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## Study of the alteration mechanisms of (U,Th)O<sub>2</sub> and (U,Pu)O<sub>2</sub> mixed oxides under storage conditions

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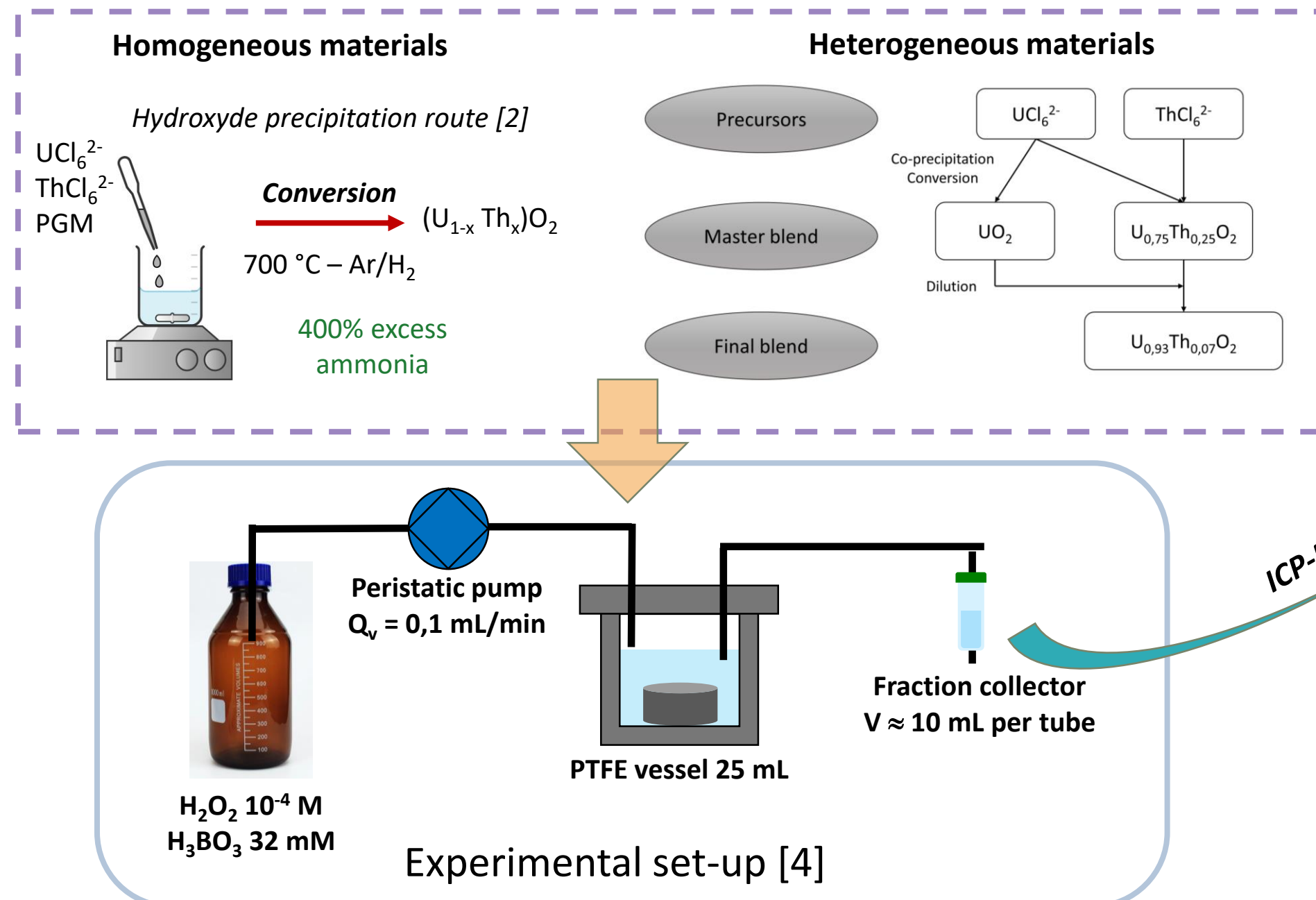
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## INTRODUCTION

