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Oxide phase characterization in simulated high burn-up UO_2 fuels in the early stages of a nuclear severe accident

MRS Spring Meeting 2018 | Claire Le Gall
CEA Cadarache | DEN | DEC | SA3E | LAMIR

C. Le Gall¹, E. Geiger², O. Proux³, M. Rovezzi³, P.L. Solari⁴, M. Hunault⁴, V. Klosek¹, C. Riglet-Martial¹, J. Léchelle¹, F. Audubert¹, Y. Pontillon¹, J.-L. Hazemann⁵

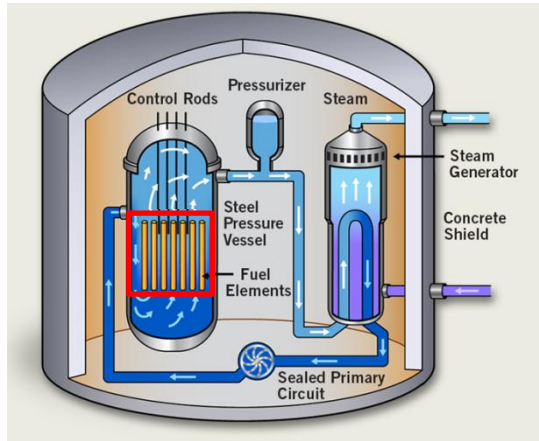
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April 2-6, 2018 – Phoenix, Arizona

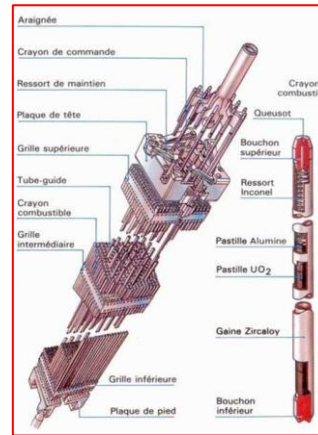


NUCLEAR FUEL FISSION

Fission products



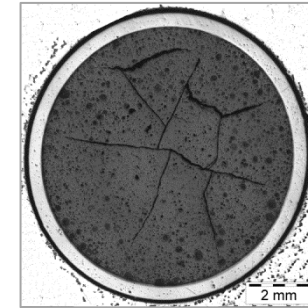
Pressurized Water Reactor (PWR)



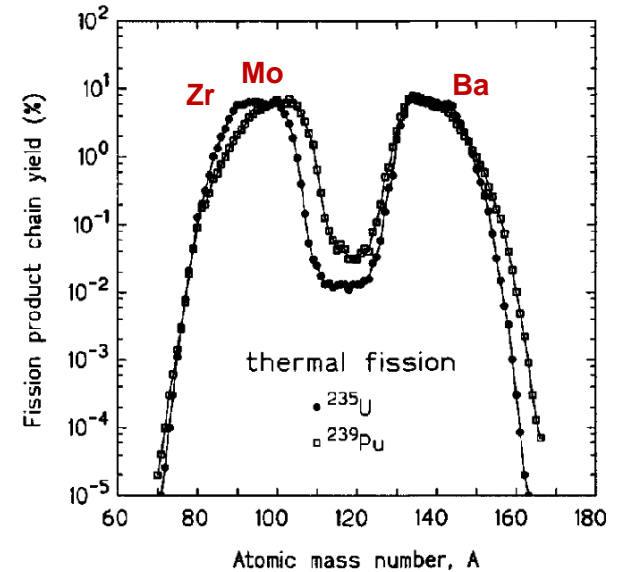
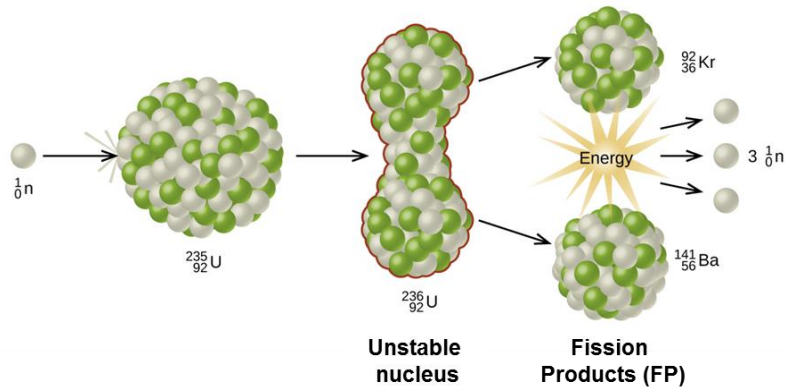
Fuel assembly



