The Cu-Dy (Copper-Dysprosium) System

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Equilibrium Diagram

The equilibrium phases of the Cu-Dy system are: (1) the liquid, L, without any miscibility gaps; (2) the fcc terminal solid solution, (Cu), with negligible solid solubility of Dy in (Cu); (3) the Dy-rich bcc terminal solid solution, (βDy), based on the equilibrium phase of pure Dy between 1381 and 1412 °C; (4) the Dy-rich cph terminal solid solution (αDy), stable between –187 and 1381 °C; (5) the Dy-rich orthorhombic terminal solid solution, (α'Dy), stable below –187 °C; (6) the high-temperature hexagonal phase, Cu_7Dy, stable between 775 and 860 °C; (7) Cu_5Dy, present in two allotropic modifications (the lower temperature cubic form is stable up to ~930 °C, and the high-temperature hexagonal form is stable between 930 °C and the peritectic melting temperature of ~965 °C); (8) Cu_9Dy_2, with an unknown crystal structure and stable up to the congruent melting temperature of 970 °C; (9) Cu_7Dy_2, with an unknown crystal structure and stable between 855 and 905 °C; (10) the orthorhombic phase, Cu_2Dy, stable up to the congruent melting temperature of 890 °C; and (11) the most Dy-rich intermediate phase, CuDy, with a cubic structure and stable up to the congruent melting temperature of 955 °C.

[64Cop] investigated the Dy-rich region of the Cu-Dy system and proposed the formation of a eutectic between (αDy) and CuDy at ~775 °C. The solid solubility of Cu in Dy was estimated to be <0.2 wt.% Cu (<0.5 at.% Cu). Subsequently, [82Fra] investigated this system as a part of a systematic study of the Cu-RE phase diagrams. Alloys were prepared by melting 99.9% pure Dy and 99.999% pure Cu under pure argon in an in-

Fig. 1 Assessed Cu-Dy Phase Diagram

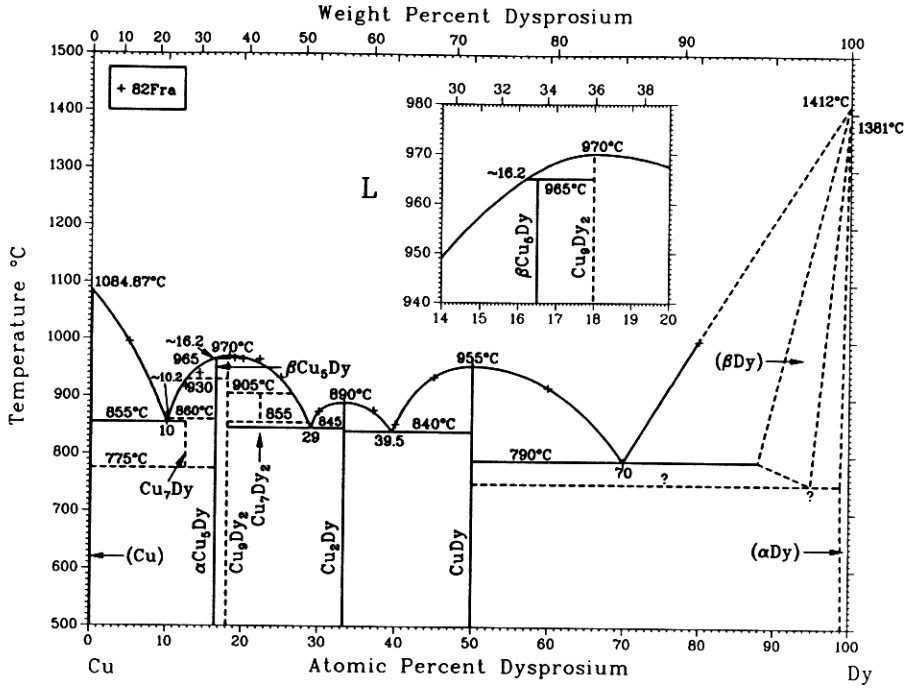

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Table 1 Cu-Dy Experimental Liquidus Data

