Water vapor corrosion of EBC candidate materials

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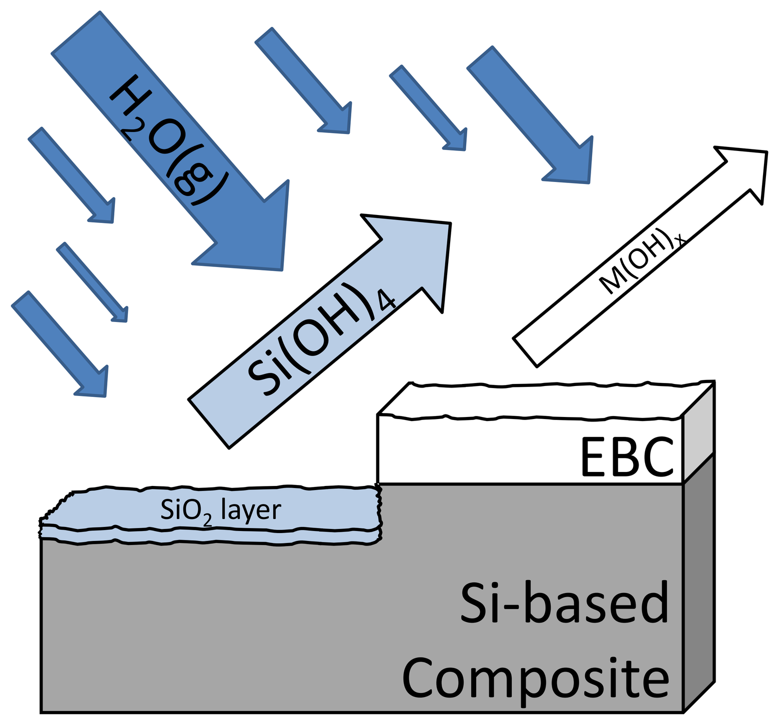
Motivation:

One of the major goals for gas turbine engines is to increase the engine efficiency which necessitates operating at higher temperatures. Increased engine temperatures have been addressed through silicon-based ceramics and composites which have shown great promise as replacements for hot-stage alloy components of gas turbine engines. However, these Si-based materials are susceptible to the effects of water vapor, (Ca-Al-Mg-Si-O) CMAS interaction, and oxidation, among other issues at high temperature. Hence, environmental barrier coating (EBC) materials are sought after to protect next generation high temperature engine components. This study focuses on one aspect of high temperature degradation in EBC materials, namely the recession caused by the interaction of the coating with water vapor.

Background:

- In 1997, Opila *et al* demonstrated that Si-based composites were vulnerable to recession by water vapor at elevated temperature (i.e. oxidation and hydroxide formation. See Eqs. at right)
- A protective layer, composed of rare earth silicates (RES), was proposed to protect against recession.[3]
- It was found that mono-silicates (i.e. RE_2SiO_5) tended to have better protection against recession over di-silicates (i.e. $RE_2Si_2O_7$) which led to their use in the current study.

