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Flash spark plasma sintering of ultrafine yttria-stabilized zirconia ceramics

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ABSTRACT

The microstructure evolution during high heating rate 'flash' spark plasma sintering (SPS) of ultrafine yttria-stabilized zirconia powder is presented. Consolidation during SPS took place in a single-stage, the activation of preferable interaggregate densification, and thus preserving microstructural features of initial powder. Different sintering stages were identified during conventional SPS, showing a distinctive role of processing temperature. An explanation and analysis of the driving forces behind these observations was attempted.

Recent interest in ultrafast consolidation using field-assisted consolidation techniques such as flash sintering, microwave sintering and spark plasma sintering (SPS) showed that metals, ceramic and their composites can be consolidated with great efficiency saving both time and energy during such materials processing [1–10]. It is obvious, that in terms of sintering theory such quick consolidation is often attributed to various phenomena mainly originating from specific electromagnetic field – mater interaction. In addition, in case of the SPS applied pressure changes the consolidation mechanism and also alters temperature distribution inside specimens.

Furthermore, recent efforts using 'flash' sintering show that consolidation time of ceramic materials may be sufficiently decreased [7–12]. Originally, flash sintering (FS) experiments were focused mainly on consolidation of various oxide ceramics. Recent experiments show that by using modification to original flash sintering techniques non-oxide such as SiC, ZrB_2 ceramics may be consolidated using 'flash' regime [10–12]. Such quick flash spark plasma sintering consolidation of ZrB_2 ceramics reported by Grasso et al. [11], in particular, requires additional exploration. It was shown that ultra-fast consolidation of zirconium diboride might be completed within minutes using flash SPS (FSPS). Due to abandoning of graphite mold-punches set-up heating or flashing mode during FSPS is mainly controlled by ceramic sample which in case of conductive ZrB_2 allowed fast heating rate. Unlike conductive ZrB_2 similar studies on 3YSZ have not been reported yet, greatly to the fact that temperatures of at least of 700 °C are required to achieve sufficient level of ionic conductivity. We believe that this FSPS consolidation should be different from conventional SPS (i.e. SPS using a graphite die, where a powder mixture is a subject of additional heating from the graphite die), given that in the former case flash phenomena (i.e. electric runaway, high-heating rate) may also occur. Thus, it is the primary objective of the present study to explore the difference between flash SPS and regular SPS while using identical hardware (Sumitomo Dr Sinter, Japan) and ultrafine powder (~60 nm aggregates of ~9 nm primary crystallites [13,14]). Secondly to explore effect of processing conditions on microstructure evolution observed during consolidation of ultrafine and uniform 3YSZ powder.

For the FSPS experiments we used ultra-fine 3YSZ powder synthesized according to Vasylkiv et al. [13,14]. Moreover we selected such powder due to the fact that densification of this 3YSZ powder was explored using the flash, microwave, spark plasma sintering, and conventional sintering techniques [7,14–17]. Due to fine primary crystallite size (7–11 nm) as well as uniform aggregation (40–80 nm spherical or lenticular aggregates) the powder is known for its low-temperature sinterability [13–15]. Furthermore, this particular 3YSZ powder also has a distinctive particularity – it is impossible to consolidate this powder into dense ceramics at high-temperatures (>1400 °C) – due to immediate aggregation and formation or high-interagglomerate porosity. Hence in case of conventional sintering, where change in powder system curvature and specific surface area reduction governs the consolidation process, this powder can be consolidated into dense ceramic only at low temperature ~1150 °C using long dwell times >10 h.

It is logical exploring what benefit might FSPS provide as compared with conventional SPS (CSPS), as well to explore microstructure evolution of 3YSZ during ultra-fast consolidation.

In the present case, fine 3YSZ powder was consolidate to produce dense samples using spark plasma sintering technique in conventional

