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Heats of Formation of Solid Indium-Lead Alloys

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Abstract

Heats of formation of solid indium-lead alloys at 315°K were determined by liquid tin solution calorimetry. The alloys were found to form endothermically, except, possibly, the (In) phase, where $\Delta H \sim 0$. Partial molar heats of solution of indium and lead in tin were also determined.

Introduction

Indium and lead form wide-ranging solid solutions in one another, with a single intermediate phase, β, as shown in Figure 1.[1] The crystal structures of all three solid phases are based on the face-centered cubic structure of Pb, but for (In) the c axis is elongated about 7.6%, and for β, c is shortened by about 7%, making the two latter phases face-centered tetragonal.[6]

The thermodynamic properties of the liquid phase are reasonably well established, but solid heats of formation have been determined only by a quantitative differential thermal analysis method[2] which does not have high precision. It was therefore decided to measure heats of formation by liquid tin solution calorimetry.

Experimental

The tin, lead, and indium used were all reported to be 99.999% pure.

Nine 12-gram samples of indium-lead alloys were prepared. Weighed quantities of metal were mixed and sealed in borosilicate glass tubes containing an atmosphere of helium plus 4% hydrogen. Melting was done approximately 20° higher than the liquidus temperature. After melting, the alloys were shaken vigorously and quenched in ice water. No loss in weight during melting was found. After cold working, the alloys were again sealed in borosilicate glass tubes and homogenized for seven days at 10° to 15°C below the solidus. Filings were taken from both ends of each ingot, mixed together, and strain annealed at 100°C for 15 minutes. X-ray diffraction showed the phases to be homogeneous and to have lattice constants corresponding to their compositions.

The liquid tin solution calorimeter has been described previously,[5] so a very brief description of the method will suffice. About 500 mg of alloy are dropped into the calorimeter containing about 200 gm of Sn at about 650°K. From the measured heat effect is subtracted the heat effects of corresponding amounts of the pure components. The result is the heat of formation of the alloy at the temperature from which it is dropped; in this case 315°K.

Three to five runs per day are made in the calorimeter. After four days, the calorimeter is dismantled and a fresh tin bath added. This is allowed to come to a steady state in temperature over the weekend and a second series begun. Each day one of the samples was pure tin, to calibrate the heat capacity of the calorimeter. A small increase of heat capacity as the amount of liquid metal increases proves the calorimeter is functioning

properly. Near the beginning of a series, samples of pure Pb and pure In are dropped; also near the end, when the amount of dissolved material is about 1.25 atomic percent. The heat effects, within experimental error, were independent of concentration.

Subtracting the known heat contents of the pure liquid metals from their heat effects when dissolving in tin, one obtains the partial molar heats of solution at 650°K:

$$\Delta\bar{H}_{Pb} = 1402,\ 1410,\ 1419,\ 1420,\ 1406,\ 1393,\ 1436,\ 1391$$

$$\Delta\bar{H}_{In} = -188,\ -176,\ -149,\ -160,\ -171,\ -172,\ -163,\ -161$$

From which we obtain:

$$\Delta\bar{H}_{Pb,\,650°K,\,x_{Sn}=1} = 1410\ (\pm 11)\ \text{cal/g-atom}$$

$$\Delta\bar{H}_{In,\,650°K,\,x_{Sn}=1} = -168\ (\pm 8)\ \text{cal/g-atom}$$

The uncertainties are twice the standard deviation of the mean. These values compare with 1360[3] and -203[4] cal/g-atom, respectively, found in the literature at near the same temperature.

The experimental values of the heats of formation are given in Table 1 and are plotted in Figure 2 along with the values of Heumann and Predel[2] calculated from liquid heats of formation and the heats of fusion of the alloys measured by differential thermal analysis. The agreement is relatively good.

The present values are to be preferred, since liquid tin solution calorimetry is known to be a more accurate method.

Acknowledgment

Stanley Ross gave valuable assistance in the measurements.

