

# 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

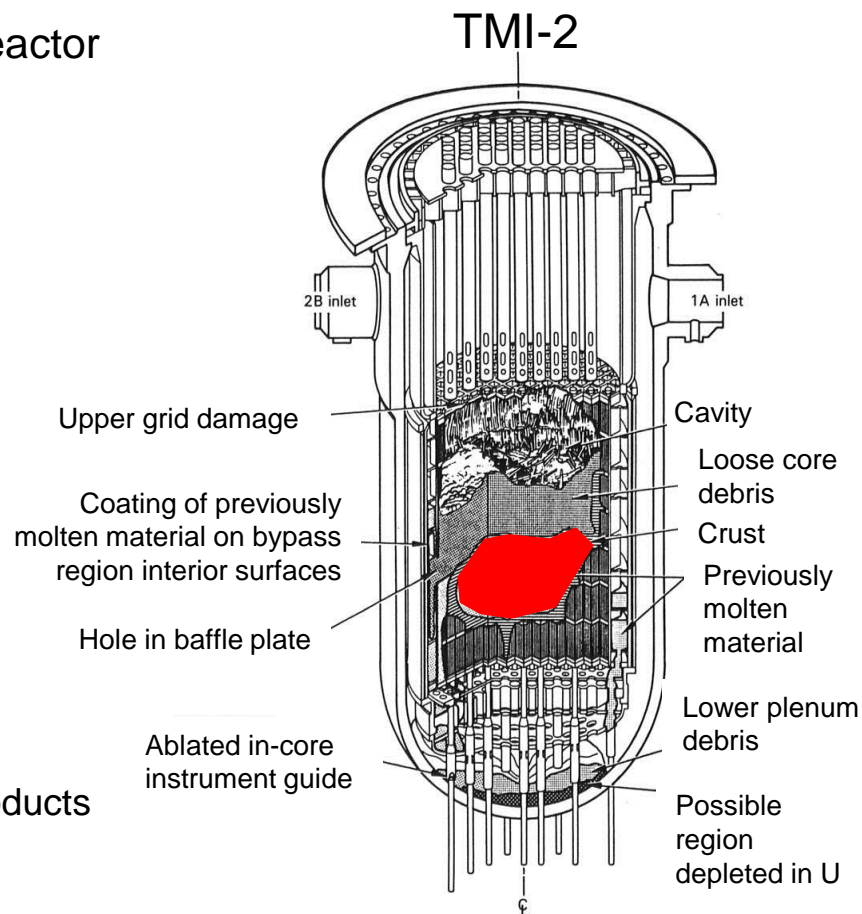
### Interactions:

MOx + Zircaloy + stainless steel + Inconel + B<sub>4</sub>C + Fission Products

### In vessel corium composition:

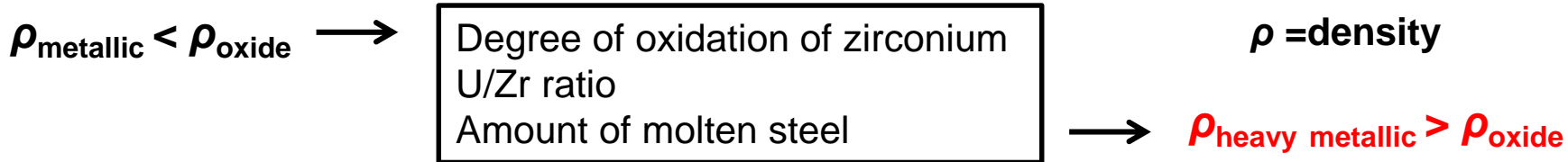
U-Zr-O-Fe-Cr-Ni-Ag-Cd-In-B-C-FPs-... (basically most of the period table)

“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)

