Development of ATTILHA - Experimental setup for high temperature investigations of U-containing systems (...soon)

A. Quaini, S. Gosse, T. Alpettaz, E. Lizon a Lugrin, C. Gueneau, S. Poissonnet, P. Bonnaillie

DE LA RECHERCHE À L'INDUSTRIE

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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

↓

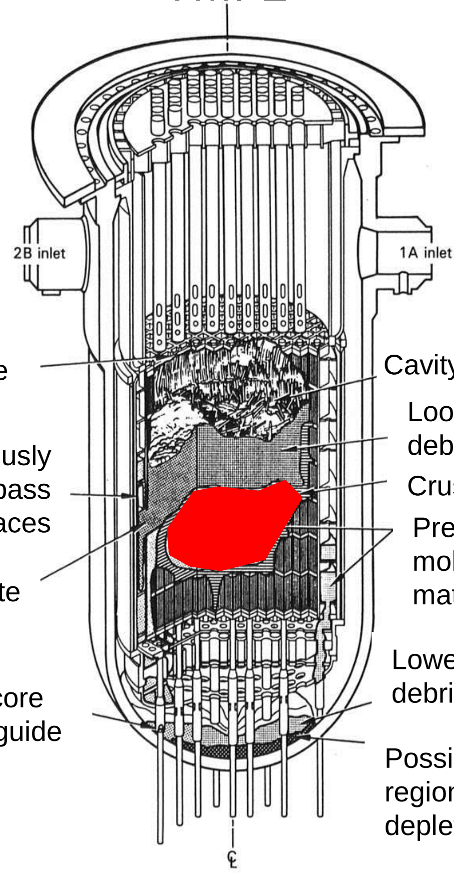
Fusion of the core and internal structure

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

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

TMI-2



PWR cross section

In-vessel core Melt configuration

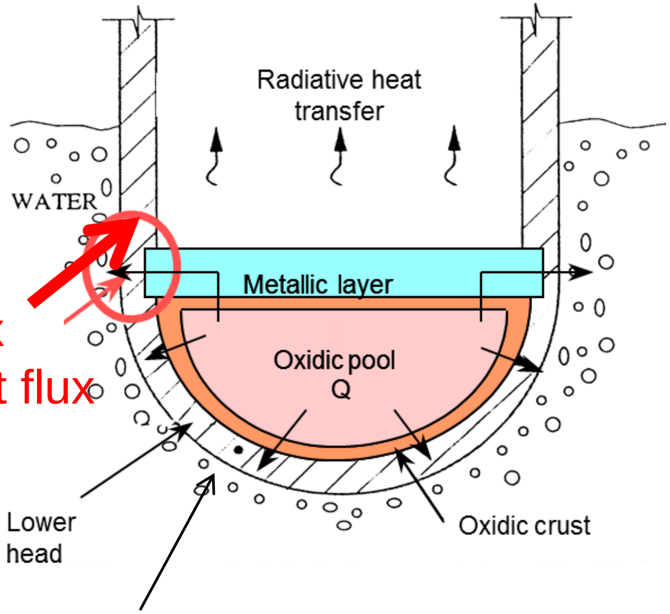
Issue 1: In-vessel core Melt configuration (Miscibility gap in the in-vessel corium liquid)

$\rho_{metallic} < \rho_{oxide}$ →

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

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

ρ =density



Max heat flux

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

Ex-vessel Core Melt configuration

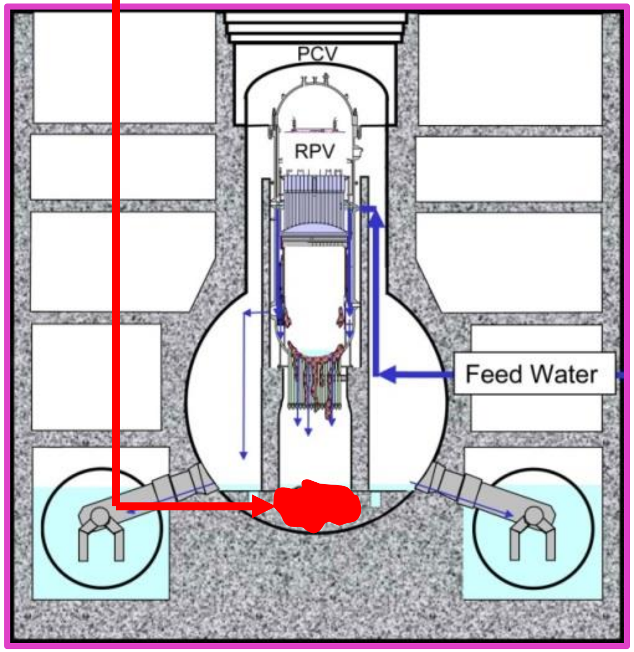
Issue 2: Ex-vessel core melt configuration

Once the reactor vessel has failed…
Last retention option: the Ex-vessel relocation of the molten core in a lateral core catcher
The molten core (T≈2400°C) pours on the containment concrete

The **Molten Corium Concrete Interaction** (MCCI) starts:

- The components of the concrete (CaO, SiO_2, Al_2O_3, MgO, H_2O, CO_2) are added to the already complex in-vessel system (U-Zr-O-Fe)
- As a first approximation the **Fe-U-Zr-Al-Ca-Si-O** system is representative of an ex-vessel corium

Fukushima



A better thermodynamic description of the ex-vessel corium sub-systems is needed to improve the thermal and thermo-hydraulics codes accuracy

Need of exp. data at very high temperatures 1500°C ≤ T ≤ 3200°C

Development of a specific exp. setup

EXPERIMENTAL TECHNIQUES

Development of ATTILHA Setup

ATTILHA Setup

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

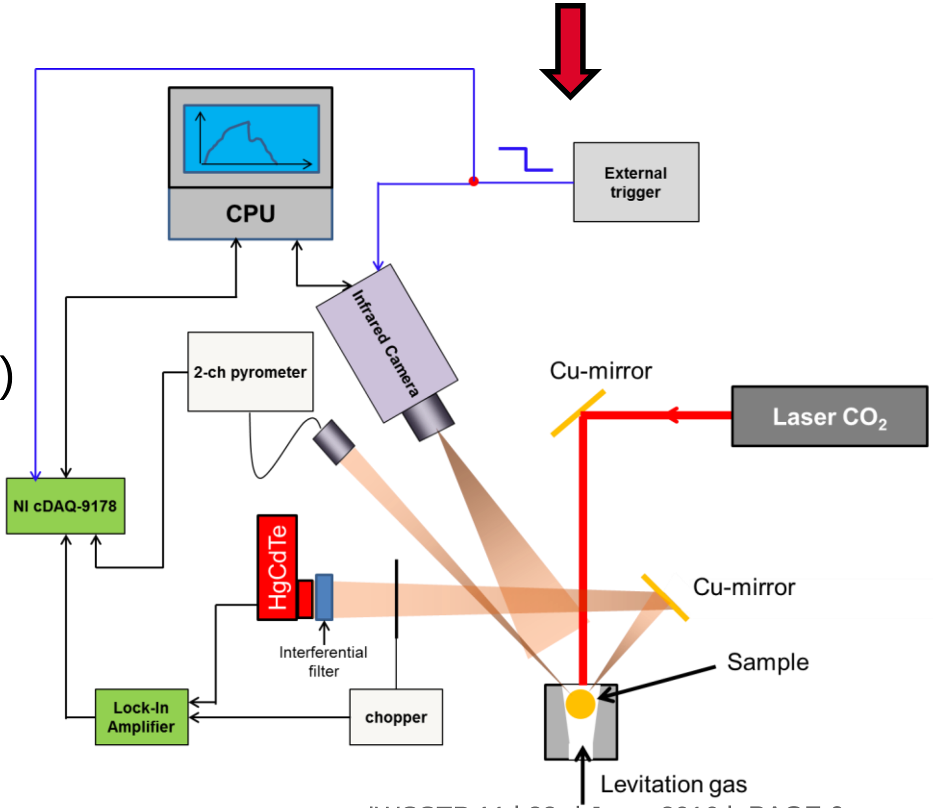
ATTILHA: Development of a setup for high solid/liquid transitions

