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

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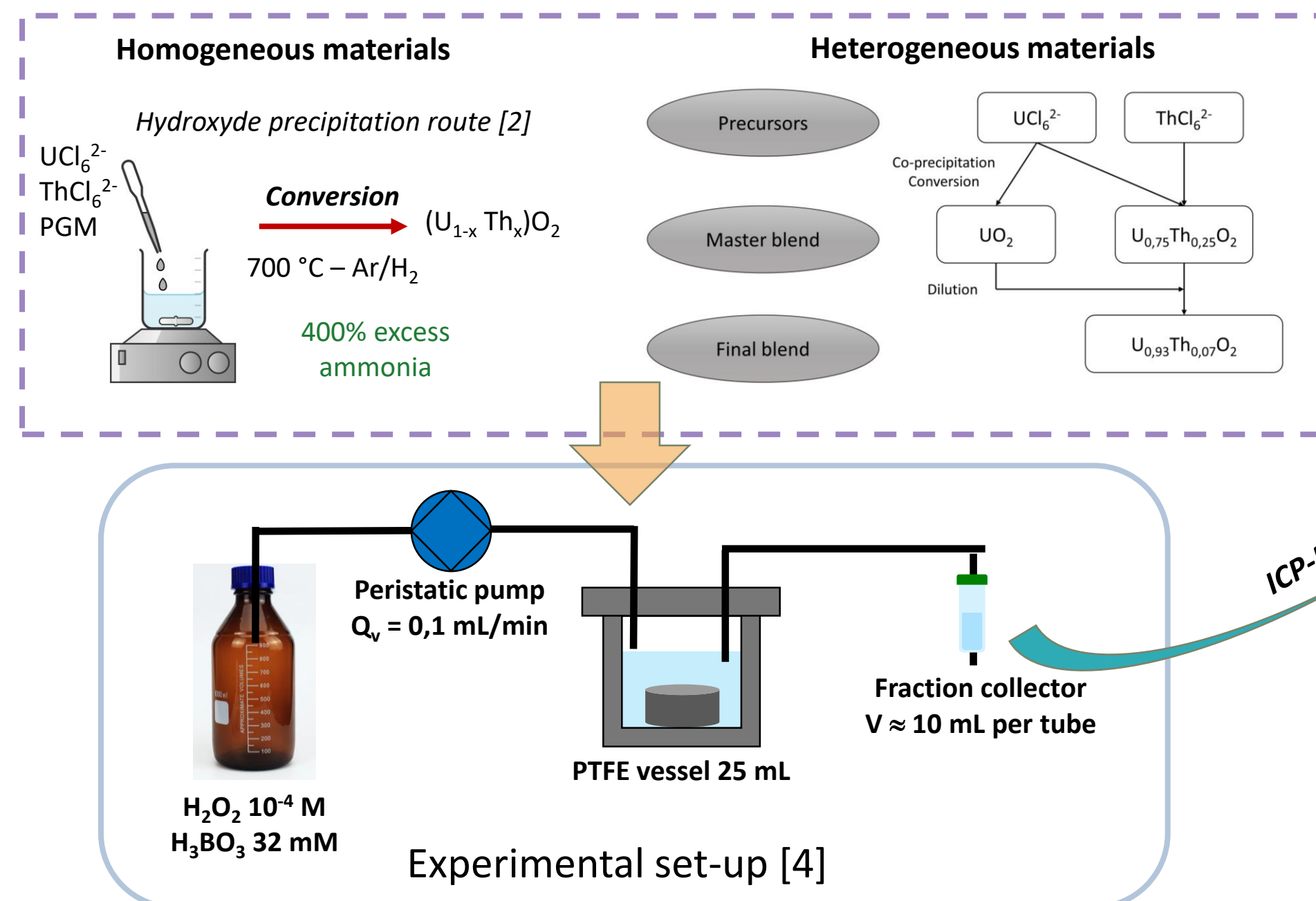
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INTRODUCTION

