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Viscosity of Hg-In Amalgam Near the Melting Point

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Accurate measurements of the viscosity of Hg-In amalgams have been carried out using an oscillating viscometer and their interpretation is briefly discussed.

1 INTRODUCTION

In a previous paper¹ the viscosity of the amalgam Hg-In has been investigated in the temperature range from 30°C to 120°C, using a special kind toroidal oscillating viscometer.

In Ref. 1 it has been found that the viscosity rises rather fastly with the decreasing of temperature: it seemed therefore interesting to investigate the viscosity behaviour of the above amalgam going down in temperature as near as possible to the liquidus/(liquidus + solid) curve in the phase diagram.

As a matter of fact the eventual influence of residual short range correlations on viscosity should become more important in the neighbourhood of liquid-solid transition.²

In order to perform such an investigation some details of the experimental apparatus used in Ref. 1 had to be modified.

The new experimental apparatus is described briefly in Section 2. In Section 3 the experimental results are given and are discussed in Section 4.

2 EXPERIMENTAL APPARATUS

The toroidal oscillating viscometer used in the present investigation is the same as employed previously. A complete account of the viscometer and of the entire experimental apparatus can be found in Ref. 1.

In order to reach -15°C , the viscometer has been placed in contact with a completely different thermostat. In fact the copper cylindrical box containing the viscometer, which oscillates in argon atmosphere at ~ 300 mbar, is sunk in a thermostatic bath, whose fluid is a special kind of silicon oil.†

The fluid temperature can be regulated continuously from 40°C to -15°C , and the fluctuations are not greater than 0.1°C . The temperature of the copper box is measured by means of a thermocouple, connected to an electronic voltmeter (type Coreci PEP 2.15 D) and graphically recorded versus time. To be sure that the viscometer and the copper box are in thermal equilibrium a convenient time delay (~ 12 h) is introduced, from the instant when the thermostat fluid changes temperature and the viscosity measurements are performed. Adopting such a procedure, the temperature of the amalgam inside the viscometer is perfectly constant during each measurement and is known with an accuracy of a few tenths of a degree centigrade.

As previously, the viscometer oscillations have been recorded using a light beam follower (Photodyme graphispot), and the recordings were good enough to allow a determination of the damping coefficient δ , with an accuracy $\sim 5\%$.

3 EXPERIMENTAL RESULTS

The viscometer dampings δ have been measured for each alloy composition (3, 15, 30, 45 atomic percent) at fourteen different temperatures from 40°C to -15°C .

For each damping the viscosity η has been calculated using formula (1) of Ref. 3 which for convenience is written below

$$\frac{I\sqrt{2}}{4\pi^3 a^2 R^3 \rho} \left[\left(1 + \frac{T^2}{T_0^2} \right) \delta - 2 \frac{T}{T_0} \delta_0 \right] = G_1(q) - \delta G_2(q) + \frac{a^2}{R^2} G_3(q). \quad (1)$$

In the above formula I is the moment of inertia of the empty oscillating system, (T , T_0 ; δ , δ_0) are the periods and logarithmic decrements with and without liquid respectively; G_1 , G_2 , G_3 are universal functions of $q = (2\pi\rho a^2/\eta T)^{1/2}$ tabulated in Ref. 3, ρ is the liquid density and finally a , R are the radius of the toroidal channel and of the torus respectively.

The experimentally determined logarithmic decrements are introduced in Eq. (1).

† Silicon Fluid AK5-B. H. Schilling (Germany).

