

Structural optimization of cast in-situ aluminum matrix composites: challenges and opportunities

V. B. Deev*, Professor, Chief Researcher³, Professor at the Faculty of Mechanical Engineering and Automation¹, Professor at the Department of Metal Forming², e-mail: deev.vb@mail.ru
E. S. Prusov, Associate Professor, Department of Functional and Constructional Materials Technology³
E. Kh. Ri, Professor, Head of the Department of Foundry Engineering and Metal Technology⁴

¹ Wuhan Textile University, Wuhan, China.

² National University of Science and Technology "MISIS", Moscow, Russia.

³ Vladimir State University named after Alexander and Nikolay Stoletovs, Vladimir, Russia.

⁴ Pacific National University, Khabarovsk, Russia.

In this work, a comprehensive analysis was performed to assess the impact of chemical and physical modifying influences on the structural formation of cast metal matrix composites with in-situ formed reinforcing phases, using the pseudo-binary Al – Mg₂Si system as a case study. Relevant problematic issues were identified, and prospective approaches to controlling the structural-morphological parameters of aluminum matrix composites during melting and crystallization were determined. Recent advancements in investigating the influence of thermal-rate and electromagnetic pulsed processing of melts on the structure of in-situ aluminum matrix composites were summarized. An evaluation of the influence of various modifying elements on the structure of pseudo-binary aluminum alloys and composites was provided. The feasibility and effectiveness of employing alkali and alkali-earth metals for the modification of Al – Mg₂Si aluminum matrix composites were confirmed, replacing expensive rare-earth metals. Additionally, it was demonstrated that lithium yields the most significant modifying effect, considering a complex assessment of its impact on the structure.

Key words: aluminum matrix composites, in-situ reinforcement, structure formation, modification, structural-morphological parameters, physical and chemical effects

DOI: 10.17580/nfm.2024.01.07

Introduction

Cast aluminum matrix composites are considered as a promising and prospective alternative to traditional casting ferrous and non-ferrous alloys for the fabrication of critical components in various industrial sectors, including automotive, aerospace, and others [1–3]. Advanced experiences demonstrate that controlled formation of heterophase reinforced structures is a promising approach to enhance the structural properties and operational characteristics of products [4, 5]. A major impediment to the widespread industrial adoption of cast aluminum matrix composites lies in technological challenges associated with the poor wetting of exogenous reinforcing particles by aluminum melts, particle agglomeration during melt stirring, gas and oxide entrapment from the melt surface, and other issues [6–8]. One research direction aimed at addressing these challenges involves the creation of aluminum matrix composites with endogenous reinforcing phases characterized by enhanced thermodynamic stability, better interfacial adhesion, more uniform distribution of reinforcing components, and, in most cases, lower production costs due to the exclusion of specialized equipment [9].

However, the primary concern in designing cast in-situ aluminum matrix composites is the rational selection of the initial component systems, based on which their synthesis is feasible through chemical interactions between precursors and matrix melts or in crystallization processes. The intermetallic compound Mg₂Si stands out due to its comparatively high melting temperature (1085 °C), high hardness (4.5 GPa), low density (1880 kg/m³), and high elastic modulus (120 GPa), thus exhibiting higher specific stiffness compared to most other intermetallic compounds, which determines its potential and prospects as a reinforcing phase in the production of aluminum matrix composites [10]. Importantly, composite materials of the Al – Mg₂Si system can be easily obtained under traditional metallurgical processes without using powder precursors, solely through the rational selection of temperature-concentration conditions for composite synthesis, significantly simplifying the technology and reducing the cost of casting production [11]. However, in such processes, the formed primary Mg₂Si particles are characterized by large sizes and unfavorable morphology. Shifting towards high concentrations of Mg₂Si (20–25 wt.% and above) leads to the formation of branched dendritic-like complexes, substantially reducing the mechanical properties of the obtained materials and limiting their practical application.

*Correspondence author.

