Radon Assay and Purification Techniques

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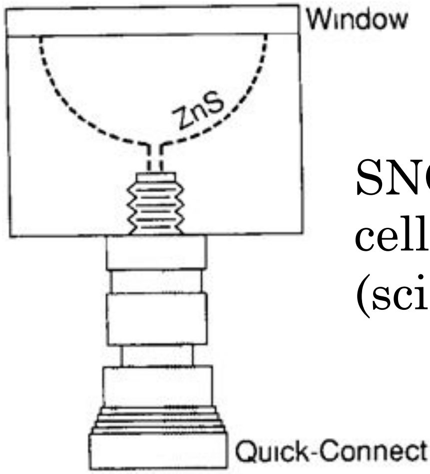
Low Radioactivity Techniques 2013

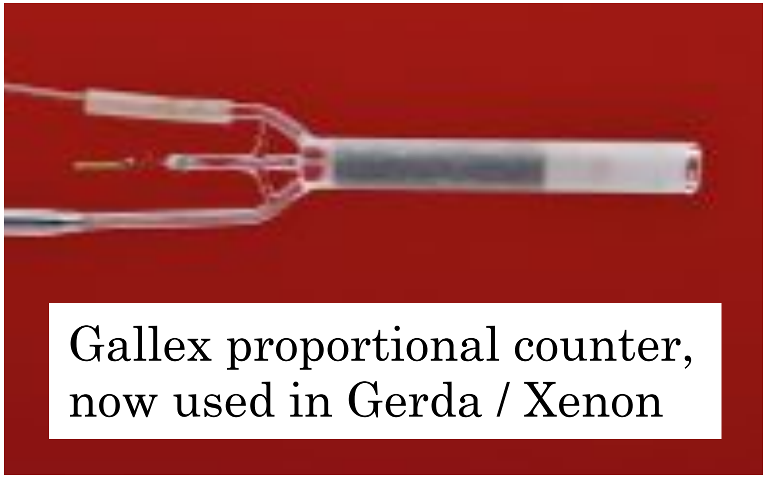
Radon Facts

- Radon …
  - … is radioactive (no stable isotopes).
  - … is everyvery (as U/Th traces are everywhere).
  - … is noble (good diffusion, no chemistry).
  - … is different (the only gaseous heavy element).
  - … is threefold in nature (^{222}Rn, ^{220}Rn, ^{219}Rn).
  - … has no sons, but many (radioactive) daughters.

Low Background Counting Techniques that require Sample Preparation



SNO Lucas cell (scintillation)

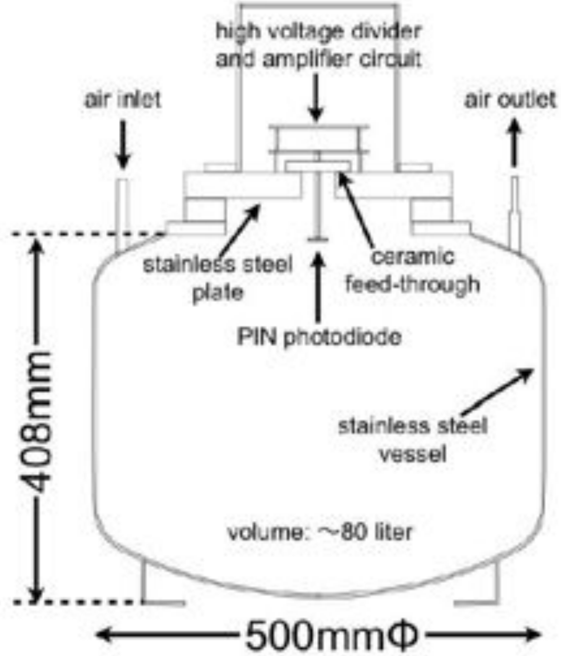


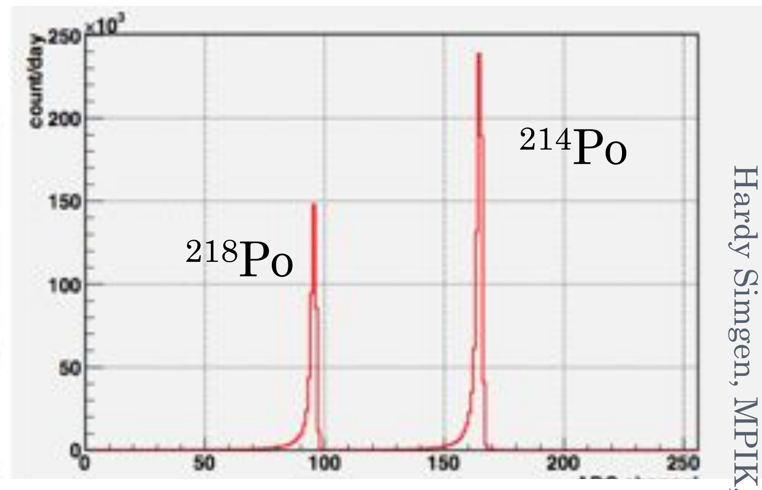
Gallex proportional counter, now used in Gerda / Xenon

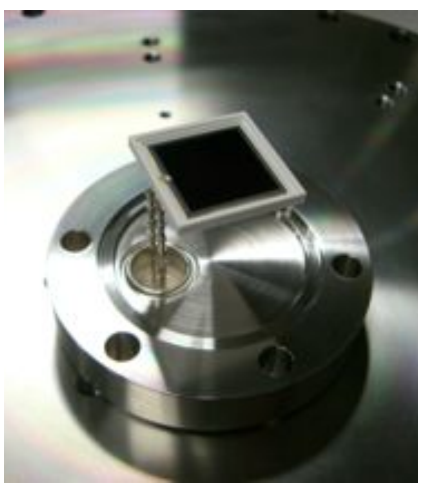
- CONs:
  - Work-intensive sample preparation without radon pollution and without radon losses.
  - Limited to ^{222}Rn.
- PROs:
  - Lowest possible background: Few ^{222}Rn atoms detection limit.
  - Universal: solid, liquid and gaseous samples.

Electrostatic Chambers

Pictures provided by Y. Takeuchi (XMASS)

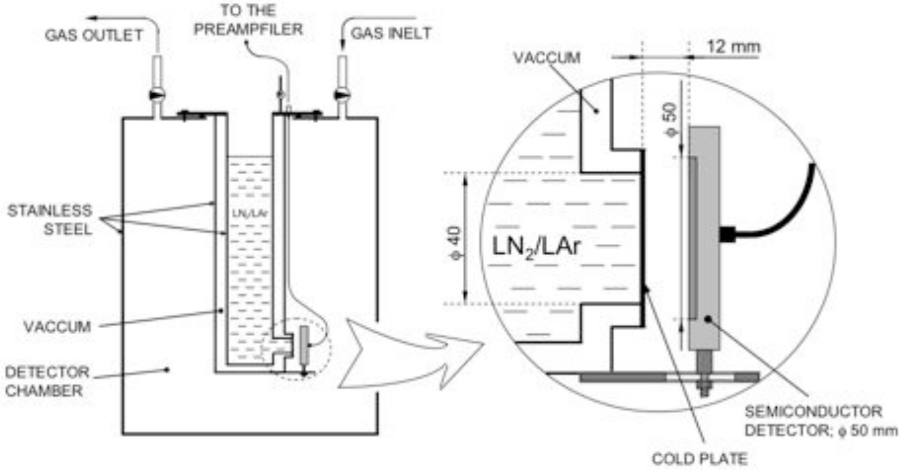






- Good energy resolution
- May be used for ^{222}Rn, ^{220}Rn and ^{219}Rn
- Commonly used in our field:
  - Super-K: NIM A 421 (1999) 334
  - SNO: NIM A 421 (1999) 601
  - Borexino: NIM A 460 (2001) 272
  - NEMO, EXO, XMASS, Gerda, …

CRYOGENIC RADON DETECTOR



- Detector : ORTEC diode, 40 mm diameter
- Cold plate : 30 mm diameter,
- Cooling : LN_2/LAr
- Cryo-vessel : 12 L
- Chamber : ~1 L, metal-sealed
- Material : Stainless steel

- Detection efficiency : 24 %
- Resolution (5.5 MeV) : 56 keV
- Background (^{222}Rn) : ~0.8 cpd
- Detection limit : ~20 µBq

Information provided by G. Zuzel (Cracow)

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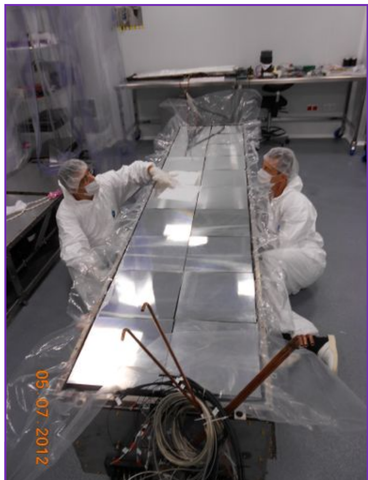
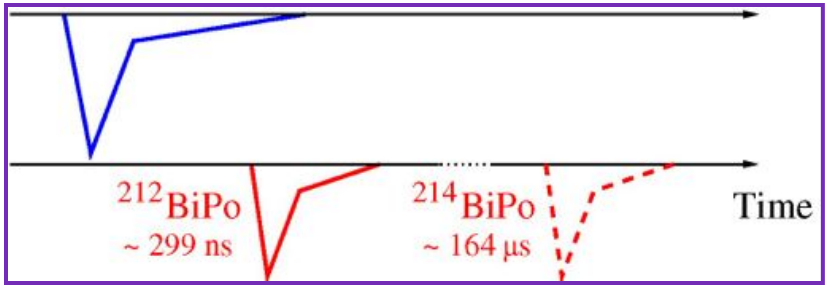
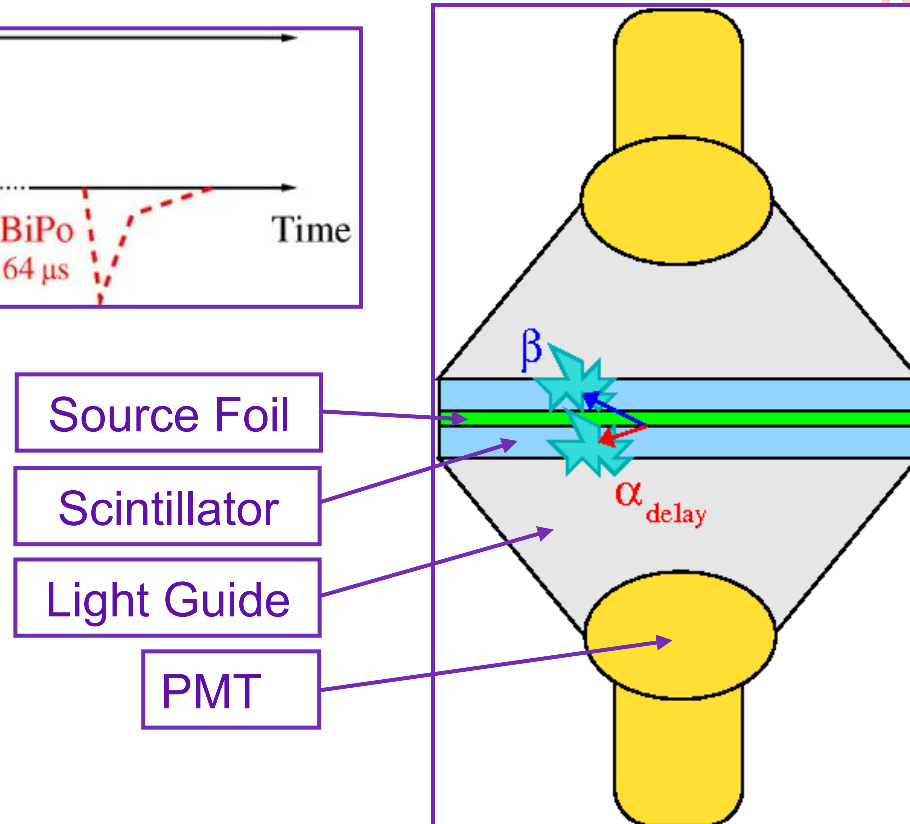
The BiPo Detector for ^{208}Tl and ^{214}Bi Detection in the SuperNEMO Source Foils.

