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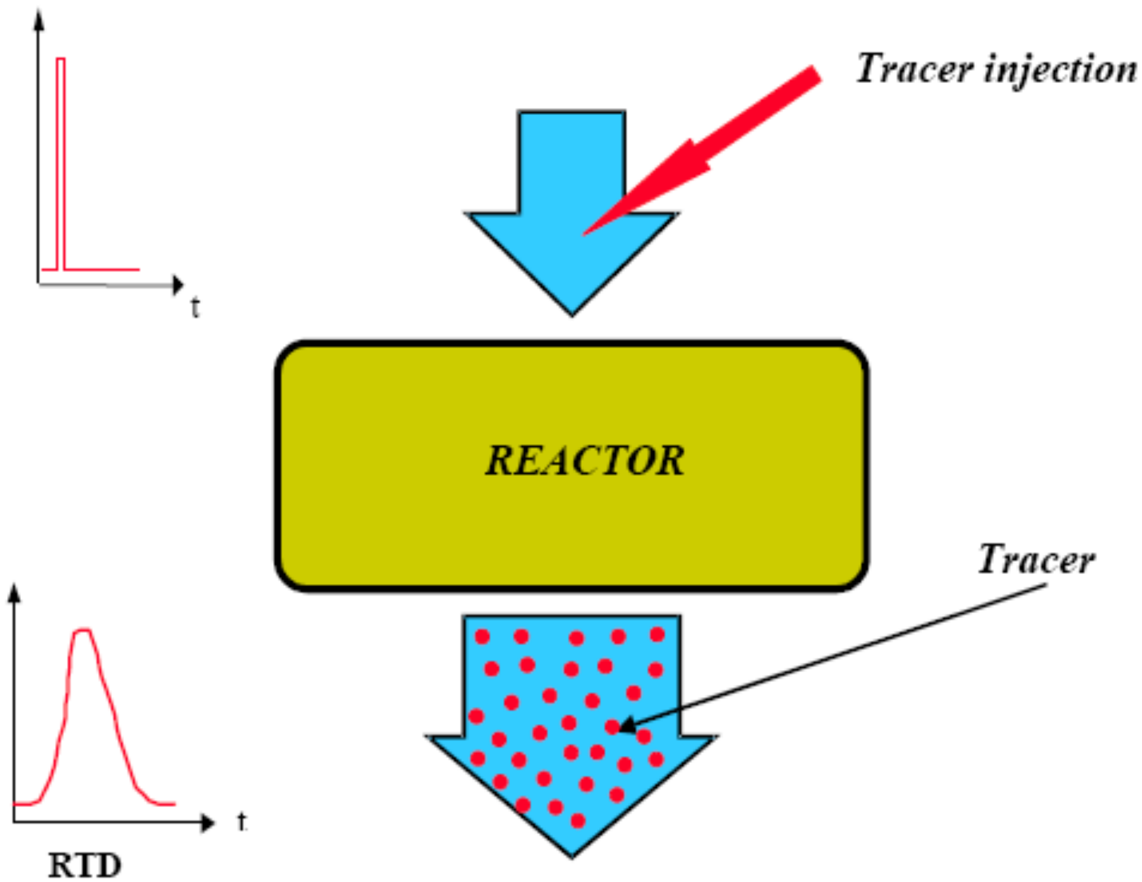
Radiotracers Applications

RADIOTRACERS

A tracer is any substance whose atomic or nuclear, physical or chemical properties provide for following of behavior of various physical or chemical processes.

There are many kinds of tracers; radioactive tracers are the only used for online diagnosis of industrial reactors.



RADIOTRACER SELECTION

Factors that are important in the selection of a radiotracer are given as follows:

- ***Physical/chemical form and properties of tracer with respect to the system under investigation***
- ***Half-life of tracer***
- ***Specific activity of tracer***
- ***Type and energy of radiation emitted***
- ***Availability and cost of tracer***
- ***Method of measurement (on-line or sampling)***
- ***Handling of radioactive materials, radiological protection/regulations.***

Intrinsic (or chemical) tracers, *are molecules containing an isotope of one of the molecule's natural elements. For example, in the case of water, Tritium ($^{1}H\ ^{3}H^{16}O$) measured by nuclear techniques (in practice liquid scintillation counting) is an intrinsic tracer. In this case, the water molecule is traced from the inside, in the intimacy of it nucleus, consequently the water tracer will (in practice) follow all movements and reactions of water itself.*

Extrinsic (or physical) tracers, *are made up of atoms or molecules supposed to share the same dynamic characteristics and, in general, the same mass flow behavior as the investigated medium. For example, in case of water, $Na^{131}I$- and $^{99m}TcO_4$ are examples of extrinsic tracers.*

Extrinsic tracers are mostly employed in industry.

