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Semi-Solid Consolidation Processing of Rapidly Solidified Powders for Fabrication of High Strength AZ91 Magnesium Alloys

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Rapidly solidified AZ91 magnesium alloy was fabricated by a single roll technique. Microstructure of the alloys were composed of a fine-grain supersaturated α–Mg solid solution with a small amount of β-$Mg_{17}Al_{12}$ precipitates dispersed. The ribbons were successively crushed into fine powders using mechanical grinding apparatus and preformed at 673 K using pulse current sintering machine. The preformed compacts were extruded into a bar using semi-solid consolidation process at 723 K. The pressure needed to extrude preformed AZ91 compacts was 110 MPa about 40% lower than that of as-cast alloys. Semi-solid consolidated specimen after heat treatment showed high tensile strength 350 MPa. These were mainly attributed to grain refinement. The process proposed in this study led to the improvement of formability and also tensile strength for AZ91 alloys.

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Keywords: *rapid solidification, AZ91 magnesium alloy, semi-solid consolidation process, microstructure, formability, tensile strength*

1. Introduction

Magnesium alloys have the great potential for a variety of applications, for example, high-performance aerospace and automotive industries *etc.*, owing to a low density. They have high specific strength, but the strength itself of the alloys is less than that of aluminum alloys. Therefore, to improve the mechanical property of magnesium alloys is required.

By the way, magnesium alloys are very active in molten state, so SF_6 gas is used to prevent the combustion in foundries. Because SF_6 gas is one of the poisonous gases, the utilization has been suppressed. Semi-solid forming processing of magnesium alloys, for example thixo-molding, is now in practical use. This process makes it possible to produce near-net-shaped magnesium alloys products without SF_6 gas. In thixo-molding process, magnesium alloys are consolidated at high temperature nearby liquidus line, because high volume fraction of liquid phase is required. This leads to coarse grains, and higher strength of magnesium alloys is not expected well.

Rapid solidification processing is potentially one of the best methods to improve mechanical property of magnesium alloys. In a few decades, some researchers investigated mechanical properties of rapidly solidified Mg–Al–Zn alloys consolidated by extrusion.[1–4] They reported the tensile strength of Mg–Al–Zn alloys was clearly improved compared with that of the alloys prepared by ingot metallurgy. And they clarified the strengthening of the alloys was attributed to the obtained microstructures and/or non-equilibrium structures. But the powder metallurgy processing accompanied extrusion process is unfavorable for fabricating near-net-shaped products. So it is important for a wider industrial use to establish the processing which enables us to attain the improvement of mechanical property and formability.

The present authors have proposed semi-solid consolidation processing to produce near-net shaped Mg–Al–Zn alloys with high strength.[5] And also this process enables us to consolidate materials with no SF_6 gas. In the previous paper, machined and successively mechanically ground AZ91 magnesium alloy (Mg–9 mass% Al–1 mass% Zn) powders were extruded around the semi-solid temperature with a small volume fraction of liquid phase. The tensile strength of the compacts extruded at 723 K showed above 300 MPa and an excellent elongation of 14% was achieved. And also near-net-shaped product, gear part, was successfully consolidated with no crack. Therefore this process has the potential to prepare AZ91 alloys with near-net-shaped products as well as high tensile strength.

In this study, in order to improve the tensile strength, semi-solid consolidation processing of rapidly solidified AZ91 magnesium alloys was applied, and the mechanical property as well as formability of the alloys was investigated.

2. Experimental Procedure

2.1 Preparation of rapidly solidified AZ91 alloys

Commercial AZ91 alloy was rapidly solidified by remelting in a graphite crucible under an argon atmosphere and then melt-spinning onto the outer surface of the rotating copper wheel. The gap between the crucible nozzle and the wheel surface was maintained in 0.5 mm. The rotation speed of copper wheel was varied from 6 to 24 ms^{-1}. Special nozzle of the graphite crucible having 14 holes with the size of 0.3 mm in diameter was applied. It enabled us to obtain sound ribbons with no crack and smooth surfaces.

