Effect of multiple forging on the microstructure and properties of an as-cast Cu–Ni–Si alloy with high Ni and Si contents

Jinlong Zhang†, Zhenlin Lu†, Lei Jia†, Hui Xie, Xin Wei and Shiping Tao∗

1 School of Materials Engineering, Xi’an Aeronautical University, Xi’an 710077, People’s Republic of China
2 School of Materials Science and Engineering, Xi’an University of Technology, Xi’an 710084, People’s Republic of China
† Authors to whom any correspondence should be addressed.
E-mail: zjl24931@163.com and lhzl2002@xaut.edu.cn

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Abstract

A Cu–Ni–Si alloy with high Ni and Si contents was prepared by the traditional melting and casting method, and then multiple forging and ageing were conducted to investigate their effect on the microstructure and properties. The results show that reticular Ni31Si12 phases are located on the grain boundaries of the dendritic α-Cu(Ni, Si) solution matrix in the as-cast Cu–Ni–Si alloy because of the high Ni and Si contents, and some rice-like Ni2Si phases precipitate in the interior of α-Cu(Ni, Si) grains during cooling. With increasing number of forging passes, the morphology of the α-Cu(Ni, Si) matrix changes from dendirites to elongated dendrites and then equiaxed grains. The Ni31Si12 phase changes from reticular to irregular and then particle-like, while the Ni2Si phase gradually disappears. As a result, the hardness increases continuously up to 18 forging passes, while the electrical conductivity first increases and then decreases significantly. The hardness and electrical conductivity achieve the highest values with 18 forging passes and a subsequent ageing treatment at 450 °C for 4 h, and the corresponding microstructure comprises an equiaxed α-Cu(Ni, Si) matrix with microscale Ni31Si12 particles and sub-microscale Ni2Si precipitates.

1. Introduction

Copper and its alloys have been widely used owing to their excellent electrical and thermal conductivities, excellent ductility, high strength, and good corrosion resistance [1, 2]. In particular, high-strength and high-conductivity copper alloys have become indispensable core materials in high-tech fields and are widely used in various energised product parts [3]. With the rapid development of modern industry and the information industry in particular, energised products are becoming increasingly miniaturised and lightweight, and the requirements for electrical conductivity, thermal conductivity, and load-bearing properties are constantly increasing; consequently, it is urgent to upgrade copper alloys [4–6]. Among the series of developed alloys, Cu–Ni–Si alloys are considered some of the most promising high-strength and high-conductivity materials owing to their high strength and conductivity and good hot and cold workability [7–13].

Cu–Ni–Si alloys are strengthened by Fleisher strengthening and Orowan strengthening through the precipitation of Ni–Si phases [14–19]. Therefore, increasing the quantity of Ni–Si precipitates in the alloy is considered to be an effective way to improve the strength of Cu–Ni–Si alloys. For this reason, in recent years, some scholars [20–24] have attempted to increase the Ni and Si contents in the alloy and increase the number of precipitated phases, achieving good results. However, when the Ni and Si contents increase, a reticular phase easily forms in the alloy structure. The reticular phase not only hinders the movement of free electrons and reduces the electrical conductivity of the alloy, but also significantly reduces the mechanical properties of the alloy, greatly deteriorating the mechanical properties and electrical conductivity. There have been attempts to crush the Ni–Si reticular phase using the multiple forging method, which achieved good results [25]. However, many technological processes are required, resulting in long preparation cycles and high costs.
Therefore, in this study, Cu–Ni–Si alloys with high Ni and Si contents were used as the research object. Based on an in-depth analysis of the microstructure and phase composition of the as-cast alloy, combined with research on the hot deformation behaviour of the alloy [26], the homogenisation heat-treatment link of the original process was removed, and the hot deformation of the as-cast alloy was directly conducted by multiple forging. The microstructural evolution of the as-cast alloy during multiple forging was analysed, and the relationship between the number of forging deformation passes and the properties was established. The mechanism by which multiple forging improves the microstructure of the alloy was revealed, which provides a reference for the preparation of high-strength and high-conductivity Cu–Ni–Si alloys.

