

# Comparison of Creep Behavior in Alumina Based Ceramics Densified by Spark Plasma Sintering and Hot Pressing

E. Karamian<sup>1\*</sup>, A. Bataille<sup>2</sup> and S. Abdollah<sup>3</sup>

<sup>1,3</sup> Department of Materials Engineering, Najafabad Branch, Islamic Azad University, IAUN, P.O.Box 517, Isfahan, Iran

<sup>2</sup> UMET, Unité Matériaux et Transformations, UMR CNRS 8207, University of Lille Science and Technology, F-59655 Villeneuve d'Ascq Cedex, France

## Abstract

Spark plasma sintering (SPS) method, as plasma activated sintering, is a method applicable for rapid sintering of metals and ceramics. Owing to the advantage of rapid heating, the alumina ceramics obtained by SPS have a grain size and density comparable to those of hot pressed ones. The increase of densification rate may be related to some difference in ion transport characteristics. This study describes creep behavior and deformation mechanisms of alumina based materials densified by SPS, and somewhat comparison is made with hot pressing (HP) at high temperature. Pure alumina was densified by SPS at 65 MPa (1200 °C) and 45 MPa (1400 °C) by HP. The grain size of the alumina HPed was twice more than the grain size of the sample SPSeD. Compressive creep tests in air were performed at different true stresses up to 100 MPa and different temperatures (1200 °C-1400 °C). The grain growth was found to be more active during creep of SPS alumina than the creep of HP alumina. Generally, the fineness of SPS materials microstructure speeded up all processes related to diffusion. The ratio of dynamic to static grain growth in SPSeD samples was twice more than HPed samples during creep test.

**Keywords:** Creep, Alumina ceramics, Sintering, Spark plasma sintering (SPS), Hot pressing (HP).

## 1. Introduction

The process of hot deformation of alumina base ceramic is limited by grain growth. Static and dynamic changes in grain size during deformation contribute to the overall growth which usually reduces continuously the super plastic capacity of the considered material. Different approaches have been experimented in order to control the grain growth during deformation. The first approach is using some compounds (such as MgO and ZrO<sub>2</sub>) in order to lower the grain boundary mobility and impede the grain growth of alumina. MgO has been added to alumina in order to lower the grain boundary mobility through solute drag and ZrO<sub>2</sub> particles have been particularly useful in impeding the grain growth of alumina by second phase pinning. However, the MgO effect is limited and one problem arises with the addition of ZrO<sub>2</sub> particles: their effectiveness is related to the homogeneity of their distribution which can evolve during the deformation of the material. Small carbon particles reduce greatly

the grain growth during the sintering and compression experiments. The super-plasticity of the carbon-containing alumina is greatly improved<sup>1)</sup>.

The second approach is using the new technique (such as SPS method) to obtain fine-grained ceramics during sintering process of ceramic materials. Hot-pressing (HP) has been the widely used sintering technique for producing highly dense UHTCs (Ultra-high temperature ceramics)<sup>2)</sup>. Spark plasma sintering (SPS) has recently been considered as an alternative method to obtain dense and fine-grained ceramics at low temperatures. During SPS of the alumina ceramics, the heating rate is very high ( $\geq 150$  °C/min) and the holding time at sintering temperature is short (3–10 min). Owing to the advantage of rapid heating, the alumina ceramics obtained by SPS have a grain size and density comparable to those of HPed ones. For example, a fully dense (a relative density of ~100%) alumina with a grain size of 0.5  $\mu\text{m}$  was obtained at 1200 °C by SPS<sup>3,4)</sup>. Spark plasma sintering (SPS) has emerged as a nonconventional powder consolidation method in densifying a number of poorly sinterable ceramics in very short times<sup>5)</sup>. In this technique, densification is stimulated by the application of a pulsed electric field, combined with resistance heating and pressure. Several experiments have emphasized that high-rate and possibly low-temperature sintering are indeed characteristics of this technique<sup>2)</sup>.