UO₂ or (U,Pu)O₂ fuel in its Zr alloyed cladding



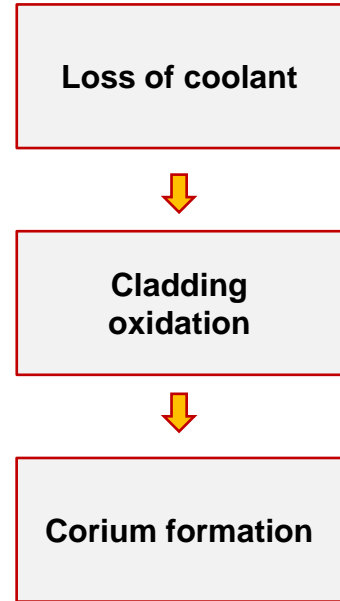
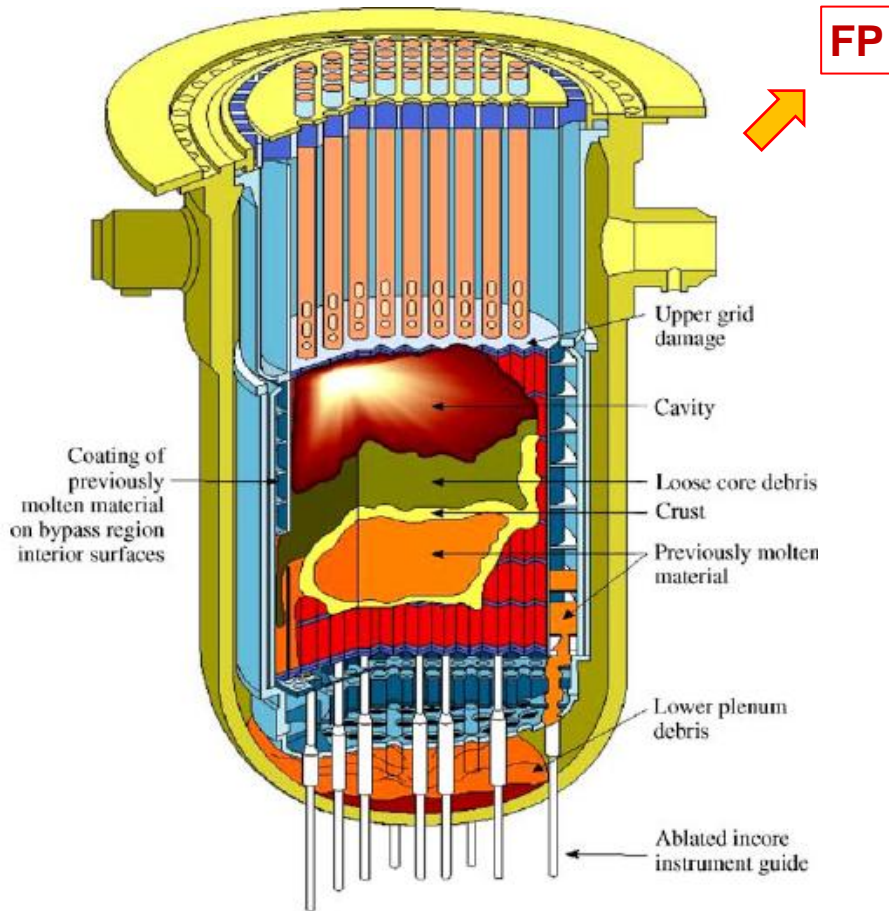
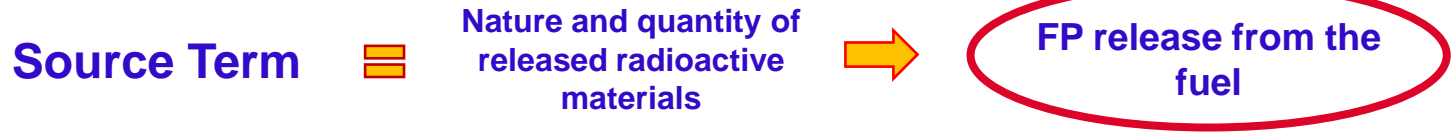
Fission yield: FP most probably produced



