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# DEVELOPMENT OF NEW CASTING MAGNESIUM-BASED ALLOYS WITH INCREASED MECHANICAL PROPERTIES

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***Обґрунтовано вибір легувальних елементів для розробки нових жароміцних магнієвих сплавів з підвищеним рівнем механічних властивостей. Встановлено залежність морфології і топології інтерметалідних фаз від хімічного складу магнієвих сплавів і їх вплив на структуру і властивості металу. Проведена кількісна та якісна оцінка структурних складових сплаву МЛ5, легованого Nd, Ge, Si, Y, Sc, Zr, Ti і Hf, досліджено їх вплив на структуру, механічні властивості і жароміцність магнієвого лиття***

***Ключові слова: магнієвий сплав, легувальні елементи, структура сплаву, інтерметаліди, механічні властивості, жароміцність***

***Обоснован выбор легирующих элементов для разработки новых жаропрочных магниевых сплавов с повышенным уровнем механичных свойств. Установлена зависимость морфологии и топологии интерметаллидных фаз от химического состава магниевых сплавов и их влияние на структуру и свойства металла. Проведена количественная и качественная оценка структурных составляющих сплава МЛ5, легированного Nd, Ge, Si, Y, Sc, Zr, Ti и Hf, исследовано их влияние на структуру, механические свойства и жаропрочность магниевого литья***

***Ключевые слова: магниевый сплав, легирующие элементы, структура сплава, интерметаллиды, механические свойства, жаропрочность***

## 1. Introduction

Scientific and technological advance leads to continuous machinery complication, which contributes to an increase of metal intensity in mechanisms and equipment. Therefore, weight reduction in engineering structures presents a challenge to the modern mechanical engineering [1]. One of the ways to solve this problem is application of magnesium alloy castings for the manufacture of units and sub-assemblies as a substitute for existing iron and still cast components. Utilization of castings from magnesium alloys makes it possible to substantially reduce the machinery weight, which results in a higher efficiency, payload and lower energy input. Ever increasing requirements to operating reliability and service life of the modern machinery lead to the necessity of improving mechanical properties and performance characteristics of magnesium alloy castings [2].

Therefore, development of the magnesium alloys with an increased level of properties represents a relevant objective.

## 2. Literature review and problem statement

At present, a number of researches on studying an influence of alloying elements on physical and mechanical properties and performance characteristics of the magnesium alloys are being carried out [3–5]. However, the number of the works dedicated to studying the nature of strengthening of such alloys resulting from their alloying and inoculation as well as its relationship to the structural condition of the metal is limited.

Basic methods of attaining a high strength in cast alloys while retaining a sufficient ductility level are as follows:

1. Formation of complex alloyed solid solutions.
2. Formation of an optimum structure through heat treatment.
3. Strengthening of metals and alloys with dispersed particles.

In order to attain high properties of the cast magnesium alloys, a combination of all three methods is required.

Metals with fully built-up d-electron shell, e. g. zinc, lead, tin etc, are readily dissolved in magnesium, both in liquid and solid state. These metals form with magnesium a range of compounds, which for the IV and V groups of the Mendeleev periodic system table are subject to the valence rule, whereas magnesium forms with the II and III group elements the phases, which are not subject to the valence rule. The elements of the III group (Sc, Y, RE elements) with incomplete d-shells form intermediate phases and readily dissolve in a liquid state; on the contrary, the solubility of the phases in solid-state magnesium is weak [6, 7].

The complex alloyed SS are formed due to solubility of elements in magnesium, when their atomic diameters are to a certain extent similar; according to Hume-Rothery's rule [8], the atomic diameters shall differ by a maximum of 15 percent. If this rule is not met, atom binding energies of both solvent and alloying elements decrease and due to distortion of the matrix lattice their solubility is reduced.

Another important prerequisite for the solubility of an element in the base metal [9–11] is a small electro negativity difference, which shall not exceed 0.2–0.4.

Among the variety of alloying elements only few of them, having a favorable factor of atomic diameter (≤ 15 %) and electro negativity (≤ 0.4) ratios, are capable of forming SS with magnesium, thus strengthening it (Table 1) [12].

