Synthesis and Processing by the Spark Plasma Method
SIMULATION OF CONTACT RESISTANCES INFLUENCE ON TEMPERATURE DISTRIBUTION DURING SPS EXPERIMENTS

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ABSTRACT

The behavior of the Spark Plasma Sintering/Synthesis (SPS) apparatus, which represents an effective tool for sintering/synthesizing advanced materials, is simulated in this work. A step-by-step heuristic procedure is proposed since several, concomitant physico-chemical phenomena, for example heat transfer and generation, electric current transport, and stress-strain mechanics along with chemical transformation and sintering, take place during SPS processes. In this work we consider the SPS behavior of specific sample configurations characterized by the absence of powders. This approach permits to determine the electric and thermal resistances experimentally evidenced in the horizontal contacts between stainless steel electrodes and graphite spacers as functions of temperature and applied mechanical load. Horizontal contact resistances between graphite elements are experimentally found to be negligible and, accordingly, they are not modeled. Model reliability is tested by comparing numerical simulations with experimental data obtained at operating conditions far from those adopted during fitting procedure of unknown parameters. The proposed model can be successfully compared from a quantitative point of view to the measured temperature, voltage once rms current, geometry are taken into account.

INTRODUCTION

SPS is an effective process for the sintering/synthesis of advanced materials like ceramics, metals, polymers and semiconductors. Basically, it consists in heating up the powder sample shaped into a die inserted between two water cooled electrodes (tanks) by means of a pulsed electric DC forced to pass through, while uniaxially pressuring the system in order to facilitate sintering processes and guarantee electric circuit closure.

In the technical literature, SPS is considered a thermo efficient sintering process since highly dense products in relatively short times are attainable. 4 A volumetric heating rate due to joule effect, in contrast to the conductive heat transport applied in conventional sintering systems, permits a quick temperature rise able to enhance the mass transport mechanisms responsible for sintering phenomena, thus improving consolidation rate and minimizing grain coarsening. The latter aspect leads to improved mechanical, physical and optical properties of final sintered products. 5

While an updated review of modeling approaches adopted to simulate the behavior of SPS apparatus is reported elsewhere 1, a reliable mathematical model of SPS can be obtained in our view by separately analyzing an increasing complex system behavior in the framework of a step by step procedure, where physico-chemical phenomena, previously excluded, are gradually introduced along with their unknown model parameters. This approach allows one to independently fit the complete set of unknown parameters of the comprehensive SPS model, thus avoiding the masking effect given by the various phenomena involved in the whole process. In this work, the first step of this ideal approach is carried out by taking into account heat transfer phenomena, and current distribution. In particular,
the evaluation of the predominant electric and thermal contact resistances is carried out by comparing model results with experimental data obtained when appropriate samples characterized by the absence of powders are used. Specifically, explicit dependence of horizontal electric and thermal contact resistances on applied load and local temperature is obtained.

EXPERIMENTAL SECTION

A SPS apparatus 515S model (Sumitomo Heavy Industries Ltd., Japan) is used for the experimental runs. The power supply is a DC pulse generator which is reported to provide a maximum current and voltage equal to 1500 A and 10 V, respectively, while the mechanical load applied through an hydraulic system can be varied between 3 and 50 kN. Specifically, current pulses of 3.3 ms fixed duration are generated. Operator is free to select the pulse sequence, i.e. number of ON pulses (from 1 to 99) vs. number of OFF pulses (from 1 to 9) that represent periods of time with zero current. Typically, a 12/2 sequence is adopted (as prescribed by Sumitomo). This choice corresponds to the repetition of a sequence of 12 ON pulses followed by 2 OFF pulses for a total sequence period of 46.2 ms (i.e. 3.3 x 14 ms). It should be mentioned that no specifications are available regarding the measured current and voltage, i.e. average or rms values. Referring to Figure 1, the sample is inserted into a die placed between two plungers that are not in direct contact with the stainless steel rams, but spacers are typically inserted in between. From the electric point of view, the end parts of the rams are connected to an electric generator through copper bars and wires. Spacers, plungers and die are made of AT101-grade graphite (ATAL, Italy) which guarantees relatively high electric and thermal conductivities, i.e. lower power dissipation, higher heat transfer to powder specimen, and quicker cooling step. The use of graphite limits the attainable pressure level to a value less than 100 MPa, while the vacuum chamber permits to avoid chemical oxidation of graphitic elements. As it may be seen in Figure 1, the vacuum chamber is made of two coaxial cylinders both jacketed with cooling water circulation. A vacuum level down to 10 Pa is attainable with the SPS 515S model. Rams are made of stainless steel (AISI 304) and cooling water flows through them, as depicted in Figure 1, where the corresponding horizontal section a-a of the water circuit is also shown.