Slide provided by H. Gómez (SuperNEMO)



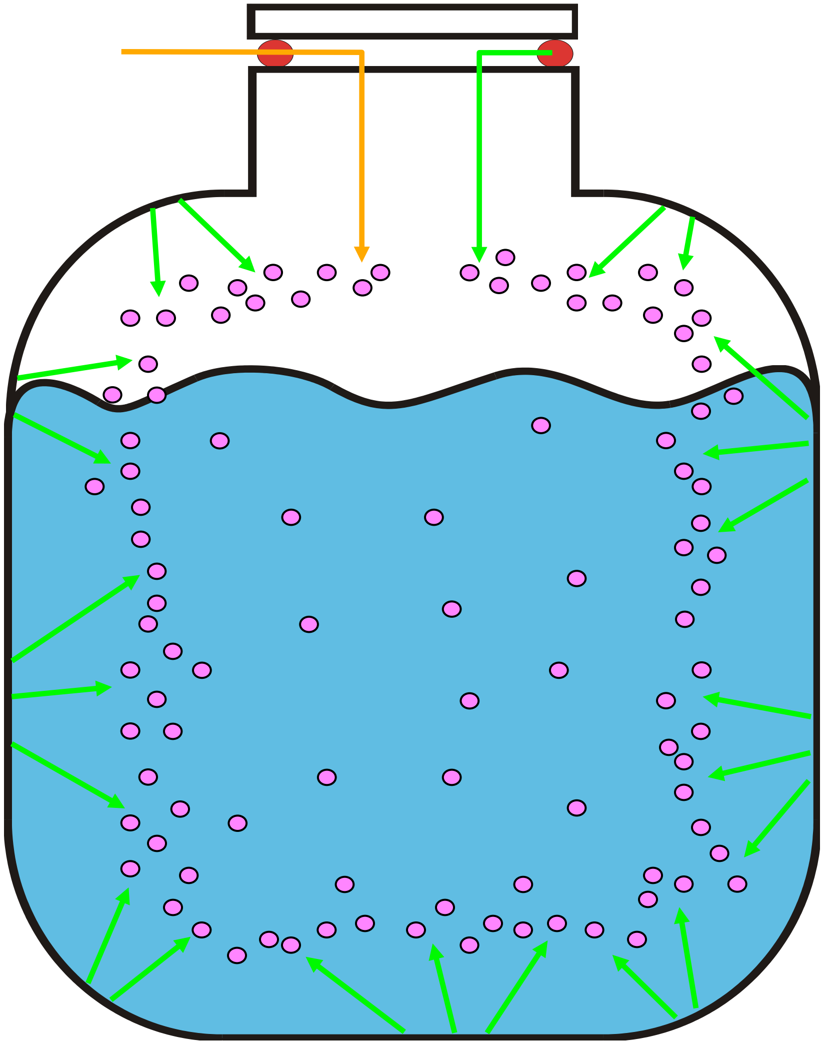




- Detection principle → BiPo $\beta - \alpha$ delayed coincidence detection

Radon Sources in your Experiment

- Intrinsic radium (uranium / thorium) contamination
- Diffusion through seals
- Emanation from vessel and instrumentation

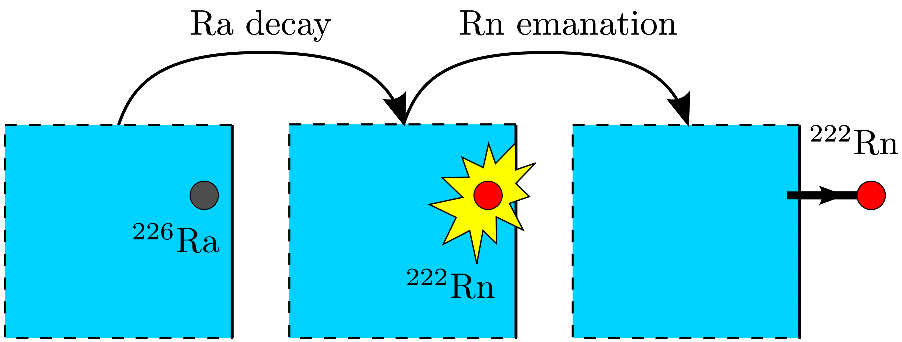


Example: The GERDA LAr Stainless Steel Cryostat

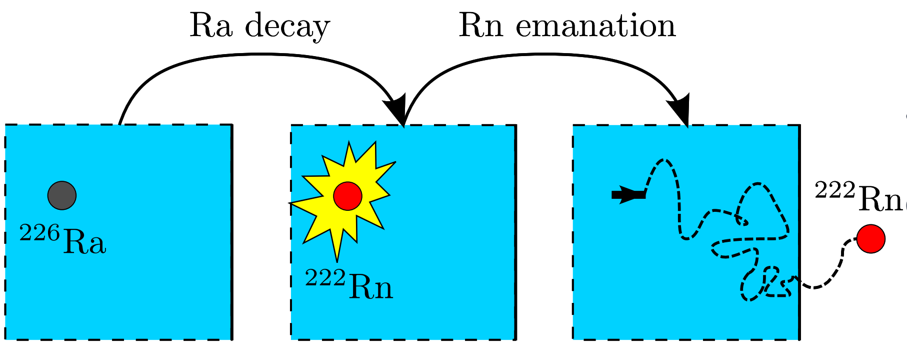
No.	Date	Description	Average [mBq]
1	Nov 07	After construction and cleaning, no N_2 mixing	23.3 ± 3.6
2	Mar 08	After additional cleaning	13.7 ± 1.9
3	Jun 08	After Cu mounting	34.4 ± 6.0
4	Nov 08	After wiping of Cu / steel surfaces.	30.6 ± 2.4
5	Sep 09	After mounting of shroud, manifold, compensator, and cryogenic tubing	54.7 ± 3.5

- see Eur. Phys. J. C 73 (2013) 2330

RADON EMANATION



a) Recoil driven emanation



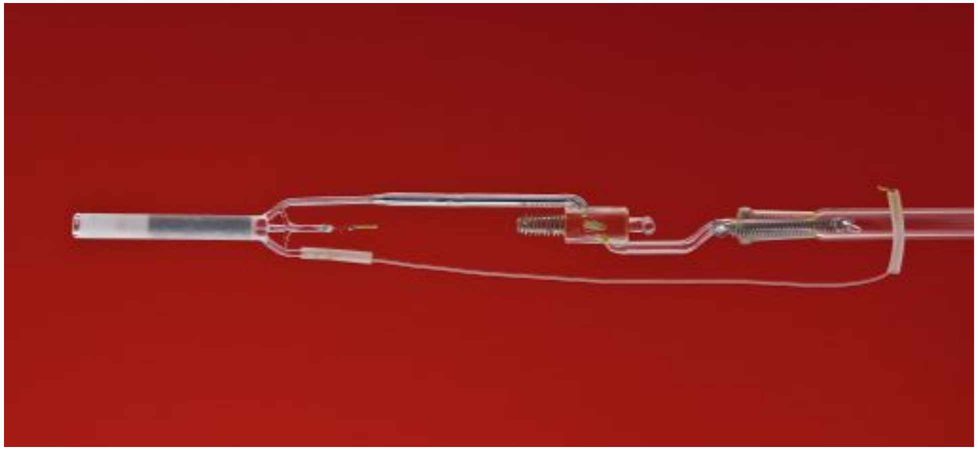
b) Diffusion driven emanation

- Two Steps:
  - Radon generation by radium decay
  - Radon release by recoil or diffusion.
- Diffusion only relevant for “soft” materials (negligible for metals).
- For high purity materials: Emanation due to bulk impurities (gamma ray spectroscopy) usually negligible → Surface impurities crucial!

Material Screening with the Radon Emanation Technique

- Complementary to bulk activity screening techniques (HPGe spectroscopy, mass spectrometry, NAA, …)
- Depending on environment: Humidity, Temperature, …
- Emanation efficiency strongly dependent on radon diffusion in the sample and surface structure: Variations between $<0.01\%$ and $\sim 30\%$.
- Becomes more and more important in our field (e.g. noble gas experiments).

