Mechanical properties of 2.0–3.5 mol% \( Y_2O_3 \)-stabilized zirconia polycrystals fabricated by the solid phase mixing and sintering method

Hiroshi OHNISHI, Hironori NAKA, Tohru SEKINO, Yuichi IKUHARA and Koichi NIHARA

NIKKATO CORPORATION, 3–2–24, Oriono-cho, Sakai-ku, Sakai, Osaka 590–0001
1Institute of Multidisciplinary Research for Advanced Materials (IMRAM), Tohoku University, 2–1–1, Katahira, Aoba-ku, Sendai 980–8577
2Institute of Engineering Innovation, School of Engineering, The University of Tokyo, 2–11–16, Yayoi, Bunkyo-ku, Tokyo 113–8656
3Extreme Energy-Density Research Institute, Nagaoka University of Technology, 1603–1, Kamitomioka-machi, Nagaoka-shi 940–2188

The crystal phase and mechanical properties of 2.0 to 3.5 mol% \( Y_2O_3 \)-stabilized zirconia polycrystals sintered from agitator milled powders (solid phase mixing, SPM) were investigated. Powders containing 2.5 and 2.8 mol% \( Y_2O_3 \) showed higher sinterability than those containing 2.0 and 3.5 mol% \( Y_2O_3 \). The thermal expansion curve of the 2.5 and 2.8 mol% \( Y_2O_3 \) samples exhibited a monoclinic to tetragonal phase transformation during the heating stage, with a linear relationship observed during cooling. However, a hysteresis was not observed during the heating and cooling stages when the \( Y_2O_3 \) content and sintering temperature were increased. The 2.0 mol% \( Y_2O_3 \) sample exhibited the transformation and hysteresis, even though the sintering temperature was increased. The relationship between the flexural strength and grain size for the sample with 2.0 mol% \( Y_2O_3 \) was not linear, but exhibited a peak. The flexural strength of the 2.5 and 2.8 mol% \( Y_2O_3 \) samples increased as the grain size increased, and then over a certain grain size the flexural strength remained constant and independent of the grain size. The flexural strength and fracture toughness of the 2.0 mol% \( Y_2O_3 \) sample might be dominated by microcrack-toughening. On the other hand, those for the 2.5 and 2.8 mol% \( Y_2O_3 \) samples were dependent not only on the stress-induced transformation, but also on the compressive residual stress and microcrack-toughening caused by the transformation. The relationship between the fracture toughness and transformed depth of the SPM sample was non-linear and exhibited a peak; however, that for the 2.8 mol% \( Y_2O_3 \) sample was linear.

©2008 The Ceramic Society of Japan. All rights reserved.

Key-words : Solid phase method, Flexural strength, Fracture toughness, Compressive residual stress, Microcrack-toughening, Stress-induced transformation, Transformed depth

1. Introduction

The sinterability and microstructure of zirconia prepared by wet dispersing and mixing technique using an agitator mill (solid phase mixing, SPM) were previously investigated by the present authors and compared with samples prepared by the liquid phase precipitation method (LPM).1,2 2.8 mol% \( Y_2O_3 \)-stabilized \( ZrO_2 \) polycrystals prepared by SPM had higher sinterability and finer grains than those prepared by LPM, because both the stabilization of \( ZrO_2 \) by \( Y_2O_3 \) and diffusion proceed during the sintering process. No monoclinic phase was observed in the LPM sample. For the SPM sample, the monoclinic phase was detected by X-ray diffraction (XRD); however, it was concluded on the basis of the thermal expansion curve that the monoclinic phase was essentially absent. As the distribution of \( Y^{3+} \) ions in \( ZrO_2 \) grains was more inhomogeneous for the SPM sample than for the LPM sample, and varied with sintering temperature, the microstructure and crystal phase of the SPM sample were considerably different from those of the LPM sample.1,3

The mechanical properties of these SPM and LPM samples were also investigated.1 In the case of the SPM sample, not only stress-induced transformation toughening, but also compressive residual stress and microcrack-toughening contributed to the fracture toughness. The fracture toughness of the SPM sample was higher than that for the LPM sample, reaching a maximum value of 9.5 MPa \( \sqrt{m} \). The toughening effect occurring in the SPM sample differed from that of the LPM sample, because the microstructure and crystal phases of the samples were considerably different. Although the grain size of the SPM sample was smaller than that of the LPM sample, the transformed monoclinic phase fraction of the SPM sample was higher than that of the LPM sample. The fracture toughness of LPM sample is mainly influenced by stress-induced transformation,3,4 which is increased with the grain size increased; the fracture toughness of SPM sample is dominated by the effect of superposition for each toughening effect. It is, therefore, very interesting that the contribution of the toughening effects, such as stress induced transformation characteristics and microcracking, on the fracture toughness were varied depending on the grain size for the present zirconia ceramics.5 Generally it is known that toughening mechanisms are fixed according to the grain size.