![Diagram of SPS experimental set-up](image)

Figure 1: Schematic representation of SPS experimental set-up (not in scale).
A new data acquisition system has been designed and installed for independently measuring instantaneous (pulsed) values of electric current and voltage, from which calculating average and rms values. In particular, referring to Figure 1, an open loop Hall effect current transducer has been used (LEM HAX 2500-S, nominal primary current 2500 A, rms. maximum primary current 5500 A, bandwidth 25 kHz, accuracy 1 % at nominal current) along with a voltage isolation amplifier (DATAFORTH DC4A1-09, Input range -40 to +40 V, bandwidth 3 kHz, accuracy 0.03 % of full scale). The latter one is connected to the copper bars right close to the stainless steel electrodes. The output signals of these transducers are fed to a data acquisition board (200 KS/s, 12-Bit, 16 Analog Input Multifunction, National Instruments) connected to a PC, where a specifically designed Labview (National Instruments) virtual instrument is installed. This data acquisition system is able to collect instantaneously current and voltage measurements and calculate the corresponding average and rms values (sampling time \( t = 0.5 \) s), along with all the other variables typically measured in SPS processes (i.e. time, temperature, displacement, load, and gas pressure).

Specific sample configurations characterized by the absence of powders are considered in this work. In particular, we used the graphite cylindrical samples reported in Table I, along with the size of upper and lower stainless steel electrodes provided with SPS 5155 model. Graphite samples have been inserted between rams during experimental runs. It should be noted that sample IV consists of two big spacers, two small spacers and one monolithic block in order to avoid vertical contact resistances, mimicking two plungers slid into a die.

**MODELLING SECTION**

Due to heat losses by radiation from lateral surfaces as well as heat removal by cooling water in axial direction, along with variations of cross sections, a 2D model for the energy balance of SPS technique is proposed, while radial symmetry is considered. Vertical symmetry cannot be assumed due to different heights of stainless steel electrodes and the corresponding cooling circuits. Although isotropic materials are considered, temperature variation in radial and axial directions induces spatial gradients of thermophysical properties like electric and thermal conductivities and coefficient of thermal expansion.

The energy balance in cylindrical coordinates \((r, \theta)\) related to the stainless steel rams as well as the graphite samples depicted in Figure 1 and Table I is given by:

\[
\rho_i C_{rs} \frac{\partial T}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left[ r k \frac{\partial T}{\partial r} \right] + \frac{\partial}{\partial z} \left[ k \frac{\partial T}{\partial z} \right] + \frac{1}{\rho_{ms}} \left( \left( \frac{\partial \varphi}{\partial r} \right)^2 + \left( \frac{\partial \varphi}{\partial z} \right)^2 \right) \quad i = \begin{cases} \text{Stainless Steel} \\ \text{Graphite} \end{cases}
\]

with the initial condition \( T = T_0 \) at \( t=0 \), while boundary conditions are reported in Figures 2 and 3. The meaning of the other symbols is reported in the Nomenclature Section. Only contact resistances at stainless steel-graphite interfaces are considered, with a local joule heat \((q_c)\) due to electric contact resistance, which has been equally split between the materials at the interface (i.e. \( f = 0.5 \) is considered when solving the model). The following steady-state conduction model:

\[
\frac{1}{r} \frac{\partial}{\partial r} \left( \frac{1}{\rho_{ms}} r \frac{\partial \varphi}{\partial r} \right) + \frac{\partial}{\partial z} \left( \frac{1}{\rho_{ms}} \frac{\partial \varphi}{\partial z} \right) = 0 \quad i = \begin{cases} \text{Stainless Steel} \\ \text{Graphite} \end{cases}
\]

