

New Method to Measure Thermal Shock Resistance in Ceramics Using a Piezo-Spectroscopic Technique

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A new method for assessing the critical temperature in thermally shocked ceramics is proposed. It is based on the measurement of stress relaxation of residual stresses as a consequence of thermal shock. The change in the stress-field is determined by piezo-spectroscopic technique. The technique is described and the results analyzed. The values obtained are compared with those obtained by the conventional method based on strength degradation measured on test pieces quenched at different temperatures. The agreement among the data is very good.

I. Introduction

THE evaluation of thermal shock resistance of ceramics is crucial for components to be used at medium or high temperature or for tribological applications.

Several methods have been proposed^{1–6} and the results are directly related to the method used. In order to overcome this inconvenience, an international agreement has established a conventional standardized method.⁷ The standard is based on Hasselman unified theory^{8,9} and consists of measuring the flexural strength of bars after thermal shock by quenching in water. This method enables us to determine the onset of loss in strength in relation to increasing initial temperature. Because of the nonductility of ceramics, a gradient throughout the thickness of a component induces thermal stresses which cannot be easily relaxed. If these stresses exceed the strength of the material, then cracks may develop and grow to relieve the strain. Hasselman identified the critical temperature ΔT_c at which unstable crack growth initiates. At this temperature, a drastic strength drop occurs due to the released elastic energy being converted into kinetic energy.

For a very rapid thermal shock, the most commonly used parameter is represented by

$$R = \frac{\sigma_f(1 - \nu)}{E \cdot \alpha} \quad (1)$$

where R is a thermal shock parameter of the first type and corresponds to the temperature difference, ΔT , which results in thermal stress on the surface equal to the strength of the material; σ_f is

the fracture strength, ν is the Poisson's ratio, E is the Young's modulus and α is the coefficient of thermal expansion (CTE).

The widely used test method consists in quenching a set of bars at different temperatures in water at 20°C, then breaking the samples in flexural mode and identifying the ΔT at which the strength of the material has a drastic drop of more than 30% of its original value. This standardized method implies the use of at least 30 samples, permitting five test pieces to be used at each of the five testing temperatures, plus five for the measurement of initial strength of unshocked material. Though largely used, this method has some disadvantages: (i) it requires a large number of oppositely prepared samples, (ii) it cannot allow multiple shock measurements, (iii) it cannot be applied to coatings and (iv) most importantly, it cannot permit thermal shock measurements on real components.

For these reasons we thought to develop an alternative non-destructive method based on using a piezo-spectroscopic technique (PS) to measure the change in the stress field of a material as a consequence of thermal shock. The idea is based on the consideration that when a thermal gradient exceeds the ΔT_c and the thermal strain is relieved by the development and growth of cracks, the residual stress distribution in the material should change. The driving force for crack propagation is derived from the stress-field within the thermal shock specimen.⁸ Because a crack affects the compliance of a body¹⁰ the formation of a crack not only modify the local stress distribution but also reduces the stress level through the body.^{8,10} Then, under severe thermal conditions, the presence of cracks reduces the stress level. The formation of cracks in a stressed body is equivalent to the lowering of effective Young's modulus.¹⁰ A decrease in Young's modulus leads to a relaxation of the stress level, which in turn decreases the driving force for crack propagation.⁸

If it is possible to measure the residual stress-field before and after a thermal shock, it should be possible to associate the ΔT_c to an evident change in the stress distribution. This method can be applied especially to laminated structures or films and coatings (i.e., thermal barriers) where large residual stresses are present in the prepared materials.

The evaluation of stresses by PS relies on the measurement of the shift of spectral lines relative to their unstrained condition. The spectral shift, $\Delta\nu$, of fluorescence lines in Al_2O_3 and of the Raman bands under uniaxial stress can be expressed, to a linear approximation, as

$$\Delta\nu = \Pi_{ij}\sigma_{ij} \quad (2)$$

where Π_{ij} is the tensor of the PS coefficients and σ_{ij} is the stress tensors.

