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Direct microwave sintering of pure alumina in a single mode cavity: Grain size and phase transformation effects

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ABSTRACT

A comparative study between conventional and direct microwave sintering of pure α and γ nanoscale alumina powders has been performed to identify possible microwave effects on densification and microstructure changes. Microwave sintering experiments have been performed in a 2.45 GHz single mode cavity furnace allowing for an accurate control of the thermal cycle and for a continuous measurement of specimen dimensional changes, so that direct comparison with conventional sintering can be achieved. Special attention has been given to the influence of particle grain size and of the γ to α phase transformation occurring during heating of γ alumina powder on the sintering behaviour. Experimental data unequivocally showed a significant effect of microwaves on α powders (lower onset temperature of densification and smaller activation energy) and on γ powders (lower phase transformation temperature). However, the sintering trajectory in grain size vs density diagram is similar in conventional and microwave sintering. Therefore, microwave heating was not beneficial to obtain dense alumina with very fine grains. Microwave effects have been explained through the ponderomotive force induced by the electromagnetic field and acting on diffusion and phase transformation mechanisms.

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1. Introduction

Microwave sintering as a rapid processing technique has been extensively investigated in the past decades [1–4]. The reported benefits of this technique on material microstructure and properties are mainly related to its specific heating process. During microwave heating, the material couples with the electric field, absorbs the electric energy in its bulk and transforms it into heat. The coupling behaviour depends on the dielectric loss factor of the material. According to the magnitude of this factor, a material can be classified as opaque (conductor), transparent (low loss insulator) or absorbing (high loss insulator). At room temperature, most oxide dielectric ceramics have a low loss factor and they heat up very slowly under microwaves. This is the case for alumina or zirconia that have a loss factor at room temperature of tan δ ~ 0.045% [5] and tan δ ~ 0.4% [6], respectively at 2.45 GHz. The loss dielectric factor generally increases with increasing temperature. In most studies displayed in the literature, the use of a susceptor with a high dielectric loss factor allows heating the green specimen up to a temperature at which it shows a significant coupling with the microwaves. In that case the heating process is a combination of conventional radiative heating by the susceptor and direct microwave heating; this process may be referred to as "hybrid microwave sintering".

Concerning alumina, the dielectric properties of this material depend strongly on doping elements such as magnesium [7] and yttrium [8] or non-controlled impurities especially in the microwave frequency range. In the literature, most studies deal with doped powders sintered in a multimode cavity comprising a susceptor. In these conditions, Brosnan et al. [9] found a spectacular effect of microwaves on the densification of a mixture of α and γ powders doped with Y_2O_3 and MgO. Full densification was obtained 250 °C below the one required in conventional sintering,

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whereas the sintering trajectory in a grain size vs relative density diagram was similar with both heating techniques. It should be noted however that the heating cycles in microwave and conventional experiments were not the same. Also the temperature of the specimen during microwave sintering could have been underestimated due to inadequate calibration of the pyrometer. Brosnan et al. proposed that microwaves enhance diffusion mechanisms. Xie et al. [10] reached the same conclusion after working on a pure α alumina powder in the same conditions. Dé et al. [11] showed that a pure α alumina powder sintered in a multimode cavity containing a susceptor is finer and more homogeneous than after conventional sintering at very high heating rate but they assumed that it was only a consequence of thermal effects. On the opposite, Zhao et al. [12] did not observe any difference between the mechanical properties of pure α alumina samples sintered either by conventional heating or by microwave heating in a single mode cavity, once again with a susceptor. They unsuccessfully tried direct microwave heating, i.e., without a susceptor. As far as we know, the only authors that presented direct microwave sintering experiments with α alumina powders used high frequency fields (28 GHz and above). Sudiana et al. [13] sintered pure α alumina powder under microwaves at 28 and 300 GHz. They showed a decrease of the sintering temperature that depended on the frequency. This decrease was higher at 28 GHz than at 300 GHz, contrary to what was expected since the dissipated power is supposed to increase with increasing frequency. Fliflet et al. [14] also heated pure α alumina using a high frequency of 35 GHz but they did not observe any effect of microwaves on the densification behaviour. Therefore, whatever the frequency is, the effects of microwaves on sintering are variable and dubious.

Concerning metastable alumina, numerous conventional sintering studies have been conducted with ultrafine powders to try obtaining dense nano-crystalline material. However, these studies do not lead to materials with smaller grain than the ones obtained starting from alpha powders [15]. Possible benefit of microwaves has been investigated by a few authors. Rybakov et al. [16] observed an 80 °C decrease of the temperature of phase transformation under microwaves and they found that the extent of phase transformation depends on the microwave intensity. On the contrary, Freim et al. [17] obtained neither an effect of microwaves on the γ to α phase transformation nor a smaller grain size after similar densification in comparison with conventional sintering.

