Features of structure formation and thermal expansion of high alloys of the Al–Si–Cu system

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Abstract: The study results of the copper alloying effect on the microstructure and thermal coefficient of linear expansion (TCLE) of high aluminum alloys for special purposes based on the Al – (20÷30)% Si system are presented. It was found that alloy building of Al – 20% Si with copper in large quantities eliminates the linear expansion anomaly characteristic to the binary alloy and leads to a significant decrease in the thermal expansion coefficient over the entire temperature range of the tests. It was shown that Al–30%Si–20÷40% Cu alloys have stable TCLE in the low-temperature test interval. Metallographic analysis of high alloys showed that copper introduced in amounts of more than 20% contributes to a change in the morphology of the silicon phase and triple eutectic.

1. Introduction
The development and implementation of “breakthrough” technical projects in modern instrument-making industries is determined, first of all, by the materials properties. To date, the best achievement in the field of light alloys with low values of the thermal coefficient of linear expansion (TCLE) is sintered aluminum alloys (SAA) containing 25-30% of alloying elements such as Si, Ni, Fe. The production of alloys of the SAA type is a very expensive process, involving the use of ultrahigh crystallization rates ($\nu_{\text{crist.}} \geq 10^5 ^{\circ}\text{C/s}$), briquetting of powders and hot pressing of blanks [1].

High alloys of the Al– (20÷30)% Si system are special-purpose alloys, because have low TCLE values. To increase the mechanical properties and further more significantly decrease the thermal expansion coefficient in the Al–Si binary alloys, additional alloying elements are introduced, usually having lower TCLE than aluminum [2, 3].

Among alloying elements that reduce the TCLE of Al – Si alloys, copper occupies an important place. Studies of the thermal expansion features of Al–Si and Al–Cu binary alloys show that they can have a wide range of thermal expansion coefficients. The value of the thermal expansion coefficient is primarily determined by the content of alloying elements, as well as technological factors such as melt processing and crystallization conditions [4-7]. Silicon and copper reduce the thermal expansion coefficient of aluminum alloys when their content significantly exceeds the ultimate solubility in aluminum [8, 9].

According to the equilibrium state diagram, in the Al–Si–Cu system, up to 5% of copper can be in solid solution. In industrial alloys based on aluminum, the copper content does not exceed 6% for deformed and 10% for cast alloys. But in the literature there is very little information about the effect of a high copper content on the thermophysical properties of Al–Si alloys. It is known that the increase in the copper content up to 8–10% increases the strength at high temperatures and reduces the creep rate of aluminum, although there is still no consensus on when copper is more effective: if it is in a
solid solution or as part of the CuAl$_2$ phase [10, 11]. Therefore, it is of interest to study the thermal expansion of high alloys Al – Si – Cu with a copper content significantly above the solubility limit.

As for the phase composition of Al–Si–Cu alloys, ternary compounds are not formed in this system. The CuAl$_2$ phase and the silicon phase are in equilibrium with the aluminum solid solution [12]. The CuAl$_2$ phase crystallizes directly from the melt at a copper content of 53.3% at a temperature of 591 °C. According to the phase diagram of the Al–Cu state, it has a limited existence interval of 52.5–53.9% Cu, which does not reach the stoichiometric copper content (54.2%), which is due to the defect of the crystal lattice. The CuAl$_2$ phase has a tetragonal lattice; its TCLE is $\alpha_{27-127} = 15.9 \times 10^{-6}$ deg$^{-1}$ and $\alpha_{27-527} = 17.2 \times 10^{-6}$ deg$^{-1}$ [13]. In the Al–Si–Cu ternary system in equilibrium at a temperature of 525 °C and concentrations of alloying elements of 27% Cu and 5% Si, the triple eutectic reaction L $\rightarrow$ (α)+Si+CuAl$_2$ occurs [14].

In the case of nonequilibrium crystallization, the Si phase and the CuAl$_2$ phase are detected at much lower concentrations of copper and silicon compared to the state diagram. Usually, in the molten state, as a result of degeneration of the eutectic, the CuAl$_2$ phase has the form of veins; colonies of triple eutectics (α+Si+CuAl$_2$) are rarely found [15]. During the heat treatment, the silicon phase is more prone to spheroidization than the CuAl$_2$ phase.

