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

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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

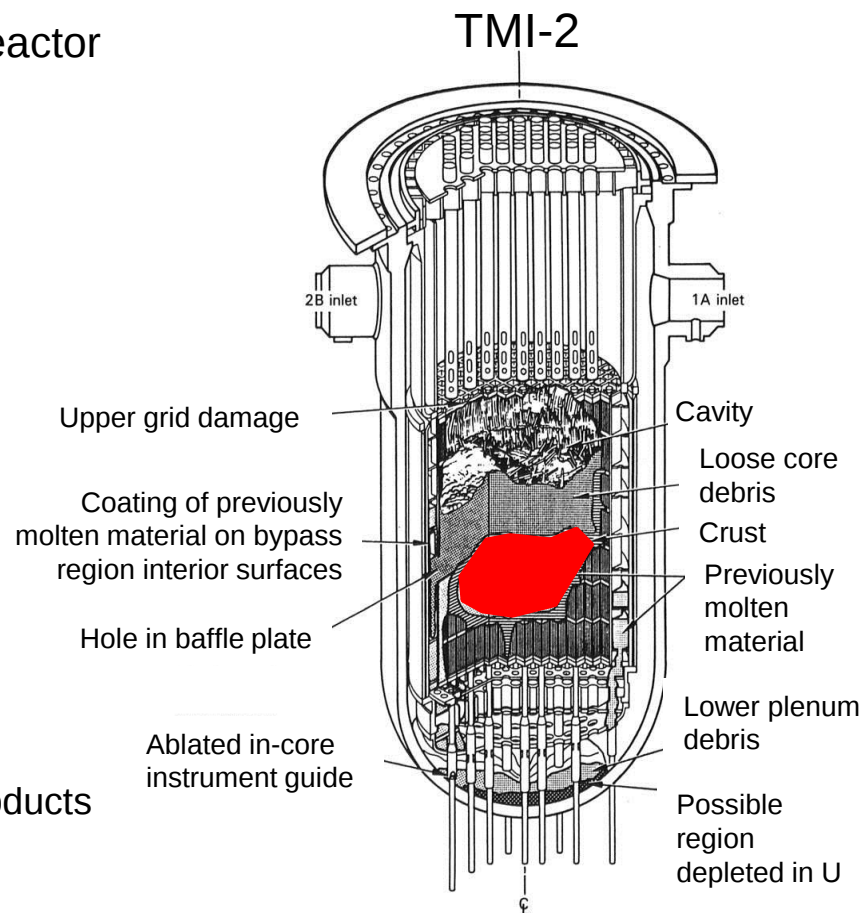
Interactions:

MOx + Zircaloy + stainless steel + Inconel + B₄C + 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**



PWR cross section

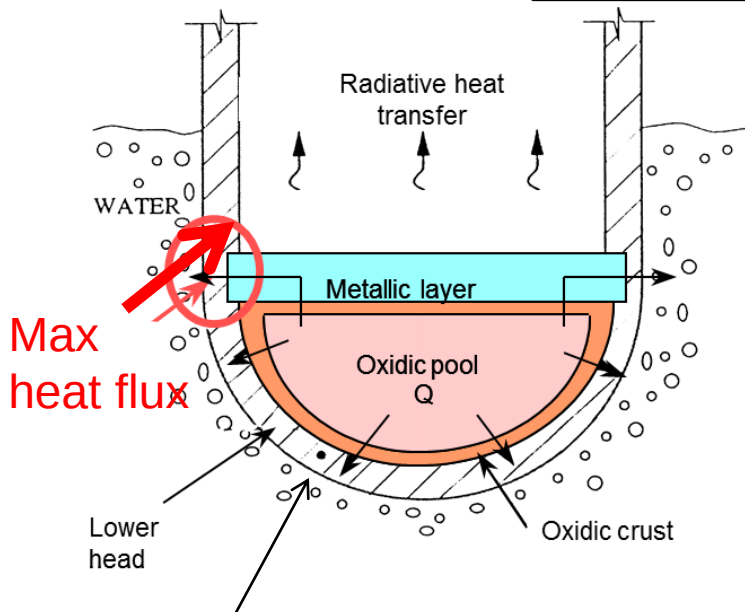
Issue : In-vessel core Melt configuration (Miscibility gap in the in-vessel corium liquid)

