

Commissioning an X-Ray Detector System for Spectral Analysis of Tritium-Filled Targets

Introduction

Both glass and plastic targets filled with deuterium–tritium mixtures (DT) are used at LLE for research into inertial confinement fusion experiments. These targets are imploded with the 60-beam OMEGA Laser System. The permeation-filling process can take up to a week since the targets must be pressurized to tens of atmospheres without crushing the fragile, thin-wall shells. The overall time required to fill targets depends on the permeation time constant. Typically five or six permeation time constants are used to fill targets to the desired pressure. Although the fill pressure and DT ratio are known precisely, a nondestructive method to verify that the targets actually contain the expected fill pressure at shot time is currently unavailable. Two factors can lead to underfilling a target: (1) The permeation time constant is underestimated so sufficient time is not allowed for the charging pressure to equilibrate with the gas pressure inside the target, or (2) a defect develops in the shell, causing the target to depressurize more rapidly than predicted by the permeation time constant.

An x-ray detection system (XDS) originally developed to measure bremsstrahlung of tritium β decay from the surface of metals^{1–3} has been modified and developed with the capability of measuring the pressure of DT fuel inside a target just prior to the shot. This system permits rapid, nondestructive identification of underpressurized targets. This article details the design of the system, discusses preliminary measurements of x-ray emission from glass and plastic targets, and demonstrates that the method can achieve accurate, repeatable measurements.

Equipment Design Features

The XDS comprises three primary components: a high-resolution, dual-axes imaging system for repeatable, accurate target positioning; a helium enclosure with triple-axes micrometer positioning; and an Amptek silicon drift detector (SDD). The SDD was fit with a silicon nitrate (Si_3N_4) window to measure x-ray energies from 200 eV to 40 keV. The detector features a 25-mm² silicon drift diode with a measured 130-eV full-width-at-half-maximum resolution at 5.9 keV.

The arrangement of the equipment is illustrated in Fig. 148.15. The origin of the optical axes was defined by centering the window of the detector on the x, y origin in one camera's field of view (FOV) and the side view of the detector window in the z origin of the other camera. The detector was then retracted 25.4 mm in the z direction so that targets placed in the center of both cameras' FOV would be positioned 25.4 mm in front of the detector with a repeatable precision of 10 μm . During all measurement campaigns, the environment was purged with helium at a flow rate of 4 L/min to provide a

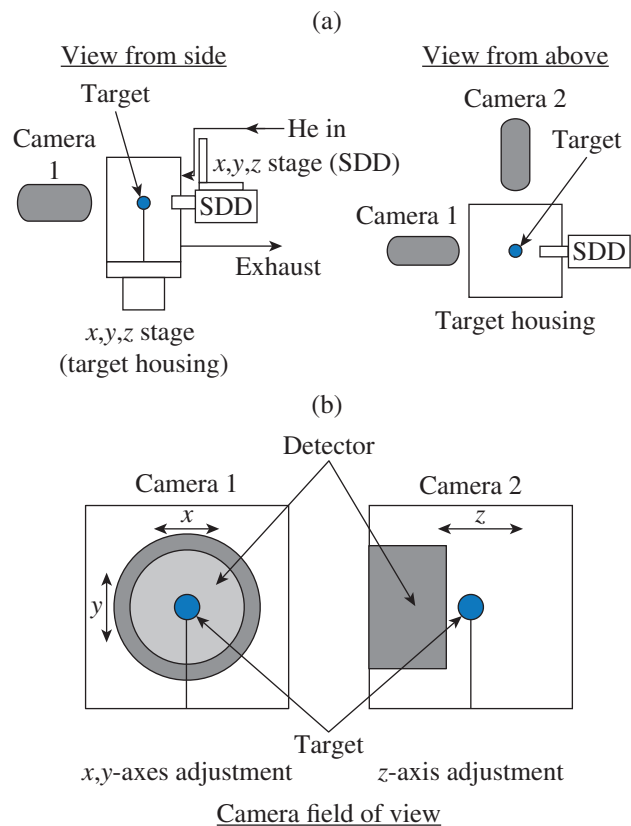
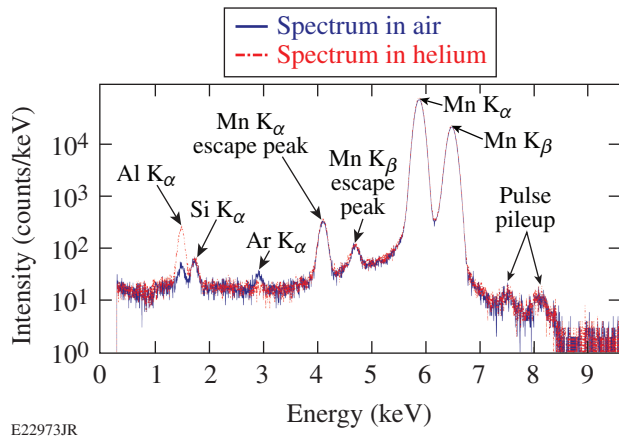