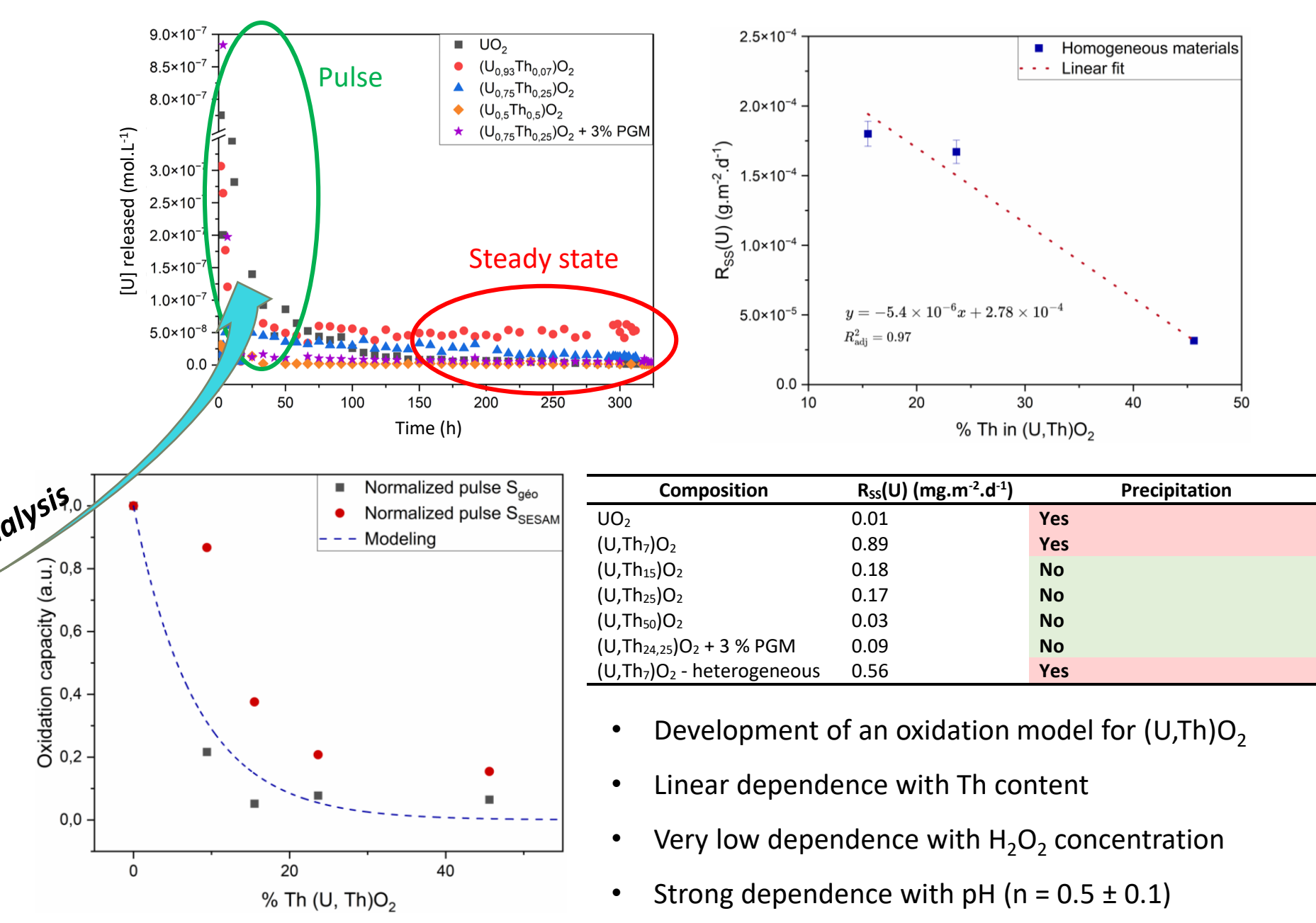
In France, the single recycling of irradiated fuel leads to the medium-term storage of irradiated **Mimas[®] MOx** (Mixed Oxide) fuel assemblies in pools, with a view to recycling or final disposal. During this **underwater storage**, attention is given to the potential impact of a **defect in the cladding**. 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

