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Metallurgy

Influence of manganese additives on the microstructure of the Al-Fe-Si alloy system synthesized through arc surfacing with a consumable electrode

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ABSTRACT

Modern technologies used for the synthesis of various alloys, including aluminum-based metal-ceramics, require detailed and comprehensive studies, especially in the case of alloys that have not yet found widespread industrial application due to a lack of sufficient scientific data. The Al-Fe-Si alloy system is of particular interest due to the simplicity of its composition and the wide variety of phases that form depending on the ratio of the alloy's base components. The intermetallic Al-Fe-Si metal-ceramic alloy, with an increased simultaneous content of both iron and silicon, was synthesized by arc surfacing with a consumable electrode. This article presents experimental studies of the metallographic analysis of the Al-Fe-Si alloy, enriched with both iron and silicon, along with manganese additives. Studying the effect of manganese in this specific alloy composition allowed for an in-depth assessment of the morphology of intermetallic compounds, phase distribution, and overall structural stability. Preliminary phase composition modeling helped identify the phases in the synthesized alloy. It was found that a minor addition of manganese could stabilize the microstructure and result in the formation of coalesced intermetallic particles. Further investigation of its effects on phase transformations and structure will provide insights into optimizing compositions for broader applications in conditions of high loads and temperatures.

Keywords: Al-Fe-Si, microstructure, ThermoCalc software, intermetallic phases, arc surfacing with a consumable electrode.

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Introduction

Aluminum alloys maintain a leading position among promising materials for the development of durable, lightweight, and wear-resistant components that meet the increasingly stringent requirements of modern mechanical engineering. The technologies, methodologies, and approaches to strengthening these alloys are diverse and have attracted considerable attention from researchers worldwide [[1], [2], [3], [4]]. Of particular interest are alloys within the Al-Fe-Si system, which have gained significant attention due to their potential for property enhancement through alloying and modification, as well as their compatibility with additive manufacturing techniques [5]. Additive manufacturing of alloys not only offers rapid production and prototyping capabilities but also allows for the creation of nanoscale inclusions and intermetallic compounds, enabling the achievement of hybrid properties such as low weight combined with high hardness, and the production of multilayer composite structures.

The growing relevance of Al-Fe-Si alloys is driven by several key attributes:

- Exceptional wear resistance and hardness,
- Lightweight combined with high strength,
- Excellent heat resistance and thermal stability,
- Superior corrosion resistance.

Even minor additions of alloying elements can significantly influence the microstructure and properties of these alloys [[6], [7], [8], [9], [10]]. Modifications in the morphological structure of phases and phase composition can be effectively achieved through the strategic incorporation of alloying elements. These additions stabilize the structure, modify phase distribution, and promote the formation of intermetallic compounds with varying dispersions and structures [[11], [12], [13], [14]].

These properties are particularly crucial for the development of advanced alloys with superior performance characteristics in industries such as aerospace, automotive, and space exploration.

While there is a wide variety of aluminum alloys, most contain a relatively limited set of alloying elements. The influence of these elements is substantial, ultimately imparting new or specialized properties to the alloys. Alloying elements can be broadly categorized into three groups: primary alloying elements, auxiliary additives, and impurities. Depending on the specific alloy, the same element may serve multiple functions [15].

Primary alloying elements commonly found in aluminum alloys include magnesium, zinc, copper, silver, and silicon. These elements are considered "primary" because they are added in substantial quantities and play a key role in determining the microstructure and primary properties of the alloy.

However, among these, silver and germanium are rarely used as primary alloying components due to economic factors. Silver is a precious and costly metal, and germanium, although valuable, is primarily used in the semiconductor industry. Furthermore, the addition of these elements does not confer significantly advantageous properties compared to other, more commonly used alloying elements.