Figure 148.15
 Layout of the x-ray detection system. SDD: silicon drift detector.

tenfold volume exchange per minute within the enclosure to minimize air contamination of the chamber and to purge any tritium that permeated from plastic targets.

The response of the SDD was calibrated in air and helium using an ^{55}Fe source. The resulting spectrum is shown in Fig. 148.16. Calibration in air shows the suppression of lower-energy K_α lines such as aluminum and the presence of the argon K_α , as expected. K_α lines from manganese, the ^{55}Fe decay product, are also clearly visible in both the air and helium cover gases. A ^{109}Cd source was used to calibrate the detector over the full energy range of the tritium spectrum.



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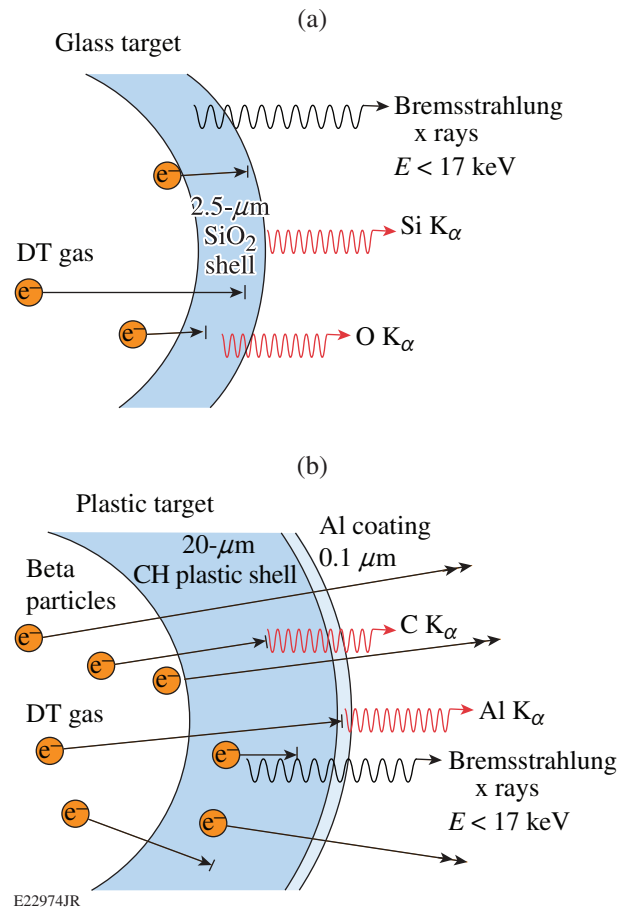
Figure 148.16 The response of the silicon drift detector to an ^{55}Fe source in air and helium.

