

# Ternary Rare-Earth Aluminum Systems With Copper: A Review and a Contribution to Their Assessment

P. Riani, L. Arrighi, R. Marazza, D. Mazzone, G. Zanichchi, and R. Ferro

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Constitutional data of the R-Cu-Al systems, with R = rare earth, are summarized and discussed. Crystal structures of the phases formed in the binary boundary systems and in the ternary systems are assessed, and the phase equilibria observed in the reported ternary systems are reviewed with special attention given to the isothermal and polythermal sections of the phase diagrams. Some regularities observed in the trends of the constitutional properties of the ternary R-Cu-Al alloys are briefly described.

## 1. Introduction

Several R-T-Al systems (R = rare earth and T = transition element) have been the object of experimental investigations and reviews due to their general characteristics and properties and to their present or potential application. Their isothermal sections generally present a high number of intermediate phases. The crystal structures of these phases have been studied by several researchers, and some criteria have been defined for their rational descriptions. In the framework of an approach to the systematics of the rare-earth ternary alloys, a review of the R-Cu-Al systems is presented here. From a technological point of view, the Al-rich alloys could be especially interesting: the beneficial effects of Cu-additions (for instance, with 4-5 at.% Cu are heat-treatable alloys that can reach quite high strengths and ductility, depending on the contents of other minority elements) and rare-earth additions (which improve tensile strength, heat and corrosion resistance and extrudability) have been described [2000Kea and 2000Gsc]. Literature, however, seems to be lacking with regard to properties and applications of Al-alloys containing small additions of both Cu and rare-earth metals. One special point has been discussed, concerning the glass-forming capability of R-Cu-Al alloys. A detailed description and discussion has been reported by [1997Ino] in the framework of a general presentation of amorphous alloys based on R-T-Al systems (T = transition metal, such as Fe, Co, Ni, Cu) (see also [1989Cah]) These alloys form glasses over wide compositional ranges (see Fig. 1). In the course of a systematic investigation of several groups of ternary R alloys, the study of the R-Cu-Al systems has now been started in our laboratory. As a first contribution, a critical review of the relevant literature is reported here. In a subsequent paper experimental results obtained on selected R-Cu-Al alloys will be reported.

P. Riani, L. Arrighi, R. Marazza, D. Mazzone, G. Zanichchi, and R. Ferro, Sezione di Chimica Inorganica e Metallurgia—Dipartimento di Chimica e Chimica Industriale via Dodecaneso 31, 16146—Genova (Italy). Contact e-mail: ferro@chimica.unige.it.

## 2. Binary Boundary Systems

### 2.1 Aluminum-Copper System

The Al-Cu system has been studied by several authors. All the results up to 1983 concerning phase equilibria, transformations, crystal structures, metastable phases, and thermodynamic data were reported in the assessment by Murray [1994Mur]. Due to the complexity of the phase diagram, many uncertainties still remained in the assessed version.

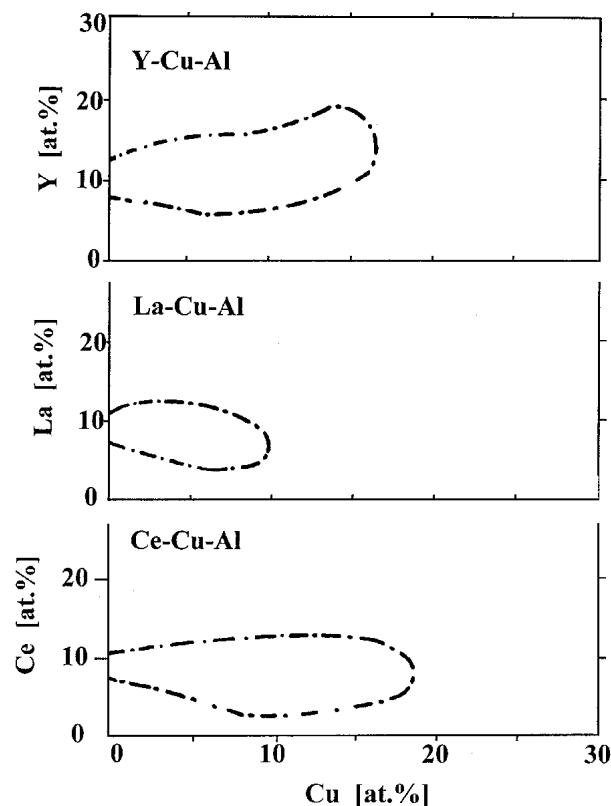


Fig. 1 Compositional ranges for formation of an amorphous phase in Al-Cu-Y, Al-Cu-La and Al-Cu-Ce systems (from [1997Ino])

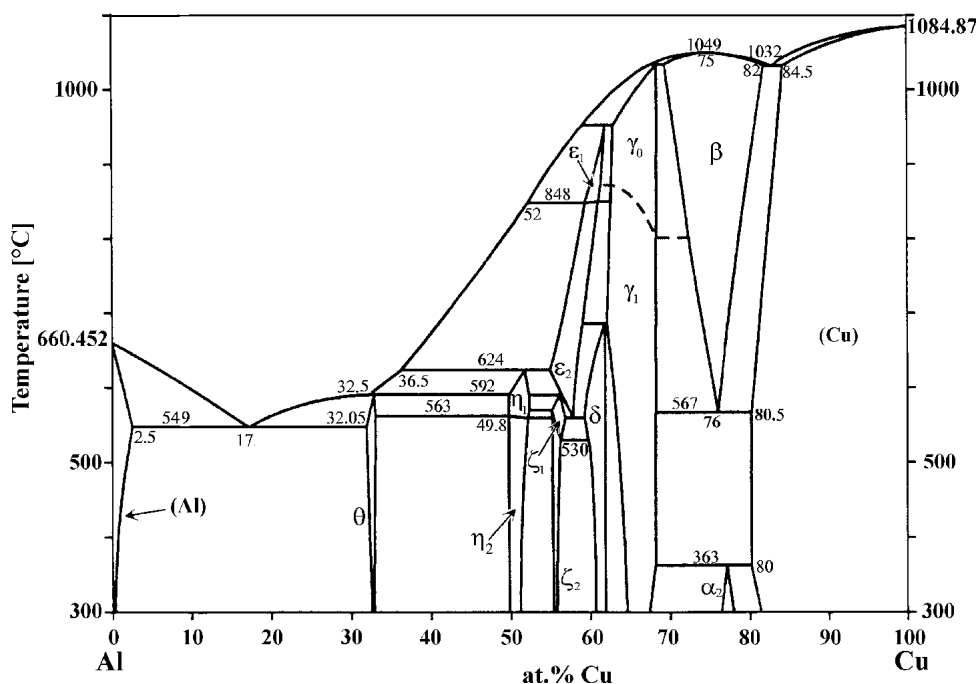


Fig. 2 Al-Cu phase diagram (after [1998Liu] with modifications from [1996God])

After 1983, phase equilibria between solid alloys and liquid were measured in the Al-rich region by [1984Ben] (maximum solid solubility of Cu in Al 2.87 at.% Cu) and by [2000Lia] (2.7 at.% Cu), while phase transformations in several Cu-12.4%Al alloys were studied with differential thermal analysis (DTA) by [1984Boj]. Gödecke and Sommer [1996God] re-determined the phase equilibria in the composition range between 31 and 37.5 at.% Cu by DTA, magneto-thermal analysis, magnetic susceptibility measurements versus temperature (MTA), and optical microscopy. A new solubility range is proposed [1996God] for the  $\text{Al}_2\text{Cu}$  ( $\theta$ ) phase, which is formed by a peritectic reaction at 592 °C: from 32.05 to 32.6 at.% Cu at 549 °C (the largest homogeneity range) and from 32.4 to 32.8 at.% Cu at 250 °C. Moreover [1996God] studied the metastable  $\text{Al}_2\text{Cu}_{(m)}$  phase with composition of 32.75 at.% Cu and the respective metastable phase equilibria, and [1996Che] determined the solidification curves in the Al-rich Al-Cu system.

Liu et al. [1998Liu] studied the phase equilibria in the temperature range 500-1000 °C and in the composition range 40-85 at.% Cu using a diffusion couple technique, differential scanning calorimetry (DSC), and high-temperature x-ray diffraction (XRD). Significant differences in the phase equilibria in the composition range 60-75 at.% Cu were found: the  $\beta_0$  phase does not exist at ~1000 °C in the composition range near to 70 at.% Cu; the transformation observed in the temperature range 800-900 °C in the composition range 62-68 at.% Cu is not a first-order reaction but a kind of second-order ordering reaction between  $\gamma_1$  ( $\text{D8}_3\text{-Cu}_9\text{Al}_4$  type) and  $\gamma_0$  ( $\text{D8}_2\text{-Cu}_5\text{Zn}_8$  type); the temperature of the eutectoid reaction  $\beta \rightarrow \alpha + \gamma_1$  is  $559 \pm 1$  °C and the invariant reactions  $\gamma_0 \rightarrow \beta + \gamma_1$  and  $\gamma_0 + \varepsilon_1 \rightarrow \gamma_1$  were not confirmed. The structure of alloys in the  $\beta + \gamma_0$  region

was also studied by Hurtado et al. [1998Hur]. The version of the Al-Cu phase diagram by [1998Liu] has also been reported by Okamoto [2000Oka]; Fig. 2 is based on this version, slightly corrected according to [1996God]. The stable phases formed in this system are listed in Table 1.

The Al-Cu phase diagram was calculated theoretically for the first time [1978Kau] by using the Calphad method but considering only four of the eleven intermetallic phases. Later [1991Che] considered six intermetallic phases and obtained an improved diagram, but it was still too simple to describe the complexity of the actual Al-Cu phase diagram. Subsequently, further thermodynamic assessment was reported by [1998Sau].

The following miscellaneous data on the Al-Cu system may be noteworthy.

Different authors have experimentally and theoretically studied the behavior under pressure of Al-rich alloys; for instance, [1995Som] studied the formation of Al-Cu solid solution and bulk properties and Kagaya et al. studied for Al-Cu alloys the specific heat at constant volume [1996Kag] and the solidus curve under pressure [2000Kag, 2001Kag]. Al-Cu alloys were investigated in a high-pressure apparatus (2.2 and 3 GPa) by [1993Yam], who carried out a calculation of the shifting of phase boundaries under pressure on the basis of the molar volumes of the phases.

Zrudsky et al. [1971Zru] measured the specific heats of a series of copper-rich Al-Cu samples over the temperature range 1-14 K while Lee and Sommer [1985Lee] determined the partial enthalpies of mixing of liquid Al-rich alloys at 1191 K using a high-temperature solution calorimeter.

Meetsma et al. [1989Mee] refined the crystal structure of the  $\text{Al}_2\text{Cu}$  compound by means of single-crystal x-ray dif-

Table 1 Al–Cu System: Solid Phases

Phase	Composition, at.% Cu	Temperature Range, °C	Space Group	Pearson Symbol and Prototype	Notes	Lattice Parameters, pm		
						<i>a</i>	<i>b</i>	<i>c</i>
(Al)	0 to 2.5	<660.452	<i>Fm</i> $\bar{3}m$	<i>cF4</i> -Cu	at 0 at.% Cu (T = 298 K)	404.96	...	...
$\theta$	32.05 to ~32.6(a)	<592	<i>I4/mcm</i>	<i>tI12</i> -Al <sub>2</sub> Cu	at ~33 at.%	606.7	...	487.7
$\eta_1$	49.8 to 52.4	624-560	<i>Pbn</i> or <i>Cmmm</i>	<i>0</i> *32	at 49.8 at.%	410	1202.4	865
$\eta_2$	49.8 to 52.3	<563	<i>C2/m</i>	<i>mC20</i>	at 49.8 at.% $\beta = 55.04^\circ$	1206.6	410.5	691.3
$\zeta_1$	55.2 to 56.8	590-530	<i>Fmm2</i> (b)	<i>oF88-4.7</i>	at ~57 at.%	812.7	1419.9	999.3
$\zeta_2$	55.2 to 56.3	<570	<i>Imm2</i> (c)	<i>oI24-3.5</i>	at ~56 at.%	409.7	703.1	997.9
$\varepsilon_1$	59.4 to 62.1	958-850	...	<i>c</i> **	...	...	...	...
$\varepsilon_2$	55.0 to 61.1	850-560	<i>P6<sub>3</sub>/mmc</i>	<i>hP6-x</i> -Ni <sub>2</sub> In	at ~57.4 at.%	414.6	...	506.3
$\delta$	59.3 to 61.9	<686	<i>R</i> $\bar{3}m$	<i>hR</i> *	...	...	...	...
$\gamma_0$	63 to 69	~1040-800	<i>I</i> $\bar{4}3m$	<i>cI52</i> -Cu <sub>5</sub> Zn <sub>8</sub>	[1998Liu]	...	...	...
$\gamma_1$	62.5 to 69	<-900	<i>P</i> $\bar{4}3m$	<i>cP52</i> -Al <sub>4</sub> Cu <sub>9</sub>	at 68 at.% [1991Vil]	870.23	...	...
$\beta$	69.5 to 82.0	1049-567	<i>Im</i> $\bar{3}m$	<i>cI2</i> -W	at 75.7 at.%	294.6	...	...
$\alpha_2$	76.5 to 78	<-363	...	related to D0 <sub>22</sub>	at 76.4 at.%	366.8(b)	...	368.0(d)
...	...	...	...	structure	at 77.9 at.%	366.6	...	367.5
(Cu)	80.3 to 100	<1084.62	<i>Fm</i> $\bar{3}m$	<i>cF4</i> -Cu	at 85 at.% [1997Vil]	365.4	...	...
...	...	...	...	...	at 100 at.% (T = 298 K)	361.46	...	...

If not otherwise stated, the data are from [1994 Mur]

(a) data of composition and temperature range by [1996God], (b) [2002Gul] previously described as hexagonal ( $a_{\text{hex}} = a_{\text{ort}}$ ,  $c_{\text{hex}} = c_{\text{ort}}$ )

(c) [2002Gul], previously described as monoclinic with similar cell edges and  $\beta = 90.38^\circ$ .

(d) subcell data.

fraction. The structures of the  $\zeta_1$  and  $\zeta_2$  phases have been refined by [2002Gul]. Whiting and Tsakiroopoulos [1997Whi] investigated the growth and morphological evolution of the Cu-Al eutectoid by scanning electron microscopy (SEM), transmission electron microscopy (TEM), and confocal laser scanning microscopy (CLSM); they suggested that termination migration and conventional coarsening are the dominant mechanisms of morphological evolution in the temperature range 515-555 °C and concluded that the choice of the operative mechanism in a given grown lamellar structure is affected by both crystallography and interfacial structure.

A study of the influence of mechanical alloying on the lattice parameter is given in [1996Gri].

A comparison of Auger electron spectroscopy (AES) and extended x-ray absorption fine structure (EXAFS) analysis of a thin Al-Cu layer on Al substrate was presented by [1998Moz]. The influence of precipitates on plastic anisotropy in two-phase Al-Cu alloys was assessed and modeled by [1998Bar1]. The primary dendrite growth of Al-15% Cu alloy films between solid plates was investigated by [1999Miz]. A description of the behavior of Al-bronzes and brasses during deformation in a wide range of temperature and strain rates was given by [2001Gro].

A study of the growth rate of intermetallic compounds at the interface of cold roll bonded Al/Cu bimetal at 250 °C was carried out by [2001Abb].

A study of the intermetallic powders of Cu-Al system

obtained by the so-called self-disintegration method was carried out by [2001Bin]. Calculated phase diagrams of aluminum alloys from binary Al-Cu to multicomponent commercial alloys were presented by [2001Yan].

## 2.2 Aluminum-Rare Earth Systems

The phase diagrams of the R-Al (with R = Sc, Y, La, Ce, Pr, Nd, Sm, Gd, Dy, Ho, Er, and Yb) systems were studied by different authors. Literature data up to about 1988 regarding these systems were reported by Gschneidner and Calderwood [1988Gsc, 1989Gsc]. For R = Eu, Tb, Tm, and Lu, no experimental diagrams are available; an extrapolated version of the phase diagram of these systems, obtained on the basis of the systematic variation of thermodynamic properties across the lanthanide series, was suggested by [1993Fer] for Tb, [1991Oka] for Eu, and [1990Mas] for Tm and Lu. In the following, a summary of each system with updating is stated. For the R-Al systems with R = Y, Gd, Ho, and Yb, the phase diagrams reported by [1988Gsc, 1989Gsc] are still current, while for the systems with R = Sc, La, Ce, Pr, Eu, Tb, Dy, Tm, and Lu, new versions of the phase diagrams are collected in [2000Oka]. For the (Nd, Sm, Gd, Er)-Al systems, the assessed versions of the phase diagrams based on new literature data are shown in Fig. 3.

The crystal structure data for the R-Al phases are reported in Table 2. Indications about other stoichiometries and structures have been reported (for instance RAl<sub>4</sub> for

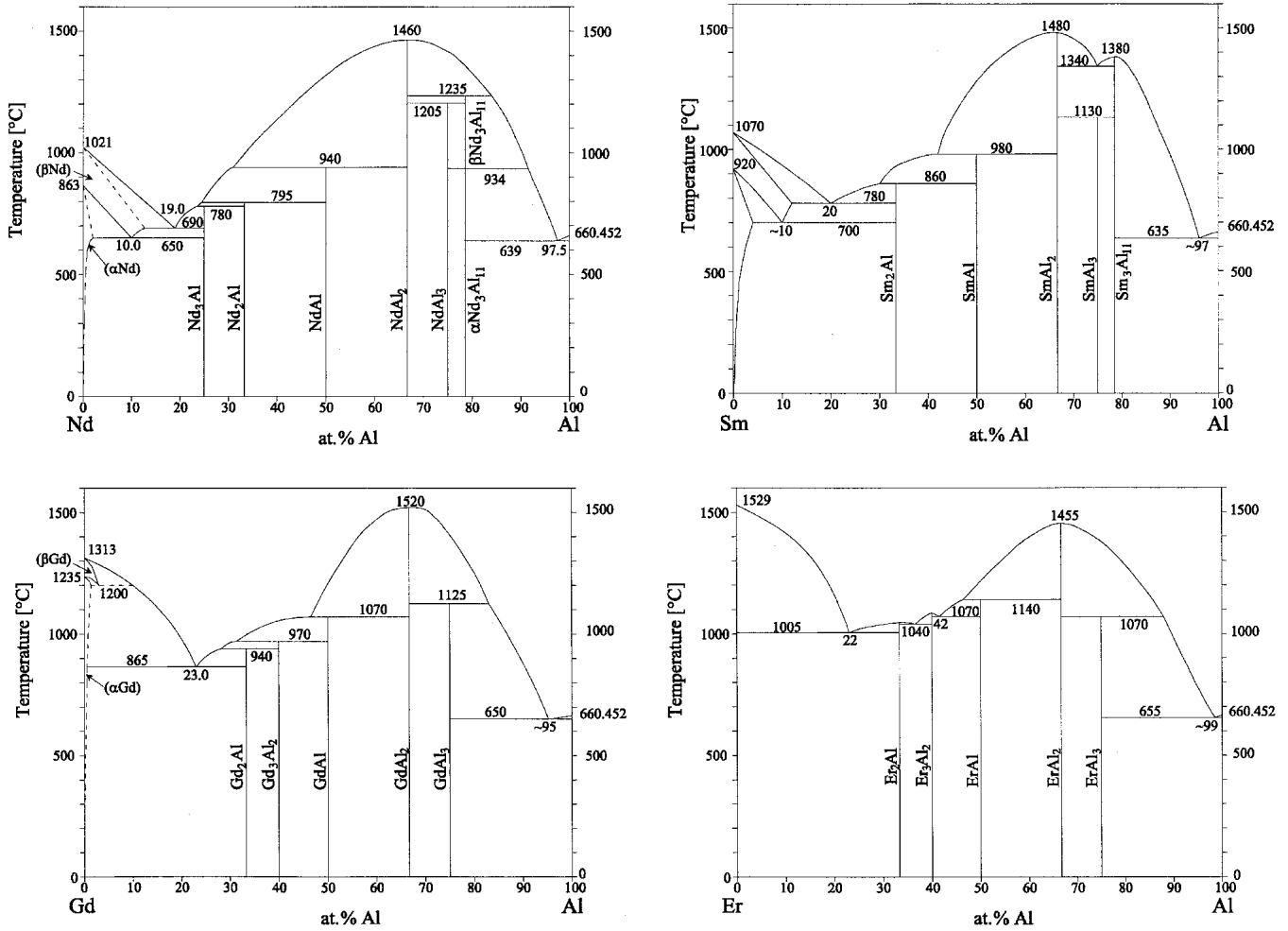


Fig. 3 R-Al phase diagrams (R = Nd, Sm, Gd, Er)

trivalent R,  $R_3Al$  for heavy R) but these have not been confirmed.

