

# In-situ formation of solidified hydrogen thin-membrane targets using a pulse tube cryocooler

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## Introduction

Recent simulation studies [1] have suggested the presence of new ion acceleration mechanisms when a high-contrast laser pulse interacts with a nanometric target, known as the Laser Breakout Afterburner (BOA) mechanism [2]. The BOA regime manifests when the target becomes relativistically transparent such that in a thin target the electrons are still interacting with the laser field – the propagation of which ‘pushes’ the electrons driving a Buneman-instability imparting a kinetic energy transfer onto the ions at the rear surface of the target [3]. It has been shown via PIC simulations that BOA can theoretically generate ion beams on the order of hundreds of MeV. Such simulations show that the heaviest species present in the target governs the ion velocity of the interaction; thus a low-Z, high purity target is ideal and therefore there is a necessity to produce solidified hydrogen thin films. Simulation suggests that for the BOA regime to be realized in solid hydrogen, due to its low-Z, a target thickness of 1-4 $\mu\text{m}$  is ideal. This paper highlights the Central Laser Facility’s successes in producing in-situ solidified hydrogen thin-membrane targets.

## Target growth procedure

The method used for the growth of hydrogen targets was the condensation method (as opposed to the use of casting plates or extrusion) [4]. This was decided upon for several reasons: lower gas pressures are mandatory for use within the laser interaction chamber, quick target growth after system-cool down and higher spatial stability of the hydrogen target could be ensured. Hydrogen growth by condensation is achieved by injecting gaseous hydrogen into a sealed environment in which there is a cold surface. The surface is then cooled below the boiling point of  $\text{H}_2$  which causes condensed hydrogen liquid to fill a target substrate which is then further cooled below the triple point temperature (13.8K) to solidify the liquid into a thin  $\text{H}_2$ -ice layer.

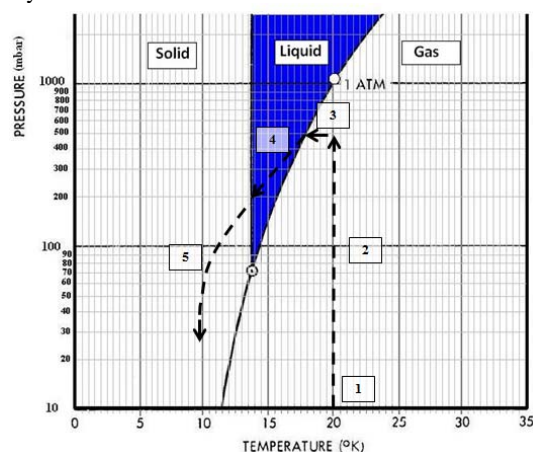


Figure 1. Phase diagram showing the growth procedure which was devised and followed for in-situ target production.

Referring to figure 1 there are five relevant stages within the followed growth procedure:

1. The initial state of the system. Regulating the temperature through a temperature controller at  $20\pm 0.3\text{K}$  (above the boiling point of hydrogen).
2. Hydrogen gas injection. The growth chamber is slowly filled with hydrogen until the pressure reaches  $\sim 500\pm 20$  mbar. This ensures there is a significant pressure to ensure the hydrogen could enter liquid phase as liquefaction causes an inherent pressure decrease.
3. Once there is an appreciable pressure inside the growth chamber the temperature is dropped and a steady inflow of gas is injected into the chamber to alleviate the pressure drop due to a fall in temperature. When the gas:liquid phase boundary is passed, droplets of hydrogen liquid can be observed flowing down the coldhead/target mount.
4. As liquefaction occurs there is a sudden pressure decrease within the chamber. Liquid hydrogen begins to flow/drip down the coldhead and over the target aperture; the colder the temperature falls, the lower the vapour pressure and the more viscous the liquid  $\text{H}_2$  becomes.
5. When the liquid:solid phase boundary is reached ( $14\pm 0.3\text{K}$  measured experimentally), dependent on the rate of cooling, one or more nucleation sites can be seen in the liquid. From such points a crystalline structure of solidified hydrogen begins to form eventually freezing into a thin layer over the aperture.

