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Transformation and Stress in Y-TZP joined with Metallic Materials

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Abstract

Y-TZP(Yttria containing Tetragonal Zirconia Polycrystal) and metallic materials(type SUS304 stainless steel and Mo) were joined, using brazing alloy(Ag-Cu-Ti) sheet. Transformed fraction(Tf) in Y-TZP from tetragonal to monoclinic phase was investigated in relation with the thermal stress. Y-TZP disks having different Y_2O_3 contents, grain size were prepared for changing their transformability. Y-TZP disks having various thickness were joined with different metals in order to change the thermal stress states. Following results were obtained: Tf became large when thermal stress state on Y-TZP surface was tensile. Coarse grained Y-TZP joints with low Y_2O_3 content had large Tf and dependence of Tf on the thermal stress was small. Tf in Y-TZP joints increased when cooled at smaller rate and was larger than in the unjoined Y-TZP(under the condition of tensile state in Y-TZP). The cooling rate dependence was related with the isothermal characteristics of the transformation, which was accelerated by the thermal stress.

Keywords : Y-TZP, joint, thermal stress, transformation, dilatometry

1. Introduction

Y-TZP has excellent strength and fracture toughness[1], comparing with the other ceramic materials. Moreover, it has a superior properties against corrosion[2] and abrasion[3]. Various application has been taken into consideration, such as the insulation of combustion chamber for diesel engines[4], thermal barrier coatings[5], etc. Their application is possible to be extended by making a combined use with metallic materials, for example, by joining with each other. Generally, in the practical fabrication of the joints, thermal stress[6] is generated due to the difference in thermal expansion constants. Many attempts have been made to lower the stress, such as by inserting a intermediate layer[7] and by grading[8] the composition and microstructures in the neighbor of the metal/ceramics interfaces. The analysis of the stress distributions is important for the interfacial designs, and there are many studies[9,10]reported.

Zirconia undergoes transformation from high temperature tetragonal(t) phase to low temperature monoclinic(m) phase(t-m transformation) upon cooling, accompanying volume expansion of 4%[11]. This is induced under the presence of applied stress[12], therefore, occurrence of the transformation is dependent on the stress states in the joints. On the other hand, the stress distribution in the joints is altered by the transformation. Consequently, they are interrelated with each other. It is of importance to estimate the amount of t-m transformation taken place during the fabrication process of the joints. In order to consider the occurrence of the transformation in the joints, the following characteristics of t-m transformation have to be taken into consideration:

1, The transformation is induced by (tensile)stress.
2, The transformation occurs readily in the larger grains[13].
3, The transformation is depressed with an increase of Y_2O_3 content[14].
4, The transformation has an isothermal characteristics [15,16]

This study is intended to investigate the transformation of Y-TZP disks joined with metallic materials. Y-TZP disks having different thickness, Y_2O_3 contents and grain sizes were prepared. They were joined with stainless steel of type SUS304 and Mo, having constant thickness. The former material has the larger thermal expansion constant than Y-TZP and the latter has the less. Variation of Y-TZP thickness and using different metals are intended to change thermal stress distributions in the joints. On the other hand, variations of Y_2O_3 contents and the grain size were intended for controlling their transformability. Lastly, the joints were cooled from the joining temperature at different rates in order to control the rate of isothermal transformation. Joints with coarse grained Y-TZP *in this study* does not have practical significance because of high susceptibility to transformation and having lower strength. However, they were utilized to investigate the effects of various factors on the transformation in Y-TZP joined with metallic materials.

2. Experimental

2-1, Specimen preparation

Y-TZP disks were cut from the sintered compact rods of ZrO_2-Y_2O_3 produced by Tosoh Inc. The compacts having different Y_2O_3 contents(2,3,4mol%: 2Y,3Y,4Y) were prepared.

Rods of commercial stainless steel(type SUS304) and Mo(3 nines) were utilized as the metallic materials. Diameters of the rods were 1.5mm, and they were cut into disks. Y-TZP had various thickness between 0.4 and 5mm, while the thickness of metals were 3mm and kept constant. The metal surface was wet-polished with alumina fine grains of 1 μm. Y-TZP surface was polished with diamond paste(finished with Kulzer diamond polishing agent MM140). Y-TZP pieces were annealed in air at 1773K for 24h in order to change their grain size. The grain size and microstructures of the Y-TZP specimens were observed by means of transmission electron microscope(TEM).