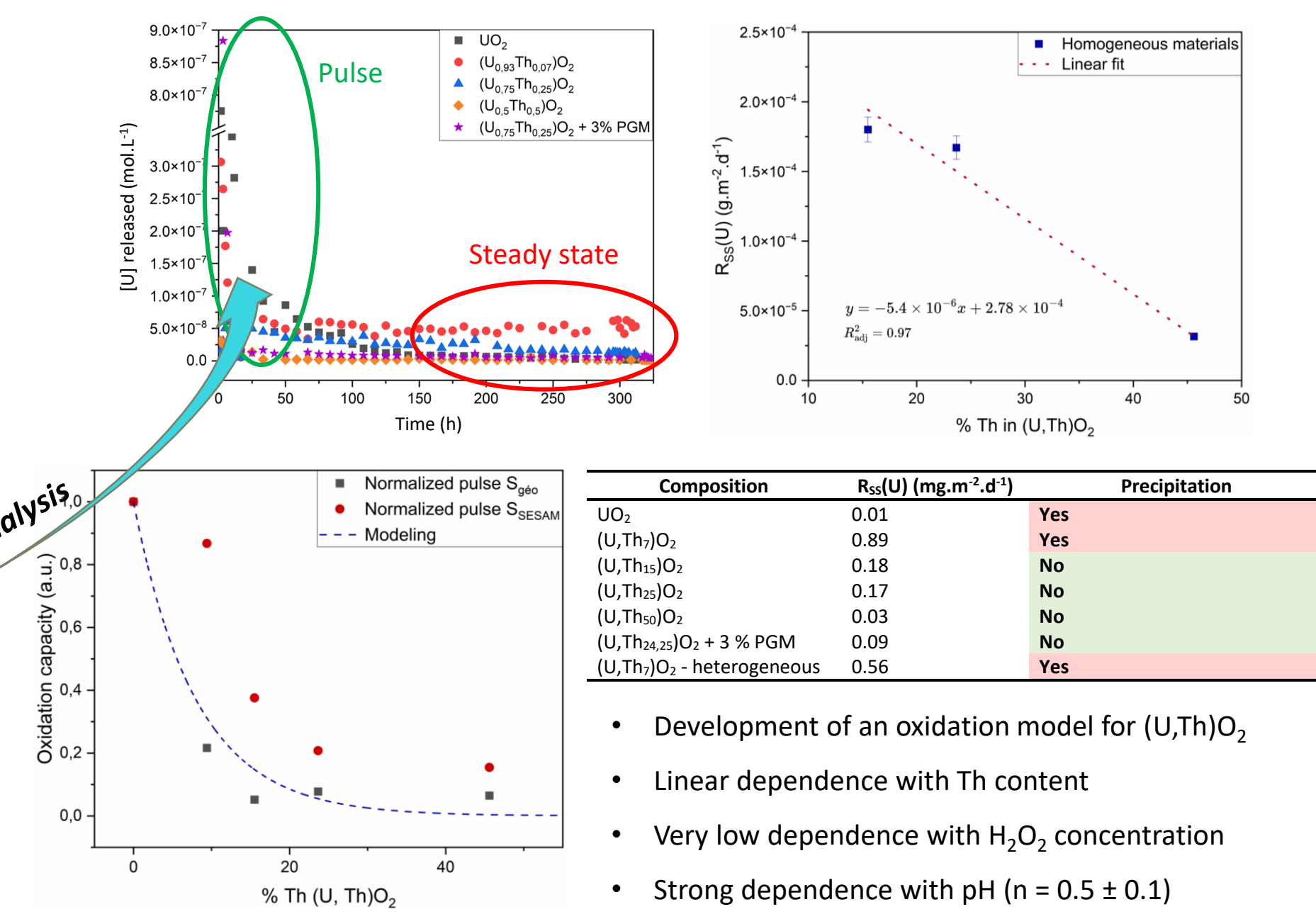
In France, the single recycling of irradiated fuel leads to the medium-term storage of irradiated **Mimas<sup>®</sup> MOx** (Mixed Oxide) fuel assemblies in pools, with a view to recycling or final disposal. During this **underwater storage**, attention is given to the potential impact of a **defect in the cladding**. Given that the pool water is oxidizing due to the presence of **radiolytic products** such as H<sub>2</sub>O<sub>2</sub> and OH<sup>\*</sup>, leaching by this water promotes the **oxidative dissolution** of uranium, followed by its reprecipitation as **studtite** [(UO<sub>2</sub>)(O<sub>2</sub>)(H<sub>2</sub>O)<sub>2</sub>].<sub>2</sub>(H<sub>2</sub>O). 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)O<sub>2</sub> MATERIALS

