

Role of Ni-rich intermetallics evolution in the physical properties of Al-Si-Ni alloys: Elastic modulus and thermal expansion behavior

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ABSTRACT

The effects of Ni-rich intermetallic compounds (IMC) evolution on the elastic modulus (E) and coefficient of thermal expansion (CTE) of Al-18Si-xNi alloys ($x = 4-28$ wt%) were systematically investigated under cooling rates of 5 and 25 K/s. At high Ni contents, the Ni-rich IMCs evolve from a single Al_3Ni phase into multiphase structures consisting of Al_3Ni , $\text{Al}_6\text{Ni}_3\text{Si}$, and Al_3Ni_2 . Since these IMCs constitute a large volume fraction, the alloys were treated as a composite of compliant α -Al matrix and stiff intermetallic reinforcements. The analyzed E and CTE were correlated with phase fractions quantified by Scheil simulation, XRD, and EBSD, together with phase-specific properties measured by nanoindentation and high-temperature XRD. Al_3Ni_2 (235 GPa and $7.7 \times 10^{-6}/\text{K}$) and $\text{Al}_6\text{Ni}_3\text{Si}$ (190 GPa and $7.4 \times 10^{-6}/\text{K}$) were identified as the dominant stiffening and low-CTE constituents. Their sequential formation at 20 and 24 wt%Ni altered the compositional dependence of both properties. The resulting phase fractions, and thus the physical properties, were highly sensitive to cooling rate. Composite models employed in this study successfully predicted experimental trends of E and CTE. With increasing Ni content, the elastic response shifted from the Voigt-Reuss lower bound, dominated by the α -Al matrix, toward the Hashin-Shtrikman and Mori-Tanaka predictions, indicating cooperative load sharing among interconnected IMCs. In contrast, the measured CTE showed the best agreement with the constraint-based Turner and Kerner models. Increasing the cooling rate to 25 K/s promoted the retention of Al_3Ni_2 while suppressing subsequent IMC evolution, which weakened the stiffening effect but further reduced the CTE. These results indicate that the connectivity and solidification-dependent evolution of Ni-rich IMCs, together with their volume fraction, control the physical properties of high-Ni containing Al-Si-Ni casting alloys. Since the investigated cooling rates correspond to thick and thin sections of permanent-mold castings, section thickness should be considered in tailoring the stiffness and thermal expansion of such components.

1. Introduction

Al-Si casting alloys are increasingly adopted in automotive and aerospace structural components owing to their excellent castability, high-temperature strength, and relatively low coefficient of thermal expansion (CTE) [1–3]. While extensive efforts have focused on improving mechanical properties to meet light-weighting demands for next-generation mobility, further enhancement of physical properties, particularly elastic modulus (Young's modulus, E) and thermal expansion behavior, remains essential for replacing conventional steel components. Elastic modulus, which quantifies stiffness and dimensional stability under load, is determined principally by interatomic bonding. Therefore, conventional matrix-strengthening strategies (e.g.,

solid-solution or precipitation hardening) increase the yield strength markedly while altering E only marginally. Thus, introducing high-modulus reinforcing phases is an effective approach to enhancing E . Increasing the Si content to increase the fraction of Si can improve stiffness, but may also reduce ductility, resulting in a significant trade-off. In addition, Al-based metal matrix composites (MMCs) reinforced with high-modulus ceramic particles such as SiC and Al_2O_3 offer an alternative route to high stiffness, but their industrial implementation remains limited by processing complexity, heterogeneous reinforcement clustering, degraded formability, and poor recyclability [4,5]. An *in situ* alloying strategy capable of intrinsically increasing E while retaining conventional casting routes is therefore of substantial scientific and technological interest.

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Alloying Al–Si with transition metals, particularly Ni, provides such a route by introducing stiff and thermally stable intermetallic compounds (IMCs) owing to the negligible solid solubility of Ni in Al [3]. In near-eutectic Al–Si alloys, the addition of 2–6 wt.% Ni has been reported to increase E nearly linearly by approximately 1.9 GPa per wt.% Ni, primarily through the load-bearing effect of hard Al_3Ni intermetallics [6]. High Ni additions have been reported to affect fluidity and solidification behavior of Al–Si alloys [7–9]. In particular, the formation of primary Al_3Ni has been shown to reduce the fluidity of Al–Ni–Si alloys, since the primary crystals formed at high temperatures impede the flow of the remaining liquid [9].

CTE is another key physical property because excessive thermal mismatch at elevated temperature shortens component lifetime by promoting loss of dimensional tolerance, seizure, or interfacial damage [10]. In multicomponent Al–Si alloys, the bulk CTE is controlled by the intrinsic thermal expansion characteristics and volume fractions of the constituent phases. Hypereutectic Al–Si alloys therefore exhibit inherently low CTE owing to the high fraction of hard Si [11]. However, further increasing the Si content to reduce CTE raises the liquidus temperature and promotes the undesirable coarsening of primary Si (Si_p), which degrades both castability and mechanical reliability [3,12]. Vijayan [13] reported that in high-Si alloys (≈ 23 wt% Si), minor Ni additions refine and modify Si_p morphology and thereby indirectly reduce CTE. Ni additions thus offer an alternative pathway to concurrently increase E and reduce CTE, by substituting excess Si with low-CTE, high-stiffness Ni-rich IMCs [11,14–16]. Although Si is more effective in reducing the CTE and density per unit volume fraction, excessive Si additions are limited by the increased liquidus temperature and the coarsening of primary Si. In contrast, Ni-rich IMCs provide substantially higher stiffness, although Ni increases the material cost and alloy density. Therefore, the benefit of Ni additions should be evaluated in terms of the combined improvement in E , specific modulus, and CTE.

Previous studies on Al–Si–Ni alloys have focused almost exclusively on hypoeutectic or eutectic compositions with relatively low Ni contents (<10 wt%), in which Al_3Ni is the only Ni-rich IMC and mechanical strengthening and thermal stability are attributed to the single phase [17–20]. Extending Ni into the hypereutectic systems with high-Ni additions changes the problem qualitatively rather than quantitatively. As the Ni content increases, the IMC fraction increases substantially, while the Ni-rich intermetallics evolve from a single Al_3Ni phase into multiphase structures. Our recent work on Al–18Si–xNi alloys reported that $\text{Al}_6\text{Ni}_3\text{Si}$ and Al_3Ni_2 form sequentially above approximately 20 and 24 wt%Ni, respectively, producing core-shell and needle-like layered structures in which the three intermetallics coexist [12]. Under these conditions, the physical properties unlikely to simply depend on the fraction of a single reinforcing phase, as they do at low Ni contents. Rather, the type of IMC formed, its connectivity within the multiphase aggregate, and its retention under a given solidification condition may all control the macroscopic response. Nevertheless, quantitative structure-property relationship that resolve these contributions in high-Ni containing hypereutectic Al–Si–Ni alloys have not yet been established.

Therefore, the present study aims to systematically investigate the effects of Ni-rich IMC evolution on the physical properties of hypereutectic Al–Si casting alloys, with particular emphasis on elastic modulus and thermal expansion behavior. Ni content was varied from 4 to 28 wt % in an Al–18Si base alloy to span the compositional range over which the type and fraction of Ni-rich IMCs change markedly, and each alloy was solidified at two different cooling rates (5 and 25 K/s) to clarify the influence of phase constitution. In order to resolve the contribution of each phase, the intrinsic elastic modulus of every constituent phase was measured by nanoindentation and the phase-specific CTE by high-temperature X-ray diffraction, while the phase fractions were quantified independently by Scheil simulation, XRD, and EBSD phase mapping. The measured properties were then compared with composite models of

increasing physical content. Through this approach, we evaluate the contribution of IMC fraction from that of the solidification-dependent IMC architecture.

2. Experimental

2.1. Alloy preparation

Al–18Si–xNi (wt.%) alloys with Ni contents ranging from 4 to 28 wt% were prepared using a steel step mold casting, following the procedure described in Refs. [3,12]. The same alloys reported previously were selected for the present study, so that the phase-specific correlations established here can be related directly to the Ni-dependent solidification behavior and microstructural evolution documented in that work [12]. Therefore, detailed alloy compositions, solidification sequences, and the microstructural characterization are not reproduced here. Only the information required for the present property analysis is summarized.

2.2. Measurement of phase fraction

In this study, phase fractions were quantified using multiple approaches. Thermodynamically predicted fractions of the Ni-rich IMCs as a function of Ni content were taken from the Scheil simulations, which were performed using Thermo-Calc software with TCAL9 database [21]. X-ray diffraction (XRD) were carried out using $\text{Cu-K}\alpha$ radiation ($1.5406 \text{ \AA}$), over a 2θ range from 10 to 140° , with a step size of 0.0131° and a total acquisition time of 1 h. Phase fractions were determined from the diffraction data by the derivative difference minimization (DDM) method [22]. In addition, the IMCs were analyzed by field-emission scanning electron microscopy (FE-SEM, Mira 1 LHM, TESCAN) equipped with an electron backscatter diffraction (EBSD, Oxford) detector. The analysis conditions were an accelerating voltage of 15 kV, a step size of $1.2 \text{ }\mu\text{m}$, and a magnification of $\times 200$, and only EBSD data with an indexing rate of 95% or higher were used for analysis. For each alloy, phase fractions were quantified from five independently acquired low-magnification EBSD phase maps collected over representative areas of each alloy to ensure that the measured values were statistically representative of the bulk microstructure, and the standard deviation was reported together with the quantitative results.