In order to join these materials, a brazing alloy(Cusil ABA: 63Ag-35Cu-2Timass %, thickness: 0.05mm) sheet was utilized. The specimen pieces were set in a zig made of stainless steel and they were placed in a silica tube, which was heated in an electric resistance furnace. Joining was conducted in Ar(99.995% in purity) atmosphere at temperature of 1113K(20deg above the melting temperature of the brazing alloy) for 1.8ks without applying pressure, then cooled at different rate.

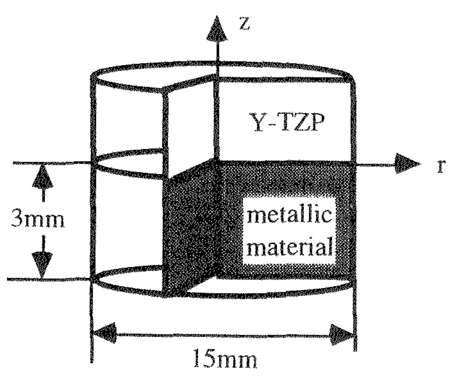


Fig. 1 : Schematic illustration of joints and r, z coordinates.

2-2, Detection of transformation

In this study, transformation in the unjoined Y-TZP compact bars(having scales of typically, 3 x 3 x 15 mm)were detected by means of thermal dilatation measurement. The measurements were conducted between the room temperature and around the joining temperature of 1113K. In thermal cycles of dilatation measurements, heating rate was set constant to 0.4deg/s. The specimens were kept at 1113K for 1.8ks then cooled at different rates.

The transformation was also determined by means of X-ray diffraction(XRD). The XRD profiles were obtained from the central region on the Y-TZP surface of the joined disks(as shown schematically in Fig. 1) and on the unjoined bars. For estimation of transformed fraction(Tf), the following equation was used:

$$Tf(\%) = (I_{\bar{1}11m} + I_{111m}) / (I_{\bar{1}11m} + I_{111m} + I_{111t}) \times 100$$

,where I_{hkl} stands for the intensity of (hkl)XRD peak(in a unit of CPS), and m,t stand for monoclinic and tetragonal phase of zirconia, respectively.

2-3, Calculation of thermal stress distribution and measurement of the residual stress

In order to estimate the thermal stress generated by the difference in thermal expansion coefficients between Y-TZP and metallic materials, axisymmetric thermoelastic finite element analysis(FEM) was conducted. The FEM program used was listed in the text[17]. The r,z coordinates taken for the analysis are shown in Fig. 1. Material properties listed in Table 1 were input for calculation. Residual stress on Y-TZP surface was measured by X-ray method[18].

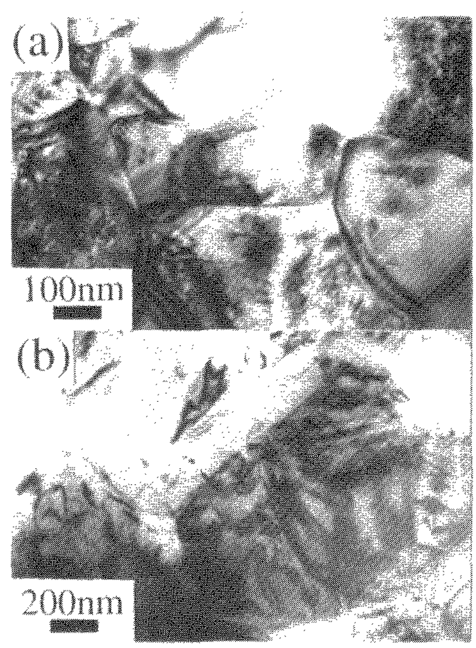


Fig. 2 : TEM photographs of specimens.
(a) as-received and (b) annealed at 1773K for 84.6ks.

3. Results

3-1, Transformation behavior of the unjoined Y-TZP

Microstructures of the as-received and the annealed Y-TZP(ZrO_2-2mol%Y_2O_3:2Y) specimens were observed by means of TEM, their photographs are shown in Fig. 2. The average grain size of Y-TZP was about 0.4μm and 1.4μm, respectively. In the latter case, the most of the grains transformed in the specimen preparation process and plenty of twins were observed in monoclinic phase. The Tf in various Y-TZP specimens was measured by means of X-ray diffraction before joining. They are listed in Table 2. The Tf was larger in the annealed (coarse grained)specimens than in the as-sintered (fine grained) specimens. However, it was noticeable that considerable fraction(Tf: 33.5%) was obtained in the fine grained 2Y specimens when they had undergone the same heat treatment as that of joining(cooled at 0.03deg/s). The dynamic transformation behavior was detected by means of thermal dilatometry. The displacements of the specimen length with variation of temperature are shown in Fig. 3.

