Progress in NUCLEAR SCIENCE and TECHNOLOGY, Vol. 2, pp.284-288 (2011)

ARTICLE

Design of Accelerator-Based Solutions to Produce ^{99}Mo Using Lowly-Enriched Uranium

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IBA Company investigates the feasibility of using charged particle accelerators to irradiate lowly-enriched Uranium targets in order to produce ^{99m}Tc/^{99}Mo generators. A 350 MeV proton accelerator coupled to a sub-critical reactor, with or without spallation target, can produce half of the ^{99}Mo worldwide demand. An alternative solution based upon a 24 MeV electron beam to produce a third of the world demand is also being pursued.

KEYWORDS: nucleon-induced fission, photofission, Uranium, ^{99m}Tc/^{99}Mo generator, accelerator-driven system, MCNPX

I. Introduction

The ^{99m}Tc radionuclide is today the most frequently used radioisotope in nuclear medicine for single photon emission computed tomography (SPECT). It is obtained by the decay of the 66 hours half-life radionuclide ^{99}Mo. This isotope is obtained with a high-specific activity by a complex production chain relying on the irradiation of highly-enriched Uranium (HEU) targets in nuclear research reactors. The recent problems encountered by the HFR (Petten) and NRU (Chalk River) research reactors lead in 2009 to a serious crisis in the production of ^{99m}Tc/^{99}Mo generators.

Ion Beam Applications (IBA) Company already started in 1995 to think about new ways to produce ^{99}Mo based upon accelerators instead of nuclear reactors. At that time, a collaboration between IBA and the Belgian center for nuclear studies (SCK-CEN) led to the ADONIS (Accelerator Driven Optimized Nuclear Irradiation System) concept based upon a 150 MeV, 2 mA H^- cyclotron.[1] With the revival of the ^{99}Mo shortage, this concept was recently revisited and new neutronic calculations were performed by IBA and SCK-CEN.

ADONIS uses a spallation target to produce a high-flux of neutrons and to generate neutron-induced Uranium fissions. In consequence, that system requires the use of HEU targets. A closer look to the cross sections for Uranium fission induced by high-energy protons and neutrons indicates that ^{235}U and ^{238}U exhibit very similar fission cross sections for nucleons above 10 MeV. We then considered the possibility to shoot a proton beam directly on lowly-enriched Uranium (LEU) targets with a ^{235}U fraction below 20% to benefit from both the proton-induced and the neutron-induced fissions.

Beside proton accelerators used for radioisotope production and proton therapy, IBA is also active in the field of electron accelerators used for industrial applications. It has developed the Rhodotron TT1000 used to produce high-energy X-rays.[2] Able to deliver 5 to 7 MeV beams with an intensity of 100 mA, it is the most powerful electron accelerator used for medical devices sterilization.[3] Based upon its experience, IBA agreed to collaborate with the Advanced Medical Isotope Corporation (AMIC) in the US to develop a new approach for ^{99}Mo production. This new system uses an electron beam impinging on a high-Z conversion target to produce a large flux of high-energy X-rays. These X-rays are then used to irradiate a vessel containing LEU salts diluted into a heavy water (D_2O) solution. A large thermal neutron flux is obtained thanks to the neutrons produced by photonuclear reactions on Deuterium and those generated by Uranium fissions.

Studies performed on the ADONIS solution are described in Section II. The performance figures of the system based upon proton-induced fission are presented in Section III. Finally, the electron accelerator-based system is considered in Section IV.

II. The ADONIS Solution

ADONIS relies on a high-energy high-current cyclotron coupled to a subcritical assembly.[1] The proton beam impinges on a conical spallation target made in Tantalum to generate a high-intensity neutron flux. The Ta target is surrounded by 4 cylindrical layers of HEU targets interleaved with Beryllium moderation rings. The targets have a cylindrical shape with an outer radius of 1.1 cm and a length of 20 cm. They are identical to those used in production today in nuclear reactors and contain 4 g of ^{235}U each. They are immersed in heavy water (D_2O) for cooling. The subcritical core contains a total of 150 HEU targets and is surrounded by a thick Be reflector. The ADONIS core is modeled with MCNPX 2.5.0 and is shown in **Fig. 1**.[4] MCNPX simulations are based upon LA150 nuclear data evaluations for proton and neutron transport in water.[5] The nuclear data libraries ENDF/B.VI.0 and ENDF/B.VI.2 are used for neutron transport in Be and ^{235,238}U, respectively. Thermal $S(\alpha,\beta)$ tables

