The Cu-Hg (Copper-Mercury) System

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The Cu-Hg phase diagram shown in Fig. 1 is derived primarily from the works of [71Lug] and [63Jan1]. It is characterized by the presence of a wide region of immiscibility in the liquid and a nearly vertical liquidus at the Hg-rich end. The (Cu) terminal solid solution has a limited homogeneity, and the (Hg) solid solution has almost none. An intermediate phase is present below 128 °C. The stoichiometry of this phase is believed to be Cu₂Hg. The crystal structure and lattice parameter values for Cu-Hg phases are listed in Tables 1 and 2. No thermodynamic data are available on the solution phases.

The Cu-Hg phase diagram reported by Lagscheider and Jangg [71Lug] and by Jangg and Palmn [63Jan1] was not based on the standard pressure of 1.01 kbar (1 atm). At temperatures above the boiling point of Hg (356.6 °C) [80CRC], the phase boundaries were determined at pressures higher than 1.01 kbar. The pressure was maintained constant at a fixed temperature, but it was increased as the equilibrating temperature increased. The resulting change in the phase boundaries relative to the standard 1.01 kbar condition is not known, as the molar volumes of the alloys and the system pressures during the measurements are not known. However, judging from the report by the authors, up to a maximum pressure of 100 kbar, the corresponding corrections needed (particularly for the composition axis) are, perhaps, not very significant.

Fig. 1 Assessed Cu-Hg Equilibrium Diagram

Table 1  Cu-Hg Crystal Structure Data

<table>
<thead>
<tr>
<th>Phase</th>
<th>Composition, at% Hg</th>
<th>Pearson symbol</th>
<th>Space group</th>
<th>Strukturbericht designation</th>
<th>Prototype</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Cu</td>
</tr>
<tr>
<td>CuHg</td>
<td>0</td>
<td>aF4</td>
<td>Fm3m</td>
<td>A1</td>
<td>Cu</td>
</tr>
<tr>
<td>(αHg)</td>
<td>100</td>
<td>hR 52</td>
<td>R3m</td>
<td>A10</td>
<td>aHg</td>
</tr>
<tr>
<td>(βHg)</td>
<td>100</td>
<td>hR 1</td>
<td>R 3m</td>
<td>A10</td>
<td>aHg</td>
</tr>
<tr>
<td>(γHg)</td>
<td>180</td>
<td>d2</td>
<td>I4/mmm</td>
<td>A10</td>
<td>αFa</td>
</tr>
</tbody>
</table>

(αHg) Formed from αHg by strain-induced (martensitic) transformation at 4.2 K; structure not known, but believed to be simple rhombohedral [65Wea]; γHg reverts to αHg at 50 K.

Table 2  Cu-Hg Lattice Parameter Data

<table>
<thead>
<tr>
<th>Phase</th>
<th>Composition, at% Hg</th>
<th>Lattice parameters, nm</th>
<th>Comment</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>(Cu)</td>
<td>0</td>
<td>a 0.36147</td>
<td>...</td>
<td>[Landolt]</td>
</tr>
<tr>
<td>CuHg</td>
<td>46</td>
<td>c 90.425</td>
<td>90° 31.7</td>
<td>[69Lin]</td>
</tr>
<tr>
<td>(αHg)</td>
<td>100</td>
<td>d 70° 44.6</td>
<td>70° 44.6</td>
<td>[57Bar]</td>
</tr>
<tr>
<td>(βHg)</td>
<td>100</td>
<td>e 70° 44.6</td>
<td>70° 44.6</td>
<td>[59Ato]</td>
</tr>
<tr>
<td>(γHg)</td>
<td>100</td>
<td>f 70° 44.6</td>
<td>70° 44.6</td>
<td>[68Ab}</td>
</tr>
</tbody>
</table>

(a) Transition of αHg → βHg results in 2.6% volume decrease at 78 K.

