European Summer School 2017

Université franco-allemande Deutsch-Französische Hochschule

"Radiochemistry & Nuclear Instrumentation (Low Level Radioactivity)"

Strasbourg, France, August 21-25, 2017

Low-level neutron measurements and dosimetry

Francesco d'Errico

Scuola di Ingegneria, Università di Pisa, Pisa, Italy
Yale University School of Medicine, New Haven, CT USA

francesco.derrico@ing.unipi.it or francesco.derrico@yale.edu

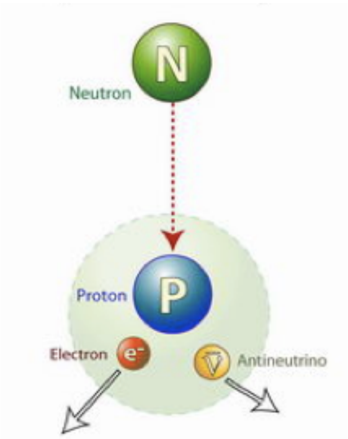
Overview

- Neutron basic concepts and natural sources
- Neutron interactions and reactions
- Neutron energy deposition in tissue
- Neutron monitoring and dosimetry
  - Moderated thermal neutron sensors
  - Tissue equivalent proportional counters
  - Superheated emulsions
- Photoneutron and SNM measurements

Neutron basic concepts and natural sources

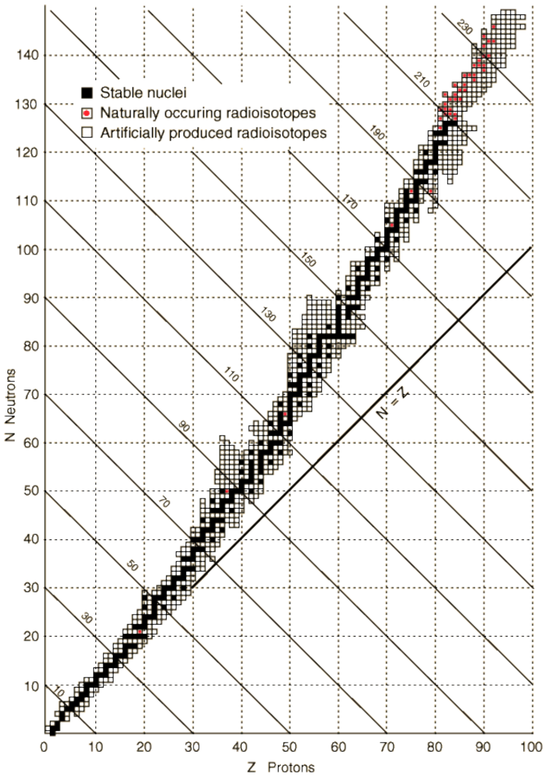
Neutrons - General Properties

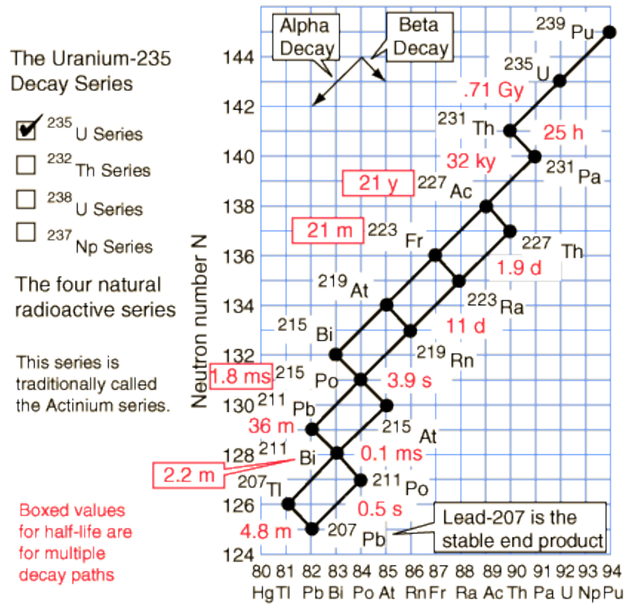
- The neutron is stable only inside the nucleus.
- A free neutron is unstable and decays β^- with a half life of about 10 min.
- 10 min is very long compared to the interaction times (in a nuclear reactor the half-life of a neutron is ~ 10^{-3} s).
- A free neutron may be considered stable, in the most important engineering applications of neutrons.



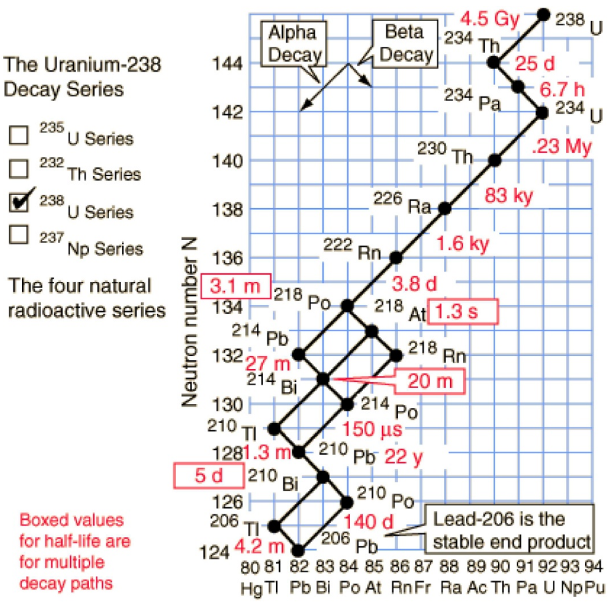
$$m_n = 939.5656 \quad MeV$$
$$m_p = 938.2723 \quad MeV$$
$$m_e = 0.510999 \quad MeV$$
$$0.7823 \quad MeV = Q \quad for \quad n \rightarrow p + e^-$$

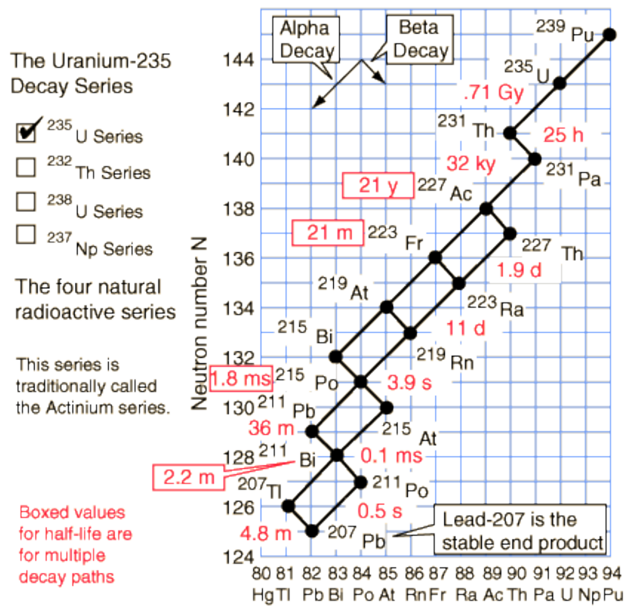
Neutrons from Radioactive decays?





More Radioactive decays





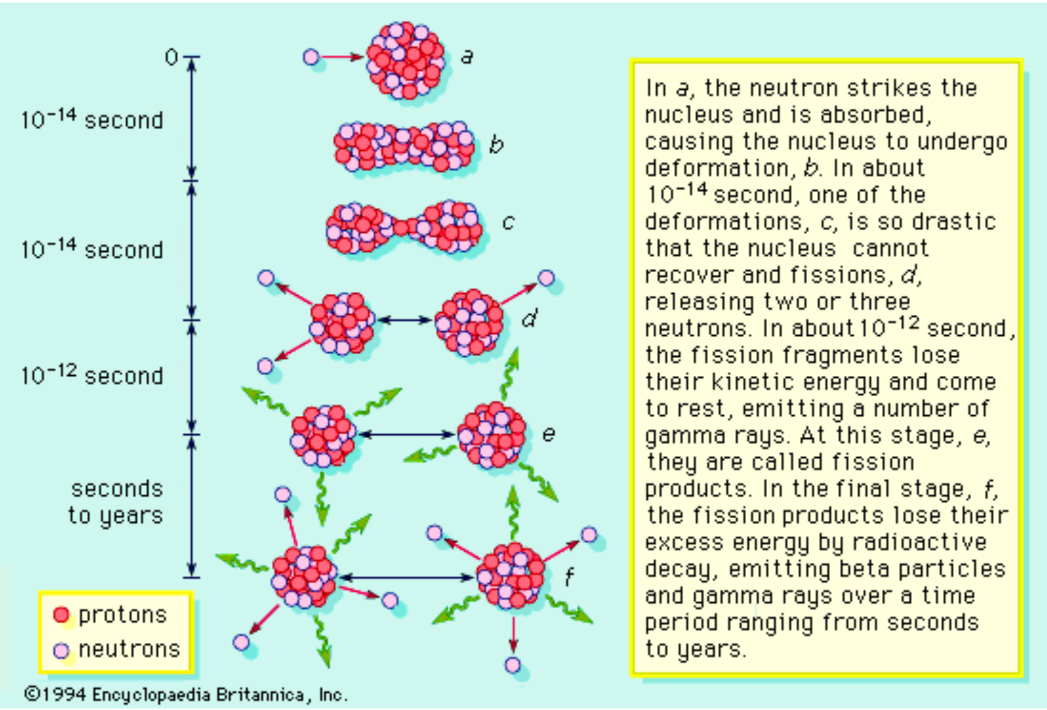
Spontaneous nuclear fission

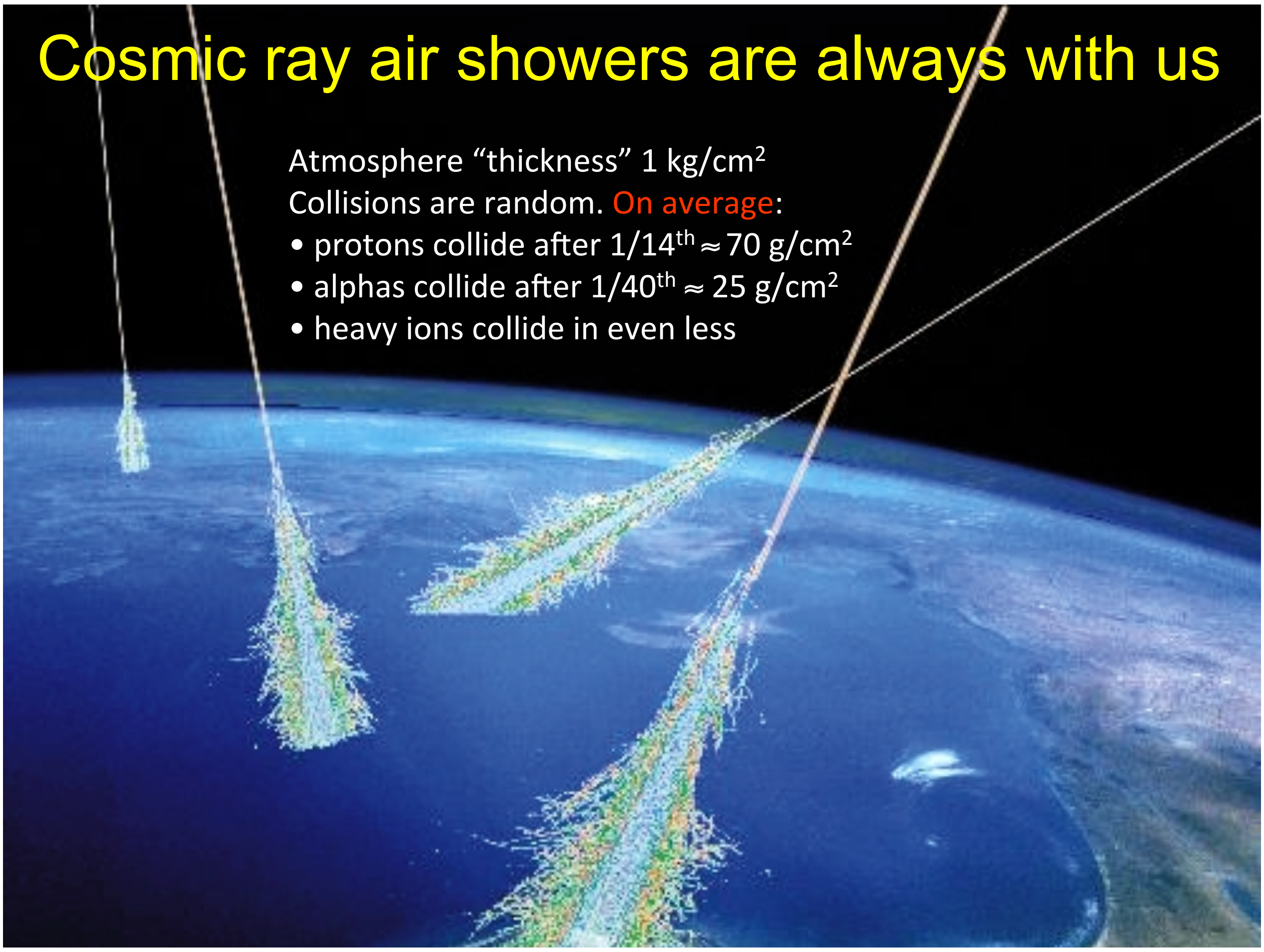
Nuclide	Half-life	Fission prob. per decay	Neutrons per fission	Neutrons per gram-second	Spontaneous half life
^{235}U	7.04×10^{8} years	2.0×10^{-9}	1.86	3.0×10^{-4}	3.5×10^{17} years
^{238}U	4.47×10^{9} years	5.4×10^{-7}	2.07	0.0136	8.4×10^{15} years
^{239}Pu	2.41×10^{4} years	4.4×10^{-12}	2.16	0.022	5.5×10^{15} years
^{240}Pu	6569 years	5.0×10^{-8}	2.21	920	1.16×10^{11} years
^{250}Cm	8300 years[7]	0.8	3.31	1.6×10^{10}	N/A
^{252}Cf	2.638 years	3.09×10^{-2}	3.73	2.3×10^{12}	N/A

In practice Pu-239 will invariably contains a certain amount of Pu-240 due to the tendency of Pu-239 to absorb an additional neutron during production. Pu-240's high rate of spontaneous fission events makes it an undesirable contaminant. Weapons-grade plutonium contains no more than 7.0% Pu-240.

