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**A MODEL FOR TRITIUM AND DEUTERIUM TRANSPORT IN
TARGETS FOR NEUTRON GENERATORS**

C. M. Logan

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LAWRENCE LIVERMORE LABORATORY
University of California, Livermore, California, 94550

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A MODEL FOR TRITIUM AND DEUTERIUM TRANSPORT IN TARGETS FOR NEUTRON GENERATORS

Abstract

The observed behavior of titanium tritide targets used for 14-MeV neutron generation indicates that two mechanisms for tritium motion are active. A model is proposed based on thermally induced motion of tritium and deuterium in gamma

phase hydride plus a radiation-induced component dependent on the incident ion current density. The model describes the observed behavior and permits speculation regarding target life under different operating conditions.

Introduction

There is increasing need for intense sources of 14-MeV neutrons. Two major applications are for cancer therapy and for materials studies related to D-T fusion reactors. Machines to generate these neutrons utilize the fusion of deuterium and tritium. Usually the tritium is contained in some type of target while deuterium ions are accelerated and directed into the tritium-bearing target to produce the D-T fusion, although systems have been suggested which would accelerate the tritium instead. In order to achieve high flux densities in small samples, it is necessary to have as the neutron source a target design which tolerates intense ion beams of small diameter and permits sample placement near it.

The most successful source of this type is the Lawrence Livermore Laboratory's rotating target. In normal use, atomic deuterium ions are accelerated to 400 keV and impinge on a rotating target.

The beam is typically less than 1 cm in diameter and contains about 15 mA of deuterons (~6 kW of beam power). This power is deposited in the front 3 μm of target thickness, and the heat it produces is conducted to the backing and removed by a flow of cooling water over the back surface. The normal target has a 10- μm (4.5-mg/cm²) vapor-deposited layer of titanium tritide on a copper alloy backing. The tritium content of a new target approaches 1.5 tritium atoms per titanium atom. Such targets contain a very high density of tritium atoms. A fully stoichiometric compound of TiT_2 has a tritium atom density twice that of liquid tritium at 20°K.

There are many innovative features of the cooling and rotating systems of the LLL neutron source which have been described elsewhere.¹⁻⁴ In this paper I intend to describe the observed behavior of the target used in this source and other similar systems and to formulate a

tritium-transport model which will explain the behavior and hopefully permit

prediction of target performance under different operating conditions.

The Target Environment

The gas pressure at the front target surface (on the beam side) must be about 10^{-5} torr or less to permit beam transport to the target. Early proposals for solid targets assumed that the target temperature could not be permitted to exceed about 250°C, and indeed for stationary, steady-state operation, target life is severely shortened by such temperatures. This target, because of its rotation at 1100 rpm, is subjected to a repeating cycle of a rapid temperature rise as it rotates through the beam, a rapid quench as the heat is conducted to the copper backing, and a gradual cooling as the target rotates around to the beam spot again. I have modeled the system in a one-dimensional transient heat transfer calculation using a finite difference code at LLL. For a given set of assumptions representative of the most severe operating conditions, the calculated temperature history is displayed in Fig. 1. The peak temperature in the spike is seen to be considerably above the supposed limit, but does not stay there for very long. The calculation of the base temperature at the end of a cycle is uncertain because of uncertainty in the heat transfer to the water. The accuracy of the calculated temperature rise in the peak is limited mostly by uncertainty about the beam power density profile, and is expected to be representative of a current of 15 mA incident with full width at half maximum (FWHM) of 0.3 cm.

The bombarding deuterons lose energy to electrons as they move through the target material and come to rest at a depth of about 1.7 mg/cm^2 . An estimate of the implanted distribution (before diffusion) is shown in Fig. 2. As the deuterons move through the titanium-tritium layer, about one in 3×10^5 reacts with a tritium atom and creates a 14-MeV neutron. Some deuterons interact with titanium atoms and cause energetic recoils which then can cause further displaced atoms. This "radiation damage" from ion bombardment is most severe near the end of the deuteron range and will result in each titanium atom

Fig. 1. Calculated front-surface temperature vs time for the LLL rotating target.

being displaced several hundred times over the lifetime of the target. I have written a computer code to calculate this damage rate from Rutherford scattering as a function of deuteron energy in titanium. From range-*vis*-energy data⁵ it is then possible to determine the damage rate as a function of depth in the target. This is also given in Fig. 2. These values become uncertain at low energy because of energy spread in the moving deuterons. I have used energy loss and range data from Ref. 5 for metallic titanium in this paper. A more precise approach would use weighted

data for titanium and tritium, but this would make little difference for our purposes, and in view of the uncertainty in the data it is not appropriate.