The objective of this study is to analyze the influence of chemical and physical modifying treatments on the structure formation of cast metal matrix composites with endogenous crystallization-originating reinforcing phases, using the Al – Mg₂Si system as an example.

1. Actual problems of structure control of aluminum matrix composites with endogenous crystallization-originating reinforcing phases

As demonstrated by numerous research findings, the Mg₂Si phase in the Al – Mg – Si ternary system can exist in two forms: the eutectic Mg₂Si phase within the eutectic structure (Al + Mg₂Si) and primary Mg₂Si crystals. The Al – Mg₂Si system is thermodynamically described as a binary pseudo-eutectic system with a pseudo-eutectic point at 13.9 wt.% Mg₂Si [12]. Beyond this concentration, Mg₂Si crystals precipitate in the transitional two-phase region as the primary phase. In the hypoeutectic compositions of the pseudo-binary Al – Mg₂Si system, crystallization initiates in the two-phase region $L + \alpha$ -Al. It is noteworthy that the transition from the eutectic Mg₂Si phase to the primary crystallizing particles, while controlling their dispersion and morphology, is crucial for obtaining cast aluminum matrix composites. For instance, the composition of Al + 15 wt.% Mg₂Si is close to the pseudo-eutectic point and (according to Thermo-Calc simulation) is characterized by a transitional two-phase zone $L + Mg_2Si$ in the temperature range of 594 to 578 °C. Further increasing the Mg₂Si content to 20 and 25 wt.% extends the temperature range of the solid-liquid region (673 to 578 °C and 680 to 579 °C, respectively).

It has been shown [13] that depending on external conditions, the Mg₂Si phase can crystallize in various morphologies, including octahedral, funnel, truncated octahedral, cube, and dendrite shapes. In conventional melting and casting technologies, primary Mg₂Si crystals form imperfect octahedral or dendritic complexes growing along the first stable dendrite in the $\langle 100 \rangle$ direction [14]. The increase in the Mg₂Si phase content is accompanied by an increase in its size and a change in morphology. Unfavorable morphology leads to stress concentration at the sharp edges of dendrites, initiating crack formation, thereby reducing the mechanical properties of composites and limiting their potential applications. Experimental data confirm that the anisotropic growth rate of Mg₂Si crystals can be artificially controlled by varying external growth conditions, allowing closer approximation to equilibrium morphology (faceted octahedron).

Thus, the Al – Mg – Si ternary system within the aluminum corner serves as the basis for creating cast aluminum matrix composites reinforced with Mg₂Si particles. Understanding the thermodynamic peculiarities of crystallization in the quasi-binary intersection of Al – Mg₂Si is a necessary condition for controlling the structural-morphological characteristics of the resulting composites. In particular, a promising approach to altering the

morphology of the Mg₂Si phase may involve searching for temperature-concentration conditions for its formation in a compact and favorable form by controlling interfacial thermodynamics at the “melt – Mg₂Si particle” boundaries, which can be achieved by applying various physical influences to liquid and crystallizing composite melts. Another important direction is the search for modifying additives that can act as centers for the crystallization of the Mg₂Si phase or restrict the growth of its primary crystals.

2. Classification of known methods for modifying in situ aluminum matrix composites through the application of physical treatments and their characteristics

Processing composite melts using various physical treatments represents a promising approach to controlling the structural and morphological characteristics of cast aluminum matrix composites with endogenous reinforcing phases. Depending on the applied processing method and the state of the melt during treatment, qualitative effects can be achieved, such as reducing the size of forming reinforcing phases, breaking agglomerates, altering phase morphology to a more compact form, increasing the overall quantity of reinforcing particles, and improving the uniformity of their distribution within the casting volume [15]. It is pertinent to categorize the methods of applied physical treatments on the base of the melt state during processing into treatment during melting, pouring, and crystallization in the casting mold (Fig. 1).