2.2 Preparation of AZ91 preformed compacts by pulse current sintering

Rapidly solidified ribbons were mechanically ground for 90 ks in an argon atmosphere to obtain fine powders. The powders were compacted into a disk measuring 10 mm in diameter and 10 mm in height using pulse current sintering at 673 K for 300 s in a vacuum. Pulse current sintering enables us to consolidate metal powders for a short time ($\sim$ 600 s). So, we can easily obtain high dense compacts consisting of fine grains. Furthermore, because sintering is performed using a graphite die in a vacuum, metals powders can be consolidated in a reducing atmosphere. As the result, oxidation to the pow-

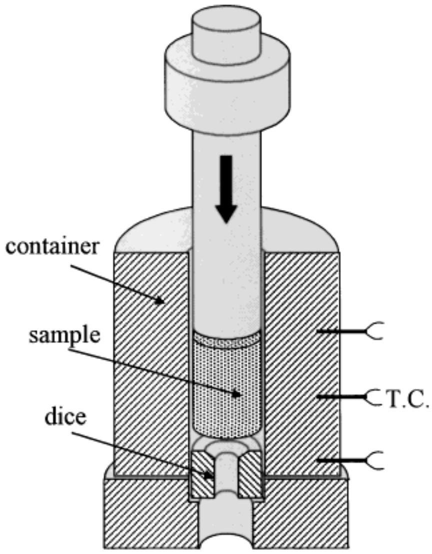

Fig. 1 Schematic illustration of semi-solid consolidating apparatus.

ders can be easily prevented. Preforming temperature 673 K is the minimum temperature to make porosity of the compacts less than 1 vol%. The reason to prepare high dense preformed compacts is described as follows; if the performed compacts contain a large amount of porosity, cracks are easily formed on the surface of extruded compacts. This is caused by residual gases trapped in pores.

2.3 Preparation of AZ91 alloy compacts by semi-solid consolidation processing

A disk was extruded at 723 K under a semi-solid state containing a very low liquid volume fraction with an extrusion ratio of 16:1. Figure 1 shows the schematic illustration of the apparatus for semi-solid consolidation processing. Consolidating temperature 723 K corresponds to the temperature which 10 vol% of liquid phase is produced in Mg–Al binary phase diagram. It is necessary to disperse the liquid phase homogenously in the preformed compact in order to extrude it such a low temperature. Therefore, infrared imaging furnace was applied for heating the samples so that the temperature in the samples was able to be held homogenously. Container for holding the samples and jigs for extrusion were made of hardened steel.

2.4 Evaluation of ribbons and consolidated compacts

Microstructure of rapidly solidified ribbons and extruded compacts was investigated first on metallographic cross-sections by optical microscope and electron probe micro-analyzer. Then the fine microstructure was investigated in detail using transmission electron microscope operating at 200 kV. The thin foils were prepared by ion thinning. Structural change was characterized by X-ray diffractometer with Cu–Kα radiation. Tensile strength of the extruded compacts were measured by Instron-type tensile testing machine at a strain rate of 10^{-4} at room temperature.

