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# Viscosity of Hg-TI Amalgam

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#### **1** INTRODUCTION

Accurate viscosity measurements about metallic alloys, besides being of great interest for the metallurgical industry, can be considered complementary instruments to X-ray diffraction or to neutron experiments in the structural studies of the liquid.

As summarized by A. R. Ubbelhode,<sup>1</sup> the experimental results, obtained by E. Gebharalt, were the following (p. 238-239):

1) In systems of a complete solubility and in eutectic systems, we found that the viscosity-isotherms depend monotonously on the composition of the alloys at temperatures above the melting point. (Examples: Au-Ag, Cu-Au, Ca-Ag, Al-Zn, Pb-Sn, Pb-Sb, etc.)

2) In systems forming intermetallic compounds in the solid state, we found a maximum in the viscosity-isotherms at the same concentrations where the intermetallic compounds form in the solid state—Figure 1 shows this phenomena in the Mg-Pb system, with corresponding viscosity-isotherms.

This could be interpreted in the sense that the maxima in the viscosityisotherms are due to a pre-freezing phenomena which would indicate the influence of the forces that lead to the formation of the intermetallic compound  $Mg_2Pb$  in the solid state. If the temperature is increased above 1000°C the viscosity becomes a monotonous function of the composition and a maximum is not observed any more. When investigating the systems: Cu-Sn, An-Sn, Mg-Sn, and Mg-Al, we found in a similar way a maximum in the viscosityisotherms at the concentration of an intermetallic compound.

Binary compounds arouse, therefore, particular interest. Among them, besides Hg-In<sup>2,3</sup> Hg-Tl (group IIIb) presents, both from the experimental and from the theoretical point of view, some very attractive features:



FIGURE 1 Phase diagram and viscosity-isotherms of the Mg-Pb system.



FIGURE 2 Phase diagram of the Hg-Tl amalgam.

a) The phase diagram (Figure 2), rather well known<sup>4,5,6</sup> shows in the solid phase intermetallic compounds with a maximum between two eutectics.

b) Special attention was paid to the question of the composition at which the maximum (33.3 atomic percent Tl (Hg<sub>2</sub>Tl) Ref. 4; 28.57 atomic percent Tl (Hg<sub>5</sub>Tl<sub>2</sub>) Ref. 6) is located.<sup>4,5,6</sup>

c) The density versus composition is well known.<sup>7</sup>

d) Accurate viscosity measurements were performed, in the last few years, from room temperature to 160°C.<sup>7,8</sup>

e) A test of the experimental apparatus employed has confirmed its feasibility.

For the above reasons we have performed some accurate measurements on the viscosity of Mercury-Thallium alloys, both as a function of composition and temperature.

In Section 2 of the present paper the experimental apparatus is described briefly. 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. 2. In order to reach  $-20^{\circ}$ C, the viscometer has been placed in contact with a completely different thermostat. In fact, the cylindrical box containing the viscometer, which oscillates in argon atmosphere at  $\sim 300$  mbar, is immersed in a thermostatic bath, whose fluid is a special kind of silicon oil.<sup>9</sup>

The fluid temperature can be regulated continuously from 50°C to  $-20^{\circ}$ C, and the fluctuations are  $\pm 0.25^{\circ}$ C. The temperature of the box is measured by means of a thermocouple, connected to an electronic voltmeter (type Coreci Pep 2.15D) and graphically recorded versus time. To be sure that the viscometer and the box are in thermal equilibrium a convenient time delay (~6 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 accurately constant during each measurement and it is known to within a few tenths of a degree centigrade. As previously, the viscometer oscillations have been recorded using a light beam follower (Photodyne graphispot), and the recordings allowed a determination of the damping coefficient  $\delta$ , with an accuracy<sup>9</sup> of  $\pm 0.5$ %.

Atomic percent Tl content	0%	2%	4.32%	6 % <b>*</b>	6 % <sup>b</sup>	11%	15.86%
<i>E.</i>	0.647	0.693	0,763	0.843	0.823	0.938	1.052
Ċ	0.512	0.512	0.485	0.456	0.471	0.442	0.412
$(1-r^2) \times 10^{-3}$	0.2	5.01	3.6	6.6	3.7	5.6	4.1

TABLE I

<sup>a</sup> Measurements made from 2-2-79 to 8-3-79.

<sup>b</sup> Measurements made from 31-3-79 to 11-5-79.

#### **3 EXPERIMENTAL RESULTS**

The viscometer dampings have been measured for each alloy composition (2; 4.32; 6; 11; 15.86 atomic percent) at different temperatures from 50°C to -20°C.

For each damping, the viscosity  $\eta$  has been calculated using formula (1) of Ref. 10, 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 = 28,346.53 c.g.s. is the total moment of inertia of the system obtained by putting onto the crucible some calibrated disks; T,  $T_0$  are the periods with and without liquid, respectively;  $\delta$ ,  $\delta_0$  are the logarithmic decrements with and without liquid, respectively;  $\rho$  is the density of liquid;  $\eta$  is the viscosity of the liquid; a = 0.348 cm is the inner radius of channel; R = 2.69 cm is the radius of the torus; q is the dimensionless parameter given by:  $a(2\pi\rho/\eta T)^{1/2}$ ;  $G_1$ ,  $G_2$ ,  $G_3$  are universal functions of q, which are given in the paper of Ref. 10.

The experimentally determined logarithmic decrements are introduced in Eq. (1). In order to get q (and from q to get  $\eta$ ) it is necessary, however, to know  $\rho$ .