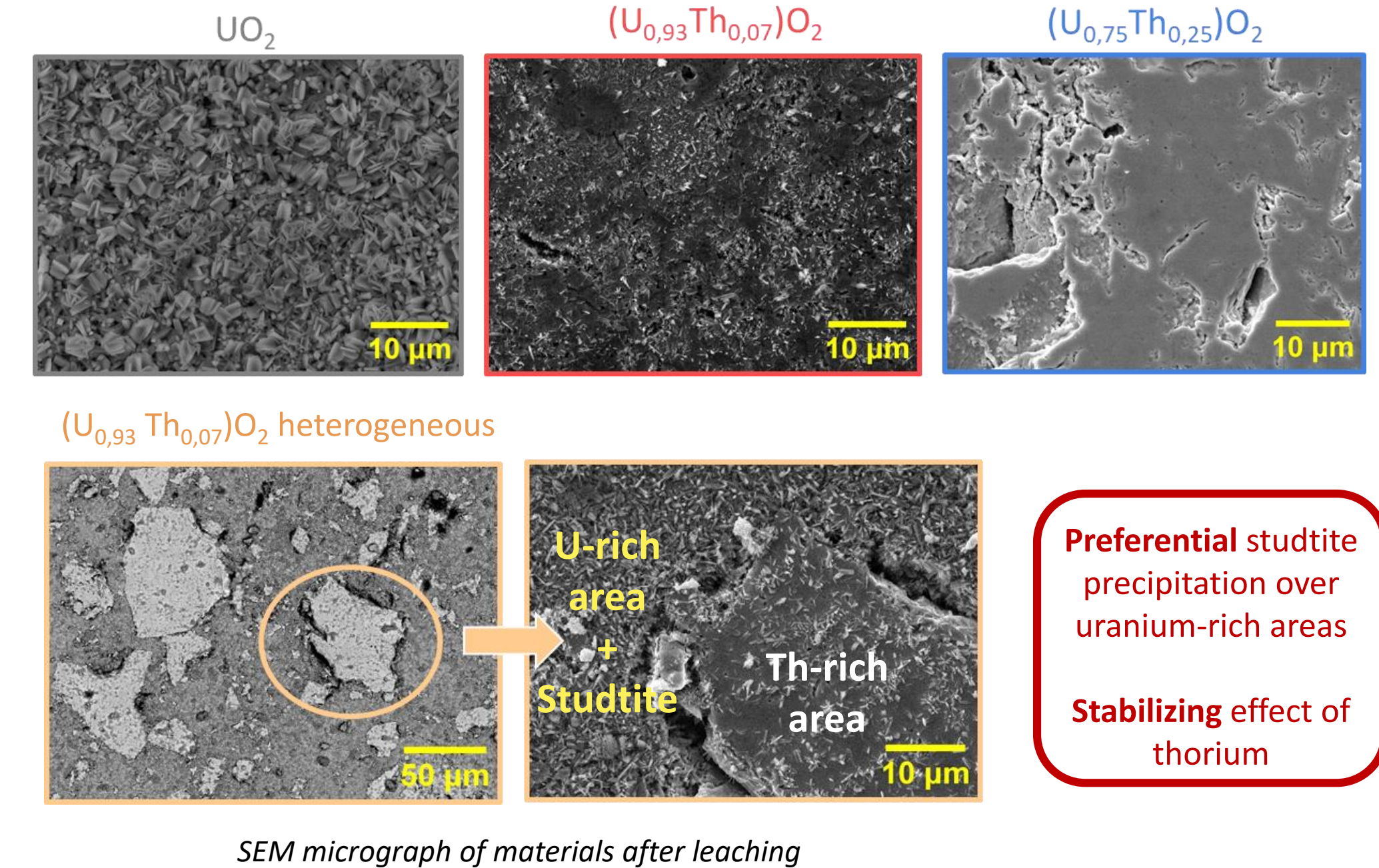
Synthesis and experimental set-up



Dynamic leaching experiment

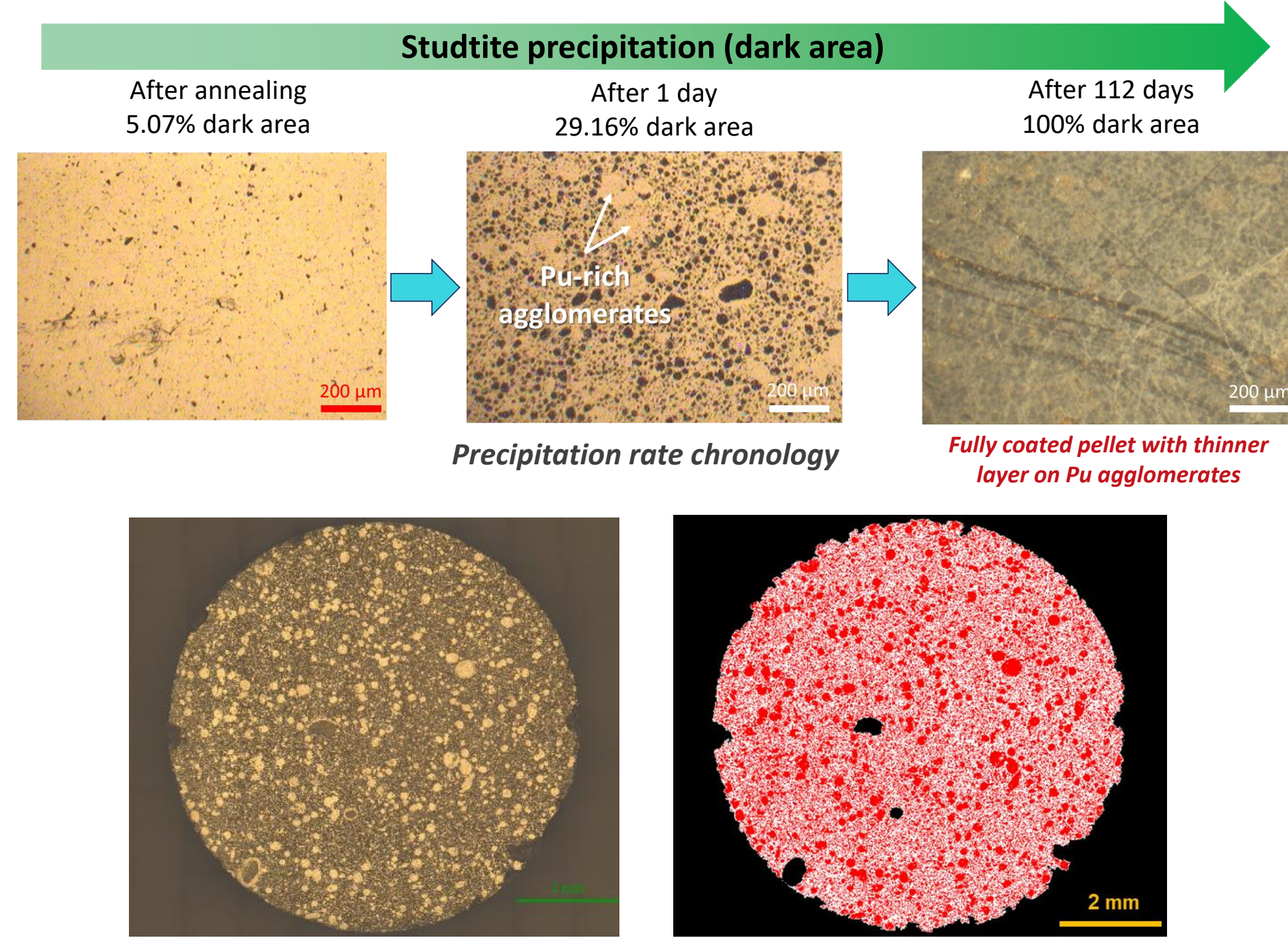


