Italian National Agency for New Technologies, Energy and Sustainable Economic Development

ENEA Projects for Medical Isotope Production and Noble Gas Measurements at the ENEA Nuclear Research Reactors, Italy

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Overview

- **ENEA projects for medical isotope production (^{99}Mo)**:
  - SORGENTINA RF project - fusion based device
  - MOLY project – neutron activation of ^{98}Mo at the TRIGA RC-1 research reactor

- **Preliminary considerations for radioxenon measurements at ENEA**:
  - measurements of radioxenon emissions at the ENEA nuclear research reactors TRIGA and TAPIRO
  - current status and planned improvements of the ENEA system for noble gas measurements

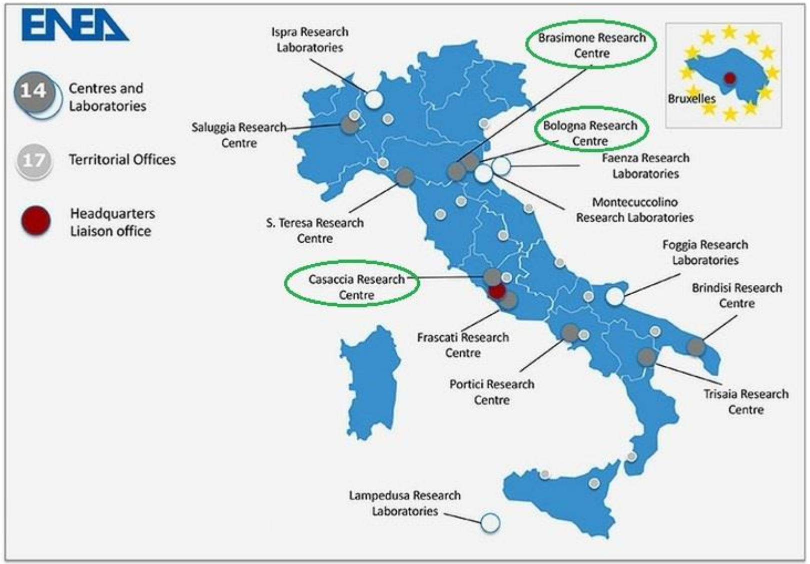
ENEA projects for medical isotope production (^{99}Mo)

Recent studies have been conducted at ENEA (Italy) for two different projects for contributing to the supply chain of medical isotopes.

1) The **SORGENTINA** project(*) was already presented at the WOSMIP 2021:

   a fusion-based device will be located at the ENEA Brasimone Research Centre (BRC), north-centre of Italy, about 60 km from Bologna.

   (*)Agostini, P. et al. SORGENTINA-RF project: fusion neutrons for ^{99}Mo medical radioisotope. Eur. Phys. J. Plus 136, 1140 (2021)



2) The **MOLY** project(**) is a feasibility study to produce ^{99m}Tc from neutron activation using the TRIGA RC-1 research reactor at the ENEA Casaccia Research Centre (Rome).

   (**)Aronica, O. et al. Il Progetto MOLY: una via italiana per la produzione del radiofarmaco ^{99m}Tc, Energia, ambiente e innovazione, 3/2017, DOI 10.12910/EAI2017-050

ENEA Sorgentina Project: a fusion-based device

- Scope: design and development of a prototypical medium intensity D-T 14-MeV fusion neutron source mostly dedicated to the production of medical radioisotopes with a special focus on ^{99}Mo.
- It will be a prototype plant to assess the production route.
- The production of molybdenum-99 was already verified on laboratory scale.
- Design and development of a medium power 14 MeV fusion neutron source relying on a rotating target and a deuterium-tritium ion accelerator:

$$^{2}H + ^{3}H \rightarrow ^{4}He + n + 17.1\ MeV$$

- Neutron irradiation of about 10 kg of natural molybdenum:

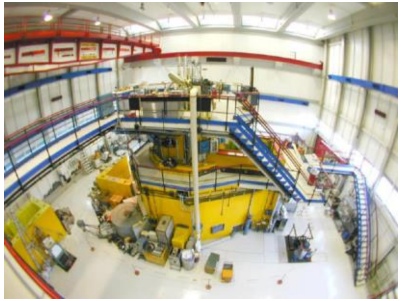
$$^{100}Mo(n,2n)^{99}Mo$$


ENEA Brasimone Research Centre

ENEA MOLY Project at the TRIGA research reactor

The **MOLY** project is a feasibility study to produce ^{99m}Tc from neutron activation using the TRIGA RC-1 research reactor (1 MW TRIGA Mark II) at ENEA Casaccia (Rome).