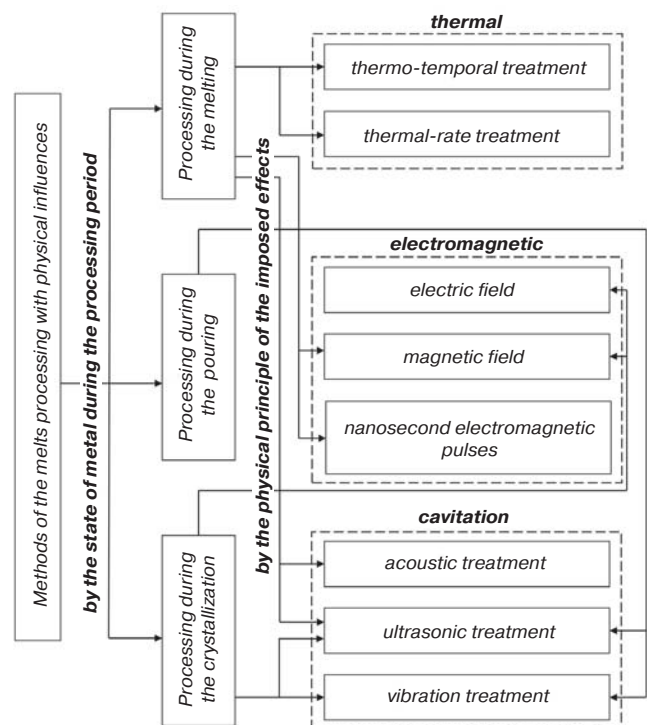


Fig. 1. Classification of physical methods for melts processing based on the metal state during treatment and the physical principle of the applied treatment [16]

Thermal treatments (thermo-temporal and thermal-rate treatments), electromagnetic treatments (processing with electric and magnetic fields, application of nanosecond electromagnetic pulses), and cavitation effects (acoustic, ultrasonic, vibrational treatments, etc.) are distinguished.

Physical modifying treatments offer several advantages compared to traditional chemical methods of processing melts, particularly in terms of a more favorable environmental situation, the avoidance of costly modifiers, and the preservation of the chemical composition of the melt during melting. This, unlike technologies using modifier elements, prevents the accumulation of excessive impurities during subsequent remelting, etc. [17–19]. However, physical treatments on melts cannot be implemented arbitrarily; they require scientific justification for their application and optimization of processing parameters in each specific melting technology and modifying treatment of metal matrix composites based on aluminum alloys. Additionally, in many cases, the modifying effect after processing composite melts with physical methods is lower than expected, which can be overcome by simultaneously combining multiple technological options and methods of physically treating melts, as well as by jointly using physical methods with traditional chemical modifiers. In the latter case, a significant increase in the efficiency of the modifying action of chemical additives can be achieved through the additional application of physical treatments to melts during melting or crystallization.

3. General facts about the influence of thermal-rate and electromagnetic pulse processing of melts on the structure of in-situ aluminum matrix composites

In recent years, a significant volume of experimental data has been accumulated regarding the impact of processing parameters on the molten state of endogenously reinforced metal matrix composites based on cast aluminum alloys (Al – Mg₂Si system) using thermal-rate and electromagnetic pulse treatments, and the effects of these treatments on the formation of structure and the desired set of properties. Specifically, the application of thermo-temporal and thermal-rate treatments to Al – Mg₂Si melts, with an isothermal hold time of 30 minutes at 900 °C, results in a substantial refinement of endogenous reinforcing Mg₂Si inclusions and an increase in their overall quantity [20]. The fractional content of the Mg₂Si phase significantly influences the changes in structural-morphological parameters during thermal-rate treatment of Al – Mg₂Si melts. In the hypereutectic compositions range of pseudobinary Al – Mg₂Si system, raising the melt overheating temperature to 900 °C leads to a pronounced refinement of all structural components. Further temperature increase to 950 °C results in some coarsening of α -solid solution and Mg₂Si crystals. The melt cooling rate to the pouring temperature should be maintained in the range of 70–90 °C/min. Based on the accumulated experience of thermal-rate processing of composite melts, achieving

