Au-Tc Crystal Structure Data

Phase	Approximate composition, at.% Tc	Pearson symbol	Strukturbericht designation	Space group	Prototype
(Au)	0	*cF4*	*A1*	*Fm3m*	Cu
(Tc)	? to 100	*hP2*	*A3*	$P6_3/mmc$	Mg

Au-Tc evaluation contributed by **H. Okamoto** and **T. B. Massalski,** Department of Metallurgical Engineering and Materials Science, Carnegie-Mellon University, Pittsburgh, PA 15213, USA. Work was supported by the International Gold Corporation Limited (InterGold) and American Society for Metals (ASM). Literature searched through 1983. Part of the bibliographic search was provided by ASM. Professor Massalski is the AMS/NBS Data Program Editor-in-Chief for Binary Alloys, and also Category Editor for binary gold alloys, jointly with Dr. Okamoto.

The Cu-Sr (Copper-Strontium) System

63.546 87.62

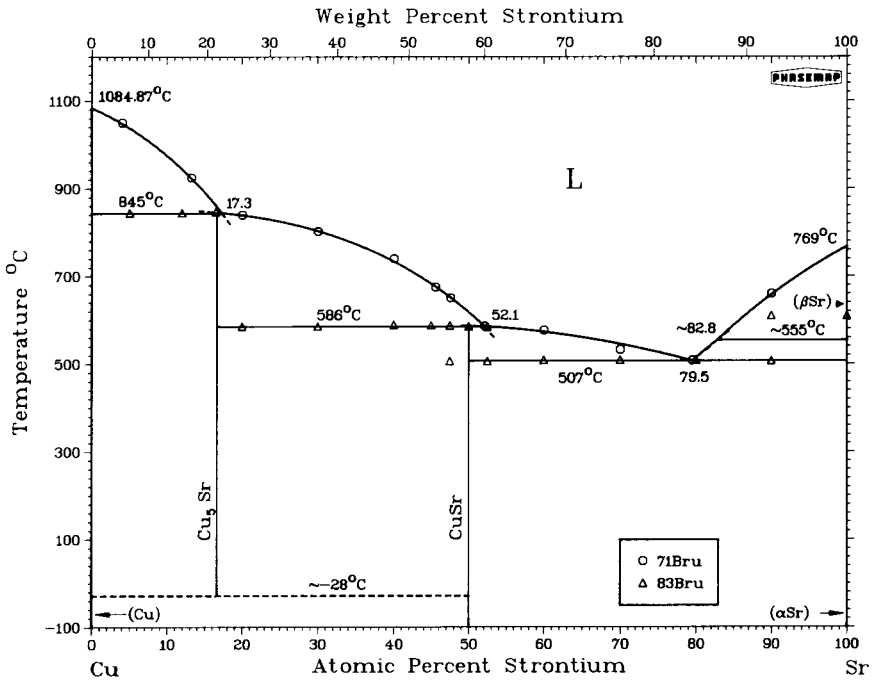
By D. J. Chakrabarti and D. E. Laughlin
Carnegie-Mellon University

The assessed Cu-Sr equilibrium diagram in Fig. 1 has been obtained from the reported work of Bruzzone [71Bru]. Cu and Sr are virtually insoluble in one another in the solid state, while being soluble in all proportions in the liquid state. Two intermediate phases having the stoichiometries Cu_5Sr and CuSr are formed by peritectic transformations at 845 and 586 °C, respectively. The crystal structures of Cu_5Sr and CuSr are isotypic with $CaCu_5$ and BaCu, respectively. No reports of thermodynamic properties of Cu-Sr alloys are available.

Equilibrium Diagram

The equilibrium phases in the Cu-Sr system are: (1) the liquid, miscible in all proportions and stable down to the eutectic at 507 °C at 79.5 at.% Sr; (2) the fcc solid solution, (Cu), with negligible solubility of Sr; (3) the bcc solid solution, (βSr), with negligible solubility of Cu and stable down to ~555 °C; (4) the fcc solid solution, (αSr), with negligible solubility of Cu and stable below ~555 °C; (5) the hexagonal stoichiometric phase Cu_5Sr, stable be-

Fig. 1 Assessed Cu-Sr Phase Diagram



Cu_5Sr eutectoid decomposition temperature of ~−28 °C is from calculations in this work (see text); metastable extensions of the liquidus curves are shown as dashed lines. D. J. Chakrabarti and D. E. Laughlin, 1984.