2.3. Measurement of macroscopic properties

The as-cast ingots were cut into $40 \times 15 \times 3 \text{ mm}$ specimens for measuring elastic modulus. The dynamic Young's modulus, E (GPa) was determined by the impulse excitation technique (IET) according to ASTM E1876 [23]. Each rectangular specimen was subjected to forced vibration, and the resonance frequency of the flexural mode was recorded. The modulus was then evaluated as

$$E = 0.9465 \left(\frac{mf^2}{w} \right) \left(\frac{l^3}{t^3} \right) T_1 \quad (1)$$

where m is the specimen mass, f the measured resonance frequency (Hz), and w , l , t the width, length, and thickness of the specimen, respectively. T_1 is the correction factor for flexural mode, which accounts for the specimen thickness and Poisson's ratio, and was calculated according to ASTM E1876 [23]. The specific modulus ($10^6 \text{ m}^2/\text{s}^2$) was calculated using the density obtained from the apparent volume of the specimens.

For thermal expansion measurement, cylindrical specimens of $\phi 6 \times 25 \text{ mm}$ were machined from the same ingots. The coefficient of thermal expansion (CTE, $\times 10^{-6}/\text{K}$) was measured from 25°C to 200°C at a heating rate of $5^\circ\text{C}/\text{min}$ using a thermomechanical analyzer (TMA, TMA 402F1, NETZSCH). All measurements were performed under a high-purity argon atmosphere to protect the specimens from oxidation.

2.4. Measurement of phase-specific properties

Composite-model prediction requires the intrinsic elastic and thermal properties of every constituent phase. To relate the measured elastic modulus to the evolving fraction of Ni-rich IMCs, the Young's modulus of each phase was measured by nanoindentation (iMicro, KLA) using Berkovich indenter tips at a peak load of 5 mN. The modulus was calculated in accordance with ASTM E2546 [24] from

$$E_r = \sqrt{\pi} S / (2\beta\sqrt{A}) \quad (2)$$

where E_r is the reduced modulus, which accounts for elastic deformation of both the specimen and the indenter. The contact area A was determined from the indentation depth using the indenter geometry, the contact stiffness S from the slope of the unloading curve, and the indenter shape correction factor β was taken as 1.034 for the Berkovich indenter.

Because the eutectic Si (Si_e) lamellae are finer than the plastic zone generated beneath the indenter, their modulus cannot be measured in isolation and was instead deconvoluted from the composite response of the indented volume. The plastic zone radius (r_p) was estimated from Johnson's expanding cavity model [25,26]:

$$r_p = a_c \left\{ (1/6(1-\nu)) [E/\sigma_y \tan(90^\circ - \theta) + 4(1-2\nu)] \right\}^{1/3} \quad (3)$$

where a_c is the contact radius and θ is the half-cone angle to the surface, taken as 70.3° for the equivalent cone of a three-sided pyramid Berkovich indenter. Values of a_c were obtained for each indentation by the Oliver–Pharr method [26,27]. A Poisson's ratio $\nu = 0.25$ and an elastic modulus $E = 123.51$ GPa, both determined by IET on the 28Ni alloy, were used consistently for all alloys. The yield strength was determined as 333.3 MPa, derived from the hardness presented in Table 1 using the Tabor theory [28]. Each indent was then enclosed by a circle of radius r_p . The area fraction of eutectic Si_e and α -Al matrix within this circle was measured, and the hardness value of Si_e was extracted by applying the rule of mixtures.

The thermal expansion of each individual phase was determined by high-temperature XRD (Empyrean, Malvern Panalytical) performed *in situ* from room temperature to 400°C in 100°C increments. The lattice parameters of the α -Al matrix, Si, Al_3Ni , $\text{Al}_6\text{Ni}_3\text{Si}$, and Al_3Ni_2 phases were refined from the diffraction peaks recorded at each temperature according to their respective crystal structures [37], and the specific CTEs were obtained from the temperature dependence of the lattice parameters.

2.5. Composite-model calculation of effective properties

To systematically assess the influence of Ni-rich IMC evolution on the macroscopic properties of hypereutectic Al-Si-Ni casting alloys, the effective elastic modulus and CTE were estimated using composite-model approaches. The phase fractions determined in Sections 2.2 and the phase-specific properties determined in Section 2.4 were used as inputs, allowing the predictions based on thermodynamic, XRD-, and

EBSD-derived fractions to be compared directly against the measured values. The governing equations and physical assumptions of the models employed are summarized in Table 2.

3. Results

3.1. Quantitative phase fractions of Al-18Si-xNi alloys

Fig. 1 shows EBSD phase maps of as-cast Al-18Si-xNi alloys with various Ni contents solidified at different cooling rates. The microstructures consist of Si particles and Ni-rich IMCs (Al_3Ni , $\text{Al}_6\text{Ni}_3\text{Si}$, and Al_3Ni_2). As the Ni content increases from 4 to 16 wt%, both the size and the volume fraction of Al_3Ni increase significantly. When the Ni content exceeds 20 wt%, the morphology of the Ni-rich IMCs changes significantly. For example, the IMCs in the 20Ni alloy exhibits a cloud-like morphology (Fig. 1(c–g)). At 24 wt% Ni, the cloud-like IMCs disappear and needle-like IMCs form (Fig. 1(d–h)). Microstructural analysis revealed that the cloud-like IMCs consist of a binary mixed structure composed of $\text{Al}_6\text{Ni}_3\text{Si}$ and Al_3Ni , whereas the needle-like IMCs consist of a ternary mixed structure composed of Al_3Ni_2 , $\text{Al}_6\text{Ni}_3\text{Si}$, and Al_3Ni , respectively. In addition, the cooling rate significantly influences the area fractions of $\text{Al}_6\text{Ni}_3\text{Si}$ and Al_3Ni_2 formed at the centers of the mixed structures, resulting in a clear difference in the phase distribution within the mixed IMCs. A detailed investigation of the microstructural evolution and phase formation in Al-18Si-xNi alloys with Ni contents ranging from 4 to 28 wt% was presented in our previous study [12]. Therefore, only a brief description of the representative microstructures relevant to the present work is provided here.

To evaluate these effects quantitatively, phase fractions were first predicted using Scheil simulations. The calculated phase fractions of the Al-18Si-xNi alloys are presented in Fig. 2. The simulations predict the onset of $\text{Al}_6\text{Ni}_3\text{Si}$ and Al_3Ni_2 formation at approximately 20 and 24 wt% Ni, respectively, which corresponds to the transition between different primary phase fields in the Al-Si-Ni ternary phase diagram. The calculated results indicate that the formation of $\text{Al}_6\text{Ni}_3\text{Si}$ progressively reduces the fraction of Al_3Ni , while the subsequent formation of Al_3Ni_2 decreases both Al_3Ni and $\text{Al}_6\text{Ni}_3\text{Si}$ fractions. However, thermodynamics-based simulations are unlikely to fully reproduce the experimentally observed microstructures because diffusion-controlled transformations and peritectic reactions significantly influence phase stability during solidification [12].

Therefore, quantitative phase analysis (QPA) was performed using XRD combined with the derivative difference minimization (DDM) method, and the results were further validated using EBSD phase mapping. Fig. 2 compares the phase fractions obtained from Scheil simulations, XRD-DDM analysis, and EBSD phase mapping for alloys solidified at cooling rates of 5 and 25 K/s. Although the measured phase fractions generally follow the trends predicted by the Scheil simulations, quantitative phase analysis based solely on XRD may involve uncertainties arising from segregation, particle-size effects, and peak overlapping. Therefore, EBSD phase mapping was additionally employed as a complementary technique to directly quantify phase fractions from the as-cast microstructures.

Table 1
Nanoindentation hardness and elastic properties of constituent phases in the Al-18Si-xNi alloys.

Phase	Hardness (GPa)	Young's modulus, E (GPa)		Bulk modulus, K (GPa)	Shear modulus, G (GPa)	Poisson's ratio (ν)	Aspect ratio (ξ)
		Measured	Ref				
α -Al	1.03	79.95	84.6 [29], [30]	83.28	29.83	0.34 [31]	-
Primary Si, Sip	11.81	160.91	162.0 [32]	111.74	63.85	0.26 [33]	1.37
Eutectic Si, Sie	1.23 (with α -Al)	111.46	129.5 [29]	77.40	44.23	0.26 [33]	1.63
Al_3Ni	7.36	158.62	166.9 [34]	80.11	67.79	0.17 [35]	2.61 (4–16Ni) 1.72 (20–22Ni)
$\text{Al}_6\text{Ni}_3\text{Si}$	11.32	190.14	-	121.88	76.67	0.24	1.92 (24–28Ni)
Al_3Ni_2	12.37	235.16	228.0 [36]	186.63	91.15	0.29 [35]	1.72 (20–22Ni) 1.92 (24–28Ni) 1.92

Table 2Composite models used for predicting elastic modulus (E) and coefficient of thermal expansion (CTE).[38], [39], [40], [41], [42], [42], [42].

