Conf-901007--64

UCRL-JC--103940

DE91 011154

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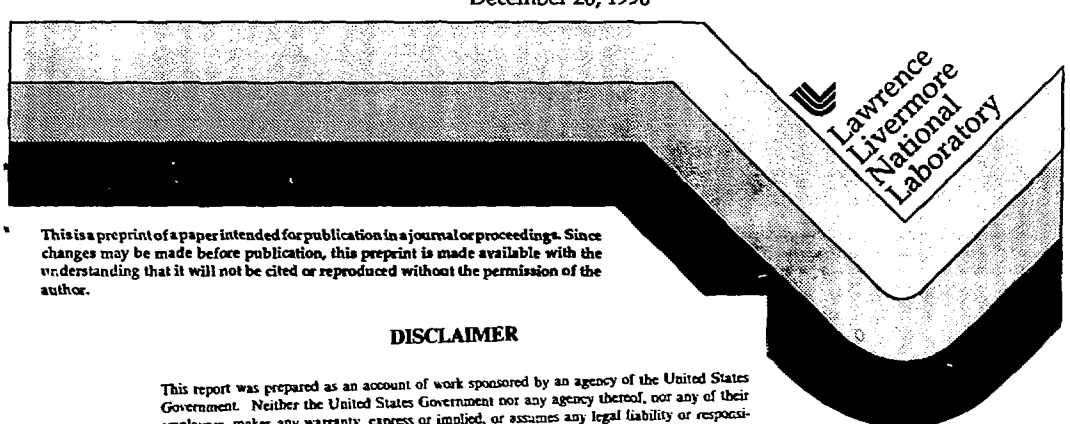
This paper was prepared for submittal to the
Ninth Topical Meeting on the Technology of Fusion Energy
Oak Brook, IL
October 7-11, 1990

December 20, 1990



NEUTRONICS ANALYSIS FOR HYLIFE-II

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ABSTRACT

A preliminary neutronics analysis of the HYLIFE-II reactor concept gives a tritium breeding ratio of 1.17 and a system energy multiplication factor of 1.14. Modified SS-316 (in which Mn is substituted for Ni) is superior to Hastelloy X and Hastelloy N as a first-wall material considering He generation, dpa-limited lifetime, and shallow-burial index. Since Flibe is corrosive to Mn metals, however, a favorable first-wall material is yet to be decided on. Flibe impurities considered (e.g., inherent impurities and those arising from wall erosion or secondary-coolant leakage) do not increase the hazard to the public over that of pure Flibe. The main issues for HYLIFE-II are the high shallow-burial index (106) and the requirement to contain some 99.7% of the ^{18}F inventory to prevent its release to the public.

INTRODUCTION

A major concern with the HYLIFE reactor concept[1] is the safety concern presented by a large amount of liquid lithium. The HYLIFE-II concept[2] seeks to remove this concern by using the molten salt Flibe (Li_2BeF_4) as the coolant. A detailed neutronics analysis is necessary because Flibe is four times more dense than liquid lithium, contains the neutron multiplier beryllium, and contains only about 28 at.% lithium. Although the conceptual design of the chamber has evolved enough to permit the necessary two-dimensional calculations to be carried out, important preliminary conclusions are obtained here with a one-

*Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore National Laboratory under contract number W-7405-ENG-48.

dimensional model. Tritium-breeding requirements will determine the Flibe fall thickness, because the thickness necessary to protect the first wall from radiation damage is less than that required to have a breeding ratio greater than 1. A stainless steel similar to the modified SS-316 used here (in which Mn is substituted for Ni) is an acceptable choice for the first structural wall in addressing radiological concerns. But Flibe attacks Mn, so a substitute for the modified SS-316 must be found. A preliminary analysis of the site-boundary dose after an accident indicates that the reactor may be built without nuclear-grade construction.

