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Metal Organic Frameworks: A new class of scintillators for radioactive gas detection

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Abstract— The detection of radioactive gases, ⁸⁵Kr, ²²²Rn, and ³H, represents a significant challenge in nuclear detection due to their inherent volatile nature. No convenient one-size-fits-all detection system exists for the online measurement of these three gases. We hence propose a new class of scintillators with high surface areas, which optimizes the interaction between the radioactive gas and the scintillator. Metal Organic Frameworks (MOF) are ideal porous candidates for this application. They are the material with the highest known surface areas, up to 8000 m²g⁻¹, and have been studied for their fluorescent and scintillating properties [1]. To the best of our knowledge, the use of MOFs as scintillating sponges to detect radioactive gases has only been theorized. We were able to experimentally demonstrate, using advanced coincidence techniques (TDCR) and a homemade metrological test bench [2], that MOFs are indeed scintillating in the presence of all three targeted gases.

Keywords—MOF, scintillator, detection, radioactive gas, porous

I. INTRODUCTION

Since the discovery of radioactivity in 1896, the detection of radioactive sources has become vital for radioprotection. Radioactive gases represent an even harder challenge due to their volatile nature. The three radioactive gases targeted here are ⁸⁵Kr, ²²²Rn and ³H due to their impact on our society. The detection of ⁸⁵Kr is of an utmost importance for the radioprotection of the workers of nuclear power plants and reprocessing plants and also as sign of the core's structural health. As for ²²²Rn, it is the second leading cause of lung cancer following tobacco. ³H detection is not only an indicator of nuclear military activity, but also, civil activities especially with its predicted increasing use as primary fuel for fusion reactors (ITER).

Proportional gas counters, diffusion chambers and liquid scintillation are all techniques used to detect one or two of these gases. However, those techniques have a few major disadvantages. Even though, proportional gas counters allow for online detection, the setup can be quite bulky, as it requires a pressurised gas bottle. Liquid scintillation only allows for remote detection. This inadequate pool of detectors show that there exists a true demand for a one-size-fits-all online detection system for radioactive gases.

We propose here using porous scintillators as a new class of scintillator. The idea is to mimic liquid scintillation where the radionuclide and the scintillating component are in direct contact with one another. A porous material will allow for a more effective intersection between the scintillator and the gas. The porous scintillators explored here are Metal Organic

Frameworks (MOF). To the best of our knowledge, the use of MOFs as scintillating porous architectures to detect radioactive gases has so far only been theorised.

II. MATERIAL

Metal Organic Frameworks are hybrid crystalline Inorganic-Organic materials made up of inorganic metal clusters linked together by organic ligands via strong ionic-covalent interactions giving rise to a cage-like structure.

MOFs are ideal candidates for radioactive gas detection due to the following outstanding properties:

A. Porosity and high specific surface area

The cage like structure of MOFs lead to porous sites ideal for gas adsorption. Due to these pores, MOFs are the materials with the highest known specific surface areas, up to 8000m²g⁻¹ [4].

B. Fluorescence and scintillating properties

Fluorescent MOFs have been studied in literature. Different fluorescence and scintillating mechanisms exist in a MOF: ligand-based, metal-based, ligand-to-ligand charge transfer, metal-to-metal charge transfer, ligand-to-metal charge transfer and metal-to-ligand charge transfer [5]. In 2009, [1] first used MOFs for the detection of ionizing radiation.

C. Hydrogen Storage

Out of our targeted gases, ³H is the trickiest to detect due to extremely low mean energy of its β particles (5.7 keV). MOFs have been vastly studied throughout the years for the storage of hydrogen, which is chemically identical to ³H [6].

D. Structural diversity

By changing the identity of the metal centre and/or ligand, one can obtain an almost infinite variety of MOFs with different physical properties.

We explored the synthesis of a wide array of MOFs mainly those already known in literature. The aim however, is to produce millimetric single crystals of MOFs to ensure the purity of our samples and hence scintillation mechanism. We target MOFs with an emission centered around 280-420 nm since the photomultiplier tubes (PMT) used in our detection system in Fig. 2 have a maximum quantum efficiency in that wavelength range.

