

Temperature dependence of the heat capacity and change in the thermodynamic functions of strontium-alloyed AK1M2 alloy

Izatullo N. Ganiev¹, Suhrob E. Otajonov², Nasim F. Ibrohimov³, M. Mahmudov²

1 V. I. Nikitin Institute of Chemistry, Academy of Sciences of the Republic of Tajikistan, 299/2 Aini Str., Dushanbe 734063, Tajikistan

2 Khujand State University name after B. Ghafurov, 1 Mavlonbekov Passage, Khujand 735700, Tajikistan

3 Tajik Technical University named after academic M. S. Osimi, 10 ac. Rajabovho Str., Dushanbe 734042, Tajikistan

Corresponding author: *Izatullo N. Ganiev (ganiev48@mail.ru)*

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Abstract

The temperature dependence of the specific heat capacity and change in the thermodynamic functions of strontium-alloyed ultrahigh-purity aluminum base AK1M2 alloy have been studied in “cooling” mode over the 298.15–900 K range. Mathematical models describing the evolution of these properties of the alloys in the abovementioned temperature range with change in alloying addition concentration have been obtained. The heat capacity, enthalpy and entropy of the alloys increase with temperature, decrease with an increase in the alloying addition concentration to 0.5 wt.% and grow with a further increase in the alloying addition concentration. The Gibbs energy of the alloys has an inverse dependence: it decreases with an increase in temperature and grows with an increase in the alloying addition concentration to 0.5 wt.%.

Keywords

AK1M2 alloy, strontium, “cooling” mode, heat capacity, enthalpy, entropy, Gibbs energy

1. Introduction

Numerous works have dealt with the physicochemical properties of industrial purity aluminum alloys [1–12], but the situation concerning ultrahigh-purity aluminum alloys is different. However, ultrahigh-purity aluminum and its alloys have found applications in microelectronics for IC fabrication.

The design and industrial fabrication of ICs are the greatest advantage of modern microelectronics. We now

have ICs incorporating whole devices and even systems within a single semiconductor crystal.

However far not any device can be fabricated using semiconductor technologies due to their limited capabilities regarding the fabrication of stable passive elements possessing a wide range of working parameters. Therefore the development of semiconductor technologies is accompanied by the improvement of another design and

technology solution for the fabrication of microelectronics devices which has already allowed combining semiconductor microchips and discrete semiconductor devices with passive film elements for designing microelectronics devices with a wide range of functional capabilities. ICs containing, along with film elements, active semiconductor elements fabricated using semiconductor technologies in the form of independent design elements are referred to as hybrid integrated circuits (HICs). HICs have a number of advantages compared with ICs: they are cheaper than semiconductor ones for small-batch production and provide for a wide range of ratings, lower tolerance and better electric parameters of passive elements. HICs allow the use of any discrete components including semiconductor ones. Hybrid technologies provide for the fabrication of devices having sufficiently high power [2, 4, 13, 14].

An important issue of microelectronics is providing stable, reproducible and reliable HICs. The problem of increasing device reliability is complex and multifaceted. One way to solve it implies the design and implementation of new materials and technologies.

Metallic substrates used in the industry nowadays include can be based on coated steel, polyimide lacquer coated steel, titanium and aluminum alloys. The use of steel and titanium substrates has a number of disadvantages restricting the range of their potential applications.

Of greatest practical interest are anodized aluminum base substrates combined with interconnections on polyimide films which allow fabricating multilevel interconnections and provide for efficient heat removal and the required structural strength. High-strength aluminum alloys based on high- and ultrahigh-purity aluminum are actually used. The alloying additions in these alloys should be susceptible to anodic oxidation by analogy with aluminum. To allow achieving a substrate surface purity of 13–14 grade for subsequent anodizing, the alloys should have highly homogeneous structure and composition across the entire wafer. Therefore high contents of alloying additions are undesired [2, 4, 13, 14].