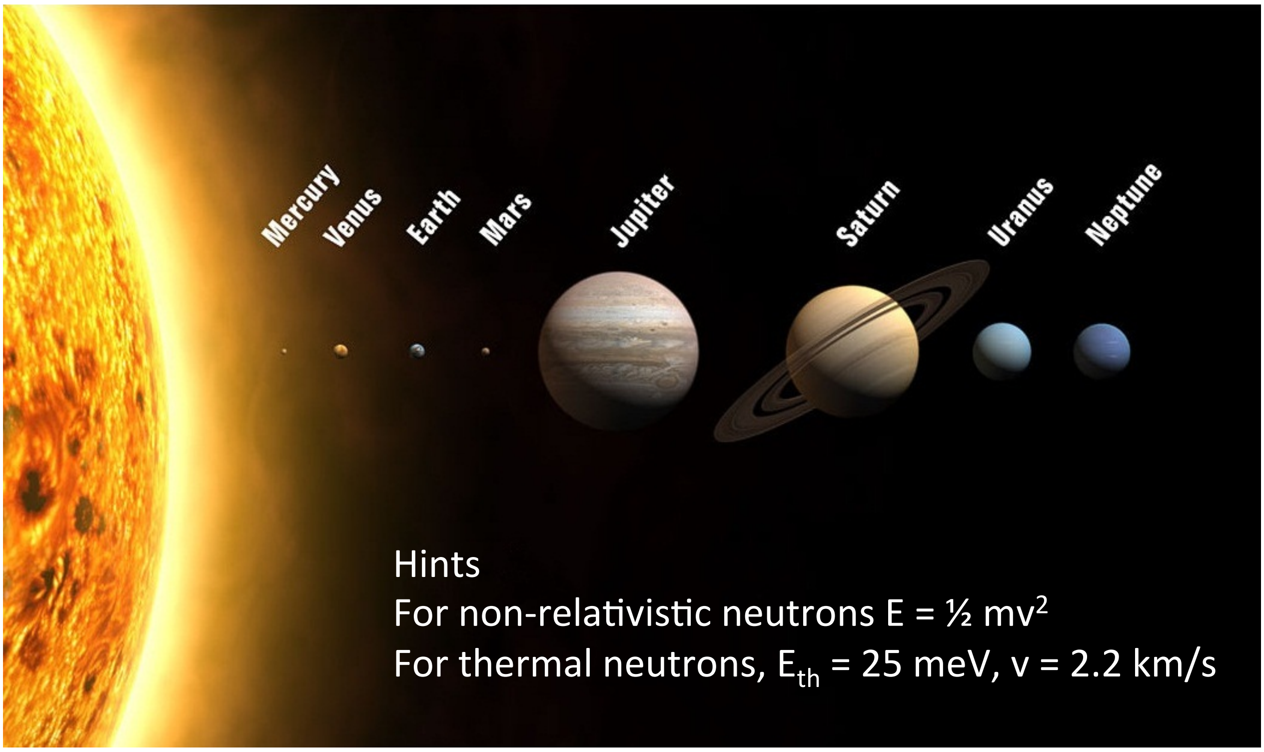
Source Wikipedia

Nuclear fission



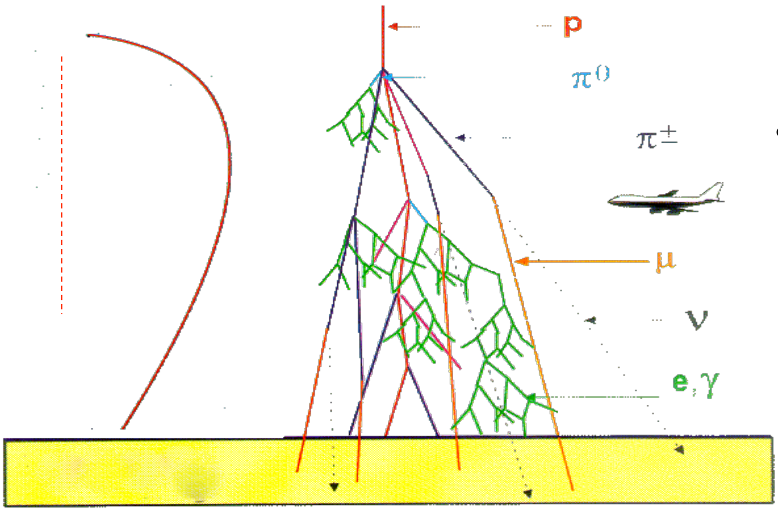


…and solar neutrons?



Cosmogenic neutrons

Shower maximum at ~18 km



- Under 20 km altitude neutrons dominate as cause of so called Single Event Effects in avionic systems.
- In mountains and at sea level there are enough of them to be a real concern for ground-based electronics that play vital roles (e.g. in computers, pace makers, cars, voting machines, power devices in locomotives..)

Why do Medical Physicists care about neutrons?

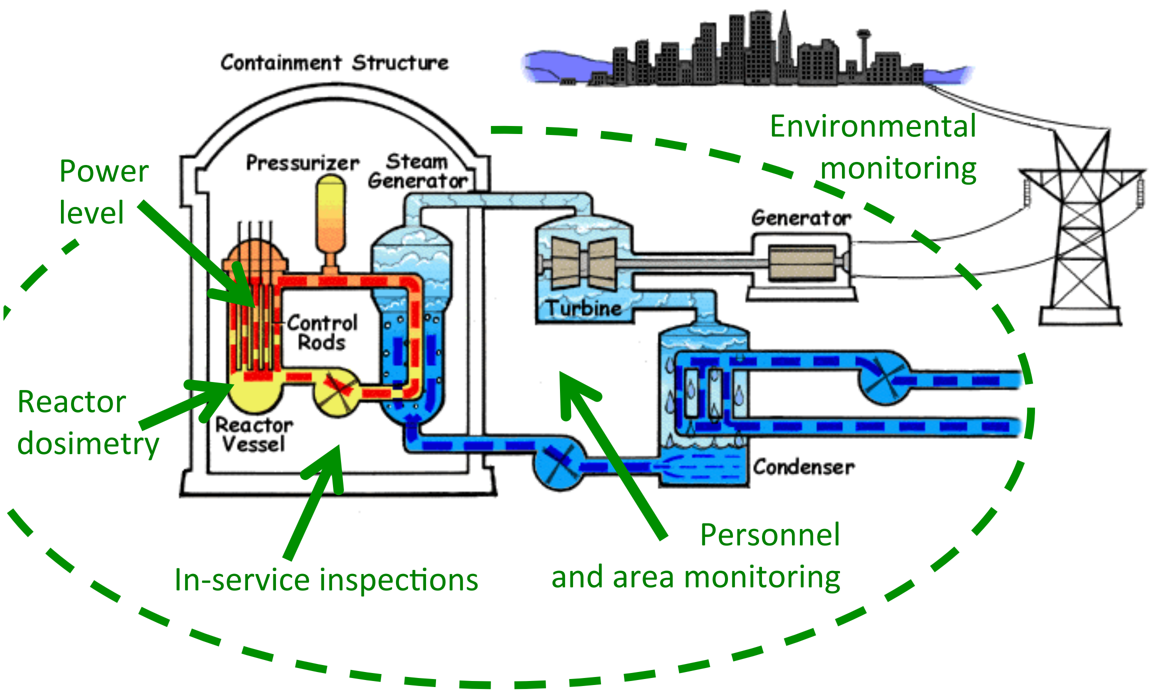
- Neutrons in Medicine
  - Neutron Therapy
  - Neutrons Contamination in X-Ray and Hadron Therapy

- Unwanted patient dose
- Shielding Considerations
- Neutron Dose

Why do Nuclear Engineers care about neutrons?

- Neutrons in Nuclear Power Plants
  - Chain reaction
  - Radiation damage/activation
  - Personnel/area monitoring

Neutron measurements at/around a nuclear plant

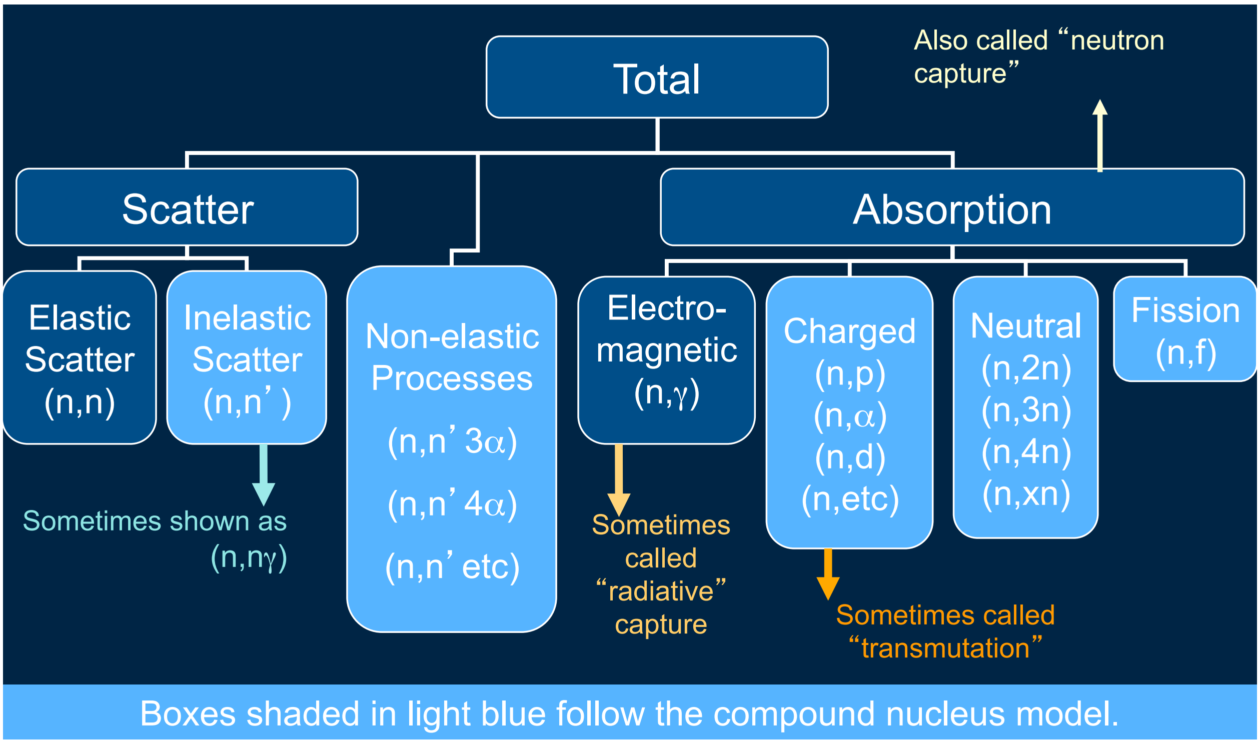


Neutron interactions and reactions

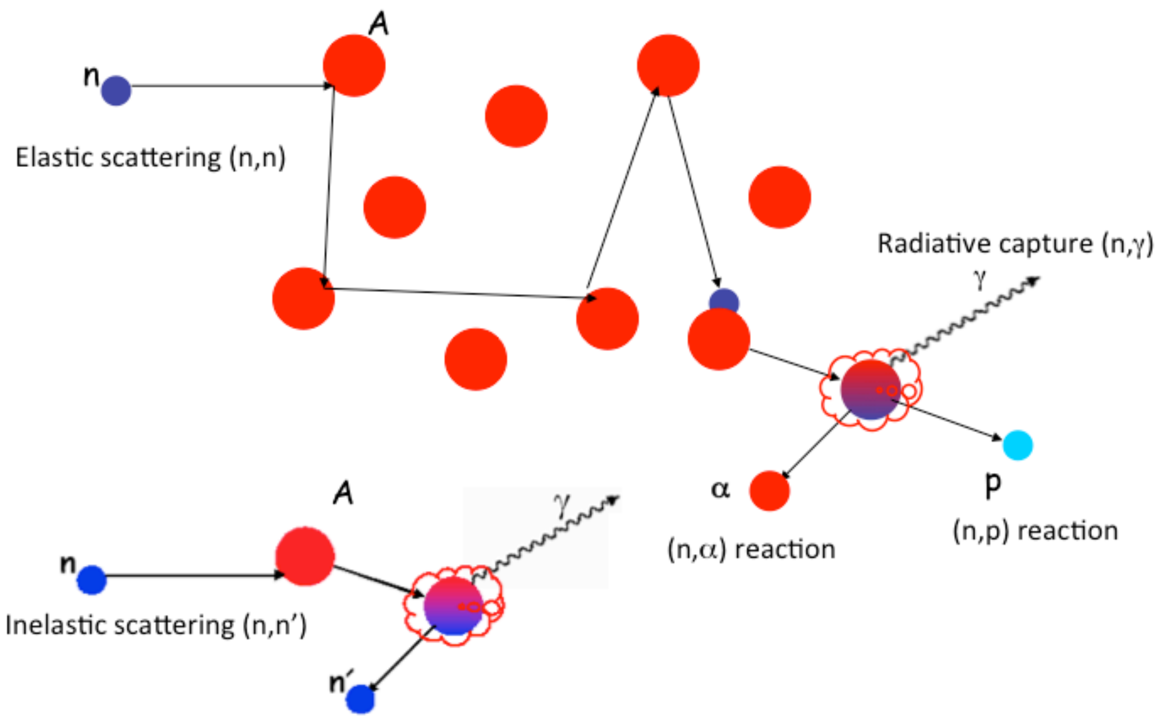
Neutrons – More General Properties

- Neutrons are Neutral
  - Can Not interact by Coulomb forces
  - Can travel through several cm of material without interacting.
- Neutrons interact with nuclei of absorbing material (they do not interact with orbital electrons).

