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Analysis of the sintering stresses and shape distortion produced in co-firing of CGO-LSM/CGO bi-layer porous structures

*De Wei Ni *, Vincenzo Esposito, Cristine Grings Schmidt, Tesfaye Tadesse Molla, Kjeld Bøhm Andersen, Andreas Kaiser, Severine Ramousse, Nini Pryds*

Department of Energy Conversion and Storage, Technical University of Denmark - DTU, Building 775, P.O. Box 49, Frederiksborgvej 399, DK-4000 Roskilde, Denmark.

** Contact : dwei@risoe.dtu.dk*

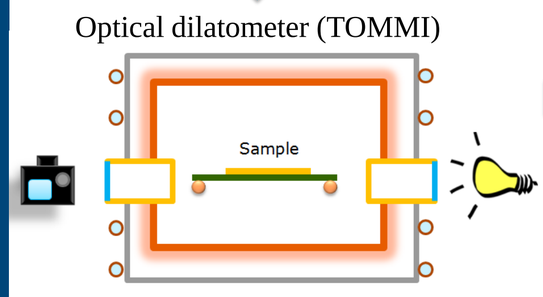
Motivation

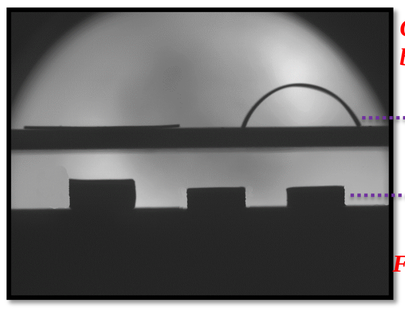
Gadolinium-doped cerium oxide (CGO) and lanthanum strontium manganate (LSM) are electro-ceramics materials with high potential for several electrochemical applications such as solid Oxide Fuel Cell (SOFC), gas separation membranes, and flue gas purification devices. Especially for novel electrochemical flue gas purification devices, multilayer structures with alternating porous layers of CGO and a LSM/CGO mixture are used to achieve specific functional requirements. In a manufacturing process of such ceramic multilayer devices, co-firing is one of the critical steps as many defects such as cracks, de-lamination and shape distortion can result as a consequence of sintering mismatch stresses caused by the strain rate difference between layers. This work seeks to understand the underlying mechanisms that occur during the co-firing of porous CGO-LSM/CGO bi-layer laminates, by evaluating the sintering mismatch stress and distortion development through modeling and experiments.

Experimental procedure

Tape Casting + Lamination
Porous CGO and LSM/CGO layers

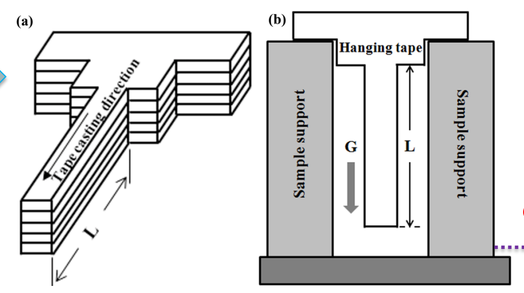
Optical dilatometer (TOMMI)





Co-firing of bi-layer

Free sintering



Vertical sintering

Co-firing of bi-layer laminate : Cai's Model

- Normalized curvature rate:

$$\dot{k} = \frac{d((t_1+t_2)/r)}{dt} = \frac{6(m+1)^2 mn}{m^4n^2 + 2mn(2m^2+3m+2)+1}\Delta\dot{\varepsilon}$$

- Sintering mismatch stress (viscous stress):

$$\sigma_1 = \left[\frac{m^4+mn}{n^2+2mn(2m^2+3m+2)+m^4}\right]\hat{\sigma}_1$$

$$\hat{\sigma}_1 = \frac{\eta_1}{1-\nu_1}\Delta\dot{\varepsilon}_{2-1} = \left[\frac{m^4n^2+2mn(2m^2+3m+2)+1}{6(m+1)^2 mn}\right]\eta_1\dot{k}$$

