# DETERMINATION OF THERMAL-ELECTRICAL PARAMETERS OF GRAPHITE DIE FOR SPARK PLASMA SINTERING

**Fausto Kenzo Chinen, fausto.chinen@poli.usp.br**
**Julio Cezar Adamowski, jcadamow@usp.br**
Department of Mechatronics and Mechanical Systems Engineering,
Escola Politécnica da Universidade de São Paulo,
Av. Prof. Mello Moraes, 2231, 05508-900, São Paulo, SP, Brazil

***Abstract.*** *Functionaly Graded Materials (FGMs) are materials characterized by a variation in composition or microstructure over volume. These variations can cause changes in the material's properties that are explored in order to design new materials with specific purposes. One technique used to produce such materials is Spark Plasma Sintering (SPS). SPS is a sintering technique where pulsed direct electric current passes through a graphite sintering die, generating heat by Joule effect. The fact that the heat is generated much closer to the sample causes greater heating and cooling rates compared to conventional sintering techniques, such as Hot Pressing, allowing the manufacturing of FGMs by stacking layers of different material compositions. The sintering process of the stacked materials requires a precise control of the temperature gradient of both the sample and the die. This can be attained by designing dies with controlled shapes using thermal and electrical properties of the graphite. The existing studies rely on numerical simulation of the heating and the heat transfer but all of them used mean material properties instead of the actual properties of the specific graphite from their suppliers. Also, these studies simplify the SPS machine's heat transfer without specifying how the machine actually removes heat from the system. This work presents a methodology to obtain the thermal and electrical properties of the graphite and thermal parameters of the SPS machine by combining experimental and numerical simulation. This is done by solving the inverse problem of adjusting the parameters of the simulation to better translate the experimental process. Material properties and the machine's heat transfer parameters are set as 11 variables in an optimization problem where the goal is to minimize the mean square difference between simulated values and experimental data. The simulation is accomplished using the finite elements software ANSYS and the nonlinear unconstrained optimization algorithm Simplex is run in the software MATLAB. The simulation includes nonlinear thermo-electrical analysis computing a new temperature distribution each 1 second. The experimental data was acquired using a pyrometer pointed to known points on the graphite surface. The parameters are obtained using a single graphite plug and the results are used to simulate the temperature distribution of a different validation arrangement, showing reasonable agreement with experimental measurement of external surface temperature.*

***Keywords****: Spark Plasma Sintering (SPS), Functionally Graded Materials (FGM), Temperature Distribution, Inverse Problem, Graphite Die*

## 1. INTRODUCTION

Spark Plasma Sintering (SPS), also known as Field Activated Sintering Technique (FAST) and Pulsed Electric Current Sintering (PECS), is a novel sintering technology that is able to sinter samples quicker and sometimes better than in similar processes (Omori, 2000; Shen et al., 2002, Khor et al., 2003), such as conventional hot pressing (HP). The main difference between HP and SPS is that in conventional HP, the sample is placed in a press inside a chamber where an electrical resistor heats the hole chamber, heating the die and the sample while in the SPS process, the heating is caused by Joule effect when a pulsed direct electric current is passed trough the conductive die and through the sample, if conductive. While the physical mechanisms responsible for this improvement in sintering times is still controversial (Zhang et al., 2005; Tiwari, Basu and Biswas, 2008), the usefulness of the quicker sintering is widely recognized in several different material manufacturing (Omori, 2000; Shen et al., 2002, Lee et al., 2003, Khor et al., 2003, Kumar, Cheang and Khor, 2003).

The high sintering speed is especially useful to the manufacturing of Functionally Graded Materials (FGM) since the speed of the process causes greater temperature gradients which are needed to successfully sinter different materials with different sintering temperatures. The composition of the FGM is then controlled by the initial powder composition and placement.

In order to better control the process, one has to control the temperature gradient. This can be done by using dies with controlled shapes designed to attain one specific temperature under specific process' conditions. The design of dies can be helped by computer simulation of the thermal-electrical states during a SPS process. Simulations of the temperature distribution in the graphite die used in SPS sintering have already been done (Anselmi et al., 2005; Matsugi et al., 2004; Vanmeensel et al., 2005; ; Yucheng and Zhengyi, 2002; Zavalingos et al., 2004) but these studies simplified the process by not modeling accurately how the heat transfers occur inside the chamber or by using materials' thermal-electrical properties without specifying how they vary with the temperature. These properties also are different considering different models and different graphite suppliers.

