Modelling of Micro/Macro Densification Phenomena of Cu Powder during Capacitor Discharge Sintering

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Abstract: Capacitor Discharge Sintering (CDS) is an ultrafast Electric Current Assisted Sintering method (u-ECAS) suited for electrically conductive powders. It is characterized by relatively short processing times (milliseconds range) and much lower sintering temperatures than the melting point of the powders. However, the CDS basic phenomena are not fully understood yet, neither at the macroscale (i.e. equipment scale), nor at the microscale (i.e. particle scale). Both the short processing time and the closed-die system pose severe difficulties in experimental monitoring.

Computer modeling, on the other hand, offers an effective mean to design new experiments and to overcome experimental difficulties, thereby enhancing the fundamental understanding of the CDS process. From a modeling viewpoint, the CDS model introduces a new class of non linear problems, which involves not only a complex bulk multiphysics, but also an intricate contact multiphysics. Both are intimately coupled across the spatial scale of the system, from the microscopic to the macroscopic level.

This work presents a micro/macro CDS model based on the Comsol Multiphysics package applied to copper powder. The overall CDS model couples the relevant partial differential equations governing the electrical, thermal and stress-strain fields with densification kinetic equation.

Keywords: CDS, capacitor discharge sintering, ultrafast ECAS, current assisted sintering, multiphysics, stress-strain, copper powder.

1. Introduction

Capacitor Discharge Sintering (CDS) is a novel, clean and energetically efficient method to rapidly and fully consolidate electrically conductive materials under relatively high pressures by using one single high intensity current pulse. The electric pulse is discharged by a capacitor bank in milliseconds [1]. Typically, green bodies or loose powders are uniaxially pressed using a confining die and two punches which also work as electrodes. The used CDS apparatus may generate current peaks of up to $5 \cdot 10^5$ A or current density of 10^8 A/m^2[2].

CDS is commonly operated in air. Short processing times help prevent undesired powder oxidation and reduce thermal radiation losses. Grain coarsening, which is common in most conventional sintering methods, is limited or even negligible in CDS. In fact, CDS is proved to be particularly suited for the densification of nanostructured powders [2], although a wide range of conventional (i.e. microcrystalline) metals, alloys and cermets can be also successfully sintered with a single pulse. These features confer to CDSed components unexpectedly enhanced properties compared to counterparts sintered with conventional processes.

However, the intricate CDS multiphysics over both the macroscale and the microscale is still veiled. The high speed of the process and the closed-die apparatus make on-line process monitoring very difficult. Experiments show that the current path across the punch/powder/die system is more sensitive to contact resistances than to bulk resistances. This is true both at the microscopic level, i.e. between particles, and at the macroscopic level. The latter involves the contact between the punch, the die and the powders. The initial (green) density, the particle size and the chemical surface conditions of the powders are of paramount importance to define the transient current flow and the corresponding heating and sintering of the compact. It is found that the powder density steeply changes during CDS.

Densification kinetic, indeed, is a complex function of imposed current and pressure. Both process parameters act on the sintering microstructure through a complex balance of electric, thermal and stress/strain fields. These interact microscopically and macroscopically, in the bulk as well as along the contact interfaces of the CDS constituent parts. Joule heating starts at particle level and rapidly propagates by conduction to macroscopic parts of the assembly. Heating involves stress-strain phenomena due to thermal differential dilatation and shrinkage phenomena which, in turn, non-linearly affect the material properties of the powder.

The purpose of this work is to physically model the complex multiphysics of CDS and to apply it in the case of a copper powder using the Comsol Multiphysics package.

2. Physical Phenomena in CDS

As said above, it is the precise balance of bulk and contact multiphysics phenomena at the micro- and macro-scale that determines the sintering of conductive powder by CDS. The accurate prediction of individual phenomena is crucial to evaluate the overall transient of the process and to reliably predict the microstructure and properties of the sintered material.

Essentially, the sintering apparatus includes a graphite die, two copper electrodes (punches), and the powder compact (Fig. 1). Experimental observations suggest that the typically short consolidation times of sintering by CDS cannot be solely explained by diffusion phenomena. Before the actual sintering cycle, the copper powders are pre-compacted using a relatively high pressure. Subsequently, they are simultaneously subjected to the same loading force and a current pulse. The relatively high imposed pressure and the overall softening of the particles, due to the decrease of the yielding stress with temperature, are considered to be the main responsible for copper densification during CDS, resulting in the plastic deformation of the particles, especially at the softened contact interfaces.