2. Materials and methods of research

As an object of study, ternary alloys of the Al–Si–Cu system were chosen at different ratios of Si and Cu. Alloys were smelted in laboratory conditions in furnaces with heaters made of silite, subject to all rules for preparing the charge and conducting melting. The aluminum charge was melted, silicon was introduced into it in the amount of 20 and 30%, as well as copper in the amount of 20, 30 and 40%. Alloys were poured into an aluminum chill mold ($t_m = 20$ °C). The pouring temperature was 1100 – 1200 °C.

Samples for dilatometric and metallographic studies were made from the obtained ingots. TCLE was determined using a differential optical photo-recording dilatometer of the Schevenar system, the determination error was $\pm 0.1 \times 10^{-6}$ deg$^{-1}$. Metallographic analysis was performed on an OLYMPUS GX-5 optical microscope, at magnifications from 200 to 1000 times. Electron microscopy and X-ray microanalysis were carried out using a scanning electron microscope Carl Zeiss AG - EVO 50 Series.

3. Results and discussion

A metallographic analysis of ternary Al – 20% Si – Cu alloys showed that the combined introduction of silicon and copper prompts the formation of smaller primary crystals of the silicon phase than that of a binary alloy (figure 1, a, b). It is seen that between the crystals of the silicon phase there is a dispersed eutectic and solid solution dendrites. It should be noted that in the ternary alloy the size of dendritic branches increases and the distance between them decreases.

![Figure 1](image1.png)

**Figure 1.** Microstructure of the Al – 20% Si alloy (a); Al-20% Si-% 20 Cu (b); Al-20% Si-% 20 Cu (c).

The microstructure of the Al–20% Si–20% Cu alloy obtained with a high spatial resolution using a scanning electron microscope allows us to study in more detail the fine structure of the triple eutectic located between the primary crystals of the silicon phase (figure 1, c). It can be seen that the eutectic
has a fine needle structure with an average particle size of eutectic silicon of not more than 10 µm. Primary silicon crystals have a predominantly polyhedral shape, but acquire a complex configuration with cavities inside. Sections of α-solid solution are in the form of dendrites.

The results of X-ray microanalysis of the Al–20 % Si–% 20 Cu alloy showed that in some parts of the eutectic a high percentage ratio of Cu and Al is observed (37% and 52%, as well as 41% and 47%, respectively), which confirms the presence of particles in them CuAl₂ phases. Thus, silicon is present in the composition of the triple eutectic (α+Si+CuAl₂) in the form of needle-shaped particles, and copper is mainly included in the eutectic in the form of the CuAl₂ phase (figure 2).

![Figure 2. Microstructure and elemental composition of the Al–20 % Si–20 % Cu alloy in typical sample zones: siliceous phase (a); CuAl₂ phase (b, c).](image)

The effect of copper on the microstructure of Al–30% Si–Cu alloys was studied. The results of metallographic analysis are presented in figure 3. It was found that when from 20 to 30% of Cu is introduced into the alloy, the crystals of the siliceous phase mainly have the form of plates, between which a large needle eutectic and dendrites of the α-solid solution are observed. With the introduction of 40% Cu, the crystals of the siliceous phase acquire an irregular shape with many sites of increased etchability. Small sections of the eutectic are visible between the crystals of the siliceous phase, but dendrites of the α-solid solution are completely absent.

![Figure 3. Microstructure of alloys based on Al–30% Si with different copper contents: without Cu (a), 20% Cu (b), 30% Cu (c), 40% Cu (d).](image)

To study the structural features of high alloys, a metallographic analysis was performed using SEM (figure 4). It was established that the introduction of copper into the Al–30% Si alloy, in the microstructure of which, in addition to the eutectic, large particles of the silicon phase are initially present, contributes to an increase in the size of these particles. It was found that alloying of such
alloys with copper in large quantities (20–40% Cu) leads to a decrease in the solubility limit of silicon in aluminum, i.e. the conditions for the formation of alloys change.

![Figure 4](image)

**Figure 4.** The microstructure of alloys based on Al–30% Si with different Cu contents: 20% Cu (a), 30% Cu (b), 40% Cu (c).

