Investigation of U-Zr-O and Fe-Zr-O systems by a laser heating approach

A. Quaini, C. Gueneau, S. Gosse, D. Manara

DE LA RECHERCHE À L'INDUSTRIE

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DE LA RECHERCHE À L'INDUSTRIE

Context – PWR Severe Accident

Out of nominal functioning of a **P**ressurized **W**ater **R**eactor may lead to a nuclear accident

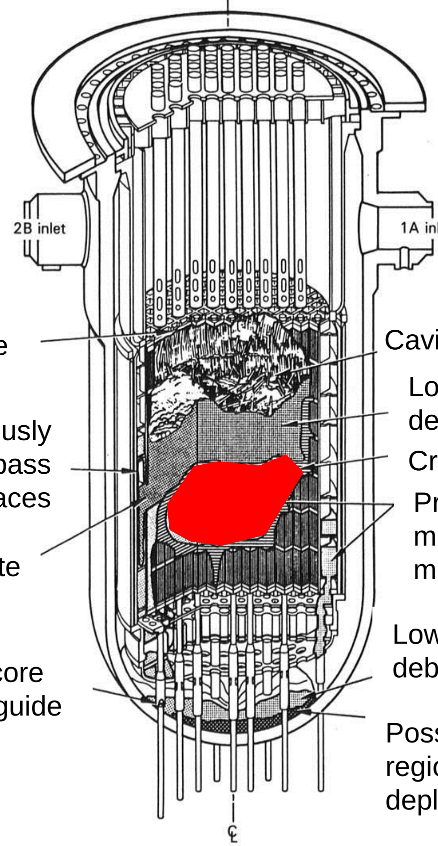
↓

Severe accident (SA): the normal functioning of the nuclear reactor is not re-established

↓

Fusion of the core and internal structure

TMI-2



PWR cross section

Interactions:

MOx + Zircaloy + stainless steel + Inconel + B_4C + Fission Products

In vessel corium composition:

U-Zr-O-Fe-Cr-Ni-Ag-Cd-In-B-C-FPs-… (basically most of the period table)

"Prototypic" in-vessel corium system: U-Zr-Fe-O

In-vessel core Melt configuration

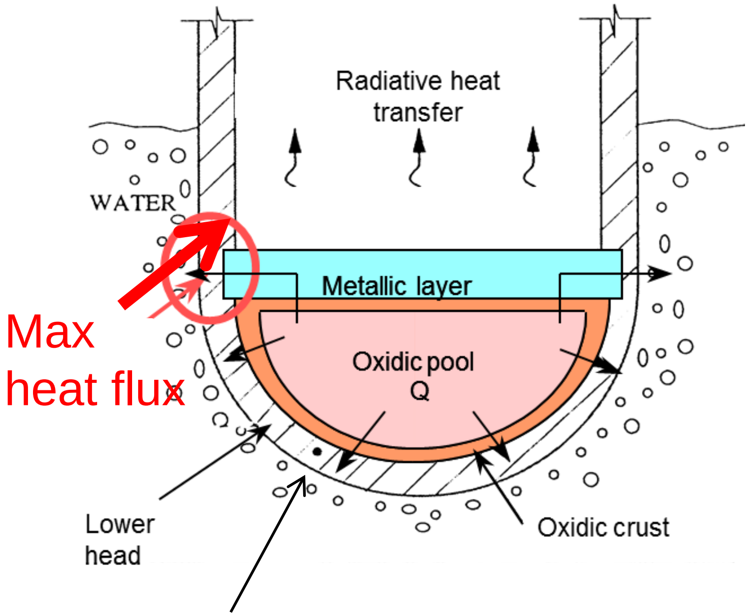
Issue : In-vessel core Melt configuration (Miscibility gap in the in-vessel corium liquid)

$\rho_{metallic} < \rho_{oxide}$ →

- Degree of oxidation of zirconium
- U/Zr ratio
- Amount of molten steel

→ $\rho_{heavy\ metallic} > \rho_{oxide}$

ρ =density



Heavy metal layer below the oxidic pool

Seiler et al., Nucl. Eng. Des. 2007

Decrease of the thickness of the upper metallic layer

- Enhanced focusing effect due to high radiative fluxes
- **Failure of the reactor vessel**

⇓

Knowledge of the chemical composition of liquids
$T_{solidus}$ and $T_{liquidus}$ are paramount for density calculations

Experimental technique

Sample preparation

Experimental results

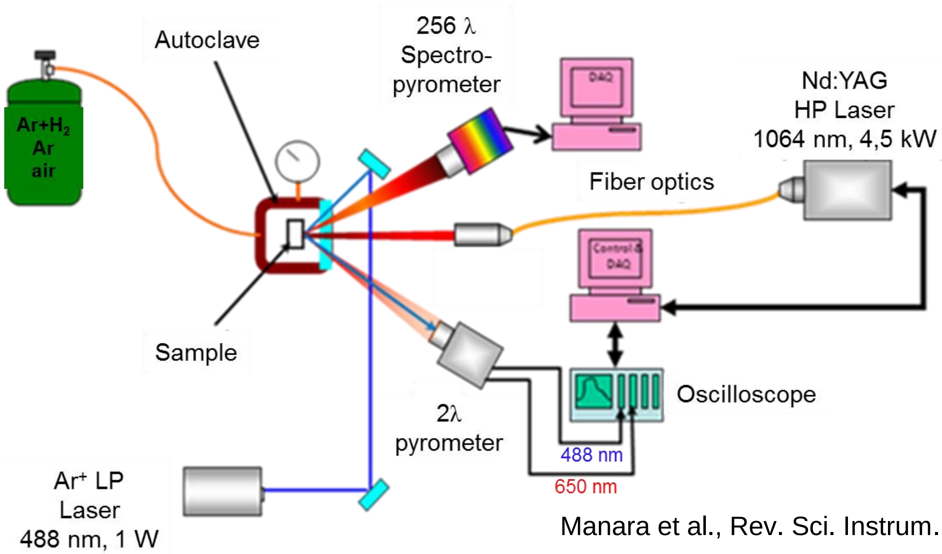
Conclusions

Experimental technique - Laser heating setup

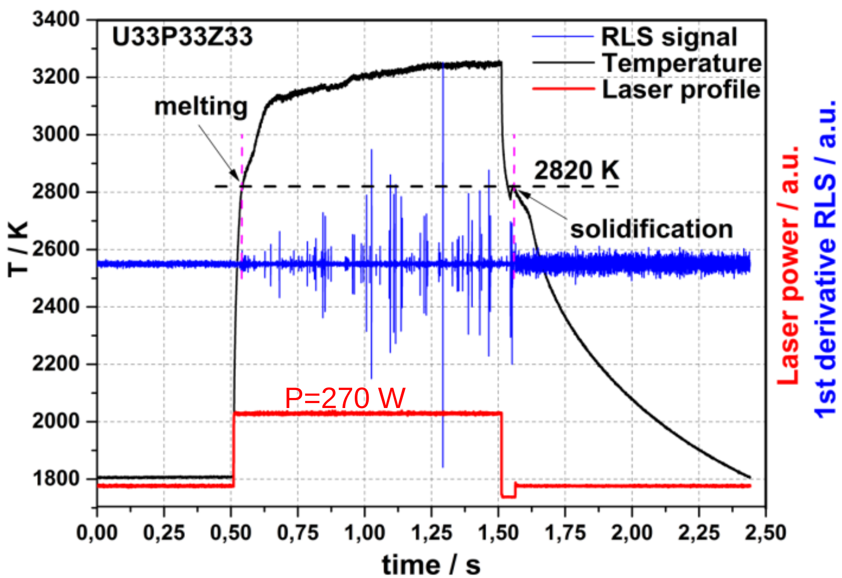
→Rapid measurements
→High power → High temperature
→Containerless
→Reducing/oxidising atmosphere

