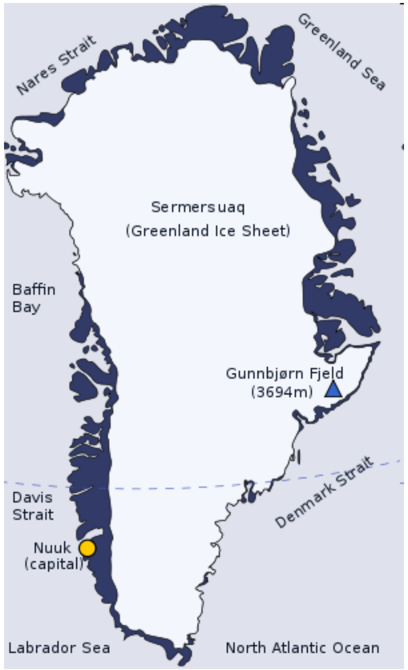
Tracers: A tool to study movement of matter

Example:

Hydrology Study of the Ice Sheet
In Greenland



Question:
How does the summary melting water reach the ocean

Radio-Tracers: A tool to study atomic motion in matter

Radio-Tracer methods find applications in nearly every field of science

- **Technical areas**: automobile industry, oil industry, chemical industry
- **Physical sciences :** physics, chemistry, geosciences, meteorology,…
- **Life science fields:** medicine, biology, physiology, nutrition, toxicology biotechnology, agriculture,,,,

Tracers: A tool to study movement of matter

- **Tracers** are used to follow the movement of atoms or molecules in matter and of macroscopic amounts of substances.

- Both **stable and radioactive** isotopes are used as tracers

- In the case of stable tracers the instrument most commonly employed is the **mass spectrometer**, a device that can determine the relative abundance of various isotopes in a sample.

- In the case of radioactive tracers (radio-tracers) the observations are made by analyzing **the radiation emitted by radioactive isotopes.**

Use of Radio-Tracers in Industry.

- Radioisotopes are used to **test material parts** and products such as metals, tire rubber, and engine oil **for wear**.
- Radiotracers are used to trace down **sources of pollution.**
- **Small leaks** can be detected in complex systems such as power station heat exchangers or oil pipelines in a refinery.
- **Mixing efficiency of industrial blenders**
- **Flow rates of liquids and gases in pipelines** can be measured accurately, as can the flow rates of large rivers.
- In agricultural laboratories, radioisotopes are used to determine how **plants take up nutritional materials** or fertilizers to improve the efficiency.
- **The age of water** obtained from underground bores

IAEA
International Atomic Energy Agency

TECHNICAL REPORTS SERIES No. 423

Radiotracers for industrial applications

Radio-nuclide	Half life	Radiation (MeV)	Daughter nuclide	Chemical forms
^{41}Ar	1.8 h	γ (1.29)	^{41}K	elementar
^{82}Br	35.3 h	γ (0.78)	^{82}Kr	CH_3Br
^{113m}In	1.7 h	γ (0.39)	^{113}In	$InCl_3$
^{56}Mn	2.6 h	γ (0.85; 1.81; 2.11)	^{56}Fe	Mn(II)-acetat, $Mn(NO_3)_2$; MnO_2, Mn (elementar)
^{24}Na	15.0 h	γ (2.75)	^{24}Mg	Na-acetat, Na-carbonat; Na-salicylat, Na-naphthenat
^{82}Br	35.3 h	γ (0.78)	^{82}Kr	NH_4Br, KBr; Br-benzol, Tetra-Br-benzol
^{140}La	40.3 h	γ (1.60)	^{140}Ce	La-napthenat; La_2O_3

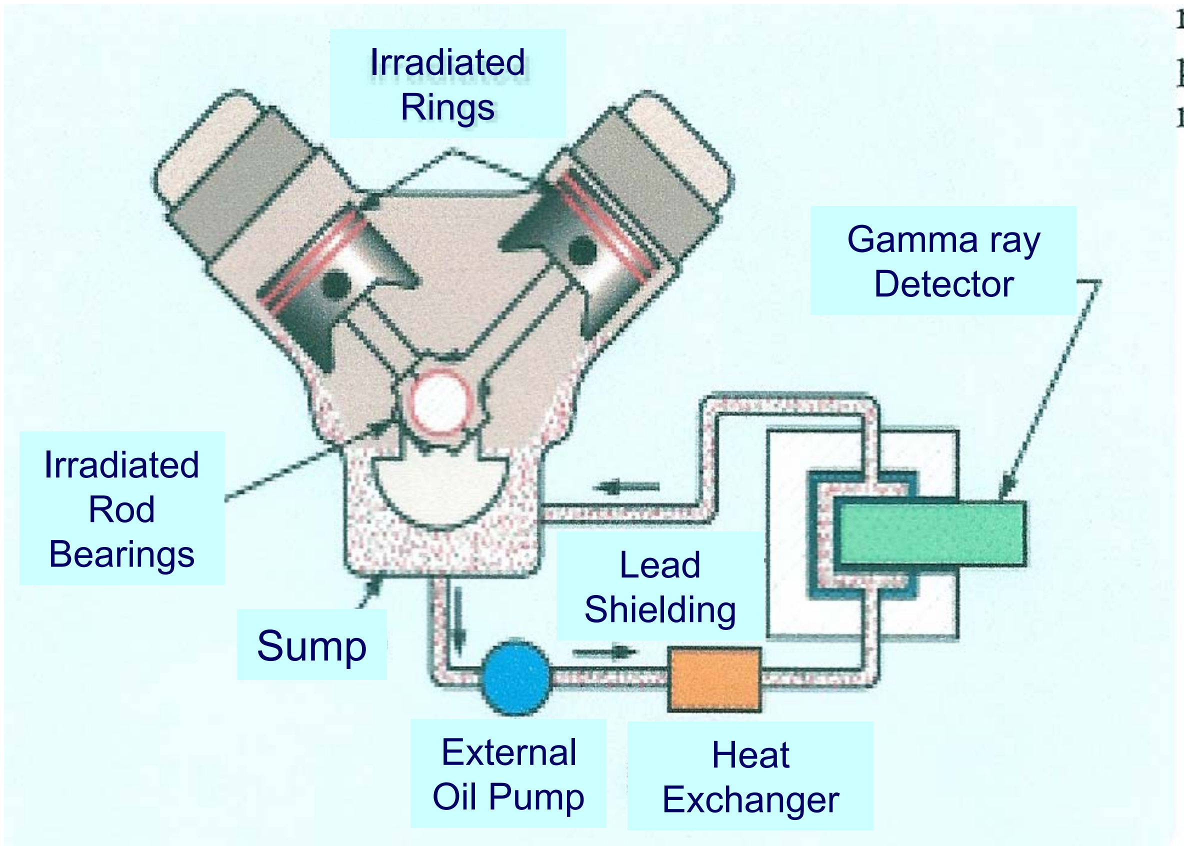
Wear studies in Automobile Industry Using Radioactive Tracers

Bulk Activation
Involves irradiating the entire part by exposing it to **thermal neutrons** in a nuclear reactor to produce suitable radio-isotopes

Surface or Thin Layer Activation
Involves irradiating a thin layer of atoms in the surface of the target part, perhaps 10 to 150 micrometers deep, by bombarding it with a **high energy beam of charged particles**.

Tritiation
Involves replacing some of the hydrogen atoms in lube oil with radioactive tritium atoms (3H) through catalytic exchange, and measuring by means of liquid scintillation counting. Tritium tracing is a tool for measuring engine oil consumption accurately over short periods of operation.

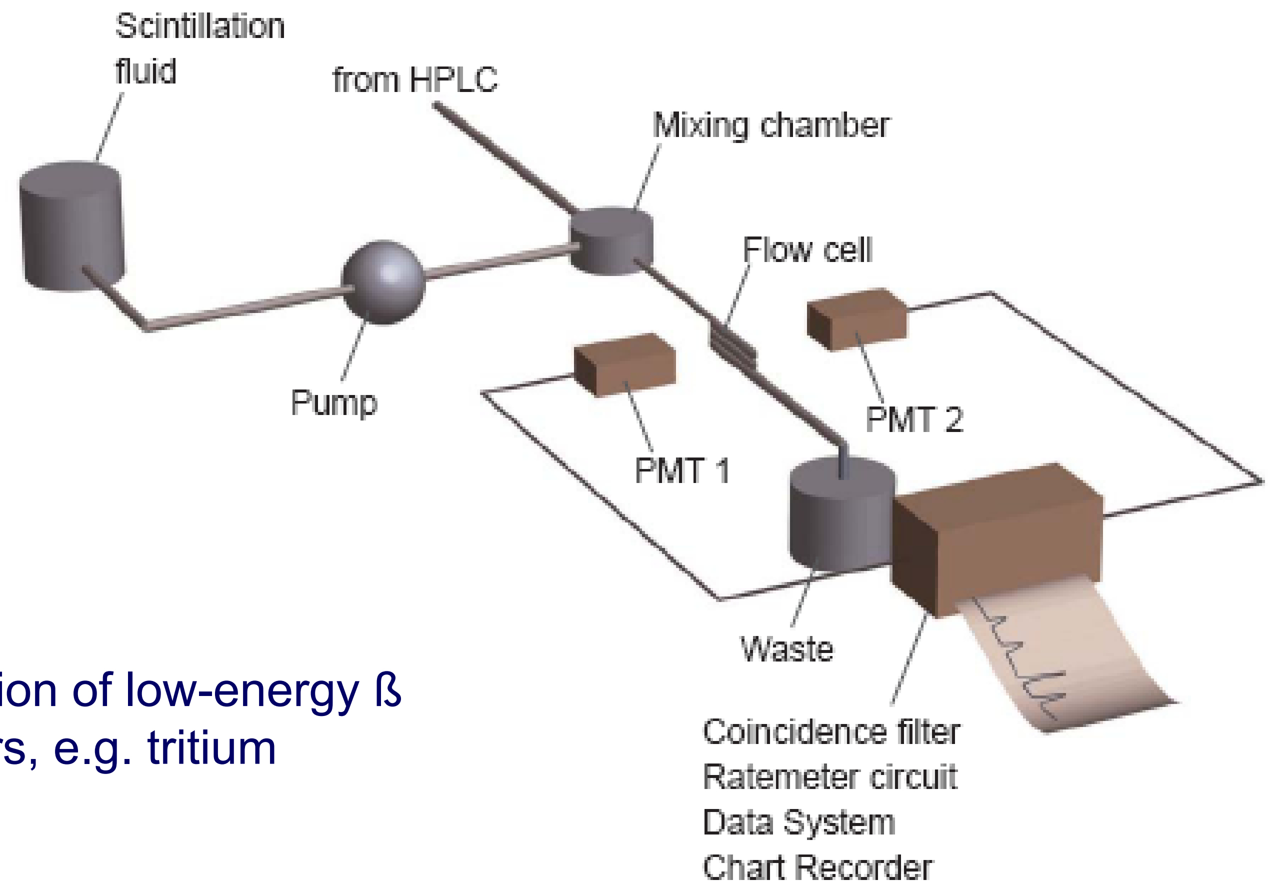
Experimental Set-Up for Motor Wear Studies



Liquid scintillator flow detector system

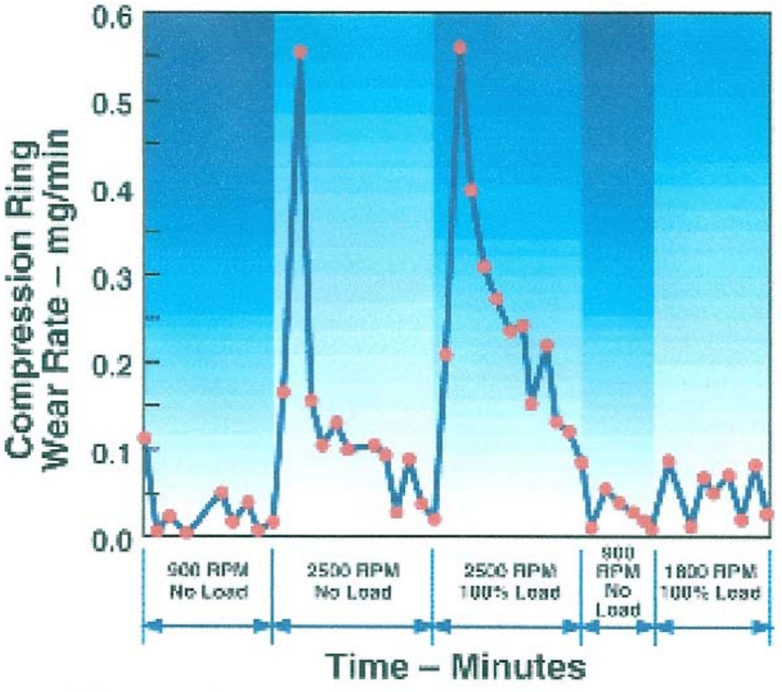
Liquid solutions of one or more organic scintillators in an organic solvent :

