

The Cu-Pb (Copper-Lead) System

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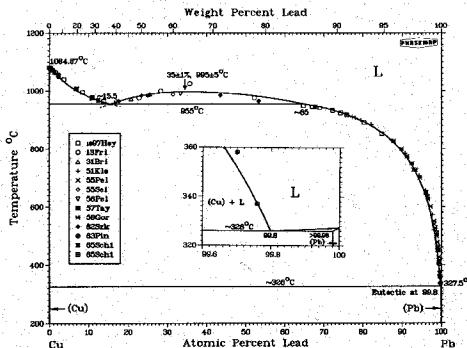
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Equilibrium Diagram

The equilibrium phases in the Cu-Pb system, shown in Fig. 1, are: (1) the liquid that is miscible in all proportions at higher temperatures, but develops a miscibility gap below about 1000 °C; (2) the fcc solid solution (Cu), with very limited solubility of Pb; and (3) the fcc solid solution (Pb), with presumably negligible solubility of Cu.

Liquidus. The liquidus in the Cu-Pb system has been studied extensively, and the results, for the most part, are in good accord (see selected data in Table 1). However, the results for the miscibility gap, including the critical point and the monotectic invariant compositions, show large scatter, so that the accepted results for these must be taken as tentative (see Table 2).

Fig. 1 Assessed Cu-Pb Phase Diagram with Selected Experimental Data



D. J. Chakrabarti and D. E. Laughlin, 1984.

The Cu-rich end of the liquidus was determined by Heycock and Neville [1897Hey] by thermal analysis. Although the freezing point of Cu reported by them was quite low (1080.8 °C), as compared to the accepted value of 1084.87 °C [81BAP], the liquidus data are still in good agreement with those of [65Sch1] and [56Pel]. Both authors used the technique of sampling from the liquid, which in this instance consisted of Cu-rich liquid in equilibrium with the (Cu) phase and supersaturated with Pb. The samples obtained at various temperatures were subsequently chemically analyzed for the compositions of the liquidus.

The liquidus on the Pb-rich side beyond the miscibility gap has an increasingly steep downward slope that becomes nearly vertical near pure Pb, where it forms a eutectic. [1897Hey] determined this part of the liquidus by thermal analysis, but the measurements could not be extended above 85 at. % Pb, corresponding approximately to 858 °C, as this method failed to detect the increasingly weak thermal arrest at higher Pb contents. Kleppa and Weil [51Kle], followed by [55Pel], [57Tay], [58Gor] and [63Pin], used the more sensitive technique of sampling (from the liquid) and determined the entire Pb-rich portion of the liquidus in good agreement with each other (see Table 1). In this sampling method, Cu was added to Pb and was allowed to saturate by partial dissolution in the molten Pb at various temperatures. The composition of the sampled liquid was determined at each temperature to define the liquidus. The data of [51Kle] overlap the temperature range of the data of [1897Hey], in good agreement with the latter. The data of [51Kle] also agree with the limited data of [65Sch1] above 906 °C, also obtained by the sampling method. Thus, the liquidus up to ~15 at. % and above ~65 at. % Pb have been fairly well established.

Heycock and Neville [1892Hey] claimed that the depression in the melting point of Pb on adding Cu reached a maximum of 1.2 °C at 0.195 at. % Cu. Subsequent studies confirmed the occurrence of a eutectic transformation at the composition indicated by [1892Hey]. The corresponding transformation temperature, as derived from the data of [1892Hey], is not correct, however, as the melting point of Pb reported by them was 326.5 °C, as compared to the presently accepted value of 327.502 °C [81BAP]. Both [1897Hey] and [13Bor] obtained thermal arrests at 326 °C and estimated the eutectic composition to be less than 0.2 and 0.18 at. % Cu, respectively, in reasonable agreement with [1892Hey]. Pelzel [55Pel] reported the eutectic temperature to be at 325.5 °C. The eutectic composition reported by him was, however, too high (0.36 at. % Cu), compared to all the other results. [51Kle] obtained 0.2 at. % Cu for the eutectic composition from the extrapolation of the liquidus to the eutectic temperature, assumed at 326 °C. Based on the literature data (Table 2), the accepted eutectic temperature and composition values have been established at about 326 °C and 0.2 at. % Cu, respectively.

