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STUDY OF LIQUID VISCOSITY BY MEANS OF VIBRATION METHOD

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Foreign Technology Division
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by

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A. N. Solov'ev

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13. ABSTRACT

The viscosity of Ga during supercooling was measured. Ga was melted, after which the temp. was quickly lowered to a given value. It is then held const. for 2-3 hrs. as the viscosity was measured. Ga was subsequently heated to $95\text{-}8^{\circ}$ and again the viscosity was detd. Quick raising and lowering of temp. guarantees preservation of solid phase residues at temps. greater than the m.p. (29.8°) and do not allow the emergence of the solid phase during cooling. The viscosity of Ga at $0\text{-}100^{\circ}$ following an empirical equation in a range of $0\text{-}100^{\circ}$ is obtained: $\eta = 0.658 \exp 559.4/I$. The mean sq. error of estn. measurement is 1.4%. Results agree with x-ray data. The viscosity of Ga during supercooling changes in the same way as with a temp. greater than the m.p.-structurel changes are absent. Branching due to the temp. of viscosity dependence is not noted. Causes of disparity with other exptl. data are not detd. Viscosity of antifreeze-65 is measured between 15.5 and 72°. The exptl. value of viscosity decreases approx. exponentially with increase in temp. from 11.3 to 2.0 cp.

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О о	*О о*	O, o	Ю ю	*Ю ю*	Yu, yu
П п	*П п*	P, p	Я я	*Я я*	Ya, ya

* ye initially, after vowels, and after ъ, ь; e elsewhere. When written as ё in Russian, transliterate as yё or ё. The use of diacritical marks is preferred, but such marks may be omitted when expediency dictates.

STUDY OF LIQUID VISCOSITY BY MEANS OF VIBRATION METHOD

V. N. Genrikh, A. B. Kaplun, and
A. N. Solov'ev

Introduction. Viscosity is one of the most important characteristics of liquids. It is closely connected with the structure and nature of the thermal motion of molecules of the liquid and, therefore, by studying it we can obtain significant information on the nature of the liquid state. On the other hand, many processes in chemical, metallurgic, and other areas of industry occur in a liquid phase, and viscosity can be used as a technological characteristic of the process.

In view of this, a number of works devoted to the experimental study of viscosities of liquids, improvement of the old and development of the new measurement methods is constantly increasing. The improvement in the viscosity measurement methods is toward increasing the temperature interval, increase in accuracy, decrease in the measurement time and automation of the measurement process.

The most promising method for measuring viscosity is the vibration method. The theory of the method and description of certain variants of the viscometer are given in our works [1, 2, 3, 4, 5]. The questions concerning a further improvement of this method, new constructional solutions and investigation results of certain liquids is the aim of this work. In order to establish a coherency of this composition, we will quickly review the bases of this theory and the general system of the viscometer.

Bases of the Theory. Let a thin plate with surface S, mounted on a long bar and elastic extensions with a total mass m_0 (Fig. 1), be undergoing laminar harmonic vibrations under the action of force $F_0 \sin \omega t$, while submerged in a limitless liquid with viscosity η and density ρ (examined is a purely newtonian liquid). In addition to the harmonic excitation force, the system is affected by the inertia $F_u = (m_0 + m_\eta)\ddot{x}$, where x - a shift from the position of equilibrium at any instant; $m_\eta = 2\sqrt{2}\, S\sqrt{\frac{\rho\eta}{\omega}}$ - the adjoint liquid mass. Value $m_\eta \ddot{x}$ characterizes the contribution to the inertia due to the part of liquid set in motion near the plate.

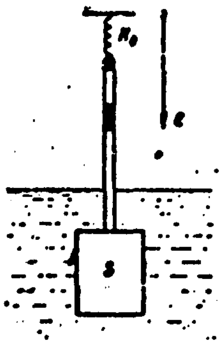

Fig. 1.

Resistance of the elastic element with rigidity k_0

$$F_c = k_0 x.$$

Friction force of the liquid against the lateral surface of the plate

$$F_{\eta} = 2S\sqrt{\frac{\rho\eta\omega}{2}}\,\dot{x}$$

Friction force of gas against the lateral surface of the bar and extensions

$$F_{\eta \text{б}} = 2S_1\sqrt{\frac{\rho_1\eta_1\omega}{2}}\,\dot{x}$$

S_1 - lateral surface of parts of the suspended system which are in friction with gas, ρ_1 - gas density, and η_1 - gas viscosity. Friction force in elastic element

$$F_{y} = k_1\dot{x}$$

Force of head resistance

$$F_{\text{л}} = k_2\dot{x}$$

Surface tension force applied to the bar at a place where it comes out of the liquid

$$F_{c} = c\sigma x$$

where σ - surface tension coefficient, c, k_1, k_2 - constants.

The equation of motion of the suspended system will be written as

$$\ddot{x} + a_1\dot{x} + a_2 x = a_3 \operatorname{Sin}\omega t \qquad (1)$$

The complete solution of a nonuniform linear equation with constant coefficients of the 2nd order (1) has the form

$$x = A_0 e^{-\frac{a_1}{2}t} \mathrm{Sin}(\omega_0 t + \varphi) + \frac{a_3}{\sqrt{(\omega^2 - a_2)^2 - \omega^2 a_1^2}} \mathrm{Sin}\left[\omega t - \mathrm{arctg}\frac{\omega a_1}{\omega^2 - a_2}\right] \quad (2)$$

where A_0 - initial amplitude, φ - initial phase of natural vibrations of the suspended system, which are determined from the original conditions; in particular, they can equal 0 or can have any arbitrary value reasonable for experiment conditions, and ω_0 - natural frequency of free vibrations of the suspended system.

In the stabilized regime of vibrations, with an increase in t the first term in (2) tends rapidly toward zero, and several seconds after the beginning of vibrations the motion will be described basically by the second term only.

In our designations

$$a_1 = \frac{2S\sqrt{\frac{\rho\eta\omega}{2}} + 2S_1\sqrt{\frac{\rho\eta_1\omega}{2}} + k_1 + k_2}{m_0\left(1 + \frac{2S}{m_0}\sqrt{\frac{2\eta\rho}{\omega}}\right.}$$

$$a_2 = \frac{k_0\left(1 + \frac{c\sigma}{m_0}\right)}{m_0\left(1 + \frac{2S}{m_0}\sqrt{\frac{2\eta\rho}{\omega}}\right.}$$

$$a_3 = \frac{F_0}{m_0\left(1 + \frac{2S}{m_0}\sqrt{\frac{2\eta\rho}{\omega}}\right)}$$

As seen from solution (2), the suspended system undergoes harmonic vibrations at frequency ω, equal to the vibration frequency of the excitation force with amplitude

$$A = \frac{a_3}{\sqrt{(\omega^2 - a_2)^2 + \omega^2 a_1^2}}$$

and with a shift in phases relative to the excitation force phase

$$\varphi = \operatorname{arc\,tg} \frac{\omega a_1}{\omega^2 - a_2}$$

A detailed analysis of the possible measurement methods is given in our work [4]. Phase-frequency version turned out to be the most suitable for high viscosities. In this case, the frequency is selected such that phase shift φ is equal to 90°. And then the calculation formula will assume the form of

$$\eta \rho = \left(\frac{m_0^2 \, \omega_0}{8 S^2} \right) \cdot \frac{(\lambda^2 - 1)^2}{\lambda}$$

where $\lambda = \frac{\omega_0}{\omega}$ – dimensionless frequency.