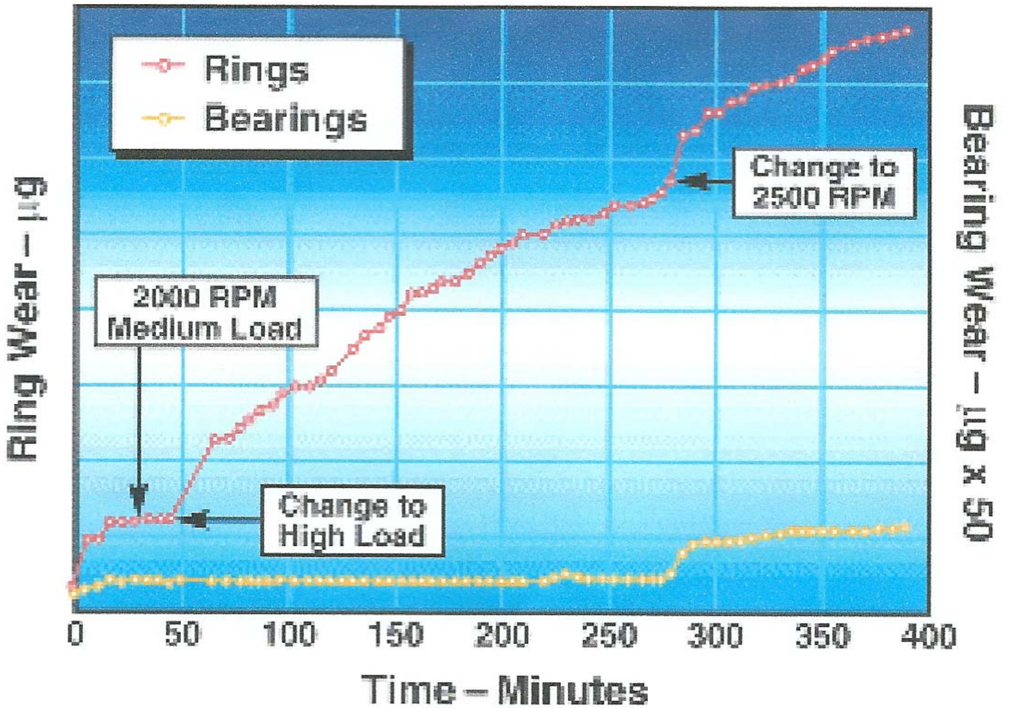
The typical solutes are fluors such as ($C_{18}H_{14}$), ($C_{20}H_{14}N_{2}O$), ($C_{24}H_{22}N_{2}O$), .
The most widely used solvents are toluene , xylene , benzene



Detection of low-energy ß emitters, e.g. tritium

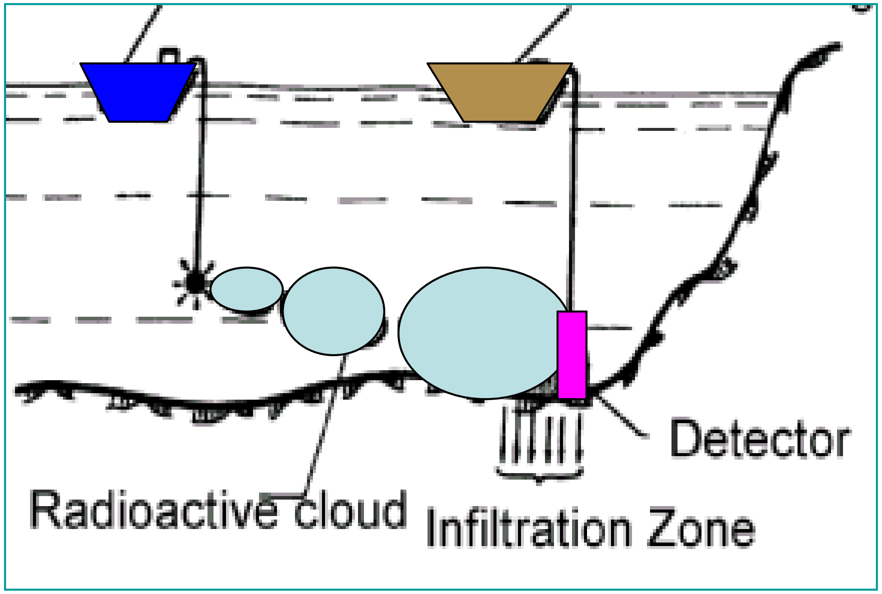
Some Results of Radio-Tracer Motor Wear Studies





Radio-Tracer Leak Detection

Dam seepage



METHODOLGY

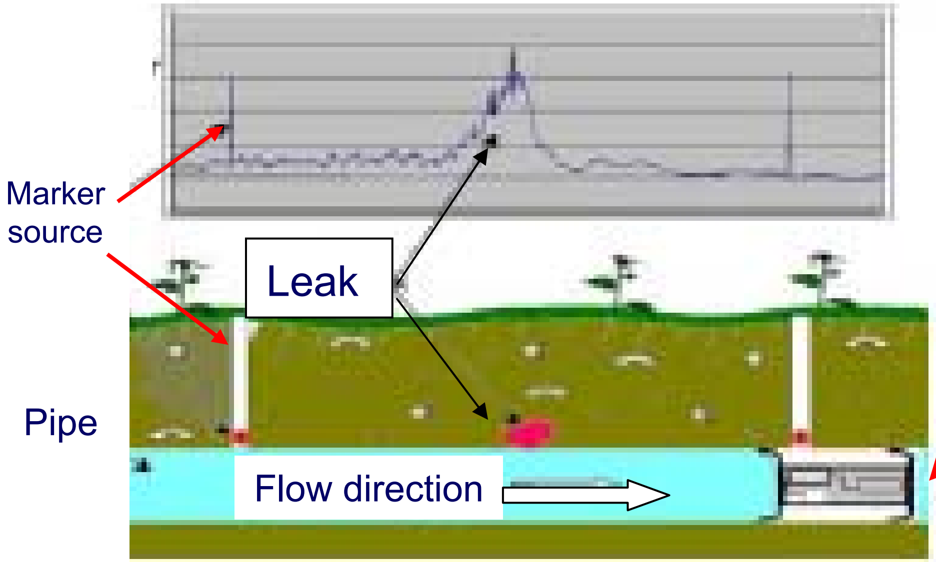
Gamma emitting radioisotope [e.g., ^{82}Br ($T_{1/2}$ = 35.3 hr, E_γ = 619, 1317 keV) or ^{131}I ($T_{1/2}$ = 8.02 d, E_γ = 365 keV)] injected at a point of the reservoir near to the bottom

Migration of the radioactive cloud is monitored using submerged scintillation detectors suspended from boats

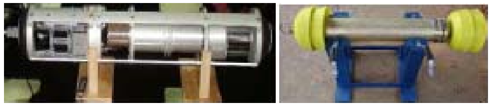
When the cloud reaches the infiltration zone, the tracer disappears after a certain period of time

Radio-Tracers in the Oil Industry

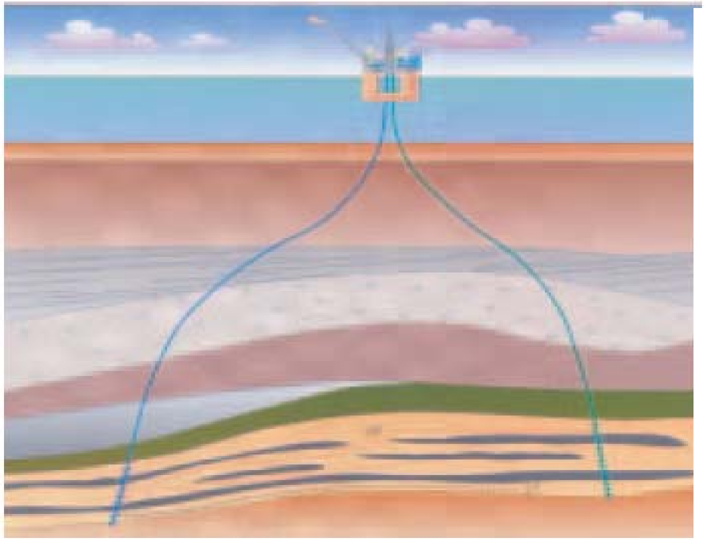
Leak Detection in underground pipelines



Traveling Detection „Pig“



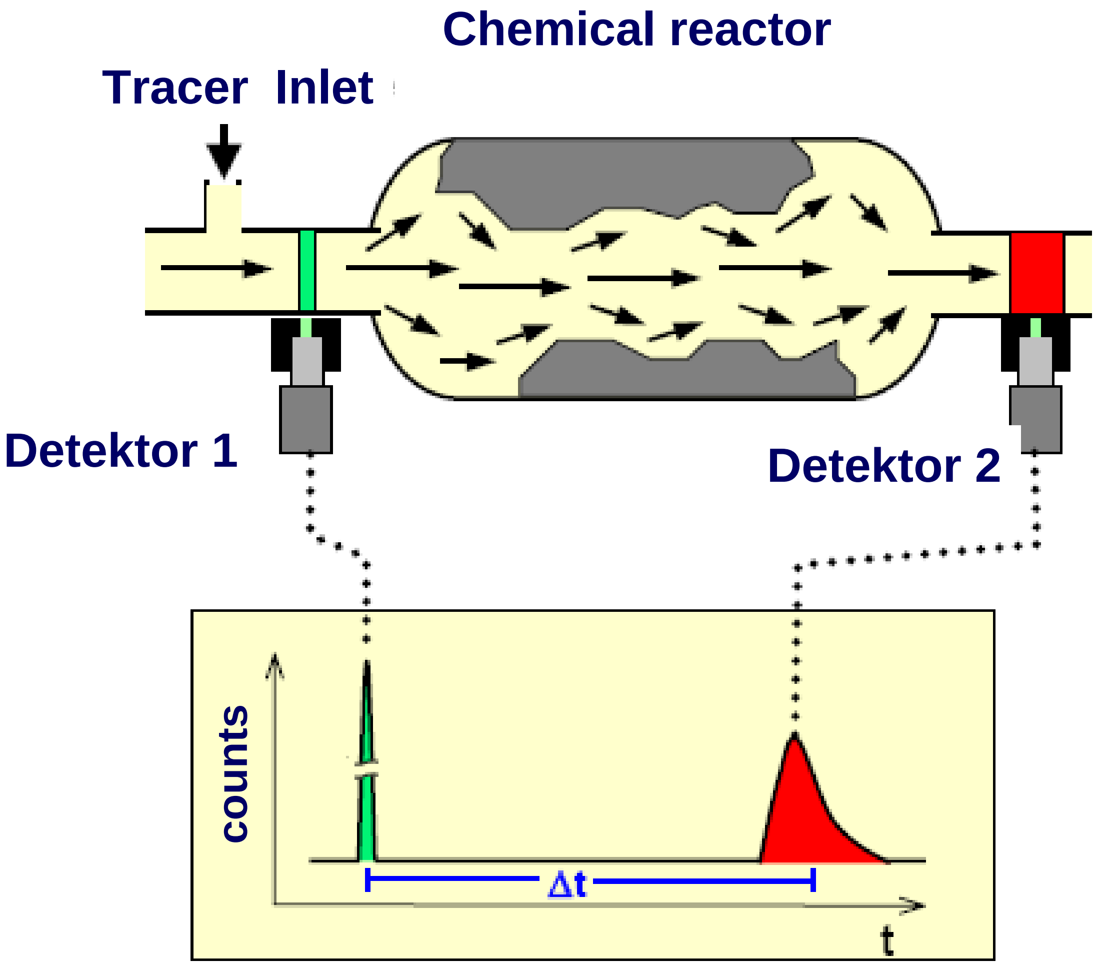
Flow Detection in layered Oil Reservoirs



