Development of a higher power cooling system for lithium targets

Phoenix, B.; Green, Stuart; Scott, Malcolm; Bennett, J.R.J.; Edgecock, T.R.

DOI:

[10.1016/j.apradiso.2015.07.050](https://doi.org/10.1016/j.apradiso.2015.07.050)

License:

Creative Commons: Attribution-NonCommercial-NoDerivs (CC BY-NC-ND)

Document Version

Peer reviewed version

Citation for published version (Harvard):

Phoenix, B, Green, S, Scott, M, Bennett, JRJ & Edgecock, TR 2015, 'Development of a higher power cooling system for lithium targets', *Applied Radiation and Isotopes*. <https://doi.org/10.1016/j.apradiso.2015.07.050>

[Link to publication on Research at Birmingham portal](#)

Publisher Rights Statement:

Eligibility for repository: checked 07/10/2015

General rights

Unless a licence is specified above, all rights (including copyright and moral rights) in this document are retained by the authors and/or the copyright holders. The express permission of the copyright holder must be obtained for any use of this material other than for purposes permitted by law.

- Users may freely distribute the URL that is used to identify this publication.
- Users may download and/or print one copy of the publication from the University of Birmingham research portal for the purpose of private study or non-commercial research.
- User may use extracts from the document in line with the concept of 'fair dealing' under the Copyright, Designs and Patents Act 1988 (?)
- Users may not further distribute the material nor use it for the purposes of commercial gain.

Where a licence is displayed above, please note the terms and conditions of the licence govern your use of this document.

When citing, please reference the published version.

Take down policy

While the University of Birmingham exercises care and attention in making items available there are rare occasions when an item has been uploaded in error or has been deemed to be commercially or otherwise sensitive.

If you believe that this is the case for this document, please contact UBIRA@lists.bham.ac.uk providing details and we will remove access to the work immediately and investigate.

Development of a higher power cooling system for lithium targets

B. Phoenix, S. Green, M.C. Scott, J.R.J. Bennett,
T.R. Edgecock

PII: S0969-8043(15)30131-7
DOI: <http://dx.doi.org/10.1016/j.apradiso.2015.07.050>
Reference: ARI7079

To appear in: *Applied Radiation and Isotopes*

Received date: 7 February 2015
Revised date: 13 May 2015
Accepted date: 27 July 2015

Cite this article as: B. Phoenix, S. Green, M.C. Scott, J.R.J. Bennett and T.R. Edgecock, Development of a higher power cooling system for lithium targets *Applied Radiation and Isotopes* <http://dx.doi.org/10.1016/j.apradiso.2015.07.050>

This is a PDF file of an unedited manuscript that has been accepted for publication. As a service to our customers we are providing this early version of the manuscript. The manuscript will undergo copyediting, typesetting, and review of the resulting galley proof before it is published in its final citable form. Please note that during the production process errors may be discovered which could affect the content, and all legal disclaimers that apply to the journal pertain.

Development of a higher power cooling system for Lithium targets

Phoenix, B ^{a,*}, Green, S^a, Scott, MC^a, Bennett, J.R.J.^b, Edgecock, T.R.^c

^a*School of Physics and Astronomy, University of Birmingham, Edgbaston,
Birmingham, UK*

^b*STFC Rutherford Appleton Laboratory, Chilton, UK*

^c*University of Huddersfield, Queensgate, Huddersfield, UK*

1. Introduction

The Dynamitron accelerator at the University of Birmingham has been in service for more than 40 years. For the last decade it has been used primarily as a neutron source for research work related to Boron Neutron Capture Therapy (BNCT). The accelerator is routinely used at proton currents of 1mA at 2.8MeV, producing a neutron source intensity of 1.37×10^{12} n/s [6], although currents of up to 1.5mA have been achieved.

Historically clinical BNCT trials worldwide have relied on fission reactor based neutron sources [1] [2] [3] [4] [5]. However there are two factors which make them unattractive for widespread hospital use. One is their cost and relative inflexibility and the other is the public perception of the dangers of nuclear reactors. As a result, the use of accelerators for neutron production has been widely investigated, a summary of accelerator based BNCT worldwide has recently been published by Kreiner [7].