Several experimental thermodynamic data, as enthalpies of formation and specific heats of some R-Al intermetallic compounds, are reported in Table 3 and 4. The  $\Delta_f H$ , even if measured at a higher temperature, has been reported at 25 °C. The calculated enthalpies of formation of R-Al (R = La-Lu) alloys and intermetallic compounds have been reported by [1996Ouy]; the calculations were made on the basis of Miedema's semi-empirical theory. A review on the enthalpies of formation of binary Laves phases has been presented by [2002Zhu].

Moreover some other papers dealing with thermodynamics and magnetic properties may be cited, and these are listed system by system in the following.

**2.2.1 Sc-Al.** The phase diagram of this system was recently revised by [1999Cac] with a combination of experiments and thermodynamic calculations, and it was sketched by [2000Oka].

[1983Zvi] used mass spectrometry to study the heats of vaporization and vapor pressures of R-Al (R = Sc, Y, La, and Nd) alloys at temperatures less than or equal to 1727 °C. [1994Zub] measured the dissolution enthalpy of pure

solid scandium in liquid Al at 800 °C and in the mole fraction range of Sc up to  $10^{-2}$ . [1999Lu] calculated the activity of scandium in Al-Sc alloys and some thermodynamic functions ( $\Delta H$ ,  $G^E$ ,  $S^E$ ) of the alloys at 1460 °C. Later, [2001Ast] reported the results of a theoretical study on the structural, vibrational, and thermodynamic properties of solid-phase Al-Sc alloys based on first principle calculations of electronic free energies and ionic vibrational spectra.

[1997Zak] investigated the kinetics of decomposition of a solid solution of Sc in Al. [1997Nak] studied the age-hardening and precipitation process of  $Al_3Sc$  in an Al 0.23 wt.% Sc alloy by hardness measurements, electrical resistivity measurements, and TEM observations. [1998Fuk] carried out an XRD study and measurement of the compressive yield strength on  $Al_3Sc$  intermetallics and made TEM observations of the nature of dislocations. The solidification behavior of dilute Sc containing Al alloys has been investigated by [1998Nor], while a systematic creep study was undertaken for the binary intermetallic  $Al_3Sc$  in the temperature range 400-927 °C under a constant compressive stress ranging from 30-300 MPa by [2000Har]. More recently [2001Hyd] in-

Table 2 Crystal Structure of Phases Observed in the R–Al Systems With R = Rare Earth

Phases	Sc	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	Structural Type
R <sub>3</sub> Al	...	...	A <sup>HT</sup>	A <sup>LT</sup>	A <sup>LT</sup>	A	...	...	...	...	...	...	...	...	...	...	A <i>hP8</i> -Ni <sub>3</sub> Sn
...	...	...	...	B <sup>HT</sup>	B <sup>HT</sup>	...	...	...	...	...	...	...	...	...	...	...	B <i>cP4</i> -AuCu <sub>3</sub>
R <sub>2</sub> Al	C	D	...	...	D	D	D	...	D	D	D	D	D	?	...	D	C <i>hP6</i> -Ni <sub>2</sub> In
...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	D <i>oP12</i> -Co <sub>2</sub> Si or
...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	<i>oP12</i> -PbCl <sub>2</sub>
R <sub>3</sub> Al <sub>2</sub>	...	F	...	...	...	...	...	...	F	F	F	F	F	F	...	F	F <i>tP20</i> -Zr <sub>3</sub> Al <sub>2</sub> or
...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	<i>tP20</i> -Gd <sub>3</sub> Al <sub>2</sub>
...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	G <i>cP2</i> -CsCl
...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	J <i>oC8</i> -CrB
RAl	G	J	K	K	M <sup>LT</sup>	M	M	N	M	M	M	M	M	M	...	M	K <i>oC16</i> -CeAl
...	J(a)	...	...	...	K <sup>HT</sup>	...	...	...	...	...	...	...	...	...	...	...	M <i>oP16</i> -DyAl
...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	N <i>oP20</i> -EuAl
RAl <sub>2</sub>	O	O	O	O	O	O	O	O	O	O	O	O	O	O	O	O	O <i>oF24</i> -Cu <sub>2</sub> Mg
RAl <sub>2,4</sub>	...	...	P <sup>HT</sup>	...	...	...	...	...	...	...	...	...	...	...	...	...	P <i>hP3</i> -AlB <sub>2</sub>
...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	Q <i>cP4</i> -AuCu <sub>3</sub>
RAl <sub>3</sub>	Q	R <sup>LT</sup>	R	R	R	R	R	...	R	S	V <sup>LT</sup>	U	Q	Q	Q	Q	R <i>hP8</i> -Ni <sub>3</sub> Sn
...	...	S <sup>HT</sup>	...	...	...	...	...	...	S <sup>HP</sup>	U <sup>HP</sup>	U <sup>HT</sup>	Q <sup>HP</sup>	...	...	...	...	S <i>hR36</i> -BaPb <sub>3</sub>
...	...	...	...	...	...	...	...	...	...	...	Q <sup>HP</sup>	...	...	...	...	...	U <i>hR60</i> -HoAl <sub>3</sub>
...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	V <i>hP16</i> -Ni <sub>3</sub> Ti
R <sub>3</sub> Al <sub>11</sub>	...	...	W <sup>LT</sup>	W <sup>LT</sup>	W <sup>LT</sup>	W <sup>LT</sup>	Y	...	...	...	...	...	...	...	...	...	W <i>oI28</i> -αLa <sub>3</sub> Al <sub>11</sub>
...	...	...	Y <sup>HT</sup>	Y <sup>HT</sup>	Y <sup>HT</sup>	Y <sup>HT</sup>	...	...	...	...	...	...	...	...	...	...	Y <i>tI10</i> -BaAl <sub>4</sub> (Al-deficient)
RAl <sub>4</sub>	...	...	...	...	...	...	...	Z	...	...	...	...	...	...	...	...	Z <i>tI10</i> -BaAl <sub>4</sub>

HT = high temperature, LT = low temperature, HP = high pressure

(a) The crystal structure of the ScAl phase has been described either as *cP2*-CsCl type or *oC8*-CrB type, and the uncertainty still remains (see [1999Cac])

investigated the effect of cooling rate on the morphology of primary Al<sub>3</sub>Sc intermetallic particles in Al-Sc alloys using electron microscopy while [2001Mar] studied the nanoscale structural evolution of Al<sub>3</sub>Sc precipitates in Al(Sc) alloys. Finally the behavior of Al-Sc alloys in hydrogen at an equilibrium pressure of 0.1 MPa was analyzed by [2001Ant].

**2.2.2 Y-Al.** An assessed version of the Y-Al phase diagram was reported by [1989Gsc]. Subsequently [1990Kon] studied the Al-rich side of the diagram up to 4 at.% Y but found no significant updating to be necessary.

The heat of vaporization and vapor pressures of Y-Al alloys at temperatures less than or equal to 1700 °C were studied by means of mass spectrometry by [1983Zvi]. The partial enthalpies of mixing of liquid Al-rich alloys were determined at about 927 °C using a high-temperature solution calorimeter by [1985Lee]. In the thermodynamic calculation of the Y-Al-C system, Gröbner et al. [1995Gro] reviewed the literature data of the Y-Al system and thermodynamically optimized this system with the BINGSS program [1977Luk, 1992Luk].

[1994Fol] studied the supersaturation of the Al<sub>2</sub>Y Laves phase obtained by rapid solidification while [1994Al-k] studied the phase constitution in melt-spun Al-10 wt.% Y by XRD and analytical electron microscopy. Moreover [1995Nin] investigated the metastable extension of solid solubility of yttrium in aluminum by measurement of the lattice spacing of the rapidly solidified alloy, and [1995Lat] investigated its amorphous-crystalline phase transformation by means of XRD and DSC. [1996Nor] studied the magneto-resistance of Al<sub>90</sub>Y<sub>10</sub>, and [1998Che] studied the diffusion of La, Nd, Dy, Yb and Ga in Y-Al garnet.

**2.2.3 La-Al.** The phase diagram of the La-Al was sketched by [2000Oka] on the basis of the new experimental data of [1990Kon] and [1996Sac]; [1990Kon] studied the Al-rich side of the diagram up to 4 at.% La, while Saccone et al. [1996Sac] re-determined the phase diagram in the region between 65 and 100 at.% La using differential thermal analysis (DTA), metallographic analysis, quantitative electron probe microanalysis (EPMA), and x-ray examinations. [1971Dee] studied the low-temperature heat capacity of LaAl<sub>2</sub> in the temperature range 8-300 K while [1972Leb] carried out an electromotive force (emf) study of the thermodynamic properties of La-Al melts in the temperature range 680-850 °C. The enthalpies of formation of liquid binary La-Al alloys were investigated by [1981Esi], while the partial enthalpies of mixing of liquid Al-rich alloys were determined by [1985Lee] at about 927 °C using a high-temperature solution calorimeter. Moreover, mass spectrometry was used by [1983Zvi] to study the heat of vaporization and vapor pressure of La-Al alloys at temperatures less than or equal to 1727 °C. [1994Wan] optimized the La-Al system on the basis of the experimental thermodynamic values reported in literature, while [2001Cac] carried out a thermodynamic modeling and optimization of the La-Al system on the basis of the more recent literature information available.

The metastable extension of solid solubility of La in Al by measurement of the lattice spacings of rapidly solidified alloys were investigated by [1995Nin]. The suppression of superconductivity by C addition to La<sub>3</sub>Al was investigated by [1995Rav]. The magneto-resistance of La<sub>90</sub>Al<sub>10</sub> and its strong enhancement due to small crystalline precipitates

**Table 3 Enthalpies of Formation (at 25-30 °C) of Some Intermetallic R-Al Compounds**

Phase	$\Delta_f H^\circ$ , kJ/mol at	Measurement Method	Reference
Al <sub>3</sub> Sc	-59.8	Acid solution cal.	[1990Pya]
Al <sub>3</sub> Sc	-43.5	Direct drop cal.	[1999Cac]
Al <sub>2</sub> Sc	-94.1	Acid solution cal.	[1990Pya]
Al <sub>2</sub> Sc	-48.0	Direct drop cal.	[1999Cac]
Al <sub>1.78</sub> Sc	-47.7	HT cal.	[1991Jun]
AlSc	-41.1	HT direct synthesis cal.	[1994Mes]
AlSc	-62.0	Acid solution cal.	[1990Pya]
AlSc	-46.0	Direct drop cal.	[1999Cac]
AlSc <sub>2</sub>	-28.2	Acid solution cal.	[1990Pya]
AlSc <sub>2</sub>	-37.0	Direct drop cal.	[1999Cac]
YAl <sub>3</sub>	-47.1	Combustion cal.	[1960Sny]
YAl <sub>3</sub>	-46.4	Liquid Al solution cal.	[1997Tim]
YAl	-53.5	e.m.f. (solid state galvanic cell)	[2000Bor]
YAl	-87.8	Combustion cal.	[1960Sny]
Y <sub>3</sub> Al <sub>2</sub>	-40.0	HT direct synthesis cal.	[1993Mes1]
Y <sub>3</sub> Al <sub>2</sub>	-46.9	Liquid-Al solution cal.	[1997Tim]
YAl <sub>2</sub>	-50.4	HT cal.	[1991Jun]
YAl <sub>2</sub>	-80.9	Combustion cal.	[1960Sny]
YAl <sub>2</sub>	-53.5	Liquid-Al solution cal.	[1997Tim]
La <sub>3</sub> Al <sub>11</sub>	-41.0	Direct isoperibolic cal.	[1997Bor]
La <sub>3</sub> Al <sub>11</sub>	-37.5	HT direct synthesis cal.	[1993Mes2]
LaAl <sub>3</sub>	-44.0	Direct isoperibolic cal.	[1997Bor]
LaAl	-46.0	Direct isoperibolic cal.	[1997Bor]
LaAl <sub>2</sub>	-50.5	Direct isoperibolic cal.	[1997Bor]
LaAl <sub>2</sub>	-54.2	Liquid Al solution cal.	[1985Col]
LaAl <sub>2</sub>	-49.9	HT cal.	[1991Jun]
Ce <sub>3</sub> Al <sub>11</sub>	-41.0	Direct isoperibolic cal.	[1991Bor]
Ce <sub>3</sub> Al <sub>11</sub>	-39.5	Liquid Al solution cal.	[1987Som]
CeAl <sub>2</sub>	-52.2	Liquid Al solution cal.	[1985Col]
CeAl <sub>2</sub>	-48.9	Liquid Al solution cal.	[1987Som]
CeAl <sub>2</sub>	-50.0	Direct isoperibolic cal.	[1991Bor]
CeAl <sub>2</sub>	-53.0	HT direct cal.	[1995Cac]
CeAl	-46.0	Direct isoperibolic cal.	[1991Bor]
Ce <sub>3</sub> Al	-16.4	Liquid Al solution cal.	[1987Som]
Ce <sub>3</sub> Al	-27.0	Direct isoperibolic cal.	[1991Bor]
PrAl <sub>2</sub>	-54.2	Liquid Al solution cal.	[1985Col]
Nd <sub>3</sub> Al <sub>11</sub>	-41.0	Direct isoperibolic cal.	[1993Bor]
NdAl <sub>3</sub>	-45.0	Direct isoperibolic cal.	[1993Bor]
NdAl <sub>2</sub>	-53.0	Direct isoperibolic cal.	[1993Bor]
NdAl <sub>2</sub>	-53.6	Liquid Al solution cal.	[1985Col]
NdAl	-50.0	Direct isoperibolic cal.	[1993Bor]
Nd <sub>2</sub> Al	-36.5	Direct isoperibolic cal.	[1993Bor]
Nd <sub>3</sub> Al	-27.5	Direct isoperibolic cal.	[1993Bor]
SmAl <sub>3</sub>	-48.0	Direct isoperibolic cal.	[1995Bor]
SmAl <sub>2</sub>	-54.3	Liquid Al solution cal.	[1985Col]
SmAl <sub>2</sub>	-55.0	Direct isoperibolic cal.	[1995Bor]
SmAl	-49.0	Direct isoperibolic cal.	[1995Bor]
Sm <sub>2</sub> Al	-38.0	Direct isoperibolic cal.	[1995Bor]
EuAl <sub>2</sub>	-36.0	Liquid Al solution cal.	[1985Col]
GdAl <sub>3</sub>	-43.4	Liquid Al solution cal.	[1988Col]
GdAl <sub>2</sub>	-53.2	Liquid Al solution cal.	[1988Col]
GdAl <sub>2</sub>	-53.2	Liquid Al solution cal.	[1985Col]
GdAl <sub>2</sub>	-51.4	Liquid Al solution cal.	[1987Som]

(continued)

**Table 3 Enthalpies of Formation (at 25-30 °C) of Some Intermetallic R-Al Compounds (continued)**

Phase	$\Delta_f H^\circ$ , kJ/mol at	Measurement Method	Reference
GdAl	-39.4	Liquid Al solution cal.	[1987Som]
GdAl	-42.9	Liquid Al solution cal.	[1988Col]
Gd <sub>3</sub> Al <sub>2</sub>	-33.3	Liquid Al solution cal.	[1988Col]
Gd <sub>2</sub> Al	-29.0	Liquid Al solution cal.	[1988Col]
Gd <sub>2</sub> Al	-34.7	Liquid Al solution cal.	[1987Som]
TbAl <sub>2</sub>	-52.4	Liquid Al solution cal.	[1985Col]
DyAl <sub>2</sub>	-52.7	Liquid Al solution cal.	[1985Col]
HoAl <sub>2</sub>	-52.5	Liquid Al solution cal.	[1985Col]
ErAl <sub>2</sub>	-50.5	Liquid Al solution cal.	[1985Col]
ErAl <sub>2</sub>	-49.1	Liquid Al solution cal.	[1987Som]
TmAl <sub>2</sub>	-51.0	Liquid Al solution cal.	[1985Col]
YbAl <sub>3</sub>	-32.5	Liquid Al solution cal.	[1983Pas]
YbAl <sub>3</sub>	-32.5	Direct isoperibolic cal.	[1997Bor]
YbAl <sub>2</sub>	-25.7	Dynamic diff. cal. and Knudsen effusion weight loss	[1978Pal]
YbAl <sub>2</sub>	-36.4	Liquid Al solution cal.	[1983Pas]
YbAl <sub>2</sub>	-39.5	Direct isoperibolic cal.	[1997Bor]
YbAl <sub>2</sub>	-38.2	Liquid Al solution cal.	[1985Col]
LuAl <sub>2</sub>	-52.6	Direct cal.	[1995Mes]

Notice the scattering of the experimental data obtained by acid solution calorimetry or combustion cal.

was studied by [1996Nor] while low-resistivity La-Al alloy thin films were investigated by [1996Tak1]. Finally the effect on solidification and microstructure of solidification condition in wedge chill casting and Bridgman growth were investigated for Al 5.7-19 wt.% La alloys by [1999Haw].

**2.2.4 Ce-Al.** [1990Kon] studied the Al-rich side of the diagram up to 4 at.% Ce, and [1996Sac] investigated the phase diagram in the region between 65 and 100 at.% Ce using DTA, metallographic analysis, quantitative EPMA, and x-ray examinations. The accepted phase diagram reported in [2000Oka] was drawn on the basis of these data. A thermodynamic modeling and optimization of the Ce-Al system on the basis of available literature information was carried out by [2001Cac].

A few comments about thermodynamic and magnetic properties may be noteworthy.

The low-temperature heat capacity of CeAl<sub>2</sub> was studied by [1971Dee] in the temperature range 8-300 K, and later the specific heat and spin-lattice relaxation rate of CeAl<sub>2</sub> were theoretically explained by [1996Hud]. The magnetic properties of the CeAl<sub>2</sub> and CeAl<sub>3</sub> intermetallic compounds were extensively studied by different authors. For instance, [1995Des] evaluated the effect of uniaxial pressure on magnetic susceptibility of CeAl<sub>2</sub>; [1995Lee] studied the magnetic properties and the electronic structure of CeAl<sub>2</sub> by magnetic susceptibility measurements and <sup>27</sup>Al pulsed NMR spectroscopy; [1996Glo] studied the break junctions of the heavy fermion antiferromagnet CeAl<sub>2</sub>; and [1998Smi] studied the electronic state of cerium in the CeAl<sub>2</sub> Laves phase. A high-pressure nuclear magnetic resonance (NMR) study of heavy fermion antiferromagnet CeAl<sub>2</sub> was carried out by [1996Kon], while a NMR study of

Table 4 Specific Heat of Selected R<sub>3</sub>Al and RAl<sub>2</sub> Compounds

Phase	Measurement T range [K]	C <sub>p</sub> [J/mol at K]			Method	Ref.
		A + B × 10 <sup>-3</sup> (T - C)				
La <sub>3</sub> Al (liq)	868-923	A = 51.1	B = -25.7	C = 298	DSC	[2001Bor]
αPr <sub>3</sub> Al	423-593	A = 23.9	B = 17.1	C = 298	DSC	[2001Bor]
βPr <sub>3</sub> Al	613-833	A = 21.9	B = 31.2	C = 298	DSC	[2001Bor]
Nd <sub>3</sub> Al	323-833	A = 22.3	B = 20.5	C = 298	DSC	[2001Bor]
EuAl <sub>2</sub>	400-1200	A = 24.8	B = 4.6	C = 273	Drop cal.	[1983Mer]
ErAl <sub>2</sub>	400-1200	A = 24.5	B = 4.7	C = 273	Drop cal.	[1983Mer]
YbAl <sub>2</sub>	400-1200				Drop cal.	[1983Mer]
$25.2 + 4.6 \times 10^{-3} (T - 273) + 40.7 \exp \frac{-327}{(T - 273)} \left[ \frac{1}{2(T - 273)^{0.5}} + \frac{327}{(T - 273)^{1.5}} \right]$						

the magnetically inhomogeneous ground state of CeAl<sub>3</sub> was carried out by [1995Gav], and new  $\mu$ SR results on the magnetic structure of CeAl<sub>2</sub> were presented by [1999Sch]. [1996Sha] investigated the population of 4f-shell and valence of cerium in CeAl<sub>3</sub> and CeAl<sub>2</sub> heavy fermion systems, and [1996Hun] studied the ground state of CeAl<sub>3</sub> at low temperature. Corsépius et al. [1997Cor] reported literature data on the physical properties of the heavy fermion CeAl<sub>3</sub> compound up to 1995. The effect of pressure on magnetoresistance of the heavy fermion compound CeAl<sub>3</sub> was studied by [1996Oom]. Moreover, low-temperature effects on the magnetic spectral response of CeAl<sub>3</sub>-based systems were studied by [1996Ale], and finally the magnetization density in Ce<sub>3</sub>Al<sub>11</sub> was studied by [1995Mun].

The following miscellaneous data on the Al-Ce system may be noteworthy.

