COMPACT ACCELERATOR BASED NEUTRON SOURCE FOR TECHNETIUM-99M PRODUCTION*

R.Seviour†, University of Huddersfield, Huddersfield, UK
H. Owen[1], University of Manchester, Manchester, UK
I. Bailey[1], University of Lancaster, Lancaster, UK
[1]also the Cockcroft Institute, Warrington, UK

Abstract

The radioisotope Technetium-99m (^{99m}Tc) is used in 85% of all nuclear medicine procedures. ^{99m}Tc is produced from its precursor Molybdenum-99 (^{99}Mo), whose production is nearly all from one of only five ageing research reactors. Recently a number of accelerator-based methods have been proposed to fill this gap and to diversify this supply chain. In this paper we present a compact (4 m) 10 mA, 3.5 MeV accelerator design, to generate ^{99}Mo via low-energy neutron bombardment of ^{98}Mo. We consider a Li(p,n) target for neutron production, and propose the use of a novel moderator to optimally shift the target output neutron spectrum into the epithermal region of the ^{98}Mo. This paper specifically focuses on numerical studies for an optimised target design capable of handling the thermal load.

INTRODUCTION

Nuclear medicine is the one of the principle hospital imaging techniques in medicine (after X-ray Computed Tomography and ahead of Magnetic Resonance Imaging), and 85% of the 36 million annual worldwide procedures utilise ^{99m}Tc; a typical ^{99m}Tc procedure might be cardiac imaging to assess risk following a heart attack. The European market is around 9 million procedures annually, where the cost of the ^{99m}Tc in each procedure is around £10. The short 6-hour half-life of ^{99m}Tc is diagnostically convenient, and its metastable nature means that it only emits 143 keV gamma rays upon decay, limiting the ancillary dose to patients (unlike other radionuclides). Not found in nature, ^{99m}Tc is derived from the precursor ^{99}Mo, which is presently mostly derived commercially from fission products from five ageing research reactors [1].

The primary reactor production chain for ^{99}Mo involves the irradiation of highly-enriched uranium (HEU) targets, chemical separation of the ^{99}Mo from the fission products, and packaging in a ^{99}Mo/^{99m}Tc generator. The resulting ^{99}Mo has a large specific activity typically exceeding 3000 Ci/g. However, since 2008 there has been a worsening supply problem due to ageing and failure these reactors, particularly the Canadian NRU reactor which presently provides half of global production [1, 2]. In 2010 it was reported that 65% of global production was facing shutdown. Whilst many of the shutdowns are carefully planned to avoid overlaps, there have been unscheduled events. There are ongoing efforts to adapt other reactors to produce ^{99}Mo but there is scientific and commercial consensus that this fragile end of the supply chain should be diversified by also developing accelerator-based production (see e.g. [3, 4]).

ALTERNATIVE PRODUCTION METHODS

Until around 2009 the most prominent proposed accelerator scheme was to use photofission of a ^{235}U or ^{238}U-containing target using bremsstrahlung photons from a typically 100 mA, 50-100 MeV electron linac/tungsten target, followed by hot-cell ^{99}Mo extraction similar to that for reactor Mo [5, 6]; depleted or LEU targets better meet IAEA requirements but produce roughly twice the fission waste of HEU, but in both cases the specific activity is similar to that for reactor ^{99}Mo. The large electron current offsets the low bremsstrahlung/photofission cross-sections, but the target is consequently difficult to cool. The principle advantage of the photofission method is that the extraction and generator production are largely the same as for reactor ^{99}Mo. The photofission route may not be cost-effective, and several alternative methods have been proposed [3, 7–12]. Direct cyclotron production of ^{99m}Tc in enriched ^{100}Mo targets using the ^{100}Mo(p, 2n)^{99m}Tc reaction has been proposed (e.g. by TRIUMF and Uni. of Alberta) using c.17-24 MeV protons [4, 13, 14]; licensing of the resultant ^{99m}Tc is progressing [4, 15–18]. The direct route means that the ^{99m}Tc must be made close to its point of use, i.e. regional rather than transnational.