Research has explored the effects of additional additives such as germanium, cerium, and scandium on the strengthening of solid solutions, structural refinement, and phase distribution in aluminum alloys [[16], [17], [18], [19]]. While these studies have yielded promising results, the practical application of these elements is limited by factors such as high cost, complex processing techniques, and the challenges associated with the purification of the final material. The economic viability of using these elements for large-scale production remains questionable.

A particularly promising avenue of research is the effect of small amounts of manganese (approximately 1.5%) on the phase composition, mechanical, and technological properties of Al-Fe-Si alloys containing high levels of both iron and silicon [[20], [21], [22]]. This study investigates the microstructure and phase morphology of an Al-Fe-Si alloy, incorporating manganese as a cost-effective and accessible alloying element. The aim of this research is to evaluate the impact of manganese alloying on the final microstructure, phase distribution, and phase dispersion, particularly in the context of high iron and silicon content.

Experimental part

The Al-Fe-Si alloy system was synthesized via surfacing with a consumable electrode [[23], [24], [25]]. This additive manufacturing technique was selected based on the specific advantages of the process. Key benefits of this method include high processing speeds, reliable oxidation protection through the use of flux, the ability to incorporate additional alloying elements via flux components, and the flexibility to achieve diverse alloy compositions without the need for preparing a separate filler material. Furthermore, the process is autonomous. However, certain limitations are associated with this method, such as the formation of pores during synthesis, quantitative constraints on the introduction of alloying elements, and directional heat dissipation, which, coupled with gravitational effects, results in a structural orientation in the vertical direction.

The experimental setup involved the following equipment:

A VDM-1202 welding rectifier with an RB-301 ballast rheostat, which served as the power source for melting the alloy components;

A stationary filter and ventilation table (SS-1200/SP) for removing gaseous products generated during synthesis. The alloy components were melted by creating an electric arc with a welding current of 290 A.

Detailed experimental procedures are described in [26]. To synthesize the alloy, grade 3 steel was employed as the iron source, also serving as a consumable electrode. AD31 aluminum alloy was used as the source of aluminum and silicon, with the latter provided in powder form, which was crushed and sifted to obtain a particle size of -500 µm. Silicon was placed between layers of aluminum, each 3 mm thick. The silicon layer was applied wet by weight, resulting in layers approximately 1-1.5 mm thick. The entire stack was covered with a flux layer 20-30 mm thick. Manganese was introduced into the system through the flux in small amounts.

Two types of AN-348 flux were used: fused and sintered. Despite having identical compositions, the fused flux more actively participates in the reaction, leading to alloy saturation with approximately 1.5% manganese, along with a similar quantity of silicon. After cooling, the synthesized material exhibited a slag crust with a characteristic glassy appearance, which was easily removed during cooling. In contrast, the use of sintered flux resulted in more pronounced interaction between the base components and those present in the flux. This was accompanied by the saturation of the slag crust with iron atoms, and the formation of transition metal-ceramic compounds directly in the interface between the material and the flux. The slag crust in this case proved more difficult to remove, and the base metal was saturated with no more than 0.2% manganese, with no significant enrichment in silicon. Furthermore, the volume of gaseous synthesis products was considerably higher.

Templates were cut from the samples synthesized using this method for the preparation of microsections. Sample preparation followed standard procedures, including grinding and polishing using Struers equipment, with subsequent examination under an Altami optical microscope. The actual composition of the alloys was determined using an Olympus VantaElementS X-ray fluorescence analyzer. To gain a more comprehensive understanding of phase formation, phase transformations from the melting point to room temperature were modelled using ThermoCalc software. Specifically, the TCAL8: Al-Allous v8.2 database was utilized to predict the number of phases formed for the actual composition of both alloys.

Results and Discussion

Table 1 presents the composition of the synthesized alloys. The alloy doped with manganese is referred to as AlFeSiMn, while the alloy with manganese at the impurity level is designated as AlFeSi.

