Processing of bulk Al7075 alloy by spark plasma sintering

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Abstract. The main advantages of powder metallurgy processing route are the possibility to produce near-net-shape compacts and to minimize the finish machining and material loss. The main problem in particle consolidation process is to suppress porosity, to remove oxide layers, and to retain the microstructure of powder materials. Spark plasma sintering (SPS) combines concurrent uniaxial pressure and direct heating by a pulsed DC current. Sintering occurs at relatively low temperatures for a short time and does not influence significantly the microstructure in the interiors of original powder particles. The efficiency of SPS in producing compacts with low porosity might be dependent on the distribution of particle size in original powder material. The gas atomized Al7075 powder was sieved to several charges and then sintered by SPS. Microstructure of sintered compacts was studied by light and scanning electron microscopy. The phase composition was investigated using X-ray diffraction. The mechanical behaviour was tested by bending tests.

1 Introduction
Powder metallurgy (PM) represents a very effective tool for processing metallic materials that can achieve properties superior to their ingot metallurgical counterparts. Atomization techniques make possible to produce powder particles at cooling rates up to $10^5 \text{ Ks}^{-1}$ which ensures the formation of materials far from thermodynamic equilibrium. The content of dissolved alloying elements might be enhanced significantly above the equilibrium solid solubility limit [1] and the microstructure of atomized powder particles is frequently fine-grained. The above mentioned aspects can improve especially the strength of PM materials.

Powder consolidation represents the main problem in the PM processing route. The current consolidation methods (e. g. high isostatic pressing) require generally a high temperature and a long time of sintering. During this period, the material tends to reach its thermodynamic equilibrium and many gains from atomization can be lost. The spark plasma sintering/field assisted sintering technology (SPS/FAST) [2] combines applied uniaxial pressure with heating by low voltage pulsed DC current flowing through the sample. High current density and large Joule heat can be evolved at contact points of powder particles where the temperature can highly exceed the set one. Sintering occurs at these points and the interiors of powder particles are nearly unaffected and retain the microstructure formed during atomization. This short time of exposition to elevated temperatures helps to avoid undesirable processes like recrystallization or grain growth. It is well known that the density and microstructure of sintered compacts can be influenced by many parameters of SPS (applied stress, heating rate, holding time, sintering temperature) [3]. On the contrary, there are scarce
data on the influence of powder particle size distribution which can play an important role especially at the beginning of sintering during re-arrangement of powder particles. The commercial Al7075 alloy was chosen for our experiments. This alloy is a typical precipitation strengthened material which derives its high strength from the precipitation sequence occurring either at room temperature (natural ageing) or at elevated temperatures (artificial ageing) after solution treatment. Our recent research has confirmed that fully dense compacts can be prepared from gas atomized powders using spark plasma sintering [4]. A bimodal microstructure with numerous small grains located along boundaries of original powder particles was observed after sintering [5]. There are two possible explanations how such bimodal microstructure can be formed:

- The small powder particles present in the gas atomized powder are re-arranged during the initial stage of sintering
- New small grains are formed during sintering at contact points between original powder particles where a high current density can result in a local melting and following solidification of the material

To solve this problem, the original gas atomized powder with a broad distribution of particle sizes was sieved to several charges and these charges were sintered separately. The microstructure, phase composition, porosity level, and mechanical properties were studied on these samples.

2 Experimental
The nitrogen atomized Al7075 powder was supplied by Nanoval GmbH & Co. KG, Berlin. The powder was sieved into 5 charges (<20 μm, 20–25 μm, 25–32 μm, 32–40 μm, and 40–50 μm) in an aqueous environment. The powders were sintered using pulsed electric current sintering system – model SPS 10-4 (Thermal technology, USA) at 425 °C under the pressure of 100 MPa for 4 minutes. The phase composition was verified by X-ray diffraction (XRD) using a diffractometer D8 Discover (Bruker AXS). Qualitative Rietveld analysis was performed to estimate the weight fraction of identified phases.

The microstructure of sintered compacts was studied by scanning electron microscopy (SEM) using microscopes EVO MA 15 (Carl Zeiss SMT) and FEI Quanta 200F. Image analysis was performed for porosity estimation. The chemical composition was determined by energy dispersive spectroscopy (EDS) and is given in Table 1. The arrangement of grains in sintered compacts was verified by light microscopy (LM) using microscope Olympus IX70. The three-point bending tests were performed at room temperature using Instron 1362 machine.

| Element | Zn | Mg | Cu | Al |
|---------|----|----|----|----|
| Wt. %   | 6.2| 2.7| 1.8| Balance |

3 Experimental results and discussion
Figure 1 shows the SEM micrograph of powders of the finest (<20 μm) and coarsest (40–50 μm) charges, respectively. The typical feature of the finest charge is a tendency to agglomeration of powder particles to objects of the size approaching nearly 1 mm. The coarsest charge contains not only powder particles exceeding 40 μm but also numerous very fine particles. The distribution of particle sizes is given in figure 2.

