

Xe collection and purification from air in three types of porous materials

C. Gueibe^{1,2}, J. Rutten¹, J. Camps¹, N. Hermanspahn³, D. Moyaux⁴, W. Schroevers², D. Minta⁵ and S. Schreurs²



INTRODUCTION

More efficient and selective adsorbents for Xe collection and purification could provide new alternatives for noble gas monitoring in the IMS.

Silver-exchanged zeolites and metal-organic frameworks have never been investigated to collect and purify Xe directly from atmospheric air.

METHODS/DATA

Comparison of Ag-exchanged Zeolites (AgZs), Metal-Organic Frameworks (MOFs) and Activated Carbon (AC)

1. Characterization by SEM/EDX, PXRD, TGA & N₂/CO₂ adsorption
2. Xe and air breakthrough
3. Thermal desorption

START

RESULTS

- Highest ever reported Xe adsorption capacity in air on AgZs
- Unprecedented Xe/air selectivity on AgZs
- Decrease in Xe adsorption on AgZs in humid conditions
- Highest Xe concentration thermally recovered on AgZs

CONCLUSION

Ag-exchanged zeolites are currently the most efficient and selective adsorbents to collect and purify xenon from atmospheric air.

They could be used as a single filter to collect and purify xenon from dry atmospheric air, which could simplify and reduce the power consumption of IMS noble gas systems.



Part of this research was funded by the EU through Council Decision 2018/298/CFSP.

P3.2-803

Please do not use this space, a QR code will be automatically overlaid

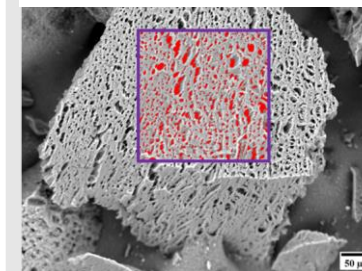
Introduction: porous adsorbents in IMS noble gas monitoring systems

Noble gas monitoring systems are a crucial component of the International Monitoring System (IMS) for the verification of the CTBT. They are monitoring the atmosphere for potential **radioxenon** releases originating from nuclear tests. The **efficient collection and purification of trace levels of xenon in air (i.e. 87 ppb)** on porous adsorbents is essential for their detection capability.

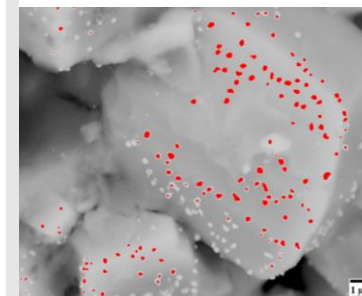
The first systems in the IMS used pre-purification techniques to remove moisture and CO₂ followed by AC columns to collect and further purify Xe. In some new systems, AgZs have replaced some of the AC columns, after the necessary pre-purification, due to their much higher Xe adsorption capacity at room temperature. The current systems require a **complex and energy demanding purification process**. More efficient and selective adsorbents could simplify the systems and reduce their power consumption. For instance, recent studies on a new class of porous materials, namely MOFs, have demonstrated high Xe selectivity over other gas components although in conditions different than for IMS applications.

No literature has been published about the investigation of AgZs or MOFs for the **collection and purification of Xe directly**, i.e. without pre-purification, **from atmospheric air**. Such a direct Xe collection and purification process could significantly **simplify the systems** and **reduce their power consumption**.

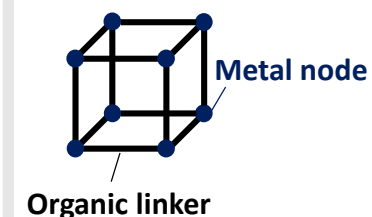
Activated carbon [1]



Ag-Zeolite [1]