NEUTRONICS MODEL

The model for these calculations is shown in Fig. 1. Although the HYLIFE-II chamber will be cylindrical, the one-dimensional approach taken here determines the conditions for the midplane of the cylinder, which will see the greatest flux. A full two-dimensional treatment will be carried out when the parameters of the Flibe fall are better defined. Generally, it is expected that a two-dimensional calculation will yield a higher TBR and greater activation (as much as 35%) of the Flibe and the first wall.[3] The inner radius of the Flibe fall is 20 cm. The thickness is 55 cm (a 90% packing fraction is assumed), giving a 49.5-cm effective thickness. The chamber radius is 3.5 m. The calculations are for a single chamber. We assume a repetition rate of 6 Hz and a target yield of 450 MJ, giving a fusion power of 2700 MW. With a 70% capacity factor, this corresponds to a neutron source term of 6.71×10^{20} n/s. The composition is F, 57.3 at.%; Li, 28.2 at.%; and Be, 14.5 at.%. Impurities considered in the Flibe (in addition to those inherent to Flibe) included contributions from an anti-corrosion agent (MoF_6), secondary-coolant leaks ($NaBF_4$), heavy-ion target material (Ta), and erosion from the first wall (SS-316).

TRITIUM BREEDING

The one-dimensional calculations give a total tritium-breeding ratio (TBR) of 1.17. No

enrichment in ^{6}Li is required. A two-dimensional calculation would yield a higher TBR because neutrons will have a somewhat longer path length in the Flibe in the cylindrical geometry, and the Flibe mass may be as much as 4 times greater. Breeding is ~90% in ^{6}Li; the remainder is in ^{7}Li. About 85% of the tritium (TBR = 1.02) is produced in the Flibe fall; the remainder is produced in the Flibe designed to cool the structures. The radial extent of Flibe was set at 55 cm for the one-dimensional calculations to achieve a TBR > 1 in the fall. Only 0.002 neutrons per source neutron escape the main reactor system. This indicates that 30 cm of C is more than sufficient as a reflector. A total of 1.65 neutrons per source neutron are absorbed in the system. Most of the neutron multiplication (92%) takes place in the Flibe; the remainder occurs in the fuel capsule as a result of (n,2n) reactions with deuterium. Driver-port leakage will reduce the energy multiplication factor and tritium breeding ratio by factors of approximately two and three times the driver-port solid angle fraction, respectively.[4] The solid angle fraction is estimated to be 0.2%.

SYSTEM ENERGY MULTIPLICATION FACTOR

The system energy multiplication factor (SEMF) can be determined by tracking the energy deposition using transport calculations in which exoergic reactions are included. This is the case with the code TARTNP. Table 1 lists the total energy deposition by region, including the α-energy deposited in the fuel region. The system multiplication factor is $M = 1.14$. This value is not expected to change significantly with a two-dimensional calculation.

Irradiation of the Flibe at an average flux of 5.11×10^{15} n/cm^2-s gives rise to short-lived activation products that deposit their β- and γ-decay energy in the coolant. This contribution increases the energy multiplication factor to $M = 1.17$. This contribution is due almost entirely to the continuous decay of ^{16}N (60 MW of the 75-MW

total). A means to further increase the SEMF would be to introduce Mn into the Flibe in sufficient quantities to take advantage of the decay of ^{56}Mn (formed by the (n,γ) reaction). The Mn will remain in solution in Flibe.[5] Although we recommend that this option be considered, we suspect that only substantial increases in the SEMF (because it is about 1) will significantly effect the cost of electricity (COE). Further, the introduction of a medium-Z material will very likely increase the shallow-burial index, the decay thermal power, and the dose rate during shutdowns, perhaps defeating an improved COE.

ANALYSIS OF WALL MATERIALS

Three first-wall materials—Hastelloy X, Hastelloy N, and a special SS-316—were compared for activation and other effects. Their compositions were taken from Refs. 6-8. The major constituents of these three alloys are given in Table 9.

Gas Production

Table 2 shows the gas production in the three wall materials at the end of 30 yr. These calculations, done with the code TARTNP[9] and the evaluated nuclear-data library ENDL,[10] do not consider the migration of gas out of the metal; migration must be considered before the effect of gas production on strength can be fully assessed. The original HYLIFE study[1] considered the production of 500 at. ppm of He as the allowable damage limit for 2-1/4 Cr–1 Mo ferritic steel. We adopt the same value and conclude that only SS-316 will have a 30-yr lifetime.