$$\rho_{\text{metallic}} < \rho_{\text{oxide}} \longrightarrow$$

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

ρ = density

$$\longrightarrow \rho_{\text{heavy metallic}} > \rho_{\text{oxide}}$$



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

Heavy metal layer below the oxidic pool
Seiler et al., Nucl. Eng. Des. 2007

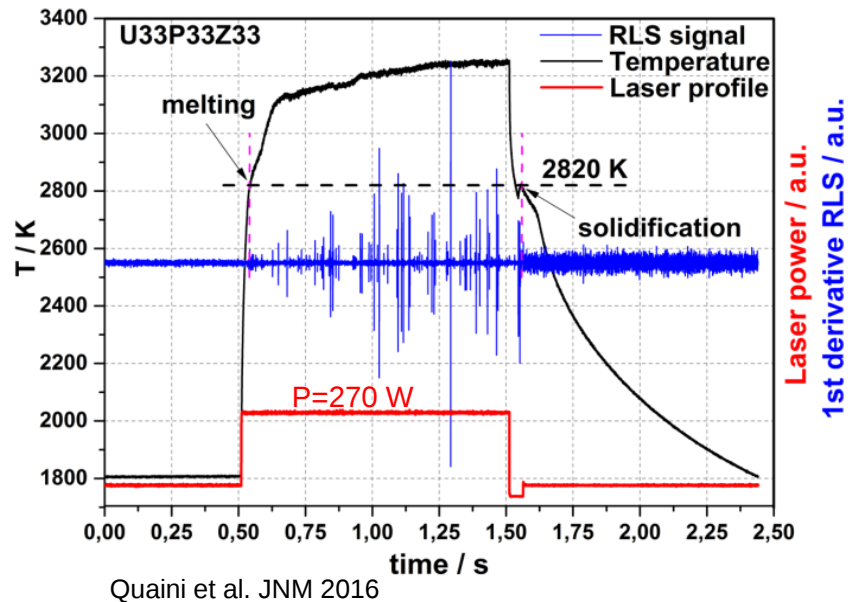
Experimental technique

Sample preparation

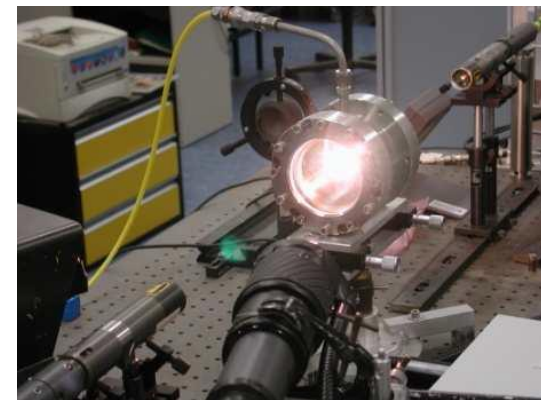
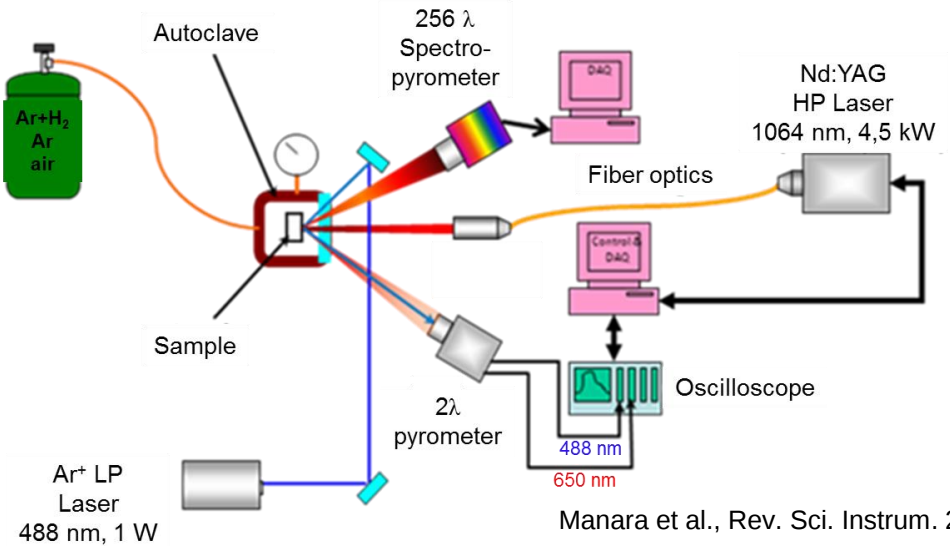
Experimental results

Conclusions

- Rapid measurements
- High power → High temperature
- Containerless
- Reducing/oxidising atmosphere
- Oxygen potential is not controllable



JRC-SNFU's Laser heating setup



De Bruycker, PhD thesis 2010

Sample preparation

Sample preparation – U-Zr-O samples

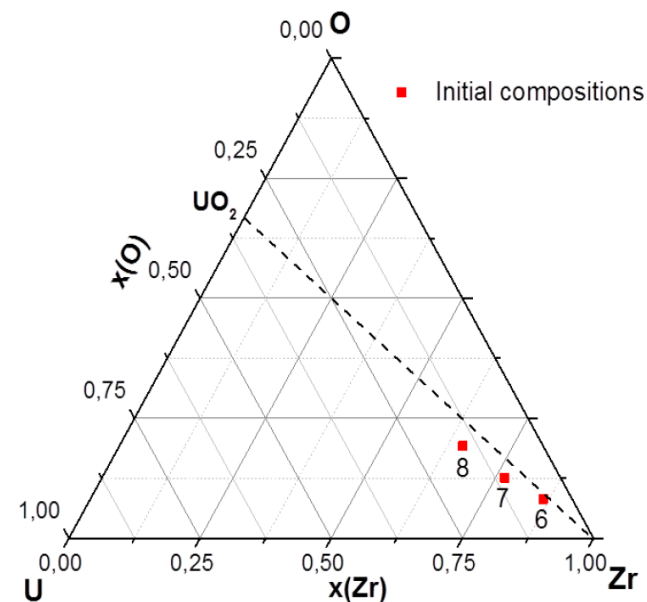
Sample	at% O	at% U	at% Zr	mass / g	Δm / g	Δm / m %
OUZr_6	8.2	5.5	86.3	1.2196	0.0002	0.016
OUZr_7	12.6	10.6	76.8	1.0384	0.0017	0.164
OUZr_8	19.2	15.3	65.5	0.9636	0.0013	0.135

→ Arc furnace

→ U-met, Zr-met, powder of ZrO_2

→ less than 100 ppm of residual O_2 in the furnace chamber

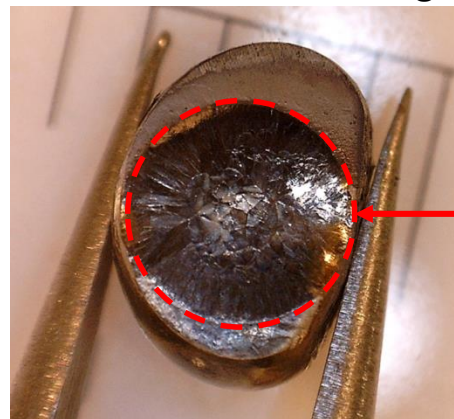
→ mass loss after arc furnace melting is negligible



Before laser heating



After laser heating



Sample preparation – Fe-Zr-O samples

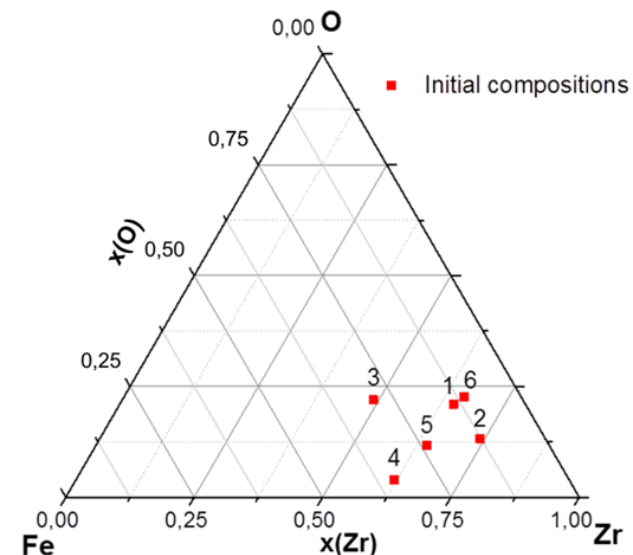
Sample	at% O	at% Fe	at% Zr	mass / g	Δm / g	$\Delta m/m$ %
OFeZr_1	21.0	14.0	65.0	0.7806	0.0020	0.156
OFeZr_2	13.2	12.7	74.1	0.7644	0.0140	1.832
OFeZr_3	21.9	29.1	49.0	0.5559	0.0063	1.133
OFeZr_4	3.9	34.1	62.0	0.5801	0.0006	0.103
OFeZr_5	11.7	23.8	64.5	0.6772	0.0030	0.443
OFeZr_6	22.6	11.1	66.3	0.5463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