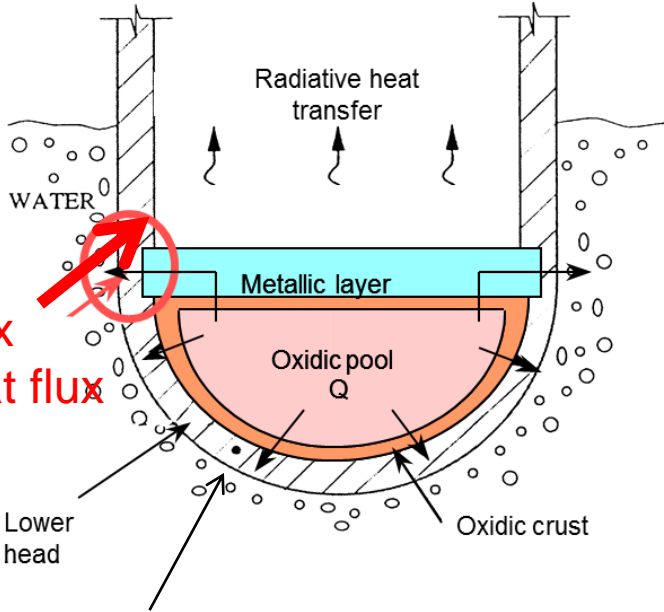


Decrease of the thickness of the upper metallic layer

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



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



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

**Experimental technique**

**Sample preparation**

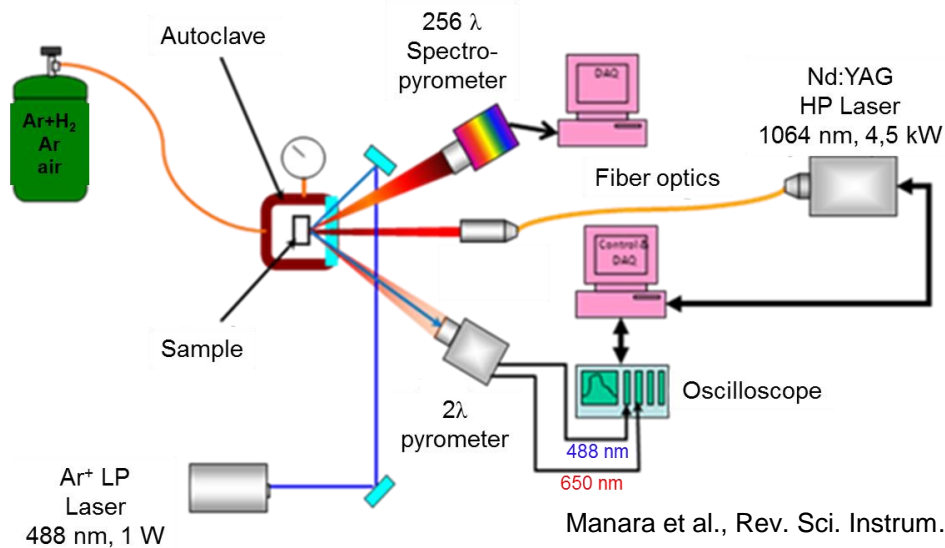
**Experimental results**

**Conclusions**

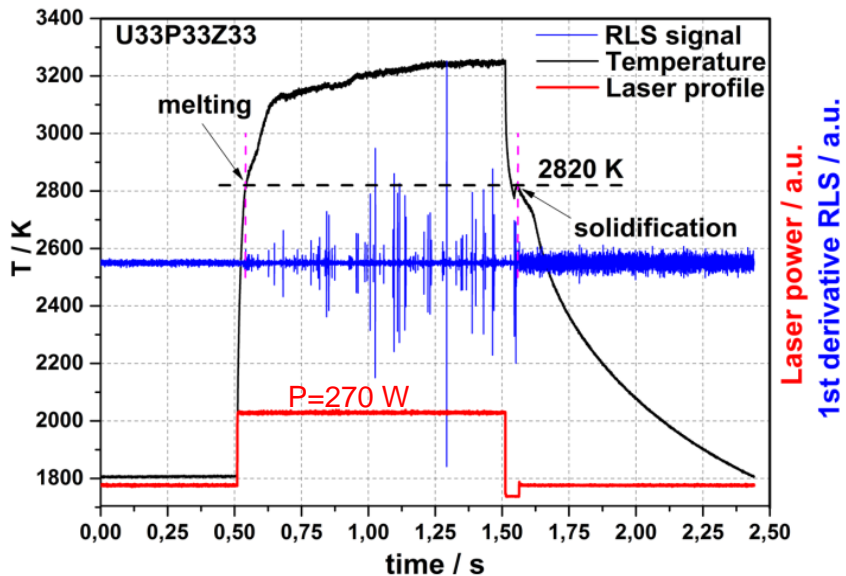
# Experimental technique - Laser heating setup

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

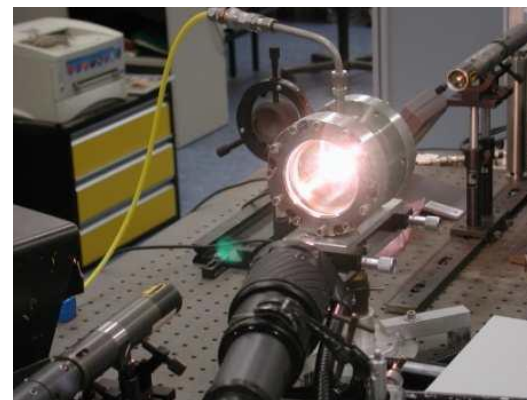
## JRC-SNFU's Laser heating setup



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

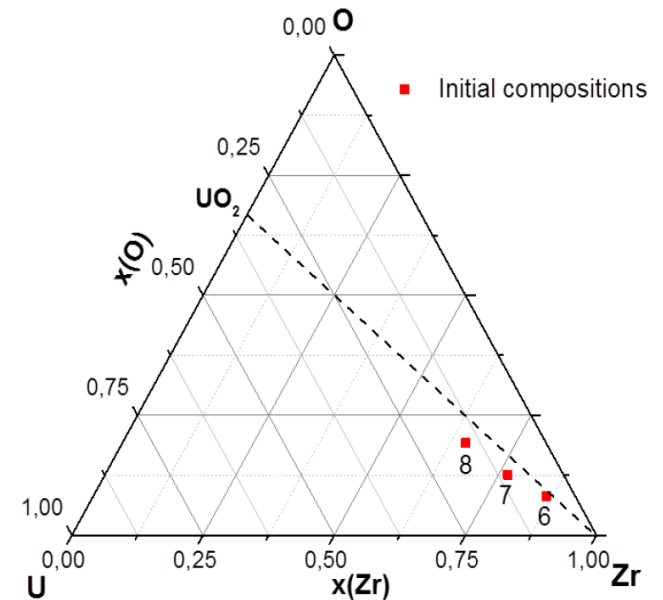
Sample	at% O	at% U	at% Zr	mass / g	$\Delta m$ / g	$\Delta m$ / m %
OUZr_6	8.2	5.5	86.3	1.2196	0.0002	0.016
OUZr_7	12.6	10.6	76.8	1.0384	0.0017	0.164
OUZr_8	19.2	15.3	65.5	0.9636	0.0013	0.135

→ Arc furnace

→ U-met, Zr-met, powder of  $ZrO_2$

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

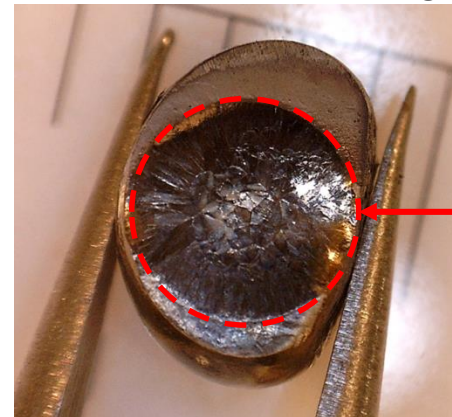
→ mass loss after arc furnace melting is negligible



Before laser heating



After laser heating



# Sample preparation – Fe-Zr-O samples

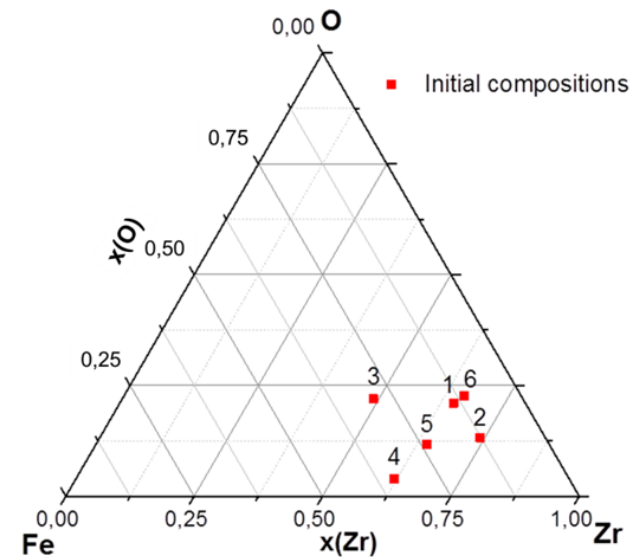
Sample	at% O	at% Fe	at% Zr	mass / g	$\Delta m$ / g	$\Delta m/m$ %
OFeZr_1	21.0	14.0	65.0	0.7806	0.0020	0.156
OFeZr_2	13.2	12.7	74.1	0.7644	0.0140	1.832
OFeZr_3	21.9	29.1	49.0	0.5559	0.0063	1.133
OFeZr_4	3.9	34.1	62.0	0.5801	0.0006	0.103
OFeZr_5	11.7	23.8	64.5	0.6772	0.0030	0.443
OFeZr_6	22.6	11.1	66.3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