Procedure

- Injection of a suitable tracer
- Injection of a cleaning fluid
- Passage of the detection „Pig“

Tracers in the chemical industry

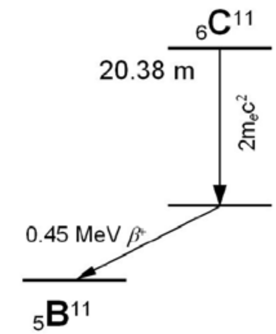


Measurement of

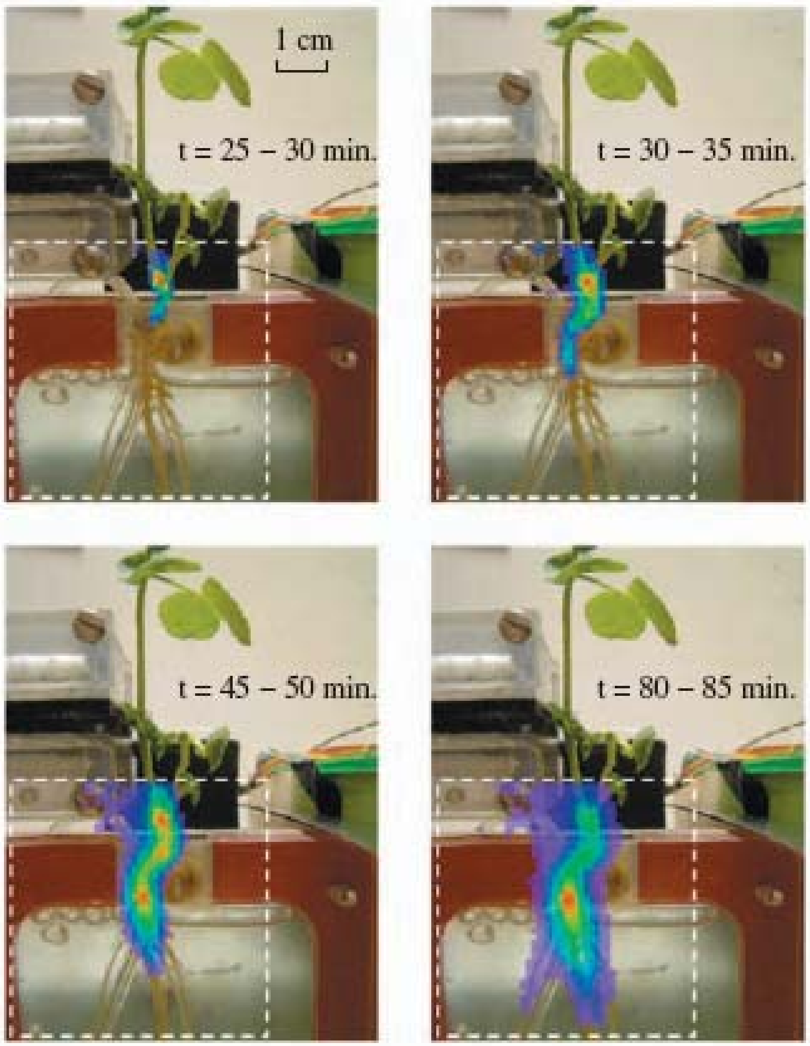
- Flow rates
- Average dwell time
- Dwell time distribution

- Flow volume
- Flow characterisitics
- Reflux
- Short-circuit currents
- Dead zones

Radiotracer applications in biological research



Production of energy-storing monosaccharides by photosynthesis in the leaves of plants



Accumulation of 11C-labeled monosaccharides in the lower shoot and root of a bean plant as a function of time

In this experiment a leaf was sealed in a cuvette into which air with a mixture of $^{12}CO_2$ and $^{11}CO_2$ flowed.

The ^{11}C up-take and the formation and distribution of monosaccharides was observed as a function of time by positron emission spectroscopy

Radiotracer used in biological research

Radioisotopes: Decay Products and Energies			
Isotope	**Emission**	**Energy (MeV)**	**Half Life ($t_{1/2}$)**
Tritium (^{3}H)	β	0.019	12.3 yrs
Carbon (^{14}C)	β	0.156	5730 yrs
Sulfur (^{35}S)	β	0.167	87.2 days
Phosphorus (^{32}P)	β	1.710	14.3 days
Phosphorus (^{33}P)	β	0.249	25.3 days
Iodine (^{125}I)	γ	0.178	59.9 days

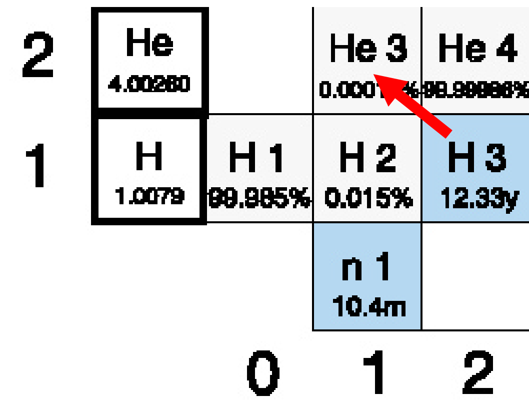
Tritium – an important tracer in biology, chemistry, hydrology, oceanography, …..

Tritium is a radioactive isotope of hydrogen,
and therefore an ideal tracer for water movement.

ß- decay of tritium

$$^{3}_{1}T \Rightarrow {}^{3}_{2}He^{+} + \beta^{-} + \bar{\nu} \qquad T_{1/2} = 12.32\ y$$

$0 \leq E_{\beta} \leq 18.6$ MeV



Natural source of tritium:

Reaction of nitrogen with cosmic ray neutrons in the atmosphere

$$^{14}_{7}N + {}^{1}_{0}n \rightarrow {}^{3}_{1}T + {}^{12}_{6}C$$

Oxidation to **HTO (tritiated water)** and precipitation as rain

Before 1954: concentration of **1-10 T per 10^{18} H atoms**

Total amount on earth: 1.8 kg

Man-made Tritium

$$^{6}_{3}Li + n \rightarrow {}^{4}_{2}He\ (2.05 MeV) + {}^{3}_{1}T\ (2.75 MeV)$$

Exothermic reaction,
used for large scale production

$$^{10}_{5}B + n \rightarrow 2\,x\ {}^{4}_{2}He + {}^{3}_{1}T$$

Boron control rods in reactors

$$^{2}_{1}D + n \rightarrow {}^{3}_{1}T$$

In heavy water reactors

Nuclear Fission of ^{235}U, ^{233}U, ^{239}Pu

Fusion reactions

$$^{3}_{1}T + {}^{2}_{1}D \rightarrow {}^{4}_{3}He + {}^{1}_{0}n + 18\ MeV$$

$$^{2}_{1}D + {}^{2}_{1}D \rightarrow {}^{3}_{1}T + {}^{1}_{1}H + 4.03\,MeV$$

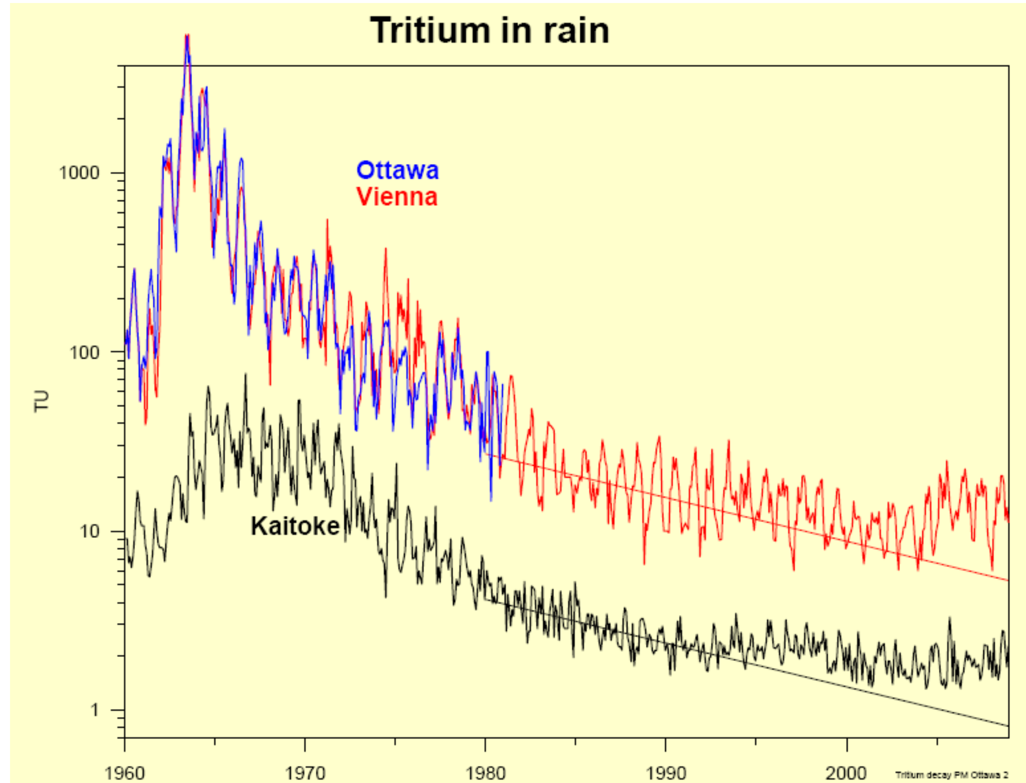
Tritium production in thermonuclear („hydrogen") bombs

^{235}U —Fission→ Kr + Ba + 2n

$^{6}Li^{2}D$ —Fission of ^{6}Li→ $^{3}T + ^{2}D + ^{4}He$

—Fusion→ ^{4}He + n + 17.6 MeV

^{14}N → $^{12}C + ^{3}T$



Total production between 1951 and 1976: 6.5 GCi = 690 kg

Applications of tritium tracers

Biological research and chemical reactions

Tritium is used as a hydrogen tracer and as a molecular label in studies of metabolism, biosynthesis, and cytology.
Tritiated thymidine and other nucleotides and nucleo-sides have been extensively used in studies of the formation of DNA and RNA

Hydrological studies

Tritium is an ideal tracer for water movement. Tritium is therefore much used in hydrology and oceanography

Some studies depend on natural tritium or that introduced by weapons testing.
In other cases large amounts of tritium are deliberately added.

Investigations include the distribution of groundwater in oil fields; the tracing of springs, rivers, and lakes; water seepage and loss from reservoirs; and the movement of glaciers

Applications of tritium tracers

Biological research and chemical reactions

Tritium is used as a hydrogen tracer and as a molecular label in studies of metabolism, biosynthesis, and cytology.
Tritiated thymidine and other nucleotides and nucleo-sides have been extensively used in studies of the formation of DNA and RNA

Hydrological studies

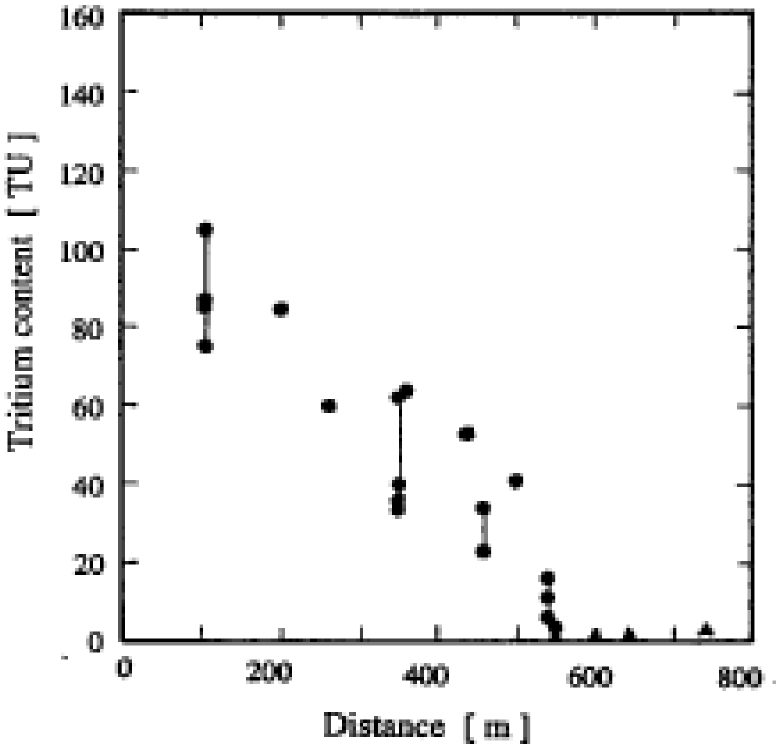
Tritium is an ideal tracer for water movement. Tritium is therefore much used in hydrology and oceanography

Some studies depend on natural tritium or that introduced by weapons testing.
In other cases large amounts of tritium are deliberately added.