Hence, although many studies have been conducted on microwave sintering of ceramics, the reasons for specific microwave effects on sintering and even the existence of such effects is still under debate. This is because most of the research works lack systematic and valuable comparison between conventional and microwave sintering for identical thermal cycles, mainly due to limited or inaccurate experimental data (in particular, unreliable temperature measurement and no dilatometry measurement). Also, the use of hybrid heating makes the identification of microwave effects more difficult, since the nature and geometry of susceptors directly affect the electromagnetic field distribution and intensity inside the samples.

In a recent paper, Croquesel et al. [18] presented a single mode cavity furnace that permitted direct microwave heating of pure alumina powder. This cavity has been optimized with an impedance tuner so as to provide maximum electric power to the specimen and with a motorized plunger for the control of thermal cycle. Also it includes an optical dilatometry device that allows following the shrinkage of the specimen in the course of sintering and a pyrometer that has been properly calibrated for reliable temperature measurement of the specimen. With this equipment, microwave sintering can be rigorously compared with conventional sintering and specific effects of microwaves, if any, can be evidenced.

The goal of this study is thus to clear up the effect of microwaves on the sintering of alumina powders thanks to this original setup. Several α and γ alumina powders with various grain sizes have been sintered by direct microwave heating and conventional heating with exactly the same thermal cycles. Comparing the densification curves and microstructures obtained in each case allows us identifying specific microwave effects. Concerning γ alumina, the process of phase transformation during heating is particularly analysed. Finally, explanations of identified microwave effects are proposed by using the ponderomotive force theory.

2. Materials and experiments

High-purity commercial Baikalox (Baikowski International, France) α-Al_2O_3 powders with different specific surface areas (19 and 6 m^2/g, corresponding to 80 and 250 nm equivalent crystallite sizes, respectively) and γ-Al_2O_3 powders containing different amounts of 100 nm alpha particles (9.4 and 3.3 wt%) were used as starting materials. In the following, α powders will be called A_{19} and A_6, respectively, whereas γ powders will be called G_9 and G_3 respectively. Table 1 displays the median particle size, D_{50}, the specific surface area, S_{BET}, the equivalent BET crystallite size, D_{BET}, the α content and the residual impurity contents (Na, K, Fe, Si, Ca) for each powder.

Cylindrical compacts with 8 mm diameter and 8 mm thickness for conventional sintering and with 8 mm diameter and 4 mm thickness for microwave sintering were pressed by double-effect uniaxial pressing (35–50 MPa) followed by cold isostatic pressing (200–450 MPa). The pressure was adapted to each powder so as to obtain green compacts with the same relative density, 48%. The relative density is defined as the weight density divided by the weight density of the fully dense material. For γ powders, the final relative density is calculated using the theoretical density of the α phase with the assumption that the sintered material is fully transformed to α phase. Compacts were heated in a conventional furnace at 2.5 °C/min up to 600 °C with a dwell time of 1 h in air to remove possible organic additives and absorbed water. Total weight loss after this treatment was about 2% for α-powders and about 4% for γ powders.

The parameters of sintering experiments are shown in Table 2. Conventional sintering was carried out in air using a dilatometer (Setsys Evolution TMA, SETARAM, France) at constant heating rate of 1.6, 4, 10 and 25 °C/min for A_{19} powder and 25 °C/min for A_6, G_3 and G_9 powders. Samples were heated up to 1550 °C with a holding time of 5 min. Microwave sintering was performed in a 2 kW, 2.45 GHz single mode cavity (SAIREM, France) equipped with standard WR 340 waveguide and samples were located in a position where the electric field was supposed to be maximal. Details of the overall process, including experimental conditions, were previously reported by Croquesel et al. [18]. A_{19} samples were heated up to 1510 °C at 10, 56 and 158 °C/min and 1550 °C at 25 °C/min with a holding time of 5 min A_6, G_3 and G_9 samples were heated up to 1550 °C at 25 °C/min with a holding time of 5 min.

Initial and final relative densities were measured by the geometrical method and the Archimedes method, respectively. Density changes were calculated from the sample diameter variation continuously measured by optical dilatometry [18]. For this purpose, the shrinkage was assumed to be isotropic. The microstructure of sintered samples was observed on longitudinal cross sections by scanning electron microscopy (Ultra 55, Zeiss, Germany). Apparent grain size was measured by the intercept method on at least 250 grains on polished and thermally etched surfaces for high densities samples (>90% T.D.) and on fractured surfaces for samples with a lower density. A statistical correction factor of 1.56

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