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► **To cite this version:**

Yannis Cheref, Raphaël Marie-Luce, Pavlo Mai, Frédéric Lerouge, Sylvie Pierre, et al.. Preparation of nanoparticle-based aerogels for real-time radioactive gas detection. Summer School, 17th International Conference on Scintillating Materials and their Applications (SCINT2024), Jul 2024, Milan (Italie), Italy. . <hal-04863366>

HAL Id: hal-04863366

<https://hal.science/hal-04863366v1>

Submitted on 31 Jan 2025

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Preparation of nanoparticle-based aerogels for real-time radioactive gas detection

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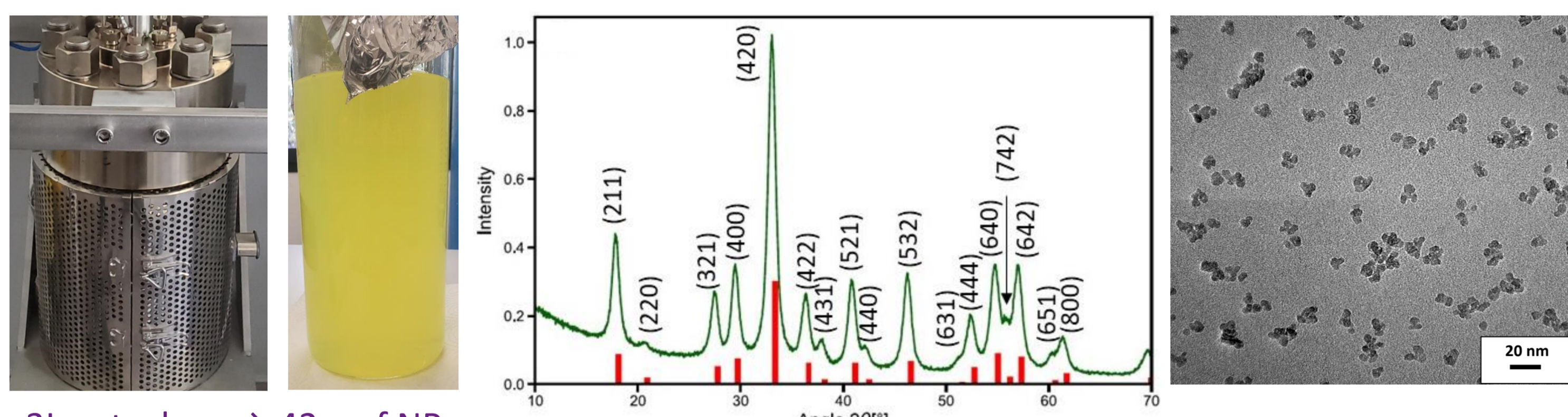
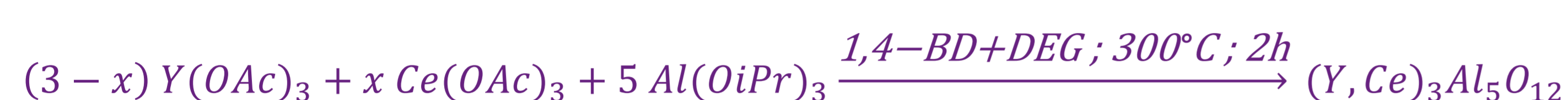
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The measurement of pure β emitting radioactive gases such as ^3H and ^{85}Kr is of major importance for the nuclear safety authorities, and will meet an increasing demand with the expansion of nuclear-based energy production. Due to the short penetration length of β electrons in air, gaseous β emitters must be mixed with radiosensitive elements to enable detection. These are either gas-gas mixtures in an ionization chamber, or gas-liquid mixtures in liquid scintillation. However, none of these existing methods combine real-time analysis, sensitivity to multiple gases, and ease of deployment for on-site measurements.