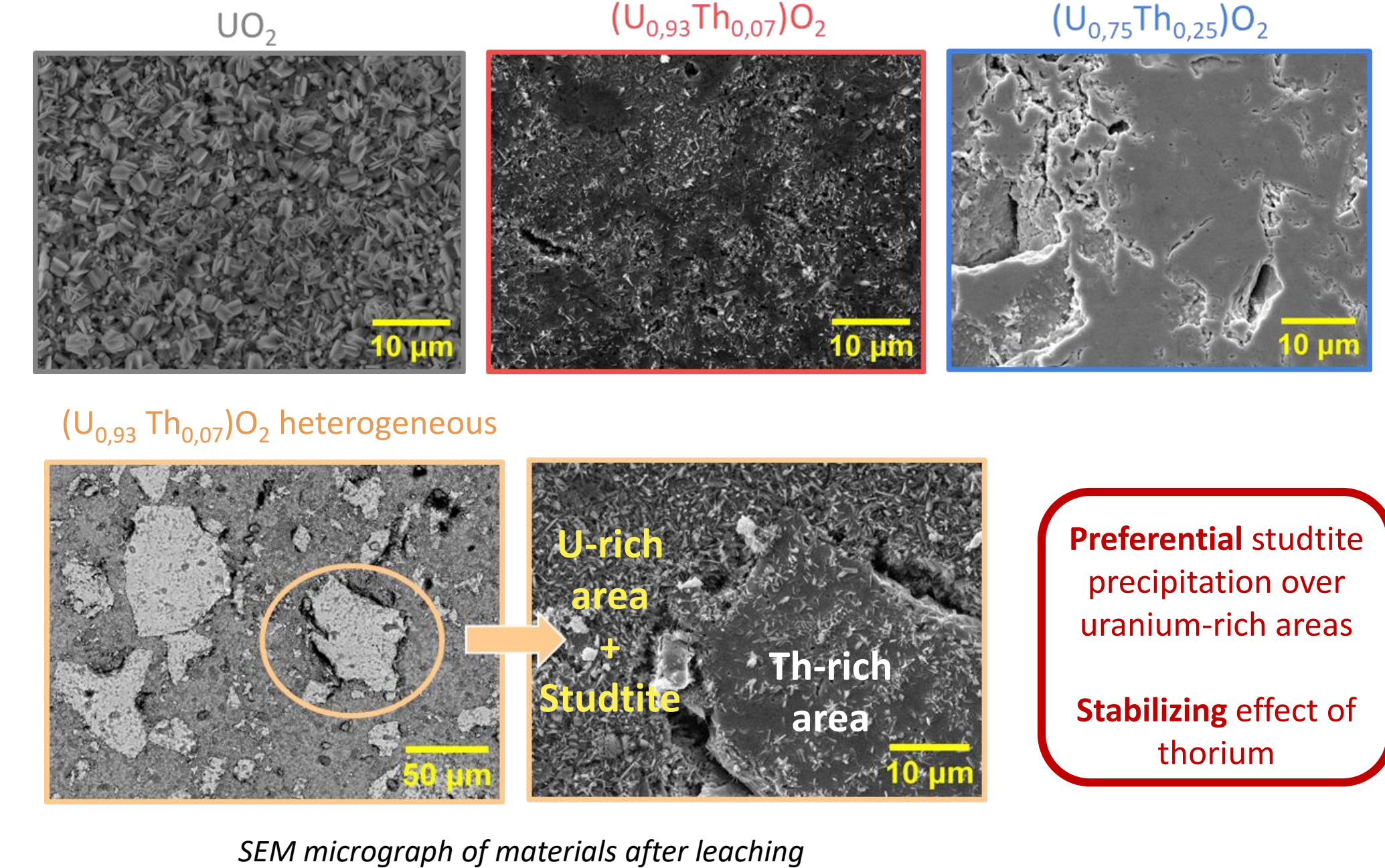
### Synthesis and experimental set-up



### Dynamic leaching experiment

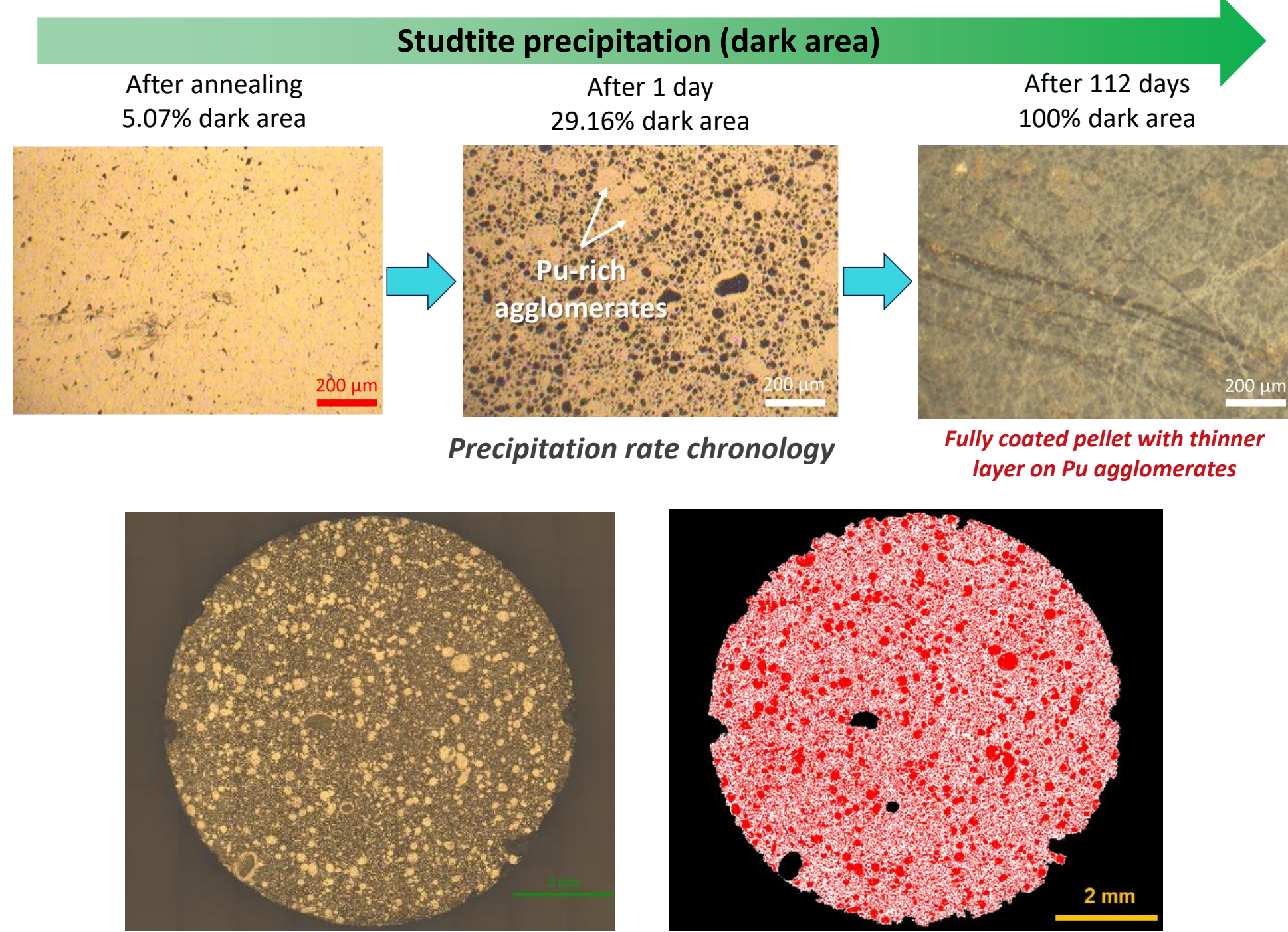