In order to determine the viscosity the vibration frequency of the system submerged and not submerged in liquid (ω_0, ω) should be measured when the phase shift is equal $\pi/2$. The phase-frequency version is suitable only for the relatively high viscosities, since, in essence, the change in frequency is connected with a change in the "adjoint mass" of liquid.

In the case of low viscosities, the most suitable is the resonance-amplitude version. In this case the frequency is selected such that the amplitude is maximum. A working formula can be derived from the condition of amplitude maximum. For low viscosities, when $\lambda \approx 1$ and phase shift $\varphi = \pi/2$, the working formula is simplified considerably and assumes the form

$$\sqrt{\rho \eta} = \frac{C_1}{A} + C_2 \qquad (6)$$

Thus, in order to determine the viscosity it is sufficient to determine only the amplitude of vibrations. The method is more suitable for a relative version, i.e., when constants C_1 and C_2 are determined during the calibration tests on a liquid with known density and viscosity values. The resonance-amplitude version has one more significant advantage: it is very easy to build an automatic instrument for a continuous measurement of viscosity. Actually, if the vibration system is employed as a unique "tonometer", i.e., as a frequency stabilizer in the drive-pulse generator system, then the generator frequency will always be automatically stabilized equal to the resonance frequency of the system, and in order to determine viscosity it is only necessary to measure the amplitude of vibrations. A version based on the attenuation of free vibrations of the suspended system is also possible for the measurement of low viscosities. If the forced vibrations in the system are excited at an amplitude of A_0, and then the excitation is eliminated, then, independently of the frequency of free vibrations ω_0, depending only on the mechanical properties of the suspended system.

If we assume that $F_0 Sin\,\omega t = 0$, then the second term in formula (2) will equal zero, and the attenuating vibrations will be described by formula

$$x = A_0 e^{-\frac{a_1}{2}t} Sin(\omega_1 t + \varphi) \tag{7}$$

The viscosity enters coefficient a_1, which for low viscosities, i.e., when $m_\eta \ll m_0$, has the form

$$a_1 = \frac{S\sqrt{\omega_0}}{m_0\sqrt{2}} \cdot \sqrt{\rho\eta} + \frac{S}{m_0}\sqrt{\frac{\rho_1 \eta_1 \omega_1}{2}} + \frac{k_1}{2m_0} + \frac{k_2}{m_0}.$$

From this it follows that in order to determine the viscosity, we have to know the attenuation decrement. In essence

$$\delta = -\frac{1}{n} \ln \frac{A_n}{A_0} \tag{8}$$

here n is a number of periods,

$$\sqrt{\rho\eta} = C_1'\delta - C_2' \tag{9}$$

Under experiment conditions, coefficients C_I' and C_2' can be easily made constant and it is convenient to use the relative version of the method. Since in reality the decrement is very large, then the measurement time is only a few seconds. Therefore, in order to determine the decrement one can use either the recording on the loop oscillograph or determine the attenuation time between the two fixed amplitude values.

Pick-up and Measuring System. Since all mechanical vibration parameters of the suspended system depend on the properties of the examined medium, then in order to realize any version of the method it is necessary to convert the mechanical vibrations into electric which are considerably easier to measure. In this case, the appropriate converter (pick-up) must meet very rigid requirements, owing to the complex operating conditions. The basic difficulties are in the following:

1. The amplitude of mechanical vibrations is small (on the order of $3 \div 30 \mu\kappa$) and can change several times during the measurement.

2. The vibration amplitude pick-up must operate in a low-frequency field of the excitation coil.

3. The mechanical vibration frequency of the suspended system can change several times.

4. The space occupied by the pick-up must be minimum.

5. The pick-up temperature can vary over a considerable range.

6. The pick-up must have a minimum inverse effect on the vibration system i.e., it must be contactless.

Of the large number of vibration-displacement pick-ups, the most suitable for our purposes turned out to be the high-frequency inductance pick-up with frequency modulation. The structure of the viscometer pick-up is illustrated in Fig. 2.

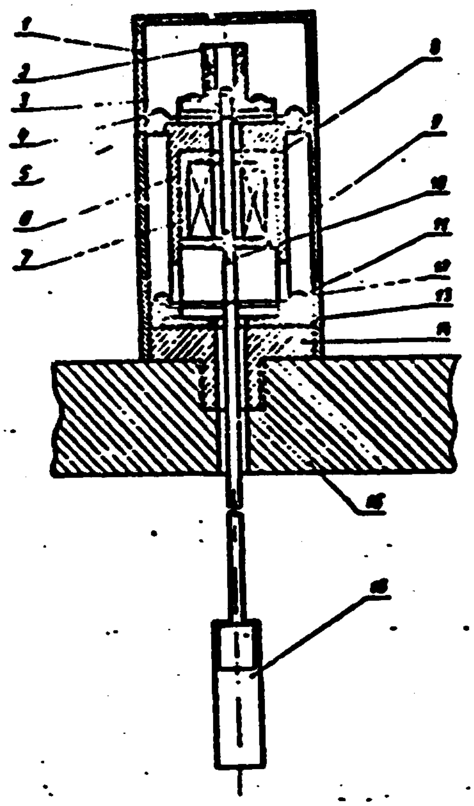

Fig. 2.

Plate probe (16) welded to a thin-walled tube made from stainless steel or other material is submerged into the test liquid up to a certain level. The tube is suspended on wire extensions (5, 12) made from string steel, which are fastened with screws (11, 4) to housing (6). A permanent magnet (10) made from alloy "alnico" is embedded inside the tube. The upper free end of the tube enters the high-frequency inductance coil (2) up to approximately 1/3 of its length. The vibration of the core results in frequency modulation of the high-frequency oscillation (in our case, 8 MHz) of the generator. As is known, the inductance of the coil with a core depends on the magnetic permeability of the core, its shape and position relative to the coil. For the cylindrical coil and magnet a linear segment can be easily separated from this relationship. If the inductance coil is connected to the generator circuit, then the generator frequency will vary according to the law of motion of the core. In the region of high frequencies the core can also be made from a nonmagnetic material, particularly from stainless steel.

The principal electric circuit of the viscometer and the operation of the high-frequency pick-up are described in [2, 3]. Let us examine the block-diagram of measurements, which was used in the amplitude version of the method (Fig. 3). The frequency-modulated signal from generator (1) enters frequency detector (2). The frequency detector converts the change in generator frequency into a change in voltage so that the signal at the frequency detector output is proportional to the amplitude of mechanical vibrations of the core rigidly connected to the suspended system. Further, this signal proceeds to voltage amplifier (3). Then the signal is limited with respect to amplitude by means of tube limiter (4) and is fed to the excitation coil of the operating or control vibrator [O. V. and C. V.] (P. B. and K. B.) through power amplifier (5); the excitation coil field interacting with the permanent magnet of the suspended system induces vibrations in the probe. Thus, the whole system represents an electronic mechanical generator with positive feedback. Due to the limiter, the excitation coil always receives a signal of definite magnitude and, in the case of low viscosities, the frequency of the self-excited oscillator is virtually equal to the resonance frequency of mechanical vibrations of the suspended system. The system has the capability to measure vibration amplitude of both alternating and direct currents. In the first case, a tube voltmeter (B3-7) is connected to the frequency detector output; in the second case, the signal from the frequency detector is fed to a high-ohm dc potentiometer (P-307), or the automatic potentiometer [EPP-09] (ЭПП-09), or other recording instrument through rectifier (6).

