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A HIGH YIELD NEUTRON TARGET FOR CANCER THERAPY

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A HIGH YIELD NEUTRON TARGET FOR CANCER THERAPY

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Summary

A rotating target has been developed that has the potential for providing an initial yield of 10^{13} neutrons per second by the $T(d,n)He^4$ reaction, and a useable lifetime in excess of 600 hours. This yield and lifetime are indicated for a 300 KV and 30 mA deuteron accelerator and a 30 μ m thick titanium tritide film formed of the stoichiometric compound TiT_2 . The potential for extended lifetime is made possible by incorporating a sputtering electrode that permits use of titanium tritide thicknesses much greater than the deuteron range. The electrode is used to remove in situ depleted titanium layers to expose fresh tritide beneath. The utilization of the rotating target as a source of fast neutrons for cancer therapy is discussed.

Introduction

Fast neutron radiation effects on cancer cells have been under investigation for many years and treatment of tumors in human patients has been taking place in Hammersmith Hospital in London since 1967 with encouraging results.^{1,2,3} Accumulated evidence indicates that this type of radiation offers certain advantages over X- or gamma-rays for radiotherapy treatment of deep-seated tumors.¹ However, few neutron sources are available today that produce a sufficient neutron intensity for this purpose. A further problem, concerning accelerators that utilize titanium tritide targets and the $T(d,n)He^4$ reaction for neutron production, is the generally short useable lifetime of these targets.

A recent experimental study was conducted that compared currently available neutron sources to determine which source was best suited for cancer therapy use.¹ The sources surveyed included a fission reactor, three cyclotrons of various sizes that utilized the $Be^9(d,n)B^{10}$ reaction, and a deuteron accelerator that produced neutrons by the $T(d,n)He^4$ reaction. The study concluded that a deuteron accelerator that produces 2.24 pJ (14 MeV) neutrons at an initial rate of at least 4×10^{12} neutrons per second was the best choice of neutron source. A minimum target lifetime of 30 hours was suggested.

Targets designed for neutron yields greater than 10^{12} neutrons per second usually have rotating surfaces to distribute the beam over a large area in order to reduce the power density.^{4,5} The temperature of the titanium tritide must not be allowed to exceed the decomposition temperature. The increased area, however, also provides a large inventory of titanium tritide for extended target lifetime. Neutron yields as high as 2×10^{12} neutrons per second and a half-life of 400 mA-hr (50 hrs at 8 mA) have been attained for a rotating disk target.⁴

We chose a rotating cylinder configuration that has a large target area, but our design also provides for use of a target film much thicker than the range of the deuteron beam. A sputtering electrode permits full use of this thick film by removing depleted titanium layers to expose fresh tritide beneath the depleted

region. The advantage of this configuration is that one thick titanium tritide layer is equivalent to several thin layers with each thin layer having a thickness equal to the range of the accelerated deuteron. The problems presented by the thick target are twofold. The target must be (1) adequately cooled, and (2) the sputtering of the target surface must not degas tritium from the film.

The purpose of this report is to present a description of the target we have developed and to report the work that we have done toward solving the above problems.

Target Description

Design Description

A neutron producing target system, which we have identified as the High Yield Neutron Target (HYNT), is designed for use with a 300 kV and 30 mA deuteron accelerator. The objective of the design is to provide a tritium loading of sufficient capacity to give a useful neutron production lifetime that approaches the indicated 2000 hour lifetime of the target's high vacuum Rulon-A* seals. These seals are the most critical mechanical component of the system.

In order to achieve the required titanium tritide target lifetime in a reasonable sized system, a cylindrical active area of 547 cm^2 is provided which can be coated with titanium tritide to a thickness of 30 μ m. For full utilization of the tritide layer, which is thicker than the range of the beam deuterons, a sputter electrode is provided to remove titanium surface layers that have been depleted of tritium. For accurate measurement of the tritide target layer remaining, a film thickness monitor is incorporated into the design.

