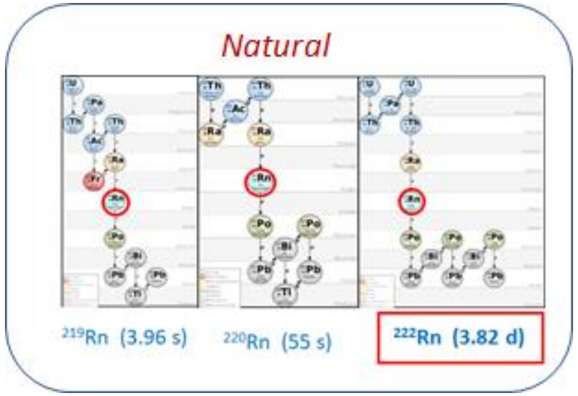
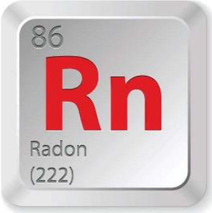
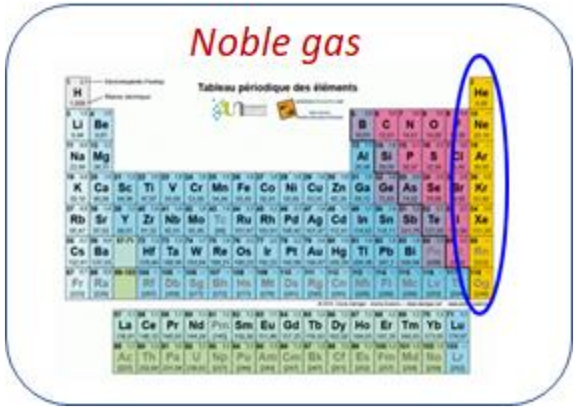
The IRENE Project
(Innovative mateRials for Extreme radon capture)

GDR Duphy, WP2, Lyon
Hichem Tedjditi

11/06/2025



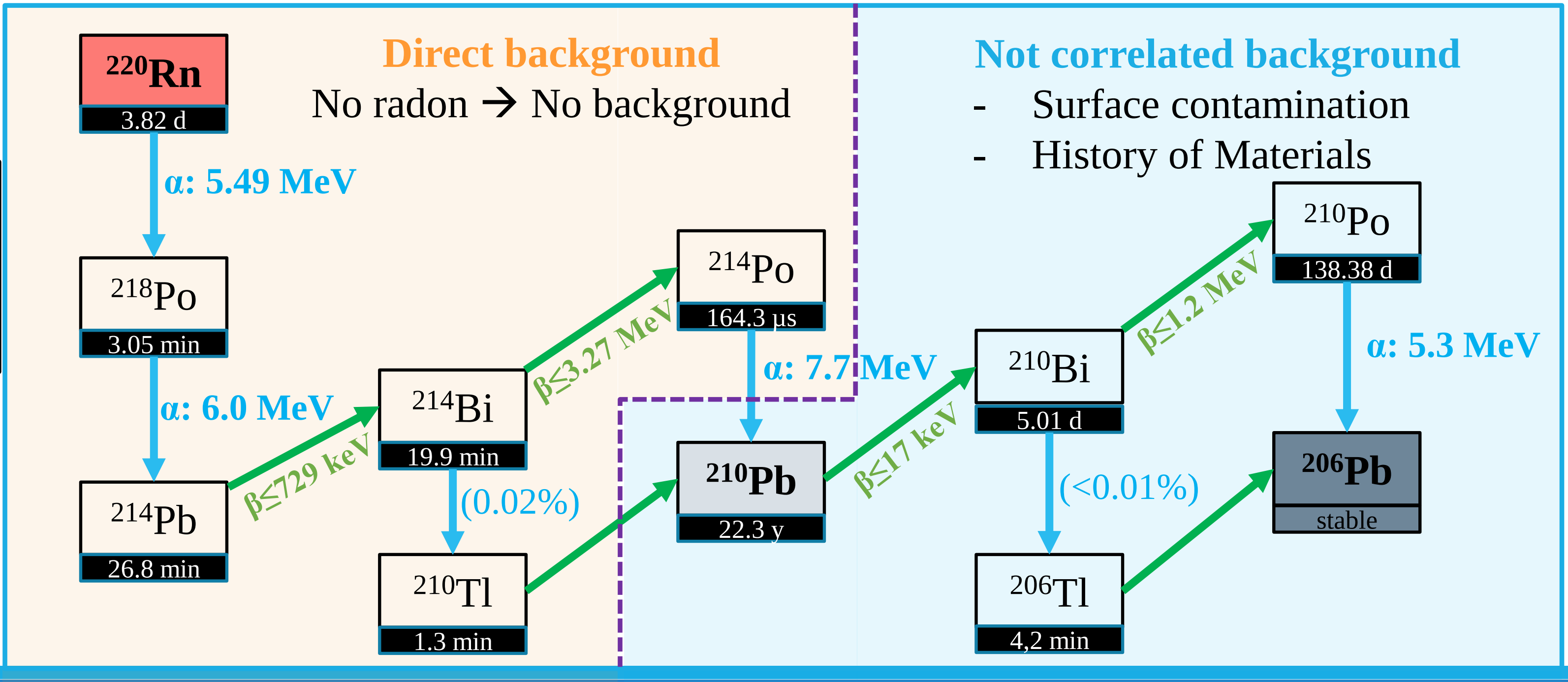


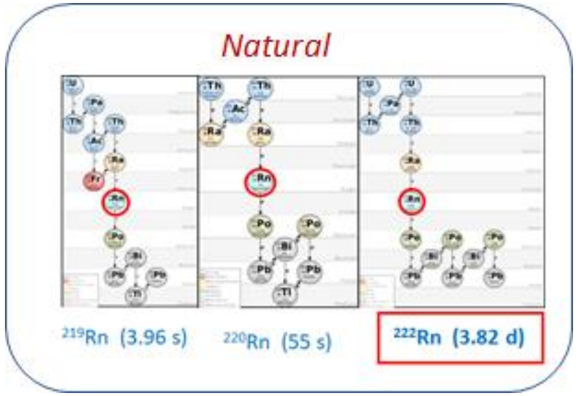


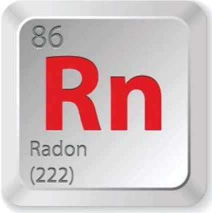
Natural, Radioactive, Noble gas from the U and Th chains

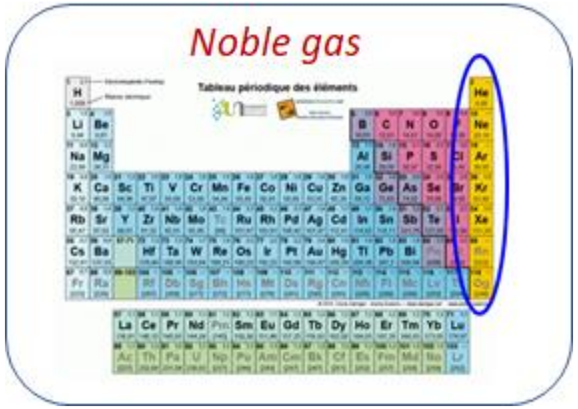
- Easy to transport, complicated to capture
- Short half-life, high intrinsic activity

→ α from 5 to 8 MeV
→ β ≤ 3.27 MeV
→ γ ≤ 3.18 MeV









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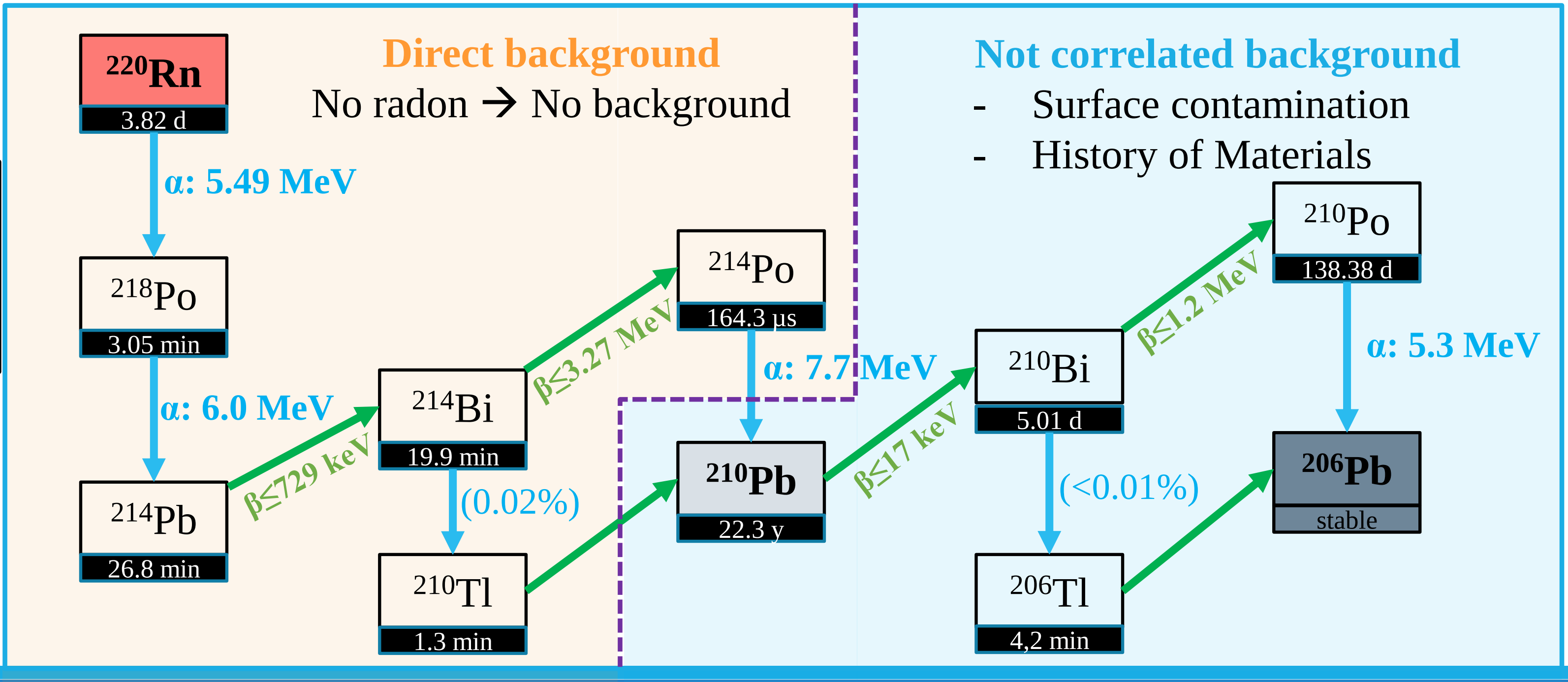

