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PRODUCTION, PROPERTIES AND PROSPECTS OF APPLICATION OF MODERN MAGNESIUM ALLOYS

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ABSTRACT

A literature review of the current state of magnesium alloy production technology is given, alloy properties and the influence of alloying elements on them are analyzed, microstructure and the main phases forming in the cast and wrought magnesium alloys are determined, a classification of modern magnesium alloys of foreign production is presented, further paths of their development are outlined, and problems in magnesium alloy application are defined. Magnesium alloying by other elements (Al, Mn, Zn, Si, Re, etc.) or strong nanoparticles allows significant improvement of its existing properties so as to expand its scope of application. Current problems in magnesium alloy application (flammability, combustibility, surface durability, bi-solubility, corrosion resistance, weldability) are solved by alloying, controlling the chemical and structural-phase composition, application of the respective modes of thermomechanical treatment and surface hardening.

KEYWORDS: electrolysis, metallothermic reduction, Bunsen and Pidgeon processes, cast and wrought magnesium alloys, corrosion, biocompatibility, biodegradation

INTRODUCTION

It is impossible to imagine the progress of modern industry without development and application of new structural materials and, primarily, magnesium alloys. Magnesium and its alloys are structural materials, having a unique complex of mechanical, physical and service properties, which allows their application in advanced areas of modern manufacturing — aircraft and space industries, power engineering, defense industry, medicine and electronics [1–3].

Magnesium as an alkaline-earth metal, has a shiny or silvery-white appearance, and high reactivity, and it is never found in a free state [4, 5]. Easy availability of magnesium ores (brucite — 69.1 % MgO, forsterite — 57.3 % MgO, magnesite — 47.8 % MgO, etc.) and unique technologies of producing and restoration ensured a wide application of this metal.

Magnesium alloys are used to produce cases of instruments, pumps, torches and cabin doors. Most of the helicopter fuselages, manufactured by Sikorsky Aircraft (Lockheed Martin Company, USA), are almost completely made from magnesium alloys. In rocket engineering they are used to produce the bodies of rockets, fairing, stabilizers, and fuel tanks, as magnesium alloys do not have enough time to overheat in short flight, due to a high heat capacity (2.5 times higher than that of steel), despite the low melting temperature (650–680 °C). Magnesium alloys form the basis of such structures that allows a significant reduction of the rocket weight by 20–30 % and increase flight range, respectively.

An important industry, where magnesium alloys are used, is nuclear engineering. Owing to their ability to absorb thermal neutrons, no interaction with uranium and good heat conductivity, magnesium alloys are used to manufacture shells of heat generating elements in nuclear reactors.

Magnesium alloys are widely used in production of household and sporting goods, and office equipment [6].

Magnesium and its alloys play an important role in organic chemistry and pharmaceuticals [7]. Deformable magnesium alloys are used with success in structural elements of flying vehicles (aircraft, rockets, shells, etc.) that ensures reduction of product weight and improves their flight characteristics.

Owing to its excellent biological properties, particularly its biodegradability *in vivo*, magnesium is a promising biomaterial [8] for treatment and prosthetics and at surgical intervention.

The objective of this work is to analyze and generalize the metallurgical features of magnesium alloy production, establish the influence of the main alloying elements and microstructural components on their structural properties and determine the main problems and further application prospects.

METALLURGY

OF MAGNESIUM ALLOY PRODUCTION

There are two main methods of magnesium production: electrolysis of molten anhydrous magnesium chloride and metallothermic restoration of magnesium oxide by ferrosilicium [9]. Typical processes using electrolytic and thermal methods are illustrated in Figure 1.

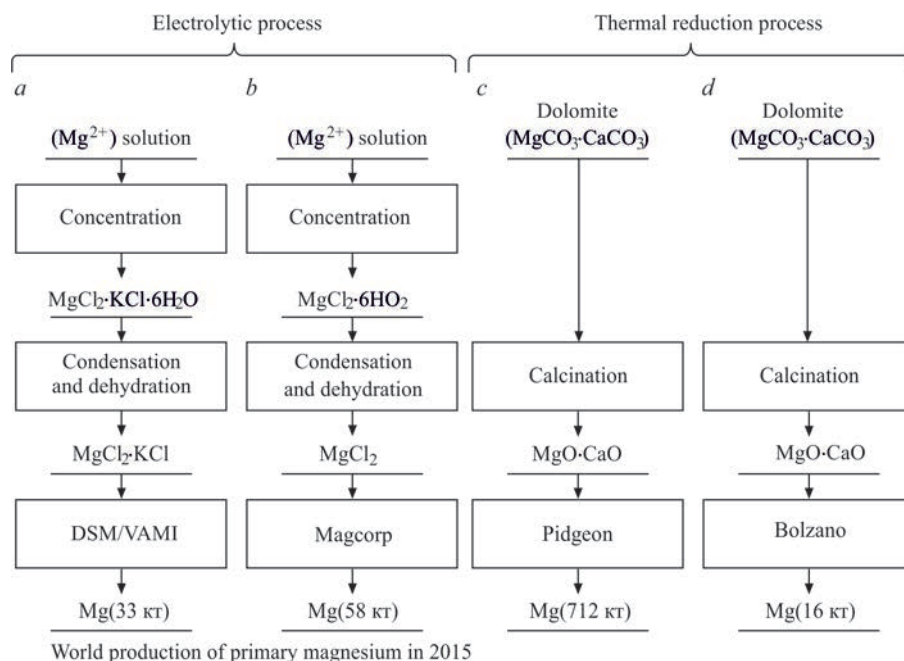
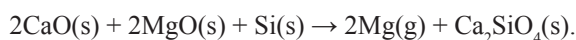


Figure 1. Block-diagram of the processes of primary metallic magnesium production: *a* — DSM/VAMI; *b* — Magcorp; *c* — Pidgeon

In the process of magnesium chloride electrolysis, first the hydrometallurgical preparation of the initial raw material (anhydrous magnesium chloride) is performed with its further feeding directly through the electrolyzers.

