National Center for Nuclear Research





Status of Molybdenum Program in National Centre for Nuclear Research

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MARIA RESEARCH REACTOR Molybdenum Program



- * NCBJ is willing to provide solution ensuring sustainable production and distribution Mo-99 in both <u>mid and long</u> <u>terms</u> based on irradiation HEU & LEU targets in MARIA RR and using the new MPF (Molybdenum Processing Facility)
- Molybdenum Program provided in NCBJ includes:
 - 1) Technology of irradiation targets in MRR: HEU/LEU-based,
 - 2) Molybdenum Processing Facility (MPF) located on NCBJ site: LEU-based production Mo-99 & Mo-99/Tc-99m generators;
 - 3) Participation on existing programs including "*Developing techniques for Molybdenum-99 production using neutron activation*" (collaboration with JAEA, MURR and IAEA)

BACKGROUND INFORMATION: ⁹⁹Mo Production



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- European production was based on HEU target irradiation on five reactors: HFR (Petten, NL), BR2 (Mol, B), MARIA (PL), LVR-15 (Rez, CR) andd OSIRIS (Sacley, FR);
- Reactors were operated as a supply network within Europe in conjunction with two Mo-99 processing facilities operated by Mallinckrodt (Petten, NL) and IRE (Fleurus, B);
- Supply elsewhere in the world is not-worked and exist as a number of single supply lines of geographically isolated reactor processing facilities (Canada, South Africa and Australia, Argentina);

• single break in the supply chain completely stops Mo-99/Tc-99m supply

• With the ageing of the European supply network it is necessary to consider new reliable supply sources.

BACKGROUND INFORMATION: ⁹⁹Mo Irradiators World Net

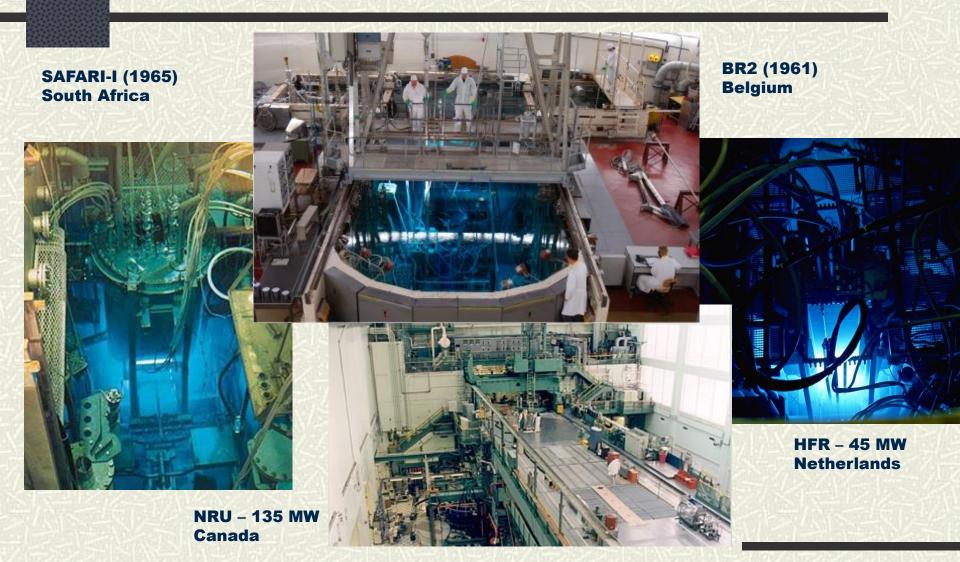


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Reactor	Country	Operating days	Forecasted date of operation			
HFR	Netherlands	280	2022			
BR-2	Belgium	140	2022			
MARIA	Poland	180	2030			
OSIRIS	France	200	2015			
LVR-15	Czech Republic	200	2029			
NRU	Canada	300	2016			
OPAL	Australia	290	2015			
SAFARI-1	South Africa	300	2027			
RA-3	Argentina	330	2022			

