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# Design of alumina-zirconia composites with spatially tailored strength and toughness

Yunfei Chang[a], Raul Bermejo[b], Oldřich Ševeček[c], Gary L. Messing[a,*]

[a] Department of Materials Science and Engineering, Pennsylvania State University, University Park, PA, 16802, United States
[b] Institut für Struktur-und Funktionskeramik, Montanuniversität Leoben, Leoben, Austria
[c] Institute of Solid Mechanics, Mechatronics and Biomechanics, Brno University of Technology, Brno, Czech Republic

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## Abstract

Composites of $Al_2O_3$–5 vol.% $t$-$ZrO_2$ (ATZ) and $Al_2O_3$–30 vol.% $m$-$ZrO_2$ (AMZ) layers were designed with 3–1 connectivity to explore the effect of spatially-dependent residual stress and layer distribution on mechanical behavior. ATZ composites with 'shallow' and 'deep' regions of AMZ, defined relative to the distance from the surface, were fabricated. Four-point bending tests on indented 3–1 composites showed crack arrest in the first compressive AMZ layer and a fracture strength nearly independent of indent size (i.e. minimum strength); the failure occurring in the region with thicker outer ATZ layers ('deep' region). Region dependent crack growth resistance was measured on SEVNB specimens and compared to theoretical predictions using a fracture mechanics model. Spatially tailored constant strengths were obtained, ranging between 148 MPa and 470 MPa; the maximum value corresponding to a 'shallow' region with a relatively thicker AMZ compressive layer embedded close to the tensile ATZ surface. The 3–1 design concept allows the fabrication of 'deep' and 'shallow' embedded regions within a unique composite architecture, thus providing a preferential path for crack propagation, opening new possibilities for design of composite structures with spatially-tailored crack growth resistance.

*Keywords:* Connectivity; Composites; Residual stresses; Fracture strength; Toughness

## 1. Introduction

The variable strength in brittle materials such as glasses and ceramics is related to the different size of critical defects, from specimen to specimen, introduced during processing, from machining or occurring in service. The strength cannot be characterized as a single value but as a distribution function, which is related to the defect size distribution in the material.[1] Increasing strength in glasses and ceramics can be attained by reducing the size of critical defects (e.g. through colloidal processing)[2] or introducing surface compressive residual stresses (e.g. strengthening in glass)[3,4] to enhance the resistance of the material to crack propagation. However, significant reduction of strength variability may not be achieved. Rather than reducing critical flaw size, a "flaw-tolerant" approach has been developed to build ceramic composites by combining layers of two different materials/microstructures in a periodic architecture with 2–2 multilayer connectivity[d].[5–13] Two main approaches regarding the fracture energy of the layer interfaces are particularly useful, which aim to generate "weak" or "strong" interfaces. In particular, 2–2 layered composites designed with strong interfaces can present enhanced mechanical behavior through microstructural design

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[*] Corresponding author. Tel.: +01 814 865 2262; fax: +01 814 865 8262.
*E-mail addresses:* messing@ems.psu.edu, messing@matse.psu.edu (G.L. Messing).

[d] Connectivity is defined as the number of dimensions in which each component (e.g. material or microstructure) is self-connected.[5] In two-component composites, a 2–2 composite is one in which both components are self-connected in two dimensions, i.e. a laminate. More details on the connectivity and its schematic illustration are available in the supplemental file.

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(e.g. grain size, layer composition) and/or due to the presence of compressive residual stresses, acting as a barrier to crack propagation. Under certain conditions, the compressive stresses may arrest the propagation of surface cracks, yielding a so-called "threshold strength", i.e. a minimum stress level below which failure does not occur despite the presence of relatively large cracks.

Threshold strength was first reported for alumina-mullite layered composites, where the propagation of indentation cracks under bending was arrested between two compressive layers.[14] The concept of designing ceramics with a "minimum strength" was further extended to other alumina-based 2–2 layered composites (e.g. alumina–$Si_3N_4$, alumina–zirconia) under different loading configurations.[7,8,10,11] In alumina–zirconia systems, high compressive stresses in the embedded layers are developed during cooling from the sintering temperature because of the differences in thermal expansion coefficients between layers of different composition.[15] The profile of the tensile and compressive stresses can be tailored by combinations of composition, stacking sequence and layer thickness.[7,10,15–17] Recent advances in fracture mechanics modeling have shown that an optimized non-periodic 2–2 layered architecture can result in pronounced crack growth resistance and minimum strength using thin outer tensile layers and thicker internal compressive layers.[18] Although much progress has been achieved in improving the resistance to crack propagation in 2–2 multilayer composites, the potential of complex architectural design has not been sufficiently exploited or explored. This is very important because spatial control and composite connectivity of the individual layers of a composite are ultimately the key to developing and controlling useful and unique properties.

