Development of new Al-Cu-Si alloys for high temperature performance

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Abstract

In a quest for developing new lightweight metal alloys that can perform excellently at elevated-temperatures (from 300°C to 400 °C), a ternary eutectic Al-Cu-Si alloy was exploited to gain a deeper understanding of the alloy system and its suitability for high temperature applications. The studied alloys, with chemical composition of Al-27%Cu-5%Si (by weight percent) with Ni addition in the range of 0 to 1.5%wt, were cast in a rapid solidification casting technique. The solidification characteristics of the alloy was studied using the Thermo-Calc software. Microstructures were characterized in a scanning electron microscope coupled with energy dispersive spectrometry (SEM-EDS). Finally, the elevated-temperature tensile properties of the alloys were investigated. Comparing the microstructures and mechanical properties of the Al-Cu-Si(-Ni) alloys with conventional A319 Al- alloy, the refined microstructure with dispersed Ni intermetallic particles formed in the as-cast Al-Cu-Si(-Ni) alloys delivers improved elevated temperature properties. In particular, the yield strength and ultimate tensile strength of the new alloy with 1.5% Ni at 400˚C were observed to be 220% and 309% higher, respectively, than for conventional A319 reference alloy. Copyright © 2017 VBRI Press.

Keywords: Eutectic composite structure, casting, solidification, elevated-temperature performance, thermo-calc, tensile properties.

Introduction

In spite of several developmental advances in Al-Si alloys, the existing alloys cannot still meet the requirements placed on heat resistance and high temperature performance of cast components [**1**]. This has led to several research efforts [**1-4**] in recent years to proffer solutions to the challenges. Yang *et al.* [**3**, **5**] performed studies on eutectic and near-eutectic Al-Si alloys with the aim of improving the mechanical properties through addition of different alloying elements such as copper, nickel and magnesium, which form various intermetallic phases with a complex morphological structure. These intermetallic phases (e.g. Mg_2Si, Al_2Cu, Al_3Ni, Al_3CuNi and Al_7Cu_4Ni) have contributed substantially to the excellent properties of these alloys. The improved properties of the alloys include good corrosion resistance and abrasion, high strength-to-weight ratio and low coefficient of thermal expansion. The addition of transition alloying elements, like copper and nickel, is the most practical and efficient technique of enhancing the mechanical properties of Al-Si alloys, particularly the elevated temperature performance of piston alloys. The effect of Si, Cu, Mg and Ni on the micro-hardness of Al-12%Si-4%Cu-2%Ni-2.6%Mg piston alloy has been investigated [**5**], and the results indicate that solute in solid solution in the α-Al matrix provides a greater contribution to micro-hardness than the formation of intermetallics. Moreover, the mechanical and physical properties of Al-Si piston alloys strongly depend on the types, morphologies and distributions of second phases [**2**]. Nickel is recognized as the most effective element in improving the elevated-temperature properties of Al–Si piston alloys [**2**, **6**]. Li *et al.* [**2**] have shown that the presence of Al_3Ni, Al_3CuNi and Al_7Cu_4Ni phases have much bigger contributions to the elevated temperature properties of Al–Si piston alloys due to their better thermal stability, mechanical properties, morphologies and distributions. For instance, if nickel is added to Al-Si alloys, a much finer dispersion of nickel intermetallic particles or a much higher volume fraction of the coarse compound would be required to achieve efficient strengthening of the alloys [**7**]. Yang *et al.* [**3**] also concluded that, with the increasing of Cu content, Ni-phases translate from Al_3Ni (ε-phase) or Al_3CuNi (δ-phase) to Al_7Cu_4Ni (γ-phase), and their morphologies change from short-strip to reticular and then annular or semi-annular shape, which consequently have a great effect on the mechanical properties. In a related study, Shaha *et al.* [**4**] investigated the microstructure and tensile properties of Al-7%Si-1%Cu-0.5%Mg cast alloy with additions of Ti, V and Zr at temperatures up to 300°C and compared with those of the commercial A380 grade. It is observed that the microstructure of both alloys consisted of Al dendrites surrounded by Al-Si eutectic containing within its structure, the ternary Al-Al_2Cu-Si phase. Due to the chemistry modification of Al-7%Si-1%Cu-0.5%Mg cast alloy, its strength was higher by 20%-40% and ductility higher by 1.5-5 times than the A380 reference grade.

