On the radiation resistance and thermal durability of silver-exchanged zeolites for trapping radioxenon

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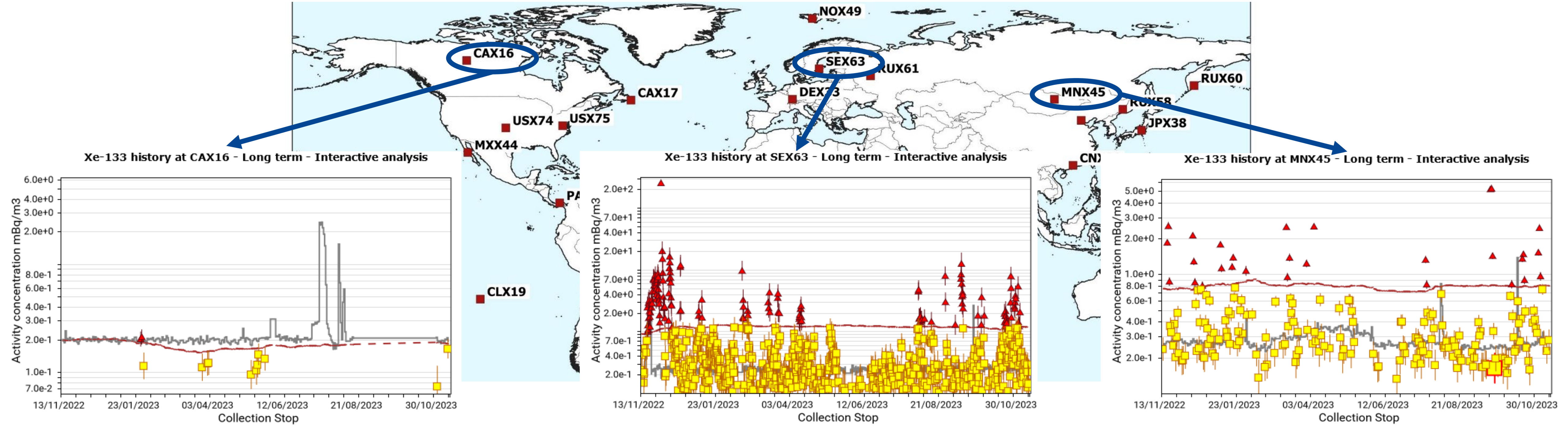
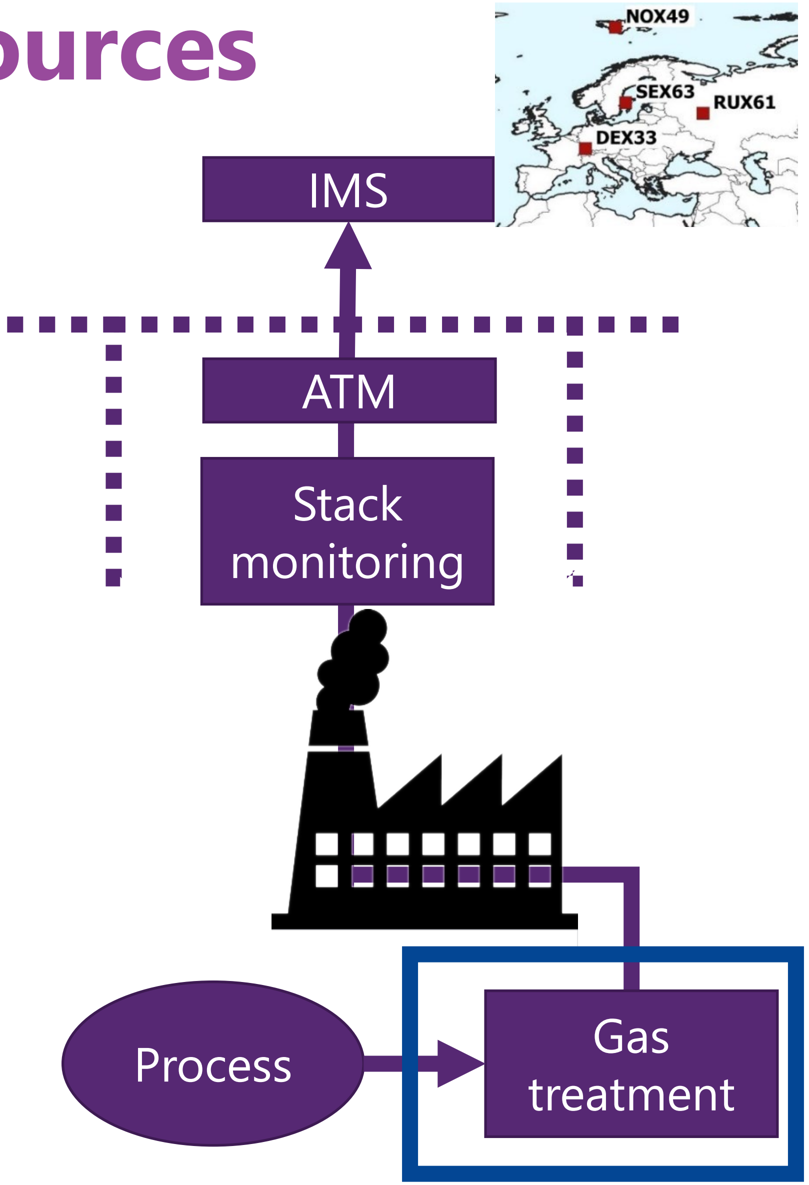
Introduction

