



Single crystal and sintered alumina corrosion in liquid sodium

J.-L. Courouau, M.-C. Steil, J. Fouletier, F. Rouillard, V. Lorentz, P. Bonnaillie, A. Muccioli, J. Unger, S. Tricoit, M. Tabarant

► To cite this version:

J.-L. Courouau, M.-C. Steil, J. Fouletier, F. Rouillard, V. Lorentz, et al.. Single crystal and sintered alumina corrosion in liquid sodium. HTCPM 2016 - 9th International Symposium on High-Temperature Corrosion and Protection of Materials, May 2016, Les Embiez, France. hal-02445715

HAL Id: hal-02445715

<https://cea.hal.science/hal-02445715>

Submitted on 20 Jan 2020

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

SINGLE CRYSTAL AND SINTERED ALUMINA CORROSION IN LIQUID SODIUM

Jean-Louis COUROUAU^a, Marlu-César STEIL^{b,c}, Jacques FOULETIER^{b,c}, Fabien ROUILLARD^a, Véronique LORENTZ^a, Patrick BONNAILLIE^d, Anthony MUCCIOLI^a, Jonathan UNGER^a, Sandy TRICOIT^a, Michel TABARANT^e

^a: Den-Service de la Corrosion et du Comportement des Matériaux dans leur Environnement (SCCME), CEA, Université Paris-Saclay, F-91191, Gif-sur-Yvette, France

^b: Univ. Grenoble Alpes, LEPMI, F-38000 Grenoble, France

^c: CNRS, LEPMI, F-38000 Grenoble, France

^d: Den-Service de Recherches de Métallurgie Physique (SRMP), CEA, Université Paris-Saclay, F-91191, Gif-sur-Yvette, France

^e: Den-Service d'Etudes Analytiques et de Réactivité des Surfaces (SEARS), CEA, Université Paris-Saclay, F-91191, Gif-sur-Yvette, France

May 15-20 2016, Les Embiez, France

Context and objectives

Ceramic materials for the core catcher:

Alumina is one of the candidate under studies for the 4th nuclear reactor programme (ASTRID) (Zirconia being the reference ceramic)

Apart from thermomechanical effects, the corrosion proceed by reaction with liquid sodium or its impurities (mainly oxygen), giving either homogeneous corrosion or localized corrosion if reactants are concentrated at the grain boundaries (GB).

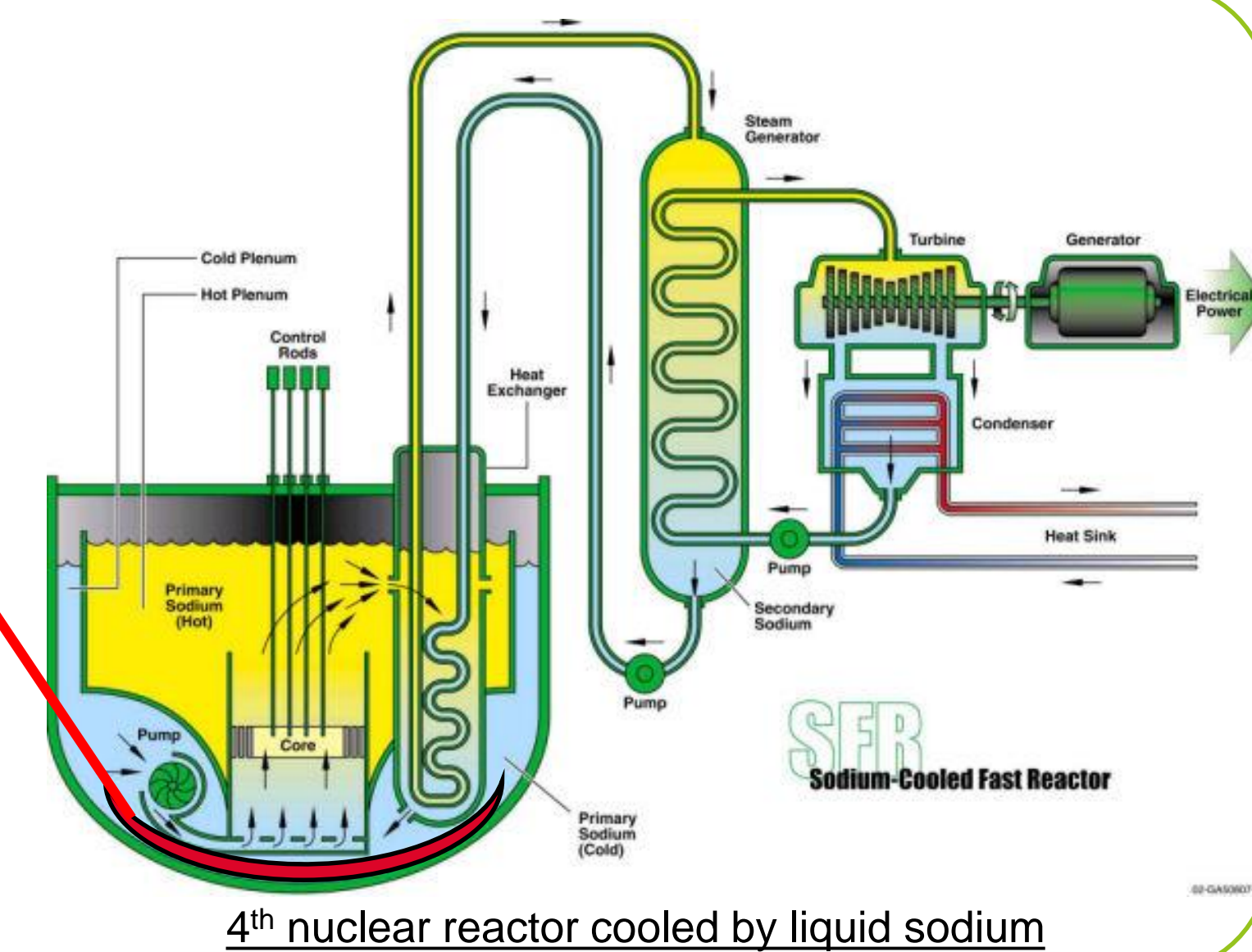
Silica traces segregated at the GBs are reported to enhance fast intergranular corrosion (IGC) in alumina that forbid the study of other corrosion phenomenon [Kano-95].

