Available online at www.sciencedirect.com
ScienceDirect
Journal of the European Ceramic Society 27 (2007) 1307–1311
www.elsevier.com/locate/jeurceramsoc

Elastic properties and damping behaviour of alumina–alumina/zirconia laminates

K. Lambrinou [a,*], T. Lauwagie [b], F. Chalvet [c], G. de Portu [c], N. Tassini [d], S. Patsias [d], T. Lube [e], O. Van der Biest [a]

[a] Department of Metallurgy and Materials Engineering, Katholieke Universiteit Leuven, Kasteelpark Arenberg 44, Heverlee B-3001, Belgium
[b] Department of Mechanical Engineering, Katholieke Universiteit Leuven, Celestijnenlaan 300b, Heverlee B-3001, Belgium
[c] Instituto di Scienza e Tecnologia dei Materiali Ceramici, Consiglio Nazionale delle Ricerche, via Granarolo 64, Faenza 48018, Italy
[d] Dynamics Research Group, Department of Mechanical Engineering, University of Sheffield, Mappin Street, Sheffield S1 3JD, United Kingdom
[e] Institut für Struktur und Funktionskeramik, Montanuniversität Leoben, Franz-Josef-Straße 18, Leoben A-8700, Austria

Available online 9 June 2006

Abstract

This work focuses on the elastic properties and damping behaviour of alumina–alumina/zirconia laminates and their constituent layers as a function of temperature. The laminates were made by warm pressing and sintering tape-casted layers of alumina and alumina/zirconia (60/40 vol.%). The laminate stacking sequence and thickness of the constituent layers were tailored so as to have compressive residual stresses on the laminate surface, improving thus the laminate wear resistance. The temperature dependence of the elastic properties and damping behaviour of the laminates was determined using the impulse excitation technique (IET). Furthermore, the measured elastic properties were validated with the help of a finite element model (FE-model).

Keywords: Tape casting; Al_2O_3; ZrO_2; Mechanical properties; Spectroscopy

1. Introduction

Ceramics are excellent wear-resistant materials, since they combine high hardness, low density, and chemical inertness with the ability to operate under extreme conditions (e.g. high temperatures, aggressive environments, etc.), but their performance is limited by their brittleness. Since the performance of wear-resistant materials is related to the properties of thin surface layers, improving the surface toughness is a promising way to ameliorate the tribological behaviour of ceramics. An effective method to improve the surface toughness of a material is to produce laminated structures, which are tailored so as to induce compressive residual stresses at the surface, based on the thermophysical properties (i.e. coefficient of thermal expansion, and shrinkage upon sintering) of the constituent materials.[1]

Laminated composites made by warm pressing and sintering alternating layers of alumina (Al_2O_3) and alumina/zirconia (Al_2O_3/ZrO_2, 60/40 vol.%) produced by tape casting have shown to possess a promising wear-resistance.[1] The constituent materials of these laminates exhibit different shrinkages during sintering, and different thermal expansion coefficients ($\alpha \approx 9 \times 10^{-6}\,^{\circ}C^{-1}$ for pure Al_2O_3, and $\alpha \approx 10 \times 10^{-6}\,^{\circ}C^{-1}$ for 60/40 vol.% Al_2O_3/ZrO_2 in the 25–1400 °C temperature range). The described differences are sufficient to induce residual stresses without exceeding the values that lead to spontaneous cracking during the processing of alumina–alumina/zirconia (AZA) laminates.[1] Since the performance of AZA laminates relies on residual stresses, it is challenging to be able to predict the evolution of these stresses as a function of temperature. Predicting the temperature dependence of the residual stresses in AZA laminates can be done with finite element modelling, provided that the thermal expansion coefficient, shrinkage upon sintering, and elastic properties (as a function of temperature) are known for each of the laminate constituent materials (i.e. the pure alumina, and the 60/40 vol.% alumina/zirconia). This study focused on determining the elastic properties and damping behaviour of alumina (A) and alumina/zirconia (AZ) laminates as a function of temperature. In the next step of this investigation, the determined elastic properties

* Corresponding author at: IMEC, Kapeldreef 75, B-3001 Leuven, Belgium. Tel.: +32 16 281743; fax: +32 16 288500.
E-mail address: klambrin@imec.be (K. Lambrinou).

0955-2219/$ – see front matter © 2006 Elsevier Ltd. All rights reserved.
doi:10.1016/j.jeurceramsoc.2006.04.124

will be introduced in a finite element model (FE-model) of the AZA laminates to calculate the residual stresses, and to predict, in this way, the performance of these wear-resistant laminates as a function of temperature.

