

Concentration dependence of sodium alloys based on tin surface tension

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Abstract. The concentration dependence of the surface tension (ST) for alloys of the Sn-Na system in the range of compositions with a content of 0.06 to 5.00 at.% Na is studied by the large droplet method using high-purity components and a corresponding ST isotherm for $T = 573 \text{ K}$ is constructed. It has been established that small additions of sodium to tin significantly reduce ST of the studied melts. Calculations of sodium adsorption in alloys have shown that there is a maximum on the adsorption curve corresponding to alloys with a content of about 1.5 at.% Na in tin.

1. Introduction

Great interest in the study of alloys with the participation of alkali metals due to the possibility of their use in many areas of new technology. In particular, tin-sodium melts are considered as promising coolants for nuclear reactors, for the development and creation of new power-intensive chemical current sources, etc [1]. However, the thermophysical properties of alloys of the system Na-Sn is poorly studied [2, 3], in this connection it is necessary to obtain precise data on the thermophysical properties of this system. The present work is devoted to the study of the concentration dependence of the surface tension (ST) $\sigma(x)$ of sodium alloys on the basis of tin.

In the literature there are only a few papers devoted to the investigation of the ST of alloys of the Na-Sn system [4, 5]. The influence of sodium (from 0.03 to 4.6 at.% Na) on ST Sn was studied in [4]. But it has several disadvantages: a) in measurements of ST, the authors of [4] used tin of technical purity for the preparation of alloys; b) due to the lack of information about the density of Sn-Na alloys in the literature, which are necessary for the calculations of ST, the authors of [4] made a rough assumption: since the Na concentrations in the alloys with Sn are small, the density of alloys should not differ significantly from the density pure tin. In this approximation, the ST of all 10 sodium alloys studied with tin was calculated.

In the framework of a simple statistical model based on the formation of complexes and on the concept of the presence of a layered structure near the phase boundary, the isotherm of the Sn-Na alloys is plotted in [5] in the total composition interval, in which both the minimum and the maximum were found, figure 1. The data $\sigma(x)$ obtained in [5] for the concentration regions of the reciprocal dilute solutions of the Sn-Na system are quite strictly subject to the criteria for the surface activity of the components in binary metallic melts [6] and, in this respect, they should be considered correct. However, it is not possible to verify the reliability of the presence of a maximum and a minimum on the isotherm $\sigma(x)$, unfortunately, due to the lack of experimental data on $\sigma(x)$ of the Sn-Na system in the complete concentration range of the compositions. In the present paper, the task is set to refine the isotherm of



$\sigma(x)$ Na alloys based on Sn using the components of improved purity and modern techniques and experimental technology.

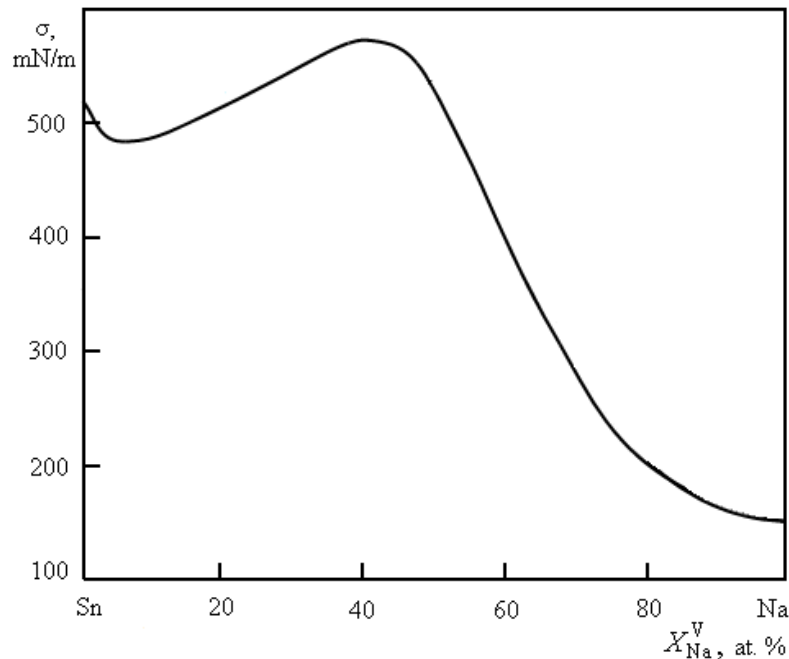


Figure 1. Concentration dependence of the surface tension (873 K) of alloys of the Sn-Na system [5].

