Review Article

A Review on Magnesium Alloys and their Manufacturing Methods - A Future Need

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Abstract:

The present focus of automobile and aerospace industries is on development of fuel efficient systems. This drives the development of light weight alloys and cost-effective manufacturing of components, having desired shape and size, from these alloys. These alloys should have low density along with reasonably good strength and stiffness; offered by magnesium alloys both in soluble and insoluble form. With a weight approximately 30% lower than aluminum and 60% lower than steel, magnesium is the lightest structural metal. The impressive strength to weight ratio of the magnesium demands future research work to leverage its applications. In present practice world-wide, most of the magnesium alloy castings are produced through the High Pressure Die Casting (HPDC) process which incurs huge investment on tooling design and manufacturing. However, a few attempts has been made by the researchers to manufacture magnesium alloy components using sand and other sand-based casting processes, encountering quality related problems. The present paper identifies development of quality and cost-effective casting methods for manufacturing of magnesium alloy components as a major research problem. In the first part of the paper, a brief review of literature on magnesium alloy casting manufacturing methods is presented along with their advantages and limitations. The second part of the paper presents future research directions along with supporting theoretical background for effective manufacturing of magnesium alloy castings with investment casting route. Major problem in the investment casting of magnesium alloys is the interfacial reaction between magnesium melt and ceramic oxide mould due to the higher affinity of magnesium to oxygen. These reactions deteriorates surface properties of magnesium alloy castings that is the major challenge faced by investment casters. This article suggest that the modification in investment casting process to produce successful magnesium castings with suppression of interfacial reactions is essential that will also allow automotive and aircraft industries to get advantage for production of lighter and economical products with high precision.

Keywords: Magnesium Alloys, Processing of Magnesium Alloys, Manufacturing Methods, Magnesium Casting.

INTRODUCTION

Magnesium (Mg) is one of the most abundant materials on the earth and when used as a material for manufactured components, it provides a unique combination of properties, including light weight, strength and stiffness. In addition, magnesium alloys can offer good castability, superior machinability, good weldability and excellent damping characteristics. To produce magnesium components using casting route is one of the economical and major manufacturing processes that can be used in variety of applications (Yang et al., 2007; Sahoo, 2011).

Properties of Pure Magnesium

Table 1 lists some important physical properties of pure magnesium. Pure magnesium is mainly used as an alloying element. Mechanical properties of pure magnesium are shown in Table 2, from that it can be observed that pure magnesium is soft and mechanically weak that requires alloy development to enhance mechanical properties (Friedrich and Mordike, 2006).

Magnesium Alloys

Main commercial magnesium alloys include the AZ series (Mg-Al-Zn), AM series (Mg-Al-Mn), AE series (Mg-Al-RE), EZ series (Mg-RE- Zn), ZK series (Mg-Zn-Zr), and WE series (Mg-RE-Zr). Commercial cast magnesium alloys for automotive applications belong to AZ and AM series alloys (AZ91D, AM50A, and AM60B) (Yang et al., 2008).

Magnesium alloys are used to produce components for Automotive, Aerospace, Medical, Sports, Electronic, and other applications.

Table 1. Physical Properties of Pure Magnesium (Magnesium encyclopedia)

Property	Value
Melting point	650°C ± 2
Boiling point	1107°C ± 10
Latent heat of fusion	0.37 MJ/kg
Latent heat of evaporation	5.25 MJ/kg
Heat of combustion	21.5 MJ/kg
Specific heat At 20°C At 600°C	1013 J/kgK 1178 J/kgK
Electrical resistivity at 20°C	4.45 μΩcm
Thermal conductivity at 25°C	155 W/kg K
Linear coefficient of thermal expansion at 20°C	$25.2\times10^{-6}\ K^{-1}$
Density At 20°C At 600°C At 650°C (solid) At 650°C (liquid)	$1.738\ g/cm^3$ $1.622\ g/cm^3$ $1.65\ g/cm^3$ $1.58\ g/cm^3$
Volume change during solidification	4.2%
Volume change during cooling 650-20°C	5%

Table 2. Mechanical Properties of Pure Magnesium (Magnesium encyclopedia)

