW due to radiation damage. The target materials keep accumulating over the 6 FPY FW lifetime, reaching a thickness of 8 µm for the laser target coatings and 5 cm for the more massive HIB hohlraum wall materials. It seems likely that the x-rays will melt most of the HIB hohlraum materials, leaving only ~1 mm sticking on the wall[8] if the FW temperature remains below 1000°C. The molten materials would run down the FW, accumulate at the bottom of the chamber, and eventually be removed for disposal or recycling.

The irradiation history for the target coatings and hohlraums can be represented as a pulsed history with a single pulse using the target neutron flux and 10^9 pulses over the 6 FPY period using the lower and softer FW flux. This is a conservative approach because in the actual case, not all materials get re-irradiated for the entire 6 FPY lifetime of the FW. This results in slightly overestimated fluence-dependent responses such as the waste disposal rating (WDR). The model for the chamber structure is relatively simple. The activation responses for the FW and blanket are calculated using the 10^9 pulses over the 6 FPY period and the spatial distribution of the neutron flux. Both models explicitly include the effect of the 85% system availability. Table 2 summarizes the key parameters considered in the activation analysis. Note that the fluence-dependent WDR is not sensitive to the FW location as long as the material-dependent end-of-life (EOL) fluence remains fixed at 21 MWy/m^2. This means a larger chamber would call for a lower wall loading and a longer FW lifetime and will have a comparable WDR to the 4 m radius base case. A target yield of 160 MJ has been considered in this analysis. The upper value of 400 MJ will not alter the main conclusions of the analysis.

Table 2. Key Parameters for Activation Analysis

Target yield	160 MJ
FW radius	4 m
Neutron wall loading	3.5 MW/m^2
SiC/SiC FW lifetime	6 FPY
Rep-rate	6 Hz
Number of shots	190 million/y
Capsule outer radius	1.95 mm – laser 2.34 mm – HIB
Thickness of sticking target material on FW @ EOL	8 µm – laser 1 mm – HIB
Overall system availability	85%

The activity and WDR were computed using the ALARA pulsed activation code[9] and the FENDL-2 175 neutron group transmutation cross-section library.[10] The neutron flux throughout the chamber was calculated with the DANTSYS[11] discrete ordinates transport code and the FENDL-2 175-neutron 42-gamma group coupled cross-section library.[12] The computational model included the essential components that influence the analysis, namely the armor, FW, blanket, and shield as arranged in Figure 1. During operation, the coating and hohlraum wall materials (not shown in the figure) will continue to accumulate on the armor, reaching a maximum thickness of 8 μm and 0.1 cm for the laser and HIB targets, respectively. Highly pure materials were assumed for the target. The impurities for the SiC/SiC structure are taken from Reference 13.

As a top-level requirement for the ARIES power plants, all components should meet both Fetter's[14] and 10CFR61 NRC[15] waste disposal limits for Class C low-level waste.[13] A computed volume-average WDR ≤ 1 at the end of a 100-year storage period means the component qualifies for shallow land burial as low-level waste. The WDRs reported herein are based on Fetter's limits as they are more restrictive than the NRC's for all materials considered in this analysis.

5. Results

As mentioned earlier, a few millimeter thick tungsten or carbon armor protects the surface of the FW from target x-rays and debris. Figure 3 displays the WDR for the armor for two cases: a separate first wall (A/FW) and a FW attached the blanket (A/FW/B). The reported results are for highly pure armors (without impurities) and fully compacted waste. The carbon armor offers a lower WDR than the W armor. The main long-lived radionuclides contributing to the WDR of the W, C, and SiC are ^{186m}Re, ^{14}C, and ^{26}Al, respectively. From the design standpoint, it seems desirable to integrate the FW with the blanket and in this case, the armor has a relatively small impact on the already low WDR of the SiC FW/blanket (0.017). This means considerations other than the radiological issues (e.g., evaporation of the armor by target x-rays) will determine the preferred armor material. A 0.2 cm thick W armor has been considered in further activation analyses.

