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C. Le Gall, E. Geiger, O. Proux, M. Rovezzi, P.L. Solari, M. Hunault, V.  
Klosek, C. Riglet Martial, J. Léchelle, F. Audubert, et al.

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# Oxide phase characterization in simulated high burn-up $\text{UO}_2$ 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 Gall<sup>1</sup>, E. Geiger<sup>2</sup>, O. Proux<sup>3</sup>, M. Rovezzi<sup>3</sup>, P.L. Solari<sup>4</sup>, M. Hunault<sup>4</sup>, V. Klosek<sup>1</sup>, C. Riglet-Martial<sup>1</sup>, J. Léchelle<sup>1</sup>, F. Audubert<sup>1</sup>, Y. Pontillon<sup>1</sup>, J.-L. Hazemann<sup>5</sup>

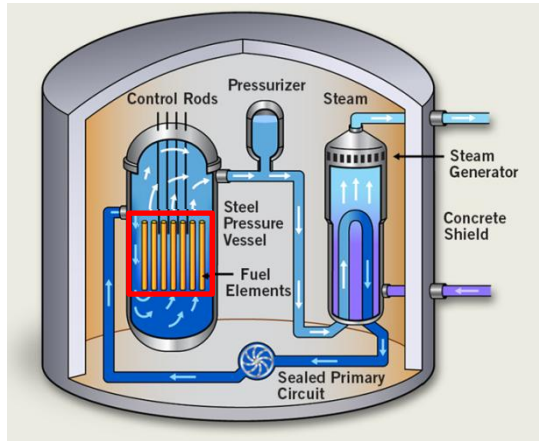
<sup>1</sup>CEA / DEN / DEC, <sup>2</sup>DCCE / RMC Canada, <sup>3</sup>OSUG-UGA, <sup>4</sup>MARS / SOLEIL Synchrotron, <sup>5</sup>Institut Néel / CNRS UGA

April 2-6, 2018 – Phoenix, Arizona

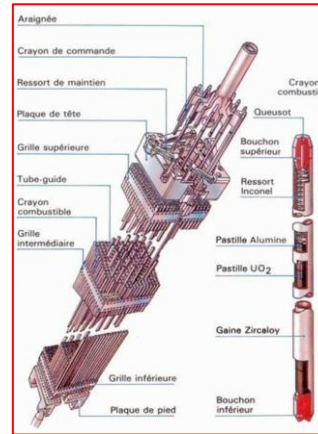


# NUCLEAR FUEL FISSION

## Fission products



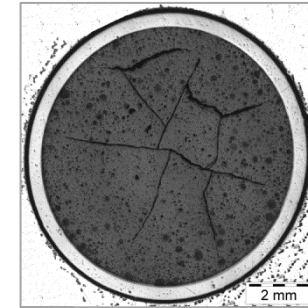
Pressurized Water Reactor (PWR)



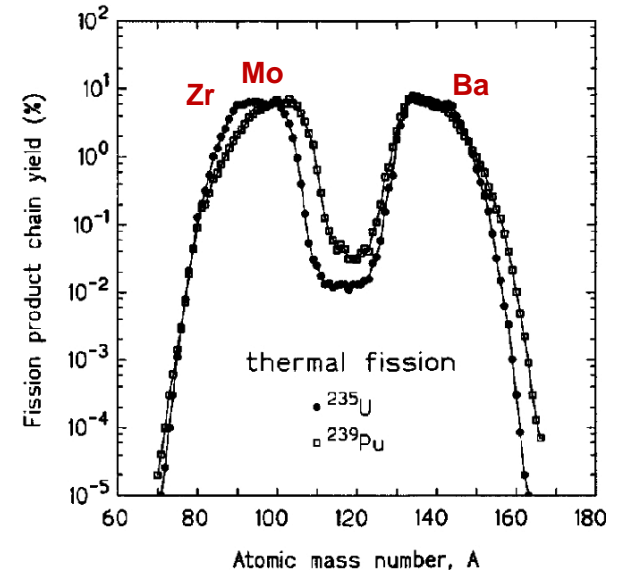
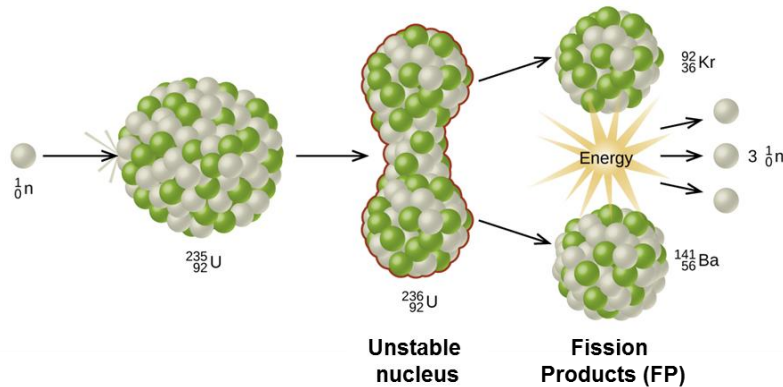
Fuel assembly



