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ALUMINUM AND ALUMINUM ALLOYS

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SPECIAL FEATURES OF STRUCTURE FORMATION AND PROPERTIES OF SPECIAL HIGH-ALLOY ALLOYS OF THE Al – Si – Cu SYSTEM

V. K. Afanas'ev,[1] M. V. Popova,[1] and M. A. Malyukh[1]

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The special features of formation of structure in alloys Al – (20 – 50)% Si – (2 – 60)% Cu and the distribution of the alloying elements in them are studied. Metallographic analysis is made using optical and scanning electron microscopy and x-ray diffractometry. The density of the alloys is determined. The Al – Si – Cu compositions with low and steady values of the temperature coefficient of linear expansion (TCLE) $\alpha = (9.0 – 4.0) \times 10^{-6}\ K^{-1}$ in the range of 50 – 450°C are chosen. It is shown that the density of these alloys does not exceed 3500 kg/m^3. The results of the study may be used for developing low-TCLE alloys for special fields of instrument engineering.

Key words: high-alloy alloys, microstructure, silicon phase, copper aluminides, temperature coefficient of linear expansion, density.

INTRODUCTION

Successful development of many high-technology industries, such as precision machine tool building, engine making, instrument engineering, and high-vacuum technique, require light alloys with low thermal expansion. This property provides constancy of the shape of the articles in the operating temperature range and is characterized by quantity α, i.e., the temperature coefficient of linear expansion (TCLE).

Most advanced alloys with specified TCLE are invars (alloys based on the Fe – Ni and Fe – Ni – Co systems) [1, 2]. Invars possess TCLE $\alpha < 3.5 \times 10^{-6}\ K^{-1}$. However, these low values are preserved in a rather narrow temperature range (0 – 100°C). In addition, invars have some drawbacks such as a high density (about 8000 kg/m^2) and a reduced corrosion resistance [3]. The demand for improvement of the properties of this class of materials is growing. For example, researchers of the Institute for Problems of Superplasticity of the Russian Academy of Sciences (Ufa) work at invars with nanocrystalline structure formed by severe plastic deformation [4 – 6]. The articles serving in nonstationary magnetic fields should be produced from alloys exhibiting no ferromagnetism and having a high corrosion resistance and low density. Titanium-based nonmagnetic invar alloys possess satisfactory mechanical properties and a low TCLE, and their density does not exceed 5400 kg/m^3 [7, 8].

Aerospace engineering requires light nonmagnetic corrosion-resistant alloys with a low TCLE and a density not exceeding 3500 kg/m^3. Such materials are created by introducing elements with TCLE much lower than that of aluminum ($\alpha_{20–100} = 24 \times 10^{-6}\ K^{-1}$ in the range of 20 – 100°C) into aluminum. In the first turn, these are silicon, which possesses a quite low TCLE ($\alpha_{20–100} = 3.2 \times 10^{-6}\ K^{-1}$) and a low density (2330 kg/m^3), copper, and transition metals Ti, Zr, V, Cr etc. [9 – 12].

The best achievements in the field of creation of light alloys with a low TCLE are sintered aluminum alloys fabricated by the methods of powder metallurgy [13, 14]. These are hypereutectic silumins containing 25 – 30% Si and 5 – 7% other alloying elements. The TCLE of such silumins ($\alpha = (13.5 – 15.5) \times 10^{-6}\ K^{-1}$) is close to that of steels, and their density is 2720 – 2780 kg/m^3. However, many articles should be produced today from light nonmagnetic alloys with TCLE not exceeding $(6 – 10) \times 10^{-6}\ K^{-1}$ in the range of their operating temperatures.

[1] Siberian State Industrial University, Novokuznetsk, Russia (e-mail: m.popova@rdtc.ru).

