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Microstructure and mechanical properties of air atomized aluminum powder consolidated via spark plasma sintering

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ABSTRACT

Two air atomized aluminum powders, one of commercial purity and the other magnesium-doped (0.4 wt%), were processed by SPS and conventional PM means. An investigation of SPS processing parameters and their effect on sinter quality were investigated. A comparison with conventionally processed PM counterparts was also conducted. Applied pressure and ultimate processing temperature bore the greatest influence on processing, while heating rate and hold time showed a minor effect. Full density specimens were achieved for both powders under select processing conditions. To compliment this, large (80 mm) and small (20 mm) diameter samples were made to observe possible up-scaling effects, as well as tensile properties. Large samples were successfully processed, albeit with somewhat inferior densities to the smaller counterparts presumably due to the temperature inhomogeneity during processing. An investigation of tensile properties for SPS samples exhibited extensive ductility (~30%) at high sintering temperatures, while lower temperature SPS samples as well as all PM processed samples exhibited a brittle nature. The measurement of residual oxygen and hydrogen contents showed a significant elimination of both species in SPS samples under certain processing parameters when compared to conventional PM equivalents.

1. Introduction

Spark plasma sintering (SPS) is a material processing technology, in which powdered materials are consolidated into parts using the simultaneous application of pressure and electrical current. Unlike conventional press and sinter powder metallurgy (PM), where compaction and sintering are separate operations [1], SPS processing provides concurrent application of these stages. Specifically, a uniaxial pressure applied in conjunction with a pulsed direct-current flow through the powder and/or die is the fundamental concept of the process. While the exact underlying mechanisms of consolidation remain in dispute, several of those more commonly accepted are outlined by Hulbert et al. [2]. In systems where the current is able to pass through an electrically conductive powder, such as in aluminum, the heating is known to occur through Joule heating [3]. From this, relatively high heating rates are achievable, allowing very high thermal gradients to develop from the core to the surface of individual particles [4]. It has been hypothesized that during the initial stages of SPS, the interfaces of adjacent spherical particles have minimal contact area, and will consequently have high current densities. The joule heating effect is therefore concentrated at these contact points, resulting in a temperature far exceeding that of the set point [4], while the core remains relatively cool. In essence, the surfaces of particles are sintered, while the core is exposed to minimal thermal effects.

Regarding the relationship between aluminum and SPS, there has been extensive work completed on increasing the strength of aluminum [5]. Specifically, SPS processing of nano-grained materials has been shown to produce aluminum alloys with relatively high strengths [6,7] as expected from a Hall–Petch standpoint. However, the tensile behavior of these nano-materials is more complex than this relationship leads it to be. For instance, research on wrought aluminum processed by accumulative roll bonding (ARB) has shown that as grain size is progressively reduced towards nano-sized territory, the susceptibility to a yield drop phenomena increases [8]. Essentially, when the grain size is reduced to or below 1 μm, the material rapidly becomes plastically unstable and can no longer maintain uniform ductility. While the microstructure of ARB and SPS processed aluminum differ with regards to the existence of prior particle boundaries, this phenomenon has shown to be evident in SPS processed pure aluminum [9].

While the majority of aluminum SPS research pertains to the processing of nano-structured powders, there is a growing list of studies that are concerned with SPS response of aluminum

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powders in the as-atomized condition. Here, the effect of the oxide-based film that invariably exists on the surface of aluminum particles has been a key focal point [10–14]. In general, lower SPS temperatures result in metal/oxide/metal bonding, where adjacent powder particles can remain separated by the original refractory film [10,14]. With higher temperatures, however, an increased ability to eliminate and/or disrupt the oxide-based layer has been observed, resulting in a high frequency of direct metal/metal bonding between powder particles. This ameliorated bonding appears to be a product of the physical breakdown of the hard oxide layer from the mechanical pressure applied [13]. However, in addition to this mechanism, Kwon et al. have noted a trend of decreasing residual oxygen content with increasing sintering temperature, far below that of the original powder [10]. This trend is unlike traditional PM, where the net concentrations of oxygen are more apt to increase during sintering as the compact reacts with the trace levels of oxygen invariably present in the gaseous sintering atmosphere.

Similar to conventional PM, it is now known that magnesium additions are beneficial to the sintering behavior in SPS [15–18]. In small amounts, magnesium reportedly reacts with the aluminum oxide skin to produce a magnesium-based oxide by-product of spinel ($MgAl_2O_4$) and/or periclase (MgO). This chemical action presents yet another means of disrupting the oxide film. As such, the relative density and tensile properties are substantially improved when the base powder is doped with magnesium [16]. In this study, a direct comparison of the conventional and SPS sintering behaviors of nominally pure aluminum powder and another doped with 0.4 wt% magnesium was undertaken. The investigation emphasized their response to select process parameters including applied pressure, heating rate, hold time, hold temperature and specimen size. These results have been quantified by physical and mechanical property tests supported by microstructural observations.

