Numerical Simulation of Thermoelectric Phenomena in
Field Activated Sintering

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Dedications

To my parents, plain hard-working people in the society,

and priceless treasure in my life.
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Abstract
Numerical Simulation of Thermoelectric Phenomena in Field Activated Sintering
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The field activated sintering technique (FAST) is a powder consolidation process that involves the application of pulsed current and pressure. Compared with hot pressing and hot isostatic pressing, FAST is a demonstrated rapid densification process that has the potential to minimize grain growth, which is crucial to achieving excellent mechanical properties. In this study, a coupled thermal-electric-densification framework for the design and optimization of FAST-type processes has been proposed and implemented into a finite element package (ABAQUS). A detailed evaluation of the accuracy of the model has been carried out for electrically conductive and insulating materials. The evaluation, compared with experimental measurements, indicated that the model provided a reasonably accurate prediction of the thermal and electric responses. A discrete finite element method was also developed to determine the effective thermal conductivity in a particulate system with a realistic microstructure. In addition to the usual examination of its dependence on relative density, an anisotropy of conductivity was found as a result of the application of non-isostatic stress. This was confirmed qualitatively by experimental observation. Finally, an exploration of FAST simulation of a complex shape part was attempted.
CHAPTER 1. INTRODUCTION

1.1 Motivation

Powder consolidation techniques such as hot pressing (HP) (Figure 1.1) or hot isostatic pressing (HIP) often need a long heating period of up to several hours, leading to grain coarsening of the microstructure and preferred segregation of certain phases at grain boundaries [1.1]. In contrast, the field activated sintering technique (FAST) [1.2] has proven to be a rapid densification process that has the potential to minimize grain growth, which is crucial to achieving improved mechanical properties. The recent emphasis on nanocrystalline materials has increased the interest in FAST.

In FAST-type processes, the sintered part is produced from powders via the simultaneous application of pressure and external electric current and/or electric field as shown in Figure 1.1.

Even though there are numerous efforts of experimental studies on different materials, and some recent attempts in modeling (see Section 1.2.2 for details), the understanding of the mechanisms in FAST-type process is still limited. Most specimens made from FAST are small (~10 mm thick) and in regular shapes (e.g., disk or cylinders). In order to extend FAST-type processes from laboratory scale to production successfully, it is important to be able to predict phenomena such as thermal transport, electric conduction and densification. In other words, there is a need for a design and optimization tool for FAST-type processes.
In the following section, the historical progress of sintering under electric current and/or electric field is first reviewed. The different FAST variations are then compared. The current understanding of the underlying physical mechanisms is also summarized.

1.2 Literature review

The application of external electric current to assist sintering was initiated by Taylor in 1933, who incorporated the idea of resistance sintering during the hot pressing of cemented carbides [1.3]. Later, Cramer patented a resistance sintering method to consolidate copper, brass and bronze in 1944 in a spot welding machine [1.4]. The concept of compacting metallic materials to a relatively high density (> 90% of theoretical) by an electric discharge process was originally proposed by Inoue in the 1960s [1.5]. Inoue argued that a pulsed current was effective for densification at the initial sintering stages for low melting point metals (e.g., bismuth, cadmium, lead, tin) and at the later sintering stage for high melting metals (e.g., chromium, molybdenum, tungsten). In the United States, Lenel also used a spot welding machine for the sintering of metals [1.6]. In addition to continuous pulses (e.g., Inoue’s method), some researchers also investigated a single discharge method, i.e., the powders were densified by a single discharge generated from a capacitor bank. In the late 1970s, Clyens et al. [1.7], Raichenko et al. [1.8] and Geguzin et al. [1.9] studied the compaction of metal powders using electric discharge compaction (EDC) or
electric discharge sintering (EDS). The single discharge method is only satisfactory for producing relatively porous parts. In all the methods cited (i.e., resistance sintering, single discharge method, and continuous pulse method), electrically conductive powders are heated by Joule heating generated by an electric current. In order to confine the electric current within the powders, the die has an insulated interior wall surface (e.g., flakes of mica or coating a non-conductive lubricant) or by making the die from an insulating material (e.g., glass, mica, Bakelite, porcelain). In 1990 Sumitomo Heavy Industries Ltd. (Japan), based on the design of Inoue [1.5], developed the first commercially operated plasma activated sintering (PAS) and spark plasma sintering (SPS) machines with punches and dies made from electrically conductive graphite [1.10]. One of the salient features of these machines was that, in addition to electrically conductive powders, high density was also achieved in insulating materials. In 1995, a process called electroconsolidation® was commercialized in the United States [1.11]. In electroconsolidation®, the specimen is immersed in a bed of graphite granules inside a die. Graphite particles act as a "pseudo-fluid" transmitting mechanical pressure to the specimen. They also serve as heating elements by Joule heating when current is transmitted through them. The roughness of the specimen depends on the size of the graphite particles. Despite the fact that uniaxial pressure is applied, compared with other FAST-type processes, the specimen in electroconsolidation is exposed to a "pseudo-isostatic" pressure when embedded in free-flowing particulates that act as a pressure-transmitting medium inside the die.
The principal differences between these techniques are shown in Figure 1.2 in terms of the applied electric current and applied pressure [1.10, 1.12]. Resistance sintering, in most cases, involves the application of direct current (DC) through an electrically conductive powder (Figure 1.2 a). Constant or no pressure is applied. In the latter case, the powders are compacted into a preform before resistance sintering. High Energy High Rate Processing (HEHR) and pulsed electro-magnetic method convert pulsed electric/magnetic energy to a mechanical pulse and concentrate the pulse in the compaction zone (Figure 1.2 b). Electric discharge compaction (EDC) and electric discharge sintering (EDS) use currents generated from a bank of capacitors. During the discharge, the voltage oscillates and eventually diminishes. The powder is subjected to a constant pressure during the process (Figure 1.2 c). In plasma assisted sintering (PAS) process, a pulsed direct current is normally applied at room temperature for a short period of time followed by a constant DC applied during the remainder of the sintering process (Figure 1.2 d). This procedure is often referred to in the literature as a "single pulse cycle process", with a typical 30 to 60 ms pulsed current of approximately 1000 A for 60 to 90 s. In the SPS process, a pulsed DC (36 ms on and 6 ms off pulses of 0.5 to 10 kA intensity) is applied repeatedly from the beginning to the end of the sintering cycle (Figure 1.2 e). In this case the procedure is referred to as a “multiple pulse cycle process”. The basic difference between SPS and EDC is in the number and voltage of discharges: multiple discharges in the former and only one discharge in the latter. For these techniques, it has been shown that an electric discharge per se does not consolidate powders to high densities and, therefore, some additional effects are needed to increase the final density (e.g.,
pressure application and/or higher temperature). For electroconsolidation, a direct current is usually used (Figure 1.2 f).

In this work, the focus is on SPS only. SPS will be used interchangeably with FAST. Currently, laboratory scale FAST experiments have succeeded in sintering a large variety of materials from polymers (thermosetting polyimides), metals (nickel, tungsten), intermetallic compounds (TiAl, NiAl, Nb3Al, FeSi2) to composites [1.2]. FAST sintered materials included ceramic oxides (Al2O3, superconductor) and nonoxide-ceramics (AlN, Si3N4, WC, TiN, YBCO) [1.2]. More importantly, nanocomposites synthesized by FAST have demonstrated exceptional mechanical properties. For example, nanocomposites of single-wall carbon nanotubes with a nanocrystalline alumina (Al2O3) matrix were sintered at 1150°C. A fracture toughness of 9.7 MPa m¹/², nearly three times that of pure nanocrystalline alumina, has been reported [1.13]. β-sialon, a solid solution based on β-Si₃N₄, demonstrated a high damage tolerance due to an interlocking microstructure sintered at 1600 °C [1.14]. It is noteworthy that these nanocomposites cannot be prepared without FAST [1.13-1.16]. Therefore, FAST can be the enabling process in the fabrication of components made of advanced materials, such as nanocrystalline particles. In this way, the full promise of this unique class of materials is expected to be realized.

1.2.1 Early understanding of FAST from experiments

The major advantage of FAST in comparison with conventional sintering (e.g., HP) (see Figure 1.1) is that sintered parts with high relative density are achieved
(a) in a shorter time, a key factor of retention of microstructures, and (b) at a claimed lower temperature. Several explanations have been proposed for the effect of FAST:

1. **Plasma generation**

   It was originally claimed by Inoue and the FAST process inventors that the pulses generated sparks and even plasma discharges between the particle contacts, which was the reason that the processes were named spark plasma sintering and plasma activated sintering [1.5, 1.10, 1.17]. They claimed that ionization at the particle contact due to spark discharges developed “impulsive pressures” that facilitated diffusion of the atoms at contacts. Groza suggested that a pulsed current had a cleaning effect on the particle surfaces based on the observation of a grain boundary without oxidation formed between particles [1.18]. Munir and Smaltzried argued that the application of an electric field enhanced grain growth and therefore densification [1.19]. Whether a plasma is generated or not has not yet been confirmed directly by experiments. Therefore, there is no conclusive evidence for the effect of a plasma generation in FAST [1.20]. The occurrence of a plasma discharge is still debated, but it seems to be widely accepted that occasional electric discharges may take place on a microscopic level [1.21]. The rough morphology observed at the contacts [1.22], and the sudden increase of conductivity of compacts subjected to an increasing electric field [1.23] implied the discharge or electrical breakdown at the particle contacts. However, all these observations are from experiments with significant field strength, e.g., $10^6$ V/m [1.23]. Discharge patterns were also observed at the sample surface [1.24]. However, the occurrences of these events are sporadic.
The schematic of electric field across the particle contact is shown in Figure 1.3. The electric field follows the relationship of $1/R_p$ [1.25] within the particle with radius $R_p$. In the gap, the field decays rapidly and is reasonably assumed to be $1/R_p^n$, where $n>1$.

Field amplification at the interface is also shown in Figure 1.3. The magnitude of field amplification could be related to the electric conductivity of powders. For electrically conductive powders, the electric field decreases substantially within the particle, so the field amplification at the gap may not be remarkable. In contrast, the field amplification for electrically insulating powders, as discussed below, could be large enough to cause sparking.

Under typical FAST conditions, approximately 1V is applied on a specimen of 10 mm thick. The average contact number through the thickness is about 1000 when 10 µm powders are considered. Taking conductive path or tortuosity into account, voltage drop per contact is about 0.001 V and the total electric field across single contact is 100 V/m. This is much below the threshold of sparking (~ $10^6$V/m [1.23]). For electrically insulating powders, the potential difference, 0.001 V, could be completely applied on the gap. As the gap is small enough, e.g., less than $10^{-4}R_p$, then local field at the contact could be as high as $10^7$V/m which is high enough to generate sparks.

2. Electroplastic effect (Electron wind)

Metal powders have been observed to exhibit a lower yield strength under an electric field. Raichenko et al. [1.8] and Conrad [1.26] independently studied
electroplastic phenomena, i.e., the interaction between moving electrons and metal atoms. Yield stress decreased with increasing intensity of the electric field. This effect, however, was observed at an electric field strengths of the order of $10^9$ V/m which is much higher than in a typical FAST condition (<100V/m or about 0.001 V per contact [1.23]).

3. Joule heating at the particle contacts

Joule heating due to the passage of electric current through particles assists in the welding of the particles under mechanical pressure. Nagae et al. [1.22] claimed oxide layer destruction by measuring the electrical resistivity of the specimen. The oxide layers were supposed to fracture due to the high temperature generated at contacts by Joule heating. However, because the size of the powder is typically small, the heat is rapidly conducted away from the contact. For alumina powders with diameter of 0.5 µm, the time to equilibrium temperature is about $10^{-9}$ s, which can be estimated by $(d/2)^2/\alpha$, where $d$ is diameter of the powder and $\alpha$ is diffusivity.

4. Pulsed current

Current understanding of the effect of pulse frequency on compact density is still incomplete. The typical approach is to compare the temperature or density of a compact processed by direct and pulsed currents, respectively. Inoue [1.5] claimed that there was a frequency-dependent effect in his patent. Nishimoto et al. [1.27] showed that the densification rate of Fe and Ni based alloy was about 5% faster with pulsed current compared with direct current. A recent comparison of Al powder
densified using DC and pulsed currents (300, 10k, and 40k Hz) by Xie et al. [1.28]
showed no significant difference.

Nagae et al. [1.22] argued that pulsed DC may have a non-thermal effect. It
could remove the trapped gas at lower temperature that prevented sintering of gas
atomized aluminum powder. He claimed that, during the initial part of the sintering
process, the sparks and/or plasma discharges cleaned the surfaces of the powders
from adsorbing CO₂ and H₂O. In subsequent stages, the presence of cleaned and
activated surfaces was expected to enhance grain boundary diffusion which promoted
transport of material and thus facilitated densification and grain growth.

5. Excessive mechanical pressure

Nygren et al. [1.29] claimed that the mechanical pressure applied in FAST
was higher than that used in normal hot pressing processes. It is generally accepted
that application of mechanical pressure is helpful in removing pores from compacts
and enhances diffusion. However, there is no reason to believe that there is a different
effect in FAST than in conventional sintering under pressure.

6. Ambiguity in specimen temperature

The advantage of FAST is often demonstrated in a claimed low temperature
compared with conventional HP or HIPing [1.22, 1.30 - 1.34]. For example, Tamari
et al. [1.32] observed that SPS reduced the sintering temperature for SiC by 200 °C.
In many references the exact conditions for conventional hot pressing are barely
mentioned. Among other differences, the rate of heating may be substantially
different in the various techniques. In FAST, extremely rapid heating rates of up to 1000 °C/min can be achieved. In conventional sintering, typical heating rates are usually very low (<50-80 °C/min), especially in tubular resistance heaters that surround the die assembly. Usually, temperatures above 1100 °C are read by a pyrometer viewing the surface of the die. This temperature is not representative of the temperature within the specimen. Some recent publications indicated that there may be a difference between the actual specimen and measured temperature [1.35 - 1.39]. For example, Tomino et al. [1.39] reported a temperature difference between the specimen center and the die surface of ~200 °C for copper and ~110 °C for alumina. There are also reports of microstructural inhomogeneities within FAST-type processed specimens, e.g., larger grains at the periphery of WC specimens than in the center [1.40], and faster densification of Al powder at the specimen surface than at the center [1.41]. Such observations may suggest indirectly that higher temperatures exist at the surface than at the center, or there are compositional gradients due to the interdiffusion of the die and specimen materials.

