A Reinvestigation of the Tritium Inventory in the Carbon Structure of the SOMBRERO Direct-Drive Conceptual Design Study

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Introduction

This paper summarizes additional studies made during 1999 to evaluate the tritium inventory in the carbon fiber composite (CFC) structure of SOMBRERO's design published in 1992. Briefly, the CFC material forms the complete reactor structure (Figure 1). The reactor wall is divided into five channels from front to rear with each channel filled with flowing small pellets of lithium oxide (Li_2O). These particles transport thermal energy from the reactor walls to a heat exchanger located below the reactor. This Li in the pellets absorbs neutrons during the pellet burn phase, generating tritium to resupply the fuel cycle. Helium at the rate of 2 m^3/s flows upward through the channels and transports the newly formed tritium to a collecting system external to the reactor.

The gas phase in these channels described above can become a chemical reducing agent to the Li_2O pellets, forming Li and LiH vapors, which would react with the graphite structure. Therefore, H_2O

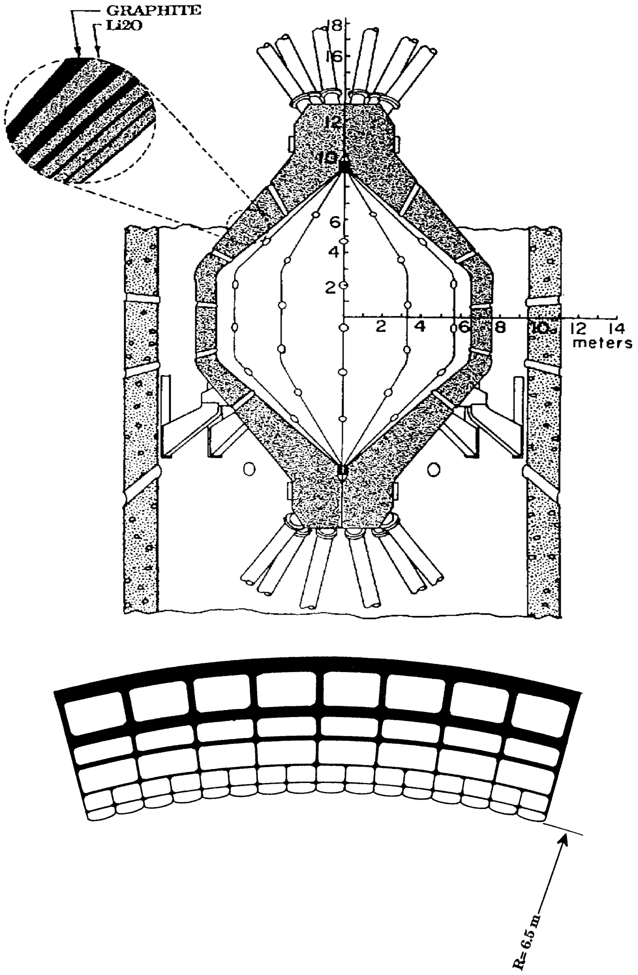


Figure 1. Schematic design of the SOMBRERO reactor.

(steam) was added as an oxidant to the helium purge stream. An additional advantage of the H_2O addition is that it catalyzes the desorption of tritium from the breeder pellets to form the stable compounds HTO and H_2 in the gas, which are recovered in the external circuit.

The determination of the tritium inventory in the graphite structure must consider several factors. These items are discussed in the following sections:

1) The oxidation of the graphite by H_2O and HTO,
2) The solubility of HT in the graphite, and
3) The retention of HT in the traps formed in the graphite by the energetic neutrons released during the target burn phase.

Oxidation of Graphite by Water

As previously noted, water vapor is added to the fuel channels of the reactor in order to retain the fuel (Li_2O) in an oxidized state and to form the stable compound HTO. The addition of H_2O, however, does oxidize the graphite structure via the chemical reaction: $C + HTO \rightarrow CO + HT$.

The initial experimental study of this reaction (Hirooka and Iami, 1982) indicated that this oxidation rate was slow (Figure 2). Subsequently, measurements over a wide range of temperatures and steam pressure (Smolik, et al., 1992) showed that the Hirooka measurements were low by a factor of ~100 and that the oxidation rate was controlled by the water pressure to the first power. Smolik's data were confirmed by the measurements of Iami, et al. (1992). Based on this information, a series of oxidation rates at several values of water vapor pressure were calculated (Figure 2).