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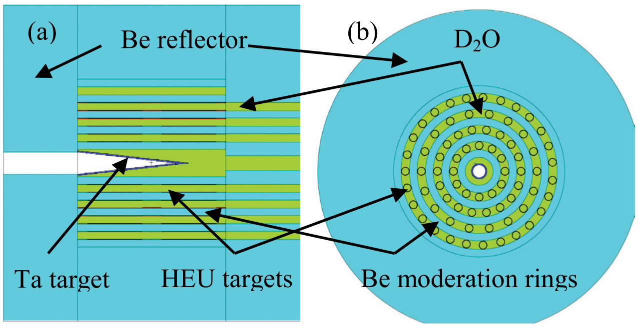

Fig. 1 Schematic view of the ADONIS central core as modeled in MCNPX: (a) Side view; (b) front view

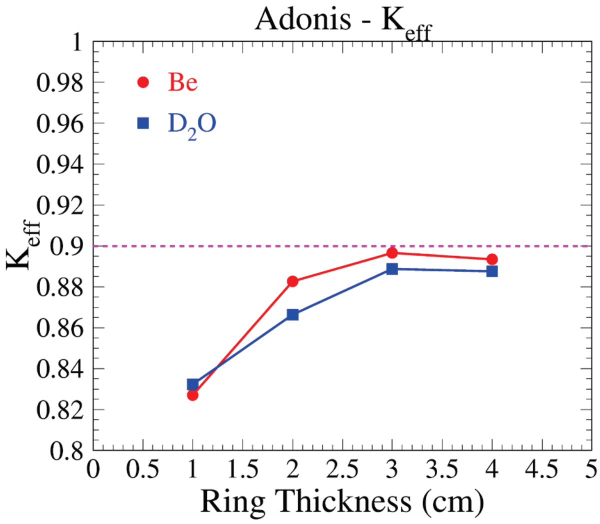

Fig. 2 Evolution of k_{eff} factor as a function of moderation ring thickness using either Be or D_2O

are used to describe the thermal neutron scattering in Be and heavy water.

Criticality calculations indicate that the k_{eff} value of the target assembly varies strongly with the thickness of Be moderation inner rings. As shown in **Fig. 2**, similar k_{eff} values can be obtained using either Be or D_2O. The thickness of the external Be reflector is also an important parameter to optimize the thermal neutron amplification factor and a thickness of 20 cm was chosen. However, for a total of 150 HEU targets, the k_{eff} value never exceeds 0.9 and the system remains thus far from criticality.

The evolution of ^{99}Mo specific activity as a function of irradiation time is presented in **Fig. 3** for beam energies of 150 MeV and 350 MeV and the same beam intensity of 1 mA. The specific activity is averaged over the targets, taking into account some target reshuffling optimized to obtain a uniform activity. Burn-up effects are also taken into account, explaining the change of production rate observed after 1 day of irradiation. The targets are removed from the core after 6 days of irradiation as the gain in activity obtained after that period becomes marginal.

Because of the long delays existing in the supply chain between end of irradiation (EOI) and delivery to the hospital, ^{99}Mo calibrated activity is reported 6 days after EOI. This study indicates that a 350 MeV, 1 mA beam is needed in order to produce 5,000 ^{99}Mo 6-day Ci/week. That amount represents half of the weekly needs for North America and Western Europe together.

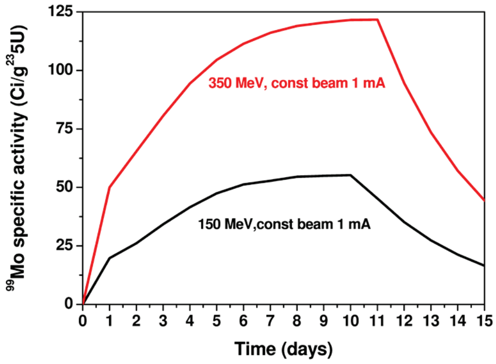

Fig. 3 Evolution with irradiation time of the average ^{99}Mo specific activity obtained in ADONIS targets for beams of 150 MeV and 350 MeV

