A PROTON-DRIVEN, INTENSE, SUBCRITICAL, FISSION NEUTRON SOURCE

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To replace the ageing nuclear reactors used today for radioisotope production, research and industrial applications, we propose to use a spallation neutron source, with neutron multiplication by fission. A 150 MeV cyclotron could be used to produce a proton beam hitting a molten Pb-Bi primary target, surrounded by a water moderator, producing about 1 primary neutron per incident proton. The primary spallation neutrons, moderated, would strike a number of secondary targets containing a subcritical amount of ^{235}U. Typical thermal neutron fluxes at the targets location would be around 2 10^{14} n/cm².s. Such a system can be used, for example, to supply the world demand in ^{99}Mo, a fission product of ^{235}U distributed as generators of ^{99m}Tc, the most frequently used radioisotope in nuclear medicine. The non-critical nature of the system would make it more acceptable for the public than a nuclear reactor and should simplify the licensing process. Price, cost of operation, of disposal of radiowaste and of decommissioning should also be advantageous compared to nuclear reactors. The present paper presents the progress achieved in the design of the proposed system.

1. Introduction

Technicium 99-m is, by a large extent, the most widely used radioisotope in nuclear medicine. In Europe only, approximatively 7 million ^{99m}Tc studies are conducted each year. The ^{99m}Tc is normally supplied to the hospital as $^{99}Mo \Rightarrow ^{99m}Tc$ generators. The ^{99}Mo has a half life of 66 hours, versus 6 hours for the ^{99m}Tc, making the logistics of distribution much more practical for the ^{99}Mo generator than for the short-lived ^{99m}Tc. Most of the ^{99}Mo used in nuclear medicine is obtained as a fission product of ^{235}U. The chain of 99-mass radioisotopes is obtained in 6.074 % of the fissions made by thermal neutrons.

The world production of fission ^{99}Mo is today made in a very small number of research reactors which are getting quite old and are due, in the next years, for a major refurbishment or for decommissioning. The problems related to the future availability of reactors suitable for the production of medical radioisotopes, mainly ^{99}Mo, for research, and for industrial applications, has prompted a renewed interest on alternative, accelerator based methods of production.

2. The direct accelerator production of ^{99m}Tc and ^{99}Mo

The direct accelerator production of ^{99m}Tc or of ^{99}Mo by proton bombardment of ^{100}Mo between 15 and 70 MeV has been studied during the last years[1].

Although the "direct" ^{99m}Tc yield is high, the ^{99}Mo generator yield is low and the resulting product is unlikely to be separable from the ^{100}Mo target material. The proton bombardment of ^{100}Mo offers thus no alternative to the present neutron production of ^{99}Mo.

Due to its short 6 hours half life, the directly made ^{99m}Tc would require a fundamental change in the logistics of distribution. An "Instant Tech." production unit could only be a regional scale facility. A loss of 2...3 half life or 75% to 85% of the produced activity seems unavoidable. The specific activity of "Instant Tech." is expected to be lower than that of generator ^{99m}Tc at the time of use and further investigations are needed to know if this lower specific activity meets the clinical needs. In any case, it is very likely that the diagnostic radio pharmaceuticals that have been FDA licensed to be labelled with generator produced ^{99m}Tc, would require a significant relicencing if used with "instant Tech".

These reasons explain why this method hasn't been yet adopted by the industry. In all instances, a ^{99}Mo source will always be needed to supply remote areas.

3. An accelerator based source of fission ^{99}Mo

A number of reasons favor the continued use of ^{235}U fission reaction for the production of ^{99}Mo:

-the high production yield, resulting from the large cross-section of fission of ^{235}U by thermal neutrons.

-the very high density of activity in the irradiated samples, allowing to perform the separation chemistry on reasonably low amounts of material. The saturation yield of ^{99}Mo for pure ^{235}U irradiated in a thermal neutron flux of $2*10^{14}$ n/cm^2 is around 335 Ci/gram. The total world production of

2.7 10^4 Ci/week requires to process around 100 gr of irradiated ^{235}U each week. This is a quite reasonable amount.

-the possibility to continue to use the existing - and very expensive - fission ^{99}Mo chemical separation facilities.

-the possibility to avoid or minimize the re-licencing process for all radioactive diagnostic drugs labeled with ^{99m}Tc from ^{99}Mo=>^{99m}Tc generators.

There are, however, downsides to the use of ^{235}U fission ^{99}Mo:

-many other fission radioisotopes are produced at the same time as ^{99}Mo. Fortunately due to the relatively short irradiation time of a target (typically one week), the production of long lived radioisotopes is, relatively, minimized. However the process produces significant amounts of short and medium lived radioactive waste.