From the functional point of view, the dependence of properties on connectivity is especially pronounced in ceramic components that are fabricated with complex connectivities for piezoelectric transducer, magnetic field sensor, low temperature co-fired electronic package and solid oxide fuel cell applications etc.[5,19–22] However, from the structural integrity point of view, residual stresses related to the combination of different materials (e.g. metal electrodes, ceramic parts) may initiate cracks, which can propagate during service and reduce the component functionality.[23] The development of structural composites with various connectivities (e.g. 3–1, 1–1) in three-dimensional structures might allow access to unique mechanical properties never before achieved in 2–2 composites. To our knowledge, few studies focused on designing and understanding the properties of structural composites with such higher levels of complexity are available in the literature.

In this work, we explore novel 3–1 connected composites to understand how this connectivity can be used to affect the resistance to crack propagation and fracture behavior in a ceramic composite. Alumina–zirconia was chosen as the model system due to its highly useful mechanical properties that warrant further exploration. Four types of 3–1 connected alumina–zirconia based composites with 'shallow' and 'deep' regions, defined relative to the distance of the embedded compressive layers from the surface, were fabricated. Crack propagation was investigated by four-point bending of indented 3–1 specimens and compared to that of 2–2 composites. The effects of location and thickness of the embedded compressive layers on the fracture strength and fracture toughness of 3–1 composites were determined. A fracture mechanics model, based on a weight function analysis, was implemented to interpret the results.

## 2. Experimental

The 3–1 connected alumina–zirconia composites consist of two components: layers of 95 vol% alumina and 5 vol% $Y_2O_3$-stabilized zirconia (ATZ), and embedded layers of 70 vol% alumina and 30 vol% monoclinic zirconia (AMZ). In 3–1 connected alumina–zirconia composites, ATZ and AMZ components are self-connected in three and one dimensions, respectively. The addition of 5 vol% tetragonal zirconia in ATZ has the effect of limiting alumina grain growth during sintering. The 30 vol% of monoclinic zirconia was used in the AMZ layers to generate a large strain mismatch between the AMZ and ATZ layers as a result of the ~5% volume change during the tetragonal-monoclinic transformation at ~730 °C upon cooling,[24] and thus induce a high residual compressive stress when embedded in the ATZ matrix.[24,25]

The composite was assembled with a symmetric and non-periodic distribution of the embedded AMZ layers. The symmetric architecture of the composite avoids warpage during sintering. The non-periodic design enables the distribution of layers at different locations and depths within a particular region, while having a constant total volume ratio between the two materials. The design and properties of the four types of composites, including the AMZ layer thicknesses ($t_{AMZ}$) and the corresponding first layer thickness ratios ($t^{1st}_{ATZ}$:$t^{1st}_{AMZ}$), are shown in Fig. 1 and listed in Table 1. The total volume ratio of ATZ:AMZ materials is 12.0 ± 0.2 for all 3–1 composites studied. Two main regions can be defined in each 3–1 composite, hereinafter referred to 'shallow' and 'deep' regions, associated with the distance ($t^{1st}_{ATZ}$) from the surface to the first AMZ embedded layer. The nomenclature used for the four types of 3–1 composites is AF, BE, CD and GH, where the regions A, B, C, D, E, F, G and H correspond to depths ($t^{1st}_{ATZ}$) of 1222 μm, 802 μm, 458 μm, 382 μm, 382 μm, 382 μm, 382 μm and 150 μm for the first AMZ embedded layers, respectively. The AMZ layer is 75 μm thick in all cases except for GH where the AMZ layers are 150 μm thick in region H.

### 2.1. Fabrication of 3–1 alumina–zirconia composites

Composites were produced from 0.28 μm α-$Al_2O_3$ powders (AKP50, Sumitomo Chemical Co. Ltd., Tokyo, Japan), 0.60 μm yttria stabilized zirconia powder (TZ-3Y, Tosoh, Yamaguchi, Japan), and 0.30 μm monoclinic zirconia powder (TZ-0, Tosoh, Yamaguchi, Japan). The ATZ and AMZ tape casting slurries were prepared by ball milling the powders for 48 h in a 50:50 (weight ratio) xylenes ± ethanol solution containing blown menhaden fish oil. Binder and plasticizers (polyvinyl butyral, butyl benzyl phthalate, and polyalkylene glycol) were then added, followed by an additional 24 h milling/mixing step.[26]