Advanced Accelerator Applications

BRIDGING SCIENCE WITH LIFE

Physics for Health, CERN, February 2010

Technical evaluation of an accelerator-driven production of Mo-99 for Tc-99m generators at CERN (MolyPAN project)

S. Buono, N. Burgio, **L. Maciocco**

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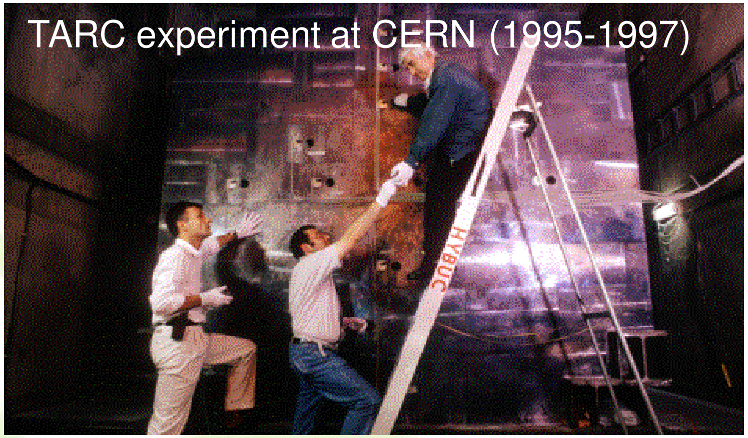
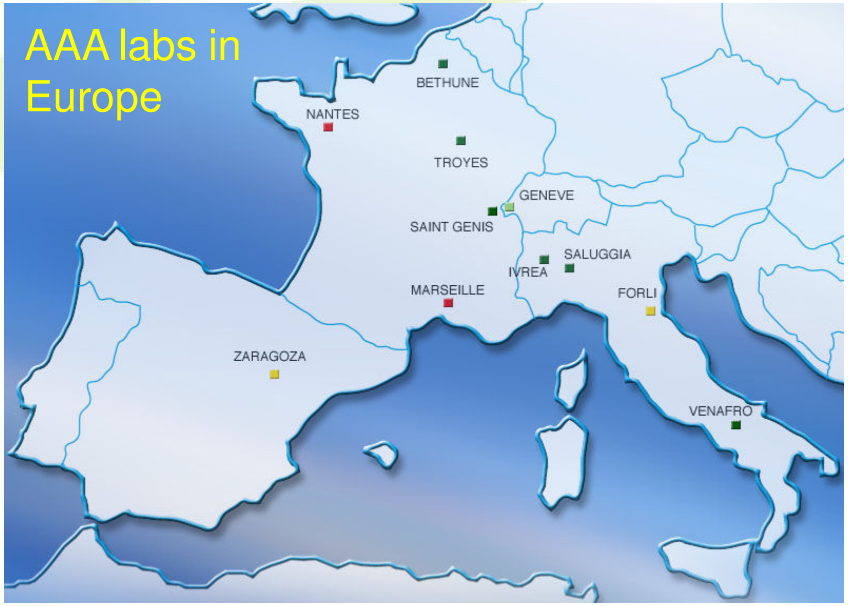
Advanced Accelerator Applications (AAA)

- AAA has been founded in 2002 (it can be considered a CERN "Spin-off")
- The idea at the origin of AAA was the exploitation of the Adiabatic Resonance Crossing patent, developed at CERN by Carlo Rubbia and his team (which included AAA founder Stefano Buono), with the aim of efficiently producing short-lived neutron-activated elements for medical applications using particle accelerators



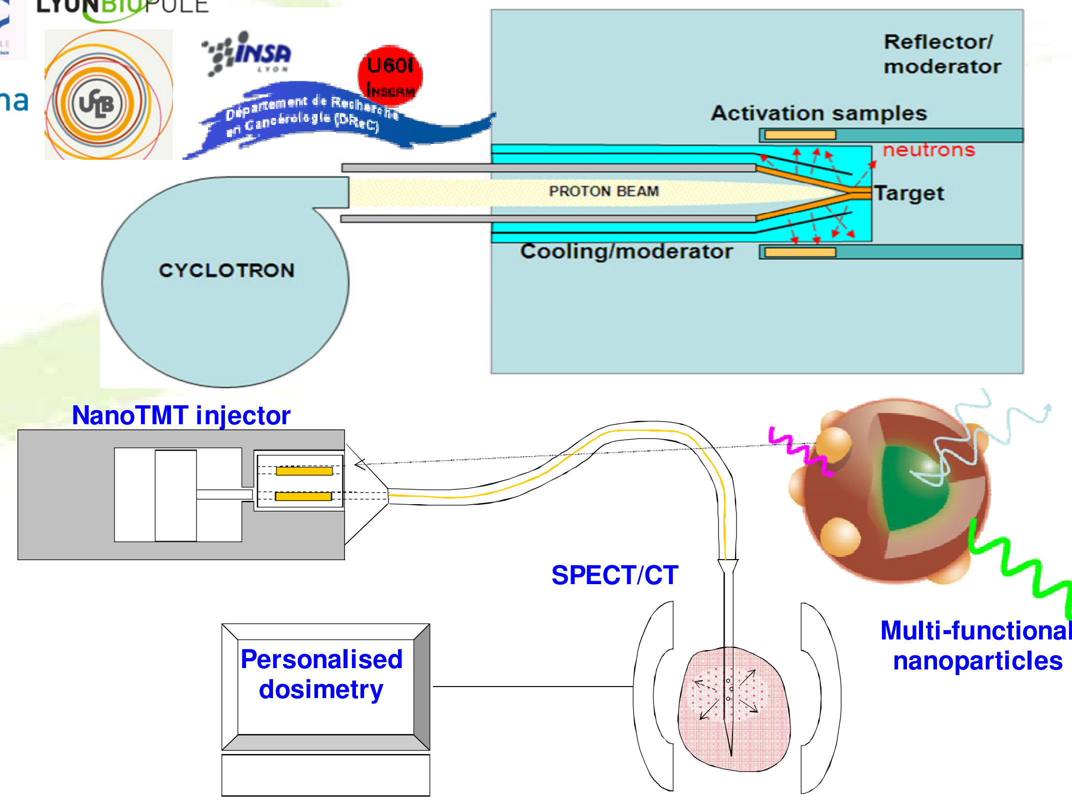
- In other to support its R&D activity, a commercial activity has been set up for the production and distribution of FDG for PET scan
- AAA is nowadays a European leader in the PET tracers production and distribution.
- Currently AAA is involved in 27 research projects with 69 partners (46% private and 54% public)
- AAA's R&D focuses on innovative diagnostic (molecular imaging) and therapeutic (personalised medicine).



Application of the ARC method in Brachytherapy

- Since 2003 AAA and its partners are working on the development of an innovative brachytherapy technique using nanoparticles activated in a cyclotron driven neutron activator (see poster session)

THERANEAN project



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Mo-99 production from a particle accelerator

- The worldwide supply chain of Mo-99 is essentially based on the production from 5 nuclear research reactors that are approaching the end of their operational lifetime
- The ageing of the installations are inducing unscheduled shutdowns that provoke severe shortage of Mo-99, hitting the nuclear medicine community
- As a consequence, the feasibility of a Mo-99 production cycle based on particle accelerators is being assessed, as it may presents many advantages in terms of safety, cost, time to market as well as environmental and proliferation issues

