# RECENT ADVANCES IN THE DESIGN OF A CYCLOTRON-DRIVEN, INTENSE, SUBCRITICAL NEUTRON SOURCE

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# **1 INTRODUCTION**

Most of the nuclear reactors used today for radioisotope production (mainly <sup>99</sup>Mo for nuclear medicine), research and industrial applications, are due, in the next years, for a major refurbishment or for decommissioning. This problem has prompted a renewed interest on alternative neutron production methods.

In particular, the need for reliable sources of <sup>99</sup>Mo is becoming more and more urgent. The importance of the production of <sup>99</sup>Mo is related to the world consumption of <sup>99</sup>Mo/<sup>99m</sup>Tc generators. The availability of Tc generators is crucial: Technetium 99-m is, by a large extent, the most widely used radioisotope in nuclear medicine.

Most of the <sup>99</sup>Mo used in nuclear medicine is obtained as a fission product of <sup>235</sup>U, in a very small number of research reactors which are getting quite old. As an attractive, competitive alternative to nuclear reactors, the authors recently proposed [1] a subcritical, cyclotronbased spallation neutron source, with neutron multiplication by fission. The present paper presents some recent advances in the design of this system, together with the design corresponding to the optimal configuration for <sup>99</sup>Mo production.

### 2 ADONIS : AN ALTERNATIVE TO NUCLEAR REACTORS

The proposed ADONIS-system (Accelerator Driven Optimized Nuclear Irradiation System) is designed for the production of fission <sup>99</sup>Mo, among other applications. It is based on [1] and includes the following elements:

- a H<sup>-</sup> cyclotron, able to accelerate 2 mA of beam at 150 MeV with low acceleration losses and almost 100% extraction efficiency;
- 2) a beam transport system, transporting the proton beam without losses to a neutron source;
- 3) a neutron source including:
  - a) a primary beam target, where the proton beam strikes a molten Pb-Bi target, producing spallation (mostly evaporation) neutrons;
  - b) a water moderator surrounding the primary target;

c) a number of secondary targets made of highly enriched <sup>235</sup>U. The neutron multiplication obtained in such a system can be shaped by the amount of secondary targets, but will ultimately remain far from criticality.

If necessary, to allow for different applications with very different source designs, or for backup reasons for example, more than one neutron source can be connected to the same accelerator.

Due to the inherent limitation of the maximum amount of uranium put into this system, its behavior is non-critical. The concept of sub-criticality indicates the completely different nature of this system compared to reactors. The use of a cyclotron as a driver also allows the quasi-instantaneous shut-down of the system if necessary. The combination of sub-criticality, externally driven neutron source, and the design of the system itself makes ADONIS inherently safe.

#### 2.1 The cyclotron

The proposed 150 MeV, 2 mA, H cyclotron takes advantage of the experience of Ion Beam Applications which has built more than 15 lower energy (30 MeV), H<sup>-</sup> cyclotrons for radioisotope production. IBA also proposes today high intensity versions of these cyclotrons, with final extracted currents up to more than 2 mA. A key component of these versions is a higher brightness H<sup>-</sup> multicusp ion source developed for IBA by AEA Technology in Culham (GB) [2]. The proposed cyclotron will use such an improved multicusp ion source, able to produce 25 mA of H<sup>-</sup>.

Experimental results and calculations were presented previously [3], showing that the space charge limit for current designs of H<sup>-</sup> cyclotrons was between 5 and 10 mA of beam current. These results show also that very high beam loading of the cyclotron RF system - up to 80% - is possible. The results of another IBA accelerator, the Rhodotron [4] show that mainline to RF efficiencies in excess of 70% can be achieved at 200 kW RF power and 107 MHz.

The total power efficiency of such a 150 MeV, 2 mA  $H^-$  cyclotron could therefore reach 50%, i.e. a total electrical power of only 600 kW for 300 kW of beam power.

The problem of the electromagnetic dissociation of H<sup>-</sup> imposes the use of lower magnetic fields at higher energies. For a 150 MeV cyclotron, the maximum sector field would be 1.1 T, and the average field .6 T at the center. This would result in a pole radius of 2.75 m, and an external diameter of 8 m for the accelerator.

#### 2.2 The production of the primary neutrons

The 150 MeV, 300kW proton beam is used to produce spallation neutrons in a molten Pb-Bi target (see next paragraph).

A first estimation of the number of neutrons per incident proton was obtained through interpolations using available ([5], [6] and [7]).

This first estimation was confirmed by more detailed calculations of the neutron production from the spallation reaction and the neutron transport using the computer codes HETC and DORT. The calculations are described in detail elsewhere [8]. As a result of these calculations, the total primary neutron yield is estimated to be 0.8 neutrons per incident proton at 150 MeV.