^{222}Rn Emanation Measurements at MPIK Heidelberg



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Laurentian-EXO Rn Counting Facility

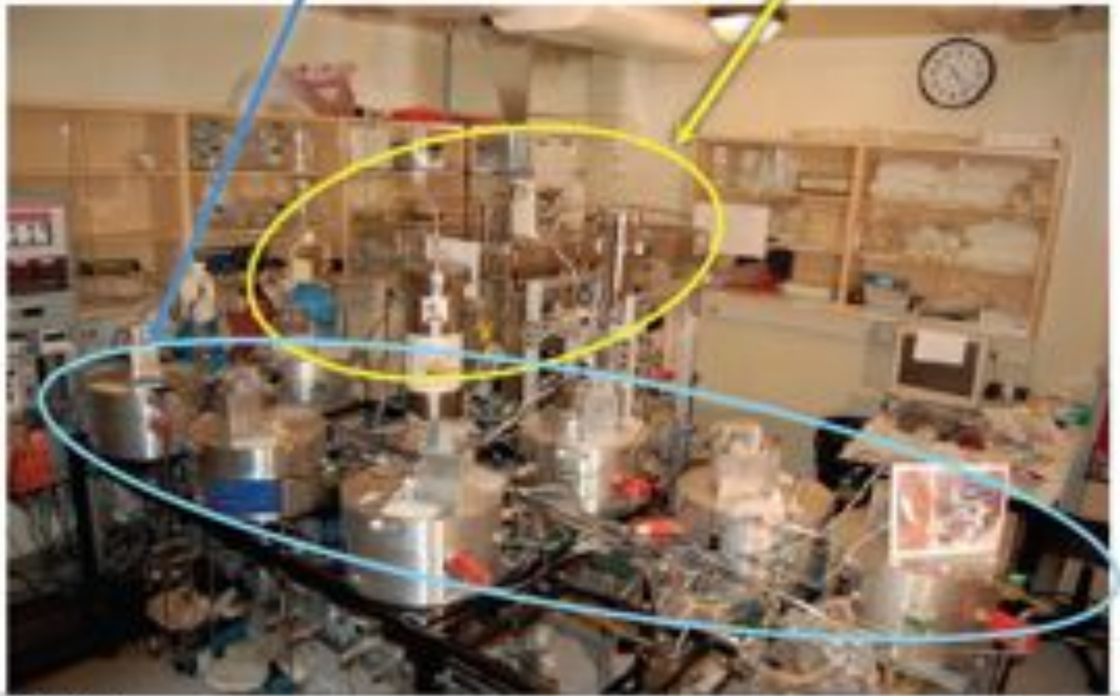
Maintain+operate 8 low background ElectroStatic Counters:

- Built+used for SNO, converted to lower bgnd for EXO
- Sensitivity ~10 ^{A}Rn/day (A=222, 220, 219)
- 1 ESC in use at WIPP (mat. screening + Rn barrier monitoring)
- 7 ESCs in use at SNOLAB:

  6 for mat. screening, 1 for Rn trap work)

Facility on hold since Oct'12 to fix issue with room air quality. Restart May'13

Slide provided by J. Farine (EXO)



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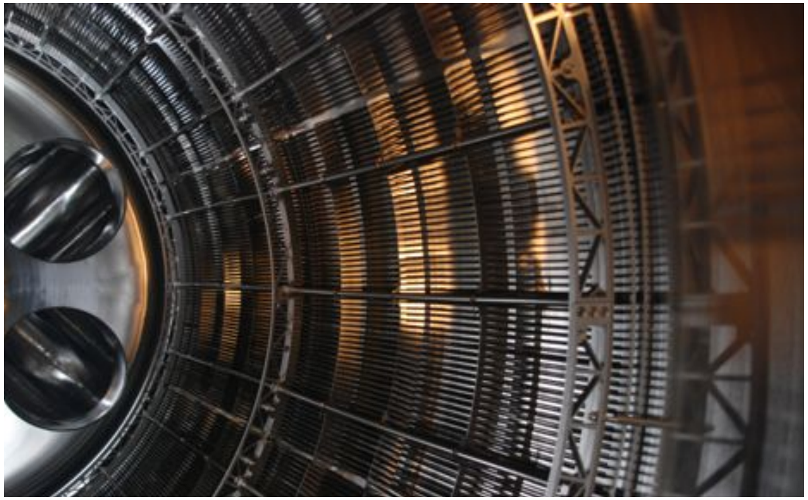
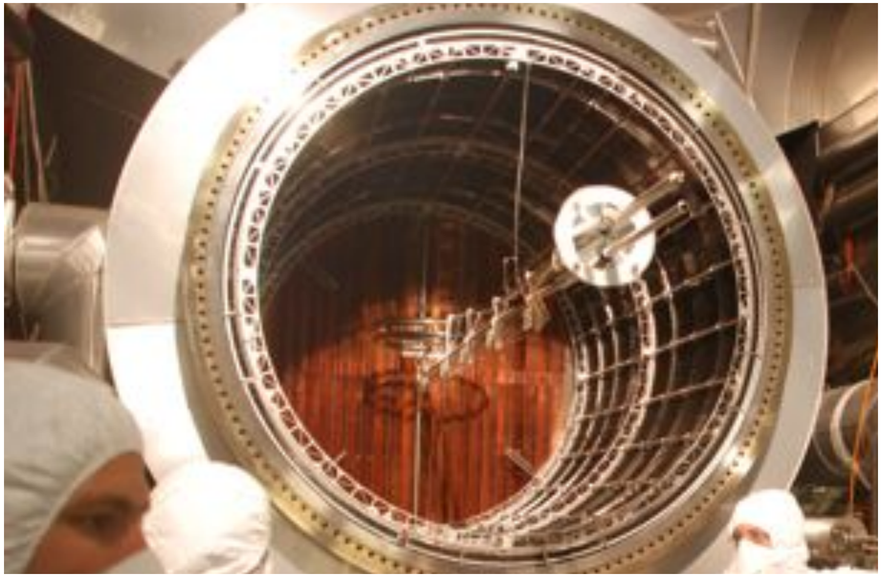
KATRIN Getter Radon Emanation





- Getter material is Zr alloy which contains traces of radioactivity.
- Typically very particular disequilibrium of radioactivity in Zr: $^{231}Pa > ^{227}Ac\ (= ^{223}Ra) > ^{235}U > ^{226}Ra$

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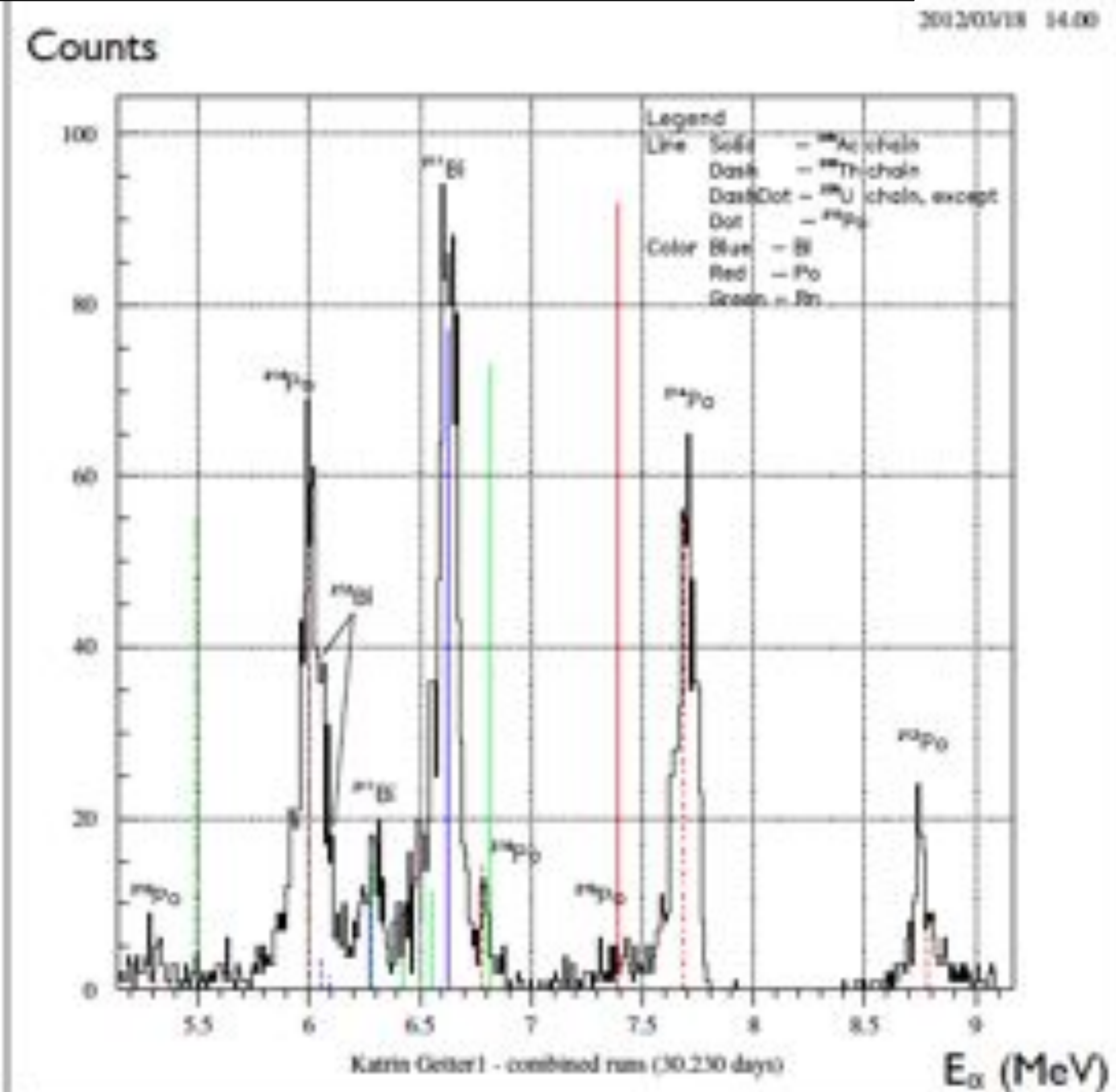
Slide provided by J. Wolf (KATRIN)