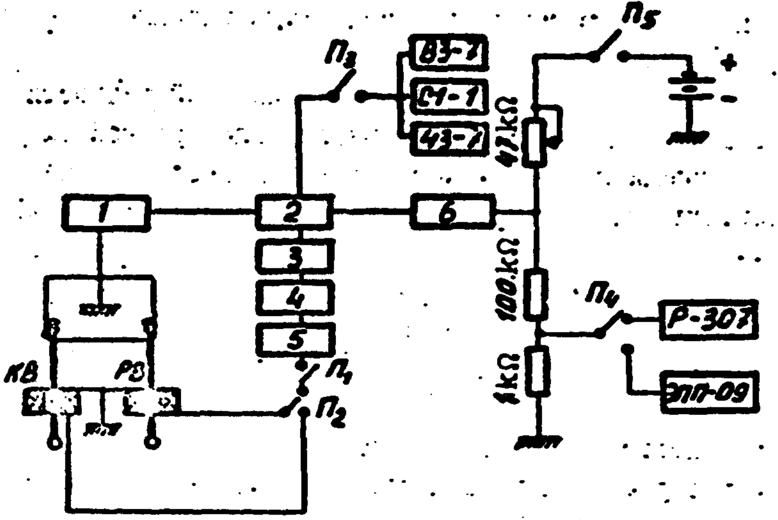

Fig. 3.

There is a number of ways for increasing the measurement accuracy by using electronic circuits. The most known are the selection of the highly stable tubes and radio components, introduction of thermocompensation, operation at lower filament and anode voltages, introduction of negative feedbacks, etc. However, as a rule these methods are rather complicated and do not always lead to desirable results. In particular, the industrial vibrometers have a vibration amplitude measurement error on the order of 5-10%.

The measurement system is constructed such as to achieve maximum measurement accuracy by the simplest means. The stability of the electronic system is checked by means of the control viscometer. The high-quality coils of the operating viscometer (O. V.) and control viscometer (C. V.) are connected in parallel. This system has the capability to switch off the vibrations of both suspended systems in order to measure the background. Both viscometers have an identical structure. The probe of the control viscometer is placed into the ethylene glycol, the temperature of which is maintained constant with an accuracy of up to 0.1°C. Since, in this case, all changes in the control signal are caused only by the instability of operation in the system, then by changing the amplification coefficient it is possible to achieve constancy in the signal of the control viscometer. The control can be accomplished by any potentiometer in the low-frequency amplifier circuit, since the whole system and the elastic element operate on the linear segments of the characteristics. In measuring the amplitude via potentiometer P-307, the control signal is established with an accuracy of up to ±0.2%. With such an electronic system control method, there is no need for the highly stable power sources (anode batteries and filament accumulators), and this enables us to use a standard power unit BC-12. The signal from frequency detector (2) can be given to the control terminals through switch Π_3. Here we can connect one of the following: voltmeter for rough amplitude measurement (we used instrument of the type B3-7, class 1.5), oscillograph for visual observation of the form of vibrations, frequency meter to control the vibration frequency, or other control and measurement instruments. The automatic potentiometer EPP-09 is switched-on by the switch $П_4$; a compensation system switched-on by switch $П_5$ is provided for expanding the instrument scale.

Studies were carried out on the stability and accuracy of the vibrational viscometer with a control vibrator. These studies have shown that viscosity measurement accuracy in the area of room temperatures, using the amplitude method, can be up to ±1% [4].

Naturally, the measurement systems of the phase-frequency method and the method of attenuating vibrations are different from those compiled above. Nevertheless, the structure of the viscometer pick-up unit and the principle circuit of the vibration amplitude pick-up is the same in all versions. This enables us to realize all three versions of the method on one unit by means of simple external switchings, depending on the viscosity range. The experimental verification of the amplitude and phase-frequency versions was described earlier in [4].

Let us examine the block-diagram of the non-stationary method for measuring liquid viscosity (method of attenuating oscillations) (Fig. 4). In the test, according to formula (9), one has to measure the time in which the amplitude of free vibrations decreases from A_0 to A_1. To measure time we used the electronic readout timer consisting of the generator Г3-3 and conversion unit [PST-100] (ПСТ-100). Vibrations of the system were produced by the usual method. The initial amplitude A_0 was established. Then, by means of the relay [RES-9] (РЭС-9) the field circuit was shut-off and the scaling circuit was turned on simultaneously. Upon reaching the predetermined amplitude A_1, the scaling circuit

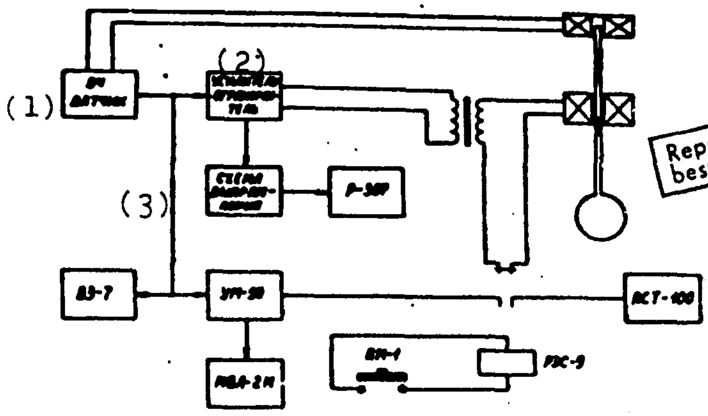

Fig. 4.
KEY: (1) B4 pick-up; (2) amplifier limiter; (3) rectifier circuit.

was shut-off and the excitation coil was turned on. The attenuation time was determined by the known generator frequency (ν= 1000 Hz) and the readings of the PST-100. By repeating all operations the readings on counter PST-100 were accumulated up to 10 measurements. The fixing of A_0 and A_1 was accomplished visually by means of voltmeter [VZ-7] (B3-7).

In another version of the system the initial amplitude was measured by the P-307, the variable signal with free attenuation was fed to the PST-100, and the discontinuation of pulse counting was ensured when the actuation level was reached in the PST-100, and this was taken as A_1.

The experimental points in toluene, acetone, and air are plotted in Fig. 5. $\sqrt{\rho\eta}$· and $\frac{1}{N}$ are plotted on the coordinate axes, where N - number of complete natural vibrations of the suspended system between the A_0 and A_1 levels. As seen from the figure, all points are in accordance with the theory and are satisfactorily arranged with respect to the calibration curve.

