Tritium Permeability of the Al Target Cell

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July 10, 2012

Introduction

The goal is to develop a safe tritium target cell for use in Hall A at Jefferson Lab for the 12-GeV electron beam experiments [1, 2] E12-10-103 and E12-11-112. Our overall philosophy for developing the conceptual design and safety devices has been to minimize the amount and density of tritium necessary for the experiment and to keep the systems and procedures as simple and reliable as possible. Also, the scattering chamber will serve as a secondary containment device. In this report we address the issue of tritium absorption and solid-state diffusion through the aluminum alloys of the target cell. Aluminum is one of the three metals considered suitable for tritium gas storage. With respect to solid-state diffusion, Al is considered superior to stainless steels due the low solubility and diffusivity of hydrogen in the metal and in the oxide layer that is invariably present on the surface.[3] The objective of calculations presented in this report is to estimate the maximum possible tritium permeation rate as a function of temperature in the cell consistent with established scientific principles and available data. Therefore, conservative assumptions are used throughout and all calculations are based on an extreme value on the least favorable side of the uncertainty range for each parameter.

Design of the target cell

The plan is to develop a tritium gas target for use in Hall A at Jefferson Lab during the 12-GeV electron beam era. The target cell would consist of 3.7 E+13 Bq (1000 Ci) of tritium gas in a completely sealed system. The tritium gas will be contained in 12.5 mm diameter aluminum cylinder, 250 mm long, made from a commercial alloy (AA 7075-T6 or AA 2024-T3511) as shown in Fig. 1. The aluminum windows for beam entrance and exit would be 0.254 mm (0.010 in) and 0.457 mm (0.018 in) thick, respectively, and the windows in the cylindrical body would be 0.457 mm (0.018 in) thick. These thin windows provide the dominant path for tritium diffusion from the primary containment cell. The net gas pressure in the container will be 1.42 MPa (14 atm) at room temperature and would double after all of the tritium decays into helium-3. However, the half-life for this decay is 12.3 y and the gas in not expected to be in the cell for more than one year. During operation of the electron beam, the target cell will be cooled with cryogenic helium gas (45 K). The electron beam current on the target cell will be limited to 25 μA and the temperature of the target cell windows should not exceed 180 K during operation. Thus, the highest temperatures the cell will be exposed to while containing tritium are the ambient temperatures of operation and storage.

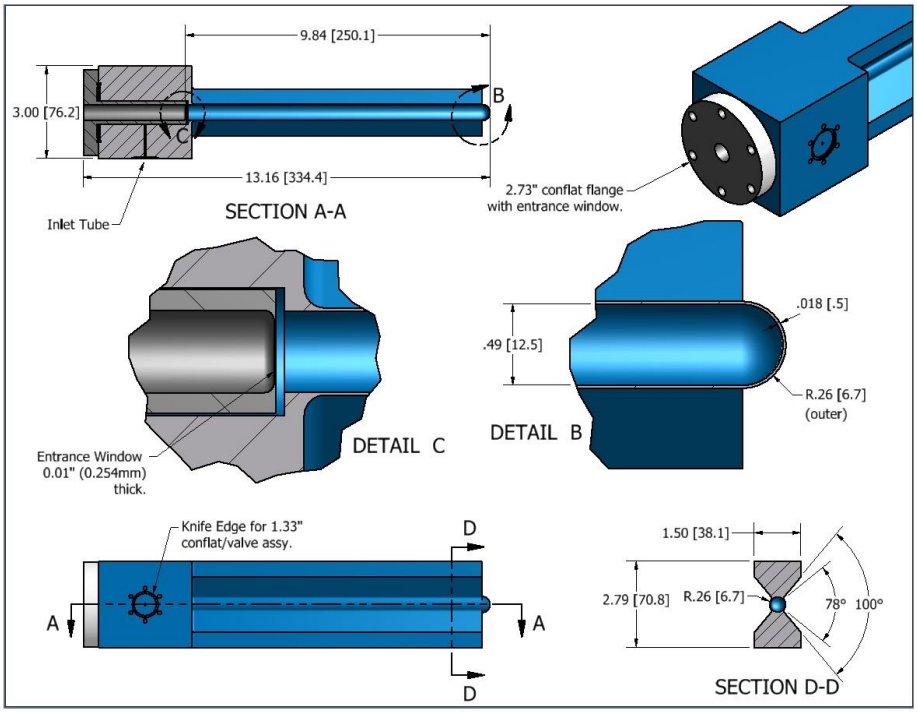

Fig. 1. Design of an individual tritium gas target cell for Jefferson Lab.

