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Influence of conductive secondary phase on thermal gradients development during Spark Plasma Sintering (SPS) of ceramic composites

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ABSTRACT

Spark Plasma Sintering (SPS) has attracted a lot of interest in recent years owing to its ability to enable the densification of a broad range of materials in a very short processing time. It is well documented in the literature that the very high heating rates that can be applied with this technology can lead to the apparition of large thermal gradients in the tool and thus affect the homogeneity of the compact.

In the present study, the influence of the compact thermal and electrical properties on the thermal gradients was studied. Al_2O_3, AlN and TiC powders were used to produce series of Al_2O_3-TiC and AlN-TiC composites (0, 25, 50, 75, 100 vol%TiC) showing different electrical and thermal conductivities. Two pyrometers were used in order to observe and measure the thermal gradients and the percolation of the current during sintering at a high heating rate and without insulation.

Electrical conductivity measurements were carried out on samples presenting different relative densities. This samples were obtained through interrupted sintering cycles at temperatures below and above the identified percolation threshold temperature.

It was shown that high thermal gradients can appear during SPS depending on the processing parameters (dimensions of the die and heating rate) but also on the composition of the compact (proportion of conductive phase) and on its density.

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1. Introduction

SPS (Spark Plasma Sintering) is a rapid sintering technique that has attracted a lot of interest in recent years. It is a hot pressing technology that enables the densification of a broad range of materials in a very short processing time and at lower temperatures compared with more conventional sintering technologies. The sintering cycle is carried out in a chamber under vacuum or inert atmosphere. The tool (cylindrical die and punches) material is usually graphite. An electric current is directly applied to the punches and die system which heats by Joule effect (as well as the powder if it is electrically conductive) at very high heating rates (up to 200–1000 °C/min depending on the equipment used [1]). The low thermal inertia of the system and the absence of a massive insulation within the water cooled vessel, allow the shortening of the cooling step as well, thereby reducing significantly the overall thermal cycle.

Due to the very high heating rates, thermal gradients can appear in the SPS tool between its centre and the external part of the die [2] leading to a lack of homogeneity in the sample. Indeed, the temperature difference in the compact should ideally be close to zero in order to obtain a homogeneous sample.

Thermal gradients depend on several parameters like the conductivities of both the sample and the die, the sample dimensions, the current distribution during sintering, etc. SPS suppliers have already found solutions aiming to reduce these gradients, for example by using spacers between the powder and the punches (in order to influence the current distribution) or by using a thermal insulation around the die. They also have partially solve this problem for larger specimens (> 40 mm) through the use of hybrid systems aiming to reduce the radial thermal gradients by secondary resistive or inductive heating (this is of major interest for conductive materials).

However, it is sometimes necessary to add a correction to the measured temperature in order to be sure that the sample reach the desired temperature. This correction is dependent on the SPS

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equipment and on the way of controlling the temperature [3]. The electrical properties of the compact to be sintered has a great influence on the radial temperature gradient that can be observed in the system and in the sample. In the case of a high electrical conductor, the main part of the current flows through the sample while, on the contrary, the current is forced to flow through the die in the case of a strong electrical insulator [4–6]. The temperature control can thus be complicated when the sample goes from an insulating to a conductive behaviour during the densification process [7,8]. For example, a composite (powder or bulk) made of both an electrically conductive and an electrically insulating phases can be conductive or not. The electrical conductivity of the whole compact depends on the amount of conductive phase and on the number of contacts between conductive grains. This is known as "site-bond percolation" [9]. The electrical behaviour of the compact can also change over time, during the densification stage. Indeed, an insulating composite can become conductive if the amount of conductive phase is high enough and if the number on contacts between conductive grains increases during densification. It was also shown in the literature that the percolation threshold depends on the conductive phase (filler) grain size [10]. As a consequence, as the current applied in SPS contributes to the direct heating of the die/punches tool by Joule effect, it can also contribute directly to the heating of the compact in case of percolation.

The objective of this study was to highlight the creation of thermal gradients in the SPS tool during the sintering of ceramic composites and their influence on the temperature control during the densification process.

Al_2O_3-TiC or AlN-TiC composites should behave in a different manner regarding to direct heating of the samples and to the heat transfer to and/or from the die/punches tool. Indeed, Al_2O_3, AlN and TiC have opposite thermal and electrical properties (Table 1). Al_2O_3 is an electrically and thermally insulating material whereas AlN is thermally conductive and electrically insulating. TiC is both electrically and thermally conductive.

Unlike alumina [14], aluminium nitride is difficult to sinter without additives as reviewed elsewhere [15]. Indeed, AlN is a covalent compound with limited atomic mobility and it is thus difficult to fully sinter this material at low temperatures. Moreover, it is known that AlN decomposes at temperatures higher than 1600 °C [16]. That is why AlN is usually densified under high pressure with additives, the most used sintering aids being Y_2O_3, CaO or CaF_2 [17].