Also shown in Figure 2 are the allowable corrosion rates for the first wall channel and the blanket channels of the SOMBRERO reactor. These limits are based on restricting the corrosion to a maximum of 10% of the thickness of the channel walls during their useful lifetime of ~ two years.

Because the first wall channel operates at such a high temperature, the P_{H2O} in this channel is limited to 2×10^{-5} atm (2 Pa) and is maintained at this value in all of the channels.

The production rate of tritium in the breeder particles is 2×10^{-3} gram atoms T/s. Therefore, H_2O must be added at the same rate to form 2×10^{-3} moles/s of HTO in the gas phase. At $P_{H2O} = 2$ Pa, the volume of H_2O which must be added is 5.9 m^3/s in the first channel and 4.2 m^3/s combined for the four blanket channels.

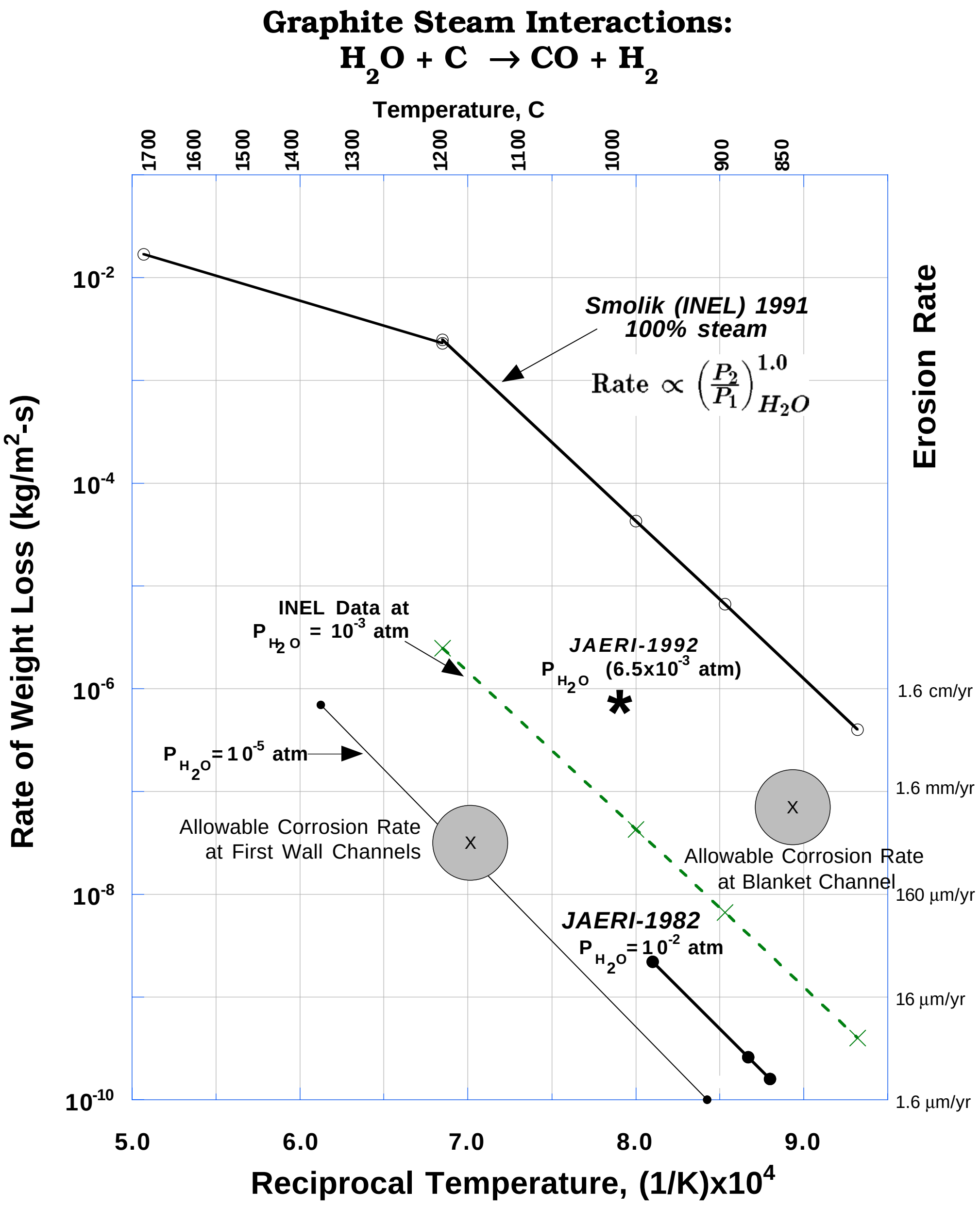


Figure 2. Erosion rates of graphite with steam.

