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AN ULTRASONIC STUDY OF LIQUID INDIUM-CADMIUM ALLOYS

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A number of workers, notably Turner et al (1973) and Maier and Steeb (1973), have shown that the compressibility of a liquid metal alloy is affected by the molecular clustering which persists in some melts. Activity measurements, Iwa, Joasoo and Blairs of liquid indium-cadmium alloys show large negative departures from ideality for melts of approximately 75% cadmium content. These measurements, esentially of the vapour pressure of cadmium atoms above the melt, show that for this concentration the atoms are more tightly bound to the liquid than expected and indicate the presence of a special bonding condition. Anomalies are also found in the electrical resistivity of indium-cadmium alloys of the same concentration, Predel and Berka (1976). They suggest that the intermetallic compound $InCd_3$, present in the solid state, is present in the form of $InCd_3$ molecular clusters in the liquid state. We have now made measurements of the sound velocities of a range of In-Cd alloys, at temperature up to 700°C, as a further investigation of clustering in this system.

Numerous workers have made ultrasonic studies of molten indium and cadmium. They have shown the sound velocity in indium to exceed that in cadmium by no more than 3%. As the sound velocities in the alloys of indium and cadmium would be expected to fall between those of the constitutents, it was necessary to measure them with an accuracy of at least 0.1%.

Satisfactory measurements were made by the adoption of the pulse-echo overlap technique, Papadakis (1967), which has proved so successful in studies of sound velocities in solids. The apparatus used for these measurements is shown schematically in Fig. 1. A 15cm long vitreous silica delay rod D was used to separate the transducer T from the hot molten metal. To minimise the deleterious effects of the long delay rod, measurements were made at the unusually high frequency of 50MHz. The pulse-echo overlap technique was used to measure the time interval between the first echo from the end of the delay rod and the echo from the stainless steel reflector R immersed in the melt. The position of the crucible, containing the liquid metal sample S was changed, during each sound velocity measurement, using a motorised micrometer. Typically, the acoustic path was changed by a total of 1cm and time interval measurements were made for each m.m. change. The velocity of sound in the liquid metal was obtained from a linear regression analysis of the 11 data pairs obtained. The micrometer was adjusted continually at a rate of 0.25mm/min so that the same cycle-for-cycle overlap of the RF bursts, comprising the two echoes, was maintained at each setting of the crucible. The magnitude of the echoes from the melts was limited by the extent of the oxide film at the delay rod-liquid metal interface. This was minimized most effectively by conducting the experiments in a reducing atmosphere of hydrogen.

The absolute accuracy of measurement achievable using the apparatus was assessed by measurements of the sound velocity in mercury at room temperature. A value of 1452 ± 0.13ms^{-1} at 20°C was obtained which is in good agreement

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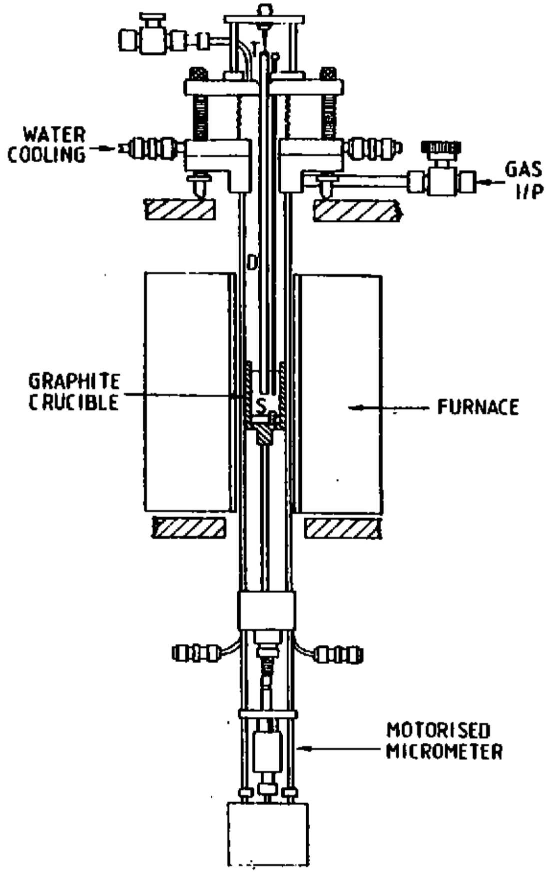

Fig. 1. Schematic diagram of apparatus used to make sound velocity measurements in liquid metals.

with the accepted figure of $1451 \pm 0.3 ms^{-1}$ obtained by Hubbard & Loomis (1928).