The phase composition of all compacts is given in figure 3. The compact prepared from the finest charge exhibits only Al matrix diffraction peaks. The absence of diffraction peaks from intermetallic phases might reflect the segregation free microstructure of the finest powder particles due to very rapid solidification. Some undulations of the diffraction curve can be explained by the presence of aluminium oxide hydroxide. This phase can be formed during sieving process occurring in water. All other compacts contain the MgZn2 phase (~3 vol. %) and also the Al2CuMg phase (~1 vol. %).
Figure 1. SEM micrographs of sieved powders, charge < 20 µm (a), charge 40 - 50 µm (b).

Figure 2. The distribution of particle size.

Figure 3. A part of the XRD pattern for sintered compacts.
The microstructure of the compact prepared from the finest charge is completely different from all other compacts (figure 4). The corresponding micrograph reveals that nearly no necks between original powder particles were formed during SPS. The measurement of porosity revealed the volume fraction of pores exceeding 10%. No coarse precipitates were observed in the interior of powder particles and the smallest ones seem to be even free of precipitates. This observation agrees well with the result of XRD analysis. The extent of porosity decreases with increasing size of powder particles. The compact prepared from the coarsest charge exhibits the volume fraction of pores not exceeding 1%. Two types of precipitates can be distinguished - coarser precipitates are located especially at the boundaries between original powder particles and finer precipitates are located in the interior of original powder particles, either along cell boundaries or homogeneously in the cell interiors.

The EDS analysis was performed to find the distribution of individual elements within the sintered compacts. Figure 5 documents an abnormally enhanced amount of oxygen at places between original powder particles in the finest compact (exceeding even 10 wt. %). These places are also rich in Mg. The distribution of other alloying elements (Zn, Cu) is homogeneous within the measurement accuracy. Much lower oxygen content was found in the coarsest compact (up to 0.5 wt. %). For comparison, the oxygen content below 0.2 wt. % was found in the compact prepared from non-sieved powder. The increased oxygen content in sieved materials can result from sieving process performed in aqueous environment. An enhanced Mg content was observed both at boundaries and in precipitates. Cu was found especially in the coarsest precipitates (see arrows), Zn mostly in finer precipitates. It can be concluded on the base of XRD analysis that coarse precipitates are probably
formed by the Al$_3$CuMg phase and the finer, more homogeneously distributed, precipitates are formed preferentially by the MgZn$_2$ phase.

Figure 5. Distribution of Al (a), O (b), and Mg (c) in the finest charge, distribution of Al (d), O (e), Mg (f), Cu (g), and Zn (h) in the coarsest charge, corresponding secondary electron image in (i).

Figure 6 shows the typical microstructure of the compact from the coarsest charge. At some places a perfect contact was formed immediately between large powder particles, at other places colonies of smaller grains were observed. SEM micrograph in figure 4b documented presence of imperfect contacts at these places. It can be therefore concluded that these small grains are small powder particles re-arranged during the initial stage of sintering [6] and not new grains formed from the melt during sintering.

Three-point bending tests at room temperature were used to estimate the ability of plastic deformation of sintered compacts. Whereas the compact prepared from non-sieved powder (containing a broad distribution of particle sizes) revealed plastic deformation, all compacts prepared from sieved powders exhibited fracture already within the region of elastic deformation. A tendency to increasing fracture stress with increasing powder particle size was observed. The explanation can be found in the increased oxide hydroxide phases in sieved materials. Simultaneously, the presence of powder particles with different sizes in non-sieved material improved the re-arrangement of powder particles during sintering and, therefore, less volume fraction of pores.
4 Conclusions

Powder particle size influences the phase composition of sintered Al7075 compacts. Higher solidification rate in small particles results in a segregation free microstructure, coarser particles contain numerous precipitates of the MgZn$_2$ and Al$_2$CuMg phases. Sieving of the powder material to different charges in aqueous environment supports formation of oxide hydroxide layers along boundaries of original powder particles, especially in the finest material. The compact prepared from the finest charge contains the highest volume fraction of this phase and exhibits the worst ability to form necks and also the worst deformation behaviour in bending tests. On the contrary, broader distribution of powder particle sizes supports sintering and leads to the best plasticity of sintered compacts. Re-arrangement of powder particles of different sizes plays an important role in the sintering process.

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