Tsukuma et al.5 investigated the mechanical properties of yttria-stabilized tetragonal zirconia polycrystals (Y-TZP) containing 2–4 mol% \( Y_2O_3 \) prepared by LPM; however, those of Y-
TZP prepared by SPM have been scarcely investigated. It is expected that the mechanical properties of SPM samples would show considerable differences from those of the LPM sample, because of the different toughening effects caused by the microstructure and crystal phases. A detailed investigation of the mechanical properties of samples with varying Y2O3 content prepared by SPM may provide the possibility of further improvement of the fracture toughness. Therefore, the purpose of this investigation is to clarify the relationships between transformation, grain size and the mechanical properties of SPM samples in comparison to LPM samples.

2. Experimental procedure

ZrO2 powders were prepared by SPM of ZrO2 powder (> 99.9%; surface area 10 m2/g) manufactured by the hydrolysis method with Y2O3 (> 99.9%; surface area 12 m2/g) and Al2O3 (> 99.9%; surface area 7 m2/g) powders. The powders were mixed in proportion to contain 2.0–3.5 mol% Y2O3 and 0.25 wt% Al2O3. Sintering was performed at 1250 to 1450°C for 2 h in air. The powder preparation, pressing and sintering conditions have been previously reported. Table 1 shows the chemical composition of each sample with 2.0, 2.5, 2.8 and 3.5 mol% Y2O3.

The density of the sintered samples was measured by the Archimedes method using distilled water. The phase compositions of the ground, polished and fractured surfaces were analyzed using XRD. The average grain size was calculated from scanning electron microscope (SEM; Model S–800, Hitachi, Ltd., Tokyo) images using the intercept method. Thermal expansion behavior was measured using a dilatometer (Thermo Plus 2 TMA8310, Rigaku Corporation, Osaka) at a constant heating and cooling rate of 2°C/min up to 1200°C, with 99.5 wt% Al2O3 used as a standard sample.

The residual stress on the surface was measured by XRD analysis. The flexural strength was evaluated using a 3-point bending test, and the fracture toughness was measured by the single etched notched beam method (SENB) for three-point bending. Details regarding the measurement methods and conditions for each property have been previously reported.1,2

3. Results and discussion

3.1 Density and grain size

The density and average grain sizes of the 2.0, 2.5, 2.8 and 3.5 mol% Y2O3 samples are shown in Fig. 1 as a function of the sintering temperature. Figure 2 shows typical SEM micrographs of each sample sintered at 1300°C. Density of over 6.0 g/cm³ was achieved for the 2.5, 2.8 and 3.5 mol% Y2O3 samples; however, the density of the 2.0 mol% Y2O3 sample was less than 5.9 g/cm³, even though the sintering temperature was increased to 1450°C. It is probably that the 2.0 mol% Y2O3 sample contained a larger monoclinic phase fraction than the other samples, as discussed later.

The grain size was increased with the decrease in Y2O3 content for the same sintering temperature. Since the content of Y3+ ions in the ZrO2 grain interior increased and stabilization proceeded during sintering with increased Y2O3 content and sintering temperature,11 grain growth might be prevented by an increase in the Y2O3 content, especially at lower sintering temperature. The relationship between the grain size and the sintering temperature was observed, and the sintering temperature dependence of the grain size became stronger with an increase in the Y2O3 content, because the tetragonal and cubic phase frac-
tion increased, due to the progress of stabilization at lower sintering temperature.

### 3.2 Crystalline phase

XRD patterns of polished surfaces of 2.0 and 3.5 mol% Y₂O₃ samples sintered at 1300 and 1400°C are shown in Fig. 3. Also, detailed diffraction patterns ranging from 72 to 75.5° are enlarged and shown in Fig. 4 for all samples. Neither crystalline phase except zirconia nor impurity phase was detected. Diffraction patterns for the tetragonal and cubic phase of each sample were clarified with an increase in the sintering temperature, and the intensity of the cubic phase diffraction peaks increased by the further progress of stabilization through sintering as Y₂O₃ content increased.

