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A SELF-REPLENISHING TRITIUM TARGET for NEUTRON GENERATORS

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ABSTRACT

The pressing need for 14-MeV neutron generators which produce high neutron yields for long periods of time motivated a search for better methods of target preparation. The neutron yield from two reactions ($T[d,n]^4\text{He}$ and $D[d,n]^3\text{He}$) under a variety of conditions was studied.

Using deuteron beams on a variety of target materials, it was possible to show that no material, including sheet Ti, retained enough deuterons to produce $D(d,n)$ neutrons in amounts competitive with loaded Ti targets. However, an unloaded Ti target built up activity to the same level as a loaded target. Gold overcoats produced a small increase in yield, but not significant enough to warrant further study.

Implanting ions along channeling directions in single crystal copper produced a yield fifty percent higher than polycrystalline copper, but still significantly lower than standard targets and target materials.

Supercooled tritium targets show no longer life nor enhancement in yield than those operated at ambient temperature. From this result, the conclusion is reached that radiation damage by the bombarding deuterons is the dominant mechanism in target decay. Chemical bonds binding the tritium in the matrix are broken, and the tritium then diffuses out of the target material.

A new concept in target design was investigated. It uses the principle of self-replenishment with tritium through a silver-palladium membrane. Three targets of this type were tested; the third producing a high and sustained yield of 14-MeV neutrons. The yield surpassed the best "standard" target tested after only 16 minutes.

of operation. It was run for a period slightly less than ten hours with a yield generally above 2×10^{10} neutrons/sec (one mA/cm² @ 185 kV).

The silver palladium, self-replenishing target principle offers the most potential of obtaining high yields with long half-lives. Indeed, the third prototype tested in this series of measurements could be used immediately in small machines using beam currents on the order of one milliamperere. More research and development is required to maximize available parameters and to see if high-beam densities in small targets will also be successful. These studies are necessary before an appropriate machine can be designed or built.

INTRODUCTION

Whatever the application, neutrons produced by an accelerator or neutron generator have advantages over neutrons from other sources. These neutrons are monoenergetic or nearly so, the limit being imposed by accelerator instability and by energy loss of the bombarding particles in the target material. The $T(d,n)^4\text{He}$ and the $D(d,n)^3\text{He}$ reactions are most commonly used to produce these neutrons. The $T(d,n)$ reaction is a prolific producer of neutrons. At the maximum cross section (110 keV), the neutrons in the forward direction have about 14.8 MeV of energy, with yields about 5×10^{10} neutrons per mA-sec of bombarding particles. Some energy variation is available by choice of angle; neutron energy in the backward direction is about 13.3 MeV. Flux densities are not strongly dependent on angle, there being only about 15% fewer neutrons in the backward direction than in the forward direction.

The $D(d,n)$ reaction produces only one or so percent as many neutrons. These neutrons have approximately 3 MeV of energy, and both

flux densities and energies are much more strongly angular dependent than T(d,n) neutrons. The cross section for this reaction increases with energy to at least 300 keV. Thus, an accelerator capable of operation in the 100 to 300 keV range may be a prolific producer of neutrons for experimental purposes.

There is a pressing need for reliable, inexpensive, high neutron yield D-T accelerators. Fast neutrons appear to be superior to x and gamma rays for radiotherapy purposes.¹ Most solid tumors have hypoxic regions which are more resistant to x or gamma radiation than to fast neutrons having a higher LET. Large animal radiobiology research is needed to provide insight as to radiation effects on humans. RBE values (relative to ⁶⁰Co) for larger animals for fast neutrons appear to be less than one for LD 50/30, but much greater than one for specific responses.²

Further definitive neutron transport, secondary gamma-ray production, and shielding data for 14-MeV neutrons are needed to better define weapons radiation effects. Comparison of Operation HENRE data to calculations at intermediate distances (one-half mile) appear to be good.^{3,4} However, data are needed at more practical distances (three-fourths to one mile).

Machines capable of producing on the order of 10^{13} n/sec for long periods of time (30 to 40 hrs), using small targets, are needed for the above tasks. The only machine that has approached the above requirements was the HENRE machine.⁵ The initial yields were 1 to 4×10^{13} n/sec, but the half-lives were on the order of one hour. The target size was 1000 cm².

The major problem preventing the development of an accelerator to do the above tasks is the target. A brief survey of target preparations showed that there were many promising methods of preparing targets that had not been fully explored. A team, composed of ORNL and EG&G employees, evaluated several alternative methods to target preparations, both by study and by experimentation.

BACKGROUND

Tritium targets display rapid decay during deuteron bombardment as seen in Figure 1. Associated particles were counted as a measure of neutron yield from the $T(d,n)$ reaction; the figure is from an unpublished paper of 1964.⁶ Many quantities must be controlled and specified for such a measurement. Bombarding voltage was 150 kV, beam current 500 μ A. Coolant was water at 17°C on the back side of the 10 mil copper substrate. An elementary calculation shows that the tritium was not used up by the reaction. Beam density was one mA/cm², and heat density 150 watts/cm². In about four hours, neutron yield was less than ten percent of initial; at this point $D(d,n)$ neutrons began to be an important part of the neutron yield, minimizing the usefulness of the target.

For any application, a constant neutron yield would be desirable, and even essential for some applications. An understanding of the mechanisms involved in the decay process then would be very helpful in developing long-lived targets. Two of the authors (Haywood and Banta) independently made what would seem to be a key observation. The machines involved employed diffusion pumps and forepumps, so that it was possible to measure the tritium content of the exhaust gas. Both sets of observations showed a stepwise increase in the rate of tritium released when

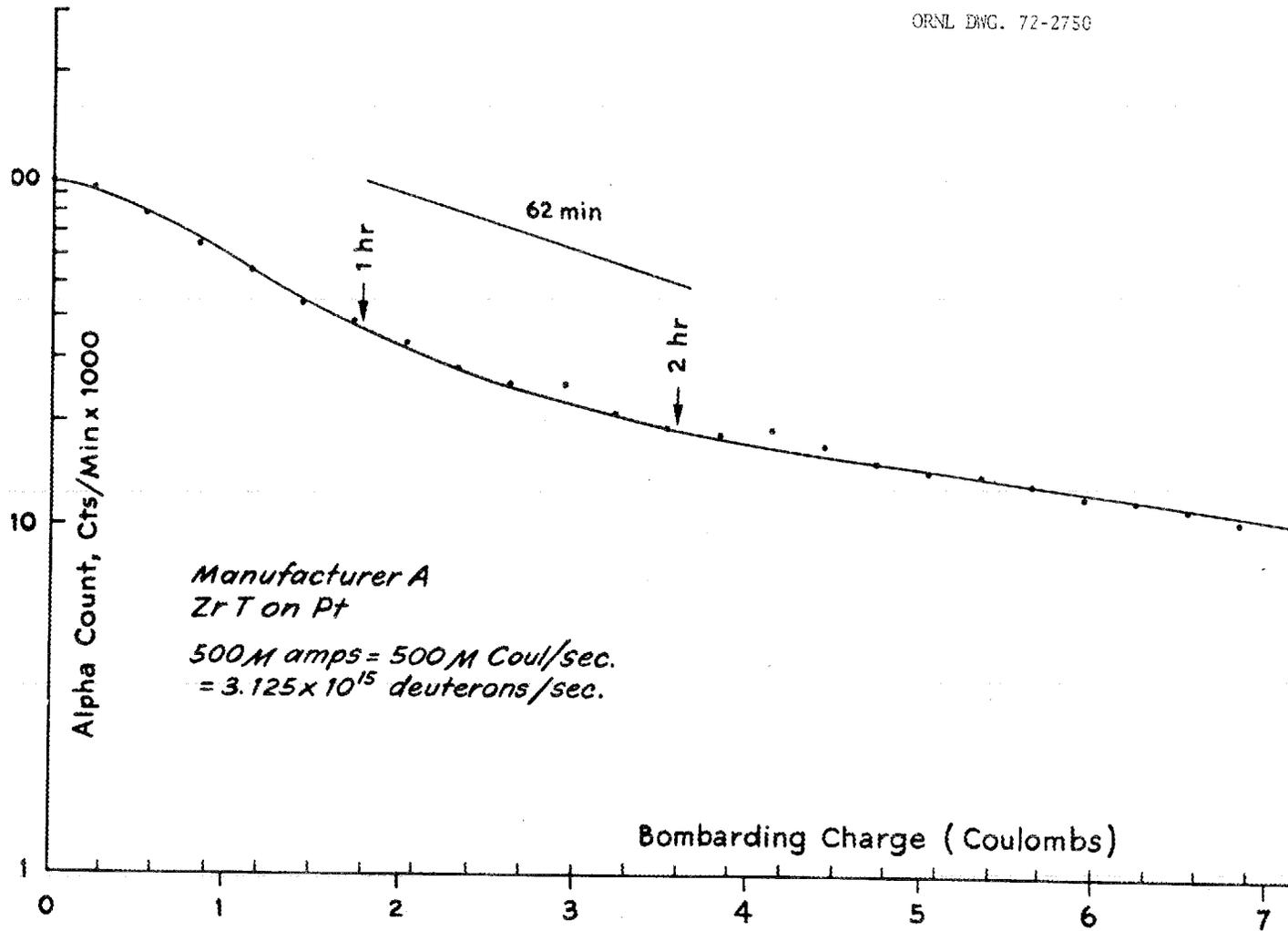


Fig. 1. Neutron Yield (Represented as the Associated Alpha Particle Count Rate) as a Function of Bombarding Charge and Operating Time from a Typical Target.

the beam was allowed to hit the target, along with a stepwise decrease when the beam was removed. The preparation of tritium targets for these machines normally involved vacuum evaporation of, for example, titanium on a 10 mil copper substrate, and reacting the titanium with tritium. The tritium was thus chemically bound to the target material, and the substrate cooled during bombardment to dissipate the heat developed. If it is assumed that the chemically-bound tritium is no health hazard, a grave error is made. In atmosphere, tritium is lost from the target, presumably at about the rate of exchange processes with air; also, flakes of tritide are lost from the target and may contaminate the environment.

The above observation about the release of tritium under bombardment allows the development of a hypothesis which may be tested by observation. A very large number of deuterons strike a limited area of the target, and are driven in to a maximum range of several hundred microns. Considerable heat is developed locally, so that conditions are most favorable for exchange processes to go on. Hence, deuterons replace the chemically-bound tritons; these tritons are then free to diffuse through and out of the target material. Local conditions would seem to favor both exchange and diffusion processes, since both are presumed to be related to temperature. Further, the tritides are likely to be spongy and particulate and to be poor heat conductors. Temperature drop through the copper substrate may be shown under conditions quoted to be only a few degrees centigrade, but locally, temperatures could be quite high, and direct observation is nearly impossible.