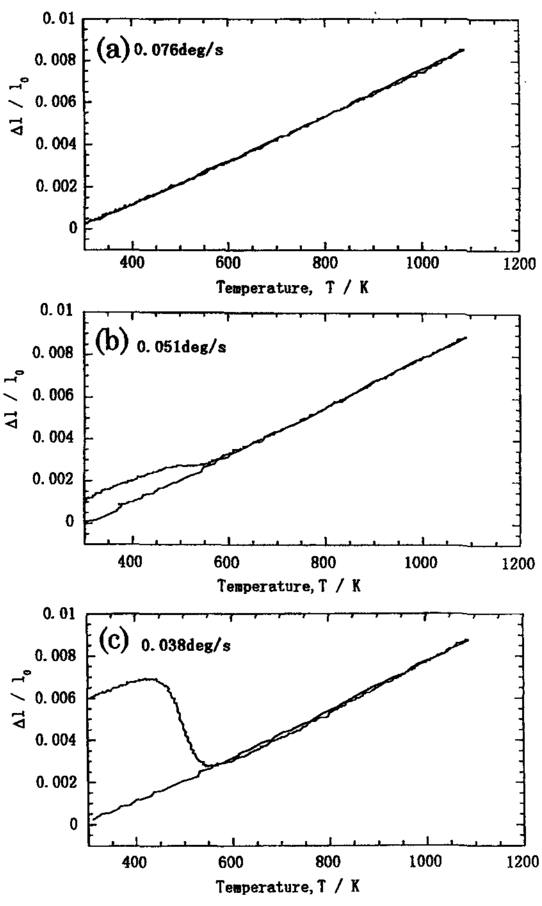


Fig. 3 : Dilatation curves of unjoined coarse grained 2Y-TZP, obtained at various cooling rates.

Y-TZP bars were heated to 1113K(joining temperature), then cooled at different rates. No transformation was detected by dilatometry from the fine grained Y-TZP, regardless of the cooling rates. Therefore, the transformation(Tf of 33.5%) in fine grained 2Y-TZP(listed in Table 2) is supposed to have occurred on the specimen surface, and not in the bulk. The Tf detected by XRD was always larger than the Tf by dilatometry.

In the dilatometry curves, abrupt increase in the specimen length occurred at about 570K due to the onset of t-m transformation, when cooled at lower rate than 0.051deg/s. The Tf increased as the cooling rate decreased. Dependence of Tf on the cooling rate is supposed to be related with the isothermal characteristics of the transformation in Y-TZP. It was noticeable that the transformation start temperature(M_s point) remained almost unchanged. The specimens contained cracks, however, they were comparatively small[13] in population, considering the degree of dilatation.

3-2, Stress states in Y-TZP / metallic materials joints

Different thermal stress states in the joints were obtained by changing the thickness and by selection of metallic materials. Thermal stress distribution due to the temperature difference(ΔT) between the joining temperature (1113K) and M_s point were calculated by FEM. In the central region of the Y-TZP disk surface, radial stress component

Table 1. Values of material properties used for FEM analysis.

	α /x10^{-6}K^{-1}	E/ GPa	ν
Y-TZP	9.0	200	0.3
SUS304	18.7	195	0.3
Mo	5.7	320	0.38

Table 2. Transformed fraction(Tf) and residual stress in the unjoined Y-TZP

	Tf / %	σ_{rr} / MPa
a) Fine grained Y-TZP (as-ground)	4.5	-120
b) Fine grained Y-TZP (heated to 1173K, then cooled at 0.03deg/s)	33.5	-71
c) Coarse grained Y-TZP (heated to 1173K, then cooled at 0.03deg/s)	60	<50
d) Coarse grained Y-TZP (heated to 1173K, then cooled at 0.1deg/s)	<5	<50

