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Ra-226 Re-use / Recycle Presentations and Round Table US DOE Isotope Program, IRP

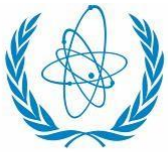
Web Meeting, May 31, 2024

Radiation protection issues associated to design and operation of facilities using Ra-226 for production of isotopic generators for alpha-radioimmunotherapy



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My brief background in relation with Ra-226

- Radiological properties of radium 226
- The radium 226 and radon 222 radiation protection
- Radon 222 measurements methods and examples of equipment
- Alternative to Ac-225/Bi-213: **Lead 212 alpha-immunotherapy: A French Approach but many similar design/operation concerns**
- Some design requirements for a facility dealing, for instance, with with Ra-226
- Design / Operation for process steps, for normal, incidental and accidental situations
- Management of secondary waste
- Conclusion: Safety first



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Outlines

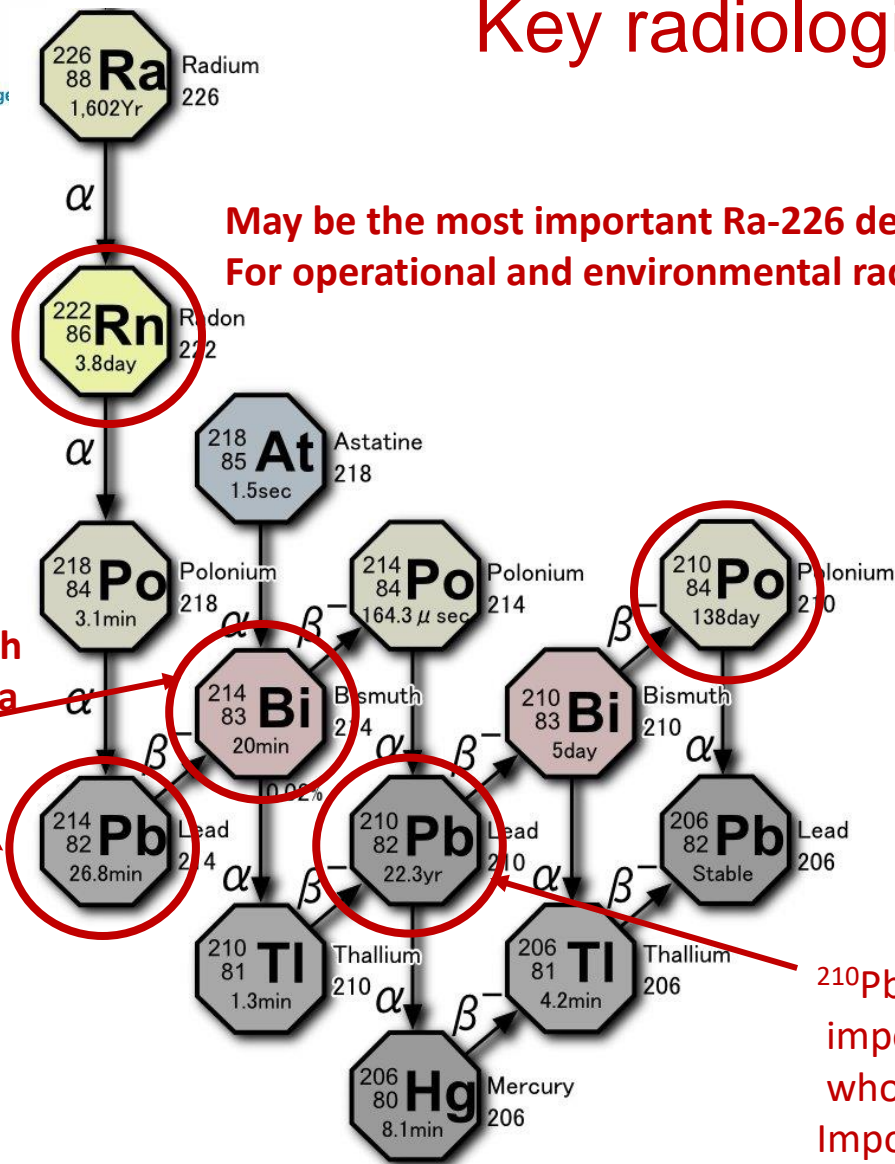
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Radium 226 decay chain: Key radiological properties