### Post-mortem characterization



## STATIC MODE LEACHING OF MIMAS MOx FUEL

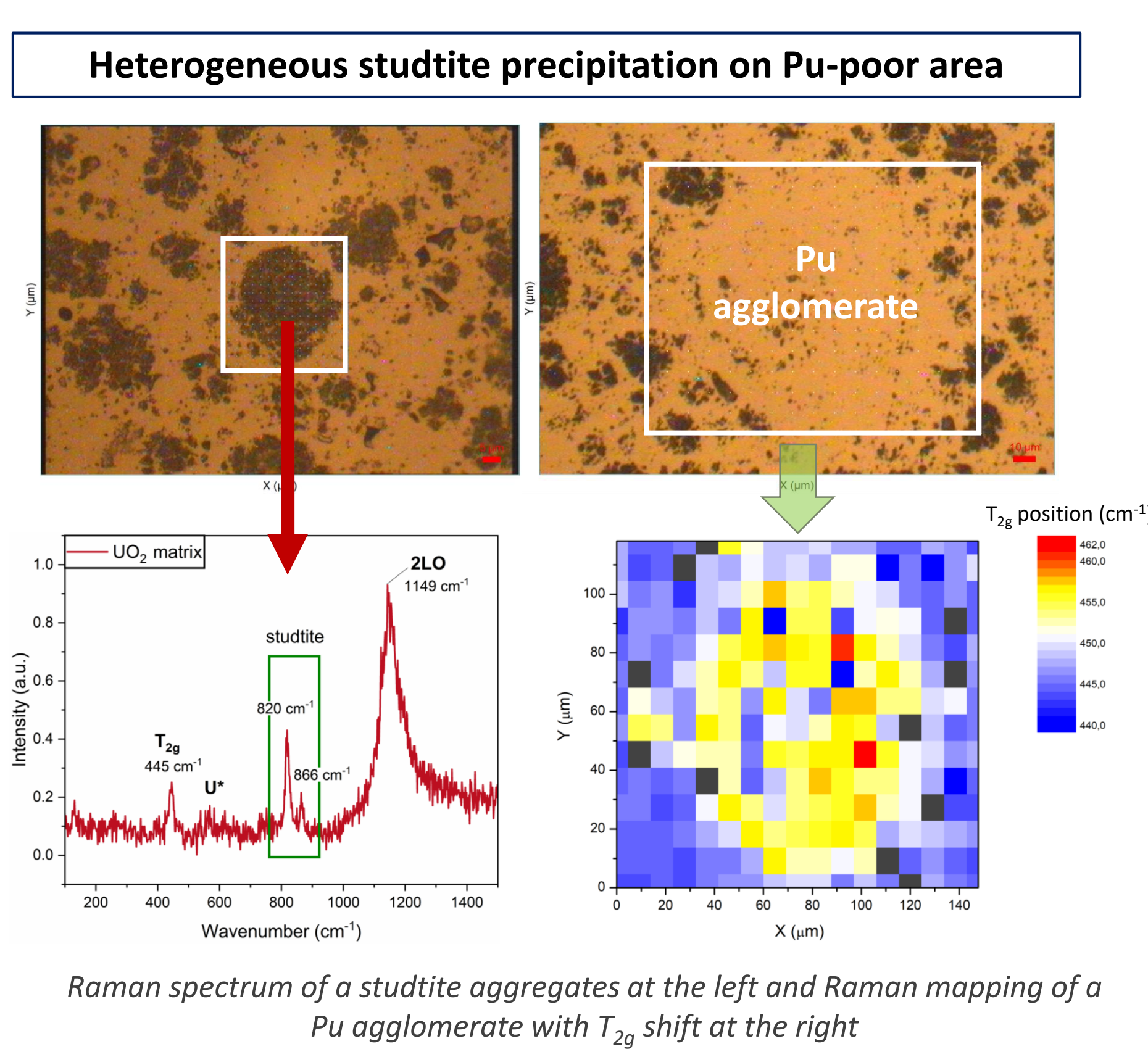
### Optical analyses



