UNIVERSITY OF MICHIGAN

NERS/BIOE 481

Lecture 04
Radioisotope Sources

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Henry Ford Health System
RADIOLOGY RESEARCH

III. Sources of Radiation (Continued)
E) Radioactive Isotopes
1) The atomic nucleus
2) Nuclear decay
3) Activity (decay rate)

Nuclear Notation

Mass number $A = Z + N$

Chemical symbol

$${}^{A}_{Z}X$$

Atomic Number (Proton number)

N = neutron number

The nucleus is comprised of a similar number of protons and neutrons.

Neutrons
neutral charge
1.008665 AMU

Protons
+ charge
1.007276 AMU

$${}^{16}_{8}O$$

Oxygen 16
16 nucleons
8 protons
8 neutrons

Radius of the atom and the nucleus for copper 63.

Cu Atom Size

In lecture 02, the radius of the outer shell electrons (M shell) for copper was deduced from the unscreened Bohr model;

$$r_m = \alpha_H \left(n^2/Z\right)$$

$$r_{Cu} = .52917\left(3^2/29\right)$$

$$r_{Cu} = .16, Angstroms$$

α_H is the 'Bohr radius'

Cu Nucleus Size

Scattering experiments suggest that nuclei are roughly spherical and have essentially the same density:

$$r_A = r_o A^{1/3}$$

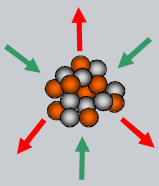
$$r_0 = 1.2\times10^{-15}, m$$

$$r_{63} = 4.8\times10^{-5}, Angstroms$$

ρ_n = 2.3 x 10^{14} g/cm^3

1 fermi = 10^{-15} m =1 femtometer (fm)= 10^{-5} Angstroms

Nuclear Forces and Stability

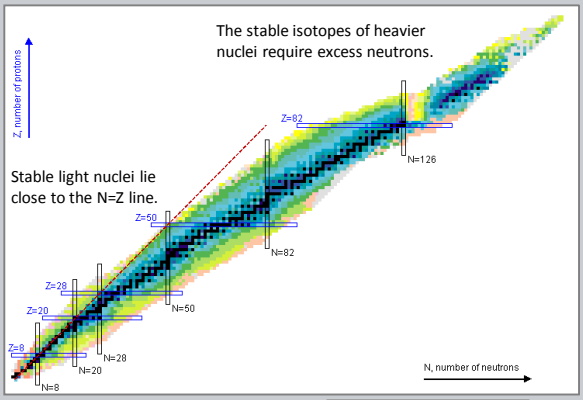


- Nucleons are held in the nucleus by a 'strong force' with a 'short range' of about 1 fermi.
  Yukawa (1935) proposed that the short range strong force came about from the exchange of a massive particle which he called a meson. The strong interaction is now thought to be mediated by gluons, acting upon quarks and anti-quarks.
  https://en.wikipedia.org/wiki/Strong_interaction
- Strong repulsive forces result from the charged protons in the nucleus.
- Quantum mechanical descriptions of the nucleus explain why the most stable configurations tend to result when neutrons and protons are paired.

^{4}He

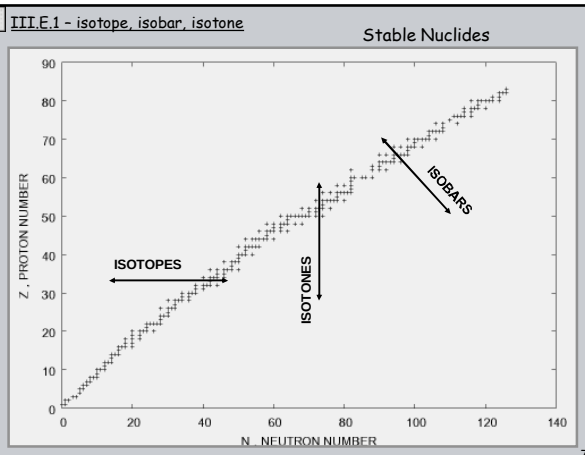
Nuclei with even numbers of protons and neutrons are particularly stable.