Data on the density of the Hg/Tl alloy have been obtained from Ref. 7 and conveniently elaborated to get  $\rho$  at each temperature and at each alloy composition.

The applied methods of least square and of linear regression have given  $\rho = 1.450$  gr for ml relative to the 4.32 atomic percent of Tl in agreement with  $\rho_{20} = 1.433$  of Table II of Abowitz and Gordon.<sup>8</sup>

The analysed experimental results are plotted in Figure 3. In Figure 4 the solid lines represent the dependence of  $\eta$  on the alloy composition,  $\eta$  being derived from the exponential best-fit (see Section 4) of the experimental data plotted in Figure 3.



FIGURE 3 Viscosity *n* versus temperature. [■ Viscosity Hg-Tl 6 at. percent Tl by measurements made from 2-2-79 to 8-3-79]. [○ Viscosity Hg-Tl 6 atomic percent Tl by measurements made from 31-3-79 to 11-5-79].

°C	$\eta_1$ (cp)	$\eta_2$ (cp)	$(\eta_1 - \eta_2)$ (cp)
-20	2.44	2.42	0.02
-10	2.29	2.28	0.01
-5	2.22	2.21	0.01
0	2.16	2.15	0.01
5	2.10	2.09	0.01
10	2.04	2.04	-
20	1.94	1.94	-
30	1.85	1.85	-
40	1.77	1.77	_
50	1.70	1.70	-

TABLE II



FIGURE 4 Viscosity  $\eta$  versus amalgam composition.

### 4 DISCUSSION OF THE EXPERIMENTAL RESULTS

The experimental results, collected in Figures 3 and 4 show the following qualitative aspects:

a) The temperature behaviour of the viscosity of 2 atomic percent Tl apparently shows small, smooth oscillations; however, the fluctuations disappear almost completely at different compositions.

b) In Figure 3, the diagrams of 6 atomic percent Tl show two sets of different measurements (performed on the same specimen with an interval of time of  $\sim 1$  month) which overlaps perfectly (see Table II). Besides the value of the viscosity of the 4.32 atomic percent Tl presently obtained at 20°C is in agreement inside the experimental errors with the value obtained in the same conditions in Ref. 8.

c) The viscosity rises smoothly with the increase of the Thallium content. In order to understand the meaning of the experimental result (a), an Arrhenius type law

$$\eta = Ce \frac{E_{\eta}}{R\theta} \tag{2}$$

has been used to fit the data.



FIGURE 5 Plots as a function of the amalgam composition: of the:a) behaviour of C versus  $q_{T1}$ . b) behaviour of  $E_{\eta}$  versus  $q_{T1}$  in the temperature ranges  $-20 \rightarrow 50^{\circ}$ C. c) of the indetermination parameter  $(1-r^2)$ .

The parameters determined by a least squares method are collected in Table I. It must be pointed out that the activation energy of Figure 2 increases almost linearly with the Tl content, differently from the Hg/In amalgam<sup>2,3</sup> in the same composition range. In the second and third rows of Table I, the values of  $E_{\eta}$  (in K cal/mol) and C are given respectively for each amalgam composition. In the fourth row the corresponding values of  $1 - r^2$  are



FIGURE 6 Deviations  $\ln(\bar{\eta}/\eta)$  of the experimental viscosity from the exponential law versus  $1/R\theta$  for each amalgam composition.

reported where  $r^2$  is the coefficient determination,<sup>11</sup> which lies between 0 and 1, and is a measure of the goodness of the exponential regression: the more  $r^2 \rightarrow 1$ , the more the experimental points approach the exponential law. The above data are also plotted in Figure 5.

It must be pointed out that Eq. (2) fits the experimental data much better than for the Hg/In amalgam. The deviations from the best fit are plotted in Figure 6.

d) Small undulations in Figure 4 are superimposed on a linear behaviour, obeying the law, valid for a simple two-component mixture.<sup>12</sup>

$$\eta = \eta_{\mathrm{TI}} q_{\mathrm{TI}} + \eta_{\mathrm{Hg}} q_{\mathrm{Hg}} \tag{3}$$

where  $\eta_{\text{Hg}}(\eta_{\text{Tl}})$  is the viscosity of Mercury (Thallium) at the same temperature and  $q_{\text{Hg}}(q_{\text{Tl}})$  is the Hg(Tl) atomic fraction. According to what has been pointed out in the case of Hg/In amalgam, there are compositions which show a viscosity greater than predicted by relation (3), which can be attributed to the interatomic bonds. The writer intends to perform, in the near future, measurements on the same amalgam near the 30% atomic composition at which an intermetallic compound appears in Figure 2.

#### SUMMARY ·

Using an oscillating viscometer, accurate measurements on the Hg-Tl amalgam viscosity have been performed, at various temperatures, ranging from  $-20^{\circ}$ C to  $50^{\circ}$ C and for different amalgam compositions (2; 4.32; 6; 11; 15. 86 atomic percent Tl).

It has been found that the temperature behaviour of the viscosity have only very small, smooth oscillations. The viscosity rises smoothly with the increase of the thallium content. The theoretical and experimental method is appropriate for making accurate viscosity measurements at the melting point.

The exponential law in Eq. (2) has been used to fit the data. The fit is much better than for the Hg/In system.

The viscosity calculation of 6 atomic percent of Tl, at different times, has demonstrated the reproducibility of the data.

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