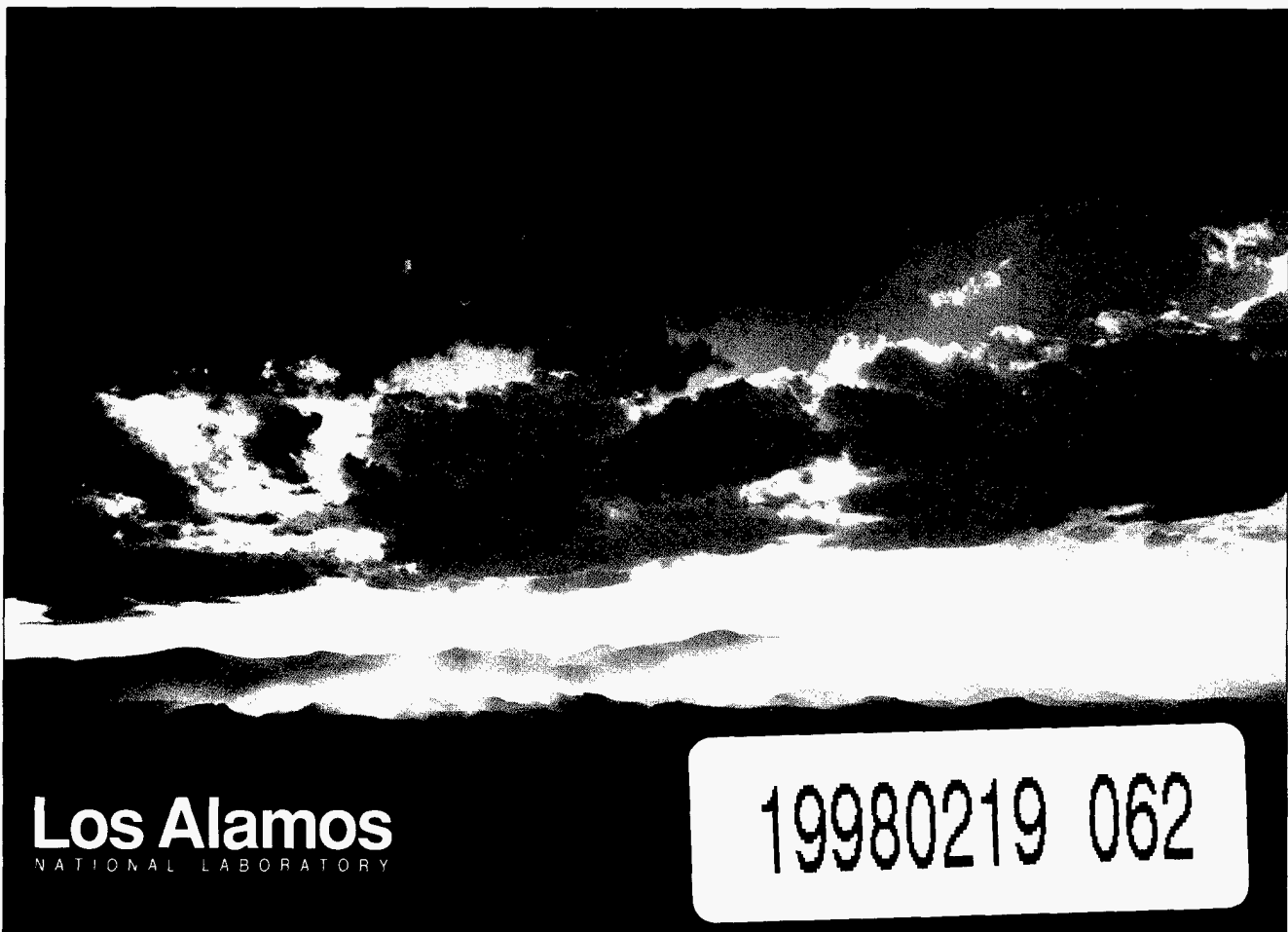LA-UR-97-2464
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TRITIUM IMPLANTATION IN THE ACCELERATOR PRODUCTION OF TRITIUM DEVICE

by

Russell B. Kidman

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Los Alamos
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TRITIUM IMPLANTATION IN THE ACCELERATOR PRODUCTION OF TRITIUM DEVICE

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ABSTRACT

We briefly describe the methods we have developed to compute the magnitude and spatial distribution of born and implanted tritons and protons in the Accelerator Production of Tritium (APT) device. The methods are verified against experimental measurements and then used to predict that ~16% of the tritium is implanted in the walls of the APT distribution tubes. The methods are also used to estimate the spatial distribution of implanted tritium, which will be required for determining the possible diffusion of tritium out of the walls and back into the gas stream.