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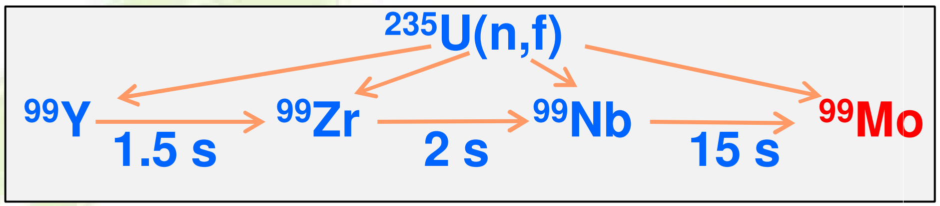
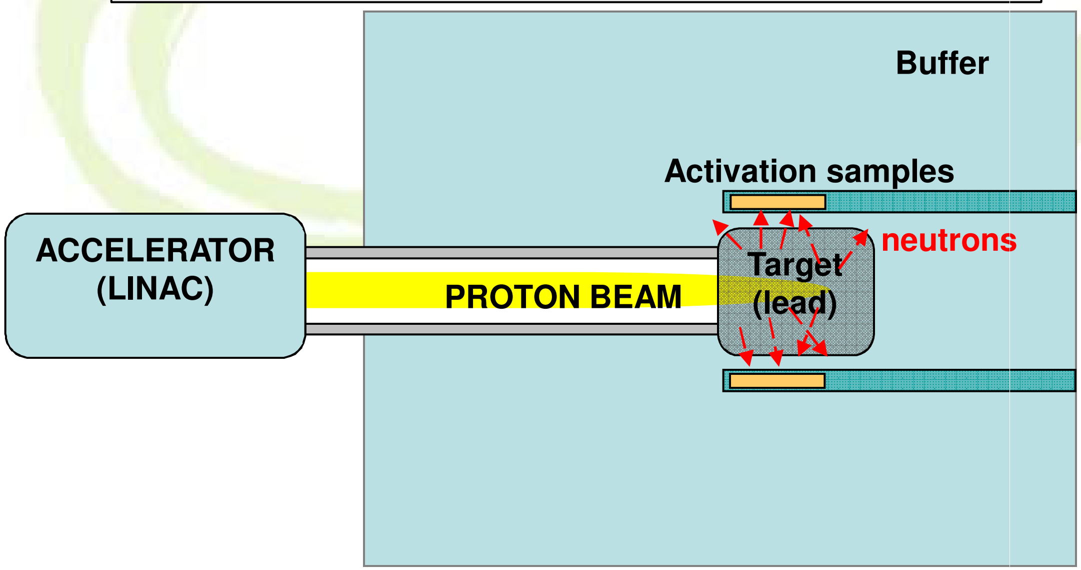
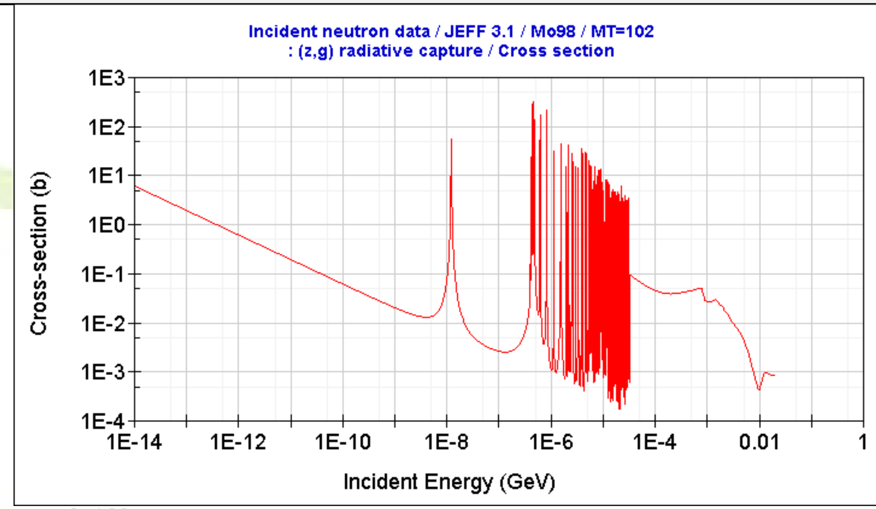
Principle of operation

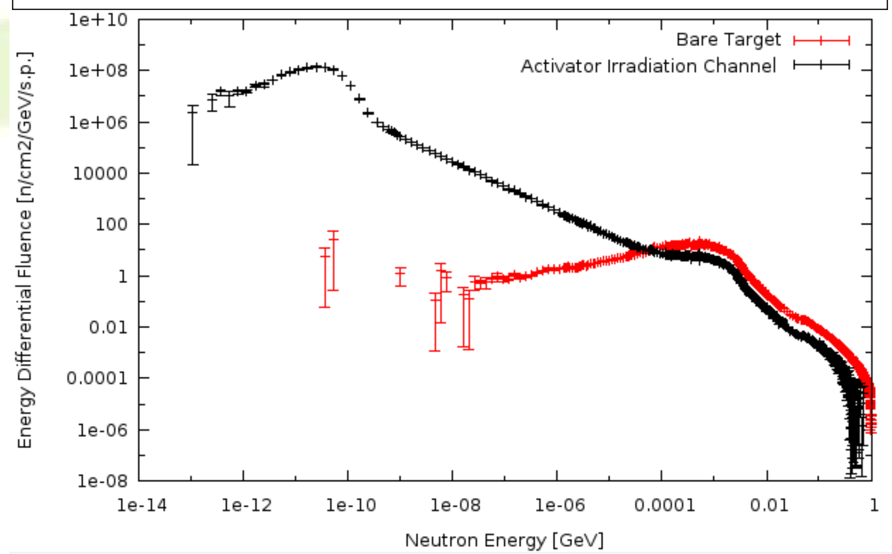
- Objectives
  - ✓ Progressive neutron moderation with minimum absorption
  - ✓ Neutron confinement
  - ✓ Most suitable neutron spectrum for the relevant reactions





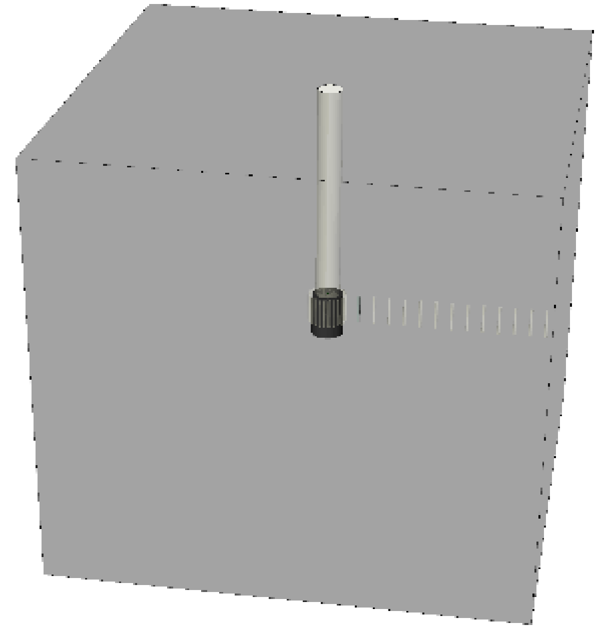
^{98}Mo(n,γ) → ^{99}Mo

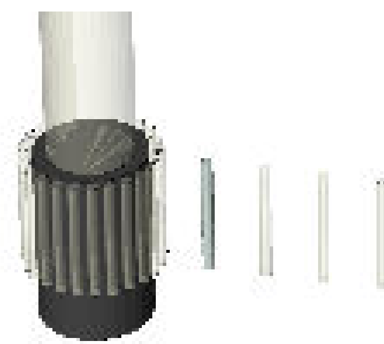




Monte Carlo model of the 1 GeV activator

- MCNPX 2.5.0 (JEFF 3.1 cross sections) and FLUKA 2008.3B were used in a comparative way
  - The activator model is made of 4 main components
    - ✓ Proton beam line
    - ✓ Cylindrical liquid Lead target (h=30 cm, r=10 cm)
    - ✓ Buffer block (300 x 300 x 300 cm^3)
    - ✓ Irradiation channels (r= 1 cm, h= 20 cm) uniformly filled with the activation sample. First ring at 1 cm from the target, next series placed at a radial step of 10 cm
  - Three activator configurations were analysed
    - ✓ Lead buffer (best ARC effect, worst moderation)
    - ✓ Graphite buffer (good moderation, low absorption)
    - ✓ Water buffer (best moderation, worst absorption)
  - Neutron target materials
    - ✓ Natural (23.8%)/ Enriched (99.9% ^{98}Mo) Mo oxide
    - ✓ Low (3% ^{235}U) /High (100% ^{235}U)-Enriched Uranium





Summary of activator performances

- According to Monte Carlo codes, the graphite configuration gives the best results both for neutron-capture and for fission production of ^{99}Mo

Case	Saturation yield [GBq/g/mA]			
	Nat Mo	Enriched Mo	LEU (3%)	HEU (100%)
Lead	9	28	6	-
Graphite	15	46	106	1730
Water	6	27	62	-

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Effect of LEU target optimisation (MCNPX)

- A major improvement of the HEU/LEU yield can be obtained
  - ✓ by using the highest possible enrichment for LEU (20%)
  - ✓ with an optimisation of the target assembly (factor 20 on LEU-20% according to FLUKA) due to reduction of self-shielding effects
  - ✓ In the following estimations, a factor 0.3 has been applied to these figures (structural components, cooling, temperature effects etc.)