Equilibrium Diagram

The equilibrium phases in the Cu-Hg system include:

- The liquid, L, which manifests a region of immiscibility below ~ 101 °C, extending to 660 °C and from 15 to ~ 90 at. % Hg.
- The fcc solid solution, (Cu), which extends to 5 at. % Hg at 660 °C.
- The rhombohedral solid solution, (αHg), which is stable below approximately ~ 38.8 °C and has negligible solubility of Cu.
- The rhombohedral intermediate phase, γ, presumably of stoichiometry Cu₀.₀₄Hg₀.₉₆, which forms peritectically at 128 °C; the lower temperature limit of stability of γ is not known.

Liquids and Solidus. Information on the liquidus within the miscibility gap and at Cu-rich compositions is derived from detailed investigations of Lugscheider and Jangg [71Lug]. The Hg-rich portion of the liquidus is based on careful studies of Jangg and Palman [63Jan1].

The occurrence of two temperature-invariant transformations at 660 and 128 °C was established by [71Lug], based on thermal arrests in DTA measurements in several specimens between 7 and 70 at.% Hg. However, the sensitivity of the DTA method was not adequate for the determination of the miscibility gap boundaries, because of the relatively small enthalpy changes that accompany these phase transformations in the liquid. Instead, modification of the sensitive sampling method was used by the authors. An equiatomic liquid alloy sample was formed by complete dissolution of Cu in Hg. This liquid was equilibrated in the miscibility gap region at successively lower temperatures, and the corresponding compositions of the liquids were determined. Separation of the two liquid layers was effected by gravity, through prolonged holding in a vertical tube at the selected temperatures and rapid solidification of the layers. The excess liquid was distilled off prior to chemical analysis. The results indicate a miscibility gap that extends from 15 to about 90 at.% Hg at the monotectic temperature of 660 °C. The critical point occurs at about 810 °C and between 50 and 55 at.% Hg (Fig. 1). The monotectic point at 15 at.% Hg was found to coincide with the composition at which the heat evolution curve showed a sharp change [71Lug]. Although the above measurements were made at temperatures above the boiling point of Hg and under Hg vapor pressures up to 100 kbar, the results indicate a fair degree of consistency.

The Cu-rich portion of the liquidus was determined by [71Lug] based on alloys containing 3.7, and 10 at.% Hg. Specimens from each of these compositions were equilibrated at temperatures 700, 600, and 500 °C, respectively, using a technique similar to that for the miscibility gap determination. Occurrence of a single (solid or liquid) or two (solid and liquid) phases in the specimen was noted from the color differences. This method, however, could not be very accurate. The approximate liquidus and solidus boundaries drawn based on this method are shown in Fig. 1.

The calculated maximum limiting slope of the liquidus at (Cu), corresponding to the zero solid solubility of a solute in (Cu), is ~ 11.76 °C per at. % solute, for the enthalpy of fusion of Cu [45.3 kJ/mol] and the fusion temperature of Cu [Tf(Cu)] is 1358.02 K. Because Hg has finite solid solubility in (Cu) [71Lug], the corresponding initial slope should be lower than the above, and not higher, as suggested by the results of [71Lug]. Thus, the curvature of the liquidus must change its sign near Cu, to conform with the thermodynamic requirements of the much reduced initial slope at (Cu). This has been indicated in Fig. 1.

The Hg-rich portion of the liquidus was determined by [63Jan1] between room temperature and 550 °C, using a modified version of the sampling technique. Cu was added to Hg and allowed to saturate by partial dissolution in the
Table 3  Cu-Hg Liquidus and Solidus Data

