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Study of the alteration mechanisms of (U,Th)O₂ and (U,Pu)O₂ mixed oxides under storage conditions

Aurélien Perrot, Sandrine Miro, Laurent Claparede, Aurélien Canizares, Lola Sarrasin, Hélène Arena, Christophe Jegou, Nicolas Dacheux

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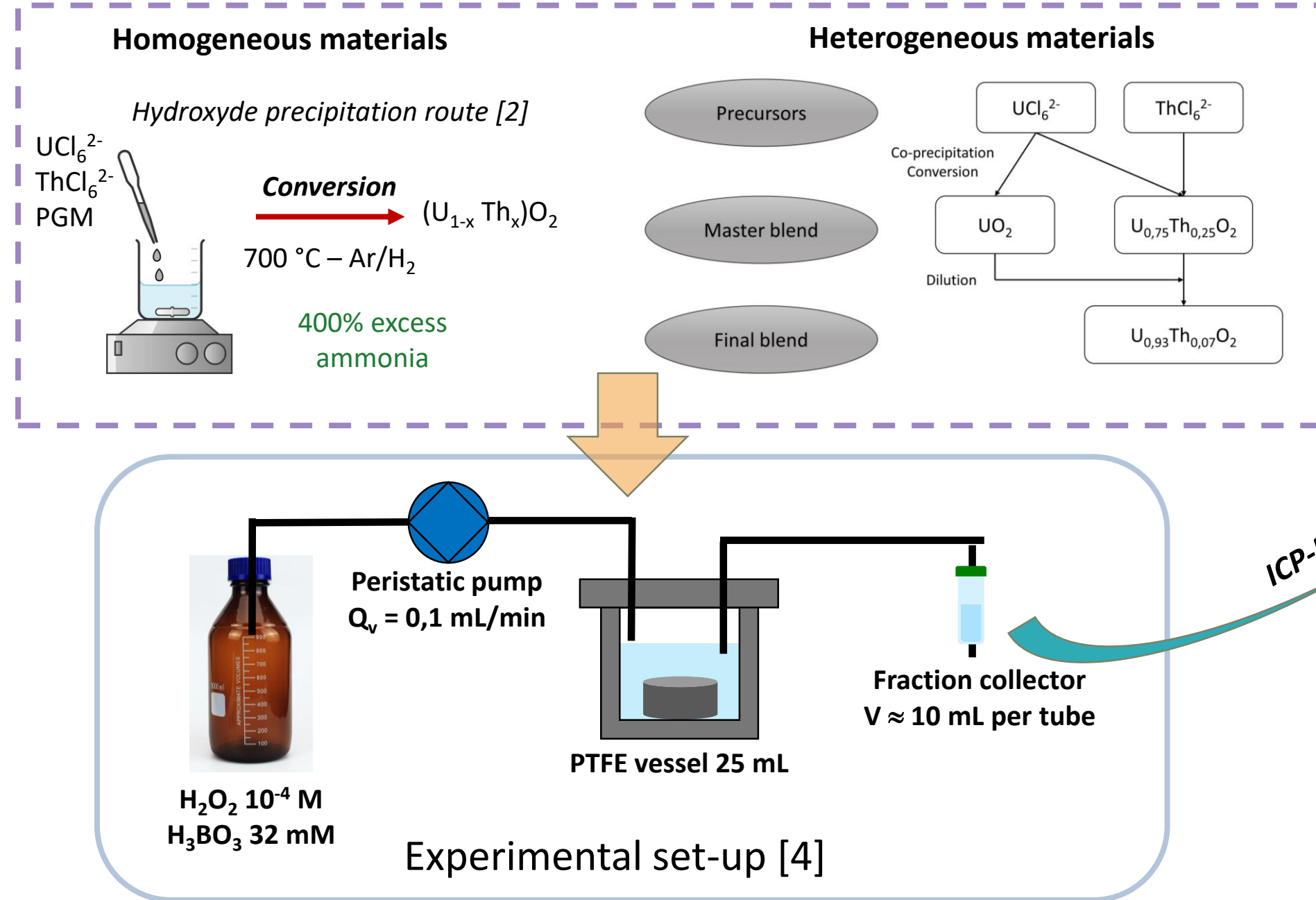
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INTRODUCTION