- Neutron activation of ^{98}Mo: **$^{98}Mo(n,\gamma)^{99}Mo$**
- Technical-financial feasibility and sustainability of the ^{99m}Tc production for local/national provision.
- The project is based on an agreement between ENEA and Perma-Fix Environmental Services which provides for the production of Technetium-99m by exploiting the potential of the TRIGA RC-1 Casaccia research reactor and a process developed by Perma-Fix Medical.
- The ^{99}Mo-^{99m}Tc generators will be supplied to nuclear medicine centres and would ensure some local provision to cover a large part of the national needs.



ENEA MOLY Project

- ENEA will manage all stages of the production process, from the irradiation and neutron activation of Molybdenum to the construction of the generator. However, maintenance and renovations works are needed to adapt the laboratories to the standards of GMP ("Good Manufacturing Practice“).
- The goal is to create a group of high-level competences for the production of radiopharmaceuticals finalizing national and international agreements with public and private entities, also involving national stakeholders in the radiopharmaceutical field.

ENEA MOLY Project:
Preliminary evaluations of ^{99}Mo production

- A preliminary evaluation was performed to estimate the ^{99}Mo production assuming the irradiation of a molybdenum target (98.4% enriched in ^{98}Mo) in the reactor central channel with different irradiations modes:
  - 120 consecutive hours → 30 GBq/g
  - 6 hours/day, 5 days/week → 9 GBq/g
- Such values were confirmed by preliminary experimental results by scaling the results obtained on samples with shorter irradiation times and using molybdenum with natural isotopic.
- As soon as molybdenum targets enriched in ^{98}Mo will be available, the tests will be repeated.

Radioxenon Emissions from Nuclear Research Reactors

M.B. Kalinowski et al., Global Radioxenon Emission Inventory from Nuclear Research Reactors, Pure Appl. Geophys. 178 (2021), 2711–2739:

"The contribution of NPPs and MIPFs to the global radioxenon emission inventory is fairly well understood. NRRs have yet to be systematically assessed. … This source type needs to be taken into consideration and further studies are required to get a better understanding of the following questions:

- *What mechanisms of radioxenon formation and release can explain the variability of releases from NRRs and what are the maximum release amounts on 1 day that can be expected?*
- *Identify which individual NRRs may have an impact on which specific regional IMS stations."*

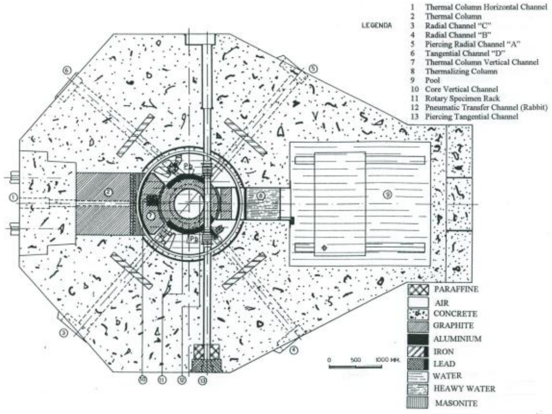
I. Hoffman et al., Medical isotope production, research reactors and their contribution to the global xenon background, Journal of Radioanalytical and Nuclear Chemistry (2018) 318:165–173:

"Although medical isotope production dominates the global radioxenon background, analysis of the emissions of the underlying research reactors show that they are also significant contributors to the radioxenon background and may become increasing important in CTBT monitoring if emissions from medical IPF decline."

Measurements of radioxenon emissions will be planned at the ENEA nuclear research reactors TRIGA and TAPIRO to contribute to the assessment of the emission inventory of NRRs and to the evaluation of noble gas background in Italy.

