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Experimental and calculation investigation on severe accidents in PWR reactors

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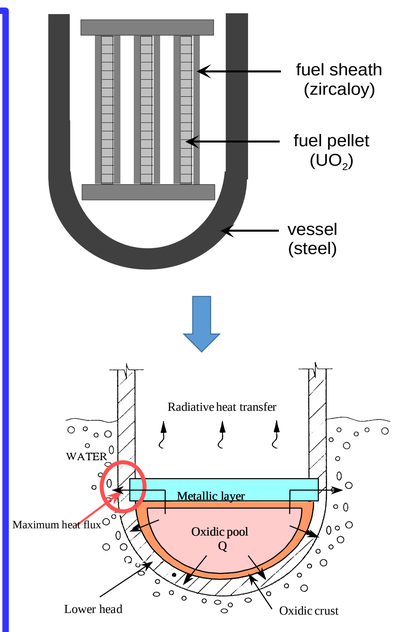
[2] *European Commission, Joint Research Centre (JRC), Safety of Nuclear Fuel Unit*

CONTEXT AND PURPOSE OF THE WORK

During a severe accident in a PWR reactor, the oxide nuclear fuel (UO_2 or $(U,Pu)O_2$) reacts at high temperature with the zirconium alloy clad the steel control rods cladding, the neutronic absorbers (Ag-Cd-In, B_4C) and the Inconel spacer grids leading to the formation of the so-called "In-vessel corium" – a mixture of solid and liquid phases – in the lower-head of the reactor vessel. Depending on several parameters, such as the U/Zr ratio and the amount of dissolved steel, the corium forms a pool in the bottom of the vessel, constituted of two liquid phases, metallic and oxide. The formation of such a configuration of the corium pool is due to the existence of a miscibility gap in the liquid state. The fractions and compositions of these liquid phases have to be well known in order to model the thermal hydraulic properties of the pool.

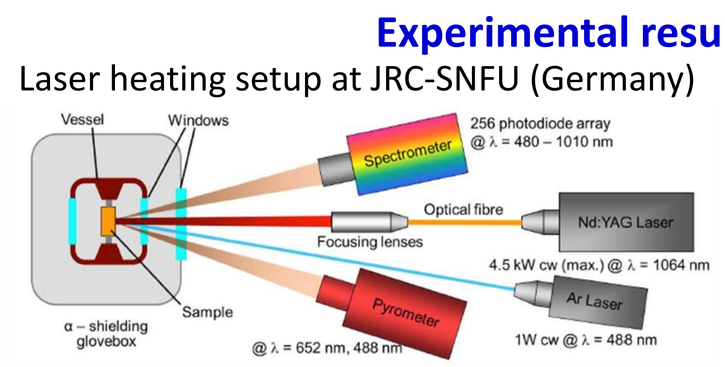
The aim of the present work is to experimentally investigate the thermodynamic properties of the U-Pu-Zr-Fe-O system representative for the in-vessel corium to improve the thermodynamic modelling using the Calphad method. It has been shown experimentally that the oxygen potential strongly influences the liquidus/solidus curves when a UO_2-rich mixture is considered. By thermodynamic calculations it is possible to show that Pu preferentially segregates in oxide phases regardless of the temperature.

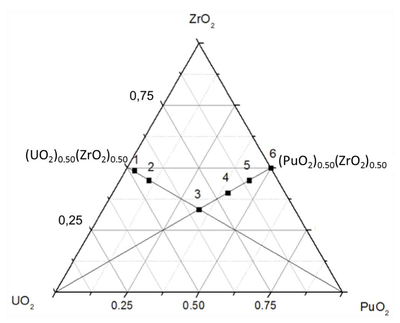
In parallel a novel experimental setup has been developed to study prototypic corium at very high temperatures (T>2500K). This setup called ATTILHA is based on the laser heating technique coupled with aerodynamic levitation. It is adapted to investigate metallic and oxide system as well as oxide-metallic system. Recent results have been obtained on the miscibility gap of the Fe-Zr-O system.