Miscibility Gap. A miscibility gap is formed in the Cu-Pb liquid near the midcomposition range. In spite of several studies, the boundaries of this gap have not been well established. This is shown in Table 2, along with the reported purity of the materials used in these works. The extremely high critical temperature (>1500 °C), reported by [13Bor] from the electrical resistivity measurements, is apparently not correct, as all subsequent studies have shown. Most of the other studies [13Fr1, 15Bog, 31Bri, 62Szk] were based on the sampling method, in which a liquid of known composition was formed by complete dissolution of Cu in liquid Pb. This liquid was subsequently equilibrated in the two-phase region at successively lower

Table 1 Selected Liquidus Data in the Cu-Pb System

Reference	Temperature, °C	Composition, at.% Pb	Experimental method	Reference	Temperature, °C	Composition, at.% Pb	Experimental method			
[1897Hey]	1080.8	0	Thermal analysis	[55Pel]	635	96.56	Chemical analysis of liquid			
	1079.3	0.15			540	98.22				
	1077.1	0.32			500	98.80				
	1071.6	0.78			450	99.09	DTA			
	1062.8	1.55			370	99.53				
	1055.1	2.26			988	31.5				
	1039.2	3.70			984	23.5				
	995	8.42			965	17				
	977.6	10.92			[56Pel]	1008	7.1	Microscopy and chemical analysis of liquid		
	963.6	13.86				990	~33.35(b)			
	948.8	64.7				[57Thy]	510	98.84	Chemical analysis of liquid	
	945.1	67.05					460	99.16		
	941.1	68.94					410	99.48		
	932.2	72.20					[58Gor]	826.8	87.82	Chemical analysis of liquid
	924.0	74.30						800	89.90	
	917.0	75.90						774	91.32	
	890.8	80.30						750	92.47	
851.8	84.80	726.8	93.43							
326.0	98.03(a)	700	94.54							
327.3	100(a)	650	96.10							
[13Fri]	1025	35.8, ...	626.8	96.62				Chemical analysis of liquid		
1000	28.4, ...	600	97.27							
975	22.8, 52.2	550	98.16							
[31Bri]	989	25.6	526.8	98.45						
986	25.2	500	98.80							
972	20.8	478	99.0							
[51Kle]	920.2	75.40	450	99.23						
918.7	75.81	426.8	99.38							
910.3	77.30	[62Szk]	985	25.2, 43.5	Chemical analysis of liquid					
907.5	78.05		965	17.7, 53.3						
897.8	79.40		[63Pin]	403	99.45	Chemical analysis of liquid				
883	82.10			358	99.70					
825.9	87.72			337	99.76					
783.9	90.82			[65Sch1]	1065	1.2	Chemical analysis of liquid and DTA			
746.2	92.5				1050	2.4				
726.9	93.06				1008	6.8				
707.2	94.42				975	10.8				
656.2	95.94				968	12.4				
641.8	96.14				945	67.6				
640.9	96.29				DTA data	1003	36.6, ...			
572.9	97.74					998	27.8, ...			
520	98.19					981	20.6, ...			
505.7	98.75					964	..., 60.8			
378	99.65									
360.2	99.69									
[55Pel]	727	92.88						Chemical analysis of liquid		

NOTE: Temperatures shown for the experimental data are as reported and have not been corrected to the 1968 temperature scale (IPTS-68). Accepted results are taken from [51Kle], [58Gor], and from data of other authors shown in boldface type.

(a) Solidus data. (b) Estimated from microscopy work; result differs from that by chemical analysis of liquid.

temperatures, and the corresponding compositions of the liquid were determined. The data for the critical point from these studies vary from 975 to 1025 °C and from ~31.5 to 36.5 at.% Pb.

The critical temperature (1025 °C), reported by Friedrich and Waelhert [13Fri], is perhaps high. Both Briesemeister [31Bri] and Szkoda [62Szk], as well as the thermal analysis [65Sch1] and thermodynamic calculations [65Sch2] data by Schurmann and Kaune, reported it to be around 1000 °C. DTA measurements by Seith, Jönen, and Wagner [55Sei] and optical microscopy by Pelzel [56Pel] gave a value of 990 °C. A sensitive method, employed by [67Jac], consisted of measuring the temperature at which an

abrupt change in the pressure of air bubbling through the Cu-Pb liquid occurred because of the formation of layers (with different densities) in the liquid. Using this method, a critical temperature of 980 ± 5 °C was determined for the bath composition of 34.3 at.% Pb. The actual temperature could be somewhat higher, considering the possibility of some undercooling effect during the bubbling experiment. Thermodynamic calculations by [80Tym], based on their vapor pressure data, gave a critical temperature of 983 °C. The weight of evidence from the above works suggests that the critical temperature lies somewhere between 990 and 1000 °C; therefore, the accepted temperature has been established at 995 ± 5 °C. The suggested critical temperature between 1100 and 1200 °C by