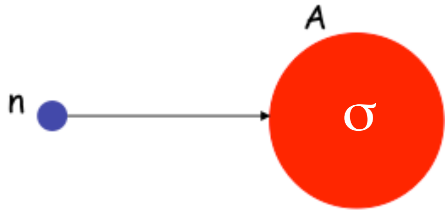
Overview of Neutron Interactions Scatter and Absorption



Neutron interaction types



Reaction Cross Sections



- A neutron reaction cross section quantitatively describes the probability of a particular interaction occurring between a neutron and matter.
- When the reaction cross section is defined microscopically per nucleus, it is denoted by σ and has S.I. Units = cm^2
  - Common unit for reaction neutron cross sections is the **barn** **($10^{-24}cm^2$)**.
- Reaction cross sections are BOTH energy and interaction type dependent.

Reaction Cross Sections

- Macroscopic cross section, Σ, probability per unit path length that a particular type of interaction will occur.

$$\boxed{\Sigma = N\sigma}$$

  - σ = microscopic cross section, cm^2
  - N = number of nuclei per unit volume, nuclei/cm^3

- All processes can be combined to calculate Σ_{total}, probability per unit path length that any type of interaction will occur.

$$\boxed{\Sigma_{Total} = \Sigma_{scatter} + \Sigma_{rad.capture} + \ldots..}$$

Exponential Attenuation

- Neutrons are removed exponentially from a collimated beam by an absorbing material.

$$I = I_o e^{-\boxed{\Sigma_{total}}\,t}$$

$\sigma_{total} N$

where

N number of absorber atoms per cm^3 (atomic density)

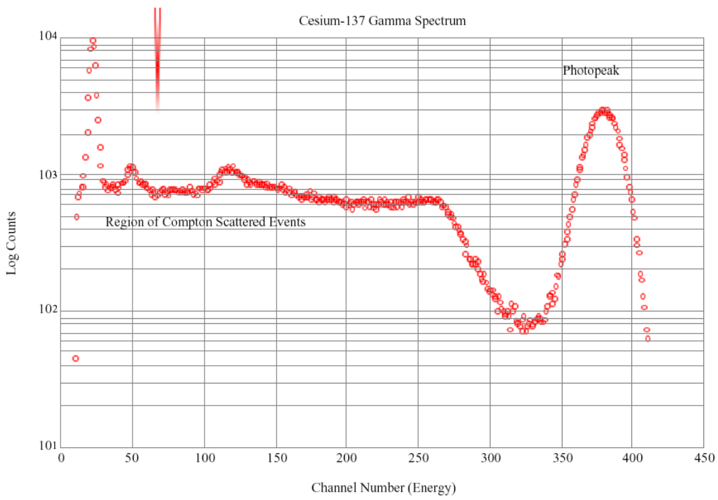
σ microscopic cross section for the absorber, cm^2

t absorber thickness, cm

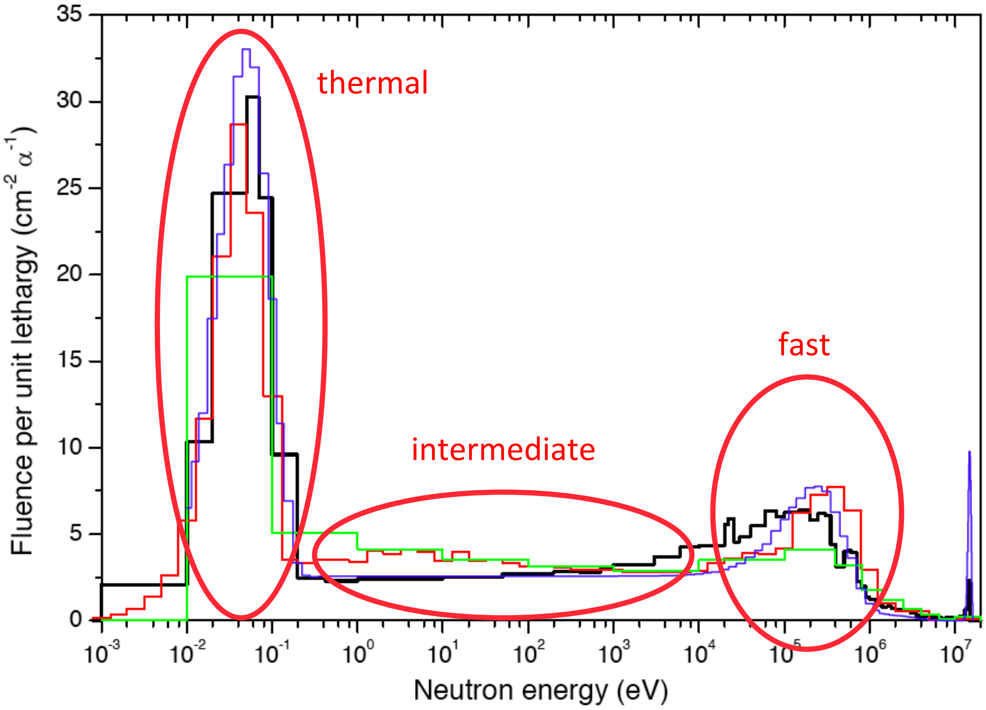
How does this help us detecting neutrons?

Photon detection

- Neither photons nor neutrons are directly ionizing.
- To detect them they must be converted to directly ionizing particles
  - Photons interact primarily with electrons
  - Reliance on photoelectric effect
  - The photo-electron acquires all the energy of the gamma ray



Energy spectrum of reactor neutrons



Neutron Interactions are Energy Dependent

- There is no real equivalent of the photoelectric effect for neutrons
- Lower energy neutrons (thermal or near thermal) are likely to undergo absorption reactions with atoms in their environment.
- Fast neutrons are most likely to undergo scatter interactions with atoms in their environment.
  - Elastic Scatter – dominate for lower energy fast neutrons
  - Inelastic Scatter – above 1 MeV

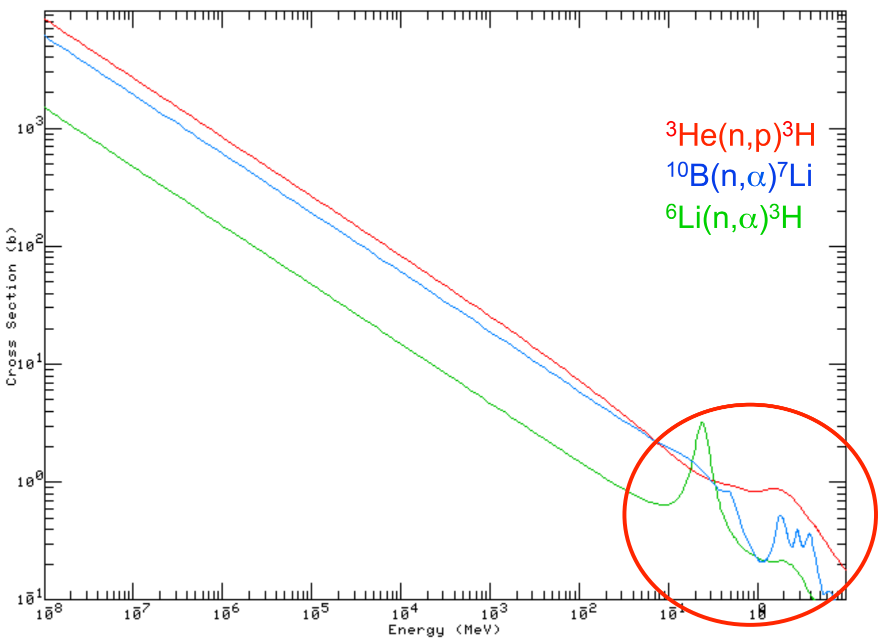
Neutron Detection

- At low energies some reactions with low-Z nuclei can be used to transfer the neutron energy to charged particles: e.g.

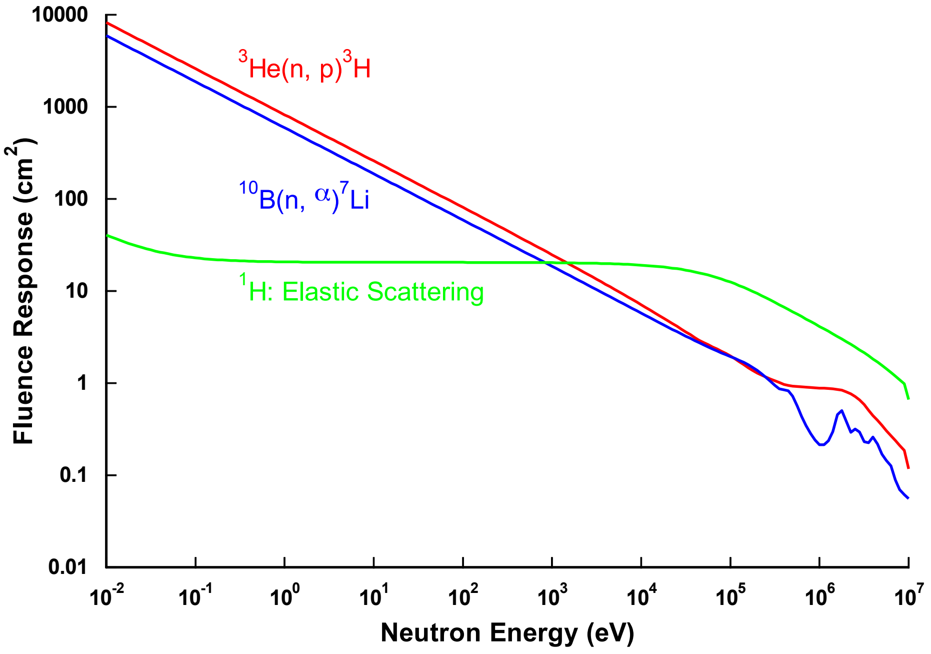
Reaction	Q value
$^{3}He(n,p)T$	764 keV
$^{6}Li(n,\alpha)T$	4.78 MeV
$^{10}B(n,\alpha)\ ^{7}Li$	2.79 MeV

- Note: both products are charged and can be detected

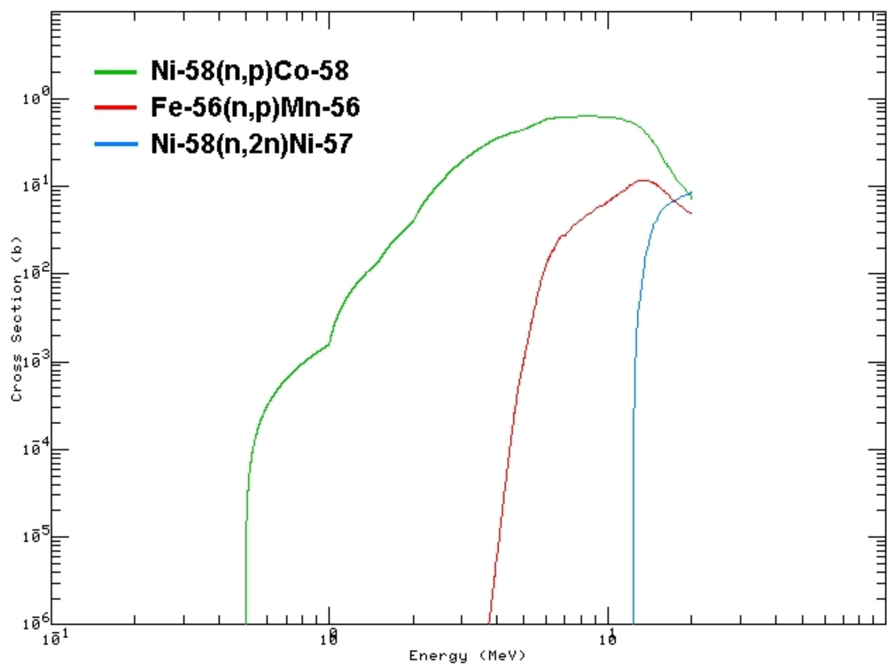
^{3}He, ^{10}B and ^{6}Li cross sections



Some capture vs scatter cross sections



Fast Neutron Absorption (Activation)



Neutron energy deposition in tissue

Neutron Interactions in tissue

- Neutrons are indirectly ionizing and but give rise to densely ionizing (high LET) particles: recoil protons, α-particles, and heavier nuclear fragments
  - These particles then deposit dose in tissue.

- The type of interaction and the amount of dose deposited in the body is strongly dependent on neutron energy and absorbing material.
  - The most common elements in the human body are **Hydrogen, Carbon, Nitrogen,** and **Oxygen**.