Eutectic Al-Cu-Si system may offer a great possibility to significantly enhance the mechanical properties of this alloy system since eutectic alloy solidifies at a constant temperature which makes it easy to process. By reducing the eutectic spacing, the mechanical properties of eutectic alloys are improved. Moreover, various researchers [**2**, **5**, **8**, **9**] have concluded that eutectic and near-eutectic Al-Si alloys possess the excellent abrasion and corrosion resistance, the low coefficient of thermal expansion and high elevated temperature strength, which has resulted to a wide range of applications of Al-Si alloys in the automobile sector. Cast eutectic Al-Si alloy has been widely employed in the manufacture of piston for petrol engines due to its high wear resistance, low expansion and low density [**10**]. However, most of the Al-Si alloys are unsuitable for elevated temperature applications since their tensile and fatigue strengths are usually lowered than desired strength at a temperature range of 250°C - 400°C. Above the temperature of 250°C, the alloy's microstructure strengthening mechanism becomes unstable, rapidly coarsen and solubilize, which make these alloys to possess undesirable microstructure for high temperature applications. As a consequence, the alloy lacks the coherency between the Al solid solution lattice and the precipitated strengthening particles and therefore reduce the possibility to use this alloy at elevated temperature [**11**]. For instance, the tensile strength of A319-Al alloy, one of the most commonly used heat resistant Al alloy, increases slightly to the highest value of 225 MPa at around 200°C, but when the temperature rises to 300°C and 400°C , the tensile strength decreases drastically to 90.8 MPa and 25.6 MPa, respectively [**12**]. This behavior of the alloy would make it undesirable for the components serving at a temperature higher than 250°C.

In a recent study, Park *et al.* [**13**, **14**] developed alloys and used different casting methods that could generate highly refined eutectic microstructures in the casting. A bulk ultrafine eutectic structure was obtained in as-cast Al-27%Cu-6%Si ternary alloy at cooling rates ranging from 10^2 to 10^3 K/s, yielding eutectic spacing of 300-700 nm in the material. The microstructures were comprised of cellular eutectic (α-Al + Al_2Cu) in a nanocrystalline matrix and exhibited high-strength with high-plasticity. The mechanical properties of these alloys at ambient temperature were determined by a compression testing technique and a high strength of 1.1±0.1 GPa as well as high plasticity of 11±2% were obtained. However, the elevated-temperature mechanical properties of the alloy were not reported.

The aim of the present study is to develop a high-temperature performance eutectic Al-Cu-Si(-Ni) alloy suitable for components serving at a temperature greater than 250˚C. Ternary eutectic Al-Cu-Si alloy with the chemical composition of Al-27%Cu-5%Si with minor additions of Ni (0-1.5%wt) is studied. This alloy is produced through a high cooling rate casting technique. The solidification behaviour of the alloy was examined by Thermo-Calc calculations. In addition, the microstructures of the alloys were characterized in a scanning electron microscope coupled with an energy dispersive spectrometer (SEM-EDS). Finally, the effect of the microstructures on the mechanical properties of the alloy at elevated temperatures is discussed and compared with the conventional A319-Al alloy.

Novelty Statement: A revolutionary eutectic Al-Cu-Si(-Ni) cast alloy was designed and produced through a rapid solidification casting method. The as-cast alloy possesses a unique microstructure that significantly enhances its elevated-temperature tensile properties, which are superior to that of the commercial heat-resistant Al-A319 reference alloy.

Table 1. Chemical composition of the alloys (wt%).