	Tensile strength MPa	Tensile yield strength MPa	Compressive yield stress MPa	Elongation % 50 mm	Brinell Hardness 500 kp/10 mm
Sand cast thickness 13 mm	90	21	21	2-6	30
Extrusion thickness 13mm	165-205	69-105	34-55	5-8	35
Hard rolled sheet	180-220	115-140	105-115	2-10	45-47
Annealed sheet	160-195	90-105	69-83	3-15	40-41

Magnesium has a long history of automotive use, the first automotive magnesium application was the racing engine pistons for "Indy 500" in 1921 developed by Dow Chemical in the United States (Alan, 2013). During the time of world war I & II, magnesium was used extensively for german military aircrafts. Consumption of magnesium in automotive sector accelerated and reached to a peak in 1971, the major applications were in the air-cooled engine and gearbox castings (Brown, 2008). Currently, magnesium has made significant gains in world-wide interior applications, replacing mostly steel stampings in instrumental panels, steering wheels and steering column components. Europe is aggressively expanding the use of magnesium in engine blocks and transmission cases using recently developed creep-resistant magnesium alloys. Only a limited number of body and chassis components are currently made of magnesium, which presents a great opportunity for magnesium to expand its applications in light weight vehicle construction.

Magnesium Vision 2020 originated from a meeting of the United States Automotive Materials Partnership (USAMP), Automotive Metals Division (AMD) proposes to reduce the weight of an average 1524 kg vehicle by 131.5 kg, by replacing 285.75 kg of current aluminum, iron and steel with 154.25 kg of magnesium (Magnesium Vision 2020, 2011).

Processing Methods

Different routes of magnesium alloy processing are casting, forming, joining, and machining. Magnesium alloys can be processed by casting processes like sand casting, die casting, investment casting, squeeze casting, stir casting, semi-solid metal casting, In-situ synthesis, etc (Gupta et al, 2007). Statistically, more than 90% of the magnesium alloy structural components are produced by casting process (Yang et al., 2008).Due to excellent properties and potential application of magnesium alloys, there was continuous increase in magnesium casting production noted in past two decades. It can be seen that from year 1995 to 2015 annual magnesium casting production is almost doubled as shown in graph given in Fig.1 (AFS casting census). However there is still limited production of magnesiumcasting as compared to other ferrous and non-ferrous metals.

According to 49th census of annual world casting production December, 2015 published by AFS annual Mg casting production was about 160808 metric tons which contributes only 0.153% in total ferrous & nonferrous annual casting production as shown in Fig.2 (AFS casting census).

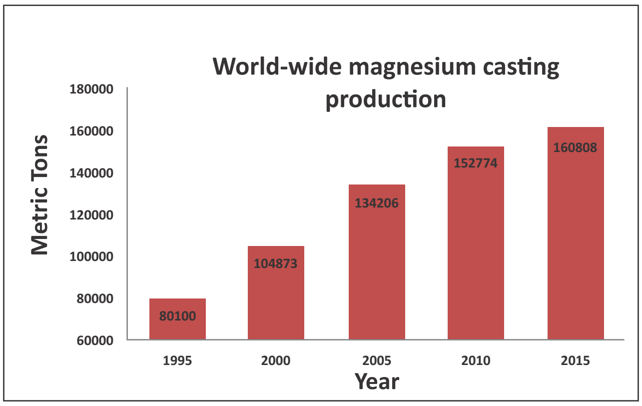

Fig. 1. Year wise worldwide magnesium casting production in metric tons (AFS casting census)

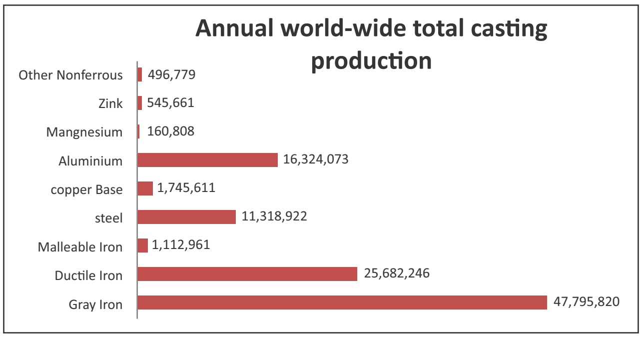

Fig. 2. Annual worldwide total casting production in metric tons for year 2015 (AFS casting census)

LITERATURE REVIEW

The present paper identifies casting as a major processing technique for production of magnesium alloys. Process-wise review of the literature is presented in this section.