coupled with the boundary conditions reported in Figures 2 and 3, is adopted for describing the electrical behavior inside the SPS system. Only contact resistances at stainless steel-graphite interfaces are considered, while equipotential conditions for electrode surfaces in contact with copper bars (cf.
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Figure 1) have been adopted. The resistive portion of the rms voltage ($\varphi$) is given by the equation $\varphi = R \frac{V}{I_{RMS}}$, where resistance is determined from the measured average voltage and electric current ($R = \frac{V}{I}$). The pseudo isostatic equilibrium model adopted to simulate the mechanical behavior of SPS systems is reported elsewhere for sake of brevity.

Table I: Samples configurations and dimensions (not in scale) of graphite and stainless steel elements of the SPS system investigated.

|     | Thermocouple | 8 cm | | Thermocouple | 8 cm |
|-----|--------------|------||               |      |
| I   |              | 1.45 cm | | | 1.45 cm |
|     | Thermocouple | 2 cm | | Thermocouple | 0.75 cm |
|     | Thermocouple | 3 cm | | Thermocouple | 0.75 cm |
|     | Thermocouple | 3 cm | | | |
| III | (two big spacers + two small spacers + one plunger) | | IV (two big spacers + two small spacers + one monolithic die & plungers ensemble) |
| Lower electrode | 26 cm | | Upper electrode | 16.4 cm |

The resulting system of differential-algebraic-integral equations has been solved by FEM numerical technique. In particular, the commercial software COMSOL MULTIPHYSYCS® 3.2 has been adopted (numbers of DOF and elements equal to about 25000 and 3000, respectively). Parameters used for computations are reported in Table II and Figure 4. In particular, heat transfer coefficient ($h$) is calculated as discussed elsewhere while heat graphite thermal conductivity determination deserves a comment, since different data are reported in the literature. In this work, $h(T)$ is evaluated by averaging the temperature dependences given by the two different references available in the literature consistent with the only value (100 W m$^{-1}$ C$^{-1}$ at 25°C) provided by ATAL vendor for the graphite AT101. The only unknown model parameters remain therefore thermal and electric conductances, $C_T$ and $C_E$, at the horizontal contacts between stainless steel electrodes and graphite spacers (cf. see
boundary conditions in Figures 2-3). The determination of the dependence of these two parameters on temperature and applied mechanical load is described in the following section.

\[
-k_n \nabla T \cdot \vec{n} = h (T - T_0)
\]
heat loss by cooling water
\[
\nabla \phi \cdot \vec{n} = 0
\]
no current external to circuit

\[
\frac{\partial T}{\partial r} = 0
\]
radial
\[
\frac{\partial \phi}{\partial r} = 0
\]
symmetry

\[
-k_n \nabla T \cdot \vec{n} = h (T - T_0)
\]
heat loss by cooling water
\[
\nabla \phi \cdot \vec{n} = 0
\]
no current external to circuit

\[
T = T_0
\]
\[
\phi = \phi_0
\]

\[
\int_{S_{pol}} \frac{1}{\rho_{pol}} \nabla \phi \cdot \vec{n} \, dS = I_{res}
\]

\[
-k_i \nabla T \cdot \vec{n} = \eta_i \nu \left( T^4 - T_i^4 \right)
\]
heat loss by radiation

\[
i = \begin{cases} 
\text{Stainless Steel} \\
\text{Graphite}
\end{cases}
\]

\[
\nabla \phi \cdot \vec{n} = 0
\]
no current external to circuit

Figure 2: Initial and boundary conditions (except those involving contacts resistances) for the energy balance and steady-state electric conduction model equation (not in scale).

RESULTS AND DISCUSSION

In what follows, the results related to the comparison between experimental data and model results will be illustrated by considering both the fitting procedure adopted to evaluate the unknown parameters and the prediction capability of the proposed SPS apparatus model.