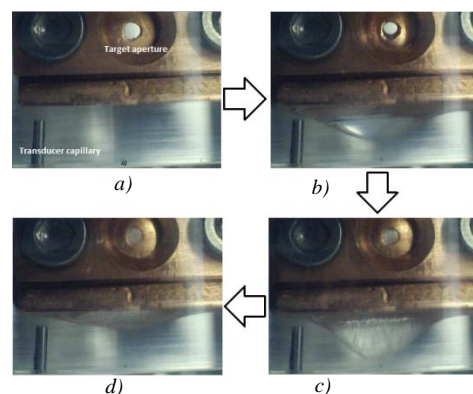
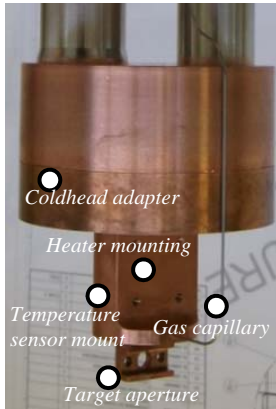


Figure 2. Time-lapse of the growth process. a) Initial state of system at 20K (stage 1). b) Liquid-phase (stage 4). Note the droplet which subtends from the bottom of the coldhead and the liquid ring over the target aperture. c) Freezing of the liquid film (stage 5). d) Solid  $\text{H}_2$  slowly sublimates due to low vacuum conditions within growth chamber. (Images taken with a USB-powered microscope).

## Initial system design

Successful hydrogen-ice growth was achieved by using a pulse tube cryocooler with a modified target mount which comprised

a copper coldhead beneath which a growth chamber was sealed, initially with an indium ring and bolt connection. (Consequently the chamber was fixed in place and only used as an early proof of principle and the seal was modified later in the project.) The growth chamber had a 1mm capillary which allowed the inletting of hydrogen gas which cooled within the environment and through careful temperature control could pass through the relevant phase boundaries to condense and solidify over an aperture within the coldhead. This gas was injected manually using a hand-valve system and was vented with a pump connected to the same capillary. The initial design is shown in figures 3 and 4.

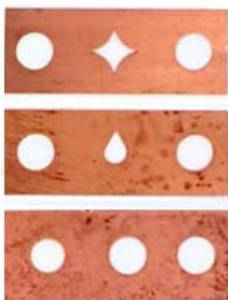


**Figure 3.** First design of cryocooler copper coldhead and target mount adapter showing the relevant components of the coldhead.



**Figure 4.** Lower section of the cold assembly with the growth chamber in place.

The target foils (figure 5) were necessary to promote condensation and growth of the target over an aperture in its centre. The foils were designed to be easily replaceable after the laser interaction with the hydrogen target which could cause damage to the foil and were also positioned horizontally to provide as much thermal contact to the coldhead as possible. Several designs were machined for future testing of the seeding of liquefied hydrogen over the target aperture, however, for the scope of this project a circular aperture was used.



**Figure 5.** Horizontally mounted copper target foils of various central aperture shapes; spiderweb (top), teardrop (centre) and circular (bottom).

The main difficulties realized with this particular system design were:

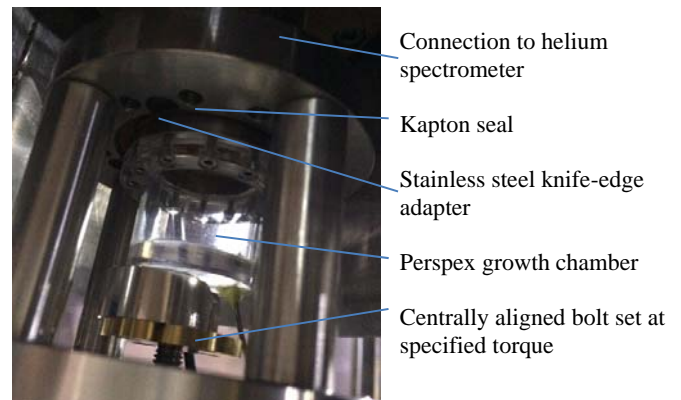
- *Seeding the liquid to form over the target aperture.* Due to the embedded geometry of the target foil – which was sandwiched within the target mount – flowing the liquid over the target centre was erratic and often would take multiple attempts. The design was revised along with several other modifications.
- *Fixed/bolted growth chamber.* The ultimate focus of the experiment was to research the laser interaction with the hydrogen target which required that the growth chamber was removable in-situ and not fixed in place to enable exposure of the target to the laser.

- *Inaccuracies and non-repeatability of gas injection.* Using a manual gas injection mechanism led to the observation that maintaining a specific pressure within the growth chamber was difficult. The fluctuations in pressure due to the high sampling rate of the transducer lead to an appreciable error in pressure readout from the display (with no data logging software installed at that time).

