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ScienceDirect

Ceramics International 42 (2016) 325–333

CERAMICS INTERNATIONAL

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Using a novel microstructure generator to calculate macroscopic properties of multi-phase non-oxide ceramics in comparison to experiments

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Received 29 April 2015; received in revised form 18 August 2015; accepted 24 August 2015
Available online 12 September 2015

Abstract

A novel method to generate microstructures and calculate thermal and elastic properties in non-oxide ceramics, namely in Aluminum Nitride (ALN), Silicon Nitride (Si_3N_4), and Silicon Carbide (SiSiC) is presented. Structural features like dihedral angle (ALN), anisotropic material properties of grains (Si_3N_4), and multiscale structure (SiSiC) are considered. To ensure that the simulated structures are close to the real materials, several samples of all ceramics were prepared by the artifact-free method of cross section polishing and imaged by scanning electron microscopy (SEM). By image analysis chord lengths, phase fractions, connectivity between grains or elongation of grains were obtained. The same parameters were extracted from 2-dimensional sections of 3-dimensional representative volume elements produced by a structure generator. Structure generation followed closely the structure formation in the real process. It was repeated until close agreement between experimental and theoretical structures was obtained. Then, thermal and mechanical properties were calculated by finite element simulations. The calculated material properties showed good agreement to the obtained experimental data.

Keywords: B. Grain boundaries; B. Microstructure-final; C. Mechanical properties; C. Thermal conductivity; Finite element simulations

1. Introduction

Material properties of ideal, single-phase materials can be obtained either by measurements or from first principle simulations. But as soon as real polycrystalline multi-phase materials are of interest, it immediately becomes very complex to calculate the macroscopic properties of the whole structure ab initio. Moreover, measurements cannot be used if the range of material properties is to be estimated for multi-phase materials with novel microstructures.

However, multi-phase materials have been of enormous interest since the very beginning of the industrial revolution due to the use of steel as a construction material. The technical development showed the need for an underlying description of materials and inspired the development of theoretical models. Bruggeman describes how to apply the linear theory of elasticity to multi-phase materials, combining parallel and serial arrangements of the phases [1]. Further improvements in modeling date from the 1960s by Hashin and Shtrikman: They applied a displacement approach called "variational principle", to calculate upper bounds and a stress approach to determine lower bounds [2]. They proved by experiments on an alloy with particles embedded in a matrix that the predicted bounds apply to materials with inhomogeneous composition.

G. Ondracek developed a model concept to calculate effective thermal, elastic or electric properties of multi-phase materials with simple structures: ellipsoidal particles embedded in a matrix phase or two interpenetrating phases [3]. This model is helpful, where a coarse approximation to real structures is sufficient. However, only due to the enormous progress in computing power going along

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http://dx.doi.org/10.1016/j.ceramint.2015.08.114
0272-8842/

with the development of efficient algorithms, the realistic treatment of multi-phase material became feasible during the 1990s.

Since then, numerous research groups have investigated microstructure generation and calculation of macroscopic properties. A survey on the literature published until 2003 is given in [4]. Some typical approaches published thereafter and dealing with ceramic phases are described below.

The research group around K. Chawla (University of Alabama, Birmingham, USA) digitalizes microstructures of cermets (SiC grains, embedded in aluminum) and calculates the stress–strain curves of these microstructures [5]. The digitalization of the samples is realized by serial sectioning with X-ray tomography. A variation of the microstructure is not possible in this simulation and is therefore realized by simplified models which use ellipsoidal objects embedded in a secondary phase. A better spatial resolution can be obtained by the combination of focused ion beam and scanning electron microscopy [6], but again the representation of the real structure does not allow for variations.

The group around D.S. Smith (University of Limoges, France) uses Monte Carlo simulations to estimate the thermal conductivity of porous ceramics and biomaterials. For the simulation of porous ceramics, spherical pores are introduced randomly in a representative volume element (RVE). As model system for biomaterials they use irregular shaped 3D particles, fill them on a random basis into a container and cut a RVE out of the container [7–9]. Both model systems allow the calculation of effective thermal conductivities.