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Grabner¹¹ proposed that, for a single crystal and uniform stress state the Π_{ij} is a covariant tensor, identical in form in equivalent coordinate systems. For the single crystal these are defined by its point symmetry group. He also suggested that Π_{ij} is a symmetric tensor reflecting the symmetry of the stress tensor $\sigma_{ij} = \sigma_{ji}$. Thus, the resulting PS tensor is diagonal in the orthogonal crystallographic reference frame, if referred to the principal axes directions

$$\Pi_{ij} = \begin{bmatrix} \Pi_{11} & 0 & 0 \\ 0 & \Pi_{22} & 0 \\ 0 & 0 & \Pi_{33} \end{bmatrix} \quad (3)$$

In particular, for Cr^{3+} in sapphire single crystal, the standard reference frame is defined with 3-axis parallel to the crystallographic direction c -axis, the 1-axis parallel to the a -axis and the 2-axis parallel to the m -axis. Based on the assumption that the PS effect of R -lines is controlled by the point symmetry of Cr^{3+} ions in the trigonal sapphire crystal, the identity $\Pi_{11} = \Pi_{22}$ follows.¹²

Therefore, in polycrystalline materials without any significant texture and a uniform state of stress, with a good approximation, the Eq. (2) can be rewritten as:

$$\Delta\nu = \frac{1}{3} \Pi_{ii} \sigma_{ii} \quad (4)$$

where Π_{ii} is the trace of the PS matrix and σ_{ii} is the first invariant of the stress tensor ($\sigma_{ii}/3$, commonly referred to as the mean normal stress).

Therefore, if Π_{ii} is known, the spectral shift $\Delta\nu$ can be considered as a direct measure of the normal stress within the volume probed by the laser beam for each spectra acquisition.

In this communication we report the variation of the residual stresses as a function of ΔT of bars obtained from an alumina-zirconia composite (AZ) and quenched in water. The change of stress field distribution associated with the different temperatures has been compared with strength measured by bending bars quenched at the same ΔT .

II. Experimental Procedures

(1) Materials and Specimen Preparation

AZ samples were prepared in the volume ratio of 60/40. Both Al_2O_3 powder (Alcoa A16-SG, Alcoa Aluminum Co., New York, NY) and zirconia powder (TZ3Y-S, Tosoh Corp., Tokyo, Japan) doped with 3 mol% Y_2O_3 (usually referred to as 3Y-TZP), with an average particle size of 0.3 μm , were ball milled in ethanol using Al_2O_3 milling media. After drying, the powders were uniaxially pressed and then cold pressed isostatically at 3000 bar in the form of bars with geometrical dimensions of approximately 31.5 \times 5.1 \times 3.7 mm (length \times depth \times thickness, respectively). Finally, all the samples were sintered at 1550°C for 1 h with heating rate of 50°C/h to 600°C and 200°C/h from 600° to 1550°C.

After these preparation procedures, the test pieces presented a density of about 99% of the theoretical one. Finally, the sintered bars were slightly ground and the long edges chamfered.

(2) Thermal Shock and Strength Measurement

Eight bars for each ΔT (200°, 250°, 260°, 275°, 300°, 400°, and 600°C) were shocked according to the European Standard prEN 820-3.⁷ Six further bars were prepared to evaluate residual stress distribution and strength at room temperature (RT). For each ΔT , four bars were used to determine uniaxial flexural strength by means of a fourpoint bending test; the other four bars were analyzed by means of piezo-spectroscopy in order to evaluate the residual stress field.

(3) PS Measurements

The residual stress field distribution on the surface of the samples was determined by using piezo-spectroscopy (PS) technique

related to the characteristic R1 band produced by the chromophoric fluorescence of Cr^{3+} impurities in Al_2O_3 .

The choice of an AZ composite as a study material is due to the fact that, as a consequence of the different CTE of alumina and zirconia, the grains of the former undergo compressive residual stresses. Moreover, if alumina is used as stress sensor, it is easier to measure a microscopic stress field as a contribution of the different grains in the composite.

Generally, the macroscopic stress in this composite should be zero at equilibrium. However, the presence of a small fraction of monoclinic phase (identified by X-ray diffraction), due to the grinding of the test pieces, generates the presence of residual stress on a macroscopic scale. Therefore, the value of Π_{ii} measured for the composite can be used to assess the macrostress.