2. Materials and methods

A Cu–Ni–Si alloy with a nominal composition of 90 wt% Cu, 8.33 wt% Ni, and 1.67 wt% Si was prepared by melting and casting with a KPS-160/1-type intermediate frequency induction furnace, where the raw materials were pure Cu (99.9 wt%), pure Ni (99.9 wt%), and pure Si (99.99 wt%). The actual chemical composition of the as-cast alloy was measured using a SparkCCD 6500 optical emission spectrometer (OMS), and the result was 89.87 wt% Cu, 8.42 wt% Ni, and 1.73 wt% Si, which is close to the nominal composition. The ingot was cut into several rectangular specimens with a size of 18 × 14.7 × 12 mm³, and then multiple forging was conducted at 863 °C with a true strain (ε) of 0.4 for a single pass, which was determined according to previous work on the hot forging behaviour of Cu–Ni–Si alloy [25]. The sample was first heated at 863 °C for 5 min and then placed into a steel channel die with a height of 12 mm and width of 14.7 mm. Subsequently, the forging operation was performed until the height of the sample was reduced from 18 to 12 mm, after which the sample was removed from the die. This process, that is, heating the sample and forging the height from 18 to 12 mm, is referred to as one pass. This process was repeated multiple times to increase the number of forging passes. Subsequently, the forged Cu–Ni–Si alloy was subjected to isothermal ageing at 450 °C for 2, 4, and 6 h in an electrical resistance furnace and then cooled in air.

Samples for microstructural observation were taken from the middle of the forged specimens along the direction parallel to the forging direction. An Olympus GX51 inverted optical microscope (OM), JSM-6510A analytical scanning electron microscope (SEM), Tecnai G2 F20 S-TWIn transmission electron microscope (TEM), and Aeris X-ray diffraction (XRD) analyser were used to characterise the microstructure. The electrical conductivity was measured using a Sigma 2008C digital eddy current conductivity meter with a resolution of 0.01% IACS and a measurement accuracy of ±1%. The hardness was measured using a TMVS-1 automatic turret digital microscopic Vickers hardness tester.

3. Results and discussion

3.1. Microstructural characterisation of the as-cast alloy

Figure 1 shows the microstructure of the as-cast Cu–Ni–Si alloy observed under a low-power OM. The Cu–Ni–Si alloy microstructure was composed of well-developed dendritic grains (figure 1(a)) with different orientations presented by different colours. High-magnification observations indicated that there were interconnected reticular phases in the dendrite arm space, and some needle-like precipitates were distributed uniformly inside the dendrite grains (figure 1(c)). XRD analysis indicated that the microstructure of the as-cast Cu–Ni–Si alloy mainly consisted of three phases, where the highest peak intensity was observed for the α-Cu(Ni, Si) solid solution, and the remaining two phases were Ni₃₁Si₁₂ and Ni₂Si (figure 1(d)). According to the morphology and XRD analysis results of the Cu–Ni–Si alloy, the secondary phases in the as-cast Cu–Ni–Si alloy was Ni₃₁Si₁₂ and Ni₂Si, which were reticular at the grain boundary and granular in the grain, respectively. To further confirm these secondary phases with different morphologies and locations, TEM observation was necessary.

Figure 2 shows bright- and dark-field images and the selected area electron diffraction (SAED) patterns of the as-cast Cu–Ni–Si alloy. In figure 2(a), the width of the secondary phase is approximately 0.5 μm, which is similar to that of the secondary phase distributed among the dendrites in figure 1(c). By calibrating the SAED pattern, this phase was confirmed to be hexagonal Ni₃₁Si₁₂ (cell parameters are a = 0.667 nm, b = 0.667 nm, c = 1.228 nm, α = 90°, β = 90°, and γ = 120°), and its crystal zone axis is [1 1 0 1]. In the dark-field image shown in figure 2(b), there are a large number of small rice-like secondary phases with a length of less than 0.19 μm, which is consistent with the morphology of the intragranular phase in figure 1(c). The SAED pattern indicates that this rice-like phase is orthorhombic Ni₂Si (cell parameters are a = 0.707 nm, b = 0.501 nm, c = 0.373 nm, α = 90°, β = 90°, and γ = 90°), and the crystal zone axis is [1 0 0], which is parallel to the crystal zone axis of the Cu(Ni, Si) matrix ([0 0 1]₁₃[Ni₃₁Si₁₂][1 0 1]₁₃Cu). Based on the above analysis, the microstructure of the as-cast Cu–Ni–Si alloy can be finally confirmed: an α-Cu(Ni, Si) solid-solution matrix with a dendritic...
morphology, reticular Ni$_3$Si$_{12}$ phases distributed at the grain boundaries of the $\alpha$-Cu(Ni, Si) matrix, and rice-like Ni$_2$Si precipitates uniformly distributed inside the $\alpha$-Cu(Ni, Si) grains.