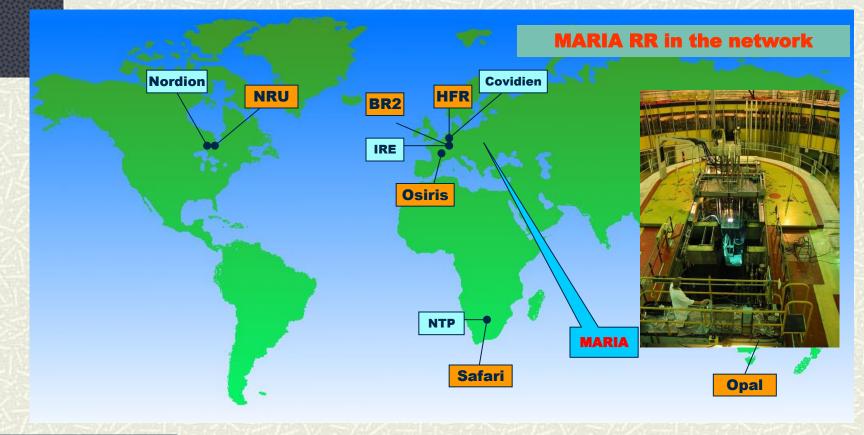
Radioisotope production Mo-99 Main irradiation facilities

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Reactors and Mo-99 Processing Facilities around the world





RR in operation

- BR2, Belgian Nuclear Radiopharmacy Centre, Belgium
- OSIRIS, France, French Atomic Energy Commission (CEA)
- NRU, National Research Universal Reactor, Canada
- HFR, Nuclear Research & Consultancy Group (NRG), The Netherlands
- SAFARI-1, South African Nuclear Energy Corporation (NESCA), S.Africa
- OPAL ANSTO, Australia

- **Mo-99 Processing Facilities**
- COVIDIEN, USA / The Netherlands
- MDS Nordion, Canada
- Nuclear Technology Products (NTP), S. Africa
- The Institute for Radio Elements (IRE), Belgium

MARIA RESEARCH REACTOR Molybdenum Program



• General assumption: production rate of ⁹⁹Mo should be proportional to fission power generated from uranium targets in irradiation rig:

A – ⁹⁹Mo activity, P – fission power, λ - decay constant ⁹⁹Mo, γ = 0.0611 – production efficiency of ⁹⁹Mo per one fission act; E_f = 3.244 · 10⁻¹¹ J – generated energy per one fission act:

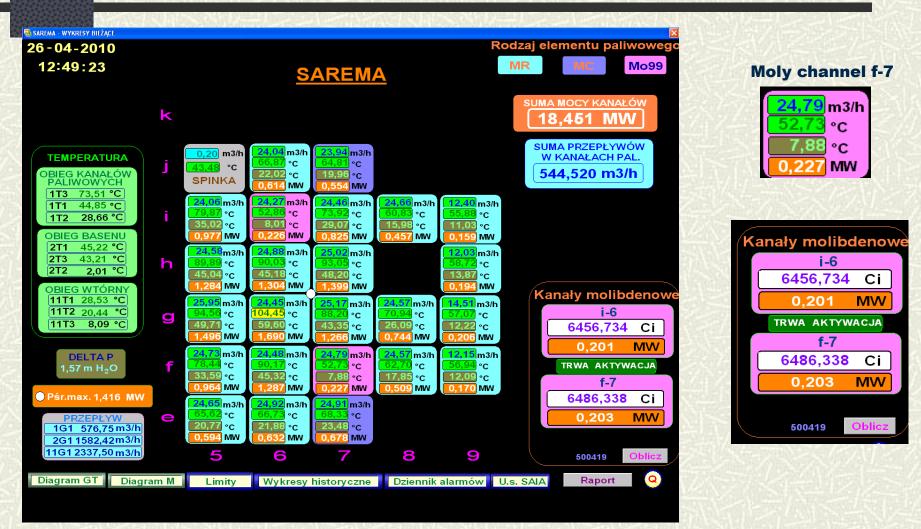
$$\frac{dA}{dt} = \alpha P - \lambda A \qquad \alpha = \gamma \lambda / E_{f}$$
(1)

- Alfa constant based on physical properties and ORIGEN code calculation: $\alpha = 5.47 \text{E3}[\text{Bq/Ws}]$ (2)
- Integrating equation (1) within the irradiation time (0,t) we receive a relation of activity of ⁹⁹Mo to be formed from the fission power generation in the plate.