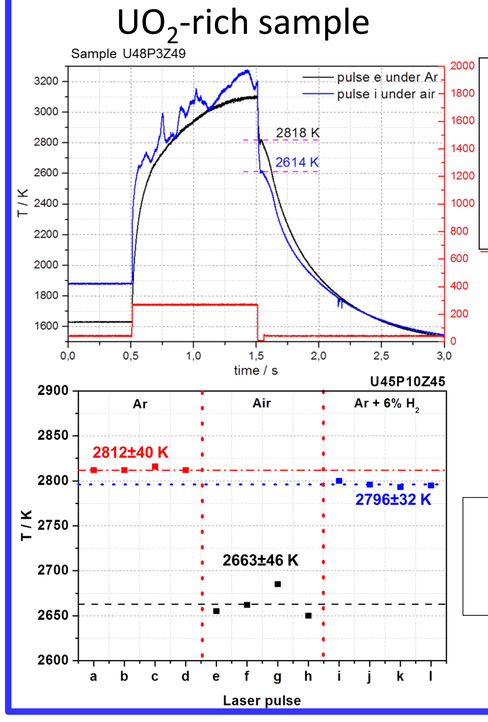
Experimental results [1]

Laser heating setup at JRC-SNFU (Germany)





UO_2-rich sample

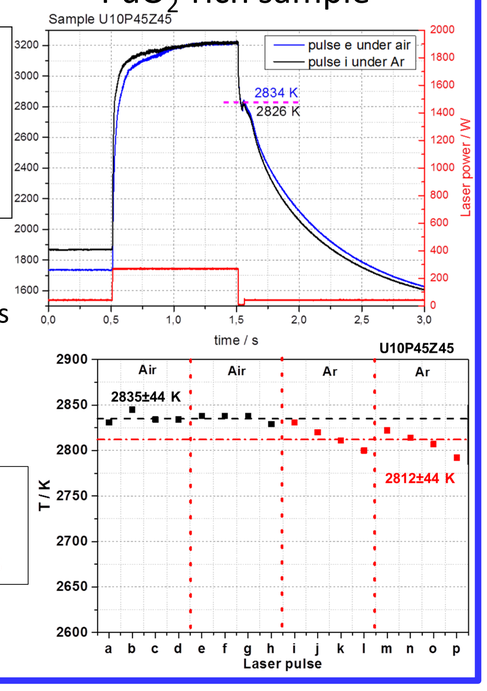


Experimental atmosphere has an effect on the melting behavior of UO_2-rich samples

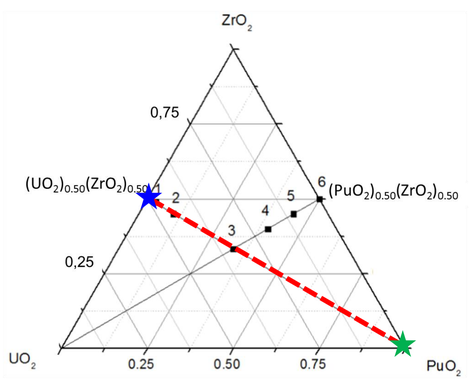
O/(U+Pu+Zr) ratio moves towards hyper-stoichiometry during experiments under air

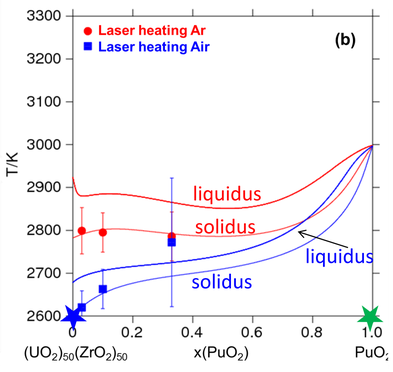
Very good repeatability regardless of the experimental conditions

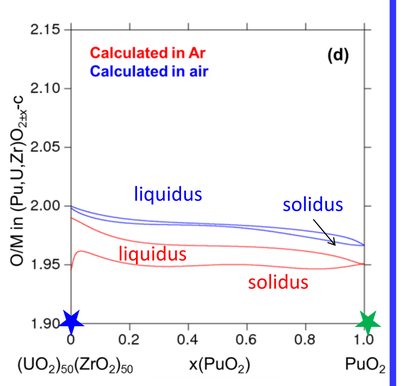
PuO_2-rich sample



Thermodynamic calculations [1]