Case	^{99}Mo Saturation yield [GBq/g/mA]	
	HEU (100% ^{235}U)	LEU (19.99% ^{235}U)
Thick uniform cylinder (r= 1 cm h=20cm)	1730	707*
Thin hollow cylinder 125 um thick (r_{max}= 0.9 cm, h=20 cm).	42600	14800

* Extrapolated from results with LEU-3%

Comparison of reactor and activator ^{99}Mo production

Input data Derived data

Production method	Flux	Target	Mo-99 Satur. Yield	Irrad Time	Target mass per batch	Proc time	A-EOP per g of Mo	A tot EOI (EOI)	Weekly capac. (6 days)	Share of world market
	[n/cm2/s]		Ci/g	days	g	days	Ci/g	kCi	kCi	%
Reactor (typical n flux in Petten)	2.0E+14	HEU-98%	269.8	4	300	1	CF	51.4	11.3	113%
		LEU-20%	93.7	4	300	1	CF	17.9	3.9	39%
		Mo-98 ox	0.7	4.5	2000	0.5	0.4	1.0	0.2	2%
GRAPHITE activator 3x3x3 m (1 GeV-1mA)	3.2E+14	HEU-98%	345.4	4	364	1	CF	79.9	17.6	176%
		LEU-20%	120.0	4	364	1	CF	27.7	6.1	61%
		Mo-98 ox	1.0	4.5	20000	0.5	0.6	13.3	2.9	29%

- Comparison based on 1 mA current (comparable neutron fluxes), but technically possible to use higher currents (up to 4 mA?)
- Maximum LEU enrichment (20%)

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- Reactor LEU fission yields based on typical thermal neutron cross-section (possibly overestimated even for optimised LEU targets)
- HEU yield (exact figure not found in literature) scaled with the ^{235}U content and with the yield ratio HEU/LEU from activator Monte Carlo simulation (self-shielding effects)
- Activator fission yields based on results on optimised HEU/LEU target scaled of a factor 0.3 to take into account effects of target structure, cooling etc.

Comparison of reactor and activator ^{99}Mo production

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- Reactor $^{98}Mo(n,\gamma)^{99}Mo$ cross section estimated equal to the thermal cross section. Effect of epithermal neutrons (factor 2 according to *Ryabchicov et al.*) considered compensated by self-shielding effects.
- Activator $^{98}Mo(n,\gamma)^{99}Mo$ yield estimated with MCNPX (the most conservative estimate) at full load (self-shielding effect included)

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Comparison of reactor and activator ^{99}Mo production

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- Based on a 5-days weekly cycle
- 1 day (24h) of processing time for fission-produced Mo
- 0.5 days (12h) processing time assumed for activated Mo

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- Typical value of HEU load in reactor ~300 g according to available information
- HEU/LEU load of 364 g in the activator calculated with full load of optimised (thin) target in first ring.
- Simulations carried out at full load in the Mo-oxide irradiation (to take into account distance and neutron self-shielding). 10 times less loading capacity considered in reactor

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- ^{99}Mo delivered as Ammonium Molybdate in the HEU-LEU cases after target processing. Not relevant for specific activity in generators (carrier free)
- Carrier mass equal to target mass for ^{98}Mo. Max theoretical amount of Mo in a 10 g Alumina generator = 0.2 g.
- ~300 mCi standard generators theoretically possible with ^{98}Mo in a 4 mA activator, but in general alternative generator technology necessary for ^{98}Mo activation

Comparison of reactor and activator ^{99}Mo production

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- According to the present estimation (not taking into account processing losses), a continuous operation of a reactor like Petten-HFR loaded with 300 g of **HEU** would cover 113% of the total demand (to be compared with actual figures)
- Better figures are obtained with the activator, mainly due to the higher neutron flux and to the higher assumed loading capability, allowing to largely cover the total world demand with 1 mA of proton current on 364 g of HEU

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Comparison of reactor and activator ^{99}Mo production

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		Mo-98 ox	1.0	4.5	20000	0.5	0.6	13.3	2.9	29%

- Concerning LEU-^{99}Mo production, results in the activator are scaled of a factor 3 with respect to HEU (same factor assumed for the reactor, to be compared with actual figures)
- The global world demand could be covered with 364 g-load of LEU-20% and a 2 mA activator

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Comparison of reactor and activator ^{99}Mo production

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		Mo-98 ox	1.0	4.5	20000	0.5	0.6	13.3	2.9	29%

- Concerning **^{98}Mo(n,γ)^{99}Mo production**, much better figures are found with the activator at 1 mA clearly due to assumed higher loading capabilities (but also to better specific yield)
- **The global world demand could be covered with 20 Kg-load of ^{98}Mo-oxide and a 4 mA activator.** The **load of 40 kg** is likely feasible without degrading the system performances. In this case **2 mA would be enough to cover the world demand**

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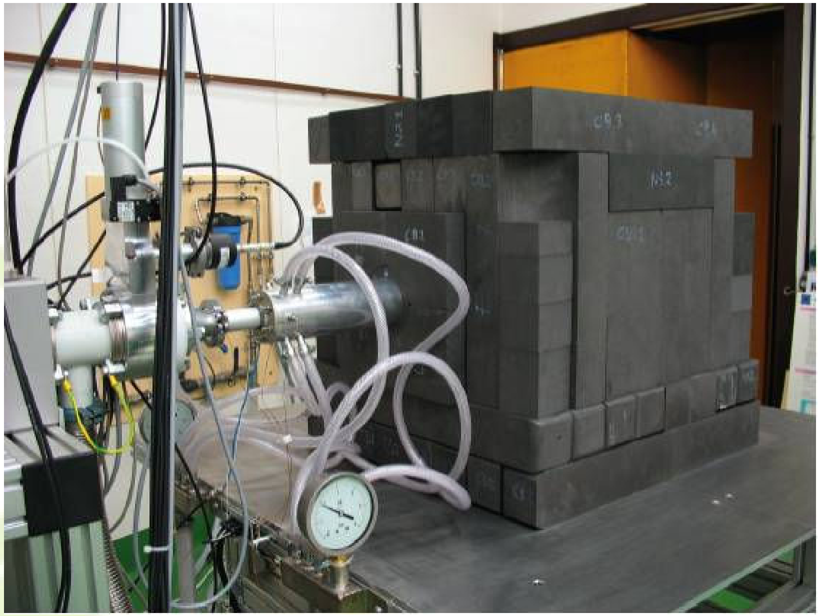
Conclusions