-to minimize the amount of material to be chemically separated, and to minimize the production of actinides by neutron capture on ^{238}U, the targets use high enrichment, bomb grade ^{235}U. Despite the small amount used, this is undesirable from a non proliferation stand-point and imposes a strict security for the use, transport, inventory and accounting of the target material.

Following[2] we present here an accelerator based neutron source for the production of fission ^{99}Mo, and of other fission or reactor produced isotopes. This production system includes the following elements:

1) a cyclotron able to accelerate 1.5 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 one of several possible neutron sources

3) a primary beam target, where the proton beam strikes a molten Pb or Pb-Bi target, producing spallation (mostly evaporation) neutrons. The expected neutron yield at 150 MeV is 0.96 neutron/proton

4) a water moderator surrounding the lead target

5) a number of secondary targets made of highly enriched ^{235}U. Because the mass of ^{235}U is strictly subcritical, and arranged so as to produce the highest possible reactivity, any perturbation to the system would reduce the reactivity. The mechanical layout of the system would be such that the introduction of additional targets would be a mechanical impossibility.

A first, possible target assembly was presented elsewhere[2]. In this design, the primary target is surrounded in all directions by a water moderator, in order to thermalize the primary spallation and secondary fission neutrons. The secondary targets, made of highly enriched ^{235}U encapsulated in an appropriate cladding, are tube shaped in order to improve the heat dissipation. Each secondary target is placed at the end of a special handling rod, and located in an individual aluminium water cooling tube providing high velocity water cooling. The secondary targets are located at close distance from the primary spallation target.

4. The 150 MeV, 1.5 mA Cyclotron

The description of the 150 MeV, 1.5 mA cyclotron is not the subject of this presentation. It is however important to note that Ion Beam Applications has built more than 15 lower energy (30 MeV), high current (0.5 mA) H^- cyclotrons for radioisotope production. Such cyclotrons are used today by all major radiopharmaceutical companies for the production of medical radioisotopes. High intensity versions (1 mA or more) of the CYCLONE 30 are now available. A key component of this upgrade is higher brightness H^- multicusp ion sources developed for IBA by AEA Technology in Culham (G.B.)[3].

Recently experimental results and calculations were presented[4] showing that the space charge limit for current designs of H^- cyclotrons was between 5 and 10 mA of beam current. These results show also that very high beam loadings of the cyclotron RF system - up to 80% - are possible. The results of another IBA accelerator, the Rhodotron[5] 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, 1.5 mA H^- cyclotron could therefore reach 50%, i.e. a total electrical power of only 450 kW for 225 kW of beam power.

The problem of the electromagnetic dissociation of H^- 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. The alternative design of a positive ion cyclotron with the auto-extraction system proposed by Yves Jongen et al. in another communication to this conference (Paper N° CC-4) would have the advantage of significantly reducing this size.

5. The production of the primary (spallation) neutrons

The 150 MeV, 150 to 225 kW proton beam is used to produce spallation neutrons in a molten Pb or Pb-Bi target. The neutron production yield from protons on lead targets has been measured by Bell et al.[6] at energies below 80 MeV, and by Carpenter[7] between 0.5 and 1.5 GeV. Using the cross-sections of reference 6 at low energy, assuming a constant cross-section of 1.61 Barn above 70 MeV and fitting the neutron multiplicity at higher energies to meet the results of reference 7, we find the neutron yield/incident proton in the 0-200 MeV range (see fig. 1).

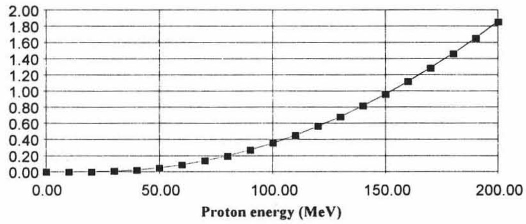

Figure 1 : Calculated neutron yield per incident proton on lead in the 0-200 MeV range.

Incidentally, a measurement of the total neutron yield at 100 MeV made by Lone et al.[8] shows a value of 0.35 neutron/incident proton at 100 MeV, in excellent agreement with our crude calculation.

The 225 kW beam power will be handled by an appropriate flow of liquid (molten lead for example) maintained through the target, resulting in a temperature rise of 200°C. The liquid lead will be circulated in closed loop.

6. Moderator, reflector, secondary targets and neutron calculations

The neutron transport, thermalisation and multiplication in this assembly were simulated using numerical neutron transport codes at SCK-CEN. The results of neutronics calculations using the DTF-4[9] and DORT[10] codes show that a k_{eff} of 0.75 is obtained with 30 targets containing each 4gr of ^{235}U, or a total of 120 gr of ^{235}U, yielding a neutron multiplication factor of 4 but staying far enough from a critical mass. More details on these calculations are presented elsewhere[11].