Die Casting

Manufacture of parts using die casting is relatively simple, especially suited for a large quantity of small to medium sized castings. High pressure die casting of magnesium is a proven manufacturing process by which products are being produced in volume, from tiny electronic parts to truck transmission cases with the unique ability to transform the injected molten magnesium into an accurately dimensioned and smoothly finished form in the shortest possible cycle time (Friedrich and Mordike, 2006). About 80% of all magnesium alloy castings made worldwide are manufactured with High Pressure Die Casting (HPDC) route. Due to higher fluidity magnesium alloys may be used for castings with thinner walls (1–1.5 mm) than is possible with aluminium (2–2.5 mm) or plastics (2–3 mm). Latent heat of fusion per unit volume of magnesium is two-third times lower than that of aluminium, therefor magnesium cools rapidly.

Iron from the dies has very low solubility in magnesium alloys, which is beneficial because it reduces any tendency to die soldering (Polmear et al., 2017) However, there are some limitations with HPDC process. High Pressure die cast magnesium alloy contains both shrinkage and gas micro porosity. Leo et al. (2005) have suggested that turbulence created during die filling under high pressure is the main reason for porosity defects that limits further heat treatment of the produced castings as that porosity expands during heat treatment which leads to the formation of cracks. Balasundaram and Gokhale (2001) have observed that the microstructure of the cast Mg alloys almost always contains some porosity, and in some cases, it may also contain processing defects such as hot-cracks, oxide skins, and flux inclusions. In die casting of magnesium alloys hot tears generally nucleated in the casting skin regions and propagated towards casting interior. This defect of hot tears generally found with mould temperature of upto 220°C, with further increase in mould temperature hot tearing decreases significantly (Bichler et al., 2008). A mold coating that does not react with magnesium melt is a critical step in advancing magnesium die coating technology for high volume applications. Mold coating research has indicated that traditional coating react with molten magnesium and formation of magnesium oxides and other reaction compounds rapidly deteriorates coating life. Use of coatings developed with fluorspar and silicate based binders found to maintain longevity and lower wear rate of die due to reduced reactions with magnesium. (Lafay and Robison, 2007)

The main disadvantage of the die casting is the very high capital cost involved. The primary and supporting tooling required is costly as compared to other casting processes. Therefore, to make die casting an economical process, a large production volume is needed. In addition, due to high heat conductivity of metal moulds, castings continue to suffer from the low specific heat capacity of magnesium, which results in rapid freezing and difficult to filling of thin-wall castings. Therefore, the process is limited to high-fluidity metals (Degarmo, 2003).

Sand Casting

The basic principles of sand casting for magnesium alloys do not differ greatly from those of other commercial metals. However, to produce castings of satisfactory quality, it is necessary to take into account some of the significant differences in physical properties between magnesium and other common metals. The molten magnesium reacts readily with many basic moulding materials including silica sand and moisture. Molten magnesium oxidizes readily forming voluminous oxides, which may be entrained in the metal. Density of magnesium is lower than other conventional casting metals. Therefore, in simple gravity casting, there may relatively little metallostatic pressure to fill the mould. To compensate for low volume heat capacity, risers need to be relatively large to provide adequate feeding (Friedrich and Mordike, 2006).

The sand mould leaves bad surface finish, due to sand indentation and oxidizing medium, which often requires further processing (Rao, 2003). Bichler and Ravindran (2010) found that alloying with elements of limited solubility and miscibility often results in inhomogeneous casting microstructure and contributes to poor casting quality. These defects, along with solidification shrinkage porosity, limit the range of casting designs and the utilization of Mg alloys in many applications. Takamori et al. (2008) have observed that reactions were found lesser on the magnesium alloy casting casted with chromite sand mold as compared to the casting with silica sand mold. They also suggested from their experiment that the wash of silica sand mold with boron nitride helps to suppress the interfacial reactions to some extent. Winardi et al. (2008) have investigated the effects of magnesium melt reactivity in incresing he tendency of blow defects in magnesium castings. They have analysed the gas evolution from cores made with various types of binder, coating and additives. During expermiments cores were immersed in magnesium melt to measure the volume and rate of evolved gas from reactions.