2. Materials

Two powders were considered in this work. One was commercial purity aluminum powder while the second was aluminum pre-alloyed with 0.4 wt% magnesium. These will be referred to in this work as 'Al' and 'Al–0.4Mg' respectively. Both powders were produced at Ecka Granules GmbH (Feurth, Germany) through gas atomization. The nominal starting chemistries are shown in Table 1. The average particle sizes of the powders were 133 μm for Al and 97 μm for Al–0.4Mg. To accurately calculate the extent of densification that occurred during sintering experiments, a concise theoretical density was calculated for each powder. This was done using a rule of mixtures approach based on the nominal composition and the measured oxide contents of the starting raw powders in accordance with the methods employed by the Aluminum Association [19]. Hence, full theoretical density values of 2.707 g cm^{-3} and 2.699 g cm^{-3} were calculated for Al and Al–0.4Mg respectively. In SPS experiments, all powders were processed in the as-received state. However, when a press-and-sinter PM approach was employed, each powder was admixed with a powdered lubricant (1.5 wt% Licowax C; Clarient Corporation) to facilitate die compaction.

3. Experimental

Samples processed by SPS were consolidated using a Model 10^{-3} unit manufactured by Thermal Technologies Inc. This was completed in a vacuum atmosphere (pressure $< 8 \times 10^{-3}$ kPa)

Table 1
Concentrations of the elements detected in raw powders (wt%).

	Composition (wt%)					
	Si	Fe	Cu	Mg	O	H
Al	0.072	0.105	0.001	0.002	0.3000	0.0069
Al–0.4Mg	0.029	0.110	0.001	0.398	0.1730	0.0045

with graphite tooling so as to yield a sintered disc with nominal dimensions of 20 mm diameter × 3 mm thick. The graphite die was ISO-Carb85 with a thermocouple hole drilled into the lower punch to within 2 mm of the sample surface. Sinter profiles involved heating rates of 50 K min^{-1}, 100 K min^{-1} or 500 K min^{-1} to an isothermal hold temperature (400–600 °C) where samples were held for 30 s, 120 s, or 300 s. Current was pulsed-DC, with a 36 ms on, 8 ms off pulse profile. Sintered samples were then furnace cooled to ambient under the vacuum atmosphere. A uni-axial pressure of 50 MPa was applied to each sample through out the entire heating/cooling cycle. Larger (80 mm diameter × 15 mm) pucks were also consolidated via SPS but at FCT Systeme GmbH (Frankenblick, Germany). In an effort to maximize the relevance to lab-processed specimens, the majority of SPS processing parameters remained fixed for the larger specimens. These included a heating rate of 50 °C/min, sintering hold time of 120 s, and a uni-axial pressure of 50 MPa that was applied through the full sintering cycle. Isothermal sintering temperature was the key variable assessed with pucks produced at set points of 400, 450, 500, and 550 °C.

For conventional 'press and sinter' PM processing, an Instron 5594-200HVL load frame with a capacity of 1 MN was utilized for die compaction. All specimens were compacted at a fixed pressure of 200 MPa using rigid tooling that incorporated a floating-die concept. The geometries considered were flat dog bone specimens for tensile properties and discs (30 mm diameter × 4.5 mm height) for general sintering studies. Sintering was carried out under a slightly positive (30 kPa) pressure of nitrogen using a three-zone horizontal tube furnace equipped with a vacuum-tight stainless steel chamber. This chamber was evacuated and backfilled with high purity nitrogen (99.999%) twice, prior to the start of the heating cycle. A constant flow of nitrogen (34 m^3 h^{-1}) was then maintained during the entire sintering cycle. This thermal profile began with a 20 min isothermal hold at 400 °C for delubrication purposes. This was followed by a secondary hold for sintering (20 min at 630 °C) and cooling to ambient in a water jacketed section of the tubular retort. The heating rate was nominally 10 °C/min.

Preliminary characterization included density measurements (MPIF standard 42) and apparent hardness using the Rockwell H scale. Samples were ground planar with 240 grit SiC abrasive paper prior to hardness measurements, but after density observations. Sections were then cold-mounted in epoxy and polished using a standard series of abrasive papers and diamond compounds. Final polishing was obtained using non-agglomerating colloidal silica on a Vibromet vibrating polisher for up to 24 h. Optical imaging was completed using an Olympus model BX51 light microscope. All samples were etched with Keller's reagent prior to imaging. To assess tensile properties, round tensile bars were machined from the 80 mm SPS samples while conventionally sintered tensile samples were of a flat dog bone geometry. All machined specimens were prepared with a geometry that was compliant with ASTM E8-M [20]. Dog bone tensile samples were tested on the same load frame used for pressing, with a 50 kN load cell and a 25 mm gauge length extensometer. For machined round bars, a servo-hydraulic Instron load frame equipped with a 100 kN load cell was used. In both cases, an Epsilon Technology axial extensometer (model 3542) collected strain data, up to and