To keep the tritide well below its decomposition temperature, a high water coolant flow rate of $2.21 \times 10^{-3} m^3/sec$ (35 gallons per minute) is provided. A narrow 1 mm wide passageway beneath the copper target substrate is used to ensure turbulent flow for maximum transfer rate of beam energy to the coolant. Finally, the target is made in the form of a cylinder that rotates at a speed of 1100 rpm to distribute the beam energy over the surface of the cylinder.

Description of Target System

A complete description of the HYNT is given in Ref. 6. Figure 1 is a drawing of the HYNT showing the coolant passageways, the sputtering electrode, and the motor. The motor is electrically insulated from the target housing. Figure 2 shows the HYNT, including the thickness monitor, attached to the beam tube of the 300 KV accelerator. A description of the thickness monitor is given in Ref. 7. The replaceable copper target cylinder is also shown in Fig. 2.

The entire target drum can be easily removed from the housing as shown in Fig. 3. The ball bearings and seals are secured in the endplates and are removed with them. A view of the water inlet endplate is shown in

*Trade name of Sealol Corporation for a teflon-glass composition.

Fig. 4. This view shows the carbon water face seal in the background, the ball bearing, and the Rulon-A high vacuum seal in the foreground.

The motor drive end of the target drum, showing the three stainless steel seat rings, can be seen in figure 5. The large ring mates with the Rulon-A high vacuum seal, the two smaller rings mate with carbon water seals. The seal seats are secured to the aluminum drum by an epoxy adhesive, then ground and lapped flat. The copper target cylinder is a forging of a copper-zirconium alloy.

The sputtering electrode can be seen in Figs. 1, 2, and 3. The electrode dimensions, shield spacings, and titanium deuteride etch rates were measured⁸ using an experimental mock-up of the HYNT.

Component Evaluation and Testing

1000 Hour Seal Test

The lifetimes of the Rulon-A and carbon seals of the HYNT were determined by rotation of the target cylinder at 1100 rpm, under accelerator vacuum conditions, for a period of 1000 hours. The air leak past the Rulon-A vacuum seals during this test was 2×10^{-10} m³ (STP)/sec. Details of the seal test are given in Ref. 6.

The original thickness of the Rulon-A and carbon seal material was 0.152 cm. After the 1000 hour test, slightly more than half of the Rulon-A material remained, while the carbon seals showed a wear of only 0.012 cm. Therefore, the maintenance free lifetime of the HYNT, determined from the Rulon-A seal wear, can be expected to be about 2000 hours.

Sputtering Electrode Evaluation

The purpose of this electrode is to remove tritium depleted layers of titanium to expose the fresh tritide material beneath. We were concerned, however, that possibly the act of sputtering itself might cause degassing of tritium from the target. We set out to prove that such degassing did not occur. For this purpose a 3.45 μ m thick titanium film was plated onto the copper target cylinder.⁶ A titanium deuteride target was then formed by deuteron implantation with an accelerator voltage of 250 kV at a deuteron beam current of 5 mA.⁶ The beam is composed of approximately 60 percent D₁⁺, 20 percent D₂⁺, and 20 percent D₃⁺ deuterium ions. Figure 6 shows the approximate implanted position of these ions in the titanium target film. The titanium deuteride composition in the target film was approximately TiD_{0.53} after the implantation.

Removal of the 3.45 μ m thick titanium deuteride film was done with the sputtering electrode.⁶ Sputtering was done at an argon pressure of about 5 N/m² with a voltage of from 2 to 5 kV applied to the electrode. The beam tube was isolated from the accelerator vacuum system by a gate valve during the sputtering.

Each sputter removal of a thin layer of the deuteride was followed by a brief probing with a 1.5 mA accelerator beam of deuterons having an energy of 0.010 pJ (100 keV), and a thickness measurement of the deuteride film remaining using the X-ray thickness monitor described in Ref. 7. The diameter of the probe beam was less than 2 cm or approximately half the diameter of the beam used for deuteron implantation. The neutron output due to the deuterons by the D(d,n)He³ reaction was monitored by a BF₃ counter. At 100 kV potential, only the D₁⁺ ions contributed to the production of neutrons.