- Radioxenon is a key component for the verification of the CTBT
- Detection capability of the IMS noble gas component depends on
  - Number and distribution of stations (31/40)
  - Minimum Detectable Concentration (< 1mBq/m^3 for Xe-133)
  - **Background level from civilian sources at individual stations**



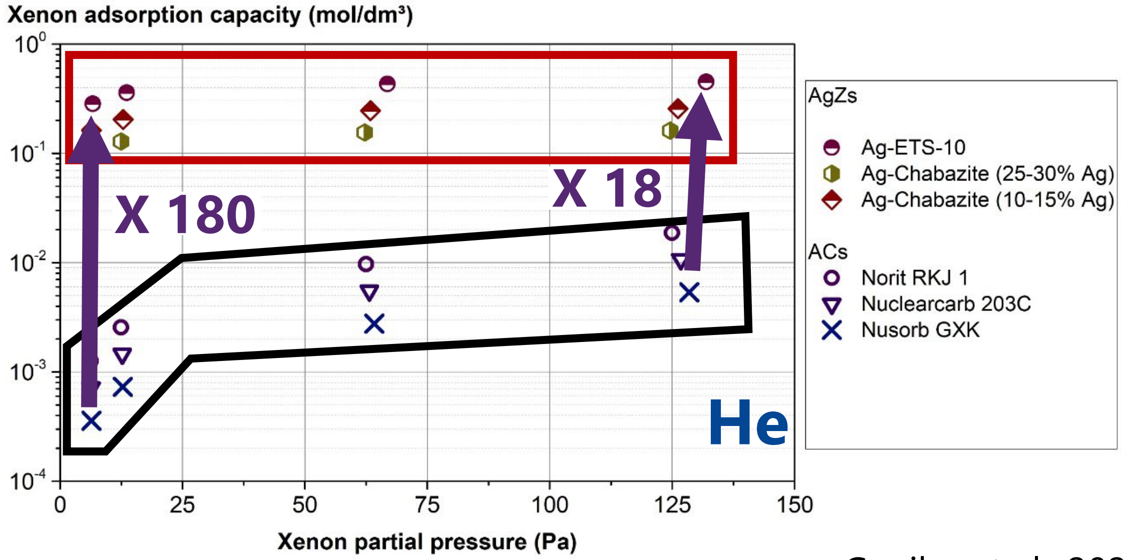
Minimizing the impact of civilian sources

- Further improve IMS stations to maximize the screening capabilities for the four CTBT-relevant isotopes
- Better understand the sources contributing to the civilian background
- Use of stack monitoring for predicting the civilian background by Atmospheric Transport Modelling
- Further reduce radioxenon emissions from civilian sources (specifically to minimize the impact on the CTBT)