	Model	Equation	Parameter calculation	Physical meaning	ref
E	Voigt-Reuss	upper	$E_a = E_m V_m + E_p V_p$	Rule of mixtures bounds; Bound estimates from iso-strain/stress assumptions	[37]
		lower	$\frac{1}{E_a} = \frac{V_m}{E_m} + \frac{V_p}{E_p}$		
	Hashin-Shtrikman	upper	$K_a = K_m + \frac{1-V_m}{\frac{1}{K_p-K_m} + \frac{3V_m}{3K_m+4G_m}}, G_a = G_m + \frac{1-V_m}{\frac{1}{G_p-G_m} + \frac{6(K_m+2G_m)V_m}{5(3K_m+4G_m)G_m}}$	Theoretical upper/lower bounds considering phase interaction	[39]
		lower	$E_a = \frac{9K_a G_a}{(3K_a + G_a)}$ $K_a = K_p + \frac{1-V_p}{\frac{1}{K_m-K_p} + \frac{3V_p}{3K_p+4G_p}}, G_a = G_p + \frac{1-V_p}{\frac{1}{G_m-G_p} + \frac{6(K_p+2G_p)V_p}{5(3K_p+4G_p)G_p}}$		
	Mori-Tanaka	$K_a = K_m + \Sigma \left[\frac{V_p(K_p-K_m)}{1 + \frac{3(1-V_m)(K_p-K_m)}{3K_m+4G_m}} \right], G_a = G_m + \Sigma \left[\frac{V_p(G_p-G_m)}{1 + \frac{3(1-V_m)(K_p-K_m)}{G_m(1 + \frac{9K_m+8G_m}{6(K_m+2G_m)})}} \right]$		Mean-field model considering matrix-inclusion interaction	[40]
Halpin-Tsai	$E_a = E_m(\frac{1 + \xi \eta V_p}{1 - \eta V_p})$	$\eta = \frac{E_p/E_m - 1}{E_p/E_m + \xi}$	Semi-empirical morphology-based load-transfer model	[38]	
CTE	Voigt-Reuss	upper	$\alpha_a = \alpha_m V_m + \alpha_p V_p$	Idealized bounds for constrained thermal expansion	[51]
		lower	$\frac{1}{\alpha_a} = \frac{V_m}{\alpha_m} + \frac{V_p}{\alpha_p}$		
	Turner	$\alpha_a = \frac{\alpha_m K_m V_m + \sum \alpha_p K_p V_p}{K_m V_m + \sum K_p V_p}$	Phase constraint model based on bulk modulus	[51]	
	Kerner	$\alpha_a = \alpha_m + \frac{V_p(\alpha_p - \alpha_m)(K_p + \frac{4}{3}G_m)}{K_m + \frac{2}{3}G_m + V_p(K_p - K_m)}$	Interphase shear constraint model for thermal expansion	[51]	

Note: E = elastic modulus, K = bulk modulus, G = shear modulus, η = elastic modulus contrast parameter, V = volume fraction, ξ = aspect ratio, α = coefficient of thermal expansion (CTE), ν = Poisson's ratio. Subscripts: a = alloy, m = matrix (Al), and p = particle.

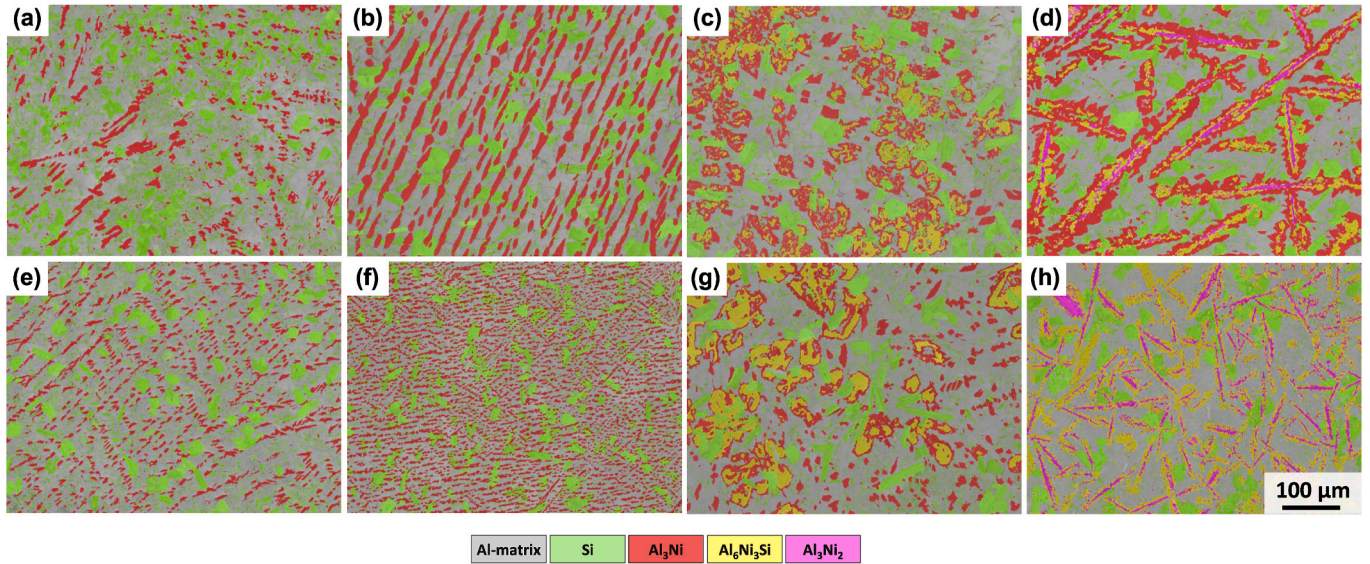


Fig. 1. EBSD phase maps of Al-18Si-xNi alloys solidified at cooling rates of (a-d) 5 K/s and (e-h) 25 K/s: (a, e) 4Ni, (b, f) 16Ni, (c, g) 20Ni, and (d, h) 24Ni.

As can be qualitatively observed in Fig. 1, substantial changes were observed in the fractions of the Ni-rich IMCs with increasing Ni content. As shown in Fig. 2(a), the fraction of Al_3Ni continuously increases as the Ni content increases from 4 to 16 wt% regardless of cooling rate. However, once the Ni content exceeds 20 wt%, where $\text{Al}_6\text{Ni}_3\text{Si}$ formation (black arrow) becomes significant, the fraction of Al_3Ni decreases noticeably. This decrease is particularly pronounced in the EBSD results, indicating that the formation of $\text{Al}_6\text{Ni}_3\text{Si}$ suppresses the stability and retention of Al_3Ni in high Ni containing alloys. A similar tendency was also observed for $\text{Al}_6\text{Ni}_3\text{Si}$. As shown in Fig. 2(b), the fraction of $\text{Al}_6\text{Ni}_3\text{Si}$ initially increases with increasing Ni content, but subsequently decreases once Al_3Ni_2 formation (red arrow) becomes significant at 24 wt% Ni. These results demonstrate that the sequential formation of higher-

order Ni-rich IMCs strongly influences the stability and fraction evolution of the preceding intermetallic phases. A clear cooling-rate dependence was also observed in the phase fractions of the Ni-rich IMCs. As can be seen in Fig. 2(a), the fraction of Al_3Ni in the alloys solidified at 25 K/s becomes significantly lower than that in the alloys solidified at 5 K/s beyond a critical Ni concentration. In contrast, the fractions of $\text{Al}_6\text{Ni}_3\text{Si}$ and Al_3Ni_2 become relatively higher under the faster cooling condition, as shown in Fig. 2(b) and (c). This suggests that rapid solidification suppresses diffusion-controlled peritectic reactions associated with the phase transformation of Al_3Ni_2 to $\text{Al}_6\text{Ni}_3\text{Si}$, thereby altering the final phase constitution depending on cooling rate [12]. These results are consistent with those shown in Fig. 1(c-g) and (d, h).