Table 1 Experimental vs Calculated Liquidus in the Cu-Sr System

Reference(a, b)	Experimental liquidus: Composition, at.% Sr	Experimental liquidus: Temperature, °C	Calculated liquidus [this work]: Temperature(c), °C	Calculated liquidus [this work]: Composition(d), at.% Sr
Liquid-(Cu) equilibrium				
[81BAP]	0	1084.87	1084.9	...
	1.1	1075	1075.7	...
	4.0	1050	1049.1	2.8
	6.3	1025	1024.5	4.9
	8.2	1000	1001.3	6.9
	10.0	975	976.7	8.8
	11.7	950	950.8	10.7
[71Bru]	12.0	940	939.3	...
	13.2	925	925.6	12.6
	14.6	900	899.9	14.4
	15.9	875	874.2	16.2
	17.1	850	848.7	18.0
Liquid-Cu_5Sr equilibrium				
	17.3	845	845.4	...
[71Bru]	20	840	...	20.8
	25	825	824.8	25.3
[71Bru](e)	30	815	...	...
[71Bru]	30	802	803.2	30.0
	34.6	775	776.1	33.7
	38.2	750	749.2	36.8
[71Bru]	40	740	...	...
	41	725	724.2	39.6
	43.4	700	699.9	42.2
	45.6	675	675.0	44.6
	47.6	650	650.3	46.8
	49.4	625	625.9	49.0
	51.2	600	599.8	51.0
[71Bru]	52.1	586	586.0	52.1
Liquid-CuSr equilibrium				
[71Bru]	52.1	586	586.0	...
[71Bru]	60	576.5	577.0	60.2
	65	565	564.9	65.4
	69.75	550	549.3	70.1
[71Bru](e)	70	532	...	...
	74.6	530	529.7	75.0
	76.6	520	520.7	77.0
	77.6	515	516.0	77.9
	79.5	507	506.8	79.4
Liquid-(Sr) equilibrium				
[81BAP]	100	769	769	...
	97.75	750	749.1	98.3
	96.9	740	740.6	97.5
	95	720	720.2	95.7
	93.3	700	700.3	94.1
	91.7	680	680.4	92.4
[71Bru]	90	660	658.1	90.8
	88.7	640	640.5	89.3
	87.2	620	619.5	87.8
	85.8	600	599.5	86.3
	84.5	580	580.7	84.8
	83	560	558.7	83.4
	81.7	540	539.6	81.9
	80.4	520	520.4	80.4
	79.5	507	507.2	79.4

(a) Data from [71Bru] are given, as read out from authors' Fig. 2. (b) Experimental liquidus data without any indicated reference correspond to interpolated values from the data of [71Bru], as read out from the authors' Fig. 2. (c) Temperatures are calculated using Eq 1 to 4, based on experimental/interpolated compositions given in the second column in each respective row. (d) Compositions are calculated using thermodynamic modeling parameters given in Table 5 and pertaining to temperatures given in the third column in each respective row. (e) These data from [71Bru] show scatter and were not used either for modeling or for deriving Eq 1 to 4.

tween 845 and approximately −28 °C; and (6) the hexagonal stoichiometric phase CuSr, stable at all temperatures below 586 °C.

The lower temperature limits of stability for Cu_5Sr and CuSr were not determined experimentally and have been estimated, based on the thermodynamic modeling of the system (see Thermodynamics section).