TRIGA Reactor: General Atomic Mark II

TRIGA	
Power	1 MW (1st start-up at 100 kW in 1960, upgrade 1 MW in 1967)
Fuel	Uranium – ZrH alloy
Enrichment	20 % 235U
Moderator	ZrH, H2O
Coolant	Demineralized water in natural Coolant convection
Reflector	Graphite
Max Neutron Flux (central channel)	~ $3*10^{13}$ neutrons cm^{-2} s^{-1}

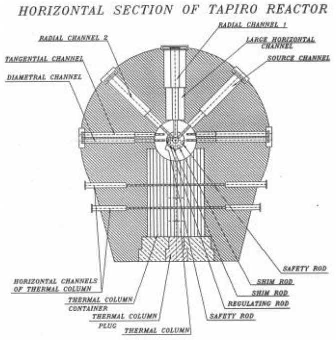


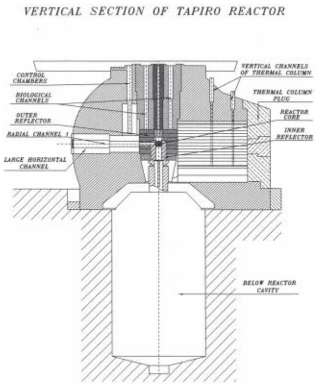
TAPIRO: Fast Neutron Research Reactor

"TAratura PIla Rapida potenza zerO (Fast Pile Calibration at 0 Power)"

- Fast source reactor, based on the concept of AFSR (Argonne Fast Source Reactor - Idaho Falls)
- Start-up: 1971

TAPIRO	
Power	5 kW
Fuel	Uranium-molybdenum alloy
Enrichment	93.5% U235
Cladding	Stainless steel
Coolant	He
Reflector	Copper
Max Neutron Flux (diametral channel)	~ $3*10^{12}$ neutrons cm^{-2} s^{-1}
Biological shield	near spherical, high density concrete





ENEA research reactors: Main fields of applications

- Test of innovative neutron detectors
- Neutron damage (Gen IV)
- Electronic hardness qualification (aerospace and accelerators)
- Neutron propagation (Gen IV)
- Radiation protection dosimetry
- Boron Neutron Capture Therapy
- Forensic application (INAA)
- Neutron cross section measurement (minor actinide management)
- Education and training

Radioxenon measurements at ENEA:

Preliminary Sampling Strategy at the ENEA research reactors

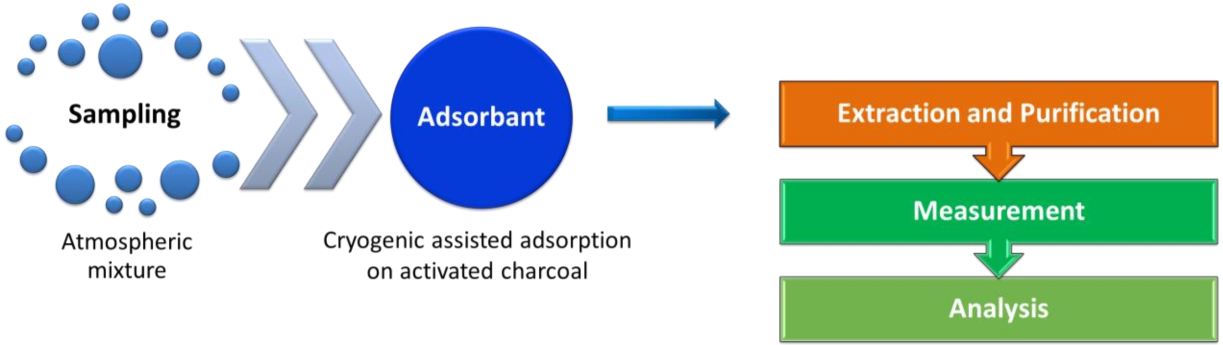
NRRs are frequently shutdown and restarted due to many reasons related to the conducted experiments or the operational working schedules, with possible puff emissions depending on the specific activity that is conducted.

- A few representative measurements will be planned within the reactor facilities and then converted to a release rate, extrapolating to the emission for a whole year.
- The experimental schedule will include:
  - background samples (after some days without reactor operation)
  - and samples during normal reactor operations.
- Air samples will be collected at different locations around the reactors at some distances from the reactor core, in the reactor hall and on top of the reactor pool (TRIGA).
- Some measurements at the stack will be also considered.
- Samples will be collected at different reactor modes:
  - reactor off
  - and normal operations with the reactor at full power.

ENEA system for noble gas measurements:
Current status and planned improvements

The system is located at the ENEA Brasimone center (Bologna, Italy), in the north-centre part of Italy by the Brasimone lake, at about 850 m altitude.