Composition, at.% Dy	Temperature, °C	Composition, at.% Dy	Temperature, °C
5.0	995	30.0	874
10.0	855	33.3	890
12.5	916	37.1	877
14.3	940	40.0	853
18.2	970	45.0	935
18.9	966	50.0	955
20.0	965	60.0	918
22.2	965	70.0	790
25.0	933	80.0	1000

From [82Fra].

duction furnace. The alloys were examined by differential thermal analysis (DTA), metallography, and X-ray analysis. Thermal analyses were performed with heating and cooling rates of 20, 10, and 5 °C/min. Because elemental lattice parameters did not change significantly on alloying, [82Fra] concluded that terminal solid solubilities are negligible in the Cu-Dy system. Also, [82Fra] observed the congruent formation of Cu_9Dy_2, Cu_2Dy, and CuDy and the peritectic formation of Cu_7Dy, Cu_5Dy, and Cu_7Dy_2. Four eutectics were reported: Cu-Cu_7Dy (855 °C, 10 at.% Dy); Cu_9Dy_2-Cu_2Dy (845 °C, 29 at.% Dy); Cu_2Dy-CuDy (840 °C, 39.5 at.% Dy); and CuDy-(αDy) (790 °C, 70 at.% Dy). In addition, [82Fra] reported the occurrence of a thermal effect at ~815 °C in alloys containing ~37.1 to 50 at.% Dy, and have attributed it to a separation in the Cu_2Dy-CuDy eutectic. A similar effect was observed in the Cu-Gd system by [83Car1].

[82Che] also investigated the Cu-Dy system by means of X-ray diffraction and DTA. Their alloys were prepared by melting 99.9% pure Dy and 99.999% pure Cu under argon in an induction furnace, followed by heat treatment at temperatures between 740 and 800 °C for 2 to 3 weeks. DTA studies were conducted at a heating and cooling rate of 10 °C/min. [82Che] reported the peritectic formation of Cu_7Dy, Cu_2Dy, and CuDy and the congruent formation of Cu_5Dy. In addition, the authors reported the formation of two eutectics: Cu-Cu_7Dy (880 °C, ~7.4 at.% Dy) and CuDy-Dy (808 °C, ~74.1 at.% Dy). The solid solubility of Dy in Cu is reported to be negligible. On the other hand, the solid solubility of Cu in Dy is estimated to be <0.2 wt.% Cu (<0.5 at.% Cu) at room temperature, and ~5 wt.% Cu (~11.9 at.% Cu) at the eutectic temperature of 808 °C. This latter value could be the solubility of Cu in the bcc phase of Dy (βDy), similar to that observed for (βGd) in the Cu-Gd system by [83Car1].

The reports of [82Fra] and [82Che] are in agreement with regard to the existence of Cu_5Dy, Cu_2Dy, and CuDy, along with the high-temperature stability of Cu_7Dy. In addition, melting points reported for Cu_5Dy

Table 2 Special Points of the Assessed Cu-Dy Phase Diagram

Reaction	Compositions of the respective phases, at.% Dy			Temperature °C	Reaction type	Reference
(Cu) ↔ L		0.0		1084.87	Melting point	[Melt]
L ↔ (Cu) + Cu_7Dy	10.0	~0	12.5	855	Eutectic	[82Fra]
L + Cu_5Dy ↔ Cu_7Dy	~10.2	16.67	12.5	860	Peritectic	[82Fra](a)
Cu_7Dy ↔ Cu + Cu_5Dy	12.5	~0	16.67	775	Eutectoid	[82Fra]
L + Cu_9Dy_2 ↔ Cu_5Dy	~16.2	18.18	16.67	965	Peritectic	[82Fra](a)
L ↔ Cu_9Dy_2		18.18		970	Congruent	[82Fra]
Cu_9Dy_2 + L ↔ Cu_7Dy_2	18.18	~26.7	22.22	905	Peritectic	[82Fra](a)
Cu_7Dy_2 ↔ Cu_9Dy_2 + L	22.22	18.18	~28.7	~855	Catatectic	[82Fra](a)
L ↔ Cu_9Dy_2 + Cu_2Dy	29.0	18.18	33.33	845	Eutectic	[82Fra]
L ↔ Cu_2Dy		33.33		890	Congruent	[82Fra]
L ↔ Cu_2Dy + CuDy	39.5	33.33	50.0	840	Eutectic	[82Fra]
L ↔ CuDy		50.0		955	Congruent	[82Fra]
L ↔ CuDy + (αDy)	70.0	50.0	~88.1	790	Eutectic	[82Fra](b)
(αDy) ↔ (βDy)		~100		1381	Allotropic	[79Bea, 86Gsc]
(βDy) ↔ L		100		1412	Melting point	[79Bea, 86Gsc]