- According to preliminary Monte Carlo estimations, a Lead target-graphite buffer activator coupled with a proton LINAC would allow to cover the whole world demand of 99Mo with
  - ✓ 350 g of LEU-20% and a proton beam of 2 mA (700 g of LEU and 1 mA possible)
  - ✓ 20 Kg of ^{98}Mo-oxide and a proton beam of 4 mA.
- Available design studies (Energy Amplifier, Eurotrans, ESS, Eurisol) and experimental evidences (Megapie) support the technical feasibility of a 1-4 MW liquid metal target.
- The problem of the nuclear-waste (activated lead target) management must be considered
- Alternative technologies for the use of ^{98}Mo-oxide carrier in ^{99m}Tc generators are necessary (encouraging results are available for PZC-based generator)
- Technical and economical aspects need to be further assessed

Comparative results of MCNPX and FLUKA on the INBARCA activator

- A comparison of ^{98}Mo(n,γ)^{99}Mo yields with experimental results on the IBARCA activator show a tendency of both codes to underestimating experimental results, especially with harder spectra (Target alone)
- In the case of the enriched Mo oxide, MCNPX underestimates the experimental results of a factor 6



Case	Sample	Saturation yield [kBq/g/µA]		
		MCNPX	FLUKA	EXP
Target alone	Nat Mo foil	19	78	307
Lead-buffer		395	319	629
All graphite		401	558	1000
	Enriched Mo-98 oxide	338	Cross sec. NA	1890

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36 MeV-10 mA (CLUSTER type) ^{99}Mo prod capacity

Input data Derived data

Production method	Flux	Target	Mo-99 Satur. Yield	Irrad Time	Target mass per batch	Proc time	A-EOP per g of Mo	A tot EOI (EOI)	Weekly capac. (6 days)	Share of world market
	[n/cm2/s]		Ci/g	days	g	days	Ci/g	kCi	kCi	%
36 MeV – 10 mA (CLUSTER) activator	1.2E+13	Mo-98 ox	0.05	4.5	1000.0	0.5	0.31	0.347	0.076	0.8%
70 MeV – 10 mA (CLUSTER) activator	3.0E+13	Mo-98 ox	0.13	4.5	1000.0	0.5	0.76	0.870	0.190	1.9%

- Data at 36 MeV based on experimental data from the INBARCA activator
- Yield at 70 MeV extrapolated on the base of simulated neutron flux (factor 2.5)

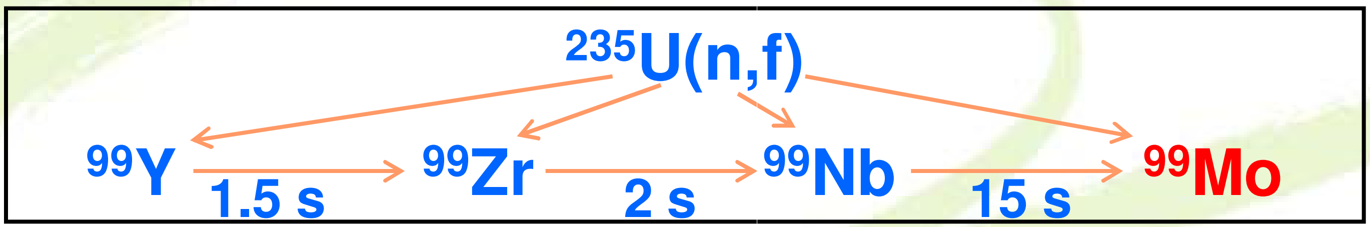
Comparative results of MCNPX and FLUKA

	$^{98}Mo(n,\gamma)^{99}Mo$ on natural Mo oxide [atoms/sp]			Fission on ^{235}U [fissions/sp]		
Activator Model	MCNPX	FLUKA	MCNPX/ FLUKA	MCNPX	FLUKA	MCNPX/ FLUKA
Lead Buffer	7.10E-06	3.15E-05	1/4.5	0.39	0.33	1.2
Graphite Buffer	1.11E-05	5.09E-05	1/4.6	0.99	0.83	1.2
Water Buffer	4.28E-06	2.22E-05	1/5.2	0.49	0.44	1.1

- Higher $^{98}Mo(n,\gamma)^{99}Mo$ yield estimated by Fluka (factor 5), possibly related to cross-section modeling in the resonance region
- Rather good agreement on ^{235}U fission yields (slightly better figures from MCNPX)
- **Both codes estimate the best yield for both reactions in the graphite-buffer configuration**

99Mo fission yield

- The ^{99}Mo yields from ^{235}U(n,f) has been estimated considering the decay chain of all the fission products decaying into ^{99}Mo



- Calculations confirm the cumulative 6% yield of ^{99}Mo per fission typical of thermal neutron fission also for the activator spectrum

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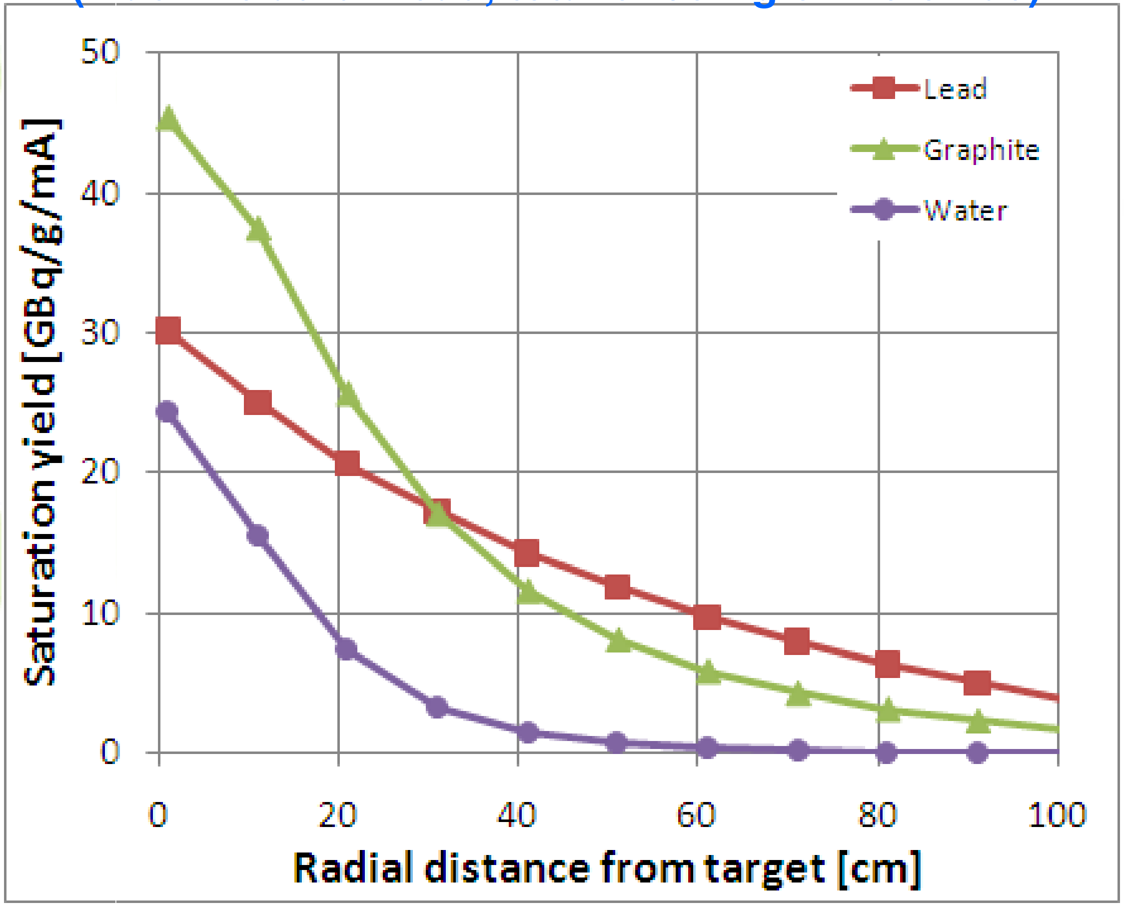
^{98}Mo oxide loading capacity