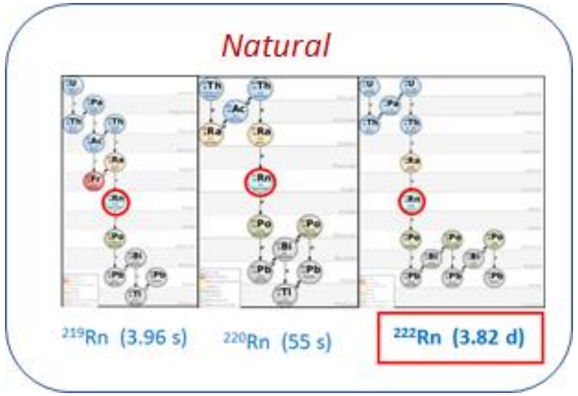

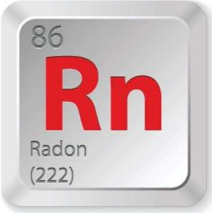

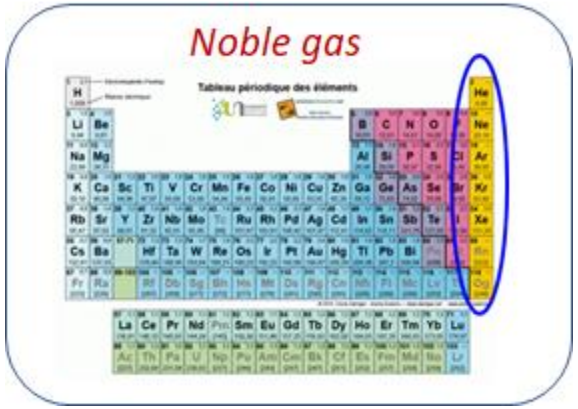

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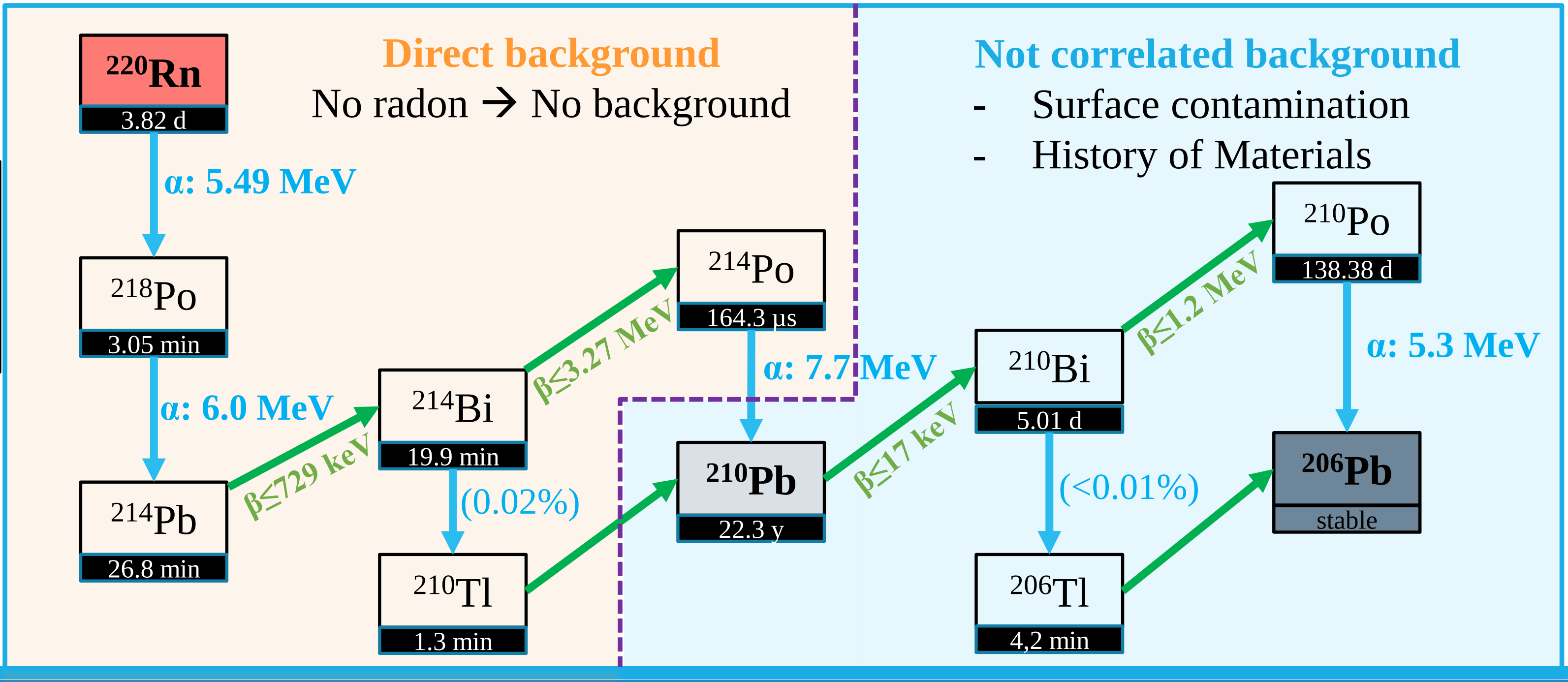

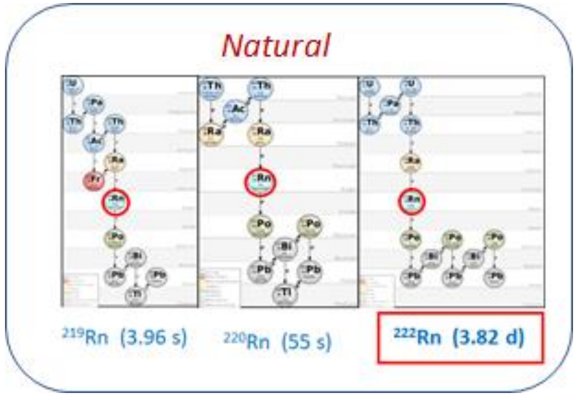

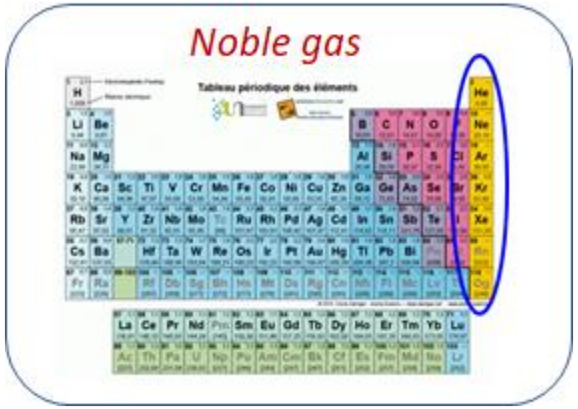

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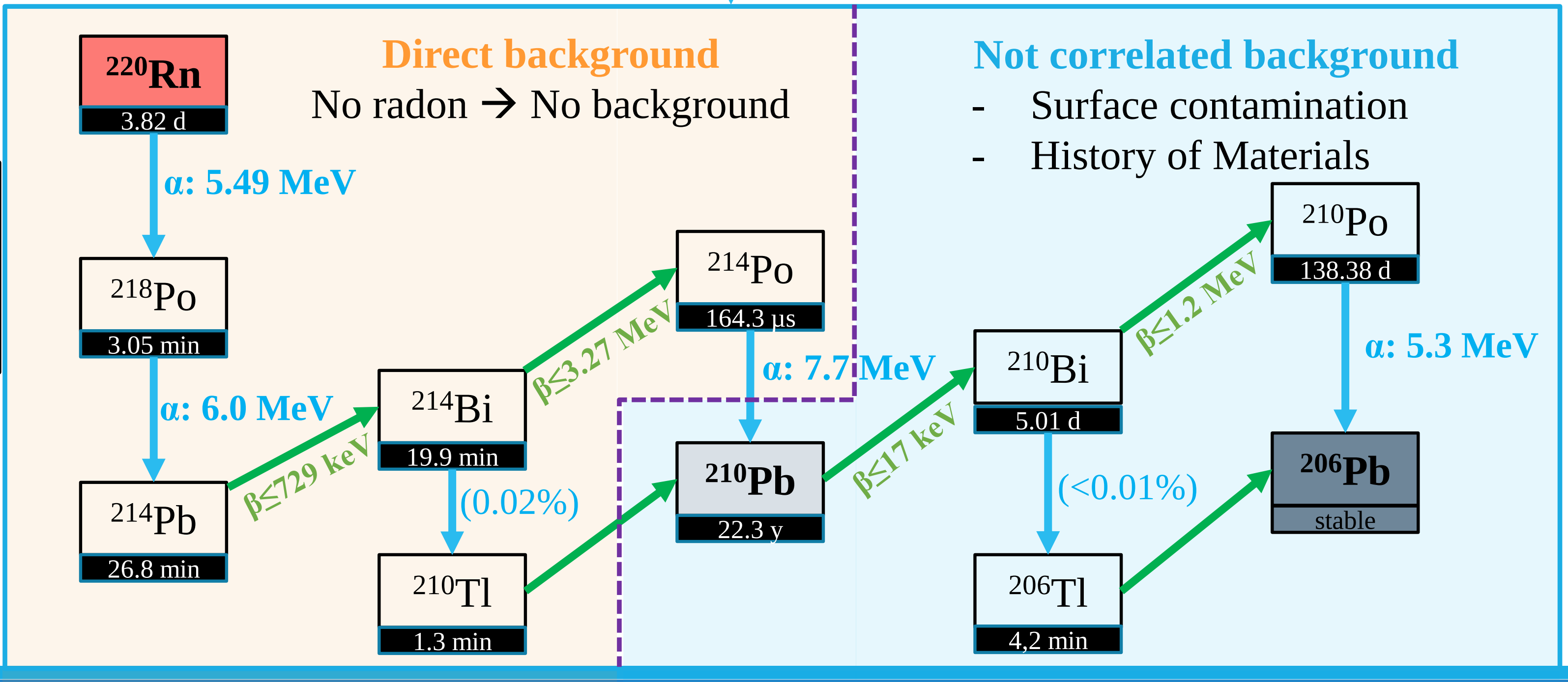

→ **α from 5 to 8 MeV**
→ **β ≤ 3.27 MeV**
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Radon Capture

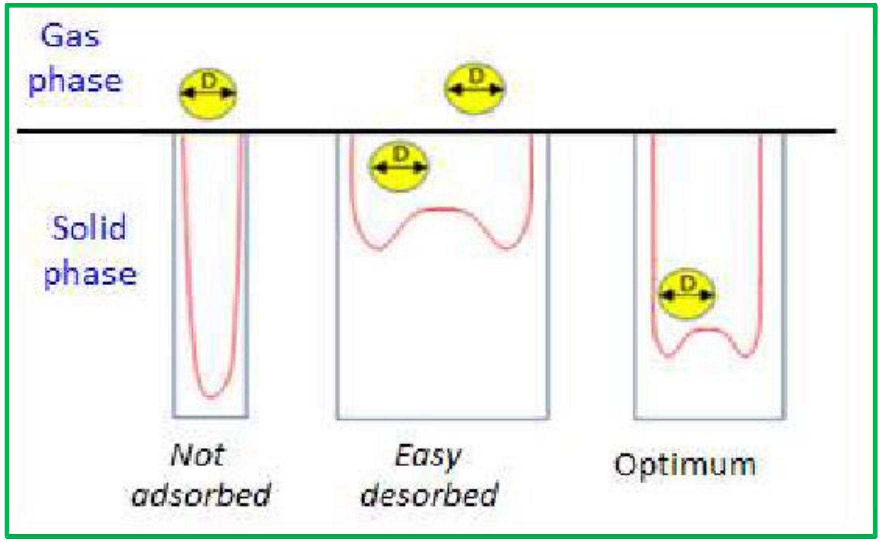
Rn is a Noble gas:

- No chemistry → Physical process

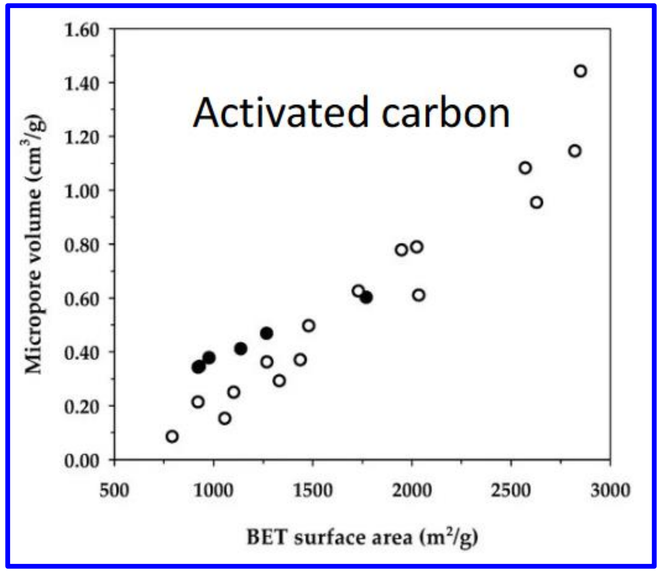
Physical adsorption:

→**Surface** capture by Van der Waals forces

→Use of porous materials: very high surface materials, up to 4000m²/g



Need to optimized Porosity



IRENE history

→ 2015 : Study of Rn capture in 50 commercial or research microporous materials (SuperNEMO)

- Activated charcoals
- Carbon molecular sieves
- Carbon aerogels (Granada Univ.)
- Molecular cages - CC3 (Liverpool Univ.)
- MOF
- Zeolites (Al, Si) (Marseille, Mulhouse Univ.)
- Dedritic polymers (Marseille)

Aix*Marseille université

CPPM

CiNaM

supernemo collaboration

UNIVERSITÉ D'AIX-MARSEILLE

ÉCOLE DOCTORALE PHYSIQUE ET SCIENCES DE LA MATIÈRE (ED352)

CPPM/CINaM

Thèse présentée pour obtenir le grade universitaire de docteur

Mention : Matière Condensée et Nanosciences

Étude et développement de substrats microporeux pour l'adsorption du radon et son application en physique du neutrino

NOËL Raymond

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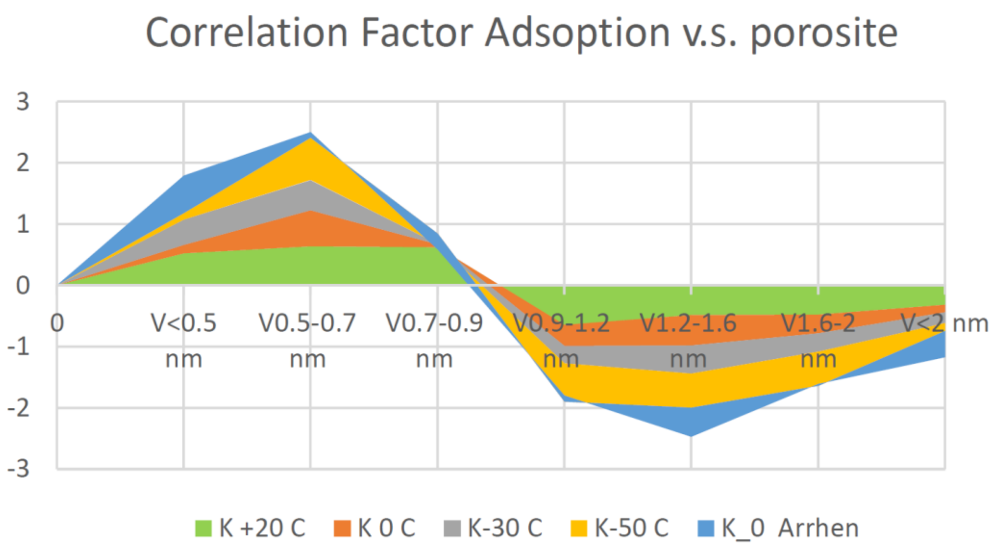
Porosity from 0.5 to 2 nm