Spectral Analysis of SiO_2 , CH, and CD Targets

Decay betas in tritium-filled targets generate both fluorescent and bremsstrahlung x rays as they interact with the shell material. These interactions are illustrated in Fig. 148.17 for both glass (SiO_2) and plastic targets. The permeability of tritium through glass is very low at room temperature and very little tritium is present in the enclosure. Additionally, the solubility of tritium in glass is very low and very little tritium resides inside the glass wall of the shell. Almost all decay betas originate in the DT gas and interact with the glass to generate x rays. These electrons have insufficient energy to penetrate the glass wall. By contrast, plastic is highly soluble and permeable to tritium. A 100-nm Al permeation barrier is applied to the shell's exterior to reduce tritium losses from the targets to manageable levels. The plastic contains a significant amount of tritium, and measureable amounts of tritium escape into the enclosure. In this case, decay betas originate in the gas and the plastic and can penetrate the aluminum overcoat together with x rays and tritium molecules.

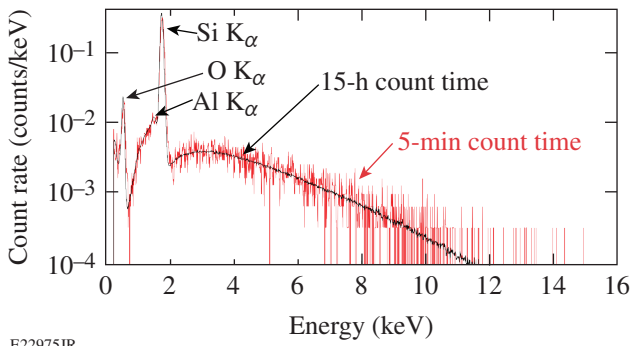
The emission spectra of glass targets are dominated by the fluorescence from silicon and oxygen atoms in the shell's wall. Two spectra from a glass target filled with 10 atm of 38.3% tritium in DT are shown in Fig. 148.18: one for a 5-min measurement time and one for a 15-h measurement time. The longer measurement time yielded a higher-resolution spectrum with more-discernible features, notably the Al K_α line from the housing of the detector itself. Figure 148.19 illustrates that once the DT gas has leaked out of the glass target, the emission spectrum drops to background levels, demonstrating that the amount of tritium dissolved into the glass is negligible.

Three glass-target silicon K_α lines were measured for 5 min and compared. Two of these targets were imploded on OMEGA later on the same day. As shown in Fig. 148.20, the two silica targets had a Si K_α count rate within 5% of each other and produced neutron yields within 10% of each other on OMEGA.



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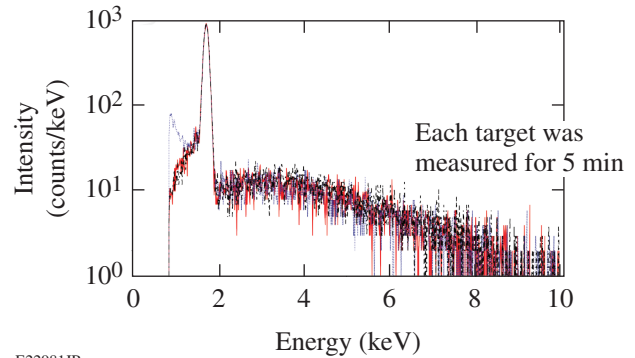
Figure 148.17 Illustration of the interactions decay betas undergo in glass and plastic targets.



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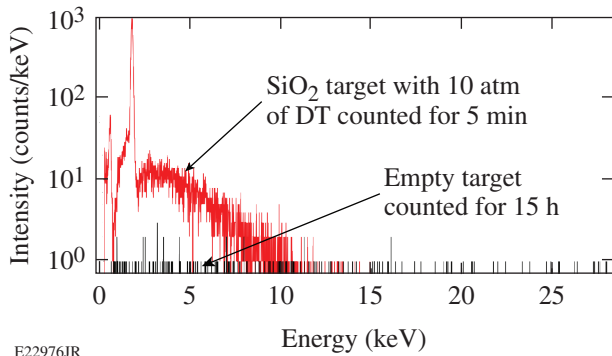
Figure 148.18
Comparison of a 5-min emission spectrum and a 15-h emission spectrum collected from a glass target containing 3.83 atm of T.