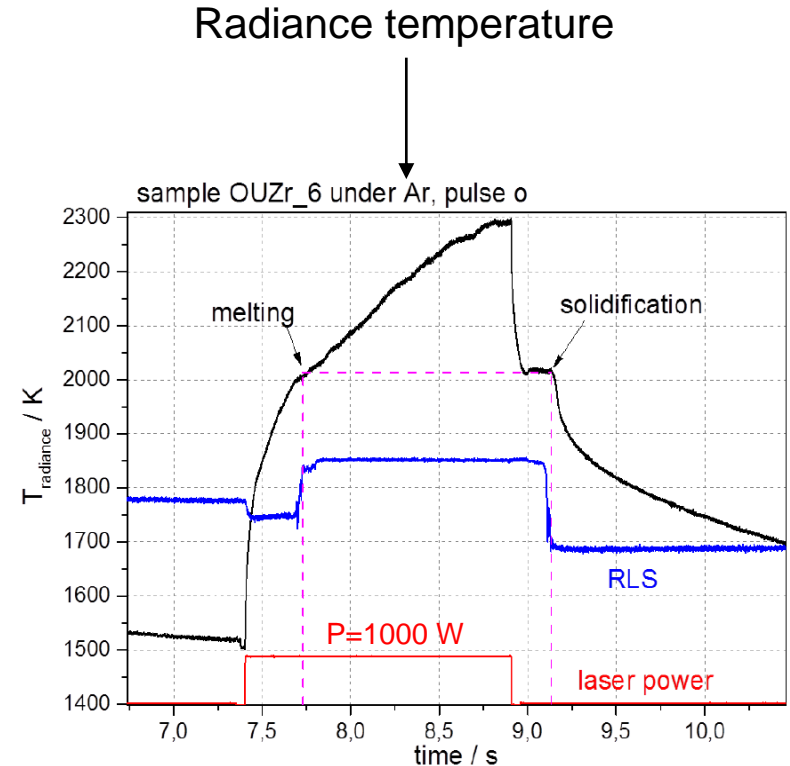
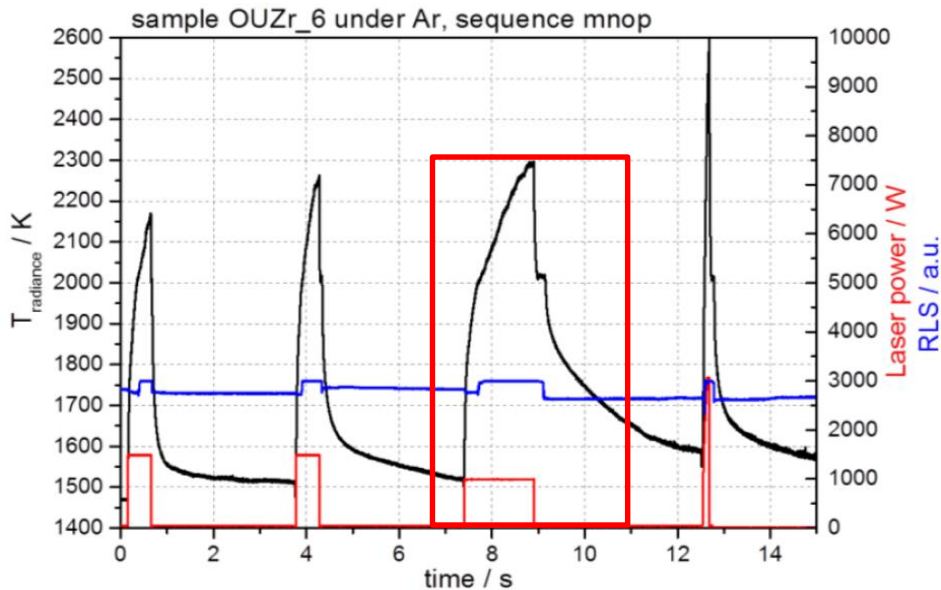
→ mass loss is significant for samples 2, 3 and 6





# Experimental Results

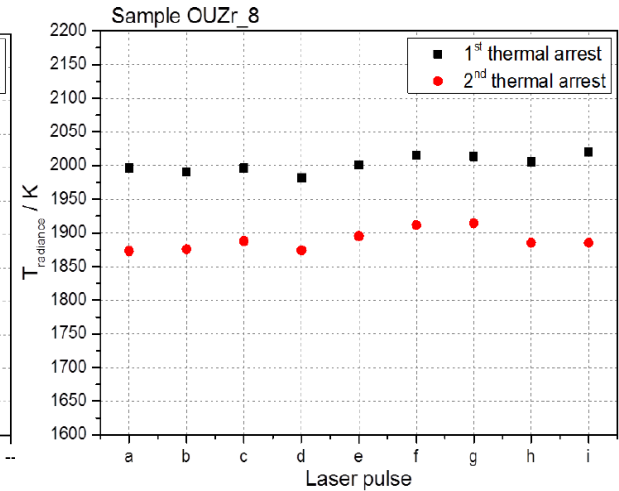
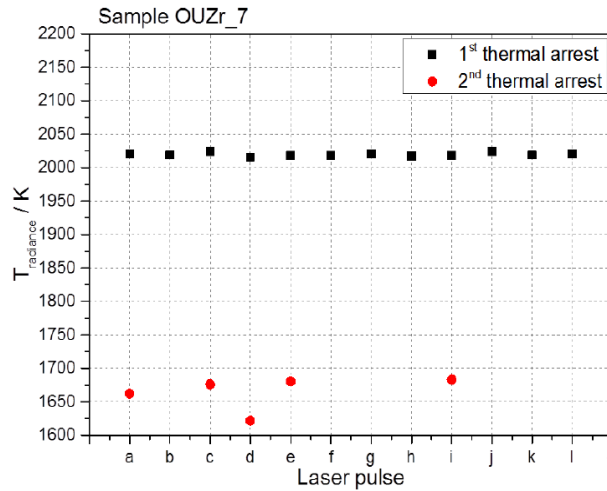
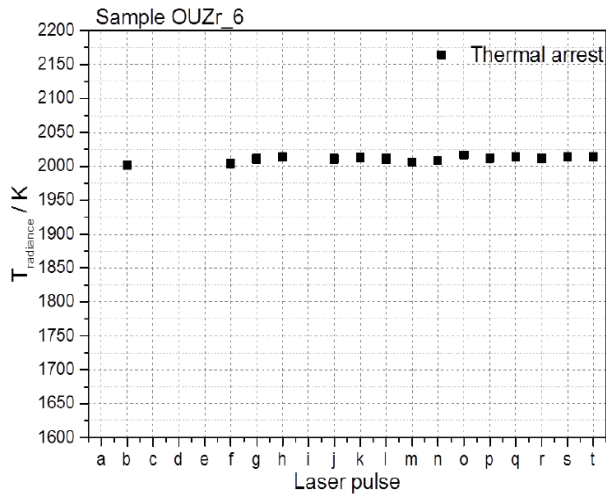
Sample 6:  $U_{0.055}Zr_{0.863}O_{0.082}$  (Under Ar)  
4 laser shots