In his time, R. Bunsen improved the Faraday process of metallic manganese production to achieve continuous separation of chlorine and magnesium. Prevention of chlorine and magnesium recombination was critically important for success, as anhydrous magnesium chloride is hygroscopic, which may lead to formation of undesirable oxides and oxychlorides during direct dehydration [9]. Production of anhydrous magnesium chloride with a minimal quantity of oxychlorides remains to be a technology problem nowadays [10]. Scheme of Bunsen electrolytic process is shown in Figure 1 [11].

The second method of metallic magnesium production is thermal reduction. Unlike electrolysis, intensive heating during thermal reduction eliminates the need for complex preparation of the initial material [12]. The thermal process proceeds at higher temperatures (above 1200 °C) by metallothermic reduction reaction, in which silicon and/or aluminium remove magnesium in the vapour form from the oxide. Magnesium oxide is usually supplied in the form of calcined dolomite ($\text{MgO} \cdot \text{CaO}$), sometimes enriched in calcined magnesite (MgO). The main reaction of producing magnesite is as follows:



This reaction is highly endothermic and it runs in the periodic mode. Production processes run under vacuum at a lower temperature, so as to limit the

damage to the structure materials and stop unwanted side reactions in gas system.

There exist several variants of the thermal process. The three main thermal processes are Pidgeon process, using a retort with external heating; Magnetherm process — electric resistance heating using an electrode; Bolzano process, where the layer of briquettes consists of ferrosilicium and lime, and the fine fractions react in the furnace using internal electric heating.

In the Pidgeon process dolomite and ferrosilicium are formed into briquettes using a retort with external heating to produce magnesium vapours. Magnetherm process calcinates a mixture of dolomite, ferrosilicium and alumina, in order to produce the same byproduct. Magnesium vapours, produced by both the methods, are cooled and condensed separately before extraction. Nowadays, Pidgeon process is the most widely used one, due to its ability to produce high purity magnesium and a large quantity of its raw material — dolomite [13].

Application of vacuum is a common feature for the above thermal processes and metallic magnesium is condensed from the vapour phase. Magnetherm process is the only one, where the waste products are removed from the furnace as slag.

The main advantage of thermal processes over the electrolytic ones consists in that the main source of magnesium is dolomite, which requires simple calcination, unlike complex cleaning, which is required by the electrolytic method for production of anhydrous magnesium chloride.

On the other hand, the main disadvantages of the thermal processes include a relatively high cost of ferrosilicium reducing agents, and in particular, alu-

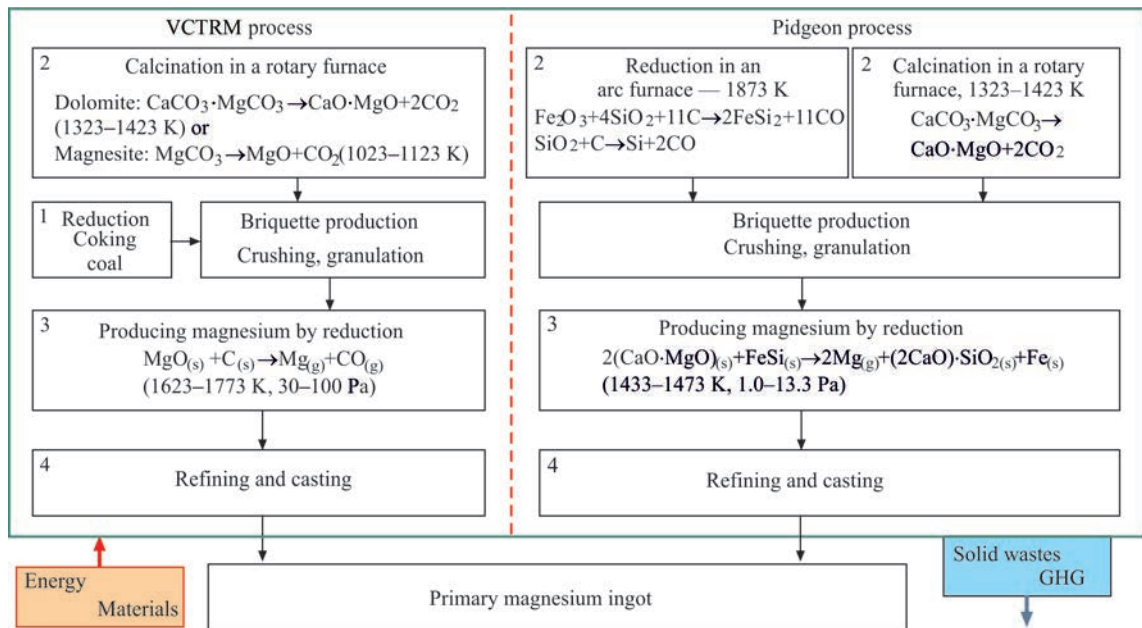


Figure 2. Schematic representation of working stages of Pidgeon and VCTRIM processes of magnesium production

minium, and periodic operation, required because of vacuum, causing lower productivity and leading to air ingress, resulting in magnesium loss.

