Y-TZP ceramics with tailored toughness

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Abstract. Despite the considerable improvement in the understanding of transformation toughening accomplished in the last three decades, it remains an important challenge to be able to control and tune the tetragonal ZrO_2 phase transformability and associated toughness of polycrystalline tetragonal zirconia (Y-TZP) ceramics. The problem of controlling the toughness of Y-TZP is investigated in the present paper by comparing the mechanical behaviour of a number of ceramics derived from commercial co-precipitated and yttria-coated zirconia starting powders as well as a range of experimental powder mixtures, obtained by mixing monoclinic and 3 mol % yttria-stabilised powders. Based on the experimental results, a simple route to tailor the toughness of Y-TZP ceramics is reported. The effectiveness of this approach was investigated and the microstructural origin influencing transformation toughening is elucidated. The difference in toughness is explained in terms of the tetragonal grain size and the overall amount and distribution of yttria in the sintered ceramics. The overall yttria stabiliser content is of primary importance, the yttria distribution however was found to be an additional important microstructural variable influencing the transformation toughness of the investigated Y-TZP ceramics.

Introduction

Toughness control and optimisation is considered as one of the key criteria in the development of Y-TZP ceramics for structural applications. To this end, the powder processing route plays a major role. At present, both co-precipitated and yttria-coated powders are commercially exploited in the fabrication of TZP ceramics [1]. In recent years, more attention is given to Y_2O_3-coated zirconia starting powders for the production of TZPs [2]. The material properties of Y-TZP ceramics obtained from yttria-coated zirconia starting powders are reported to be more attractive than those of the ceramics based on co-precipitated powders, sintered under the same conditions. However, yttria-coated zirconia powders are no longer commercially available. In this paper, a novel route to engineer the toughness of Y-TZP ceramics by means of "powder mixing" is described.

Experimental Procedure

The commercial zirconia starting powders include 2.8 mol % yttria-coated (Tioxide grade YZ5N (Tio3)), 3 mol % yttria co-precipitated (Tosoh grade TZ-3Y (T3)) and 2 mol % yttria co-precipitated powder (Tosoh grade TZ-2Y (T2)). Co-precipitated 3Y-TZP powder (T3) was mixed with Y_2O_3-free monoclinic powder (Tosoh grade TZ-0) in order to obtain powder mixtures with an overall Y_2O_3-content of 1.75 (TM1.75), 2 (TM2) and 2.5 mol % (TM2.5). The powders were mixed on a multidirectional mixer for 24 hours in propanol, using alumina milling cylinders to break the agglomerates. The densification of the ceramic samples was accomplished either by hot pressing in vacuum (1450°C, 1 h) or by pressureless sintering of cold isostatically pressed (3 minutes at 300 MPa) powder compacts in air (1450°C, 2 h).

The Vickers hardness (HV_{10}) was measured on a Zwick hardness tester with an indentation load of 10 kg. The fracture toughness (K_{Ic}) was calculated from the measured radial cracks around the

HV indentations, according to the formula of Anstis et al. [3]. The reported H_V and K_{IC} values are the mean of at least 5 indentations. The E-modulus of the ceramic specimens was measured by the resonance frequency method using a Grindo-Sonic. The density was measured in ethanol. Microstructural investigation of the thermally etched specimens was performed using SEM, XRD and electron microprobe analysis (EPMA). From the digitised SEM micrographs of thermally etched samples, some 1150-1400 grains were counted. The grain size distribution was fitted with the Extreme Type I (Gumble) function according to the model described elsewhere [4].

Results and Discussion

The mechanical properties of the hot pressed ceramics are summarised in Table 1. SEM observations of polished surfaces revealed that the obtained ceramics are fully dense, except the TM1.75 ceramic that shows cracks. XRD analysis of the polished surfaces revealed the predominant presence of the t-ZrO_2 phase in all samples. A very small amount (2.7 %) of m-ZrO_2 was found in the powder mixture grade TM2, whereas a relatively large amount (40.3 %) of m-ZrO_2 is observed in the T2 ceramic.

The mechanical properties, listed in Table 1, reveal that the toughness of the powder mixture grades increases with decreasing overall yttria content down to 2 mol %. At lower yttria levels, the toughness is strongly decreased. The toughness of the experimental TM2 ceramic is much higher than that of the co-precipitated T2 ceramic and even better than that of the yttria-coated Tio3 ceramic. At an overall yttria content below 1.75 mol %, the fracture toughness, hardness as well as density are decreased, due to spontaneous transformation of t-ZrO_2 grains during cooling from the sintering temperature.