[1995Nin] investigated the metastable extension of solid solubility of cerium in Al by measurement of the lattice spacings of the rapidly solidified alloys, while [1996Wat] investigated the microstructure and thermal stability of melt-spun Ce-Al alloy ribbons. Baricco et al. [1993Bar] carried out a thermodynamic investigation of glass forming in the Ce-Al system and later [1998Bar2] studied the thermodynamics of homogeneous crystal nucleation in Ce-Al metallic glasses. Moreover, [1996Li] studied the hydrogen-induced amorphization in Ce-Al alloys, and [1998Cro] studied anodic oxidation of Ce-Al alloys containing up to 27 at.% Ce. The effect on solidification and microstructure of solidification condition in wedge chill casting and Bridgman growth was investigated for Al 5.7-19 wt.% Ce alloys by [1999Haw]. [2001Aka] investigated the optical properties of Al-3 at.% Ce alloy. [2001Dab] studied the Ce-based conversion layers on Al alloys, and [2002Zha1] carried out a microstructural characterization and microhardness of rapidly solidified Ce-Al alloys. [2003Zha] studied the effect of ejection temperature and wheel speed on the microstructure of melt-spun Al-20Ce alloy.

**2.2.5 Pr-Al.** The phase diagram reported in [2000Oka] was drawn on the basis of work by [1990Kon] and [1996Sac]. [1990Kon] studied the Al-rich side of the diagram up to 4 at.% Pr, while [1996Sac] studied the phase diagram in the region between 65 and 100 at.% Pr using DTA, metallographic analysis, quantitative EPMA and x-ray examinations; the results are in good agreement with

previous literature data. Later [2001Yin] developed a thermodynamic optimization of the Pr-Al system using the THERMOCALC program in fairly good agreement with the experimental data.

The low-temperature heat capacity of PrAl<sub>2</sub> was studied by [1971Dee] in the temperature range 8-300 K, and low resistivity Pr-Al alloy thin films were investigated by [1996Tak1]. The effects of the crystal field on the effective exchange parameter in PrAl<sub>2</sub> were investigated by [1995Cal].

The metastable extension of solid solubility of praseodymium in Al by measurement of the lattice spacings of the rapidly solidified alloys was evaluated by [1995Nin].

**2.2.6 Nd-Al.** Saccone et al. [1996Sac] studied the Nd-rich region (70-100 at.% Nd) using differential thermal analysis, metallographic analysis, quantitative EPMA, and x-ray examinations. The Al-rich part of the system was recently investigated by different authors [1990Kon, 1997Kal, 1999Haw, 2001God]. In particular there was no agreement concerning composition and temperature of the eutectic reaction  $L \leftrightarrow Al + \alpha\text{-Nd}_3\text{Al}_{11}$ . By means of thermal and magneto-thermal analysis, and microstructure and x-ray analysis, Gödecke et al. [2001God] confirmed the eutectic composition proposed by [1990Kon] and [1999Haw] at 2.5 at.% Nd but modified the temperature (639 °C). The new version of the phase diagram is shown in Fig. 3.

Thermodynamics were investigated by several authors; the low-temperature heat capacity of NdAl<sub>2</sub> was studied by [1971Dee] in the temperature range 8-300 K; the heats of vaporization and vapor pressure of Nd-Al alloys at temperatures less than or equal to 1727 °C were investigated by [1983Zvi] using mass spectrometry. Thermodynamics of the solid and/or liquid phases were also investigated by [1976Zvi], who studied enthalpies of mixing of the liquid at 977-1277 °C by high-temperature calorimetry; [1977Ker], who studied enthalpies of mixing by the emf method at 760 °C; [1979Kob1, 1984Kob], who studied partial Gibbs energies of Nd in two-phase fields at 527 and 727 °C by the emf method; and [1979She], who studied partial Gibbs energies of Al in the liquid at 1427 °C by vapor pressure measurements. The Nd-Al system was optimized by [1996Cla], [1996Wan], [1998Bar2] (modeling also the amorphous phase), and [2001Cac]; in the last paper more recent literature data were considered.

Low-resistivity Nd-Al alloy thin films were investigated by [1996Tak1]. The magnetic properties of the Nd<sub>2</sub>Al intermetallic compound were studied by [1999Li1]. Baricco et al. [1993Bar] carried out a thermodynamic investigation on glass forming in the Nd-Al system, and later [1998Bar2] studied the thermodynamics of homogeneous crystal nucleation in Nd-Al metallic glasses. [1995Nin] investigated the metastable extension of the solid solubility of neodymium in Al by measurement of the lattice spacings of the rapidly solidified alloy while [1996Wat] studied the microstructure and thermal stability of melt-spun Nd-Al alloy ribbons.

**2.2.7 Sm-Al.** The Al-rich part (66-100 at.% Al) of this system was studied by Buschow and Van Vucht [1965Bus,1967Bus] and by Casteels [1967Cas]. More recently, [1990Kon] studied the Al-rich side of the diagram up to 5 at.% Sm. This part of the phase diagram is significantly different from the corresponding region of the other R-Al systems. Saccone et al. [1998Sac] investigated the complete composition range of this phase diagram by means of XRD, DTA, light optical microscopy (LOM), SEM and EPMA; nevertheless some uncertainties still remain in the Al-rich part of the phase diagram due to experimental difficulties. The phase diagram is sketched in Fig. 3.

[2001Ada] studied the anisotropic spin form factor of SmAl<sub>2</sub>.

The crystallization behavior in rapidly solidified Sm-Al alloys was studied by [1996Fol] and [1998Riz], while the metastable extension of the solid solubility of Sm in Al was investigated by measurement of the lattice spacings of the rapidly solidified alloys by [1995Nin]. Moreover Sm-Al alloy thin films with low resistivity and high thermal stability for microelectronic conductor lines were studied by [1996Tak2].

**2.2.8 Eu-Al.** No experimental Eu-Al phase diagram is available except for the Al-rich side up to 4 at.% Eu determined by [1990Kon]. An extrapolated version of this phase diagram was suggested by [1991Oka]. The antiferromagnetic structure of the EuAl<sub>2</sub> intermetallic compound was studied by [1997Oul]. [1995Nin] investigated the metastable extension of solid solubility of europium in Al by measurement of the lattice spacings of the rapidly solidified alloys.

**2.2.9 Gd-Al.** Saccone et al. [2000Sac] investigated the phase diagram in the composition region up to 66.7 at.% Al using the same techniques as [1996Sac]; [2001Gro] and [2003Cac] carried out thermodynamic calculations of the Gd-Al phase diagram using the CALPHAD method. The new version of the phase diagram is shown in Fig. 3.

The following notes about physical properties and metastable conditions may be noteworthy. The low-temperature heat capacity of GdAl<sub>2</sub> was studied by [1971Dee] in the temperature range 8-300 K. Specific heat and magnetic entropy associated with magnetic ordering in GdAl<sub>2</sub> were studied by [1987Sah]. Phase composition and thermodynamic properties of Gd-Al alloys were studied by [1979Kob2] by emf measurements; thermal expansion and heat capacity of GdAl<sub>2</sub> were investigated by [1976Sle]. Moreover, the partial enthalpies of mixing of liquid Al-rich alloys were determined at about 927 °C using a high-temperature solution calorimeter by [1985Lee].

Magnetic properties and electronic structure of GdAl<sub>2</sub> were extensively studied by several authors: [1995Lee] by magnetic susceptibility measurements and <sup>27</sup>Al pulsed NMR spectroscopy, [1996Sza] using photoelectron spectroscopy, and [1999Str] by <sup>155</sup>Gd-Mossbauer spectroscopy under pressure. Li et al. [1999Li1,2 and 2000Li] investigated the magnetic properties of Gd<sub>2</sub>Al in amorphous and crystalline states. [1995Nin] investigated the metastable extension of the solid solubility of Gd in Al by measurement of the lattice spacings of the rapidly solidified alloy,

**2.2.10 Tb-Al.** No phase diagram is available for Tb-Al; [1993Fer] gave a theoretical prediction for this system. Vapor pressures of Al in the Tb-Al system were measured and several thermodynamic characteristics calculated on the basis of the measured values by [1985Zvi]. For the TbAl<sub>2</sub> intermetallic compound the specific heat and magnetic entropy associated with magnetic ordering in sintered compound were studied by [1987Sah], while the heat capacity in the temperature range from 1.5-20 K in external magnetic fields up to 7.5 T was studied by [1986Sch].

**2.2.11 Dy-Al.** The partial experimental Dy-Al phase diagram from 66.6-100 at.% Al was reported by [1988Gsc]. Later [2000Sac] investigated the system in the composition region up to 66.7 at.% Al using conventional DTA, LOM, SEM, XRD techniques, and [2003Cac] computed the phase diagram with the CALPHAD method.

The thermodynamic and magnetic properties of the DyAl<sub>2</sub> phase are the subject of several papers; the low-temperature heat capacity and thermal properties were studied by [1977Ino] and in external magnetic fields up to 7.5 T by [1986Sch]; specific heat and magnetic entropy associated with magnetic ordering in a DyAl<sub>2</sub> sintered compound were studied by [1987Sah]; and [2000Ran] studied the anomalies in the magnetocaloric effect. In addition, [1995Nin] investigated the metastable extension of the solid solubility of dysprosium in Al by measurements of the lattice spacings of the rapidly solidified alloys, and [1996Tak2] studied Al-Dy alloy thin films with low resistivity and high thermal stability for microelectronic conductor lines.

**2.2.12 Ho-Al.** The Ho-Al phase diagram, reported by [1988Gsc], was studied experimentally from 0 to 85 at.% Ho by [1965Mey]. More recently [2003Cac] optimized this system with the Calphad method. The computed phase diagram showed poor agreement with the experimental results of [1965Mey] but good agreement with homologous Gd and Dy systems. For the HoAl<sub>2</sub> intermetallic compound, the low-temperature heat capacity in external magnetic fields up to 7.5 T was determined by [1984Sch], while the specific heat and magnetic entropy associated with magnetic ordering in the sintered compound were studied by [1987Sah].

**2.2.13 Er-Al.** The Er-Al phase diagram was re-investigated in the 50-80 at.% Er composition range by [2002Sac] and optimized by [2002Cac]. Several discrepancies in the invariant equilibrium temperatures compared with those reported in [1988Gsc] can be observed; some uncertainties still remain with regard to the invariant temperatures and melting behavior of the Er<sub>2</sub>Al phase (peritectic or congruent melting). This new version is sketched in Fig. 3.



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The thermodynamic properties of the  $\text{ErAl}_2$  intermetallic compound were studied by different authors: [1971Mac] studied the low-temperature specific heat, [1987Sah] investigated the specific heat and magnetic entropy associated with magnetic ordering in the sintered compound, [1977Ino] studied the low temperature heat capacity and thermal properties, and [1986Sch] studied the low-temperature heat capacity in the temperature range from 1.5 to 20 K in external magnetic fields up to 7.5 T. [1995Nin] investigated the metastable extension of the solid solubility of Er in Al by measurement of the lattice spacings of the rapidly solidified alloy.

**2.2.14 Tm-Al.** No experimental phase diagram is available, though [1990Mas] reported an extrapolated one.

[1986Sch] studied the low-temperature heat capacity of  $\text{TmAl}_2$  in the temperature range from 1.5 to 20 K in external magnetic fields up to 7.5 T, while [2000Bra] studied transport properties of  $\text{TmAl}_3$  and  $\text{TmAl}_2$ .

**2.2.15 Yb-Al.** The accepted phase diagram is that reported by [1989Gsc]. More recently, [1990Kon] studied the Al-rich side of the phase diagram up to 4 at.% Yb, and found the Al-richest eutectic point to be slightly different (3.5 at.% Yb) from the previously reported value (4 at.% Yb [1989Gsc]). [1995Nin] investigated the metastable extension of solid solubility of Yb in Al by measurements of the lattice spacings of the rapidly solidified alloy.

The more recent informations about Yb-Al intermetallic compounds were reported in the following: Polarized neutron diffraction study on the magnetism in  $\text{YbAl}_3$  was carried out by [2000Hie]. The magnetic susceptibility of  $\text{YbAl}_3$  was measured by [2000Bra]. The effect of pressure on transport properties of  $\text{YbAl}_3$  was studied by [2001Oha]. Several physical properties of  $\text{YbAl}_3$  (susceptibility, magnetization, specific heat, 4f occupation number, Hall effect, magneto-resistance for single crystal) were studied by [2002Cor]. Optical properties and electronic structure of a single crystal of  $\text{YbAl}_2$  were studied by [2000Lee].

**2.2.16 Lu-Al.** A schematic Lu-Al phase diagram was reported by [1993Oka]; there are five intermetallic compounds but the melting temperatures are still unknown.

As for the  $\text{LuAl}_2$  compound, several physical properties were studied; the magnetic properties and the electronic structure were determined by [1995Lee and 2002Kwa] by magnetic susceptibility measurements and  $^{27}\text{Al}$  pulsed NMR spectroscopy, the low temperature heat capacity and thermal properties were studied by [1977Ino], and optical properties and electronic structure of a single crystal of  $\text{LuAl}_2$  were examined by [2000Lee].

### 2.3 Copper-Rare Earth Systems

Literature data up to 2001 are reported in an assessment by [2002Ria]. Crystal structure data are reported in Table 5.

Several other papers were published, mainly on magnetic studies by different authors. [2001Luo] studied the effect of crystalline electric field on the Néel temperatures of  $\text{RCu}_2$  compounds ( $R = \text{Tb to Tm}$ ) theoretically: [2002Gyg] investigated the magnetic structure of  $\text{GdCu}_2$  by zero-field  $\mu\text{SR}$  spectroscopy on a single crystal between 5 K and  $T_N = 39.6$  K. [2002Svo] studied the antiferromagnetic order-

ing in  $\text{TmCu}_2$ . [2002Vej] examined the magnetic properties of  $\text{SmCu}_2$  single crystal. [2002Rot] modeled the magnetostriction in  $\text{RCu}_2$  compounds using the McPhase simulation program. Moreover, [2002Rei] studied the electronic transport properties of  $\text{NdCu}$  using point-contact spectroscopy, and [2003Hen] studied the lattice dynamics of  $\text{YCu}_2$  by inelastic neutron scattering on a single crystal.

Regarding amorphous phases, [1993Sum] carried out a comparative study of heavy fermion behavior in amorphous  $\text{CeCu}_6$  and  $\text{LaCu}_6$ , [1999Sum] studied the structures, electronic states, and low-temperature properties of amorphous Ce-Cu alloys, and [1999Tan] studied the effects of La, Nd, and Sm additions on the structures and properties of rapidly solidified copper alloys. Also, [2001Ilk] studied the electron-quasiparticle interaction function using point-contact spectroscopy and electrical resistivity of the cubic phase of melt-spun  $\text{TmCu}_5$ , [2002Ilk1] investigated the point-contact spectra of hetero-contacts between the cubic melt-spun  $\text{TbCu}_5$  and Cu, and [2002Ilk2] combined point-contact spectroscopy and temperature dependence of the electrical resistivity to study the electron-quasiparticle interaction function and transport properties of the cubic  $\text{HoCu}_5$  prepared by melt spinning.

## 3. Ternary R-Cu-Al phases

### 3.1 Comments on the Experimental Techniques Used in the Preparation and Study of the Ternary R-Cu-Al Alloys

Typical purities of the metals used by different authors were stated generally to be in the ranges of  $R = 98.6\text{-}99.9\%$  and  $\text{Al and Cu} = 99.6\text{-}99.999$  mass%. Alloy samples were generally prepared by melting the pure metals together in an electric-arc furnace on a water-cooled Cu hearth under a purified argon atmosphere. Another method used by several authors [1961Gla, 1968Zar, 1969Zar, 1972Dri] for the alloys in the Al-rich corner is melting the pure elements together in a resistance furnace in alumina crucibles under a eutectic  $\text{LiCl} + \text{KCl}$  (or  $\text{LiF}$ ) flux and, after thermal treatment, quenching in toluene. Different annealing temperatures were used: from 250 to 650 °C for the isothermal sections and higher temperatures—for instance 800 °C—for determination of the crystal structure of the phases, for different periods of time, typically between 400 and 2000 h.

The isothermal sections of the different systems were generally studied by XRD and in some cases by metallography, micro-hardness measurements, and electric resistivity.

The ternary compounds identified in these systems were generally studied by XRD with powder or single-crystal methods. Polythermal sections of the (Sc, Y, La, Ce, Nd)-Cu-Al systems were studied by DTA and metallography, and in some cases (Y) by micro-hardness measurements. Magnetic, electric, and thermodynamic property and Mössbauer measurements were carried out for different stoichiometries in several systems.

### 3.2 Crystal Structure Types Observed in the R-Cu-Al alloys

A high number of intermediate phases have been observed in all of the R-Cu-Al systems. These phases gener-

**Table 5** Crystal Structure of the Phases Observed in the R–Cu Systems With R = Rare Earth

Phases	Sc	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	Structural type
R <sub>2</sub> Cu	...	...	...	...	...	...	...	A	...	...	...	...	...	...	...	...	A <i>oP12</i> -Ca <sub>2</sub> Cu
RCu	B	B	C	C	C	C	C	C	B	D <sup>LT</sup>	B	B	B	B	C	B	B <i>cP2</i> -CsCl
...	...	...	...	...	...	...	B(a)	...	...	B <sup>HT</sup> (b)	...	...	...	...	...	...	C <i>oP8</i> -FeB
...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	D <i>tP2</i> -HgMn
RCu <sub>2</sub>	E	F	G	F	F	F	F	F	F	F	F	F	F	F	F	F	E <i>tI6</i> -MoSi <sub>2</sub>
...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	I <sup>HP</sup>	...	F <i>oI12</i> -CeCu <sub>2</sub>
...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	G <i>hP3</i> -AlB <sub>2</sub>
...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	I <i>hP12</i> -MgZn <sub>2</sub>
R <sub>2</sub> Cu <sub>7</sub> (c)	...	X	...	...	...	X	?	...	X	?	X	?	(d)	?	X	?	X struct. unknown
R <sub>14</sub> Cu <sub>51</sub> (e)	...	...	...	J	...	...	...	...	...	...	...	...	...	...	...	...	J <i>hP68-x</i> -Gd <sub>14</sub> Ag <sub>51</sub>
R <sub>6</sub> Cu <sub>23</sub>	...	...	...	...	...	...	...	...	...	K <sup>HP</sup>	K <sup>HP</sup>	...	...	...	K <sup>HP</sup>	K <sup>HP</sup>	K <i>cF116</i> -Th <sub>6</sub> Mn <sub>23</sub>
RCu <sub>4</sub>	X	?(f)	L(g)	M	M	M	M	...	...	...	...	...	...	...	...	...	L <i>tI98</i> -LaCu <sub>4</sub>
...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	M <i>oP20</i> -CeCu <sub>4</sub>
R <sub>2</sub> Cu <sub>9</sub> (h)	...	...	...	...	...	...	...	...	N	?	N	?	(d)	?	N	?	N <i>mC</i> -7450
RCu <sub>5</sub>	...	(i)	P	P	P	P	P	P	Q <sup>LT</sup>	Q <sup>LT</sup>	Q <sup>LT</sup>	Q <sup>LT</sup>	Q	Q	P(j)	Q	P <i>hP6</i> -CaCu <sub>5</sub>
...	...	...	...	...	...	...	...	...	P <sup>HT</sup>	P <sup>HT</sup>	P <sup>HT</sup>	P <sup>HT</sup>	...	...	Q <sup>HP</sup>	...	Q <i>cF24</i> -AuBe <sub>5</sub>
RCu <sub>6</sub>	...	...	R <sup>LT</sup>	R <sup>HT</sup>	R <sup>LT</sup>	R <sup>LT</sup>	S	...	S	...	...	...	...	...	...	...	R <i>mP28-α</i> LaCu <sub>6</sub>
...	...	...	S <sup>HT</sup>	S <sup>HT</sup> (b)	S <sup>HT</sup> (b)	S <sup>HT</sup> (b)	...	...	...	...	...	...	...	...	...	...	S <i>oP28</i> -CeCu <sub>6</sub>
RCu <sub>7</sub>	...	U(k)	...	...	...	...	...	...	U <sup>HT</sup>	U <sup>HT</sup>	U <sup>HT</sup>	...	...	...	...	...	U <i>hP8</i> -TbCu <sub>7</sub>
RCu <sub>13</sub> (l)	...	...	V <sup>HT</sup>	...	V	...	...	...	...	...	...	...	...	...	...	...	V <i>cF112</i> -NaZn <sub>13</sub>

(a) Conflicting data were reported for the SmCu phase about the stability of its crystal structure according to cooling rate, impurities, etc.

(b) For the TbCu, CeCu<sub>6</sub>, PrCu<sub>6</sub> and NdCu<sub>6</sub> phases the HT forms are stable above -157, -43, -6, -118 °C respectively

(c) Suggested for some R as a phase stable at high temperature. Observed by DTA only for Y, Nd, Gd, Dy, Yb; assumed by analogy for Sm, Tb, Ho, Tm, Lu

(d) These formulae have been assumed on the basis of analogy with the other R by [1994Sub], ascribing these formulae to the phases described as ErCu<sub>x</sub> and ErCu<sub>y</sub> by [1970Bus].

