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ULTRASONIC VELOCITY IN Ga-Pb LIQUID ALLOYS[1]

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Ultrasonic velocity V_S in Ga-Pb liquid alloys at frequency of 30.3MHz has been measured in the range from monotectic temperature up to 1100 K using a pulse phase technique with varying the acoustic path length. It was stated that curves V_S(T) are linear for all the investigated alloys above the phase separation temperature. The temperature coefficients dV_S/dT are negative. The liquid coexistence curve on the phase diagram of the Ga-Pb system is determined on the basis of the V_S(T) measurements.

1. Introduction

The immiscible liquid metal systems seems to be interesting for the investigation of the nature of phase transitions and critical phenomena. However, investigations of these systems were not very numerous up to now because it was difficult to establish the immiscibility region for a fusible alloys. Recently, the possibility of determination of the immiscibility region on the basis of the V_S(T) measurements using a pulse phase technique with varying the acoustic path length [1] has been pointed out.

In this paper, the investigation of the temperature dependences of the ultrasonic velocity V_S(T) for Ga-Pb liquid alloys and determination of the position of the liquid-liquid coexistence curve with help of these data are presented.

2. Experimental Method

The ultrasonic velocity measurements have been carried out by means of a pulse phase technique. This technique is based on the electric interference of coherent waves transmitted through the measuring cell with melt and the reference cell filled by water. The cells are connected in parallel. The measuring cell is made in form of a quartz vertical container and is attached to the lower buffer rod. The upper buffer rod can be moved

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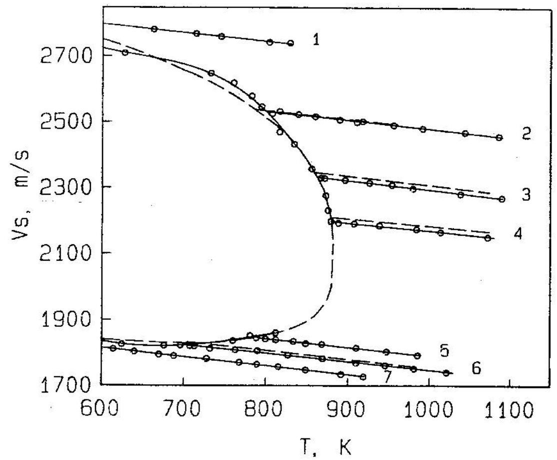

Fig.1. Temperature dependences of the ultrasonic velocity V_S for $Ga_{100-x}Pb_x$ liquid alloys: (1) x=0, (2) 10, (3) 20, (4) 30, (5) 80, (6) 90, (7) 100 at.%. o, present results: - - -, results of Regel et al. [1]. The cupola-like curves correspond to the ultrasonic velocity along the binodal.

by means of a micrometer screw. The buffer rods are made of a clear-fused quartz and are placed along container vertical axis. The construction of the reference cell is analogeous.

When the upper buffer rod of the measuring cell is shifted, a sum signal transmitted through both cells traces out maxima and minima of interference pattern. Completing the information obtained from the minima separation L which are equal to integer multiples of the wavelength (nλ) with the knowledge of the carrier-wave frequency f, the velocity may be computed as $V_S = \frac{L}{n} f$. This velocity refers to the region of the melt between the initial and final positions of the upper buffer rod. Since this distance L, which is of the order of 2-3 mm, is known to be within ±0.005 mm from measurements, the velocity can be determined with accuracy of $\pm0.2\%$. From the measurement of the ultrasonic velocity V_S at different heights H from the lower rod, the dependence of V_S versus H can be determined.

3. Results and Discussion

The ultrasonic velocity measurements were carried out for liquid Ga-Pb mixtures in both Ga-rich and Pb-rich concentration ranges of the miscibility gap. The temperature dependences of the ultrasonic velocity V_S(T) for the investigated alloys are shown in Fig. 1. The data obtained for liquid gallium and lead are also presented for comparison. The ultrasonic velocity in the alloys increases linearly with temperature lowering until the phase separation temperature $T_S(x)$, where gravity separates the melt into two liquid layers. The linear temperature dependence of V_S in liquid Ga-Pb alloys above the phase separation temperature demonstrates a typically metallic behaviour. On cooling below the $T_S(x)$ point the V_S(T) curve diverges into two branches: the lower branch describes the ultrasonic velocity of the Pb-rich phase while the upper branch describes V_S(T) of the Ga-rich phase.

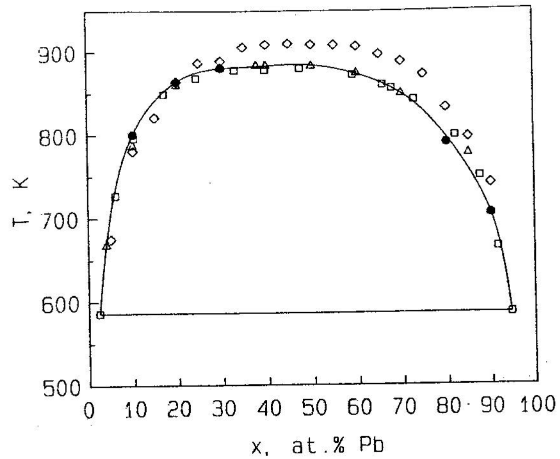

Fig.2. Liquid-liquid coexistence curve of the Ga-Pb system. •, present work; △, Regel et al. [1]; □, Predel [2];◇, Sokolovskii et al. [3]. The binodal is plotted by using results of the present work and data of [1,2].

Our values of ultrasonic velocity V_S are slightly lower than the data of Regel et al. [1] obtained at a frequency of 7 MHz also using a pulse phase technique.

In order to determine the position of the liquid-liquid coexistence curve on the phase diagram of the Ga-Pb system, the ultrasonic velocity was measured along vertical axis. When the alloy splits up and two liquid layers are present, a jump of V_S forms on the phase boundary. Measuring curves V_S(H) at various temperatures we observed an appearance of the phase boundary.

The phase separation curve of the liquid Ga-Pb system determined on the basis of the V_S(T) measurements is shown in Fig. 2. It is in good agreement with the earlier studies [1-3], except for the Ga-80 at.% in the Ga–Pb alloy for which the phase separation temperature is sufficiently lower than in [3].

After heating the separated melt up to the temperature just above the immiscibility region or mixing of the components at the same temperature we observe that ultrasonic velocity changes greatly along the vertical axis. The inhomogenity of the medium examined by the ultrasonic velocity, vanishes after mechanical stirring of the sample, after heating the melt up to a specific temperature or after isothermal exposition ~ 10 hours slightly above the phase separation temperature. We have found that the influence of the mechanical stirring on homogenization process is more efficient compared to the temperature and time influences. The thermal and time stabilities of this inhomogeneity of V_S increase when the alloy concentration approaches the critical point.

4. Conclusions

'he ultrasonic velocity in Ga-Pb liquid alloys has been measured at a frequency of 0.3 MHz. Using the pulse phase technique with varying the acoustic path length it is ossible to determine the position of the liquid-liquid coexistence curve on the phase iagrams. It was stated that the inhomogeneity of the ultrasonic velocity along vertical xis above the immiscibility region remains for a long time. Correct investigations of iquid immiscible alloys throughout the region of the miscibility gap require the melt to e preliminarily homogenized. Mechanical stirring and the temperature-time exposition an be used for this purpose.

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