

Thermodynamic study of (U-Pu-Zr-O-Fe) corium system

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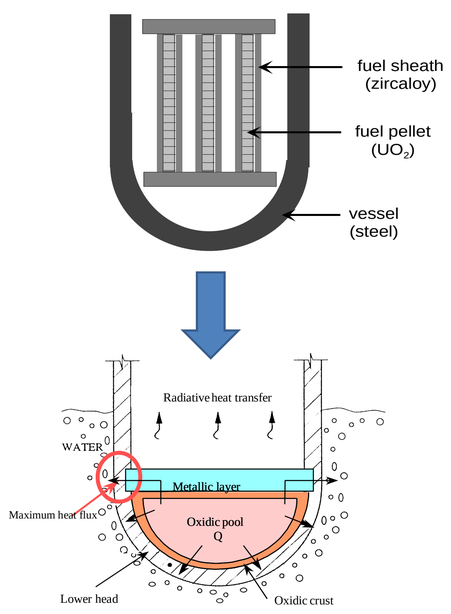
Thermodynamic sudy of the (U-Pu-Zr-O-Fe)

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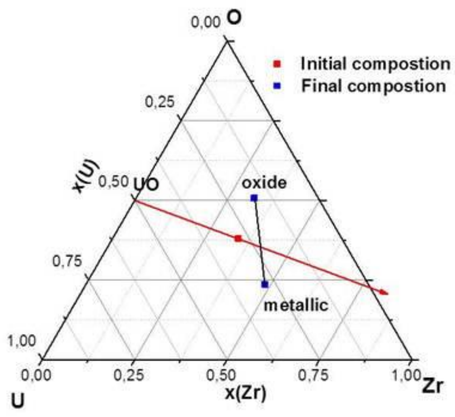
CONTEXT AND PURPOSE OF THE WORK

In case of severe accident in a pressurized water reactor (PWR), the nuclear oxide fuel ($UO_{2\pm x}$ or $(U,Pu)O_{2\pm x}$) could react with the Zr-alloy cladding and subsequently with the stainless steel vessel to form a mixture of solid and liquid phases, designated as "**corium**". The high temperature thermodynamic properties of this complex mixture have to be properly established to predict the corium behaviour during a severe accident. The approach is to develop a thermodynamic database using Calphad method on the (**U-Pu-Zr-O-Fe**) system that is representative for $UO_{2\pm x}$ or $(U,Pu)O_{2\pm x}$ fuel, Zr-alloy cladding and steel vessel. A critical analysis of the available experimental data was first performed. Then an experimental work was carried out to provide new phase diagram data in U-Zr-O, Fe-Zr-O, UO_2-PuO_2-ZrO_2 sub-systems. Heat treatments in the U-Zr-O and Fe-Zr-O systems allowed for highlighting the existence of miscibility gaps in the liquid state in both systems. The advanced laser heating setup developed in ITU was used to measure temperatures of solid/liquid transitions in the UO_2-PuO_2-ZrO_2 sub-system. All the experimental data together with literature data were used to improve the thermodynamic modelling of the (U-Pu-Zr-O-Fe) system. Both experimental and calculated results will be presented on the different corium systems.

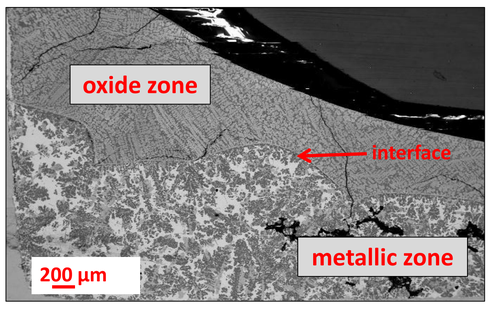


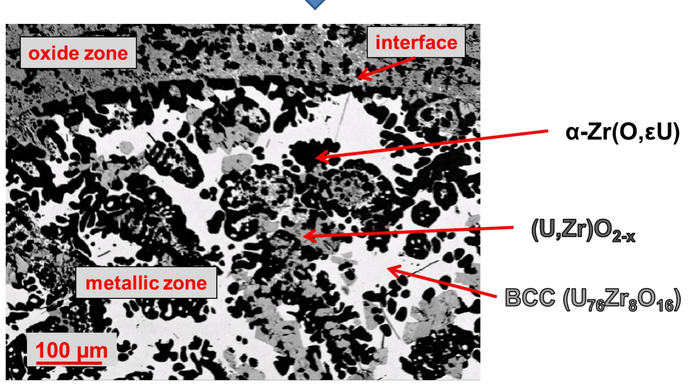
MISCIBILITY GAP IN THE U-ZR-O SYSTEM

- 4 samples (mixtures of U, Zr, ZrO_2) heated at 2567 K for 45 minutes in W crucibles ➡ 2 metallic and oxide liquids
- Characterization using SEM, EDS, WDS ➡ experimental tielines