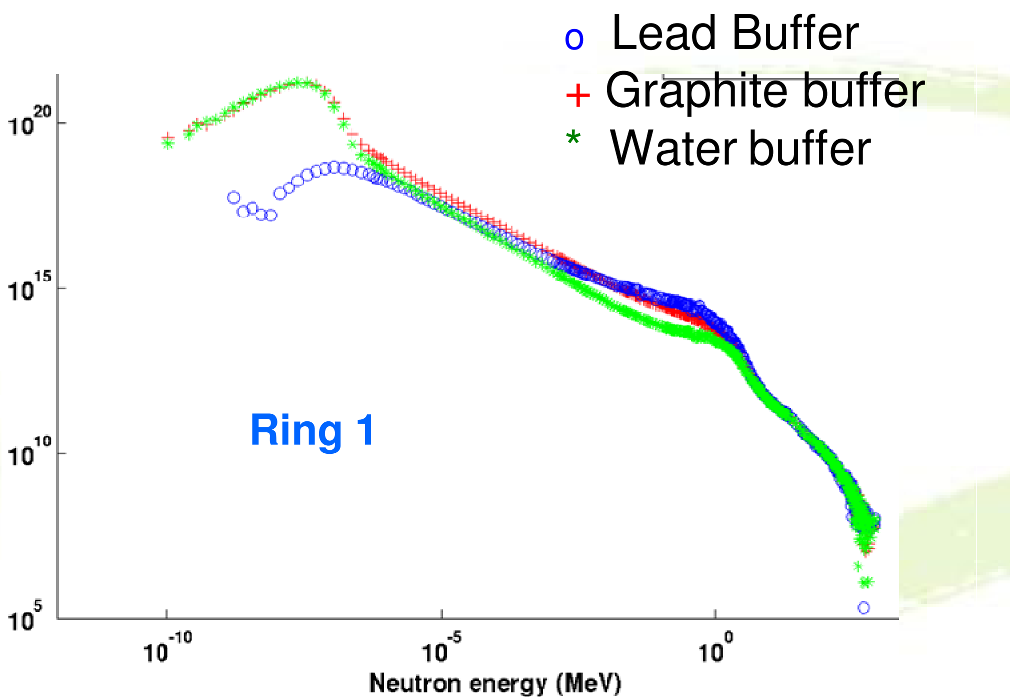
- As expected, Lead shows the smoother reduction of the yield with the distance (ARC effect)
- Graphite shows a factor 0.6 decrease in the first 3 rings, so guaranteeing a good loading capability (minimum 20 Kg in the first 3 rings)
- Water show a high neutron absorption with a rapid decrease of the yield with the distance (lower loading capabilities)

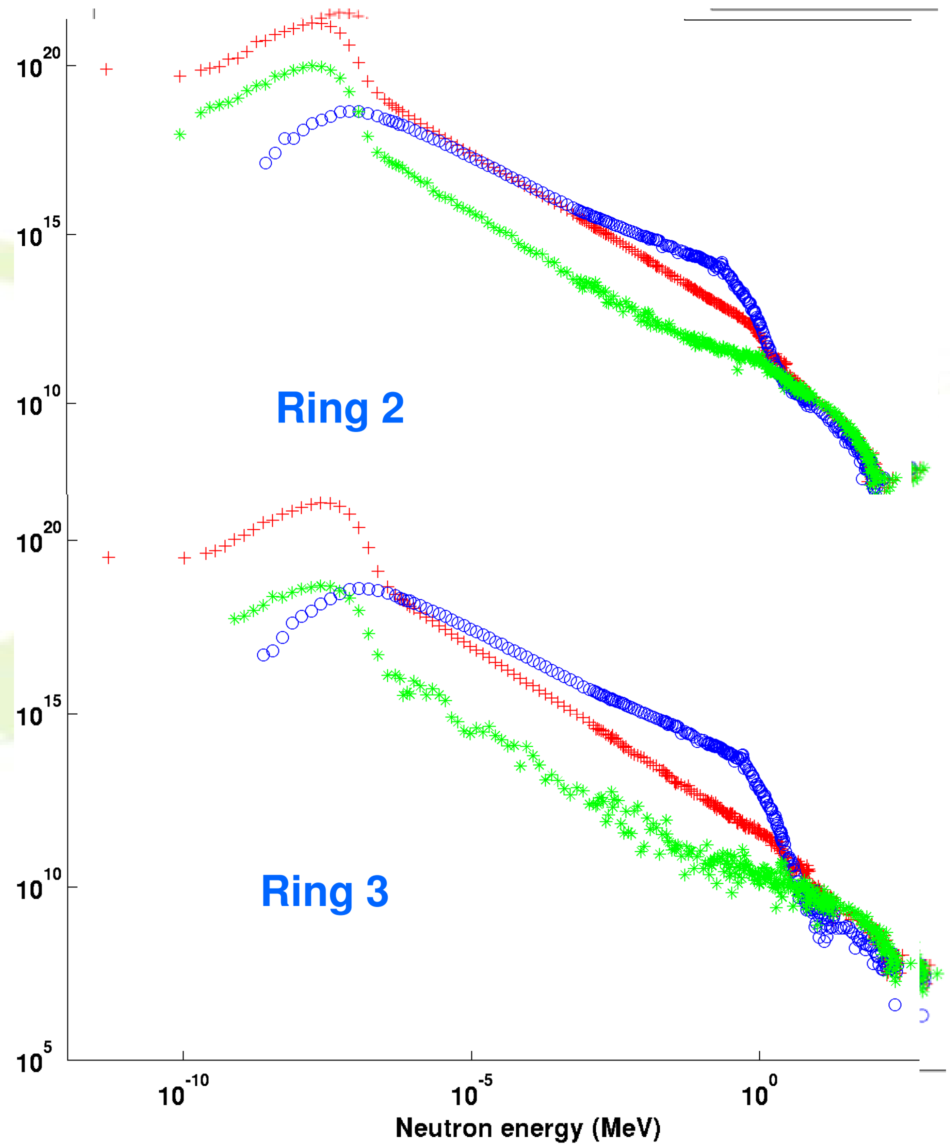
Variation of the 98Mo(n,γ)99Mo yield with the distance from the target in the three configurations (machine at full load, total of 95 Kg of Mo oxide)



Neutron flux variation with distance`

Differential flux (n/cm²/MeV/mA)





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Provision of ^{98}Mo oxide / LEU

- The possibility to produce large amounts of ^{98}Mo oxide should be assessed
- The cost of a small quantity (~1 g) is at present around 2.5 $/mg. This would imply a cost of 50 M$ per 1 activator load of 20 Kg. A large scale production of ^{98}Mo oxide is expected to significantly reduce the price (at least a factor 10 giving a cost in the order of 5 M€ per load)
- The technical/economical feasibility of recuperating the ^{98}Mo-oxide from the used generators should be assessed
- A cost analysis for the supply of LEU at different enrichment should be done