The specific activities for the Au coating and Au/Gd hohlraum material condensed on the FW are displayed in Figures 4 and 5, respectively. We have done some work to characterize the importance of the exposure of the coating/hohlraum materials to a single shot with the high flux of the target (curve I) versus the extended irradiation (6 FPY) with the lower FW flux (curve II). We observed that during the short burn time (tens of picoseconds), the interaction of the target materials with the source neutrons results in the highest activity almost immediately at the shutdown of the machine. This activity is dominated by the short-lived radionuclides and within a few minutes the activity drops by nearly 3-5 orders of magnitude. The activity of the target materials plated on the FW that have been re-irradiated during subsequent shots dominates the total activity at longer times after shutdown (> 10 min). This last process generates long-lived radioisotopes that are the dominant contributors to the WDR of the coating/hohlraum materials sticking on the FW. The other candidate coating/hohlraum materials exhibit similar behavior to Au as illustrated in Figures 6 and 7.

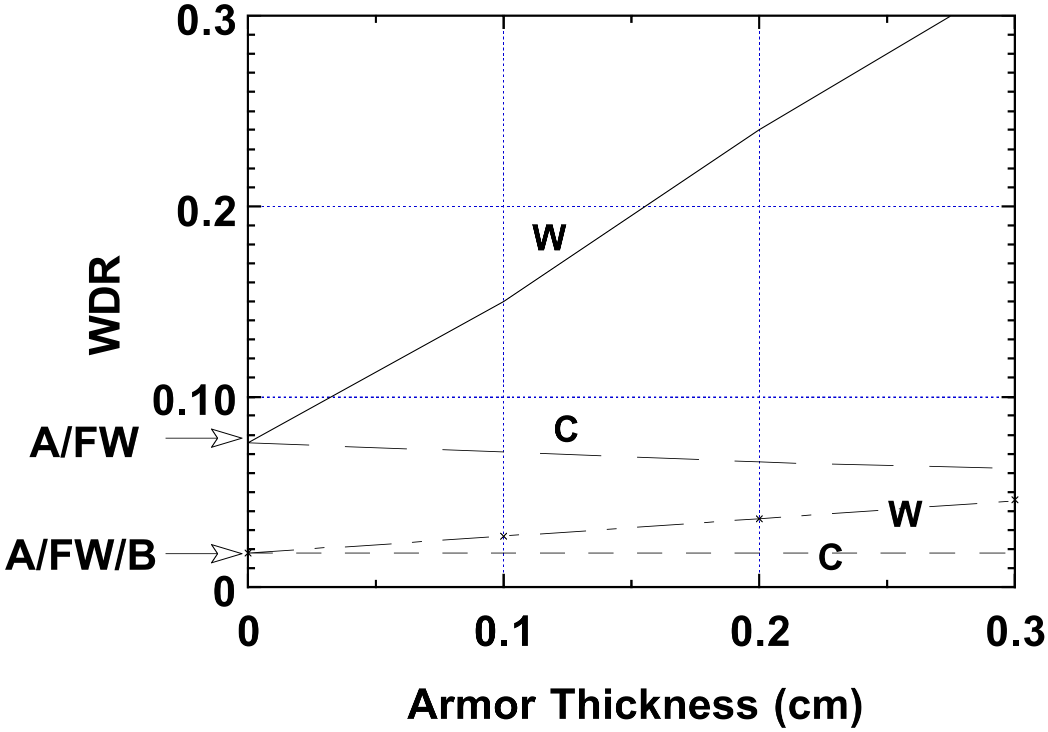

Figure 3. Impact of armor materials on waste disposal rating of FW and blanket.