TABLE 1

Experimental Values of Heats of Formation at 650°K

$$(1-x)In_{(s)} + xPb_{(s)} = In_{1-x}Pb_{x(s)}$$

x_{Pb}	Phase	ΔH, experimental	ΔH, selected*
0.10	(In)	43, -18, -5, -42	-0(±36)
0.20	β	191, 199, 154, 188	190(±12)
0.30	β	279, 257, 270	270(±12)
0.40	(Pb)	306	300(±12)
0.50	(Pb)	321, 296, 303	310(±12)
0.60	(Pb)	275	280(±12)
0.70	(Pb)	251, 241	250(±12)
0.80	(Pb)	182, 193	190(±12)
0.90	(Pb)	63, 41, 107, 91	100(±12)

*The selected values were chosen from a plot of ΔH versus x_{Pb}. The uncertainties are twice the standard deviation from the curve.

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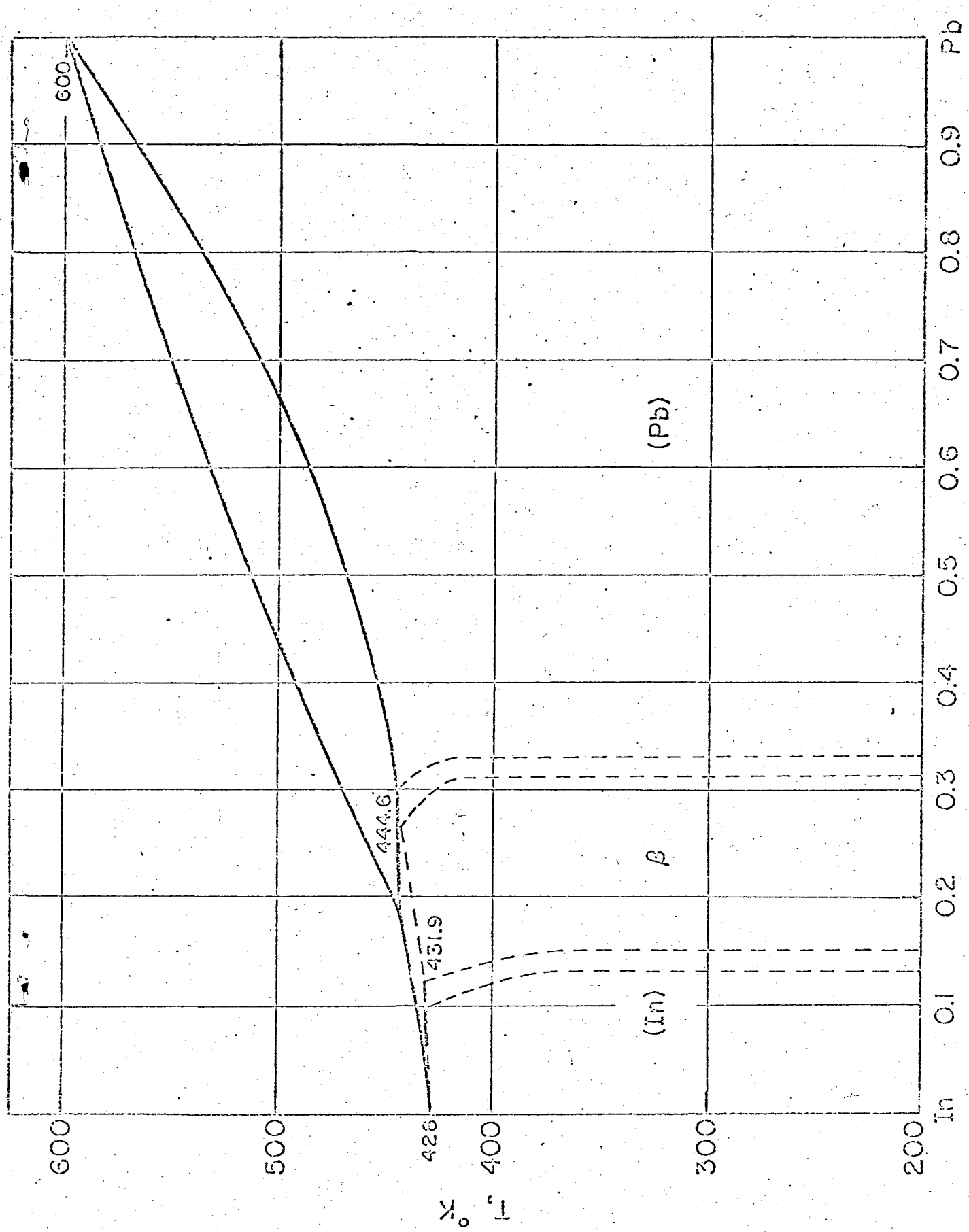