*Corresponding author

Email address: ben@bnct.co.uk (Edgecock, T.R.)

2. Target and Cooling System Design

Neutrons are produced at the Birmingham facility via the lithium-7 (p,n) reaction, with a thick natural lithium target and typically a 2.8MV accelerating voltage. The choice of the thickness of the lithium layer is a trade off between a number of factors including cooling performance, mechanical properties and gamma ray production.

In the target design described in this paper, all protons stop in the lithium layer. This has the significant advantage of avoiding concerns over blistering of the backing layer, which is a serious problem in some materials and can lead to lithium separating from the backing [8]. Experience of running these thick lithium targets has demonstrated that proton implantation in, and blistering of, the lithium itself has a negligible impact on target performance even over many months of running.

The Li(p,n) reaction has a threshold of 1.882 MeV. Protons below this energy are therefore not producing neutrons but can still produce gamma rays via inelastic scatter from the lithium nuclei. Targets thinner than the proton range reduce this additional undesirable dose to the patient by ensuring the protons only slow to the threshold energy in the lithium, doing the remaining slowing down in the heavy backing layer. Birmingham's design reduces this additional photon dose by incorporating a 20mm thick layer of antimony-free lead into the beam shaping assembly immediately surrounding the target [11]. This layer reduces the photon dose due to scattering in the lithium by 97%.

Lithium is a poor thermal conductor, with a thermal conductivity of $84.8 \text{ Wm}^{-1}\text{K}^{-1}$. It also has poor mechanical properties and degrades rapidly in contact with air or water. These properties make target construction more

of a challenge than with some other candidate materials. Birmingham's target consists of a 0.8mm thick layer of lithium metal on an oxygen free copper backing. Within the copper backing are thermocouple channels used to monitor the target temperatures; these are 1mm diameter holes spark etched into the 2mm thick copper backing which extend into the area underneath the lithium. In routine operation 0.5mm diameter, insulated, k-type thermocouples (RS part number 444-1247) are inserted in these channels and data is read using a National Instruments NI 9213 module. This temperature data is used both to aid in beam positioning and profiling, and as an input into the safety monitoring system, which controls the beam interlocks.

The mechanical and thermal bond between the copper and the lithium layer is critical for good target performance, and a method has been developed which forms a lithium-copper intermetallic layer with negligible thermal resistance [11]. To produce this, a mechanical pressing and heating regime is used, noting that the final target quality is very sensitive to the parameters in both of these processes. The target preparation methodology has been extremely successful at current Dynamitron powers despite the detailed composition of the lithium-copper intermediate layer not being known. It is possible that this layer is an alloy or solid solution with significantly different thermal properties to lithium. Experimental work investigating the physical properties of this layer in more detail is currently ongoing.

Cooling of the target is provided by a submerged jet of heavy water impinging directly on to the copper backing, as shown in figure 1. Simulations and associated experimental verifications which lead to this design have been previously described in the literature [11]. The cooling set-up has proved to

Figure 1: The existing submerged jet cooling system

be capable of maintaining a solid lithium target with heat loads of up to 4.2kW, equating to a heat flux of over 3 MW/m^2 . In order to have a clinical facility capable of irradiating patients in a reasonable time, upgrading the Birmingham system was considered necessary, with the aim of having a target capable of remaining solid up to at least 3mA and, ideally, 5mA.

The current system is extremely robust, requires minimal maintenance and has relatively low cost. A desire to preserve these characteristics has lead to an investigation of a number of relatively minor upgrades rather than

a radical redesign of the whole cooling system.

3. Cooling tests

3.1. Heating methodology

Representative tests of cooling system upgrades require a heat source capable of reproducing both the power density and the power input of the Dynamitron. Initial tests were made with a resistive heater; a gold plated silicon chip soldered directly to a copper target blank. It was found that temperature gradients across the heating element under load caused thermal stresses leading to rapid mechanical failure of the heating element.