An alternative route is to utilise neutron capture via ^{98}Mo$(n,\gamma)^{99}$Mo in either natural Mo or enriched ^{98}Mo. Initially fast neutrons from a suitable proton conversion target are moderated to epithermal energies to overlap the resonance peaks in the ^{98}Mo capture cross-section [19, 20]; spallation targets have been proposed at high energy that give many neutrons per proton (as advocated by Buono et al. [21]) but these require large (300 m) and expensive accelerator infrastructure. Low-energy targets may also be used. AAA presently produce small quantities of commercial radioisotopes (not yet ^{98}Mo) using a c.30 MeV cyclotron with a ^{9}Be(p,n)^{9}B target [20, 22]. Owen has examined the use of an existing high current (1-10 mA), 3 MeV proton source with a ^{7}Li(p,n)^{7}Be target; in both Be and Li targets the resonance overlap is limited by $1/r^2$ effects in the moderator assembly (so a small target assembly is required), but a graphite reflector can significantly boost the overall neutron capture rate [23]. The same targets might also utilise a subcritical uranium neutron multiplier [7, 24], but these

* Work supported by the UK Science and Technology Facilities Council under the grant ST/I002247/1
† r.seviour@hud.ac.uk

targets are likely more difficult to obtain regulation for. The difficulty in isotopically separating the ^{99}Mo from the target is well-known, but may not be needed if the capture rate is large enough; more likely, one may extract ^{99m}Tc directly from the targets similar to the cyclotron route.

PROPOSED SYSTEM

Here we consider the well-known ^{7}Li(p,n)^{7}Be reaction from protons with 2.5 MeV incident energy; at this energy several accelerator technologies may be used to deliver currents up to 10 mA (electrostatic or RFQ-based) in a compact (c.4m) footprint. The low energy and small target allows the use of a compact neutron moderator assembly (NMA) to shift the fast neutron energies into the epithermal (resonant) region for the ^{98}Mo(n,γ)^{99}Mo reaction. Studies show that ^{98}Mo secondary targets placed in a suitable location within the NMA can give diagnostically-useful specific activities for realistic currents and irradiation times. By comparison, a ^{9}Be(p,n)^{9}B target study (using MCNPX and GEANT4) predicts that a 10 mA extracted proton current at 30 MeV gives specific activities of 230 Ci/g/hr [23]. However, 10 mA currents at 30 MeV will be very difficult to achieve from a cyclotron. We propose a high current proton source, a low energy beam transport (LEBT) and RFQ (Figure 1).

Figure 1: Schematic overview of our proposed accelerator isotope system.

Accelerators based on RFQs offer many advantages over other technologies; RFQs can in principle transport intense beams up to 100 mA with high beam transmission . This gives RFQs a flexibility to operate over a very wide parameter space; RFQs have demonstrated the capability of accelerating protons up to 8 MeV [25]. The parameters of our proposed RFQ are: input energy 60 keV, output energy 4 MeV, current 10 mA, duty factor 6 – 100%, RF frequency 352.2 MHz, maximum Es 33MV/m, beam power 170 kW, structure power 420 kW, total power 590 kW, total length 4.75 m.

The ^{7}Li(p,n) reaction is well understood with extensive experimental data and well-developed, verified, numerical models [26, 27]; protons at 2.5MeV on ^{7}Li(p,n) have a c.300 mb cross-section, producing 5.5$x10^{-8}$ neutrons per incident proton. The fast neutron spectrum from a Li target allows the use of a compact Neutron Moderator Assembly (NMA) to shift the fast neutron energies into the epithermal resonant part of the ^{98}Mo neutron capture cross-section. The NMA we propose is based upon the design of Brett [23] adapted from a NMA used for BNCT [28]. This design consists of three main components: (1) primary target (proton to neutron production); (2) Moderator (Neutron moderation from the primary to secondary target; (3) Secondary target (Neutron capture target). An overview of our NMA is shown in figure 2. The primary target is surrounded by a layered lead/Fluental moderator (Al+AlF3+Li) into which irradiation slugs of ^{98}Mo may be inserted at a radius optimising the overlap neutron spectrum with the ^{98}Mo capture cross section. The NMA is surrounded by a Graphite reflector and Li-loaded polyethylene shield to maximise the neutron flux at the secondary targets.

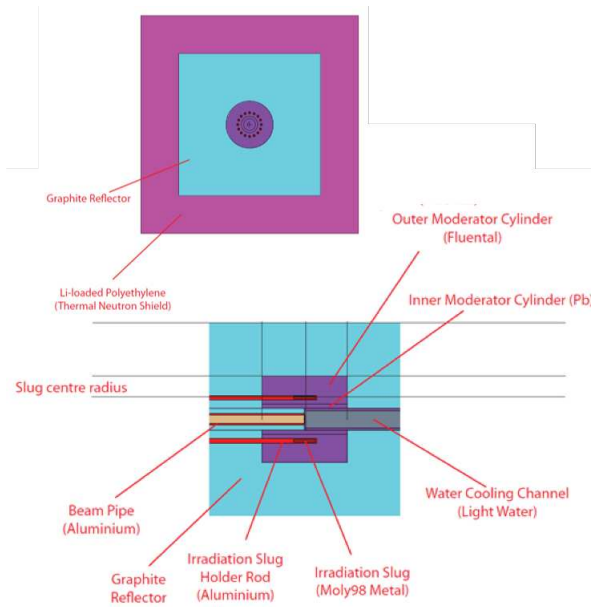

Figure 2: Schematic overview of the primary target, neutron moderation assembly and secondary target slug locations; the entire assembly is a cube approximately 60 cm on a side.

