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Modeling of the temperature distribution of flash sintered zirconia

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Recently, the number of published papers on the sintering technologies activated by current have increased exponentially. In particular, it has been reported that the application of electric field as high as 120 V/cm permitted the instantaneous full densification of yttria stabilized tetragonal zirconia at the unusual low temperature of 850°C. The mechanisms of the so called flash sintering phenomenon are elucidated by analyzing the temperature distribution of the bulk sample under the application of the electric field.

Key-words : FEM simulation, Flash sintering, Zirconia

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1. Introduction

Electric Current Assisted/Activated Sintering (ECAS)[1] is a class of consolidation methods in which the electric and the thermal fields are combined to enhance interparticle bonding and densification. The primary purpose of imposed electric currents is to provide the required amount of resistive heat. Moreover, electric currents may additionally enhance powder sintering by activating one or more concurring mechanisms, such as surface oxide removal, electromigration and electroplasticity. The use electromagnetic and electrical fields have been demonstrated effective to enhance the sintering kinetics in several techniques.[1]

Recently a new type of sintering method named "Flash Sintering" has been reported.[2] Cologna et al. showed that yttria stabilized tetragonal zirconia can be fully densified in a 5 s at 850°C under the application of a DC electrical field. However, by employing conventional sintering several hours at temperature higher than 1200°C are needed to achieve fully dense material.[3] The flash sintering phenomenon was explained by the local Joule heating at grain boundaries, which promotes grain boundary diffusion enhancing the sintering kinetic. Similarly Prette et al. extended the flash sintering method to fully densify Co_2MnO_4 materials.[4]

The present work aims to predict the temperature distribution of the zirconia samples under the flash sintering condition by means of FEM simulations.

2. Experimental procedure and simulation method

2.1 Experimental

In order to understand the phenomenon of the flash sintering, the experimental conditions as described in Ref. 2 were simulated by the finite element model. The green sample was bone shape, as shown in **Fig. 1**, the gage section was of 21 mm with a rectangular cross section of 3 × 1.58 mm^2. The employed powders were 3% mol yttria stabilized tetragonal zirconia TZ-3Y[5] (the calcination temperature was 650°C, the crystallite size was 8–12 the mean secondary particle size was 40–70 nm) and non-stabilized undoped zirconia powder TZ-0 (Tosoh Corporation, Japan). The initial green sample was around 50% of relative density. The flash sintering was performed in air furnace under the application of a constant DC voltage as shown in Fig. 1. The sample was suspended in the furnace by means of two electrically insulated platinum wires connected to the handles of the dog bone specimens. The furnace temperature

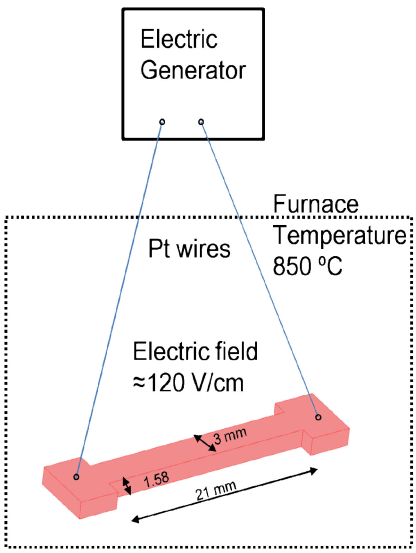

Fig. 1. (Color online) Schematic of bone shaped sample geometry and the experimental set-up of flash sintering process. The preheating furnace temperature was 850°C.

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was raised up to 850°C and consequently the electric field of 120 V/cm was applied to the sample. Below 850°C no electric filed was applied, in fact, the ionic conductivity of TZ-3Y is exponentially dependent on the temperature.

2.2 Simulation method

In order to account the thermal effect during the sintering, we simulated process just before the electric current shut down, when the electric power supplied by the generator rapidly increased.[2] We believed that the sudden increase of power was responsible of the flash sintering phenomenon under the electric field ranging from 60 to 120 V/cm.[2] In particular, as reported by Cologna et al.,[2] in the case of electric field as high as 120 V/cm, the maximum power supplied by the generator was 70 W which corresponded to a current flowing through the sample of about 0.3 A. The application of high voltage (60 to 120 V/cm) generated significant Joule inside the sample, however, was quite difficult to perform a reliable measurement of the sample local temperature due to the high heating rate (1000°C/min) and the small cross section the sintering sample. Thus, the FEM model was indispensable to overcome the intrinsic difficulties of the temperature measurements.

The overall set of governing equations along with initial conditions and boundary constraints were solved using a commercial finite-element package. Maxwell's and the energy balance equation were simultaneously solved with inherent coupling terms.