CARBOTHERMAL REDUCTION AS A NEW PRODUCTION TECHNOLOGY

The modern Pidgeon technological process is known by its complex control and high operational costs. Moreover, it consumes a tremendous amount of energy. It necessitated a search for more efficient and sustainable production technologies. An alternative production method is vacuum carbothermal reduction of magnesium (VCTRIM) process, when magnesium reacts with carbon with formation of magnesium and carbon monoxide vapour [14] (Figure 2).

Despite the fact that this production process has passed large-scale production trials in the middle of

the XX century, it attracted more attention only in the last decades, as carbon is a less expensive reducing agent than ferrosilicium is. It is readily available and in case of success the total production cost will be much lower than with Pidgeon method [11]. Therefore, condensation of magnesium vapors and their separation from carbon monoxide remains a serious problem. Moreover, the researchers' interest to solar carbothermal reduction is growing (Figure 3) [15]. Investigations showed that this process uses less energy and releases less hydrocarbon gas, compared to Pidgeon method (5.31 against 15.9 kg CO₂).

SECONDARY PRODUCTION OF MAGNESIUM

With increase of magnesium application all over the world, greater volumes of magnesium wastes are to be anticipated. Magnesium is readily recycled and its

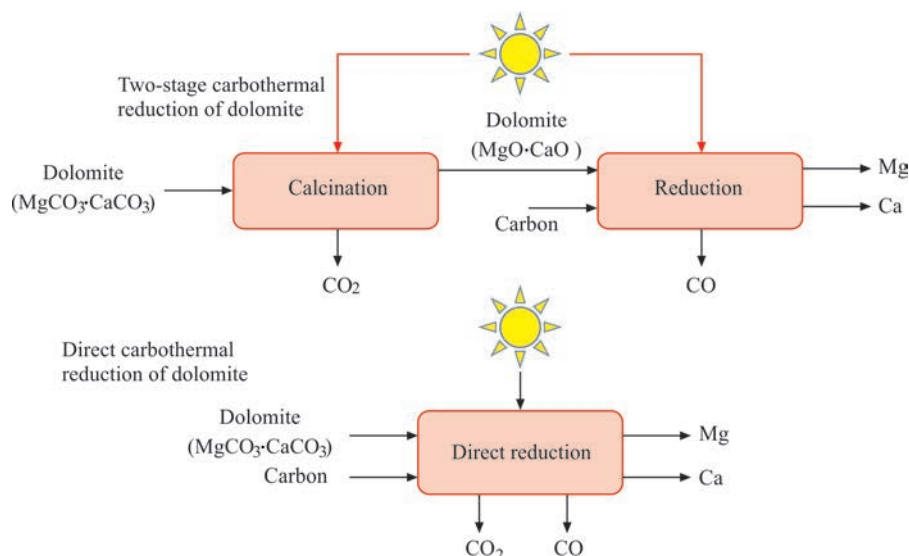


Figure 3. Block-diagram of the processes of solar carbothermal reduction of magnesium

recycling can become an additional supply source for the main production. Nowadays there exist both the flux and fluxless technologies of magnesium recycling. Recycled magnesium preserves the same chemical, physical and mechanical characteristics, as the initial material had. Moreover, magnesium recycling consumes only 5 % of the energy, required for its production. Use of secondary magnesium also reduces carbon emissions and saves material in a renewable economy. Magnesium alloys, however, can be easily recycled only in the case, if their composition is stable and does not contain any impurities [16]. It explains why most of the recycled magnesium is obtained from the wastes of die-cast magnesium alloys, when almost 30 % of magnesium is removed during the production process. To enable real progress of magnesium recycling, it is necessary to develop a recycling process, which would be commercially viable and environmentally friendly. At present, magnesium recycling methods include distillation, salt-free or salt-based remelting, solid-state recycling and direct conversion.

3D PRINTING

AS A NEW PRODUCTION PROCESS

Additive manufacturing or 3D printing is a new method of producing magnesium alloys. In general, the interest to 3D printing of metals grows due to its ability to adjust and optimize the composition of each alloy, in keeping with its application, and obtain the best possible result. Printing alloys in a more complex configuration to suit the anatomic geometry of patient tissues, so as to promote an accelerated growth of the cells, proliferation and regeneration of bones, can be an example [17].

Another example can be printing of complex-shaped parts from magnesium alloys. It could not be achieved earlier using traditional production, which restricted its development. More over, unlike the traditional production methods, part manufacturing using additive technologies can also greatly reduce the technical complexity and cost of individual production, while increasing its effectiveness.

