ERRATUM

Erratum: Corrigendum to "Phenomenological analysis of densification mechanism during spark plasma sintering of MgAl₂O₄" [J. Mater. Res. 24, 2011 (2009)]

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This recently published paper reported how spark plasma sintering (SPS) of a stoichiometric alumina–magnesia spinel (MgAl₂O₄) powder, shaped by slip casting, was studied in vacuum, in the 1200–1300 °C temperature range. The other experimental parameters were a heating rate of 100 °C/min, an applied macroscopic compaction pressure of 25 MPa, a soak time of 15 min, and the use of the standard 12:2 pulse configuration.

In the discussion section, we tried to determine what could be the mechanism(s) involved during the SPS experiments we completed and to correlate the suspected mechanism(s) to the microstructure of the sintered MgAl₂O₄ samples.

Assuming an approach similar to Mukherjee for the creep of dense metals, it has been proposed that the SPS kinetic equation can be written as:

$$\frac{1}{\mu_{eff}} \frac{1}{D} \frac{dD}{dt} = K \frac{e^{-\frac{Q_d}{RT}}}{T} \left(\frac{b}{G} \right)^p \left(\frac{\sigma_{eff}}{\mu_{eff}} \right)^n \quad , \qquad (1)$$

where D is the instantaneous relative density of the compact, t is the time, μ_{eff} is the instantaneous shear modulus of the compact, K is a constant, R is the gas constant, T is the absolute temperature, Q_d is the apparent activation energy of the mechanism controlling densification, b is the Burgers vector (close to the lattice parameter), G is the grain size, and σ_{eff} is the instantaneous effective stress acting on the compact.

It was also proposed that μ_{eff} and σ_{eff} can be written as:

$$\mu_{eff} = \frac{E_{th}}{2(1 + \nu_{eff})} \frac{D - D_0}{1 - D_0} \quad , \qquad (2)$$

$$\sigma_{eff} = \frac{1 - D_0}{D^2(D - D_0)} \sigma_{mac} \quad , \qquad (3)$$

where E_{th} is the Young's modulus of the theoretically dense MgAl₂O₄ material, ν_{eff} is the effective Poisson's ratio, D_0 is the starting green density of the powder compact, and σ_{mac} is the macroscopic compaction pressure.

To discriminate the mechanisms controlling the densification of the spinel powder during SPS, it is necessary to determine the values of the Q_d, p, and n parameters in Eq. (1).

Rearranging Eq. (1) yields:

$$Ln \left[\frac{T}{\mu_{eff}} \frac{1}{D} \frac{dD}{dT} \frac{dT}{dt} \right] = -\frac{Q_d}{RT} - pLn(G) + nLn\left(\frac{\sigma_{eff}}{\mu_{eff}} \right) + K' \quad , \qquad (4)$$

where $\frac{dT}{dt}$ is the heating rate during the SPS experiment, K' is a constant, and G is given by:

$$G(nm) = \alpha e^{\beta D} \qquad (5)$$

with $\alpha = 0.0018$ *and* $\beta = 5.3586$.