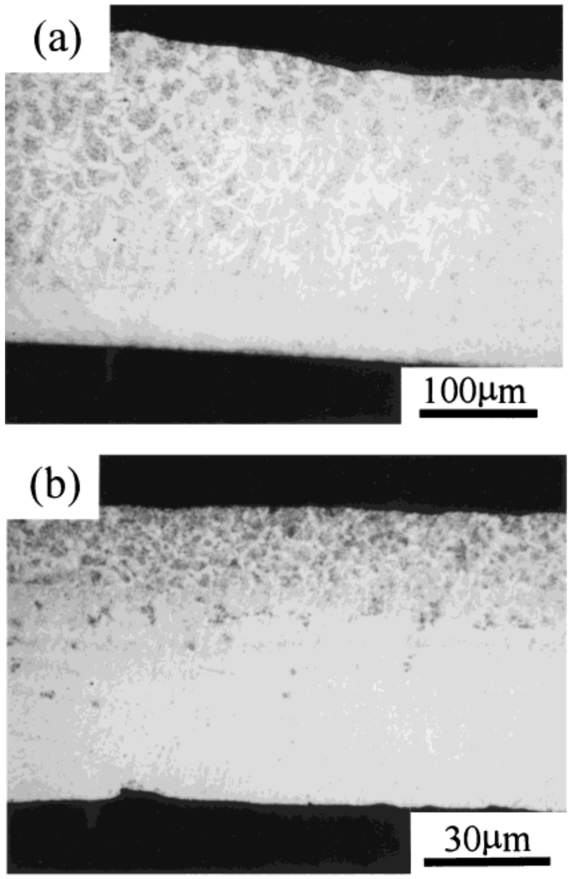

Fig. 2 Optical-micrographs of as-rapidly solidified ribbons prepared at the rotation speed of copper wheel (a) 6 ms^{-1} and (b) 24 ms^{-1}.

3. Results and Discussion

3.1 Microstructure of rapidly solidified AZ91 ribbons

The thickness of ribbons decreased with increasing the rotation speed of copper wheel, 3–4 mm wide and 200 μm for 6 ms^{-1} to 70 μm for 24 ms^{-1}, and did not change at above 24 ms^{-1}. Figure 2 shows the cross-sections of rapidly solidified AZ91 ribbons. Optical micrographs of both ribbons prepared at 6 ms^{-1} and 24 ms^{-1} indicate a lot of striations developing from the surface contact with the copper wheel (lower side of the figure) toward the other side. This suggests unidirectional solidification. The length of striations was measured to be about 60 μm for 6 ms^{-1} and about 30 μm for 24 ms^{-1}. Equiaxed grains can be seen from the front of striations to the top surface. Grain size of the equiaxed grains was measured to be almost the same as the spacing between the striations. Grain size decreased with increasing the rotation speed of copper wheel, too.

Figure 3 shows a bright field image of rapidly solidified AZ91 ribbons prepared at the rotation speed of 24 ms^{-1}. Grain size of the matrix was measured to be 0.5–2 μm, and finely dispersed particles of about 20 nm in the grains were seen. Matrix and nano-particles were analyzed using electron dispersed spectroscopy as follows; matrix: Mg 93.2 at%, Al 6.8 at%, nano-particles: Mg 56.1 at%, Al 32.8 at%, Mn 11.1 at%. Mn is added to improve the castability of AZ91 alloys.

From the result of X-ray diffraction analysis, two phases

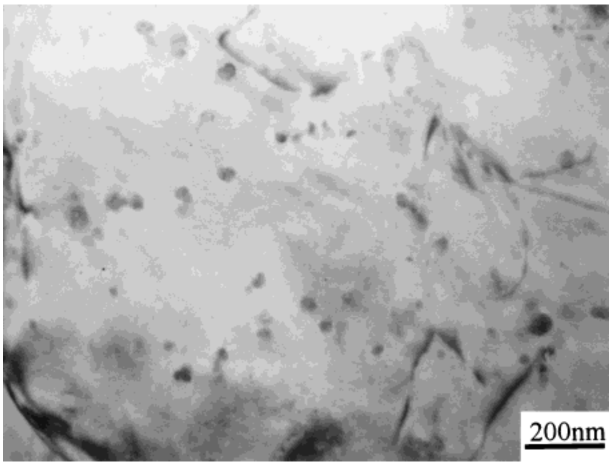

Fig. 3 Bright field image of as-rapidly solidified ribbon prepared at $24\,\mathrm{ms^{-1}}$.

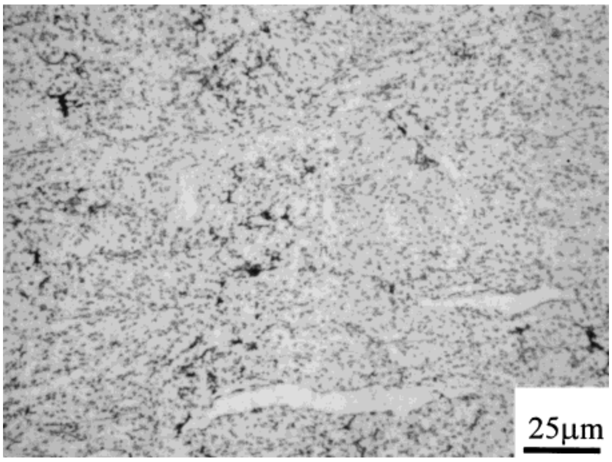

Fig. 4 Optical micrograph of preformed AZ91 specimen.