Nuclide Chart – Color coded by decay time (Black = stable)

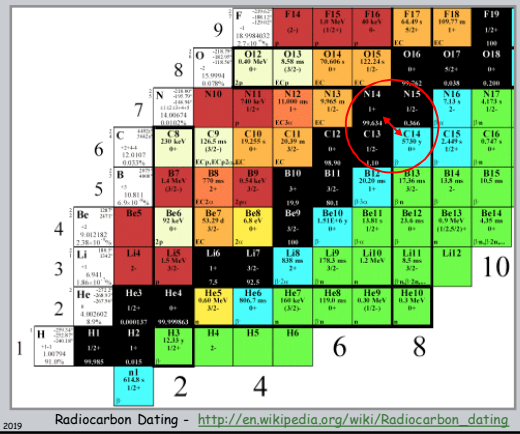


http://www.nndc.bnl.gov/nudat2/

III.E.1 – isotope, isobar, isotone



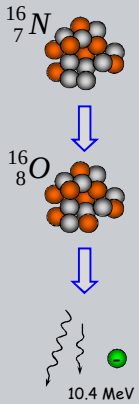
III.E.1 – Table of isotopes



Radiocarbon Dating - http://en.wikipedia.org/wiki/Radiocarbon_dating

III.E.2 – Nuclear Decay

- An unstable nuclei may spontaneously change its neutron to proton ratio and become more stable.
- The resulting 'daughter' nuclide has slightly less mass than the 'parent' nuclide.
- The energy equivalent to the mass difference is released as energetic photons (gamma rays) or light particles (electrons, positrons) in which case the nucleons in the daughter are the same as the parent.
- Some heavy nuclides may eject a particle with 2 protons and 2 neutrons, an alpha particle, with a net reduction of nucleons in the daughter nucleus.



III.E.2 – Beta Decay

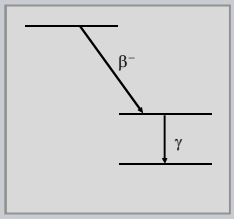
ν – The neutrino is an elementary particle with minimal mass and no charge.

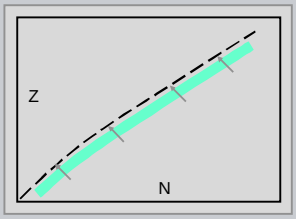
Reconfiguration of the nucleus

β^- (beta) emission

$$n \Rightarrow p^+ + e^- + \nu + energy$$

Decal Level Scheme





Effect:

Neutron ➔ Proton

After decay, the daughter isotope is often in a excited state that relaxes with gamma ray emission

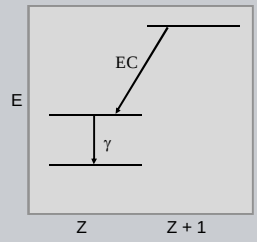
III.E.2 – Electron Capture

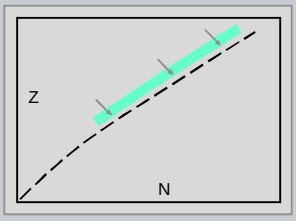
Reconfiguration of the nucleus

Electron Capture

$$p^+ + e^- \Rightarrow n + \nu + energy$$

Decal Level Scheme





Effect:

Proton ➔ Neutron

Electron capture is more probable for high Z nuclides as a method to reduce the proton number and increase neutrons.

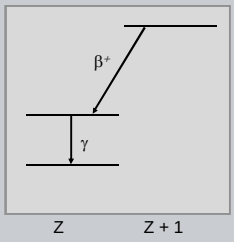
III.E.2 – Positron Decay

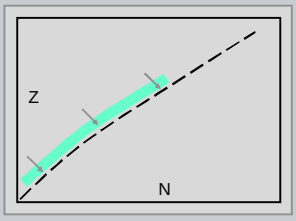
Reconfiguration of the nucleus

β^+ (positron) emission

$$p^+ \Rightarrow n + e^+ + \nu + energy$$

Decal Level Scheme





Effect:

Proton ➔ Neutron

Positron decay is more probable for low Z nuclides as a method to reduce the proton number and increase neutrons.