As in most ECAS [1] methods, porosity significantly affects the electrical resistivity of the powder compacts. Even when copper punches are used, copper powders dissipate significantly more heat than punches or die. This results in a steep increase in the powder temperature during the early stage of the densification process. The bulk and contact properties of the powders are also affected by the temperature field. In general, other than electrical conductivity and yield stress, also thermal conductivity and specific heat are assumed to be functions of temperature and porosity.

In general, powder densification occurs by material flow along the particle interfaces or by mechanical collapse of particles. As the sintering temperature can be relatively high, though always much lower than the melting point of the powder, primary densification may be enhanced by creep flow. Depending on surface contamination or the presence of insulating oxide films, the contact conductivity (electric and thermal) may be unsuitable for the desired current flow. In the presence of such insulating interface films, the efficiency of CDS sintering is severely decreased.

As the powder shrinks, the electrodes and other large, electrically connected constituent

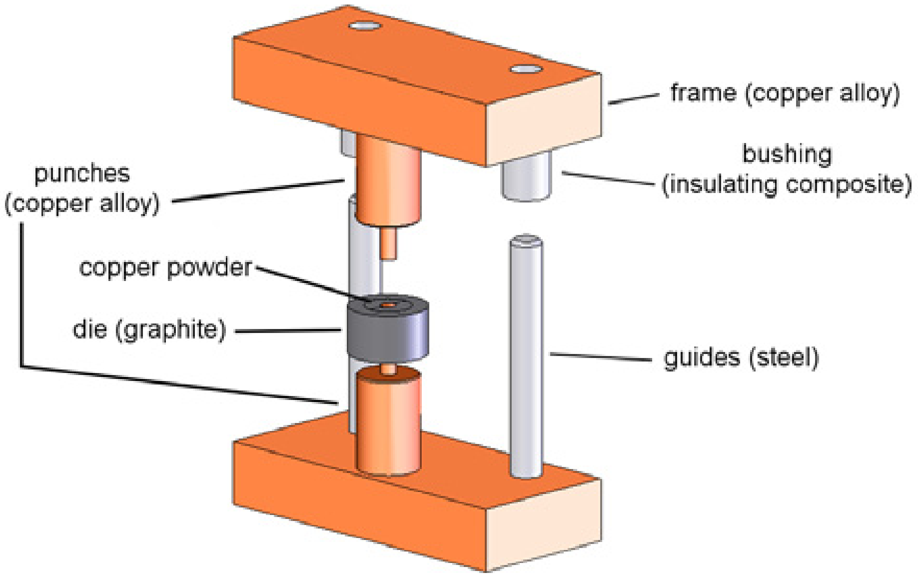

Figure 1. Schematic of the CDS assembly.

blocks function as formidable heat sinks. Consequently, the powder is rapidly quenched in less than one second by conduction. This rapid quenching hinders grain growth, which is a typical problem in most conventional sintering processes. As a result, the final CDS microstructure closely resembles the initial powders.

3. The CDS Model

The model has to couple the electro-thermo-mechanical behavior both at the macroscopic level of the machine and at the microscopic level of the sintering particles. The different models are illustrated below. The overall scheme of the developed model is shown in Fig. 2.

3.1 The macroscopic models

Due to the symmetry of the CDS geometry only a quarter of it requires to be solved. A 2D axial-symmetry is considered in the calculations. The powder compact is assumed to be a continuous medium at the macroscopic level, although its properties vary element-by-element according to the microscopic model. The CDS model is driven by an imposed DC pulse of specified current intensity and shape. The current is carried to the powder by the two electrodes. Upon discharge, the powders constitute the major region of dissipation, due to their very high initial resistance.

As the imposed current pulse (see Figure 3) is time dependent, the Poisson equation can be integrated across the system by treating the time as an external parameter rather than as an independent variable [*Conductive media DC*, 3]. This parameter ranges over the whole thermal cycle, from 0 to 0.1 s. For the implementation, the current-time profile is entered as a current density at the upper electrode end boundary. The macroscopic electric conduction across each system domain is controlled by the local electric conductivity, which is temperature dependent and, in the case of the powder domain, also function of porosity.

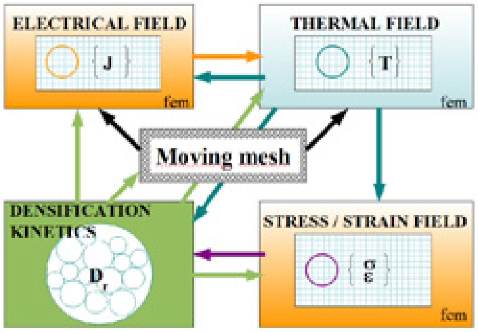

Figure 2. Scheme of the micro/macro multiphysics and moving mesh coupling taken into account in the CDS model.