(a) Liquidus composition was obtained by interpolation of experimental data in Fig. 1. (b) Composition of (βDy) is from [82Che].

Table 3 Cu-Dy Experimental Lattice Parameters

Phase	Crystal structure	Lattice parameters, nm a	b	c	Reference
Cu_7Dy	Hexagonal	0.4932	...	0.4156	[71Bus](a)
Cu_6Dy	Cubic	0.7027	...	...	[69Bus]
		0.7022	...	...	[82Fra]
Cu_9Dy_2	(b)	0.4999	...	1.394	[82Fra]
$Cu_{23}Dy_6$	Cubic	1.214	...	...	[84Tsv](c)
Cu_2Dy	Orthorhombic	0.4300	0.6792	0.7300	[63Sto]
		0.4303	0.6802	0.7289	[82Fra]
CuDy	Cubic	0.3461	...	...	[61Bae]
		0.3460	...	...	[64Cha]
		0.3444	...	...	[65Pie]
		0.3461	...	...	[65Dwi]
		0.3455	...	...	[71Bel]
		0.3455	...	...	[82Fra]

(a) High-temperature phase; reported by [82Fra] to be stable between 775 and 860 °C. (b) Structure based on a tetragonal cell. (c) Th_6Mn_{23} type; formed only at high pressures (7.7 Gpa).

and Cu_2Dy are in close accord. However, [82Fra] reported the peritectic formation of Cu_5Dy and the congruent formation of Cu_9Dy_2, Cu_2Dy, and CuDy. In contrast, [82Che] observed the congruent formation of Cu_5Dy and the peritectic formation of both Cu_2Dy and CuDy. Moreover, [82Che] did not observe the phases Cu_9Dy_2 and Cu_7Dy_2. Because there is no other independent phase diagram investigation to resolve the conflicting data of [82Fra] and [82Che], one has to rely on systematics of alloying behavior of Cu with the heavy lanthanides [85Gsc] (see also "The Copper-Rare Earth Systems," in this issue). In this context, the pattern of Cu-Dy phase relationships reported by [82Fra] closely follows that reported for Cu-Gd [83Car1], Cu-Er [70Bus], and Cu-Y [81Cha]. As such, the assessed phase diagram for the Cu-Dy system, shown in Fig. 1, is drawn from the data reported by [82Fra]. The solubility values of Cu in (αGd) at room temperature and in (βGd) at the eutectic temperature are tentatively accepted from [82Che]. Elemental melting points have been adjusted in accordance with the accepted values listed in [Melt] for Cu and in [78Bea] and [86Gsc] for Dy. [82Fra] did not indicate the (αDy) ↔ (βDy) transformation temperature in their phase diagram, and the accepted value in Fig. 1 is from [78Bea] and [86Gsc].

