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Fracture mechanisms of structural and functional multilayer ceramic structures

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Abstract. The fracture of mechanically loaded ceramics is a consequence of material critical defects located either within the bulk or at the surface, resulting from the processing and/or machining and handling procedures. The size and type of these defects determine the mechanical strength of the specimens, yielding a statistically variable strength and brittle fracture which limits their use for load-bearing applications. In recent years the attempt to design bio-inspired multilayer ceramics has been proposed as an alternative choice for the design of structural components with improved fracture toughness (*e.g.* through energy release mechanisms such as crack branching or crack deflection) and mechanical reliability (*i.e.* flaw tolerant materials). This approach could be extended to complex multilayer engineering components such as piezoelectric actuators or LTCCs (consisting of an interdigitated layered structure of ceramic layers and thin metal electrodes) in order to enhance their performance functionality as well as ensuring mechanical reliability.

In this work the fracture mechanisms in several structural and functional multilayer components are investigated in order to understand the role of the microstructure and layered architecture (*e.g.* metal-ceramic or ceramic-ceramic) on their mechanical behaviour. Design guidelines based on experiments and theoretical approaches are given aiming to enhance the reliability of multilayer components.

Introduction

It is well known that ceramics are very brittle and their strength is very sensitive to the presence of defects [1-4]. This is made very clear in glass, which is one of the strongest manufactured materials when the surfaces are free from flaws (as with glass fibres), but under ordinary conditions "glass" is a synonym for fragility. The increased number of engineering design constraints, driven by the growing product requirements, as well as the greater range of advanced materials now available face the designer with complex choices for selecting a material to meet the performance of a particular system.

The demanding requirements for advanced devices have involved in many cases the combination of material classes (such as metals, ceramics and polymers) to fulfil the performance in a given system. In these cases, the fabrication of components having two different materials has been a challenge, not only from the viewpoint of the structural integrity (*i.e.* mechanical resistance) of the part but also from its functionality. For instance, the integration of electrodes into ceramic parts as in the case of multilayer piezoelectric actuators for a reduction of emissions in diesel engines requires a precise location of the interdigitated structure as well as high piezoelectric properties of the piezo-ceramic to fulfil the performance in service [5, 6] (the functionality of such devices is associated with the mechanical resistance of the ceramic layers; if cracks propagate between electrodes the device cannot fulfil its function anymore [7]). Another example is the case of Low Temperature Co-fired Ceramics (LTTCs), where the co-sintering of ceramic substrates and metal electrodes and vias at relative low temperatures (*i.e.* ca. 950°C) has enabled the improvement of wireless communication systems (*e.g.* mobile phones, GPS-technology) at relative low costs (in this case the mechanical reliability of the ceramic substrate conditions the performance limits of the microelectronic device) [8].

In many cases, the functionality of the system relies on the structural integrity of its constituents, in other words, the mechanical resistance of the part subjected to thermo-, mechanical- and eventually electrical loads. In this regard, the low mechanical reliability of ceramic materials along with their low resistance to crack propagation (*i.e.* catastrophic failure) has drawn the attention of engineers and material scientist in an attempt to improve their mechanical properties and thus enhance the performance of the system. Some of these issues have been overcome in high performance ceramics by combining ceramics with other ceramics, metals or polymers in layered or other composite configurations [10-12]. Such strategy has been inspired by nature, for instance the extraordinary toughness and strength of mollusc shells (Fig. 1), which are related to the fine-scale structure of the shell; a laminate of thin calcium carbonate crystallite layers consisting of 99% calcium carbonate ($CaCO_3$) and tough biopolymers, arranged in an energy-absorbing hierarchical microstructure [9]. The strength and toughness of such layered structures are significantly higher than their constituents, yielding for instance toughness values of one order of magnitude higher [13, 14]. Nevertheless, the replication of architectural features that are found in nature, at the micro and nano scales, into real macro scale structural engineering materials at a reasonable cost, is still a challenge. It is in fact necessary to build these high toughness architectural features by using more durable materials since they should possess additional functional and/or structural properties as, for example, high temperature stability and specific functionality. The concept of layered ceramics and functionally graded materials and coatings could allow tailoring of the surface and bulk properties of advanced engineering components with the purpose of enhancing their structural integrity as well as adding multi-functionality, which would translate into higher efficiency and better performance of these components.