2. Work methodology

2.1. Preparation of laminates

For the needs of the present study, two types of sheets were prepared by tape casting: alumina (A), and alumina/zirconia (AZ). The starting materials used for the manufacturing of the tapes were a high purity (99.7%) alumina powder (Alcoa A16-SG, Alcoa Aluminium Co., NY, USA), and a zirconia powder doped with 3 mol.% Y_2O_3 (Zirconia TZ-3YS, Tosoh Corp., Tokyo, Japan); both powders had an average particle size of 0.30 μm. The two powders were mixed with the appropriate solvents, binder, dispersant and plasticizer to obtain alumina and alumina/zirconia slips suitable for tape casting. The tapes were dried, punched, and the produced sheets were stacked by thermo-compression (30 min at 80 °C under a pressure of 30 MPa) in the desired sequence to produce A (10 layers of pure alumina), AZ (10 layers of alumina/zirconia, 60/40 vol.%), and AZA (9 layers: A/A/AZ/A/AZ/A/AZ/A/A) laminates. The 'green' laminates were first heated at 3 °C/h between 200 and 600 °C to allow the burnout of the binder, and were subsequently sintered to nearly full density (2–3% residual porosity) at 1550 °C for 1 h (heating and cooling rate during sintering: 30 °C/h). Rectangular bars were cut out of the A, AZ and AZA laminates to make the test specimens; the mass and dimensions of these bars are given in Table 1.

2.2. Impulse excitation technique

The impulse excitation technique (IET) is a resonant-based, non-destructive, standard test method for the determination of the elastic properties of materials at ambient temperatures.[2] With this technique, a beam-shaped test sample is excited by a mechanical impulse, its vibration is captured, and the digitised vibration signal is processed with the RFDA software (IMCE, Diepenbeek, Belgium). This software assigns a vibration of the form $x(t) = x_0 e^{-kt} \sin(2\pi ft + \phi)$ to a predefined number of resonant frequencies. An algorithm simulates the measured signal as a sum of these transient sinusoidal signal components, optimising iteratively the initial amplitude (x_0), the exponential decay (k), the resonant frequency (f) and the phase (ϕ) of every signal component. Having reached a predefined convergence criterion, the software provides the resonant frequencies and associated k values of all the measured vibration modes. An internal friction (Q^{-1}) value is calculated from each k, based on relation $Q^{-1} = k/\pi f$. Finally, the elastic material properties are derived from the resonant frequencies of the fundamental flexural and torsional vibration modes, using the identification formulas of the ASTM standard E 1876-99.[3]

The A, AZ and AZA laminates were first excited in both out-of-plane flexural and torsional vibration at room temperature. Subsequently, all three laminates were excited in out-of-plane flexural vibration at high temperatures in nitrogen (N_2); the employed heating cycle – the aim of which was to simulate as close as experimentally possible the laminate sintering cycle – was the following: heat at 5 °C/min to 1100 °C → heat at 1 °C/min to 1550 °C → dwell for 5 min at 1550 °C → cool at 1 °C/min to 600 °C → cool at 5 °C/min to room temperature.

2.3. Finite element modelling

As mentioned in Section 2.2, IET was used to identify the properties of the A and AZ laminates: their Young's modulus (E) and internal friction were measured as a function of temperature, while their shear modulus (G) and Poisson's ratio (ν) were only measured at room temperature. Next, a finite element model (FE-model) of the AZA laminate was constructed, where the input elastic properties were the ones identified from the pure A and AZ laminates. The FE-model was made using brick elements with quadratic shape functions, and assumed a perfect bonding between the constituent layers. The mesh density of the model was determined with a convergence analysis. This analysis revealed that the computation of the fundamental flexural frequency of the sample required a model size of $15 \times 1 \times 9$ elements. In order to validate this FE-model, the computed resonant frequency of the fundamental flexural mode was compared with the measured one; the validation of the FE-model was performed over the temperature range of interest (i.e. from room temperature to 1550 °C), at temperature intervals of 100 °C. It must also be noted that the resonant frequencies of a beam-shaped sample are not influenced by the residual stresses in the material.