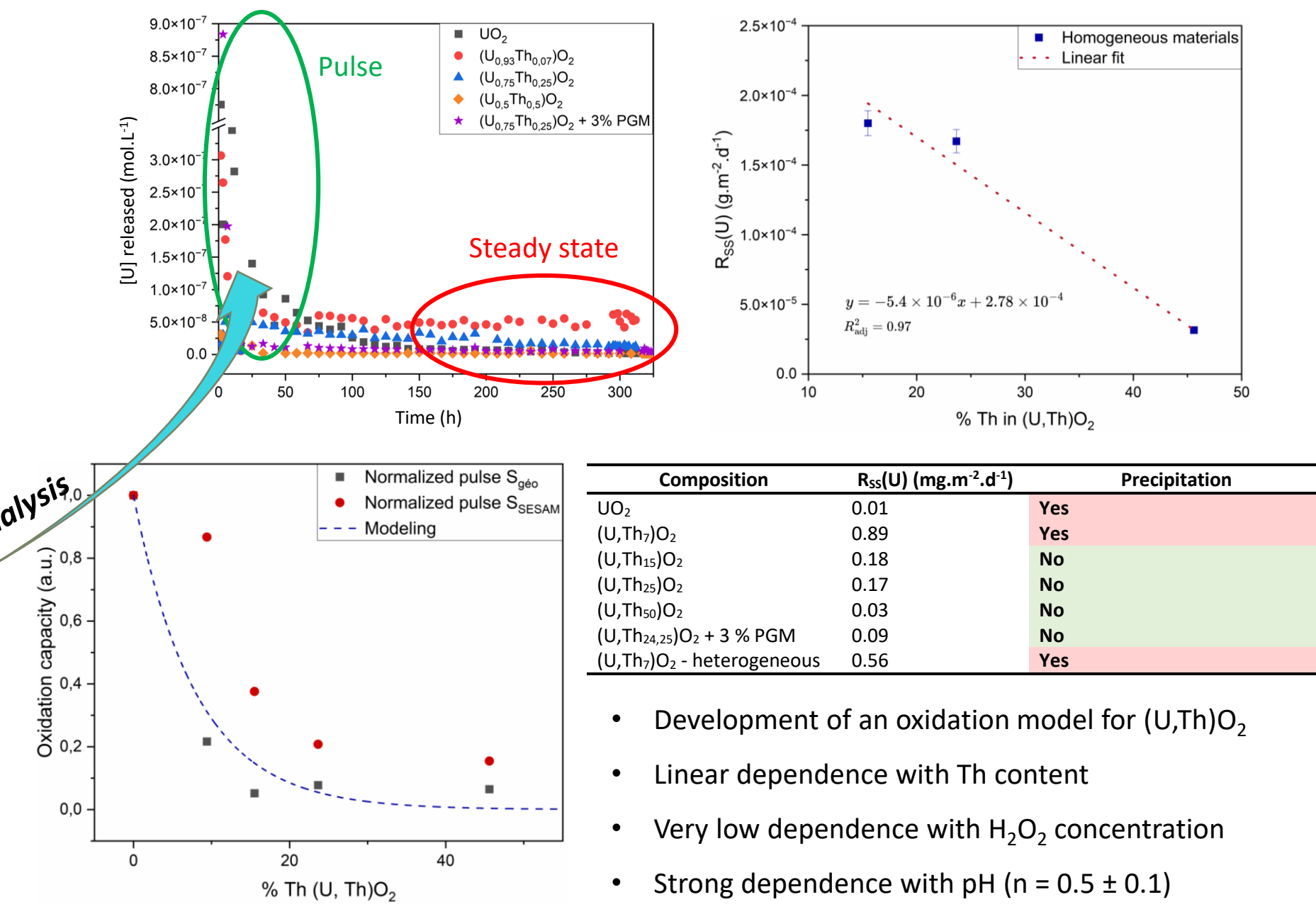
In France, the single recycling of irradiated fuel leads to the medium-term storage of irradiated **Mimas[®] MOx** (Mixed Oxide) fuel assemblies in pools, with a view to recycling or final disposal. During this **underwater storage**, attention is given to the potential impact of a **defect in the cladding**. Given that the pool water is oxidizing due to the presence of **radiolytic products** such as H₂O₂ and OH^{*}, leaching by this water promotes the **oxidative dissolution** of uranium, followed by its reprecipitation as **studtite** [(UO₂)(O₂)(H₂O)₂].₂(H₂O). This research is focused on the degradation mechanism of the fuel under oxidative environment and is useful in the context of evaluating the long-term radiological consequences for situations involving defective rods.

