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Spark Plasma Sintering tool design for preparing alumina-based Functionally Graded Materials

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Abstract

A way to produce Functionally Graded Materials (FGM) is by means of Spark Plasma Sintering (SPS) and specifically designated tools. These new tools permit a current density modulation and therefore a temperature variation along the *z*-axis. The key feature relies on a varying die section. FEM modelling has given the suitable range of die dimensions between the top and the bottom to obtain a given temperature gradient (around 300 °C) out of roughly a 15 mm height. Experiments conducted in different configurations (with or without samples) and the measurement of the associated thermal gradient led to improvements of the mould (in particular the introduction of a counter-piston). By the use of this specific mould, an alumina showcasing a microstructure gradient was sintered: from a dense face exhibiting 1.3 µm grains to a porous one composed of 200 nm grains.

Keywords: Spark Plasma sintering; Functionally Graded Material; FEM modelling; Alumina

1. Introduction

A Functionally Graded Material (FGM) is a material presenting a gradient of composition, phase, porosity or texture which leads to a gradient of properties such as hardness, density, thermal conductivity, elasticity, etc. [1]. The integration of materials with different functional properties within the same product is a growing area in manufacturing industries, especially in the fields of nuclear energy [2], defence [3] or aeronautics [4]. Other potential applications are tools, environmental sensors, biomaterials, optical and electronical materials [5]. The functional gradient most often originates from a chemical composition gradient [6]. Kieback et al. [7] classified them into two categories: (i) a gradient of chemical composition in a single-phase material, when a solid solution between the chemical constituents exists in the composition range; (ii) a gradient of the various phase volume fractions in a multi-phase material. FGMs offer the advantage of optimizing functionality by a suitable distribution of the materials/microstructures within a unique product while removing stress singularities at sharp bi-material interfaces. Indeed, a compositional gradient provides a "smooth and mastered" transition between different materials which limits thermal expansion differences and reduces thermal stress under various solicitations. With ultrasound and hardness tests in Ni/Cu FGMs, Rubio et al. [8] demonstrated that a mechanical property gradation was achieved along the direction of stacking. Zhang et al. [9] used finite element modelling to optimize the compositional distribution in Al_2TiO_5/Al_2O_3-based FGMs and therefore found this "smooth and mastered" transition.

A possible way to produce FGMs is powder metallurgy, via the thermal processing of multilayer powder stacking [10]. For example, porosity gradients were in general produced by varying the particle size in the stacking [11] or the blowing agent (such as NaCl) fraction in the layers [12]. However, Spark Plasma Sintering (SPS) permits the production of such materials from a unique powder thanks to the features of the process itself [13]. Indeed, during an SPS cycle, the temperature distribution within the sample depends on its electrical