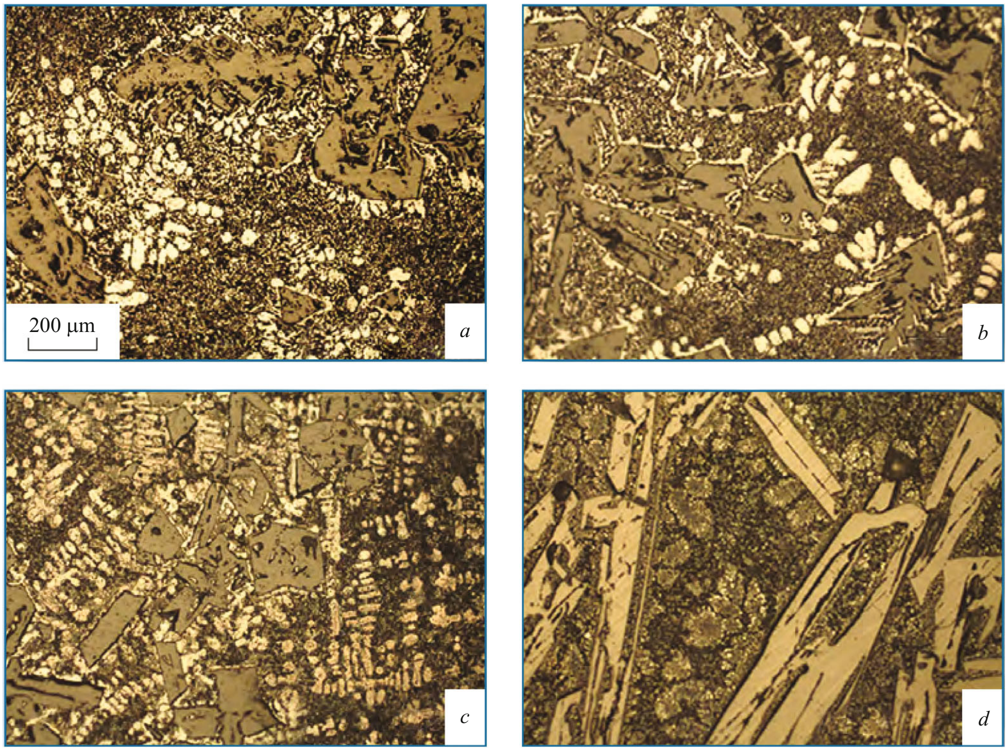


Fig. 1. Microstructure of Al – Si alloys without copper (*a*, *b*) and with copper addition (*c*, *d*) (scanning electron microscopy): *a*) Al – 20% Si; *b*) Al – 30% Si; *c*) Al – 20% Si – 20% Cu, and *d*) Al – 30% Si – 20% Cu.

The results of many-years' systematic studies in the field of creation of light alloys with regulated values of TCLE give us grounds to choose the Al – Si – Cu system for special-purpose alloys with specified TCLE. Silicon and copper lower considerably the TCLE of aluminum alloys when introduced in a content exceeding much their limiting solubility in aluminum [15 – 17].

However, the reported data on the effect of high contents of silicon and copper on the thermal expansion of aluminum alloys are quite scarce [18, 19]. For the most part, the studies have been devoted to the effect of these elements on other thermophysical properties, for example, the thermal conductivity and the heat transfer coefficient [22 – 22].

Formation of the structure and properties of high-alloy alloys is determined by the nature of their components and by the degree of likeness of their physicochemical properties. However, they depend much on the processes of production and treatment of the alloys, and this makes it impossible to design theoretically the compositions for alloys possessing specified values of TCLE in the temperature range of service of the article.

The aim of the present work was to study the special features of variation of the structure and the temperature coefficients of linear expansion of alloys of the Al – Si – Cu system, where the silicon content was varied from 20 to 50% and the copper content was varied from 2 to 60%.

METHODS OF STUDY

The alloys were melted in laboratory SShOL-type shaft resistance furnaces with silicon carbide heaters in alundum crucibles. The blends were composed of commercially pure aluminum A7, silicon Kr0 and copper M1.

Having melted the aluminum, we introduced silicon in small batches. When the silicon dissolved, we added copper and raised the temperature of the melt gradually to 1100 – 1200°C. After a 1 – 15-min hold, we removed the slag from the surface of the melt and poured the latter into a cold aluminum mold (the cooling rate was about 50 – 80 K/sec). The ingots obtained were used to make specimens for metallographic and dilatometric analyses.