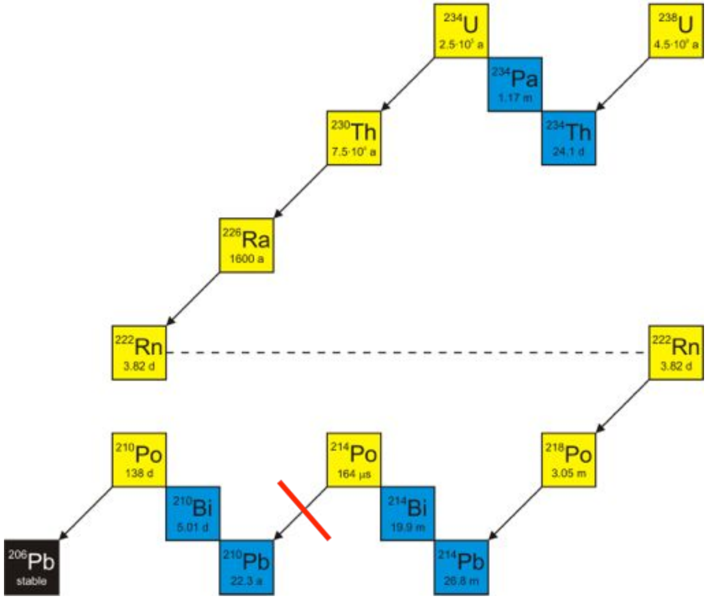
KATRIN Getter SAES ST-707

Slide provided by J. Farine (Laurentian)



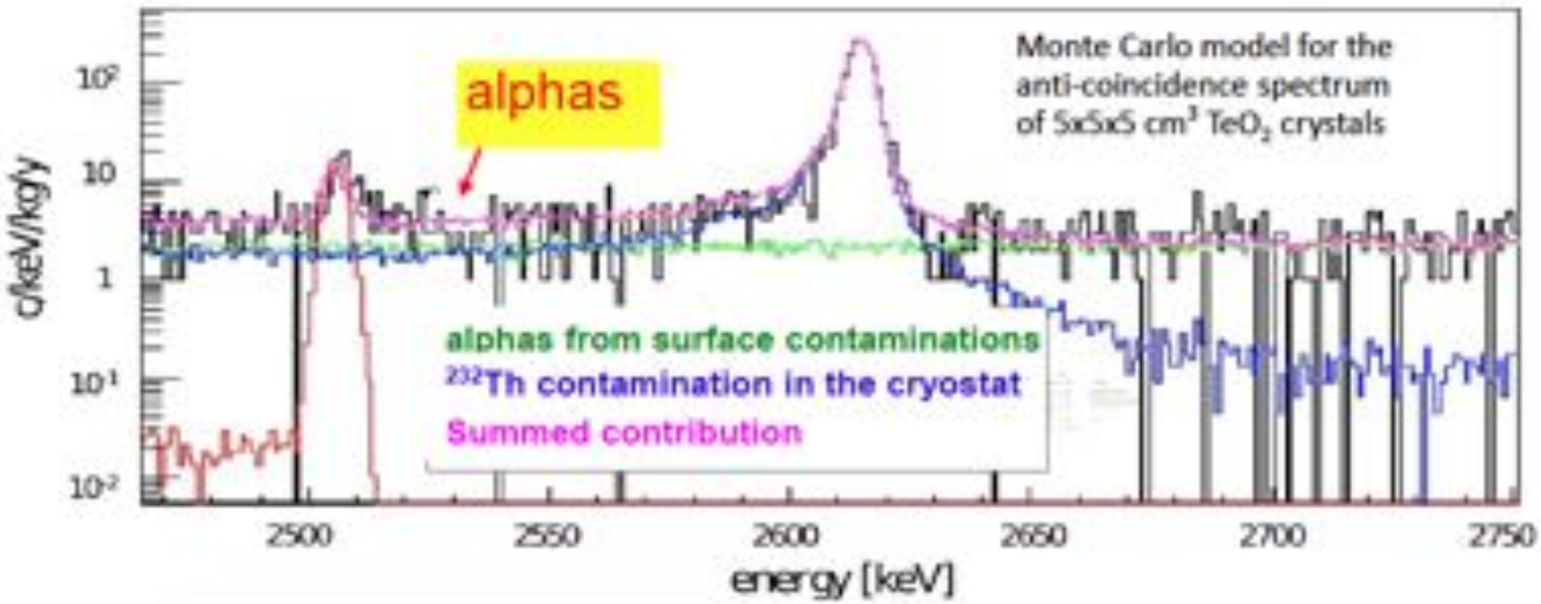
- Container design and construction at KIT
- Design optimises the transport of ^{219}Rn (24% avge)
- 30m of getter placed inside
- Shipped to Sudbury
- Counted for 30.230 days in flow-through mode
- Strong Ac signal (^{219}Rn)
- E scale set by U peaks
- E scale consistent throughout
- Relative intensities also consistent

^{222}Rn Daughters



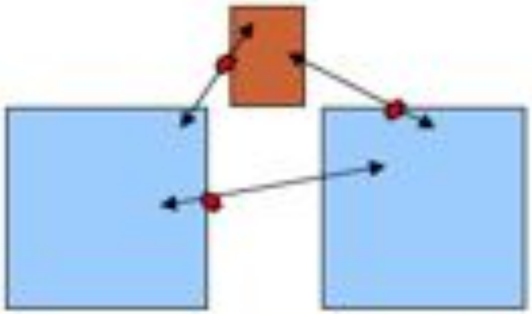
- ^{222}Rn decay chain broken at ^{210}Pb (t_H = 22.3 y).
- ^{210}Pb, ^{210}Bi, ^{210}Po may be present without direct support by ^{222}Rn.
- Deposition from ambient air.
  → Exposure history crucial.
- Only weak/low energy gamma emitters present in last part of ^{222}Rn chain. → hard to detect.
- Poorly understood chemistry (in particular ^{210}Po).
  → difficult to develop dedicated cleaning procedures.

The Background in the Cuoricino ROI



b = 0.169 ± 0.006 c/keV/kg/y

^{232}Th in the cryostat (γ) [1,2]	30 ± 10 %
Contamination on **crystal surface** [1,2]	10 ± 5 %
Contamination on **Cu surface** [1,2]	50 ± 20 %



The main issue are degraded alphas from crystal and copper surfaces

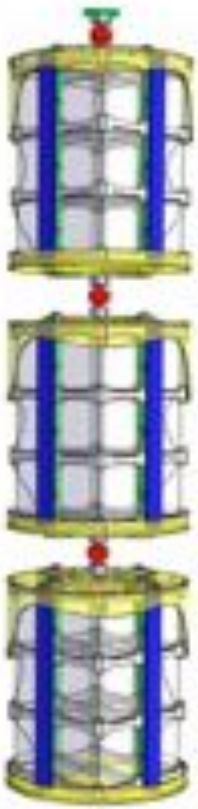
[1] C. Arnaboldi et al., Phys. Rev. Lett. 95, 142501 (2005)
[2] C. Arnaboldi et al., Phys. Rev. C 78, 035502 (2008)

Slide provided by A. Giuliani (CUORE)

Background from Cu: TTT test

Bolometric test to compare the effect in the ROI of **3 different copper surface treatments**

Crystals from Cuoricino array **fully reprocessed** according to the new CUORE standards

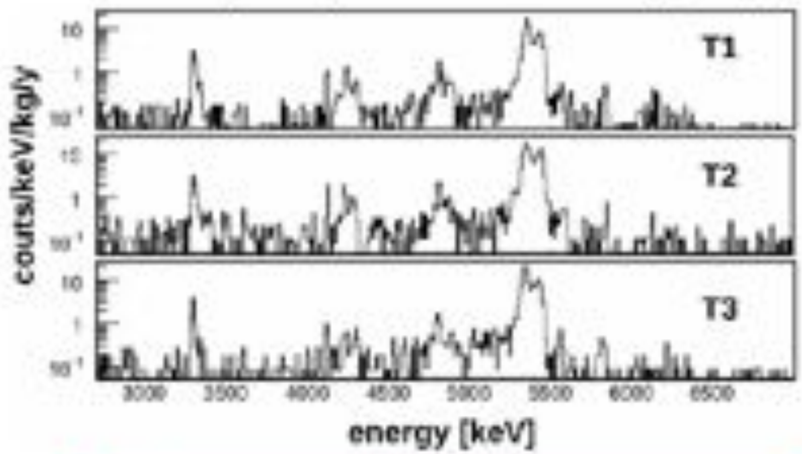


T1: Polyethylene
- Soap
- H_2O_2+ H_2O + citric acid
- 70 mm of polyethylene

T2: Chemical treatment
- Soap
- Electro-erosion
- Chemical etching
- H_2O_2+ H_2O + citric acid

T3: Plasma cleaning
- Tumbling
- Electro-polishing
- Chemical etching
- Magnetron Plasma etching

Total spectra (no anticoincidence applied)



Best result obtained for T1 and T3

Bkg in the (2.7÷3.9) MeV region after anti-coincidence cut:

=> 0.052 ± 0.008 c/keV/kg/y

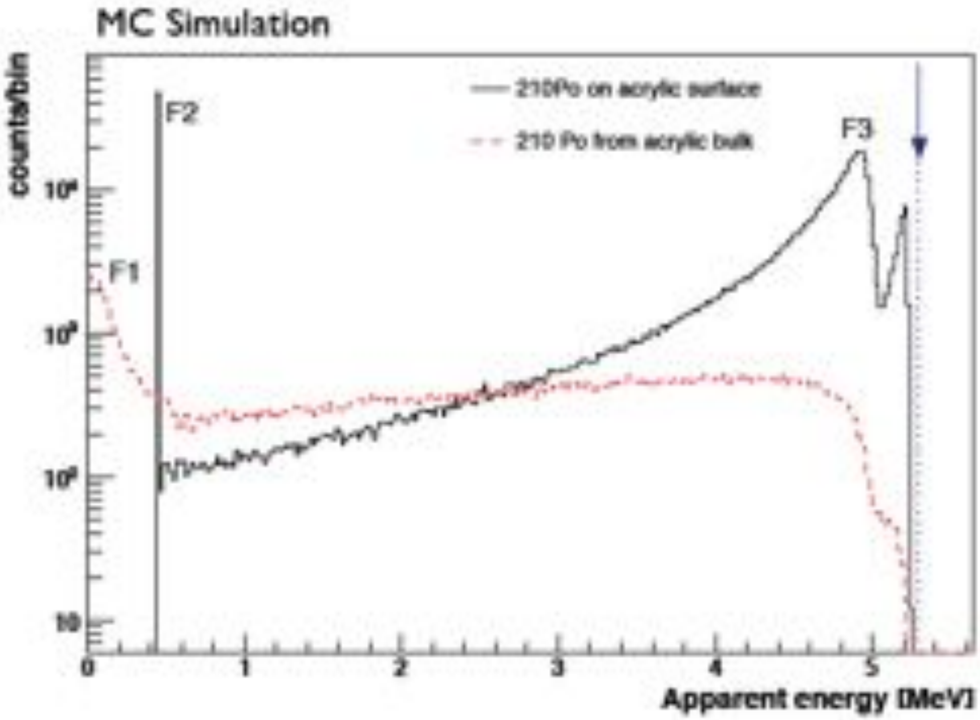
^{232}Th / ^{238}U on Cu: < $7 \cdot 10^{-8}$ Bq/cm^2

