INVESTIGATION OF THE DENSITY OF LIQUID LEAD, CESIUM, AND GALLIUM BY THE GAMMA-METHOD

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In investigating the density of metals in the liquid state, use has been made of the contactless gamma-method based on the weakening of penetrating radiation by the metal. The method is widely used in technology for measuring the density of solids and liquids [1, 2] at low temperatures, and recently it has begun to be used in physicochemical studies of melts at high temperatures [3, 5]. An analysis of the published work shows that the potentialities of the gamma-method have not yet been fully exploited.

The experimental setup for determining the density of melts by the gamma-method is described below, and the results of density measurements on three liquid metals are given.

Calculated relationships. Under actual experimental conditions in the high-temperature apparatus, there are in the path of the gamma beam from the source to the detector, besides the layer of material being investigated (with thickness δ, density ρ, and mass attenuation coefficient μ), "parasitic" absorbing media, such as parts of the apparatus, crucible walls, and the gas filling the body of the furnace. Under these circumstances the intensity of the primary radiation entering the detector (for an initial intensity I of the narrow beam) is

$$I_g = I \exp\left(-\mu\rho\delta - \sum_i \mu_i\rho_i\delta_i\right) \qquad (1)$$

where the terms μ_i, ρ_i, and δ_i, represent the absorbing power of the "parasitic" media.

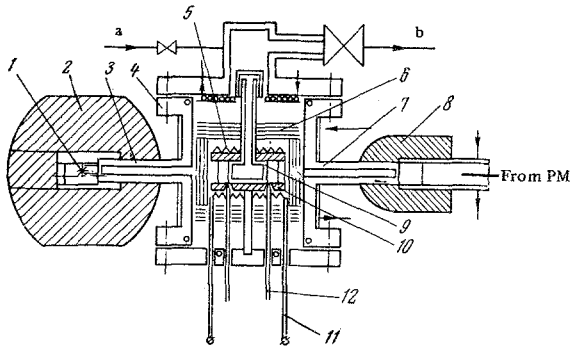

Fig. 1

Since the accuracy of the absorption coefficients found in tables is low, it is best to determine $\Sigma\mu_i\rho_i\delta_i$ experimentally in the absence of the liquid. It is convenient to use a relative variant of the method, i.e., to determine the temperature dependence of the density. The formula for the relative method is

$$\rho = \rho_1 \frac{1+\alpha t_1}{1+\alpha t} \frac{\ln I_0/I}{\ln I_{01}/I_1} \qquad (2)$$

where ρ and ρ_1 are the densities of the liquid at temperatures t and t_1 respectively; α is the coefficient of linear expansion of the crucible material; I_0 and I_{01} are the radiation intensities recorded by the dectector at temperatures t and t_1 in the absence of the liquid; and I and I_1 are the intensities at t and t_1 when the liquid is placed in the crucible. I_0 and I_{01} have to be determined by direct experiment.

If the discrete method of recording the radiation is applied and the radiation intensity is directly proportional to the mean counting rate n, Eq. (2) assumes the form

$$\rho = \rho_1 \frac{1+\alpha t_1}{1+\alpha t} \frac{\ln n_0/n}{\ln n_{01}/n} \qquad (3)$$

where n_0, n_{01}, n, and n_1 are the mean counting rates corresponding to the intensities I_0, I_{01}, I, and I_1.

Experimental setup. A diagram of the apparatus intended for the investigation of the density of liquid metals, including alkali metals, is shown in Fig. 1.

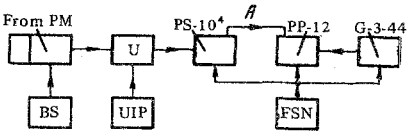

Fig. 2

The metal to be investigated is placed in cylindrical crucible 9 having thin, flat end walls. The crucible containing the metal is put into thick-walled nickel block 10, whose purpose is to equalize the temperature; four thermocouples are embedded in the inner surface of this block, while electric heater 5 is wound on the outside of the block. The assembly consisting of the heater and the block containing the crucible is surrounded by series of thin metallic screens 6 and is mounted on the lower part of cylindrical furnace body 4, in which a vacuum down to 10^{-5} mm Hg or an inert-gas pressure up to 200 atm can be achieved. The crucible is accurately located along the axis of the radiation beam by means of adjusting screws.

The narrow beam of γ-quanta is limited by a system of two coaxial collimators—one near source 3 and one near detector 7—with a hole 5 mm in diameter; these are welded into the body of furnace 4. The total length of the beam's path from the radiation source to the detector is 550 mm.

Radiation source 1 (Co^{60} with an activity of ~1 Ci was used) is put in a rotating device attached to the source collimator and can occupy two rigidly fixed positions: "working" and "storage." In the first case it is situated exactly opposite the hole in the collimator, and in the second case it is moved aside through an angle of ~30°.

The measuring part of the setup consists of apparatus for determining the temperature of the metal under investigation, the pressure in the body of the furnace, and the intensity of the radiation.

The temperature of the metal was measured with four thermocouples (PP or KhA) in conjunction with an R-307 or EPP-09 potentiometer; the pressure in the furnace with a standard class-0.4 manometer and the vacuum with a VIT-1A vacuum meter.

