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On Joule heating during spark plasma sintering of metal powders

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Joule heating as a primary heating source mechanism was probed during spark plasma sintering (SPS) of pure metal powders (Fe, Ni and Cu). Resistance to electric path was estimated from voltage–current measurements obtained online during these experiments. Resistance was observed to saturate at the same value irrespective of the type of metal powder, after attaining a sintering temperature of ~$0.3T_m$. This saturation in resistance is attributed primarily to the Joule heating that occurs at the graphite-foil and punch in an SPS system.

Keywords: Spark plasma sintering; Joule heating; Electrical resistivity; Densification

Spark plasma sintering (SPS) has emerged as an advanced sintering technique in the last few decades, primarily because of its versatility to sinter a variety of materials to full density with retention of grain size at much lower homologous temperatures in a shorter duration compared to conventional processes. These advantages are attributed largely to localized internal heat generation mechanisms in SPS as compared to external heat supply in conventional sintering [1–6]. The formation of spark followed by plasma in the vicinity of pores is considered to be one of the many mechanisms involved in SPS. However, ambiguity on the existence of plasma still persists in the literature [7–9]. Another probable mechanism is Joule heating, which is a promising heat generation mechanism in the case of metals. Joule heating occurs because of resistance to electric current. The maximum resistance offered to the electric path is believed to come from particle–particle contacts [10–12]. However, these contacts start diminishing with the progress of densification due to pore closure. Consequently, the resistance to electrical path due to these contacts also decreases and eventually vanishes once the material attains near-full density. The resistance to the electrical path also involves contributions from graphite–die punch system and graphite foil–powder contacts [13]. Table 1 lists the various locations in a SPS system where Joule heating is anticipated to occur, as reported in the literature. However, the relative contributions of various sources have not yet been explicitly elucidated. Therefore, online monitoring of the variation of voltage and current may give an insight to understand the Joule heating during SPS experiments. Thus, in the present study, an attempt has been made to probe the Joule heating sources by measuring resistance during the densification of three different pure metals. The variation in the resistance was evaluated by online measurement of voltage and current during densification.

Fe, Ni and Cu powders with purity >99.5% and size <325 mesh (Loba Chemie Ltd.) were sintered in Dr Sinter SPS-5000 machine (Sumitomo Metals, Japan). Graphite die-punch (20 mm inner diameter) with graphite foil inserted in between the punch and powder was used to consolidate these powders. A schematic of a typical die-punch setup is shown in Figure 1a. A constant uniaxial pressure of 10 MPa was applied throughout the experiment. Sintering was performed in vacuum (~10 Pa) to avoid oxidation of the powders at a temperature of $0.7T_m$ (T_m = melting point of pure metal). The desired temperature was reached with a ramp rate of 100 K min^{-1} and the temperature was measured using a k-type thermocouple inserted in the graphite die at a position of 4 mm away from the inner diameter. The displacement, voltage (V) and current (I) profiles were monitored continuously during the experiments and were acquired for further analysis.

The displacement data obtained during the SPS experiment was used to calculate instantaneous relative density (D_i) of pellet using the relation $D_i = D_f(h_f/h_i)$, where D_f is the final relative density of the sample measured by Archimedes' method while the theoretical density of pure metal was used as a reference, h_f and h_i are the final and instantaneous height of the pellet, respectively. The

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Table 1. The prominent Joule heating region(s) in SPS for conducting materials, as reported in the literature.

Joule heating region	Refs.
Graphite die-punch contacts/interfaces	[4,14–17]
Particle–particle contacts/internal heating	[4,10]
Punches	[18]
Graphite die-punch assembly	[19,20]

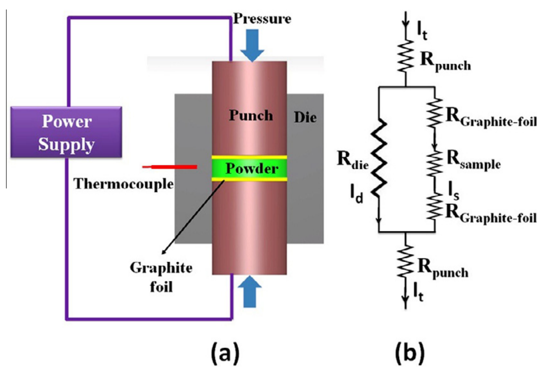

Figure 1. (a) Schematic of a typical graphite die-punch setup used in the present study of SPS experiments. (b) An equivalent Ohmic circuit for SPS die-punch setup in the present study.