To measure merely macrostress, data were analyzed according to the approach suggested by previous authors.^{13,14} The specific Π_{ii} value related to the R1 band was calculated by a standard fourpoint bending calibration procedure. Details of the calibration procedures were given in a previous report.¹⁵ Stress mapping was performed on the surface of AZ monolithic bars, which underwent different thermal shock cycles. To collect fluorescence spectra, the spectrometer apparatus (ISA, T 64000 Horiba/Jovin-Yvon, Tokyo, Japan) employed in the present experiments used an argon-ion laser as an excitation source, operating at a wavelength of 488 nm with a power of 400 mW. Details on the apparatus and procedures can be found elsewhere.¹³

Macroscopic stress distributions were measured by collecting square maps of spectra on the specimen surface. Several measurements were done on each samples. The map sizes were both 500 $\mu\text{m} \times$ 500 μm and 2 mm \times 2 mm (to analyze a larger area of the sample). In the first case the square mesh was characterized by a 5 μm step, in the second case, steps of 20 μm were used. The frequency shifts were obtained by subtracting the center frequency of the peak obtained from unstressed test pieces from the center of the peak recorded in each point of the maps. The data reported are the average values of at least three measurements (in different zones) on each sample of the four used for each ΔT .

III. Results and Discussion

The mean strength values obtained by means of four-point bending on bars quenched at different temperatures as a function of ΔT are reported in Fig. 1.

From this diagram, it appears evident that a sharp drop in mean strength higher than 30% of the initial mean strength occurs at $\Delta T = 260^\circ\text{C}$.

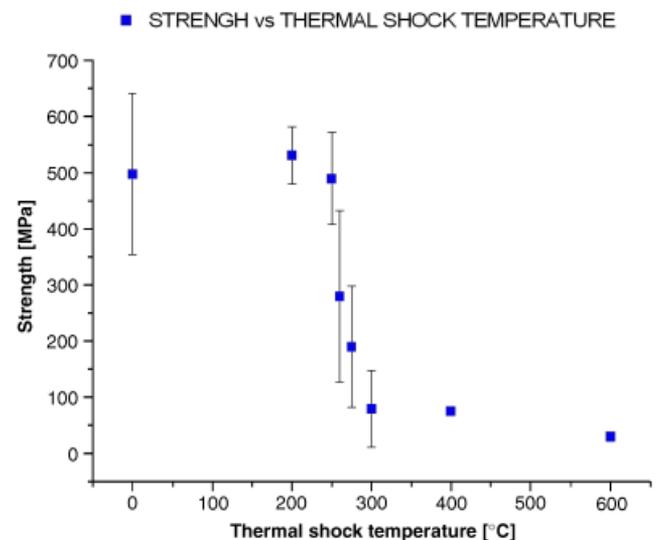


Fig. 1. Change in flexural strength after quenching from various temperatures which shows a sharp decline at a critical temperature as expected from the theory.

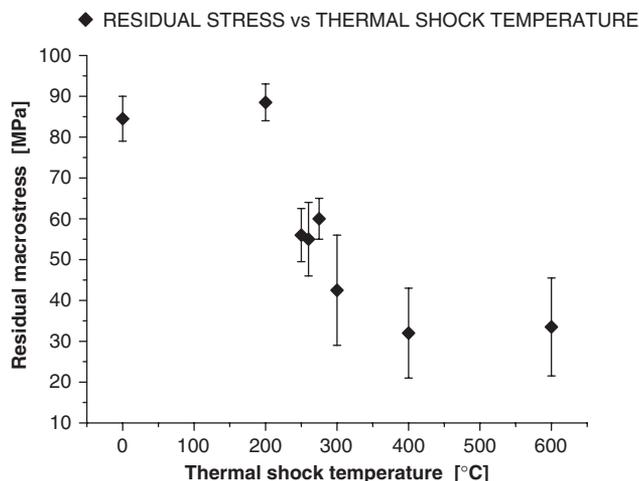


Fig. 2. Change in residual macrostress as a function of different thermal-shock temperatures. Similarity with Fig. 1 is evident.

The residual macrostresses have been calculated from PS measurements using the PS coefficient $\Pi_{\text{hydro}} = 9.0 \pm 0.1$ ($\text{cm} \cdot \text{GPa})^{-1}$ and R1 initial peak position equal to $14407.81 \pm 0.02 \text{ cm}^{-1}$.

As mentioned before, the small amount of macrostress of about 90 MPa can be observed on the surface of nonshocked samples due to the presence of some monoclinic grains. After thermal shock the stress distribution changes. A behavior very similar to the conventional diagram $\sigma - \Delta T$, reported in Fig. 1, can be observed considering the variation of residual stresses as a function of ΔT , as shown in Fig. 2. Also in this case, the drop in residual stress can be located at a $\Delta T \cong 260^\circ\text{C}$.