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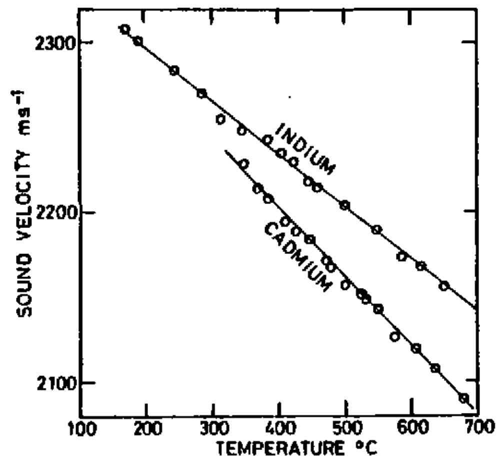

Fig.2. Measurements of sound velocities in pure liquid indium and cadmium.

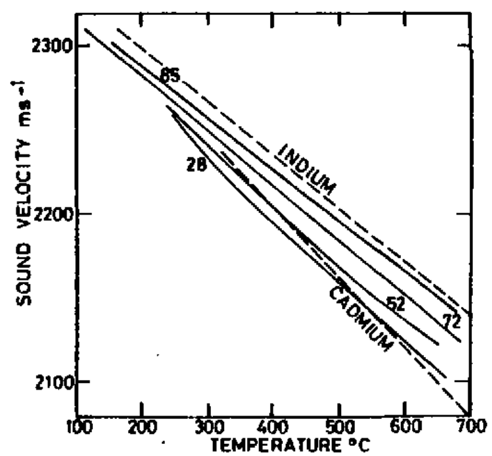

Fig. 3. Measurements of sound velocities in liquid alloys of 28, 52, 72 and 85 weight per cent indium in cadmium. Sound velocities in pure indium and cadmium are indicated by the broken lines.

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Measurements of pure cadmium and indium are shown in Fig. 2. The lines fitted through the data are:

$$U(Cd)ms^{-1} = (2472 \pm 4) - (0.404 \pm 0.005)(T/K)$$

$$U(In)ms^{-1} = (2445 \pm 3) - (0.314 \pm 0.006)(T/K)$$

The increased uncertainty in sound velocity at high temperature has been traced to the thermal instability of the melts. A thermal drift of 1K during the measurement period was found lead to an error, by the differential thermal expansion of the components of the apparatus, of 2.5μ in the total displacement of the crucible by the micrometer. The temperature of the melt was maintained to about ±1K of the required values during the measurement periods. This amount of thermal instability introduced an average uncertainty to our measurements of ~0.1%.

The velocities of sound in alloys of indium and cadmium are shown in Fig. 3. To avoid confusion, the lines fitted through the data, rather than data points, are presented. It is evident that the alloys of higher cadmium content deviate greatly from the rule of mixtures. Similar negative departures from ideality were found by Maier and Steeb for Mg-Sn, Al-Sn and Fe-C alloys of compositions which are known to show significant molecular clustering in the liquid state. The range of compositions for the departures from ideality in the indium-cadmium alloys coincides with the large activity anomalies found by Iwa, Joasoo & Blairs. The reduction in sound velocity for melts containing inhomogeneities, in the form of molecular clusters, is predicted by the Landau relationship between compressibility K and the fluctuation in atomic volume ΔV

$$\overline{\Delta V^2} = kTVK$$

where V is volume, T temperature and k Boltzmann's constant.

On the completion of a series of measurements of the densities of the indium-cadmium alloys it will be possible to estimate their isothermal compressibilities, using the sound velocity measurements presented here. These compressibilities will then be used, with the activity measurements, to complete a partial structure factor analysis of the In-Cd system of the type obtained for the Cu-Sn alloys by Turner et al.

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