### 1.2.2 Early attempts at modeling FAST

In addition to experiments, some researchers have performed numerical studies. Early attempts to simulate FAST were performed by Wang et al. [1.38], Mori et al. [1.42], Fessler et al. [1.43], Heian et al. [1.44], and Matsugi et al. [1.45]. In all these works, only the specimen [1.38, 1.43, 1.44] or specimen with punches and die [1.42, 1.45] were considered. Wang et al. [1.38] assumed that the specimen was a
cylinder and applied a one-dimensional heat transfer solution. In [1.42], this problem was considered under the restrictive assumption that all current flowed through the specimen. Fessler et al. [1.43] examined electroconsolidation considering thermal and electric analyses to be decoupled by assuming that the electrical properties do not depend on temperature. The electric problem was solved first, and Joule heating was calculated. Then the results were passed to a thermal transfer algorithm. Heian et al. [1.44] and Matsugi et al. [1.45] used a partial difference method. The major limitation of these models was that only the specimen and punch/die assembly were modeled. The boundary conditions selected were also often not realistic. For example, in [1.44], a potential difference as high as 15 V was applied directly to the top and bottom surface of the specimen. The conclusions regarding temperature distribution and reaction kinetics from such simulations would be problematic.

1.2.3 Critical needs

In the long term, the central claim of FAST that causes “activated” sintering needs to be verified directly. Since plasma generation has proven to be difficult to detect, a different approach is followed.

This work is motivated by the need to analyze and evaluate the exact electric current path and temperature distributions in FAST. It is possible that electrically conductive and insulating powders may respond differently in FAST and this difference may also be crucial to the evolution of porosity during sintering. Moreover the true conditions (e.g., voltage, temperature) to which the specimen is subjected
need to be understood. Currently, such details are evaluated only indirectly (for example, voltage drop across the whole SPS system and temperature measurements on the surface of the die). At the same time, despite the fact that the fundamental mechanisms are not clear, FAST appears to be successful on a laboratory scale. Further advances and scale-up, however, require the ability to design complex shape parts. In these cases, the effect of the specimen geometry on the electric current and heat conduction is not trivial.

### 1.3 Goals of this work

The main aim of the present study is to provide physical insight into the thermal energy and electric transport phenomena in FAST. The ultimate goal is to provide a design tool to control and optimize the FAST-type process.

Electricity, heat transfer and densification phenomena occurring during FAST are fully coupled, as shown in Figure 1.4. The coupling of thermal and electrical phenomena\(^1\) originates from: (a) the temperature dependence of electrical conductivity for all components of the system, and (b) the Joule heating generated by the electric current. Densification is obviously temperature activated. The electroplastic effect, or the interaction between moving electrons and dislocations, has been observed in metals under high current densities \((10^5 \sim 10^8 \text{ A/mm}^2)\) [1.26].

The implementation of a fully coupled thermo-electrical-densification simulation is difficult, at least in the context of commercial finite element codes such

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\(^1\) The thermoelectric coupling of the Seebeck and Peltier effects [1.46] are not considered here.
as ABAQUS. Although there is a way to model this coupling for simple geometries, the focus in this study is on thermoelectric coupling.

With these issues in mind, the specific goals of this study are outlined below:

- An approximate lumped resistance model to facilitate a quick simulation of FAST has been proposed. This procedure provides a reasonable insight for the electric current and Joule heating distribution. The details of the lumped resistance model are presented in Chapter 2.

- A coupled thermal-electric-densification framework for the design and optimization of the FAST-type process has been proposed and implemented into a finite element package (ABAQUS). The accuracy of the model has been examined for electrically conductive and insulating materials. The details of the framework and evaluation are given in Chapter 3.

- Simulation results showed that the heating of the specimen was achieved primarily by conduction. This realization provided the motivation to examine heat transfer in compacted powders. A discrete finite element method was developed to determine the effective conductivity in a particulate system with a realistic microstructure. In addition to the usual examination of its dependence on relative density, the anisotropy of conductivity as a result of the application of non-isostatic stress was examined. This was confirmed qualitatively by
experimental observations. The details of the discrete finite element method are presented in Chapter 4.

- A temperature closed-loop controlling algorithm has been implemented in the finite element package (ABAQUS). The temperature control function, which is employed in the FAST-type system, was simulated successfully. The subroutine is given in Appendix A.

- A procedure for the characterization of electric and thermal gap conductances has been developed. For the same system geometry, a single calibration may be sufficient, and substantially reduces the calibration effort. The details are given in Appendix B.

- A preliminary study of FAST for a complex shape part has been conducted. The results are presented in Appendix C.

The conclusions of this work are summarized in Chapter 5, where suggestions for future research are also outlined.

The impact of this work is threefold:

- A proper evaluation of temperature gradients in FAST is achieved. This allows for: (a) an unbiased comparison with traditional sintering techniques – underestimation of specimen temperature by a pyrometer measuring die surface temperature leads to an incorrect comparison, and (b) the possibility for optimization of the processing of large size specimens, where the presence of large temperature gradients within the sample may be detrimental to the properties of the final product.
• The electric field in the specimen is estimated. This provides a realistic starting point for further investigation of other phenomena (e.g., the conditions for generation of a plasma or a discharge).

• The discrete element model enables further understanding of thermoelectric phenomena in other processes, such as selective laser sintering (SLS), resistance welding, and in the reliability of electronic components.
Figure 1.1 Schematic representation of (a) HP and (b) FAST.
Figure 1.2 Variations between different field activated sintering techniques. (a) – (e) are modified from [1.10].

(a) Resistance sintering
I/S ~ 10^4 kA/m^2
P ~ 1 MPa
< 350 s

(b) High Energy High Rate Processing (HEHR)
U ~ 5 - 10 kV
P ~ 0 MPa
< 3 s

(c) Electric Discharge Compaction (EDC)
U ~ 5 - 10 kV
I/S ~ 100 kA/m^2
P ~ 1 MPa
100 µs

(d) Plasma Assisted Sintering (PAS)
Pulsed DC current, I ~ 1000 A
P ~ 100 MPa
~ 50 s
< 500 s

(e) Spark plasma sintering (SPS)
Pulsed current, I ~ 1000 A
P < 100 MPa
< 500 s

(f) Electroconsolidation
DC, I/S ~ 1500 kA/m^2
P ~ 70 MPa
< 3000 s
Figure 1.3 Schematic of electric field along two neighboring particles. $R_p$ is particle radius.
Figure 1.4  Schematic of the coupling of the thermo-electrical-densification model used in this work.
CHAPTER 2. SIMPLIFIED LUMPED ELECTRIC RESISTANCE MODEL

2.1 Introduction

A simplified model can be useful due to the possible constraint on computational power, time and available data. In this chapter, a simplified lumped electric resistance model of FAST-type system is presented which provides a first order estimate of the electric current flow in the system.

2.2 Simplified lumped electric circuit model

Each graphite component in FAST was modeled as an electric resistor. Given the operating frequency (~ 20 kHz), the system exhibits resistive behavior only, i.e., there are no capacitive and inductive elements. The origin of induction or capacitance based effects comes from the particulate nature of the powder. Preliminary calculations show that such effects became important only at gigahertz levels of frequency. As electric current flows through these components, an equivalent lumped electric resistance circuit is formed by connecting different resistors following all possible current paths. By considering a lumped electric resistor, we assume that there is no spatial distribution of electric field within the resistor and that the resistors are connectedly by perfect conductors.
In FAST, each graphite component is in the shape of either a cylinder or a tube. The formulae for the resistance of a homogeneous material of uniform cross section carrying a steady current are described in most electromagnetic fields textbooks (e.g., [2.1]). Note that the resistance depends not only on the geometry of the resistor but also the direction along which current flows. In the case that current flows along the axial direction, the resistance is calculated as

\[
R = \frac{1}{\sigma A} \frac{L}{A}
\]  

(2.1)

where \( \sigma \) (\( \Omega \text{m} \)^{-1}), \( L \) (m), \( A \) (m^2) are electrical conductivity, length and cross sectional area of the resistor, respectively. When the current flows along the radial direction of a tubular resistor, the resistance is expressed by

\[
R = \frac{1}{\sigma} \frac{\ln(R_{\text{out}} / R_{\text{in}})}{2\pi L}
\]  

(2.2)

where \( R_{\text{in}} \) and \( R_{\text{out}} \) are the inner and outer radii of the tube, respectively.

Joule heating is calculated by

\[
P = I^2 R
\]  

(2.3)

where \( I \) (A) is the electric current flowing through the resistance \( R \) (\( \Omega \)).
2.3 Results and discussion

Figure 2.1 shows the schematic of a FAST system and its corresponding simplified lumped electric resistance model. The geometry of each component is given in Table 2.1.

Figure 2.1(b) shows that the specimen and die are connected in parallel. The ratio of current flowing through the specimen and die is inversely proportional to the ratio of their resistances:

\[
\frac{I_{\text{die}}}{I_{\text{sp}}} = \frac{R_{\text{sp}}}{R_{\text{die}}} \tag{2.4}
\]

The relation is plotted in Figure 2.2. Note that the ratio of the current is determined by the resistance ratio, not the resistivity ratio since the geometry of the component also comes into the picture. When sintering an insulating powder, e.g., alumina or for a low relative density during the initial sintering stage, the resistance of the specimen is high. Therefore, little or no current flows through the specimen unless the die is insulated on its internal surface. On the other hand, when sintering conductive powders, or as the relative density increases, the resistance of the specimen is reduced, more current is conducted within the specimen. In general, it is fair to say that, due to the porosity or due to powder surface contamination, the resistivity of any powder compact, including conductive ones, would be higher than that of graphite.
The voltage drop across the specimen is only a small fraction of the total voltage difference applied on the system, $V_{tot}$. The voltage across the specimen increases monotonically with $R_{sp}$. Its maximum value, $V_{sp}^{\text{max}}$, occurs when $R_{sp}$ approaches infinity (i.e., an insulating specimen):

$$V_{sp}^{\text{max}} = V_{tot} \frac{R_{die}}{R_{die} + R_{p} + R_{sys}}$$  \hspace{1cm} (2.5)$$

This means that if the goal is to maximize the voltage across the specimen for an insulating specimen, the resistance of the die should be maximized ($R_{die} >> R_{p} + R_{sys}$). This can be achieved by insulating the interior wall surface of the die.

The amount of Joule heating generated within in each component is given in Table 2.2. The maximum Joule heating is generated in the punches independently of the resistivity of the specimen. This happens because the punch offers the smallest cross sectional area which makes the corresponding resistance high, thus dominating the overall resistance. For the specific values of the resistances in the circuit shown in Figure 2.1, the punches generate approximately 70\% of the Joule heat in the system (Table 2.2). Maximum Joule heat is qualitatively consistent with the experimental observations. That is, the punches experience the highest temperature, as shown in Figure 2.3.

Assuming a constant current $I_0$ transmitting through the system, Joule heating in the specimen normalized by $I_0^2 R_{die}$ is plotted in Figure 2.2:
\[
\frac{P_{sp}}{I_0 R_{die}} = \frac{R_{die} R_{sp}}{(R_{die} + R_{sp})^2}
\]  

(2.6)

Joule heating in the specimen increases with the resistance ratio, \(R_{die}/R_{sp}\), till it reaches the maximum fraction, 0.25. For higher values of \(R_{die}/R_{sp}\), the Joule heat decreases.

The maximum Joule heating in the specimen, \(P_{sp}/P_{tot}\), is readily calculated when the following condition is met:

\[
R_{sp} = R_{die} \sqrt{\frac{R_{sys} + R_p}{R_{sys} + R_p + R_{die}}}
\]  

(2.7)

When the resistances of the system and punch are high (in our case, \(R_{sys} + R_p \gg R_{die}\)), then it is easily shown that the maximum Joule heating in the specimen occurs when the specimen resistance is equal to that of the die as explained by the maximum power transfer theorem [2.2].

2.4 Conclusions

In this chapter, a simplified lumped electric resistance model of a FAST-type system has been studied. The results can be applied to (a) estimate the electric current distribution and resulting Joule heating for sintering electrically conductive and insulating powders or for the powders in which the resistivity evolves due to
densification during sintering. Electric current path is determined by the resistance ratio of specimen to die. Note that both the resistivity and geometry of the components determine the resistance; and (b) identify the potential heat source, i.e. the component with maximum Joule heating. Maximum Joule heating in the specimen is achieved when its resistance is equal to that of the die.
Figure 2.1 (a) Schematic of the current flow in FAST; (b) Equivalent electric circuit. Radiation and convection are ignored here.
Table 2.1 Geometry, function and materials of each component.

<table>
<thead>
<tr>
<th>Component</th>
<th>Resistance group</th>
<th>Function</th>
<th>Material</th>
<th>Outer Diam./ Inner Diam./ Thickness (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>$R_{sys}$</td>
<td>Water cooled ram</td>
<td>Graphite</td>
<td>OD: 120, Th.: 200</td>
</tr>
<tr>
<td>B</td>
<td></td>
<td>Large disc, current transfer</td>
<td>Graphite</td>
<td>OD: 155, Th.: 20</td>
</tr>
<tr>
<td>C</td>
<td></td>
<td>Small disc, spacer</td>
<td>Graphite</td>
<td>OD: 120, Th.: 20</td>
</tr>
<tr>
<td>D</td>
<td></td>
<td>Spacer, thermal buffer</td>
<td>Graphite</td>
<td>OD: 76.2, Th.: 40</td>
</tr>
<tr>
<td>E</td>
<td>$R_p$</td>
<td>Punch</td>
<td>Graphite</td>
<td>OD: 19.1, Length: 25.4</td>
</tr>
<tr>
<td>F</td>
<td>$R_{die}$</td>
<td>Die</td>
<td>Graphite</td>
<td>OD: 44.6, ID: 19.1, Length: 38.1</td>
</tr>
<tr>
<td>G</td>
<td>$R_{sp}$</td>
<td>Specimen</td>
<td>Alumina, Graphite</td>
<td>OD: 19.1, Th.: 6.0</td>
</tr>
</tbody>
</table>
Figure 2.2 Current ratio and Joule heating.
Table 2.2  Joule heating generated in the components according to Figure 2.1. \( \rho_{\text{graphite}}=1.67\times10^{-5} \ \Omega m \) and \( \rho_{\text{Al}_2\text{O}_3}=10^{13} \ \Omega m \) at room temperature, \( R_{\text{sys}}, R_{\text{sp}} \) and \( R_p \) use Eq.2.1; \( R_{\text{die}} \) is composed of three parts along the height – top and bottom parts which are in contact with punches and are computed using Eq.2.2, middle part which is next to specimen and is calculated using Eq. 2.1).