Table 2 Compositions and Temperatures at Invariant Points in the Cu-Pb System

Reference	Monotectic		Eutectic		Critical		Method/comment
	Temperature, °C	Composition, at.% Pb	Temperature, °C	Composition, at.% Pb	Temperature, °C	Composition, at.% Pb	
[1892Hey]	1.2 below melting point of Pb	0.195	Freezing point depression(a)
[.897Hey]	954	17	65	326	<0.2	...	Thermal analysis(b)
[13Bor]	955 to 956	14.7	79.5	326	0.18	>1500	Electrical resistivity(c)
[13Fri]	953	21	57	1025	35.8 Chemical analysis(d)
[15Bog]	940	14	67	975	...
[31Bri]	...	16 to 17(f)	79(f)	1001	36.5 Chemical analysis(d)
[33Nis]	957	25	67	(g)
[51Kle]	67(h)	...	0.2(j)	...	Saturation solubility(k)
[52Bis]	Electrical resistivity(m)
[55Pel]	325.5	0.36	...	Saturation solubility
[55Sei]	...	15(f)	65(f)	990	34 DTA(n)
[56Pel]	955 ± 0.5	16(p)	64	990	33.4 Optical microscopy(p)
[58Abd]	1100 < T < 1200	...
[62Szk]	1000	31.5 Chemical analysis(d)
[65Sch1]	1002	35.5 Thermal analysis
[65Sch2]	952	14.9	64.6	1004	35.5 Thermodynamic calculations(q)
[67Jac]	980 ± 5	...
[80Tim]	...	16.3	65.3	983	36.3 Thermodynamic calculations(s)
[82Esd]	1054	36.4 Thermodynamic calculations
Accepted (this evaluation)	955	15.5 ± 0.5	65	326	0.2	995 ± 5	35 ± 1 See text for explanations

(a) Melting point of Pb reported by the authors is 326.5 °C. (b) Melting point reported by the authors is 327.3 °C. (c) Quoted in [31Bri]. (d) Sampling from two-melt layers held in equilibrium. (e) Quenching of liquid droplets from different temperatures; 99.97% Cu and 99.5% Pb starting material. (f) Estimated values from extrapolation of liquidus to $T_{\text{mon}} = 954$ °C. (g) Quoted in [56Pel]. (h) Value from extrapolation of liquidus to 954 °C. (j) Value from extrapolation of liquidus to 326 °C. (k) Starting material compositions: 99.8% Cu and 99.997% Pb. (m) Boundary of miscibility gap was found to lie between 1007 and 994 °C for liquid alloy of 36 at.% Pb. (n) Starting material compositions: electrolytic Cu and 99.99% Pb. (p) Chemical analysis of the liquid samplings indicate a higher Pb content (20.4 at.%) at monotectic point than that estimated by microscopy. (q) Based on heat content measurement data. (r) On 34.3 at.% Pb alloy made from electrolytic Cu and 99.95% Pb. (s) Based on vapor pressure data using dew point method.

[58Abd], based on the observed constancy of the vapor pressure of Pb with composition in this temperature range, was not supported by other works.

An accurate critical composition of a miscibility gap is often difficult to determine experimentally, and the reported compositions in Table 2 are approximate, because they have been derived from either interpolation or extrapolation of the measured composition data. The presence of a shallow gap at the top, as in the Cu-Pb system, will probably introduce errors in such estimations. This may account, to a certain extent, for the large difference existing between the reported critical composition results, as shown in Table 2. Thermodynamic calculations, by contrast, can offer accurate determination of the critical composition and temperature, provided the selected thermodynamic variables used in the calculation are consistent with the features of the experimental phase diagrams at other points. The critical composition calculated by

[65Sch2] was 35.5 at.% Pb at 1004 °C, and that calculated by [80Tim] was 36.3 at.% Pb at 983 °C. These composition results are comparable with some of the earlier experimental data [13Fri, 31Bri] and the thermal analysis results of [66Sch1]. However, the results are higher than some of the recent measurements [55Sei, 56Pel, 62Szk].