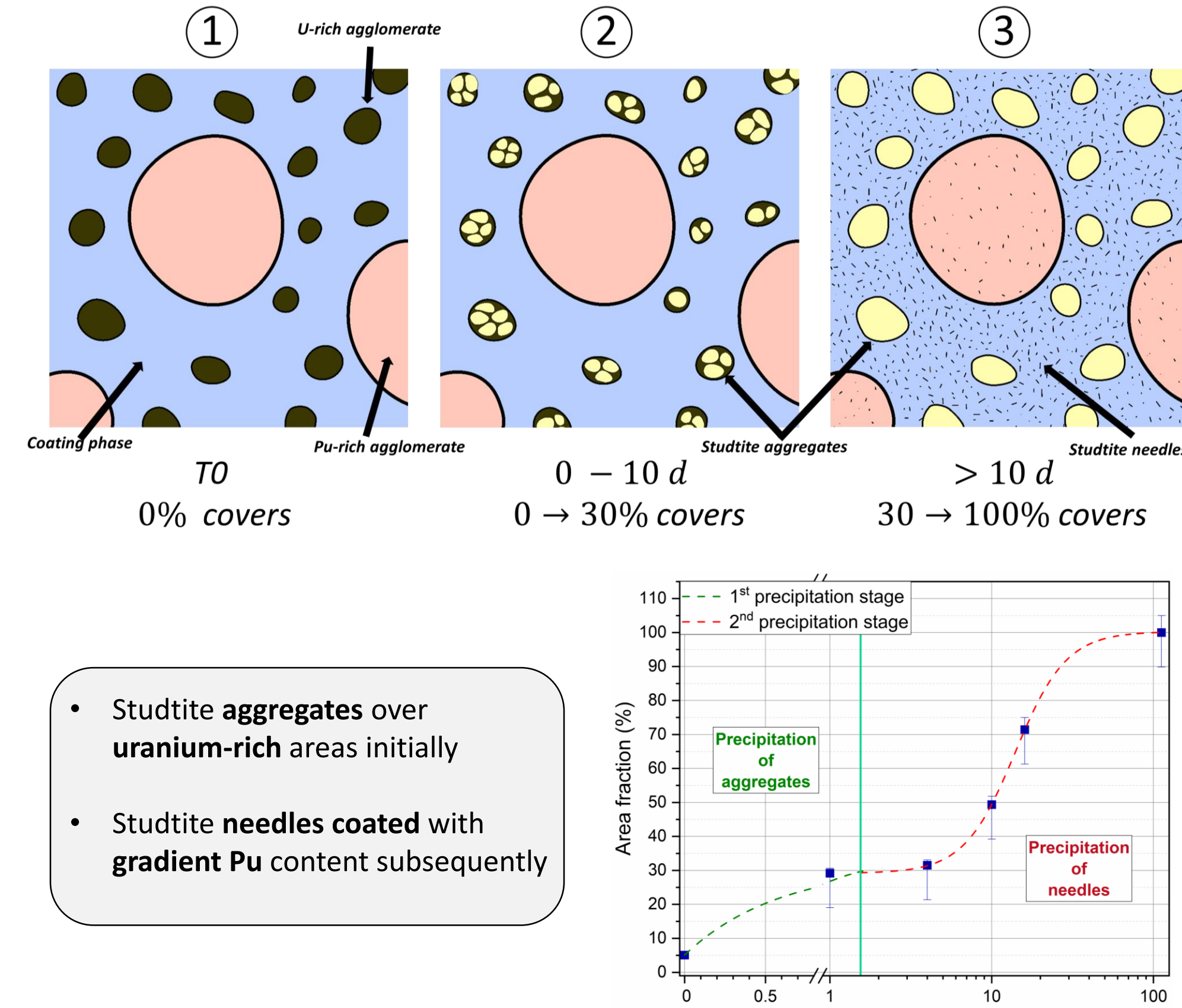
Segmentation of altered areas of the MOx pellet after studtite dissolution at the end of leaching by the Weka Fiji Plugin [3]