Figure 5 reports the direct comparison between temperature and voltage temporal profiles when sample I and sample II are used under the same operating conditions, i.e., a rectangular profile of rms current (amplitude 1200 A, 35 min duration) at 12/2 pulse sequence, and an initial mechanical load equal to 3 kN. It clearly follows that horizontal contact resistances between graphite elements can be neglected. Figure 6 reports the same comparison when a graphite foil (0.13 mm thick, Alfa Aesar) is inserted at the contacts of sample I with stainless steel electrodes.
@ Contact Interfaces

\[
\frac{1}{\rho_{d,2}} \frac{\partial \varphi}{\partial z} - \frac{1}{\rho_{d,1}} \frac{\partial \varphi}{\partial x} = - \frac{1}{\rho_{d,1}} \frac{\partial \varphi}{\partial z} = -j
\]

\[
q_n = f \ C_e (\varphi_1 - \varphi_2) \leftrightarrow q_e = \ C_e (T_1 - T_2)
\]

\[
q_{e2} = (1 - f) \ C_e (\varphi_1 - \varphi_2)
\]

\[
-k_e \ \frac{\partial T}{\partial z} = q_e - q_n \quad -k_e \ \frac{\partial T}{\partial z} = q_e + q_{e2}
\]

\[
\left. \frac{\partial \varphi}{\partial z} \right|_{A} = \left. \frac{\partial \varphi}{\partial z} \right|_{B}
\]

\[
\left. \frac{\partial T}{\partial z} \right|_{A} = \left. \frac{\partial T}{\partial z} \right|_{B}
\]

Figure 3: Boundary conditions involving contacts resistances for the energy balance and the steady-state electric conduction model (not in scale).

The experimental runs are repeated several times as reported in Figures 5 and 6 in order to appreciate the reproducibility level obtainable with SPS systems. Since significant differences are found in terms of both temperature and voltage temporal profiles, it is apparent that both thermal and electric contact resistances between graphite and stainless steel elements need to be taken into account. Indeed, according to Madhusudana,\textsuperscript{21} conducting interstitial or filler material inserted in the gaps left by actual solid-solid point contact, increases the real surface of contact between interfacing elements, thus reducing conduction resistances. Therefore, in our case the presence of graphite foil reduces the relevant horizontal contact resistances, so that lower temperature and voltage temporal profiles are obtained. Presumably, machined graphite parts used in this work possess a lower surface roughness than that of stainless steel electrodes provided with the SPS 5158 model. It is worth noting that, in order to experimentally highlight the presence of thermal and electric contact resistances in horizontal position, the lowest applicable mechanical load and the higher nominal contact surface among the available samples (cf. Table 1) have been used.

According to Madhusudana\textsuperscript{21} and Babu et al.,\textsuperscript{22} thermal and electric conduction conductances are related to temperature and applied mechanical load as follows:

\[
C_t(T, P) = \alpha_t \ k_{\text{hard}} \left( \frac{P}{H_{\text{hard}}} \right)^{\gamma_P}
\]  

(3)
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\[ C_L(T,P) = \alpha_C \sigma_{d,horn}(P/H_{horn})^2 \]  

where \( P = F/S \) represents the mechanical pressure uniformly applied at the contact surface area \( S \) between graphite and stainless steel.