Échantillons	S_{BET} N_2 (m^2/g)	$V_{<0,5\,nm}$ (cm^3/g)	$V_{0,5\text{-}0,7\,nm}$ (cm^3/g)	$V_{0,7\text{-}0,9\,nm}$ (cm^3/g)	$V_{0,9\text{-}1,2\,nm}$ (cm^3/g)	$V_{1,2\text{-}1,6\,nm}$ (cm^3/g)	$V_{1,6\text{-}2\,nm}$ (cm^3/g)	$V_{<0,7\,nm}$ (cm^3/g)	$V_{0,7\text{-}2\,nm}$ (cm^3/g)	$V_{<2\,nm}$ (cm^3/g)
Carbosieve SIII (1)	1062	0,076	0,233	0,062	0,018	0,020	0,000	0,308	0,102	0,410
Carboxen 1000 (2)	812	0,064	0,161	0,044	0,001	0,000	0,000	0,225	0,045	0,270
Carboxen 569 (3)	299	0,074	0,011	0,000	0,000	0,000	0,000	0,085	0,000	0,085
K48S (4)	799	0,074	0,138	0,034	0,033	0,032	0,006	0,212	0,105	0,317
Nucleacarb 208C 5TEDA (5)	1298	0,053	0,136	0,084	0,087	0,106	0,044	0,189	0,321	0,510
SHIRASAGI G2x4 (6)	1383	0,051	0,161	0,096	0,090	0,103	0,033	0,212	0,322	0,534
Carboact (7)	1096	0,077	0,206	0,053	0,033	0,048	0,022	0,283	0,156	0,439
Environcarb 207C (8)	1073	0,075	0,181	0,058	0,046	0,050	0,011	0,256	0,165	0,421
K48 (9)	793	0,078	0,137	0,026	0,034	0,034	0,005	0,215	0,099	0,314
Nuclearcarb 208 5KI3 (10)	1315	0,057	0,145	0,081	0,084	0,110	0,046	0,202	0,321	0,522
Carboxen 1012 (11)	1140	0,074	0,145	0,054	0,076	0,078	0,016	0,219	0,223	0,443
Carboxen 1021 (12)	585	0,083	0,049	0,020	0,037	0,026	0,017	0,132	0,100	0,231
Carboxen 1016 (13)	77	0,000	0,000	0,000	0,000	0,000	0,000	0,000	0,000	0,000
Carboxen 1018 (14)	804	0,061	0,155	0,045	0,010	0,024	0,014	0,216	0,092	0,308
Aquacarb 207C (15)	1040	0,063	0,165	0,066	0,044	0,052	0,019	0,228	0,181	0,409
Carbosieve G (16)	1273	0,074	0,183	0,112	0,046	0,050	0,025	0,257	0,233	0,490



Optimal porosity:

- 0.5 – 0.7 nm (Rn 0.4 nm)

Small effect on the chemical composition (Sulphur)

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→ 2018 : Xenon / Radon adsorption with macromolecular cages: Cryptophane

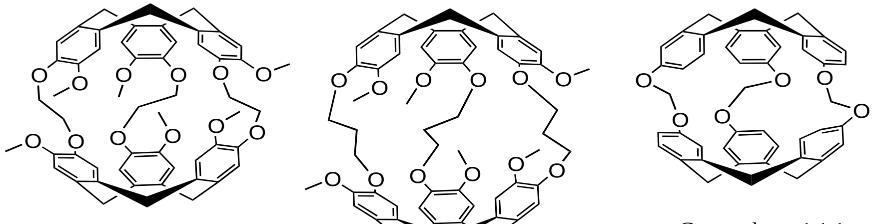
- Used for the complexing of Xe
- Adjustable size

Atomic Radius

Gas	Van der Waals (nm)	Covalent
Xe	0.216	0.130
Rn	0.220	0.145

Compétition Xe / Rn

Three types of cage

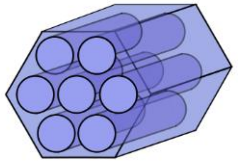


crptophane A
ou cryptophane 2,2,2

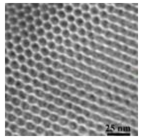
crptophane E

Cryptophane 1,1,1

Two supports



Silice MCM-41
Ø = 2,6 nm
1000 m²/g



Silice SBA-15
Ø = 6,5 nm
500 m²/g

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Rn Adsorption (CPPM)

Adsorbant	K (m^3/kg) @-30°C
YC-125: Cryptophane A/MCM-41	103,2 ± 25
YC-126: Cryptophane A/SBA-15	60 ± 15
YC-127: Cryptophane E/MCM-41	24 ± 6
YC-128: Cryptophane E/SBA-15	2 ± 6
Crystals of Cryptophane	0
AC K48 special	66,6 ± 5

Xe Adsorption (MADIREL)

Echantillon	Adsorption Xe (mmol/g)
Cryptophane A (MCM-41)	0.176 ±0.010
Cryptophane A (SBA-15)	0.123 ±0.006
Charbon actif Silcarbon K48	2.31 ±0.04

13.12

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→ 2020 : Radon adsorption on silver zeolite

Three commercial Ag Zeolite

Zeolite	Supplier	Chemical formula	Silver content (wt%)	Pore size (Å)
Ag-ZSM-5	Riogen	$Ag_{4.8}[(AlO_2)_{4.8}(SiO_2)_{91.2}] \cdot 16H_2O$	7.9	5.2 × 5.7 and 5.3 × 5.6
Ag-13X	Sigma Aldrich	$Ag_{84}Na_2[(AlO_2)_{86}(SiO_2)_{106}] \cdot xH_2O$	35	10 × 10
Ag-ETS-10	Extraordinary Adsorbents Inc.	$Ag_{16}[Si_{40}Ti_8O_{104}]$	30	4.9 × 7.6

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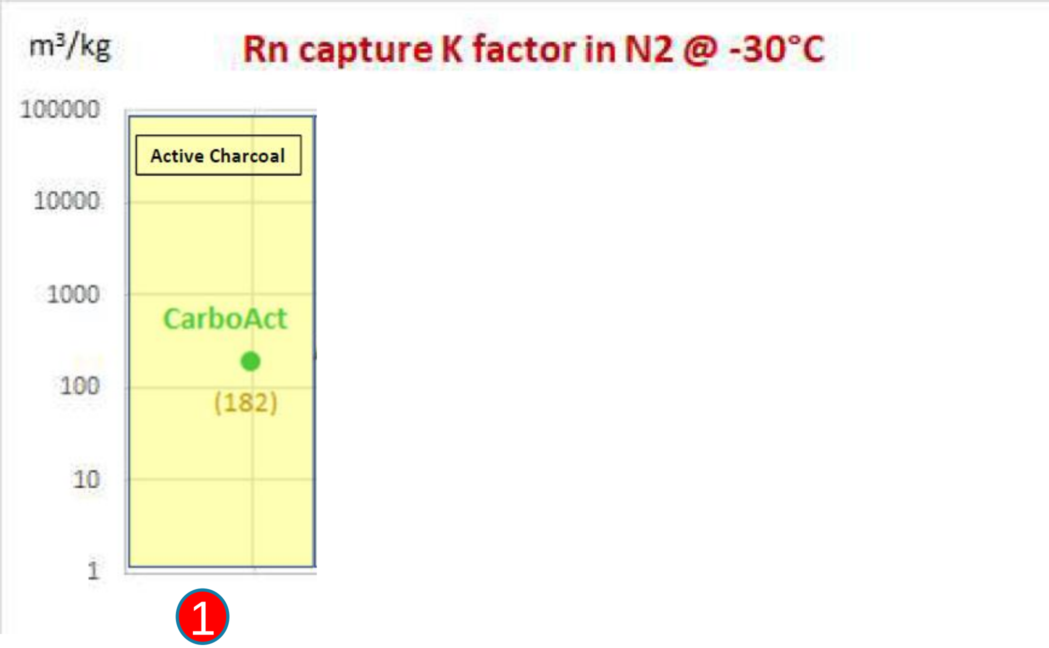
$K = \frac{Rn\ in\ the\ Adsorbent}{Rn\ in\ the\ Gas}$ [m³/kg]



1 Reference charcoal (carboAct)

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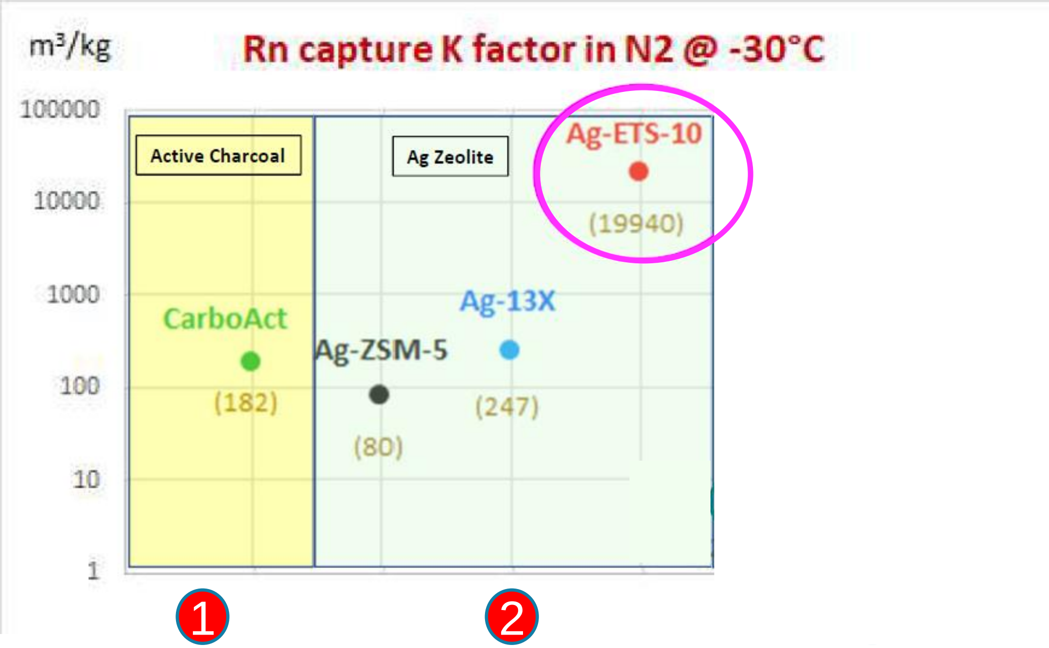

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- Ag-13X
- Ag-ETS-10

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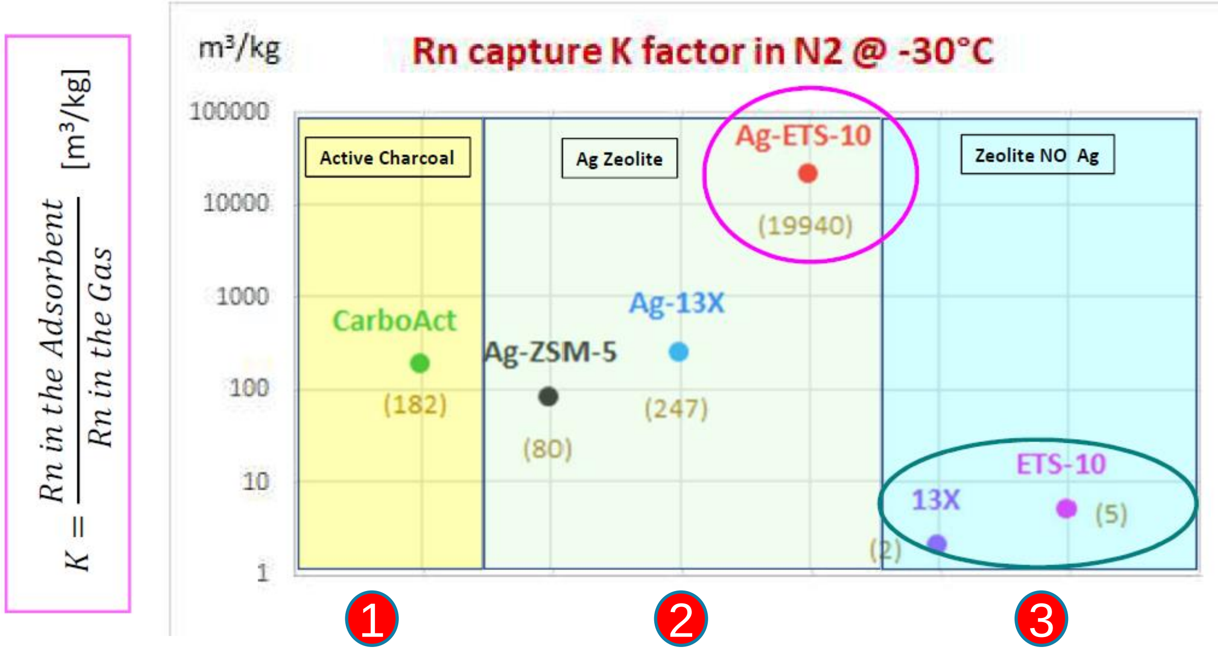

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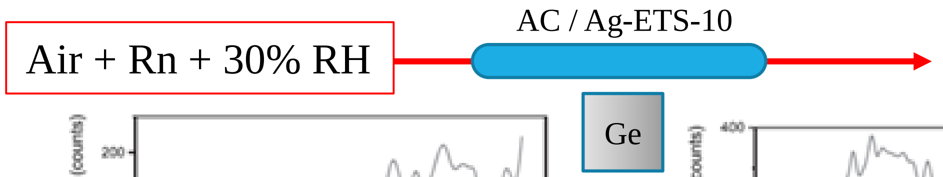
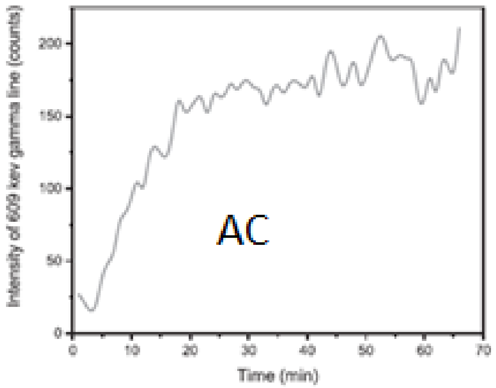
High ^{226}Ra concentration

