Effects of Residual Stresses on Strength and Crack Resistance in ZrO$_2$ with Alumina Coating

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Abstract: By covering a thin Al$_2$O$_3$ coating on ZrO$_2$ substrate, compressive stress caused by the mismatch of thermal expansion coefficients between the coating and substrate was introduced in the surface layer of Al$_2$O$_3$-ZrO$_2$ pre-stressed ceramics (marked as A$_3$Z$_6$ pre-stressed ceramics). Vickers indentation test was carried out to check the crack resistance in the surface layer and substrate influenced by the residual stresses. The enhancement of the crack resistance in surface layer results in a high flexural strength and excellent damage tolerance. Both theoretical analysis and experimental results show that the compressive stress and crack resistance in the surface layer increase with the increasing ratio of the cross-sectional area of ZrO$_2$ substrate to Al$_2$O$_3$ coating. Due to the residual compressive stress existing in Al$_2$O$_3$ coating, a high flexural strength of (1207±20) MPa was measured for ZrO$_2$ specimens coated with 40 μm Al$_2$O$_3$. The flexural strength is 32% higher than that of monolithic ZrO$_2$, and about triple the value of Al$_2$O$_3$. Meanwhile, compared to ZrO$_2$, the A$_3$Z$_6$ pre-stressed ceramics exhibit perfect thermal shock resistance.

Key words: Al$_2$O$_3$-ZrO$_2$ pre-stressed ceramics; compressive stress; flexural strength; damage tolerance

Zirconia (ZrO$_2$) materials have been widely applied in fuel cell, refractory materials, and construction materials due to the prominent mechanical and chemical properties such as high hardness, strength, good wear-resistant, fine oxygen ionic conductivity, excellent heat-insulating and corrosion protective$^{[1-3]}$. With the rapid advances of engineering applications, ceramic materials with higher strength and reliability are greatly needed. Therefore, significant improvements are essential to fabricate zirconia components with higher strength and excellent damage tolerance to improve the lifetime and safety of ceramic components. Many efforts including reinforcement strengthening (such as the incorporation of fibers, particles, and whiskers) and transformation toughening were applied to improve the strength and toughness of ZrO$_2$$^{[4-6]}$. Among them, the way to enhance the strength and damage tolerance of brittle ceramics by introducing residual compressive stress in the surface layer was more attractive because of its remarkable strengthening effect and convenience$^{[7-8]}$. It is well-known that the fracture of ceramics is mainly caused by crack propagation in the surface layer. Thus, the key to improve the strength and damage tolerance of ceramics is to enhance the resistance to crack propagation in the surface$^{[9]}$. By covering a coating with lower coefficient of thermal expansion (CTE) than the substrate, compressive stress will be generated in the coating layer after sintering. Then the compressive stress will hinder the crack extension, thereby improve the strength and damage tolerance of ceramics.

In order to illuminate the strengthening mechanism, it is important to understand the effect of residual stress on crack propagation. As it is well known that indentation deformation is strongly linked to the crack initiation and crack propagation. For the crack propagation behavior investigation, Vickers indentation proposed by Palmqvist in the late 1950s has been widely used...
because of its simplicity and efficiency\textsuperscript{[10]}. By comparing the growth of crack from the indentations made in unstrained and stressed materials, the effect of residual stress on crack propagation could be studied.

In this work, hot-pressed sintering (HP) was used to fabricate ZrO\textsubscript{2} ceramics covered by Al\textsubscript{2}O\textsubscript{3} coating with strong interface bonding and high density. For comparison, monolithic ZrO\textsubscript{2} ceramics were also prepared by the HP process using the same technical parameters. The effects of different ratio of cross-sectional area of substrate to coating (marked as S/s) on residual stresses and the crack resistance were further investigated.