Table II: Model parameters.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>( C_{p,o} )</td>
<td>see Figure 4</td>
<td>11</td>
</tr>
<tr>
<td>( C_{p,n} )</td>
<td>see Figure 4</td>
<td>12</td>
</tr>
<tr>
<td>( E_o )</td>
<td>( 1.1 \times 10^4 ) [N mm(^{-2})]</td>
<td>ATAL vendor</td>
</tr>
<tr>
<td>( E_n )</td>
<td>( 19.3 \times 10^5 ) [N mm(^{-2})]</td>
<td>13</td>
</tr>
<tr>
<td>( f )</td>
<td>0.5</td>
<td>9</td>
</tr>
<tr>
<td>( h )</td>
<td>4725 [W m(^{-2}) K(^{-1})]</td>
<td>This work</td>
</tr>
<tr>
<td>( H_o )</td>
<td>3.5 \times 10^6 [Pa]</td>
<td>ATAL vendor</td>
</tr>
<tr>
<td>( H_n )</td>
<td>1.92 \times 10^5 [Pa]</td>
<td>14</td>
</tr>
<tr>
<td>( k_o )</td>
<td>see Figure 4</td>
<td>ATAL vendor; 12; 15</td>
</tr>
<tr>
<td>( k_n )</td>
<td>see Figure 4</td>
<td>16</td>
</tr>
<tr>
<td>( T_o )</td>
<td>298.15 [K]</td>
<td>This work</td>
</tr>
<tr>
<td>( \alpha_o )</td>
<td>see Figure 4</td>
<td>17</td>
</tr>
<tr>
<td>( \alpha_n )</td>
<td>see Figure 4</td>
<td>13</td>
</tr>
<tr>
<td>( \eta_o )</td>
<td>0.85</td>
<td>6; 8; 9; 18</td>
</tr>
<tr>
<td>( \eta_n )</td>
<td>0.4</td>
<td>19</td>
</tr>
<tr>
<td>( \nu )</td>
<td>5.67 \times 10^4 [W m(^{-2}) K(^{-1})]</td>
<td>20</td>
</tr>
<tr>
<td>( \rho_o )</td>
<td>1750 [kg m(^{-3})]</td>
<td>ATAL vendor</td>
</tr>
<tr>
<td>( \rho_n )</td>
<td>8000 [kg m(^{-3})]</td>
<td>12</td>
</tr>
<tr>
<td>( \rho_{d,o} )</td>
<td>see Figure 4</td>
<td>ATAL vendor</td>
</tr>
<tr>
<td>( \rho_{d,n} )</td>
<td>see Figure 4</td>
<td>12</td>
</tr>
<tr>
<td>( \nu_o )</td>
<td>0.33</td>
<td>This work</td>
</tr>
<tr>
<td>( \nu_n )</td>
<td>0.29</td>
<td>13</td>
</tr>
</tbody>
</table>

The parameters \( k_{horn} \) and \( \sigma_{d,horn} \) take into account the temperature dependence of contact conductances, and are expressed as follows using the harmonic mean of the individual thermal and electric conductivities of graphite and stainless steel:

\[ k_{horn}(T) = \frac{2 k_o(T) k_n(T)}{k_o(T) + k_n(T)} \]  

\[ \sigma_{d,horn}(T) = \frac{2 \sigma_{d,o}(T) \sigma_{d,n}(T)}{\sigma_{d,o}(T) + \sigma_{d,n}(T)} \]
Figure 4: Thermophysical properties of AT 101 (ATAL) graphite: heat capacity (a), electrical resistivity (b), thermal conductivity (c), and coefficient of thermal expansion (d), and AISI 304 stainless steel: heat capacity (e), electrical resistivity (f), thermal conductivity (g), and coefficient of thermal expansion (h). Dots represent experimental data taken from the literature as specified in Table II, while lines are the corresponding fitting curves adopted for the numerical simulation.
Figure 5: Comparison between sample I and sample II in terms of temporal profiles of measured temperature ($r = 0, z = 28 \text{ cm}$) a) and rms voltage b), when a rms current step of 1200 A is applied for 35 min with a initial load of 3 kN.

Figure 6: Comparison between sample I with and without graphite foils at stainless steel/graphite interfaces in terms of temporal profiles of measured temperature ($r = 0, z = 28 \text{ cm}$) a) and rms voltage b), when a rms current step of 1200 A is applied for 35 min and 25 min, correspondingly, with a initial load of 3 kN.

