



A novel upcycling strategy to Al alloys: Si removal via Mg-assisted suspension based fractional crystallization

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ARTICLE INFO

Keywords:

Upcycling
Al-Si hypoeutectic alloys
Mg addition
Si removal
Thermochemical
Microstructure
Suspension-based fractional crystallization

ABSTRACT

With the electrification and lightweighting of the automotive industry, Al-Si cast alloys scraps from end-of-life internal combustion engine vehicles is rapidly accumulating, severely disrupting the aluminum recycling cycle. This trend underscores the urgent need for advanced upcycling strategies that can remove Si with low energy and cost, thereby enabling the sustainable recycling of casting alloys. In this study, we proposed a Mg-assisted suspension based fractional crystallization method to achieve selective separation of Si from Al-Si alloys. Thermochemical modeling first guided alloy selection and process windows. A systematic experimental study evaluated the effects of Mg content and cooling rate on Si removal. The optimal process parameters are 20 wt% Mg addition with a cooling rate of 30 °C/h, achieving a Si removal degree of 63.0%. In addition, microstructural characterization demonstrated the separation mechanism. In Al-Si systems with high Mg content (e.g., 15 wt% Mg and 20 wt% Mg), polyhedral primary Mg₂Si precipitates as a single solid phase during the initial solidification stage and floats upward due to its lower density than the melt, thereby enabling selective Si separation. In contrast, eutectic Mg₂Si forms with α -Al, where it becomes entrapped within the α -Al dendritic network and thus cannot separate. Comprehensive assessment indicates that Mg can be recovered via vacuum distillation, thereby reducing net material costs. Additionally, this process has the potential to effectively control Si and Mg content while maintaining low energy consumption. Overall, this approach provides a technically feasible, economical, and environmentally friendly pathway for large-scale upcycling of Al-Si cast scraps

1. Introduction

As the second most produced and widely used metal after steel, global aluminum output reached 111 million tons in 2024 (73 million tons primary and 38 million tons secondary) and continues to grow [1,2]. Aluminum production is among the most energy- and greenhouse gas (GHG)-intensive industrial activities [3]. In 2023, the primary aluminum smelting consumed 4% of total global electricity [4,5]. The global aluminum industry emitted 1116 million tons of CO₂-equivalent, accounting for 2% of total GHG emissions [6,7]. In contrast, aluminum recycling typically requires only 5% of the energy of primary production and emits 0.5 t CO₂-equivalent per ton of recycled aluminum (3–5% of primary-route emissions) [8,9]. Therefore, expanding aluminum recycling is essential for improving both economic efficiency and environmental performance.

Aluminum is widely favored in automotive manufacturing for its high strength-to-weight ratio, corrosion resistance, and recyclability [10,11]. With the ongoing advancement of electrification and light-

weighting, the automotive industry's demand for wrought aluminum alloys, such as sheets and extrusions, is increasing, whereas the use of casting aluminum alloys in conventional internal combustion engine vehicles (ICEVs) components, such as engines and gearboxes, is declining [12–14]. ICEVs typically have a service life of 10–15 years [15,16]. As ICEVs reach end-of-life in the coming decades, a considerable surplus of Al-Si cast scraps (typically containing 7–10 wt% Si) is anticipated, which could amount to approximately 3.6 million tons by 2040, thereby significantly disrupting the balance of the aluminum [17]. Upcycling, which transforms low-grade aluminum cast scraps into high-quality wrought alloys or even pure aluminum, offers a promising strategy to rebalance the aluminum cycle and sustainably managing surplus casting aluminum scraps.

In casting aluminum alloy scraps, silicon is the most prevalent impurity, typically present at hypoeutectic levels ranging from 5 to 10 wt %. Due to its extremely low solubility in aluminum at room temperature, excess Si precipitates as hard and brittle eutectic Si particles or Si-containing intermetallic compounds, such as β -Al₅FeSi, α -Al₃Fe₂Si, and

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<https://doi.org/10.1016/j.seppur.2026.137458>

Received 14 November 2025; Received in revised form 19 February 2026; Accepted 1 March 2026

Available online 3 March 2026

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Mg₂Si [18,19]. These particles act as crack initiation sites, leading to embrittlement and significantly degrading mechanical performance, including reduced impact toughness [20], ductility [21–23], elongation [24], and fatigue resistance [25]. Even after modification and heat treatment, Al–Si alloys cannot match the combined ductility and toughness of wrought aluminum alloys. This inherent limitation poses a major obstacle to the upcycling of aluminum cast scraps in the automotive industry. For instance, wrought alloys such as automotive body sheet (ABS) materials impose strict impurity thresholds, with Si typically restricted to 0.2–1.0 wt% [26]. Therefore, developing efficient strategies to selectively remove silicon from Al–Si casting alloys is essential for Al upcycling.

Several approaches have been proposed to reduce Si or upgrade Al–Si cast scraps. Soluble anode electrolysis can produce aluminum with a purity comparable to primary aluminum [27–29]. However, the technique is always associated with high energy consumption, severe corrosion of electrodes and reactor materials, and environmental concerns. Lu et al. [17] proposed a solid-state electrolysis process for upcycling ADC12 casting alloy, achieving 99.9% purity aluminum with a recycling rate of 95.6%. However, the energy consumption remained significantly higher than that of remelting process (approximately 13 times) and challenges related to salt handling and process continuity persist [30]. Tamura et al. [31] imposed a constant electromagnetic force (EMF) in directional solidification, which upgraded the Al-7.4 wt % Si alloy to Al-3.6 wt% Si. However, the process requires external electric and magnetic field systems, leading to high equipment investment and complex maintenance, and the inherently slow directional solidification limits throughout. Moreover, for Al–Si alloys near the eutectic composition, this method becomes ineffective due to the limitations imposed by the Al–Si phase diagram and the corresponding crystal structure. Sun et al. [32] combined semi-solid isothermal treatment with centrifugal filtration separation to achieve an increase in aluminum content of the coarse AlSiCu alloy from 86.5% to 99.4%. Although promising, the narrow processing window and composition sensitivity hinder the scalability, and centrifugal separation typically entails high capital cost and strict process control.

Suspension-based fractional crystallization is a metallurgical purification and separation method in which crystals nucleate and grow freely suspended in the melt, rather than forming a solid layer of crystal at the interface [33]. Compared with layer-based fractional crystallization, this approach provides a much larger solid/liquid interfacial area and more efficient mass transfer, leading to higher purification efficiency, higher productivity, shorter operation cycles, and lower energy consumption [34]. It has been successfully applied in the purification of commercially pure aluminum, as demonstrated in industrial processes such as the Alcoa and Corus crystallizers [35]. However, its application to aluminum cast scraps with high Si contents remains challenging, primarily due to the complex thermodynamics and multi-phase competition of multi-component melts. In hypoeutectic Al–Si alloys, silicon predominantly precipitates with the α -Al phase in the form of eutectic Si phase, where Si forms an interconnected plate-like or fibrous morphology physically coupled to the α -Al matrix [36,37]. Consequently, this Si phase does not exist as discrete, free crystals but as eutectic aggregates co-grown with α -Al, which largely eliminates the separation window and prevents the conventional suspension crystallization method from effectively separating silicon. To address impurity-related challenges, metal addition strategies, such as addition with Mn or Cr, have been widely adopted in aluminum scraps to remove detrimental elements like Fe through formation of intermetallic compounds [38–40]. Studies on Al-Si-Mg systems have shown that magnesium, as a common alloying element in Al alloys, promotes the formation of Mg₂Si intermetallic compounds during solidification [41–43], which suggests a feasible pathway to capture Si into a distinct intermetallic phase and enable separation. Nevertheless, there is currently a lack of systematic research on the removal of Si from aluminum alloys through the addition strategy.

This study proposes a novel upcycling approach for the selective separation of Si from multicomponent Al–Si melts. The experimental design was informed by thermochemical modeling, enabling effective Si removal through the optimization of key process parameters. Systematic microstructural characterization combined with theoretical analysis was conducted to elucidate the phase separation and flotation-driven purification mechanism. The energy consumption of the method was evaluated to assess its feasibility for future industrial-scale applications. This work provides both theoretical insight and parametric foundations for the development of scalable upcycling strategies for resource recovery of aluminum scraps.

2. Materials and methods

2.1. Thermochemical modeling

In this study, basic FTLite database [44] from the commercial software FactSage 8.3 were employed using the Equilib module to perform thermochemical calculations. This model was developed to predict the possible formed solidified phases in each sample, as well as the solidification conditions, i.e., the temperature-dependent evolution of equilibrium and non-equilibrium phases in Al-Si-Mg melts with varying Mg addition contents. These were calculated using equilibrium cooling and Scheil-Gulliver cooling, respectively. Thermochemical calculations were further utilized to guide the design of experiments involving Al-9%Si-x% Mg systems with different Mg contents, and to provide theoretical support for interpreting the resulting phase transformation behaviors. It should be pointed out that the stable intermetallic phases in Al-Si-Mg samples are labeled as FCC-Al₁, aC1 and Beta in FactSage 8.3, corresponding to α -Al, cubic Mg₂Si, β -Al₃Mg₂, respectively.