A plot of the neutron output produced by the deuteron probe, as a function of target thickness, is shown in Fig. 6. The length of the line extending to the right of each datum point is the approximate range of the D₁⁺ deuteron (1.35 μ m) in the film. The deuterium distribution appears to fall off toward the copper substrate. However, the rapid fall off in neutron count rate at film thicknesses near the copper substrates does not represent the true deuterium distribution, because a fraction of the D₁⁺ ion path length is in the copper substrate. The neutron count rate per unit of path length of the D₁⁺ probe ion in the titanium deuteride is approximately the same as that of the front layers of the target. A reasonably flat deuterium distribution is indicated. Therefore, it is evident that the sputtering has not caused any significant degassing of deuterium from the titanium deuteride.

TiD₂ Film

During the building and testing of the HYNT, we conducted a companion research program aimed toward the formation of a continuous TiD₂ film. We succeeded in forming a continuous film of TiD₂ that bonded to copper as well as some other substrate materials. Film thicknesses of greater than 30 μ m were formed.

During the TiD₂ development program, films were formed that varied in composition from TiD_{0.7} to TiD₂. Photomicrographs taken of some of these surface films illustrate a contrast in surface appearance between the two phase titanium/titanium deuteride structure and the single phase titanium deuteride structure. For example, Fig. 7 shows a replica of the surface of a TiD_{1.03} film. Both titanium metal and titanium deuteride phases coexist in the film. Note the warped and rough appearance of the surface. The crystallites of this specimen ranged in size from 0.1 to 1 μ m. Now compare this surface with Fig. 8 which shows the surface of a TiD_{1.76} film, and Fig. 9 which shows a TiD_{1.85} film at higher magnification. Both photomicrographs illustrate the continuous nature of the near TiD₂ films. The crystallites of these films ranged in size from 0.1 to 0.6 μ m and showed a uniform growth pattern.

The electrical resistivity of a TiD₂ film was measured to be 60×10^{-6} ohm-cm. Reported resistivity values for pure titanium vary over a range of $80-42 \times 10^{-6}$ ohm-cm.⁹ Therefore the electrical resistivity of the TiD₂ film is approximately the same as for the bulk titanium metal. A value for the thermal conductivity of the TiD₂ film of 0.12 W/cm K is calculated in Ref. 6 using the measured electrical resistivity value and the Wiedemann-Franz law.¹⁰ The calculated thermal conductivity of TiD₂ is an approximate agreement with a near room temperature thermal conductivity value for bulk titanium of 0.154 W/cm K (at 320 K).⁹

Heat transfer calculations, using the thermal conductivity factors described in the previous paragraph, were made in Ref. 6 for a beam power density of 400 W/cm² and a 30 μ m thick TiD₂ film on the 1350 μ m copper substrate. This power input caused a maximum target surface temperature rise of 40 K.

Assuming a target cooling water temperature of 275 K, the HYNT TiD₂ surface temperature at a 400 W/cm² beam power input will be approximately 315 K. The dissociation temperature will limit the allowable target surface temperature of the titanium deuteride. Extrapolation of data from Ref. 11 indicates a dissociation temperature for TiD_{1.5} of approximately 450 K at a pressure of 5×10^{-4} N/m². This is the approximate pressure existing at the target during operation. If we

assume a dissociation temperature of 450 K, our target surface temperature can be increased by 135 K over the surface temperature caused by the 400 W/cm² power input. Or, we will have a total allowable surface temperature rise of 175 K above the temperature of the cooling water.

A 30 mA deuteron beam, accelerated by 300 kV potential, will deliver 9 kW of power to the target. If the 9 kW is contained in a circular beam spot with a beam power density of 400 W/cm², the beam spot diameter will be 5.4 cm.

Discussion

Deuterium rather than the radioactive tritium was used in our work because it was necessary at this time that we avoid radioactive contamination of the accelerator system. Since deuterium and tritium are chemically identical, the results we have found for the titanium deuteride compound should be directly applicable to the titanium tritide compound.

Potential Yield and Lifetime of HYNT

Our 300 kV deuteron accelerator has a beam ion composition of 60 percent D₁⁺, 20 percent D₂⁺, and 20 percent D₃⁺. Using a 30 mA deuteron beam of this composition and the neutron yield data for the T(d,n)He⁴ reaction on a TiT₁ target film, a maximum theoretical 2.24 pJ (14 MeV) neutron yield of 5.4x10¹² neutrons per second is calculated.¹² Use of a TiT₂ film instead of the TiT₁ film could potentially increase the maximum yield to 1x10¹³ neutrons per second.

