



20th DISCUSSION MEETING ON THERMODYNAMICS OF ALLOYS
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BLED - SLOVENIA

Investigation of the metal-oxide part of U-Pu-O system at high temperature

Caroline Denier^{1,2}, Bénédicte Michel¹, Marc Barrachin¹, Luka Vlahovic²

¹Autorité de Sûreté Nucléaire et de Radioprotection (ASNR), Cadarache, France

²European Commission, Joint Research Centre (JRC), Karlsruhe, Germany



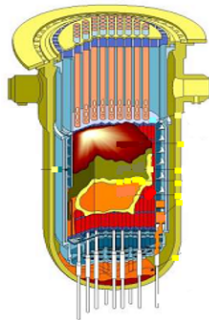
Context

MOX fuels behaviour under severe accident conditions

MOX fuel = Mix of uranium and plutonium oxide $(U,Pu)O_2$

Enable partial re-use of the nuclear fuel

Used in French 900 MWe reactors, planned extension 1300 MWe reactors



TMI-2 vessel final state

Severe accident conditions



temperature increase $> 2500\text{ K}$



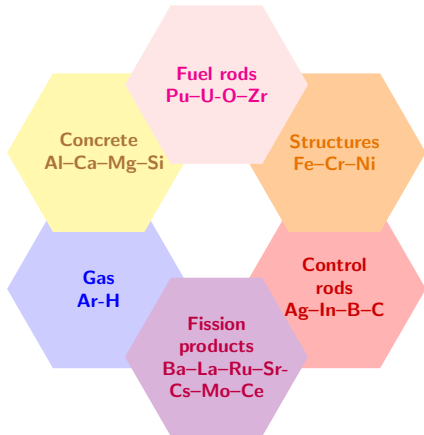
corium formation

Distribution of Pu between different phases in the corium ?

NUCLEA database

a **non-ideal** thermodynamical database for in-vessel and ex-vessel corium applications

NUCLEA-26



- ▶ 24 chemical elements,
- ▶ developed using the CALPHAD method,
- ▶ covering all compositions ranging from the metallic to the oxidic state (all binary and pseudo-binary systems assessed, 44 ternary systems fully modeled and 27 sections derived from quaternary systems modeled, higher systems extrapolated),

but data are very scarce for some systems

U-Pu-O ternary system

- Demixing of U-Pu-O corium into metallic and oxide phases

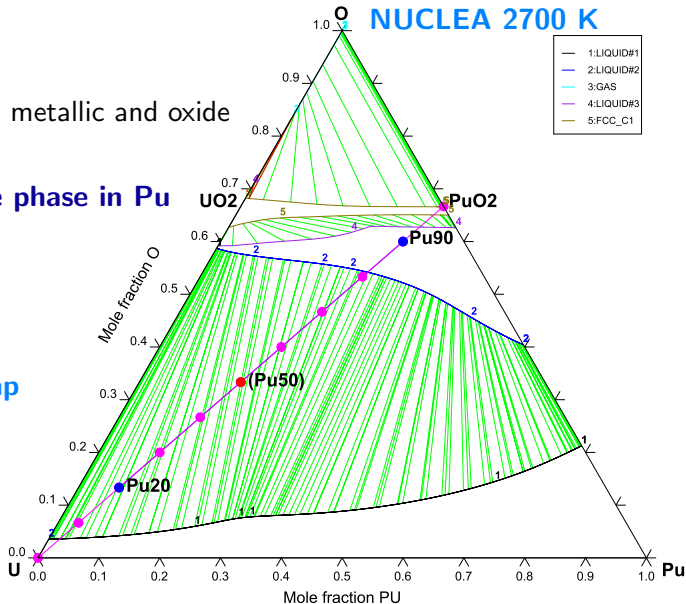
⇒ possible enrichment of one phase in Pu

