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

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To cite this version:

C. Le Gall, E. Geiger, O. Proux, M. Rovezzi, P.L. Solari, et al.. Oxide phase characterization in simulated high burn-up UO_2 fuels in the early stages of a nuclear severe accident. The 2018 MRS Spring Meeting & Exhibit, Apr 2018, Phoenix, United States. cea-02338951

HAL Id: cea-02338951

https://cea.hal.science/cea-02338951

Submitted on 21 Feb 2020

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DE LA RECHERCHE À L'INDUSTRIE

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

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April 2-6, 2018 – Phoenix, Arizona

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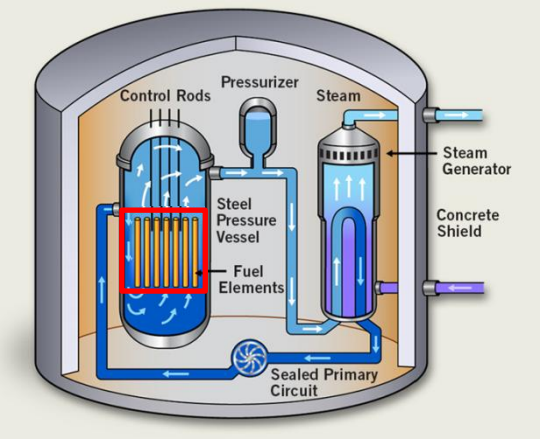
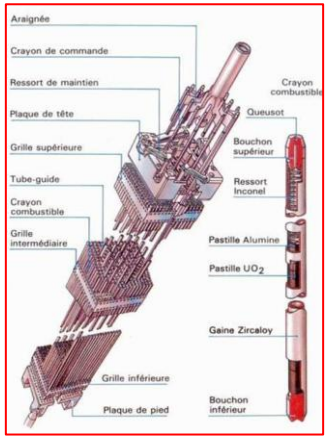
NUCLEAR FUEL FISSION

Fission products

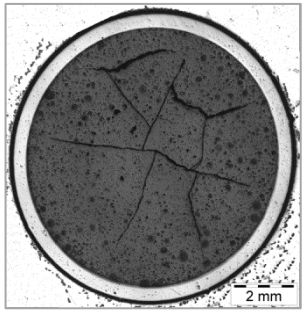


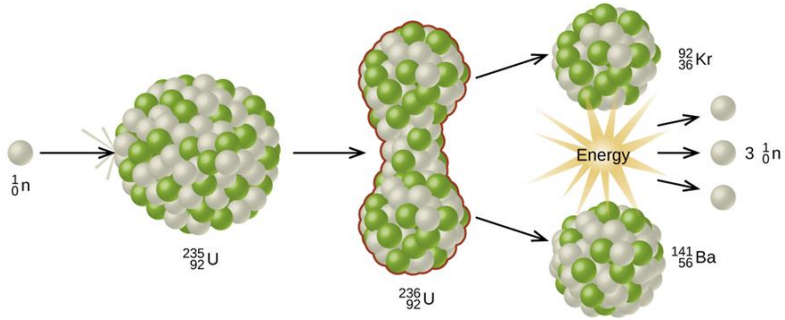
Pressurized Water Reactor (PWR)



Fuel assembly

UO_2 or $(U,Pu)O_2$ fuel in its Zr alloyed cladding

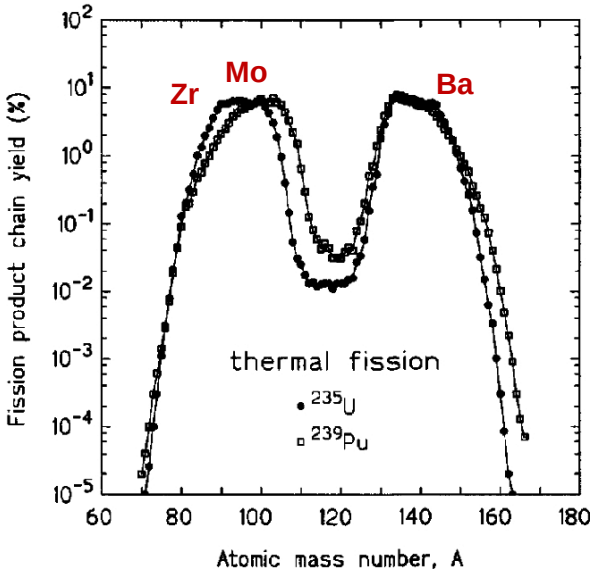




Unstable nucleus

Fission Products (FP)