Tritium Trapping in Irradiated Carbon Fiber Composites

Continued irradiation of graphite with energetic neutrons displaces carbon atoms from their normal position in the graphite structure and forms vacancies or voids, which can trap the hydrogen isotopes in the structure. Experimental measurements of the tritium retention as a function of radiation damage have been performed (Kwast et al., 1996, and Causey et al., 1996). In each of these experiments, carbon specimens were sealed in metal capsules and irradiated for various periods in a research reactor. Following the irradiation, the capsules were shipped to a tritium laboratory where the capsules were opened and the contents exposed to tritium gas. Following this

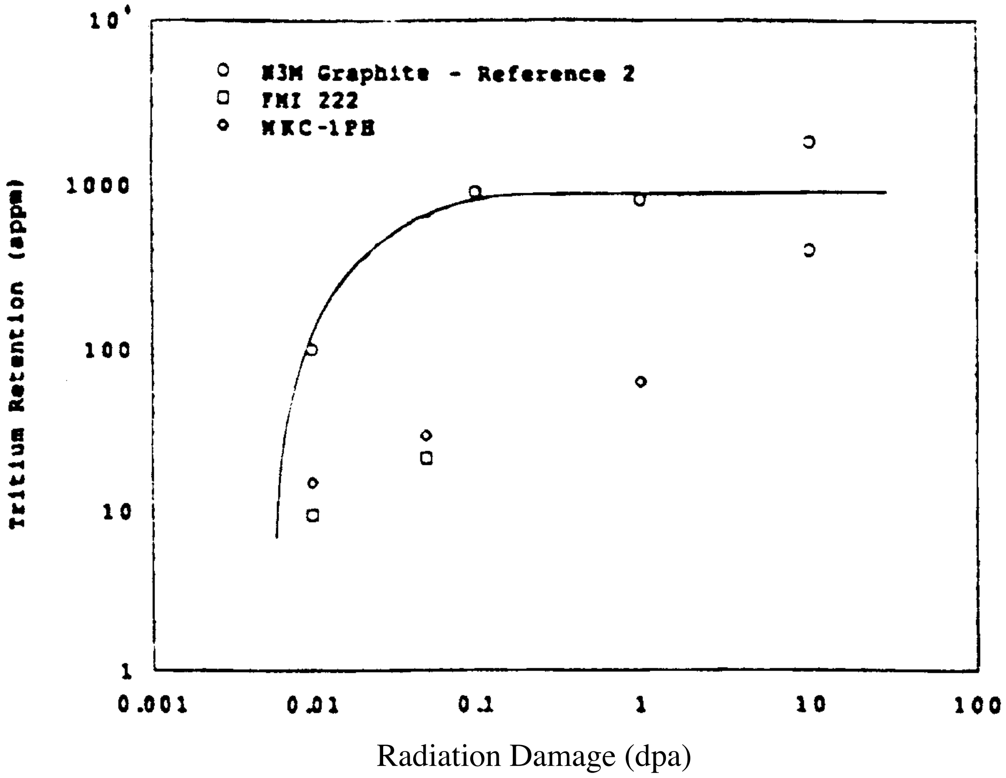


Figure 3. Comparison of tritium trapping in irradiated carbon composites and graphites.

exposure, the excess tritium gas was removed and the carbon specimens were heated to release the tritium trapped in the irradiated carbon.

The results (Figure 3) from Causey et al. indicate that the trapping in carbon composites formed with carbon fibers is nearly one order of magnitude less than for bulk graphite specimens. Radiation damage calculations for the SOMBRERO blanket indicate that the radiation damage would be 16 dpa/FPY at the front-facing wall of the first channel and 12 dpa/FPY for the rear portion of the channel for an average of 14 dpa/FPY. Based on Figure 3, this radiation damage would trap 240 appm per FPY of (T+H) in 20 Mg of carbon, for a total of 1.2 kg of tritium per 2 FPY.