Fission occurs in nuclear fuel pellets

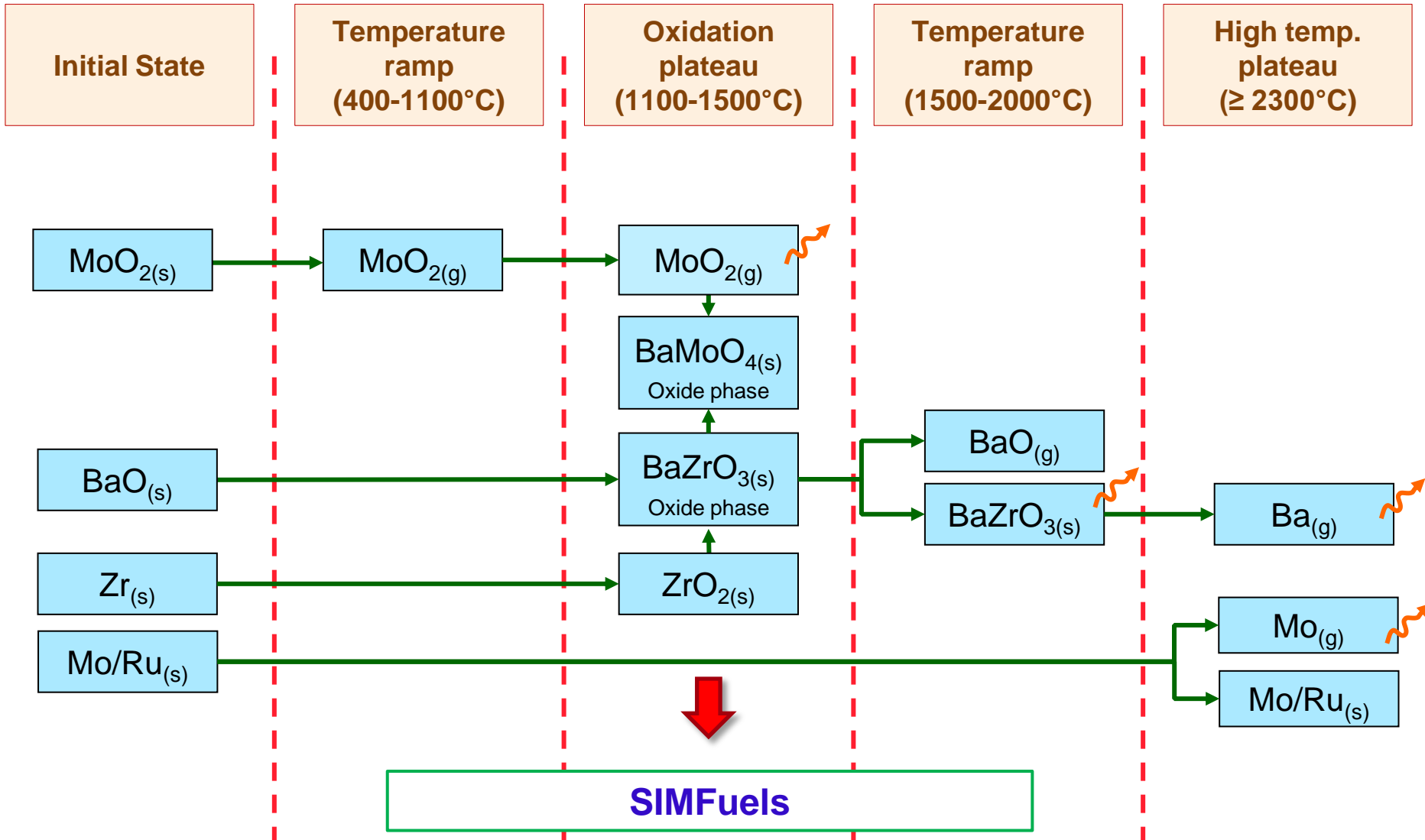
SEVERE ACCIDENT PHENOMENA

Source term



STUDY OF FPs BEHAVIOR

Mechanism suggested in the literature



SIMFuels SAMPLES

Fabrication data

Depleted UO_2 + 11 oxides
(FPs surrogates)



Sintering at 1650°C
during 2h under pure H_2

Elements	Ba	Ce	La	Mo	Sr	Y	Zr	Rh	Pd	Ru	Nd
Concentration (at%)	0,26	0,61	0,20	0,51	0,13	0,06	0,60	0,03	0,42	0,64	0,91