The results of X-ray microanalysis of Al–30% Si–20–40% Cu alloys showed that a high percentage ratio of Cu and Al is observed in certain zones of the eutectic. Figure 5 shows that between the crystals of the silicon phase there are sections of a Cu solid solution in Al and zones with a high copper content (40% Cu and 49% Al) or (38% Cu and 53% Al), which are the CuAl₂ phase. Such changes in the structure lead to a significant decrease in TCLE from 12.4·10⁻⁶ to 6.0·10⁻⁶ deg⁻¹ in the range of 50–150 °C (figure 6).

![Figure 5](image)

**Figure 5.** Microstructure and elemental composition of Al–30% Si–20% Cu alloys in typical sample zones: siliceous phase (a); CuAl₂ phase (b); α-solid solution (c).

The effect of copper on the thermal expansion of Al–20% Si–Cu and Al–30% Si–Cu alloys was studied. It was established that the Al–20% Si–20% Cu alloy has stable TCLE in the low-temperature test interval. An increase in the copper content from 20 to 50% contributes to a smooth decrease in the thermal expansion coefficient of ternary alloys over the entire temperature range of the tests, as can be seen in figure 6, a. It should be noted that alloying Al–20% Si alloy with copper in large quantities eliminates the linear expansion anomaly characteristic of the double alloy and leads to a significant decrease in the
thermal expansion coefficient over the entire temperature range of the tests. So, the average TCLE of the Al–20% Si–20% Cu alloy in the range of 50–200 °C is $14.4 \times 10^{-6}$ deg$^{-1}$, while the TCLE of the Al–20% Si–30% Cu alloy is $13.1 \times 10^{-6}$ deg$^{-1}$. For the Al–20% Si–40% Cu alloy, a decrease in the thermal expansion coefficient to $9.6 \times 10^{-6}$ deg$^{-1}$ was established. The greatest decrease in the thermal expansion coefficient is observed for the Al–20% Si–50% Cu alloy; here, the thermal expansion coefficient decreases to $6.0 \times 10^{-6}$ deg$^{-1}$.

Most significantly, copper stabilizes the high-silicon alloys TCLE with a content of 30–50% Si. Figure 6b shows that alloying of the Al–Si binary alloy with copper in the amount of 20% allows us to obtain slightly varying TCLE in the range 50–400 °C: $\alpha = 12.0 \div 12.5 \times 10^{-6}$ deg$^{-1}$. An increase in copper content to 30% makes it possible to reduce the TCLE of the ternary Al–30% Si–30% Cu ternary alloy over the entire test temperature range: from $\alpha_{50} = 6.0 \times 10^{-6}$ deg$^{-1}$ to $\alpha_{450} = 6.8 \times 10^{-6}$ deg$^{-1}$.

![Figure 6. Thermal expansion of Al–Si–Cu alloys: Al–20% Si–Cu (a), Al–30% Si–Cu (b).](image)

It should be noted that for the Al–30% Si alloy, the introduction of copper of more than 30% is inappropriate since the increase in copper content up to 40% does not lead to a further decrease in the TCLE values, but its increase is observed in the high-temperature test interval.

4. Conclusion

With the introduction of large amounts of copper, the conditions for the structure formation of hypereutectic alloys Al–(20–30)% Si change. Copper introduced in amounts of more than 20% contributes to an increase in size and a change in the shape of the silicon phase, as well as a change in the structure of the triple eutectic.

Building of alloys Al– (20–30)% Si with copper in the amount of more than 20% leads to a significant decrease in the thermal expansion coefficient of ternary alloys over the entire temperature range of the tests. It was established that the Al–30% Si–20% Cu alloy has practically constant thermal expansion coefficients over the entire test temperature range of $12.5 \div 12.0 \times 10^{-6}$ deg$^{-1}$, which is typical for invar alloys [16]. This allows such alloys to be used when paired with steel products.
Thus, the joint alloying of aluminum with silicon and copper allows low and stable TCLE in the low and medium temperature test ranges to be obtained: Al–30% Si–20% Cu alloy has $\alpha = 12.0 \pm 12.5 \cdot 10^{-6}$ deg$^{-1}$ in the range of 50–400 °C; Al–30% Si–30% Cu alloy has $\alpha = 6.0 \cdot 10^{-6}$ deg$^{-1}$.

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