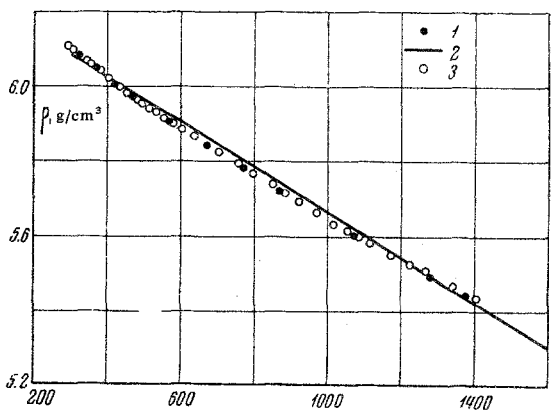

Fig. 3

A block diagram of the apparatus for measuring the radiation intensity is given in Fig. 2.

The radiation detector is a scintillation counter made from a NaI (Tl) single crystal 40 mm in diameter and 40 mm thick, and a FEU-13 photomultiplier.

Table 1

Density of Liquid Lead

T,°K	ρ, g/cm³
645.7	10.64
690.1	10 54
717.4	10.52
838.4	10.38
949.8	10.20
1019	10.14
1109	10.03
1152	9.95
1246	9.85
1329	9.75

The impulses from the photomultiplier PM enter the amplifier U, for which a USS block slightly modified from a PP-8 radiometer was used. The amplified impulses are fed to a PS-10 000 at whose input the signals from the PM and the amplifier are separated. The working signal is calculated by a scaling circuit.

A separate electronic meter circuit is used to measure accurately the time intervals; it consists of a precision G-3-44 generator and a PP-12 counting device. The two counting devices (i.e., the scintillation impulse counter PS-10 000 and the time meter PP-12) are started and stopped by a single NP-contact on the PS-10 000.

In the recording of the radiation with a scintillation detector, the principal error is known to be introduced by the instability of the amplification coefficient of the FEU; consequently special measures were taken in our experiments to increase the stability.

The detector was mounted in a housing that had a thermostatic jacket. The PM was fed from a battery source (BS) of about 5 amp. hr capacity, and the operating voltage was chosen from the flattest part of the plateau of the counting characteristics, which were obtained experimentally (up to counting rates of 60 000 counts/sec). In addition we found that most consistent results and good reproducibility during prolonged operation (several days) could be obtained by using a special method of feeding the FEU, in which the working voltage was not applied continuously but with interruptions during which the PM "rested". The voltage is not entirely removed from the FEU during the "rest" period; it only decreases to a value at which the amplitude of the scintillation impulses becomes approximately equal to that of inherent noises of the FEU at the working voltage.

Table 2

Density of Liquid Cesium

T°,K	ρ, g/cm³	T,°K	ρ, g/cm³
306.7	1.836	785.7	1.562
315.3	1.829	797.9	1.552
323.9	1.826	852.7	1.526
366.1	1.802	870.9	1.519
369.7	1.800	884.0	1.508
435.6	1.762	906.2	1.497
483.2	1.736	977.2	1.460
521.1	1.715	987.5	1.447
526.4	1.712	1013.2	1.434
541.8	1.703	1043.6	1.422
587.1	1.684	1041.4	1.416
622.3	1.657	1076.5	1.394
669.3	1.631	1077.2	1.399
689.8	1.627	1115.0	1.370
752.2	1.589	1125.2	1.372

Experimental procedure and treatment of the results. The same method of measuring the radiation intensity was used throughout. A reduced voltage (the "rest" voltage) was applied to warm up the PM and at the same time the measuring apparatus was switched on. After the electronics warmed up and the furnace reached the required temperature conditions, the working voltage was applied to the PM. While the necessary conditions were being established in the PM (10–12 min), the counting rate was measured 6–7 times with exposures of 100–300 sec; the time intervals between the measurements and the time intervals from the moment of switching on the working voltage that supplies the PM to the beginning of the first measurement were recorded. The external conditions in operating the PM were kept the same in all the measurements. At the end of the measurements the voltage applied to the PM was reduced to the "rest" voltage at which the PM was under a different temperature regime during the furnace output.

Before the experiments with the metal were begun, the whole furnace assembly including the empty crucible was installed in the apparatus and calibration measurements were made, i.e. the background was determined (this was about 2% of the working level in all the experiments) and so was the initial counting rate n_0 at room temperature and at various gas pressures in the body of the furnace (from 10^{-5} mm Hg to 15 atm). It was shown in preliminary experiments that the change in the residual "parasitic" absorptions with temperature is negligible.

After the calibration had been completed, the crucible was filled with metal and the main experiments were begun, in which the temperature dependence of the counting rate was determined.

