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MEASURING THE MELTING POINTS OF ALUMINUM
AND COPPER AT PRESSURES UP TO 18,000 KILOGRAMS
PER CENTIMETER

- USSR -

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Until recently, our information on the effect of super high pressure upon the melting point of metals was limited to the data obtained by Bridgeman /1,2/, who investigated certain low-melting metals (potassium, sodium, lithium, rubidium, caesium, mercury and gallium) at pressures up to 12,000 kilograms per square centimeter and bismuth up to 17,000 kg/cm².

Since 1953, the Institute of Crystallography, Academy of Sciences USSR, has been conducting investigations on the phase transformation of metals under conditions of super high pressure. The developed method for obtaining and measuring a high temperature inside a conduit of a vessel under super high pressure made it possible to widen the temperature range at the investigation of the fusion of metals under super high pressure /3/. The melting points of tin and lead at pressure up to 34,000 kg/cm² /4/, antimony, cadmium, tin and thallium at pressures up to 30,000 kg/cm² /5/, and bismuth at pressures up to 30,000 kg/cm² /6/ were measured. The above measurements were conducted on metal specimens placed in a heated crucible, in an isopentane medium in a super high-pressure intensifier canal. The melting points of the above-mentioned metals, with the exception of bismuth, gallium and antimony, rise with the pressure. In the case of bismuth and gallium, the melting point drops only until the triple point: alpha-phase-beta-phase-fusion is reached.

In the case of antimony, the lowering of the melting point is observed during the entire pressure interval under investigation.

Data on the measurements of the melting points of germanium at pressures up to 180,000 kg/cm², carried out, apparently, in apparatus for the synthesis of diamonds, were published in 1955 /7/. The germanium sample was placed into a

small graphite cylinder, which at the same time had the function of the heating element. The pressure was determined with the aid of a calibration curve, plotted on the polymorphic transformation points of bismuth, thallium, caesium and barium. The temperature was measured with a platinum-platinrhodium thermocouple. It was found that the melting point of germanium is reduced throughout the pressure interval.

Experimental Part

The present work sets forth the results of measuring the melting points of aluminum and copper at temperatures up to 18,000 kg/cm². To obtain temperatures above 600°C in the intensifier canal in the isopentane medium is difficult, due to the fact that the isopentane cracks and the coke liberated during this process closes the turn of the heating coil.

Therefore, on carrying out the tests which combine super high pressure with high temperature, it is expedient to apply gas as a pressure transmitting medium.

Along with the intensifier mentioned above /8/, two types of apparatus, permitting the creation of super high gas pressure in conjunction with high temperature, were developed at the Institute of Crystallography.

In the first of these, carbon dioxide was the pressure transmitting medium. It was used for the study of the pressure and temperature dependence of the polymorphic transformation of black phosphor /9/. In the second apparatus, nitrogen or argon were used as pressure transmitting media. The conical high pressure vessel in this apparatus is fed by a device which permits the feeding of gas at pressures up to 2,000 kg/cm², and then cuts off the contents of the cone's canal from the gas supply system.

The pressure dependence of the melting point of aluminum and copper was determined in the apparatus of the second type. Pure aluminum (99.99 percent) and copper (99.995 percent) were used in the investigations.

A schematic diagram of the intensifier canal is shown in Figure 1.

The aluminum sample was fused in a graphite crucible (1) 10 millimeters in diameter and 14 millimeters high, placed within a heating element (2) of nichrome wire. To prevent the graphite from closing the turns of the nichrome coil, the crucible was separated from the latter by a thin layer of mica. The melting point of aluminum under pressure was measured with the aid of a differential platinum-platinrhodium thermocouple (3). To prevent the "hot" junction of the thermocouple from diffusing in the fused aluminum, the