III.E.3 – Isotope decay rate

- For a sample with N nuclei of a radioactive nuclide, the rate of decay in disintegrations per second is given by a differential equation for amount of activity, **A**, based on a decay constant, λ (1/sec).
- This simple differential equation is easily solved for N and therefore for A.
- The time for a radioactive sample of A_o activity to decay to an activity equal to 1/2 of A_o is known as the half-life, $T_{1/2}$

$$A_{(t)} = \lambda N_{(t)} = -dN_{(t)}/dt$$

$$A_{(t)} = \lambda N_{(0)} e^{-\lambda t} = A_{(0)} e^{-\lambda t}$$

$$T_{1/2} = \frac{\ln(2)}{\lambda} = \frac{.693}{\lambda}$$

The SI unit for activity (**A**) is the Becquerel (Bq) which is equivalent to 1.0 disintegrations per second. The traditional unit of activity in the Curie (Ci) where 1.0 Curie is equal to 3.7×10^{10} disintegrations/second or 37 GBq. For radionuclides used in nuclear imaging, activities are often in the range from 1 to 20 mCi (40 to 800 MBq).

III.E.3 – Gamma ray emission fraction

- It is important to recognize the difference between the gamma ray emission rate of a radioactive sample and the decay rate in Bq.
- For example, Fluorine-18 decays with a 110 minute halflife to stable Oxygen-18. The decay may occur by either electron capture (EC) or positron (β^+) decay. The β^+ decay occurs for .967 of the decays with the remaining fraction occurring for EC.
- Since each β^+ decay produces 2 511 keV gamma rays, radioactive decay produces an average of 1.93 gamma rays per disintegration ,

$$\dot{N} = f_\gamma A = 1.93\, A$$

III.E.3 – Total gamma emission

- It is often necessary to determine the total number of gamma rays emitted over a period of time.
- This is obtained by integrating the exponential decay relation over the time period, T.

$$N = \int_0^T N(t)dt = \int_0^T f_\gamma A(t)dt$$

$$= f_\gamma A_0 \int_0^T e^{-\lambda t} dt$$

$$= f_\gamma \frac{A_0}{\lambda}\left(1 - e^{-\lambda T}\right)$$

III.F – Isotope Production (17 - charts)

III. Sources of Radiation (Continued)

F) Isotope Production

1) Production Methods
2) Radioisotope generators
3) Reactor production of 99Mo

III.F.1 – Production Methods

Radionuclides are produced by transforming the neutron-proton composition of a nuclide.

Four methods are:

- Nuclear fission
  The byproducts of nuclear fission from spent reactor fuel elements are separated to obtain radioisotopes.
- Neutron capture
  Neutrons, typically from a nuclear reactor, are absorbed by a target material to create a radioactive product.
- Charged particles bombardment
  Energetic charged particles, typically protons or deuterons, strike a target material.
- Nuclear decay
  A radioactive parent nuclide is used to generate a radioactive daughter product.

III.F.1 – Nuclear fission

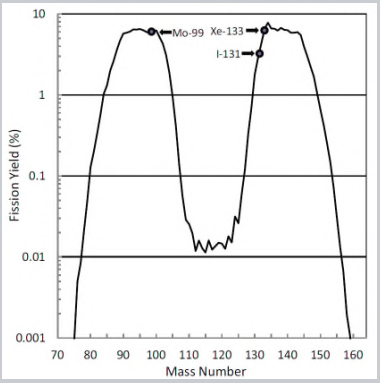
Reactor Produced – Fission Byproducts

$$^1_0n + {}^{235}_{92}U \Rightarrow {}^{99}_{42}Mo + {}^{133}_{50}Sn + 4\,{}^1_0n$$

Isotope production by separation of fission by products produces radioactive material that is prone to contamination with unwanted radioactive isotopes

Reactor produced radioisotopes:
neutron rich
beta decay

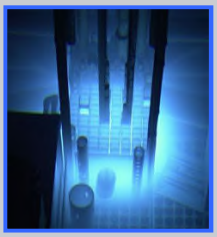
- Molybdenum-99 (66 h):
  Used as the 'parent' in a generator to produce technetium-99m.
- Xenon-133 (5 d):
  Used for pulmonary (lung) ventilation studies.
- Iodine-131 (8 d):
  Widely used in treating thyroid cancer.