Numerical simulations using MCNP5 and Geant4 indicate c.0.2 Ci/g/hr/mA of 99Mo activity may be obtained at the secondary targets for a given proton current at the primary target. With sufficient cooling it is thought that a solid Li target may support currents up to 10 mA (i.e. 30 kW beam power) [29], with around 3 kW demonstrated so far. Our concept primary target design is shown schematically in figure 3 and consists of 20cm radius 1mm thick Li target covered with a 60μm Al layer, all on a Cu back plate 3mm thick. The stopping power of the Al film cover was modelled using SRIM [30], showing that a 3 MeV proton beam passing though the 60μm Al layer produce 2.5 MeV incident protons At the Li target surface.

To determine the effects of heating and cooling in the primary target the commercial simulation package Comsol was used to determine effective heat transfer properties calculated from the respective properties of the solid and fluid materials that make up the primary target structure. This model also models the effect of the interfaces between the materials in the model to simulate the contact thermal conductance coefficients, dependent on the applied stress, on the specific conductivity in the gap, and also by accounting

ISBN 978-3-95450-147-2

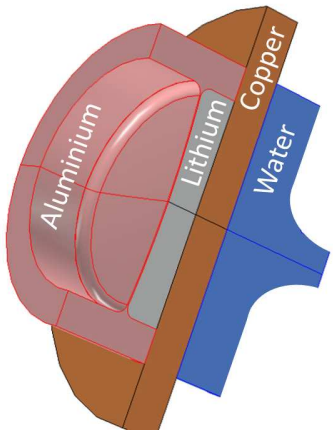

Figure 3: Schematic overview of our proposed target.

for the surface-to-surface radiation contribution between surfaces separated by small gaps. In our model the primary target is actively water cooled with a high water flow rate; in Comsol the thermal effects of fluid flow are taken into account at interfaces between laminar flow and turbulent flow by using high/low-Reynolds $\kappa - \epsilon$ turbulent models, where natural buoyancy effects due to temperature differences are accounted for by assuming a non-isothermal flow.

Simulations were conducted assuming a steady state heat load of 30kW from the proton beam on to the Al film of the primary target. The results of the optimised target simulation are shown in figure 4, where it was found that the maximum temperature in the Li target was below 170 deg C, compatible with the 180 deg C melting point of the lithium target. This was achieved with the target actively cooled by injecting water at the base of the target system, at 0.1 Bar, 15 deg C at 0.3 m/s.

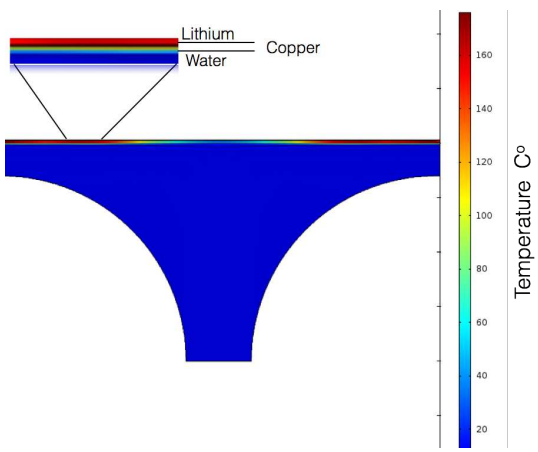

Figure 4: Expected temperature distribution of the primary target at 30 kW beam power load.

CONCLUSION

Using the proposed ^{7}Li(p,n) reaction allows lower-energy proton beams to be used compared to conventional accelerator approaches for technetium production, but with a higher possible beam current, enabling this scheme to achieve a similar specific activity to the 30MeV cyclotron ^{9}Be(p,n)^{9}B target discussed above. The combination of this target with our novel moderator assembly results in potentially clinically-useful production rates of 99mTc, although measurements of the specific activity and extracted purity would need to be undertaken. Although physically larger than the equivalent cyclotron target, the proposed target assembly is not unduly large, and could be used at laboratories with electrostatic proton sources but no cyclotron; for example co-existing with other uses of high-flux neutrons such as for materials characterisation or security. Hence this scheme can help to diversify supply of 99mTc to more locations than possible only with reactors or cyclotrons.

REFERENCES

[1] P. Gould, "Medical isotope shortage reaches crisis level", in *Nature*, vol. 460, p. 312, 2009.