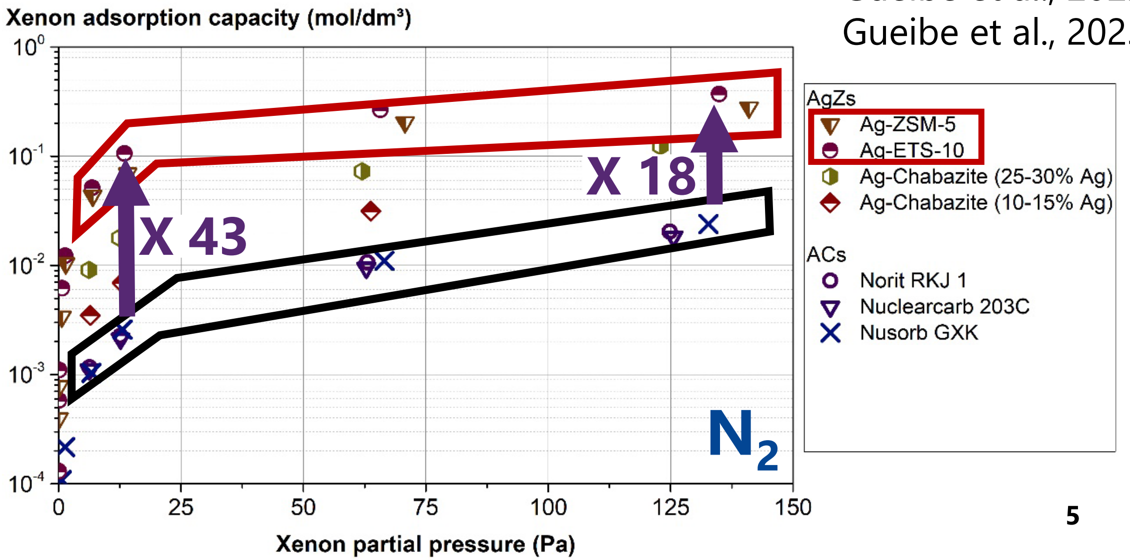


Further reduce radioxenon emissions ?

- Silver-exchanged zeolites (AgZs) are more efficient than activated carbon
  - Room temperature
  - $P_{xe} < 1000$ Pa
  - Xe in He
    - And in N_2 (also Ar)

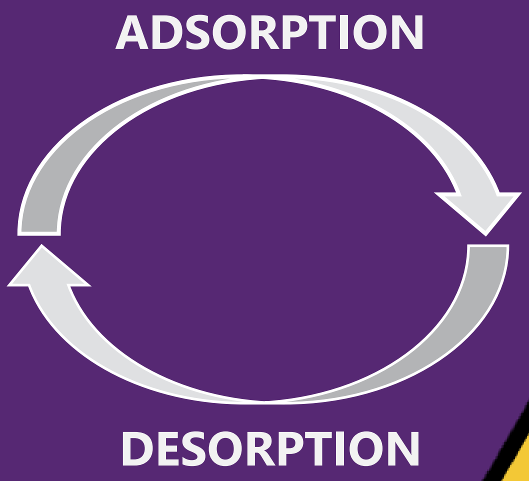


Gueibe et al., 2022
Gueibe et al., 2023



But AgZs are more sensitive to moisture
➔ Moisture traps are needed

What is their radiation resistance and thermal regeneration durability ?