In Russian and international practice, the technology of thin metallic films for ICs is currently undergoing a transition from single-component metallic materials to high-purity metal base alloys containing two or more alloying additions. This transition is absolutely reasonable since the use of pure metals as semiconducting materials may lead to a number of technological and operation deviations which can be avoided by microalloying. The application of microalloying technologies for aluminum alloys faces a number of problems unawareness or ignorance of which may cause negative consequences. These problems mainly include:

- choice of type and optimum composition of alloying additions;
- purity of alloying components which is currently a complex task;
- availability of stable technologies and reliable equipment for obtaining high-purity alloys and absence of sufficiently developed theoretical basis for the choice of required compositions.

High-purity aluminum base alloys with well-studied nature, structure and properties would greatly improve the performance of the devices. They would furthermore favor broadening of high-purity aluminum application domains in other branches of science and technology, and exhibit earlier unknown properties. Therefore research efforts aimed at exploring potential applications of the new ultrahigh-purity aluminum base alloys are important and quite timely [14, 15].

Unfortunately, researchers currently pay undeservedly small attention to the development of theoretical fundamentals for the choice of required alloy compositions such as the physicochemical properties of ultrahigh-purity aluminum base alloys. The systems in question include e.g. the AK1 aluminum–silicon alloy and the AK1M2 aluminum–copper alloy with alkaline–earth metal additions [14, 15].

The aim of this work is to study the effect of temperature and strontium content on the heat capacity and thermodynamic characteristics of ultrahigh-purity aluminum base AK1M2 alloy.

2. Experimental

Measuring the heat capacity and its temperature dependence is an important tool in the study of alloys. There are few if any literary data on the heat capacity of multicomponent aluminum alloys.

Below we present experimental data on the temperature dependence of the specific heat capacity of the strontium alloyed AK1M2 alloy (Al + 1wt.% Si + 2wt.% Cu). Since a monotonic change in test material temperature can hardly be attained in “heating” mode due to a wide range of external factors (furnace power supply voltage, ambient heat capacity etc.), i.e., the experiment has a multifactor character, we considered “cooling” mode as the most suitable and simple approach from this viewpoint.

We measured the heat capacity of the alloys on an instrument the operation of which is based on the method of a C-calorimeter with a heat gage and an adiabatic enclosure. The heat capacity measurement method and the instrument design were reported earlier [16–20].

The specific heat capacity of metals was measured using the Newton–Richman cooling law. Each body the temperature of which is higher than the ambient one is cooled down, with the cooling rate being controlled by the body’s heat capacity and heat emission factor. The heat flow passing through the heat gage is assessed based on the temperature gradient at the heat gage and the thermal conductance of the heat gage as determined in independent graduated experiments for a copper reference specimen. The temperature range is up to 900 K. Taking two metal rods of a specific shape one being the reference one (its heat capacity and cooling rate must be known) and comparing the cooling curves (temperature vs time functions) of these specimens one can determine the heat capacity of the other rod based on its cooling rate.

The quantity of heat δQ lost by the pre-heated body with the mass m as a result of its cooling by dT degrees can be determined using the formula

$$\delta Q = C_p^0 m dT, \quad (1)$$

where C_p^0 is the standard specific heat capacity of the material of the body at a constant pressure.

The energy is lost via the body surface. One can therefore accept that the quantity of heat δQ_S lost via the body surface over the time $d\tau$ is proportional to the time, the surface area S and the difference between the temperature T of the body and the temperature T_0 of the environment:

$$\delta Q_S = -\alpha(T - T_0) S d\tau, \quad (2)$$

where α is the heat emission factor. If the body emits heat in such a way that the temperature of all its points varies similarly, we can write

$$\delta Q = \delta Q_S; C_p^0 m dT = -\alpha(T - T_0) S d\tau. \quad (3)$$

Equation (3) can be represented in the following form:

$$C_p^0 m \frac{dT}{d\tau} = -\alpha(T - T_0) S. \quad (4)$$

Assuming that for a small temperature range C_p^0 , α , T and T_0 do not depend on the coordinates of specimen surface points heated to the same temperature and the similar temperature of the environment, we can write Eq. (4) for two specimens:

$$C_{p_1}^0 m_1 S_1 \alpha_1 \left(\frac{dT}{d\tau} \right)_1 = C_{p_2}^0 m_2 S_2 \alpha_2 \left(\frac{dT}{d\tau} \right)_2. \quad (5)$$