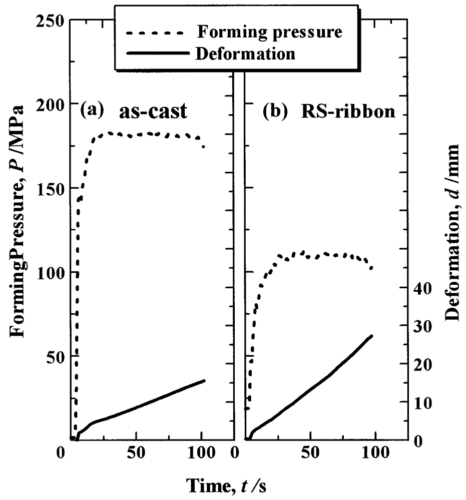

Fig. 5 Relationship between pressure, deformation and time for (a) as-cast AZ91 alloy and (b) RS-ribbon AZ91 alloy when semi-solid consolidation extrusion processing is carried out.

were identified. One of them was α–Mg solid solution, which X-ray peaks were shifted to lower angles, and one another $Mg_{17}Al_{12}$ compound. Equilibrium value of the room temperature solid solubility of Al in Mg is reported less than 1 at% in Mg–Al binary system. This reveals matrix phase is supersaturated α–Mg solid solution. Therefore, it is concluded that the structure of rapidly solidified AZ91 ribbon is composed of supersaturated α–Mg solid solution with nano-sized $Mg_{17}Al_{12}$ particles dispersed. Structure of rapidly solidified AZ91 alloys obtained in this study is roughly the same as that reported in the previous study.[6)]

3.2 Preparation of performed compacts by pulse current sintering

Rapidly solidified AZ91 ribbons were powdered into about 20 μm of particle size by mechanical grinding in an argon atmosphere. X-ray diffraction patterns of the obtained powders revealed broader peaks than those of as-ribbons. And also X-ray profiles of mechanically ground powders didn't change for more than 90 ks. This suggests the grain size in powders did not change. Mechanically ground powders thus obtained were preformed at 673 K using pulse current sintering method. Figure 4 shows the optical micrograph of the preformed AZ91 alloy compact. Porosity of the obtained compact was evaluated to be less than 2 vol%. The structure of preformed compact was composed of α–Mg solid solution with the size of 2–3 μm and $Mg_{17}Al_{12}$ precipitates with the size of 0.5 μm.

3.3 Formability of AZ91 alloy by semi-solid consolidation processing

The temperature dependence of deformation of as-cast and RS AZ91 alloys was investigated by thermo-mechanical analyzer under the constant load 1.25 kPa in argon flow. Both of the samples began to abruptly at around 710 K. Therefore semi-solid consolidating temperature was determined to be 723 K.

Figure 5 shows the time dependence of forming pressure, dislocation of cross head of semi-solid consolidation apparatus for preformed AZ91 alloy above mentioned and as-cast AZ91 alloy. Initial forming pressure necessary to transform preformed AZ91 alloy was measured to be 110 MPa, while 180 MPa for as-cast AZ91 alloy. That is to say, as for preformed AZ91 alloy, it is possible to extrude the sample at lower forming pressure. The reason is considered as follows; preformed AZ91 alloy has fine structures, so thin liquid phase is uniformly covered on the surface of α–Mg solid solution particles. This leads to thixotropy phenomenon. Therefore this proposed process enabled us to extrude AZ91 alloy at lower forming pressure.

Semi-solid forming processes are popularly in industrial use as a consolidating technique of magnesium alloys. But a volume fraction of liquid phase is over than 60% in most semi-solid processes, so dispersion of solid phase particles in the extruded samples can be easily non-uniform. In particular, liquid phase is inhomogenously dispersed in AZ91 alloy with larger grain sizes and is often squeezed out at the pointed end of extruded materials. But such a phenomenon wasn't observed in this study. This is because the structure of preformed AZ91 alloy was composed of fine grains 2–3 μm.