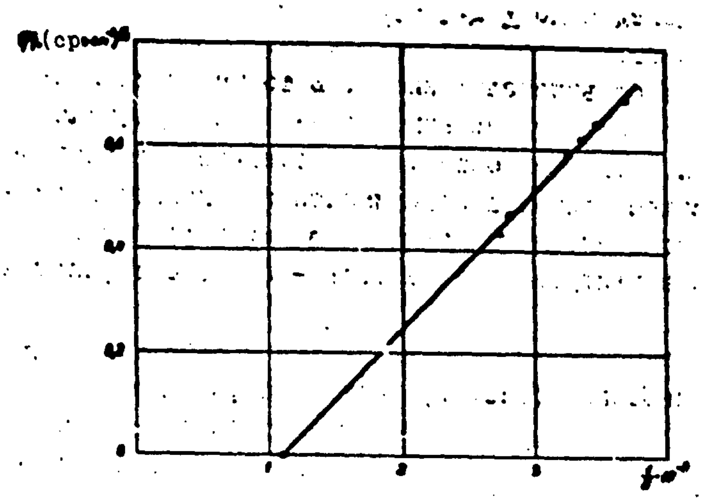

Fig. 5.

The vibration method for measuring viscosity has a number of peculiarities which must be considered in order to obtain satisfactory experimental results. Below are examined certain operational features of the vibration viscometer revealed during a prolonged operation of the high-temperature unit used for measuring viscosities of metals.

Assembly Requirements of the Electrical Part of the Viscometer.

In view of the fact that high-frequency inductive pick-up is used as the vibration amplitude pick-up, the mounting of the electronic unit must be rigid. This is due to the fact that any change in the position of connecting wires or the pick-up components will entail a change in the circuit parameters of the high-frequency generator or decoder. In order to avoid the microphone effect and to exclude the influence of accidental shocks and jarring, the unit must be placed on soft rubber. Due to the very fine resonance tuning of the generator and detector circuits, we were unable to reproduce the readings by switching the high-frequency measurement coils of the operating and control viscometers. In view of this fact, the measurement coils of the operating and control viscometers are connected in parallel and are in constant contact with the generator. The monitoring of the system is accomplished by the alternative excitation of natural vibrations in the control and operating viscometers by switching the excitation coils. Even though the parallel connection of the measurement coils cuts the sensitivity of the vibration amplitude pick-up in half, nevertheless, it relieves the experimenter from a tedious operation of resonance tuning of the generator and detector with each switching from the control to the operation, and it is also conducive to a more stable operation of the system.

The measurement coils of the operating and control viscometers should be connected to the electronic unit by coaxial cable only. The cable should be rigidly fastened in order to prevent its accidental movements during the viscometer operation. Under actual conditions the experimental installation can be a considerable distance from the measuring system. Cables 8-10 m in length can be used in the system described above. The cable length can be increased considerably if a high-frequency matching transformer is used. The lead-in of the high-frequency cable into the working space (under the vacuum cover) of the viscometer is accomplished by means of a separate vacuum seal; furthermore, both the core of the cable as well as its braiding ("ground") are connected to the measurement coil by rigid well-secured wires, not necessarily by a cable. Under no circumstance should any other wires be permitted to pass through the same connector with a high-frequency wire, for example, wires of thermocouples, excitation coil, etc. It is also not permissible that the high-frequency wires be close, much less intertwine, with other wires. This is due to the fact that the effect of the adjoined wires on the generator circuit, for example, on its quality rating depends on the position of not only that part of them which is close to the high-frequency coil and lead-in wires but also on the position and connection of wires outside the cover.

Structural Features of Viscometer.

Due to resonance forced mechanical vibrations can arise in certain installation parts under the influence of probe vibrations. Since additional energy is spent on the excitation of idle vibrations and the power of the excitation system is constant, then this leads to a decrease in the vibration amplitude of the probe. The effect of idle vibrations is especially noticeable when measuring low viscosities (less than one centipoise), when the friction force acting on the probe on the part of a liquid is small. Therefore, the unit must be constructed so as to ensure a minimum magnitude and univocacy of the energy losses. It is necessary to ensure that all parts of the installation were rigidly attached and did not move with respect to one another during the measurement. The support frame of the installation and the assembly plate must be sufficiently massive; it is necessary to attempt to lower the center of gravity of the installation.

In order to decrease the effect of accidental shocks, the frame is mounted on soft rubber or felt; preevacuation pumps which are in close proximity to the installation should also be mounted on felt. If these conditions are observed, no special foundation or mounting of the viscometer on a main wall is necessary in order for the viscometer to operate. Strips of vacuum rubber (4) were used to decrease the contact between the pick-up casing (1) and support plate (5); the mounting diagram is illustrated in Fig. 6. It is necessary to emphasize that the fastening screws (2) can loosen during the vibration and, therefore, they should be secured using, for example, slot washers (3).

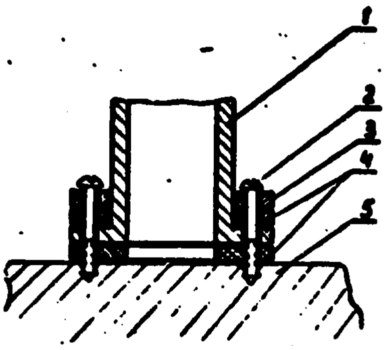

Fig. 6.

Viscometer Calibration and Measurement.

The calibration of the instrument is usually done with air or organic liquids with known viscosity and density. During this the following should be considered. As a rule, thermostatic control is provided to avoid overheating of the extensions; sometimes the cooling of the installation is accomplished with tap water. In the summer when the temperature and humidity are high, water vapors from the air can form condensation on the internal parts of the installation, including the probe. Therefore, the thermostatic control temperature must be above the dew point. Water vapor condensation can also occur with a decreased calibration liquid temperature.

On the other hand, vapor condensation of the calibration liquid is also possible when its temperature is increased. We were unable to raise the temperature of such volatile liquids as acetone and toluene to above 40-50°C, and water - 70°C; a further increase in temperature is accompanied by the condensation of the calibration liquid on the probe. In order to establish a new temperature regime for the calibration liquid, a considerable amount of time is required. Therefore, in order to speed up the calibration we used a container in which four glasses are soldered in one jacket. The glasses are filled with four calibration liquids; the temperature in all four glasses is identical.

The calibration is accomplished by changing the calibration liquid in which the probe is submerged, at a constant temperature. This method has enabled us to carry out the calibration in several tens of minutes. The measurements themselves do not have any significant features. A two to three-hour heating is necessary for normal operation of the system; the thermostats of the control and operational viscometers are turned on simultaneously. The measurement is done in the following order. At first the temperature of the control viscometer and the plate of the operating viscometer is checked; then a determined control signal value is set. If the viscosity of the test liquid is low, then the operating signal does not reach the maximum value at once; the time before the maximum is reached ranges from several seconds up to a minute. In order to decrease the chance of error, the measurement is accomplished several times. It is not necessary to check the system any sooner than every 5-10 min.

Investigation of Viscosity of Several Liquids.