[2] Report to the European Commission on the Supply Shortage of Radiopharmaceuticals, October 2008.

[3] H.L. Owen *et al.*, "Options for UK Technetium-99m production using accelerators", in *Proc. of IPAC'14*, Dresden, Germany, 2014.

[4] B. Neilly *et al.*, "Future supply of medical radioisotopes for the UK report 2014", arXiv:1501.03071, 2015.

[5] G Bennett, *et al.*, in *Nuclear Technology*, vol. 126, no.1, p. 102, 1999.

[6] "Making medical isotopes - report of the task force on alternatives for medical-isotope production", TRIUMF, Vancouver, Canada, pp. 1-94, 2008.

[7] 'Production technologies for molybdenum-99 and technetium-99m', IAEA-TECDOC-1065 (1999).

[8] K. Bertsche, "Accelerator production options for 99Mo", in *Proc. of IPAC'10*, Kyoto, Japan, paper MOPEA025, p. 121.

[9] M. de Jong, "Producing medical isotopes using X-rays", sin *Proc. of IPAC'12*, New Orleans, LA, 2012, paper THXA01, p. 3177.

[10] T.J. Mackie, *Nucl. Med.*, p. 53 and Suppl. 1, p. 1480.

[11] J.A. Osso Jr, *Curr. Radiopharm.* vol. 5, p. 178, 2012.

[12] G.E. Dale *et al.*, in *AIP Conf. Proc. 1525 (CAARI'12)*, p. 355, 2013.

[13] M.C. Lagunas-Solar *et al.*, in *Appl. Rad. Isotopes*, vol. 42, no. 7, p. 643, 991.

[14] T. Ruth, in *La Physique au Canada*, vol. 66, no.1, p. 16, 2010.

[15] K.M. Gagnon, "Cyclotron production of technetium-99m", PhD thesis, University of Alberta, 2012.

[16] X. Hou, A. Celler, J. Grimes, F. Benard, and T. Ruth, "Theoretical dosimetry estimations for radioisotopes produced by proton-induced reactions on natural and enriched molybdenum targets", *Phys. Med. Biol.*, vol. 57, p. 1499, 2012.

[17] O. Lebeda, E.J. van Lier, J. Stursa, J. Ralis, and A. Zyuzin, "Assessment of radionuclidic impurities in cyclotron produced 99mTc", in *Nucl. Med. Biol.*, vol. 39, p. 1286, 2012.

[18] T.J. Morley, *et al.*, "An automated module for the separation and purification of cyclotron-produced 99mTcO4", in *Nucl. Med. Biol.*, vol. 39, p. 551, 2012.

[19] C Rubbia., CERN, Geneva, Switzerland, Rep. CERN/LHC/97-04, 1997; US Patent App. 10/985,323, Jan 2004.

[20] P. Froment *et al.*, in *Nucl. Inst. Meth. A*, vol. 493, p. 165, 2002.

[21] S. Buono, "An accelerator-driven production of Mo-99 at CERN as a long-term solution to the current world crisis in diagnostic medical imaging", AAA White Paper, 2009.

[22] K. Abbas *et al.*, in *Nucl. Inst. Meth. A*, vol. 601, p. 223, 2009.

[23] D. Brett, H. Owen and M. Gill, "Resonant neutron capture for radioisotope production", internal report, Manchester, 2010.

[24] P.J. D'hondt *et al.*, in *Proc. SPIE*, vol. 2867, p. 505, 1997.

[25] J.W. Staples *et al.*, "Design of an RFQ-based neutron source for cargo container interrogation", in *Proc. of Linac'06*, paper TUP005, p. 253.

[26] H. Liskien and A. Paulsen, "Neutron production cross sections and energies for the reactions 7Li(p,n)7Be and 7Li(p,n)7Be", in *Atomic Data and Nuclear Data Tables*, vol. 15, p. 57, 1975.

[27] C. Lee and X. Zhou, "Thick target neutron yields for the 7Li(p,n)7Be reaction near threshold", in *Nucl Inst. Meth. B*, vol. 152, p. 1, 1999.

[28] C. Culbertson *et al.*, "In-phantom characterisation studies at the Birmingham Accelerator-Generated epIthermal Neutron Source (BAGINS) BNCT facility", *Appl. Rad. Isotopes*, vol. 61, p. 733738, 2004.

[29] R. Edgecock, "Accelerator-driven boron neutron capture therapy", in *Int. J. Mod. Phys. A*, vol. 29, no. 14, p. 1441004, 2014.

[30] J.P. Biersack and L. Haggmark, in *Nucl. Instr. and Meth.*, vol. 174, p. 257, 1980.

ISBN 978-3-95450-147-2