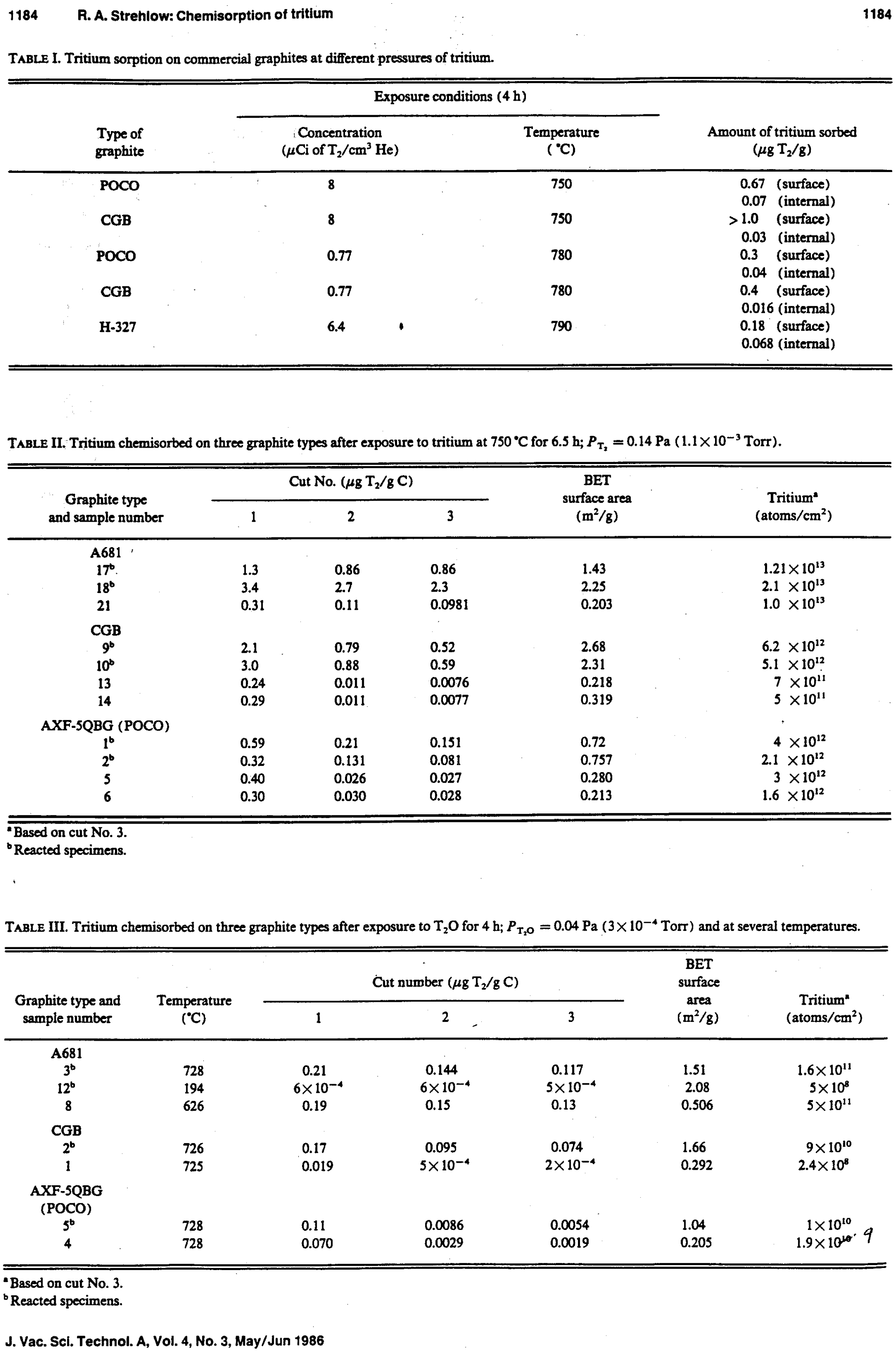
For the remainder of the blanket, the calculated radiation damage in the carbon structure decreases from 12 dpa/FPY to essentially zero at the back edge of the reactor, giving a weighted average of 4 dpa/FPY, comparable to 125 appm (H+T) per FPY for the total 430 Mg remaining carbon structure. For the two-year life of the outer blanket, the total tritium retained would be 13.4 kg. For the total carbon structure, the tritium inventory would be 14.6 kg (T) for 2 FPY if no countermeasures were taken to reduce the tritium trapping.