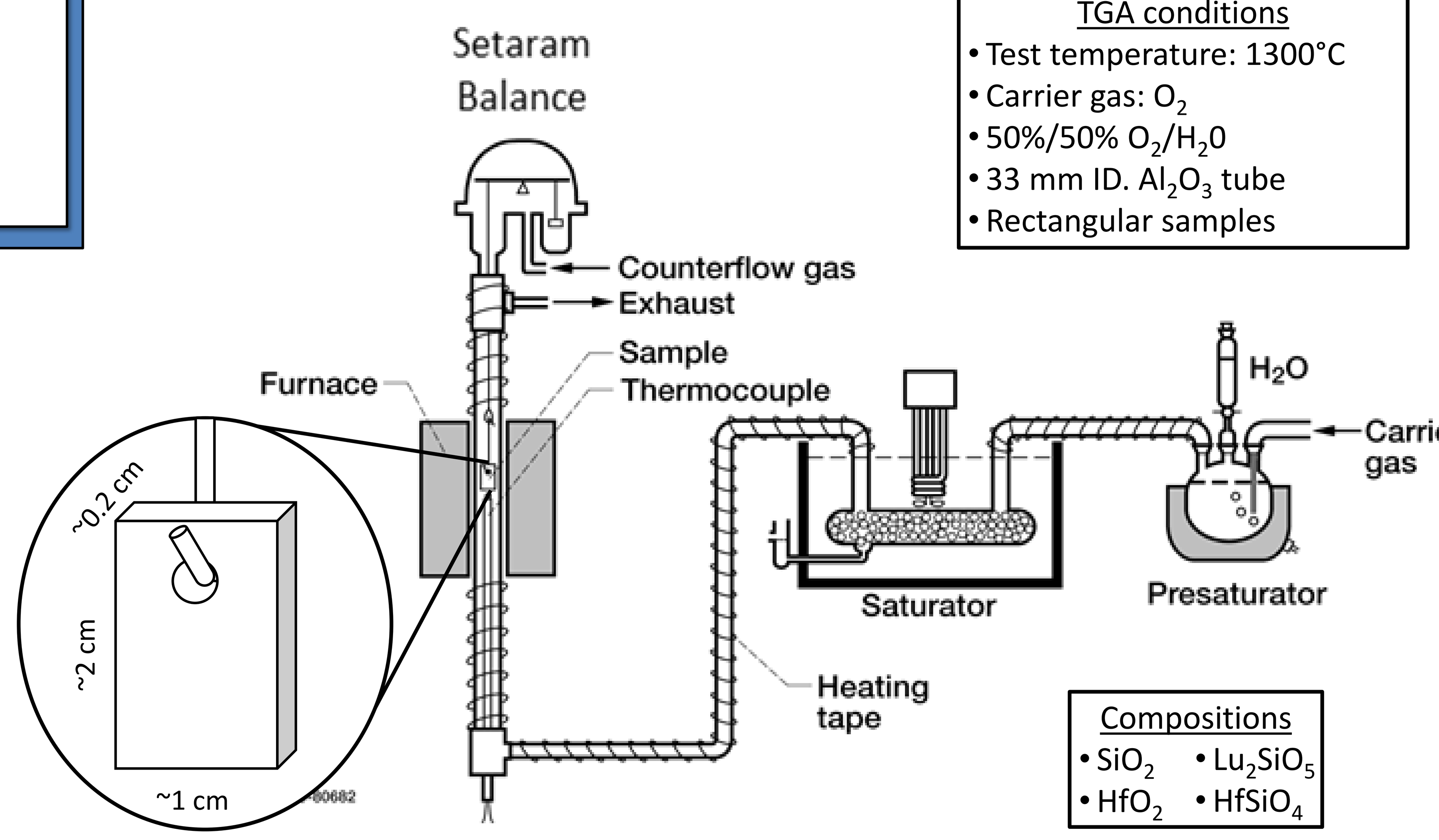


Recession at high temperature due to water vapor[2]

$$SiC + 3/2\,O_2(g) = SiO_2 + CO(g) \quad (1)$$

$$SiO_2 + 2H_2O(g) = Si(OH)_4(g) \quad (2)$$

Experimental setup:[4]



TGA conditions
- Test temperature: 1300°C
- Carrier gas: O_2
- 50%/50% O_2/H_2O
- 33 mm ID. Al_2O_3 tube
- Rectangular samples