Slow Neutron Interactions with Tissue

$^{1}H(n_{th},\gamma)^{2}H$

- It releases a 2.22 MeV γ-ray that irradiates the surrounding tissue
- It is one of the two important interactions by which thermal neutrons deposit energy in tissue
- Often seen as a background gamma-ray in power and research reactors

$^{14}N(n_{th},p)^{14}C$

- It releases 626 keV and contributes approximately 1% of the total dose equivalent in soft tissue for neutron energies less than 10 MeV
- the absorbed dose is delivered locally at the interaction site, since the ranges of proton and ^{14}C recoil nucleus are short

$^{23}Na(n_{th},\gamma)^{24}Na$

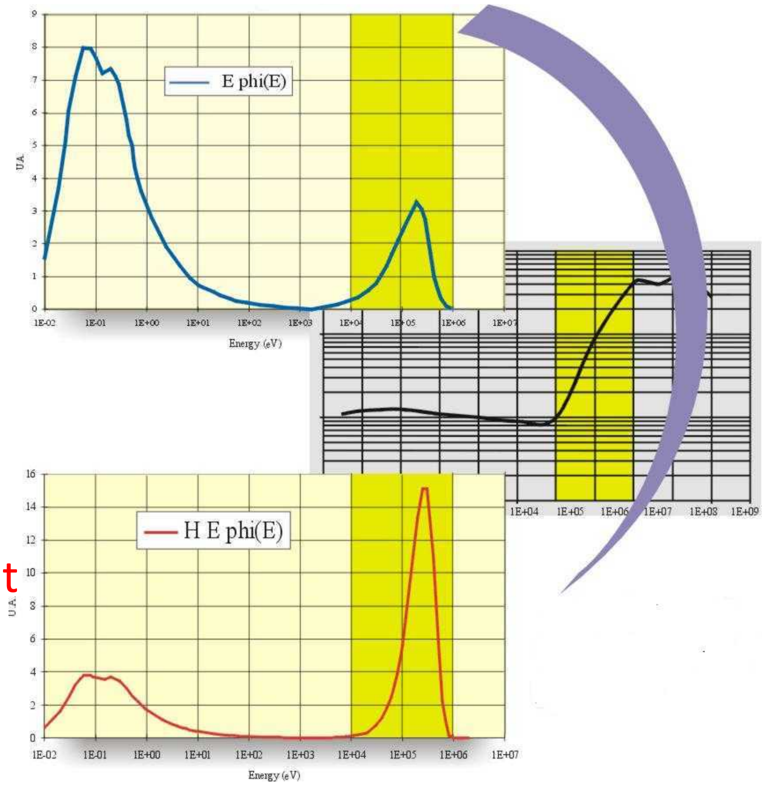
- The decay of ^{24}Na (half-life = 15 h) yields two γ's of 100% intensity: 1.37 and 2.75 MeV)
- It activates human blood sodium and can be used to quick-sort personnel after a suspected criticality

Fast Neutron Interactions in tissue

- Fast neutrons interact with hydrogen via elastic processes and result in the release of energetic protons
- Higher energy neutrons interact with carbon and oxygen via nonelastic processes and result in the release of charged α-particles, *(n,n'3α)* and *(n,n'4α)*.
  - These α-particles then deliver dose to tissue

Neutron radiation protection dosimetry

Neutron fluence spectrum around a reactor



Dose-equivalent weighted spectrum

Ideal neutron dosimeter response, H*(10)/Ø

Neutron Reactions Important in Health Physics

$^{3}He(n_{th},p)^{3}H$

- It is the basis for the use of ^{3}He as a gas in several types of neutron proportional counters

$^{6}Li(n_{th},t)^{4}He$ or $^{6}Li(n_{th},\alpha)^{3}H$

- It releases a tritium nucleus (triton) and a helium nucleus (α particle)
- it is used in many neutron detection instruments, including thermoluminescent dosimeters (TLDs)

$^{10}B(n_{th},\alpha)^{7}Li$

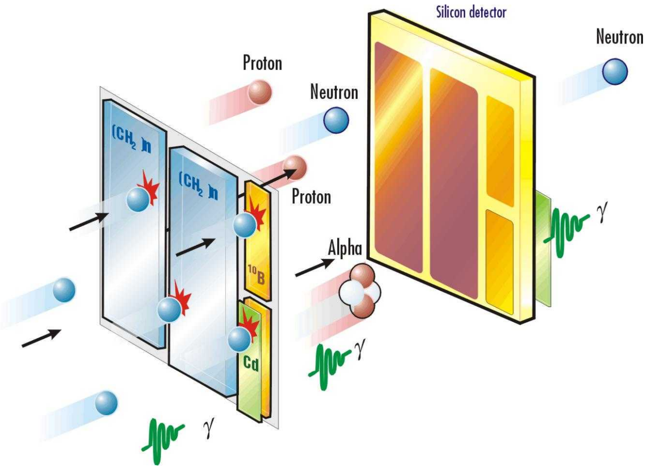
- It is used in neutron shielding and as the basis for neutron detectors utilizing BF_3 gas or boron-lined counter tubes

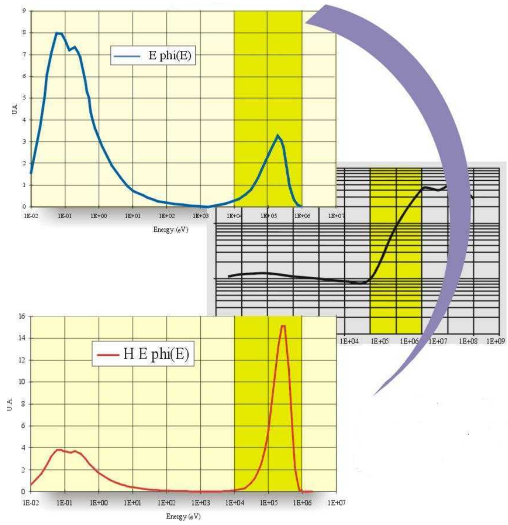
$^{1}H(n,n)^{1}H$

- It is used in neutron moderation/shielding and in organic (solid or gas) detectors and dosimeters

Neutron dosimetry approaches

- Design of systems mimicking the fluence to dose equivalent conversion coefficient
- Measurements of LET spectra and convolution over Q(L).





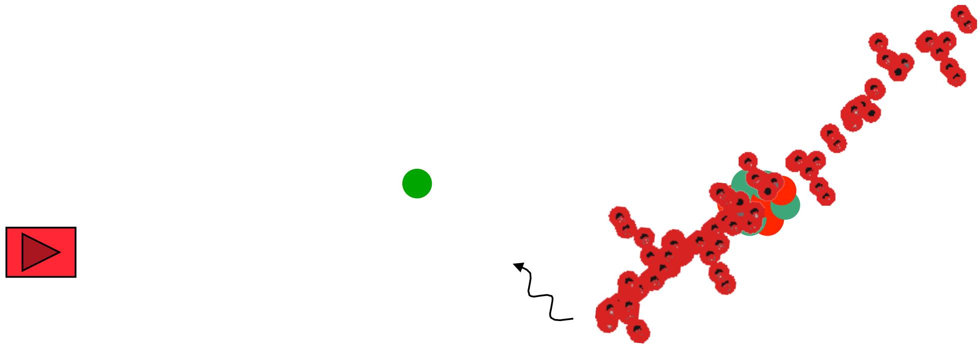
- Utilization of physical phenomena resembling dose (equivalent) deposition in tissue.

Neutron monitoring and dosimetry

- Moderated thermal neutron sensors
- Tissue equivalent proportional counters
- Superheated emulsions

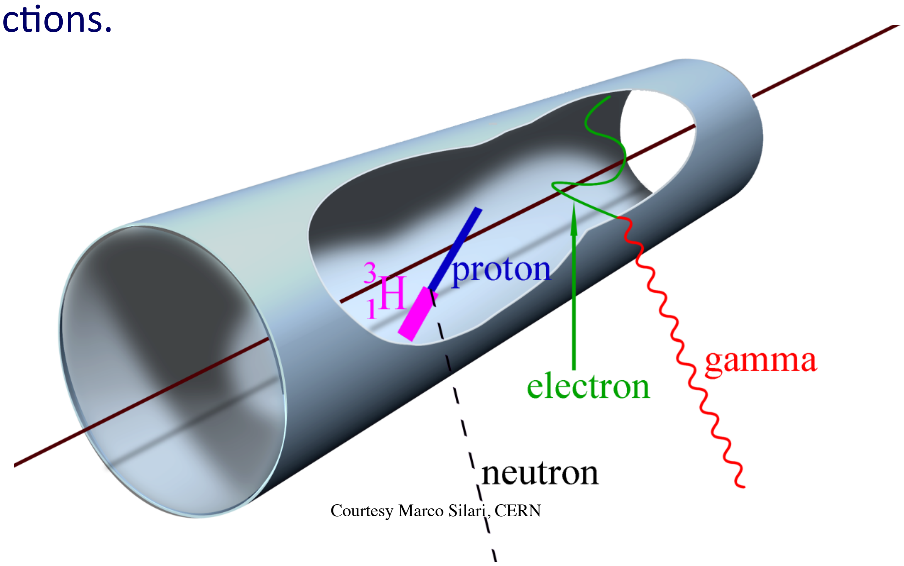
Neutron detection basics

- Neutrons are detected through:
  → secondary charged particles
    → generated via elastic or inelastic reactions with nuclei.



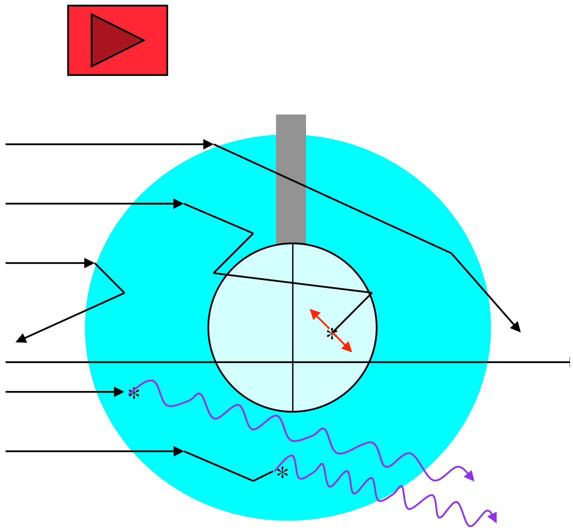
Neutron proportional counters

- Gamma rays can interact in the walls and produce electrons in the gas, but the energy loss of electrons is small (≈ 2 keV/cm), so that these pulses are much smaller than those due to neutrons;
- A pulse amplitude threshold can thus eliminate most gamma interactions.

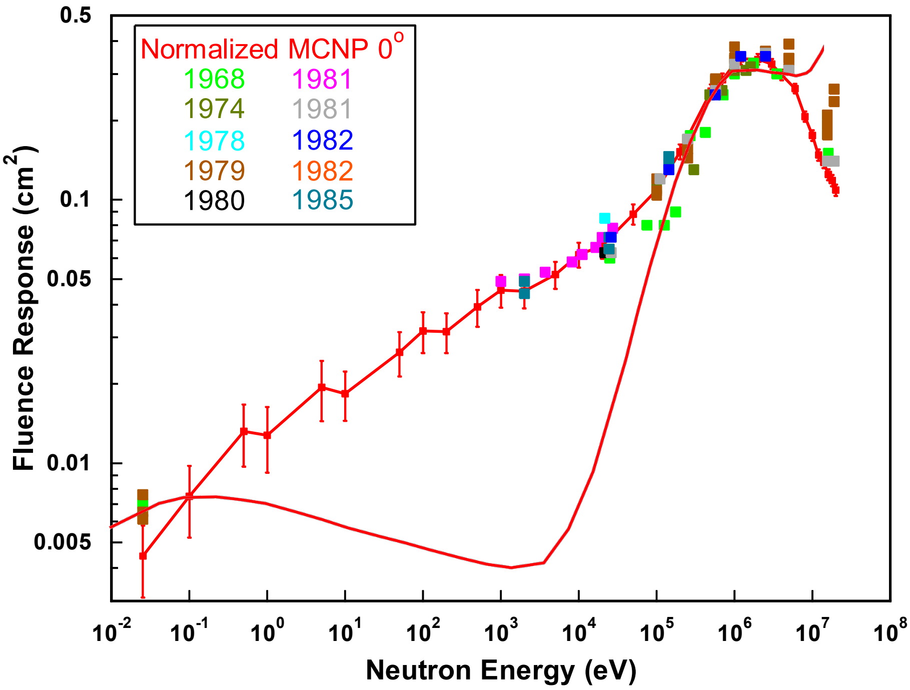


Neutron area survey instruments

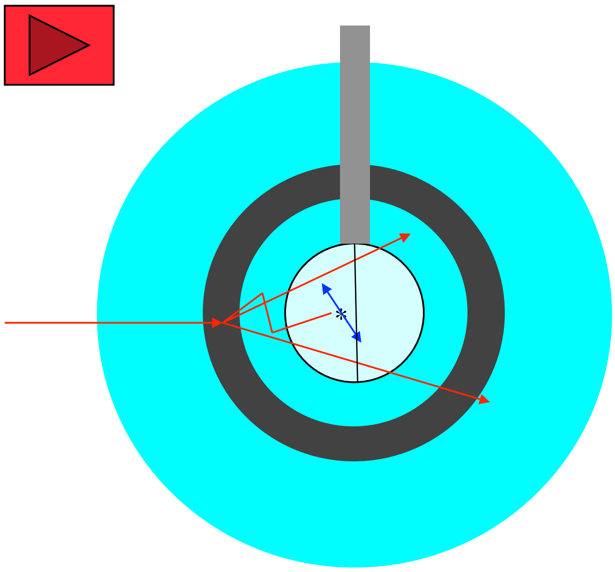
- Thermalize the neutron field using large mass of hydrogenous material
- Correct the over-response to intermediate energy neutrons via an absorbing layer (e.g. boron or cadmium) in the moderator
- Prevent an over-suppression of the thermal neutron response via holes in the absorbing layer
- More sophisticated designs not in widespread use



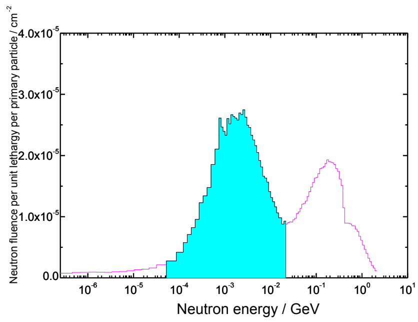
Fluence response R/Φ of a survey monitor (Leake design) MCNP and experiment



Detection of high-energy neutrons (LINUS)

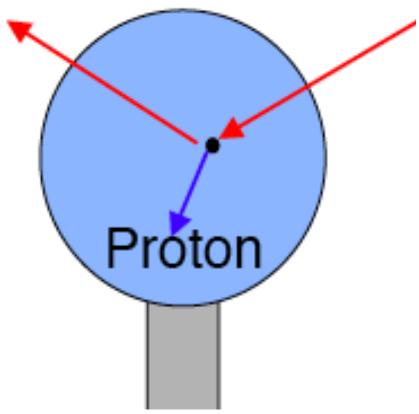


- The instrument response can be extended to high-energy neutrons (up to a few GeV) by coupling an attenuator shell of high-mass number to the moderator. Evaporation neutrons play a fundamental role for improving the response to HE neutrons.
- This technique is applied to BSS, rem-meters, liquid scintillators and superheated emulsions.

