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Protective overcoatings on thin-film titanium targets for neutron generators

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ABSTRACT

We have developed a thin-film coating for neutron generator targets that can be loaded with deuterium or tritium at low temperatures (100 °C), and at gas/Ti ratios greater than 1.7. The key to this improvement is the addition of a thin palladium overcoat at the end of the titanium deposition. This overcoat prevents the oxidation of the titanium film, yet still allows loading to take place at low temperatures. A palladium overcoat of just 50–100 Å is sufficient to protect the titanium, while presenting a minimal energy loss to incident ions. We have just begun producing targets using this method, and see the possibility of substantial improvement in neutron generator efficiencies.

1. Introduction

Deuterium or tritium-loaded titanium films are frequently used in neutron generator targets [1]. One of the drawbacks of titanium is that it readily forms a tenacious surface oxide that prevents the absorption of D or T into the underlying film. The usual procedure for loading targets is to heat them to 400–500 °C in the loading gas for up to several hours, which disrupts the surface oxide layer and allows the gas to react with the bulk of the Ti film. Instead of relying on high-temperature processing to "activate" the titanium [2], we deposited a thin overcoat of palladium during the vacuum-coating process to protect the film from oxidation. Palladium was chosen for the overcoat due to its high diffusion constant for hydrogen isotopes. This allowed the films to be loaded to ratios greater than 1.7 at temperatures near 100 °C. This Pd overcoat should also protect the titanium from oxidation after loading. Unlike bare titanium targets that need to be protected from oxidation by storage in an inert atmosphere, we expect that Pd-coated targets can be stored in air without degradation.

Due to the novelty of the Pd overcoat, we did not know a priori the processing conditions that would be required to load the targets. Therefore, we designed a UHV chamber that could heat the substrates to >500 °C in pressures up to 100 Torr (13 kPa). This temperature also would allow the recovery of tritium from spent targets by heating them with the chamber evacuated.

We present here, the description of the processes used and results of fabricating and loading our first targets with deuterium.

2. Target design

We chose titanium as the target material as it has the capability of holding a higher density of hydrogen than the other commonly used metal-hydride target materials (Er, Zr, etc.) [3]. Titanium also retains the hydrogen at moderate temperatures, so target heating during operation of the neutron generator is less of a concern. The Ti film thickness of 5000 Å was selected to be somewhat less than the penetration depth of deuterium ions at the 100 kV accelerating potential we expected to use in our system. For a 5000 Å layer of $TiD_{1.7}$, SRIM calculations show that for a D^+ beam with initial energy of 100 keV, 90% of it will be deposited in the film.

Palladium was chosen for the overcoat for its high diffusion rate for hydrogen isotopes at moderate temperatures. Since it will only readily pass hydrogen, it is often used to purify hydrogen gas supplies [4]. For our application, the thickness of the overcoat is a compromise. The film has to be thick enough to protect the underlying titanium from oxidation, while thin enough to prevent excessive energy loss in the incident ions. Since the exact thickness where sputtered palladium will form a continuous, protective film over a titanium base layer was not known a priori, we tested several thicknesses. Based on SRIM calculations of a $\sim$1 keV loss for an incident 100 keV D^+ ion beam onto 50 Å Pd layer,we decided the energy loss would be acceptable for a 100 Å overcoat and deposited and tested 100, 75 and 50 Å overcoats. Due to the low concentration of deuterium remaining in the Pd film at the temperatures and pressures used in the neutron generator, few

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fusion reactions will occur in the thin overcoat. In comparison, a bare titanium film will form an oxide very rapidly when exposed to air, producing a "dead" layer on non-overcoated targets as well. Although the thickness of this native oxide layer can be around 80 Å, one study has measured dead layers from 500 Å to as much as 3000 Å on deuterated titanium films [5]. In this regime, a good approximation of energy loss is to scale linearly with thickness and density. Therefore, the energy loss should be approximately 0.8 keV for 80 Å of oxide, but as high as 30 keV for a 3000 Å thick dead layer. Since the fusion cross-sections increase rapidly with energy in the 100 keV range, a reduction in the energy loss in the thinner Pd layer has the potential to significantly improve the performance of thin-film targets when used with low to moderate ion energies.