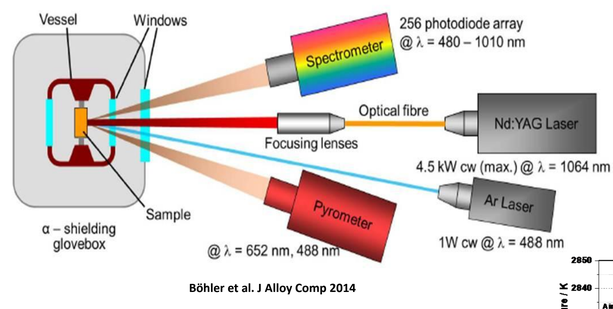


Evolution of the sample overall composition due to « UO » vaporization



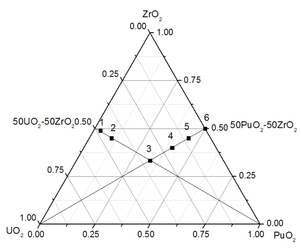


SOLIDIFICATION TEMPERATURE IN UO_2-ZrO_2-PuO_2 SYSTEM

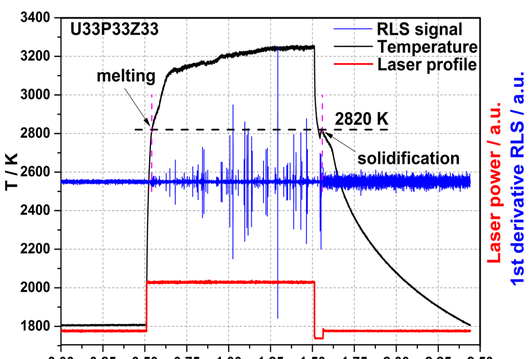


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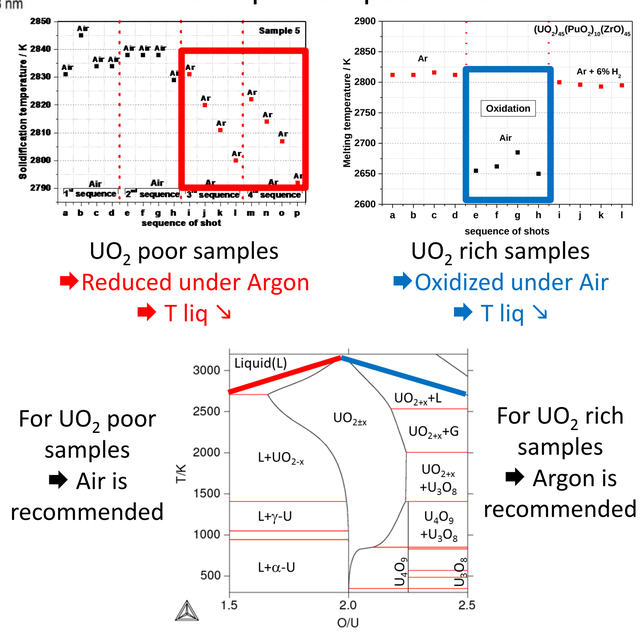
Laser heating setup in ITU (Karlsruhe)