Investigations include the distribution of groundwater in oil fields; the tracing of springs, rivers, and lakes; water seepage and loss from reservoirs; and the movement of glaciers

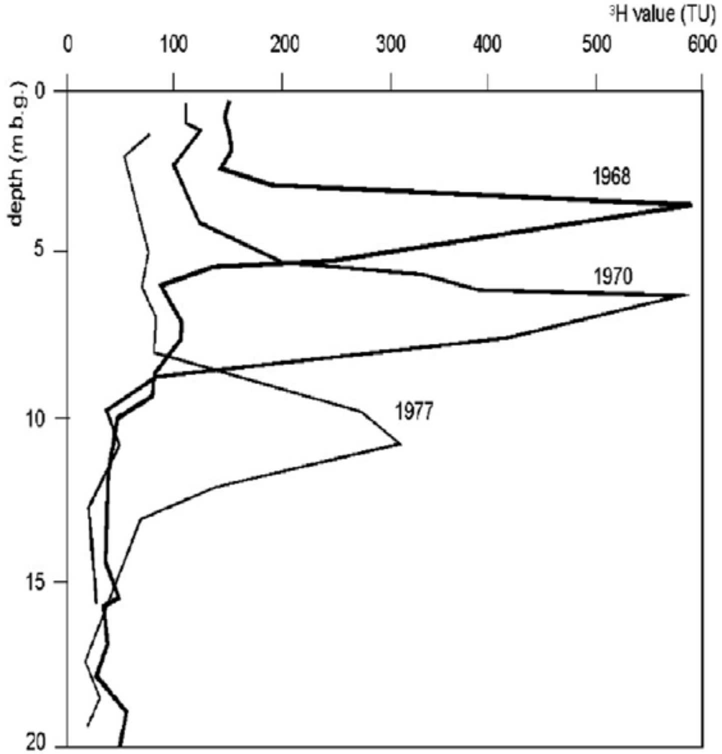
Tritium in hydrology - Examples:

Tritium in groundwater
River infiltration of the Reno River, Italy, 1973



River water penetrates ground water up to a distance of 600 m

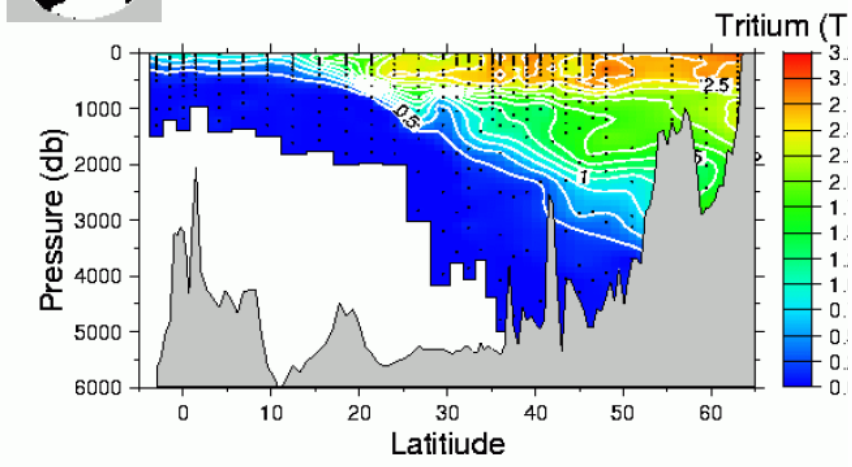
Tritium migration in unsaturated chalk, England



Determination of recharge rate

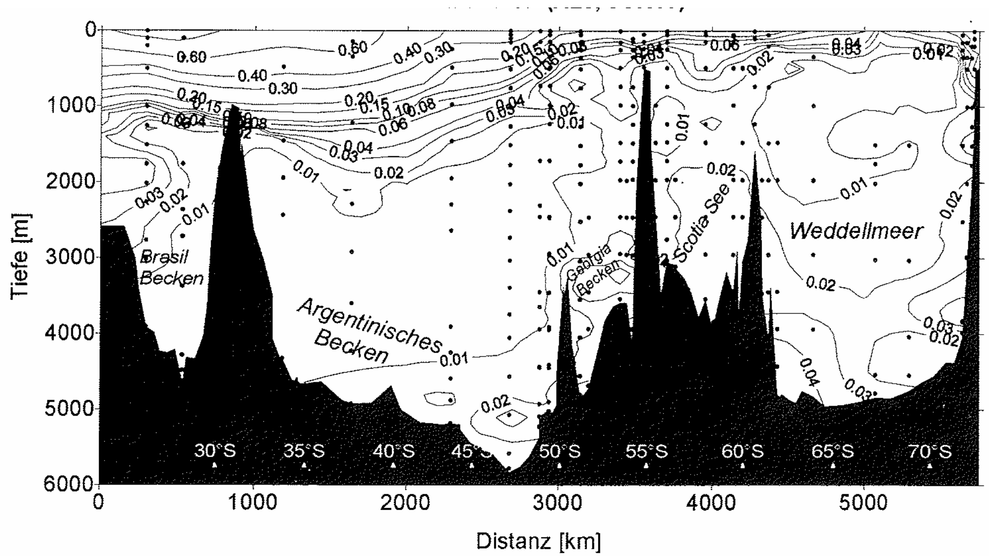
Tritium Distribution in the North Atlantic

Tritium penetration to > 3000 m depth in region of North Atlantic Deep Water (NADW) production

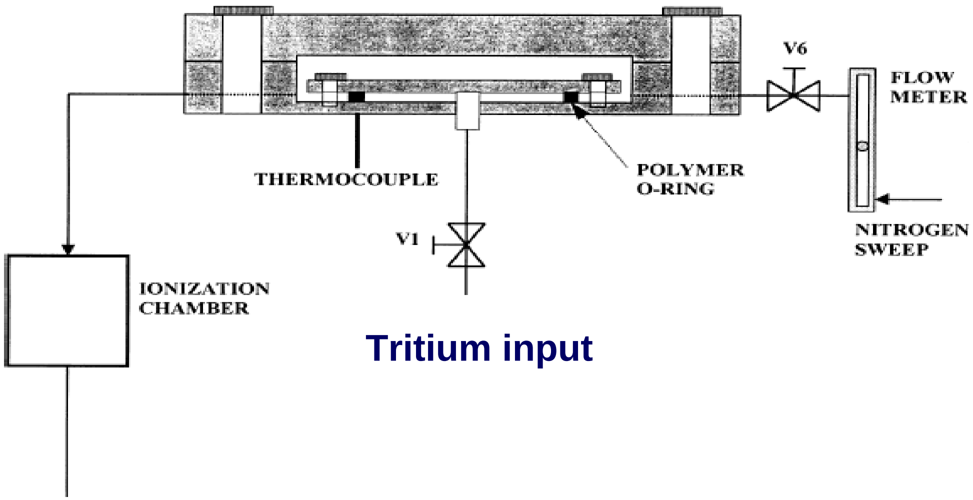


from: L-DEO Envir. Tracer Group, WOCE page (http://www.ldeo.columbia.edu/~etg/text/woce_proj.htm

Tritium Distribution in the South Antlantic

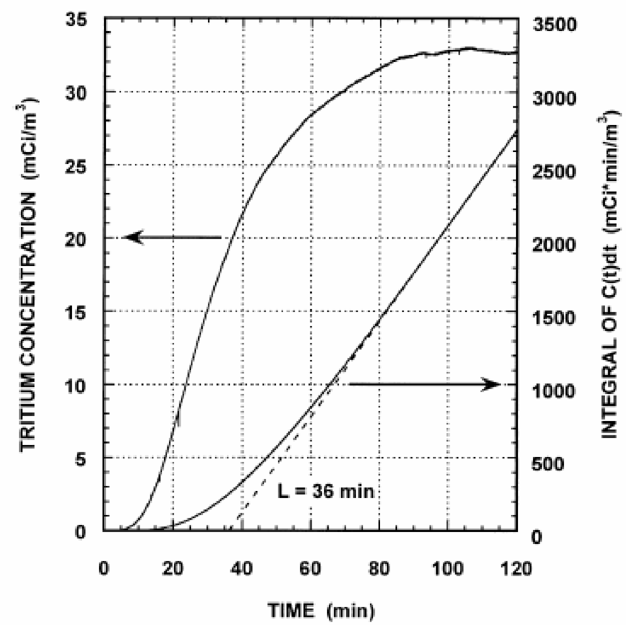


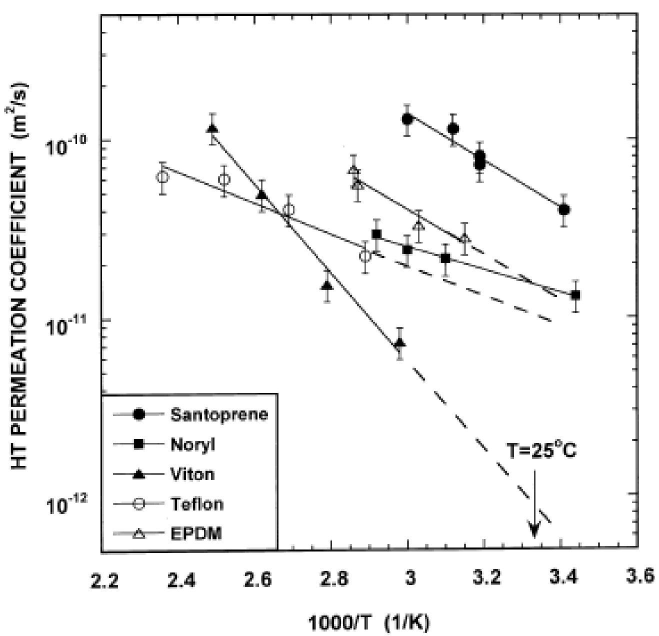
Tritium as a tracer for the measurement of hydrogen permeation in polymeric materials



Tritium input

Characteristic permeatioin curve for Noryl at 60^0C
Time interval: 1 s.