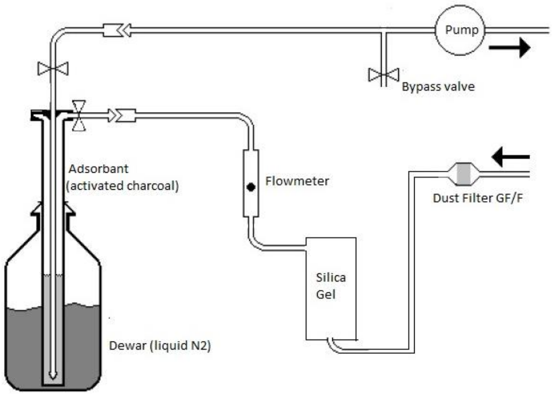
It consists of three separate units: air collection (sampling and adsorption), sample processing (gas extraction and purification) and measurement (gamma spectrometry):



ENEA system for noble gas measurements:
Sampling equipment

The sampling equipment (based on cryogenic adsorption on activated charcoal) was constructed along the lines of a similar system developed at the Freiburg laboratory (BfS, Germany) - *E.Nava et al., The ENEA noble gas laboratory: status of implementation, J Radioanal Nucl Chem (2013) 296:1163–1167*

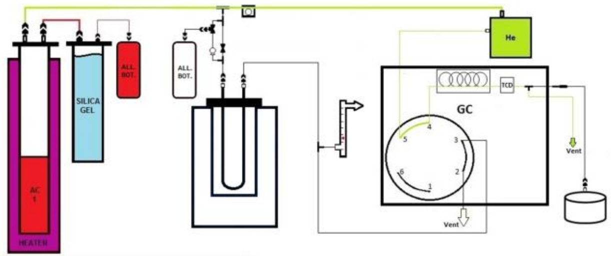
Sampling scheme



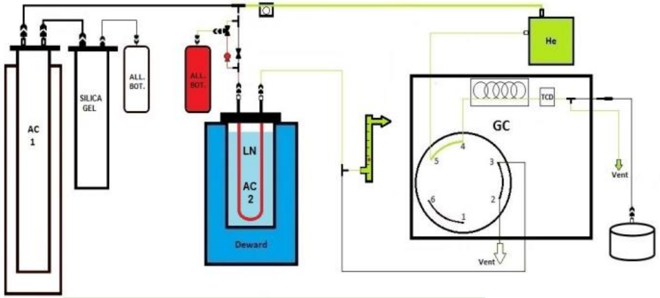
Sampling set-up

Adsorbant cartridge

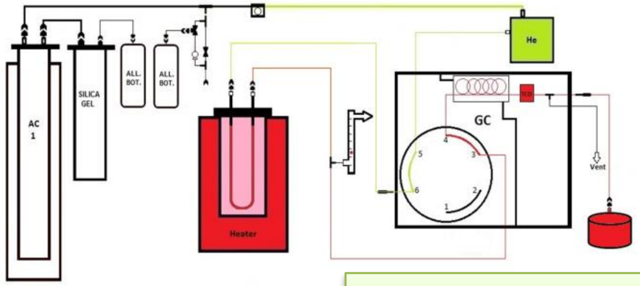
ENEA system for noble gas measurements:
Sample Processing



1. Extraction



2. Concentration



3. Separation

Gas chromatograph
Agilent 7890B

ENEA system for noble gas measurements: **Sample Measurement**

- High-purity germanium, coaxial detector p-type 'Extended Range' with an end-cap made of carbon fibre
- Low-background shield
- CANBERRA cryo-cycle cryostat