Target number	Target ID	Si K_{α} rate	Neutron yield ($\times 10^{13}$)
SiO ₂ -1	ISE-3Q14-01-8	31.9	3.02
SiO ₂ -2	ISE-3Q14-01-5	31.3	n/a
SiO ₂ -3	ISE-3Q14-01-27	33.5	3.42



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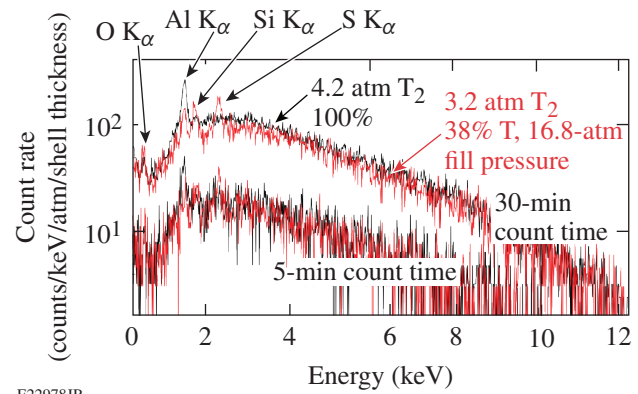
Figure 148.20
Five-minute spectra of the three glass targets listed in the table. These targets were fielded on OMEGA.



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Figure 148.19
A 5-min spectrum collected from a glass target containing 10 atm of DT compared to a 15-h spectrum collected from glass target that has lost its DT.

Fluorescence does not dominate emission spectra from plastic targets. Figure 148.21 compares two plastic targets in the energy range from 2.5 keV to 12 keV. One target was filled with 4.2 atm of T₂ at Lawrence Livermore National Laboratory (LLNL) and then shipped to LLE at liquid nitrogen temperature for attachment to a stalk. The second target was filled at LLE with 16.7 atm of 38.3% tritium in DT, or an equivalence of 3.2 atm of T₂ gas. This target was attached to its support stalk using ~80 ng of epoxy prior to filling with DT. Both targets exhibited strong bremsstrahlung emission with fluorescence K_α peaks for aluminum and oxygen. Additionally, the target filled at LLE presented silicon and sulfur K_α lines. Tritium soaked into the silicon stalk and the epoxy while the target was filled with tritium at LLE and subsequently fluoresced the silicon and the sulfur present in the epoxy. The larger amplitude of the aluminum K_α line found in the emission spectrum of the LLNL target may be attributed to the higher tritium pressure and thinner shell wall in that target by comparison to the LLE target.



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Figure 148.21
Five-minute spectra collected from two plastic targets: one filled to 3.2 atm of T₂; the other filled with 4.2 atm of T₂.

Beta particles that emerge from the plastic targets can interact with the detector window if the detector is within the beta range. This effect is demonstrated in Fig. 148.22, where emission spectra from plastic and silica targets are compared at 2.54 cm and 1.27 cm from the detector. The silica target exhibits the same bremsstrahlung spectrum at 1.27 cm as it does at 2.54 cm since no betas escape the shell. On the other hand, the emission spectrum from the plastic target develops a more-pronounced bremsstrahlung signature than the silica target at 1.27 cm when the target is close enough to the detector for escape betas to interact with the detector window.

