Spark plasma sintering novel tooling design: temperature uniformization during consolidation of silicon nitride powder

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Spark Plasma Sintering (SPS) of silicon nitride is affected by temperature non-uniformities within the powder compact, resulting in density and microstructure inhomogeneities. A double-pyrometer experimental setup reveals a temperature disparity of 100–200°C between the overheated outer surface of the die and the bottom of the upper punch, as a consequence of the electric current concentration through the die’s wall characterizing the SPS of non-conductive powders utilizing conventional SPS tooling. A novel tooling design, consisting in the tailored drilling of axial cylindrical or ring-shaped holes within the punch, is individuated and optimized through a campaign of fully-coupled thermal, electrical and mechanical finite element simulations. The analysis of the numerical results, experimentally assessed, allows for a comprehensive understanding of the phenomena underlying radial temperature distributions in SPS and leads to the development of a technological solution for the uniformization of temperature distribution.

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1. Introduction

Spark Plasma Sintering (SPS) technology, a particularly efficient process for the densification of powder compacts with the aid of electric current, has gained growing attention in the last decade.1,2) Interest has frequently shifted from the specimen to the entire tooling setup, usually constituted by graphite components, since experimental and numerical research has shown how significantly the overall tooling conditions affect the powder-specimen final properties, in terms of densification and microstructure.

Many efforts have consequently been dedicated to SPS process optimization. In this area, most of the conducted studies concern a progressive refinement of the sintering regime parameters—heating rate, holding time-step, preset temperatures, externally applied pressure, utilization of sintering aids—in order to gradually improve the densified specimen properties,3) enhance certain specific characteristics of the final product,4) or realize opportunely tuned functionally-graded materials.5) In certain cases, optimization resulted from the combination of SPS with other techniques in a tailored sequence,6) while other studies concerned more fundamental aspects, leading to the formulation or review of apposite theoretical models.7,8) A few coupled modeling with experimental verification,9–11) sometimes considering even more innovative current-assisted sintering techniques, such as high-voltage electric discharge consolidation, in whose context Grigoryev and Olevsky analyzed the behavior of interparticle contact zones.12,13)

Among the approximately 700 yearly publications addressing current-assisted sintering techniques, only a few works investigated on the role played by the tooling setup characteristics14) and on the relative possibility of optimization, treated with experimental approaches, numerical modeling or a combination of both. Such studies analyzed the consequences of modifying the constituent material,15–16) the geometrical design of the components,17–19) the manufacturing machine’s combination with characterization technologies,20,21) or the scalability of the process.22–23) The scalability aspect revealed the fundamental importance of understanding how thermo-electrical phenomena distribute within the powder specimen and the entire setup, since an increase in the characteristic dimensions unfailingly causes the development of augmenting internal temperature gradients, non-uniformities that so far have been assessed only for small sample diameters.24) Some more specific analyses were conducted in order to customize the die shape for the production of complexly shaped axial symmetrical parts25) and of functionally graded materials,26) evidencing how extensive and versatile SPS-tooling optimization procedures can be. More efforts are needed in order to attain satisfactory results in terms of reproducible production of bulk parts with homogeneous property distributions, suitable for industrial production.

When considering Spark Plasma Sintering or Field-Assisted Sintering Technique (FAST) equipment, certain distinctions have been individuated and defined, in the context of comprehensive reviews of the various methodologies and patents released since the introduction of these electrically-assisted densification technologies.27–30)

A setup optimization process needs to follow a thorough analysis of the conventional configurations’ conditions, in which the influencing factors can be pointed out, classified and investigated. The present work aimed at mitigating thermal gradients in the powder sample, therefore a preliminary study of the internal
temperature distributions was performed.

Because of the SPS device intrinsic difficulties in measuring the sample actual temperatures, after a few experimental approaches to the temperature distributions' reconstruction,[31,32] numerical tools have revealed to be particularly effective in compensating for this lack of experimental data.[30] Finite element models have been developed, capable to simulate the electrical and thermal evolutions of the specimen and tooling components during the SPS procedures.[33–38]

In order to accurately recreate the experimental conditions, fully-coupled models, combining electrical current, heat transfer, mechanics and densification kinetics, need to be implemented. A variety of studies have addressed this issue, providing indispensable information on the inherent mechanisms of such innovative densification techniques.[39–46]

Our study arose from an issue of raising temperature disparities when dealing with sintering on the macro-scale (specimen diameter of several cm, as an order of magnitude). Specifically, non-negligible thermal gradients were appearing in the specimen cross-section radial direction, which could be experimentally identified thanks to a double-pyrometer setup.