Post-mortem characterization



STATIC MODE LEACHING OF MIMAS MOx FUEL

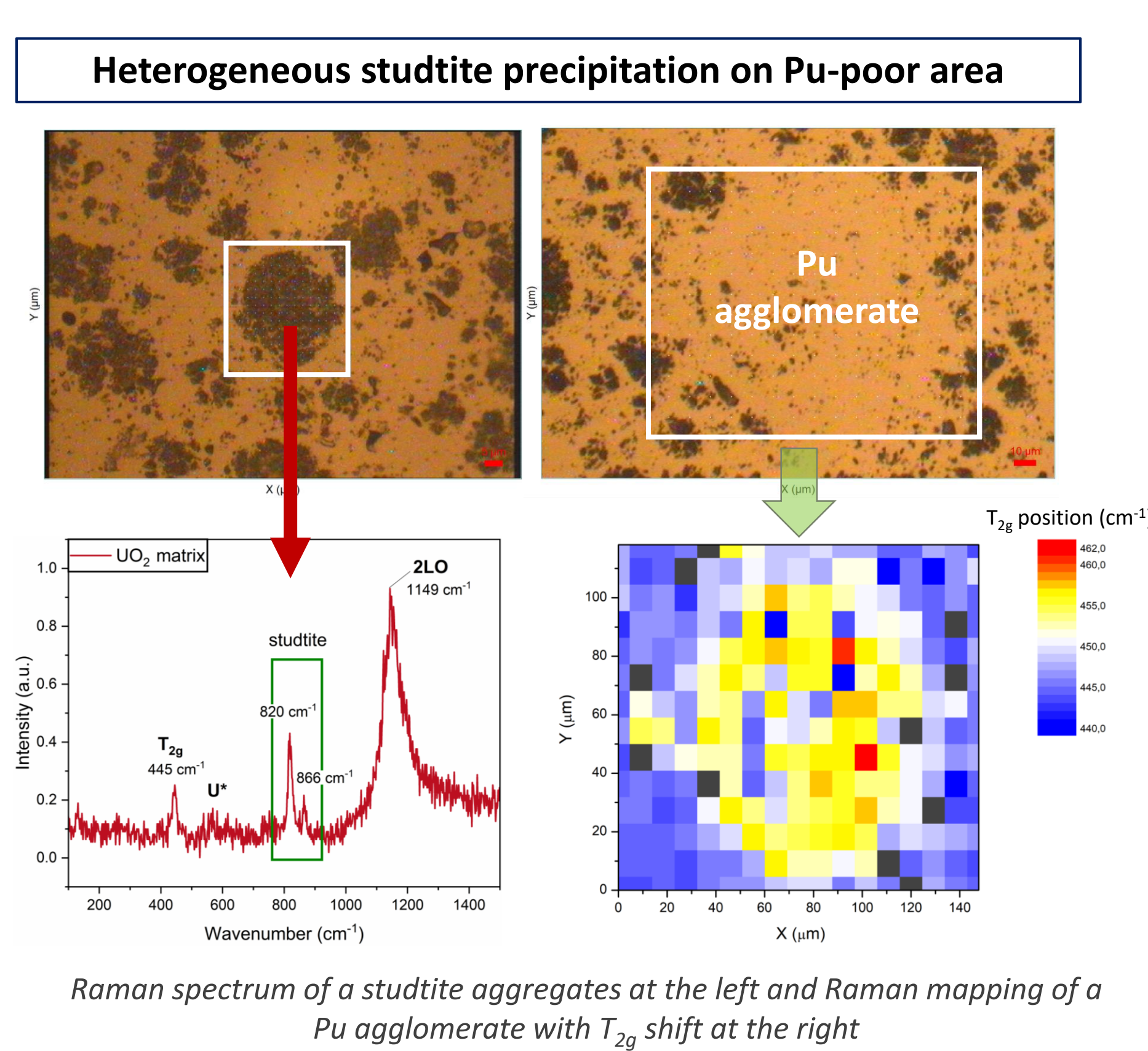
Optical analyses