⇒ risk of return in criticality

- no experimental data in the metal-oxide part of the system

⇒ extent of the miscibility gap purely hypothetical

⇒ Melting temperature measurements on the U-PuO₂ line by laser flash method



Samples production

mix U and PuO_2 powders

press into pellets

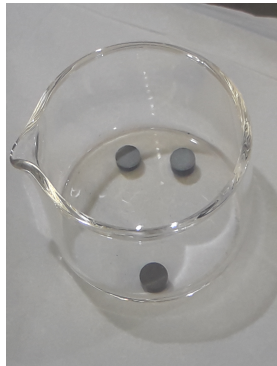
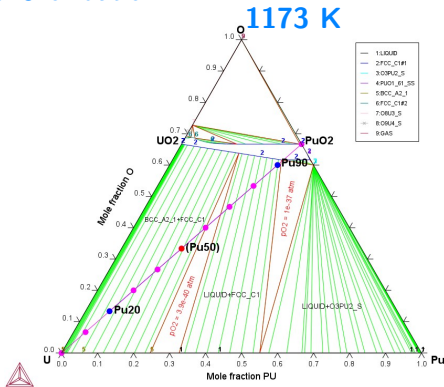
sintering at 1173 K

- ▶ U powder (produced by hydride-dehydride of a U metal plate)
- ▶ under purified argon $\Rightarrow$ avoid U oxidation

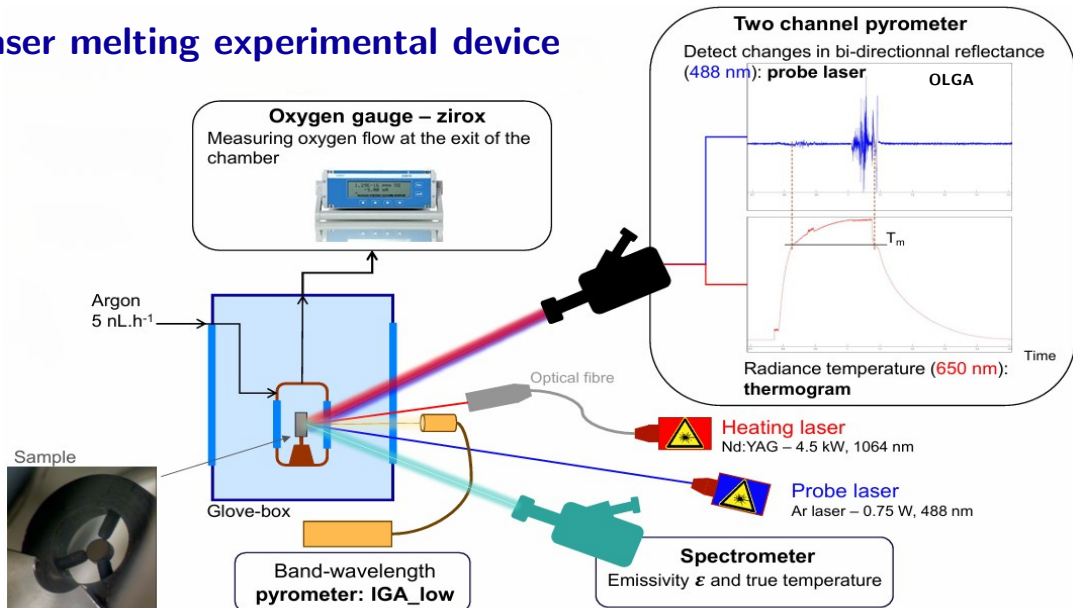
$\Phi \sim 5 \text{ mm}$
 $h=1-2 \text{ mm}$

	Pu90	Pu20
at-% U	10	80
at-% Pu	30	6.67
at-% O	60	13.33
Pu/(U+Pu)	0.75	0.08
O/M	1.5	0.15

$\Rightarrow$ Pu50 not possible (friable sample, especially after sintering (swelling))



Laser melting experimental device



Pu90 Results



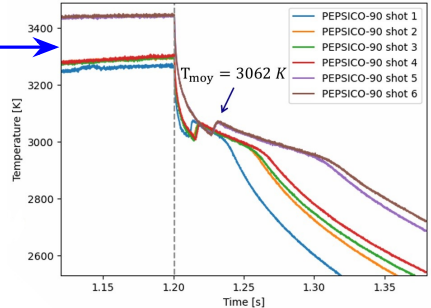
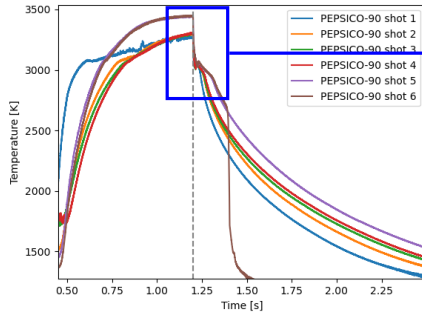
Before