The dilatometric analysis was performed using an OLYMPUS GX-5 optical microscope (OM) and a Carl Zeiss AG-EVO 50 XVP scanning electron microscope with an X-ACT microanalyzer.[2] The phase composition was determined by an x-ray diffraction analysis with the help of a DRON-2 diffractometer in iron K_α radiation. The density of the alloys was assessed by hydrostatic weighing using an ADV-200M analytical balance. The thermal expansion of the

[2] The electron microscopic studies were performed using the equipment of the common access center "Structure, Mechanical and Physical Properties of Materials" of the Novosibirsk State Technical University.

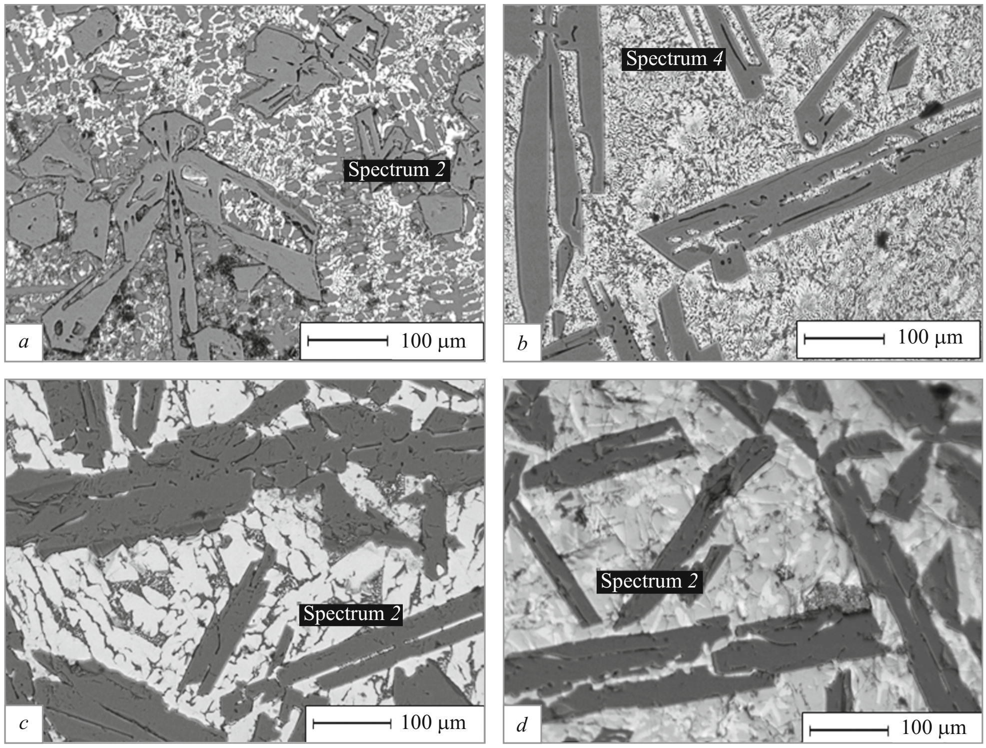


Fig. 2. Microstructure of alloys Al – 20% Si – 20% Cu (*a*), Al – 30% Si – 20% Cu (*b*), Al – 30 Si – 30% Cu (*c*), and Al – 30% Si – 40% Cu (*d*) (SEM).

alloys was studied using a Chevenard differential optical dilatometer in the 20 – 450°C temperature range. The true TCLE at the specified temperature was calculated by the tangent method. The total error of its determination was $\pm 0.10 \times 10^{-6}\ K^{-1}$.

