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Contact resistances in spark plasma sintering: From in-situ and ex-situ determinations to an extended model for the scale up of the process

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ABSTRACT

Heating in spark plasma sintering is a key point of this manufacturing process that requires advanced simulation to predict the thermal gradients present during the process and adjust them. Electric and thermal contact resistances have a prominent role in these gradients. Their determination is difficult as they vary with pressure and temperature. A calibration method is used to determine all of the contact resistances present within tools of different sizes. Ex situ measurements were also performed to validate the results of the in-situ calibrations. An extended predictive and scalable contacts model was developed and reveals the great importance and diversity of the contact resistances responsible for the general heating of the column and high thermal gradients between the parts. The ex/in situ comparison highlights a high lateral thermal contact resistance and the presence of a possible phenomenon of electric current facilitation across the lateral interface for the high temperatures.

1. Introduction

The spark plasma sintering technique (SPS), also known as the field-assisted sintering technique (FAST), belongs to the hot pressing technologies, where a uniaxial pressure and a pulsed direct current is applied to the die. Over the past decade, SPS has been successfully used for a wide variety of materials in the main class [1,2] (metals and alloys, ceramics, polymers and composites). The general advantages of spark plasma sintering compared to traditional hot isostatic pressing or hot pressing, are [3,4] high heating rates, short processing time and the possibility to minimize grain growth known to improve the physical, optical or mechanical properties of materials, and the attainment of high densification [5].

The main difficulties of this technology are to control the temperature and densification field in the sample. The Finite Element Modeling (FEM) of the process is a solution to predict and adjust the internal physical parameters to the target objective. These simulations are developed on numerical codes containing: i) an electro-thermal (ET) component to predict the temperature field and ii) a mechanical component (M) to predict the powder densification. Most of the time, for the ET component pure resistive heating is considered without any inductive effects which is a good approximation of the phenomena [6–9]. These models are able to predict the behavior of the electric current (different depending on the electric conductivity of the sample), the area of high heat generation often located in the punches or the presence of hot spots. One of the most difficult phenomena to determine is the electric and thermal contact resistances (ECR and TCR) present at all the inner interfaces of the SPS column. These ECR and TCR result from non-ideal interfaces between the different parts with a certain roughness or from the presence of another material. Several authors have pointed out the importance and diversity of these contacts [10–13]. Anselmi-Tamburini et al. [14] suggested the punch/sample ECR is negligible for high pressure but pointed out the importance of the punch/die contact resistance. This last interface is very difficult to study because the lateral pressure governing a large part of the behavior of this contact is influenced by the thermal expansion of the punch, the gap in the punch/die interface and the possible compaction of the third material usually present at this interface (graphite foil, etc.). All of these parameters imply a very low contact pressure and are difficult to determine. For this reason, some authors chose to access the ECR and TCR by calibration of the temperature field [10,11,15,16] or by measurement of the overall column resistance in different configurations

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Nomenclature

$\vec{J}$	Current density A m^{-2}	
$\vec{E}$	Electric field V m^{-1}	
κ	Thermal conductivity W m^{-1} K^{-1}	
σ	Electrical conductivity Ω^{-1} m^{-1}	
ρ	Density kg m^{-3}	
Cp	Specific heat capacity J kg^{-1} K^{-1}	
T	Temperature K	
σ_s	Stefan-Boltzmann's constant 5.6704.10^{-8} W m^{-2} K^{-4}	
ϕ_r	Radiative heat flux W m^{-2}	
ε	Emissivity 0.80 for graphite	
T_e	Emission surface temperature K	
T_a	Chamber wall temperature K	
ϕ_c	Conductive heat flux W m^{-2}	
$T_{inconel}$	Inconel wall temperature K	
T_w	Water temperature K	
h_c	Convective coefficient W m^{-2} K^{-1}	
J_c	Contact current density A m^{-2}	
$\dot{q}_c$	Contact heat flux W m^{-2}	
U_i (i equal 1 or 2)	Contact face electric potential V	
T_i (i equal 1 or 2)	Contact face temperature K	
ECR	Electric surface contact resistance Ω m^2	
TCR	Thermal surface contact resistance m^2 K/W	
θ	Porosity	
ρ_e	Electrical resistivity Ω m	
P	The contact pressure Pa	

[13,17]. These two in situ approaches have the advantage of quickly allowing the establishment of a heating model very close to experiment. But, the main drawback of these approaches is their domain of validity restrained to the experimental conditions of the model determination.

In order to approach a more generalized model of SPS heating, in this work we considered two approaches:

- in situ calibrations of the ECR and TCR for different geometries of the system 'punch, die, and sample' to extend the model and take into account the scaling effect.
- ex situ measurements of the ECR and TCR in different conditions of pressure and temperature to validate the in situ determinations and subsequently extend the model.