In this case, using a 1 ... 1.5 mA beam of 150 MeV protons on the primary lead target, a thermal neutron flux of 2 10^{14}n/cm^2.s should be obtained at the secondary fission targets. The resulting thermal fission power would be 600 ... 720 kW. In a flux of 2 10^{14}n/cm^2.s, a properly designed ^{235}U target would reach a saturation activity of 335 Ci of ^{99}Mo/gram. With a loading of 100 gr, and replacing the targets on a weekly basis, the weekly production of ^{99}Mo would be 2.77 10^4 Ci, or 6,000 Ci/week with a 6-day post calibration. This amount represents approximatively the world weekly demand in ^{99}Mo.

It should be noticed that the decay time of ^{235}U in a thermal neutron flux of 2 10^{14}n/cm^2.s is approximatively 160 days. By replacing the targets on a weekly basis, the reactivity of the assembly will hardly vary, and most of the ^{235}U content of the targets will be unused and could be recycled. The short irradiation time, and the small amount of ^{238}U in the targets will minimize the production of actinides and, generally, of low lived radioisotopes.

7. Preliminary engineering design of the primary (spallation) target

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 and conduction of the target material itself.

One of the items of concern is the preheating and controlled cooling down of this heavy metal liquid. If one considers a closed circuit, without any dilatation possibilities, one will experience major problems to control the start-up and cool-down stage of operation. We are therefore considering a drain tank where the liquid can be pre-heated and cooled down in a controlled manner.

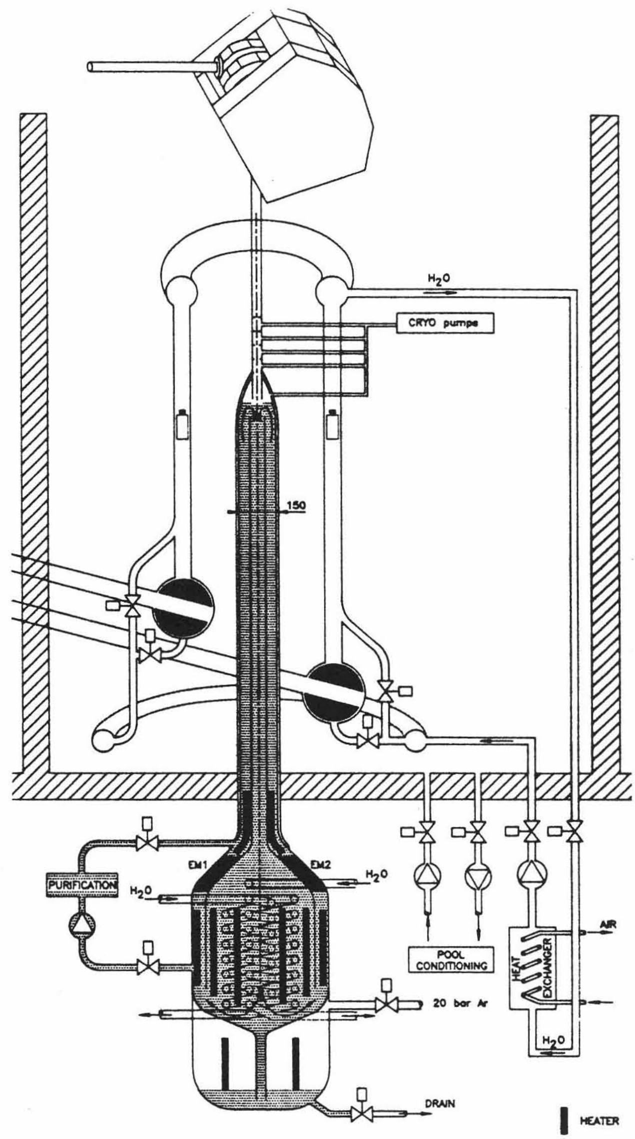

Figure 2: Preliminary design of the target.

The proposed target[12] 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. In the present design, the volume of the fluid is about 1.5 cubic meters.

Points presently under consideration include, among others, the nozzle design, the materials compatibility, the purification requirements, the neutron damage in the lead-bismuth nozzle container and the long term corrosion of this container.

8. Conclusion

We have shown the possibility to replace the nuclear reactors currently used for the production of fission-based radioisotopes, for research and for industrial applications, by a cyclotron driven spallation neutron source, with neutron multiplication by fission. The system would be far enough from a critical mass and would present, therefore, an unquestionable safety. As far as the production of ^{99}Mo is concerned, one such system could in principle supply the world demand and would cost significantly less than a commercial, 10 MW isotope production reactor. Costs of operation, radioactive waste management and decommissioning would be significantly lower also. Finally, the non critical nature of the system would make it more acceptable for the public opinion than a nuclear reactor, and should simplify the licensing process as well.

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