[55Sei] used high-purity material for the alloys and determined the gap boundaries in detail. However, the relative insensitivity of the DTA method near the top of the gap boundary, caused by relatively small changes with temperature of the enthalpy value, makes the absolute accuracy of their results somewhat uncertain. The accepted composition has been established tentatively at 35 ± 1 at.% Pb, as a compromise between the calculated results of [65Sch2] and the experimental results of [65Sch1] and [55Sei], because the width of the gap at the monotectic temperature given by these works is comparable with the accepted one (see below).

[1897Hey] observed a thermal arrest at 954 °C, corresponding to the monotectic transformation. Subsequent measurements confirmed the occurrence of this transformation, and the reported results overlapped between 953 and 957 °C, except for the one by [15Bog], which was too low (see Table 2). The accepted monotectic temperature has been established at 955 °C, as obtained by [56Pel].

There is considerable uncertainty regarding the composition of the Pb-rich liquid at the monotectic temperature. [1897Hey] reported the composition at 954 °C to be 65 at.% Pb, compared to 67 at.% reported by [15Bog], [33Nis], and [51Kle], (the latter from extrapolation of the measured liquidus data from the Pb-rich end up to 954 °C), 65 at.% by [55Sei], and 64 at.% by [56Pel]. Some of the other reported results were unacceptably too high or too low (see Table 2). The accepted composition has been tentatively established at about 65 at.% Pb, in keeping with both the experimental as well as the thermodynamically calculated values of [65Sch2] and [80Tim].

The probable composition of the Cu-rich liquid at the monotectic point is between 14 and 17 at.% Pb, while still higher values were also reported in the literature. Pending further experimental verification, the monotectic composition is tentatively accepted to be 15.5 ± 0.5 at.% Pb, in keeping with the experimental results of [55Sei] and [56Pel], as well as the calculated results of [65Sch2] and [80Tim].

Terminal Solid Solubility. Cu and Pb have very restricted mutual solubility in the solid state. The solubility of Pb in (Cu) does not exceed 0.09 at.% at 600 °C, as determined by X-ray and microscopy by [46Rau]. The solubility of Cu in (Pb) is probably less than 0.023 at.%, because precipitates were observed by optical microscopy in cast and rolled specimens of that composition [38Gre].

The suggestion by [63Kim] of at least 5 at.% Pb being soluble in Cu between 900 and 1000 °C, based on vapor pressure measurements, is apparently not correct.

Metastable Phases

There is no report of any metastable phase in the Cu-Pb system. A possible metastable extension of the terminal solid solubility of Pb in (Cu), amounting to 3.3 to 4.0 at.%, was observed by [50Rau] in electrolytically deposited Cu-Pb alloys. Metastable extensions of the liquidus curves, below the monotectic point, are shown by dashed lines in Fig. 1.

Crystal Structure and Lattice Parameters

Cu and Pb both have fcc structures, but because of the large difference in Goldschmidt atomic radii (37%), they are nearly immiscible in each other in the solid state. No lattice parameter data exist for the solid solutions. Pb undergoes an allotropic transformation to the cph structure under pressure, corresponding to 130 kbar at room temperature [69Tak].

The crystal structure and lattice parameter values for the elements are presented in Table 3.

Thermodynamics

Thermodynamic Measurements. The thermodynamic properties of the Cu-Pb liquid alloys were measured as a function of temperature and composition by several authors and were summarized in [Hultgren, Binary]. The selected enthalpy (ΔH) and excess entropy ($^e\Delta S$) values at 1473 K, given by [Hultgren, Binary] were based primarily on the heat content measurements by Schurmann and Kaune [65Sch1] and agree closely with the corresponding values calculated by Kleppa [52Kle] from the phase diagram. These variables show large positive deviations from ideality. The results of [66Yaz] (based on vapor pressure measurements by the transportation method), of [30Kaw] (by the calorimetric method), and of [73Kir] (by the emf method) are in qualitative agreement with the selected values, but are considerably more endothermic. The emf results of [71Dee] gave even higher values for these variables and must be rejected. The vapor pressure data of [58Abd] and [63Kim] showed considerable scatter with respect to the selected values and are also inconsistent with the phase diagram.