Table 1 lists the experimental data for the Cu-Dy liquidus boundaries from [82Fra]. The various invariant reactions observed for the Cu-Dy system are summarized in Table 2. The various eutectic and congruent melting temperatures in Table 2 are in good agreement with the systematics observed for the corresponding temperatures in the other Cu-lanthanide

Table 4 Cu-Dy Crystal Structure Data

Phase	Composition, at.% Dy	Pearson symbol	Space group	Strukturbericht designation	Prototype
(Cu)	0	*cF4*	$Fm\bar{3}m$	*A1*	Cu
Cu_5Dy(HT)	~16.67	*hP6*	*P6/mmm*	$D2_d$	$CaCu_5$
Cu_5Dy(LT)	~16.67	*cF24*	$F\bar{4}3m$	$C15_b$	$AuBe_5$
Cu_2Dy	~33.3	*oI12*	*Imma*	...	$CeCu_2$
CuDy	~50	*cP2*	$Pm\bar{3}m$	*B2*	CsCl
(α'Dy)	100	*oC4*	*Cmcm*	...	α'Dy
(αDy)	100	*hP2*	$P6_3/mmc$	*A3*	Mg
(βDy)	100	*cI2*	$Im\bar{3}m$	*A2*	W

Table 5 Cu-Dy Lattice Parameter Data

Phase	Composition, at.% Dy	Lattice parameters, nm *a*	*b*	*c*	Comment	Reference
(Cu)	0	0.36146	...	...	At 25 °C	[Massalski]
Cu_5Dy(HT)	~16.67	0.502	...	0.408	(a)	[83Car2]
Cu_5Dy(LT)	~16.67	0.7025	...	...	...	[69Bus, 82Fra]
Cu_2Dy	~33.3	0.4300	0.6792	0.7300	...	[65Sto]
CuDy	~50	0.3455	...	...	...	(c)
(α'Dy)	100	0.3595	0.6184	0.5678	At −187 °C	[78Bea, 86Gsc]
(αDy)	100	0.35915	...	0.56501	At 25 °C	[78Bea, 86Gsc]
(βDy)	100	0.398	...	...	(b)	[78Bea, 86Gsc]

(a) Lattice parameters were estimated by [83Car2] from the systematics of crystallographic data for the Cu-lanthanide systems. (b) Determined by extrapolation to 0% solute of *a* vs composition data for Dy-Mg alloys. (c) [61Bae, 64Cha, 65Pie, 65Dwi, 71Bel, 82Fra].

systems (see "The Copper-Rare Earth Systems," in this issue).

Metastable Phases

[79Mcg] prepared amorphous thin films with the composition $Cu_{0.55}Dy_{0.45}$ by sputtering from arc-melted targets, and by thermal evaporation from Cu and Dy sources, followed by deposition on liquid nitrogen-cooled sapphire substrates. The films so obtained were 500 to 1000 nm thick.

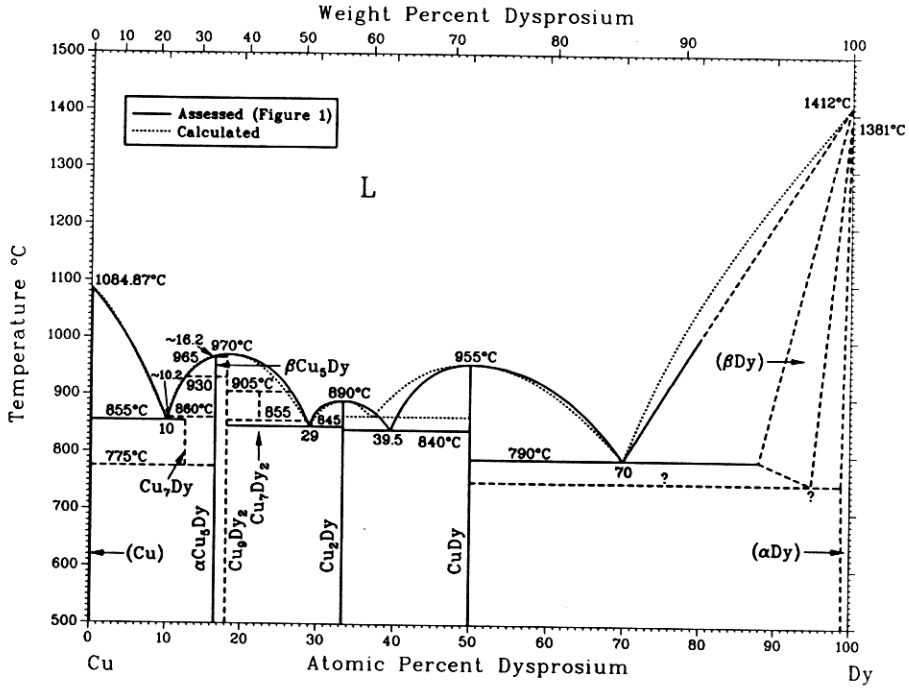
Crystal Structures and Lattice Parameters

