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Subcritical crack growth in multilayer Low Temperature Co-fired Ceramics designed with surface compressive stresses

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ABSTRACT

The strength of Low Temperature Co-fired Ceramics (LTCC) can be affected by the environmental conditions under which the material is loaded. In this work, the strength degradation associated with Subcritical Crack Growth (SCCG) mechanisms is investigated in several multilayer LTCC architectures designed with surface compressive residual stresses. The magnitude of the residual stresses was tailored combining two different LTCC materials. Biaxial strength measurements using the ball-on-three-balls method performed at room temperature in water (as reference environment) showed a clear increase in the characteristic strength with the compressive residual stress in the surface layer. The strength distribution in dependence of the surface stresses in the outer layer can be represented by a three-parameter Weibull distribution, thus providing a "threshold strength" (i.e. minimum strength) for the material. In addition, the use of compressive stresses in LTCCs introduces a threshold intensity factor for the SCCG behaviour, below which no environmental assisted cracking can occur.

1. Introduction

Low Temperature Co-fired Ceramics (LTCC) designates a group of ceramic materials where sintering temperatures below 900 °C can be achieved [1] making the co-sintering with good conducting metals like silver, copper or silver-palladium-alloys feasible. A special case of LTCC is substrates where crystalline ceramic (e.g. alumina) grains are embedded in a glassy matrix. In general, LTCCs are made by laminating green ceramic-based tapes together with screen-printed internal metallization (e.g electrodes) and vias as connection between different planes. LTCCs with internal 3D metal structures can be utilized as functional components and used as ceramic circuit boards, for instance in mobile phones or as WLAN-, Bluetooth-, or RADAR-antennas, as well as in biomedical sensors and devices [2,3]. The LTCC technology provides components with improved thermal and mechanical stability (in terms of stiffness and shape) compared to the widely used polymer-based printed circuit boards (PCB) technology. Therefore, ceramic circuit boards are often used in harsh environments such as elevated temperatures, high mechanical loading and at heavy vibrations. Consequently, high strength and mechanical reliability of LTCC-components are mandatory.

Important mechanical failure mechanisms of LTCCs are (i) brittle fracture and (ii) subcritical crack growth (SCCG). In both cases cracks start at small flaws in the microstructure (e.g. large crystalline grains, pores, surface defects such as scratches), which can be described as cracks in the framework of the linear elastic fracture mechanics. In the first case (brittle fracture) when the so-called stress intensity factor (SIF) at the crack tip, K_I, reaches the fracture toughness, K_c, of the LTCC material, the crack grows almost at the speed of sound (i.e. fast fracture). The second case (SCCG) is common in many ceramic materials containing a glassy phase. SCCG precedes brittle fracture in a loading region where the SIF is smaller than K_c. The SCCG of cracks needs time and may cause delayed failure of components. Note that at small loadings the crack growth rate may become very slow (rates as small as 10^{-8} m/s have been measured [4]). Of course the lifetime of LTCC components depends on the SCCG-rate.

Microstructural features such as the glass phase content (amorphous or crystallized), ceramic particle size, shape and distribution, can affect the fracture toughness and SCCG behaviour [5]. Strength as well as lifetime can be increased by higher fracture toughness.

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But the size of the fracture initiating flaws (fracture origins) has also a significant influence on strength as well as on SCCG-rate. Since the size of critical flaws differs from specimen to specimen (or component to component) the strength and lifetime also differ. The scatter of strength is described in general by a two-parameter Weibull distribution [6].

Increasing strength in brittle components can be attained by reducing the size of critical defects (e.g. through colloidal processing) [7], or by introducing compressive residual stresses at the surface (e.g. strengthening in glass such as Gorilla® glass [8]). Both approaches have been successfully exploited in many different structural ceramic systems (see for instance [9–15]). In laminate ceramics, the use of different materials in the layers (having a different coefficient of thermal expansion, CTE) generates residual stresses, which may be used to improve the mechanical properties of the laminates. However, to the authors' knowledge, no attempts have been made up to date to tailor the mechanical properties of LTCCs through a layered architecture. In this work, a novel strategy to increase the strength and lifetime of LTCC substrates is presented. Multilayer architectures with compressive residual surface stresses were produced by combining two LTCC materials with similar composition but having fillers with different shapes. Thickness and arrangement of the different layers is used to tailor the residual stresses. The most important parameter is the volume ratio of the layer materials. The strength distribution is described by a three-parameter Weibull distribution and the (two parameter) single power law used to describe SCCG-rates transforms into a three parameter law. Special focus is given to the effect of surface compressive residual stresses on the subcritical crack growth parameters. It is shown that the compressive residual stress in the surface layer introduces a lower bound for the strength as well as a threshold for the SCCG of the LTCC material.