Metal-Organic Framework

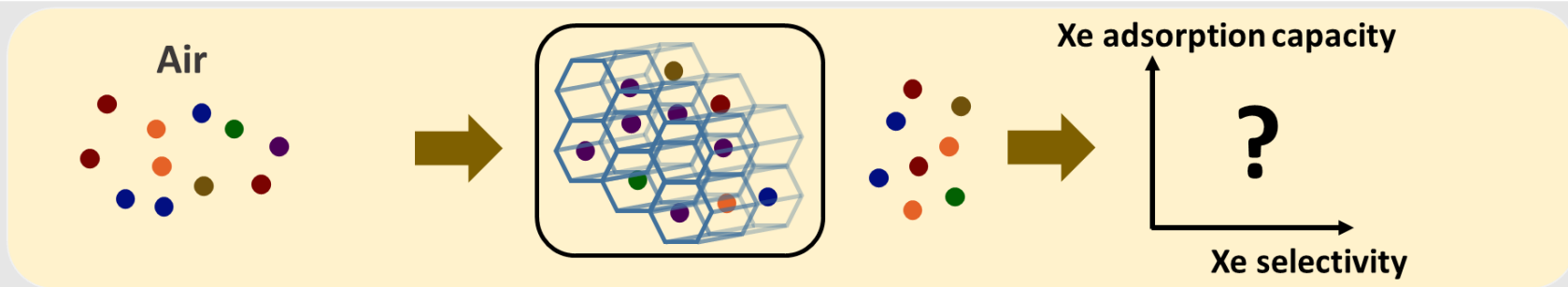


Objectives

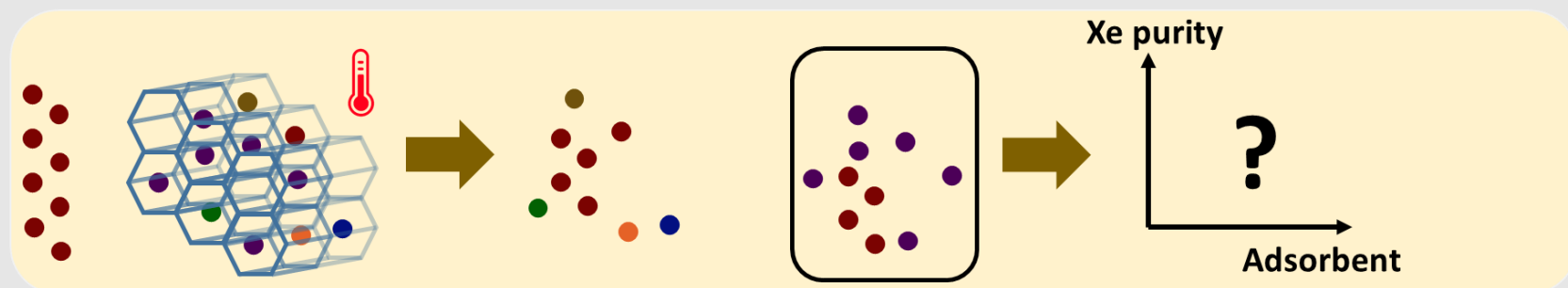
First time investigation into the use of AgZs and MOFs for the **collection and purification of Xe directly from atmospheric air** to potentially simplify or reduce the power consumption of IMS noble gas monitoring systems. The aim is to answer the following questions:

1. Are the adsorbents, in pelletized or granulated form, acquired in this work in agreement with properties reported in the literature for the same adsorbents ?
2. How efficient and selective are MOFs and AgZs, compared to AC, in collecting Xe from atmospheric air ?
3. How easy and in which purity can we thermally recover the collected Xe from the adsorbents ?

②
Xenon
collection



③
Xenon
purification



N₂
O₂
Ar
CO₂
Kr
Xe

Selection of adsorbents based on commercial availability

- a) Silver-exchanged zeolites (AgZs): Ag-ETS-10 and Ag-ZSM-5
- b) Activated Carbon (AC): Nusorb® GXK
- c) Metal-Organic Frameworks (MOFs): HKUST-1 and Ni-DOBDC