The studies examined in this work were done on a high-frequency installation (Fig. 7). In this installation the pick-up unit (3) is set in the area of room temperatures. It is mounted on a massive support plate (7) and is removed from the working space (13) at 450 mm. The installation also has a system of refrigerators and shields (9), which provides normal operating conditions for the electrical part of the viscometer. In this installation, we can create a vacuum on the order of 10^{-5} mm Hg or pressure up to 50 atm. The main part of the installation remains invariable and only the filling system and certain auxiliary units were changed.

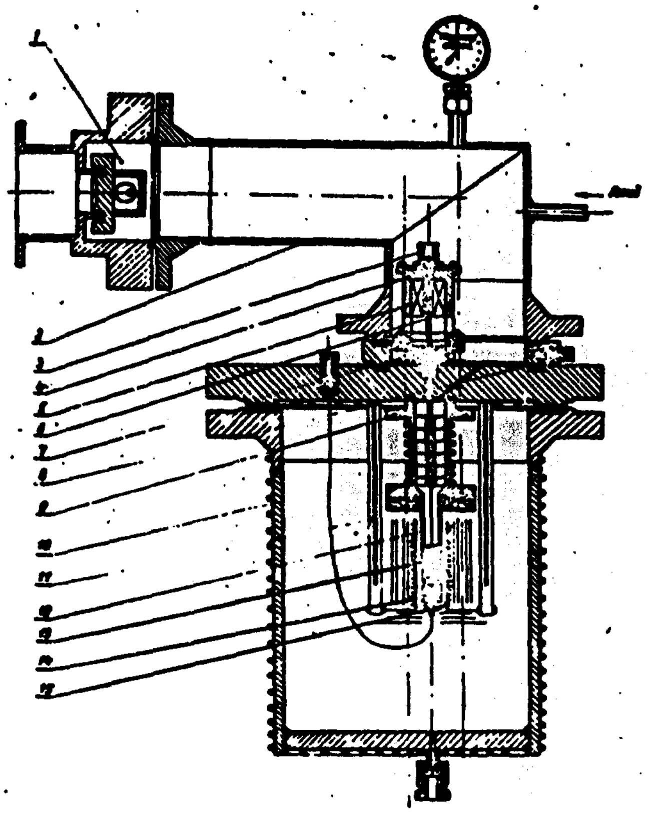

Fig. 7. 1 - valve; 2 - tube with top cover; 3 - high-frequency coil; 4 - pick-up housing; 5 - excitation coil; 6 - permanent magnet; 7 - bar; 8 - slip ring with electrical leads; 9 - condenser; 10 - lower cover; 11 - electrical lead; 12 - crucible; 13 - probe; 14 - introduction of liquid metal; 15 - heater.

Viscosity Measurement of Gallium in the Region of Supercooling. The question concerning structural reconstructions occuring close to the melting point has been discussed in the literature for quite some time now. In the opinion of some authors [6], there should be remnants of solid phase in the liquid phase immediately after melting; it would appear as though the "subcrystals" were floating. A breakdown of these "subcrystals" can occur in the time comparable to that of the experiment. The "precrystallization" processes during cooling and subsequent hardening of the liquid correspond to the "post-melting" processes connected with inheritance of the solid phase during liquid heating. These occurrences should be particularly noticeable in metals having a tendency toward supercooling. Since viscosity is a structurally sensitive property, then the atomic rearrangement must elicit a change in viscosity in time. If the time of the experiment is less than the time of the assumed rearrangements, then the viscosity change in time can be detected experimentally. The study of viscosity in the region of supercooling can yield definite information on the nature of thermal movement. The region of supercooling has a metastable nature, and it can turn out that the temperature course of viscosity in this region is different from that above the melting point. In studying these processes, it is of interest to use a continuous method for measuring viscosity, such as the vibration method. Earlier, T. L. Gotgil'f [6] has observed a viscosity change in time on a number of objects, including gallium. G. I. Goryaga [7] did not observe such change in gallium. Both studies were done using the method of torsional vibrations of a cup. Only G. I. Goryaga has studied gallium viscosity below the melting point. Furthermore, data on gallium viscosity are very contradictory. For example, Goryaga's data are 65% lower than those in [8]. In view of this fact, we have attempted to measure the viscosity of gallium in the 0-100°C temperature range. Gallium is a very suitable study object: it has a low melting point 29.8°C, high tendency toward supercooling, and low vapor pressure. Moreover, at the present time gallium and its components find ever wider use, especially in the semiconductor technology.

The viscosity of gallium was studied on a high-temperature installation. A glass cup with double walls, between which water was pumped from thermostat U-8, was in an air-tight connection with a refrigerator flange (Fig. 8). Prior to the filling a vacuum on the order of 10^{-5} mm Hg, which was maintained throughout the measurement, was created in the space formed by the upper cover, refrigerator housing, and cup. The filling was accomplished through a kovar tube connected to the auxiliary container with liquid gallium. First, the stainless steel container is evacuated to 10^{-2} mm Hg, and then a low residual pressure of an inert gas, necessary in order to establish a certain level of gallium in the working space, is created in it by means of the system of valves. The change in the gallium level during measurement was accomplished in the same way. To remove mechanical impurities from gallium, a stainless steel filter is embedded into the bottom of the auxiliary container. The temperature was measured by a platinum resistance thermometer. The measurements were carried out in the following order. The gallium (class Kh-4 containing 99.9% of the basic substance) was melted; after which the temperature was rapidly raised to a certain level and then maintained constant for 2-3 hours. The vibration amplitude of the probe was measured continuously throughout this time. After this, the gallium was heated to 95-98°C and the measurements were taken again for 2-3 hours. A rapid rise and fall of the temperature should have preserved the remnants of the solid phase during heating and prevent the appearance of the solid phase during cooling. The temporary holding at maximum temperature should have resulted in a stabilized state of liquid gallium. We have observed that at a constant gallium temperature the vibration amplitude of the probe during the 2-3 hours decreases by approximately 2 times and then remains constant. However, if the gallium level is rapidly lowered (2-3 mm) and then raised to the original value the vibration amplitude becomes maximum and assumes the initial value. Evidently, the decrease in vibration amplitude is due to the effect of the gallium surface film. It could be actually

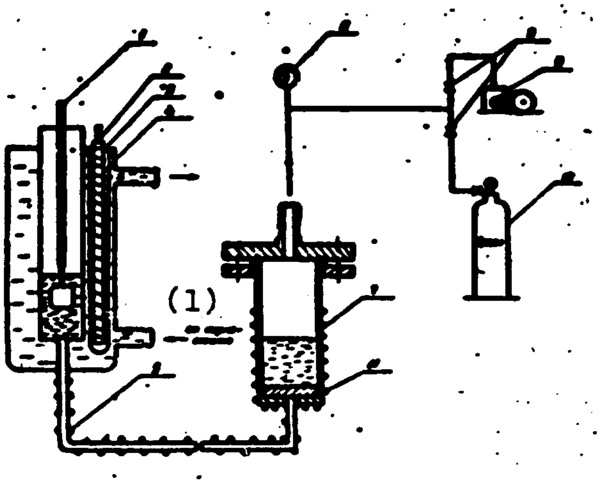

Fig. 8.
KEY: (1) Illegible.