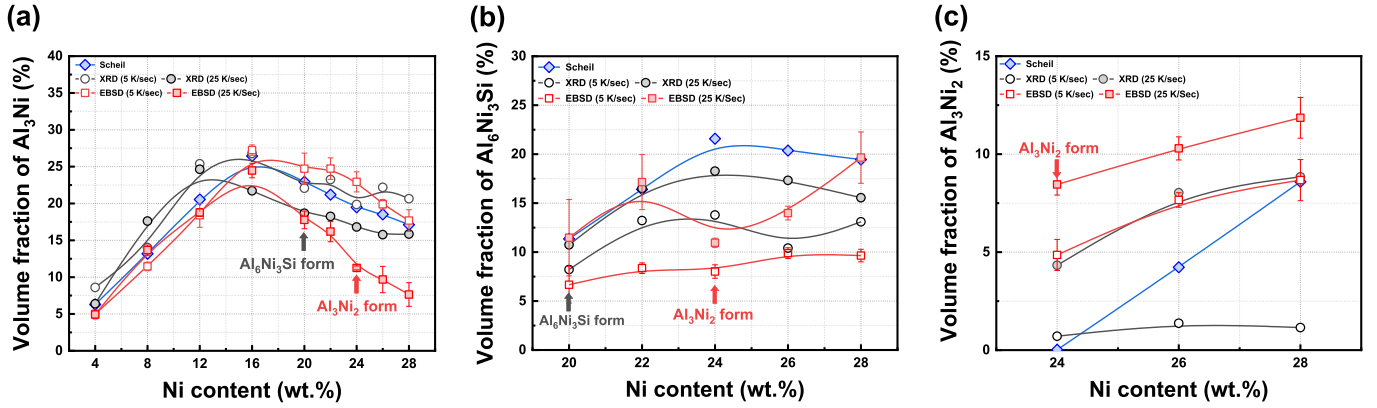


Fig. 2. Phase fractions of the constituent Ni-rich IMCs as a function of Ni content in Al-18Si-xNi alloys, determined by Scheil simulation, XRD, and EBSD phase mapping for alloys solidified at cooling rates of 5 and 25 K/s: (a) Al_3Ni , (b) $\text{Al}_6\text{Ni}_3\text{Si}$, and (c) Al_3Ni_2 .

3.2. Physical properties of Al-18Si-xNi alloys

Fig. 3 shows the elastic modulus (E) and specific modulus of the Al-18Si-xNi alloys solidified at cooling rates of 5 and 25 K/s as a function of Ni content. As the Ni content increases from 4 to 28 wt%, the E values continuously increases from 86.21 to 123.51 GPa for the alloys solidified at 5 K/s (Fig. 3(a)) and from 86.04 to 118.74 GPa for those solidified at 25 K/s (Fig. 3(b)). Up to 20 wt% Ni, the elastic modulus increases nearly linearly with slopes of approximately 1.47 and 0.92 GPa per wt.% Ni for the alloys solidified at 5 and 25 K/s, respectively. Once $\text{Al}_6\text{Ni}_3\text{Si}$ forms above 20 wt% Ni (black arrow), the increasing trend becomes more pronounced, exhibiting slopes of approximately 2.19 and 1.42 GPa per wt.% Ni for the alloys solidified at 5 and 25 K/s, respectively. However, beyond 24 wt% Ni (red arrow), as shown in Fig. 3(b), where Al_3Ni_2 formation occurs, the increase in elastic modulus becomes more gradual, particularly in the alloys solidified at 25 K/s. In these alloys, the elastic modulus increases by only ~ 0.34 GPa per wt.% Ni above 24 wt%Ni, accompanied by a slight decrease in specific modulus from 36.87 to $36.75 \times 10^6 \text{ m}^2/\text{s}^2$. This decrease is attributed to the retention of the high-density Al_3Ni_2 under rapid solidification, which increases the alloy density more significantly than the elastic modulus.

Fig. 4 shows the coefficient of thermal expansion (CTE) of the Al-18Si-xNi alloys solidified at cooling rates of 5 and 25 K/s as a function of Ni content. The average CTE measured between 25 and 200°C continuously decreases with increasing Ni content, from 20.88 to $14.49 \times 10^{-6}/\text{K}$ for the alloys solidified at 5 K/s (Fig. 4(a)) and from 20.92 to $13.35 \times 10^{-6}/\text{K}$ for those solidified at 25 K/s (Fig. 4(b)). Similar to the elastic modulus behavior, the CTE also exhibits slope changes at

Ni contents of 20 and 24 wt%. At 5 K/s, the slope decreases from 0.35 to 0.17, whereas at 25 K/s it decreases from 0.34 to 0.19. However, beyond 24 wt% Ni, as shown in Fig. 4(a) and (b), the slope increases to 0.33 for the alloy solidified at 25 K/s, while remaining only 0.02 for the alloy solidified at 5 K/s, indicating a much steeper reduction in CTE for the 25 K/s alloy.

The elastic modulus and CTE exhibit similar compositional trends, with changes in slope occurring at the alloy compositions where $\text{Al}_6\text{Ni}_3\text{Si}$ and Al_3Ni_2 begin to form as primary phases. Furthermore, above 24 wt% Ni, where Al_3Ni_2 formation becomes significant, clear cooling rate dependence emerges in both properties even at the same composition. These results indicate that the evolution of Ni-rich IMCs during solidification influences the physical properties of the Al-18Si-xNi alloys.

3.3. Physical properties of phases in Al-18Si-xNi alloys

To investigate the origin of the physical-property variations in the Al-18Si-xNi alloys, the elastic modulus and thermal expansion behavior of the constituent phases were analyzed. The Al-18Si-28Ni alloy, which contains all phases observed in the investigated alloy system, was selected for this analysis.

Fig. 5(a) shows representative nanoindentation marks obtained from the α -Al matrix, primary Si (Si_p), eutectic Si (Si_e) and the Ni-rich IMCs (Al_3Ni_2 , $\text{Al}_6\text{Ni}_3\text{Si}$, and Al_3Ni phases) in the Al-18Si-28Ni alloy. The Ni-rich IMCs and primary Si exhibit significantly smaller indentation sizes than the α -Al matrix. Direct nanoindentation analysis of eutectic Si was challenging due to the fine microstructural scale and the influence

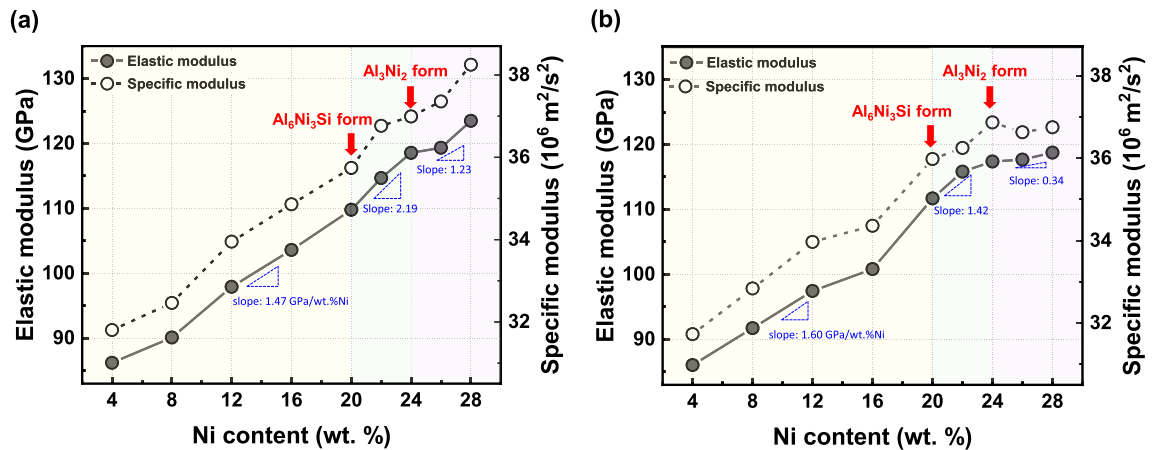


Fig. 3. Elastic modulus and specific modulus of Al-18Si-xNi alloys as a function of Ni content for alloys with microstructures dominated by Al_3Ni , $\text{Al}_6\text{Ni}_3\text{Si}$ and Al_3Ni_2 . (a) 5 K/s; (b) 25 K/sec

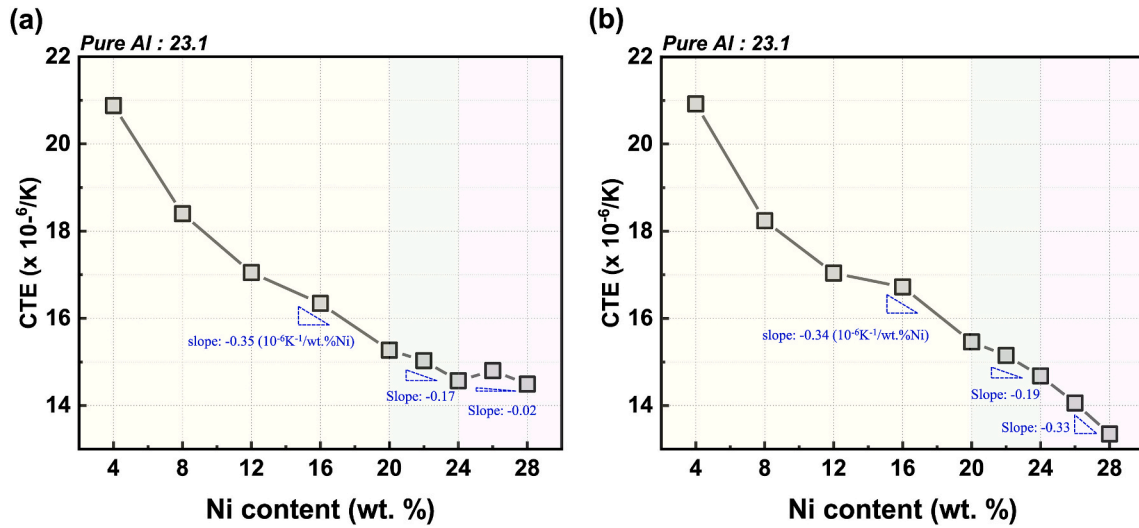


Fig. 4. Coefficient of thermal expansion of Al-18Si-xNi alloys as a function of Ni content for alloys with microstructures dominated by Al_3Ni , $\text{Al}_6\text{Ni}_3\text{Si}$ and Al_3Ni_2 . (a) 5 K/s; (b) 25 K/s