Tritium Trapping in Irradiated Carbon When HTO is Present

The above discussion assumed that the tritium existed as molecular HT in the He gas. However, in SOMBRERO, the tritium exists in the oxide form HTO, which cannot dissolve in graphite. Alternatively, HTO can corrode graphite as shown in Figure 2, and release HT, which can be absorbed in the structure. An experiment analogous to these conditions was performed by Strehlow (1986) in which he exposed 1 cm cubes of several grades of graphite to T_2O at a pressure of 0.04 Pa, and a temperature of ~750°C for four-hour exposures as shown in Figure 4. In a subsequent experiment, specimens of the same types of graphite were exposed to molecular T_2 at a pressure of 0.14 Pa for 6.5 hours with the results shown in Figure 4. In order to compare these results, the values for tritium dissolution from T_2O were corrected to the same pressure and exposure time as used for the T_2 experiment based on Atsumi's observation (Atsumi, et al., 1998) that tritium dissolution in graphite scaled to the square root of the tritium pressure and to time of exposure to the first power. These corrected values for each type of graphite were compared with the dissolution of tritium when molecular T_2 gas was used. These results (Table 1) indicated that, on average, the tritium dissolved in the graphite due to T_2O was only 10% as much as when T_2 was utilized.

Strehlow's values can be compared also with the calculated amount of T_2 released from the T_2O due to the erosion of the graphite. For the temperature and pressure of the T_2O, Figure 5 indicates that the corrosion of graphite would be 10^{-14} kg/m^{2}s. For the four-hour exposure, the corrosion of graphite is 1.2 x 10^{-12} g.at.(C)/cm^2. Because 2 g.at.(T) are released for each g.at.(C) and there are 6 x 10^{23} atoms/g.at.(T), the free tritium released is 1.4 x 10^{12} atoms (T)/cm^2. Comparing this value with the experimental values determined by Strehlow (Table 2) indicates that only approximately 8.7% of the tritium released from the T_2O was absorbed in the graphite, very similar to the ratio shown in Table 1. Strehlow suggested that the reason for the low absorption of the tritium in the graphite is caused by the oxidation process of T_2O, which attacks the reactive sites on the carbon surface. These sites are normally the preferential sites for tritium to enter the carbon structure, hence destroying these sites prevents the absorption of tritium.

Based on the above information, the tritium retention in the carbon fiber composite structure of SOMBRERO can be estimated. For the first and second walls, the average temperature is ~1050°C



1184 **R. A. Strehlow: Chemisorption of tritium** 1184

TABLE I. Tritium sorption on commercial graphites at different pressures of tritium.

Type of graphite	Exposure conditions (4 h) Concentration (μCi of T_2/cm^3 He)	Temperature (°C)	Amount of tritium sorbed (μg T_2/g)
POCO	8	750	0.67 (surface)
			0.07 (internal)
CGB	8	750	> 1.0 (surface)
			0.03 (internal)
POCO	0.77	780	0.3 (surface)
			0.04 (internal)
CGB	0.77	780	0.4 (surface)
			0.016 (internal)
H-327	6.4	790	0.18 (surface)
			0.068 (internal)

TABLE II. Tritium chemisorbed on three graphite types after exposure to tritium at 750 °C for 6.5 h; $P_{T_2} = 0.14$ Pa (1.1×10^{-3} Torr).

Graphite type and sample number	Cut No. (μg T_2/g C) 1	2	3	BET surface area (m^2/g)	Tritium[a] (atoms/cm^2)
A681					
17[b]	1.3	0.86	0.86	1.43	1.21×10^{13}
18[b]	3.4	2.7	2.3	2.25	2.1×10^{13}
21	0.31	0.11	0.0981	0.203	1.0×10^{13}
CGB					
9[b]	2.1	0.79	0.52	2.68	6.2×10^{12}
10[b]	3.0	0.88	0.59	2.31	5.1×10^{12}
13	0.24	0.011	0.0076	0.218	7×10^{11}
14	0.29	0.011	0.0077	0.319	5×10^{11}
AXF-5QBG (POCO)					
1[b]	0.59	0.21	0.151	0.72	4×10^{12}
2[b]	0.32	0.131	0.081	0.757	2.1×10^{12}
5	0.40	0.026	0.027	0.280	3×10^{12}
6	0.30	0.030	0.028	0.213	1.6×10^{12}

[a] Based on cut No. 3.

[b] Reacted specimens.