The powder employed was silicon nitride, a material that has raised increasing interest in the recent years, due to its multiple applications, from the bio-materials to the space engineering field. Several research studies have revealed the successful application of SPS to this material system,[47–49] some also investigating on the role played by the pulsed electric current during the powder consolidation process.[50]

In our investigation, silicon nitride was treated as an interesting source of experimental and numerical data for the analysis of radial temperature distributions in a macro-scale setup for the SPS of a non-conductive material, serving as input for the development of a novel tooling design, in which the punch geometry was optimized by drilling a specific array of holes, with geometrical parameters individuated by means of an extensive campaign of FEM simulations.

2. Experimental procedures

All experiments were performed with an FCT HPD25, a FAST/SPS device for the production of medium size samples, and a tooling-setup made of R7710 graphite (SGL Carbon SE, Wiesbaden, Germany), characterized by strength values of 250 MPa under compression and 80 MPa in bending.

The components constituting the graphite tooling were two 35 mm long punches, 62 mm in diameter, endowed with an axially-centered, 10 mm large and 30 mm deep circular hole, to allow for the temperature measurement; a 48 mm long and 10 mm thick die; two tapered spacers (also called heat protection plates), each 45 mm long, with an upper basis diameter of 62 mm at the interface with the punches, and a through hole of 10 mm diameter, again to consent temperature readings. At the interfaces between die, punches and specimen, a double layer of 0.25 mm thick graphite foil (Sigraflex, SGL Carbon SE, Germany) was inserted to ensure the stability of electro-mechanical contacts and to minimize the reactions of the powder compact with the graphite of the tooling. Such graphite paper has a substantial impact on temperature evolution, because of its anisotropic behavior, induced by the flake-like structure of the raw graphite material employed for the calendaring of the thin foil. The graphite paper’s in-plane electrical conductivity is two orders of magnitude higher than the through-plane one, and its properties are dependent on the applied pressure.[51]

The device was equipped with two optical pyrometers, providing for the temperature readings, from 400°C until the completion of the process, in two separate points of the setup. The axially mounted leading pyrometer A controlled the temperature profile set on the machine (operating in temperature control mode) and a radially adjusted pyrometer B was used to simultaneously measure the temperature at the outer surface of the die. Since the die was surrounded by a 10 mm thick, 80 mm long graphite felt thermal insulation (Sigratherm, SGL Carbon SE, Germany), aimed at minimizing heat radiation losses, a through circular hole of 7 mm diameter was produced into the felt, in order to get access to die surface and allow the corresponding temperature measurement. Note that the felt was chosen to be longer than the die (48 mm), with the objective of minimizing the additional radiation losses due to the exposed surfaces of the punches.

Figure 1 offers a representation of the complete tooling setup and of the pyrometers’ exact location.

The powder utilized was a silicon nitride-based composite of 96 wt% α-Si₃N₄ (Silzot HQ, AlzChem AG, Germany, 80% α-Si₃N₄ content, d₅₀~1.3 μm), with 2 wt% of Al₂O₃ (AKP50, Sumitomo Coal Mining Ltd., Tokyo, Japan) and Y₂O₃ (Grade C, Treibacher, Althofen, Austria), ground in isopropanol (planetary ball mill, 3 h, 300 rpm, agate furniture). The amount of additives was appositely chosen to be such that a significant but incomplete densification was attained, accompanied by a dissolution-diffusion-precipitation mechanism, leading to a transformation from low temperature trigonal alpha to high temperature hexagonal beta phase. Such phase transformation is regulated by the time and intensity of the exposure to high temperatures, and can be therefore utilized to estimate the local temperature distribution within the composite during consolidation.[52]

For each experiment, 50 grams of silicon nitride composite powder was utilized, loosely poured into the cavity of the SPS tooling. A heating rate of 100°C/min from 400 up to 1750°C was applied, followed by an isothermal dwell time (holding time step) of 5 min at 1750°C. The uniaxial pressure was increased from 8 to 50 MPa between 900 to 1000°C and was kept constant until the end of holding time. The sample was then freely cooled down to room temperature and cleaned by sand blasting to remove the adhering graphite foil.

Data about axial displacements, electrical power and temperature were collected and provided an important insight on the specimen’s evolution during the procedure. From the displace-
Fig. 2. Standard configuration temperatures. Experimental data show a high disparity between pyrometer A and B throughout the whole SPS process for the conventional tooling-setup.