Sr is very reactive in air, and the Cu-Sr alloys behave likewise. Thus, contamination is always a problem with these alloys, particularly in the liquid. The Sr used by

Bruzzone [71Bru] was purified by distillation. The purity of Sr and Cu used was 99.8 and 99.999%, respectively [83Bru]. The melting point (773 °C) and lattice parameter values (0.6082 nm) of βSr, reported in [71Bru], are comparable with the recent literature data (769 °C [81BAP] and 0.6084 nm [58Sch]). The same was also true for the Cu being used.

The alloys were prepared under argon atmosphere, using Fe crucibles for compositions above 60 at.% Sr and Mo crucibles for Cu-rich compositions [71Bru]. The differential thermal analysis (DTA) method was used during cooling to determine the liquidus and the temperature-invariant boundaries. The rate of cooling was 2 to 4 °C/min, while the melts were stirred prior to cooling. The temperature was measured to an accuracy of ±1 °C, and the composition was determined by chemical analysis. The crystal structures of the phases were studied by X-ray diffraction.

Liquidus and Solidus. The DTA measurements by [71Bru] defined the liquidus boundaries and indicated, from the occurrence of primary thermal arrests, the existence of four temperature-invariant transformations in the Cu-Sr system. The liquidus accepted in this evaluation has been based on the data of [71Bru] for the alloys and on the compilation in [81BAP] for the melting points of Cu and Sr at 1084.87 and 769 °C, respectively.

Although experimental data are not available close to pure Cu and pure Sr, the liquidus drawn by interpolation of data points from the nondilute regions has been used to estimate the initial slopes of the liquidus at Cu and at Sr. These have been found to be −10.5 ± 0.5 °C/at.% Sr and −10.5 ± 0.5 °C/at.% Cu, respectively. Using the van't Hoff's relation, the corresponding initial slopes of the solidus at Cu and Sr (calculated from the enthalpy of fusion and melting points of the respective metals and assuming Raoult's law is satisfied in the liquid and in the solid in both the (Cu) and (Sr) phases) are approximately -10^4 °C/at.% Sr and -3×10^4 °C/at.% Cu, respectively. The results indicate very little mutual solubility between Cu and Sr, which is consistent with the phase diagram by [71Bru] that shows the absence of any significant solubility fields in the (Cu) and (Sr) phases.

The four temperature-invariant transformations indicated by [71Bru] are the two peritectic transformations at 845 and 586 °C associated with the formation of Cu_5Sr and CuSr phases: the eutectic transformation at 507 °C and the polymorphic transformation of (βSr) at ~555 °C. The type of transformation at ~555 °C is not known. This can be either a metatectic or a peritectic transformation, depending on whether the βSr ⇄ αSr transition temperature is lowered or raised in the alloys, compared to that in Sr (see comment by Massalski [83Mas]). Because the thermal measurements were made during cooling, the actual transformation temperatures could be somewhat higher, if undercooling effects were present. However, the lack of significant scatter in the primary thermal arrest data minimizes such a possibility, although the thermal arrest observed at ~47.5 at.% Sr at 507 °C is indicative of incomplete peritectic transformation at 586 °C. The transformation temperatures in Fig. 1, except for the low-temperature decomposition of Cu_5Sr, have been accepted from [71Bru]; the low-temperature invariant is from calculations in the present evaluation. The composition of the liquidus given by [71Bru] at the invariant temperatures 586 and 507 °C were 52.1 and 79.5 at.% Sr, respectively. For the other temperatures, the tentative compositions obtained from interpolation of his liquidus data were ~17 at.% Sr at 845 °C, and ~82.8 at.% Sr at ~555 °C. The experimental liquidus and solidus data, as transcribed from the figure of [71Bru] and on which Fig. 1 is based, are presented in Table 1. The detailed thermal analysis results of [71Bru] are presented in Table 2 (see [83Bru]). The temperature-invariant transformations are shown in Table 3.

Table 2 Thermal Analysis Results of Cu-Sr Alloys [83Bru]

Specimen composition, at.% Sr	Melting temperature, °C	Secondary arrest temperatures, °C		
0	1084	...	...	...
5	1040	845	...	...
12	942	845	...	...
16.5	847	...	...	...
20	840	...	585	...
30	815	...	586	...
40	740	...	589	...
45	680	...	588	...
47.5	648	...	587	(506)
50	...	...	585	...
52.5	...	...	584	506
60	575	...	...	509
70	532	...	...	509
80	508	...	...	508
90	660	...	610	507
100	773	...	610	...

