Composites Part B 119 (2017) 79–89

Contents lists available at ScienceDirect

Composites Part B

journal homepage: www.elsevier.com/locate/compositesb

Sol-gel temperature dependent ductile-to-brittle transition of aluminosilicate fiber reinforced silica matrix composite

L.W. Yang [b, *, 1], J.Y. Wang [b, 1], H.T. Liu [a, **], R. Jiang [a], H.F. Cheng [a]

[a] Science and Technology on Advanced Ceramic Fibers and Composites Laboratory, National University of Defense Technology, Changsha 410073, China
[b] IMDEA Materials Institute, c/Eric Kandel 2, 28906 Getafe, Madrid, Spain

ARTICLE INFO

Article history:
Received 16 January 2017
Received in revised form
20 March 2017
Accepted 21 March 2017
Available online 24 March 2017

Keywords:
Ceramic matrix composite
Nanoindentation
Fiber/matrix bond
Damage tolerance
Mechanical properties

ABSTRACT

A thorough understanding in the micro mechanical properties of the fiber, the fiber/matrix and the matrix of the composite material, especially in response to different processing temperatures, is critical to explore the strengthening and toughening mechanisms of the novel oxide fiber reinforced oxide ceramic matrix composites. This work contributes this by studying the sol-gel temperature dependent micro mechanical properties of an aluminosilicate fiber reinforced silica matrix composite using novel nano-mechanical methods like nanoindentation and push-in techniques. In the sol-gel temperature range of 600–1200 °C, the aluminosilicate fiber was comparably stable in mechanical properties, though a microstructure transition from amorphous SiO_2+ γ-Al_2O_3 to crystallized mullite+γ-Al_2O_3 was found at 1000 °C. The silica matrix was amorphous in microstructure, but were more crystallized as the sol-gel temperature increased, which subsequently led to an enhanced mechanical property at higher temperature. The interfacial shear strength was small in the temperature range of 600–1000 °C, and was increased slightly from ~50 MPa at 600 °C to ~84 MPa at 1000 °C. The interfacial shear strength at 1200 °C was high (~256 MPa), due to the interfacial reaction occurring at this temperature. Correlating these micro mechanical properties to the macro fracture resistance that was quantified by the three-point bending test, an interface mediated toughening mechanism was finally concluded to explain the sol-gel dependent fracture resistance of the composite and the ductile-to-brittle transition observed in the temperature range.

1. Introduction

Oxide fiber reinforced oxide ceramic matrix (oxide/oxide) composites are promising for high temperature applications in oxidizing atmospheres due to their excellent oxidation resistant property and thermal stability [1–15]. Particularly, the aluminosilicate (AS) fiber reinforced oxide/oxide (AS/oxide) composites have shown excellent mechanical properties with high strength and damage tolerance, due that firstly, the AS fiber is more thermally and mechanically stable than other oxide fibers like quartz fibers, silica fibers, etc. [4]; secondly, the interface interaction of the composite can be tailored by proper interphases like pyrocarbon, BN [5,6,15,16]. The mechanical properties of the AS-oxide/oxide composites are mainly dominated by the micro mechanical properties of the AS fiber, the fiber/matrix interface and the oxide matrix, which are highly dependent on the composite fabrication conditions like processing temperature of the matrix [17–20]. The functional properties of the AS-oxide/oxide composites are also developed recently. For instance, the AS fiber reinforced silica matrix (AS/SiO_2) composites have been widely applied as wave transparent materials, due to their low dielectric constant and loss tangent and the excellent electrical insulations of the composite constituents [21–24].

Full understanding in the macro mechanical response of the oxide/oxide composite when subjected to various external loadings requires accurate measurements of the micro mechanical properties of the fibers, matrix and interface, which till now is still not well defined due to the difficulties to perform the micro-mechanical testing in small length scales [11,25–28]. While novel nano-mechanical testing techniques like nanoindentation [29–33], fiber push-in/push-out [31,34–41], micro-cantilever bending [29],

* Corresponding author.
** Corresponding author.
E-mail addresses: yang.lingwei@imdea.org (L.W. Yang), xzddlht@163.com (H.T. Liu).
[1] These authors contributed equally to the work.

http://dx.doi.org/10.1016/j.compositesb.2017.03.043
1359-8368/

etc. have been widely applied to acquire the modulus, strength and toughness of the fiber and matrix and the shear properties of the fiber/matrix interface, they are still scarcely employed in the novel oxide/oxide composite systems to help to understand fundamentally the macro mechanical performance and to accelerate the design and development of novel oxide/oxide composites [11]. For instance, recent studies have shown that the 1200 °C fabricated AS/SiO_2 composite is brittle in three-point bend response due to the strong interfacial interaction by the elemental diffusion at this temperature, which can be subsequently hindered by the pyrocarbon interphase [6]. However, instead of fabrication of the time-consuming and expensive interphase, a more efficient method to toughen the composite lies in tailoring the processing temperature to obtain optimized composite strength with appropriate micro-mechanical properties. However, no further work was performed in this composite so far to study the dominating micro mechanical properties especially when the processing temperature was changed properly.