Reactor Produced – Neutron Capture

$$n + {}^{98}Mo \Rightarrow {}^{99}Mo + \gamma$$

Isotope production by neutron capture produces radioactive material that is relatively free of other radioactive contaminants



Ford Nuclear Reactor
Phoenix Memorial Lab
University of Michigan

Mo-99 for Medical Imaging, National Academies Press, 2016

- Fission:
  Uranium fission is considered to be the "gold standard" process for producing Mo-99 because the production process is highly efficient, especially when highly enriched uranium (HEU) is used; and the Mo-99 produced has a high specific activity (>1,000 curies per gram [Ci/g]), making it suitable for use in conventional technetium generators.
- Neutron Capture:
  Neutron capture is a less efficient process for producing Mo-99 than is fission because the neutron capture cross section for Mo-98 is over three orders of magnitude smaller than the fission cross section for U-235. Moreover, Mo-99 produced by neutron capture has a lower specific activity (typically 0.1-1 Ci/g), too low for use in conventional technetium generators.

However, present concern regarding the use of HEU has lead to new technology for separating Mo-99 from neutron capture targets.

Charged Particle Bombardment

$${}^{1}_{1}p + {}^{18}_{8}O \Rightarrow {}^{18}_{9}F + {}^{1}_{0}n$$

Energetic charged particles from an accelerator strike a target to produce a radioisotope from reaction that add the charged particle to the nucleus and remove neutrons.

(p , α)	(p , n)	(p , 2n)
(d , α)	(d , n)	(d , 2n)
…	…	

Cyclotron produced radioisotopes:
proton rich, positron/EC decay

- ^{11}C (20 min), ^{13}N (10 min), ^{15}O (2 min)
  Short lived positron emitter synthesized with organic compounds for specialized PET imaging.
- Fluorine-18 (110 min):
  Positron emitter commonly synthesized with Fluoro-2-Deoxyglucose, FDG, for PET imaging procedures.
- Thallium-201 (73 h):
  Used for diagnosis of coronary artery disease, heart conditions such as heart muscle death, and for location of low-grade lymphomas.

Radioisotope daughter product

$${}^{99}_{42}Mo \Rightarrow {}^{99}_{43}Tc^{m} \Rightarrow {}^{99}_{43}Tc + \lambda$$

β^-, 66 hr — Isomeric, 6 hr

Some isotopes can be conveniently obtained by collected the daughter product resulting from the decay of a radioactive parent.

Technetium resulting from 99Mo decay, exists in a metastable state that decays to the ground state with a half life of 6 hours.

- A radioactive series involves the decay of a parent isotope, A, to a daughter isotope, B, with decay constant λ_A. and isotope B decays to C with decay constant λ_B;

$$A_{(t)} \xrightarrow{\lambda_A} B_{(t)} \xrightarrow{\lambda_B} C_{(t)}$$

- The number of atoms of A is described by;

$$A_{(t)} = A_{(o)} e^{-\lambda_A t}$$

- The number of atoms of B depends on the production from the decay of A and the loss by the decay of B. This can be described by a differential equation;

$$\frac{dB_{(t)}}{dt} = A_{(t)}\lambda_A - B_{(t)}\lambda_B = \left(A_{(o)} e^{-\lambda_A t}\right)\lambda_A - B_{(t)}\lambda_B$$

Note: A(t), B(t), and C(t) refer here to the number of atoms of A, B, and C

- Following the derivation in Evans (pg 477), the solution of this differential equation for B will be of the form ;

$$B_{(t)} = A_{(o)}\left(h_A e^{-\lambda_A t} + h_B e^{-\lambda_B t}\right)$$

- If this expression and it's derivative, dB/dt, is substituted into the prior differential equation and terms collected, we get;

$$e^{-\lambda_A t}\left(h_A\lambda_A - \lambda_A + h_A\lambda_B\right) = 0$$

- And therefore; $h_A = \dfrac{\lambda_A}{\lambda_B - \lambda_A}$

- The coefficient h_B depends on the value of $B_{(t)}$ at time t. For $B_{(t)} = 0$ at $t = 0$, $h_B = -h_A$ and thus;

$$B_{(t)} = A_o \frac{\lambda_A}{\lambda_B - \lambda_A}\left(e^{-\lambda_A t} - e^{-\lambda_B t}\right)$$