2 different ATTILHA configurations: Contactless → Aerodynamic levitation
Containerless

Acquisition of data on corium systems

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

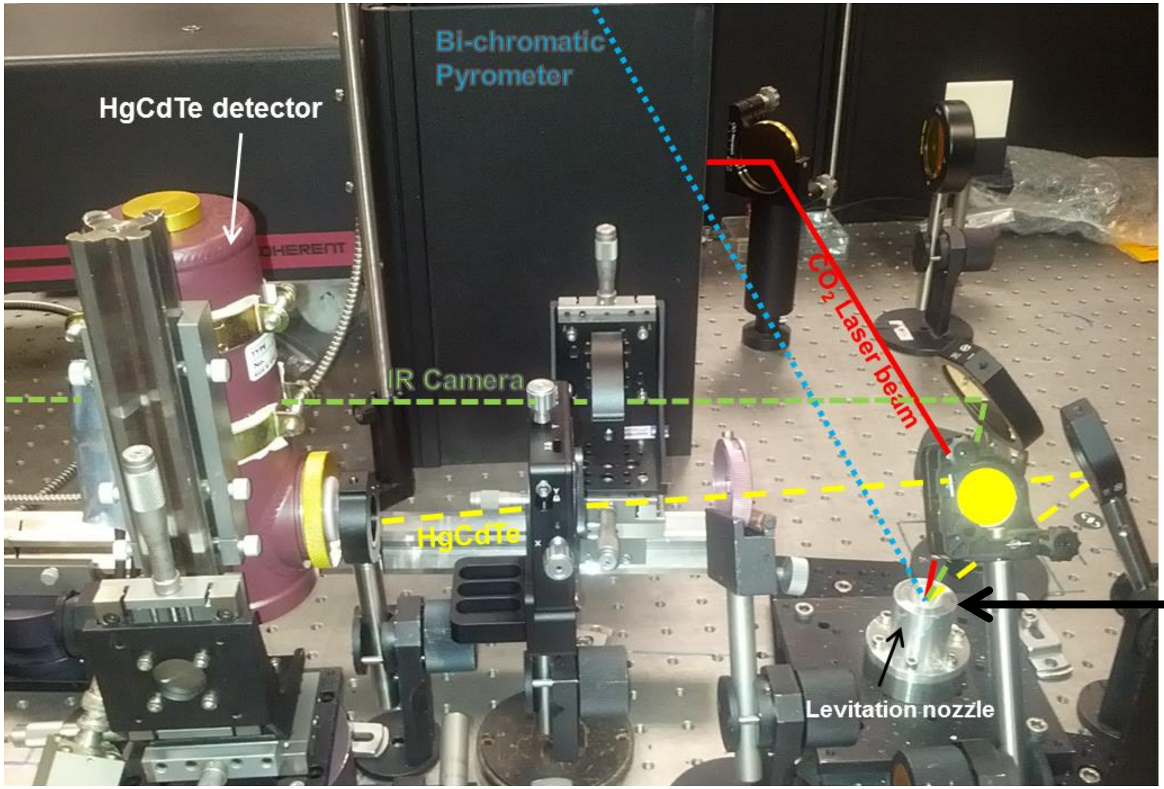
All the instruments are synchronized
Validation on transitions in oxide systems
Al_2O_3
Al_2O_3-ZrO_2

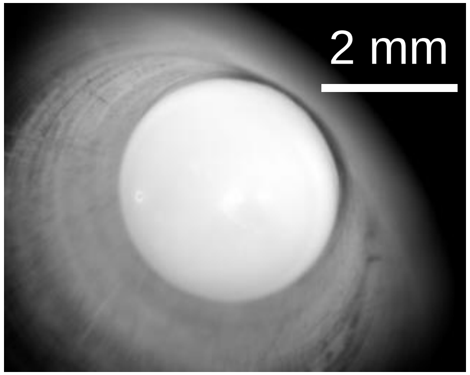


ATTILHA setup
Aerodynamic levitation

Heating device:	CO_2 laser
Temperature measurement:	HgCdTe IR detector (2 - 22 µm, filtered by narrow band filters) Bichromatic pyrometer (0.8-1.05 µm)
Image recording:	High speed calibrated (300°C-1500°C) IR Camera (@ 3.99 µm)

Aerodynamic levitation configuration in a convergent/divergent nozzle





Levitating liquid Al_2O_3 droplet in the nozzle

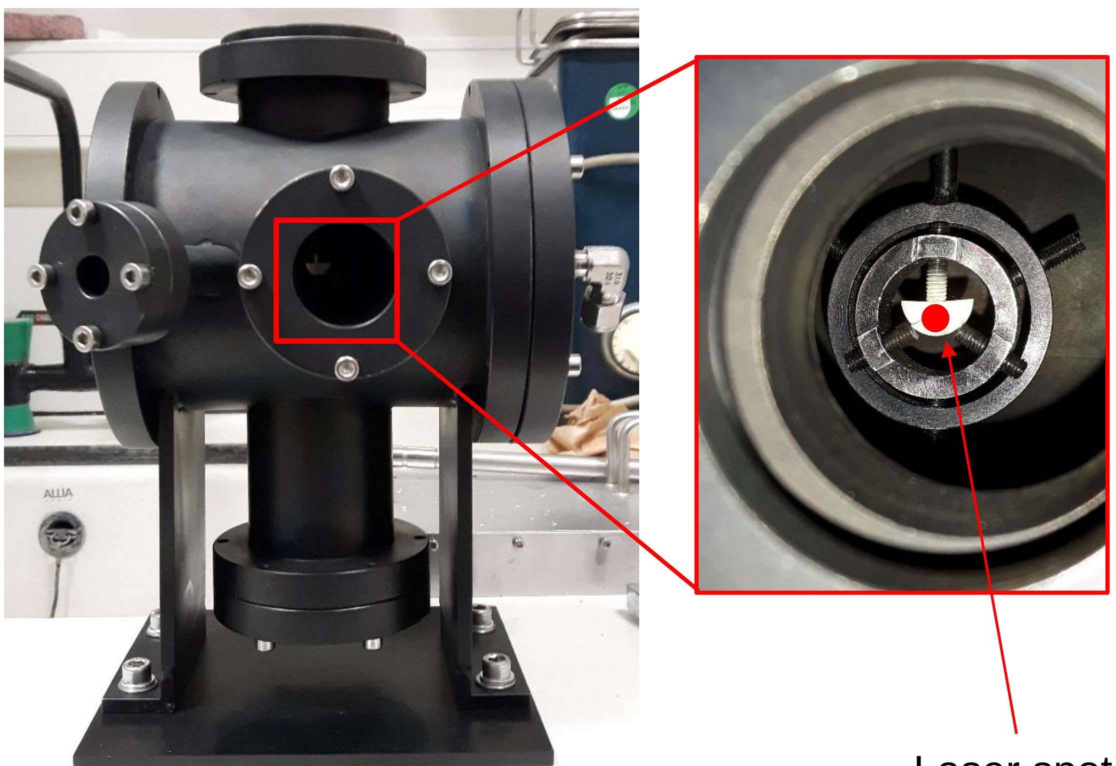
ATTILHA setup

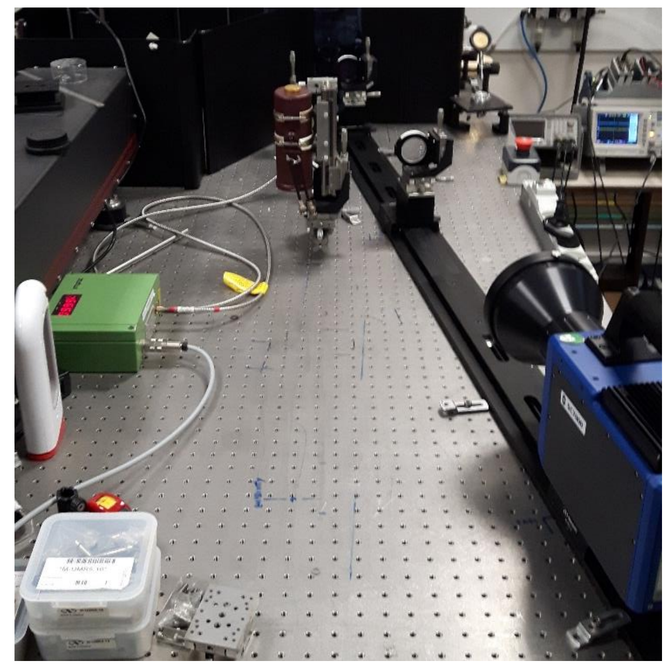
Containerless

Heating device: CO_2 laser

Temperature measurement: HgCdTe IR detector (2 - 22 µm, filtered by narrow band filters)
Bichromatic pyrometer (0.8-1.05 µm)