Table 1

Atomic radii (Ar), element electro negativity (E/N) in relation to magnesium [12]

| Periodic system group | Element | Electron configuration | $A_{rEl}$, nm. | $(A_{rMg} - A_{rEl})\ A_{Mg}$, % | $E/N_{El}$ | $\lvert E/N_{Mg} - E/N_{El}\rvert$ |
|---|---|---|---|---|---|---|
| 2a | Mg | $3s^2$ | 0.160 | – | 0.56 | – |
| 3a | Sc | $3d^14s^2$ | 0.164 | 1.2 | 0.53 | 0.03 |
|  | Y | $4d^15s^2$ | 0.181 | −13.1 | 0.48 | 0.08 |
|  | Nd | $4f^46s^2$ | 0.182 | −13.8 | 0.95 | 0.39 |
| 4a | Ti | $3d^24s^2$ | 0.146 | 8.8 | 0.61 | 0.04 |
|  | Zr | $4d^25s^2$ | 0.160 | 0 | 0.57 | 0.01 |
|  | Hf | $4f^{14}5d^26s^2$ | 0.159 | 0.6 | 0.50 | 0.06 |
| 4b | Si | $3s^23p^2$ | 0.136 | 15.0 | 0.83 | 0.27 |
|  | Ge | $3d^{10}4s^24p^2$ | 0.137 | 14.4 | 0.84 | 0.28 |

In order to increase the high-temperature strength of an alloy by alloying, the melting temperature of alloying elements shall be higher than that of the alloy base [13]. Therefore, the possible candidates for the alloying of the magnesium alloys to improve both physical-mechanical properties and high-temperature strength are the following elements: Nd, Ge, Si,Y, Sc, Zr, Ti, Hf.

Genesis of magnesium – alloying elements equilibrium diagrams (from germanium to hafnium) (Fig. 1) shows that as the alloying element melting temperature increases, the equilibrium diagrams transform from eutectic to peritectic type. Besides, the increase of the element melting temperature is accompanied with a simultaneous increase in the melting temperature of transition phases, which results in a heterogeneous structure resistant to increased temperatures.

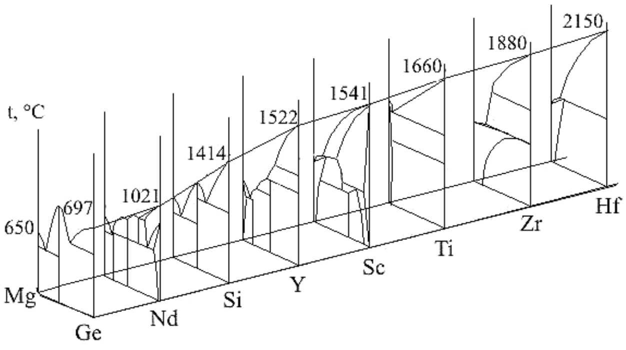


Fig. 1. Genesis of phase diagrams for the magnesium — favourable alloying elements with magnesium

In the known studies on the effect of temperature modes of the cast solution processing on the anisotropy of the properties and corrosion resistance [14–16], a critical analysis on the influence of various alloying elements on the physical, mechanical and performance properties of magnesium alloy castings is needed. The phase stability of casting after doping was investigated [17–19] but the nature of this influence, the relationship with the structural state of the metal was not clarified. Therefore, the study of the influence of alloying elements on the structure formation, mechanical and special properties of magnesium alloy castings, the study of nature and the relationship of these factors is a promising task of modern mechanical engineering.

## 3. Research aim and objectives

The aim of the present work is development of new and improvement of existing magnesium alloys with increased properties, based on the atomic and electron interaction of their elements, improvement of the casting quality by controlling their structural characteristics, which ensure reliable and durable operation of equipment.

To attain the aim, the following issues were addressed:

1. Definition of optimum alloying element content in ML5 magnesium alloy.
2. Study of chosen alloying element influence on the structure formation in the alloy as well as on the morphology and topology of the phases being formed.
3. Definition of relationships associated with an influence of quantity and a shape of the intermetallic phase on the mechanical properties and high-temperature strength of the magnesium cast products.