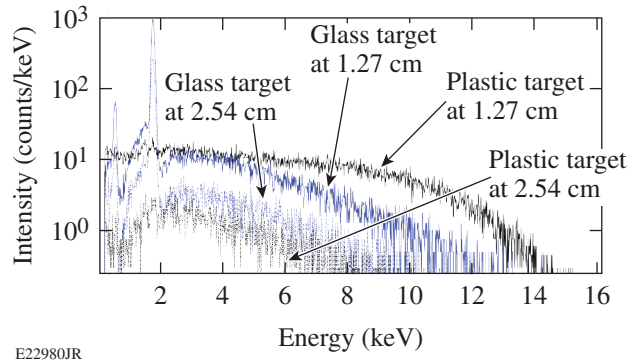


Figure 148.22 Comparison of spectra collected from glass and plastic targets at two distances from the detector to illustrate how decay betas escape from plastic targets interacting with the detector.

Correlation Between Plastic Target Activity and Pressure

Gas loss from plastic targets is approximately exponential. The permeation time constant is measured for each target using D₂ gas at General Atomics after fabrication. These time constants are then used to guide the tritium-filling rate. Once charged to the desired pressure with tritium gas, the targets are placed in cold storage to reduce permeant losses. Targets at LLNL are stored at 77 K, increasing the permeation time constant by a factor of about 1100, while targets at LLE are stored at 150 K, increasing the permeation time constant by a factor of about 800.

The x-ray spectra of five plastic targets were taken continuously while they were outgassing at room temperature to correlate the tritium activity against the gas pressure. All targets were measured at 2.54 cm from the detector face to suppress bremsstrahlung from the detector membrane. The environment was purged with helium at a flow rate of 4 L/min during measurements to prevent a buildup of tritium in the housing

as targets outgassed. Two targets (A1 and B1) were measured immediately after removal from cold storage during the diagnostic development campaign and then again after five days of outgassing at 24-h intervals for an additional five days. Three targets (A2, B2, and B3) were measured continuously upon removal from cold storage for a period of six days to obtain the energy/time spectrum. The targets in Group A were filled at LLE with 16.7 atm of DT (38.3% tritium) or an equivalent T₂ fill pressure of 3.2 atm while the targets in Group B were filled at LLNL with T₂. Estimates of tritium content in the targets at the time of the initial x-ray emission measurement and the shell composition are provided in Table 148.V.

All targets were mounted at LLE. For the ease of comparison, DT-filled targets are from here on reported in their equivalent T₂ concentrations.

The time evolution of the spatially integrated count rate of the targets takes on the shape of a double-exponential function as seen in Fig. 148.23. Decay betas from two sources are most likely responsible for the shape of the activity-time curve: decay betas from tritium gas inside the shell and decay betas from tritium dissolved in the target shell. The decay in activity is the result of losing tritium through diffusion through the shell or the dissolution of tritium out of the shell into the helium gas flow away from the detector. The two sources are referred to as the “gas activity” and the “shell activity” to differentiate between the two sources and their effect on the time evolution of the spectrum. The time constant for the gas activity component is expected to be short and dependent on the gas pressure in the target. The shell activity is expected to have a very long time constant since the tritons are most likely chemically bound in the plastic. A 40-h measurement of the change in the baseline activity suggests the time constant for the shell activity is of the order of 3.6 years.

Table 148.V: Tritium-fill pressure and the estimated tritium pressure in each target at the time of the x-ray measurement.

Target ID	Tritium-fill pressure (atm)	Estimated target pressure at time of x-ray measurement (atm)	Shell composition
A1	3.2*	2.9	CD
A2	3.2*	2.4	CH
B1	5.0	3.6	CH
B2	5.1	3.2	CD
B3	12.2	12.2	CD

*Actual fill pressure of 16.7 atm using 38.3% T in DT.

