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The sintering mechanism in spark plasma sintering – Proof of the occurrence of spark discharge

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Typical sintering experiments were conducted to understand the spark plasma sintering (SPS) mechanisms. Based on the results of the direct visual observations and characteristic microstructure analysis, we believe that spark discharge does indeed occur during the SPS process. The high-temperature spark plasma could be generated in the microgaps due to the discharge effect. Fast and efficient sintering can be achieved under the combined action of spark discharge, Joule heating, electrical diffusion and plastic deformation effect in the SPS process.

Keywords: Spark plasma sintering (SPS); Discharge; Spark plasma; Sintering mechanism

Spark plasma sintering (SPS), also known as the field-activated sintering technique and pulsed electric current sintering, is a comparatively novel sintering process that allows fabrication of bulk materials from powders using a fast heating rate (up to 1000 °C min^{-1}) and short holding times (in most cases 0–10 min) at low sintering temperatures (200–300 °C lower than most of the conventional sintering techniques) [1–9]. Because of its great advantages, SPS is by far the most popular of the ultrarapid sintering techniques, and is used to process nanostructured materials, amorphous materials, intermetallic compound, metal matrix and ceramic matrix composites, highly refractory metals and ceramics, etc., which are difficult to sinter by common methods [9–17].

Extensive efforts have been made in investigating SPS and developing it as a promising technique for rapid densification of advanced new materials with various applications [18–22]. However, limited data are available on the sintering mechanism involved in the SPS process. In addition, a significant gap exists in the fundamental understanding of the SPS mechanisms. This gap is due to the complexity of the thermal, electrical and mechanical processes that may be involved during SPS, in addition to their dependence on the SPS parameters [23]. In particular, the existence of spark plasma and occurrence of discharge in the sintering process are highly controversial.

Most researchers believed that during the SPS process a high electric–pulsed current is applied on the electrodes (as shown in Fig. 1), and the microscopic electrical discharges in the gaps between the powder particles generate plasma, causing sintering [24,25]. The spark discharge can effectively eliminate adsorptive gas and any impurities that are present on the surface of the powder particles, and can easily destroy the oxide films on the particle surface [1,25–27], leading to an enhancement of the thermal diffusion ability of the sintered material [10,24,28,29]. In addition, Joule heating and plastic deformation effects contribute to the densification of the powders [30–32]. Therefore, sintering

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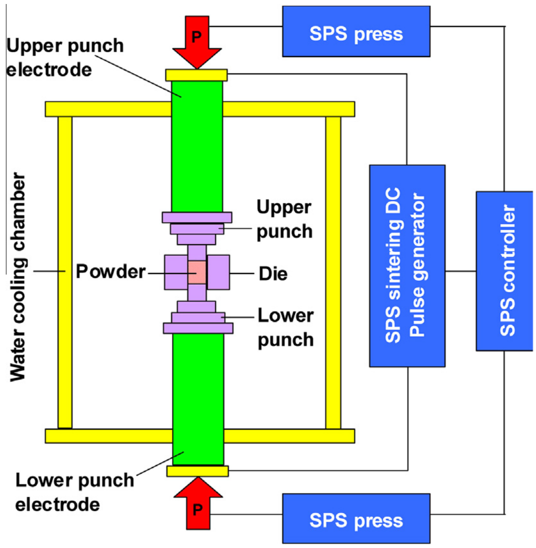

Figure 1. A schematic of the SPS process.

efficiency is remarkably improved and grain growth is typically restricted in the SPS process compared with the commonly used hot pressing (HP) and hot isostatic pressing (HIP) approaches [11,30,33–36]. However, to date there has been no clear evidence demonstrating the occurrence of discharge and the existence spark plasma during the SPS process. Therefore, a number of researchers have questioned the narrative. In particular, Hulbert et al. [37] gave an experimental demonstration of the absence of spark discharge and plasma during the SPS process. They used a number of different methods, including in situ atomic emission spectroscopy, direct visual observations and ultrafast in situ voltage measurements, under a variety of SPS conditions, to investigate the existence of spark plasma and occurrence of discharge in the sintering process. They concluded that there is no plasma, sparking or arcing present during the SPS process, either during the initial or in the final stages of sintering. The exact nature of the SPS mechanism is therefore still under debate.