these cooling rates is possible by adding the unheated pieces of pre-prepared material of the same composition into the melt in the range of 15–25% of the total processed melt mass. During thermal-rate treatment of Al – Mg₂Si melts in the hypereutectic composition range, the primary Mg₂Si crystal refinement is attributed to changes in crystallization thermodynamic conditions due to increased undercooling and the (presumably) activation of insoluble oxide particles (spinel) for heterogeneous nucleation [21]. The modifying effect of thermal-rate treatment on the solid solution grains and eutectic components is associated with increased melt microhomogeneity at high-temperature overheats and the fixation of this effect through rapid melt cooling to the casting temperature, as well as the introduction of additional potential crystallization centers by adding unheated charge materials to the melt [22].

During elaboration of the parameters of nanosecond electromagnetic pulse treatment it was shown that at melting temperature regimes used for industrial aluminum alloys of the Al – Mg – Si system, melt treatment in the range of generator amplitudes from 5 to 15 kV has a strong modifying effect on the constituents of the matrix alloy structure but minimal impact on the sizes and morphology of primary Mg₂Si crystals [23]. Based on the obtained data, it is recommended to apply combined treatments to composite melts through thermal-rate treatment and the imposition of nanosecond electromagnetic pulses. This approach allows influencing the structural-morphological parameters of all phases in cast Al – Mg₂Si aluminum matrix composites. Increasing the amplitude and frequency of the nanosecond electromagnetic pulse generator results in solid solution and eutectic component refinement, and increased Mg and Si content in the eutectic. Higher amplitudes reduce the quantity and sizes of eutectic inclusions inside primary Mg₂Si crystals, and increase the microhardness of the primary phase to levels around 600–650 HV, close to the microhardness of defect-free Mg₂Si crystals. The impact of nanosecond electromagnetic pulses on metallic melts is described from the perspective of the quasicrystalline model of melt structure, suggesting that irradiation induces energy fluctuations, alters the near-order structure of atoms, and shortens their duration of existence, reducing the disordering temperature of the structure [24–26]. An increase in melt undercooling enhances the rate of solid phase nucleation centers formation per unit volume of the melt.

The study [27] explores the structure features of cast ingots of Al + 25 wt.% Mg₂Si composites obtained under various frequency and amplitude parameters of nanosecond electromagnetic pulse exposure during crystallization. In all cases, Mg₂Si particles exhibited dendritic morphology and demonstrated anisotropic behavior during crystal growth, typical for pseudobinary aluminum matrix composites with high primary Mg₂Si phase content. Nanosecond electromagnetic pulse treatment during crystallization increases the observed quantity of primary

Mg_2Si particles in the ground area by more than an order of magnitude. The average linear size of particles, determined as the Feret diameter, in the untreated composite was $147\ \mu m$. Imposing nanosecond electromagnetic pulse on the crystallizing composite melt under the tested frequency and amplitude parameters allows reducing the average sizes of primary phase particles to values ranging from 40 to $65\ \mu m$. Additionally, radical morphological changes in the pseudobinary eutectic during the crystallization of melts under nanosecond electromagnetic pulse influence was noted.

Generally, the observed effects of thermal-rate and electromagnetic pulse treatment on the size and morphological characteristics of structural components in aluminum matrix composites based on the pseudobinary Al – Mg_2Si system may have significant practical implications for improving the melting and casting processes. This could lead to enhanced mechanical and operational properties of cast products, thereby increasing interest in these promising materials from the commercial sector.

4. Application of chemical action modifiers for controlling the structure of in-situ aluminum matrix composites

Over the recent years, extensive research has been conducted to investigate the impact of various modifying elements on the structure of pseudo-binary aluminum alloys and composite materials within the Al – Mg_2Si system. During this investigation, the notably high effectiveness of incorporating minor additions of rare-earth metals has been substantiated. This effectiveness is manifested in the reduction of average sizes and alteration of the morphology of primary Mg_2Si particles, as well as in the modification of the pseudo-eutectic ($\alpha + Mg_2Si$) microstructure. Several tested modifiers for the primary particles

of the reinforcing phase in Al – Mg_2Si composites, along with the outcomes of their application, are summarized in **Table 1**.