<table>
<thead>
<tr>
<th>Reference</th>
<th>Method</th>
<th>Temperature, °C</th>
<th>Composition, at.% Hg</th>
<th>Temperature, °C</th>
<th>Composition, at.% Hg</th>
</tr>
</thead>
<tbody>
<tr>
<td>[71Lug]</td>
<td>Direct</td>
<td>800</td>
<td>43.5 60</td>
<td>770</td>
<td>33.4 75.6</td>
</tr>
<tr>
<td></td>
<td></td>
<td>750</td>
<td>34.9 80.1</td>
<td>700</td>
<td>19.2 85.9</td>
</tr>
<tr>
<td></td>
<td></td>
<td>650</td>
<td>53.9</td>
<td>610</td>
<td>97.7</td>
</tr>
<tr>
<td>[63Jan1]a</td>
<td>Direct</td>
<td>450</td>
<td>98.4 500</td>
<td>98.97</td>
<td>99.47</td>
</tr>
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<td></td>
<td></td>
<td>350</td>
<td>99.24</td>
<td>300</td>
<td>99.47</td>
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<td></td>
<td></td>
<td>250</td>
<td>99.66</td>
<td>200</td>
<td>99.81</td>
</tr>
<tr>
<td></td>
<td></td>
<td>150</td>
<td>99.89</td>
<td>100</td>
<td>99.961</td>
</tr>
<tr>
<td>[67Lev]</td>
<td>Electrometric</td>
<td>50</td>
<td>99.977</td>
<td>40</td>
<td>99.998</td>
</tr>
<tr>
<td>[74Lun]</td>
<td>Electrometric</td>
<td>20</td>
<td>99.99</td>
<td>70</td>
<td>99.97</td>
</tr>
<tr>
<td></td>
<td></td>
<td>100</td>
<td>99.952</td>
<td>40</td>
<td>99.998</td>
</tr>
<tr>
<td></td>
<td></td>
<td>20</td>
<td>99.963</td>
<td>70</td>
<td>99.97</td>
</tr>
<tr>
<td></td>
<td></td>
<td>60</td>
<td>99.975</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Note: RT = room temperature.  
(a) As transcribed from the author’s figure and reported in [Shunk].  
(b) As transcribed from the author’s figure.

Table 4  Solubility of Cu in Hg at Room Temperature

<table>
<thead>
<tr>
<th>Solubility, at.% Hg</th>
<th>Method</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0098</td>
<td>Direct</td>
<td>[98Hum]</td>
</tr>
<tr>
<td>0.0076</td>
<td>Direct</td>
<td>[10Ric]</td>
</tr>
<tr>
<td>0.0101</td>
<td>Electrometric</td>
<td>[27Tam]</td>
</tr>
<tr>
<td>0.008</td>
<td>Direct</td>
<td>[32Irv]</td>
</tr>
<tr>
<td>0.0101</td>
<td>Colorimetric</td>
<td>[55Liu]</td>
</tr>
<tr>
<td>0.0022</td>
<td>Direct</td>
<td>[55Sel]</td>
</tr>
<tr>
<td>0.006</td>
<td>Electrometric</td>
<td>[63Jan1]</td>
</tr>
<tr>
<td>0.008</td>
<td>Electrometric</td>
<td>[63Jan2]</td>
</tr>
<tr>
<td>0.0087 ± 0.003</td>
<td>Electrometric</td>
<td>[65Cha]</td>
</tr>
<tr>
<td>0.010</td>
<td>Electrometric</td>
<td>[67Lev]</td>
</tr>
<tr>
<td>0.0097</td>
<td>Electrometric</td>
<td>[69Boi]</td>
</tr>
</tbody>
</table>

Note: Accepted results are shown in boldface type.

The Cu-rich solidus below the monotectic temperature (660 °C) was determined by [71Lug] from the variations in the lattice parameter values by XRD. The results show (Fig. 1 and Table 3) that the solid solubility of Hg in (Cu), perhaps negligible at room temperature [29Kat], increases to a maximum of 5 at.% at 660 °C and remains nearly constant at that value up to ~ 750 °C. The distinct break in the solubility curve at 660 °C obtained in these studies is in agreement with the thermal analysis results, which confirm the invariant temperature to be 660 °C. The X-ray results of the solubility above 660 °C would indicate a retrograde behavior. However, because the data are limited and not definitive, they are discarded in favor of a smooth nonretrograde-type solidus curve, obtained from the high-temperature measurements by these authors, as described previously. The high solubility value (9 at.% Hg) at ~ 150 °C suggested by [23Tam] is apparently incorrect.

Solubility of Cu in Liquid Hg at Room Temperature. Several reports are available on the solubility of Cu in Hg at room temperature. The inherently low solubility value is partly responsible for the scatter in the reported results given in Table 4. The solubility values reported are generally high. In many instances, this arose due to the incomplete filtration of the Cu-Hg amalgam (especially when using chamois-leather filter that contain varying pore sizes), so that some of the microcrystalline solid Cu pieces also passed through the filter, resulting in higher analysis for Cu in the liquid. By contrast, the use of a sintered Jena glass filter by Irving and Russell [32Irv] gave lower values and better consistency.

