

Enhance the heating rate of the PECS process by inserting a conducting body into BN powder compact

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Pulsed electric current sintering (PECS), or the so-called spark plasma sintering (SPS), is a novel sintering process with the characteristic of short firing cycle. The process can be applied to densify metallic or ceramic bodies within a matter of minutes instead of several hours. The process is similar to the conventional hot pressing in the way that the powder compact is heated with the application of an external load. Nevertheless, instead of using an external heating source, a pulsed electric current is passed directly through the electrically conducting, such as graphite, mold.

Tokita *et al.* claimed that the pulsed current generates spark discharge and even plasma between conducting particles [1]. They suggested that the generation of spark discharge or plasma removes the adsorbed species from the surfaces of particles during the initial stage of PECS. The grain boundary diffusion is thus enhanced in the later stage of the process. However, Wang *et al.* [2] claimed that the pulsed electric current sintering on alumina is similar to that of hot-pressing. The pulsed electric current heated the graphite mold first by Joule heat, then the heat transferred inside into the powder compact. As the heating rate is fast, a temperature gradient exists. A dense outer region is thus observed within an alumina specimen after PECS [2]. Munir *et al.* suggested that the presence of pulsed electric field contributes to the fast densification [3]. Shen *et al.* suggested that the discharge can still take place between the non-conducting particles [4].

Many attempts have been made to explore the sintering mechanism of PECS. Though systematic studies have been conducted [1–5], the mechanisms for the densification and grain growth are far from clear. Nevertheless, most previous studies agreed upon the significance of PECS technique to the industrial production. In the present study, the presence of spark discharge between conducting particles is confirmed by inserting a conducting body into nonconducting particles.

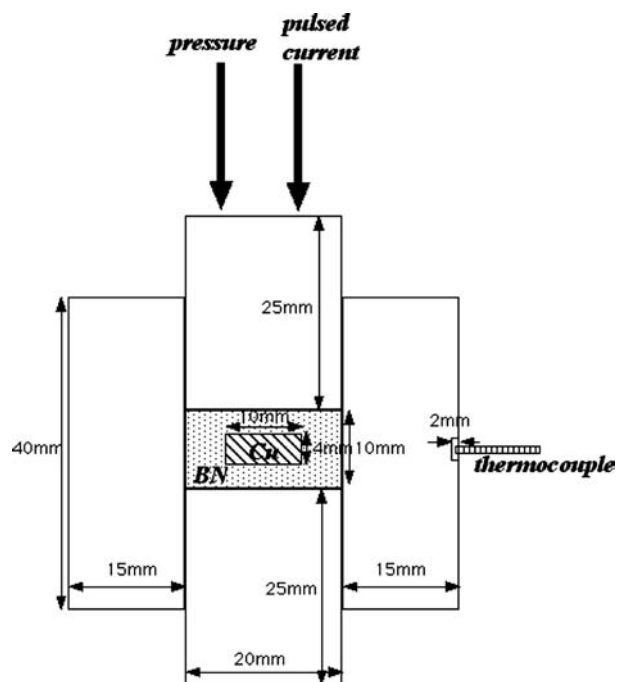


Figure 1 Schematic of the experimental setup for the PECS process.

Several copper specimens were prepared by using a PECS furnace (Model SPS-1050, Sumitomo Coal Mining Co., Tokyo, Japan). The Cu particles with average size of 20–30 μm were packed in a WC-Co die with an inner diameter of 10 mm. The punches were also made of WC-Co. An external pressure of 100 MPa was applied uniaxially on the powder compact throughout the PECS process. The peak temperature was 400 $^{\circ}\text{C}$. The heating rate was 100 $^{\circ}\text{C}/\text{min}$ and the dwell time 1 min. The final dimensions of the Cu specimen were 4 mm in thickness and 10 mm in diameter. The Cu specimens were nearly fully dense after PECS.

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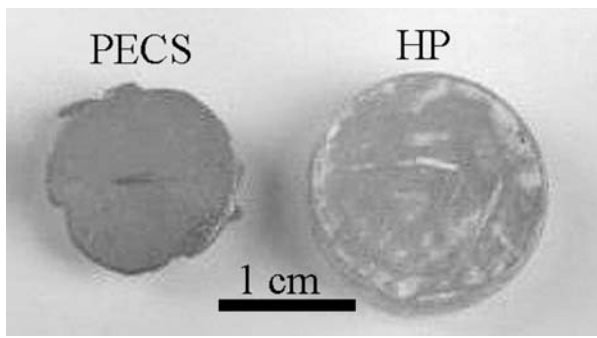


Figure 2 Morphology of the Cu specimen after removing the surrounding BN.

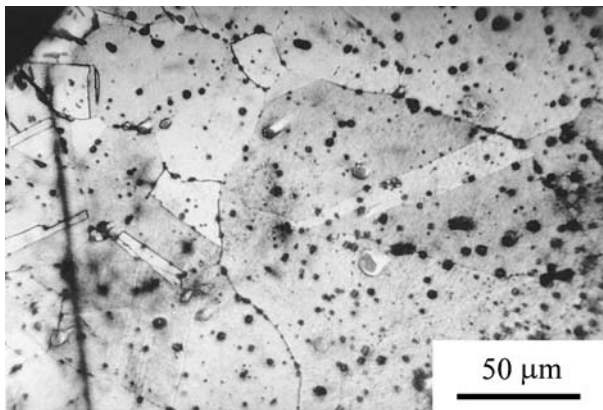


Figure 3 Microstructure of the Cu specimen after HP at 1080 °C for 1 hr.