2.2. Materials preparation

Based on thermochemical modeling, four alloy compositions were developed to investigate the effect of Mg content on phase formation and evolution during solidification process, i.e., Al-9%Si-0%Mg, Al-9%Si-5%Mg, Al-9%Si-7%Mg, Al-9%Si-10%Mg, Al-9%Si-15%Mg, and Al-9%Si-20%Mg. The raw materials used in the experiments were commercial pure Al blocks (99.7%), high purity Mg blocks (99.97%) and Al-20%Si master alloy blocks (all expressed in wt% hereafter, unless otherwise specified). For alloy samples preparation with different contents of Mg, commercial pure aluminum and Al-20%Si master alloy were melted with the necessary amount of pure magnesium in each case in a clay-graphite crucible inside an inductive-heated furnace at 750 °C and held for 30 min to assure the complete mixing and homogeneity of the alloy. The clay-graphite crucible consists primarily of 40–47 wt% graphite, 10–18 wt% SiC, 10–18 wt% Al₂O₃, and 20–30 wt% SiO₂. The inner wall was coated with BN as a non-wetting barrier to reduce direct melt-crucible contact and suppress interfacial reactions. To avoid the oxidation and evaporation, the melt was covered with commercial Mg-salt flux (Flux 5) under purging argon gas. Flux 5 is a fused chloride-fluoride salt mixture near the MgCl₂-KCl-NaCl eutectic, consisting of approximately 46% MgCl₂, 23% KCl, 26% NaCl, and 5% CaF₂. After removing the salt from the surface, the Al-Si-Mg melt was cast into a rectangular steel mold to obtain the target alloy with uniform composition. The chemical compositions of the mixed alloys are shown in Table 1.

2.3. Experimental procedures

To investigate the effect of the Mg addition and cooling rate on phase formation and the separation behavior of the impurity element Si during solidification, a suspension-based fractional crystallization furnace was employed (see Fig. 1 left). Although structurally similar to a conventional resistance furnace, it is termed “suspension-based fractional crystallization” according to its solidification mode, where partial

Table 1

Chemical compositions of Al–Si alloys with different Mg contents used in experiments (wt%).

Alloy	Element, wt%				
	Si	Mg	Fe*	Σ (Cu, Mn, Cr, Ni, Ti)	Al
#1 Al-9%Si-0%Mg	9.04	<0.01	0.22	<0.02	Bal.
#2 Al-9%Si-5%Mg	9.10	5.33	0.21	<0.02	Bal.
#3 Al-9%Si-15%Mg	9.07	14.25	0.17	<0.02	Bal.
#4 Al-9%Si-20%Mg	9.31	19.58	0.18	<0.02	Bal.
#5 Al-9%Si-7%Mg	9.14	7.08	0.18	<0.02	Bal.
#6 Al-9%Si-10%Mg	9.11	10.51	0.17	<0.02	Bal.

* The detected Fe originates from the raw materials, with ~0.15% Fe in commercially pure Al and ~0.25% Fe in the Al-20% Si master alloy.

crystals nucleate and grow while suspended in the melt. The furnace uses resistance heating through a cylindrical graphite element, with a ceramic fiber insulation layer providing excellent thermal stability. The high thermal conductivity and heat capacity of the graphite chamber allow prolonged heat retention and slow, uniform cooling of the melt, enabling the free suspension and growth of crystalline particles characteristic of a suspension-based fractional crystallization process. An approximately 400 g of each Al-Si-Mg sample was charged into a small graphite crucible ($\phi 5.5 \times H12.0$ cm) and placed in the middle of the furnace. The pre-alloyed samples were heated up to 30–50 °C above respective liquidus temperature and held for 0.5 h to ensure complete and homogeneous remelting of the alloys. The use of graphite crucible and graphite heating elements as well as an excellent heat-sealed system in this furnace guaranteed uniform distribution within the melt and providing very slow cooling rates. The melt was protected by a continuous flow of high-purity argon gas. The temperature of the sample was permanently monitored and regulated in real time by a programmable temperature controller with a type K thermocouple located inside the melt. The detailed experimental parameters including initial melt temperature and cooling rate are provided in Table 2, where 30, 60, and 120 °C/h were chosen as slow, medium, and fast controlled-cooling rates to study their effect on primary Mg_2Si growth and flotation separation.

2.4. Analysis and characterization

After horizontal sectioning, the top, middle, and bottom regions of each sample were analyzed, as illustrated in Fig. 1 (right). The chemical composition of the experimental samples was determined by SPECTRO MAXx optical emission spectrometer (SPECTRO Analytical Instruments, Germany). Phase composition was conducted via X-ray diffractometer

(XRD, Bruker AXS D8 Advance). The microstructure and distribution of phases were analyzed using scanning electron microscopy coupled with energy-dispersive X-ray spectroscopy (SEM-EDS, FEI Helios NanoLab G3 CX).

3. Results and discussion

3.1. Thermochemical calculations to estimate the effect of Mg content on the Si removal efficiency

There are three key criteria that must be satisfied in order to gain an effective silicon removal by addition of the element X: 1. This additive should readily react with Si to form a stable Si-rich intermetallic phase with a sufficiently high mass fraction to promote the removal of Si; 2. the Si-rich intermetallic compound phase should precipitate as a single phase at the earliest possible stage of solidification process. If a substantial number of α -Al dendrites or other intermetallic compounds form prior to or simultaneously with the target intermetallic phases, there is a high risk that the target intermetallic phases will be entrapped within these solid phases, thereby reducing the separation efficiency and even completely preventing it; 3. as this study employed a simple suspension-based fractional crystallization method to separate Si, the density of the target intermetallic compounds should differ sufficiently from that of molten aluminum to support the separation process additionally by gravity settling or buoyant flotation.

As shown in Fig. 2(a) and Fig. 3(a), in the Al-9% Si system without Mg addition, α -Al is the prior phase to begin precipitating from the liquidus at 604.0 °C and continues down to the eutectic temperature of 578.8 °C. The following eutectic reaction occurs isothermally at 578.81 °C, resulting in the simultaneous precipitation of eutectic Si and α -Al until completely solidification. Therefore, thermochemical calculations theoretically indicate that Si cannot be separated from Al-9%Si hypoeutectic system through a simple solidification process. In Al-9%Si-5%Mg system (Fig. 2(b) and Fig. 3(b)), α -Al remains the primary phase, precipitating first from the liquidus at 587.7 °C during the second

Table 2

Experimental parameters of initial melt temperature range and cooling rate.

Alloys	Initial melt temperature, °C	Cooling rate, °C/h
#1 Al-9%Si	630	30, 60, 150
#2 Al-9%Si-5%Mg	630	30, 60, 150
#3 Al-9%Si-15%Mg	730	30, 60, 150
#4 Al-9%Si-20%Mg	750	30, 60, 150
#5 Al-9%Si-7%Mg	630	60
#6 Al-9%Si-10%Mg	660	60

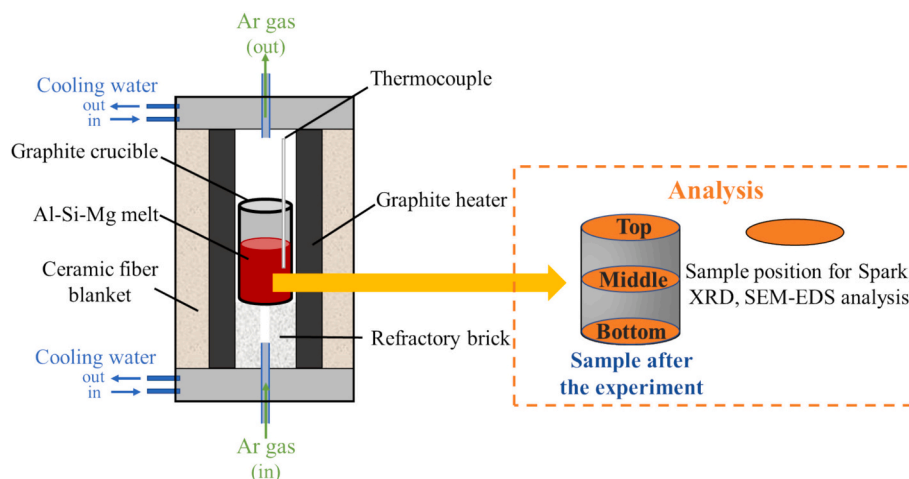


Fig. 1. (a) Schematic diagram of suspension-based fractional crystallization furnace (left) and sample positions for analysis (right).