I. INTRODUCTION

In the current Accelerator Production of Tritium (APT) concept, tritium is produced when spallation neutrons are captured by the ^{3}He gas that is circulated throughout the APT in aluminum distribution tubes. Ideally, the materials in the APT (lead, tungsten, water, aluminum, and stainless steel) are arranged to quickly slow the neutrons to thermal energies where ^{3}He has a high capture cross section. When a thermal neutron is captured by ^{3}He, a 191 KeV triton and a 573 KeV proton are produced. Depending on where these tritons and protons are born, they may have enough energy left to implant themselves in the aluminum tube walls. The magnitude and spatial distribution of the born and implanted tritons and protons are the subjects of this paper.

We proceed by intoducing the methods that we have developed to predict the production and implantation distributions of the tritons and protons in any arbitrarily shaped container. We then proceed to test the methods against the experimentally measured tritium implantation in several different containers. Finally, the methods are used to predict tritium implantation losses, distributions, and mitigation effects for the APT.

II. METHODOLOGY

In this section, we briefly describe the analytical and numerical methods we use to compute the fraction of tritons implanted in the walls of various containers.

A. Analytic Implantation

We begin by defining D as the critical distance a triton will have to travel in the container gas before it can no longer become implanted in the container walls. The critical distance is a function of the triton energy and the type and pressure of the gas in the container. In general, the critical distances are equivalent to the stopping lengths or ranges of tritons in gasses given by the computer code TRIM.[1]

To have some flexibility in testing the effects of different spatial distributions of triton production, we will assume that triton production, p(r), is given by :

$$p(r) = a + br + cr^2 \text{ tritions/cc} , \tag{1}$$

where r is the distance from a tube axis or sphere center, and a, b, c are constants. Different spatial distributions are invoked by simply adjusting the constants a, b, c.

With the above assumptions for triton production distribution and trition range, and with the assumptions of isotropic trition emission and a straight line slowing down, it can be shown that the fraction, F_{cyl}, of tritons implanted in the tube walls of an infinitely long cylinder is given by

$$F_c = \frac{D\left(\begin{array}{c}120aR + 120bR^2 + 120cR^3 - 40bRD \\ -80cDR^2 + 5bD^2 + 30cRD^2 - 4cD^3\end{array}\right)}{40R^2\left(6a + 4bR + 3cR^2\right)}, \tag{2}$$

where R is the radius of the cylindrical tube. The integrations necessary to derive Eq. (2) could not be performed exactly, and it was necessary to neglect terms $\leq D^2/R^2$. This is not a bad assumption because for any practical case $D \ll R$. The assumption of straight line slowing down is also reasonable because the slowing down is almost entirely electronic and not neutronic. Note that for the case of homogeneous triton production $(b=c=0)$, the fraction of tritons lost becomes the useful and easily remembered formula

$$F_c(\text{for homogeneous production}) = \frac{D}{2R} = \frac{\text{triton range}}{\text{tube diameter}}. \tag{3}$$

Under the same above conditions, it can be shown that the fraction, F_{sph}, of tritions implanted in the walls of a spherical container is given by

$$F_s = \frac{\left(\begin{array}{c}\frac{a(R^3-[R-D]^3)}{6} - \frac{a(R^2-D^2)(R^2-[R-D]^2)}{8D} + \frac{a(R^4-[R-D]^4)}{16D} \\ +\frac{b(R^4-[R-D]^4)}{8} - \frac{b(R^2-D^2)(R^3-[R-D]^3)}{12D} + \frac{b(R^5-[R-D]^5)}{20D} \\ +\frac{c(R^5-[R-D]^5)}{10} - \frac{c(R^2-D^2)(R^4-[R-D]^4)}{16D} + \frac{c(R^6-[R-D]^6)}{24D}\end{array}\right)}{\frac{aR^3}{3} + \frac{bR^4}{4} + \frac{cR^5}{5}}. \tag{4}$$

These analytic formulas are very useful for predicting the triton implantation in containers that approximate long tubes or spheres. Analytic derivations for other shapes, even finite length tubes, are very difficult and complicated.