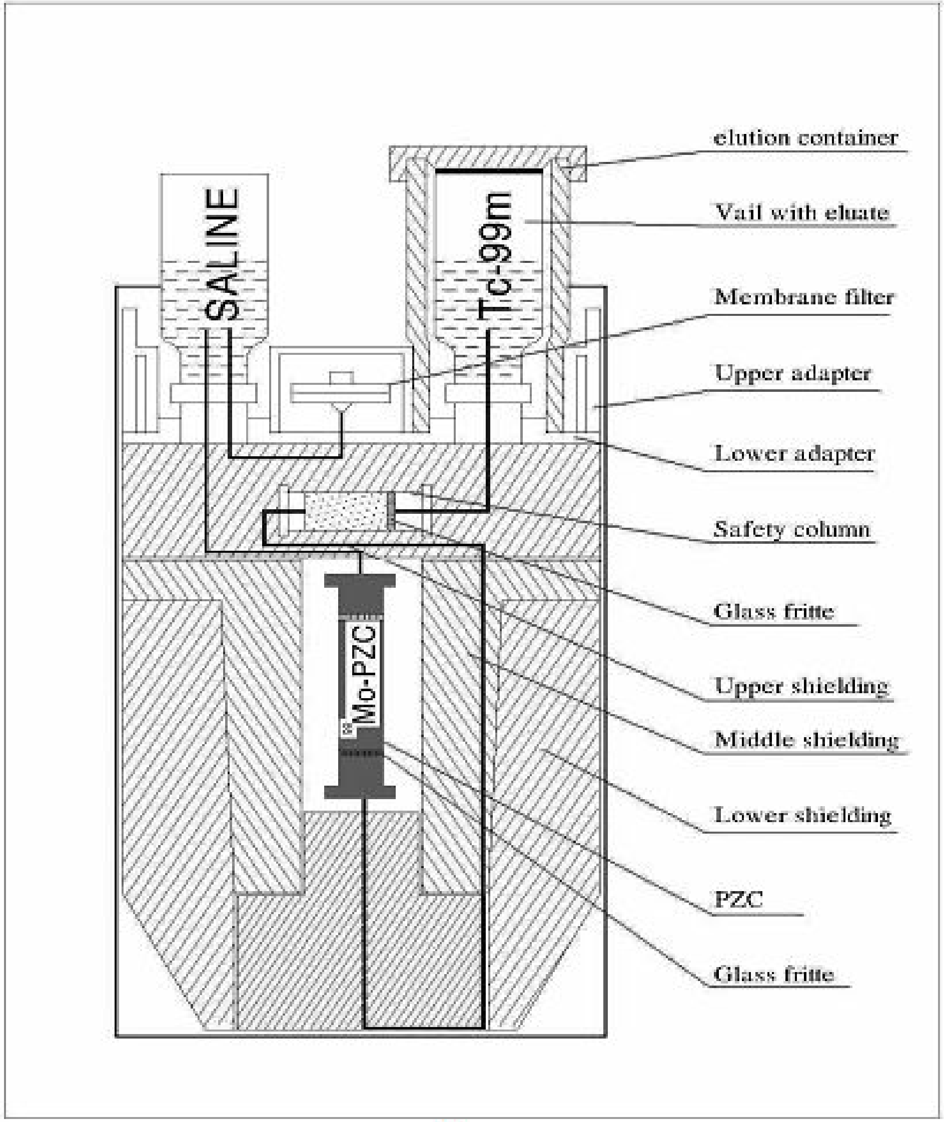
Alternative methods for 98Mo-produced generators

- Post-elution concentration techniques
  - ✓ Elution with Acetone instead of saline solution
  - ✓ Tandem-type generator
- Zirconium-based adsorbent instead of alumina column
  - ✓ Zr gel-based generators
  - ✓ Poly Zirconium Compound (PZC)-based generators

Advanced Accelerator Applications

The ^{99}Mo-^{99m}Tc PZC generator

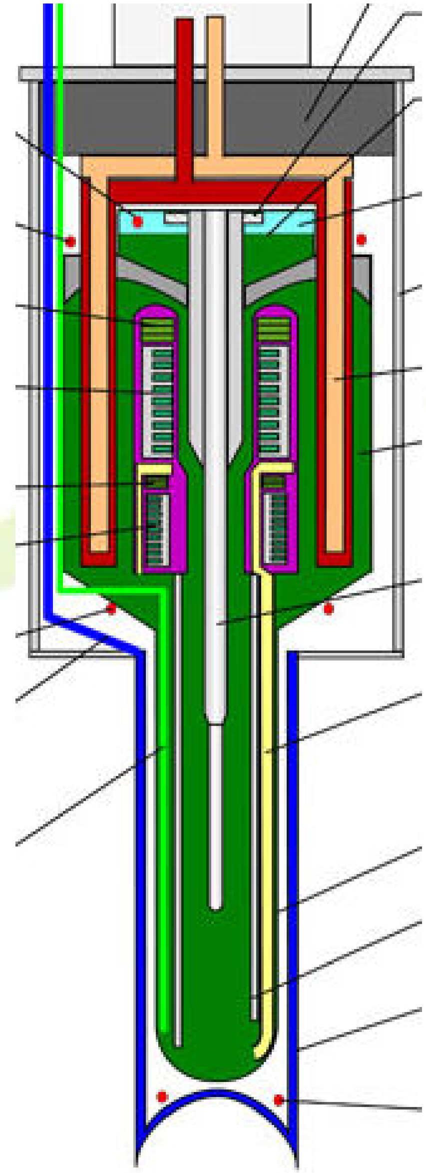
- First proposed in 1994 (JAERI-KAKEN) and extensively tested and optimised by the FNCA (Forum for Nuclear Cooperation in Asia) research group in 2003-2006
- Capable of trapping up to 250 mg of Mo per g of PZC (more than 10 times higher of Al columns)
- Pre-clinical and clinical tests results reported by FNCA partners are in compliance with European pharmacopeia



Advanced Accelerator Applications

Feasibility of a 1-4 MW liquid-metal target: MEGAPIE

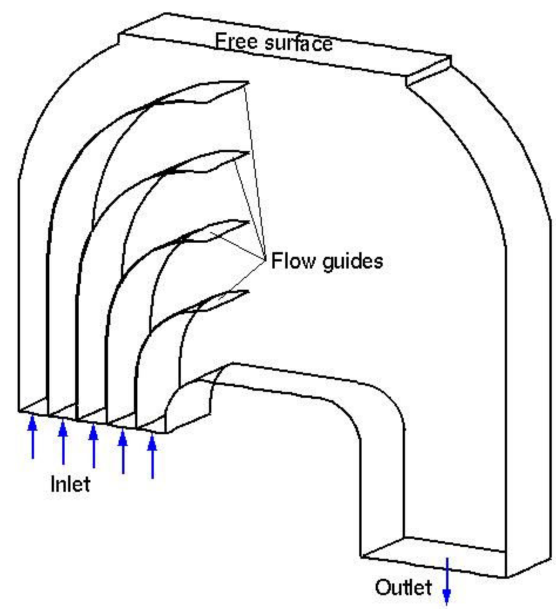
- **Designed to be coupled with a 590 MeV - 2 mA cyclotron (PSI-SINQ)**
- **(Dual) Window-type Lead-Bismuth target (D~20 cm, L~5 m)**
- **Electromagnetic pumps**
- **The MEGAPIE target has been completed and routinely tested in 2006 in PSI at 590 MeV and 1.3 mA.**
- **AAA people have participated to the design and the engineering of the target**