In the context of the Al – Mg – Si – La system, we investigated the influence of La in concentrations up to 1 wt.% on the phase composition and processes of structural formation [46]. A significant modifying effect of La on the eutectic phase Mg_2Si was observed. The shape of the latter changed slightly after the addition of 0.1 wt.% La, but at higher concentrations, it evolved from plate-like structures to thin flakes. This effect was attributed to the adsorption of the triple lanthanum-containing phase $AlLaSi$ (presumably Al_2LaSi_2) at the solid/liquid interface, inhibiting the growth of Mg_2Si .

In the near-eutectic composition range, the modifying effect of lanthanum can be traced in the case of the Al + 15 wt.% Mg_2Si aluminum matrix composite (**Fig. 2**). Samples (hereinafter) were obtained by pouring a composite melt of nominal composition at $720\ ^\circ C$ into a vertical steel mold with an internal diameter of 20 mm and a height of 100 mm (cooling rate $\sim 10\ K/s$). The addition of lanthanum leads to a reduction in the average sizes of primary Mg_2Si crystals (measured by Feret diameter) from $\sim 38.4\ \mu m$ in the initial state to $\sim 27.5\ \mu m$ at 0.3 wt.% La.

The aforementioned premises necessitate further development of efficient and economical methods for modifying the impact on all structural components of Al – Mg_2Si composites. It should be noted that a significant number of tested modifiers for primary particles of the reinforcing phase in Al – Mg_2Si system composites do not yet indicate a complete solution to the issue of controlling their structure and properties. Key indicators of the effectiveness of modifying impact should include the average size of Mg_2Si primary particles, crystal shape, aspect ratio, and the number of particles per unit surface area. However, in many cases, it is not possible to compare the effectiveness of modifying additives based on published data, as many significant parameters are not reported, and the conditions of experiments vary significantly. The literature also lacks information on the combined influence of chemical and physical modifying actions on the structure formation of metal matrix composites based on the Al – Mg_2Si system, highlighting the relevance of continuing research in this direction to discover new efficient and economical methods for controlling the solidification structure and properties of these promising materials.

5. Assessment of the prospects of alkali and alkaline earth metals application for modifying in-situ aluminum matrix composites

Significant practical interest lies in exploratory research aimed at identifying chemical modifying additives that can serve as alternatives to expensive rare earth metals in the production of Al- Mg_2Si composites. The selection of alkali and alkaline earth metal additives should be based on an evaluation using techno-economic criteria and

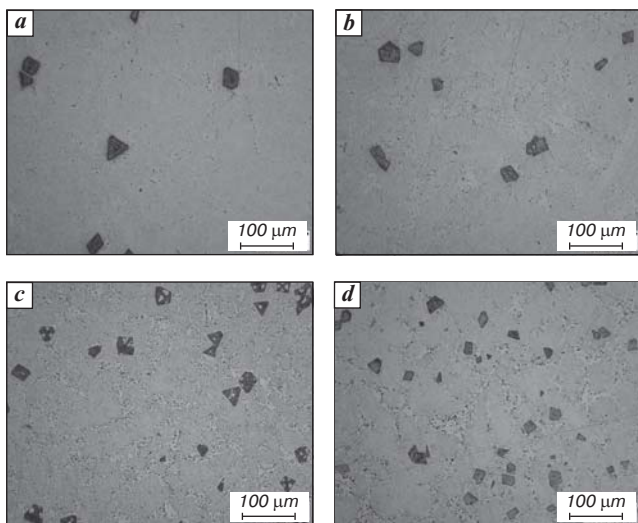


Fig. 2. Microstructure of the Al + 15 wt.% Mg_2Si aluminum matrix composite: without modification (a); 0.1 wt.% La (b); 0.2 wt.% La (c); 0.3 wt.% La (d)

review data. The economic feasibility assessment of modifier usage was performed by constructing and analyzing diagrams of distribution based on indicators of element extraction and production (HHIp) and estimated reserves of elements (HHIr) using the Herfindahl-Hirschman Index [47]. Data on element production volumes and reserves from the United States Geological Survey (USGS) were used to estimate corresponding HHIp and HHIr values.