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**

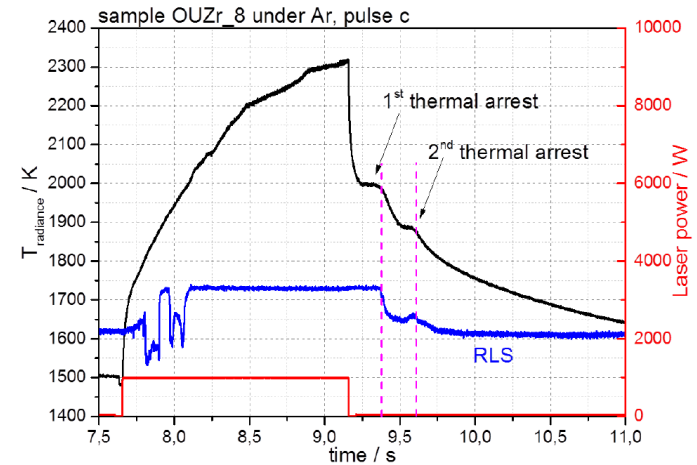
# Experimental results – U-Zr-O system



Good repeatability especially on the first thermal arrest

Uncertainty on radiance temperature in less than 0.5 %

Sample	Radiance temperature / K	
	1 <sup>st</sup> inflection	2 <sup>nd</sup> 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	$\epsilon(650 \text{ nm})$
OUZr_6	0.243±0.05
OUZr_7	0.264±0.05
OUZr_8	0.40±0.08

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

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

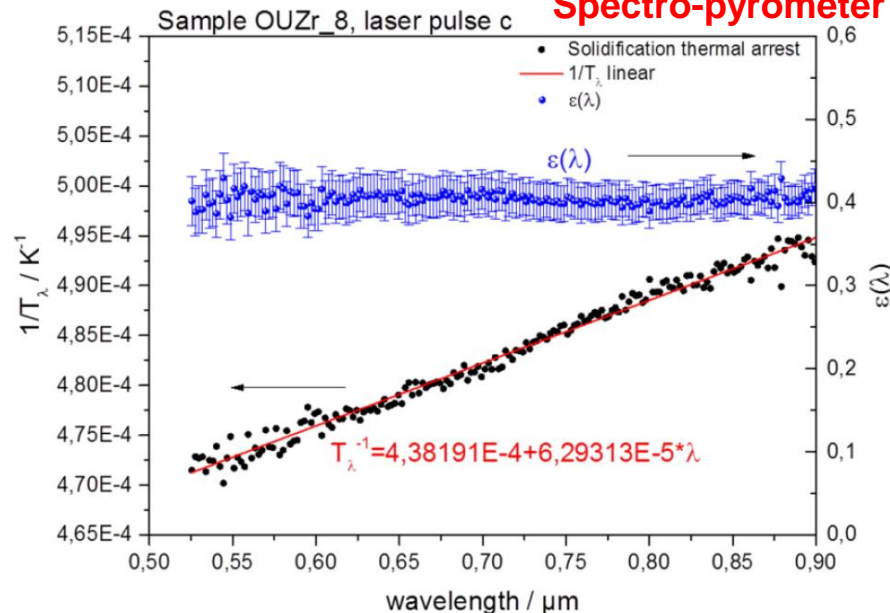
~~Polynomial  $\epsilon_{\text{sample}} = \sum_{l=0}^n \alpha_l \lambda^l$~~

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

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

$$y = y_0 + m * x$$

## Spectro-pyrometer results



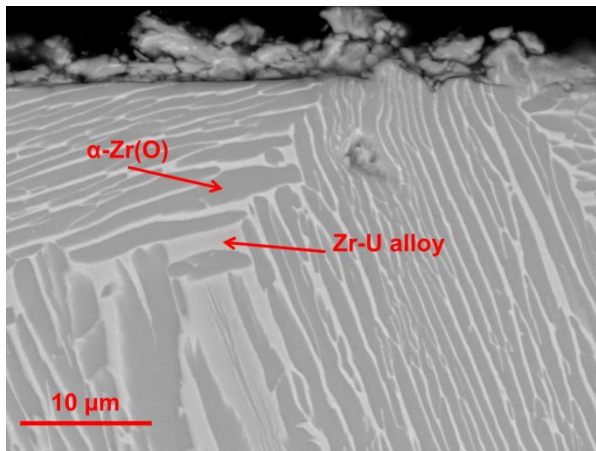
# Experimental results – emissivity again??