Slide provided by A. Giuliani (CUORE)

Removal of ^{222}Rn daughters from Cu, SS and Ge Surfaces

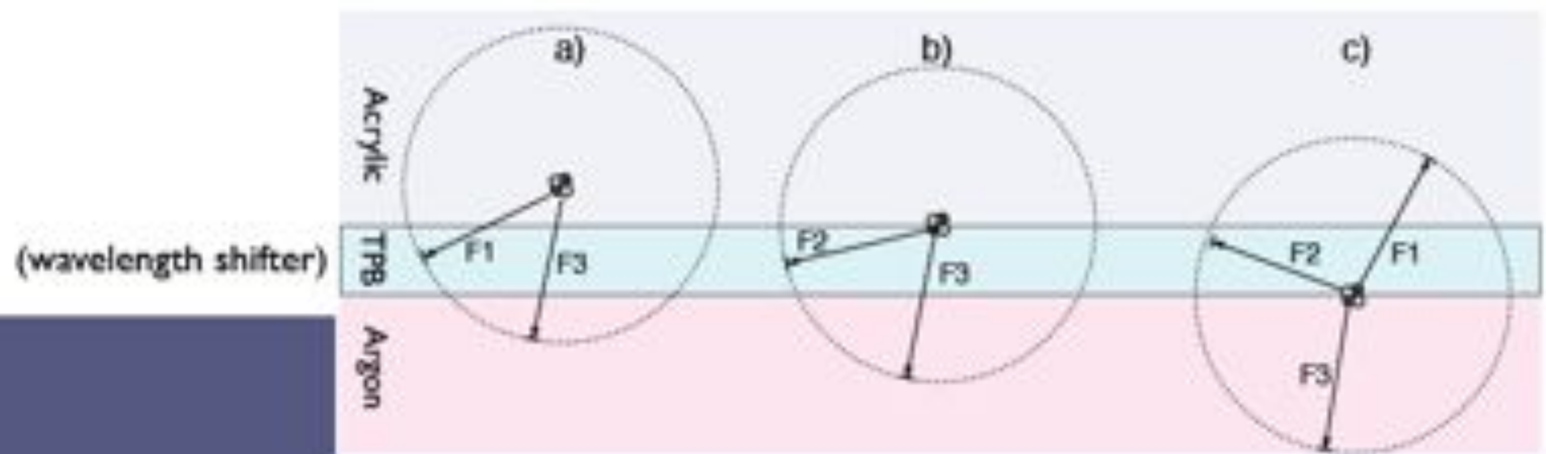
- Zuzel and Wójcik: NIM A 676 (2012) 140 and Zuzel et al.: NIM A 676 (2012) 149.
- Different recipes and procedures tested.
- Some general observations:
  - Electropolishing is more effective than chemical etching.
  - ^{210}Po removal is most difficult: For instance no reduction of ^{210}Po from Cu surfaces by etching.
  - Explanantion: Re-deposition of polonium during the etching process.
- Would be nice to continue the study with more (non-metallic) materials and different recipes.

DEAP-3600 has studied the backgrounds from alpha particles emitted by radon daughters in or on the acrylic and the TPB layer.



The spectra of those decays can reach down to energies relevant for WIMP search.

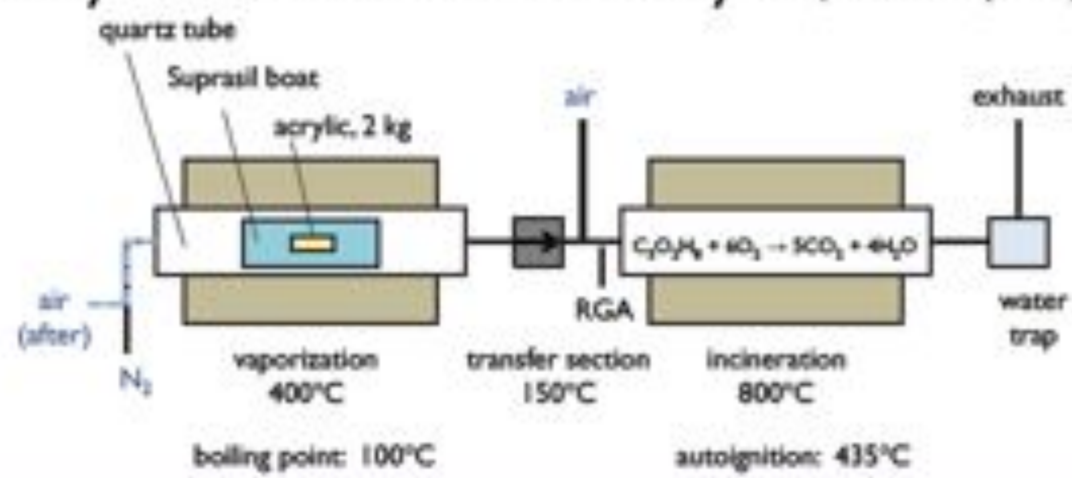
Slide provided by T. Pollmann (DEAP)



DEAP 3600

DEAP-3600 controls and verifies contamination with radon daughters through:

Assay of the manufactured acrylic. (C. Nantais's poster)



Radon-reduced Working Environment

- If long-lived ^{222}Rn daughters are a problem for your experiment, you would like to work in ^{222}Rn-free environment.
- Best: N_2 atmosphere, but people can't work there
- Alternative 1: Synthetic air!
  - Mix O_2 from gas cylinders with boil-off N_2
  - <0.1 mBq/m^3 ^{222}Rn (Borexino)
  - Expensive: Not for permanent usage
- Alternative 2: Rn-reduced air
  - Strip radon from (dried) air with cryogenic activated carbon column
  - typically 1 mBq/m^3
  - Applied by Super-K, NEMO, CUORE, Darkside

Radon Reduction System for Ambient Air

The DARKSIDE plant

Slide provided by C. Galbiati (Darkside)

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Darkside Cleanroom

Obtained <30 mBq/m^3 in >100 m^3 cleanroom volume (~factor 1000 reduction)

Slide provided by C. Galbiati (Darkside)

Radon Removal from Nitrogen and Argon to Concentrations below 1 microBq/m^3

- Cryo-adsorption is powerful and simple technique to purify gases.
- Based on different binding energy of bulk gas and contaminant
- If difference is strong, bulk gas adsorption and contaminant adsorption are independent → no interference
- Activated carbon is ideal adsorber due to
  - huge surface area (~1000 m^2/g)
  - non-polar surface
  - high chemical purity and low ^{222}Rn emanation rate

Radon Removal from Nitrogen and Argon to Concentrations below 1 microBq/m^3

- Liquid nitrogen purification plant for Borexino
- <0.5 μBq/m^3 at 100 m^3/h production rate
- Appl. Rad. Isot. 52 (2000) 691.

- In Gerda experiment: <1μBq/m^3 at 18 m^3/h achieved for gaseous argon.
- Appl. Rad. Isot. 67 (2009) 922.

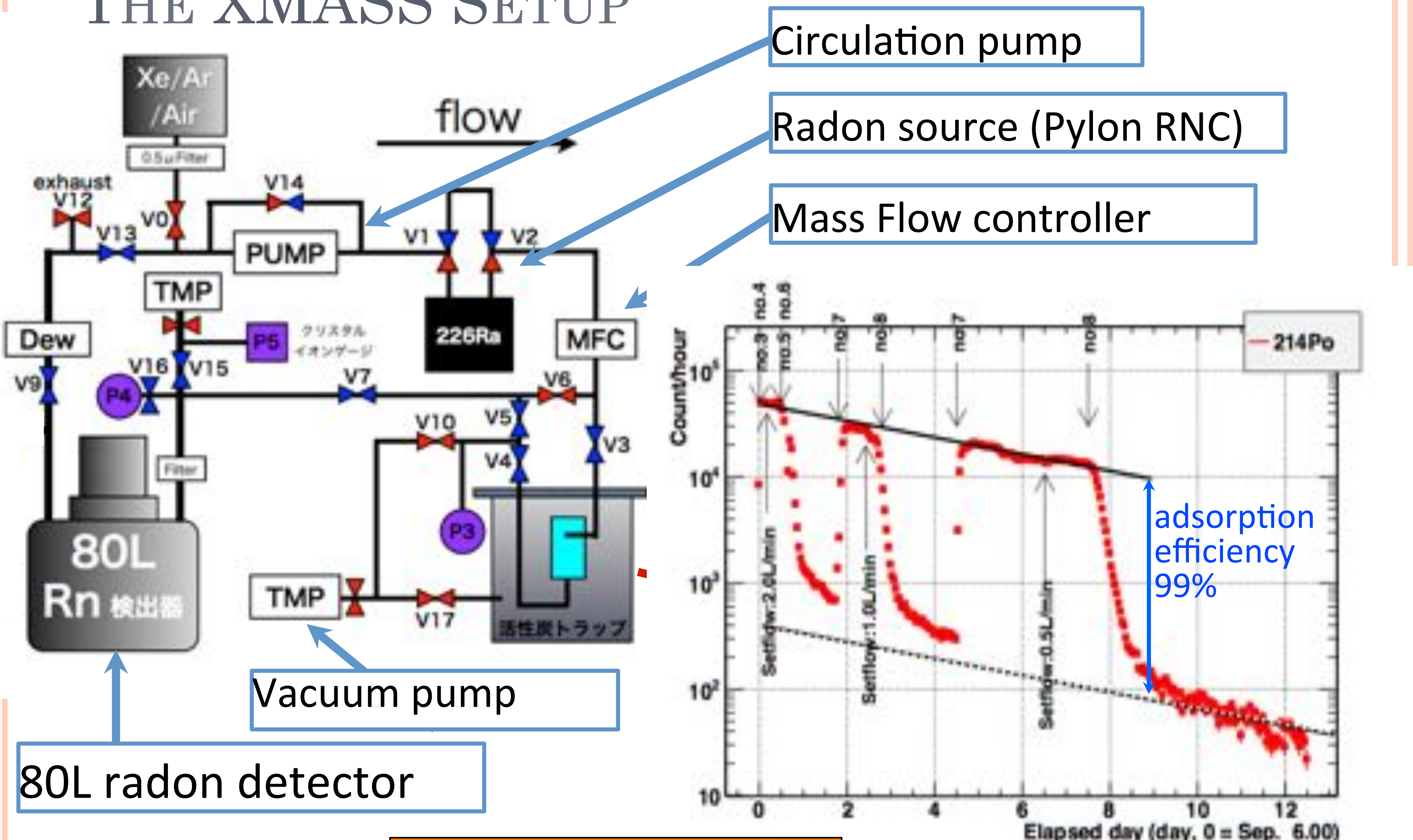
Radon Removal from Xenon

- Xenon nowadays an attractive target material
- What works well for nitrogen/argon (adsorption) should also work for xenon, but:
  - Xenon and radon are very similar → hard to separate.
  - Relatively high temperature of LXe compared to N_2/Ar.
  - Xenon is very expensive
    - Tests are more difficult.
    - Xenon adsorption is critical (adsorbed fraction is unusable).