Investment Casting

The investment casting process (also known as 'lost wax casting' or 'precision casting') is one of the ideal liquid metal processing methods for producing the near-net-shape structural parts for the aluminum alloys and nickel-based super alloys. Recently, there have been attempts by several researchers to develop investment casting process to achieve higher quality in magnesium alloy castings by using different techniques and methods. Investment casting is one of the reasonable solutions for magnesium alloys that suffer from difficulties in forming in the solid state due to the hexagonal lattice structure of magnesium (Yang et al., 2007; Rao, 2003). Hong et al. (2000) have performed experiments with different mould preheating temperature and pouring temperature to analyse its effects on tensile strength and hardness. Lun Sin et al. (2007) have poured magnesium alloy AZ91 in investment moulds by top and bottom filling systems. They observed that top filled specimens exhibit flow marks on surface that are the surface defects usually associated with turbulent flow whereas, in bottom filled specimen these defects were absent. Elsayed et al. (2010) have suggested that use of Al-Ti-B based grain refiner enhances grain refinement and mechanical properties of Mg alloy AZ91 casting. The main problem with the investment casting of magnesium alloys has been the high tendency of magnesium to react with ceramic investment casting mold. This reaction causes the surface finish of the casting to deteriorate, which is the main issue for the majority of researchers and magnesium foundries.Kim M. and Kim Y. (2002) have suggested from their investigation that magnesium melt dissolves mould materials into melt. Oxygen dissolution may increase hardness on surface of solidified casting while, dissolution of silicon and zirconium may results in inclusions in the produced casting. Idris et al. (2014) have developed in-situ melting and solidification technique for magnesium investment casting found to be reduced reactions to some extent, but problems related to inhomogeneous fusion and solidification of granules were noted with this technique. Lun Sin and Dube (2004) have observed from experimentation that vacuum assisted investment casting for magnesium alloy helps to improve fluidity of magnesium melt to fill thin wall castings and to obtain better surface quality. Andres et al. (2011) have analyzed the effectiveness of protective SF_6 gas and inhibitors $NaBF_4$ and KBF_4 on interfacial reactions of magnesium castings. Arruebarrena et al. (2007) have poured magnesium AZ91 alloy in investment molds with different proportion of SF_6 gas with CO_2 carrier gas and observed that 9% SF_6 concentration eliminated oxidation on casting surface at greater extent. Under the SF environment, liquid Mg wets the solid MgO and forms the stable and denser layer of MgO. This dense layer minimizes exposed surface area of the melt to environment reducing the chances of burning and oxidation (Cashion et al., 2002). Higher global warming potential about 23,900 of SF_6 demands use of some alternative protective gas in magnesium sand and investment casting. Okhuysen (2011) has evaluated effectiveness of Novec 612 (fluorinated ketone – $C_6F_{12}O$) gas as an alternative of SF_6. This gas protects the melt more effectively since each molecule of Novec 612 contains twice as many fluorine atoms as SF_6. Thus, it can be used at a much lower concentration than SF_6 (0.015 to 0.4 volume % or 150 to 4000 ppm V compared to 0.7 to 6% SF_6). In contrary the molecules of Novec 612 breaks down faster at the higher temperatures therefore, single point source of it during melting and pouring is not sufficient (less coverage).

In order to prevent the reaction of magnesium with oxygen, it is first necessary to prevent atmospheric oxygen from contacting the metals, and it is necessary to prevent reaction between the metal and the mould materials. The metal-mould reaction can be prevented by an oxide-free mould being used or inhibitor/barrier being present to prevent the reaction (Sahoo, 2011).

Summary

Although, almost 90% of magnesium alloy castings are manufactured using metal moulds especially High Pressure Die Casting (HPDC) route, it encounters number of problems. Process includes huge cost on tooling design and development and is mainly limited by size and weight of the components. In addition, better understanding about melt fluidity under high pressure is desirous to get quality castings. On the other hand, sand (silica) mould based process route offers lower process cost but suffers from problem of reaction at melt-mould interface, poor surface quality and dimensional accuracy. In this situation, economical and precision manufacturing of magnesium components using Investment Casting (IC) process would be a potential alternative to explore. However IC process suffers from many limitations offered by the sand mould based processes. Future research as presented in the next section is required to address these limitations.