Inductive heating was then tried using a large commercial induction heater capable of supplying a nominal 50kW (Minac 25 supplied by EFD). Initial attempts were made to couple the power directly into the copper target but this proved impractical as the heating unit was optimised for steel. A 5mm steel plate was then silver brazed to a copper boss, which was then vacuum brazed to a copper target blank (figure 2). Heat flow down the copper boss was calculated from several thermocouple readings down the axis of the bar, and heat variation across the target region was monitored by thermocouples in the same configuration as used with the normal, lithium, target. This arrangement comprises a central thermocouple surrounded by six equally evenly spaced thermocouples at a radial distance of 20mm. Tests showed that a maximum heat input of approximately 8kW was possible before the surface of the steel layer began to melt.

Several approaches to gas flame heating have also been employed. First a solid copper boss brazed to the back of the target was used, the boss being

Figure 2: Target blank prepared for induction heating

Figure 3: Gas flame heating arrangement.

heated with multiple oxyacetylene torches. However, the required power input could not be achieved. The most recent tests have been done using a small (2mm diameter) pinpoint gas flame and a jig which allowed repeatable positioning anywhere on the target surface (figure 3). This has proved very successful indeed allowing rapid and reproducible scans to be made of the variation of heat transfer coefficient across target surface.

3.2. Pumping Power

The cooling performance of the jet system is dependant on the jet velocity, its diameter and the distance of the input nozzle from the target. Pumping

Figure 4: Variation of target temperature with flow rate. Data for both heating with a copper boss and induction heater and with a gas flame

in the primary loop of the Birmingham system is provided by a centrifugal pump with all stainless steel internals. A recent upgrade (Grundfos model CRN3-17 to model CRN3-19) has increased the maximum flow rate from 20l/min to 40l/min. The dependence of average target temperature on flow rate is shown in figure 4.

The cooling loop currently installed uses standard 22mm diameter copper tubing with soldered joints to link the target to a 6kW chiller. However, any further increases in pumping power are likely to exceed the maximum working pressure of this system, and will require a redesign.

3.3. Binary Ice

Binary ice, also known as slurry ice, liquid ice, or pumpable ice, is a suspension of sub-millimetre scale ice crystals in a water/anti-freeze mix. It has been widely used in the fishery industry and as a cooling and thermal storage medium, so commercial production units are readily available. Also, in plate heat exchangers it has been shown to have an advantage in cooling performance over water [13]. Binary ice appeared to offer significantly increased heat removal properties over normal water so its use for our application has also been investigated.

Tests were carried out using the unmodified submerged jet geometry. Binary ice was provided by a Cooltech GmbH system and tests were carried out at their workshops by the authors. A standard binary ice mix consisting of a nominal 15% ice (by mass), 10% ethanol and 75% water was used. Ice crystal size at time of production from this type of scraped ice generator is approximately 100 microns [14]. The ice mix was stored in a large buffer tank throughout the several days experiments so there was likely to be some variation in ice fraction and crystal size [9]. The cooling system was configured so that it could be switched between the closed loop ice system and mains water run to waste. The pumping system was the same in both cases and flow rate was monitored with an in-line impeller flow meter. Target heat input was provided by the EFD induction heater described earlier.

It is clear from the data presented in figure 5 that in this system there was no significant difference between binary ice and water. The enthalpy of fusion of ice is 6.01 kJ mol^{-1} , compared to the heat capacity of water of $75.3 \text{ J (K mol)}^{-1}$ but this large difference in effective thermal capacity is not reflected

Figure 5: Comparison of binary ice cooling with water at 20L/min pumping rate

in the heat transfer results. No monitoring of return flow temperature or ice fraction was in place. It is possible that melting ice reduced the return flow temperature.

It is thought that this lack of improvement can be explained by the short residence time of the ice crystals in contact with the target plate. Simple calculations show that for a flow rate of 20L/min this is of the order of 1 millisecond, too short for any significant melting to take place. Results in the literature for a submerged jet system with a different phase change coolant (encapsulated paraffin wax) support this conclusion, as it was found that the cooling performance due to melting was strongly flow rate dependant [12]. An alternative explanation may be a layer of water which prevents the binary ice melting in contact with the surface.

4. Conclusions

Increasing the maximum pumping rate in our existing Dynamitron target cooling system reduces target temperatures for a given power by a factor of approximately 1.4. This is thought to be the maximum practical pumping power without significant system redesign.