B. Numerical Implantation

While contemplating containers of odd shapes and odd production distributions, another method of computing implantation was developed. It involves numerical Monte Carlo calculation with a code like MCNP.[2]

Implantation is modeled by setting up the exact geometry of the container and simulating the initial triton source distribution with neutrons of an appropriate energy and with the correct spatial and directional distributions. No materials are used. Rather, the interior of the container is given an importance of 1, while the exterior of the container is given an importance of 0. The key to this method is to give the neutrons a cutoff time that just allows the neutrons time to travel the triton range before they are killed. Because of their spatial and directional distributions, the neutrons can have different path lengths in the interior of the container before they are killed (which represents tritons stopped in the container gas) or escape the container surface (which represents tritons that reach the container surface). A tally on the container surface will count only the neutrons that reach the surface and represents the tritons that are implanted in the container wall.

The sophisticated MCNP geometry and source modules make it easy to model even complicated containers and triton source distributions. Because there are no neutron interactions with materials in this method, millions of neutrons can be run in 2–3 minutes, which usually gives adequately small statistical uncertainties. Variations in container geometry, gas loading, triton range, and spatial and directional distributions are readily investigated. This numerical method has been verified by reproducing the results for tubes and spheres that have the above analytic solutions. It is a very practical method for estimating triton or proton implantation losses.

C. Implantation Distributions

To investigate the subsequent buildup or diffusion of implanted tritons, it is necessary to estimate the initial spatial distribution of the tritons that are implanted in the container walls.

An extension of the numerical method introduced above solves this problem for us. Instead of modeling the container out to just the interior surface, the "gas" interior is extended 1 triton range further. Several tally surfaces are inserted between the original interior surface and the new extended surface. The difference between two adjacent surface tallies represents the tritons implanted in that shell. After MCNP has been run to obtain the shell tallies, the surface distances beyond the original interior container surface are then shrunk by the ratio of the triton range in the wall material to the triton range in the gas material. The resulting shell positions and volumes are combined with the shell tallies to give the desired spatial distribution of tritons in the container wall.

III. EXPERIMENT

A tritium production experiment has been carried out at Los Alamos National Laboratory in which the tritium implantation has been measured in numerous spheres, capsules, and tanks filled with various gas mixtures at various pressures. We have used our methods to calculate these same implantation losses, and the results

are shown and compared with the measurements in Table 1. All calculations assumed that the tritons were born with a kinetic energy of 0.191 MeV (i.e. from ^{3}He capture of thermal neutrons), and except for the spheres, all calculations assumed that the triton production was spatially uniform within the containers. In general, our predictions are only ~10% lower than the measured implantations.

The theoretical predictions for several capsules were only about 60% of the measured values. We suspect that the energy of the triton-producing neutrons in these capsules could be above thermal. This would give the tritons more than 0.191 MeV of kinetic energy, which would give them a better chance of becoming implanted. This could explain part of the discrepancy. When the neutron flux in these capsules becomes available, the theoretical predictions will be upgraded by using more appropriate triton energies.

Because experimental results for the spheres were not yet available, the situation was used as an opportunity to make some theoretical predictions before measured results are known. Figure 1 shows the calculated triton production density as a function of radius for the ^{3}He Sphere 2 container. This profile was used to compute the 12.1% triton implantation loss shown in Table 1. If a flat (uniform) profile were used, the predicted loss would be 9.2%. Figure 2 shows the predicted triton and proton implantation distributions in the aluminum wall of Sphere 2. The effect of uniform and nonuniform (Fig. 1) source distributions on these implantation distributions is also shown in Fig. 2. The proton results were included because they may affect the wall matrix, which in turn could affect the subsequent diffusion of tritons in the wall material. The protons were assumed to be born with a kinetic energy of 0.573 MeV.

Implantation estimates for the real-time ^{3}He half cylinder show the utility of the numerical method. The main part of the tank is a half cylinder, which was easy to model on MCNP and showed that 31.5% of the tritons were implanted. However, the half-tank also had sizable supply lines. These supply lines were easy to add to the MCNP model and the overall implantation loss was 33.3%. An analytic derivation of these same results would have been very difficult.

IV. APT

The size and pitch of the ^{3}He distribution tubes will vary from region to region in the APT. The exact geometries have not been set; however, a reasonable set of distribution tube diameters has been assumed for the

various tritium-producing regions to use our methods to estimate the tritium implantation in the APT. The results are shown in Table 2. The first column shows the percent of total tritium produced in each of the five main tritium producing regions of the APT. The second column shows the distribution tube diameter assumed for each region. The third column is the result of applying our implantation methods, and for each region shows the fraction of tritium implanted in the distribution tube walls. (For these calculations it was assumed that tritium production was radially flat in the distribution tubes, that the initial triton energy was 0.191 MeV, that the ^{3}He gas was at 100 psia and 305 K, and that the resulting triton range was 0.257 cm.) The fourth column simply multiplies columns 1 and 3 to give the percent of total tritium production lost in each region. The overall result is that ~14.7% of the tritium is implanted in the aluminum tube walls. Allowing for the 10% discrepancy mentioned above, we estimate that ~16% of the tritium is actually implanted in the aluminum tube walls.