May be the most important Ra-226 decay product
For operational and environmental radiation protection

Very high radiotoxicity such as poisons,
according to recent events

Both have high
Energy gamma
rays

^{210}Pb : 22,3 years half-life, but doesn't
impede fast secular equilibrium of the
whole ^{226}Ra chain (few days)
Important as cumulate in air filters



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Radiation protection concerns when dealing with ^{226}Ra from disused sources

Main Ra-226 radiological hazards are:

- External exposure from gamma rays emitted by radium 226 and its progeny

Co-60	7,80E-04
Ra-226	
averaged	3,40E-04
Cs-137	8,70E-05

Unit: mSv/h/MBq at 1m

Gamma constant of Ra-226 in equilibrium with its its progeny

- Internal exposure from:
 - Ingestion of radium 226 (and its decay products)
 - Inhalation of radon 222, especially its daughter products



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Exposition from radium 226 and radon 222

- ❑ Radium 226: alpha emitter (~4.9 MeV) + **185 keV gamma ray** → **external exposure mainly from progeny products when in equilibrium:**
352 keV (38%), 609 keV (46%), 1120 keV (15%) and **1764 keV (15%)**

- ❑ Radon 222: main internal exposure
 - Equivalent dose delivered by radon 222 itself by inhalation *is negligible*
 - **Equivalent dose is delivered by short-lived decay products of radon 222**

- Inhalation of 1 Bq ^{222}Rn exposes an individual to $5.56 \text{ E}^{-09}\text{J}$ (34 710 MeV)
= sum of **potential alpha energies** of its progeny

- Explains high radiotoxicity of radon and related high risk of lung cancer for exposed people such as workers in uranium (or other) mines.



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Exposure from radon 222 and its progeny

Radon 222 decay products				Energy per alpha particle		For 1 Bq of Rn-222	
				MeV	E ⁻¹² Joules	MeV/Bq	E ⁻¹⁰ J/Bq
		Half-lives					
Polonium	218	3,05	minutes	13,69	2,19	3615	5,79
Plomb	214	26,8	minutes	7,69	1,23	17850	28,6
Bismuth	214	19,9	minutes	7,69	1,23	13250	21,2
Polonium	214	164	Micro-seconds	7,69	1,23	2,0E ⁻⁰³	3,0E ⁻⁰⁶
Total						34715	55,6



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Radon 222 measurement methods (very summarized)

- Sampling radon on a short time (~1 hour) in a specific place; **scintillation cells** (with ZnS), or **gamma spectrometry** of sample taken in a standardized container, **alpha counting**
- Integrated measurement methods that needs a longer sampling time ~1 week to integrate daily variations of volumic concentration: **Solid State Nuclear Trace detectors, use of charcoal then liquid scintillation counting or gamma spectrometry, electrostatic devices,**
- Continuous sampling and measurement, which should be adapted to the dynamics of radon generation and transport phenomena: **ionization chamber (gaz circulation, or diffusion) or gamma spectrometry; sampling on filter and alpha spectrometry**
- *Methods are codified in national and international standards*

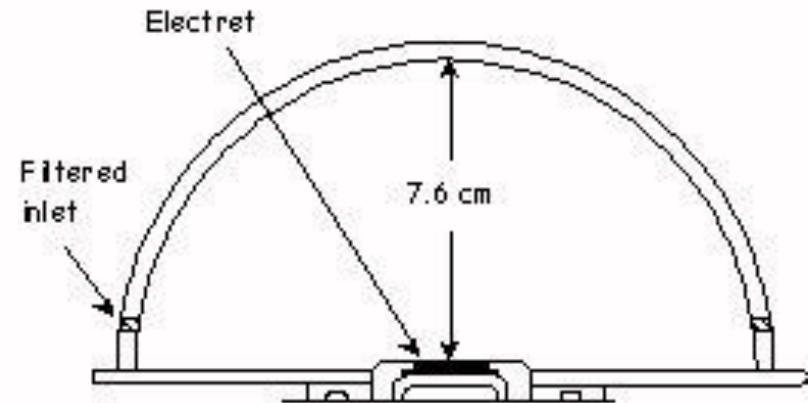


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Passive Radon Measurement Methods

Passive monitoring techniques (Solid State Nuclear Trace detectors [SSNTD](#) and ion chamber detectors) and continuous monitoring techniques (Solid State [Silicon](#) and [Germanium](#) Detectors).