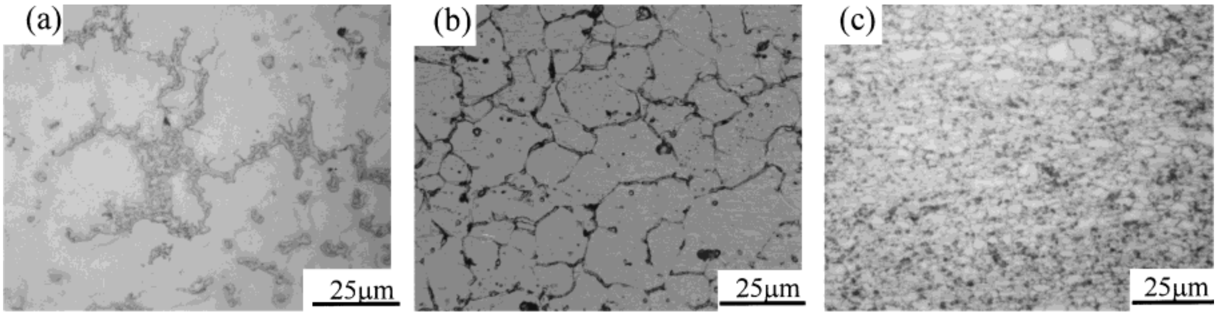

Fig. 6 Optical micrographs of AZ91 alloys prepared by (a) as-cast, (b) semi-solid consolidated as-cast and (c) semisolid consolidated RS-materials.

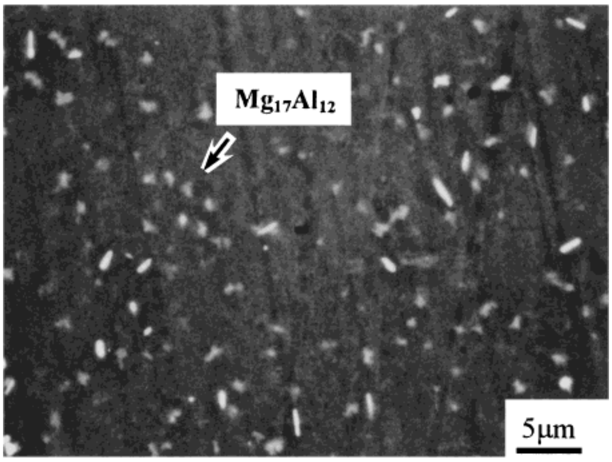

Fig. 7 Back scattering electron image AZ91 alloy prepared by semi-solid extrusion.

3.4 Mechanical properties of AZ91 alloys prepared by semi-solid consolidation

Figure 6 shows the optical micrographs of (a) as-cast AZ91 alloy prepared by a permanent mold casting, (b) semi-solid consolidated as-cast and (c) RS AZ91 alloy. The structure of as-cast AZ91 alloy is made up of large grains of 10–50 µm. This is probably attributed to the large latent heat of fusion generating when the liquid phase is solidified. On the other hand, semi-solid consolidated as-cast AZ 91 alloys has smaller grains of 5–20 µm, and semi-solid consolidated RS AZ91 alloy consist of fine grains of 2–3 µm. In this process, because of a small volume fraction of liquid phase at forming temperature 723 K, the latent heat of fusion is much smaller than that in permanent mold casting process. This leads to suppression of coarsening of grains.

The structure of semi-solid consolidated RS AZ91 alloy was investigated in detail using electron probe micro-analyzer. Figure 7 reveals a backscattering electron image of the alloy, showing finely dispersed $Mg_{17}Al_{12}$ precipitates in α–Mg solid solution matrix. Transmission electron microscopy observation clarified α–Mg solid solution matrix consisted of fine grains of 2–3 µm. According to energy dispersion spectroscopy analysis, Al composition in the matrix was analyzed as 4.6 at%, which value suggests Al supersaturated in Mg matrix. It is concluded that AZ91 alloy obtained in semi-solid consolidation process was made up of fine-grained supersaturated α–Mg solid solution and $Mg_{17}Al_{12}$ compound.