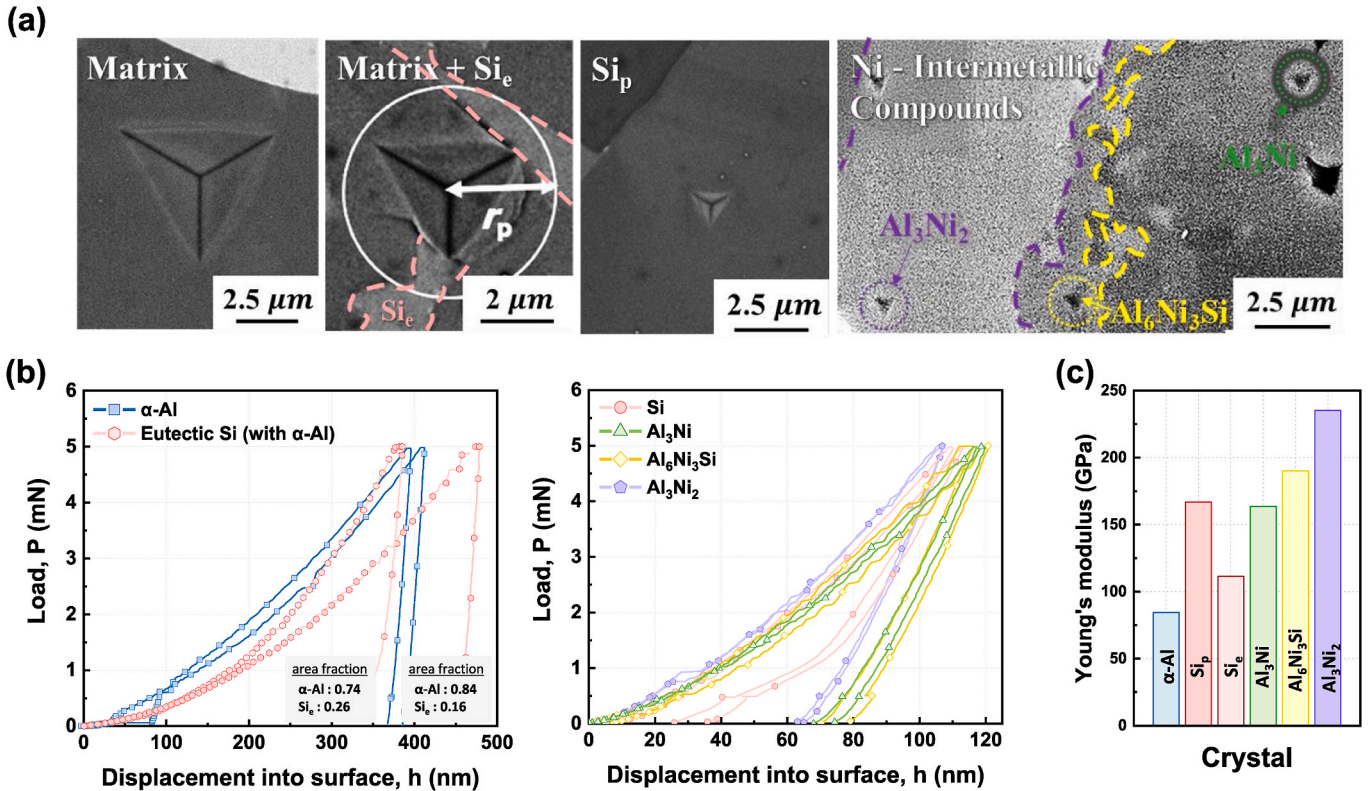


Fig. 5. (a) SEM image showing the indentation marks on Ni-rich IMCs, $\alpha\text{-Al}$ matrix, eutectic Si, and primary Si used for the application of Johnson's cavity expansion model. (b) Corresponding load-displacement (P-h) curves. (c) Young's modulus of the constituent phases.

of the surrounding $\alpha\text{-Al}$ matrix within the plastic deformation zone. Therefore, the modulus and hardness of eutectic Si were estimated using a rule-of-mixtures approach based on the calculated plastic-zone radius (Eq. (3)).

The corresponding load-displacement (p-h) curves are shown in Fig. 5(b). Al_3Ni_2 exhibits the smallest displacement under identical loading condition, followed by $\text{Al}_6\text{Ni}_3\text{Si}$, Si_p , and Al_3Ni whereas the $\alpha\text{-Al}$ matrix shows the largest displacement. Fig. 5(c) and Table 1 summarize the elastic modulus values measured for each phase. The elastic modulus values of the $\alpha\text{-Al}$ matrix, Si_p , Al_3Ni , and Al_3Ni_2 are 79.95, 160.91,

158.62, and 235.16 GPa, respectively. The measured values are in good agreement with previously reported data [29,30,32,34,36,43]. Notably, the elastic modulus of the ternary $\text{Al}_6\text{Ni}_3\text{Si}$ phase was measured to be 190.14 GPa, for which comparative experimental data are currently unavailable in the literature. The elastic modulus of the Si_e was estimated at 111.46 GPa using a rule-of-mixtures approach based on the phase fraction within the plastic zone radius.

Fig. 6 shows the high-temperature XRD profiles of the Al-18Si-28Ni alloy measured between 25 and 400 $^{\circ}\text{C}$. With increasing temperature, the $\alpha\text{-Al}$ peaks shift toward lower 2θ values, whereas the Si and Ni-rich

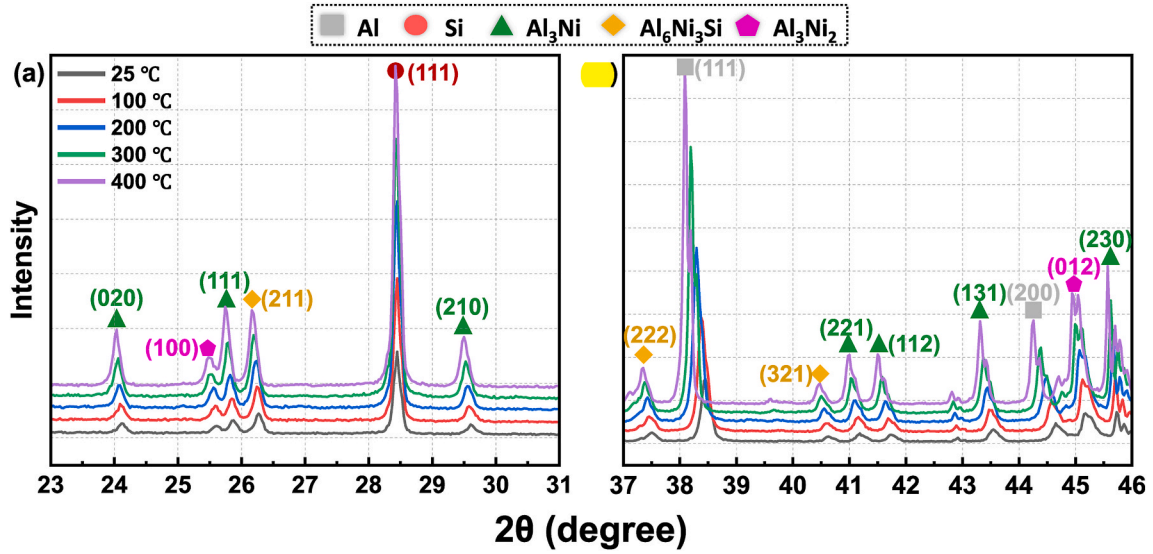


Fig. 6. X-ray diffraction patterns of Al-18Si-xNi alloys measured at different temperatures: (a) low-angle and (b) high-angle 2θ regions.