variation of relative density with homologous temperature shows a typical S-curve with final densities of >85% of the theoretical density (Fig. 2). Also, Figure 2 illustrates the variation of resistance (right y-axis) as a function of homologous temperature (T/T_m), which will be discussed later. The maximum densification of all powders occurred in the range ~0.4–0.6T_m. The applied pressure (10 MPa) and sintering temperature (0.7T_m) are much lower than those used in conventional sintering techniques [21]. Such a fast-paced densification at lower temperatures must be attributed to the underlying heating mechanisms during SPS [22]. Aman et al. [23] have demonstrated the contribution of localized phenomena in the densification of Cu by pressureless SPS. However, the effect of densification mechanisms is not considered in our study, which concentrates on understanding the mechanisms/localized heat sources in SPS [24]. Thus, further discussion in the present paper will only deal with Joule heating which is a prominent phenomenon during field-assisted sintering of metals.

Joule heating occurs due to the resistance offered by the material to the electric path, and is directly proportional to the product of the square of current (I) flowing through the material and the resistance (R) it offers (i.e. I^2R). In this SPS experiment, a DC electric pulse sequence of 12:2 ms was used. Among the various possible locations at which Joule heating occurs in SPS, the Joule heating occurring at particle–particle contacts is considered to play a significant role in the sintering of metal powders [4,10]. Thus, the instantaneous relative density of the powder affects the resistance and hence plays a role in the densification process. From the V–I profile, the total resistance (R) is calculated as $R = V/I$ (assuming Ohmic nature only). The variation of the resistance with relative density of the metal is shown in Figure 3. It is interesting to note that the resistance offered at the beginning of sintering is much higher and decreases drastically with the progress of densification [25,26]. Intriguingly for all the three metals, the resistance saturates nearly to a constant value ($3 \times 10^{-3}\,\Omega$) with respect to relative density and remains constant as sintering progresses. Thus, the following main observations can be made from Figure 3:

1. In the initial stages of sintering, the resistance offered to the electric path is significantly higher, which could be due to the contribution from the large number of particle–particle contacts with thin oxide layer on the surface in addition to other contacts (graphite-die–punch, punch–foil, and powder–foil).
2. After the metal powder achieves a critical relative density, the resistance to the electric path saturates to a lower value. It appears that the resistance becomes insensitive to metal powder properties once the sample reaches this critical value of relative density.

In order to understand the variation of the resistance during SPS, the Ohmic circuit for SPS should be considered [27]. An Ohmic circuit for the SPS die punch setup of the present study is shown in Figure 1b. The total current (I_t) is the sum of the current flowing through the die (I_d) and the current flowing through the sample (I_s). During SPS of metals, as the density increases, minimum current flows through the die (i.e. $I_d \ll I_s$). Thus, the current flowing through the sample is assumed to be the total current (i.e. $I_s \approx I_t$) [11]. In such a scenario, Joule heating must be the resultant of the resistance offered by the punch, graphite –foil and sample assembly. The resistivity of the graphite -punch and foil was assumed to be constant with

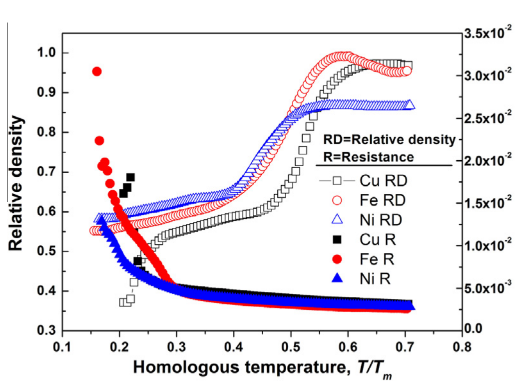

Figure 2. Variation of relative density and resistance with homologous temperature for Fe, Ni and Cu. The resistance saturates at ~0.3T_m which is related to the dielectric breakdown of the oxide layer on the metal powder.

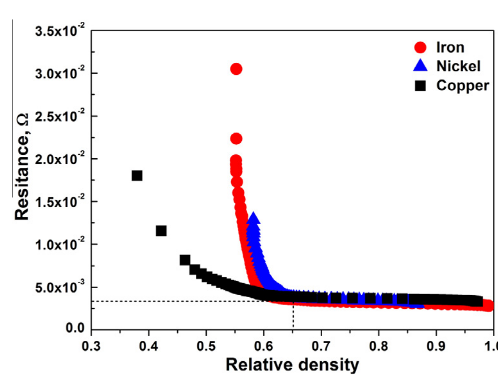

Figure 3. The variation of resistance with relative density of three different metals Fe, Ni and Cu. The dotted lines indicate that the resistance value for all three metals saturates to same value with relative density.