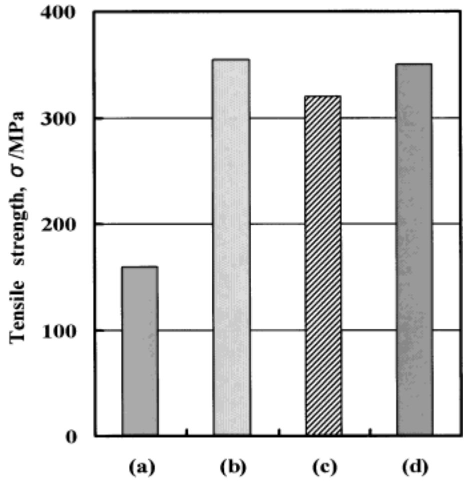

Fig. 8 Tensile strength of specimens prepared by (a) as-cast, (b) semi-solid consolidated as-cast, (c) semi-solid consolidated RS-materials and (d) semi-solid consolidated RS-materials after heat treatment.

3.5 Mechanical property of AZ91 alloy prepared by semi-solid consolidation processing

Figure 8 shows the tensile strength of (a) as-cast AZ91 alloy prepared by a permanent mold casting, (b) semi-solid consolidated as-cast and (c) semi-solid consolidated RS AZ91 alloy. Semi-solid consolidated RS AZ91 alloys showed high strength 320 MPa, which was more than double that of as-cast alloy. This is mainly due to grain refinement and solid solution strengthening as shown in Fig. 6. Furthermore, the tensile strength of (d) thermally treated at 443 K for 57.6 ks shown in Fig. 8 was measured to be 350 MPa, which was 30 MPa higher than that of as-consolidated specimen (c). This is probably due to fine $Mg_{17}Al_{12}$ precipitates from α–Mg matrix. Therefore, it is possible to strengthen as-consolidated samples in terms of adequate heat treatment. By the way, semi-solid consolidated as-cast AZ91 alloys revealed higher strength of 350 MPa than semi-solid consolidated RS AZ91 alloys. This is because the latter has more pores than the former between the particles. So reduction of pores leads to the improvement of the tensile strength.

H. Gjestland *et al.*[7] reported the mechanical properties of extruded RS-AZ91 alloys. They obtained higher tensile strength of 420–480 MPa at the extrusion ratio of 20:1, at the container temperature of 473–523 K, at the extrusion speed of 0.6–1.8 m/min. The structure of the obtained materials consisted of fine grain of 0.4–1 µm in terms of lower forming temperature relative to semi-solid consolidation process in this study. This leads to higher tensile strength. The tensile strength of semi-solid consolidated RS-AZ91 alloys shows lower than that of extruded RS-AZ91 alloys. But the extrusion process is unfavorable for fabricating near-net-shaped products with complicated shapes. On the other hand, proposed semi-solid consolidation process enables us to prepare AZ91 alloys with near-net-shaped products with complicated shapes such as gear part.[5)] In previous study, machined and successively mechanically ground AZ91 magnesium alloy powders were used for semi-solid consolidation. Initial forming pressure necessary to transform preformed RS-AZ91 alloy was measured to be 110 MPa, which was about 25 MPa lower than that of AZ91 alloy compact in the previous study. This reveals near-net-shaped products with complicated shapes such as gear part should be successfully fabricated. It is concluded that semi-solid consolidation processing using rapidly solidified ribbons makes it possible to produce AZ91 alloy with high strength and also good formability.

4. Conclusion

The following conclusions are drawn.

(1) Rapidly solidified AZ91 alloys have fine structures consisting of supersaturated α–Mg solid solution with the grain size of 0.5 μm, also with $Mg_{17}Al_{12}$ precipitates of 20 nm dispersed in α–Mg matrix.

(2) Preformed AZ91 alloy compacts of rapidly solidified ribbons were successfully fabricated by semi-solid consolidation process with no crack at 39% lower pressure than as-cast AZ91 alloy. This is referred to structure refinement.

(3) Tensile strength of AZ91 alloy prepared by semi-solid consolidation process revealed 320 MPa, about twice as high as that of as-cast AZ91 alloy. The value was improved to 350 MPa by means of adequate heat treatment.

(4) Proposed semi-solid consolidation processing has the potential to prepare AZ91 alloys with near-net-shaped products as well as high tensile strength.

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