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Influence of temperature and humidity on the strength of low temperature co-fired ceramics

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Abstract

Strength degradation in glass and ceramic materials is related to subcritical crack growth mechanisms acting at the crack tip during mechanical loading. In this work the effect of humidity and temperature on the strength of a commercial low temperature co-fired ceramic was investigated using a biaxial testing procedure. Experiments were performed in argon and in air at different stress rates between 25 and 125 °C. The effect of humidity on strength was assessed at room temperature varying only the relative humidity. The sole effect of temperature was evaluated in argon at high stress rates. The combined effect of humidity and temperature was determined in air, testing at different temperatures. Results showed the existence of an inert strength of the material at room temperature. Measurements in ambient air showed a counterbalance effect of temperature and humidity yielding an almost constant strength for this material between 25 and 125 °C.

Keywords: LTCC; Ceramic matrix composites; Biaxial strength; Temperature; Humidity

1. Introduction

Low temperature co-fired ceramics (LTCC) consist of ceramic grains (i.e. alumina) embedded in a silicate glass matrix. Due to the glass content, low sintering temperatures (i.e. below 900 °C) can be achieved.[1] This makes the sintering of the LTCC tapes together with high-conductivity metals such as copper, silver or silver–palladium-alloys feasible. The LTCC-technology provides components with improved electrical (e.g. low dielectric loss factor), thermal and geometrical behaviour compared to the widely used polymer laminate based printed circuit board (PCB) technology. To give some examples, laminates of LTCC tapes with internal 3D metal structures can be utilised as functional components or 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] They are often used in relatively harsh environments (e.g. at elevated temperature, heavy mechanical loading, and significant vibrations). Therefore, the functionality of LTCC based components depends on their mechanical strength and resistance to crack propagation in a given environment.

It is well known that glass-containing materials are susceptible to subcritical crack growth (SCCG) (sometimes called SCG "slow crack growth" and also known in metals as "stress corrosion"), where cracks can grow under an applied stress intensity factor, K_I, well below the toughness, K_{Ic}, of the material. In ceramics, this phenomenon is related to the presence of humidity. Polar molecules, such as water, interact with the strained crack tip, thus weakening the strength of the bonds at the crack tip.[4–10] This may cause a decrease in the measured strength of the material (i.e. strength degradation). Such a phenomenon has been also reported in LTCC or in SOFC materials loaded under mechanical stress, especially in humid environments.[11–15]

Among the different models proposed to describe the crack growth in these materials, a direct chemical attack of the environment on the crack tip seems to have the strongest experimental support.[7,9] Depending on the crack growth rate and environment, different mechanisms of SCCG exist, which can be recognised by different slopes in a double logarithmic plot of the v–K_I curve,[6] where v represents the crack growth rate and K_I the

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applied stress intensity factor at the crack tip. At a relatively low K_I, a *reaction–rate–controlled region* (region 1) dominates the crack growth rate,[5] which can be empirically described by a single power-law relation (i.e. Paris law)[5,16]:

$$v = \frac{da}{dt} = v_0 \cdot \left(\frac{K_I}{K_{Ic}}\right)^n, \quad (1)$$

where a is the crack size, t is the time, K_I and K_{Ic} are the applied mode I stress intensity factor and the fracture toughness respectively, n is the SCCG exponent and v_0 is a material constant (i.e. critical crack growth velocity) for a particular environment. At a higher stress intensity factor (or in the case of relatively dry environments) the crack growth rate increases to a level limited by the rate of diffusion of water molecules to the crack tip, and thus a *transport–controlled region* (region 2) governs the growth rate in the v–K curve.[5] At very high K_I, close to K_{Ic}, (region 3), the bonds at the tip can break even without the assistance of water molecules and mechanically activated bond breaking dominates.[17] In addition to the effect of humidity, thermal energy can assist the fracture of bonds, resulting in a stepwise crack growth (referred to as "bond trapping" in the literature).[8,18,19] In summary, a combination of thermal and chemical processes can assist the subcritical growth of cracks in these materials.