The optimization of the punch geometry was performed by means of finite element simulations, operated in the COMSOL Multiphysics® environment, thanks to the software’s distinguishing capabilities of simultaneously combining a variety of physical processes. A three dimensional, fully coupled, electrical-thermal-mechanical model, capturing the essential physics of the SPS experimental procedures previously conducted, was developed. Thanks to the presence of the two temperature probes (pyrometer A and B) in the experimental setup, two sets of data were available for calibration. The model validation consisted in providing the software with the experimental current profile as input, and subsequently assessing that the temperatures observed were consistent with the experimental data for material and contact properties. In addition to this, it is important to remember that a numerical simulation will invariably introduce an error in the process reconstruction. The PID controller simulation will introduce an error, as well as the SPS process simulation itself. Thus, instead of inserting such two errors and moreover having to assess the coherence of the numerical PID-produced current profile with the experimental data, we chose to employ directly the experimental current data as input. We believe that the reliability of a modeling framework lies in guaranteeing the best possible matching with experiments as far as the conditions experienced by the SPS tooling and powder are concerned, more than in reproducing the operational mode of the SPS machine (temperature or current-control). By using the experimental current profile as an input, together with a whole set of experimental data for material and contact properties, we are ensuring the maximum possible adherence to reality. Once two separate sets of temperature data (profiles of pyrometers A and B) are found to be coincident between experiments and simulations, we can be confident that our modeling framework is reliable. Such confidence was further confirmed by the second set of experiments (§4.1), posterior to the initial model validation operated with the conventional punch geometry, and by additional experiments conducted in the broadening of this study.29

In each simulation the entire SPS tooling setup was reconstructed, comprehensive of powder compact specimen, punches, die and spacers, as represented in Fig. 1. The presence of the graphite foil, located at the specimen-punch, specimen-die and punch-die interfaces, was simulated through an appropriate distributed impedance contact condition, while the graphite felt thermal insulation was replicated with the corresponding boundary condition.

The FEM simulation of Spark Plasma Sintering procedures requires the coupled implementation of an electrical module, describing the current flowing through the SPS setup, together with a thermal one, responsible for heat transfer and Joule heating phenomena, a kinetic section, describing the application of external loads, the description of the interactions between components and the introduction of the powder compact constitutive behavior, and finally a mathematical section, reconstructing the densification kinetics by means of a user defined partial differential equation (PDE). The required equations are presented here. For the DC current distribution we use:

$\mathbf{J} = \sigma (-\nabla V)$  

where $\mathbf{J}$ (A/m²) is the current density, $\sigma$ (S/m) the electrical conductivity and $V$ (V) the voltage.

The heat transfer is given by:

$\rho_c C_p \frac{dT}{dt} = \nabla \cdot (k \nabla T) + Q$  

$Q = \mathbf{J} \cdot \mathbf{E}$

in which $C_p$ is the heat capacity [J/(kg °C)], $T$ (°C) the temperature, $k$ [W/(m°C)] the thermal conductivity, $Q$ the heat source term (W/m²), $\mathbf{E}$ the electric field (V/m), and the density $\rho_c$ (volumic mass, kg/m³) is expressed as a function of the theoretical density of the bulk material $\rho_{th}$ (kg/m³) and the
porosity $\theta$, volume fraction of voids in the domain, in the following form:

$$\rho_{\text{eff}} = \rho_0 \rho = \rho_0 (1 - \theta)$$  \hspace{1cm} (4)

The constitutive equation for the powder specimen has the form taken from the continuum theory of sintering, in which the behavior of the material was selected to follow the power law creep model:

$$\sigma_{ij} = A_n W^{m-1} \left[ \dot{\varepsilon}_{ij} + \left( \frac{\psi}{3} + \frac{1}{2} \dot{\varepsilon}_{kk} \delta_{ij} \right) \right] + P_L \delta_{ij}$$  \hspace{1cm} (5)

where $\sigma_{ij}$ (Pa) are the stress tensor components, $A_n$ (Pa·s$^m$) is the power law creep coefficient, related to the material’s activation energy ($Q$) and to temperature through an Arrhenius-type relationship, $W$ (s$^{-1}$) is the equivalent strain rate, $m$ the power law creep strain rate sensitivity exponent, $\dot{\varepsilon}_{ij}$ (1/s) the strain rate tensor components, $\psi$ the normalized shear viscosity, $\dot{\varepsilon}_{kk}$ the normalized bulk viscosity, $P_L$ (Pa) the sintering stress and $\delta_{ij}$ is the Kronecker delta.

The above-mentioned sintering parameters are given as functions of porosity as follows:

$$W = \sqrt{\frac{\psi^2 \dot{\varepsilon}_{kk}^2 + \psi \dot{\varepsilon}_{kk}^2}{1 - \theta}}$$  \hspace{1cm} (6)

$$\psi = (1 - \theta)^2$$  \hspace{1cm} (7)

$$\dot{\varepsilon}_{ij} = \frac{2}{3} (1 - \theta)^3$$  \hspace{1cm} (8)

$$P_L = \frac{3\alpha}{r_0} (1 - \theta)^2$$  \hspace{1cm} (9)

where $\dot{\varepsilon}$ (1/s) is the strain rate tensor deviator, $\alpha$ (1/m$^2$) the surface tension and $r_0$ (m) the particle radius.