[71Bus] reported the high-temperature formation of Cu_7Dy, with its subsequent decomposition to Cu_5Dy and elemental Cu on annealing at temperatures near 700 °C. From X-ray diffraction and density measurements on Cu_7Tb, [71Bus] concluded that Cu_7RE phases (RE = Gd, Tb, Dy, Y) form with a structure closely related to that of the hexagonal $CaCu_5$ type, with 22% of the Ca sites substituted at random by pairs of Cu atoms, resulting in the actual stoichiometry $Cu_{5.44}RE_{0.78}$. [71Bus] have not reported the temperature range of existence of Cu_7Dy, but the phase is shown to be unstable below 700 °C. Although [82Fra] could not prepare Cu_7Dy as a single phase through normal melting techniques, thermal analysis and metallography of alloys containing 10 to 14.3 at.% Dy revealed that Cu_7Dy forms peritectically from the liquid and Cu_5Dy at ~860 °C, and subsequently decomposes eutectoidally at ~775 °C.

[69Bus] reported the occurrence of the cubic $AuBe_5$-type structure in both as-cast and annealed alloys with the stoichiometry Cu_5Dy. In addition, [69Bus] mentioned the existence of Cu_5Dy with the hexagonal $CaCu_5$-type structure at high temperatures. There are no experimental lattice parameter data for the hexagonal $CaCu_5$ form. However, [83Car2] predicted its lattice parameters to be $a = 0.502$ nm and $c = 0.408$ nm. These values were determined by [83Car2] on the basis of the systematic variation observed for the cube root of the unit formula volume with respect to the ionic radii of the lanthanide element. The phase diagram of [82Fra] shows a thermal effect at ~930 °C corresponding to the stoichiometry of Cu_5Dy. This thermal effect can be attributed to the change in crystal structure of Cu_5Dy from the cubic $AuBe_5$ type to the hexagonal $CaCu_5$ type.

[82Fra] observed the occurrence of a phase at ~18.2 at.% Cu and assigned it a stoichiometry Cu_9Dy_2 on the basis of thermal analysis and metallography. Preliminary X-ray measurements on platelike crystals of Cu_9Dy_2 revealed a tetragonal symmetry with lattice parameters $a = 0.4999$ nm and $c = 1.394$ nm (minimum). Although no structural details were obtained, [82Fra] observed that the powder patterns of Cu_9Dy_2

Fig. 2 Assessed vs Calculated Cu-Dy Phase Diagram



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were similar to those of Cu_9Gd_2 [83Car1] and Cu_9Yb_2 [71Ian]. In addition, [82Fra] determined that Cu_9Dy_2 and the $AuBe_5$-type cubic Cu_5Dy are dimensionally related with $a(Cu_9Dy_2) = a(Cu_5Dy)/2^{1/2}$ and $c(Cu_9Dy_2) = 2a(Cu_5Dy)$.

[82Fra] detected the occurrence of a phase with the approximate stoichiometry Cu_7Dy_2 at ~22.3 at.% Dy on the basis of thermal effects only. This phase was found to exist with a limited temperature range of stability and formed peritectically at ~905 °C, with subsequent decomposition at ~855 °C.

The phases Cu_2Dy and CuDy formed congruently at 890 and 955 °C, respectively. Cu_2Dy formed with the orthorhombic $CeCu_2$ structure, whereas the equiatomic phase CuDy formed with the cubic CsCl structure.

