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On the control of the liquid-phase distribution in multi-material assemblies processed by liquid-phase sintering

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Abstract

Proper control of composition gradients in components processed by liquid-phase sintering requires the control of the migration of the liquid phase during sintering. A model considering isotropic interface energies has been developed for expressing the variation of the solid–liquid interface area with liquid volume fraction during the final stage of liquid-phase sintering (*i.e.* when residual porosity has disappeared). The model allows the computation of the driving forces for shape accommodation as a function of the dihedral angle ψ, liquid volume fraction, u, particle volume, V_p, and average particle coordination, n_c. It particularly enlightens the coupling between the effects of ψ and of n_c. The model allows a new insight into the control of microstructural evolution during liquid-phase sintering of assemblies of dual-phase materials with different average particle size. Taking $\psi = 0°$ for the WC–Co system, the model was found to predict with a good accuracy the distribution of Co at equilibrium in assemblies made by heat-treating at 1400 °C under protective atmosphere stacks of cylinders of WC–Co cermets differing in mean WC particle size and cobalt content.

Keywords: Thermodynamics; Modelling; Interface wetting; Liquid-phase sintering; Liquid infiltration

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1. Introduction

Capillarity phenomena play a major role in the evolution of systems consisting of a finely dispersed solid phase embedded into a percolating liquid phase. Such systems are relevant for many material processing technologies, such as, in particular, thixocasting, thixoforming, and liquid-phase sintering. Proper understanding of the thermodynamics of capillarity is a condition for the modelling of the kinetics of microstructural evolution in the presence of a liquid phase (e.g. [1–5]).

A point of particular interest is the redistribution (or "migration") of a liquid phase during heat-treatment of materials with gradients of composition and particle size. The type of system investigated in this work is sketched in Fig. 1. This paper focuses on the capability of control of the liquid-phase distribution in such systems. The key of the phenomenon is "shape accommodation", *i.e.* the fact that solid particles tend to assume the shape which corresponds to the minimisation of interface energy. We consider only the evolution of the system after the elimination of residual porosity: as suggested in Fig. 1, the material thus consists of an aggregate of solid particles immersed in a liquid that completely fills the interstices between the particles.

The subject will first be illustrated by a presentation of experimental results on the processing of bi-material assemblies of WC–Co cermets with different particle sizes. The final objective is to allow control of gradients of liquid fraction in multilayered materials via a proper choice of the particle sizes in each layer. Enlightened by the experimental observations, we will deepen the discussion of a model that has been introduced in a previous work [6], in which the analysis was restricted to cases with a dihedral angle ψ equal to zero (see definition of ψ in Fig. 1). The model revisits previous analyses (e.g. [7]) by treating the average particle coordination n_c as a continuous variable. Although the analysis could be extended to more complex cases [6], the model assumes isotropic interface energies. Hence, when the fluid volume fraction exceeds a critical value, the equilibrium

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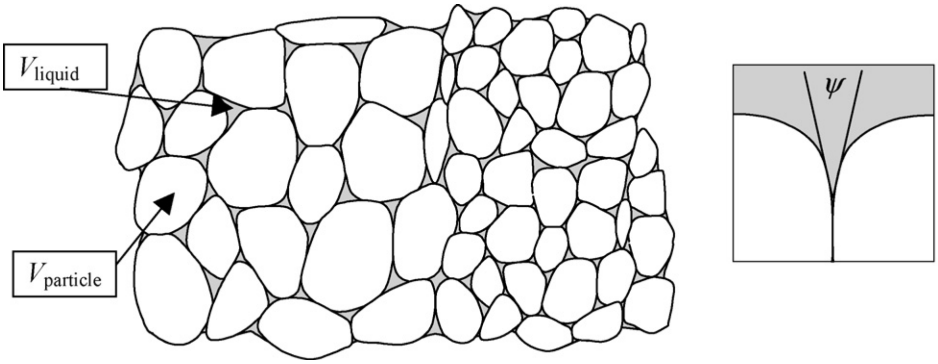