→Oxygen potential is not controllable

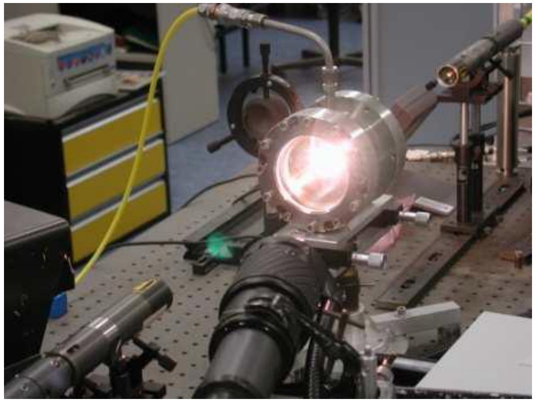
JRC-SNFU's Laser heating setup



Manara et al., Rev. Sci. Instrum. 2008



Quaini et al. JNM 2016



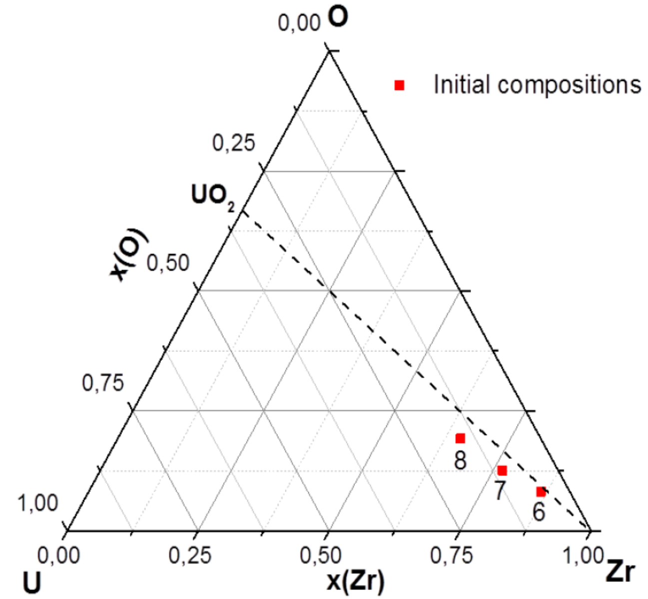
De Bruycker, PhD thesis 2010

Sample preparation

Sample preparation – U-Zr-O samples

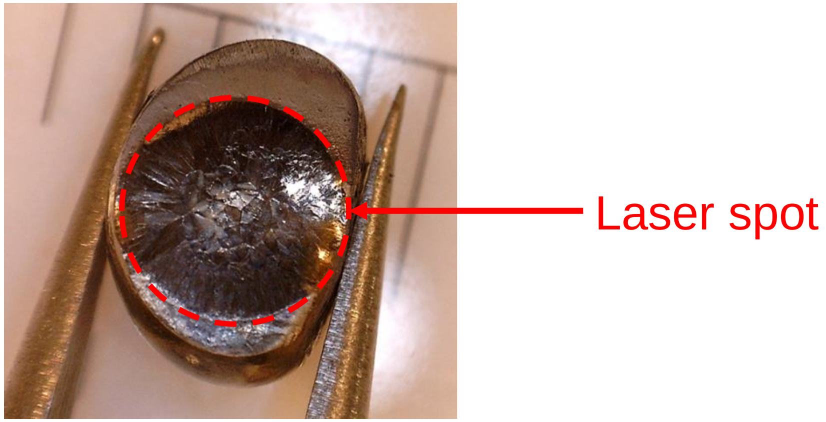
Sample	at% O	at% U	at% Zr	mass / g	Δm /g	Δm /m %
OUZr_6	8.2	5.5	86.3	1.2196	0.0002	0.016
OUZr_7	12.6	10.6	76.8	1.0384	0.0017	0.164
OUZr_8	19.2	15.3	65.5	0.9636	0.0013	0.135

→Arc furnace
→U-met, Zr-met, powder of ZrO_2
→less than 100 ppm of residual O_2 in the furnace chamber
→mass loss after arc furnace melting is negligible



Before laser heating

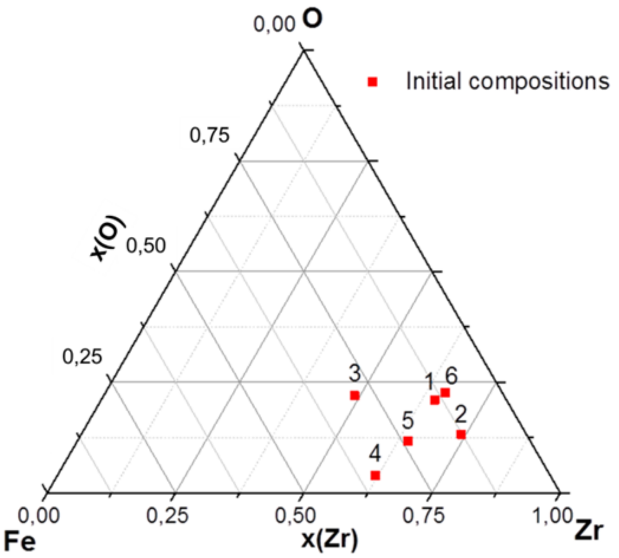
After laser heating



Sample preparation – Fe-Zr-O samples

Sample	at% O	at% Fe	at% Zr	mass / g	Δm / g	Δm/m %
OFeZr_1	21.0	14.0	65.0	0.7806	0.0020	0.156
OFeZr_2	13.2	12.7	74.1	0.7644	0.0140	1.832
OFeZr_3	21.9	29.1	49.0	0.5559	0.0063	1.133
OFeZr_4	3.9	34.1	62.0	0.5801	0.0006	0.103
OFeZr_5	11.7	23.8	64.5	0.6772	0.0030	0.443
OFeZr_6	22.6	11.1	66.3	0.5463	0.0069	1.263