Any interactions between the liquid metal and the structural materials must be well understood and predicted to guarantee a 60 years service life-time as well as resistance to incidental conditions.

Compare polycrystal to monocrystal to define IGC-free ceramic material, to identify corrosion phenomena and quantity first corrosion rates vs. T and [O]

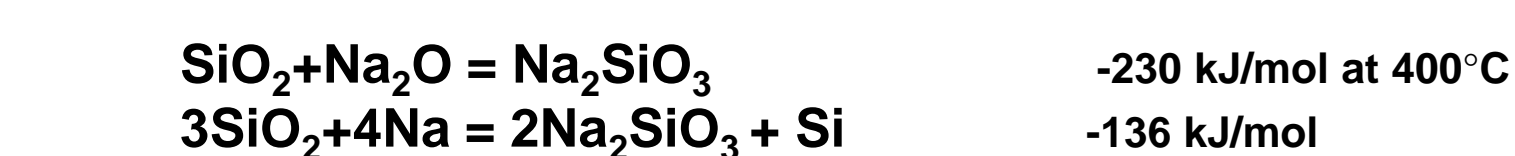
Core catcher : collect and allow cooling down all of melted reactor materials in case of reactor meltdown

Conditions: in contact with liquid sodium, 60 years, 400°C, [O]<~10 wt. ppm as potential casing of the ceramic material cannot be inspected nor repaired

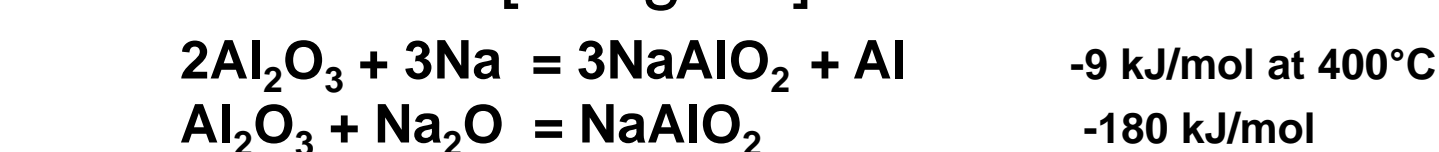


Thermochemistry

- Aluminate and silicate are more stables at any reactor operating conditions, meanings kinetics control is required
- Kinetics for silicates formation is reported as fast for T>~300°C [Jung-83]