UO<sub>2</sub> or (U,Pu)O<sub>2</sub> fuel in its Zr alloyed cladding



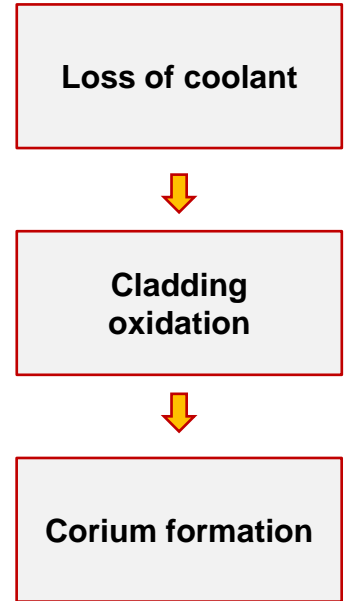
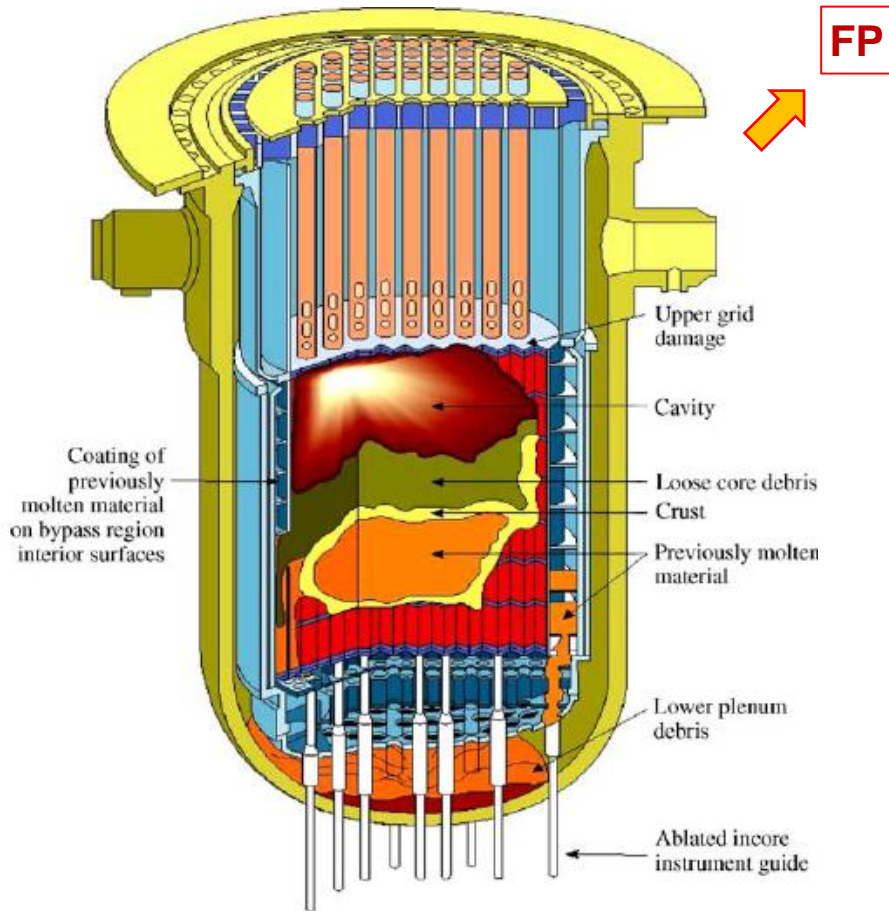
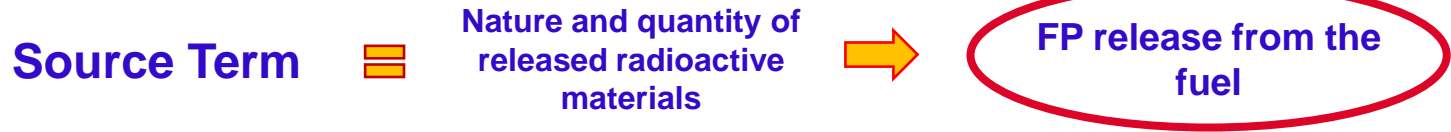
Fission yield: FP most probably produced