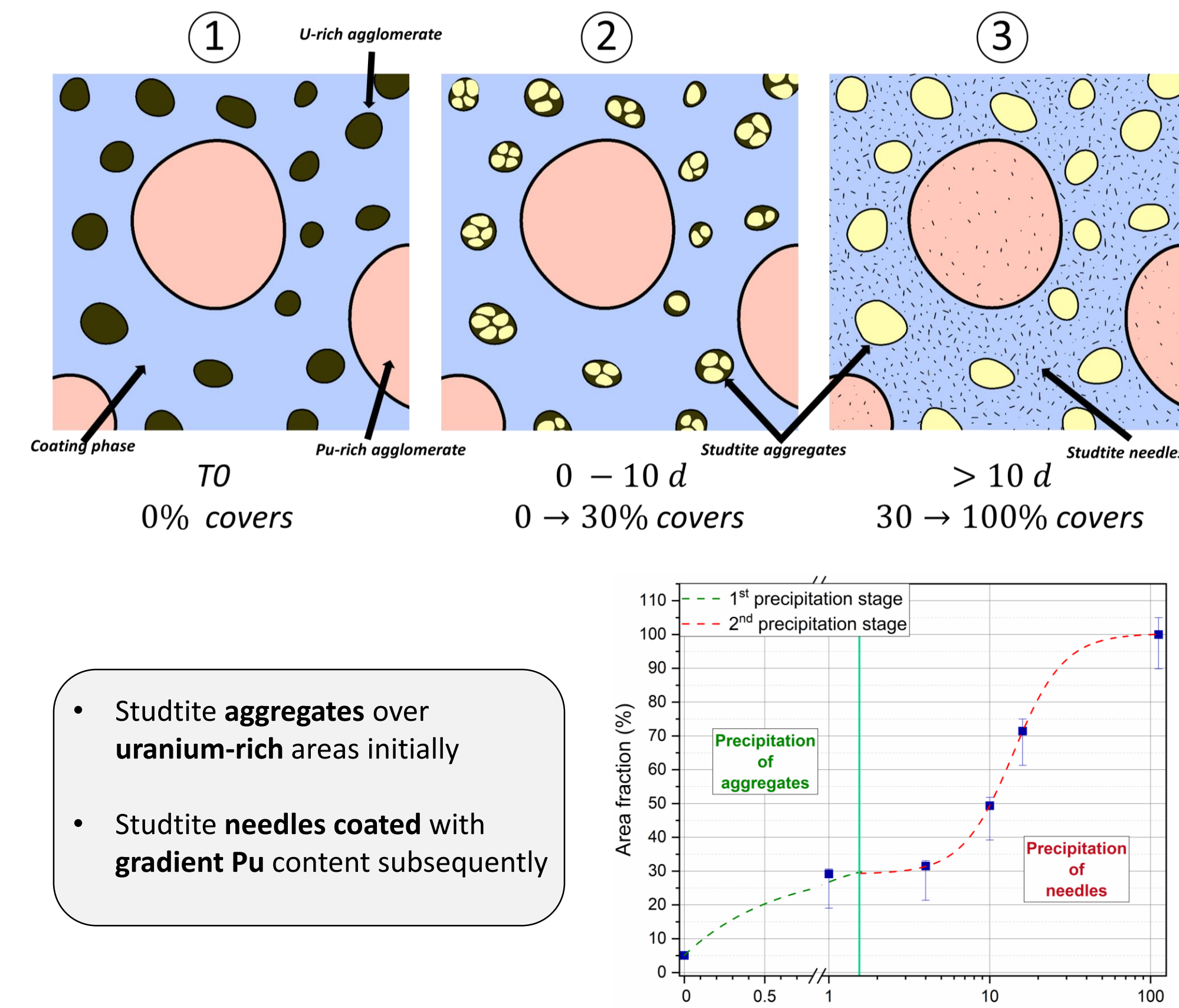
Segmentation of altered areas of the MOx pellet after studtite dissolution at the end of leaching by the Weka Fiji Plugin [3]