Image recording: High speed calibrated (300°C-1500°C) IR Camera (@ 3.99 µm)





Development approach

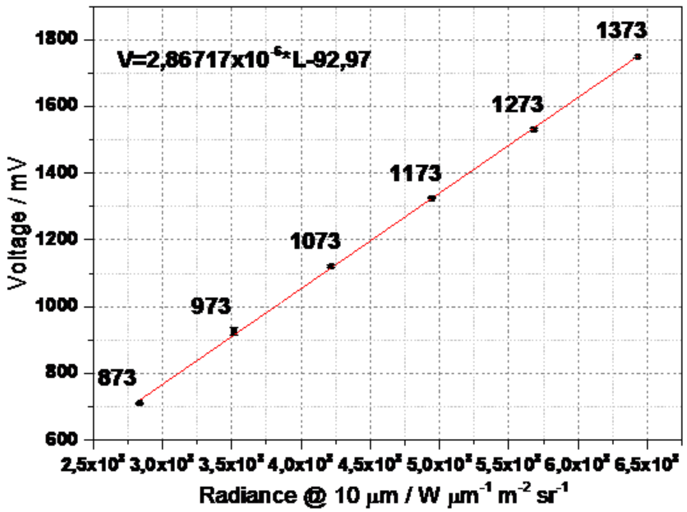
→ Optimisation of the instrumentation position on the optical table

CO_2 laser – HgCdTe detector – Rapid infrared camera – Pyrometer

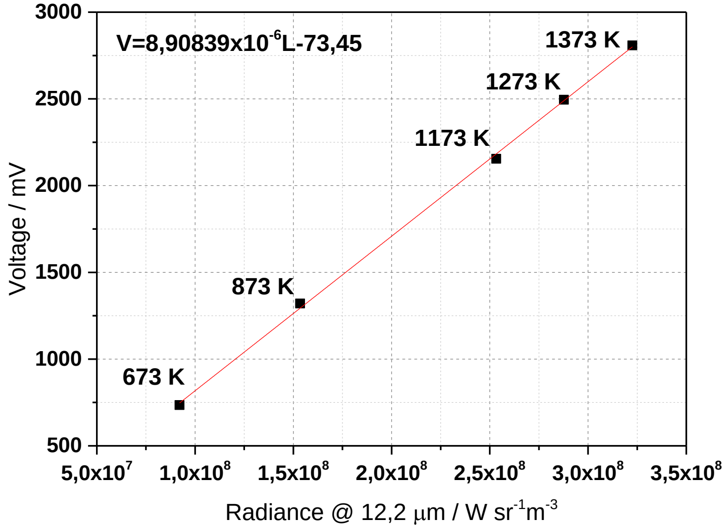
→ Calibration at 10 µm and 12.2 µm

Christiansen point Al_2O_3: 10 µm; ZrO_2: 12.7 µm

@ 10 µm



@ 12,2 µm



→ Error analysis (collaboration with Institut de Mathématiques de Toulouse)

Development approach

→ Optimisation of the instrumentation position on the optical table

CO_2 laser – HgCdTe detector – Rapid infrared camera – Pyrometer

→ **Verification of the manufacturer calibration against a qualified black-body**

→ **Python image processing routine for emissivity estimation (IMT-Toulouse)**

Development approach

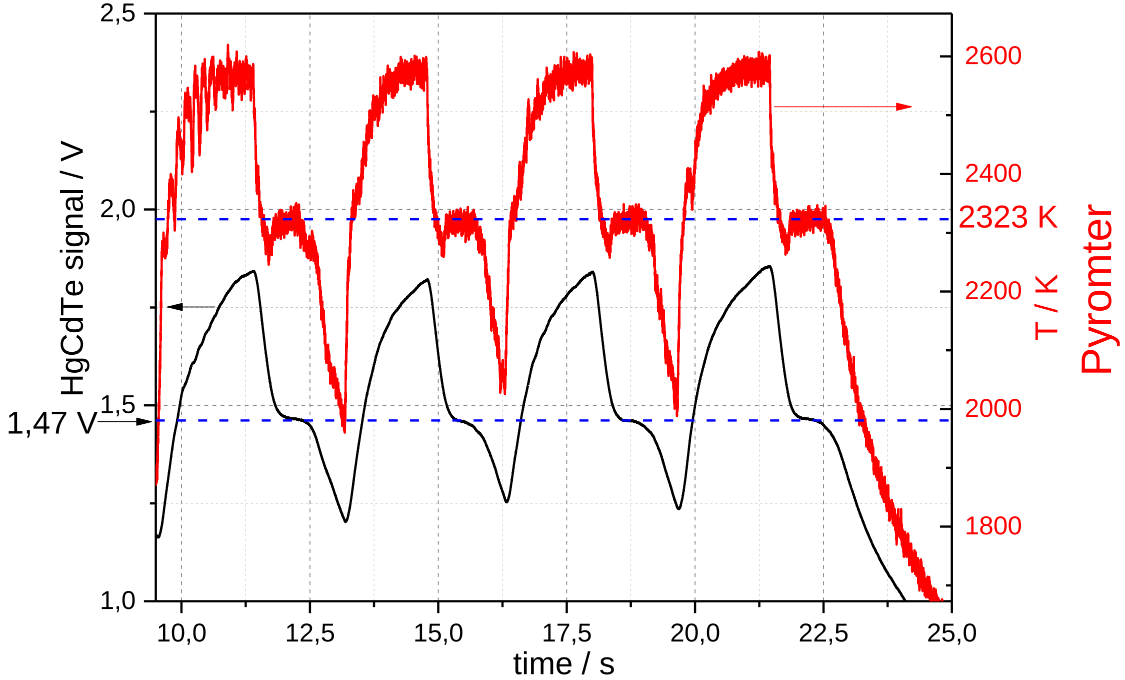
→ Optimisation of the instrumentation position on the optical table

CO_2 laser – HgCdTe detector – Rapid infrared camera – Pyrometer

→ Validation of the setup

→ Al_2O_3 (T=2323±25 K)