FUTURE RESEARCH DIRECTIONS

Investment casting process allows near net shape and the degree of freedom in design is larger in contrast to pressurized casting processes, such as die casting and semi-solid metal processing. The high reactivity of molten magnesium requires specific melting and casting processes as well as specific moulding materials. Furthermore, the study to determine optimum casting parameters such as casting and mould temperatures is also necessary (Hong et al., 2015). This section, presents future research work required to suppress the mould-metal reactions and to manufacture quality magnesium castings using Investment Casting (IC) process.

Suppression of Mould-Metal Reactions with Unconventional Stable Ceramic Oxides

Investment Casting moulds are primarily manufactured by applying number of coats (depending on mould thickness required) of slurries consist of ceramic oxides and binders over suitable pattern material. Subsequently, pattern material is drained out to get hollow mould cavity. Magnesium has higher affinity towards oxygen and it undergoes reactions when comes in contact with ceramic oxides. These reactions are primarily responsible for the failure of investment moulds as well as deterioration of casting surface properties and characteristics.

Cingi (2006) has investigated thermal stability for various oxides and found that ceramic oxides with higher thermal stability found to resist the reactions to some extent. Idris et al. (2013) have suggested that use of Inhibitors and protective gas also helps to overcome the reactions. Vyas and Sutaria (2016) have suggested Alumina as a face coat oxide material instead of conventional Zircon Flour face coat from the results of TGA/DSC analysis, to suppress the exothermic interfacial reactions. Modifications in conventional investment mould materials and exploration of new mould materials along with new mould development methods may be helpful in suppressing interfacial reactions to produce components of enhanced quality economically with cut down in consumption of costly protective gases.

Evaluation of Mould-metal Reactions based on Interfacial Solidification Analysis

Time temperature cooling curves can facilitate in quantification of degree of reaction at melt-mould interface during solidification. In general, these reactions are exothermic in nature due to generation of MgO. The liberated heat retards the initial solidification rate and further enhances degree of reaction as liquid magnesium gets more time before it solidifies.

Thermal history at the melt-mould interface can be recorded by placement of K type thermocouple at the interface and connecting it with data acquisition system. Cooling curves for different mould materials can be compared that helps to determine heat transfer characteristics of moulds prepared with different face coat slurry material.

Influence of Mould and casting geometry on Mould-Metal Reactions

Mould thickness depends upon number of layers of ceramic invested on wax pattern. As the thickness varies permeability of mould also varies. Therefore role of permeability for interfacial reactions needed to be evaluated. Investment moulds can be poured with $SF_6 + CO_2$ protected magnesium melt. Moulds can also be purged with protective gas prior to pouring by giving continuous supply at pouring cup (Idris et al., 2013). After pouring and during solidification, there may be the access of oxygen to melt from atmosphere through the permeable pores of mould. Oxygen is not responsible to only initiate the reaction but also facilitates continuation of exothermic reaction, leading to generation of local heat. Mould permeability can also be increased by addition of nylon fibers in the slurry as fibres are burned out during firing of shell, which creates pores (Jones and Yuan, 2003).

Interfacial reactions may also be influenced by casting section thickness. As section thickness changes modulus of casting changes and therefore solidification rate at interface also changes. The contact period of melt with mold increases the chances of metal mold reactions. Quantifiaction of reactions on surfaces of stepped geometry casting with different section thickness may help to detrmine the effect of casting thickness on reactions. Cooling curve at interface may helpful to justify the reactions at interface.

Evaluation of Reactive Wetting Kinetics at Elevated Temperature for Metal/Ceramic Oxide System

Understanding high-temperature wetting behavior between liquid metal (melt) and solid ceramic (mould material) is critical for improving the surface quality of the cast products.