→Arc furnace
→Fe-met, Zr-met, powder of ZrO_2, powder Fe_2O_3
→less than 100 ppm of residual O_2 in the furnace chamber
→mass loss is significant for samples 2, 3 and 6

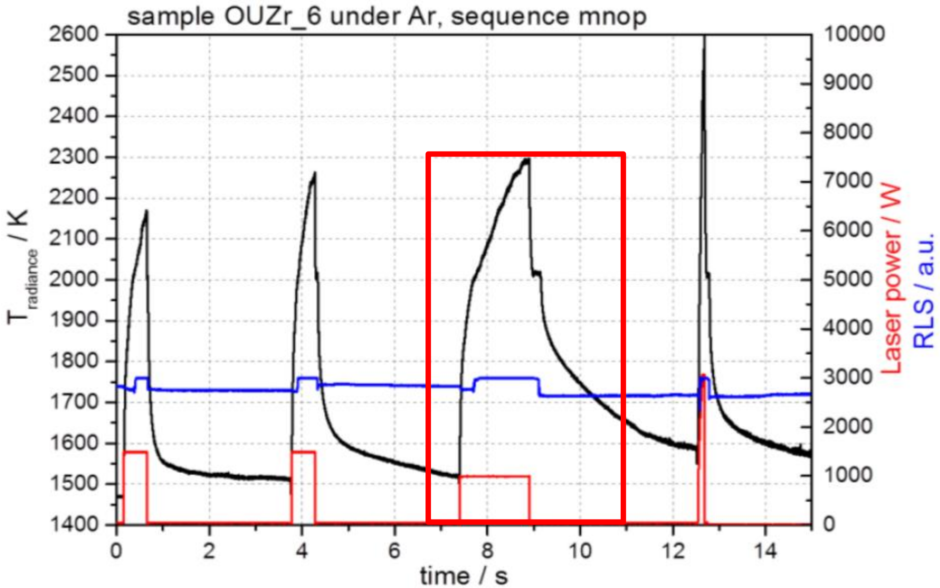


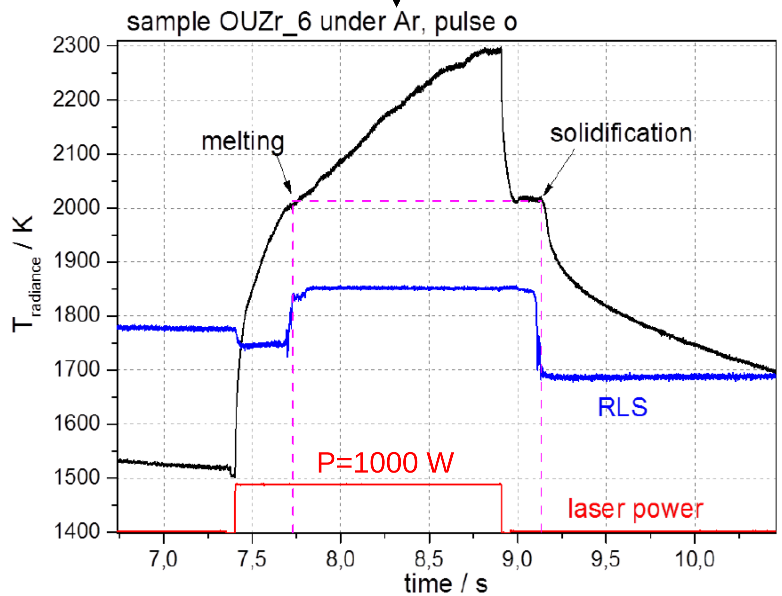
Experimental Results

Experimental results – U-Zr-O system

Sample 6: $U_{0.055}Zr_{0.863}O_{0.082}$ (Under Ar)
4 laser shots

Radiance temperature

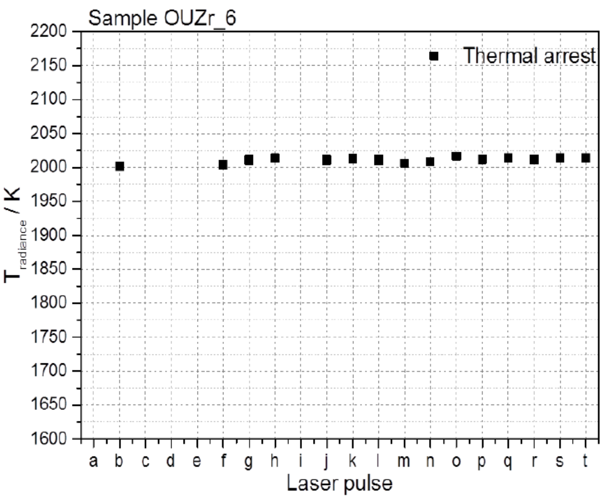


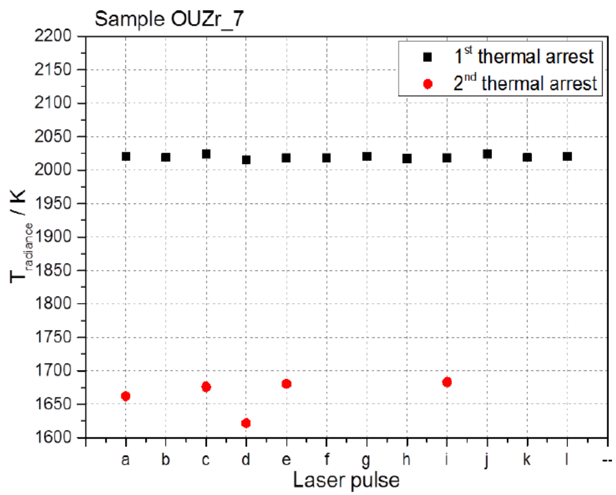


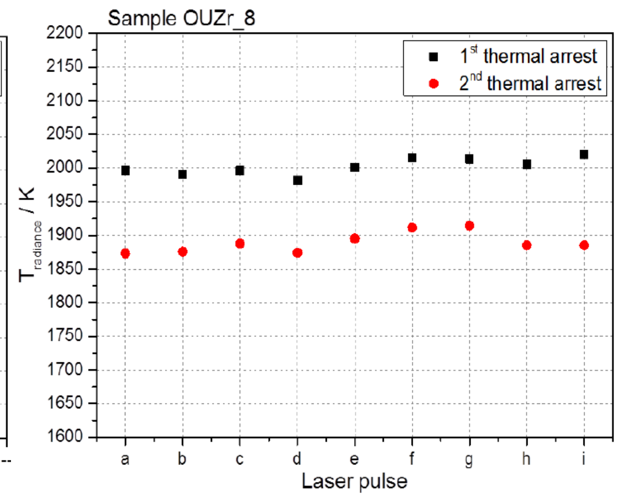
RLS: allows to detect changes in the reflectivity of the sample → solid/liquid
→ Good correspondence between melting and solidification