**Fission occurs in nuclear fuel pellets**

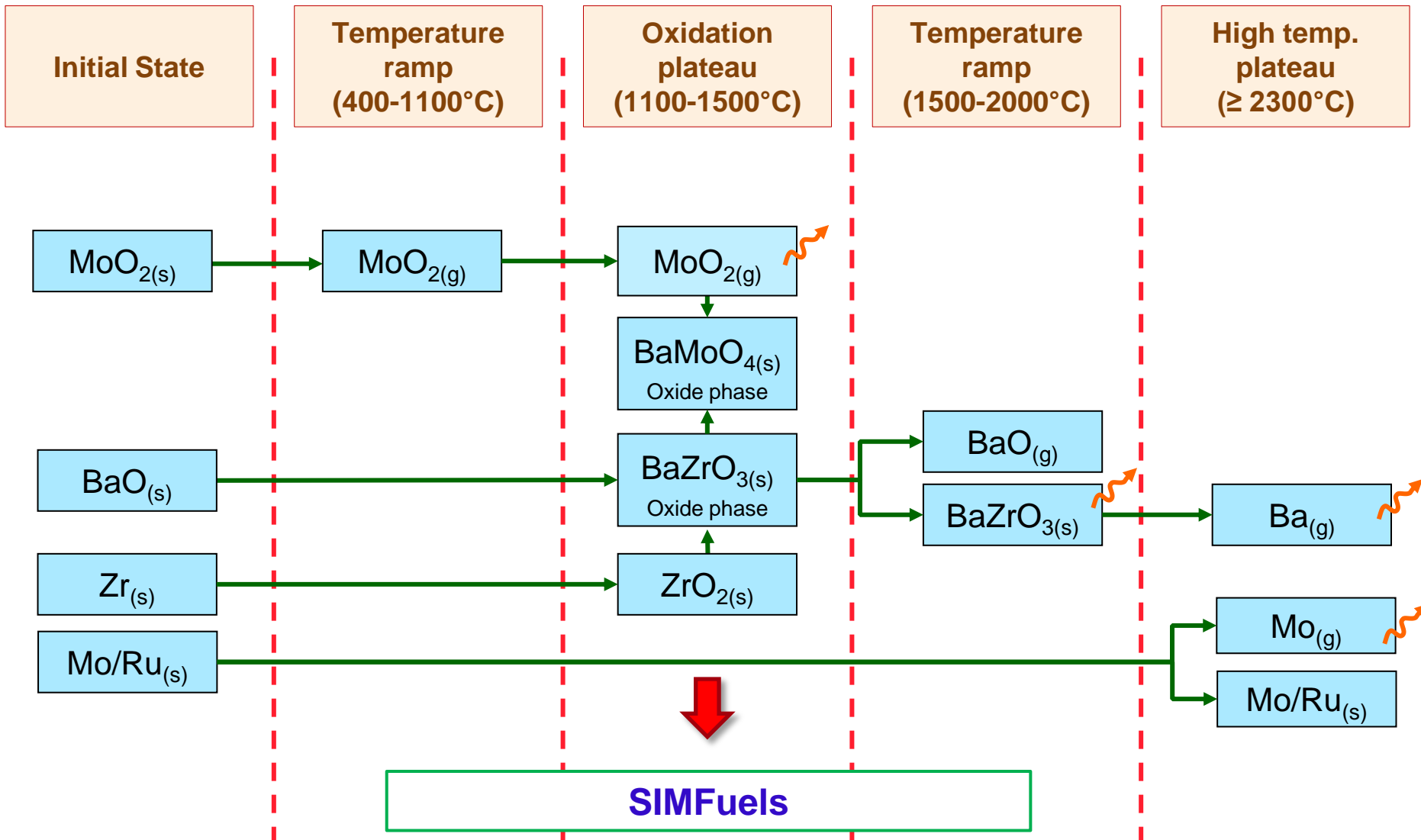
# SEVERE ACCIDENT PHENOMENA

Source term



# STUDY OF FPs BEHAVIOR

## Mechanism suggested in the literature



# SIMFuels SAMPLES

Fabrication data

Depleted  $\text{UO}_2$  + 11 oxides  
(FPs surrogates)