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

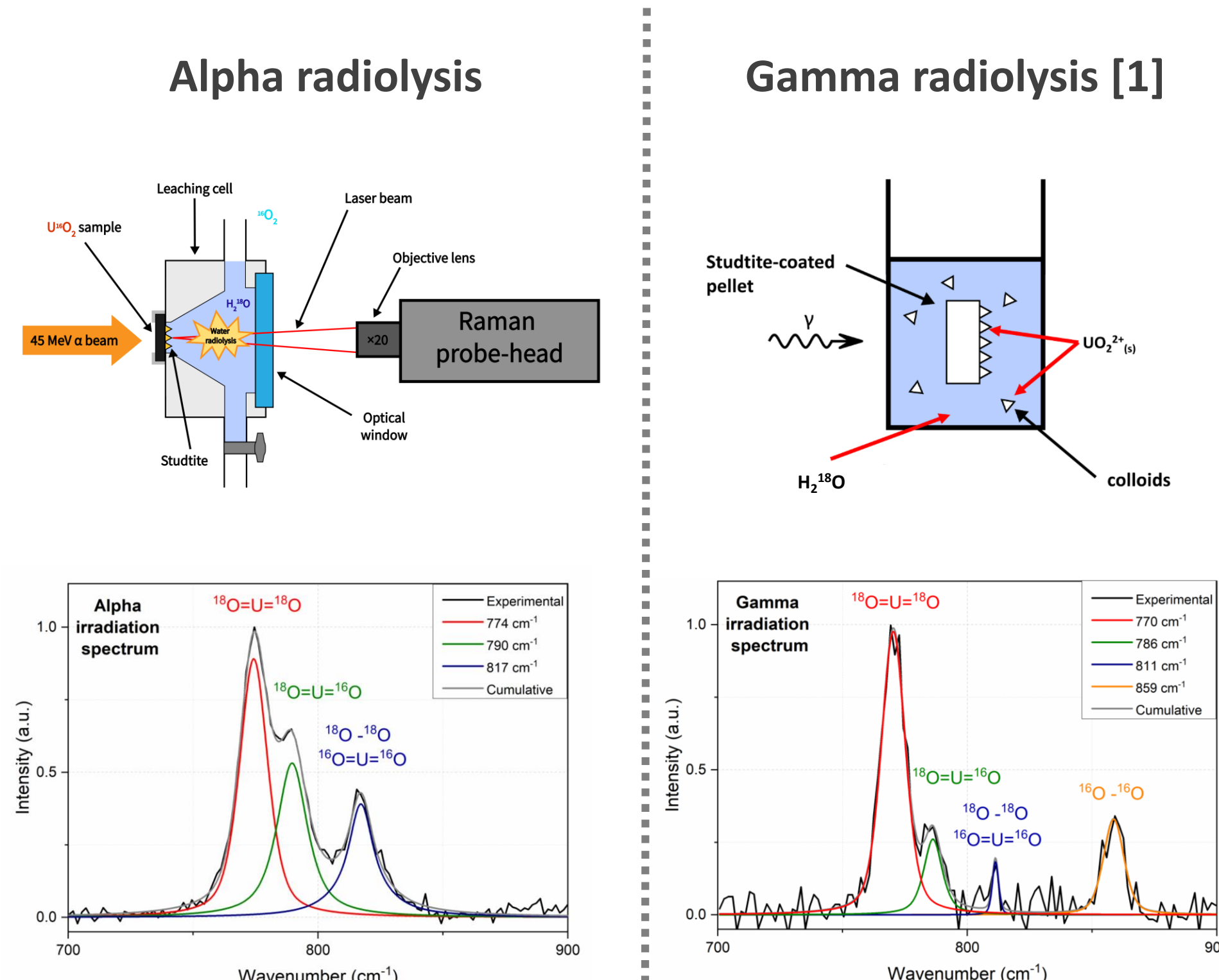