In a previous work,[13] the SCCG behaviour of a commercial LTCC material was investigated. The SCCG parameters were estimated from biaxial bending tests conducted in ambient air (about 40% relative humidity) at different stress rates. The SCCG exponent was $n = 35 \pm 3$, which is in agreement with other similar LTCC materials reported in literature.[11,12] In this work, the combined effect of humidity and temperature on the strength of the same LTCC material was evaluated at different temperatures (25 °C, 75 °C and 125 °C) and different atmospheres using an adapted ball-on-three-balls (B3B) testing jig. Samples were tested in ambient air at room temperature (RT) and different relative humidity (RH) to quantify the effect of humidity on the strength. The sole effect of temperature was evaluated in argon at different temperatures and relatively high stress rates. In addition, fracture toughness measurements were performed at 25 and 125 °C to assess the influence of temperature on the fracture toughness of the LTCC substrate.

2. Experimental procedure

2.1. Material of study

A commercial LTCC material, referred to as LTCC-MKE, was provided by the company TDK-EPC (Deutschlandsberg, Austria). It consists of Al_2O_3 particles (*approx.* 40 vol.% and 2 μm mean grain size) embedded in a silicate based matrix phase containing Ca, Na, Si, K, B and Al, where the degree of crystallisation after sintering exceeds 90%. A thorough microstructural characterisation can be found in previous work.[13] In Fig. 1 a cross-section of the LTCC material prepared using Focused Ion Beam is shown. The alumina particles (dark-grey) are distributed in the matrix (light-grey). For the biaxial strength measurements rectangular plate specimens (dimension ca. 11.0 mm × 9.6 mm × 1 mm) were used. To obtain a good statistical significance, between 8 and 30 specimens were tested for each condition.

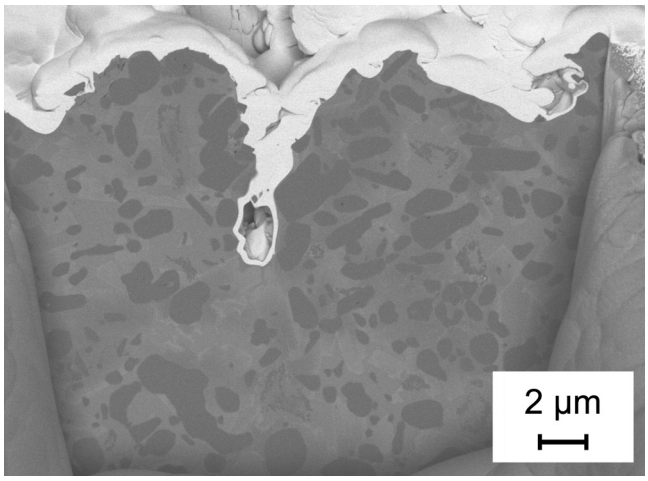

Fig. 1. Cross-section of an LTCC material prepared using a Focused Ion Beam. The alumina particles (dark-grey) are distributed in the glass matrix (light-grey).

2.2. Mechanical testing setup

2.2.1. Strength measurements

The biaxial strength was determined using the ball-on-three-balls (B3B) testing method, which provides very low measurement uncertainties (±1% or less),[20,21] compared to other methods such as flexural four-point bending or ring-on-ring tests.[22] It should be noted that too high measurement uncertainties may cause a large scatter in the measured strength data. As a consequence, a Weibull modulus higher than $m = 20$ can hardly be determined by simple bend beam testing.[23] Such a high precision in strength measurements is also mandatory for an accurate determination of the SCCG exponent with a reasonable small number of tested specimens.[13,24]

During the B3B tests, the "as-sintered" rectangular plates were symmetrically supported by three balls on one face and loaded by a fourth ball in the centre of the opposite face. The four balls had a diameter of 8 mm giving a support radius of 4.619 mm. A pre-load of 20 N was applied to hold the specimen between the four balls. For further details on the testing procedure see Refs. [20,21,25] The tests were conducted under displacement control using a universal testing machine (Messphysik MIDI 25-5, Fürstenfeld, Austria). The corresponding stress rates were calculated from the load–displacement curves. Tests were performed in air and/or in argon atmosphere, at three different temperatures (25 °C, 75 °C, 125 °C) and different stress rates (ranging from 0.7 to 700 MPa/s), depending on the investigated effect: (i) To quantify the effect of humidity on strength, several LTCC samples were tested in ambient air with a constant stress rate (8 MPa/s) at different relative humidity (RH) conditions (i.e. 12% RH, 40% RH and 74% RH); the room temperature during the tests being constant (23 ± 3 °C). (ii) To assess the possible existence of an inert strength of the material (i.e. strength