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Spark Plasma Sintering and Hot Extrusion of Aluminium Alloy Powder

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ABSTRACT

The Spark Plasma Sintering (SPS) is a promising sintering technology to produce dense bulk pre-compacts from micro- or nano-structured aluminium alloys at lower temperatures and shorter sintering times. The densification behaviour and sintering response of an atomized Al-Si alloy sintered by SPS was investigated. A high density of bulk material with porosity less than 1% can be prepared by SPS with a temperature of 450°C, holding for 2-5min and a pressure of 170 MPa. The particle size of the prealloyed powder influencing its electrical resistance plays a crucial role at the finding of the optimum processing parameters in case of a power-controlled sintering regime. In addition, the homogeneity of the microstructure across the diameter of the sintered samples are investigated and correlated with in-situ measurements of the temperature distribution within the samples. Residual porosity is mainly localised at the margin close to the wall of the die, corresponding to the lowest measured temperatures. The influence of the process parameters on the structure and tensile properties of subsequently hot extruded material was studied. The mechanical tensile properties of the SPSed compacts are significantly lower compared to that of the extruded material due to some residual porosity and the weakness of the joint between the initial atomized alloy powder particles. The extruded P/M material offers superior mechanical strength to the comparable compositions of known die cast piston alloys.

INTRODUCTION

Rapidly solidified P/M Al-Si alloys with higher content of silicon and transition metals are expected to be applied for automotive engine components such as pistons or connecting rods thanks to a low coefficient of thermal expansion, high wear resistance and excellent mechanical properties. Rapid solidification is an effective way to get a supersaturated solid solution, thermally stable dispersoids, metastable phases or amorphous phase, but limiting the thickness of the rapidly solidified structure, which is therefore available only within fine powders, thin ribbons or flakes. The challenge of producing a bulk product from such prealloyed powders is the need to perform a complete consolidation at the lowest possible temperature in a short time with preservation of the unique nano-/microstructure to the bulk material.

In this context, the recently developed field assisted sintering technology offers unique advantages in material fabrication compared to the conventional P/M techniques. It belongs to a class of sintering techniques that employ electric current to assist sintering [1], and Spark Plasma Sintering (SPS) is one of the most recent versions of these techniques [2-4]. Regardless the controversy to the presence/absence of sparking [5], important technological benefits such as short processing time, fewer processing steps, elimination of the need for sintering aids, and near net shape capability are unchallenged. In addition, the use of high heating rates and short dwell times can minimise grain growth, which often leads to improved material properties.

Several experimental studies [6-16] on the effect of pulsed current sintering on microstructure and mechanical behaviour of pure and alloyed aluminium powders demonstrated the capability of SPS in maintaining a very fine microstructures as prerequisite for a excellent mechanical performance of the bulk material. The breakdown of the oxide layers on the aluminium powder particles is one of the challenges for successful sintering of aluminium. The available experimental results suggest, the SPS process can promote the

elimination of the closed oxide layers and therefore promote the sintering by localised heating at the contact areas in combination with a sufficient pressure on the powder. In addition, the necessary degassing step for P/M aluminium should be also feasible in a short time by employing the sintering cycle to remove the adsorbed gas on the particles effectively. But, the measured tensile mechanical properties of aluminium compacts consolidated by SPS scatter between lower and comparable with that of an corresponding wrought or extruded compact [7, 11, 14, 15]. Therefore, it seems, that no comprehensive understanding regarding the influence of the SPS sintering process parameters and material characteristics exists today.
In this paper, a hypereutectic Al-Si alloy has been processed by SPS and subsequent hot extrusion to investigate the effect of relevant processing parameters. The effects of these parameters were investigated by means of microstructural analysis and tensile tests.

EXPERIMENTAL

The starting material was gas atomized powder sieved to <50µm of an Al-19Si-3,0(Fe,Ni)-2,5(Cu,Mg) alloy (supplied by ECKA Granulate GmbH, Germany) with a melting range of (530…680)°C and a density of 2,72 g/cm^3.
The pre-compacts were fabricated by SPS of the powder with a direct resistance heating for very fast heating using FCT-HP D 250 of FCT Systeme GmbH, Germany (Fig.1). The samples were heated by a pulsed electric current which flows through the punch-die-sample-assembly using a high current and low voltage. The current was forced through the powder charge by lining the inner sides of the steel die with a layer of an electrical insulator.