1. General characterizations of the acquired samples

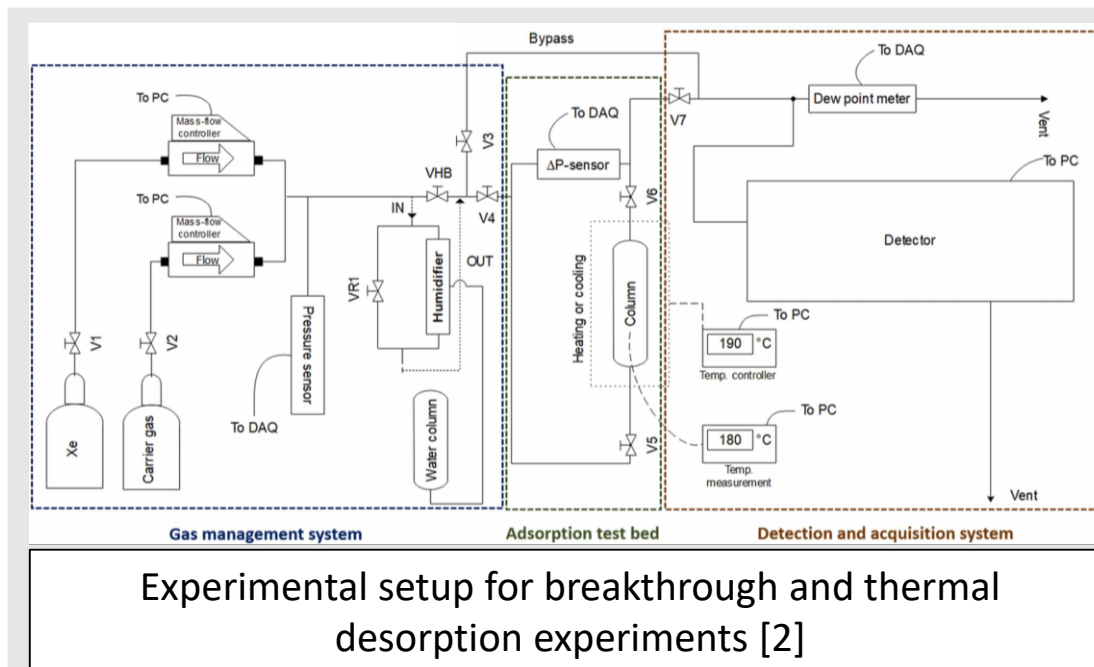
- Morphology and composition by SEM/EDX, crystallinity by PXRD and thermal stability by TGA-MS
- Microporosity by N₂ adsorption isotherm at 77 K and CO₂ adsorption isotherm at 273 K

2. Investigation of Xe collection at room temperature

- 250 ppm Xe in N₂ breakthrough in dry and humid (5% and 50% R.H.) conditions
- 100 ppb Xe in N₂ & air breakthrough

3. Investigation of Xe purification

- Thermal desorption under N₂ after air adsorption (without Xe breakthrough)
- Thermal desorption under N₂ after adsorption of 250 ppm Xe in nitrogen spiked with Rn-222



Results: xenon collection from air

- Figure 1: Decrease in Xe adsorption capacity on AgZs by a factor 30 in 50% R.H. compared to dry conditions
- Figure 2: Significantly higher Xe adsorption capacity (at 100 ppb Xe in N₂) on both AgZs (■) compared to literature data
 - Similar capacity in air (not shown here)
- Figure 3: Significantly higher Xe/N₂ selectivity in air on both AgZs (■) compared to literature data

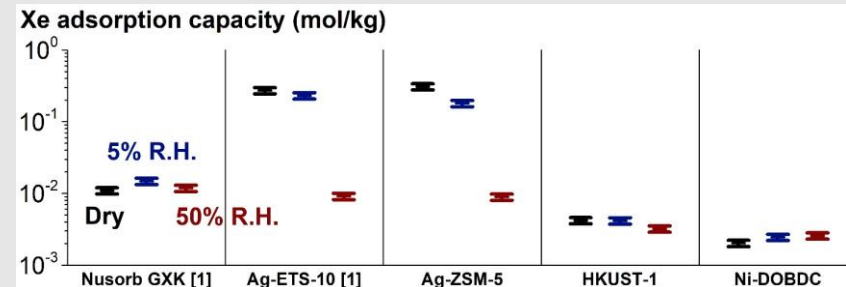


Figure 1 – Xe adsorption capacity as a function of relative humidity on the five adsorbents.