This study aims to reveal the SPS mechanism and provide direct evidence for the occurrence of spark discharge and the existence of plasma in the SPS process, using typical SPS experiments, including sintering of ceramic powders, metal powders and metal–ceramic composite powders. We hope that our investigation can quell the controversy over the occurrence of spark discharge in the SPS process and contribute to the understanding of the nature of the SPS process.

Six graphite punches with different cross-section areas were used in this investigation. Three of them were placed on a die, with the other three underneath it. The diameters of the first, second and third punches were 150, 120 and 80 mm, respectively. Their heights were 20, 20 and 40 mm, respectively. A cylindrical graphite die with an outside diameter of 60 mm, an inside diameter of 30 mm and a height of 50 mm was used in a 0.5 Pa vacuum chamber. A layer of TiB_2 powders with an average particle diameter of 4.5 μm and a thickness of about 0.3 mm was distributed uniformly on the surface of the third punch. DR.SINTER type SPS-3.20 equipment (Sojitz Machinery Corporation, Tokyo, Japan) with a pulse duration of 3.3 ms was used for the sintering. The holding compressive pressure level applied was 1 MPa. A direct pulse current was gradually applied up to 1200 A within the first minute, then maintained at 1200 A throughout the sintering process. A KT 19.43 infrared thermometer (Heitronics Infrarot Messtechnik GmbH, Wiesbaden, Germany) with a measuring range of 0–2500 °C was used to determine the sintering temperature (T) of the die.

Figure 2 presents the sintering die and punches in the water cooling chamber of the SPS system at different sintering stages. When the pulse current was applied up to 760 A (the corresponding sintering time $t = 38$ s), the first spark discharge point was observed on the interface between the second and third punches. After a further 15 s, the second discharge point appeared. Figure 2a shows the two discharge points with a sintering time of 60 s. Here the temperature at the centre of Point ① was 1027 °C and that at Point ② was 1256 °C. However, the sintering temperature at the surface centre of the die was only 337 °C. The spark discharge at Point ① became progressively weaker

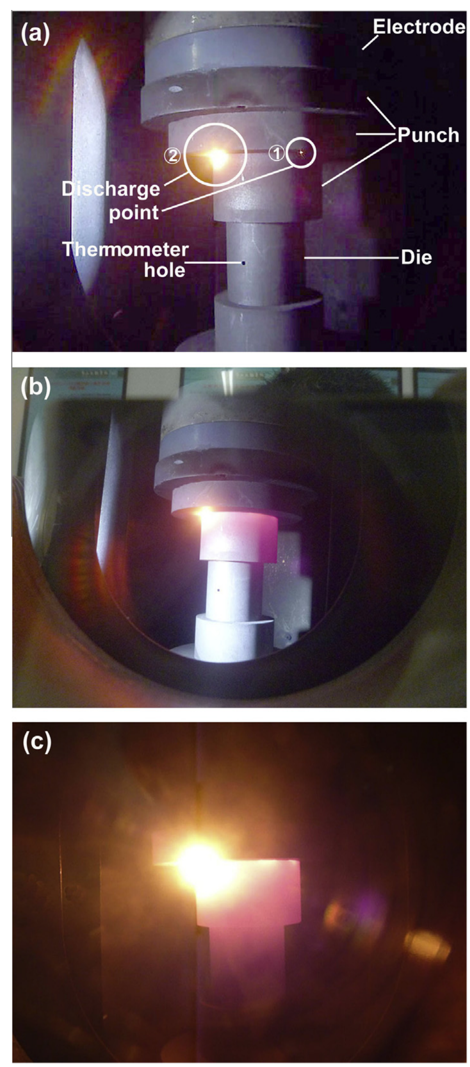

Figure 2. Discharge occurred at the interface between the second and third punch on the die at different sintering stage: (a) $t = 60$ s, $T = 337$ °C; (b) $t = 120$ s, $T = 586$ °C; (c) $t = 240$ s, $T = 992$ °C.