3. Results and discussion

Table 2 presents the room temperature elastic properties (E, G and ν) and internal friction (Q^{-1}) of the A and AZ laminates as determined by IET. Figs. 1–3 present the output of the high-temperature IET tests for the A, AZ and AZA laminate, respectively; obviously, the fundamental flexural frequency of all three laminated specimens decreased with temperature, in

Table 1
Mass and dimensions of the A, AZ and AZA test specimens (rectangular bars)

	Mass (g)	Thickness (mm)	Width (mm)	Length (mm)
A laminate	0.808	1.758	2.767	42.344
AZ laminate	1.035	1.727	3.136	40.424
AZA laminate	0.953	1.788	3.019	41.988

Table 2
Room temperature elastic properties and internal friction of the A and AZ laminates

	E (GPa)	G (GPa)	ν	Q^{-1}
A laminate	391.8	157.8	0.24	0.00005
AZ laminate	306.5	121.9	0.26	0.00023

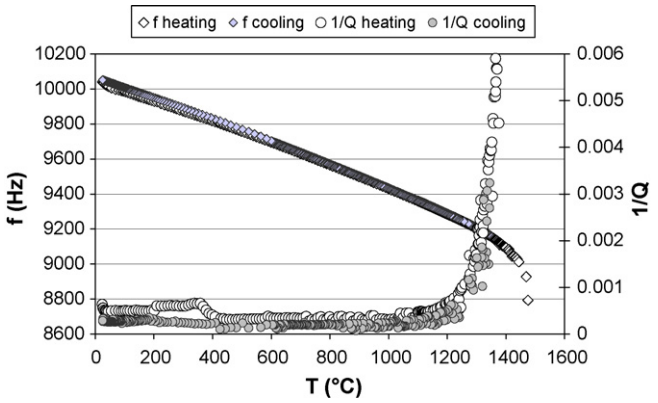

Fig. 1. Plot of the fundamental flexural frequency f and internal friction Q^{-1} of the A laminate as a function of temperature.

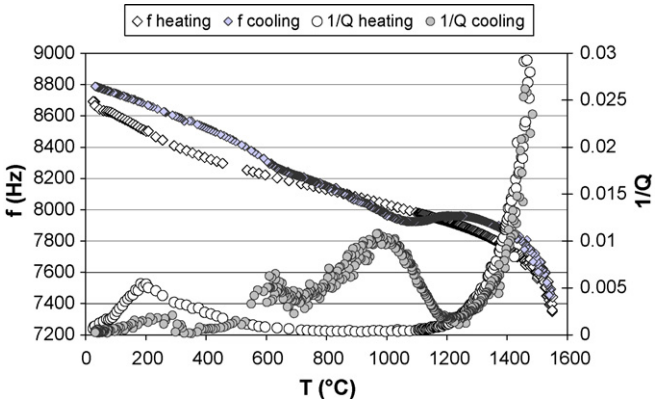

Fig. 2. Plot of the fundamental flexural frequency and internal friction of the AZ laminate as a function of temperature.

contrast with the internal friction that increased abruptly above ~1200 °C. The frequency values of Figs. 1 and 2 were used to calculate the Young's modulus of the A and AZ laminates as a function of temperature; the results of these calculations are presented in Fig. 4. As may be seen from Fig. 4, the exposure of laminates A and AZ to high temperatures caused additional sintering, which lead to an increase of 0.24% and 2.79% in the room temperature E of the A and AZ laminate, respectively.

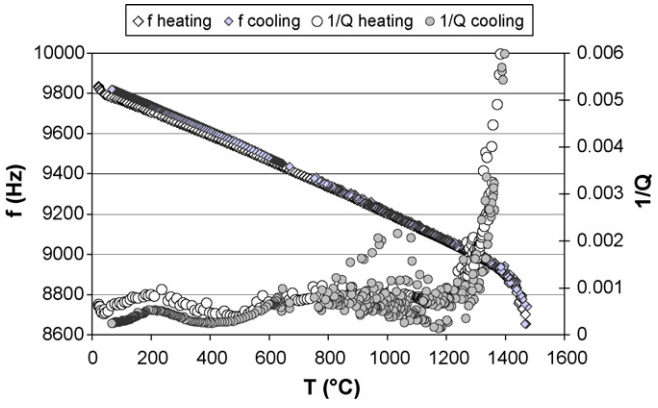

Fig. 3. Plot of the fundamental flexural frequency and internal friction of the AZA laminate as a function of temperature.