(σ_{rr}(=$\sigma_{\theta\theta}$))was significant and the other component(σ_{rz}) was almost zero[18]. As an example, σ_{rr} distribution in the Y-TZP/SUS304 steel joint is shown in Fig. 4, in which both disks have the same thickness of 3mm. σ_{rr} in the interfacial area of Y-TZP was negative(compressive) and inversed to be positive(tensile) in the surface area. Calculated values of σ_{rr} at the central region of Y-TZP disk are plotted in Fig. 5 for the different joints. In two types of material pairs, the σ_{rr} values had opposite dependence on the thickness of Y-TZP. The sense of stress component on the Y-TZP surface changed from compression to tension as the increase of Y-TZP thickness in the joints with SUS304 steel, and vise versa in the joints with Mo. The measured σ_{rr} on the fine grained Y-TZP is also plotted in the figure. The measured stress was compressive in all the cases. The tensile states on Y-TZP surface might induced the transformation and converted into the compressive states because of the volume expansion.

3-3, Dependence of Tf on Y_2O_3 contents.

Fine grained Y-TZP disks having different thickness and Y_2O_3 content were joined with SUS304 steel disks. The relationships between the Y-TZP thickness and Tf are plotted in Fig. 6. The Tf decreased with Y_2O_3 contents, and no monoclinic phase was detected in 4Y-TZP. In this study, 2Y-TZP was investigated intensively, because the Tf was larger and detection of transformation was easier. Monoclinic phase on 2Y-TZP having thickness of 2mm was about 30%. In this thickness ratio, σ_{rr} on Y-TZP was estimated not to be large(less than 200MPa in Fig. 5, open circle). It is noticeable that Tf of about 30% was closer to the values in the unjoined Y-TZP after the treatment(Table 2). The Tf was larger when thermal stress(σ_{rr}) was positive(tensile, 3mm) and the Tf was smaller in the negative (compressive, 1mm) case.

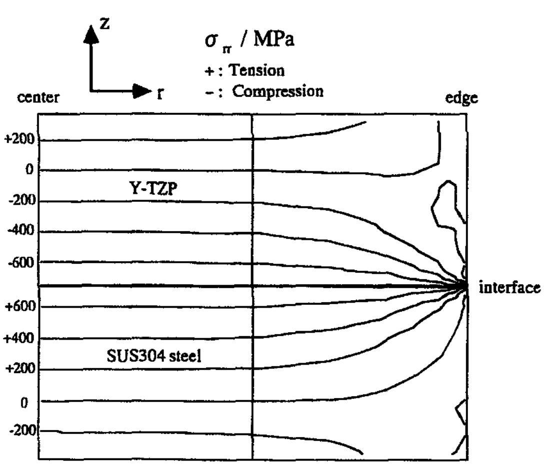


Fig. 4 : Thermal stress distribution in theY-TZP/ SUS304 steel joints, calculated by means of FEM.

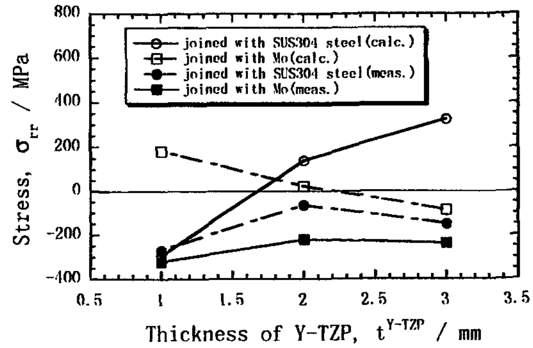


Fig. 5 : Calculated and measured σ_{rr} at the central part on the surface of Y-TZP disk with different thickness, joined with SUS304 and Mo.