RADIOTRACER ADVANTAGES ARE:

- high detection sensitivity for extremely small amounts: 1 Ci of ^{131}I- weighs 8 µg, while 1 Ci of ^{82}Br- weighs only 0.9 µg; that's why, when injected, they do not disturb the dynamics of the system under investigation;

- radiotracer can be measured from the outside of a pipe or vessel;
-disappearance of the tracer from the medium under investigation through radioactive decay provides for a repetition of experiments on the same location with the same tracer, all while pollution declines to a minimum;

- radiotracer can be selective; several tracers may be employed simultaneously and measured with field spectrometers;

- emission of radiation is a specific property of the radionuclide, not affected by interference from other materials in the system.

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Gas phase

Non-nuclear techniques	*Radiotracers*
Traces Used: Reactive Gases	Recommended : Gaseous radiotracers
Cl_2, SO_2, NO_2, SF_6, etc.	Ar-41, Kr-79, Br-82 (as CH_3Br)
Advantages	
-Easy availability -Simple analysis	-High selectivity -Low detection limit -In-situ/On-line measurement (no sampling needed)
Disadvantages	
-Poor selectivity -Poor detection threshold -Statistically representative sample	-Availability -Costs -Radiation safety regulations
Applications in WWTP: (1) Aeration tanks, (2) Biological filters, (3) Disinfection unit, (4) Anaerobic digester	

Liquid phase

Non-nuclear techniques	*Radiotracers*
Tracers used:	Radiotracers recommended:
Electrolytes (NaCl solution) - conductivity Dyes (Rhodamine, Fluorescence) - color Acids & Alkali - pH	$K^{82}Br$, NH_3Br, ^{99m}Tc, ^{113m}In-EDTA, ^{46}Sc, I (131&125), $^{24}Na_2CO_3$
Advantages	
- Easy availability - Cheap	- No interaction with WWTP treatment - Low detection threshold - On-line measurement - No limitations due pH, conductivity, color - Some radiotracers are readily available.
Disadvantages	
- Not suitable for colored, conducting liquids - Stratification possibility - Large threshold detection concentration - Possible interference with WWTP operation	- Radiation safety regulation - Relatively expensive detection equipment
Applications in WWTP: (1) Central collection/Flow rates, (2) Equalization tank, (3) Flash mixer, (4) Clarifiers, (5) Aeration vessel (ASP), (6) Anaerobic digesters, (7) Dispersion of discharge in water bodies.	

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Solid phase

Non-nuclear techniques	*Radiotracers*
Tracers used	Radiotracers recommended
No known solid tracers Current method: sampling, filtering, drying, weighing	In-113m, Tc-99m, Au-198, etc.
Advantages	
	- Same as in case of liquid phase radiotracers - Can be independently detected without interference with gas and liquid detection
Disadvantages	
- Tedious - Statistically representative sampling	- Radiation safety regulation - Relatively expensive equipment
Applications in WWTP: (1) Collection networks, (2) Sand and grit removal, (3) Clarifiers, (4) Biological reactors (aerobic and anaerobic) (5) Discharge networks	

Common types of radionuclide sources

Currently most radionuclides are produced from three types of sources:

- nuclear research reactors,
- radionuclide generators
- cyclotron facilities.

  (generators still need a reactor or cyclotron source to produce the parent radionuclide)

	Nuclear Reactors	**Generators**	**Cyclotrons**
Principle of production	Target material inserted in the neutron flux field undergoes fission or neutron activation transmuting into radionuclide of interest	Long-lived parent radionuclide decays to short-lived daughter nuclide of interest. Daughter nuclide elution follows in pre-determined cycles	Target material irradiation by charged particle beams. Inducing nuclear reactions that transmute the material into radionuclide of interest.
Transmutation base	Neutrons	Decay	Mostly proton beam
Advantages	- High production efficiency - Centralized production: one research reactor able to supply to large regions	- Available on site, no need for logistics - Mostly long shelf life - Easy to use - Limited radioactive waste: returned to manufacturer after use	- High specific activity in most cases - Small investment in comparison to nuclear reactor - Little long-lived radioactive waste
Disadvantages	- Extremely high investment cost - High operational costs - Considerable amounts of long-lived radioactive waste - Long out-of-service periods - Public safety concerns	- Supplies in cycles according to possible elution frequency; - Trace contaminants of long-lived parent nuclide in eluted product	- Complex logistics needed for short-lived produced radionuclides - Radionuclide production limited depending on installed beam energy

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Producing technetium

Tc-99m is produced by the radioactive decay of molybdenum-99 (Mo-99). Mo-99 is currently made in nuclear research reactors through the fission (splitting) of enriched uranium. Neither isotope can be stockpiled because they decay rapidly: the amount of radiation emitted by Tc-99m halves every 6 hours, and the yield of Tc-99m obtained from Mo-99 halves every 66 hours. Mo-99 is loaded into dispensers called generators, which are shipped to the end-users. Tc-99m is extracted from the Mo-Tc99m generator by flushing it with saline (saltwater). A generator typically lasts 1-2 weeks.