3. Titanium/palladium deposition

The active films were vacuum deposited onto molybdenum disks using magnetron sputtering sources. Since the titanium will react with any residual air or water vapor in the chamber during the deposition process, we chose a small vacuum coating system with low base pressure ($<2\times10^{-7}$ Torr), and used process parameters that produced a reasonably high deposition rate ($\sim$2 Å/s). The system was also operated with a high flow of spectroscopy-grade argon across the substrate during coating (32 sccm), which further reduced the amount of residual gas able to react with the titanium film during coating.

Since opening the chamber to change source materials between the titanium deposition and the palladium overcoat would have allowed an oxide layer to form, we used a chamber with two sputtering sources. The chamber selected also has the hardware in place to rf sputter-etch, which we used before deposition to ensure good adhesion of the titanium layer to the molybdenum substrate.

The thickness of the Ti films were determined after the coating by measuring a step on a piece of silicon wafer coated at the same time as the target, using a profilometer. The palladium films were too thin to be measured directly with any accuracy, so their thickness was inferred using the deposition rate calculated from test coatings deposited for longer times, and scaled to the short deposition time required for the overcoat. The first three target coatings were 1.4 in. in diameter and 4100 Å thick, and the fourth measured 4200 Å.

4. Hydrogen loading chamber design

In order to load a titanium target with hydrogen isotopes, we needed a small vacuum chamber that had the ability to provide heat to the target while under vacuum. Since we plan to load targets with tritium, its design needed to be consistent with safe handling practices. To that end, the chamber uses all metal seals (ConFlat and VCR fittings), and has no electrical feed-throughs, which eliminates the possibility of their failure and release of tritium. The chamber was designed to have a minimum internal volume, 9.2 cu. in. (150 cc), to reduce the tritium inventory during the loading process. This relaxed the lab safety requirements for its use. Fig. 1 shows a cross-section view of the bottom part of the chamber.

The external vacuum envelope is made from a stock, 4.5-in. dia. ConFlat half-nipple. The substrate stage is at the top of a reentrant tube, and is heated by a 200 W, flat-spiral heating element on the air side of the stage. The stage is 2 in. in diameter, matching the maximum planned target diameter. The 3-in. long, 304 L stainless steel tube and the annular gap around it provides thermal insulation for the stage. Due to concerns of carbide precipitation when heated near 500 °C, the stage was made from 321 stainless, which is stabilized against it by the addition of titanium to the alloy. The heating element is designed for 110 vac, and has a thermocouple attached to it for safety monitoring. There is a small hole drilled up from the bottom of the stage to within 60

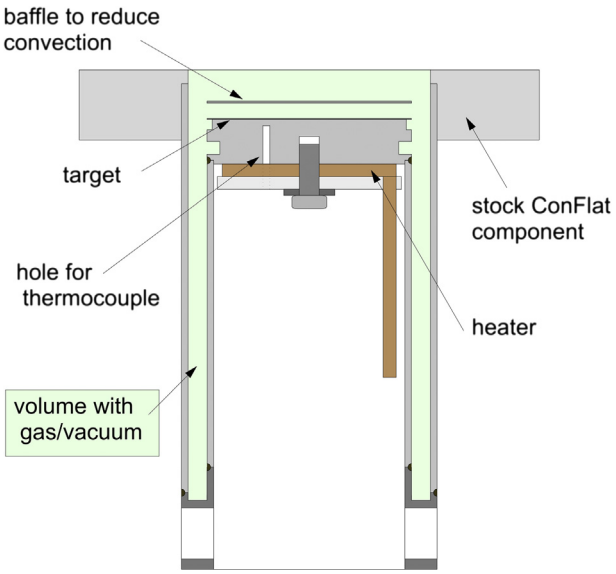

Fig. 1. Schematic of the hydriding chamber, showing several design features. It is designed to hold 2-in. dia. targets, with a minimum internal volume.

Fig. 2. Photo of the hydriding chamber during initial testing. At the tritium-loading facility, it will be connected to their control and data collection system, along with their mid-vacuum and tritium supply lines.