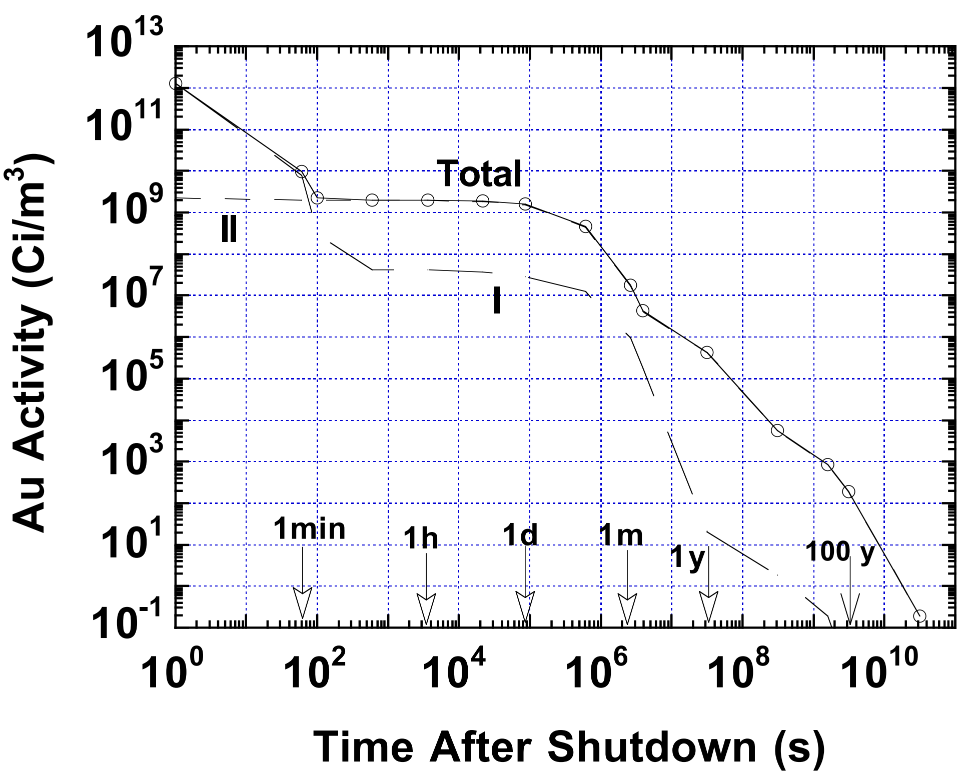

Figure 4. Activity of gold as a function of time after shutdown.

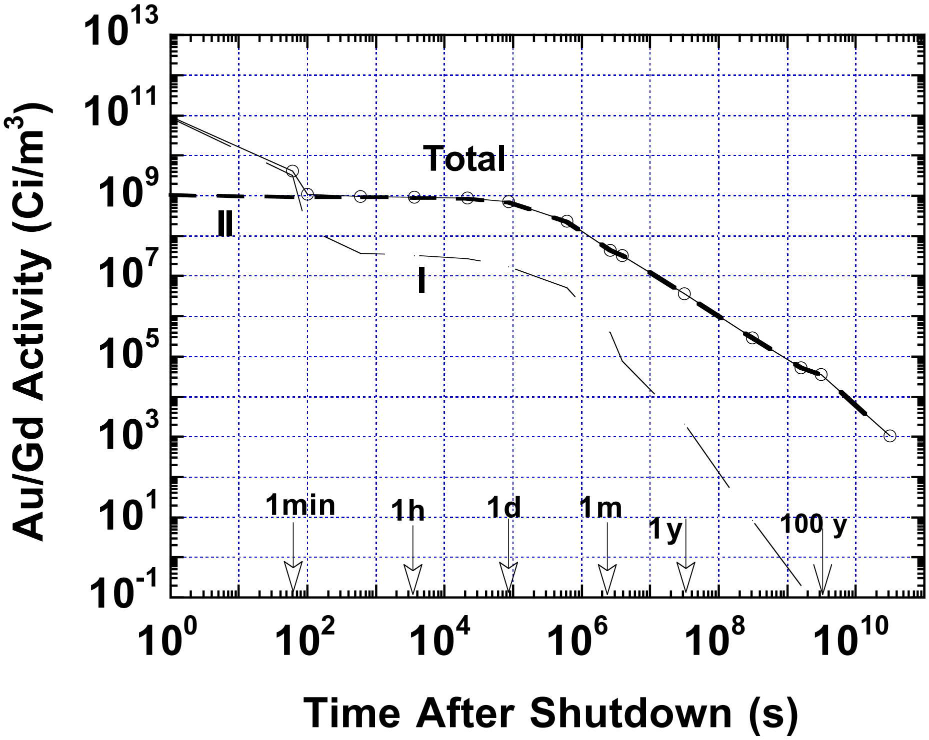

Figure 5. Activity of Au/Gd as a function of time after shutdown.