TABLE III. Tritium chemisorbed on three graphite types after exposure to T_2O for 4 h; $P_{T_2O} = 0.04$ Pa (3×10^{-4} Torr) and at several temperatures.

Graphite type and sample number	Temperature (°C)	Cut number (μg T_2/g C) 1	2	3	BET surface area (m^2/g)	Tritium[a] (atoms/cm^2)
A681						
3[b]	728	0.21	0.144	0.117	1.51	1.6×10^{11}
12[b]	194	6×10^{-4}	6×10^{-4}	5×10^{-4}	2.08	5×10^{8}
8	626	0.19	0.15	0.13	0.506	5×10^{11}
CGB						
2[b]	726	0.17	0.095	0.074	1.66	9×10^{10}
1	725	0.019	5×10^{-4}	2×10^{-4}	0.292	2.4×10^{8}
AXF-5QBG (POCO)						
5[b]	728	0.11	0.0086	0.0054	1.04	1×10^{10}
4	728	0.070	0.0029	0.0019	0.205	$1.9\times10^{\cancel{10}}$ 9

[a] Based on cut No. 3.

[b] Reacted specimens.

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Figure 4. Chemisorption of tritium on graphite after exposure to T_2O or T_2 (Strehlow's 1986 report).

Table 1. Comparison of the chemisorption of tritium in graphite from either molecular T_2 or T_2O.

Graphite Type	Sample No.	Tritium Species	Tritium Conc. atoms/cm^2	Adjusted T_2O Conc.	Conc. Ratio T atoms from T_2O/T_2
A681	#17	T_2	1.2×10^{13}		
	#3	T_2O	1.6×10^{11}	4.8×10^{11}	4×10^{-2}
	#21	T_2	1×10^{13}		
	#8	T_2O	5×10^{11}	15×10^{11}	15×10^{-2}
CGB	#13	T_2	7×10^{11}		
	#2	T_2O	9×10^{10}	2.7×10^{11}	39×10^{-2}
	#14	T_2	5×10^{11}		
	#1	T_2O	2.4×10^{8}	7.2×10^{8}	0.14×10^{-2}
AXF	#5	T_2	3×10^{12}		
	#1	T_2O	1×10^{10}	3×10^{10}	1×10^{-2}
	#6	T_2	1.6×10^{12}		
	#4	T_2O	1.9×10^{10}	5.7×10^{10}	3.6×10^{-2}
				Average	10.5×10^{-2}

Table 2. Experimental values for tritium absorption in graphite from T_2O compared with the amount of T_2 released from T_2O by the erosion of graphite (pressure $T_2O = 0.04$ Pa; T = 750°C, exposure time = 4.0 hours).

Graphite Sample	Tritium Expt. Conc. atoms/cm^2	Calc. Tritium atoms/cm^2 Released by Erosion	Ratio (Expt./Calc.)
A681			
#3	1.6×10^{11}	1.44×10^{12}	1.1×10^{-1}
#8	5×10^{11}	1.44×10^{12}	3.4×10^{-1}
CGB			
#2	9×10^{10}	1.44×10^{12}	6.25×10^{-2}
#1	2.4×10^{8}	1.44×10^{12}	1.67×10^{-4}
AXF			
#5	1×10^{10}	1.44×10^{12}	6.9×10^{-3}
#4	1.9×10^{10}	1.44×10^{12}	1.3×10^{-3}
		Average	8.7×10^{-2}