MULTIPARAMETRIC STUDY OF (U,Th)O₂ MATERIALS

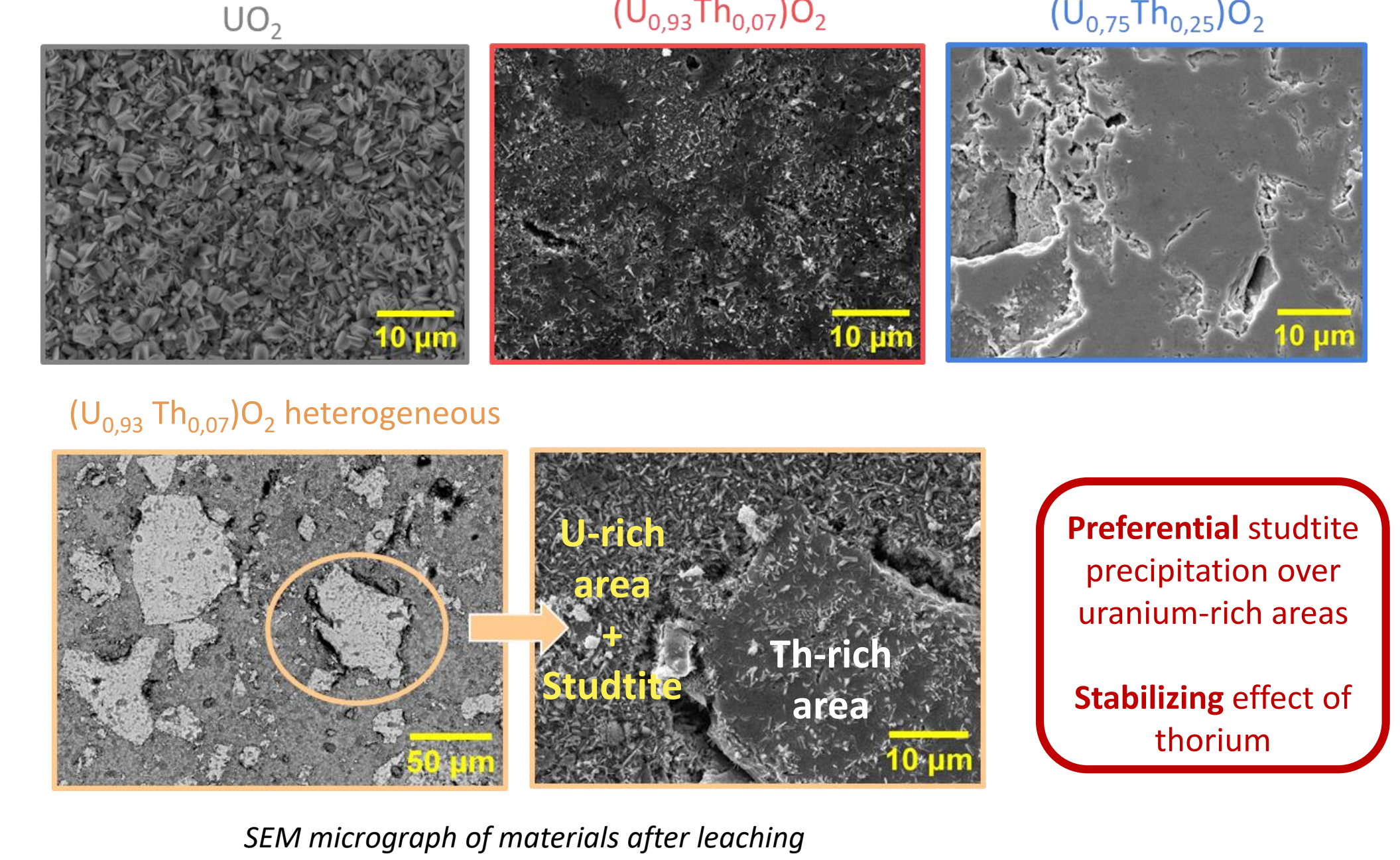
Synthesis and experimental set-up