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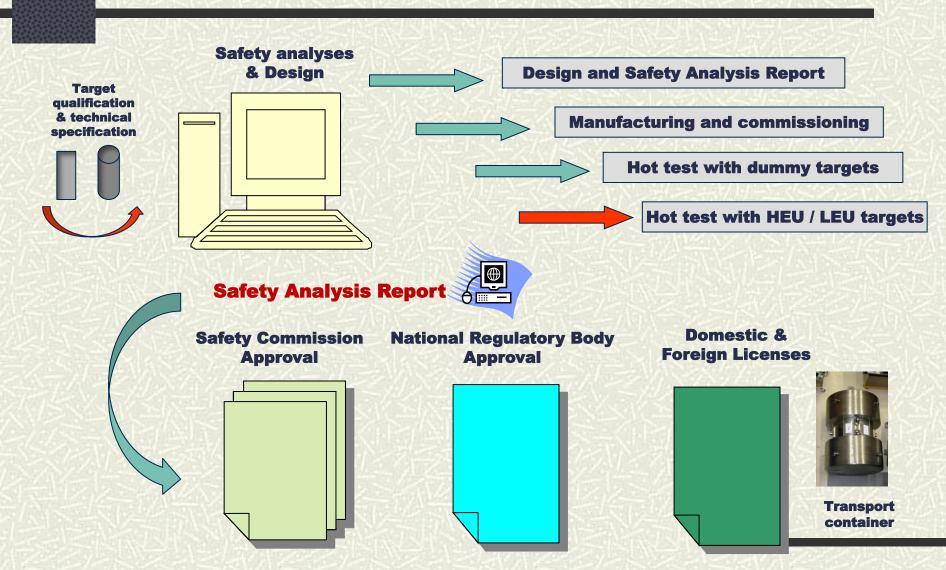


Screen of SAREMA system – reactor MARIA

MARIA RESEARCH REACTOR The Safety Analysis, Test & Manufacturing for Certification of new HEU targets