The accurate prediction of the influence of electric currents on the temperature field is crucial to understand the mechanism of the flash sintering process. The current flow in the TZ-3Y material is governed by the continuity equation, which follows from Maxwell's equations:[6]

$$\nabla \cdot [-\sigma(T, RD)_i \nabla U] = 0 \quad (1)$$

with

$$\vec{E} = -\nabla U \text{ and} \quad (2)$$

$$\vec{J} = \sigma(T, RD)\vec{E} \quad (3)$$

where $\vec{J}$, $\vec{E}$, U, σ and RD are current density vector, electric field vector, electric scalar potential, electrical conductivity, relative density.

Joule heating[6] is referred to as an irreversible phenomenon. It is a dissipative volume heat generation ($\dot{Q}_V$) due to current flow resistance:

$$\int_{V_i} \dot{Q}_V \mathrm{d}V = \int_{V_i} \vec{J} \cdot \vec{E} \mathrm{d}V = \int_{V_i} \sigma(T, RD)_i |\vec{E}|^2 \mathrm{d}V = -\int_{V_i} \sigma(T, RD)_i \, (\nabla U)^2 \mathrm{d}V \quad (4)$$

The heat generation term is coupled to Eq. (1) through the electrical conductivity. The latter depends on temperature and the relative density. The heat generation term is also coupled to heat conduction equation[6] which, in the integral form reads:

$$\int_{V_i} [\rho C_p(T, RD)]_i \frac{\partial T}{\partial t} \mathrm{d}V = \int_{V_i} \nabla \cdot (k(T, RD)_i \nabla T) \mathrm{d}V + \int_{V_i} \dot{Q}_V(T, RD, U)_i \mathrm{d}V \quad (5)$$

where ρC_p, T, t, k, $\dot{Q}_V$, are heat capacity, temperature, time, thermal conductivity generated heat per unit volume V, respectively.

Equations (1)–(5) can be solved simultaneously in the system domain provided that initial conditions, boundary conditions along with materials properties functions of temperature in TZ-3Y sample are defined. Heat capacity, thermal electrical, and thermal conductivity were assumed to be functions of the temperature. The simulation took into account the heat convection and radiation occurring inside the furnace.[6] The temperature dependent thermo-physical properties (i.e., specific heat, electric and thermal conductivity) of the TZ-3Y employed in the FEM simulations are given in references.[7)–9] The simulation did not take into account the dimensional changes of the sample during sintering, however, the experimentally measured voltage and current profiles accounted implicitly the effect of the densification. The prediction of the electric ant the temperature fields was crucial to understand the flash sintering process.

3. Results and discussion

Figure 2 shows the temperature distribution at the mid-thickness plane of TZ-3Y sample after 3 s discharge. The electric power generator was 70 W, the electric field applied to the sample gage section was 120 V/cm and the DC current intensity was 0.3 A. As can be seen in Fig. 2, the application of the high electric power rapidly heated the sample, the temperature raised from 850 to 1600°C in 3 s. The latter can explain the almost instantaneous shrinkage of the TZ-3Y powder.[2] In the flash sintering temperature equilibrium is rapidly reached when the heat losses by radiation and convection are equal to Joule heating source.

The results presented by Cologna et al. did not take into account the significant effect of local temperature increase inside the sample as shown in Fig. 2. During the sample heating under constant electric field, the current density flowing through the TZ-3Y sample is progressively increased.[2] The sudden increase of electric current corresponded to the flash (short time) sintering phenomenon. In fact, it is well known that by increasing the temperature over 850°C the electric conductivity grows exponentially with the temperature,[7),8] consequently the application of an electric field as high as 120 V/cm led to a significant

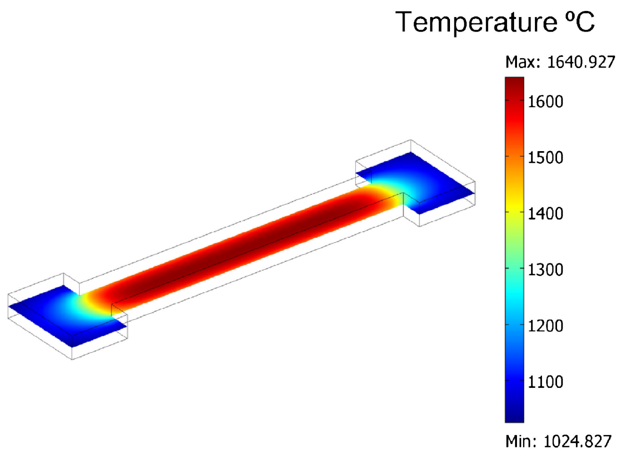

Fig. 2. (Color online) FEM simulation of the TZ-3Y sample temperature distribution assuming the application of 70 W electric power generator for 3 s. The electric field along the sample gage section was 120 V/cm. The furnace-sample temperature before the application of the 120 V/cm electric field was 850°C.