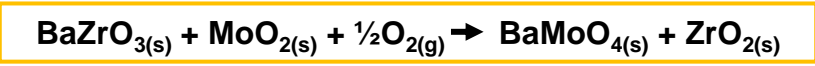
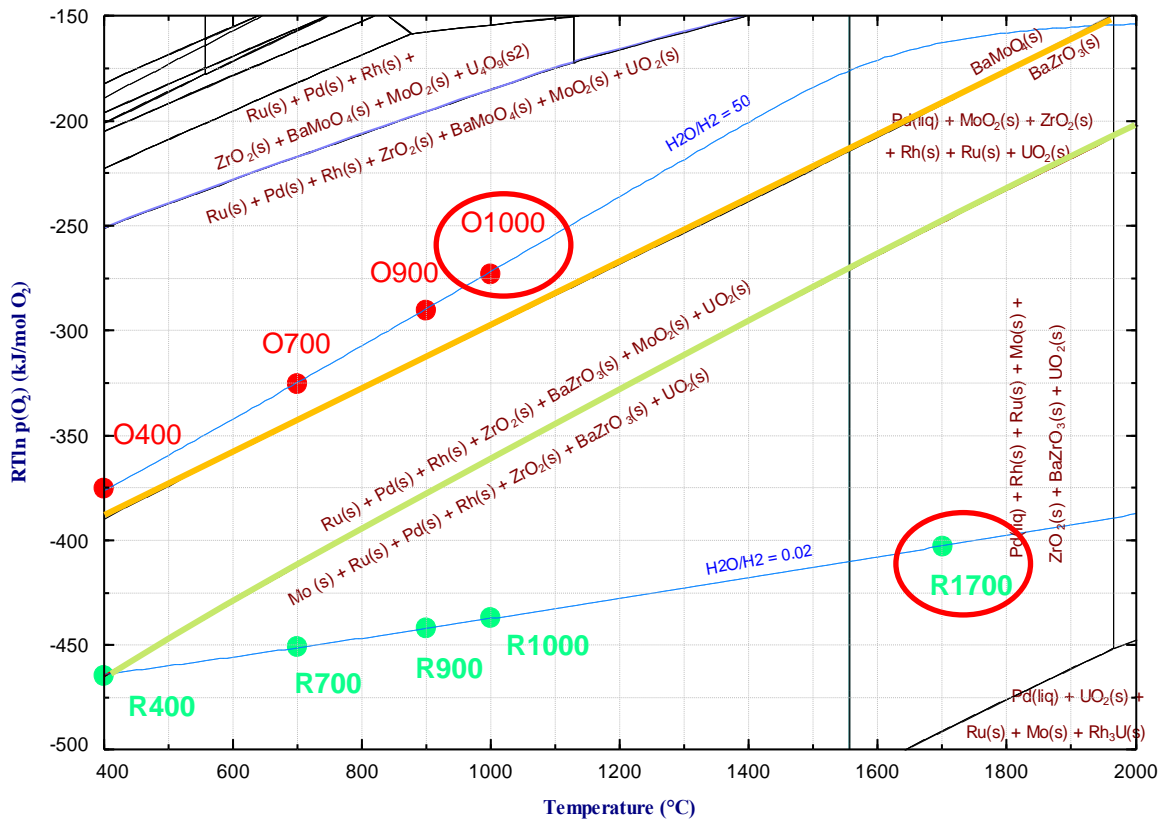


Concentrations representative of an irradiated fuel
with a burn-up of $76 \text{ GWd.t}_{\text{HM}}^{-1}$

SIMFuels SAMPLES

Thermal treatments

Ba - O₂ - Mo - Zr - Ru - Rh - Pd - U

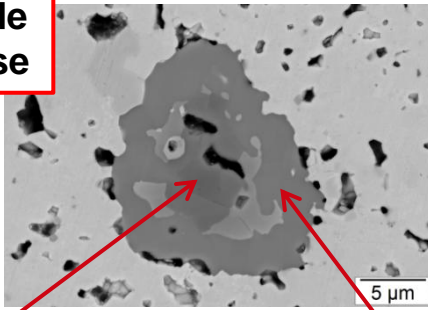




CHARACTERIZATIONS SEM-EDX

Sample as sintered

Oxide phase

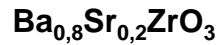


$$O/Zr \approx 2$$

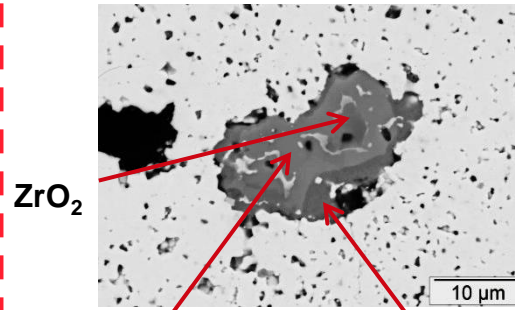


$$(Sr+Ba)/Zr \approx 1$$

$$O/Zr \approx 2.5$$



1000°C, -292 kJ.mol⁻¹

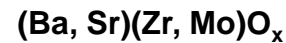


$$(Ba+Sr)/Mo \approx 1$$

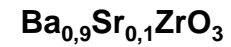
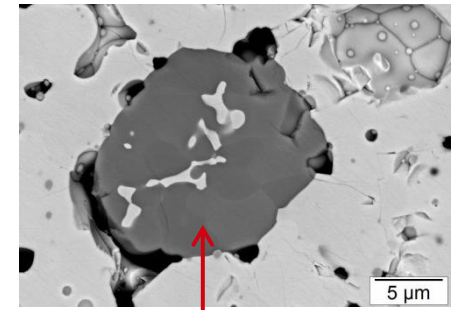
$$(Ba+Sr)/Zr \approx 0.6$$