The temperature dependencies of the various materials properties are reported in Table 1 for the R7710 graphite components and in Table 2 for the silicon nitride powder. The dependence on porosity of the specimen’s material data could not be univocally assessed and was therefore not included in the present simulations. The graphite data were provided by the producer (SGL Carbon SE, Wiesbaden, Germany), while for the silicon nitride case they were partly taken from the literature and partly experimentally measured. Specifically, differential scanning calorimetry was utilized for the heat capacity, while the thermal conductivity was found as the product of density, heat capacity and thermal diffusivity, which was in turn individuated by means of laser flash analysis. Electrical conductivity and power law creep parameters were taken from the literature.

The imposed boundary and initial conditions reflected the actual experimental framework employed during the SPS procedures, and they are listed according to the section in which they are defined.

In the electrical currents module, the initial voltage was set to zero, insulation of the outer tooling surfaces was included, the bottom surface of the lower spacer was grounded, the current input (extracted from the SPS machine readings during the experimental process) was applied to the top surface of the upper spacer, and the opportune electrical contact resistance was imbedded at the interfaces between the tooling components as a function of temperature and pressure, in the following form:

$$n \cdot J = \frac{\Delta V}{R_c}$$  \hspace{1cm} (10)

and

$$R_{c,\text{th}} = (4.15 - 0.21 \cdot \ln T) \left( \frac{19}{\rho} \right) \times 10^{-7} \Omega \cdot m^2$$  \hspace{1cm} (11)

$$R_{c,v} = (22.59 - 0.006 \cdot T)(1.15 - 0.008 \cdot p) \times 10^{-7} \Omega \cdot m^2$$  \hspace{1cm} (12)

where $n$ is the outward unit vector normal to the interface surface, $\Delta V$ (V) the voltage drop across the interface, $R_{c,\text{th}}$ and $R_{c,v}$ (mΩ·cm$^2$) the horizontal and vertical interface contact resistances, respectively, and $p$ (MPa) the pressure.

As for the heat transfer module, the initial temperature was set to be 25°C, the same constant temperature of 25°C was imposed at the bottom surface of the lower spacer and at the top surface of the upper one, corresponding to the cooling effect of the circulating water in the SPS machine, the outer surface of the die was thermally insulated, as the presence of the graphite felt imposes, ideal thermal contact between layers was implemented, since the role of thermal contact resistance was proven to be negligible, and a heat radiation boundary condition was imposed at the external surfaces, according to the Stefan-Boltzmann law, with a value of 0.8 for graphite emissivity. The perfect insulation
condition used to simulate the presence of the graphite felt is a first approximation that provided satisfactory results in this context, but can be perfected with a radiation at low emissivity for future purposes.

Concerning mechanics and densification kinetics, an initial porosity of 30% was introduced (in accordance with the experiments), the bottom surface of the lower spacer was fixed, the top surface of the upper spacer was subjected to an externally applied pressure equal to the load utilized in the experiments, and the contact between the different tooling components was ensured.

The first step of the FEM model construction consisted in the calibration of the model according to the available experimental data. The SPS machine provides readings of the parameters involved in the sintering process. The current profile was therefore extracted and used as the input for the FEM computations. The data fitting was evaluated from the temperature readings standpoint. As reported in Section 2, two optical pyrometers were utilized to obtain the temperature evolution during the processing time: pyrometer A, the leading one, focused on the lower point of the opposite hole in the top punch, and pyrometer B, aiming at a mid-point of the die outer surface. Two sets of point-temperature readings were therefore available to be employed in the verification of the FEM outcomes, since in COMSOL® we were able to monitor this parameter’s evolution with time in the same two locations. By slightly calibrating the contact resistances, a good fitting between experimental data and numerical results was obtained, as shown in Fig. 3.

Once such agreement had been reached, a modeling-based optimization of the tooling was performed by modifying the punch geometry. The conventional configuration was substituted by alternative solutions, consisting in the drilling of holes according to specific patterns, an alteration that was allowed by the compressive strength of the SPS setup graphite. Two main families of novel punch geometries were identified, one provided with an annular array of axial cylindrical holes, the other with a variable number of concentric ring-shaped holes, both represented in Fig. 4. From this point on, we will refer to this first modified configuration as “Holes”, to the second as “Rings”, while the conventional one will be addressed as “Full”.

For both the Holes and Rings cases, several combinations of the main geometrical parameters were investigated. Several variations were created, by changing the values of the dimensions marked as D and H in the figure. H is the height of the holes, while D is their diameter in the Holes configuration, and the series of concentric rings diameters in the Rings case.

The following values were applied, in all their possible combinations, for a total of 24 case-studies. A few others were later considered, in the very final stage of the optimization process.