Basically, the use of harmonic mean is a consequence of the series combination of the two interfacing materials, which characterize the real contact. Analogously, in this work the parameter $H_{\text{harm}}$ is the harmonic mean between graphite and stainless steel hardnesses:

$$H_{\text{harm}} = \frac{2H_G H_S}{H_G + H_S}$$  \hspace{1cm} (7)

The remaining parameters appearing in Eqs. 3-4 (i.e., $\alpha_r$, $\beta_r$, $\alpha_z$, and $\beta_z$) are the only adjustable ones in our model. They depend on surface roughness and plastic behavior of contact between graphite and stainless steel interfaces. These four parameters have been fitted by direct comparison between modeling results and experimental data in terms of temporal profiles of resistive portion of rms voltage measured between electrodes' ends, $\phi_h$, and temperature measured by a thermocouple placed in axial position inside sample I, as depicted in Table I. In particular, experimental runs conducted under applied initial mechanical loads in the range 3-50 kN, at a constant $f_{\text{cycle}}$ of 1200 A at 12/2 pulse cycle,
have been performed. Experimental data for the case of 3, 20 and 50 kN are compared with modeling results in Figures 7-9, respectively, while fitted parameters are reported in Table III along with the 95\% confidence band and correlation coefficients.

Figure 7: Comparison between experimental data and model results in terms of temporal profiles of temperature a) taken at the center of the lower big spacer \((r = 0, \ z = 28 \text{ cm})\), resistive portion of rms voltage b), when sample 1 is submitted to a rms electric current step of 1200 A for 20 min, with a load of 3 kN.

Figure 8: Comparison between experimental data and model results in terms of temporal profiles of temperature a) taken at the center of the lower big spacer \((r = 0, \ z = 28 \text{ cm})\), resistive portion of rms voltage b), when sample 1 is submitted to a rms electric current step of 1200 A for 20 min, with a load of 30 kN.

Figure 9: Comparison between experimental data and model results in terms of temporal profiles of temperature a) taken at the center of the lower big spacer \((r = 0, \ z = 28 \text{ cm})\), resistive portion of rms voltage b), when sample 1 is submitted to a rms electric current step of 1200 A for 20 min, with a load of 50 kN.
Table III: Fitted parameters of contact conductances along with 95% confidential band, and correlation coefficient R.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value ± Error</th>
<th>R</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\alpha_r$</td>
<td>22810 ± 4.6 [m$^{-1}$]</td>
<td>0.9368</td>
</tr>
<tr>
<td>$\beta_r$</td>
<td>1.08 ± 0.23</td>
<td></td>
</tr>
<tr>
<td>$\alpha_e$</td>
<td>64 ± 1.38 [m$^{-1}$]</td>
<td>0.9609</td>
</tr>
<tr>
<td>$\beta_e$</td>
<td>0.35 ± 0.05</td>
<td></td>
</tr>
</tbody>
</table>

The corresponding values of electric and thermal conductances obtained in the range of temperature and mechanical load investigated in this work are $0.25±1.7 \times 10^6 \Omega^{-1} \text{m}^{-2}$ and $0.007±2 \times 10^3 \text{W m}^{-2} \text{K}^{-1}$, respectively. It is worth noting that constant values for electric and thermal contact conductances in horizontal position equal to $1.25 \times 10^7 \Omega^{-1} \text{m}^{-2}$ and $2.4 \times 10^3 \text{W m}^{-2} \text{K}^{-1}$, respectively, were obtained by Zavallangos et al. These discrepancies may be ascribed to the fact that Zavallangos et al. considered the electrodes made of graphite (instead of stainless steel), whose thermophysical properties are rather different from those used in the present paper. A satisfactory simulation of SPS system behavior in terms of temperature, voltage is obtained, especially at higher applied mechanical loads, when temperature and voltage decrease due to lower contact resistances.

Model reliability is further tested by comparing model results with experimental data obtained using samples III and IV and adopting operating conditions significantly far from those used during fitting procedure. In particular, the $I_{	ext{rms}}$ value is decreased from 1200 A to 670 A and 980 A. It is worth noting that also sample geometry is changed and, accordingly, a different joule heat generation is expected to produce different temperature distribution, since electric resistance varies as function of cross section surface area. For the same reason, voltage changes as well. The obtained results are shown in Figures 10-11.

Figure 10: Comparison between experimental data and model predictions in terms of temporal profiles of temperature a), resistive portion of rms voltage b), when sample III is submitted to a rms electric current step of 670 A for 20 min, with a load of 3.5 kN. Temperature is taken at three different locations, i.e. center of lower big spacer ($r = 0, z = 28 \text{ cm}$), center of lower small spacer ($r = 0, z = 31.5 \text{ cm}$), and center of plunger ($r = 0, z = 34 \text{ cm}$).