The relationship between the monoclinic, tetragonal and cubic phase fractions on the ground and polished surfaces and the grain size for each sample sintered at 1275–1440°C is shown in Fig. 5. The monoclinic phase fraction on these surfaces decreased with the increase in the grain size and Y₂O₃ content.

The monoclinic phase fraction of the 2.0, 2.5 and 2.8 mol% Y₂O₃ samples showed a tendency to decrease as the grain size increased, and the tetragonal phase fraction increased simultaneously. In the case of the 2.5 and 2.8 mol% Y₂O₃ samples, a drastic decrease in the monoclinic and cubic phase fractions and an increase in the tetragonal phase fraction were observed. While, for the 2.0 mol% Y₂O₃ sample, the decrease in the monoclinic phase fraction and increase in the tetragonal phase fraction were less pronounced than that for the 2.5 and 2.8 mol% Y₂O₃ samples, and the cubic phase was not detected by XRD, because the amount of stabilizer is too small to form the cubic phase.

The dependence of the monoclinic phase fraction on the grain size for the 3.5 mol% Y₂O₃ sample was almost the same as that for the other samples. However, the tetragonal phase fraction was not dependent on the grain size and remained almost constant, while the cubic phase fraction increased.

It was reported that in conventional Y-TZP prepared by LPM, the monoclinic phase fraction on a ground surface, which is transformed by grinding stresses, increases as the grain size increases, because the tetragonal phase is easily transformed to the monoclinic phase when the grain size is increased. On the contrary to the LPM sample, the monoclinic phase fraction on the ground surface in the SPM sample decreased in inverse proportion to the grain size. The monoclinic phase fraction of the SPM samples probably depended on the progress of stabilization of ZrO₂ by Y₂O₃, rather than the acceleration of the transformation by grinding stresses. Accordingly, it is reasonably considered that the stress induced tetragonal to monoclinic phase transformation becomes to be difficult due to the progress of sta-

---

**Fig. 3.** XRD patterns of polished surfaces for the samples with 2.0, 2.5, 2.8 and 3.5 mol% Y₂O₃ sintered at 1300 and 1400°C.

**Fig. 4.** XRD patterns of polished surfaces for the samples with 2.0, 2.5, 2.8 and 3.5 mol% Y₂O₃ sintered at 1300 and 1400°C.

**Fig. 5.** Relationship between the monoclinic, tetragonal and cubic phase fractions and the grain size of samples with 2.0, 2.5, 2.8 and 3.5 mol% Y₂O₃.
bilization by the increase in Y$_2$O$_3$ content and the corresponding increase in grain size. This phenomenon might also contribute to the fraction of transformed monoclinic phase by surface grinding. However, the monoclinic phase fraction was still detected on the polished surface by XRD, even though the grain size increased. This suggests that the transformed zone created by grinding stresses was deep and could not be removed even by polishing, as discussed later.

### 3.3 Thermal expansion

The thermal expansion curves of the 2.0 mol% Y$_2$O$_3$ sample sintered at 1300 and 1440°C, and the 2.5 and 2.8 mol% Y$_2$O$_3$ samples sintered at 1275, 1300 and 1440°C are shown in Fig. 6. The monoclinic to tetragonal phase transformation during the heating stage, and the tetragonal to monoclinic phase transformation during the cooling stage were observed in the 2.0 mol% Y$_2$O$_3$ sample, as with conventional partially stabilized zirconia, regardless of the sintering temperature. This expansion behavior indicates that the 2.0 mol% Y$_2$O$_3$ sample probably contains the monoclinic phase. The martensitic temperature (A$_m$), at which the monoclinic to tetragonal phase transformation is observed, decreased with increasing sintering temperature. It is suspected that the stabilization of ZrO$_2$ by Y$_2$O$_3$ progressed and ZrO$_2$ was more stabilized with the increased sintering temperature and the resulting in decrease in the A$_m$ temperature. The monoclinic to tetragonal phase transformation was observed during the heating stage for the 2.5 and 2.8 mol% Y$_2$O$_3$ samples sintered at 1275°C and the 2.5 mol% Y$_2$O$_3$ sample sintered at 1300°C, similar to that observed for the 2.0 mol% Y$_2$O$_3$ sample. However, a linear expansion curve was observed during the cooling stage, in contrast to the 2.0 mol% Y$_2$O$_3$ sample.