RESULTS AND DISCUSSION

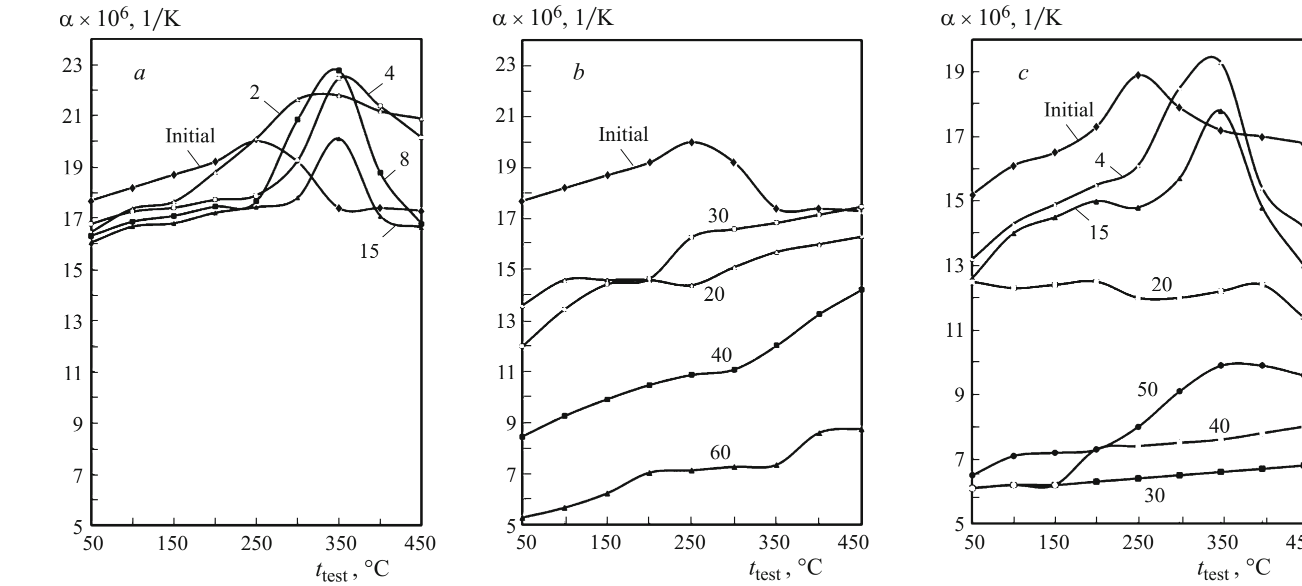
We studied two groups of alloys, i.e., (*1*) silumins with 20 and 30% Si and (*2*) high-silicon silumins with 40 and 50% Si. The copper content was varied from 2 to 60%in the alloys of the first group and from 4 to 50% in the alloys of the second group.

Alloys Al – (20 – 30)% Si – nCu. In the binary aluminum alloys with 20 or 30 % Si, the primary crystals of the silicon phase (SPC) have a compact polyhedral shape (see Fig. 1). A rather large content of a fine (α + Si) eutectic is located between the SPC). Dendrites of an α-solid solution formed due to the nonequilibrium crystallization of the alloy are also detectable (Fig. 1*a* and *b*).

Analysis of the microstructure of copper-alloyed Al – 20% Si alloys allowed us to establish that the introduction of up to 10% Cu refines the SPC without changing the sizes of the other structural components. Elevation of the copper content to 15 and 20% (up to 60%) causes growth of the SPC again, first to the size of those in the binary alloy and then to a larger size. In addition, the study of the microstructure and of the elemental composition of the phases with the help of SEM with a microanalyzer showed formation of a ternary (α + Si + $CuAl_3$) eutectic in the alloys containing ≥ 20% Cu and reduction of the size of the dendrite arms of the α-solid solution and of the distance between them (Fig. 2*a*).

By the data of the metallographic analysis of alloys Al – 30% Si – *n*Cu, the addition of 1 – 4% copper reduces the sizes of the SPC; above 4% Cu these sizes grow abruptly. The shape of the SPC changes from a polyhedral one to a lamellar one, and solid-solution dendrites do not form at all (Fig. 4*d*). Comparison of the microstructures of the alloys based on Al – 20% Si and Al – 30% Si shows that the highest growth in the sizes of the crystals of the silicon phase is typical for the alloys containing ≥ 20% Cu.

A characteristic feature of the structure of alloy Al – 30% Si – 20% Cu is formation of an exceptionally fine ternary eutectic (α + Si + $CuAl_2$) between lamellar SPC (Fig. 2*b*). When the copper content is increased to 30 and 40%, the eutectic acquires a coarser structure, and its volume fraction decreases strongly due to formation of particles of a $CuAl_2$ θ-phase (Fig. 2*c* and *d*). The data of the x-ray diffraction and microscopic x-ray spectrum analyses show that these particles are a subtraction solid solution based on an intermetallic $CuAl_2$ phase, because the concentration of copper in its varies from 37 to 41% (the stoichiometric content of copper corresponding to formula $CuAl_2$ is equal to 54.2%) [23].