Whether using different methods of preparation of the amalgam also has any bearing on solubility results, as claimed by [32Irv], is not certain, because equilibrium solubility should remain the same. Solubility as deter-
Table 5 Cu-Hg Thermodynamic Parameters, J/mol, T in K

Lattice stability parameters

\[ \Delta G^\alpha L = 13.054 - 9.613T \] [Hultgren, E]  
\[ \Delta G^\beta L = 2295.5 - 9.80T \] [Hultgren, E]  
\[ RT \ln y_{\text{Cu}} = 2400 - 1.46T \] [This work]

where \( \alpha \) is fcc and \( \beta \) is rhombohedral.

Integral molar excess Gibbs energy of liquid [This work]

\[ G^\infty(\text{Cu}) = X(1-X)(33,545 + 2069X - 15.487T) \]
\[ G^\infty(\text{Hg}) = (132,200 \pm 400) + T(328.4 \pm 0.5) \] [71Lug]

where \( X \) is the atomic fraction of Hg.

Note: Mole for compound refers to respective atomic units as elementary entities; standard states are pure liquid Cu and pure liquid Hg.

Table 5. Cu-Hg Thermodynamic Parameters, J/mol, T in K

<table>
<thead>
<tr>
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</thead>
<tbody>
<tr>
<td>( \Delta G^\alpha L ) = 13.054 - 9.613T [Hultgren, E]</td>
</tr>
<tr>
<td>( \Delta G^\beta L ) = 2295.5 - 9.80T [Hultgren, E]</td>
</tr>
<tr>
<td>( RT \ln y_{\text{Cu}} ) = 2400 - 1.46T [This work]</td>
</tr>
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where \( X \) is the atomic fraction of Hg.

Note: Mole for compound refers to respective atomic units as elementary entities; standard states are pure liquid Cu and pure liquid Hg.

mined by the electrometric method (by noting the dilution level of the amalgams at which the potential drop measured at a calomel electrode reached a constant value) in general showed somewhat higher values, about 0.01 at. % Hg. The direct method is one in which the saturated amalgams were filtered and the filtrate analyzed for the metal (by colorimetric or gravimetric method), following the removal of excess Hg by distillation/volatilization. This method gave lower values [for (1896Hun)] and more reliable values, as indicated in the results of [32irv] and [63Jaw]. The solubility of Cu in (Hg) at room temperature is accepted as 0.006 at. %, based on the latter two studies.

Intermediate Phase. Tammann and Stassfurth [25Tam] observed several conflicting (thermal) arrests in their thermal measurements on the electrolytically prepared Cu-Hg amalgams of wide composition ranges. Based on X-ray analysis, they reported the presence of at least one compound, with the CuHg composition and a decomposition temperature of 96 °C. [Hansen] recorded several earlier reports of the observation of intermediate phases in the Cu-Hg system. After the studies of [25Tam], other works confirmed the occurrence of a stable intermediate phase, but differed on whether the composition is CuHg, 28Tcr, 53LiH, or CuHg [35Schn]. Kato [29Kat] designated it \( \gamma \) phase and determined its Cu-rich composition limits to be between 54 and 57.4 at. % Cu. The Cu-Hg amalgams, in this work, were prepared electrolytically and subjected to 10,000 kbar pressure to remove the liquid phase from the solid amalgam. [69Lin] proposed the composition to be Cu\(_{56}\),\(^{\gamma}\)Hg\(_{44}\), based on the measured density of 12.6 ± 1 g/cm\(^3\) and the assumption of a γ brass-type structure of this phase. Lindahl and Westman [69Lin] made further revision of the stoichiometry to CuHg. This is consistent with the more precise density data (13 g/cm\(^3\)) of [35Schn], with the rhombohedral structure of the phase, and with the composition limits given by [29Kat]. Lugeholder and Jang [71Lug] also ascribed the same stoichiometry (CuHg) to this phase on the basis of the observed peak in the heat evolution curve at this composition. Thermal arrests observed in the DTA studies by these authors indicate that the phase is formed at 123 °C, presumably by the peritectic reaction between the Hg-rich liquid and the Cu phase (Fig. 1). The lower temperature limit for the stability of the CuHg\(_{\alpha}\) is not known.