2. Experimental/computational methodology

2.1. SPS calibration experiments

All the in situ experiments for the calibrations were performed on the SPS machine (Dr. Sinter 2080, SPS Syntex Inc, Japan) of the Plateforme Nationale CNRS de Frittage Flash located at the University of Toulouse III-Paul Sabatier. The SPS column studied has six different types of contacts to be calibrated (Fig. 1). A graphite foil called papyex (® Mersen) is introduced at the punch/die, punch/sample and sample/die interfaces for easy removal of the sample and to ensure a good electrical contact. At the electrode/spacer interface, two graphite foils are introduced. The sample is 99.99% α-alumina powder (reference TM-DAR, Taimei Chemicals Co. Ltd, average initial grain size of 0.14 μm). For each test the rms value of the current delivered is measured by a Rogowki coil sensor (Power Electronic Measurements, CWT60) and temperatures are measured with K type thermocouples at different points of the SPS column.

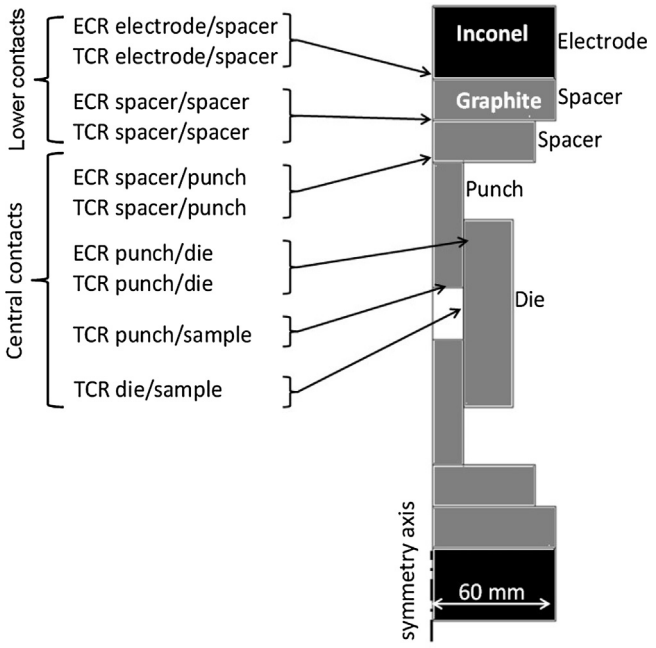

Fig. 1. Overview of the SPS column and the location of the ECR & TCR for the different inner contact interfaces.

Six calibration experiments were performed for in situ estimation of the ECR and TCR (Fig. 2). Three of them were devoted to the determination of the electrode/spacer and spacer/spacer contacts (also named lower contacts). All of these contacts are perpendicular to the applied load direction, then the resulting ECR and TCR can be related to the pressure exerted at each of these contacts. The geometrical configurations reported in Fig. 2a–c use simple graphite punches of 20, 30, 50 mm diameter placed between graphite spacers.

The three other experiments are devoted to the identification of the spacer/punch, punch/sample, sample/die and punch/die contacts (also named central contacts). The sample, die and punches geometries are reported in Fig. 2d–f are homothetically increased for punch diameters of 10, 20 and 30 mm. A constant pressure is applied during the whole cycle (including cooling), this allows verification of the TCR during the cooling step, a pure thermal stage of the cycle. A pressure of 100 MPa is applied for the calibration of the lower contacts. The aim of this high pressure is to minimize the TCR (punch/spacer) and it allows a strong heat flux to run through the lower contacts to be calibrated. The thermal cycle is a 100 K/min ramp up to 1000 °C and release of the current for the cooling stage.

Concerning the calibration of the central contacts, a constant pressure of 50 MPa is applied during the whole thermal cycle. This pressure is most useful for classical applications as it allows the densification of a wide range of materials without risk of die failure. The thermal cycle imposed at the die surface is the following: increase of the temperature from room temperature up to 1100 °C with a ramp of 100 K/min, a dwell of 1 min at 1100 °C and then the current is stopped for the cooling stage. For these configurations, the graphite felt that is classically added at the external die surface is not used here. This graphite felt decrease the die thermal radiative losses and then decrease the radial thermal gradient between the central column parts (punches, sample) and the edge of the die [12]. Even if this is beneficial for the sample homogeneity, for the calibration purpose, the impact of the vertical contact resistances on the temperature field is more distinguishable without the graphite felt. The higher is the ECR and TCR impact on the temperature field, the more accurate is the calibration of these ECR and TCR.

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