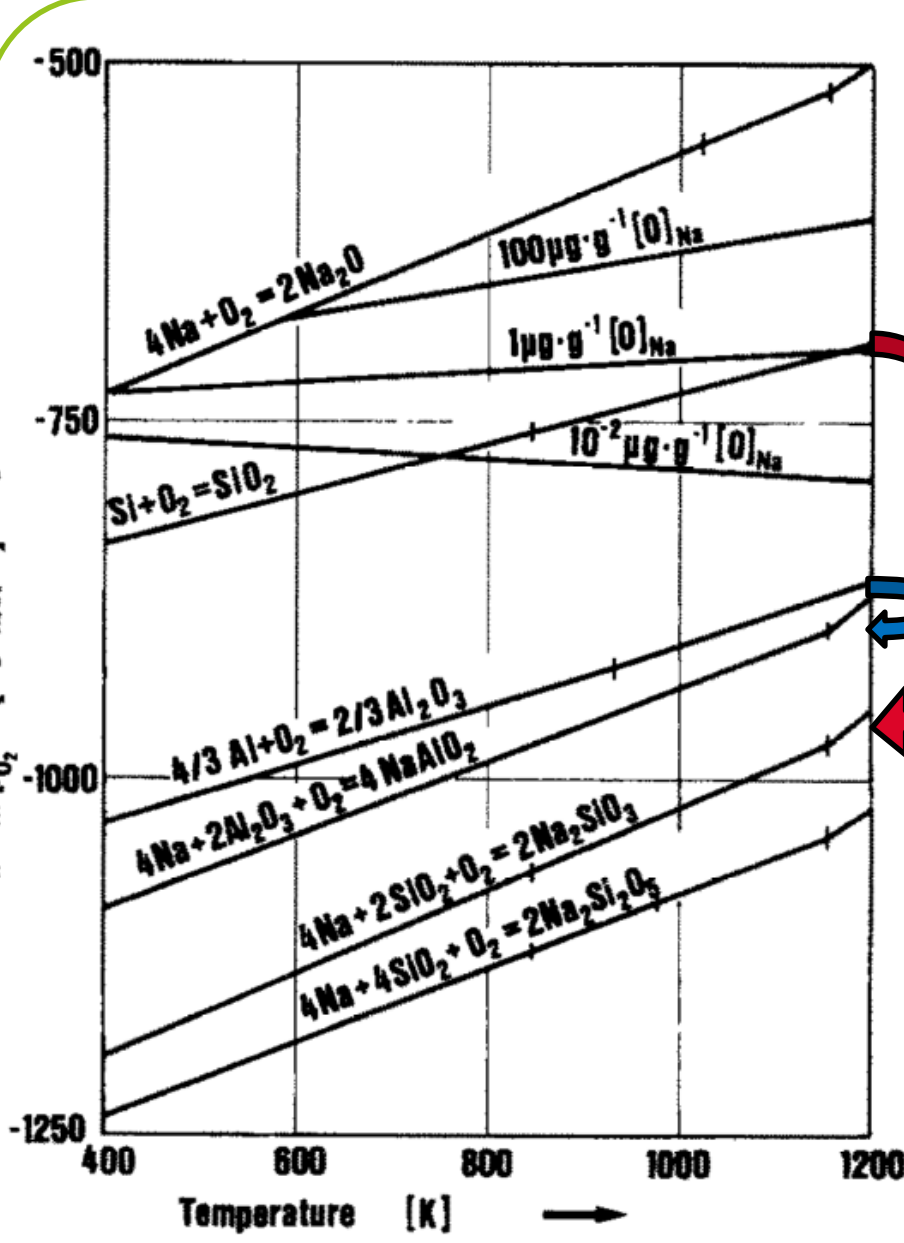


- Almost no data available for alumina, reaction not identified [Jung-83]



This study :

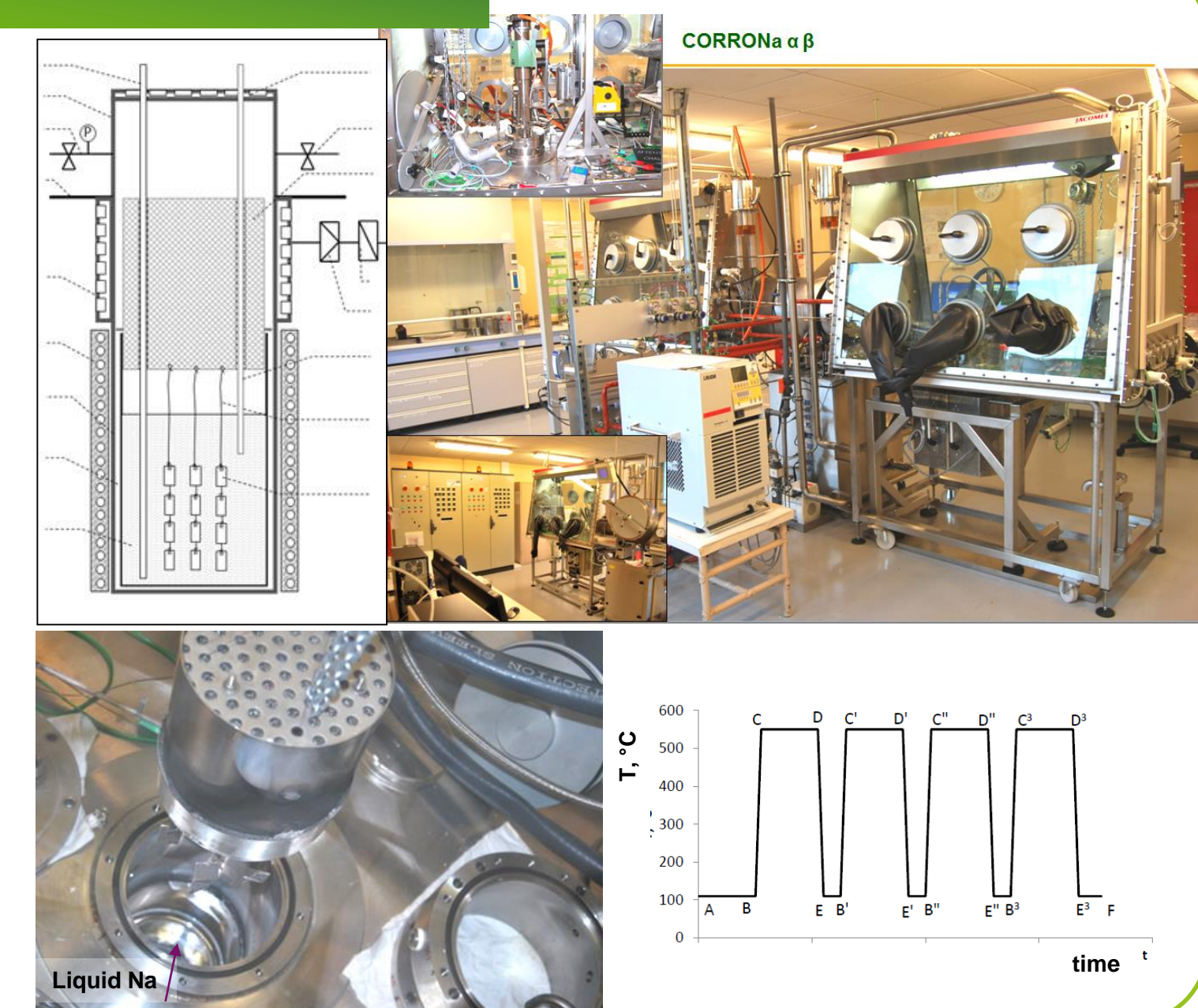
T & [O] effects on 'dissolution' of alumina and IGC