Surface recession from ion sputtering should be insignificant. A sputtering ratio (atoms lost per incident ion) of about 10^{-3} would be expected⁶ and would remove about 0.01 mg/cm² for each mA-hr/cm² of exposure. While surface erosion is not an important consideration, sputtering would effectively remove surface oxide or carbon deposits and could change surface morphology.

Fig. 2. Neutron yield, displacement damage, and deuteron implantation vs depth in target, for 400-keV deuterons.

Neutron Yield vs Depth

The variation of the $T(d,n)\alpha$ reaction cross section: $\sigma(E)$ with deuteron energy is well known.⁷ The neutron yield $Y(E)$ in the target at any energy is given by

$$Y(E) = K \frac{\sigma(E) N_T}{dE/dx}, \quad (1)$$

where E is the energy of the moving deuteron, dE/dx is its rate of energy loss, and N_T is the tritium density. For a con-

stant tritium density, the relative neutron yield as the deuteron moves through the layer is given in Fig. 2. Tritium deeper than about 1.5 mg/cm^2 does not produce neutrons, and that contained near the front surface is relatively ineffective. This distribution is for a 400-keV incident energy. A lower incident energy would in effect remove the front portion of this distribution.

Observations of Target Parameters and Target Behavior

I know of no direct observations or measurements of tritium profiles, deuterium profiles, or density profiles of used targets. The available evidence comes from observed neutron yields under various circumstances.

If we consider a titanium layer with 1.5 tritium atoms per titanium atom, there would be about 7×10^{22} tritium atoms per cm^3 . Since the useful range is $3.3 \times 10^{-4} \text{ cm}$, there are about 2.5×10^{19} tritium atoms per cm^2 available over the useful range. If we use a target until its yield drops to about 70-80% of that of a new target we have implanted in it about 5.0 mA-hr/cm^2 of deuterons, which is several times as many deuterium atoms as the number of tritium atoms originally available.

The burnup of tritium from D-T reactions is negligible. About one atom in 5000 over the first 1.5 mg/cm^2 of target material is utilized. However, tritium is lost from the target, and observations of the accumulation in the vacuum pumping system indicate that between 0.1 and

0.2 of a tritium atom are emitted for each incident deuteron.² There has been one poorly documented observation of the gas evolution from an operating target.⁸ A mass spectrograph indicated a nearly constant gas flow of molecular hydrogen. It was initially dominated by T_2 , then changed to mixed DT, and finally was dominated by D_2 near the end of the useful target life.

Careful observations of the Livermore system indicate a slight rise in neutron yield for a short time when starting up with a fresh target. This is followed by a period of fairly rapid decay and a second period with more slowly varying yield.⁴ Figure 3 is reproduced from Ref. 4. The reference to small and large radius applies to the target. The target velocity is about a factor of two higher at the large radius. This decay with two separate time constants has also been observed at other laboratories⁹ for fixed (low-intensity) targets using lower bombarding energies.

Figure 4 (from Ref. 2) shows the neutron yield for various bombarding

energies for fresh and used targets. The fresh target shows a yield consistent with uniform tritium concentration over the 1.5-mg/cm^2 useful range. The measured yield at 200-keV bombarding energy would be sensitive to any tritium depletion near the front surface, and the measurements indicate that any surface layer containing no tritium must be less than $0.3\text{ }\mu\text{m}$ thick. Other laboratories have also observed surface depletion in $0.1\text{-}\mu\text{m}$ layers.¹⁰ After some use at 400-keV bombarding energy, the target yield has decreased. If at this point the bombarding energy is increased, the yield decreases further. After prolonged use at

Fig. 3. Neutron yield decay curves for various thicknesses of the titanium tritide layer at the front of the target. Yield is plotted as a function of cumulative bombardment.

Fig. 4. Neutron yield vs deuteron energy for targets after various exposures to 400-keV deuterons.

400 keV this phenomenon is so pronounced that decreasing the bombarding energy actually increases the yield. An estimate of tritium profiles which would account for this behavior is given in Fig. 5. This constant reduction factor for yield at 200 and 300 keV is a strong indication that no steep gradients exist in the first 1.0 mg/cm^2 of target material, and the

Fig. 5. Tritium concentration vs depth in target for various target use histories.

sharp loss of yield for 450-keV deuterons indicates that a severe depletion occurs between 1.0 and 1.5 mg/cm² from the front surface.

Studies have been made^{4,11,12} on the effect of the thickness of the titanium layer. There is little effect on initial yield so long as the layer thickness is at least equal to the deuteron range. However, the life of the target is improved by thicker titanium layers up to thicknesses about 2.5 times the deuteron range. Thicknesses greater than this decrease the useful target life.