The surface tension is determined by the method of the lying drop with an error of about 2% on samples of alloys from high-purity components (Sn-000 and 99.993% Na) in a vacuum of 10^{-4} Pa. The whole-soldered measuring cell used by us makes it possible to prepare alloys and measure ST in the same conditions of static vacuum without opening and depressurization in the course of experiments, and, most importantly, it ensures the thermodynamic equilibrium of the surfaces of alloys with its own saturated vapor. The features of the experimental setup and the measurement procedure are described in our papers [7, 8].

2. Results of the study ST sodium alloys based on tin

The results of measuring the ST of sodium based on tin alloys in the range of compositions studied by us graphically presented in figure 2. It shows that the obtained isotherm of the ST $\sigma(x)$ is a smooth falling curve, i.e. additions of sodium to tin lead to a significant decrease in the ST of alloys. In particular, the addition of 1 at.% Na to Sn reduces the ST of tin by 80 mN/m, i.e. by 15 %, which indicates a rather high surface activity, manifested by sodium at a low content in alloys with tin.

As noted above, there is only one work [4], devoted to the experimental study of the ST alloys of the Sn-Na system, whose data are also shown in figure 2 in the form of light circles. In figure 2 that our data on the isotherm $\sigma(x)$ are in qualitative agreement with the results of [4], but are located systematically somewhat lower than the $\sigma(x)$ isotherm obtained in [4]. The maximum deviation of the compared ST isotherms reaches 20 mN/m, which exceeds the experimental error by several times.

Using the ST $\sigma(x)$ isotherm obtained in the experiments, we estimated the limiting surface activity of sodium α_{Na} in alloys with tin at a temperature of 573 K:

$$\alpha_{\text{Na}} = \left(\lim_{\Delta x_{\text{Na}} \rightarrow 0} \frac{\Delta \sigma}{\Delta x_{\text{Na}}} \right)_T,$$

where $\Delta \sigma$ is the change in ST depending on the composition of the melt, and x is the concentration of sodium in atomic fractions.

It turned out that α_{Na} in alloys with tin it amounted to about 15 N/(m \times at. of a share), which indicates a higher surface activity exhibited by sodium with its small content in tin than the limiting surface activity α_{Na} calculated from the data of [4].

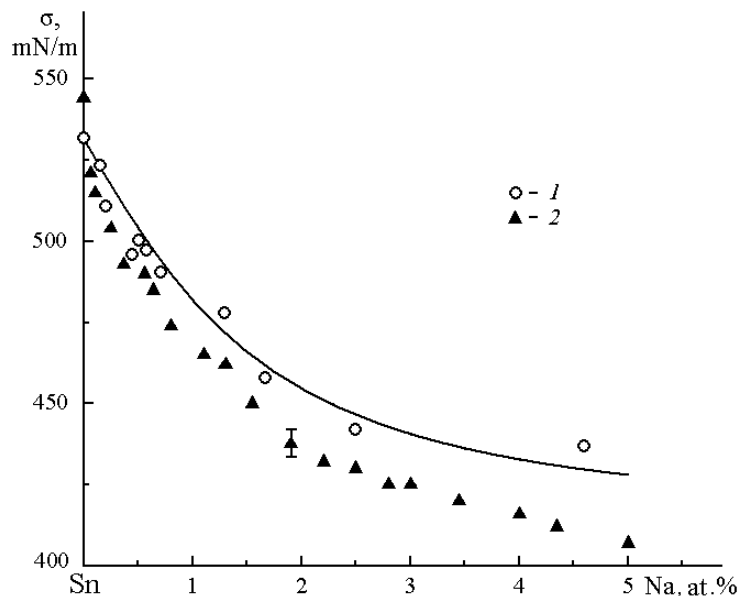


Figure 2. Concentration dependences of the surface tension of sodium alloys based on tin at a temperature of 573 K: 1 - data [4], 2 - our data.

