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Characterization of residual compressive stresses in layered ceramics by positron annihilation spectroscopy

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Abstract: Microstructural characterization of stress states of alumina layers subjected to residual compressive stresses was performed by positron annihilation spectroscopy (PAS). Al₂O₃-t-ZrO₂ three layered samples were appropriately designed and processed by sequential slip casting with symmetric structure. The zirconia contents inside the core layer were selected in order to generate compressive stresses of variable intensity in the outer Al₂O₃ layers. PAS results highlighted a correlation between positron annihilation parameters and microstructural changes related to the generation of residual stresses.

Keywords: Residual stress; Al₂O₃; Positron annihilation spectroscopy

1. Introduction

Residual stresses greatly affect the structural performance of advanced ceramic materials, for this reason their characterization is of key importance to define the properties of a ceramic component.¹ Information on residual stresses, besides being useful in the study of the microstructure of materials, provides important data to optimize their performance and reliability.

Although various techniques are used to study the presence of residual stresses in ceramics, these methods have limitations for a simple and comprehensive evaluation. The classical theory of elasticity or finite element method (FEM) often do not provide the real final residual stresses, as some data can be missed in the initial considerations about the materials. Results provided by indentation tests, widely employed to perform direct measurements in coatings and multilayers of traditional and advanced ceramics, may be unreliable due to the difficulty of measuring

the effects of the residual stresses generated and quantifying the magnitude of residual surface stresses. Piezo-spectroscopic methods are usually applied to local stress measurements.²⁻⁸ Also, low angle X-ray diffraction (XRD) measurements give an estimation of the residual stresses in a range of a few microns beneath the sample surface.⁹ Nevertheless, it is well known that stresses at this distance are not representative of the whole stress state as the presence of the free outer surface is an important condition to take into account.

To measure residual stresses inside the material or as a function of depth, successive X-ray diffraction measurements have to be performed, following layer removal and applying subsequent correction of the effect of removed material that is difficult to estimate with accuracy. Penetrating non-destructive technique using neutron sources and synchrotron radiation provide measurements of bulk residual stresses^{7,8} and their profiling.^{10,11} However, these high energy methods, although widely used, require big installations and complex equipments.

Positron annihilation spectroscopy (PAS) is a non-destructive technique used to study the presence of defects in a material and their nature,¹² and it is based on the analysis of the annihilation radiation of electron-positron pairs in a sample. When a

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positron from a β^+ decay of a radioactive isotope source goes into a solid, it diffuses in the material and, after a time of the order of ~ 100 ps and getting a penetration depth of the order of ~ 100 μm , it annihilates with an electron emitting two photons of 511 keV. The analysis of the annihilation radiation provides information on the concentration of defects in the material and their nature. Indeed, in the presence of open volume defects, the local electron density and the local electron momentum distribution in the material vary respect to the crystal free of defects. When the positron annihilates in an open volume defect, the energy and momentum of the annihilation photons and positron lifetime change in a characteristic way.

Two methods of positron annihilation measurements are of particular concern for investigating defects in the bulk of a material: positron annihilation lifetime spectroscopy (PALS) and coincidence Doppler broadening (CDB).

PALS is based on the measure of positron lifetime in the material, and results reflect the local electron density at the annihilation site and give information about the size of the defects and their concentration in the material. The CDB measurements of the annihilation radiation provide data about the momentum distribution of electrons taking part in the positron–electron annihilation and lead information on the electronic structure of materials and the chemical surroundings of the annihilation sites.¹³

In the present work positron annihilation spectroscopy (PAS) is proposed as an accessible and simple technique to investigate residual stresses in ceramics, based on the correlation found between the annihilation parameters and microstructural changes related to the stress states which the material is subjected. Positron annihilation experiments were performed on Al_2O_3 layers using Al_2O_3 – t-ZrO_2 layered samples properly designed to test the ability of PALS and CDB techniques to provide information on microstructural characteristics associated with the stress state of ceramic materials.

