

# Natural Analogues of Actinide Ceramic Waste Forms

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10-14 September 2018  
Trieste, Italy



# Performance criteria for waste forms

- 1. Introduction

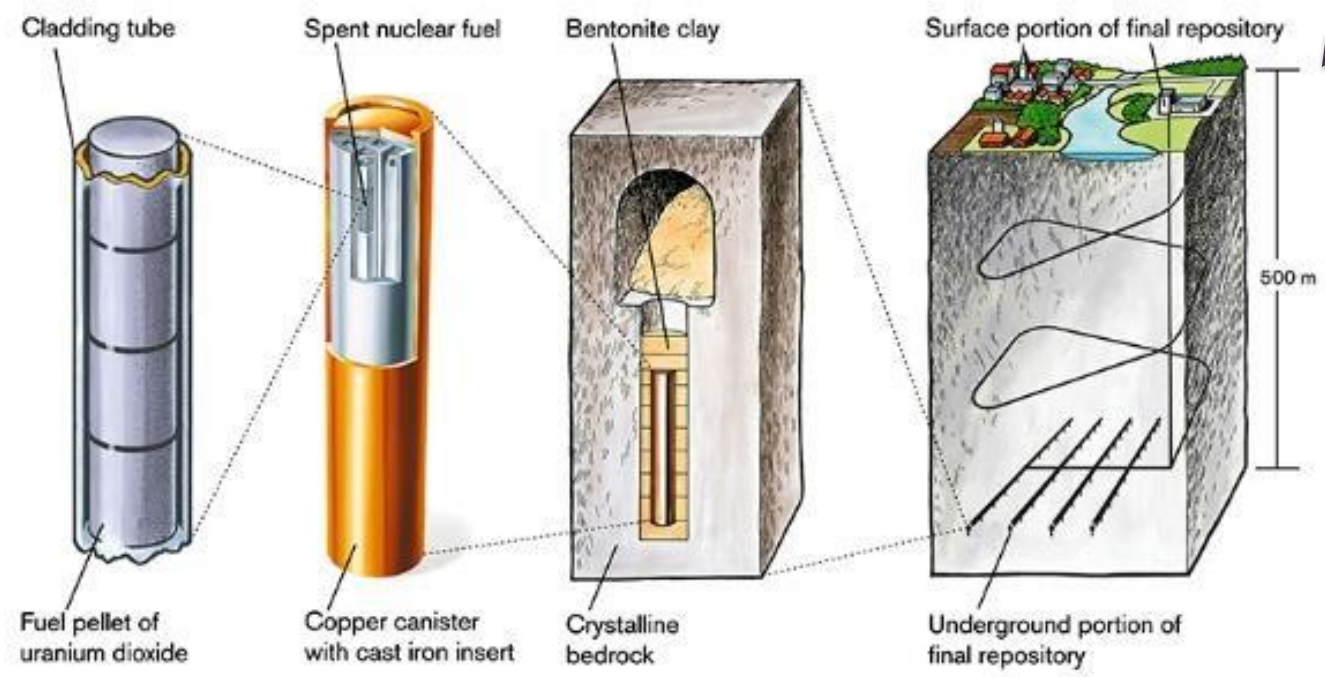
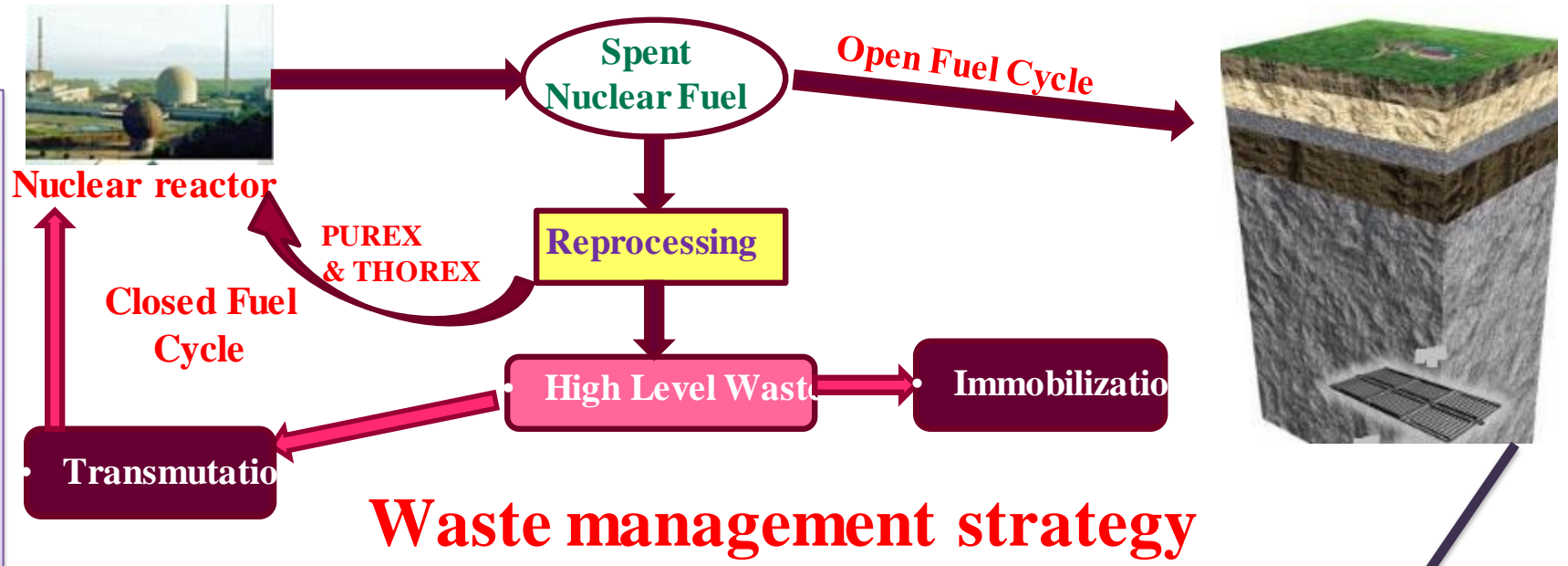
- 2. Samples in conditions