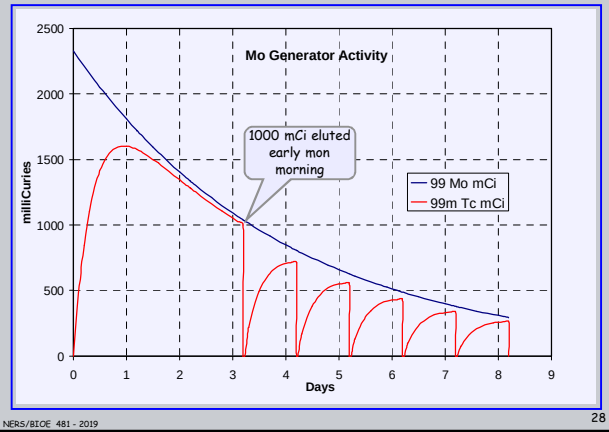
- For technetium generators, 99-Mo as A decays to 99m-Tc as B with a probability of 0.876 and a half-life of 66 hours. Otherwise it decays directly to stable 99-Tc.
- The metastable 99m-Tc relaxes to the 99-Tc ground state with a 6 hour halflife by emitting a 140 keV gamma ray (see Sorenson, App C).
- After several days, the second exponential in the preceding equation becomes zero and the 99m-Tc activity reaches equilibrium

$$A^{Tc}_{(t)} = \left(\lambda_B B_{(t)}\right) = 0.876\frac{\lambda_B}{\lambda_B - \lambda_A}\left(\lambda_A A_o e^{-\lambda_A t}\right)$$

The 99m-Tc activity in disintegrations per second, $B^{Tc}(t)$, is thus equal to $(0.876*1.1=0.96)$ times the 99-Mo activity, $A^{Mo}(t)$.

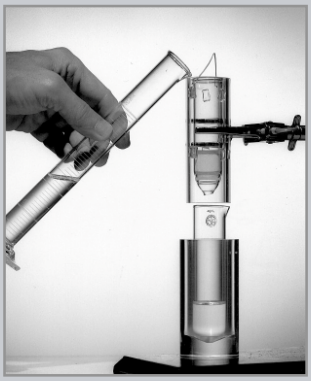
- If all of the 99m-Tc is eluted from the generator at time Te , the 99m-Tc activity will return to equilibrium ($t > T_e$) as described by;

$$A^{Tc}_{(t)} = 0.96A^{Mo}\left(_{T_e}\right)\left(e^{-\lambda_{Mo}(t-T_e)} - e^{-\lambda_{Tc}(t-T_e)}\right)$$



- A simple 99m-Tc generator can be made by permanently adsorbing 99-Mo material on a porous aluminum oxide (alumina) column as molybdate.
https://en.wikipedia.org/wiki/Molybdate

- The 99m-Tc daughter product, which is chemically present as pertechnetate, is separated by washing with a sodium chloride solution.



An original 99m-Tc generator, BNL, 1958

- In a generator device, the column is a small tube contained within a lead shield.
- Sterile saline in a medical vial is placed on a needle connected to the input line.
- A vacuum vial is then placed on a needle connected to the output line to draw the saline through the column.

99m-Tc generator, Lantheus 2015

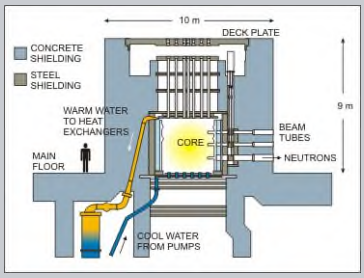
III.F.3 – Reactor Production of 99-Mo

- Over 70% of medical radioisotope examinations use 99m-Tc obtained from 99-Mo generators.
- Over 92,000 technetium generators were sold in the United States in 2005 supplying 22.9 million doses of Tc-99m radiopharmaceutials (Bio-Tech Systems, 2006).
- Until recently, most of the US supply of 99-Mo for medical generators has been produced by the irradiation of Highly Enriched Uranium (HEU) targets in a reactor. Either,
  - the National Research Universal (NRU) Reactor operated by Atomic Energy of Canada, Ltd. (AECL) at its Chalk River, Ontario, site or
  - the High Flux Reactor (HFR) operated by the Nuclear Research and Group at the Petten, Netherlands, site.

Both reactors are over 40 years old.

III.F.3 – Canadian NRU reactor

On Nov 18, 2007, AECL shut down the NRU reactor for 5 days of maintenance. During the shutdown, inspectors discovered that AECL had been operating the reactor without upgraded backup power systems for the cooling pumps and ordered the reactor to remain shut down until installation of these backup systems.