Yucheng and Zengi (2002) modeled the temperature distribution analytically but in a steady-state condition, presenting high temperature gradients inside the sample and substantial difference between the temperature of the sample and the temperature measured in the die, used in the control loop. Matsugi et al. (2004) presented results from a finite differences method analysis at steady-state, comparing conductive and insulator samples. Zavaliangos et al. (2004) presented good results from a finite element method analysis that included thermal and electrical resistances in some of the surfaces. The resistances were estimated in previous work by Zhang (2003) and the comparison was made based on one experimental point. Anselmi et al. (2005) presented results from finite volume method analysis and compared input from thermocouple and pyrometer. Vanmeensel et al. (2005) compared finite elements method result and experimental data from measurement of temperature of one point over time with and without superficial resistances. Tiwari, Basu and Biswas (2008) modeled the temperature distribution with different material properties but made no comparison with any experimental data.

This work presents a methodology to obtain the material's thermal-electrical properties of the graphite and the thermal parameters of a modeled version of the SPS machine by solving the inverse problem of determining unknown finite element method parameters using commercial finite element method software and nonlinear unconstrained optimization algorithm.

## 2. EXPERIMENTAL PROCEDURE

The experiments were carried out using a commercial SPS machine (Dr. Sinter SPS-1050 from Sumitomo). The machine consists of a uniaxial press inside a vacuum chamber, the voltage is applied between both rams of the press and the current flows through the graphite spacers and graphite die. The temperature is controlled by a PID controller which input is the temperature curve programmed and the feedback is the temperature measured by a thermocouple or by a pyrometer. The output of the PID controller is the electrical current that passes through the apparatus. The SPS-1050 has a current maximum of 5000 A and a maximum force of the press of 100 kN. Data from temperature, displacement, displacement rate, axial force applied, voltage between the rams, electrical current and chamber pressure are computer logged. Typical arrangement for a SPS process using the pyrometer as temperature sensor is shown in the Fig. 1.

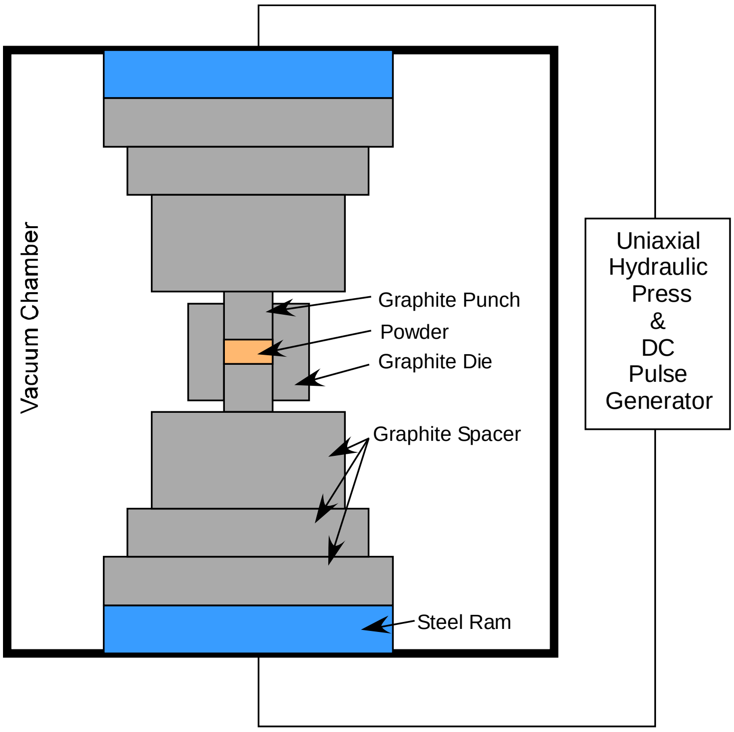


Figure 1. Typical arrangement for a SPS process

The main concept of this method is to use an optimization algorithm to match the numerical results to the experimental data, thus calibrating the simulation. The chosen parameters were the specific heat capacity, the thermal conductivity and the electrical resistivity of the graphite varying with the temperature, modeled as a polynomial expression of grade 3, 3 and 4 respectively. To model the water-cooled rams, it was considered a disk of steel with thickness to be determined as the final parameter.