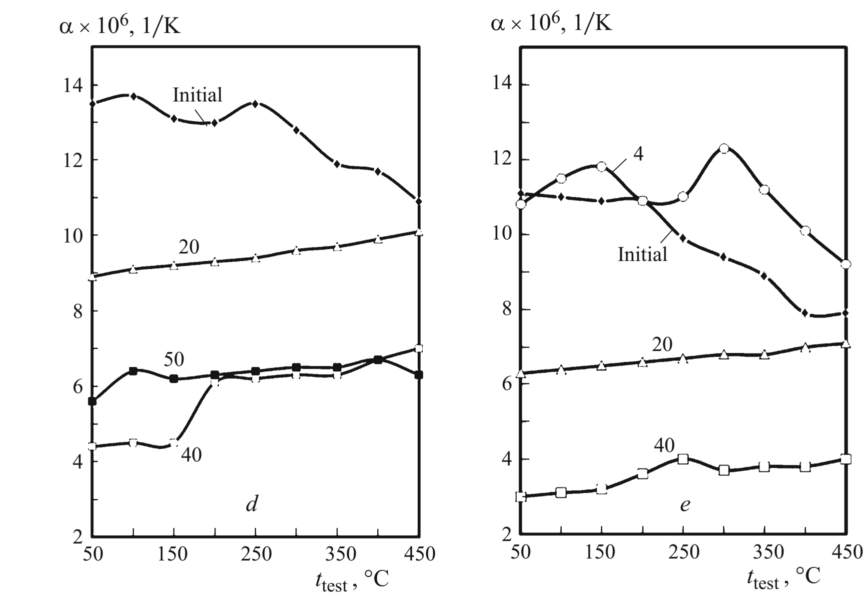


Fig. 3. Temperature dependences of the TCLE of alloys based on Al – 20% Si (*a*, *b*), Al – 30% Si (*c*), Al – 40% Si (*d*) and Al – 50% Si (*e*) and alloyed with different contents of copper (given at the curves in %): Init. — initial alloy (without copper).

We studied the effect of copper on the thermal expansion of alloys Al – 20% Si and Al – 30% Si. The TCLE of the binary alloys varied nonmonotonically with increase of the test temperature. In the range of 250 – 300°C they exhibited some anomaly of linear expansion, which consisted in considerable growth of the TCLE in the narrow range of the test temperatures (Fig. 3*a* and *c*). Copper, like silicon, has a TCLE lower than that of aluminum, and therefore its content in the ternary alloys should be reduced. The dilatometric analysis showed that when the copper content in alloy Al – 20% Si was increased from 2 to 15%, the TCLE decreased gradually at the test temperatures 50 – 250°C. However, at a higher test temperature, the TCLE of the ternary alloys did not obey the additivity rule; the values of this coefficient exceeded that for the binary silumin. The anomaly of the linear expansion typical for the binary alloy became more manifested and shifted toward the test temperatures of 300 – 400°C (Fig. 3*a*). Introduction of over 20% Cu into the Al – 20% Si alloy lowered the TCLE quite substantially in the whole of the range of the test temperatures (Fig. 3*b*). For example, the average TCLE of the binary Al – 20% Si alloy in the range of 50 – 200°C was $\bar{\alpha}_{50-200} = 18.8 \times 10^{-6}\ K^{-1}$, whereas at 50% Cu it fell to $\bar{\alpha}_{50-200} = 6.0 \times 10^{-6}\ K^{-1}$.

The TCLE-stabilizing action of copper was higher in alloy Al – 30% Si (Fig. 3*c*). Alloying of Al – 30% Si with 20% copper provided steady TCLE in the ternary alloy in the whole of the range of the test temperatures, i.e., $\bar{\alpha}_{50-450} = 12.2 \times 10^{-6}\ K^{-1}$. At 30% copper in the alloy, the TCLE fell to $\bar{\alpha}_{50-450} = 6.4 \times 10^{-6}\ K^{-1}$. It should be noted that the increase of the copper content to 40 and 50% not only lowered the TCLE still more, but also raised it somewhat in the high-temperature range (Fig. 3*c*).