Ellingham diagram [Jung-83]

Experimental

- Static liquid sodium** (2.3 kg- Mo crucible)
- Sintered alumina : Degussit AL23 - 99.7% purity (60 ppm SiO₂), no open porosity, tube (OD4- ID2 mm) and Sapphire supplied by Rubis SA(D20 mm rod)
- Sodium chemistry control**
 - Purification (hot trapping by Zr foil at 630°C – 75h: <0.1 wt. ppm to 1-5 ppm)
 - Decantation at low T (105°C – 100h : 40 ppm)
 - Oxygen addition (Na₂O): 200 ppm
- Temperature: 450 °C & 550 °C**
- Duration: from 250 h to 4600 h**

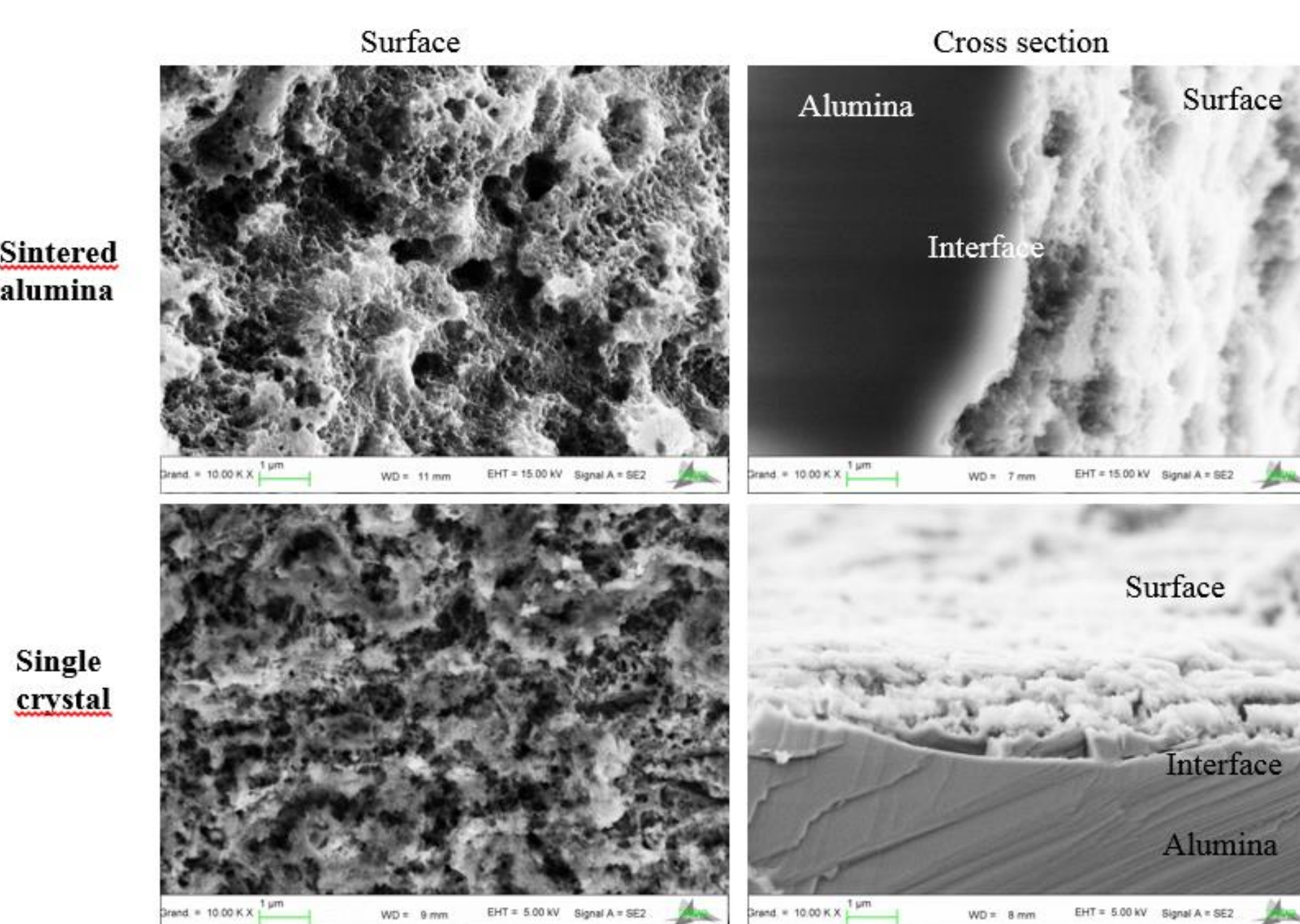


Morphologies

Surface and fractured cross section of sintered alumina & Sapphire

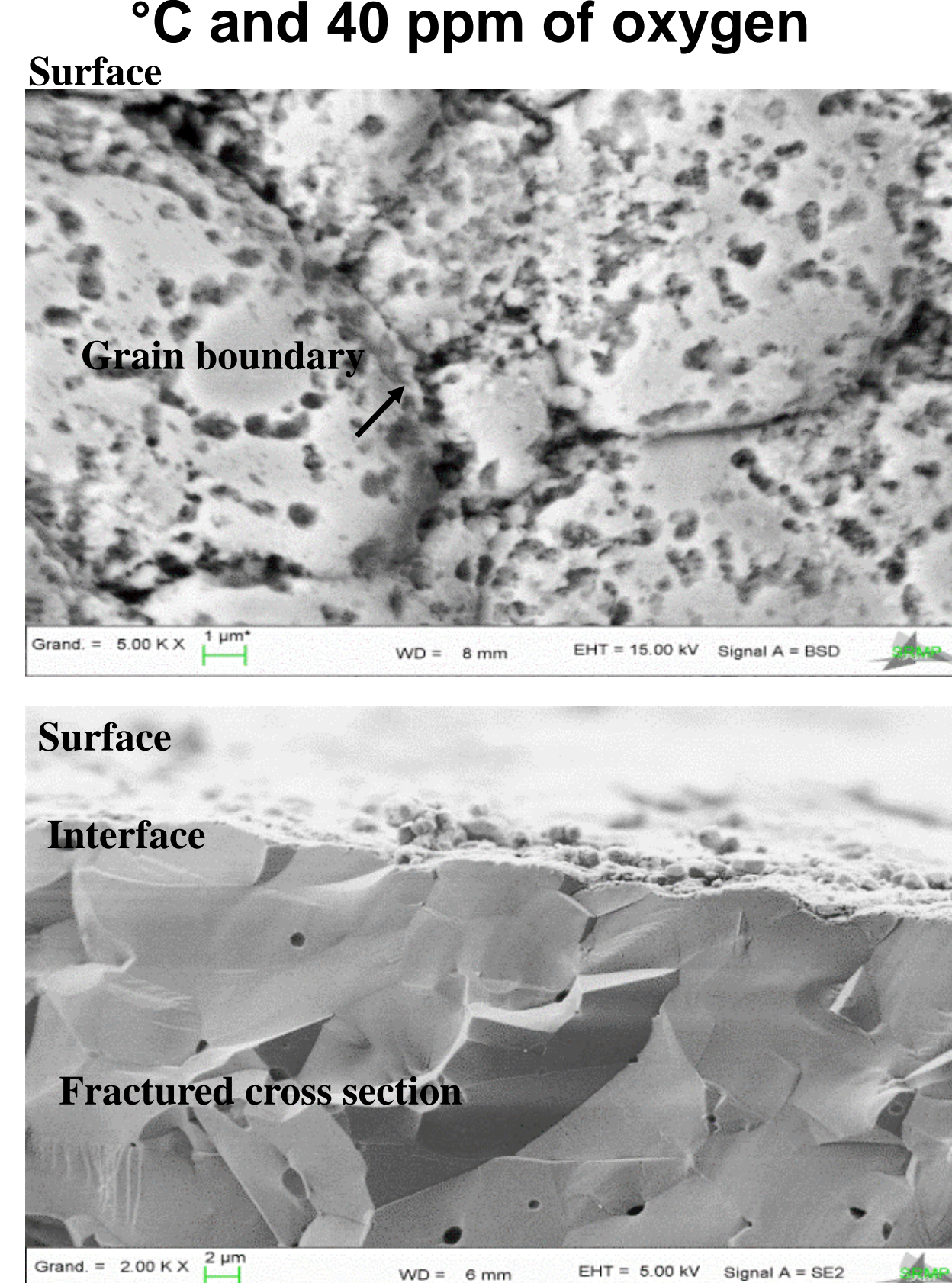
Exposure at 550 °C, 1-5 ppm of oxygen and 3000 h

Sintered alumina after 1000h at 450 °C and 40 ppm of oxygen



Fractured surfaces are trans-granular for all conditions up to the interface, demonstrating the absence of IGC