Booth⁴ has published lifetime data for a titanium tritide target with an initial yield of 2x10¹² neutrons per second. The target was in the form of a disk which rotated at 1100 rpm. The half-life of the target was 3.3 mA-hr/cm². Using this half-life value, our 30 mA beam current, and the total useable area of the HYNT, an estimated half-life for one target layer, with a thickness equal to the D₁⁺ ion range, is calculated.

$$\text{HYNT half-life (one target layer)} = 3.3 \text{ mA-hr/cm}^2 \times \\ 547 \text{ cm}^2/30 \text{ mA} = 60 \text{ hours}$$

We have calculated the range of a 0.048 pJ (300 keV) D₁⁺ ion in the titanium tritide film to be about 3 μm by using data of Gunnerson, et al.,¹³ and Warshaw.¹⁴ A 30 μm thick TiT₂ film is sufficient for 10 target layers.

The TiT₂ film on the HYNT cylinder has been designed to accommodate two 5 cm diameter beam spots. The beam will be absorbed on a band, the width of the beam spot, on the rotating target surface. A flexible bellows section should be installed in the beam tube a few feet from the HYNT. The HYNT is then supported by a mechanism that provides lateral movement of the entire HYNT along the axis of the cylinder relative to the beam. The mechanism should include a position indicator and be operated remotely. By this method, the beam can easily be moved to fresh target material whenever maximum neutron output is needed. The beam can then be returned to the used target band for use with experiments that do not require the high neutron output.

Potential Use of the HYNT for Cancer Therapy

The Brennan study¹ concluded that the best neutron source for cancer therapy was a deuteron accelerator that produced 2.24 pJ (14 MeV) neutrons at an initial rate of at least 4x10¹² neutrons per second. A minimum target lifetime of 30 hours was suggested.

The HYNT is capable of surpassing the neutron yield and lifetime requirements set forth by Brennan. The targets indicated 2000 hour vacuum seal lifetime and its ability to use thick, large area target films has been demonstrated. TiD₂ target films that adhere well to the substrate and can be adequately cooled have been made. For purpose of illustration of the capability of the HYNT, we will assume that our 300 kV and 30 mA accelerator will be used, in conjunction with the HYNT with a TiT₂ film, for fast neutron radiotherapy purposes.

Assuming an initial yield of 1x10¹³ neutrons per second, the target will be useful for only about one half-life, or until the tritium loss has reduced the yield to 4x10¹² neutrons per second. As described in the previous section, however, the target cylinder can accommodate two bands the width of the beam spot. One fresh band of the target can be used for cancer therapy treatment of patients until the neutron yield has dropped to an unacceptable level of approximately 4x10¹² neutrons per second. This neutron yield is still useful for radiobiological experiments, or for the production of radioisotopes for medical research or radiotherapy applications. The accelerator can be run continuously on this partly depleted target band for several more half-lives until the neutron yield is no longer useful. Meanwhile, irradiation of patients at higher neutron levels can continue by simply displacing the beam to the other fresh target band during the irradiation. Following each patient irradiation, the beam spot will be immediately moved to the used target band for continued irradiation of experiments. When the entire cylinder surface has been depleted of tritium to the depth of the beam deuteron, the titanium of the surface will be removed by the sputtering electrode until a fresh TiT₂ film is again exposed. The irradiation procedure will then continue as before.

A 30 μm thick TiT₂ film is sufficient for 10 target layers and will have a useful lifetime for radiotherapy treatment of patients (one half-life) of approximately 600 hours. If we assume that the target is useful for at least two more half lives (1200 hr) for other irradiations as described in the preceding paragraph, the overall useful target life will approach 2000 hours, the indicated lifetime of the Rulon-A seals. The long lifetime is important for hospital applications since most hospitals do not have the facilities required to replace a tritium contaminated target cylinder. Target changing would occur at the same time the seals, bearings, and associated hardware would require changing. It would be advisable to have, as a minimum, two complete target assemblies so that the entire HYNT can be exchanged. Irradiations can then continue again in a matter of a few hours. The used HYNT may then be rebuilt at leisure.

