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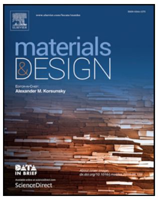


Solid state recycling of pure Mg and AZ31 Mg machining chips via spark plasma sintering

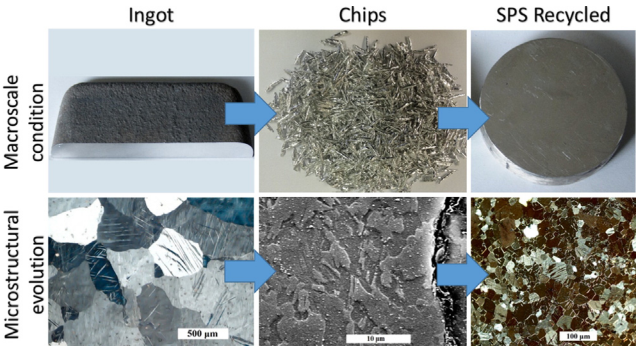
Dimos Paraskevas [a],*, Sasan Dadbakhsh [a], Jef Vleugels [b], Kim Vanmeensel [b], Wim Dewulf [a], Joost R. Duflou [a]

[a] *KU Leuven, Department of Mechanical Engineering, Celestijnenlaan 300A, B-3001 Heverlee, Belgium*
[b] *KU Leuven, Department of Materials Engineering (MTM), Kasteelpark Arenberg 44, B-3001 Heverlee, Belgium*

HIGHLIGHTS

- Spark plasma sintering is found to be a successful solid state recycling technique for the consolidation of Mg alloy chips.
- Fast and full densification/consolidation of Mg alloy machining chips directly into bulk semi-finished products was acheived.
- The total recycling route resulted in a finer microstructure than that of the parent material.
- The strong bonding and the finer microstructure of the recycled samples improved the mechanical properties.

GRAPHICAL ABSTRACT



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ABSTRACT

This work investigates the applicability of spark plasma sintering (SPS) as a solid state recycling technique for magnesium alloy scrap. In this respect, machining chips from pure Mg and AZ31 Mg alloy ingots are chemically cleaned, cold compacted and SPSed directly into bulk specimens. It is found that SPS can successfully establish full densification and effective metallurgical bonding between chips without altering compositional constituents. This is attributed to the dynamic compaction during sintering as well as to the disruption of the chips' surface oxide film due to SPS electric current based joule heating. Apart from the successful consolidation, microstructural analysis of the initial Mg ingots, chips and SPS recycled material reveals that the SPS microstructure was finer than that of the original ingots due to significant deformation induced grain refinement during machining. As a result, the recycled materials had a higher compression and shear strength than that of the starting ingot material. The findings indicate that SPS is an effective alternative method for solid state recycling of magnesium alloy scrap.

1. Introduction

Magnesium and its alloys have the lowest density of all structural metallic materials, with high specific strength, good machinability and dimensional stability. This leads to many applications, replacing heavier materials in aerospace, automotive, and transportation industries [1]. In addition to their lightweight applications, their good electro-magnetic shielding characteristics increase their usage in housing and electronic devices like mobile or laptop cases. Such a wide range of applications results in large amounts of scrap in the form of chips from Mg alloy machining (e.g., a 'buy-to-fly' ratio as high as 15-20 is not uncommon in aerospace applications). Therefore, increasing the recycling efficiency of Mg alloys is a relevant challenge. In fact, it is worth mentioning that

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* Corresponding author.
E-mail address: dimos.paraskevas@kuleuven.be (D. Paraskevas).

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magnesium is included in the latest European Union's raw material supply criticality list [2], which was motivated by its wide range of applications, the lack of viable substitutes, and the dependence on imports and trade.

Melting-based recycling of Mg alloys, especially in the form of chips, poses various challenges related to the thermodynamic constrains during melt purification, energy consumption and material losses. Hiraki et al. [3] simulated the Mg smelting processes based on chemical thermodynamic analysis, indicating which elements can be removed and how efficient the impurity can be controlled during metallurgical recycling. They concluded that the refining options are very limited for Mg, compared to other metals (such as ferrous metals), with only a few alloying and impurity elements that can be refined. Regarding the eleven alloying elements of Magnesium alloys, only Yttrium can be removed with oxidation from the melt to some extent. Consequently, open loop recycling (alloy mixing) results in quality and dilution losses during remelting (similar to aluminium remelting [4]). Moreover, metal losses during successive life cycle stages (recycling, semi-fabrication and part manufacturing) are also a major challenge. In fact, the high affinity of Mg to oxygen results in significant unrecoverable oxidation losses (since the metal property is lost) during remelting, especially for light-gauge scrap like chips due to their high surface-to-mass ratio.

The idea of recycling Mg alloy scrap by plastic deformation, in particular via hot extrusion, was patented by Stern in the USA already in 1944 [5]. Since then, many researchers focused on solid state recycling (**SSR**) of magnesium alloy chips directly into semi-finished products through various plastic deformation routes, and mainly by hot extrusion or extrusion based techniques (e.g. friction stir extrusion) [6–13]. As the Mg surface is always covered by a chemically stable and hard oxide layer, plastic/shear deformation aims at the physical disruption and dispersion of the oxide film to allow the formation of metallic bonds.