Alloy	Si	Cu	Ni	Fe	Al
1	5.43	27.47	0	0.11	Balance
2	5.04	26.75	0.78	0.09	Balance
3	5.21	26.07	1.05	0.10	Balance
4	4.97	26.63	1.50	0.10	Balance

Experimental

Material preparation and casting

The eutectic Al–27%Cu–5%Si-x%Ni (x = 0 - 1.5%wt.) alloys were prepared in a batch-wise manner by melting and mixing a pure aluminum ingot (99.95% purity), Cu chips (99.9% purity), and Al-50%Si and Al-20%Ni master alloys in a graphite crucible at 730°C. Alloys with 0%, 0.7%, 1.0% and 1.5% Ni were produced. The melt was prepared under a controlled atmosphere to prevent oxidation, and was thoroughly stirred to ensure melt homogeneity. The molten alloy was skimmed and poured into a copper mould to obtain 16 cm long cast rods with 9 mm diameter. The chemical composition (in weight percent) of each batch of as-cast Al-27%Cu-5%Si-x%Ni eutectic alloys was determined in a spectrometer (Spectromax CCD LMXM3) and are shown in **Table 1**. The remelting of the cast rods were conducted in an electrical resistance furnace. Prior to remelting, the rods were inserted into 20 cm long steel tubes with 10 mm inner diameter which were coated with boron nitride on the inside to prevent iron diffusion into the melt during remelting. The samples were held for 30 minutes at 680°C and then quenched directly in a cold water-ice mixture. The steel tubes were cut open to remove the solidified alloys for microstructural characterization and mechanical testing. Thermo-Calc 3.0.1 software was applied to investigate the influence of Ni additions on the solidification behaviour of ternary Al-27%Cu-5%Si(-Ni) alloy using the TCAL2 database (TCS Al-based alloys database version 2) [**15**].

Microstructural characterization and mechanical testing

Samples were taken from the cast alloys and polished for microstructural investigation using standard metallographic procedures. The microstructures were studied in a scanning electron microscope (JEOL JSM-7001F) equipped with energy dispersive spectroscopy (SEM-EDS).

The mechanical properties of the cast alloys at elevated temperatures (300°C and 400°C) were evaluated by tensile testing. The tensile test samples had a gauge length of 25 mm and a diameter of 6 mm according to ASTM-E8 standard. The tests were carried out in a Zwick/Roell Z100 testing machine at a strain rate of $10^{-3}s^{-1}$. An axial 25 mm clip-on extensometer was attached to the testing specimen at the gauge section. Specimens were heated to the desired temperature in a high temperature furnace and held at the test temperature for 15 minutes prior to testing. Two samples were tested for each alloy at each temperature.

Results and discussion

Phase evolution from thermodynamic calculations

Fig. 1 shows the Thermo-calc results for the Al-27% Cu-5%Si alloy with (a) 0% Ni, (b) 0.7% Ni, (c) 1.0% Ni and (d) 1.5% Ni, respectively. The figure depicts the mole fraction of the different phases as a function of temperature during solidification of the alloys as computed by the Thermo-Calc software. The thermodynamic calculation for the 0% Ni alloy (**Fig. 1a**) shows that primary Al_2Cu intermetallic phase starts forming at 527.6°C and continues to grow until a maximum of 1.57% solid fraction is obtained at 525.9°C. At this temperature, the binary eutectic reaction starts with L → α-Al + Al_2Cu. The binary eutectic reaction proceeds until the temperature has decreased to 521.7°C, giving of a maximum solid fraction of about 18.6%. At 521.7°C, the solidification of the alloy is completed with a ternary eutectic reaction - L → α-Al + Al_2Cu + β-Si - and the remaining 81.4% liquid solidifies completely.

For the other alloys, which all contain Ni addition, it is noted from **Fig. 1(b-d)** that a Ni-rich intermetallic phase (Al_7Cu_4Ni) forms first from the liquid metal. The precipitation temperature of the Al_7Cu_4Ni phase is dependent on the Ni concentration. The temperatures at which the Al_7Cu_4Ni intermetallic begins to form at 0.7%, 1.0% and 1.5% Ni are 546.9°C, 556.9°C and 572.8°C, respectively.