<table>
<thead>
<tr>
<th>System ( R_{\text{sys}} )</th>
<th>Punch ( R_p )</th>
<th>Die ( R_{\text{die}} )</th>
<th>Specimen ( R_{\text{sp}} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Resistance ( \Omega m )</td>
<td>1.0</td>
<td>3.0</td>
<td>0.5</td>
</tr>
<tr>
<td>Current</td>
<td>( I_0 )</td>
<td>( I_0 )</td>
<td>( R_{\text{sp}} ) ( I_0 )</td>
</tr>
<tr>
<td>Joule heat</td>
<td>( I_0^2 R_{\text{sys}} )</td>
<td>( I_0^2 R_p )</td>
<td>( \frac{(R_{\text{sp}})^2 R_{\text{die}}}{(R_{\text{die}} + R_{\text{sp}})^2} I_0^2 )</td>
</tr>
<tr>
<td>Joule heat fraction</td>
<td>0.24</td>
<td>0.70</td>
<td>0.03</td>
</tr>
<tr>
<td>(graphite)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Joule heat fraction</td>
<td>0.22</td>
<td>0.67</td>
<td>0.11</td>
</tr>
<tr>
<td>(alumina)</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Figure 2.3 Image of punch and die of SPS during heating stage (courtesy of Sumitomo Heavy Industries, Ltd. Japan)
3.1 Introduction

In the previous chapter, the one-dimensional, steady state electrical conduction of a simplified lumped electric resistance model of a FAST-type system was considered. It provided only a first order estimate of the electric field distribution. Electricity, heat transfer and densification phenomena present during FAST are fully coupled as shown in Figure 1.4. The coupling of thermal and electrical phenomena originates from: (a) the temperature dependence of electric conductivity for all components of the system, and (b) the Joule heating generated by electric current. Densification is obviously temperature activated. Electroplastic effect, or the interaction between moving electrons and dislocations has been observed in metals under high-density electric current.

Coupling between heat transfer, electricity condition and densification renders the temperature field complicated. In FAST, the fast heating rate (up to 1000 °C/min) makes the temperature field even harder to predict due to the transient response.

As discussed in Chapter 1, electric current application may create favorable conditions for the generation of micro sparks between contacts which may remove the impurities on the powder particle surface. However, information on electric field strengths at the particle level is still missing. In order to know the spatial and temporal information of temperature field, electric field and densification in FAST-type processes, a framework of modeling addressing these phenomena is needed.
In this chapter, the procedure will be outlined and its accuracy will be evaluated by comparing the prediction of temperature and electric fields for electrically conductive and insulating materials against corresponding experimental measurements. This work will provide a basis of design tool for FAST of complex shape and large size parts.

3.2 Modeling framework

3.2.1 Theory

As discussed in Chapter 1, electric conduction, heat transfer and densification phenomena are coupled in FAST. The thermal-electrical coupling can be described by the well-known energy balance equation for the transient heat transfer [3.1]:

\[
\int_v \rho c_p \frac{\partial \theta}{\partial t} dV = \int_v \nabla \cdot (k \nabla \theta) dV + \int_v \dot{q}_e dV + \int_S (\dot{q}_{e_{\text{conv}}} + \dot{q}_r + \dot{q}_{e_{\text{ec}}}) dS \quad (3.1)
\]

where \( V(\text{m}^3) \) is any control volume enclosed by a surface \( S(\text{m}^2) \); \( \rho(\text{kg/m}^3) \) the density; \( c_p(\text{J/kg-K}) \) the specific heat; \( \theta(\text{K}) \) the temperature; \( k(\text{W/m-K}) \) the thermal conductivity; and \( t(\text{s}) \) the time. The left hand term and first term on the right hand side are explained in most heat transfer textbooks (e.g. [3.2]). The last two terms of Eq. (3.1) represent heat generation within the volume \( V \) and heat exchange with the surroundings through the surface \( S \). For this problem, heat is generated throughout
the volume \( V \) by Joule heating represented by \( q_e (W/m^3) \). Surface heat fluxes that correspond to heat conduction from neighboring volumes, heat transfer by convection, heat transfer by radiation and interfacial heating effects are represented by \( q_e \) (W/m\(^2\)), \( q_{\text{conv}} \) (W/m\(^2\)), \( q_r \) (W/m\(^2\)) \( q_{\text{ac}} \) (W/m\(^2\)), respectively. These variables will be discussed in detail in Section 3.2.2.

Joule’s Law in a three-dimensional form is written as [3.3]:

\[
q_e = \vec{E} \cdot \vec{J} \tag{3.2}
\]

where \( \vec{E} \) (V/m) and \( \vec{J} \) (A/m\(^2\)) are electric field intensity, electric current density, respectively which are related by Ohm’s law [3.3]:

\[
\vec{J} = \sigma \vec{E} \tag{3.3}
\]

where \( \sigma \) (\( \Omega \)m\(^{-1}\)) is the electrical conductivity. In general, \( \sigma \) can be anisotropic as in the case of extruded graphite and this can be easily incorporated into the model. The electric field, \( \vec{E} \), can be also expressed by the negative gradient of the potential [3.3]:

\[
\vec{E} = -\nabla \varphi \tag{3.4}
\]
Substitution of Eq. (3.3) and (3.4) into Eq. (3.2) renders the rate of internal energy generation per unit volume due to Joule heating as a function of the electric potential, $\varphi$, only:

$$
\dot{q}_e = (-\nabla \varphi) \cdot \sigma (-\nabla \varphi) = (\nabla \varphi) \cdot \sigma (\nabla \varphi) \quad (3.5)
$$

### 3.2.2 Numerical implementation

The entire FAST system was modeled by the finite element model shown in Figure 3.1. Each component, e.g., punch, die, specimen, etc., was discretized by several finite elements to allow for non-uniform electric and temperature fields.

Thermal conduction, electric conduction and densification problems are coupled when the thermal and electric properties are functions of temperature and relative density. Simultaneous coupled solution of the thermal and electric fields is possible with the commercial finite element package ABAQUS [3.1]. There are two specific difficulties in the model: (a) how to incorporate densification into the governing equation (Eq. 3.1); and (b) how to model the interfacial thermal and electric transport properties between different components. The solutions of these problems are discussed below.

The implementation of a general fully coupled framework that incorporates thermal conduction, electric conduction and densification is currently not feasible for two reasons:
• The modeling of sintering alone is possible in the form of continuum models calibrated by experiments [3.4]. The effect of the electric field on sintering, however, is an open problem. As discussed in Section 1.2.1, such an effect might be present at very high electric fields which are much higher than the typical fields in FAST.

• Numerical implementation of the fully coupled model is complicated. At present, a coupled thermo-electrical analysis in ABAQUS cannot include displacement degrees of freedom. In order to take into account the motion of the punches due to densification of the specimen, the simulation is split into a number of steps.

Although incorporation of a sintering model into the simulation is possible, given the complexity in calibration, the problem is circumvented by retrieving the densification history from experiments. In this way, this work does not predict sintering, but analyze the thermoelectric problem, not only by including the effect of density in the simulations but also by taking into account the dimensional changes attendant to densification. In this case, the performance of the thermoelectric part of the simulation is evaluated independently to prevent from possible errors that the sintering model could have introduced. There is, however, one assumption that the density of the specimen is considered to be uniform. The majority of simulations presented here concern specimens with cylindrical geometry and the history of the specimen dimensions, and their densities are approximated in a stepwise manner (Figure 3.2). In each step a coupled thermal-electrical analysis is performed with fixed specimen dimensions and punch position. At the end of each step, the specimen
height is decreased based on the prescribed curve, i.e., displacement of the punch. A new mesh is used to match the size reduction due to sintering. The top and bottom punches and loading trains also move closer by half distances correspondingly without changing the size of mesh. Mapping of the nodal temperatures and voltages at the loading trains is required from one step to the next. A linear mapping technique is found to be adequate because the temperature variation within the specimen is relative small. Internal energy is conserved because mass is unchanged.

In order to predict the electric and thermal fields correctly, interfacial thermal and electric transport properties between different components have to be identified. Across a perfect interface, the temperature and the electric potential are continuous functions. When two real surfaces are brought into contact, however, geometric irregularities and surface deposits prevent perfect contact. Consequently, both temperature and electric potential are discontinuous across an imperfect interface. In other words, a finite temperature or voltage difference is sustained across the contact (Figure 3.3).

The notions of electric [3.5] or thermal contact resistance [3.6] are applied to explain the discontinuity of temperature and voltage. The thermal flux [3.6] and the electric current flux [3.5] across the interface are

\[ \dot{q}_c = h_c (\theta_1 - \theta_2) \]  \hspace{1cm} (3.6)

and

\[ J = \sigma (\varphi_1 - \varphi_2) \]  \hspace{1cm} (3.7)
respectively. \( h_g (W/m^2-K) \) and \( \sigma_g (\Omega m^2)^{-1} \) represent the thermal and electric gap conductances, respectively. \( \theta \) (K) and \( \varphi \) (V) are temperature and electric potential, respectively. The subscripts 1 and 2 denote the two parts in contact.

The thermal, \( R_{c \text{th}}^\theta \) (K /W), and electric, \( R_{c \text{el}}^\varphi \) (\( \Omega \)), contact resistances can be defined as [3.5, 3.6]:

\[
R_{c \text{el}}^\varphi = \frac{1}{S_A \sigma_g} \quad \text{and} \quad R_{c \text{th}}^\theta = \frac{1}{S_A h_g}
\] (3.8)

where \( S_A \) (m\(^2\)) is the apparent contacting area (Figure 3.3).

In addition to the drop of temperature and voltage, the heat flux is also discontinuous at the interface as Joule heat is generated due to the electric contact resistance [3.1]:

\[
\dot{q}_{ec} = J(\varphi_1 - \varphi_2) = \sigma_g (\varphi_1 - \varphi_2)^2
\] (3.9)

As the result, the heat flux across the contacting surfaces of two parts (1) and (2) in Figure 3.3 can be written as:

\[
\dot{q}_1 = \dot{q}_c - \frac{1}{2} \dot{q}_{ec} = h_g (\theta_1 - \theta_2) - \frac{1}{2} \sigma_g (\varphi_1 - \varphi_2)^2
\] (3.10)
\[
\dot{q}_2 = \dot{q}_c + \frac{1}{2} \dot{q}_{ec} = h_g (\theta_1 - \theta_2) + \frac{1}{2} \sigma_g (\varphi_1 - \varphi_2)^2
\]  

(3.11)

where the Joule heating due to the electric contact resistance is assumed to be evenly divided between the two contacting surfaces. The heat flux difference across the interface, \( \dot{q}_2 - \dot{q}_1 \), is \( \dot{q}_{ec} \). If there is no electric current and only heat flows across the imperfect interface, then the heat flux will be continuous since no thermal energy is generated or absorbed there.

Thermal boundary conditions on the surface, \( S \), can take any of the following forms: (a) specified boundary temperature, and/or (b) specified surface heat flux. The latter can correspond to heat conduction to or from neighboring volumes or interfaces, heat transfer by convection, heat transfer by radiation. In this problem heat transfer by convection, \( \dot{q}_{conv} \), is zero since FAST is usually performed in a vacuum chamber.

Radiation occurs in the system and is given by

\[
\dot{q}_r = \sigma_s \varepsilon [(\theta_1)^4 - (\theta_2)^4]
\]  

(3.12)

where \( \sigma_s \) is the Stefan-Boltzmann constant, \( 5.669 \times 10^{-8} \) W/m\(^2\)-K\(^4\); \( \theta_1 \) is the temperature of the emitting surface; \( \theta_2 \) is the temperature of the absorbing surface and \( \varepsilon \) is the emissivity. The problem of evaluating radiation exchange in closed cavities has been examined in [3.7, 3.8]. All exposed surfaces can exchange heat by radiation (Figure 3.1). Heat transfer by radiation between the exposed surfaces of rams,
spacers, punches, dies and interior chamber surfaces are taken into account using a geometric viewing factors algorithm [3.9] that is readily available in commercial finite element codes (e.g. ABAQUS).

For the electric boundary condition, although a number of variations of FAST are performed under alternating or pulsed direct currents, the simulations presented here involved direct current (or RMS equivalent). The effect of pulsed current can be broadly classified as follows:

- “Sintering activation” by plasma/spark generation.
- Increase of the effective resistance of the powder compact due to the introduction of equivalent capacitive and inductive impedances.

Preliminary calculations show that for typical powders these impedances become important only at frequencies of the order of 1GHz which are much higher than the typical frequencies used in FAST (~ 20 kHz).

Given that this study does not intend to predict sintering directly but rather the temperatures and electric fields that may drive sintering, it is believed that, to the first order approximation, the temperature distribution does not depend strongly on the type of current if the equivalent power supplied to the system is the same. In all simulations, the end of the lower ram is grounded and the potential is set to be zero. A potential, $\varphi$, is applied on the end of upper ram and is specified to generate a preset time-temperature profile, $\theta_{\text{surface}}$, on the outer die surface (Figure 3.1). In the experiment, this is realized by a closed-loop control system. A proportional controller is implemented in the form of an ABAQUS User Subroutine [3.1]. The temperature
of a node at the surface of the die that corresponds to the thermocouple reading is passed as feedback into the closed loop algorithm. The output voltage is adjusted by $\Delta \varphi$, which is proportional to the error, i.e. the difference between temperature read, $\theta_{\text{surface}}$, and the preset temperature, $\theta_{\text{preset}}$, as shown in Equation (3.13):

$$\Delta \varphi = -\alpha (\theta_{\text{surface}} - \theta_{\text{preset}}) \quad (3.13)$$

where $\alpha (\text{V/}^\circ\text{C})$ is a tunable constant, that represents the gain of the closed loop control system. A balance between accuracy of control and simulation time renders $\alpha$ to be $10^{-6} (\text{V/}^\circ\text{C})$. Note that the exact value of $\alpha$ depends on the system examined. The numerical implementation of the closed-loop control system in ABAQUS is given in Appendix A.

The initial temperature of all components is 25 °C. During the simulation, the temperatures of the ends of the rams, the outer surface of chamber are taken to be constant at 25 °C (Figure 3.1). The walls of the chamber are considered to be electrically insulating.

### 3.3 Evaluation of the model

The model was evaluated by comparing the spatial distribution and evolution of temperature in two cases: (a) sintering of alumina powder which is electrically insulating; and (b) the heating of a fully dense graphite cylinder. These two materials
represent two extremes of the resistivity, against corresponding experimental measurements.

