LETTERS TO THE EDITOR

The Density of Dilute Solutions of Copper in Liquid Lead*

In a previous investigation (*1*) the concept of interstitiality in liquid metal solutions (*2*) was tested via precision density measurements on dilute lead-nickel solutions. It was found that the partial molal volume[1] of nickel decreased with decrease in nickel content and approached a value of zero at high dilution. These results were shown to be consistent with a proposed solution process of the nickel in the molten lead (*1*). As a continuation of this work, dilute lead-copper solutions were investigated in a similar manner, and the present communication gives the results obtained.

The experimental procedure used to measure the densities of lead-copper solutions was the same as that described in a previous paper, and the lead and copper used were of chemical purity comparable to that of the lead and nickel used in the previous investigation (*1*).

TABLE I

DENSITY FOR LIQUID LEAD-COPPER SOLUTIONS

At % (Cu)	Temp (°C)	Density (g/cc)	At % (Cu)	Temp (°C)	Density (g/cc)	At % (Cu)	Temp (°C)	Density (g/cc)	At % (Cu)	Temp (°C)	Density (g/cc)
0.352	651.3	10.273		758.0	10.150		651.5	10.268		750.8	10.158
	652.0	10.270		758.8	10.150		653.0	10.266		752.7	10.155
	652.1	10.272		797.6	10.103		656.7	10.268		793.8	10.107
	657.1	10.268		801.0	10.104		702.1	10.215		796.0	10.102
	696.2	10.218		803.1	10.101		702.2	10.212		797.2	10.106
	697.4	10.214		803.5	10.098		703.2	10.211		798.1	10.106
	698.5	10.217	0.972	650.7	10.274		703.8	10.210		798.4	10.101
	703.2	10.216		651.0	10.270		749.4	10.160	3.444	650.0	10.266
	704.6	10.211		651.2	10.276		750.9	10.155		650.8	10.263
	747.6	10.158		654.5	10.268		752.8	10.158		651.8	10.267
	747.8	10.159		697.1	10.216		755.3	10.156		653.2	10.262
	749.8	10.158		698.4	10.220		794.6	10.110		656.6	10.259
	751.0	10.156		702.3	10.214		799.1	10.105		696.9	10.214
	752.3	10.158		702.5	10.212		800.6	10.104		697.3	10.217
	793.9	10.101		703.7	10.209		800.8	10.101		698.0	10.210
	795.8	10.108		745.4	10.164	2.600	644.0	10.275		699.7	10.212
	796.2	10.105		749.2	10.160		645.5	10.277		702.8	10.208
	797.8	10.103		750.8	10.158		646.2	10.271		703.2	10.205
	799.2	10.097		751.7	10.158		648.8	10.268		747.4	10.156
0.702	650.5	10.275		752.3	10.155		649.7	10.272		749.3	10.154
	653.6	10.271		752.5	10.151		698.3	10.216		751.0	10.157
	653.8	10.272		790.1	10.110		698.7	10.215		752.7	10.150
	653.8	10.274		794.2	10.105		699.2	10.218		753.5	10.151
	701.5	10.216		795.0	10.108		700.3	10.214		792.6	10.105
	703.2	10.213		796.6	10.109		700.6	10.212		795.8	10.101
	703.7	10.210		800.8	10.100		701.8	10.208		796.5	10.104
	704.8	10.214		801.3	10.097		745.9	10.163		797.4	10.100
	751.1	10.162	1.572	649.2	10.271		746.7	10.159		799.1	10.102
	753.9	10.156		650.2	10.273		749.8	10.162		799.4	10.108

Table I gives density data for the liquid lead-copper solutions that were obtained in the present investigation. It was found that, over the composition region investigated, the molal volumes of lead-copper solutions that were calculated from the experimentally determined densities were

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[1] The partial molal volume of nickel is the rate of change in volume produced by the addition of nickel to the lead-nickel solution at constant pressure, temperature, and concentration.