IMC peaks exhibit relatively smaller shifts. The corresponding lattice-parameter changes are summarized in Fig. 7(a). Detailed crystallographic information for the constituent phases is listed in our previous study [12]. The α -Al matrix exhibits the largest lattice expansion with increasing temperature, with the lattice parameter increasing from 4.05 to 4.09 Å ($\sim 1.0\%$), while Si shows negligible variation, remaining nearly constant at ~ 5.43 Å. The Ni-rich IMCs exhibit relatively small lattice-parameter changes compared with the α -Al matrix. In particular, $\text{Al}_6\text{Ni}_3\text{Si}$ shows only a slight increase from 8.32 to 8.34 Å ($\sim 0.25\%$), whereas Al_3Ni and Al_3Ni_2 exhibit anisotropic lattice expansion depending on crystallographic direction. For Al_3Ni , the a-, b-, and c-axis lattice parameters increase from 6.62 to 6.70 Å (1.33%), 7.38 to 7.40 Å (0.32%), and 4.81 to 4.85 Å (0.70%), respectively. Similarly, Al_3Ni_2 exhibits relatively small lattice expansions, with the a-axis increasing from 4.04 to 4.07 Å (0.68%) and the c-axis from 4.90 to 4.92 Å (0.33%). The lattice-parameter variations were used to estimate the average CTE values of each phase, as summarized in Fig. 7(b) and Table 3.

Due to the random crystallographic orientation in polycrystalline casting alloys, average CTE values were used for interphase comparison. The resulting CTE values follow the sequence: $\text{Al} > \text{Al}_3\text{Ni} > \text{Al}_3\text{Ni}_2 > \text{Al}_6\text{Ni}_3\text{Si} > \text{Si}$.

Table 3

Coefficient of thermal expansion of constituent phases in the Al-18Si-xNi alloy system.

Phase	Coefficient of thermal expansion, CTE ($10^{-6}/\text{K}$)
Al	$a = b = c = 24.6$
Si	$a = b = c = 2.44$
Al_3Ni	$a = 28.6, b = 7.55, c = 22.4$
$\text{Al}_6\text{Ni}_3\text{Si}$	$a = b = c = 7.39$
Al_3Ni_2	$a = b = 8.77, c = 5.65$

4. Discussion

The elastic modulus and coefficient of thermal expansion (CTE) of multiphase Al alloys depend on the volume fraction, morphology, spatial distribution, and intrinsic properties of the constituent phases, as well as on defects such as porosity and interfacial imperfections [11,14,43,44]. In the present Al-18Si-xNi alloys, the Ni-rich IMCs occupy a large volume fraction, so that the alloy can be treated as a composite consisting of a compliant α -Al matrix and stiff intermetallic reinforcements.

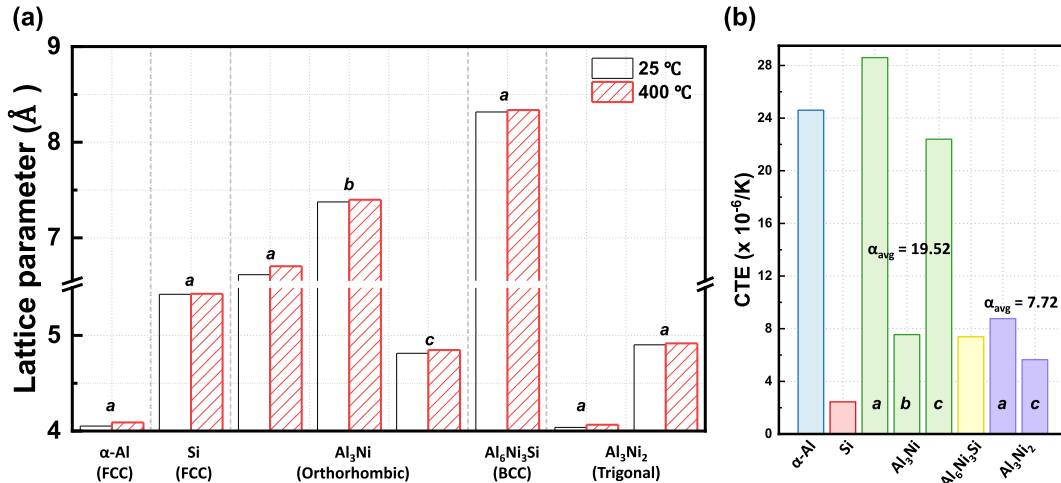


Fig. 7. (a) Lattice parameters and (b) coefficient of thermal expansion of the constituent phases in Al-18Si-xNi alloys measured at 25 and 400 °C.

4.1. Effect of Ni-rich IMC evolution on elastic modulus

In the present Al-18Si-xNi alloys, as shown Fig. S1, the sequential formation of Ni-rich IMCs transforms the microstructure from a matrix-dominated alloy into an increasingly interconnected multiphase system [12]. Then the elastic response should evolve from iso-stress condition, in which deformation is accommodated by the soft matrix, toward a constrained, iso-strain like condition in which load is shared cooperatively among stiff IMCs. To test this hypothesis, the measured elastic modulus was compared with representative composite models, Voigt-Reuss (V-R), Hashin-Shtrikman (H-S), Halpin-Tsai, and Mori-Tanaka models [38–41]. The governing equations and assumptions are summarized in Table 2, and the elastic constants of the constituent phases in Table 1. The bulk modulus (K) and shear modulus (G) of each phase were calculated using $K = E/3(1-2\nu)$ and $G = E/2(1+\nu)$, respectively. The Poisson's ratio (ν) of each phase was obtained from crystallographic databases and literature [31,33,35], except for $\text{Al}_6\text{Ni}_3\text{Si}$, for which no value is available. A value of 0.24 was assumed based on its intermediate hardness between Al_3Ni and Al_3Ni_2 , and on the increased electron density and strengthened interatomic bonding associated with Si incorporation, which is expected to raise Poisson's ratio within the range observed for Al–Ni intermetallic compounds [45,46].

At low Ni levels (4–16 wt%Ni), the measured elastic modulus lies close to the V-R lower bound irrespective of cooling rate or phase-fraction measurement method. This tendency is consistently reproduced using phase fractions obtained from Scheil simulations, XRD analysis, and EBSD phase mapping (Figs. 8 and 9). Since the Reuss model (i.e., V-R lower bound) assumes an iso-stress deformation condition [47], the α -Al matrix accommodates most of the elastic strain, while Si and Al_3Ni act as mechanically isolated reinforcements with limited load-transfer efficiency [39]. With increasing above ~20 wt%, however, the measured elastic modulus departs progressively from the V-R lower bound and approaches the H-S, Halpin-Tsai, and partially V-R upper bound (Voigt model). This transition appears only in the XRD- and EBSD-based predictions (see Fig. 8(b and c) and 9(b,c)), whereas the Scheil-based predictions remain relatively close to the V-R lower bound even at high Ni (Fig. 8(a) and 9(a)). Therefore, the effective elastic response is highly sensitive to the actual solidification microstructure rather than equilibrium phase fraction. The progressive shift toward H-S and Voigt indicate a transition from matrix-dominated deformation toward cooperative stress sharing among multiple stiff IMCs, because these models represent constrained deformation and iso-strain-type load sharing [48,49]. A comparable transition occurs at 25 K/s (Fig. 9), although it is less complete, suggesting that increasing cooling rate suppresses the development of mechanically effective IMC networks even when high modulus phases are present.

This evolution is closely associated with the sequential formation of

$\text{Al}_6\text{Ni}_3\text{Si}$ and Al_3Ni_2 . Unlike the low-Ni alloys, which contain primarily isolated Al_3Ni particles, the high-Ni alloys contain multiple Ni-rich IMCs with substantially larger phase fractions and enhanced microstructural connectivity. In addition, the Ni-rich IMCs in the mixed structures exhibit specific orientation relationships with low interfacial misfits (4.95% for $\text{Al}_6\text{Ni}_3\text{Si}/\text{Al}_3\text{Ni}$ and 0.65% for $\text{Al}_3\text{Ni}_2/\text{Al}_6\text{Ni}_3\text{Si}$) [12], suggesting that the semi-coherent IMC/IMC interfaces facilitate stress transfer within the interconnected IMC aggregates. As a result, matrix constraint, and inter-phase stress transfer become more significant with increasing Ni content. The improved agreement with the H-S and Mori-Tanaka models, both of which incorporate the bulk and shear moduli of all phases, suggests that elastic interaction among the IMCs, rather than their fraction, determines the effective stiffness [24]. The reasonable agreement with the Halpin-Tsai model further indicates that the morphology and distribution of IMCs also influence the efficiency of load transfer, thereby contributing to the overall elastic response of the alloy [41]. The shape parameter ξ changes from 2.61 for the isolated Al_3Ni particles at low Ni contents to 1.72 and 1.92 for the cloud-like and needle-like mixed structures, respectively (Table 1). However, the Halpin-Tsai model, which accounts only for the particle morphology, underestimates the measured E at high Ni contents, indicating that the connectivity of the IMC aggregates provides an additional contribution to the stiffening beyond the morphological effect. An additional important observation is the distinct slope changes in composition-modulus relation near 20 and 24 wt% Ni, corresponding to the appearance of $\text{Al}_6\text{Ni}_3\text{Si}$ and Al_3Ni_2 (See Figs. 1 and 2). It should be noted that the modulus increase becomes less pronounced at high cooling rate after Al_3Ni_2 formation despite the high elastic modulus of Al_3Ni_2 . As discussed in our recent study [12], rapid solidification suppresses the diffusion-controlled transformation of Al_3Ni_2 into $\text{Al}_6\text{Ni}_3\text{Si}$ ($\text{Al}_3\text{Ni}_2 + \text{Liq.} \rightarrow \text{Al}_6\text{Ni}_3\text{Si} + \text{Si}$), which alters the formation and distribution of $\text{Al}_6\text{Ni}_3\text{Si}$ around Al_3Ni_2 . As a result, the fractions of $\text{Al}_6\text{Ni}_3\text{Si}$ and Al_3Ni_2 at high Ni contents are higher, and that of Al_3Ni is lower, at 25 K/s than at 5 K/s (Fig. 2). Nevertheless, the measured E is lower at 25 K/s, indicating that the cooling-rate dependence of E cannot be explained solely by the phase fractions. Although the retained Al_3Ni_2 contributes to overall stiffness, the associated changes in the morphology and connectivity of the Ni-rich IMCs can partially offset this beneficial effect and consequently alter the load-sharing behavior within the alloy.