Sintering at  $1650^\circ\text{C}$   
during 2h under pure  $\text{H}_2$

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

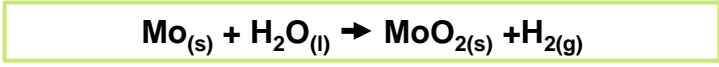
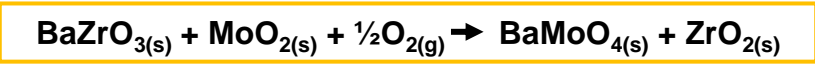
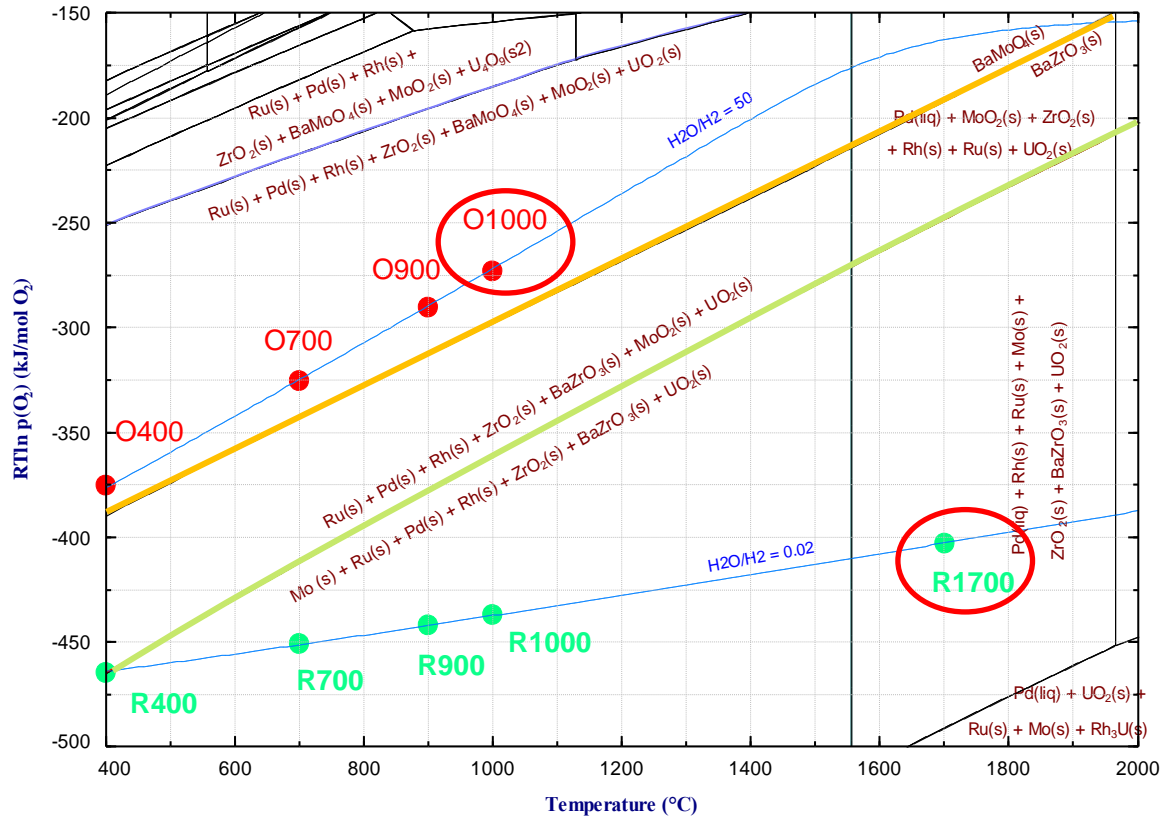


Concentrations representative of an irradiated fuel  
with a burn-up of  $76 \text{ GWd.t}_{\text{HM}}^{-1}$

