UDC 669.018.44:669.715

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ANALYSIS OF METAL SYSTEMS FOR DEVELOPING CREEP-RESISTANT ALUMINUM ALLOYS. A REVIEW

This article considers the role of the main alloying elements that have the most significant impact on the formation of aluminum alloys physical and mechanical properties for operation at high temperatures. Significant interest in research and prospects of casting compositions based mainly on eutectic systems and elements or compounds that can positively affect the structure and phase composition of alloys to increase their high temperature strength is shown. The analysis and comparison of the known systems Al–Si, Al–Cu, and Al–Mn with the systems Al–Fe, Al–Cr, Al–Ce, Al–Ni, Al–Ni–La, and others potentially suitable for creation of creap-resistant compositions are reviewed. Peculiarities of the structural-phase state, physical and mechanical characteristics of binary aluminum alloys, and the influence of the considered components on these indicators, depending on their content alloys obtaining conditions are considered. It is noted that the eutectic system Al–Ca, despite some similarities with other considered systems, does not meet the necessary requirements for the creation of alloys, operating at elevated temperatures. However, experimental cast alloys of this system are characterized by high specific strength and ductility. The following trends in the development of heat-resistant aluminum alloys have been identified: the use of almost insoluble elements and elements with a low diffusion coefficient in aluminum; creation of eutectic type complex-alloyed multicomponent alloys; creation of rapidly crystallized multicomponent alloys, which are subjected to further pressing and sintering. The development of cast alloys of Al–Fe–Mn–Ni, Al–Fe–Cr–Mn–Ni, and Al–Ni–La systems with nanostructured thermally stable eutectic deserves special attention and seems to be the main trend in reviewed question. The need for research related to microalloying and modification of such alloys is indicated.

Keywords: *material development, creap-resistant aluminum alloys, cast aluminum alloys, eutectic systems.*

In nowadays, mainly used creep-resistant aluminum-based alloys are related to Al–Si, Al–Si–Cu, Al–Si–Cu–Ni, and some similar systems. Nevertheless, the limit of their application temperatures is about 200–300 ^{0}C and they are also characterized by low mechanical properties. Due to rising demands for modern materials, the development of new creep-resistant aluminum alloys looks like an important task. According to this, it is necessary to overview the main principles of such materials creation and to learn some useful information about basic systems for their development [1–5].

Al–Cu system. Copper belongs to the most important alloying element for aluminum alloys due to its effective solid-state hardening. The presence of the intermetallic compound Al2Cu (θ-phase) determines the possibility of dispersion hardening during alloy age-hardening. The maximum solubility of copper in aluminum at the temperature of eutectic transformation is equal to 5.65 % wt., and the normal copper grade concentration in most aluminum alloys is in the range of 2–6 % wt. [4, 5].

With the increasing percentage of copper, the heat resistance of the alloy significantly increases (at 250–300 ^{0}C) due to the strengthening of the interatomic bonds of the solid solution based on aluminum [5, 6]. This is confirmed by the data according to which binary parts of Al_5Cu and $Al_{6.5}Cu$ are used under conditions of moderate temperatures (up to 200–225 ^{0}C) [4, 7]. At higher operating temperatures, θ-phase coagulation occurs, which significantly reduces the strength of the alloy and negatively affects the high-temperature strength [1, 8–10]. Stabilization of the Al_2Cu phase with scandium, zirconium, and erbium, as well as complex multi-stage heat treatment, allows obtaining more stable nanoscale inclusions in the alloy structure, which retain their size and slightly increase the alloy operating temperature to ~ 300 ^{0}C [11].

The Al–Cu system has an aesthetic type of state diagram [3, 10], and copper itself has a positive effect on reducing the coefficient of linear thermal expansion of aluminum alloys, which are positive features for alloying cast heat-resistant alloys [8, 11]. The addition of copper also reduces the degree of tetragonality of phases with a structure of the DO_{22} type, which has a positive effect on the mechanical properties of heat-resistant alloys [2, 12]. At the same time, it should be noted that copper reduces the melting point of iron-containing eutectics and the iron solubility in a solid solution based on aluminum, as well as reduces the corrosion resistance of aluminum alloys [1, 6, 13].