Hydrogen solubility in aluminum

A review[4] of hydrogen solubility, diffusion, and trapping in aluminum indicates significant disparity in the values reported for these quantities in the literature. It is believed that these discrepancies are primarily due to variations in surface oxides, surface reactions, and microstructural defects associated with variations in sample preparation and experimental conditions. The apparent hydrogen solubility as a function of temperature and pressure is given by

$$C_H = C_0 \, \mathrm{sqrt}(P/P_0) \exp(-\Delta H_s/RT) \qquad (1)$$

where P is the pressure, P_0 is the standard state pressure (1.0 atm), T is the temperature, ΔH_s is the heat of solution, and C_0 is a constant. Using eq. (1) to extrapolate elevated temperature hydrogen solubility measurements for pure aluminum to room temperature (25 °C) and a hydrogen pressure of 0.10 MPa (1 atm) indicates that the solubility of hydrogen in pure aluminum will range between 1.7 E-11 and 1.1 E-16 in terms of the unitless mole ratio of atomic hydrogen to aluminum ($X_{H/Al}$).[4,5] Taking the original solubility measurements from the literature cited in this review[4] finds 132 measurements made between 650 °C and 350 °C. Regression analysis of these collected data yields an estimated solubility at 25 °C and P = 1.42 MPa (14 atm) of log ($X_{H/Al}$) = -12.16 ± 0.19 with a correlation coefficient (r) of 0.94. The 95% confidence interval for this fit indicates that solubility of hydrogen in pure aluminum at this temperature and pressure is less than log ($X_{H/Al}$) = -11.78. However, this extrapolation assumes there is no fundamental change in the physical processes limiting solubility and that pure aluminum can be used to represent solubility in commercial alloys. Unfortunately, there is very little data on the solubility of hydrogen in solid Al alloys and the published data is on binary alloys. While commercial aluminum alloys contain a mass fraction of aluminum of over 90%, the existing data

indicates that alloying elements can increase hydrogen solubility by a factor of 2 or 3 over the concentration range of commercial alloys.[4] While a solubility increase with alloying element concentration may include hydrogen trapped at interfaces or intermetallic phases in the microstructure, it was decided that the very conservative (high) estimate for the solubility of hydrogen at room temperature (25 °C) and pressure (0.10 MPa), $X_{H/Al}$ = 1.0 E-09, will be used in subsequent calculations.

Hydrogen diffusion coefficient in aluminum

There is also a large variation in data reported for the diffusion of hydrogen in aluminum and these variations are attributed to the same factors influencing solubility measurements.[4] Most of these studies were performed at elevated temperatures and the results are extrapolated to room temperature using the Arrhenius relation:

$$D = D_0 \exp(-Q/RT) \qquad (2)$$

where Q is the effective activation energy for hydrogen diffusion and D_0 is the pre-exponential factor. Using the constants determined in these studies indicates that the diffusion coefficient for pure aluminum at 25 °C is between 2.5 E-11 m^2/s and 1.4 E-15 m^2/s if one assumes the measurements indicating a diffusion coefficient below 1.0 E-20 m^2/s at 25 °C were due to diffusion in the oxide becoming rate limiting.

The measurements more relevant to the present discussion are those for Al alloys 2024 and 7075. These measurements are summarized in Table 1. Gest and Troiano[6] studied hydrogen diffusion in AA 7075-T6 by galvanostatic permeation. However, Gest showed that these permeation fluxes were independent of foil thickness indicating that there could be a bias from surface effects. Braun *et al.*[7] also studied AA 7075-T6 by electrochemical permeation at room temperature and concluded that the diffusivity is two orders of magnitude faster, (2.3 to 6.0) E-11 m^2/s, than that reported by Gest and Troiano. Therefore, the faster diffusion rate end of the range reported by Braun *et al.*[7] will be used in subsequent calculations to represent the diffusion rate of hydrogen in aluminum alloys.