→ Arc furnace

→ Fe-met, Zr-met, powder of ZrO_2 , powder Fe_2O_3

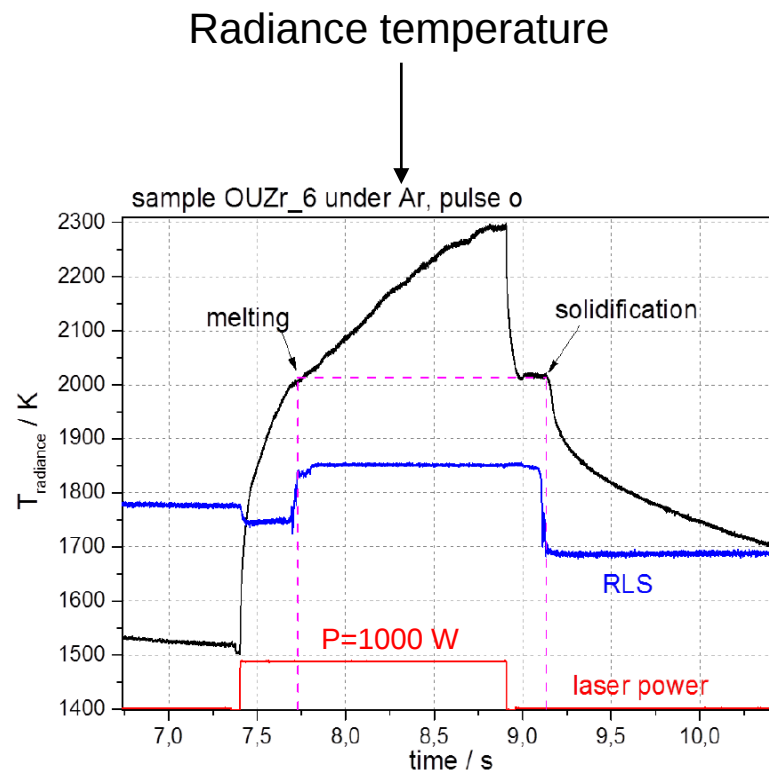
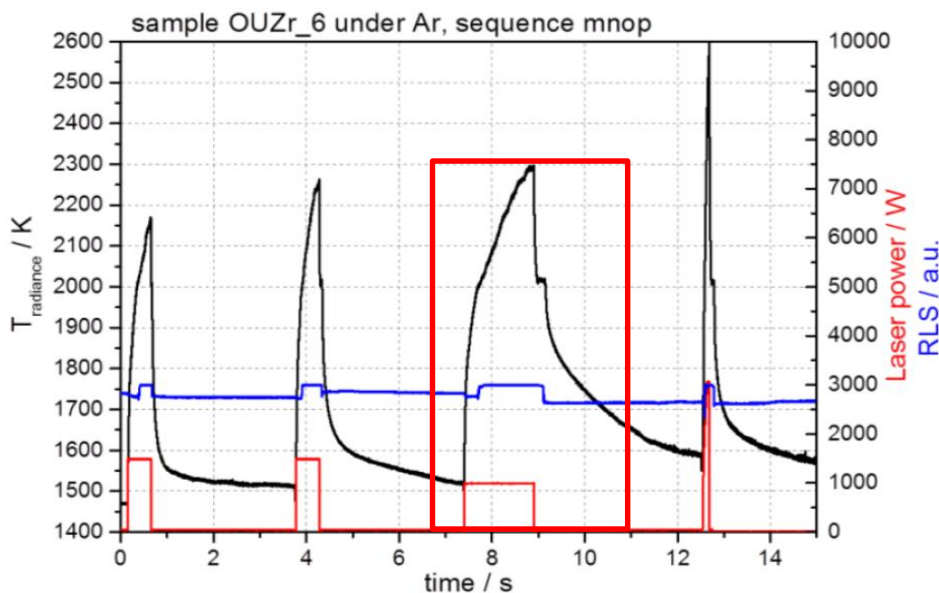
→ less than 100 ppm of residual O_2 in the furnace chamber

→ mass loss is significant for samples 2, 3 and 6



Experimental Results

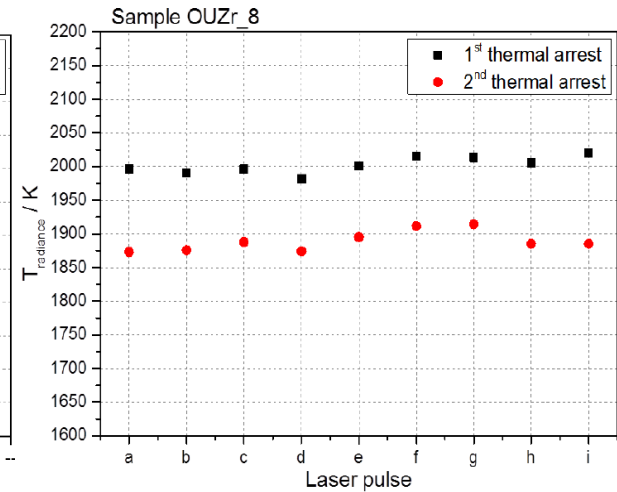
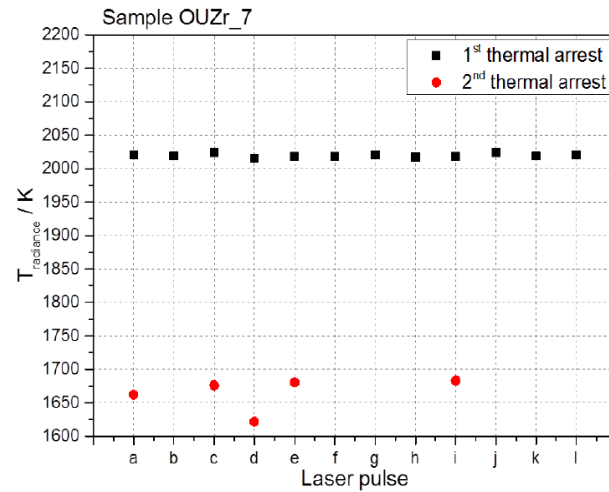
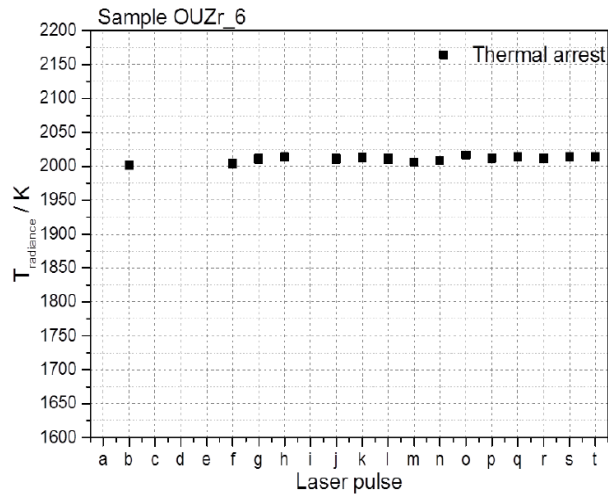
Sample 6: $\text{U}_{0.055}\text{Zr}_{0.863}\text{O}_{0.082}$ (Under Ar)
4 laser shots