Table 3 Temperature-Invariant Transformations in the Cu-Sr System

Transformation	Temperature, °C	Compositions(a), at.% Sr		
Peritectic				
(Cu) + L ⇄ Cu_5Sr	845	~0	17.3	16.7
Cu_5Sr + L ⇄ CuSr	586	16.7	52.1	50
Eutectic				
L ⇄ CuSr + (αSr)	507	79.5	50	~100
Peritectic or metatectic				
L + (βSr) ⇄ (αSr)	~555	~82.8	~100	~100 (<(βSr))
(βSr) ⇄ L + (αSr)	~555	~100	~82.8	~100 (>(βSr))
Eutectoid				
Cu_5Sr ⇄ (Cu) + CuSr	~−28	16.7	~0	50

(a) Compositions for the phases are given in the order they appear in column 1.

Analytic polynomial representations of the liquidus curves have been obtained by the least-squares analysis in the different composition ranges. These are shown in Eq 1 to 4, below. In deriving these equations, the melting point values for Cu in Eq 1 and for Sr in Eq 4 were set equal to 1084.9 and 769 °C, respectively. The fit between the calculated and the experimental liquidus is within ±2 °C in most of the ranges, as can be seen in Fig. 1 and in Table 1, where X is atomic fraction of Sr and T is in °C.

0 to 17.3 at.% Sr:

$$T = 1084.9 - 792.5\,X - 2061\,X^2 - 8039\,X^3 \quad \text{(Eq 1)}$$

17.3 to 52.1 at.% Sr:

$$T = 863.9 - 89.1\,X + 269\,X^2 - 2152\,X^3 \quad \text{(Eq 2)}$$

52.1 to 79.5 at.% Sr:

$$T = 184.4 + 1763.8\,X - 2281\,X^2 + 721\,X^3 \quad \text{(Eq 3)}$$

79.5 to 100 at.% Sr:

$$T = 769 - 818.4(1 - X) - 3576(1 - X)^2 + 6502(1 - X)^3 \quad \text{(Eq 4)}$$

Intermediate Phases. Two intermediate phases of stoichiometry, Cu_5Sr and CuSr, that form at peritectic temperatures of 845 and 586 °C, respectively, were identified by [71Bru]. These have been indicated as line compounds and are presented as such in Fig. 1. The possible existence of any solubility range in these phases has not been examined.

Crystal Structure and Lattice Parameters

The crystal structures and accepted lattice parameters of Cu, Sr, and of the intermediate phases, Cu_5Sr and CuSr, are shown in Table 4. [53She], [56Hir], and [58Sch] observed three allotropic forms of Sr, having the transition temperatures at 213 ± 3 °C and 602 ± 8 °C, corresponding to the cph ⇄ fcc and bcc ⇄ cph structures. Peterson and Colburn [66Pet], however, showed that the cph Sr is a H-stabilized binary phase and that H_2 also increases the bcc ⇄ cph transition and the melting temperatures of Sr. Using vacuum distilled Sr metal of better than 99.7% purity (containing 0.4 mol% SrH_2) and DTA measurements during heating, they obtained, after correcting for the residual H content in the sample, a temperature of 555 °C for the transition from bcc to fcc lattice of Sr. The corresponding temperature found in DTA measurements during cooling was 540 °C, in agreement with [52Rin]. The melting temperatures of Sr (L ⇄ bcc Sr) reported were 768 °C [66Pet], 770 °C [52Rin], and 774 °C [58Sch], as compared to the accepted value of 769 °C taken from [81BAP]. Thus, the thermal arrests at 610 °C, obtained by [71Bru] on Sr metal and on the Cu-Sr alloy, are probably related to impurity effects, as is the additional transformation point for Sr shown at 225 °C in their figure. The bcc ⇄ fcc transition temperature of Sr has been accepted from [66Pet] to be 555 °C, as shown in Fig. 1, and the associated invariant transformation for Cu-Sr alloys has been indicated at the same temperature, in the absence of any experimental data.