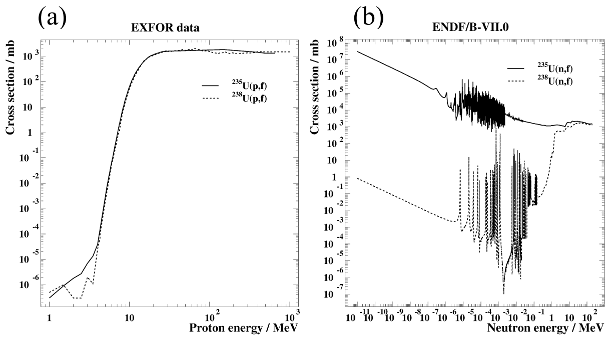

Fig. 4 Nucleon-induced ^{235}U and ^{238}U fission cross sections: (a) with protons; (b) with neutrons

III. Proton-Induced Fission

The ADONIS concept mostly relies on ^{235}U fission induced by thermal neutrons. The use of Ta for the spallation target leads to the emission of an intense flux of evaporation neutrons with energy below 10 MeV. Those neutrons subsequently get thermalized inside the core and captured by ^{235}U. However, ^{235}U and ^{238}U both exhibit a rather large fission cross section for nucleons above a few MeV. As shown in **Fig. 4**, the proton-induced fission cross section reaches about 1.5 barn above 20 MeV while the neutron-induced fission cross section exceeds 1 barn above 10 MeV. It is thus interesting to try and benefit from these large fission cross sections by shooting directly the proton beam on the U targets instead of using a spallation target. In addition, the interaction of protons with U also leads to the emission of evaporation neutrons that can be thermalized and captured by ^{235}U.

To check the validity of the Bertini intra-nuclear cascade model used in MCNPX to describe proton-nucleus reactions, we compared MCNPX predictions with data measured by Meier et al. for protons impinging on thick ^{238}U target.[6] The predicted and measured neutron doubly differential cross sections are compared in **Fig. 5** and are found in very good

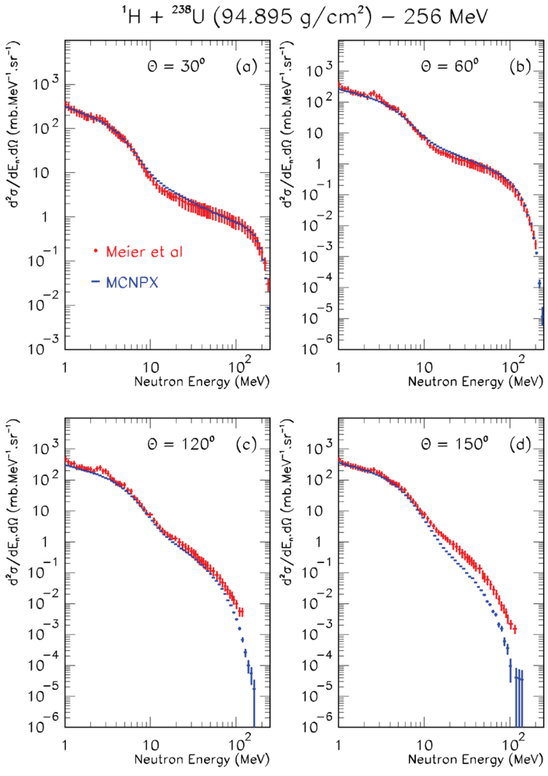

Fig. 5 Comparison of the neutron doubly differential cross section calculated with MCNPX and measured by Meier et al. for 256 MeV protons impinging on a 94.895 g/cm^2 thick ^{238}U target [6]

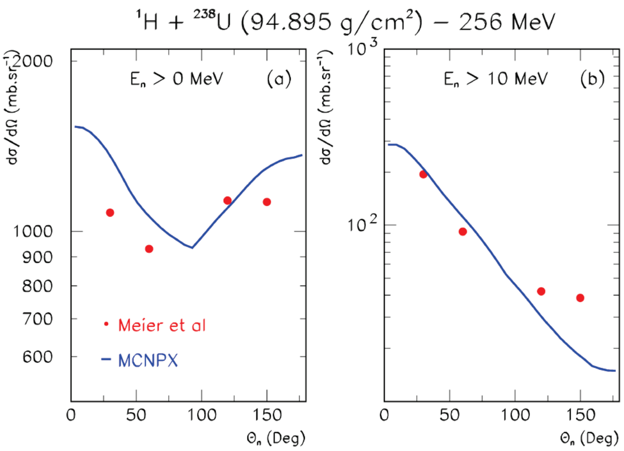

Fig. 6 Comparison of the angular differential cross section calculated with MCNPX and measured by Meier et al. for 256 MeV protons impinging on a 94.895 g/cm^2 thick ^{238}U target

agreement. **Figure 6** also demonstrates that the predicted and measured angular differential cross sections agree very well, both for evaporation and spallation neutrons.