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

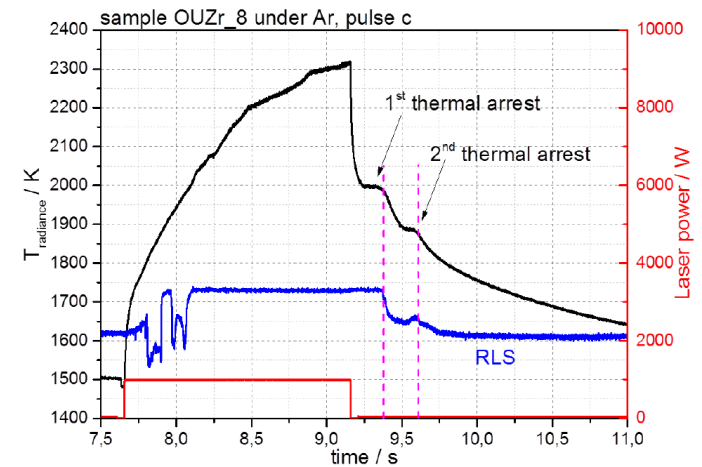
Experimental results – U-Zr-O system



Good repeatability especially on the first thermal arrest

Uncertainty on radiance temperature in less than 0.5 %

Sample	Radiance temperature / K	
	1 st inflection	2 nd 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?

Our case:

Sample	
	$\varepsilon(650 \text{ nm})$
OUZr_6	0.243 ± 0.05
OUZr_7	0.264 ± 0.05
OUZr_8	0.40 ± 0.08

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

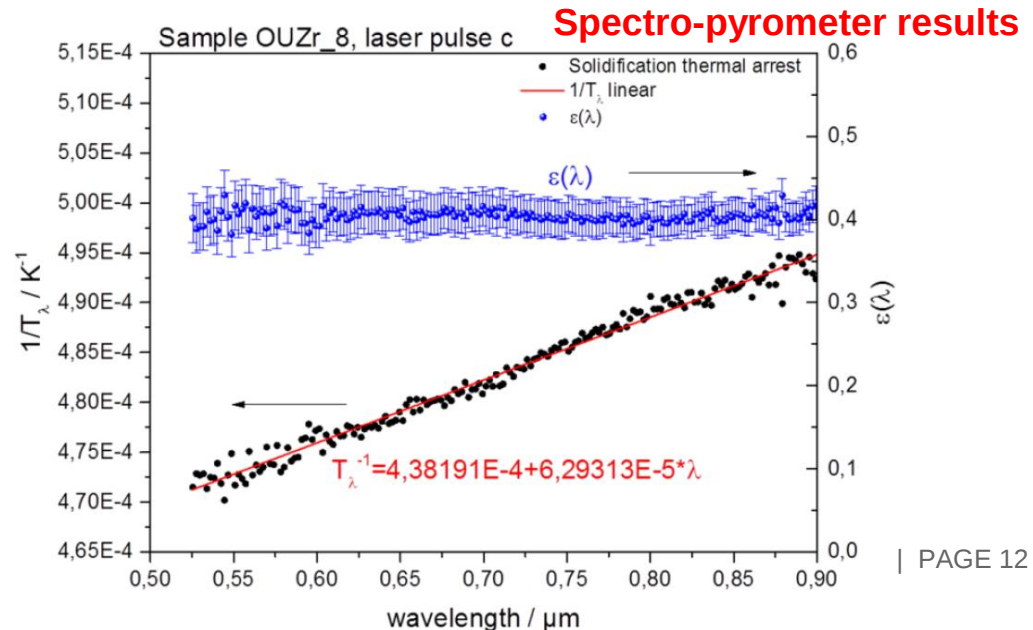
~~Physical expression: $\varepsilon_{\text{sample}} = f(T, \lambda, \dots)$ (Hagen Rubens, Drude, ...)~~ Good for pure metals

~~Polynomial $\varepsilon_{\text{sample}} = \sum_{l=0}^n \alpha_l \lambda_l$~~

Grey body: $\varepsilon_{\text{sample}} = \text{constant}$

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

$$y = y_0 + m * x$$

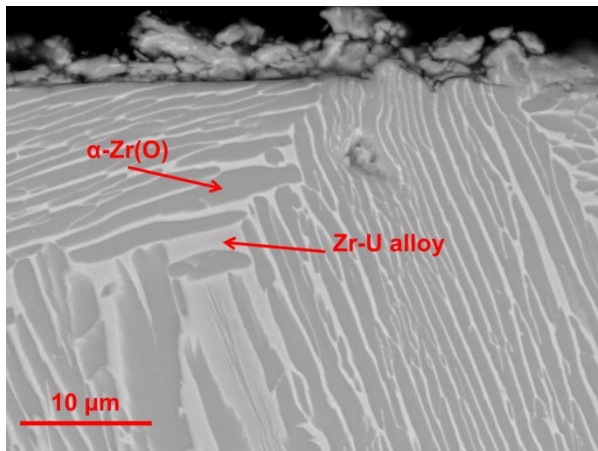


Sample	$\varepsilon(650 \text{ nm})$
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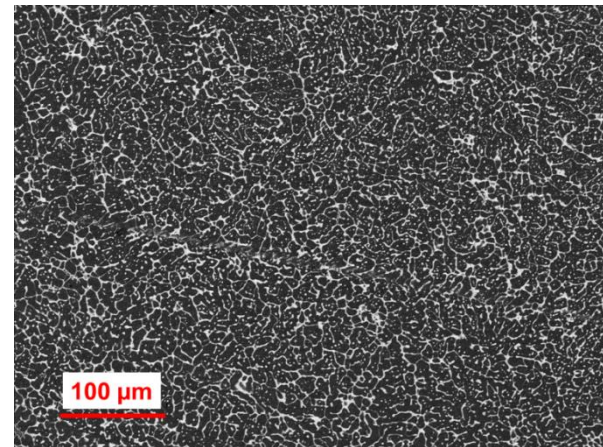
Significant difference between sample 6&7 and 8

Could be related to :