The algorithm was run considering the data set of an initial, simpler, experiment arrangement and the results were compared to a second arrangement, more complex and similar to the actual SPS process, in order to validate the method.

## 2.1. Initial arrangement

The initial arrangement used a simple graphite punch instead of the whole graphite die with punches. The temperature was measured by a pyrometer, pointed to five known points of the graphite punch at heights of 0 mm, 2 mm, 4 mm, 6 mm and 8 mm from the central plane. Figure 2 illustrates the arrangement and the elements are described in Tab. 1.

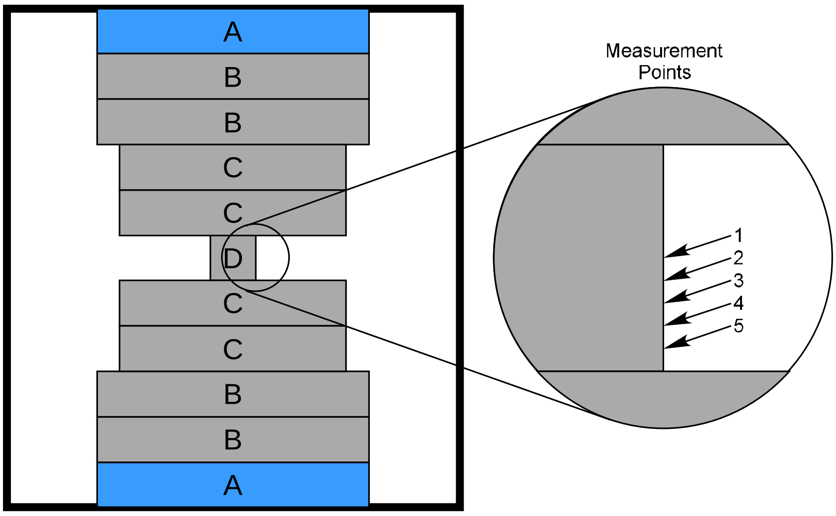


Figure 2. Initial arrangement

Table 1. Details of the elements of the initial arrangement

| Elements | Function | Material | Diameter | Height |
|---|---|---|---|---|
| A | Press ram and electrode | Stainless Steel | 120 mm | - |
| B | Spacer, thermal buffer | Graphite | 120 mm | 20 mm |
| C | Spacer, thermal buffer | Graphite | 100 mm | 20 mm |
| D | Punch | Graphite | 20 mm | 20 mm |

As the temperature of just one height could be logged at a time, the heating and cooling had to be done 5 times, changing the point that the pyrometer pointed at. To maintain initial conditions, long cooling times were used between the 5 different runs.

As the 5 runs had to be very similar, the temperature could not be used as feedback for the control loop. Therefore, the process was run as fixed current of 1415 A for 60 s. The current was set so that the temperature was high enough to have a considerable amount of reads of the pyrometer, which readings begin at 570 °C, but not too high, as the system would take too much time to cool down. In all runs, the vacuum pressure was stable at around 12 Pa and the axial pressure was stable at 30MPa. The data was logged at 1 Hz.

## 2.2. Optimization and numerical simulation

The optimization algorithm was run in the commercial software Matlab (The Mathworks Inc.). The 15 parameters were initially set to values from literature (Anselmi, 2005) and normalized. The nonlinear unconstrained Simplex algorithm called the numerical simulation program through command line, passing the values of the parameters as well. The numerical simulation run in the commercial software ANSYS (ANSYS, Inc.) using the element PLANE67, which is axissimetric nonlinear thermal-electrical element. The simulation was run with time steps of 1 s and total time of 120s. It was also used the central horizontal plane as symmetry plane. The model used is shown in Fig. 3. The temperature at the bottom of the ram was considered constant at 23 °C and also the temperature of the chamber was considered 23 °C for radiation heat transfer purpose. No convection was considered as it is negligible in chamber pressure of 12 Pa.