This implies that increasing Ni content in the eutectic Al-27%Cu-5%Si alloy raises the liquidus temperature of the alloy. For the alloy with 0.7% Ni, **Fig. 1(b)** demonstrates that the Al_7Cu_4Ni intermetallic phase starts to grow at 546.9°C and continues until a temperature of 531.5°C is reached, after which it begins to transform in a peritectic reaction to Al_2Cu intermetallic according to the peritectic reaction: L + Al_7Cu_4Ni → Al_2Cu. However, the last liquid solidifies in ternary reaction at 523.5°C according to L → α-Al + Al_2Cu + β-Si. The alloy containing 1.0% Ni solidifies in a similar way as shown in **Fig. 1(c).** It is noticed from the figure that the Al_7Cu_4Ni phase first precipitates at 556.9°C, and this Ni-rich phase transforms to Al_2Cu intermetallic in a peritectic reaction at 528.9°C. The final liquid solidifies in a ternary eutectic reaction like the alloy with 0.7% Ni.

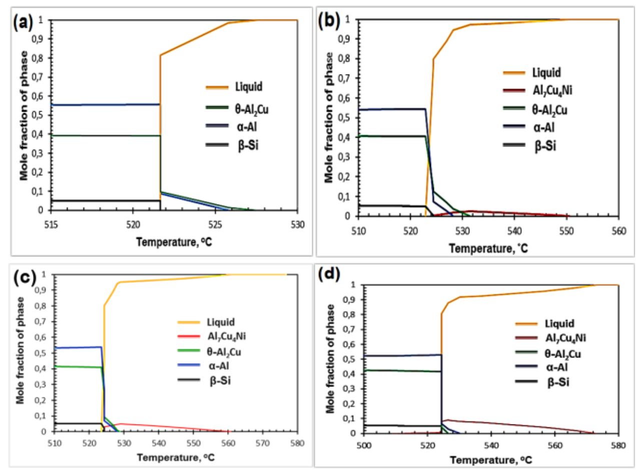

Fig. 1. Thermo-Calc calculation of the solidification sequence for Al-27%Cu-5%Si with (a) 0%Ni, (b) 0.7%Ni, (c) 1.0%Ni and (d) 1.5%Ni.

In the Al-27%Cu-5%Si-1.5%Ni alloy **(Fig. 1d),** the precipitation of Al_7Cu_4Ni commences at 572.8°C and continues to grow without a peritectic transformation. At 530.1°C and 526.5°C, α-Al and Al_2Cu are respectively formed from the remaining liquid and the final liquid freezes in a quaternary eutectic reaction at 524.4°C: L → α-Al + θ-Al_2Cu + β-Si + Al_7Cu_4Ni. Below 516.9°C, the quaternary phase transforms into ternary eutectic phase like the other Ni-containing alloys.

In summary, it is observed from this study that increasing Ni content in the Al-27%Cu-5%Si alloy decreases the amount of binary eutectic that forms. Instead, a Ni-rich intermetallic phase (Al_7Cu_4Ni) forms first during solidification of the Ni-containing Al-Cu-Si alloys.

Microstructural analysis

The microstructures of the alloys were examined using SEM-EDS method and are shown in **Figs. 2** and **3**. In the figures, the dark contrast structure is the α-Al phase; the bright contrast represents θ-Al_2Cu intermetallic phase while the gray bulky reticular or annular structure is the Ni-containing phase (Al_7Cu_4Ni). A bimodal eutectic composite structure was developed in the as-cast alloys. This is visible by closely examining the SEM micrograph of the 0% Ni alloy **(Fig. 2a).** The binary eutectic (α-Al + θ-Al_2Cu) cells are seen to be surrounded by the ternary eutectic (α-Al + θ-Al_2Cu + β-Si) matrix structure, which vividly agrees with the Thermo-Calc calculation results. The inset picture in **Fig. 2(a)** obtained at higher magnification evidently depicts the bimodal eutectic structure consisting of cellular binary eutectic and very fine ternary eutectic matrix. The composition analyses of these alloys have shown that the alloys are composed of three distinct phases namely: α-Al, θ-Al_2Cu and β-Si.