On the other hand, SPS has recently been

\* Corresponding author:

Tel: +98 331 2291111

Fax: +98 331 2291016

Email: ekaramian@mpt.iaun.ac.ir

Address: Department of Materials Engineering, Najafabad Branch, Islamic Azad University, IAUN, P.O.Box 517, Isfahan, Iran

1. Assistant Professor

2. Professor

3. M.Sc. Student

considered as an alternative method to obtain dense and fine-grained ceramics at low temperatures. SPS, also known as plasma activated sintering, is a method applicable for rapid sintering of metals and ceramics. Owing to the advantage of rapid heating, the alumina ceramics obtained by SPS have a grain size and density comparable to those of HPed ones. The increase of densification rate may be related to some difference in ion transport characteristics.

In fact, super-plastic deformation of fine grained alumina is a boundary diffusion process mainly involving grain boundary sliding. Also, cavitations may become an important accompanying process for deformation when the mean grain size is over a few micrometers. From this basis, one of the necessary conditions for dense alumina to exhibit important deformation capabilities is to start with a submicrometre microstructure. The finer initial microstructure will lead to the better plastic behavior <sup>6)</sup>.

The aim of this paper is to assess the creep and deformation behavior of alumina based ceramics densified by SPS and somehow compare it to the behavior of HPed ones.

## 2. Experimental procedure

The starting material used in the study was an atomized commercial 500 ppm FeO-doped  $\alpha$ -alumina powder (Baikowski, SM8), as can be seen Figure 1. The nominal composition of the alumina powder is given in Table 1.

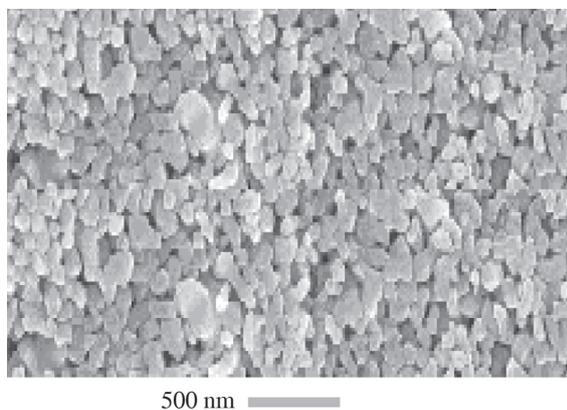


Fig. 1. Micrograph of Pure alumina- $\alpha$  (Baikowski, SM8, Mean average size 300 nm).

From this powder, two types of dense alumina materials were produced. The first type was produced

by HP method, whereas for the second type, SPS method was employed. Discs were hot-pressed (45 MPa) during 30 min at  $T=1450\text{ }^\circ\text{C}$  in a graphite die with the heating rate being  $10\text{ }^\circ\text{C}/\text{min}$ . SPS was performed under 45 MPa at  $1200\text{ }^\circ\text{C}$ . The obtained alumina discs had a diameter of 30 mm and a height of 10 mm.

The density of the resulting billets was determined by weighing in alcohol using the Archimedes' method. Final densities were found to be greater than 99.4% of theoretical for all billets, and 95.6% of theoretical for samples FeO-doped. Samples for deformation experiments were cut from the pressed billets and the faces were made parallel. Typical sample sizes had a transverse square section of  $3\times 3\text{ mm}^2$  and a height of 7 mm.

Compressive creep tests in air were carried out at different true stresses up to 100 MPa and different temperatures ( $1200\text{--}1400\text{ }^\circ\text{C}$ ). From length change versus time curves, true strain rates versus true strain curves were obtained. The microstructures of densified and strained specimens were characterized by scanning electron microscopy (SEM). For SEM investigations, diamond polished cross sections were thermally etched at  $T=1270\text{ }^\circ\text{C}$  for 30 min to reveal the microstructure (Figure 2). The grain sizes were obtained by multiplying the square root of the average grain surface by 1.38 <sup>7)</sup>. More than three hundred grains were numbered for each grain size calculation.

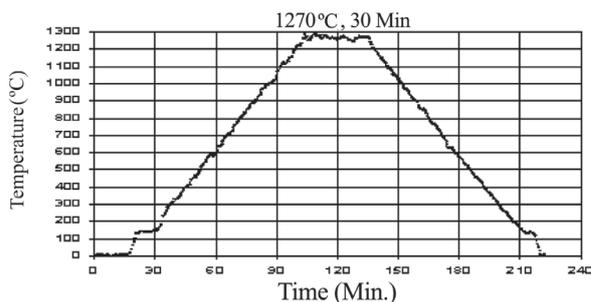


Fig. 2. Graph of thermal etch for SEM investigations.