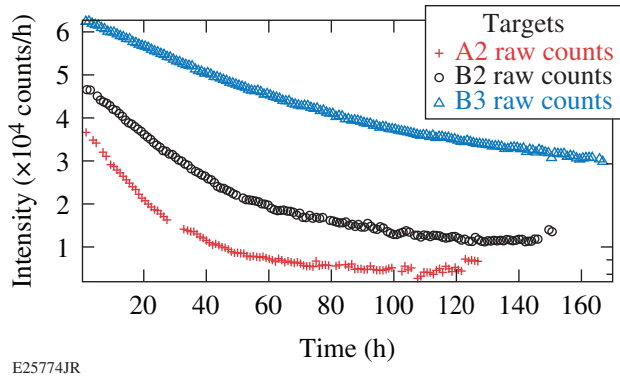


Figure 148.23
The time evolution of the spectrally integrated count rate of three plastic targets outgassing at room temperature.

The spectra were fit with weighted double-exponential functions after reaching the baseline shell activity and using 3.6 years as the half-life for the shell’s exponential function. Using the measured coefficients, the gas and shell activity were deconvolved from the spectra by subtracting each from the raw data as shown in Fig. 148.24. The measured permeation time constants for the gas activity source are provided in Table 148.VI and compared against the values provided by General Atomics. In each case the DT time constants are longer by at least 50% than the reported time constants determined with D₂ gas.

The pressure of the targets at each measurement interval was calculated using the reported soak time and fill pressures and the measured DT time constant, assuming the tritium loss rate from the target to be exponential. Table 148.VII compares the soak and fill pressures. The calculated target pressure evolution was plotted against the deconvolved gas activity evolution and is shown in Fig. 148.25. Pressure correlates linearly with activity as expected. The slopes of the pressure–activity curves

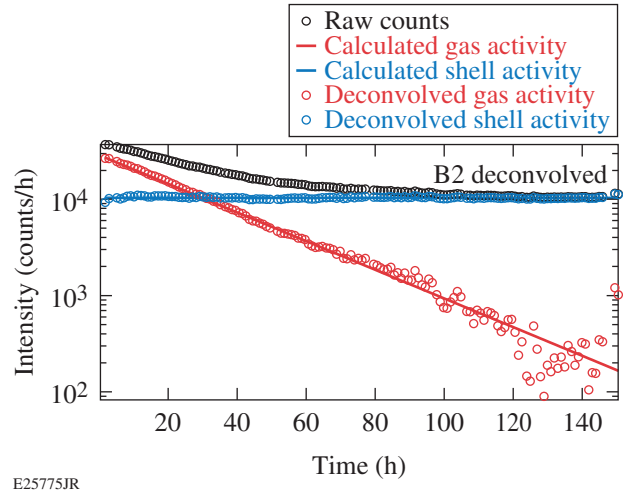


Figure 148.24
The continuous x-ray spectrum from a plastic target deconvolved to show the contributions from the DT gas inside the target and from DT gas dissolved in the plastic.

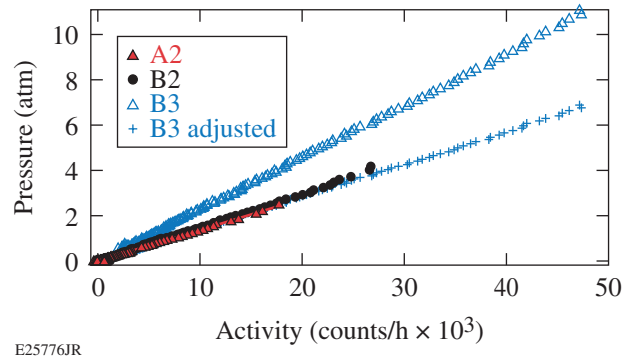


Figure 148.25
The calculated DT pressure loss from a plastic target compared against the x-ray emission from the residual DT inside the plastic target.

Table 148.VI: Comparison of the D₂ reported time constant with the measured time constant using DT.