Small static targets have been bombarded under various cooling conditions.^{11,13} These targets experience a constant

temperature while being bombarded. They fail quickly at 253°C, have an acceptable lifetime at 115°C, and a somewhat better lifetime (decay rate reduced ~30%) when Freon-cooled to below room temperature. For the Livermore rotating target system, by contrast, cooling with chilled water results in no observable change in target life.¹⁴

Some attempts have been made to run targets of this type with mixed beams of atomic and diatomic ions. The neutron yield drops very rapidly under these conditions.¹⁴ No effect on target life is observed from the elimination of oil diffusion pumping,¹⁴ indicating that surface

carbon deposits are not present or do not influence target life.

When targets are made with thicker titanium layers, the tritium loading that

can be achieved increases and in some cases approaches stoichiometry for layers thicker than 10 mg/cm^2 (Refs. 15 and 16).

The Titanium-Hydrogen System

Most of the literature on the titanium-hydrogen system has dealt with low hydrogen concentrations. The phase diagram shows that for equilibrium below about 300°C two phases exist. The alpha phase of titanium is hexagonal-close-packed. The solubility of hydrogen in the alpha phase is about 0.0005 atom fraction at room temperature¹⁷ and reaches a maximum of approximately 0.08 near 300°C . The gamma phase compound TiH_2 has a face-centered-cubic fluorite-type structure and exists with a composition as low as

50-60% hydrogen. No appreciable difference is known to exist for the titanium-deuterium or titanium-tritium systems.^{16,18}

Impurities have been observed to have dramatic effects on phase boundaries and solubilities. Oxide layers are apparently formidable barriers to hydrogen diffusion.¹⁸

Since the temperature exceeds 300°C for only very short times and interesting tritium concentrations are greater than 30%, we are probably always dealing with a mixture of alpha and gamma phases or pure gamma.

A Proposed Mechanism for Target Degradation

The evolution of tritium and deuterium from targets of this type cannot be surface-limited. One would not expect a surface layer (such as oxide) to survive the displacements and sputtering caused by the deuterons. If a surface barrier existed with relatively easy internal diffusion, no appreciable hydrogen concentration gradients could exist. We observe that the implanted deuterium must move rather slowly and that it displaces tritium at the implantation depth.

The loss of tritium is partially thermally activated. The rate of target decay is dependent on temperature for other conditions being equal, but targets also show yield drop when used at room temper-

ature. These targets can be stored for prolonged periods at room temperature without yield loss, so no appreciable thermal degradation occurs at that temperature. Part of the titanium loss must occur from a mechanism like radiation-enhanced (nonthermal) diffusion.

There is a strong experimental and theoretical basis¹⁹ for describing the jump frequency of a hydrogen atom in the gamma phase by the expression

$$\nu = \frac{1}{2}(2-X)(36 \times 10^{12}) \exp(-0.37/kT), \quad (2)$$

where X is the number of tritium atoms per titanium atom. The hydrogen atoms occupy the tetrahedral sites in the fcc

lattice. When $X = 2$ all the tetrahedral sites are occupied. Since no thermally created vacancies are present the jump frequency is proportional to the fraction of vacant sites, $\frac{1}{2}(2 - X)$.

I suggest that the incident deuterons can modify the jump frequency in two ways: 1) The collision cascades create vacancies not related to the tritium content so that the concentration of vacant sites also depends on the average incident deuteron current density ρ_d . 2) The kinetic energy of the cascade also causes "jumps" which are not thermally activated and which have a frequency proportional to current density. With these two considerations the expression for jump frequency becomes

$$\nu = \left[\frac{1}{2}(2 - X) + K_1 \rho_d \right] (36 \times 10^{12}) \times \exp(-0.507/kT) + K_2 \rho_d \quad (3)$$

Since for the cases of interest X never approaches 2 and the lifetime of a collision vacancy is short, $K_1 \rho_d \ll \frac{1}{2}(2 - X)$ and we can neglect the contribution from the $K_1 \rho_d$ term.

A very rough estimate of the $K_2 \rho_d$ term can be made from several observations. These targets can be soldered at 100°C without serious damage,¹³ yet they lose tritium when used at -20°C. The average current density on the LLL rotating target is similar to that used on fixed targets at about $30 \mu\text{A}/\text{cm}^2$. A very rough guess is that at a temperature of 127°C the thermal jump frequency and the deuteron-induced jump frequency are equal. If we let $X = 1.2$, $\rho_d = 30 \mu\text{A}/\text{cm}^2$, $T = 400^\circ\text{K}$, and evaluate Eq. (3) for equal contributions, $K_2 = 2 \times 10^5$ (jumps/sec/ $(\mu\text{A}/\text{cm}^2)$). This value seems very large

when compared to the energy deposition. If all the energy of each 400-keV deuteron were transferred to tritons requiring 1/2 eV to jump, we would have 8×10^5 jumps induced by each incident deuteron. Since $1 \mu\text{A}/\text{cm}^2$ is 6×10^{12} deuterons/ $\text{cm}^2\text{-sec}$ and there are 3×10^{19} tritons/ cm^2 over the deuteron range, about 0.2 jump/sec (for each atom) would be induced for each $\mu\text{A}/\text{cm}^2$ of deuterons — only one millionth of the value $K_2 = 2 \times 10^5$ that was estimated above by Eq. (3). Since very small energy transfers may cause jumps, and since a 1/2-eV atom may jump many times, this large value is not energetically impossible. It may be this high because some of the energy lost to the electrons in the compound effectively reduces the activation energy for motion of tritium.