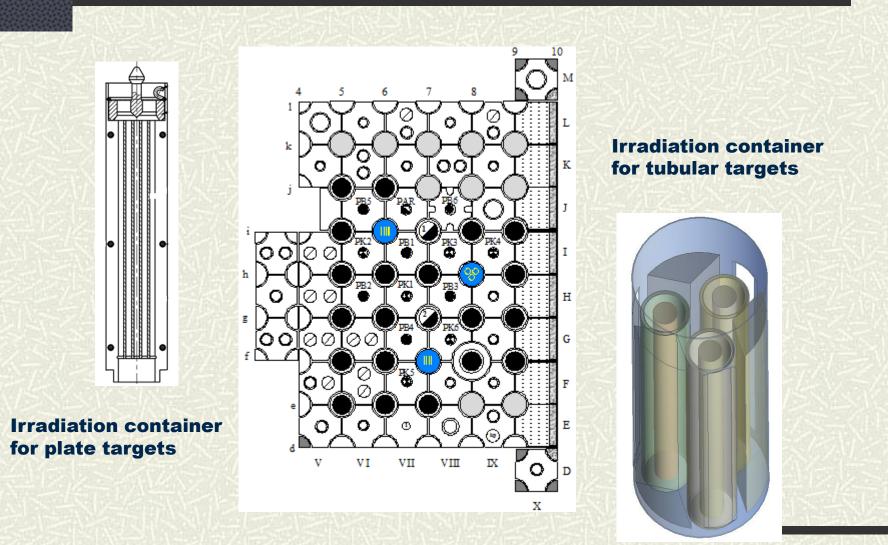


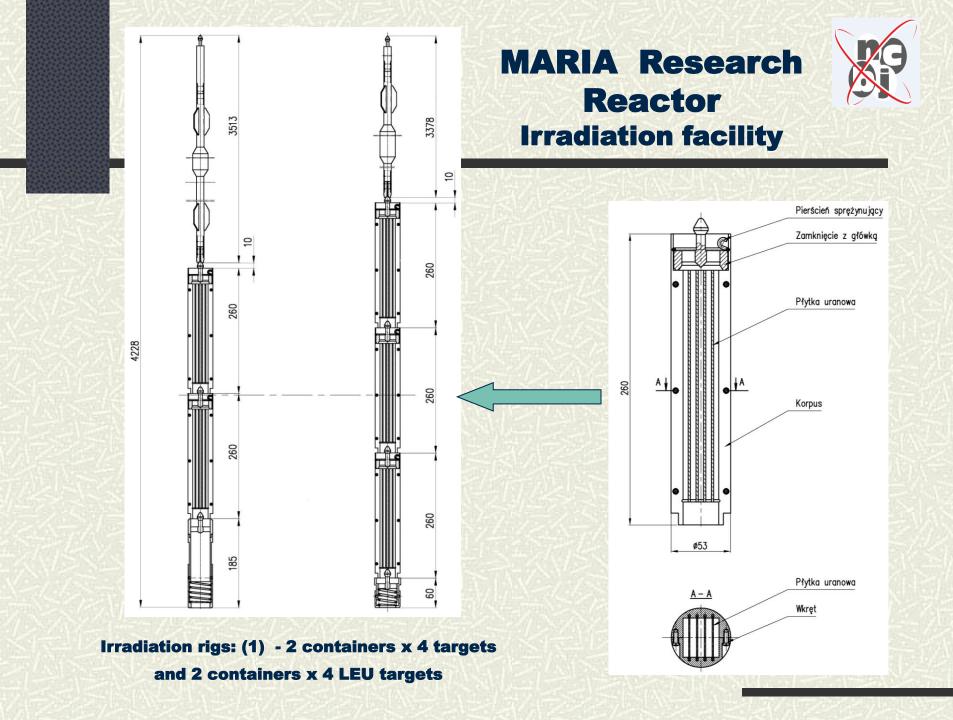
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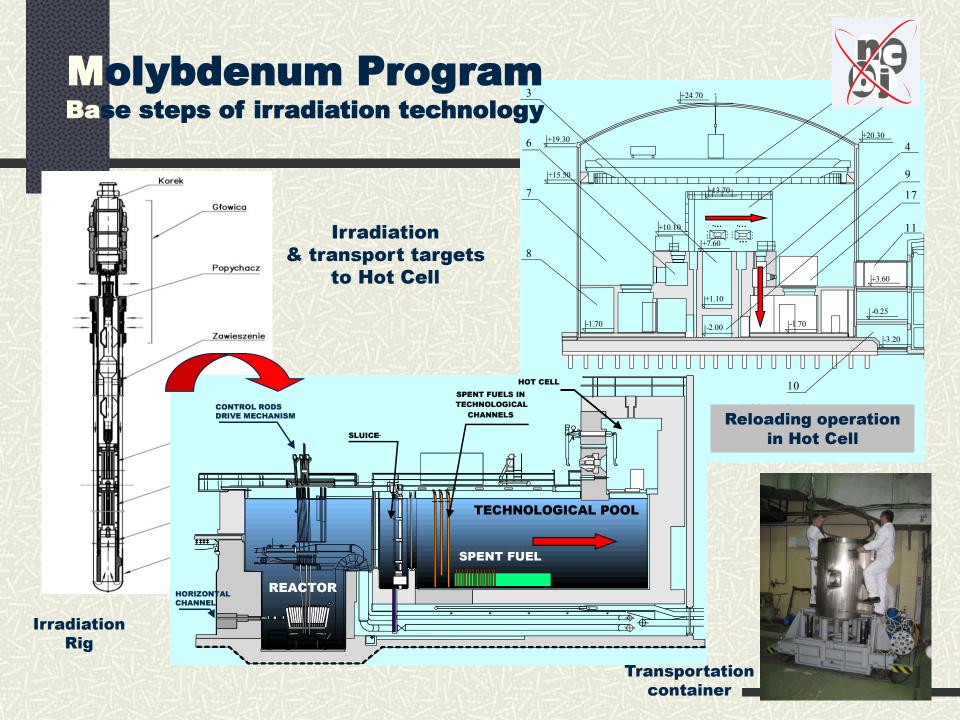