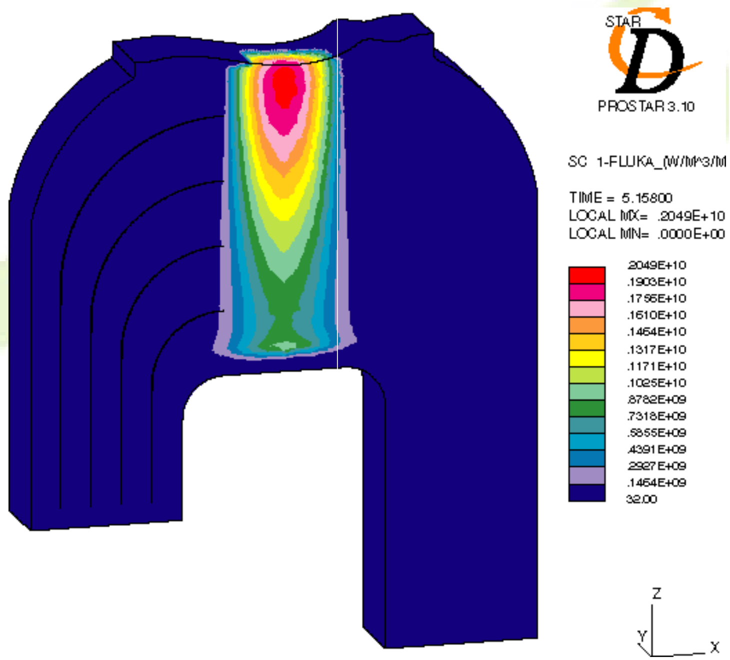


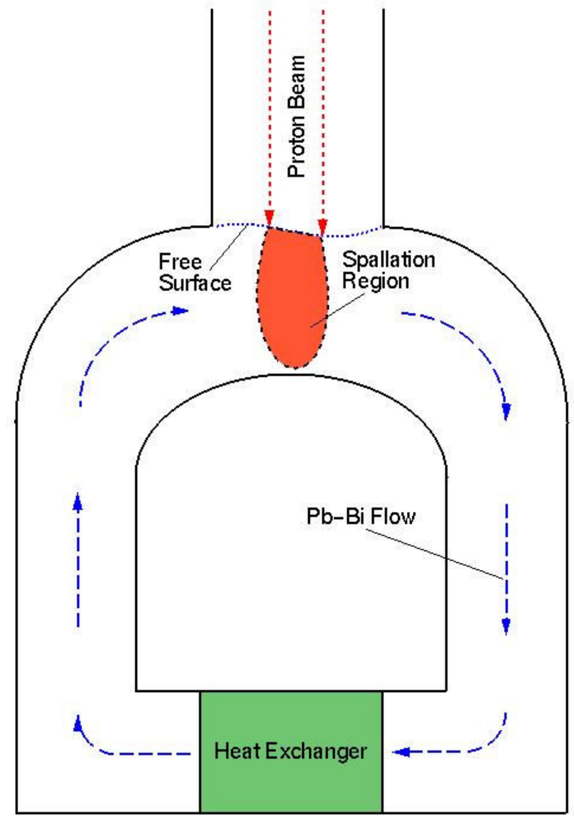
Advanced Accelerator Applications

Feasibility of a 1-4 MW liquid-metal target: ENERGY AMPLIFIER Windowless Target

- Designed to be coupled with a **600 MeV - 6 mA cyclotron**
- **Windowless-type Lead-Bismuth target (D~30 cm)**
- **Simulated with AAA in-house moving-mesh free-surface algorithm (the only existing complete CFD simulation of a windowless target**





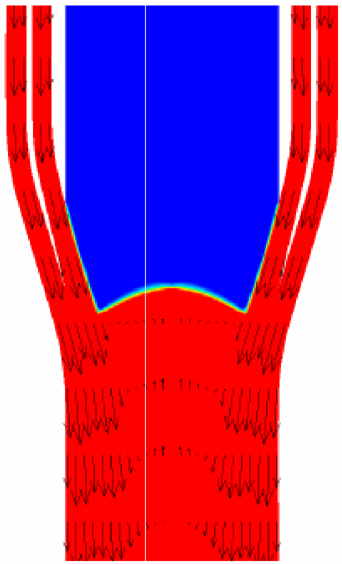


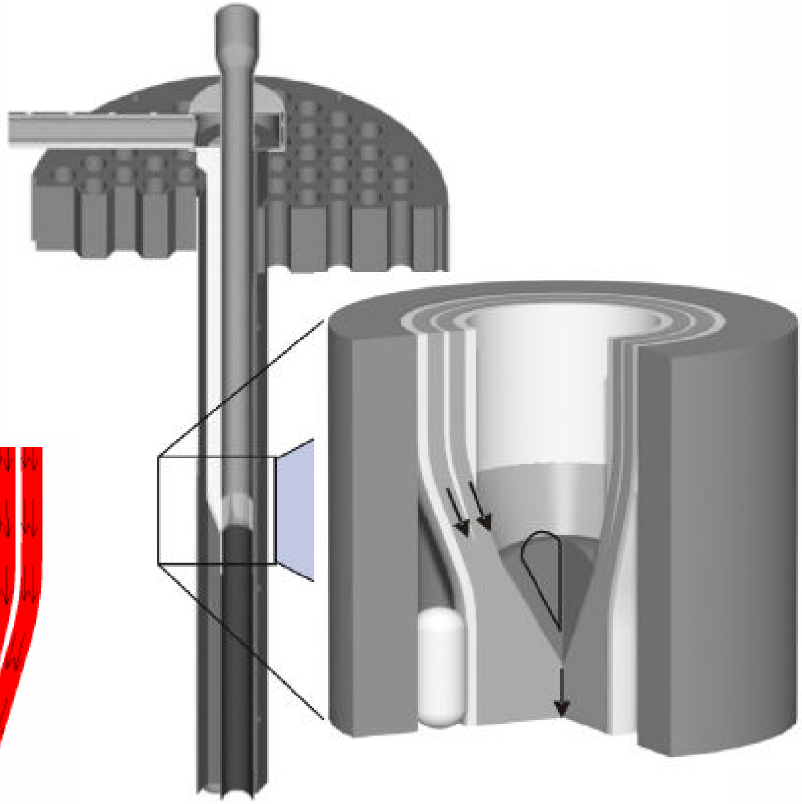
Temperature field

Advanced Accelerator Applications

Feasibility of a 1-4 MW liquid-metal target: EUROTRANS Windowless Target

- Designed to be coupled with a 600 MeV - 6 mA cyclotron
- Windowless-type Lead-Bismuth target (D~15 cm))
- Needs a swept beam to avoid heat deposit in the central recirculation region
- Design still in progress