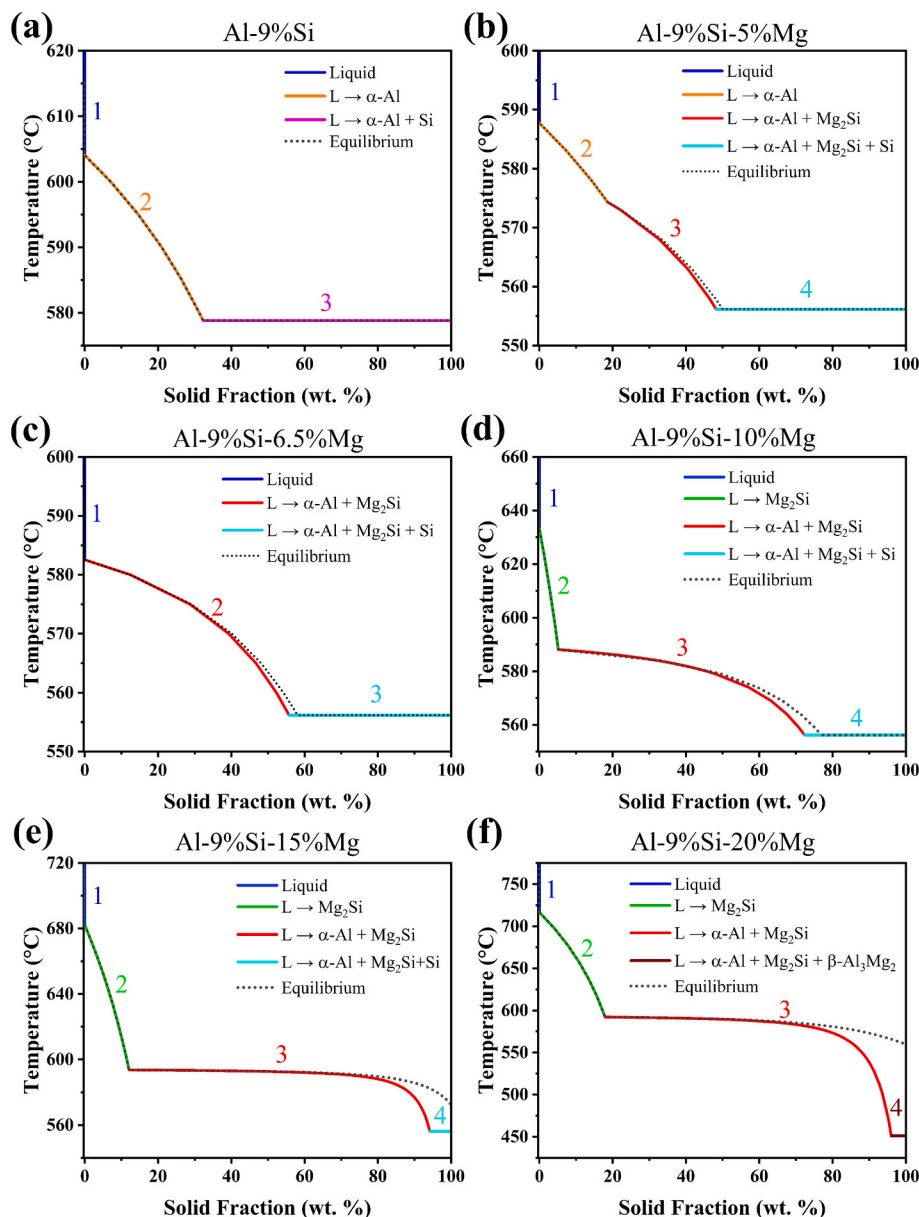


Fig. 2. Non-equilibrium (Scheil-Gulliver model) and equilibrium solidification curves of (a) Al-9%Si, (b) Al-9%Si-5%Mg, (c) Al-9%Si-6.5%Mg, (d) Al-9%Si-10%Mg, (e) Al-9%Si-15%Mg, (f) Al-9%Si-20%Mg, predicted by Factsage 8.3.

cooling stage (587.7–574.3 °C), corresponding to the initial solidification stage. In the third cooling stage (574.3–556.2 °C), α -Al and Mg_2Si form simultaneously as a binary eutectic constituent. In the fourth cooling stage, at the isothermal temperature of 556.16 °C, a ternary eutectic constituent (α -Al + Mg_2Si + Si) precipitates. As shown in Fig. 2 (a-c) and 3(a-c), when the Mg content in the Al-9% Si-x% Mg system increases from 0 to 6.5%, α -Al remains the primary phase, however, the phase fraction of α -Al formed as the initial single phase decreases progressively from 32.3% to 0%, while the phase fraction of Mg_2Si phase in the solidified alloy increases from 0% to 9.2%. Although a considerable amount of Si-rich intermetallic phase (Mg_2Si) forms during solidification of the Al-9%Si-x%Mg system ($0 < x < 6.5$), these Mg_2Si precipitate simultaneously as eutectic phase with α -Al or with α -Al and Si during the second solidification stage after α -Al is already formed as the primary solid phase. This is contrary to the second separation criteria mentioned above, as these secondary eutectic Mg_2Si is easily encapsulated within the α -Al matrix, making it difficult to be separated effectively.

The addition of 6.5% Mg marks a critical turning point in the

solidification phase transformation of Al-9%Si-x%Mg system. When the Mg content exceeds this threshold, as shown in Fig. 2(d, e) and Fig. 3(d, e), Mg_2Si phase becomes the primary phase, which precipitates as the single phase during the initial solidification stage. Fig. 3(c-e) further indicate that the primary Mg_2Si formed during this stage increases from 0% to 5.2% and 12.1% as the Mg content rises from 6.5% to 10% and 15%, respectively. The formation of these primary Mg_2Si intermetallic compounds fully satisfies the three selection criteria outlined above. Since the density of Mg_2Si (1.99 g/cm³ [45]) is significantly lower than that of the aluminum melt (2.20–2.50 g/cm³, depending on temperature and composition [46]), these primary Mg_2Si particles can easily float to the melt surface to achieve an effective separation. When the Mg content exceeds 17.05%, essentially all Si impurity in the system, except for a trace amount dissolved in α -Al (the solubility of Si in aluminum is 0.05 at.% at 300 °C [47]), combines with Mg to precipitate as Mg_2Si phase. As shown in Fig. 2(f) and 3(f) for the Al-9%Si-20%Mg system, Mg_2Si phase precipitates as primary phase during the second cooling stage (716.63 to 592.16 °C), accounting for 17.9% of the Al-Si-Mg system. In the fourth