Important to select best adsorber material

Radon Removal from XENON: The XMASS Setup

Slide provided by Y. Takeuchi (XMASS)

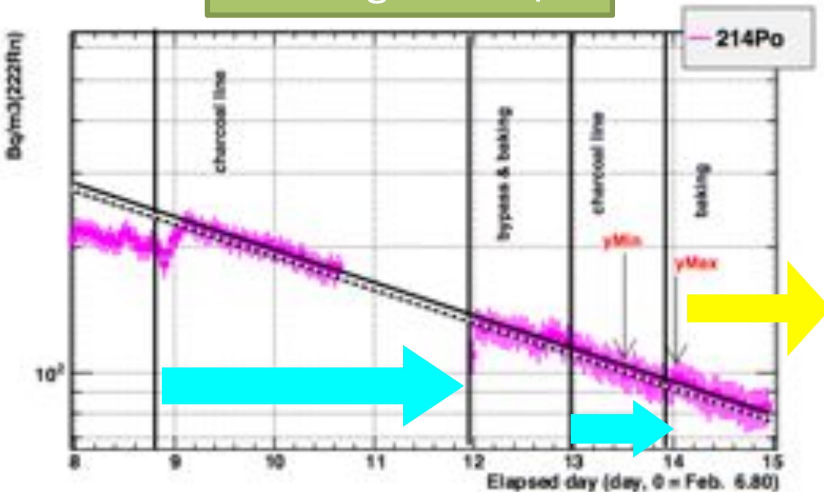


see NIM A 661 (2012) 50

SOME VERY PRELMINARY DATA

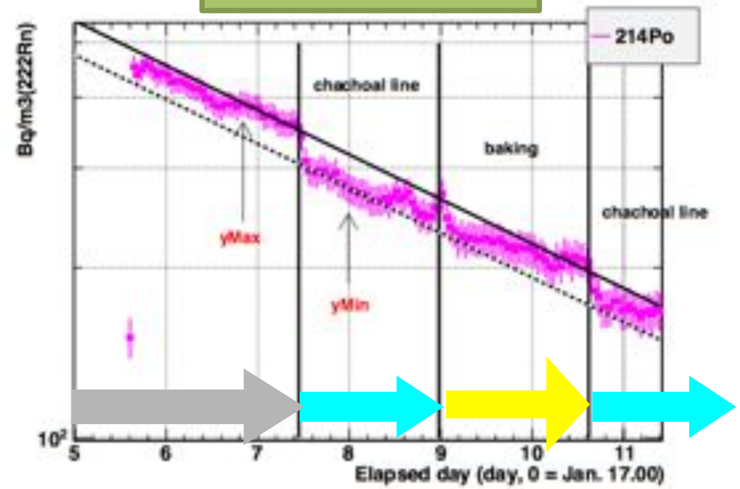
Slide provided by
Y. Takeuchi (XMASS)