(e) Partially disordered, metastable?

(f) Suggestions have been given about Y<sub>2</sub>Cu<sub>6</sub> stoichiometry or alternatively about an off-stoichiometric extension of the YCu<sub>4</sub> phase.

(g) The structure of this phase [1991Vil] corresponds to (80Cu + 18La) in the unit cell with an atomic ratio Cu/La of 80/18 ≅ 4.4.

(h) Superstructure related to AuBe<sub>5</sub>-type subcell is given for Yb [1996Cer], possibly Gd and Dy are isostructural to Yb. This stoichiometry has been assumed by analogy for Tb, Ho, Tm, Lu.

(i) The YCu<sub>5</sub> compound as a possibly metastable phase has also been described.

(j) For the hexagonal form of YbCu<sub>5</sub> a variant corresponding to hP20 YbCu<sub>6.5</sub> with ordered vacancies has been described [1972Hor].

(k) The Cu-richest phase in the Cu–Y system has been alternatively described as YCu<sub>7</sub> or YCu<sub>6</sub>. It probably corresponds to a certain range of solid solutions around these two formulae (-12 up to 16 at.% Y).

(l) Possibly metastable phases?

HT = high temperature, LT = low temperature, HP = high pressure

ally pertain to the composition range lying between 7 and 33 at.% rare earth metals. Their stoichiometries and some crystal structure data are listed in Table 6. A few comments and remarks about the different compounds, reported in the following in order of increasing quantity of R, may be noteworthy. However, see also the comments reported in the following paragraphs for the different R–Cu–Al systems.

**NaZn<sub>13</sub>-Type Phases.** These phases correspond to an ideal atomic ratio R/(Cu+Al) = 1:13 (corresponding to 7.1 at.% R), and are characterized by the large cubic cell of the cF112 NaZn<sub>13</sub>-type (space group *Fm*  $\bar{3}$  *c*) with Na in *8a* and Zn in *8b* + *96i*. A very high co-ordination number, 24, is observed around Na, while it is 12 and 10 for the two sites respectively occupied by Zn. In the ternary alloys, the large R atoms are in the Na position and for Cu and Al a random distribution in the *b* and *i* sites has generally been suggested. The crystal structure and magnetic properties of these phases were determined by Felner [1980Fel and 1982Fel] for samples with a nominal composition of RCu<sub>6</sub>Al<sub>6</sub>. However, these were probably multiphase samples. For

CeCu<sub>6</sub>Al<sub>6</sub>, a significant larger cell parameter *a* = 1187 pm, in comparison with *a* = 1182.2 pm [1985Cor1,1985Cor2], was reported by [1980Fel]. This can be related to the stoichiometry CeCu<sub>6.5</sub>Al<sub>6.5</sub> proposed by [1985Cor1, 1985Cor2]. For R = Pr, Nd, and Sm formulae such as RCu<sub>6+x</sub>Al<sub>6</sub> have been proposed with *x* values close to 0 [1980Fel] and 1 [1978Zar, 1975Zar and 1984Pre] with a stoichiometric ratio changing from 1:12 to 1:13. A large homogeneity range, corresponding to Al/Cu substitution, has been suggested for the systems with La [1969Zar] and Eu [1992Yan].

**ThMn<sub>12</sub>-Type Phases.** A review of structure, magnetic, and related properties of the ThMn<sub>12</sub>-type compounds of rare earths and actinides was published by [1996Sus]. This structure corresponds to a body-centered tetragonal cell with 2 Th in *a* and sets of 8 Mn in *f*, *i*, *j* (space group *I4/mmm*). A high coordination, 20, was also observed in this structure around the large Th atoms, while around Mn coordination numbers of 12 and 14 were observed. This structure was observed in the ternary alloys for Sc, Y, and all the

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Table 6 Crystal Structure of Phases Observed in the R–Cu–Al Systems With R = Rare Earth

Phases	Sc	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	Structural Type
$\tau_1$ $\sim$ RCu <sub>7-x</sub> Al <sub>6+y</sub>	...	...	A <sub>SS</sub>	A	A	A	A	A <sub>SS</sub>	...	...	...	...	...	...	...	...	A <i>cF112</i> -NaZn <sub>13</sub>
$\tau_2$ R(Cu,Al) <sub>12</sub>	B <sub>SS</sub>	B	B	B	B <sub>SS</sub>	B	B	B	B	B <sub>SS</sub>	B <sub>SS</sub>	B	B	B	B <sub>SS</sub>	B <sub>SS</sub>	B <i>tI26</i> -ThMn <sub>12</sub>
$\tau_3$ R(Cu,Al) <sub>11</sub>	...	C	...	...	C	C	...	...	C	...	...	...	...	...	...	...	C <i>tI48</i> -BaCd <sub>11</sub>
$\tau_4$ $\sim$ RCu <sub>6,6</sub> Al <sub>4,4</sub>	...	D	...	...	...	...	...	...	D	D	...	...	...	...	...	...	D <i>oF*</i> -Tb(Cu <sub>0,58</sub> Al <sub>0,42</sub> ) <sub>11</sub>
$\tau_5$ $\sim$ R <sub>2</sub> Cu <sub>10</sub> Al <sub>7</sub> $\sim$ R <sub>2</sub> Cu <sub>7</sub> Al <sub>10</sub>	...	E	...	E <sub>SS</sub>	E	E	E	...	E <sub>SS</sub>	E <sub>SS</sub>	E <sub>SS</sub>	E	E <sub>SS</sub>	E	E <sub>SS</sub>	E <sub>SS</sub>	E <i>hR57</i> -Th <sub>2</sub> Zn <sub>17</sub>
$\tau_6$ R <sub>4</sub> (Cu <sub>0,26</sub> Al <sub>0,74</sub> ) <sub>33</sub>	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	F	F <i>tI*</i> -Yb <sub>4</sub> (Cu <sub>0,26</sub> Al <sub>0,74</sub> ) <sub>33</sub>
$\tau_7$ R(Cu,Al) <sub>6</sub>	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	G	G <i>tI14</i> -YbMo <sub>2</sub> Al <sub>4</sub>
$\tau_8$ R <sub>2</sub> Cu <sub>7,5</sub> Al <sub>3,5</sub>	H	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	H <i>h**</i>
$\tau_9$ R <sub>3</sub> Cu <sub>9</sub> Al <sub>7</sub>	I	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	I <i>o**</i>
$\tau_{10}$ R <sub>6</sub> Cu <sub>16,4</sub> Al <sub>13,9</sub>	J	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	J <i>h**</i>
$\tau_{11}$ R(Cu <sub>1-x</sub> Al <sub>x</sub> ) <sub>5</sub>	...	K?	K <sup>o</sup>	K <sup>o</sup>	K <sup>o</sup>	K <sup>o</sup>	K <sup>o</sup>	L	K <sup>o</sup>	K <sup>o</sup>	K	K	K	K	K <sup>o</sup>	K	K <i>hP6</i> -CaCu <sub>5</sub>
...	...	...	...	...	...	...	...	K <sup>o</sup>	...	...	...	...	...	...	...	...	L <i>hP6</i> -CeNi <sub>2</sub> Al <sub>3</sub>
$\tau_{12}$ RCu <sub>2</sub> Al <sub>2</sub>	M	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	M <i>o**</i>
...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	N <i>tI10</i> -BaAl <sub>4</sub>
$\tau_{13}^{\wedge}$ $\sim$ RCu <sub>0,5</sub> Al <sub>3,5</sub> -RCuAl <sub>3</sub>	...	N <sub>SS</sub>	N <sub>SS</sub>	N <sub>SS</sub>	N <sub>SS</sub>	N	N	...	O	...	...	...	P	P	N	...	O <i>tI10</i> -BaNiSn <sub>3</sub>
...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	P <i>oI10</i> -HoCuAl <sub>3</sub>
$\tau_{14}$ R <sub>6</sub> (Cu,Al) <sub>23</sub>	...	...	...	...	...	...	...	...	...	...	Q	Q	Q	Q	Q	Q	Q <i>cF116</i> -Th <sub>6</sub> Mn <sub>23</sub>
$\tau_{15}^{\wedge}$ R <sub>3</sub> (Cu,Al) <sub>11</sub>	...	R <sub>SS</sub>	...	...	...	...	...	...	R	R	R	R	...	...	...	...	R <i>oI28</i> -La <sub>3</sub> Al <sub>11</sub>
$\tau_{16}$ RCu <sub>2</sub> Al	S	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	S <i>cF16</i> -MnCu <sub>2</sub> Al
$\tau_{17}$ R <sub>3</sub> Cu <sub>5</sub> Al <sub>4</sub>	...	...	T	...	...	...	...	...	...	...	...	...	...	...	...	...	T <i>h**</i>
$\tau_{18}$ $\sim$ RCu <sub>0,9</sub> Al <sub>2,1</sub>	...	U	...	...	...	...	...	...	V	V	V	V	V	V	V	V	V <i>hR36</i> -Ca <sub>3</sub> Cu <sub>2</sub> Al <sub>7</sub>
...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	V <i>hR36</i> -PuNi <sub>3</sub>
$\tau_{19}$ $\sim$ RCu <sub>0,83</sub> Al <sub>1,27</sub>	...	...	...	...	...	...	...	W	...	...	...	...	...	...	...	...	W <i>hP93</i> -Ca <sub>30</sub> Cu <sub>25</sub> Al <sub>38</sub> <sup>+</sup>
$\tau_{20}$ RCuAl	...	...	...	...	...	...	Y	...	Y	Y	Y	...	...	Y	Y	Y	X <i>hP12</i> -MgZn <sub>2</sub>
...	...	X	Y	...	Y	Y	Z <sup>HP</sup>	...	Z <sup>HP</sup>	Z <sup>HP</sup>	Z <sup>HP</sup>	Y	Y	Z <sup>HP</sup>	Z <sup>HP</sup>	Z <sup>HP</sup>	Y <i>hP9</i> -ZrNiAl
...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	...	Z <i>cF24</i> -Cu <sub>2</sub> Mg
$\tau_{21}$ RCu <sub>0,6</sub> Al <sub>1,4</sub>	AA	...	...	...	...	...	...	AA	...	...	...	...	...	...	...	...	AA <i>hP24</i> -Ni <sub>2</sub> Mg

Due to the complexity of the different systems and to some ambiguities in literature data, this table is only a summary. The interested reader is invited to read both the paragraphs dealing with the different structures and those dealing with the systems.

The progressive number of each  $\tau$  phase is shown in the various isothermal sections where all the phases identified are shown, even those of unknown structure; their compositions are indicated in the different sections.

<sup>o</sup> Connected to the binary phase

<sup>+</sup> Described by [1991Daa] as *hP93*-Ca<sub>7</sub>Cu<sub>6</sub>Al<sub>7</sub>

<sup>^</sup> See text 3.2.1 ( $\sim$ RT<sub>4</sub> phases)

ss Solid solution

HP high pressure

lanthanides. In the isothermal sections reported in the literature for the different systems, generally, the composition RCu<sub>4</sub>Al<sub>8</sub> is given (for La 1:2:10). According to [1980Fel] and [2001Duo] the ThMn<sub>12</sub> type structure is observed (for Gd to Lu and Y) also for the composition RCu<sub>6</sub>Al<sub>6</sub>. Magnetic properties of GdT<sub>4</sub>Al<sub>8</sub> and GdT<sub>6</sub>Al<sub>6</sub> (T = Cr, Mn, Cu) and crystallographic site occupation were described by [2001Duo]; for the light R for a composition close to 1:6:6 the NaZn<sub>13</sub>-type structure was described by [1980Fel]. From the literature, it is not always clear whether the two compositions 1:4:8 and 1:6:6 correspond to two different phases or if they are the limits of a solid solution range. In his review, Suski [1996Sus] noticed that the authors usually distinguish three types within these phases RT<sub>x</sub>Al<sub>12-x</sub> with T = transition element and x = 4,5,6; he remarked, however, that this is only a simplification for easier presentation of the results and solid solutions with 3 < x ≤ 6, or sometimes even higher x values, were reported. In a number of systems, moreover, these phases were explicitly determined as solid solutions. For instance in the determina-

tion of the isothermal section at 650 °C of the Tb-Cu-Al system, [1995Kuz] identified the ThMn<sub>12</sub> structure for a phase having a small range of composition TbCu<sub>12-x</sub>Al<sub>x</sub> (6.8 ≤ x ≤ 7.2). Notice, however, that generally the isothermal sections were studied at temperatures between 400 and 600 °C while Felner and Duong prepared their samples at a higher temperature (with long annealing times at T ≥ 800 °C).

*BaCd<sub>11</sub>-Type Phases.* This structure corresponds to a body-centered tetragonal cell in the space group *I4<sub>1</sub>/amd* with Ba in *4a* and Cd in *4b*, *8d*, and *32i*. A coordination of 22 is observed around Ba, while the coordination around Cd ranges from 10 to 14. For ternary alloys, this phase with the ideal BaCd<sub>11</sub>-type structure was described for PrCu<sub>7,6</sub>Al<sub>3,4</sub> ([1978Zar]), NdCu<sub>8,6</sub>Al<sub>2,4</sub> ([1969Bod]) and for GdCu<sub>7,8</sub>Al<sub>3,2</sub>, with Gd in *4a*, Cu in *8d* and (Cu + Al) in *4b* and *32i* by [2001Gum]. For Y, Gd, and Tb, an orthorhombic variant of this structure was observed (YCu<sub>6,8</sub>Al<sub>4,2</sub> [2003Kra], GdCu<sub>6,6</sub>Al<sub>4,4</sub> [2001Gum], and TbCu<sub>6,4</sub>Al<sub>4,6</sub> [1995Kuz]) with similar values of the coordination num-

bers. For samarium, a  $\sim\text{SmCu}_6\text{Al}_5$  phase of an unknown structure, was described by [1983Pre].

*Th<sub>2</sub>Zn<sub>17</sub>-Type Phases.* In this rhombohedral structure, the following positions of the hexagonal cell (space group  $R\bar{3}m$ ) are occupied: Th in  $6c$ , Zn in  $6c$ ,  $9d$ ,  $18f$ , and  $18h$ . The coordination number around Th is 19, and that around the Zn atoms ranges between 12 and 14. For the Gd-Cu-Al alloy the structure was described [1982Pre, 1988Pre and 2001Gum] as corresponding to Gd in  $6c$ , Cu in  $9d$  and a mixed occupation (Cu + Al) in the other sites. This structure has been described, with the exception of Sc, La, and Eu, for all the entire rare earths series with phase stoichiometry varying slightly through the series. The stoichiometry variation is illustrated by the invariant stoichiometries:  $\sim\text{R}_2\text{Cu}_7\text{Al}_{10}$  for Pr, Sm, and Ho [1978Zar; 1982Pre],  $\sim\text{R}_2\text{Cu}_8\text{Al}_9$  for Nd [1975Zar], and  $\sim\text{R}_2\text{Cu}_{10}\text{Al}_7$  for Tm alloys [1993Ste1]; For the systems given by Ce, Gd, Tb, Dy, Er, Yb, and Lu, larger or smaller solid solution ranges (within the previously reported composition values) have been proposed. As for the Th<sub>2</sub>Zn<sub>17</sub>-type compound in the Y-Cu-Al system, two compositions have been suggested: Y<sub>2</sub>Cu<sub>9</sub>Al<sub>8</sub> [1968Zar] at 400 °C and Y<sub>2</sub>Cu<sub>12.0-10.5</sub>Al<sub>5.0-6.5</sub> solid solution [2003Kra] at 550 °C. According to [1978Pop], the structure Th<sub>2</sub>Ni<sub>17</sub>-type was observed in the Gd system at  $\sim\text{Gd}_2\text{Cu}_6\text{Al}_{11}$ . This phase however was not confirmed by [1988Pre] and [2001Gum].

*CaCu<sub>5</sub>-Type Phases.* The crystal structure of these phases has been studied by [1978Tak] for the composition  $\text{RCu}_4\text{Al}$  with R = La to Sm and Gd to Tm. The structure corresponds to Ca in  $1a$  and Cu in  $2c + 3g$  in the hexagonal cell (space group  $P6/mmm$ ). The coordination numbers are 18 around Ca and 12 around Cu atoms. In the ternary phase, see for instance Lu-Cu-Al as described by [1992Kuz1], R is in  $1a$  and Cu and Al are mixed in the other two positions. In the ternary R-Cu-Al systems we may find this structure either as an extension of the binary  $\text{RCu}_5$  phase or as a real ternary phase. We may notice that in binary R-Cu systems structural transitions from  $\text{CaCu}_5$  to  $\text{AuBe}_5$  structure types are observed. Binary compounds  $\text{RCu}_5$  with R = Y, La to Eu, and Yb belong to the  $\text{CaCu}_5$ -type structure and those with R = Er, Tm, Lu belong to the  $\text{AuBe}_5$ -type. Compounds with Gd, Tb, Dy, and Ho can form in both types, with temperature-dependent polymorphic transformation; for Yb an  $\text{AuBe}_5$  type form is obtained under high pressure. This has an influence on the ternary system R-Cu-Al. The existence of compounds with  $\text{CaCu}_5$ -type structure in binary R-Cu systems leads to the formation of a considerable number of solid solutions in the corresponding ternary system with Al (R = La-Sm, Gd, Tb, Yb, and possibly Y). If a compound with a  $\text{CaCu}_5$ -type structure does not exist in the binary system as a stable phase at the temperature considered, a ternary compound  $\text{R}(\text{Cu},\text{Al})_5$  is formed. This is the case in the R-Cu-Al systems with R = Dy, Er, Lu, and possibly Ho.

Notice however that, in the binary systems presenting the two 1:5 structures (Gd to Ho and Y), generally the  $\text{CaCu}_5$  modification is stable at high temperature and the  $\text{AuBe}_5$ -type at room temperature. The transformation temperatures reported in the literature are really higher than those at which the isothermal sections have been established. The

reported data are, however, uncertain, and moreover, metastable situations cannot be excluded during the preparation of the ternary alloys.

In the Eu system, both a solid solution based on  $\text{EuCu}_5$  and a ternary phase described as pertaining to the  $\text{CeNi}_2\text{Al}_3$  type (ordered?) were observed [1992Yan].

*$\sim RT_4$  Phases.* In the composition range around 20-21 at.% R, a few interrelated, crystal structures were described. For Y, Yb and La to Sm, a structure related to the body-centered tetragonal  $\text{BaAl}_4$ -type was proposed. For this structure, the following positions are occupied in the space group  $I4/mmm$ : Ba in  $2a$  and Al in  $4d + 4e$ .

According to Hulliger [1995Hul] a partially ordered structure 1:1:3 is exhibited by  $\text{CeCuAl}_3$  [Ce in  $2a$ , Al in  $4d$  and (Cu + Al) in  $4e$ ], in the same space group  $I4/mmm$ ,  $\text{CeCuAl}_3$ -type. A derivative fully-ordered structure ( $\text{BaNiSn}_3$ -type) may be described corresponding, in the space group  $I4mm$ , to Ba in  $2a$ , Ni in  $2a$ , and Sn in  $2a + 4b$ . According to Hulliger [1995Hul], prolonged annealing at a relatively low temperature would convert the  $\text{CeCuAl}_3$ -type ordering into the complete  $\text{BaNiSn}_3$ -type order. The results of a refinement of neutron powder diffraction patterns showed that this  $\text{CeCuAl}_3$  compound does not crystallize as a disordered form of the  $\text{ThCr}_2\text{Si}_2$  structure but belongs to the more ordered  $\text{BaNiSn}_3$ -type [1996Moz]. For  $\text{GdCuAl}_3$  the crystal structure described as  $\text{BaAl}_4$  by [1988Pre] and assumed to be  $\text{BaNiSn}_3$ -type by [1994Mul], was not confirmed by [2001Gum]; however the compound  $\text{Gd}_3\text{Cu}_{2.1}\text{Al}_{8.9}$  with a closely similar composition and the related  $oI28$   $\text{La}_3\text{Al}_{11}$ -type structure, was found. Similar behavior was observed and/or proposed for the  $\sim\text{RCuAl}_3$  phases with the heavy rare earths (Gd to Ho) with the exception of Yb. For this element and Lu, in the same range of composition, a phase with the  $cF116$ - $\text{Th}_6\text{Mn}_{23}$  was observed instead [1993Ste2 and 1992Kuz1]. Notice that, previously, the structure type  $oI10$ - $\text{CeNi}_{2+x}\text{Sb}_{2-x}$  ( $\text{Ce}_3\text{Ni}_7\text{Sb}_5$ ,  $\text{HoCuAl}_3$  etc), a  $\text{BaAl}_4$ -type variant, was also proposed for these phases in the system with the heavy rare earths [1988Kuz]. Subsequently, however, [1995Kuz] discussed the structure of the Tb compounds and proposed the structure  $\text{La}_3\text{Al}_{11}$  for the  $\text{Tb}_3\text{Cu}_{1.2}\text{Al}_{9.8}$  composition, while [2000Ste] determined the structure  $\text{La}_3\text{Al}_{11}$  for  $\text{Dy}_3\text{Cu}_{2.6}\text{Al}_{8.4}$ ,  $\text{Ho}_3\text{Cu}_{2.4}\text{Al}_{8.6}$  and [2003Kra] found the solid solution  $\text{Y}_3\text{Cu}_{2.7-2.0}\text{Al}_{8.3-9.0}$  with a crystal structure of the  $\text{La}_3\text{Al}_{11}$ -type. Probably the same structure is also formed by the other heavy rare earths.