Fission yield: FP most probably producted



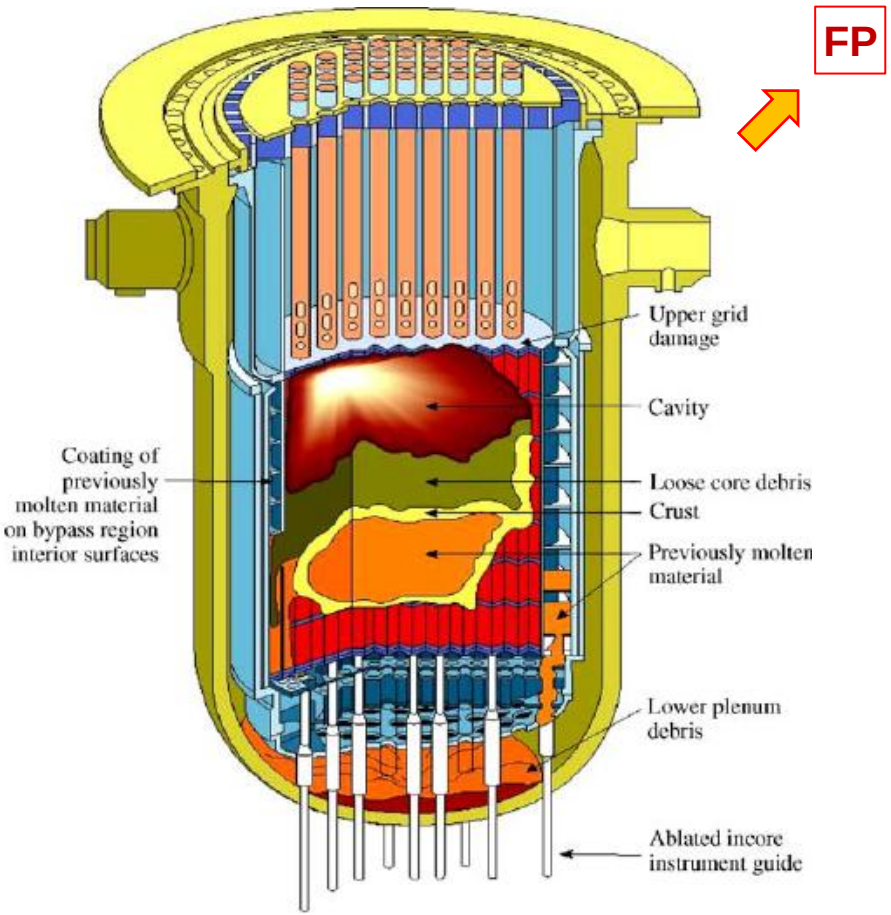
Fission occurs in nuclear fuel pellets

D. Heinze *Ph.D thesis* **(2010)**

SEVERE ACCIDENT PHENOMENA

Source term

Source Term = **Nature and quantity of released radioactive materials** → **FP release from the fuel**



