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Abstract

Full Text

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SURFACE TENSION OF MELTS OF THE INDIUM—ANTIMONY SYSTEM

(Presented by Academician I. I. Chernyaev, May 20, 1962)

Establishing a regular correspondence between the singular points of the phase diagram and of the surface-tension—composition diagram is of interest, since it would make it possible to investigate more deeply the connection between the structure of the liquid and solid phases, as well as between the surface and bulk properties of a substance (1-11). Recently several works have been published (12-14) that refute the opinion that the surface tension of liquid metals is not a structure-sensitive property.

Fig. 1. Temperature dependence of the surface tension of indium. *1*—data of Melford and Hoar (16), *2*—data of P. P. Pugachevich and O. A. Timofeevicheva (17), *3*—data of the authors.

The study of the surface tension of indium—antimony melts over a wide concentration range was carried out by us in order to determine whether, in this system with the congruently melting intermetallic compound indium antimonide, there exists a correspondence between the singular points on the phase diagram and on the surface-tension—composition diagram, and also to obtain experimental data on the surface tension of melts whose composition differs from the stoichiometric one. The latter is of practical importance in connection with the growth of single crystals of indium antimonide from melts, the composition of which does not remain constant during crystal growth.

Measurements of the surface tension (σ) were made by the maximum-pressure method in a gas bubble, essentially in the manner described in (15). In our work, the formation of gas bubbles was carried out at the ends of sharply pointed quartz capillaries with a wall thickness at the cut of 0.01 mm, i.e., about 1%

Fig. 2

Figure 2: Fig. 2

Fig. 3

Figure 3: Fig. 3

of the smaller diameter of the capillary, which eliminated difficulties associated with selecting the effective capillary radius for calculating σ. The measurements were conducted in an argon atmosphere at a pressure of ~ 100 mm Hg. The maximum pressure in the gas bubble was determined with a U-shaped manometer using a KM-10 cathetometer. Polyphenylmethylsiloxane (PFMS-2) vacuum oil, possessing low viscosity and low vapor pressure, was used as the manometric liquid. The total maximum error

was 1.2%, i.e., about 7.0 dyn/cm. Since all measurements were carried out with the same capillaries, the error in determining the effective radii of the capillaries (1%) could not have had a significant effect on the relative positions of the experimental points. To prepare the alloys, indium of 99.999% purity and antimony of 99.995% purity were used, as well as crystals and single crystals of indium antimonide grown by us from the melt by the Czochralski method.

Measurements of the value of σ for the melts were begun by studying the temperature dependence of the surface tension of indium in the temperature range 200–800°. In the literature there were no data on the dependence of σ of indium on temperature in such a broad temperature interval; as for the temperature range below 500°, then, as can be seen from Fig. 1, our data are in good agreement with the data (16, 17).

Fig. 2. Temperature dependence of the surface tension of antimony according to the data of Birkumshaw (1) and according to the authors' data (2).

Subsequently, the surface tension of antimony was also measured in the temperature range 650-800°, and the data obtained (Fig. 2)

Fig. 3. Dependence of the surface tension of melts on temperature. Concentration of antimony in at.%:
1 –0.00; 2 –1.24; 3 –3.83;
4 –8.97; 5 –12.92; 6 –16.60; 7 –19.70; 8 –24.5; 9 –29.7; 10 –32.3;
11 –34.7; 12 –37.2; 13 –40.1; 14 –42.4; 15 –44.5; 16 –46.3;
17 –47.9; 18 –49.2; 19 –50.0; 20 –50.9; 21 –52.1; 22 –54.4;
23 –58.5; 24 –62.9; 25 –66.8; 26 –69.4; 27 –85.1; 28 –100.

practically coincided with the data of (9). The surface tension of indium–antimony melts was studied in the temperature interval 550–700°. In all, the temperature dependence was studied for 28 alloys of different concentrations—from pure indium to pure antimony (Fig. 3). Over the entire temperature interval investigated, antimony proved to be surface-active with respect to indium.

Fig. 4. Surface tension of melts in the indium—antimony system. 1 —at 550°; 2 —at 700°

Figure 4: Fig. 4. Surface tension of melts in the indium—antimony system. 1 —at 550°; 2 —at 700°

According to our data, the surface tension of indium antimonide at a temperature of 550° was 418 dyn/cm. In addition, experiments were carried out in which a rod of single-crystal indium antimonide of known diameter was melted at one end, and from the weight of the detached drops of melt it was possible to calculate σ of indium antimonide at the melting point, using the Lohnstein—Harkins corrections (1). The surface tension of indium antimonide determined in this way was 425 ± 20 dyn/cm.

Fig. 4. Surface tension of melts in the indium—antimony system. 1 —at 550°; 2 —at 700°

As can be seen from Fig. 4, where the isotherms of surface tension are shown, the existence in the indium—antimony system of the intermetallic compound indium antimonide is reflected in the surface-tension isotherms the more clearly, the closer the temperature is to the melting point of indium antimonide.

It is interesting to note that, according to the data of work (18), where the viscosity of indium—antimony melts was studied, in the concentration region corresponding to the formation of indium antimonide, clearly expressed maxima were observed on the viscosity isotherms. As can be seen from Fig. 4, with an increase in the antimony content in the melts in the concentration region close to the intermetallic compound, the value of the derivative of the surface tension with respect to concentration changes rather sharply, passing in absolute value through a minimum. This change in the value of $\partial\sigma/\partial C$ should be reflected in the adsorption isotherm (Γ_{Sb}) of antimony on the surface of the melt, since the formula for calculating Γ_{Sb} has the form:

$$\Gamma_{\mathrm{Sb}} = -\frac{1}{RT} C(1-C) \frac{\partial\sigma}{\partial C} \frac{1}{1 + \dfrac{\partial \ln f}{\partial \ln C}}, \tag{1}$$

where f is the activity coefficient, $\Gamma_{\mathrm{Sb}} + \Gamma_{\mathrm{In}} = 0$.

Using the data of work (19) on the activities of the components in indium—antimony melts, with the aid of formula (1) it is easy to show that the adsorption of antimony, as the concentration changes from 0 to 100%, passes twice through a maximum value, while in the concentration region close to the stoichiometric composition a relative minimum of adsorption is observed, which apparently is connected with the existence in the melt of a certain tendency toward the formation of a quasi-ordered structure. From Fig. 4 it is seen that σ of indium antimonide melts alloyed with antimony is smaller than σ of indium antimonide melts alloyed with indium.

In conclusion, we express our gratitude to M. S. Mirgalovskaya for her constant interest in the work.

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Note: Figure translations are in progress. See original paper for figures.