The Cu specimen was then inserted into the center of a boron nitride powder compact, as shown in the schematic in Fig. 1. A graphite mold with outer diameter of 50 mm and inner diameter of 20 mm was used during PECS. The load applied was 30 MPa. The peak temperature was 1080 °C, the dwell time 1 min. The temperature was measured by using a *K*-type thermo-couple that inserted into a hole on the outer surface of the mold. The depth of the hole was 2 mm. The final thickness of the specimen was 10 mm.

A comparison experiment was also conducted by using a hot-pressing technique. The Cu specimen prepared by PECS was also inserted into a BN powder compact, then hot-pressed at 1080 °C with a load of 30 MPa. A graphite mold with the same dimensions as those of the PECS mold was also used; the dwell time at hot-pressing was 1 hr.

The BN was not fully dense after either PECS or HP treatments. The Cu specimen could be easily removed from the BN powder compact. The cross-section of the Cu specimen was polished and etched in 10% HCl solution, then observed with an optical microscope.

The morphology of the Cu specimen after PECS and HP is shown in Fig. 2. The Cu specimen after HP remains its original shape and size. However, a serious shape distortion is noted for the Cu specimens after PECS treatment. Furthermore, the size of the PECS specimen is much smaller than that of the HP specimen. Many small copper droplets are noted on the inner surface of the graphite mold, especially at the gap between punch and mold. It indicates that the temperature of Cu specimen during PECS treatment is far higher than the temperature measured from the outer surface of the graphite mold. The temperature is not only higher than the melting point of Cu but also high enough to vaporize some Cu.

The cross-section of the Cu specimens after HP and PECS is shown in Figs 3 and 4, respectively. Though the dwell time at the peak temperature during PECS is much shorter than that at the HP temperature, the grain size of the Cu specimen treated with PECS is much larger than that of the specimen treated with HP. It implies that either the temperature of the conducting Cu specimen or the grain growth rate is increased significantly by using BN powder surrounding. There are many debates on the coarsening behavior during PECS process. Systematic studies had been conducted [1–5]; these studies recognized the importance of controlling the processing parameters during PECS. Among the processing parameters, the peak temperature, pressure and heating rate are found to be the three most influential

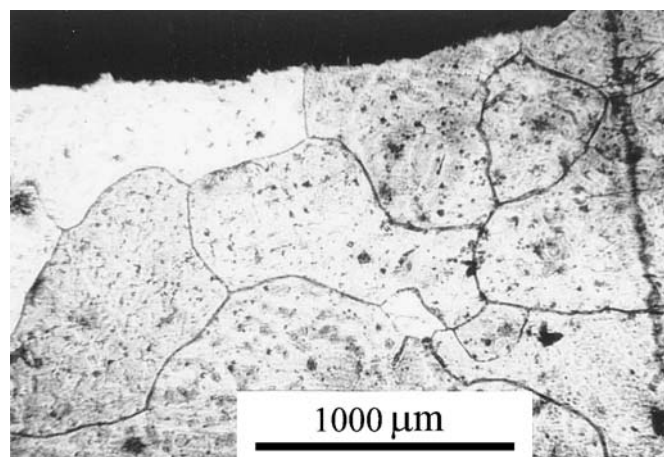


Figure 4 Microstructure of the Cu specimen after PECS at 1080 °C for 1 min.

parameters. Raising the peak temperature and pressure during PECS can usually increase the final grain size. Nevertheless, the role of heating rate on coarsening behavior remains unclear. The increase of heating rate has been reported to result in finer grain size [2, 4, 6–8]. However, a fast heating rate has also been reported as the key parameter to enhance grain growth [5]. A recent study proposed that the coarsening behavior during PECS is far more complicated than one would expect [9]. Zhou *et al.* indicated that higher heating rate induces higher coarsening rate at the early stage and a lower rate at late stage of sintering. In any case, since the shape of the Cu specimen after PECS is distorted and the existence of Cu drops at the gap between punch and mold, it indicates that the temperature of the Cu specimen during PECS is much higher than that of the graphite mold. The coarsening of the Cu specimen prepared by PECS process is likely contributed mainly by the resulting high temperature. A separate experiment has also been conducted by inserting titanium diboride (TiB₂) powder compact into the BN powder bed. A sharp temperature increase has also been observed for the TiB₂ specimen [10]. Though the Cu specimen was not in contact with the graphite mold during PECS and the time at the peak temperature during PECS was very short, 1 min, the temperature and heating rate on the embedded Cu specimen are significantly increased. BN is an electrical insulator; the presence of BN powder bed should affect little on the electrical field around the Cu specimen. Therefore, the pulsed electric current can still induce spark discharge between Cu particles. It implies that the spark discharge is the dominant mechanism for the increase of heating rate. Furthermore, BN is also a good thermal insulator; the heat generated within the Cu powder compact by the spark discharge is confined within the porous BN powder bed. Therefore, the temperature of the Cu specimen is high. A recent study demonstrated that the addition of 1 vol.% Ni nanoparticles could enhance the grain growth of alumina matrix during PECS [11]. It also suggested

that the incorporation of conducting particles into non-conducting powder compact could introduce an extra heating mechanism into the non-conducting specimen.

The heating rate during pulsed electric current sintering (PECS) is very fast, >100 °C/min. In the present study, a methodology is introduced to further enhance the heating rate. An electrical conducting body, Cu specimen, was inserted into a non-conducting BN powder compact; both the heating rate and peak temperature on the Cu specimen are consequently increased. The increase of heating rate is related to the presence of spark discharge within the conducting particles.

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