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Soviet-era science, translated into English

# CHEMISTRY

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1960

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Fig. 1

Figure 1: Fig. 1

**Abstract****Full Text****CHEMISTRY****O. A. TIMOFEEVICHEVA and P. P. PUGACHEVICH****TEMPERATURE DEPENDENCE OF THE SURFACE TENSION OF GALLIUM***(Presented by Academician I. I. Chernyaev, 27 IV 1960)*

The surface tension ( $\sigma$ ) of gallium was first measured by Richards and Boyer <sup>(1)</sup>, who, using the sessile-drop-shape method, found that  $\sigma$  of gallium at 30°C is equal to 358.2 dyn/cm. Later Frumkin and Gorodetskaya <sup>(2)</sup> pointed out that this value is too low and determined it at the boundary with an electrolyte solution, at the maximum of the electrocapillary curves, to be 592 dyn/cm. Mack, Davis, and Bartell <sup>(3)</sup>, by the drop-shape method, found that  $\sigma$  of gallium in an atmosphere of hydrogen and carbon dioxide at a temperature of 30.5–40° is equal to 735 dyn/cm. Recently Korolkov <sup>(4)</sup>, using the method of maximum pressure in a gas bubble, determined that at a temperature exceeding the melting point of gallium by 50–80°, its surface tension is equal to 715 dyn/cm.

**Fig. 1**

We measured the surface tension of gallium in vacuum by the method of maximum pressure in a drop over the interval from 30 to 500°. The measurements were carried out in a gravitational apparatus, which was described in <sup>(5)</sup>. For this purpose a glass system was assembled, schematically shown in Fig. 1. After about 10 cm<sup>3</sup> of solid metallic gallium had been loaded into funnel 1, tube 2, containing this funnel, was sealed, and a high vacuum was created in the system. After prolonged thermal-vacuum treatment of the gravitational apparatus 3 and reservoir 4, the gallium in funnel 1 was melted with the aid of a cylindrical electric furnace placed over tube 2 and, being filtered from oxides, flowed through the narrow end of funnel 1 and tube 6 into beaker 5, in which it was then heated to a high temperature and held at this temperature for several hours. This promoted degassing of the metal, partial destruction of organic compounds contaminating the gallium, and distillation of some volatile impurities. As our experience shows, degassing and purification from impurities were especially significant if the metal was heated in quartz apparatus at 1000°C.

After heating the metal, the gravitational apparatus 3 with reservoir 4 was sealed off from the vacuum installation along the lines 7–7, 8–8, then the molten-

Fig. 2. Temperature dependence of the surface tension of gallium

Figure 2: Fig. 2. Temperature dependence of the surface tension of gallium

Fig. 3. Temperature dependence of the surface tension of tin and its alloys with tellurium according to the data of Pokrovskii and Kristian

Figure 3: Fig. 3. Temperature dependence of the surface tension of tin and its alloys with tellurium according to the data of Pokrovskii and Kristian

The refined gallium from beaker 5 was poured into gravitational apparatus 3; the latter, while hot, was sealed off from reservoir 4 along line 9–9, and the surface tension of gallium was measured in the apparatus analogously to the way this was done in the investigation of the surface tension of indium (<sup>5</sup>).

**Fig. 2.** Temperature dependence of the surface tension of gallium

In calculating the surface tension it was necessary to know the density of gallium at different temperatures, but we did not carry out these measurements and used the data of Spells (<sup>6</sup>).

**Fig. 3.** Temperature dependence of the surface tension of tin and its alloys with tellurium according to the data of Pokrovskii and Kristian. Tellurium concentration in atomic percent: 1–0.0037; 2–0.0046; 3–0.0104; 4–0.0121; 5–0.0140; 6–0.0177; 7–0.0364; 8–0.0289; 9–pure tin

**Table 1**

**Temperature dependence of the surface tension of gallium**

Temperature, °C	Surface tension, dyn/cm: $\sigma_{\text{exp}}$	Surface tension, dyn/cm: $\sigma_{\text{eq}}^{(1)}$	$\Delta\sigma = \sigma_{\text{exp}} - \sigma_{\text{eq}}^{(1)}$
30	707.0	706.6	+0.4
46	706.8	706.6	+0.2
63	706.1	706.5	–0.4
77	706.1	706.4	–0.3
93	706.3	706.2	+0.1
101	704.9	706.1	–1.2
132	704.9	705.5	–0.6
166	704.6	704.8	–0.2
196	705.2	703.8	+1.4
199	704.0	703.7	+0.3
247	703.0	701.9	+1.1
288	700.3	700.0	+0.3
338	695.9	697.3	–1.4
384	693.5	694.3	–0.8
424	690.9	691.4	–0.5
473	687.9	687.4	+0.5