The heating part of a SPS run, for temperatures between 940 and 1300 °C, was exploited to determine what could be the values of Q, p, n, and K' that are the key parameters of Eq. (4). Indeed, in the function of the obtained values, the hypothesis about the involved densification mechanism can be formulated. Unfortunately, a mistake was presented in the calculations that were originally published. Therefore, the purpose of this corrigendum is to report the correct results and the possible
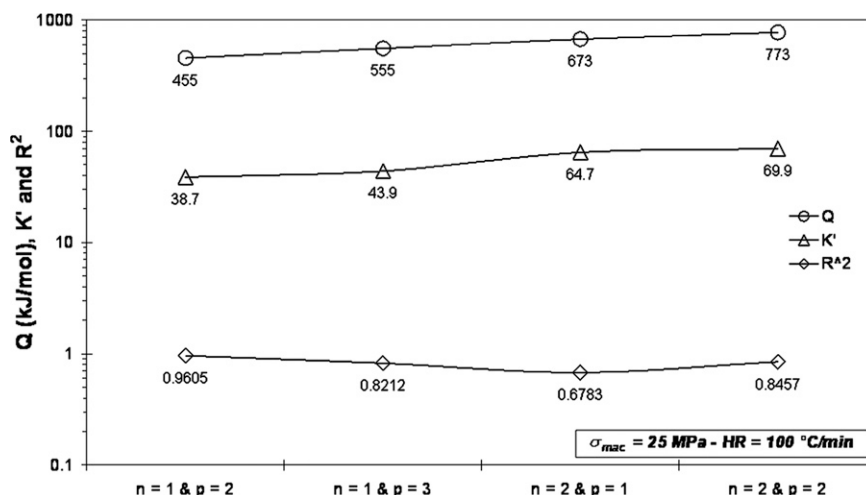
FIG. 1. Q_d, K', and R^2 values obtained for the different couples (n, p) imposed are shown. R^2 is the highest when n and p have values of 1 and 2, respectively.

impact of this mistake on other calculations/interpretations published in the original paper.

Let us exploit correctly the heating part of an SPS run for temperatures between 940 and 1300 °C. Imposing n and p values (as reported in the published paper, these parameters have precise values that determine what is the densification mechanism that could be claimed), it is possible, using the Excel Solver function, to calculate the corresponding Q_d and K' values that enable the left side of Eq. (4) to be equal to its right side. Then, plotting $Ln\left[\frac{T}{\mu_{eff}}\frac{1}{D}\frac{dD}{dT}\frac{dT}{dt}\right]$ as a function of $-\frac{Q_d}{RT} - pLn(G) + nLn\left(\frac{\sigma_{eff}}{\mu_{eff}}\right) + K'$ should provide a straight line passing through the origin (we forced the passage through the origin during the calculation), whose regression coefficient R^2 can be obtained.

Q_d, K', and R^2 values obtained for the different couples (n, p) imposed are given in Fig. 1. R^2 is highest when n and p have a value of 1 and 2, respectively. In that case, Q_d has a value of 455 ± 20 kJ/mol and K' is $\sim 38.7 \pm 1.8$. In agreement with theoretical values for n and p and also because the apparent activation energy is close to the one for oxygen self-diffusion in monocrystalline stoichiometric spinel (see the published paper for precisions), we propose that grain boundary sliding, accommodated by lattice diffusion of the O^{2-} anions, governs densification of our spinel samples during the heating portion of the SPS experiments we performed (between 940 and 1300 °C).

The densification mechanism invoked in this corrigendum is indeed close to the one claimed in the published paper. The only difference concerns the quality of sources and sinks of vacancies involved in the diffusion process. Originally erroneous calculations told us that sources and sinks were not perfect. Now, with correct calculations, sources and sinks should be perfect, minimizing strongly the possible contribution of an interface-reaction step in the densification mechanism, at least during the heating portion of an SPS run.

One may now wonder about an interface-reaction contribution to the densification mechanism at the beginning of soaks at the different sintering temperatures, as proposed in the published paper. At this point, we decided to stick with the scenario established in the published paper because it fits perfectly with the microstructure of the sintered samples observed by transmission electron microscopy. Another point comforts also our analysis. Additional calculations have been completed. For that, p has been fixed to 2 (correct value obtained from the analysis of the heating portion of an SPS run, see above). Then, an average value of $n = 1.6$ and an apparent activation energy of 570 ± 25 kJ/mol for sintering temperatures in the range of 1200–1275 °C are obtained. Because in that case, n is not close to 1 and clearly between 1 and 2, it confirms the contribution of an interface-reaction step to the densification mechanism, as originally proposed in the published article.

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J. Mater. Res., Vol. 24, No. 11, Nov 2009