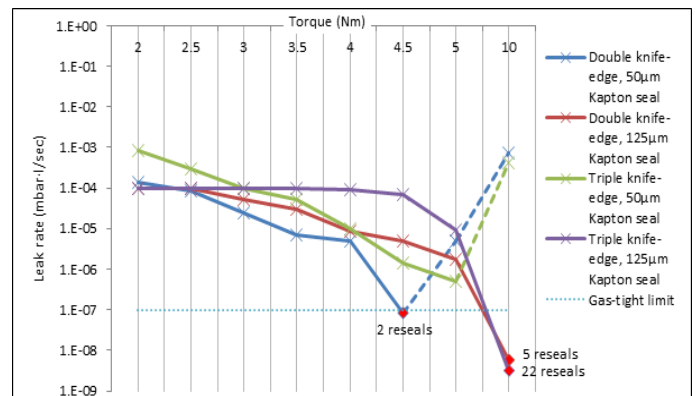
#### Design modifications:

In order to address the experimental issues with the initial system design several design modifications were made in order to realize a working cryogenic targetry apparatus suitable for fielding in a laser interaction chamber. The primary concern was to design and implement a dynamic sealing mechanism to enable the removal of the growth chamber from the coldhead, expose the target to the laser, and then reseal the growth chamber, all operated remotely from the control room of the laser facility.

To address the above issues, a novel sealing mechanism, as shown in figure 6, was designed and tested for efficacy. Two and three-bladed knife-edge adapters were machined to be attached to the rim of the growth chamber which would be forced into a polyimide (Kapton) film of sufficient thickness. The design was first tested for efficacy by forcing a centrally aligned bolt under differing torques on the underside of the growth chamber which pressed the blades against the Kapton. The two and three bladed adapters were each leak-tested, with a helium spectrometer, against two different thickness of Kapton film for varying torques as shown in figure 7.



**Figure 6.** Knife-edge and Kapton sealing leak-rate testing mechanism.



**Figure 7.** Leak rates of both a double vs. triple-edged knife adapter upon 50µm and 125µm Kapton film thicknesses when various torques were applied to a bolt forcing the knife-edges into the film. For successful gas-tight seals, the number of repeatable seals is shown.

From the results shown in figure 7 there were only three gas-tight configurations of the system. Of the three successful tests a triple-knife edge adapter under 10Nm of torque forced upon 125µm of Kapton provided the most leak tight seal of

$3.2 \times 10^{-9}$  mbar-l/sec and was repeatable 22 times before the Kapton film failed and needed replacing.

A motor driven system was then designed to satisfy the parameters above. It consisted of a remotely controlled driving mechanism delivering 10Nm torque and thus enabled a gas-tight seal in the growth chamber. The motor was attached on the underside of the system and was sufficiently insulated to avoid undesirable heat loading by using glass fibre rods to fix it in place with the coldhead. The growth chamber was also redesigned from stainless steel (optical windows were fitted) in order to withstand the force exerted by the motor.

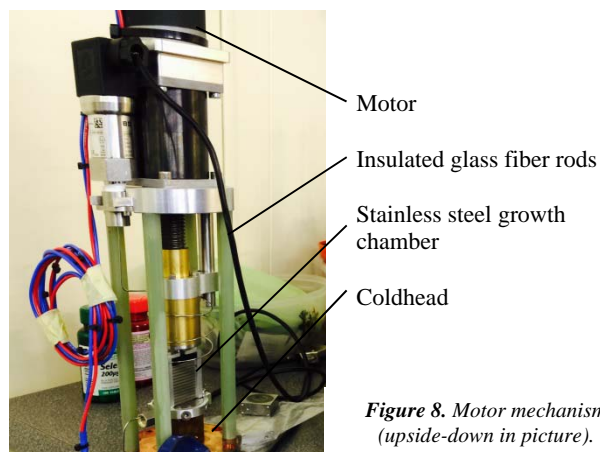


Figure 8. Motor mechanism (upside-down in picture).

Due to the inaccuracies and unrepeatability of the growth procedure through manual operation of the gas injection an automated gas flow control system was designed including LabVIEW control software, with the assistance of Alex Ortner (Technische Universität Darmstadt). This enabled all of the systems required for the growth process to be operated from a single UI and through integration of mass flow controllers allowed a constant, accurate pressure to be maintained which is crucial to keep the target in a stable environment.