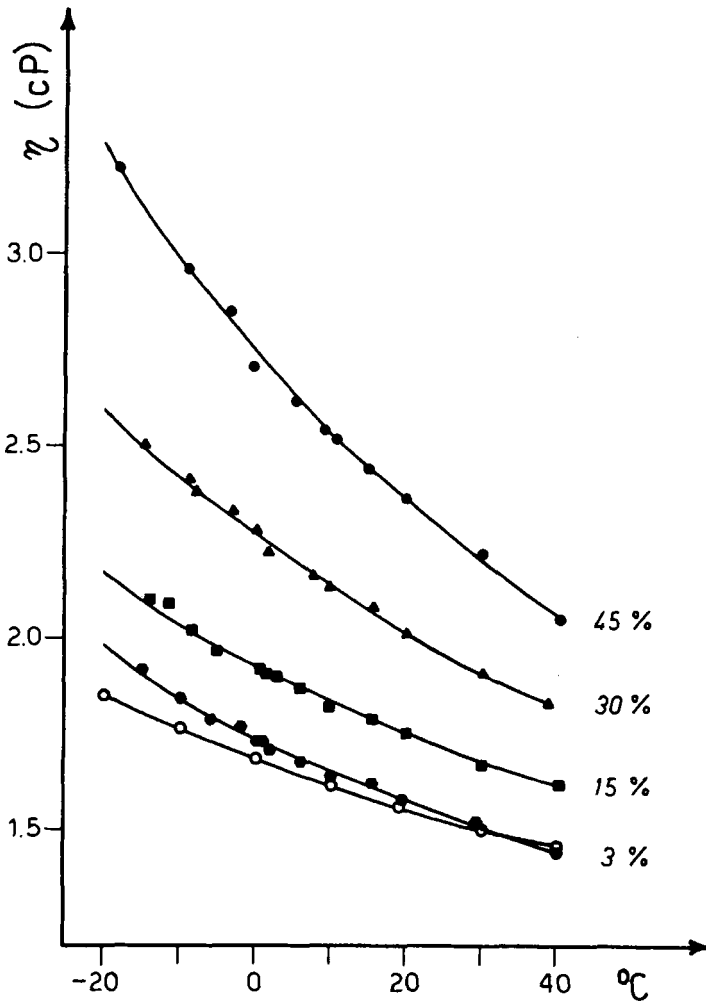


FIGURE 1 Experimental data of the viscosity η versus t ($^{\circ}\text{C}$) and q_{in} , together with an exponential best fit.

In order to get q (and from q to get η) it is necessary, however, to know ρ . Data on the density of Hg/In alloy have been obtained from Ref. 4 and conveniently elaborated to get ρ at each temperature and at each alloy composition.

The analysed experimental results are plotted in Figure 1. In Figure 2 the solid lines represent the dependence of η on the alloy composition, η being derived from the exponential best-fit (see Section 4) of the experimental data, plotted in Figure 1.

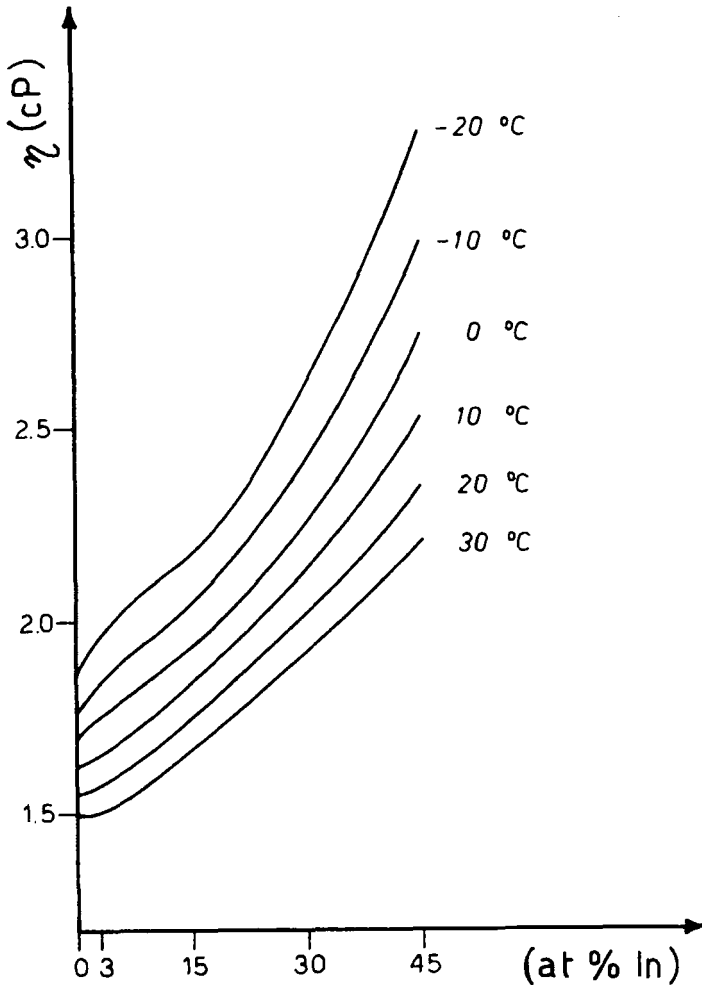


FIGURE 2 Behaviour of η versus q_{In} , deduced from the best fits plotted in Figure 1, at various temperatures from 30°C to -20°C. It must be observed that for $q_{In} = 45\%$ and $t = -20^\circ\text{C}$ the amalgam begins to solidify.