- 3. Behavior of Samples in experiment

- 4. Conclusions



INTRODUCTION



# Performance criteria for waste forms

- Must maintain mechanical integrity
- Must be radiation resistant
- Must have an acceptable thermal conductivity (especially important for HLW)
- Must be chemically flexible (usually)
- Must be capable of high waste loading
- Must have a low leach rate in foreseeable groundwater conditions

## Synthetic minerals- Matrices for Actinide Waste Immobilization

Vitreous matrices of Na-glass

Phosphate glasses  
Borosilicate

**Ceramic-Synroc**

**Ceramic – Ti-pyrochlore**

**Ceramics based on zircon/zirconia**

**Gadolinia-stabilized cubic zirconia**

**Tetragonal zirconia,  $(\text{Zr}, \text{Pu})\text{O}_2$**

# What will be the immobilization happening in the future?

## Radiation-Induced Swelling

### Examples of Swelling

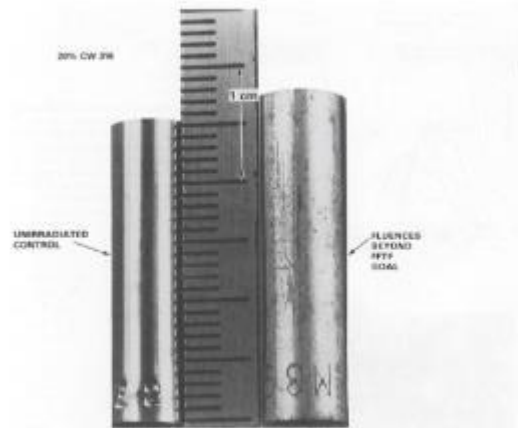


Figure 6-24. Excess observed swelling ( $\approx 10\%$  linear,  $\approx 33\%$  volumetric) in irradiated 20% cold worked AISI 304 cladding tube at  $1.5 \times 10^{17} \text{ n cm}^{-2}$  ( $E > 0.1 \text{ MeV}$ ) at  $311^\circ\text{C}$  in ATR-HTR after Strassburg et al., 1992). Note that, in the absence of physical constraints, all relative proportions are preserved during swelling.