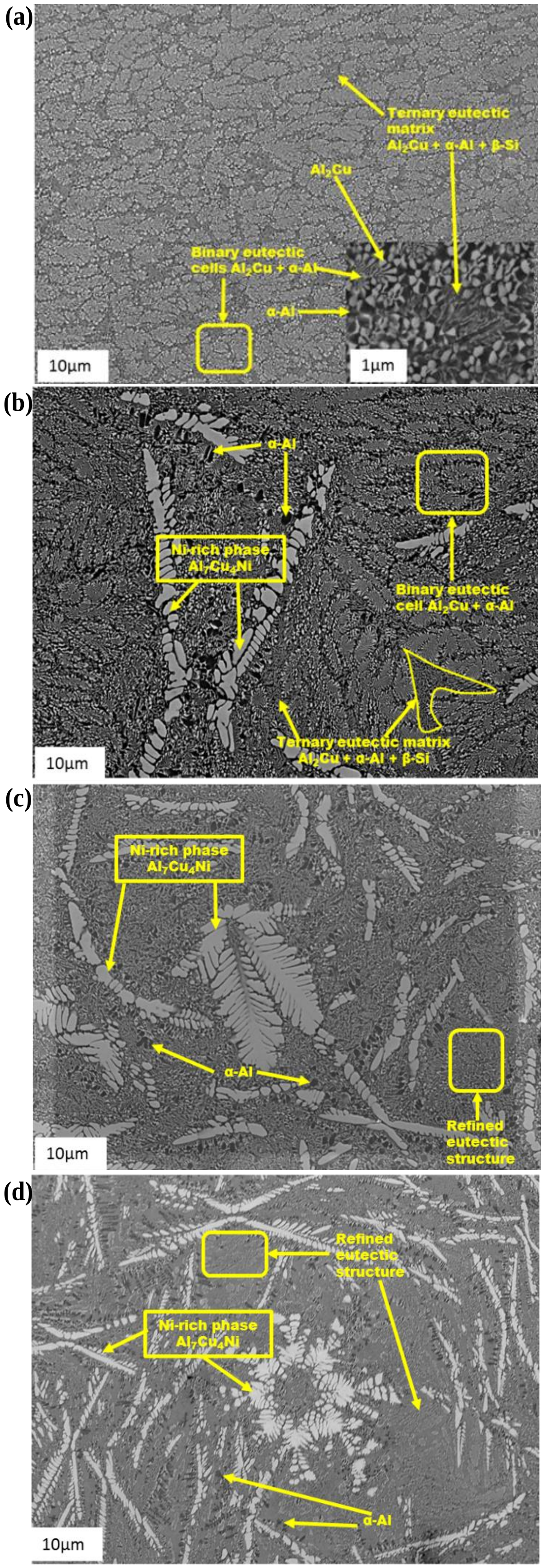

Fig. 2. SEM micrographs of as-cast Al-27%Cu-5%Si alloys with (a) 0% Ni, (b) 0.7% Ni, (c) 1.0% Ni and (d) 1.5% Ni.

However, it is nearly impossible to differentiate between the α-Al and β-Si phases in the SEM images because both phases show a dark colour in the SEM micrographs, which is probably due to their relatively similar atomic weights. The SEM-EDS mapping results shown in **Fig. 3** demonstrates that Si is present as fine particles in the ternary eutectic matrix. The Si crystals are found to be uniformly distributed in the region surrounding the binary eutectic colonies.

The θ-Al_2Cu phase shows a bright contrast in the images in **Fig. 2**. It is apparent from the SEM micrographs presented in **Fig. 2** that the alloys containing Ni additions exhibit a significantly different morphology compared to the base alloy with no Ni addition. Ni-rich (Al_7Cu_4Ni) intermetallic phases are observed in addition to the binary and ternary eutectic structures seen in the base alloy. The Al_7Cu_4Ni intermetallic phase is bulky in nature and exhibits two different morphologies – one with a dendritic structure and the other looks like a broad flat leaf. The volume fraction of Al_7Cu_4Ni intermetallic phase is observed to increase with increasing Ni content.