3.3.1 Experimental work

All experiments presented here were performed by members of Professor Joanna Groza’s group at the University of California, Davis. Experiments were carried out using a commercial Sumitomo SPS sintering machine (Model 1050), which is a 20 ton vacuum uniaxial-press, capable of evacuating the process chamber down to 5 Pa. The power supply can provide a pulsed direct current up to 5000 A through the rams to a graphite die set that contains the specimen. A stack of conductive graphite discs is placed between the punches and water-cooled rams (Figure 3.1).

A pulsed square wave current with on/off cycles of 10-99 ms (on-time) / 1-9 ms (off-time) is available. A typical pulse duration in the SPS machine is about 3 ms. Experiments reported here employed a square wave of 36 ms on and 6 ms off. Power is controlled through the application of voltage difference on the ends of rams in order to produce a preset time-temperature profile through a closed-loop control system. The controlling temperature is monitored up to 1100 °C via a sheathed 13% Rh/Pt-Pt thermocouple attached onto the outer die surface (Figure 3.1). A single-color pyrometer is used for higher temperatures. In all experiments, current, voltage, ram displacement and load were computer logged.
In order to determine the actual temperature in the specimen, four different sets of experiments were performed. In all cases, a pressure of 15 MPa was applied on the ends of the rams. Heating rate of 50 °C/min was controlled at the outer die surface in all experiments. Two thin graphite foils (UCAR Carbon Company Inc., Wilmington, DE) were placed on the top and bottom of the specimen. In all experiments, power was controlled by the outer die surface temperature measurements, i.e., a closed-loop control system which controls the voltage difference is applied in such a way that a preset time-temperature profile at the outer die surface is followed.

In the first experiment, loose BAIKALOX ultra fine α-Al₂O₃ powder (Baikowski International Corporation) with an average particle size of 0.1 µm was sintered in a graphite die/punch assembly. No lubricants or additives were used. No thermocouples were inserted in the specimen.

In the second experiment, a solid graphite cylinder (Tokai Carbon Co. Ltd., Japan) with an embedded 13% Rh/Pt–Pt thermocouple instead of a powder specimen was used. The use of a thermocouple wire within compacting powder is usually problematic. Therefore, a solid material was selected to facilitate temperature measurement within the specimen. The inserted thermocouple was used to compare the actual temperature in the specimen with the temperature at the die surface. Note that graphite is a material with high thermal and electrical conductivity compared to typical powder compacts.
For the solid graphite cylinder specimen, three additional heating rates of 25, 110, and 220 °C/min, were applied to investigate the possible effect of heating rate on the actual specimen temperature.

Two more experiments were performed using silicon and lithium silicate. Since reliable temperature reading of thermocouples is only limited to below 1100 °C, indirect temperature estimates were provided by the melting of the powders of silicon (Alfa Aesar, Ward Hill, MA, 99.9985%, melting point 1201 °C) and lithium silicate (Alfa Aesar, Ward Hill, MA, 99.8%, melting point 1410 °C). No thermocouples were inserted in these compacts. Melting may occur as the powders are heated. This event can be detected from the ram displacement curve that exhibits a sudden increase in the rate of displacement.

**3.3.2 FE model**

Due to the axial symmetry of the arrangement, only one half of the cross-section was required. The cross section was modeled using continuum, axially symmetric, 4-node linear, coupled thermo-electrical elements, DCAX4E [3.1]. The finite element mesh generated for this cross-section is shown in Figure 3.1 and was comprised of 1895 elements. The meshes of the specimen/punch/die assembly were discretized with smaller elements to reveal detailed spatial information.

The electric conductivity, thermal conductivity and specific heat of fully dense graphite [3.10] and Al₂O₃ [3.11] are shown in Figure 3.4 and 3.5. The
anisotropy of the conductivities of the graphite foil [3.12, 3.13] are shown in Table 3.1.

To avoid the complexity of a sintering model, density evolution is assumed to be spatially homogeneous which is reasonable for a small aspect ratio specimen. Density evolution during sintering was extracted from the experimental curve of punch position. The effect of the evolving density during sintering on thermal and electric properties was taken into account. A simple linear model [3.14] for both thermal and electric conductivities of porous specimens was used:

$$\frac{k_{\text{eff}}}{k_s} = (2RD - 1) \quad \text{and} \quad \frac{\sigma_{\text{eff}}}{\sigma_s} = (2RD - 1) \quad (3.14)$$

where $k_{\text{eff}}$ and $\sigma_{\text{eff}}$ are effective thermal and electric conductivities at relative density $RD$, respectively. $k_s$ and $\sigma_s$ are fully dense thermal and electric conductivities which are given in Figure 3.4 and 3.5.

Relative density is related to the height of a cylindrical specimen by mass conservation:

$$\frac{RD}{RD_0} = \frac{V_0}{V} = \frac{H_0}{H} \quad (3.15)$$

where $V$ and $H$ are the current volume and height of specimen at relative density $RD$, respectively. $RD_0$ is the initial relative density.
Thermal and electrical contact resistances were implemented using gap thermal (Eq. 3.6) and electric (Eq. 3.7) conductances in ABAQUS. The calibration of the electric contact resistances in the punch/specimen/die system was based on a series of experiments without specimens reported in detail in Appendix B. These experiments showed that the contacts along axial direction exhibited a higher contact resistance than the transverse contacts (Figure 3.1). A possible explanation for this behavior was that contact resistances depended on the applied pressure on them [3.15]. The electric gap conductances along the axial and transverse directions at 25°C are 1.25×10⁷ and 7.5×10⁶ (Ωm²)⁻¹, respectively. In general, these conductances are temperature dependent, reflecting the conductivities of the contacting materials and geometrical changes of the asperities of the contacting surfaces due to applied or induced loads. To simplify the calculations, the temperature dependence of the electric gap conductance is assumed to be identical to the corresponding temperature dependence of the contacting materials.

The coupling between thermal and electrical phenomena renders the calibration of thermal contact resistance less straightforward than the electric contact resistance. For this reason, a set of results based on no thermal contact resistance is presented first, and subsequently the possible effect of thermal contact resistance based on a parametric study is discussed.

The commercial finite element code ABAQUS was used to perform the coupled thermo-electrical analysis. The coupled thermo-electrical equations were solved for both temperature and electric potential at the nodes. Electric and
temperature fields inside the element were interpolated with the same interpolation function [3.1].

Maximum temperature change to be allowed in an increment, DELTMX, was taken to be 10K. The initial time increment is 0.01s based on the relation:

$$\Delta t > \frac{\rho c}{6k} (\Delta l)^2$$

(3.16)

where $\rho$ (kg/m$^3$), $c$ (J/kg-K), $k$ (W/m-K) and $\Delta l$ (m) are density, specific heat, thermal conductivity and characteristic length of an element, respectively. This relation defined the minimum stable time step for the heat transfer analysis.

The 6.3 version of ABAQUS/Standard was used. The total CPU time was less than one hour on a 1GHz 1GB RAM PC.

### 3.3.3 Experimental results

In the experiment involving a fully dense graphite cylinder instead of a powder specimen, the interior and die surface temperatures were measured simultaneously by a thermocouple in the center of the specimen, $\theta_{center}$, and a thermocouple or a pyrometer on the outer die surface $\theta_{surface}$. The positions of the thermocouple and the pyrometer are shown in Figure 3.6. With thermocouple readings limited to $\theta_{surface} < 1100$ °C, higher temperatures were indirectly estimated by measuring $\theta_{surface}$ at the onset of melting of powders for which the melting points are known (1410 °C for lithium silicate and 1201 °C for silicon). These melting
events were observed in the ram displacement curve (Figure 3.7). The first onset of melt formation was indicated by a moderate and steady increase in displacement. Onset of melting commenced at the top surface of the sample and corresponded to a position denoted as $\theta_{\text{top}}$ in the schematic included in Figure 3.6. The second event noted in Figure 3.7 corresponded to the extrusion of melt out of the die. As the temperature increased further, more liquid became available and eventually reached the gap between punches and die. Given that the first melting event detected in the ram displacement curve represented the temperature on the top surface of the sample, two additional points are included in Figure 3.6, one for silicon and one for lithium silicate. The excess of the interior temperature ($\theta_{\text{center}}$) over die surface temperature ($\theta_{\text{surface}}$) increased considerably from 65 °C at 650 °C to 240 °C at 1180 °C. The presence of such a temperature gradient also implied that the heating rate in the center of the specimen was higher than that of the die surface by about 70%.

### 3.3.4 FE model results

In this section, attention was focused on the accuracy of the model by comparing predictions against experimental data. In particular, the following effects will be discussed: (a) material properties on the temperature difference between the specimen and the die surface; (b) heating rate on the abovementioned temperature difference; (c) resistivity of specimen on the distribution of electric and thermal fields; and (d) electric gap conductance on the energy efficiency.
The effect of material properties on the temperature difference

Figure 3.8 shows the evolution of temperature on the surface of the die used to control the voltage applied to the load train. The result of the simulation was not a prediction but rather an indication that the numerically implemented control algorithm indeed allowed the system to follow faithfully a prescribed temperature history.

As discussed before, the coupling between thermal and electric phenomena rendered the calibration of thermal contact resistance less straightforward than the electric contact resistance. For this reason, a set of results with no thermal contact resistance was first presented.

The modeling confirmed the experimental observation that the punches act as heat generating sources and the die and specimen as heat sinks as shown in Figure 3.6. Thus, $\theta_{\text{top}}$ is the highest temperature within specimen. The specimen experiences lower temperatures for positions closer to the die ($\theta_{\text{edge}}$). Both model and experiment suggest an increase of this effect with higher temperature. However, the overall differences found in the experiments exceed the modeling results by a factor of 3. The finite element simulation with no thermal contact resistance shows a similar trend with the experimental data, an almost linear increase of internal temperature with surface temperature.

The slope of the experimentally measured relation between the two temperatures was higher than the predicted one. The difference between the experimental and model predictions of the temperature difference from the specimen
center to die surface, $\theta_{\text{center}} - \theta_{\text{surface}}$ (Figure 3.6), may be attributed to the following, as illustrated in Figure 3.9:

(a) Overestimation of the thermal diffusivity of the die. A lower thermal diffusivity obviously will amplify the thermal gradients. For a 50% lower thermal diffusivity the corresponding temperature difference increased by about 80% and reduced the difference between the numerical and experimental results (Figure 3.9). Although it is possible that the presence of porosity (12%) may reduce the thermal diffusivity of graphite, the corresponding difference in temperature should not be more than 20% than the one currently predicted.

(b) Underestimation of the emissivity of the die surface. Emissivity is not easy to measure and depends on many parameters such as surface roughness. Using emissivity of $\varepsilon=1.0$, i.e., a black body, instead of the value $\varepsilon=0.8$, resulted in a 10% increase of the temperature difference at a temperature of the die surface of about 900 °C. Given that the value of 0.8 is more realistic [3.16], it is clear that this was not the reason for the discrepancy.

(c) The presence of thermal contact resistance on the inner vertical surface of the die. This concept is analogous to the electric contact resistance that has been clearly proven in the previous work. When there is an electrical contact resistance, then, by default, there is also a thermal contact resistance. To evaluate this possibility while keeping the simulation tractable, the ratio of axial over the transverse thermal gap conductance is chosen to be scaled in the same way as that of electric gap conductance. By varying thermal gap
conductance, it is possible to match exactly the experimental data (Figure 3.6). Note that the incorporation of the thermal contact resistance does not change significantly the prediction of electric current flow.

Parametric studies showed that a combination of (a) and (c) was the origin of the discrepancy.

The results presented above show that measuring the temperature on the surface of a die does not provide useful readings for temperatures above 1000 °C. In fact, relying on $\theta_{\text{surface}}$ without calibration makes the internal temperatures unpredictable, depending on the electrical and thermal properties and geometry of the die set configuration.

Given that in the temperature range under investigation (specimen temperature < 1550 °C), the specimen temperature in the center, $\theta_{\text{center}}$, exhibits an almost linear correlation with $\theta_{\text{surface}}$, it is possible to derive calibration curves with a minimal set of internal temperature measurements when using a pyrometer. A single calibration test with thermocouples embedded in a solid cylinder made of a predensified or a fully dense compact will suffice to predict the temperature difference, $(\theta_{\text{center}} - \theta_{\text{surface}})$. As long as the die geometry remains the same, a single calibration may be sufficient, reducing the calibration effort substantially.

There are two main implications of the observed temperature difference between die surface and specimen center:

(a) Comparisons between FAST and conventional hot pressing or HIPing have not been done on the basis of die surface temperatures. Given that the temperature gradients in conventional hot pressing are lower than those
observed in FAST due to the slower heating, it is possible that some of the claimed advantages of FAST are actually due to the higher temperatures in the specimen;

(b) The presence of significant temperature gradients in FAST implies that great care should be taken with larger or complex shape specimens. Further developments in the modeling approach are particularly important for the proper usage of FAST for parts with complex geometries.

The design of FAST operations for large or complex shape specimens would benefit from accurate finite element calculations. Simple design and geometry changes can help to reduce the gradient inside the specimen by balancing Joule heat generation and heat transfer. Specifically, the following measures may be considered:

(a) Minimizing heat losses of the die by using thermal insulation on the die surface.

(b) Reducing interface resistance between die and punches by avoiding the trapping of powder particles, providing good surface finish where the punch and die are in contact or using a highly conductive liner.

(c) More complex arrangements of the die set and the use of thermal insulation can be modeled by finite element analysis in order to tailor the current path and Joule heat generation for a particular material, specimen size and geometry.
The effect of heating rate on the temperature difference

The evolution of the microstructure of conducting and insulating powders are different at various heating rates. The effect of heating rate on the densification and grain growth of $\alpha$-$\text{Al}_2\text{O}_3$ and MoSi$_2$ powder using FAST has been studied by Stanciu et al. [3.17]. The study showed that, for alumina, which is an insulating material, the final grain size was inversely proportional to the heating rate. While for the electrically conductive MoSi$_2$, the grain size was independent of the heating rate. Fast heating rates favor densification over grain growth in coarsening sensitive systems. The systems are characterized by a higher activation energy for diffusion than for grain growth, such as Al$_2$O$_3$ and ZnO. A high heating rate will not allow surface diffusion to coarsen the structure at low temperatures. Instead, the sintering powders are quickly taken to high temperatures where densification becomes predominant. Temperature plays the most important role in alumina coarsening. In contrast, no dependence of grain size on heating rates was observed in MoSi$_2$ [3.17]. Therefore, it is important to understand the temperature of the specimen at different heating rates.