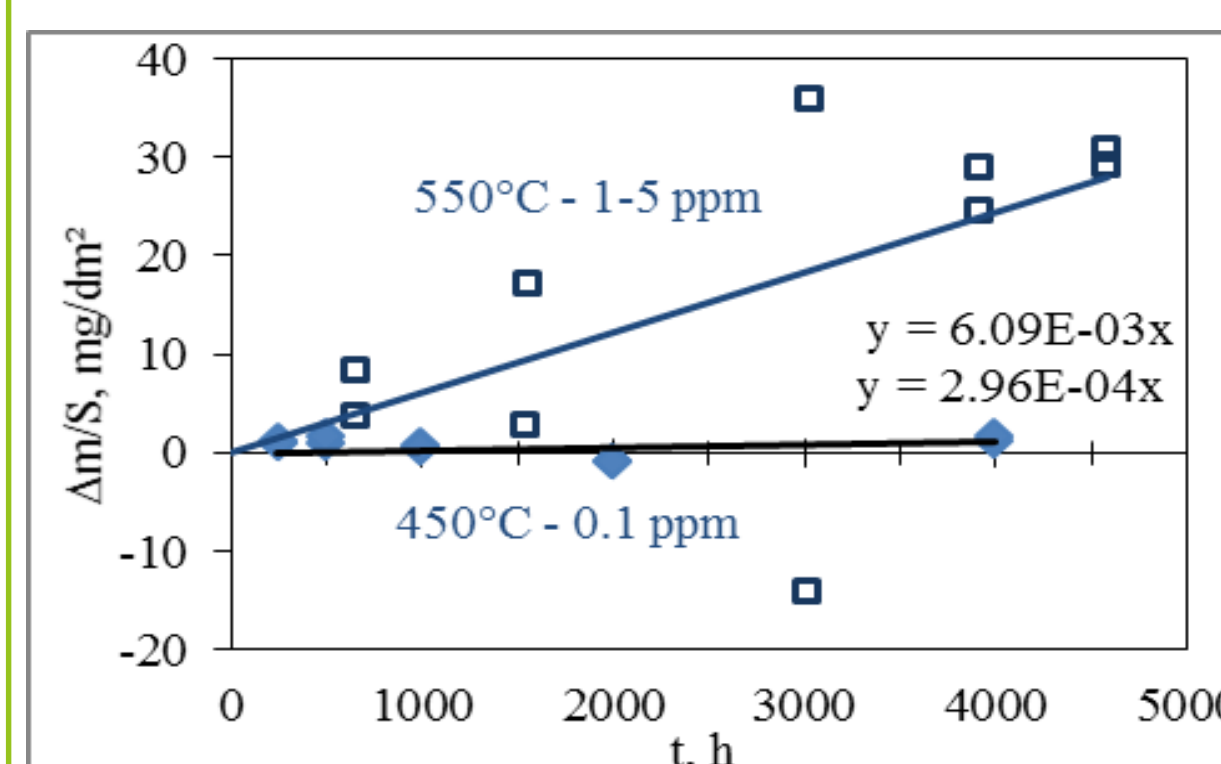
Pure and dense sintered alumina presents the same behavior as the single crystal to the condition that the grain boundary corrosion is negligible, which is the case for this high purity grade of sintered alumina



Porosity dimensions tend to increase with time and temperature (in relation with sodium residues after ethanol cleaning) eventually covering all of the surface

Corrosion rates

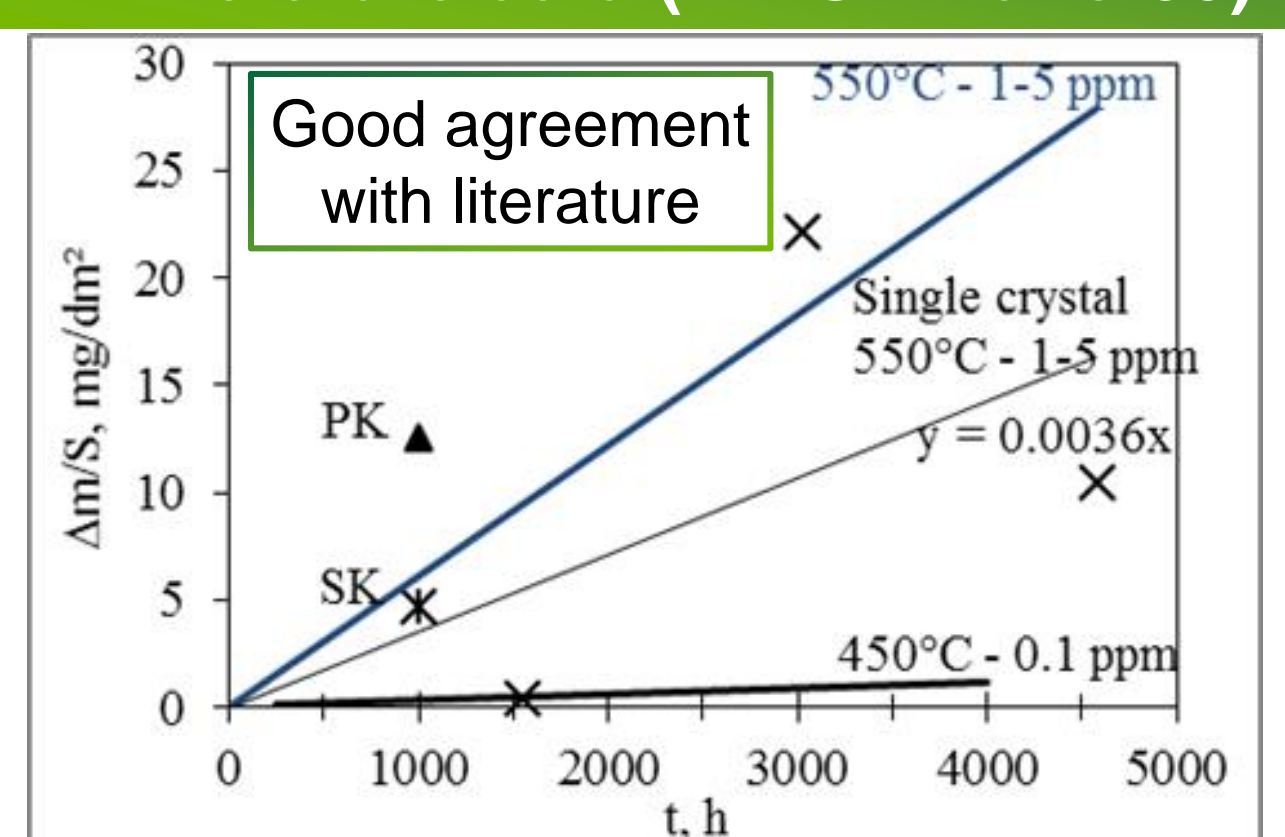
Weight loss of sintered alumina at 450°C and 550°C at low O