According to theoretical calculations, both alloys consist of three primary phases at room temperature. The AlFeSiMn alloy at room temperature comprises approximately 40% of the β phase, 38% of the θ phase, and 22% of the θ_2 phase. The key difference between the θ and θ_2 phases lies in the increased silicon content in the latter. A minor quantity of impurity atoms is present in the form of nickel aluminide and other phases, such as silicides, along with a small number of carbides, collectively contributing to less than 1% of the total composition.

For the AlFeSi alloy, the phase composition at room temperature also includes three main phases: approximately 43% β, 43% θ, and 14% FCC aluminum. Similar to the AlFeSiMn alloy, impurity atoms in this system form aluminides, silicides, copper-containing intermetallics, and carbides, with these phases contributing to less than 1% of the total composition.

Table 1 - Alloys compositions

Alloy	Alloys compositions					
	Fe	***Si***	***Cu***	***Mn***	***Ni***	***Al***
AlFeSiMn	33.6	5.18	-	1.57	0.19	Ball
AlFeSi	29.5	3.72	0.02	0.18	0.02	Ball

The analysis of the microstructure of the synthesized samples revealed good agreement with the results of phase modeling. The AlFeSiMn alloy is characterized by the formation of two distinct types of α-phase: the αh-hexagonal intermetallic phase, which shows only minor dissolution of impurity atoms, including manganese, and the αc-cubic intermetallic phase, in which iron atoms are partially substituted by manganese atoms, resulting in a morphological change of the phase. At a temperature of approximately 700 °C, the total amount of the αc-phase does not exceed 20%, while the hexagonal modification accounts for about 60% of the volume fraction.

For the AlFeSi alloy, the αc-phase content is limited by the small amount of manganese and remains below 1%. However, at room temperature, when cooling occurs under non-isothermal conditions, small amounts of the α-phase may remain as residual quantities. As expected, both alloys contain the θ-phase intermetallic compound, primarily composed of Al and Fe. The θ-phase forms predominantly by separating from the melt at temperatures above the eutectic point, contributing to the basic structure of the alloy. As phase transformations occur, the quantity of the θ-phase decreases at around 500 °C, and for the AlFeSiMn alloy, this phase fully dissolves, only to reform at a temperature of approximately 450 °C (Fig.1.).

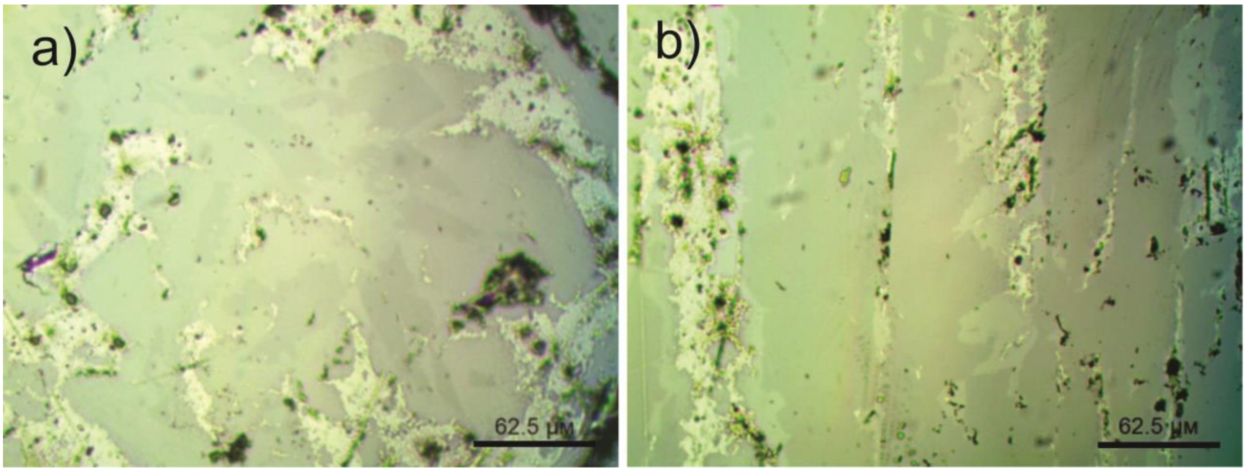

Figure 1 - Microstructure of AlFeSiMn (a) and AlFeSi (b) alloy, intermetallic region
This area contains intermetallic particles, which are β/θ/$θ_2$ phases (a)
or a combination of β/θ phases(b) (gray area).
The space between the particles is occupied by FCC (light area)

