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ESTIMATE OF TRITIUM PERMEATION OUT OF TPBAR WASTE CONTAINER (U)

Elliot A. Clark

26 August 2004

Westinghouse Savannah River Company
Aiken, SC 29808

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Materials Technology Section

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ESTIMATE OF TRITIUM PERMEATION OUT OF TPBAR WASTE CONTAINER

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ESTIMATE OF TRITIUM PERMEATION OUT OF TPBAR WASTE CONTAINER

Elliot A. Clark

Executive Summary

The rate of tritium permeating out of a proposed waste container for the Commercial Light Water Reactor- Tritium Extraction Facility is estimated. This estimate conservatively assumes that all of the residual tritium in the already extracted Tritium Producing Burnable Absorber Rods (TPBARs) is available as diatomic gas to permeate the outer container. The permeation rate is calculated for each year after disposal, at two temperatures determined by a separate thermal analysis of this waste form in disposal. All the residual tritium permeates out of the container in 17 years at 200 F and in 24 years at 175 F. Results of this estimate will be used in a Special Analysis to assess waste disposal options.

Introduction

The Commercial Light Water Reactor program will produce tritium for Defense Programs using Tritium Producing Burnable Absorber Rods (TPBARs) installed in a light water reactor. Neutron irradiation of the rods in the reactor creates tritium by fission of the isotope lithium-6, and the tritium will be trapped in the TPBAR by zirconium and zircaloy getters. Tritium will be thermally extracted from the TPBARs in the new Tritium Extraction Facility at the Savannah River Site, using furnaces designed for this purpose. After extraction, there will be a small amount of residual tritium in the spent TPBARs. The spent TPBARs will be placed inside several containers, and finally will be contained by a carbon steel container designed to shield gamma radiation coming from activated stainless steel cladding of the TPBARs.

In all likelihood the residual tritium in the extracted TPBARs will remain chemically bound as a hydride in the zirconium and zircaloy getters. This analysis evaluates the hypothetical worst-case scenario of all the tritium being immediately released from the getters as tritium gas, and this gaseous tritium permeating the outer waste container. (The interior nested containers of the TPBARs, which include the consolidation container and the furnace extraction basket, are not sealed and so provide no permeation barrier.) The outer container is considered to be the tritium permeation barrier in this analysis. The amount of tritium permeating the outer container each year will be input to a so-called Special Analysis to assess where this waste form can be disposed of.

Analysis

The current design of the outer container is a rectangular box made of SA-516, Grade 70 carbon steel (Fig. 1). Three permeation paths are considered. First, tritium permeates the side walls and bottom of the container. The wall thickness is 13 inches on all sides and the bottom. The internal wall surface that is exposed to the hypothetical tritium gas is a box 36” by 36” by 202”. Second, tritium permeates the outer seal lid. This lid is welded to one end of the container and is 47” by 47” by 1” thick (Fig. 1). (No credit is taken for the bolted upper lid, which provides radiation shielding

for the top, to reduce permeation.) Third, tritium permeates the weld used to attach the outer seal lid. The weld is considered to be ½" deep, ½" wide, and has a length of 4* 47" or 188".

The permeability is defined by

$$\Phi \equiv D \times S \qquad 1$$

in which D is the diffusivity of tritium in the material (cm^2/s) and S is the solubility (here cc H_2 @ STP/cc material). The unit of Φ is cm^2- cc H_2 @ STP /s/cc material. The maximum permeation rate through a material of thickness L is the so-called steady state flux:

$$Flux = \frac{\Phi}{L} \qquad 2$$

The flux is the amount of tritium (cc H_2 @ STP/cm^2/s) permeating per unit area per unit time. Because of the rapid diffusivity of hydrogen isotopes in this material, the steady state permeation rate is achieved in about one week in 13" thick walls (much faster for the thinner seal lid and weld paths) and so steady state equations describe the permeation occurring over years very accurately.

The diffusivity of hydrogen isotopes in iron is represented by

$$D = D_0 * e^{-\frac{Q}{RT}} \qquad 3$$

where D_0 is the pre-exponential factor, Q the activation energy of diffusion, R is the Gas Constant and T the absolute temperature. The solubility of hydrogen isotopes in iron is given by Sievert's Law:

$$S = S_0 * \sqrt{p} * e^{-\frac{\Delta H}{RT}} \qquad 4$$

in which S_0 is a constant, p the partial pressure of the solute (in atmospheres), and ΔH the heat of solution. Values of the constants Q, D_0, S_0, and ΔH are assumed to be those of protium, ^1H, for pure iron [1]. The solubility, diffusivity, and permeability of tritium in the material of construction of the container is assumed to be the same as that of pure iron at moderate temperatures (ferrite, or alpha iron). This assumption is reasonably conservative because, in the limited number of systematic studies regarding diffusivity of hydrogen in iron-based alloys, the major alloying elements decrease or have no effect on hydrogen diffusivity [2].