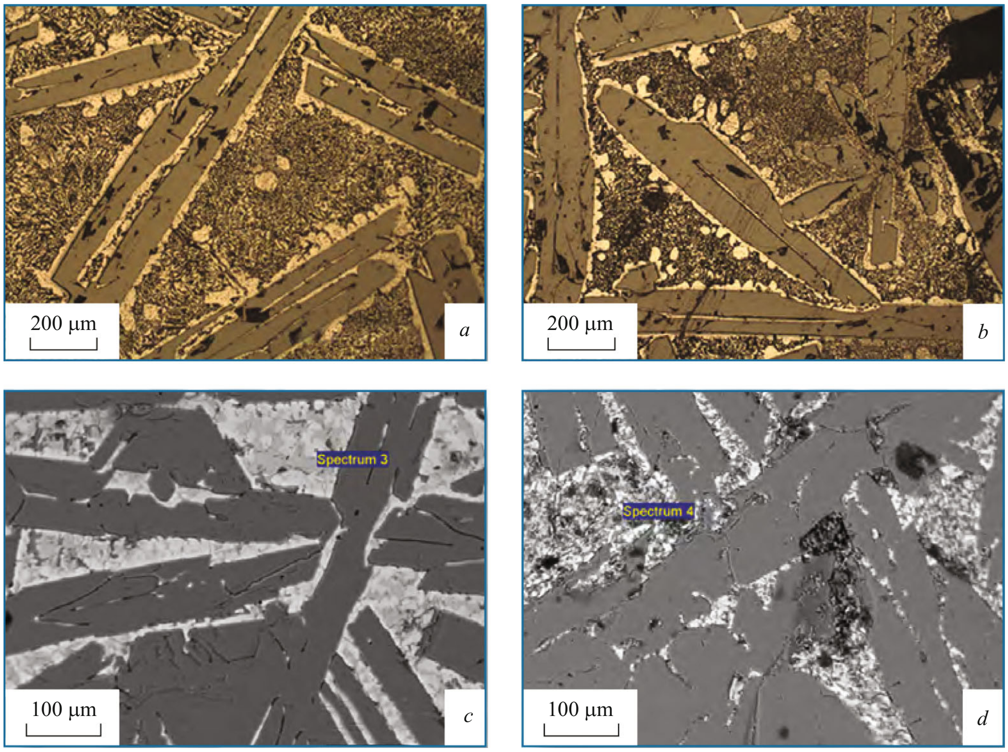


Fig. 4. Microstructure of Al – Si alloys without copper (*a*, *b*) and with copper addition (*c*, *d*) (scanning electron microscopy): *a*) Al – 40% Si; *b*) Al – 50% Si; *c*) Al – 40% Si – 40% Cu, and *d*) Al – 50% Si – 30% Cu.

Generalization of the results of the metallographic, x-ray spectrum and dilatometric analyses allows us to infer that the TCLE of the ternary alloys Al – (20 – 30)% Si – *n*Cu decreases considerably if their structure bears not only a great number of silicon primary crystals, but also particles of a $CuAl_2$ intermetallic. These phases create a rigid skeleton that lowers the thermal expansion of the alloys in a the whole of the tested temperature range (Fig. 2*c* and *d*).

Alloys Al – (40 – 50)% Si – nCu. The high-silicon aluminum alloys with 40 and 50% Si possess an initially low TCLE in the low-temperature test range as compared to the other silumins (Fig. 3*d* and *e*).

The structure of the binary alloys is characterized by coarse lamellar crystals of a silicon-bearing phase from 100 to 1000 μm in size, between which we observe an acicular (α + Si) eutectic (see Fig. 4). The primary crystals of the silicon phase are enframed with light dendrites of an α-solid solution formed due to the depletion of the melt of silicon during the formation of the SPC in the process of crystallization of the melt (Fig. 4*a* and *b*).

Alloying with copper in an amount ≥ 20% promotes formation of coarser SPC in the alloys and increase of their volume fraction. The ternary ($\alpha + Si + CuAl_2$) eutectic is present in the alloys in a very low amount. Intermetallic particles are located between the PSC (Fig. 4*c* and *d*). By the data of the x-ray diffraction analysis, the ternary Al – (40 – 50)% Si – *n*Cu alloys contain not only particles of a stable $CuAl_2$ phase but also bear metastable Cu_9Al_4 and CuAl intermetallics.