Ref: Garner, Ch. 6, Irradiation Performance of Cladding and Structural Steels in Liquid Metal Reactors, of "Nuclear materials part 1", Vol 10A, Published by VCH, Germany

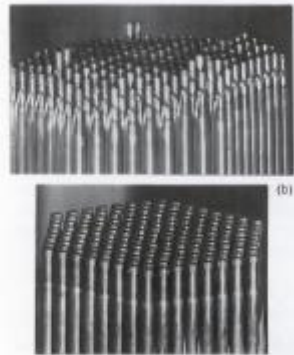


Figure 6-131. (a) Top of a bundle of D9 fuel pins irradiated to a peak fluence of  $2.1 \times 10^{17} \text{ n cm}^{-2}$  ( $E > 0.1 \text{ MeV}$ ), showing varying length of pins in response to gradients across the bundle in flux and temperature and also to small variations in pin fabrication history and composition. (b) An undistorted fuel pin assembly with accretion HT9 cladding at  $1.9 \times 10^{17} \text{ n cm}^{-2}$  ( $E > 0.1 \text{ MeV}$ ) after Makenas et al., 1990a).







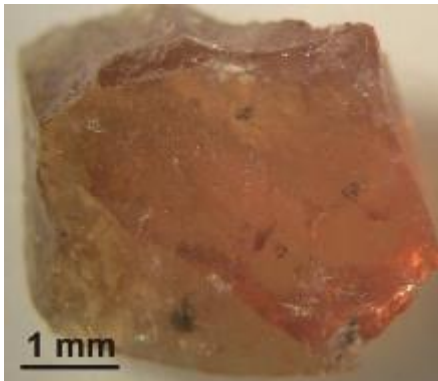


- Metamict state was first defined by Broegger (1893) in a Danish encyclopedia. A lot of studies have been carried out to understand properties of metamict minerals in the past.
- Metamict minerals are characterized by amorphous states but initially they were crystalline. Due to admixture of natural radioactive elements such as U and Th, their crystal structures were destroyed.
- Metamict minerals can be considered as natural analogues of ceramic nuclear waste-forms affected by radiation damage (also natural chemical alteration).
- Therefore, the study of metamict minerals helps to understand behavior of waste forms under geological conditions.

## 2. Samples in conditions

- It concludes radioactive elements, especially, actinides
- Natural crystal, or pre-state is a crystal
- Stable in the geological conditions
- Grain is not small, however, the biggest is the best!

What else conditions?



From nepheline syenites from the Khibiny alkaline massif in the Kola Peninsula, Russia



From granite pegmatites of Karelia, Russia

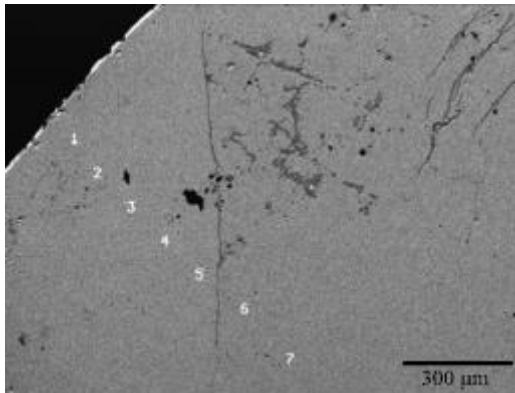
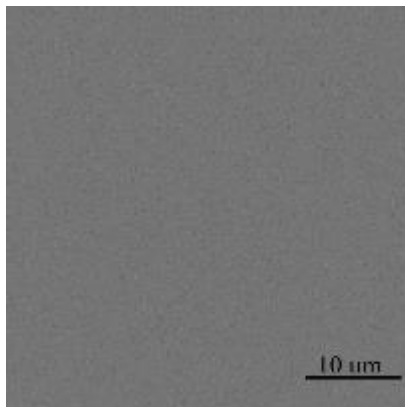