Figure 1. Fracture of a mollusc shell [9].

In this work, experimental studies are presented on the mechanical behaviour of several ceramic-ceramic, ceramic-metal multilayer systems, where the influence of the microstructural architecture on the fracture response of the material is discussed aiming to optimise the design of structural and functional ceramic based components. The fracture behaviour of three different material systems have been investigated under applied bending stresses, using the four-point bending (4PB) [15] and/or the Ball-on-three-balls (B3B) testing methods [16, 17].

Multilayered ceramic structures

Layered ceramics with tailored residual stresses fabricated by sequential slip casting of stable suspensions have been investigated [18]. They are composed of 5 thick layers of alumina (named A) alternated with 4 thin layers of alumina-zirconia (referred to as B). The differential thermal strain between layers, due to the phase transformation of the zirconia particles during cooling down from sintering, promote alternated tensile-compressive biaxial residual stresses within the A and B layers, respectively. The magnitude of such residual stresses has been estimated by FE analysis of the cooling down from the sintering temperature. The thermo-physical properties of the layers were evaluated on monolithic specimens with the same layer composition and introduced in the model. The biaxial stresses resulted in about +100 MPa in the A layers and –650 MPa in the B layers, for a given composition and layer architecture [19]. It has been shown in previous works that the compressive stresses can enhance the resistance of the material against crack propagation in comparison to monolithic alumina based ceramics [19-23]. The resistance to crack propagation has been studied on such multilayer systems using indented specimens under four point bending. Figure 2 shows typical fracture behaviour of such layered structure. The fracture energy of the composite is increased through crack deflection/bifurcation within the compressive layers.

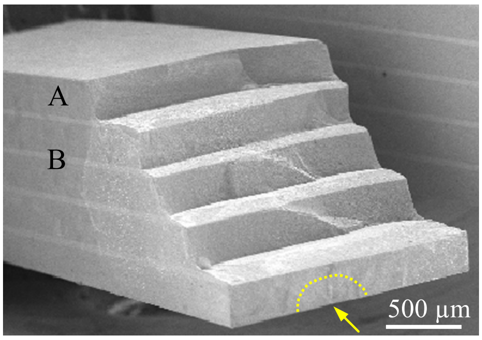


Figure 2. Step-like fracture of a layered ceramic where the initial crack is deflected through the compressive layers.

In some cases, propagating cracks can even be arrested at the interface of the compressive layers, yielding a minimum failure stress (*i.e.* higher mechanical reliability) below which the material does not fail. An example of this phenomenon is shown in Fig. 3, where through-the-thickness thermal shock cracks originated at the surface of the laminate propagated only through the first layer.

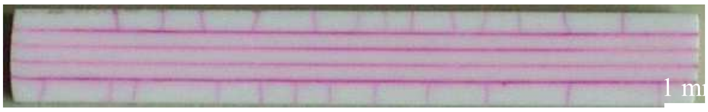
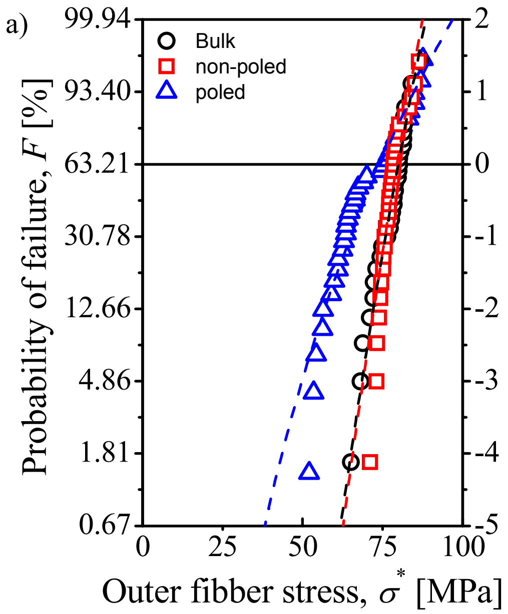


Figure 3. Thermal shock cracks arrest at the compressive layer of a laminate for a $\Delta T = 300$ °C [24, 25].