Holes:
- D: 8, 10, 12, 14 mm
- H: 10, 20, 30 mm

Rings:
- 1 Ring
  - D: 24–45 mm
  - H: 10, 20, 30 mm
- 2 Rings
  - D: 20–30–40–50 mm, 18–32–38–52 mm
  - H: 10, 20, 30 mm
- 3 Rings
  - D: 18–25–32–39–46–53 mm
  - H: 10, 20, 30 mm

For each simulation, the sintering route followed what has been applied in the experimental framework. The new outer surfaces resulting from the presence of the holes in the punch were provided with the same surface radiation boundary condition introduced in the thermal module for the external surfaces of the whole tooling setup.

Several attempts have been performed in order to stabilize the simulations outcomes. A free tetrahedral mesh was finally selected, with a number of “normal”- or “fine”-sized elements ranging between 12,000 and 25,000, automatically generated by the software. Such definitions of the mesh sizes are reported as denominated in the software itself, which uses a qualitative indication of the mesh refinement grade that spans from “extra-coarse” to “extra-fine”.

4. Results

4.1 Experimental optimization

The first three simulations run—Full, Holes with D = 12 mm and H = 20 mm, Rings with D: 20–30–40–50 mm and H = 20 mm—offered promising results. In accordance with what was available from experimental procedures, i.e. the temperature readings in correspondence to the two points A and B (named after the relative pyrometers), the parameter selected for comparison between the different geometries was the temperature disparity $\Delta T = T(B) - T(A)$. Such disparity appeared lowered, during the holding time, from an average value of 120°C in the Full case, to 80°C for the Holes, to even 15°C for the Rings, a tendency that was confirmed by means of a second set of experiments, in which the conventional Full punch was replaced.
by the two abovementioned optimized geometries (a photograph of the modified punches is given in Fig. 5). A representation of the successful application of the new configurations is offered in Fig. 6, where the $\Delta T$ evolution with time for the three cases experimentally implemented is plotted together with the temperature profile corresponding to the imposed sintering regime.

From this second set of experiments one can also see that during the heating ramp $\Delta T$ slowly increases with time, nevertheless staying into a relatively low range of 25–35°C (corresponding to $T(A) = 600$ and 900°C, respectively), but that it suddenly raises when the applied pressure reaches its maximum value of 50 MPa. The most interesting time step, though, is the isothermal dwelling, at the beginning of which the peak of 200°C temperature disparity for the conventional Full case is successfully lowered down to 64°C in the Holes configuration and 32°C for the Rings one. This beneficial tendency appears stabilized with time, since after 2 min of holding a $\Delta T$ of 115, 54 and 13°C is seen in the Full, Holes and Rings setup, respectively. The agreement between experiments and modeling was confirmed to be satisfactory, with a maximum discrepancy of $\pm 25°C$ reached in the Holes case at certain stages of the holding time.

It’s worth noticing that the punch geometry results do not have any significant impact on the monitored densification of the sample (the variations in final density are below 5%, and even lower during the heating ramp), whereas the electrical power shows some differences among the Full, Holes and Rings configurations experimentally analyzed, as expectable when changing the cross-sectional area through which the current can flow. Table 3 compares the cross-section surface values, absolute and relative, for the three considered setups, together with the corresponding final density of the specimen.

The samples’ density values reported in Table 3 were obtained through the Archimedes’ method. The specimens were then cut to analyze the alpha silicon nitride content by means of XRD (CuK$_\alpha$) measurements (Fig. 7), and determine the local density distribution (Fig. 8).

The alpha content quantitative analysis was performed with Autoquan (GE Inspection Technologies, Fairfield, CT) and according to the Rietveld’s method, underpinning the results of temperature measurement. For all the three configurations the alpha content, acting as an indicator for the local temperatures history (the lower the alpha amount, the longer the exposure to local overheating was), is in the range of 40% at specimen center, which is consistent with the temperature data retrieved from the controlling pyrometer A, located very close to this position. With increasing distance from the sample central area, the values of alpha Si$_3$N$_4$ content show a decrease in every configuration, but the most significant reduction is shown in the Full case, evidencing the strong impact of temperature gradients on the final

**Table 3. Cross-section areas and final sample densities. Data for the setups utilized during experiments. A decrease in cross-section area leads to a slightly lower final density**

| Configuration | $A_{cm^2}$ | $A_{rel}$ | Density $g/cm^3$ |
|---------------|------------|-----------|-----------------|
| Full          | 29.11      | 100.00    | 3.06            |
| Holes         | 20.07      | 68.92     | 2.91            |
| Rings         | 18.12      | 62.23     | 2.89            |

**Fig. 5.** Alternative punch designs. The photograph shows the experimentally implemented Holes (left) and Rings (right) punch configurations.

**Fig. 6.** Temperature gradients in the modified setups. Experimental data on temperature disparities in the alternative configurations confirm the gradients mitigation anticipated by FEM modeling.

**Fig. 7.** Alpha-Si$_3$N$_4$ content as a function of the distance from the sample’s center. The radial decrease of the alpha phase is a consequence of the higher temperatures reached at the specimen’s edge.