Table 1. Diffusion rates for hydrogen in Al alloys at room temperature.

Alloy (Al Assn. Dsgn.)	Experimental Technique	Diffusion Coefficient (m^2/s)	Reference
7075-T6	Permeation	2 E-13	Gest and Troiano
2024-T3	Potential Permeation	4 E-11	Braun *et al.*
7075-T6	Galv. Permeation	2.3 E-11	Braun *et al.*
7076-T6	Potential Permeation	2.5 E-11	Braun *et al.*
7075-T6	Current Pulse	6 E-11	Braun *et al.*

Tritium permeation from cell

The permeation rate of tritium from the cell will be dominated by the flux through the relatively thin windows. For the current design, the electron beam entrance window is the thinnest section being 0.254 mm (0.010 in) thick with an area of 0.0012 m^2 (1.2 cm^2) and the other windows are the 0.457 mm (0.018 in) thick with a total area of 0.00455 m^2 (45.5 cm^2). The window support consists of a 3.175 mm (0.125 in) thick walled tube with a surface area of 0.00436 m^2 (43.6 cm^2) that must also be taken into account. The permeation estimates will assume a temperature of 25 °C which is a conservative estimate since the target will be cooled with liquid nitrogen during operation. It is also assumed that solid-state diffusion of tritium through the metal is rate limiting, the interior surface of the metal is in equilibrium with the gas inside the cell, and the concentration on the outside surface is zero. Since both surfaces will be covered with a native aluminum oxide film with a lower solubility and diffusion rate than the metal, these are conservative assumptions.

The concentration of hydrogen per unit volume in aluminum, n_H, is given in terms of the solubility of hydrogen in aluminum, C_H, and the amount of aluminum per volume, n_{Al} as

$$n_H = n_{Al} C_H \tag{3}$$

Then, the permeation, J, of hydrogen through Al is given by

$$J = n_H D A/d \tag{4}$$

where A is the area of the thin windows and d is the window thickness of the Al. The results of permeation calculations assuming the fastest diffusion coefficient from Table 1 are given in Table 2. These calculations assume an activity for tritium of 1.07 E+15 Bq/mol (4.819 E-20 Ci/atom), a tritium concentration on the inside surface of $X_{H/Al}$ = 1.00 E-9, an aluminum concentration of 1.00 E+5 mol/m^3 (6.02 E+22 atoms/cm^3), a pressure in the cell of 1.42 MPa (14 atm), and that the tritium atom will diffuse at a rate that is $3^{-1/2}$ times that of the hydrogen atom.

Table 2. Maximum expected permeation rates for tritium from cell.

Cell Location	J mol(T)/s (atoms/s)	Activity Release Bq/s (Ci/s)
Entrance window	6.14 E-15 (3.7 E+9)	6.6 (1.8 E-10)
Exit windows	1.29 E-13 (7.7 E+10)	137.5 (3.8 E-9)
Window tube	1.77 E-14 (1.1 E+10)	19.2 (5.2E-10)
Total	1.53 E-13 (9.2 E+10)	163.3 (4.5 E-9)

At this permeation rate, the release of tritium to the secondary containment would be 2.1 E+8 Bq/y (5.6 mCi/y) from the upstream window, 4.3 E+9 Bq/y (120 mCi/y) for the 0.457 mm (0.018 in) thick windows, and 6.1 E+8 Bq/y (16.4 mCi/y) for the 3.174 mm (0.125 in) window tube support for an estimated total of 5.1 E+9 Bq/y (142 mCi/y) for permeation through the cell walls.

Conclusions:

The maximum possible tritium release rate from the target cell was estimated by reviewing the scientific literature on the solubility and diffusivity of hydrogen in aluminum, the assessment of the uncertainty range for each parameter, the selection of a value to represent the least favorable extreme, and conservative assumptions. The maximum total tritium loss from the target cell by permeation through the thin windows and window tube support was estimated at 5.1 E+9 Bq/y (142 mCi/y). Thus, it appears that the diffusion of tritium through the aluminum target windows will not be problematic in terms of tritium release. The leak rate from the two seals is estimated to be 2.47 E+9 Bq/y (66.8 mCi/y) and from the valve as 1.12 E+10 Bq/y (304 mCi/y).

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