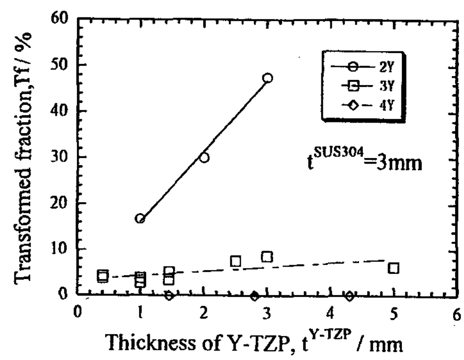
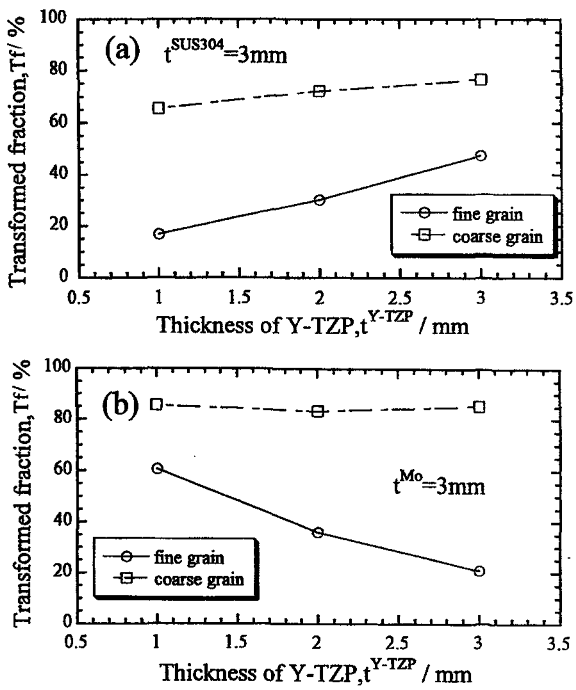


Fig.6: Relationship Y-TZP thickness and transformed fraction(Tf) in Y-TZP having different Y_2O_3 content.

3-4 , Dependence of the transformed fraction on the thickness ratio of the joints.

Y-TZP(2Y) disks having different thickness and grain size were joined with SUS304 steel and Mo disks. The relations between the Tf and the thickness are plotted in Fig. 7(a),(b). As the thickness of fine grained Y-TZP increased, Tf increased in the SUS304 steel joints and decreased in the Mo joints. The Tf on the Y-TZP surface was related with the thermal stress states in the joints(estimated by FEM analysis), namely, the Tf was larger when σ_{rr} was positive(tensile) and smaller when σ_{rr} was negative(compressive). The Tf on the coarse grained Y-TZP are also included in Fig. 7(a),(b). The Tf was larger than the fine grained Y-TZP cases, and the thickness dependence was less obvious. Especially, in the Y-TZP/Mo joints, the dependence was weaker. In these joints, thermal stress σ_{rr} in the interfacial area of Y-TZP is expected to be always tensile. Transformation occurred in the interface area readily, because of the larger grain size. The volume expansion in the interfacial region caused the successive transformation in the surface region. Consequently, large Tf was obtained in the 3mm Y-TZP case, even FEM calculation predicted the compressive thermal stress states on the surface.



F ig. 7 : Transformed fraction(Tf) on the surface of Y-TZP having various thickness in the joint pairs of (a) Y-TZP/SUS304 steel, and (b) Y-TZP/Mo.

3-5, Dependence on the cooling rate from the joining temperature

Unjoined Y-TZP having coarse grains had the larger Tf when cooled at the lower rate(Fig. 3). It is of interest to study the relationship between the cooling rate and the Tf in the joined states. Coarse grained Y-TZP(1mm thickness) joined with metallic materials were cooled at different rates from the joining temperature. The relationship between the cooling rate and the Tf is plotted in Fig. 8. The unjoined coarse grained Y-TZP had critical cooling rate of about 0.05deg/s, above which transformation was not detected. However, transformation occurred in Y-TZP disks joined with Mo when cooled at large rate of 1.13deg/s, as shown in Fig. 8.

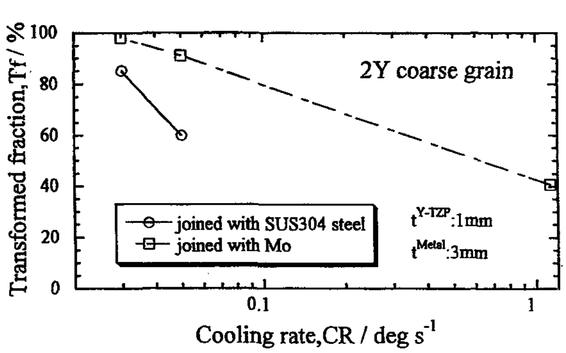


Fig. 8 : Relationship between cooling rate and transformed fraction(Tf) in the coarse grained Y-TZP joined with metallic materials.

The Tf decreased by increasing the cooling rate. It should be noted, however, that the successful joining was possible only when cooled at not too low rate, because otherwise the large fraction of transformation occurred, causing the large volume increase and the joints debonded.