Producing magnesium alloys by additive technologies is promising. However, studies on how magnesium alloys can be printed in 3D printer are rather limited, as it is known to be related to several problems [18]. First, magnesium is a chemically reactive and combustible material. It has a low evaporation temperature that complicates mixing and printing of magnesium alloys of a desirable composition, density, strength, biocompatibility and corrosive behaviour. Preparation of the initial material is dangerous, as magnesium powders have a high explosive tendency. At present, two typical methods of additive manufacturing to produce parts

from magnesium alloys are wire arc additive manufacture (WAAM) and selective laser melting (SLM), known as powder layer melting [18].

ALLOYING, STRUCTURE

AND PROPERTIES OF MAGNESIUM ALLOYS

Magnesium is the lightest engineering metal. Pure magnesium has a very low density (1.74 g/cm^3) and it demonstrates a higher strength-to-weight ratio and better ductility and castability than aluminium or steel. Compared to other metals and polymer materials, magnesium is not toxic and it has better values of heat and electric conductivity, vibration and shock absorption, cushioning capacity, as well as good workability, and it can be formed by any established method. Its disadvantage, however, is its corrosion susceptibility. It quickly corrodes under two conditions: when the alloy consists of specific metal impurities, or under the impact of aggressive kinds of electrolyte. Technologies of coating deposition (electrochemical, conversion, anodizing) were developed and used to overcome this feature of magnesium alloys [19]. Pure magnesium (99.8 %) is ideally suitable for metallurgical and chemical purposes, but it has to be alloyed for application in engineering structures, so as to enhance its properties without damage for the key characteristics [6].

The best production method is melting and casting magnesium alloys in an inert vacuum atmosphere, as magnesium solidifies better than other metals do. More over, a chemically inactive environment can prevent any contamination by reactive gases. Solid solution and dispersion strengthening by another phase is as a rule used to strengthen the magnesium alloys [20]. The alloy components and their chemical composition can influence improvement of magnesium alloy physical properties.

ALLOYING ELEMENTS

Aluminium and zinc are the most common and prevailing alloying elements. Both aluminium and zinc are sufficiently cost-effective and readily soluble in magnesium. Aluminium can be used to improve the strength and hardness and to widen the alloy melting range, while at the same time lowering its corrosion susceptibility. The respective range of strength and ductility can be achieved by changing the aluminium content in the alloy [21]. When applied together with zinc, it can further increase the room temperature strength of the alloy.

Zinc addition alone can increase the magnesium alloy castability and improve their dimensional stability. When zinc together with magnesium alloys is mixed with such impurities as nickel and iron, it can

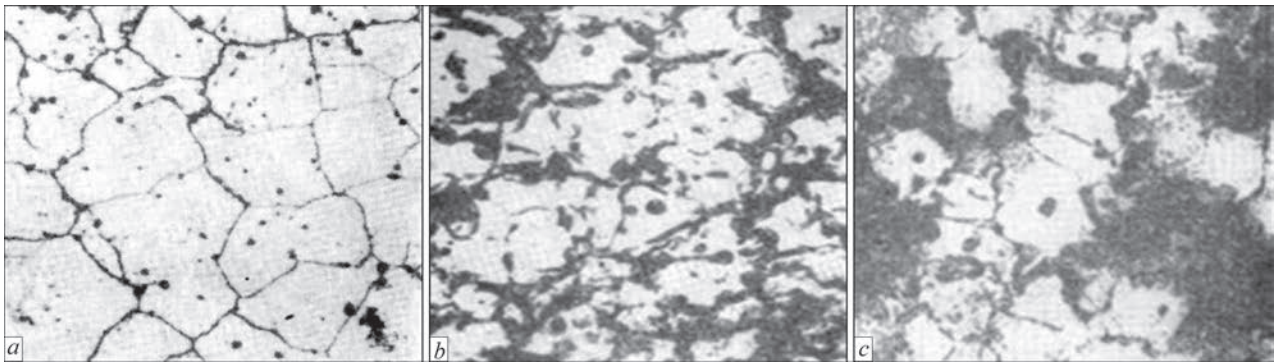


Figure 4. Microstructure ($\times 100$) of cast magnesium alloy ML5 (9 % Al; 0.5 % Mn; 0.8 % Zn) after different heat treatment modes: *a* — in the cast state; *b* — after quenching; *c* — after quenching + aging

increase the alloy corrosion resistance. Zinc does not form any independent phases with the alloying component — manganese. An γ -phase ($Mg_{17}Al_{12}$) forms in MA2-1 alloy. This phase precipitates at the alloy recrystallization as a result of heat treatment or welding, as well as during decomposition of the oversaturated solid solution, becoming part of a pseudoeutectic. The pseudoeutectic is mostly located along the grain boundaries. Primary $Mg_{17}Al_{12}$ phase is a strengthening phase. It is preserved in the alloy after deformation and welding. Some authors established that $Mg_{17}Al_{12}$ γ -phase has the ability of enrichment in zinc and additionally in aluminium, forming $Mg_{17}(Al, Zn)_{12}$ intermetallic phase, which is represented by coarse precipitates.

Excessively high concentrations of aluminium lead to ductility lowering and impair the alloy deformability in the hot and cold states. Zinc improves not only strength, but also relative elongation of the alloys with aluminium. However, at more than 1.5 % Zn content, pressure workability of alloys of Mg–Al–Zn system deteriorates. Manganese is added to these alloys predominantly to improve the corrosion resistance.