Layered ceramics are a suitable system for generating different levels and distribution of residual stresses in the ceramic components. Many intrinsic and extrinsic features^{11–13} can modify the residual stress state, but the most common approach is associated with differences in thermal expansion coefficients between adjacent layers. The stress field in the laminates can be appropriately controlled designing the thickness, composition and sequence of the layers.^{14–16} It has been proved that compressive stresses on surface layers may improve mechanical strength and increase the fracture toughness, as the state of compression is able to impede surface cracking.^{15,17,18} On the other hand, if compressive residual stresses are induced in the inner layers, tolerance to damage and, therefore, reliability are promoted.^{14,19}

In the present study laminated samples were properly tailored with a symmetric structure, i.e. two Al_2O_3 external layers and the middle one consisting of a composite of Al_2O_3 with variable amounts of stabilized tetragonal zirconia (t-ZrO_2). Differences in thermal expansion coefficients of the layers provided tensile–compressive states during cooling of sintering, giving alumina external layers in compression, and the internal one in a tensile state. The thickness of the layers was designed to ensure

high residual stresses, and the annihilation of all positrons in the alumina layers to be investigated.

2. Materials and methods

Laminated materials consisting of three layers were appropriately designed: the external ones composed of pure alumina with thicknesses of 400 μm and the inner layer, 4 mm thick, made of a composite of alumina with variable amounts (30, 40 and 50 vol.%) of t-ZrO_2 stabilized with 3 mol% yttria (3Y-TZP). The 1:10 ratio between the thicknesses of the layers allowed developing an intense compressive stress field in the external layers and a less intense tensile stress field in the intermediate layer. Considering that the implantation range of ^{22}Na positrons is about 200 μm in ceramics,²⁰ and the diffusion length for the thermalized positrons even less of 200 μm ,²¹ the thickness of 400 μm for the outer layers was chosen to ensure that the annihilation of positrons was practically accomplished in the external Al_2O_3 layers adjacent to the ^{22}Na positron source. Then, any contribution from annihilations in the inner layer and at the layer interface would be meaningless.

The samples were fabricated by using the technique of sequential slip casting, a versatile and suitable method to make multilayered materials.²²

The starting powders, used to produce the samples, were $\alpha\text{-Al}_2\text{O}_3$ (Condea HPA 0.5, USA), and TZ-3YS (Tosoh, Japan), hereafter respectively denoted by A and TZ. A–A(TZ)_{X vol.%} denoted the three-layer Al_2O_3 – t-ZrO_2 samples.

Suspensions, containing 38 vol.% of solid in deionized water and 0.8 wt% of an acrylic-based polyelectrolyte (Duramax D-3005, Rohm & Haas, USA) as dispersant, were prepared by ball milling. The percentage of dispersant was chosen on the basis of preliminary studies.^{23–25}

The shaping stage of the samples was preceded by the study of the casting kinetic of each suspension to obtain the thickness of the layers as a function of the casting time.²²

70 mm \times 70 mm plates were slip cast on a plaster mould. Green bodies were pre-sintered at 900 °C for 1 h, cut into square samples ~ 10 mm \times 10 mm, and then sintered at 1500 °C for 2 h. The heating and cooling rates were of 5 °C min^{-1} .

The density of the sintered samples, measured by both the Archimedes and the helium pycnometry method, was found to be $\sim 98\%$ of the theoretical density.

The phases present in the sintered samples were determined by X-ray diffraction, carried out in a Philips PW 3830 NL diffractometer using $\text{Cu K}\alpha_{1,2}$ radiation. X-ray patterns revealed that only $\alpha\text{-Al}_2\text{O}_3$ and t-ZrO_2 phases were present.

The microstructure of the sintered samples was analyzed by scanning electron microscopy (SEM) (Zeiss DSM-950, Germany) on polished and thermally etched surfaces of the cross sections.

Positron annihilation measurements at room temperature were made using a ^{22}Na positron source of ~ 10 μCi sealed between two kapton foils (10 μm thick) and sandwiched between the outer Al_2O_3 surfaces of two identical samples.