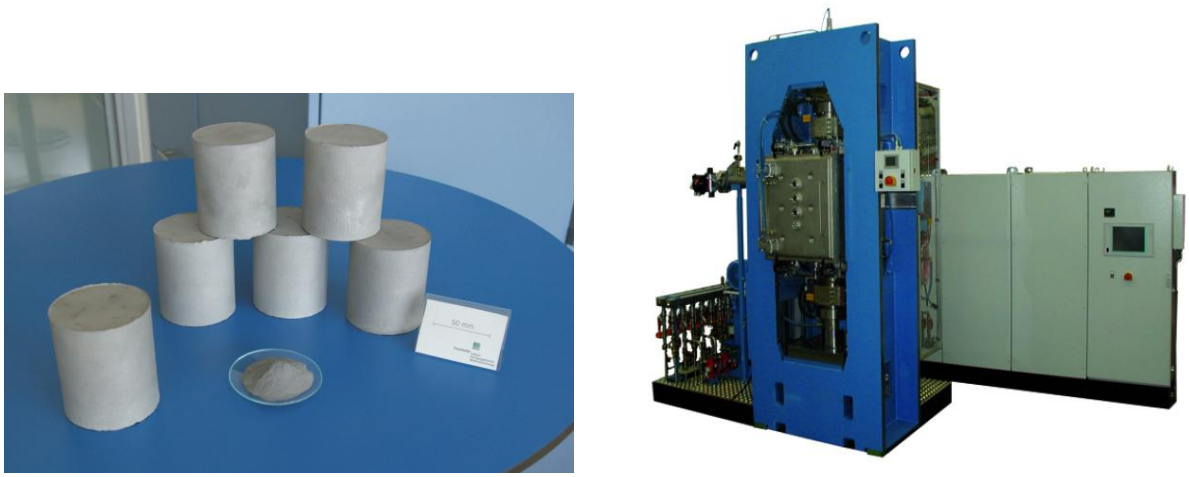

Figure 1. Aluminium pre-compacts (Ø 75mm, h 85mm, 99,3% theoretical density) consolidated by SPS using the FCT-HP D 250 of FCT Systeme GmbH, Germany.

All runs were executed in dynamic vacuum using one heating step from room temperature to the desired sintering temperature. After the desired holding time had elapsed, the power was shutt off and the assembly was allowed to cool down to room temperature. The pressure ranging between 170…220MPa was applied in two variants: 1. constant pressure for the whole sintering process, and 2. a low initial pressure of about 68 MPa was applied during the heating to 450°C and raised to the maximum value after several minutes. The second processing variant should lead to a more pronounced degassing of the powder which is necessary for a successful compaction. In some special experiments, additional thermocouples were placed inside the sinterbody to measure the temperature distribution during the SPS process [17]. Subsequently, the sintered pre-compacts were deformed by direct hot extrusion at 400°C to rods of 25mm in diameter with a extrusion ratio of 10:1.
The bulk density of the materials was measured by using a method based on Archimedes' law and compared with the theoretical density. Microstructures of the sintered and deformed materials were studied by optical microscopy. Tensile samples with a rectangular gauge area

of 31mm^2 and a gauge length of 35mm were machined from the SPSed pre-compacts and the extruded rods. Tensile properties were measured at room temperature using the ZWICK 1476-universal testing machine with a strain rate of 0,008s^{-1}. The Young`s modulus was determined by using the resonance frequency method. The Brinell hardness tests (HB2,5/62,5) were done at room temperature with a EMCO tester M4U-250.

RESULTS AND DISCUSSION

The relative densitiy of the SPSed pre-compacts is about 99,3% of the theoretical density independent from the sintering schedule. Residual porosity is clearly visible within the compact close to die (Fig. 2a). The optical micrographs reveal a very fine microstructure of the pre-compacts, the silicon therein having an average particle diameter of not more than about 4µm. In addition, the microstructure shows partly undeformed nearly original spherical powder particles, but in well contact among each other.