$$O/Zr \approx 4$$

$$O/Mo \approx 5$$



1700°C, -426 kJ.mol⁻¹

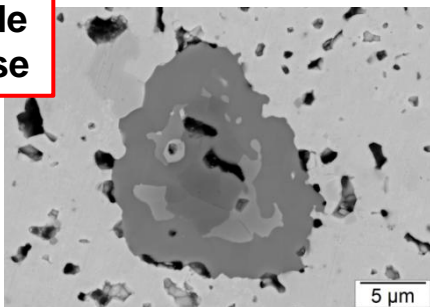




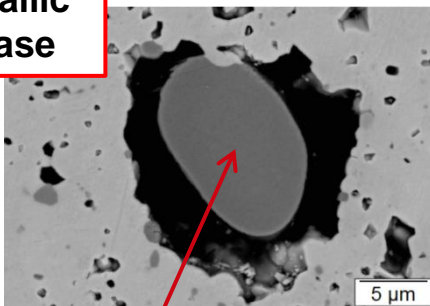
CHARACTERIZATIONS SEM-EDX

Sample as sintered

Oxide phase

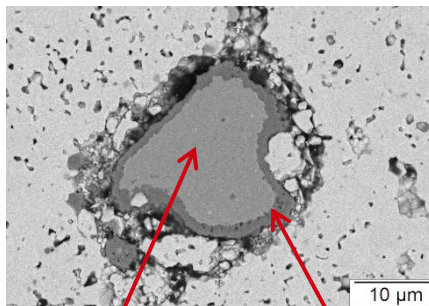
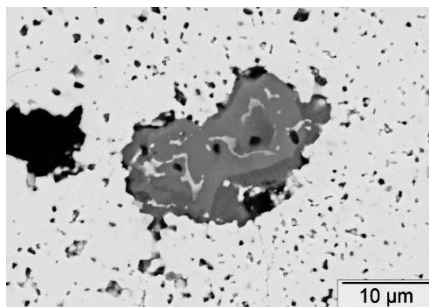


Metallic phase



100% Mo

1000°C, -292 kJ.mol⁻¹



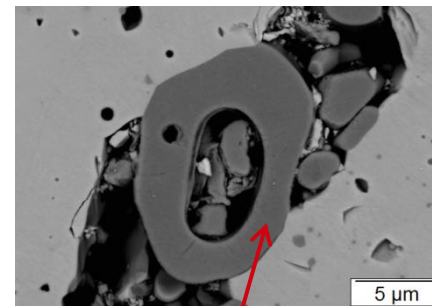
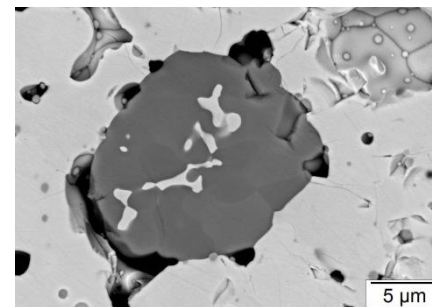
100% Mo