Using Eq. (5) for two specimens (the reference one and any other) having similar sizes $S_1 = S_2$ and surface conditions implies taking their heat emission factors to be equal: $\alpha_1 = \alpha_2$. Then

$$C_{p_1}^0 m_1 \left(\frac{dT}{d\tau} \right)_1 = C_{p_2}^0 m_2 \left(\frac{dT}{d\tau} \right)_2. \quad (6)$$

Therefore knowing the specimen masses m_1 and m_2 , the reference and specimen cooling rates

$\left(\frac{dT}{d\tau} \right)_1$ and $\left(\frac{dT}{d\tau} \right)_2$ and the specific heat capacity of the

reference specimen $C_{p_1}^0$ one can calculate the heat capacity of the other material $C_{p_2}^0$:

$$C_{p_2}^0 = \frac{C_{p_1}^0 m_1 \left(\frac{dT}{d\tau} \right)_1}{m_2 \left(\frac{dT}{d\tau} \right)_2}. \quad (7)$$

To confirm the validity of the above assumption, we measured specimen temperature vs cooling time curves for aluminum and copper [21]. The results proved to be in a good agreement with literary data [22].

Within this work we studied the effect of strontium on the heat capacity and change of thermodynamic functions of the AK1M2 alloy. The alloy specimens were prepared from A5N Grade ultrahigh-purity aluminum (99.999 % Al), single crystal silicon, copper and aluminum base master alloy containing 10.0 wt.% strontium. The contents of strontium in the alloy specimens were (wt.%) 0.01, 0.05, 0.1, 0.5 and 1.0. The master alloy was added to the alloy specimens in SShOL type open shaft furnaces. The alloy was then cast to 30×16 mm cylindrical specimens.

3. Results and discussion

The experimental specimen temperature vs time curves were described using equations of the following type:

$$T = ae^{-b\tau} + pe^{-k\tau}, \quad (8)$$

where a , b , p and k are the constants and τ is the cooling time.

Differentiating Eq. (8) with respect to τ we obtain the specimen cooling rate equation:

$$\frac{dT}{d\tau} = -abe^{-b\tau} - pke^{-k\tau}. \quad (9)$$

Using Eq. (7) we write the following equations for the temperature dependence of the heat capacity of the AK1M2 alloy:

$$C_p^0 = -961.11 + 5.33T - 3.9 \times 10^{-3}T^2 + 1.88 \times 10^{-6}T^3,$$

and for the strontium alloyed alloys (in wt.%):

– AK1M2 + 0.01 % Sr:

$$C_p^0 = -500.18 + 5.73T - 5.9 \times 10^{-3}T^2 + 2.57 \times 10^{-6}T^3;$$

– AK1M2 + 0.05 % Sr:

$$C_p^0 = -686.19 + 6.27T - 6.7 \times 10^{-3}T^2 + 2.92 \times 10^{-6}T^3; \quad (10)$$