Emissivity is needed to convert radiance temperature into true temperature

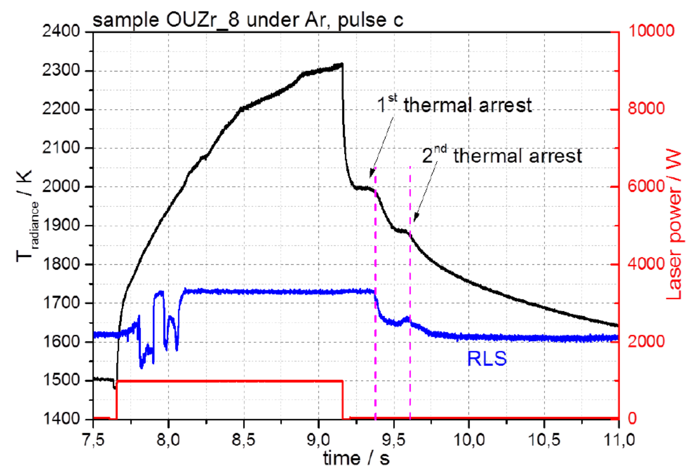
Experimental results – U-Zr-O system







Good repeatability especially on the first thermal arrest



Uncertainty on radiance temperature in less than 0.5 %

Sample	Radiance temperature / K	
	1st inflection	2nd inflection
OUZr_6	2011±11	//
OUZr_7	2020±10	1665±26
OUZr_8	2002±16	1889±18

…and the true temperature?

Experimental results – emissivity again??

What can we do without **any knowledge** of the functional relation between optical properties, wavelength and temperature and without any **experimental data** coming from the literature?

Sample	
	ε(650 nm)
OUZr_6	0.243±0.05
OUZr_7	0.264±0.05
OUZr_8	0.40±0.08

Our case:

~~combination of the metallic components: $\varepsilon_{sample}=A\varepsilon_U+B\varepsilon_{Zr}$~~ Good for dielectrics

~~Physical expression: $\varepsilon_{sample}=f(T,\lambda,\ldots)$ (Hagen Rubens, Drude, …)~~ Good for pure metals

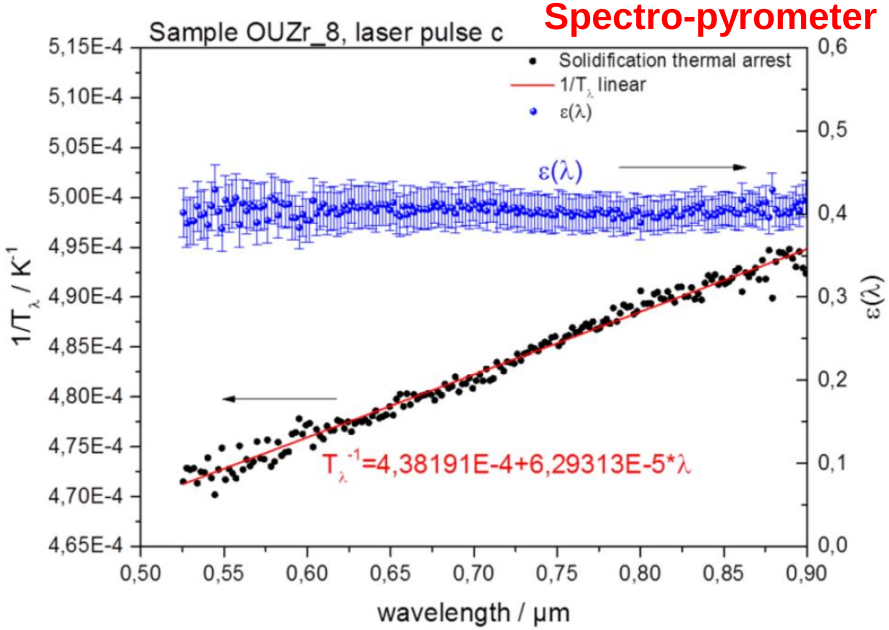
~~Polynomial $\varepsilon_{sample}=\sum_{i=0}^{n}\alpha_i\lambda_i$~~

Grey body: ε_{sample}=constant

$$\frac{1}{T_\lambda}=\frac{1}{T}-\frac{\lambda}{c_2}\ln(\varepsilon)$$

$$y=y_o+m*x$$

Spectro-pyrometer results



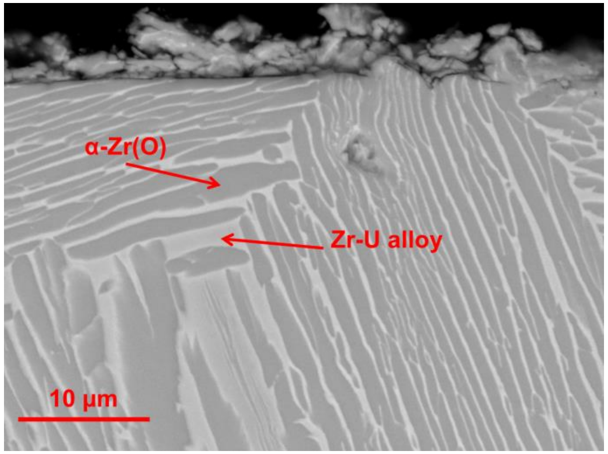
Experimental results – emissivity again??

Sample	
	ε(650 nm)
OUZr_6	0.243±0.05
OUZr_7	0.264±0.05
OUZr_8	0.40±0.08

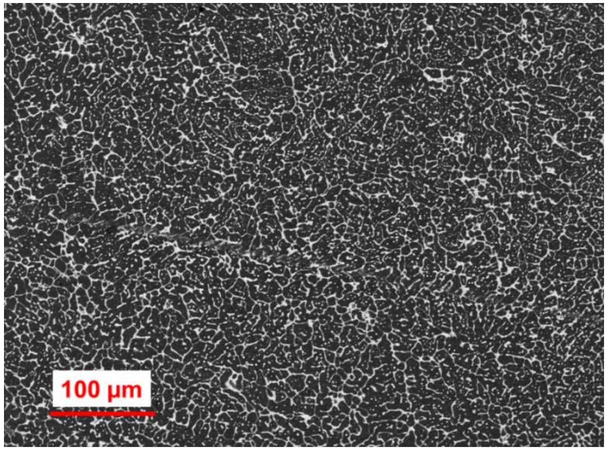
Significant difference between sample 6&7 and 8

Could be related to :

- the phases formed during solidification
- Applicability of the grey body assumption