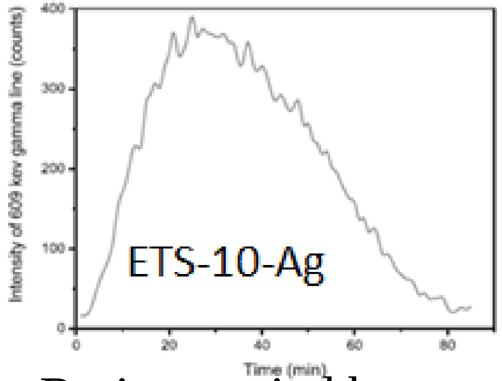
Echantillon	226Ra (Bq/kg)
K48	< 0,25
NuclCarb 5TEDA	<0,3
ENvCarb 207C	0,28 +- 0,17
Shirasagi	0,164 +- 0,023
CarboAct	0,0023 +- 0,0019
ETS-10-AG	0,988 +- 0,05

High sensitivity to H_2O





Rn reach equilibrium



Rn is emptied by water

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Other results

@ room temperature

Sample	Reference	K factor [m3/kg]
Ag-ETS-10	Heinitz et al.	3400
Ag-ZSM-5	Heinitz et al.	3500
8 Ag-FER-B	Takeuchi et al.	6500

AC @ 20°C : ~ 10 m^3/kg

Very good results but not well understood

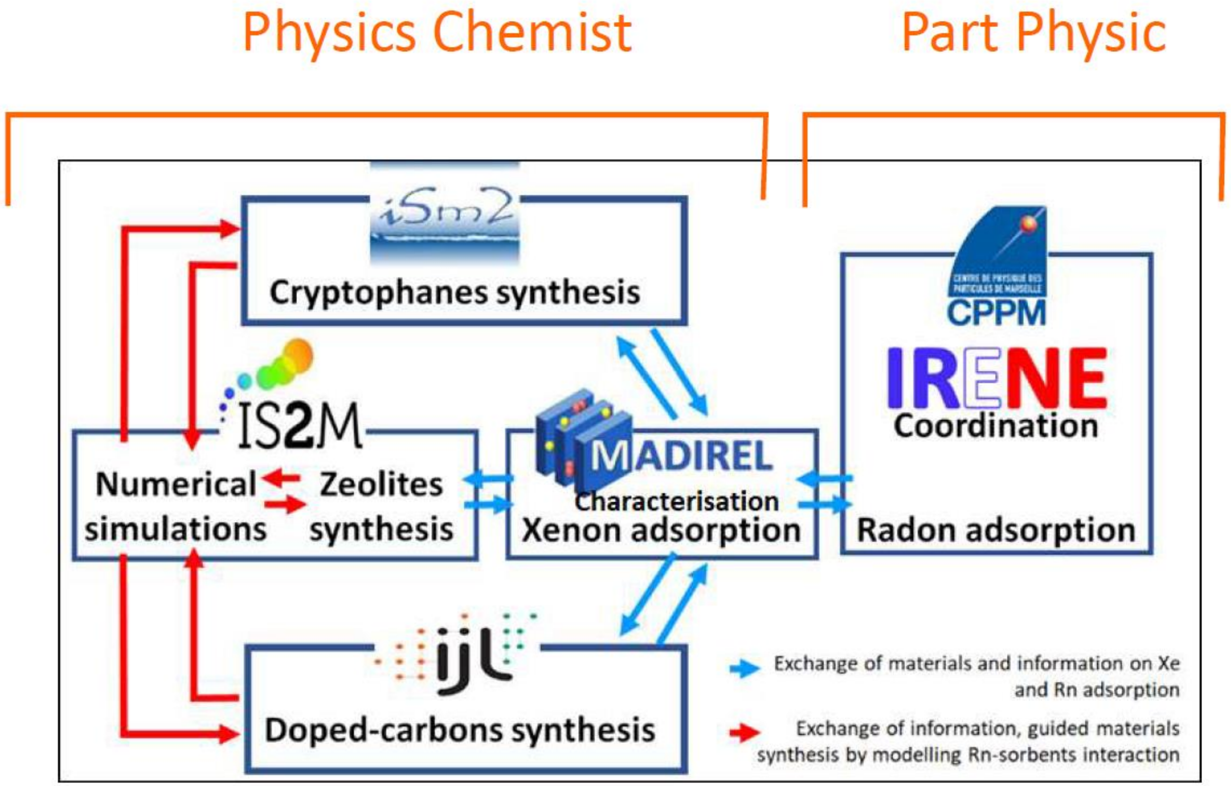
Can we do better ?

- Higher adsorption
- Improved radiopurity
- Lower sensitivity to water

IRENE Project

Innovative mateRials for Extreme radoN capturE

Understanding and optimising the capture of Rn in new materials



Project start in February 2024

Philosophy:

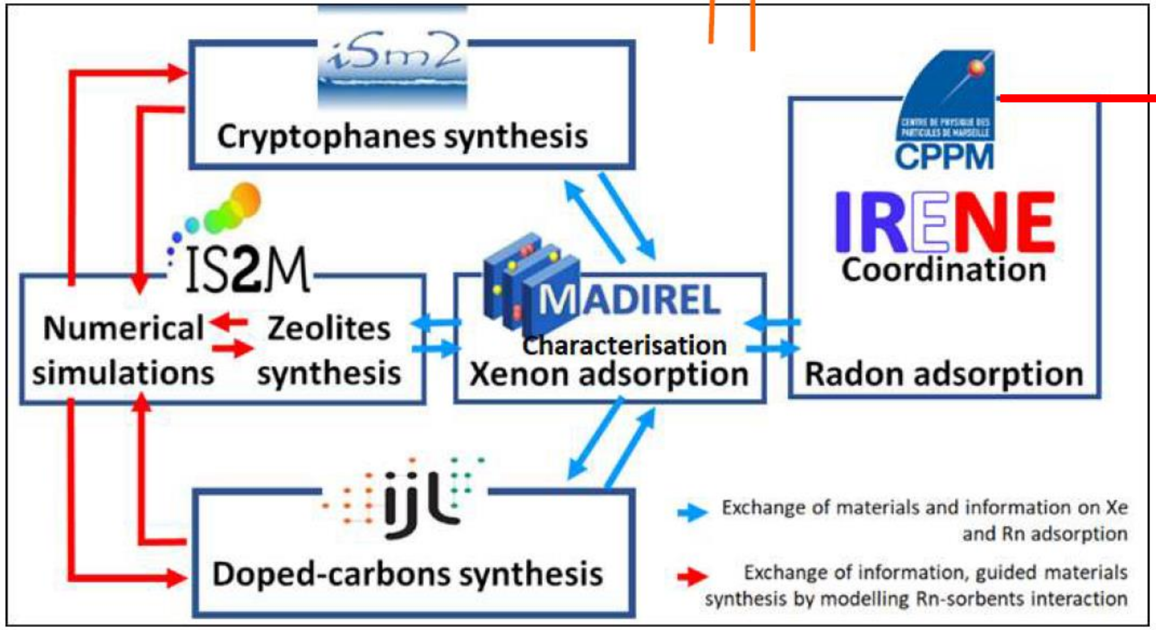
- Feedback between the 5 laboratories
- Modelling as a guide
- 3 families of adsorbents:
  - Zeolites
  - Mol. Cages
  - Carbon based
- Xenon as a reference gas
- Radon capture in N_2,He and Xe

IRENE Project

Innovative mate**R**ials for **E**xtreme rado**N** captur**E**

Understanding and optimising the capture of Rn in new materials

Physics Chemist   Part Physic



Radon adsorption measurement

Radon adsorption coefficient "K"

K is the volume of radon a sample can capture:

- $K\left[\frac{m^3}{kg}\right] = \frac{C(Rn)_{Sample}[^{Bq}/_{kg}]}{C(Rn)_{gas}[^{Bq}/_{m^3}]}$

The retention time τ is the time it take for Rn to exit the adsorbent

- $\tau\ [s] = \frac{m[kg]*K\left[^{m^3}/_{kg}\right]}{\phi\left[^{m^3}/_{s}\right]}$

And the reduction factor R is define as:

- $R = \frac{Rn_{out}}{Rn_{in}} = e^{-\frac{\ln(2)}{T_{1/2}(Rn)}*\tau}$

K can be expressed as an Arrhenius-type law:

- $K = K_0\, e^{\frac{Q}{RT}}$

K increased exponentially with respect to 1/T

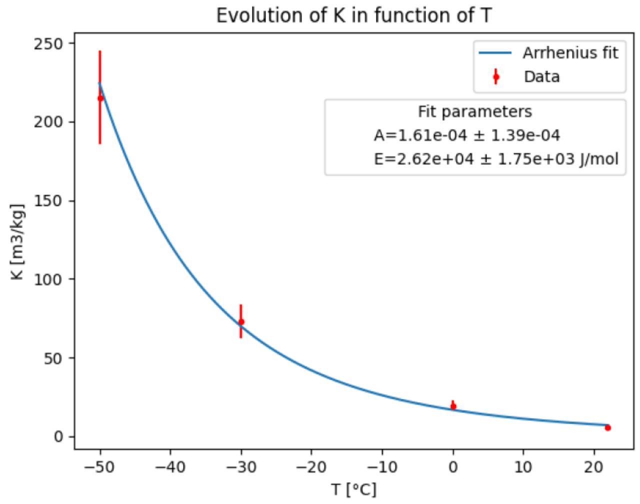


Figure 3.1: Graph of the variation of K with respect to the temperature for an adsorbent measured at CPPM. The fit is an Arrhenius-type law with factor $A = 1.61*10^{-4} \pm 1.39*10^{-4}$ and $E = 26247.17 \pm 1747.08\, J/mol$.

How do we measure the K coefficient of Materials

Dynamical adsorption method

Similarly to chromatography on a column, we force Rn carried by a neutral gas (N_2, Ar, He, etc) through the adsorbent. When it is totally filled with Rn, we measure the Radon concentration inside with an HPGe. We can then calculate the K coefficient described previously.

- Measure the absolute K coefficient
- Reproductible measurement
- Take a long time (1 sample/day/bench)

- **Done in 3 step:**
  1. Activation of the sample
  2. Radonisation
  3. HPGe analysis

Activation

Objective:
- Ensure the reproducibility
- Maximum radon adsorption

Purpose:
- Removal of all traces of physically and chemically adsorbed molecules or atoms

How:
- Heating of the sample under vacuum or flow of neutral gas

Temperature and condition depends on adsorbent:
- AC/Zeolite → 200°C+
- MOF & Molecular cage → <100°C

CPPM activation bench:
- Temperature between 20 and 250°C
- Flow of N_2/Ar/He
- Vacuum

Heat control

Flow controller

Heater

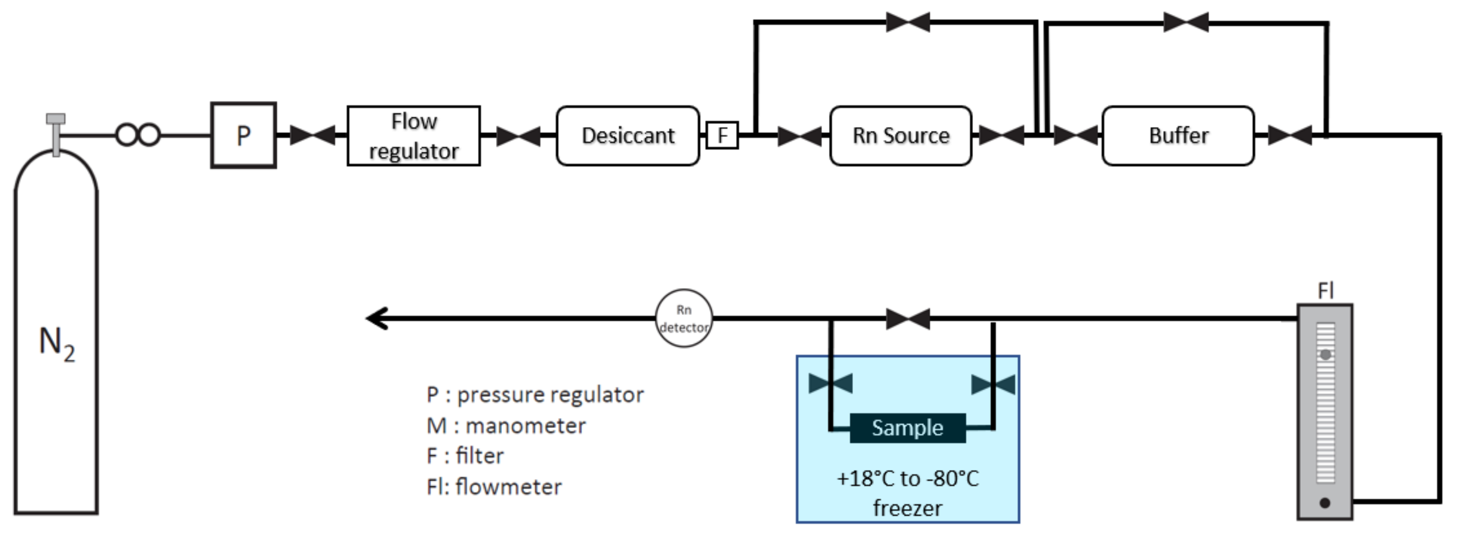
Radonisation

Set up:

- Send a constant concentration of radon through the sample
- Control of radon concentration with a Rn detector at the output
- Sample cooled at up to -80°C

CPPM platform bench:

- “Strasbourg”, open loop, A = 932 ±18 Bq/m3;
- “Marseille”, closed loop, A = 1240 ±41 Bq/m3;