First exploration on Ag-ETS-10

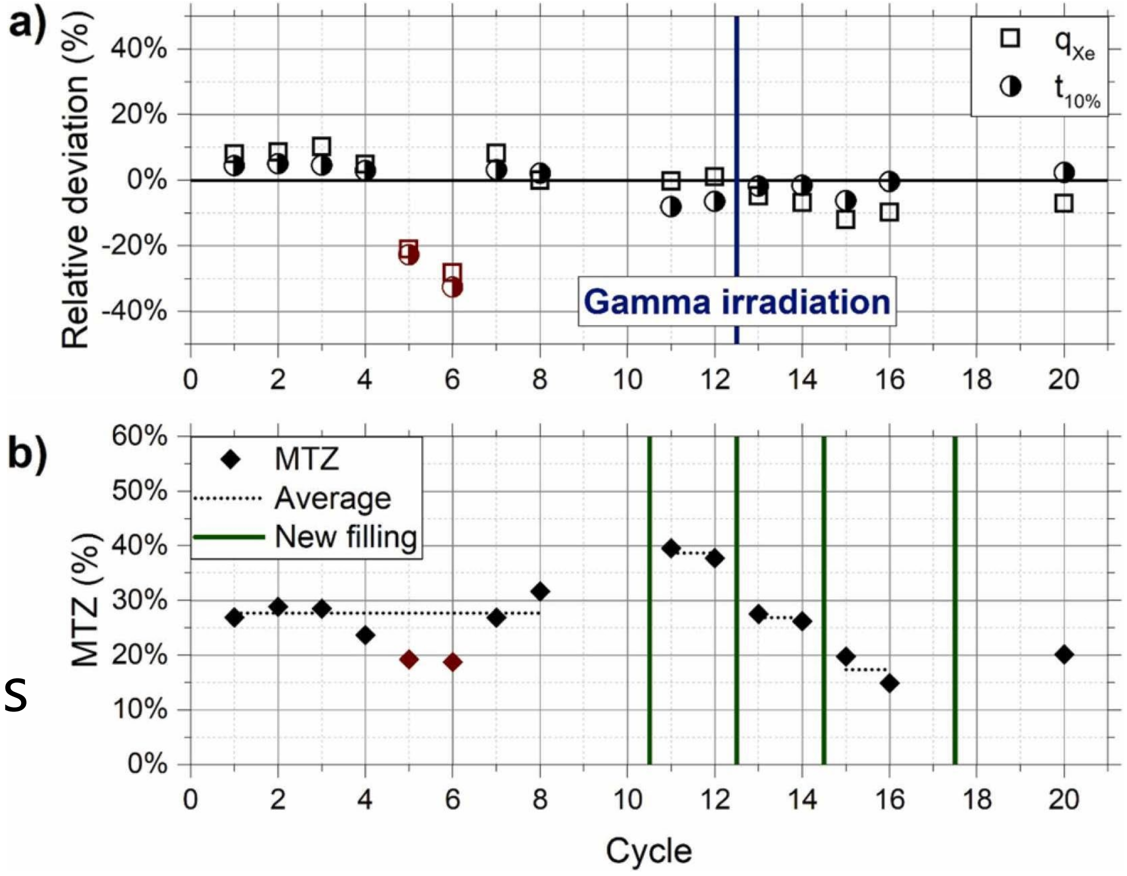
Gueibe et al., 2022

- Thermal regeneration durability
  - Regeneration at 170 – 235°C under He
  - Adsorption of 1000 ppm Xe in He

➡ **No significant variation on q_{Xe} & $t_{10\%}$**

- Radiation resistance
  - External gamma irradiation of 1 MGy
  - “Only” a few hours of operation at MIPFs

➡ **No significant variation on q_{Xe} & $t_{10\%}$**



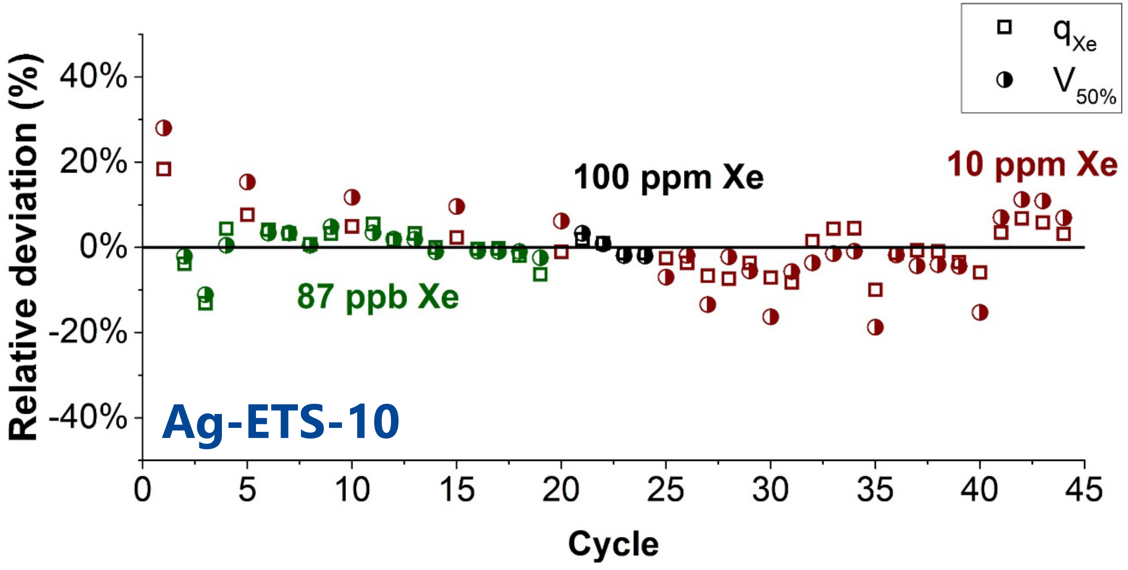
Variations on Mass Transfer Zone (MTZ) are due to packing