This work contributes this by studying the processing temperature dependent micro and macro mechanical properties of the AS/SiO_2 composite fabricated by the sol-gel method in a temperature range of 600–1200 °C [42]. The microstructures of the composites evolved with the sol-gel temperature were studied mainly by the x-ray diffraction (XRD), scanning electron microscopy (SEM) and the transmission electron microscopy (TEM). The sol-gel dependent modulus and the strength of the AS fiber and the SiO_2 matrix were quantified directly in the composite form by the instrumented nanoindentation. The interfacial shear strength as a function of the processing temperature was studied by the fiber push-in tests. The macro fracture resistances of the composite fabricated at different temperatures were finally measured by the macro three-point bend (TPB) method, and were correlated to the micro-mechanical properties to shed light on the underlying strengthening and toughening mechanisms in this typical oxide/oxide composite.

2. Experimental and modeling procedures

2.1. Composite fabrication process

The AS/SiO_2 composites were fabricated by reinforcing the 3D woven AS fiber preforms (ALF 3025T fiber fabrics, from Nitivy ALF Co., Japan) with the sol-gel processed SiO_2 matrix in argon atmosphere and at temperatures of 600, 800, 1000 and 1200 °C, respectively. Note temperatures lower than 600 °C would lead to inadequate gelate reaction of the sols, while temperatures higher than 1200 °C could result in severe mechanical degradation of the AS fiber [17]. The as-received AS fiber preform was ~40% in volume fraction [7,43,44], having a weaving structure schematically shown in Fig. 1. The sol-gel process was as follows: firstly, the as-received AS fiber preforms were infiltrated in the silica sols (Zhejiang University, Hangzhou, China) for >4 h in vacuum to allow adequate sol infiltration into the preform interiors [17]. After that, the infiltrated preforms were pyrolyzed at target temperatures for 30 min in argon to fabricate the SiO_2 matrix, after dried at 200 °C for 2 h.

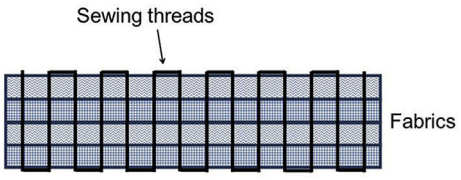

Fig. 1. Schematic of the AS fiber preform employed in the AS/SiO_2 composite in this study.

Finally, the process was repeated for >12 times until the pore closure, which was indicated by a <1% weight increase during subsequent sol-gel cycles. The sol-gel process was typical, and has been widely used to fabricate the oxide/oxide composites [3,17,45]. The densities of all the composites after the sol-gel processes were ~1.92 g/cm^3, leading to similar porosities in the composites, despite of the differences in the processing temperature. Smooth cross-sectional surfaces were fabricated from the AS/SiO_2 composites by the traditional grinding and polishing processes, in order to implement the nanomechanical testing.

2.2. Microstructure characterization technique

The microstructures of the AS fiber and the AS/SiO_2 composites were characterized in this work by the XRD (Cu Kα, D8 ADVANCE diffractometer) and the TEM (FEI Talos AS00X). The morphologies of both were characterized by the optical microscopy (Olympus BX51) and the SEM (field emission SEM in a Helios 600i FEI system). The chemical composition of the AS/SiO_2 composite was measured by the energy dispersive X-ray spectroscopy (EDS, Oxford INCA).

2.3. Micro-mechanical property testing method

Instrumented nanoindentation was performed on the individual AS fiber and the SiO_2 matrix in the finely polished surfaces of the AS/SiO_2 composite to measure the corresponding Young's modulus and hardness. The tests were performed in a 950i Triboindenter™ system (Hysitron Inc, Minneapolis, MN), employing a Berkovich diamond indenter. Details of the indenter geometry can be assessed in Ref. [46]. The area function of the indenter was calibrated prior to the measurements using the procedure by Oliver and Pharr [47] to eliminate the effect of the roughness of the indenter. The system is capable of positioning the indenter precisely to where to be indented using an *in-situ* scanning probe microscopy (SPM) imaging mode. This technique was employed in this work to perform the nanoindentation on the AS fibers. In addition, the compliance of the testing environment can be a problem due to the complexity of the composite structure where high density AS fibers were distributed randomly in the SiO_2 matrix. To eliminate this, a partial loading-unloading nanoindentation procedure (load controlled mode, maximum load: 10 mN) was employed to obtain depth independent mechanical properties. Each cycle involves loading for 1 s and unloading for 1 s after holding at peak load for 1 s. Despite that the loading rate for each cycle was different, the mechanical properties were expected to be strain rate independent, due to the brittle nature of the ceramic fiber and matrix. Finally, the Oliver and Pharr method [47] was employed to extract the Young's modulus and the hardness of the AS fiber and SiO_2 matrix based on the experimental force-penetration depth curves.

Fiber push-in tests were performed in the composite cross-sections also in the Triboindenter™ system, but using a 3 μm flat diamond punch, to quantify the interfacial shear strength. The test was in displacement control mode, with a constant displacement rate of 30 nm/s up to a maximum depth of 2 μm. A minimum of 10 tests was performed for each composite sample. The interfacial shear strength was extracted from the push-in force-displacement curves based on the analytical shear-lag model [34,35].

2.4. Macro-mechanical testing methods

TPB tests were performed to quantify the macro fracture resistance of the AS/SiO_2 composites as a function of the sol-gel temperature, following the ASTM C1341-06. The tested specimens were machined by the metal-bonded diamond cutting blade to a thickness of ~3 mm, a width of ~4 mm and a length of ~50 mm. The