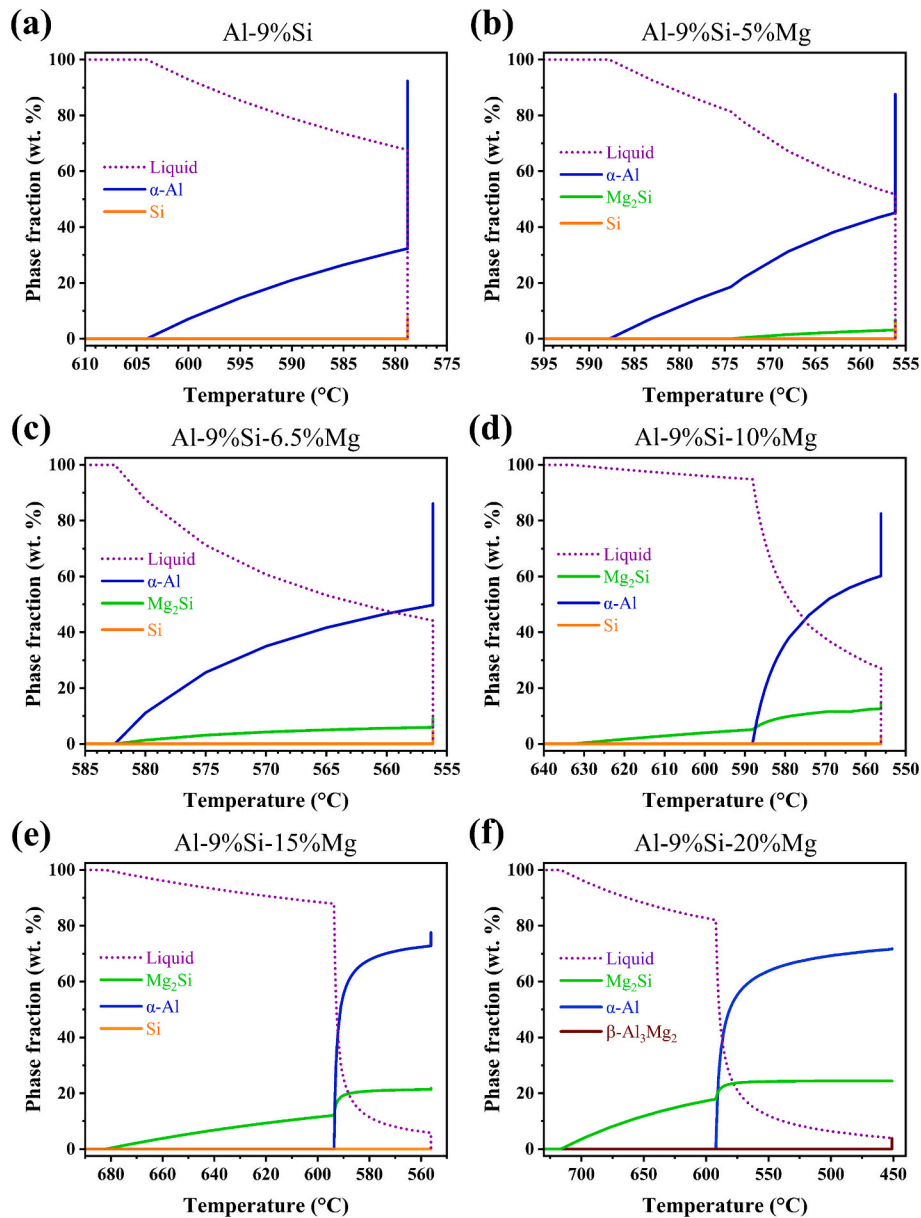


Fig. 3. Non-equilibrium (Scheil-Gulliver model) solidified phase fraction (wt%) for aluminum alloys with different Mg content (a) Al-9%Si, (b) Al-9%Si-5%Mg, (c) Al-9%Si-6.5%Mg, (d) Al-9%Si-10%Mg, (e) Al-9%Si-15%Mg, (f) Al-9%Si-20%Mg, predicted by FactSage 8.3.

stage at the isothermal temperature of 451.0 °C, the excess Mg combines with Al to form the β - Al_3Mg_2 phase (3.8% of the total mass), which forms together with α -Al and secondary Mg_2Si phases.

Based on these solidification predictions derived from FactSage, the theoretical Si removal degree ($\eta_{T,\text{Si}}$) can be estimated under the assumption that only the single primary Mg_2Si phase formed during the first solidification stage is completely removed by flotation, while the Si and Mg remaining in the residual liquid are considered unremovable impurities. The theoretical Si removal degree (%) is calculated as:

$$\eta_{T,\text{Si}} = \frac{C_{\text{Si},i} - C_{\text{Si},\text{res}}}{C_{\text{Si},i}} \times 100\% \quad (1)$$

where $C_{\text{Si},i}$ (wt%) is the initial Si content in the system, and $C_{\text{Si},\text{res}}$ (wt%) is the residual Si content in the liquid phase after the primary Mg_2Si solidification stage.

As shown in Fig. 4, the residual Si content in the liquid phase after the primary Mg_2Si solidification stage decreases markedly with increasing Mg addition content, from 9.0% at 6.5% Mg to 1.33% at 25%

Mg, corresponding to an increase in the theoretical Si removal degree from 0% to 88.4%. In contrast, the residual Mg content in the liquid phase after the primary Mg_2Si solidification stage exhibits rise over the same Mg range from 6.5% at 6.5% Mg to 14.35% at 25% Mg. The above thermochemical calculations confirm that the addition of Mg is an effective strategy for removing Si from Al–Si casting alloys. However, these results emphasize the need to balance the improvement in Si removal degree with the potential accumulation of residual Mg in practical processing.

3.2. Experimental investigations on the effect of Mg content on the Si removal efficiency

3.2.1. The effect on macrostructure, phase formation, and chemical composition

Fig. 5(a–f) show the macrostructure of longitudinal cross sections of Al–Si alloy samples with different Mg contents at a cooling rate of 60 K/h, while the corresponding XRD patterns for the sampling positions are

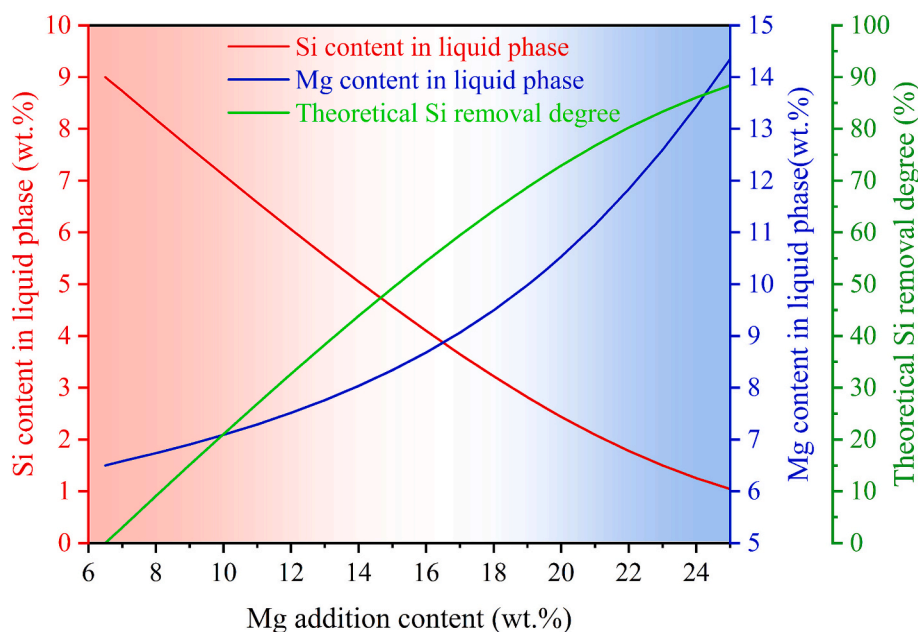


Fig. 4. Theoretical Si removal degree and residual Mg and Si contents in the liquid phase as a function of Mg addition content, calculated by FactSage thermochemical simulations.

illustrated in Fig. 5(g). The gas porosities and shrinkage cavities in the solidified samples are due to the conducting experiments under very slow solidification rate as well as lack of vacuum conditions.

The longitudinal section of the Al-9%Si-0%Mg sample shows a macroscopically uniform ingot with coarse α -Al dendrites distributed throughout the cross-section and no stratification. The XRD pattern proves that the bottom of the Al-9%Si-0%Mg sample mainly consists of α -Al and Si phases. In the Al-9%Si-5%Mg sample; though and despite the absence of any obvious stratification the pattern of the bottom region shows diffraction peaks of the latter two phases in accompanying with Mg_2Si . To verify the thermodynamic prediction that primary Mg_2Si precipitation and thus Si separation starts once the Mg addition exceeds the critical level of 6.5%, the experiments on Al-9%Si-7%Mg at a cooling rate of 60 K/h were performed. The result (Fig. 5(c)) shows that when the Mg content slightly exceeds the predicted separation threshold to 7%, a thin Mg_2Si -rich layer begins to form at the top surface, indicating the onset of separation in agreement with the FactSage calculations. With further increasing Mg to 10%, a markedly thicker and more continuous intermetallic-rich layer is observed (Fig. 5(d)), confirming that the separation effect becomes progressively stronger as the amount of primary Mg_2Si increases.

When the Mg addition is increased to 15%, as seen in Fig. 5(e), abundant dark intermetallic particles cluster near the top surface and along the sidewalls, evidencing distinctive gravity-driven macro-segregation. XRD patterns verify that these particles are Mg_2Si and the top region exhibits much stronger Mg_2Si diffraction peaks than the middle region, consistent with flotation and accumulation of the low-density Mg_2Si phase in the upper part of the ingot. At these positions the Si reflections are not detectable, indicating that Si was largely consumed to form Mg_2Si below the XRD detection limit. Furthermore, the crucible wall acted as a preferential site for heterogeneous nucleation, facilitating the formation of Mg_2Si particles. Once nucleated, these particles tended to adhere to the crucible wall and grow, thereby hindering their flotation. Therefore, localized accumulation of Mg_2Si particles were also observed in the vicinity of the crucible wall. When the Mg content in the sample further increases to 20%, as in Fig. 5 (f), the upper region of the ingot exhibits a markedly higher areal fraction of Mg_2Si . Part of the Mg_2Si particles coarsen and develop into larger dendritic morphologies, in contrast to the mainly polyhedral Mg_2Si observed in Al-9%Si-15%Mg

sample. In the Al-9%Si-15%Mg and Al-9%Si-20%Mg samples, a clear stratification was observed, characterized by a distinct Mg_2Si accumulation layer at the top region of both samples. As shown in Fig. 2(f) and Fig. 3(f), thermochemical calculations with Scheil-Gulliver cooling model for Al-9%Si-20%Mg predict the formation of 3.17 wt% β - Al_3Mg_2 at the end of solidification, suggesting that β - Al_3Mg_2 phase could precipitate in the middle and bottom position of sample. However, XRD pattern of the middle region of Al-9%Si-20%Mg sample shows no resolvable β - Al_3Mg_2 diffraction peak. This is primarily attributed to (i) the very low local fraction of this phase, which is likely below the detection limit of laboratory XRD equipment, and (ii) the back-diffusion of Mg [48], as a high-diffusivity element, into the solid during the terminal stage of solidification enriches Mg in α -Al and expands the lattice parameters of α -Al, resulting in a slight shift of the α -Al peaks at 78.227° and 82.435° toward lower 2θ values, as indicated by the dashed line in Fig. 5(g).

