Microwave Sintering of Micro- and Nano-Sized Alumina Powder

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In order to get very dense material, it is convenient to mix micro- and nanoparticles so that the smallest grains fill the space between grains of bigger size. Microwave heating is well-suited to sintering compact of this type because the threshold process which appears in the nanoparticles induces homogeneous heating, further relayed by electrical conductivity in the entire compact.

Introduction

When traditional sintering techniques are used it is difficult to obtain dense materials with nanometric or submicrometric grain sizes. Microwave sintering was proposed as an alternative technique to overcome the problems of conventional fast heating. However, various problems usually occur when ceramics are sintered by direct microwave heating [1]. Hybrid heating techniques, which combine direct microwave heating with infrared heat sources were proposed [2].

Microwave sintering of micro- and nano- alumina powders mixed together was studied in a recent past, in the frame of a cooperation between the Universities of Krasnoyarsk and Toulouse-INP. Toulouse worked with Pechiney powders and Krasnoyarsk with homemade powders by explosion in a bomb. The aim was to obtain pills as dense as possible. To aid in understanding the process of microwave sintering, we created a 3D model corresponding to the experiment performed.

Experimental Procedures

The materials used were two kinds of alumina powders: nano-sized and submicron fractions. Alumina nanopowders were synthesized by the explosion technique described in [3]. The properties of these powders were studied in detail in [4, 5]. As a submicron fraction a commercial P172SB alumina powder (Pechiney, France) was used. Properties of the powders are summarized in the Table 1.

The composition of nano and microsized fraction that gives the best mechanical properties, was experimentally determined to be $Al_2O_3^c$(P172SB)–10% vol. $Al_2O_3^f$ [6]. The powders were mixed, and then compacted into a disk of diameter 16 mm by cold uniaxial pressing at 200 MPa.

A special microwave oven was designed to introduce homemade lossy ceramics to produce infrared heating of the sample surface necessary to get a homogeneous temperature profile. The heating process included different steps: a heating slope of 100°/mn up to 1300°C, a dwell for 30 mn (to transform $\delta+\theta$ alumina into α phase), the last slope of 100°/mn up to 1600°C and a last dwell during 30 mn. The temperature slowly decrease in the oven over the course of 2.5 hours. The temperature in the microwave furnace was controlled by a pyrometer directly focussed on the surface of the sample.

Table 1. Properties of Alumina Nanopowders

Properties of powders	Nanosized Al_2O_3	Submicron Al_2O_3 P172SB
Particle-size	$d_{0,5}$ = 0,077 μm	$d_{0,5}$ = 0,52 μm
Distribution	σ = 1,68	σ = 1,58
Morphology	spherical	lamellar
Specific surface area S, m^2/g (BET)	20,7	7,1
Phase composition	$\delta+\theta$	α
Density, g/cm^3	3,72	3,97
Bulk density, γ, g/cm^3	0,3	1,3

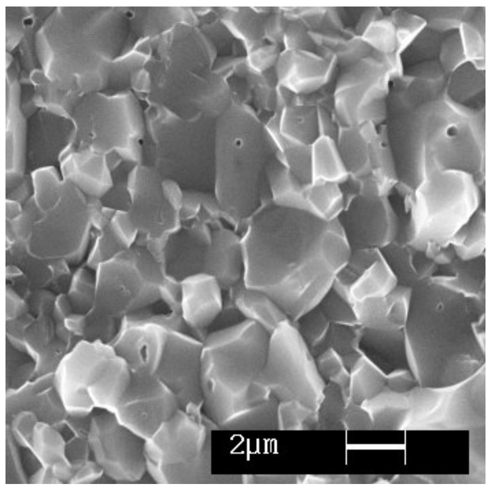

Fig. 1 SEM micrographs of the ceramic $Al_2O_3^c$(P172SB) – $Al_2O_3^f$ obtained.

The final densities of the compacts were measured by the Archimedes method. The sintered samples reached densities of 98% with uniform microstructures without significant grain growth. The microstructures of the microwave-sintered samples were observed by scanning electron microscopy (SEM) (Fig. 1).