Simulation of FAST of solid graphite cylinder specimens at heating rates of 25, 50, 110, and 220 $^\circ\text{C}/\text{min}$, respectively, were carried out and compared with experiments to investigate the possible effect of heating rate on the actual specimen temperature. Figure 3.10 shows that the heating rate, for both simulation and experiments within the specimens, was linearly proportional to the heating rate on the die surface. The actual heating rate was higher than the die surface temperature of 70%. The linear relation between the heating rate in the specimen and on the die
The effect of resistivity of specimen on the distribution of electric and thermal field

In this section, numerical simulations compared with experiments were reported for specimens made from alumina powders with initial and final heights of alumina of 8 mm (RD ~ 0.5) and 6 mm (RD ~ 0.67), respectively and fully dense graphite. For comparison with graphite, unless specified, the results of alumina at the end of sintering are presented.

Figure 3.11 shows the evolution of overall resistance of the system (including the punches, spacers, etc.). There was a discrepancy in the initial sintering stage. For alumina, it was attributed to the presence of nano-size alumina powders at the interface between the die and punch during filling. The contacts evolved with increasing temperature. Note that the corresponding predicted resistance of the graphite cylinder was in good agreement with experimental data.

Figures 3.12 to 3.15 show the electric field at stage B, as defined in Figure 3.8. Although the magnitude of the electric field varied during the process, the pattern of the electric field was practically independent of time for the specific application simulated here. Figure 3.12 shows the electric potential gradient in the system and indicates clearly that a large potential difference existed across the specimen for insulating powders. There was a minor radial variation of electric potential gradient (~ 50 V/m) in the non-conductive powders. This condition is often claimed to cause micro spark or plasma generated at particle contacts. The potential difference along
the alumina specimen height (6 mm) is about 10 percent of the total voltage applied to the system (~0.3 V). For the solid graphite cylinder, the potential difference accounted for only approximately 3 percent of the total voltage due to its low resistivity.

The information on electric field in the specimen made it possible to examine the physical mechanisms that enhance the densification in FAST. Even though several interpretations have been suggested [3.18], it is speculated that three major mechanisms are involved: (a) field emission; (b) voltage breakdown or discharge; and (c) thermal breakdown. These mechanisms will be discussed in detail below.

Field emission refers to the emission of electrons that are stripped from parent metals or semiconductors in the presence of an applied or induced electric field. The electric field is about 100 V/m across the alumina powders and 10 V/m for the graphite cylinder, which is not high enough to activate emission (~10⁶ V/m). Geometric field enhancement makes emission possible when acicular particles, geometric asperities at the particle surface or nanosize particles are present. A field enhancement factor, \( \beta \), can be used to define the localized electric field [3.19]:

\[
E = \beta \frac{U}{d}
\]  

(3.17)

where \( U \) (V) is the voltage difference and \( d \) (m) is the distance between two particle centers close to each other. \( U/d \) is the electric field with no geometric enhancement and it can be estimated from simulation (100 V/m for alumina powders and 10 V/m for graphite cylinder). When nano-size particles are considered and two neighboring
particles are close together (e.g., the ratio of the distance between the surfaces to the particle radius is about $10^{-4}$), then the geometric enhancement factor could be as high as $10^9$.

In addition to geometric enhancement, it also appears that the emission is affected by absorbed gases [3.20]. The strength of emission changes as gases are adsorbed and desorbed. The renewed adsorption of gases often reactivates the emission after a thermal cycle. Therefore, the effects of pulsed current frequency and processing atmosphere are worth investigating in further studies.

The electric field was not high enough to breakdown vacuum (the dielectric strength of vacuum is larger than $10^6$ V/m). However, both the geometric factors as discussed above, contaminants and defects can enhance the field strength [3.21]. Surface contaminants are capable of altering the activation energy for breakdown. Plasma could also be produced by electron impact ionization of neutral gases which can melt particle surfaces and/or an entire particle.

The high contact resistance at the contact generates Joule heating when current flowed into the particle. Accumulation of Joule heat between two touching particles, if heat can not be dissipated quickly away from contact, could induce thermal breakdown of the contact.

In summary, it is believed that nano size insulating powders have the highest probability of micro spark occurrence. This is consistent with the observations of Ozaki et al. [3.22] who investigated the evolution of the current during FAST of different size aluminum powders ($<$10 µm and $>$100 µm, respectively) using BN (electrically insulating) and graphite (electrically conductive) dies. Discharge or
breakdown was detected by observing a sudden current increase in the system (> 600A) when fine aluminum powders (10 µm) were sintered using a BN die. No discharge was observed when a graphite die was used since it could not sustain a large electric field across the specimen.

The electric current density, as shown in Figure 3.13, was higher in the punches and exhibited a maximum at the outside rim as a result of the large cross sectional area of the die. The specimen and the die were essentially a parallel electric network. The current path depends primarily on the ratio of resistance of specimen to die, i.e., the electric conductivity of the die and specimen, and the conductive cross sectional areas of each component. Because the graphite specimen is equally conductive with the die, the majority of the current (~ 80%) was diverted to the die which had a larger cross sectional area. For a less electrically conductive specimen, either due to high resistivity (e.g. alumina) or for a loose powder, practically all the current would be diverted into the die.

Figure 3.14 shows the magnitude of the horizontal and vertical components of the electric current in the punch/die assembly with a graphite cylinder in the place of the specimen. Results showed that the direction of the current was essentially in the axial direction. The only area with a current flow in the transverse direction was at the top of the punch and at the contact between the punch and the top surface of the die. Some limited current flow occurred along a graphite foil on the top and bottom of the specimen due to the high conductivity of the foil along its plane, but it cannot be seen for the contour levels selected. The component of current density normal to the
punch/die interface was continuous as required. There is no reason for the component of current density tangent to the interface to be continuous across the interface.

Figure 3.15 shows that the amount of Joule heating in the punches was about 16 times higher than the corresponding Joule heating inside the specimen at the center of the die. The electric conductivity of the specimen did not affect significantly this result for the geometry selected. Because the majority of Joule heat generation occurred at the punches, the specimen was indirectly heated by thermal conduction from the punches. The graphite foil also exhibited a relatively large Joule heating, but its volume was small and the resulting effect on the spatial temperature variation was limited. It is noteworthy, however, that the graphite foil provided slightly better temperature uniformity.

Both electric current and Joule heat are solely dependent on the local resistivity distribution in the system. Similar to electric current, Joule heating (Figure 3.15) was highest at the ends of the punches due to the small diameter of the punches and the resulting high resistance. Noticeably, Joule heat was zero within the specimen for alumina and very small for the graphite cylinder. Figure 3.13 shows that essentially no current passed through the alumina specimen and Joule heating in the specimen was practically zero. For the graphite cylinder, even though a certain amount of current flowed through the specimen (Figure 3.14), the resulting Joule heat was relatively small due to the small resistance of the specimen.

The effect of electrical conductivity of the specimen on the Joule heating was further investigated by performing a parametric study by varying the conductivity of the specimen, for the geometry and condition of experiment described in Section 3.3.
Figure 3.16 shows that Joule heating in the specimen accounted for less than 1 percent of the total Joule heating in the system. The Joule heating was only generated when the electric conductivity of the specimen was very close to that of graphite for the specific geometry examined. Again this was consistent with the maximum power transfer theorem [3.23], as discussed in Chapter 2.

Without heat transfer (conduction, radiation), the temperature increase due to Joule heating was proportional to the local resistivity. Heat transfer modifies the initial temperature distribution based on Eq 3.1. The temperature distributions within the die, punches and specimen are shown in Figure 3.17 for two instances illustrated in Figure 3.8, just after the beginning of heating and at the late stage of the heating. As discussed above, regardless of the electrical conductivity of the specimen, the punches generated the majority of the heat. The highest temperature in the system developed in the punches during the early heating stages. The heat was generated partially conducted into the specimen and partially lost in the machine (upper and lower graphite spacers) which were water cooled. As time progresses, the temperature in the specimen increased due to thermal conduction from the punches. In addition, surface radiation was a secondary heat loss for the specimen/die assembly. This pattern of heat flow resulted in a temperature differential between the die surface and specimen center with the specimen center at a higher temperature than the control temperature on the surface of the die. For this simulation the maximum predicted difference between the center of the specimen and the surface was about 100 °C [3.24].
Significant temperature gradients were found in the radial direction for electrically conductive and insulating specimens because of the heat flux pattern, i.e., conduction from the punch and heat loss through the die towards the surface of the die. It was more pronounced during the initial stages of sintering due to the relatively low density of the specimen. Compaction and reduction in the aspect ratio of the specimen may improve the temperature homogeneity (see Chapter 4 for more discussion).

The heat flux vectors during the heating stage is plotted in Figure 3.18. The observed pattern was consistent with the previous discussion on temperature evolution. Calculation when the die surface temperature is around 700 °C showed that only about 15% of the total electrical energy input was effectively used to heat up the specimen (heat flux into the specimen). More than 2/3 of the total energy was lost to the water cooled rams through heat conduction. The radiation from the die end, die side and punch end accounted for 3%, 15% and 3%, respectively. The relative importance of the various heating loss routes is shown in Figure 3.19 for various temperature levels. At low temperature heat conduction through the loading train is the main heat loss path. The role of radiation becomes more important at higher temperatures, as intuitively expected. Therefore, it has to be controlled for very high temperature applications.

*The effect of electric gap conductance on the energy efficiency*

The presence of contact resistances also implied localized Joule heating at those interfaces. Various levels of contact resistance correspond to different spatial
distributions of power. Low contact resistance, especially at the punch/spacer interface, would be associated with increased heating away from the specimen, i.e., reduced efficiency.

Figure 3.20 shows the total energy input into the system (normalized by $mC_p\Delta T$, temperature ramp from 25 °C to 1100 °C) as a function of electric gap conductance. There was a transition of energy efficiency in the range of electric gap conductance of $10^4$ (Ωm$^{-2}$)$^{-1}$. Above this level, the conversion of electric energy was not most efficient. Below this level, as discussed before, there was a higher demand on the electric power.

Modification of gap conductance can be achieved by using interfacial materials. Interfacial materials between contacting interfaces may be deliberately introduced to either increase or reduce the contact resistance. Examples include foils, wire screens, metallic and other coatings, greases and lubricant films. A summary of materials and corresponding properties is available at [3.6].

### 3.4 Conclusions

In this chapter, a coupled thermal-electric-densification framework for the design and optimization of FAST-type processes has been proposed and implemented into a finite element package (ABAQUS). A detailed evaluation of the accuracy of the model has been conducted for electrically conductive and insulating materials. The evaluation, compared with experimental measurement, indicated that the model
provided reasonably accurate predictions of the thermal and electric responses and pattern of thermal transfer.

It was found that the typical specimen encountered a higher temperature than that of the surface of the die (~ 100 °C), which is often used as a reference temperature. Therefore, the often quoted reduction in the sintering temperatures with respect to other techniques such as hot pressing or HIPing can be at least partially attributed to the lower temperatures measured on the surface of the die than that actually exist within the specimen. Both experimental and numerical results showed that an almost linear correlation existed between the die surface temperature and the inside temperature. This correlation allows the calibration of the temperature inside the specimen against the die surface temperature.

Simulation showed that, for sintering of alumina powder, almost all the electric current was diverted into the die. In contrast, approximately 20 percent of the total electric current flowed into the graphite specimen. Despite this, the specimens are mainly heated by the Joule heat generated at the punch ends, regardless of specimen’s electrical conductivity. As a result, the temperature field in the punch/die assembly basically does not depend on the electrical conductivity of the small specimen. It is noted that the transition of the specimen to large and complex shapes is not trivial. It is expected that the method presented here should be indispensable for the design of such operations.
Figure 3.1 Geometry of the SPS machine and the corresponding finite element mesh (right) and schematic boundary conditions (left, see later for details) for the simulations. Curved arrows represent potential radiation. Contact resistances are also marked. $\theta_{\text{surface}}$ is the controlling temperature. $\phi$ is the controlled potential applied to the system. The letters A - G refer to the components specified in Table 2.1. Letter H refers to the graphite foil (UCAR Carbon Company Inc., diameter 19.1 mm, thickness 135 $\mu$m). Voltage difference is applied at the end of component A (top and bottom).
Figure 3.2  Experimental curve for the displacement of the punch during sintering of alumina powders (after considering the thermal expansion effect in the system). Step-wise displacement is used in the simulation. Mapping of temperature from each step to the next is performed.
Figure 3.3 Temperature or voltage drops at an imperfect interface with apparent contact area $S_A$ due to thermal contact resistance or electrical contact resistance as heat, $\dot{q}_c$, or electric current flux, $J$, flows across the interface, respectively. Joule heat, $\dot{q}_{ec}$, is also generated and evenly distributed into two parts. $\dot{q}_1$ and $\dot{q}_2$ are the heat fluxes in part 1 and 2 respectively.
Figure 3.4  Temperature dependence of bulk graphite properties [3.10]. Density is 1850 kg/m³ [3.10].
Figure 3.5 Thermal and electrical properties of fully dense alumina [3.11]. Density is 3980 kg/m³ [3.11].
Table 3.1  Orthotropic material properties of graphite foil [3.12, 3.13].

<table>
<thead>
<tr>
<th>Property</th>
<th>Direction</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal conductivity</td>
<td>In-Plane</td>
<td>140 W/m-K</td>
</tr>
<tr>
<td>Thermal conductivity</td>
<td>Thickness</td>
<td>5 W/m-K</td>
</tr>
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<td>In-Plane</td>
<td>$111111 \ (\Omega m)^{-1}$</td>
</tr>
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<td>Electrical conductivity</td>
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<td>$10204 \ (\Omega m)^{-1}$</td>
</tr>
<tr>
<td>Specific heat</td>
<td>In-Plane and Thickness</td>
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</tr>
<tr>
<td>Density</td>
<td>In-Plane and Thickness</td>
<td>1120 kg/m$^3$</td>
</tr>
</tbody>
</table>
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Figure 3.18  Vector plot of heat flux (HFL) of alumina powders (left) and graphite cylinder (right) at stage B in Figure 3.8. The length and density of vectors express the intensity of heat flux. 1: Conduction loss to machine spacer; 2: Radiation loss through die top; 3: Radiation loss through die wall and 4: Radiation loss from punch end.
Figure 3.19  The relative contributions of various mechanisms of heat loss versus the controlling temperature.
Figure 3.20 Effect of electric gap conductance on energy efficiency. Energy input is normalized by $mc_p\Delta T$. Temperature ramp is from 25 to 1100 °C. Note that the x-axis is a log scale. Contact resistance increases to the left.
CHAPTER 4. DISCRETE FINITE ELEMENT MODEL

4.1 Introduction

In Chapter 3, it was shown that in FAST the specimen is not heated by Joule heating within itself, even if the material is electrically conductive. The specimen is actually heated through heat conduction from the punches. Because most materials sintered using FAST are rather hard to densify under the applied pressure alone (approximately 10 ~ 50 MPa), their thermal diffusivity is determined by their high porosity in the early stages of the process. For a rapid process like FAST, if larger and more complex larger samples are to be considered, understanding the thermal conduction of the compact is essential. In addition to the specific problem in hand, thermal properties of low relative density materials are important in other fields such as sintering [4.1], selective laser sintering (SLS) [4.2], microwave sintering [4.3], and powder beds under low pressure [4.4].