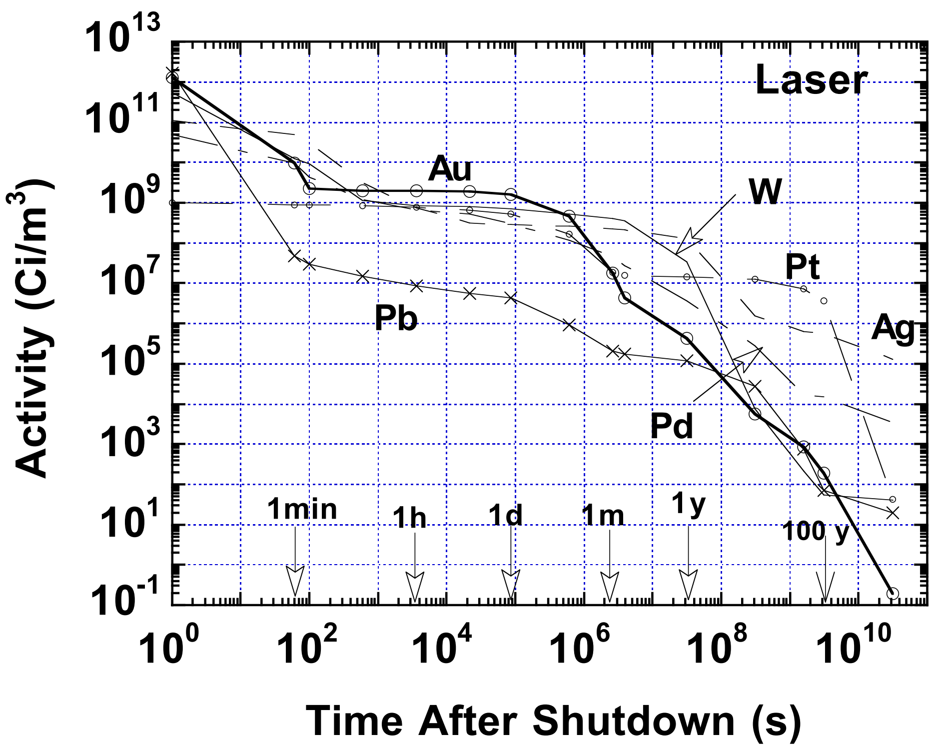

Figure 6. Activity of coating materials as a function of time after shutdown.