# SIMFuels SAMPLES

## Thermal treatments

Ba - O<sub>2</sub> - Mo - Zr - Ru - Rh - Pd - U

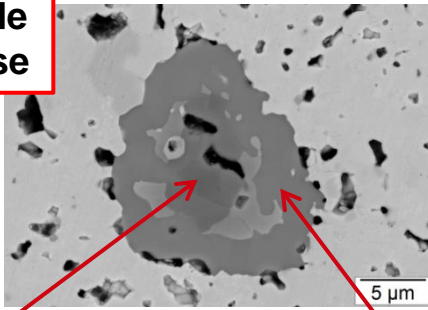




# CHARACTERIZATIONS SEM-EDX

Sample as sintered

Oxide phase



$O/Zr \approx 2$



$ZrO_2$

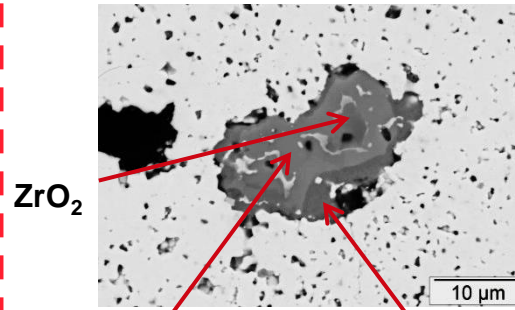
$(Sr+Ba)/Zr \approx 1$

$O/Zr \approx 2.5$



$Ba_{0,8}Sr_{0,2}ZrO_3$

1000°C, -292 kJ.mol<sup>-1</sup>



$ZrO_2$

$Ba_{0,8}Sr_{0,2}ZrO_3$

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

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

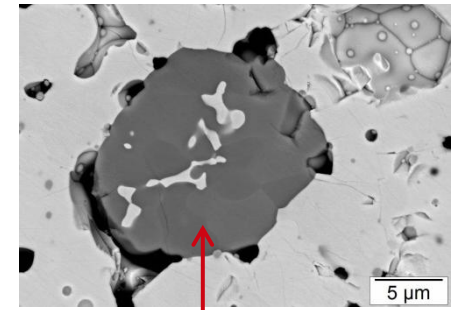
$O/Zr \approx 4$

$O/Mo \approx 5$



$(Ba, Sr)(Zr, Mo)O_x$

1700°C, -426 kJ.mol<sup>-1</sup>



$Ba_{0,9}Sr_{0,1}ZrO_3$

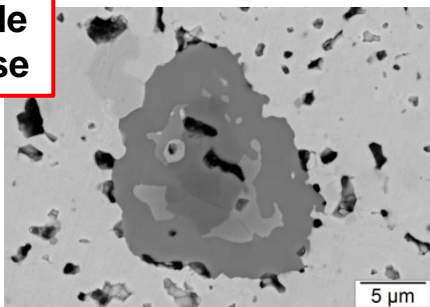




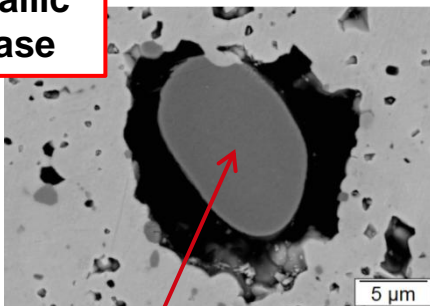
# CHARACTERIZATIONS SEM-EDX

Sample as sintered

Oxide phase

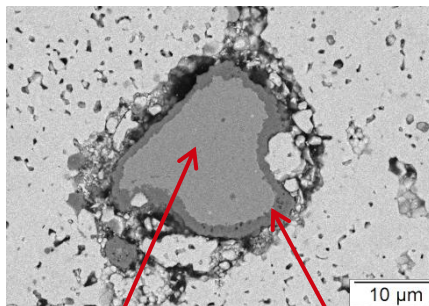
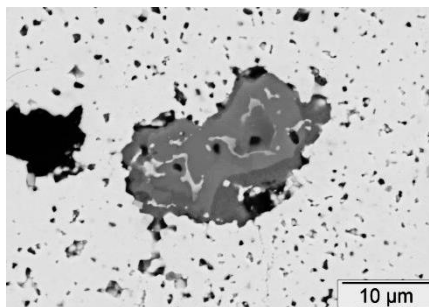


Metallic phase



100% Mo

1000°C, -292 kJ.mol<sup>-1</sup>



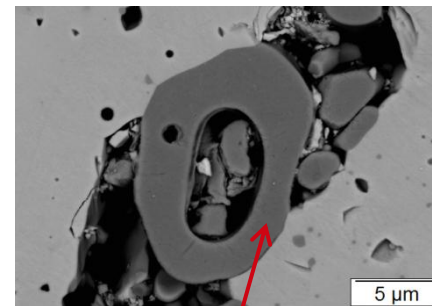
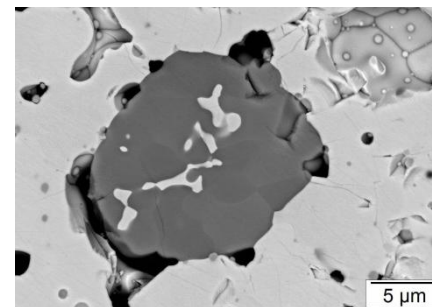
100% Mo