[84Tsv] obtained a phase with the stoichiometry $Cu_{23}Dy_6$ by the application of a constant pressure of 7.7 GPa at high temperatures to a stoichiometric mixture of the constituent elements. The resulting phase was indexed on the basis of the cubic Th_6Mn_{23} structure with space group $Fm\bar{3}m$ and with a lattice parameter a = 1.214 nm.

Table 3 lists the experimental values for the lattice parameters of the various Cu-Dy phases. Crystal structure data pertinent to the Cu-Dy system are summarized in Tables 4 and 5.

Thermodynamics

No experimental thermodynamic data are available for the Cu-Dy system. In the present modeling, the integral Gibbs energy of mixing of the liquid was derived from the experimental eutectic data of [82Fra] (10.0 at.% Dy, 855 °C and 70 at.% Dy, 790 °C), with the assumptions that mutual solid solubilities are negligible and that the liquid behaves like a subregular solution. The elemental lattice stability parameters used in the calculation are listed in Table 6. The Gibbs energies of formation of Cu_5Dy, Cu_9Dy_2, Cu_2Dy, and CuDy were then determined from the various invariant temperatures, as well as the Gibbs energy of mixing of the liquid. In all instances, the phases were assumed to be line compounds. Table 6 shows the values of the various parameters.

The calculated phase boundaries, derived from the Gibbs energy functions in Table 6, are compared in Fig. 2 with the experimental phase boundaries. In general, the calculated liquidus is in good accord with the experimental data, with the exception of the region between Cu_2Dy and CuDy. The calculated Cu_2Dy + L/L and L/L + CuDy liquidus is shifted with respect to the experimental data toward higher Cu contents. The resultant Cu_2Dy-CuDy eutectic is estimated to lie at 37.1 at.% Dy and 858 °C, as compared to the experimental data of 39.5 at.% Dy and 840 °C. This deviation, however, is not serious, and the ther-

Table 6 Thermodynamic Properties of Cu-Dy Phases

Lattice stability parameters for Cu(a)

$G^0(Cu, L) = 0$

$G^0(Cu, fcc) = -13\,054 + 9.613\,T$

Lattice stability parameters for Dy(b)

$G^0(Dy, L) = 0$

$G^0(Dy, bcc) = -11\,060 + 6.563\,T$

$G^0(Dy, cph) = -15\,220 + 9.078\,T$

Integral molar Gibbs energies(c)

$G(L) = X(1-X)(-87\,555 + 43\,352\,X) + RT[X \ln X + (1-X) \ln (1-X)]$

$\Delta_f G(Cu_7Dy) = -12\,731 + 0.10\,T$

$\Delta_f G(Cu_5Dy) = -18\,965 + 2.56\,T$

$\Delta_f G(Cu_9Dy_2) = -26\,113 + 7.53\,T$

$\Delta_f G(Cu_2Dy) = -21\,732 + 0.57\,T$

$\Delta_f G(CuDy) = -36\,654 + 10.67\,T$

Note: Standard states: pure liquid Cu and pure liquid Dy. Gibbs energies are expressed in J/mol, and temperatures are in K. X is the atomic fraction of Dy. Mol refers to the atom as the elementary entity.
(a) From [Hultgren,E]. **(b)** From [83Cha]; melting and transformation temperatures are from [78Bea] and [86Gsc]. **(c)** From the phase diagram [this work].

modynamic functions of Table 6 are considered adequate to reproduce the Cu-Dy equilibrium diagram.

The enthalpy data from the present thermodynamic analysis are compared in Table 7 with the enthalpies of formation evaluated with the semi-empirical model of [80Mie] and [83Nie]. In almost all instances, the Miedema model predicts enthalpy values that are much more exothermic than the values obtained in the present calculation.

Table 7 Calculated Enthalpies of Formation of Cu-Dy Intermediate Phases vs Theoretical Estimates Based on Miedema's Model

Phase	Enthalpy of formation, kJ/mol Present modeling	Miedema model(a)
Cu_7Dy	-12.7	-27.5
Cu_5Dy	-19.0	-32.2
Cu_9Dy_2	-26.1	-33.9
Cu_2Dy	-21.7	-46.1
CuDy	-36.7	-46.7

Note: Standard states are liquid Cu and liquid Dy.
(a) From [83Nie].