## 4. Heat production procedure and research methods

The magnesium alloy was melted in an induction crucible and holding furnaces with a portioned alloy sampling. Incremental master alloy additions consisting of the corresponding elements (0; 0.05; 0.1; 1.0 %) were added into the melt, which was heated up and poured into cast test specimen.

The specimens were then subjected to T6 heat treatment.

The UTS and elongation of the test specimen were determined on P5 tensile-testing machine at room temperature. The stress-rupture strength at a temperature of 150 °C and applied stress of 80 MPa was determined on AIMA 5-2 stress-rupture testing machine on the test specimen with the gauge diameter of 5 mm.

The chemistry of the magnesium alloy castings was checked with optical emission spectrometers «SPECTROMAXx» and «SPECTROMAXxF», photoelectric spectrometers MFS-8 and TFS-36, «SPECTRO XEPOS» X-ray fluorescent spectrometer.

The macro- and microstructure of the researched alloys was studied by the light microscopy («Neophot 32», «OLYMPUS X 70» microscopes) and using «Videotest-Struktura 5.0» soft-and-hardware complex on the basis of Axiovert 40MAT metallurgical microscope.

Qualitative and quantitative evaluation was made by a standard method of volume percentage calculation.

A micro X-ray spectrum analysis of the magnesium alloy structure constituents was performed using JSM-6360LA electron microscope.

## 5. Studying of alloying element influence on structure formation, mechanical properties and high-temperature strength of the magnesium alloy

The base magnesium alloy versions representing a number of different micro alloying modifications were characterized by a similar chemical composition (7.6 % A1; 0.28 % Mn; 0.35 % Zn; 0.02 % Fe; 0.005 % Cu; 0.04 % Si).

The macro fractographic study of the cast test specimen fractures of the pure magnesium showed a presence of a brittle coarse-crystalline structure (Fig. 2, *a*). The micro structure of the standard ML5 alloy was distinctly fine (Fig. 2, *b*) and an addition of the alloying elements into the alloy resulted in a matted fine-crystalline structure (Fig. 2, *c*).

The microstructure of ML5 alloy was a δ-solid solution with a presence of eutectic of $\delta+\gamma(Mg_{17}Al_{12})$ – type and intermetallic compound $\gamma(Mg_{17}Al_{12})$ (Fig. 3).