If the hypothesis developed here has any validity, a helpful measure would be to move the beam around on the target. This may be done by systematically moving either the beam or the target. It is indeed found that target life is much enhanced by such measures,⁷ but if one is considering large beam currents and large targets, for example Operation HENRE,⁸ different and more effective measures are indicated. If the beam (half an ampere on a 1000 cm² target) is allowed to spread and cover the whole target, moving either beam or target only results in a decreased neutron yield.

Another hypothesis may be offered to explain tritium target decay: Bombarding deuterons may break the bonds which bind the tritium in the target matrix. Once released, these atoms would diffuse out of the target material as in the previous hypothesis. The two hypotheses may seem closely related, although there are some vital differences. If the first were dominant, target decay would be strongly related to temperature. If the second were dominant, the strong variable would be beam density.

There have been few efforts to investigate the physical and chemical processes which constitute the cause, or causes, of tritium target decay. Compartmentalization of effort is a major cause of this omission. The laboratory group or commercial concern involved in tritium target production usually has little or no feedback from the experimenter. The experimenter produces neutrons for some experimental purposes and is little concerned about the mechanism of decay. The present effort was to investigate, by any means available, the decay processes and, if possible, produce a tritium target which would show longer life than the available targets.

It was learned recently⁹ that one research group had obtained some preliminary data that indicated the neutron yield for the D(d,n) reaction, with the target at liquid nitrogen temperature, may be substantially greater than that for a target which was kept near ambient temperature. This group tentatively concluded that by supercooling the target and bombarding with very intense beam currents, it may be possible to obtain an atomic ratio of 5:1 for hydrogen in copper. This, in turn, led to the supposition that a T(d,n) yield of $>10^{12}$ n/sec could be realized easily by mixing tritium and deuterium in the ion source and accelerating this mixture into the supercooled target. Essentially this would form a high pressure gas bubble in the copper, thus providing the needed quantity of target atoms.

It was therefore decided that several experiments would be conducted with liquid nitrogen as coolant. Clearly then, the hypotheses regarding tritium consumption as a function of target temperature and that deuteron buildup in supercooled substrates could be explored.

A self-diffusing technique of replenishing the tritium through a silver-palladium membrane was also studied. This technique shows much promise, above all other studied, for obtaining high-yield, long-lived targets.

EQUIPMENT AND APPARATUS

Experiments in this series were conducted at the ORNL Health Physics Division's DOSAR Low Energy Accelerator. The accelerator, in this case, is a Texas Nuclear Model No. 9999 neutron generator which is described elsewhere.¹⁰

Two types of target-cooling systems were developed which are discussed in detail. In addition, a self-diffusing gas target which was utilized in the $T(d,n)$ experiments represents a revolutionary approach in target design; it is described also.

Fortunately, it was not necessary to modify the beam tube in order to accomplish this work. The liquid-nitrogen-cooled targets were mounted as shown in Figure 2. A 3-in. diameter stainless steel beaker was used as the basic housing. The beaker was built in such a way that flanges could be welded as needed for attachment to the beam tube and for providing ready access in changing targets.

There were two design objectives to be achieved with nitrogen cooling. One of these was obviously to lower the target temperature while maintaining a vacuum seal; the other was to provide an adequate supply of liquid nitrogen to the reservoir behind the targets. Both of these were accomplished in the following way. A target holder was fashioned from a stainless steel plate. A 3/4-in. diameter by 1/2-in. deep reservoir was machined into this piece. This reservoir was filled with a plug of copper whose surface had several narrow slits 0.015-in. wide by 0.80-in. deep machined in the direction of liquid flow. The copper served two purposes: First, it provided a good sink for heat which was conducted from the back surface of the target; second, the narrow slits provided a larger area of contact with the liquid nitrogen and further induced a substantial pressure drop across the diameter of the target. This design promoted uniform conduction of heat away from the back side of the target. A 1/4-in. stainless steel coolant tube was welded to both the entrance and exit side of the target holder reservoir.

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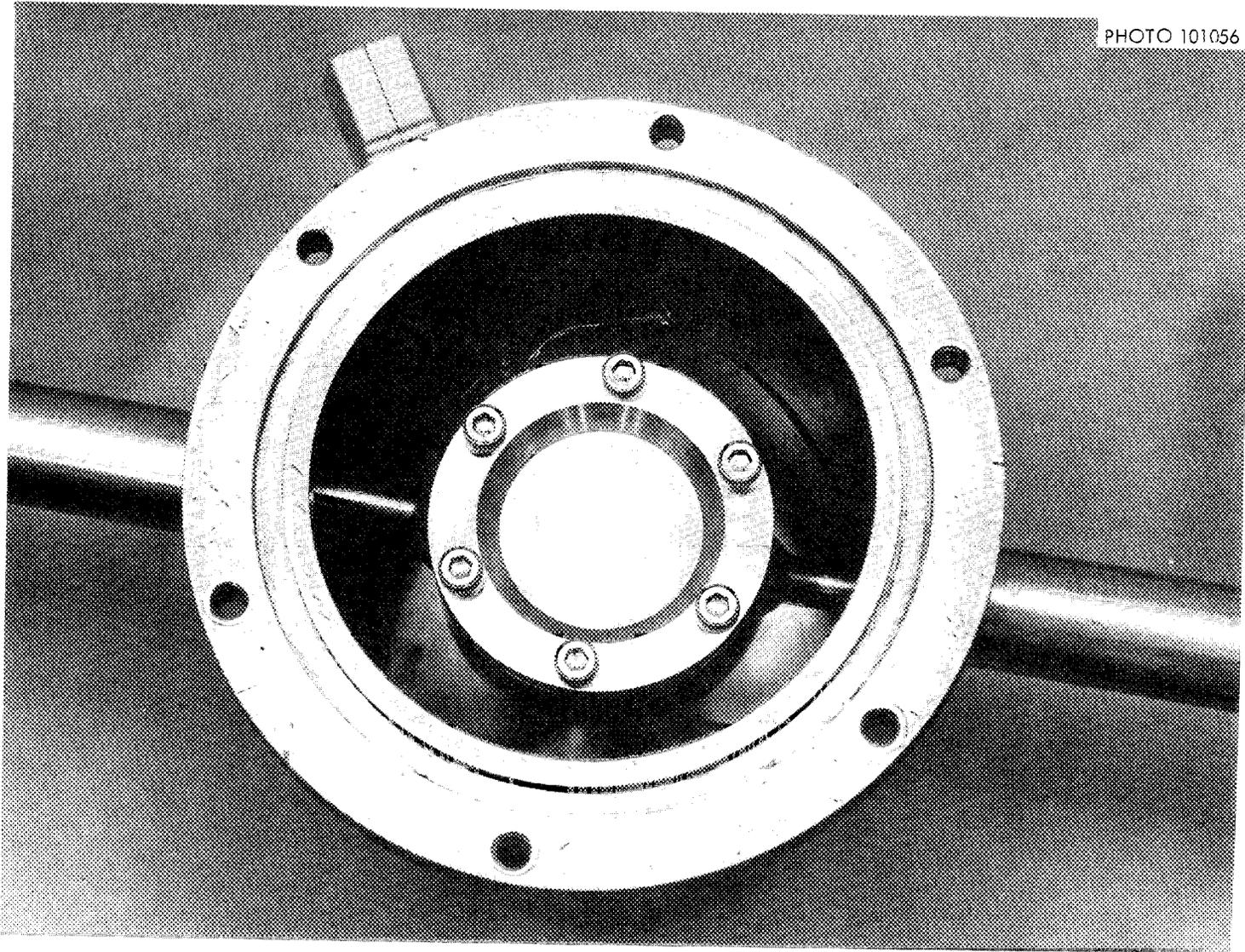


Fig. 2. View of Target Holder Used for the Supercooled Experiment.

These 1/4-in stainless steel fill-and-exit tubes were extended coaxially through 5/8-in thin wall stainless steel tubing. These were welded together about 6 inches away from the housing, so that overall outside temperatures would be near ambient.

All of the targets used in the supercooled experiment were 1/4-in. diameter and were about 10 to 20 mils thick. These were mounted on the target holder with a relieved clamp ring. An adequate vacuum seal was maintained by using indium wire instead of organic O-rings.

Liquid nitrogen was supplied from a 160-liter pressurized dewar at the rate of about 2 liters per minute. Fifteen minutes were required for cooling the target to -150°C prior to bombardment.

A thermocouple was attached mechanically to the target holder at the closest practical point. Covar seals were used to feed the thermocouple leads through the target housing as shown in Figure 3. Thermocouple signals were monitored with a standard laboratory potentiometer, referenced to a 0°C junction.

Near the end of Operation HENRE, a suggestion¹¹ was made to the USAEC that targets replenished with tritium during use could maintain high yields as well as provide much longer half-lives. This could be accomplished by diffusing tritium gas through a permeable barrier, such as a Pd-Ag alloy. A barrier foil would then act as a valve, passing tritium when heated by the beam, but blocking the flow of gas when the foil temperature was reduced.

A metal composition of 80% Pd and 20% Ag was chosen in order to retain dimensioned stability in a hydrogen environment. Foils were prepared in thicknesses ranging between 0.010 in. and 0.030 in. The overall target size was kept as small as practical, having both a fill port and