### 3.2. Microstructural evolution during multiple forging

Figure 3 shows the microstructure of the as-cast Cu–Ni–Si alloy after various numbers of multiple forging passes. After one forging pass, the dendritic morphology remained easily visible, and the grains were compressed
to flattened and elongated shapes, as shown in figure 3(a). High-magnification observations of the grain boundary of the $\alpha$-Cu(Ni, Si) matrix revealed that the reticular Ni$_{31}$Si$_{12}$ phase was slightly broken, as shown in the inset of figure 3(a). When the number of forging passes increased to 12, the microstructure was markedly different from that of the as-cast state and that after one-pass forging. The dendritic morphology changed to elongated equiaxed grains, and the morphology of the Ni$_{31}$Si$_{12}$ phase correspondingly changed from a reticular shape to a striped or elliptical shape. Upon further increasing the number of forging passes to 24, the $\alpha$-Cu(Ni, Si) matrix was intensively refined and presented a perfectly equiaxed structure, the reticular Ni$_{31}$Si$_{12}$ phase was completely broken into a small particle-like phases, and the small Ni$_2$Si precipitates could not be observed. The SAED pattern from the TEM analysis also demonstrated that the small particles were the Ni$_{31}$Si$_{12}$ phase (figure 3(d)).

Based on the processing conditions and microstructural evolution information, it can be concluded that multiple forging can lead to the refinement of the $\alpha$-Cu(Ni, Si) matrix grains and reticular Ni$_{31}$Si$_{12}$ phase on the grain boundaries of the matrix, and it also results in the disappearance of Ni$_2$Si precipitates. The refinement process of the $\alpha$-Cu(Ni, Si) matrix and Ni$_{31}$Si$_{12}$ phase can be divided into two stages. First, the $\alpha$-Cu(Ni, Si) matrix is elongated under the action of compressive stress when the number of forging passes is small, which can also break the Ni$_{31}$Si$_{12}$ phase because of its poor plasticity, as shown in figure 3(a). Second, as the number of forging passes increases further, plastic deformation accumulates, and the morphology of the $\alpha$-Cu(Ni, Si) matrix changes from dendritic to equiaxed through mechanical shearing and dynamic recrystallisation. In this process, the Ni$_{31}$Si$_{12}$ phase is also rotated and spheroidised, resulting in changes in both the morphology and size. The disappearance of Ni$_2$Si precipitates can be attributed to the heat treatment before forging, which is close to the solution temperature of Cu–Ni–Si alloys [25].

### 3.3. Variation in electrical conductivity and hardness during multiple forging

Figure 4 shows the variation in both the electrical conductivity and hardness of the as-cast Cu–Ni–Si alloy after multiple forging for different numbers of passes. With increasing number of forging passes, the electrical conductivity first increased and then decreased, while the hardness increased continuously until it slightly
decreased when the number of forging passes exceeded 18. According to the electron mechanism, electrical conductivity is affected by phonon scattering, dislocations, solution atoms, precipitates, grain boundaries, interfaces, and impurities [27, 28]. However, for precipitation-strengthened Cu–Ni–Si alloys, the most important factors are the solution atoms and precipitates [29]. Considering the microstructural evolution of the as-cast Cu–Ni–Si alloy after multiple forging for different numbers of passes, it can be easily confirmed that the increase in electrical conductivity is due to the destruction of the reticular Ni$_3$Si$_{12}$ phase, which is located on the grain boundaries and has poor conductivity; hence, it has a very negative effect on the electrical conductivity [20]. The decrease in electrical conductivity is the result of the dissolution of Ni$_2$Si precipitates because the atoms degrade the electrical conductivity considerably more in solid solution than as precipitates [30]. In contrast, the increase in hardness is the result of the dispersed Ni$_3$Si$_{12}$ phase and solution strengthening, while the slight decrease in hardness may be due to the coarsening of equiaxed grains after the maximum degree of grain refinement is reach at 18 forging passes.