O/Mo ≈ 2



MoO<sub>2</sub>

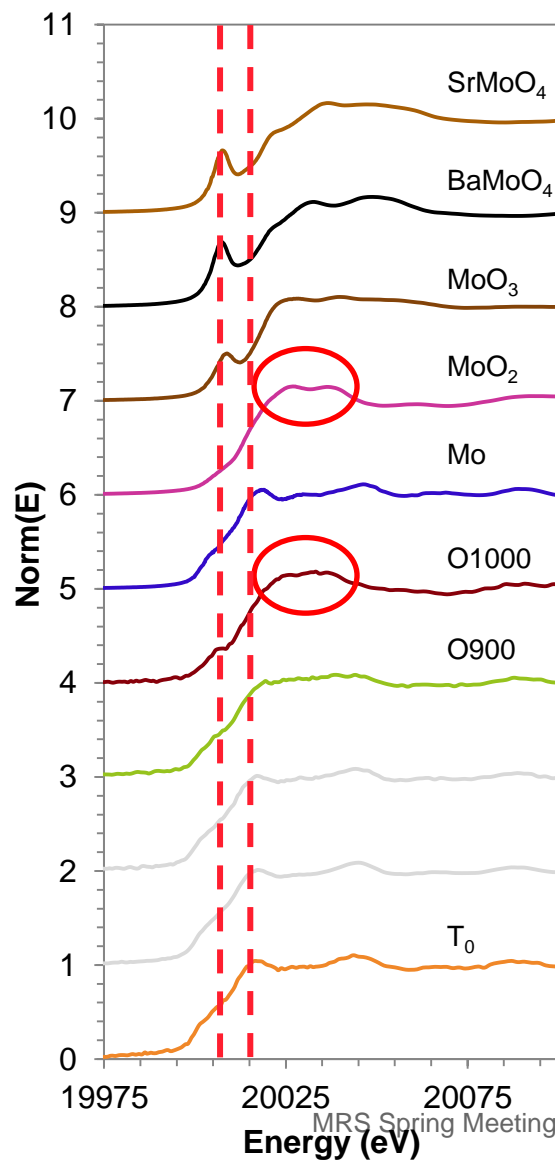
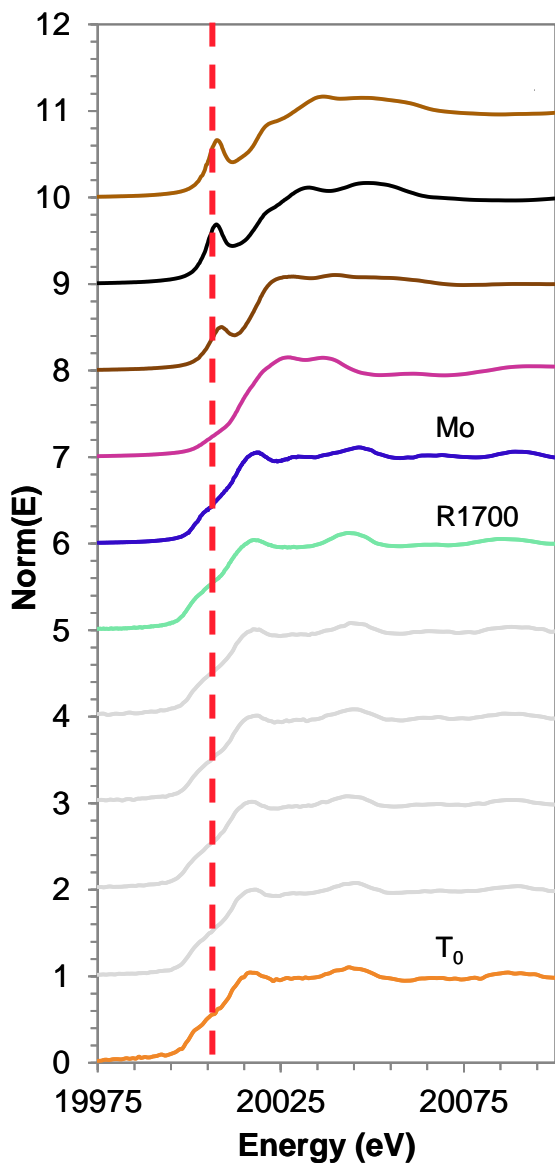
1700°C, -426 kJ.mol<sup>-1</sup>



