Pressure Effects on Temperature Distribution during Spark Plasma Sintering with Graphite Sample

Salvatore Grasso¹₂³, Yoshio Sakka¹₂³,* and Giovanni Maizza⁴

¹World Premier International Research Center Initiative (WPI Initiative) on Materials Nanoarchitronics (MANA), NIMS, Tsukuba 305-0047, Japan
²Graduate School of Pure and Applied Sciences, University of Tsukuba, Tsukuba 305-0047, Japan
³Nano Ceramics Center, Fine Particle Processing Group, National Institute for Materials Science (NIMS), Tsukuba 305-0047, Japan
⁴Dipartimento di Scienza dei Materiali ed Ingegneria Chimica, Politecnico di Torino, Corso Duca degli Abruzzi 24, I-10129 Torino, Italy

The influence of applied pressures on temperature distribution in punch/die/graphite/sample assembly during SPS current control mode operation was systematically investigated by coupling experiments and computer modeling. Combined experimental and numerical results showed that the peak temperature and the temperature difference existing between the sample and the die outer surface progressively decreased with increasing of applied pressure from 5 to 80 MPa. This behavior was attributed to the strong change of the electric and thermal contact resistances at the punch/die interface due to punch Poisson deformation. [doi:10.2320/matertrans.M2009148]

(Received April 23, 2009; Accepted May 25, 2009; Published July 8, 2009)

Keywords: spark plasma sintering, finite element, multiphysics modeling, contact pressure, graphite compact sample

1. Introduction

Spark Plasma Sintering (SPS) is an electric current assisted sintering technique characterized by an intricate coupled bulk and contact multiphysics.¹⁻⁵ The electric, thermal and stress/strain fields are intimately coupled not only within the volume of the SPS machine constituent parts (i.e. graphite punches, die and spacers) but also along their contact interfaces. The material properties (i.e. electrical resistivity, thermal conductivity, density and specific heat) together with the Joule heat source term work as strong coupling factors between involved physical fields. Most published SPS models¹⁻⁵ assumed bulk materials properties as at least non linear functions of temperature.

Conversely to bulk multiphysics, our understanding on contact multiphysics is incomplete both theoretically and experimentally and only a limited number of papers dealt with the full coupling of electric, thermal and displacement multiphysics at the contact interfaces. Nevertheless, contact problems have been for long time subject of theoretical and practical interest in many engineering applications such as spot welding,⁶ furnace electrodes,⁷ carbon brushes, electrical joints and switches.⁸

Imperfect contacts caused by surface roughness, texture, impurities, insulating layers or oxides,⁹⁻¹³ may involve complex behavior of thermal and electrical resistances at the interfaces which can be very difficult to predict. Basic contact theories⁹⁻¹³ suggest that contact resistance is function of the actual contact surface area, electrical resistivity and interface temperature. The actual contact surface area depends on the contact pressure and usually is only a small fraction of the apparent contact area.

In a SPS apparatus, the main contact interfaces are located between the graphite elements and between the punches and the powder sample. Graphite performs quite well as an electric contact material since it exhibits good electric conductivity, high temperature oxidation resistance, good thermal/mechanical stability at high temperature¹² and high melting point.¹³ However, graphite has a relatively low bearing capacity. Typically, the maximum loading pressure admitted to SPS (punch and die) graphite is in the range 80–140 MPa.

Yet, the effects of external pressure have not been systematically investigated in terms of contact pressure or contact resistances between graphite elements (i.e. punch, spacers and die) and between them and the sample. Indeed, external pressure strongly affects current distribution including both bulk and interface heat generation and heat flow across the contact interfaces.

Experimental and modeling studies reported that SPS horizontal contact resistances are smaller than vertical ones.²⁻⁴,¹⁴ The latter were found to be the main cause of temperature difference developed between the sample core and the outer die surface.¹,⁵ Anselmi-Tamburini et al.¹⁵ studied experimentally the influence of applied pressure on the electric resistance of punch/die/sample assembly in the case of both copper and alumina compact samples. They reported a lower overall electric resistance (measured at room temperature) for copper than for alumina sample up to 50 MPa but surprisingly the overall electric resistance was the same for both samples at higher pressures. The reason for such behavior was not studied in detail. The present authors attributed this behaviour to the change of the punch/die fit tightness as a result of applied pressure.

