Study on the behaviors of high-temperature oxidation and embrittlement of fuel cladding under air-steam conditions

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Introduction:

Fuel cladding may be exposed to air-steam atmosphere during sever accidents (SA) of NPP. Extensive calculation work performed by SA codes showed that current SA analysis needs to be refine. Recent experimental work showed much higher complexity of the oxidation process of Zr-alloy fuel cladding exposed under N_2-containing atmospheres, involving ZrN formation and its oxidation, which accelerates the oxidation of fuel cladding substantially compared to pure steam, *e.g.* [1-7]. It may consequently lead to higher production of heat and hydrogen, as well as to enhance embrittlement of the fuel cladding. Kinetic data and embrittlement of fuel cladding in N_2-containing atmosphere appears to be crucial in the process of refining the SA analysis.

Objective: To assess the oxidation behavior & mechanical properties of Zry-4 fuel cladding as a function of the air fraction in steam.

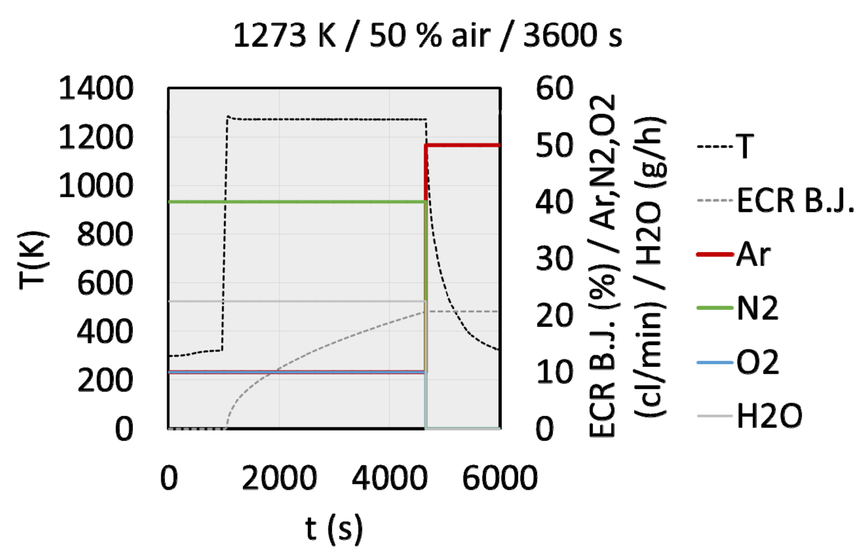
Experimental:

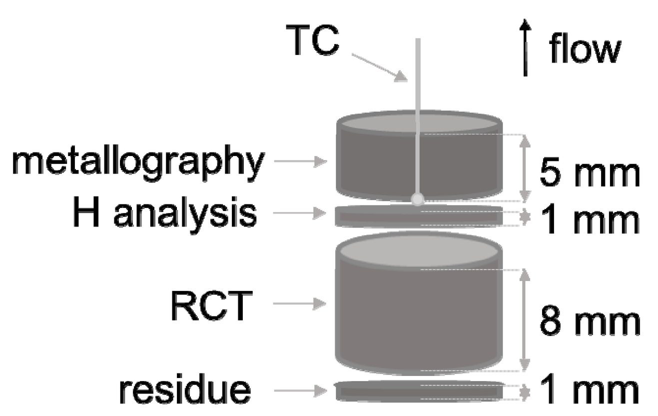
Non-irradiated, low-tin PWR 17x17 Zry-4 cladding tube (length ~ 15 mm, OD ~ 9.50 mm, wall thickness ~ 0.64 mm)

Air = 80 % N_2 + 20 % O_2 (≤ 1 L/min), steam (≤ 220 g/h), Ar - carrier gas (≤ 1 L/min)

Vertical tubular infrared furnace (quartz tube ID ~ 4 cm), Pt-Rh TC spot-welded on sample outer surface