Arrhenius plot of tritium permeation for five tested polymer

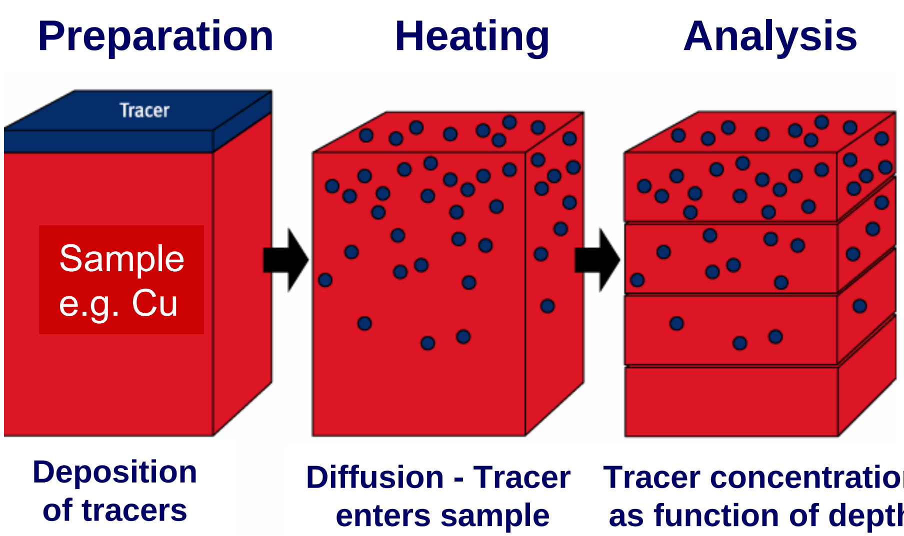
D.O. Stodilka et al. / International Journal of Hydrogen Energy 25 (2000) 1129±1136

Physics applications of Radiotracers: Studies of Self- and Impurity Diffusion

Detailed understanding of solid state diffusion (atomic motion) is of great technical interest

Examples: Doping of semiconductors with donors and acceptors
C diffusion in steel

Steps of a radiotracer study

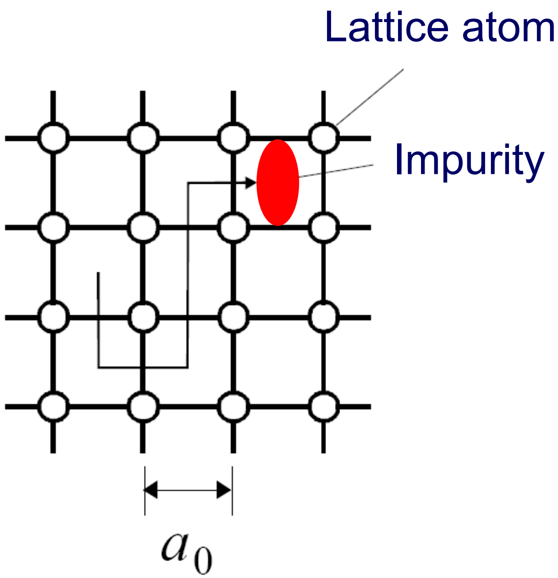


Tracer deposition:
Evaporation
Implantation

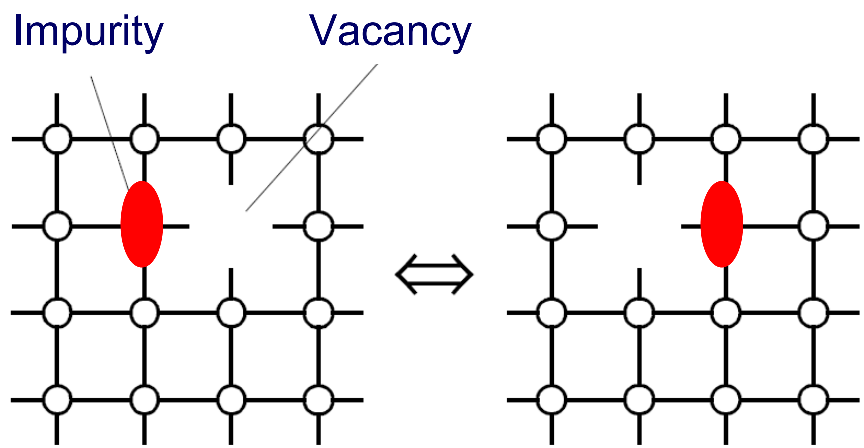
Depth profile:
Ion ablation by
sputtering

Mechanisms of Impurity Diffusion

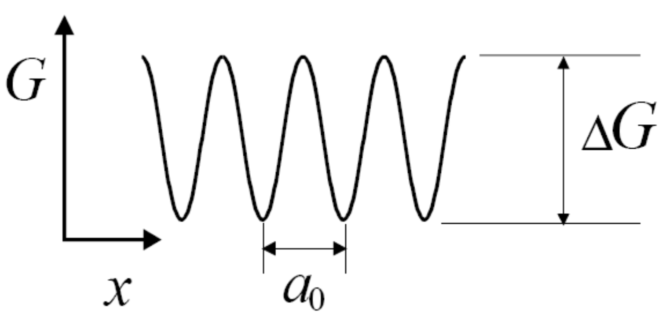
Interstitial Mechanism



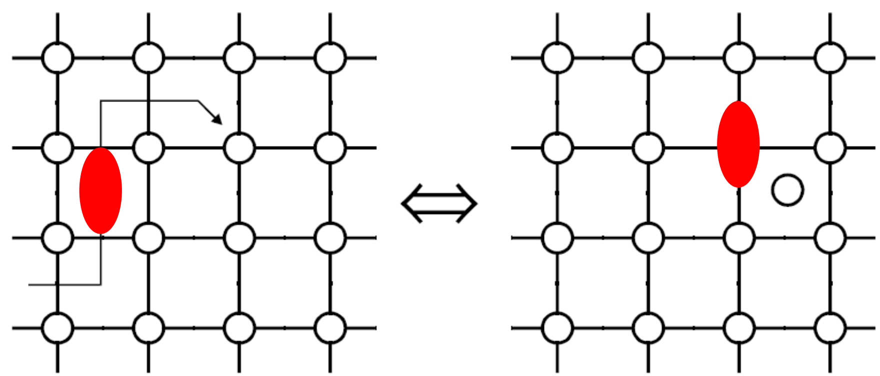
Vacancy Mechanism



Potential barriers



Replacement Mechanism

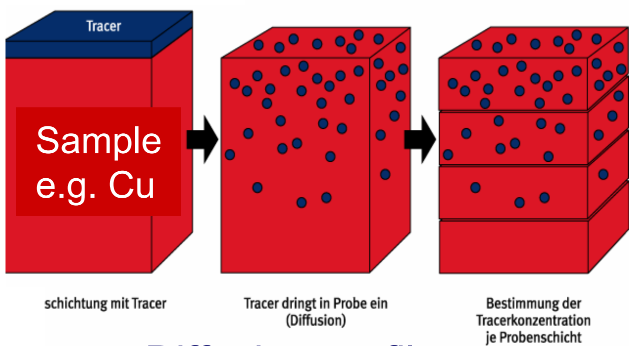


Diffusion coefficient : $D \propto \exp(-\Delta G/k_B T)$

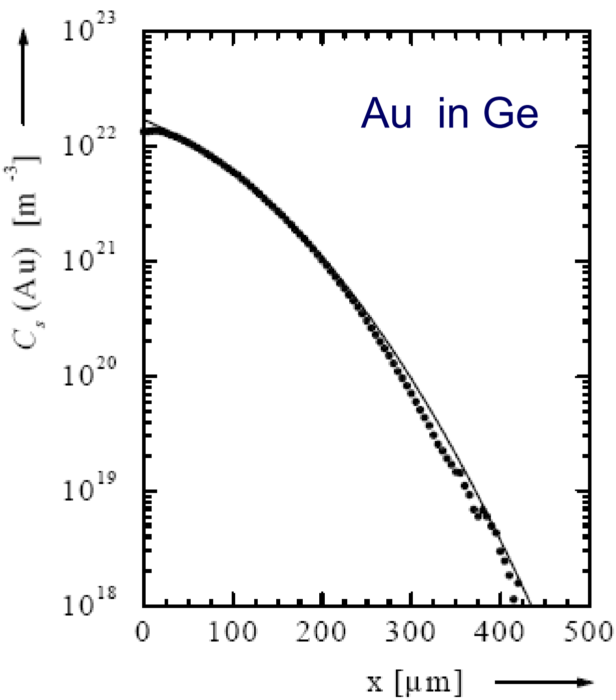
from A. Strohm, MPI Stuttgart

Diffusion studies using Radio-Tracers

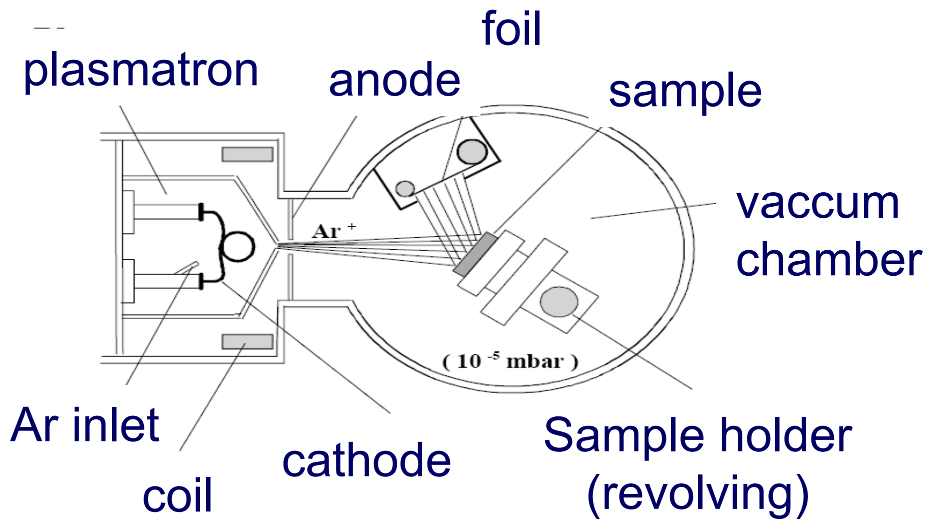
Deposition by Evaporation Heating Analysis