Due to 99-Mo shortages, the Canadian Parliament intervened and allowed the reactor to restart on Dec 16, 2007.

III.F.3 – Canadian NRU reactor

- In May 2009, a heavy water leak at the base of the reactor vessel was detected prompting a shutdown of the reactor. The leak was due to corrosion. The reactor was defueled and drained of all of it's moderator (heavy water). Remote tooling was created and used to repair all recognized areas of vessel wall thinning, caused by corrosion. The reactor returned to service in Aug 2010.

- The shutdown occurred when only one of the other four worldwide medical isotope reactors was producing 99-Mo, resulting in a worldwide shortage.

http://en.wikipedia.org/wiki/National_Research_Universal_Reactor.
http://www.nrucanada.ca/

III.F.3 – Canadian NRU reactor

2010: Canada initiates a new long term strategy for Mo99
- Cease production of 99Mo in October 2016 and move away from Government involvement in 99Mo production.
- Support research, development and demonstration of nonreactor based technology.

2015: Canada announced contingency to supply 99Mo from NRU after October 2016 until permanent shutdown of NRU.

NRU Time Line:
- End routine production of 99Mo October 31, 2016
- Establish and maintain stand-by 99Mo capability between November 01, 2016 and March 31, 2018
- Continue other operations including neutron beam research, non 99Mo isotope production, and operating of experimental fuel loop until March 31, 2018
- Permanent shutdown of NRU reactor March 31, 2018

III.F.3 – Netherlands HFR reactor

http://www.pallasreactor.com/?lang=en

The HFR reactor was shut down in late Aug 2008 after gas bubbles of unknown origin were discovered in the primary cooling system. An investigation determined that the bubbles were the product of corrosion of an aluminum sleeve where it contacted concrete. The reactor was not restarted until Feb 16, 2009.

- The HFR was then shut down for 5 weeks in July 2009 for maintenance.
- Another shutdown occurred from Feb to Aug of 2010 to repair cooling water pipework. This created worldwide shortage of medical isotopes since the NRU reactor in Canada was also shutdown during this time period.
- A replacement reactor named PALLAS is expected to operate in 2023.

III.F.3 – Medical Isotope production & US policy

- Use of HEU targets for 99-Mo production is a source of concern in relation to US regulations designed to reduce the risk of terrorism.
- Reliable supply of 99-Mo is presently of high concern due to the age and stability of the reactors now being used.
- Alternative methods using Low Enriched Uranium (LEU) and alternative reactor facilities are detailed in a report released by the National Research Council.

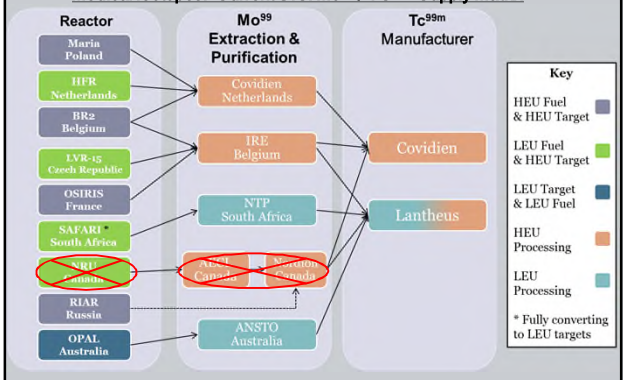
"Medical Isotope Production Without HEU", National Academy of Sciences, Committee on Medical Isotope Production Without HEU, National Research Council, 2009.

https://www.ncbi.nlm.nih.gov/books/NBK215149/

- On January 2, 2013, President Obama signed into law the American Medical Isotopes Production Act of 2011 (AMIPA) as part of the 2013 National Defense Authorization Act. The AMIPA encourages the domestic production of LEU Mo-99 and provides for the eventual prohibition of the export of HEU from the United States.
- In addition, the Centers for Medicare Services (CMS) recently stipulated in the 2013 final Medicare payment rules, for Medicare Hospital Outpatients, that CMS will provide incremental reimbursement for every Tc-99m diagnostic dose produced from non-HEU sourced Mo-99.
- On January 9, 2013, Lantheus announced that the Company has added a Low-Enriched Uranium (LEU) Tc 99m Generator to the Company's nuclear imaging product portfolio.