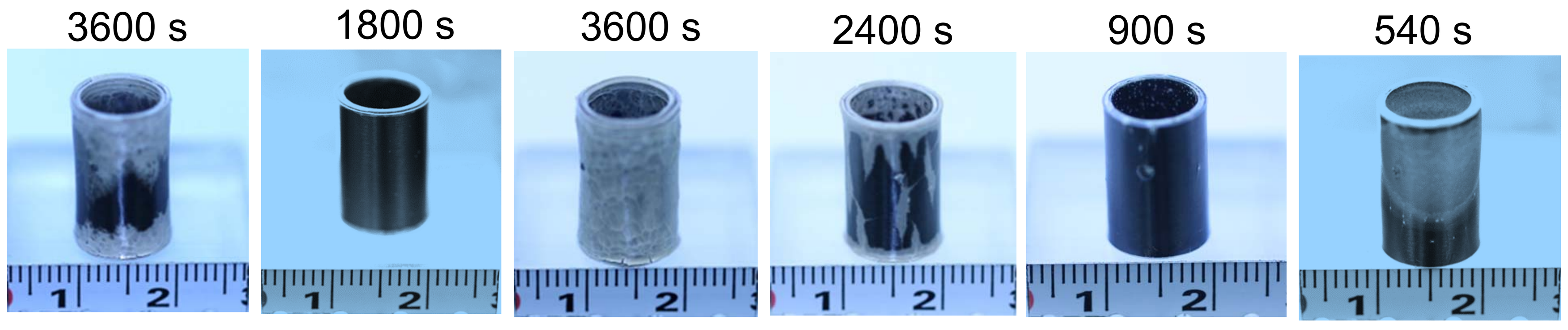
Heating rate = 10 K/s, T = 1273-1473 K, cooling-down naturally in opened furnace