4 DISCUSSION OF THE EXPERIMENTAL RESULTS

The experimental results, collected in Figures 1 and 2, show the following qualitative aspects.

a) The temperature behaviour of the viscosity is quite similar to the one measured at higher temperature: however the fluctuations observed previously have disappeared almost completely.

TABLE I

At percent In content q_{In}	0%	3%	15%	30%	45%
E_{η}	0.647	0.818	0.793	0.934	1.211
c	0.512	0.385	0.448	0.405	0.294
$(1 - r^2) \times 10^{-3}$	0.2	8.7	8.1	3.8	3.0

- b) The viscosity rises smoothly with the increase of the indium content.
- c) The viscosity minimum, for an alloy composition in the neighborhood of 3 atomic percent indium found previously, is still present but disappears rapidly at lower temperatures.†

In order to understand the meaning of the experimental result a) an Arrhenius type law

$$\eta = ce^{E_{\eta}/RT} \tag{2}$$

has been used to fit the data. The parameters determined by a least square method are collected in Table I.

In the second and third rows of Table I the values of E_{η} (in kcal/mol) and of c are given respectively for each amalgam composition. In the fourth row the corresponding values of $1 - r^2$ are reported, where r^2 is the coefficient of determination,⁶ which spans between 0 and 1 and is a measure of the goodness of the exponential regression: the nearer $r^2 \rightarrow 1$, the more the experimental points approach the exponential law.

In Figure 3 the values of E_{η} are plotted as a function of q_{In} , together with the corresponding values obtained in Ref. 1. It appears that for $q_{In} < 30\%$ the value of E_{η} that fits the data in the lower temperature range ($-15 \div 40^{\circ}\text{C}$) could fit the data also in the higher temperature range ($30 \div 130^{\circ}\text{C}$). On the contrary for $q_{In} > 30\%$ E_{η} becomes appreciably different in the above two temperature ranges. This means that there is a breakdown of the simple Arrhenius type law when the indium content becomes sufficiently great.

As a matter of fact for $q_{In} = 45\%$ the experimental viscosity data may be fitted using a linear combination of at least two different exponentials.

The behaviour of η versus q_{In} , given in Figure 2 are fitted by a straight line, according to the rule valid for a simple two component "mixture."

$$\eta = q_{In}(\eta_{In} - \eta_{Hg}) + \eta_{Hg} = a_0(t) + a_1(t)q_{In} \tag{3}$$

† The minimum observed in Ref. 1 is actually less pronounced: in fact in present work a more careful determination of the viscometer parameters a, R, I , gives rise to a 5% increase of the viscosity values.

Present calibration has been tested by measuring the viscosity of pure mercury, which turned out to be in complete agreement with values given in the literature.⁵

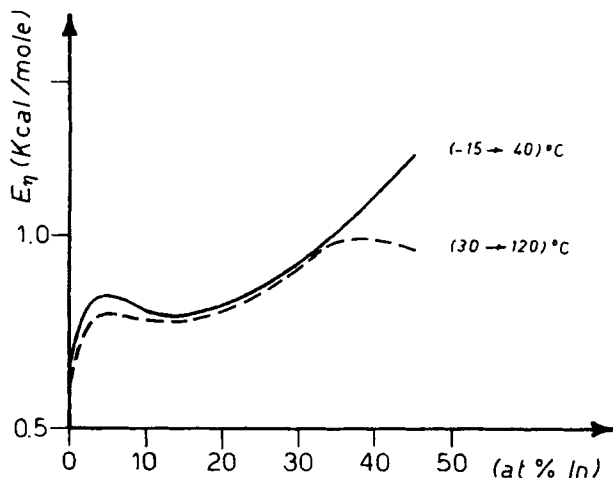


FIGURE 3 Behaviour of E_{η} versus q_{In} in the two temperature ranges: $(-15 \rightarrow 40)^{\circ}\text{C}$, $(30 \rightarrow 120)^{\circ}\text{C}$.

which indicates a direct proportionality between the viscosity of the alloy and the In content.