Loss of coolant

↓

Cladding oxidation

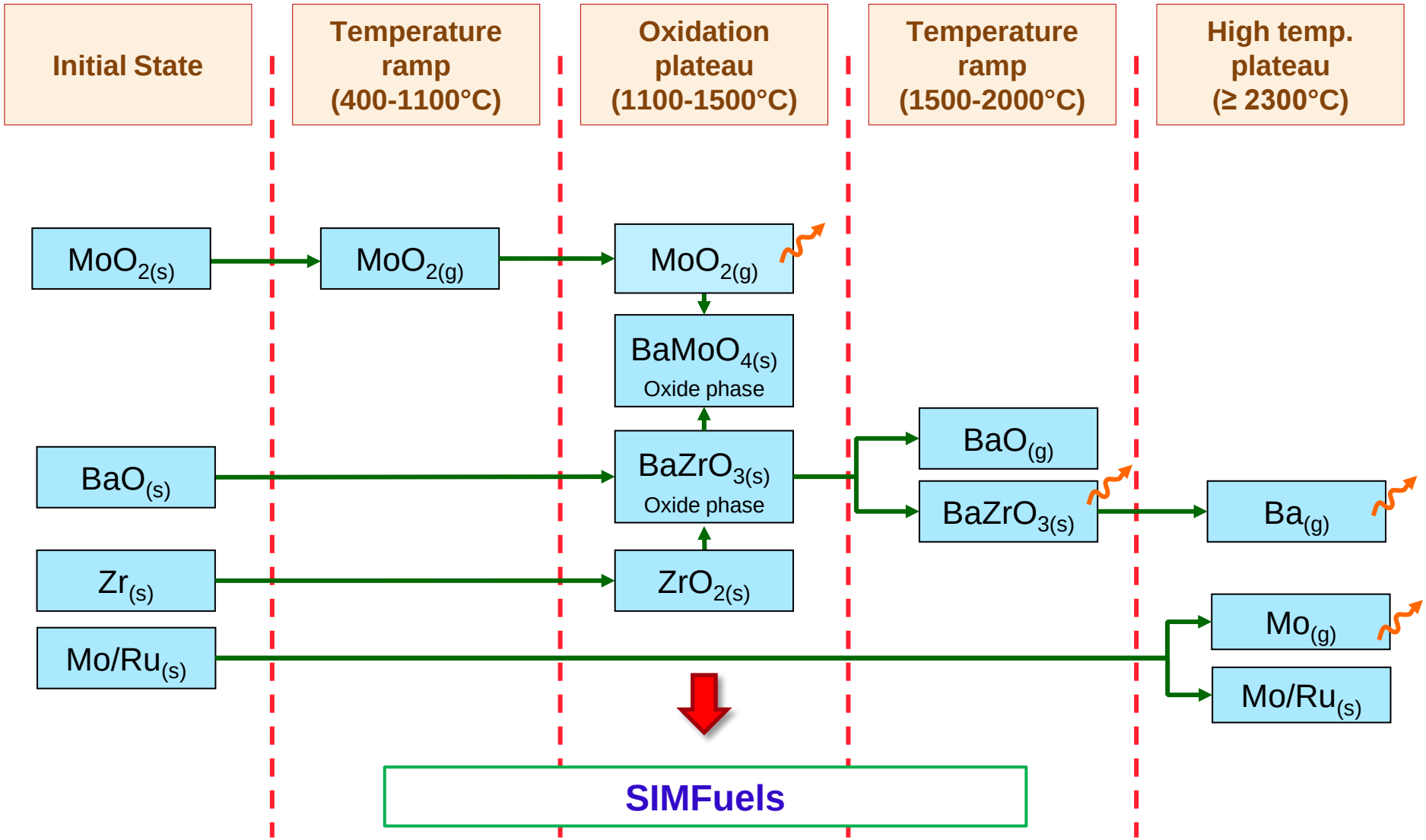
↓

Corium formation

J. P. Van Dorsselaere et al., *NED 236* **(2006)**
B. R. Sehgal, *Nuclear safety in LWR* **(2012)**

STUDY OF FPs BEHAVIOR

Mechanism suggested in the literature



G. Nicaise, *8th International Conference on Candu Fuel* **(2003)**
G. Nicaise, *NT DPAM/SEMIC 2004/37* **(2004)**
E. Geiger, *PhD thesis* **(2016)**
H. Kleykamp, *J. Nucl. Mater.* **(1985)**

SIMFuels SAMPLES

Fabrication data

Depleted UO_2 + 11 oxides
(FPs surrogates)

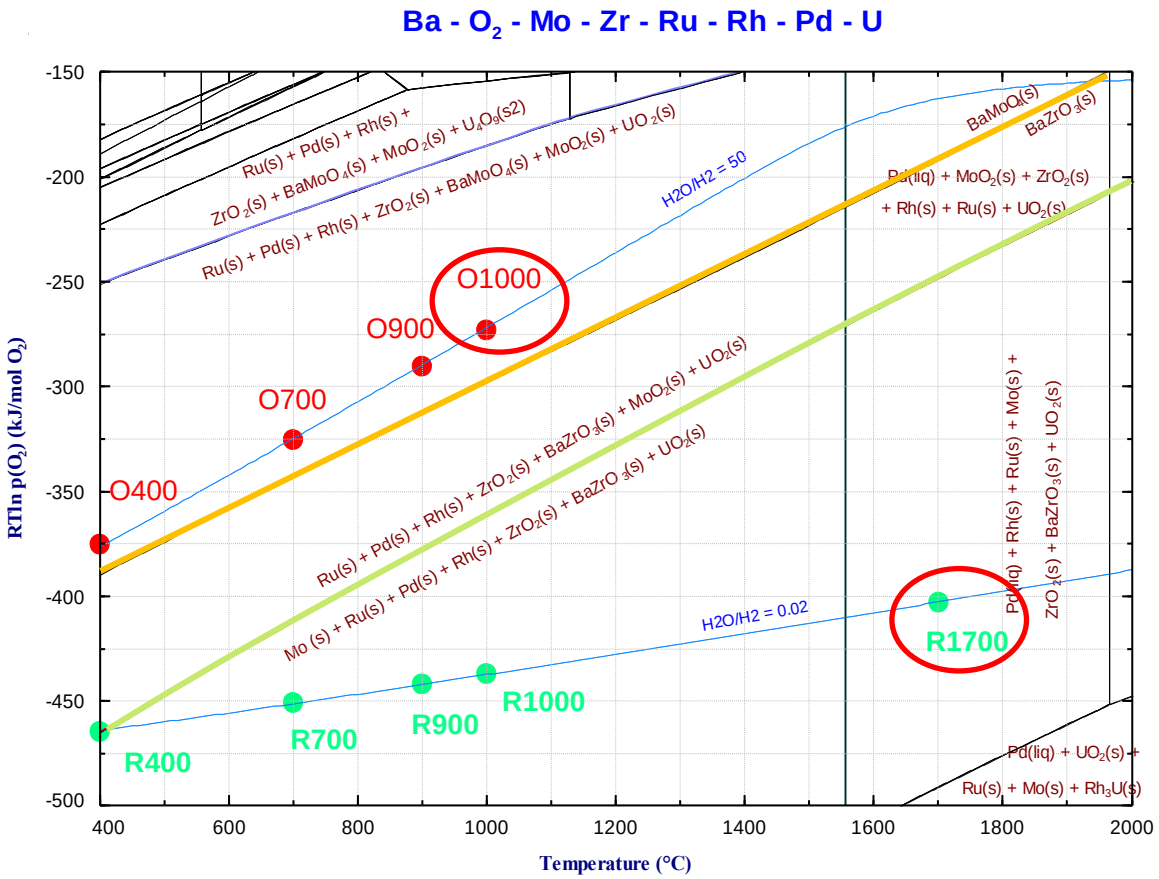
Sintering at 1650°C
during 2h under pure H_2

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 $GWd.t_{HM}^{-1}$

SIMFuels SAMPLES

Thermal treatments



$$BaZrO_{3(s)} + MoO_{2(s)} + \tfrac{1}{2}O_{2(g)} \rightarrow BaMoO_{4(s)} + ZrO_{2(s)}$$