**Fig. 8.** Specimen density as a function of the distance from the sample’s center. The density radial distribution provides an additional confirmation of the presence of thermal gradients throughout the sample.
outcome microstructure and local properties (trend confirmed by density measurements), while the Holes and Rings cases succeeded in lowering these disparities.

Together with the XRD technique, the local microstructure evolution has been analyzed by Field Emission Scanning Electron Microscopy (FESEM) with a Zeiss Ultra 55 for all the three configurations here implemented. FESEM specimens have been obtained from the sample’s edge and central area. They have been subsequently ground and observed with an energy and angle selective backscattered electrons detector (BSE), which guarantees a good representation of silicon nitride thanks to the selective contrast between the main Si$_3$N$_4$ material and oxide additive phase. The relative images are shown in Fig. 9.

Besides the still present porosity in this appositely non-completely densified material, for all the three configurations the sample central area shows larger grains, belonging to alpha phase, whereas the smaller elongated or hexagonal-shaped grains were the results of the dissolution-diffusion-precipitation mechanism leading to beta phase formation. The latter are located at oxide-phase rich areas (bright contrast region). The XRD analysis revealed that the amount of beta Si$_3$N$_4$ phase located at the sample center was not being altered when changing configuration.

In the Full configuration case, a 100% amount of beta phase has been detected in the proximities of the edge, namely the area in which the highest local temperatures were located. Only for this configuration the micrographs reveal that a significant grain growth took place, by means of an Ostwald ripening process, while for the Holes and Rings configuration no significant difference in grain size was detected when moving along the radial direction. These microstructural characterization results are coherent with XRD measurement.

The slight gradients in density along the radial direction, which in the Rings case seem to show an opposite trend, were considered of negligible entity with respect to the phases content and grain size inhomogeneities. The density variations are in a range of 3–5% (Fig. 8), opposed to the 25–30% difference in the alpha-phase content (Fig. 7) and to a significant grain growth at the edge for the Full case (Fig. 9).

The obtained combination of effects, namely an improvement in temperature and properties distributions accompanied by minor modifications of the power output and final overall density when utilizing the modified punches, stimulated a broadening of our study. Room appeared to be left for further geometry optimization, aimed at both finding an ideal technological solution to the
thermal gradients issue, and answering a series of interrogatives arisen in view of the phenomena described in this section—such as how current density and temperature non-uniformities are correlated, what material characteristics have a stronger impact on heat transfer, how temperature distributes inside the whole cross-section of the specimen, what are the mechanical strength limitations to the punch geometry alterations.

4.2 Further optimization

In view of such outcomes, a second series of FEM simulations was run, this time implementing all the remaining Holes and Rings options listed in Section 3. This broader part of the study allowed a refining of the geometry optimization process and a deeper understanding of the phenomena underlying the distribution of temperatures in the SPS tooling setup, together with an assessment of the impact of each of the parameters playing a role in the uniformization of temperatures, focusing in particular on the radial direction.

The modeling results are reported in Figs. 10 and 11. For the purpose of comparison with experiments, both figures depict the time evolution of the same parameter $\Delta T$, namely the difference between the temperatures at points B and A, where pyrometers were located. Notice that we focused on the holding time step, neglecting the heating ramp. Such a choice was based on the relevance of the holding with respect to the previous heating ramp, in terms of temperature stabilization and specimen densification.

The denominations for the Holes case are based on the holes diameter D, while the Rings case allows a qualitative classification, based on the number of concentric ring-shaped holes. In the 2 Rings case, two combinations of the D parameter values were attempted, therefore they are distinguished by addressing one as “2 Small Rings” (D: 20–30–40–50 mm) and the other one as “2 Large Rings” (D: 18–32–38–52 mm). Note that this second punch design is the one in which most of the material is removed, even more than in the 3 Rings case.

The graphs are given following an order of decreasing cross-section area (larger amount of removed material) for both Holes and Rings, but in the latter case the first two configurations, 1 Ring and 2 Small Rings, present an equal value, meaning that the current density flowing through the interface is the same in the two cases, which therefore differ only for the current distribution inside the punches. By current distribution we denote the path followed by the current within the punches space.

Observing these plots, one can immediately infer how a more homogeneous current distribution has a significant beneficial effect on the lowering of the temperature disparities, while current density through the radial cross-section area doesn’t seem to play an important role. The first statement is a consequence of the comparison between the two upper plots in the Rings configuration: as mentioned above, 1 Ring and 2 Small Rings present the same cross-section area, but $\Delta T$ results to be higher in the former case. The impact of current density deserves a more careful analysis, which will be enounced later. Another immediate observation can be drawn with regards to the effect of a change in the holes’ depth, since in all cases an increase in H leads to a decrease in the temperature gradients.