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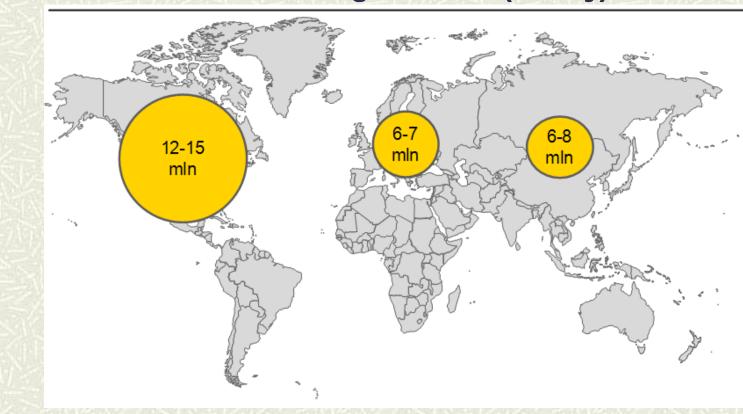


NARIA RESEARCH REACTOR Molybdenum Program



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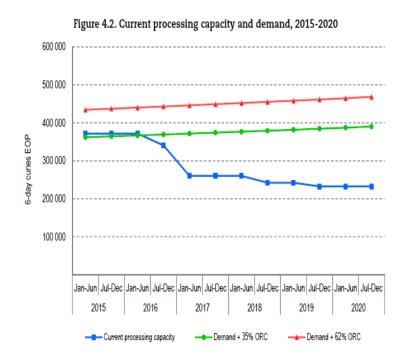
Number of medical procedure around the world based on ⁹⁹Mo/ ^{99m}Tc generators (2009 y)

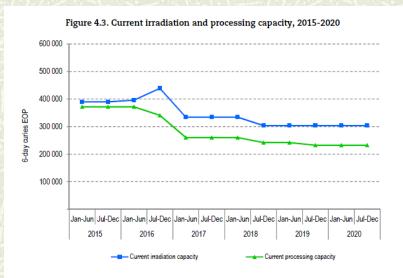


Source: Prelminary report on supply of radioisotopes for medical use and current developments in nuclear medicine, EC 2009

BACKGROUND INFORMATION: ⁹⁹Mo Production







BACKGROUND INFORMATION: ⁹⁹Mo Production



Processing facilities should be located close to the reactors:

- the primary reasons for this is minimize the transport of high activity fissile nuclear materials;
- reduce as much as possible the decay loss of Mo-99 (the quantity of Mo-99 produced decay by 1% per hour from the EOB);
- the processing site should be licensed for waste management.
- The main European Mo-99 producers: COVIDIEN and IRE plants are located at reactor site or on reasonable distance:
 - COVIDIEN plant is located at the HFR site; PALLAS location will be same
 - IRE plant is located close to BR-2; MYRRAHA location will be same,
 - Plans for new plant be located in Munich (TUM site next to FRM II) and / or Cadarache (CEA site next to JHR).

"Molibden – Świerk" Project

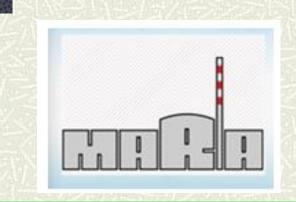


Project defined as follows:

- Design and construction of the two parallel technological lines for Mo-99 production with LEU targets (UAI_xAI) irradiated in reactor MARIA (2x500 Ci /w 6-days Mo-99) will be placed in a new building near reactor;
- Establishing of the new line for Mo-99/Tc-99m generators manufacturing, which will be placed inside the POLATOM building (capacity of 500 units /week)