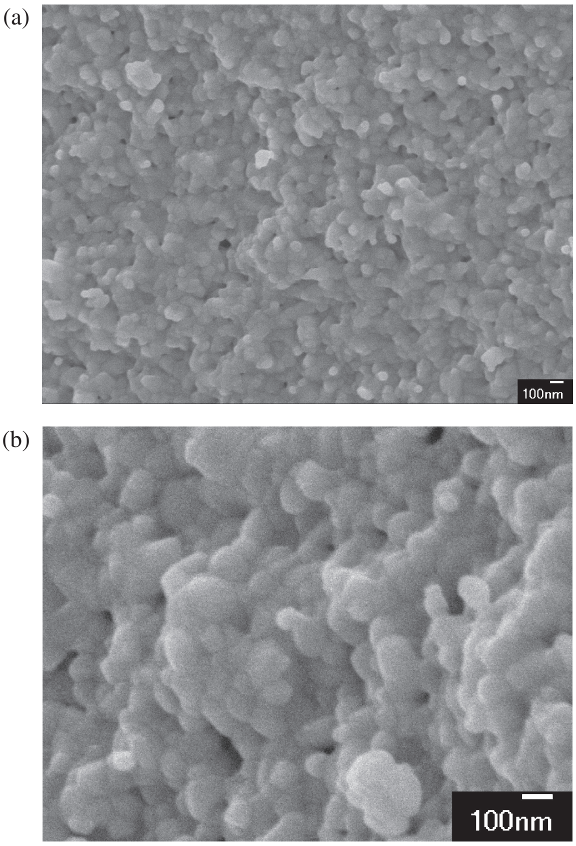

Fig. 3. FESEM photograph of the fracture surface of the TZ-3Y flash sintered sample for 10 s at (a) low and (b) high magnification. The furnace-sample temperature before the application of the 120 V/cm electric field was 850°C. The high electric conductivity of the sample generated sudden increase of the sample temperature (see Fig. 2). The good level of densification was attributed to the local sample temperature.

sample overheating which corresponded to an almost instantaneous sample shrinkage.

Figure 3 shows the microstructure of the flash sintered TZ-3Y sample. The discharge time was about 10 s. The grain size was below 100 nm and the density as measured by SEM micrograph was as high as 98%. For comparison we carried out flash sintering experiments of undoped zirconia TZ-0 powder. In the case of the TZ-0 the sample behaved essentially as an electric insulator and no significant Joule heating occurred under the application of the electric field of 120 V/cm. By comparing the microstructure of TZ-3Y and TZ-0 (see Figs. 3 and **4**), both sintered under the application of an electric field of 120 V/cm at the furnace temperature of 850°C, the higher level of densification in TZ-3Y sample could be attributed to local sample overheating rather than to intrinsic effect of the applied electric field.

4. Conclusion

The flash sintering process was simulated in the case of stabilized and undoped zirconia powders. It is shown that the high level of densification obtained in the case of yttria stabilized

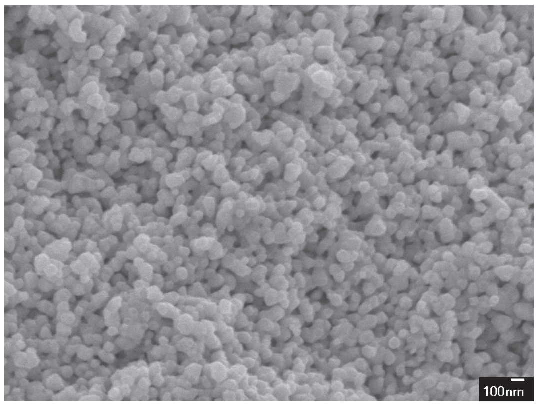

Fig. 4. FESEM photograph of the fracture surface of the flash sintered ZS-0 (without yttria) sample. The furnace temperature before the application of the electric field of the 120 V/cm was 850°C. The ZS-0 was electrically insulating and no Joule overheating occurred. No significant effects of the electric field were observed.

tetragonal zirconia sample was attributed to the local sample overheating. In the case of non stabilized sample the application of the electric field did not affected significantly the powder densification. The flash sintering phenomena was ascribed to the sudden temperature increase inside the sample rather than to the intrinsic effect of the electric field.

The flash sintering process in combination with the appropriate modeling tools might be suitable for sintering material at very high heating rate (i.e., 10^4°C/min) in order to inhibit the grain growth. Furthermore the flash sintering process, because of the highly localized heating and the short processing time, is highly energy saving. The modeling tools are fundamental to limit the thermal stress inside the samples during sintering and cooling. By means of computer simulation tools, the physical principle of the flash sintering has been elucidated in term of bulk Joule heating of the sample.

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