Conclusions

The design of the HYNT has been presented along with the results of component tests. Specifically, we have demonstrated the following. Our seal lifetime tests have shown that, mechanically, the HYNT has an indicated lifetime of about 2000 hours at 1100 rpm. We have shown that a continuous film of TiD₂ can be made to a thickness of 30 μm. Our heat transfer calculations indicate that a 30 mA and 300 kV deuteron beam (9 kW of power) impinging on a 5.4 cm diameter spot of the rotating HYNT cylinder, coated with a 30 μm TiD₂ film, will produce only a 40 K rise in target surface temperature. We have demonstrated that the sputtering electrode of the HYNT can effectively remove deuterium-depleted titanium layers to expose fresh underlying layers of titanium deuteride. Furthermore, it has been shown that this can be accomplished without degassing

deuterium from titanium deuteride beneath the target surface. The method permits utilization of the full 30 μm Tid_2 target film.

The HYNT has the potential of providing a neutron yield, by the $\text{T}(\text{d},\text{n})\text{He}^4$ reaction, of 10^{13} neutrons per second. This yield is indicated for a 300 kV and 30 mA deuteron accelerator and titanium tritide film formed of the stoichiometric TiT_2 compound. The neutron yield is based on theoretical yields for the $\text{T}(\text{d},\text{n})\text{He}^4$ reaction. For fast neutron radiotherapy applications, a mode of operation can be chosen whereby the HYNT can have a useful life approaching the 2000 hour lifetime of the Rulon-A seals. In this mode, only about 600 hours of the target life is used at the high neutron production levels required for radiotherapy treatment of patients. The remaining target life is used at lower neutron levels for radioisotope production or other experiments. Therefore, target changing can occur at the same time that the HYNT will be removed for seal and bearing replacement.

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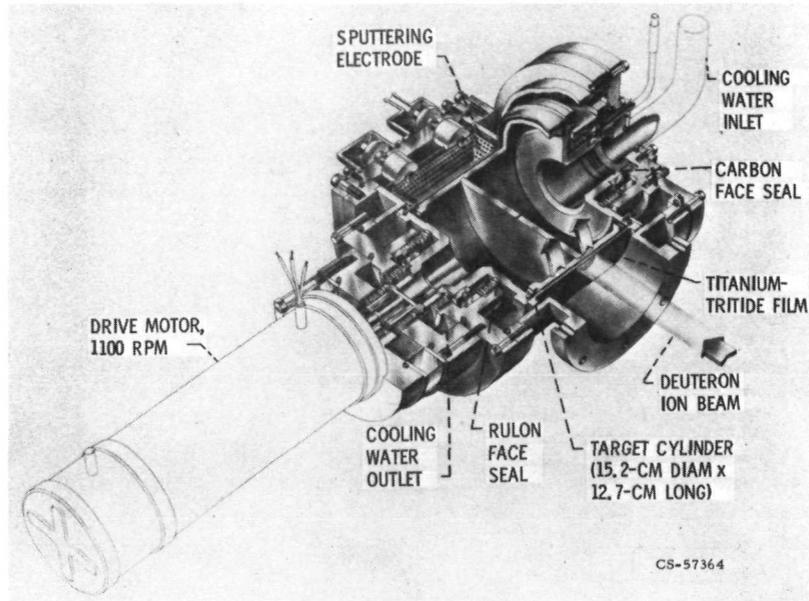


Figure 1. - The high yield neutron target (HYNT).