To study the feasibility of this concept, a simple target assembly was modelled in MCNPX using targets made of 0.5 mm thick metallic Uranium foils with 3 or 5 cm radius. Only LEU targets were considered with ^{235}U fraction below 20%. The targets are stacked to form a cylinder and are separated by 1 mm thick water channel for cooling. The target stack is surrounded by a water volume but no reflector is

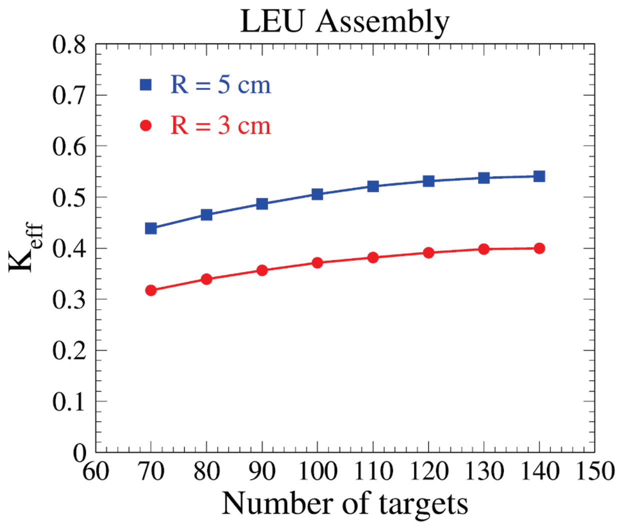

Fig. 7 Comparison of the k_{eff} values obtained for 3 cm and 5 cm radius targets as a function of the number of targets included in the system

added in order to limit the k_{eff} value of the system. The proton beam is scanned on the first target to cover the target surface. The nuclear data libraries used for the MCNPX simulations are the same as those used in the ADONIS study.

MCNPX simulations demonstrate that the k_{eff} value of such assembly varies with the number of targets in the range 0.3 to 0.4 and 0.45 to 0.55 for target radii of 3 cm and 5 cm, respectively (**Fig. 7**). The system is thus very far from criticality and will not be considered as a critical reactor from licensing point of view.

Considering primay beam energies of 200 MeV and 350 MeV, the ^{235}U and ^{238}U fission rates are estimated using an analytical evaluation for the proton-induced fissions and MCNPX for the neutron-induced fissions (the use of a tally multiplier with a F4 tally requires a measured fission cross section that is not available for protons). In ^{235,238}U fissions induced by thermal neutrons, ^{99}Mo is produced with a constant fission rate of 6%. In practice, mass distribution measurements for nucleon-induced fissions suggest a reduction of this ^{99}Mo production rate at intermediate energies down to 4%.[7] However, the majority of fissions generated in our system correspond to thermal neutron capture on ^{235}U and the rate overestimation due to the use of a constant fission rate of 6% is small.

Figure 8 exhibits the evolution with the ^{235}U fraction of the ^{99}Mo total activity obtained at end of beam after 66 h of irradiation with 200 and 350 MeV, 1 mA proton beam. This activity reaches 10,700 Ci for a ^{235}U content of 20%. It is reduced to 2,900 Ci with a beam energy reduced to 200 MeV and the same beam intensity.

Considering irradiation periods of 66 h followed by 6-day decay periods, the 6-day calibrated ^{99}Mo activities obtained per week are summarized in **Table 1** for different beam energies and target radii but a constant beam current of 1 mA. As for the ADONIS concept, it turns out that a 350 MeV, 1 mA beam is needed in order to produce about 5,000 ^{99}Mo 6-day Ci/week.