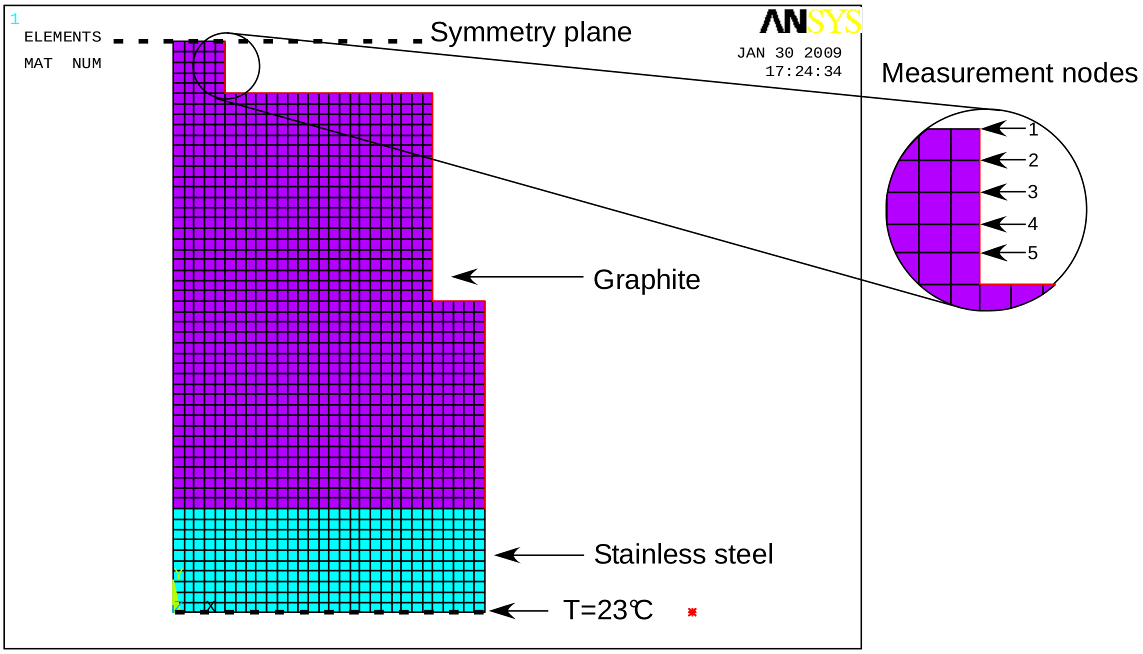


Figure 3. Simulation model for the initial experiment

### 2.3. Validation arrangement

The results of the inverse problem solving were validated with a more complex arrangement. This arrangement was similar to the first one except for the use of two more punches and graphite contact resistances. The temperature was, again, measured by a pyrometer, pointed to known heights of the graphite punches. Figure 4 illustrates the arrangement and the elements are described in Tab. 2.