Oxidising and Reducing conditions → good agreement with experimental data

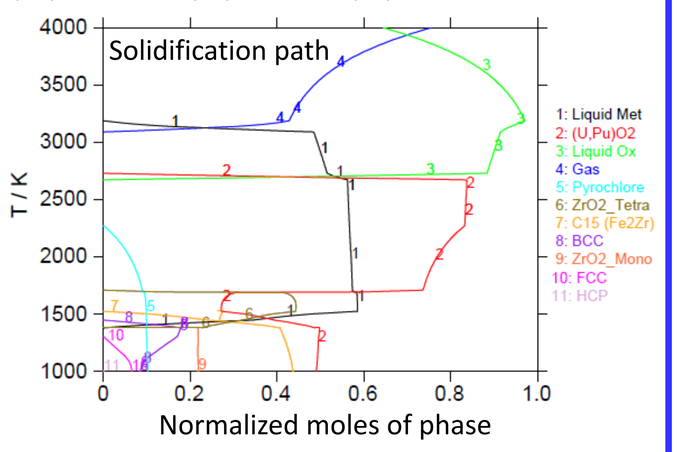
Oxidising conditions →$T_{solidus}$ ↑ adding PuO_2
→O/M in the $(U,Pu,Zr)O_{2\pm x}$ varies along solidus/liquidus depending on the atmosphere
→ PuO_2 is never stoichiometric along solidus/liquidus under air /Ar

Application calculation – interaction MOx+Zry+Steel

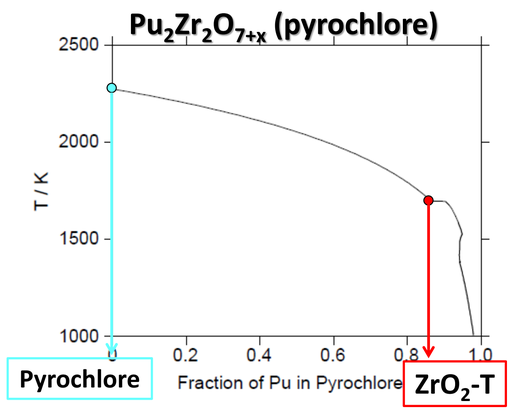
Composition of a prototypic PWR corium [2]

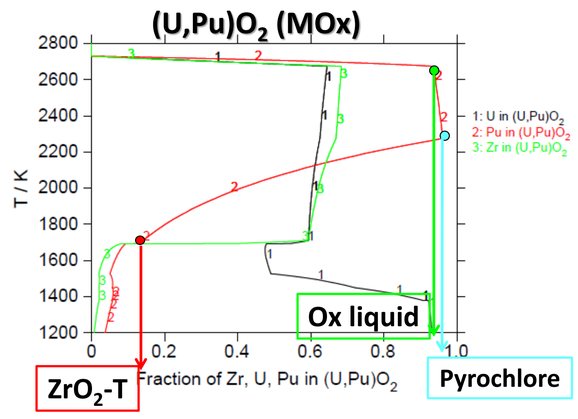
Component	wt %
$(U_{0.90}Pu_{0.10})O_2$	52.5
30% oxidized Zr	24.4
$Fe_{0,71}Cr_{0,18}Ni_{0,11}$	23.1

Conditions for the calculations:
Fuel + Zr: $m(PuO_2)$=5.25, $m(UO_2)$=47.25, m(Zr)=15.988, $m(ZrO_2)$=9.26
Stainless steel: m(Fe)=16.401 m(Ni)=2.541 m(Cr)=4.158



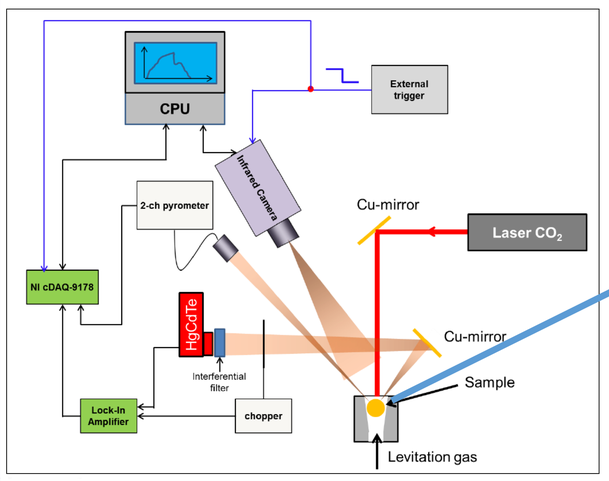
Formation of oxidized liquid: T=2673 K
Formation of metallic liquid: T=1381 K
Major low T condensed phases (T<1500 K):
- Metallic C15-Fe_2Zr, BCC (Cr,Ni), FCC (Ni,Fe)
- Oxide ZrO_2-Mono, $(U_{0.975}Pu_{0.005}Zr_{0.020})O_2$, **$Pu_2Zr_2O_{7+x}$ (pyrochlore)**