– AK1M2 + 0.5 % Sr:

$$C_p^0 = -636.37 + 5.93T - 6.1 \times 10^{-3}T^2 + 2.66 \times 10^{-6}T^3;$$

– AK1M2 + 1 % Sr:

$$C_p^0 = -965.58 + 6.99T - 7.7 \times 10^{-3}T^2 + 3.39 \times 10^{-6}T^3.$$

Figure 1 shows temperature dependences of the specific heat capacity (in J/kg × K) of the AK1M2 alloy containing different quantities of strontium. It can be seen from Fig. 1 that the heat capacity of the alloys increases with temperature and decreases with an increase in the strontium concentration. To calculate the changes in the temperature dependences of the enthalpy, entropy and Gibbs

energy we used integrals of the specific heat capacity as written in the following equations:

$$S^0(T) - S^0(298.15 \text{ K}) = \int_{298.15}^T \frac{C_p^0(T)}{T} dT, \quad (11)$$

We obtained the following polynomials describing changes in the temperature dependence of the AK1M2 alloy enthalpy:

$$H^0(T) - H^0(298.15 \text{ K}) = -99095.92 - 961.11T + 5.33T^2 - 3.9 \times 10^{-3}T^3 + 1.88 \times 10^{-6}T^4,$$

and for the strontium containing alloys, wt.%:

$$\begin{aligned} & \text{-- AK1M2 + 0.01 \% Sr:} \\ & H^0(T) - H^0(298.15 \text{ K}) = -223956.5 - 500.18T + 5.728T^2 - 5.9 \times 10^{-3}T^3 + 2.567 \times 10^{-6}T^4; \\ & \text{-- AK1M2 + 0.05 \% Sr:} \\ & H^0(T) - H^0(298.15 \text{ K}) = -198738.9 - 686.19T + 6.27T^2 - 6.7 \times 10^{-3}T^3 + 2.92 \times 10^{-6}T^4; \\ & \text{-- AK1M2 + 0.5 \% Sr:} \\ & H^0(T) - H^0(298.15 \text{ K}) = -196838.8 - 636.37T + 5.931T^2 - 6.1 \times 10^{-3}T^3 + 2.657 \times 10^{-6}T^4; \\ & \text{-- AK1M2 + 1 \% Sr:} \\ & H^0(T) - H^0(298.15 \text{ K}) = -156311.1 - 965.58T + 6.991T^2 - 7.7 \times 10^{-3}T^3 + 3.396 \times 10^{-6}T^4. \end{aligned} \quad (12)$$

Figure 2 shows temperature dependences of the enthalpy (in kJ/kg) of the strontium containing AK1M2 alloy calculated using Eqs. (12). The enthalpy of the alloys increases with temperature and decreases with an increase in the strontium content. We obtained the following equations describing the changes in temperature dependence of the entropy of the AK1M2 alloy:

$$S^0(T) - S^0(298.15 \text{ K}) = -2727.02 - 961.11 \ln T + 10.67T - 5.85 \times 10^{-3}T^2 + 2.512 \times 10^{-6}T^3;$$

and for the strontium containing alloys, wt.%:

$$\begin{aligned} & \text{-- AK1M2 + 0.01 \% Sr:} \\ & S^0(T) - S^0(298.15 \text{ K}) = -2719.55 - 500.73 \ln T + 11.46T - 8.85 \times 10^{-3}T^2 + 3.423 \times 10^{-6}T^3; \\ & \text{-- AK1M2 + 0.05 \% Sr:} \\ & S^0(T) - S^0(298.15 \text{ K}) = -2951.72 - 686.19 \ln T + 12.55T - 10.05 \times 10^{-3}T^2 + 3.894 \times 10^{-6}T^3; \\ & \text{-- AK1M2 + 0.5 \% Sr:} \\ & S^0(T) - S^0(298.15 \text{ K}) = -2817.31 - 636.37 \ln T + 11.86T - 9.15 \times 10^{-3}T^2 + 3.543 \times 10^{-6}T^3; \\ & \text{-- AK1M2 + 1 \% Sr:} \\ & S^0(T) - S^0(298.15 \text{ K}) = -3261.93 - 965.58 \ln T + 13.98T - 11.55 \times 10^{-3}T^2 + 4.529 \times 10^{-6}T^3. \end{aligned} \quad (13)$$

Table 1 shows the experimental temperature dependence of the entropy (in J/kg × K) of the strontium containing AK1M2 alloy calculated using Eqs. (13). The entropy of the alloys increases with temperature and decreases with an increase in the strontium content to 0.5 wt.%.

