Materials Science and Engineering A 393 (2005) 170–178

MATERIALS SCIENCE & ENGINEERING A

www.elsevier.com/locate/msea

On the mechanical properties of alumina–epoxy composites with an interpenetrating network structure

M.T. Tilbrook, R.J. Moon, M. Hoffman*

School of Materials Science and Engineering, University of New South Wales, Sydney, NSW 2052, Australia

Received 17 June 2004; received in revised form 4 October 2004; accepted 4 October 2004

Abstract

Methods of predicting effective mechanical properties of composites with an interpenetrating network structure are currently not well understood, particularly for cases in which the constituent materials have widely differing properties. Alumina–epoxy composites with an interpenetrating composite structure have been produced via an infiltration process and the elastic properties were measured via the impulse excitation technique. A strong dependence of properties on composition and processing was observed. Properties were compared with several mixing law predictions made using compositional data obtained from microstructural analysis. The effective medium approximation (EMA) was shown to predict properties adequately, whilst others models proved inappropriate.

Keywords: Interpenetrating network composite; Effective properties; Impulse excitation technique; Effective medium approximation

1. Introduction

Composites potentially represent an advance in optimisation of material performance, by offering a combination of properties of several different constituent materials. Underpinning composite design is an understanding of the relation between mechanical properties of the constituent materials and the effective mechanical properties of the resulting composite material. For a particular composite, this depends on the internal geometry and may be derived theoretically [1–5], though experimental verification of effective properties is crucial to accurately relate material composition and properties.

Composites with an interpenetrating network structure are of particular interest as they have been found to demonstrate higher strength, toughness and wear resistance when compared to other composite structures [6,7]. Traditionally, composite materials tended to display a fibre/matrix, layered, laminar or particle/matrix type structure. Correspondingly, theoretical models based upon such geometries have proliferated [1–5], whilst work on interpenetrating structures has been more limited [8].

This issue is of particular interest for functionally graded materials (FGMs), which incorporate a spatial variation of composition and properties [9–11], where experimental verification of local mechanical properties is difficult. Furthermore, FGMs often possess an interpenetrating network structure [12]. Recent interest in graded materials has motivated the development of semi-empirical models, based on micromechanics and image analysis [13–15].

Several generic models may potentially be applicable to interpenetrating network-structured composites, including the isostress- and isostrain-approximation models, Hashin–Shtrikman bounds, and the effective medium approximation (EMA). In addition, several models have been proposed for composites with two continuous phases with simple regular internal geometries [8,16]. For a more rigorous treatment of mixing laws, see the work of Torquato et al. [17], Reiter et al. [11] or Suresh and Mortensen [10].

The simple isostress and isostrain relations provide a first approximation of the bounds for effective Young's modulus [17]. These are:

$$E^* = v_1 E_1 + v_2 E_2 \tag{1}$$

---

* Corresponding author. Tel.: +61 2 9385 4223; fax: +61 2 9385 5956.
E-mail address: mark.hoffman@unsw.edu.au (M. Hoffman).

0921-5093/$ – see front matter © 2004 Elsevier B.V. All rights reserved.
doi:10.1016/j.msea.2004.10.004

$$E^* = \left(\frac{v_1}{E_1} + \frac{v_2}{E_2}\right)^{-1} \tag{2}$$

where E is the Young's modulus, (*) represents the effective composite property, v the volume fraction and subscripts 1 and 2 denote each phase. Replacing Young's modulus, E, in these equations with bulk and shear moduli, K and G, leads to the Voigt and Reuss model expressions.

The Hashin–Shtrikman bounds [18] were used by Torquato et al. to successfully predict the mechanical properties of interpenetrating boron carbide–aluminium composites [17]. The effective medium approximation (EMA) [4] was used by Hoffman et al. to predict mechanical properties of aluminium–alumina-interpenetrating composites [19], although the validity of this was not confirmed experimentally. Tuchinskii developed a simplified unit-cell model for composites with two continuous phases [16]. This was observed by Jedamzik et al. [20] to fit experimental results for interpenetrating structured tungsten–copper composites. A similar approach was applied by Feng et al. [21] to interpenetrating composites with more than two continuous phases.