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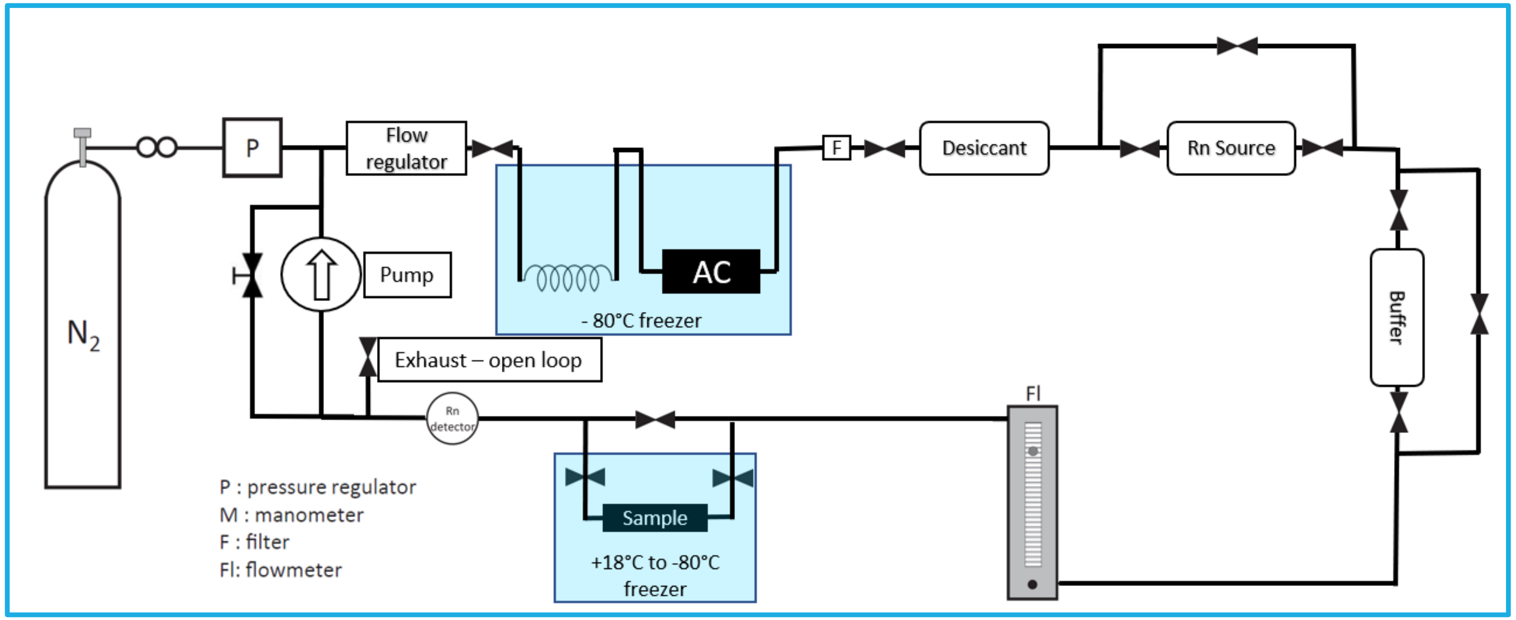
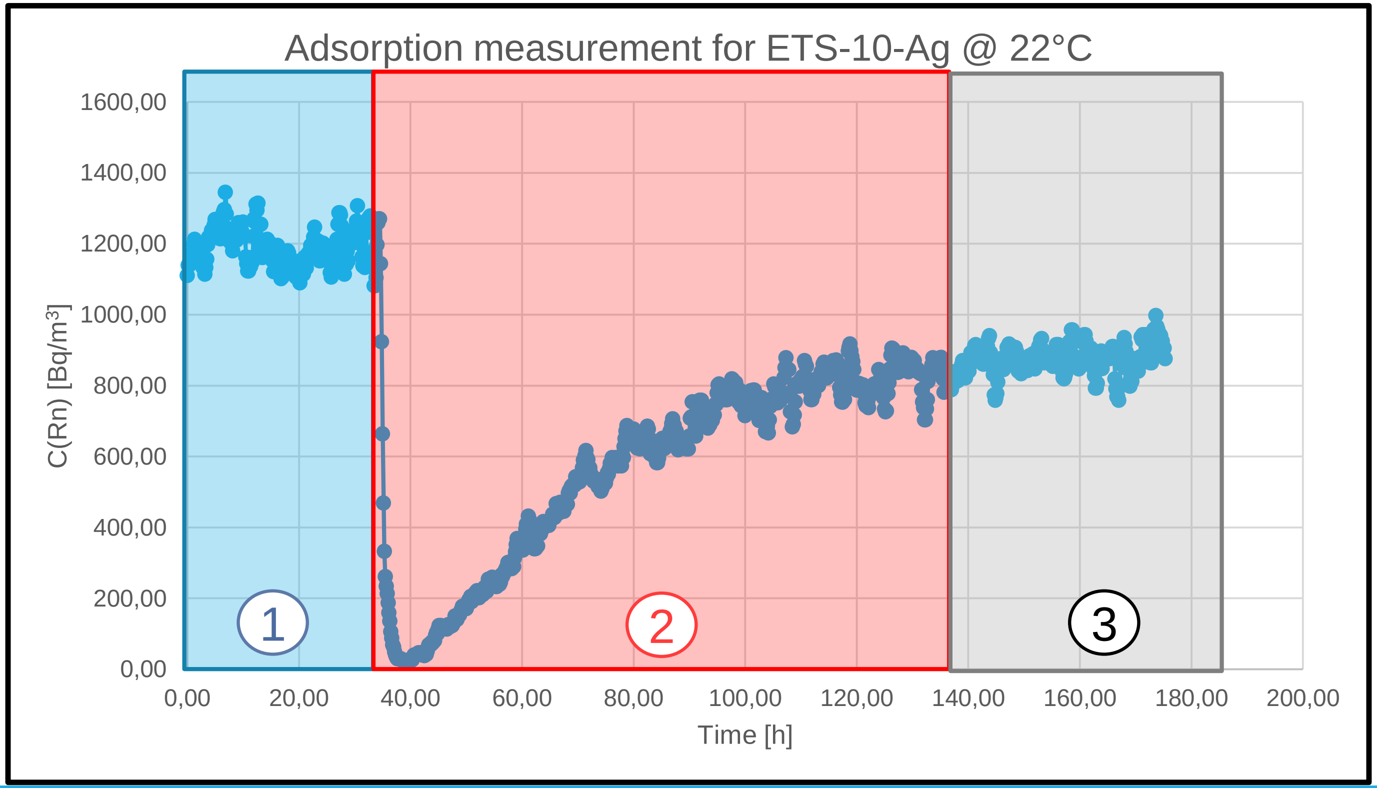


Radonisation

Typical Rn concentration change:

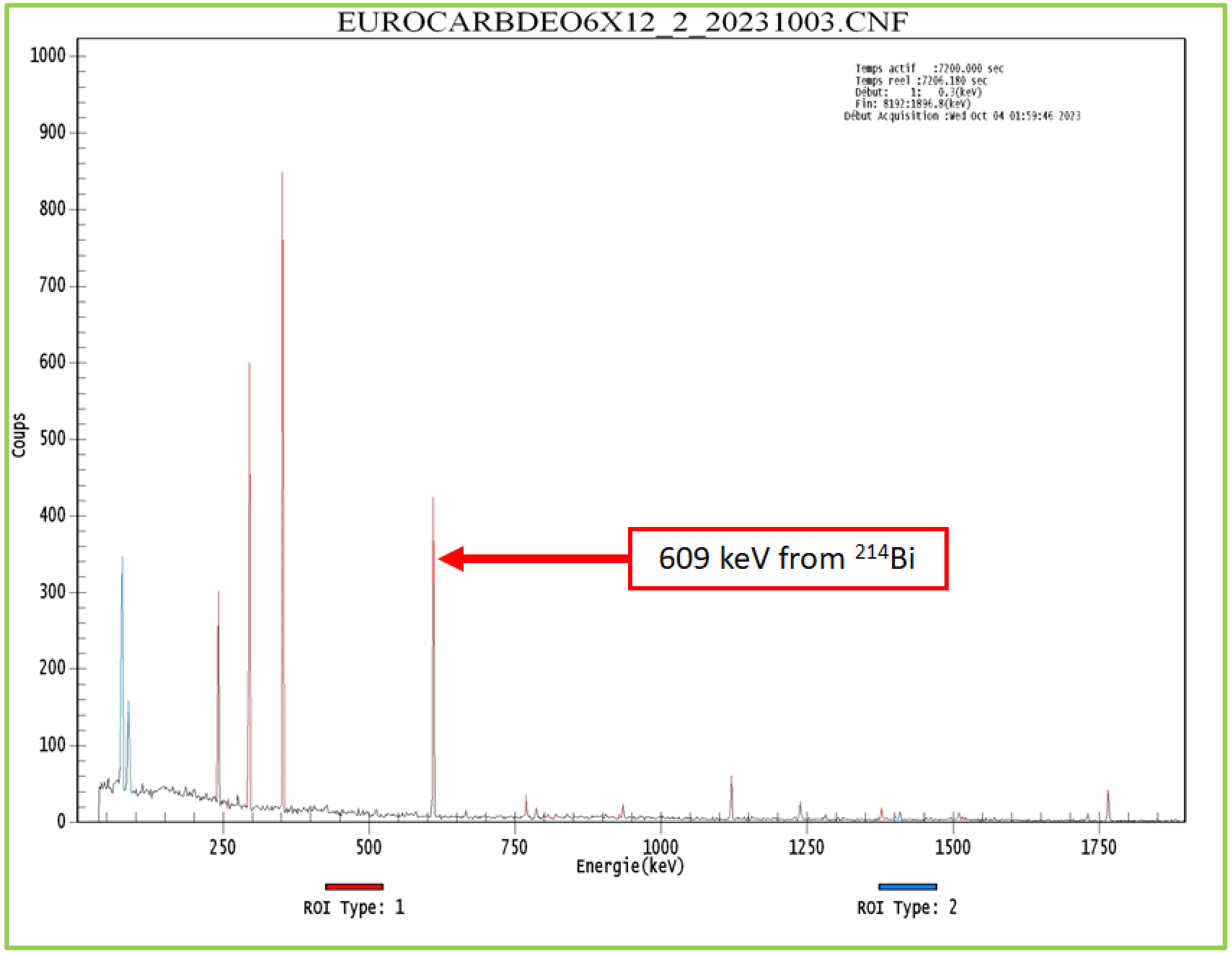
1. Carrier gas is radonised to a concentration C1, “initialization”
2. Flushed through the column, “adsorption”
3. C(Rn) stabilized at a value C2, “saturation”



HPGe measurement

Measurement of the Radon inside the sample by gamma spectrometry

→ 100 cm^3 HPGe shielded by 15cm lead shielding
→ Measurement of ^{214}Bi 609 keV gamma line
→ A_{sample} in Bq/kg

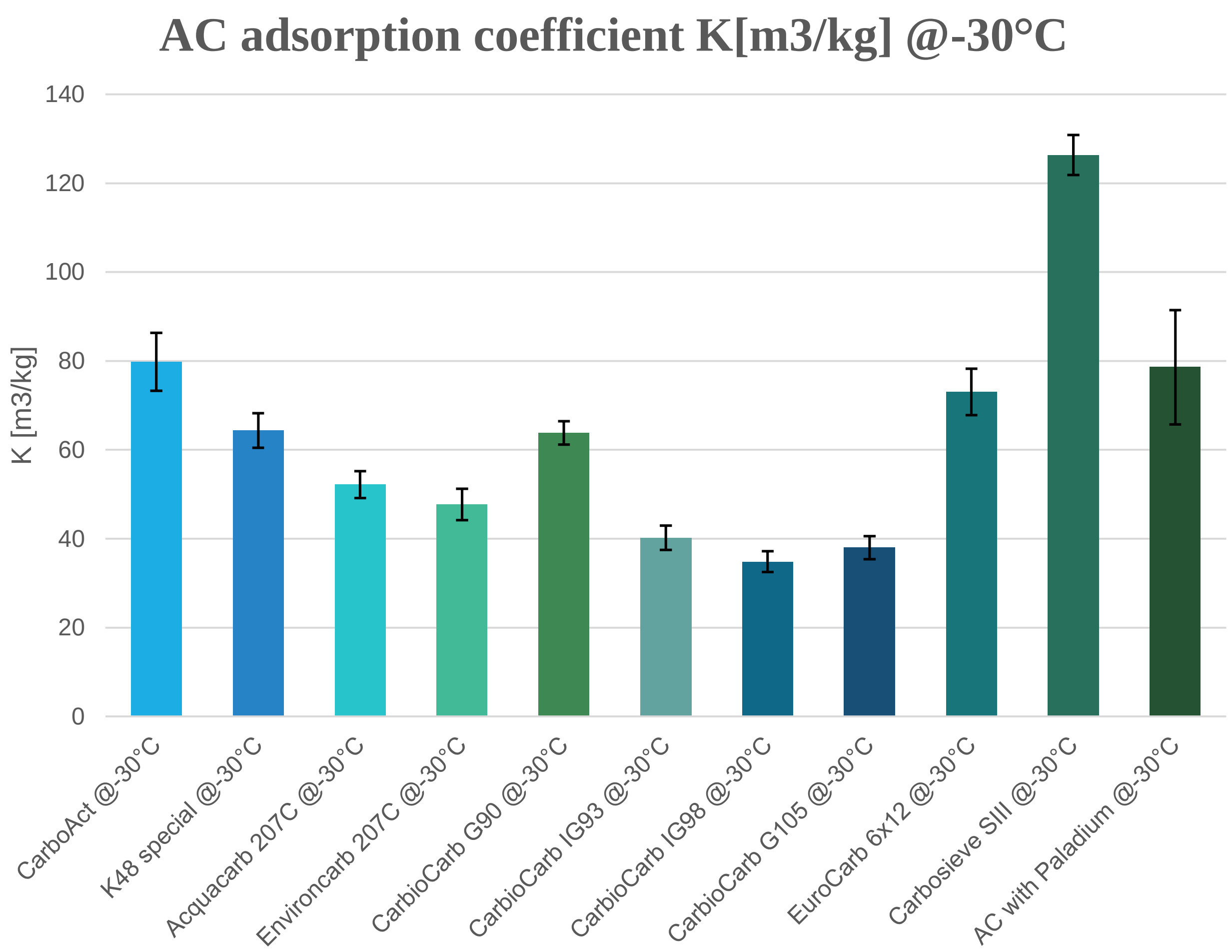


Adsorption coefficient measurement

Measurement of:

- Active Charcoal
- Molecular sieves
- Carbon aerogels
- Molecular cages
- MOF
- Zeolite
- Doped charcoal
- Etc

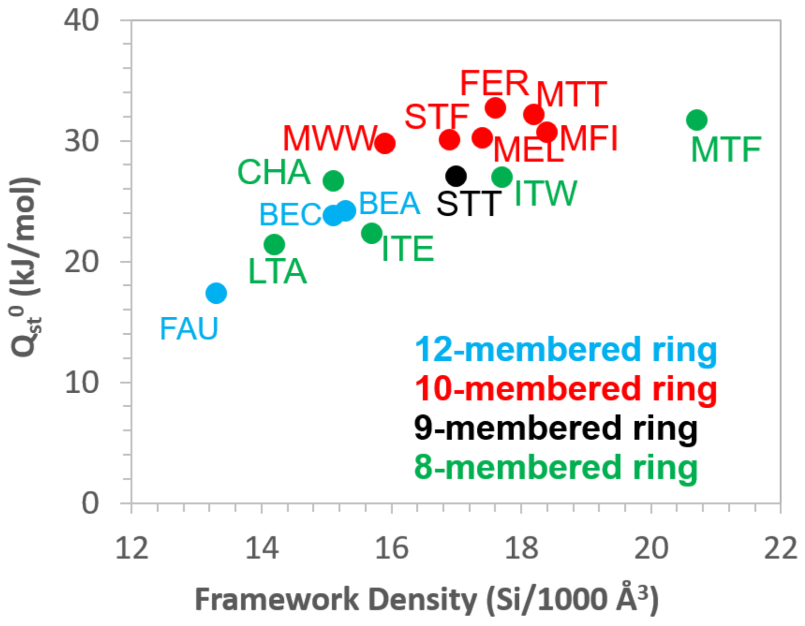
From Ambient temperature to -50°C

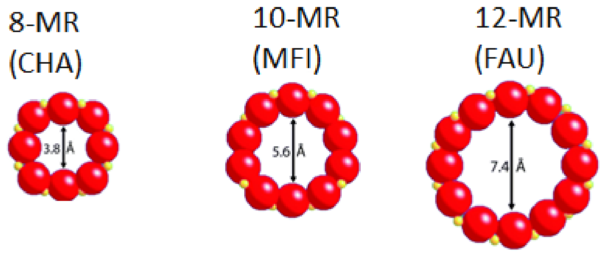


IRENE Project

Preliminary results from simulation

Goal: Find the zeolite with the best performance for Rn adsorption.





Zeolites	Rapport Si:Al min
FER	>6
MWW	>18
MTT	>10
MEL	>20
MFI	>13
STF	>20

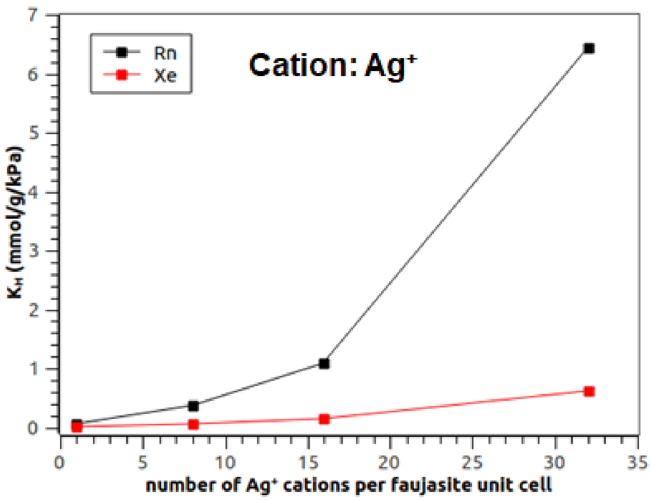
→ Zeolite with 10MR are the best candidate for Rn adsorption

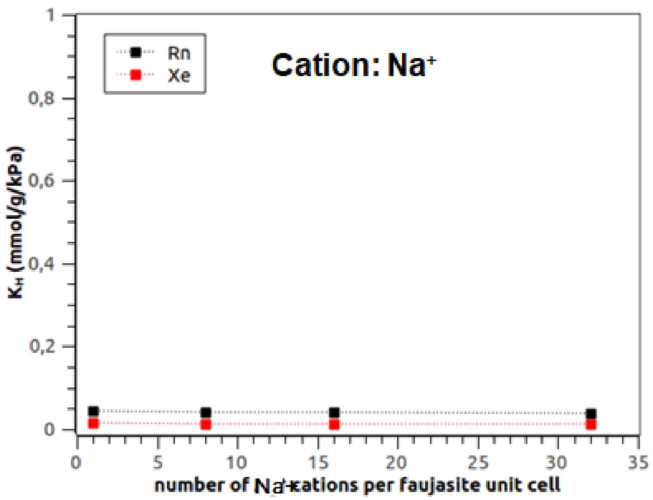
IRENE Project

Preliminary results from simulation

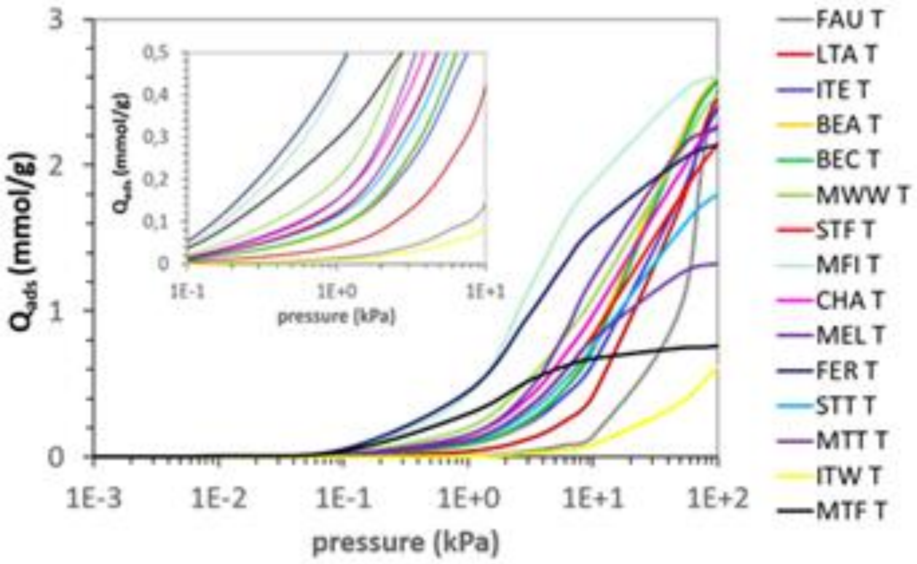
How chemical composition of zeolite influences Rn & Xe adsorption

FAU





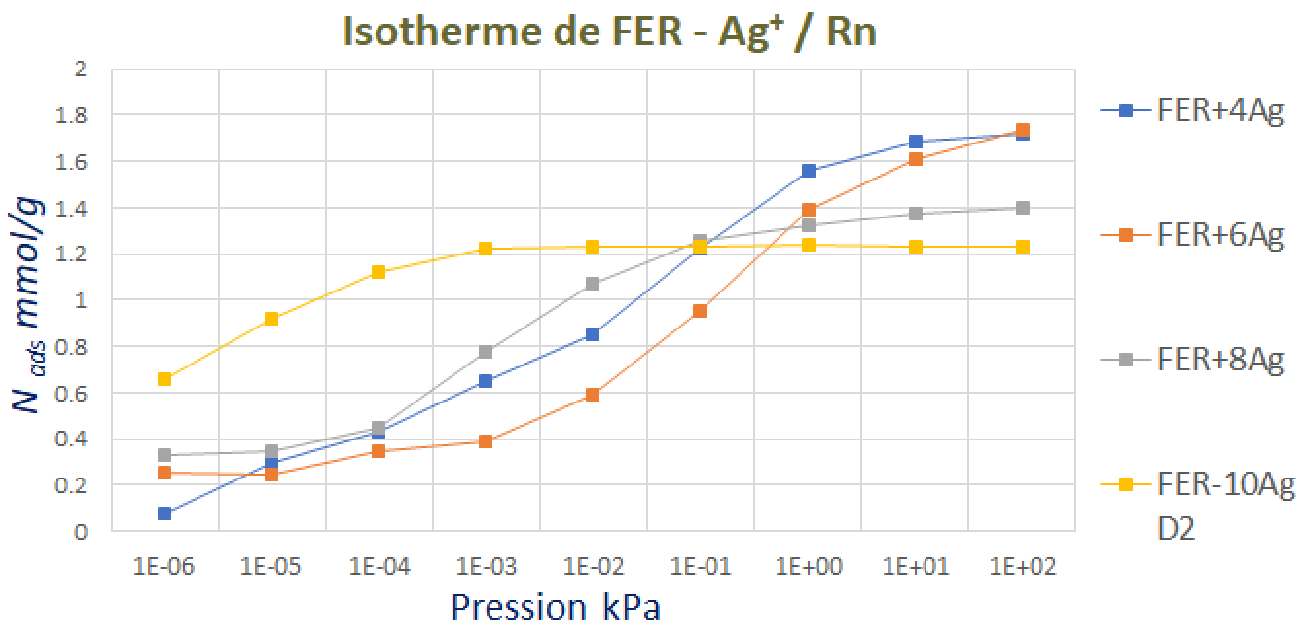
Simulated Rn adsorption isotherms @ 298K for several zeolite structures

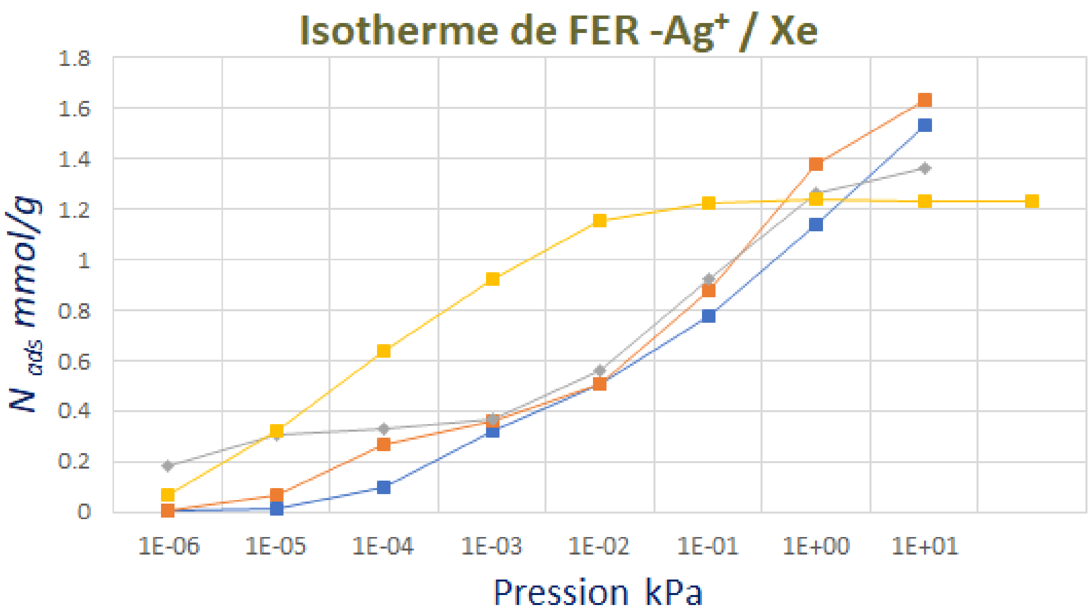


IRENE Project

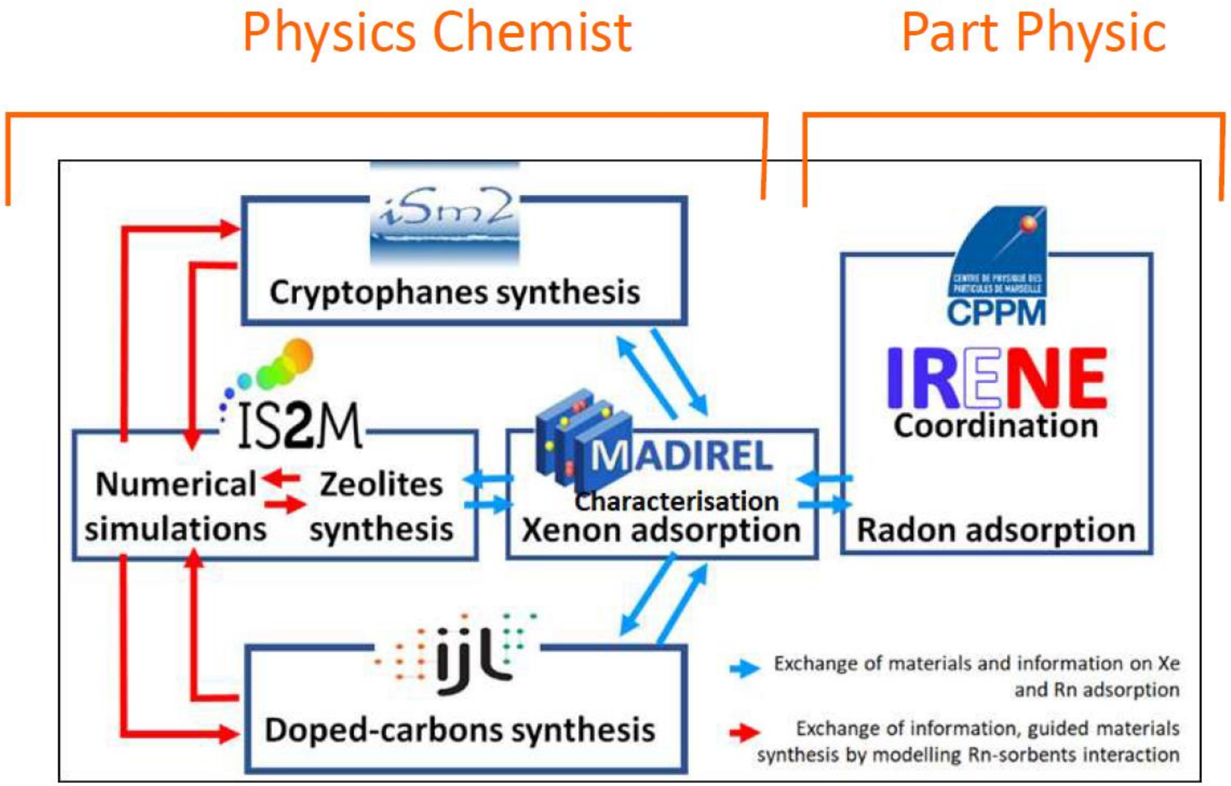
Preliminary results from simulation

Effect of the Si/Al ratio on Rn and Xe adsorption properties for FER with 4,6,8,10 Ag^{+} cations