The samples for thermal expansion measurements were prepared by grinding under the same conditions as that for the flexural strength measurement, so that the transformed monoclinic phase caused by grinding stresses was present in the surface layer of the samples. The transformed monoclinic phase in the surface layer would affect the thermal expansion behavior, because the sample size (i.e. 3 × 3 × 20 mm) was very small for thermal expansion measurements. Therefore, this expansion behavior is estimated as follows. The monoclinic to tetragonal phase transformation in the surface layer during the heating stage appears in the thermal expansion curve when the transformed monoclinic phase fraction is large and/or the transformed zone is deep. The 2.5 mol% Y$_2$O$_3$ sample sintered at 1440°C and the 2.8 mol% Y$_2$O$_3$ samples sintered at 1300 and 1440°C displayed a linear expansion curve during both the heating and cooling stages. The sintering temperature, at which this linear expansion behavior was observed, decreased with the increase in Y$_2$O$_3$ content. In the sample sintered at lower temperature, insufficiently stabilized regions by Y$_2$O$_3$ might exist in some ZrO$_2$ grains as reported previously. Since these regions might be easily transformed by lower grinding stresses, the transformed zone should be deep. As a consequence, the transformed monoclinic by grinding stresses appeared during the heating stage. On the other hand, it seems that the thermal expansion curve of the sample sintered at higher temperature is linear, because of the progress of stabilization with increasing sintering temperature and/or the Y$_2$O$_3$ content. This is demonstrated by the decrease in the A$_m$ temperature of the 2.0 mol% Y$_2$O$_3$ sample as the sintering temperature increased.

As the result, it is suggested that for the samples with Y$_2$O$_3$ contents over 2.5 mol%, the monoclinic phase is essentially absent, or if present, it is too small a quantity to affect the thermal expansion behavior.

### 3.4 Relationship between monoclinic phase of the fracture surface and residual stress

#### 3.4.1 Monoclinic phase on the fracture surface

The relationship between the residual stress and transformed monoclinic phase fraction on the fracture surface for the 2.0, 2.5, 2.8 and 3.5 mol% Y$_2$O$_3$ samples are plotted as a function of grain size in Fig. 7.

The monoclinic phase found on the fracture surface for the 2.5, 2.8 and 3.5 mol% Y$_2$O$_3$ samples was regarded as a transformed monoclinic phase, because the these samples did not contain the monoclinic phase before fracture. In contrast, for the 2.0 mol% Y$_2$O$_3$ sample, the monoclinic phase was confirmed after sintering as mentioned in relation to the thermal expansion behavior. Therefore, the transformed monoclinic phase fraction was calculated by subtracting the monoclinic phase fraction observed on the polished surface from that measured on the fracture surface.

The transformed monoclinic phase fraction increased proportionally to the grain size for the 2.0 mol% Y$_2$O$_3$ sample, as for the previously reported Y-TZP prepared by LPM. However, the mechanism of the increase of the transformed monoclinic phase against the grain size seems to be different between LPM and SPM samples. In the case of the LPM sample, solid soluted Y$_2$O$_3$ concentration was decreased when grain size increased, and then the tetragonal grains tended to transform easily. While, the transformed monoclinic phase fraction of 2.0 mol% Y$_2$O$_3$
sample increased as the grain size increased (see Fig. 7). This suggests that Y₂O₃ concentration solid soluted into ZrO₂ grains increased as the grain size increased and the stabilization progressed, and resulting in the increase of critical stress for stress-induced transformation. Because of this stabilization of the tetragonal grains, monoclinic phase fraction between polished and ground surface for the 2.0 mol% sample was almost the same (see Fig. 5), which indicated that the grinding stress was lower than the critical stress for the transformation. On the contrary, these grains in 2.0 mol% Y₂O₃ sample presumably transformed by higher stress during fracture, and hence the transformed monoclinic phase in the fractured surface was increased according to the grain growth.