It is seen that the developed model is able to entirely predict SPS system behavior in terms of temperature, voltage, and displacement temporal profiles. In order to further highlight the importance of taking into account contact resistances, Figure 11 reports model predictions when the fitted contact resistances are neglected (i.e. model equations are solved by setting very high contact conductances).
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In this particular case, temporal profiles of temperatures and voltage are underestimated. As expected, the larger discrepancy is related to temperature taken in axial position of lower graphite small spacer, i.e. the one closer to the relevant contact resistances. Moreover, cooling dynamics, occurring once the electric current is turned off, is overestimated so that displacement rises relatively too quickly to the initial value. It is apparent that sample 1V has been chosen to make these considerations, since, among those investigated in this work, is the most similar in size to the actual configuration used when sintering/synthesis experiments are performed.

![Graph 1](image1.png)

**Figure 11**: Comparison between experimental data and model predictions in terms of temporal profiles of temperature a), resistive portion of rms voltage b), and displacement c), when sample 1V is submitted to a rms electric current step of 980 A for 10 min, with a load of 3.5 kN. Temperature is taken at two different locations, i.e. center of lower small spacer (x = 0, z = 31.5 cm) and inside the die (x = 1.1 cm, z = 32.25 cm).

**CONCLUDING REMARKS**

In this paper, the quantitative description of SPS system behavior in the case of the absence of powders is addressed in the framework of a novel heuristic approach. In particular, electric and thermal resistances in horizontal contacts as functions of temperature and applied mechanical load have been obtained by direct comparison between model results and experimental data. The electric behavior of the system is described in details by highlighting the importance of considering rms electric current and voltage (whenever pulse cycle is adopted) when joule effect needs to be quantitatively determined. Model reliability is for the first time tested by comparing model predictions with experimental data obtained at operating conditions significantly far from those ones considered during fitting procedure. The proposed model has been successfully compared from a quantitative point of view to the measured temperature, voltage and displacement when rms current, geometry and initial mechanical load are set. This work represents the first step towards the development of a complete model for the simulation of SPS system behavior where sintering/synthesis of powders sample takes place.

**ACKNOWLEDGMENTS**

IM (Innovative Materials) Srl, Italy is gratefully acknowledged for granting the use of SPS apparatus.

**NOMENCLATURE**

- \( C_p \) electric contact conductance, [\( \Omega^{-1} \text{m}^2 \)]
- \( C_{p,g} \) graphite heat capacity, [J kg\(^{-1}\) K\(^{-1}\)]
- \( C_{p,st} \) stainless steel heat capacity, [J kg\(^{-1}\) K\(^{-1}\)]
- \( C_T \) thermal contact conductance, [W m\(^{-2}\) K\(^{-1}\)]
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\[ E_g \]  \quad \text{graphite Young's modulus, [N mm}^{-2}\text{]} \]

\[ E_s \]  \quad \text{stainless steel Young's modulus, [N mm}^{-2}\text{]} \]

\[ F \]  \quad \text{mechanical load, [N]} \]

\[ f \]  \quad \text{fraction of localized joule heat due to electric contact resistance} \]

\[ H_g \]  \quad \text{graphite hardness, [N m}^{-2}\text{]} \]

\[ H_{\text{mean}} \]  \quad \text{harmonic mean between graphite and stainless steel hardness, [N m}^{-2}\text{]} \]

\[ H_s \]  \quad \text{stainless steel hardness, [N m}^{-2}\text{]} \]

\[ h \]  \quad \text{heat transfer coefficient, [W m}^{-2}\text{K}^{-1}\text{]} \]

\[ I \]  \quad \text{instantaneous electric current, [A]} \]

\[ I \]  \quad \text{temporal average electric current, [A]} \]

\[ I_{\text{rms}} \]  \quad \text{temporal root mean squared electric current, [A]} \]

\[ j \]  \quad \text{rms current density, [A m}^{-2}\text{]} \]