IRENE Project



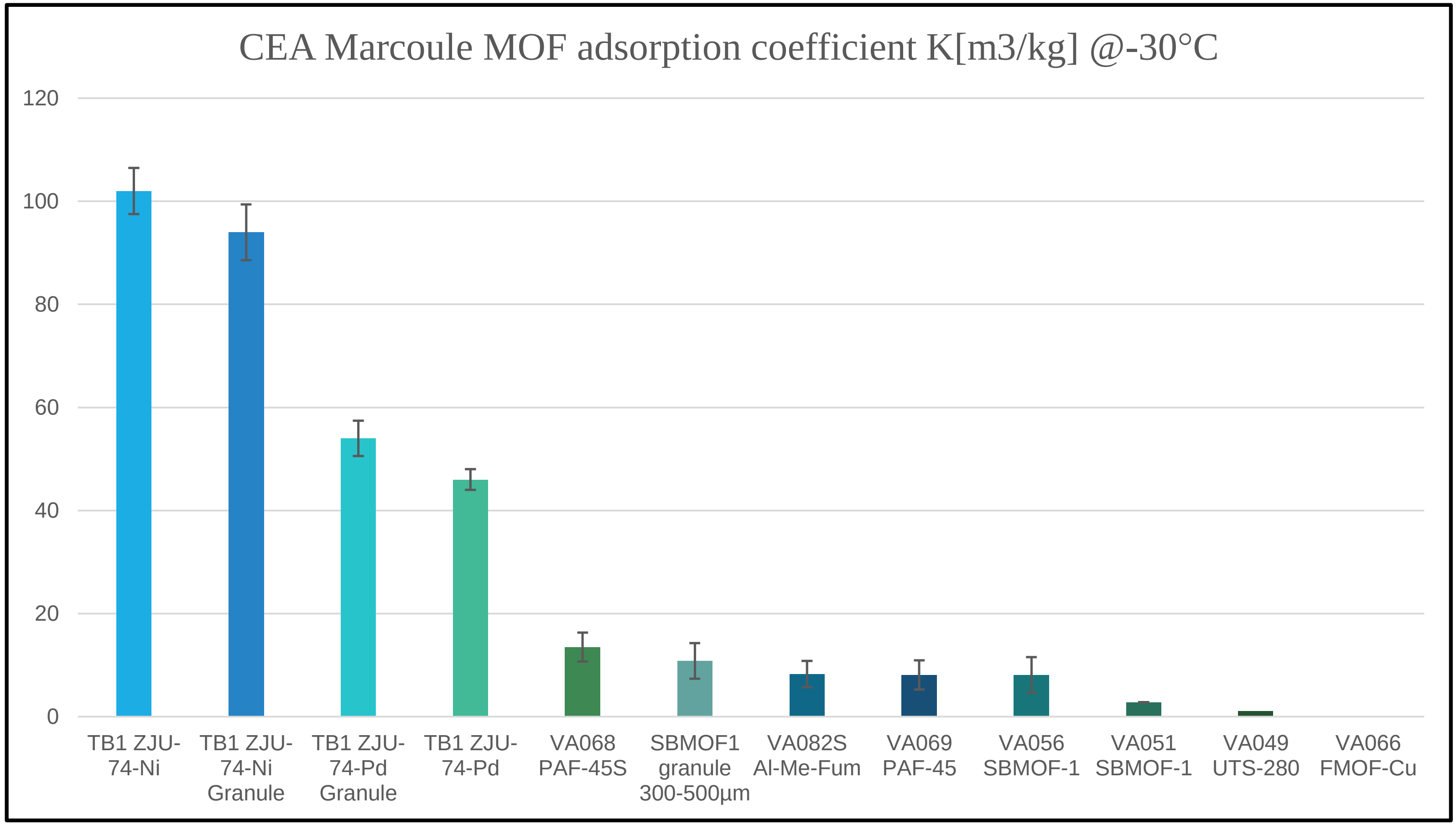
Current phase

- Production of Adsorbent doped with Metal

IRENE Project

Adsorbent measurement

12 MOF from CEA Marcoule :
- MOF with Ni is the most performant



IRENE Project

Measurement of Extremely Adsorbent at high Temperature:

- For extremely adsorbent materials, low temperature measurement is complicated.
- Maximum K measurable for 0,2g is 39 x10^3 m^3/kg; 10^4 m3/kg for 0,8g.

- We can lower the mass, but it increase the uncertainty and enhanced the sensibility to humidity

- K can be expressed as an Arrhenius-type law:
  - $K = K_0\, e^{\frac{Q}{RT}}$

Goal: Estimate the K factor of extremely adsorbent materials at low temperature by measuring K at high temperature and with an higher mass.

→ Adsorption at high temperature is a advantage for the Rn purification of gas mixture.

IRENE Project

Measurement of Extremely Adsorbent at high Temperature:

Preliminary Results with ETS-10-Ag

- ETS-10-Ag measured at -5, 22, 40, 80, 100 and 150°C, mass between 0,8 and 0,9g

Temperature [°C]	K[m³/kg]
-30	19940 ± 5980,8
-12	9951,9 ± 2404,6
-5	6593,8 ± 684,6
22	2332,1 ± 165,5
40	989,1 ± 63,1
80	186,5 ± 11,7
100	136,8 ± 8,0
150	29,3 ± 2,3

From DOI: 10.1093/ptep/ptad160

→ ETS-10-Ag K factor is still significant at high temperature !

IRENE Project

Measurement of Extremely Adsorbent at high Temperature:

Preliminary Results with ETS-10-Ag

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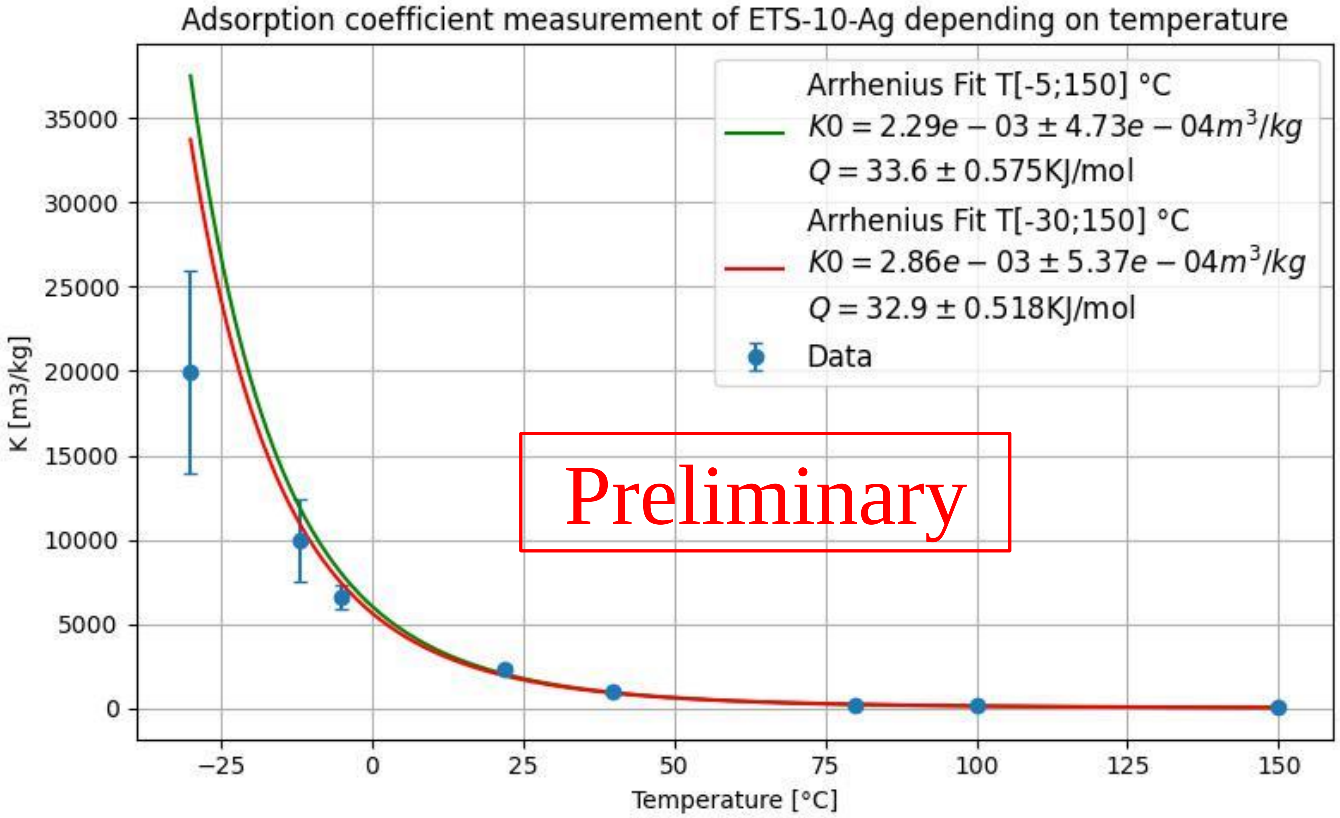

IRENE Project

Measurement of Extremely Adsorbent at high Temperature:

Preliminary Results with ETS-10-Ag

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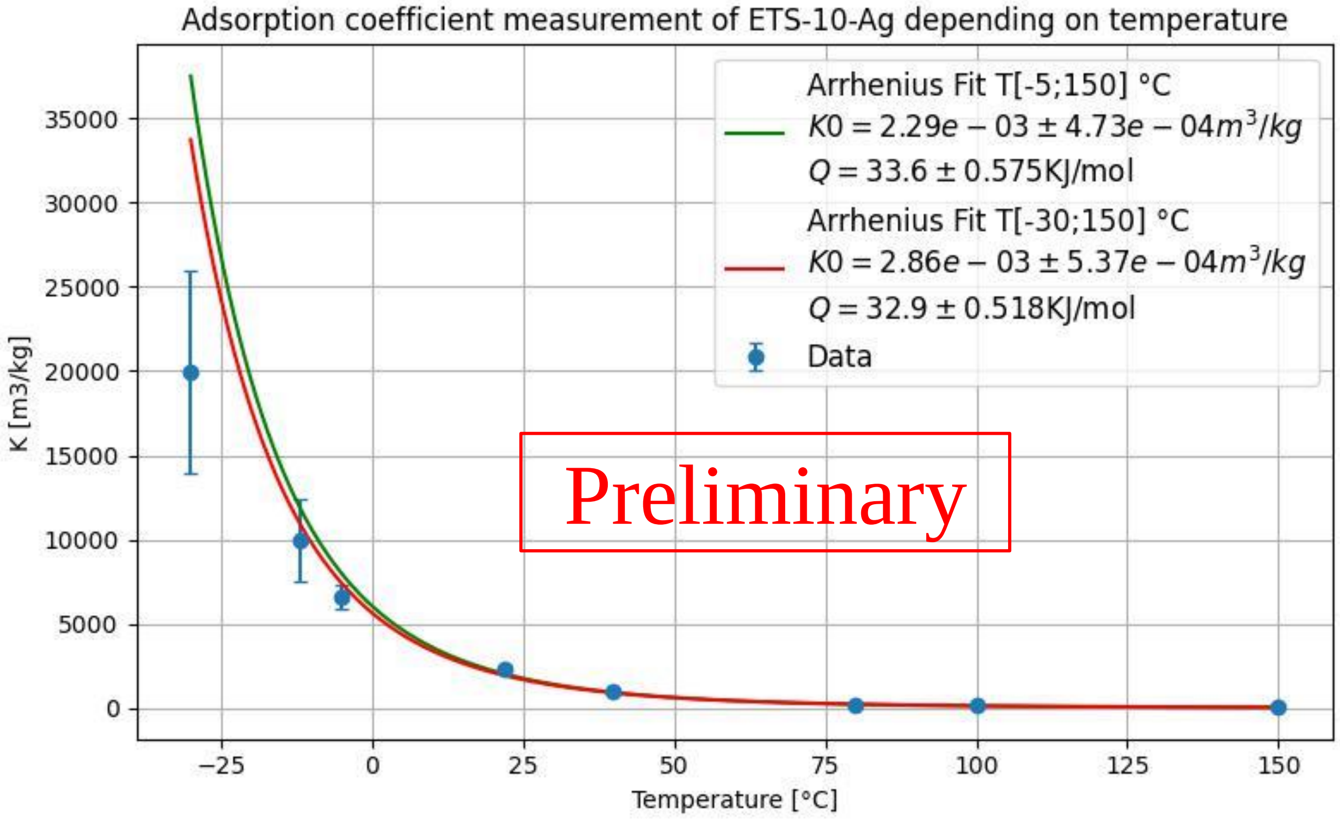


- Fit is in accordance with data up to -12°C.
- Need more measurement at inflexion point to improve K0 and Q estimation.