The expression for jump frequency is then

$$\nu = 1.5 \times 10^{13} \exp(-0.507/kT) + 2 \times 10^5 \rho_d \quad (4)$$

This is plotted vs $1/T$ in Fig. 6 for $\rho_d = 30 \mu\text{A}/\text{cm}^2$.

We can see from this that for this model there is little point in attempting to lower the target temperature below 80°C unless the purpose is to limit the temperature excursion in the peak or to operate in a particular temperature range required by the cooling fluid used.

The diffusion coefficient is proportional to the jump frequency so that an appropriate measure of diffusion-induced motion is $(\nu t)^{1/n}$, where t is the time and n is between 1 and 2. For a given temperature history it is then possible to define an average value of $(\nu t)^{1/n}$ called Δ which is

$$\Delta \equiv \frac{\int (v t \, dt)^{1/n}}{\int (dt)^{1/n}}. \quad (5)$$

This value is the appropriate figure of merit for a given temperature cycle. For simple time cycles we can evaluate this expression and we see that for $n = 2$, 10^{-3} sec at 1000°K causes the same diffusion-controlled motion as 4 sec at 400°K . More complicated cycles require computer evaluation.

The value of n depends on the way concentration gradients change. For a "diffusion couple" where the concentration is initially discontinuous, the appropriate value for n is 2. For steady-state equilibrium flow (constant gradients) the flux is proportional to $v t$ and n should be 1.

Fig. 6. Jump frequency of a tritium atom as a function of inverse temperature.

Discussion

There is a widespread impression that tritium in these targets diffuses forward from regions beyond the range into the useful part of the target material.^{3,9,11} The evidence cited for this is that lowest decay rates are observed when the tritiated layer thickness is about 2.5 times the deuteron range.

I believe, however, that such resupply does not occur in any significant amounts and that tritium in the target beyond the deuteron range is probably still there when the target is discarded.

Deuterons are implanted at about 1.7 mg/cm^2 depth with a spread of only about 0.1 mg/cm^2 (for 400-keV incident energy), so in a very short time there is an enormous hydrogen concentration peak at this depth. We have shown that this large driving force does drive tritium from this

region toward the forward part of the target, which results in the initial small increase in neutron yield. Tritium also will be driven toward the back of the target, and in order for this tritium to resupply the front regions it will have to diffuse up a very large concentration gradient. But this cannot occur. Thus it appears that the region in the target beyond the deuteron range improves target life by serving as a dump for excess deuterium, thereby reducing the concentration gradient which drives tritium toward the front surface. If a target is used with the backing material just beyond the deuteron range, the deuterium piles up at the interface, doubles the concentration gradient, and results in rapid target deterioration. The oxide at the interface is a very effective diffusion barrier, and

the solubility of hydrogen in the copper matrix is about 10^{-6} at 700°K .²⁰ If on the other hand the titanium tritide layer is very thick, it increases the target temperature because of its low thermal conductivity ($\sim 4\%$ that of copper), and target life is again shortened.

The decrease in target decay rate after some usage is usually ascribed to the loss of all the tritium that is not "bound" in the delta phase. The very low solubility of hydrogen in the alpha phase obviously precludes this explanation, since losing all of the "dissolved" tritium would not result in an observable yield drop. I believe the correct explanation for the observed behavior is the gradual achieving of equilibrium concentration gradients, so that the effective value of the parameter $1/n$ in Eq. 5 changes from $1/2$ to 1.

This discussion has assumed that hydrogen motion in gamma-phase titanium tritide is the limiting mechanism for target behavior. Since some alpha-phase titanium

metal may be present it may influence the model. Hydrogen diffuses much more readily in alpha titanium than in the gamma phase.²¹ Further, hydrogen solubility is very low in the alpha phase so that no appreciable deuterium or tritium inventory resides there. I believe that the presence of the alpha phase does not compromise the validity of this model, but it may make it impossible to calculate the actual concentrations present in a target. A continuous alpha matrix would serve as a "pipeline" for deuterium and tritium which had escaped from the gamma phase. This may create a mechanism for discharging deuterium from deep in the target without "flushing out" the tritium in the useful region.

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