In contrast, the transformed monoclinic phase fraction of the 2.5 and 2.8 mol% Y₂O₃ samples was almost constant regardless of the variation in grain size. For the 3.5 mol% Y₂O₃ sample, the decrease of the transformed monoclinic phase fraction was inversely proportional to the grain size. The transformed monoclinic phase fraction and grain size for the LPM sample was proportional relation in any compositions. For all SPM samples was not linear, and exhibited a peak. The flexural strength of the 2.5 and 2.8 mol% Y₂O₃ samples increased up to a certain grain size, and was independent on the compressive residual stress. The relationship between the flexural strength and grain size over a certain grain size and remained constant. The relationship between the compressive residual stress and grain size of the 2.5 and 2.8 mol% Y₂O₃ samples showed the same behavior. The flexural strength of the 2.5 and 2.8 mol% Y₂O₃ samples increased with a decrease in the compressive residual stress over the grain size showing the maximum compressive residual stress. That of the 2.5 and 2.8 mol% Y₂O₃ samples increased up to a certain grain size, and was dependent on the grain size over a certain grain size and remained constant. However, the flexural strength of 2.5 and 2.8 mol% Y₂O₃ samples with smallest grain size was low despite of low compressive residual stress.

The relationship between the flexural strength and the transformed monoclinic phase fraction for the 2.0, 2.5 and 2.8 mol% Y₂O₃ samples could not observed, on the other hand, the flexural strength of the 3.5 mol% Y₂O₃ sample increased with decreasing the transformed monoclinic phase fraction in contrast of the LPM sample.²⁻³

### 3.5.2 Fracture toughness
The dependency of the fracture toughness on the grain size for all SPM samples was not linear, and exhibited a peak. The fracture toughness was more dependent on the grain size than the flexural strength. While, the fracture toughness of the 2.0 mol% Y₂O₃ sample showed little dependence on the transformed monoclinic phase fraction and compressive residual stress as same as the flexural strength. Claussen et al.²⁻³ investigated the relationship between the fracture toughness and volume fraction of ZrO₂.
in Al₂O₃ matrix with a dispersion of unstabilized ZrO₂ particles. They reported that the microcrack-toughening improved the fracture toughness, but had little effect on the flexural strength. In the case of the 2.0 mol% Y₂O₃ sample, large amount of the monoclinic phase fraction was present within the sample despite of the grain size as shown in the thermal expansion behavior, and resulting in involvement of microcracks. The amount and size of microcracks caused by the transformation were probably influenced by the variation of Y₂O₃ content in ZrO₂ grains as with the variation of the As temperature in the thermal expansion behavior. This behavior for the fracture toughness of the 2.0 mol% Y₂O₃ sample was similar to that reported by Claussen et al.⁶ Accordingly, the flexural strength and fracture toughness of the 2.0 mol% Y₂O₃ sample seems to be mainly dominated by the microcrack-toughening. Since the increase of the transformed monoclinic phase fraction was found, but lower than other SPM and LPM samples, it is estimated that the contribution of stress-induced transformation toughening was much lower than those of the microcrack-toughening.

The samples with 2.5 and 2.8 mol% Y₂O₃ exhibited a strong relationship between the fracture toughness and the compressive residual stress, and the fracture toughness showed a maximum value at a certain grain size, at which the compressive residual stress also reached a maximum value. In addition, the stress-induced transformation toughening might strongly contribute to the fracture toughness, because the transformed monoclinic phase fraction was larger than that of the LPM sample. Furthermore, as the transformed monoclinic phase by grinding stresses was present within the sample, the microcrack-toughening might have influence on the fracture toughness as same as the stress-induced transformation toughening. The fracture toughness of the 2.5 mol% Y₂O₃ sample reached a maximum value of 9.9 MPa√m, which is much larger than that for Y-TZP prepared by LPM.⁴ And, the fracture toughness of the 2.5 mol% Y₂O₃ sample was larger than that of the 2.8 mol% Y₂O₃ sample, too. It is attributed that the compressive residual stress and the transformed monoclinic phase fraction improving the fracture toughness of the 2.5 mol% Y₂O₃ sample was larger than that of other samples. The fracture toughness of the 2.5 and 2.8 mol% Y₂O₃ samples decreased with increasing the grain size over the grain size showing a maximum fracture toughness. This suggests that each toughening effect was reduced by the decrease of the compressive residual stress and the transformed monoclinic phase fraction due to the progress of the stabilization of tetragonal grains.