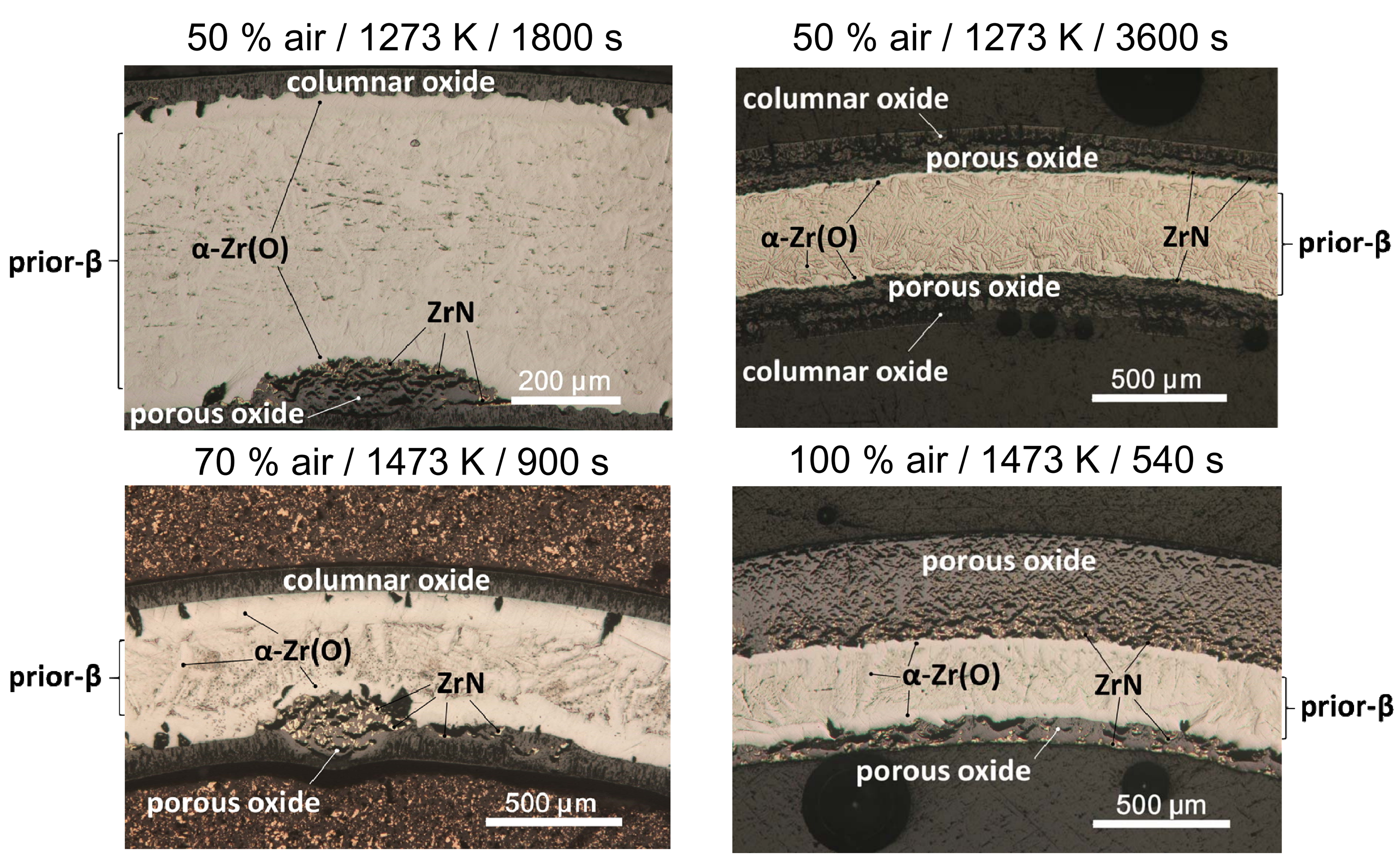


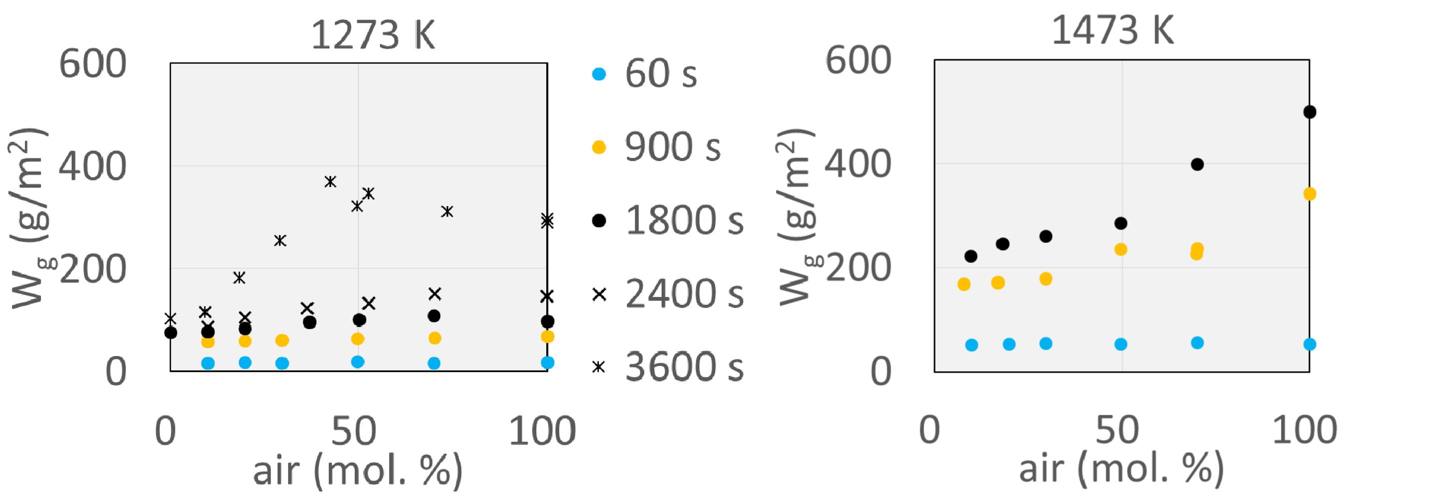


Results:

30 % air 1273 K 3600 s	50 % air 1273 K 1800 s	50 % air 1273 K 3600 s	70 % air 1273 K 2400 s	70 % air 1473 K 900 s	100 % air 1473 K 540 s

