Study of materials for improved adsorption of xenon at IMS radionuclide stations

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ABSTRACT

Abstract:

Radioxenon monitoring systems are a crucial component of the verification system of the Comprehensive nuclear-Test-Ban Treaty. As part of the International Monitoring System, these systems are monitoring the atmosphere for potential xenon releases originating from nuclear explosions. The efficient adsorption and desorption of the xenon isotopes in adsorbents is essential for the detection capability of these systems.

The SCK CEN was contracted by the CTBTO under the EU JA VII program to perform a fundamental comparative study of commercial xenon adsorption materials (Activated Carbons, Silver-exchanged Zeolites, Metal-Organic Frameworks) which, depending on the results, may be used as future alternatives for noble gas monitoring at IMS stations with the aim of higher detection capability.

Activated Carbon (AC)

Nusorb GXK

Silver-exchanged Zeolite (AgZ)

Ag-ETS-10 (SAUNA III)

Ag-ZSM-5 (SPALAX-NG)

Metal-Organic Framework (MOF)

Ni-DOBDC

HKUST-1

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INTRODUCTION

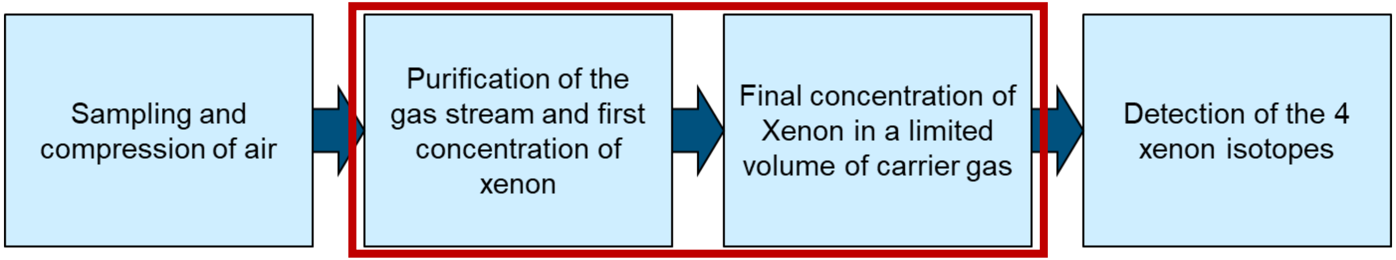
A new class of adsorbents, Metal-Organic Frameworks, has recently been investigated for Xe/Kr separation

- Could these MOFs have the potential to enhance the Xe collection and purification in IMS NG systems ?
- Comparison with three reference materials: Nusorb GXK (AC), Ag-ETS-10 (AgZ) and Ag-ZSM-5 (AgZ)

Important criteria for Xe adsorption and desorption in IMS noble gas monitoring stations

- Efficient Xe collection from air (adsorption)
- Efficient separation of Xe from other gaseous components (desorption)
- Durability against adsorption/desorption cycles

Figure 1 - Summary of the four stages of IMS NG systems to measure radioxenon in the atmosphere.



Potential future alternatives for IMS noble gas systems to:

- Decrease detection limits
- Reduce the energy consumption

?

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METHODS

Xe adsorption

- Xe breakthrough measurements on adsorbents in different conditions such as: Xe concentration, carrier gas, moisture content, impurities, flow rate, …

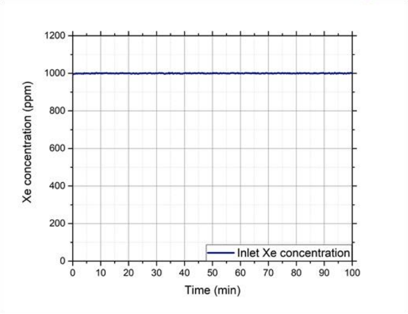
Xe thermal desorption

- Separation of Xe from other gaseous components (O_2, CO_2, Ar and Rn)
- Xe enrichment in the carrier gas and recovery for its successive detection

Durability against adsorption/desorption cycles

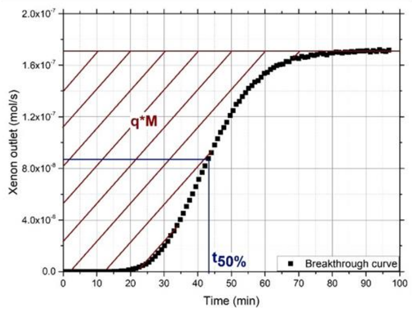
Figure 2 – The gas mixture (Xe, carrier gas and eventually impurities) is sent to the adsorption column and the Xe outlet is measured. Based on the obtained Xe breakthrough curve, the Xe adsorption capacity (q in mol/g) and retention time at a certain percentage of the inlet (e.g. $t_{50\%}$) are calculated. The adsorbent can be thermally desorbed.