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III. METHOD

The MOFs synthesised were characterized structurally (X-Ray Diffraction, porosity and surface area measurements, thermogravimetric analysis) and photophysically (photoluminescence excitation and emission, and time correlated single photon counting for the half-life of emission).

Promising candidates were then test on a unique homemade metrological gas bench test shown in Fig 2 [2].

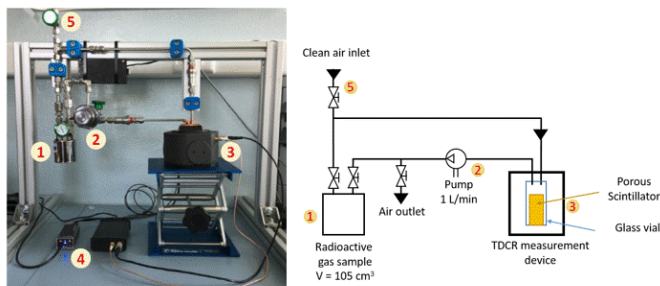


Fig. 1: Illustrating the homemade metrological test bench.

A radioactive atmosphere of either ^{222}Rn , ^{85}Kr or ^3H stored in vial **1** is circulated throughout the closed circuit until it reaches the MOF sample in the Triple-to-Double coincidence ratio (TDCR) measurement device labeled **3**. The TDCR counter is a precise metrological device, which allows to record the logical sum of the double coincidences (D) as well as the triple coincidences (T) of photons emitted by the MOF between the three PMTs mentioned above. The T/D ratio gives the efficiency of the scintillation yield of the MOF. After the gas is circulated in the system, another set of measurements are recorded while the system is being rinsed with dry air. This allows us to monitor whether the MOF releases the radioactive gas from its pores and hence is a reusable scintillator.

IV. RESULTS AND DISCUSSION

A. ^{85}Kr

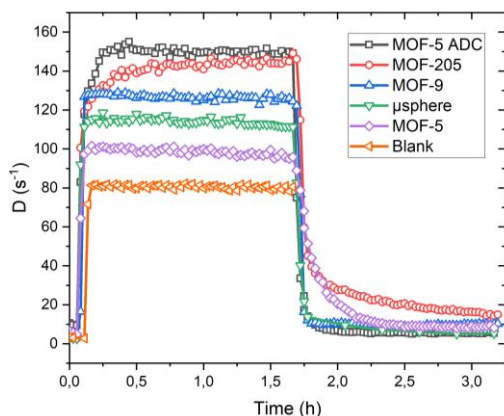


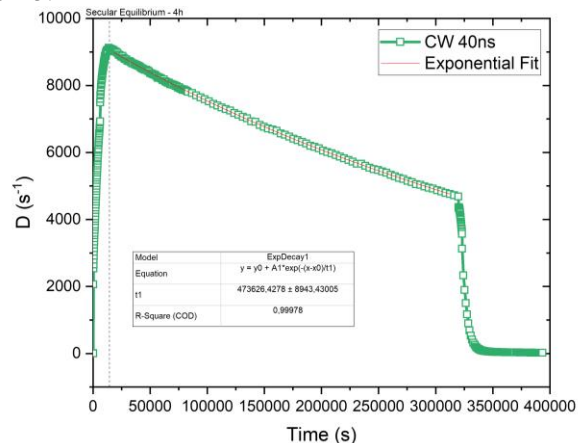
Fig. 2: Results of 10kBq of ^{85}Kr detection by various MOFs.

Fig 3. demonstrates that the photon count rate for our various MOFs is higher than the blank measurement (system without MOFs, only Cherenkov light produced), proving that the MOFs are indeed scintillating in the presence of 10kBq ^{85}Kr .

We were also able to show with further experiments using MOF-205, that reproducible measurements were obtained with the same activity and that increasing the injected ^{85}Kr activity linearly increases the average photon count plateau.

B. ^{222}Rn

Not only where we able to detect ^{222}Rn with MOF-205, but we were able to experimentally measure the half-life of ^{222}Rn at 3.7997 days with an exponential fit, the official value being 3.8215.



V. CONCLUSION

To conclude, we were able to show for the first time that MOFs are potent scintillators for radioactive gas detection. We were also able to compare the scintillating performance between different MOFs. This work is also the first proof of concept of the use of MOFs for ^{222}Rn detection and half-life measurement of a radionuclide.

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