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*Indicates key paper.

#Indicates presence of a phase diagram.

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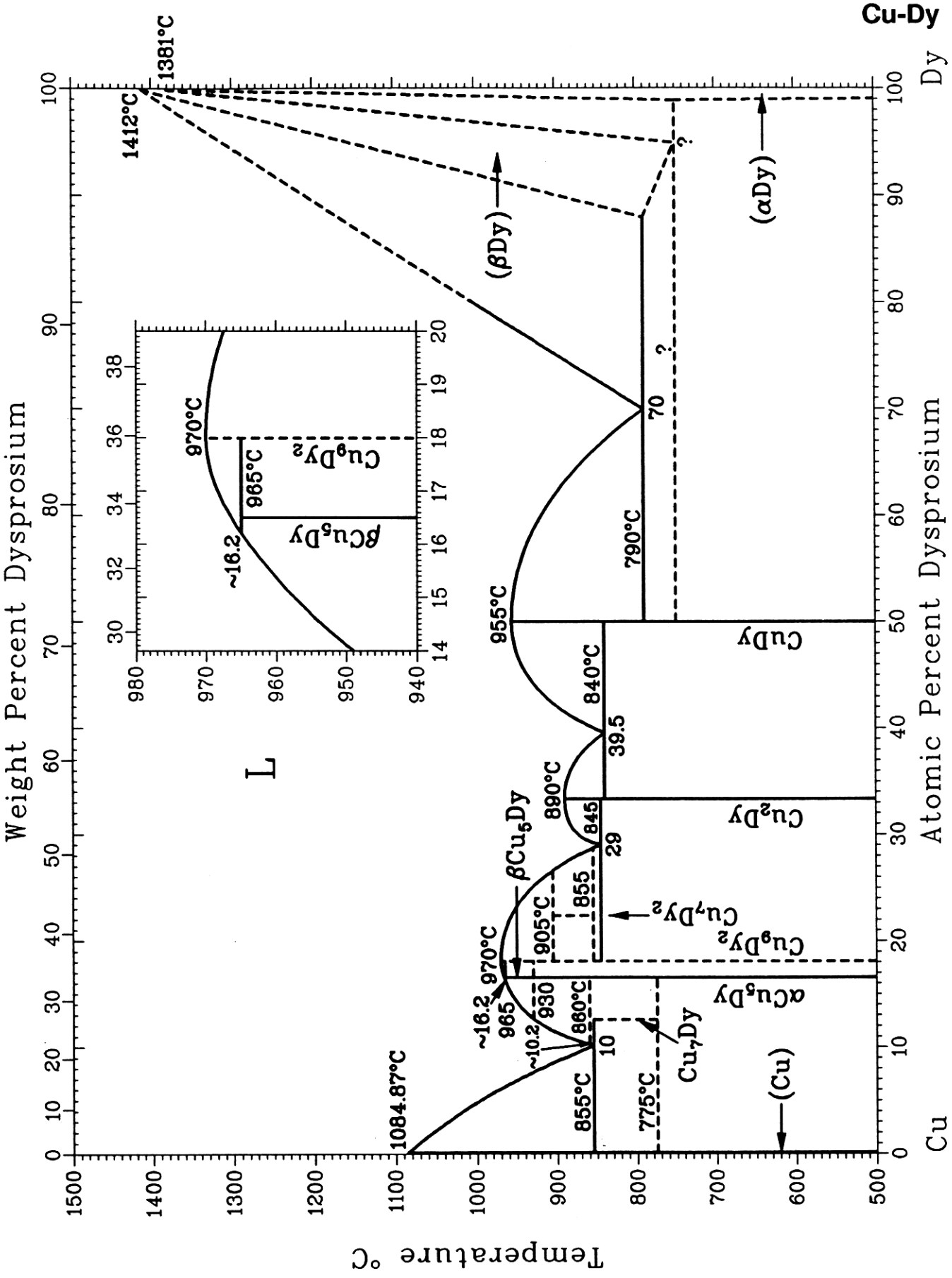