Uranium targets irradiation HEU/LEU COVIDIEN Mo-99 Mo-99

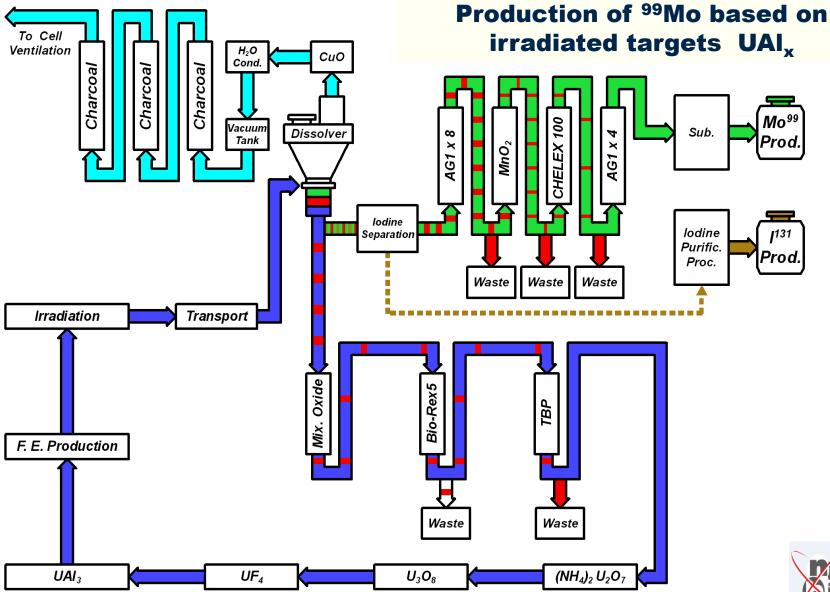




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Target Specification: "MOLIBDEN 2010" Program

target specification		HEU	LEU	LEU	LEU
target core material type of target		UAIx-AI plate	U₃Si₂-AI plate	UAIX-AI plate	UO ₂ -AI cylinder
target dimension core meat dimension	mm mm	203 x 40 x 1.45 191 x 27.5 x 0.85	203 x 40 x 1.35 186 x 29.5 x 0.75	203 x 40 x 2.00 186 x 29.5 x 1.4	Ø _{out} = 25.0 Ø _{in} = 24.4
m eat thickness enrichment	mm %	0.85 93	0.75 19.75	1.4 19.75	0.6 19.75
U density U _{total} loading	gU⊤/cm³ g	1.1 5.1	4.8 23.8	2.6 11.7	3.8 11.7
²³⁵ U loading	g	4.7	4.5	2.3	5.3
amount of Al in the core amount of Al in the target	g g	11.6 20.3	6.9 20.3	10.4 20.3	bd. bd.
number of targets (batch) total mass of ²³⁵ U (batch)	g	8 37.6	8 36.0	8 31.6	6 37.6