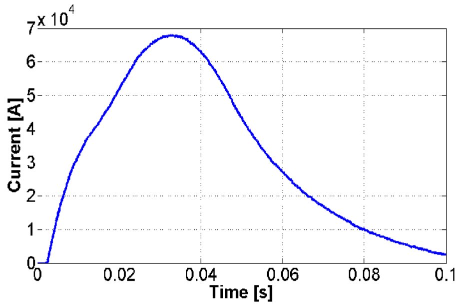

Figure 3. Profile of the experimental current pulse.

The major portion of the applied current pulse is converted into heat inside the powder. A minor portion of it is dissipated inside the constituent parts of the CDS system. The heat, once generated, is rapidly extracted from the powder via thermal conduction through the electrodes and all other copper constituent parts. Heat transfer through the CDS system is described by the standard transient heat conduction partial differential equation [*Heat transfer by conduction*, 3]. This equation includes the Joule heat source term dissipated in each conductive region of the system, as calculated by the electrical model. At each "thermal" time step, the Poisson equation is integrated in space taking the current value of the current-time profile as initial condition.

The thermal behavior in each region of the system is controlled by the local thermophysical properties (i.e. thermal conductivity, specific heat) which are function of temperature and, in the case of powder, also function of porosity (or relative density). Due to the rapid thermal cycling in CDS, the constituent parts made of copper alloy and the powder are assumed to be mainly cooled by heat conduction. The temperature of the support elements stays near room temperature throughout the CDS cycle. Heat losses by convection and radiation are reasonably assumed negligible.

The mechanical model predicts the stress-strain field in the compact [*Axial symmetry, Stress-strain*, 4]. For each time step, the stress-strain field is updated to account for the shrinkage and the change of material properties. It assumes the powder to behave as isotropic elasto-plastic medium. The yield stress of the copper powder is assumed to be temperature and porosity dependent. The imposed mechanical pressure (200 MPa) is assumed constant in time. It is applied at the upper electrode end boundary.

During CDS, the powder shrinkage alters the geometry of the system. To handle the inherently large powder displacements, a *moving mesh* technique is invoked. This is a built-in function in Comsol, which is called at each time step. It allows the powder region to homogeneously shrink of the fractional densification displacement computed by the microscopic model in the current time step. In addition, it updates the upper punch position by displacing it downward of the same fractional densification displacement. Accordingly, it recalculates the internal nodal points positions inside the punch and powder regions. This assumes an actual requirement where the sliding speed of the punches equals the shrinkage rate in the compact. The fractional macroscopic shrinkage is averaged over the whole powder region from the relative density field computed with the aid of the microscopic model.

In summary, the electric model is intrinsically coupled to the thermal model by the heat source term and the material properties. The thermal model computes the real transient across the powder and the constituent parts. At each thermal time step defined by the thermal field, the steady state electric model solves Poisson's equation with the imposed current corresponding to the defined time step. Convection and radiation losses in air can be reasonably ignored.

3.2 The microscopic model

The original work of Arzt et al. [5] for HIP process is adapted here for predicting copper powder densification during CDS. It includes three basic mechanisms, namely, plastic deformation, power-law creep and grain boundary diffusion from particle interfaces to pore surfaces. The model assumes a random packing of initially spherical, homogeneously sized contacting particles. The increase in the overall contact surface area between particles at each time step determines the fractional densification.

For a random packing of spheres the average contact force *f* is given by [5]:

$$f = \frac{4\pi R_{p0}^2}{Z D_r} p \qquad [1]$$

where p is the external pressure, R_{p0} is the initial particle radius, Z is the average number of contacts per particle and D_r is the relative density of the powders. The resulting effective pressure acting over each particle contact (including as additional driving forces the surface tension and the pressure of residual gas trapped in the pores) is:

$$p_{eff} = \frac{f}{a} + p_s - p_g \qquad [2]$$

where

$$p_s = \gamma\left(\frac{1}{\rho} - \frac{1}{x}\right), \quad \rho = \frac{x^2}{2(R_{p0} - x)} \qquad [3]$$

and

$$p_g = p_0 \frac{(1 - D_{rc}) D_r}{(1 - D_r) D_{rc}}$$

where a is the surface of a single contact area, γ is the surface energy, x is the radius of a contact area, p_0 is the outgassing pressure and $D_{rc} = 0.9$ is the relative density at which only closed porosity remains in the compact.