3.4. Post heat treatment of the multiple-forged alloys

Based on the variations in both hardness and electrical conductivity with the number of forging passes, it is difficult to obtain a balance between them, which impedes the goal of preparing Cu alloys with high strength and high electrical conductivity. Considering the purpose of multiple forging, that is, destroying the reticular Ni$_3$Si$_{12}$ phase and refining the matrix grains, 18 times is confirmed to be the best number of forging passes. Figure 5(a) shows the evolution of the hardness and electrical conductivity of the 18-pass-forged Cu–Ni–Si alloy during isothermal ageing at 450 °C for different times. For the as-cast alloy, the initial electrical conductivity was 21.4% IACS, and the hardness was 142 HV, which are considerably lower than those of the aged alloy. With increasing ageing time, both the electrical conductivity and hardness first increased considerably; then, the conductivity increased slightly, and the hardness decreased slightly. The conductivity reached a maximum value of 37.3% IACS after ageing for 6 h, and the hardness reached a maximum value of 221.6 HV after ageing for 4 h. This suggests that a good balance can be achieved by the combination of multiple forging and isothermal ageing, while the slight decrease can be attributed to the coarsening of precipitates caused by over-ageing. Figures 5(b) and (c) show the corresponding microstructure and XRD spectrum of the Cu–Ni–Si alloy with the best combination of electrical conductivity and hardness. The microscale Ni$_3$Si$_{12}$ particles are uniformly dispersed among the equiaxed α-Cu(Ni, Si) solution matrix, and a large number of sub-microscale Ni$_2$Si precipitates are present in the α-Cu(Ni, Si) grain interior. These results suggest that a good balance between the electrical conductivity and hardness of Cu–Ni–Si alloys with high Ni and Si contents can be achieved by using multiple forging and ageing, where the role of the former process is to break the reticular Ni$_3$Si$_{12}$ phase at the grain boundary, and that of the latter is to promote the precipitation of Ni and Si atoms as the Ni$_3$Si phase. The SAED pattern from the TEM analysis also demonstrates that the small particles correspond to the Ni$_3$Si$_{12}$ phase, as shown in figure 3(d).
4. Conclusions

Multiple forging was conducted on an as-cast Cu–Ni–Si alloy with high Ni and Si contents, and its influence on the microstructure and properties was investigated. The underlying mechanism was also revealed, and the main conclusions are as follows.

(1) The as-cast Cu–Ni–Si alloy mainly consisted of a dendritic $\alpha$-Cu(Ni, Si) solution matrix, reticular Ni$_{33}$Si$_{12}$ phases at the grain boundaries, and rice-like Ni$_3$Si phases in the grain interior of the $\alpha$-Cu(Ni, Si) matrix.

(2) Multiple forging refined the $\alpha$-Cu(Ni, Si) matrix grains and reticular Ni$_{33}$Si$_{12}$ phases on the grain boundaries of the matrix through mechanical shearing and dynamic recrystallisation, while the Ni$_3$Si precipitates disappeared owing to the pre-heating treatment before forging.

(3) With increasing number of forging passes, the hardness increases continuously up to 18 passes owing to the refinement of the microstructure and dissolution of Ni$_3$Si precipitates into the matrix. In contrast, the electrical conductivity first increased because of the destruction of the reticular Ni$_{33}$Si$_{12}$ phase and then decreased because of the dissolution of Ni$_3$Si precipitates into the solid solution.

(4) A good balance between hardness and electrical conductivity was achieved by applying an ageing treatment to the Cu–Ni–Si alloy after multiple forging for 18 passes, and the corresponding microstructure comprised an equiaxed $\alpha$-Cu(Ni, Si) matrix with microscale Ni$_{33}$Si$_{12}$ particles and sub-microscale Ni$_3$Si precipitates.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

ORCID iDs

Jinlong Zhang https://orcid.org/0000-0002-6334-1043
Lei Jia https://orcid.org/0000-0002-2910-4925

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