4 heating/cooling runs

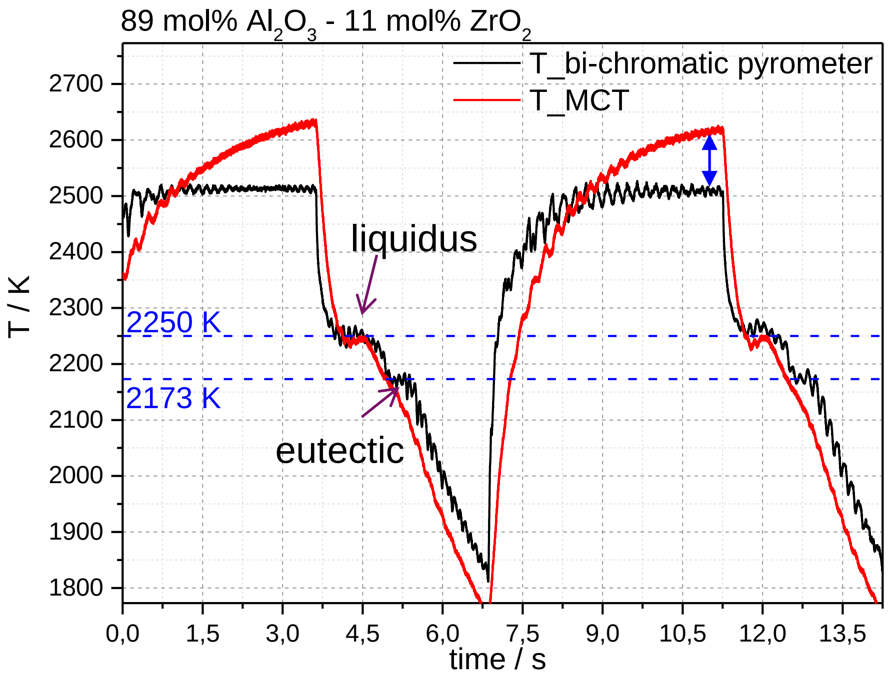


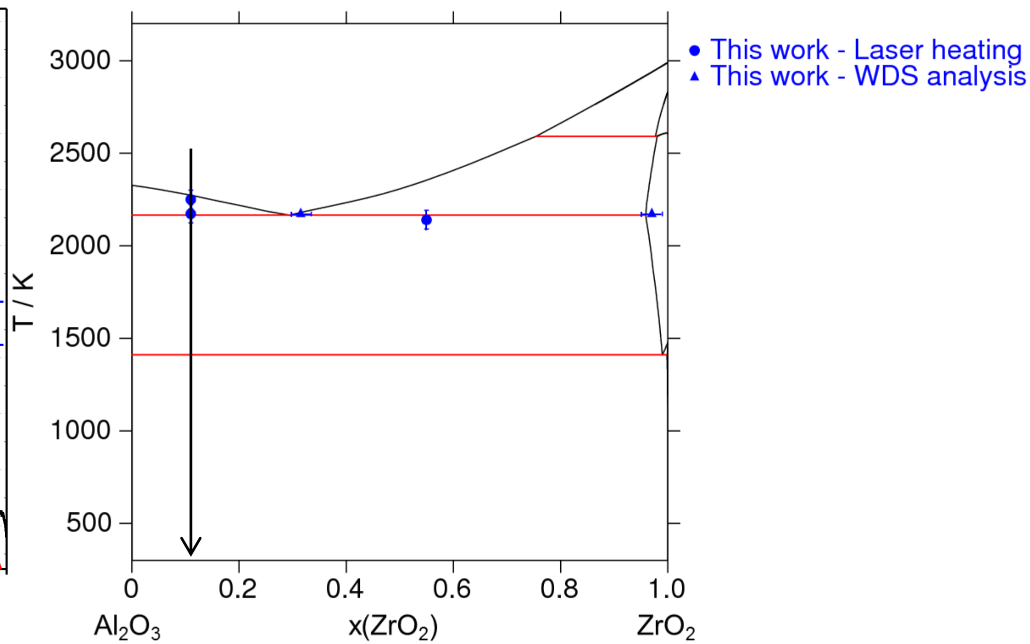
EXPERIMENTAL RESULTS

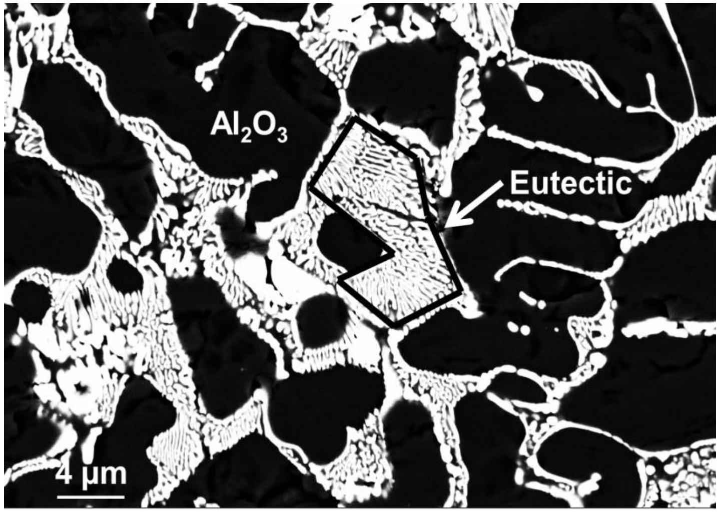
Some Preliminary Results using ATTILHA setup on the systems:

Al_2O_3-ZrO_2

Results: Al_2O_3-ZrO_2







$T_{liquidus}$ + $T_{eutectic}$ → good agreement with literature

Eutectic composition → good agreement with literature

↕ ⟶ Semi-transparent behaviour

EXPERIMENTAL RESULTS

Some Preliminary Results using ATTILHA setup on the systems:

Al_2O_3-CaO-ZrO_2

Preparation of the samples Al_2O_3-CaO-ZrO_2

Samples are made from sintered pellets of raw oxide powders:

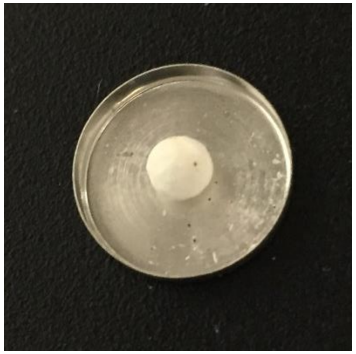
Al_2O_3/CaO/ZrO_2 (52-41-07 in mol %) +
≈ 1% mass. Of Zinc Stearate

No mass loss during sintering for Al_2O_3-CaO-ZrO_2

Al_2O_3/CaO/ZrO_2 Sample

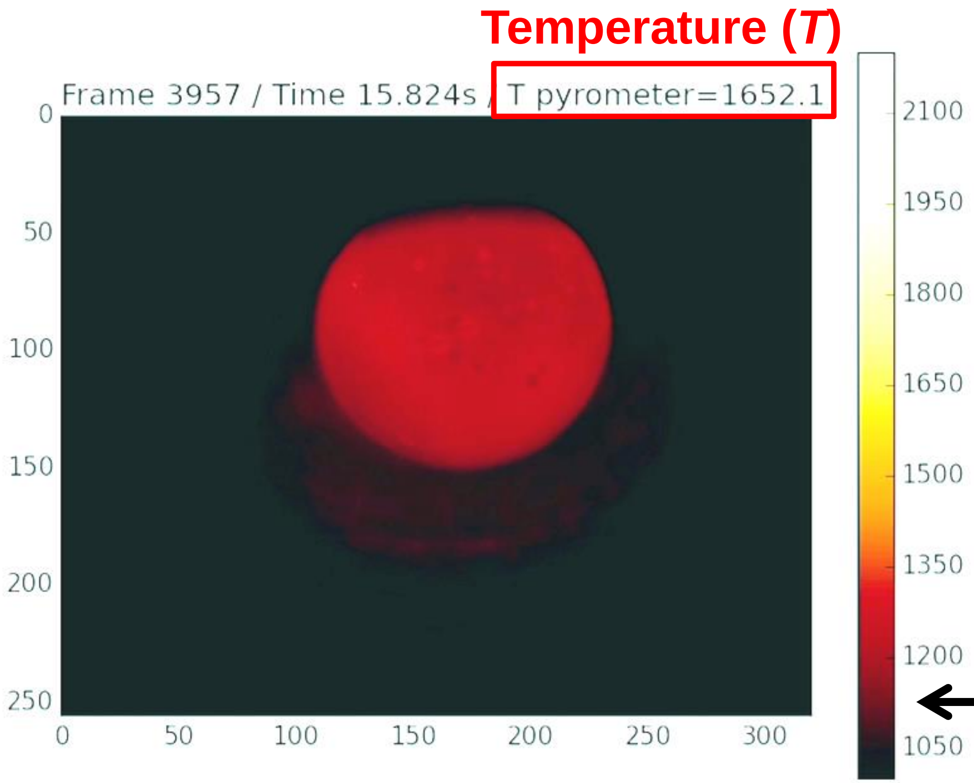
After sintering

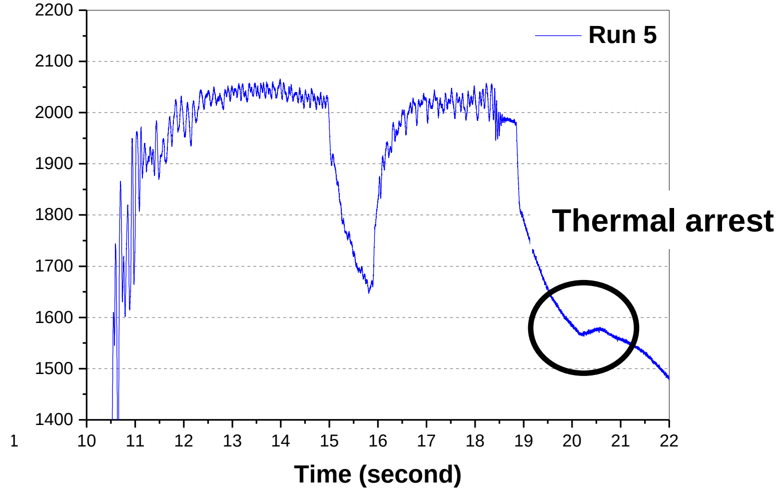
After laser melting



Al_2O_3-CaO-ZrO_2 – Radiance results

Development of a Python code for image processing and emissivity estimation





Radiance Temperature (T_L) scale (at 3.99 µm)