From diorite of Jiangsu, China

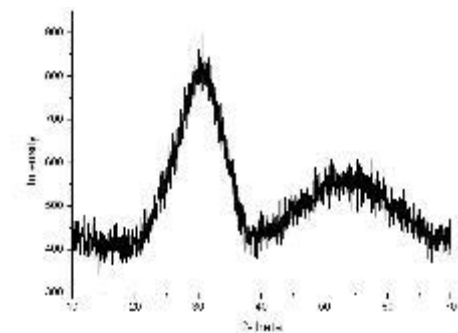
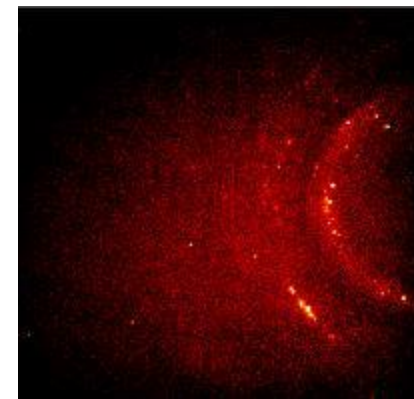
### 3. Behavior of Samples in experiment

U, Th are included in these samples, just their contents aren't the same. Even though their initial state show some common phenomena:

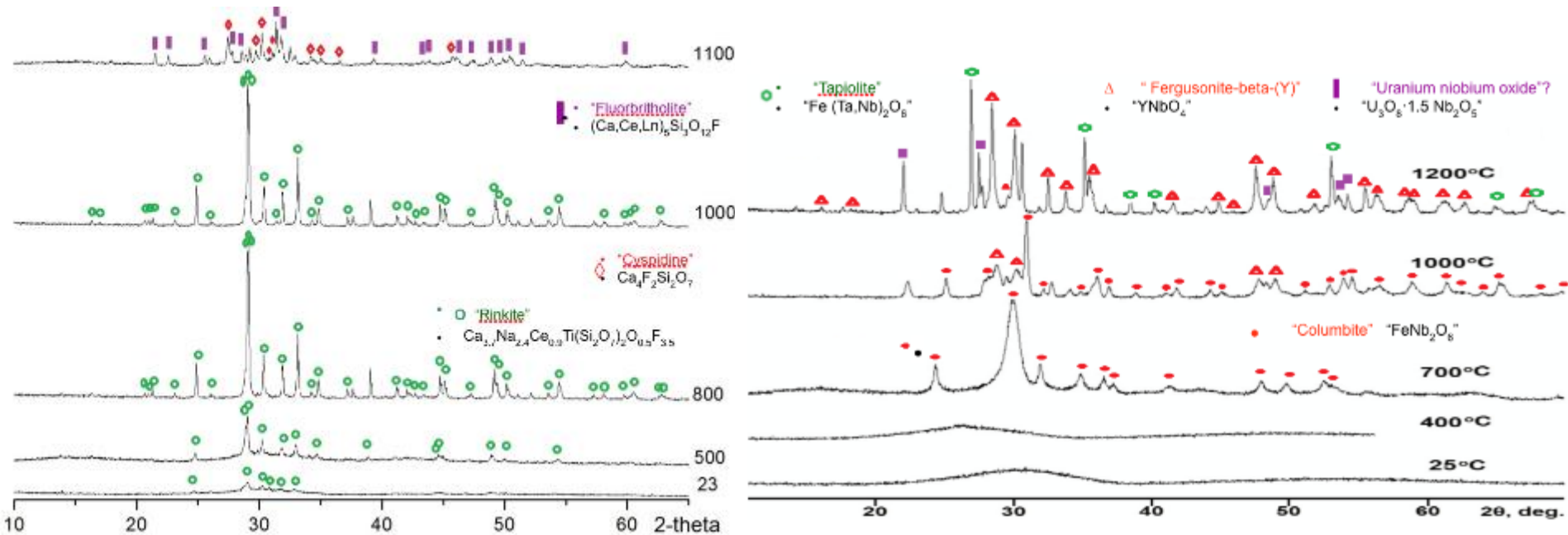
SEM Micrograph —Homogeneous matrix



The diffraction pattern



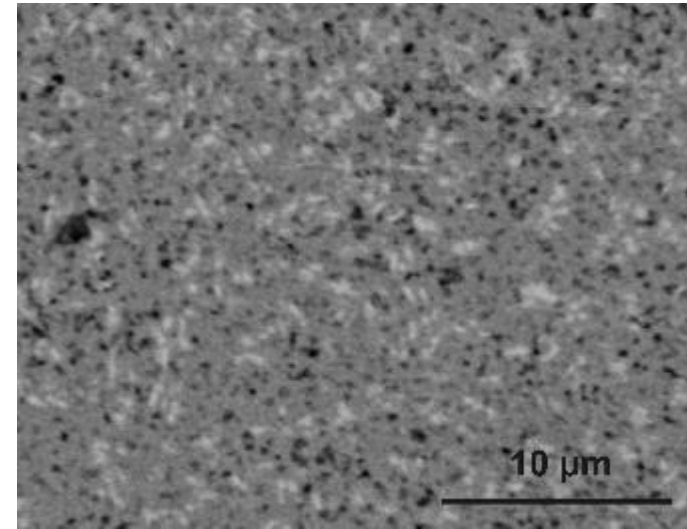
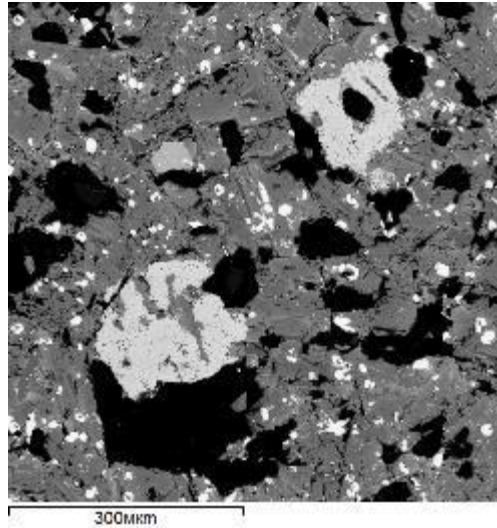
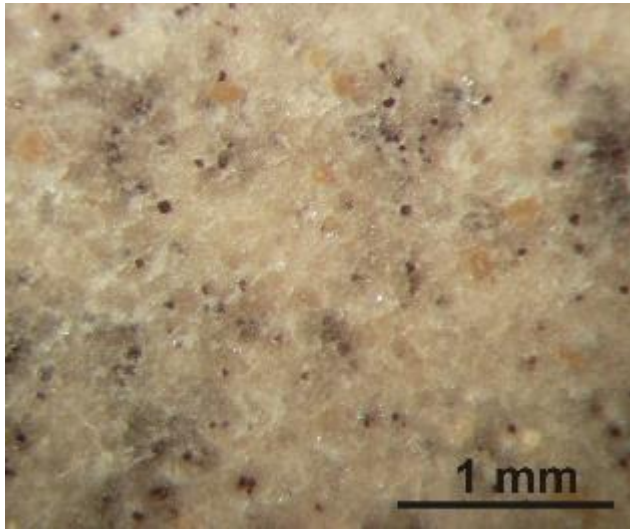
### 3. Behavior of Samples in experiment