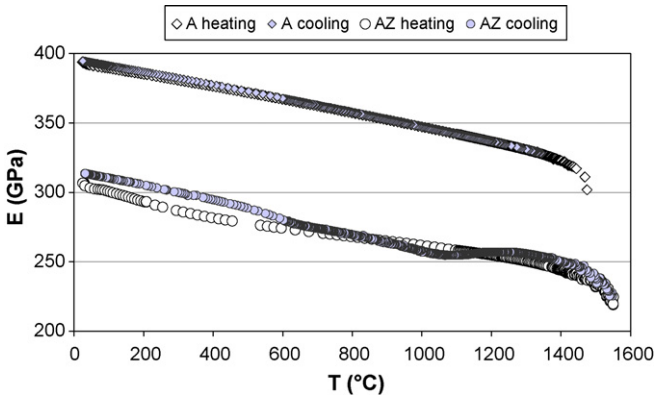

Fig. 4. Plot of the Young's modulus E of the A and AZ laminates as a function of temperature.

Very interesting is the plot of the internal friction of the AZ laminate as a function of temperature (Fig. 2). One may immediately notice the presence of two distinct internal friction peaks: one that forms during cooling at ~970 °C, and another that forms both during heating and cooling at ~210 °C. These two peaks may be related to phase transformations in the 3 mol% yttria-stabilised zirconia ceramic (3Y-TZP), which represents 40 vol.% of the AZ laminate; the peaks are also present but less prominent in the internal friction curve of the AZA laminate (Fig. 3) due to the smaller volume fraction (15.4 vol.%) of the 3Y-TZP constituent material. The first Q^{-1} peak may be attributed to the transformation of tetragonal zirconia (t) to monoclinic (m) zirconia, which occurs during cooling at ~950 °C in pure zirconia, being accompanied by a shear strain of ~0.16 and a volume expansion of ~4%.[4] It is already known that the martensitic $t \rightarrow m$ phase transformation starts at ~1170 °C during the cooling of pure zirconia,[2] so it is not surprising that the related damping peak begins close to that temperature in the case of the AZ laminate. The exact nature of this damping peak has not yet been perfectly understood, but it has been attributed to energy-dissipating grain boundary movements.[2] The onset

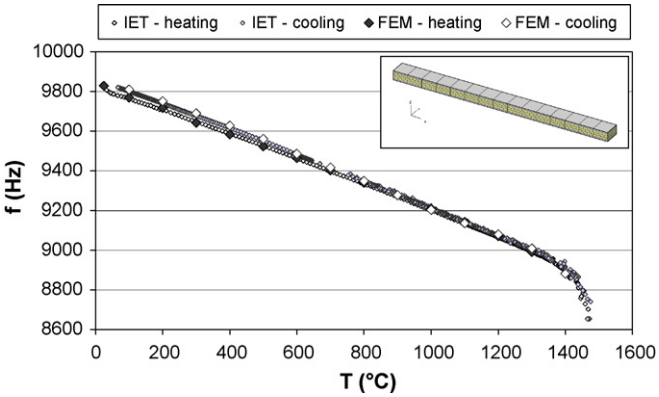

Fig. 5. Comparison between the IET-measured (IET) and the FE-model-predicted (FEM) fundamental flexural frequency of the AZA laminate as a function of temperature. The inset drawing shows the FE-model of the AZA laminate.

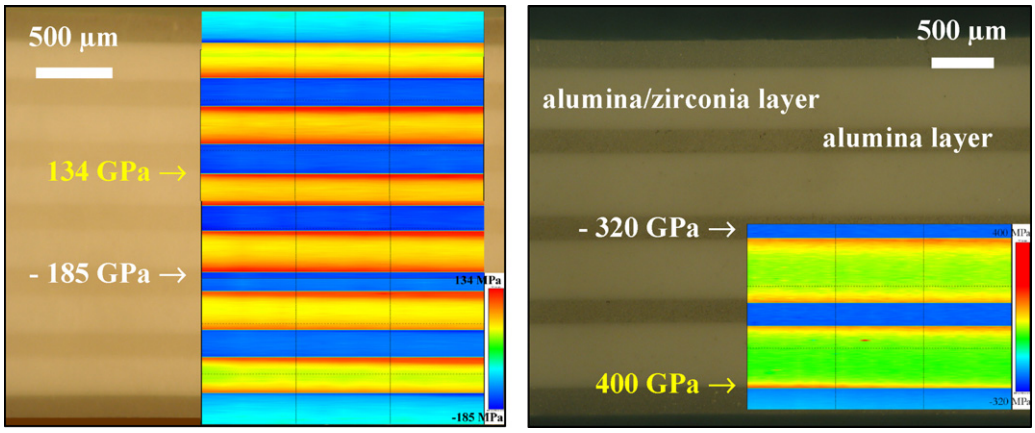

Fig. 6. Residual stress maps constructed by a piezo-spectroscopy technique for two different AZA laminates.