Supposing linear kinetics for low O content:

- 0.14 mg/dm²/d or 1.4 μm/y at 550 °C
- 0.007 mg/dm²/d or 0.07 μm/y at 450 °C

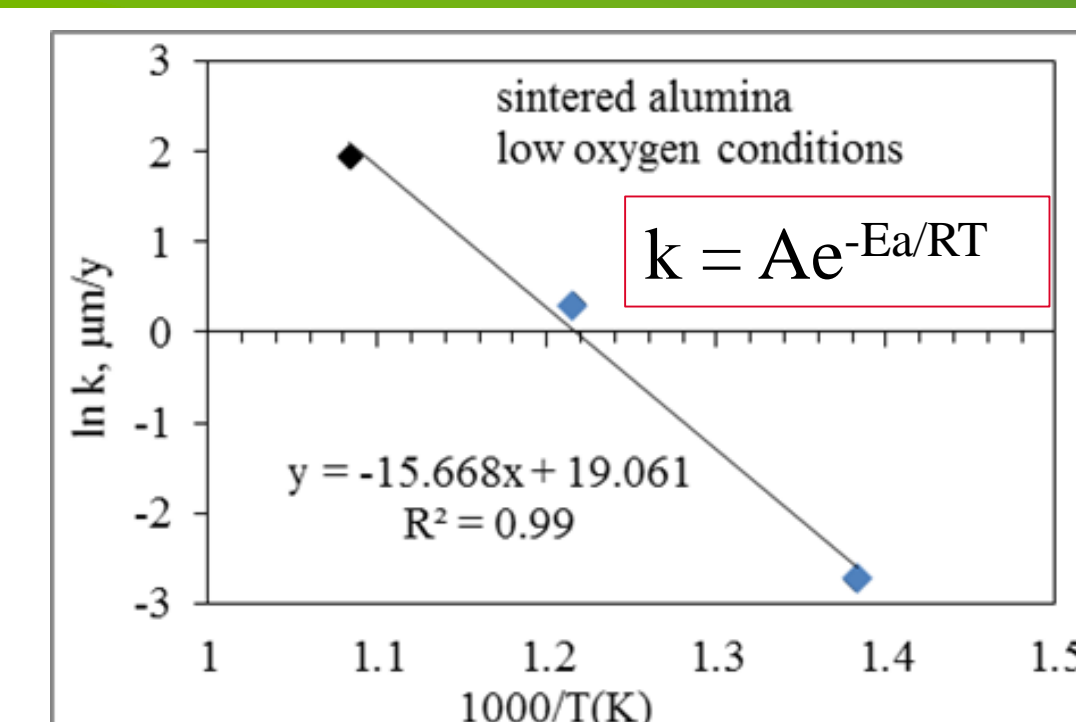
Single crystal weight loss at 550°C compared to sintered alumina and literature data (PK-SK Kano-95)