Shirasagi X2M 4/6



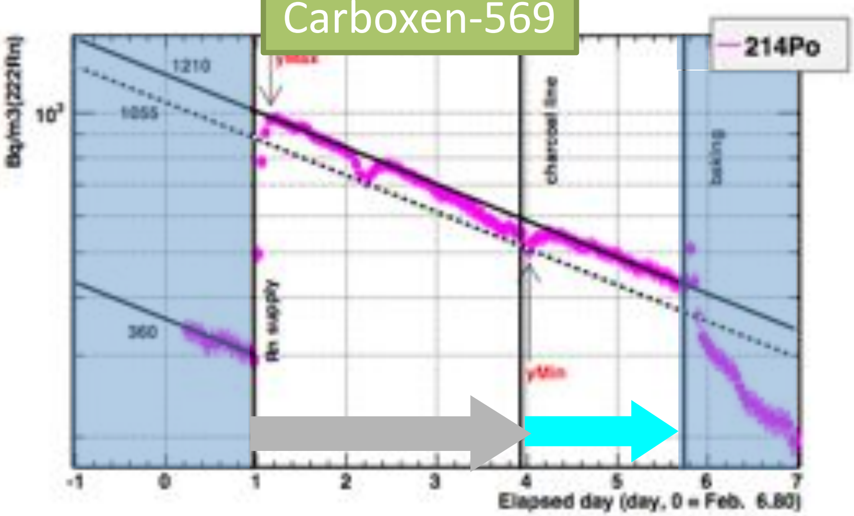
adsorption efficiency: 0-4%

Carboxen-1021



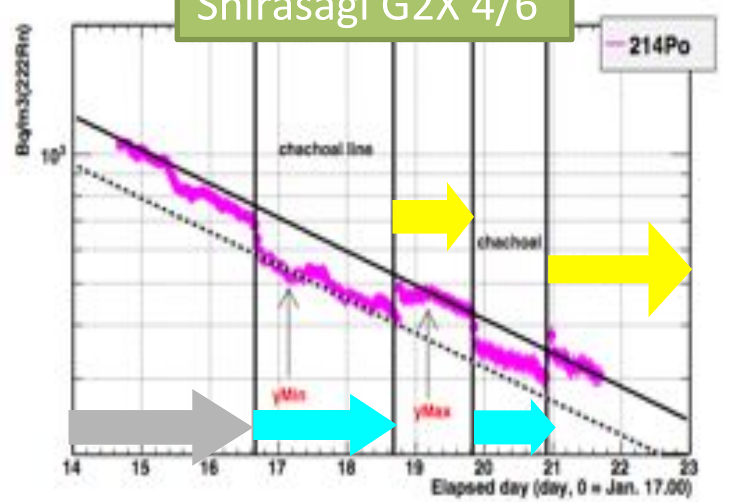
adsorption efficiency: 6-13%

Carboxen-569



adsorption efficiency: 0-13%

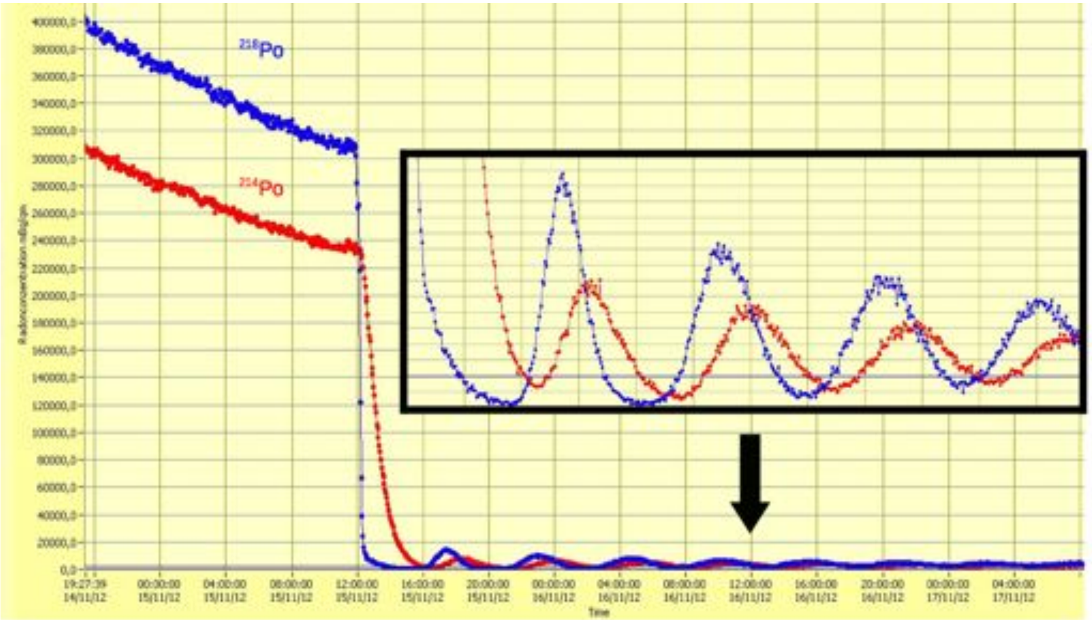
Shirasagi G2X 4/6



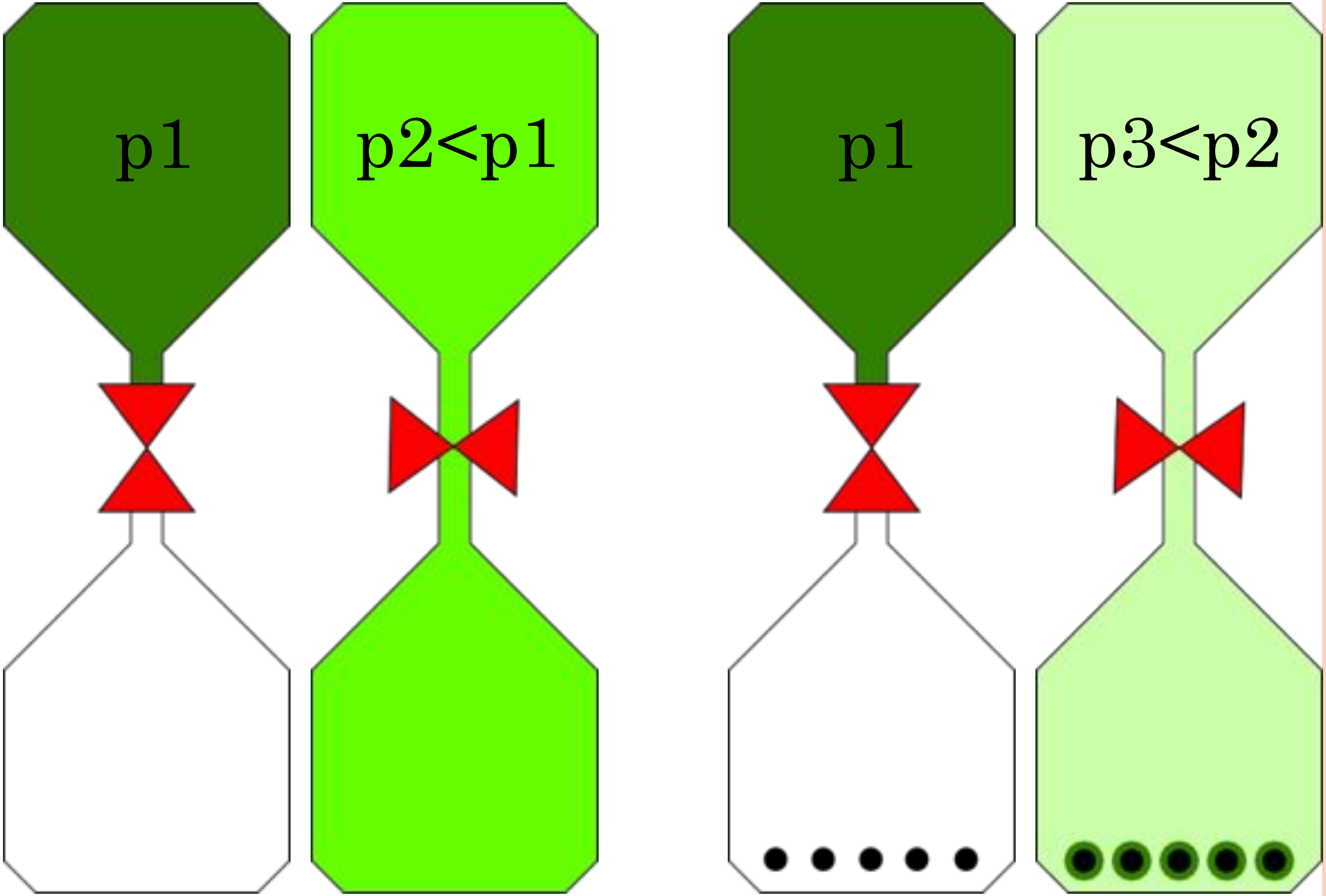
adsorption efficiency: 7-23%

Radon Removal from Xenon: Work in EXO and XENON1T

- EXO:
  - Use Cu (Ni) spheres instead of carbon.
  - EXO-200 does no longer need a Rn trap.
  - Work will be resumed for nEXO.
- XENON1T:
  - Similar setup as XMASS.
  - Use plateau and oscillations to extract adsorption parameters.



A different Approach: Adsorption Equilibrium Measurements



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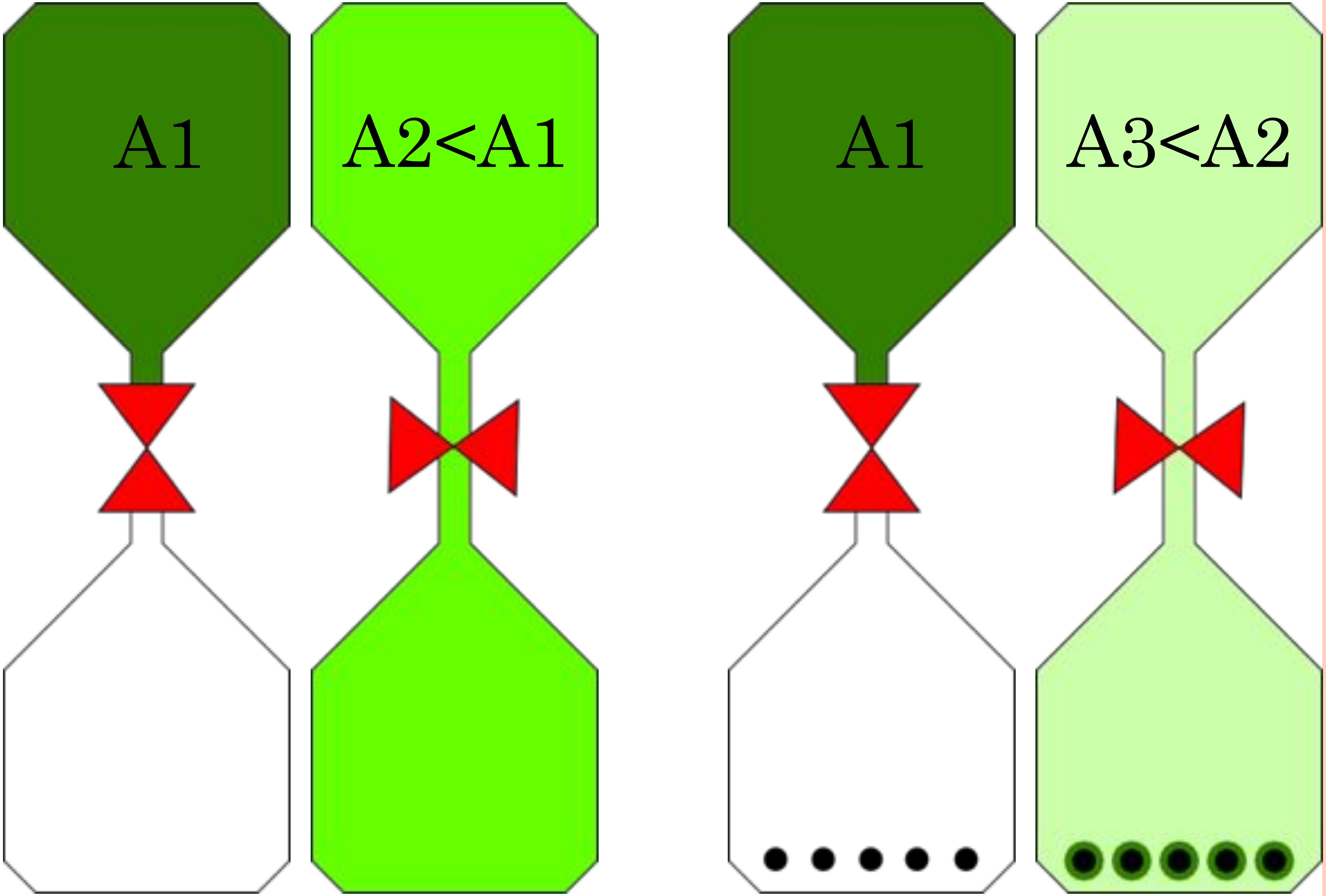
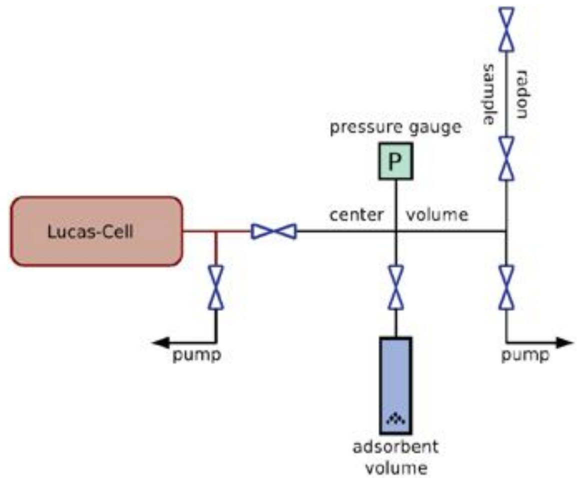
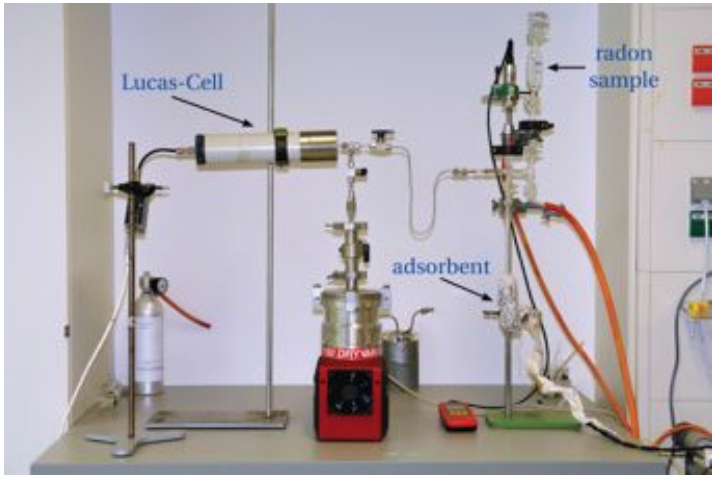


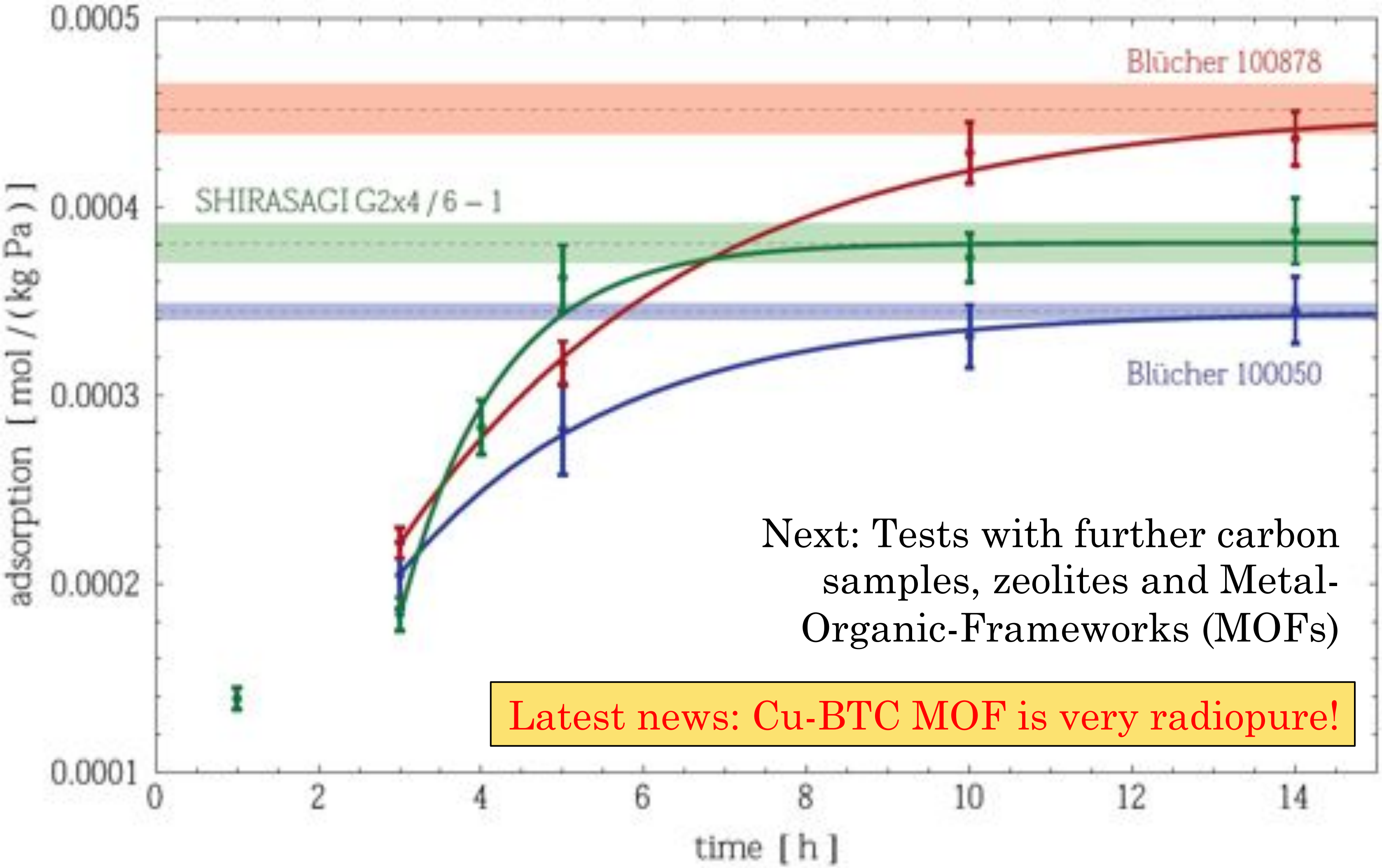
MPIK Device for Adsorption Equilibrium Measurements