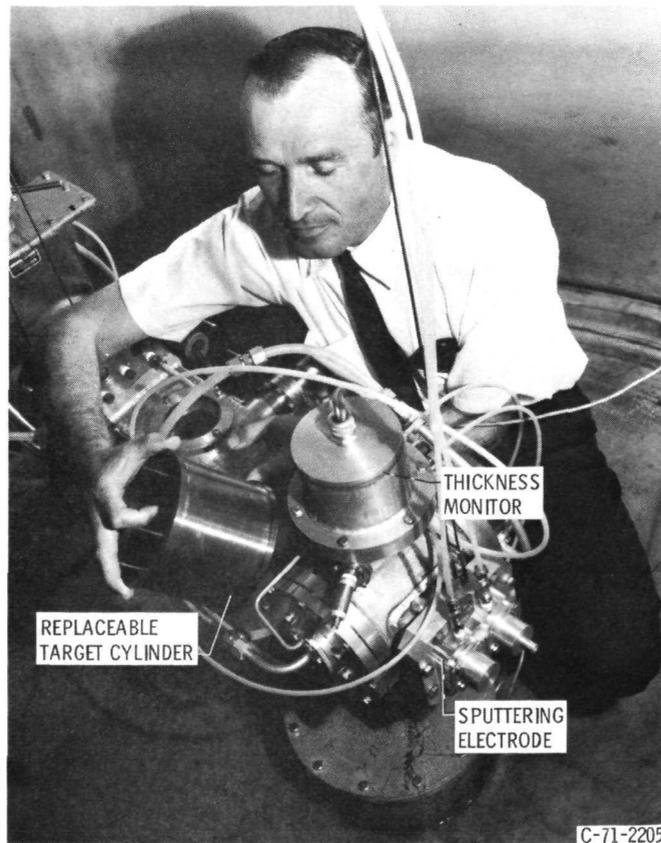


Figure 2. - The HYNT attached to the 300 KV, 30 mA accelerator beam tube.

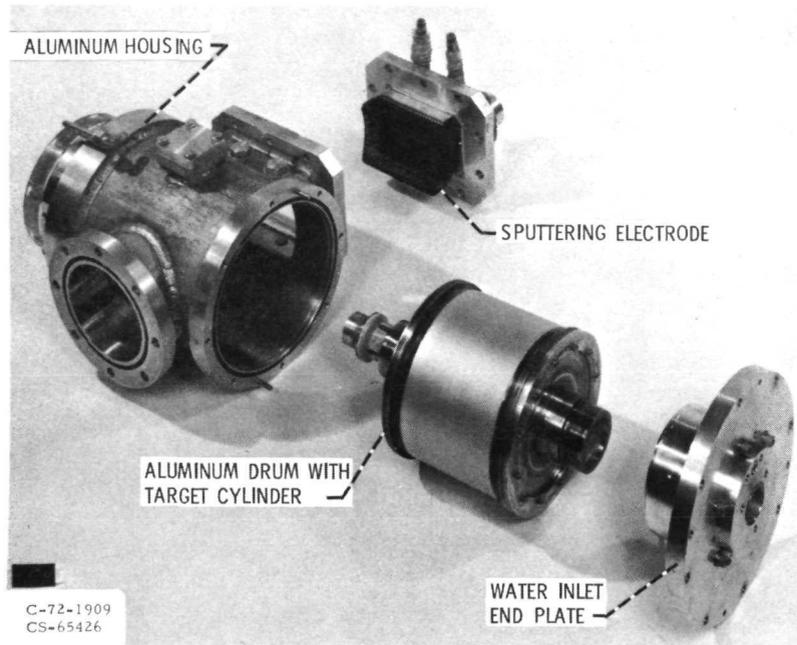


Figure 3. - Dissassembled view of HYNT less motor and thickness monitor.

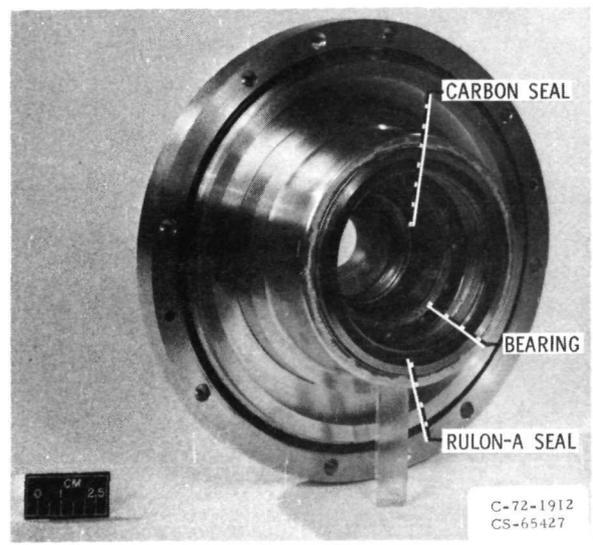


Figure 4. - Water inlet endplate showing installed seals and bearing.