Tests of a binary ice, phase change, coolant did not show a performance difference from water in the submerged jet system. The reason for this was not clear. Experimental work with binary ice is ongoing in order to better understand the system and to potentially design a cooling system which better exploits its properties. It is likely that investigations of binary ice cooling in the near future will have to remain experimental. The flow and thermal characteristics of binary ice mixtures are not modelled accurately by

existing CFD packages, so cooling performance is hard to predict. The vast majority of existing work is with ice slurries at much lower flow rates and pressures than are likely to be needed in accelerator applications. Technical challenges may therefore exist, both with pumping and mechanical properties of targets, which have yet to be discovered.

Extrapolation of the target temperature curve in figure 5 suggests that the current cooling system could potentially handle 3mA of beam current if the heat load was evenly distributed over the entire target surface. The existing beam steering system rasters the proton beam spot over a relatively small proportion of the target surface area. Future work will look at better matching this distribution of power delivery to the cooling profile, and increasing the total surface area of the target.

5. Acknowledgments

The authors would like to thank all at Cooltech, and particularly the late Joachim Paul, for all their help and advice and for making their facilities available for our use.

6. References

- [1] Busse, Paul M., et al. "A critical examination of the results from the Harvard-MIT NCT program phase I clinical trial of neutron capture therapy for intracranial disease." *Journal of neuro-oncology* 62.1-2 (2003): 111-121.
- [2] Chadha, M. D., et al. "Boron neutron-capture therapy (BNCT) for glioblastoma multiforme (GBM) using the epithermal neutron beam at

- the Brookhaven National Laboratory.” *International Journal of Radiation Oncology* Biology* Physics* 40.4 (1998): 829-834.
- [3] Sauerwein, Wolfgang, et al. “Organisation and management of the first clinical trial of BNCT in Europe (EORTC Protocol 11961).” *Strahlentherapie und Onkologie* 175.2 (1999): 108-111.
- [4] Yamamoto, T., et al. “The status of Tsukuba BNCT trial: BPA-based boron neutron capture therapy combined with X-ray irradiation.” *Applied Radiation and Isotopes* 69.12 (2011): 1817-1818.
- [5] Hatanaka, Hiroshi, and Yoshinobu Nakagawa. “Clinical results of long-surviving brain tumor patients who underwent boron neutron capture therapy.” *International Journal of Radiation Oncology* Biology* Physics* 28.5 (1994): 1061-1066.
- [6] Campbell, J., and M. C. Scott. “Absolute neutron yield measurements for protons on Li, Cu, Co and Be from threshold to 3 MeV.” *Proceedings of the fourth conference on the scientific and industrial applications of small accelerators*. 1976.
- [7] Kreiner, Andres Juan, et al. “Present status of Accelerator-Based BNCT.” *Reports of Practical Oncology & Radiotherapy* (2014).
- [8] Astrelin, V. T., et al. “Blistering of the selected materials irradiated by intense 200keV proton beam.” *Journal of Nuclear Materials* 396.1 (2010): 43-48.
- [9] Pronk, P., et al. “Time-dependent behavior of different ice slurries during storage.” *International Journal of Refrigeration* 28.1 (2005): 27-36.

- [10] Savidou, A., et al. "Proton induced thick target γ -ray yields of light nuclei at the energy region 1.0–4.1 MeV." *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* 152.1 (1999): 12-18.
- [11] Brown, Adam V., and Malcolm C. Scott. "Development of a high-power neutron-producing lithium target for boron neutron capture therapy." *International Symposium on Optical Science and Technology. International Society for Optics and Photonics*, 2000.
- [12] Wu, W., et al. "Jet impingement and spray cooling using slurry of nanoencapsulated phase change materials." *International Journal of Heat and Mass Transfer* 54.13 (2011): 2715-2723.
- [13] Bellas, J., I. Chaer, and S. A. Tassou. "Heat transfer and pressure drop of ice slurries in plate heat exchangers." *Applied Thermal Engineering* 22.7 (2002): 721-732.
- [14] Wang, M. J., and N. Kusumoto. "Ice slurry based thermal storage in multifunctional buildings." *Heat and Mass Transfer* 37.6 (2001): 597-604.