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

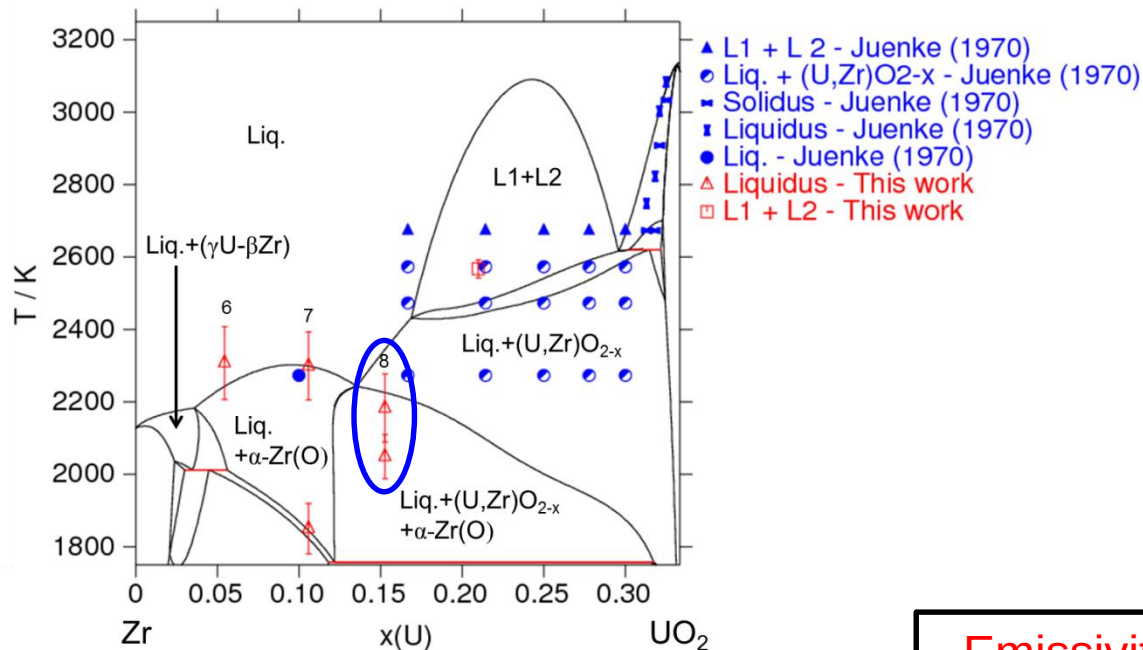


Sample 6



Sample 8

Experimental results



A. Quaini, PhD Thesis, INP Grenoble 2015

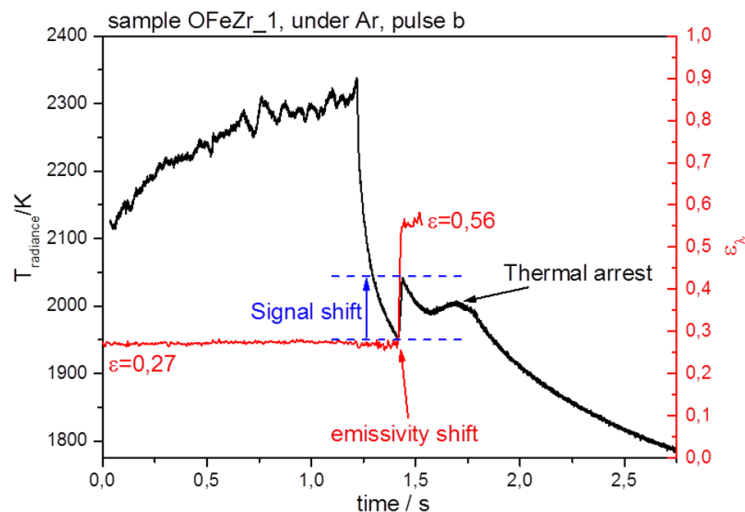
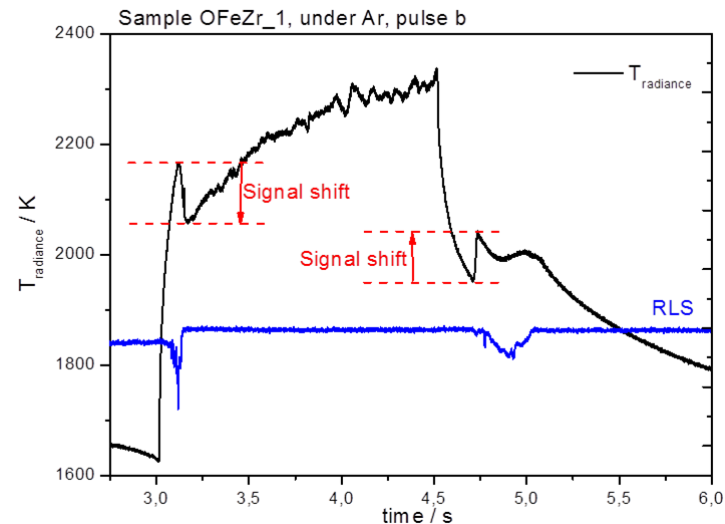
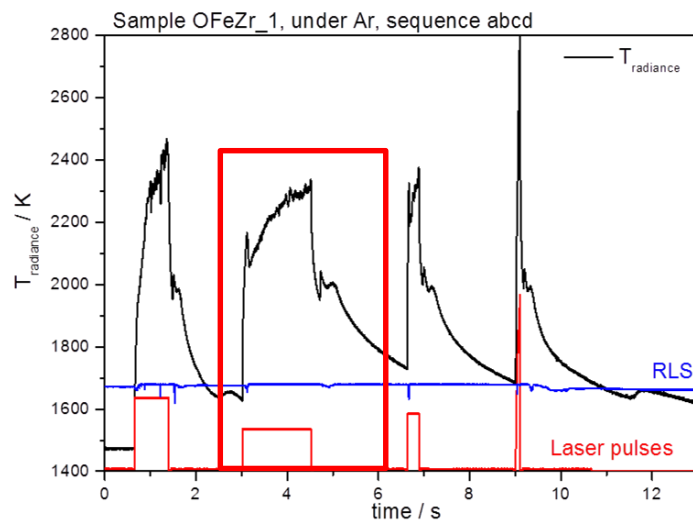
Zr-UO₂ isopleth section:

- 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 ($\epsilon=0.40$)	Experimental result / K ($\epsilon=0.20$)	Calculation / K
1 st thermal arrest	2183	2343	2345
2 nd thermal arrest	2049	2190	2220
Difference	134	153	125

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_r / K	$\varepsilon(652 \text{ nm})$	True Temperature / K
	1 st inflection		1 st 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