Fig. 5 (h) and 5 (i) illustrates the variation of Si and Mg contents at the top, middle, and bottom regions of Al-9%Si-x%Mg samples with different contents of Mg additions. At 0% Mg and a cooling rate of 60 K/h, the Si contents at the top, middle, and bottom positions of the Al-9%Si sample remain nearly identical, as shown by the black line in Fig. 5(h), indicating no Si segregation. This is consistent with previous thermochemical predictions that α -Al and Si co-precipitate and cannot be separated, as also reflected in the uniform longitudinal cross-section shown in Fig. 5(a). Similarly, with 5% Mg additions at 60 K/h, both Si and Mg contents remained uniform across the ingot height, indicating the absence of stratification of Si-rich and Si-lean.

In the Al-9%Si-7%Mg sample, the top region exhibited a pronounced enrichment of both Si and Mg, whereas the middle and bottom regions showed only slight decreases relative to the initial composition. The Si removal degree in the middle and bottom regions were only 2.0% and 2.3%, respectively. This limited refinement is attributed to the fact that 7% Mg produces only a small fraction of primary Mg_2Si , which preferentially agglomerates and accumulates at the top to form a thin and compact Mg_2Si -rich layer. As a result, strong compositional enrichment is observed locally at the top, while the removal of Si in the remaining regions is modest. With further increasing Mg to 10%, the separation effect becomes stronger. The Si contents in the middle and bottom regions decreased to 7.74% and 7.60%, corresponding to Si removal

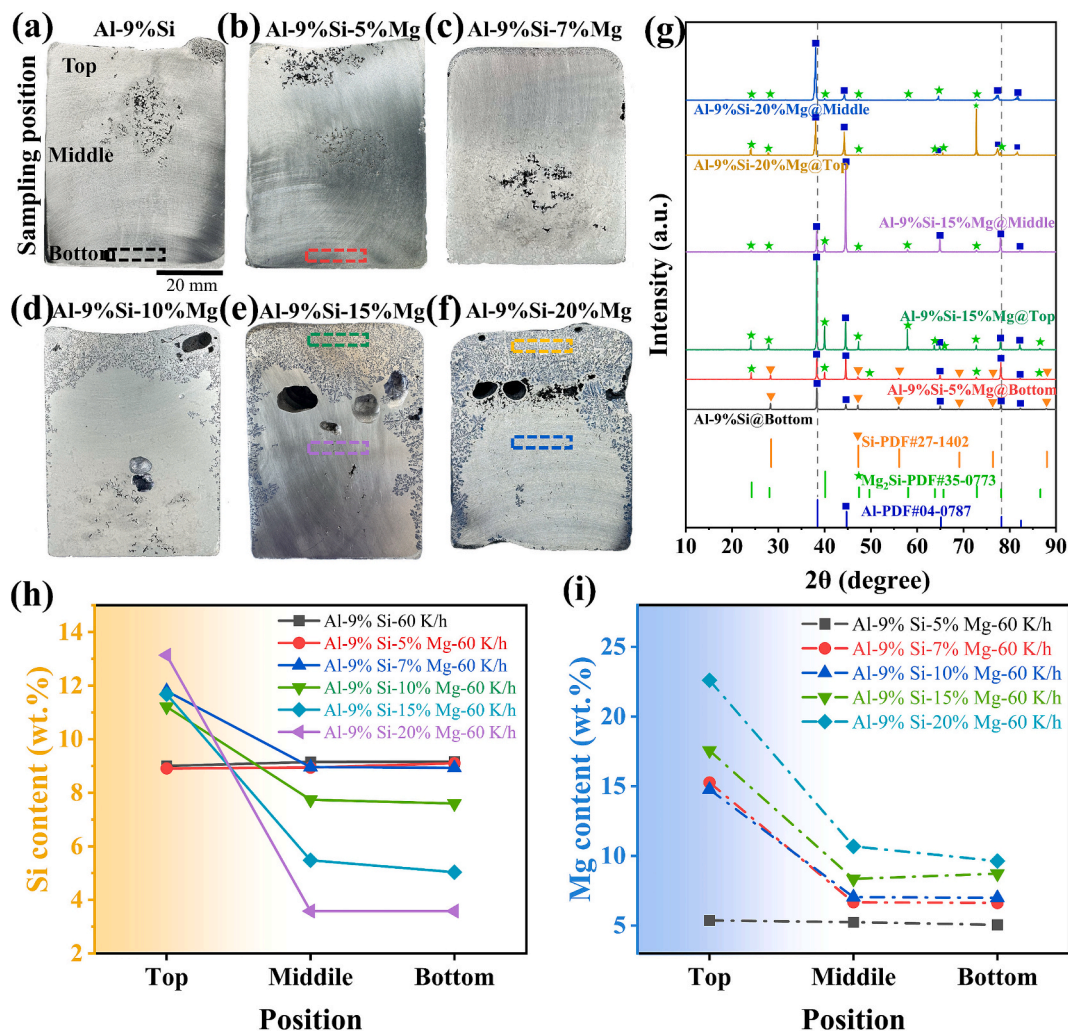


Fig. 5. Macrostructure of longitudinal sections of samples with different Mg contents at a cooling rate of 60 K/h: (a) Al-9%Si, (b) Al-9%Si-5%Mg, (c) Al-9%Si-7%Mg, (d) Al-9%Si-10%Mg, (e) Al-9%Si-15%Mg, (f) Al-9%Si-20%Mg, (g) corresponding XRD patterns, (h) Si content, and (i) Mg content at different sample positions as a function of Mg addition.

degree of 15.0% and 16.6%, respectively. This is consistent with enhanced formation and separation of primary Mg_2Si and the associated redistribution of Mg and Si. Meanwhile, the Mg contents in the middle and bottom regions increased to 7.04% and 6.99%.

When the Mg addition is increased to 15%, clear stratification is observed: Si and Mg are significantly enriched in the top region, while the middle and bottom regions exhibit much lower and nearly identical concentrations. The top region of the Al-9%Si-15%Mg sample with a cooling rate of 60 K/h contains 12.32% Si and 19.15% Mg due to the accumulation of Mg_2Si -rich phases. In comparison, the Si content in the middle and bottom decrease to 5.48% and 5.05%, respectively, which is close to the calculated Si concentration of the residual liquid (5.20%, in Fig. 4). Correspondingly, the Si removal degrees at the middle and bottom reach 39.6% and 44.5%, respectively, demonstrating effective Si separation under 15%Mg additions. When the Mg addition is further increased to 20%, the Si concentration at the top position becomes higher than that of the Al-9%Si-15%Mg sample. This corresponds with the coarser and more densely packed Mg_2Si particles observed at the top region in Fig. 5 (f), whereas the Si content in the middle and bottom regions is markedly lower. At a cooling rate of 60 K/h, the Si content in the top region of the Al-9%Si-20%Mg sample reaches 13.14%, while the middle and bottom both decrease to 3.58%. The corresponding Si removal degree in the middle and bottom region thus reaches 61.5%. Although Si elimination is more effective in these regions, the Mg

content is simultaneously higher with increasing Mg addition. Specifically, the Mg contents in the Al-9%Si-20%Mg sample are 22.6% at the top, 10.68% at the middle, and 9.63% at the bottom, all of which are higher than those in the Al-9%Si-15%Mg sample.

3.2.2. Microstructural characterization

The SEM images with EDS elemental point and mapping analyses of Al-9%Si-x%Mg ($x = 0, 5, 15, 20$) samples at a cooling rate of 60 K/h is shown in Fig. 6. As shown in Fig. 6(a), in the Al-9%Si-0%Mg, numerous eutectic Si flakes were distributed throughout the Al matrix. Due to the presence of 0.22 wt% Fe impurity in the sample, bright and fine needle-like $\beta\text{-Al}_5\text{FeSi}$ phases were observed. When the Mg content reaches 5% (Fig. 6(b)), the eutectic Mg_2Si phase with the dark Chinese-script-like (skeleton-like) morphology appeared abundantly within the $\alpha\text{-Al}$ matrix, forming an irregular network structure. Since these Mg_2Si precipitates simultaneously with $\alpha\text{-Al}$ through eutectic reactions, they are difficult to separate by flotation during solidification. Meanwhile, with the addition of Mg, part of the $\beta\text{-Al}_5\text{FeSi}$ phase transforms into polyhedral $\pi\text{-Al}_8\text{FeMg}_3\text{Si}_6$, as revealed by EDS analyses.