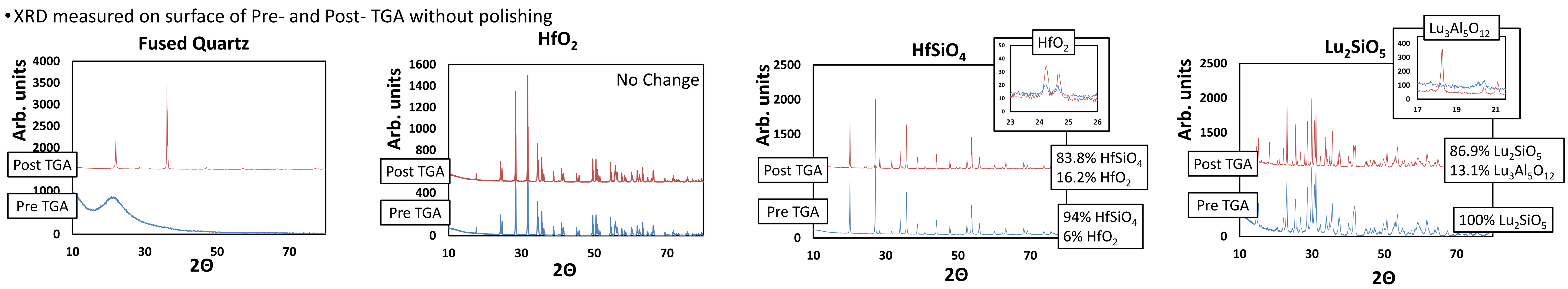
Compositions
- SiO_2
- Lu_2SiO_5
- HfO_2
- $HfSiO_4$

Sample preparation and experimental methodology:

- To replicate water vapor conditions in an engine and measure recession, thermo-gravimetric analysis (TGA) was used under the conditions listed to the right.
- Samples are hot pressed in graphoil between 1500-1600°C.
- Samples are cut into rectangles, polished to remove carbon, and a hole is then drilled for hanging in TGA. If necessary, samples are annealed to remove infiltrated carbon.
- TGA was used to measure the weight change in bulk samples while a 50%/50% mixture of H_2O/O_2 was flowed over the sample.
- Analyses are made with X-ray diffraction and scanning electron microscopy (SEM) to understand how the surface changes after exposure
- The recession is then compared to a model using a simple boundary layer expression programmed into Excel.