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

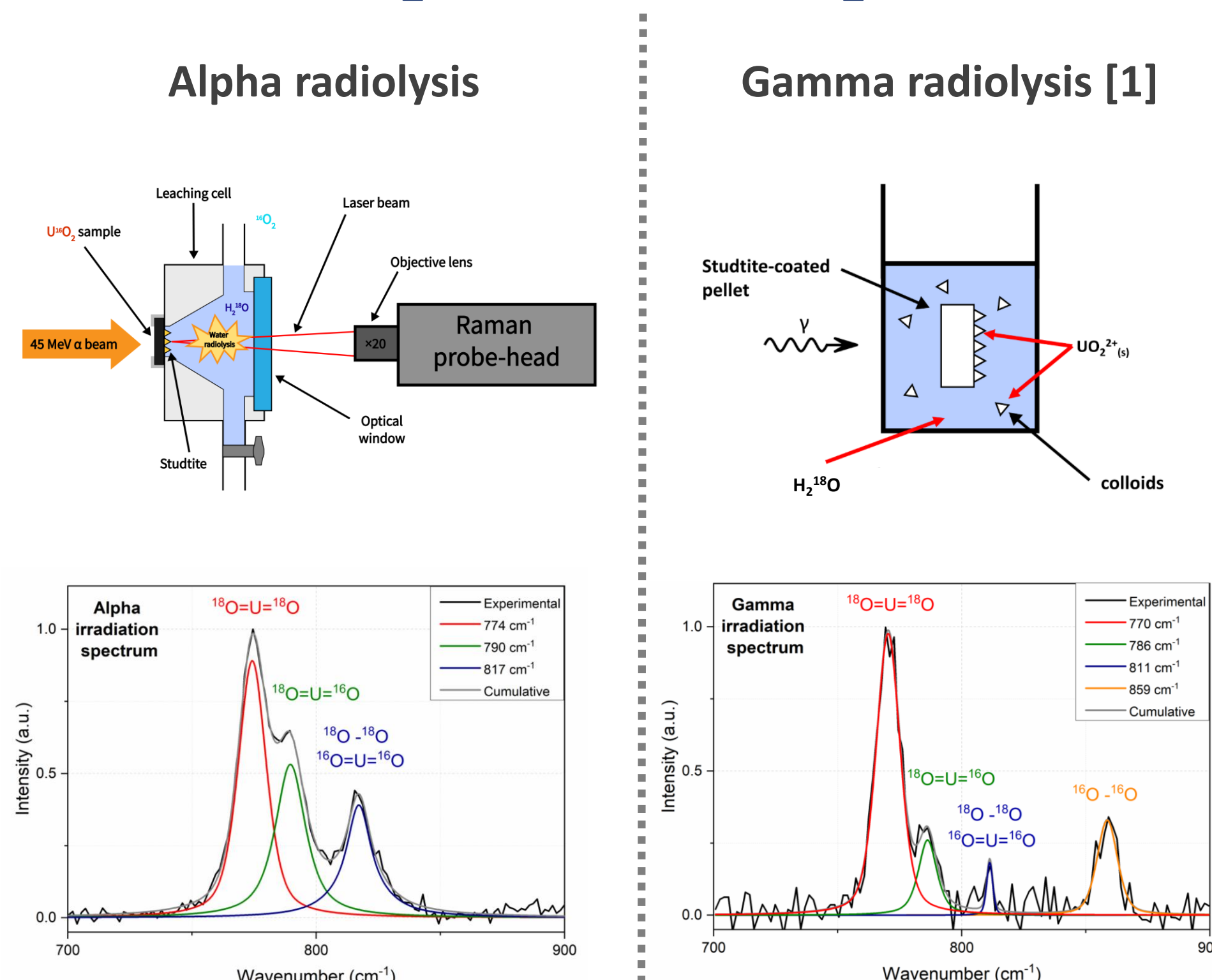