### Precipitation process



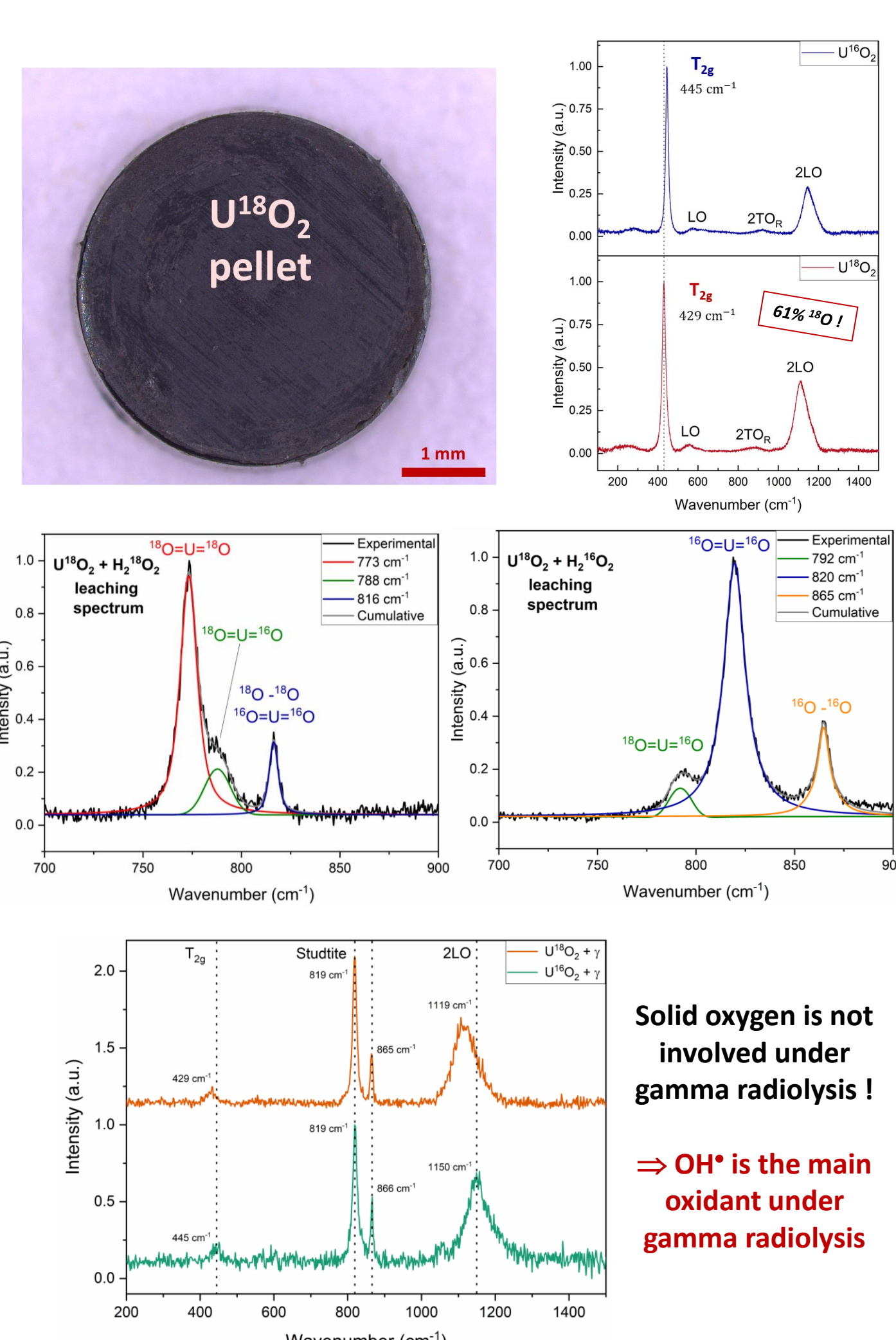
## MECHANISTIC STUDIES OF STUDDITE FORMATION