- Simultaneous measurement of xenon adsorption (pressure) and radon adsorption (activity).
- Elaborated procedure to avoid disturbance by Rn daughters.
- Robust, reproducible results: see S. Bruenner, Diploma Thesis (2013) University of Graz / MPIK.

^{222}Rn Adsorption from a Rn/Xe Mixture on three Activated Carbons



Next: Tests with further carbon samples, zeolites and Metal-Organic-Frameworks (MOFs)

Latest news: Cu-BTC MOF is very radiopure!

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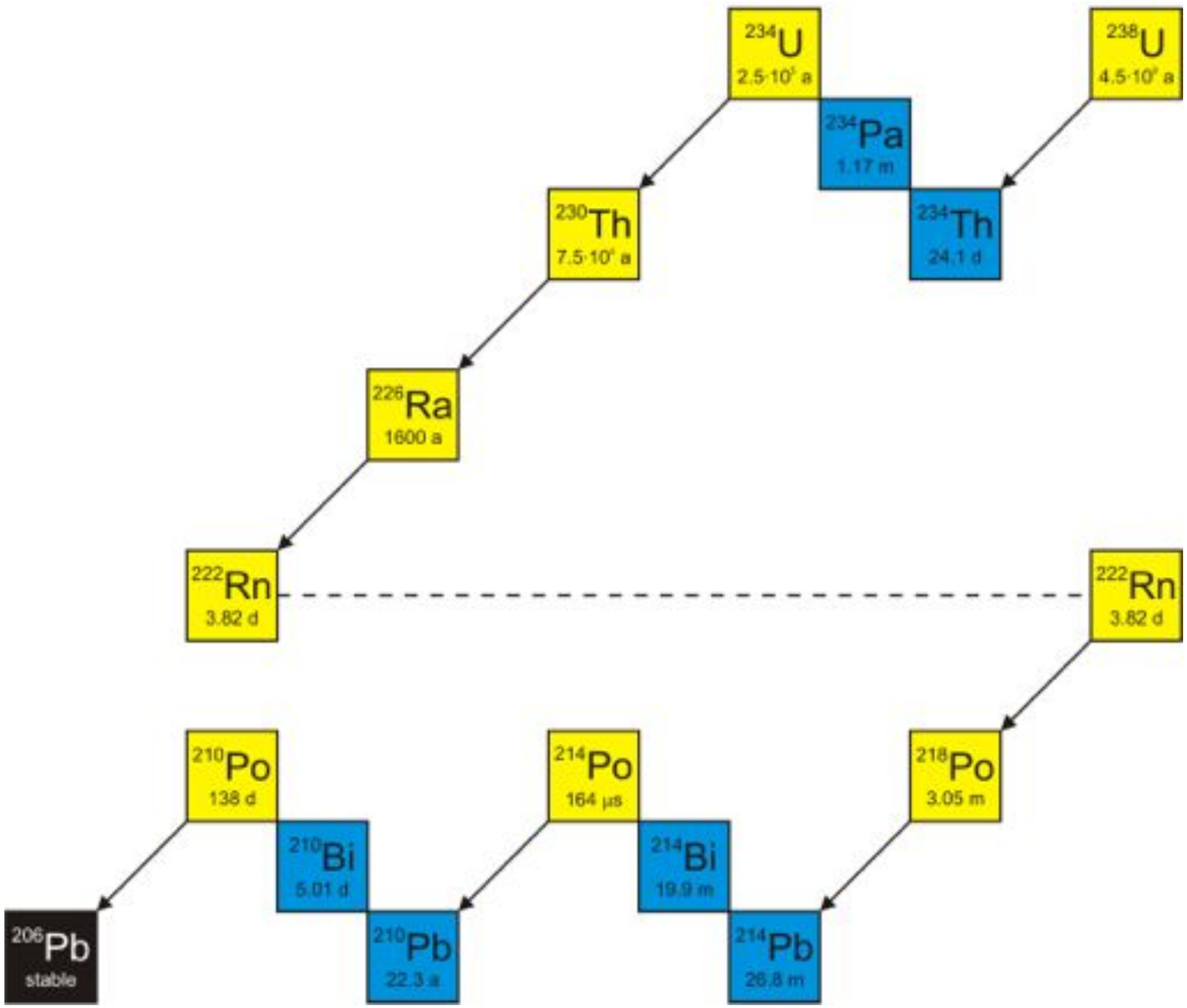
THANK YOU!

- J. Farine (EXO)
- C. Galbiati (Darkside)
- A. Giuliani (CUORE)
- H. Gomez (SuperNEMO)
- T. Pollmann (DEAP)
- Y. Takeuchi (XMASS)
- J. Wolf (KATRIN)
- G. Zuzel (GERDA)

for slides and very helpful discussions.

^{222}Rn decay chain



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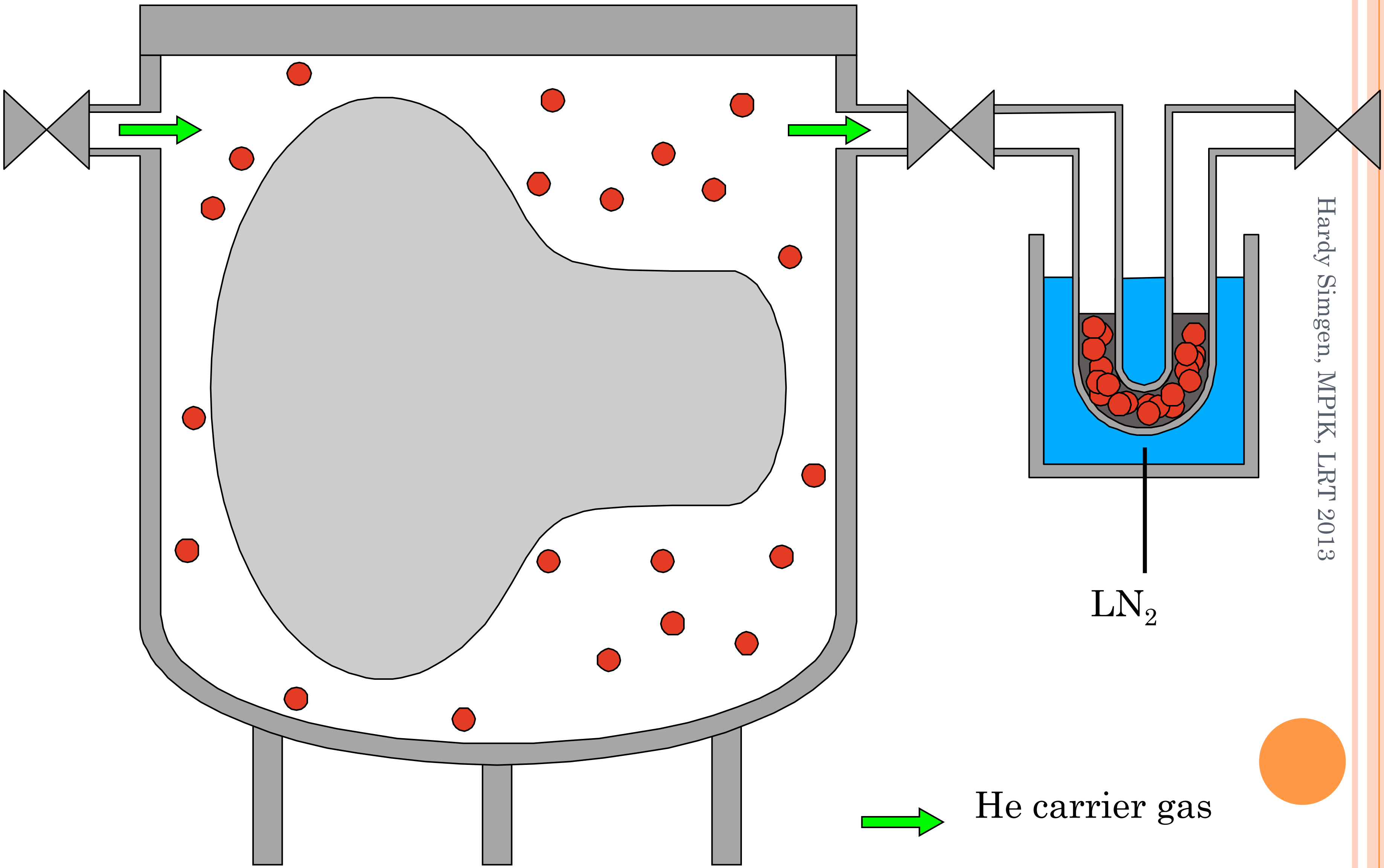
Material Selection: The ^{222}Rn Emanation Technique



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