Nuclear reactors producing >90% of the world's Mo-99 are:

HFR Netherlands	38%	2024
BR-2 Belgium	26%	2026
Safari-1 South Africa	21%	2030
MARIA Poland	15%	2030
OPAL Australia	15%	2057
LVR-15 Czech Republic	14%	2028
NRU Canada	*Previously 30%, now none. Closed Oct 2016*	

When a target is under irradiation in a nuclear reactor, the activation per second can be represented by:

$$dN^*/dt = \Phi \sigma N_T$$

where:

- Φ is the neutron flux ($n/cm^2 s$)
- σ is the activation section (neutron capture cross-section, 10^{24} barn)
- N^* is the number of activated atoms (atoms/g)
- N_T is the total number of atoms present in the target (atoms/g)

Since the product radioisotope starts decaying with its own half-life, once production starts, the net growth rate of active atoms can be written as:

$$dN^*/dt = \Phi \sigma N_T - \lambda N^*$$

where: $\lambda = \ln 2/T_{1/2} = 0.693/ T_{1/2}$ is the decay constant of the being created radioisotope, and $T_{1/2}$ is its half-life.

The above equation can be solved to determine the value of radioactive atoms at the end of irradiation time t_i, as follows:

$$N^* = \sigma \Phi N_T[1- \exp(-\lambda t_i)]$$

Surface labelling

Adsorption of a radionuclide on surface of a solid has been used as a labelling method for sand particles and many powdered materials.

Solid particles are first soaked in stannous chloride ($SnCl_2$) and then placed in an aqueous solution of gold chloride containing radioactive ^{198}Au. The gold exchanges with tin, through a reduction-oxidation process, to produce labelled particles.

Another simple method for surface labelling is to absorb, soak or sprinkle the material with radioactive solution. This method has been used for labelling coal and refractory materials.

With surface labelling methods, unlike direct activation, the activity becomes proportional to the surface area of the material rather than its mass, and it thus depends on the grain size distribution.

Some fine materials (<100 microns) like silt, cement, carbon black, aluminum powder, etc. labelled with Au-196, Tc-99m or In113 can be considered as mass labelling.

Major Radiotracers used in Industry

Isotope	Half-life	Radiation, Energy(MeV)	Chemical Form	Tracing of phase
Tritium (^3H)	*12.6 y*	*Beta, 0.018(100%)*	*Tritiated water*	*Aqueous*
Sodium-24	*15 h*	*Gamma:1.37(100%)2.75(100%)*	*Sodium carbonate*	*Aqueous*
Bromine-82	*36 h*	*Gamma: 1.32 (27%)*	*Ammonium bromide,* *p-dibrom-benzene,* *Dibrobiphenyl* *CH_3Br*	*Aqueous* *Organic* *Organic* *Gases*
Lanthanum-140	*40 h*	*Gamma: 1.16 (95%), 0.92 (10%)* *0.82(27%), 2.54 (4%)*	*Lanthanum Chloride,* *Lanthanum oxide*	*Solids*
Gold-198	*2.7 d*	*Gamma: 0.41 (99%)*	*Chloroauric acid*	*Solids /aqueous*
Mercury-197	*2.7 d*	*Gamma: 0.077(19%)*	*Mercury metal*	*Mercury*
Iodine-131	*8.04 d*	*Gamma: 0.36 (80%), 0.64 (9%)*	*Potassium or Sodium iodide,* *Iodobenzene*	*Aqueous* *Organic*
Chromium-51	*28 d*	*0.320 (9.8%)*	*Cr-EDTA, $CrCl_3$*	*Aqueous*
Technetium-99m	*6 h*	*Gamma: 0.14 (90%)*	*Sodium pertechnetate TcO_4^-*	*Aqueous*
Scandium-46	*84 d*	*Gamma: 0.89(100%)1.84(100%)*	*Scandium oxide,* *Scandium chloride,* *$ScCl_3$ (Sc^{3+})*	*Solids*
Xenon-133	*5.27 d*	*Gamma: 0.08 (100%)*	*Xenon*	*Gases*
Krypton-85	*10.6 y*	*Gamma: 0.51(0.7%)*	*Krypton*	*Gases*
Krypton-79	*35 h*	*Gamma: 0.51 (15%)*	*Krypton*	*Gases*
Argon-41	*110 min*	*Gamma: 1.29 (99%)*	*Argon*	*Gases*