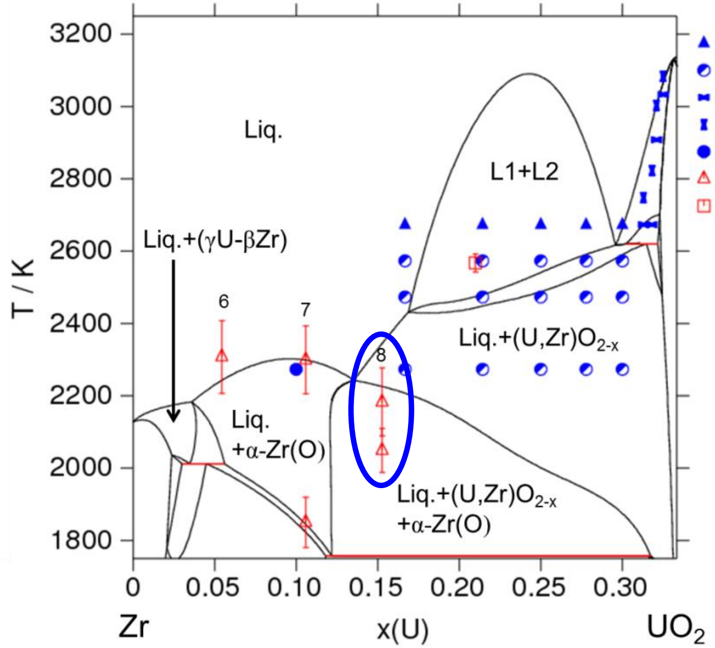


Sample 6



Sample 8

Experimental results



A. Quaini, PhD Thesis, INP Grenoble 2015

Zr-UO_2 isopleth section:

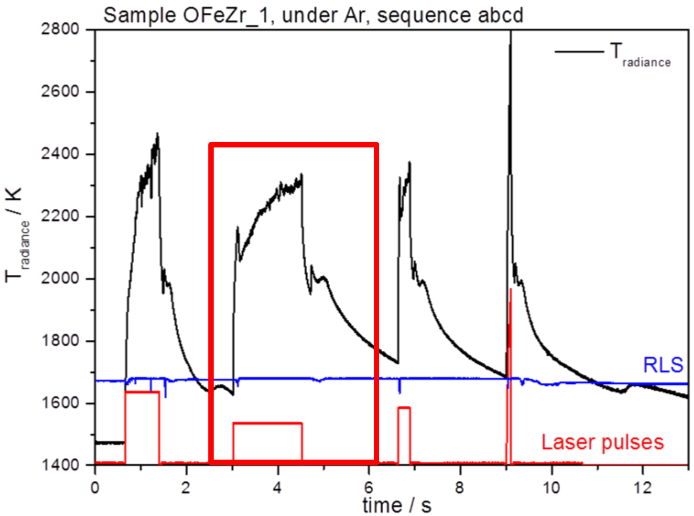
- Calculated using CEA database
- Laser heating data were not used for the assessment
- Good agreement between calculation and exp data on samples 6&7
- Disagreement with sample 8

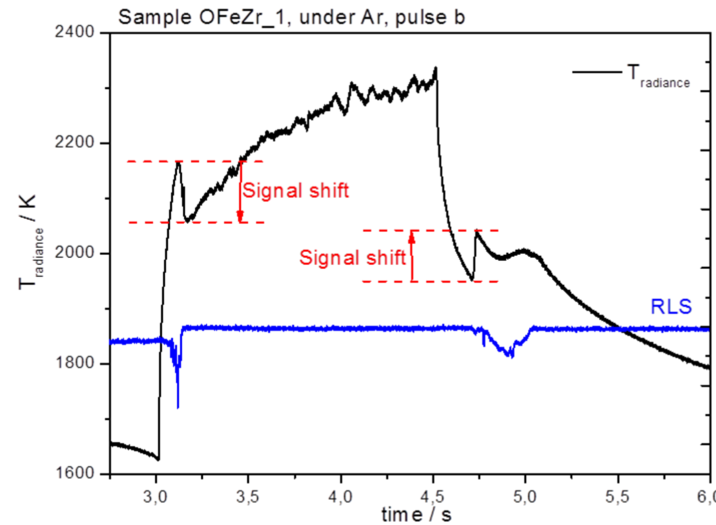
Emissivity may radically change the meaning of a temperature measurement

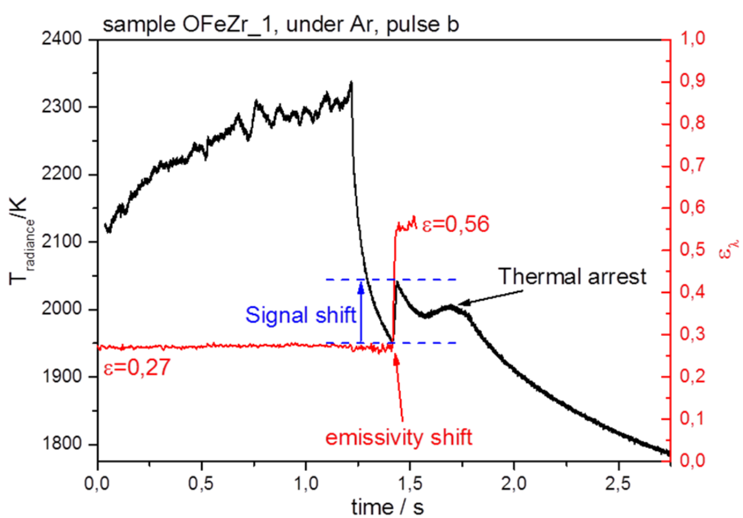
Sample OUZr_8	Experimental result / K (ε=0.40)	Experimental result / K (ε=0.20)	Calculation / K
1st thermal arrest	2183	2343	2345
2nd thermal arrest	2049	2190	2220
Difference	134	153	125

Experimental results – Fe-Zr-O

Fe-containing samples are less stable face to rapid heating → vaporisation is easier







Sharp variation of the thermogram → sol/liq
→ Emissivity effect!!!! (again ☺)

Sample	T_λ / K		True Temperature / K
	1st inflection	ε(652 nm)	1st inflection
OFeZr_1	1989±18	0.56±0.11	2098±86
OFeZr_2	1883±29	0.56±0.11	1980±94
OFeZr_5	1847±11	0.55±0.11	1943±72

The uncertainty on ε contributes for 80% on the overall error bar on true temperature

Conclusions

Conclusions

Melting behavior of Zr-rich corner of the U-Zr-O and Fe-Zr-O systems

Solidification and melting temperatures have been measured using optical pyrometer and RLS technique

Emissivity is the main source of uncertainty→ it strongly affects the error bars on true temperature

The current results are in good agreement with thermodynamic calculations performed using the U-Zr-O CEA database