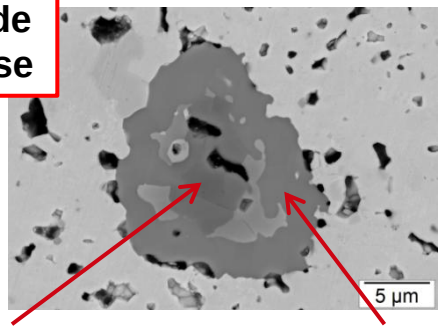
$$Mo_{(s)} + H_2O_{(l)} \rightarrow MoO_{2(s)} + H_{2(g)}$$

CHARACTERIZATIONS

SEM-EDX

Sample as sintered

Oxide phase



O/Zr ≈ 2

ZrO_2

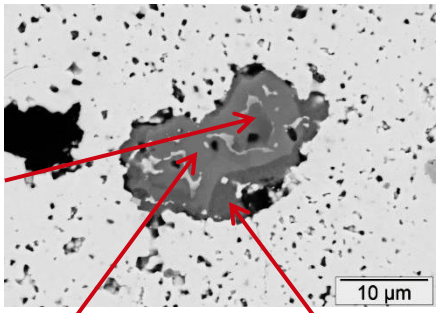
(Sr+Ba)/Zr ≈ 1

O/Zr ≈ 2.5

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

1000°C, -292 kJ.mol^{-1}

ZrO_2



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

(Ba+Sr)/Mo ≈ 1

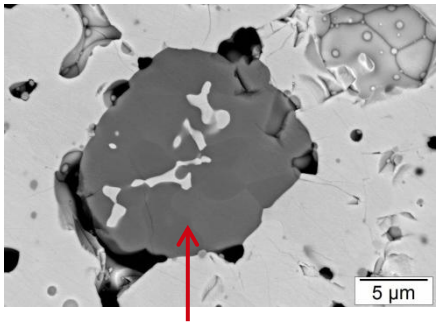
(Ba+Sr)/Zr ≈ 0.6

O/Zr ≈ 4

O/Mo ≈ 5

(Ba, Sr)(Zr, Mo)O_x

1700°C, -426 kJ.mol^{-1}



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

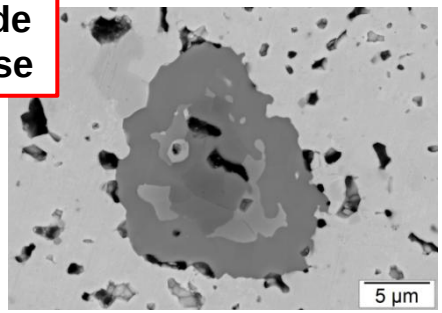
C. Thouzellier internship **(2017)**, H. Kleykamp, *J. Nucl. Mater.* **(1985);** E. Geiger et al. *ERMSAR proceedings* **(2015)**; E Geiger et al. *J. Nucl. Mater. (accepted)*