At room temperature, the microstructure of the Al-Fe-Si alloys with 1.5% and 0.18% manganese consists of intermetallic phases, primarily represented by compounds of the Al3Fe or Al13Fe4 type, or the θ-phase. These phases are present as large intermetallic particles with a lamellar (blocky) morphology, distributed throughout the entire area of the samples.

Upon cooling from the synthesis temperature, particles of the α-phase and other intermetallic compounds, formed with the involvement of impurity elements in the alloy, are precipitated on the existing θ-phase. As a result of interphase interactions within the liquid-solid solution, the formed particles continue to grow until the fcc aluminum is almost completely depleted of impurity atoms. The remaining liquid then crystallizes according to the typical behavior seen during the solidification of aluminum alloys with eutectic composition.

The θ and β phases present in the microstructure are not distinguishable through phase contrast; thus, it is more accurate to refer to the θ-phase as a θ/β complex (the grey phase in the micrographs). The color contrast observed within the θ/β phases does not correspond to phase contrast but rather to thickness contrast and therefore does not represent the boundaries between the individual phases. In the alloy with 0.18% Mn, the θ/β-phase displays a directional distribution across the entire sample, with a coarse, large-needle structure, having a length of over 200 µm and a width up to 60 µm.

In contrast, the alloy with 1.5% Mn exhibits a fragmented, coagulated, and rounded θ/β-phase structure with smaller dimensions (~100 µm in length and 30-60 µm in width). By comparing Fig 1a with Fig 1b, one can observe the transformation of the acicular structure of the θ-phase induced by the presence of manganese. Although complete suppression of β-phase formation under the influence of manganese is not observed, the disappearance of the θ-phase upon cooling in the temperature range of 550-450 °C leads to a fundamental change in the morphology of the intermetallic particles.

The light regions in the micrographs correspond to FCC aluminum, which aligns with the results of phase composition modelling. Despite the absence of FCC aluminum in the AlFeSi alloy model, its presence can be attributed to the nonequilibrium crystallization conditions; it is a product of phase transformations and represents a residual phase. A detailed examination of the FCC aluminum regions reveals a eutectic structure.

Within the FCC aluminum regions, intermetallic inclusions in both alloys are part of the eutectic. These inclusions have a nanodispersed dimension: in the alloy with 1.5% Mn, they range from 50 to 100 nm, while in the alloy with 0.18% Mn, they range from 100 to 500 nm.

The eutectic within the FCC region exhibits distinct morphological features. In the alloy with 1.5% Mn (Fig.2.), the eutectic precipitates display a fragmented, coagulated structure.