The radially flat production assumption of Table 2 is particularly suspect. This was investigated with a detailed simulation of a 2.3622-cm diameter distribution tube in the decoupling region. Figure 3 shows the resulting radial profile of tritium production. Obviously, the production distribution is not radially flat. Triton and proton wall implantation fractions with an assumed flat radial tritium production density are 0.109 and 0.342, respectively; however, they become 0.119 and 0.355, respectively, with the nonflat radial profile of Fig. 3. Thus, the decoupling fraction loss of Table 2 should probably be increased to 0.127 to account for the nonflat radial production profile in that region. The production profiles in the other tritium producing regions of the APT have not yet been determined.

The radial deposition of tritium within the gas and wall of a distribution tube is of great interest and can be calculated with the numerical methods developed in this paper. To demonstrate, we choose a tube located at the peak tritium production location in the very-high-power-lead blanket. At this location, the average tritium production rate is 2.95e13 tritons/cc/s, which we will assume is radially uniformly distributed. The 1.4097-cm diameter tube is filled with ^{3}He gas at 100 psia and 305 K, which gives the 0.191 MeV tritons a range of 0.257 cm. The range of 0.191 MeV tritons in aluminum is 1.765e-4 cm. With these assumptions, our numerical method gives the radial distribution of the triton deposition rate in the tube gas as shown in Fig. 4. Note how the deposition rate begins to fall below the production rate just where tritons are born within range

of the tube wall. The radial distribution of the triton implantation rate in the tube wall is shown in Fig. 5.

The fate of the implanted tritium is not yet known. Do the walls act like a sponge and continuously absorb and retain the implanted tritium, or does the implanted tritium build up to a certain level and then diffuse back into the gas or through the wall to the water coolant? These questions form the next phase of this work. In case tritium does not diffuse back into the gas, there are some mitigation measures that can be used to reduce tritium implantation. Figure 6 shows how implantation is reduced as one uses larger distribution tubes. Figure 7 shows how implantation is reduced by increasing the gas pressure by adding various heavy gasses to the ^{3}He gas. Of course, one is not totally free to implement such measures because they usually come with an engineering tradeoff penalty elsewhere in the APT.

ACKNOWLEDGMENT

REFERENCES

1. James F. Ziegler, *TRIM - The Transport of Ions in Matter*, IBM Research code version 95.06, Yorktown Heights, New York (January 9, 1995).

2. Richard E. Prael and Henry Lichtenstein, "User Guide to LCS: The LAHET Code System," Los Alamos National Laboratory Report LA-UR-89-3014 (September 1989).

TABLE 1
THEORETICAL TRITON-IMPLANTATION ESTIMATES FOR THE T/P EXPERIMENT
(Assuming initial triton kinetic energy = 0.191 MeV)