O/Mo ≈ 2



MoO₂

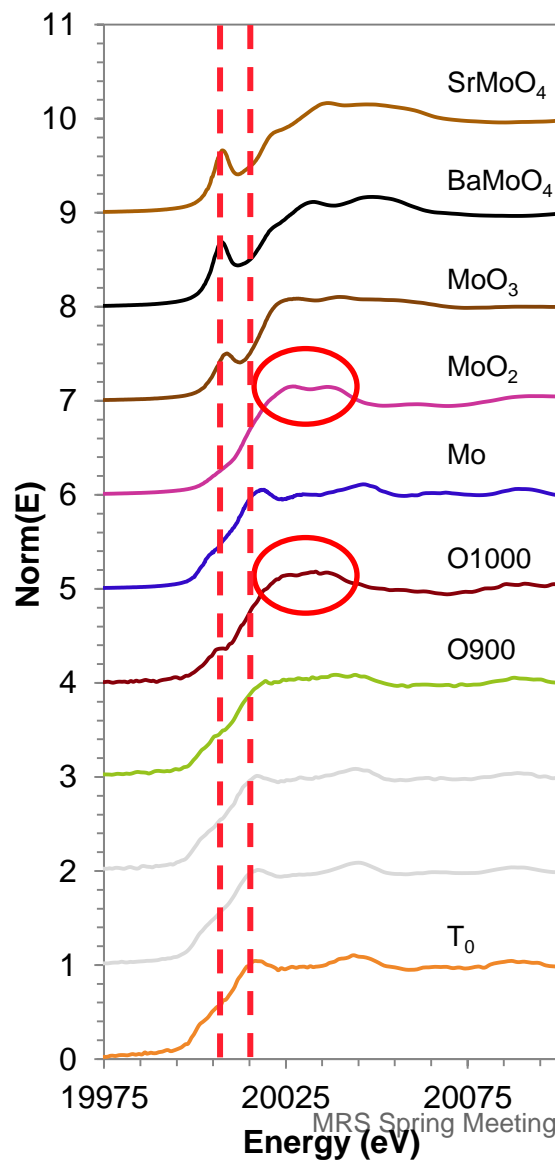
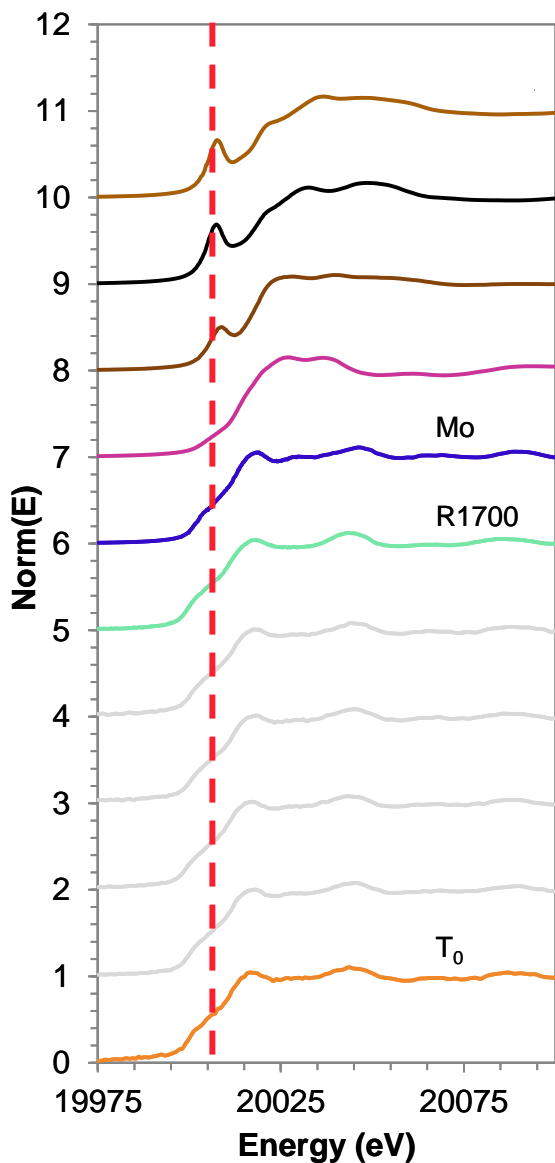
1700°C, -426 kJ.mol⁻¹