The experimental temperature dependences of the Gibbs energy of the AK1M2 alloy and the strontium containing alloys (wt.%) are as follows:

$$\begin{aligned} & \text{-- AK1M2:} \\ & G^0(T) - G^0(298.15 \text{ K}) = -99095.92 + 1765.92T - 5.334T^2 + 1.95 \times 10^{-3}T^3 - 6.279 \times 10^{-7}T^4 + 961.11T \ln T; \\ & \text{-- AK1M2 + 0.01 \% Sr:} \\ & G^0(T) - G^0(298.15 \text{ K}) = -223956.5 - 2219.37T - 5.728T^2 + 2.95 \times 10^{-3}T^3 - 8.557 \times 10^{-7}T^4 + 500.19T \ln T; \\ & \text{-- AK1M2 + 0.05 \% Sr:} \\ & G^0(T) - G^0(298.15 \text{ K}) = -198738.9 + 2265.53T - 6.275T^2 + 3.35 \times 10^{-3}T^3 - 9.734 \times 10^{-7}T^4 + 686.19T \ln T; \\ & \text{-- AK1M2 + 0.5 \% Sr:} \\ & G^0(T) - G^0(298.15 \text{ K}) = -196838.8 + 2180.94T - 5.931T^2 + 3.05 \times 10^{-3}T^3 - 8.858 \times 10^{-7}T^4 + 636.37T \ln T; \\ & \text{-- AK1M2 + 1 \% Sr:} \\ & G^0(T) - G^0(298.15 \text{ K}) = -156311.1 + 2296.34T - 6.991T^2 + 3.85 \times 10^{-3}T^3 - 1.1322 \times 10^{-6}T^4 + 965.58T \ln T. \end{aligned} \quad (14)$$

Table 2 shows the experimental temperature dependence of the Gibbs energy (in kJ/kg) of the strontium containing AK1M2 alloy calculated using Eqs. (14).

High-purity aluminum alloys e.g. AK1M2 are recommended for electronics and semiconductor materials applications. This alloy is used for example in electron beam

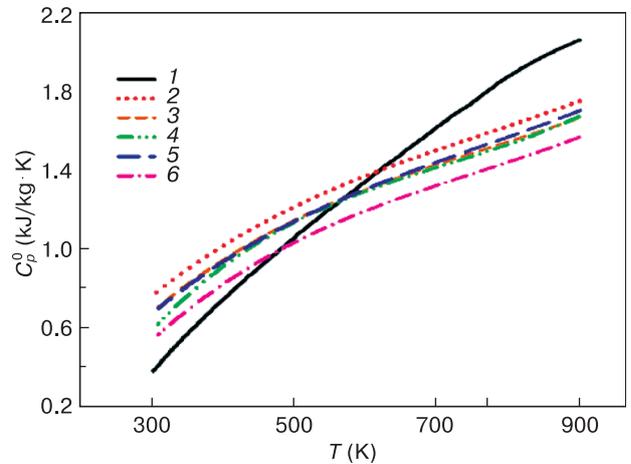


Figure 1. Temperature dependences of the specific heat capacity of the strontium containing AK1M2 alloy: (1) 0; (2) 0.01 % Sr; (3) 0.05 % Sr; (4) 0.1 % Sr; (5) 0.5 % Sr; (6) 1 % Sr.

Table 1. Temperature dependence of the entropy of the strontium containing AK1M2 alloy.