The experimental results reveal that the overall yttria content needs to stay above 1.75 mol %, when tailoring the toughness of Y-TZP by the addition of yttria-free ZrO_2 powders to 3 mol % yttria co-precipitated powders.

It should be pointed out that the toughness ranking of the pressureless sintered Y-TZP grades is the same as for the hot pressed samples, demonstrating the effectiveness of the powder mixture approach under normal sintering conditions for oxide ceramics.

It is well recognised that stress induced transformation is the predominant toughening mechanism in Y-TZP ceramics [5]. The physical modifications in the microstructure and their impact on the mechanical properties of Y-TZP ceramics prepared by the powder mixture approach are identified and elucidated.

Ceramic	HV_{10} [GPa]	E [GPa]	ρ [g/cm^3]	grain size [µm]	K_{IC10} [MPa m$^{1/2}$]
Tio3	12.1 ± 0.2	204	6.07	0.199	8.7 ± 0.3
T3	11.9 ± 0.2	194	6.08	0.311	2.5 ± 0.1
TM2.5	12.6 ± 0.1	203	6.05	0.350	5.7 ± 0.1
TM2	11.9 ± 0.1	215	6.07	0.491	10.3 ± 0.5
T2	10.9 ± 0.4	215	6.06	0.429	5.9 ± 0.1
TM1.75	9.6 ± 0.1	175	5.83	0.724	3.7 ± 0.2

Table 1. Properties of the Y-TZP ceramics, hot pressed at 1450°C for 1 h in vacuum.

It is well reported that the transformation toughening contribution to the total toughness of Y-TZP ceramics exhibits a substantial grain size dependence [6]. The fracture toughness of co-precipitated powder based as well as powder mixture based grades increases with increasing mean grain size (see Table 1). When the increase in fracture toughness is solely related to an increased grain size, below a critical maximum grain size [7], the differences in toughness can be attributed to a reduced yttria content, either generated in the co-precipitation process (T2 versus T3) or established by powder mixing (TM2, TM2.5 versus T3). Moreover, the addition of m-ZrO_2 to a 3

mol % Y_2O_3 co-precipitated powder appears to be an efficient way to increase the grain size, resulting in an increased fracture toughness.

Literature data [8] show that the fracture toughness for a co-precipitated powder based 2Y-TZP ceramic with an average grain size around 0.5 μm is about 6 MPa $m^{1/2}$, which is in agreement with the value obtained for the T2 ceramic. This value however is well below the excellent fracture toughness of 10 MPa $m^{1/2}$ obtained for the TM2 ceramic. This observation illustrates that the mechanical behaviour of the powder mixture grades is not in agreement with the common experience on co-precipitated powder based Y-TZP.

Furthermore, a high transformation toughness of 9 MPa $m^{1/2}$ is obtained for the Tio3 ceramic at a finer tetragonal grain size of 0.19 μm, a grain size at which co-precipitated t-ZrO_2 grains are very stable and hardly susceptible to transformation.

On the basis of the above observations, it can be concluded that the excellent fracture toughness of the yttria-coated and powder mixture based ceramics can not only be explained by the grain size effect. Therefore, other microstructural variables need to be considered.

EPMA point analysis revealed a broad and symmetric yttria distribution around 3 mol % yttria in the yttria-coated Tio3 ceramic, with a significant amount of t-ZrO_2 grains having a low yttria content (1-3 mol %). These low yttria-content grains should be considered as very susceptible to transformation, since the transformability increases with decreasing yttria content. TEM observations revealed a core-shell microstructure in the t-ZrO_2 grains of yttria-coated powder based Y-TZP [9]. It is also suggested in literature that the enhanced transformability of the core-shell microstructure is responsible for the high fracture toughness of coated Y-TZP [10].