A distribution diagram of elements based on Herfindahl-Hirschman Index values for production volumes and estimated reserves was compiled (Fig. 3).

In general, if either the HHIp or HHIr index exceeds 6000, the use of such components becomes high-risk in terms of the uncertainty of future supplies [48]. From this perspective, the preferred additives for use are considered to be Ba, Sr, Na, Ca, and Li. In turn, additives Bi, Sc, and

Table 1
Some tested modifiers for primary particles of the reinforcing phase in Al – Mg₂Si composites and the results of their application

| Element | Optimal amount | Composite | Effect | Ref. |
|----------------------------------|----------------|----------------------------|--|--------------|
| Bi | 0.4 wt. % | Al – 20%Mg ₂ Si | Mg ₂ Si particle size decreased by 35% from 179.40 to 115.92 μm Average particle number increased from 12 to 38 pcs/mm ² | [28] |
| Li | 0.3 wt. % | Al – 15%Mg ₂ Si | Reduction of Mg ₂ Si particles from 30 to 6 μm Refining of the pseudo-eutectic | [29] [30] |
| Sb | 0.8 wt. % | Al – 20%Mg ₂ Si | Mg ₂ Si particle size decreased from 179.40 to 134.65 μm Average number of particles increased from 12 to 29 pcs/mm ² | [28] |
| | 0.91 wt. % | Al – 20%Mg ₂ Si | Mg ₂ Si particle size decreased from 54.2 to 46.7 μm Modification of pseudo-eutectic | [31] |
| Sr | 0.01 wt. % | Al – 20%Mg ₂ Si | Mg ₂ Si particle size decreased from 179.40 to 125.55 μm Average number of particles increased from 12 to 35 pcs/mm ² | [28] |
| | 0.05% | Al – 25%Mg ₂ Si | Severe fragmentation and change of morphology to polygonal (quantitative estimates of average size are not given) Strength increases from 90 to 120 MPa | [32] |
| Sc | 0.25 wt. % | Al – 11%Mg ₂ Si | Reduction of pseudo-eutectic Mg ₂ Si from 12.4 to 1.8 μm Modification of aluminum solid solution grains Strength increases from 269 MPa to 334 MPa | [33] |
| Eu | 0.1% | Al – 18%Mg ₂ Si | The particle size of Mg ₂ Si decreased from ~90 μm to ~16 μm | [34] |
| Ba | 0.2 wt. % | Al – 13Mg – 7Si | Reduction of Mg ₂ Si particle size from 65.3 to 35 μm Change of shape to perfectly cubic | [35] |
| Ca | 0.15% | Al – 20%Mg ₂ Si | Reduction of Mg ₂ Si particle size from 100 to 44.1 μm Modification of the pseudo-eutectic | [36] |
| Gd | 0.5 wt % | Al–15%Mg ₂ Si | Reduction of Mg ₂ Si particles from 30 to 12 μm Change of shape to polyhedral Decrease in volume fraction of Mg ₂ Si particles Decrease of interplastic distance in pseudo-eutectic from 1.8 μm to 1 μm | [37] |
| | 1.0 wt % | Al – 15%Mg ₂ Si | Reduction of Mg ₂ Si particles from 40 μm to 20 μm Decrease in aspect ratio from 1.34 to 1.25 Increase in particle number from 495 to 1167 pcs/mm ² | [38] |
| Gd + melt thermal-rate treatment | 1.0 wt. % | Al–15%Mg ₂ Si | Reduction of Mg ₂ Si particles from 40 to 13 μm with additional thermal-rate treatment at 850 °C | [39] |
| Nd | 0.5 wt. % | Al – 18%Mg ₂ Si | Reduction of Mg ₂ Si particles from 47.5 to 13.0 μm Change in shape from dendritic to polygonal Change of morphology of pseudo-eutectic | [40] [41] |
| Pr | 0.7% | Al – 18%Mg ₂ Si | Reduction of Mg ₂ Si particles from 65 to 17 μm Change of morphology to polyhedral Increase in strength from 191 to 262 MPa and ductility from 0.73 to 1.9% | [42] |
| Ni | 5 wt. % | Al–15%Mg ₂ Si | Reduction of Mg ₂ Si particles from 42 μm to 17 μm Refining of the pseudo-eutectic | [43] |
| Cu | 3.0 wt. % | Al – 15%Mg ₂ Si | Reduction of crystallization interval Increase in strength up to 290 MPa | [44] |
| HfP (Al – 7Hf – 1.2P) | 2.50% | Al – 20%Mg ₂ Si | Reduction of Mg ₂ Si particles from 121.70 μm to 19.20 μm Elimination of coarse Mg ₂ Si dendrites | [45] |