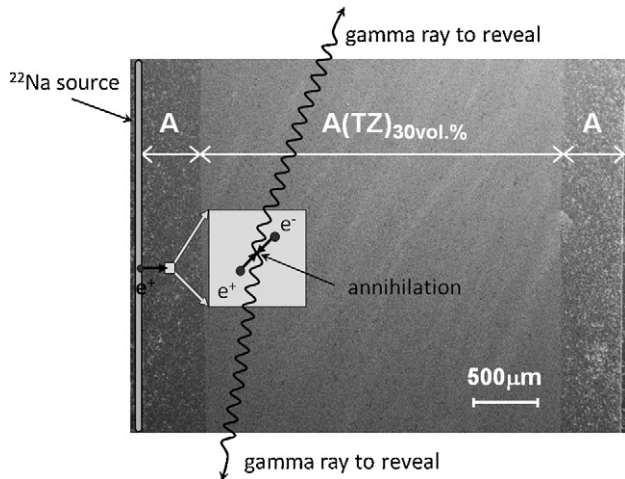


Fig. 1. SEM micrographs of the cross section of A–A(TZ)_{30 vol.%} sample. The sketch displays the positron–electron annihilation in the middle of the alumina layer.

Positron lifetime spectra were recorded with a conventional fast–fast coincidence spectrometer equipped with two NE111 plastic scintillators. CDB measurements were performed using two HP Ge detectors in timing coincidence. The technical details of the procedures for both the positron lifetime and CDB measurements can be found in Ref. 23.

3. Results and discussion

The intensity of the residual stresses was calculated following the classic theory of elasticity and using the corresponding values for the thermal expansion coefficients and elastic modulus reported in the literature, a Poisson coefficient of 0.3, and considering that residual stresses start to be developed at 1200 °C.^{1,5} The evaluation of the residual compressive stress in the outer alumina layers resulted in –477, –646, –1.035 MPa, while the tensile ones in the central A(TZ)_{X vol.%} layers were 95, 129 and 207 MPa for X = 30, 40, 50 respectively.

The SEM image of the cross section shown in Fig. 1 reveals a well defined interface and good adherence between the layers and no delamination between adjacent layers. Moreover, no evidence of cracks was found in any of the layers of the A–A(TZ)_{X vol.%} samples for X = 30 and 40. However, two cracks in the alumina layers of the A–A(TZ)_{50 vol.%} sample were observed at the apexes of a transverse fracture running through the whole central layer, as Fig. 2 shows. This fracture is attributed to the intense tensile state developed in the central layer due to its high zirconia content. Although this fracture appears to be blocked at the interfaces, cracks still enter into the alumina layers and propagate up to halt at around the middle of the layers as Fig. 2 reveals. The appearance of the fracture in the A(TZ)_{50 vol.%} layer and the crack propagation in the A layers cause the relaxation of the tensile–compressive state.