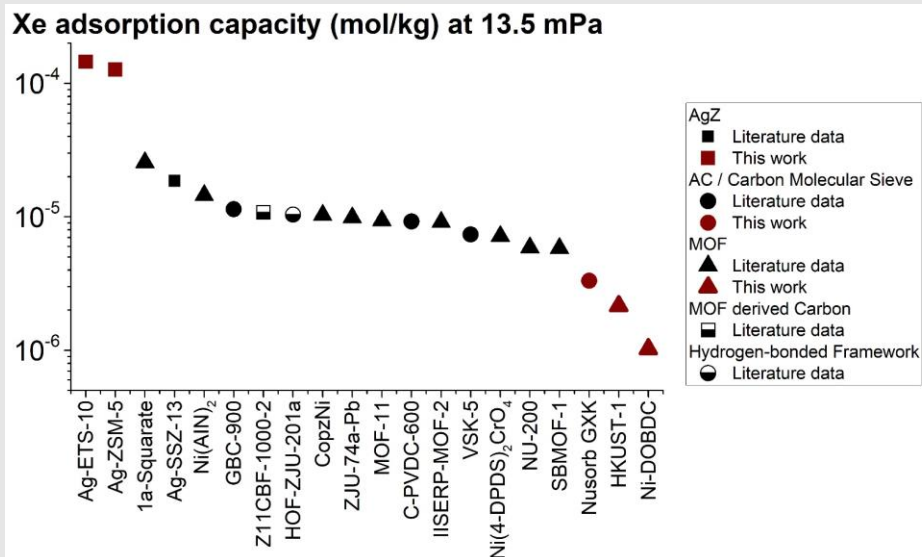


Figure 2 – Xe adsorption capacity (100 ppb Xe in N₂) on the adsorbents in this work compared to the literature.

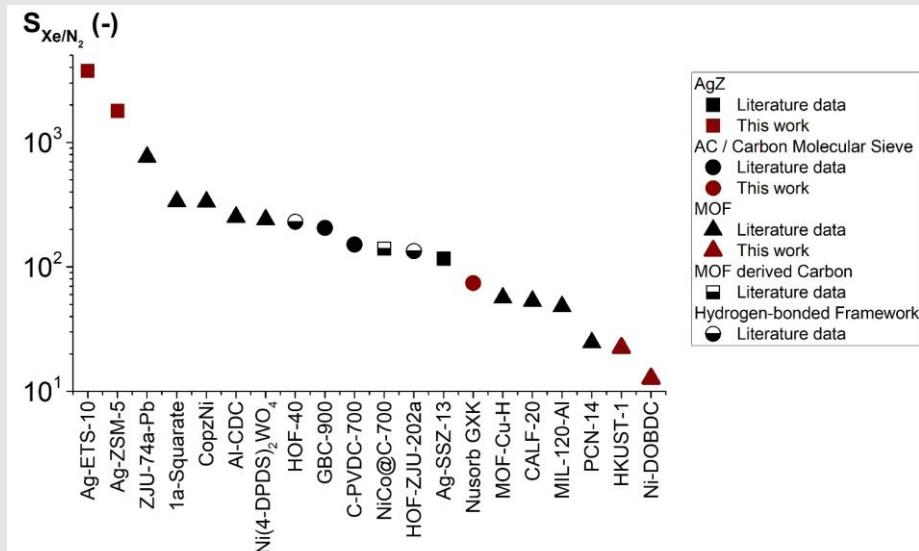


Figure 3 – Xe/N₂ selectivity in air on the adsorbents in this work compared to the literature.

Results: xenon purification from dry air

- Figure 4: Significantly higher Xe concentration (◆) after a single thermal desorption cycle with 13 vol. % Xe in the gas recovered on Ag-ETS-10
- Figure 5: Impressive Xe/Rn thermal separation in both AgZs BUT it requires higher temperatures than MOFs/AC

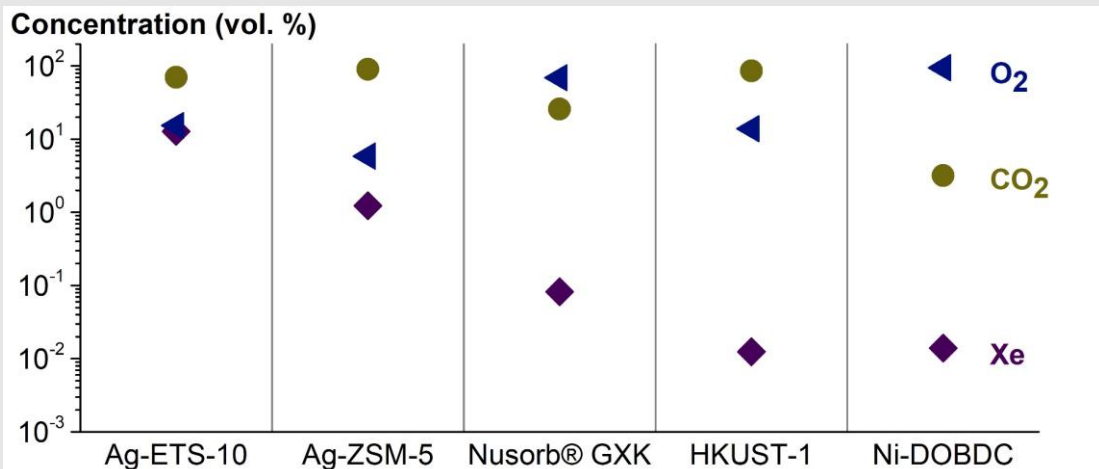


Figure 4 – Comparison of the Xe, O₂ and CO₂ concentration in the recovered gas (containing 90% of the collected Xe in air) after a single thermal desorption cycle.