Shallow-Burial Index

The codes ORLIB[11] and FORIG[12] were used with TARTNP to calculate the shallow-burial index (SBI), the decay thermal power, and the dose rate. The shallow burial indices are taken from 10 CFR 61 (Ref. 13). Those nuclides not covered by 10 CFR 61 are taken from Fetter et al[14]. The SBI for

SS-316 at discharge after 30 yr is nearly 16 times lower than that for Hastelloy X (1700 vs 106). The SBI for Hastelloy N (12,260) is another factor of 7 higher than that for Hastelloy X, or about 120 times higher than that for SS-316. The responsible nuclides are the same for all three wall materials, ^{94}Nb and ^{99}Tc. (The value used for ^{99}Tc was 0.2 Ci/m^3 and taken from Ref. 14.) The differences lie in how much of each is produced. Table 3 lists the inventories of these nuclides for each wall material after 30 yr.

The difference in the nuclide inventories arises from the different mass fractions of Mo and Nb in the wall compositions. The SBI for Hastelloy X could be reduced to about that of Hastelloy X if the Nb mass fraction were reduced to the same value (0.0001) and the Mo value was reduced from 13% to the allowable minimum of 11%. (Hastelloy X has 9% Mo.) It is unclear whether a 16 times higher SBI for Hastelloy than for SS-316 is a disadvantage because both require deep-burial disposal. The Mo in SS-316 is for corrosion resistance. It is uncertain whether a steel can be found for this application that will have an SBI less than 1 because most steels contain Mo for corrosion resistance. A possible candidate is the low activation steel, Fenelon[8]. Even if a steel without Mo were found to be compatible with Flibe, the N content of the steel may result in deep disposal because of ^{14}C production. A 2-D calculation may show these SBIs are actually underestimated. This is because although the volume of steel will be 20% greater in a cylindrical geometry, the softer spectrum (due to a longer neutron path length in the Flibe) may cause more activation.

Decay Thermal Power

Figure 2 compares the decay thermal power from the candidate first wall materials for 100 yr after shutdown. (Only Hastelloy X is shown because it is very similar to Hastelloy N.) The SS-316 value varies from 3 times greater than the Hastelloy steels to nearly 100 times less at the 100-yr

point. The peak decay heating value of 12 MW (SS-316) is too low to cause melting of the steel. (The maximum possible temperature is around 1000°C). The dominant nuclide during irradiation for SS-316 is ^{56}Mn, which supplies nearly all of the decay thermal power. For both Hastelloy X and Hastelloy N, the major contributor (over one-half) is ^{60}Co. For Hastelloy X, the remaining thermal power is mainly due to ^{56}Mn, while for Hastelloy N, the remaining thermal power arises about equally from ^{56}Mn, ^{58}Co, ^{95}Nb, ^{99}Mo, and ^{101}Mo, all produced by irradiation of the Ni. This is to be expected, because Hastelloy N has almost twice the Ni as Hastelloy X. The nuclide ^{63}Ni is responsible for 72% of the SS-316 decay heat at 100 yr and for 85% and 73% of the decay heat for Hastelloy X and Hastelloy N, respectively, after 100 yr.

Dose Rate

After shutdown and during dismantling, the biological dose rate to workers is a factor in determining ease and cost of disposal. Figure 3 shows the dose rate at the wall surface 100 yr after shutdown. After shutdown, ^{59}Fe, ^{54}Mn, and ^{60}Co, each dominate the dose rate in turn for SS-316; ^{60}Co dominates right up to 100 yr, after which ^{94}Nb dominates. For Hastelloy, ^{60}Co dominates throughout up to 100 yr, after which ^{94}Nb also dominates. (Again, only Hastelloy X is shown because it is similar to Hastelloy N.) Continuous worker exposure to the activated wall is permissible only for a dose rate below 2.5 mrem/h, so these walls will always require shielding, limited or controlled access, and special handling.

Displacements per Atom

The displacements per atom (dpa's) were calculated using dpa cross sections from the SPECTER computer code.[15] The cross sections used were those for Fe, which differ very little from those of Ni, Mo, Cr, and Mn, the major constituents of the three walls. This result should therefore be within an estimated 10 to 20% of the actual value

for each wall. The calculated values are 2.74 dpa/yr and an energy-averaged cross section of 127 displacement-barns. A threshold of 200 dpa was used for 2-1/4 Cr–1 Mo steel in a preliminary analysis by Meier.[16] Any of the three walls will therefore survive a 30-yr lifetime (70% capacity) without failure due to dpa damage. Meier found that the limiting rate of 9.5 dpa/yr was achieved with 31 cm of Flibe (full density). The minimum thickness required for HYLIFE-II to breed sufficient tritium may be larger (~50 cm, although a two-dimensional calculation may reduce this thickness by as much as 10 cm).