Some radiotracers induced by direct activation of solids

Irradiated material	*Induced radionuclides*
Coal	^{46}Sc, ^{59}Fe (after decay of ^{56}Mn, ^{24}Na)
Clinker, cement	^{24}Na
Cracking catalyst	^{140}La
Gold ore	^{198}Au, ^{59}Fe, ^{42}K, ^{140}La, ^{56}Mn, ^{24}Na, ^{46}Sc, ^{51}Cr
Copper ore	^{64}Cu, ^{42}K, ^{140}La, ^{24}Na
Carbon black	^{24}Na

Commonly used radiotracers in aqueous solutions

Radionuclide	Half-life	γ radiation of interest (MeV)	Chemical form
^{24}Na	*15 h*	*1.37 (100%), 2.75 (100%)*	Na_2CO_3, $NaHCO_3$
^{56}Mn	*2.6 h*	*0.85 (100%), 1.8 (30%), 2.1 (20%)*	$MnSO_4$
^{82}Br	*36 h*	*0.55 (70%), 1.32 (27%)*	NH_4Br
^{198}Au	*2.7 d*	*0.41 (99%)*	$HAuCl_4$
^{131}I	*8.04 d*	*0.36 (80 %), 0.64 (9%)*	*NaI, KI*
^{51}Cr	*27.8 d*	*0.325 (9%)*	*Cr-EDTA complex*

Common liquid radiotracers for organic media

Liquid tracer	*Chemical form*	*Boiling point (^oC)*
Para-dibromo- benzene	$C_6H_4{}^{82}Br_2$	219
Bromo-dodocane	$C_{12}H_{25}{}^{82}Br$	240
Ammonium-iodide	$NH_4{}^{131}I$	220
Ammonium- bromide	$NH_4{}^{82}Br$	235
Bromo-naphthol	$^{82}BrC_{10}H_6OH$	130
Iodo-benzene	$C_6H_5{}^{131}I$	188
Sodium-iodide	$^{24}Na^{131}I$	1304

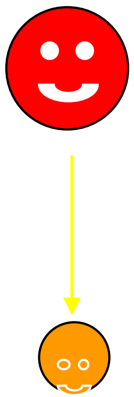
Major Problem in Radiotracer Applications for countries without nuclear reactor

- *Timely availability of a tracer is the greatest barrier to the use of radioactive tracer. Many urgent applications can not be carried out.*
- *Countries with no radioisotope production facilities need to import the radiotracers.*

Solution ?