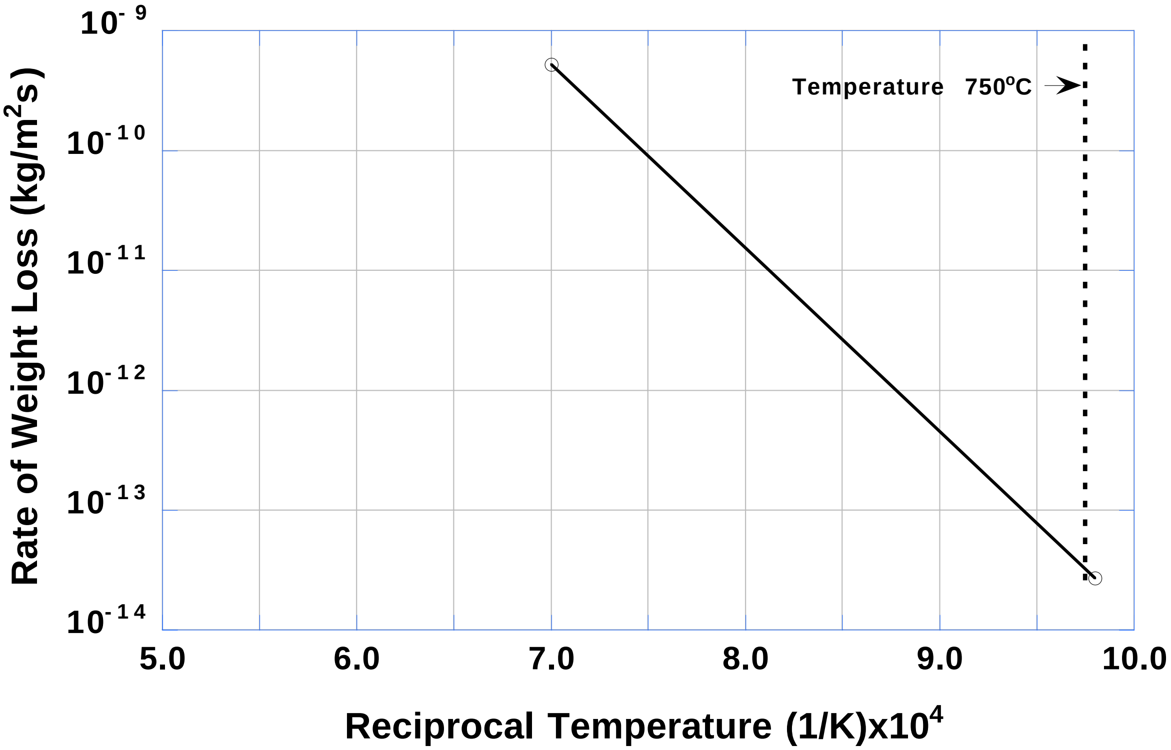

Figure 5. Erosion rate of graphite with low steam pressure, 4 x 10^{-7} atm H_2O (extrapolated from Smolik's data in Figure 2).

with a combined weight of 20 Mg and subjected to pressure of 2 Pa of HTO. Based on the erosion data (Figure 2), the rate weight loss of carbon on the inside of the first wall is 2 x 10^{-9} kg/m^{2}s.

The surface area inside the first wall is 735 m^2; hence, the erosion rate is 4 x 10^3 g.at.C/yr. For each g.at.C eroded, one mole of HT (bearing one g.at.T) is released from HTO. According to the analysis of Strehlow's experiment, only 8.7% (4.4 mole % HT) of the tritium is absorbed by the carbon [i.e., 176 g.at.T/yr (0.53 kg T/yr)]. The same erosion and T absorption occurs on the inside face of the second wall so that for a two year life, the T absorption would be 2.12 kg (T).

Similarly, for the outer section of the blanket, the average temperature is only 850°C. The erosion rate (1 x 10^{-11} kg/m^{2}s) for 8 carbon walls is 1.85 kg/yr, which releases 154 moles of (HT)/yr. For 4.4 mole % HT absorption and a two year life, the tritium absorption is 0.041 kg(T). For the total carbon structure, the tritium inventory is 2.16 kg (T).

The above calculations were based on the assumption that the concentration of HTO in the gas phase was constant from the bottom to the top of each of the graphite reactor channels. In fact, the water vapors enter the bottom of each channel as molecular H_2O and gradually form HTO as the gases rise through each channel and are not fully reacted to form HTO until the gases reach the top of the reactor. As a result, the dissolution of tritium in the graphite channels is not uniform, but nearly zero at the bottom and reaches the calculated solubility only near the top of the channel. As a result, the average tritium concentrate in the graphite channel would be only 50% of the calculated value, giving a total inventory in the reactor of only 1.08 kg at the end of its two year lifetime.

Summary

The tritium inventory in the carbon structure when tritium is in the oxidized form as HTO is only 4.4% as much as when the tritium is in the non-oxidized state as T_2. This result is due to the fact that HTO is not soluble in carbon, but must be reduced to the molecular state (HT). The only mechanism to reduce HTO is by oxidation of the carbon. During the oxidation process, the reactive sites of the carbon are destroyed. These sites are normally entryways for tritium into the carbon structure. As a result, sufficient H + T atoms are not available to satisfy all of the radiation-induced traps in the carbon structure, which could retain nearly 15 kg of tritium in two years while only 1 kg is available by the reduction of HTO by carbon.

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