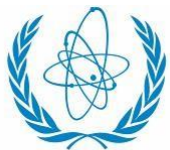


Electret ion chambers are commonly used **for passive** radon measurements.

Two types of E-PERM devices (the S- and H-chambers) are used (see www.radelec.com)



Kit for Radon-222 spot measurement using scintillation flasks



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Radiation protection: Active measurements of radon 222 - Some examples:

- ✓ Continuous measurements for Rn-222 (Radonova)



Corentium: Alpha spectrometry as Rn-accumulative method



AlphaGuard spectrometry as Rn-accumulative method
Detector: 0.62 L pulsed ionization chamber +
Alpha spectroscopy



ATMOS (radonocva) pumps filtered air into a pulsed ion chamber

Many other equipment exist!

Radiation protection: Active measurements of radon 222 - Some examples of equipment

✓ Portable/personal electronic dosimeters



e.g. the AlphaE (Radonova)
3 pulses/hour at 100 Bq/m³ thus
compatible with the 300 Bq/m³ limit



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Exposure from radium 226

Radiation protection means for process steps

- ✓ External exposure: use of shields, e.g. some process steps in a shielded glove box or a 'light' hot cell depending on the ^{226}Ra source term (depending on maximum activity used in a single step).
- ✓ Glove box should be shielded, e.g. with leaded plexiglass panels.
- ✓ Glove boxes / hot cells should have nuclear ventilation and filtration (ISO 17873:2004).
- ✓ Filtered air should be released in a stack to dilute radon in order to limit dose impact to the reference (critical) group.
- ✓ In many countries, it is mandatory to quantify amounts of gaseous (Rn-222) releases



Exposition from radium 226 and radon 222

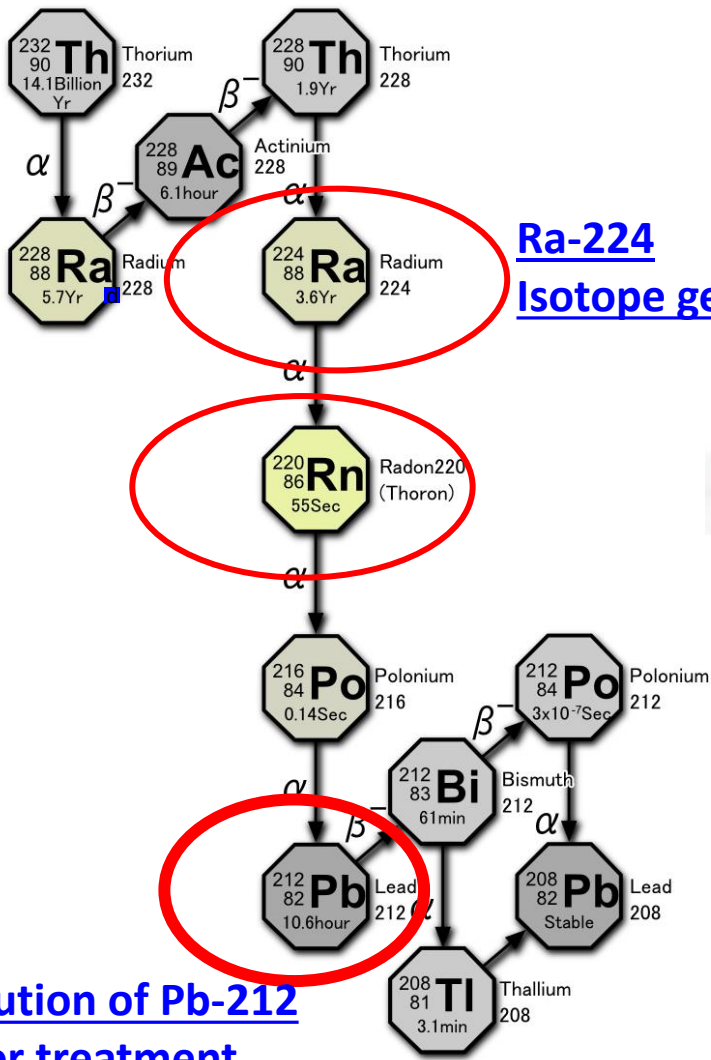
Radiation protection for process steps

- ✓ In most cases, Personal Protective Equipment (PPE) will be limited as the main effort is put on collective radiation protection.
- ✓ Personal dosimetry: passive and active → **mandatory**, even for radon dosimetry when needed
- ✓ Permanent control in the process premises of:
 - ✓ Airborne alpha-beta contamination,
 - ✓ Airborne radon 222 volumic concentration (recommended maximum level: 300 Bq/m^3 , if no specific radon PPE)
 - ✓ Ambient gamma dose rate at safety relevant places