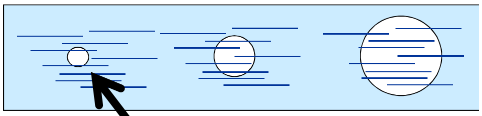


Tissue Equivalent Proportional Counters (TEPC)

- Cylindrical or spherical housing (Depending on Type and Model)
- Uses 2 - 5 inch Hollow Sphere of A150 TE Plastic in stainless steel
  - Low pressure TE Proportional Gas
  - Typically Simulates 1-2 μm Tissue Sphere – Bragg-Gray Principle
  - Measures lineal energy distribution
  - Microprocessor Multi-Channel Analyzer
- Converts specific energy absorption to Dose Equivalent
  - QFs from ICRP recommendations



Bragg-Gray cavity theory



The main assumptions of this theory are:

- the cavity dimensions are so small compared to the range of charged particles within it so that the fluence of charged particles inside the cavity is not perturbed by the presence of the cavity
- there are no interactions of uncharged particles in the cavity so that the absorbed dose deposited in the cavity is due to the charged particles that cross the cavity

Under these conditions:

$$\frac{D_w}{D_g} = \frac{\int_{T_{\min}}^{T_{\max}} \left(\frac{d\Phi}{dT}\right)_w \left(\frac{dT}{\rho dx}\right)_{c,w} dT}{\int_{T_{\min}}^{T_{\max}} \left(\frac{d\Phi}{dT}\right)_w \left(\frac{dT}{\rho dx}\right)_{c,g} dT} = \bar{\bar{S}}_g^{\,w}$$

D_w is the absorbed dose in the medium w
D_g is the absorbed dose in the cavity g
$\left(\frac{d\Phi}{dT}\right)_w$ is the fluence energy distribution of the electrons in the medium

The symbol $\bar{\bar{S}}_g^{\,w}$ has the double bar to indicate that this ratio of average stopping-powers considers both the average over the photon-generated electron spectrum and the changes in this spectrum due to the continuous loss of kinetic energy in the materials.

Absorbed Dose (D)

- The energy absorbed in a material (usually tissue) divided by the mass of the material

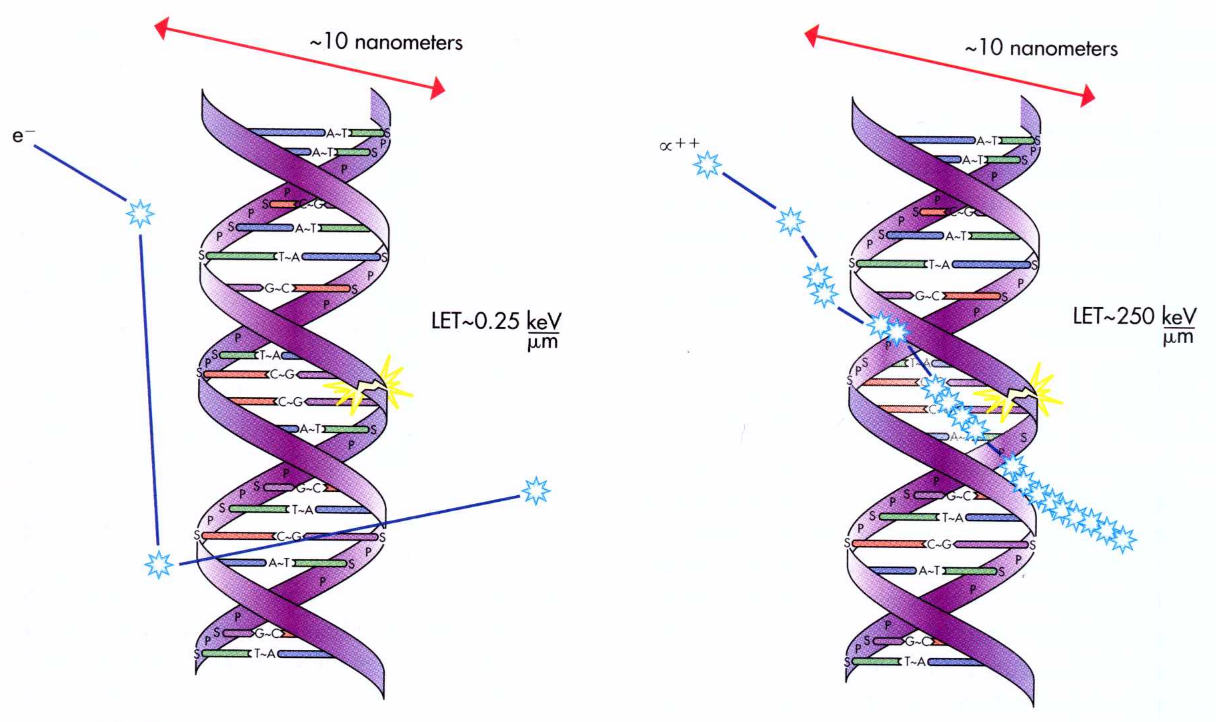
$$D = \frac{d\varepsilon}{dm_{material}} \qquad D_T = \frac{\int_{m_T} D\,dm}{m_T}$$

- Unit is the *gray,* Gy [J/kg] (SI), or the *rad* (US)

1 Gy = 100 rad

- Defined for all types of ionizing radiation in any medium
- Does not account for energy deposition pattern (ionization density)

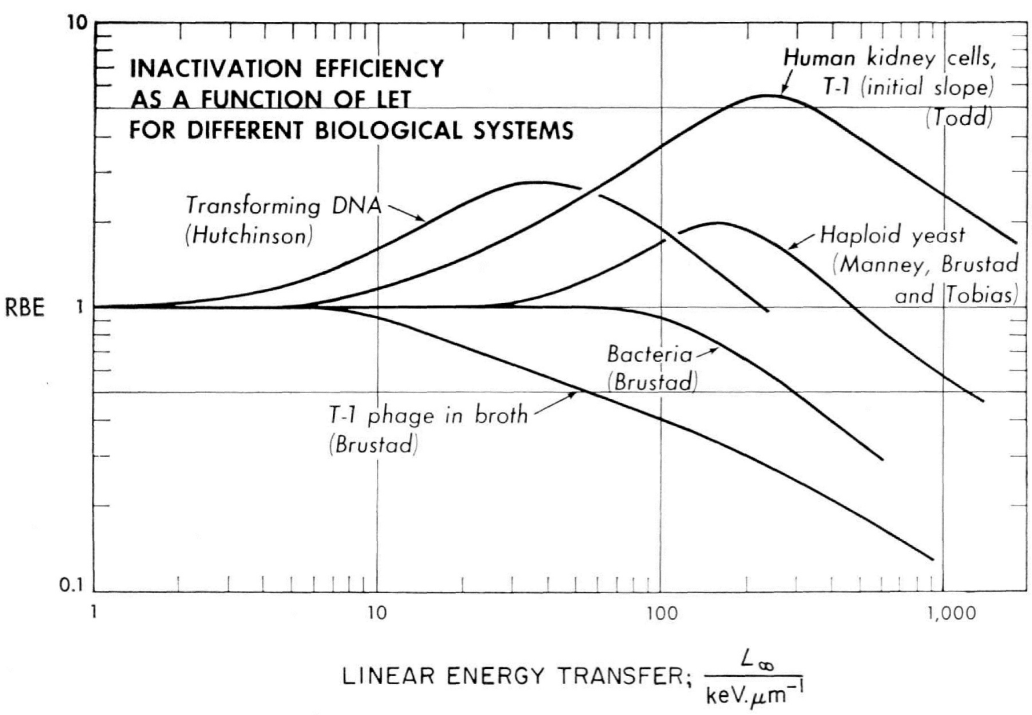
Linear energy transfer (LET)



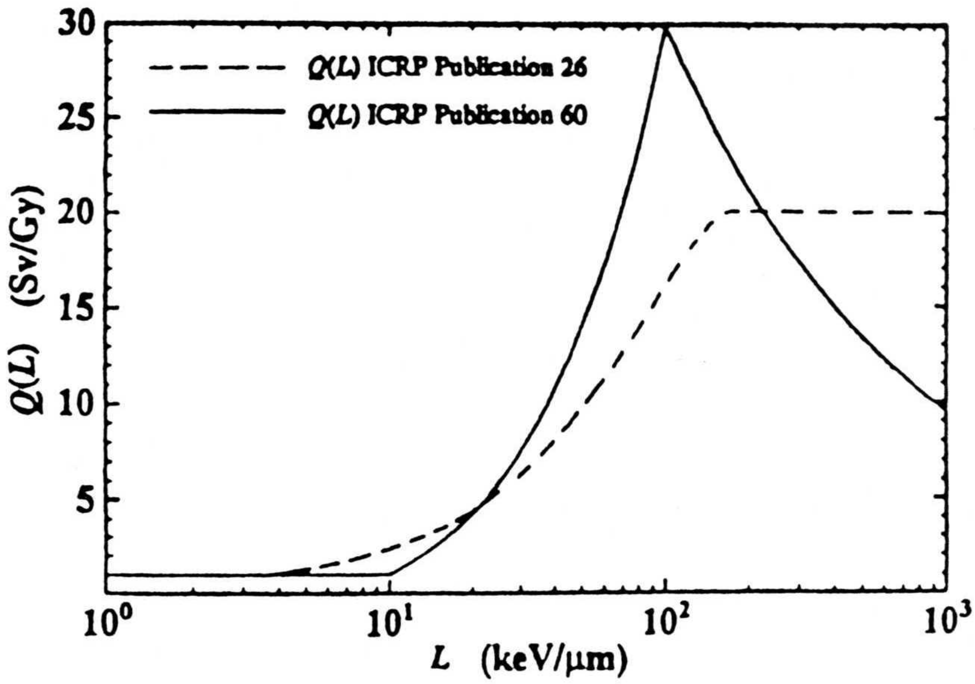
Radiation quality

- A physical quantity which describes biological effects for all radiation qualities does not (yet) exist
- Absorbed dose needs “correction factors”
  - relative biological effectiveness (RBE)
  - quality factors (Q)
- RBE
  - dependent on biological endpoint and object
  - dependent on many physical and biological parameters (radiation type, dose, dose rate, cell cycle, oxygen supply, …)
- Q
  - used only for stochastic effects at low doses and dose rates
  - defined by ICRP and ICRU

Linear energy transfer *versus* relative biological effectiveness



Quality factors



From absorbed dose to dose equivalent

- *Absorbed dose*: The energy absorbed in a material (usually tissue) divided by the mass of the material:

$$D = \frac{d\varepsilon}{dm_{material}} \qquad D_T = \frac{\int_{m_T} D\,dm}{m_T}$$

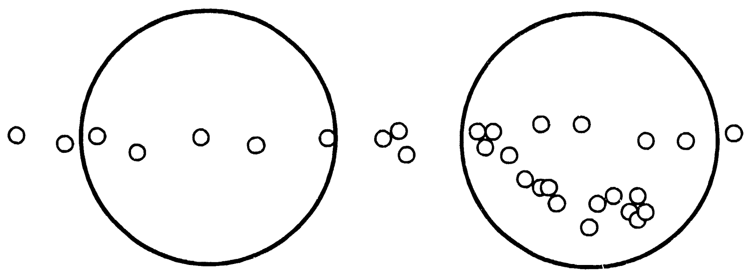
  – Unit is the *gray,* Gy [J/kg] (SI), or the *rad* (US), 1 Gy = 100 rad
  – Defined for all types of ionizing radiation in any medium

- *Dose equivalent*, dose weighted by an LET-dependent "*quality factor*" Q

$$H = D\,Q$$

  – Defined at a point in a reference phantom
  – Unit of measure is the *sievert,* Sv [J/kg] (SI), or the *rem* (US), 1 Sv = 100 rad

Ionization produced along the path of a neutron recoil proton in a TEPC



Small energy deposit

Large energy deposit

Path of recoil proton with constant linear energy transfer

$\bar{l} = 4V/S$

Lineal Energy, y

$y = \varepsilon / \bar{l}$

Microscopic quantity which can be experimentally measured by TEPC

Stochastic quantity

Linear Energy Transfer, L

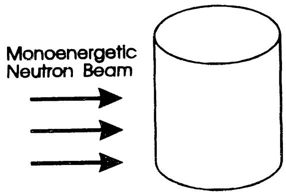
$L = dE / dx$

Macroscopic quantity-

average of many measurements or calculated value

Comparison of methods to obtain neutron dose equivalent

From Conversion Factors



$$H = \sum C(E) \quad \varphi(E)$$

Calculated Value

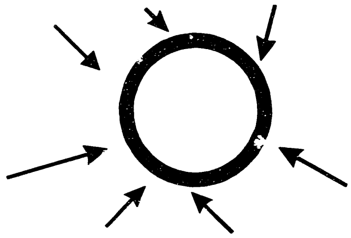
$H(n,\gamma)$ included in neutron dose

Interpolated C(E) values for Different neutron energies

H_{max} at different depths for each neutron energy

Conservative estimate

From TEPC Measurements



$$H = \bar{Q} \quad D$$

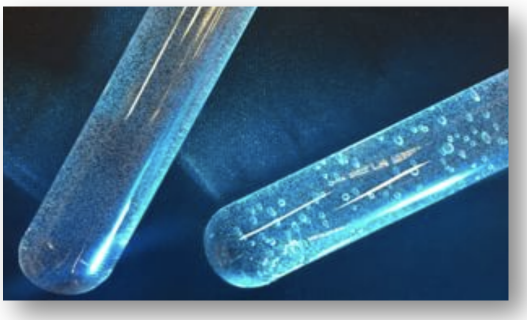
Point in space measurement (kerma)