## 3. Results

### 3.1. Creep tests ( $\dot{\epsilon}$ - $\epsilon$ curves)

Compression creep tests were carried out in air at a temperature range ( $1200\text{--}1400\text{ }^\circ\text{C}$ ) and under true stresses up to 100 MPa using length change versus time curves, and true strain rates versus true strain curves. The graphs of true strain rates versus true strain ( $\dot{\epsilon}$ -  $\epsilon$ ) at the temperature of  $1200\text{ }^\circ\text{C}$  and  $1300\text{ }^\circ\text{C}$  under stress true rate of 30 MPa for samples of alumina densified by SPS at  $1300\text{ }^\circ\text{C}$  and  $1400\text{ }^\circ\text{C}$  and under the same

Table 1. Chemical and physical properties of Alumina- $\alpha$ +500 wt. ppm FeO before atomization

Element	Fe	Na	K	Si	Average particle size (nm)	Mean grain size, Crystalline size (nm)	BET $\text{gr}/\text{m}^2$
ppm	500	17	38	8	400	50	10

true stress for samples densified by HP are presented (Figures 3 and 4).

As a matter of fact, the graph corresponding to the creep SPS alumina at 1300 °C and 30 MPa is very comparable to the graph of creep HP alumina at 1400°C and the same stress level.

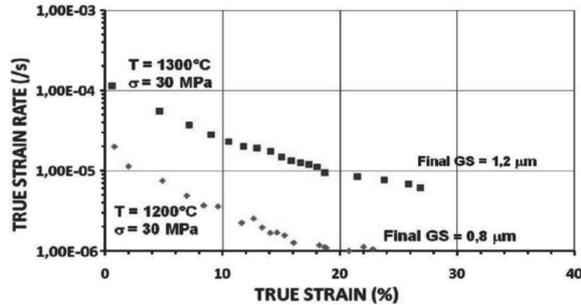


Fig. 3. True strain rate versus true strain curves ( $\dot{\epsilon}$ -  $\epsilon$ ) at the temperatures of 1200 °C and 1300 °C,  $\sigma$  =30 MPa for SPS alumina.

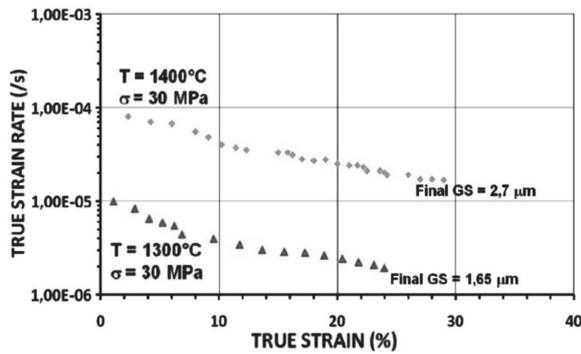


Fig. 4. True strain rate versus true strain curves ( $\dot{\epsilon}$ -  $\epsilon$ ) at the temperatures of 1300 °C and 1400 °C,  $\sigma$  =30 MPa for HP alumina.

Concerning the calculation grain size of deformed samples observed by SEM at different temperatures,  $\sigma$  =30 Mpa, and the final deformation ~%25, the graph of grain growth was obtained in both types of samples (Figure 5). The grain growth was twice more active during creep of SPS alumina than creep HP alumina.

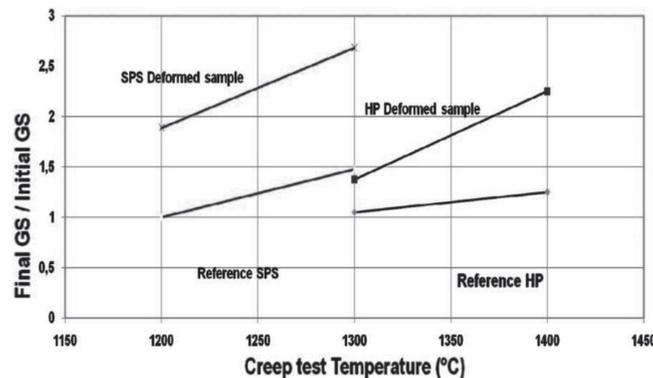


Fig. 5. The static and dynamic grain growth, reference and deformed samples, respectively at different creep temperatures,  $\sigma$  =30 MPa and final deformation ~%25 for both SPS alumina and HP alumina.