*PuNi<sub>3</sub>-Type Phases.* Notice that for compositions close to 25 at.% R, different structures were described for a few R-compounds ( $cF16$ - $\text{MnCu}_2\text{Al}$  for Sc [1996Nak], and an unknown hexagonal structure for  $\sim\text{La}_3\text{Cu}_5\text{Al}_4$  [1986Zhu]). For Y and all the R from Gd to Lu the  $hR36$   $\text{PuNi}_3$ -type (or  $\text{NbBe}_3$ -type) was observed ([1992Kuz2] and [2001Gum]). In a refinement of the structure of  $\sim\text{HoCuAl}_2$  the following positions were observed [1992Kuz2] in the space group  $R\bar{3}m$ : Ho in  $3a + 6c$ , Cu in  $6c$ , Al in  $3b$  and (Al + Cu) in  $18h$ . The large atoms have coordination number 16 and 20, and the others 12. Moreover [2003Kra] suggested for the  $\text{Y}_3\text{Cu}_2\text{Al}_7$  compound a completely ordered structure of the  $\text{Ca}_3\text{Cu}_2\text{Al}_7$ -type (superstructure of  $\text{PuNi}_3$ -type).

*$\sim\text{RCuAl}$  Phases.* For nearly all the rare earths, the phases

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RCuAl, hP9 ZrNiAl-type (Fe<sub>2</sub>P derivative), were described by [1968Dwi]. The coordination number ranges from 9 to 18.

For compositions close to 1:1:1, the following structures were also observed: hP93 Ca<sub>30</sub>Cu<sub>25</sub>Al<sub>38</sub> for ~EuCu<sub>0.83</sub>Al<sub>1.27</sub>, the MgZn<sub>2</sub>-type for scandium and the MgCu<sub>2</sub>-type (as a high-pressure form) for Sm, Gd, Tb, Dy, Tm, Yb, and Lu [1992Yan, 1965Tes and 1987Tsv]. A MgNi<sub>2</sub>-type phase was observed for RCu<sub>0.6</sub>Al<sub>1.4</sub> (R = Sc, Eu) [1996Nak (cited by Red Book), 1992Yan].

**3.2.1 Comments About the Crystallo-Chemistry of the R-Cu-Al Phases.** A first comment concerns the smooth trend we may notice, as a function of the R concentration, in the coordination number around the R atoms themselves. We have (see Table 6 and the previous comments) higher and higher values of this number as the concentration of the partner atoms increases and the R concentration decreases. Together with high stoichiometric ratios, we reach values such as 24, 22, 20, etc. for the number of atoms arranged around the large R atoms.

Another remark may arise from a comparison between these R-Me<sub>1</sub>-Me<sub>2</sub> alloys and selected binary R-Me alloys where a single partner is combined with the R. Particularly interesting may be a comparison with the binary alloys of “divalent” metals (Mg, Zn, Cd etc.). A peculiarity of these alloys is that compounds are generally formed in the composition range with R ≤ 50 at.% down to very small R concentrations.

In the case of Zn, for instance, a number of the following stoichiometries (and crystal structures) may be found: RZn (*cP2*-CsCl and *tP2*-HgMn types), RZn<sub>2</sub> (*oI12*-CeCu<sub>2</sub>), RZn<sub>3</sub> (*oP16*-YZn<sub>3</sub>), R<sub>3</sub>Zn<sub>11</sub> (*oI28*-La<sub>3</sub>Al<sub>11</sub>), R<sub>6</sub>Zn<sub>23</sub> (*cF116*-Th<sub>6</sub>Mn<sub>23</sub>), R<sub>13</sub>Zn<sub>58</sub> (*hP142*-Gd<sub>13</sub>Zn<sub>58</sub>), RZn<sub>5</sub> (*hP6*-CaCu<sub>5</sub> and *hP36*-ErZn<sub>5</sub>), R<sub>3</sub>Zn<sub>22</sub> (*tI100*-Pu<sub>3</sub>Ni<sub>22</sub>), R<sub>2</sub>Zn<sub>17</sub> (*hR57*-Th<sub>2</sub>Zn<sub>17</sub> and *hP38*-Th<sub>2</sub>Ni<sub>17</sub>), RZn<sub>11</sub> (*tI48*-BaCd<sub>11</sub> or *hP42*-SmZn<sub>11</sub>), RZn<sub>12</sub> (*tI26*-ThMn<sub>12</sub>), RZn<sub>13</sub> (*cF112*-NaZn<sub>13</sub>). We see that this sequence has several similarities (in stoichiometries, crystal structures) with that previously described for the R-(Cu,Al) alloys. It may perhaps have some significance that, on average, we have nearly the same valence electron number per each atom of the partner (or of the combination of the two partners): two electrons every Zn atom and for a (Cu,Al) mixture, an average number between one and three for each atom.

### 3.3 Phase Equilibria in the Ternary R-Cu-Al Systems

In the following, for a number of systems, the phase equilibria will be presented mainly as results from isothermal section determinations. For each system, a few comments will be reported; some preliminary general remarks, however, may be useful. As explained in the previous paragraphs these systems are quite complex and some uncertainties and/or ambiguities currently exist in their diagrams.

We have to remark that generally their analysis required long and accurate investigations, which in many cases, were performed by preparing 100 or even 200 alloys for one system. Unfortunately, very few details were often given about the experimental procedures, so for instance, no list of the prepared alloys was reported. It is not generally known,

therefore, for a particular tie-triangle in the isothermal sections, how many alloys were included in it. Therefore it may be difficult to understand whether such a triangle is firmly established by a number of mutually consistent observations, or on the other hand, it is a weak spot of the section.

In addition, in the literature, comments are often lacking about the real coherency of the data with those given in previous papers or with those accepted for the binary systems involved. As a consequence we have found, in a number of cases, disagreements and inconsistencies that are not easily explicable and correctable. If, for instance, we have binary diagrams involving a phase sequence in disagreement with the accepted literature data for the given temperature, it is not clear whether we have to adjust this sequence and the equilibria involved, or if we have to modify the temperature value. It was decided therefore to report the various data as found in the literature without any substantial change, adding in specific cases some comments.

For the ternary R-Cu-Al systems, partial or complete isothermal sections were determined for the large majority of the rare earths. These are reported in Fig. 4(a-d). For a few systems (R = Sc, Y, La, Ce, Nd) data concerning the liquidus were also reported; a summary of this information is presented in Fig. 5(a) and 5(b). The crystal structures of the ternary phases are listed in Table 6.

For the different systems the following comments may be noteworthy.

**3.3.1 Sc-Cu-Al.** Several investigations of the phase equilibria in this system were performed. The isothermal section at 500 °C between 0-33 at.% Sc, proposed by [1976Pre] is shown in Fig. 4(a). Note that the Al-Cu edge is not in agreement with the accepted Al-Cu system. Several ternary phases were identified. The following phases were described as compounds of fixed stoichiometry: ~ScCu<sub>4</sub>Al<sub>3</sub> (unknown structure), ~Sc<sub>3</sub>Cu<sub>13</sub>Al<sub>4</sub> (unknown structure), ~ScCu<sub>3</sub>Al<sub>2</sub> (unknown structure), Sc<sub>3</sub>Cu<sub>8</sub>Al<sub>7</sub> (unknown structure), τ<sub>12</sub>-ScCu<sub>2</sub>Al<sub>2</sub> (orthorhombic), ~ScCu<sub>1.4</sub>Al<sub>0.6</sub> (unknown structure), τ<sub>20</sub>-ScCuAl (MgZn<sub>2</sub>-type), τ<sub>16</sub>-ScCu<sub>2</sub>Al (MnCu<sub>2</sub>Al-type), while the τ<sub>2</sub>-ScCu<sub>6.6-4.0</sub>Al<sub>5.4-8.0</sub> (ThMn<sub>12</sub>-type) was a solid solution. Subsequently Gladyshevskii et al. [1992Gla] investigated the crystal structures of ~Sc<sub>3</sub>Cu<sub>13</sub>Al<sub>4</sub>, Sc<sub>3</sub>Cu<sub>8</sub>Al<sub>7</sub>, and ~ScCu<sub>3</sub>Al<sub>2</sub>, refining their composition to τ<sub>8</sub>-Sc<sub>2</sub>Cu<sub>7.5</sub>Al<sub>3.5</sub> (hexagonal), τ<sub>9</sub>-Sc<sub>3</sub>Cu<sub>9</sub>Al<sub>7</sub> (orthorhombic), and τ<sub>10</sub>-Sc<sub>6</sub>Cu<sub>16.4</sub>Al<sub>13.9</sub> (hexagonal), respectively.

Later [1996Nak] investigated the crystal structure of the τ<sub>21</sub>-ScCu<sub>0.6</sub>Al<sub>1.4</sub> (MgNi<sub>2</sub>-type) compound using XRD on a single crystal obtained by annealing an Sc<sub>30</sub>Cu<sub>30</sub>Al<sub>40</sub> alloy for 1000 h at 500 °C. The Al-rich region of the isothermal sections at 450 and 500 °C were subsequently studied by [1991Kha] (see Fig. 4a): solid solution ranges were investigated and a different trend of the tie-lines from that of [1976Pre] was suggested; in particular, a tie-line connecting Al with a ternary phase (ScCu<sub>4.5</sub>Al<sub>5.8</sub> probably phase τ<sub>2</sub> of Table 6) was proposed. A similar trend was suggested also by Yunusov [1992Yun], who studied the composition region between Al-CuAl<sub>2</sub>-ScCuAl-ScAl<sub>2</sub> investigating some polythermal sections and the liquidus surface (see Fig. 5a). The coordinates of the invariant points are listed in Table 7. Notice that in this composition region Yunusov's version is

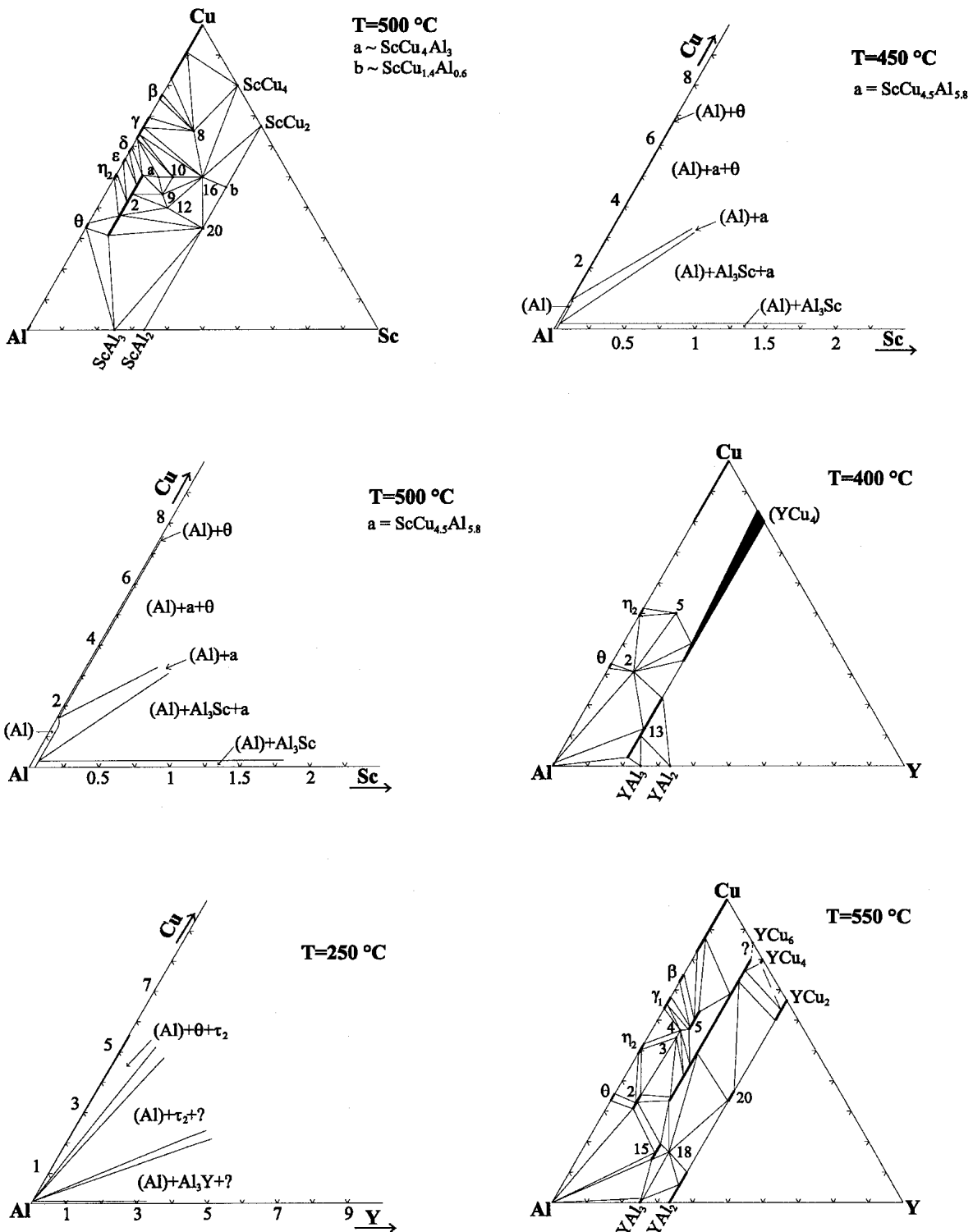


Fig. 4(a) Isothermal sections at the stated temperatures of the ternary R-Cu-Al systems: R = Sc, Y. The identification numbers of the different phases correspond to those listed in Table 6.

different from that by [1976Pre]; we have tie-lines connecting Al with  $\tau_2$  (solid solution ThMn<sub>12</sub>-type) and  $\tau_2$  not only with ScAl<sub>3</sub>, but also with ScAl<sub>2</sub>.

Toropova [1992Tor] studied a part of the Al-rich region up to about 1.2 at.% Sc. The solidus surface of Sc-Cu-Al system was studied in the Al-rich range.

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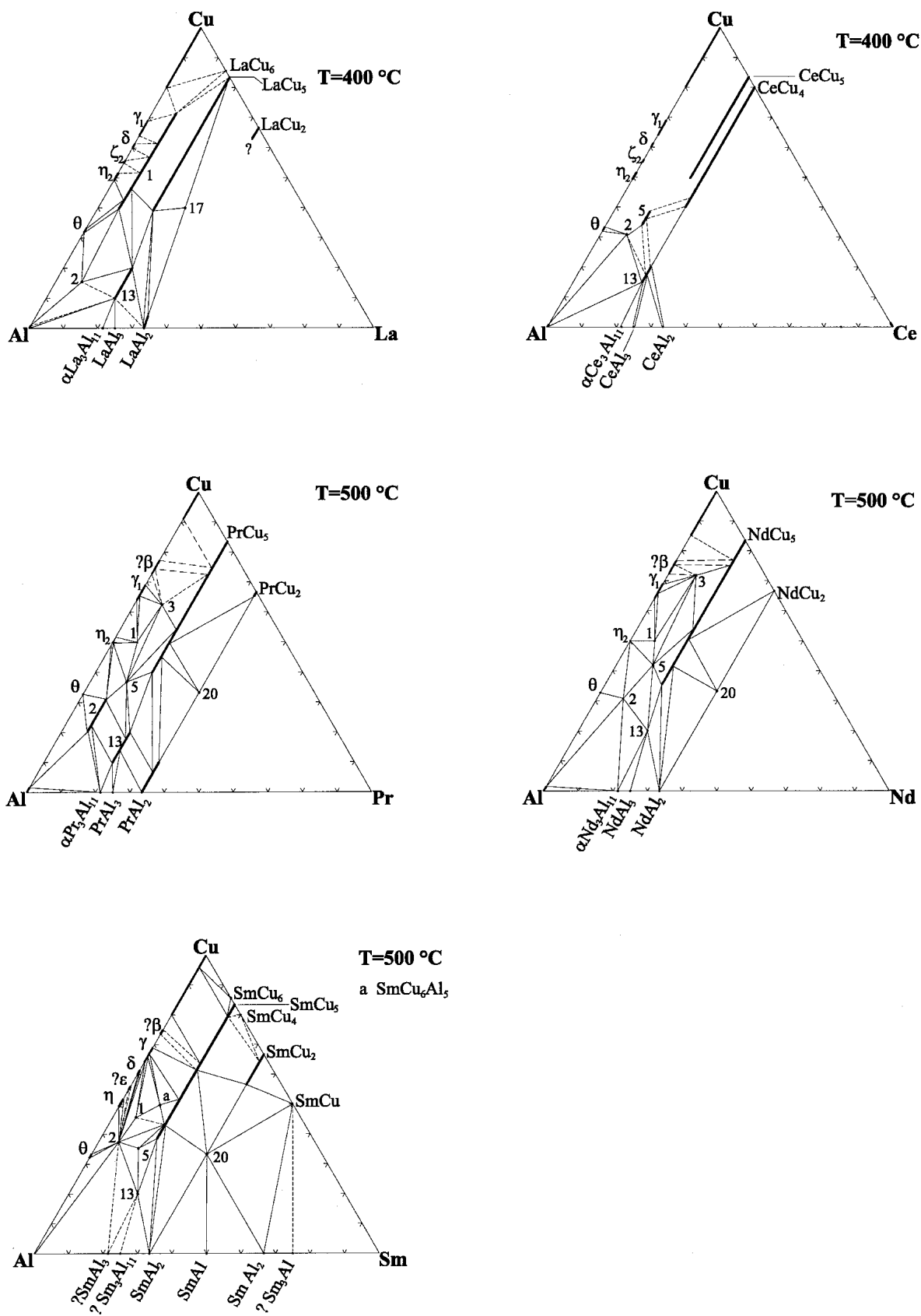


Fig. 4(b) Isothermal sections at the stated temperatures of the ternary R-Cu-Al systems: R = La, Ce, Pr, Nd, and Sm. The identification numbers of the different phases correspond to those listed in Table 6.

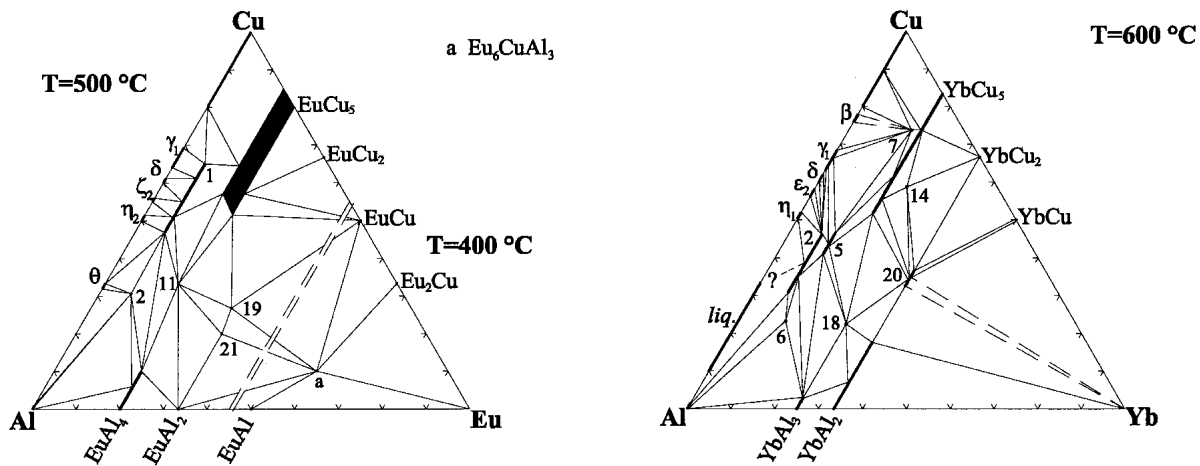


Fig. 4(c) Isothermal sections at the stated temperatures of the ternary R-Cu-Al systems: R = Eu, Yb. The identification numbers of the different phases correspond to those listed in Table 6.