Piezostacks

Modern multilayer piezoelectric actuators used in advanced injection systems consist of a stack of piezoceramic layers (*e.g.* PZTs) with interdigitated metallic electrodes in between. The main feature of the actuator's functionality is associated with the domain switching process triggered by an electrical field (ferroelectricity) and/or mechanical loading (ferroelasticity). However, the mechanical stresses occurring during service of these electro-mechanical converter components can lead to failure of the device [26]. Information about the strength of the pure ceramic and the multilayer structure is necessary to assess the performance limits of such actuators. Four point bending tests have been performed on (i) non-poled bulk ceramic specimens, (ii) non-poled and (iii) poled multilayer metal-ceramic composites, to assess the influence of the microstructure on the strength of the material. Figure 4 shows the strength distribution vs. the probability of failure of the three different structures, represented in a Weibull diagram. Due to the non-linear behaviour of PZTs during bending, the true outer fibber stress, σ^*, has been estimated from the elastically calculated bending stress, σ_e, according to *Fett et al.* [27].



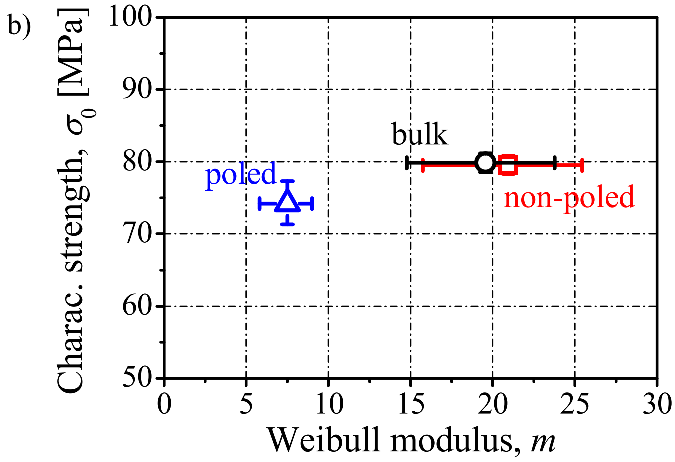


Figure 4. a) Strength distribution vs. probability of failure in a Weibull diagram and b) characteristic strength vs. Weibull modulus of (i) non-poled bulk ceramic specimens, (ii) non-poled and (iii) poled multilayer metal-ceramic composites.

It can be observed that the flexural strength and reliability is not affected by the presence of electrodes (compare (i) with (ii)). However the poling state of the material does affect its strength distribution. A fractographic analysis of broken specimens can be seen in Fig. 5 corresponding to the three different samples [28]. It can be observed that the lower Weibull modulus in the poled specimens (iii) is associated with the coexistence of both inherent natural flaws and relative large cracks (see close-up in Fig. 5c) generated during the poling operation.