Exact quantitative agreement between measurement and prediction is not expected. The composite models employed in this study assume isotropic constituent phases, perfect interfacial bonding, and homogeneous strain/stress fields. These assumptions are not fully satisfied in Al–Si–Ni cast alloys because of complex phase morphologies, imperfect interfaces, crystallographic anisotropy, and residual porosity, which may cause the measured values to fall below the V-R lower bound (Reuss

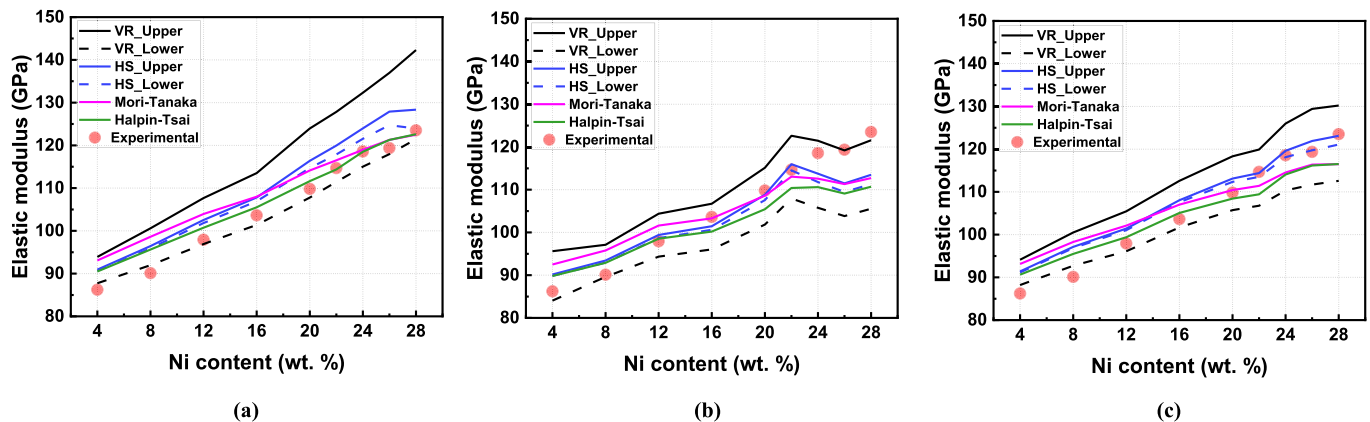


Fig. 8. Comparison between experimentally measured and model-predicted elastic modulus of Al-18Si-xNi alloys solidified at 5 K/s using phase fractions obtained from (a) Scheil simulations, (b) XRD, and (c) EBSD.

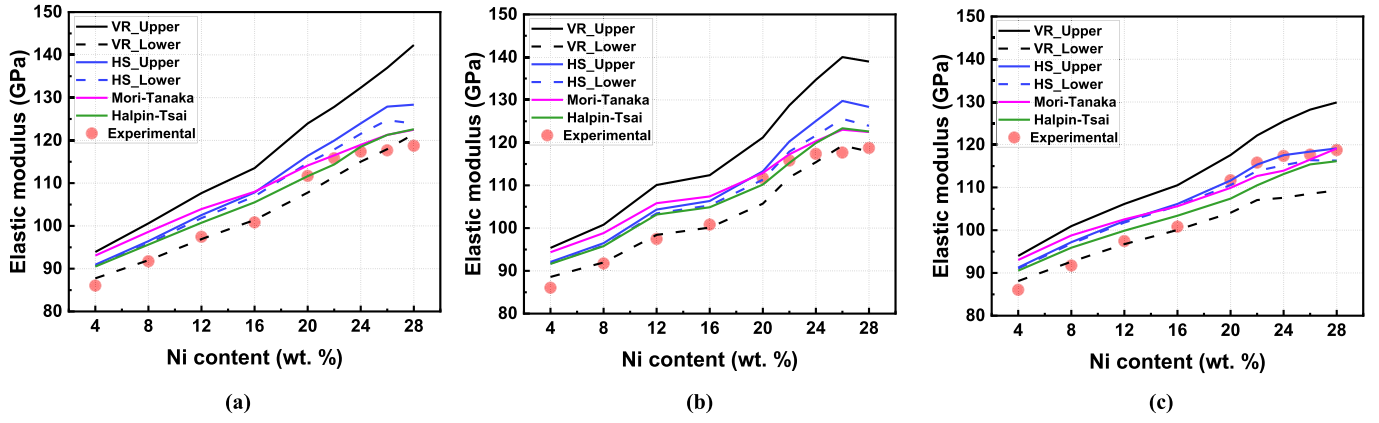


Fig. 9. Comparison between experimentally measured and model-predicted elastic modulus of Al-18Si-xNi alloys solidified at 25 K/s using phase fractions obtained from (a) Scheil simulations, (b) XRD, and (c) EBSD.

model) [50]. The elastic modulus obtained by impulse excitation technique (IET) was calculated using apparent rather than density of the specimens, and the modulus assigned to eutectic Si also involves additional uncertainty from the rule of mixtures estimate and from the intrinsic elastic anisotropy of Si [51]. More fundamentally, however, all models employed here describes only matrix-reinforcement interaction, whereas in the alloys with high Ni content, Al_3Ni , $\text{Al}_6\text{Ni}_3\text{Si}$, and Al_3Ni_2 coexist as layered particles where stress is transmitted through direct IMC-IMC contacts. The residual deviation at high Ni is thus not merely experimentally scatter but a signature of a network effect that matrix-inclusion models cannot clearly account for. Overall, the results demonstrate that the macroscopic elastic response of highly alloyed Al-Si-Ni casting alloys is controlled not simply by IMC fraction, but more importantly by the interaction, connectivity, and solidification-dependent evolution of Ni-rich intermetallic compounds.

4.2. Effect of Ni-rich IMC evolution on thermal expansion behavior

Like the elastic modulus, the CTE of the Al-18Si-xNi alloys is influenced not only by the volume fraction of constituent phases but also by the elastic interactions that develop during thermal expansion. To understand the governing mechanisms, the experimentally measured CTE values were compared with theoretical predictions obtained using the Voigt-Reuss (V-R), Turner, and Kerner models [42]. (Tables 2 and 3).

The V-R model averages the phase CTEs by volume fraction and neglects inter-phase constraint, thereby overestimating the thermal expansion of multiphase alloys. The Turner model incorporates the bulk modulus of each constituent phase to account for elastic constraint

during volumetric deformation, and the Kerner model further considers shear stresses generated by CTE mismatch between phases.

For the alloy solidified at 5 K/s (Fig. 10), the Turner and Kerner models provide the closest agreement with the experimental CTE values, particularly when experimentally determined phase fractions obtained from XRD and EBSD are employed (Fig. 10(b and c)). In contrast, predictions based on Scheil phase fractions (Fig. 10(a)) deviate increasingly at high Ni, indicating that the actual thermal expansion behavior is influenced by microstructural factors beyond the thermodynamic phase fractions predicted by the Scheil simulation.

A similar tendency is observed for the alloy solidified at 25 K/s (Fig. 11). Although the overall compositional dependence remains similar, the experimental values show slightly better agreement with the Turner and Kerner models over a wide composition range. This improvement is particularly evident for the XRD- and EBSD-based predictions (Fig. 11(b and c)), indicating that the actual phase evolution with rapid solidification is more accurately reflected by experimentally measured phases fractions than by Scheil simulations. This suggests that the CTE behavior is controlled by the actual solidification microstructure rather than by thermodynamic phase fractions.