In the SEM-EDS image of the top region of the Al-9%Si-15%Mg sample (Fig. 6(c)), numerous black particles with dendritic or polyhedral morphologies were clearly observed, with sizes ranging from 250 to 500 μm . EDS analysis confirmed these coarse particles to be primary Mg_2Si , consistent with the characteristic morphology of primary Mg_2Si

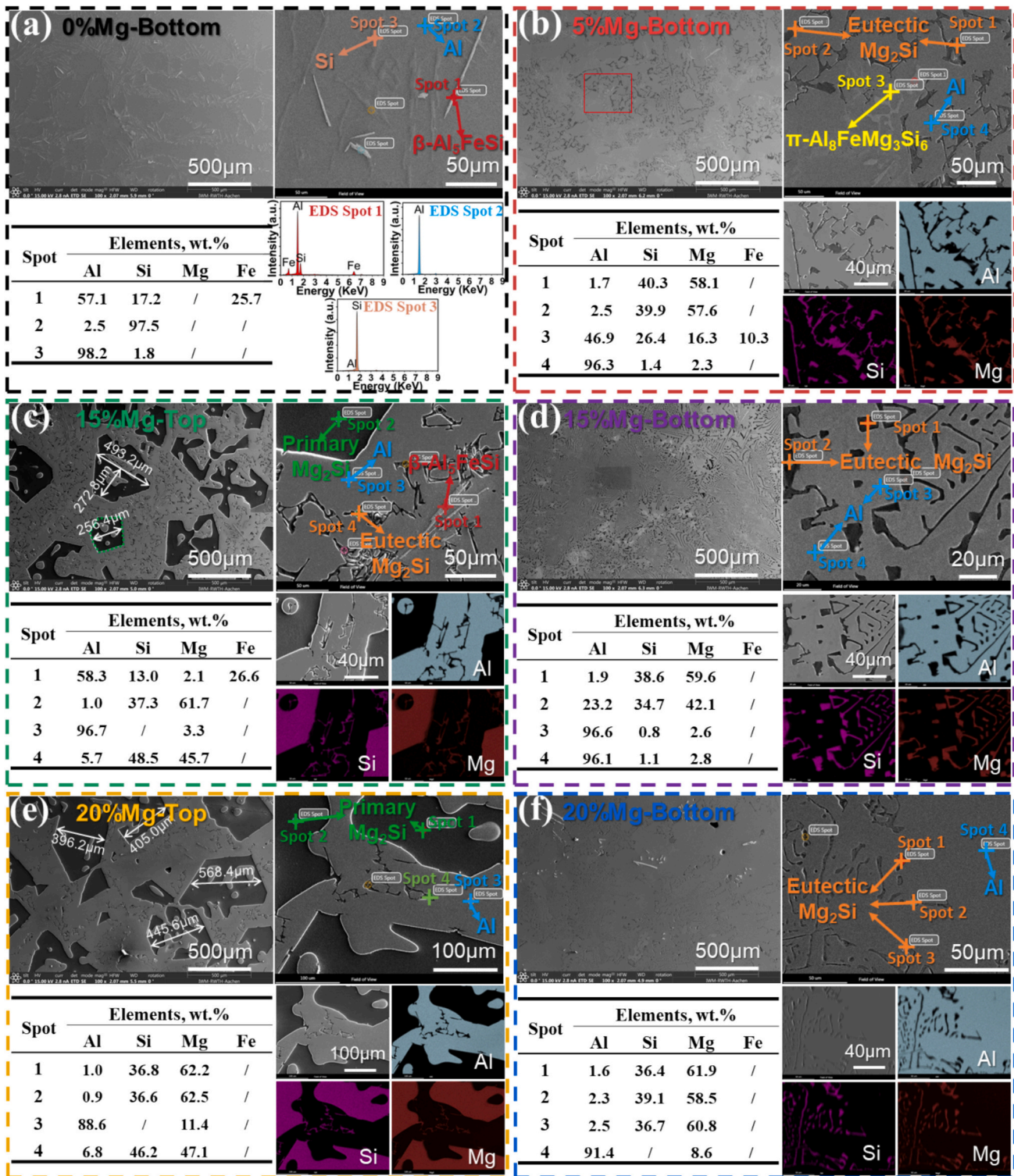


Fig. 6. SEM images with EDS elemental point and mapping analyses of Al-9%Si-x%Mg samples with varying Mg additions at a cooling rate of 60 K/h: (a) Al-9%Si-0% Mg-bottom position, (b) Al-9%Si-5%Mg-bottom position, (c) Al-9%Si-15%Mg-top position, (d) Al-9%Si-15%Mg-bottom position, (e) Al-9%Si-20%Mg-top position, (f) Al-9%Si-20%Mg-bottom position.

reported in the literature [49,50]. These primary Mg₂Si particles float upwards during the early stage of solidification and accumulate in the top layer of the sample due to their low density. In addition, numerous fine and Chinese-script-like eutectic Mg₂Si particles were dispersed within the gray α -Al matrix, which are much smaller than the primary Mg₂Si and consistent with the morphology of eutectic Mg₂Si shown in Fig. 6(b). Consequently, the top region of this sample exhibited the

typical structural feature of primary Mg₂Si enrichment accompanied by dispersed eutectic Mg₂Si in Al matrix. The bottom region of Al-9%Si-15%Mg sample, as shown in Fig. 6(d), contained no primary Mg₂Si but instead exhibited a uniform distribution of fine, skeleton-like eutectic Mg₂Si within the α -Al matrix. These observations indicate that primary Mg₂Si segregates to the top due to flotation, while secondary eutectic Mg₂Si remains embedded in the α -Al matrix, as it precipitates

concurrently with α -Al during solidification.

In the top region of the Al-9%Si-20%Mg sample (Fig. 6(e)), numerous coarse primary Mg_2Si particles with sizes of 400 to 600 μm were enriched, which are larger than those in the top position of Al-9%Si-15%Mg sample, while the fraction of eutectic Mg_2Si within the α -Al matrix is markedly reduced. In the bottom region of the Al-9%Si-20%Mg sample, as shown in Fig. 6(f), consisted mainly of fine, skeleton-like eutectic Mg_2Si phases uniformly distributed within the α -Al matrix, similar to the bottom position of Al-9%Si-15%Mg sample but with a lower eutectic Mg_2Si fraction. These features are consistent with thermochemical calculations from FactSage, indicating that increasing the Mg content from 15% to 20% promoted the formation of primary Mg_2Si while suppressing secondary eutectic Mg_2Si . Furthermore, EDS analysis of Spot 4 in Fig. 6(f) revealed that the Al matrix contains 8.6 wt% Mg, which explains the leftward shift of the Al diffraction peaks observed in the XRD patterns (Fig. 5(g)) with increasing Mg content, as lattice expansion occurs due to Mg incorporation.

3.3. Effect of cooling rate on the Si removal efficiency

3.3.1. Chemical composition

Fig. 7(a–c) presents the Mg and Si contents at the top, middle, and bottom region of Al-9%Si- x %Mg systems ($x = 5, 15, 20$) samples solidified at cooling rates of 30, 60, and 150 K/h. For the alloy with 5% Mg (Fig. 7(a)), the Si and Mg contents remained essentially uniform across the ingot height regardless of cooling rate, indicating that Si cannot be separated under these conditions due to thermochemical constraints. When the Mg addition increases to 15% (Fig. 7(b)), the Si and Mg concentrations in the top region of sample increased as the cooling rate decreased, whereas the middle and bottom regions were basically unaffected. The Si and Mg concentrations in the top region of the sample with a cooling rate of 30 K/h reached 12.32% and 17.56%, respectively, compared with 11.20% and 17.25% at 150 K/h. As the Mg addition was

increased to 20% (Fig. 7(c)), the segregation of Mg and Si in the top region was strongly dependent on the melt cooling rate, i.e., reducing the cooling rate significantly enhanced enrichment of Si and Mg at the top position of sample. The primary Mg_2Si accumulated more densely in the top region of the sample with a lower cooling rate of 30 K/h, where the Si and Mg contents reached 14.70% and 28.14%, respectively. In contrast, within the range of cooling rates investigated in this study, the Si and Mg contents in middle and bottom regions of sample were relatively insensitive to the cooling rate.