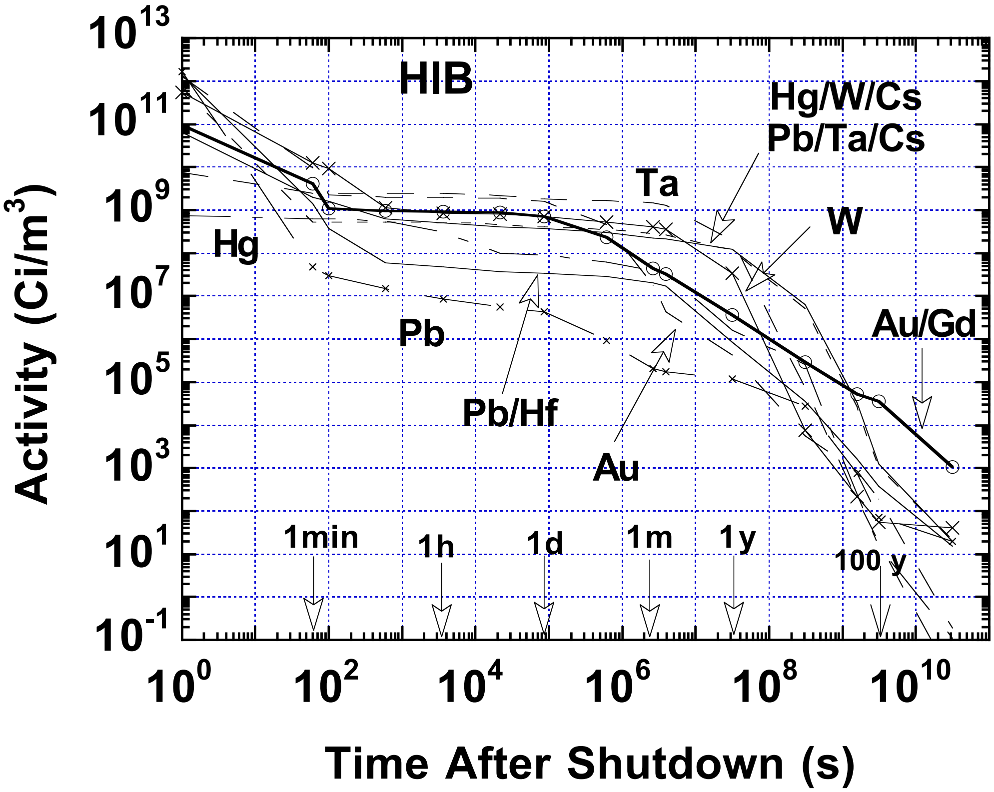

Figure 7. Activity of hohlraum materials as a function of time after shutdown.

We have determined the WDR at 100 y following shutdown, the end of institutional control at the disposal site. The volume-average WDRs are summarized in Tables 3 and 4 for the coatings (C) and hohlraum walls (H) only and for two other cases, which are C/H combined with W armor on a separate FW and C/H combined with a W/FW attached to the blanket. The main long-lived radionuclides contributing to the WDR are included between parentheses. These radionuclides have well-defined primary production pathways beginning with the original isotopes as identified in Table 5. One notes immediately that the gold-plated FW qualifies as Class C low-level waste. The silver and gadolinium generate high-level waste (WDR >>1) even when the WDR is averaged over the entire FW/blanket. Of interest is that even very thin layers of 1 µm Ag and 10 µm Gd on the FW cause waste disposal problems. Admittedly, it is feasible to separate the small amount of ^{108m}Ag and ^{158}Tb radioisotopes from the waste stream and dispose of them as high level waste. However, the high cost of the isotopic separation process could be prohibitive. If palladium is the preferred coating for laser targets, the palladium plated FW and blanket should be disposed of as a single unit to meet the Class C waste management requirements.

Table 3. Waste Disposal Rating for Target Coating Materials Condensed on the SiC/SiC Structure of a Laser-Driven ARIES-IFE Chamber

	Coating Materials	C/W/FW	C/W/FW/B
	---	0.24	0.04
Au	0.87 (^{194}Hg)	0.24	0.04
W	1.03 (^{186m}Re)	0.24	0.04
Pb	3.6 (^{208}Bi)	0.24	0.04
Pt	169 (^{192n}Ir)	0.35	0.05
Pd	4.6×10^3 (^{108m}Ag)	3.3	0.4
Ag	1.7×10^5 (^{108m}Ag)	114	12.4

Table 4. Waste Disposal Rating for Hohlraum Materials Condensed on the SiC/SiC Structure of a HIB-Driven ARIES-IFE Chamber

	Hohlraum Materials	H/W/FW	H/W/FW/B
	---	0.24	0.04
Au/Gd (50:50)*	1.2×10^4 (^{158}Tb)	924	107
Au	0.87 (^{194}Hg)	0.28	0.043
Pb	3.6 (^{208}Bi)	0.5	0.068
Hg	0.4 (^{194}Hg)	0.25	0.04
Ta	0.06 (^{182}Hf)	0.22	0.04
W	1.03 (^{186m}Re)	0.3	0.045
Pb/Ta/Cs (45:20:35)	1.5 (^{208}Bi)	0.34	0.05
Hg/W/Cs (45:20:35)	0.26 (^{194}Hg, ^{186m}Re)	0.24	0.04
Pb/Hf (70:30)	2.9 (^{208}Bi)	0.44	0.06

* atom %

To understand the tradeoff between the hohlraum materials and target performance, Meier and Callahan-Miller[16] examined the sensitivity of the conventional and close-coupled target parameters to the hohlraum wall materials of the HIB system. There are three considerations for the hohlraum wall materials: the energy loss to the ion beam, the driver energy/cost, and the incremental change in the cost of electricity (COE). Table 6 shows the results for the potential candidates relative to Au/Gd, the best material currently known. From the physics and design standpoint, a combination of Pb/Ta/Cs can have an energy loss almost as low as the Au/Gd. Both mixtures possess salient properties of high opacity and low heat capacity, offering the lowest energy loss, driver cost, and COE. However, the neutron-induced radioactivity of Gd is excessive to the extent that even an extremely thin layer of 10 µm Au/Gd condensed on the dry wall will inhibit the disposition of the chamber structure as low-level waste. This comparison suggests that other combinations of materials can work nearly as well as Au/Gd. In particular, the Hg/W/Cs and Pb/Hf mixtures have 4% higher losses and an insignificant cost penalty. Single materials would also offer attractive safety features and result in a reasonable 3-5% increase in COE.