100% Mo

# CHARACTERIZATIONS

## X-ray Absorption Spectroscopy at Mo K-edge



No strong evolution of Mo XANES reducing conditions



Metallic Mo

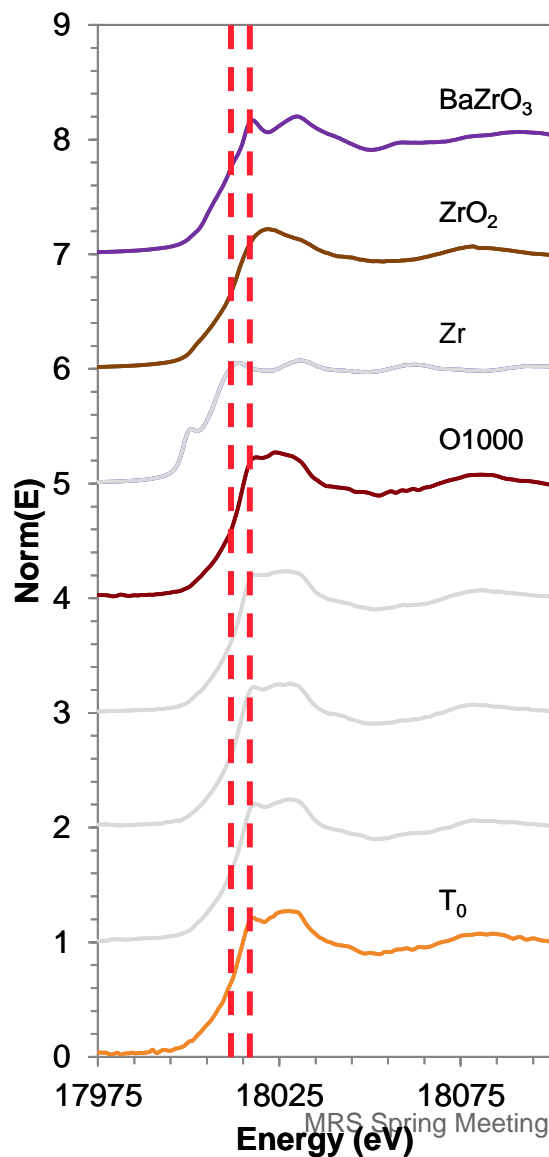
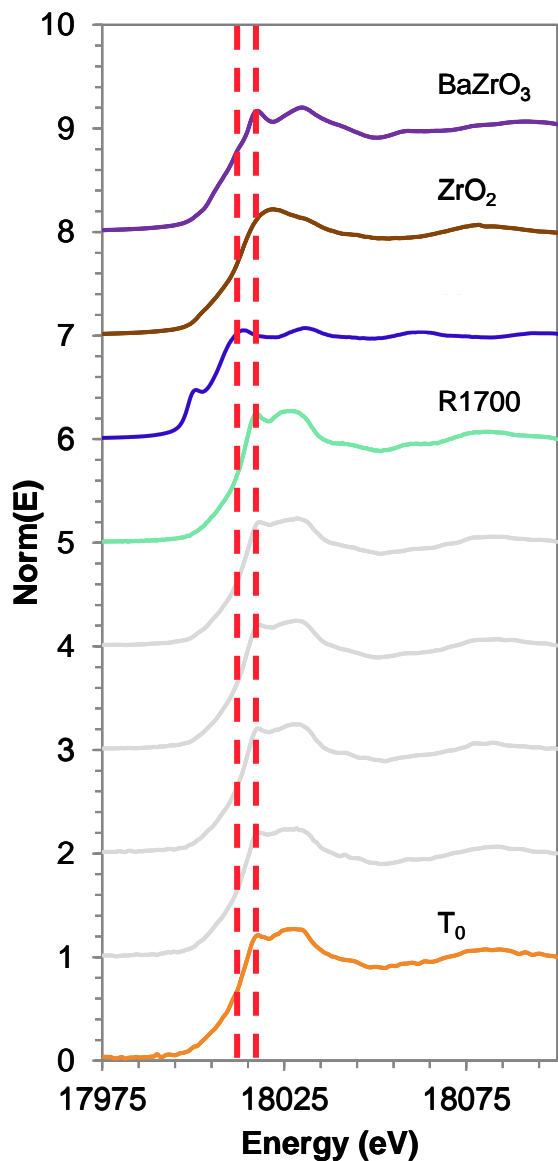
Evolution of Mo local environment from 900°C in oxidizing conditions



Consistent with Mo oxidation, MoO<sub>2</sub> 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 BaZrO<sub>3</sub>  
formation and stabilization

Zr XANES evolution from  
1000°C in oxidizing conditions



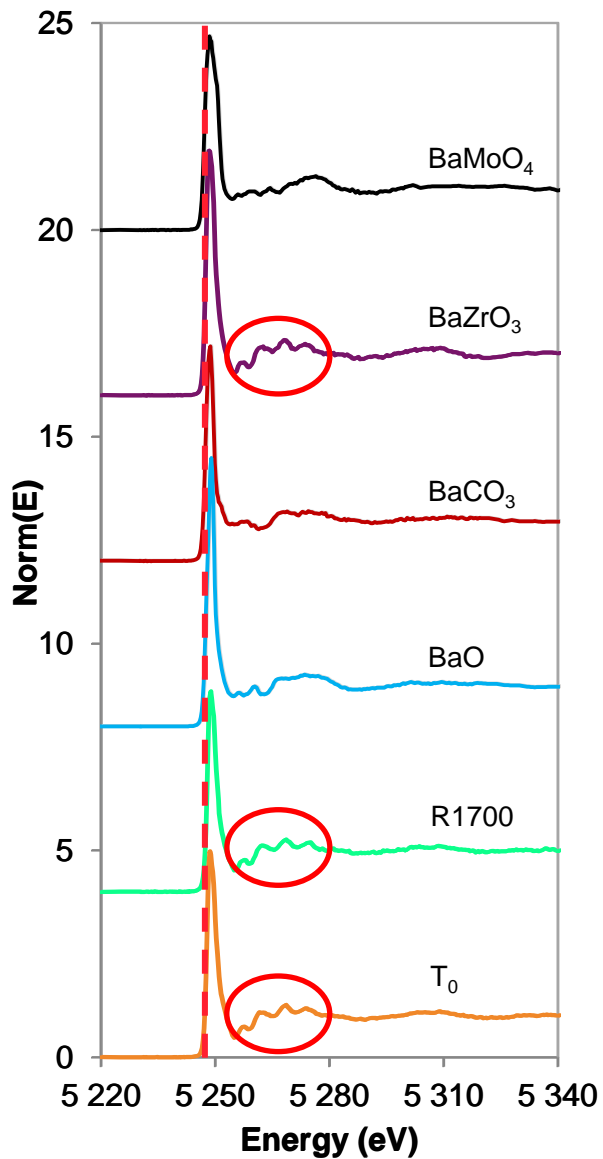
Destruction of part of the  
BaZrO<sub>3</sub> phase and formation of  
ZrO<sub>2</sub> ?