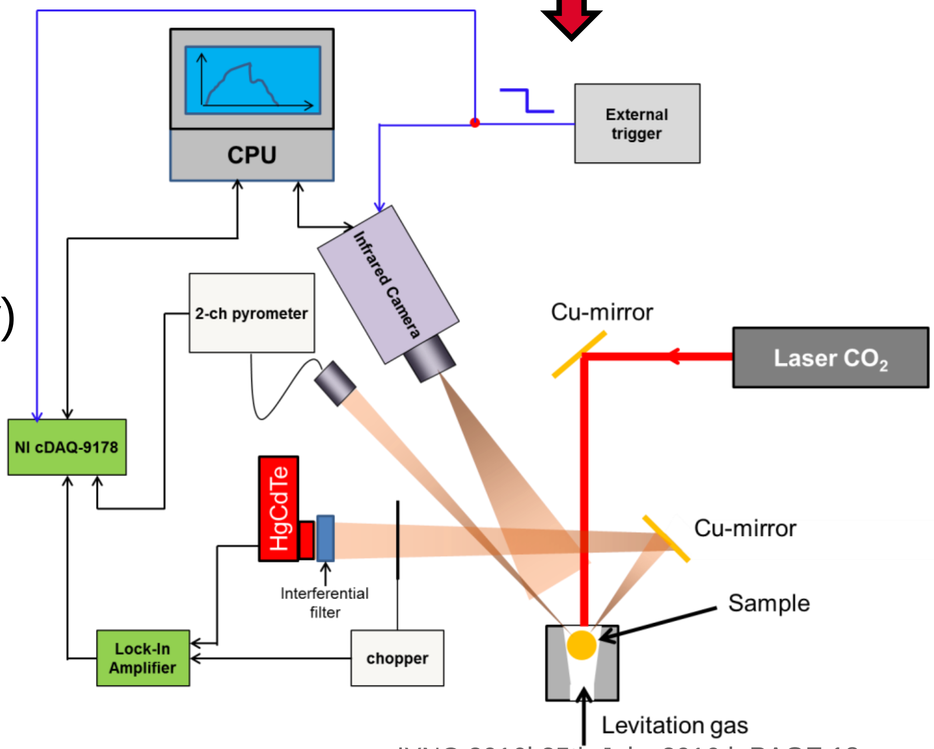
ATTILHA Setup

Advanced **T**emperature and **T**hermodynamics **I**nvestigation by a **L**aser **H**eating **A**pproach

…the God's scourge

ATTILHA: Development of a setup for high solid/liquid transitions

2 different ATTILHA configurations: Contactless → Aerodynamic levitation
Containerless

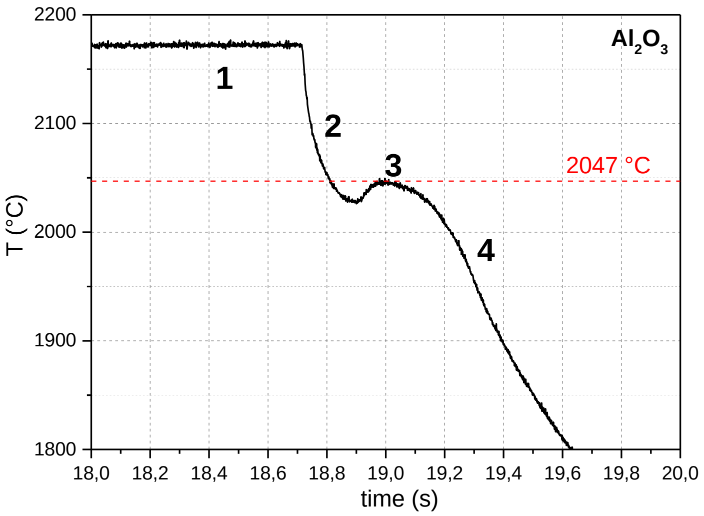


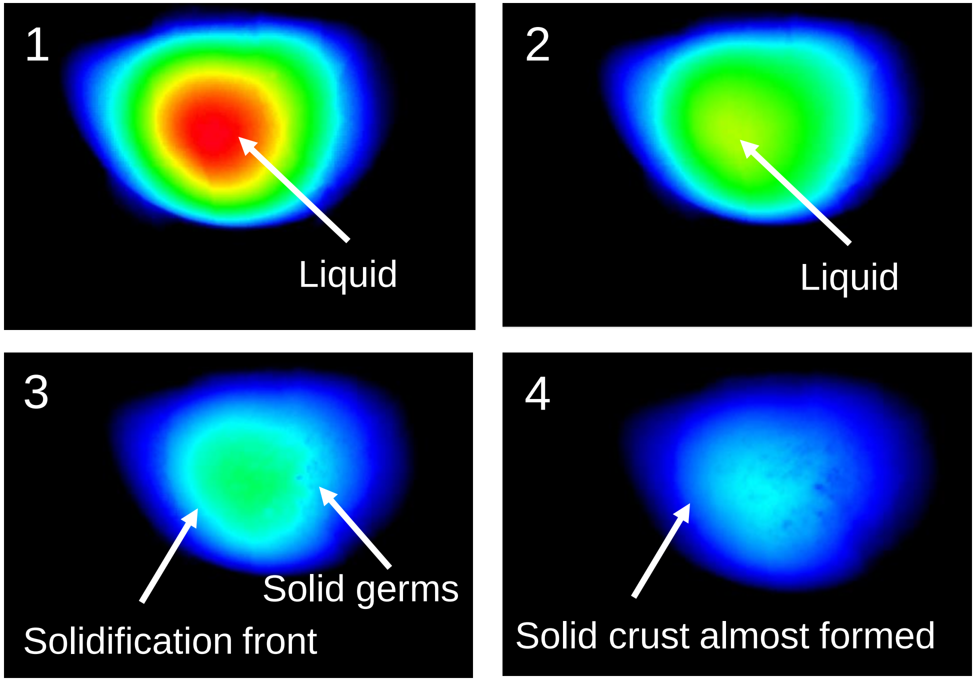
Acquisition of data on corium systems

- Phase diagram data (liquidus, solidus)
- Thermo-radiative properties (IR emissivity)

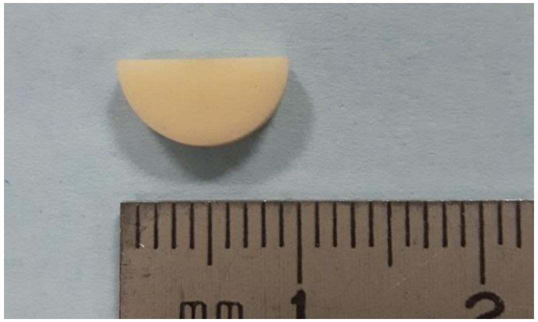
All the instruments are synchronized
Validation on transitions in oxide systems
Al_2O_3
Al_2O_3-ZrO_2