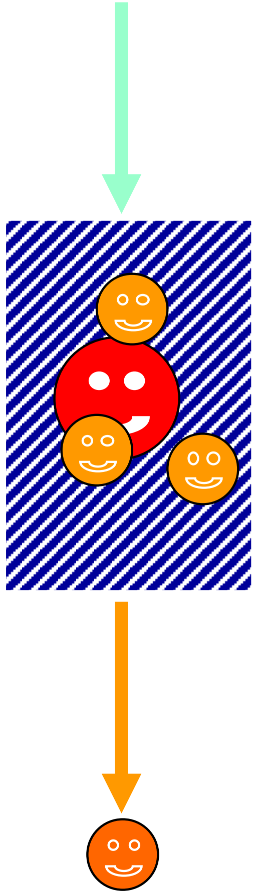
Radionuclide Generators

Generator principles

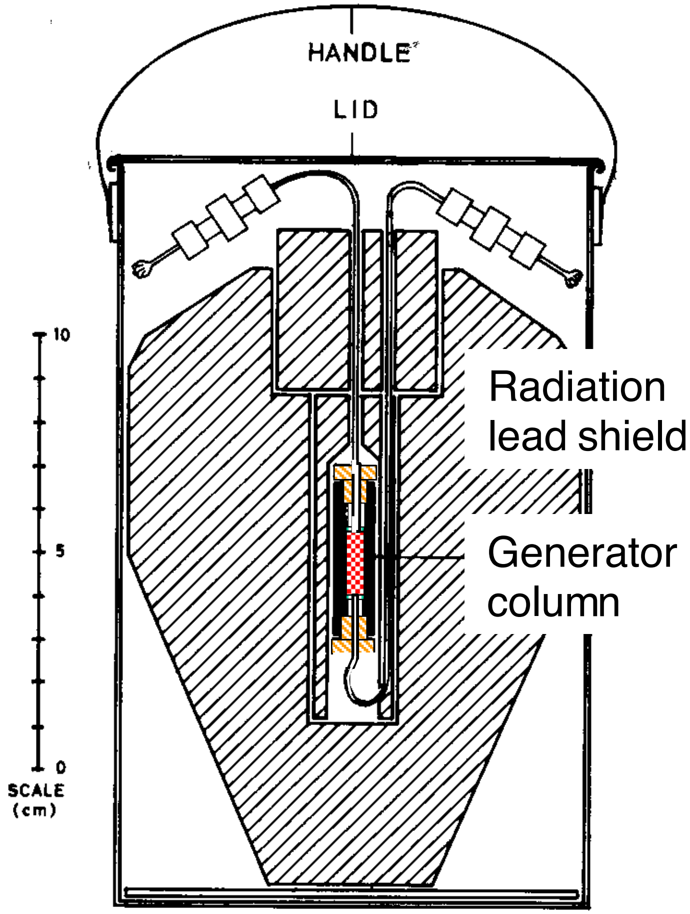


Mother radionuclide

Daughter radionuclide



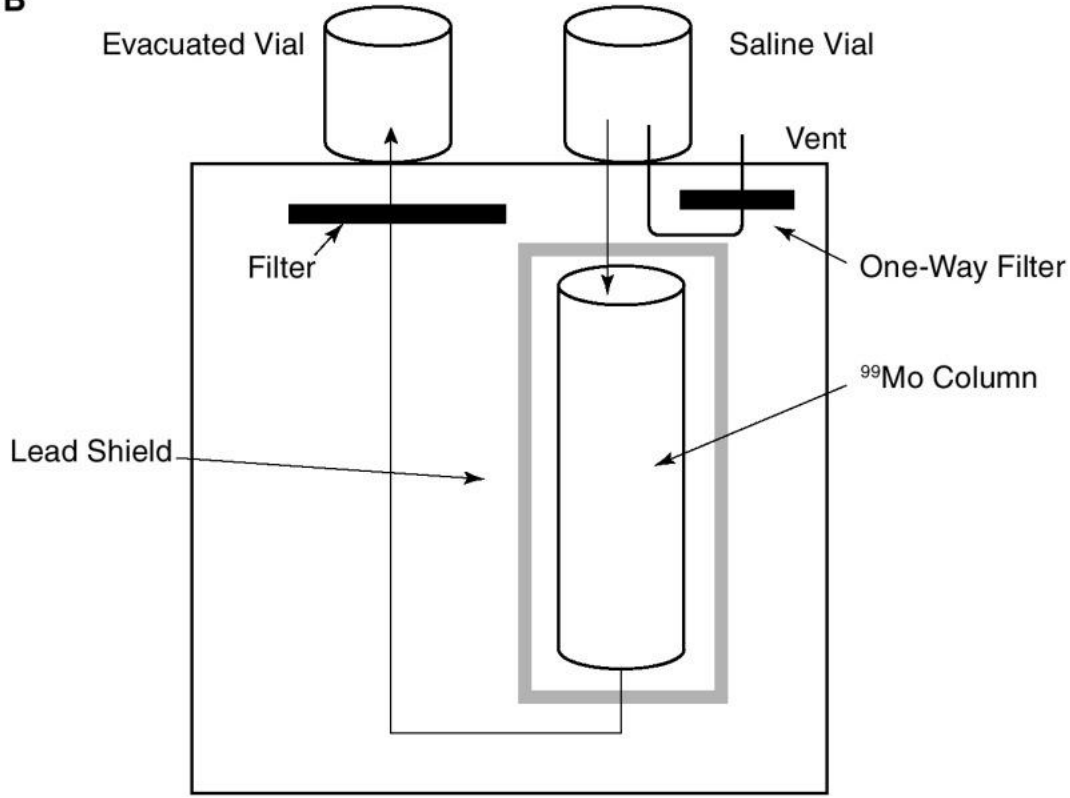
^{99m}Tc is eluted as an anion TcO_4^- (perthecnetate)



22 million medical diagnostic procedures per year in more than 100 countries

A

B



a) External view of a technetium generator produced by the Australian Nuclear Science and Technology Organisation
b) Schematic diagram showing the internal structure of a typical technetium generator.