Alternative to Ac-228: Pb-212 alpha-radio immunotherapy

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Elution of Pb-212 for treatment

AdvanCell Isotopes ²¹²Pb Generator



Ra-224/Pb-212 generator from US DoE





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Example of the Maurice Tubiana Laboratory in France

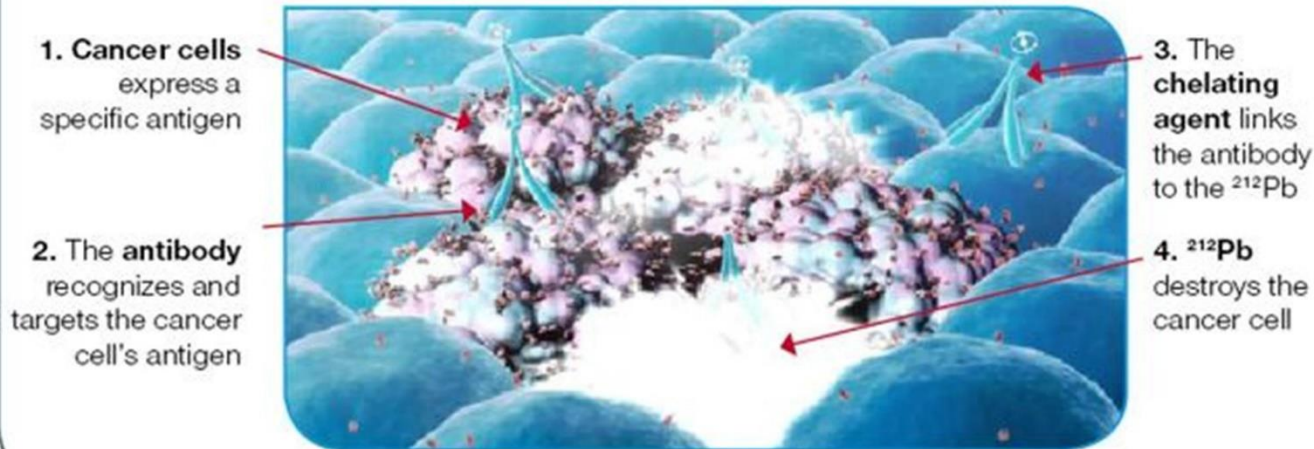
- French Orano Group has developed alpha-radio immunotherapy based on Lead 212, daughter product of Th-232 → Ra-228 → Th-228b → .. → Ra-224 generator to deliver Pb-212 to the patient
- Reason: After the 2ndWW, France used monazite ore to develop the French Civilian and Defence nuclear programs based on uranium, but large amounts of Th-232 were generated as a by-product and stored at CEA facility.
- Process was developed by highly skilled chemists in La Hague spent fuel reprocessing Plant
- Pre-pilot plant operated in Bessines Orano site (former uranium mine) for production of the first Ra-224 generator to start human in-vivo testing in the US (Alabama)
- Pilot production plant built at Bessines (Laboratoire Maurice Tubiana)
- Production plant built in Texas (US)

Alpha-Radioimmunotherapy with Lead 212

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How ^{212}Pb Works

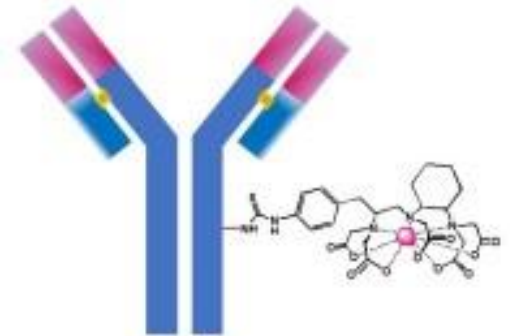
An example of TAT: Radioimmunotherapy using ^{212}Pb compounded with an antibody to attack and destroy cancer cells



Antigen: A substance which provokes an immune response.