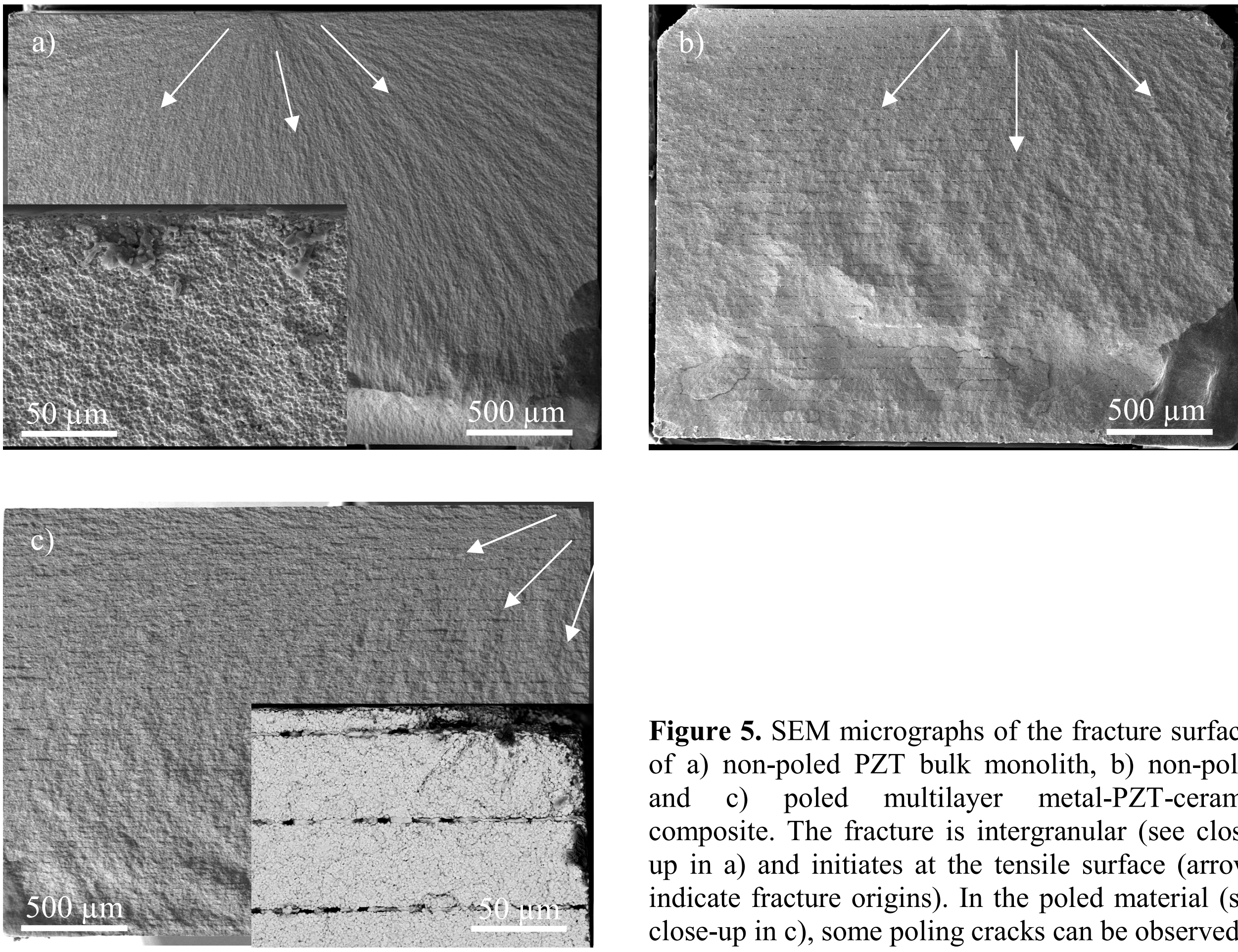
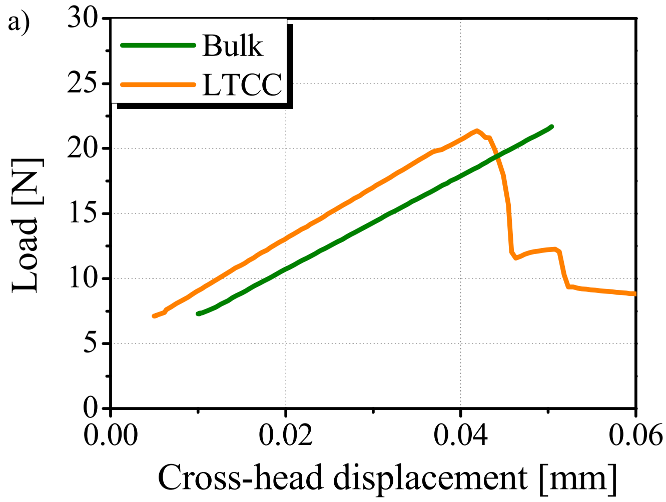


Figure 5. SEM micrographs of the fracture surfaces of a) non-poled PZT bulk monolith, b) non-poled and c) poled multilayer metal-PZT-ceramic composite. The fracture is intergranular (see close-up in a) and initiates at the tensile surface (arrows indicate fracture origins). In the poled material (see close-up in c), some poling cracks can be observed.