Precipitation process



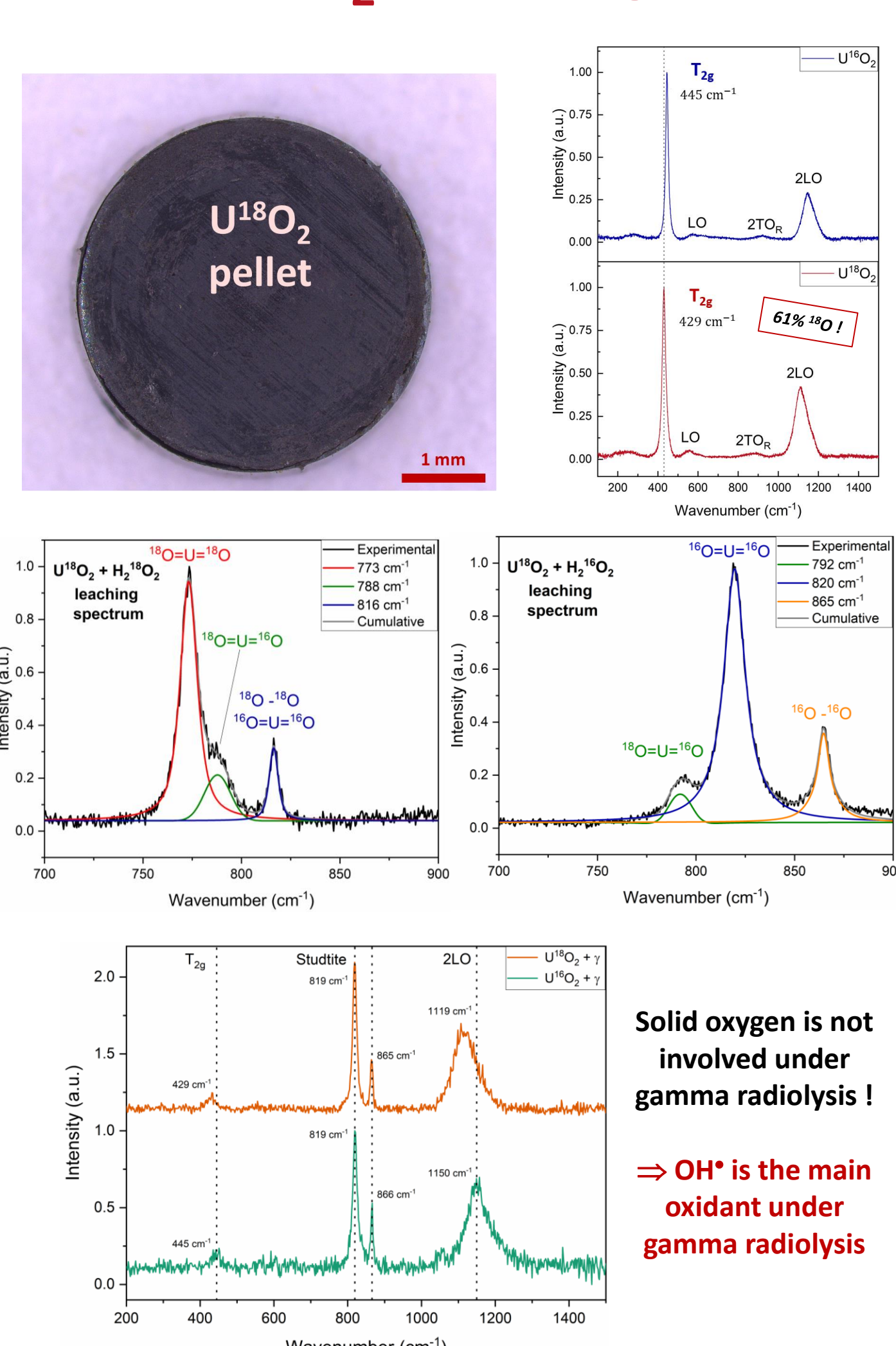
MECHANISTIC STUDIES OF STUDDITE FORMATION