Since the Si and Mg contents in the middle and bottom region of the samples were nearly identical, the Si content used for calculating the removal degree was taken as the average of the Si contents in these two regions. The real Si removal degree (%) of the sample was then calculated as:

$$\eta_{Si} = \frac{C_{Si,i} - (C_{Si,M} + C_{Si,B})/2}{C_{Si,i}} \times 100\% \quad (2)$$

where $C_{Si,i}$ (wt%) is the initial Si content in the system, $C_{Si,M}$ and $C_{Si,B}$ (wt %) is the Si content in middle and bottom positions of the sample after solidification, respectively.

Fig. 7(d) shows the variation of the real Si removal degree and Mg_2Si accumulation layer thickness with cooling rate. Within the cooling rate range of 30–150 K/h, the overall Si removal degree was weakly dependent on cooling rate. In the Al-9%Si-15%Mg system, the average Si removal degree was 43.5%, whereas increasing the Mg content to 20% enhanced the Si removal degree to 62.2%. However, Fig. 7(d) shows that the thickness of the primary Mg_2Si accumulation layer at the top region of ingot varied markedly with cooling rate at Mg additions of 15% and 20%. As the cooling rate decreased, the primary Mg_2Si accumulation layer became significantly thinner. In the Al-9%Si-15%Mg system, the layer thickness decreased from 28.7 mm at 150 K/h to 18.1 mm at 30 K/h. Similarly, in the Al-9%Si-20%Mg system, the thickness

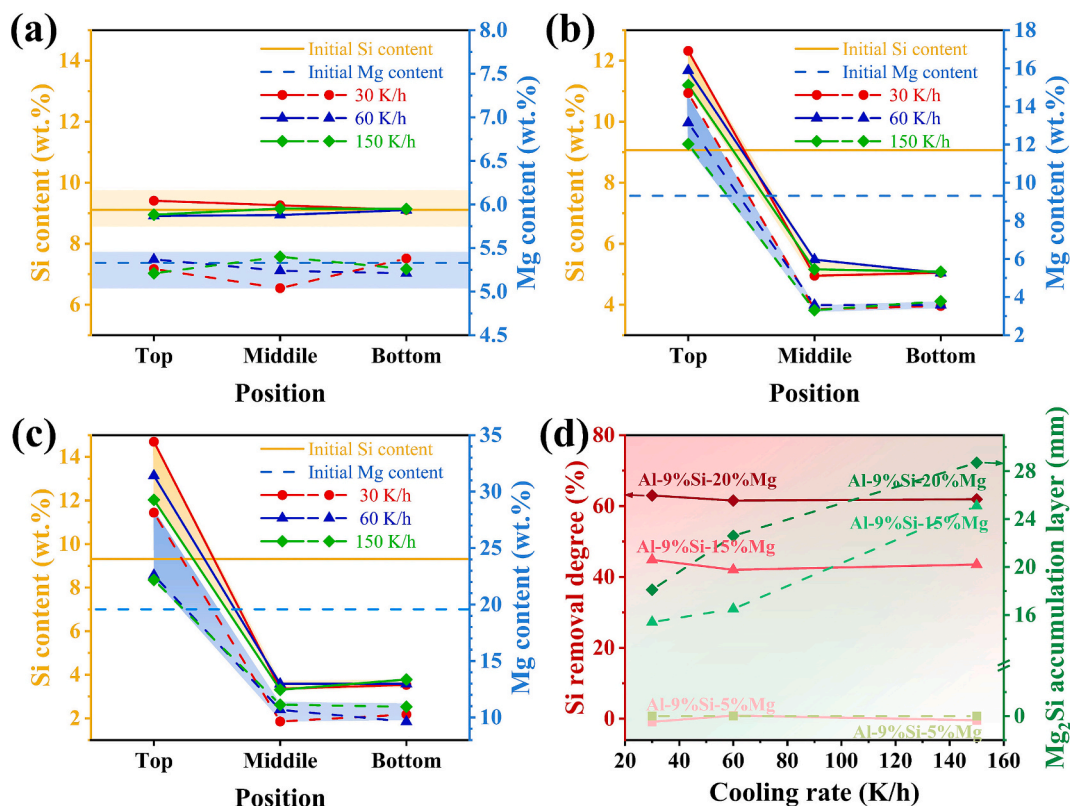


Fig. 7. Mg and Si contents at the top, middle, and bottom of samples solidified at different cooling rates of 30, 60, and 150 K/h: (a) Al-9%Si-5%Mg, (b) Al-9%Si-15%Mg, (c) Al-9%Si-20%Mg, and (d) variation of Si removal degree and primary Mg_2Si accumulation layer thickness with cooling rate.

decreased from 25.1 mm at 150 K/h to 15.4 mm at 30 K/h. With lower cooling rate, the primary Mg₂Si particles at the top position exhibited coarsening with reduced interparticle spacing, which leads to the formation of a compact but thinner enrichment layer. This morphology enhanced the effective enrichment of Si, thereby improving the yield of the low-Si regions of the samples.

3.3.2. Floatation velocity calculation

The results presented in sections 3.2 and 3.3 demonstrate that a dense primary Mg₂Si aggregation layer formed in the top region of the Al-9%Si-15%Mg and Al-9%Si-20%Mg samples and the thickness of this layer was found to depend on the melt cooling rate. This observation indicated that the development of the accumulation layer is strongly associated with the floatation behavior of primary Mg₂Si particles. Stokes' law is commonly employed to calculate the sedimentation/floatation velocity of particles in fluids, under the assumption that the particles are perfect spheres. In practice, however, primary Mg₂Si exhibits irregular polyhedral morphologies. To improve the accuracy of the velocity estimation, a shape factor (*K*) was introduced. Accordingly, the modified equation for the sedimentation (or floatation) velocity of non-spherical particles can be expressed as follows [51]:

$$V = \frac{K_V(\rho_S - \rho_L)gD_V^2}{18\eta} \quad (3)$$

where *V* is the terminal settlement (or floatation) velocity of particle, *K_V* is the volume shape factor that accounts for the deviation from spherical geometry, ρ_S and ρ_L are the density of Mg₂Si particle (1.99 g/cm³ [45]) and the melt (ρ_L =2.2 g/cm³, Al-9%Si-15%Mg, 680 °C [52]), η is the dynamic viscosity of the melt (approximately 1.2×10^{-3} N·s/m² [53,54]), *D_V* is the volume-equivalent diameter of the irregular particle (same volume as a sphere).

The volume shape factor (*K_V*) is commonly related to the particle sphericity (ϕ), which quantifies the degree of similarity between a particle and a perfect sphere [55].

$$K_V = \phi^{0.83} \quad (4)$$

Sphericity is defined as the ratio of the surface area of the equivalent sphere (*A_{sph}*) to the actual surface area of particle (*A_{sph}*) [56]:

$$\phi = \frac{A_{sph}}{A_s} \quad (5)$$

For a sphere with volume-equivalent diameter (*d_n*), where *A_{sph}* is calculated by:

$$A_{sph} = 4\pi(d_n/2)^2 \quad (6)$$

The above equation requires three-dimensional surface area information to accurately calculate the true 3D sphericity. Due to lack of three-dimensional data, a two-dimensional approximation was employed by using the circularity parameter in this work. Circularity (also called surface sphericity) can be considered a 2D equivalent of sphericity in that it compares the perimeter of a particle projection to its area [57,58]. The equation for circularity (*C*) is defined as follows [59]:

$$C = \frac{4\pi A}{P^2} \quad (7)$$

where *P* is the projected perimeter of the particle in its direction of motion, *A* is the projected area in the direction of motion.

SEM images of particles were analyzed by Image J software to extract the projected area and perimeter along the direction of motion. The primary Mg₂Si particle outlined by the green dashed line in Fig. 7(c) was selected as a representative example for calculation. The results revealed that the projected area of a representative particle was 57,954.8 μm², with a corresponding perimeter of 932.8 μm. Substitution of these values into the above Eq. (7) resulted in an approximate circularity of 0.84.

Similarly, in this study, the volume-equivalent diameter of irregular

particles was approximated using a two-dimensional approach based on the SEM image. Specifically, the equivalent diameter in the two-dimensional scale is defined as the diameter of a circle having the same projected area as the particle, and can be expressed as:

$$D = \sqrt{\frac{4A}{\pi}} \quad (8)$$

Based on the Eq. (8), the equivalent diameter of the particle was determined to be 271.6 μm. By substituting the obtained parameters into Eq. (3), the sedimentation velocity of this particle was 6.5 mm/s. Consequently, the floatation time for this particle from the bottom to the top of the melt with the floatation distance considered as 7 cm was 10.7 s, demonstrating an exceptionally rapid floatation process. With a lower cooling rate, primary Mg₂Si particles have more time to grow and coalesce during floatation, which promotes the formation of a denser accumulation layer in the upper region, thereby resulting in a reduction in its thickness.