Shot 3



Shot 6

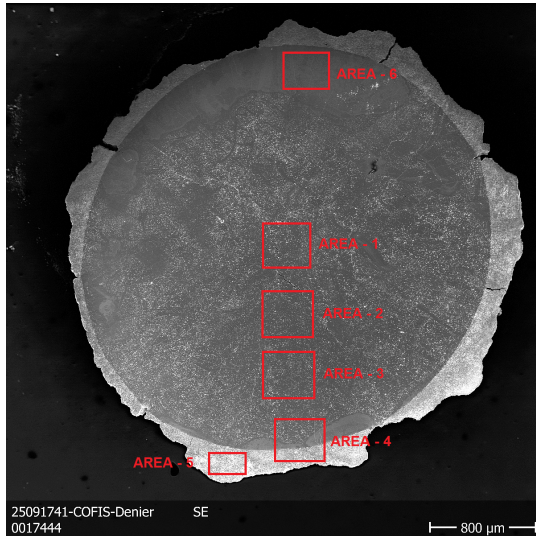


- ▶ collapse of the molten zone after cooling (shot 6)
- ▶ sample failure during shot 7
- ▶ oxidation during laser shots

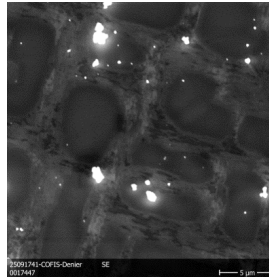
- ▶ Olga pyrometer
- ▶ $\epsilon = 0.7$ measured by spectrometer

⇒ **a reproducible thermal event**
(undercooling ⇒ liquidus)

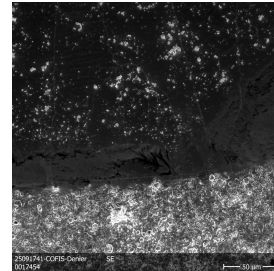
Pu90 SEM Measurements



Area 1 (2,3)



Area 4



Area	Pu / (U+Pu)	O/M
<i>Theoretical</i>	0.75	1.5
Sintered sample	0.82 ± 0.06	0.15
1, 2, 3 – molten pool	0.83 ± 0.06	0.20
4, 5, 6 – not molten	0.61 ± 0.03	0.28

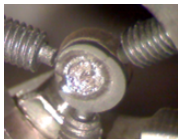
Pu20 Results



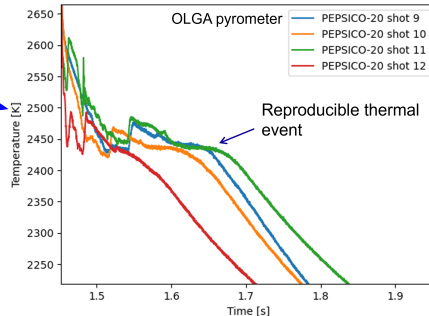
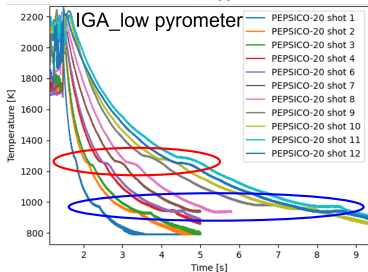
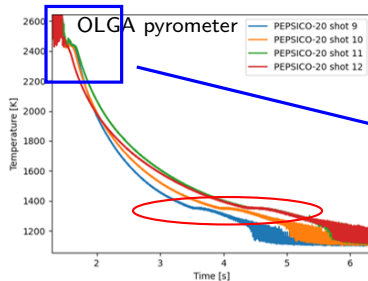
Before



