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/m³ 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
  - $P_{\text{xen}} < 1000 \text{ Pa}$
  - Xe in He
    - And in $N_2$ (also Ar)

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

Gueibe et al., 2022
Gueibe et al., 2023
What is their radiation resistance and thermal regeneration durability?
First exploration on Ag-ETS-10

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

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

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

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

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

Gueibe et al., 2022
New thermal durability investigation

- 44 cycles on Ag-ETS-10
  - Regeneration at ±210°C (+ test at 260°C) under N₂ (+ test with air)
  - Ads.: 0.087, 10 and 100 ppm Xe in air
  - No significant variations on \( q_{\text{Xe}} \) & \( t_{10\%} \)

- 43 cycles on Ag-ZSM-5
  - Regeneration at ±210°C (+ test at 260°C) under N₂ (+ test with air)
  - Ads.: 0.087 and 10 ppm Xe in air
  - Variations on \( q_{\text{Xe}} \) & \( t_{10\%} \) likely due to regeneration duration
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

<table>
<thead>
<tr>
<th>Characterization</th>
<th>Ag-ETS-10 Thermal cycles</th>
<th>Irradiation</th>
<th>Ag-ZSM-5 Thermal cycles</th>
<th>Irradiation</th>
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<td>SEM/EDX</td>
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<td>$^{27}$Al NMR</td>
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<td>$^{29}$Si NMR</td>
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<td>Microporosity</td>
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</table>

Graphs showing Xe relative outlet vs. time for Ag-ETS-10 and Ag-ZSM-5 before and after irradiation.
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
Thank you for your attention!