ACTIVATION OF FLIBE

We examined two radiological hazards associated with using Flibe as a blanket for the HYLIFE-II ICF reactor: the dose at the site boundary in the event of an accident (using the code HOTSPOT[17]) and the SBI. The accident scenario releases all significant nuclides present at shutdown with a 1-m/s wind speed toward a site boundary at 1 km. We assumed a 30-m release height and Pasqual D atmospheric conditions.

Five cases of Flibe activation were analyzed: pure Flibe, Flibe with those impurities Be may contribute (mainly O at 1000 wt ppm; 0.14 wt% in all), pure Flibe with 10 wt ppm MoF_6 (a possible anticorrosive in the reactor), pure Flibe with 10 wt ppm $NaBF_4$ (the secondary coolant), and Flibe with 430 wt ppm Mn-modified SS-316 steel (for an erosion rate of 0.5 mm/yr for 1 yr). These cases were examined separately to determine the contribution of each set of impurities to the base case of pure Flibe. We assumed uniform distribution of contaminants throughout the Flibe (e.g., in the fall and wall coolant regions of the reactor). The Flibe mass was 54.3 Mt and was continuously irradiated.

Site-Boundary Dose

The nuclides produced in pure Flibe listed in Table 4 contribute significantly to the accident site-

boundary dose. (The dose from a 100% release of all the rest of the nuclides is estimated be $\ll$1 rem and is therefore not considered).

We assume that all the ^{14}C is released as CO_2 and that all the ^{18}F is released as F_2. These results suggest that it is important to demonstrate that the release fractions can be reduced by factors of about 3×10^{-3} for ^{18}F and 0.4 for ^{14}C. This might be done by containment or by showing that the material is chemically bound to inert material in an accident. Tritium will provide a site-boundary dose of 16 rem for each kilogram of the nuclide released (as HTO).

When Flibe impurities are considered, additional nuclides are produced that can increase the accident site-boundary dose. They are listed in Table 5.

Again, safety analysis must show that either far less than 100% of these nuclides will be released or that the impurities in Flibe can be reduced enough to prevent their production. With 10 wt ppm MoF_4, enough ^{99}Mo is produced that a 100% release will cause a 0.5-rem dose. Therefore, 10 wt ppm is perhaps an allowable upper limit unless far less than 100% is released in an accident. For 10 wt ppm $NaBF_4$, enough ^{22}Na is produced to cause a 0.03-rem site-boundary dose. A much higher secondary-coolant weight fraction in the Flibe can probably be tolerated. Erosion of the SS-316 wall material results in several nuclides that could affect the accident site-boundary dose. They are listed in Table 6.

Shallow-Burial Index

The shallow-burial index for pure Flibe is 11, due entirely to ^{14}C. After 30 yr of irradiation the solidified coolant would have to be disposed of by deep burial. Impurities in the Flibe do not increase the SBI. For 150 to 500 wt ppm MoF_4, the Flibe will also require deep burial due to ^{99}Tc. Neither the secondary coolant, the lead, nor the steel erosion products are likely to affect the SBI.

ACTIVATION OF TANTALUM: HEAVY-ION TARGET MATERIAL

A concern unique to ICF is the potential for a radiological hazard associated with exposing large amounts of high-Z material (61 kg per day) to very high neutron fluxes (5.8×10^{19} n/cm^2 per shot) in the center of a reactor such as HYLIFE-II. We have made a first-order analysis of the hazards of the activation of target debris. We examined Ta because it is the only high-Z material under consideration that is soluble in Flibe. We assume that each capsule has a radius of 4 mm and is 35 μm thick (twice the thickness in g/cm^2 recommended by ref. 18, to be conservative).[18] Further, we assume that a week's worth of Ta (428 kg) is the total inventory required to support target fabrication and injection. This means that the Ta is continuously removed from the Flibe, held for one week, and then sent back to the target factory. The advantage of this scheme is that short-lived nuclides decay away over that week, making handling easier and avoiding the build-up of long-lived nuclides by transmutations of activation products of pure Ta. Each target is irradiated only about 1560 times during the reactor life (i.e., once a week for 30 yr). After 30 yr, 7.5% of the Ta has been converted to Hf and 2% to W. Furthermore, the Ta is now only 94% ^{181}Ta and 6% ^{180}Ta. Natural Ta is virtually all ^{181}Ta. This isotopic change is significant because the thermal neutron absorption cross section of ^{180}Ta is 30 times larger than that of ^{181}Ta. This is important for the Ta that exists in the Flibe in a steady-state quantity and is therefore continuously irradiated.