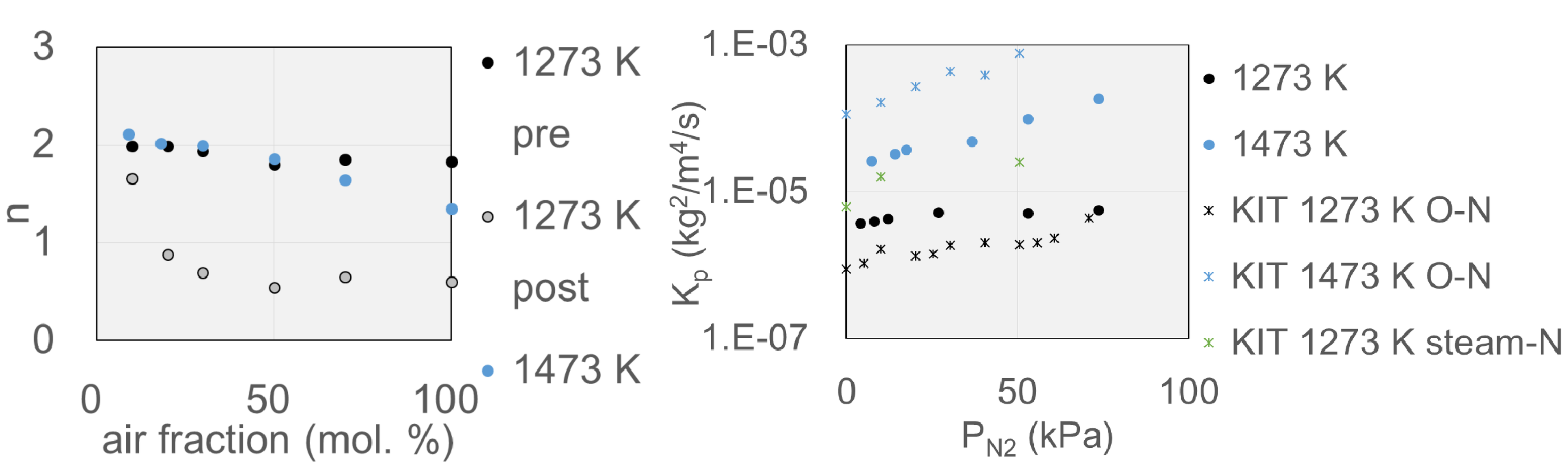




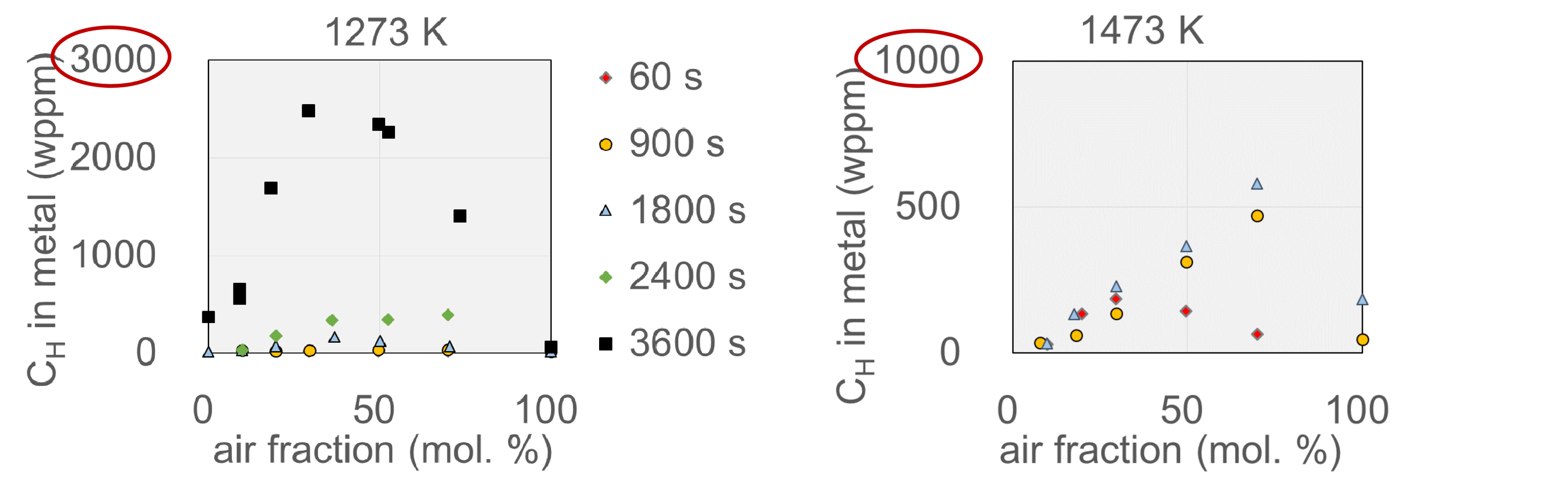


- Effect of the air fraction on W_g is substantial: tendencies differ depending on T (effect of partial pressure of O_2 & flow rate)