Many researchers have attempted to measure or model the effective thermal conductivity of particulate materials. In [4.5 – 4.8], the effect of porosity and size on the effective thermal conductivity was investigated. Hadley [4.9], Tehranian et al. [4.4] and Gorbis et al. [4.10] investigated the effect of applied external loads on powder bed conductivity. Bounds method has been applied to derive the effective thermal conductivity of mixture of two phase composites [4.11]. Mean field or Effective Medium Theory (EMT) was also used to calculate conductivity [4.12]. The complexity of derived or proposed expressions depends on how much information on
pore structures is taken into account. A recent summary of these expressions is available in [4.13]. Argento and Bouvard [4.14] studied the conductivity evolution of monosize spheres during isostatic densification. Conductivity evolution between two particles was first calculated by finite element method and was used to derive an expression relating effective conductivity and relative density. This study only considered densification under isostatic stress condition.

Most of the thermal conduction work has been limited to compacts under either isostatic stress or to thermal properties in axial direction under die compaction. The result may not be applicable to large and complex shape compacts which experience non-isostatic stress. Compaction under non-isostatic stresses produces a microstructure which, at least geometrically, has directional properties. Here we are interested in the anisotropy of thermal properties.

In this chapter, the effect of applied pressure on the thermal properties of partially compacted powders is studied. In addition to the usual examination of their dependence on relative density, special attention is given to the potential of anisotropy as a result of the application of non-isostatic stress.

### 4.2 Method

In general, compared to high relative density problems, the understanding of the thermal properties of powder compacts close to their packing density is incomplete, due to the importance of the role of contact geometry and thermal
behavior of the contacts. Homogenization techniques need to incorporate more information on microstructural geometry while bound approaches [4.11] are weak for this problem since the lower bound is zero.

For this purpose, the Discrete Element Method (DEM) is more appropriate because it takes into account the connectivity of powders in a compact. It is well known that the distribution of external forces in the compact is highly inhomogeneous [4.15]. As a result, the thermal conductivity of the compact is inherently related to the applied stress that produces the microstructure. Moreover, it is now accepted that non-isostatic stresses result in a morphological anisotropy, which in turn is at least partially responsible for the observed anisotropy of the mechanical performance [4.16]. It is intuitive that a similar anisotropy may also exist in the thermal response.

In this chapter, the coupling between the mechanical and thermal response and the possibility of thermal anisotropy due to non-isostatic stress is explored. To this end, a multiparticle finite element simulation is employed. This method does not make any compromise with respect to particle geometry or material behavior, but it is very computationally expensive. Thus, the discussion is restricted to two-dimensional (2D) problems. Here we present preliminary results from simulations with uniform size circular particles. An attempt to demonstrate experimentally the anisotropy in conductivity of die compacts is also presented.
4.3 Simulation setup

The coupling of pressure with the thermal properties of compacted powders is studied by a two-step simulation approach as shown in Figure 4.1: (a) a 2D compaction simulation was performed using Discrete Finite Element Method (DFEM), a multiparticle finite element analysis; (b) the predicted geometry of the compact was used for a thermal analysis that predicted the thermal conductivity of the material. Details of the simulation procedure are given below.

4.3.1 Mechanical compaction

Discrete Finite Element Method and its advantages in studying the compaction of powders are discussed in detail in [4.17, 4.18]. For completeness, only the aspects of the simulation that are relevant to this work are reported here.

The analysis presented here uses finite elements and examines the behavior of 400 particles under macroscopic stress. Each particle is discretized into a finite element mesh as shown in Figure 4.2, which consists of 67600 4-node plain strain reduced integration elements, CPE4R [4.19], with a specially designed grid that is denser in the periphery in order to properly simulate the interparticle contacts. The perimeter of each particle is composed of 72 nodes. Each particle has 169 nodes and 132 elements. The model was implemented in ABAQUS/Explicit [4.19]. Further discussion about this geometry and its effectiveness for simulating compaction is
presented in [4.17, 4.18, 4.20]. While many shapes can be examined by this technique, results on circular particles are presented here.

An initial isotropic configuration is achieved by: (a) generating a random configuration of particles at a low density (relative density is about 0.4 for 2D problem) by placing particle centers to random positions selected by a random number generator within a predescribed box. Rigid dies are then used in a finite element simulation to “densify” the particle collection. The finite element analysis uses the same conditions as the subsequently described compaction simulations. This “precompaction” step is carried out until the onset of plastic strain at one of the particles. At this point the simulation is stopped (relative density is 0.764) and this is considered to be the initial configuration. This initial configuration is indeed isotropic. It is verified that, in the subsequent mechanical and thermal simulations under isostatic stress, the resulting properties remain isotropic.

To study the effect of compaction on thermal properties, representative microstructures are generated at different relative densities under closed die and isostatic compactions. Each particle is considered to be elastoplastic with a Young’s modulus to yield strength ratio of 100 and Poisson’s ratio of 0.3. Mass scaling [4.19] has been used to accelerate the simulations.

The densification simulation is free of the simplifying interparticle assumptions which are typical in classic DEM simulations. Interactions between particles and dies are assumed to be frictionless. The interparticle friction is modeled by a Coulomb model with a coefficient of friction of 0.5. A maximum shear stress $\tau_{\text{max}} = \sigma_Y/\sqrt{3}$ is imposed to simulate sliding by yielding on a thin surface layer of the
powders when the shear stress on the surface is high. This condition prevents the
development of high shear stresses that are predicted by the Coulomb friction law
under high stresses normal to the contact. The same interparticle behavior is imposed
on all potential contacting particle pairs. Due to the rearrangement of particles during
compaction, the actual contact pairs are not known \textit{a priori}. Therefore all particle
pairs initially within a distance of five particle radii are continuously checked for
contacts.

Compaction is achieved by moving rigid surfaces until the powder reaches
near full density, as shown in Figure 4.3. The ratio of kinetic to potential energy is
controlled to be less than 5\% to avoid any inertia effects that may be exaggerated by
mass scaling.

4.3.2 Thermal property extraction

The effective thermal conductivity of compact, $k_{\text{eff}}$, is determined using
Fourier’s diffusion law

$$\dot{q} = -k_{\text{eff}} \nabla \theta$$

(4.1)

where $\dot{q}$ is heat flux (W/m$^2$) and $\theta$ is temperature (K).

The thermal conductivity of fully dense material, $k_s$, is defined as

$$\dot{q}_s = -k_s \nabla \theta$$

(4.2)
Then, for a fixed temperature gradient, the normalized effective conductivity is

\[
\frac{k_{\text{eff}}}{k_s} = \frac{\dot{q}}{\dot{q}_s}
\]

Intermediate mesh configurations were obtained from the compaction simulation and were used for the thermal simulation which provided data for the calculation of the thermal conductivity as shown in Figure 4.1. Steady-state heat transfer analysis with 67600 diffusion continuum 2D 4-node elements, DC2D4, in ABAQUS/Standard [4.19] was employed.

A temperature difference imposed on the nodes of the top and bottom layers of particles shown in Figure 4.4. In order to remove the wall effect, the boundary temperatures were assigned to the nodes within the following ranges for conductivity in compaction direction:

\[
y_{\text{max}} > y > y_{\text{max}} - 3R \text{ (on the top layer)} \tag{4.4a}
\]

\[
y_{\text{min}} < y < y_{\text{min}} + 3R \text{ (on the bottom layer)} \tag{4.4b}
\]

where R is the radius of the particle. \(y_{\text{min}}\) and \(y_{\text{max}}\) are minimum and maximum y coordinates, respectively.

Similarly, in the transverse direction, the temperature difference was applied to the left and right layers, respectively:
\[ x_{\text{max}} > x > x_{\text{max}} - 3R \text{ (on the right layer)} \quad (4.5a) \]

\[ x_{\text{min}} < x < x_{\text{min}} + 3R \text{ (on the left layer)} \quad (4.5b) \]

where \( x_{\text{min}} \) and \( x_{\text{max}} \) are minimum and maximum x coordinates, respectively.

For the fully dense material of the same dimension, the heat flux that sustains the same \( \Delta \theta \), in the compaction direction, is

\[ \dot{q}_s = -k_s \frac{\Delta \theta}{H} \quad (4.6) \]

where \( H' = H - 6R \); \( H \) is the height of the compact.

In ABAQUS, the thermal contact interaction between particles was modeled by a gap conductance, \( h_g \) (W/m\(^2\)-K), and gap clearance, \( d \) (m) as shown in Figure 4.5. A parametric study of conductivity with different values of gap conductance and gap clearance showed that thermal conductivity is independent of gap conductance \( h_g \) over a relatively large range from \( 10^4 \) to \( 10^8 \) W/m\(^2\)-K. Therefore, the gap conductance \( h_g \) is set to be \( 10^8 \) W/m\(^2\)-K in this study. The need of using a finite gap clearance over which conduction is allowed when two neighboring nodes lie within the gap comes from the corresponding tolerance of the mechanical compaction simulation. A small gap clearance may result in lack of heat transfer at contacts that are mechanically closed. A large gap clearance artificially enlarges the contact area. Using a parametric study, the best compromise is a value for the gap clearance of 0.5% of the particle
diameter. The number should be adequately small to avoid exaggeration of the contacts but at the same time it should be large enough to be insensitive to the tolerance of the mechanical contact discretization.

4.4 Experimental procedure

In order to validate the simulation, the electric conductivity of pressed aluminum powders is measured. Mathematically, the electric conductivity is exactly the same as the thermal counterpart under steady state condition [4.21]. Aluminum powder (Valimet, Inc., Stockton, CA) was die-compacted using an Instron (Model 1127) at various pressures from 150 MPa to 380 MPa. Zinc stearate in an acetone solution was used as a die-wall lubricant. No lubricant was applied within the powder. Figure 4.6 shows the typical morphology of the initial powder. The powders have a spherical shape with an average particle diameter of 500 µm. Following die compaction, four-point probe measurements (Cascade® C4S-57, MicroTech) [4.22, 4.23] were carried out on the top and side of the compact, as illustrated in Figure 4.7.

4.5 Results and discussion

4.5.1 Mechanical compaction

A detail of the deformed mesh at a relative density of 0.93 is shown in Figure 4.8(a). Under high relative density and high constraint (coefficient of friction 0.5), the rearrangement of particles is limited. In die compaction, particles are more
compressed in the axial direction than the transverse one. Many particles exhibit near ellipsoidal shapes with short axes along or close to the compaction direction. Similar particle shapes are observed in the pressed compact as shown in Figure 4.8(b).

In isostatic compaction, particles indent each other with less rearrangement than in die compaction because they are constrained by neighbors from all directions. As a result, the particles remain more or less equiaxed. In contrast, particle contacts in the direction of die compaction are more heavily deformed.

The evolution of the average coordination number \([4.20]\), defined as the average number of contacts per particle is plotted in Figure 4.9. The average coordination number is shown to be a monotonically increasing function of relative density and does not depend on the mode of compaction.

The normalized macroscopic compaction stress, \(\Sigma/\sigma_Y\), for isostatic and die compaction as a function of relative density is shown in Figure 4.10. For both isostatic and die compaction, the resultant stresses increase with relative density. The stress in the die compaction direction is higher than the isostatic case at the same relative density. This is consistent with previous modeling \([4.24]\) and experimental observations \([4.25]\).

### 4.5.2 Evolution of effective thermal conductivity

Figure 4.11 shows that, for isostatic compaction, conductivity does not depend on the direction. A similar dependency of conductivity on the relative density is observed for both die and isostatic compactions. The only difference is that, for die
compaction, conductivity along the die compaction direction is higher than isostatic compaction while that in the transverse direction it is lower than the isostatic case.

The anisotropy of conductivity can be expressed as the ratio of conductivity along the compaction direction over that along the transverse direction. This ratio is shown in Figure 4.12. For isostatic compaction, there is practically no anisotropy. In contrast, in die compaction, the conductivity ratio exhibits two distinct stages. At lower relative density (RD < 0.81), the ratio increases with the densification. At a relative density of 0.81, the anisotropy reaches maximum with the conductivity along the die compaction direction 20% higher than in the transverse direction. At higher relative densities (0.81 < RD < 1), the ratio decreases with increasing density.

In the early stage, the ratio of radial to axial stress is very small. As a result, contacts along the compaction direction are larger than in other direction. This explains the early rise of the conductivity ratio. As densification progresses, first the ratio of radial to axial stress increases. More importantly the cross section of the particles in the transverse direction increases. These two effects contribute to the reduction of anisotropy as densification progresses.

It is believed that the anisotropy would be larger in 3D. This is supported by preliminary calculations in 2D and 3D unit cells. Because this anisotropy appears to be present in the early densification stages, it is expected to be of practical importance for hard powders, e.g. tungsten or ceramics, which do not densify significantly under cold pressing before sintering.

It is also believed that this anisotropy may be even higher at lower triaxialities (i.e., under compaction with less hydrostatic component than die compaction).
Experimental measurement of electric conductivity (Figure 4.7) also shows that anisotropy develops in die compaction. The measured conductivity ratio is given in Figure 4.13. There are no data available at lower relative density due to low strength of the green part. The results, however, shows clearly that anisotropy of conductivity exists at intermediate relative density and is reduced as it approaches near fully dense. Of course the surface oxide present on aluminum powder will also affect the level of anisotropy.

Compaction not only increases the effective thermal conductivity, but also modifies the heat flux distribution. Figure 4.14 shows the evolution of heat flux normalized by the maximum heat flux at fully dense (0.035 W/m³ in this study). At lower relative density, only a few conductive paths exist for heat transmission through the compact. At higher relative density, large contact areas result in a more homogenous heat flux. Therefore, application of stress will make the heat flux and the resultant temperature field more uniform within the specimen.