It is also noticed in **Fig. 2** that addition of Ni to the base alloy composition has a profound refining effect on the eutectic structures. It is worth noting as well from the micrographs that addition of Ni leads to the formation of a dendritic α-Al phase in the alloys (**Fig. 2(b-c)**). In general, the microstructures of the alloys reveal a bimodal distribution of both binary and ternary eutectic phases which is also consistent with the thermodynamic calculations (**Fig. 1**). The SEM-EDS analysis of the alloys depicts a cellular eutectic structure of α-Al and Al_2Cu phases intermingled together while the ternary eutectic structure represents the main matrix, which is enriched in Si. Moreover, **Fig. 3a** displays the SEM-EDS images with x-ray spectra taken from various spots as highlighted in the figure.

The bright blocky dendritic structure was identified as $Al_{68.3}Cu_{30.7}Ni_{0.6}Si_{0.5}$, the dark phase was recognized as $Al_{94.7}Cu_{3.9}Si_{1.4}$ which is enriched in Al while the bright phase enriched in both Al and Cu was identified as $Al_{77.9}Cu_{19.4}Si_{1.8}$. Based on the SEM-EDS results, the various phases identified in the studied alloys can be recognized as, (i) the bright blocky structure is the Al_7Cu_4Ni intermetallic phase, (ii) the dark contrast represents the α-Al phase, and (iii) the bright alternating structure represents the primary θ-Al_2Cu intermetallic phase. Si crystals were not distinctly observed in the alloys due to their fineness, but are present as fine particles in the ternary eutectic matrix **(Fig. 3b)**.

Mechanical properties

The tensile properties of the as-cast alloys were determined at temperatures of 300˚C and 400˚C, and the results are shown in **Figs. 4-6**. The results are the mean values based on the duplicate measurements. It is observed that the alloys exhibit excellent mechanical properties at elevated temperatures (300˚C and 400˚C). For instance, the yield strength (YS), ultimate tensile strength (UTS) and the elongation (EL) of the base alloy (no Ni addition) at 300˚C reach 96 MPa, 117 MPa and 10.3%, respectively.

(a)

(b)

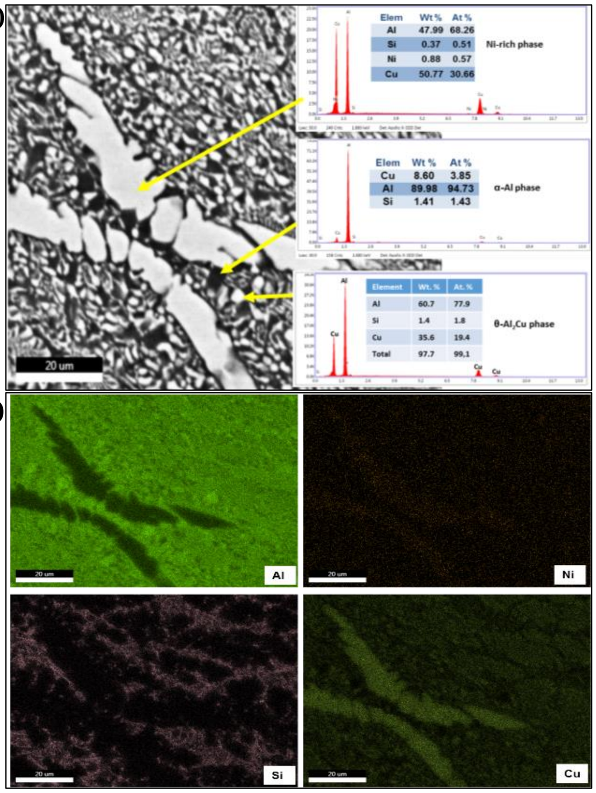

Fig. 3. SEM-EDS analysis of as-cast Al-27%Cu-5%Si-0.7%Ni alloy showing Ni-rich phase, α-Al phase and θ-Al_2Cu phase.

