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Influence of Sintering Conditions on Mechanical and Thermal Properties of Cordierite–ZrO_2 Composites

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Abstract

Bend strength, fracture toughness and thermal diffusivity of cordierite composites with 30 vol.% of either unstabilized or Y_2O_3*-stabilized* ZrO_2 *sintered at 1400°C have been determined as a function of sintering time. Depending on sintering time, the phase composition varies from cordierite + monoclinic* ZrO_2 *to spinel + zircon. Cordierite with tetragonal* ZrO_2 *sintered for 1 h exhibits a fracture toughness of* $\sim 4{\cdot}1\ MPa\sqrt{m}$*, while composites consisting essentially of spinel and zircon with both inter- and intragranular monoclinic* ZrO_2 *particles show the highest fracture toughness (* $\sim 4{\cdot}7\ MPa\sqrt{m}$ *). These composites are obtained when cordierite is sintered with unstabilized* ZrO_2 *at 1400°C for >4 h. They also exhibit the highest thermal diffusivity (*$48 \times 10^{-3} cm^2/s$*).*

Biegefestigkeit, Bruchzähigkeit und Temperaturleitfähigkeit von Cordierit mit 30 Vol.% entweder unstabilisiertem oder mit 3 Mol% Y_2O_3*-stabilisiertem* ZrO_2 *wurden als Funktion der Sinterzeit bestimmt. Je nach Sinterzeit bei 1400°C variiert die Phasenzusammensetzung von Cordierit + monoklines* ZrO_2 *bis Zirkon + Spinell. Proben, die 1 h gesintert waren, bestehen aus Cordierit mit tetragonalem* ZrO_2*; sie weisen eine Bruchzähigkeit von* $4{\cdot}1\ MPa\sqrt{m}$ *auf. Die höchste Zähigkeit (*$4{\cdot}7\ MPa\sqrt{m}$*) zeigen Proben, die ursprünglich unstabilisiertes* ZrO_2 *enthalten und die nach >4 h bei 1400°C hauptsächlich aus Spinell und Zirkon mit sowohl inter- wie intragranular dispergierten monoklinen* ZrO_2*-Teilchen bestehen. Diese Proben zeigen auch die höchste Temperaturleitfähigkeit (*$48 \times 10^{-3}\ cm^2/s$*).*

En fonction du temps de frittage, on a déterminé la résistance à la flexion, la ténacité et la diffusibilité thermique de composites à base de cordiérite, contenant 30% en volume de ZrO_2*, soit non stabilisée, soit stabilisée par* Y_2O_3*, et frittés à 1400°C. La composition phasique varie en fonction du temps de frittage, passant de cordiérite +* ZrO_2 *monoclinique à spinelle + zircon. La cordiérite avec de la zircone tétragonale, frittée pendant 1 h, présente une ténacité de* $\simeq 4{\cdot}1\ MPa\sqrt{m}$*, tandis que les composites formés essentiellement de spinelle et zircon, avec des particules de* ZrO_2 *monoclinique inter- et intragranulaires, possèdent la ténacité la plus élevée (* $\simeq 4{\cdot}7\ MPa\sqrt{m}$ *). Ces composés sont obtenus quand la cordiérite est frittée avec de la zircone non stabilisée à 1400°C pendant plus de 4 h. Ils présentent également la diffusivité thermique la plus élevée (*$48 \times 10^{-3}\ cm^2/s$*).*

1 Introduction

The mechanical properties of cordierite ($2MgO.2Al_2O_3.5SiO_2$) can be improved by dispersions of

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either unstabilized (m-ZrO_2) or Y_2O_3-doped ZrO_2 (t-ZrO_2) particles.[1-4] Trace amounts of both zircon and spinel were detected in sintered cordierite–ZrO_2 composites[1,2] which form according to the reaction:[5]

$$5ZrO_2 + Mg_2Al_4Si_5O_{18} = 2MgAl_2O_4 + 5ZrSiO_4 \qquad (1)$$

Recently, it has been demonstrated[6] that in cordierite–m-ZrO_2 composites sintered between 1200 and 1400°C for up to 48 h, ZrO_2 particles react in a cordierite matrix to form zircon and spinel. Similar compositional changes were observed in the cordierite–t-ZrO_2 composites sintered at 1200 and 1300°C. However, when cordierite–t-ZrO_2 composites were sintered at 1400°C, the reaction between cordierite and ZrO_2 particles was severely limited, which is explained by the formation of an Al_2O_3–SiO_2–Y_2O_3 intergranular phase acting as a chemical reaction barrier. Due to the diffusional loss of Y, though, ZrO_2 is increasingly destabilized, leading to transformation to m-symmetry.

The intention of the present work was to study the influence of microstructural changes on mechanical and thermal properties of cordierite–ZrO_2 composites sintered at 1400°C for up to 48 h.