### U<sup>16</sup>O<sub>2</sub> leaching in H<sub>2</sub><sup>18</sup>O

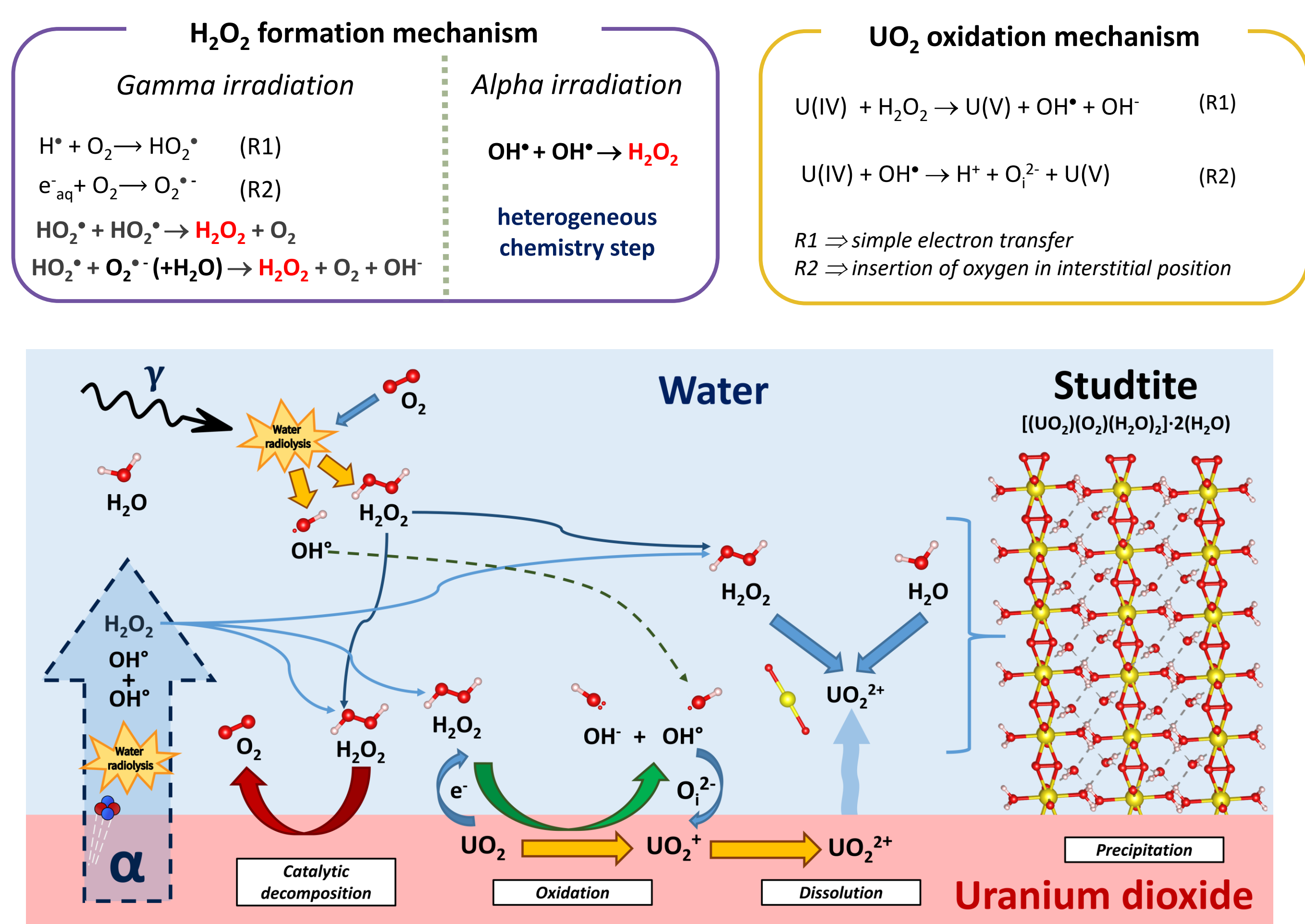


- Peroxide bond isotope differs in alpha vs. gamma radiolysis
- Stability of the peroxide bond between H<sub>2</sub>O<sub>2</sub> in solution and studtite has been shown in a complementary experiment
- ⇒ Dependence of the studtite formation process on LET effects

### U<sup>18</sup>O<sub>2</sub> leaching



### Studtite formation mechanism



## REFERENCES

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[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)O<sub>2</sub> materials in aqueous solutions containing H<sub>2</sub>O<sub>2</sub>. 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)