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conductivity, its relative position to the die, the characteristics of the die, the heating rate, and even the applied pressure. Anselmi et al. [14] investigated the current and temperature distributions in alumina and copper specimens both by simulation and experiments. Significant differences in Joule heat distribution were observed for both cases. Vanmeensel et al. [15] observed an electrical behaviour change in ZrO_2-based composites containing TiN (electrically conductive) once percolation has occurred. Minier et al. [16] showed that nickel specimens heated using identical sintering parameters exhibited different microstructures depending on the material of the mould that was used (alumina or graphite). In alumina and nickel samples, they also observed that the larger the die diameter, the bigger the alumina grains and the finer the nickel ones [17]. However, this statement is only true for a given sample size as Rathel et al. [18] showed by studying the influence of the die dimensions on the microstructure of Si_3N_4 and WC larger samples. The non-linear correlation between die sizes and temperature distributions was also demonstrated by Wang et al. [19] in their calculations on alumina compacts. Hulbert et al. [20] fabricated porous boron carbide, by offsetting the die relative to the punches. They did not measure the extent of the temperature gradient but estimated it by numerical modelling (405 °C over 15 mm in height). Carneiro et al. [21] produced cutting tools in Al_2O_3–ZrO_2 + WC–Co or Al_2O_3–TiC + WC–Co. Belmonte et al. [22] went further by varying the contact sections between the mould and the graphite stack (lower plunger completely enclosed in the die) to control the $\alpha \rightarrow \beta$ phase transformation in silicon nitride specimens. The thermal gradient (between 50 and 150 °C) was only confirmed by the microstructure once again. They increased the thermal gradient (higher than 150 °C afterwards) by placing an electrical insulator barrier (boron nitride) between the powder compact and the upper punch [23]. Others varied the graphite section along the z-axis [24]. With such a pre-designed mould, Omori et al. [25] and Hong et al. [26] successfully produced Cu/PI and ZrB_2–SiC/ZrO_2(3Y)-based FGMs respectively. Holland et al. [27] used high heating rates (1000 °C/min) to obtain a radial temperature gradient in boron carbide hollow cylinders. The internal surface was then fully dense whereas an open network of pores ($<$90% dense) was present on the external surface. For a symmetrical set-up Yucheng et al. [28] proposed a model to calculate the axial and radial thermal gradients. These equations were clearly dependent on the heating rate. The calculated and experimental values of the radial temperature gradient are not in good agreement due to the steady-state conditions for the calculations. Wei et al. [29] showed in TiB/Ti-based FGMs that the heating rate increase has also led to a stress gradient increase. Lee et al. [30] showed by simulation that, in the case of the rapid water quenching of porous alumina (porosity of 20%), the thermally induced stress was mainly determined by the elastic modulus of the microstructure. Consequently, in their example, a partially sintered alumina was found to be advantageous to prevent thermal shock damage. Mignard et al. [31] evaluated the critical thermal shock temperature difference of coarse grain porous alumina to be 700 °C. Grasso et al. [32] investigated the pressure effect on the temperature distribution in the case of a graphite specimen and concluded that the vertical contact resistance (between punch and die) directly affected the temperature gradient along the radius of the assembly. The temperature gradient decrease was associated with an external pressure increase because pressure influences the electrical and thermal fields by modifying both the electrical and thermal contact resistances and the characteristics of the powder compact. Zavaliangos et al. [33], Vanmeensel et al. [34] incorporated thermal contact resistance in their model to obtain a match between simulated results and experimental data. The moving mesh approach respectively allowed Maizza et al. [35] and Song et al. [36] to simulate the geometrical changes due to sintering in the case of tungsten carbide and iron powders. Recently, the literature has presented investigations of temperature homogeneity in more complex geometries than merely asymmetrical ones [37].

In the present paper, the main objective is to design SPS tools to induce large thermal gradients within a powder compact. Given that it is expected that the latter will depend on the thermal gradient inside the die, the first part of this work will deal with temperature measurements inside a classical die for different configurations (with or without samples). From these thermal considerations, a specific die (including a counter-piston) was designed and the possibility of inducing a large thermal gradient inside an alumina powder compact was evaluated through the evolution of the microstructure. This gradient close to 300 °C corresponds to the temperature difference between the onset and the end of densification of the powder. Although many studies correlate experimental and numerical data, the analysis was quite limited. The radial temperature gradients were correlated with the microstructure evolution and/or with at most two temperature measurements. In this study, we propose four temperature measurements that highlight the possible experimental variations in the process.

2. Experimental procedure

FEM modelling was performed using Dassault systèmes software (Abaqus® 6.13) and an axisymmetrical thermal-electric model, to design an adapted SPS tool (die dimensions especially), capable to create such a wide temperature gradient. The die shape is inspired by studies published by Tokita [13], with a graphite axially-varying section. This die is hereafter referred to as a classical gradient mould (Fig. 1a). The inner diameter of the die was set to 20 mm and its height to 43 mm. The smallest section was also set: the outer diameter was 32 mm and its height was 20 mm. The numerically modelled SPS assembly was the configuration (without sample) shown in Fig. 1b. The initial and boundary conditions were as follows

1. The initial temperature was set at 296 K.
2. The temperatures of the upper and lower copper electrodes were held constant at 296 K.
3. Lateral heat losses through conduction or convection were not taken into account. All the lateral surfaces of the assembly are