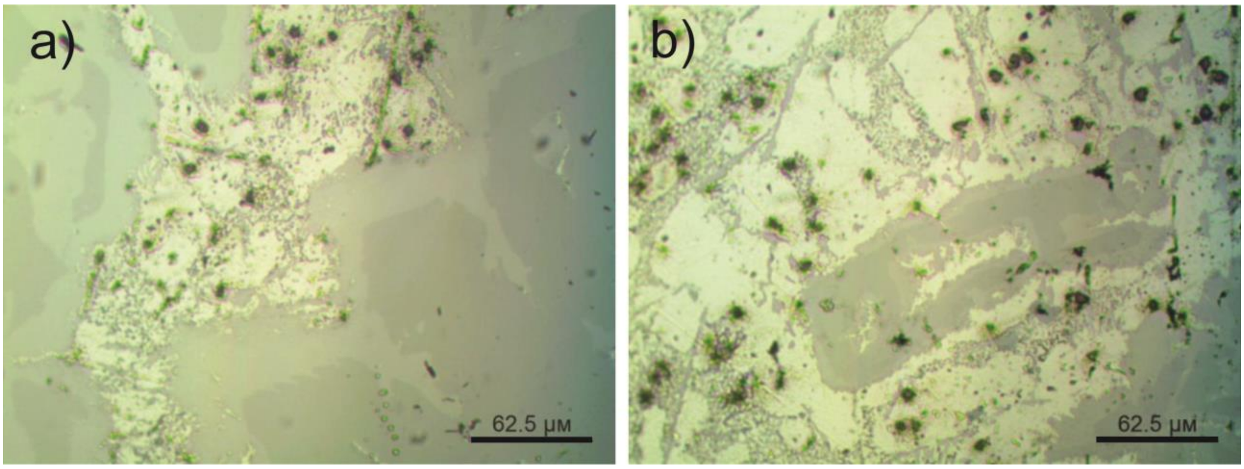

Figure 2 - Microstructure of AlFeSiMn (a) and AlFeSi (b) alloy, eutectic region
The space between the intermetallic particles is filled with an FCC phase containing a eutectic

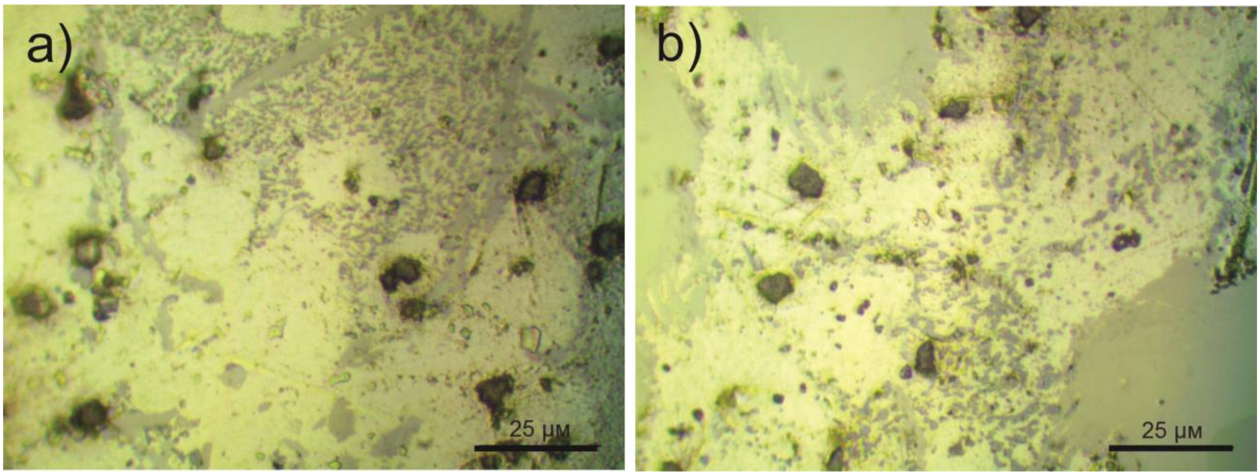

Figure 3 - Microstructure of AlFeSiMn (a) and AlFeSi (b) alloy, eutectic region (greater magnification)

In contrast, the eutectic in the AlFeSi alloy with 0.18% Mn exhibits a characteristic needle-like morphology. Upon cooling from the synthesis temperature, two phases – α and β – are precipitated simultaneously within the θ-phase that forms inside the eutectic. These phases constitute complex eutectic mixtures of solid solutions, including intermetallic phases with additional phases based on (Fe, Si) Al. In these phases, aluminum, iron, silicon, and small amounts of manganese are present. Manganese, in combination with other elements, is incorporated into the structure, stabilizing the phases and influencing their shape and distribution.