Detector Canberra GX6020

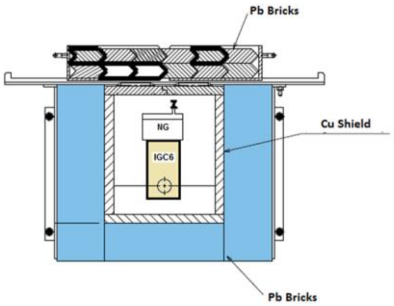
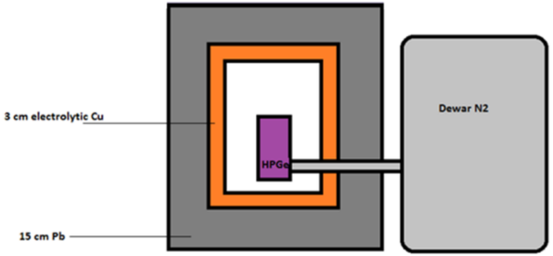
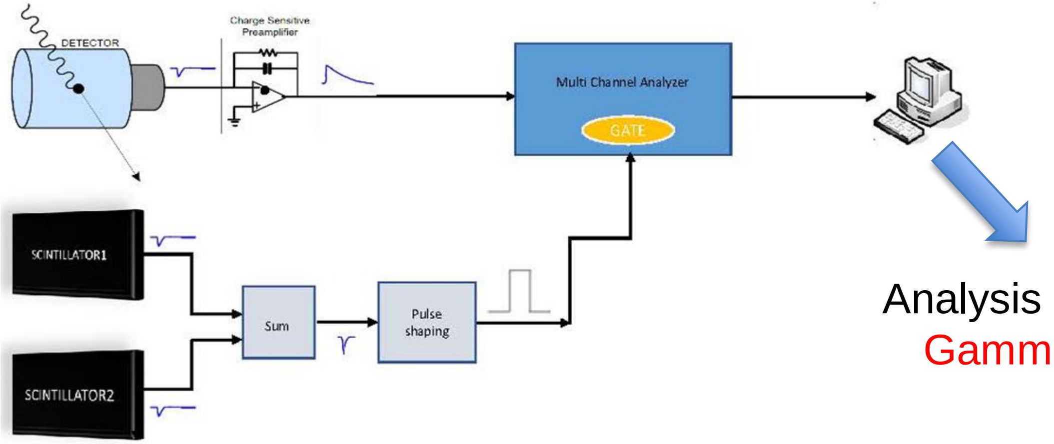


FIG.3



ENEA system for noble gas measurements:
Sample Measurement – Anticoincidence System



Analysis Software:
GammaVision

Two NUVIA plastic scintillators were placed above the shielding.

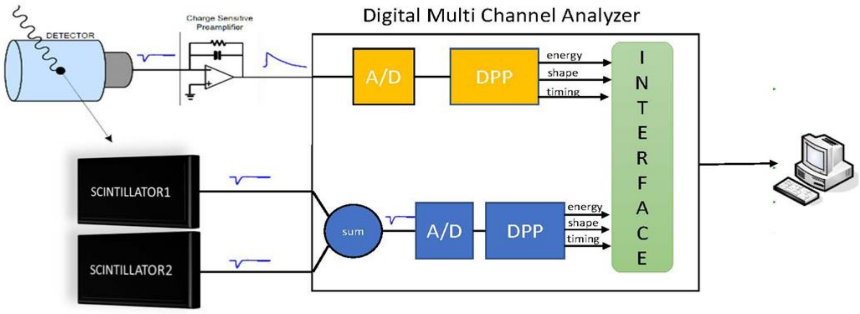
The anticoincidence system developed at the ENEA laboratory is able to implement the anticoincidence mode, with improved sensitivity and detection limits for the four xenon radioisotopes.

ENEA system for noble gas measurements:
Sample Measurement – Further improvements

The analog–digital system has been replaced with a fully digitized Multi Channel Analyzer:

CAEN Tools for Discovery
Electronic Instrumentation

→ **Digital Multi Channel Analyzer**



ENEA system for noble gas measurements:
Plans for improvements

The ENEA noble gas systems has been participating in the CTBTO Xe PTE intercomparison exercises since 2012.

Issues to be addressed and plans for improvements

- Sampling device:
  - at the moment only one adsorbent cartridge is available, multiple cartridges would be necessary for the measurement campaigns at the research reactors;
  - the sampling time is quite long (about 1-2 weeks), possible upgrade of the air pump will be considered
- Sample processing:
  - possible gas leakage during sample transfer to the cell measurement
- Measurement system:
  - further works should be conducted to better characterise the detector response to the low energy gamma peaks of radioxenon isotopes with the anticoincidence system active;
  - new design of the measurement cell along the lines of a system developed by Seibersdorf Laboratories (Austria)

Summary and way forward

- Some noble gas measurement campaigns will be scheduled at the ENEA nuclear research reactors for contributing to the noble gas background assessment.
- The ENEA noble gas system will be adapted and optimized for these measurements.
- Collaborations proposals for measurements with other systems will be welcomed.

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