We demonstrate a new gas-solid mixture approach using inorganic aerogels as nanoporous scintillators. This method combines the above criteria, allowing efficient real-time measurement of ^3H and ^{85}Kr activity. Here, we focus on the preparation of the scintillation aerogels, from 10g-scale nanoparticle synthesis to supercritical drying.

1. Solvothermal synthesis of Ce:YAG nanoparticles



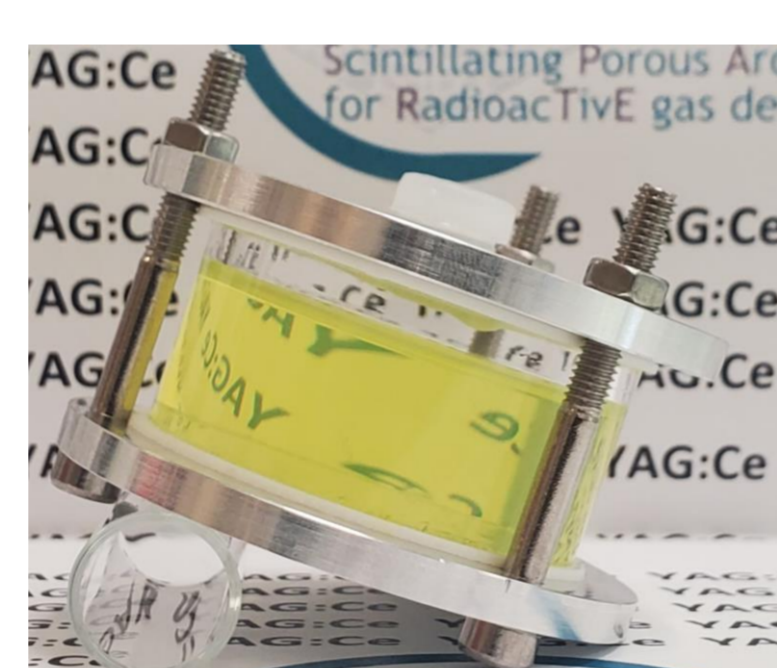
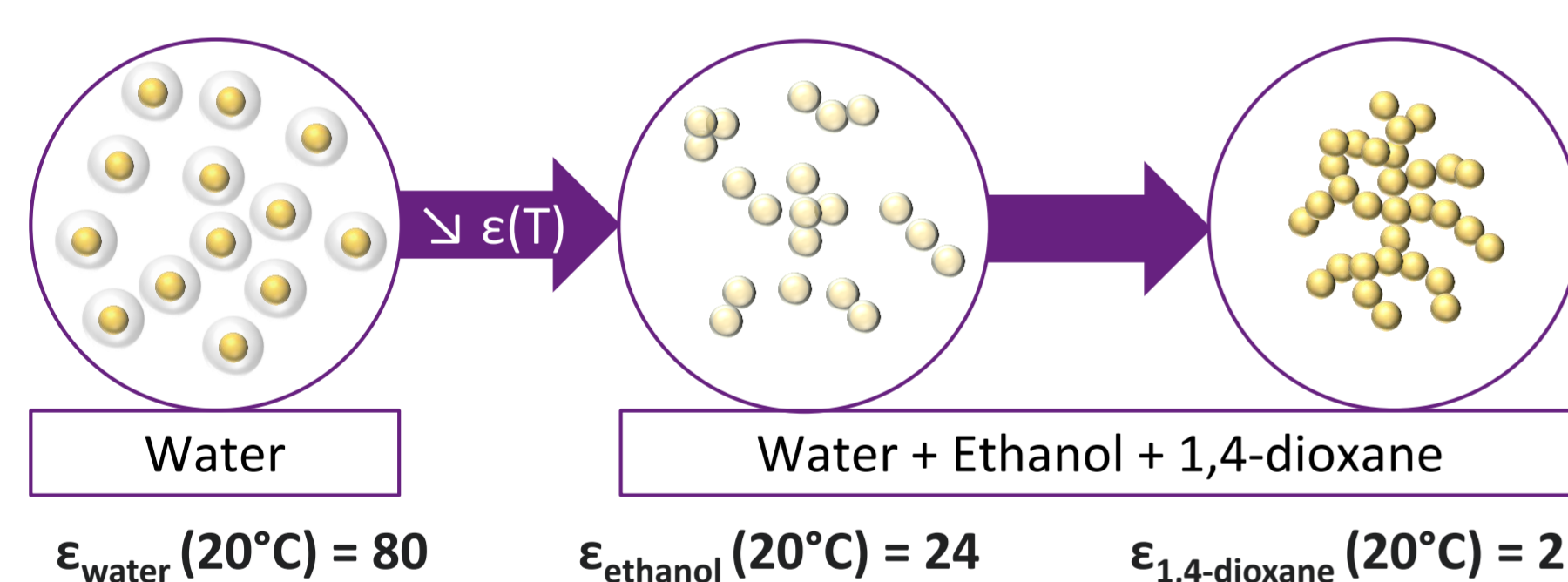
3L autoclave \rightarrow 42 g of NP