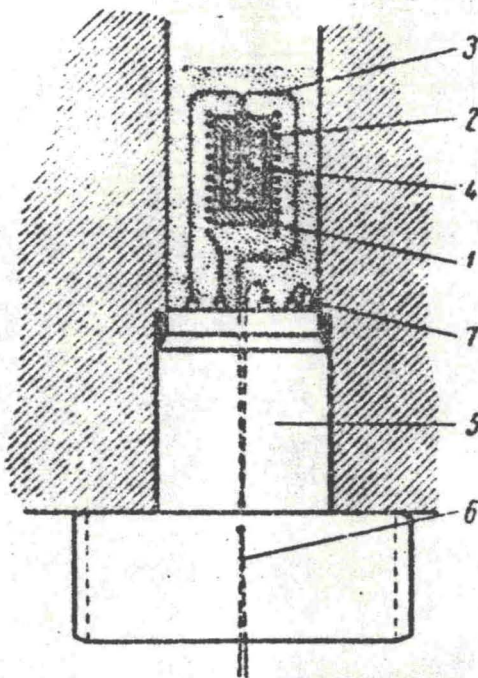


Fig. 1. Schematic cross-section of the measuring device inside the canal of a super high-pressure vessel.

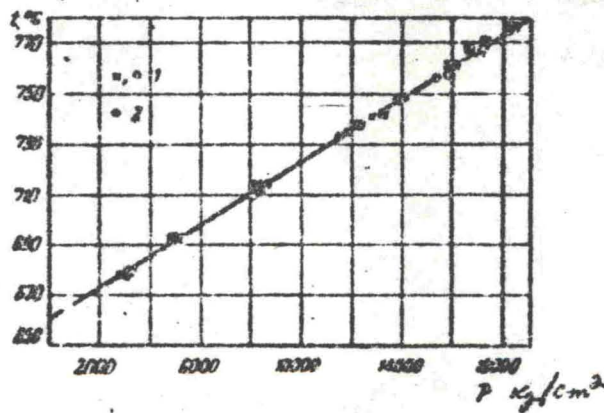


Fig. 2. Pressure dependence of the melting point of aluminum:

1. in an argon atmosphere,
2. in a nitrogen atmosphere.

former was placed in a thin quartz jacket (4) fastened to the graphite crucible cover. The "cold" junction of the thermocouple was deeply inserted into the electric lead-in (5).

Its temperature was measured with the second thermocouple (6) introduced into the body of the electric lead-in from outside, whereby the distance from the junction of this thermocouple to the "cold" junction of thermocouple (3) did not exceed six millimeters. To heat-insulate the furnace from the walls of the cone's canal, all the free space in the intensifier canal, with the exception of the upper part where the piston was moving, was filled with a fine aluminum oxide powder.

The electro motive force of both thermocouples was measured with two potentiometers of the PP-1 type. The pressure in the intensifier canal was measured by a manganin manometer coil mounted on the electric lead-in (7). The resistance of the manganin coil was measured by a type MVL-47 resistance bridge. The measurements of pressure and temperature were carried out within an accuracy of ± 100 kg/cm² and $\pm 2^{\circ}\text{C}$ respectively.

The experiments on the melting of aluminum under pressure were carried out in nitrogen and argon media (in view of the possibility of a reaction between the aluminum and nitrogen with the generation of nitride). The measurements are presented in Figure 2. From Figure 2, it is evident that the melting point of aluminum rises linearly with the pressure in the argon and nitrogen media.

To obtain a higher temperature at the determination of the pressure dependence of the melting point of copper, the nichrome coil was replaced by one made of tungsten, and the quartz jacket was replaced by a steel "pocket" with a four-millimeter thick wall to protect the "hot" junction of the thermocouple. The over-all scheme and method of computation remained unchanged.

In these tests, nitrogen served as a pressure transmitting medium. The results of the measurements are presented in Figure 3. The accuracy of measurements of temperature in the $1,050 - 1,250^{\circ}$ range is estimated at $\pm 5^{\circ}\text{C}$. Within the limits of experimental error the melting point of aluminum and copper rise linearly with the pressure. For aluminum the value of dT/dP amounts to 6.3×10^{-3} degrees \times cm²/kg, and for copper it amounts to 4.6×10^{-3} degrees \times cm²/kg.

