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Fig. 1

Figure 1: Fig. 1

Abstract

Full Text

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THE DEPENDENCE OF THE SURFACE TENSION OF THALLIUM ON TEMPERATURE

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The investigation of the temperature dependence of the surface tension σ of thallium was carried out by us in order to resolve certain difficulties associated with the fabrication of semiconductor devices (^{1}). The surface tension of thallium had previously been studied in the work of Sauerwald and Schmidt (^{2}), but these authors indicate that they were unable to obtain results reproducible in time; namely, σ of thallium increased over the course of 1-2 hr from 357.0 to 496.0 dyn/cm at 320°, which apparently was connected with methodological shortcomings.

Fig. 1

To measure the temperature dependence of σ of thallium we used the maximum-pressure method, which is well substantiated theoretically. The theory of this method, developed by Cantor (^{3}), requires the use of capillaries whose wall thickness at the cut is infinitely small. In view of the high toxicity of thallium, we decided to investigate its surface tension in a sealed glass apparatus. However, the fabrication from glass of capillaries satisfying the requirements of the theory (^{3}) is very difficult. If, for measuring σ of liquids that do not wet the capillary material, one uses the maximum-pressure method in a drop, and for measuring σ of liquids that wet the capillary material, one applies the method of maximum pressure in a bubble, then capillaries with a finite wall thickness may be used.

Since the wettability of glass by thallium at different temperatures was unknown to us, an uncertainty arose in choosing between the two variants of the maximum-pressure method. To resolve these difficulties, one of us (O. A. Timofeevicheva) constructed a sealed glass rotating apparatus which, unlike those previously described in the literature (^{4,5}), made it possible not only to carry out measurements of σ by the maximum-pressure method in a drop, but also, at any stage of the experiment, to switch to measuring σ by the maximum-pressure method in a gas bubble and back again, without opening the apparatus.

After careful high-vacuum treatment of the apparatus (Fig. 1), mounted on a rotating frame, the required amount of the molten metal under investigation is transferred through tube 18 into vessels 1 and 2, and the tee is sealed off along the line 19—19′; through tube 3 the apparatus is filled ···

with inert gas to a pressure slightly below atmospheric, is sealed off from the vacuum apparatus along line 4—4′, and is placed in an air thermostat. To measure the surface tension by the method of maximum pressure in a gas bubble, the instrument is rotated in the plane of the drawing clockwise so that part of the metal flows through tube 5 into reservoir 6 and closes the end of capillary 12, and through tube 7 into reservoir 8 and from it into manometer 9. After the instrument is returned to its initial position, the metal remaining in vessel 2, by rotating the instrument about the axis XX_1 away from oneself through an angle somewhat exceeding 90°, is poured into vessel 1 through tubes 10, 11, and the instrument is returned to the position shown in the drawing. Then the metal, under the action of gravity, flowing through constriction 13, gradually closes the openings of tubes 11 and 7, and a closed volume of gas is created in the instrument, bounded by the surfaces of the metal in vessel 2, capillary 12, and bend 14 of manometer 9. As the metal flows into vessel 2, the pressure in the indicated volume increases, while in reservoir 6 and the space connected with it it decreases; at the cut of capillary 12, located in the liquid, a gas bubble is formed. The pressure difference in the bubble and above the surface of the metal in vessel 6, which is recorded by manometer 9, leads to gas bubbles beginning to detach from the tip of capillary 12 when the maximum pressure is reached. When the instrument is rotated about the axis XX_1 away from oneself through an angle slightly greater than 90°, the pressure in all its parts is equalized, and the metal from vessel 2 flows into vessel 3. When the instrument is returned to its initial position, the measurements can be repeated, and this can be done many times. Knowing the distance from mark 15 to the cut of capillary 12, and measuring the distance from the same mark to the metal level in vessel 6, one can determine the depth of immersion of the capillary. To calculate σ by Cantor' s formula, it remains to add the difference of the liquid levels in manometer 9, the radius of capillary 12, and the density of the liquid under investigation at the measurement temperature.

The transition to measurement by the method of maximum pressure in a drop is carried out by simply rotating the instrument about the axis X—X_1 away from oneself through 180°. The metal from vessel 2, now located in the upper part of the instrument, will slowly enter, through tube 16 and constriction 17, capillary 12 and manometric tube 8. In this case the gas pressure in all parts of the instrument, owing to the system of connecting tubes, is identical and constant. When the maximum pressure in the drop is reached, droplets of liquid begin to fall from the tip of capillary 12. Having measured the height of the metal column corresponding to this pressure, equal to the distance from the cut of capillary 12 to the surface of the liquid in manometric tube 8, and also knowing the density of the liquid at the temperature of the investigation and the radius of the capillary, one can calculate the surface tension by Cantor' s formula. To

Fig. 2

Figure 2: Fig. 2

repeat the measurements, the instrument is tilted away from oneself through an angle of 90°; in this case the liquid from vessels 8 and 6 flows into vessel 2; the instrument is rotated through 90° in the reverse direction, which makes it possible to proceed to the next measurement.

In many cases, in the course of an experiment it becomes obvious that the metal under investigation, introduced into the instrument, does not wet the glass; then, to simplify the work of measuring σ, there always remains the possibility of sealing off the instrument from the vacuum apparatus before filling it with inert gas and carrying out the measurements only by the method of maximum pressure in a drop under high-vacuum conditions, i.e., in the same way as in the gravitational instrument (^{4}). Precisely such a possibility arose in the investigation of the surface tension of thallium.

After the molten metal, filtered in a vacuum of $\sim 1 \cdot 10^{-5}$ mm Hg from oxides, had flowed into the instrument, it was found that in the temperature interval 303—500° thallium does not wet glass. This allowed us, in the present case, to seal off the instrument from the vacuum apparatus without filling it

with an inert gas, and to carry out, by the maximum-bubble-pressure method, measurements of σ for liquid thallium in vacuum in equilibrium with its own saturated vapor.

The density values for molten thallium at the temperatures of the investigation, needed for calculating σ by Cantor' s formula, were taken from work (^{6}).

In contrast to the results obtained by Sauerwald and Schmidt (^{2}), the surface tension of thallium in our experiments, which were carried out over a period of 7 days, remained unchanged. The thermal dependence of σ for thallium in the temperature interval investigated, from 310 to 500°, proved to be linear and can be represented by the formula

$$\sigma = 464.5 - 0.080\,(t - 303). \tag{1}$$

The constants of equation (1) were calculated by the method of least squares; the largest deviation of the experimental points from the straight line expressed by equation (1) is less than 0.2 dyn/cm, i.e. $\approx 0.05\%$.

Fig. 2

As can be seen from Fig. 2, which contains the results of our investigation, as well as the temperature dependences of the surface tension of gallium and indium according to data (^{7,8}), the surface tension of the elements of the gallium subgroup decreases with increasing atomic weight, analogously to the elements of the alkali-metal subgroup (⁹).

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