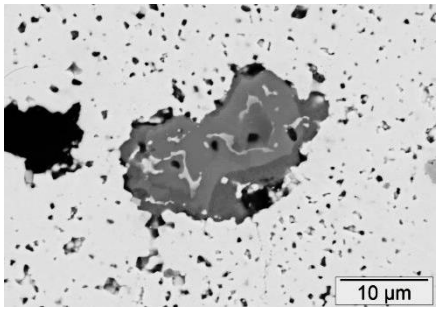
CHARACTERIZATIONS

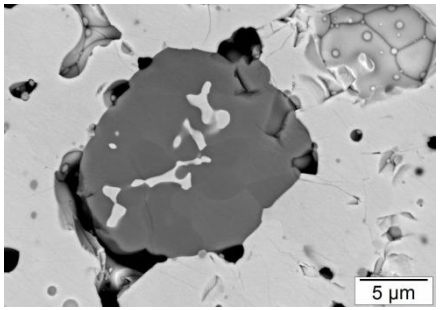
SEM-EDX

Sample as sintered	1000°C, -292 kJ.mol^{-1}	1700°C, -426 kJ.mol^{-1}

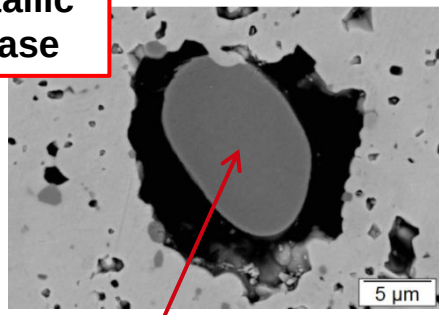
Oxide phase



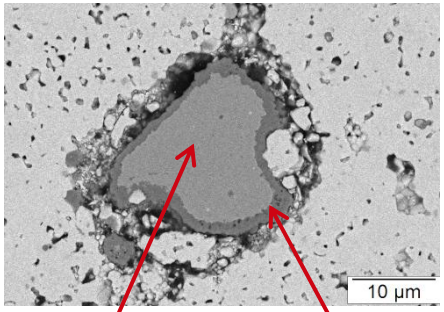




Metallic phase



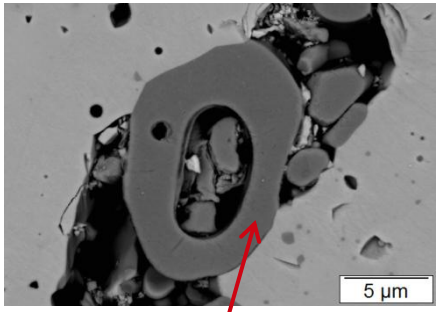
100% Mo



100% Mo

O/Mo ≈ 2

MoO_2

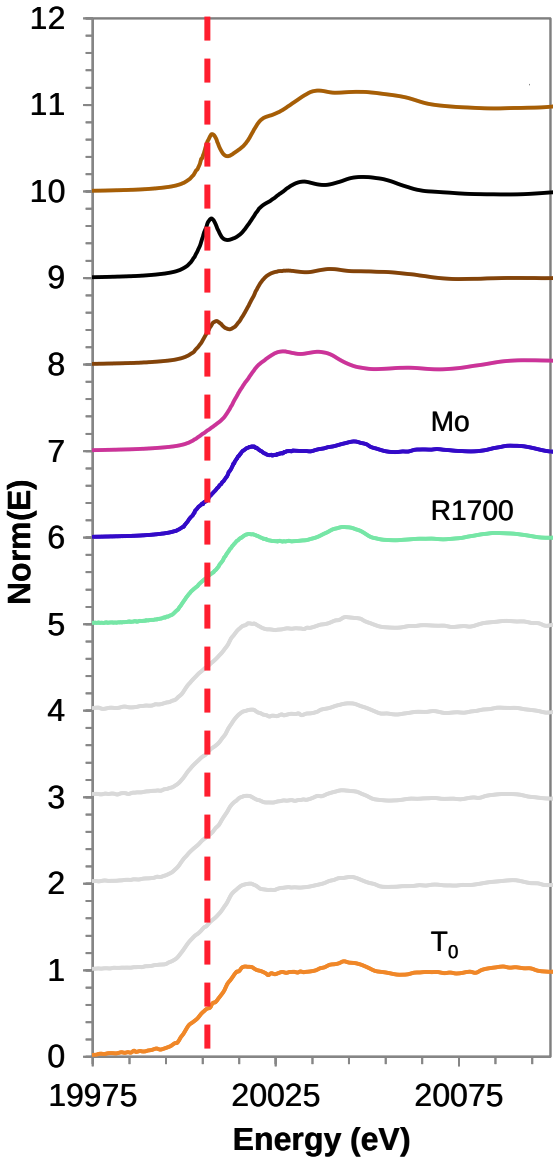


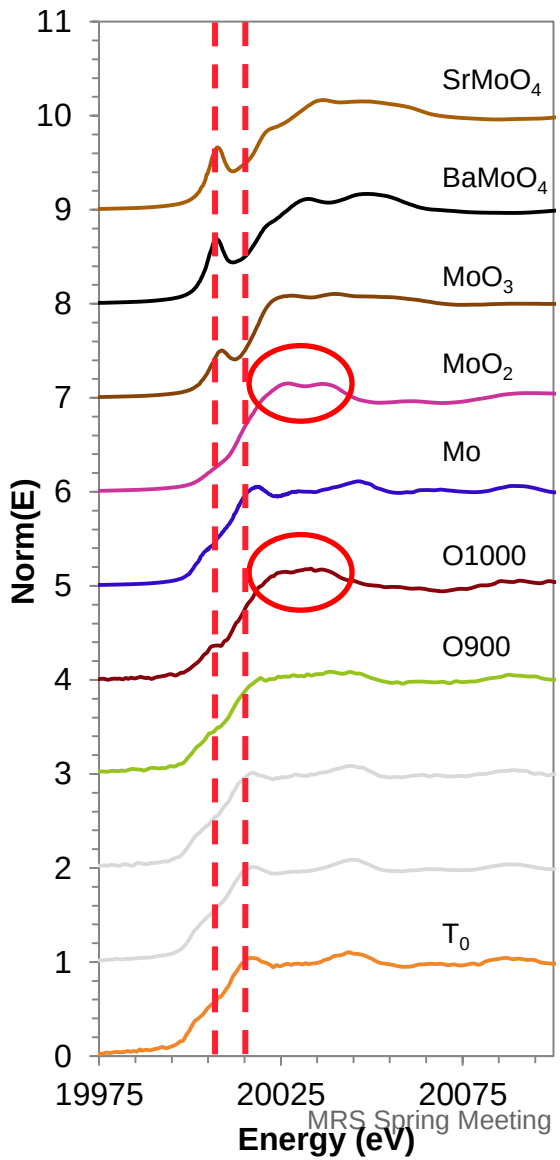
100% Mo

C. Thouzellier internship **(2017)**, H. Kleykamp, *J. Nucl. Mater.* **(1985);** E. Geiger et al. *ERMSAR proceedings* **(2015)**; E Geiger et al. *J. Nucl. Mater. (accepted)*