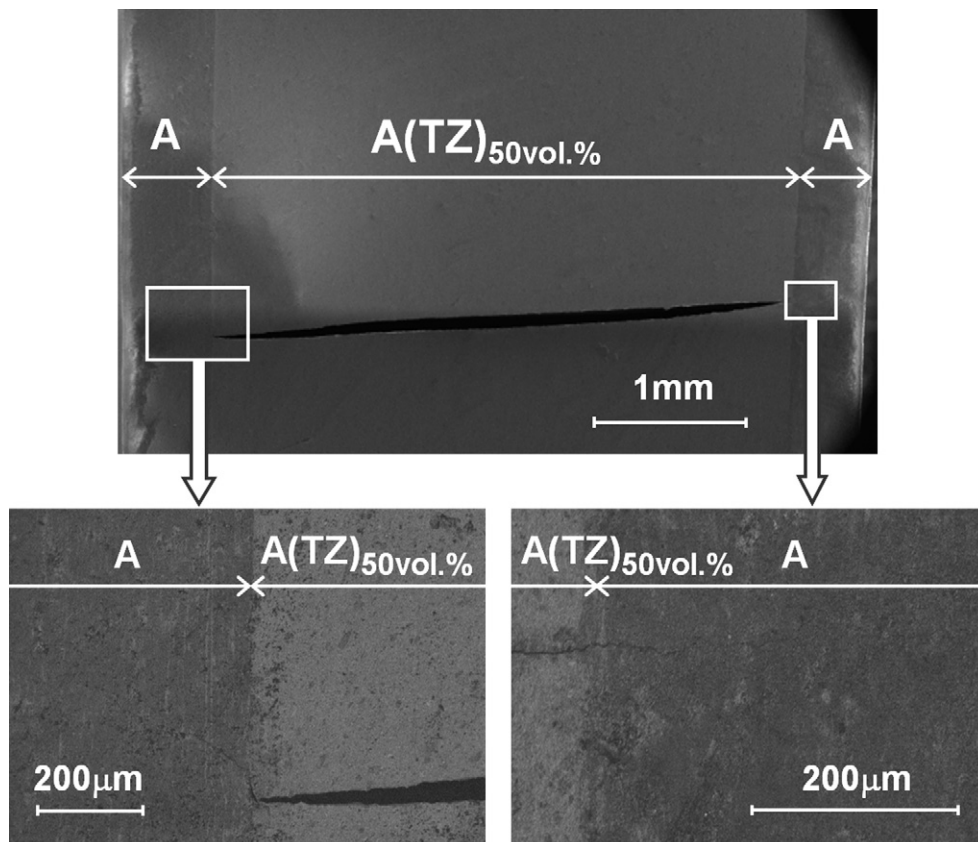


Fig. 2. SEM micrographs showing the fracture in the middle layer of the A–A(TZ)_{50 vol.%} sample. The detail micrographs on the bottom point out the crack propagating in Al₂O₃ layers.

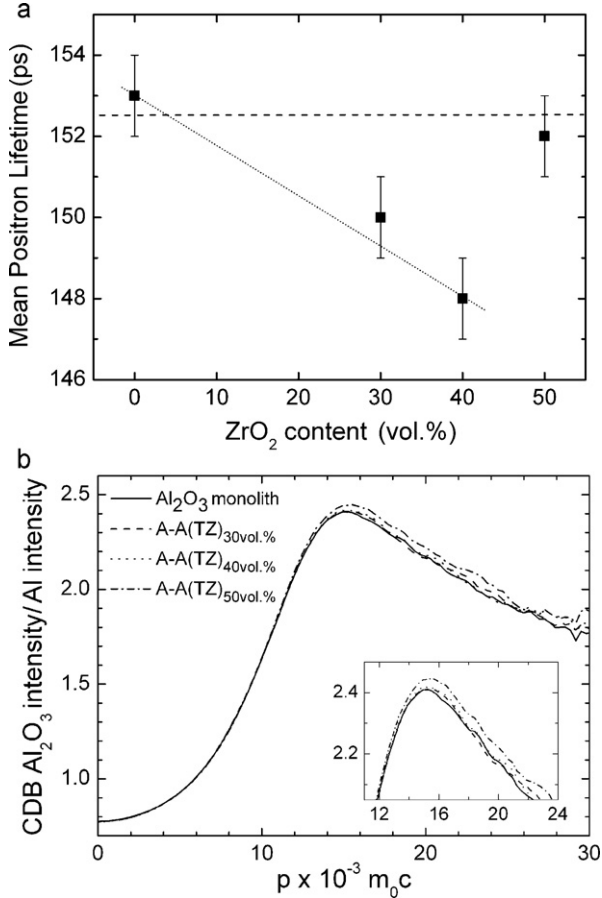


Fig. 3. Representation of PAS measurement results. (a) Mean positron lifetime as a function of the ZrO₂ content in the central layer A(TZ)_{X vol.%}. The lines are a guide for the eyes. (b) CDB ratio spectra for the whole A-A(TZ)_{X vol.%} series. The detail points out variations of intensity of the peak.

PALS and CDB techniques were applied to investigate the relation between the change in positron lifetime and CDB intensity and the microstructural changes related to compressive states of different intensity in alumina.