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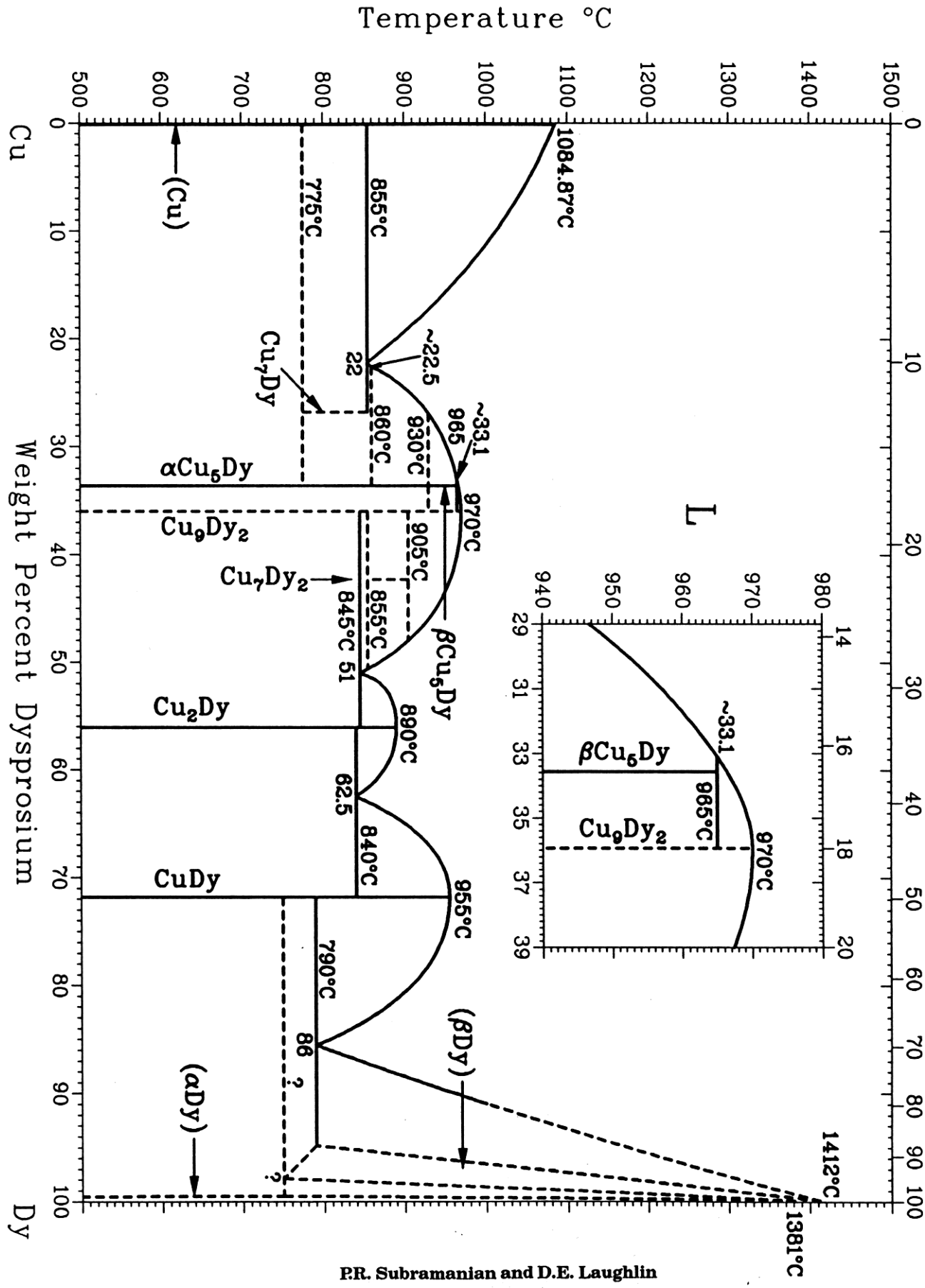

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