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

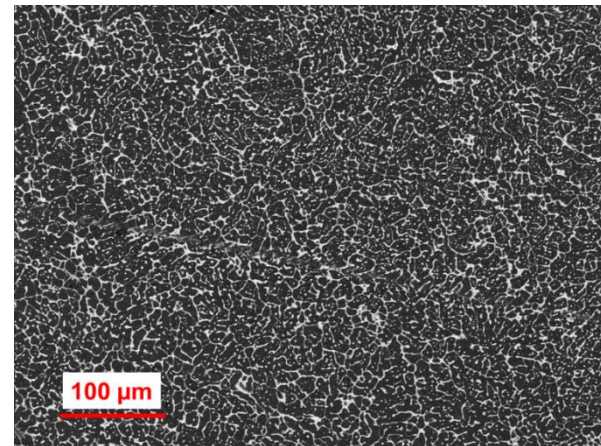
Significant difference between sample 6&7 and 8

Could be related to :

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

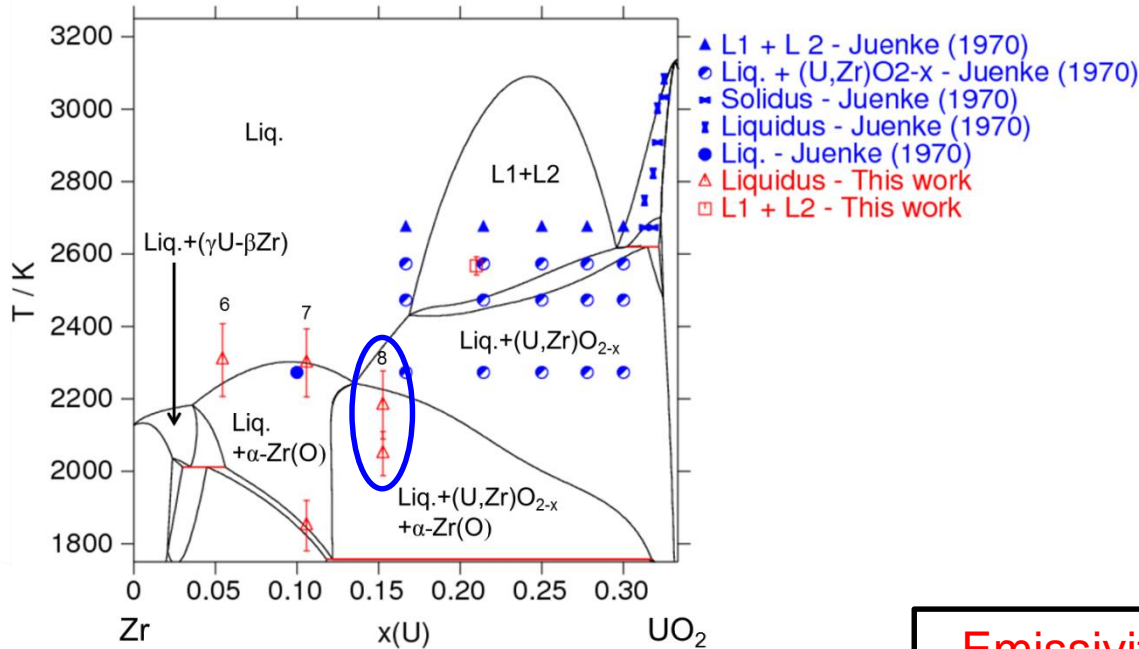


Sample 6



Sample 8

# Experimental results



A. Quaini, PhD Thesis, INP Grenoble 2015

## Zr-UO<sub>2</sub> isopleth section:

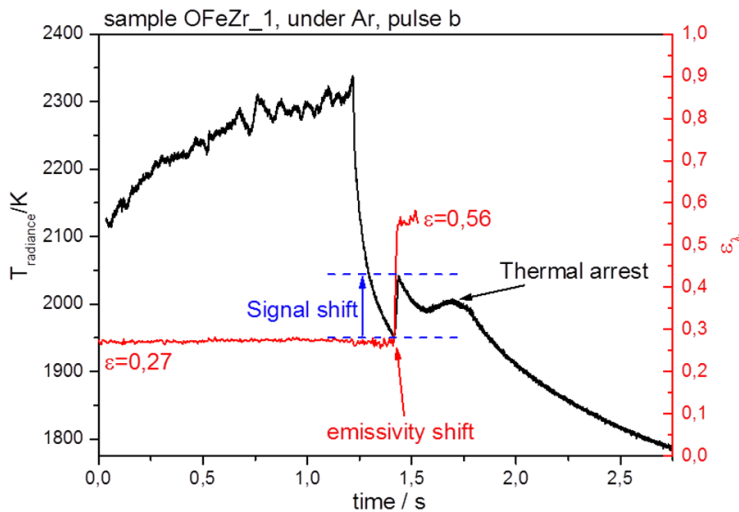
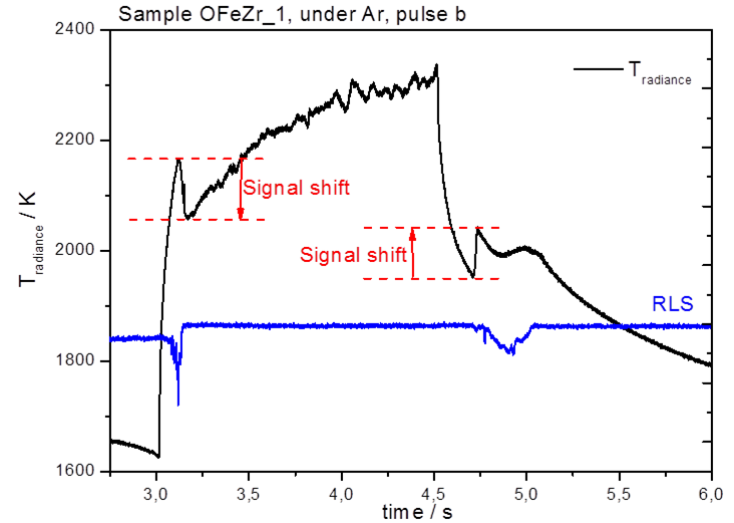
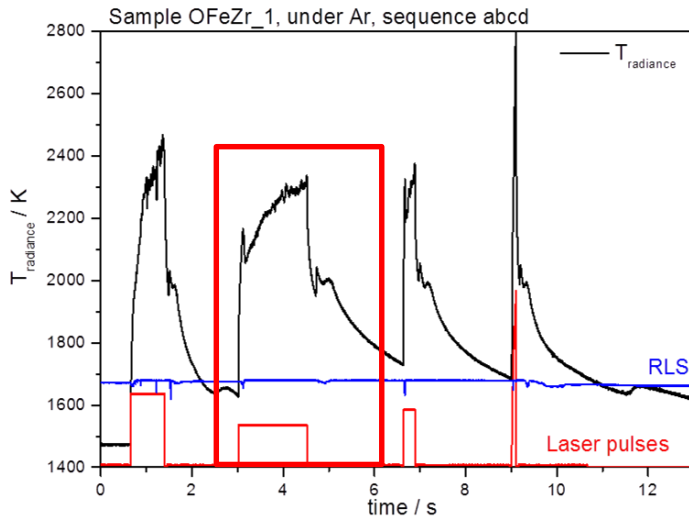
- Calculated using CEA database
- Laser heating data were not used for the assessment
- Good agreement between calculation and exp data on samples 6&7
- Disagreement with sample 8