To make a first-order assessment of the possible contribution of the stored Ta inventory to the site-boundary dose in an accident scenario, we compute the dose from a complete release of all Ta immediately after the last shot under the same conditions as above. Although it is likely that the Hf and W will be removed from the Ta over time,

we include these elements to provide a worse-case result, listed in Table 7.

A final issue is the effect of residual Ta in the Flibe where it is continuously irradiated over the reactor lifetime. For a 50–wt ppm Ta concentration in the Flibe (75 kg), we determined the effect on the site-boundary dose for complete release of the nuclides produced. We set the ^{181}Ta isotopic fraction at 97.5% and the ^{180}Ta fraction at 2.5% because the recycled Ta target material has this isotopic composition after 15 yr in the reactor. The results are listed in Table 8.

CONCLUSION

A reasonable (yet conservative) release fraction is 1% for all nuclides bound in the Flibe or first structural wall (FSW). Applying this criterion to the site-boundary doses calculated above yields a cumulative maximum dose of 20 rem. Therefore, only the ^{14}C and ^{18}F appear to be areas of concern here. Both the FSW and the solidified Flibe will require deep burial after a 30-yr life. The TBR is 1.17, indicating that a large excess of tritium can be expected from this reactor. The fall can be thinned, however, to reduce the TBR to the desired level because only ~30 cm of Flibe provides sufficient protection of the FSW. The desired FSW is yet to be determined because Flibe attacks the Mn in this SS-316. Still, it is desirable that the wall material have the neutronic properties of manganese-modified SS-316.

ACKNOWLEDGMENTS

I acknowledge the helpful assistance of W. R. Meier (W. J. Schafer Associates), R. W. Moir (LLNL), and R. L. Bieri (MIT). Work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

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Figure Captions

Fig. 1. Neutronics model for HYLIFE-II.

Fig. 2. Decay thermal power of a 2.5-cm-thick first wall for two candidate first-wall materials for the HYLIFE-II reactor after 30 yr of irradiation at 4.78×10^{14} n/cm^2-s.

Fig. 3. Biological dose rate at the surface of two candidate first-wall materials for the HYLIFE-II reactor after 30 yr of irradiation at 4.78×10^{14} n/cm^2-s.

Table 1. Energy deposition for HYLIFE-II.

Region	Total energy deposited (MeV)
Flibe fall	12.93
Wall coolant (Flibe)	0.50
First structural wall	0.16
Wall coolant (Flibe)	0.41
Graphite reflector	0.07
Reflector coolant (Flibe)	0.11
Vacuum vessel	0.01
Target	5.81
Total	20.00
Leakage	0.003

[20.00/17.61 MeV = 1.14 (SEMF)]

Table 2. Gas production (in at. ppm) for three candidate first wall materials after 30 yr (70% capacity). Flux is 4.78 $\times 10^{14}$ n/cm^2-s.

Gas	Hastelloy X	SS-316	Hastelloy N
H	2927	849	4305
He	1550	349	2326

Table 3. Inventories of nuclides that determine the SBI for three wall materials after 30 yr of operation and 100 yr of decay.

Wall	Nuclide	Inventory (Ci)	Index
SS-316	^{94}Nb	39.4	106
	^{99}Tc	39.0	
Hastelloy X	^{94}Nb	64.1	1,700
	^{99}Tc	1170.0	
Hastelloy N	^{94}Nb	7746.0	12,260
	^{99}Tc	1693.0	

Table 4. Accident site-boundary dose from complete release of nuclides produced by the activation of pure Flibe.

Nuclide	30 yr inventory (Ci)	Dose (rem)
^{10}Be	4.82×10^{1}	0.3
^{14}C	7.32×10^{4}	2.5
^{18}F	2.92×10^{8}	340.0

Table 5. Accident site-boundary dose from complete release of nuclides produced by the activation of impure Flibe.