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

Advanced Temperature and Thermodynamics Investigation by a Laser Heating Approach



...the God's scourge

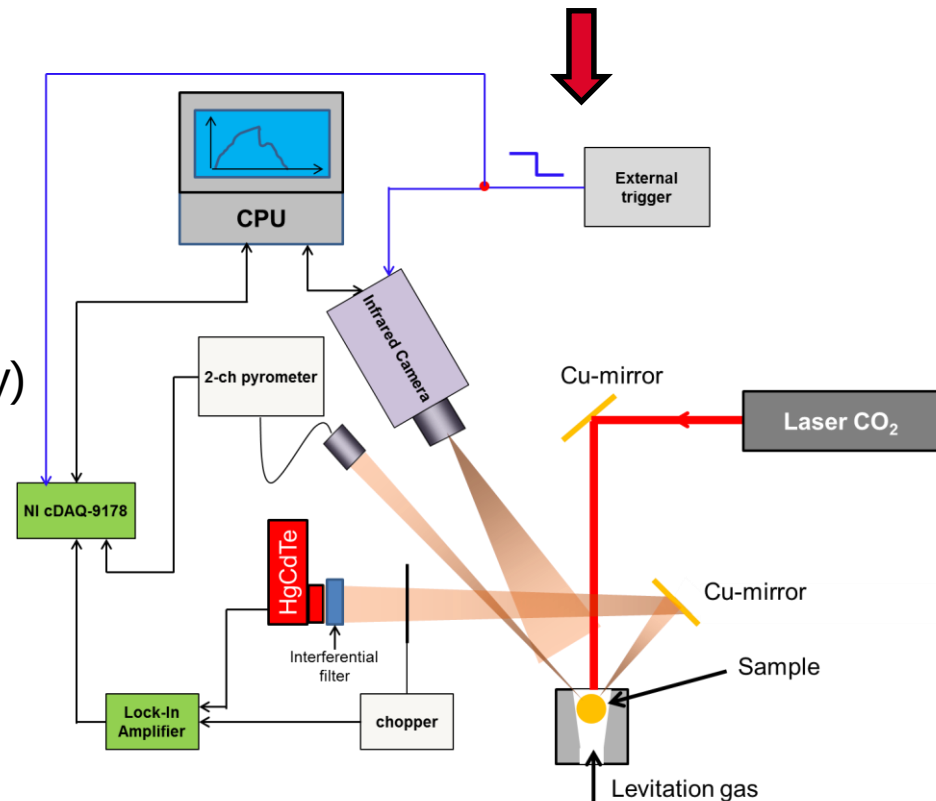
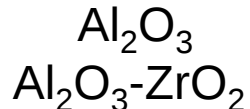
ATTILHA: Development of a setup for high solid/liquid transitions

2 different ATTILHA configurations: Contactless → Aerodynamic levitation
Containerless

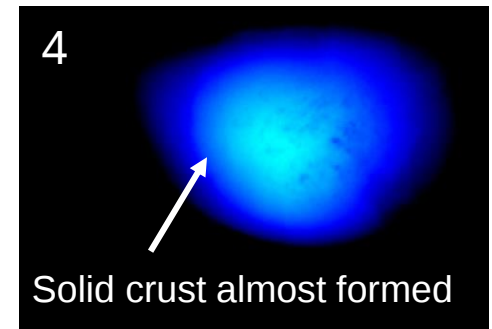
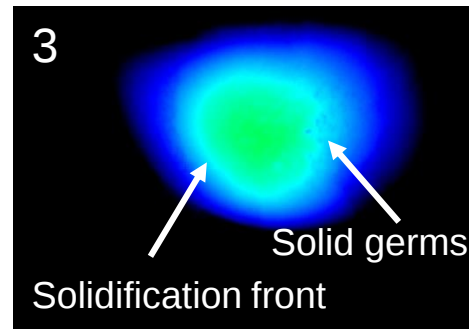
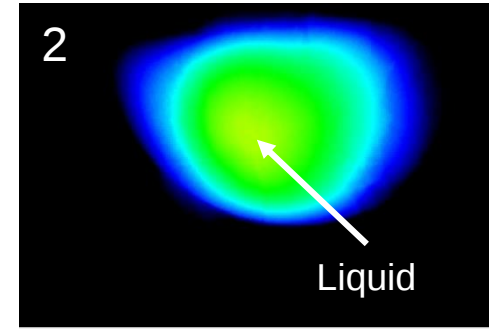
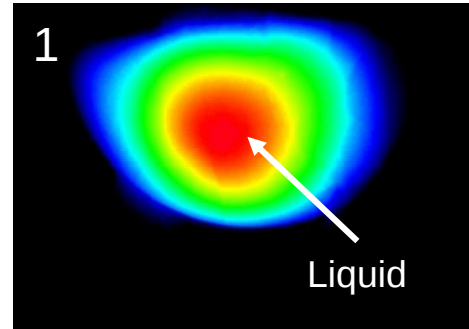
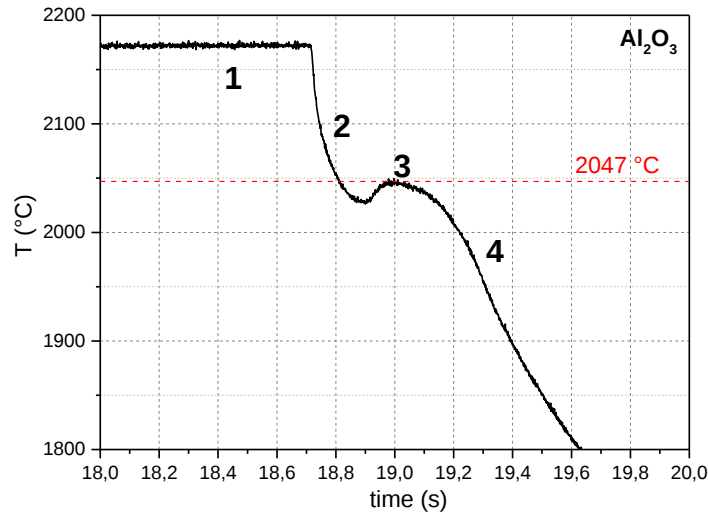
Acquisition of data on corium systems

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

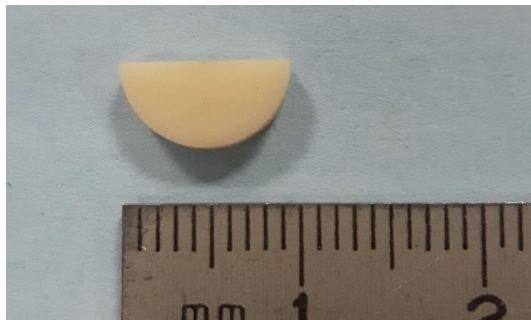
All the instruments are synchronized
Validation on transitions in oxide systems