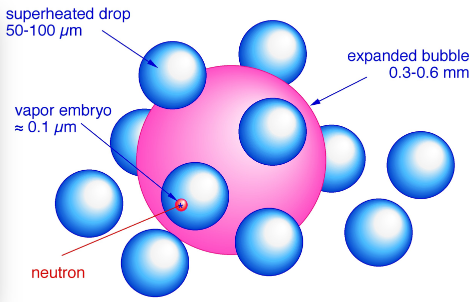
$H(n,\gamma)$ included in gamma dose

Neutron recoils measured

Measured at single depth (thickness of wall)

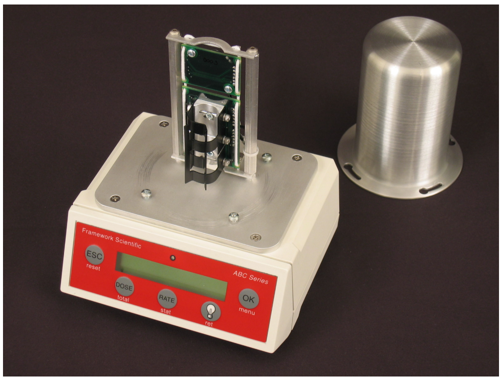
Superheated emulsions

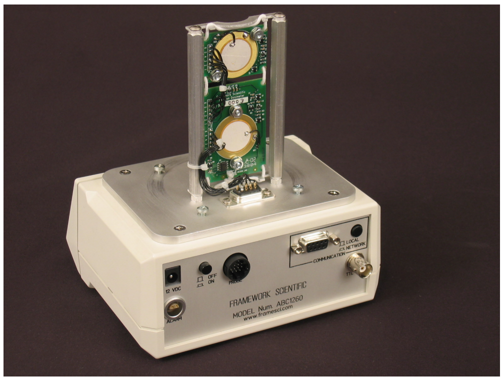




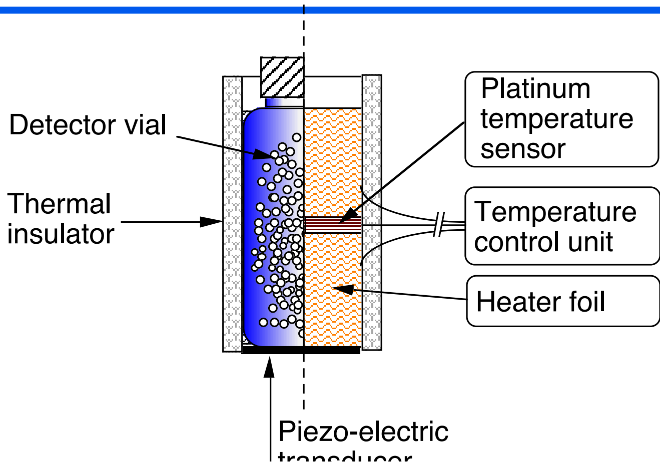
- Fluorocarbon droplets kept in a steady superheated state by emulsification in compliant aqueous, polymers.
- Bubble nucleation triggered by neutrons above selectable threshold energies (e.g. above interrogation energy).
- Can be totally insensitive to photons.

Some current technologies





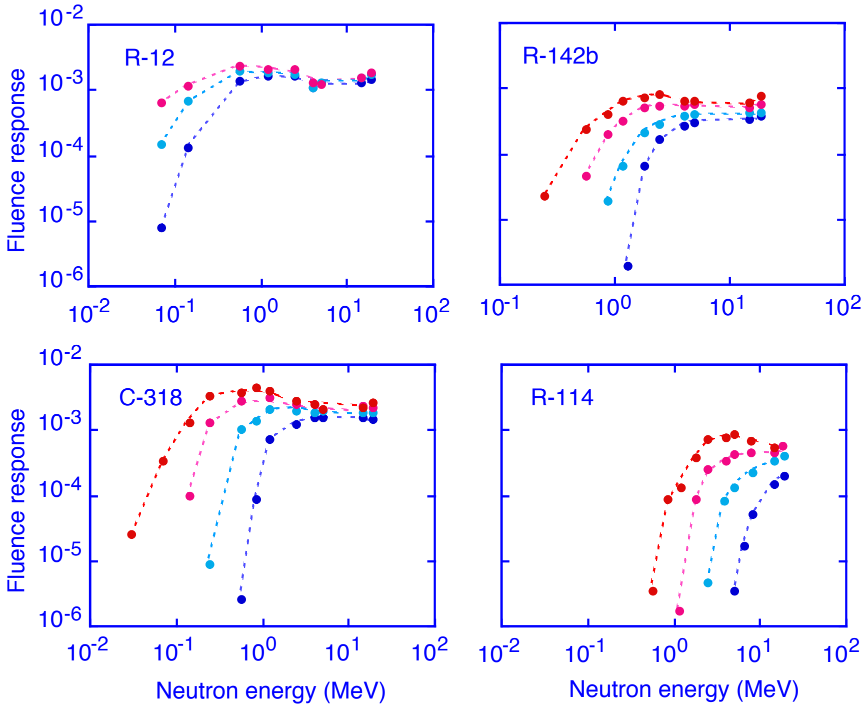
Neutron response studies



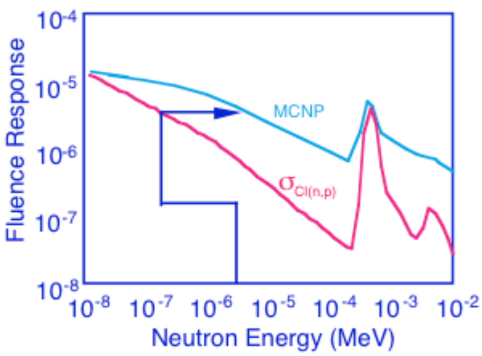
Chemical name	Chemical abstracts number	Refrigerant number	Empirical formula	Boiling point T_b (°C)[a]	Critical point T_c (°C)
Dichlorotetrafluoroethane	76-14-2	R-114	$C_2Cl_2F_4$	3.65	145.7
Monochlorodifluoroethane	75-68-3	R-142b	$C_2H_3ClF_2$	−9.14	137.15
Octafluorocyclobutane	115-25-3	C-318	C_4F_8	−6.99	115.22
Dichlorofluoromethane	75-71-8	R-12	CCl_2F_2	−29.76	111.8
Tetrafluoroethane	811-97-2	R-134a	$C_2H_2F_4$	−26.07	101.2
Hexafluoropropylene (HFP)	116-15-4	–	C_3F_6	−29.40	85.0
Monochloropentafluoroethane	76-15-3	R-115	C_2ClF_5	−39.17	79.9
Octafluoropropane	76-19-7	R-218	C_3F_8	−36.65	71.95

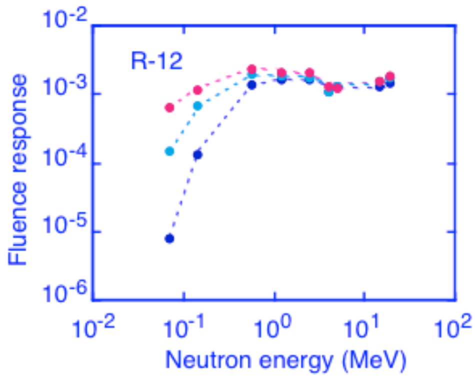
[a] At atmospheric pressure (101 kPa).

Fluence response of R-12, R-142b, C-318, R-114 vs temperature and neutron energy



Response of dosimetric (chlorine-bearing) emulsions to fast and slow neutrons





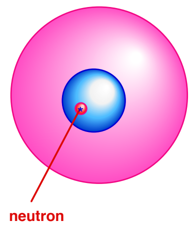
$^{35}Cl(n,p)^{35}S$ Q = 615 keV

- p receives 598 keV
- ^{35}S receives 17 keV (range 0.04 μm)

Skripov's theory of radiation-induced nucleation in bubble chambers

Critical bubble radius based on the thermodynamics of isothermal spontaneous nucleation (Gibbs):

$$R_c = 2\sigma/(p'' - p') \approx 2\sigma/[(p_s - p')(1 - v'/v'')]$$



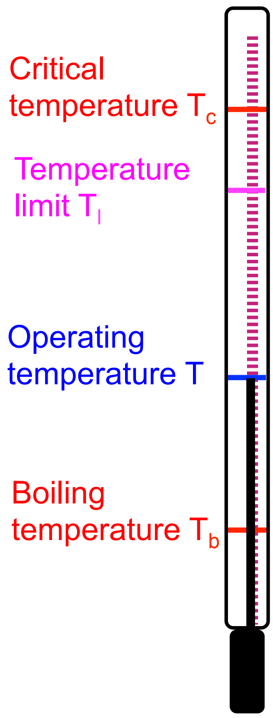
Vaporisation energy required in the heterogeneous nucleation by an ionising particle (Skripov):

$$W_{rev} = \frac{16\pi\sigma^3}{3(p_s - p')^2(1 - v'/v'')^2}\left[1 + \frac{2\Delta H}{(p_s - p')(v'' - v')} - \frac{T}{\sigma}\frac{d\sigma}{dT}\right]$$

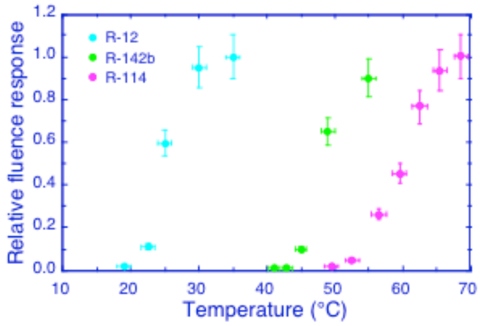
Reduced superheat:
$s = (T - T_b)/(T_c - T_b)$

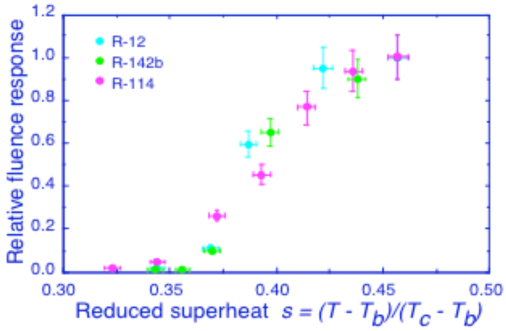
T_b is the boiling point, above which a fluid is either a vapor or a superheated liquid (lower limit of superheat)

T_c is the critical temperature, above which the liquid phase can no longer exist, and a fluid becomes a gas (theoretical upper limit for the superheated state)



Thermal neutron sensitization vs temperature and vs reduced superheat

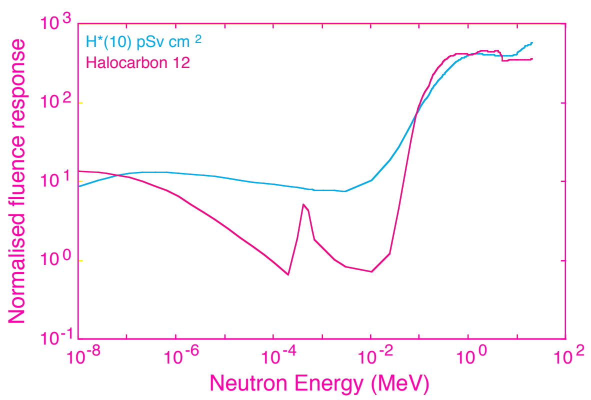


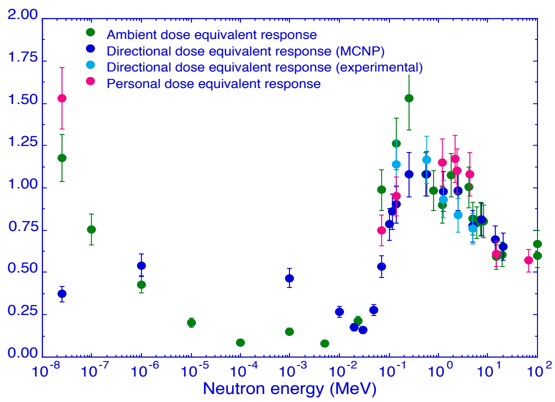


Halocarbon	T(°C)	s	W_o(keV)	R_c(μm)
R-12	17.0	0.330	17.0	0.047
	17.5	0.334	16.1	0.046
	18.0	0.337	15.2	0.044
R-142B	41.0	0.343	17.8	0.044
	41.5	0.346	16.9	0.043
	42.0	0.350	16.0	0.042
R-114	50.5	0.330	17.8	0.047
	51.0	0.333	16.8	0.046
	51.5	0.337	15.9	0.045

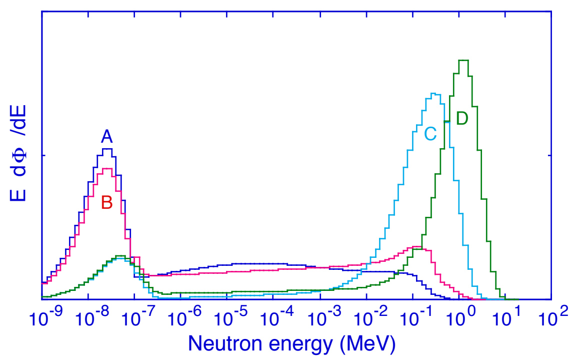
s = reduced superheat, W_o = vaporization energy, R_c = critical radius

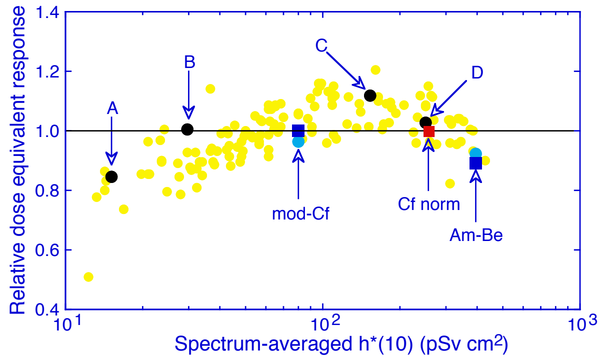
Dose equivalent response of R-12 to monoenergetic neutrons