Nuclide	30 yr inventory (Ci)	Dose (rem)
^{46}Sc	4.7×10^3	1.7
^{54}Mn	3.7×10^4	3.8
^{56}Mn	2.9×10^5	1.6
^{59}Fe	9.8×10^3	1.5
^{57}Co	1.7×10^4	2.0
^{58}Co	2.0×10^4	2.3
^{60}Co	1.1×10^4	26.0
^{65}Zn	8.8×10^3	2.5

Table 6. Accident site-boundary dose for complete release of Mn SS 316 activated erodants (erosion for 1 yr) after 30 yr.

Nuclide	30 yr inventory (Ci)	Dose (rem)
^{54}Mn	9.3×10^4	9.55
^{56}Mn	8.4×10^5	4.6
^{55}Fe	4.2×10^5	8.0
^{59}Fe	1.3×10^4	2.0
^{58}Co	5.0×10^3	0.56
^{60}Co	8.4×10^3	20.0

Table 7. Site-boundary dose for complete release of Ta heavy-ion target material.

Nuclide	Activity released (Ci)	Dose (rem)
^{181}Hf	1.4×10^4	3.20
^{179}Ta	3.1×10^5	32.0
^{180m}Ta	3.1×10^6	4.6
^{182}Ta	4.4×10^4	26.0
^{181}W	6.7×10^4	0.18

Table 8. Site boundary dose for complete release of Ta material in the Flibe.

Nuclide	Activity released (Ci)	Dose (rem)
^{181}Hf	1.1×10^3	0.26
^{182}Ta	6.4×10^2	0.38
^{183}Ta	4.8×10^2	0.041
^{185}W	2.4×10^4	0.32
^{187}W	7.0×10^3	0.066
^{184}Re	2.4×10^2	0.016
^{186}Re	2.3×10^4	1.20
^{188}Re	7.6×10^4	0.24

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^{59}Fe	9.8×10^3	1.5
^{57}Co	1.7×10^4	2.0
^{58}Co	2.0×10^4	2.3
^{60}Co	1.1×10^4	26.0
^{65}Zn	8.8×10^3	2.5

Table 6. Accident site-boundary dose for complete release of Mn SS 316 activated erodants (erosion for 1 yr) after 30 yr.

Nuclide	30-yr inventory (Ci)	Dose (rem)
^{54}Mn	9.3×10^4	9.55
^{56}Mn	8.4×10^5	4.6
^{55}Fe	4.2×10^5	8.0
^{59}Fe	1.3×10^4	2.0
^{58}Co	5.0×10^3	0.56
^{60}Co	8.4×10^3	20.0

Table 7. Site-boundary dose for complete release of Ta heavy-ion target material.

Nuclide	Activity released (Ci)	Dose (rem)
^{181}Hf	1.4×10^4	3.20
^{179}Ta	3.1×10^5	32.0
^{180m}Ta	3.1×10^5	4.6
^{182}Ta	4.4×10^4	26.0
^{181}W	6.7×10^4	0.18