Investigations conducted over the recent years showed that magnesium alloy doping by rare-earth metals (REM) allows an essential improvement of their strength properties.

Magnesium alloys doped by REM of cerium (La, Ce, Pr, Nd, Pm, Sm, Eu) and yttrium (Sc, Y, Gd, Tb, Dy, Ho, Er, Tm, Yb) groups have their advantages and disadvantages, if compared by their properties. In the alloys with metals of the cerium group, lower strength values are achieved, but shorter aging time is required to obtain maximum strengthening. In alloys with metals of yttrium group a higher level of strengthening is achieved, but after a long-time soaking at aging and a higher content of expensive rare-earth metals. In a number of cases, simultaneous application of REM of both the groups is rational for doping the magnesium alloys.

Magnesium alloy microstructure in the initial state consists predominantly of equiaxed grains of 100 to

1000 μm size in the inner regions and on the boundaries, where dispersed and dispersive strengthening phases are located. These phases can have an important role in the magnesium alloy properties after conducting the respective heat treatment (Figure 4).

The dimensions and distribution of the phases forming during casting or during the subsequent stages of heat or thermomechanical treatment, depend on the respective heat treatment parameters. Dispersed particles counteract grain growth according to Zener–Straw model, in which the grain size grows with reduction of the size of particles and increase of their volume fraction. The effect of grain boundary fixation by Zr dispersoids and without them in the magnesium alloy is given in Figure 5. In magnesium alloys without aluminium application, zirconium is often used as a dispersing element, while in alloys of Mg–Al system with aluminium, it is rational to use magnesium with formation of AlMn intermetallics. Zr additives are also effective as grain refiners during casting that enables production of fine-grained magnesium alloys.

A considerable part of magnesium alloy products is produced by the extrusion method. As a result, such magnesium alloys differ by their microstructure that is related to the processes of grain recrystallization on the second phase clusters, extruded at higher temperatures (Figure 6).

The structural-phase composition of magnesium alloys is mainly determined by their alloying system. There exist a number of solidification phases forming in the equilibrium state with magnesium solid solution, namely $Mg_{17}Al_{12}$ (Mg–Al system), Mg_2Ca (Mg–Ca system), Mg_2Sn (Mg–Sn system), Mg_4Ag (Mg–Ag system), Al_2Re and Al_2Ca (Mg–Al–Ca–Re system), $MgCu_2$ (Mg–Cu–Zn system), $Mg_6Ca_2Zn_3$ (Mg–Ca–Zn system), X_MgReZn (Mg–Gd–Zn and Mg–Y–Zn system), PHI_AlMgZn (Mg–Al–Zn system). The following metastable phases can also form in the magnesium alloys: $MgZn_2$ (Mg–Zn system) and Mg_7Re (Mg–Re system).

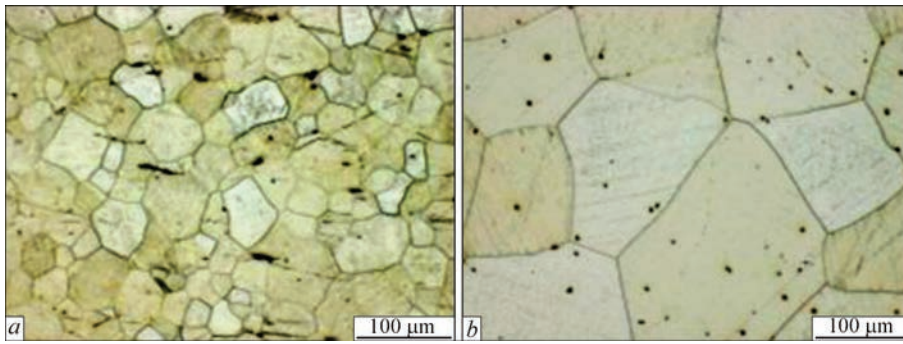


Figure 5. Microstructure of magnesium alloy with REM, after forging and heat treatment: *a* — with zirconium (~ 0.4 wt.%); *b* — without zirconium

A feature of magnesium alloys is a lower rate of diffusion of the majority of the components in the magnesium solid solution. Low diffusion rates promote development of dendrite liquation, require a long soaking time at heating, facilitate solid solution fixation at hardening, but complicate decomposition of oversaturated solid solutions at aging.

Magnesium alloys have a high ductility in the hot state, and deform well when heated. For wrought alloys diffusion annealing is usually combined with heating for pressure treatment. Magnesium alloys are readily cut, ground and polished.

The disadvantages of magnesium alloys are poor casting properties, proneness to gas saturation, oxidation and ignition during casting. Special fluxes are used to prevent defect formation at casting and welding, and small calcium additives (up to 0.2 %) are applied to reduce porosity, and beryllium additives (0.02–0.05 %) — to reduce oxidizability.

Alongside the traditional alloying method, considerable progress has been made over the recent years in application of nanocomposites based on magnesium alloys, which additionally strengthen the alloys [22]. The characteristics of these alloys are affected by both the alloying components and the nanoparticles, reinforcing the alloy [23]. Increased researchers' interest in this area is associated with the fact that strengthening by nanoparticles can improve the magnesium strength and ductility without any negative consequences.