100% Mo

CHARACTERIZATIONS

X-ray Absorption Spectroscopy at Mo K-edge



No strong evolution of Mo XANES reducing conditions



Metallic Mo

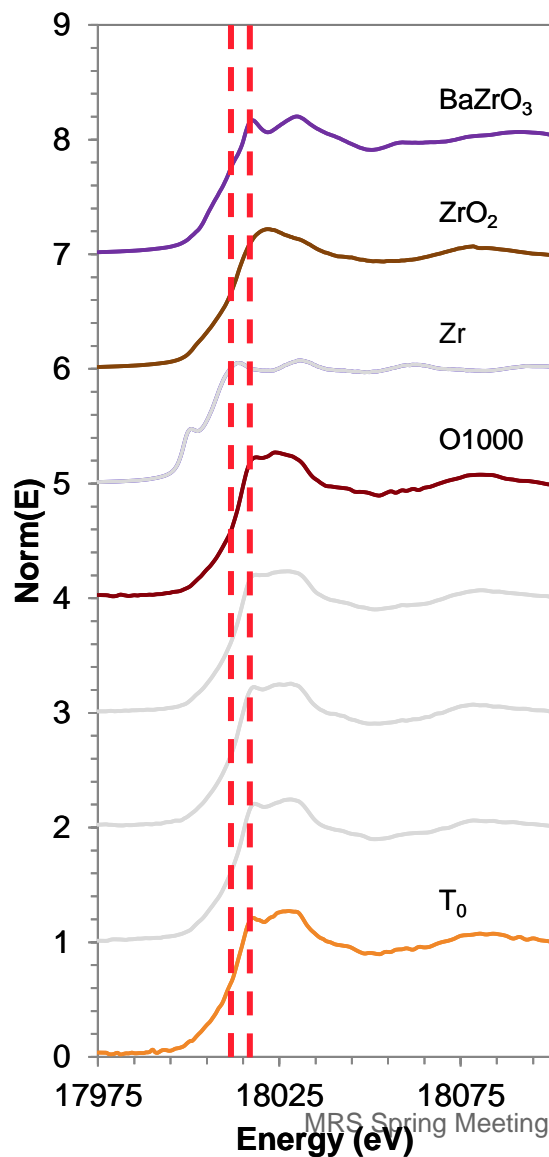
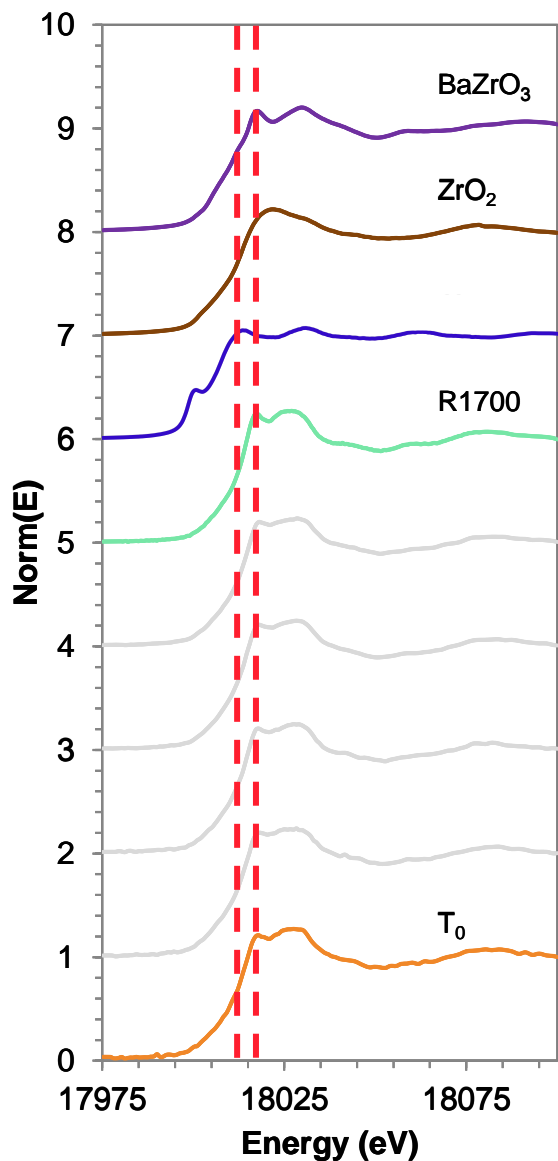
Evolution of Mo local environment from 900°C in oxidizing conditions



Consistent with Mo oxidation, MoO₂ formation and the oxide phase evolution

CHARACTERIZATIONS

X-ray Absorption Spectroscopy at Zr K-edge



Zr XANES evolution at 1700°C
in reducing conditions



Consistent with the
continuous BaZrO₃
formation and stabilization

Zr XANES evolution from
1000°C in oxidizing conditions



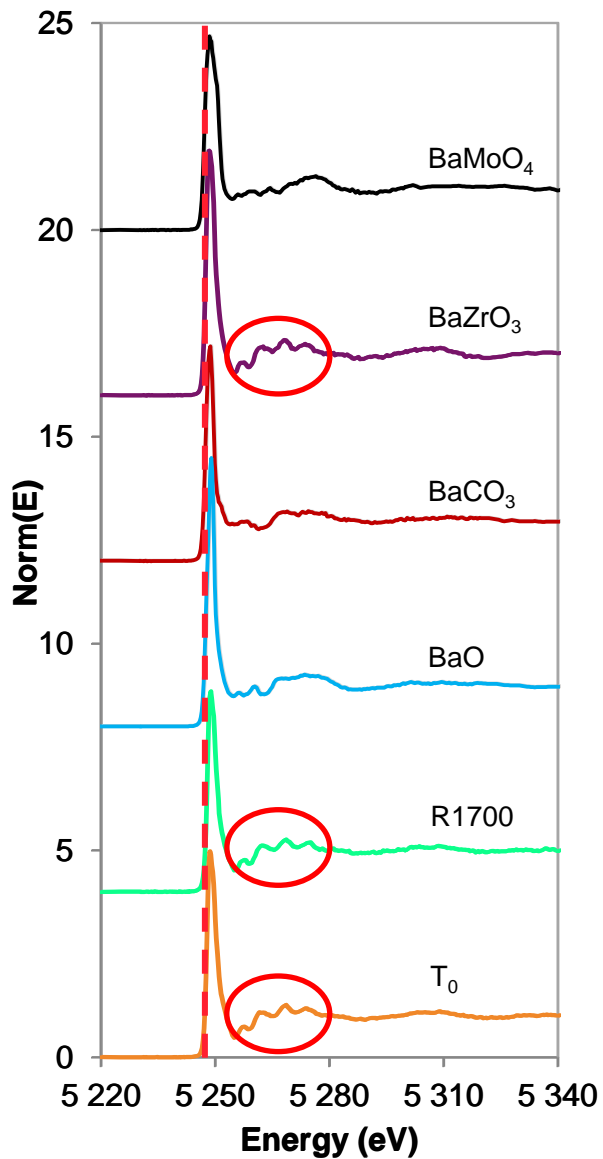
Destruction of part of the
BaZrO₃ phase and formation of
ZrO₂ ?