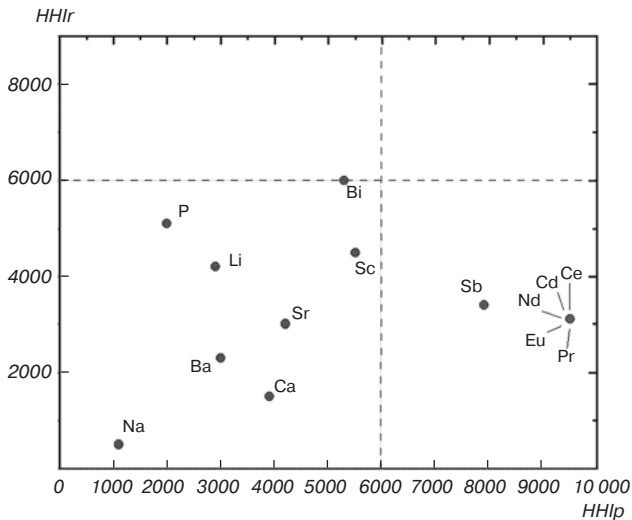


Fig. 3. Evaluation of economic feasibility of using popular modifying elements for Al – Mg₂Si aluminum matrix composites

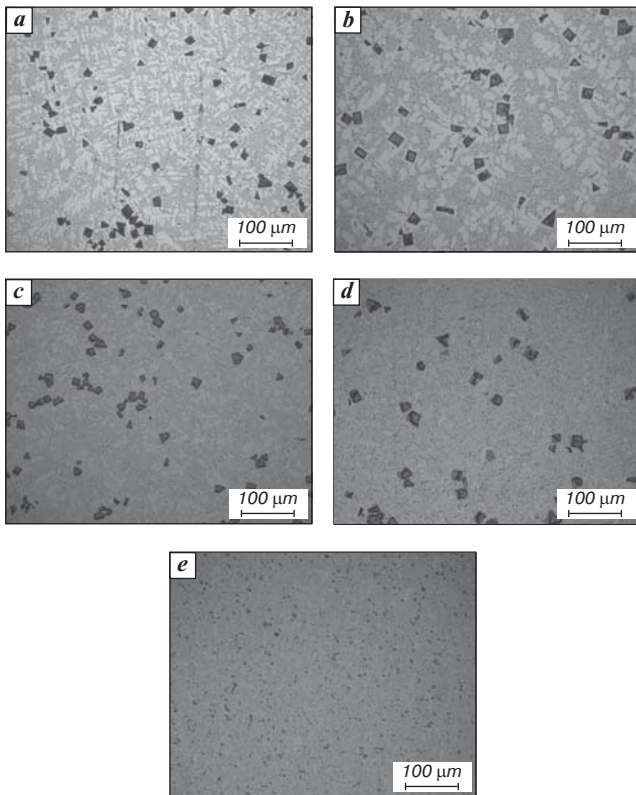


Fig. 4. Microstructure of Al + 15 wt.% Mg₂Si aluminum matrix composite after addition of modifiers (rational quantities determined experimentally): 0.15 wt.% Ba (a); 0.2 wt.% Sr (b); 0.25 wt.% Na (c); 0.25 wt.% Ca (d); 0.3 wt.% Li (e)

P were excluded from consideration in the current analysis due to their proximity to the threshold values of the HHI indices (~6000), although this does not restrict the potential application of these additives in cases where economic factors do not play a decisive role.