1 Experimental
1.1 Material synthesis
Commercial zirconia with an average particle size of \(D_{95}=220\) nm (3mol% Y\textsubscript{2}O\textsubscript{3}, G3Y-0200O, Shandong Sinocera Functional Materials Co. Ltd., China) and alumina with an average particle size of \(D_{95}=220\) nm (SAO-020A-M, Shandong Sinocera Functional Materials Co. Ltd., China) were used as the raw materials in this research. Alumina powders, deionized water, and dispersant (Isobam104 with a molecular weight of 55,000–60,000, provided by Kuraray Co., Ltd., Osaka, Japan) were added to produce alumina slurry, and their mass contents were 30%, 69.2%, and 0.8%, respectively. The mixture was mixed using a planetary mill (YXQM-1L, MITR, China) for 24 h in zirconia containers with the ball-to-powder weight ratio of 10:1 and rotational speed of 150 r/min.

To fabricate the A\textsubscript{C}Z\textsubscript{S} pre-stressed ceramics by hot-pressed sintering, graphite die with a diameter of 50 mm was uniformly sprayed with the above alumina slurry. And the thickness of the Al\textsubscript{2}O\textsubscript{3} coating was about 40 \(\mu\)m. Then zirconia powder was put into the Al\textsubscript{2}O\textsubscript{3}-coated graphite die followed by applying a constant pressure of 10 MPa. The specimens were heated up to 1450 °C with the pressure of 30 MPa for 60 min. Both the ZrO\textsubscript{2} specimens with a dimension of 3 mm\(\times\)4 mm\(\times\)36 mm and the A\textsubscript{C}Z\textsubscript{S} pre-stressed ceramics with the size of 3.6 mm\(\times\)4 mm\(\times\)36 mm were machined (the original thickness of A\textsubscript{C}Z\textsubscript{S} pre-stressed ceramics prepared by hot-pressed sintering was 3.6 mm).

1.2 Characterization
Morphology of the specimens was observed by using a scanning electron microscope (SEM) (Merlin, Zeiss, Germany). The flexural strength was measured by the three-point bending tests with a span length of 30 mm and a crosshead speed of 0.5 mm/min (according to ISO 14704:2000). The residual strengths were investigated to evaluate the thermal shock resistance after quenching the specimens from different temperatures (200 °C, 250 °C, 300 °C, 350 °C, 400 °C) to 20 °C water (according to ASTM C1525-18). The measured value of strength is the average of 5 separate measurements. In order to study the effect of residual stress on crack propagation behavior, the specimens with Al\textsubscript{2}O\textsubscript{3} coatings of different thicknesses (40, 30, 20 \(\mu\)m) were used, because the residual stresses change with the coating thickness. The Vickers indenter (Tukon2500B, Wilson, America) was used to generate the indentation and indentation cracks under different loadings.

2 Results and discussion
The A\textsubscript{C}Z\textsubscript{S} pre-stressed ceramics were designed as the schematic diagram for plates in Fig. 1(a). To generate compressive stresses in the surface layer, Al\textsubscript{2}O\textsubscript{3} was applied as the coating material, because it has a lower coefficient of thermal expansion (CTE) than ZrO\textsubscript{2}. Due to CTE difference between Al\textsubscript{2}O\textsubscript{3} coating and ZrO\textsubscript{2} substrate, residual compressive stress was generated in the Al\textsubscript{2}O\textsubscript{3} coating during the cooling process of the A\textsubscript{C}Z\textsubscript{S} pre-stressed ceramics. The way to fabricate pre-stressed ceramics is also available for any ceramic components of various shapes, including cylinders, tubes and spheres, as illustrated in Fig. 1(a). As for structural ceramics, the interface bonding
generates significant impacts on mechanical properties. Fine interface bonding manner was investigated from the continuous and tight interface illustrated in Fig. 1(b). The cross-sectional SEM image of the $A_cZ_S$ pre-stressed ceramics indicates that the thickness of Al$_2$O$_3$ coating is near 40 μm (while the thickness for ZrO$_2$ substrate is 3.6 mm). Therefore, by employing HP sintering, $A_cZ_S$ pre-stressed ceramics with a good interface bond between Al$_2$O$_3$ coating and ZrO$_2$ substrate can be successfully fabricated.