observed that the surface film appeared as though it was structurized. During lowering it could be observed that the surface film of the gallium appeared as though it were being "hung-up" on the probe. The vibration amplitude increase to the maximum occurred sometimes as a result of accidental shocks. In view of the above mentioned fact, the viscosity factor was calculated using the maximum vibration amplitude values. The maximum amplitude values at a given temperature were reproduced within the error limits of the experiment independently of how this temperature was reached. In all, four series of experiments were carried out in which 33 points were plotted. Each point represents an average of 5-7 measurements. The experiment results and the data of other authors are given in Fig. 9. Table No. 1 shows the experimental viscosity values of gallium. The experimental values of all four series of experiments fit on one curve within the error of measurement (1-1.5%). Let us note that one series of experiments was obtained under conditions of continuous cooling of the metal without the intermittent holdings. Since the investigations were carried out over a narrow temperature range, the processing of the measurement results was accomplished by a simple exponential formula. In the temperature range of 0-100°C, the experimental values are described by the equation

$$\eta = 0{,}658 \exp \frac{5594}{T}$$

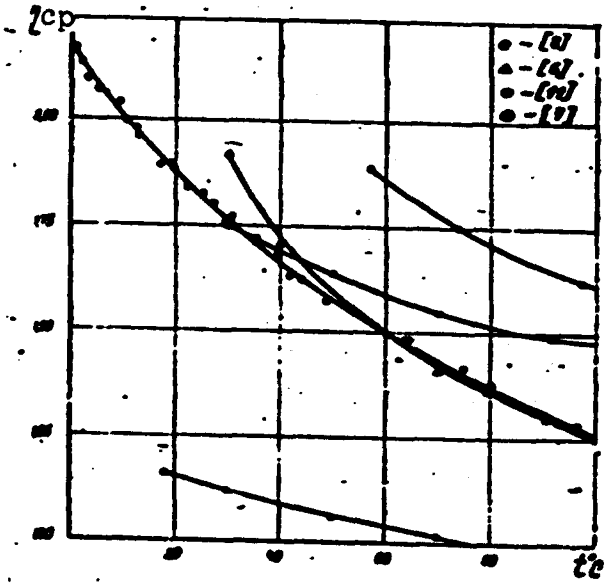

Fig. 9.

The mean square measurement error is 1.4%. Thus, in the supercooling region the viscosity of gallium changes according to the same law that is applicable above the melting point. This indicates the absence of structural changes in this region which is in accordance with the radiographic data in [9], and also, that the nature of thermal motion in the region of supercooling remains invariable. The same result was obtained by G. I. Goryagà [7] who has studied gallium viscosity above 18°C.

In our experiments, in contrast to work in [6], the branching of temperature dependence of viscosity was not observed. The apparent increase in viscosity detected under all conditions at constant temperature is actually connected with the effect of

Table No. 1.

No.	t, °C	η, cp	No.	t, °C	η, cp
I	93	I,290	I8	I7,2	I,89I
2	54,6	I,592	I9	35,5	I,7I6
3	35,5	I,720	20	48,7	I,375
4	30,2	I,767	2I	70,[illegible]	I,4I6
5	25,2	I,827	22	90,8	I,30I
6	22,3	I,84I	23	75,3	I,4I6
7	I9,6	I,884	24	5,3	2,075
8	I3,0	I,960	25	3,I	2,I00
9	I0,9	I,995	26	0,9	2,I74
I0	2,3	2,I44	27	30,6	I,77
II	9,3	2,043	28	44,0	I,625
I2	I2,7	I,982	29	4I,7	I,634
I3	I9,2	I,896	30	30,6	I,753
I4	27,6	I,804	3I	63,6	I,485
I5	39,5	I,684	32	96,6	I,285
I6	60,0	I,495	33	64,6	I,485
I7	79,7	I,364			

the surface film at a point where the probe comes out of the liquid. It is possible that the quasicrystalline formations in gallium were subjected to the mechanical breakdown in the immediate vicinity of the vibrating probe. The difference in the results can also be due to the varying purity of the examined metal and varying displacement rate which is considerably greater in the vibration method than in the method of torsional vibrations. We should mention that at the present time the reasons for large differences between the various experimental data are not explained.

Viscosity of Cooling Liquid "Antifreeze-65". The cooling liquid "65" (ГОСТ-159-52) [ГОСТ = GOST = All Union State Standard] is widely used as a working liquid in the cooling systems of equipment. Despite its wide use, there are no data in the literature on the viscosity of the cooling liquid "65", which makes the designing of the cooling systems more difficult. Measuring the viscosity of antifreeze presents certain difficulties, since the light components evaporate faster than the heavy components during the measurement, and the quantitative composition of the cooling liquid can change continually during the experiment. The measurements were done on the same installation used to measure gallium. The viscometer was calibrated using ethylene glycol, butyl alcohol, and water. A portion of the test liquid was poured into the working cup and heated to a certain temperature; to avoid distillation the cup was tightly covered with a plug. Then the liquid was raised rapidly up to a certain level and the vibration amplitude of the probe was measured. After 2-3 measurements the liquid portion was changed. The measurements were carried out in the 15.5-92°C temperature range. Figure 10 shows the results of the experiments. The deviation from the interpolated curve does not exceed ±3%. We note that the scatter of the experimental points is caused, basically, not by the experimental errors but by the change in composition of the cooling liquid during the measurements. The viscosity values taken from the interpolation curve are presented in Table No. 2. It also shows the data for temperatures up to -50°C obtained by extrapolation using the Frenkel formula.

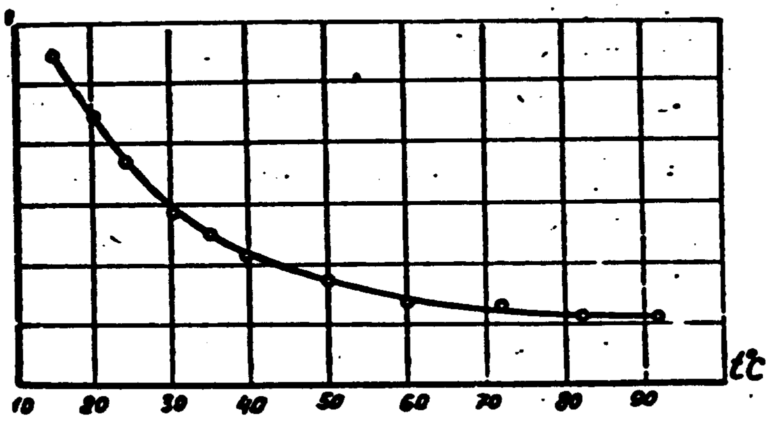

Fig. 10.