Comparison of results with those obtained in a previous study on alumina layers subjected to tensile stresses of variable intensity,²³ allowed to find a correlation between PAS parameters and microstructural changes related to residual stresses.

PALS measurements were performed and all positron lifetime spectra acquired were properly fit to only one component: the average lifetime, a statistically accurate parameter adequate to monitor changes in the lifetime spectra, as indicated by the small value of χ^2 (always <1.2) obtained when analyzing the lifetime spectra with the POSITRONFIT program.²⁶

The average positron lifetime values are given in Fig. 3a as a function of the t-ZrO₂ content in the central layer of A-A(TZ)_{X vol.%} samples. It can be observed a reduction of positron lifetime, from 153 ± 1 ps to 148 ± 1 ps, for a zirconia content lower than 50 vol.%. The positron lifetime for the A-A(TZ)_{50 vol.%} recovers the value corresponding to the monolithic sample. The decreasing trend of the positron lifetime is due to the compressive state that leads to a reduction of the crystal lattice parameters, causing an increase in the electronic density

of the material with an increase of packing of the atoms. In the A-A(TZ)_{50 vol.%} sample the fracture observed (Fig. 2) relaxes the residual compressive stresses in the outer alumina layers that recover the lattice parameter and atomic density values, and thus the average positron lifetime, of the monolithic sample (see Fig. 3a).

The present results were compared with those obtained in a previous PALS characterization of the Al₂O₃ tensile state in layered samples with a similar structure, containing monoclinic zirconia in the central layer.²³ Lifetime measurements on alumina layers under tension in alumina-monoclinic zirconia (m-ZrO₂) layered samples previously reported²³ showed that the average positron lifetime value increased with the addition of m-ZrO₂ to the middle layer. We linked this increase to the reduction of the electronic density¹² due to an expansion of the crystal lattice, associated with the most intense state of tension of alumina layer, and to positron annihilation in structural defects related to the presence of cracks propagating from the surface. In particular, we stated that for these materials aluminum mono-vacancies V_{Al} and more complex defects, such as divacancies ($V_{Al}-V_{Al}$), (V_O-V_{Al}) or complex vacancies – impurities, contributed to the increase in the average lifetime value.²³

CDB measurements were performed to obtain information about the chemical surrounding of the positron annihilation sites. The CDB spectra from the Al₂O₃ layers in the A-A(TZ)_{X vol.%} samples are shown in Fig. 3b. They represent the ratio of the number of positron annihilating in alumina respect to the aluminum reference sample, versus the momentum shift of electron-positron annihilation pairs.

It can be observed that all spectra show a peak centered at $\sim 15 \times 10^{-3} m_0 c$, which is attributed to positron annihilations with valence electrons of O²⁻ ions.²⁷ The intensity of the CDB spectra in the low-momentum region is lower than 1, and there are not significant differences among the different ratio spectra. In the central region of the peak, there is no appreciable intensity change, except for the A-A(TZ)_{50 vol.%} sample which exhibits cracks in the Al₂O₃ layers. In our previous CDB analysis of tensile states in alumina²³ we observed that the intensity of the CDB peak increased with the ZrO₂ content in the presence of cracks, accompanied by an increase of the positron lifetime, in the presence of structural defect associated with O²⁻ ions, such as cation vacancies or dislocations induced by tensile residual stresses.

4. Conclusions

PALS and CDB measurements performed on alumina layers subjected to residual compressive stresses of different intensity allowed finding a correlation between spectroscopic parameters and microstructural changes related to residual stress fields generated during sintering. Positron lifetime and CDB spectra were acquired on external alumina layers of layered samples properly designed to be investigated by PAS. The data obtained were analyzed using the information supplied by SEM morphological observations. The present positron annihilation results, along with the ones for alumina layers subjected to internal tensile stresses reported in Ref. 23, reveal that mean positron lifetime

and CDB spectra intensity are sensitive to (i) the formation of microstructural defects associated with ion O^{2-} , and especially cation vacancies, and dislocations, and (ii) changes in crystal lattice parameters related to tensile and compressive states.

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