To investigate the effect of residual stress on crack propagation behavior, Vickers indentation tests were carried out on the ZrO$_2$ ceramics and the $A_cZ_S$ pre-stressed ceramics with different ratios of sectional area ($S_s/S_c$ is the ratio of substrate to coating in the schematic diagram of plates shown in Fig. 1(a)). By comparing the length and expanded direction of crack in Fig. 2(b) and Fig. 3, the indentation diagonal half-length $a$ and the crack length $c$ (illustrated in Fig. 2(a)) can be easily obtained through the observation of indentation morphology. On this basis, the indentation fracture resistance ($K_{I FR}$) used to describe the resistance to indentation cracking can be calculated by Eq. (1) [13]. Since the residual stresses are distributed anisotropically, the test conditions for determine the crack resistance does not satisfy all the application requirements of Eq. (1) mentioned in ISO 14627. It is effective to evaluate the crack resistance at different directions in the surface layer and substrate by comparing the measured values of $K_{I FR}$.

$$K_{I FR} = 0.000978 \left( \frac{E}{H_v} \right)^{0.5} \left( \frac{P}{c^{1.5}} \right)$$  \hspace{1cm} (1)$$

Where, $K_{I FR}$: the indentation fracture resistance value (MPa·m$^{1/2}$); $E$: modulus of elasticity (GPa); $H_v$: Vickers hardness (GPa); $P$: pressing in load (N); $c$: half of average of crack length (mm); $a$: half of average of diagonal line length of indentation (mm).

As for the indentation on the polished surface of ZrO$_2$ shown in Fig. 2(b), the cracks propagate outward along the diagonal direction. According to Eq. (1), the calculated $K_{I FR}$ of ZrO$_2$ ceramics was 9.10 MPa·m$^{1/2}$ (as shown in Table 1). However, different indentation morphology was presented at the $A_cZ_S$ pre-stressed ceramics with different thicknesses of Al$_2$O$_3$ coating in Fig. 3. As shown in Fig. 3(a-c), the crack in Al$_2$O$_3$ coating layer propagates along the direction parallel to the interface, while the crack in ZrO$_2$ substrate propagates along the direction perpendicular to the interface. The above phenomenon might be attributed to the compressive stress existed in the Al$_2$O$_3$ coating, while the tensile stress existed in the ZrO$_2$ substrate, as shown in Fig. 4. And it is well known that the compressive stresses could restrain crack extension and the tensile stress could promote crack initiation and growth [7-8]. Therefore, the cracks perpendicular to the interface were restrained in Al$_2$O$_3$ coating layer, while those were promoted in ZrO$_2$ substrates. Moreover, it was found that the length of crack in Al$_2$O$_3$ surface layer decreases with the increased value of $S_s/S_c$. The measured $K_{I FR}$ of Al$_2$O$_3$ coatings and ZrO$_2$ substrates varies with the coating thickness, as shown in Table 1. The result suggests that the variations in the length of crack is the consequences of varied residual stress in the $A_cZ_S$ pre-stressed ceramics with different values of $S_s/S_c$. Combined Eqs. (3) and (4), the residual stress in the Al$_2$O$_3$ coatings and ZrO$_2$ substrates could be calculated.
\[ \sigma_c = \left( \frac{S_c}{S_s} \right) \cdot \left[ 1 - \frac{E_s \cdot \alpha_s}{E_c \cdot \alpha_c \cdot \left( 1 + \frac{E_s}{E_c} \right)} \right] \cdot E_s \cdot \alpha_s \cdot \Delta T_c \]  

(3)

In any cross-section of the sample, the tensile stresses in the substrate and the compressive stresses in the coating should keep balance,

\[ \sigma_c S_c = \sigma_s S_s \]  

(4)

where \( S_s, E_s, \alpha_s \) are the values of the cross-section areas, elastic modulus and CTE of the substrate, \( S_c, E_c, \alpha_c \) are those of the coating, respectively. \( \Delta T \) the temperature difference, in this test: \( \Delta T = T_{\text{sintering}} - T_{\text{room}} = 1425^\circ \text{C} \).