American Society for Testing Materials (ASTM) proposed a system of designations for identification

of magnesium alloys and determination of their composition (Figure 7) [24].

All the magnesium alloys are divided into two large groups, namely cast and wrought. Among the cast alloys, a group of alloys produced by die casting (AZ91, AS41, AE42) stands out. Wrought magnesium alloys are divided into those deformed by forging, rolling or extrusion.

Performed analysis of published sources showed that development of magnesium alloys gradually moved along the path of application of more complex compositions, in order to obtain specific properties, namely strength (superlight, high-strength), toughness (thermally or thermomechanically hardened), creep, thermal expansion, wear, etc. (Figure 8) [25].

Magnesium alloys with aluminium are AZ31 and AZ91. AZ31 alloy is widely used in aircraft construction due to its low mass density and good mechanical properties. At the same time, AZ91 alloy remains to be one of the most popular casting alloys owing to its high strength, excellent corrosion resistance and good castability. Mechanical properties of AZ31 and AZ91D magnesium alloys are given in Table 1.

Performed analysis of publications showed that the main problems are to be anticipated in welding cast magnesium alloys (AZ63F, ZE41F, ZK51A, ZK61A), as a result of development of dendritic liquation, chemical and structural heterogeneity in them, formation of pores and discontinuities and strengthening phase segregation (Table 2), whereas forged magne-

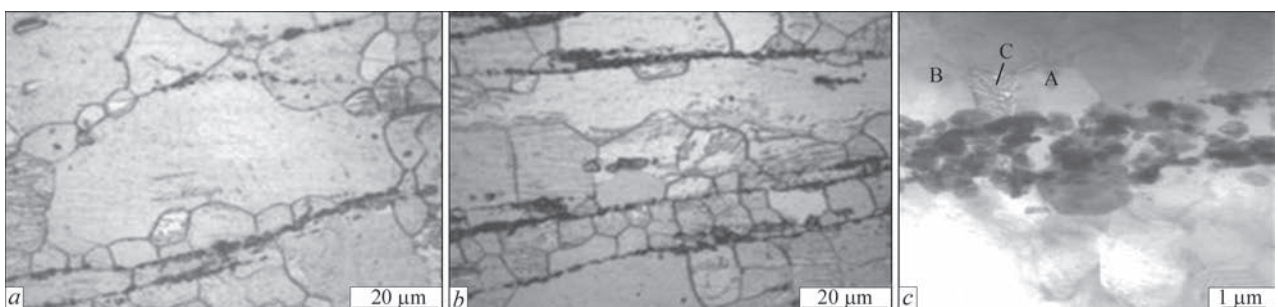


Figure 6. Microstructure of magnesium alloy AZ31 + 0.8 wt.% Sr, extruded at the temperatures of 250 (*a*), 350 °C (*b*) and recrystallization on particles of Al-Sr phases (*c*)

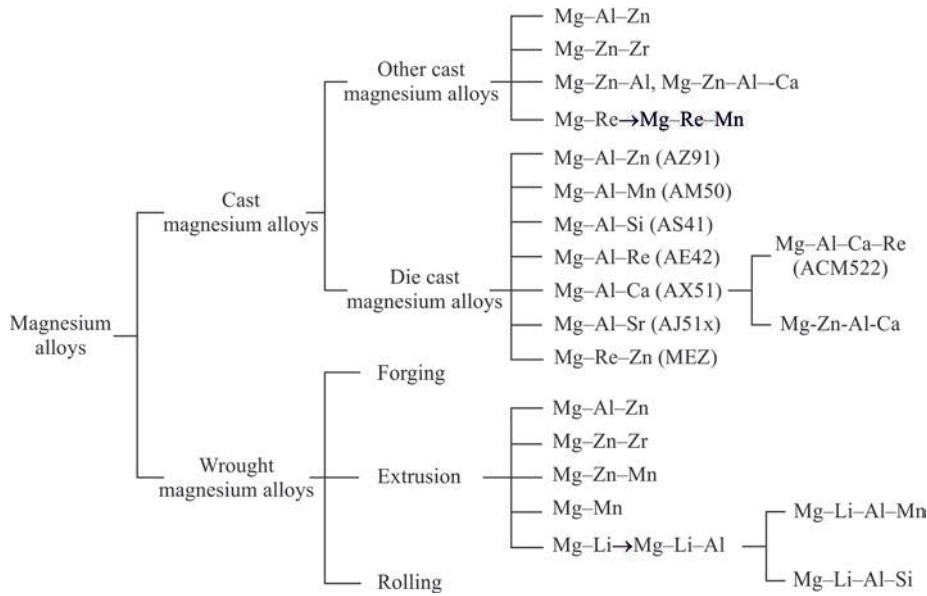


Figure 7. Classification of foreign magnesium alloys [24]