A least squares method has been used to fit the viscosity versus q_{In} .

In Table II the values of a_0 , a_1 , and of their corresponding relative standard errors S_0/a_0 , S_1/a_1 are indicated. In the sixth row the viscosity of pure mercury are indicated, which are to be compared with a_0 .

It is possible to see by inspection, that the relation (3) represents fairly well the behaviour of the viscosity above room temperature while going down in temperature the linear approximation becomes worse.

This result is easily understood; in fact relation (3) disregards completely any interaction between ions of the two elements of the alloy.†

For what concern point c , that is the existence of a minimum in the viscosity for low values of q_{In} , it must be pointed out that the minimum disappears as soon as it drops below room temperature.

To see more clearly where the minimum comes from, in Figure 4 the values of c versus q_{In} are plotted.

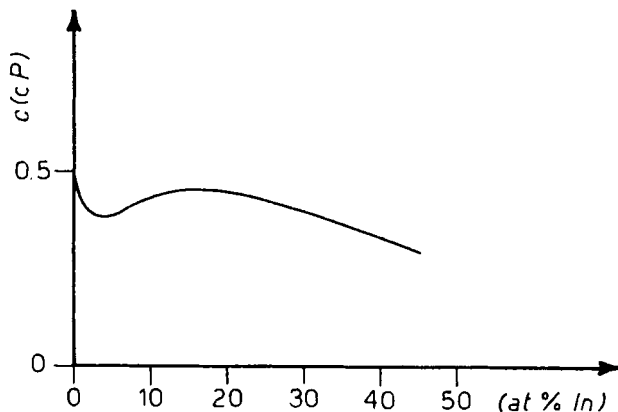
In the figure it appears that c drops rapidly as soon as q_{In} becomes different from zero and reaches clearly a minimum. On the other hand according to the "hole theory" c should be proportional to the transfer probability from the actual position of the ion to an empty "hole."

The lowering of c could therefore be interpreted as a partial filling of the "holes" in the undisturbed mercury matrix.

† As a matter of fact in order to describe the behaviour of η versus q , a 2nd order polynomial has usually been employed.⁶

TABLE II

Temperature (<i>t</i>)	-20	-10	0	10	20	30
a_0	1.812	1.725	1.648	1.579	1.519	1.464
S_0/a_0	4.1×10^{-2}	3.3×10^{-2}	2.7×10^{-2}	2.1×10^{-2}	1.7×10^{-2}	1.5×10^{-2}
a_1	3.007×10^{-2}	2.616×10^{-2}	2.288×10^{-2}	2.012×10^{-2}	1.77×10^{-2}	1.573×10^{-2}
S_1/a_1	9.8×10^{-2}	9.8×10^{-2}	8.7×10^{-2}	6.7×10^{-2}	5.9×10^{-2}	5.4×10^{-2}
η Hg	1.855	1.764	1.685	1.615	1.554	1.499

FIGURE 4 Behaviour of c versus q_{In} .

By increasing still further the indium content, a distortion of the solvent matrix sets in, creating more "holes."

SUMMARY

Using an oscillating viscometer, accurate measurements on the Hg-In amalgam viscosity have been performed, at various temperatures, ranging from -15°C to 40°C and for different amalgam compositions (3, 15, 30, 45 at percent In).

It has been found that the temperature behaviour of the viscosity is quite similar to the one measured at higher temperature. The viscosity rises smoothly with the increase of the indium content.

The viscosity minimum, for an alloy composition in the neighborhood of 3 atomic percent indium found previously is still present but disappears rapidly at lower temperatures.

The exponential law

$$\eta = c \exp\left(\frac{E_{\eta}}{RT}\right)$$

has been used to fit the data. The values of E_{η} are plotted as a function of q_{In} . It appears that for $q_{In} < 30\%$ the value of E_{η} that fits the data in the lower temperature range ($-15^{\circ}\text{C} \div 40^{\circ}\text{C}$) could fit the data also in the higher temperature range ($30^{\circ}\text{C} \div 130^{\circ}\text{C}$). For $q_{In} > 30\%$ E_{η} becomes appreciably different in the above two temperature ranges.

The experimental results are discussed and some preliminary conclusions are drawn.

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