Table No. 2

t, °C	η, cp	Notation	t, °C	η, cp	Notation
-50	534	Extrapolated data	30	6,03	Experimental data
-40	255	- " -	35	5,08	- " -
-20	132	- " -	40	4,35	- " -
-20	71	- " -	45	3,80	- " -
-10	40	- " -	50	3,38	- " -
0	24	- " -	60	2,80	- " -
10	14	- " -	70	2,48	- " -
15	11,30	Experimental data	80	2,25	- " -
20	9,05	- " -	90	2,10	- " -
25	7,30	- " -	100	2,00	- " -

Sodium Viscosity at the Melting Point. As is known, the existing methods for measuring the viscosity of liquids do not permit the measurement of viscosity in the immediate region of their melting points, due to the fact that in all known methods, the time of one measurement at a constant temperature is too long. In this regard, the vibration method has indisputable advantages since it allows approaching the melting point, and especially the solidification point, as close as desired. And the viscosity of liquids at melting points is of unquestionable theoretical and practical interest.

Up until now, the viscosity value of metals at melting points could only be obtained by the extrapolation of the experimental data, using the formulas which describe well the viscosity behavior of a liquid close to the melting point, for example, according to the Andrade formula (10). The validity of such extrapolations still requires the experimental confirmation. We took the advantage of the continuity of the vibration method in measuring the viscosity and the virtually instantaneous readout of amplitude of the forced vibrations of the probe in order to study the viscosity value of sodium in the immediate region of its solidification point.

The studies were done on the installation (Fig. 7) which was somewhat modified to suit the conditions of the experiment. Stainless steel crucible (1) in Fig. 11 with a 30 mm inside diameter is adjoined to condenser (9) so that it is an air-tight connection (Fig. 7). Two conical tubes (4), one for illumination and the other for the observation of the level and purity of the metal surface, are welded to the sides of the crucible. The external surface of the crucible is tightly fitted with a copper jacket (5) in order to equalize the temperature along its length. Glycerin from thermostat U-8 was fed through a coil wound above the temperature-control unit. The temperature was measured by the chromel-alumel thermocouple and platinum thermometer of resistance. The thermocouple was introduced into the center of the melted liquid 3-4 mm below the probe by means of a thin-walled shell (7) welded to the walls of the crucible. The thermometer of resistance was placed between the copper jacket and coil. The metal was filled from the bottom through stainless steel T-junction (8). One T-junction union is connected to the crucible, the second to sylphon valve (14), and the third to the sampler for metal analysis (10) or to the calibration liquid filling system. Sylphon batcher filled with sodium (12) is connected to one of the unions of sylphon valve (13). The filling of the batcher was done at the distillation unit. We used class "4" sodium containing 99.8% of the basic substance. Before filling it with the calibration liquid and sodium, the installation was thoroughly evacuated for several hours to (2-3) 10^{-5} mm Hg and then filled with purified argon up to a pressure of 1.3 atm.

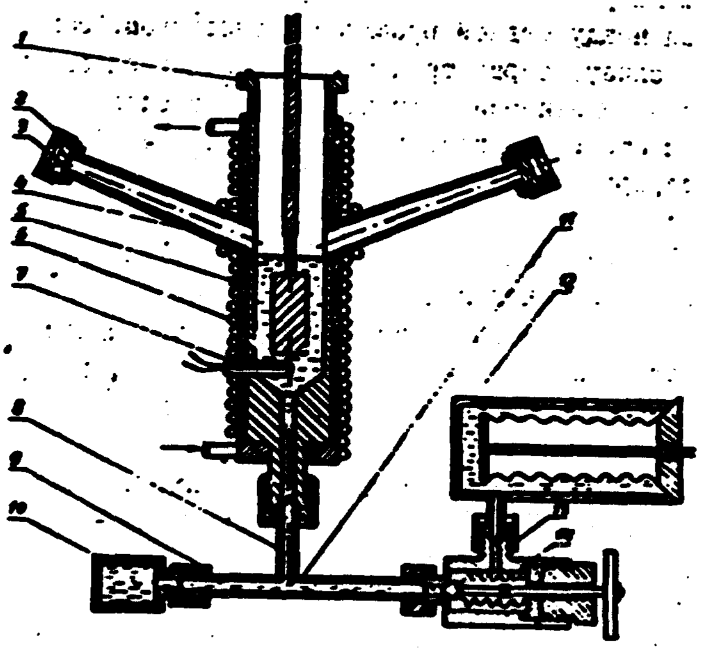

Fig. 11.

The viscometer calibration was carried out under the conditions identical for the measurements, i.e., with the batcher with sodium connected and its sylphon valve closed during the calibration. In the preceding experiments we were convinced that in the interval $\sqrt{\rho\eta}$ = 0.5-0.8 (cp cm^{-3})$^{1/2}$ the calibration of our viscometer strongly depends on the distribution and tightening of various units and can vary by ±3% in terms of $\sqrt{\rho\eta}$ if anything is changed after the calibration. In our work the only nut on the second union of T-junction (9) was loosened three times: during the filling with toluene, water, and when the sampler (10) was connected. Water and toluene were our calibration liquids. The maximum deviation of points from the calibration curve was ±0.5%.

The calibration can also be done using the known viscosity values in the literature, if these data are sufficiently reliable for any temperature region. This is the direction we have taken, since we are interested in the temperature course of the viscosity close to the melting point and not the absolute values. As reference we used the data in [12].

As a result of high chemical activity of alkali metals their surface, in case the inert gas in the installation is not sufficiently pure, is quickly covered with a film, representing their complex union with oxygen, nitrogen, and hydrogen. These films decrease the amplitude of forced vibrations by several percent, therefore, we paid a special attention to purifying the argon from the water and gas vapors. First the argon was bubbled through an eutectic sodium-potassium mixture, and before its introduction into the installation it was passed through a heated (700-800°C) filter made from metallic titanium filings. The beginning of the film formation on the sodium surface is indicated by the developing dependence of the amplitude of forced vibrations on time (amplitude is decreased).

Even under the most thorough purifying conditions, some contamination can still occur on the walls of the connecting tubes, valves, and crucible. Therefore with a first filling, as a rule, a film of oxides and contaminations could be observed. Near the melting point the sodium does not wet the probe. Therefore, after the first filling it is possible to discard the contaminated part of the metal and to refill the crucible. If necessary, the operation can be repeated.

In this manner the probe turns out to be thoroughly cleaned from the contaminations. In this case, there appears a "slip" phenomenon which is examined in detail in the next section exemplified by mercury. In contrast to mercury, the vibration amplitude of the plate in the sodium changes in time even if the plate is polished. Sometimes, for several hours the vibration amplitude was greater than the normal (i.e., determined by the viscosity and density of the sodium) and remained virtually constant. Due to this fact, we were unable to achieve the reproducibility of the indicators with a non-wetted probe. The wetting of the probe was accomplished when the sodium was heated to 500-600°C, which could be observed well by the change in the form of the meniscus. After the wetting, the amplitude of vibrations was stable and was reproduced well.

Figure 12 shows the experimental values of sodium viscosity from the melting point to 150°C. The mean square deviation of points from the mean curve is ±0.5%. The measurements were done at the stationary points during the rise or fall of the temperature, and also after the solidification and a subsequent melting of the sodium. All series of measurements coincided within the mean square deviation. Up to 98°C, i.e., 0.4° before the melting point, there were no anomalies of viscosity observed. Then a sharp decrease in the vibration amplitude was observed (isolated points on the graph of the temperature dependence of viscosity, Fig. 12). An anomalous decrease in the vibration amplitude in the vicinity of the solidification point can be caused to a lesser degree by three reasons.

