Concentration dependence of surface properties and molar volume of multicomponent system indium-tin-lead-bismuth

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Abstract. The results of an experimental research of surface properties of the four-component system indium-tin-lead-bismuth are presented. The researches under discussion were carried out in a combined device in which the surface tension ($\sigma$) is measured by the method of maximum pressure in a drop, and density ($\rho$) is measured by advanced aerometry. Measurement errors are 0.7 % for surface tension measurement, and 0.2 % for density measurement. The study of the concentration dependence of $\sigma$ in this system has revealed the influence of the third and fourth components upon the characteristics of surface tension isotherms of the binary system indium-tin. It was found out that with an increase in the content of the third and fourth components the depth of the minimum on the surface tension isotherms of the indium-tin system decreases. On the basis of the concentration dependence of $\sigma$ the phenomenon of concentration bufferity is revealed. It is shown that despite the complex character, isotherms of $\sigma$ on beam sections of a multicomponent system do not contain qualitatively new features in comparison with the isotherms of these properties in lateral binary systems.

1. Introduction.
A lot of experimental data on the properties of pure metals and binary systems have been collected by now [1]. Theoretical equations [2,3-4] have been received allowing to describe the concentration dependence of surface tension of various binary systems. Information about temperature and concentration dependences of surface tension of metals and binary alloys makes it possible to define the adsorption of components, surface entropy and composition, minimally possible and effective thickness of a surface layer [6].

However, theoretical and experimental research of the surface properties of multicomponent systems is rather insufficient. There is no strict theory for a quantitative description of the dependence of surface tension on concentration and temperature. The available theoretical equations either contain physical values the experimental definition of which presents great difficulties, or in many cases do not describe the surface properties of alloys even qualitatively. Strict methods allowing calculation of thermodynamic parameters of the surface layer of multicomponent alloys on the basis of the concentration dependences of surface tension and molar volumes are not developed.
At the same time, most materials used nowadays are multicomponent alloys, which makes an overall study of their properties essential. In the process of solving many technological problems it is necessary to consider the influence of several components on the properties of materials, but surface-active substances even in rather small quantities can considerably change the structure and properties of surface layers [7]. Therefore, the study of surface properties of multicomponent systems was and remains to be of vital importance.

2. Surface tension and molar volume in the indium-tin-lead-bismuth system.
The experimental research of the surface tension of the indium-tin-lead-bismuth system was performed in a combined device in which the surface tension is measured by the method of maximum pressure in a drop, and density is measured by advanced aerometer. The metals used in the experiments are indium In-000; tin - 000; lead Pb-0000; bismuth Bi-000. This system was chosen because the surface tension isotherms of the lateral binary system indium-tin had a complex character. In particular, the concentration dependence of the surface tension is characterized by a flat minimum in the vicinity of the equiatomic concentration. Other lateral binary systems are characterized by smooth isotherms of surface tension. Besides the binary systems studied by us, we used the data on the binary and ternary systems forming the In-Sn-Pb-Bi system which were obtained in our laboratory before.

The calculations made for binary and ternary systems In-Sn-Bi; In-Pb-Bi; Sn-Pb-Bi [8] have shown that the concentration dependence of surface tension is well described by a polynomial of the third degree. Therefore, we used this model for the research of a four-component system [9]. We use the method of a simplex lattice to define the factors of the polynomial. Fig. 1. shows the points of the plan, which concludes the units of a simplex matrix for a model of the third order. Data on the surface tension of four-component alloys are necessary to analyze and verify the adequacy of the equation of regression. To solve this problem we studied the temperature dependences of the surface tension and density of seventeen alloys whose structures lay on the sections In:Sn:Pb=1:1:1.

The results of the calculations have shown that the concentration dependences of the surface tension of the system in question has some features inherited from the minimum on the surface tension isotherms of the lateral binary system In-Sn. This fact makes it necessary to verify the accuracy of the received equation.

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**Figure 1.** The arrangement of points of the plan of experiment in the units of a simplex matrix for a model of the third order.

**Figure 2.** Isotherms of the surface tension and molar volumes in system indium-tin-lead – bismuth
After comparing the experimental isotherms of surface tension and the ones that were calculated on the basis of the received polynomials, we see in fig 2 that the calculated values and experimental data coincide within the error of measurement.

Besides, the chosen model completely describes the influence of the fourth component on the position of extremums in the isothermal surfaces of the three-component systems In-Sn-Pb and In-Sn-Bi surface tension. Hence, the model of the third order adequately reflects the real dependence of surface tension on the composition in the four-component system under discussion. A common feature of the isothermal $\sigma$ surfaces of lateral three-component systems is a "hollow" in the area of joining the indium-tin components. The study of the influence of the fourth component on this phenomenon is of great interest.

Fig.3 displays the isolines of the surface tension in the system under study with a constant content of bismuth. These diagrams clearly show the influence of bismuth content on the isothermal $\sigma$ surface in the three-component system indium-tin-lead.

It was found out during the experiment that the "hollow" on the isothermal $\sigma$ surface of the In-Sn-Pb system smoothes out when bismuth content is low (less than 0.05 of molar fraction). The further increase in the content of bismuth does not change isothermal surfaces of $\sigma$ considerably.

For the indium-tin-bismuth system isothermal $\sigma$ surfaces smooth out only when the content of lead is more than 0.1 of molar fraction. It is due to the fact that the limiting surface activity of lead in indium-tin-bismuth alloys is lower than that in the indium-tin-lead system.

![Figure 3](image-url)

**Figure 3.** The isolines of the surface tension (mN/m) in indium-tin-lead-bismuth system at 623 K and $x_{Bi} =$: a-0.0; b-0.2; c-0.4 of molar fraction.
Thus, the influence of the third or fourth component on the isotherm of surface tension in a four-component system depends on their limiting surface activity. Any signs of mutual influence of impurities have not revealed. Joint influence of the third and fourth components on the "hollow" in the surface tension isotherm of indium-tin system is defined by their total concentration in the alloys. At an increase of the content of bismuth and lead in the indium-tin-lead-bismuth system the process of alignment of the relief of the isothermal $\sigma$ surface occurs.

Molar volumes of In-Sn-Pb-Bi alloys have positive deviations from additive values (fig.2). However, in the absolute value they do not exceed the deviation of molar volumes from additivity in lateral binary systems. Isotherms of surface tension and molar volumes in the system under study do not contain qualitatively new features in comparison with the isotherms of these properties in lateral binary systems. Another important result is that the surface tension of four-component systems is well described by the equation received for ideal solutions. Thus, the more components there are in the alloys, the closer are the theoretical and experimental values. Hence, in this system with an increase in the number of the components the surface layer of the alloys comes nearer to the ideal one. The temperature dependences of the surface tension and density in the alloys under study are linear with the negative temperature factor.

4. References

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