Lantheus TechneLite (LEU):
http://www.technelite.com/home.html

Medical Isotopes: Current U.S. Mo^{99} / TC^{99m} Supply Matrix

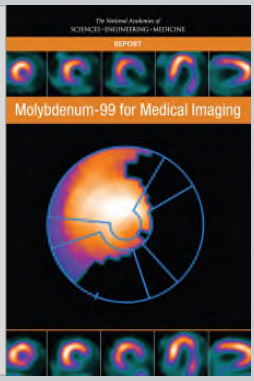


LANL Mo99 meeting, dec 2011, Miles Pomper presentation

Since 2009, the Department of Energy's National Nuclear Security Administration (DOE-NNSA) has signed cooperative agreements with five private-sector companies to assist their efforts to develop domestic capabilities to supply molybdenum-99 (Mo-99) without the use of highly enriched uranium (HEU) targets.

- BWX Technologies
- General Atomics (GA)
- General Electric-Hitachi (GEH)
- NorthStar Medical Radioisotopes
- SHINE Medical Technologies

The objective is to develop the capability to supply more than half of the domestic need for Mo-99.



National Acadamies Press, 2016

Neutron generators

- With support from the US DOE, Shine Medical Technologies is developing an accelerator driven subcritical assembly for Mo-99m production using neutron generators from Phoenix Nuclear Labs (PNL). Both are located in Wisconsin.
- Shine's production facility will operate as many as eight PNL neutron generators simultaneously.
  - 2017 – laboratory construction started.
  - 2018 – First accelerator delivered.
  - 2021 – Mo-99m supply projected
- Lantheus Medical Imaging Inc recently has an agreement with Shine for the future supply of Mo-99m.

http://phoenixnuclearlabs.com/

NorthStar Medical Radioisotopes

NNSA agreements to develop domestic Mo-99 production using two different processes:

- Photon-induced transmutation of molybdenum-100 (100Mo(γ,n)99Mo) using photons produced with linear accelerators.
- Neutron capture of molybdenum-98 (98Mo(n,γ)99Mo) using neutrons produced in a research reactor.

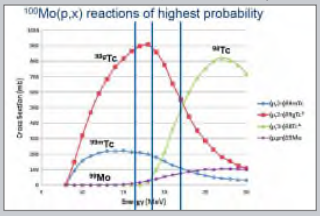
The FDA approved the NorthStar Tc-99m generator in 2018.

The RadioGenix Generator utilizes a Tc-99m-selective resin column to recover sodium pertechnetate (NaTcO4) containing Tc-99m from an alkaline solution of Mo-99. The was designed specifically for use with the low-specific-activity Mo-99 produced by NorthStar.

https://www.northstarnm.com/

Cyclotron Production

- New approaches are being investigated in Canada to produce Tc99m directly by small cyclotrons located within hospital facilities. Multi-institutional consortia in Alberta and with TRIUMF have been funded.
- The $^{100}Mo(p,2n)^{99g}Tc$ reaction is used with proton bombardment of an enriched 100Mo target.
- The key innovation for the implementation of this technology resides with the target plate technology.



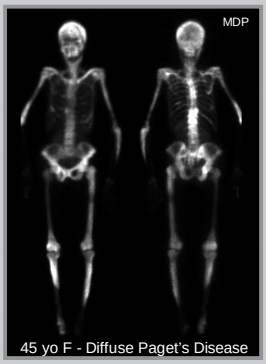
- Target plates consist of approximately 80 mg of ^{100}Mo that is sintered and pressed into the backing plate.
  - TRIUMF usees a ^{100}Mo target thickness of around 300um on a tungsten backing plate for use with both the ACSI TR19/TR24 and GE PET-TRACE cyclotrons.
  - Alberta uses an aluminium backing plate that fits in the ACSI T24 cyclotron.