2 Experimental

Samples of cordierite and 30 vol.% of either monoclinic (m) or tetragonal (t) ZrO_2 were prepared by attrition milling, isostatic pressing and sintering. Details of sample preparation are given in Ref. 6. Strength of bars (3·5 × 4·5 × 50 mm) was measured in four-point bending with spans of 30 and 10 mm. Fracture toughness was measured by the indentation strength in bending technique.[7] The indented samples (load 100 N) were broken in three-point bending with a 16 mm span. The tensile surface of the samples was polished to a 1 μm finish. The crosshead speed was 10 μm/min in all cases. Each data point represents an average of 6 measurements.

The fracture surfaces were examined by SEM. Laser flash technique[8] was used to measure thermal diffusivity of the 10 mm diameter by ~1 mm thick disk-shaped specimens. Details of the equipment and procedures have been published elsewhere.[9]

3 Results and Discussion

Strength and fracture toughness of cordierite composites with m-ZrO_2 ('m'-composite) and t-ZrO_2 ('t'-composites) are presented in Figs 1 and 2. The thermal diffusivity for these samples is given in Fig. 3. The sintering time dependencies of these properties are difficult to interpret and to correlate with the respective microstructures which is essentially due to the complicated phase changes taking place in the system.

3.1 Strength and toughness of 'm'-composites

A strong increase in strength and especially toughness is observed for 'm'-composites sintered at 1400°C. Sintering for 8 and 24 h results in a maximum strength of ~285 MPa and maximum fracture toughness of ~4·7 MPa$\sqrt{m}$, respectively. Because of the extensive phase changes taking place at this sintering temperature, the toughness and strength dependency can no longer be related to a specific toughening or strengthening mechanism

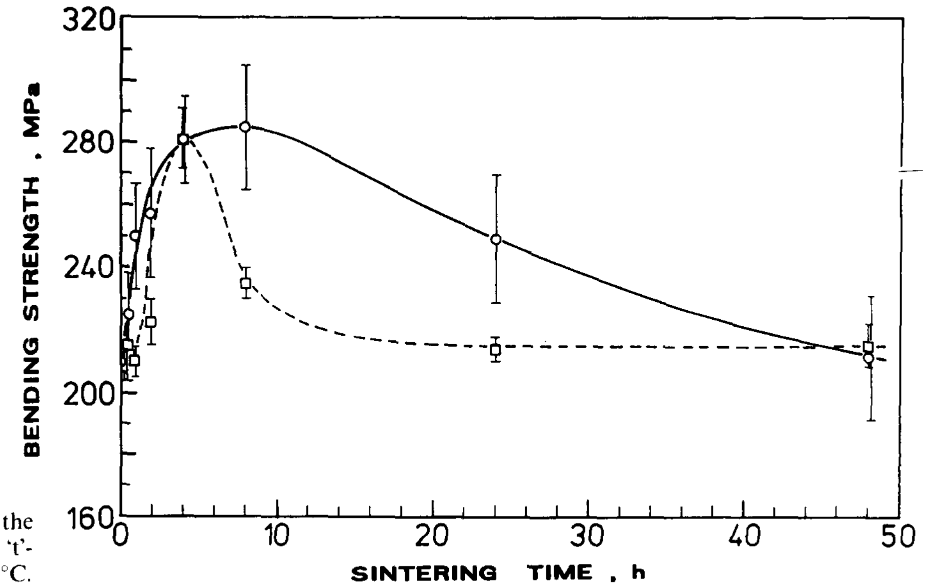

Fig. 1. Effect of the sintering time on the bending strength of (○) 'm'- and (□) 't'-composites. Sintering temperature: 1400°C.

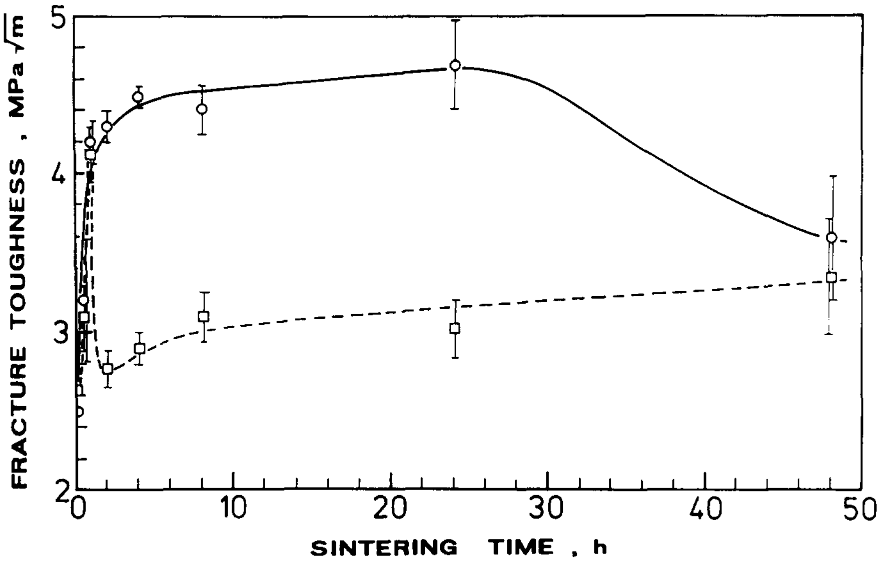

Fig. 2. Effect of the sintering time on the fracture toughness of (○) 'm'- and (□) 't'-composites. Sintering temperature: 1400°C.

prevailing in the cordierite matrix. It can merely be stated that multiphase composites,[6] consisting predominantly of spinel and zircon with both intra- and intergranular m-ZrO_2 particles, exhibit values of toughness far greater than that of the respective single phases.