Low Temperature Co-fired Ceramics

LTCCs are layered ceramic based components, which consist of a complex three-dimensional micro-network of metal structures embedded within a glass-ceramic substrate. The low sintering temperature in LTCCs (*i.e.* below 950 °C) can be achieved by using a glass matrix with a low melting point, allowing a liquid phase sintering of the glass ceramic composite material [29]. The metallisation both at the surface and within the part can influence the strength of the component. This effect has been investigated using the ball-on-three-balls tests, where special locations of the LTCC, where the internal architecture may differ from place to place, have been tested under biaxial bending [30]. Results have been evaluated using Weibull statistics and a fractographic analysis of broken specimens has been performed to determine the mode of fracture of the components and the influence of the internal architecture (*i.e.* metal-electrodes, pads, ceramic layers) on the mechanical reliability of the LTCC part. In Fig. 6 the fracture features of a LTCC component tested using the B3B test are shown. A step-wise fracture (similar to the case of multilayered ceramics) can be observed (Fig. 6a) in comparison to the brittle and catastrophic failure of a bulk glass-ceramic taken as reference. The maximal applied stress is distributed in a small region of the specimen (see Fig. 6b) in order to test a specific location of the part. In Fig. 6c the cross-section of the fracture surface corresponding to the specimen plotted in Fig. 6a is presented. It can be appreciated how the metallization inside the LTCC influences the crack path. This could be used as a mechanism to increase the resistance to crack propagation in functional ceramics.



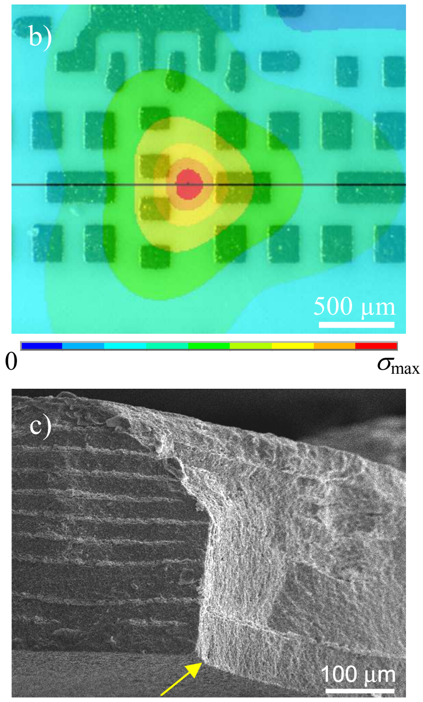


Figure 6. a) Load vs. cross-head displacement curves of a bulk glass-ceramic and a LTCC with metallization; b) Maximal stress distribution during B3B test at the tensile surface of a LTCC; c) Cross-section of a fracture LTCC after B3B test (arrow indicates fracture origin).

Outlook

The experimental findings reported above are only examples of layered structures which are conceived in different ways according to the end application. The know-how gained in ceramic-ceramic and ceramic-metal systems such as those mentioned above, in terms of fracture response under bending, could be applied to other multilayer devices where the layered architectural design concept is also available; systems that could be used as functional components and where the life time performance may be significantly enhanced through a tailored multilayer design.

The understanding of the conditions under which energy dissipating mechanisms such as crack deflection and/or interface delamination occur and the influence of the layered architecture (*e.g.* geometry, composition of layers, elastic and physical properties of the layers) on the crack propagation must be assessed in order to obtain tougher and more resistant materials. In this regard, the benefits of high failure resistance materials designed with weak interfaces and the strength reliability enhancement (flaw tolerance) of residual stress based materials designed with strong interfaces could be combined in a unique multilayer architecture [31]. For instance, weak interfaces in piezo-actuators could act as locations which would favour crack propagation, thus avoiding the propagation of cracks between electrodes (short circuit). In other applications involving relatively aggressive environments (*e.g.* high relative humidity) degradation phenomena such us Subcritical Crack Growth (SCG) can lead to failure of the component well under the predicted strength of the material, lowering then its mechanical reliability [32, 33]. The design of such components with “protective” layers (*e.g.* a compressive layer to stop the propagation of subcritical propagating cracks) could increase its life time and reliability.

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