4.6 Conclusions

The evolution of the thermal conductivity of compacts under different compaction modes, i.e., isostatic and die compactions, has been studied. The simulations show that the effective thermal conductivity increases with relative density, as expected. However, anisotropy of conductivity develops in the specimen under die compaction. For the 2D circular powders with isotropic intrinsic
conductivity, the effective conductivity of the compact in the compaction direction is higher than the transverse one and it may be as high as 20\%. Experimental measurements also confirmed the existence of an anisotropy of conductivity in a die compacted specimen.

Conductivity is determined by the microstructure that evolves according to the processing route. In isostatic compaction, it is reasonable to assume that all of the contacts are of similar character. In die compaction, the anisotropy is due to the combination of the number of paths along which the heat is conducted and the average contact size along these paths which are higher in compaction direction.

The ramification of these results is that relative density is not the only internal variable describing the state of compaction in non-isostatic stress state. Even though both compacts have the same volume fraction, their effective properties depend on microstructural information beyond that contained in the volume fraction of pores.
Figure 4.1 Schematic of the procedure of electric conductivity measurements of deformed compacts.
Figure 4.2  (a) Discrete Finite Element Method (DFEM) mesh of 400 particles; (b) localized deformed mesh.
Figure 4.3 (a) Undeformed mesh; (b) deformed mesh after isostatic compaction; (c) deformed mesh after die compaction.
Figure 4.4 A temperature difference is imposed on the nodes of the top and bottom layers of particles in the compaction direction.
Figure 4.5 Heat can be transferred between two particles if the distance is within the gap clearance, \( d \). The heat transfer property between the two particles is defined by the gap thermal conductance, \( h_g \).
Figure 4.6  SEM image of as-received aluminum powders.
Figure 4.7 Schematic of four-point probe measurement of conductivity of compact in the transverse and axial directions.
Figure 4.8  (a) FEM mesh of compact under uniaxial compaction (2D, RD ~ 0.93).  
(b) SEM image of side view of pressed compact (3D, RD ~ 0.77). The arrows denote the compaction direction.
Figure 4.9  Evolution of average coordination number during compaction.
Figure 4.10 Evolution of compaction stress normalized by yield strength of powder.
Figure 4.11 Evolution of effective thermal conductivity normalized by the thermal conductivity of the powder.
Figure 4.12  Evolution of the ratio of the effective thermal conductivity in the axial direction to that in the transverse direction (simulation, 2D).
Figure 4.13  Evolution of the ratio of the effective electric conductivity in the axial direction to that in the transverse direction (experiment, 3D).
Figure 4.14 Evolution of heat flux (normalized by maximum heat flux at fully dense) at various relative density during isostatic compaction.
CHAPTER 5. CONCLUSIONS AND FUTURE WORK

We have formulated a coupled thermal-electric-densification framework for the design and optimization of FAST-type process and implemented into a finite element package (ABAQUS). This model includes temperature and porosity dependent thermal and electric properties which couple the thermal and electric models together with Joule heating. Additional features of the framework include (a) a temperature closed-loop controlling algorithm has been implemented into the finite element package (ABAQUS). The temperature control function, which is actually employed in the FAST-type system, is realized (see Appendix A); and (b) a procedure to characterize electric and thermal gap conductances has been developed, which is critical to proper prediction of temperature and electric fields (see Appendix B).

In addition to the general framework, an approximate lumped resistance model is proposed to facilitate quick simulations of FAST-type process. This procedure yields fairly reasonable predictions for electric current and Joule heating distribution.

A detailed evaluation of the accuracy of the models has been conducted for electrically conductive and insulating materials. The model predictions, compared with experimental measurements, indicate that the model provides a reasonably accurate prediction of thermal and electric response.

A key finding of this work is that a typical specimen encounters a higher temperature than that of the surface of the die (~ 100 °C), which is often used as a reference temperature. Therefore, the often quoted reduction in the sintering temperatures with respect to other techniques such as hot pressing or HIPing can be at
least partially attributed to the lower temperatures measured on the surface of the die than those that actually exist within the specimen.

Another important result of the simulation is that heating of the specimen is mainly done by conduction from the hot punches rather than Joule heating within the specimen. This is due to the fact that majority of current goes around the specimen not only for insulating material (such as alumina) but also for conductive ones. Even for fully dense graphite cylinder in the place of the specimen, only approximately 20% of the current goes through the sample for the geometry examined. For lower relative densities even conductive specimens, electric current diverts into the die due to its large cross sectional area. The only possible way to increase the Joule heating within conductive specimens is to insulate the punch/die interface. Therefore the resulting temperature distribution in the FAST cases examined is nearly independent of the electric conductivity of the powder material and is determined by the effective thermal conductivity of the sample which governs the conduction of heat from the neighboring punches.

To understand the correlation between effective thermal conductivity and densification, finite discrete element method is employed. In addition to the usual examination of conductivity dependence on relative density, we found that an anisotropy of conductivity exists as a result of the application of non-isostatic stress. For 2D circular powders with isotropic intrinsic conductivity, the effective conductivity of the compact in the compaction direction is higher than the transverse one and it may be as high as 20%. This was qualitatively confirmed by experimental observations. This anisotropy appears to be present in the early densification stages.
Therefore it is expected to be of practical importance for hard powders, e.g. tungsten or ceramics, which do not densify significantly under cold pressing before sintering.

A key issue that needs to be addressed in the future is that present simulation of FAST lacks a sintering model that would be fully coupled with the thermal and electric models. In our work, the effect of sintering was taken into account in a rough and approximate way, i.e., by assuming that density within the specimen is uniform and as a result it can be estimated from the experimental data of punch displacement. Such an approach was adequate for a flat cylindrical specimen but it could be erroneous for large and complex parts. In that case, a model such as the ones described in [5.1] and [5.2] should be introduced to the finite element codes. At this point, such a possibility is not available in ABAQUS.

Our current efforts in modeling the effective thermal conductivity have been focused on two-dimensional problems due to intensive computational requirements. Additional challenge to extend current method to three-dimensional problem arises from the meshing. To the best of author’s knowledge, a consistent meshing method, i.e., application of repeatable transition layers of mesh between coarse and fine meshes, using three-dimensional block elements, is still unavailable. A conceivable solution could be the calibration of the parameters in simplified contact laws using discretized two-particle unit cell model, or a small assembly of particles, under different deformation modes, e.g., compression and shear. This solution is similar to the work by Argento and Bouvard [5.3] in that (a) two-particle unit cell model provides the contact law under compression deformation; and (b) the calibrated
parameters are used in simplified contact law and to derive transport properties, e.g., thermal conductivity.
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C.6 http://www.mamat.com/hitemp.html
APPENDIX A. ABAQUS USER SUBROUTINE

C*****************************************************************************
C
C ABAQUS USER SUBROUTINES FOR
C IMPLIMENTATION OF CLOSED LOOP TEMPERATURE CONTROLLED
C VOLTAGE

C

*NOTES:
C 1. TWO USER SUBROUTINES ARE EMPLOYED HERE:
C    DISP - APPLY CORRECTED VOLTAGE
C    UVARM - READ CURRENT TEMPERATURE
C 2. TESTED WITH ABAQUS VERSION 6.2 OR 6.3
C 3. IN MAIN INPUT FILE, THE FOLLOWING IS REQUIRED
C    TO INVOKE THE USER SUBROUTINE:
C    *MATERIAL, NAME= XXX (WHICH TEMPERATURE IS READ)
C    *USER OUTPUT VARIABLE
C        1
C    *BOUNDARY, USER
C    NODE NUMBER (WHERE VOLTAGE IS APPLIED),9
C

*DEFINITAION OF VARIBLES:

C
C VOLTAGE -- VOLTAGE DATA BLOCK
C KINC_PRE -- INCREMENT NUMBER AT PREVIOUS INCREMENT
C
C TIM_PRE -- TIME AT PREVIOUS INCREMENT
C TIME0 -- STARTING TIME OF HEATING STAGE
C TIME1 -- ENDING TIME OF HEATING STAGE. IT IS ALSO
C         THE STARTING TIME OF HOLDING STAGE
C TIME2 -- ENDING TIME OF HOLDING PERIOD
C TEMP0 -- STARTING TEMPERATURE OF HEATING STAGE
C TEMP1 -- ENDING TEMPERATURE OF HEATING STAGE. IT IS
C         ALSO THE STARTING TEMPERATURE OF HOLDING STAGE
C TEMP2 -- ENDING TEMPERATURE OF HOLDING PERIOD
C TEMP10008 -- CURRENT TEMPERATURE AT CONTROLING POINT
C TEMP_CONTROL -- CONTROLLING TEMPERATURE
C GAIN -- THE COEFFICIENT DEFINING THE RATIO OF
C         VOLTAGE ADJUSTMENT TO TEMPERATURE ERROR
C U0 -- INITIAL VOLTAGE APPLIED ON THE SYSTEM
C U_PRE -- VOLTAGE AT PREVIOUS INCREMENT
C DU -- VOLTAGE ADJUSTMENT
C DTIME -- TIME PERIOD BETWEEN CURRENT INCREMENT
C        AND PREVIOUS INCREMENT
C*****************************************************************************

SUBROUTINE DISP(U,KSTEP,KINC,TIME,NODE,NOEL,JDOF,COORDS)
INCLUDE 'ABA_PARAM.INC'
DIMENSION U(3),TIME(2),COORDS(3)
COMMON /VOLTAGE/U_PRE, TIME_PRE, KINC_PRE
COMMON TEMP10008
PARAMETER (TIME0=172.D0, TEMP0= 580.D0,
#       TIME1=786.D0, TEMP1= 1100.D0,
#       TIME2=888.D0, TEMP2=1100.D0,
#       GAIN=0.1, U0=2.0)

C
IF (KINC.GT.KINC_PRE) DTIME=TIME(2)-TIME_PRE
WRITE(*,999), KINC, KINC_PRE, TIME(2),TIME_PRE,DTIME
999   FORMAT(5HXOXOX,6H KINC=,I5,6H KPRE=,I5,6H TIME=,F10.5,
#    6H TPRE=,F10.5,6H DTIM=,F10.5)
C
C Initialization
C
IF (KINC.EQ.1) THEN
U(1)=U0
U_PRE=U0
KINC_PRE=1
TIME_PRE=TIME(2)
TEMP10008=25
RETURN
ENDIF
C
C Control command for all other increments
C
print*, 'dtime=',dtime,kinc_pre,kinc,time(1),time(2)
IF (TIME(1).LT.TIME0) THEN
   TEMP_CONTROL=(TEMP0-25.)*TIME(1)/TIME0+25.0
ELSE IF ((TIME(1).GE.TIME0) .AND. (TIME(1).LT.TIME1)) THEN
   TEMP_CONTROL=(TEMP1-TEMP0)*(TIME(1)-TIME0)/(TIME1-TIME0)+TEMP0
ELSE IF ((TIME(1).GE.TIME1) .AND. (TIME(1).LE.TIME2)) THEN
   TEMP_CONTROL=(TEMP2-TEMP1)*(TIME(1)-TIME1)/(TIME2-TIME1)+TEMP1
ELSE
   TEMP_CONTROL=25.
ENDIF
C
C Voltage correction
C
IF (TIME(1).LT.TIME0) THEN
   DU=GAIN*(TEMP10008-TEMP_CONTROL)*DTIME/300.0
   IF (TIME(1).GT.(TIME0-40.)) DU=DU/2.
ELSE IF ((TIME(1).GE.TIME0) .AND. (TIME(1).LT.TIME1)) THEN
   DU=GAIN*(TEMP10008-TEMP_CONTROL)*DTIME/800.0
   IF (TIME(1).GT.(TIME1-40.)) DU=DU/2.
ELSE IF ((TIME(1).GE.TIME1) .AND. (TIME(1).LT.TIME2)) THEN
   DU=GAIN*(TEMP10008-TEMP_CONTROL)*DTIME/800.0
   IF (TIME(1).GT.(TIME2-40.)) DU=DU/2.
ENDIF
C Voltage application
C

IF (TIME(1).LT.TIME2) THEN
  U(1)=U_PRE-DU
  IF (U(1).LT. 0) THEN
    U(1)=0.D0
  ENDIF
ELSE
  U(1)=0.D0
ENDIF

IF (KINC_PRE+1.EQ.KINC) TIME_PRE=TIME(1)
IF (KINC_PRE.LT.KINC) THEN
  U_PRE=U(1)
  KINC_PRE=KINC
ENDIF
WRITE(*,101) TIME(1),KINC, KINC_PRE
WRITE(*,102) TEMP10008,TEMP_CONTROL,U(1),-DU
101   FORMAT(5HTime=,F10.5,5HKINC=,I5,' KINC_PRE=',I5)
102   FORMAT('Tcur=',F10.5,' Tctl=',F10.5,' New V=',F10.5,' -dU=',F10.5)
RETURN
END

SUBROUTINE UVARM(UVAR,DIRECT,T,TIME,DTIME,CMNAME,ORNAME,
1 NUVARM,NOEL,NPT,LAYER,KSPT,KSTEP,KINC,NDI,NSHR,COORD,
2 JMAC,JMATYP,MATLAYO,LACCFLA)

INCLUDE 'ABA_PARAM.INC'
C
CHARACTER*80 CMNAME,ORNAME
CHARACTER*3 FLGRAY(15)
DIMENSION UVAR(NUVARM),DIRECT(3,3),T(3,3),TIME(2)
DIMENSION ARRAY(15),JARRAY(15),JMAC(*),JMATYP(*),COORD(*)
COMMON TEMP10008

C The dimensions of the variables ARRAY and JARRAY
C must be set equal to or greater than 15.

IF ((NOEL.EQ.10008).AND.(NPT.EQ.4)) THEN
  CALL GETVRM('TEMP',ARRAY,JARRAY,FLGRAY,JRCD,JMAC,JMATYP,
1 MATLAYO,LACCFLA)
  TEMP10008=ARRAY(1)
  UVAR(1)=TEMP10008
  print*,TEMP10008=',TEMP10008
ENDIF
RETURN
END
APPENDIX B. PROCEDURE FOR CALIBRATION OF CONTACT RESISTANCE

The calibration of thermal and electrical contact resistance is necessary for the proper description of the interfaces in the FE models presented in Chapter 3. The gap conductances (electrical and thermal) can be identified by specialized equipment (e.g. [B1, B2]). As an easier alternative, a set of experiments was proposed that can be performed on the same SPS machine that is used for FAST, as described in Section 3.3.1. These experiments include three configurations shown in Figure B.1 for calibration of electric gap conductance:

(a) Single punch test: a punch placed between the spacers
(b) Double punch test: two punches placed between the spacers
(c) Dummy run test: a punch/die/punch assembly without a specimen

During these experiments, the voltage, current and temperature are logged. The temperature, as shown in Figure B.1, is logged at the middle of the outer surface of the punch, next to the interface between the two punches and at the surface of the die for the three tests listed above, respectively.