Depending on the calculations, the values of residual stress in the coatings (\( \sigma_c \)) and substrates (\( \sigma_s \)) were plotted in Fig. 5. As is shown, \( \sigma_c \) increased while \( \sigma_s \) decreased with the value of \( S_s/S_c \) increasing. It can be deduced that, with the value of \( S_s/S_c \) increasing, the compressive stress in \( \text{Al}_2\text{O}_3 \) coatings increased, while the tensile stress in \( \text{ZrO}_2 \) substrates presented opposite tendency. As compressive stresses could restrain crack extension and the tensile stress could promote crack initiation and growth, the crack resistance of \( \text{AcZS} \) pre-stressed ceramics was enhanced as the value of \( S_s/S_c \) increasing. This is consistent with the crack propagation behavior shown in Fig. 2(b) and Fig. 3.

Fractographic features of \( \text{ZrO}_2 \) ceramics with and without \( \text{Al}_2\text{O}_3 \) coating (40 \( \mu \text{m} \) thickness) were observed through SEM after bending tests. As shown in Fig. 6(a) and 6(c), the fracture morphology is smooth with no ductile ridge marks. This means that the fracture mode of both \( \text{ZrO}_2 \) ceramics and \( \text{AcZS} \) pre-stressed ceramics is brittle fracture. Grain sizes of uncoated \( \text{ZrO}_2 \) ceramic and \( \text{ZrO}_2 \) substrate of \( \text{AcZS} \) pre-stressed ceramics displayed in Fig. 6(b) and (d) are in the range of 400 to 600 nm. Moreover, few pores can be found from the microstructure, indicating that the specimens prepared by the hot-pressing sintering method have high density.

Fig. 2 (a) Typical Vickers indentation of \( \text{ZrO}_2 \) ceramics; (b) Optical photograph of the indentation morphology for \( \text{ZrO}_2 \) ceramics without residual stress

Fig. 3 Optical photograph of the indentation morphology for \( \text{AcZS} \) pre-stressed ceramics with different ratio of the cross-sectional area, showing the effects of the compressive stresses in the coating and tensile stresses in the substrate
The flexure strength of ACZs pre-stressed ceramics with 40 μm thickness of Al₂O₃ coating is (1207±20) MPa, which is 32% higher than that of ZrO₂ ceramics ((821±15) MPa) fabricated by the same process and about 3-fold of the flexural strength of Al₂O₃ [14]. According to the previous reports [15-17], the Al₂O₃/ZrO₂ composites fabricated by the conventional methods, such as ZTA or ATZ, or laminated Al₂O₃/ZrO₂, mostly possess a strength with the value between that of Al₂O₃ and ZrO₂. Generally, the strength of ceramic composites may be between the reinforcement material and matrix. However, the pre-stressing design endows the ACZs ceramics with a higher strength than Al₂O₃ and ZrO₂. The results of the mechanical properties clearly show that the increase in strength is mainly attributed to the residual compressive stress in Al₂O₃ coating, thereby more energy was required for crack extension [18]. In addition, the robust interface between substrate and coating material also makes restrictions against crack growth, which has a significant effect on the improvement of the strength and toughness [19].

Another important mechanical property observed for ceramics is the damage tolerance, which represents the ability to inhibit crack extension. The residual strength of two materials was tested after thermal shock with various thermal shock temperatures, as illustrated in Fig.7. The residual strength of both ZrO₂ and ACZs pre-stressed ceramics with 40 μm thickness Al₂O₃ coating were reduced with the quenching temperature increasing. Especially, the residual strength of ACZs pre-stressed ceramics rapidly dropped from 1040 MPa to 317 MPa when the thermal shock temperature raised from 350 °C to 400 °C,
while the residual strength of ZrO₂ quenched at 350 °C was only 100 MPa. The result could be ascribed to that the compressive stresses existing in the coating layer can hinder the crack extension. Consequently, it is significant to introduce residual compressive stress in the surface layer of ceramic components to prevent crack propagation.