\[ k_g \]  \quad \text{graphite thermal conductivity, [W m}^{-1}\text{K}^{-1}\text{]} \]

\[ k_{\text{mean}} \]  \quad \text{harmonic mean between graphite and stainless steel thermal conductivities, [W m}^{-1}\text{K}^{-1}\text{]} \]

\[ k_s \]  \quad \text{stainless steel thermal conductivity, [W m}^{-1}\text{K}^{-1}\text{]} \]

\[ L \]  \quad \text{self-inductance, [H]} \]

\[ \mathbf{n} \]  \quad \text{surface area unit vector in outward direction} \]

\[ P \]  \quad \text{applied mechanical pressure, [N m}^{-2}\text{]} \]

\[ q \]  \quad \text{heat flux due to thermal contact resistance, [W m}^{-2}\text{K}^{-1}\text{]} \]

\[ q_{\text{e}} \]  \quad \text{localized joule heat flux due to electric contact resistance, [W m}^{-2}\text{K}^{-1}\text{]} \]

\[ R \]  \quad \text{electric resistance, [\Omega]} \]

\[ T \]  \quad \text{temperature, [K]} \]

\[ T_0 \]  \quad \text{initial ambient temperature, [K]} \]

\[ t \]  \quad \text{time, [s]} \]

\[ r \]  \quad \text{radial coordinate, [m]} \]

\[ S \]  \quad \text{surface area, [m}^2\text{]} \]

\[ V \]  \quad \text{instantaneous voltage, [V]} \]

\[ \bar{V} \]  \quad \text{temporal average voltage, [V]} \]

\[ V_L \]  \quad \text{instantaneous inductive voltage, [V]} \]

\[ V_R \]  \quad \text{instantaneous resistive voltage, [V]} \]

\[ V_{\text{rms}} \]  \quad \text{temporal root mean squared voltage, [V]} \]

\[ z \]  \quad \text{axial coordinate, [m]} \]

Greek letters

\[ \alpha_g \]  \quad \text{adjustable parameter in Eq. 3, [m]} \]

\[ \alpha_g \]  \quad \text{graphite coefficient of thermal expansion, [K}^{-1}\text{]} \]

\[ \alpha_s \]  \quad \text{stainless steel coefficient of thermal expansion, [K}^{-1}\text{]} \]

\[ \alpha_T \]  \quad \text{adjustable parameter in Eq. 3, [m]} \]

\[ \beta_g \]  \quad \text{adjustable parameter in Eq. 4} \]

\[ \beta_T \]  \quad \text{adjustable parameter in Eq. 3} \]

\[ \varphi \]  \quad \text{resistive rms voltage, [V]} \]
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\[ \varphi_c \quad \text{resistive rms voltage between electrodes ends, [V]} \]

\[ \eta_c \quad \text{graphite emissivity} \]

\[ \eta_{ss} \quad \text{stainless steel emissivity} \]

\[ \nu \quad \text{Stefan-Boltzmann constant, [W m}^{-2} \text{K}^{-4]} \]

\[ \rho_g \quad \text{graphite density, [kg m}^{-3} \text{]} \]

\[ \rho_{ss} \quad \text{stainless steel density, [kg m}^{-3} \text{]} \]

\[ \rho_{elec} \quad \text{graphite electric resistivity, [\Omega m]} \]

\[ \rho_{elec}^{har} \quad \text{harmonic mean of graphite and stainless steel electric resistivities, [\Omega m]} \]

\[ \rho_{elec}^{sm} \quad \text{stainless steel electric resistivity, [\Omega m]} \]

\[ \sigma_{elec} \quad \text{graphite electric conductivity \left( \frac{1}{\rho_{elec}} \right), [\Omega}^{-1} \text{m}^{-1]} \]

\[ \sigma_{elec}^{sm} \quad \text{harmonic mean of graphite and stainless steel electric conductivities, [\Omega}^{-1} \text{m}^{-1]} \]

\[ \sigma_{elec}^{sm} \quad \text{stainless steel electric conductivity \left( \frac{1}{\rho_{elec}^{sm}} \right), [\Omega}^{-1} \text{m}^{-1]} \]

REFERENCES