Dynamic leaching experiment



Post-mortem characterization



STATIC MODE LEACHING OF MIMAS MOx FUEL

Optical analyses

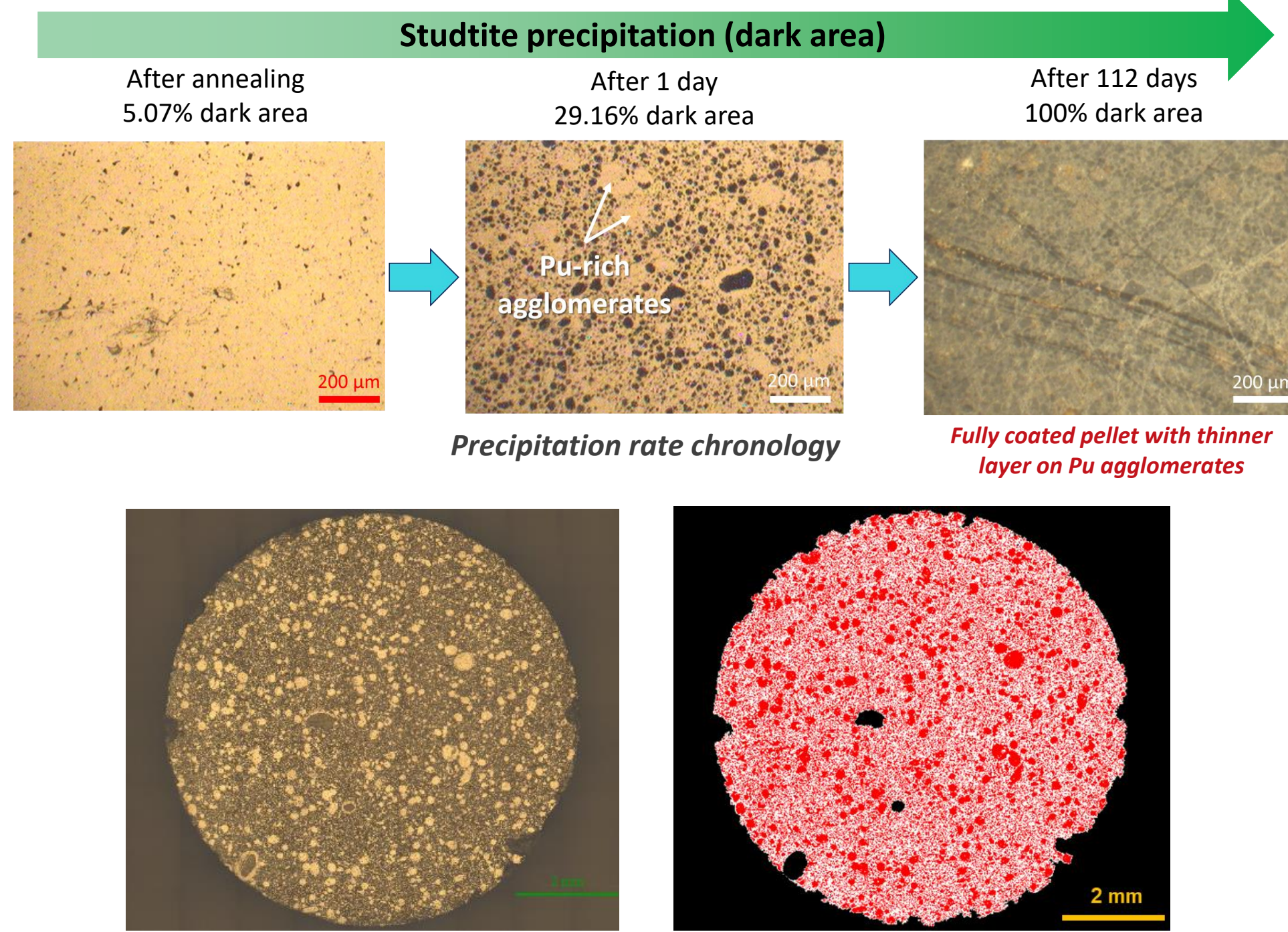
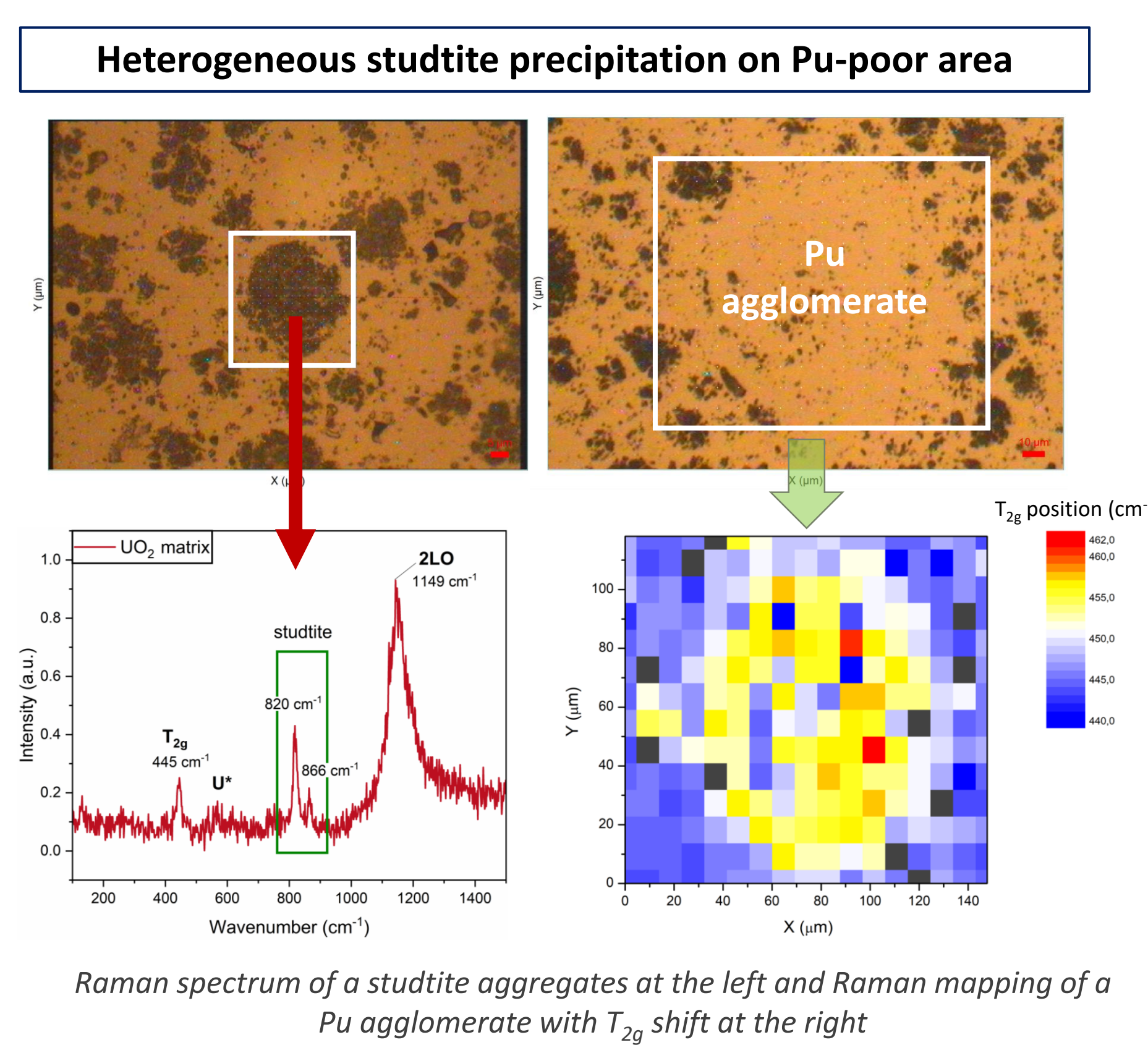
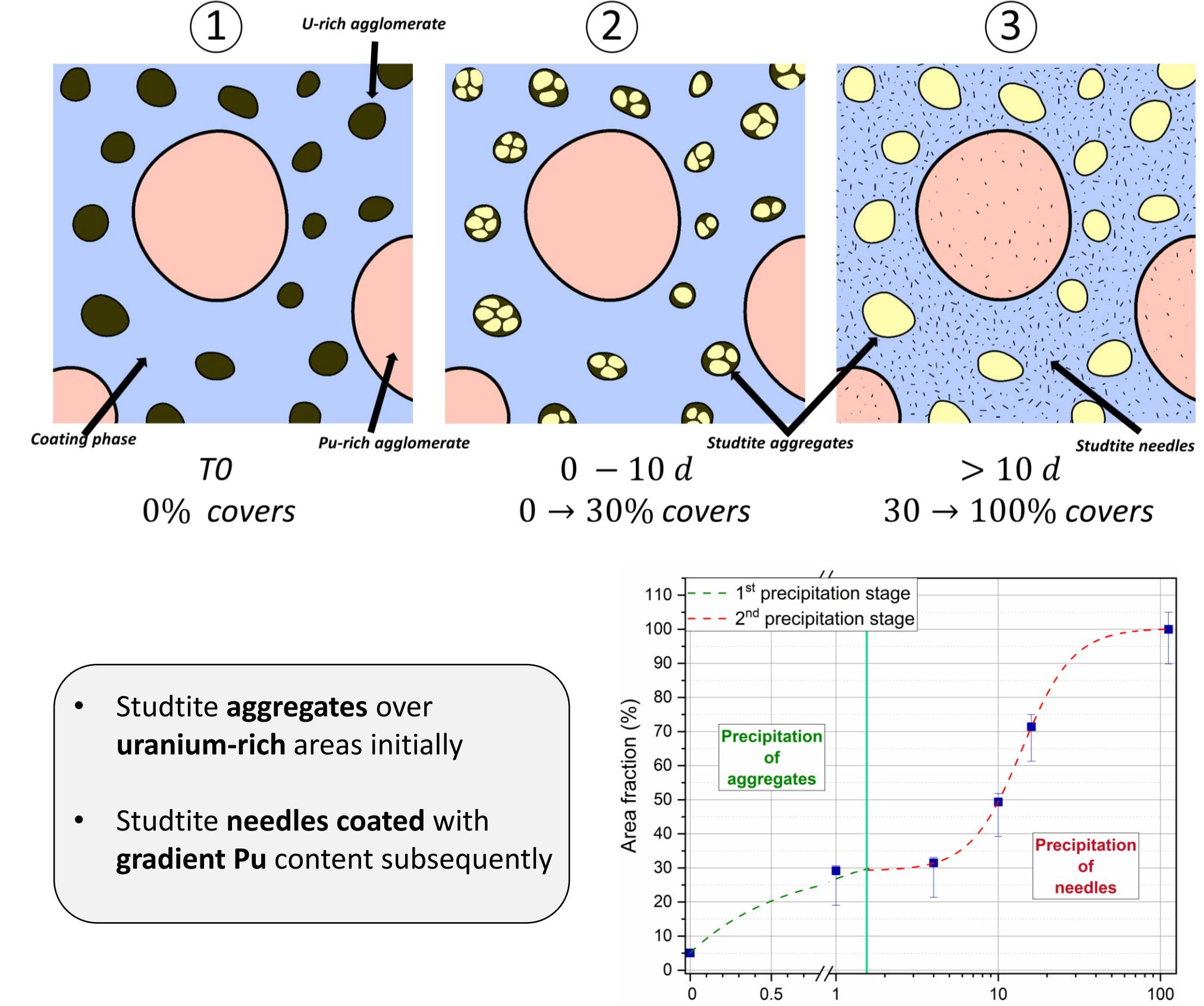