CHARACTERIZATIONS

X-ray Absorption Spectroscopy at Mo K-edge





No strong evolution of Mo XANES reducing conditions

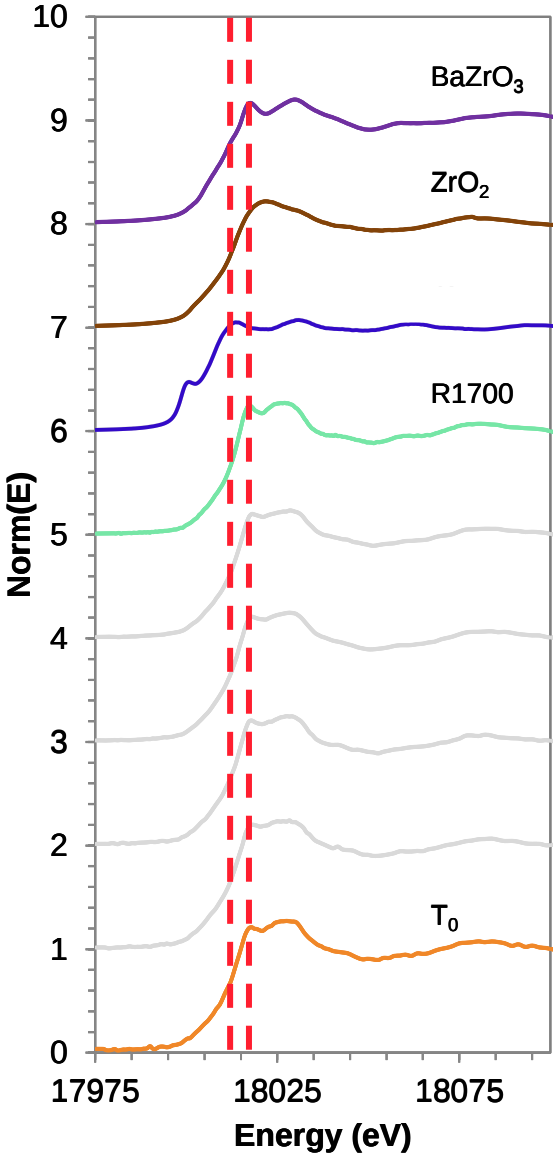
Metallic Mo

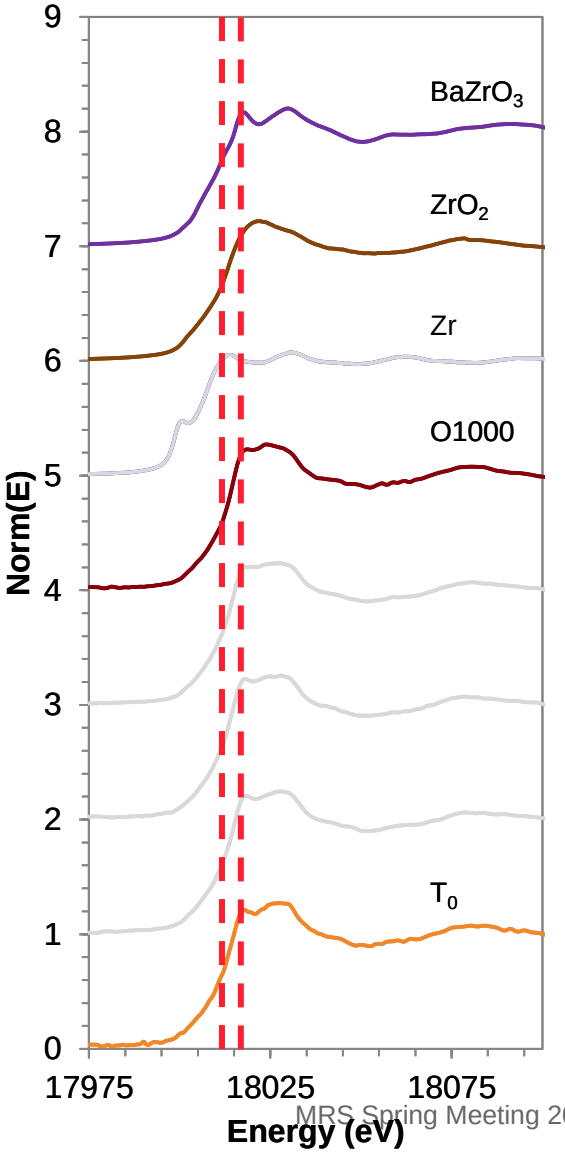
Evolution of Mo local environment from 900°C in oxidizing conditions

Consistent with Mo oxidation, MoO_2 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 $BaZrO_3$ formation and stabilization