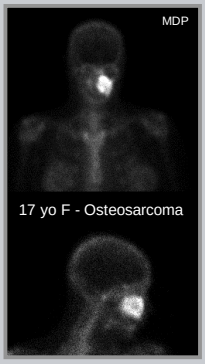
III.F – Radiopharmaceuticals (- charts)

III. Sources of Radiation (Continued)
G) Medical Radiopharmaceuticals
1) pharmaceuticals and function
2) Ideal radionuclide properties

III.F.1 – Tc-99m Bone Scans

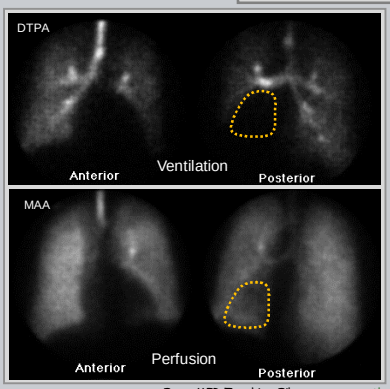
- Tc-99m Medronate (MDP)
- Tc-99m Etidronate
- Tc-99m Oxidronate (HDP)





III.F.1 – Tc-99m Lung Scans

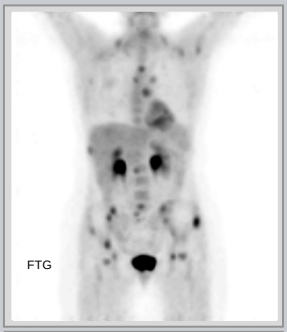
- Tc-99m Macro Aggregated Albumen (MAA)
- Tc-99m Pentetate (DTPA) aerosol



- Images from an aerosol Ventilation examination indicate absent ventilation to the left lower lung.
- A blood Perfusion exam shows only minimal decrease to the left lower lobe.
- Bronchoscopy was performed and a large mucous plug in the bronchus to the left lower lobe was removed. The patient rapidly improved.

III.F.1 – F-18 FDG

F-18 fluorodeoxyglucose (FDG)



47 yo Female with a history of breast Cancer. PET FDG images demonstrate multiple metastases that were not detected on CT or bone scans.

- The distribution of 18F-FDG is a good reflection of the distribution of glucose uptake and phosphorylation by cells in the body.
- It is retained by tissues with high metabolic activity, such as most types of malignant tumors.
- The production of ^{18}F-FDG is usually done by proton bombardment of ^{18}O-enriched water, causing a (p,n) reaction in the ^{18}O to produce ^{18}F. The ^{18}F is then collected and immediately attached to deoxyglucose in a "hot cell"
- The labeled FDG compound (T1/2 = 110 min) is rapidly shipped to points of use by the fastest possible mode.

III.F.1 – Others

Other commonly used radiopharmaceuticals

• Tc-99m Sulfur Colloid	Liver Scan
• Tc-99m Sestamibi	Cardiac Perfusion
• Tc-99m Exametazime	Brain Perfusion
• Tc-99m Succimer (DMSA)	Kidney Scan and Function
• Tc-99m Red Blood Cell	Blood Pool Imaging
• Xe-133 gas	Lung ventilation imaging
• Ga-67 citrate	inflammatory lesions
• Th-201 chloride	Heart muscle imaging

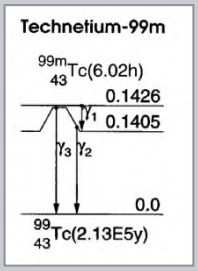
NOTE: these pharmaceutical are used for nuclear medicine imaging including SPECT. We will consider PET agents later.

III.F.2 – Ideal Radioisotope Properties.

1. Single gamma ray emitted.
2. No charged particle emission.
3. Half-life:
   - Long enough to permit imaging.
   - Short enough to reduce dose.
4. Gamma energy:
   - High enough to escape the body.
   - Low enough to permit efficient detection
5. Chemical attributes that enable useful pharmaceutical compounds

Tc-99m comes close to the characteristics desired of an ideal isotope:

- The decay scheme is dominated by a single gamma ray.
- As an isomeric transition, no charged particles are emitted from the nucleus, although low energy conversion electron are produced with low probability.
- The half life of 6 hours is convenient for daily procedures.
- The 140 keV energy escapes relatively well and is effectively detected by 3/8" thick detector crystals.
- Tc chemistry is not nearly as good as for C, N, and O. However, overtime numerous effective compounds have been developed.



- Tc 99m decay characteristics are shown in Sorenson-App.C .
- The yield of 140.5 keV gamma rays ($\gamma 2$) is 0.89 per decay.
- An internal conversion electrons (ce-[K,L,..],$\gamma 2$) is otherwise emitted with 0.11 per decay