U¹⁶O₂ leaching in H₂¹⁸O

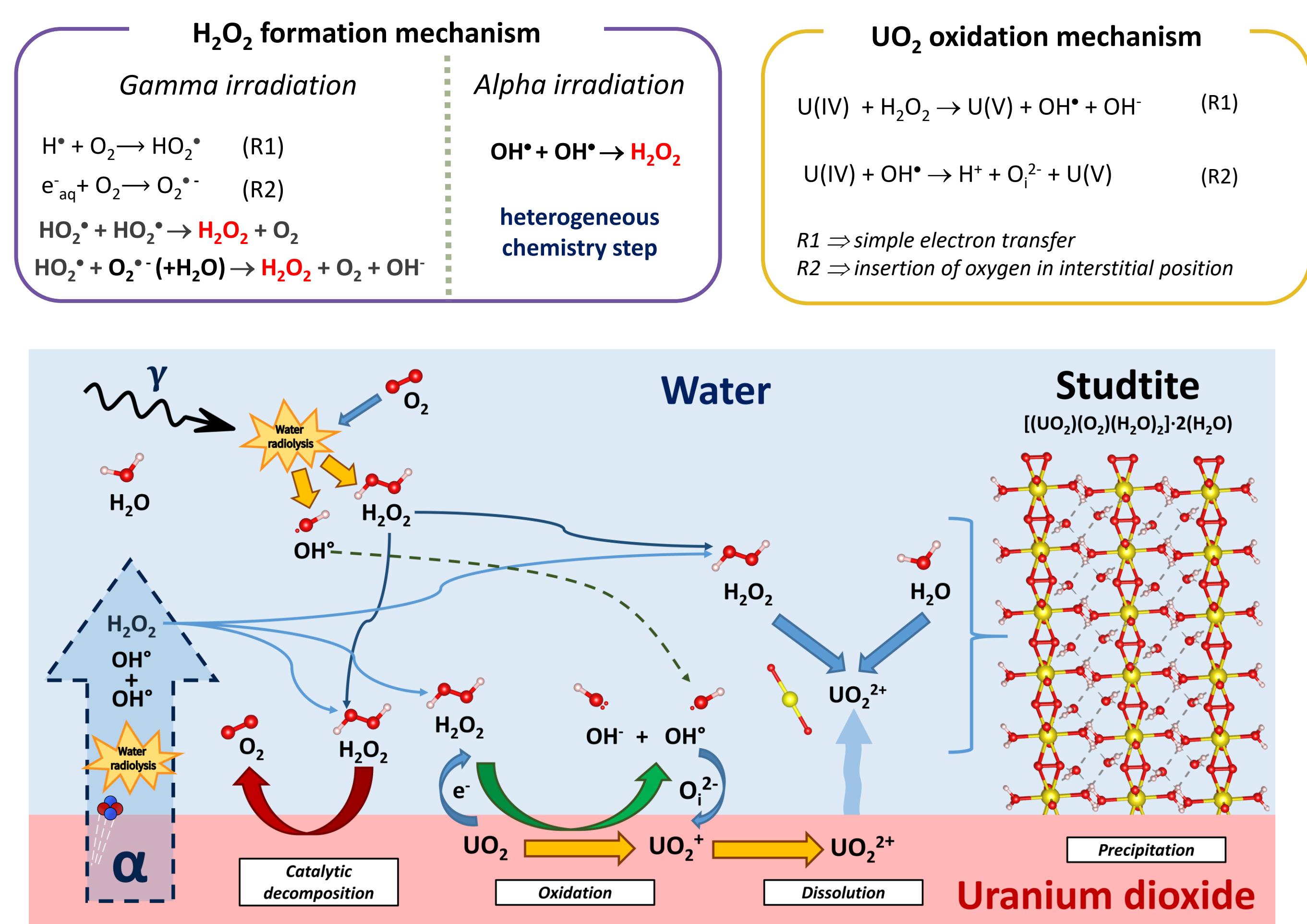


- Peroxide bond isotope differs in alpha vs. gamma radiolysis
- Stability of the peroxide bond between H₂O₂ in solution and studtite has been shown in a complementary experiment
- ⇒ Dependence of the studtite formation process on LET effects

U¹⁸O₂ leaching



Studtite formation mechanism



REFERENCES

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