Shot 8



Shot 12

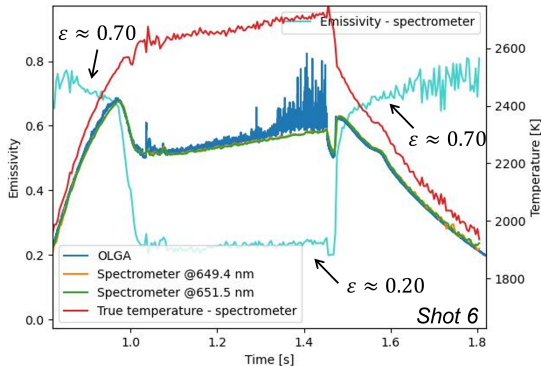


- ▶ radiance temperatures
- ▶ oxidation during laser shots

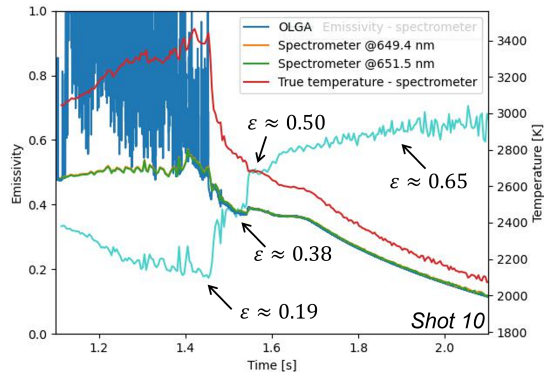
⇒ **3 reproducible thermal events (plateaux)**

Pu20 Emissivity

ϵ drop at melting



several changes in ϵ during cooling



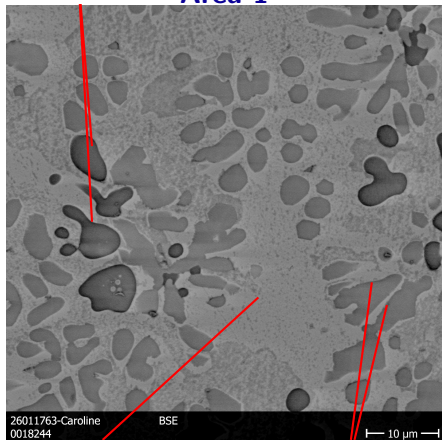
⇒ a thermal event seems to be linked to emissivity change

⇒ emissivity measurements crucial for oxide-metal experiments

Pu20 SEM Measurements

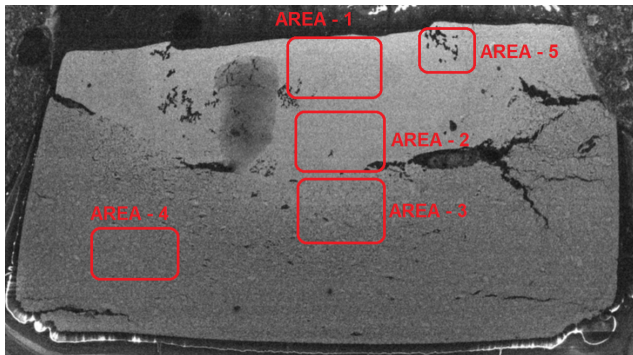
Pu 0.14
O/M 0.64-0.67

Area 1



Pu 0.09-0.11
O/M 0.15

Pu 0.06-0.03
O/M 0



Area	Pu / (U+Pu)	O/M
<i>Theoretical</i>	0.08	0.15
Sintered sample	0.16 ± 0.01	0.24
1, 2, 5 – molten pool (MP)	0.10 ± 0.01	0.22
3, 5 – surroundings of the MP	0.11 ± 0.01	0.53
4 – not molten	0.07 ± 0.01	0.66

Summary and conclusions

Pu90	Pu20
one liquid	two liquids
a reproducible thermal event: 3062 K ($\epsilon = 0.7$) (undercooling $\Rightarrow$ liquidus)	3 thermal events (plateaux): 2600 K ($\epsilon = 0.57$), 1497 K ($\epsilon = 0.2$) and at lower temperature
oxidation during laser shots	oxidation during laser shots

SEM analysis still ongoing (oxygen content difficult to quantify)

- Composition of the sintered sample ? Composition of the initial melt ? Composition of the different phases formed during thermal events ?
- origin of oxidation during laser shots : diffusion from other parts of the sample ? evaporation of metallic species ? oxygen from argon flow ?

Use of the NUCLEA database planned for data interpretation :

identify the expected phases, determine the solidification path, relate the calculated phase evolution to the experimental observations...

Summary and conclusions

Laser flash method quite useful for high temperature measurements :

- ▶ no crucible $\Rightarrow$ no contamination
- ▶ short temperature peaks $\Rightarrow$ limited evaporation (high pressure cell also possible)
- ▶ spectrometer $\Rightarrow$ confidence in temperature measurements (essential for metal-oxide tests)

BUT experiments difficult to interpret :

- ▶ composition of the initial melt still difficult to determine, especially oxygen content
- ▶ which phase transition corresponds to each thermal event ? what is the composition of the phases at the corresponding temperature ?