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 <sup>st</sup> thermal arrest	2183	2343	2345
2 <sup>nd</sup> 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	$T_r$ / K	$\varepsilon(652 \text{ nm})$	True Temperature / K
	1 <sup>st</sup> inflection		1 <sup>st</sup> 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  $\varepsilon$  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

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

2 different ATTILHA configurations: **Contactless** → Aerodynamic levitation  
**Containerless**



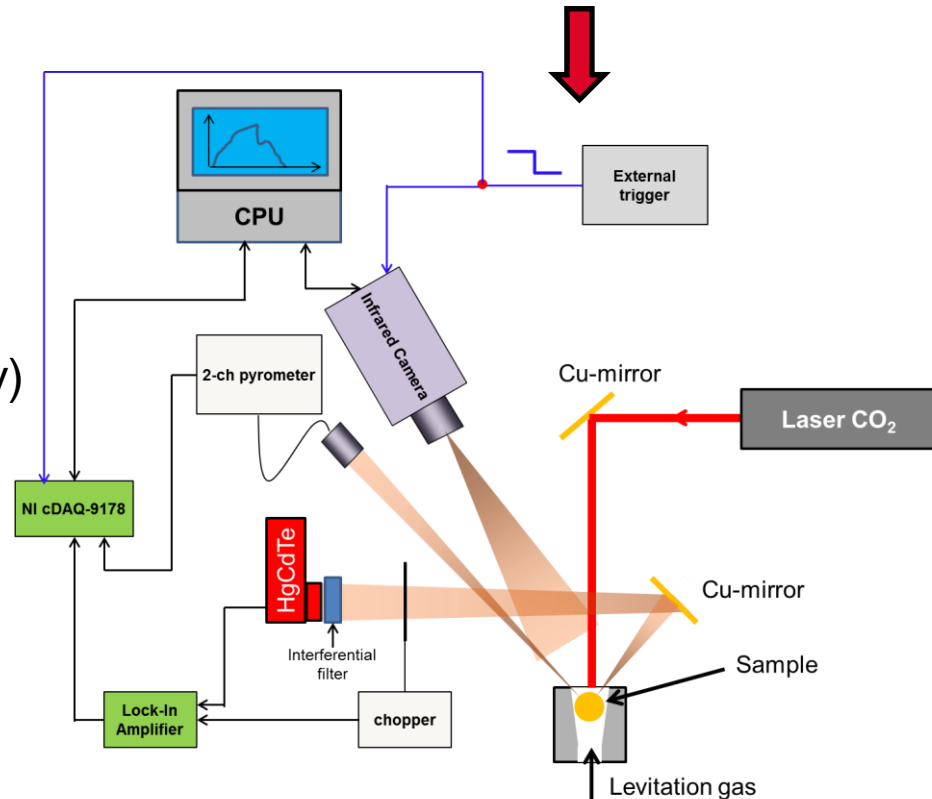
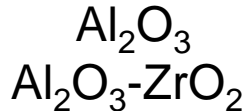
...the God's scourge

### Acquisition of data on corium systems

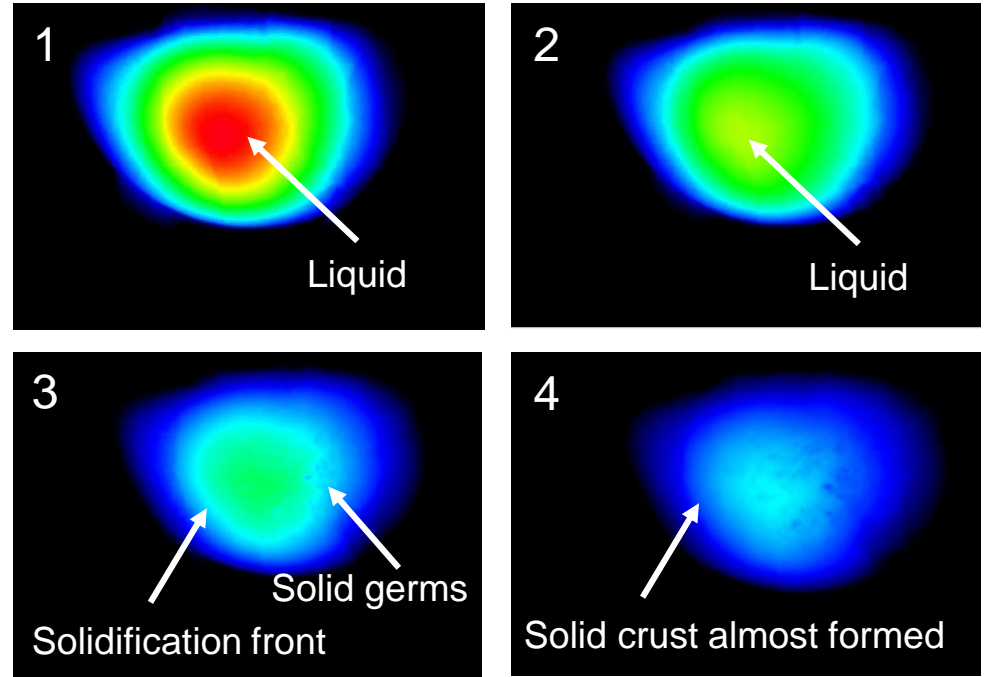
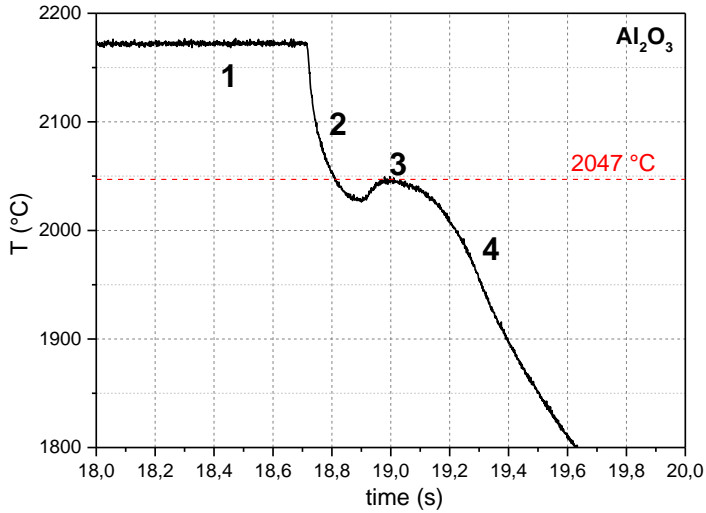
- Phase diagram data (liquidus, solidus)
- Thermo-radiative properties (IR emissivity)