The crystal structure of the Cu_5Sr phase has been identified to be isotypic with $CaCu_5$ [66Bru]. The crystal structure of CuSr was determined by Fornasini and Merlo [80For], using single crystals of the stoichiometric phase prepared from 99.5 wt.% Sr and 99.999 wt.% Cu. Their X-ray analysis was carried out by Laue and Weissenberg, and precession methods and lattice parameter values, presented in Table 4, have been obtained from rotation patterns. CuSr was found to have hexagonal symmetry, in which slabs of trigonal prisms of Sr with Cu atoms in their centers are stacked in a close-packed manner along [001]. The weak Sr-Sr bonds between the adjacent slabs makes the CuSr behave like a layered phase.

The lattice parameters of αSr and βSr have been taken from [58Sch] and may undergo slight modification with purer material. [63Mcw] and [63Jay] studied the effect of pressure on allotropic modification in Sr that showed only two forms, i.e., fcc and bcc Sr. At room temperature, increasing the pressure above 35 kbar changes the fcc αSr phase to the bcc βSr phase. According to the Le Chatelier principle, this means that the density of the bcc phase is greater than that of the fcc phase. This has been found to be true in this system, using the lattice parameter data shown in Table 4. The density of βSr at 42 kbar and room temperature is 3.33 g/cm^3, and that for αSr at 1.013 bar and 25 °C is 2.58 g/cm^3. This rather rare phenomenon also occurs in Yb [63Hal]. The works of [63Jay] on vacuum distilled high-purity Sr (>99.8% purity) at 1 atm pressure gave 557 °C for the βSr ⇄ αSr transition temperature, confirming the results of [66Pet].

Thermodynamics

No thermodynamic data are available on the Cu-Sr alloys. Because the Cu-Sr phase diagram is reasonably well established, the equilibrium boundaries between the coexisting phases can be used to derive expressions for the thermodynamic conditions of the different phases. The expressions, in turn, can be used to reproduce the phase boundaries as a check for self-consistency and also to calculate the regions of the phase diagram not determined experimentally. Both of these aspects have been attempted in this evaluation, following the approach below.

Because both the (Cu) and the (Sr) phases display virtually zero solubility, they have been assumed to be line phases, and the representation of their molar free energy has been obtained from the respective lattice stability parameter values for the solid-liquid transition, as given in [Hultgren; Elements]. The resultant expressions relative to pure liquid Cu and pure liquid Sr as standard states (used throughout the calculation) are presented in Table 5.

Table 4 Cu-Sr Crystal Structure and Lattice Parameter Data

Phase	Approximate composition(a), at.% Sr	Pearson symbol	Space group	Prototype	Lattice parameters, nm a	c	Comment	Reference
(Cu)	0	*cF*4	*Fm*3*m*	Cu	0.36147	···	(b)	[Landolt-Börnstein]
Cu_5Sr	16.67	*hP*6	*P*6/*mmn*	$CaCu_5$	0.5261 ± 0.0002	0.4058 ± 0.0002	(c)	[66Bru]
CuSr	50	*hP*8(?)	$P6_3/mmc$	BaCu	0.4341	1.538	(c)	[80For]
(βSr)	100	*cI*2	*Im*3*m*	W	0.485	···	(d)	[58Sch]
(αSr)	100	*cF*4	*Fm*3*m*	Cu	0.6084	···	(e)	[58Sch]
Pressure-stabilized form								
βSr or Sr-II	100	*cI*2	*Im*3*m*	W	0.4437 ± 0.0002	···	(f)	[63Mcw]

(a) From the phase diagram. (b) At 18 °C, on elemental Cu. (c) On single-crystal samples. (d) At 614 °C. (e) At 25 °C. (f) At room temperature, under 42 kbar pressure.