MAJOR RADIONUCLIDE GENERATORS

$^{99}Mo \rightarrow {}^{99m}Tc$, $T_{1/2}$ = 66 h → 6 h, IT 140keV

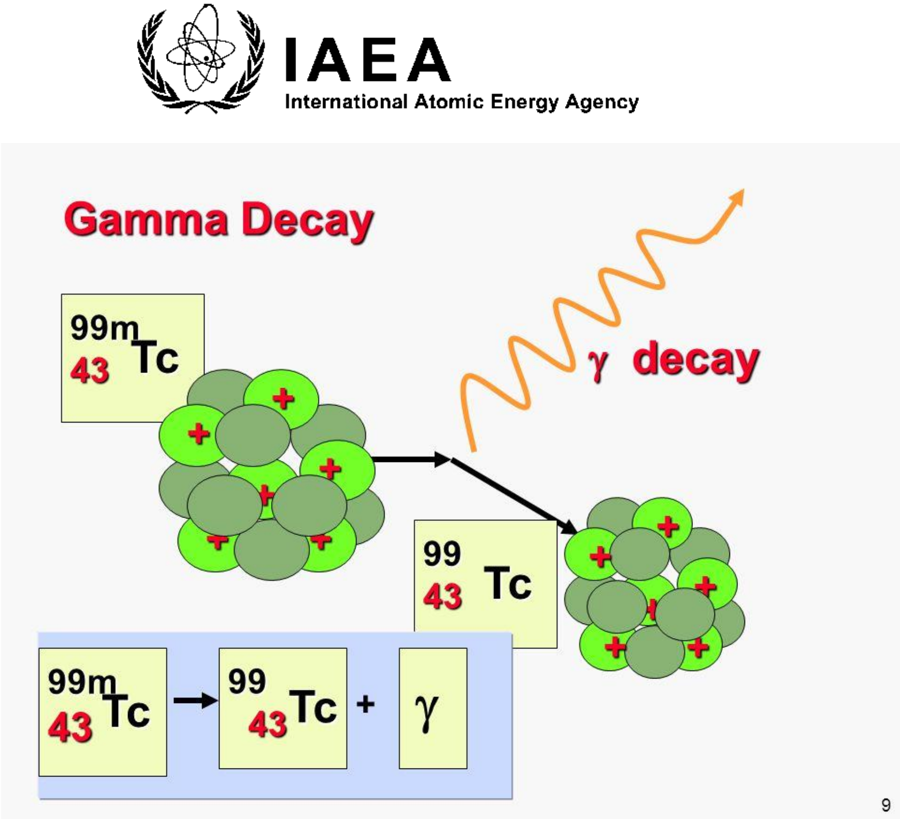
$^{113}Sn \rightarrow {}^{113m}In$, $T_{1/2}$ = 115.1 d → 99.5 min, IT 392 keV

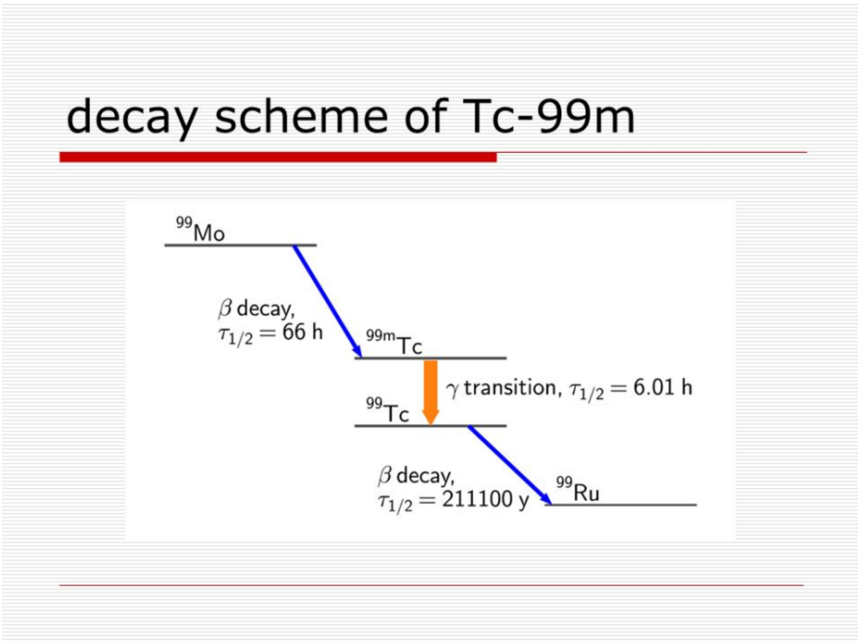
$^{137}Cs \rightarrow {}^{137m}Ba$, $T_{1/2}$ = 30.17 y → 2.55 min, IT 661.6 keV

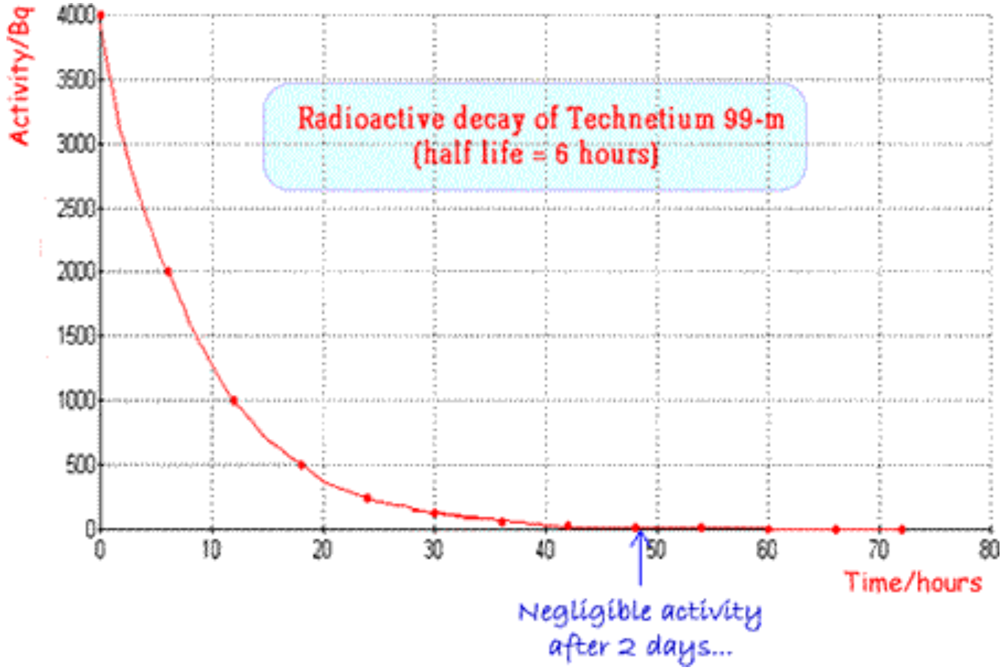
Radioisotope generator in investigation:

$^{68}Ge \rightarrow {}^{68}Ga$,

γ 1077 keV, 270.8 d 67.6 min,







Question: Into which substance does ^{99m}Tc decay when it emits gamma radiation?

a. *Tc - 99*

b. *Mb - 98*

c. *Tc - 98*

d. *Mo - 99*

Problem 1.

I-131 sodium iodide has a tbiol of 24 d. What is teff?

1/ teff = 1/ tphys + 1/ tbiol = 1/8 + 1/24 = 1/6 day-1

so.... teff = 6 d

Problem 2.

A Tc-99m compound has a teff = 1 hr. What is tbiol?

1/ tbiol = 1/ teff - 1/ tphys = 1/1 - 1/6 = 5/6 hr-1

so... tbiol = 1.2 hr

Problem 3.

A radiopharmaceutical has a biological half-life of 4.00 hr and an effective half-life of 3.075 hr. What isotope was used?

1/ tphys = 1/ teff - 1/ tbiol = 1/3.075 – 1/4.00 = 0.0752033 hr-1

Therefore tphys = 13.3 hr and the radioisotope is I-123

Technetium, 99mTc, is one of the favorites for diagnostic scans because of short physical and biological half-lives. It clears from the body very quickly after the imaging procedures.

^{99}Mo/^{99m}Tc Generator, is used extensively in nuclear medicine and is widely available worldwide; for this reason the short half-life of the ^{99}Mo is not a disadvantage. $T_{1/2}$= 6 h of ^{99m}Tc is appropriate for a wide range of studies on industrial plant. The gamma-ray (140 keV) has a half-thickness of 5 mm of steel. Thus, the ^{99m}Tc, which is usually eluted as sodium pertechnetate solution, is suitable as a tracer for vessels of wall thickness up to about 10 mm.

Typical applications in industry: RTD and flow rate measurements

^{137}Cs/^{137m}Ba Generator, is not commercially available, but home made. The parent, ^{137}Cs may be purchased from a number of suppliers ($^{137}CsCl_3$). To provide a generator system it is necessary:

(a) To attach the cesium chemically onto appropriate support medium.

(b) To provide a suitable system for eluting the ^{137m}Ba daughter.

Typical application of this generator is the flow rate measurement for calibration.

^{113}Sn / ^{113m}In Generator, is commercially available. Gamma-ray energy of ^{113m}In (392 keV) together with useful half-lives of the ^{113}Sn parent (115.1 d) and ^{113m}In daughter (99.5 min) makes this generator more suitable for many industrial applications than the ^{99}Mo/^{99m}Tc. Once eluted with HCl, $^{113m}InCl_3$ is produced, which is good tracer for solids (sediments); mixing with EDTA solution, the ^{113m}In-EDTA compound is produced which is a good tracer for water like systems.

^{68}Ge/^{68}Ga Generator, *is not commercially available*.
This is in many respects similar to the ^{113}Sn/^{113m}In generator. Due to high gamma ray energy of 1.08 MeV, ^{68m}Ga can be used as a radiolabel on suitable tracers for plants with thick-walled pipes and vessels. The half-life of the parent (287 days) is conveniently long though the daughter's half-life of 68.3 minutes is too short for studies of more than a few hours' duration. It is not known whether this generator is currently commercially available.