Target ID	D ₂ reported time constant (h)	Mass-adjusted time constant (h)*	DT time constant (h)	Ratio of measured/reported
A2	5.1	5.7	12.8	2.3
B2	11.3	13.8	20.2	1.5
B3	23.7	26.5	39.0	1.5

*GA reported half-lives converted to a DT-equivalent time constant using a root-mass difference of the hydrogen isotopologues: D₂, DT, and T₂, i.e., $\sqrt{5/4}$ and T₂ by $\sqrt{3/2}$.

for targets A2 and B2 lie within 8% of one another, while the slope for target B3 is 1.6× greater than the average slopes of the other two targets. Taking the measured tritium activity to be accurate with <1% error in the counting statistics, the initial fill pressure of B3 should be closer to 7.6 atm of T₂ as opposed to the reported 12.2 atm. The pressure–activity curve for B3 using a fill pressure of 7.6 atm of T₂ (shown in Fig. 148.25) is observed to fall in line with the curves for the other two targets. The reduced fill pressure is being determined under the assumption that targets A2 and B2 had more accurately reported fill pressures since their slopes were within 8% of each other.

The shell activity of each target was measured independently using liquid scintillation counting at the end of each measurement campaign. Each target was crushed in liquid scintillation fluid to capture any residual tritium. Each crushed shell was soaked in the liquid scintillation fluid for several days to leach tritium from the plastic. A 1-mL aliquot was extracted from the leachate and measured in the liquid scintillation counter. These activities are also summarized in Table 148.VII. The shell activity was found to be linearly proportionate to the fill pressure of the target, or the initial total activity. Additionally, the plastic walls retained ~30% of the initial target activity.

Conclusions

An XDS has been developed and successfully commissioned with an intended application of nondestructively measuring the activity in tritium-filled targets. The XDS allows for an expedient verification of tritium content in a target prior to it being loaded into the OMEGA target chamber. The measured activities of two of the glass targets have been measured to have a Si K_α activity rate within 5% of each other, which corresponded to neutron yields within 10% after being imploded by the 60-beam OMEGA Laser System. This activity in glass targets is believed to provide a suitable initial baseline for

comparison with future glass targets, allowing targets to be screened for low, or vacant, tritium content.

The T₂ permeation half-life has been measured for three plastic targets by continuously measuring the decrease in activity over a period of six days each. The measured half-lives were observed to be between 1.5× and 2.3× longer than the corresponding D₂ half-life values measured by the manufacturer, after taking into account the root–mass difference for DT and T₂. The activity of the tritium bound to the shell was observed to be proportionate to the initial fill pressure. The shell activity was ~30% of the total activity in the target.

The target fill pressure at the time of x-ray measurement was calculated taking into account the different half-lives, fill pressures, and storage times of the targets. Based on the target fill pressure, the activity of A2 was calculated to be 17% lower than A1. The measured activity of A2 was 16% less than A1 for a +1% difference. The targets from Group B exhibited significant deviation from their expected activities. The difference between the expected and measured activities is attributed to an underestimate in the permeation time constant. Adjusting for the longer time constant in the activity of B3 reconciles the apparent discrepancy in the pressure–activity curve when the B3 curve is compared against the curves measured for A2 and B2. The fill pressure of B2 at the time of measurement was calculated to be 7% less than B1, with an expected similar difference in activity. The actual measured activity for B2 was 11% greater than B1 for a difference of +18%. The actual fill pressure of B3 was calculated to be ~33% less than the value reported by General Atomic, based on the slope of the pressure versus activity over time when compared with targets A2 and B2.

The XDS is a robust, nondestructive technique for confirming the actual tritium fill pressure in either glass or plastic targets.

Table 148.VII: Comparison of the shell activity to fill pressure and total target activity.

Target ID	Soak pressure (atm)	Target pressure (atm)	Fill-to-destruction duration	Shell activity (kBq)	Shell activity/target pressure (kBq/atm)	Shell/total activity (%)
A2	3.2*	2.4	48	926	386	31
B2	5.1	3.2	46	950	297	28
B3	12.2	7.6**	53	1259	166	29

*Adjusted for the actual T₂ content.

**Fill pressure based on known soak time and DT measured time constant.

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