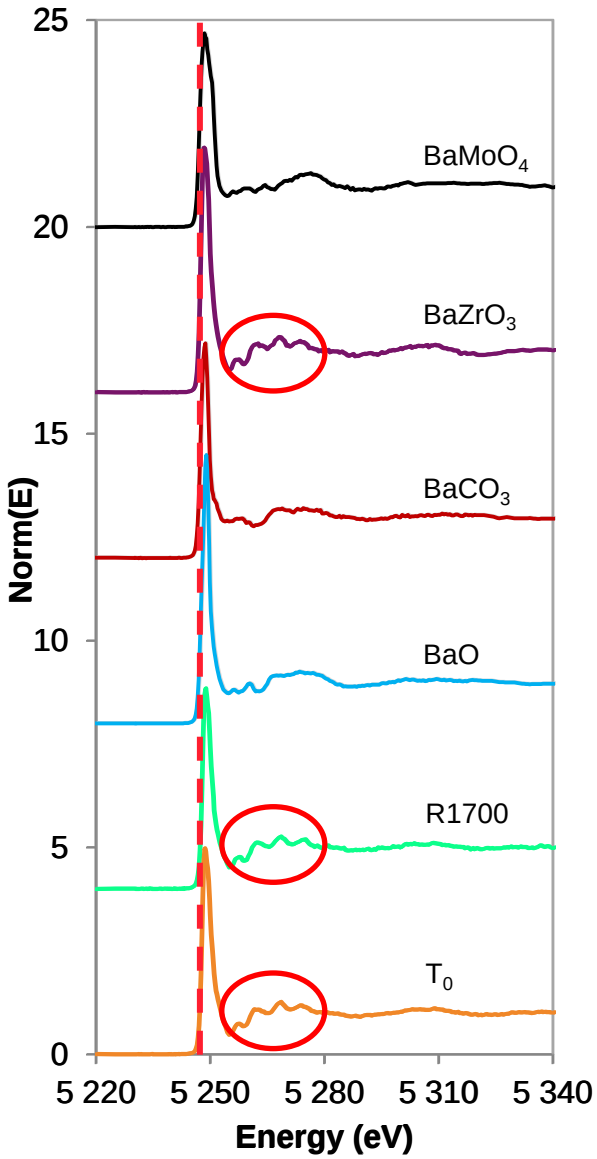
Zr XANES evolution from 1000°C in oxidizing conditions

Destruction of part of the $BaZrO_3$ phase and formation of ZrO_2 ?

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

CHARACTERIZATIONS

HERFD-XANES at Ba L_3-edge



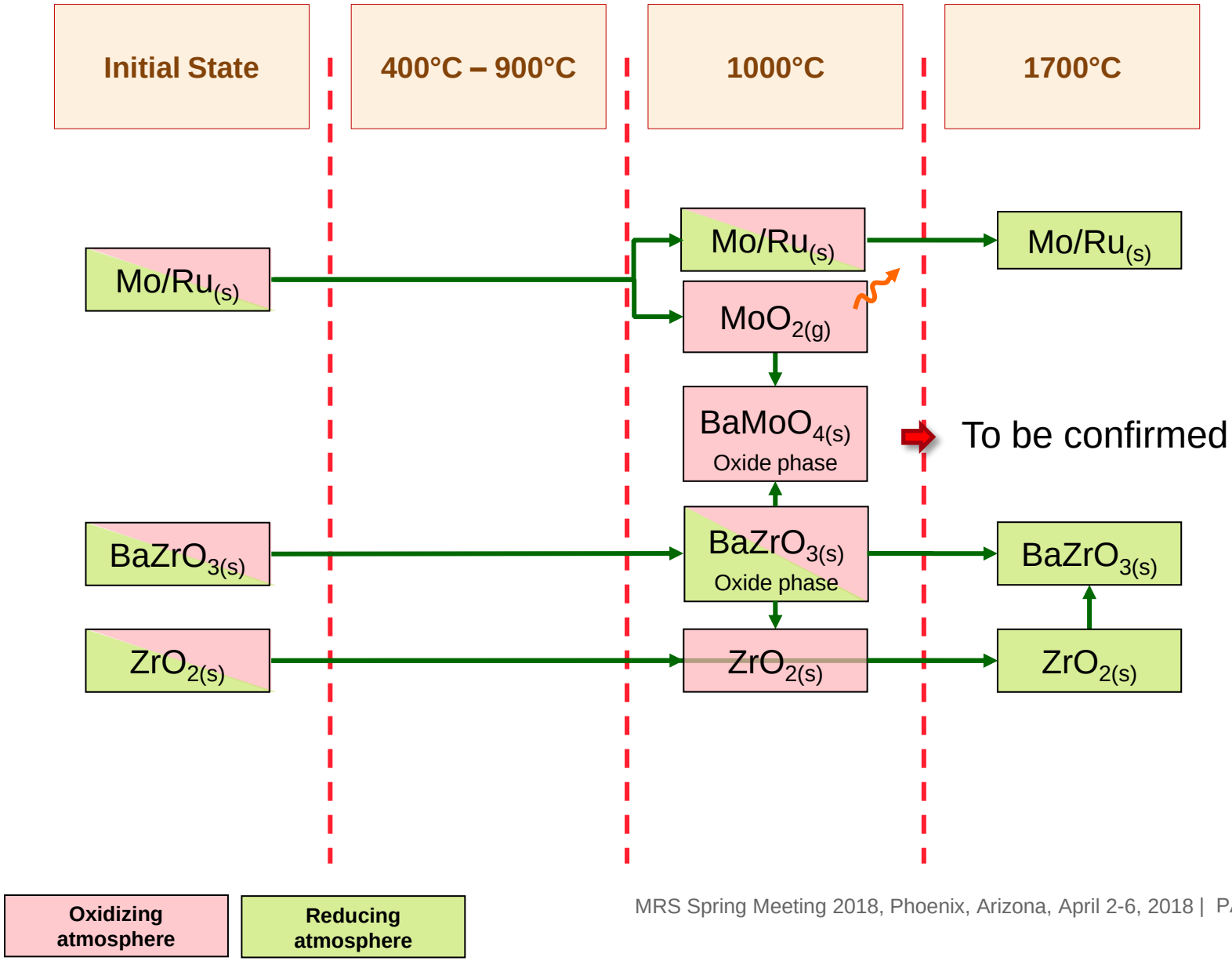
No Ba XANES evolution at 1700°C in reducing conditions