Despite the low cost and simpleness of creating Al–Cu heat-resistant alloys, such compositions have a large number of potential disadvantages. Apart from the unstable Al_2Cu phase, among them, we can also note the relatively low temperature of the eutectic transformation and the wide crystallization interval in the effective copper concentrations area. These data lead to the conclusion that copper cannot be the main alloying element for the creation of heat-resistant alloys [1, 14].

Al–Mn system. Manganese is a very important alloying element for aluminum alloys due to a number of features of positive influence on their structure and properties. It is able to reduce the harmful effects of iron in Al–Cu, Al–Si–Cu, Al–Mg, and Al–Mg–Si, as well as increase their corrosion resistance and strength. Manganese, like copper, increases the heat resistance of aluminum alloys and reduces the coefficient of linear thermal expansion of aluminum alloys, which in its turn increases the crack resistance of castings made of them [1, 6, 15, 16].

Typically, in industrial cast aluminum alloys manganese is contained in an amount of up to 0.5 % wt., and for wrought – up to 1 % wt. [1, 3, 11, 17, 18]. Like copper, manganese forms a eutectic with aluminum, but at a higher temperature. Eutectic transformation occurs at a manganese concentration of ~ 1.9 % wt. by 657 ^{0}C [3]. The solubility of manganese in aluminum at the temperature of eutectic transformation is about 1.8 % wt., and at room temperature – 0.168 % wt., so it could be considered as an effective reinforcing element [3, 6, 10, 18]. The strengthening phase is Al_6Mn, which is released from the supersaturated solid solution during age-hardening. The same phase can be initially formed and be a part of the eutectic [19].

An important feature of manganese is that it has an extremely low diffusion coefficient in aluminum, many times lower than copper and magnesium, and the compound Al_6Mn is not prone to coagulation and is stable up to 500 ^{0}C, which makes manganese an advanced material for aluminum-based heat-resistant alloys [5, 19–22].

Al–Fe system. Iron is always present as an impurity in aluminum alloys, especially in those obtained from secondary raw materials. It is considered mainly a harmful impurity in cast aluminum alloys due to the formation of unfavorable needle morphology phases [1, 5, 23]. Iron is added to some Al–Cu–Ni alloys to increase heat resistance, as well as to reduce corrosion in water vapor at high temperatures [3].

Uncertainly iron influences the technological properties of cast aluminum alloys. Its increased content in silumin helps to get rid of the metallic pickup to the molds during injection molding, so the concentration of iron in such alloys can be up to 1.5 % wt. At the same time, with an increasing amount of iron in aluminum alloys, their tendency to form gas and shrinkage porosity increases, but the coefficient of linear thermal expansion and shrinkage decreases [1, 3].

According to [19, 24, 25], the Al–Fe system is considered to be one of the most high-potential for the creation of heat-resistant cast alloys based on aluminum due to the presence of eutectic, which is formed at an iron content of 1.7–2.2 % wt. Another important factor is the implementation of mechanisms of high-temperature hardening of alloys and grinding of their crystal structure due to the separation of the primary phases of Al_3Fe and Al_6Fe during rapid cooling of the melt [3].

The eutectic of Al-Al_6Fe has a very favorable structure and mainly submicron allocations of the Al_6Fe phase of round or fibrous morphology [26–29]. Such characteristics allow us to consider Al–Fe alloys as high-potential heat-resistant materials and Al_6Fe phase as an effective strengthening component of aluminum alloys. An example given in [27] is the Al3Fe alloy, which is obtained by angular extrusion to grind the structure of the alloy and ensure its uniform structure. The size of the Al_6Fe phase emissions, in this case, was 50–100nm in diameter and 300–500 nm in length. The tensile strength of this alloy was 175MPa. The data of [28] show that the strength of the cast alloy of the Al–Mg–Fe system, which crystallized under conditions of rapid cooling of the melt, had strength values at the level of 120–213MPa and ductility $\delta \sim 2–6.2$ %.