In the present study, special attention was given to the correlation between applied pressure and temperature differences in the punch/die/sample assembly along the radius. To this purpose, the (SPS) heating process of graphite compact sample was investigated. The designed methodology combined on-line experiments with computer modeling.³,⁴ In the foregoing “pressure” rather than “applied uniaxial pressure” is selected for short, unless specified otherwise.
2. Experimental Procedure

The experiments were carried out on a SPS-1050 machine equipped with a 100 kN uniaxial press (Syntex Inc., Japan). The die, punch and sample were made of low strength molded graphite GS-203 (Toyo Tanso Co. Ltd., Japan). The outer/inner diameters and height of the die were 40/20 and 30 mm respectively. The diameter and height of punches were 19.95 and 20 mm respectively. The diameter and nominal height of graphite sample disk was 20 and 8 mm, respectively.

All experiments were carried out under current-control mode (CCm). CCm offered a direct way to physically model actual SPS cause-effect phenomena using on-line measurable quantities (imposed current, pressure, voltage-time and temperature-time). This unique feature was especially convenient during experimental calibration and validation of the finite element SPS model since it allowed to systematically compare experimental and modeling results. This step was crucial for assessing the reliability of the model.

The room temperature mechanical properties of employed graphite (for punches, sample, die, spacers) were 50 MPa flexural strength, 26 MPa tensile strength, 60 shore hardness and 9.2 GPa elastic modulus.

A constant imposed DC current of 1400 A was assumed in each experiment. The applied pressure was taken as a working parameter ranging from 5 to 80 MPa. Voltage drop was recorded from the control panel of SPS machine. The temperature at the die outer surface (A point, in Fig. 2) was measured by using an optical pyrometer. Moreover, the temperature at the sample edge (B point, in Fig. 2) was measured through a radial bore hole (across the die) using the same optical pyrometer. Accordingly, one single experiment had to be repeated twice. While the first reading provided the desired temperature measurement, the subsequent reading was used to assess measurement repeatability. The second reading was cross-linked with four repeated voltage drop measurements to definitely confirm temperature measurement repeatability.

The relative error of temperature measurements was below 0.3%, which was within 1% precision of the used pyrometer. To assure a reliable temperature measurement the bore hole spot area (7 mm diameter) was designed to be five times larger than pyrometer spot area (3 mm diameter). The two-point temperature measurement offered an inexpensive but effective means to directly measure the temperature difference between the sample core (or edge) and the die outer surface. SPS experiments were stopped when the die surface temperature reaches approximately 1650°C. Radiation heat losses were minimized by applying a 2.5 cm thick graphite felt around the outer die surface. The Poisson induced deformations of punches. The induced deformations in graphite were separately measured by using a compression test apparatus equipped with an optical displacement measuring device.

A fully coupled bulk/contact electrothermal SPS model was developed to elucidate the influence of pressure on temperature distribution in the punch/die/sample assembly for 5, 20 and 80 MPa pressure (SPS) heating. The predicted contact resistances were systematically calibrated in terms of a) voltage drop between the two water cooled rams, b) temperature profiles at both the die surface and the sample edge.

3. Results and Discussion

SPS punches subjected to a uniaxial pressure undergo an elastic deformation composed of a contraction along the longitudinal axis and an expansion along the radial direction. The latter, of specific interest here, is proportional to well known Poisson ratio. This is defined as the ratio of the transverse strain (normal to the applied load) to axial strain (in the direction of the applied load). As an initial clearance fit of 0.05 mm is normally designed between the punch and the die, the application of a uniaxial pressure to punches results in a reduced punch/die clearance. The effects of clearance decrease on electric and thermal contact resistances as a function of a uniaxial press force was not previously investigated neither theoretically nor experimentally.

Figure 1 qualitatively sketches how a vertical punch/die contact was affected by different imposed pressures under the underlined experimental conditions. A loose punch/die fit (Fig. 1(a)) was likely to occur for low applied pressures regimes (i.e. 5, 20 to 40 MPa). Conversely, a tighten fit or an interference fit (Fig. 1(b)) was likely to occur for 60 MPa or 80 MPa pressures respectively. The punch/die clearance fit was defined as the initial clearance minus the total radial punch displacement \( \delta_p \) (Table 1). Two punch/die clearance fits, \( f_1 \) and \( f_2 \), for two different pressures are shown in Fig. 1(a) and (b) respectively. As shown in Table 1, for

<table>
<thead>
<tr>
<th>Applied pressure (MPa)</th>
<th>( \delta_p ) (mm)</th>
<th>Punch/die clearance fit (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>0.007</td>
<td>0.043</td>
</tr>
<tr>
<td>40</td>
<td>0.017</td>
<td>0.033</td>
</tr>
<tr>
<td>60</td>
<td>0.032</td>
<td>0.028</td>
</tr>
<tr>
<td>80</td>
<td>0.053</td>
<td>Interference</td>
</tr>
</tbody>
</table>
pressures lower than 80 MPa no interference fit at punch/die interface was observed. The 5 MPa case was not included in Table 1 since the inherent measured displacement was negligible.