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

# CHARACTERIZATIONS

## HERFD-XANES at Ba L<sub>3</sub>-edge



No Ba XANES evolution at  
1700°C in reducing conditions

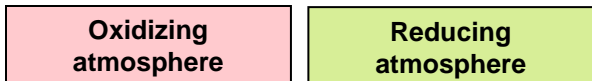
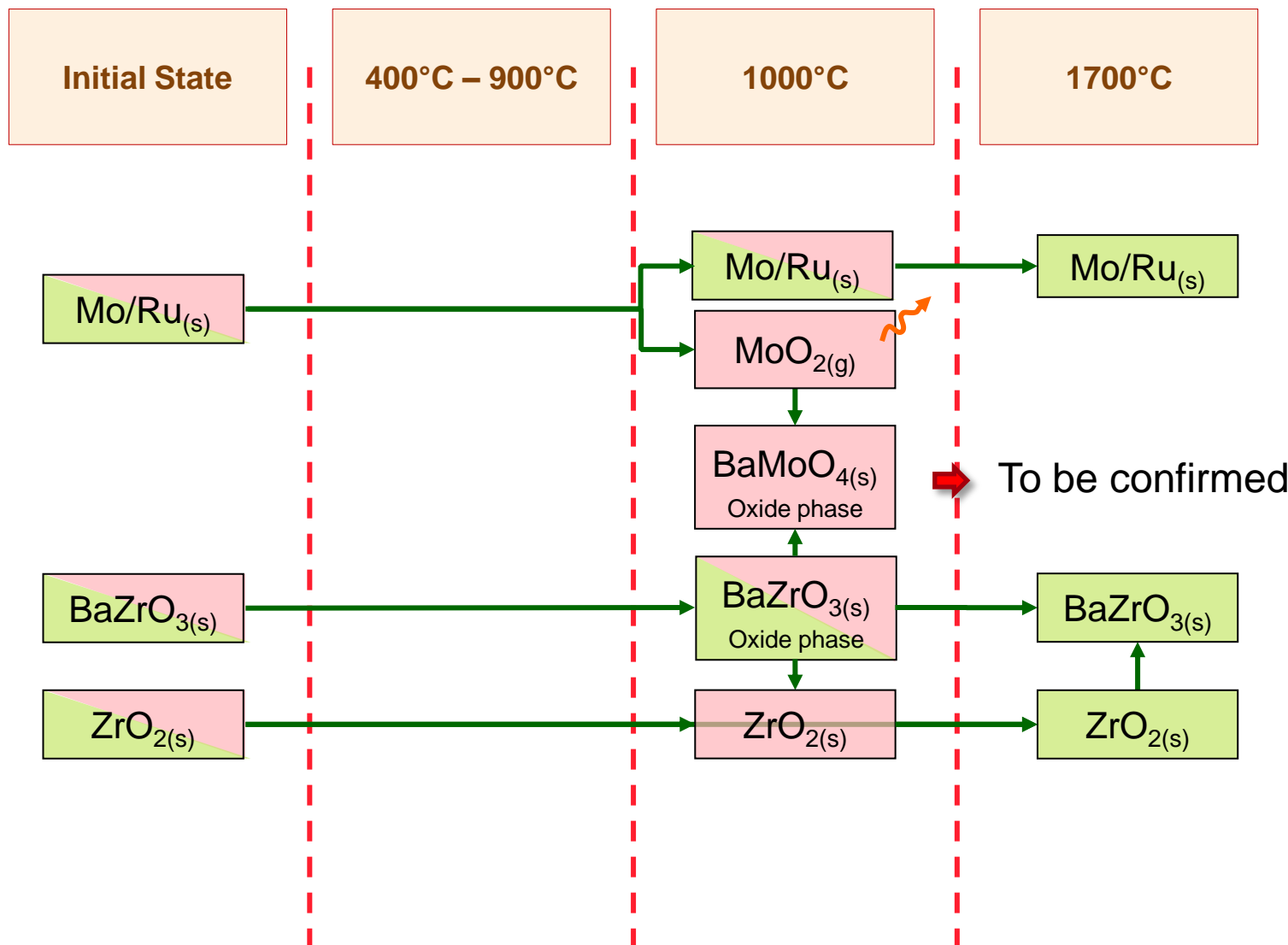


BaZrO<sub>3</sub>

XANES analyses of the O-  
samples scheduled in April  
2018

# CONCLUSIONS

## Evolution of the oxide phase





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## Principle