Results and discussion

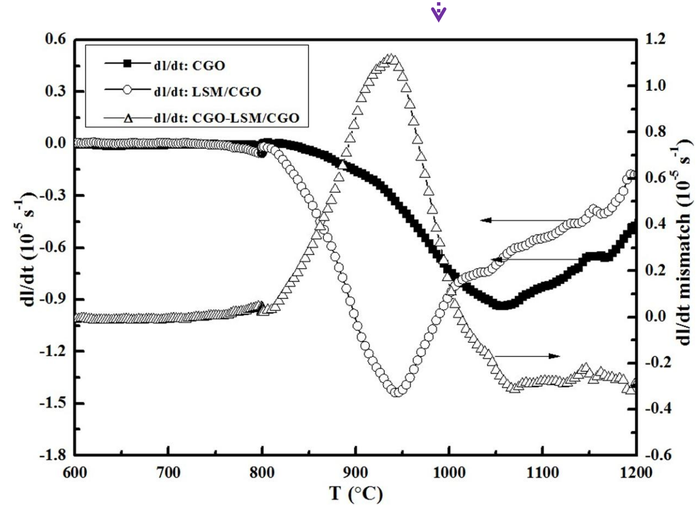


Fig.1 Free sintering: linear strain rate of porous LSM/CGO and CGO individual layers, and linear strain rate mismatch between them.

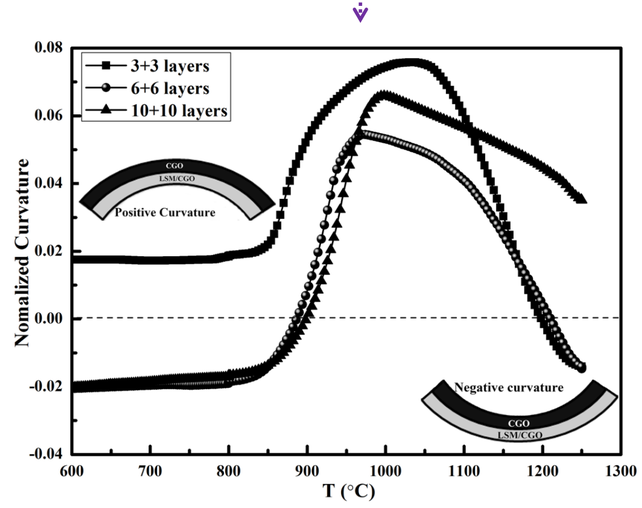


Fig.2 Experiment derived normalized curvature for the asymmetric CGO-LSM/CGO bi-layer laminates with different thickness.

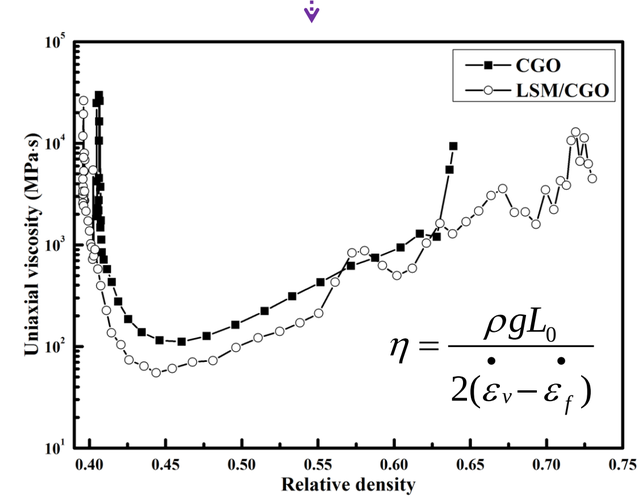


Fig.3 Experiment determined uniaxial viscosity for porous CGO and LSM/CGO tapes as a function of temperature via vertical sintering method.