### 3.2. Microstructures of densified and deformed materials (SEM micrographs)

Figures 6 and 7 show SEM micrographs of both SPS and HP alumina as densified and deformed at  $T$ =1300 °C,  $\sigma$  =30 MPa and final deformation of %30.

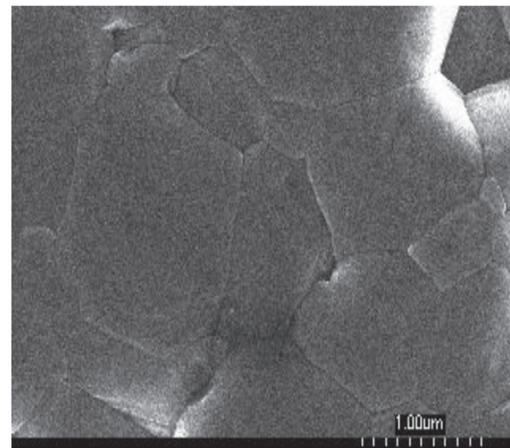
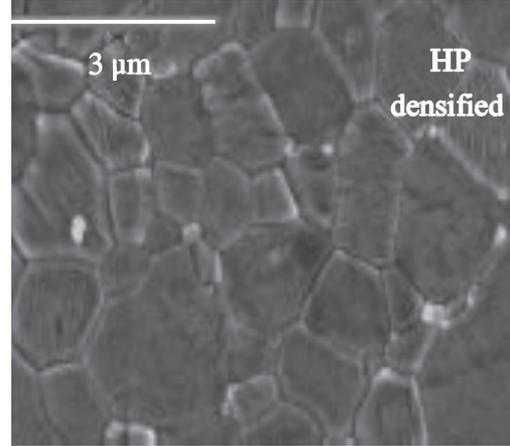


Fig. 6. SEM micrographs of HP alumina samples as densified (left) and deformed at  $T$ =1300 °C,  $\sigma$  =30 MPa and final deformation %30 (right). Grain size was increased from 1000 nm to 1220 nm for sample deformed at these conditions.

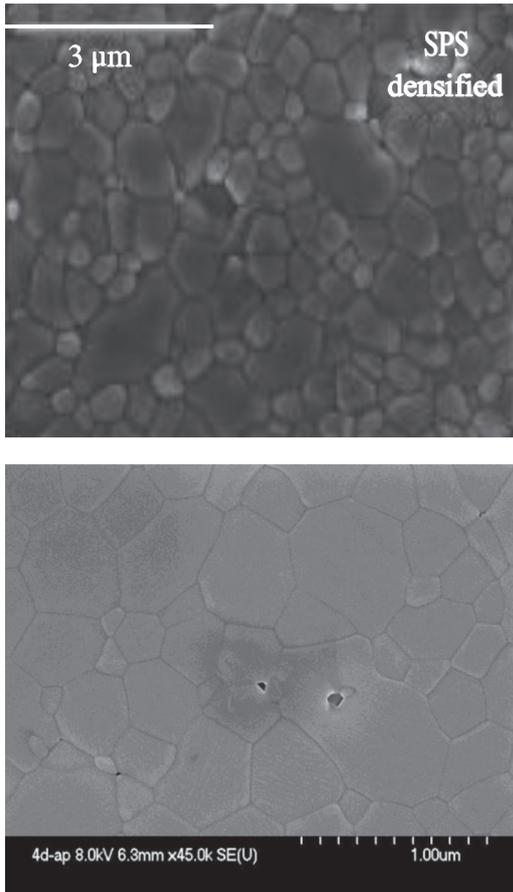


Fig. 7. SEM micrographs of SPS alumina samples as densified (left) and deformed at  $T=1300\text{ }^{\circ}\text{C}$ ,  $\sigma =30\text{ MPa}$  and final deformation %30 (right). Grain size was increased from 440 nm to 750 nm for sample deformed at those conditions.