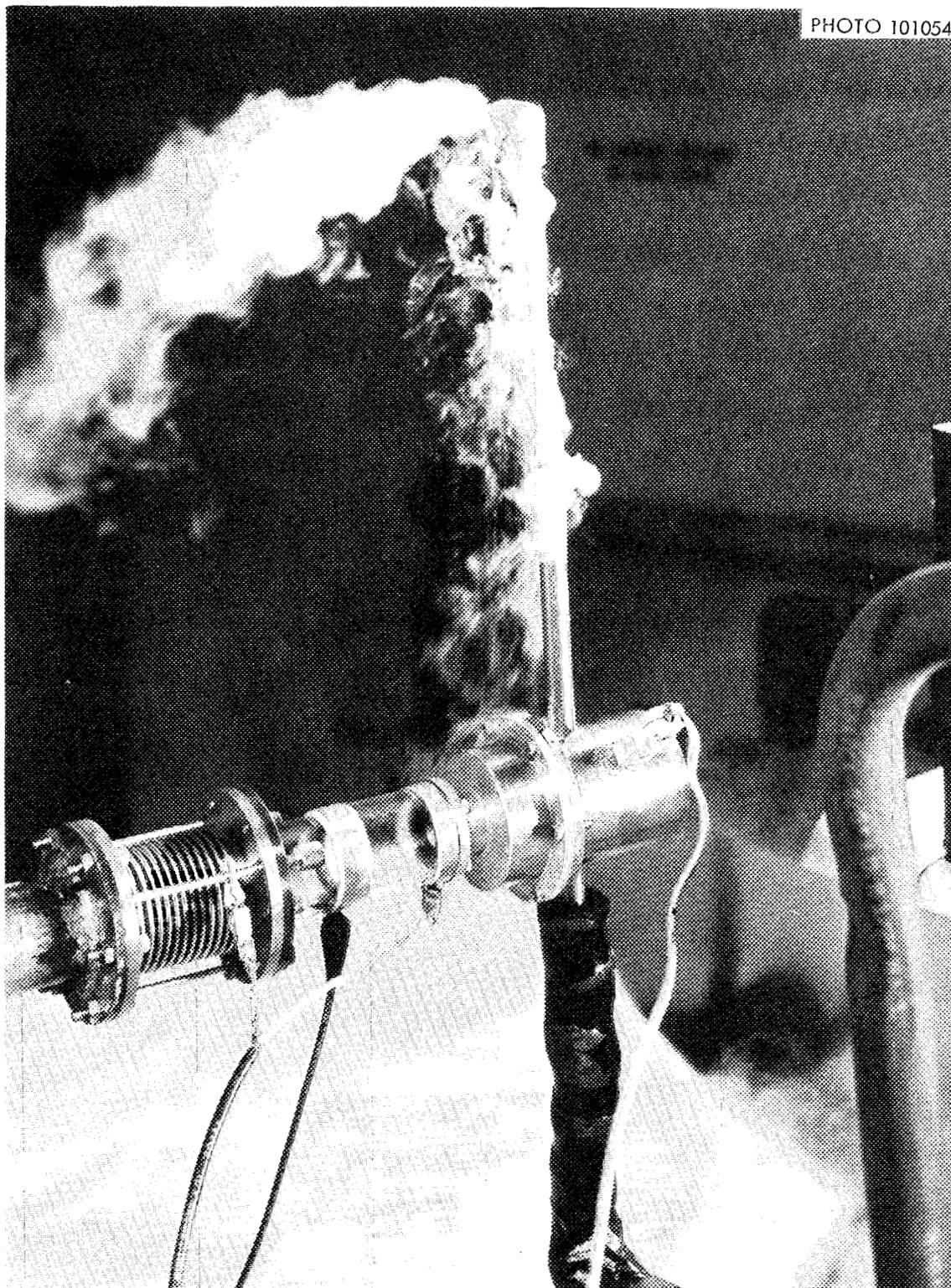


Fig. 3. View of the Supercooled Target Assembly Mounted on Neutron Generator with Liquid Nitrogen Flowing.

reservoir for tritium and having a cooling system adequate to remove 200 watts developed by beam bombardment. Details of the target are shown in Figures 4 and 5. A 7/8-in. diameter Pd-Ag foil was brazed to the copper substrate in a He atmosphere. Heat was conducted away from the foil at the rim (1/16-in. overlap) and at several points on the back of the foil where it was brazed to 1/16-in. square posts 0.065 in. high. These posts were made by machining away surrounding copper to create a gas reservoir. Cooling of the substrate and Pd-Ag foil was accomplished by circulating water through 0.052-in. channels drilled through the substrate. A 1/8" x 1-1/8" header was brazed to each end of the cooling channels for uniform water distribution. Small access holes for two thermocouples were provided near the gas fill line in order to measure temperatures on the back side of the Pd-Ag foil. Titanium was evaporated on the top surface to be used as the occluder. The entire target assembly, brazed together, measured 1-1/2" x 1-3/4" x 1/4".

EXPERIMENTAL RESULTS

General

The general purpose of this research was to investigate several possible approaches for tritium targets; to identify general trends and general conclusions rather than to obtain detailed definitive data on each target concept. Consequently, the results presented in this section reflect these objectives.

Initial experiments were conducted only on deuterium targets. This had the advantage of not dealing with tritium hazards and high neutron yields normally associated with tritium targets. To facilitate intertarget

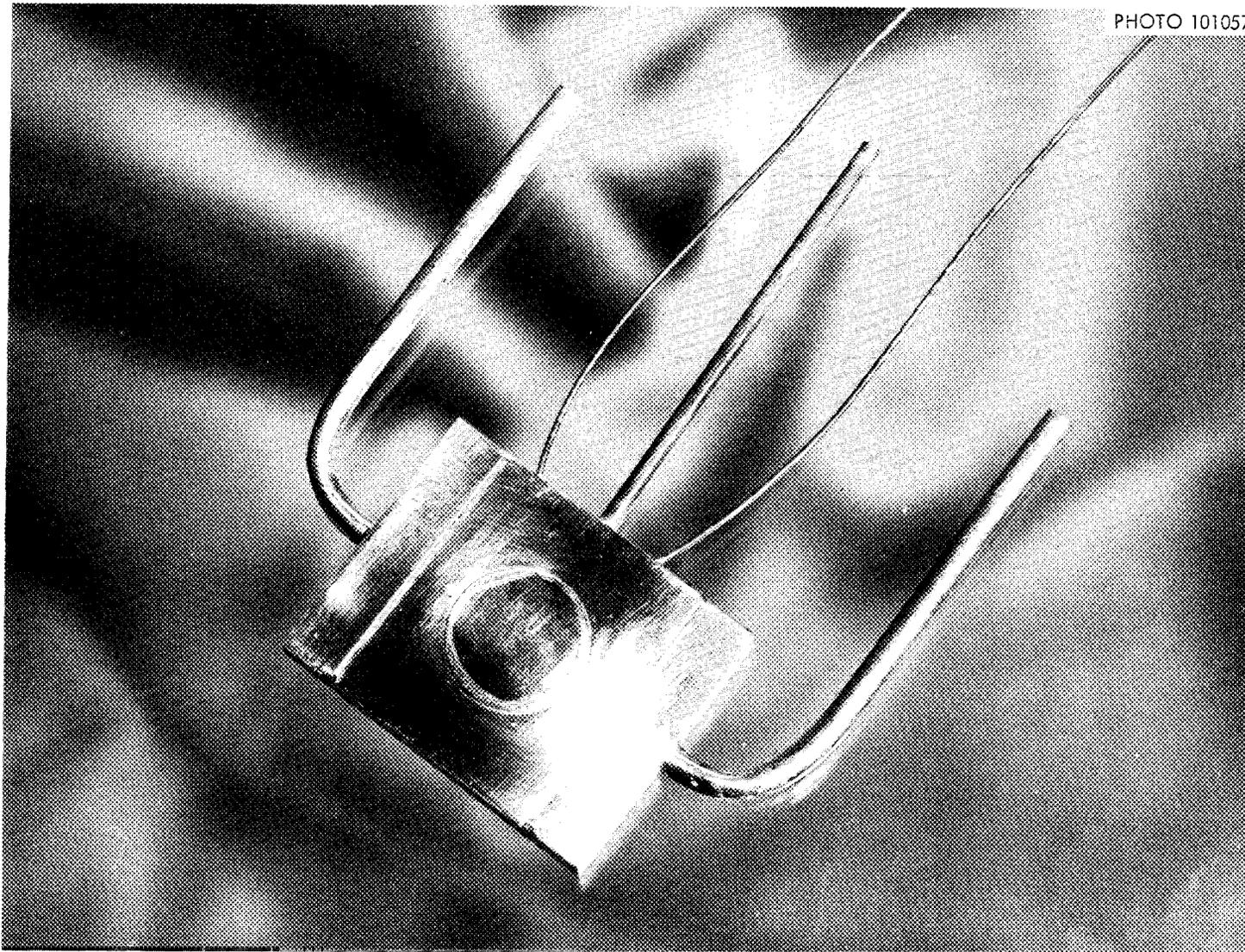


Fig. 4. View of Self-Replenishing Target Assembly Showing Ti-Coated Pd-Ag Surface, Coolant Lines, ^3H Fill Line and Thermocouples.

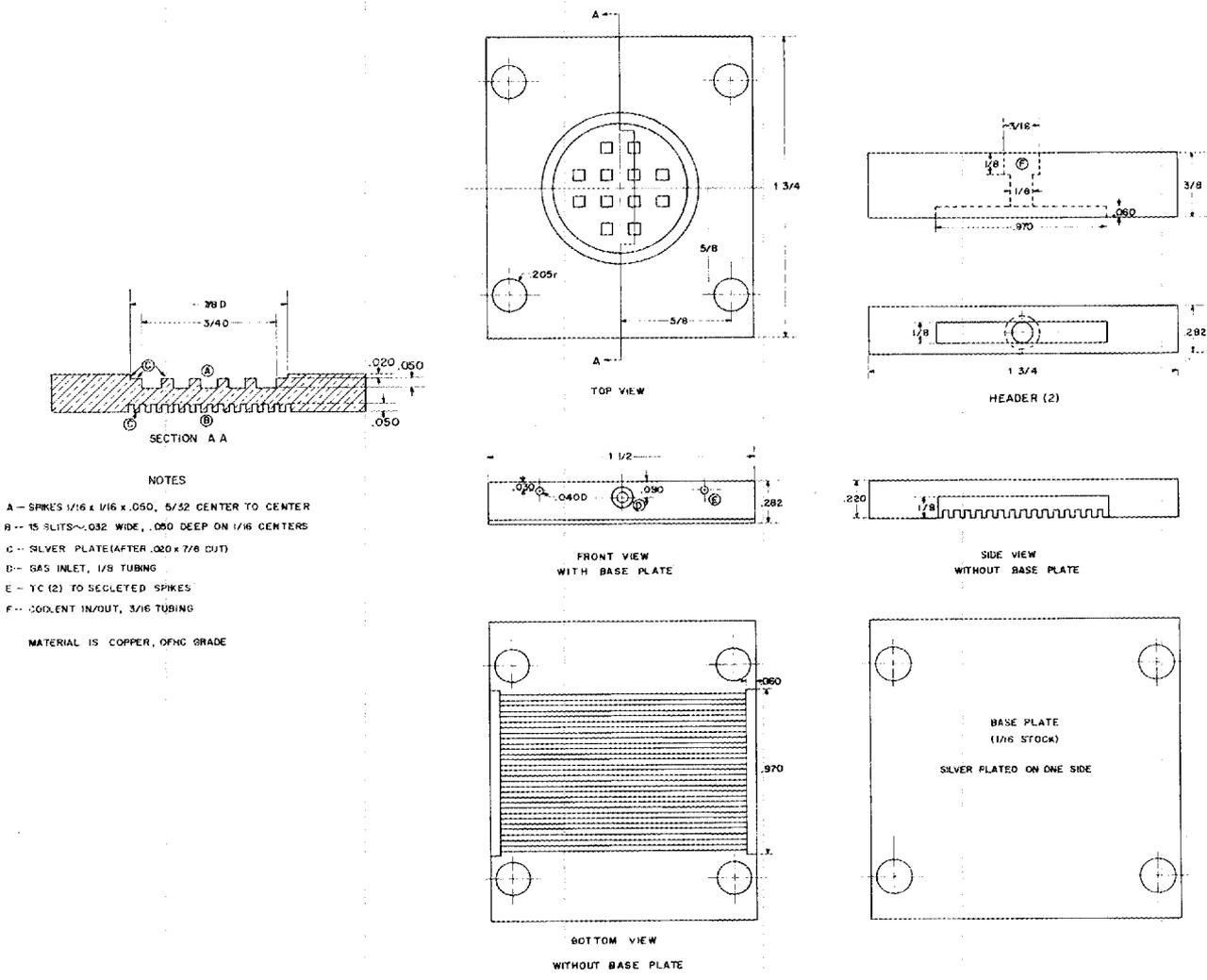


Fig. 5. Engineering Drawing of Self-Replenishing Target.

comparison, standard operating conditions of current, energy, and beam density were established. The beam was collimated just upstream from the target by a 1-cm² aperture. Bombarding particles were deuterons at all times. An R-F type ion source was used which produced particles around ninety percent atomic. Standard conditions were 0.95 to 1.0 mA of beam current at 185 kV, with the beam defocused, so that a few percent of the charged particles were stopped at the collimator. Thus, at least something was known about beam and power density on the target, a quantity which is frequently unspecified in target damage studies. After the D(d,n) studies were completed, the aperture was removed. The tightest focus produced approximately the same density as with the aperture in place. Thus, a beam density of 1 mA per cm² is quoted for all the results (except as noted). The variation in this quantity is probably contained within a factor of two.