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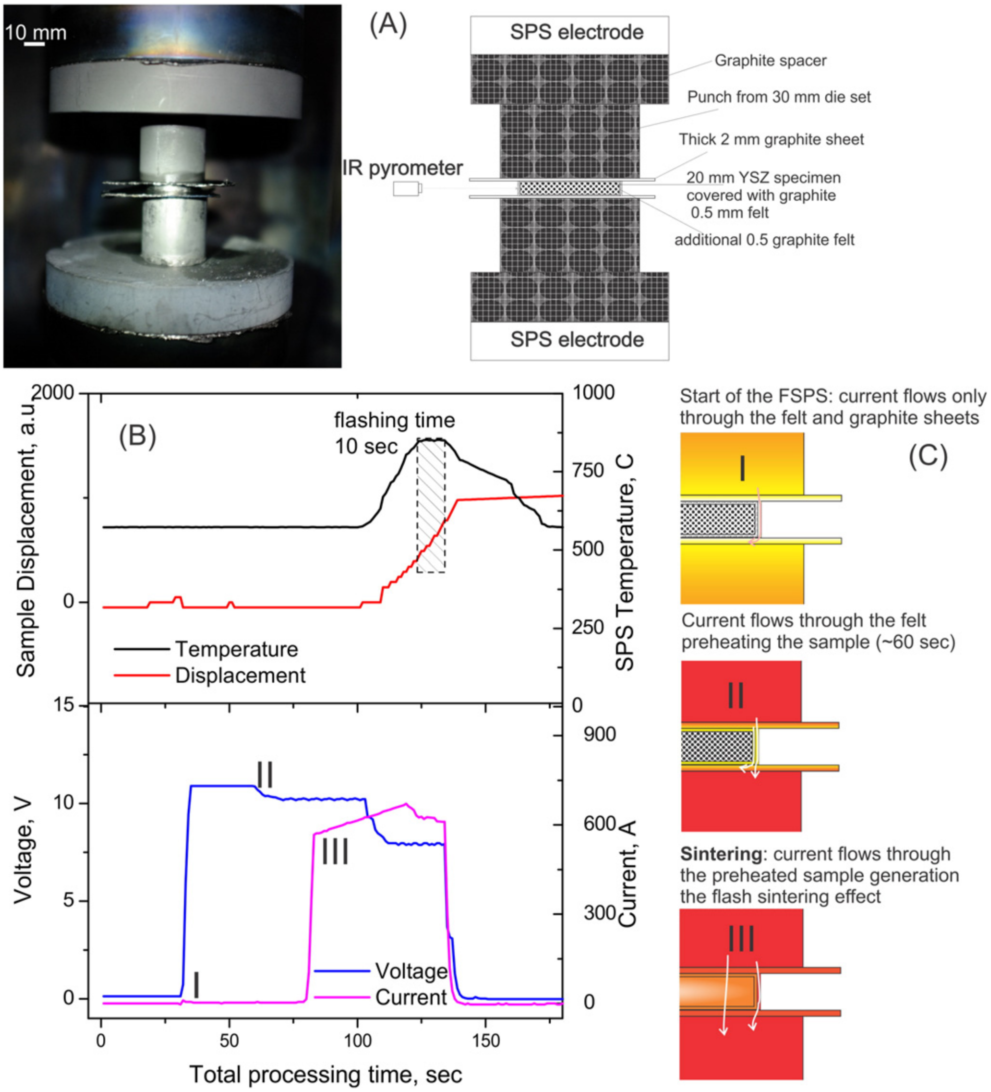

Fig. 1. Flash spark plasma sintering consolidation of YSZ ceramics using the SPS set-up without graphite die: (A) experimental set-up used during FSPS of 3YSZ ceramics, (B) lower image shows change of temperature, displacement, voltage and current during FSPS of 3YSZ. Discharge time at 850 °C is 10 s presented as dashed area. (C) Right image shows different stages of heating process during FSPS of 3YSZ ceramics. Note that at 850 °C value of current was manually decreased to prevent runaway phenomena. Until a rapid increase in current the macroscopic sparking between graphite felt and sample was clearly observed due to short-current situation in case of lack for 3YSZ sample conductivity at low temperatures. No peaks in temperature are shown since pyrometer was focused on side of specimen, and not on the graphite felt – specimen – graphite punch interface.

and flash modes, i.e. using a conventional graphite mold set and 'mold-free' processing [12], when pre-sintered green specimen was placed between two graphite punches, these acted as preheaters to initiate flash [11,12] consolidation process (Fig. 1(C)).

In a typical sintering experiment, 8 g of zirconia powder was filled into a 20 mm diameter graphite die. To perform the FSPS sintering experiments, the 3YSZ powder was partially sintered under 30 MPa at 750 °C for 1 min.

At this stage the relative density was 40 ± 5% and the samples was dense enough to be processed by FSPS – i.e. to form strong intracrystallite contacts, but remain <50% TD. To perform FSPS the sample was pressed between two graphite punches of 30 mm diameter (Fig. 1(A)). The temperature was probed by the side pyrometer focused on the side of the graphite felt using an emissivity of 0.90. During the FSPS, graphite mold was not employed, so the electric current passed entirely across the sample (after the preheating stage, Fig. 1) [12]. In order to minimize the heat loss by radiation from the sample, and to perform the temperature measurement using know emissivity vs temperature dependence, additional graphite felt was wrapped around the 3YSZ specimens during sintering.

In FSPS experiments, a constant uniaxial pressure of 20 MPa was applied. The samples were discharged under a peak power of about 1–4 kW for 5 and 10 (Fig. 1(B)) (see Movies 1 and 2). The power was rapidly decreased after the "discharge" and it was maintained for a few seconds by manually decreasing current. We also noted a macroscopic sparking between two thick graphite sheets, which occurred in case of high voltage values and low current values (i.e. <3 A).

This suggests that during this initial stage of FSPS, specimens were indirectly preheated by a short-current situation. Only when temperature and conductivity of 3YSZ specimens increased due to local increase in temperature, rapid increase in current that passed through the specimens was observed. This simultaneous increase in conductivity and temperature has similar nature to "flash" sintering or thermal runaway phenomena observed during field assisted consolidation of ceramic [1,2,6].

For comparison, the samples were also sintered using a CSPS configuration in the temperature range of 700 °C–1100 °C under a uniaxial pressure of 20 MPa, with a dwell time of 0–5 min.

Microstructural observations and analyses were carried out on fractured sample sections using SU 5500 cold-emission FE-SEM (Hitachi,