The influence of Ni additions on the elevated tensile properties of the alloy can be noticed in **Figs. 4-6** at 300˚C. With increasing Ni content from 0.7% to 1.5%, the yield strength of the alloy increases from 98 MPa to 267 MPa **(Fig. 4),** while the ultimate strength of the alloys is also increased from 208 MPa to 272 MPa **(Fig. 5),** but the elongation of the alloys decreases from 12.3% to 4% **(Fig. 6).** The enhanced mechanical properties experienced in the Ni-containing alloys at 300˚C is probably due to the presence of the Ni-rich intermetallic phase as observed in **Fig. 1(c-d)**. This observation however supports previous findings that addition of Ni to Al-Si piston alloys improves the elevated-temperature mechanical properties of the alloys, owing to the better thermal stability of the Ni-rich intermetallic phases (such as Al_3Ni, Al_3CuNi and Al_7Cu_4Ni) which are precipitated in the alloys [**2**, **6**].

By comparing the mechanical responses of the alloys at both 300˚C and 400˚C, it is noticed that the tensile properties of the alloys at 400˚C decrease generally compared with the responses at 300˚C, especially for the alloys with 1.0 and 1.5% Ni. For the 1.5% Ni alloy, the yield strength and the ultimate tensile strength decrease by 70% and 62%, respectively, when the testing temperature increases from 300˚C to 400˚C **(Figs. 4-5).** However, the elongation of this alloy increases from 4% at 300˚C to 7% at 400˚C **(Fig. 6).** Despite the decrease in the mechanical properties of the studied alloys at 400˚C, the elevated-temperature properties of these alloys are still superior to the conventional Al-Si alloys.

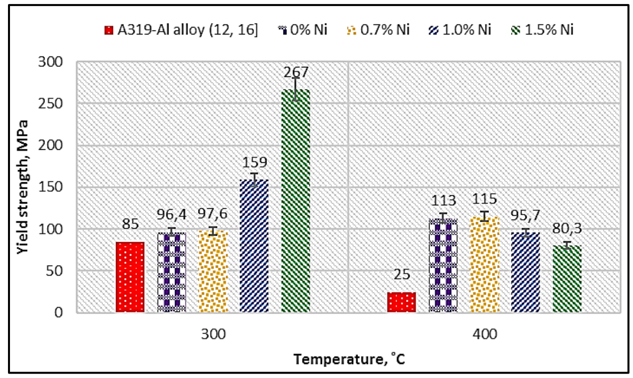

Fig. 4. Comparison of yield strengths of the present and conventional A319-Al [**12**, **16**] alloys at elevated temperatures.

Moreover, to compare the high-temperature mechanical properties of the newly developed heat-resistant Al alloys studied in this investigation with the commercially available Al-Si alloys, the results from a study of the elevated-temperature tensile properties of A319 alloy [12,16] are compared with the tensile properties of the present alloys and provided as **Figs. 4-6.** By comparing the tensile properties of A319 with that of the 1.5% Ni alloy, **Figs. 4-5** reveal that the yield strength and ultimate tensile strength of the 1.5% Ni alloy at 300˚C are higher than for alloy A319 by 214% and 199%, respectively. At 400˚C, the yield and ultimate strengths of the 1.5% Ni alloy are 220% and 309% higher, respectively, than for the conventional A319 **alloy (Table 2).** In **Fig. 6,** it can be observed that the ductility of A319 is higher than that of the 1.5% Ni alloy. However, the ductility of this alloy at 400˚C is still satisfactory.

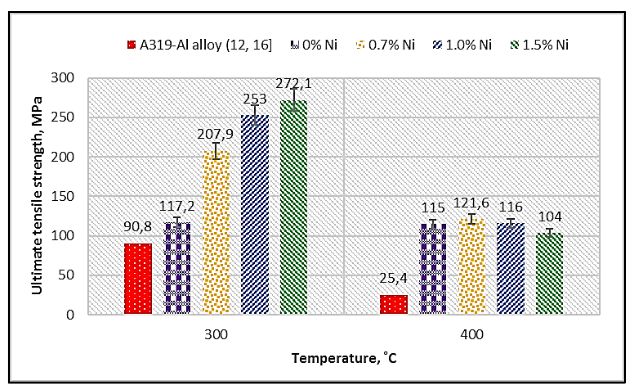

Fig. 5. Comparison of tensile strengths of the present and conventional A319-Al [**12**, **16**] alloys at elevated temperatures.