The observed transition can be understood from both microstructural and model-related perspectives. At low Ni contents, Al_3Ni is the dominant intermetallic phase, and its strong crystallographic anisotropy (as evidenced in Fig. 7 and Table 3) introduces deviations from the isotropic assumptions adopted in the prediction model. As a result, the experimental CTE values remain relatively close to the V-R bounds. With increasing Ni content, however, the formation of $\text{Al}_6\text{Ni}_3\text{Si}$ and Al_3Ni_2 , both of which have far lower intrinsic CTEs than Al_3Ni , sharply

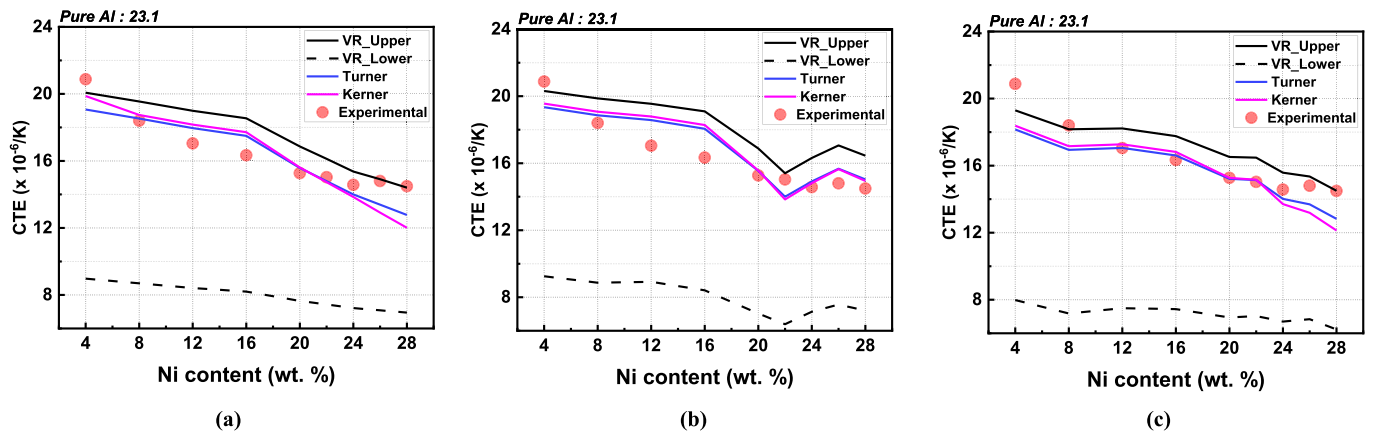


Fig. 10. Comparison between experimentally measured and model-predicted CTE of Al-18Si-xNi alloys solidified at 5 K/s using phase fractions obtained from (a) Scheil simulations, (b) XRD, and (c) EBSD.

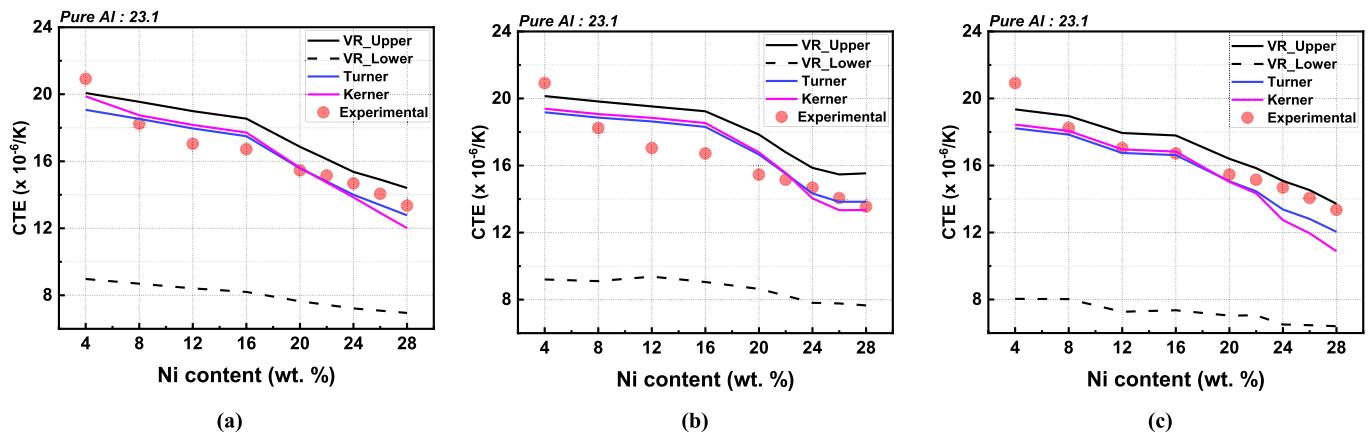


Fig. 11. Comparison between experimentally measured and model-predicted CTE of Al-18Si-xNi alloys solidified at 25 K/s using phase fractions obtained from (a) Scheil simulations, (b) XRD, and (c) EBSD.

decreases the measured CTE and progressively improved agreement with the Turner and Kerner models. As the multiphase microstructure develops, phases with relatively high CTE, such as Al_3Ni , can no longer expand freely because they are mechanically constrained by surrounding low-CTE phases, particularly $\text{Al}_6\text{Ni}_3\text{Si}$ and Al_3Ni_2 . This thermoelastic interaction enables the Turner and Kerner models to reproduce the experimentally observed CTE reduction more accurately than the V-R model. Furthermore, Al_3Ni_2 exhibits the highest bulk modulus (91.15 GPa) among the constituent phases, thereby providing stronger mechanical constraint on the surrounding matrix during thermal expansion. This characteristic further enhances its contribution to the overall reduction in alloy CTE despite its phase fraction being lower than that of Al_3Ni .

Cooling rate further modifies this behavior through its influence on phase evolution. As discussed previously [12], rapid solidification suppresses the diffusion-controlled transformation of Al_3Ni_2 into $\text{Al}_6\text{Ni}_3\text{Si}$, promoting the retention of Al_3Ni_2 while reducing the fraction of Al_3Ni . Although Al_3Ni_2 exhibits a CTE comparable to that of $\text{Al}_6\text{Ni}_3\text{Si}$, its bulk modulus is approximately 1.5 times higher (186.63 vs. 121.88 GPa), and it therefore imposes a stronger elastic constraint on the surrounding phase during thermal expansion. In addition, the reduced fraction of Al_3Ni , which has the highest CTE among the Ni-rich IMCs, further lowers the CTE. These combined effects result in a lower CTE for the high-Ni alloys solidified at 25 K/s.

5. Conclusion

In this study, the effects of Ni-rich IMC evolution on the elastic modulus (E) and CTE of hypereutectic Al-18Si-xNi casting alloys were systematically investigated under cooling rates of 5 and 25 K/s. By combining phase-specific property measurements with composite-model analysis, the relationships between solidification microstructure and physical properties were clarified. The main conclusions are summarized as follows:

1. With increasing Ni addition, the microstructural evolution Ni-rich IMCs occurs along with an increase in the fraction. Al_3Ni phase is the dominant IMC in the range of 4–16 wt% Ni; $\text{Al}_6\text{Ni}_3\text{Si}$ appears above 20 wt% Ni, and phase newly appears, and Al_3Ni_2 above 24 wt %. The cooling rate modifies the phase constitution by suppressing the transformation of Al_3Ni_2 into $\text{Al}_6\text{Ni}_3\text{Si}$. As a result, Al_3Ni_2 is retained while the fraction of Al_3Ni is reduced, demonstrating that the physical properties depend strongly on the solidification.
2. Nanoindentation and high-temperature XRD measurements showed that a modulus and CTE Al_3Ni_2 has the highest elastic modulus (235 GPa) and a low CTE ($7.7 \times 10^{-6}/\text{K}$), while $\text{Al}_6\text{Ni}_3\text{Si}$ also exhibits

high stiffness (190 GPa) and a similarly low CTE ($7.4 \times 10^{-6}/\text{K}$). The sequential formation of these IMCs is therefore responsible for the increased stiffness and reduced thermal expansion of the alloys. These findings provide a basis for tailoring the elastic modulus and CTE of Al-Si alloys through the controlled formation and retention of Ni-rich IMCs by adjusting the Ni content and cooling rate.

3. With increasing Ni content, the elastic response progressively shifts from the Voigt-Reuss lower bound toward the Hashin-Shtrikman and Mori-Tanaka predictions, indicating a transition from matrix-dominated deformation to cooperative load sharing among interconnected IMCs. In contrast, the CTE is best described by the constraint-based Turner and Kerner models, highlighting the increasing importance of interphase elastic constraint during thermal expansion. These results demonstrate that the connectivity, interaction, and cooling-rate-dependent retention of Ni-rich IMCs control the physical properties of the high-Ni containing Al-Si casting alloys.

CRediT authorship contribution statement

Yoon-Ho Lee: Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Formal analysis, Data curation. **Jung-Moo Lee:** Writing – review & editing, Validation. **Sang-Ik Lee:** Investigation, Formal analysis, Data curation. **Jae-il Jang:** Writing – review & editing, Validation. **Young-Hee Cho:** Writing – review & editing, Writing – original draft, Validation, Supervision, Project administration, Methodology, Funding acquisition, Conceptualization.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.msea.2026.151194>.

Data availability

Data will be made available on request.

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