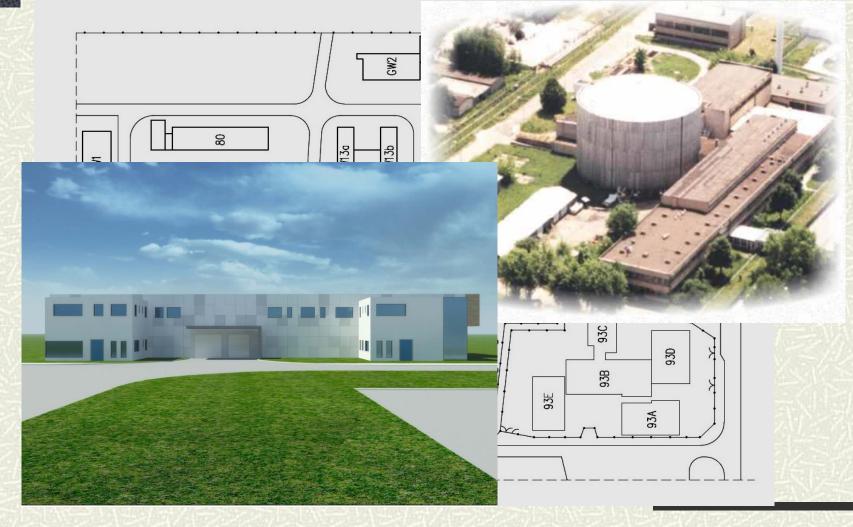




Location of the building for ⁹⁹Mo and ^{99m}Tc generators production

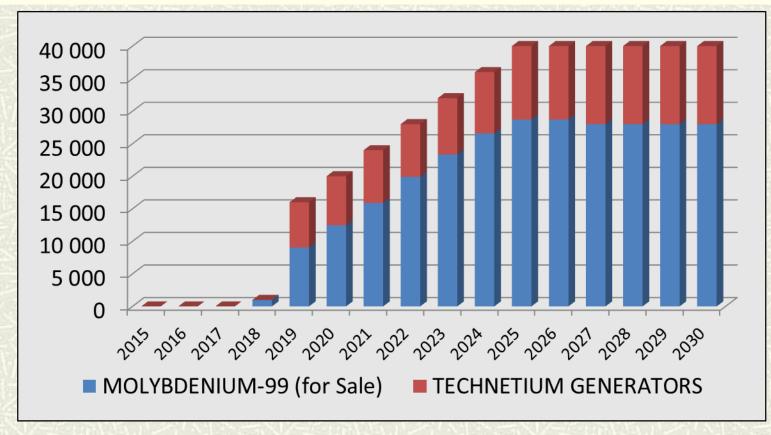


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"Molibden – Świerk" Project Financial Model

Chart 1: Production Output: Technetium Generators and Mo-99 (Ci Mo)



Source: Business Plan by NCBJ



Determining the activity of ⁹⁹Mo in irradiated uranium targets



- The measurements of temperatures and flow rate of coolant in reactor fuel channels are conducted by standard reactor measuring system SAREMA;
- Using the quantities measured the total reactor power (P) to be sum of thermal powers of fuel channels and reactor pool is calculated;
- These measurements, don't supply direct information on current fission power, to be generated in fuel channel; they are taking into account additionally heat generation due to gamma radiation from FE and the heat exchange between FE and reactor pool;
- To estimate the ⁹⁹Mo activity in molybdenum channel it is necessary to get information on real fission power to be generated in uranium plates.

Technical characteristic of uranium targets & irradiation rig



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- The uranium targets: plates with dimension of 203x40x1.45 mm; the core including fission material in the shape of dispersion in aluminum: UAl_x (x \cong 3) in Al (> 90% ²³⁵U);
- Container for irradiation of uranium target includes 8 plates positioned in special cans (four plates on each);
- Irradiation of uranium plates set takes place in the adopted for this goal fuel element of MARIA reactor;
- The internal structure of the molybdenum channel preserve the construction principle of the *Field's tube*;
- The construction of the internal fuel structure allows after reactor shutdown and removing the fuel channel head to take out and transportation of the irradiation container.

Iternative Molybdenum-99 IAE / NCBJ experience



Irradiation molybdenum targets using neutron capture method

- Irradiation facility:
- Target:
- Batch:
- Irradiation container:
- Container closing:
- Mass:
- Neutron flux density:
- •
- Yield at EOI:
- Time of irradiation:

EWA & MARIA RRs MoO₃ (powder) 3 x vials inside Al container **0** 25 mm / L=100 mm cold weld able 3x2,5g 1x10¹⁴ [n/cm² s] (MARIA) 8x10¹³ [n/cm² s] (EWA) 2.0 Ci/g (MARIA) 1.2 Ci/g (EWA) 87 h (MARIA) 1 week (EWA)

Alternative Molybdenum-99 IAE / NCBJ current status



- Most of ⁹⁹Mo is produce using fission method: 95% of the world demand is covered by this method;
- The extremely low specific activity of ⁹⁹Mo from neutron capture method makes its use less convenient;
- Difficulties in terms of safety & waste management are limitation for production fission ⁹⁹Mo;

Current status:

- NCBJ plans continuation of irradiation of targets HEU & LEU
- NCBJ plans design, construct & operate own production facility based on irradiated in the MARIA RR LEU targets
- RR MARIA is ready to restart irradiation ⁹⁸Mo targets & participate on program neutron of (n,γ) method

Alternative Molybdenum-99 IAE / NCBJ experience



Processing procedure

- Irradiated cans are opened using special cutters;
- Targets has been transferred to a solution flack containing 4N NaOH solution;
- Solution has been heated up to 60-70 °C to fascilate dissolution;
- Solution was stirred by passing compressed air;
- Solution was cooled and passed through a filter to remove any impurities;
- Result: sodium molibdate in sodium hydroxide solution;
- ⁹⁹Mo was used as a row material for conversion into medical product for extraction ^{99m}Tc.