T, K	$S^0(T) - S^0(298.15), \text{kJ/kg} \times \text{K}$				
	AK1M2	AK1M2 + 0.01 % Sr	AK1M2 + 0.05 % Sr	AK1M2 + 0.5 % Sr	AK1M2 + 1 % Sr
300	14.51	13.10	14.03	13.58	15.33
400	764.67	665.82	709.64	690.42	772.56
500	1458.11	1223.69	1297.70	1269.31	1407.48
600	2109.90	1707.25	1801.58	1771.50	1947.27
700	2735.10	2137.02	2244.63	2218.26	2419.10
800	3348.78	2533.56	2650.22	2630.83	2850.15
900	3966.02	2917.38	2860.77	3030.49	3267.58

Table 2. Temperature dependence of the Gibbs energy of the strontium containing AK1M2 alloy.

T, K	$G^0(T) - G^0(298.15 \text{ K}), \text{kJ/kg}$				
	AK1M2	AK1M2 + 0.01 % Sr	AK1M2 + 0.05 % Sr	AK1M2 + 0.5 % Sr	AK1M2 + 1 % Sr
300	-1.79	-0.94	-1.28	-1.19	-1.80
400	-137.40	-85.78	-107.08	-100.93	-138.88
500	-345.06	-230.98	-276.86	-263.28	-345.35
600	-619.86	-428.07	-501.05	-479.51	-610.33
700	-958.38	-670.67	-772.38	-742.99	-925.65
800	-1358.72	-954.41	-1085.95	-1049.28	-1285.90
900	-1820.49	-1277.8	-1439.17	-1395.99	-1688.34

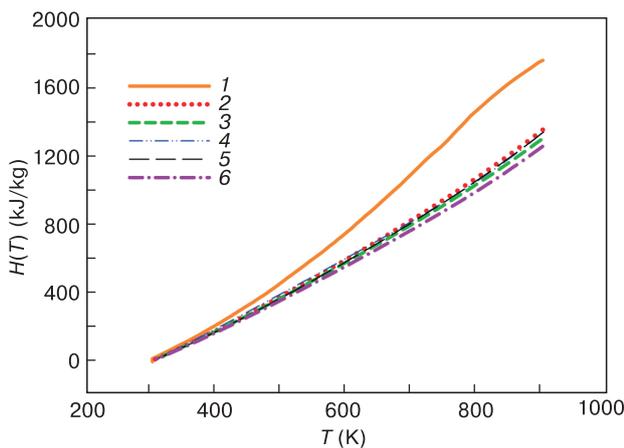


Figure 2. Temperature dependences of the enthalpy of the strontium containing AK1M2 alloy: (1) 0; (2) 0.01 % Sr; (3) 0.05 % Sr; (4) 0.1 % Sr; (5) 0.5 % Sr; (6) 1 % Sr.

tubes in the form of sheets, foil or wire. It is also used if cathode sputtering is undesired, e.g. for the fabrication of X-ray tube and cathode oscilloscope cathodes, high-voltage discharger electrodes and lamps. Due to their low density and low X-ray radiation intensity under electron bombardment, high-purity aluminum alloys are used in high-voltage electron beam devices as deflector plates and diaphragms.

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The AK1M2 ultrahigh-purity aluminum alloy is recommendable for transistor, diode and thermal resistor technologies for producing contact transient layers to germanium or silicon alloys [23–25].

4. Conclusion

The temperature dependence of the specific heat capacity and change in the thermodynamic functions of strontium-containing ultrahigh-purity aluminum base AK1M2 alloy have been studied in “cooling” mode over the 298.15–900 K range. Mathematical models describing the evolution of these properties of the alloys in this temperature range with change in alloying addition concentration have been obtained. The heat capacity, enthalpy and entropy of the alloys increase with temperature, decrease with an increase in the alloying addition concentration. The Gibbs energy of the alloys has an inverse dependence: it decreases with an increase in temperature and grows with an increase in the alloying addition concentration.

The decrease in the heat capacity of the alloys is accounted for by an increase in the heterogeneity of the alloys due to strontium alloying since strontium changes the crystallization mode of the aluminum solid solution.

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