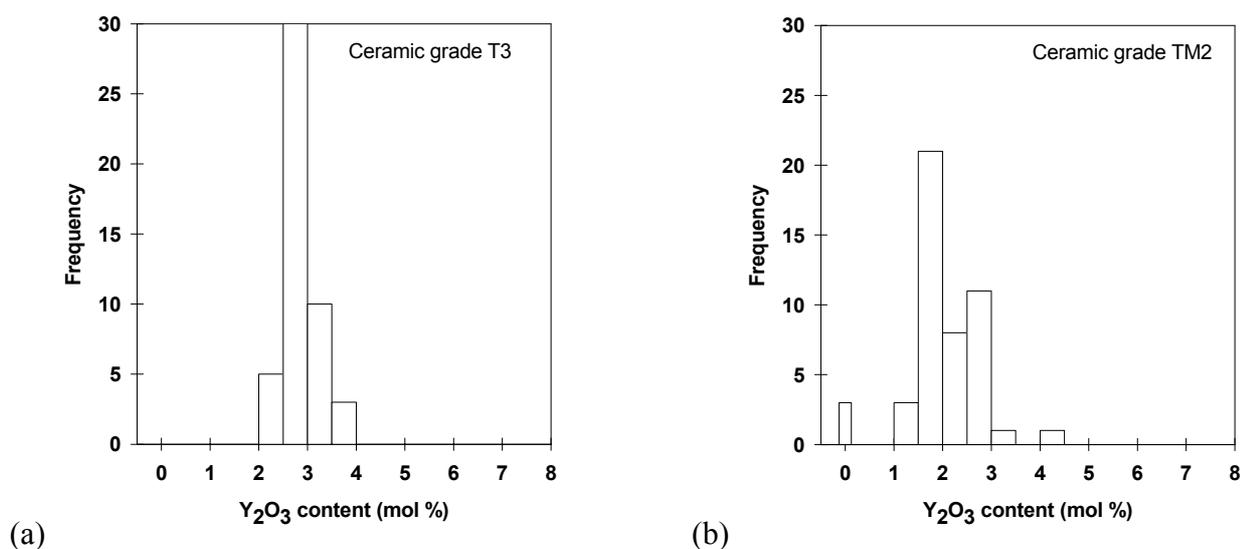(a)   (b)

Fig. 1. Yttria distributions in the hot pressed 3 mol % yttria co-precipitated powder T3 (a) and the TM2 powder mixture (b) based Y-TZP, as obtained by EPMA.

On the other hand, the yttria is homogeneously distributed in the hot pressed 3Y-TZP ceramic, as revealed by the narrow yttria distribution around 3 mol % yttria shown in Fig. 1.a. The yttria distribution of the TM2 ceramic, shown in Fig. 1.b, reveals a major frequency at 2 mol % yttria. However, many analysis results show a deviation in yttria content, including a significant number at 0 mol %. This indicating an incomplete conversion of the two starting powders (TZ-3Y and TZ-0) into new tetragonal grains with an uniform yttria content of 2 mol %. The relatively small amount of low yttria content grains can be considered as extremely sensitive to stress induced transformation.

Differences in yttria content between neighbouring grains should also enhance the local residual stress due to the anisotropy in the coefficient of thermal expansion, CTE, along the c- and a-axis of the tetragonal zirconia phase (α_c/α_a) [11,12]. Based on X-ray precision lattice parameter

measurements and finite element calculations, a large difference in the CTE anisotropy between 3Y-TZP (15 %) and 2Y-TZP (32 %) processed from homogeneous powders was observed [12]. This will lower the critical tensile stress needed to initiate the transformation, which is an important parameter in the description of stress induced martensitic transformation [5]. Once a critical tensile stress is generated in these grains during crack propagation, they will immediately transform and subsequently induce tetragonal phase transformation in the neighbouring grains in an autocatalytic manner. This will eventually result in the formation of a larger transformation zone. This kind of autocatalytic transformation is a characteristic feature of the zirconia martensitic phase transformation [13].

Two major mechanisms for enhanced fracture toughness can be considered for the powder mixture based Y-TZP ceramics. The first one is due to grain growth, originating from a reduced yttria content in the originally co-precipitated grain and the second one is due to the formation of yttria gradients in the material. Based on the above discussion, it is clear that the contribution of the second mechanism is predominant in the powder mixed grades.

Summary

The most important outcome of the present investigation is that the fracture toughness of Y-TZP can be tailored by the addition of $m\text{-}ZrO_2$ particles to 3 mol % yttria co-precipitated powder.

An excellent toughness of 10 MPa m$^{1/2}$, in combination with a fine $t\text{-}ZrO_2$ mean grain size of 0.5 μm was achieved after hot pressing for 1 hour at 1450°C in vacuum, with powder mixture grades having an overall yttria content of 2 mol %.

The grain growth, observed in the powder mixture grades, is too limited to account for the observed increase in toughness. The additional microstructural parameter contributing to an increased toughness was identified to be the inhomogeneous yttria distribution, a factor that also accounts for the excellent toughness that can be obtained with yttria-coated ZrO_2 powders.

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