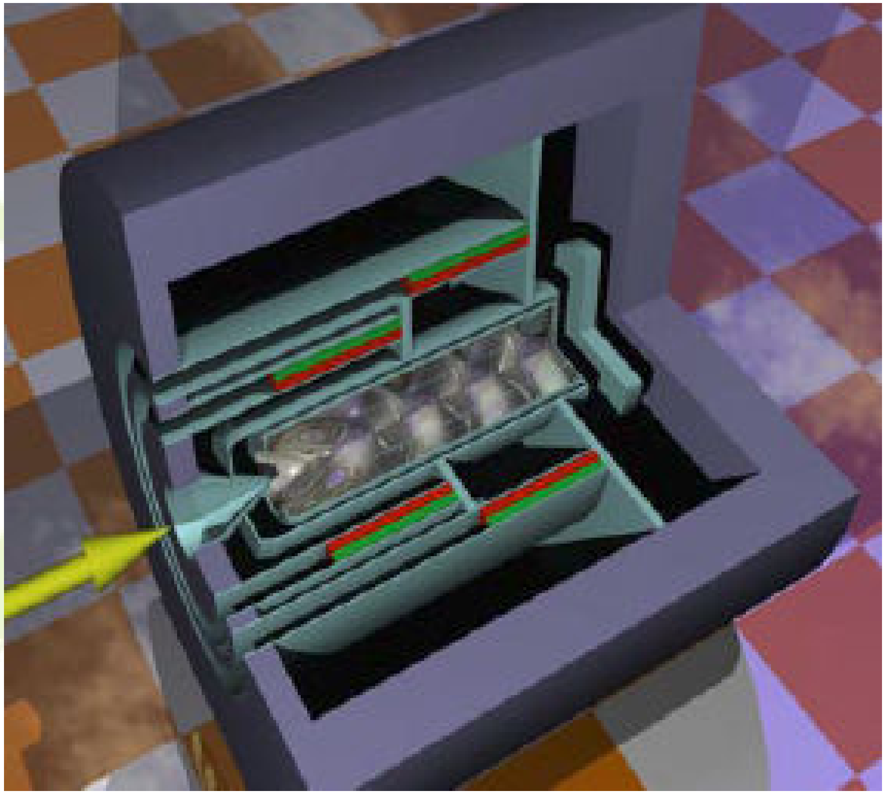




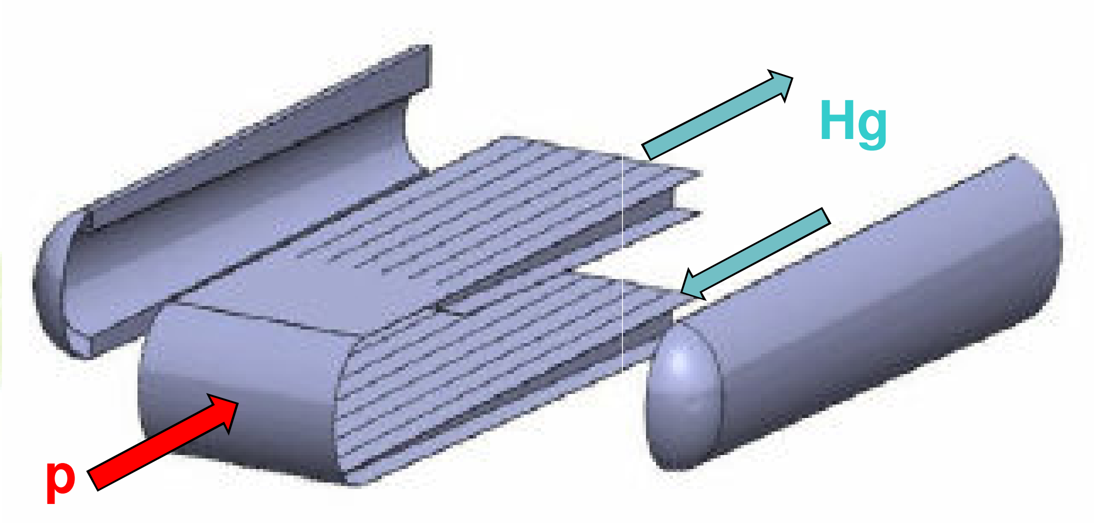
Advanced Accelerator Applications

Feasibility of a 1-4 MW liquid-metal target: EURISOL

- Designed to be coupled with a 1 GeV – 4 mA accelerator
- Window-type (D~10 cm) (also windowless considered)
- Hg or Lead-Bismuth as target material
- According to available results, preliminary design with a Hg window-type target at 1 GeV and 4 mA showed thermal and mechanical feasibility



Feasibility of a 1-4 MW liquid-metal target: EUROPEAN SPALLATION SOURCE



- Designed to be coupled with a 1/1.3 GeV – 5 mA pulsed LINAC
- Window-type (D~10 cm) Hg proton target
- Current design status to be assessed

CardioGen-82

- Contains Sr-82 in a lead shielded column
  - Sr-82 half life is 25 days
- The “daughter” is Rb-82 chloride
  - half life 75 sec
- Rb-82 kinetics:
  - After iv injection, Rb-82 rapidly clears the blood
  - Extracted similar to potassium
  - Activity in myocardium within first minute
  - Defects visualized 2-7 minutes after injection
  - Uptake is also observed in the kidney, liver, spleen, and lung
- Must be used with an infusion system
- Same delivered dose for rest and stress imaging
- On demand availability

CardioGen-82® package insert

Attenuation Corrected Tc-99m Sestamibi SPECT Compared with Rb-82 Myocardial Perfusion PET

The Results:

	PET	SPECT with AC
Overall Accuracy	90%	80%
Left anterior descending artery (LAD)	90%	76%
Left circumflex artery (LCx)	83%	78%
Right Coronary Artery (RCA)	90%	77%
All Coronaries	88%	77%
Image Quality (Excellent)	80%	24%
No Artifacts	90%	50%

J Nucl Cardiol July/August 2005 Abstracts
Abstract 34.22

Example CardioGen-82 PET/CT Protocol[9]

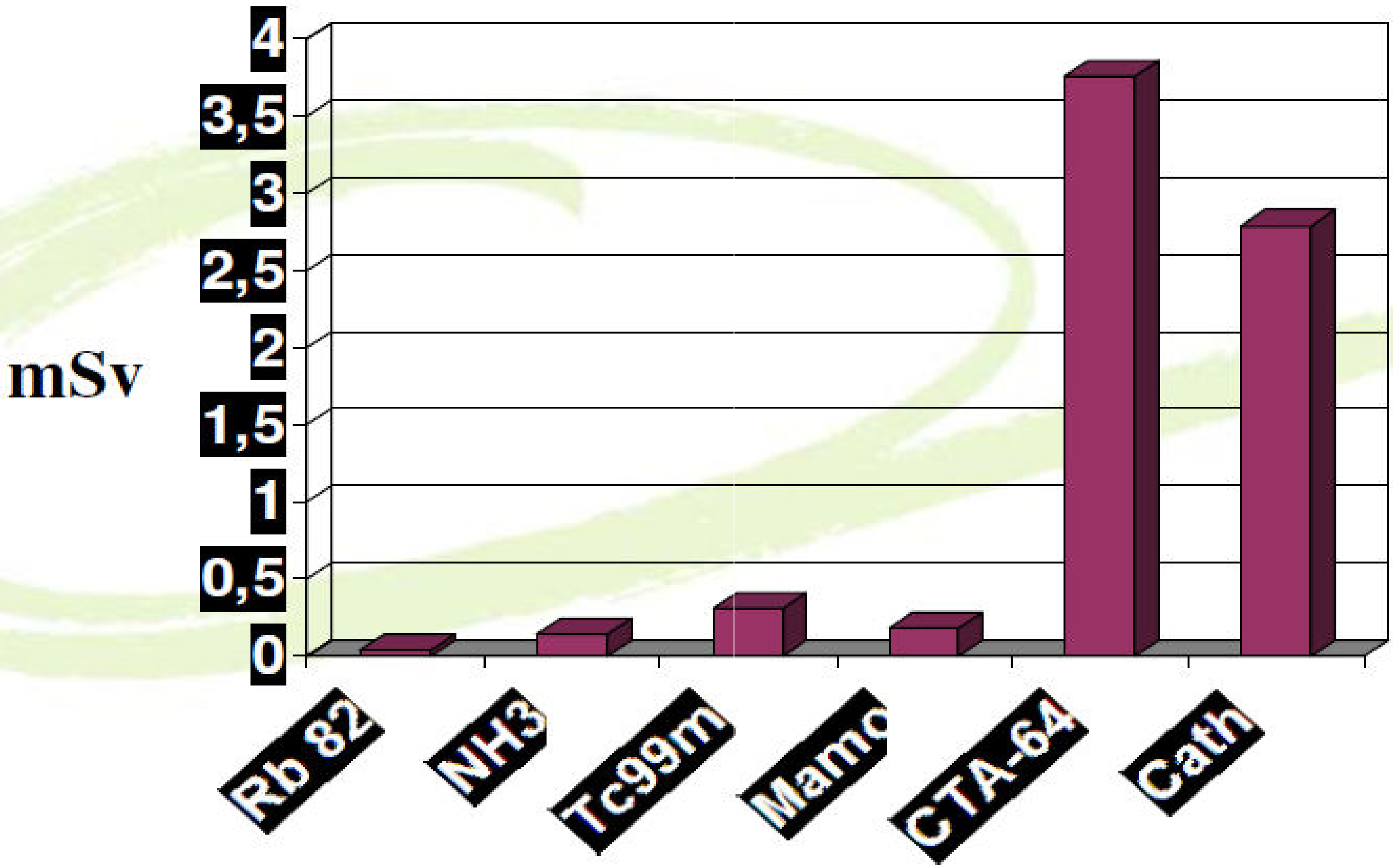
Approximately 35 minutes

Scout	Trans	90-120 sec	Gated Rest	Dipyridamole Adenosine	90-120 sec	Gated Stress	Trans	CAC	CTA
		Rb 82 (40 – 60 mCi) ↓			Rb 82 (40 – 60 mCi) ↓				
5 sec	10 sec		5 minutes	4-6 minutes		5 minutes	10 sec	10 sec	20 sec

9. Di Carli MF. Major achievements in nuclear cardiology:XI. Advances in positron emission tomography. J Nucl Cardiol. 2004;11:719-732.

A gated SPECT using Tc-99 products requires 2 steps of 1 hour each (in 2 different days for better dosimetry)

Breast and skin dose from noninvasive imaging studies



Castronovo et al. In Cardiac PET & PET/CT Imaging. Di Carli/Lipton. 2007