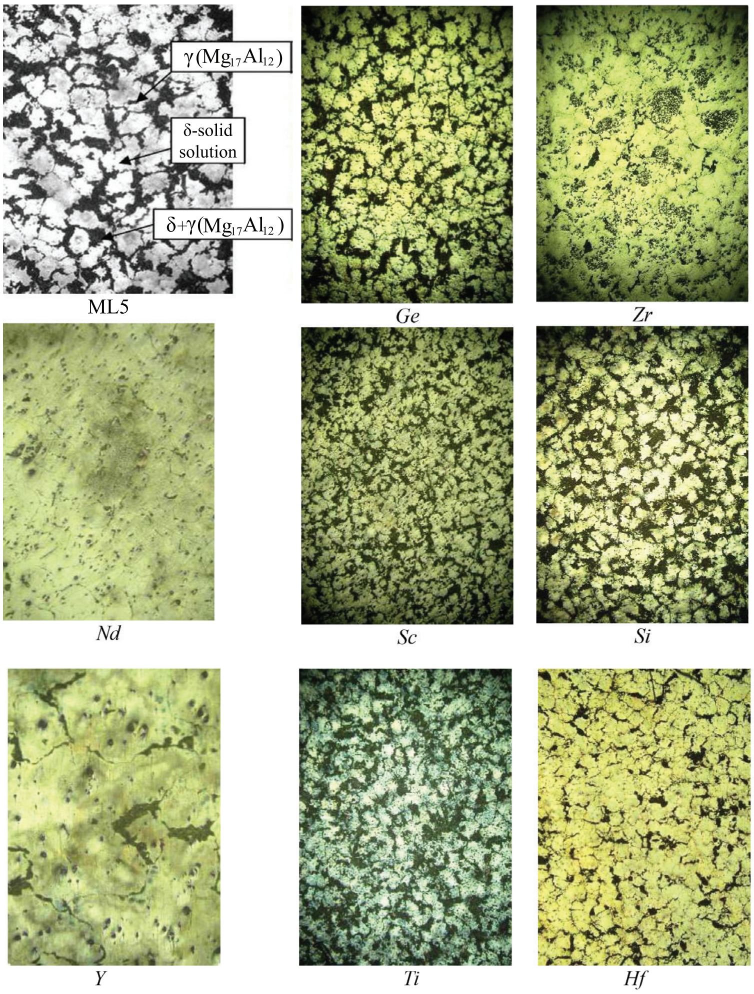


Fig. 3. Influence of the alloying elements (0.1 %, wt) on the ML5 microstructure, × 200

The addition of the alloying elements into the alloy favored the reduction of the 2nd dendrite arm spacing (Table 2), the structural constituent size and dispersing the eutectics.

Alloying of ML5 alloy with Si, Sc, Ti, Ge, Y, Zr, Nd and Hf has resulted in a reduction of the micro grain by 30...40 %, increase in a micro hardness of the structure constituents and contributed to attaining finer intermetallic phases. In each subgroup of the periodic system, the influence of the elements on the grain size refinement was more pronounced with increasing nuclear charge of the element atoms. In so doing, elements of 4a subgroup Ti, Zr and Hf caused a more intensive refinement.

*a* *b* *c*

Fig. 2. The micro structure of the test specimen fractures: *a* — 99.9 % Mg, *b* — ML5, *c* — ML5 + 0.1 % Zr, × 5

Table 2

The characteristics of the structure constituents of ML5 with different alloying elements (mean values)

| Element | Content, % wt. | 2nd dendrite arm spacing, μm. | Micro grain size, μm. | Micro hardness, HV, MPa |
|---|---|---|---|---|
| Mg (99.9 %) | – | 40 | 300 | 524 |
| Mg (99.9 %) + Al | 0.045 | 35 | 210 | 582.0 |
|  | 0.096 | 30 | 200 | 646.7 |
|  | 8.50 | 25 | 170 | 1226.5 |
| МЛ5 | 0.32 | 23 | 140 | 1257.6 |
| Sc | 0.05 | 18 | 120 | 1465.7 |
|  | 0.10 | 17 | 100 | 1547.1 |
|  | 1.0 | 16 | 90 | 1675.0 |
| Nd | 0.05 | 18 | 120 | 1290.0 |
|  | 0.10 | 17 | 100 | 1390.5 |
|  | 1.0 | 17 | 90 | 1407.6 |
| Zr | 0.05 | 17 | 105 | 1235.3 |
|  | 0.10 | 16 | 100 | 1265.6 |
|  | 1.0 | 16 | 70 | 1297.9 |
| Hf | 0.05 | 17 | ПО | 1256.6 |
|  | 0.10 | 16 | 100 | 1294.4 |
|  | 1.0 | 15 | 70 | 1321.1 |
| Si | 0.047 | 19 | 130 | 1276.5 |
|  | 0.12 | 17 | 120 | 1313.5 |
| Ge | 0.055 | 19 | 125 | 1233.4 |
|  | 0.095 | 18 | 100 | 1244.6 |
|  | 1.09 | 17 | 90 | 1287.5 |
| Ti | 0.05 | 18 | 120 | 1265.6 |
|  | 0.10 | 16 | 100 | 1270.7 |
|  | 1.0 | 16 | 100 | 1283.3 |

Lamellar (Fig. 4, *a*) and spherical (Fig. 4, *b*) intermetallic phases were present in the structure of the base alloy. Addition of the studied alloying elements into the base alloy resulted in an increase of the IC content and a change in their chemical composition (Table 3).

Besides, the addition of 0.05...0.1 % alloying elements into ML5 base alloy has led to a considerable increase in a number of spherical and not significant – lamellar ICs (Table 4).