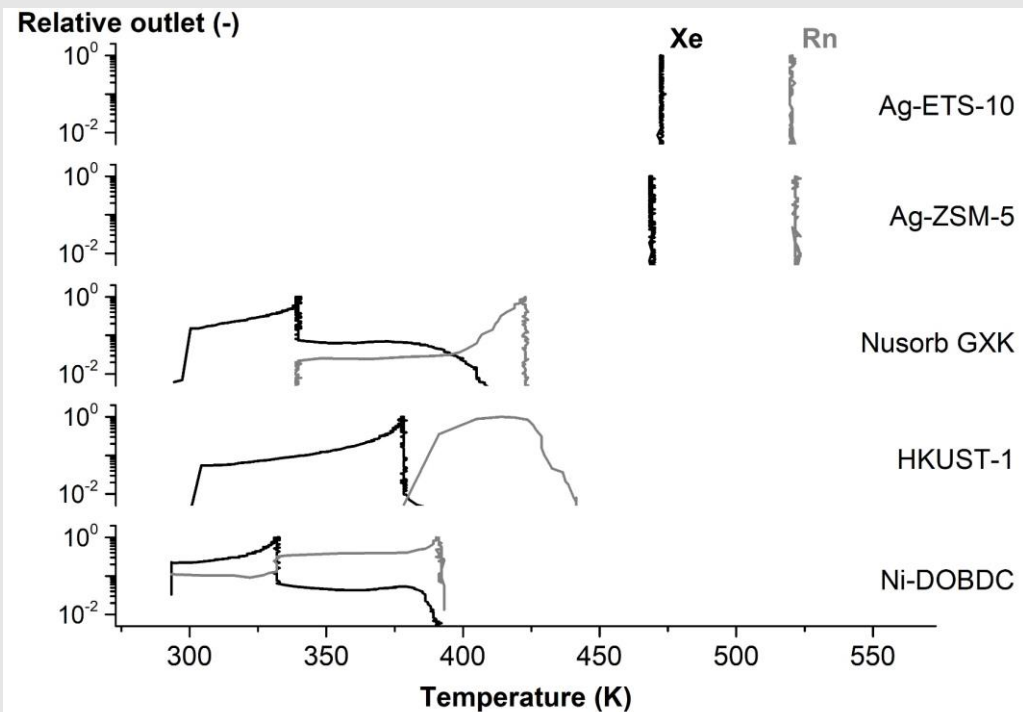


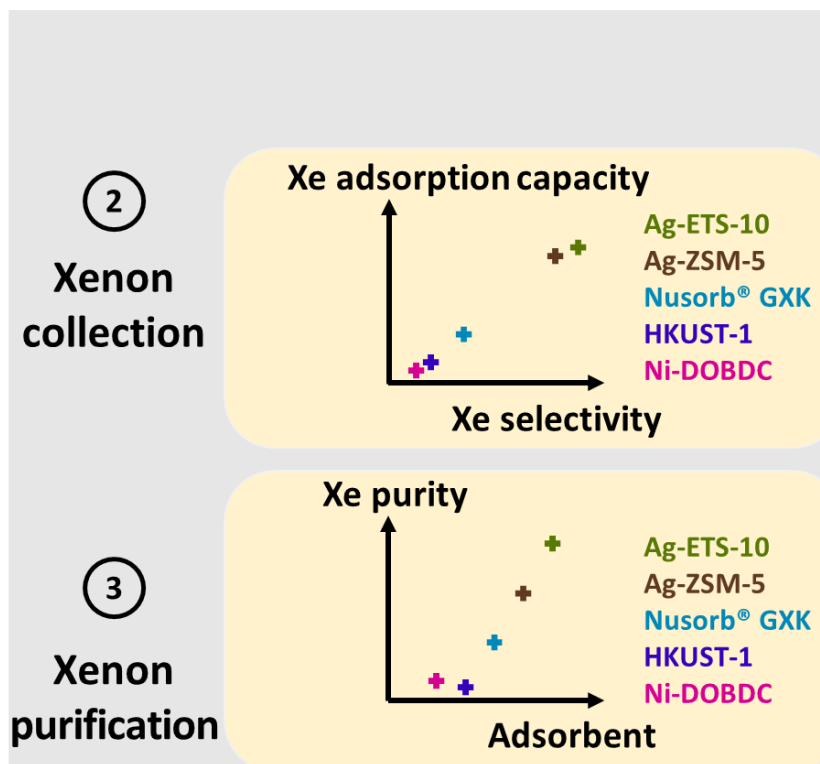
Figure 5 – Xe/Rn separation during a thermal desorption on all five adsorbents. The AgZs have a very sharp desorption profile with regard to temperature.