Thermal calculations indicate that the maximum temperature in the interior of the container (within the spent rods) is 200° F, and the maximum temperature at the interior surface of the carbon steel permeation boundary is 175° F [3]. Results of permeation rates out of the container will be given for these two temperatures. Information from the project gives the residual tritium in each rod as 133 Ci. Each furnace basket is assumed to contain 300 rods, and for the first container disposed, three of the normal extracted baskets will be disposed of. (The fourth position in the container will be occupied by the container containing the unused Lead Test Assembly test TPBARs. Tritium

permeation will be calculated for this container separately and is not considered in this analysis.) Thus, the total tritium content of the container that can permeate the carbon steel is 133 Ci/rod * 300 rods * 3 extraction baskets = 119,700 Ci. The unoccupied fraction of the total container volume inside the carbon steel wall is assumed to be 20%. The free volume v^{free} is the container volume inside the permeation boundary multiplied by the free volume fraction- the free volume is the volume the hypothetical tritium gas occupies. The amount of tritium in cc at Standard Temperature and Pressure (STP) v^{stdcc} is the amount of tritium in Ci divided by 2.589 [4]. The hypothetical tritium partial pressure in the free volume *p* of the container is found by:

$$p = \frac{v^{stdcc} \cdot T}{v^{free} \cdot 273} \qquad 5$$

in which T is the absolute temperature (Kelvin). Note the partial pressure depends on the assumed temperature in Eq. 5.

The diffusivity (Eq. 3) and solubility (Eq. 4) are calculated using the assumed temperatures and partial pressures (Eq. 5). The permeability (Eq. 1) and the flux (Eq. 2) are then calculated for each permeation path (note the different paths have different thicknesses, Eq. 2). Finally, the total amount of tritium permeating each path is found by multiplying the surface area of the path by its flux. This calculation has been implemented as a spreadsheet that results in the calculation of the steady state permeation rate, expressed in Ci/year, for each of the three permeation paths.

The rapid permeation of tritium in this hypothetical scenario results in a declining tritium partial pressure inside the container. This occurs because permeation reduces the amount of tritium in the container, v^{stdcc}, and so the partial pressure is reduced, Eq. 5, thus reducing the solubility at the internal surface, Eq. 4. To account for this, a separate spreadsheet calculates the declining permeation rate for each permeation path for each year sequentially. A constant permeation rate is used during each year, and the rate is adjusted for the next year based on the reduction of tritium in the container each year. In addition, radioactive decay of tritium inside the container further reduces the tritium partial pressure. This decay is also applied in the spreadsheet, using the tritium half life 12.3 years [4].

Table I displays the yearly tritium permeation through each path, the total permeated each year, as well as the amount remaining before and after calculating the reduction from radioactive decay, assuming 200° F. Table II displays the same results for 175° F. All the residual tritium permeates out of the container in 17 years at 200 F and in 24 years at 175 F.

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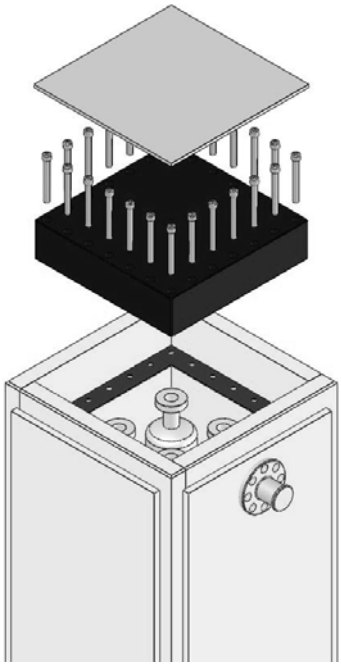