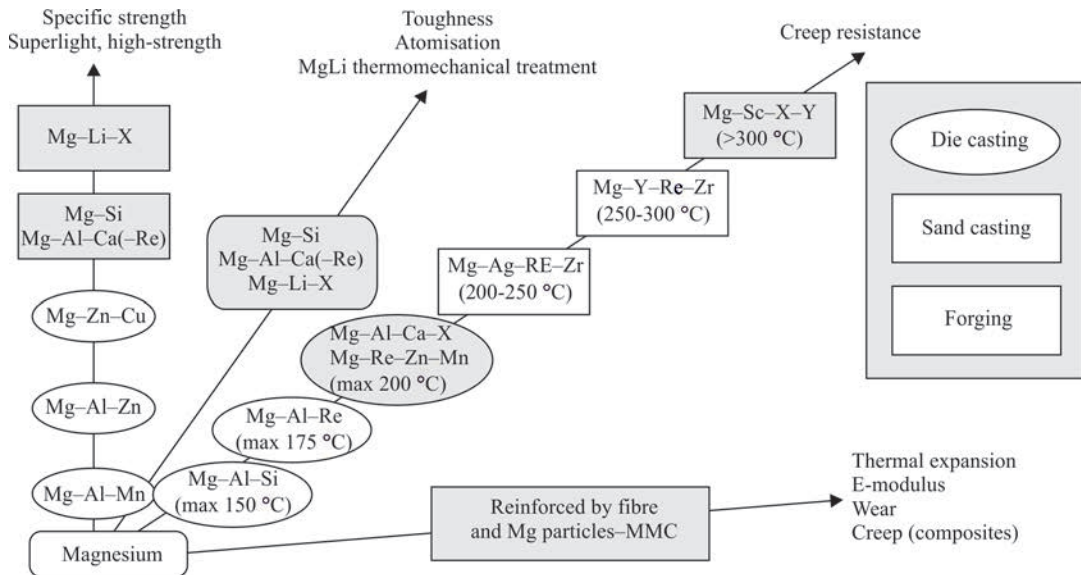


Figure 8. Directions of development of foreign magnesium alloys [25]

sium alloys (NK31A, ZE10A, ZK21A) have excellent weldability.

APPLICATION OF MAGNESIUM ALLOYS

Calculations show that application of lighter magnesium alloys instead of heavier ones (aluminium and titanium alloys or steels) can improve the aircraft fuel efficiency and reduce harmful emissions. It means considerable fuel saving and lower operating costs. Replacement of aluminium alloys by magnesium ones can en-

sure an almost 60 t weight reduction, that makes 28 % of the aircraft weight.

At the same time, limitations and critical aspects of magnesium application in modern aviation were revealed. This is predominantly related to its combustibility, surface durability and corrosion resistance.

Nowadays aluminium and its alloys still have significant advantages over magnesium and its alloys for aerospace structures. However, recent progress is applied studies of magnesium alloy flammability lead to discovery of less combustible materials that enabled

Table 1. Mechanical properties of magnesium alloys

| Material/Alloy | Density, g/cm ³ | Compressive strength, MPa | Tensile strength, MPa | Modulus of elasticity, GPa |
|----------------|----------------------------|---------------------------|-----------------------|----------------------------|
| Magnesium | 1.74 | 20–115 | 90–190 | 45 |
| AZ31 (wrought) | 1.78 | 83–97 | 241–260 | →→ |
| AZ91D (cast) | 1.81 | 160 | 230 | →→ |

Table 2. Relative weldability of magnesium alloys

| Cast into a chill mold | | | | | | | | | |
|------------------------|-------|-------|--------|-------|-------|-------|-------|-------|-------|
| AM100 | AZ63A | AZ81A | AZ91AC | AZ92A | EK41A | EZ33A | HK31A | HZ32A | OE22A |
| B | C | B | B | B | B | A | B | C | B |

Table 2 (cont.)

| Cast into a chill mold | | | | Deformed by forging | | | | |
|------------------------|-------|-------|-------|---------------------|-------|-------|-------|-------|
| ZE41A | ZH62A | ZK51A | ZK61A | HK31A | HM21A | HM31A | ZE10A | ZK21A |
| C | C | D | D | A | A | A | A | B |

Note. A — very good; B — good; C — satisfactory; D — bad.

their wider application in passenger aircraft and cars. Increase of mechanical strength of such magnesium alloys as Elektron 21 or Elektron 675, demonstrating two times higher strength than aluminium does, will also promote their further application [26].

Although the first attempts of application of magnesium and its alloys in cars date back to the beginning of the XX century, the overall fraction of magnesium materials in the automotive industry remains to be negligible, compared to steel and aluminium. Figure 9 shows the retrospective of application of magnesium and its alloys in cars of different manufacturers during 1930–2000.

The main type of cast magnesium alloys in the automotive industry is a series of alloys, based on Mg–Al system (AZ, AM), rare-earth elements (WE43, E21) and ZK grade alloys.

Moreover, the possibility of using high-strength magnesium alloys with nanocomposites is considered for higher temperature applications, as they have excellent thermal and dimensional stability [27]. However, except for cast magnesium alloys, other alloy types, for instance, sheet wrought or extruded magnesium alloys are almost not used in modern automotive industry.

Magnesium alloys found wide application in the health care and biomedical industry [24]. Magnesium is a material naturally biocompatible with the human body. It supports physiological functions in the human body, including structural stabilization of proteins, nucleic acids, cell membranes, and it also stimulates special structural or catalytic activities of proteins or enzymes [1]. Moreover, magnesium is not toxic, does not irritate the skin and promotes tissue healing.