In the presented micrographs, the FCC+eutectic region (lighter in color) represents a mixture of FCC aluminum and intermetallic compounds, which are uniformly distributed between the phases, forming transition zones (Fig.3). The light areas correspond to a multiphase eutectic composed of FCC aluminum, β-phase (Al_5FeSi), Si, θ-phase ($Al_{13}Fe_4$), and a small amount of other intermetallic phases. However, the eutectic structure in the studied samples varies. In the sample with 0.18% Mn, the eutectic exhibits a lamellar structure, with plate lengths ranging from 20 to 50 µm and thicknesses of 2 to 5 µm. In the sample with 1.5% Mn, the eutectic particles take on a granular, rounded shape with diameters ranging from 1 to 5 µm, showing an uneven distribution. For greater clarity, the main results of the study are presented in Table 2.

Table 2 - Phase and structural composition

Alloy	Volume fraction calculated, %					Microstructure parameters			
	θ	*ϑ*	*ϑ₂*	*FCC*	*Other*	*Particle sizes θ/ϑ, μm*	*FCC particle sizes, μm*	*The nature of the arrangement of particles*	*Eutectic characteristics*
AlFeSiMn	~40	~38	~22	-	<1	100x30-60	60x20	coagulated	Round granular structure, particles 1-5 μm
AlFeSi	~43	~43	-	~14	<1	200x60	200x20	directed	Lamellar structure, plates 20-50x2-5 μm

This suggests that manganese suppresses the growth of the phase in anisotropic directions, where elongated (plate-like) forms typically occur, thereby inducing an inhibition or rounding effect. In the AlFeSiMn alloy, large β-phase needles and small areas of the residual α-phase are visible within the eutectic region. Intermetallic inclusions of various morphologies are also present. In contrast, the eutectic of the AlFeSi alloy is represented by coarser, less branched particles, with no large needle-like intermetallic phases observed. Rounded oxide particles are present in the FCC aluminum regions, while the intermetallic regions are free of oxides.

Conclusions

Based on the aforementioned studies, it can be confidently stated that the morphological structure of the alloy microstructure is highly dependent on the manganese content, particularly in alloys enriched with both silicon and iron. The addition of manganese results in a more rounded phase structure and a more dispersed eutectic.

It was revealed that the addition of 1.5% manganese changes not only the composition of intermetallic phases but also their morphology. The binding of iron, silicon and aluminum atoms into a cubic intermetallic α phase in the presence of manganese leads to the complete dissolution of the primary θ phase with its repeated formation near the lower boundary of the α phase existence. Such phase transformation promotes the transition from the growth of intermetallic particles on primary crystals of the θ phase with the formation of a directional microstructure to a coagulated microstructure through phase recrystallization. The addition of manganese promotes grinding of both intermetallic particles to a size of 100x30-60 µm and eutectic particles. While in the absence of manganese, coarse intermetallic particles of 200x60 µm in size are formed, formed on primary crystals of the θ phase. As a result of phase recrystallization, the alloy also contains fcc regions, but with a significantly smaller volume fraction, no more than 5%, located between intermetallic particles. As a result of the studies, a predominantly intermetallic alloy with a more favorable microstructure was obtained, which predicts high friction properties, and increased wear resistance with the possibility of use for parts that do not experience significant loads with an operating temperature range of 20-450° C.

Therefore, investigating the effect of manganese on an aluminum alloy produced by arc remelting holds significant scientific and practical value, particularly for further study of its mechanical properties.

Conflict of interest. On behalf of all authors, the corresponding author declares that there is no conflict of interest.

CRediT author statement. **V. Andreyachshenko**: Conceptualization, Methodology, Software. Visualization, Reviewing and Editing; **Y. Malashkevichute-Brillant**: Data curation, Writing draft preparation, Investigation.