In [30], iron is evaluated as a potentially useful component to increase the heat resistance of piston alloys to be recycled. In [31–33] an example of the creation of super eutectic silumin, which contains 5 % wt. iron and shows the effectiveness of the high content of this element to increase the heat resistance and heat resistance of the alloy. In [34] there are examples of super eutectic powder silumin with an iron content of 8 % wt., but in addition, they contain 5 % wt. Mn or 1 % wt. Cr, which clearly shows the importance of high iron content to ensure a high level of heat resistance and heat resistance of aluminum alloys.

The Al–Ni system has a eutectic reaction at a concentration of Nickel of about 6 % wt., which takes place at 640 °C. In the solid state, there is a low solubility of nickel in aluminum at the level of 0.04 to 0.006 % wt. at 640 ^{0}C and 527 ^{0}C, respectively [3, 10, 35]. For this system, it is possible to implement the mechanisms of dispersing the strengthening of the aluminum matrix by thermally stable and diffusion-inactive particles of Al_3Ni intermetallic [2], similar to the Al–Fe system and some others [3, 16].

It is known that nickel is an alloying element in some standard aluminum alloys, which are used to make parts of internal combustion engines [24, 30, 36]. In addition, in recent years, alloys of Al–Ni, Al–Ni–Fe, Al–Ni–Cr, Al–Ni–Fe–Ce, and some other compositions have received considerable attention from the standpoint of their use as heat-resistant and heat-resistant materials [37–40]. The casting properties of such alloys are quite high due to the eutectic of Al+Al_3Ni. The intermetallic component of Al_3Ni of this eutectic has a fibrous morphology and has a favorable effect on the strength of alloys. However, their matrix requires additional doping due to the almost zero solubility of nickel itself.

As an additive in known industrial casting alloys, nickel is able to increase the fluidity of the alloy, give a more compact round shape to the eutectic components and bind to the θ-phase of Al_2Cu, thereby increasing its thermal stability above 250 ^{0}C [36]. Like iron, nickel shifts the point of eutectic transformation toward a lower concentration of aluminum [24]. Nickel is a very weak nucleating modifier and has a greater effect on the structure of alloys due to inhibition of grain growth. The addition of nickel alone or together with iron increases the corrosive properties of aluminum alloys in hot water and water vapor [3].

The Al–Co system can also be considered as potentially high-potential for the creation of heat-resistant cast aluminum alloys due to the presence of eutectic transformation at a Co content of about 1 % wt. and at 657 ^{0}C. Like nickel and iron, cobalt is almost insoluble in aluminum and has low diffusion activity. The eutectic of Al–Al_9Co_2 can have a fibrous structure during rapid crystallization, which has a positive effect on the mechanical properties of such alloys [3].

Cobalt can also be considered a useful additive for aluminum alloys. Its addition to silumin with a silicon content above 5 % wt. more effectively affects the leveling of the harmful effects of iron than manganese. Microalloying of aluminum and its cobalt alloys has a positive effect on the corrosion properties and in sulfuric acid and salt solutions environments. Giving cobalt slightly increases the strength of aluminum and its alloys. The Co-addition of cobalt and molybdenum increases the strength more significantly [3, 41, 42].

The Al–Cr system is also of great interest to a number of developers of heat-resistant aluminum alloys [43–45]. In early works [3] it was assumed that for this system there is a peritectic reaction between Al and Cr on the aluminum side of the state diagram. More recent studies [46, 47] indicate the presence of eutectic transformation at a chromium content of 0.8 % wt. by 661^{0}C.