The liquidus boundaries between 1084.87 and 845 °C, and between 769 and 507 °C, have been used to estimate the integral molar excess free energy expressions for the liquid. The latter has been expressed as a polynomial, with composition in the following form:

$$^{E}\Delta G^{L} = X(1 - X)\sum_{i=1}^{N}(a_i^H x_i^{i-1} - T \cdot b_i^S x_i^{i-1}) \qquad \text{Eq 5}$$

where a_i^H and b_i^S are, respectively, the coefficients of the enthalpy ($\Delta H/X(1 - X)$) and entropy ($^{E}\Delta S/X(1 - X)$) functions of the liquid and X is the atomic fraction of Sr. The coefficients have been assumed to be independent of temperature. The simultaneous linear equations, set up from the equilibrium between the liquid and the solid phases at several temperatures, have been solved by the standard multiple least-squares regression analysis (and the Gauss-Jordan reduction algorithm) to derive the values of the coefficients a_i^H and b_i^S, respectively. The number of the a^H and b^S terms has been limited to a minimum in these calculations, as a compromise between the reproducibility of the calculated diagram that improves with their increased numbers, and the simplicity of the model. A total of 37 data points have been used to derive one coefficient each for a^H and b^S that could reproduce the phase diagram satisfactorily. The resultant expression for the $^{E}\Delta G^{L}$ is as follows:

$$^{E}\Delta G^{L} = X(1 - X)(-20\,780 + 7.55\,T) \quad \text{(J/mol)} \qquad \text{Eq 6}$$

The maximum value of ΔH, according to Eq 6, is −5195 J/mol at $X = 0.5$, and the corresponding ΔG value, estimated at 1000 K, is −9070 J/mol.

From a knowledge of the molar free energy of the liquid, the molar free energy of Cu_5Sr, expressed in the form $(A + BT)$, has been estimated by considering equilibrium of the phase with the liquid at the temperatures 845 and 586 °C, corresponding to the liquid compositions of 17.3 and 52.1 at.% Sr, respectively. In a similar manner, the molar free energy of CuSr has been estimated from the least-squares fitting of the liquidus data at 586, 576.5, 550, and 507 °C, corresponding to the compositions 52.1, 60.0, 69.75, and 79.5 at.% Sr, respectively. The results for both phases are presented in Table 5.

Table 5 Thermodynamic Properties of Phases in the Cu-Sr System

All parameters expressed in J/mol

Lattice stability parameters(a)

$^{0}G_{Cu}^{L} = 0$
$^{0}G_{Sr}^{L} = 0$
$^{0}G_{Cu}^{(Cu)} = -13\,054 + 9.613\,T$
$^{0}G_{Sr}^{(\beta Sr)} = -8276 + 7.95\,T$
$^{0}G_{Sr}^{(\alpha Sr)} = -9075 + 8.92\,T$
$(^{0}G^{\beta Sr \rightarrow \alpha Sr} = -799 + 0.97\,T)$

Integral molar Gibbs free energies(b)