Inlet



Adsorbent

Outlet



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RESULTS

Xe adsorption capacity in N_2 at room temperature

- Xe concentrations from 100 ppb to 10 000 ppm
- First time that such low Xe concentrations were investigated for Metal-Organic Frameworks !

Table 1 – Ranking order of the Xe adsorption capacity at 1 ppm Xe in N_2 and ratio of the Xe adsorption capacity compared to the HKUST-1 MOF.

Material	Ratio to HKUST-1
Ag-ETS-10	70
Ag-ZSM-5	19*
Nusorb GXK	2.7
HKUST-1	1.0
Ni-DOBDC	0.40

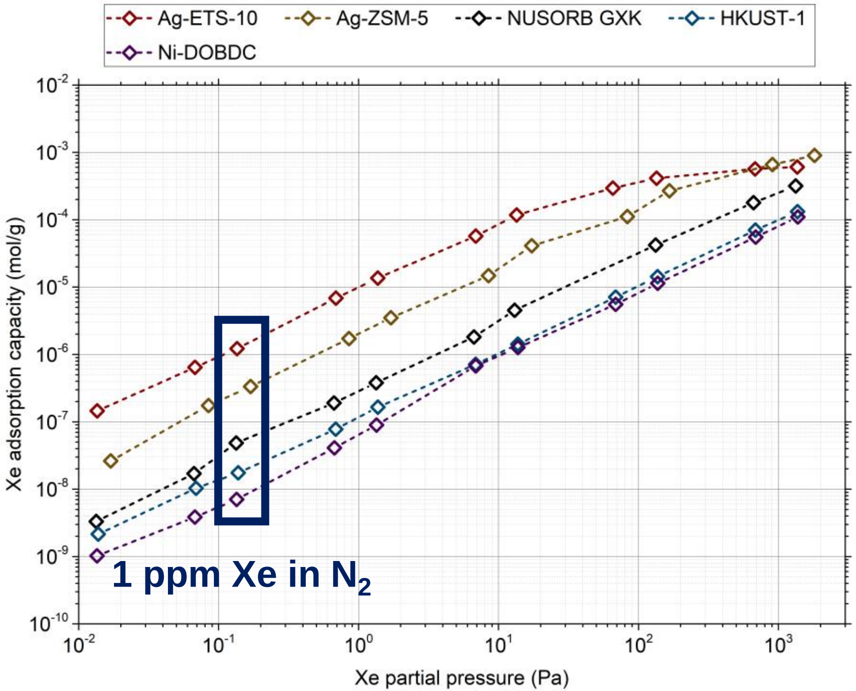

Figure 3 – Xe in nitrogen adsorption isotherms for the 5 adsorbents.

Xe adsorption capacity in air (87 ppb Xe)

- Results are equivalent !

* For the Ag-ZSM-5 a higher Xe adsorption capacity (by a factor 4) was observed during later experiments. This will be further investigated.

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RESULTS

Effect of moisture on Xe adsorption

- Adsorption of 250 ppm Xe in air at room temperature
  - Dry, 5% and 50% R.H.
- No significant effect on AC and MOFs
- **Significant decrease on AgZs at 50% R.H.**

Xe separation from O_2, Ar and CO_2 during desorption

- After adsorption of 250 ppm Xe in air at room temperature
- Very good separation on the Ag-ETS-10 material
- Poorer separation on all other materials

Table 2 – Percentage of contaminants during the thermal desorption of Xe and the corresponding % of Xe recovered in the peak.