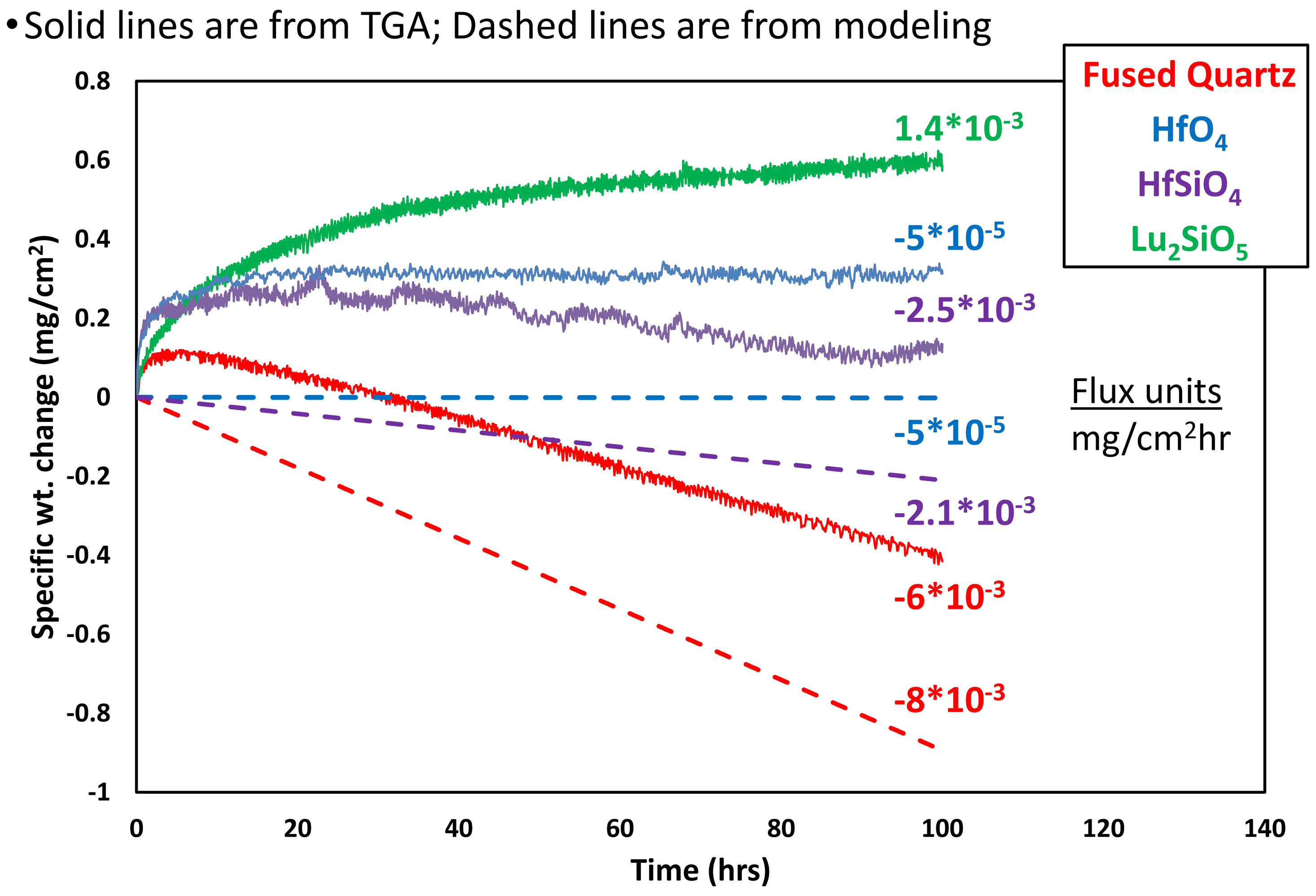
X-ray diffraction

- XRD measured on surface of Pre- and Post- TGA without polishing

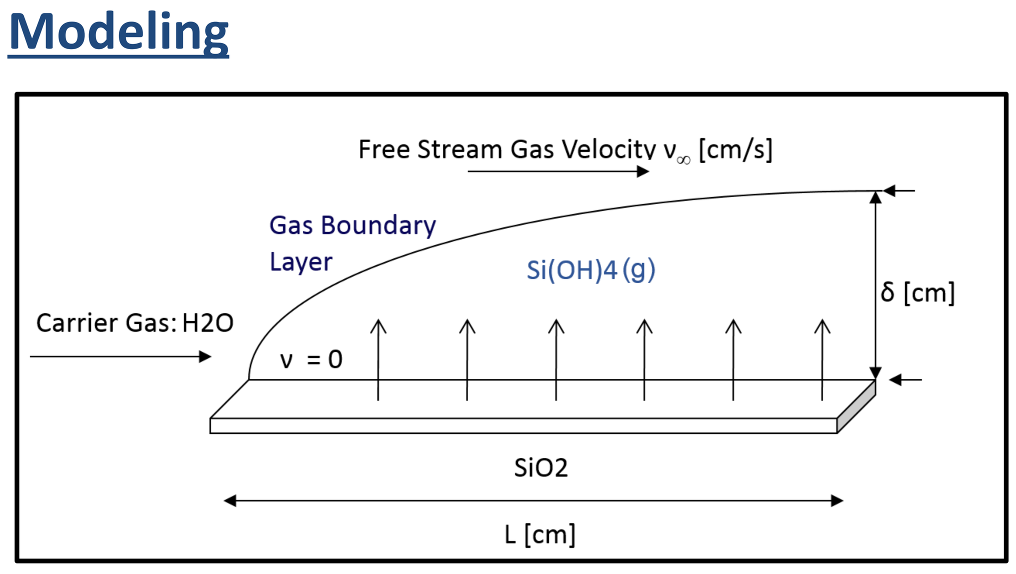


Exposure to water vapor

- Solid lines are from TGA; Dashed lines are from modeling



Modeling



$$\delta = \frac{1.5L}{(Re)^{1/2}(Sc)^{1/3}}$$

Equation for boundary layer thickness, δ [cm]