As a result the following physical and mechanical properties of sintered ceramic were obtained:

Grain size (d, μm)	5-7
Density (ρ, g/sm^3)	3.82-3.91
Young's modulus (E, GPa)	360-370
Poisson's ratio (ν)	0.23-0.26
Bending strength ($\sigma_в$, MPa)	350-400
Vickers hardness (HV, GPa)	17-20
Indentation fracture toughness, (K_{Ic}, $MPa \cdot m^{1/2}$)	5-5,6

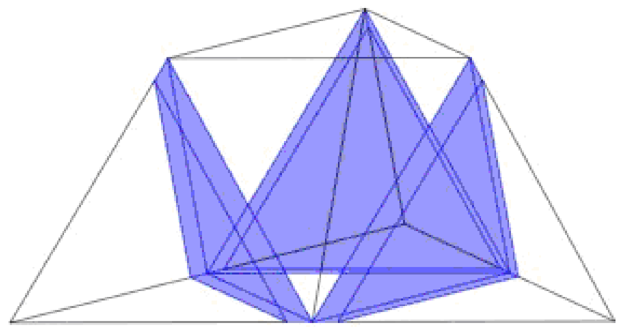

Fig. 2 Model of alumina grains

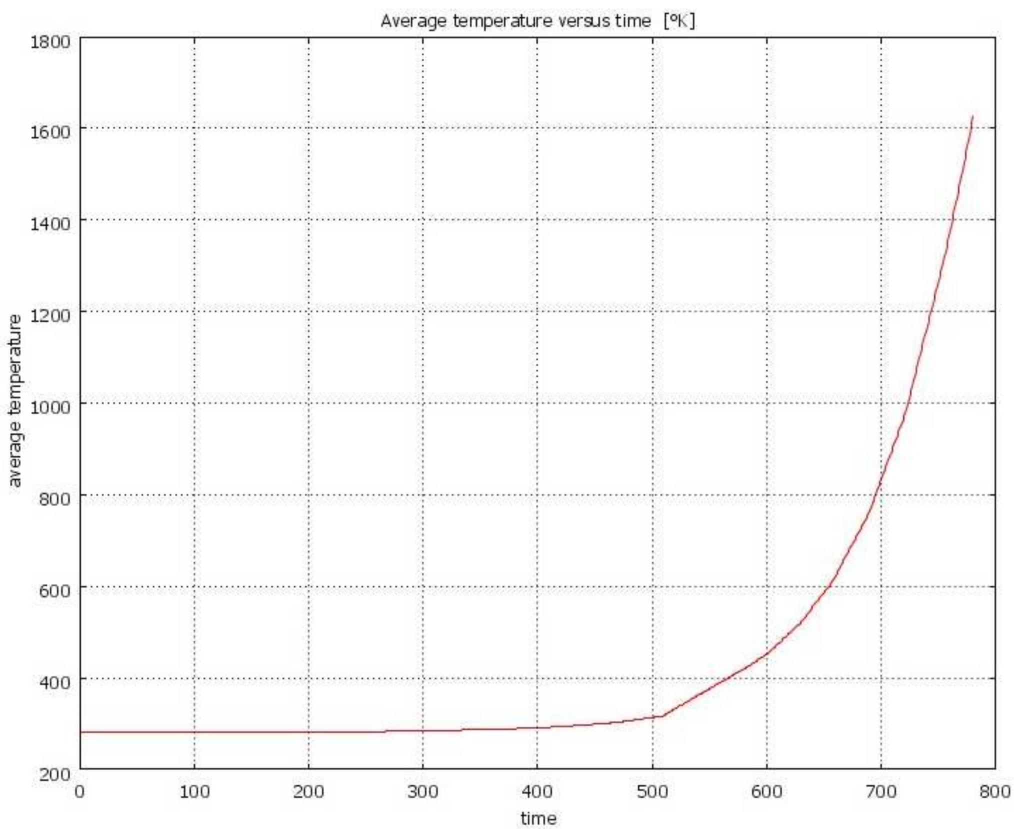

Fig. 3 Average temperature during the heating

Modeling

We created a special simulation model using Comsol Multiphysics software to describe the heating process. The model's geometry consists of a truncated pyramid (Fig. 2) comprised by 7 regular tetrahedrons (4 are kept compact to simulate a big particle) and three layers of free space inside the others, which are marked in Fig. 2 in blue color. The seven tetrahedrons represent bigger grains of alumina micropowder ($d_{0,5}$=0.52 μm), and the free space is filled by the alumina nanopowder grains of smaller size ($d_{0,5}$= 0.077 μm). The volumes of the bigger and the smaller grains are in ratio 9:1, as in the experiment.

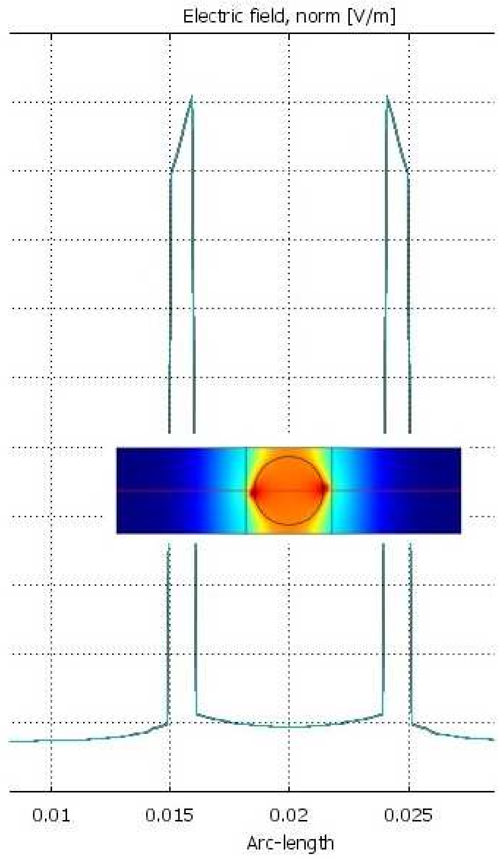

Fig. 4 Threshold process

To describe the process of sintering, the following boundary conditions are chosen: the larger base of the pyramid is "Ground", the smaller base is "Electric potential" (we put 1.2e-4 V there) and the lateral faces are "Electric insulation". The thermal source was microwave power Q_{av} and the electrical conductivity is proportional to the temperature [7]. Simulation lasted for 13 minutes, and showed that temperatures up to 1300ºC were reached, as shown in Fig. 3.