The approach of the group around T. Böhlke (KIT Karlsruhe, Germany) focuses on the mathematical perfection of algorithms to model ceramic microstructures, using Voronoi structures [10] and grain-like objects embedded into a matrix phase [11]. On the latter the elastic properties and fracture toughness of silicon nitride are simulated [12]. In both cases they generate model systems in good agreement with sample microstructures but do not consider heat transfer properties.

Commercial tools like *GeoDict* or *Digimat* are also available which digitalize microstructures of multi-phase materials. GeoDict can use computer tomography for the generation of representative volume elements or build structures from simple shaped geometric particles [13]. Digimat calculates the multi-physics nonlinear behavior of homogeneous models [14]. However, all the approaches focus on either the spatial reproduction of real microstructures or on the generation of simplified models. The former does not allow for structural variations and the latter does not take into account particular features of liquid phase sintered ceramics.

For these requirements we extended an in-house software tool called *GeoVal* which combines particle and voxel oriented methods to generate representative volume elements [15]. Combined with a meshing tool developed at Fraunhofer ISC [15] and the finite element methods already described in [4], macroscopic properties like thermal, elastic and electric properties are calculated.

With this approach it is possible to investigate the effect of changes in microstructure genesis closely related to process parameters during raw material production, forming, and sintering. In the first step, the methods have to be validated by generating microstructures which are similar to experimental structures and comparing simulated and measured material properties. The objective of the present paper is to describe experimental and simulation methods and then check the agreement between both routes. The consequences of structural variations on macroscopic properties will be discussed in a further paper.

2. Experimentals

2.1. Investigated samples

Three non-oxide ceramics, ALN, Si_3Ni_4 and SiSiC, were selected for the investigation. They cover a large fraction of possible microstructures of binary phase ceramics. In these three ceramics grains of the main phase had very different shapes in these three ceramics and secondary phase distribution showed large diversity. In ALN and Si_3Ni_4 samples, the secondary phase is formed by oxide sintering additives. Both ceramic types have low residual porosity after sintering, which was not determined separately. Judging from SEM images, the samples have a sealed porosity of 1–3%. As Young's modulus can be assumed to be linear with the porosity for nearly dense ceramics [16,17], the influence of the porosity on the samples' Young's moduli is within the error bars of the simulations and therefore neglected. SiSiC is produced by liquid silicon infiltration (LSI) of a porous SiC ceramic, which results in dense ceramics. The secondary phase consists mainly of Silicon eventually containing additionally small SiC particles formed during infiltration. Two different variants of each ceramics are investigated:

- ALN: One ALN sample produced by tape casting (referred to as AlN-TC), the other one by dry pressing (AlN-DP). As additive yttria (Y_2O_3) is used which forms yttrium-aluminates with alumina (Al_2O_3) impurities contained in the ALN powder.
- Si_3Ni_4: One Si_3Ni_4 sample with Y_2O_3 and Al_2O_3 additives (referred to as Si_3Ni_4-YA), the other Si_3Ni_4 sample with Y_2O_3, silica (SiO_2) and magnesia (MgO) (Si_3Ni_4-YSM).
- SiSiC: Two SiSiC samples with different contents of carbon (referred to as SiC-1 and SiC-2).

All samples were produced within the joint project MIKFORM [18].

2.2. Microstructural analysis

Cross section polishing (CSP) by an Argon-ion beam was chosen for an artifact-free preparation of SEM samples. The Argon ions cut into the sample at an angle of 90° [19,20]. Ten SEM images were taken per sample where each showed roughly 100 grains (see enlarged section of a SEM image in Fig. 1a). For quantitative image analysis, the SEM images were binarized (Fig. 1b). The commercially available software *ImageC* was used to carry out chord length and shape analyses.