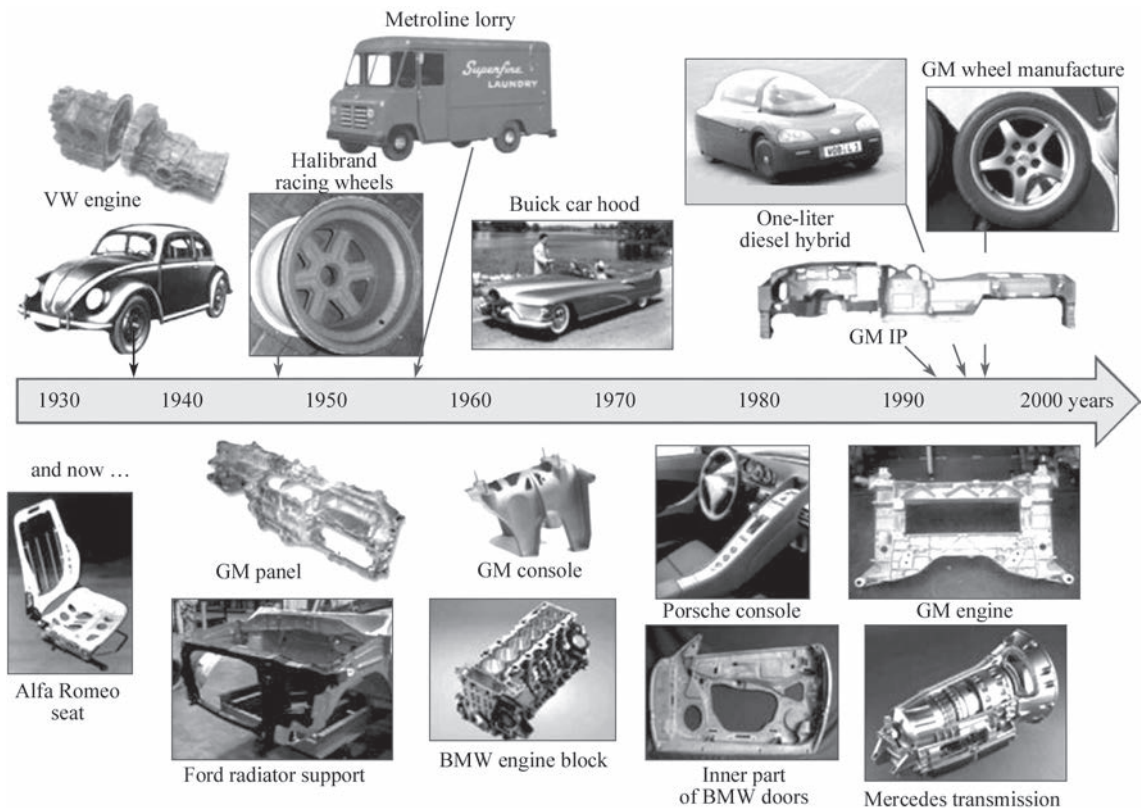


Figure 9. Retrospective of magnesium alloy application in automotive industry

Magnesium also is a bioabsorbable and bioactive material. When used as implants, magnesium alloys have a lower modulus of elasticity, than do other analogs, which is their advantage as a biomaterial. As the modulus of elasticity of magnesium is closer to the rigidity of natural bone at application, it is preferred in most cases.

Often the body requires just a temporary presence of the implant to support tissue regeneration and restoration of the impaired physical functions. Magnesium ability to biodegrade is both its greatest advantage and weakness. Magnesium quickly decomposes in a physiological environment. Such accelerated degradation can endanger the mechanical integrity and strength of the respective implants. It may also lead to undesirable reactions and can cause biotoxicity.

The disadvantages of magnesium alloy application include their wear corrosion resistance. Depending on application conditions, different kinds of corrosion can develop in the magnesium alloys, namely: galvanic, pitting, filiform, intergranular (IGC), exfoliation (EFC), crevice, stress corrosion cracking (SCC), corrosion fatigue (CF) and erosion corrosion wear [5].

Analysis of the used publications showed the key strategies for overcoming the corrosion fracture of magnesium alloys, which consist of cleaning, alloying and surface modification. These strategies use the principle of slowing down the corrosion process due to reduction of hydrogen accumulation rate and local alkalization. Despite the considerable achievements in controlling magnesium corrosion, this problem still remains urgent.

CONCLUSIONS

1. Magnesium accessibility and a unique combination of properties (low weight, specific high strength-to-weight ratio, excellent workability and castability) made it very attractive for application. Magnesium alloying by other elements (Al, Mn, Zn, Si, Re) or strong nanoparticles allowed improvement of its existing properties, so as to expand the range of its applications. These factors allowed magnesium to find its place in the aerospace, automotive, defense and nuclear industries and in medicine.

2. Existing problems in the use of magnesium alloys (flammability, combustibility, fatigue life, biosolubility, corrosion resistance, weldability) are solved by its alloying, controlling its chemical and structural-phase composition, application of the respective modes of thermomechanical treatment and surface hardening.

3. Modern methods of producing primary magnesium and its alloys are rather energy-intensive and inefficient in production, which results in expansion

of investigations in the field of additive production of magnesium alloys and use of “green” power technologies. An ecologically safe process of recycling of magnesium and its alloys also is one of the variants of manufacturing the required metal products.

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CONFLICT OF INTEREST

The Authors declare no conflict of interest

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