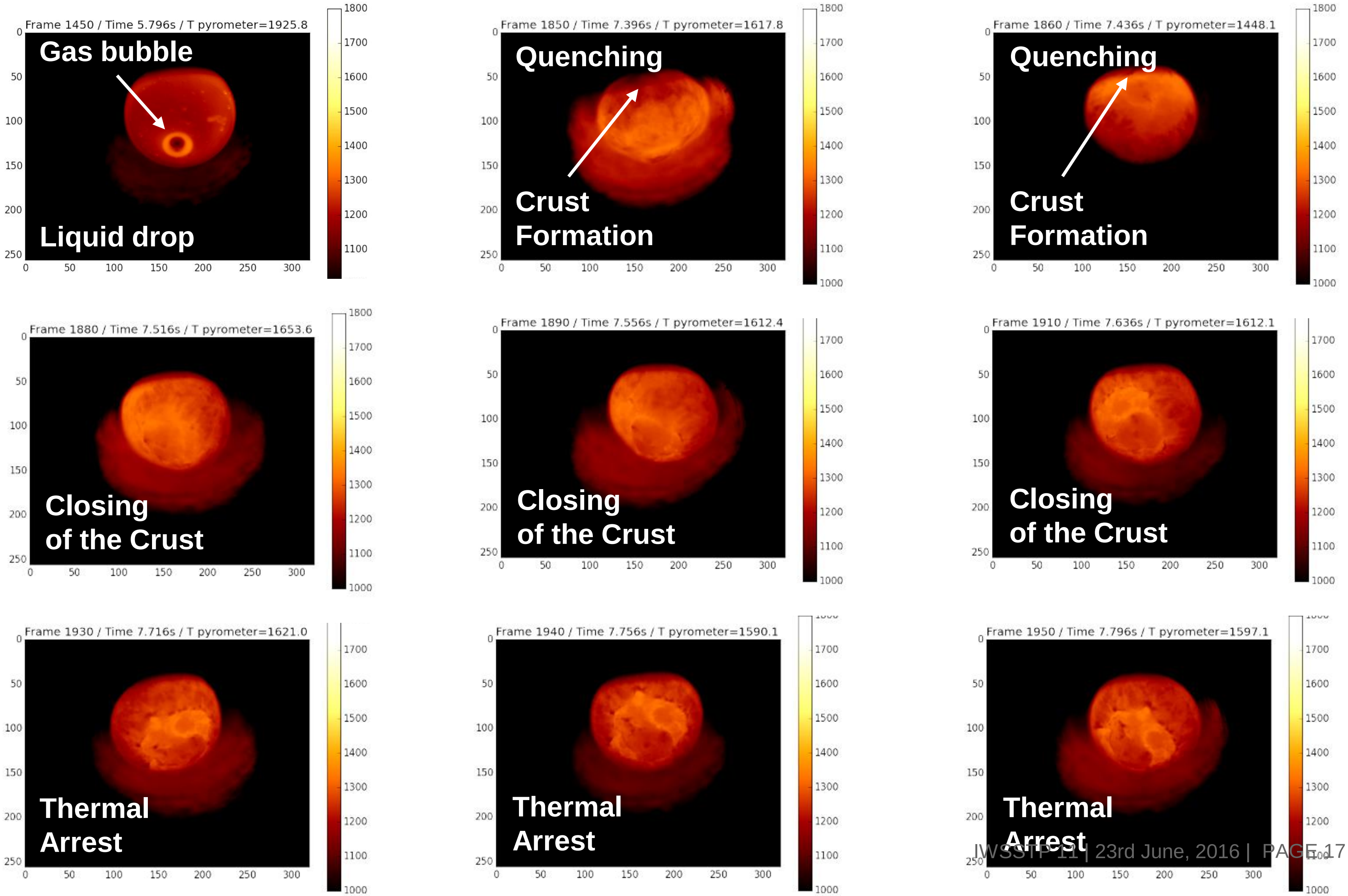
Real speed: 250 Hz

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

COMMENTS

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

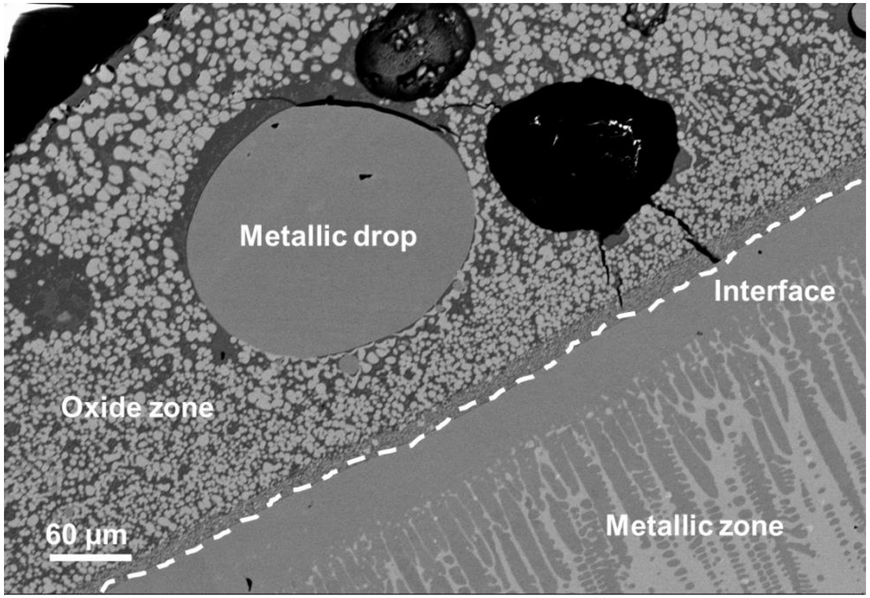
Al_2O_3-CaO-ZrO_2 – Thermal arrests



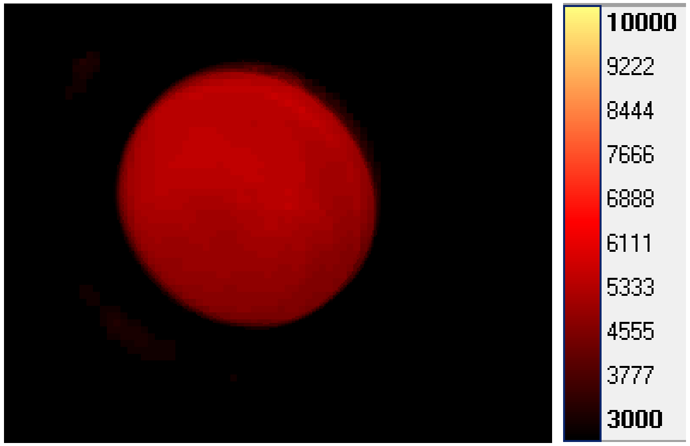
EXPERIMENTAL RESULTS

Miscibility gap in the Fe-Zr-O system

Miscibility gap in the Fe-Zr-O system



Infrared camera footage



Digital level

Real speed 200 Hz
Video player 12.5 Hz

New result

Starting composition: $Fe_{0.85}Zr_{0.15}$

↓ Levitation gas: He

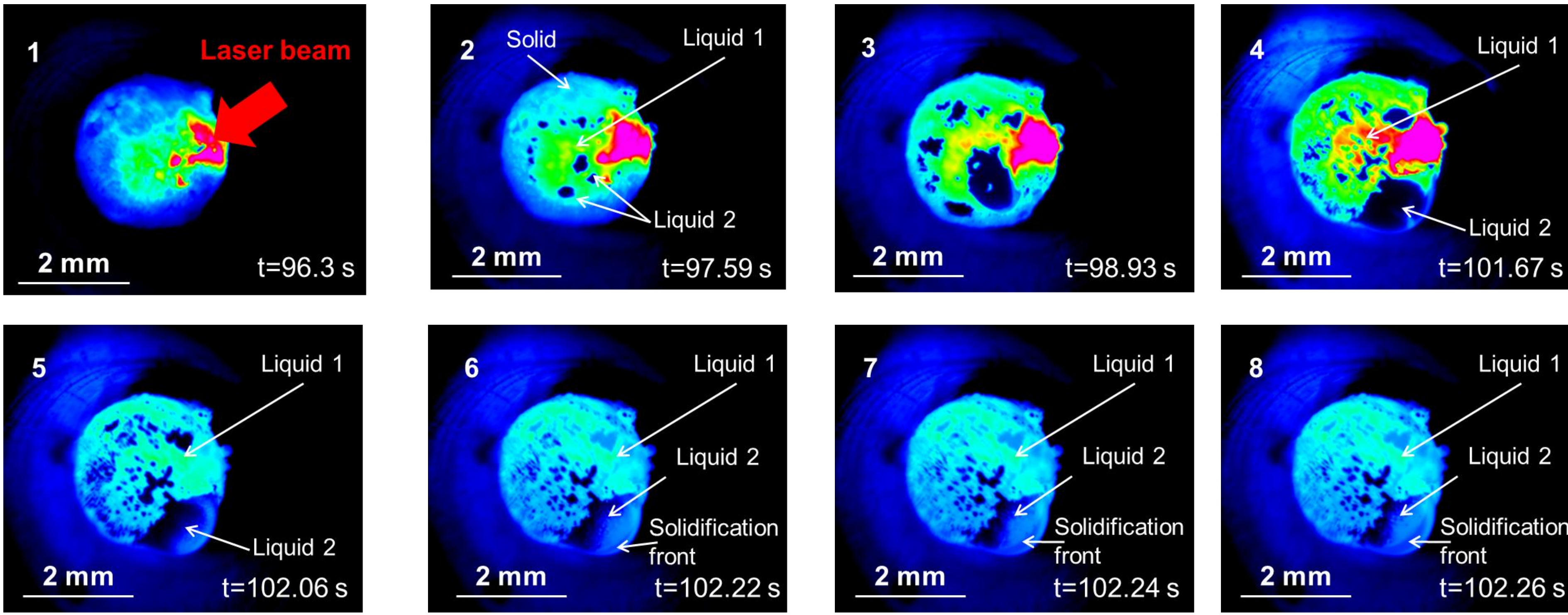
Composition moved into the ternary Fe-Zr-O system

tie-line: $Fe_{0.97}O_{0.03}$ – $Fe_{0.05}Zr_{0.32}O_{0.63}$

Observation of dynamic phenomena:
→ **Formation of 2 liquids in-situ**

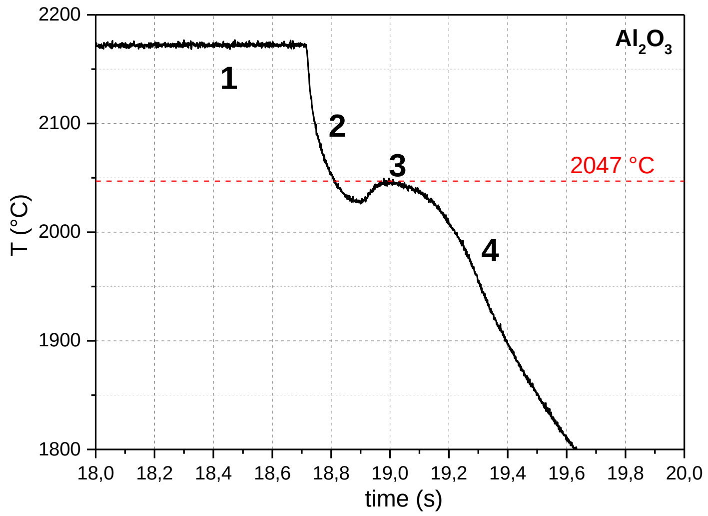
Estimation of the emissivity ratio between the two liquids
→ $\varepsilon_{oxide} \sim 2\varepsilon_{metal}$

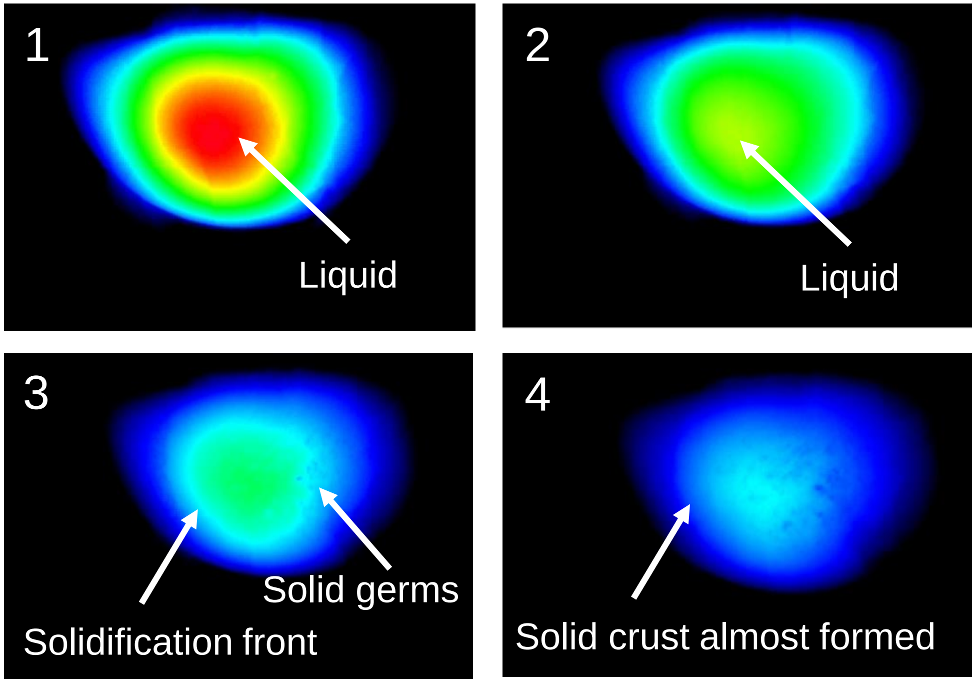
Miscibility gap



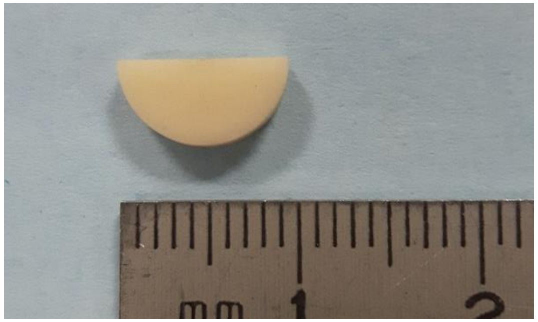
CONTAINERLESS CONFIGURATION

Al_2O_3 melting/solidification results





Before



After

Thermal gradients
Solidification front progression
Semi-transparency

Next step… **U-containing samples**

Conclusion and Perspectives

ATTILHA a laser heating setup at CEA Saclay

- Aerodynamic levitation / containerless
- Rapid measurements: Fast heating/quenching
- High temperature
- Levitation gas → Reducing or oxidizing conditions
- Gas flow in the experimental vessel → Red/Ox