3.2 Strength and toughness of 't'-composites

The 't'-composites sintered at 1400°C are the only samples where the cordierite matrix remains nearly unchanged except for slight grain growth with time. The fracture toughness shows a maximum at 4·1 MPa$\sqrt{m}$ after sintering for 1 h. The bend strength shows a small maximum at 280 MPa after 4 h where all ZrO_2 particles are still tetragonal. The toughness remains essentially at ≈ 3 MPa$\sqrt{m}$ for all sintering times longer than 1 h. This fact is somewhat surprising, since the phase symmetry of the ZrO_2 particles changes from nearly 100% tetragonal up to 4 h to predominantly monoclinic after 48 h of sintering.

3.4 Thermal diffusivity of 'm'- and 't'-composites

Figure 3 shows the influence of sintering time on thermal diffusivity at room temperature of 'm'- and 't'-composites. Only slight changes in thermal diffusivity with increasing sintering time are observed in 't'-composites sintered for up to 48 h, which is reasonable considering the small compositional changes. The thermal diffusivity of 'm'-composites, however, increases considerably with sintering time, which reflects the respective phase changes, i.e. the relative high thermal diffusivity of spinel becomes obvious. For sintering times >24 h the influence of zircon with its generally lower value of thermal diffusivity begins to dominate (cf. Fig. 4 in Ref. 6).

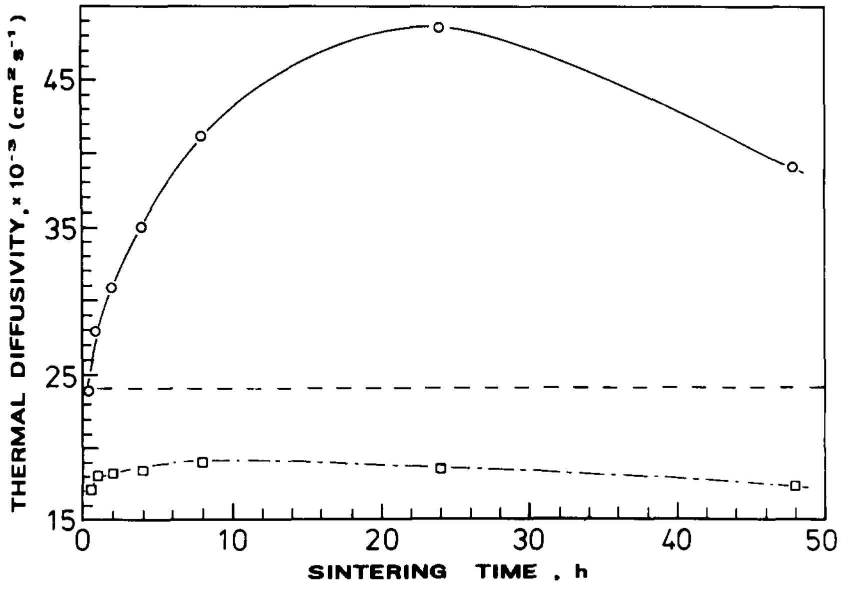

Fig. 3. Effect of the sintering time on the thermal diffusivity at room temperature of (○) 'm'- and (□) 't'-composites compared to (- - -) cordierite. Sintering temperature: 1400°C.

4 Conclusions

The phase changes occurring during sintering of cordierite with 30 vol.% of either monoclinic or Y-stabilized ZrO_2 strongly affect the mechanical properties and the thermal diffusivity. In all cases, both strength and toughness are considerably higher than that of the cordierite matrix alone (120 MPa and $1{\cdot}5\,MPa\sqrt{m}$, respectively).[2] Maximum strength of ~280 MPa is exhibited by composites with tetragonal and monoclinic ZrO_2 sintered for 4 and 8 h, respectively. Maximum fracture toughness of $4{\cdot}7\,MPa\sqrt{m}$ is attained by composites, which originally contain unstabilized ZrO_2, sintered for >4 h. These composites consist essentially of the reaction products zircon and spinel with both inter- and intragranular m-ZrO_2 particles. These 'm'-composites also show the highest thermal diffusivity. Both parameters are prerequisites for excellent thermal shock properties.

Acknowledgement

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