Table 3

Chemical composition of the IC in ML5 alloyed with different elements

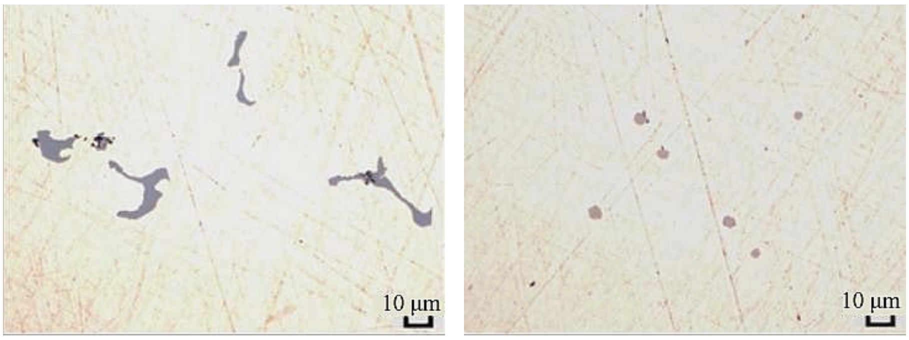
| Alloying modification | Element content, % wt. |  |  |  |  |
|---|---|---|---|---|---|
|  | Alloying element | Mn | Al | Si | Mg |
| Standard alloy | – | 1.80 | 14.85 | 1.36 | 81.99 |
| Sc | 32.65 | 6.49 | 26.87 | – | 33.99 |
| Y | 15.74 | 46.36 | 34.33 | 0.58 | 2.99 |
| Nd | 31.26 | 1.37 | 44.25 | 0.16 | 22.96 |
| Ti | 21.80 | – | 21.14 | 21.57 | 35.49 |
| Zr | 71.40 | 1.30 | 10.98 | 1.55 | 14.77 |
| Hf | 1.16 | 34.67 | 34.94 | 2.38 | 26.85 |
| Si | 52.36 | 0.02 | 1.22 | – | 46.40 |
| Ge | 19.84 | – | 2.68 | 16.83 | 60.65 |

Table 4

The ICs volume fraction (V) and size distribution in ML5 alloy containing different alloying elements

| Element | Content, % wt. | IC size group distribution $V \times 10^{-3}$, Mm |  |  |  |  |  |  |
|---|---|---|---|---|---|---|---|---|
|  |  | < 2 | 2...3.9 | 4...7.9 | 8...11.5 | 11.6...15 | 15.1...19 | Total |
| Standard |  | 6/0 | 18/54 | 36/30 | 30/12 | 30/12 | 18/0 | 138/108 |
| Y | 0.05 | 42/2 | 84/72 | 24/42 | 12/24 | 18/0 | 18/0 | 210/138 |
|  | 0.10 | 66/0 | 90/150 | 42/42 | 12/18 | 18/0 | 6/0 | 234/210 |
|  | 1.00 | 78/78 | 162/126 | 42/24 | 6/6 | 18/0 | 0/0 | 306/234 |
| Sc | 0.05 | 42/0 | 60/78 | 48/36 | 12/30 | 6/0 | 0/0 | 168/144 |
|  | 0.10 | 72/74 | 12/114 | 78/30 | 6/24 | 6/0 | 0/0 | 174/192 |
|  | 1.00 | 81/96 | 81/48 | 132/24 | 6/24 | 0/0 | 0/0 | 300/192 |
| Nd | 0.05 | 48/0 | 48/108 | 30/42 | 24/36 | 36/6 | 18/0 | 204/192 |
|  | 0.10 | 90/18 | 30/114 | 48/42 | 18/30 | 18/6 | 18/0 | 220/210 |
|  | 1.00 | 150/57 | 24/114 | 73/20 | 18/24 | 12/6 | 6/0 | 282/231 |
| Ti | 0.05 | 60/0 | 84/60 | 54/36 | 24/24 | 12/0 | 6/0 | 240/210 |
|  | 0.10 | 90/18 | 126/78 | 18/30 | 18/18 | 6/0 | 0/0 | 258/144 |
|  | 1.00 | 120/60 | 168/66 | 24/36 | 18/18 | 6/0 | 0/0 | 136/180 |
| Zr | 0.05 | 36/6 | 30/42 | 54/30 | 18/42 | 6/0 | 12/0 | 156/120 |
|  | 0.10 | 60/18 | 30/84 | 60/30 | 18/24 | 0/0 | 12/0 | 180/156 |
|  | 1.00 | 66/60 | 54/84 | 126/24 | 12/12 | 6/0 | 0/0 | 264/180 |
| Hf | 0.05 | 66/6 | 84/78 | 42/36 | 24/24 | 12/6 | 6/6 | 234/156 |
|  | 0.10 | 66/12 | 132/120 | 84/36 | 12/8 | 12/0 | 0/0 | 306/186 |
|  | 1.00 | 126/72 | 84/108 | 96/24 | 18/0 | 0/0 | 0/0 | 324/204 |