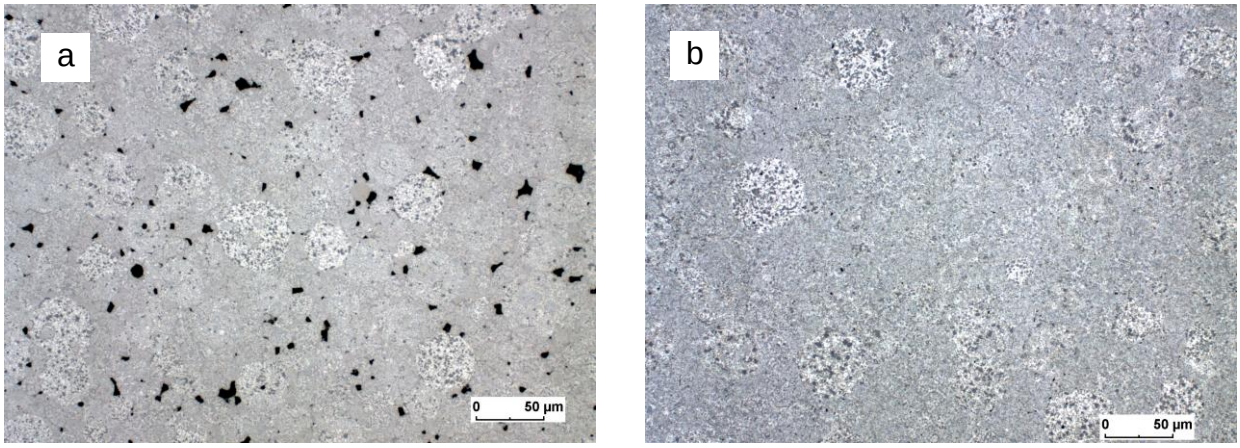

Figure 2. Optical micrographs of cross-sections at different radial positions in the middle of a SPS processed pre-compacts (a – close to the lateral shell, b – in the centre of the material).

The thermal analysis by using of additional thermocouples showed, that the temperature at the centre of the sinterbody is higher than at the margin close to the wall of the die. Especially a ring segment between 0 and 10 mm away from the die wall show significant lower temperatures compared to the inner part of the cylindrical disk. During the heating, the difference is decreases but in the thermal maximum, there is a temperature gradient with a ΔT = 44 K between the measuring points in the centre and 30 mm away (Fig. 3). The origin for this cooling effect arises from the used pressing tool setup. The pressing die is not connected to the electrical circuit, and therefore its temperature is relatively low.

For the time-controlled heating, a program with two fixed voltages (U_1, U_2) were used. Depending on the electrical resistivity of the pressed powder or the sinterbody, the maximum current was controlled by the automatic process control. This program results in two different time-current characteristics if coarse (> 50µm) or fine powders (< 50µm) were used as starting material (Fig. 4).

After the starting of the heating process (this means closing the electric circuit of the pulse generator), there was a significant delay for the generation of Joule heat. For fine powders, the delay is longer than for coarse powders. After several seconds during simultaneous increasing pressing force, the heating starts immediately. One origin for this behaviour may be the oxide layer on the surface of the aluminium alloy particles. When the layer is broken due to the deformation of the particles, the electrical circuit is closed and the heat generation starts. An evidence for this is the fact, that the power (U=const), which can be used for the heating increases much faster in case of coarse powders compared to fine powder, because the contact area is much larger.