Container	Radius (cm)	Length (cm)	Pressure (Torr)	Triton Range (cm)	Theoretical Percent of Tritons Implanted	Measured Percent of Tritons Implanted	Calculated/ Measured
SPHERES:							
2	8.274		1241.2	1.018	12.1		
3	8.274		1241.2	1.018	12.1		
4	8.274		1241.2	1.018	12.1		
5	8.274		51.7	24.425	100.0		
6	8.274		1241.2	1.018	12.1		
REAL-TIME HE HALF-CYL:							
Without supply lines	7.493	32.766	611.3	2.633	31.5		
With supply lines	7.493	32.766	611.3	2.633	33.3		
CAPSULES:							
H157 (B-1)	1.22936	2.4638	770.9	1.652	78.4	90.4	.87
H153 (B-3)	1.22936	2.4638	770.9	1.652	78.4	89.8	.87
H154 (C-3)	1.22936	2.4638	770.9	1.652	78.4	89.8	.87
H151 (C-6)	1.22936	2.4638	770.9	1.652	78.4	89.8	.87
H152 (D-6)	1.22936	2.4638	770.9	1.652	78.4	89.6	.88
H159 (D-11)	1.22936	2.4638	770.9	1.652	78.4	89.6	.88
Xe22 (B-2)	1.22936	2.4257	763.2	1.458	72.0	81.7	.88
Kr73 (B-4)	1.22936	2.4638	761.4	1.571	75.8		
Kr80 (B-6)	1.22936	2.4638	761.4	1.571	75.8	86.0	.88
Kr79 (C-1)	1.22936	2.4638	761.4	1.571	75.8		
Kr76 (D-4)	1.22936	2.4638	761.4	1.571	75.8		
Kr75 (D-9)	1.22936	2.4638	761.4	1.571	75.8	86.9	.87
He11 (B-5)	1.22936	2.4257	2287.0	.562	32.1		
He12	1.22936	2.4257	2287.0	.562	32.1		
Ar117 (C-2)	1.22936	2.4638	589.4	1.478	72.5		
Ar119 (C-5)	1.22936	2.4638	589.4	1.478	72.5	75.3	.96
Ar120 (C-7)	1.22936	2.4638	589.4	1.478	72.5		
Ar118 (D-5)	1.22936	2.4638	589.4	1.478	72.5	75.7	.96
Xe36 (C-8)	1.22936	2.4257	762.8	1.456	72.0		
Xe33 (C-11)	1.22936	2.4257	762.8	1.456	72.0	85.4	.84
Xe32 (D-3)	1.22936	2.4257	762.8	1.456	72.0		
Xe31 (D-8)	1.22936	2.4257	762.8	1.456	72.0		
H165 (C-10)	1.22936	2.4638	761.9	1.673	79.1	90.0	.88
H163 (D-1)	1.22936	2.4638	761.9	1.673	79.1	89.7	.88
H162	1.22936	2.4638	761.9	1.673	79.1	90.4	.88
H164	1.22936	2.4638	761.9	1.673	79.1	89.6	.88
H169	1.22936	2.4638	761.9	1.673	79.1	89.8	.88
Ar115 (D-10)	1.22936	2.4638	756.1	1.152	59.9	64.8	.92
Kr81	1.22936	2.4257	768.4	1.023	54.5	89.1	.61
Kr86	1.22936	2.4257	768.4	1.023	54.5	90.3	.60
Kr87	1.22936	2.4257	768.4	1.023	54.5	87.4	.62
Kr88	1.22936	2.4257	768.4	1.023	54.5	90.1	.60
Kr90	1.22936	2.4257	768.4	1.023	54.5	88.2	.62
Kr84	1.22936	2.4257	768.4	1.023	54.5		
Xe49	1.22936	2.4257	761.6	.936	50.6		
Xe42	1.22936	2.4257	761.6	.936	50.6	90.4	.56
Xe46	1.22936	2.4257	761.6	.936	50.6		

TABLE 1 (cont)
THEORETICAL TRITON-IMPLANTATION ESTIMATES FOR THE T/P EXPERIMENT
(Assuming initial triton kinetic energy = 0.191 MeV)

Container	Radius (µm)	Length (cm)	Pressure (Torr)	Triton Range (cm)	Theoretical Percent of Tritons Implanted	Measured Percent of Tritons Implanted	Calculated/Measured
CAPSULES:							
Xe53	1.22936	2.4257	763.8	.935	50.5		
Xe54	1.22936	2.4257	763.8	.935	50.5		
Xe59	1.22936	2.4257	763.8	.935	50.5		
Xe55	1.22936	2.4257	763.8	.935	50.5		
Ar123	1.22936	2.4638	751.4	.519	29.7	51.5	.58
Ar130	1.22936	2.4638	751.4	.519	29.7		
Kr94	1.22936	2.4638	758.9	1.036	54.9		
H171	1.22936	2.4638	751.5	1.694	79.8	91.8	.87
H175	1.22936	2.4638	751.5	1.694	79.8	90.9	.88
He6	1.22936	2.4257	2286.0	.562	32.1		
Ar136	1.22936	2.4638	760.3	.513	29.4		
Ar137	1.22936	2.4638	760.3	.513	29.4		
Ar138	1.22936	2.4638	760.3	.513	29.4		

TABLE 2
APT TRITIUM IMPLANTATION LOSSES
(1700 MeV protons, 13 ladders)
(16 x 160-cm beam footprint, 1-m shield all around)
(Assumption: Triton energy = .191 MeV)
(Triton range in 100 psi, 300-K ^{3}He = .257 cm (from TRIM))

Material	% of Total Tritium Produced in Each Material	^{3}He Tube Inner Diameter (cm)	Fraction of Tritium Implanted in Walls	% of Tritium Production Lost
Decoupling	23.459	2.20346	.11663	2.736
Very High Power Lead	48.102	1.40970	.18231	8.769
High Power Lead	17.871	2.36220	.10880	1.944
Low Power Lead	8.712	2.36220	.10880	.948
Reflector	1.857	2.36220	.10880	.166
Total	100.000			[a]14.600