of the $t \rightarrow m$ phase transformation is also accompanied by a decrease in the fundamental flexural frequency of the AZ sample (Fig. 2), and, consequently, by a decrease in the material's E (Fig. 4). The deterioration of the material's elastic modulus might be related to the formation of microcracks within the 3Y-TZP constituent of the AZ laminate; in fact, microcracking has been reported to accompany invariably the volume-expanding $t \rightarrow m$ phase transformation.[2]

The second Q^{-1} peak that forms during the cooling of the AZ laminate can be attributed to one of the following two phenomena: (a) an elastic dipole relaxation that has also been reported in 3Y-TZP ceramics by Roebben et al.,[2] and/or (b) a degradation of the 3Y-TZP ceramic due to its exposure to moist air.[4] The first phenomenon attributes the damping peak to the movement of oxygen vacancies around immobile yttrium substitutional cations, while the peak height (i.e. the maximum damping value) is determined by the Y^{3+} content of the zirconia lattice.[5] The oxygen vacancies and yttrium cations are believed to form an elastic dipole that reacts to the application of stress at ~200 °C in the following way: the thermally-activated oxygen vacancies move to positions of minimum strain energy. This relaxation effect has been reported to occur at 217 °C in monolithic 3Y-TZP ceramics,[2] which is very close to the temperature determined in this study for the 3Y-TZP-containing AZ laminate. The second phenomenon attributes the damping peak to the degradation of the YSZ ceramic due to its exposure to moist air or hot water. This degradation has been reported to occur at 230 °C and is related to the leaching of Y_2O_3, which leads to the moisture-triggered destabilisation of the t phase and the transformation of its surface layers to the m phase.[4] Even though the high-temperature IET test on the AZ laminate was performed in N_2, the presence of some moisture inside the IMCE furnace cannot be excluded.

Fig. 5 compares the measured and computed resonant frequencies of the AZA specimen as a function of temperature. It is obvious that there is an excellent agreement between the IET-measured flexural frequencies and the frequencies predicted by the FE-model over the whole temperature range of interest. This validates the input material parameters of the FE-model, which will be used in the next step of this investigation to simulate the development of residual stresses in the AZA laminate as a function of temperature.

4. Future work

Preliminary residual stress measurements have been performed on alumina–alumina/zirconia laminates comparable to the ones addressed in this study: these laminates were produced with the same starting materials and processing route, but had different layer thicknesses and stacking sequence (Fig. 6). The measurements were done using a piezo-spectroscopy technique at the Research Institute of Nanoscience (RIN, Kyoto, Japan), and will be compared to the residual stress values predicted by finite element modelling. Moreover, high-temperature IET tests will be performed on 'green' laminates, so as to investigate whether it is possible to arrest the development of residual stresses by following the changes in the internal friction of the studied laminates.

5. Conclusions

This study determined the temperature dependence of the elastic properties and damping behaviour of alumina (A) and alumina/zirconia (AZ) laminates. The determined elastic properties are necessary for the calculation of residual stresses in alumina–alumina/zirconia (AZA) laminates using finite element modelling; these residual stresses will be computed as a function of temperature in the next step of the investigation.

Acknowledgment

The authors acknowledge the financial support provided through the European Community's Human Potential Programme under contract HPRN-CT-2002-00203, [SICMAC].

References

1. Toschi, F., Melandri, C., Pinasco, P., Roncari, E., Guicciardi, S. and de Portu, G., Influence of residual stresses on the wear behavior of alumina/alumina-zirconia laminated composites. *J. Am. Ceram. Soc.*, 2003, **86**(9), 1547–1553.
2. Roebben, G., Basu, B., Vleugels, J. and Van der Biest, O., Transformation-induced damping behaviour of Y-TZP zirconia ceramics. *J. Eur. Ceram. Soc.*, 2003, **23**, 481–489.
3. Standard test method for dynamic Young's modulus, shear modulus, and Poisson's ratio by impulse excitation of vibration. ASTM standard: E 1876-99, July 1999.
4. Hannink, R. H. J., Kelly, P. M. and Muddle, B. C., Transformation toughening in zirconia-containing ceramics. *J. Am. Ceram. Soc.*, 2000, **83**(3), 461–487.
5. Weller, M. and Schubert, H., Internal friction, dielectric loss, and ionic conductivity of tetragonal ZrO_2 – 3% Y_2O_3 (Y-TZP). *J. Am. Ceram. Soc.*, 1986, **69**(7), 573–577.