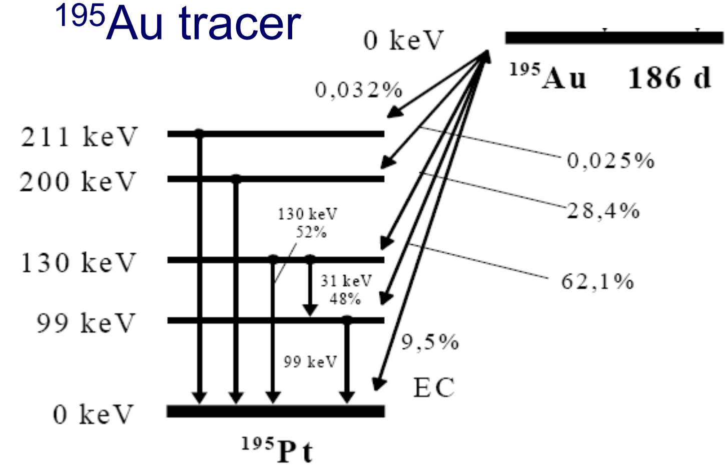
Diffusion profile
7 h @ 1000 K



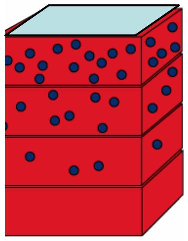
Analysis by Sputtering



^{195}Au tracer

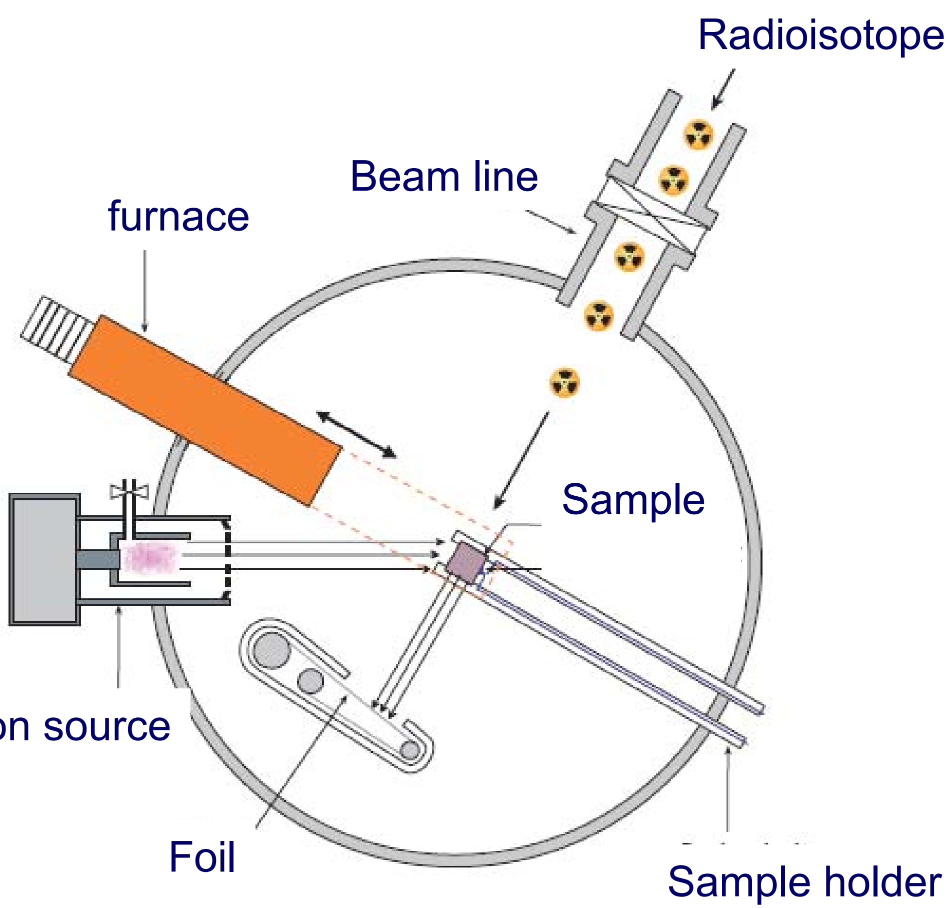


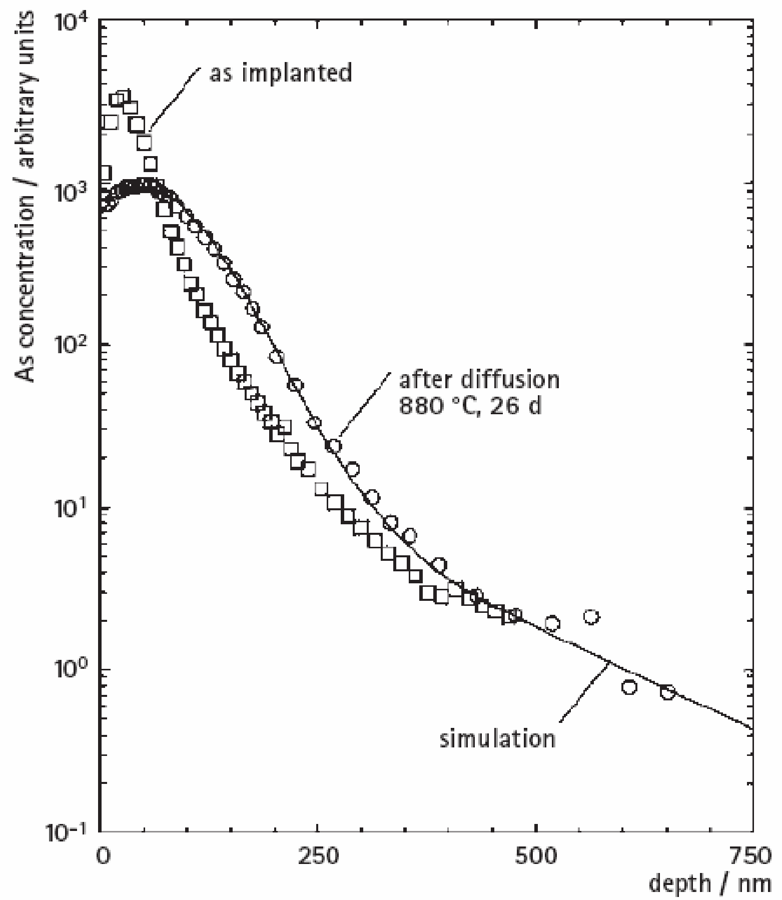
Problem with tracer deposition by evaporation: Surface barriers



Example: Al exposed to air grows a Al_2O_3 layer which modifies the diffusion results

Solution: Radio-tracer-implantation





Radio-Tracers in Medicine: Studies of Metabolism

The beginning:

George Charles de Hevesy, *Georg Karl von Hevesy*, (1 August 1885 – 5 July 1966) a Hungarian radio-chemist was the first to use radioactive isotopes in studying the metabolic processes of plants and animals, by tracing chemicals in the body by replacing part of stable isotopes with small quantities of the radioactive isotopes.

George Charles de Hevesy, received the Nobel prize in 1943 for his key role in the development of radioactive tracers to study chemical processes such as in the metabolism of animals.

Radio-tracers as instruments of medical diagnosis

Radio-tracers primarily show the physiological function of an organ as opposed to **traditional anatomical imaging** such as CT or MRI.

Radio-tracer studies are organ- or tissue-specific !!!

An example: A renogram

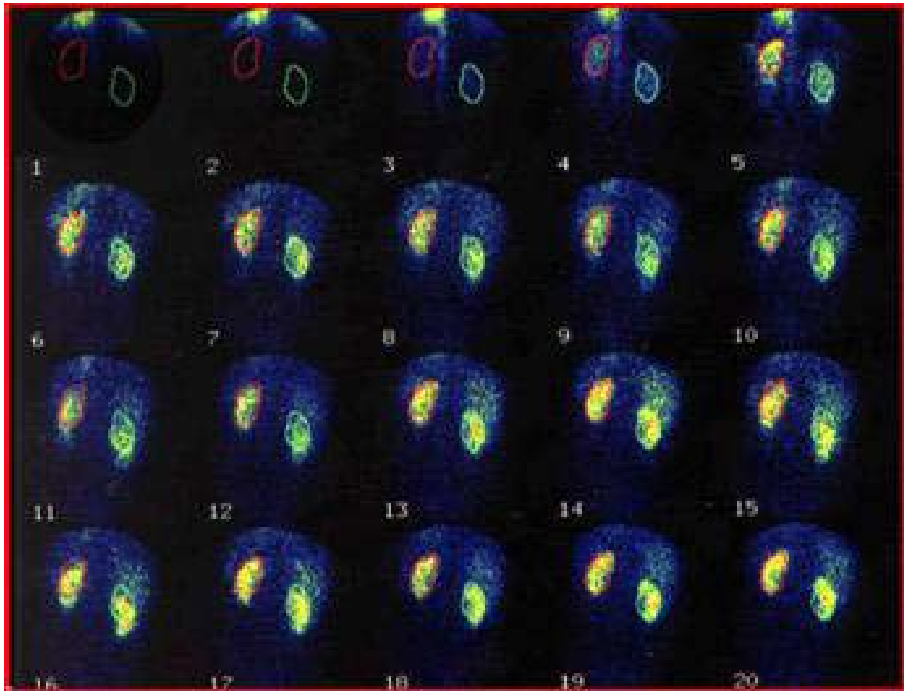
The tracer is injected into the patient.

The radioactive material is removed from the bloodstream by the kidneys.

Within a few minutes of the injection, the radiation is concentrated in the kidneys.

After 10 – 15 minutes, almost all of the radiation should be in the bladder.

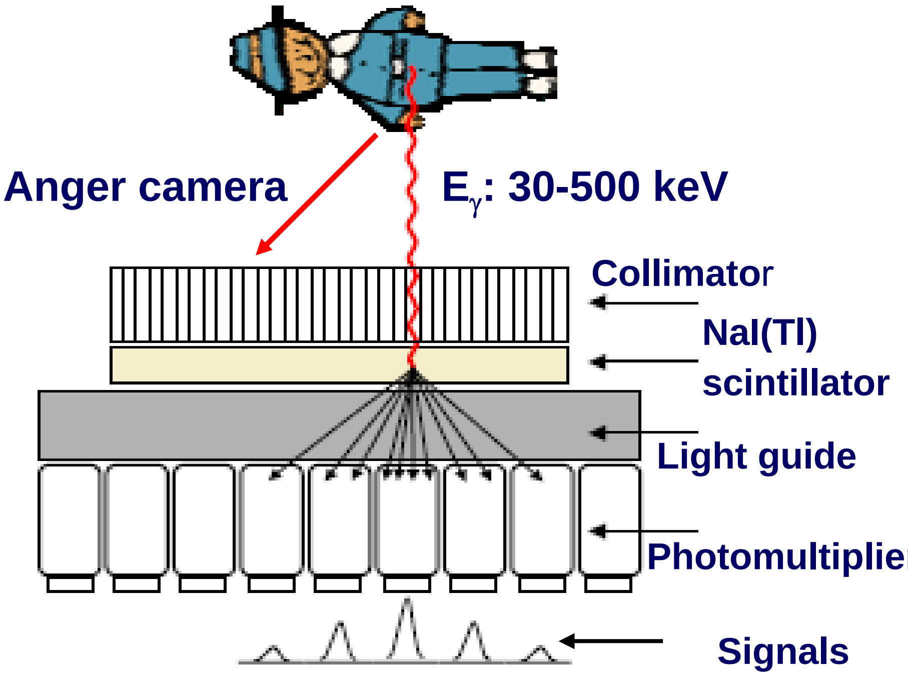
The gamma camera takes readings every few seconds for 20 minutes.

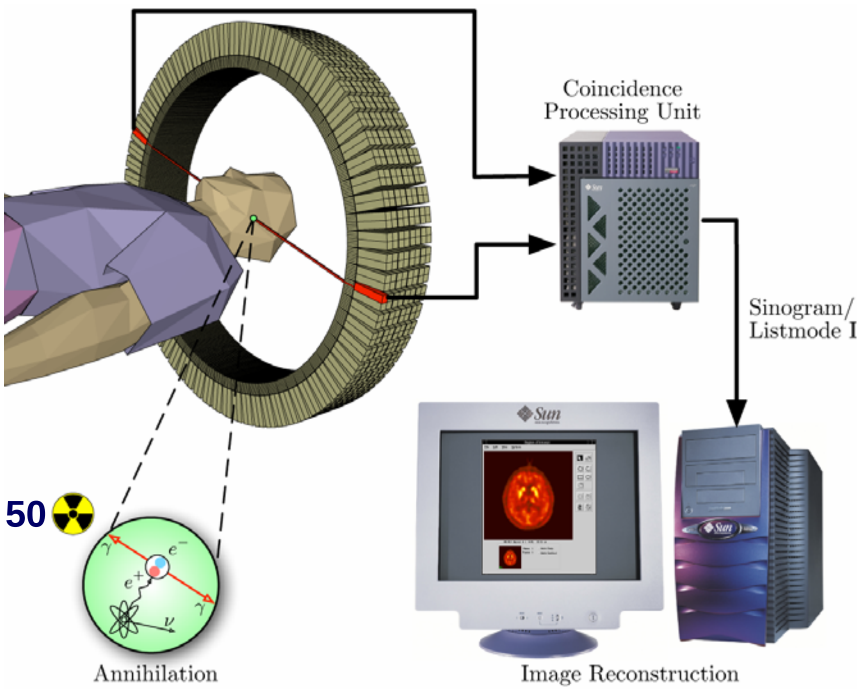


Radio-tracer diagnostic techniques

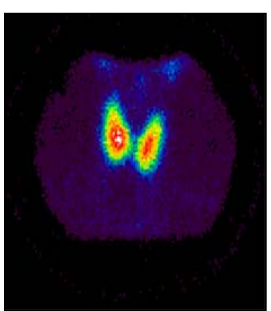
SPECT - Single-photon emission computed tomography

PET – Positron emission tomography





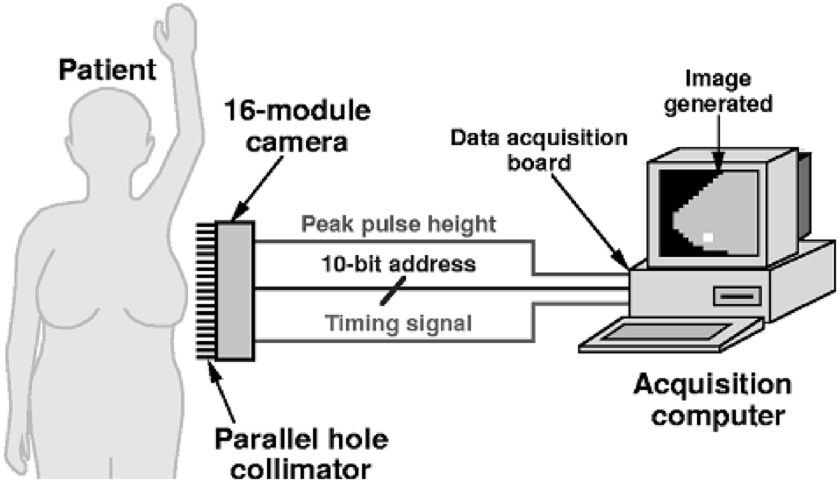
Thyroid scintigram



Less expensive

Higher resolution

Development of Compact Gamma Cameras



Design approaches:

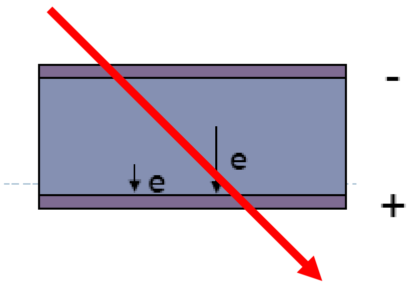
Discrete scintillator/photodiode cameras wherein the gamma rays interact in an array of optically isolated scintillation crystals coupled 1-to-1 to an array of solid-state photodetectors

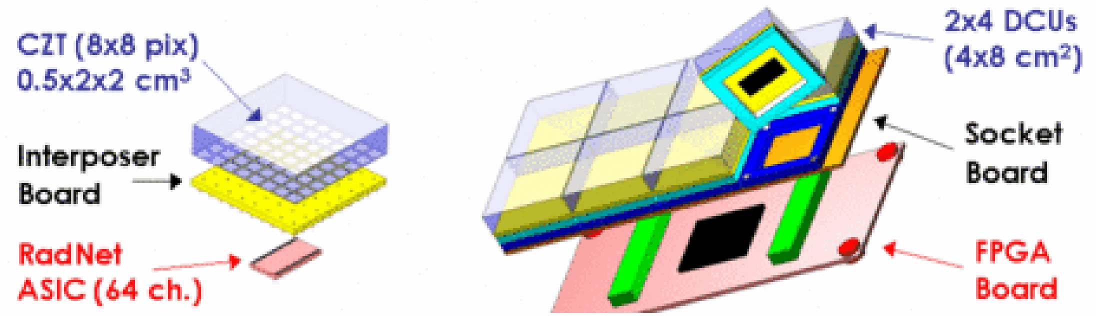
Solid-state cameras where the gamma rays interact directly with a pixellated solid-state detector such as CdZnTe (CZT)

Position-sensitive photomultiplier tube (PSPMT) cameras where the gamma rays interact in one or more scintillation crystals which are subsequently read out by a single PSPMT

Cadmium-Zink-Telluride (CZT) Semiconductor Camera

- Direct detection of gamma quanta
- Low noise
- Excellent energy resolution





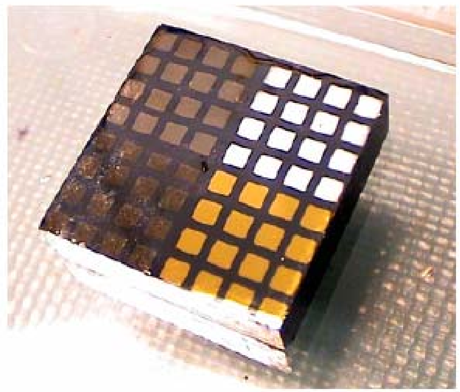
Cadmium-Zinc-Telluride CZT:

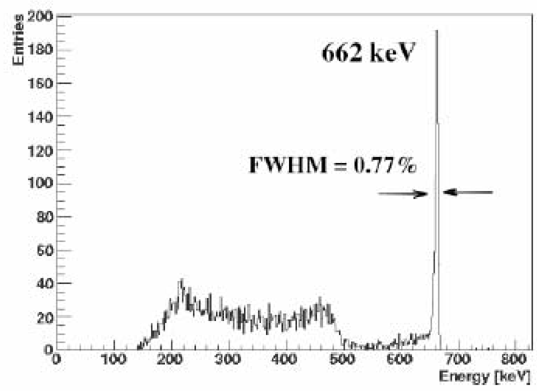
- High atomic number

  7 mm CZT = 1 cm NaI (at 140keV)
- Room temperature
- Energy resolution 6%

$E_g = 1.57$ eV

Individual pixels, size: 330µm ... 5mm

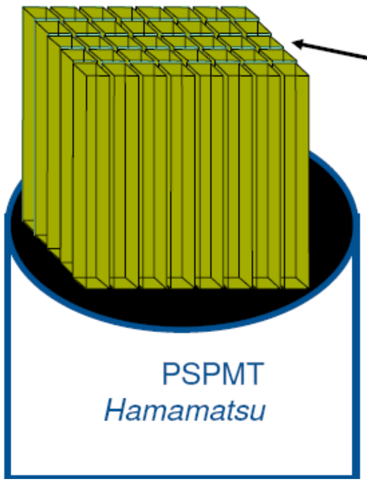




Orbotech Detector:

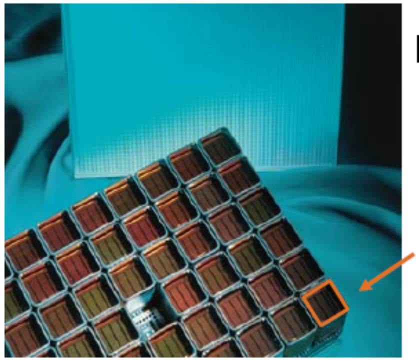
- 0.5×2×2 cm³, 64 In-Pixel (Pitch: 2.5 mm), Au-cathode.

Position-sensitive photomultiplier tube (PSPMT)/scintillator camera



I. Ziegler, TUM

Matrix of compact PSPMT

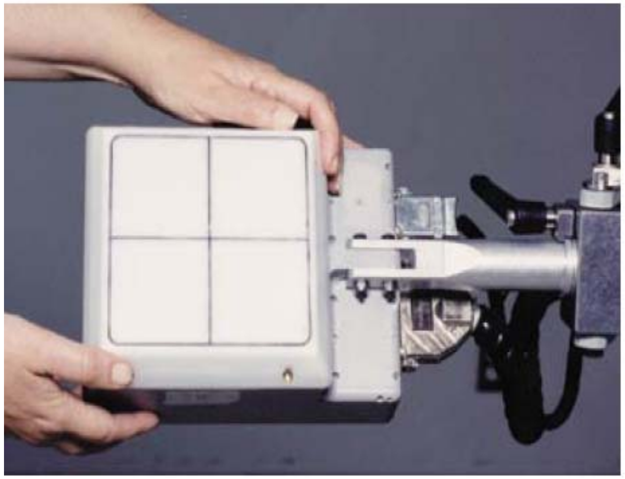


NaI(Tl) pixel: 2 x 2 x 6 mm

2,8 x 2,8 cm^2 x 2,7 cm
4 X and 4 Y wires
(Hamamatsu)

Intrinsic: < 2 mm; $\Delta E/E$(140 keV): 18%

Compact detector head



Patt et al., Majewski et al., LumaGEM, Dilon

„Single-photon“ radionuclides used in nuclear medicine

• Tc99m	140.5 keV	6.03 hours
• I-131	364, 637 keV	8.06 days
• I-123	159 keV	13.0 hours
• I-125	35 keV	60.2 days
• In-111	172, 247 keV	2.81 days
• Th-201	~70, 167 keV	3.044 days
• Ga-67	93, 185, 300 keV	3.25 days

^{99m}Technetium
The most widely used radioactive tracer isotope in Nuclear Medicine

Technetium (Tc) is one of 2 elements without a stable isotope (The other one is promethium). **$T_{1/2}$= 2x10^5 yr**

In 1871 D.I. Mendelejew predicted the existence of Tc from the systematics of his Periodic System (as Eka-Mangan)

First production in 1937 by E.G. Segrè and C. Perrier by bombarding a sample of Molybdenum for **several months** with deuterons:

$$^{98}_{42}\mathrm{Mo} + {}^{2}_{1}\mathrm{H} \Rightarrow {}^{99}_{43}\mathrm{Tc} + \mathrm{n}$$

Larger quantities of Tc can be obtained as a fission product
In nuclear reactors

$$^{235}_{92}\mathrm{U} + \mathrm{n} \Rightarrow {}^{236}_{92}\mathrm{U} \rightarrow {}^{132}_{49}\mathrm{In} + {}^{99}_{43}\mathrm{Tc} + 5\mathrm{n}$$

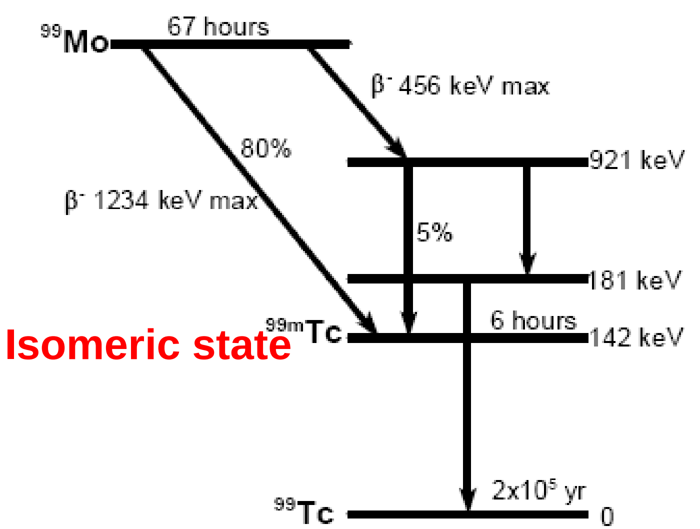
Chemical separation and reduction with H_2 gives Tc in metallic form

Properties and production of ^{99m}Tc

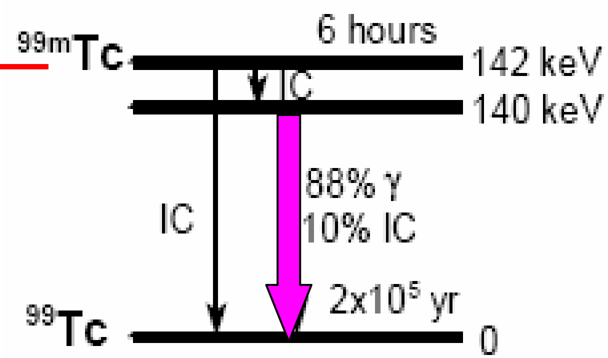
^{99m}Tc: almost pure gamma ray emission with one mono-energetic gamma ray. This makes ^{99m}Tc an extremely useful radionuclide for diagnostic in nuclear medicine

Thermal neutron capture in ^{98}Mo (Abundance=24.13 %): **^{98}Mo (n,γ) 99Mo**

The decay of ^{99}Mo

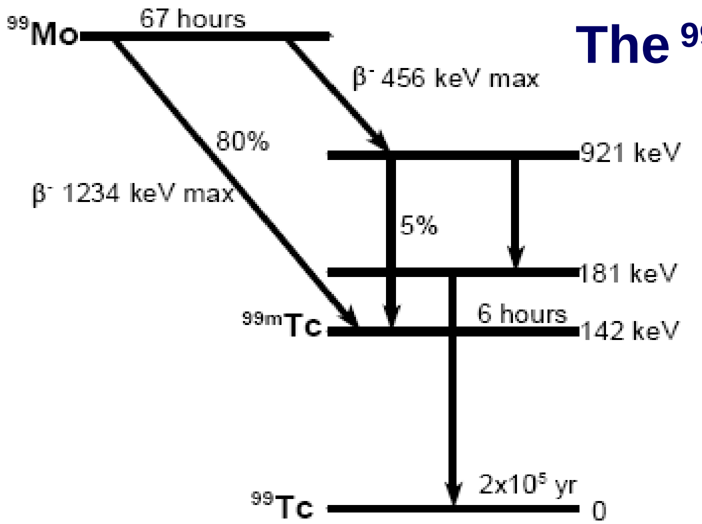


The decay of ^{99m}Tc



Monoenergetic gamma, no ß particles, short half life (6hours), radiation exposure of patients is small