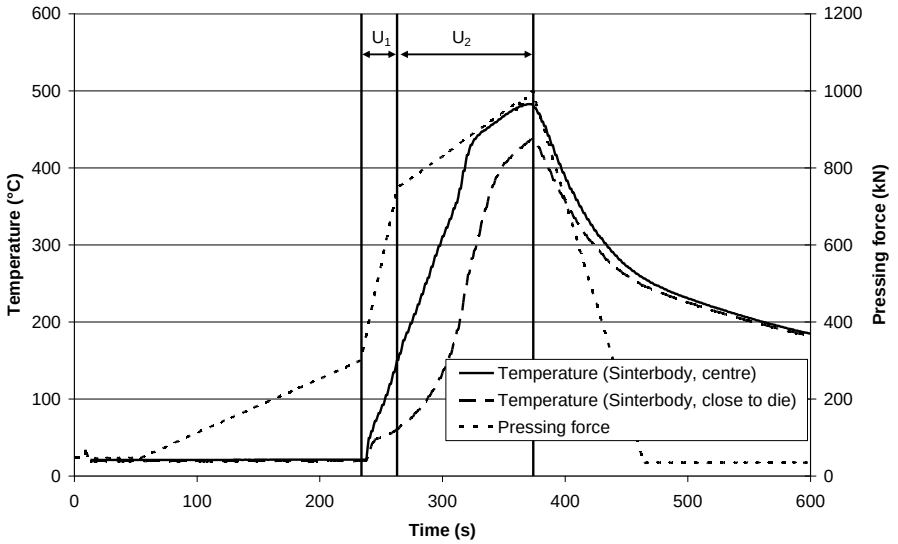

Figure 3. Time-temperature dependence inside the sinterbody during the Spark Plasma Sintering process. The measuring was done with external thermocouples, coming from the upper punch about 40 mm deep inside the sinterbody. One was placed in the centre of the sinterbody, one on the edge, about 10 mm away from the die wall.

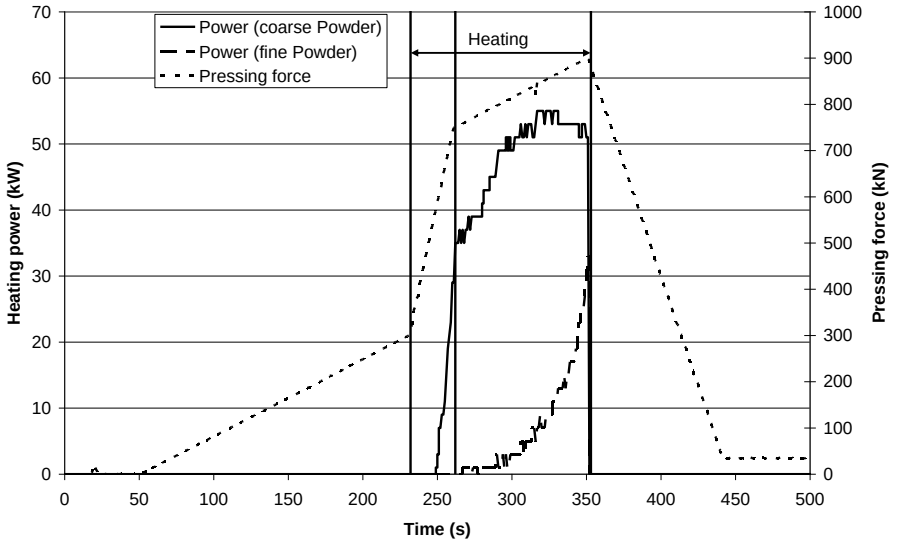

Figure 4. Time-(heating) power dependence during the Spark Plasma Sintering of Al-alloy powders with different particle size.

Figure 5 shows the microstructure at the central part of the extruded rods, exhibiting a retained fine and homogeneous microstructure, which is necessary to achieve a good mechanical performance of the consolidated material. In addition, some typical streamlines can be seen in the longitudinal section. The relative density of the extruded material is 100% theoretical density.

Table 1 presents a comparison of the mechanical properties of the materials fabricated by the SPS process and the combined consolidation, which is the combination of SPS with the direct extrusion.

Although the hardness value of the SPSed compacts is higher than that of the extruded materials, the tensile properties are significantly lower due to some residual porosity and especially the weakness of the joint between the initial atomized alloy powder particles. This confirms the results given e.g. in [14].