Sample compositions



Thermogram of sample 3 under 0,3 MPa Argon



UO_2 poor samples
➡ Reduced under Argon
➡ T liq ↘

UO_2 rich samples
➡ Oxidized under Air
➡ T liq ↘

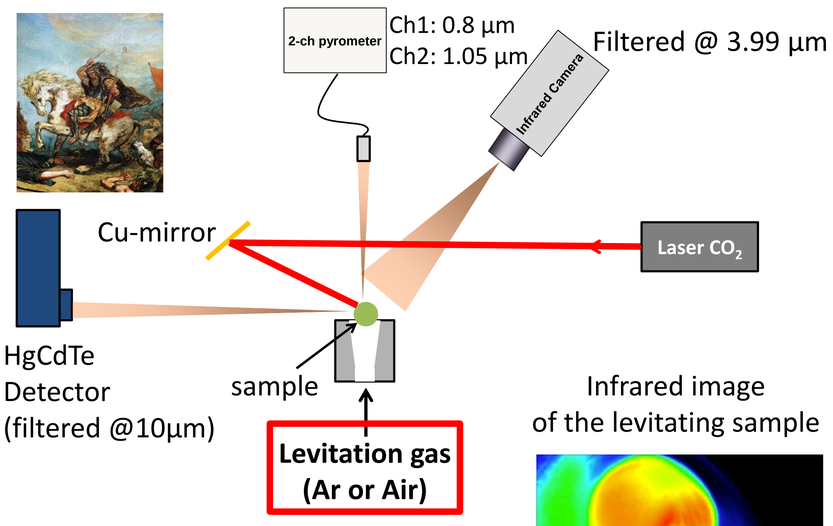
For UO_2 poor samples ➡ Air is recommended

For UO_2 rich samples ➡ Argon is recommended

Influence of the atmosphere

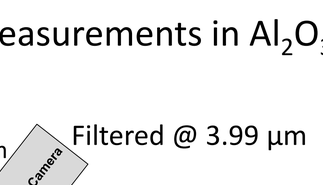
ATTILHA SETUP DEVELOPMENT

- Phase transition temperature measurements in Al_2O_3-ZrO_2 system (10 mol. % ZrO_2)

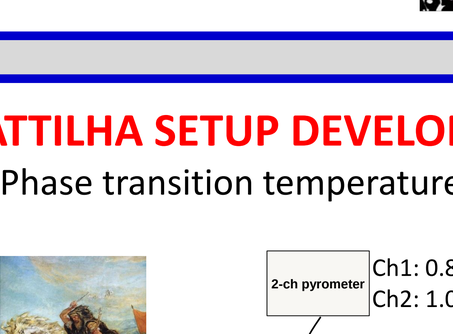


Infrared image of the levitating sample

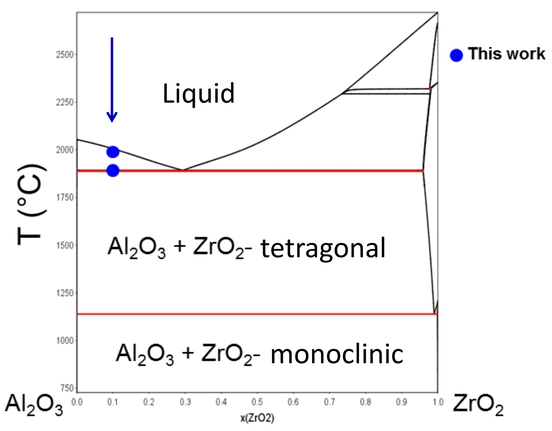
During heating
➡ Melting

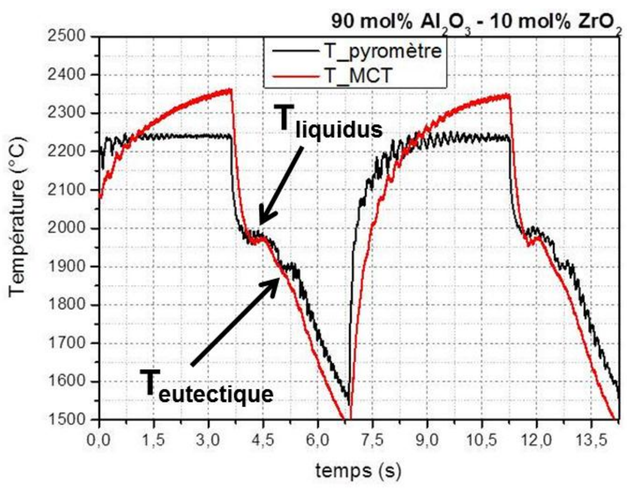


During cooling
➡ Solidification



Microstructure of the quenched sample





Thermogram under Argon

CONCLUSION

➡ **New experimental results on corium**

- Tielines in liquid miscibility gap of U-Zr-O, U-Zr-O-Fe systems
- Phase transition temperature measurements in U-Zr-O, Fe-Zr-O and UO_2-PuO_2-ZrO_2 systems (ITU)
- Development of the ATTILHA new laser heating setup

➡ **Improvement of the thermodynamic modelling**

- UO_2-PuO_2-ZrO_2 system
- U-Pu-Zr-Fe-O system

➡ **Application calculations for severe accidents**

- Solidification paths to interpret the quenched microstructure
- Corium/concrete interaction (using TAF-ID database for concrete systems)