#### 2.3 The neutron source

The proposed target assembly is schematically illustrated in fig. 1. A flowing liquid lead-bismuth eutectic alloy is currently proposed as the spallation target material. The flowing target allows the heat to be transported through convection of the target material itself. The proposed primary target is vertical with the liquid lead-bismuth flowing out of a ring-type nozzle into an open channel. Here the fluid interacts with the proton beam and is in direct contact with the vacuum. The flow exits the bottom of the target region, and is pumped through a heat exchanger, and then returns back to the target. A drain tank (not on fig. 1) is used to hold the solid and liquid alloy during start-up or shut-down: the liquid can therefore be pre-heated and cooled down in a controlled manner. Electro-magnetic pumps were chosen to deliver the forced circulation in the Pb-Bi circuit. The primary target will be surrounded in all directions by a water moderator, in order to thermalize the primary spallation and secondary fission neutrons. The <sup>235</sup>U secondary targets are planar and placed around the spallation source in a separate circuit. They are positioned in three concentric zones as illustrated in fig. 1. The optimal configuration may vary in function of the application and has been calculated with neutronic calculation codes for the case of the production of <sup>99</sup>Mo (see next paragraph).

All the above components are located in a main irradiation pool, for moderation and shielding purposes. Adjacent to the main pool is an annex pool (see fig. 1) allowing the remote handling of new and irradiated targets and used also as an intermediate storage pool. The manipulation and transfer of the uranium targets is realized by using a combined system of teleoperation and guiding tubes.



Fig. 1. Schematics of the sub-critical facility

Among the advantages of the proposed assembly, let us note the absence of an entrance window for the proton beam, the uncoupling of the primary spallation target circuit and the uranium target circuit and surrounding feeding zone, and the simple one-circuit operation for Pb-Bi serving both heat-transport needs and spallation target needs.

# 3 OPTIMIZATION OF THE SYSTEM FOR <sup>99</sup>Mo PRODUCTION

In view of this important application, the design of such a sub-critical facility was optimized for <sup>99</sup>Mo production. To obtain the maximum fission rate in the  $^{235}$ U targets - and therefore the maximum multiplication factor, these targets have to be placed in a position where the thermal flux resulting from the slowing-down of the energetic spallation neutrons is maximum. Another parameter of importance is the amount of uranium targets that can be placed in that position, in order to maximize the total <sup>99</sup>Mo production rate, but limiting the shadowing effects between them. In fact, the proposed ADONIS system results into a high operational flexibility. One can load independently different amounts of <sup>235</sup>U targets in the three irradiation zones, such that one can maximize either the total <sup>99</sup>Mo production or the specific activities. Any production scheme in between is of course possible.

The neutron transport, thermalisation and multiplication in this assembly were simulated using

numerical neutron transport codes at SCK-CEN. In particular, the DORT  $S_N$  [9] neutron transport code was used, associated to the MOL-BR2-40GR library, to calculate the multiplication factor of the ADONIS system as well as the thermal flux distribution in the water pool surrounding the spallation source, in the presence of the secondary targets or not. These calculations were completed in the fixed source mode with multiplication allowed in the fissile zones. A cylindrical geometry modeling was used to describe the Pb-Bi spallation source, the structural external double wall (Stainless Steel), the water-pool and the three rings of <sup>235</sup>U targets. By using different loading schemes, one can obtain an extended set of attainable flux distributions. One of them is shown in fig.2. The presence of two-side wings is due to the effect of the thermalisation and the reflection of fast neutrons created during the fission process in the <sup>235</sup>U plates. Typical thermal neutron fluxes at targets location are around  $6.10^{13}$  n/s.cm<sup>2</sup>.



Fig. 2. Neutron flux distribution

Table 1 presents the activity of <sup>99</sup>Mo which can be achieved for two different loading schemes of the uranium targets.

 Table 1:
 Example of Loading Schemes for <sup>99</sup>Mo

 Production (activities end of irradiation, per week)

рег week)		
Type of Loading	Total Activity (Ci)	Specific Activity (Ci/gr)
Maximum total production	35000	100
Maximum specific activity	22500	180

The maximum total production corresponds, at the end, to more than 50 % of the world's demand, while the obtained mean specific activities are comparable to those presently obtained in most nuclear reactors used for  $^{99}$ Mo production.

#### **4 CONCLUSION**

Compared to nuclear reactors, the ADONIS system is unquestionably safe. Other advantages are the modularity of the system and its flexibility in operation. As far as the production of <sup>99</sup>Mo is concerned, one such system can supply more than 50 % of the world demand. Investment, operational costs, including personnel, operational fuel and waste, and final dismantling costs, would be significantly lower also. The existing fissionmolybdenum processing technologies could be used further on without change.

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