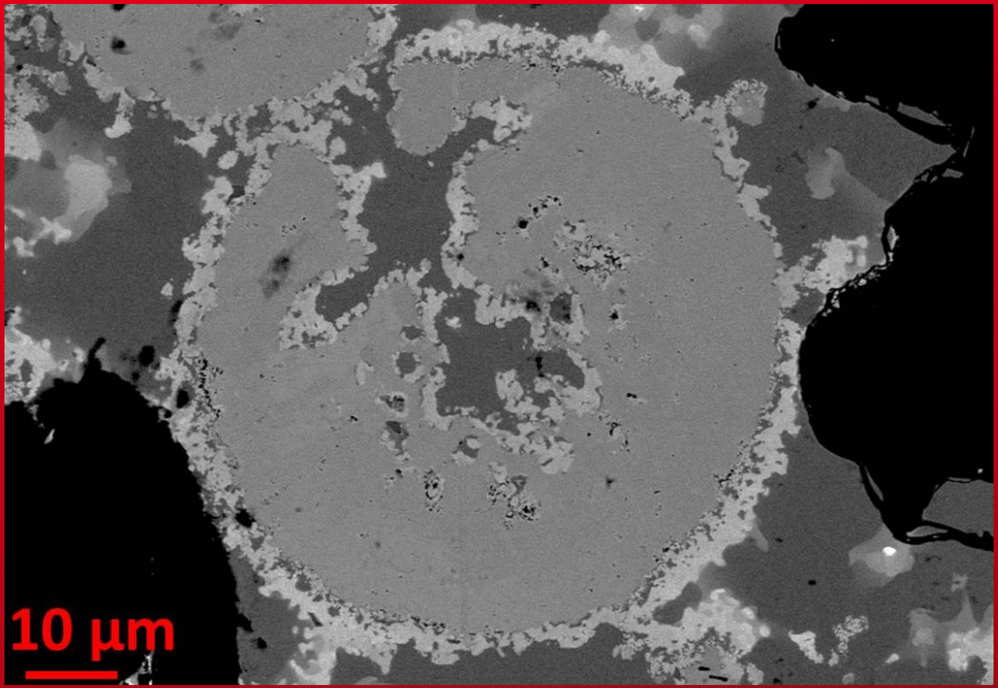
High temperature dynamic phenomena
- Solid/liquid transitions in Ex-vessel coria
- Miscibility gap in Fe-Zr-O

Validation on oxide systems
Al_2O_3 & Al_2O_3-ZrO_2
Preliminary results on the corium sub-systems
Fe-Zr-O
Al_2O_3-CaO-ZrO_2
Al_2O_3-SiO_2-ZrO_2

Open questions
- Very high temperature references?
- Supplementary T monitoring?
- Thermodynamic equilibrium?
- Influence of $P(O_2)$?

Future developments
Nuclearisation of the setup → investigation of **Uranium** containing samples
Ultra rapid camera (5 kHz) → viscosity and density measurement

THANK YOU FOR YOUR ATTENTION



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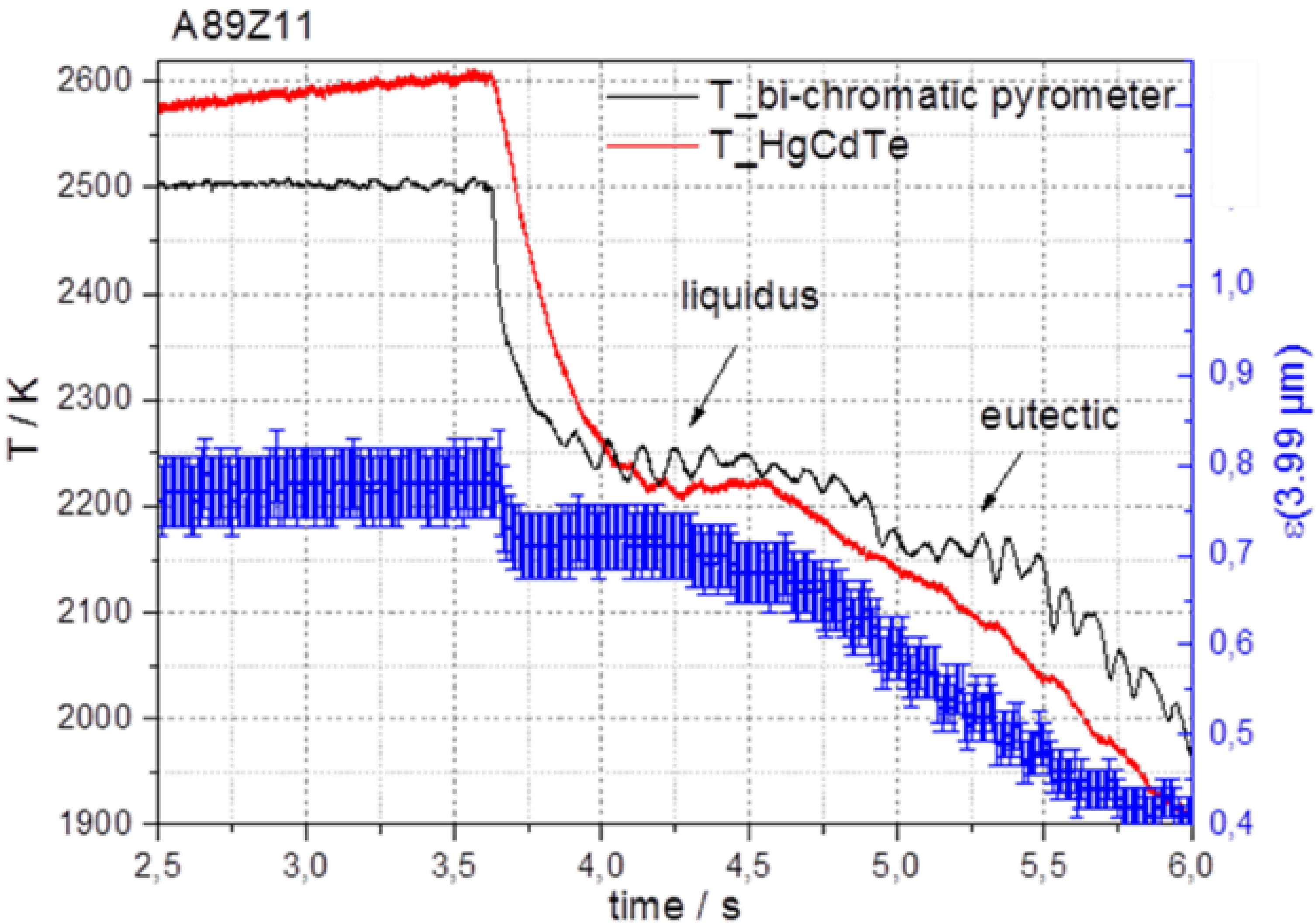
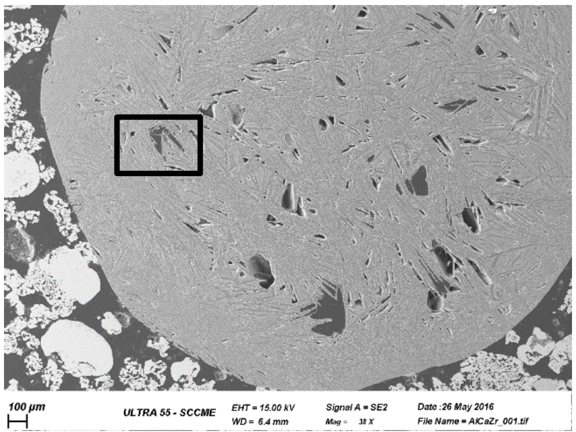
Département de Physico-Chimie
SCCME
Laboratoire de Modélisation
Thermodynamique et Thermochimie
CEA de Saclay