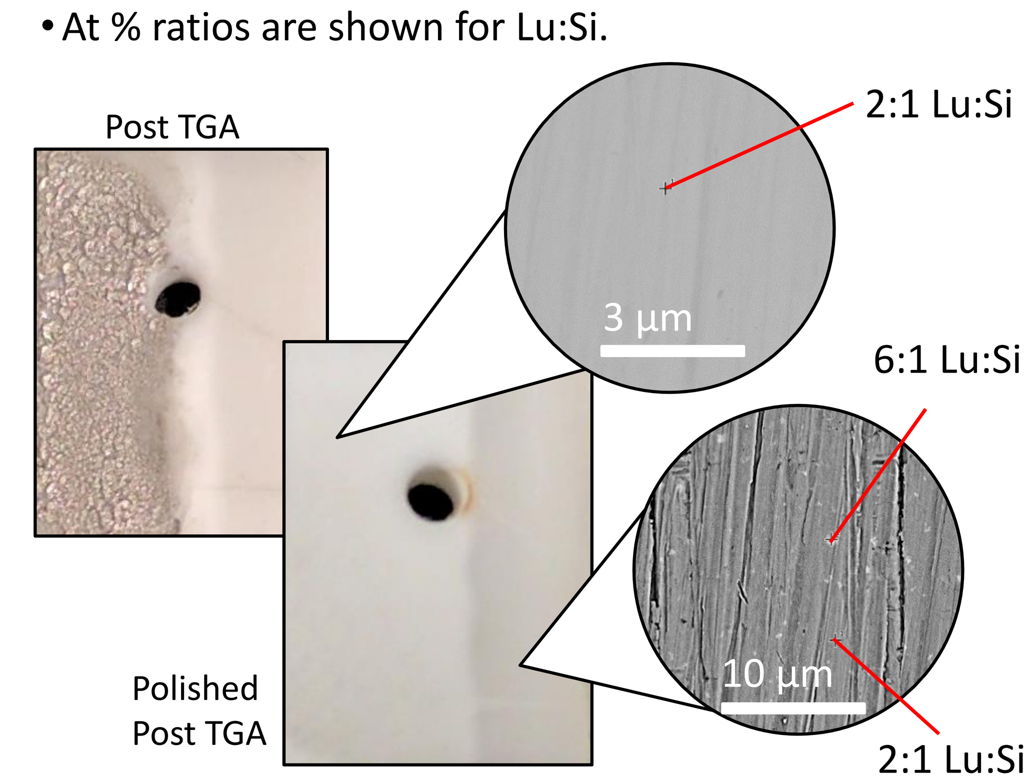
$$J = 0.664\left(\frac{v_\infty \rho_\infty L}{\eta}\right)^{0.5}\left(\frac{\eta}{D_i \rho_\infty}\right)^{0.33}\frac{D_i P_i}{R\,T\,L}$$

Equation for flux, J [mg/cm²hr] from laminar flow

Reynolds number, *Re*	Interdiffusion coefficient, D_i
Schmidts number, *Sc*	Pressure of hydroxide, P_i
Density of gas, ρ_∞	Gas constant, *R*
Viscosity, η	Temperature, *T*

Scanning electron microscopy

- To investigate $Lu_3Al_5O_{12}$ at the surface, the weight change of a half Pt coated sample of Lu_2SiO_5 was measured in TGA.
- SEM images were then taken of a polished surface to determine Al infiltration depth.
- At % ratios are shown for Lu:Si.



Conclusion:

- EBC candidate materials were tested under high temperature, water vapor conditions with a TGA.
- SiO_2 reacts to form $Si(OH)_4$ as expected (Eq. 2). $HfSiO_4$ and HfO_2 are relatively stable in the water vapor environment.
- However, Lu_2SiO_5 exhibits complex behavior due to the presence of Al_2O_3 in the TGA tube.
- Model predicts recession through water vapor, but does not account for surface reactions from contaminants.

References:

1. Opila & Hann, *J. Am. Ceram. Soc.* **80** [1] 197-205 (1997)
2. Opila, Fox & Jacobson, *J. Am. Ceram. Soc.* **80** [4] 1009-12 (1997)
3. Lee, Fox & Bansal, *J. Eur. Ceram. Soc.*, **25** 1705-1715 (2005)
4. Opila, *J. Am. Ceram. Soc.*, **77** [3] 730-736 (1994)