Densification is assumed to be instantaneous, provided that the applied pressure is sufficient to cause plastic flow in the compact; where the contact areas have grown and the yield stress is no longer exceeded, densification by plastic flow ceases. The common indentation law is adopted as yield criterion:

$$\frac{f}{a} = 3\sigma_y \qquad [4]$$

Densification proceeds by creep process after yielding at the contact areas ceases to operate. A simple power-law creep mechanism is given by:

$$\dot{\varepsilon} = \dot{\varepsilon}_0 \left(\frac{\sigma}{\sigma_0}\right)^n$$

where the parameters ε_0, σ_0 and n are material properties and σ is the equivalent (von Mises) stress computed by the macro-mechanical model. For the considered spherical particles geometry, this can be translated to a realistic creep densification rate using the following approximate equation:

$$\dot{D}_{rcr} = 5.3\, x \frac{(D_r^2 D_{r0})^{1/3}}{R_{p0}} \dot{\varepsilon}_0 \left(\frac{p_{eff}}{3\sigma_0}\right)^n \qquad [5]$$

In parallel to creep effects, sintering is assumed to be promoted by diffusion mechanisms. Matter is assumed to flow from the contact regions to the free surfaces of the particles mainly by grain-boundary diffusion. The rate of mass transfer along grain boundaries is believed to depend on two fundamental contributions: a) dislocation density generated by the severe plastic deformation, especially at particle interfaces; b) high contact temperatures developed during CDS. These two contributions strongly depend on the initial particle size R_{p0} and the number of contacts as follows:

$$\dot{D}_{rd} = \frac{12\, D_r^2 \left(\delta D_b + \rho D_v\right)}{D_{r0} R_{p0}^3\, g\left(D_r\right) kT}\, \Omega\, Z\, p_{eff} \qquad [6]$$

where $g(D_r)$ is a purely geometric term, δD_b is the boundary diffusion coefficient times the grain boundary thickness, D_v is the lattice diffusion coefficient, ρ is defined in Eq. [3], k is Boltzmann's constant, Ω is the volume of the diffusing atom and p_{eff} is the effective pressure given by Eq. [2].

The microscopic densification model is expressed in the form of a first order ODE, its coefficients changing in time as functions of the thermal and stress fields:

$$\frac{dD_r}{dt} = \dot{D}_{ry} + \dot{D}_{rcr} + \dot{D}_{rd} \qquad [7]$$

This kinetic describes the powder shrinkage behavior at the macroscopic (finite element) level. The ODE can be formulated using the *PDE mode, coefficient form* of Comsol Multiphysics [3]. The initial relative density (or porosity) is a very sensitive parameter, which significantly affects the CDS transient through the change of compact material properties. The model is solved assuming 0.45 as initial density according to experiments.

3.3 Material properties

The material properties of pure copper (powder) and of the constituent parts (copper alloys) are function of temperature, taken from a property database [6]. Thermal and electrical conductivity of the powder are both related to the particle contact areas and therefore are assumed to be functions of the relative density according to the following equation:

$$k_{contact} = \sqrt{\frac{1 - D_{r0}}{D_r - D_{r0}}} \qquad [8a]$$

$$\rho_{powder} = k_{contact} \cdot \rho_{bulk}\left(T\right) \qquad [8b]$$

where ρ_{powder} is the electrical resistivity of the powder compact, D_r is its relative density and D_{r0} is the initial relative density. This equation results from an approximated calculation of the contact areas in a regular packing of particles with increasing relative density. For $D_r = D_{r0}$, the particles are barely in contact, thus the electrical and thermal resistivity are very high. For $D_r = 1$, the porosity is negligible and the compact properties approach the bulk properties.

Heat capacity and elastic modulus are corrected for the porosity using a simple rule of mixtures. The Poisson's ratio (ν) is computed using the Clough and Shaefer equation [7]:

$$\nu = \nu_s \frac{D_r - D_{r0}}{1 - D_{r0}} \qquad [9]$$

where ν_s is the bulk Poisson's ratio.

4. Results and discussion

Figure 4 shows the isothermal map together with the electric field streamlines after 30 ms in the CDS assembly. A partial shrinkage is evidenced by the empty area in the topmost region of the upper punch. The electric field streamlines appear to be denser in the electrode and powder regions than in the die. Thus, the punches behave as the second source of heat dissipation after the powder. The electric streamlines in the die are concentrated near the powder. The resistive heating is mostly concentrated in the powder compact, as was expected from the experiments.