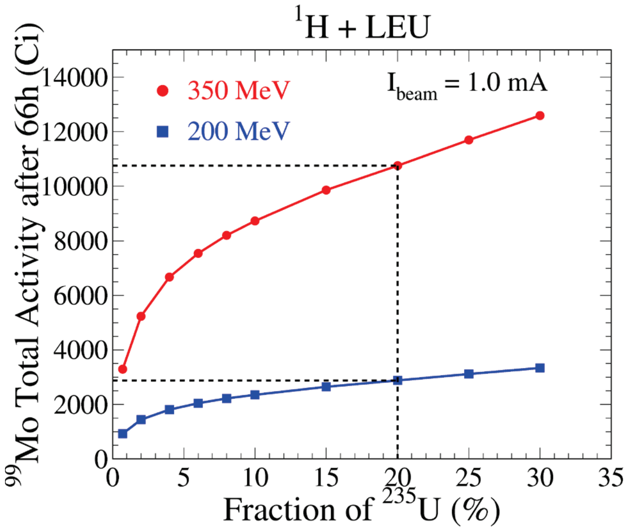

Fig. 8 Evolution as a function of ^{235}U fraction of the ^{99}Mo total activity produced after 66 h of irradiation with proton beams of 200 MeV or 350 MeV and a beam current of 1 mA. The results correspond to a target radius of 3 cm.

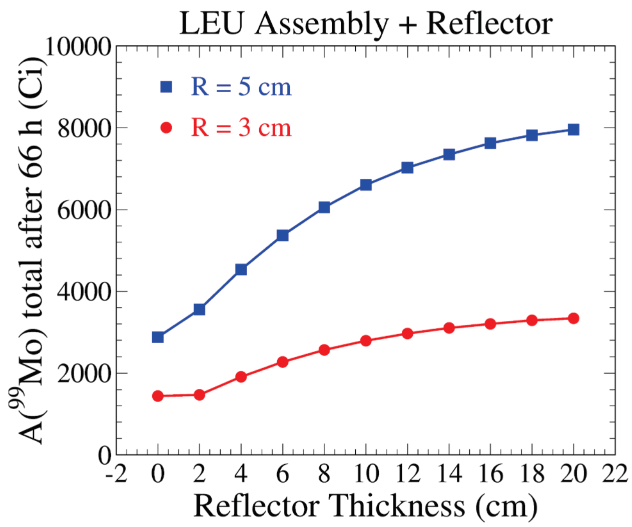

Fig. 9 Variation of the ^{99}Mo activity produced after 66 h of irradiation with the thickness of the reflector

Table 1 6-day calibrated ^{99}Mo activity obtained with the proton- induced system using a constant beam current of 1 mA

Beam energy (MeV)	Target radius (cm)	^{99}Mo activity (6-day Ci/week)
200	3	892
200	5	1470
350	3	2530
350	5	4940

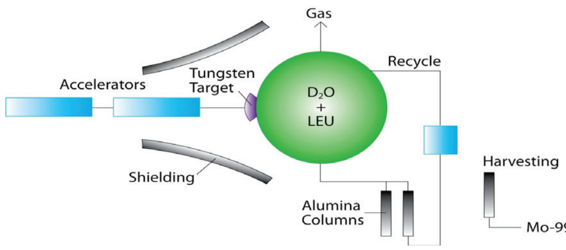

Fig. 10 Schematic view of the electron accelerator-based system designed by AMIC

The influence of a Be reflector surrounding the target assembly is also investigated. **Figure 9** demonstrates that the ^{99}Mo activity obtained after 66 h of irradiation using a 200 MeV, 1 mA beam can be multiplied by a factor 2.3 to 2.8 depending upon the target radius for a reflector thickness of 20 cm.

IV. Electron Accelerator-Based Solution

The two previous studies described in this paper involve the use of a high-energy and high-intensity proton accelerator. Although design studies for such machine have already been pursued, it does not exist today and its development would require major investments.

The new concept proposed by AMIC to produce very high ^{99}Mo activity using a medium-energy electron accelerator appears thus very attractive and MCNPX simulations are performed to prove the feasibility of that concept. This new approach is based upon the idea to create a large thermal neutron flux from an electron accelerator connected to a vessel containing a D_2O solution.[8] As shown in **Fig. 10**, the electron accelerator is used to produce high-energy X-rays from a high-Z target made in W or Ta and absorbing the primary electrons. The X-rays enter a spherical reaction vessel containing LEU salts dissolved in D_2O.

Because of the low threshold (2.23 MeV) and the significant cross section (2.2 mbarn at 5 MeV) for the $^{2}H(\gamma,n)^{1}H$ photonuclear reaction, a large flux of neutrons is released in the irradiated vessel. These neutrons are thermalized in the same vessel and can be efficiently captured by the ^{235}U atoms present in the solution. The neutron flux can be further enhanced by a neutron reflector surrounding the reaction vessel such as high-density polyethylene (HDPE) or Beryllium.

MCNPX simulations are performed using the BOFOD photonuclear data library for ^{235}U and ^{238}U.[9] Photonuclear reactions inside D_2O are simulated using the LA150U photonuclear data library from the LANL/T-16 group.[5] Neutron transport in D_2O is also performed using LA150 data library.