*Note: numerator – a volume fraction of the lamellar ICs, denominator – a volume fraction of the spherical ICs.*



*a* *b*

Fig. 4. Lamellar (*a*) and spherical (*b*) IC in ML5 alloy

At the alloying element concentration of about 1.0 % in the alloy, a quantity of spherical ICs within the grains increased slightly, whereas the increase in the quantity of the lamellar-type was considerable, which favored the grain refinement. The growth of the total amount of the ICs was accompanied with an increase of the alloy strength. A size group evaluation of the IC distribution has shown that the lamellar ICs of 4...15 μm size group dominated in the base alloy. Spherical ICs are generally represented by a size group of 2...11.5 μm. The studied alloying elements in the magnesium alloy led to a shift in the inclusion size towards smaller sized groups (up to 2–7.9 μm for the spherical and 2–11.5 μm for the lamellar). In so doing, the alloying element content increase in the alloy resulted in a higher number of IC sized less than 2 μm and lower number of IC sized more than 11.6 μm. It was established that increase in a volume percent (V) of the IC in ML5 alloy leads to a notable micro grain refinement (Fig. 5).

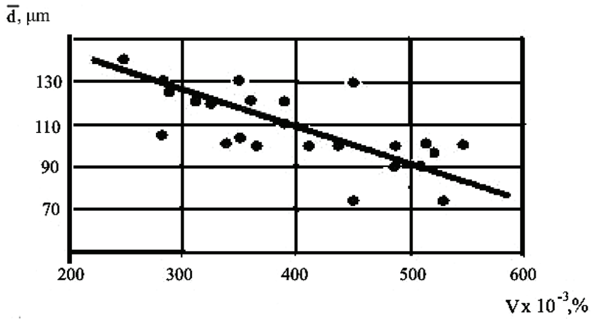
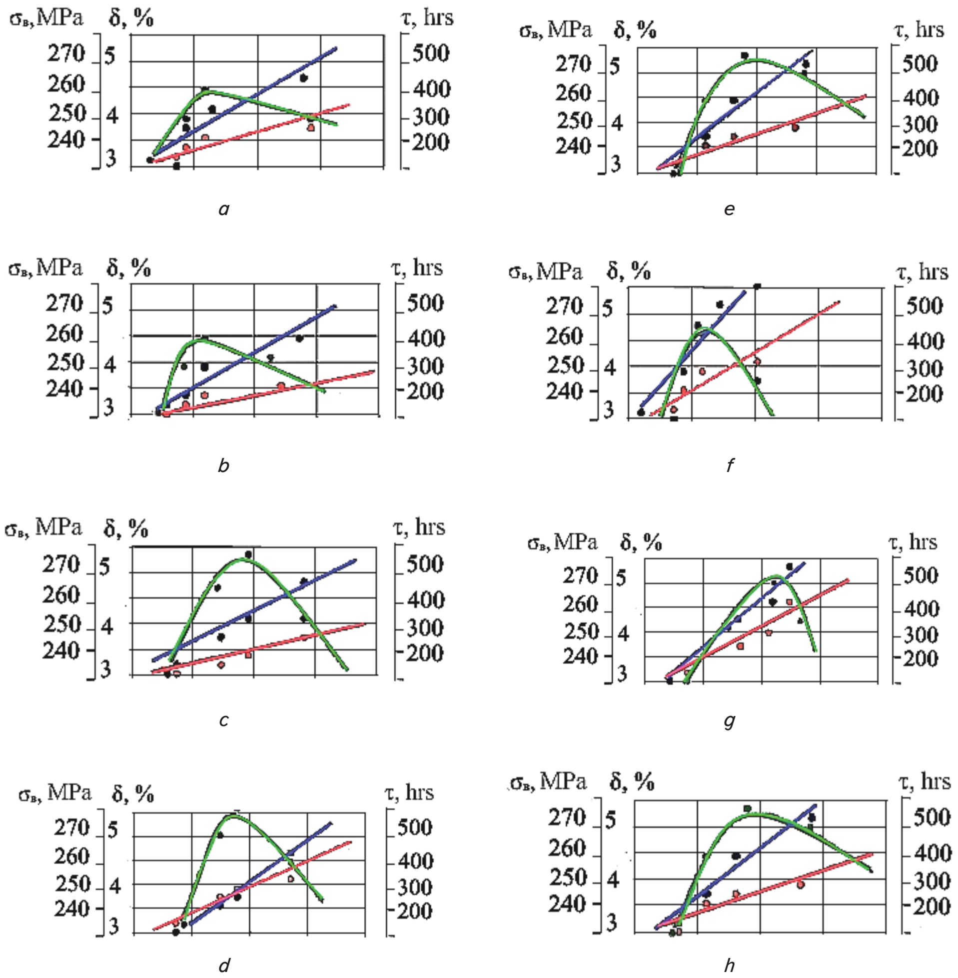