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Балқитын электродпен доғалық қаптау арқылы синтезделген Al-Fe-Si қорытпа жүйесінің микроқұрылымына марганец қоспаларының әсері

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ТҮЙІНДЕМЕ

Әртүрлі қорытпаларды, соның ішінде алюминий негізіндегі металлкерамикалық материалдарды синтездеу үшін қолданылатын заманауи технологиялар, әсіресе олар туралы ғылыми деректердің жетіспеушілігінен әлі өнеркәсіпте кең қолданыс таппағандықтан, терең және кешенді зерттеуді талап етеді. Al-Fe-Si жүйесінің қорытпаларына ерекше қызығушылық құрамның қарапайымдылығына және қорытпаның негізгі компоненттерінің арақатынасына байланысты түзілетін фазалардың алуан түрлілігіне негізделген. Темір мен кремнийдің жоғары концентрациясымен Al-Fe-Si интерметалдық металлкерамикалық қорытпасы тұтынылатын электродпен доғалық қаптау әдісі арқылы синтезделді. Мақалада темір мен кремниймен, сондай-ақ марганец қоспаларымен бір мезгілде байытылған Al-Fe-Si қорытпасының металлографиялық талдауының тәжірибелік зерттеулері келтірілген. Белгілі бір қорытпа құрамындағы марганецтің әсерін зерттеу металаралық қосылыстардың морфологиясын, фазалық таралуын және жалпы құрылымдық тұрақтылығын терең бағалауға мүмкіндік берді. Фазалық құрамды алдын ала модельдеу синтезделген қорытпадағы фазаларды анықтауды жеңілдетті. Марганецтің аздап қосылуы микроқұрылымды тұрақтандыруға және коагуляцияланған интерметалдық бөлшектерді алуға мүмкіндік беретіні анықталды. Марганецтің фазалық өзгерістер мен құрылымға әсер ету механизмдерін тереңірек зерттеу жоғары жүктемелер мен температуралар жағдайларында кеңінен қолдануға арналған композицияларды оңтайландыруға мүмкіндік береді.

Түйін сөздер: Al-Fe-Si, микроқұрылым, ThermoCalc-ті бағдарламалық қамтамасыз ету, интерметалдық фазалар, балқитын электродпен доғалық қаптау.

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Влияние добавок марганца на микроструктуру сплава системы Al-Fe-Si, синтезированного дуговой наплавкой плавящимся электродом

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АННОТАЦИЯ

Современные технологии синтеза различных сплавов, включая металлокерамические материалы на основе алюминия, требуют детального и всестороннего исследования, особенно в области сплавов, которые еще не нашли широкого промышленного применения из-за недостаточности научных данных о них. Особое внимание привлекают сплавы системы Al-Fe-Si, благодаря простоте их состава и широкому разнообразию фаз, формируемых в зависимости от соотношения основных компонентов. Интерметаллидный металлокерамический сплав Al-Fe-Si с одновременно повышенным содержанием железа и кремния был синтезирован методом дуговой наплавки с плавящимся электродом. В статье представлены результаты экспериментальных исследований металлографического анализа сплава системы Al-Fe-Si, обогащенного железом и кремнием с добавками марганца.

Изучение влияния марганца на данный сплав позволило более детально оценить морфологию интерметаллидных соединений, распределение фаз и общую стабильность структуры. Предварительное моделирование фазового состава облегчило идентификацию фаз в синтезированном сплаве. Было установлено, что введение марганца в малых количествах способствует стабилизации микроструктуры и образованию коагулированных интерметаллидных частиц. Дальнейшее исследование механизмов воздействия марганца на фазовые превращения и структуру позволит оптимизировать составы сплавов для более широкого применения в условиях высоких нагрузок и температур.

Ключевые слова: Al-Fe-Si, микроструктура, программное обеспечение ThermoCalc, интерметаллидные фазы, дуговая наплавка плавящимся электродом.

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