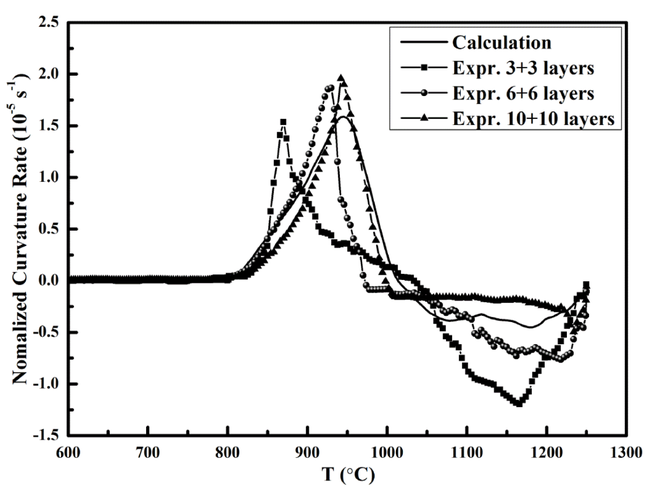


Fig. 4 Comparison between experimentally derived and theoretically calculated normalized curvature rate for the asymmetric CGO-LSM/CGO bi-layer laminates.

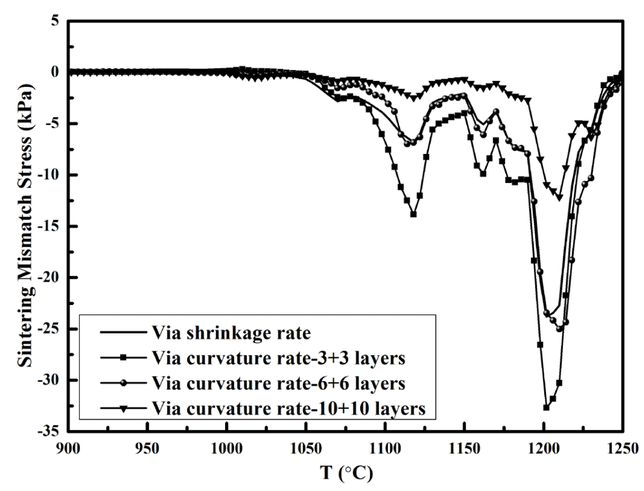


Fig. 5 Sintering mismatch stress developed in LSM/CGO layer calculated by linear strain rate difference and normalized curvature rate via Cai's model.

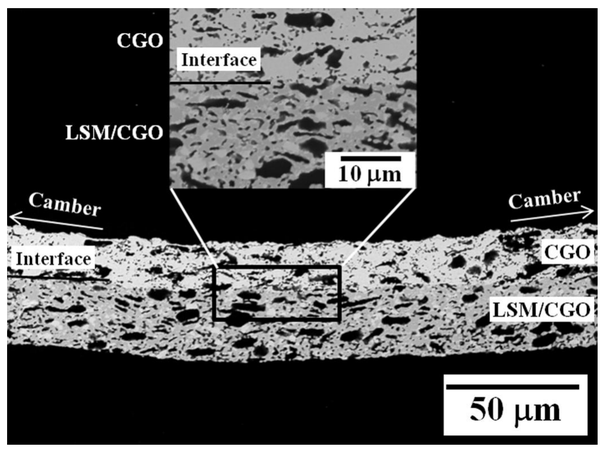


Fig. 6 SEM image of the interface for the asymmetric CGO-LSM/CGO bi-layer after sintering at 1250 °C for 4h.

Conclusion

- Uniaxial viscosity was determined via load-free sintering (*i.e.* laying on a horizontal support) and hanging sintering (*i.e.* under the effect of its own weight – a low tensile stress).
- The normalized curvature rate developed during co-firing of porous CGO and LSM/CGO bi-layer laminates were theoretically calculated via Cai's model, which corresponded with the experimental data.
- The sintering mismatch stress in co-fired CGO-LSM/CGO bi-layer were significantly lower than general sintering stresses. As a result, no co-firing defects were observed in the bi-layer laminates, illustrating an acceptable sintering compatibility of the ceramic layers.

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