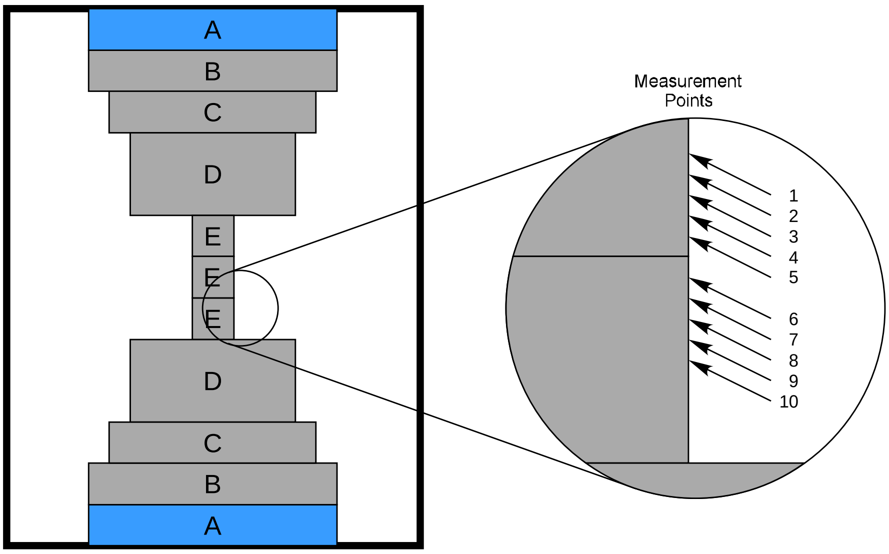


Figure 4. Validation arrangement

Table 2. Details of the elements of the validation arrangement

| Elements | Function | Material | Diameter | Height |
|---|---|---|---|---|
| A | Press ram and electrode | Stainless Steel | 120 mm | - |
| B | Spacer, thermal buffer | Graphite | 120 mm | 20 mm |
| C | Spacer, thermal buffer | Graphite | 100 mm | 20 mm |
| D | Spacer, thermal buffer | Graphite | 80 mm | 40 mm |
| E | Punch | Graphite | 20 mm | 20 mm |

As done before, the heating and cooling cycles were done 5 times, changing the point that the pyrometer pointed at. To maintain initial conditions, long cooling times were used between the 5 different runs.

The process was run as fixed current of 900 A for 60 s for the same reasons of the first arrangement. In all runs, the vacuum pressure was stable at around 12 Pa and the axial pressure was stable at 50 MPa. The data was logged at 0.1 Hz.

## 3. RESULTS AND DISCUSSION

Figure 5 compares the initial result from the simulation with the experimental data. Figure 6 compares the final result from the simulation, after 1200 iterations.