Neutron yields were determined from data taken with a long counter placed in a permanent location. Empirical calibration factors were found by normalizing the counts from the long counter to 4π neutron output, determined by fission foils (for D-D neutrons) and sulfur pellets (for D-T neutrons) placed at 0° (forward direction) during calibration runs.

At the beginning of the T(d,n) investigation, several "standard" targets were run to obtain baseline yield and half-life data. Some were fabricated by the Isotopes Division of ORNL, and some were purchased from U.S. Radium. All were composed of evaporated titanium on a copper backing. The tritium loading process is accomplished in a vacuum system containing a partial atmosphere of tritium.¹²

During the studies, information was gathered from various sources that was helpful during the experiments. A portion of this information is included in the Appendixes.

D(d,n)³He Investigations

When materials are bombarded by a deuteron beam, some deuterons penetrate to their maximum range and are retained in the target material at that depth (Appendix A). Such deuterons contribute negligibly to the D(d,n) yield, since subsequent deuterons arrive there with zero energy. However, if they diffuse toward the target surface or are stopped in the target material short of the maximum range, they are available for subsequent reactions to produce neutrons. It might be pointed out that, to an order of magnitude, only one deuteron in 10^7 in the beam results in the production of a neutron; the rest are implanted in the target material, diffuse or form bands under bombardment and local heating.

Results of the D(d,n) investigations are shown in Figure 6 and Table 1. The table shows the relative yield after saturation to 90 percent of maximum, and the time required to reach this saturation for eleven different target structures.

The first run was on a fully deuterated, thick, Ti target to establish a standard for comparison.* During the first thirty minutes, neutron output decayed five percent; thereafter, the yield increased slowly to about five percent above initial. The neutron yield was about 2.0×10^8 neutrons per second over 4π . Subsequently, there were two shutdowns of a few minutes; each shutdown resulted in a few percent decrease in neutron yield, probably due to the loss of deuterium from the target surface. These results are illustrated in Figure 6.

An identical target, which had not been deuterated, was run next. Neutron yield increased like a saturation curve, ultimately to the same

* Fully deuterated refers to a deuteron loading process by which a maximum deuterium-titanium ratio is obtained.¹²

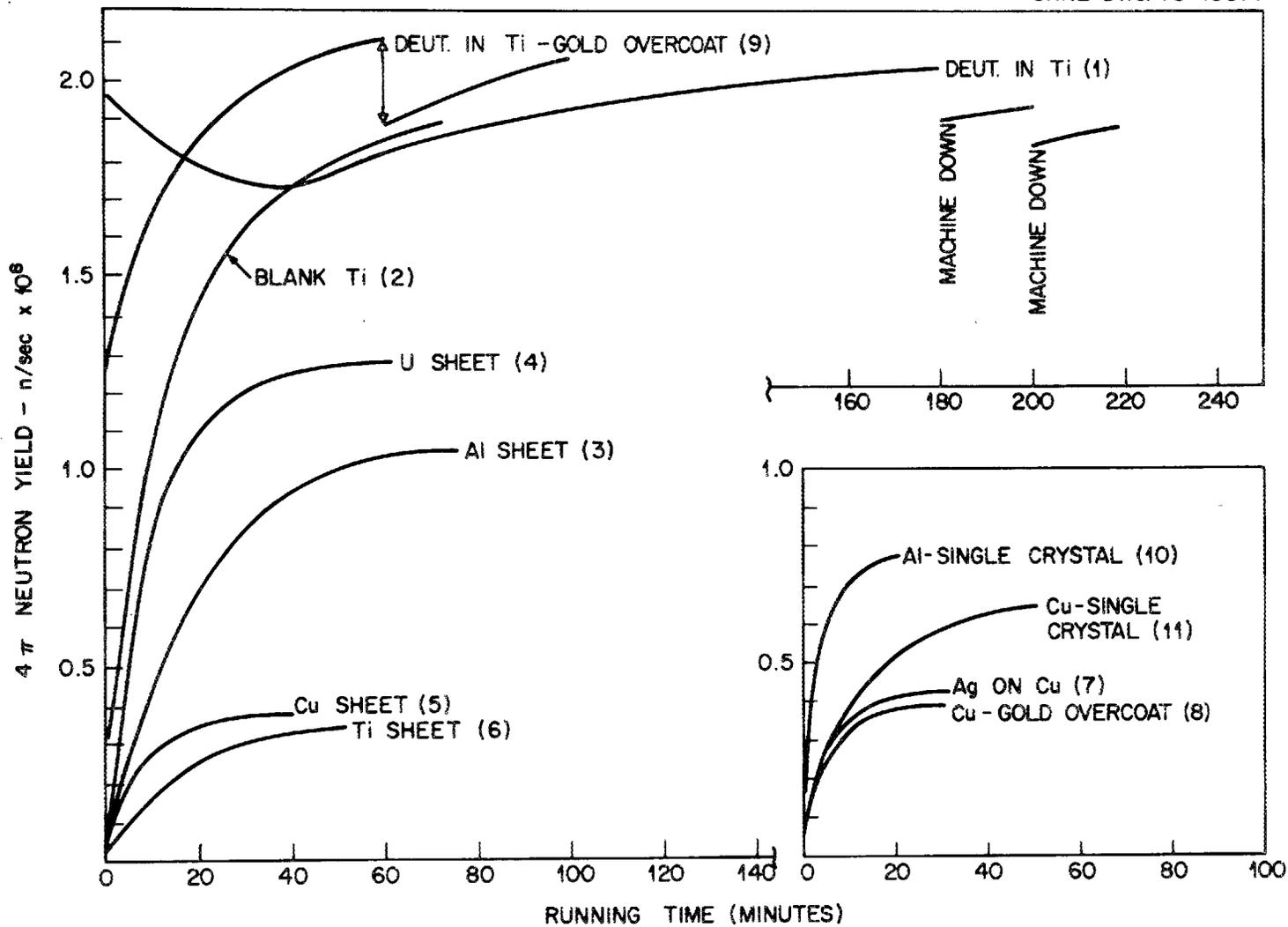


Fig. 6. Results of $D(d,n)^3\text{He}$ Reaction Experiment.

Table 1. Results of $D(d,n)^3\text{He}$ Reaction Experiment

Target	Relative (D-D) Neutron Yield	Time to 90% Saturation (min)
1. Fully-Loaded Deuterium Target, $\text{Ti}(1 \text{ mg/cm}^2)$ on copper	100	-
2. Blank Titanium Target, $\text{Ti}(1 \text{ mg/cm}^2)$ on copper	100	60
3. Plain Aluminum Sheet (40 mils thick)	48	25
4. Depleted Uranium Sheet	68	15
5. Plain Copper Sheet (13 mils thick)	19	5
6. Plain Titanium Sheet	17	30
7. Blank Silver Target ($\text{Ag}, 306 \text{ g/cm}^2$, on Cu)	23	7
8. Gold Overlayer ($100 \text{ }\mu\text{g/cm}^2$) over copper	21	10
9. Gold Overlayer ($132 \text{ }\mu\text{g/cm}^2$) over Ti on copper	113	-
10. Aluminum Single Crystal (2 mm thick)	39	10
11. Copper Single Crystal (2 mm thick)	30	15

level as the loaded target. Note that saturation time, as defined here (90 percent of maximum), is approximately 60 minutes. Buildup on a plain titanium sheet (Target 6) took 30 minutes to reach 17 percent of the yield from Target 1. A plain aluminum sheet built up in 25 minutes to 48 percent of the standard neutron yield. A plain copper sheet built up in five minutes to 19 percent of the standard. A plain uranium sheet (depleted) built up in 15 minutes to 68 percent of the standard. Silver (vacuum deposited) built up in 7 minutes to 23 percent of the standard.

There has been a common supposition that an overcoat on the target could retain the active material and still be thin enough that energy loss by the bombarding particles would not be significant. Aluminum has been used for this overcoat; gold seems a better choice because of its inert nature and ease and controllability of vacuum deposition. Indeed, a run on a gold overcoated, deuterated target did show an increase of 13 percent over the standard. However, this is well within the normal variation to be expected from one target to another. It is not known why there was a decrease in yield at one hour. A gold overcoat on a copper sheet built up in seven minutes to 21 percent of the standard.

Research at ORTEC¹³ suggested that ions might be implanted along a channeling direction in a single crystal. Average range could be expected to be several times the range in polycrystalline material, and implanted ions might be expected to be retained in these channels. The work at ORTEC had showed encouraging results. In the present experiment, a single crystal of aluminum was bombarded along the (110) direction. It built up in 10 minutes to 39 percent of the standard. However, crystal alignment along the (110) direction was not certain.

A better alignment was achieved on a single crystal of copper. The channel along the (110) direction is open over a range of angles of about ± 0.5 degrees for copper. Our alignment was within 0.25 degrees. However, this target built up in 15 minutes to only 30 percent of the standard. This result is supported by independent research by Fischer,¹⁴ in which excellent alignment along a channeling direction in copper was achieved. A 50-percent increase over polycrystalline copper is significant, but does not compete with standard targets and target materials.

To summarize the D(d,n) investigations, the materials tested differed widely in ability to retain implanted ions. Large variations in yields between polycrystalline materials and vacuum deposited materials was not expected and requires further study for interpretations. The cost of single crystals, coupled with their disappointing yields, discourages further attempts to implant ions along channeling directions. Conclusions regarding overcoats are less definite. Some yield increase was realized with gold overcoats. Future results and discussions will determine whether such marginal increase is worth the effort and cost involved.