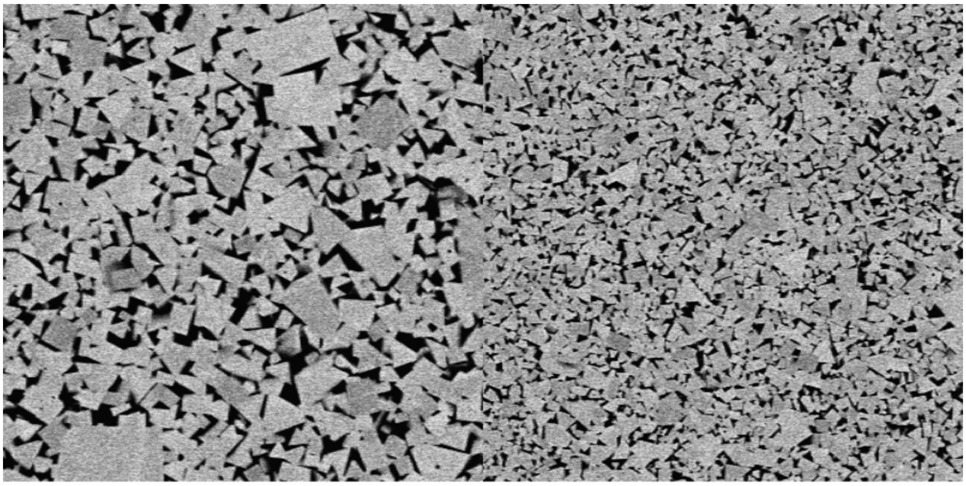
Fig. 1. Schematic representation of the solid–liquid bi-material system considered in this work and definition the dihedral angle: $\psi = 2\arccos(\gamma_{ss}/2\gamma_{sl})$ with γ_{sl} and γ_{ss} denoting the interface tensions solid/liquid and solid/solid.

shapes of the particles become simple spheres. New results of computations for the whole range of dihedral angles will be presented and analysed. The coupling between the roles of the dihedral angle ψ and the average particle coordination n_c will be especially emphasized. In Section 4, we will evaluate some applications of the model to practical situations.

2. Experimental facts

The initial motivation behind this study was the control of the processing of functionally graded WC–Co cermets for application as cutting tools and mining tools [8,9]. The phenomenon of liquid migration during liquid-phase sintering of these cermets has been the focus of a recent work by Colin et al. [10]. The starting materials in the latter work are cylinders of 11 mm diameter consisting of 4 different WC–Co cermets, combining two different mean WC particle size ($\approx$0.6 and $\approx$2.1 μm *i.e.* a size ratio S_r equal to about 3.5) and two different cobalt concentration ($\approx$6 and $\approx$15 wt.%). The cylinders had been sintered to full density by the company CERATIZIT (Mamer, Luxemburg). Pairs of these cylinders had been stacked on top of one another and bonding of this assembly was achieved by a heat-treatment for 7 h at 1400 °C under 0.5 bar of H_2. The two cylinders making the pair consisted of different cermet grades and had a different height. The variation of the composition and height of the two cylinders allows to cover a wide range of overall Co content in the final assembly. The reader is referred to Ref. [10] for additional experimental details.

In order to help figure out the experiment, Fig. 2 presents two scanning electron microscopy (SEM) images comparing the initial microstructures (*i.e.* before heat-treatment) of two cylinders which, in that particular case, consisted of a cermet with $\approx$2.1 μm WC size and $\approx$15 wt.% Co for the image on the left hand side and a cermet with $\approx$0.6 μm WC size and $\approx$6 wt.% Co for the image on the right hand side. The migration of the liquid from the one side of the bonding interface to the other side reflected in a change of the diameters and heights of the two cylinders. The migration kinetics has been monitored by measuring the evolution of these diameters and heights as a function of the heat-treatment duration. No significant further dimensional change could be measured after a couple of hours. The state reached after completion of 7 h of heat-treatment was



WC size ≈ 2.1 μm
Co content ≈15 wt.%

WC size ≈ 0.6 μm
Co content ≈ 6 wt.%

Fig. 2. Illustration of the principle of the experiment: SEM images (at the same magnification) showing the microstructures before heat-treatment of cylinders consisting of two different grades of WC–Co cermet which were brought into contact for being bonded by heat-treatment.