New thermal durability investigation

- 44 cycles on Ag-ETS-10
  - Regeneration at ±210°C (+ test at 260°C) under N_2 (+ test with air)
  - Ads.: 0.087, 10 and 100 ppm Xe in air

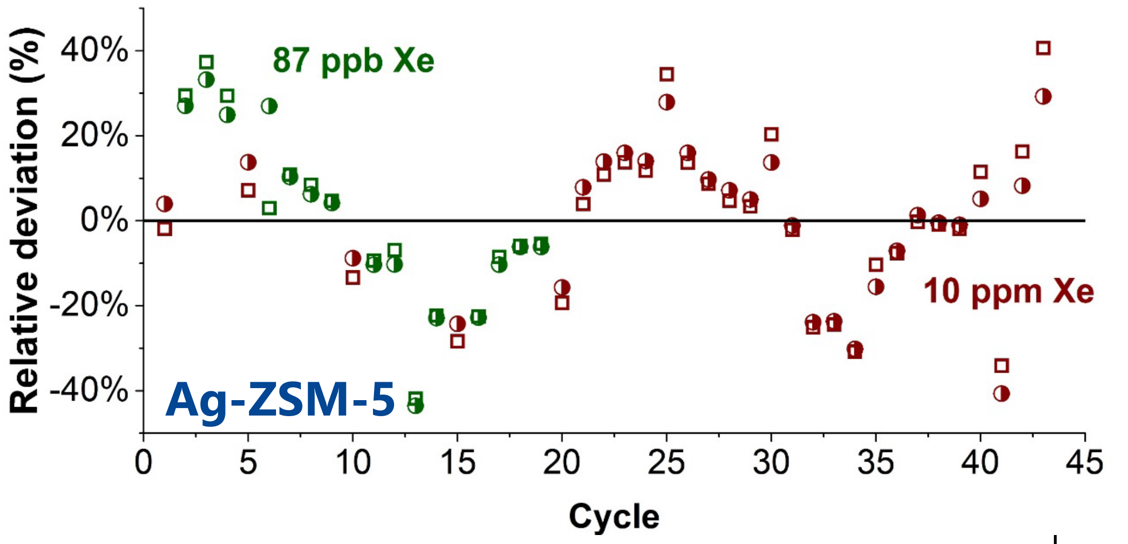
➡ **No significant variations on q_{Xe} & $t_{10\%}$**

➡ **Variations on MTZ**



- 43 cycles on Ag-ZSM-5
  - Regeneration at ±210°C (+ test at 260°C) under N_2 (+ test with air)
  - Ads.: 0.087 and 10 ppm Xe in air