As for the solid solubility of Cu and Sc in Al, indications were given by [1991Kha]; it was observed that the addition of the third element reduces the solubility of Sc in Al and of Cu in Al. The compositions of the doubly-saturated (Al) solid solution were given as follows at 500 °C: in equilibrium with  $\text{CuAl}_2$  and  $\tau_2$  1.6 at.% Cu and 0.015 at.% Sc; in equilibrium with  $\text{ScAl}_3$  and  $\tau_2$  0.21 at.% Cu and 0.045 at.% Sc.

**3.3.2 Y-Cu-Al.** A partial isothermal section at 400 °C from 0 to about 30 at.% Y, constructed by [1968Zar], is reported in Fig. 4(a). Notice that an indication of the  $\sim\text{YCu}_7$  phase is missing, the binary compound reported as  $\text{YCu}_4$  ( $\text{CaCu}_5$ -type) dissolves up to ~45 at.% Al, and that the solid solubility of the  $\tau_{13}$  is included within the range  $\text{YCu}_{1.0.25}\text{Al}_{3.3.75}$  ( $\text{BaAl}_4$ -type). Moreover, there are two compositionally invariant compounds:  $\tau_2$ - $\text{YCu}_4\text{Al}_8$  ( $\text{ThMn}_{12}$ -type) and  $\tau_5$ - $\text{Y}_2\text{Cu}_9\text{Al}_8$  ( $\text{Th}_2\text{Zn}_{17}$ -type).

Two isothermal sections at 250 and 540 °C were presented by [1971Dri] in the Al-rich corner (from 95-100 at.% Al); the 250 °C section is shown in Fig. 4(a).

Two other compounds are known for this system:  $\tau_{20}$ - $\text{YCuAl}$  ( $\text{ZrNiAl}$ -type) and  $\tau_{18}$ - $\text{YCu}_{0.9}\text{Al}_{2.1}$  ( $\text{PuNi}_3$ -type) studied by [1968Dwi] and [1992Kuz2], respectively.

Later [2003Kra] studied this system as well and constructed the isothermal section at 550 °C up to 33.3 at.% Y (Fig. 4a). Notice that the binary Al-Cu edge is not in agreement with the accepted Al-Cu binary phase diagram; the phase  $\text{YCu}_6$  is reported instead of  $\sim\text{YCu}_7$  and the  $\text{Y}_2\text{Cu}_7$  phase is missing.  $\text{YCu}_2$  dissolves up to ~6.7 at.% Al,  $\text{YAl}_2$  up to ~10 at.% Cu, and it is not clear whether  $\text{YCu}_6$  dissolves aluminum up to ~50 at.% or if this solid solution ( $\text{CaCu}_5$ -type) is a separate compound. Notice that the solid solution suggested by [1968Zar] as an extension of the binary compound  $\text{YCu}_4$  is probably based on  $\text{YCu}_6$  with the structure  $\text{TbCu}_7$ -type closely related to the  $\text{CaCu}_5$ -type. According to [2003Kra], the dissolution of Al in the compound  $\text{YCu}_6$  could cause the disappearance of crystallographic defects in the structure and a transition from the  $\text{TbCu}_7$  to the  $\text{CaCu}_5$ -type.

In this system there are four ternary solid solutions:  $\tau_2$ - $\text{YCu}_{4.6.4.0}\text{Al}_{7.4.8.0}$  ( $\text{ThMn}_{12}$ -type),  $\tau_5$ - $\text{Y}_2\text{Cu}_{12.0.10.5}\text{Al}_{5.0.6.5}$  ( $\text{Th}_2\text{Zn}_{17}$ -type),  $\tau_{20}$ - $\text{YCu}_{1.0.1.1}\text{Al}_{1.0.0.9}$  ( $\text{ZrNiAl}$ -type)—all previously reported as compositionally invariant—and  $\tau_{15}$ - $\text{Y}_3\text{Cu}_{2.7.2.0}\text{Al}_{8.3.9.0}$  ( $\text{La}_3\text{Al}_{11}$ -type)—never before reported. There are three additional ternary compounds of fixed stoichiometry:  $\tau_4$ - $\text{YCu}_{6.8}\text{Al}_{4.2}$  [ $\text{Tb}(\text{Cu}_{0.58}\text{Al}_{0.42})_{11}$ -type],  $\tau_3$ - $\text{YCu}_{6.5}\text{Al}_{4.5}$  ( $\text{BaCd}_{11}$ -type), and  $\tau_{18}$ - $\text{Y}_3\text{Cu}_2\text{Al}_7$  ( $\text{Ca}_3\text{Cu}_2\text{Al}_7$ -type superstructure of  $\text{PuNi}_3$ -type), previously reported as  $\text{YCu}_{0.9}\text{Al}_{2.1}$  ( $\text{PuNi}_3$ -type).

In an investigation by [1988Yun], a few vertical sections of the diagram were presented. As for the liquidus, [1992Ran] presented a somewhat speculative version for the Al corner. It was constructed by using the data from [1972Dri] (liquidus surface) and [1988Yun] (vertical section).

Subsequently, a slightly different and more complete version of the liquidus, from ~65 to 100 at.% Al, was presented by [1993Yun]; part of this version is shown in Fig. 5(a), after some inconsistent data close to the Al-Y edge were deleted. The co-ordinates of the invariant points are listed in Table 8.

**3.3.3 La-Cu-Al.** The isothermal section at 400 °C from 0 to 33 at.% La, constructed by [1969Zar] and reported by [1991Ran1] is shown in Fig. 4(b). The phases  $\tau_{17}$ - $\text{La}_3\text{Cu}_5\text{Al}_4$  (hexagonal) and  $\tau_2$ - $\text{LaCu}_2\text{Al}_{10}$  ( $\text{ThMn}_{12}$ -type) were reported as compositionally invariant, while  $\tau_1$  and  $\tau_{13}$  are solid solutions,  $\text{La}(\text{Cu}_{1-x}\text{Al}_x)_{13}$   $0.23 \leq x \leq 0.58$  ( $\text{NaZn}_{13}$ -type) and  $\text{La}(\text{Cu}_{1-x}\text{Al}_x)_4$   $0.75 \leq x \leq 0.875$  ( $\text{BaAl}_4$ -type), respectively.

By preparing and using a number of polythermal sections, Yunusov et al. [1990Yun] developed the liquidus surface in the Al corner (see Fig. 5b; the co-ordinates of the invariant points are listed in Table 9), and determined the congruent melting points of  $\tau_{13}$ - $\text{LaCu}_{0.5}\text{Al}_{3.5}$  and  $\tau_2$ - $\text{LaCu}_2\text{Al}_{10}$  (1000 °C).

The quasi-binary  $\text{LaAl}_2$ - $\text{LaCu}_5$  section, determined by [1986Zhu], shows the congruent melting (~1170 °C) of the phase  $\tau_{17}$ - $\text{La}_3\text{Cu}_5\text{Al}_4$  (described as a solid solution range at



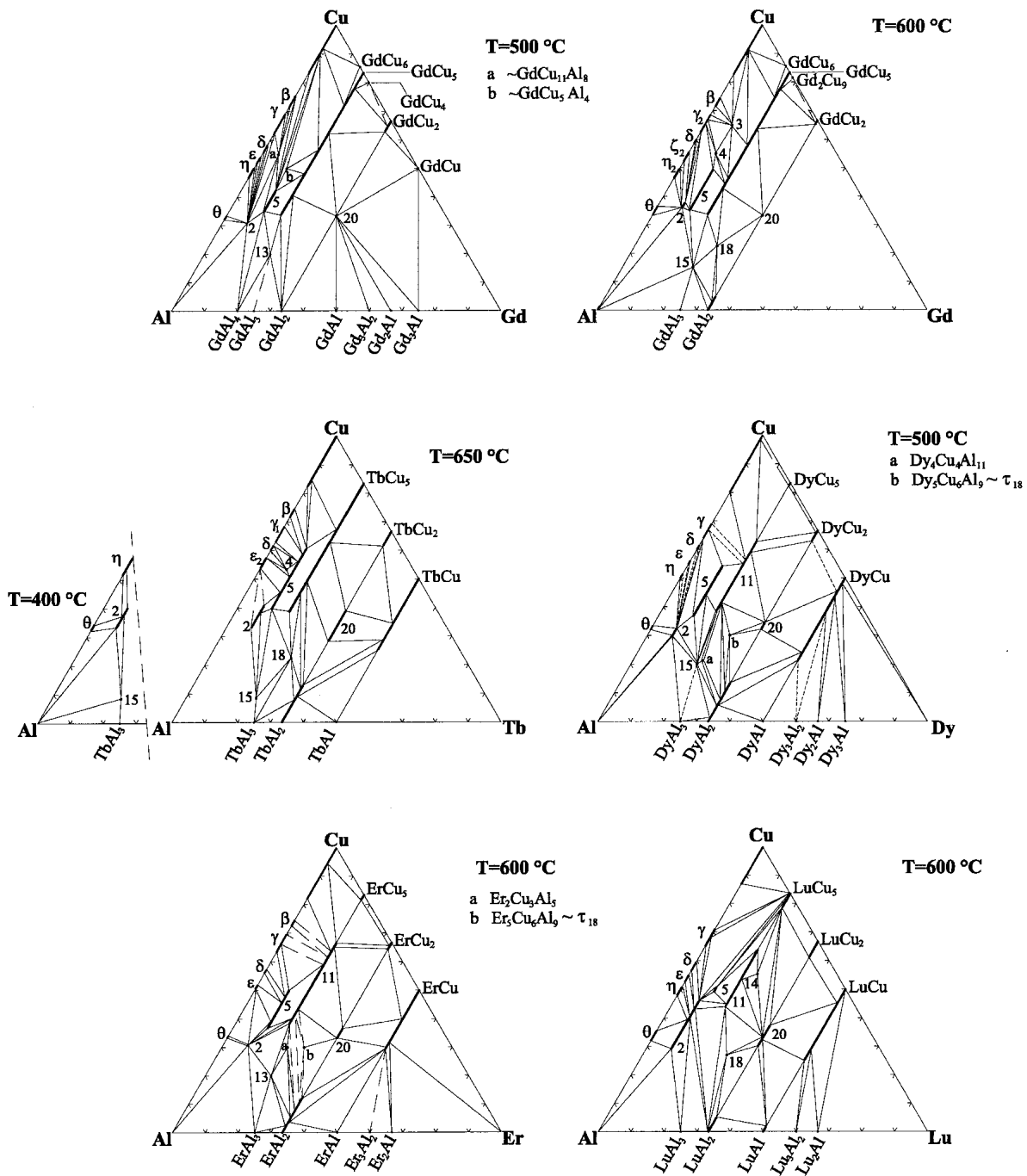


Fig. 4(d) Isothermal sections at the stated temperatures of the ternary R-Cu-Al systems: R = Gd, Tb, Dy, Er, Lu. The identification numbers of the different phases correspond to those listed in Table 6. For comments concerning the binary and ternary phases shown and for a number of uncertainties and disagreement with other literature data, see the text.

high temperature) giving two eutectics with the two binary compounds.

**3.3.4 Ce-Cu-Al.** The partial isothermal section at 400 °C, constructed by [1961Gla], is shown in Fig. 4(b), with the addition, according to [1991Rog] of the  $\text{CeCu}_5$ -based solid solution phase, which dissolves up to 33.3 at.% Al. In this isothermal section, there are a binary compound  $\text{CeCu}_4$ ,

which dissolves up to about 40 at.% Al; two ternary compounds with a composition range  $\tau_5\text{-Ce}_2\text{Cu}_{7.3-6.5}\text{Al}_{9.7-10.5}$  ( $\text{Th}_2\text{Zn}_{17}$ -type) and  $\tau_{13}\text{-CeCu}_{1-0.75}\text{Al}_{3-3.25}$  ( $\text{BaAl}_4$ -type); and a compound  $\tau_2\text{-CeCu}_4\text{Al}_8$  ( $\text{ThMn}_{12}$ -type) of fixed composition. Moreover, two other ternary compounds are known for this system:  $\tau_{20}\text{-CeCuAl}$  ( $\text{ZrNiAl}$ -type) and  $\tau_1\text{-CeCu}_{6.5}\text{Al}_{6.5}$  ( $\text{NaZn}_{13}$ -type) ([1968Dwi, 1980Fel and 1985Cor1,2]).

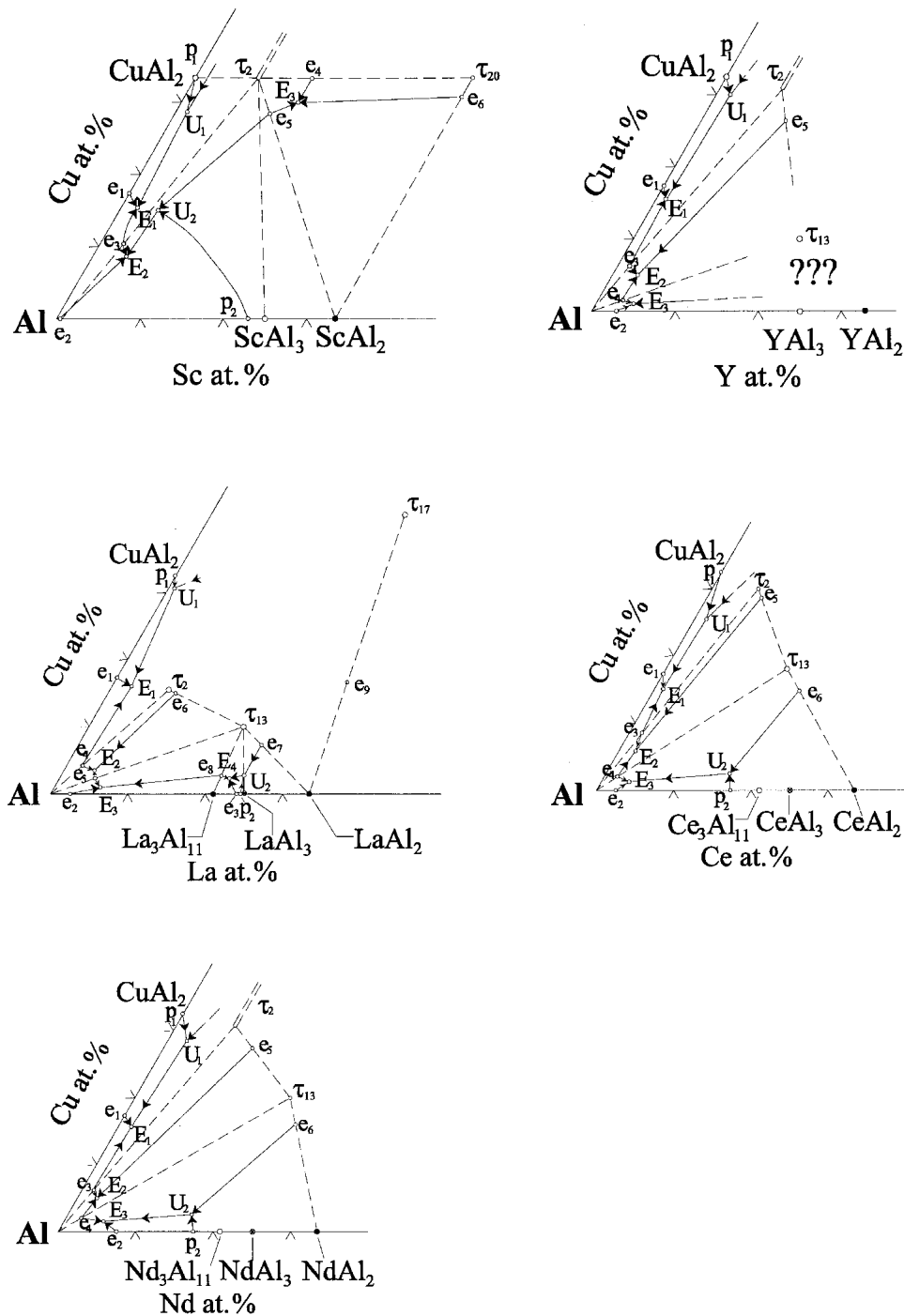


Fig. 5 R-Cu-Al liquidus surfaces in the Al-rich corner: R = Sc, Y, La, Ce, Nd, where (●) = congruent melting, (○) = peritectic formation, (⊗) = solid state reaction

A number of vertical sections of the phase diagram, Al-CeCu<sub>4</sub>Al<sub>8</sub>, Al-CeCuAl<sub>4</sub>, CeCu<sub>4</sub>Al<sub>8</sub>-CeCuAl<sub>4</sub>, and Ce-CuAl<sub>4</sub>-CeAl<sub>2</sub>, all showing a simple eutectic behavior were studied by [1991Yun]. Notice the different composition Ce-CuAl<sub>4</sub> suggested for the  $\tau_{13}$  phase. A projection of the liquidus surface was obtained by [1991Yun] and a sketch of this liquidus is shown in Fig. 5(b) with slight adjustments in the positions of the ternary phases and of the related eutec-

tics, to have them in better agreement with their accepted compositions. For the CeCuAl composition a peritectic decomposition between 600 and 650 °C was suggested [2001Che].

**3.3.5 Pr-Cu-Al.** The partial isothermal section at 500 °C, according to [1978Zar], is shown in Fig. 4(b). Notice that the binary Al-Cu edge is not in agreement with the accepted Al-Cu binary phase diagram; moreover, in the Pr-

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**Table 7 Invariant Equilibria in the Al-rich Corner of the Sc–Cu–Al System (Data From [1992Yun])**

Invariant Point	Reaction Type	Temperature, °C	Liquid Composition, at. %		
			Sc	Cu	Al
U <sub>1</sub>	L + CuAl ↔ CuAl <sub>2</sub> + τ <sub>2</sub>	565	1	29	70
U <sub>2</sub>	L + ScAl <sub>2</sub> ↔ ScAl <sub>3</sub> + τ <sub>2</sub>	850	5	15	80
E <sub>1</sub>	L ↔ Al + τ <sub>2</sub> + CuAl <sub>2</sub>	530	2	15	83
E <sub>2</sub>	L ↔ Al + ScAl <sub>3</sub> + τ <sub>2</sub>	530	4	9	87
E <sub>3</sub>	L ↔ ScAl <sub>2</sub> + τ <sub>20</sub> + τ <sub>2</sub>	898	14	30	56
e <sub>1</sub>	L ↔ Al + CuAl <sub>2</sub>	548	0	17	83
e <sub>2</sub>	L ↔ Al + ScAl <sub>3</sub>	655	0.5	0	99.5
e <sub>3</sub>	L ↔ Al + τ <sub>2</sub>	560	3	10	87
e <sub>4</sub>	L ↔ τ <sub>2</sub> + τ <sub>20</sub>	920	14	33	53
e <sub>5</sub>	L ↔ ScAl <sub>2</sub> + τ <sub>2</sub>	850?(a)	11	28	60
e <sub>6</sub>	L ↔ ScAl <sub>2</sub> + τ <sub>20</sub>	925	33	31	36
p <sub>1</sub>	L + CuAl ↔ CuAl <sub>2</sub>	591	0	33	67
p <sub>2</sub>	L + ScAl <sub>2</sub> ↔ ScAl <sub>3</sub>	1322	23 (b)	0	77

τ<sub>2</sub> = ScCu<sub>6,6-4</sub>Al<sub>5,4-8</sub> and τ<sub>20</sub> = ScCuAl  
(a) notice that this temperature value is inconsistent with E<sub>3</sub> and U<sub>2</sub> temperatures  
(b) subsequently ~21 was proposed [1999Cac]

**Table 8 Invariant Equilibria in the Al-rich Corner of the Y–Cu–Al System (Data From [1993Yun])**

Invariant Point	Reaction Type	Temperature, °C	Liquid Composition, at. %		
			Y	Cu	Al
U <sub>1</sub>	L + CuAl ↔ CuAl <sub>2</sub> + τ <sub>2</sub>	576	2	30	68
E <sub>1</sub>	L ↔ Al + τ <sub>2</sub> + CuAl <sub>2</sub>	545	1	16	83
E <sub>2</sub>	L ↔ Al + τ <sub>2</sub> + τ <sub>13</sub>	607	3	5	92
E <sub>3</sub>	L ↔ Al + τ <sub>13</sub> + YAl <sub>3</sub>	605	4	1	95
e <sub>1</sub>	L ↔ Al + CuAl <sub>2</sub>	548	0	17	83
e <sub>2</sub>	L ↔ Al + YAl <sub>3</sub>	637	3	0	97
e <sub>3</sub>	L ↔ Al + τ <sub>2</sub>	615	2	6	92
e <sub>4</sub>	L ↔ Al + τ <sub>13</sub>	615	3	2	95
e <sub>5</sub>	L ↔ τ <sub>2</sub> + τ <sub>13</sub>	910	10	27	63
p <sub>1</sub>	L + CuAl ↔ CuAl <sub>2</sub>	591	0	33	67

τ<sub>2</sub> = YCu<sub>4</sub>Al<sub>8</sub> and τ<sub>13</sub> = YCu<sub>0,5</sub>Al<sub>3,5</sub>

Cu edge, the PrCu<sub>6</sub> and PrCu<sub>4</sub> phases are missing. In this phase diagram there are four compounds of fixed stoichiometry: τ<sub>20</sub>-PrCuAl (ZrNiAl-type), τ<sub>1</sub>-PrCu<sub>7</sub>Al<sub>6</sub> (NaZn<sub>13</sub>-type), τ<sub>5</sub>-Pr<sub>2</sub>Cu<sub>7</sub>Al<sub>10</sub> (Th<sub>2</sub>Zn<sub>17</sub>-type) and τ<sub>3</sub>-PrCu<sub>7,6</sub>Al<sub>3,4</sub> (BaCd<sub>11</sub>-type). There are also two solid solutions: τ<sub>2</sub>-Pr(Cu<sub>x</sub>Al<sub>1-x</sub>)<sub>12</sub> with *x* ranging from 0.22-0.33 (ThMn<sub>12</sub>-type); and τ<sub>13</sub>-Pr(Cu<sub>x</sub>Al<sub>1-x</sub>)<sub>4</sub> with 0.125 ≤ *x* ≤ 0.25 (BaAl<sub>4</sub>-type). PrCu<sub>5</sub> dissolves up to ~45 at. % Al, PrCu<sub>2</sub> up to about 5 at. % Al, and PrAl<sub>2</sub> up to 10 at. % Cu.