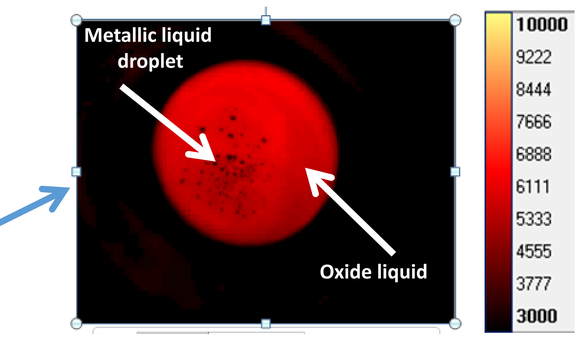


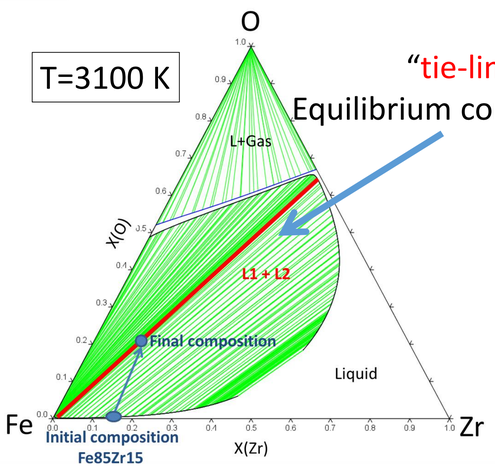
→ Relocation of Pu from MOx into $Pu_2Zr_2O_{7+x}$ during severe accident
→ Pu is mostly present in oxide phases: Oxide liquid, $(U,Pu)O_2$, Pyrochlore
→ High rate of evaporation of Pu ($Pu_{(g)}$, $PuO_{(g)}$) above 3200 K

ATTILHA setup

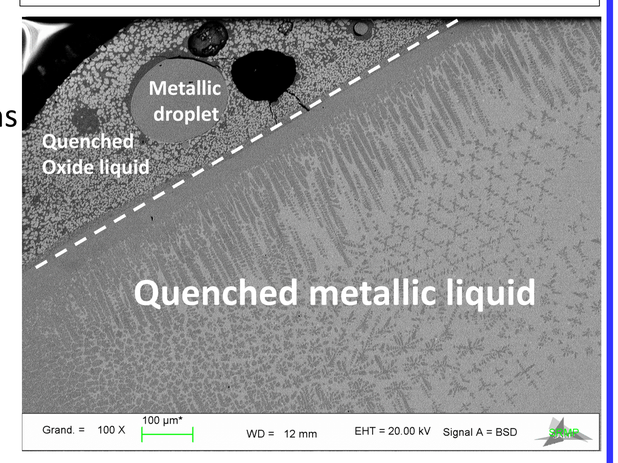


Infrared image of the levitating (Fe,Zr,O) sample under Ar





Microstructure of the quenched sample



→ In-situ observation of the formation of the miscibility gap in Fe-O-Zr

Conclusions

Experimental:

First systematic investigation of the influence of the experimental atmosphere on the high temperature behaviour of the UO_2-PuO_2-ZrO_2 system

Calculations:

Pu preferentially segregates in oxide phases…**risk of re-criticality**?

ATTILHA:

Development of a novel experimental setup dedicated to high temperature measurements → **in-situ evidence** of the formation of the miscibility gap in the Fe-Zr-O system

[1] Quaini et al. JNM 467 (2015) 660-676
[2] Steinwarz & Sappok, Nucl. Technol. 125 (1999) 363-370