The Fig. 4 shows the threshold process around the smallest grains. We assume that a thin layer of air surrounds these grains; the difference of permittivity produces a high electric field, shown by the blue lines, which starts the electric conduction simulated by $\sigma \approx \sigma_0(|E|>E_0)$, where E_0 is the threshold field. This conduction dissipates electric energy which increases the temperature shown by the two red points; after some time the conductivity/dielectric losses also appear in the micro grains.

Conclusion

This paper has shown the experimental results of the microwave sintering of a compound made of two types of alumina grains; one type of micrograin was obtained from Pechiney, while the other nanopowder was made by the University of Kranoyarsk. The objective was to get a blend as dense as possible; we got almost the theoretical value. The sintering was performed in a microwave oven fed by two 1 kW, 2.45 GHz magnetrons in an isolated chamber. Moreover, in the surroundings of the compound, microwave susceptors were added to allow for infrared heating of the surface. This (volume+surface) heating gave a good homogeneity. The choice of microwave energy was a good one as it was expected previously and later shown by modeling. In fact, heating of the nanopowder acts like seeds disseminated throughout the volume. It produces a homogeneous heat pattern when the micrograins become heated by themselves.

To obtain material of very high density it is necessary to use different granulometries, and this blend fits perfectly well with compound microwave heating to produce uniform sintering.

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