Antibody: Recognizes and targets cell-specific antigens on cancer cells.

Chelating Agent: A "molecular cage" used to attach isotopes to monoclonal antibodies.



Alternative to Ac-228: Lead 212 alpha-immunotherapy The *Maurice Tubiana* Laboratory in France



Stack for Rn-220 dilution

Limited PPE for workers as main effort is put on the shielding and **confinement** of the process equipment





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Example of the Maurice Tubiana Laboratory in France Quality control laboratory





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Exposition from radium 226 and radon 222 Radiation protection Ra-226 DSRS dismantling operations

Dismantling of ^{226}Ra sources and Ac-228 separation/purification performed in close and ventilated equipment: need for shielded glove box(es)

**No ventilation
extraction**



➔ Shielded walls (leaded plexiglass,..)

➔ Nuclear ventilation (ISO 17873) to:

- Filter radioactive particles, aerosols,
- Extract radon and drive it to an exhaust (stack), and quantify radon releases





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E.G. of Some design requirements:

Waste disposal sinks and Drainage Pipes



✓



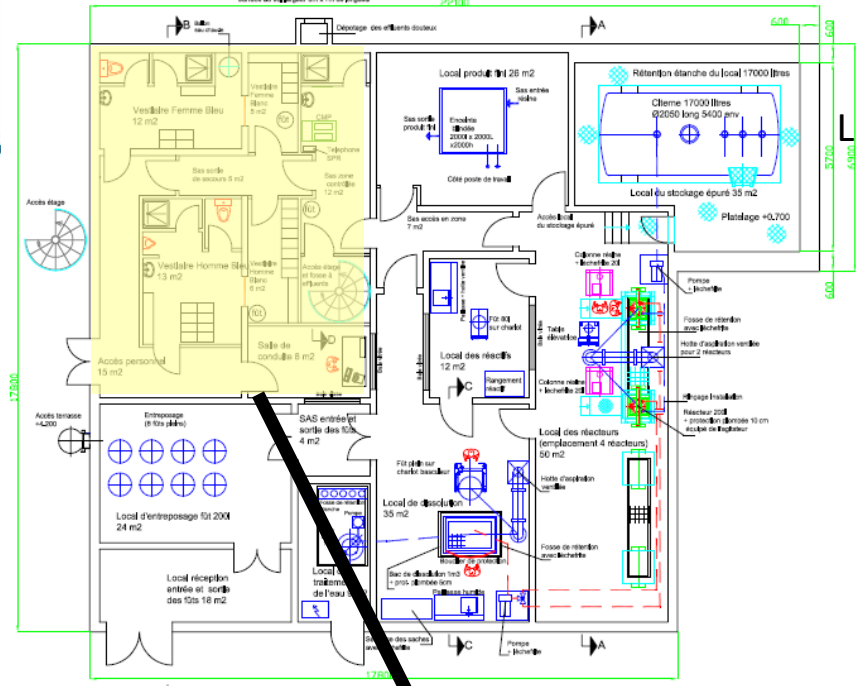
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Use of absorbent paper on benches and in glove boxes





Prendre une aire de stockage pour combler la surface au sol pour 3m x 7m de longeur