Metastable Phases

There is no report of any metastable phase in the Cu-Hg system. Indirect evidence, however, suggests the likelihood of one or more metastable intermediate phases being present. For example, Irvin and Russell [32irv] observed the formation of intermediate phases with different stoichiometries, depending upon the method of preparation of the Cu-Hg amalgams. Thus, CuHg\(_{\alpha}\) was formed by the electrolysis method, CuHg by the displacement method (involving a more reactive metal) and CuHg\(_{\beta}\) by the mechanical mixing of the finely divided Cu in Hg. However, no structural evidence has been presented to confirm this. Likewise, the reports of differing compositions of the intermediate phase \( \gamma \), described earlier, could be due to the formation of nonequilibrium or metastable equilibrium structures.

Crystal Structures and Lattice Parameters

Cu and Hg are negligibly soluble in each other in the solid state at lower temperatures. Consequently, no composition-dependent lattice parameter data on Cu and Hg phases are available. Lattice parameters for the elemental Cu and Hg (with rhombohedral structure) are presented in Table 2. A nearly linear relationship is observed between the lattice parameter and temperature for Hg, as per the measurements of [57Bar] at 5 and 78 K and of [30Meh] at 227 K. At high pressures, αHg undergoes an allotropic transformation to βHg, which has a bct crystal structure [60Ato] and lattice parameters, as shown in Table 2. The calculated density of the β phase is 14.88 g/cm\(^3\), and compared to the density of αHg (14.2 g/cm\(^3\)), such increase with pressure is consistent with the Le Chatelier principle. Although βHg is the stable form below 79 K, it does not form except under high pressures (~4 kbar), because the α → β transformation occurs sluggishly at lower temperatures [62Sch]. The occurrence of a third allotropic form of Hg, the \( \gamma \)Hg phase, was suggested by [68Abe] (see Table 1).

Regarding the intermediate phase \( \gamma \) (not to be confused with \( \gamma \)Hg), the powder XRD pattern was reported to be similar to the γ brass structure, and the lattice parameters based on this cubic unit cell were determined by [29Kat], [30Kat], and [35Schn]. The results, converted from the kx unit, were 0.9420 and 0.9425 nm, respectively. Attempts by [35Schn] to fit their observed XRD intensity data to a cubic structure model corresponding to the stoichiometry Cu\(_{56}\),\(^{\gamma}\)Hg\(_{44}\), as derived from their careful density measurements, were not successful, and the authors instead assumed the CuHg composition for the calculation.

Linental, Pilotti, and Westman [68Lin] interpreted their powder XRD data in terms of a bcc unit cell with a rhombohedral distortion and an assumed ~52 atoms per unit cell. The assumption of a rhombohedral structure with the space group R3m for the \( \gamma \) phase enabled Lindahl and Westman [69Lin] to account for their X-ray data in terms of the stoichiometry Cu\(_{56}\),\(^{\gamma}\)Hg\(_{44}\), which also conformed with their measured density data. The resulting lattice parameter value and the interaxial angle, calculated on the basis of the unit cell described by [69Lin], are presented in Table 2.

Thermodynamics

Except for the Gibbs energy of formation of the CuHg\(_{\alpha}\) phase derived from thermal analysis studies by [71Lug], (Table 5), no thermodynamic data are available on the Cu-Hg alloys. An attempt has been made in this evaluation to obtain analytic expressions for the integral molar
excess Gibbs energy of the liquid and the (Cu) phases from the experimental phase diagram. Based on these derived expressions, the boundaries in the equilibrium diagram have been calculated and compared to the experimentally determined one to check for self consistency.