As shown in **Fig. 11**, the use of a thick HDPE reflector allows obtaining k_{eff} values very close to 1. The goal is to work with a k_{eff} value of 0.99 in order to obtain a thermal neutron flux amplification factor of about 100.

The beam power needed to produce an amount of 3,000 6-day Ci/week of ^{99}Mo is evaluated using MCNPX as a function of beam energy and k_{eff} value. **Figure 12** demonstrates that, for beam energies above 20 MeV and k_{eff} value close to 0.99, a beam power limited to 450 kW is enough to fulfill that goal. For a 20 MeV machine, that corresponds to a reasonable beam current of 22.5 mA.

Parallel calculations are performed by AMIC and lead to very similar results.[10] They demonstrate that a 24 MeV elec-

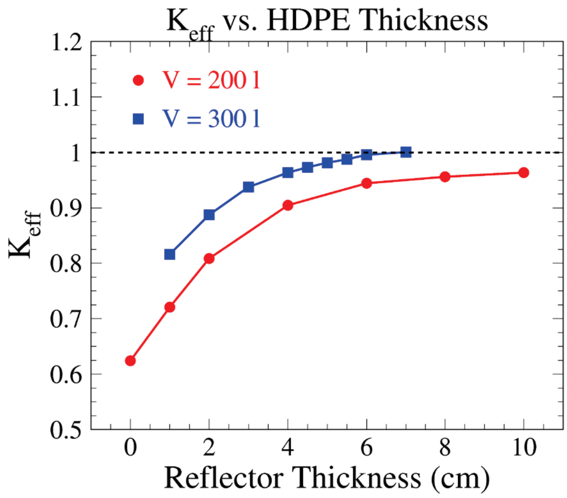

Fig. 11 Evolution with the reflector thickness (HDPE) of the k_{eff} value for different vessel volumes

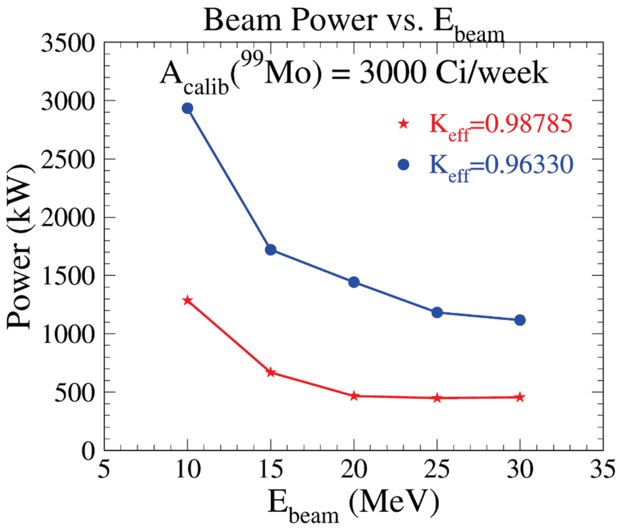

Fig. 12 Beam power needed to produce a ^{99}Mo activity of 3,000 6-day Ci/week as a function of beam energy

tron beam with a maximum current of 15.8 mA (equivalent to a beam power of 380 kW) is enough to produce a ^{99}Mo activity of 3,000 6-day Ci/week. Such a beam can be obtained by the use of two existing IBA TT300 Rhodotron connected in series.

Prototype experiments are now undertaken to confirm these neutronic calculations and validate the predictions that the system is self-limiting and cannot go critical.

Concerning the properties of the produced ^{99}Mo, specific activity, purity and by-product contents are expected to be similar to existing commercially available ^{99}Mo.

V. Conclusion

Due to the recent problems encountered by aging nuclear reactors used to produce ^{99m}Tc/^{99}Mo generators, IBA investigates the feasibility of new production methods based upon charged particle accelerators.

Solutions based on high-energy proton accelerators, with or without spallation target, offer the possibility to produce half of the ^{99}Mo world demand with a 350 MeV, 1 mA beam. However, building such an accelerator would require a major effort and a large investment.

The use of a medium-energy electron accelerator to irradiate a LEU+D_2O solution offers an elegant alternative with very interesting results. Moreover, the electron beam required to produce a very high ^{99}Mo activity of 3,000 6-day Ci/week can be generated with existing electron accelerators such as the IBA Rhodotron TT300.

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