Al₂O₃ melting/solidification results



Before



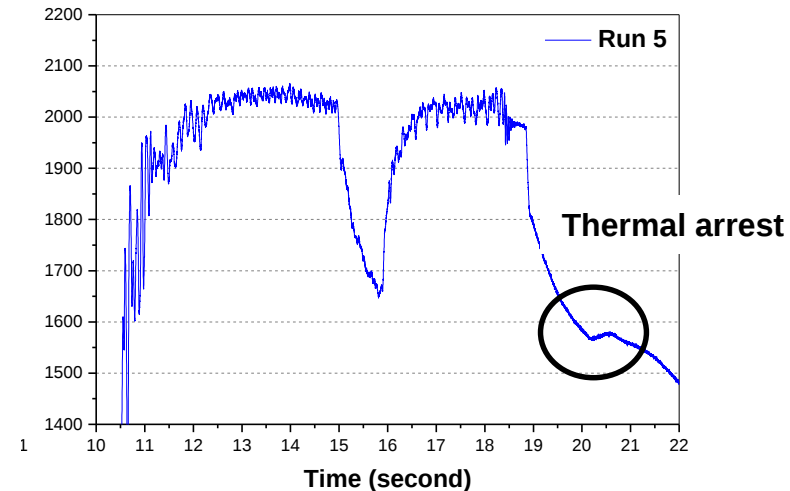
After



Thermal gradients
Solidification front progression
Semi-transparency

Next step... **U-containing samples**

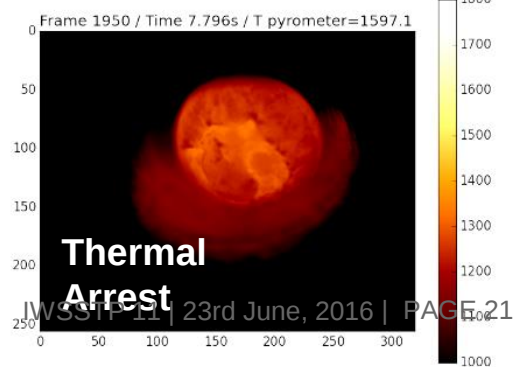
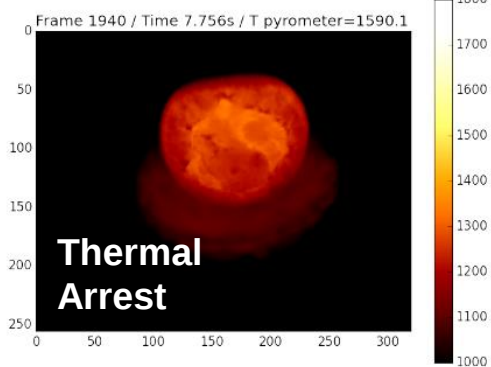
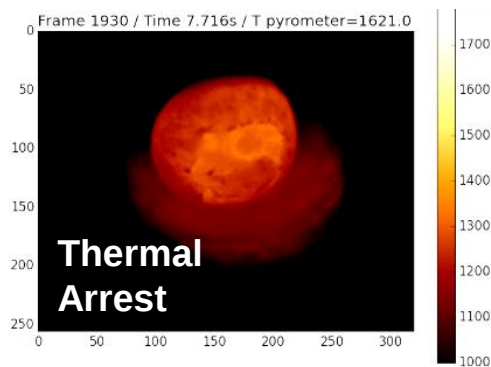
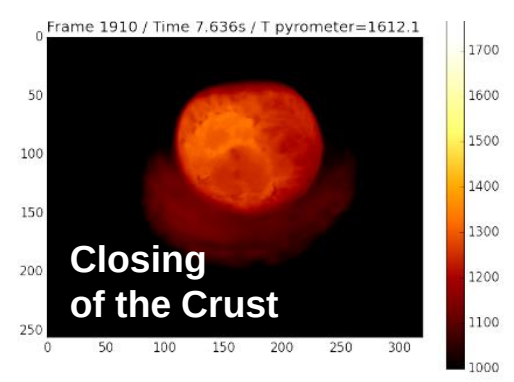
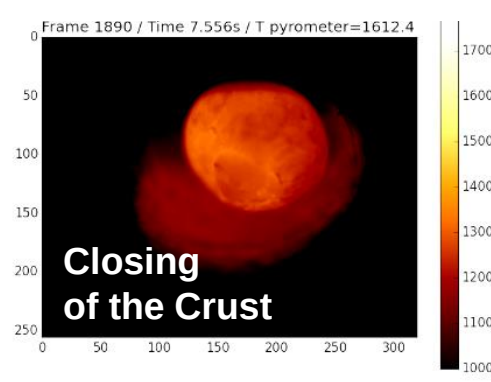
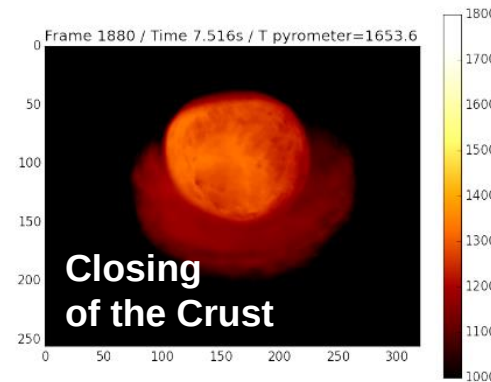
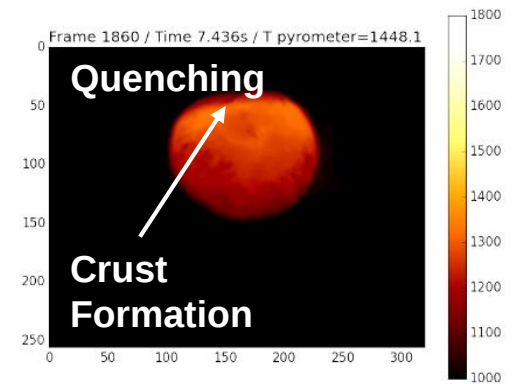
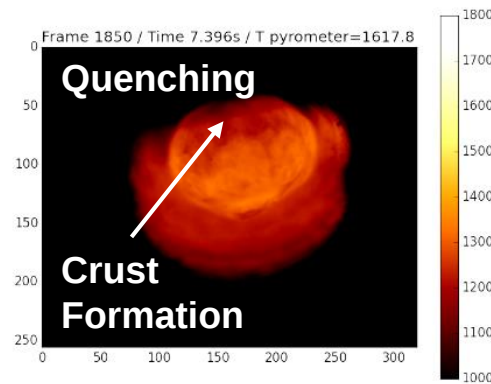
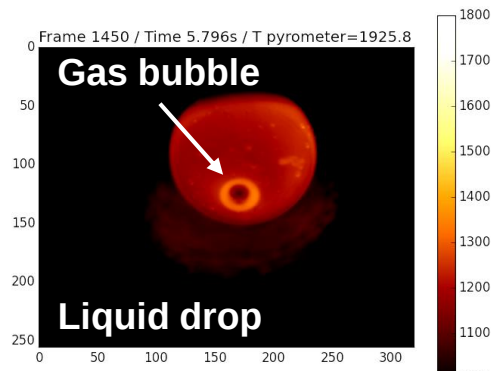
Development of a Python code for image processing and emissivity estimation