Timucin [80Tim] made careful measurements of the vapor pressure of Pb by the dew point method, using Cu and Pb of 99.99% purity. The measurements were made between 1000 and 1200 °C at 50 °C intervals and between 2 and 90 at.% Pb. The various partial and integral properties derived from these measurements were presented in their paper. The ΔH values of [80Tim] showed reasonable agreement with the selected results of [Hultgren, Binary], but the corresponding $^e\Delta S$ curve, by contrast, was considerably skewed and went through a negative deviation near the Cu end. The results of [80Tim] confirmed the conclusions of [65Sch1] and [66Yaz] that the Kopp-Neumann rule holds in these alloys, in that the partial and integral quantities are independent of temperature.

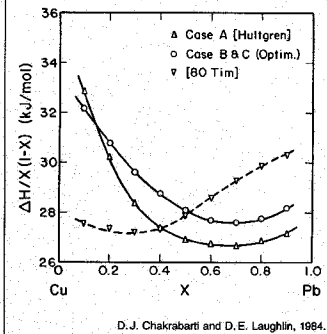
The activity coefficient of Cu, at infinite dilution in liquid Pb ($\gamma_{Cu}^{inf, L}$), was estimated by [56Lan] to be 2.23, based

Table 3 Crystal Structure and Lattice Parameter Data

Phase	Approximate composition(a), at.% Pb	Pearson symbol	Strukturbericht designation	Space group	Prototype	Lattice parameters, nm a c	Comment	Reference
(Cu)	0(b)	cF4	A1	Fm3m	Cu	0.36147 ...	(c)	[Landolt-Börnstein]
(Pb)	100	cF4	A1	Fm3m	Cu	0.49502 ...	(d)	[81Kin]
Pressure-stabilized form								
Pb	100	hP2(e)	0.3265(4) 0.5387(7)	(f)	[69Tak, 83Kin]

(a) From the phase diagram. (b) Metastable solid solubility may extend up to 3.3 to 4.0 at.% Pb (see text). (c) At 18 °C, for elemental Cu. (d) Structure is stable at atmospheric pressure up to 320 °C [41Sto] and down to 20 K [26Keel]. (e) From [83Kin]. (f) At 139 kbar, at room temperature, the change in atomic volume on transition from fcc to cph was found by [69Tak] to be -1.2%.

Fig. 2 Normalized Enthalpy, $Q = \Delta H/X(1-X)$, vs X in the Cu-Pb System



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Table 4 Thermodynamic Properties of Phases in the Cu-Pb System

Integral molar excess free energy of liquid

Case A: ${}^{\circ}\Delta G^L = X(1-X)[36450 - 41700X + 57250X^2 - 25000X^3] - T(10.89 - 34.31X + 54.53X^2 - 26.3X^3)$ (J/mol)

Case B: ${}^{\circ}\Delta G^L = X(1-X)[33875 - 18165X + 13115X^2] - T(6.94 - 9.37X + 9.78X^2)$ (J/mol)

Case C: ${}^{\circ}\Delta G^L = X(1-X)(36450 - 41700X + 57250X^2 - 25000X^3) - T(5.04 - 9.20X + 12.63X^2)$ (J/mol)

[80Tim]: ${}^{\circ}\Delta G^L = X(1-X)[27620 - 2760X + 6800X^2] - T(-1.55 + 13.80X - 5.41X^2)$ (J/mol)

Lattice stability parameter of Cu:

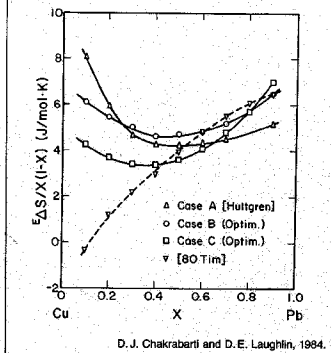
${}^{\circ}G_{Cu}^{w-1} = 7683 + 38.844T + 1.8933 \times 10^{-2}T^2 - 6.527T \ln T$ (J/mol)

where X is atomic fraction of Pb, and T is temperature in K.

on the data of saturation solubility of Cu (6.2 at.%) in liquid Pb at 657 °C, reported by [13Ber], and the relative partial molar enthalpy of Cu in liquid Pb ($\Delta H_{Cu}^{(l)}$) of 23.974 kJ/mol at 6.2 at.% Cu, reported by [30Kaw]. The value of $\gamma_{Cu}^{(l)}$ has been recalculated in this evaluation, using the presently accepted solubility value at 657 °C of 4 at.% Cu (see Fig. 1), the $\Delta H_{Cu}^{(l)}$ value at 4 at.% Cu of 25.1 kJ/mol [Hultgren, Binary], and the lattice stability parameter of Cu [see Table 4], and was found to be 3.20.