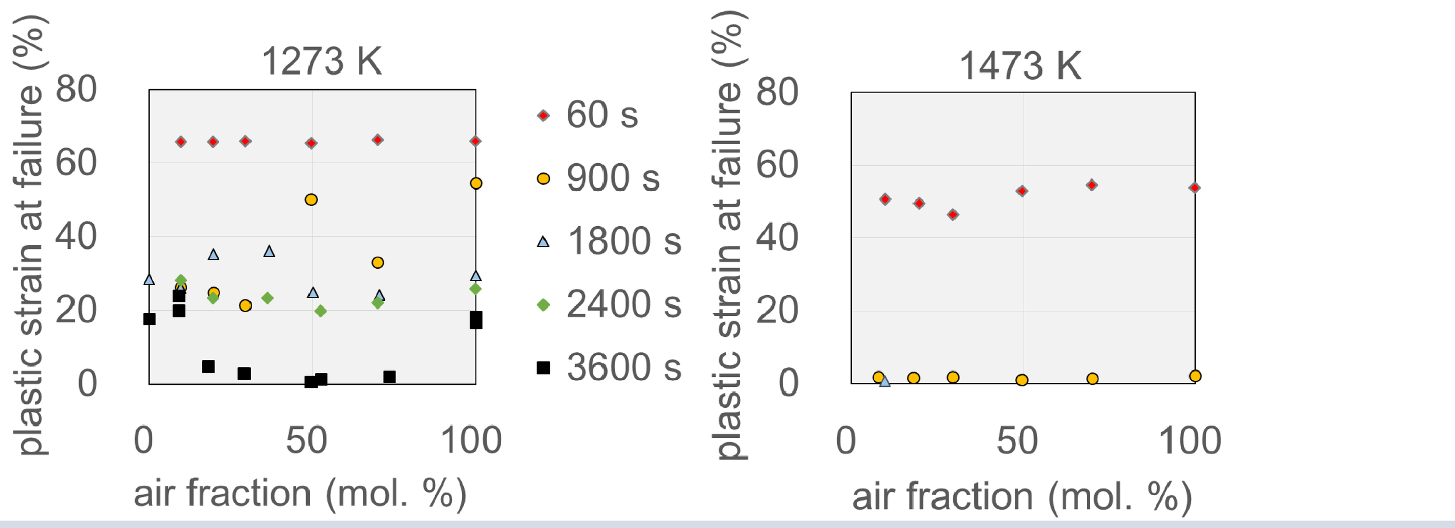
$$w_g^n = K \cdot t \qquad n = 2 \rightarrow w_g^2 = K_p \cdot t$$



- Higher air fractions in steam => significant deviation from the parabolic law due to ZrN formation
- Significant differences between JAEA & KIT => effect of experimental setting => more parameters need to be considered