Table 5. Summary of Important Activation Pathways

$$\left.\begin{array}{l} {}^{109}\mathrm{Ag} \xrightarrow{(n,2n^*)} \\ {}^{107}\mathrm{Ag} \xrightarrow{(n,\gamma^*)} \end{array}\right\} {}^{\mathbf{108m}}\mathbf{Ag}$$

$${}^{208}\mathrm{Pb} \xrightarrow{(n,\gamma)} {}^{209}\mathrm{Pb} \xrightarrow{\beta-} {}^{209}\mathrm{Bi} \left\{\begin{array}{l} \xrightarrow{(n,2n^*)} {}^{208m}\mathrm{Bi} \xrightarrow{IT} {}^{\mathbf{208}}\mathbf{Bi} \\ \xrightarrow{(n,2n^*)} {}^{\mathbf{208}}\mathbf{Bi} \end{array}\right.$$

$${}^{196}\mathrm{Hg} \left\{\begin{array}{ccc} \xrightarrow{(n,2n^*)} & {}^{195m}\mathrm{Hg} & \xrightarrow{(n,2n)} \\ & \downarrow \mathrm{IT} & \\ \xrightarrow{(n,2n)} & {}^{195}\mathrm{Hg} & \xrightarrow{(n,2n)} \end{array}\right\} {}^{\mathbf{194}}\mathbf{Hg}$$

$$\begin{array}{cccccc} {}^{110}\mathrm{Pd} \xrightarrow{(n,2n^*)} & {}^{109m}\mathrm{Pd} & & {}^{109}\mathrm{Ag} & \xrightarrow{(n,2n^*)} & {}^{\mathbf{108m}}\mathbf{Ag} \\ & \downarrow \mathrm{IT} & & (n,n^*)\uparrow\downarrow IT & & \\ \left.\begin{array}{l} {}^{110}\mathrm{Pd} \xrightarrow{(n,2n)} \\ {}^{108}\mathrm{Pd} \xrightarrow{(n,\gamma)} \end{array}\right\} & {}^{109}\mathrm{Pd} & \xrightarrow{\beta-} & {}^{109m}\mathrm{Ag} & & \end{array}$$

$${}^{181}\mathrm{Ta} \left\{\begin{array}{l} \xrightarrow{(n,y)} {}^{182}\mathrm{Ta} \left\{\begin{array}{l} \xrightarrow{(n,p^*)} {}^{182m}\mathrm{Hf} \\ \qquad\quad \downarrow \mathrm{IT} \\ \xrightarrow{(n,p)} {}^{182}\mathrm{Hf} \end{array}\right. \\ \xrightarrow{(n,2n)} {}^{180}\mathrm{Ta} \xrightarrow{(n,p) \text{ or e}} {}^{180}\mathrm{Hf} \xrightarrow{(n,y)} {}^{181}\mathrm{Hf} \xrightarrow{(n,y)} {}^{\mathbf{182}}\mathbf{Hf} \end{array}\right.$$

$${}^{194}\mathrm{Pt} \left\{\begin{array}{llll} \xrightarrow{(n,2n^*)} & {}^{193m}\mathrm{Pt} & & \\ & \downarrow \mathrm{IT} & & \\ \xrightarrow{(n,2n)} & {}^{193}\mathrm{Pt} & \xrightarrow{(n,p) \text{ or } \varepsilon} {}^{193}\mathrm{Ir} \xrightarrow{(n,2n^*)} {}^{\mathbf{192n}}\mathbf{Ir} & \end{array}\right.$$