The estimation of the electric gap conductance is based on the following assumptions:

- The temperature dependence of the gap conductances (electric and thermal) is the same with the contacting material (in this system graphite). This may not be accurate at high temperatures since creep at asperities may increase the gap conductance. This is a very convenient assumption since it reduces the number of unknowns to one.
- Two values of electric gap conductances suffice, namely one for interfaces along the loading direction, $\sigma_g^h$, and one for the transverse direction, $\sigma_g^v$. This assumption reflects the fact that contact resistance depends on pressure, as shown in [B.3]. Because all simulations and experiments discussed in this work were conducted at a single level of load (15kN), this assumption is adequate, albeit not perfect, since the stress on the punch spacer and spacer/spacer contacts are not the same.

- Determination of electric gap conductance requires a combination of experiments and simulations. In these simulations, the thermal gap conductance is assumed to be essentially infinite. This assumption is based on the consideration that the determination of thermal gap resistance requires more complex experiments [B.1]. In this work, the electric field in the system is only a weak function of the thermal contact resistances. This justifies the assumption which is made only for the simulations used to calibrate the electric contact resistance.

Based on these assumptions, the flow chart of the procedure that determines the electric gap conductances $\sigma_g^h$ and $\sigma_g^v$ is shown in Figure B.2.

Subsequently, the experiment with the graphite cylinder was used to compute $h_g^h$ as shown in Figure B.1(d). The experimentally observed temperature difference between the die surface and specimen was fitted with the predictions from the simulation by selecting a value for $h_g^h$. The value of $h_g^v$ is determined simultaneously by assuming $\sigma_g^h / \sigma_g^v = h_g^h / h_g^v$. 
This procedure requires four experiments and is adequate for the simulation of FAST of powders with low electric conductivity. In this case, the electric contact resistance is not important since no current flows through the specimen. For higher electrical conductivity materials, this contact resistance must be evaluated. One possibility is to run a test with a fully dense compact made from the material to be sintered, then scale the contact resistance with the relative density. An alternative approximation is to assume that the contact between the punch and the compact is the same as that between the graphite punches. Here the derived values

\[
\sigma_g^h = 1.25 \times 10^7 \text{ (} \Omega \cdot \text{m}^2 \text{)}^{-1}; \quad \sigma_g^v = 7.5 \times 10^6 \text{ (} \Omega \cdot \text{m}^2 \text{)}^{-1}
\]  \hspace{1cm} (B.1)

\[
h_g^h = 2.2 \times 10^3 \text{ W/m}^2\text{-K}; \quad h_g^v = 1.32 \times 10^3 \text{ W/m}^2\text{-K}
\]  \hspace{1cm} (B.2)

were used in all simulations.

In summary, with four independent calibration experiments and several simulations, the calibrated gap conductances are identified. The power, temperatures and resistances of simulations are compared with experiments as shown in Figures B.3, B.4 and B.5. In terms of electrical resistance, the overall resistance of the system would be significantly underpredicted if contact resistances were ignored [B4, B5]. With the introduction of contact resistances at the axial contacts between punches, spacers, etc., the predicted overall resistance is still underpredicted. It is only when a transverse contact resistance at the interface between the punch and die is included, that predictions match the experimentally observed values.
This calibration method provided a satisfactory result. The simulated power, die surface temperature and resistance are in a reasonable good agreement with experimental data. The values of gap conductances depend on the specific system. In this FAST system, the contact conditions between different components are similar. Therefore, a minimum calibration effort (four calibration tests) is sufficient. In other complex systems, additional temperature and voltage measurements at different locations, next to the interface, may be required to retrieve reliable data.
Figure B.1 Schematic (left) and FEM mesh of calibration tests. (a) single-punch run; (b) double-punch run; (c) dummy run and (d) graphite cylinder. The arrows point to the locations where the temperatures are logged.
Figure B.2  Flow chart for determination of gap thermal/electric conductances in calibration tests.
Figure B.3 Single-punch run calibration (experiment and simulation). Stepwise electric current is input at the end of rams. (a) resistance; (b) power; and (c) temperature.
Figure B.4 Double-punch run calibration (experiment and simulation). Stepwise electric current is input at the end of rams. (a) resistance; (b) power; and (c) temperature.
Figure B.5 Dummy run (no specimen) calibration (experiment and simulation). Inputs are temperature controlled voltage. (a) resistance; (b) power; and (c) temperature.
APPENDIX C. CASE STUDY OF FAST

C.1 Introduction

In this section, a case study, FAST of a Co-Cr alloyed ball (Figure C.1), is presented to demonstrate the effectiveness of the framework developed in Chapter 3.

The major obstacle to applying FAST on complex shape parts is the inhomogeneous density distribution due to compaction. Coupling between heat transfer, electricity conduction and densification renders the temperature field more complicated than for small size specimens. It is possible to minimize the problem by optimizing the process using modeling. The goal is to produce homogeneously densified, complex shaped sintered products.

The modeling presented in this thesis does not allow for the most general way of coupling between densification evolution in sintering and the thermoelectrical problem as discussed in Chapter 5. As a compromise, a two-step procedure was applied using the framework developed in Chapter 3: (a) a compaction simulation is performed using a porous plasticity model; and (b) the predicted geometry of the compact along with the corresponding porosity distribution was used in a FAST simulation that estimates the temperature and electric fields. Porous plasticity is assumed to be temperature independent here for simplicity. It is worth to note that this study is not intended to make an accurate prediction, but rather to demonstrate the viability of the framework developed in Chapter 3. Details of the simulation procedure are given below.
C.2 Simulation setup

*Compaction of a porous material*

The closed die compaction of a Co-Cr alloy with an initial relative density of 0.6 was considered here. Schematics of the upper and lower punches, die and powders are shown in Figure C.1. The finite element mesh used in the compaction is also given in Figure C.1.

In this simulation, due to the axial symmetry of the problem, only one half cross-section of the geometry was considered. Note that the mesh at the compact corners is refined since these regions experience severe deformation during compaction. The compact consists of 934 continuum axial symmetric 4-node reduced integration elements, CAX4R.

In this step, the isotropic porous plasticity model of Gurson [C.1] was used to simulate densification. The parameters $q_1$, $q_2$ and $q_3$ in the Gurson model and other materials properties of compact are given in Table C.1.

Punches and die are modeled as rigid elements. Compaction was achieved by moving the top punch until powders fill the cavity of the die completely. Interaction between powder and die is assumed to be frictionless. ABAQUS/Explicit [C.5] is employed.

*FAST simulation*

The numerical procedures required in this step were discussed in Chapter 3. In this simulation, we can not assume that the relative density is uniform. The variation
of relative density is estimated from the compaction simulation and is mapped onto a FAST simulation. The finite element mesh is shown in Figure C.2. Thermal conductivity and electric conductivity are taken to be functions of the relative density. A simple linear model for both thermal and electric conductivities of porous specimens was used:

$$\frac{k_{\text{eff}}}{k_s} = (2RD - 1) \quad \text{and} \quad \frac{\sigma_{\text{eff}}}{\sigma_s} = (2RD - 1)$$

where $k_{\text{eff}}$ and $\sigma_{\text{eff}}$ are effective thermal and electric conductivity at relative density $RD$, respectively. $k_s$ and $\sigma_s$ are thermal and electric conductivities, fully dense, which are given in Table C.1.

The heating profile using closed-loop controlling algorithm shown in Figure 3.8 was simulated. Other thermal and electric boundary conditions are the same as those described in Chapter 3. Electric gap conductances are taken to be the values chosen in Appendix B ($h_g^h = 1.25 \times 10^7 (\Omega \cdot m^2)^{-1}$, $\sigma_g^h = 7.5 \times 10^6 (\Omega \cdot m^2)^{-1}$).

Density evolution within the component is the result of both applied pressure during the precompaction and temperature distribution during FAST. In order to estimate the role of compaction and interfacial properties, three simulations were performed: (a) inhomogeneous density distribution after compaction and no thermal contact resistance (i.e., $h_g^h = 10^8$ W/m²-K and $h_g^v = 10^8$ W/m²-K) during FAST; (b) uniform density after compaction and no thermal contact resistance during FAST; and (c) inhomogeneous density distribution and non-zero gap thermal resistance between
specimen and die \( \left( h_g^h=10\text{ W/m}^2\text{-K} \right) \) and \( h_g^v=10^8\text{ W/m}^2\text{-K} \) in this study) during FAST.

A comparison of cases (a) and (b) illustrate the effect of non-uniform density as the result of applied pressure on the temperature inhomogeneity. A comparison of cases (a) and (c) provides insight on the effect of thermal interfacial condition on the uniformity of the temperature field.

### C.3 Results and discussion

Figure C.3 shows the hydrostatic pressure distribution at the end of compaction of cases (a) and (c). The maximum stresses occur at points A and B marked on Figure C.3. At his point, the “sharp” outer edge of the top punch is severely stressed. Therefore, the pressure in FAST is limited by the strength of punch and die material rather than the pressing capacity of the machine. High strength graphite [C.6] is required to allow for high pressure compaction.

The porosity of cases (a) and (c) is not uniform as shown in Figure C.4. The average porosity is about 0.18. The highest porosity is observed at the edge of the ball end, where minor tensile stress develops during compaction.

Co-Cr alloy powders are highly electrically conductive even at intermediate relative densities. In case (a), the electric current distribution at the end of sintering is shown in Figure C.5. It shows that even though the conductivity of the sample is high enough for most of current going through the specimen, the maximum current density occurs at the punches due to their small cross section. As a result, the ends of the punches are acting as heating elements as shown in Figure C.6. The specimen is
heated by the heat conducted away from the punches. The temperature distribution is given in Figure C.7. The maximum temperature difference within the specimen is about 86 °C.

Density inhomogeneity of the green compact of the complex shape can be improved by applying a multi-punch configuration, as shown in Figure C.8. However, the equipment for multi-punch compaction is complicated and expensive. More importantly, even though a uniform density is assumed, the resultant temperature field is similar to that from the single punch compaction as shown in Figure C.9. Again, this was due to the fact that the specimen was heated by the heat conduction from the punch. The only improvement was that the electric current, as shown in Figure C.10, was more uniform than case (a). In other words, unless variation of porosity is extreme, the assumption of a uniform density is reasonable for this work.

The edge of the ball acts as heat sink in both cases (a) and (b), as shown in Figure C.11. By modifying the heat flux, it is possible to minimize the temperature inhomogeneity. In case (c) low thermal gap conductance \( (h_g = 10 \text{ W/m}^2\text{-K}) \) at the interface between the specimen and the die was applied. In reality, this can be achieved by using a thermally insulating material, e.g., mica. The heat flux near the edge of the ball is then partially diverted as shown in Figure C.11(c). The resultant temperature field, as shown in Figure C.12, becomes more homogeneous than case (a) and (b). The maximum temperature difference is 72 °C which is about 20 % less than case (a) and (b).
C.4 Conclusions

In this appendix, the simulation of compaction and FAST of a ball shape part has been conducted. It was found that, for Co-Cr alloys, the homogeneity of relative density after compaction will not affect the temperature distribution per se due to the high thermal and electric conductivities even at intermediate relative densities. By predicting temperature field, it is possible to improve the temperature homogeneity in the specimen by modifying local insulation conditions.
Figure C.1  (a) Ball shape part; (b) schematic and geometries of punch and die for compaction of ball; and (c) FE mesh of compaction.
Table C.1 Material properties of Co-Cr alloy.

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
<th>Reference and remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density</td>
<td>8500 kg/m$^3$</td>
<td>[C.2], fully dense</td>
</tr>
<tr>
<td>Elastic modulus</td>
<td>240 GPa</td>
<td>[C.3], fully dense</td>
</tr>
<tr>
<td>Poisson’s ratio</td>
<td>0.3</td>
<td></td>
</tr>
<tr>
<td>Yield strength</td>
<td>700 MPa</td>
<td>[C.3], assume same as tensile strength</td>
</tr>
<tr>
<td>q$_1$</td>
<td>1.11</td>
<td></td>
</tr>
<tr>
<td>q$_2$</td>
<td>1.025</td>
<td></td>
</tr>
<tr>
<td>q$_3$</td>
<td>1.23</td>
<td></td>
</tr>
<tr>
<td>Initial relative density</td>
<td>0.6</td>
<td></td>
</tr>
<tr>
<td>Heat capacity</td>
<td>422 J/kg-K</td>
<td>[C.4], fully dense</td>
</tr>
<tr>
<td>Thermal conductivity</td>
<td>14.8 W/m-K</td>
<td>[C.4], fully dense</td>
</tr>
<tr>
<td>Electric conductivity</td>
<td>1030927 (Ω-m)$^{-1}$</td>
<td>[C.4], fully dense</td>
</tr>
</tbody>
</table>
Figure C.2  (a) Finite element model of FAST on the ball after compaction. For clarity, no elements and only the outlines of each component are shown; (b) Details of finite elements of punches, die and compact.
Figure C.3  Hydrostatic pressure distribution at the end of compaction of cases (a) and (c).
Figure C.4 Porosity distribution at the end of compaction of cases (a) and (c).
Figure C.5  (a) Electric current distribution of punch/die/compact at the end of FAST. (b) Detailed electric current distribution in the compact of case (a).
Figure C.6  (a) Joule heating distribution of punch/die/compact at the end of FAST.  
(b) Detailed Joule heating distribution in the compact of case (a).
Figure C.7  (a) Temperature distribution of punch/die/compact at the end of FAST. (b) Detailed temperature distribution in the compact of case (a).
Figure C.8  Multi-punch configuration. Compact with uniform density may be achieved by pushing punch 1 first and then punch 2.
Figure C.9  Temperature distribution of compact of case (b) assuming uniform relative density of 0.82.
Figure C.10  Electric current density distribution of compact of case (b) assuming uniform relative density of 0.82.
Figure C.11  (a) Heat flux of case (a); (b) Heat flux of case (b); and (c) Heat flux of case (c).
Figure C.12 Temperature (NT11, °C) distribution of compact of case (c).
VITA

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Publications

* Also 8 papers in referred Conference Proceedings.*