Conclusion

- Low-energy and low-counting rate experiments require materials with extreme purity and ultra-low radon concentration.

- Materials with adsorption capacities more than 100 times higher than the best active carbons are now commercially available but they do not have the necessary purity levels and their performance are not well understood.

- Possible improvements in radon capture can be reached by choosing the right chemical composition and good porous structure.

- The objective of the **IRENE** project is to tackle the problem of radon capture in microporous materials in a scientific and coherent way, thanks to a unique consortium of physical chemists and particle physicists

- **IRENE** is now in the phase of producing new adsorbent doped with silver and other metals

Thanks for your attention

Back up

Back up

Adsorption

Radonisation

CPPM platform bench:

- "Marseille", closed loop, A = 1240 ±41 Bq/m3;

Side Project: Recycling & Purification system

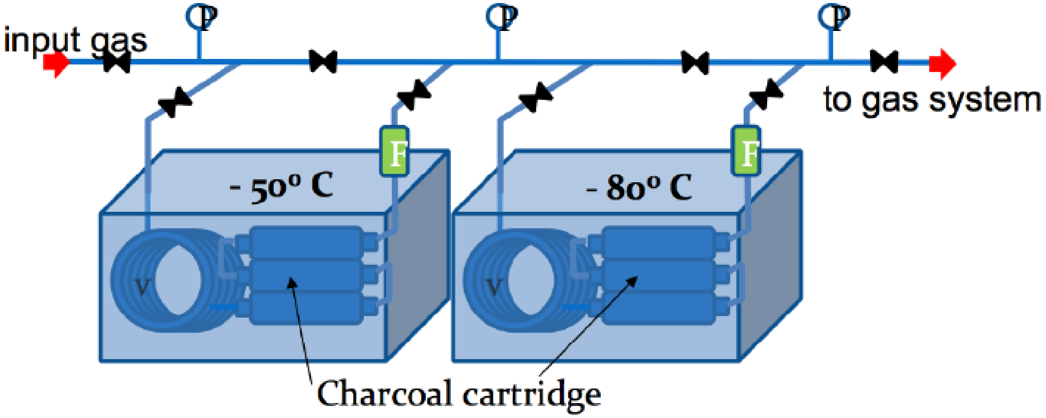
- Gas price inflation since Ukraine/Russian war
- Throw out gas is now too expensive

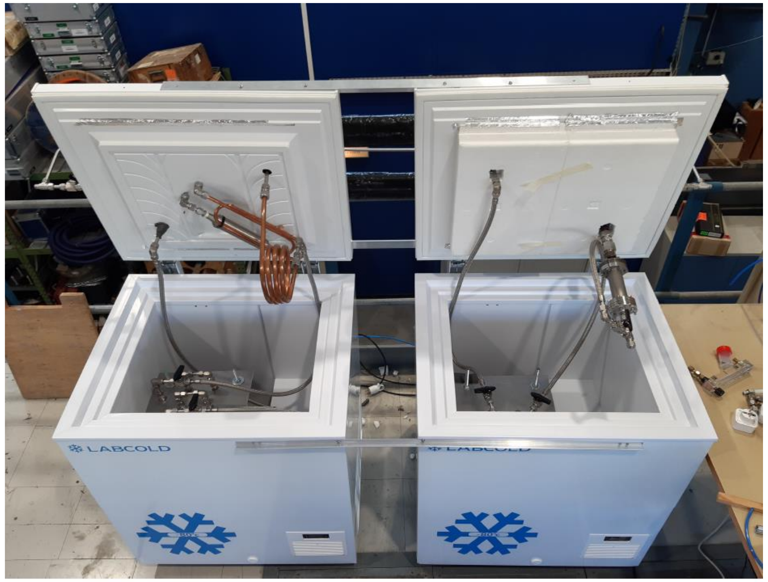
→ Need for recycling system
→ Imply purification of gas from radon and other contaminant

→ Adsorption and gas system knowledge of the CPPM radon platform fit

Three system have been made:

1. **J-Trap**: Radon removal system





Side Project: Recycling & Purification system

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Three system have been made:

1. **J-Trap**: Radon removal system
2. **GPS**: Rn removal and recycling system for XIA

XIA run with Ar from LAr evaporation:
→ 1 run cost 1k€

Objective:

- Use Ar by purifying it and recycling it to reduce the run cost.

Adsorption

Side Project: Recycling & Purification system

- Gas price inflation since Ukraine/Russian war
- Throw out gas is now too expensive

→ Need for recycling system
→ Imply purification of gas from radon and other contaminant

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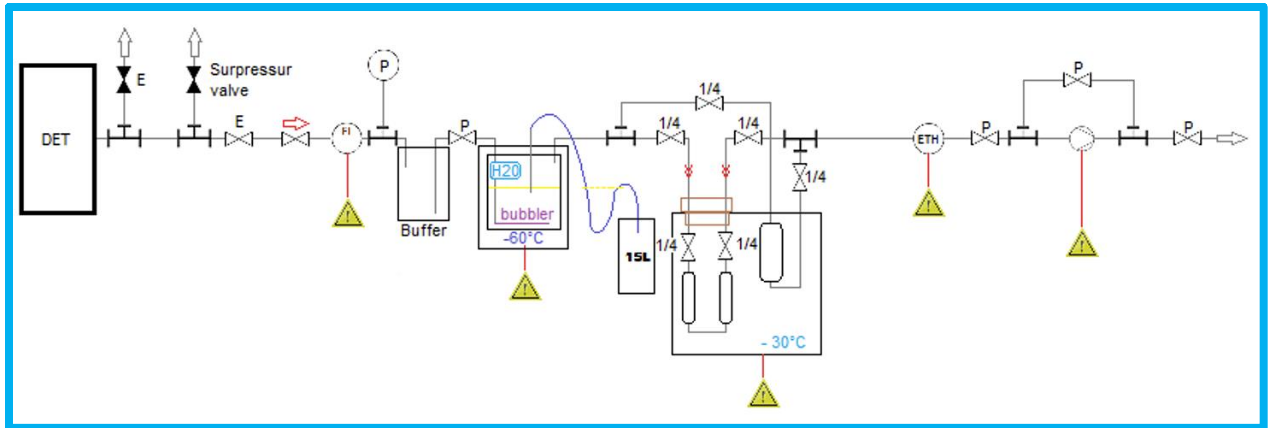
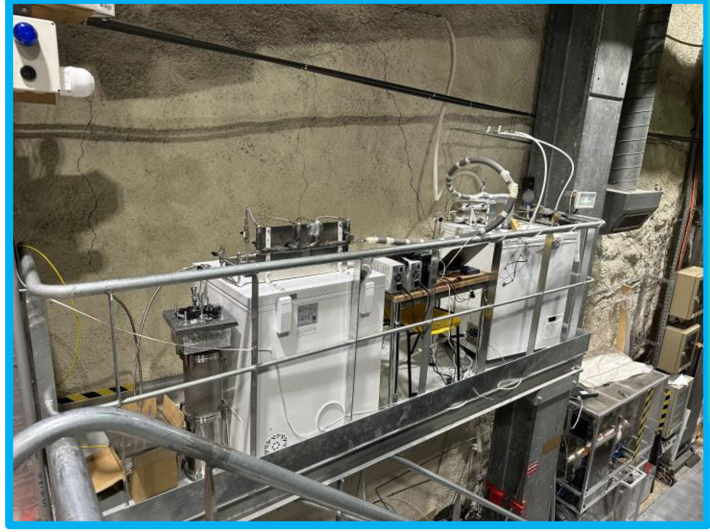
Three system have been made:

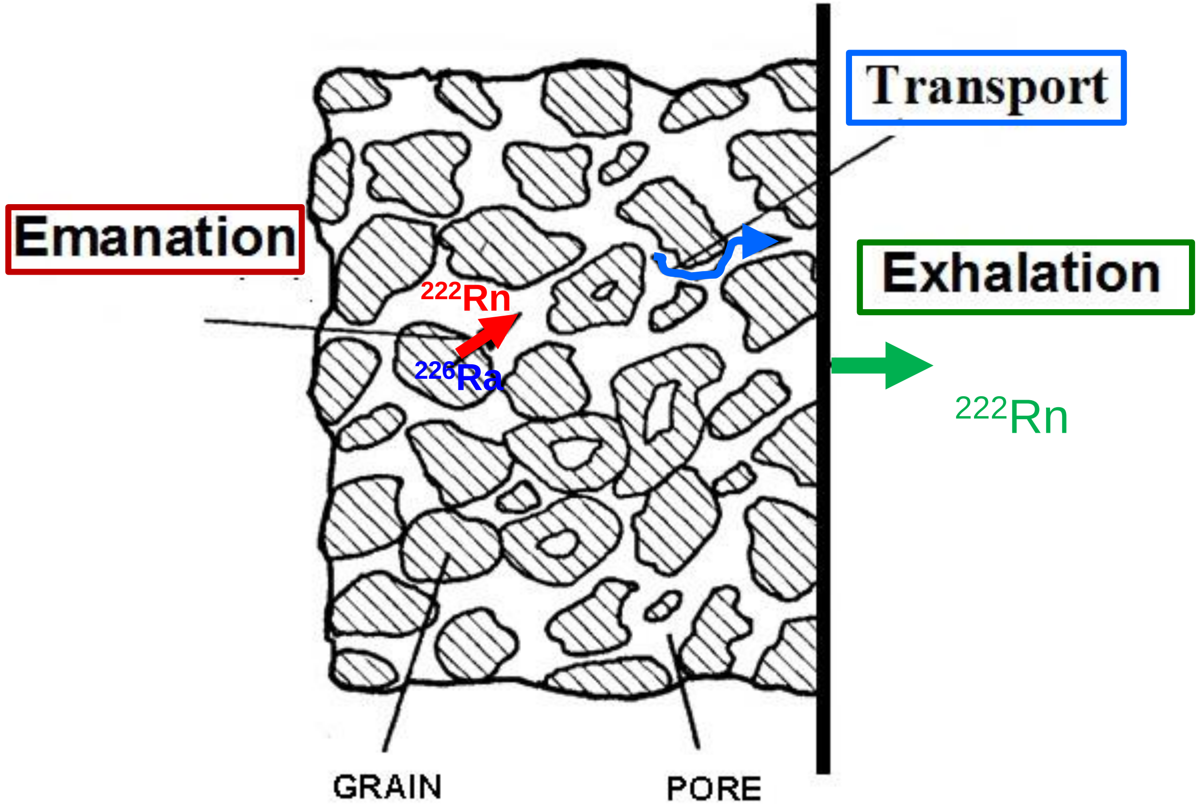
1. **J-Trap**: Radon removal system
2. **GPS**: Rn removal and recycling system for XIA
3. **NESS**: No Ethanol System in SuperNEMO





Installed at LSM August 2024

Emanation/Exhalation



Emanation (^{226}Ra α decay)

- O(5MeV) α-decay creates O(100 keV) ^{222}Rn nuclear recoil
- Recoil range 10 – 100 nm (depending on Z, density)

Transport

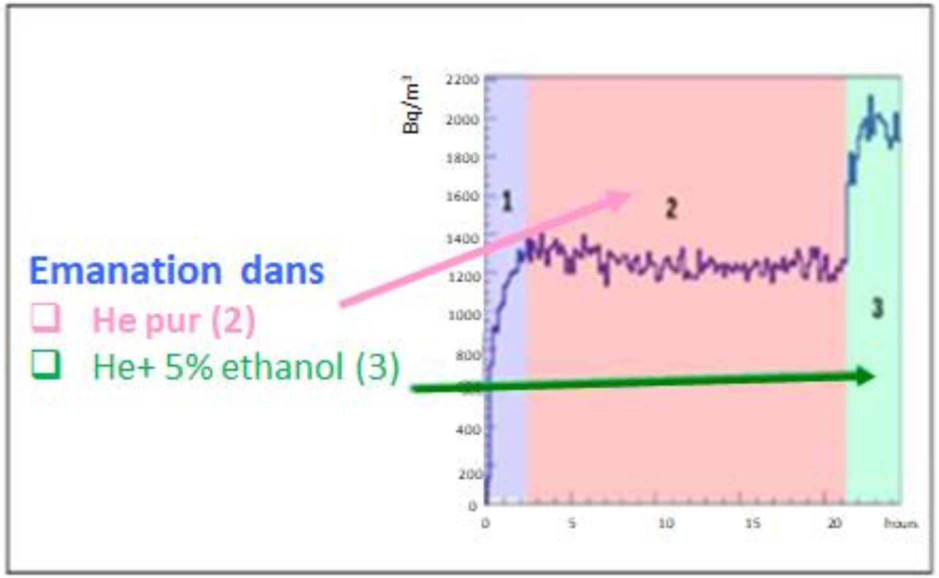
- Tortuosity
- Density
- Inter-grain media
- Strongly depends on temperature

Radon exhalation

- ^{226}Ra concentration
- Porosity
- Temperature
- Pressure
- Medium (gas /liquid)
- Self-adsorption
- …

Influence des liquides sur l'exhalation et le transport

SupeNEMO gas : He + 4 % ethanol + 1 % Ar



JUNO : Rn transport in 5 mm HDPE liner air/H_2O