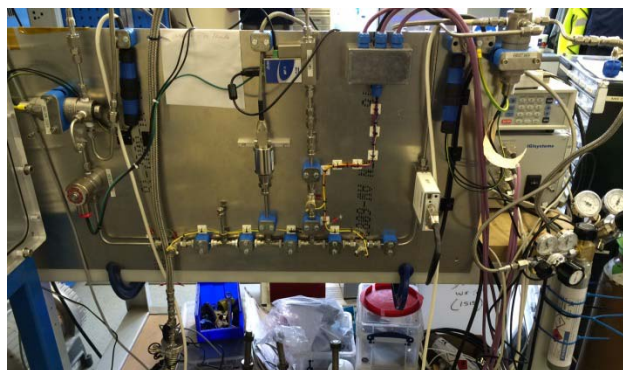


Figure 9. Gas-flow control system.

The thermal stability of the system was also improved by integrating a Lakeshore 336 temperature controller allowing four sensor inputs as well as 150W of heater power (up from 27W), which greatly decreased the warm-up time of the system and allowed more precise temperature regulation.

Due to the heat loading from the motor an improved two-stage radiation shield was designed and implemented onto the system. The two layers of the shielding comprised 2mm thick aluminium which attached to the upper 40K and lower 4.2K stages of the pulse tube respectively. By precooling the outer layer of the shield the thermal radiation impinging onto the inner, cooler shield was minimized which in turn minimized the overall radiation heat loading onto the target in the centre. Improvements to base temperature are shown in figure 10.

Shielding	Minimum Temperature (K)
No shielding	14K
One stage radiation shield and superinsulation blanket	8.4K
Two-stage radiation shield and superinsulation blanket	5.8K

Figure 10. Base temperature achievable by varying the shielding surrounding the coldhead.

Due to the volatility of hydrogen it was observed that when exposing the frozen target to vacuum the target rapidly thinned only remaining for approximately one minute before it completely sublimed. Figure 11 shows the temperature dependence on the thinning rate of hydrogen further demonstrating that as low a temperature as possible is crucial and is an ongoing subject of research for the project.

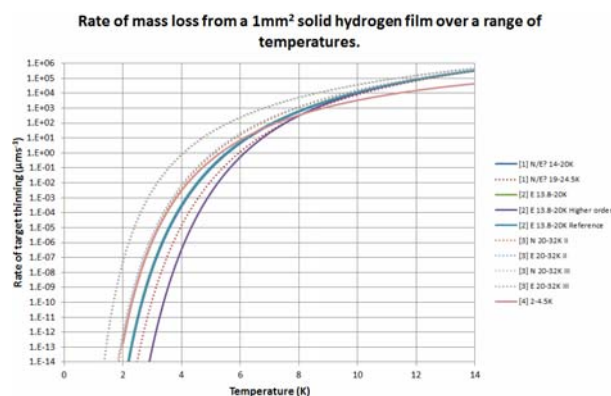


Figure 11. Thinning rate of solid hydrogen vs. temperature (credit: Patrick Gildersleve).

## Conclusions and future work

In July 2014, the system was successfully installed into Vulcan TAP and several laser shots on solidified hydrogen targets were taken which is the first time such a target has been shot in the target area. However, the rapid sublimation of the targets was an ongoing issue that is currently being researched in Vulcan's TAE facility. The cause of the sublimation is thought to arise from the escape of hydrogen gas after the target is exposed to the high vacuum, which then warms up and cryopumps back onto the target, which further heats the hydrogen and speeds up the sublimation process. A new research target chamber has been commissioned to investigate the issue through the use of two large turbo pumps and a smaller chamber volume in order to siphon the 'hot' gas as quickly as possible away from the target environment.

Within the constraints of the experiment it was difficult to assess the thickness of the target. A confocal microscope system is being researched on-site which will be installed to monitor the target thickness in real-time.

## References

- [1] Albright B J, Yin L, Bowers K J, Hegelich B M, Flippo K A, Kwan T J T, and Fernández J C, Phys. Plasmas **14** (2007), 094502-2
- [2] Yin L, Albright B J, Hegelich B M and Fernández J C, Laser and Particle Beams **24** (2006), pp 291-298.
- [3] Roth M, Bedacht S, Busold S, Deppert O, Schaumann G, Wagner F, Tebartz A ..... and Schollmeier M, *Breaking the 70MeV proton energy threshold in laser-proton acceleration and guiding beams to applications*. IPAC2014 conference 15-20 June 2014.
- [4] Tomlinson S, *Design for Production of Thin Film Solid Hydrogen Target*, CLF Annual Report 2011-2012.