$\text{Y}(\text{OAc})_3$, $\text{Ce}(\text{OAc})_3$ and $\text{Al}(\text{OiPr})_3$ are dried, added to a 17:3 mixture of 1,4-butanediene (1,4-BD) and diethylene glycol (DEG), and mixed into a homogeneous slurry.

The slurry is heated to 300°C for 2h in an autoclave, producing a yellow transparent colloidal solution of highly crystalline Ce:YAG nanoparticles. [1]

3. Gelification and supercritical drying

Addition of **low-permittivity solvents** into the concentrated colloid causes the **reduction of the Debye radii** of the nanoparticles, causing their aggregation. [2]



The solvent mixture inside the gel is replaced with acetone before **supercritical drying in CO_2** .



The obtained aerogels are **annealed in air**. This step converts the Ce^{3+} into Ce^{4+} , which are good scintillation centers. It also results in the **contraction and strengthening** of the material.

Conclusion

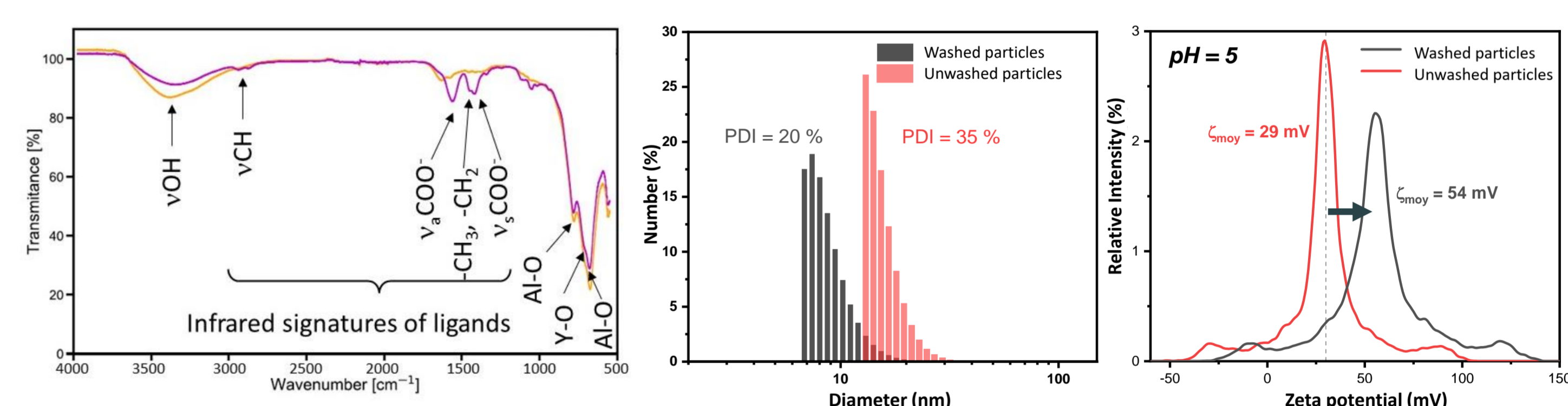
- Transparent aerogels were prepared by **aggregation and supercritical drying of a concentrated colloid** of scintillating Ce:YAG nanoparticles.
- Real-time activity measurements were performed on known gas atmospheres, showing a **sensitive and efficient detection of pure β emitting radioactive gas**.
- The **same procedure** was adapted to higher doping ratios and to different scintillating materials, allowing further optimization.