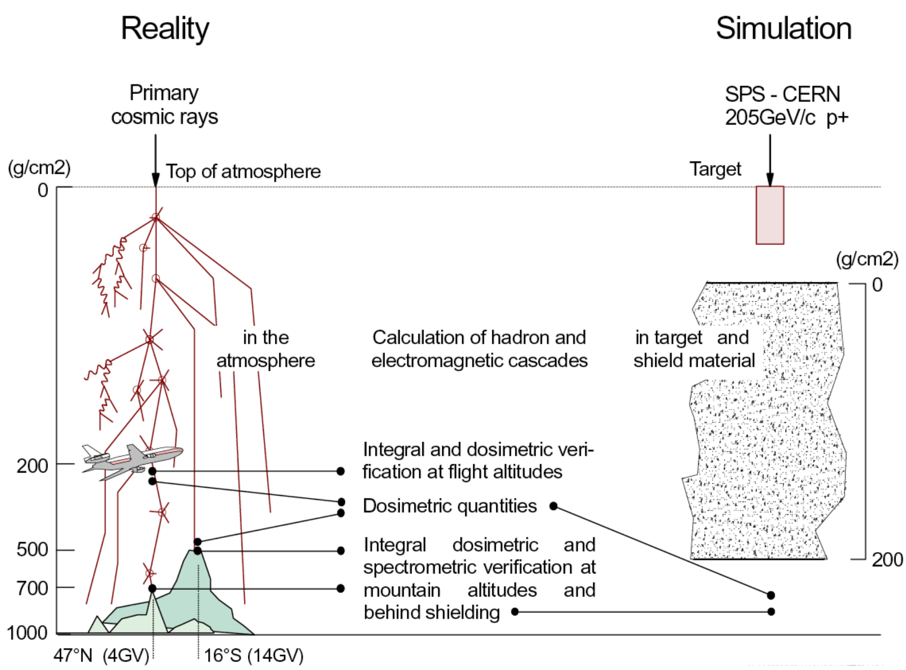


Dose equivalent response of R-12 to reactor and radionuclide spectra

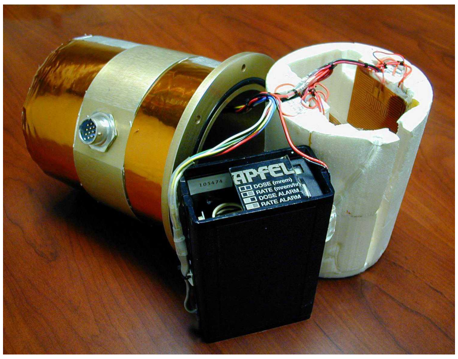


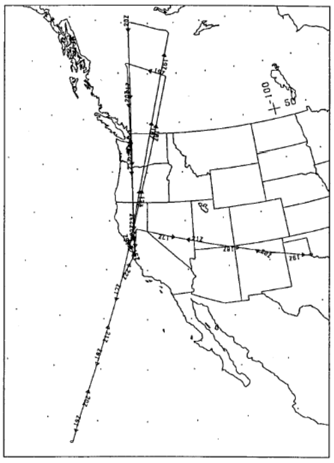


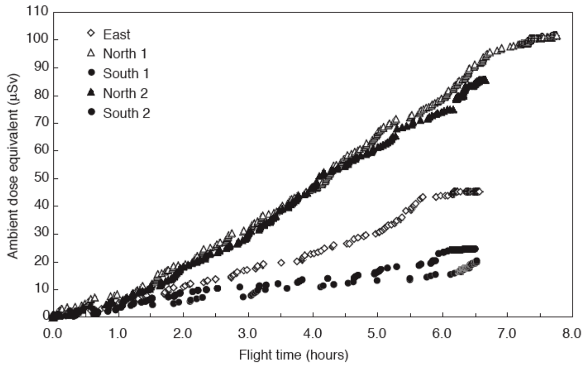
The cosmic radiation field in the atmosphere and its simulation at CERN



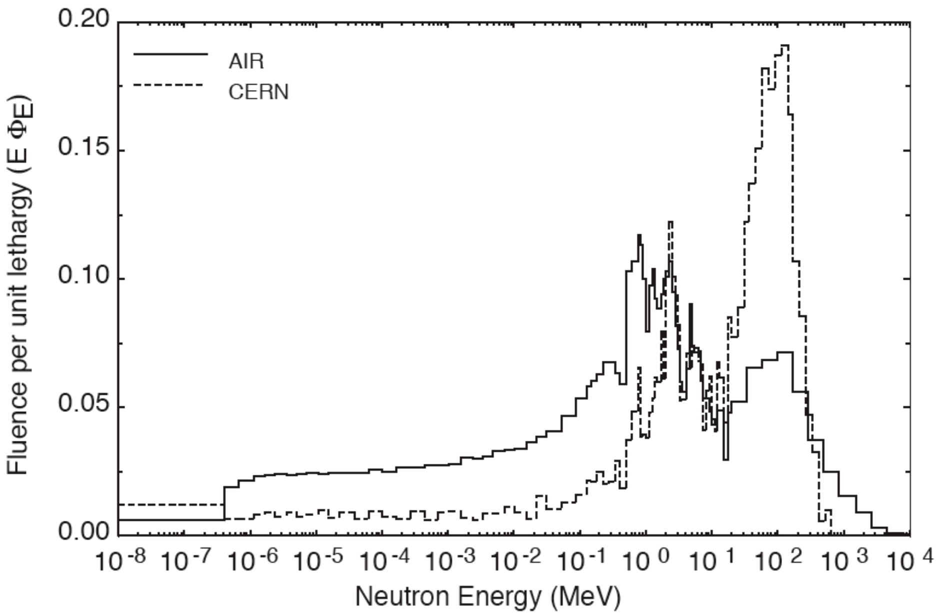
NASA cosmic radiation measurements







Comparison between neutron spectra at high-altitude and at the CERN-CERF facility

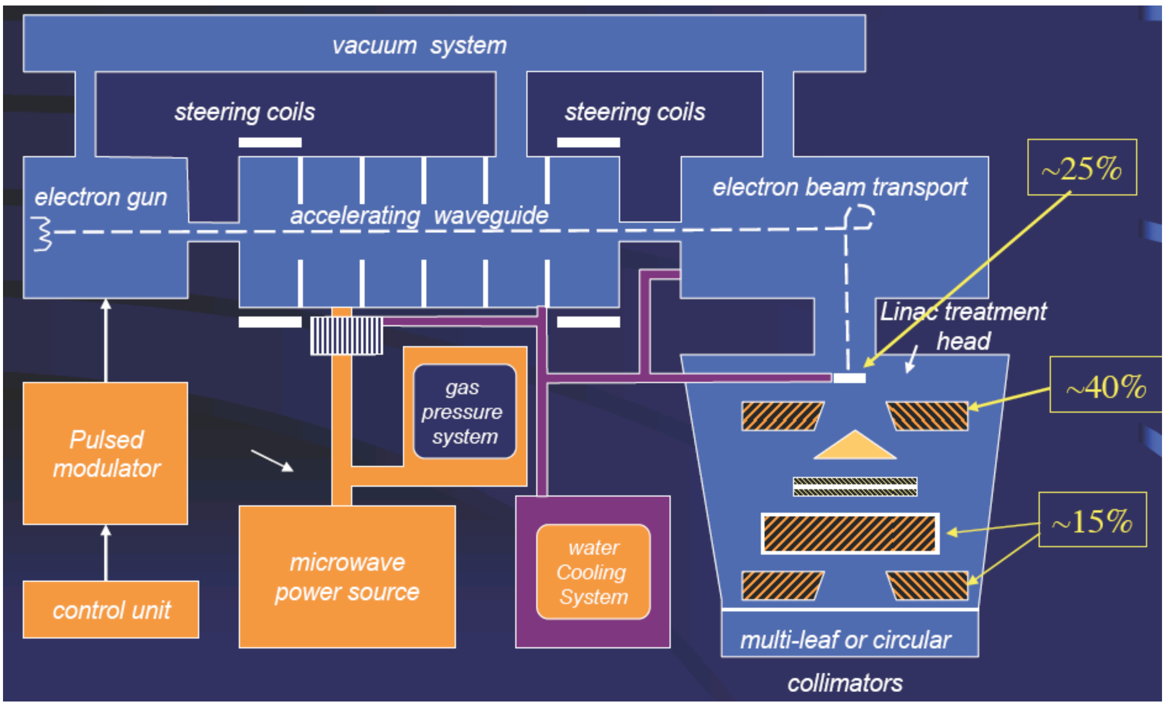


Response of various dosimeters to the high-LET field component at CERF

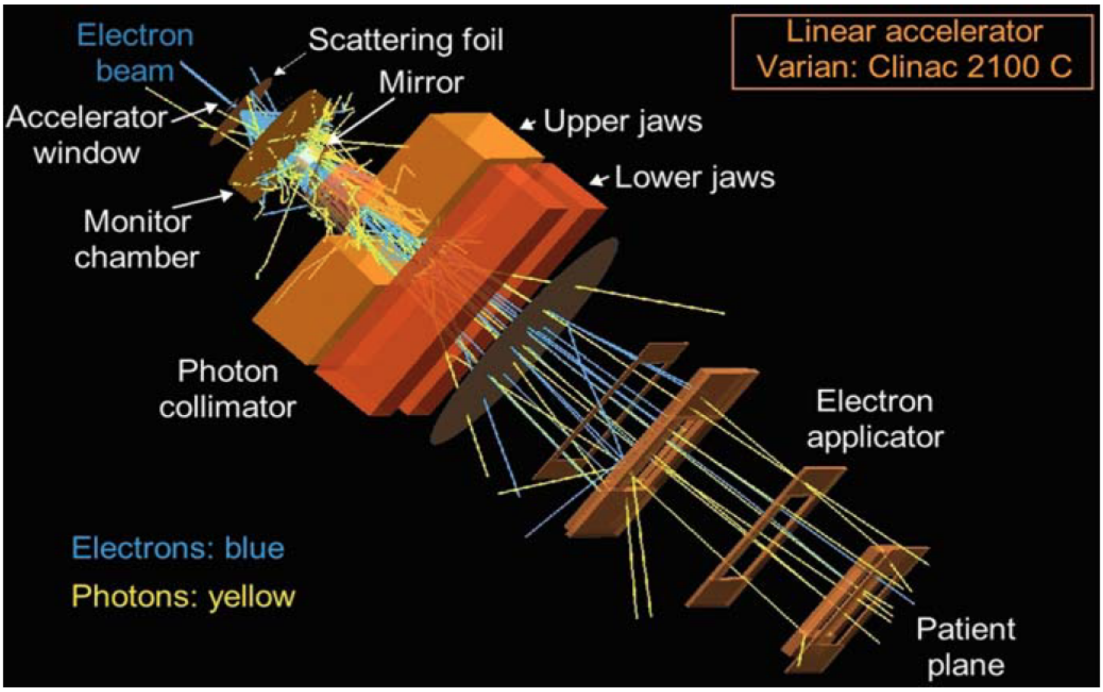
Dosimeter type	Rel. response*
^{6}LiF/^{7}LiF albedo	0.10
CR-39	0.35
NTA film	3.70
Studsvik (cylindrical)	0.53
Berthold (spherical)	0.54
Spherical LINUS (w/Pb)	0.86
Cylindrical LINUS (w/Pb)	0.93
Superheated emulsion	**0.65**
Superheated emulsion w/Pb	**0.95**

*reference values from TEPC

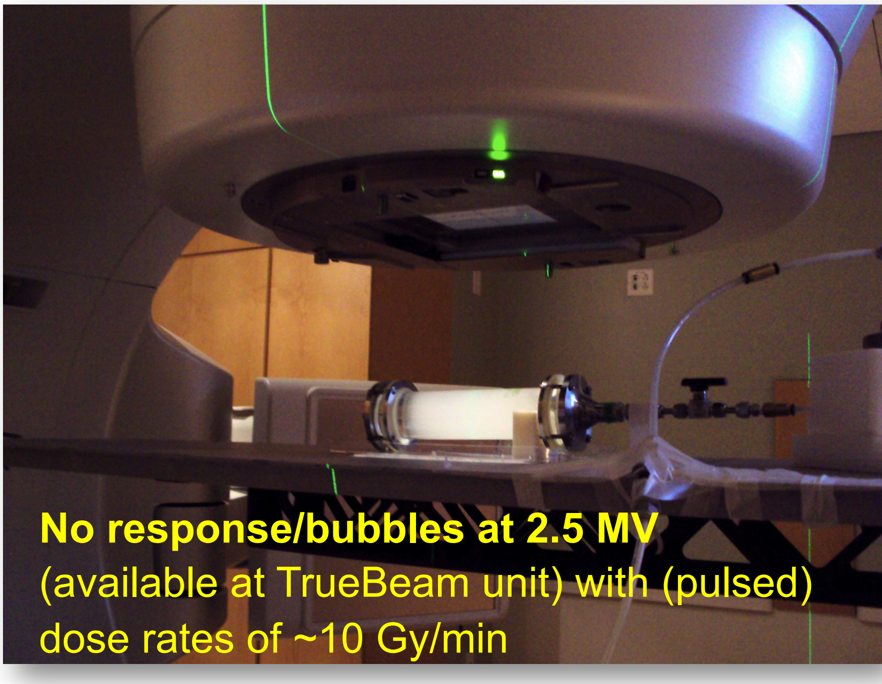
Photoneutron production in accelerator head