Generalization of the results of the dilatometric, metallographic and x-ray diffraction analyses shows that the joint introduction of silicon and copper in an amount ≥ 60% into aluminum changes the phase composition of the alloys so that their structure acquires a continuous net of a silicon phase and CuAl, $CuAl_2$ and Cu_9Al_4 aluminides. The TCLE of the aluminides is much lower than that of aluminum and their density of lower than that of copper. For example, the TCLE of the $CuAl_2$ phase is $\alpha_{27-127} = 15.9 \times 10^{-6}\ K^{-1}$ and the density is $\rho = 4340\ kg/m^3$ [24]. Therefore, the density of the alloys of the Al – Si – Cu system does not exceed $3500\ kg/m^3$ even when they contain 50% Cu. Such alloys may be used as base for developing special-purpose light alloys with specified TCLE.

The data obtained have been used to determine the possible directions of application of high-alloy Al – Si – Cu alloys with a TCLE close to that of invar alloys (see Table 1).

CONCLUSIONS

We have studied the structure and distribution of alloying elements in alloys Al – (20 – 50)% Si – (2 – 60)% Cu. By the data of the metallographic analysis, the structure of the

TABLE 1. Composition, Properties and Recommended Fields of Application of Known and Novel Alloys with Regulated TCLE

Alloy	Thermal properties		Application
	ρ, kg/m³	$\bar{\alpha} \times 10^6$, K^{-1}	
29NK (28.5 – 29.5% Ni; 17 – 18% Co)*	8000	$\alpha_{20-100} = 4.5$	Material for articles of radioelectronics (terminals, envelopes). Provides vacuum-tight seals with glasses of grades S39, S47, S52, S59
Al – 30% Si – 30% Cu**	3230	$\alpha_{20-100} = 4.6$	
Al – 50% Si – 40% Cu**	3410	$\alpha_{20-100} = 3.1$	
30NKD (29.5 – 30.5% Ni; 13.0 – 14.2% Co, 0.3 – 0.5% Cu)*	8200	$\alpha_{20-100} = 6.1$	Material for metallic components of vacuum and gas discharge devices connected to ceramic structures and integrated circuits. Connection to ceramics is made through low-temperature solders, cements and adhesives
Al – 40% Si – 40% Cu**	3420	$\alpha_{20-100} = 6.3$	
47NKhR (46 – 48% Ni; 4.5 – 6.0% Cr)*	8200	$\alpha_{20-200} = 9.1$	Alloy for making terminals and envelops of semiconductor devices. Provides vacuum tight seals with soft glasses of grades S80-1, S72-1, S90-1, S93-2
Al – 40% Si – 20% Cu**	3120	$\alpha_{20-200} = 9.1$	

* Standard alloys (the remainder Fe).

** Experimental alloys (described in the present work).

Notations: ρ) alloy density; α_{20-100} and α_{20-200}) temperature coefficients of linear expansion in the ranges of 20 – 100 and 20 – 200°C respectively.

high-alloy alloys with total content of silicon and copper exceeding 60% is characterized by coarse lamellar silicon primary crystals with particles of stable and metastable copper aluminides in between and some ternary eutectic (α + Si + $CuAl_2$).

Generalization of the results of the dilatometric and x-ray diffraction analyses allows us to state that joint introduction of silicon and copper into aluminum can reduce substantially the temperature coefficient of linear expansion (TCLE) of ternary alloys of the Al – Si – Cu system when their structure contains a great number of primary crystals of a silicon phase and some CuAl, $CuAl_2$ and Cu_9Al_4 intermetallics creating a rigid skeleton providing a size stability of the alloy. The density of such alloys does not exceed 3500 kg/m³.

The results of the dilatometric analysis allow us to determine the compositions of high-alloy Al – Si – Cu alloys with low and steady values of the TCLE ($\alpha = (9.0 - 4.0) \times 10^{-6}$ K^{-1}) in the temperature range 50 – 450°C. For example, the 40% Al – 30% Si – 30% Cu, 30% Al – 50% Si – 20% Cu and 10% Al – 50% Si – 40% Cu compositions exhibit virtually constant values of the TCLE ($\alpha = 6.4 \times 10^{-6}$ K^{-1}, 6.7×10^{-6} K^{-1}, and 3.5×10^{-6} K^{-1}, respectively) at 50 – 450°C. The studied compositions may be used as a base for developing materials with a low TCLE for special instrument engineering.

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