$G^{L} = X(1 - X)(-20\,780 + 7.55\,T) + RT[X \ln X + (1 - X)\ln(1 - X)]$

where X is atomic fraction of Sr

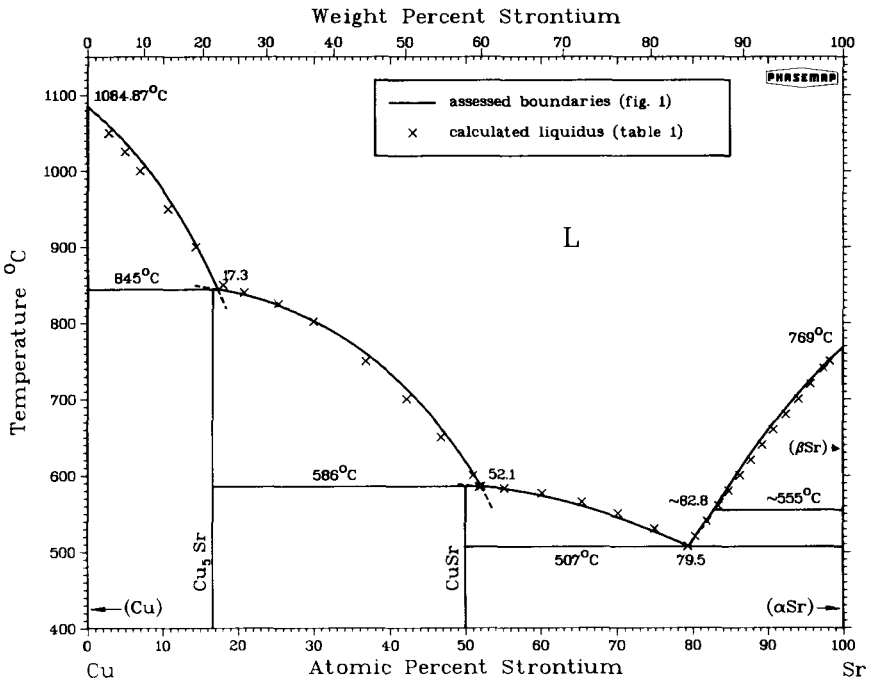
$G^{Cu_5Sr} = -18\,995 + 11.70\,T$
$G^{CuSr} = -34\,350 + 30.10\,T$

where mol for Cu_5Sr and CuSr refers to an atom as the elementary entity

Standard states: pure liquid Cu and pure liquid Sr.

(a) From [Hultgren; Elements]. (b) From the phase diagram [this work].

Fig. 2 Comparison of Assessed Phase Diagram and Thermodynamic Calculations for the Cu-Sr System



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

The calculated liquidus, based on these derived expressions and the lattice stability parameters for Cu and Sr, is shown in Fig. 2 and is tabulated in Table 1 for quantitative comparison with the experimental liquidus. The agreement between these results is very good, suggesting good consistency between the thermodynamic parameters of the different phases, as derived from the phase diagram.

The stability of Cu_5Sr and CuSr, with respect to the (Cu) and (Sr) phases, respectively, at lower temperatures, has been examined by considering the relative changes with temperature of the molar free energy of the four phases, as given in Table 5. The calculations have shown that the Cu_5Sr phase decomposes at about −28 °C into (Cu) and CuSr phases. The resultant eutectoid transformation is indicated in Fig. 1 and in Table 3. The hypothetical decomposition temperature for CuSr, however, occurred (in the metastable range) above its formation temperature, indicating this phase to coexist with (Cu) and (Sr) at all temperatures below ~−28 °C.

Suggestions for Future Experimental Work

The possible decomposition of the Cu_5Sr phase near −28 °C should be studied to help in checking the validity of the thermodynamic parameters for the different phases, as derived from the phase diagram in this work.

Using H-free, high-purity Sr (and Cu), the temperature of the $\beta Sr \rightleftarrows \alpha Sr$ invariant transformation in the Cu-Sr alloys should be determined.

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81BAP: "Melting Points of the Elements", *Bull. Alloy Phase Diagrams, 2*(1), 145-146 (1981), for melting point of elements corrected to conform to the 1968 ITPS scale. (Equi Diagram; Compilation)

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

Cu-Sr evaluation contributed by **D. J. Chakrabarti** and **D. E. Laughlin,** Department of Metallurgical Engineering and Materials Science, Carnegie-Mellon University, Pittsburgh, PA 15213, USA. Work was supported by the International Copper Research Association, Inc. (INCRA) and the Department of Energy through the Joint Program on Critical Compilation of Physical and Chemical Data coordinated through the Office of Standard Reference Data (OSRD), National Bureau of Standards. Thermodynamic calculations were done in part with the use of the program made available to the authors by Mr. E. S. K. Menon of Carnegie-Mellon University. The thermal analysis data on which the evaluation had been primarily based were made available in tabulated form by Prof. G. Bruzzone of the University of Genoa. Literature searched through 1982. Professor Laughlin and Dr. Chakrabarti are the ASM/NBS Data Program Co-Category Editors for binary copper alloys.