3.4. Mechanism of Si removal by Mg-assisted suspension-based crystallization

In this study, an upcycling method for Si removal from Al—Si scraps via Mg-assisted suspension-based crystallization was proposed. Different from conventional suspension-based fractional crystallization that primarily rely on solute distribution and diffusion, this study establishes a novel framework for Si removal by controlling phase transition reactions and intermetallic compound trapping. Specifically, Mg addition thermodynamically converts dissolved Si in the Al melt into a stable primary Mg₂Si phase, thereby transferring Si from the liquid solution into a separable carrier solid. This enables effective Si removal during solidification, whereas simple crystallization alone cannot remove Si due to its eutectic reaction in Al—Si hypoeutectic alloys.

The schematic diagram of its mechanism is shown in Fig. 8. In the first stage, Al—Si scraps and Mg additions are completely melted to form a homogeneous liquid, where Si and Mg atoms are uniformly distributed. In the second stage, as the melt continues to cool down, Mg and Si atoms nucleate and crystallize to form polyhedral primary Mg₂Si particles. Once formed, these particles rapidly rise toward the upper surface due to their lower density relative to the melt, and continue to grow as cooling process. In the third stage, the floating primary Mg₂Si particles accumulate at the top of the melt, forming a distinct Mg₂Si-rich layer and thereby achieving the Si removal from the melt. In the fourth stage, as the temperature decreases further to the eutectic point, a eutectic reaction occurs in the remaining liquid, leading to the simultaneous precipitation of α-Al and eutectic Mg₂Si. In contrast to the primary Mg₂Si phase, the eutectic Mg₂Si forms with relatively low volume fraction and becomes entrapped within the α-Al dendritic network, which inhibits the floatation and separation of eutectic Mg₂Si.

3.5. Comprehensive assessment of this method

The above results demonstrate that this method can effectively remove Si from Al—Si cast alloys. Compared with other approaches, such as molten salt electrolysis and directional solidification, this technique is simple to operate, requires no complex equipment, and is environmentally friendly. From an alloy design perspective, Mg is also a widely used alloying element in wrought aluminum alloys, which ensures compatibility with existing wrought alloy compositions and reduces the introduction of undesirable impurities [60]. However, a current limitation is the relatively high Mg content in the products, which restricts their direct use as conventional wrought alloys. Future development of high-Mg wrought alloys through tailored alloy design could expand their applicability. Furthermore, as Mg exhibits a vapor pressure approximately 8–9 orders of magnitude higher than that of aluminum [61], Mg can be easily separated from the product through

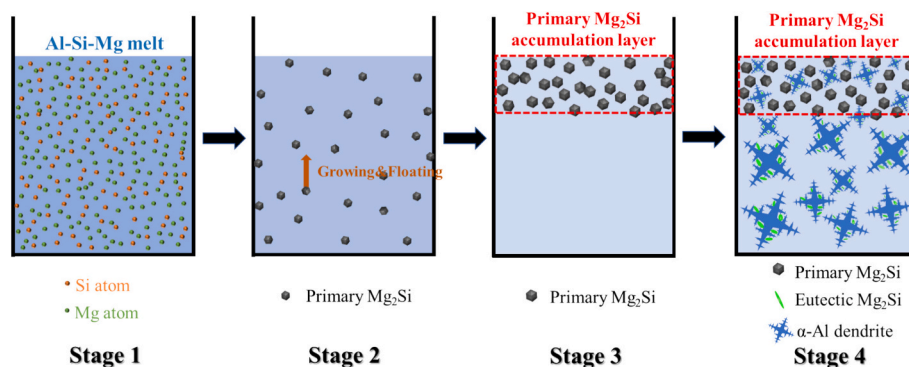


Fig. 8. Mechanism diagram of Si removal from Al alloys by Mg-assisted suspension-based crystallization.

vacuum distillation, enabling its recovery and reuse as an additive. From an economic standpoint, the material cost of this method is therefore negligible, with energy consumption being the primary factor of concern.

The main energy consumption in this method arises from the remelting of the aluminum alloy with magnesium addition and the subsequent controlled slow-cooling stage. In industrial practice, the energy required for aluminum remelting can be controlled at 390.3 kWh/t, while the energy consumption during the holding stage can be as low as 34.7 kWh/h [62]. Therefore, by approximating the entire operation (melting and controlled cooling at a rate of 60 °C/h for 2 h) as a combination of melting and two hours of holding, the total energy consumption is estimated to be around 459.7 kWh/t (0.46 kWh·kg⁻¹Al). If the energy consumption of vacuum distillation for Mg removal requires further consideration, research has demonstrated that vacuum distillation can achieve a Mg removal rate of 99.91% in an Al-10.41%Mg alloy, with an electricity consumption below 3.30 kWh/kg [63]. Accordingly, the total energy consumption of this proposed strategy including vacuum distillation is estimated to be 3.76 kWh·kg⁻¹Al, i.e., 13.54 MJ·kg⁻¹Al. For comparison with representative Al upcycling method reported in the literature, this energy demand is substantially lower than solid-state electrolysis (65.4 MJ·kg⁻¹Al [17]) and lower than the multi-pass directional solidification with electromagnetic force method (7 MJ·kg⁻¹Al) per pass, three passes to reduce Si from 7% to <1%, i.e., 21 MJ·kg⁻¹Al in total [31]), while being comparable to centrifugal separation under isothermal semi-solid conditions (9.2–10.9 MJ·kg⁻¹Al) [32]).

Experimental results indicated that a certain amount of aluminum is entrained within the primary Mg₂Si accumulation layer. Further research is required to enhance aluminum recovery rates by improving separation methods for primary Mg₂Si and aluminum, such as utilizing electromagnetic fields, filtration, or controlled stirring to further aggregate Mg₂Si phase and release the entrapped Al. The refined Al-Mg-Si product after Si removal contains an elevated Mg level. While this composition may not be directly compatible with conventional wrought alloy families without further adjustment, it nevertheless offers clear and practical valorization routes. First, the refined Al-Mg-Si product can serve as raw material for developing high-Mg Al alloys that are increasingly explored for their potential strength-ductility synergy [64]. Second, the Mg level can be reduced to meet established Al-Mg wrought alloy windows via Mg removal routes (e.g., vacuum distillation). Notably, increasing Mg addition within an appropriate range can further enhance the Si removal efficiency, enabling flexible tuning of both Si and Mg to target the desired alloy composition range.

Overall, this separation strategy demonstrates high technical feasibility, process scalability, and energy efficiency, providing a cost-effective and reliable purification route for large-scale treatment and quality improvement of Al-Si cast scraps. Future work will address the recycling and valorizing the Mg₂Si-rich fraction (e.g., for

thermoelectrics) and adjusting the Mg content of the refined Al-Mg-Si melt (e.g., by vacuum distillation), as well as exploring its potential use in high-Mg Al alloy development.

4. Conclusion

In this study, we developed a Mg-assisted suspension-based fractional crystallization strategy that enables efficient, economical, and eco-friendly upcycling of Al-Si cast scraps. The study clarifies the phase-selection window, process-microstructure-removal degree relationships, and flotation-driven separation mechanism, and evaluates the scalability of the alloy upcycling. The main findings of this study are summarized as follows:

- (1) Theoretical analysis and process optimization indicated that when the Mg content exceeds 6.5% in the Al-9%Si-x%Mg system, a single primary Mg₂Si phase formed during the initial solidification stage. Furthermore, the increase of Mg content led to a significant improvement of the Si removal degree. In the Al-9%Si-20%Mg system at a cooling rate of 30 K/h, the Si removal degree of 63% was achieved.
- (2) The reduction of the melt cooling rate mainly promoted the growth and agglomeration of primary Mg₂Si, thereby forming a denser accumulation layer of primary Mg₂Si at the top and reducing its thickness. In the Al-9%Si-20%Mg system, when the cooling rate decreased from 150 K/h to 30 K/h, the thickness of the primary Mg₂Si accumulation layer decreased from 25.1 mm to 15.4 mm. The calculated buoyant rise velocity of representative primary Mg₂Si particles is 6.5 mm/s, supporting the high efficiency of flotation-driven Si separation.
- (3) The separation and purification mechanism proceeds in two steps: (i) the Al-Si alloy and Mg additive are melted to form a homogeneous liquid with high Mg content; (ii) polyhedral or dendritic primary Mg₂Si precipitates during the first solidification stage and rapidly floats to selectively separate Si. In contrast, the eutectic Mg₂Si is entrapped within the α-Al dendritic network, thereby preventing their separation.
- (4) This proposed method enables effective Si separation from Al-Si cast scraps through a simple operation with minimal equipment requirements, offering an environmentally friendly and energy-efficient route. The refined Al-Mg-Si melt is composition-tunable, supporting cost-effective integration into multiple wrought alloys.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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