A peak at the beginning of the holding time step appears in each plot. It is due to the current profile, given as an input and presenting the same kind of spike in correspondence to the end of the heating ramp. Such phenomenon is therefore unavoidable, since the reliability and repeatability of our computations required the same experimental electrical current input in every FEM simulation, and also because it was proven, both experimentally and numerically, how this trend was responsible for the preset temperature stabilization during the holding time step. A decreasing of this effect is, however, attainable by lowering the holes depth dimensions: in all the situations considered, an increase in H corresponds to an increase in the initial peak.

Figure 12 offers a summarized version of the previous graphs. A specific time instant was chosen, $t = 1200$ s, corresponding to mid-holding, in order to compare the most significant temper-
ature measurement for every implemented geometry. Here the most successful approach clearly appears to be the Rings, with its values of $\Delta T$ reaching values of 10°C, or even negative ones, suggesting interesting hints for a further improvement of the punch geometry, eventually able to eliminate $\Delta T$ completely, while the Holes setup does not provide any decrease in $\Delta T$ higher than 40°C with respect to the Full-punch case. The impact of current density, current distribution and holes depth described above is thereby confirmed.

From this same figure, we infer how an additional slight change in the drilled holes configuration, in particular of their height, which is the parameter that appears to have the most immediate effect on temperature gradients within the SPS setup, could lead to even better results. By making a direct comparison between the three most promising designs (see Fig. 13), namely 2 Small Rings with $H = 30$ mm, 2 Large Rings with $H = 20$ mm and 3 Rings with $H = 30$ mm, circled in the upper part of the same figure, one can select the configuration which is more prone to a finalization of the geometry optimization. Aiming at decreasing both the initial temperature peak and the current distribution inhomogeneities, the most suitable setup results to be the 3 Rings one, as the lower part of the same Fig. 13 ($\Delta T$ vs. holding time) reveals.

The final stages of the punch redesign consist in a trial-and-error procedure for calibrating both the $H$ value and the current input. As stated before, up to this step of our study, the applied current needed to be equal in every case study, for comparison purposes. At this point, once the optimal configuration has been selected and requires only minor adjustments, the verification of the temperature evolution relative to pyrometer A can show that such parameter does not follow the profile imposed by the SPS machine during experimental procedures. The conclusive outcome of this novel configuration, by giving the plots of the radial temperature distribution in specimen’s central cross-section for the original Full-punch setup and the present modified Rings case. The use of the same color scale for the legend shows clearly how strong the impact of the tailored holes is.

Fig. 11. Temperature disparity evolution for the Rings configurations. $\Delta T$ vs. holding time is plotted as a function of the Rings configuration’s geometrical parameters.

Fig. 12. Summary of temperature disparities. $\Delta T$ at $t = 1200$ s (mid-holding) is considered for a comparison of all the simulated configurations.

Fig. 13. Temperature disparity evolution for the Rings configurations. $\Delta T$ vs. holding time is plotted as a function of the Rings configuration’s geometrical parameters.
Irradiated heat and temperature disparity between thermal source explained by looking at the Stefan-Boltzmann equation relating environment kept at room temperature. This is quantitatively corresponds to a more extensive amount of material exposed to an increase in the area of the outer surfaces of the tooling corresponding to an increase in the overall outer boundary surface area and to a decrease in thermal mass, both beneficial for a lowering of thermal gradients, but, at the same time, we have a detrimental effect of increased current density, which is an immediate consequence of the drilling of holes, several simultaneous effects: a reduction of this area, indeed, from the considerations listed above, one can notice how, even though these holes act as closed cells not in immediate connection with the environment kept at room temperature, they are sufficient to mitigate localized overheating, probably thanks to the emissivity properties of graphite. This conclusion has been assessed by running some simulations with and without the opposite boundary condition. The temperatures obtained showed variable differences, typically of several tens of degrees centigrade. Higher temperatures were reached in the case of absence of surface radiation, with consequent changes in the disparities among the two pyrometers.

Current distribution, as introduced before, influences thermal gradients by lowering the non-uniformities when directed in such a way to result in more spatially homogeneous temperature distributions. Here we strictly refer to the Rings configuration, which, indeed, is also the one that showed the most promising results and a series of possibilities of fine tuning and further optimization. Spatial homogeneity of the current distribution translates in the choice of a pattern of ring-shaped holes consisting of a higher number of thin holes (in the radial direction), instead of a single large one that drastically separates the current flow into two branches running far from each other, one close to the center and one to the edge of the component.