#### 4. Discussion

Creep tests were mainly carried out at  $S=30\text{ MPa}$ , and  $S=75\text{ MPa}$ . The presented graphs corresponded to the creep tests at  $S=30\text{ MPa}$  and different temperatures. The creep behaviour of SPS densified alumina under these conditions can be compared to one of hot-pressed alumina. The similarity of creep behaviour is perceptible. As a matter of fact, the curve corresponding to the creep of SPS alumina at  $S=30\text{ MPa}$  and  $T=1300\text{ }^{\circ}\text{C}$  is very comparable to one of HP alumina at  $S=30\text{ MPa}$  and  $T=1400\text{ }^{\circ}\text{C}$ .

By considering the expression of the constitutive equation of superplastic deformation, mean values of stress exponent, strain size dependant factor and activation were obtained for the SPS alumina and those comparable to the ones for HP alumina (reference 1 and present work). As follows, the calculated values were close for both materials:

$$\dot{\epsilon}(\text{SPS}) = C_1 \cdot \sigma^{2.2} \cdot \exp\left(-\frac{490\text{ kJ}}{RT}\right) \quad (1)$$

$$\dot{\epsilon}(\text{HP}) = C_2 \cdot \sigma^{1.92} \cdot d^{-2.2} \cdot \exp\left(-\frac{510\text{ kJ}}{RT}\right) \quad (2)$$

Interestingly, when the second equation was

considered, a decrease of temperature of  $100\text{ }^{\circ}\text{C}$  (from  $1400\text{ }^{\circ}\text{C}$  down to  $1300\text{ }^{\circ}\text{C}$ ) was compensated by the difference of grain size observed between SPS alumina and HP alumina. This would explain the presented similarity of creep curve. This means that creep mechanisms active in the creep of HP alumina are those active in the creep of SPS alumina.

From the analysis of grain growth at  $S=30\text{ MPa}$  and at different temperatures, the grain growth was found to be more active during creep of SPS alumina than the creep of HP alumina. This may be the counterpart of the high densification rate obtained at lower temperatures in the SPS process. The high rate limits the possibility of reaching the equilibrium in the microstructure during SPS densification. Hence the microstructure of SPS material is less stable. Also, there are several cavitations in triple grain boundary, decreasing the mobility of grain boundary or grain growth. So, the grain size of HPed samples is more stable during creep test than SPSed samples.

#### 5. Conclusion

1. The grain growth was more active during the sintering of SPS alumina than the sintering of HP alumina.
2. By considering the analysis of grain growth at  $S=30\text{ MPa}$  and at different temperatures, the grain growth was more active during the creep of SPS alumina than the creep of HP alumina.
3. The high rate limited the possibility of reaching the equilibrium in the microstructure during SPS densification. Hence the microstructure of SPS material was less stable.
4. The grain size of the HP alumina was twice bigger than the grain size of SPS sample ( $1000\text{ nm}$  versus  $440\text{ nm}$ ). Generally, the fineness of SPS materials microstructure speeded up all processes related to diffusion.
5. By considering the investigation of microstructure of HPed samples, there were several cavitations in triple grain boundary, decreasing the mobility of grain boundary or grain growth. So, the grain size of HPed samples was more stable during creep test than SPSed samples.
6. The ratio of dynamic to static grain growth in SPSed samples was twice more than HPed samples during creep test.

#### References

- [1] A. Bataille, J. Crampon and R. Duclos: *Ceram. Int.*, 25(1999), 215.
- [2] F. Monteverde: *J. Alloys Compd.*, 428(2007), 197.
- [3] Z. Shen, M. Johnsson, Z. Zhao and M. Nygren: *J. Am. Ceram. Soc.*, 85(2002), 1921.
- [4] R.S. Dohedoe, G.D. West and M.H. Lewis: *Bull. Eu. Ceram. Soc.*, 1(2003), 19.
- [5] M. Nygren and Z. Shen: *Solid State Sci.*, 5(2003), 125.
- [6] A. Bataille and J. Crampon: *J. Mater. Sci.*, 38(2003), 3245.
- [7] C. Carry and A. Mocellin: *Ceram. Int.*, 13(1987), 89.