$$\left.\begin{array}{llll} {}^{186}\mathrm{W} \xrightarrow{(n,\gamma)} & {}^{187}\mathrm{W} & \xrightarrow{\beta-} {}^{187}\mathrm{Re} & \xrightarrow{(n,2n^*)} \\ {}^{186}\mathrm{W} \xrightarrow{(n,2n^*)} & {}^{185m}\mathrm{W} & & \\ & \downarrow \mathrm{IT} & & \\ \left.\begin{array}{l} {}^{186}\mathrm{W} \xrightarrow{(n,2n)} \\ {}^{184}\mathrm{W} \xrightarrow{(n,\gamma)} \end{array}\right\} & {}^{185}\mathrm{W} & \xrightarrow{\beta-} {}^{185}\mathrm{Re} & \xrightarrow{(n,\gamma^*)} \end{array}\right\} {}^{\mathbf{186m}}\mathbf{Re}$$

$$\left.\begin{array}{r} {}^{157}\mathrm{Gd} \xrightarrow{(n,\gamma)} {}^{158}\mathrm{Gd} \xrightarrow{(n,\gamma)} \\ {}^{158}\mathrm{Gd} \xrightarrow{(n,\gamma)} \\ {}^{160}\mathrm{Gd} \xrightarrow{(n,2n)} \end{array}\right\} {}^{159}\mathrm{Gd} \xrightarrow{\beta-} {}^{159}\mathrm{Tb} \xrightarrow{(n,2n)} {}^{\mathbf{158}}\mathbf{Tb}$$

$${}^{197}\mathrm{Au} \left\{\begin{array}{lll} \xrightarrow{(n,2n^*)} & {}^{196m}\mathrm{Au} & \\ & \downarrow \mathrm{IT} & \\ \xrightarrow{(n,2n)} & {}^{196}\mathrm{Au} & \xrightarrow{\beta-} {}^{196}\mathrm{Hg} \xrightarrow{(n,2n^*)} {}^{195m}\mathrm{Hg} \xrightarrow{(n,2n)} {}^{\mathbf{194}}\mathbf{Hg} \end{array}\right.$$

Table 6. Energy Loss and Economic Impact of Hohlraum Materials (courtesy of W. Meier and D. Callahan-Miller,[16] LLNL)

Hohlraum Materials	$E/E_{Au/Gd}$	Driver Energy * (MJ)	Driver Cost ($B)	ΔDC ($B)	ΔCOE (mills/kWh)
Au/Gd	1	5.9/3.3	2.9/2.03	0	0
Pb/Ta/Cs	1.01	5.9/3.3			
Hg/W/CS	1.04	6/3.4	2.93/2.06	0.03	0.4
Pb/Hf	1.04	6/3.4			
Au	1.25	6.7/3.7	3.16/2.16	0.26/0.13	3.7/1.8
Pb	1.28	6.7/3.7			
Hg	1.26	6.7/3.7			
Ta	1.25	6.7/3.7			
W	1.25	6.7/3.7			

* conventional/close-coupled

6. Conclusions

We have evaluated the targets of ARIES laser and HIB inertial fusion designs from the safety perspective. The list of target coatings and hohlraum materials includes Au, Gd, W, Pb, Pt, Pd, Ag, Hg, Ta, Cs, and Hf. Unless stopped, then pumped out with the chamber buffer gas protecting the dry wall, these materials will condense on the first wall and change the attractive safety features of the low activation SiC/SiC composites employed for the ARIES-IFE chamber. We have shown that the gold plated first wall would qualify as Class C low-level waste. If palladium is the preferred coating for laser targets, the palladium plated FW and blanket should be disposed of as a single unit to meet the Class C waste management requirements. Only silver and gadolinium generate high-level wastes, considering a realistic thickness on the dry wall exceeding one and ten microns, respectively. On this basis, we recommend excluding the silver and gadolinium from the list since other materials can work nearly as well, then select the best material(s) based on considerations other than WDR and take a small penalty in the economics if necessary. Other design issues such as target fabrication/instability/gain, tritium retention, tritium fill time, and offsite dose during an accident could be addressed during the course of the ARIES-IFE study and may further limit the coating/hohlraum materials choice. The merits and additional cost associated with the exclusion of some materials should be evaluated with the perspective that the incremental change in COE is only 5% or less.

Acknowledgements

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