First, in the working space there can occur such a distribution of temperatures that the metal is not crystallized all at once but from the surface of the probe toward the vessel walls. In this case, the vibrations will not stop but will attenuate strongly. Another temperature distribution is also possible during which the solidification will occur in such a way that the vibrations will not stop.

Second, it can occur that in the closest proximity to the solidification point the vibration amplitude decreases due to the appearance of a surface film which, however, cannot be seen through the observation window.

And finally, the viscosity can have an anomalous temperature course connected with the precrystallization processes.

We tend to think that the anomalous decrease in the vibration amplitude close to the solidification point is connected with the first two reasons.

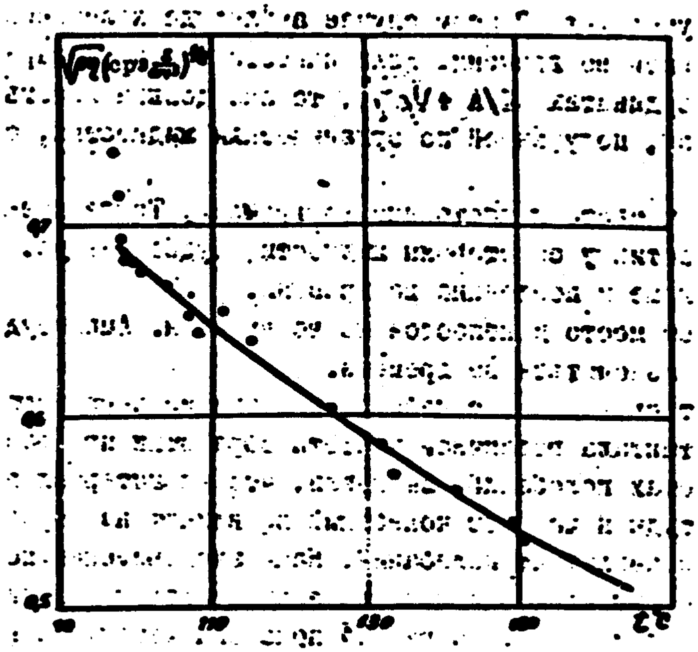

Fig. 12.

According to our thermocouple, the solidification point of the sodium studied by us turned out to be 97,6, i.e., somewhat lower than the known data [12]; apparently, this is explained by the presence of small amounts of the potassium additions, which have insignificant effect on the absolute viscosity value but can shift the melting point toward the side of lower temperatures. The relative error in the data on sodium was estimated by us to be 1.5%.

Study of the "Slip" Phenomenon.

In carrying out experiments with the non-wetting liquids, we have detected that sometimes the vibration amplitude would change significantly with time. We tentatively called this occurrence "slip" and have carried out special experiments in order to determine the reasons for occurrence of this phenomenon. The experiments were done with mercury. The measurements were done in a glass vessel with double walls, which enabled us to control the level and state of the mercury surface. The mercury temperature was maintained close to that of the room in order to decrease the time necessary for the equalization of the mercury and probe temperatures. The mercury level was raised rapidly to a given mark on the probe and, at the same time, the auto-oscillations were switched-on. The "rocking" of the mercury level did not change the vibration amplitude; consequently, the surface layer does not affect the vibrations. The time dependence of the vibration amplitude was investigated. Here, three cases are possible.

1. Slip is absent. In this case the vibration amplitude must remain constant in time. If the point is plotted on the calibration graph (on coordinates $1/A$, $\sqrt{\rho\eta}$). then it should lie on the calibration curve obtained for the organic liquids wetting the probe.

2. Slip occurs and is constant in time. Then the viscous force acting on the plate on the part of the liquid will be less, while the vibration amplitude greater and constant in time.

3. Slip occurs and is not constant in time. The vibration amplitude will also change in time.

The experiment was carried out on a set of plates made from stainless steel, molybdenum, and tantalum with varied finishing treatment at various amplitudes and vibration frequencies. We have found that neither the plate material nor the magnitude of amplitude and vibration frequency affect the nature of the vibration amplitude time dependence. Our experiments have shown that there is a slip and its presence depends only on the degree of machining of the probe. Figure 13 shows a characteristic dependence of the vibration amplitude on time for plates made from stainless steel with $\nabla 3$ treatment. As seen from the figure, the time decrease in the vibration amplitude is approximately 10% and it is almost monotonic. The change in the amplitude is greatest during the first moment after the submersion and it stabilizes after 3-4 hours. The stable amplitude quantity has a value determined by the viscosity of mercury. The point corresponding to the constant amplitude falls on the calibration graph obtained using the organic liquids.

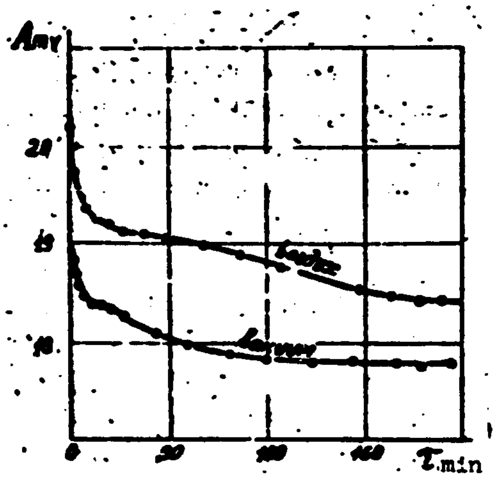

Fig. 13.

We have obtained the same results when the measurements were done under the vacuum of 10^{-2} mm Hg. However, the time required for the amplitude to stablize is 1.5-2 h, which is considerably less.

Similar results were obtained also for the molybdic cylindrical probe of approximately the same finishing treatment. Just as in the case of the stainless steel, the measurements were done at two different amplitudes differing by a factor of two and at various frequencies - 120 and 180 Hz. The nature of time dependence in each mode of operation remained constant.

A different result was obtained when we investigated the vibration amplitude time dependence for the plates with high finishing treatment **(▼6).** The studies were done on plates made from stainless steel, molybdenum, and tantalum. Throughout the measurement the vibration amplitude remained constant within the error limits of the experiment.

At the present time we are unable to propose with certainty a mechanism explaining the change in the vibration amplitude with time. It can be assumed that there is an adsorbed layer of air on the plate surface. The mercury does not come in contact with the plate immediately and it seems as though there is an air pillow between them; furthermore, the amount of adsorbed gas depends on the roughness of the plate. In time the mercury forces out the gas layer and adheres to the plate. However, this is contradicted by the fact that the slip occurs also at 10^{-2} mm Hg. However, it is entirely possible that the slip would disappear if a vacuum of several orders higher could be attained and the plate was preannealed.

An attempt to explain the vibration amplitude change in time can also be made as follows. Since mercury does not wet the plate, it does not fill the depressions on the plate surface immediately due to the surface tension forces. It would appear the contact surface of mercury with the plate is less as compared with the contact surface of a perfectly flat plate. In time the mercury flows into the depressions and the friction surface becomes normal. The vibration amplitude change in time was also observed in other liquid metals.

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