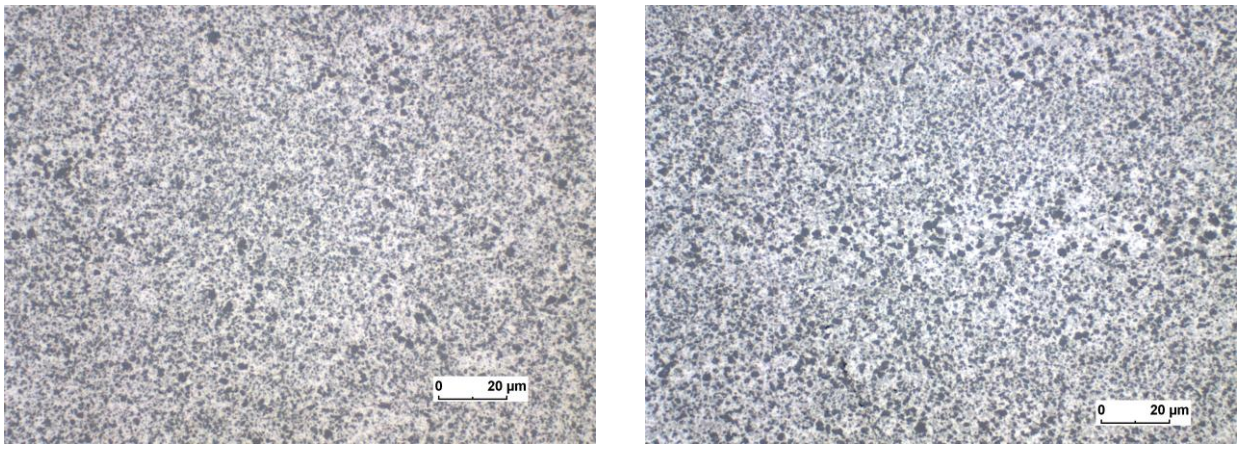

Figure 5. Optical micrographs of cross (left) and longitudinal (right) sections of a SPS/hot extruded rod.

	Relative density in %	Brinell hardness HB 2.5/62.5	Young's Modulus in GPa	UTS in MPa	Elongation A in %
SPS 2	99,3	180	n.d.	105…215	0
SPS 1 + extrusion	100	157	92	435	1,5
SPS 2 + extrusion	100	171	92	455	1,3
MAHLE 138 [18]	100	90…125	83	230…300	0,5…1,5

Table 1. Comparison of the mechanical properties of the consolidated materials produced by different methods.

The additional short degassing step shows no marked increase of the mechanical performance of the extruded material in contrast to the results given in [11, 13]. Further variations of the processing parameters are necessary for more understanding this degassing step during SPS.
The extruded P/M material offers superior mechanical strength to the comparable compositions of well known die cast piston alloys (e.g. MAHLE 138: Al-18Si-1,5Cu-1,3Mg-1,3Ni-0,5Fe).

CONCLUSIONS

A high density of hypereutectic AlSi bulk material with porosity less than 1% was achieved by SPS with a temperature of 450°C, holding for few minutes and a pressure of 170 MPa. Residual porosity is mainly localised at the margin close to the wall of the die, corresponding to the lowest measured temperatures. The mechanical tensile properties of the SPSed compacts are significantly lower compared to that of the extruded material due to the weakness of the joint between the initial atomized alloy powder particles. The selected procedure of an additional degassing step during the SPS results in only small effects on the mechanical properties of the deformed rods.
Additional variations of the relevant processing parameters of SPS and possible effects of subsequent heat treatments of the bulk material will be investigated in future.
The extruded P/M material offers superior mechanical strength compared to compositions of well known die cast piston alloys.

REFERENCES

[1] J.R. Groza, “Field assisted sintering”, In: *ASM Handbook*, 7, Powder Metallurgy, 1998, pp. 583-589.

[2] M. Tokita, “Industrial applications of advanced spark plasma sintering”, *American Ceramic Society Bulletin*, 2006, vol. 85, no. 2, pp. 32-34.

[3] M. Nygren, Z. Shen, “Novel assemblies via Spark Plasma Sintering”, *Silic. Ind. Spec.*, 2004, vol. 69, pp. 211-218.