Quenching method is strongly influenced by the geometry and the size of the samples, however, because with the PS technique we consider the effects induced on the stress distribution by thermal shock (i.e., crack formation and crack propagation) this measurement is independent from the method used to apply a thermal stress. For this reason it can be used on bodies of different shape and volume.

IV. Conclusions

A new method to evaluate the thermal shock resistance of ceramics has been proposed. It is based on the measurement of the variation of residual stresses due to thermal shock. The value of critical ΔT identified with this method has been compared with the value obtained with the conventional-standardized method

based on the measurement of flexural strength retained by the material after quenching.

The values obtained with the two methods are highly coherent.

This method is nondestructive and the preparation of appropriate test pieces is not necessary. Consequently the method can allow measurement of thermal shock, particularly on massive materials made with composites, laminated structures, and coatings that contain residual stresses. It can be used also on bulk materials assuming that machining of a component introduces residual stresses in the system.

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References

- R. W. Davidge and G. Tappin, "Thermal Shock and Fracture in Ceramics," *Trans. Br. Ceram. Soc.*, **66** [8] 405–22 (1967).
- D. III Lewis, "Thermal Shock and Thermal Shock Fatigue Testing of Ceramics with the Water Quench Test"; pp. 487–96 in *Fracture Mechanics of Ceramics 6*, Edited by R. C. Bradt, A. G. Evans, P. H. D. Hasselman, and F. F. Lange. Plenum Press, New York, 1983.
- G. A. Schneider and G. Petzow, "Thermal Shock Testing of Ceramics—A New Method," *J. Am. Ceram. Soc.*, **74** [1] 98–102 (1991).
- E. H. Lutz and M. V. Swain, "Interrelation Between Flaw Resistance, R-Curve Behaviour and Thermal Shock Strength Degradation in Ceramics," *J. Am. Ceram. Soc.*, **74** [11] 2859–68 (1991).
- T. Anderson and D. Rowcliffe, "Indentation Thermal Shock Test for Ceramics," *J. Am. Ceram. Soc.*, **79** [6] 1509–14 (1996).
- A. G. Tomba and A. L. Cavalieri, "Alumina Disks with Different Surface Finish: Thermal Shock Behavior," *J. Eur. Ceram. Soc.*, **20** [7] 889–93 (2000).
- European Standard N° prEN 820-3 (rev. 2004), Advanced Technical Ceramics—Method for Testing Monolithic Ceramics—Thermomechanical Properties—Part 3: Determination of Resistance to Thermal Shock by Water Quenching, March 2004.
- D. P. H. Hasselman, "Unified Theory of Thermal Shock Fracture Initiation and Crack Propagation in Brittle Ceramics," *J. Am. Ceram. Soc.*, **52** [11] 600–4 (1969).
- D. P. H. Hasselman, "Thermal Stress Resistance Parameters for Brittle Refractory Ceramics, a Compendium," *Bull. Am. Ceram. Soc.*, **49** [12] 1033–7 (1970).
- J. P. Berry, "Some Kinetic Consideration of the Griffith Criterion of Fracture: I and II," *J. Mech. Phys. Solids*, **8** [3] 194–216 (1960).
- L. Grabner, "Spectroscopic Technique for the Measurement of Residual Stress in Sintered Al_2O_3 ," *J. Appl. Phys.*, **49**, 580–3 (1978).
- J. He and D. R. Clarke, "Determination of the Piezospectroscopic Coefficient for Chromium-Doped Sapphire," *J. Am. Ceram. Soc.*, **78** [5] 1347–53 (1995).
- G. de Portu, L. Miccè, and G. Pezzotti, "Measurement of Residual Stress Distributions in $\text{Al}_2\text{O}_3/3\text{Y-TZP}$ Multilayered Composites by Fluorescence and Raman Microprobe Piezo-Spectroscopy," *Acta Mater.*, **53**, 1511–20 (2005).
- Q. Ma and D. R. Clarke, "Stress Measurement in Single-Crystal and Polycrystalline Ceramics Using Their Optical Fluorescence," *J. Am. Ceram. Soc.*, **76** [6] 1433–40 (1993).
- G. Pezzotti, "In situ Study of Fracture Mechanisms in Advanced Ceramics Using Fluorescence and Raman Microprobe Spectroscopy," *J. Raman Spectr.*, **30**, 867–75 (1999). □