This paper details the investigation of effective mechanical properties of alumina–epoxy composites with an interpenetrating network structure. Experimental results obtained via the impulse excitation technique are presented and several theoretical models are applied to this composite system. The alumina–epoxy composite system has been adopted as a model system due to the significantly differing elastic properties of the two phases. This enables a large stiffness variation to be attained across the composition range.

The large difference in properties of the two constituents leads to widely varying predictions from different models. Considering for instance, a composite of 50% alumina ($E = 400$ GPa) and 50% epoxy ($E = 4$ GPa) [22], the predictions from Eqs. (1) and (2) are 202 and 7.9 GPa, respectively. These are clearly extremely disparate and a more precise prediction is required. For a 50/50 composite of alumina and aluminium ($E = 79$ GPa), the predicted values, 240 and 132 GPa, respectively, differ less drastically though still significantly.

An advantage of investigating an alumina–epoxy composite system is that the increased variation, between different model predictions, enables improved identification of the appropriate choice of model for composites with interpenetrating network structure.

2. Experimental techniques

2.1. Sample processing and preparation

Alumina–epoxy FGMs were produced by infiltration of epoxy into an open porosity alumina preform. Samples were produced in the composition range 5–50% epoxy, with an interpenetrating network structure in which both phases are continuous.

The processing was conducted similarly to the method described by Cichocki et al. [23], which has been used to produce aluminium–alumina-interpenetrating composites [12]. Open-celled polyurethane (PU) foam (Bulpren S-31048, Eurofoam, Troisdorf, Germany, 90 pores/in.) was used as an imprint for the interpenetrating network structure. It was compressed from an initial density of 2.5% to obtain the required volume fractions of foam and air.

A colloidal suspension of alumina particles (99.99% Al_2O_3, Taimicron TM-DAR, Taimei Chemicals Co. Ltd., Japan) was infiltrated into the PU foam under varying pressure, cast and allowed to dry. The foam was then pyrolysed at 800 °C, leaving a ceramic green-body (approximate dimensions: 50 mm × 30 mm × 6 mm) with a network of interconnecting pores. The ceramic was sintered at 1500 °C for 1 h leading to ∼10% shrinkage in each dimension. Epoxy resin (Epofix, Struers, Germany) was infiltrated into the ceramic under varying pressure then cured at room temperature. Specimens were then ground to size (50 mm × 30 mm × 5 mm) with a 600 grit diamond-grinding wheel, and polished with an automatic polisher and diamond paste down to a diamond particle size of 1 μm in the final step. The plate specimens were later cut into small beams (50 mm × 5 mm × 4 mm approx.) with a low speed diamond saw for further testing. This process results in two continuous interpenetrating phases; the interconnecting porosity of the foam perform is mimicked by the alumina, which is a robust structure after firing, while the complete penetration of the epoxy with no discernable porosity in the composite confirms the complete interpenetration of this phase.

Several samples of an alumina particulate/epoxy matrix composite in the composition range 75–90% were also produced, for comparison with the interpenetrating-network composites. Alumina powder (Taimicron TM-DAR, as above) was mixed with epoxy resin (Epofix) by stirring before curing. Due to the viscosity of the mixture, air bubbles introduced during stirring were difficult to remove, which lead to some porosity in these samples.

Monolithic alumina and epoxy specimens were also produced for comparison. Alumina specimens were produced from a suspension of alumina particles (Taimicron TM-DAR), which was slip-cast, dried and sintered at 1500 °C for 1 h. Epoxy specimens were produced from resin (Epofix) cured at room temperature. Grinding and cutting was conducted similarly to the composite specimens.

2.2. Microstructural analysis

Optical microscopy on the polished surfaces was used to characterise the alumina and epoxy phases and to quantify the porosity. Images were obtained using a Nikon 200 microscope and digital camera, and then processed using Adobe Photoshop and Image Processing ToolKit (Version 3.0) [24]. Examples of microstructures for a range of compositions, for both the interpenetrating-network and powder composites, are shown in Fig. 1. The continuity of each phase in the