4. Discussion

4-1. The effect of thermal stress on t-m transformation

The Tf on the fine grained Y-TZP disks was dependent on the Y-TZP thickness and on the selection of metallic materials, as shown in Fig. 7a,b. This dependence was caused by the differences in the thermal stress states in the joints. Values of thermal stress(abscissa in Fig. 9) responsible for enhancing or depressing the transformation was estimated by FEM for the temperature difference between the joining temperature(1113K) and the transformation start temperature(570K: Ms). The effective temperature difference should have taken smaller, if the deformation of brazing sheet and plastic deformation in metals are taken into consideration.

In calculation, it was assumed that M_s was not altered by an applied stress. It was also assumed that the elastic properties were constant in the experimental temperature range and they do not change by annealing.

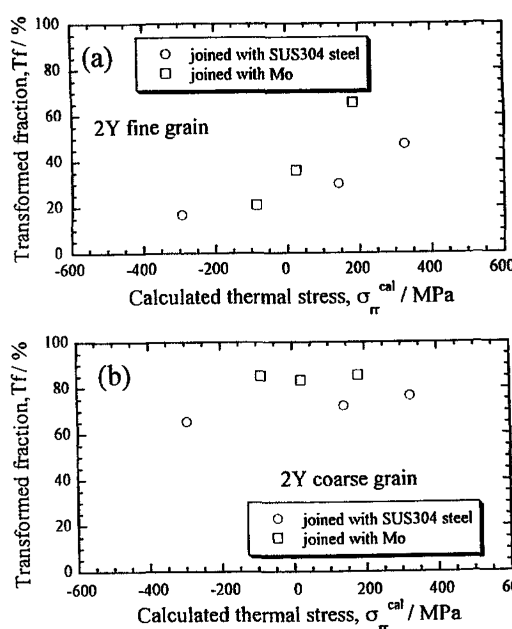


Fig. 9 : The relationship between the calculated thermal stress and the transformed fraction (Tf).
(a) in fine grained Y-TZP joints and (b) in the coarse grained Y-TZP joints.

Although the elastic constants were reported[19] to vary with temperature, the calculated thermal stress was not influenced very much within the range of their variation. The relationships between the calculated thermal stress and the measured Tf are plotted for the fine grained Y-TZP(Fig.9a) and for the coarse grained Y-TZP(Fig.9b). In the former case, Tf increased as the thermal stress on Y-TZP changed from compression to tension. The effect on the transformed fraction was evident. On the other hand, the correlation was not clear in the latter case. Large transformability and occurrence of subsidiary transformation were pointed out to be the causes for the less clear correlation.

4-2. The effect of cooling rate on the transformation fraction

In the unjoined coarse grained Y-TZP, the Tf increased as the cooling rate decreased. The same tendency was observed in the joined Y-TZP. The Tf of 40% was observed in Y-TZP/Mo joint cooled at the higher rate than 1 deg/s(Tf was less than 5%, cf. Table 2, d). These phenomena support the assumption that Y-TZP has characteristics of isothermal transformation under the presence of tensile stress, and the rate of transformation was enhanced by the tensile stress(although the effect of compressive stress was not clear in Y-TZP/SUS304 joints).

Transformation behavior in the Y-TZP joins is interpreted as follows: Under presence of the tensile stress, the incubation time before the onset of t-m transformation decreased and the rate of the transformation was accelerated. This caused the increase in Tf at the lower cooling rate. This phenomena has been analyzed, quantitatively[20] by means of the shift of TTT-diagram of the isothermal (pearlite formation)transformation, in the case of steels. The more precise and quantitative analysis are required in the future study in order to predict the transformation behavior in fabrication of the joints.

5. Conclusion

Y-TZP disks were joined with that of SUS304 steel and Mo with variation of transformability of Y-TZP, stress distribution in the joints and cooling rate from the joining temperature. Transformed fraction in Y-TZP disks was measured and their dependence on various parameters was investigated.

Correlation existed between the thermal stress(σ_{rr}) and the transformed fraction, that tensile radial stress enhanced the transformation. The transformed fraction in the joined Y-TZP was larger when cooled at lower rate. Considering the isothermal characteristics of t-m transformation, the increase in Tf at lower cooling rate was interpreted to be the acceleration of transformation rate by the applied tensile stress.

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