



# A sintering model for plasma-sprayed zirconia thermal barrier coatings. Part II: Coatings bonded to a rigid substrate

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## Abstract

The sintering model described in Part I, which relates to free-standing plasma-sprayed thermal barrier coatings, is extended here to the case of a coating attached to a rigid substrate. Through-thickness shrinkage measurements have been carried out for coatings attached to zirconia substrates, and these experimental data are compared with model predictions. The model is then used to explore the influence of the substrate material (zirconia vs. a nickel superalloy), and of the in-plane coating stiffness. Both differential thermal expansion stresses and tensile stresses arising from the constraint imposed on in-plane shrinkage can be relaxed via two diffusional mechanisms: Coble creep and microcrack opening. This relaxation allows progression towards densification, although the process is somewhat inhibited, compared with the case of a free-standing coating. Comparison of the stored elastic strain energy with the critical strain energy release rate for interfacial cracking allows estimates to be made of whether debonding is energetically favoured.<sup>1</sup>

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## 1. Introduction

The model described in Part I of this pair of papers [1] refers to unconstrained (detached) coatings. Obviously, under service conditions these coatings will remain attached to a substrate. At least in most cases, both the thickness and the stiffness of the coating will be significantly lower than those of the substrate, so that the in-plane dimensions of the coating will be forced to conform to those of the substrate. This naturally affects the progression of the sintering, and the associated changes in properties. It also tends to create residual stresses within the coating, although stress relaxation phenomena are likely to be operative at these elevated temperatures. Of course, stresses can also arise during sintering in the absence of a substrate, for example, as a result of differential shrinkage

between different parts, but in general the presence of a rigid substrate is expected to create larger stresses, plus there may be differential thermal contraction effects between substrate and coating as a consequence of changes in temperature.

There have been several previous studies of constrained sintering. For example, Bordia and coworkers published a series of papers [2–6] in the 1980s and 1990s, concerning the mechanisms by which constraint from the presence of a rigid substrate can give rise to stresses and damage, such as crack formation, and also exploring possible constitutive relations describing constrained sintering. Garino and coworkers [7–9] also attempted to quantify the effect of constraint on sintering rates, using essentially empirical relationships. Particular attention has been paid [10,11] to the anisotropic nature of the process and the recent review of Green et al. [12] also highlights this aspect. These treatments were based on the concept of a viscous Poisson coefficient. However, the handling of anisotropy in this way appeared to be rather incomplete and unsatisfactory. The effects of rigid inclusions and heterogeneities on the

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<sup>1</sup> A compiled version of the sintering model can be downloaded from [www.msm.cam.ac.uk/mmc/publications/software.html](http://www.msm.cam.ac.uk/mmc/publications/software.html).

progression of sintering have also been studied [5,13] and there have been several papers oriented towards multi-component and multi-layered systems [14,15], or towards the generation of defects such as cracks [6,14,16].

There have, however, been few attempts to develop sintering models based on the variational principle, in which the effect of constraint is incorporated. The recent paper of Hutchinson et al. [17] is aimed in this direction, but it contains no experimental information, although the modelled microstructure is oriented specifically towards plasma vapour deposited coatings. In the present paper, the theoretical framework outlined in Part I, which relates to a microstructure representative of plasma-sprayed coatings, is extended to encompass the effect of constraint. Comparisons are presented between predictions and experimental data, allowing the validity and limitations of the model to be explored.

## 2. Experimental procedures

### 2.1. Production of vacuum plasma-sprayed coatings on zirconia substrates

Samples for constrained sintering experiments were produced by vacuum plasma spraying (VPS) of yttria stabilized zirconia (YSZ) powder, with the composition given in Table 1, onto fully dense zirconia substrates. The higher particle velocities typical of VPS lead to improved coating adhesion. The spraying conditions are listed in Table 2. Technox 2000 zirconia tiles (Dynamic-Ceramic Ltd.) were employed, with dimensions  $50 \times 50 \times 5 \text{ mm}^3$  and a composition of 3 mol.% YSZ (tetragonal). They were grit blasted with alumina, in order to increase the surface roughness and promote coating adhesion. A thermocouple was spot welded onto a metallic plate, which was cemented to the back of the tile. The tile was preheated to about 600 °C immediately prior to spraying. Sintering experiments were carried out both on specimens cut from the sprayed tile and also on detached coatings obtained from the same sprayed material.

### 2.2. Dilatometry

Dilatometry was carried out in the through-thickness direction, on specimens attached to zirconia substrates, in air at 1400 °C. Dimensional changes were monitored using a DIL 402C Netzsch pushrod dilatometer. The through-thickness shrinkage of free-standing VPS and APS coatings was also measured. (Spraying conditions are given in Table 2.)

Table 2  
VPS and APS spraying conditions.

	VPS	APS
Chamber pressure (mbar)	200 (Ar)	Atmospheric
Substrate temperature (°C)	~600	~200
Plasma gun type	F4 (Plasma Technik VPS)	9 MB (Sulzer Metco Plasma System)
Nozzle diameter (mm)	8	6
Plasma gas flow rates ( $\text{l min}^{-1}$ )	Ar, 50 H <sub>2</sub> , 8	N <sub>2</sub> , 35.4 H <sub>2</sub> , 8
Carrier gas flow rates ( $\text{l min}^{-1}$ )	Ar, 4	N <sub>2</sub> , 5.2
Arc current (A)	750	500
Voltage (V)	50	78.6
Power (kW)	37.5	39.3
Stand-off distance (mm)	400 (preheat) 250 (spraying)	114

### 2.3. Microstructural examination

Microstructural examinations were carried out as detailed in Part I. It should be noted that the microstructure of the VPS coatings was rather different from that of the APS coatings shown in Part I. This is illustrated by Fig. 1, which shows low and high magnification views. It can be seen in Fig. 1b that the splat structure is rather finer than that of the APS coatings (see Fig. 1 in Part I), both in terms of the splat thickness and, particularly, in terms of typical spacing between inter-splat contact points.

## 3. Constrained sintering model framework

An extension to the nomenclature of Part I is shown in Table 3.

### 3.1. Model geometry

The model geometry of Part I is also employed here, with the additional constraint that the coating is attached to a flat, rigid substrate of infinite extent in the in-plane directions (i.e. the in-plane dimensions are much greater than the coating thickness and there are no edge effects). In the predictions presented here, the substrate is either zirconia or a nickel superalloy, with the possibility of differential thermal contraction misfit strains being absent in the first case and present in the second. The model concerns isothermal sintering, with no thermal gradients present and no externally applied forces or bending moments. It therefore relates to homogeneous constrained sintering, with uniform strain and stress fields through the thickness of the coating, no strain gradients or specimen curvatures and no strains or stresses in the substrate (due to its substantially greater stiffness and thickness).

Table 1  
Chemical composition (wt.%) of powder supplied by Sulzer Metco Inc.

ZrO <sub>2</sub>	Y <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	CaO	Fe <sub>2</sub> O <sub>3</sub>	HfO <sub>2</sub>	MgO	Th	TiO <sub>2</sub>	U
Rem.	7.41	0.02	0.07	<0.01	<0.01	1.62	<0.01	<0.002	0.08	<0.002

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