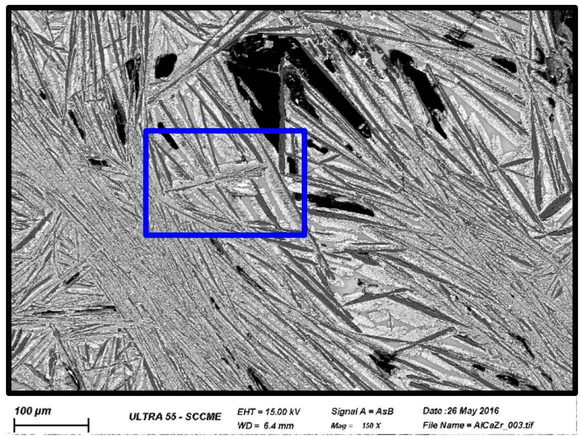
Emissivity estimation at 3.99 µm

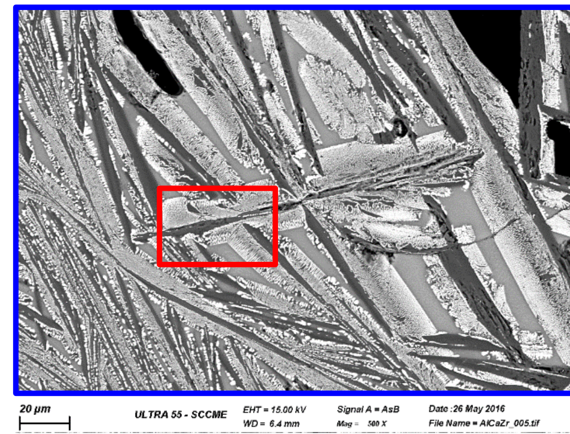


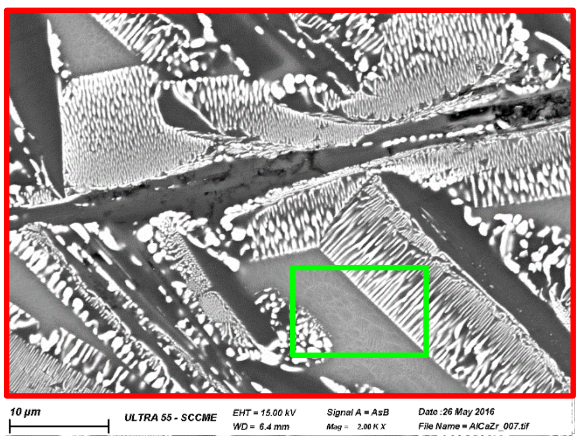
Analyses Al_2O_3-CaO-ZrO_2

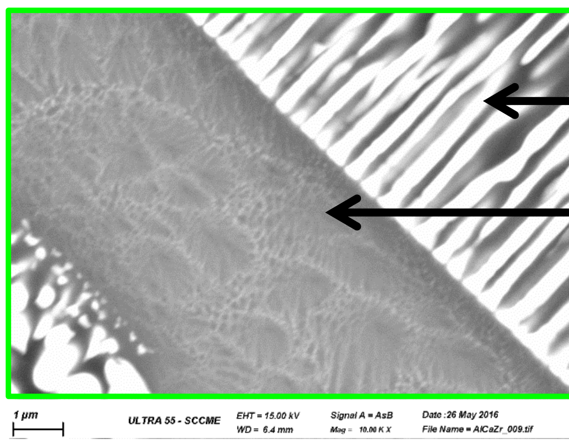
SEM observations and EDS analyses (Secondary and Back Scattered)











$(Al_{0.28},Ca_{0.13},Zr_{0.59})O_2$

Al_2O_3-CaO (+ ε Zr)

$\approx Al_{21.88}\ Ca_{14.00}\ Zr_{1.92}\ O_{62.20}$

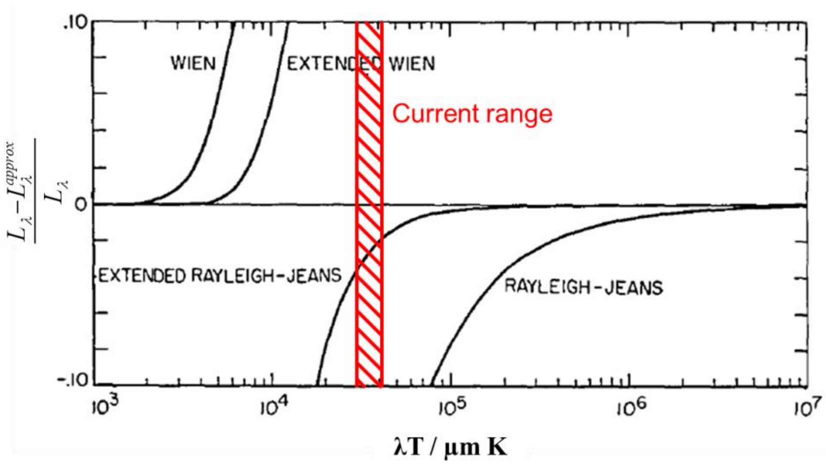
Oxygen is only estimated from EDS

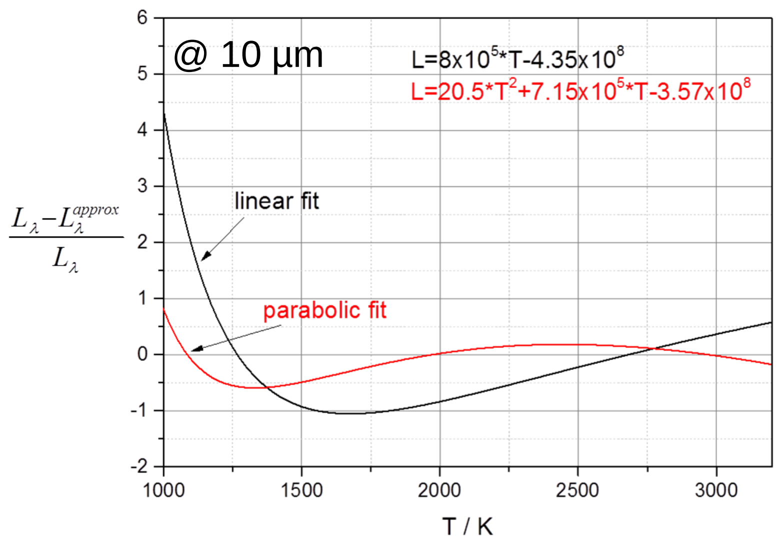
Al_2CaO_4 + ε $CaZrO_3$???

Further analyses will be conducted

Error analysis

Error analysis (collaboration with Institut de Mathématiques de Toulouse)





$$T \in \left[\hat{T} - \frac{K_2}{K_1}(e'+2\sigma) - 2\frac{K_3}{K_1}\sigma_\varepsilon , \hat{T} + \frac{K_2}{K_1}(e'+2\sigma) + 2\frac{K_3}{K_1}\sigma_\varepsilon \right]$$

For example, the true temperature of a sample with an estimated emissivity 0.9±0.045 and an estimated radiance temperature of 2000 K is 2158±98 K with a probability of 0.9025. It must be pointed out that 86 % of the reported error bar is due to the estimated emissivity.

Error analysis (collaboration with Institut de Mathématiques de Toulouse)

$$\left|\hat{T}_\lambda - T_\lambda\right| \le \frac{e}{2cT_{\min} + b} = e' = 4.2\,K$$

Maximum deterministic error on T_λ

$$T_\lambda \in \left[\hat{T}_\lambda - e' - 2\sigma, \hat{T}_\lambda + e' + 2\sigma\right]$$

With a probability of 95 % and considering the most conservative case

$$T \in \left[\hat{T} - \frac{K_2}{K_1}(e'+2\sigma) - 2\frac{K_3}{K_1}\sigma_\varepsilon, \hat{T} + \frac{K_2}{K_1}(e'+2\sigma) + 2\frac{K_3}{K_1}\sigma_\varepsilon\right]$$

With a probability of 90.25 %

$$\left|\hat{T} - T\right| \le \frac{K_2}{K_1}\left|\hat{T}_\lambda - T_\lambda\right| + \frac{K_3}{K_1}\left|\varepsilon - \hat{\varepsilon}\right|$$

$$\begin{cases} K_1 = \hat{\varepsilon}(b + 2cT_{\min}) \\ K_2 = b + 2cT_{\lambda,\max} \\ K_3 = a + bT_{\grave{u}ax} + cT_{\max}^2 \end{cases}$$