Al_2O_3 melting/solidification results





Before



After

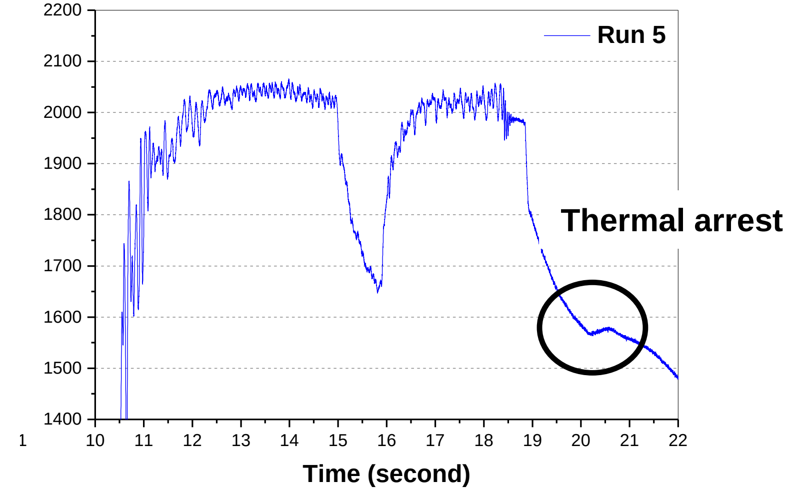
Thermal gradients
Solidification front progression
Semi-transparency

Next step… **U-containing samples**

Al_2O_3-CaO-ZrO_2 – Radiance results

Development of a Python code for image processing and emissivity estimation

Temperature (*T*)



Radiance Temperature (T_L) scale (at 3.99 µm)

