HIGH YIELD, LONG-LIVED TRITIUM TARGETS

F. F. Haywood, Oak Ridge National Laboratory
H. E. Banta, E.G.&G., Incorporated
Z. G. Burson, E.G.&G., Incorporated

Summary
In order to extend the lifetime of conventional nonrotating tritium targets (say, TiTz), a new design concept for neutron generator targets has been tested successfully. It consists of a device which allows the target area to be replenished during operation. It has been used to produce neutrons [T(d,n)He reaction] for about ten hours at a sustained 4n yield of $2 \times 10^{11}$ n/µA-s. Tritium diffuses through a palladium foil into the target area when the foil is heated by a deuteron beam. Control of the flow of tritium is accomplished by regulating gas pressure in the reservoir and water flow in the cooling system.

Introduction
Sustained high yields of 14-MeV neutrons from the T(d,n) reaction at voltages of ~ 200 kV are difficult to obtain; conventional targets decay almost exponentially with a short half-life. At beam densities of about 1 mA/cm², this half-life may be as short as twenty minutes. The investigation reported here is concerned with a self-replenishing target. The substrate is palladium, which is porous to tritium at elevated temperatures. Thus, under bombardment and/or local heating, tritium lost from the front face of the target can be replaced by tritium flowing from a reservoir behind the target. A more complete report of this work is available elsewhere.¹

The logical application of this research in the foreseeable future is in the fields of general radio-biological research and cancer radiotherapy using neutrons. One very strict requirement for a neutron generator used for this latter purpose is imposed by therapists who would be conducting clinical trials; that is, the generator must produce at least $10^{12}$ n/s for periods of 30-40 hours² (this many treatment hours before making physical changes on the machine). Such lifetimes are not possible unless there is some mechanism for replenishing the target during operation. Rotating targets on low-voltage accelerators exhibit long lifetimes only because of a much greater total target area, which is usually in an annular ring. On the other hand, long lifetimes with nonrotating targets are obtained with deuteron beams of a few MeV because of particles penetrating the target and depositing their energy in a cool substrate.

Apparatus
Experiments were performed with the target assembly mounted on the beam tube of a small positive ion accelerator. Deuterons were from an unanalyzed 1-mA beam, whose diameter at the target was 1 cm. An overall view of the assembly, which is mostly copper, is shown in Figure 1. It is 2-1/2" long, 1-3/4" wide, and 1/4" thick. It is made up of two end-cap headers (A) and a larger center piece (B) in which a shallow gas reservoir was machined. Six holes, 0.040" diameter, were drilled the length of (B) just under the gas reservoir for circulating coolant. A foil of Pd-Ag alloy (0.80 Pd, 0.875" diameter), was brazed in an inert atmosphere to form the top cover of the gas cell. Tritium was supplied through a 1/8" stainless steel tube to the cell from a uranium trap. A layer of Ti was deposited over the palladium so that tritium diffusing through the foil could form a hydride target. Neutron yields were determined using falt activation techniques; a moderated BF₃ counter was used to monitor yield as a function of time.

Experimental Results
At the beginning of this series of experiments, the authors felt a need to investigate the decay processes of conventional tritium targets. There are two hypotheses about such decay: (1) When a very large number of deuterons strike a limited target area, penetrating a moderated BF₃ counter, and, once released, these atoms would be knocked out of the target material. If the first of these two hypotheses were dominant, target decay would be strongly related to temperature. If the second were dominant, the strong dominating influence would be increase of beam density. Through a series of neutron yield vs. operating time studies for fixed beam current (1 mA) and accelerating voltage (185 kV), it was determined that there was essentially no difference in the half-life of targets when cooled with water at 17°C or liquid nitrogen (target temperature was -150°C). These results are shown in Figure 2. Also shown are for TiT₂ and SeT₂ targets cooled with water and a ScT₂ target cooled with liquid nitrogen. It is seen that there is little, if any, difference in the half-life of targets when operated at large temperature differences. From this, the conclusion is inescapable that radiation damage by bombarding deuterons is the dominant mechanism responsible for target decay.

Several self-diffusing target assemblies were fabricated and tested. The first of these was for a palladium thickness of 0.030" and 0.305 mg/cm² of Ti. Although the results did not represent optimum parameters, feasibility was again demonstrated as an earlier case.¹ The next assembly tested incorporated a 0.420" palladium substrate and a 1.0 mg/cm² layer of Ti. A maximum yield of $2 \times 10^{10}$ n/µA-s was observed over a period of two hours. During this time, the tritium pressure varied from 8" to 24.5" vacuum (Hg). A decrease of about 35 percent in neutron yield was noticed; however, it was concluded that neutron output was not strongly related to tritium pressure. The third and final target was made with a 0.030" palladium foil and Ti thickness of 0.445 mg/cm². Tritium pressure in the cell was maintained at 10" vacuum. The results of this run are also shown in Figure 2. There was an initial buildup in 20 minutes to a yield of $4.9 \times 10^{10}$ n/µA-s. This level continued for a period of three hours, thereafter a half-life of 2.5 hours was observed. After 4.8 hours, the accelerator was shut down overnight (Figure 2, Position A). The following one-hour of operation indicated the yield was offset lower and declined at near the previous half-life. As predicted, a slight beam displacement to fresh hydride (Figure 2, Position B) resulted in an increase by a factor of two in neutron output. Some of the data point fluctuations past position B are due to making slight changes in beam current and voltage. At nine hours, the voltage was increased to 200 kV, and the beam current was increased to 1.2 mA (data normalized to 1 mA).

¹Research sponsored by the U.S. Atomic Energy Commission under contract with Union Carbide Corporation.
Conclusions

It has been concluded that the dominant mechanism in limiting the lifetime of tritium targets is radiation damage to the hydride by bombarding deuterons. A self-replenishing target, utilizing a palladium-silver alloy leak for diffusing tritium into the target area, offers a unique way in which to obtain long lifetimes between target changes. For application to radiobiology research and cancer therapy, for which a neutron yield of $10^{12}$ n/s is required, this system used on a 100 kV accelerator (200-300 kV) with 50-100 mA beam currents would provide the required output. Although there was evidence of target degradation in this experiment, it would be a relatively simple matter to replace the Ti layer without opening the system. This could be done by incorporating a sputter source and Ti evaporator in the beam tube. After many hours of operation, the used Ti could be sputtered away from the palladium. By using a collimated beam of Ti vapor, the target surface would be renewed. The system would be ready then for a new cycle.

Acknowledgments

The authors wish to express gratitude to J. A. Auxier for his interest and assistance. J. E. Jobst was very helpful during the final few runs. Appreciation is expressed to E. M. Robinson for operating the generator and to J. W. Poston for providing neutron yield calibrations.

References

3. J. E. Strain, private communication.

Figure 1. View of the Self-Replenishing Target. The palladium foil is shown in the center of the assembly. The body is primarily copper and measures 2-1/2" long, 1-3/4" wide, and 1/4" thick. Water is circulated through 3/16" copper tubes, and tritium gas is supplied to the gas cell through a 1/8" stainless steel tube. Target temperatures are measured using two chromel-alumel thermocouples.

Figure 2. Neutron Yield as a Function of Operating Time for Two Conventional Targets Cooled with Recirculating Water (17°C). One Similar Target Cooled with Liquid Nitrogen (-150°C), and the Self-Diffusing Target.