Thermodynamic Calculations. The miscibility gap boundaries in the Cu-Pb system were calculated with varying degrees of success by [65Sch2], [80Tim], and [82Ead]. Modeling calculations performed in this evaluation reproduced satisfactorily the liquidus below the monotectic invariant

Fig. 3 Normalized Excess Entropy, $\beta = {}^{\circ}\Delta S/X(1-X)$, vs X in the Cu-Pb System



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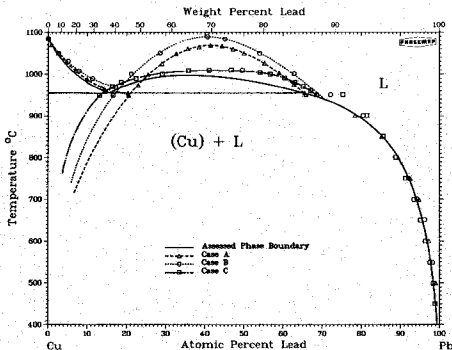
temperature, but much less satisfactorily the corresponding miscibility gap boundaries. The calculations were performed for three different sets of normalized enthalpy, $Q (= \Delta H/X(1-X))$, and normalized excess entropy $\beta (= {}^{\circ}\Delta S/X(1-X))$ values of the liquid, where X is the atomic fraction of Pb. The sources of Q and β values were as follows:

- **Case A:** selected ΔH and ${}^{\circ}\Delta S$ data at 1473 K in [Hultgren, Binary]
- **Case B:** ΔH and ${}^{\circ}\Delta S$ data from [Hultgren, Binary], optimized with respect to the experimental phase diagram in Fig. 1
- **Case C:** ΔH data from [Hultgren, Binary] and ${}^{\circ}\Delta S$ data derived from the optimization fit of the above ΔH , with respect to the experimental miscibility gap boundaries in Fig. 1

Plots of Q and β curves for the three cases and from the data of [80Tim] are shown in Fig. 2 and 3, respectively. The corresponding polynomial expressions for the liquid excess free energy are shown in Table 4. The lattice stability parameter of the solid phase, based on Cu and assumed to have negligible solubility of Pb, has been derived from the heat capacity data of Cu by [77Bar] and is also given in Table 4.

The liquidus curves in equilibrium with the (Cu) phase for Cases A, B, and C, together with the corresponding miscibility gap boundaries, have been calculated on the basis of the above data, assuming Q and β to be independent of temperature. The results presented in Fig. 4 show good agreement of the calculated liquidus, with the experimental liquidus below the monotectic temperature for Case A, whereas for Cases B and C increasing deviations are observed at higher temperatures. The miscibility gap

Fig. 4 Calculated vs Experimental Liquidus in the Cu-Pb System



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boundaries in all three cases, however, show large discrepancy with respect to the experimental boundary. The same has been experienced with the Q and β expressions derived from the data of [80Tin]. The best overall fit has been obtained with Case A, corresponding to the thermodynamic data from [Hultgren, Binary], but even this was not satisfactory. Small modifications in the compositional fit of either Q or β expressions were found to result in large and complex variations in the shape of the gap boundary.

Presumably, the shapes and the positions of the two minima in the free energy curve of the liquid are shallow and are extremely sensitive to small variations in the relative values of ΔH and ΔS . This results in large changes in composition and temperature values, corresponding to the points of common tangency between the two minima, or between one of the minima and that of the coexisting (Cu) phase. Thus, very accurate determination of the thermodynamic properties of the liquid is required for an accurate calculation of the miscibility gap boundary. The accepted liquidus and the miscibility gap boundaries, therefore, have been based entirely on the experimental phase diagram data.

The Cu-Pb system, as contributed in a short version by R. E. Johnson, was previously published in provisional form in the *Bulletin* [80Joh]. The present evaluation reviews all bibliography and data on the Cu-Pb system available in the literature through 1982 and includes information pertaining to crystal structures, metastable phases, and thermodynamics. The present authors have also performed thermodynamic assessment of certain phase boundaries. The present evaluation supersedes the earlier work.

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*Indicates key paper.

#Indicates presence of a phase diagram.

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