Additives of alkali and alkaline earth metals, selected based on the assessment of the economic feasibility of

their use, were experimentally tested. Subsequently, using the example of an Al + 15 wt.% Mg₂Si aluminum matrix composite, information is presented illustrating the influence of barium, strontium, sodium, calcium, and lithium on the dispersity, distribution, and morphological characteristics of primary Mg₂Si particles (Fig. 4). Rational concentrations of modifying additives providing the best modifying effect in terms of the evaluated structure parameters of the composites were determined based on the results of series of conducted experiments. The structural components of the aluminum matrix composite with 15 wt.% Mg₂Si in its unmodified state include primary silicides identified by dark irregularly shaped inclusions, as well as a pseudo-binary eutectic (α + Mg₂Si) and a small amount of α -solid solution dendrites. The addition of the tested alkali and alkaline earth metals led to a significant change in the structure of the composites. In particular, the introduction of barium and strontium resulted in a decrease in the fraction of the pseudo-binary eutectic and the appearance of a significant number of α -solid solution dendrites. This may be associated with the shift of critical points on the corresponding phase diagrams and requires further research to clarify the detailed mechanisms of the occurring changes. The average sizes of primary Mg₂Si phase particles (by Feret diameter) upon the introduction of the tested metals, except for lithium, remained in the range of 24...30 μ m, which generally corresponds to previously achieved results for the modifying effect when using lanthanum as a modifier for a composite of similar composition. Specifically, with the addition of 0.2 wt.% Sr, the average sizes of Mg₂Si particles were ~24.73 μ m, with 0.15 wt.% Ba ~25.53 μ m, with 0.25 wt.% Na ~25.97 μ m, and with 0.25 wt.% Ca ~30.13 μ m. It is noteworthy that a radical reduction in the particle sizes of Mg₂Si to ~7.92 μ m occurred with the addition of 0.3 wt.% Li.

In general, the obtained experimental data in conjunction with the published results of other scientific teams indicate the feasibility and efficiency of utilizing alkali and alkaline-earth metals to modify cast aluminum matrix composites of the Al – Mg₂Si system instead of expensive rare-earth metals, with the greatest modifying effect achieved with the use of lithium.

6. Prospects for enhancing the efficiency of modifying action of rare-earth and alkaline earth metal additives

Further exploration of methods to enhance the effectiveness of modifier action of alkali, alkaline earth, and rare-earth metals is advisable in the direction of combining various chemical and physical influences on melts. Investigating the regularities and mechanisms of the combined impact of chemical and physical modifying influences on the structural formation and property development of cast metal matrix composites of the Al – Mg₂Si system will be an objective prerequisite for creating new efficient and economical means of controlling the cast structure and properties of these promising materials.

The achieved results in modifying treatment open new possibilities for expanding the range of engineering components where the replacement of traditional alloys with cast aluminum matrix composites of the Al – Mg₂Si system is justified. This is particularly attributed to overcoming known drawbacks inherent in Al – Mg₂Si composite materials in the hypereutectic composition range (coarse morphology of pseudo-eutectics, large crystals of primary phases with unfavorable morphology) through the use of tested variants of melt modifying treatment. The obtained results will facilitate the broadening of practical applications for cast aluminum matrix composites with endogenous crystallization-originated reinforcing phases, forming the basis for the development of specific practical recommendations for the implementation of new materials and the technologies of their production and modifying treatment in the conditions of existing industrial manufacturing.

Acknowledgments

This research was funded by the Russian Science Foundation (Project № 20-19-00687-II, <https://rscf.ru/project/23-19-45019/>).

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