[a] Preliminary comparisons of theoretical implantation with experimentally measured implantation indicates that theoretical implantation predictions may be ~10% low. Therefore, (14.6)(1.1) = 16% may be closer to the actual implantation losses for this assumed set of ^{3}He distribution tubes.
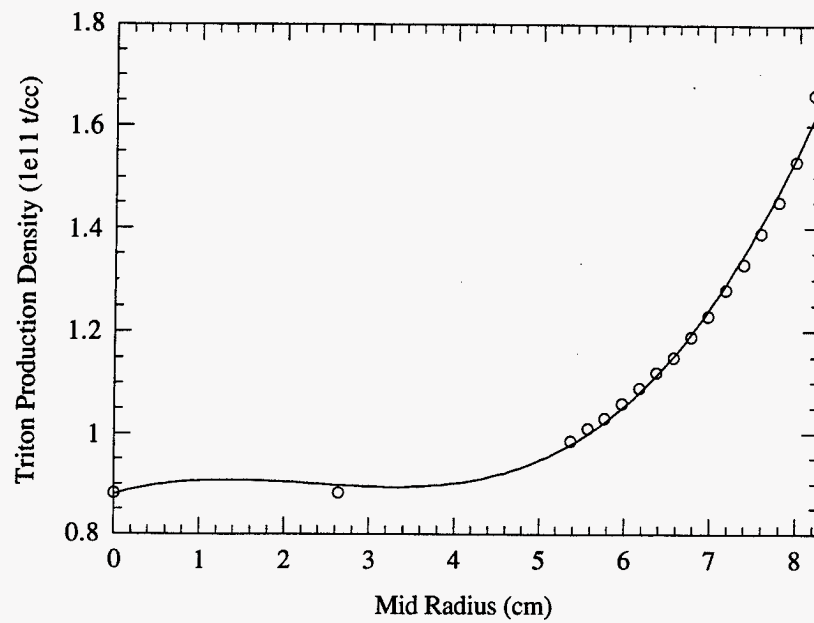
Fig. 1. The predicted triton production density as a function of radius in Sphere 2 of the tritium production experiment.
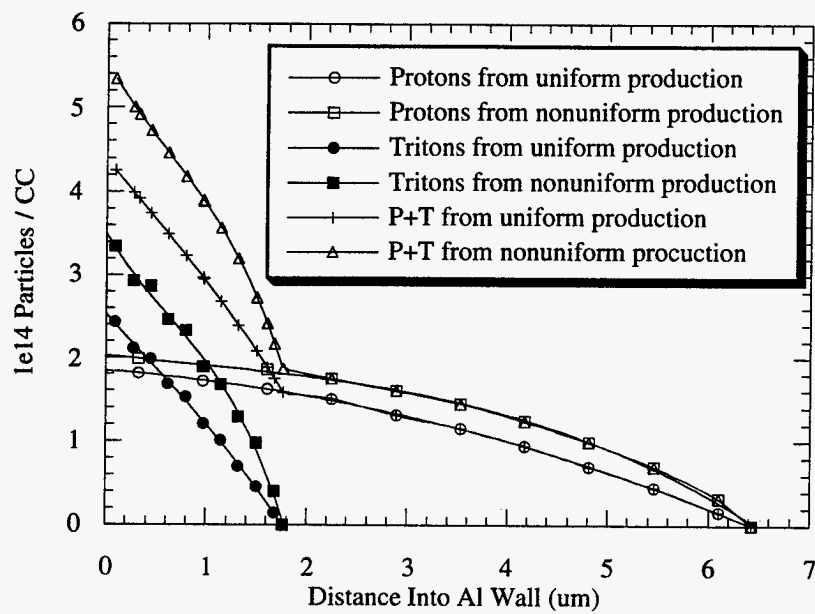