Table 3

Density of Liquid Gallium

T°,K	ρ, g/cm³	T°,K	ρ, g/cm³
295.6	6.1031	596.5	5.8940
296.0	6.1021	603.7	5.8884
307.1	6.0930	624.5	5.8776
314.4	6.0880	635.0	5.8700
321.4	6.0830	676.1	5.8457
340.5	6.0669	706.8	5.8245
359.1	6.0550	708.1	5.8240
380.5	6.0388	761.9	5.7930
404.4	6.0209	798.7	5.7682
421.3	6.0119	850.8	5.7396
436.6	5.9992	880.7	5.7162
453.4	5.9818	920.5	5.6998
472.0	5.9708	971.9	5.6627
482.8	5.9668	992.1	5.6627
485.4	5.9646	1018.2	5.6342
499.0	5.9562	1059.5	5.6176
509.0	5.9491	1085.1	5.6020
515.9	5.9386	1112.8	5.5871
522.7	5.9385	1171.0	5.5514
536.5	5.9310	1220.7	5.5290
552.3	5.9215	1263.9	5.5107
554.4	5.9179	1342.1	5.4704
583.4	5.8994	1407.7	5.4359
593.8	5.8950		

All the experiments were carried out under stationary conditions, the temperature not varying by more than 2 deg/hr. Measurements were started at the nearest possible temperature to the melting point of the metal, and at the beginning of each new day the conditions were invariably repeated in the temperature range already investigated.

The mean temperatures and the mean values of the counting rates were used in the calculations. A correction for errors in the counting device was applied to the measured counting rates, and the magnitude of the background and the change in activity of the radiation source were taken into account.

Results of measurements. The density of lead was determined mainly to check the suitability of the apparatus. A stainless-steel crucible 38 mm long was filled with "Ch"-lead conforming to specification TU 70-56 (with a controlled impurity content not exceeding 0.035%). The reference value of the density ρ_1 was derived from the equation in [6] for a temperature of 717° K. The results of the measurements, given in Table 1, can be represented by the equation

$$\rho_f\,(\mathrm{Pb}) = 10.676 - 12.887 \cdot 10^{-4}\,(T - 600.6)\ \mathrm{g/cm^3}$$

with a mean square deviation of ±0.16%.

The maximum error of a single measurement at 1320° K is estimated to be ~0.4%, taking no account of the error in determining ρ_1.

The cesium used contained 99.9% Cs. The crucible (of stainless steel and 100 mm long) was filled with cesium in an argon ambient. Owing to a failure of the heater, it was not possible to raise the temperature above 1123° K. The results of the measurements, given in

Table 2, can be represented by the equation

$$\rho_f\ (\text{Cs}) = 1.8390 - 5.665 \cdot 10^{-4}\ (T - 301.4)\ \text{g/cm}^3$$

with a mean square deviation of 0.23%. This equation differs very little from the one in [7], from which the reference point for a temperature of 306.7° K was derived.

The estimated maximum error of measurement at 1120° K is ~0.7%, taking no account of the error in determining ρ_1.

Last in this series of experiments, gallium was investigated (in a quartz crucible 60 mm long).

The results, obtained on gallium conforming to specification RETU 851-61, are presented in Table 3 and Fig. 3, in which they are compared with the data of other authors [8, 9]. Over the whole temperature range our data coincide within 0.1% with Hoather's data [8] (from which the reference point for 307.1° K was derived) and can be interpreted as consisting of three straight line sections for temperatures from
the melting point to 505° K

$$\rho_f\ (\text{Ga}) = 6.09538 - 7.208 \cdot 10^{-4}\ (T - 302.93)\ \text{g/cm}^3$$

505 to 1020° K

$$\rho_f\ (\text{Ga}) = 5.94933 - 6.122 \cdot 10^{-4}\ (T - 505)\ \text{g/cm}^3$$

1020 to 1400° K

$$\rho_f\ (\text{Ga}) = 5.63464 - 5.127 \cdot 10^{-4}\ (T - 1020)\ \text{g/cm}^3$$

The mean square deviation of our data from these relationships does not exceed 0.05% in the respective temperature ranges. The maximum error of measurement at 1400° K is estimated as 0.25% (taking no account of the error in determining ρ_1).

In a recently published paper [9], the density of liquid gallium was determined by the horizontal drop method. The authors estimated the error in their measurements as 0.6% and represent their data up to 1700° K by a single linear relationship, which coincides with our data within the limits of accuracy indicated.

The coefficient of volume expansion derived from our data changes rather sharply in the vicinity of 505 and 1020° K. These breaks should also be visible in the temperature dependences of other properties.

Such a break was in fact found by Goryaga and Morgunova [10] at about 620° K on the viscosity (η)-density curve plotted in the coordinates of A. I. Bachinskii's formula. A break is also clearly visible on the graph of $\ln\eta$ plotted against $1/T$ constructed from Spells' data [8]. A similar break was also observed at about 520° K in the temperature dependence of the surface tension according to data in [11], although the authors prefer to interpret their data by a single quadratic relationship.

There are also indications that at about 540° K anomalies occur in the temperature dependences of the electrical and thermal conductivities.

All this provides evidence that in the region of 505° K a considerable structural change takes place in liquid gallium; this is possibly due to the dissociation of Ga_2 molecules, whose existence in the liquid phase has been reported in the literature (see [12, 13]).

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