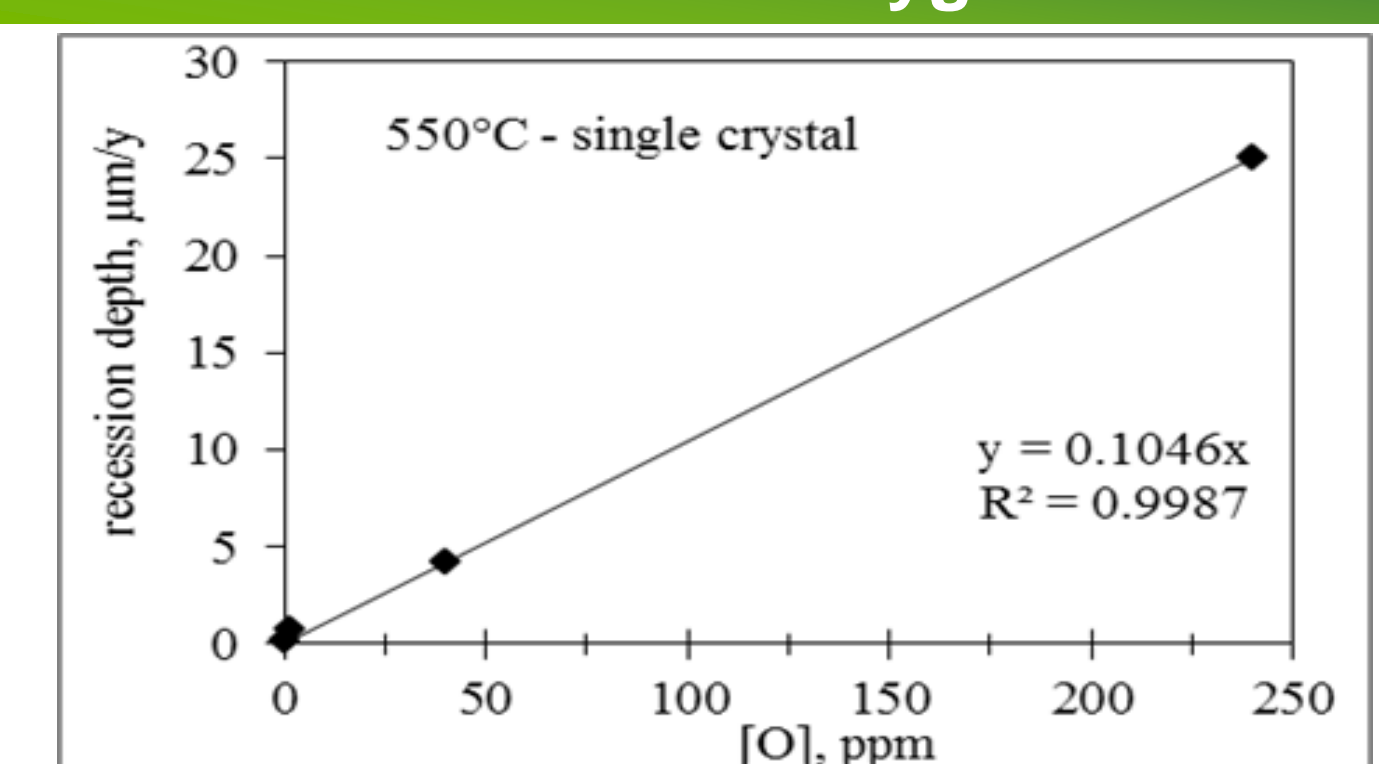
GB effect: dissolution rate twice greater for sintered alumina than for single crystal

Weight loss interpreted as equivalent to a dissolution with intermediate formation of NaAlO₂ that dissolves in Na or in water after sodium exposure

Arrhenius plot for sintered alumina at 450, 550 and 650 °C [Kano-95] in low oxygen content



Recession depth of single crystal at 550 °C as function of oxygen content



Dissolution rate: $R=0.011 \cdot [O]$ (mg/dm²/d – ppm)

Temperature effect:

Apparent activation energy of 130 kJ/mol or 1.35 eV

Dissolved oxygen effect:

Straight dependence with oxygen content:
- Main reaction is $\text{Al}_2\text{O}_3 + \text{Na}_2\text{O}(\text{d}) = \text{NaAlO}_2$
- First-order reaction kinetics with respect to oxygen content at the temperature of 550 °C

Conclusion and prospect

First step to a better understanding of the mechanisms of degradation and a guide for the elaboration or selection of ceramics that would be compatible with sodium on the long term:

- 1- High density, no open porosity and high purity required to avoid intergranular corrosion in liquid Na
- 2- Homogeneous corrosion of alumina equivalent to dissolution through a mechanism yet to elucidate
- 3- Corrosion rates of 0.07 to 0.14 μm/y assessed from weight losses measured at 450°C and 550°C in low oxygen conditions in agreement with the few available literature data
- 4- Apparent activation energy evaluated to 130 kJ/mol (1.35 eV) suggesting solid phase diffusion control as limiting step for the corrosion mechanism
- 5- First-order reaction kinetics with respect to dissolved oxygen at 550 °C

In prospect : confirm these results with additional tests, elucidation of the formation of sodium aluminate mechanism, hydrodynamics effect on corrosion rate, and extrapolation of these results to other ceramic materials such as zirconia.

Aknowledgements

The CEA R4G project, and in particular F. Serre, C. Cabet and O. Gastaldi are gratefully acknowledged for funding this research, as well as B. Duprey for having provided one weight loss on single crystal obtained on the second testing bench of the Lab (CORRONa-2).