2. Nanoparticle purification

Centrifugation \rightarrow Acid wash \rightarrow Centrifugation \rightarrow Dialysis

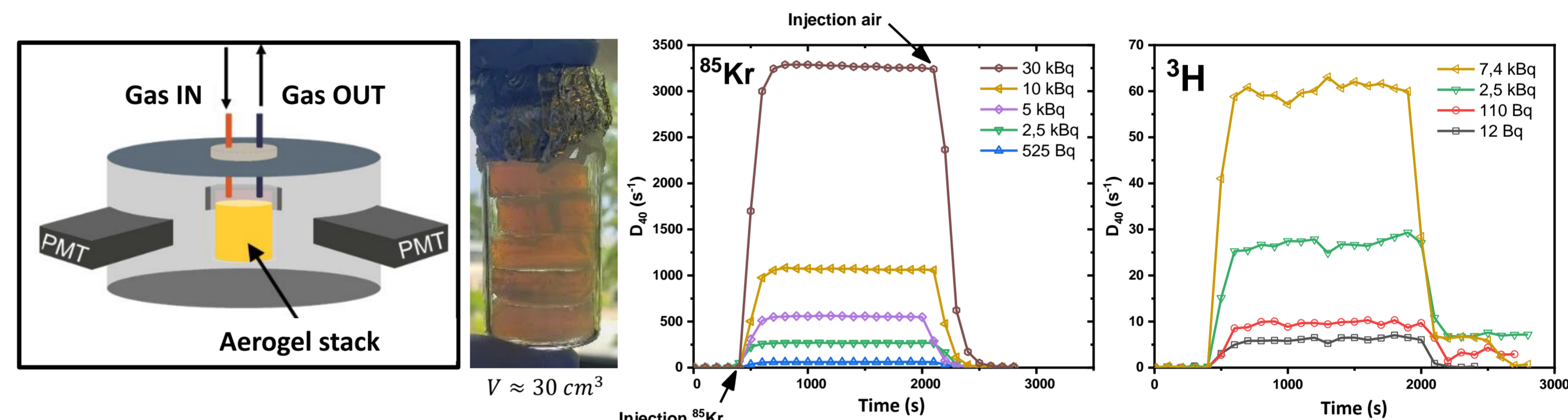
The nanoparticles are extracted from the organic solvent mixture by centrifugation and dispersed into water. The native surface ligands are then removed by a wash in HCl solution, yielding an **electrostatically-stable colloid**, which is then further washed by centrifugation, neutralized by dialysis, and concentrated by rotary evaporation to **40 %_{w/w}**.

The effects of the purification are measured in FT-IR, DLS and zetametry.