Due to its limited sinterability, TiC reduces strongly the sintering kinetics of alumina [18]. It forms an interconnected network that prevents to reach the maximum density. It is thus required to sinter at a higher temperature in order to increase the driving force for sintering, resulting however in matrix grain growth, or to heat at a lower temperature with the application of a uniaxial pressure and possibly adding sintering aids.

For Al_2O_3-TiC, the sintering aid is usually Y_2O_3 [19,20]. However, SPS could prevent the use of sintering additives but only limited information is available in the literature concerning the SPS densification of such composites [21–23]. They can be used, for example, for cutting and wear tools (with a TiC content of around 30 wt%) or as a substrate of magnetic heads due to their attractive mechanical properties (high temperature strength and thermal shock resistance) and good electrical conductivity [24].

AlN-based materials find application in refractories or in the electronic field due to their high thermal conductivity combined with their electrical insulating properties. The literature concerning AlN-TiC materials is poor as the main field of application of AlN substrate is in electronics where the electrical insulation is the major requirement. In this work, these composites have thus been chosen as a matter of comparison with Al_2O_3-TiC, to assess the influence of the thermal insulating behaviour or the heat transfer between the composite and the die.

2. Experimental

Commercial powders (Al_2O_3, AlN and TiC) were used as raw materials. Table 2 presents the grades that were chosen as well as their initial grain size (D50) and main impurities.

They were chosen for their different electrical and thermal properties. As said in the introduction, Al_2O_3 and AlN are electrically insulating powders ($\rho_{élec} > 10^{13}\ \Omega$ m) but they have different thermal conductivities: Al_2O_3 is thermally insulating ($\lambda \sim 30$–$40\ W\ m^{-1}\ K^{-1}$) while AlN presents good thermal properties ($\lambda \sim 200$–$300\ W\ m^{-1}\ K^{-1}$). TiC is a conductive material ($\rho_{élec} \sim 0.5 \times 10^{-6}\ \Omega$ m; $\lambda = 21\ W\ m^{-1}\ K^{-1}$) which was used as a second phase in Al_2O_3- and AlN-based composites in different proportions (25, 50 and 75 vol%) so that Al_2O_3-TiC and AlN-TiC composites with different electrical and thermal properties were produced.

Al_2O_3-TiC mixtures containing 25, 50 and 75 vol% of TiC were prepared by ball-milling during one night in isopropanol. Al_2O_3-TiC mixtures were dried in a rotational evaporator and sieved at 500 μm in order to break the larger agglomerates.

Due to the hygroscopic behaviour of AlN, AlN-TiC mixtures (25, 50 and 75 vol% of TiC) were mixed in dry conditions and sieved at 500 μm as well.

SPS experiments were conducted in an HPD25/1 equipment (FCT System, Germany) using a fixed pulse pattern of 10:5 ms on:off. The temperature was axially measured by an optical pyrometer focused on a point situated near the upper surface of the sample through a hole drilled in the upper punch (central pyrometer "CP").

Al_2O_3-TiC and AlN-TiC compacts were sintered at different temperatures in order to obtain samples with different densities (interrupted cycles). These sintering experiments were conducted in a die of 30 mm in diameter covered with a graphite felt insulation (in order to improve thermal insulation), with a heating rate of 200 °C/min and an applied load of 50 MPa. Table 3 summarizes the temperature and dwell time conditions of these tests. The density of each sample was measured using the Archimedes' immersion method (in water for Al_2O_3-TiC samples and in mercury for AlN-TiC samples).

The electrical conductivity of the composites was measured by the 4-points method (RZ-2001i, Osawa Science) between 25 and

Table 1
Electrical and thermal properties of Al_2O_3, AlN and TiC (from [11–13]).

Powder	λ ($W\ m^{-1}\ K^{-1}$)	$\rho_{élec}$ (Ω m)
Al_2O_3	30–40	$> 10^{15}$
AlN	320 (Pure crystal) 180–220 (Sintered)	$> 10^{13}$
TiC	21	0.5×10^{-6}

Table 2
Raw materials.

Powder	Supplier	Grade	D50 (μm)	Impurities
Al_2O_3	Alcan	P172SB	0.4	600 ppm Na_2O, 900 ppm SiO_2, 600 ppm CaO, 120 ppm Fe_2O_3, 900 ppm MgO
AlN	H.C. Starck	Grade C	0.8–1.8	Fe < 50 ppm, O < 2%
TiC	H.C. Starck	HV120	1.0–1.5	free C, O max 1.3%

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