Pr-rich alloys were studied by [2002Zha2] through differential scanning calorimetry; a ternary eutectic, L ↔ Pr + PrCu + Pr<sub>3</sub>Al, was found at the composition Pr 68 at. %-Cu 25 at. %-Al 7 at. % with an invariant temperature of 432 °C.

**3.3.6 Nd-Cu-Al.** The isothermal section at 500 °C in the composition range 0-33 at. % Nd, prepared by [1975Zar], is shown in Fig. 4(b). Notice that the binary Al-Cu edge is not in agreement with the accepted Al-Cu binary phase diagram, and in the Nd-Cu edge, the NdCu<sub>6</sub> and NdCu<sub>4</sub> phases are missing. Six compositionally invari-

ant compounds are reported: τ<sub>20</sub>-NdCuAl (ZrNiAl-type), τ<sub>1</sub>-NdCu<sub>7</sub>Al<sub>6</sub> (NaZn<sub>13</sub>-type), τ<sub>13</sub>-NdCuAl<sub>3</sub> (BaAl<sub>4</sub>-type), τ<sub>2</sub>-NdCu<sub>4</sub>Al<sub>8</sub> (ThMn<sub>12</sub>-type), τ<sub>5</sub>-Nd<sub>2</sub>Cu<sub>8</sub>Al<sub>9</sub> (Th<sub>2</sub>Zn<sub>17</sub>-type), τ<sub>3</sub>-NdCu<sub>8,6</sub>Al<sub>2,4</sub> (BaCd<sub>11</sub>-type). NdCu<sub>5</sub> dissolves up to ~50 at. % Al. A number of polythermal sections, showing a simple eutectic aspect, were studied by [1987Yun]: Al-NdCu<sub>4</sub>Al<sub>8</sub>, NdCu<sub>4</sub>Al<sub>8</sub>-NdCuAl<sub>3</sub>, and NdCuAl<sub>3</sub>-NdAl<sub>2</sub>. A projection of a partial liquidus surface prepared by [1993Yun] is shown in Fig. 5(b). Notice that in the Al-Nd edge the α,β Nd<sub>3</sub>Al<sub>11</sub> phases with the related equilibria are missing; the corresponding coordinates of the invariant points are listed in Table 10.

**3.3.7 Sm-Cu-Al.** The isothermal section at 500 °C was studied by [1983Pre]. Notice that some inconsistencies are present in the paper itself. In fact the compositions of the different phases as read from the isothermal sections are in some cases different from those reported in the crystal structure table given in the same paper. Six compounds of fixed stoichiometry have been listed by [1983Pre]: τ<sub>20</sub>-SmCuAl

**Table 9** Invariant Equilibria in the Al-Rich Corner of La-Cu-Al System (Data From [1990Yun] and [1986Zhu])

Invariant Point	Reaction Type	Temperature, (°C)	Liquid Composition, at. %		
			La	Cu	Al
U <sub>1</sub>	L + CuAl ↔ CuAl <sub>2</sub> + τ <sub>2</sub>	582	1	31	68
U <sub>2</sub> (a)	L + LaAl <sub>2</sub> ↔ LaAl <sub>3</sub> + τ <sub>13</sub>	1234	23	3	74
E <sub>1</sub>	L ↔ Al + τ <sub>2</sub> + CuAl <sub>2</sub>	542	3	16	81
E <sub>2</sub>	L ↔ Al + τ <sub>2</sub> + τ <sub>13</sub>	550	4	4	92
E <sub>3</sub>	L ↔ Al + τ <sub>13</sub> + La <sub>3</sub> Al <sub>11</sub>	590	6	1	93
E <sub>4</sub>	L ↔ LaAl <sub>3</sub> + τ <sub>13</sub> + La <sub>3</sub> Al <sub>11</sub>	885	22	2.5	75.5
e <sub>1</sub>	L ↔ Al + CuAl <sub>2</sub>	548	0	17	83
e <sub>2</sub>	L ↔ Al + La <sub>3</sub> Al <sub>11</sub>	640	2.5	0	97.5
e <sub>3</sub> (b)	L ↔ La <sub>3</sub> Al <sub>11</sub> + LaAl <sub>3</sub>	1220	24	0	76
e <sub>4</sub>	L ↔ Al + τ <sub>2</sub>	580	2.5	4	93.5
e <sub>5</sub>	L ↔ Al + τ <sub>13</sub>	610	4.5	2.5	93
e <sub>6</sub>	L ↔ τ <sub>2</sub> + τ <sub>13</sub>	770	9	15	76
e <sub>7</sub>	L ↔ LaAl <sub>2</sub> + τ <sub>13</sub>	1230	24	7	69
e <sub>8</sub>	L ↔ La <sub>3</sub> Al <sub>11</sub> + τ <sub>13</sub>	900	20.5	3	76.5
e <sub>9</sub>	L ↔ LaAl <sub>2</sub> + La <sub>3</sub> Cu <sub>5</sub> Al <sub>4</sub>	1026	30	17	53
p <sub>1</sub>	L + CuAl ↔ CuAl <sub>2</sub>	591	0	33	67
p <sub>2</sub> (b)	L + LaAl <sub>2</sub> ↔ LaAl <sub>3</sub>	1240	~25	0	75

τ<sub>2</sub> = LaCu<sub>2</sub>Al<sub>10</sub> and τ<sub>13</sub> = LaCu<sub>0.5</sub>Al<sub>3.5</sub>.

(a) Notice some inconsistencies in the temperatures reported for the invariant equilibria close to U<sub>2</sub> (U<sub>2</sub> should be at lower temperature than e<sub>7</sub>).

(b) These invariant equilibria are not in agreement with the accepted binary phase diagram, according to which the phase LaAl<sub>x</sub> (29.3 at.% La) should be involved instead of LaAl<sub>3</sub>.

(ZrNiAl-type), τ<sub>13</sub>-SmCuAl<sub>3</sub> (Al<sub>4</sub>Ba-type), τ<sub>2</sub>-SmCu<sub>4</sub>Al<sub>8</sub> (ThMn<sub>12</sub>-type), τ<sub>1</sub>-SmCu<sub>7</sub>Al<sub>6</sub> (NaZn<sub>13</sub>-type), τ<sub>5</sub>-Sm<sub>2</sub>Cu<sub>7</sub>Al<sub>10</sub> (Th<sub>2</sub>Zn<sub>17</sub>-type), and ~SmCu<sub>6</sub>Al<sub>5</sub> (unknown structure). In drawing the isothermal section, instead of τ<sub>2</sub>-SmCu<sub>4</sub>Al<sub>8</sub>, τ<sub>1</sub>-SmCu<sub>7</sub>Al<sub>6</sub>, τ<sub>5</sub>-Sm<sub>2</sub>Cu<sub>7</sub>Al<sub>10</sub>, and ~SmCu<sub>6</sub>Al<sub>5</sub>, the following compositions are reported: τ<sub>2</sub>-Sm<sub>6</sub>Cu<sub>37.5</sub>Al<sub>56.5</sub>, τ<sub>1</sub>-Sm<sub>7</sub>Cu<sub>46</sub>Al<sub>47</sub>, τ<sub>5</sub>-Sm<sub>13</sub>Cu<sub>35.5</sub>Al<sub>51.5</sub>, and ~Sm<sub>11.5</sub>Cu<sub>50</sub>Al<sub>38.5</sub>. In Fig. 4(b), the isothermal section of the Sm-Cu-Al system is shown as drawn by [1983Pre]. Notice that SmCu<sub>5</sub> dissolves up to ~46 at.% Al while SmCu<sub>2</sub> up to about 10 at.% Al. Finally for the binary boundary systems, it may be observed that Sm<sub>3</sub>Al was not identified in the accepted binary system; the corresponding tie-line is probably wrong. In the original diagram, instead of Sm<sub>3</sub>Al<sub>11</sub>, SmAl<sub>4</sub>, was proposed. In the binary Al-Cu system, notice that the β and ε phases are not stable at 500 °C. The isothermal section of this system was also reported by [1990Gla], but it is not in agreement with the data given by [1983Pre].

**3.3.8 Eu-Cu-Al.** The complete isothermal sections at 400 °C for the composition range 45-100 at.% Eu, and at 500 °C for 0-45 at.% Eu, were constructed by [1992Yan]; they are shown in Fig. 4c. There are five compounds of invariant composition and one solid solution: τ<sub>2</sub>-EuCu<sub>4</sub>Al<sub>8</sub> (ThMn<sub>12</sub>-type), τ<sub>11</sub>-EuCu<sub>2</sub>Al<sub>3</sub> (CeNi<sub>2</sub>Al<sub>3</sub>-type), τ<sub>21</sub>-EuCu<sub>0.6</sub>Al<sub>1.4</sub> (MgNi<sub>2</sub>-type), Eu<sub>6</sub>CuAl<sub>3</sub> (unknown structure), τ<sub>19</sub>-EuCu<sub>0.83</sub>Al<sub>1.27</sub> (which was described by [1988Zar] as Eu<sub>30</sub>Cu<sub>25</sub>Al<sub>38</sub> hP93 Ca<sub>30</sub>Cu<sub>25</sub>Al<sub>38</sub>-type), and τ<sub>1</sub>-EuCu<sub>6.9-9.1</sub>Al<sub>6.8-3.9</sub> (NaZn<sub>13</sub>-type). EuCu<sub>5</sub> dissolves up to ~30 at.% Al, and EuAl<sub>4</sub> up to about 10 at.% Cu. For the solid solution phases, selected values of the lattice parameters are presented according to [1992Yan] in Table 11.

**3.3.9 Gd-Cu-Al.** Isothermal sections of the system were studied by several researchers. A section at 500 °C was prepared by [1988Pre]. This is shown in Fig. 4(d); notice, however, that a slightly different version was published in an assessment by [1991Ran2], taking into account mainly the updated binary systems. In the [1991Ran2] version (not shown here), Gd<sub>2</sub>Cu<sub>9</sub> was inserted instead of GdCu<sub>4</sub>, and Gd<sub>3</sub>Al and GdAl<sub>4</sub> were deleted. In the Cu-Al system, the β phase (not stable at 500 °C) was deleted and the ε phase was substituted by ζ<sub>2</sub>; an indication was also given for the terminal solid solutions. A new version of the partial isothermal section at 600 °C was constructed experimentally by [2001Gum]; this is shown in Fig. 4d. The differences between the two diagrams may be summarized as follows: the phase previously indicated by [1988Pre] as τ<sub>13</sub>-GdCuAl<sub>3</sub> (BaAl<sub>7</sub>-type) was not found by [2001Gum] (neither at 500 nor at 600 °C); a composition τ<sub>15</sub>-Gd<sub>3</sub>Cu<sub>2.1</sub>Al<sub>8.9</sub> was instead proposed for a phase with the La<sub>3</sub>Al<sub>11</sub>-type structure. The phase τ<sub>18</sub>-GdCu<sub>0.9</sub>Al<sub>2.1</sub> (PuNi<sub>3</sub>-type) was identified by [2001Gum]. For the phase with unknown structure previously indicated by [1988Pre] as GdCu<sub>5</sub>Al<sub>4</sub>, the composition τ<sub>3</sub>-GdCu<sub>7.8</sub>Al<sub>3.2</sub> (BaCd<sub>11</sub>-type) was proposed by [2001Gum]. The phase previously indicated as ~GdCu<sub>11</sub>Al<sub>8</sub> in the English version of [1988Pre] (the original Russian version was not available to us) but cited by [2001Gum] as Gd<sub>2</sub>Cu<sub>11</sub>Al<sub>8</sub> was described as τ<sub>4</sub>-GdCu<sub>6.6</sub>Al<sub>4.4</sub> [Tb(Cu<sub>0.58</sub>Al<sub>0.42</sub>)<sub>11</sub>-type]. Finally, notice some disagreement between the accepted binary Cu-Al system and that given in [2001Gum] (Fig. 4d). In the 600 °C section by [2001Gum] no indication was given of the liquid phase in the Al-rich region. The η<sub>2</sub> phase should possibly be η<sub>1</sub>, instead of ζ, the ε phase should be observed (the phase sequence reported would be in better agreement (except for the β phase) with a temperature of ~550 °C instead of 600 °C).

**Table 10 Invariant Equilibria in the Al-Rich Corner of Nd-Cu-Al System (Data From [1993Yun])**

Invariant Point	Reaction Type	Temperature, (°C)	Liquid Composition, at. %		
			Nd	Cu	Al
U <sub>1</sub>	L + CuAl ↔ CuAl <sub>2</sub> + τ <sub>2</sub>	580	2	29	69
U <sub>2</sub>	L + NdAl <sub>3</sub> ↔ Nd <sub>3</sub> Al <sub>11</sub> + τ <sub>13</sub>	1212	16	3	81
E <sub>1</sub>	L ↔ Al + τ <sub>2</sub> + CuAl <sub>2</sub>	539	2	16	82
E <sub>2</sub>	L ↔ Al + τ <sub>2</sub> + τ <sub>13</sub>	609	3	5	92
E <sub>3</sub>	L ↔ Al + Nd <sub>3</sub> Al <sub>11</sub> + τ <sub>13</sub>	598	5	2	93
e <sub>1</sub>	L ↔ Al + CuAl <sub>2</sub>	548	0	17	83
e <sub>2</sub> (a)	L ↔ Al + Nd <sub>3</sub> Al <sub>11</sub>	637	7.5	0	92.5
e <sub>3</sub>	L ↔ Al + τ <sub>2</sub>	620	2	6	92
e <sub>4</sub>	L ↔ Al + τ <sub>13</sub>	615	2	2	96
e <sub>5</sub>	L ↔ τ <sub>2</sub> + τ <sub>13</sub>	885	12	27	61
e <sub>6</sub>	L ↔ τ <sub>13</sub> + NdAl <sub>2</sub>	1225	23	16	61
p <sub>1</sub>	L + CuAl ↔ CuAl <sub>2</sub>	591	0	33	67
p <sub>2</sub> (b)	L + NdAl <sub>2</sub> ↔ Nd <sub>3</sub> Al <sub>11</sub>	1227	17	0	83

τ<sub>2</sub> = NdCu<sub>4</sub>Al<sub>8</sub> and τ<sub>13</sub> = NdCuAl<sub>3</sub>.

(a) Notice that coordinates of this binary invariant equilibrium have been changed by [2001God]. See text paragraph 2.2.6.

(b) Notice that the accepted temperature value of this equilibrium is 1235 °C. See Fig. 3.

**Table 11 Lattice Parameters of the Solid Solution Phases in the Eu-Cu-Al System. Data From [1992Yan]**

Phase	Lattice Parameters (pm)	
	a	c
<b>EuAl<sub>4</sub>-based solid solution t110-BaAl<sub>4</sub></b>		
EuAl <sub>4</sub>	440.2	1116.3
EuCu <sub>0.375</sub> Al <sub>3.625</sub>	428.5	1129.0
EuCu <sub>0.6</sub> Al <sub>3.4</sub>	427.1	1116.7
<b>τ<sub>1</sub>-EuCu<sub>6.9-9.1</sub>Al<sub>6.8-3.9</sub> cF112-NaZn<sub>13</sub></b>		
EuCu <sub>7.0</sub> Al <sub>6.0</sub>	1191.8	
EuCu <sub>7.7</sub> Al <sub>5.3</sub>	1186.9	
EuCu <sub>9.1</sub> Al <sub>3.9</sub>	1182.6	
<b>τ<sub>11</sub>-Eu(Cu<sub>1-x</sub>Al<sub>x</sub>)<sub>5-y</sub> hP6-CaCu<sub>5</sub></b>		
EuCu <sub>4.7</sub> Al <sub>0.3</sub>	522.3	407.1
EuCu <sub>4.1</sub> Al <sub>0.9</sub>	524.4	412.1
EuCu <sub>3.8</sub> Al <sub>1.2</sub>	524.3	409.6
EuCu <sub>3.2</sub> Al <sub>1.8</sub>	528.9	416.5
EuCu <sub>4.28</sub> Al <sub>0.28</sub>	525.8	413.9
EuCu <sub>3.72</sub> Al <sub>0.83</sub>	524.7	412.5
EuCu <sub>3.44</sub> Al <sub>1.11</sub>	526.2	414.6
EuCu <sub>2.89</sub> Al <sub>1.67</sub>	530.4	416.8
EuCu <sub>3.75</sub> Al <sub>0.25</sub>	523.2	409.5
EuCu <sub>3.1</sub> Al <sub>0.9</sub>	525.7	414.5
EuCu <sub>2.5</sub> Al <sub>1.5</sub>	530.9	417.0

In both isothermal sections studied by [1988Pre] and [2001Gum] an indication of the existence of two phases with ThMn<sub>12</sub> and Th<sub>2</sub>Zn<sub>17</sub> crystal structures is given; their composition ranges are τ<sub>2</sub>-GdCu<sub>4.7-4.9</sub>Al<sub>7.3-7.1</sub> [2001Gum] (or GdCu<sub>4</sub>Al<sub>8</sub> [1988Pre]) and τ<sub>5</sub>-Gd<sub>2</sub>Cu<sub>9.4-6.7</sub>Al<sub>7.6-10.3</sub> [2001Gum] (or Gd<sub>2</sub>Cu<sub>8.0-6.7</sub>Al<sub>9.0-10.3</sub> [1988Pre]), respectively.

**3.3.10 Tb-Cu-Al.** A partial isothermal section (from 0 to 50 at.% Tb) was constructed by [1995Kuz] at 650 °C but

at 400 °C in the Al-rich region. This is shown in Fig. 4(d). Note that along the Tb-Cu edge, the Tb<sub>2</sub>Cu<sub>9</sub> compound, assumed to exist by analogy with the other heavy rare earths, is missing; the TbCu<sub>5</sub> compound dissolves up to ~45 at.% Al. Several ternary phases were identified; the τ<sub>4</sub>-TbCu<sub>6.4</sub>Al<sub>4.6</sub>, τ<sub>18</sub>-TbCu<sub>0.9</sub>Al<sub>2.1</sub> and τ<sub>15</sub>-Tb<sub>3</sub>Cu<sub>1.2</sub>Al<sub>9.8</sub> (La<sub>3</sub>Al<sub>11</sub>-type) phases were described as compounds of fixed composition [1995Kuz, 1996Ste]. For the other phases, the following composition ranges were proposed: τ<sub>2</sub>-TbCu<sub>12-x</sub>Al<sub>x</sub> from x = 6.8 to 7.2 (ThMn<sub>12</sub>-type); τ<sub>5</sub>-Tb<sub>2</sub>Cu<sub>17-x</sub>Al<sub>x</sub> from x = 5.5 to 9.4 (Th<sub>2</sub>Zn<sub>17</sub>-type) and τ<sub>20</sub>-TbCu<sub>2-x</sub>Al<sub>x</sub> with 0.85 ≤ x ≤ 1.15 (Fe<sub>2</sub>P or ZrNiAl-type) [1995Kuz]. Subsequently [1996Ste], using single crystal x-ray diffraction, determined the crystal structure of τ<sub>5</sub>-Tb<sub>2</sub>Cu<sub>8</sub>Al<sub>9</sub> as being a Th<sub>2</sub>Zn<sub>17</sub>-type and refined by the Rietveld method the atomic parameters of τ<sub>4</sub>-TbCu<sub>6.4</sub>Al<sub>4.6</sub>, a new structural type related to the BaCd<sub>11</sub>-type. This phase was previously identified by [1995Kuz].