Targets Cooled by Liquid Nitrogen

When the decision was made to study the decay of regular tritium targets at liquid nitrogen temperatures, it was decided to repeat a few of the early D(d,n) experiments of this project, but with LN cooling instead of water. A Ti blank was LN cooled and loaded by bombardment; the result is shown at the bottom of Figure 7, along with results of an identical target run at ambient temperature. Within the accuracy of the observation, no more deuterons were retained by the target at LN temperature than at ambient temperature. The target-cooling water for the

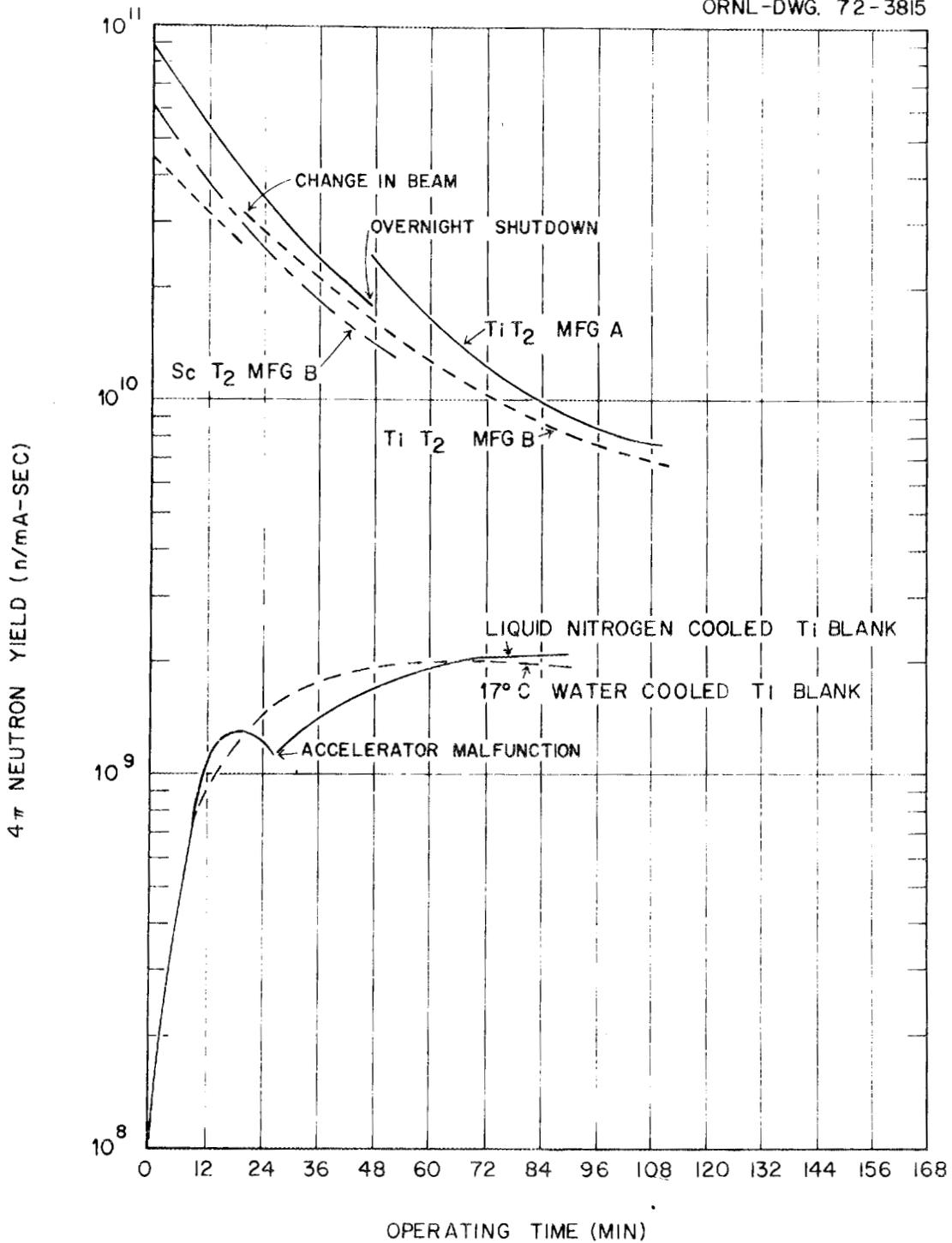


Fig. 7. Comparison of Neutron Yield from $T(d,n)^4He$ and $D(d,n)^3He$ Experiments at Ambient Temperature and at $-100^{\circ}C$.

ambient temperature target was cooled and recirculated producing an equilibrium temperature of 10°C.

Two tritium targets were run with liquid nitrogen cooling. Their results are also shown in Figure 7 (Sc and Ti). Liquid nitrogen is not a good heat transfer medium; the heat of vaporization of water is about ten times higher. Temperature was measured by the thermocouple shown in the target block in Figure 3. This temperature was kept constant at around -150°C during bombardment. This was not the temperature of the target, however. Maximum estimated target temperatures were approximately -100°C. Liquid nitrogen flow rates to maintain these temperatures were quite high. It was normal to use 160 liters of nitrogen in one hour of operation. Shutdowns, for any reason, resulted in offsets in the decay curves as seen in the D(d,n) results. This may be a result of redistribution of the available tritium in the target.

The neutron yield versus time of a standard tritium target run at ambient temperature is also shown in Figure 7 for comparison. An initial yield of about 5×10^{10} n/sec (4π) is normal. The decay curves of targets cooled by LN are, in general, parallel to those run at ambient temperature. Hence, cooling to some arbitrary temperature, much below ambient, does not significantly affect the target decay.

From this result, the conclusion is inescapable that radiation damage by the bombarding deuterons is the dominant mechanism in target decay. Chemical bonds binding the tritium in the matrix are broken, and the tritium then diffuses out of the target material. At some higher temperature, exchange processes could possibly dominate. (Ambient temperature means an input temperature of 10°C to a water coolant.)

A Pd-Ag Self-Diffusing Target

Two of the authors (Haywood and Burson) were intimately associated with the design, construction, and use of a high-yield neutron generator, which was the primary neutron source for Operation HENRE at the Nevada Test Site.⁸ As mentioned earlier in this report, targets for this generator had an active area of 1000 cm². As with all targets of this type (where the beam covers essentially the entire surface), the neutron output decayed by a factor of 2 every 45-60 minutes. The urgent need for targets with long half-lives and high yields stimulated the search for a solution to the aforementioned deficiency. The results of this series of measurements, using a silver-palladium membrane to replenish tritium in the titanium occluder, show much promise.

Three self-diffusing, Pd-Ag foil targets were fabricated and tested. The first contained a 30 mil Pd-Ag foil with 303 $\mu\text{gm}/\text{cm}^2$ of Ti vacuum deposited on it. After initial bombardment, a small air leak into the tritium system was observed. Thus, the tritium during this run was contaminated with air. The results are therefore not reproduced here. Feasibility of this technique had been demonstrated previously by J. E. Strain of ORNL;¹⁵ this run substantiated his findings.

The second target had a 20-mil Pd-Ag foil as a target substrate with 1000 $\mu\text{gm}/\text{cm}^2$ of Ti vacuum deposited on it. After an initial buildup of neutron yield to about 2×10^{10} n/sec, a general decay began (Figure 8). For a time span of 1-1/2 hours, some results are not plotted. During this interval, an effort was made to optimize parameters producing quite irregular yields. Over this period of time, tritium pressure was varied between 8 and 24.5 inches of vacuum. Neutron yield decreased by about

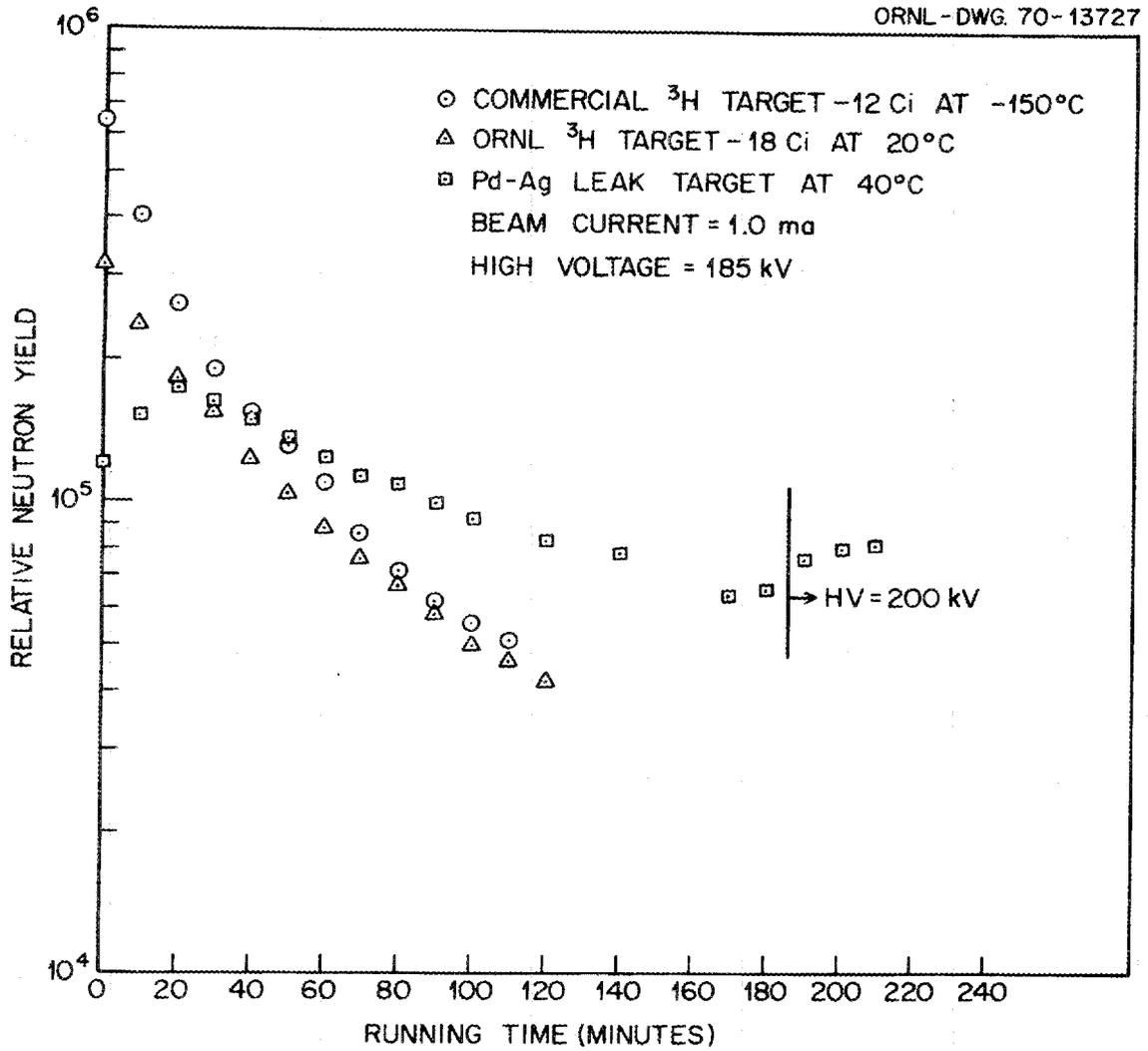


Fig. 8. Comparison of Neutron Yield from Standard Ti and Sc Targets with Neutron Yield from Prototype Self-Replenishing Ag-Pd Target.