Temperature (T)Radiance Temperature (T_L) scale
(at 3.99 μm)Calibrated from 300°C to 1500°C
Extrapolated above 1500°C

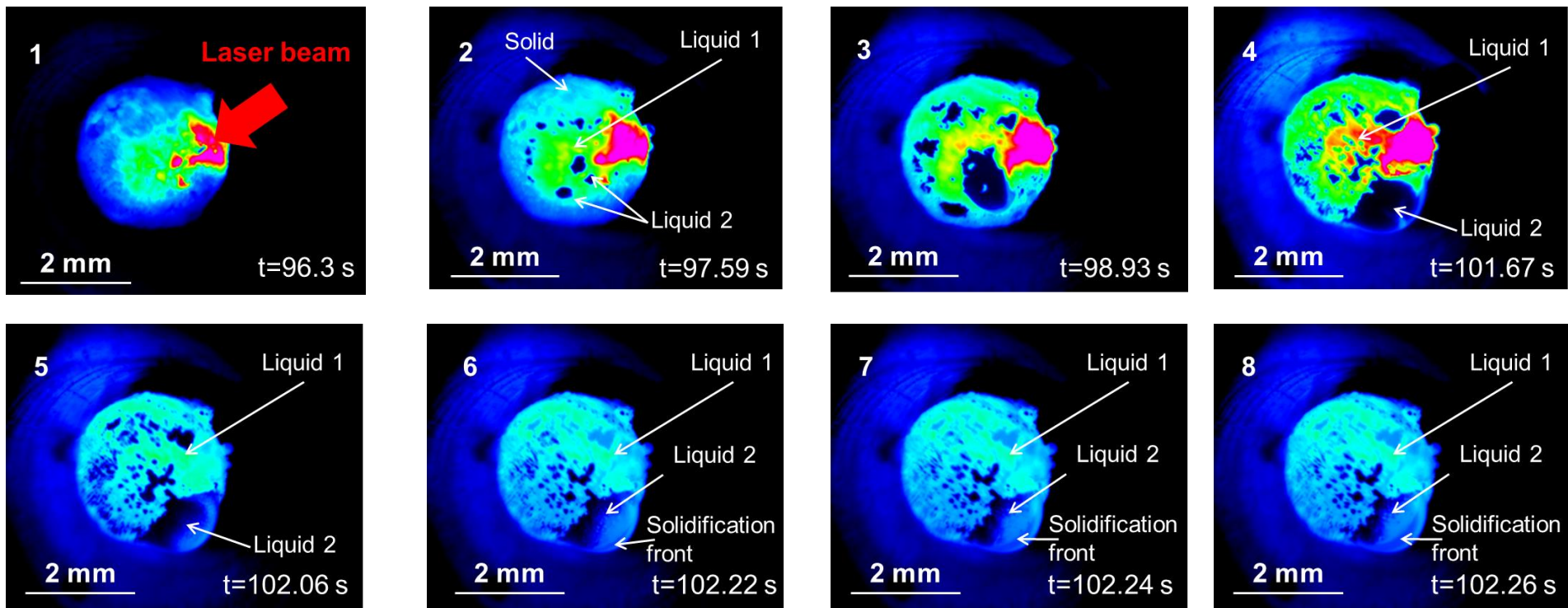
Real speed: 250 Hz

COMMENTS

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

cea den $\text{Al}_2\text{O}_3\text{-CaO-ZrO}_2$ – Thermal arrests

Miscibility gap

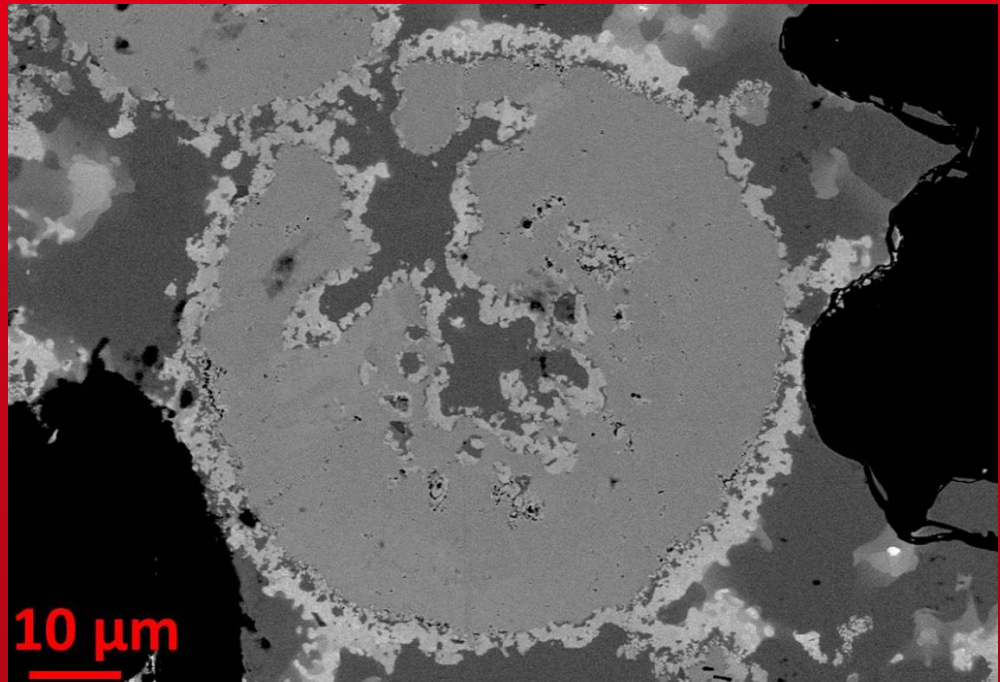


Blue: metallic liquid → less emissive

Green - Red: oxide liquid → highly emissive

Pink: overheated liquid (laser impinges directly on that zone)

THANK YOU FOR YOUR ATTENTION



Dr Andrea Quaini
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