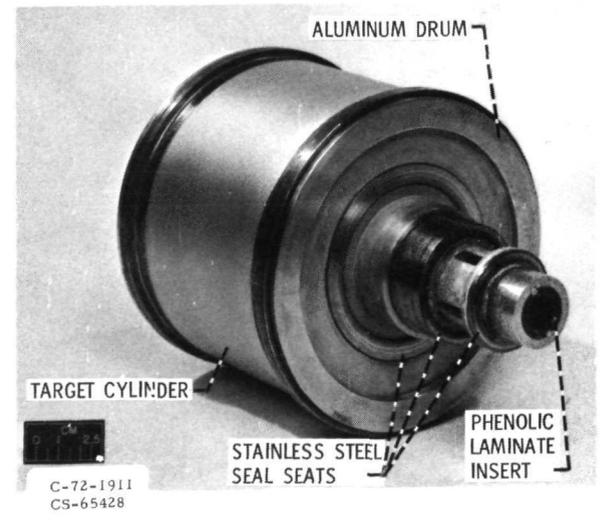


Figure 5. - Motor drive end of aluminum drum with target cylinder attached.

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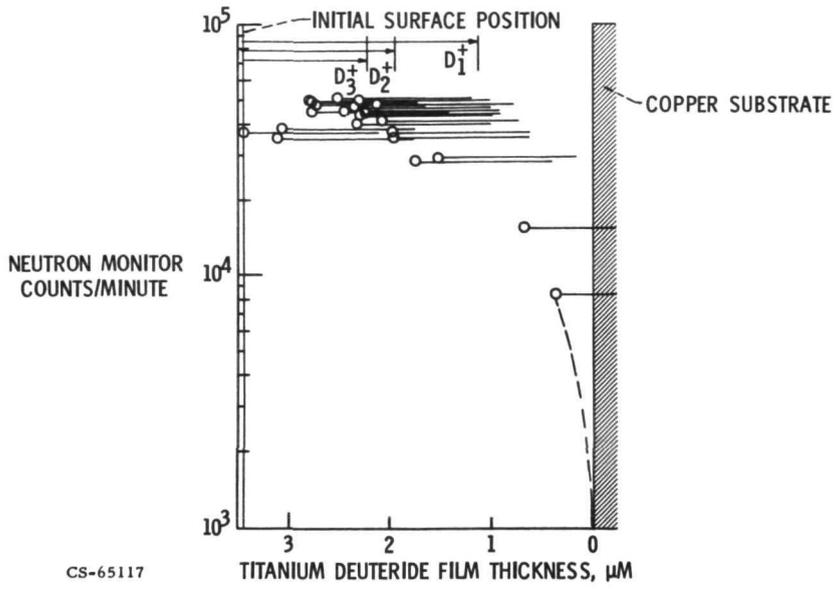


Figure 6. - Arrows indicate depth of implantation of deuterium ions. Each neutron count rate datum point is plotted at the deuteride surface. The line after each datum point represents range of deutron probe in deuteride (1.3 μM).