**3.3.11 Dy-Cu-Al.** The isothermal section at 500 °C, studied by [1989Kuz1], is given in Fig. 4(d). Notice that, instead of the binary ζ<sub>2</sub>, the phase ε, not stable at 500 °C, was reported, and that in the Al-Dy edge the indicated compound Dy<sub>3</sub>Al is probably an impurity-stabilized phase. Notice that the Dy<sub>2</sub>Cu<sub>9</sub> compound is missing. Several ternary phases were identified studying this isothermal section: τ<sub>2</sub>-~DyCu<sub>4</sub>Al<sub>8</sub> (ThMn<sub>12</sub>-type), ~DyCuAl<sub>3</sub>, τ<sub>20</sub>-DyCuAl, ~Dy<sub>4</sub>Cu<sub>4</sub>Al<sub>11</sub>, and ~Dy<sub>5</sub>Cu<sub>6</sub>Al<sub>9</sub> were described as compositionally invariant, the last two compounds having unknown structures. For the τ<sub>5</sub> and τ<sub>11</sub> phases, the following solubility ranges were determined: Dy<sub>2</sub>Cu<sub>10.3-7.0</sub>Al<sub>6.7-10</sub> and DyCu<sub>3.8-2.3</sub>Al<sub>1.2-2.7</sub>. [1992Kuz2] investigated the crystal structure of RCu<sub>0.9</sub>Al<sub>2.1</sub> (R = Y, Tb to Lu), PuNi<sub>3</sub>-type, and suggested this structure for the Dy<sub>5</sub>Cu<sub>6</sub>Al<sub>9</sub> compound. Notice that the phase given by [1989Kuz1] as corresponding to the composition ~DyCuAl<sub>3</sub> was subsequently [2000Ste] described as τ<sub>15</sub>-Dy<sub>3</sub>Cu<sub>2.6</sub>Al<sub>8.4</sub> with the La<sub>3</sub>Al<sub>11</sub>-type structure. Finally, as cited by [2001Gum], the crystal structure of

the phase  $\tau_{14}$ -Dy<sub>6</sub>(Cu,Al)<sub>23</sub> was identified as having a cubic Th<sub>6</sub>Mn<sub>23</sub>-type structure by [1990Ste].

**3.3.12 Ho-Cu-Al.** No information concerning the phase diagram for Ho-Cu-Al is available in the literature. Various papers concerning the intermediate phases and their crystal structures were published. The crystal structure of  $\tau_{18}$ -HoCu<sub>0.9</sub>Al<sub>2.1</sub> (PuNi<sub>3</sub>-type) was determined by [1992Kuz2], and other compositionally invariant compounds and one solid solution, reported in an assessment by [1991Ran3], are  $\tau_{20}$ -HoCuAl (ZrNiAl-type), HoCuAl<sub>3</sub> (HoCuAl<sub>3</sub>-type),  $\tau_{11}$ -HoCu<sub>4</sub>Al (CaCu<sub>5</sub>-type),  $\tau_5$ -Ho<sub>2</sub>Cu<sub>7</sub>Al<sub>10</sub> (Th<sub>2</sub>Zn<sub>17</sub>-type) and two compounds (or two alloys in the same solid solution range?) both with the ThMn<sub>12</sub>-type structure:  $\tau_2$ -HoCu<sub>4</sub>Al<sub>8</sub> [1979Fel] and  $\tau_2$ -HoCu<sub>6</sub>Al<sub>6</sub> [1980Fel] (see para. 3.2). Notice that the composition HoCuAl<sub>3</sub> given by [1991Ran3] was subsequently [2000Ste] described as  $\tau_{15}$ -Ho<sub>3</sub>Cu<sub>2.4</sub>Al<sub>8.6</sub> with the La<sub>3</sub>Al<sub>11</sub>-type structure. Finally, as cited by [2001Gum] the crystal structure of  $\tau_{14}$ -Ho<sub>6</sub>(Cu,Al)<sub>23</sub> was identified by [1991Ste] as being of the cubic Th<sub>6</sub>Mn<sub>23</sub>-type structure.

**3.3.13 Er-Cu-Al.** The isothermal section at 600 °C was determined by [1989Kuz2], and it is given in Fig. 4(d). Note that the Al-Cu edge is not in agreement with the accepted binary Al-Cu system and that in the Er-Cu edge the two compounds Er<sub>2</sub>Cu<sub>7</sub> and Er<sub>2</sub>Cu<sub>9</sub> are missing. These two compounds were indicated as ErCu<sub>x</sub> (peritectic formation at 940 °C) and ErCu<sub>y</sub> (congruent melting at 1010 °C) by [1970Bus] and assumed to be Er<sub>2</sub>Cu<sub>7</sub> and Er<sub>2</sub>Cu<sub>9</sub> by [1994Sub] on the basis of melting behavior consistent with that of other similar R-compounds. In this system, there are four compositionally invariant compounds and three ternary solid solutions:  $\tau_{13}$ -ErCuAl<sub>3</sub> (HoCuAl<sub>3</sub>-type), Er<sub>2</sub>Cu<sub>3</sub>Al<sub>5</sub> (unknown structure), Er<sub>5</sub>Cu<sub>6</sub>Al<sub>9</sub> (unknown structure),  $\tau_2$ -ErCu<sub>4</sub>Al<sub>8</sub> (ThMn<sub>12</sub>-type),  $\tau_5$ -Er<sub>2</sub>(Cu<sub>1-x</sub>Al<sub>x</sub>)<sub>17</sub> with  $0.44 \leq x \leq 0.59$  (Th<sub>2</sub>Zn<sub>17</sub>-type),  $\tau_{11}$ -Er(Cu<sub>1-x</sub>Al<sub>x</sub>)<sub>5</sub> with  $0.18 \leq x \leq 0.54$  (CaCu<sub>5</sub>-type), and  $\tau_{20}$ -ErCuAl with a small solubility range (ZrNiAl-type). At 600 °C, ErCu<sub>2</sub> dissolves up to ~1 at.% Al, ErCu up to ~20 at.% Al, and ErAl<sub>2</sub> up to ~13 at.% Cu. Subsequently, [1992Kuz2] found the compound ErCu<sub>0.9</sub>Al<sub>2.1</sub> (PuNi<sub>3</sub>-type) in samples annealed at 600 °C and suggested that probably the compound Er<sub>5</sub>Cu<sub>6</sub>Al<sub>9</sub> with unknown structure was ErCu<sub>0.9</sub>Al<sub>2.1</sub> due to the similarity in composition. As cited by [2001Gum], the crystal structure of the phase  $\tau_{14}$ -Er<sub>6</sub>(Cu,Al)<sub>23</sub> was identified by [1990Ste] as having a cubic Th<sub>6</sub>Mn<sub>23</sub>-type structure.

**3.3.14 Tm-Cu-Al.** No data have been reported on the phase diagram. The crystal structure data of the intermediate phases was presented by [1997Vil], who reported five point compounds:  $\tau_{20}$ -TmCuAl (ZrNiAl-type),  $\tau_{13}$ -TmCuAl<sub>3</sub> (HoCuAl<sub>3</sub>-type),  $\tau_{11}$ -TmCu<sub>4</sub>Al (CaCu<sub>5</sub>-type), and  $\tau_2$ -TmCu<sub>4</sub>Al<sub>8</sub> and  $\tau_2$ -TmCu<sub>6</sub>Al<sub>6</sub> with the same ThMn<sub>12</sub>-type crystal structure (possibly belonging to the same solution range?). Subsequently, crystal data for  $\tau_{18}$ -TmCu<sub>0.9</sub>Al<sub>2.1</sub> (PuNi<sub>3</sub>-type) and for  $\tau_5$ -Tm<sub>2</sub>Cu<sub>10</sub>Al<sub>7</sub> (Th<sub>2</sub>Zn<sub>17</sub>-type) were reported by [1992Kuz2] and [1993Ste1], respectively. Moreover, as cited by [2001Gum], the crystal structure of the phase  $\tau_{14}$ -Tm<sub>6</sub>(Cu,Al)<sub>23</sub> was identified by [1990Ste] as having a cubic Th<sub>6</sub>Mn<sub>23</sub>-type structure.

**3.3.15 Yb-Cu-Al.** The isothermal section at 600 °C was studied by [1993Ste2]. Along the Yb-Cu edge, the two compounds Yb<sub>2</sub>Cu<sub>7</sub> and Yb<sub>2</sub>Cu<sub>9</sub> are missing as well as the liquid-phase. Moreover, along the Cu-Al axis, some discrepancies with the accepted binary system may be noticed; possibly the average temperature of the investigation was slightly lower than 600 °C (metastable equilibria). Some adjustments were performed in this region and are reported as dashed lines in the diagram shown in Fig. 4(c). In this phase diagram, there are the following phases:  $\tau_{20}$ -Yb(Cu<sub>0.55-0.50</sub>Al<sub>0.45-0.5</sub>)<sub>2</sub> (ZrNiAl-type),  $\tau_2$ -Yb(Cu<sub>0.50-0.33</sub>Al<sub>0.50-0.67</sub>)<sub>12</sub> (ThMn<sub>12</sub>-type),  $\tau_{18}$ -YbCu<sub>0.9</sub>Al<sub>2.1</sub> (PuNi<sub>3</sub>-type),  $\tau_5$ -Yb<sub>2</sub>(Cu<sub>0.51-0.46</sub>Al<sub>0.49-0.54</sub>)<sub>17</sub> (Th<sub>2</sub>Zn<sub>17</sub>-type),  $\tau_7$ -YbCu<sub>5.1</sub>Al<sub>0.9</sub> (YbMo<sub>2</sub>Al<sub>4</sub>-type),  $\tau_{14}$ -Yb<sub>6</sub>(Cu<sub>0.74</sub>Al<sub>0.26</sub>)<sub>23</sub> (Th<sub>6</sub>Mn<sub>23</sub>-type), and  $\tau_6$ -Yb<sub>4</sub>(Cu<sub>0.26</sub>Al<sub>0.74</sub>)<sub>33</sub> [Yb<sub>4</sub>(Cu<sub>0.26</sub>Al<sub>0.74</sub>)<sub>33</sub>-type]. Moreover [1993Ste2] did not find in the isothermal section the YbCuAl<sub>3</sub> phase (Al<sub>4</sub>Ba-type), previously reported by [1988Kuz], and found that the previously reported YbCu<sub>4</sub>Al phase (CaCu<sub>5</sub>-type) is a solid solution of Al in the binary compound YbCu<sub>5</sub>, which dissolves up to about 35 at.% Al. Moreover, YbAl<sub>2</sub> dissolves up to about 17 at.% Cu and YbAl<sub>3</sub> less than 5 at.% Cu.

**3.3.16 Lu-Cu-Al.** The isothermal section at 600 °C was studied by [1992Kuz1] and is shown in Fig. 4(d). Note that the Al-Cu edge is not in agreement with the accepted binary Al-Cu system and that the Lu<sub>2</sub>Cu<sub>9</sub> compound, assumed to exist by analogy with the other heavy rare earths, is missing. Six ternary phases have been found, except for  $\tau_{14}$ -Lu<sub>6</sub>Cu<sub>16</sub>Al<sub>7</sub> (Th<sub>6</sub>Mn<sub>23</sub>-type) and  $\tau_{18}$ -LuCu<sub>0.9</sub>Al<sub>2.1</sub>, which are compounds of fixed stoichiometry, the other ternary phases are solid solutions:  $\tau_2$ -LuCu<sub>6-3.8</sub>Al<sub>6-8.2</sub> (ThMn<sub>12</sub>-type),  $\tau_5$ -Lu<sub>2</sub>Cu<sub>10.2-9.1</sub>Al<sub>6.8-7.9</sub> (Th<sub>2</sub>Zn<sub>17</sub>-type),  $\tau_{11}$ -LuCu<sub>3.8-2.6</sub>Al<sub>1.2-2.4</sub> (CaCu<sub>5</sub>-type), and  $\tau_{20}$ -LuCu<sub>1.1-0.9</sub>Al<sub>0.9-1.1</sub> (ZrNiAl-type).

#### 4. Comments About the Characteristics of the Isothermal Sections

The various R-Cu-Al sections show some changes on passing through the R series, and generally the R-rich alloys have not been investigated. A few general remarks however are possible, concerning the ternary phases and their equilibria with the component metals and/or the binary phases. Special attention may be given to the three component-rich corners. Considering the trivalent R, we notice that for the R-rich regions, data are generally very scarce. In the Cu-rich corner usually two-phase regions are formed: Cu solid solution in equilibrium with the RCu<sub>5</sub> or RCu<sub>6</sub> based phases. In the Al-rich corner, the (Al) phase is in equilibrium, according to the Cu/R ratio, with the  $\theta$ -Al<sub>2</sub>Cu phase, and with R<sub>3</sub>Al<sub>11</sub> (light R) or with RAl<sub>3</sub> (heavy R, Sc, and Y), and one ternary compound. (Al) is in equilibrium with the phase  $\tau_2$ -R(Cu,Al)<sub>12</sub> in all the systems. Moreover, it is in equilibrium also with the phase  $\tau_{13}$ -R(Cu,Al)<sub>4</sub> in the systems with R = La and Ce, or  $\tau_{15}$ -R<sub>3</sub>(Cu,Al)<sub>11</sub> with R = Y, Gd, and Tb. Progressive changes of the Cu/R ratio, with the trivalent R, result in a gradual variation of the structure of the solid alloy. This gives the possibility of fine tuning the characteristics of such alloys. The systems with Eu and Yb are

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slightly different, mainly in the R-rich corners where we have a ternary phase in equilibrium with the R itself.

### 5. Miscellaneous

In the following, a few notes are reported for the different phases in the R-Cu-Al systems concerning different properties such as magnetic and electrical. [1979Fel] studied the magnetism and hyperfine interactions of  $^{151}\text{Eu}$ ,  $^{155}\text{Gd}$ ,  $^{161}\text{Dy}$ ,  $^{166}\text{Er}$ , and  $^{170}\text{Yb}$  in  $\text{RCu}_4\text{Al}_8$ , and [1995Cac2] reported neutron spectroscopy studies of crystal-field interaction in  $\text{RT}_4\text{Al}_8$  compounds (R = Tb, Ho, and Er; T = Mn, Fe, and Cu). [1996Shc] carried out  $L_{\text{III}}$  x-ray absorption (77 and 300 K) and magnetic susceptibility measurements on  $\text{RCu}_4\text{Al}_8$  compounds finding peculiarities of the valence state of Ce and Yb in  $\text{RCu}_4\text{Al}_8$ , while Suski et al. [1997Sus] studied the magnetic and electrical properties of  $\text{ScCu}_{4+x}\text{Al}_{8-x}$  ( $0 \leq x \leq 2.15$ ) system. Moreover, Kang and Chen [2003Kan] calculated the preferential occupation of Cu in  $\text{ScCu}_x\text{Al}_{12-x}$  ( $4 \leq x \leq 6.15$ ), by interatomic potential using the lattice inversion method; the properties related to lattice vibration, such as the phonon density of states, specific heat, and vibrational entropy; and some mechanical properties such as the elastic constants and the bulk moduli of  $\text{ScCu}_x\text{Al}_{12-x}$ . Finally, [2000Kon] studied magnetic properties of  $\text{CeCu}_{4+x}\text{Al}_{8-x}$  ( $0 \leq x \leq 0.55$ ) by specific heat and electrical resistivity,  $^{27}\text{Al}$ -NMR and  $^{63}\text{Cu}$ -NQR measurements.

[1982Fel] investigated the magnetic properties of  $\text{RCu}_6\text{Al}_6$  (R = light rare earth) by magnetization and Mössbauer spectroscopy measurements. [2001Duo] studied the magnetic properties of  $\text{GdCu}_4\text{Al}_8$  and  $\text{GdCu}_6\text{Al}_6$  using standard magnetization and susceptibility measurements, magnetization measurements in high fields of up to 35 T, and measurements of specific heat. They also analyzed the data in terms of a simple mean-field two-sublattice model and found that the coupling between Gd moments is fairly weak and leads to antiferromagnetic ordering at rather low temperatures.

Spin fluctuations in the  $\text{Ce}_2\text{Cu}_8\text{Al}_9$  phase were studied by neutron scattering by [1999Ooh].

Magnetic and electrical properties of the  $\text{GdCu}_{5-x}\text{Al}_x$  ( $x = 0$  to 2) alloys were described by [1998Tun]; all the alloys were of the  $\text{CaCu}_5$ -type. To avoid the cubic  $\text{AuBe}_5$ -type, splat cooling was applied to  $\text{GdCu}_{5-x}\text{Al}_x$  alloys with  $x = 0$  and 0.1; for  $0.5 \leq x \leq 2.0$  it was found that the  $\text{AuBe}_5$ -type structure does not exist and these compounds melt congruently to form the hexagonal  $\text{CaCu}_5$ -type structure. Low-temperature resistivity and magnetic properties of the  $\text{YbCu}_{5-x}\text{Al}_x$  were investigated by several authors, evidencing Kondo behavior [1992Bau, 1998He, 1999Bon, 2001And, 2001He].

[1994Mul] investigated the magnetic properties and the  $^{155}\text{Gd}$  Mössbauer spectra of  $\text{GdCuAl}_3$ , which was found to order antiferromagnetically at low temperatures, while [1996Aoy] carried out an NMR study of the heavy fermion material  $\text{CeCuAl}_3$ , and [1996Kon] studied high pressure effects on heavy fermion antiferromagnet  $\text{CeCuAl}_3$  by NMR up to 15 kbar. Subsequently, Kontani et al. [1999Kon]

investigated magnetic properties of  $\text{CeCu}_x\text{Al}_{4-x}$  ( $x = 0.8, 0.9, 1, 1.1$ ) by magnetic susceptibility, high-field magnetization, specific heat, and electrical resistivity measurements using single-crystal samples.

Recent papers about magnetic properties of the  $\text{RCuAl}$  phases were reported in the following. Specific heat versus temperature was measured by [2000Jav] for a  $\text{CeCuAl}$  sample containing ~10% spurious phases; subsequently the electrical resistivity and magnetization measurements, carried out by [2001Che], revealed the occurrence of antiferromagnetic ordering in  $\text{CeCuAl}$  below  $T_N = 5.2$  K; moreover, the electronic structure was investigated by core-level photoemission spectroscopy by [1993Sin]. [1998Jav1] carried out a specific heat and magnetization study of  $\text{PrCuAl}$  and  $\text{NdCuAl}$  and found that, while  $\text{PrCuAl}$  orders antiferromagnetically below  $T_N = 7.9$  K, the magnetic behavior of  $\text{NdCuAl}$  is more complex. The ordering temperature is 18 K, and the magnetic ordering is characterized by a coexistence of ferro- and antiferromagnetic components of Nd moments. [1998Jav2] studied the magnetic properties of the  $\text{RCuAl}$  (R = Y, Ce to Sm, Gd to Tm, and Lu) intermetallic compounds by means of susceptibility, magnetization, and specific heat measurements and observed a magnetic ordering at low temperatures in most of these materials.  $\text{PrCuAl}$  and  $\text{NdCuAl}$  showed an antiferromagnetic behavior while in the heavy rare-earth compounds (R = Gd to Er) ferromagnetic coupling was found. The magnetization, electrical resistivity and alternating current (ac) susceptibility measurements, carried out by [2000Jar], provide evidence for a ferromagnetic type of order-disorder transition at 83 K in  $\text{GdCuAl}$ ; a second transition at 23 K was also found.

The magnetic properties of  $\text{TbCuAl}$  and neutron diffraction were discussed by [1996Ehl]. [1996Jav] carried out a magnetic structure study of  $\text{ErCuAl}$  and found that it orders ferromagnetically with magnetic moments along the c axis below  $T_c = 6.8$  K. A neutron powder diffraction study on  $\text{TmCuAl}$  was presented by [2002Jav]. The specific heat, thermal expansion, and electric resistivity of the  $(\text{Yb,Lu})\text{CuAl}$  compounds were measured in the temperature range 1.5-400 K by [1981Pot]. Finally, the high-pressure crystal structure of the  $\text{RCuAl}$  compounds was reported by [1987Tsv].

Atomic and electronic structure and the electronic transport mechanism in the amorphous  $\text{Al}_x(\text{Cu}_{0.4}\text{Y}_{0.6})_{100-x}$  ( $0 \leq x \leq 85$ ) were studied by [1996Fuk]. Glass-forming ability was discussed by different authors: [2000Fan] studied the glass-forming ability of R-Al-Cu (R = Sm, Y) upon melt-spinning and die-casting into a copper mold, while [2001Tan] investigated the La-rich La-Cu-Al alloys and obtained the best glass formation at 66La, 20Cu, 14Al at.%, described as a eutectic alloy.

[2001Li] studied the effect of Ce addition on the hydrogen content in a Al-32.2 Cu (mass%) eutectic alloy melt, [1996Mit] studied the interaction of  $\text{H}_2$  with  $\text{RCuAl}$  (R = Dy, Ho, Er), and [2001Kad] investigated whether Al substitution could expand the metal-atom lattice of  $\text{YCu}_5$  enough to provide a better interstitial position for hydrogen storage, but no significant hydrogen absorption/desorption characteristics were observed.

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