Fig. 5. Influence of the IC volume percent (V) on the grain size (d) in ML5 alloy

With the larger amount of the ICs, both room-temperature and high-temperature strength of ML5 alloy increased. In contrast, the ductility of the alloy displayed a nonlinear dependence on the IC content in so far as it was considerably increasing at the volume percent within a range of 300...450 × $10^{-3}$ and reducing with a further increase of their amount (Fig. 6).

It was established that not only the quantity of the precipitated intermetallic phase but also its topology and morphology exert influence on the properties of the magnesium alloy. The lamellar ICs sized less than 8 μm had a positive effect on the alloy properties whereas the spherical ones showed a positive effect with the size of up to 11.6 μm. Therefore, the spherical shape of the intermetallic compounds is more desirable for improving the properties. Spherical ICs of smaller size groups provided for better strengthening of the alloy. An increase in the ductility of the alloy was observed only with the content of the studied alloying elements of 0.05...0.1 %, when a fraction of the spherical IC increased and the grains were refined. When the content of the alloying elements in the alloy was further increased to 1.0 %, the grain refinement was accompanied with a substantial increase in the quantity of the IC, which resulted in a higher brittleness of the metal and reduced ductility.



*a* *e*

*b* *f*

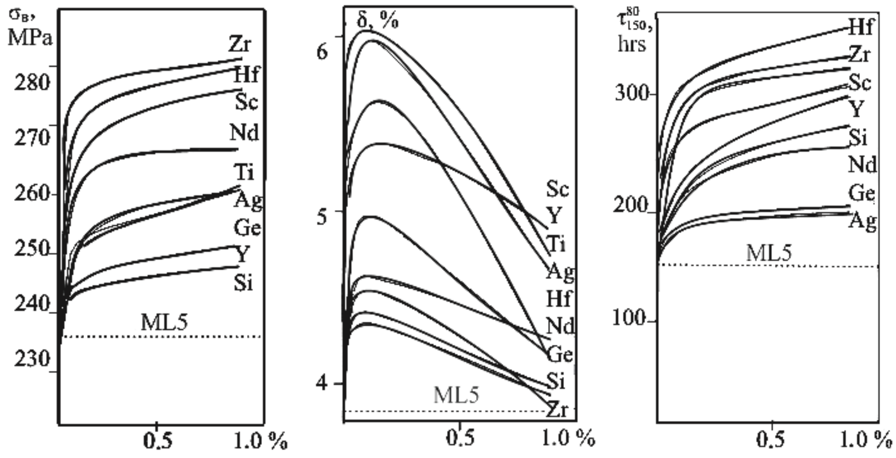
*c* *g*

*d* *h*

Fig. 6. Influence of the IC volume percent on the properties of the alloyed ML5:
*a* — Si; *b* — Ge; *c* — Nd; *d* — Ti; *e* — Y; *f* — Zr; *g* — Hf; *h* — Sc; 🟥 — high-temperature strength at a 150 °C, τ hrs; 🟦 — $\sigma_в$, MPa; 🟩 — δ, %

Depending on the alloy strengthening effect (from maximum to minimum), the alloying elements were distributed in the following series: Zr, Hf, Sc, Nd, Si, Ge, Ti, Y (Fig. 7).