Because the equilibrium boundaries within the miscibility gap are very sensitive to small variations in the Gibbs energy of the liquid, the modeling calculations were begun with these boundaries. The liquidus compositions on the miscibility gap, across tie-lines at many closely spaced temperatures (between 660 and 804 °C), were fitted to analytic expressions for the excess Gibbs energy of the liquid phase of the following form:

\[ \Delta G^{ex}(L) = X(1 - X) \sum_{i=1}^{N} \left( a^{ex}_{i}X_{i}^{i-1} - T \sigma_{i}^{ex}X_{i}^{i-1} \right) \]  
(1)

where, \( a^{ex}_{i} \) and \( \sigma_{i}^{ex} \) are, respectively, the coefficients of the enthalpy (\( \Delta H/X(1 - X) \)) and entropy (\( \Delta S^{ex}/X(1 - X) \)) functions of the liquid and \( X \) is the atomic fraction of Hg. The coefficients are assumed to be independent of temperature. The standard states were referred to the liquid phases for both pure Cu and pure Hg. The simultaneous linear equations generated were solved by the standard multiple least-squares regression analysis (and Gauss-Jordan reduction algorithm). The resultant expression for \( \Delta G^{ex}(L) \) is as follows:

\[ \Delta G^{ex}(L) = X(1 - X) \times \left( 33.545 + 2069X - 15.48T \right) \text{ J/mol} \]  
(2)

where \( T \) is in K. The maximum value of \( \Delta H \) according to Eq 2, corresponding to \( X = 0.508 \), is 8647 J/mol, and the corresponding \( \Delta G \) value estimated at 1000 K is -984 J/mol. The calculated gap boundaries at selected temperatures based on Eq 2 are shown in Fig. 2. The close agreement between the calculated and experimental gap boundaries shows self consistency in the modeling only, and not the authenticity, or otherwise, of the experimental boundary. The metastable boundaries of the miscibility gap below 660 °C, calculated using the same expression, are also shown in Fig. 2.

The lattice stability parameter for fcc Cu was obtained from [Hultgren, E]. To derive an approximation of the same for Hg in the fcc phase, (Cu) was assumed to be Henrian:

\[ a^{Cu}_{Cu} = X_{Cu}^{Cu} \]  
(3)

and

\[ a^{Cu}_{Hg} = \gamma_{Hg}^{0Cu}X_{Hg}^{Cu} \]  
(4)

where, \( \gamma_{Hg}^{0Cu} \) is the Henrian activity coefficient of Hg in the (Cu) phase. The \( \gamma_{Hg}^{0Cu} \) at several temperatures, relative to pure liquid Cu as the standard state, were derived based on Eq 2 and the phase equilibrium data between (Cu) and L phases. The resulting values of \( \gamma_{Hg}^{0Cu} \) at 933, 873, 773, and 673 K were 18.6, 22.7, 36.4, and 62, respectively. The expression for the \( RT \ln \gamma_{Hg}^{0Cu} \) obtained from the above data is shown in Eq 5:

\[ RT \ln \gamma_{Hg}^{0Cu} = 24000 - 1.46T \text{ J/mol} \]  
(5)

Based on Eq 2 and 5 and the lattice stability parameter of Cu in Table 4, \( \Delta G^{st} \) for (Cu) was derived from the multiple least-squares regression analysis of the phase equilibria data between L and (Cu) below 600 °C, as described earlier for the liquid. This is shown in Eq 6:

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**Fig. 2** Calculated Versus Experimental Cu-Hg Phase Diagram


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\[ \Delta G^\circ \text{Cu} = X(1 - X) \times (24790 - 138500X - 40.91T) \text{ J/mol} \]  

(Eq 6)

The solvus and the liquidus boundaries below the monotectic temperature (660 °C) were calculated at selected temperatures based on data in Table 5, and are shown in Fig. 2. A good reproduction of the solvus indicates that the modeling in this region is self-consistent. However, the fit for the liquidus shows increasing deviations at higher temperatures. This indicates that the thermodynamic expression for the liquid derived from the miscibility gap boundary data may not be very reliable. This is understandable, because small uncertainties in the miscibility gap boundaries can affect the derived Gibbs energy values in a major way, and vice versa. Also, no data from the L/L + (Cu) boundaries are available to model the liquid. This, perhaps, is partly responsible for the inability of the model to reproduce the monotectic reaction without severe distortion of the gap boundaries, or vice versa.

Cited References


* Indicates key paper.
# Indicates presence of a phase diagram.