In general, wettability refers to the ease with which a liquid spreads across a solid surface. Contact angle (CA) is the main parameter introduced by Young that correlates wetting with interfacial tensions of the solid, liquid and vapour interfaces using simple considerations of the equilibrium at the triple line, i.e., the line where the solid, liquid and vapour phases come in contact as shown in Fig. 3 (Duncan et al., 2005).

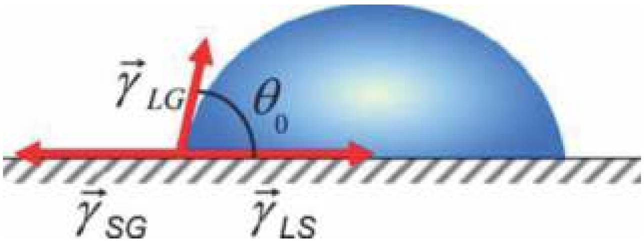

Fig. 3. Contact angle at three phase contact line

$$\cos\theta = \frac{(\sigma sl - \sigma sg)}{\sigma lg} \qquad (1)$$

At high temperature, complex chemical reactions at melt-atmosphere and melt-solid ceramic interface increases, leading to significant changes in interface structure and chemistry thus, affecting the surface properties of cast products. In non-reactive wetting systems, there is no significant-transient change in contact angle and system reaches to the equilibrium in a short time of milliseconds. Whereas, in presence of reactions, the formation of interfacial compounds lead to gradual decrease in contact angle as surface energy changed and takes more time to reach the equilibrium. In addition, dissolution of solid ceramics into liquid occurs that influences the wettability. Therefore, it is most necessary to study wetting kinetics for reactive systems described by the Equation. (2) (Eustathopoulos et al., 1999).

$$\cos\theta(t) = \cos\theta(o)\ \frac{(\Delta\sigma(t) \quad (\Delta G(t)}{\sigma lg(o) \quad \sigma lg(o)} \qquad (2)$$

$$\text{and } (\Delta\sigma(t) = \sigma sl(t) - \sigma sl(o)$$

Where, θ(0) is the equilibrium contact angle in the absence of reaction, ΔG(t) is the change of Gibbs energy per unit area released by the dissolution reaction at the interface and Δσ(t) takes into account the change in σsl brought about by the reaction.

Process temperature also affects spreading. Higher temperature decreases liquid viscosity and liquid surface tension, which assists the good spreading. Wetting rate increases with rise in temperature following the Equation. (3) (Novakovic and Ricci, 2000).

$$\sigma = \sigma_0 - d\sigma/dT\ (T - Tm) \qquad (3)$$

Where, σ_0 and σ are initial and processing temperature surface tension, respectively. $d\sigma/dT$ is temperature coefficient. T and Tm are processing and liquidus temperature of metal, respectively.

Wetting kinetics for different Mg and ceramic oxides (general form M_xO_y) system can be observed by measuring contact angle at regular time interval until it reaches to equilibrium with image captured by high speed camera. Ceramic substrates can be prepared with standard shell making process by changing primary face coat slurry mixture. Secondary coating and stuccoing can be done as per standard shell making practice.

From the wettability measurements, system that found with higher equilibrium CA can be considered as most suitable to suppress the interface reactions and that oxide M_xO_y (of liquid phobic nature) will be suggested as primary face coat slurry material for magnesium alloy investment casting.

Assessment of Matrix-Reinforcement Reactivity for Magnesium Matrix Composites

Magnesium composite is the most upcoming research focus due to its additional weight saving characteristics. The reactivity between the matrix and the reinforcement have a significant effect on the interfacial strength and hence the deformation and fracture of the composite (Lloyd, 2013). Thermodynamic stability of different reinforcements with magnesium matrix needed to be enhanced. Wettability is the main parameter concerned with bonding strength of matrix-reinforcement that demands higher wettability. Higher wetting for reactive systems is responsible for higher reactions. Therefore optimum condition with higher wettability and lesser reactions is required for magnesium composite development. Modification in particle reinforcement like metallic coating of particles may help to get better results.

CONCLUSIONS

Research approaches proposed in present paper are mainly for development of economic and higher quality magnesium alloy casting components with main focus on reducing interfacial reactivity in investment casting and composite development. These approaches will be helpful to investment casters for mould development to produce successful magnesium investment castings as well as to enhance quality and properties of magnesium castings.

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