After annealing, Recrystallization of samples are appeared



### 3. Behavior of Samples in experiment

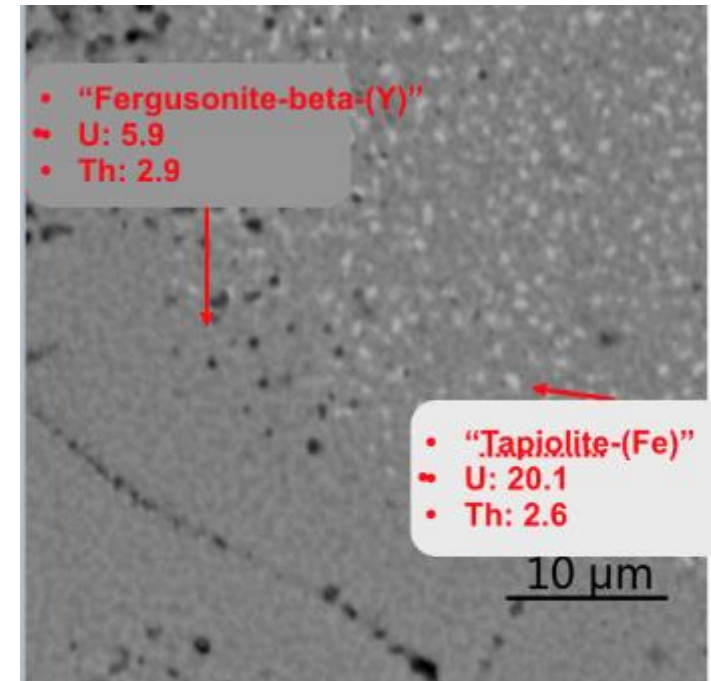
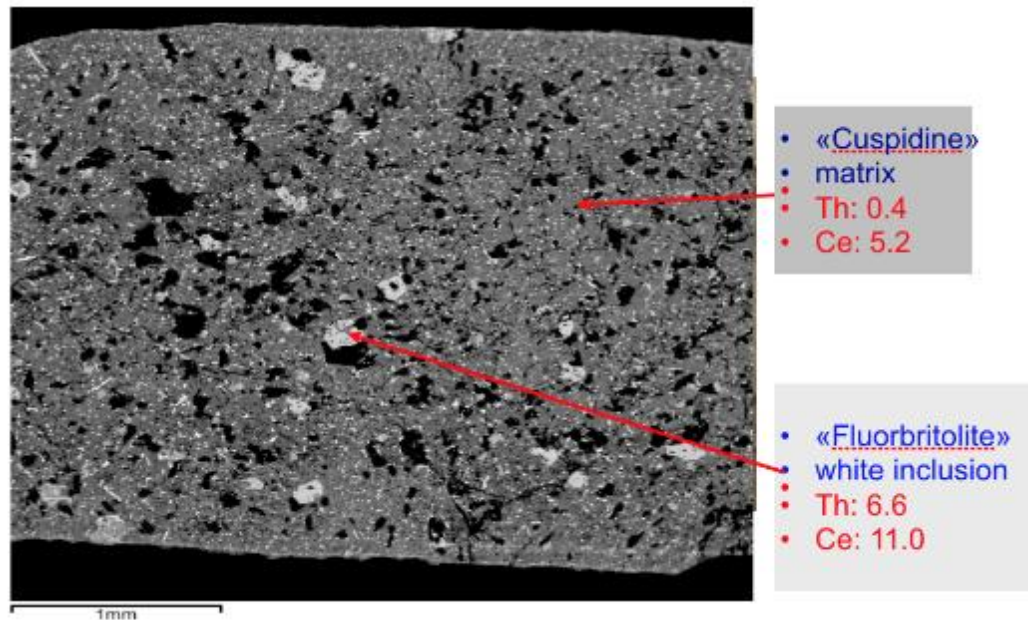


After annealing, New faces are formed

What kind of useful information can we get from this observation?

### 3. Behavior of Samples in experiment

After annealing, redistribution in different faces are found.



When condition is changed, do some radioactive elements escape from these solids  
(as a result of destruction of solid solution)?

## Conclusion

- 1) Studying metamict mineral is a possibility to know the behavior of actinide ceramics over long time (under chemical alteration and radiation damage)
- 2) The use of such samples in comparison with artificial samples is an optimal way to develop the suitable crystalline forms for actinide immobilization.
- 3) Reformed phase and the redistribution radionuclides in solids by changing environment by activity is important for making sure about the stability of actinide immobilization, or we can say, it help us to understand the synthesis actinide ceramic waste forms.

Therefore, we are looking for more suitable samples for investigation

# Thank you for your attention!



Thank you for supporting by ICTP & NRE1508!