➡ **Variations on q_{Xe} & $t_{10\%}$ likely due to regeneration duration**



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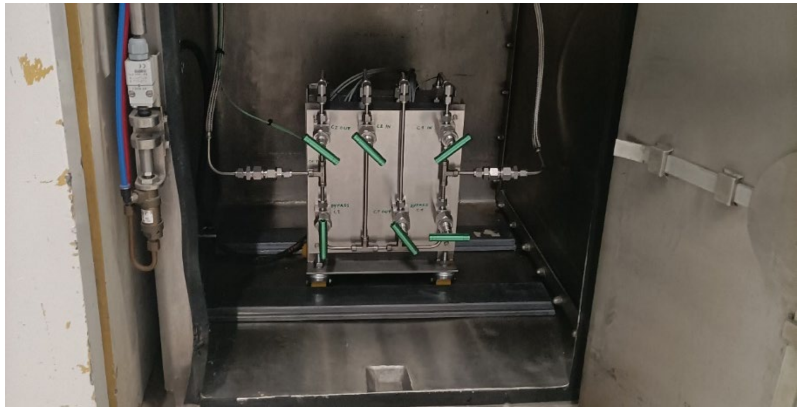
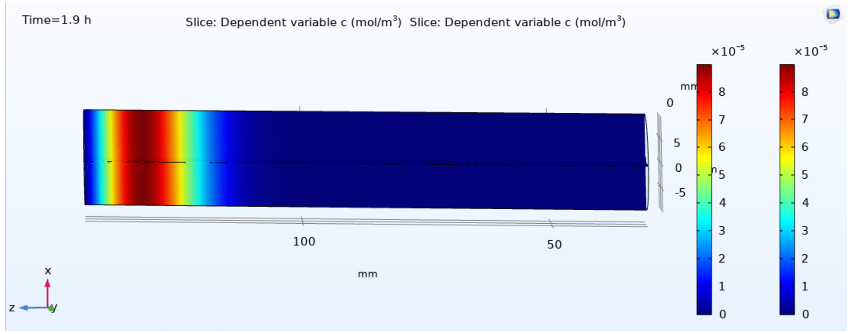
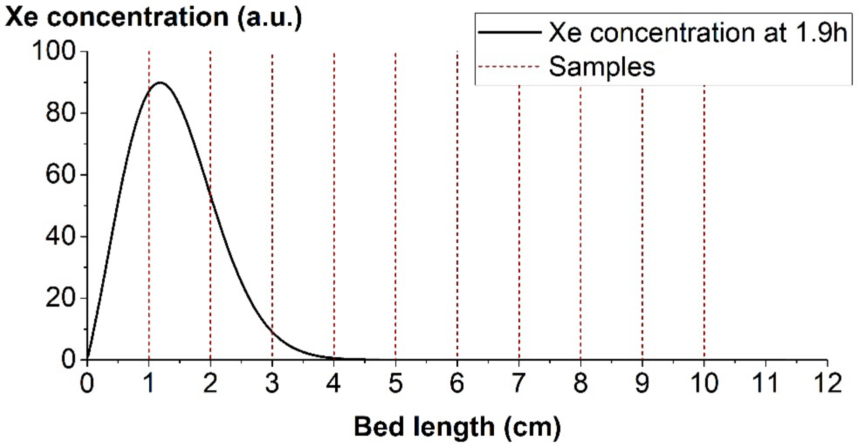
New in-situ irradiation of AgZs

Adsorption of ~ 50 TBq Xe-133 on ~ 30 g of both AgZs at IRE for 8 days

- Activity distribution estimation with COMSOL Multiphysics® (based on stable Xe experiments)
- Estimation of absorbed dose per 1 cm layer (as sampled after irradiation) by MC
  - Current estimate: 10 – 100 MGy
  - Tens – hundreds of hours of operation
- Characterization of the most irradiated sample
  - Xe adsorption at room temperature
  - SEM/EDX, PXRD, ^{27}Al- and ^{29}Si solid-state NMR and microporosity