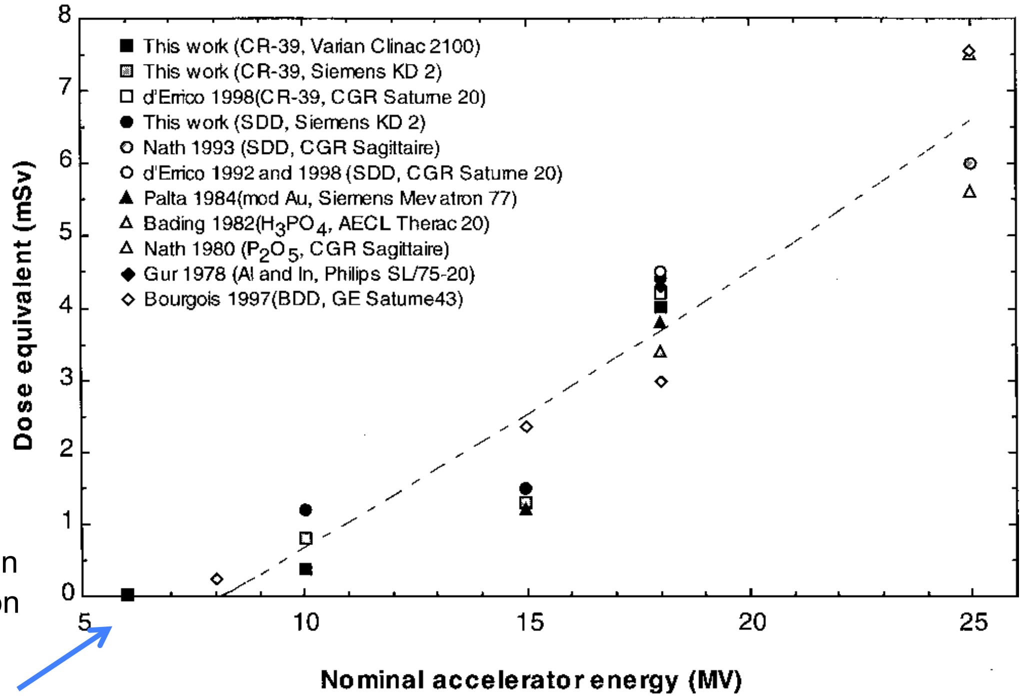
Schematic of a Varian accelerator head



X-ray irradiations at medical LINACs



X-ray tests at medical LINACs



Slight neutron contamination also at 6 MV (likely due to Be window)

Special nuclear material interdiction

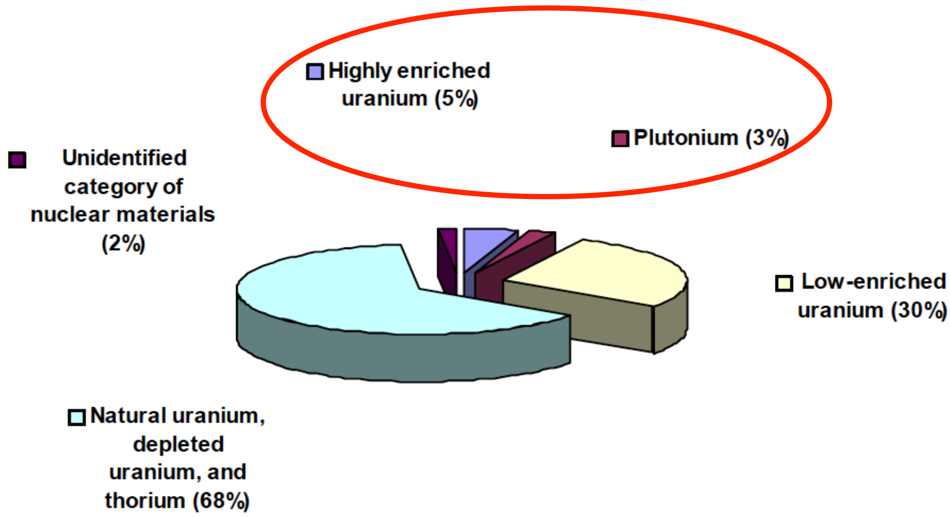
Radiation portal monitors

Radiation portal monitors provide a passive, safe, and effective means to scan traffic and cargo for nuclear and radioactive materials, while maintaining flow of commerce.

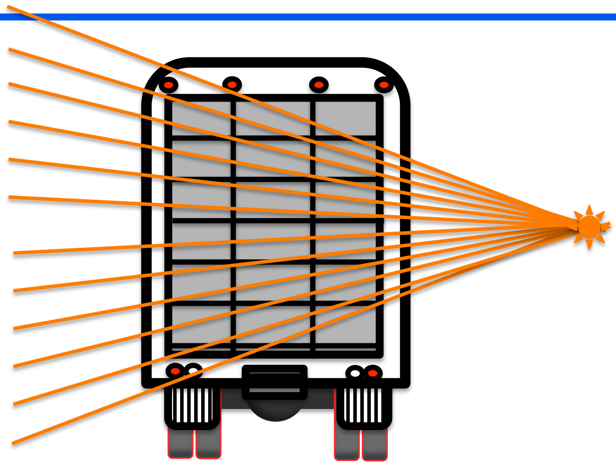
Widely implemented detectors:

- Polyvinyl Toluene (PVT) scintillators for photons
- Moderated ^{3}He counters for neutrons

Distribution of incidents involving nuclear material, 1993-2004 (IAEA)

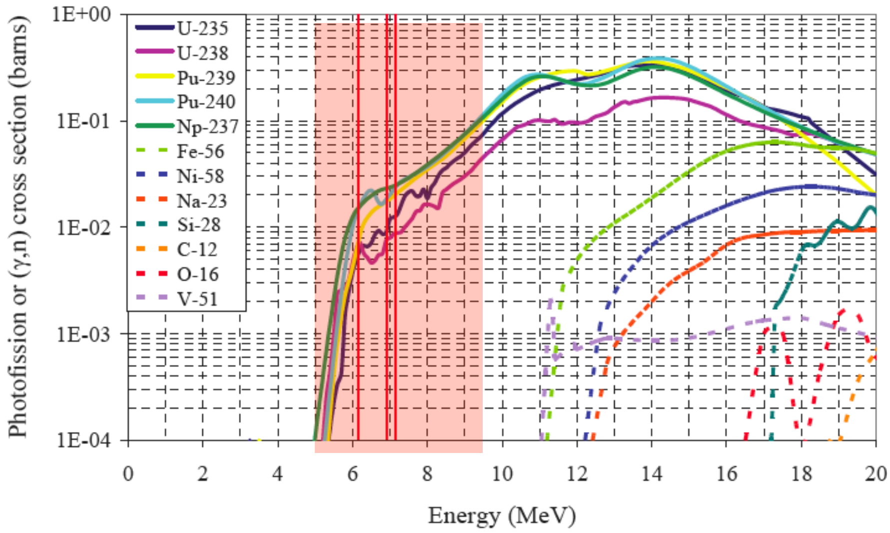


Active SNM interrogation techniques



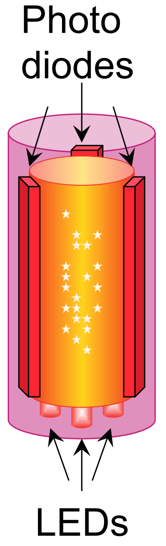
- External beams of neutrons or high-energy X-rays are used to trigger fission reactions.
- Detection of induced fission signatures (prompt and delayed) can be used to confirm the presence of SNM.
- Considered the only viable option to detect the presence of HEU; also effective in the detection of Pu-239

High-energy photon interrogation



- The photofission cross section peaks at about 14 MeV, while below 10 MeV it is < 5% of typical peak values.
- Using 14 MeV γ's would increase the yield of fission neutrons but also the neutron background from (γ,n) reactions in other materials.

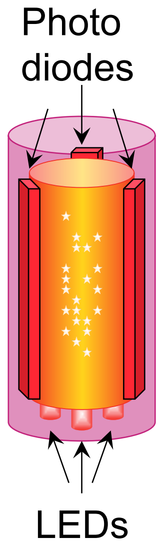
Optical readout based on scattered light

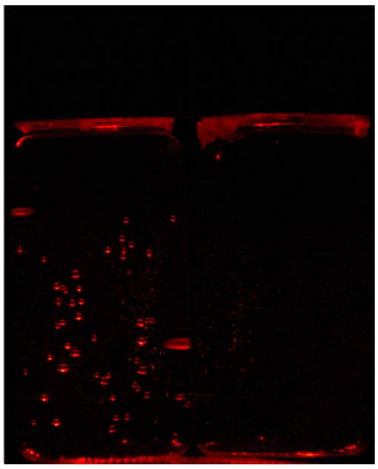


- Instant read out
- Rate insentive
- Position sensitive

Light on

Optical readout based on scattered light

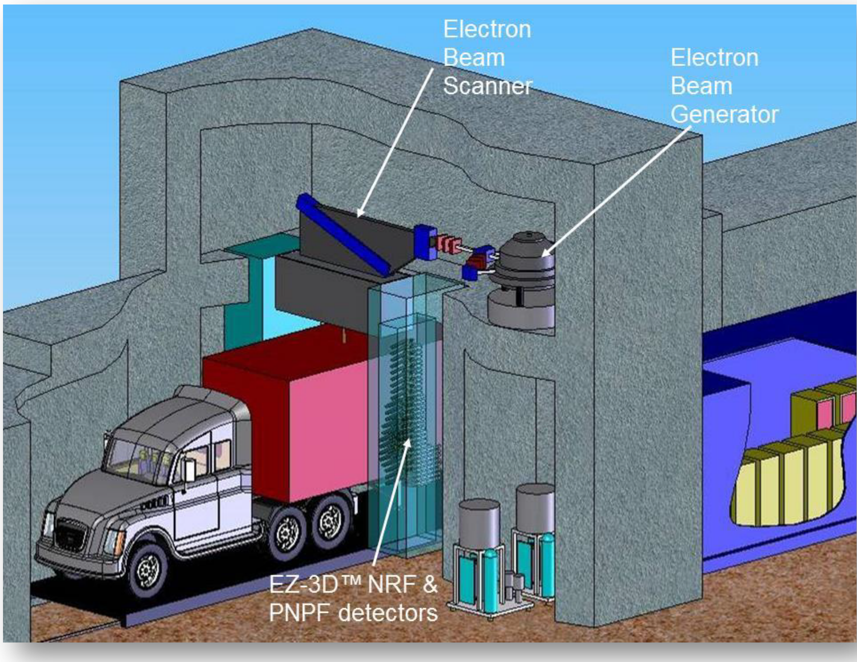




- Instant read out
- Rate insentive
- Position sensitive

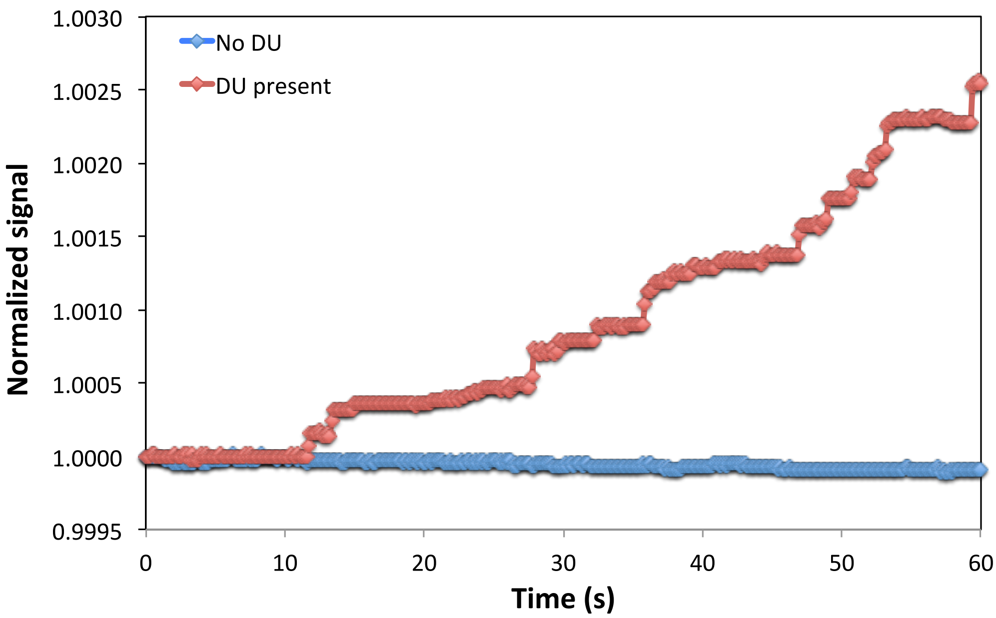
Light off

Active interrogation at Passport Systems

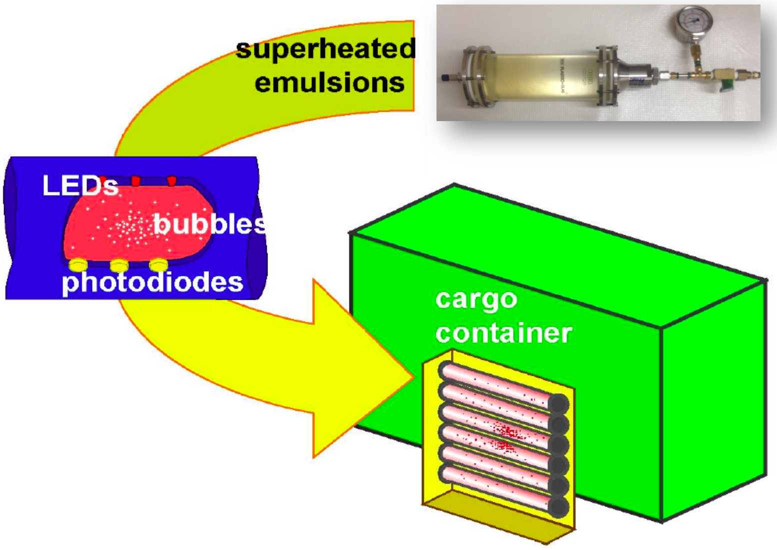


Passport Systems' CW electron accelerator based alarm resolution system

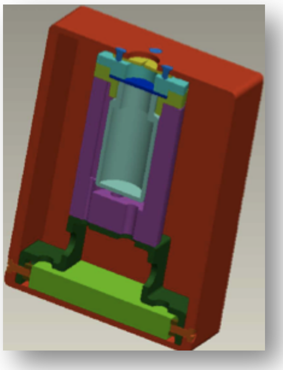
Detection of SNM by optically readout superheated emulsions



Outlook: going big and small



Electronic
personal
dosimeter



Cargo
interrogation
system