Calibrated from 300°C to 1500°C
<u>Extrapolated above 1500°C</u>

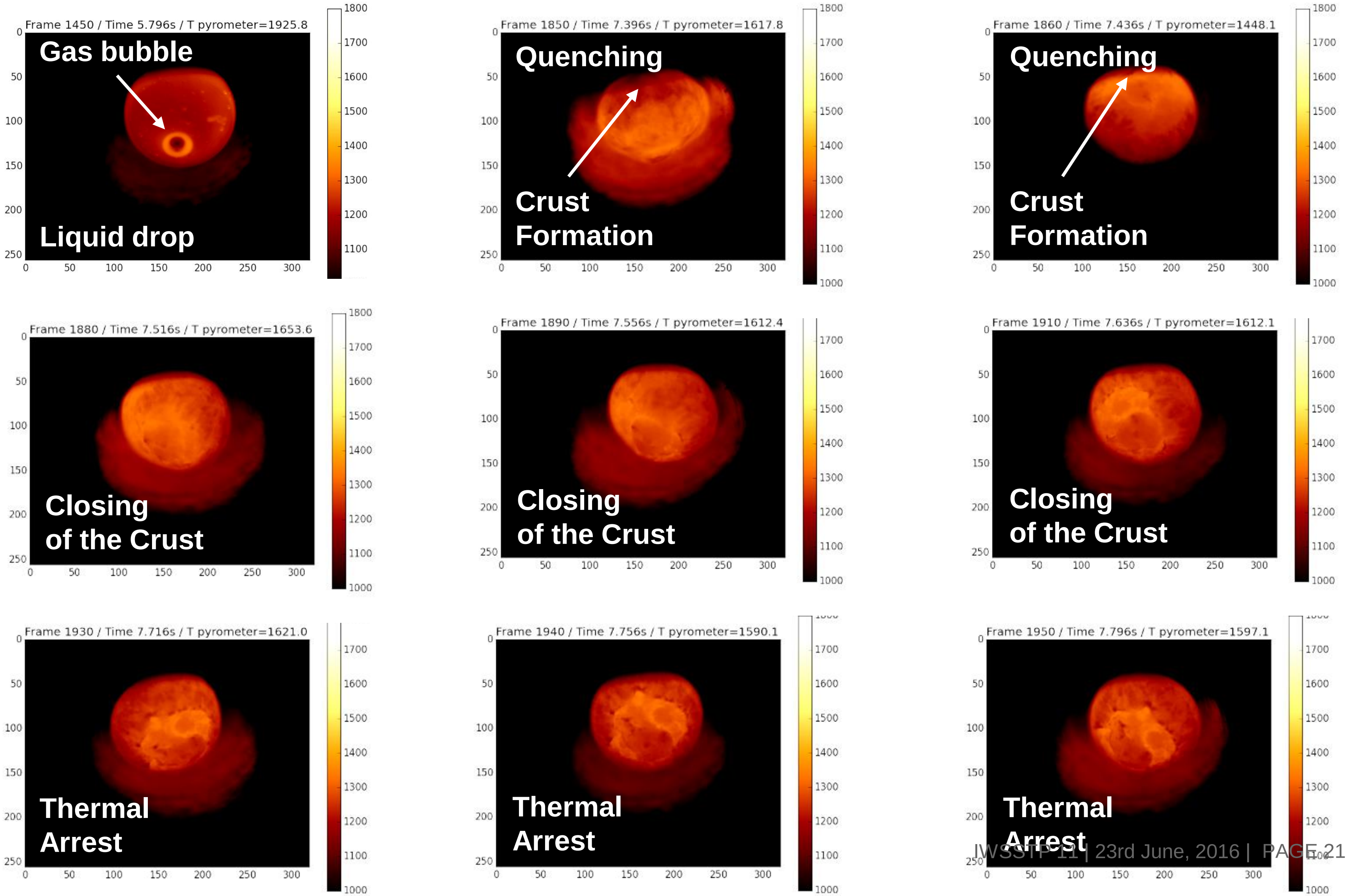
Real speed: 250 Hz

COMMENTS

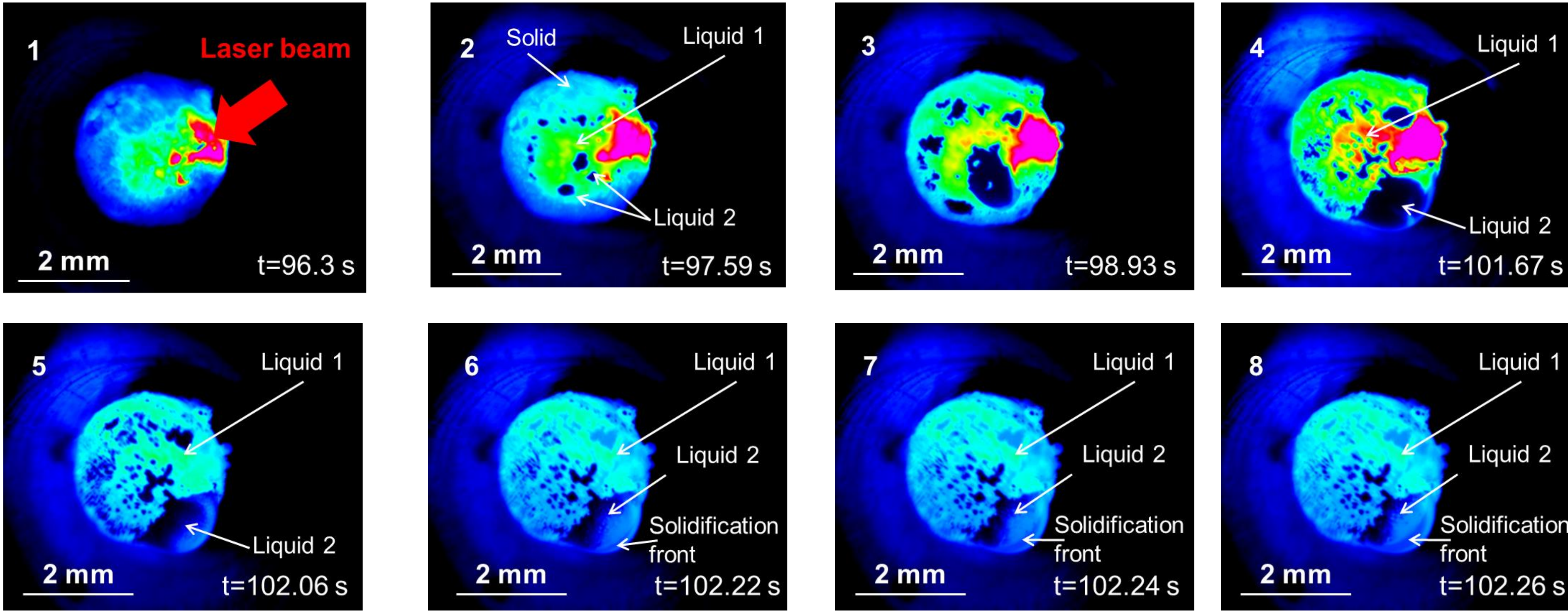
High temperature (1500°C-2500°C) calibration of the camera will be performed soon → FLIR®

HgCdTe detector → cooling system KO

Al_2O_3-CaO-ZrO_2 – Thermal arrests

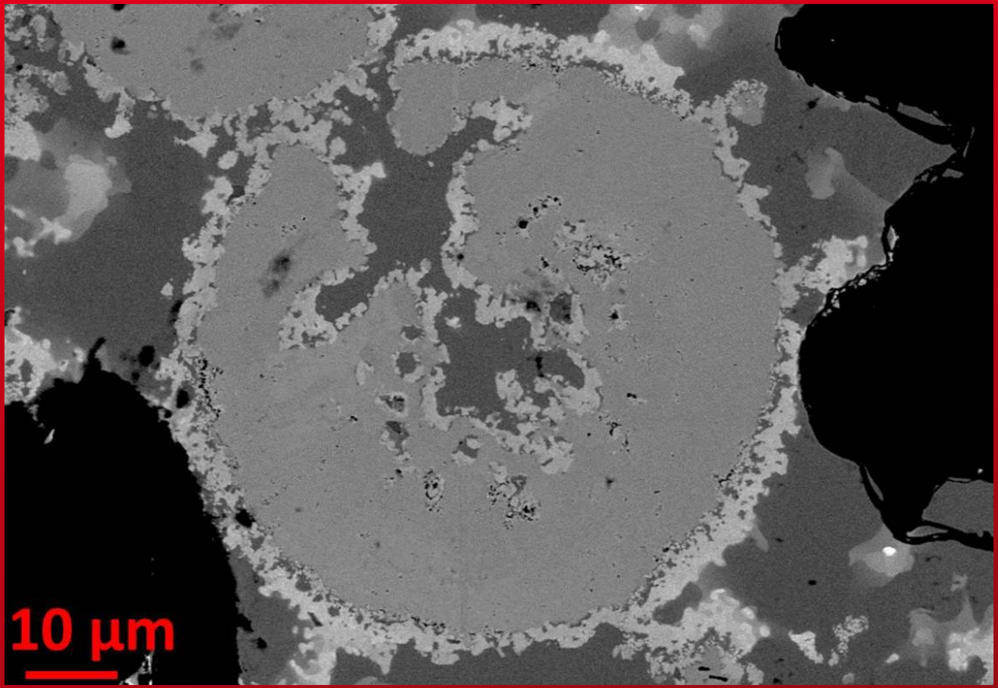


Miscibility gap



Blue: metallic liquid → less emissive
Green - Red: oxide liquid → highly emissive
Pink: overheated liquid (laser impinges directly on that zone)

THANK YOU FOR YOUR ATTENTION



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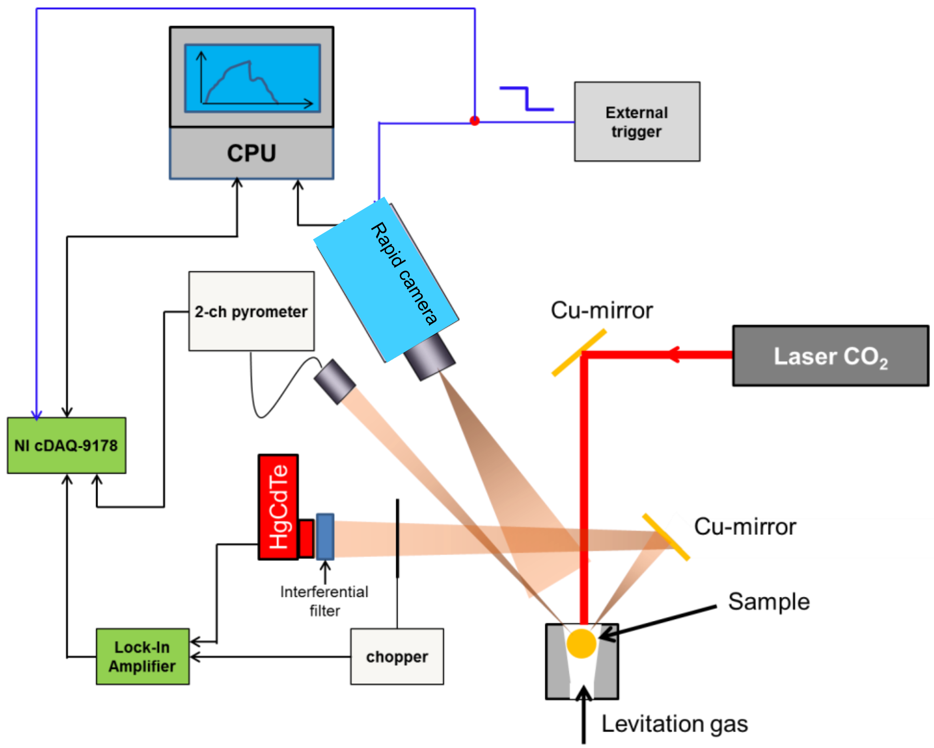
CPU
External trigger
Rapid camera
Cu-mirror
Laser CO_2
2-ch pyrometer
NI cDAQ-9178
HgCdTe
Cu-mirror
Interferential filter
Sample
Lock-In Amplifier
chopper
Levitation gas