At the concentration of the alloying elements of 0.05...0.1 % wt. also the ductility was improved. A notable improvement in the ductility was observed at 0.05...0.1 % wt. of Y, Ti, Sc, Nd, Hf; a less pronounced ductility increase was attributed to Si and Ge.



*a* *b* *c*

Fig. 7. Influence of the alloying on the room-temperature strength (*a*), ductility (*b*) and high-temperature strength (*c*) (the dotted line represents a property level of the standard alloy)

The high-temperature strength of ML5 alloy showed a distinct improvement with increasing of the alloying element melting point, from germanium to hafnium as well as with increasing their content in the alloy (Fig. 8).

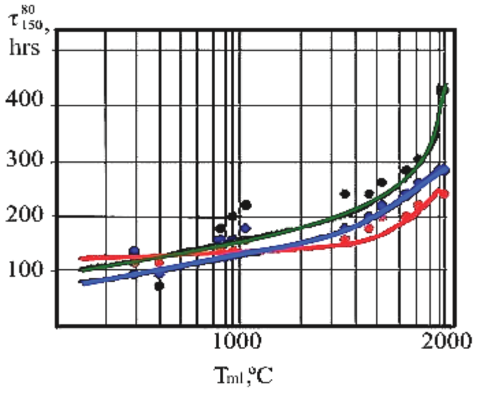


Fig. 8. Influence of the alloying element melting point on the high-temperature strength of ML5 alloy: 🟥 — 0.05 wt. %; 🟦 — 0.1 wt. %; 🟩 — 1.0 wt. %

New magnesium alloys with improved mechanical properties and heat resistance have been developed on the basis of studies [20, 21], which are industrial testing.

## 6. Discussion of the results of research of new magnesium alloys with improved properties

As a result of the undertaken research effort, it was established that alloying the magnesium alloys with Nd, Ge, Si, Y, Sc, Zr, Ti and Hf contributes to the refinement of their structure constituents. In so doing, the content of the alloying elements within a limit of 0.05–0.1 % provides for dispersed spherical IC. The integrated experimental data on the influence of the alloying elements on the magnesium alloy mechanical properties showed that alloying with Zr, Hf, Sc, Nd led to a notable strengthening whereas Y, Ti, Nd, Sc were more attributed to ductility improvement. It was established that the high-temperature strength of the magnesium alloys is defined by a quantity of the heat resistant intermetallic phase formed as a result of alloying the magnesium alloys refractory elements. The most heat resistant alloys were those alloyed with hafnium, zirconium and titanium.

Based on the results of the study, new magnesium alloys with improved properties were developed, which are currently being subjected to industrial testing trials. The use of the new alloys is advisable in aerospace, car making, instrumentation industry and other manufacturing sectors.

## 7. Conclusion

1. Based on two criteria (atomic diameters ≤ 15 %, electro negativity difference ≤ 0.4, melting point of the alloying element is higher than the one of magnesium) elements (Nd, Ge, Y, Sc, Zr, Si, Ti and Hf) have been determined, capable of improving the structure, mechanical properties and special performance characteristics properties of magnesium alloy castings.
2. The studied alloying elements in the magnesium alloys form complex intermetallic phases possessing the topology and morphology features. Alloying with the elements results in 30...40 % micro- and macro grain refinement and increased micro hardness of the structure constituents. With the content of each alloying element in the alloy 0.05...0.1 % predominantly spherical intermetallic compounds of the finer size group are formed, which lead to grain refinement and mechanical properties improvement.
3. The developed new magnesium alloy compositions [20, 21] allow for improvement in the ultimate tensile strength by 30 % and ductility by 50 % with a nearly twofold increase in the high-temperature strength.

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