Spark plasma sintering (**SPS**), also known as field activated assisted sintering (**FAST**) or pulsed electric current sintering (**PECS**), has been reported as an advanced consolidation technique for aluminium alloy chips and sheet metal scrap [14,15]. Significant metal and energy savings can be achieved when avoiding remelting, as proved by Duflou et al. [16] who performed a comparative life cycle assessment (**LCA**) of three SSR routes for aluminium alloy scrap [14,17,18]. An environmental impact reduction with a factor 2–2.5 was found for the SPS route compared with its reference remelting route, with avoided material losses representing the most important gains. Both magnesium and aluminium alloys, and especially machining chips, are subject to similar energy and material efficiency losses during metallurgical recycling and further processing. Unrecoverable metal losses have to be compensated with primary material (substitution methodology) due to the limited scrap availability. For recycling of Mg alloy machining chips via SPS the impact reduction potentials are expected to be even higher compared to aluminium since: i) Mg has higher tendency in oxidation than aluminium, meaning higher oxidation losses during remelting; and ii) the energy intensity as well as the greenhouse gas emission of the primary magnesium production is much higher compared to aluminium. Moreover, further impact reduction as well as productivity improvements can be achieved by: i) improving the tool design and energy efficiency of the process; ii) reducing the overall cycle time; iii) upscaling (larger samples), and iv) process modifications/developments in order to move from batch processing to a semi-continuous approach (e.g. Spark Plasma Extrusion).

Despite the relatively extensive research on recycling by plastic deformation of magnesium chips [6–13], no sintering-based SSR technique capable to achieve full density (from machining Mg scrap) has been reported so far. Accordingly, this work aims at extending the available SPS recycling palette to magnesium alloy scrap. Therefore, it systematically investigates the feasibility and efficiency of SPS to produce bulk and semi-finished products directly from Mg and AZ31 Mg alloy chips. It also monitors the chip consolidation by SPS and analyses the microstructure and strength of the initial ingot verses the recycled material. These provide a clear understanding of the SPS applicability to SSR Mg-based alloys.

2. Materials and experimental procedure

2.1. Materials

Feedstock machining chips (Fig. 1b) with a length of 2–4 mm and <0.1 mm thick were produced by face milling of pure magnesium and AZ31 (Mg-3Al-1Zn) alloy ingots. The as-cast Mg ingot was twin-free (as observed in Section 3.1), whereas the AZ31 Mg ingot was deformed after casting with visible twins. The machined feedstock was chemically cleaned and loaded into a SPS graphite die to be compacted into cylindrical billets of 33 mm diameter for pure Mg (Fig. 1c) and 56 mm for

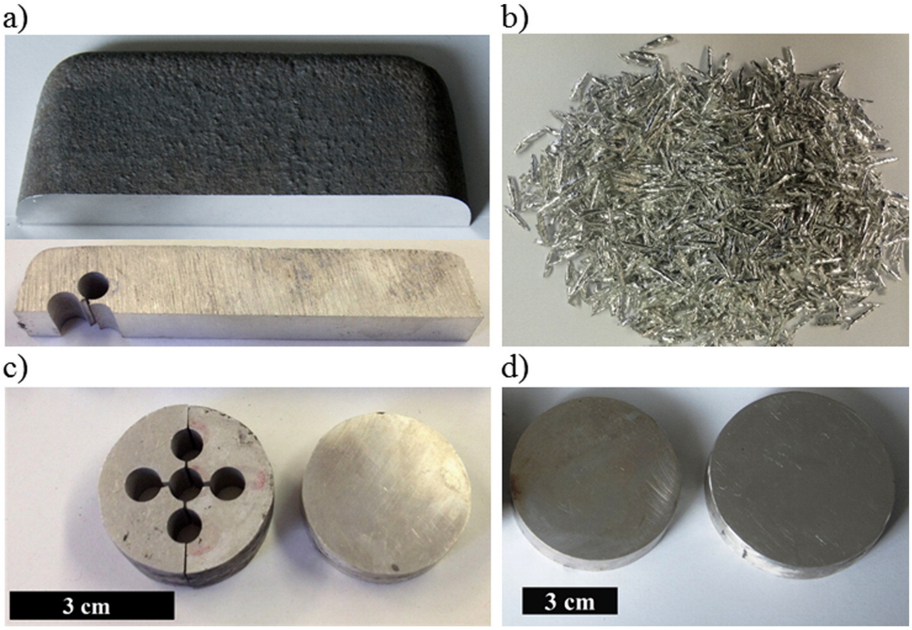

Fig. 1. (a) AZ31 (Mg-3Al-1Zn) (upper) and pure Mg (lower) starting ingots; (b) Chips produced by face milling; (c) recycled SPS Mg blank; and (d) recycled AZ31 SPS blanks.