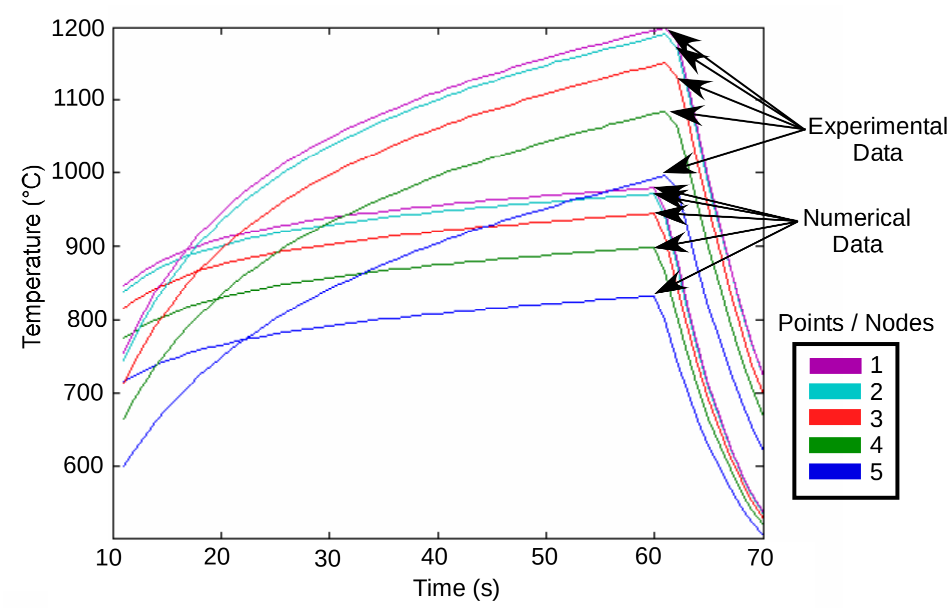


Figure 5. Initial difference between numerical and experimental data

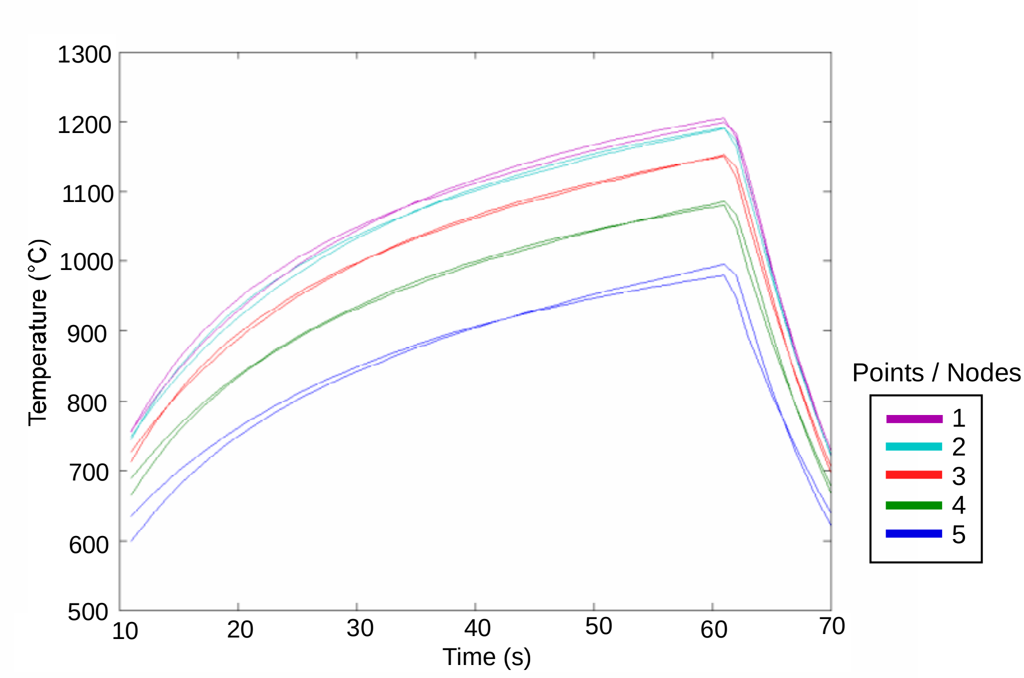


Figure 6. Final difference between numerical and experimental data

The proximity of both curve sets after the optimization process indicates that there were enough parameters in the optimization problem, allowing a good fit between the curves.

The data set that resulted from the optimization algorithm was used in the numerical model of the validation arrangement. The results of the numerical simulation are compared to experimental data in Fig. 7. The continuous lines are the numerical data and the dotted lines are the experimental data. Each line represent the temperature measured on a known point, hotter points are closer to the horizontal symmetry plane.

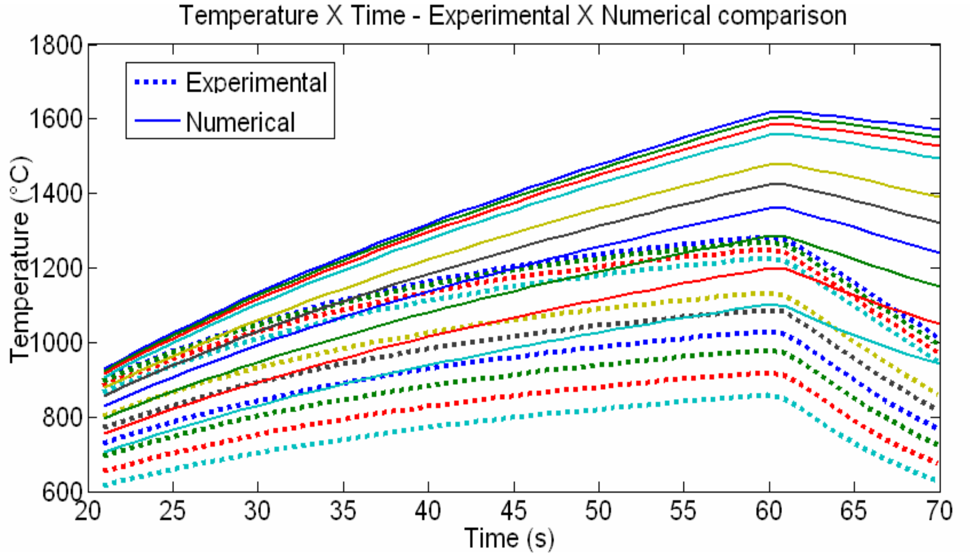


Figure 7. Difference between numerical and experimental data for validation arrangement

The difference between both curve sets is of 28% at peak temperature. One possible reason for the difference is the Thermal barrier caused by contact thermal resistance for it was not present in the initial arrangement and therefore, was not adjusted in the model.