40 percent, approximately the expected decay during the period. Hence, it was concluded that yield was not strongly related to tritium pressure. Beam energy was advanced to 200 keV for a short period, with no dramatic result. A tentative conclusion reached here was that beam density, and hence, target temperature, may have been too high for optimum operation. The target was then run for about 2-1/2 hours at a reasonably constant yield of about 10^{10} n/sec. After an overnight shutdown, the experiment was continued with no tritium behind the target; what seems to be an entirely normal target decay resulted. Two normal decay curves are reproduced on Figure 7 for comparison.

The next day, the run was continued; tritium pressure was kept as constant as possible at 2/3 atmosphere, or 10 inches of vacuum. A slowly declining yield near 6×10^9 n/sec resulted. It might be noted here that even this yield (after 7 hours) was far above what would be expected from a standard target. At the end of this period, the beam was defocused to give lower beam density and lower temperatures. This led to an increase in yield from about 5×10^9 n/sec to 1.5×10^{10} n/sec, another indication that power density may have been higher than optimum. Focusing on a new spot, rather than power density, may have also influenced the yield. Thereafter, the target was run for an hour with slowly declining yield.

At this point, the results of the two runs indicated that the titanium thickness might be very important in optimizing parameters for maximum neutron yield. The third target, therefore, contained $445 \mu\text{g}/\text{cm}^2$ of Ti vacuum deposited on a 30-mil Pd-Ag foil. Focal conditions were the same as those at the end of the previous run. Inspection showed that the

beam diameter, as seen by a burned spot on the target, was larger than minimum, but not by a large factor. Spot diameter may have increased from 6 to 8 mm, so the usual 1 mA/cm² may still be quoted within the accuracy assigned elsewhere in this report.

After the initial buildup, in about 20 minutes, the target gave a reasonably constant yield near 4.5×10^{10} n/sec for about three hours. These results were even better than expected. The yield surpassed the best "standard" target after only 16 minutes of operation. After 2-1/2 hours of operation, the yield declined with a half-life of about 2-1/2 hours. Yield at shutdown (overnight) was near 2.5×10^{10} neutrons/sec. Next morning, the yield was offset lower and declined at near the previous half-life. After one hour of operation, the target holder was displaced about 7 mm, so that the beam struck the target at the edge of the previously bombarded area. The yield immediately doubled up to about 3.5×10^{10} neutrons/sec. The yield then declined about 10 percent in two hours. For some unknown reason, the yield then declined rapidly and then became almost constant for an hour at around 2.3×10^{10} neutrons/sec.

The beam voltage was then increased to 200 kV, resulting in a few percent increase in yield. The increased range of the deuterons would produce about this much increase in yield. At 9 hours of operation, the beam was increased to 1.2 mA, resulting in a further increase in yield to about 4×10^{10} neutrons/sec. The last two data points showed a large increase. At one time, the beam was 1.5 mA. Defocusing of the beam by mutual repulsion may have caused the increase in yield, indicating that further increases may be obtained by additional defocusing. The spread of the beam probably reached portions of the target unreached before. (The collimator was not used for these runs.)

Tritium flow rates were estimated by noting the time (1.15 hr) necessary for tritium pressure to change from 8 to 24-1/2 inches of vacuum. The volume of the tritium reservoir was estimated as 1.8 cm³, and the flow was thus near 5.6 cm³/hr at STP or 12 Ci/hr. In about 18 hours of operation, about 220 Ci of activity was released into the machine. In this machine, there was a molecular sieve¹⁶ between the high vacuum pump and forepump, which absorbed a major fraction of the released tritium.

The third Pd-Ag self-replenishing target was run for a period slightly less than ten hours with a yield generally above 2×10^{10} neutrons/sec. Less than half the available target area was bombarded, indicating that sustained yields of twice this duration could be expected with this type of target.

The yield seems to be strongly dependent on the Ti thickness and much less dependent on the Pd-Ag thickness. In going from 1000 $\mu\text{g}/\text{cm}^2$ of Ti, the yield was increased by a factor of almost five. A Ti thickness such that the majority of D-T reactions take place near the Pd-Ti interface may be optimum.

The yield does not seem to be strongly dependent on tritium pressure. Variations in tritium pressure only slightly affected the yield.

Small variations in target temperature (factor of 2 in cooling water) did not influence the yield.

Three mechanisms may account for the decay in these targets:

- (1) A small layer of the target material (Ti) may have been sputtered away under bombardment;
- (2) a buildup of cracked organic material, called carbon buildup, on the target material may have occurred;
- (3) a

buildup, or replacement, of the tritium by deuterium may have occurred. It is not clear which mechanism is dominant.

SUMMARY, CONCLUSIONS, AND RECOMMENDATIONS

There is a pressing need for reliable, inexpensive, high neutron yield D-T accelerators. Fast neutrons appear to be superior to x or gamma rays for radiotherapy. Large animal radiobiology research is needed to provide indicators of radiation effects on humans. Further definitive neutron transport, secondary gamma-ray production, and shielding data for 14-MeV neutrons are needed to better define weapons radiation effects.

Machines capable of producing on the order of 10^{13} n/sec (4π) for long periods of time (30 to 40 hours), using small targets, are needed for the above tasks. The key to building such a machine is the target. The HENRE machine⁵ produced the yield, but the targets had a half-life of about an hour, and the targets were large. A brief survey of target preparations showed that there were many promising methods of preparing targets that had not been fully explored. A team, composed of ORNL and EG&G scientists, evaluated several alternative methods to target preparations.

This research has produced a prototype target that shows much promise in solving this problem. It uses the principle of self-replenishment with tritium through a silver-palladium membrane. Three targets of this type were tested; the third producing a high and sustained yield.

The results of this target are shown in Figure 9, along with the decay curve of the best "standard" target tested. The yield of the self-replenishing target surpassed the standard target after only 16 minutes

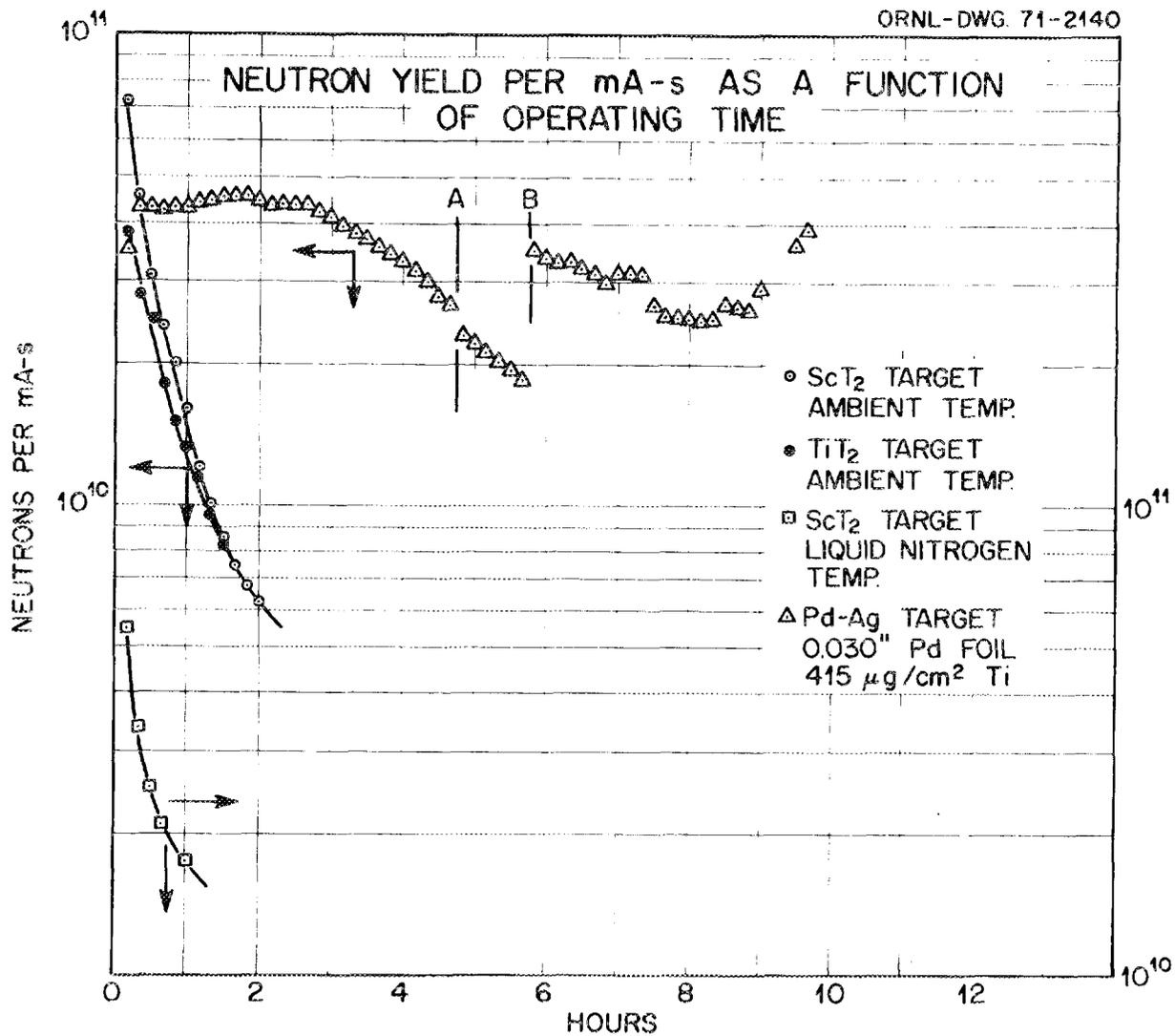


Fig. 9. Neutron Yield per mA-s as a Function of Operating Time.

of operation. The target was run for a period slightly less than ten hours with a yield generally above 2×10^{10} neutrons/sec. Less than half the available target area was bombarded, indicating that sustained yields of twice this duration could be expected with this type of target.

The yield seemed to be strongly dependent on the Ti thickness and much less dependent on the Pd-Ag thickness. In going from $1000 \mu\text{g}/\text{cm}^2$ to $440 \mu\text{g}/\text{cm}^2$ of Ti, the yield was increased by a factor of almost five. A Ti thickness such that the majority of D-T reactions take place near the Pd-Ti interface may be optimum.

The yield does not seem to be strongly dependent on tritium pressure. Variations in tritium pressure only slightly affected the yield.

Small variations in target temperature (factor of 2 in cooling water) did not influence the yield.