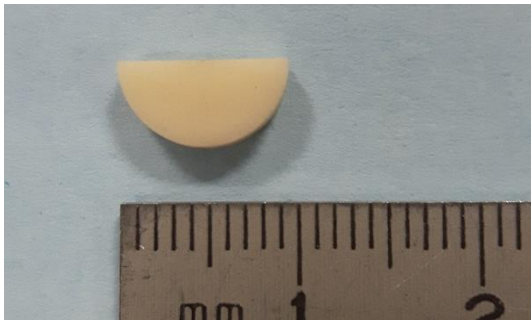
All the instruments are synchronized  
Validation on transitions in oxide systems



# Al<sub>2</sub>O<sub>3</sub> melting/solidification results



Before



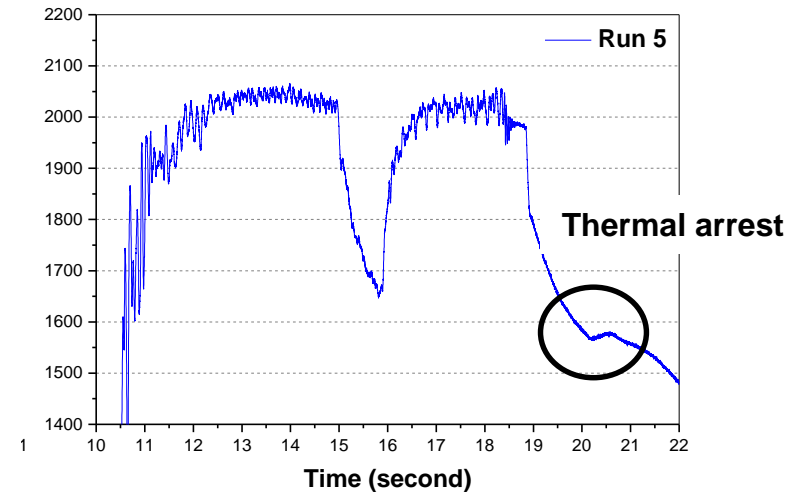
After



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  $\mu\text{m}$ )

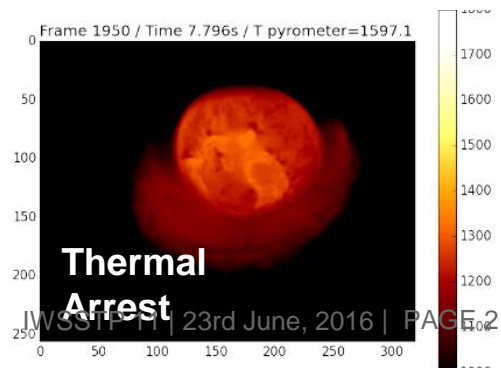
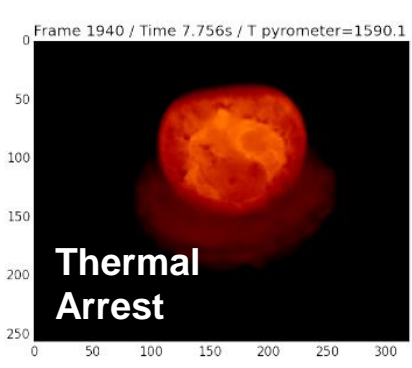
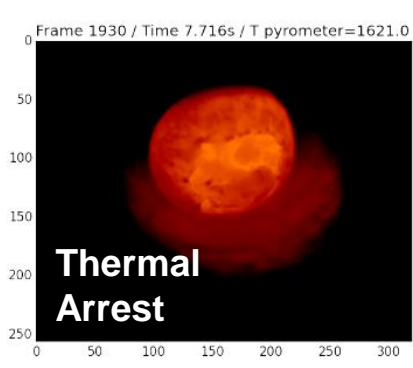
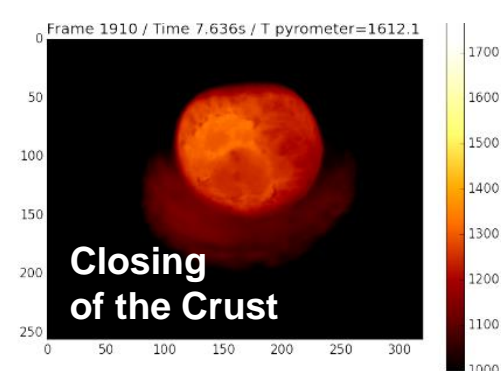
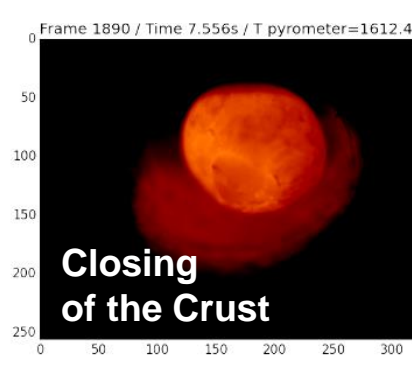
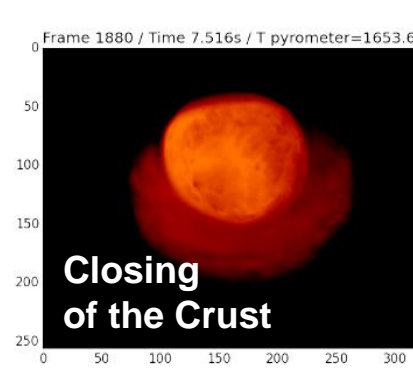
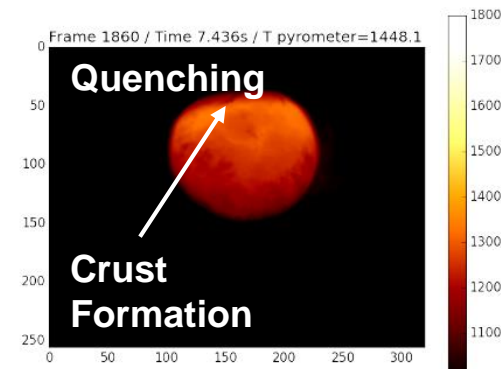
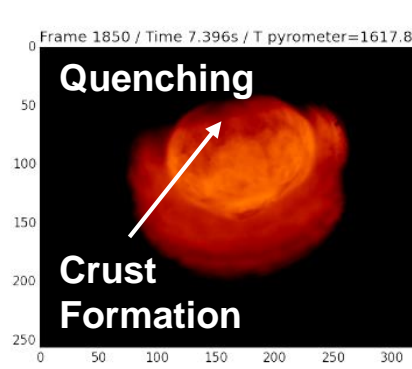
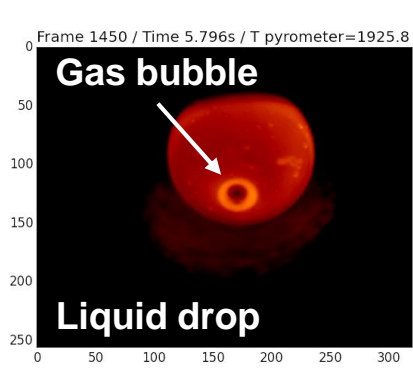
Calibrated from 300°C to 1500°C  
Extrapolated above 1500°C

