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# Optimization of glass–ceramic sealant compositions in the system $MgO–BaO–SiO_2$ for solid oxide fuel cells (SOFC)☆

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**Abstract**

The glass–ceramic with composition 55 $SiO_2$–27 BaO–18 MgO, mol.% (Mg1.5–55) presents suitable properties for application as a sealant material in IT-SOFC; good unions are achieved with the electrolyte 8YSZ and with the aluminium based interconnect material FeCr alloy. Nevertheless, sealing delaminates when applied to chromium based ferritic stainless steel Crofer 22, one of the most used interconnect material in SOFC, due to the precipitation of $BaCrO_4$ at the interface.

In this work, the union and chemical compatibility of new compositions prepared from modifications of composition Mg1.5–55 have been tested on Crofer 22. The modifications consisted on substituting $SiO_2$ for $B_2O_3$ (Mg1.5–50–5B to Mg1.5–35–20B), for $B_2O_3$ and PbO (Mg1.5–40–5B–10Pb), and $B_2O_3$ and part of MgO for ZnO (Mg1.5–50–5B–8Zn and Mg1.5–40–15B–8Zn). The characterization of the glass–ceramic materials was carried out by dilatometry, differential thermal analysis (DTA), hot-stage microscopy (HSM), X-ray diffraction (XRD) and scanning electron microscopy (SEM).

All the glasses present a suitable thermal expansion coefficient for use in SOFCs. The addition of $B_2O_3$ produces the expected decrease of viscosity and a delay in the crystallisation, which gives a better wettability of the glasses on the steel and a better union, inhibiting the direct reaction between chromium oxide and the barium rich crystalline phases to form $BaCrO_4$. At 850 °C, barium silicates crystallize in all the glasses. Barium magnesium silicates and magnesium silicate are present in the glasses without zinc and the crystallisation of barium zinc silicate is promoted in the zinc containing glasses instead. Compositions Mg1.5–40–15B and Mg1.5–40–15B–8Zn exhibit the best glass–ceramic/Crofer 22 union for the operation temperature of 850 °C. The seals are pore-less, strongly adherent to the steel and present elongated crystals which are homogeneously distributed in the glass matrix.


*Keywords:* Sealing; Glass–ceramic; Interconnect; Crofer 22; SOFC

## 1. Introduction

A key problem in the fabrication of solid oxide fuel cells of planar configuration and intermediate operation temperature (IT-SOFC) is the sealing of the electrolyte or the ceramic anode with the metallic interconnect in order to achieve a hermetic and stable cell.

The sealing material must provide tightness for avoiding the leakage of reactant gases which implicate that a series of thermal, mechanical and chemical requirements must be fulfilled. Additionally, the sealing must present special characteristics referred to electrical conductivity, fabrication, design and application. The glass–ceramic materials offer the best perspectives to satisfy the requirements [1,2].

In previous works, the composition $55SiO_2{\cdot}27BaO{\cdot}18MgO$ (mol.%) (Mg1.5–55) has been developed as a good candidate for sealing SOFC [3–5]. The bonding with the electrolyte yttria stabilised zirconia (8YSZ) and different types of interconnect materials has been studied, particularly the ferritic stainless steel Crofer 22 and the aluminium based alloy FeCr alloy. The bonding with 8YSZ and FeCr alloy is strong and stable [6].

The oxide layer formed in the surface of the steel FeCr alloy, mainly formed by aluminium oxide, reacts during the sealing with the vitreous matrix originating a strong seal with high stability versus thermal cycling. Nevertheless, the composition does not present bonding with the steel Crofer 22 due to the precipitation of $BaCrO_4$ at the interphase, with a coefficient of thermal expansion (CTE) very different to that of the materials to be joined [7]. By the other hand, in reducing atmosphere, the

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