Consideration of Results

F. E. Simon and his collaborators /10/ suggested the following equation to express the pressure dependence of the melting point of substances:

$$\frac{P}{\alpha} = \left(\frac{T}{T_0}\right)^c - 1, \quad (1)$$

where P -- pressure, T and T_0 -- melting point in $^{\circ}\text{K}$ at P pressure and atmospheric pressure respectively, α and c -- constants. For nonpolarized substances, the value α proved to approach in magnitude the so-called internal pressure, determined by the evaporation energy of a substance ()

$$\alpha = \frac{\lambda - RT}{V}, \quad (2)$$

where V -- volume of the liquid. The value c for these substances was between 1 and 2. Thus, for argon $c = 1.16$ /11/, for helium 1.554 /12/ and for nitrogen - 1.775 /11/. At $c = 1$, the melting point should rise linearly with the pressure. With the rise of c the melting curves T - P decline from the straight line towards the pressure axis. Simon /13/ made an attempt to apply equation (1) to the melting of alkali metals, by utilizing the data obtained by Bridgeman /1/. It appeared that for them the value c amounts to 3.8 to 4.8. In the meantime, all the attempts to calculate the value c on the basis of various equations of the state of liquids and solids (see /14-16/) ended up with $1 < c < 1.5$. Apparently, in the case of the fusing of metals, Simon's equation must be considered to be empirical.

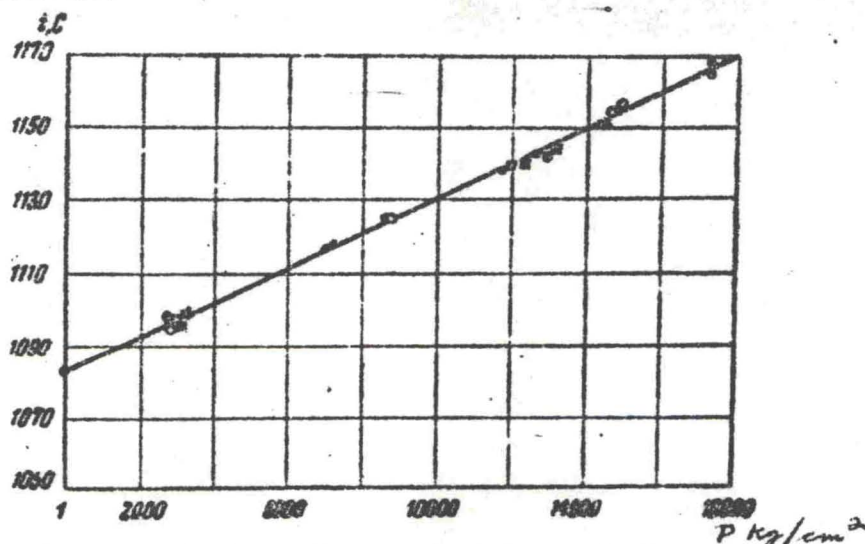


Fig. 3. Pressure dependence of the melting point of copper. Points -- results of separate experiments.

Calculations show that in the case of fusing of lead and tin /4/ the values of c amount to approximately 2.5 and 4.0 respectively. It should be noted, however, that the

values α and c are very susceptible to extremely slight variations of the melting curve; therefore, their determination by the melting curves is performed with very low accuracy.

The melting curve of thallium is also bent towards the axis of pressures /5/. In the case of copper and aluminum, and cadmium and zinc as well /5/, the pressure dependence of the melting point can be expressed --within the limits of experimental error -- by a straight line.

It is to be hoped that the further accumulation of experimental data on the melting of metals under pressure will provide a theoretical explanation for the observed laws, and will link them with the characteristics of metal constitution.

Conclusions

1. The melting points of aluminum and copper have been determined in an inert gas atmosphere at pressures up to 18,000 kg/cm². It was found that within the limits of experimental error the melting points of these metals rise linearly with the pressure.
2. The applicability of Simon's equation to the melting points of metals at high pressures has been discussed.

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