Owing to the high density and residual compressive stresses in the surface layer, ACZS pre-stressed ceramics were endowed with high flexural strength and excellent thermal shock resistance. Moreover, considering the feasibility in economy and simplification in the fabrication, the pre-stressing design to improve mechanical properties will have great practical value and prospect in the fields of structural ceramics and domestic ceramics. Besides, unlike the pre-stressed concrete and tempered glass, the pre-stressed ceramics can be machined to specific dimensions, which is more suitable for industrialized production.

Table 1 Parameters used in formulas and the results of the experiment and simulation

|                         | Elasticity modulus, E/GPa | p/N | a/μm | c/μm | H/V/GPa | K\_I,IFR/(MPa·m\(^{1/2}\)) |
|-------------------------|---------------------------|-----|------|------|---------|--------------------------|
| ZrO₂                    |                           |     |      |      |         |                          |
| AcZS with 40 μm coating | Al₂O₃ -40                 | 350 | 4.91 | 11.25| 47.50   | 17.98                   |
|                         | ZrO₂ -40                  | 220 | 19.62| 26.16| 36.88   | 13.28                   |
| AcZS with 30 μm coating | Al₂O₃ -30                 | 350 | 4.91 | 11.09| 44.38   | 18.49                   |
|                         | ZrO₂ -30                  | 220 | 19.62| 26.08| 36.05   | 13.37                   |
| AcZS with 20 μm coating | Al₂O₃ -20                 | 350 | 4.91 | 10.94| 25.00   | 19.02                   |
|                         | ZrO₂ -20                  | 220 | 19.62| 26.01| 35.87   | 13.44                   | 8.64                    |

Fig.7 Residual strength of AcZS pre-stressed ceramics and ZrO₂ ceramics, after quenching at different thermal shock temperatures

3 Conclusions

Pre-stressing design was used to enhance the strength and damage tolerance of ZrO₂ ceramics by coating a thin Al₂O₃ layer. The magnitude and direction of the residual stress were examined by Vickers indentation tests, and the crack resistance varied with the residual stress was further verified. Experiments indicated that the crack resistance of AcZS pre-stressed ceramics was enhanced with increasing ratio of the cross-sectional area of ZrO₂ substrate to Al₂O₃ coating.

On account of the residual compressive stress in the coating, the bending strength of ZrO₂ specimens coated with 40 μm thickness of Al₂O₃ was (1207±20) MPa which is much higher than that of ZrO₂ (850 MPa) and Al₂O₃ (400 MPa). After quenching from 350 °C to water, the residual strength of the AcZS pre-stressed ceramics was measured as 1040 MPa, while the measured residual strength of the ZrO₂ was only 100 MPa.

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力陶瓷的裂纹扩展阻力增加，最终导致强度和损伤容限提高；且 Al$_2$O$_3$ 预应力陶瓷表层的压应力和裂纹扩展阻力随着基体截面积与涂层截面积比值的增加而增大。当 ZrO$_2$ 基体表层的 Al$_2$O$_3$ 涂层厚度为 40 μm 时，由于表层压应力的存在而使得 Al$_2$O$_3$ 预应力陶瓷的弯曲强度达到(1207±20) MPa，相比于同种工艺下制备的 ZrO$_2$ 陶瓷强度提高了 32%，同时也是 Al$_2$O$_3$ 强度的 3 倍。此外，Al$_2$O$_3$ 预应力陶瓷也表现出很好的抗热震性能。

关 键 词： Al$_2$O$_3$–ZrO$_2$ 预应力陶瓷; 压应力; 弯曲强度; 损伤容限
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