Current density through the punch’s radial cross-section deserves a more careful analysis. In the plots reported in Figs. 10–12, at a first glance, a change in the current flow density, consequence of an alteration of the cross-section area, seems not to have a significant effect on temperature disparities. Nevertheless, from the considerations listed above, one can notice how a modification of the punch radial cross-section area leads to several simultaneous effects: a reduction of this area, indeed, which is an immediate consequence of the drilling of holes, corresponds to an increase in the overall outer boundary surface area and to a decrease in thermal mass, both beneficial for a lowering of thermal gradients, but, at the same time, we have a detrimental effect of increased current density, to which localized Joule heating phenomena are proportional. This enhancement of the Joule effects would tend to lead to local peaks of temperatures, but the augmented surface radiation and the diminished heat capacity hamper the verification of such thermal gradients. Therefore it would be incorrect to address radial cross-section

5. Discussion

From the obtained results several conclusions can be drawn concerning the identification and impact of the phenomena influencing temperature non-uniformities within a Spark Plasma Sintering tooling setup.

The main factors affecting the temperature gradients resulted to be heat capacity, surface radiation, current distribution and current density.

The heat capacity of a certain component can be qualitatively defined as its “thermal mass”, the heating storage capability of such domain, whose diminution revealed to be an efficient method to mitigate the thermal inhomogeneities within the entire tooling.

Surface radiation has been proven to act efficiently when temperature peaks manifest during SPS procedures, since an increase in the area of the outer surfaces of the tooling corresponds to a more extensive amount of material exposed to an environment kept at room temperature. This is quantitatively explained by looking at the Stefan-Boltzmann equation relating irradiated heat and temperature disparity between thermal source and destination, in which temperatures appear at the fourth power. By drilling holes inside the punch, we have automatically increased the amount of the tooling setup’s external surfaces radiating, and therefore releasing, heat. It is interesting to notice how, even though these holes act as closed cells not in immediate connection with the environment kept at room temperature, they are sufficient to mitigate localized overheating, probably thanks to the emissivity properties of graphite. This conclusion has been assessed by running some simulations with and without the opposite boundary condition. The temperatures obtained showed variable differences, typically of several tens of degrees centigrade. Higher temperatures were reached in the case of absence of surface radiation, with consequent changes in the disparities among the two pyrometers.

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area modifications, or, alternatively, current density changes, as ineffective or negligible, since we are instead having a combination of three different effects that end up compensating each other.

Consequently, a comprehensive punch geometry optimization procedure can be operated on different levels, individually or simultaneously implemented. A holes height increase has two advantageous consequences, namely an augment in boundary surface and a diminution of extra thermal mass, without presenting the downside of excessive current density localization. A more homogeneous current flow throughout the punch can be created by providing for the selected material’s removal in the form of an increased number of thinner concentric ring-shaped holes. The cross-section area parameter will, on the other side, need to be cautiously tuned in order to obtain desirable outcomes. Notice how all the redesign procedures have been based on the Rings configuration, thanks to both its versatility and its ability to achieve consistently improved results with respect to the Holes setup, by removing equivalent quantities of graphite.

At this point it is important to point out that our results have been experimentally validated based on the temperatures recorded by the two pyrometers, while a prosecution of the study is needed in order to obtain a completely satisfactory correspondence for the porosity distribution. In the experimental procedures section we explained that oxides had been added to the powder specifically to hamper densification and therefore individuate more clearly the radial thermal gradients patterns based on the alpha and beta phases-content, the effects of which are not immediate to reconstruct in numerical simulations. Interestingly, experiments revealed that the evidence of thermal gradients lied significantly more in the alpha/beta phases and grain size distributions (Figs. 7 and 9 of the manuscript) than in the density distribution (Fig. 8). The multiple experimental evidences on the temperatures level were more than sufficient to consider the novel punch design as a successful strategy when radial thermal gradients constitute an issue. Such results can be used as qualitative guidelines for the optimization of SPS tooling setups.

6. Conclusions

A radial temperature inhomogeneity issue in Spark Plasma Sintering of silicon nitride was addressed through a geometric optimization of the tooling design. A coupled experimental and numerical approach led to the development of a novel punch configuration, realized by drilling an array of circular holes or a series of concentric ring-shaped channels in the conventional graphite setup. The successful application of such modifications was experimentally assessed, while finite element simulations were implemented to further refine the outcomes, to obtain a full map of the temperature distribution in the specimen’s cross-section, and to investigate on the mechanisms influencing the thermal gradients development. It was shown that the removal of material from the punch results in a beneficial combination of heat capacity reduction and increase in the free surface area. A configuration ensuring a more homogeneous current distribution is also desirable, while a decrease in the cross-section area perpendicular to the current flow reflects in a combination of the abovementioned advantageous graphite removal with a detrimental effect of rising current density, therefore resulting in a negligible global effect.

The final optimized configuration consisted in a punch with three thin concentric ring-shaped holes (channels), able to substantially reduce internal temperature disparities, and offering interesting possibilities for further improvements, some of which investigated in Ref. 57, in terms of control of the intensity and path followed by the electric current flow.

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