It can be concluded from this investigation based on the experimental results, that the newly developed heat-resistant Al-Cu-Si eutectic cast alloy has superior elevated-temperature tensile properties compared to the conventional A319 alloy **(Table 2).** Therefore, the developed alloys could be a potential candidate material for the design of components serving at a temperature up to 400˚C. The possible components where the newly developed alloys could be used may include pistons, engine blocks and cylinders, connecting rods, cylinder liners, turbochargers etc.

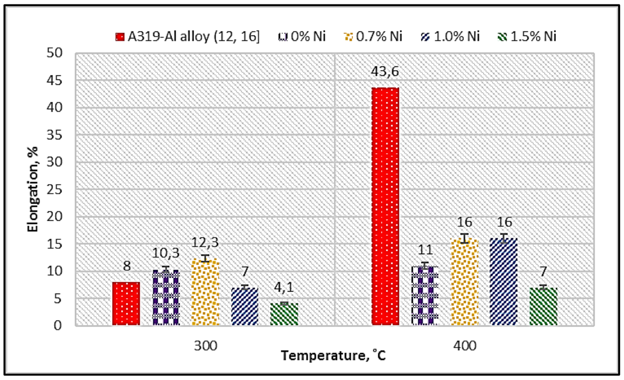

Fig. 6. Comparison of elongation of the present and conventional A319-Al [**12**, **16**] alloys at elevated temperatures.

Table 2. Tensile properties of the present and previously reported Al-alloys.

Alloys	Temp. (˚C)	Tensile properties UTS (MPa)	YS (MPa)	EL (%)	Composition (wt.%)
This work	300	272 ± 4	267± 3	4± 0.3	Al-27Cu-5Si-1.5Ni
	400	104 ± 3	80 ± 4	7± 0.2	
Al-A319 [**12**, **16**]	150	230	155	0.4	Al-3.8Cu-8.6Si-0.5Fe-0.3Mn-0.05Cr-0.023Ni-0.015Pb-0.012Sr-0.013Ti
	240	165	149	0.3	
	300	91	85	8	
	400	25	25	43	

Conclusion

A novel Al-Cu-Si(-Ni) eutectic cast alloy has been designed and investigated to examine its suitability for elevated temperature applications. The alloy, with a chemical composition of Al-27%Cu-5%Si and minor additions of Ni, was produced through a rapid solidification casting technique. The evolution of various phases during the solidification process of the developed alloys was investigated using Thermo-Calc software. The results from the thermodynamic calculations reveal that θ-Al_2Cu intermetallic phase first precipitates out of the Al-27%Cu-5%Si base alloy, followed by a binary eutectic reaction (L → α-Al + θ-Al_2Cu) and with the last melt solidifying in a ternary eutectic (θ-Al_2Cu+α-Al+β-Si) structure. It is also observed that addition of Ni to the base alloy alters its solidification profile, and a Ni-rich phase (Al_7Cu_4Ni) is produced in the alloy which forms ahead of the eutectic front at a temperature greater than the eutectic temperature. SEM-EDS analysis of the cast alloys shows a bimodal eutectic composite microstructure in the Al-27%Cu-5%Si base alloy, consisting of binary (θ-Al_2Cu+α-Al) eutectic cells surrounded by the ternary (θ-Al_2Cu+α-Al+β-Si) eutectic matrix. The SEM characterization of the alloys containing Ni addition reveals that Al_7Cu_4Ni intermetallic was formed in the alloys in addition to the composite eutectic structure, which is in good correlation with the Thermo-Calc computations. The presence of the Al_7Cu_4Ni intermetallics contributes significantly to the elevated-temperature tensile properties of the alloy. A significant increase in high-temperature tensile strength of 272 MPa was achieved in cast Al-Cu-Si(-1.5%Ni) eutectic alloy at 300˚C compared to 91 MPa attained in the reference alloy A319 at the same temperature. The yield strength and the ultimate tensile strength of the alloy containing 1.5% Ni at 400˚C were observed to be 220% and 309% higher, respectively, than for conventional A319 reference alloy.

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Author's contributions

Conceived the plan: AD, YL, AJ, SS; Performed the expeirments: SA; Data analysis: SA, SS; Wrote the paper: SA. Authors have no competing financial interests.

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