The tritium gas flow through a combination of materials, such as silver-palladium and titanium, is generally limited by the flow through the one with the least permeability; in this case, titanium (Appendix B). Of course, the yield is dependent upon the tritium density in the titanium and not on the flow. Optimum conditions will be obtained when the tritium density is at a maximum at the depth where most of the D-T reactions take place. This may explain why the yield was largely dependent upon titanium thickness rather than on the silver palladium thickness, its temperature or the tritium pressure.

Under idealized conditions, Gray¹⁷ indicates that a yield of 1.9×10^8 n/ μ Coulomb (@ 300 kV) may be obtained using titanium as an occluder. The third silver-palladium target reached about 3.9×10^7 n/ μ C

for 185 kV. Extrapolating upwards to 300 kV, this would be about 6.2×10^7 n/ μ C, a factor of 3 lower than that probably ever attainable. Thus, these targets, which were run to evaluate the feasibility, are not far from optimized for maximum yields.

Using deuteron beams on a variety of target materials, it was possible to show that no material, including sheet Ti, retained enough deuterons to produce D(d,n) neutrons in amounts competitive with loaded Ti targets. However, an unloaded Ti target built up activity to the same level as a loaded target. Gold overcoats produced a small increase in yield, but not significant enough to warrant further study.

Implanting ions along channeling directions in single crystal copper produced a yield 50 percent higher than polycrystalline copper, but still significantly lower than standard targets and target materials.

Supercooled tritium targets show no longer life nor enhancement in yield than those operated at ambient temperature. From this result, the conclusion is reached that radiation damage by the bombarding deuterons is the dominant mechanism in target decay. Chemical bonds binding the tritium in the matrix are broken, and the tritium then diffuses out of the target material.

The silver-palladium self-replenishing target principle offers the most potential of obtaining high yields with long half-lives. Indeed, the third prototype tested in this series of measurements could be used immediately in small machines using beam currents on the order of one milliamperere. More research and development is required to maximize available parameters and to determine if high beam densities in small targets will also be successful. These studies are necessary before an appropriate machine can be designed or built.

First, it is recommended that more experiments be performed, using low beam densities, to better define the parameters. In addition, definitive information should be obtained on: (1) Tritium density as a function of depth in the titanium and palladium; (2) tritium diffusion rates; (3) titanium sputtering; and (4) heat dissipation.

Similar experiments and investigations should be run for much higher beam densities. Experiments using low beam densities could be run at ORNL (DOSAR Facility). It is recommended that high beam density experiments be performed at another location where high beam machines with appropriate focusing and beam analysis capability exist.

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APPENDIX A

Deuteron Energy Loss in Metals

The energy loss of the beam in metals is nearly linear with thickness. A brief survey of the literature revealed theoretical calculations by Janni* to be applicable. Experimental data by Whaling** generally confirm these calculations.

Table A1 shows the range of 200 keV deuterons (or 100 keV protons) in some metals.

Some straggling will occur, of course, but, initially at least, most of the deuterons will come to rest at these depths. Note that for a titanium target, at least 400 $\mu\text{g}/\text{cm}^2$ is necessary to stop all the deuterons.

Table A2 shows approximate energy losses of 200 keV deuterons (or 100 keV protons) at various depths in aluminum, copper, gold, silver, and titanium.

* Joseph F. Janni, *Calculations of Energy Loss, Range, Pathlength, Straggling, Multiple Scattering, and the Probability of Inelastic Nuclear Collisions for 0.1 to 1000 MeV Protons*, AWFL-TR-65-150, September, 1966.

** Ward Whaling, *The Energy Loss of Charged Particles in Matter*, Handbuch der Physik, Bd. XXXIV, p. 13.

Table A1. Range of 200 keV Deuterons in Selected Metals

Material	Deuteron Range	
	$\mu\text{g}/\text{cm}^2$	μ
Aluminum	257	.95
Copper	519	.59
Gold	1250	.65
Silver	782	.74
Titanium	407	.91

Table A2. Energy Loss of 200 keV Deuterons at Various Depths in Selected Metals

Thickness $\mu\text{g}/\text{cm}^2$	Aluminum keV	Copper keV	Gold keV	Silver keV	Titanium keV
30	10	7	3	8	13
50	20	10	5	13	20
75	30	20	8	20	30
100	50	25	10	25	45
150	80	40	15	40	70
200	120	50	20	50	90
300	210	90	35	75	140
400		130	50	100	190
500		180	75	130	220
750			125	190	
1000			180		

APPENDIX B

Diffusion of Hydrogen Isotopes in Palladium

During the course of the study, some general information was gathered from the literature on the diffusion of hydrogen isotopes in metals, in particular, palladium. The more important data that was used in planning the experiment is discussed in this Appendix.

Hydrogen flow rates through palladium depends upon temperature and pressure. The higher the temperature and pressure, the greater the flow rate of hydrogen through the palladium membrane. However, even for low temperatures and pressures, the flow rate appears to be substantial. For a deuterium gas and a silver-palladium (25-75) foil of thickness 1 mil and 1 inch² in the area, the flow rate is 0.7 cc/sec at 348° C when the difference in the square roots of the upstream and downstream partial pressures is (1 psia)^{1/2}. When the differential is (10 psia)^{1/2}, the flow rate is 8.3 cc/sec (see Table B1).*

Flow rates are more for silver-palladium than for pure palladium. Flow rates are more by a factor of about 1.7 for hydrogen than for deuterium. The gas flow through a membrane is inversely proportional to its thickness.

The gas flow through a combination of materials is generally limited by the flow through the one with the least permeability. One experiment** of the diffusion of hydrogen through a composite membrane of copper and nickel with palladium indicated the flow rate through the entire membrane was dependent upon the flow rate through the copper and

* Englehard Industries, Inc. Technical Bulletin, Vol. 7, 1966.

** Barrier, R. M., *Diffusion In and Through Solids*, Cambridge Univ. Press, London, 1941, Chapter 4.

Table Bl. Experimental Results[†]

Specimen	Gas	Temp. (°C)	Corrected Flow (cc/sec at 10 p.s.i.a. ^{0.5})	Permeability (R) (cc/sec) mil/in. ² -p.s.i.a. ^{0.5}
Pd ^a	D ₂	301	2.74	0.321
		350	3.45	0.404
		400	4.41	0.516
		448	5.26	0.615
		499	6.19	0.724
		550	6.96	0.814
		596	8.43*	0.986
	H ₂	305	5.89	0.689
		350	6.19	0.724
		400	7.25	0.848
		450	8.35	0.977
		500	9.87	1.155
		548	11.36	1.329
		596	12.84	1.502
Pd-25 Ag ^b	D ₂	348	8.32	0.718
		403	9.22	0.796
		449	9.23	0.797
		500	9.84	0.849
		598	11.46	0.989
	H ₂	350	14.21	1.226
		403	14.42	1.244
		449	15.36	1.326
		500	16.05	1.385
		598	18.67	1.611

* Extrapolated. Last data point at 8.4 p.s.i.a.^{0.5}.

- a. 10.00" x 0.124" O.D. at start of experiment.
 Effective area 3.76 in.², 4.6-mil wall.
 0.130" O.D. x 4.44-mil wall at end of experiment.
 Averaged ratio wall thickness/area, 1.17 mil/in.².
 The averaged values were used to calculate permeability.
- b. 10.05" x 0.126" O.D.
 Effective area 3.82 in.², 3.3-mil wall.
 Wall thickness/area ratio, 0.563-mil/in.² throughout experiment.

†Technical Bulletin, Vol. 7, Engelhard Industries, Incorporated, 1966.

nickel alone. One would expect then, that the flow through a sandwich of silver palladium and vacuum-deposited titanium would depend upon the flow through the titanium. Very little information was found on the flow through titanium.

For low temperatures (below 300° C), it is important that the hydrogen isotope be reasonably pure. Many substances are adsorbed preferentially over hydrogen through palladium at these low temperatures. One of the targets tested used tritium contaminated with air. This may explain the low yield.

The atom ratio, H/Pd, is inversely proportional to temperature and varies from about 0.1 to 0.7. Of course, if the gas is tritium being diffused into a titanium substrate, bombardment by deuterium will produce a neutron yield dependent upon the tritium density in the titanium and not in the palladium. Optimum conditions will be obtained when the density of tritium in the titanium is at a maximum at the depth where most of the D-T reactions take place. This probably explains why the yield was largely dependent upon titanium thickness rather than on the silver palladium thickness, its temperatures, or the tritium pressure.

APPENDIX C

Sputtering in Single Crystals

Two articles that presented a fairly clear picture of the sputtering process were reviewed. Impinging ions collide with target atoms. These atoms tend to recoil in the forward direction with sufficient energy to undergo secondary collisions with other atoms. This second generation of recoil atoms has a greater chance of escaping in the backward direction than the first generation. These are of very low energy, but there are so many of them they account for the major portion of the sputtering yield. However, they cannot escape the surface unless they are originally located within a couple of atomic layers from the surface.*

Consequently, the deeper the impinging ions can penetrate, the smaller the sputtering yield. The result is: The sputtering yield decreases with increasing ion energies and increases with an increase in ion mass. Deuterium ions bombarding single crystal copper at normal incidence at 1 MeV energy yields a sputtering ratio of about 10^{-3} atoms/ion. Kaminsky** has given values of sputtering of copper under bombardment by deuterons. The sputtering ratio (atoms/ion) at 150 kV is about 2×10^{-3} , and increases to 2.7×10^{-3} at 100 kV. At 150 kV, an ion beam of 1 mA, bombarding a single crystal copper target for 78 minutes, would sputter off 5.6×10^{16} atoms of copper ($2 \times 10^{-3} \times 2 \times 8 \times 10^{19} = 5.6 \times 10^{16}$). If the target is 1 cm^2 , this would represent a removal of 6.6×10^{-3} microns off the surface.

* Peter Sigmund, Phys. Rev. 184, 2, 383 (1969).

** Manfred Kaminsky, Phys. Rev. 126, 4, 1267 (1962).

APPENDIX D

A Simplified Method of Estimating the D-D Component of a D-T Source

In all D-T accelerators, there are some neutrons emerging from the D-D reaction. These usually range from 1 to 10% of the neutron output. There are many instances in which an estimate of the D-D component in real time is desirable.

A simplified method was developed for estimating the D-D component of a D-T source. It was not used, however, in these experiments because baseline data on the D-D neutron yield had been established for the targets of interest and because extreme accuracy was not required in these experiments. The method is presented here for informational purposes.

The angular distribution of the neutrons emerging from the D-D reactions is peaked in the forward direction, whereas those from the D-T reaction are nearly isotropic. (See Figure D1.) To take advantage of this directionality, if a neutron counter is placed at 0° and another at about 90° , an estimate of the D-D component can be made by examining the ratio of the counts. Standard, insensitive, long counters or Hurst proportional counters would suffice, provided the counters record the same number of counts per neutron of the two reactions.