Applicability of the $Na^{131}I$ and $^{99m}TcO_4^-$ as radiotracer

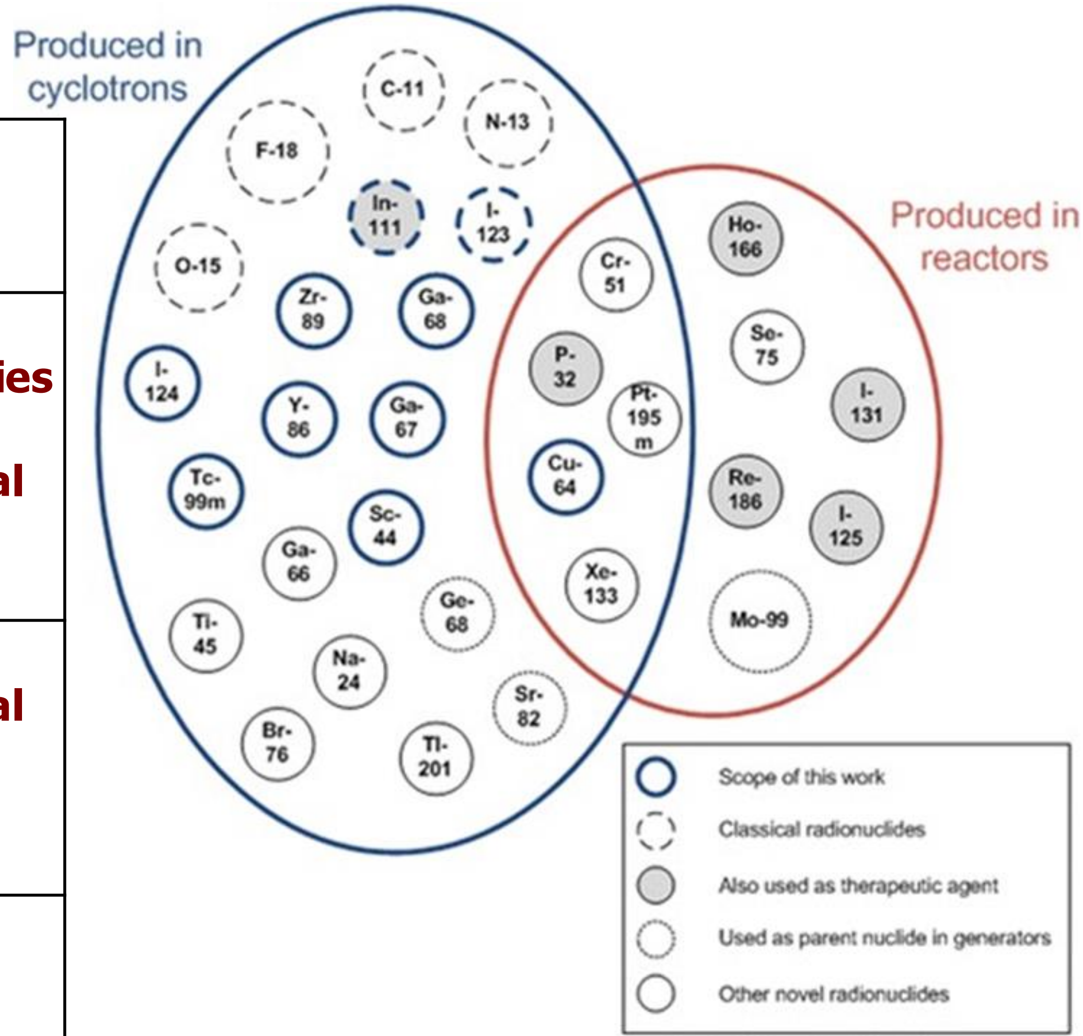
Can they be used for tracing water ? Yes

Can they be used for tracing solids? Tc only, sometimes

Can they be used for tracing organic liquids? NaI could be as iodobenzene, but how to prepare? And as alcohol?

Cyclotron produced radionuclides

Cyclotron type	Energy Range (MeV)	Approximate number	Typical location
Small medical cyclotron (SMC)	< 20 MeV	1050	- hospitals - universities - local commercial plants
Intermediate energy cyclotron	20 – 35 MeV	100	- regional commercial plants - research institutes
High energy cyclotron	> 35 MeV	50	- research institutes - cancer proton therapy centers



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Iodine-123 Radiochemical Sodium Iodide Solution

Chemical Form: Sodium iodide in 0.1N NaOH

Nuclear Reaction
Primary
124Xe (p,2n) 123Cs - 123Xe - 123I
Secondary
124Xe (p,pn) 123Xe - 123I

Radioiodines I-123

^{123}I ($T_{1/2}$ = 13.2 h, 100% EC, 83% γ 158 keV)

Today, the widely used ^{131}I therapeutic agent ($T_{1/2}$ = 8.02 d, 100% β^-, 90% β^-_{av} 192 keV, 82% γ 364 keV) is produced in nuclear reactors.

Iodine-123 is the second most widely used radioiodine and also the second most used imaging agent after ^{99m}Tc. Its popularity comes from its availability, perfect γ energy for imaging and appropriate half-life.

^{123}I is considered a classical radionuclide because of its widespread availability, well-established production method and routine production.