Calculations of sodium adsorption in alloys with tin are carried out by the known Guggenheim-Adam formula in the N-variant by the method described in [9-11], using for this purpose the experimental data obtained on the isotherm of the ST $\sigma(x)$ of sodium alloys based on tin at 573 K:

$$\Gamma_2^{(N)} = -\frac{x_2(1-x_2)}{RT} \left(\frac{\partial \sigma}{\partial x_2} \right)_T$$

in which x_2 – are the atomic fractions of the second component (sodium) under the normalization condition $x_1 + x_2 = 1$, R is the universal gas constant, and T is the temperature in K.

We note that the derivatives $(\partial \sigma / \partial x_2)_T$ are determined until recently by "manual" graphical differentiation of the isotherm $\sigma(x)$, adhering to the "first-derivative step" principle and substituting them later into the Guggenheim-Adam equation. The main drawback of such calculations is the considerable error in the calculations of adsorption, which usually reaches 15% or more [11]. To reduce the error in calculating the adsorption of the components of alloys, we have used the method of calculating the adsorption of components, which we improved, by processing the experimental data of the concentration dependence of the ST $\sigma(x)$ by the method of least squares and then differentiating it in the MathCAD package [12]. In this case, a general regression is used representing a linear combination of power functions x^m , $m=1, 3$ and an isobaric hyperbola $1/(x+\alpha)$, where the numerical parameter α is determined from the condition of regression convergence to the data obtained experimentally. As a result, the error in calculating the adsorption of components in metallic melts decreased several times by a new method and, importantly, it is possible to eliminate the timeconsuming and inefficient procedure for graphically differentiating the isotherm of the ST $\sigma(x)$.

The data of a large number of computational experiments make it possible to assert that the proposed method and procedure of approximation make it possible not only to minimize the errors in determining

the adsorption of the components of the solutions under study, but also to check the informativeness and accuracy of data processing.

From the data obtained, it follows that sodium in the region of its small content in alloys with tin exhibits a high surface activity, the maximum value of which is achieved in the composition range of ~ 1.5 at. % Na in tin, figure 3. The greatest value of adsorption obtained in this work is 10% higher than the adsorption maximum found earlier by Pokrovsky et al. at [4].

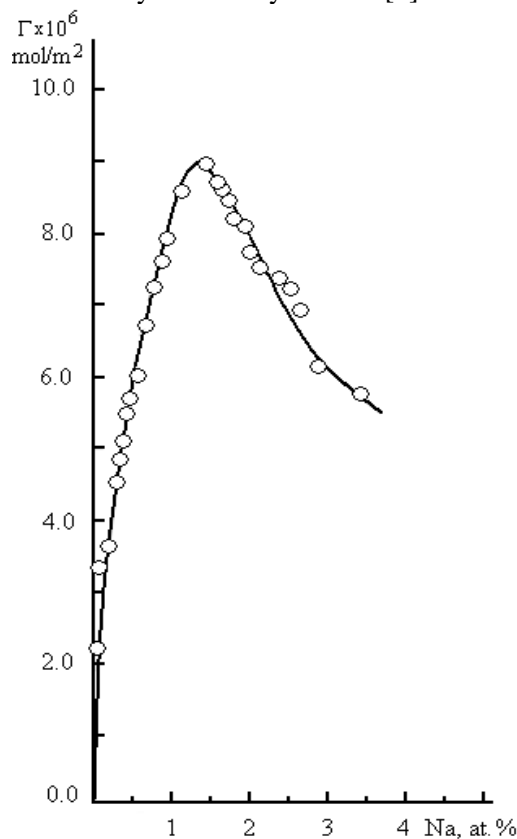


Figure 3. Results of calculations of sodium adsorption in alloys based on tin.

3. Conclusions

Under conditions of a high static vacuum, the surface tension of high purity tin, as well as of its 19 alloys with a content of 0.06 to 5.0 at.% Na, was measured and a corresponding isotherm $\sigma(x)$ was constructed for a temperature $T = 573 \text{ K}$. It has been shown that the limiting surface activity of sodium in alloys with tin is about $15 \text{ N/(m} \times \text{at. of a share)}$. Sodium in the region of its small content in alloys with tin exhibits a high surface activity, the maximum value of which is achieved in the composition region with a content of about 1.5 at.% Na in Sn, which is in agreement with the rule of generalized moments in Semenchenko and Zadumkin, as well as with other criteria for the surface activity of components in binary metallic systems.

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