The first counter should be placed at 0° . The second counter could be placed at an angle of 75° . Too often the target provides some self-shielding at 90° .

The target-counter distance of the second counter could be adjusted such that the two counters read the same for the D-T neutrons. This distance (for the 75° detector) should be closer than the 0° detector

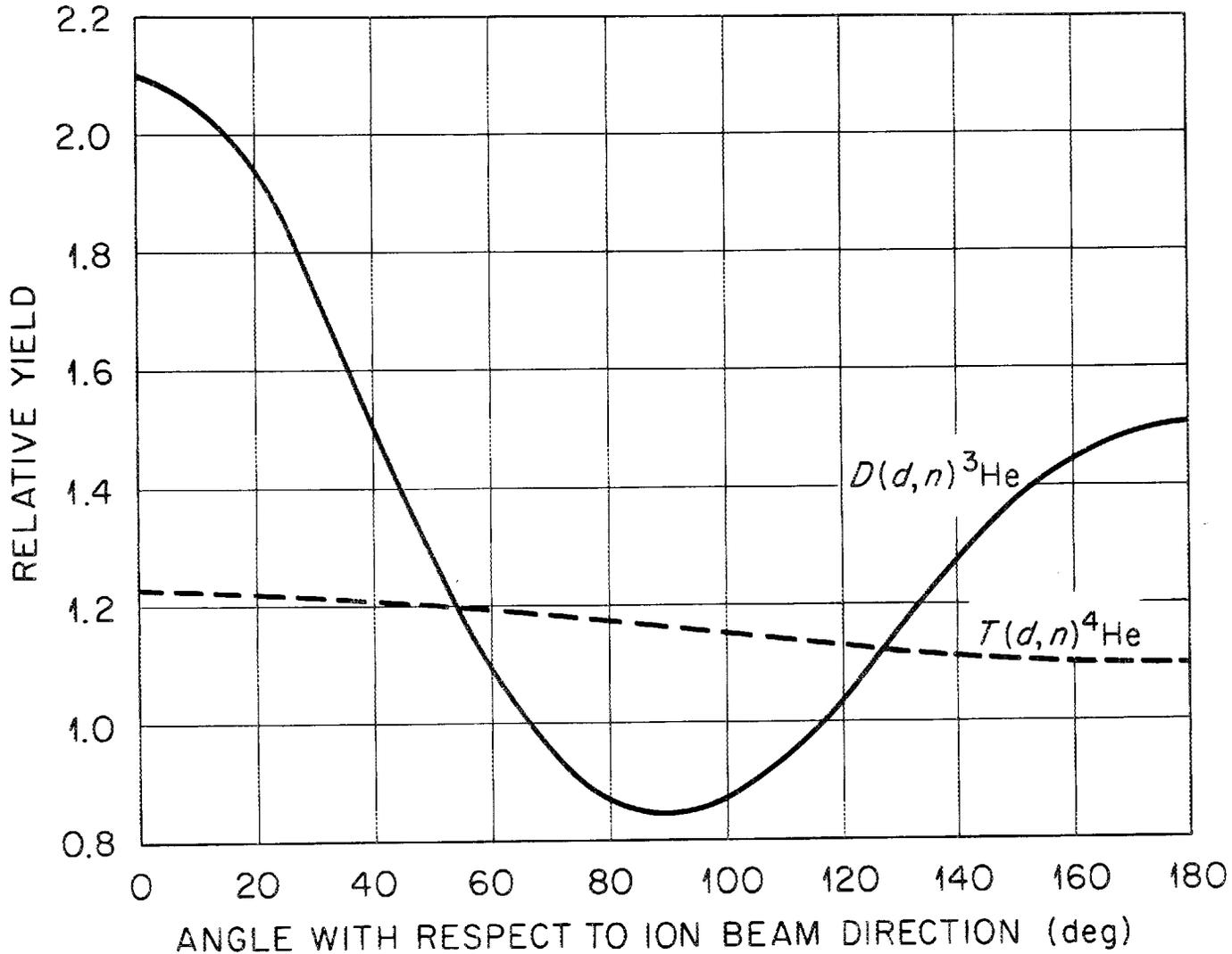


Fig. D1. Relative Directional Yield for A Thick Titanium Target - 200 kV Beam.

by 98.4%, 98.2%, 98.0%, 97.8%, 97.7%, and 97.6% for deuteron energies of 100, 200, 300, 400, 500, and 600 keV, respectively.

Next, a blank titanium (or other material) target could be put on the accelerator to produce only D-D neutrons. The ratio of counts of the two detectors should be as listed in Table D1.

Values in Figure D2 can be used for other accelerating voltages. If these counter ratios are not obtained, the source-detector distance should be adjusted until the ratios are obtained.

With the detectors at the proper distances and angles, the D-D component of the fluence at 0° for any target can be determined. Table D2 gives ratios for counter readings for various beam compositions at 0° . Other tables could be developed for other angles or specific accelerating voltages from the basic data in Seagraves* handbook. Still other tables could be developed for the 4π neutron outputs by relating the 4π output to the 0° fluences if desired.

Note that from Table D2 for a 200 kV beam, a 1% greater fluence at 0° shows a 2% D-D component. It should be easy to obtain results much less than 1% relative accuracy by obtaining 100,000 counts or so on the scalers. It is, of course, important that the two scalers record during an identical time interval.

This method would work best when neutron scattering from materials in the room are minimized.

* John D. Seagrave, *D(d,n)He³ and T(d,n)He⁴ Neutron Source Handbook*, LAMS-2162 (January 24, 1958).

Table D1. Ratio of Neutrons in Forward Direction to Those at 75° as a Function of Deuteron Energy

Energy keV	Ratio - C_{0°/C_{75°
100	1.872
200	2.249
300	2.661
400	3.015
500	3.352
600	3.656

Table D2. Relative Ratios of the 0° Neutron Fluence to the 75° Neutron Fluence for Various Beam Compositions

Beam Compositions at 0° (%)		Accelerating Voltage - kV					
D-T	D-D	100	200	300	400	500	600
100	0	1.000	1.000	1.000	1.000	1.000	1.000
99	1	1.0047	1.0056	1.0063	1.0067	1.0071	1.0073
98	2	1.0094	1.0112	1.0126	1.0135	1.0142	1.0147
95	5	1.0238	1.0286	1.0322	1.0346	1.0364	1.0377
90	10	1.0489	1.0588	1.0666	1.0716	1.0755	1.0783
80	20	1.1027	1.1250	1.1426	1.1543	1.1632	1.1700
70	30	1.1624	1.1999	1.2304	1.2508	1.2666	1.2787
50	50	1.3036	1.3844	1.4537	1.5019	1.5404	1.5704
25	75	1.5369	1.7139	1.8802	2.0050	2.1108	2.1971
0	100	1.872	2.249	2.661	3.015	3.352	3.656

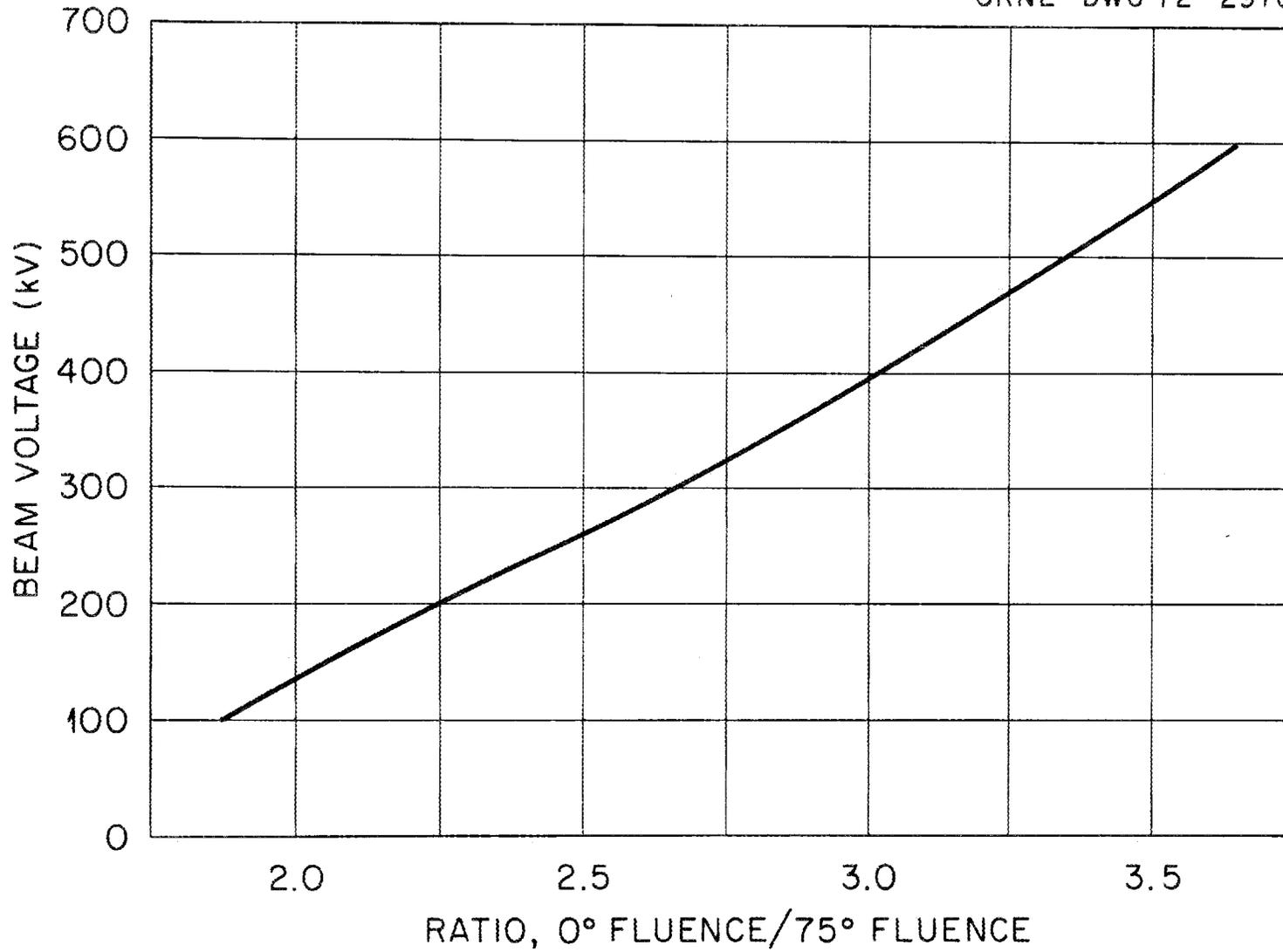


Fig. D2. Ratio of 0° Fluence to 75° for the $D(d,n)^3He$ Reaction when the Fluences from the $T(d,n)^4He$ Reaction at the Same Positions Are Equal.

1

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