Real speed: 250 Hz

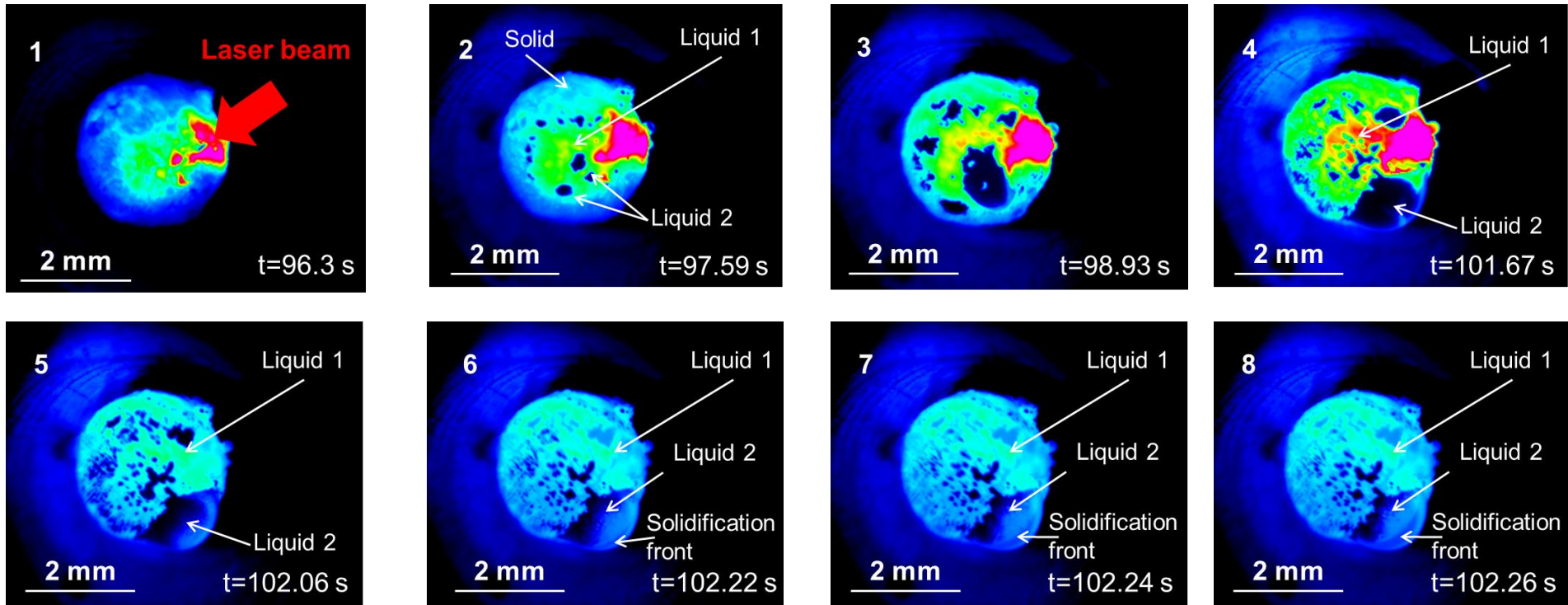
## COMMENTS

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

# cea den $Al_2O_3$ -CaO-ZrO<sub>2</sub> – Thermal arrests



## Miscibility gap



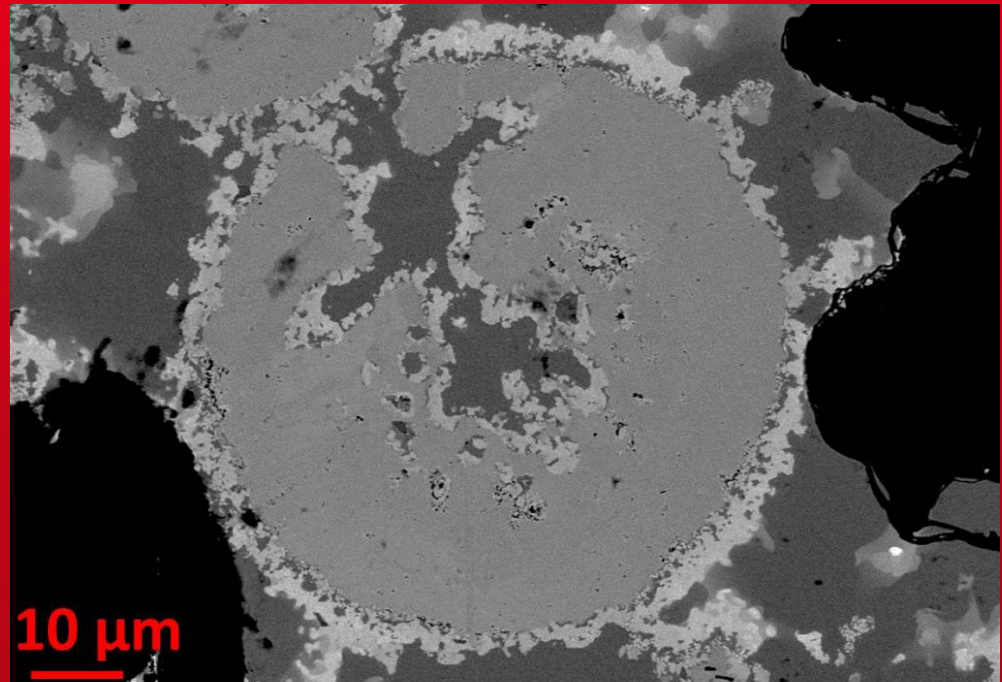
Blue: metallic liquid → less emissive

Green - Red: oxide liquid → highly emissive

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



# THANK YOU FOR YOUR ATTENTION



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