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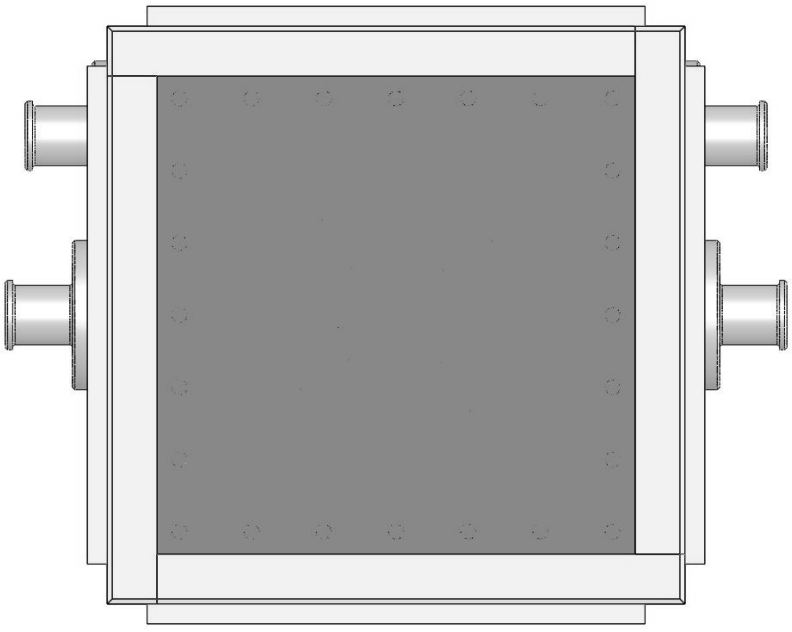

b.

Figure 1. Pictures of top of proposed waste container for CLWR-TEF: a. exploded view of upper radiation shield and outer seal lid; b. Top after welding outer seal lid.

200 deg. F Year	Sides & Bottom	Weld	Lid	Ci Permeated Each Year	Total Ci Remaining	Total Ci Decay Correct
0						119700
1	5273	407	4780	10459	109241	103255
2	4897	378	4439	9714	93540	88415
3	4532	350	4108	8989	79426	75073
4	4176	322	3785	8283	66790	63130
5	3829	295	3471	7596	55535	52492
6	3492	269	3165	6926	45565	43068
7	3163	244	2867	6274	36795	34778
8	2842	219	2576	5638	29140	27544
9	2529	195	2293	5017	22526	21292
10	2224	172	2016	4411	16881	15956
11	1925	149	1745	3819	12137	11472
12	1632	126	1480	3238	8234	7783
13	1345	104	1219	2667	5116	4835
14	1060	82	961	2102	2733	2583
15	775	60	702	1537	1047	990
16	479	37	435	951	39	36
17	92	7	83	182	0	0
18	0	0	0	0	0	0
19	0	0	0	0	0	0
20	0	0	0	0	0	0
21	0	0	0	0	0	0
22	0	0	0	0	0	0
23	0	0	0	0	0	0
24	0	0	0	0	0	0
25	0	0	0	0	0	0

Table I. Calculated yearly tritium permeation through sides, weld, and lid, total permeated each year, remaining tritium each year, and decay corrected remaining tritium each year. Temperature is 200° F. All tritium amounts in Ci (curies).

175 deg. F Year	Sides & Bottom	Weld	Lid	Ci Permeated Each Year	Total Ci Remaining	Total Ci Decay Correct
0						119700
1	3259	251	2954	6465	113235	107030
2	3082	238	2794	6113	100917	95388
3	2909	224	2637	5771	89616	84706
4	2742	212	2485	5438	79268	74924
5	2578	199	2337	5115	69809	65984
6	2420	187	2193	4800	61184	57832
7	2265	175	2053	4494	53338	50415
8	2115	163	1917	4196	46220	43687
9	1969	152	1785	3906	39782	37602
10	1827	141	1656	3623	33978	32117
11	1688	130	1530	3349	28768	27192
12	1553	120	1408	3081	24110	22789
13	1422	110	1289	2821	19968	18874
14	1294	100	1173	2567	16307	15414
15	1170	90	1060	2320	13094	12376
16	1048	81	950	2079	10297	9733
17	929	72	842	1843	7890	7457
18	813	63	737	1614	5844	5524
19	700	54	635	1389	4135	3908
20	589	45	534	1168	2740	2590
21	479	37	435	951	1639	1549
22	371	29	336	735	814	769
23	261	20	237	518	251	237
24	145	11	132	288	0	0
25	0	0	0	0	0	0

Table II. Calculated yearly tritium permeation through sides, weld, and lid, total permeated each year, remaining tritium each year, and decay corrected remaining tritium each year. Temperature is 175° F. All tritium amounts in Ci (curies).

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