Zavaliangos et al.\textsuperscript{1)} and Vanmeensel et al.\textsuperscript{2)} observed that the temperature difference between the die surface and the sample center was strongly influenced by punch/die (vertical) contact resistances. Figure 2 shows the temperature difference between A and B points ($\Delta T_{AB} = T_B - T_A$), versus either time (Fig. 2(a)) or die outer surface temperature (Fig. 2(b)). It is observed that $\Delta T_{AB}$ decreased with increasing pressure. Except for 5 MPa case, the temperature difference $\Delta T_{AB}$ progressively increased either as a function of time or as a function of die surface temperature. Moreover, the heating rate at both A and B points progressively decreased by increasing pressure.

For low and intermediate applied pressures regimes (i.e. 5, 20 and 40 MPa) a loose punch/die fit occurred. The 5 MPa curve in Fig. 2 represented a limiting case as shown by the pronounced temperature difference originated in the early transient. This was attributed to severe bulk and interface Joule heating. After this transient, $\Delta T_{AB}$ decreased. Assuming the temperature distribution at 180 s as a reference, the temperature difference for low and moderate pressures was as high as 300°C (Fig. 2(a)).

A rather different situation was expected for higher pressures, such as 60 and 80 MPa, for which a tighten fit or even an interference fit (Fig 1(b)) was observed along punch/die contact surfaces (Table 1). In case of 60 and 80 MPa, $\Delta T_{AB}$ at 180 s was 210 and 160°C respectively (Fig. 2(a)). These results were consistent with previous experimental studies\textsuperscript{3)} which investigated the effect of various initial clearances (i.e. in the range 0.02, 0.2 mm) at constant pressure. The results in Ref. 5) showed that the temperature difference between the sample edge and the die surface increased with increasing punch/die initial clearance.

Figure 3 shows the time variation of the predicted temperature difference existing between the core and the edge of the sample for 5, 20 and 80 MPa applied pressures. This temperature difference was defined as the temperature at the sample core ($T_{r=0}$) minus the sample edge ($T_{r=10}$). The computed temperatures at the sample core ($T_{r=0}$) after 180 s heating time for the three applied pressures are also marked in.

Fig. 2 Temperature difference between sample edge (B) and die outer surface (A) points as a function of time (a) and die outer surface temperature (b).

Fig. 3 Computed temperature difference across the graphite sample versus heating time for 5, 20 and 80 MPa applied pressures. This temperature difference was defined as the temperature at the sample core ($T_{r=0}$) minus the sample edge ($T_{r=10}$). The computed temperatures at the sample core ($T_{r=0}$) after 180 s heating time for the three applied pressures are also marked in.
The sample core peak temperature was 2103 and 1620°C for 5 and 80 MPa respectively, after 180 s (Fig. 3) while the temperature difference across the graphite sample was 130 and 60°C for 5 and 80 MPa respectively. Small temperature differences in the sample are required to assure uniform densification, homogeneous microstructure and minimal residual stresses after sintering.2,3)

The temperature difference and the maximum peak temperature of the SPS system progressively decreased with increasing pressure. Higher pressures make the punch/die contact mechanically tighter hence, more conductive. Typically in SPS the sample volume is a small portion (i.e. about 5%) of the whole punch/die/sample assembly. As a result the external pressure strongly controls the overall temperature distribution in the punch/dies/sample assembly through the tightness condition of the punch/die contact fit.

4. Conclusion

A fundamental study was undertaken to elucidate the role of an imposed uniaxial pressure on SPS contact resistances during SPS heating in current control mode (SPS-CCm) with graphite compact sample.

The results showed that different values of applied pressure induced significant changes in bulk electric and thermal fields, including Joule heating. The vertical punch/die contact resistance directly affected the temperature difference along the radius in the punch/die/sample assembly. The temperature difference across the punch/die/sample assembly decreased with increasing pressure. The experimental and modeling results confirmed the leading role of external pressure as a fundamental parameter to control SPS performance and material microstructure. The external pressure strongly controls the overall temperature distribution in the punch/dies/sample assembly through the tightness condition of the punch/die contact fit.

Acknowledgment

This work was supported by World Premier International Research Center Initiative (WPI Initiative), MEXT, Japan. The authors are grateful to Toyo Tanso Co. Ltd. (Tokyo) for kindly providing mechanical properties datasheet of GS-203 graphite used in this work.

REFERENCES

16) Toyo Tanso Co. Ltd., Tokyo, private communication.