Fig. 2. The predicted triton and proton implantation distributions as a function of radius in the aluminum wall of Sphere 2 of the tritium production experiment.
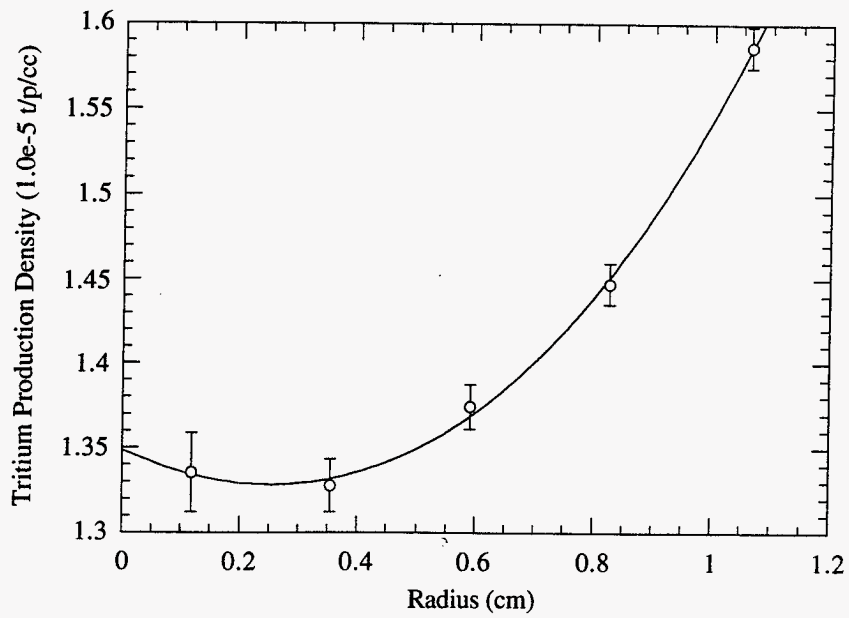
Fig. 3. Radial profile of the vertically averaged tritium production densities in the ³He gas of the highest-power, explicitly modeled, decoupling-zone tube. The tube inner radius is 1.1811 cm and is located in the inner row of decoupling tubes next to ladder 6.
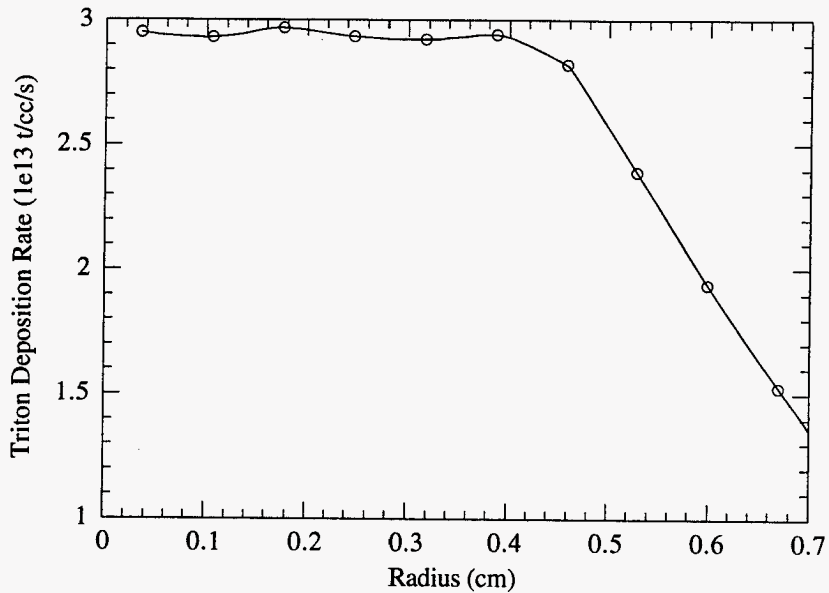
Fig. 4. The radial distribution of the triton deposition in the distribution tube ³He gas at the peak tritium production location in the very-high-power-lead blanket. The APT is at full power (100 mA of 1700 MeV protons), and the tube radius is 0.70485 cm. The triton generation rate was assumed to be radially flat at 2.95e13 tritons/cc/s.
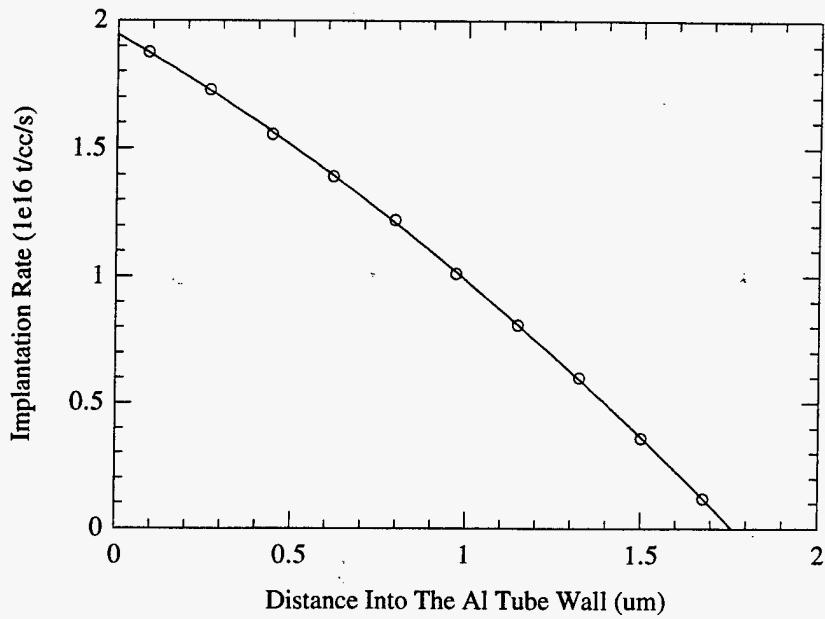
Fig. 5. Triton implantation distribution in the aluminum distribution tube walls at the peak tritium production location in the very-high-power-lead blanket. The APT is at full power (100 mA of 1700 MeV protons) and the tube radius is 0.70485 cm.
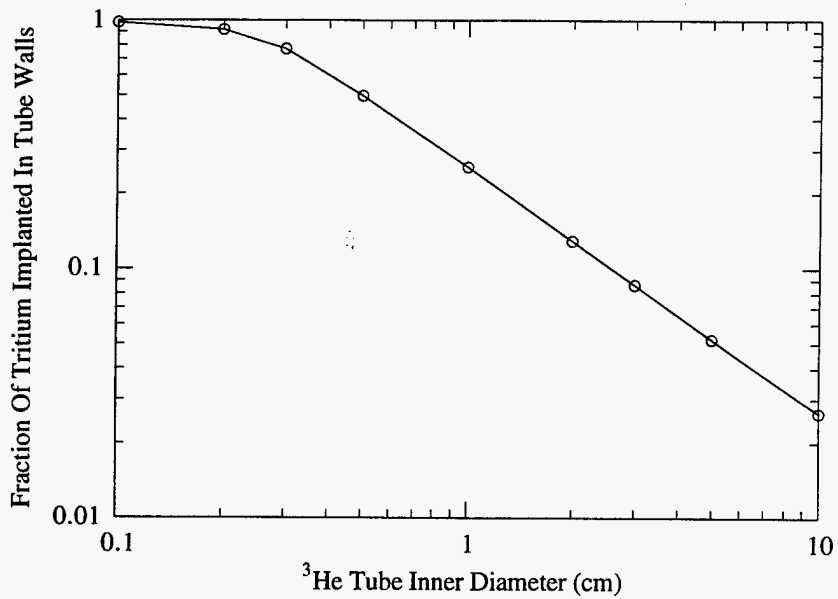