4. Radioactive gas activity measurement

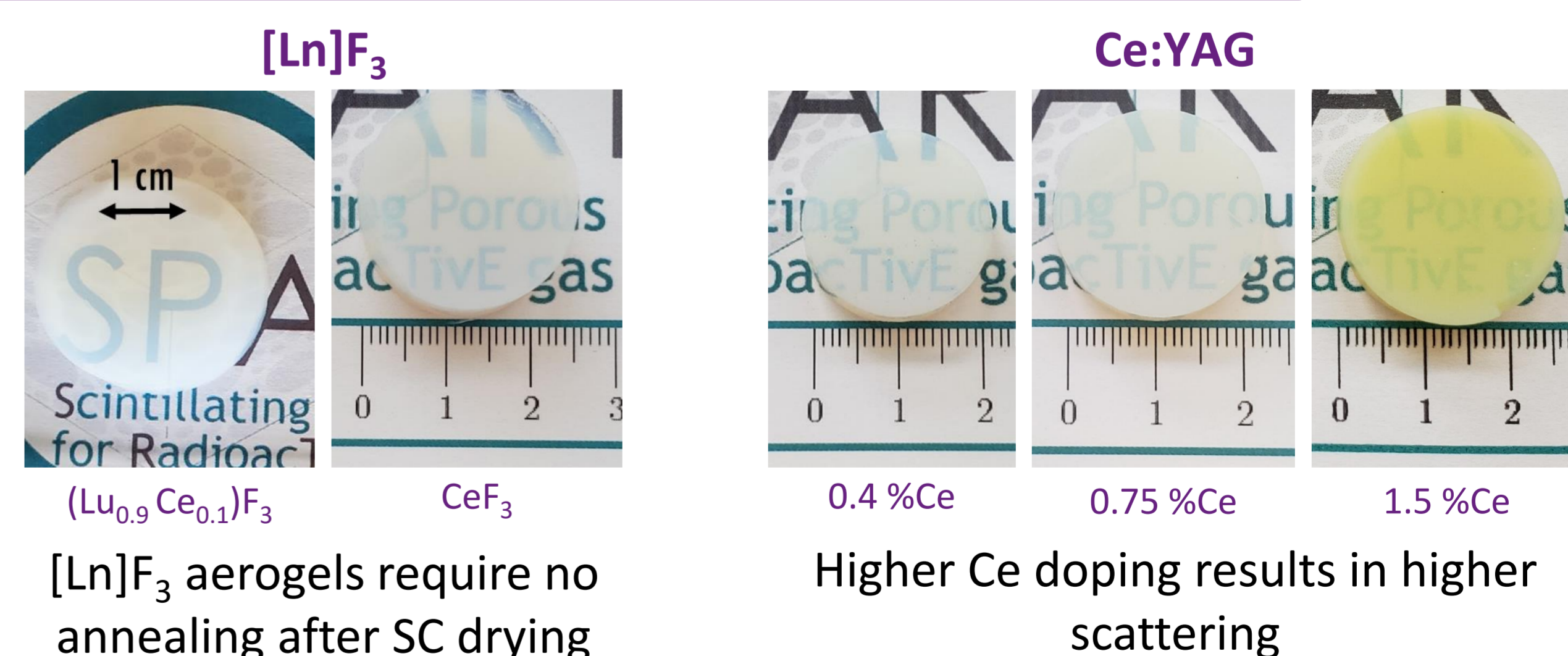
The scintillation of the aerogels in the presence of a known activity of gas is measured in **real time**, in double and triple coincidence, in 100s steps. [2,3]



For a stack of Ce:YAG aerogels with a doping ratio of 0.4 %:

- Detection threshold: **0.1 Bq/cm³**
- Efficiency for ^{85}Kr : **100 %**
- Efficiency for ^3H : **18,1 %**

5. Optimization and adaptation of the method



References

- [1] Odziomek, M., Chaput, F., Lerouge, F., Sitarz, M., Parola, S. Highly luminescent YAG:Ce ultra-small nanocrystals, from stable dispersions to thin films. *J. Mater. Chem. C* 5, 12561–12570 (2017).
- [2] Marie-Luce R., Mai P., Lerouge F., Cheref Y., Pierre S., Sabot B., Chaput F. & Dujardin C. Real-time detection and discrimination of radioactive gas mixtures using nanoporous inorganic scintillators. *Under Review* (2024)
- [3] Sabot, B., Rodrigues, M., Pierre, S. Experimental facility for the production of reference atmosphere of radioactive gases (Rn, Xe, Kr, and H isotopes). *Applied Radiation and Isotopes* 155, 108934 (2020).