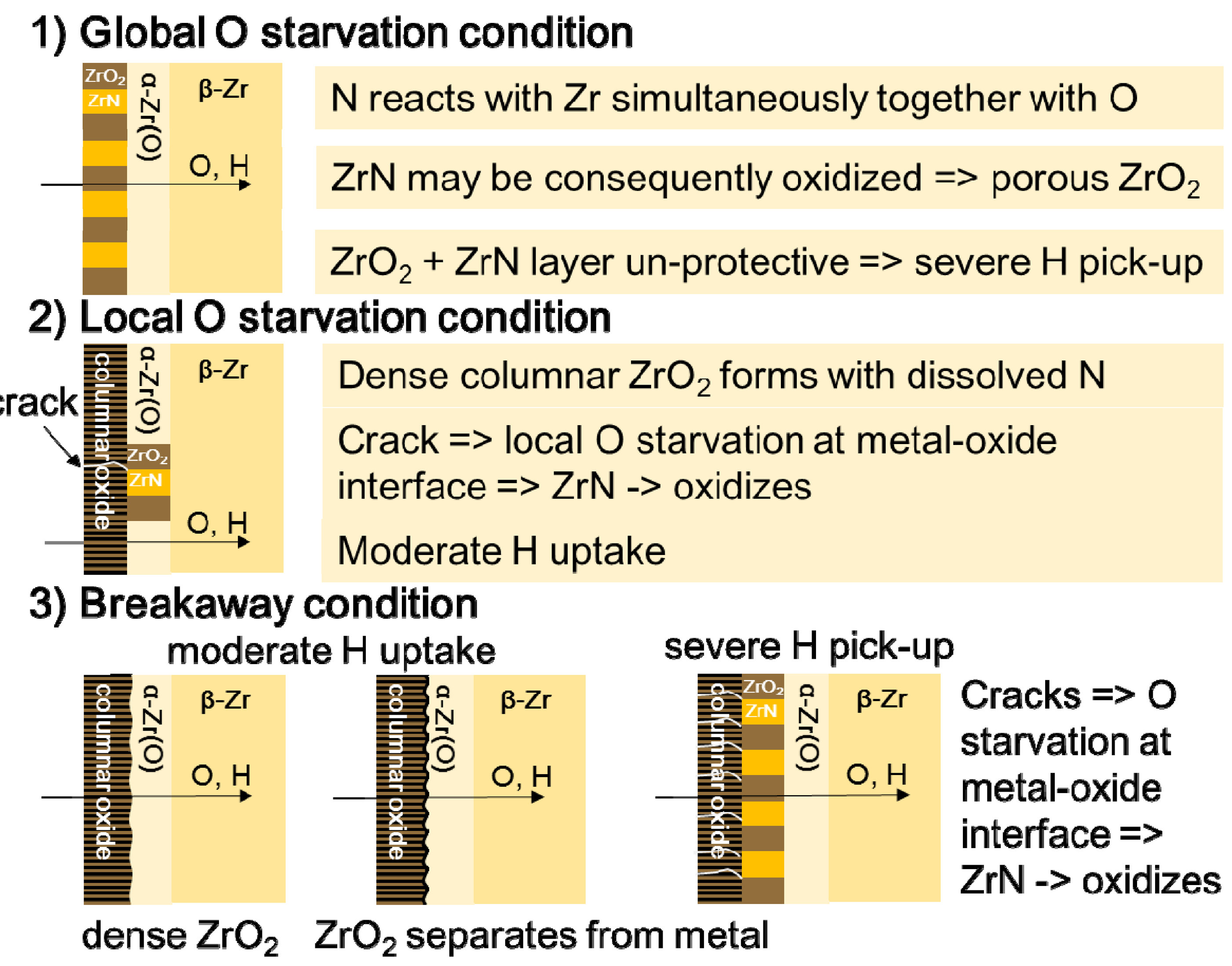


- Increasing air fraction in steam => oxide properties deteriorate (ZrN & porous oxide formation) => higher amount of H is absorbed
- Higher air fractions => lower P_{H2} => H uptake decreases



- Significant reduction of the plastic strain at failure due to large H pick-up

Oxidation mechanism in steam-air atmospheres



Conclusions:

- Air in steam affects the oxidation behavior of fuel cladding substantially through ZrN formation and its oxidation
- ZrO_2 loses its protectiveness against O, N & H => oxidation kinetics accelerates and H uptake increases substantially => plastic strain at failure may be reduced significantly
- The oxidation kinetics has been assessed in dependence on the air fraction in steam at 1273 -1473 K
- More parameters have to be considered: partial pressure of oxygen, flow rate

References:

[1] Duriez C, Dupont T, Schmet B, Enoch F. Zircaloy-4 and M5 high temperature oxidation and nitriding in air. J. Nucl. Mater. 2008; 380: 30-45.

[2] Steinbrück M. Prototypical experiments relating to air oxidation of Zircaloy-4. J. Nucl. Mater. 2009; 392: 531-544.

[3] Steinbrück M. High-temperature reaction of oxygen-stabilized α-Zr(O) with nitrogen. J. Nucl. Mater. 2014; 447: 46-55.

[4] Lasserre M, Peres V, Pijolat M, Coindreau O, Duriez C, Mardon J-P. Modelling of Zircaloy-4 accelerated degradation kinetics in nitrogen-oxygen mixtures at 850 °C. J. Nucl. Mater. 2015; 462: 221-229.

[5] Negyesi M, Amaya M. Oxidation behavior of Zry-4 in steam-air mixtures at high temperature. Proc. Top Fuel 2016; 2016 September 11-16; Boise (Idaho, USA).

[6] Negyesi M, Amaya M. Oxidation kinetics of Zry-4 fuel cladding in mixed steam-air atmospheres at temperatures of 1273 – 1473 K. J. Nucl. Sci. Technol. 2017; 54: 1143-1155.

[7] Negyesi M, Amaya M. High temperature oxidation of Zry-4 in oxygen-nitrogen atmospheres. Proc. ICONE25; 2017 July 2-6; Shanghai (China).