1. The morphology, composition, crystallinity, thermal stability and microporosity of the adsorbents acquired in this work are in very good agreement with reported data.
2. Xe adsorption & selectivity in air: values on AgZs are significantly higher than literature data
 - a) **BUT** in humid conditions, the Xe adsorption capacity decreases significantly on AgZs !
3. The thermally recovered Xe (90% yield) from dry air has the highest Xe concentration using AgZs
 - a) AgZs are very efficient for Xe/Rn separation
 - b) **BUT** more energy required than for MOFs/AC

➔ **AgZs are promising as single filter in dry air**

Future work

- Further investigate the purity of the recovered Xe gas over multiple cycles
- Investigate the durability of the adsorbents
- Investigate other, currently non-commercial, promising adsorbents



References

[1] C. Gueibe, J. Rutten, J. Camps, D. Moyaux, W. Schroeyers, M. Auer, S. Schreurs. Application of silver-exchanged zeolite for radioxenon mitigation at fission-based medical isotope production facilities. *Process Saf Environ*, 158 (2022), 576-588. <https://doi.org/10.1016/j.psep.2021.12.031>



[2] C. Gueibe, J. Rutten, J. Camps, D. Moyaux, W. Schroeyers, R. Plenteda, N. Hermanspahn, M. Daria, S. Schreurs. Xenon Collection and Separation from Atmospheric Air in Silver-Exchanged Zeolites, Activated Carbon and Metal-Organic Frameworks. Preprint available at SSRN. <https://dx.doi.org/10.2139/ssrn.4442283>



References used in figures 2 & 3

- Li et al., *Micropor Mesopor Mat*, 330 (2022) 111631.
- Li et al., *J Am Chem Soc*, 141 (2019) 9358-9364.
- Chakraborty et al., *Chem-Eur J*, 26 (2020) 12544-12548.
- Gong et al., *J Mater Chem A*, 6 (2018) 13696-13704.
- Guo et al., *Ind Eng Chem Res*, 61 (2022) 7361-7369
- Kang et al., *J Mater Chem A*, 10 (2022) 24824-24830.
- Liu et al., *Angew Chem Int Edit*, 61 (2022) e202117609.
- Zhao et al., *Sep Purif Technol*, 302 (2022) 122074.
- Liu et al., *Chem Eng J*, 453 (2023) 139849.
- Pei et al., *J Am Chem Soc*, 144 (2022) 3200-3209.
- Chen et al., *Sci China Chem*, 66 (2022) 601–610.
- Magomedbekov et al., *J Chem Eng Data*, 68 (2022) 282–290.
- Zheng et al., *Angew Chem Int Edit*, 61 (2022) e202116686.
- Yu et al., *J Mater Chem A*, 6 (2018) 11797-11803.
- Gong et al., *J Am Chem Soc*, 144 (2022) 3737-3745.
- Zhu et al., *Sep Purif Technol*, 274 (2021) 119132.
- Gong et al., *Nano Res*, 15 (2022) 7559-7564.
- Chen et al., *Angew Chem Int Edit*, 60 (2021) 2431-2438.
- Xiong et al., *J Mater Chem A*, 6 (2018) 4752-4758.
- Wei et al., *Rsc Adv*, 12 (2022) 18224-18231.
- Zhang et al., *J Chem Eng Data*, 65 (2020) 4018-4023.
- Perry et al., *J Phys Chem C*, 118 (2014) 11685-11698.



INTRODUCTION

OBJECTIVES

METHODS/DATA

RESULTS

CONCLUSION



Please do not use this space, a QR code will be automatically overlaid

P3.2-803