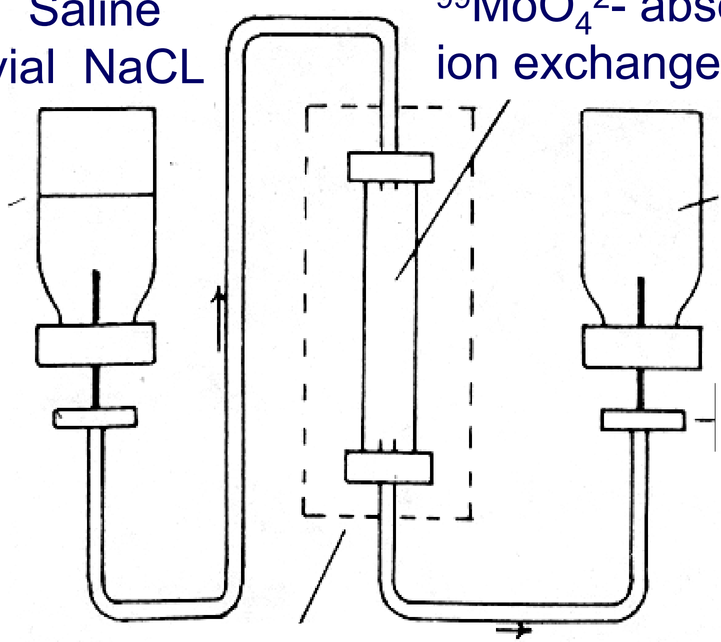
The ^{99}Mo/^{m99}Tc Generator



Tc can be separated from the parent Mo chemically, using the fact that Tc is soluble in saline solution whereas Mo is not.This is used to separate the daughter ^{99m}Tc from the parents isotope ^{99}Mo

Saline vial NaCL

$^{99}MoO_4^{2-}$ absorbed onto Al_2O_3 ion exchange column



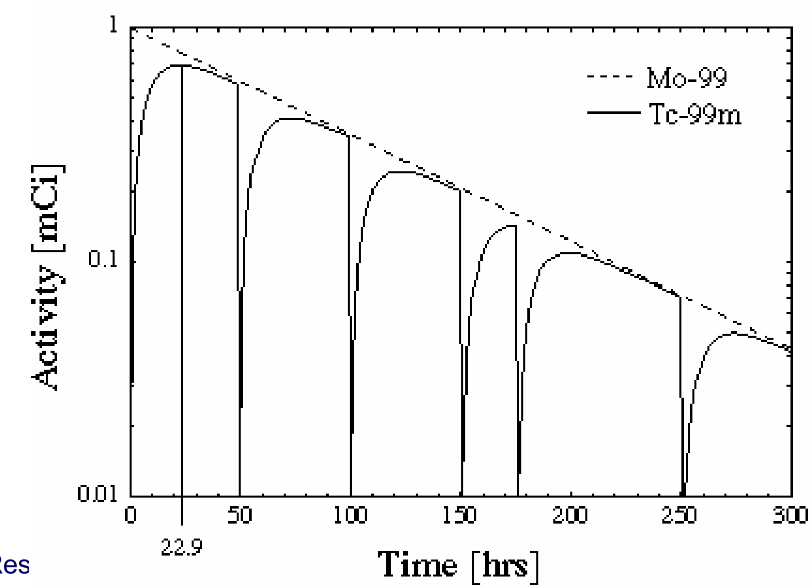
Evacuated vial

Na$^{99m}TcO_4$

Filter

Lead Shield

Multiple ^{99m}Tc elution



^{99m}Tc radiopharmaceuticals used in medical imaging

The radioactive technetium can be chelated to a number of different compounds to create specific radiopharmaceutical and optimise imaging of various structures:

^{99m}Tc **sulfur colloid:**
splenic and hepatic imaging taken up by the spleen, Kupffer cells in the liver and a small proportion by bone marrow

^{99m}Tc pertechnetate: **thyroid imaging**

^{99m}Tc teboroxime: **cardiac imaging**

^{99m}Tc labelled red blood cells; assessment of **occult gastrointestinal haemorrhage and vascular lesions**

^{99m}Tc incorporation into monoclonal antibody, **an immune system protein capable of binding to cancer cells**. Higher γ-ray intensities indicate where the tumor is.

This technique is particularly useful for detecting hard-to-find cancers, such as those affecting the intestine. **Immunoscintigraphy**

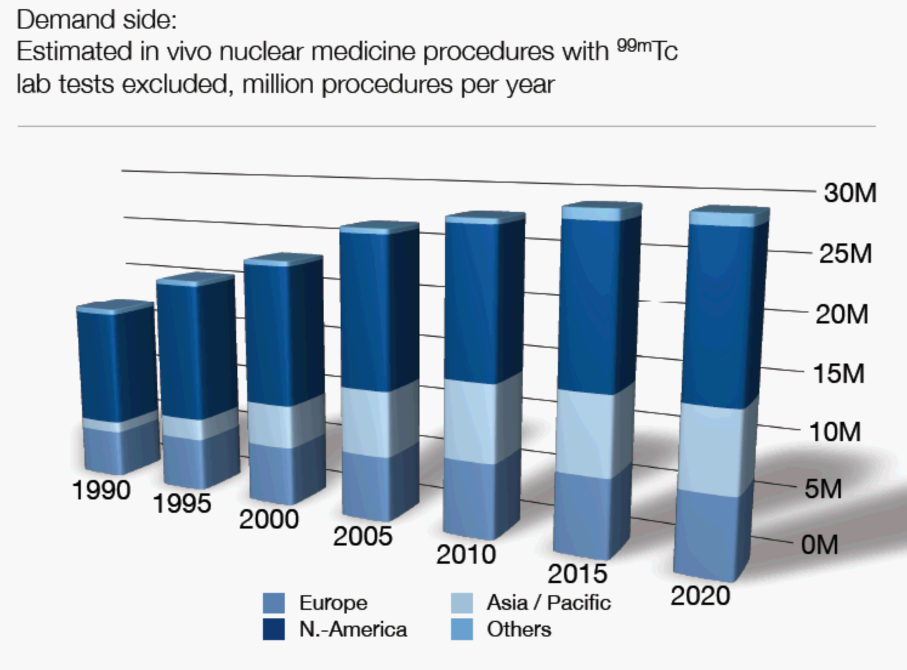
The global consumption of 99Mo/99mTc

- **99mTc** (**T1/2= 6 hours**) remains the **most widely used** radioisotope for diagnostic in nuclear medicine

- About **80%** of the nearly **30 million** annual radiodiagnostic procedures are carried out worldwide with this single isotope (**140 keV gamma rays**). :

- This percentage share is expected to continue to grow yearly by 3% in the near future due to its availability from the **very convenient** and **cost-effective 99Mo/99mTc generator**.

- The short half-life's of **99Mo (T1/2= 66 hours)** and its daughter **99mTc (T1/2= 6 hours)** clearly present a problem in terms of **reliable supply** since they can not be stockpiled.



- A **regular supply** of 99Mo/99mTc generators to hospitals or central radio-pharmacies is required.

The 99Mo/99mTc Generator Shortage

World needs : about 12.000 Ci 99Mo ‘6-days’ calibrated per week !!!

The Mo-99 Global Supply Chain

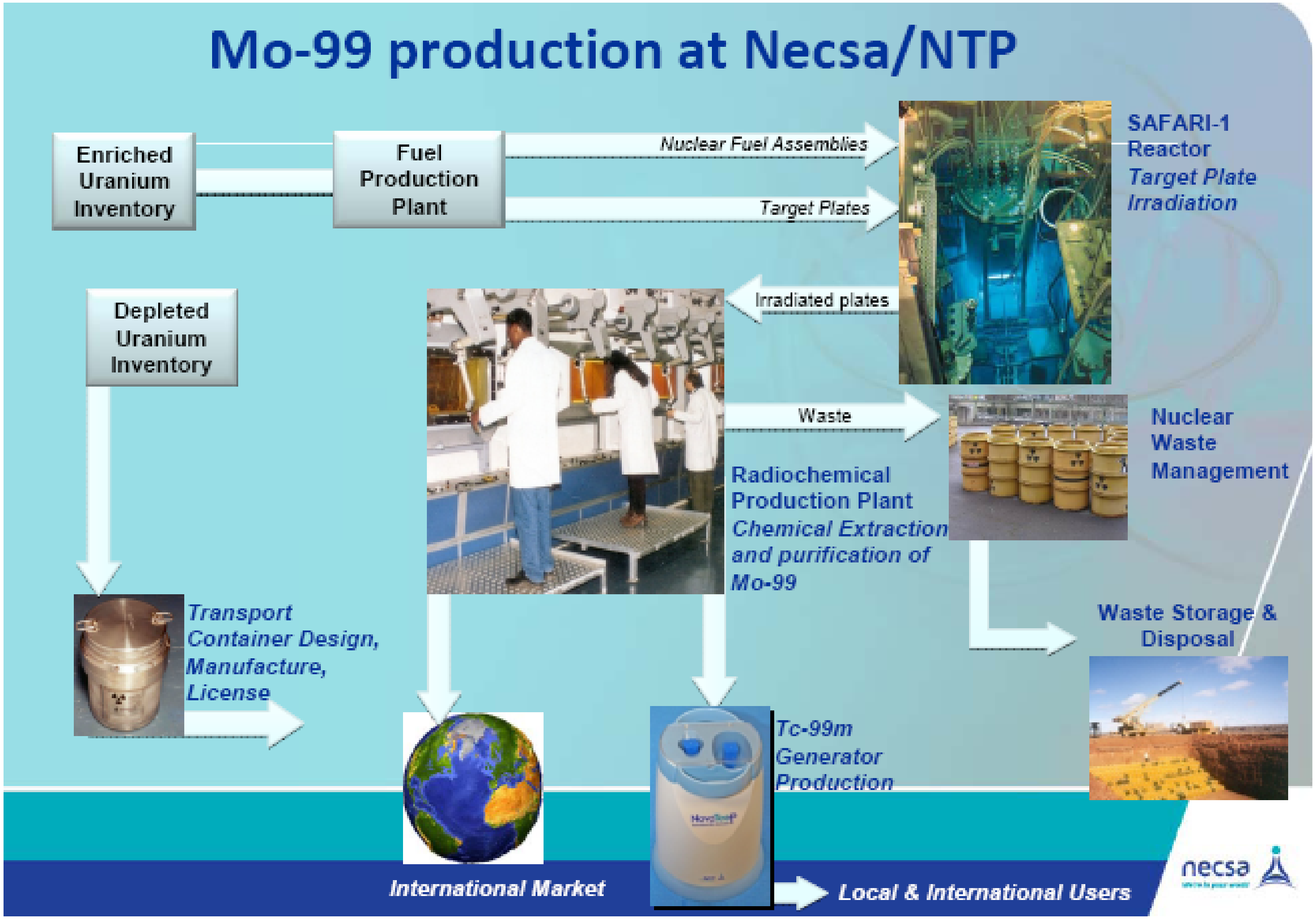
Today, only **7 major Research Reactors** are irradiating 235U targets to serve the world demand.

BR2 (Belgium)
HFR (The Netherlands)
OSIRIS (France)
NRU (Canada)
SAFARI (South Africa)
OPAL (Australia; 2009)
MARIA (Poland)

The irradiated targets are processed by **5 facilities** in the world,supplying **95%** of the bulk 99Mo.

The 99Mo/99mTc generators are manufactured on **5 main sites**.

GE HEALTHCARE (UK)
IBA CIS BIO (France)
COVIDIEN (The Netherlands)
COVIDIEN (USA)
LANTHEUS (USA


Mo-99 production at Necsa/NTP
Enriched Uranium Inventory
Fuel Production Plant
Nuclear Fuel Assemblies
Target Plates
SAFARI-1 Reactor
Target Plate Irradiation
Irradiated plates
Depleted Uranium Inventory
Waste
Nuclear Waste Management
Radiochemical Production Plant
Chemical Extraction and purification of Mo-99
Transport Container Design, Manufacture, License
Waste Storage & Disposal
Tc-99m Generator Production
International Market
Local & International Users
necsa