For the 2.8 mol% Y₂O₃ sample with the smallest grain size, the flexural strength was low, on the contrary to high fracture toughness. It is thus considered that the flexural strength and fracture toughness were dominated by the microcrack-toughening as reported at the previous investigation.⁵ While, in
the case of the 2.5 mol% Y2O3 sample with the smallest grain size, both the flexural strength and fracture toughness were low compared to those of the 2.8 mol% Y2O3 sample. The difference of the relationship between the flexural strength and fracture toughness was presumably dependent on the difference of the microcrack size. It can be estimated that the microcrack size of the 2.5 mol% Y2O3 sample was larger than that of the 2.8 mol% Y2O3 sample, because the transformed monoclinic phase fraction of 2.5 mol% Y2O3 sample was larger than that of the 2.8 mol% Y2O3 sample, and the transformation zone of the 2.5 mol% Y2O3 was deeper than that of the 2.8 mol% Y2O3 sample as discussed in relation to thermal expansion behavior.

The fracture toughness of the 3.5 mol% Y2O3 sample was slightly dependent on the grain size. The compressive residual stress of the 3.5 mol% Y2O3 sample was low, and the monoclinic phase fraction on the polished surface was smaller than that of the 2.5 and 2.8 mol% Y2O3 samples. Therefore, the fracture toughness of the 3.5 mol% Y2O3 sample might dominate by the stress-induced transformation toughening rather than the compressive residual stress and the microcrack-toughening, similarly to the SPM sample.

For Y-TZP prepared by LPM, the transformed monoclinic phase fraction increased as the grain size increased due to the decrease of the critical stress for the transformation. In addition, the fracture toughness increases with increasing the transformed monoclinic phase fraction.3,4) On the other hand, for the 3.5 mol% Y2O3 sample prepared by SPM, since the conflicting relationship between the stabilization of the tetragonal grains and the acceleration of stress-induced transformation was observed, the fracture toughness might exhibit a slight peak against the grain size by the combined effect of both these processes.

As mentioned above, the flexural strength and fracture toughness of Y-TZP prepared by SPM are probably dominated by not only the stress-induced transformation, but also the compressive residual stress and microcrack-toughening. The contribution ratio of each toughening effect is estimated to vary according to the Y2O3 content and grain size.

3.6 Relationship between the transformation depth and fracture toughness

The relationship between the fracture toughness and \( V_f \sqrt{h_f} \) (\( V_f \): tetragonal phase fraction, \( h_f \): transformation depth) for each sample is shown in Fig. 10. The transformation depth was calculated using the expression given by Kosmac.7) The relationship between the fracture toughness and \( V_f \sqrt{h_f} \) for the 2.0, 2.5 and 3.5 mol% Y2O3 samples exhibited a peak. On the other hand, that relation for the 2.8 mol% Y2O3 sample was linear.2) However, if the relation for 2.8 mol% Y2O3 sample would be obtained for wider range of \( V_f \sqrt{h_f} \), the curve might exhibit a maximum point, because the microstructure of the 2.8 mol% Y2O3 sample is essentially the same as that of the other samples.

In the case of the 2.0 mol% Y2O3 sample with large monoclinic phase fraction, the fracture toughness was mainly dependent on the microcrack-toughening as discussed above. Kosmac et al.8) investigated that in the Al2O3–ZrO2 composite with tetragonal and monoclinic phases, the relationship between the fracture toughness and \( V_f \sqrt{h_f} \), and reported the relation was not linear and exhibited a peak at a certain \( V_f \sqrt{h_f} \) as with this investigation. Hence, the fracture mechanism of 2.0 mol% Y2O3 sample with tetragonal and monoclinic phases was presumably the same as the Al2O3–ZrO2 composite reported by Kosmac et al.8) For the 2.5 mol% Y2O3 sample, the fracture toughness was dependent on the grain size, and the relationship between the fracture toughness and \( V_f \sqrt{h_f} \) exhibited a peak. The fact that the fracture toughness of the 2.5 mol% Y2O3 sample was higher than that of other samples suggests that the toughening effect by the stress-induced transformation and compressive residual stress was the highest among the all samples. The fracture toughness of the 3.5 mol% Y2O3 sample was mainly dominated by the transformation, similar to that of the LPM sample. However, the relationship between the fracture toughness and \( V_f \sqrt{h_f} \) for the 3.5 mol% Y2O3 sample differed from that for the LPM sample, and the fracture toughness showed almost constant against \( V_f \sqrt{h_f} \). It seems that the fracture toughness of the 3.5 mol% Y2O3 sample was independent on \( V_f \sqrt{h_f} \), due to the occurrence of the conflicting relationship between the stabilization of the tetragonal grains and the acceleration of stress-induced transformation, as discussed above.