2. Theoretical background

2.1. Residual stresses in ceramic laminates

Laminates are produced by laminating and sintering green ceramic sheets. If the layers consist of different materials, the thermal shrinking during cooling down to room temperature is also different in layers of different materials. One of the reasons is a different CTE of the different layer materials, but other reasons, e.g. phase transformations, may also apply. Without loss of generality we will restrict in this paper to the action of different CTEs; other effects can simply be accounted by replacing the CTE mismatch by the strain mismatch.

We analyse laminates that are free from bending after the sintering process. Symmetrically layered laminates (in relation to the mid plane), as the ones investigated here, fulfil this restriction intrinsically. We assume that the layers sinter together and that strong interfaces are built between the layers irrespective whether or not they consist of the same material. If some stress relaxation occurs (e.g. caused by diffusive processes), the reference temperature, Tref, below which residual stresses occur, is in general lower than the nominal sintering temperature, Tsint. For instance, the glass transition temperature, Tg, in this type of LTCC materials has been determined to be ca. 750 °C in previous studies, which may be considered as the onset for residual stress development in the composite material. In practice the reference temperature has to be determined by additional means, e.g. by stress measurements using X-ray diffraction or Raman scattering, which is beyond the scope of this work. Therefore, we will further consider an upper bound for the calculations and assume that the laminate is free of residual stresses at the sintering temperature (i.e. approx. 850 °C). Then, if no stress relaxation is considered, significant (in-plane) residual stresses arise in the layers during cooling down from the sintering temperature to the ambient temperature, T_0. They can be determined by the classical laminate theory [16–18]. Far from the free edges they are [19]:

$$\sigma_{\mathrm{res},i} = \frac{E_i}{1-\nu_i}(\bar{\alpha}-\alpha_i)\Delta T = \frac{E_i}{1-\nu_i}\Delta\varepsilon_i. \tag{1}$$

here, $\sigma_{\mathrm{res},i}$ is the amplitude of the residual stress and E_i, ν_i and α_i are the Young's modulus, the Poisson's ratio and the coefficient of thermal expansion of the i^{th} layer (i.e. the technical or secant modulus between T_{ref} and T_0), respectively. The ΔT is the temperature difference = $T_{\mathrm{ref}} - T_0$. The mismatch strain; $\Delta\varepsilon_i = (\bar{\alpha}-\alpha_i)\cdot\Delta T$, of the i^{th} layer depends on the difference between the average CTE, given as:

$$\bar{\alpha} = \sum_{i=1}^{N}\frac{E_i t_i \alpha_i}{1-\nu_i} \Big/ \sum_{i=1}^{N}\frac{E_i t_i}{1-\nu_i} \tag{2}$$

and the CTE of the i^{th} layer [20,21], i.e. α_i, with t_i being the thickness of the i^{th} layer and N the total number of layers. As mentioned above, if additional sources of strain mismatch (e.g. phase transformations) exist, these have to be accounted for in Eq. (1). Then, $\Delta\varepsilon_i$ is the sum of all mismatch strains caused by any reason. According to Eq. (2), the ratio between the total thickness, or more generally, the total volume of the two layer materials (V_A/V_B) and not the thickness of the individual layers is the key property for the design of the residual stresses in planar laminates (see Sestakova et al. for more details [21]).

2.2. Brittle fracture and strength statistics

For the sake of simplicity and without loss of generality, the arguments in this chapter are given for specimens loaded in uniaxial and homogenous tension. Cracks are loaded perpendicular to their surface, i.e. in mode I loading. Generalisations for non-homogeneous and multiaxial stress states and/or for arbitrary oriented cracks can be found in the literature (see for instance [22]).

Brittle fracture in ceramic materials is described in the framework of linear elastic fracture mechanics. Fracture starts at flaws in the microstructure or at the surface of the specimen (component), which are described as cracks [23,24]. The stress intensity at the crack tip is proportional to the applied stress according to:

$$K = \sigma Y\sqrt{\pi a}, \tag{3}$$

where σ is the tensile stress in the un-cracked body, Y is the geometric factor of the crack in the specimen and a is the length of the crack. It describes "the loading" of a crack. For details see standard fracture mechanic textbooks [25]. If the stress intensity reaches or exceeds a critical value (the fracture toughness, K_c), spontaneous brittle fracture occurs (Griffith/Irwin criterion: $K \geq K_c$ [26,27]). The tensile strength of a smooth ceramic specimen is limited by the crack loaded with the highest stress intensity factor, and is given as:

$$\sigma_f = \frac{K_c}{Y\sqrt{\pi a_c}}. \tag{4}$$

Since the size of the critical flaw differs from specimen to specimen, also the strength of specimens differs. This is the reason for the large inherent scatter of the strength of ceramic materials: The tensile strength is not given by a simple number but has to be described by a distribution function. Such a function has been proposed by Weibull more than fifty years ago [6,28].

To describe the fracture probability, Weibull defined the simplest analytical dependence of the applied stress, which fits well to