Image analysis
14.35% of surface fraction are Pu agglomerates
53.37% of pellet has been altered (Cut-off ≈ 8 mol.% of Pu)

Raman spectroscopy

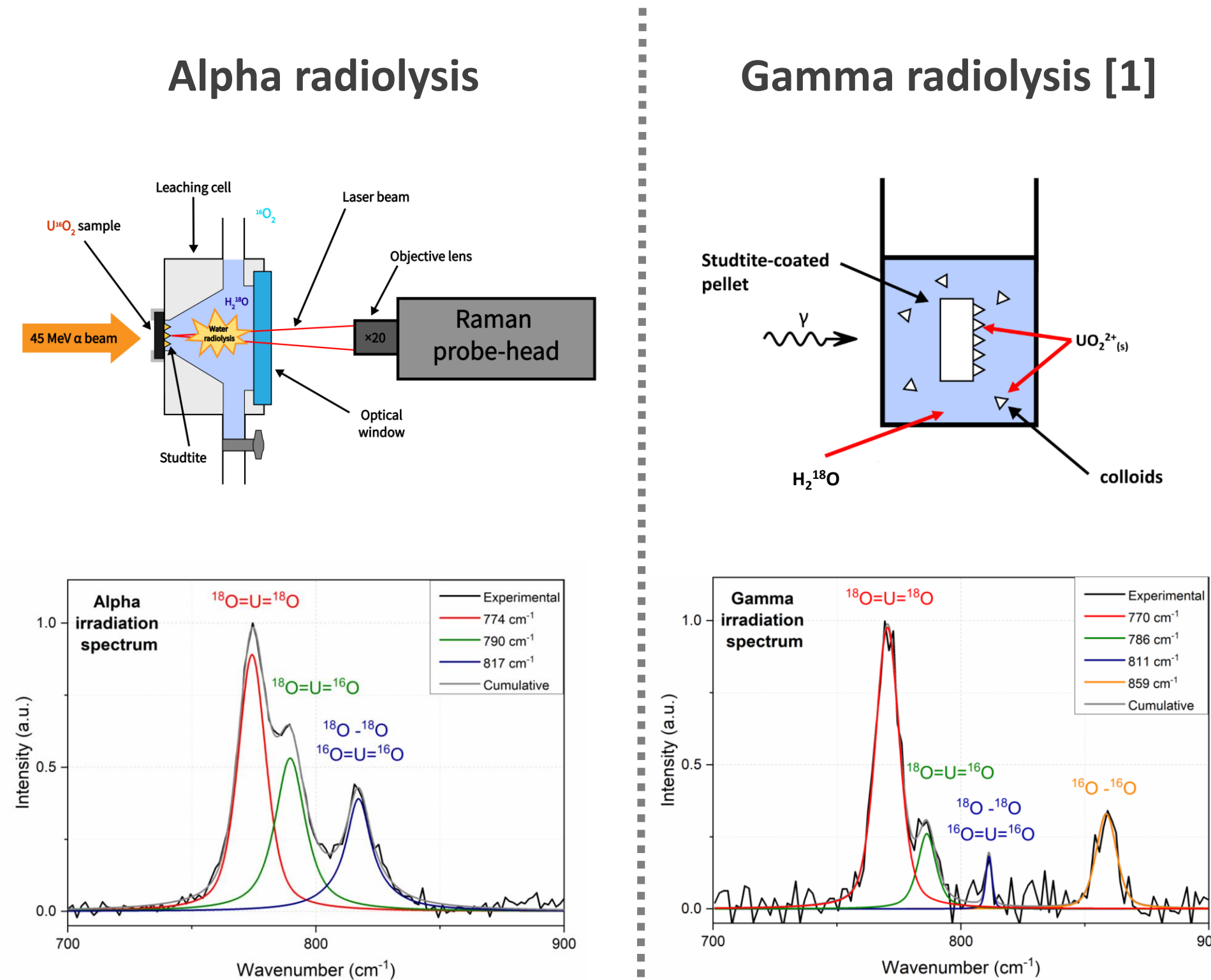


Precipitation process



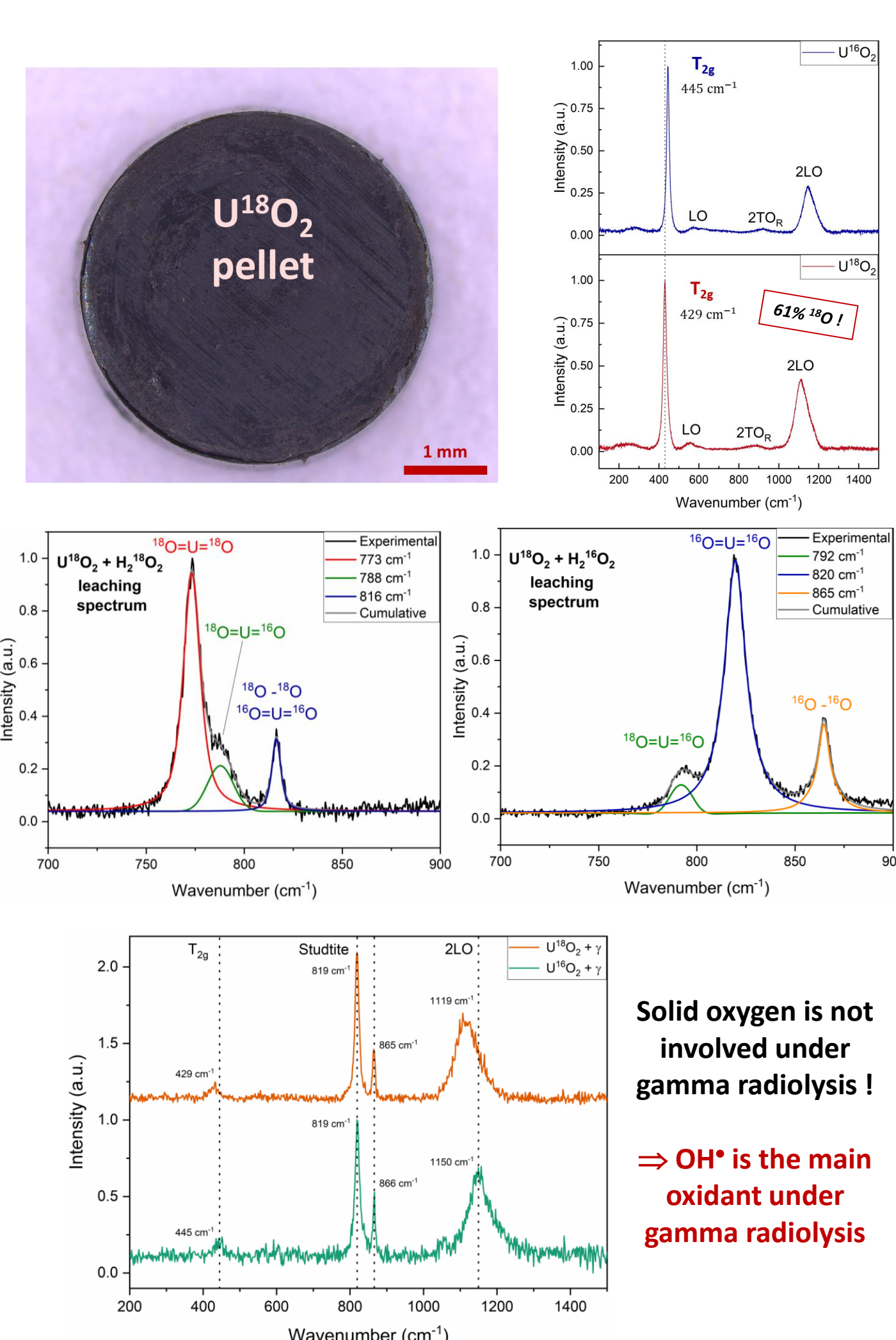
MECHANISTIC STUDIES OF STUDDITE FORMATION