McMeeking et al.8) reported that the fracture toughness was proportional to the product of the tetragonal phase fraction and the transformed zone for Y-TZP with a high homogeneous distribution of Y2O3 prepared by LPM. Mori et al.8) investigated the relationship between the fracture toughness and \( V_f \sqrt{h_f} \) for Y-TZP prepared by LPM, and reported the relationship was linear, regardless of the Y2O3 content, because the stress-induced transformation was the main contribution to fracture toughness differently to the SPM samples. In the case of the SPM samples, the fracture toughness presumably dominated not only the stress-induced transformation, but also the compressive residual stress and microcrack-toughening. This suggests that the fracture mechanism in Y-TZP prepared by SPM is not expressed by the equation reported by McMeeking et al.10) and the fracture toughness of the SPM samples should be expressed by the equation: \( K_m = K_f + K_m + K_w \) where \( K_f \), \( K_m \), and \( K_w \) correspond to the contribution by the compressive residual stress, microcrack-toughening and stress-induced transformation toughening, respectively.2)

As the contribution ratio of each effect might vary according to the Y2O3 content and grain size, the relationship between the fracture toughness and \( V_f \sqrt{h_f} \) probably exhibits a different profile from that for Y-TZP prepared by LPM.2)

4. Conclusions

The mechanical properties of 2.0–3.5 mol% Y2O3-stabilized zirconia polycrystals prepared by the solid phase method were investigated in view of various relationships with grain size, residual stress and phase transformation behavior. The following conclusions were made.

(1) The density of the 2.5 and 2.8 mol% Y2O3 samples were...
higher than the 2.0 and 3.5 mol% Y$_2$O$_3$ samples. The density of the sample with 2.0 mol% Y$_2$O$_3$ was less than 5.9 g/cm$^3$, due to the presence of the monoclinic phase.

(2) The grain size decreased with increasing Y$_2$O$_3$ content for the same sintering temperature. The difference in the grain size became small when sintering was performed at high temperature. The grain growth rate was increased with increasing the Y$_2$O$_3$ content.

(3) The tetragonal phase fraction of the 2.5 and 2.8 mol% Y$_2$O$_3$ samples was significantly increased by sintering from 1275 to 1300°C, while the monoclinic phase fraction decreased. The tetragonal phase fraction of the 2.0 mol% Y$_2$O$_3$ sample was slightly increased. For the 3.5 mol% Y$_2$O$_3$ sample, the tetragonal phase fraction was not dependent on the grain size and remained almost constant.

(4) For the 2.5 and 2.8 mol% Y$_2$O$_3$ samples, a monoclinic to tetragonal phase transformation in the thermal expansion curve was observed during the heating stage. However, the thermal expansion curve during the cooling stage was linear, regardless of the sintering temperature. On the other hand, for the 2.0 mol% Y$_2$O$_3$ sample sintered at any temperature, a hysteresis was observed in the thermal expansion curve due to the well-known reversible transformation during both the heating and cooling stages. Accordingly, it was estimated that the monoclinic phase fraction was essentially absent in the 2.5, 2.8 and 3.5 mol% Y$_2$O$_3$ samples, but was present in the 2.0 mol% Y$_2$O$_3$ sample.

(5) It is proposed that the fracture toughness is dependent not only on the stress-induced transformation, but also on a combined effect of the compressive residual stress and microcrack-toughening caused by the transformation. The fracture toughness of the sample with 2.5 mol% Y$_2$O$_3$ was 9.9 MPa$\sqrt{m}$, which is much larger than that of Y-TZP prepared by LPM. The fracture toughness of the 2.0 mol% Y$_2$O$_3$ sample was dominated by microcrack-toughening caused by the transformation during the cooling stage of the sintering process. The relationship between the fracture toughness and $V_f\sqrt{t}$ for the 2.0, 2.5 and 3.5 mol% Y$_2$O$_3$ samples exhibited a peak, but was linear for the sample with 2.8 mol% Y$_2$O$_3$.

References