$BaZrO_3$

XANES analyses of the O-samples scheduled in April 2018

CONCLUSIONS

Evolution of the oxide phase



ESRF:

P. Colomp, O. Proux, M. Rovezzi, J. L. Hazemann

MARS/SOLEIL:

M. Hunault, P. L. Solari

CEA/LLCC:

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CEA/LCU:

F. Garel, P. Matheron, N. Tarisien, X. Itlis, H. Rouquette, J. Raynal, J. Lamontagne

CEA/LAMIR:

C. Thouzellier, J.C. Richaud, M. Pontillon, G. Volle, A. Gallais During, Y. Pontillon, F. Audubert, I. Moysan

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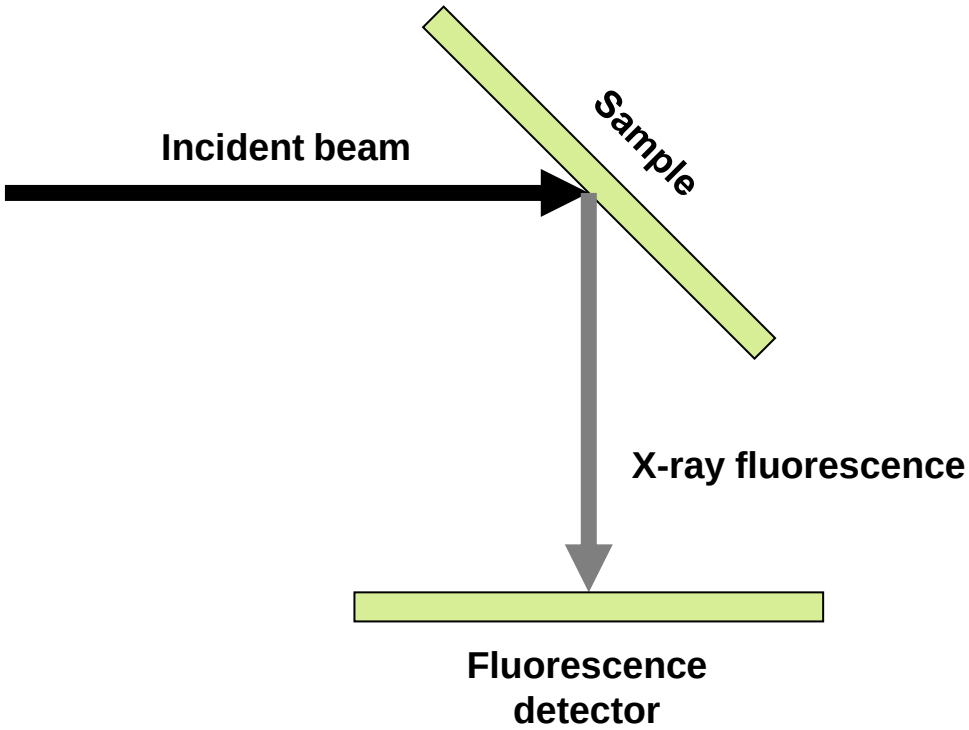
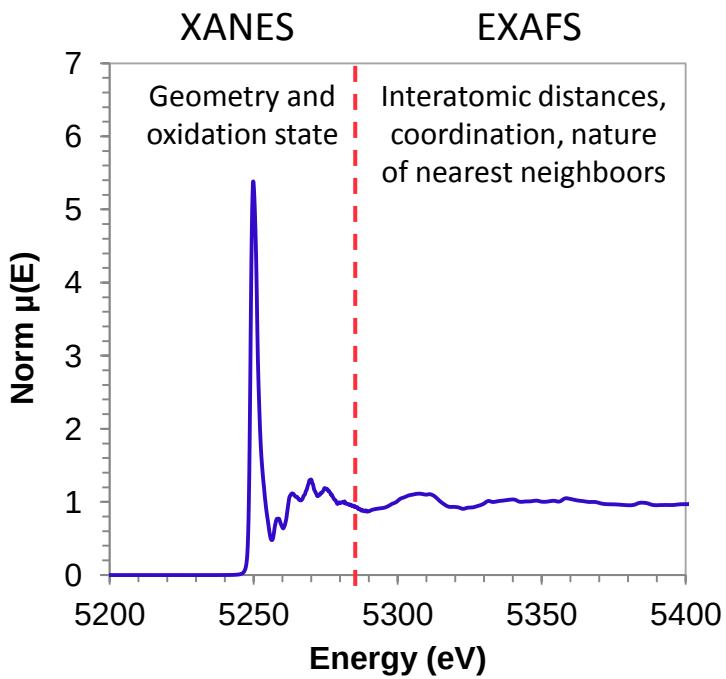
X-RAY ABSORPTION SPECTROSCOPY

Principle