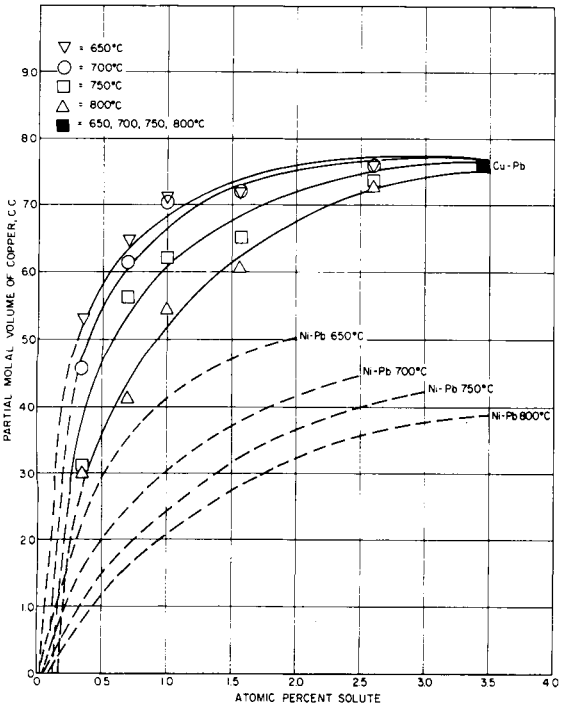

FIG. 1. Partial molal volume-composition isotherms for copper in liquid lead-copper and for nickel in liquid lead-nickel solutions.

ess than those calculated from the rule of mixtures even though less dense copper was added to the liquid lead.[2]

Partial molal volumes for lead and copper were estimated by the method of intercepts (*1*, *4*). It was found that, as in the case of the lead-nickel solutions, the partial molal volume of lead in the lead-copper solutions did not differ by more than about 0.2% from the molal volume of lead. Figure 1 gives partial molal volume-composition isotherms for copper in the lead-copper solutions, and similar curves for nickel in lead-nickel solutions are included for comparative purposes. The data indicate that the same general trend is observed for both systems; the partial molal volume of the solute extrapolates to a value of zero at high dilution, decreases with increase in temperature at constant concentration of solute, and increases with increase in solute content. These observations, in addition to the fact that the metallic radii of the solutes, their electronegativities, as well as their temperature coefficients of solubility in liquid lead (*2*) are comparable, suggests that the model used as a basis to explain the composition dependence of the partial molal volume of nickel (*1*) should be equally applicable to explain that of copper. Briefly, in each of these binary systems, the radius ratio of solute to solvent (*5*) is not far from Hägg's atomic radius ratio of 0.59 for pairs of atoms the smaller of which should fit into interstitial positions in the lattice of the larger. Furthermore, since liquid metal solutions may display a distribution in the sizes of interstitial holes (*6*), one can visualize that at high dilution the relatively small solute atoms (copper or nickel) enter interstitial sites in the quasi-lattice of the molten lead with the result that the partial molal volume of the solute approaches a value of zero. As more solute is dissolved, the partial molal volume increases with increase in composition because some of the solute atoms must enter substitutional sites (*1*). It is to be noted in Fig. 1 that the partial molal volume of copper increases more rapidly with composition than that of nickel. This is still consistent with the proposed model for the solution process (since the atomic volume of copper is about 5% larger than that of nickel) and further emphasizes the importance of the atomic size effect in liquid alloys.

[2] In the absence of experimental data on the densities of liquid copper undercooled to temperatures as low as 650°C, the densities used in the rule of mixtures calculations of the molal volumes of copper were obtained by extrapolation on a density-temperature plot using Lang's (*3*) density data.

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Reactivity Effect of (*n*, 2*n*) Reactions in D_2O*

Recent measurements (*1*) of the (*n*, 2*n*) cross section of deuterium by Hill, Goldberg, LeBlanc, and Taylor at the Lawrence Radiation Laboratory give results in good agreement with theory (*2*) and permit reasonably accurate calculations to be made of the effect of this reaction on the multiplication factor of heavy water moderated reactors. Multigroup calculations were made for an infinite sea of D_2O containing a uniformly distributed source of fission neutrons to determine not only the magnitude of the (*n*, 2*n*) effect, but also the effect of (*n*, α) and (*n*, *p*) reactions with oxygen. The effects were found approximately to cancel with a net loss of neutrons per fission neutron of 0.06%.

In the multigroup calculation, a group width (common to all groups) of 0.1 lethargy unit was employed where the

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