Temperature, °C	Surface tension, dyn/cm: $\sigma_{\text{exp}}$	Surface tension, dyn/cm: $\sigma_{\text{eq}}^{(1)}$	$\Delta\sigma = \sigma_{\text{exp}} - \sigma_{\text{eq}}^{(1)}$
505	684.7	684.5	+0.2

$$\Delta\sigma_{\text{av}} = \pm 0.6 \text{ dyn/cm}$$

The gallium of 99.9% purity investigated by us contained very small amounts of both surface-active and surface-inactive impurities. The results of our measurements of  $\sigma$  for gallium are placed in Table 1 and presented in Fig. 2. The temperature dependence of gallium, as can be seen, is not linear and may be represented by the equation

$$\sigma = 706.6 - 0.000647(t - 29.79) - 0.0000965(t - 29.78)^2, \quad (1)$$

where  $t$  is the temperature in °C; 29.78 is the melting temperature of gallium according to the data of Rezer and Hofmann (7).

As is evident from equation (1), the temperature coefficient of the surface tension of gallium not only depends on temperature but also proves to be surprisingly small in comparison with  $d\sigma/dT$  for other metals.

A nonlinear dependence of surface tension on temperature has been observed for many metals, for example, for cadmium (8,9), mercury (8,10-13), bismuth (14), copper (15), antimony (9,16), sodium (17), and selenium (18). The most probable cause of this phenomenon, in those cases where the measurement methods employed and the experimental conditions give no cause for doubt, should be considered the presence of surface-active impurities in the metal under investigation.

Fig. 4. Periodic dependence of certain properties of elements on the ordinal number in D. I. Mendeleev's table

This was convincingly shown by Pokrovskii and Kristian, who found (19) that small additions of tellurium to tin lead to a sharp change in the analytical dependence of surface tension on temperature, and that a maximum appears on the surface-tension polytherm of such alloys (Fig. 3). Metzger (20) has recently established that the temperature coefficient of surface tension  $d\sigma/dT$  for pure copper is negative; however, additions of lead to copper change the sign of  $d\sigma/dT$ .

Fig. 4. Periodic dependence of certain properties of elements on the ordinal number in D. I. Mendeleev's table

Figure 4: Fig. 4. Periodic dependence of certain properties of elements on the ordinal number in D. I. Mendeleev's table

These examples indicate that the study of the surface tension of ultrapure metals and the influence on their  $\sigma$  of small additions represents a considerable—

...of theoretical interest and has important practical significance for new fields of technology. For us, the question was also of interest as to the extent to which the data we obtained for the surface tension of gallium corresponded to the periodic dependence of surface tension on the ordinal number of the element in the periodic system of the elements. The periodicity of the change in the specific weights of the elements as a function of atomic numbers was pointed out already by D. I. Mendeleev. Later this dependence was found for effective atomic and ionic radii, melting temperatures of the elements, their ionization potentials, and other properties. In 1914 Smith<sup>(21)</sup> established an analogous dependence also for the surface tension of the elements, which was subsequently confirmed in works<sup>(22–25)</sup>. At present, the dependence of the surface tension of the elements on their position in the periodic system of D. I. Mendeleev can be shown more clearly (Fig. 4). In Fig. 4 we give not only experimental data, but also the most probable values of  $\sigma$  for certain metals, calculated by Taylor<sup>(26)</sup> from various, mainly thermodynamic, relationships. It is seen from Fig. 4 that the surface tension of the elements, as well as such of their properties as density and reciprocal compressibility, is a periodic function of the atomic number. The extremal regions of the indicated properties, as is evident from Fig. 4, fall on the same groups of elements. From comparison of these dependences it followed that the surface tension of gallium should be higher than  $\sigma$  of germanium, but lower than the surface tension of zinc, which was confirmed by our experiments. Earlier, in studying the surface tension of indium<sup>(5)</sup>, proceeding from the periodic dependence of  $\sigma$  of the elements on ordinal number, we expected that  $\sigma$  of indium should be approximately the same as the surface tension of cadmium and tin, which also found its experimental confirmation<sup>(5,27)</sup>.

These examples are not the only ones; the general regularities existing between surface tension and other properties of the elements as a function of their atomic number point to a profound connection between the surface and bulk properties of matter.

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24 IV 1960

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