Material	Xe (%)	O_2 (%)	Ar (%)	CO_2 (%)
Ag-ETS-10	91%	< 1%	< 1%	1%
Ag-ZSM-5	87%	< 1%	< 1%	**6%**
NUSORB	91%	2%	< 1%	**27%**
Ni-DOBDC	89%	**21%**	**12%**	2%
HKUST-1	90%	2%	1%	**86%**

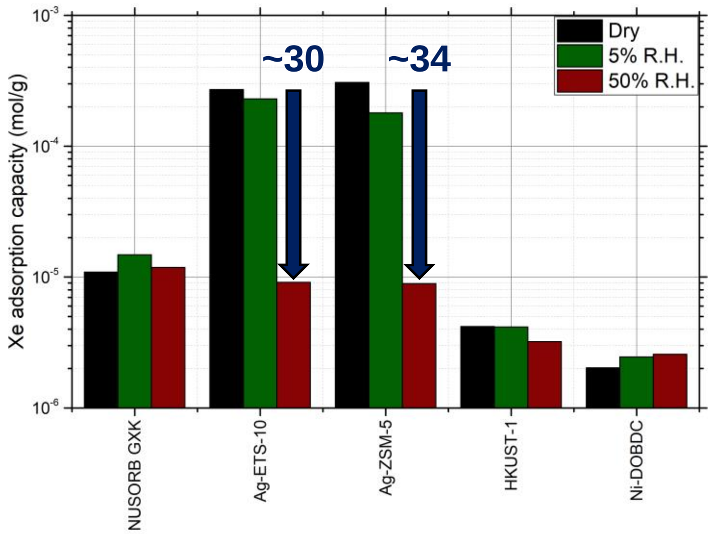

Figure 4 – Xe adsorption capacity obtained with 250 ppm Xe in air and with three moisture contents: dry, 5% and 50% R.H.

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RESULTS

Xe/Rn separation

- Good separation was obtained on all materials except: Nusorb GXK and Ni-DOBDC

Xe enrichment and recovery

- Adsorption 250 ppm Xe in N_2 and enrichment during desorption
- Best Xe enrichment on the Ag-ETS-10 & Ag-ZSM-5 materials

Durability against adsorption/desorption cycles

- Xe adsorption and desorption in various conditions
- Durability monitored through regular adsorption of 250 ppm Xe in dry air

➔No degradation observed, except on the Ni-DOBDC after the 50% R.H. adsorption and desorption

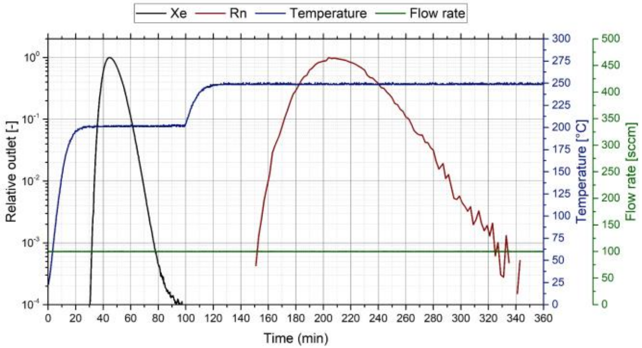

Figure 5 – Example of the Xe/Rn separation during the thermal desorption of the Ag-ETS-10.

Table 3 – Xe enrichment in the thermally desorbed peak and the corresponding % of Xe recovered in the peak.

Material	Xe (%)	Xe enrich.
Ag-ETS-10	90%	382
Ag-ZSM-5	90%	275
NUSORB	90%	54.8
Ni-DOBDC	92%	24.8
HKUST-1	90%	10.00

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CONCLUSIONS

- The two AgZs investigated are outperforming all other materials
  - Xe adsorption capacity in nitrogen, Xe separation from O_2, Ar, CO_2 and Rn, and Xe enrichment
  - Except at 50% R.H. !
- The two MOFs were not providing any advantage compared to the three reference materials
  - It was the first time these MOFs were investigated for such an application

- Many other MOFs are being developed, at laboratory scale, for Xe/Kr separation and could potentially also have interesting properties for IMS NG systems
  - New MOFs should be monitored for their application in IMS NG systems !

Full reports accessible at https://publications.sckcen.be/portal/

Gueibe, C., Rutten, J. and Camps, J. (2019). Study of materials for improved adsorption of xenon at IMS radionuclide stations: Task 1 – Selection of materials and initial comparison study. SCK CEN Open Report BLG-1214.

Gueibe, C., Rutten, J. and Camps, J. (2020). Study of materials for improved adsorption of xenon at IMS radionuclide stations: Task 2 – In-depth studies of selected materials. SCK CEN Open Report BLG-2923.