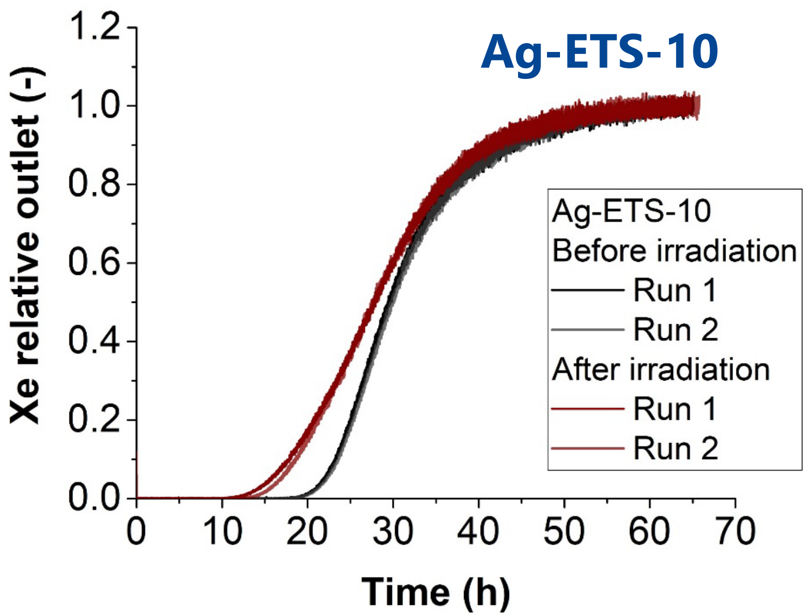


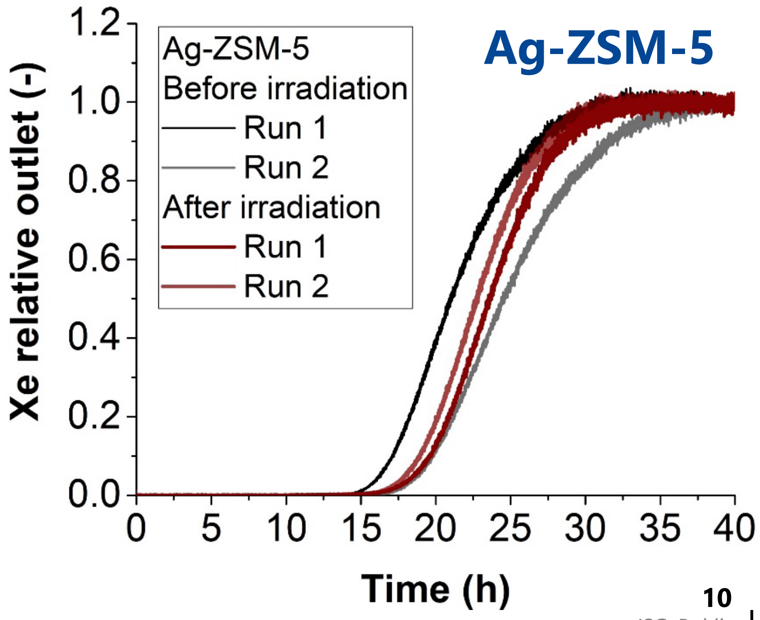


New in-situ irradiation of AgZs

- No significant degradation on the breakthrough of 10 ppm Xe in nitrogen (packing !)
- No significant differences observed by other characterizations, **EXCEPT** ^{29}Si NMR on Ag-ETS-10
  - Local changes in the Si environment in Ag-ETS-10

Characterization	Ag-ETS-10 Thermal cycles	Ag-ETS-10 Irradiation	Ag-ZSM-5 Thermal cycles	Ag-ZSM-5 Irradiation
SEM/EDX				
PXRD				
^{27}Al NMR	NA	NA		
^{29}Si NMR				
Microporosity				





Conclusions

1. Durability for thermal regeneration
   - No significant degradation observed
   - Packing of Ag-ETS-10 is important on the shape of the breakthrough
   - Variations in Xe adsorption on Ag-ZSM-5 likely from desorption duration

2. Radiation resistance
   - No significant degradation observed on Ag-ZSM-5
   - No significant degradation observed on Ag-ETS-10, **EXCEPT** on ^{29}Si NMR
     - Changes in the local environment of Si after irradiation

- Publication is being drafted

Perspectives

- Future potential work
  - Further characterizations of the irradiated samples (e.g. Ag oxidation states)
  - Further investigation on the ^{29}Si NMR result on Ag-ETS-10
  - Effect of impurities on the performances of AgZs (e.g. Cl-containing VOCs)
    - This would require a characterization of the gas stream to be treated at facilities

- New adsorbents in general could
  - Simplify mitigation systems (passive, less pre-conditioning needed, …)
  - Reduce the operation cost (room temperature, smaller systems, …)
  - Further reduce radioxenon emissions (equivalent but more efficient systems)

➡ **Ideally all three but in practice probably a trade-off between them**

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Contents lists available at ScienceDirect

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journal homepage: www.elsevier.com/locate/psep

Application of silver-exchanged zeolite for radioxenon mitigation at fission-based medical isotope production facilities

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Silver-exchanged zeolites for collecting and separating xenon directly from atmospheric air

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Thank you for your attention!