In general, chromium as an impurity is able to reduce the harmful effects of iron in silumin and inhibit the recrystallization of aluminum alloys. Its low solubility in aluminum and low diffusion coefficient, like Fe, Co, Ni, potentially makes it possible to consider chromium as a stable reinforcing element. However, the Al_7Cr phase, which is part of the Al–Cr eutectic and can be released inside aluminum grains, has a rather coarse morphology and is prone to rapid growth during crystallization. This adversely affects the strength, ductility, and toughness of the alloys. For these reasons, chromium is added to classic foundry aluminum alloys in very small quantities. Alloys containing more than 1 % wt. chromium is produced mainly by powder metallurgy. In this case, the strength and corrosion resistance of such materials is higher than that of cast analogs [3, 43, 48].

The Al–Ca system has recently become increasingly of interest to researchers due to the possibility of developing heat-resistant low-density casting alloys [49–52]. Eutectic $Al+Al_4Ca$ exists at a calcium concentration of 7.6 % wt. at 617 ^{0}C. In this case, the solubility of calcium in solid aluminum at the temperature of eutectic transformation is 0.01 % wt., which gives the right to consider it along with nickel, cobalt, iron, and some other elements as an almost insoluble and stable reinforcing component. The Al_4Ca phase has a tetragonal crystal lattice, so calcium is potentially a good reinforcer, which at the same time slightly reduces plasticity [3, 49]. The corresponding hypothesis was tested in [53] and shown that as in binary compositions as with the addition of other elements, the Al–Ca alloys are quite malleable and capable of nanostructuring after intense plastic deformation. In the same studies, it is shown that the hardness and strength of such alloys after deformation treatment begin to decrease after heating to temperatures of 200–350 ^{0}C. At the same time, there are structural-phase changes with a noticeable growth of grains, which reduces the potential of such alloys in the areas of high-temperature application, while increasing interest in them as high-strength materials.

Foundry melts of the Al–Ca system are usually doped with iron, manganese, and nickel to increase their strength at room and elevated temperatures. The addition of nickel and iron mostly affects the structure of the eutectic. To strengthen the aluminum matrix using manganese and scandium in the amount of 0.2–0.3 % wt. [54–57].

Despite the favorable characteristics of Al–Ca alloys and their structural-phase features, a significant disadvantage of alloys is that even a small content of calcium (> 0.15 % wt.) dramatically reduces the corrosion resistance of aluminum [3, 49]. Also, as mentioned above, the temperature limit of their operation does not exceed the indicators of eutectic silumins. Accordingly, the emphasis on the creation of aluminum-calcium alloys has been shifted towards the creation of high-strength compositions that can be obtained by casting [58–60].

Al–REM systems are mostly represented by Al–Ce and Al–La based alloys. Both systems are characterized by the presence of eutectic transformation – 12 % wt. at 637 ^{0}C and 12 % wt. at 642 ^{0}C for Ce and La, respectively. The solubility of both elements at eutectic transformation temperatures is ~ 0.05 % wt. and almost disappears at room temperature [1, 3, 49].

The eutectic compound Al_4Ce is characterized by a BCC-tetragonal lattice, so the addition of cerium has little effect on the strength and ductility of aluminum [3], so the development of Al–Ce alloys is closely related to the use of various alloying additives. For example, nickel, iron, and zirconium are added to form more thermally stable ternary intermetallics [61–63].

Alloys of Al–Ni, Al–Ce, and Al–Cu–Ce systems in their structure correspond to natural composites, which make them high-potential materials in the context of the use at high temperatures, but the high cost of nickel and cerium restrains their widespread industrial use [1, 63, 64].