Consistent with the oxide phase
evolution (Mo reacts with Ba)

CHARACTERIZATIONS

HERFD-XANES at Ba L₃-edge



No Ba XANES evolution at
1700°C in reducing conditions

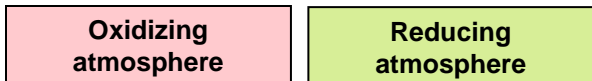
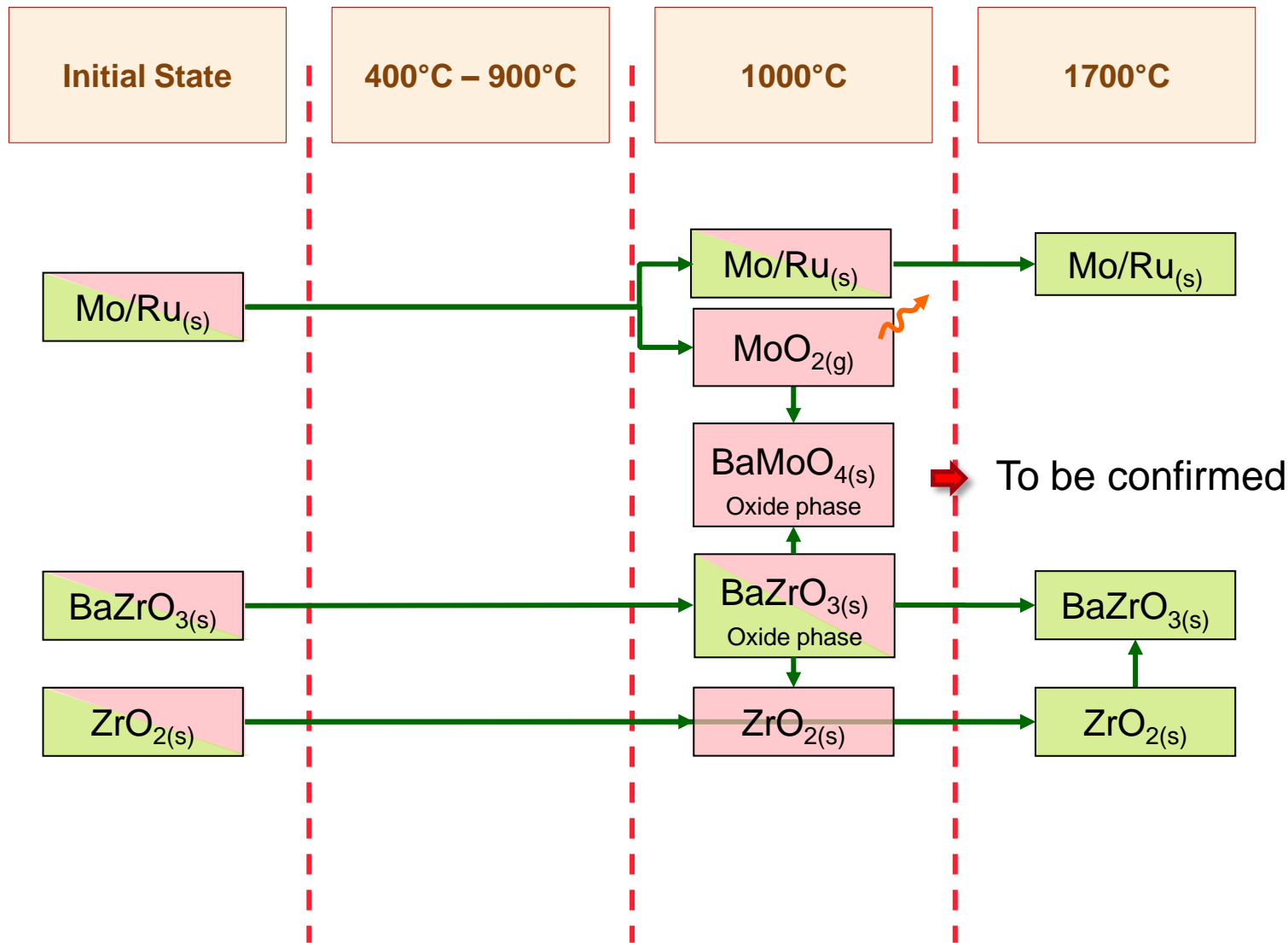


BaZrO₃

XANES analyses of the O-
samples scheduled in April
2018

CONCLUSIONS

Evolution of the oxide phase





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Principle