During CDS the powder core experiences the highest temperature and the highest densification, in agreement with experiments. The punch/powder and the powder/die interfaces are only slightly affected. The remote regions of the die do not experience a significant heating, thus confirming the experimental observations and the correctness of the boundary conditions set there. The situation is different at the topmost end of the punch, due to a fixed temperature boundary condition set at this boundary. This boundary condition attempts to reproduce the

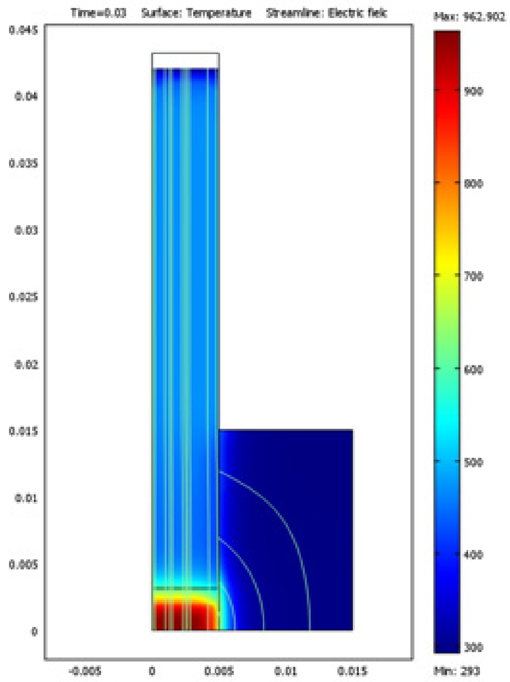

Figure 4. Isothermal map and electric field streamlines in the CDS assembly after 30 ms.

severe conduction cooling exerted by the connected constituent parts.

Figure 5 shows the detail of the isodensity map across the copper powder after 37 ms. The periphery shows a larger residual porosity. This undesired result would be probably enhanced if friction effects at particle and powder/die interfaces were included. Upon comparison of the individual sintering contributions accounted by the microscopic model, plastic flow appears to play a dominant role during CDS (figure not in included). According to Figure 6a, two densification stages can be identified. The first stage (linear) is mainly controlled by plastic deformation. The second stage, corresponding to the knee of the densification curve, is basically governed by temperature-controlled processes such as creep and diffusion. The second stage evolves towards completion at lower but steady densification rate.

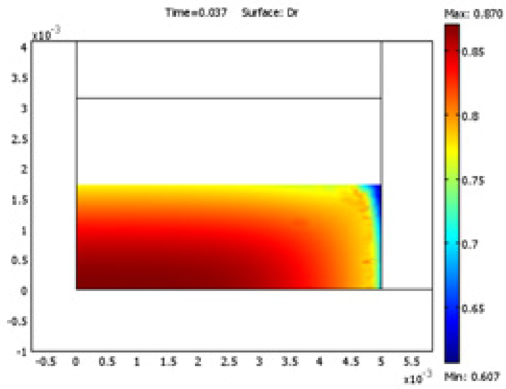

Figure 5. Detail of the relative density map in the copper powder after 37 ms.

The current peak is reached after 30 ms, whereas the temperature peak is reached later (50 ms). However, the latter's position can be influenced by the set fixed temperature boundary condition at the upper punch end. This would also affect the densification curve. Therefore, an accurate experimental evaluation of this boundary condition is required.

Computationally, despite the strong nonlinearities involved in the CDS model, including the number of the computed field variables and the moving mesh algorithm, convergence is not so critical. However, a very fine tuning of some critical parameters, such as the initial relative density, is required.

The present CDS model is validated with respect to final density measurements and will be more accurately validated against experimental measured punch displacement versus time. Further refinements of the model are required before it can be effectively used in the design of CDS molds.

5. Conclusions

The CDS process has been successfully modeled using COMSOL Multiphysics, by taking into account plastic deformation, creep

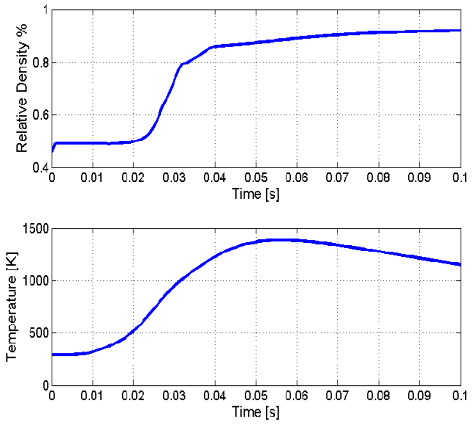

Figure 6. a) Computed relative density in the powder core; b) temperature profile in the compact core during CDS.

and grain-boundary diffusion densification factors.

CDS promotes densification in the core, rather than at the surface of the powder compact, as confirmed by experiments. The heating of the die is practically negligible. Heat-assisted plastic flow is the main responsible for the early densification. Creep and grain-boundary diffusion effects occur in the later stage of densification, at higher temperature.

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