Lanthanum in aluminum alloys is widely used as a modifying additive. For alloys of the Al–Cu system, in particular, Al_6Cu with the addition of titanium, manganese, zirconium, and boron, the addition of 0.3–1 % wt. lanthanum led to a marked increase in strength and heat resistance due to the formation of $Al_{11}La_3$ compounds in eutectic and interdendritic zones [65]. The addition of lanthanum to super eutectic silumin leads to the formation of crystals of primary silicon branched morphology [66]. Modification of the eutectic alloy Al11Si1.5Cu0.3Mg with lanthanum containing boron and strontium showed effective neutralization of these elements with the simultaneous formation of LaB6 and increasing the strength and ductility of the alloy to 270 MPa and 5.8 %, respectively [67]. Studies of modification of pre-eutectic silumin $AlSi_9Cu_3$ with lanthanum and nickel have shown that nickel is also able to enhance the positive effects of manganese and, to a lesser extent, chromium in the context of eliminating the harmful effects of iron. Lanthanum behaves in the same way. With the simultaneous addition of lanthanum and nickel, the modifying effect becomes unpredictable due to the tendency of nickel to enter the eutectic. Lanthanum may also be part of the eutectic. The complication of phase stoichiometry and eutectic leads to the heterogeneity of the structure of both cast metal and heat-treated [68].

Al–La binary alloys are considered as potentially suitable heat-resistant alloys with high corrosion resistance. They show strength at the level of 60–93 MPa at 200 ^{0}C and 13–18 MPa at 500 ^{0}C and have favorable structural-phase characteristics – finely dispersed eutectic and uniform distribution of fairly fine primary phases in the case of eutectic alloys [69]. However, to this date, preference is given to the creation and study of ternary and multicomponent lanthanum-containing alloys due to their higher level of performance.

In a conclusion, it can be said, that Fe, Mn, Cr, Co, Ni, Ce, La, and different combinations of these elements are the most promising components for the development of creep-resistant Al-based alloys. The highest attention should be given to those compositions, which may have nanostructured eutectic and solid solution precipitates, like for Al–Ni–La alloys. However, complex impacts and effects from addition alloying and microalloying of such compositions are purely discovered and need a significant number of experiments and investigations.

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Received 28.04.2021

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АНАЛІЗ МЕТАЛЕВИХ СИСТЕМ ДЛЯ РОЗРОБКИ ЖАРОМІЦНИХ АЛЮМІНІЄВИХ СПЛАВІВ. ОГЛЯД

У статті розглядається роль основних легуючих елементів, які мають найбільш значний вплив на формування в алюмінієвих сплавах комплексу фізико-механічних властивостей для роботи при високих температурах. Показаний значний інтерес до досліджень та перспектив створення ливарних композицій, які базуються головним чином на евтектичних системах та елементах або сполуках, які можуть позитивно впливати на структуру та фазовий склад сплавів для підвищення їх високотемпературної міцності. Проведено аналіз і порівняння відомих систем Al–Si, Al–Cu та Al–Mn з потенційно придатними для створення жароміцних композицій системами Al–Fe, Al–Cr, Al–Ce, Al–Ni, Al–Ni–La та ін. Розглянуто особливості структурно-фазового стану та фізико-механічних характеристик бінарних алюмінієвих сплавів та вплив на ці показники розглянутих компонентів, в залежності від їх вмісту та умов одержання сплавів. Відмічається, що евтектична система Al–Ca, не дивлячись на певну схожість з іншими розглянутими системами, не відповідає необхідним вимогам для створення сплавів, що працюють при підвищених температурах. Проте, експериментальні ливарні сплави даної системи характеризуються високою питомою міцністю і пластичністю. Визначено наступні тенденції розробок жароміцних алюмінієвих сплавів: застосування майже нерозчинних елементів та елементів з низьким коефіцієнтом дифузії в алюмінії; створення складнолегованих багатокомпонентних сплавів евтектичного типу; створення швидко закристалізованих багатокомпонентних сплавів, які піддають подальшому пресуванню та спіканню. Особливої уваги заслуговує розробка ливарних сплавів систем Al–Fe–Mn–Ni, Al–Fe–Cr–Mn–Ni та Al–Ni–La з наноструктурною термічно стабільною евтектикою. Вказано на необхідність досліджень, пов'язаних з мікролегуванням та модифікуванням таких сплавів.

Ключові слова: *розробка матеріалів, жароміцні алюмінієві сплави, ливарні алюмінієві сплави, евтектичні системи.*