Fig. 6. Reducing tritium implantation losses by increasing the tube size. The ^{3}He gas is at 100 psia and 305 K. The tubes are 250 cm long. The initial triton energy is 0.191 MeV, which gives them a range of 0.257 cm in this ^{3}He gas.
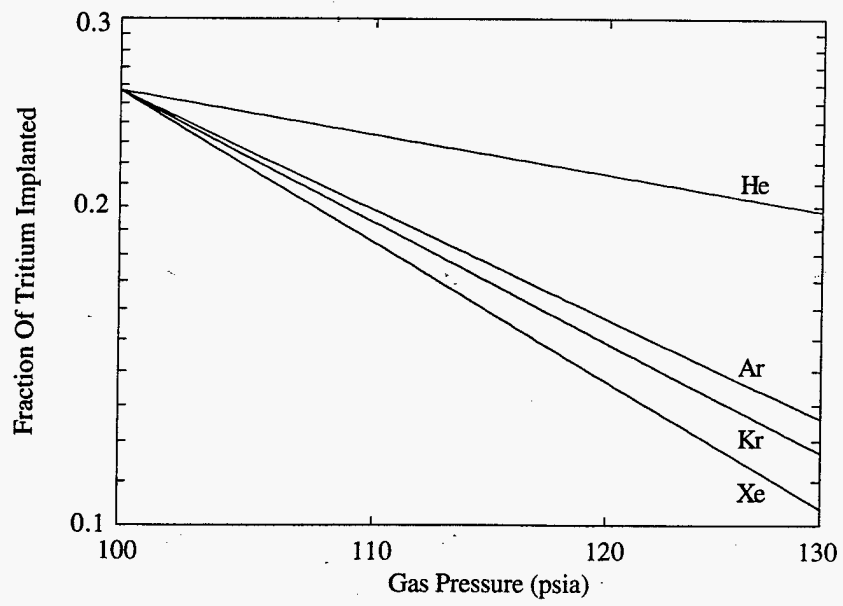
Fig. 7. Reducing tritium implantation losses by adding various heavy gasses to the original 100 psia ^{3}He gas. The tube diameter is 1 cm and the gas temperature is 305 K.

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