Although the experimental procedure had a good repeatability, it was not studied whether small disturbances in the initial curve set cause smaller or bigger disturbances in the calculated parameters. Small disturbances in the temperature curves can be caused by different cooling water temperature, by different temperatures outside the vacuum chamber wall and by small convection heat transfers among other less probable causes.

To verify the actual cause of experimental and numerical values, the initial temperature curve set must be more complex, including contact thermal resistances as well as contact electrical resistances. The effect of the contact resistance can be measured by reading the temperature profile before and after forcing a change in contact quality by applying different axial pressures. The effect of small disturbances in temperature curves can be observed by forcing small disturbances and reading the difference in the calculated parameters.

## 4. CONCLUSION

A method for characterization of graphite thermal-electrical properties and of SPS process thermal parameters was developed successfully using an optimization algorithm to solve the inverse problem. The results of the validation indicated the lack of the thermal resistance parameter that will be added in near future analysis. The results will be used to design graphite dies with specific shapes that will produce a specific temperature gradient needed for FGM materials manufacturing. Also, with the results of the analysis, one can determine the actual internal temperature of the sample being processed in spite the fact that the temperature sensors can only read external die temperature.

## 5. ACKNOWLEDGEMENTS

This work was supported by scholarship from FAPESP and by grants from PETROBRAS/ANP and CNPq.

## 6. REFERENCES

Anselmi-Tamburini, U., Gennarib, S. , Garay, J.E. and Munir, Z.A., 2005, "Fundamental investigations on the spark plasma sintering/synthesis process II. Modeling of current and temperature distributions", Materials Science and Engineering A, Vol.394, pp. 139-148

Khor, K.A., Cheng, K.H., Yu, L.G. and Boey, F., 2003, "Thermal conductivity and dielectric constant of spark plasma sintered aluminum nitride", Materials Science and Engineering A, Vol. 347, pp. 300-305

Kumar, R., Cheang, P. and Khor, K.A., 2003, "Spark plasma sintering and in vitro study of ultra-fine HA and ZrO2–HA powders", Journal of Materials Processing Technology, Vol. 140, pp. 420-425

Lee, Y.I., Lee, J., Hong, S. and Kim, D., 2003, "Preparation of nanostructured TiO2 ceramics by spark plasma sintering", Materials Research Bulletin, Vol. 38, pp. 925–930

Matsugi, K., Kuramoto, H., Hatayama, T. and Yanagisawa, O., 2004, "Temperature distribution at steady state under constant current discharge in spark sintering process of Ti and Al2O3 powders", Journal of Materials Processing Technology, Vol. 146, pp. 274–281

Omori, M., 2000, "Sintering, consolidation, reaction and crystal growth by the spark plasma system (SPS)", Materials Science and Engineering A, Vol. 287, pp. 183–188

Shen, Z., Johnsson, M., Zhao, Z. and Nygren, M., 2002, "Spark Plasma Sintering of Alumina", Journal of the American Ceramic Society, Vol. 85, pp. 1921-1927

Tiwari, D., Basu, B. and Biswas, K., 2008, "Simulation of thermal and electric field evolution during spark plasma sintering", Ceramics International, Vol. 35, pp. 699–708

Vanmeensel, K., Laptev, A., Hennicke, J., Vleugels, J. and van der Biest, O., 2005, "Modelling of the temperature distribution during field assisted sintering", Acta Materialia, Vol. 53, pp. 4379–4388

Yucheng, w. and Zhengyi, F., 2002, "Study of temperature field in spark plasma sintering", Materials Science and Engineering B, Vol.90, pp. 34–37

Zavalingos, A., Zhang, J., Krammer, M. and Groza, J.R., 2004, "Temperature evolution during field activated sintering", Materials Science and Engineering A, Vol. 379, pp. 218–228

Zhang, J., 2003, "Numerical Simulation of Sintering under Electric Field", Ph.D. Dissertation, Drexel University, Philadelphia.

Zhang, J., Tan, T., Fu, Z. and Wang, W., 2005, "Densification of Cu/Ti System by Spark Plasma Sintering", JJournal of Wuhan University of Technology - Materials Science Edition, Vol. 20, pp. 83-85

## 7. RESPONSIBILITY NOTICE

The authors are the only responsible for the printed material included in this paper.