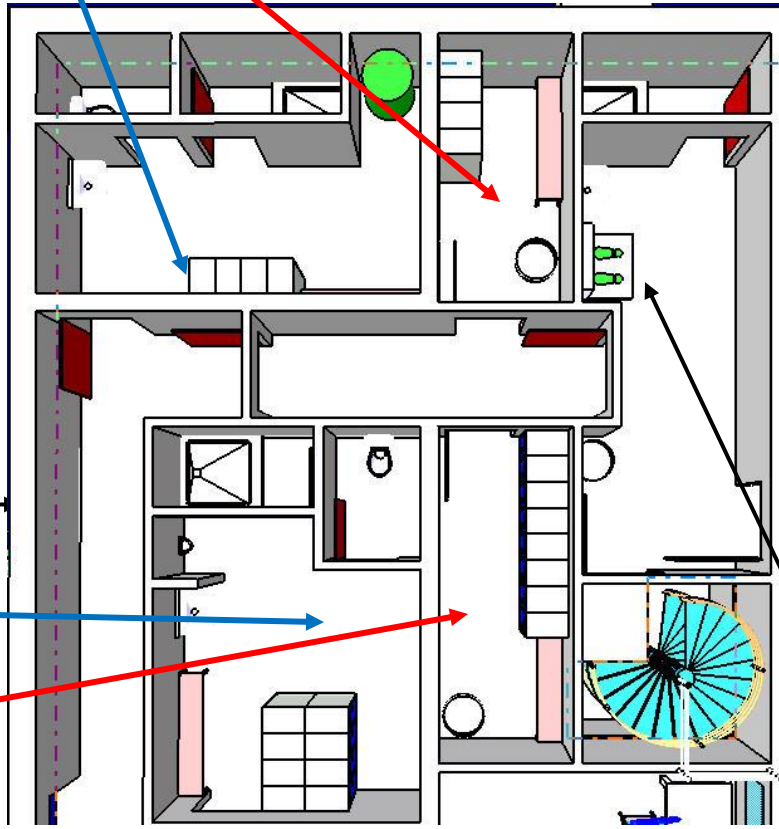


Level 0 (1)

Locker rooms in a radiochemical laboratory

Cold / Blue changing room for women

Hot / White (active) changing room for women



Changing rooms

Cold / Blue changing room for men

Hot / White (active) changing room for men

radiological monitor hands-foot-clothes



Design for process: incidental and accidental situations – some tips

Incidental/accidental situations – examples:

- If cut-off of electrical supply => static confinement for some equipment (glove boxes/hot cells); use of diesel generators
- Break-down of a glass container, ion exchange resin column, etc. trays to recover liquid spills, materials/surfaces easy to decontaminate
- Emergency teams with special PPE, special intervention trainings,
- Fire: inform/train the fire brigade to specific hazards/specific procedures/equipment needed

All these situations should be:

- ➔ described in the main safety documents SAR/EIA/safety case
- ➔ emergency plan/procedures



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Management of secondary waste

Secondary waste will be generated in the processes:

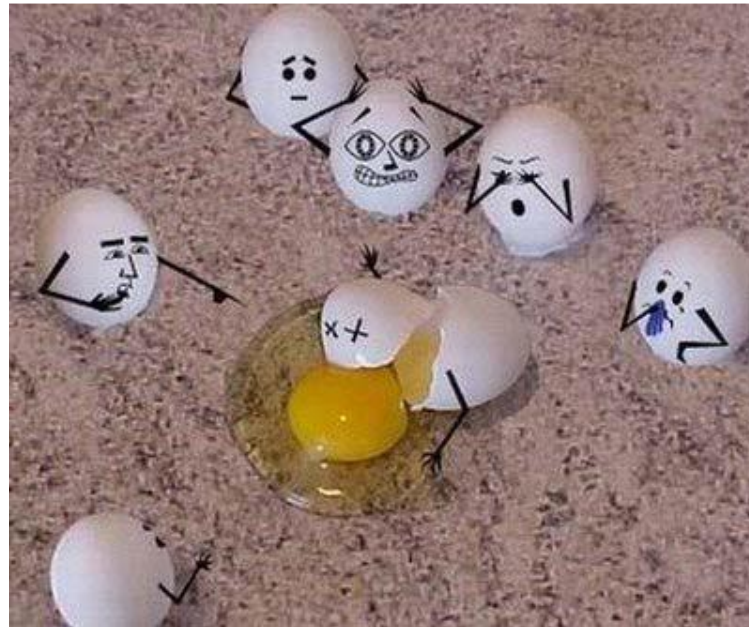
- envelopes/debris from dismantled Ra-226 sources
 - at the irradiation facility premises (targets, other)
 - Process waste: spent IER columns, laboratory glass/other equipment, wipes, etc.
 - Waste from cleanup of glove boxes / hot cells, etc.
 - Spent HEPA filters,
 - All items resulting from implementation of **Good Manufacturing Practices** and sound radiation protection practices
-
- **Secondary waste should be:**
 - Packed e.g. in plastic bags, then in 100 L or 200 L drums
 - Characterized, e.g. by gamma spectrometry and/or by the dose rate conversion method (no sampling recommended)
 - Stored in a ventilated area (radon 222 issue for Ra-226 bearing waste)



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Safety First!



Thank you for your attention