U¹⁶O₂ leaching in H₂¹⁸O

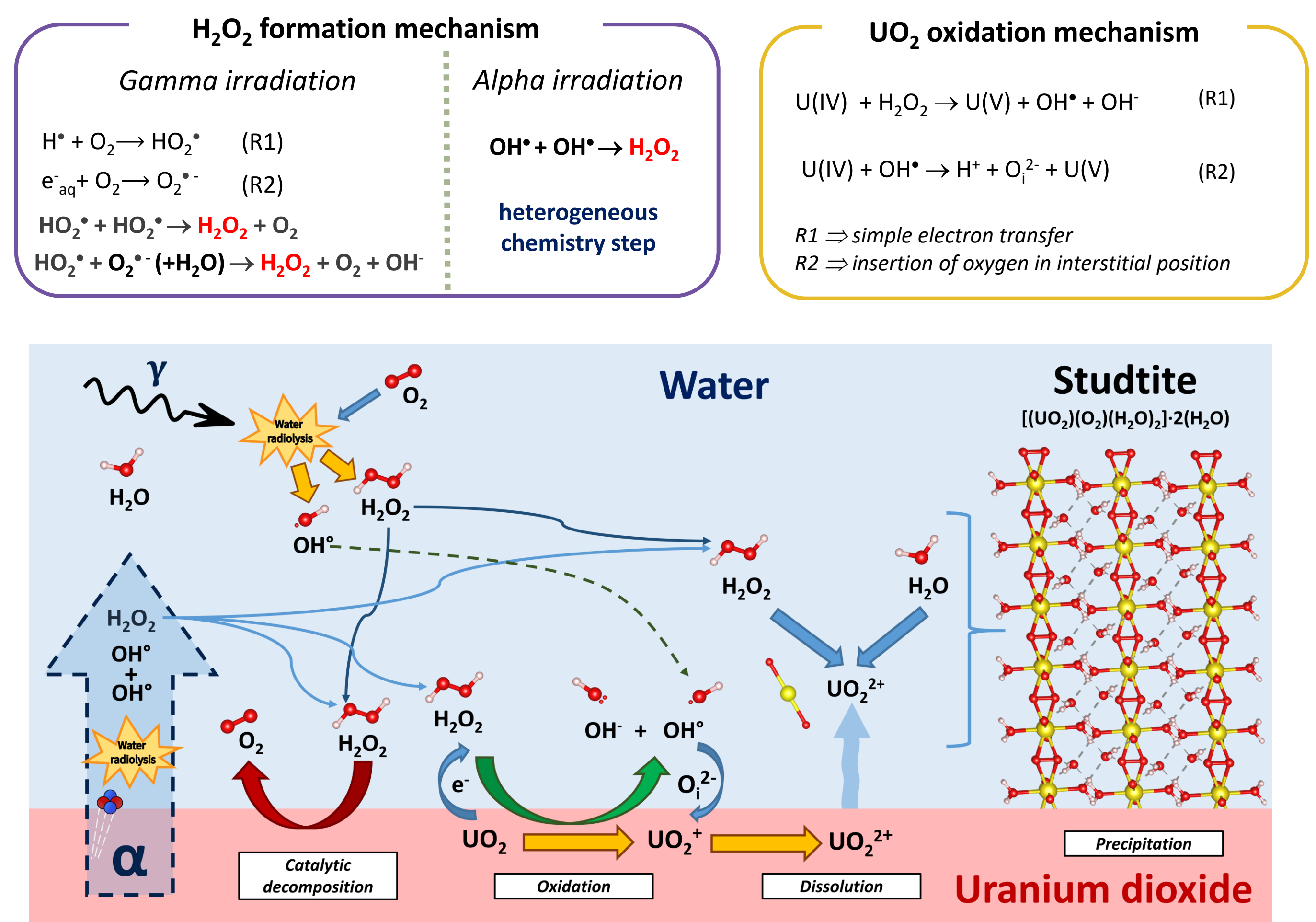


- Peroxide bond isotope differs in alpha vs. gamma radiolysis
- Stability of the peroxide bond between H₂O₂ in solution and studtite has been shown in a complementary experiment
- ⇒ Dependence of the studtite formation process on LET effects

U¹⁸O₂ leaching



Studtite formation mechanism



REFERENCES

[1] Lola Sarrasin et al. « Studtite Formation Assessed by Raman Spectroscopy and ¹⁸O Isotopic Labeling during the Oxidative Dissolution of a MOX Fuel J. Phys. Chem. C 125, (2021), p. 19209–19218.

[2] Martinez, J., et al., An original precipitation route toward the preparation and the sintering of highly reactive uranium cerium dioxide powders. Journal of Nuclear Materials, 2015. 462: p. 173–181

[3] Arganda-Carreras et al. Trainable Weka Segmentation: a machine learning tool for microscopy pixel classification. J. Bioinform. 33(15), (2017), p. 2424–2426

[4] Montaigne, T., et al., Oxidative dissolution of (U,Ce)O₂ materials in aqueous solutions containing H₂O₂. npj Materials Degradation, 2023. 7(1): p. 1–16

[5] Jégou, C., Entreposage sous eau des combustibles MOx. Etat des connaissances et besoins de R&D. 2005(NOTE TECHNIQUE DTCD/SECM/2005/24)