[4] H.U. Kessel, J. Hennicke, J. Schmidt, T. Weißgärber, B. Kieback, M. Herrmann, J. Räthel, „Feldaktiviertes Sintern „FAST“ – ein neues Verfahren zur Herstellung metallischer und keramischer Sinterwerkstoffe“, *Proc. of Pulvermetallurgisches Symposium,* Hagen, Germany, 2006, pp. 201-237.

[5] D. M. Hulbert, A. Anders, D.V. Dudina, J. Andersson, D. Jiang, C. Unuvar, U. Anselmi-Tamburini, E. J. Lavernia, A.K. Mukherjee, “The absence of plasma in `spark plasma sintering`”, *J. of Applied Physics*, 2008, vol. 104, no. 3, pp. 033305-1 – 7.

[6] J.R. Ryu, K.I. Moon, K.S. Lee, “Microstructure and mechanical properties of nanocrystalline Al-Ti alloys consolidated by plasma activated sintering”, *J. of Alloys and Compounds,* 2000, vol. 296, pp. 157-165.

[7] Y. Murakoshi, T. Sano, I. Diewwanit, Y. Nakayama, S. Miyamoto, “Effect of extrusion on aluminium-lithium alloy composites sintered by SPS process”, *Proc. of the 1st Inter. Symp. On Spark Plasma Sintering (ISSPS-1)*, 2001, pp. 29-37.

[8] O. Ohashi, G. Xie, K. Chiba, “Spark plasma sintering of pure Al powder and bonding behavior between powder particles”, *Proc. of the 1st Inter. Symp. On Spark Plasma Sintering (ISSPS-1)*, 2001, pp. 45-53.

[9] Y. Shiomi, J. Yoshino, N. Kuroishi, “SPS sintered pre-forms for high strain rate super-plastic forming”, *Proc. of the 1st Inter. Symp. On Spark Plasma Sintering (ISSPS-1)*, 2001, pp. 159-162.

[10] G. Xie, O. Ohashi, N. Yamaguchi, “Sintering behaviour of aluminium powder by spark plasma sintering”, *Trans. of the Materials Res. Soc. of Japan*, 2002, vol. 27, no. 4, pp. 743-746.

[11] T. Nagae, M. Yokota, M. Nose, S. Tomida, K. Otera, T. Kamiya, S. Saji, “Microstructure and mechanical properties of gas atomized aluminium alloy powder compact densified by pulsed current pressure sintering process”, *Materials Trans.*, 2002, vol. 43, no. 3, pp. 537-543.

[12] C.Y. Xü, S.S. Jia, Z.Y. Cao, “Synthesis of Al-Mn-Ce alloy by spark plasma sintering”, *Materials Characterization,* 2005, vol. 54, pp. 394-398.

[13] K. Sastry, “Compaction of complex aluminium alloy powders for preparing industrial components through field assisted sintering technology (FAST)”, doctoral thesis, 2006, Katholic University Leuven.

[14] K. Hummert, R. Schattevoy, “Anwendungen für PM-Aluminium – Höchstleistung bei niedriger, aber voller Dichte”, *Proc. of Pulvermetallurgisches Symposium,* Hagen, Germany, 2007, pp. 229-245.

[15] M. Zadra, F. Casari, L. Girardini, A. Molinari, „Spark plasma sintering of pure aluminium powder: mechanical properties and fracture analysis“, *Powder Metallurgy*, 2007, vol. 50, no. 1, pp. 40-45.

[16] O. Ohashi, G. Xie, „Sintering of aluminium alloy powders using pulse electric current sintering process“, *Welding International*, 2008, vol. 22, no. 10, pp. 680-685.

[17] J. Schmidt, R. Niewa, M. Schmidt, Yu. Grin, Spark Plasma Sintering Effect on the Decomposition of MgH_2, Journal of the American Ceramic Society 88(7), 2005, 1870.

[18] D.M. Röhrle, „Kolben für Verbrennungsmotoren – Grundlagen der Kolbentechnik“, Verl. Moderne Industrie, Landsberg/Lech, 2001.