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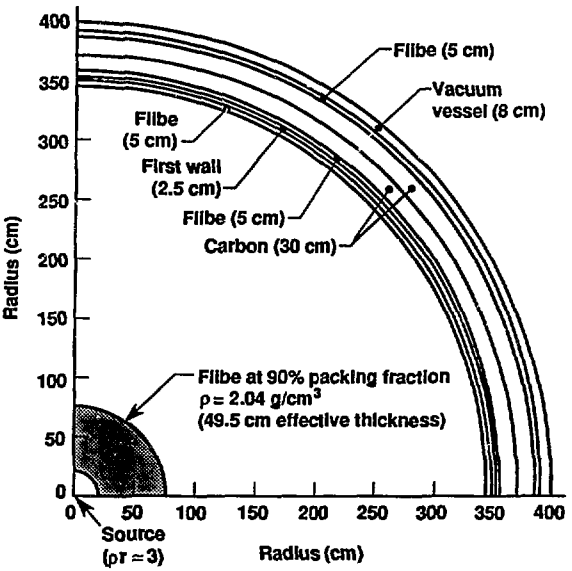
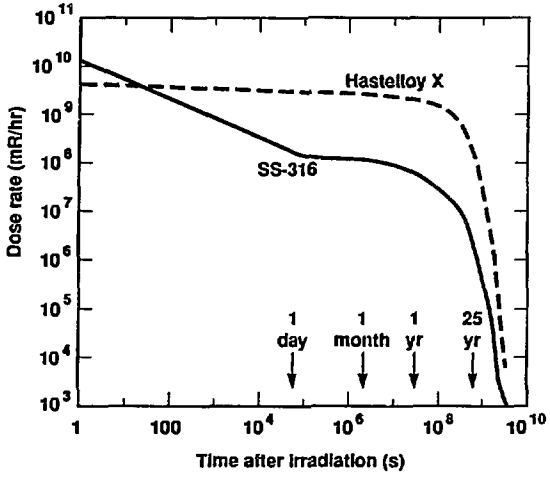
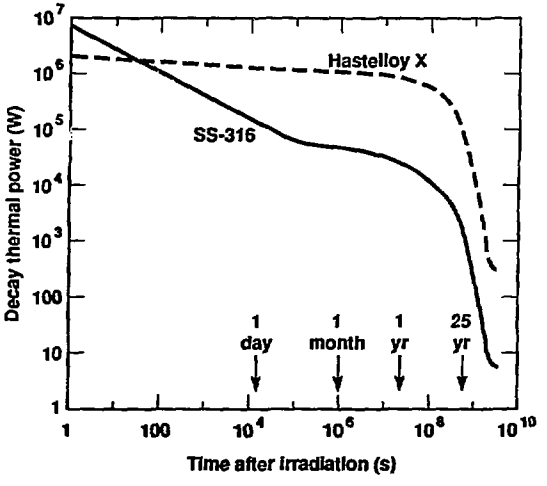
^{183}Ta	4.8×10^2	0.041
^{185}W	2.4×10^4	0.32
^{187}W	7.0×10^3	0.066
^{184}Re	2.4×10^2	0.016
^{186}Re	2.3×10^4	1.20
^{188}Re	7.6×10^4	0.24

Table 9. Wt % constituents of three candidate first walls.

Element	Hastalloy X [6]	SS316 [7]	Hastolloy N [8]
B(5)	0.005	0.005	0.001
C(6)	0.2	0.2	0.2
N(7)	0.06	0.06	0.06
Mg(12)			0.25
Al(13)	0.03	0.03	0.03
Si(14)	0.4	0.4	0.1
P(15)	0.04	0.04	0.01
S(16)	0.016	0.016	0.01
Ti(22)	0.005	0.005	0.42
V(23)	0.005	0.005	0.005
Cr(24)	22.0	10.00	8.0
Mn(25)	1.37	18.00	0.25
Fe(26)	18.5	70.13	0.1
Co(27)	1.5	0.016	1.5
Ni(28)	46.23	0.7	70.69
Cu(29)	0.03	0.03	0.03
Zn(30)	0.003	0.003	0.003
As(33)	0.02	0.02	0.02
Se(34)	0.005	0.005	0.005
Zr(40)	0.005	0.005	0.005
Nb(41)	0.01	0.01	2.0
Mo(42)	9.00	0.3	13.0
Ag(47)	0.0001	0.0001	0.0001
Cd(48)	0.0002	0.0002	0.0002
Sn(50)	0.005	0.005	0.005
Sb(51)	0.001	0.001	0.001
Hf(72)			1.58
Ta(73)	0.005	0.005	0.005
W(74)	0.6	0.01	0.6
Pb(82)	0.001	0.001	0.001
Bi(83)	0.001	0.001	0.001


Flibe (5 cm)
Vacuum vessel (8 cm)
Flibe (5 cm)
First wall (2.5 cm)
Flibe (5 cm)
Carbon (30 cm)
Flibe at 90% packing fraction
ρ = 2.04 g/cm³
(49.5 cm effective thickness)
Source
(ρr ≃ 3)
Radius (cm)
Radius (cm)
0
50
100
150
200
250
300
350
400


Dose rate (mR/hr)
10^{11}
10^{10}
10^{9}
10^{8}
10^{7}
10^{6}
10^{5}
10^{4}
10^{3}
Hastelloy X
SS-316
1 day
1 month
1 yr
25 yr
1
100
10^{4}
10^{6}
10^{8}
10^{10}
Time after irradiation (s)


Hastelloy X
SS-316
1 day
1 month
1 yr
25 yr
Decay thermal power (W)
Time after irradiation (s)