FIG. 1 PHASE DIAGRAM FOR In-Pb SYSTEM

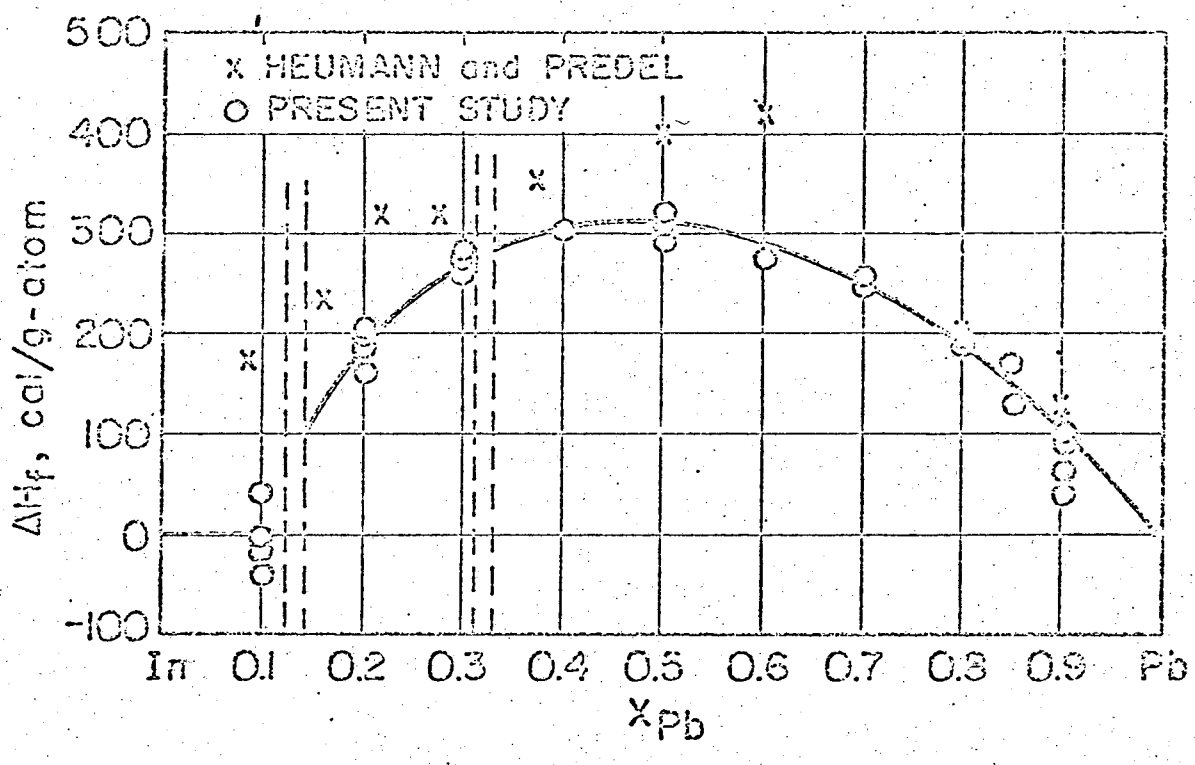

FIG. 2 HEATS OF FORMATION OF In - Pb ALLOYS AT 315°K

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