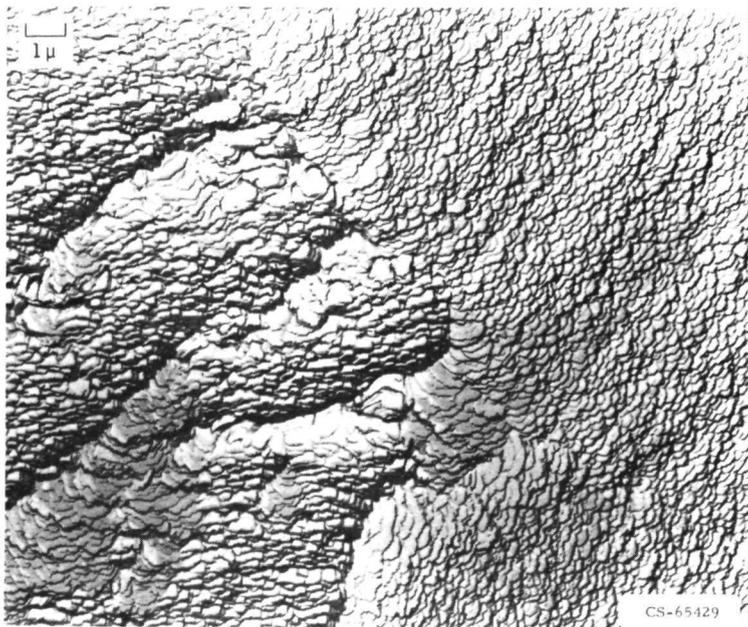


Figure 7. - A 3 micron thick film of $\text{TiD}_{1.03}$ composition.

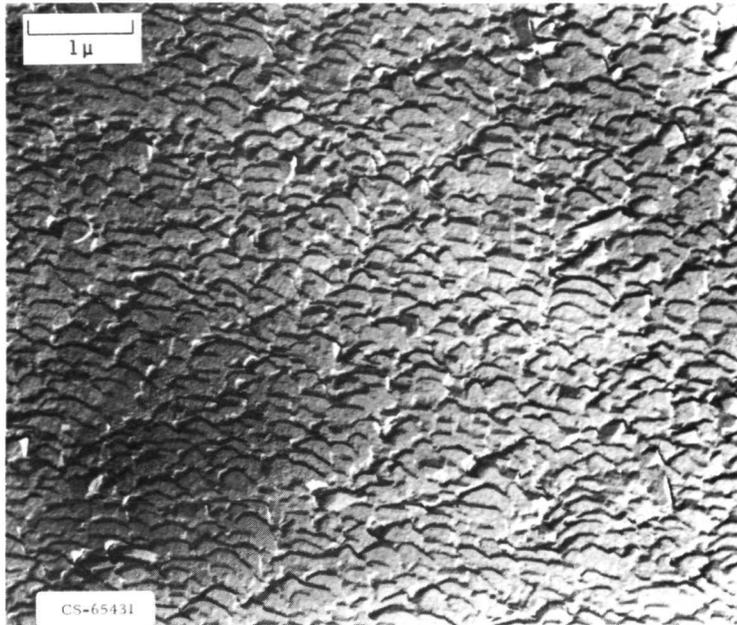


Figure 8. - A continuous 3 micron thick film of $TiD_{1.76}$ composition.

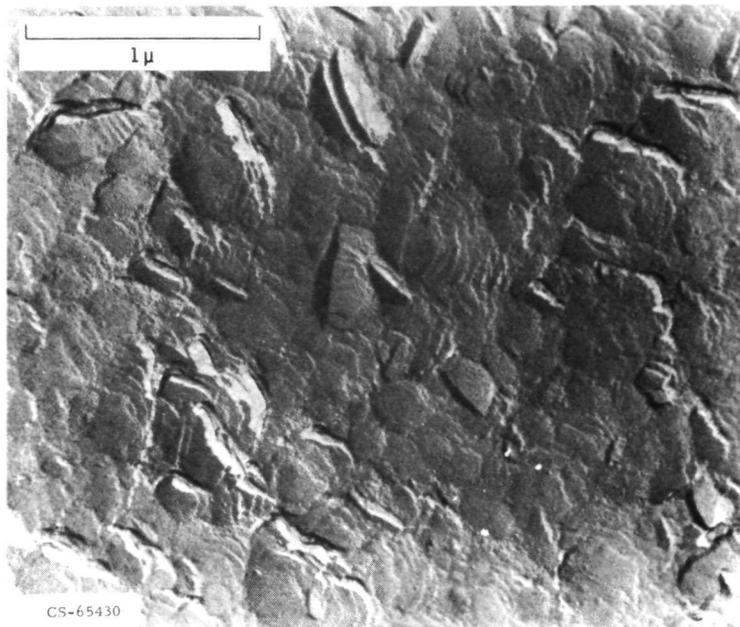


Figure 9. - A continuous 3 micron thick film of $TiD_{1.83}$ composition.