Copper – Lanthanum – Nickel

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Introduction

A summary of experimental studies of phase equilibria is given in Table 1. A large number of studies in this system are related to hydrogen absorption/desorption behavior of stoichiometric $LaNi_{5-x}Cu_x$ with $0 \leq x \leq 5$ and non-stoichiometric $LaNi_{6-x}Cu_x$ with $0 \leq x \leq 6$ phases [1974Mal, 1978Shi, 1980Bel, 1982Lia, 1983Sem, 1984End, 1985Kon, 1985Per, 1996Luo, 1997Lat, 2000Nak, 2002Jou], as they are promising candidates for hydrogen storage materials with applications in the field of rechargeable nickel/metal hydride batteries. In addition, the absorption and desorption behavior of deuterium in stoichiometric $LaNi_4Cu$ and non-stoichiometric $LaNi_5Cu$ have also been studied [1997Lat]. These studies are driven primarily by the fact that long-term electrochemical cycling stability of $LaNi_5$ in alkaline media can be substantially improved by partial replacement of La and/or Ni by another transition metal. In particular, $LaNi_{6-x}Cu_x$ compounds are very attractive due to their good long-term stability with an excellent charge/discharge capability [1992Coe].

Only recently, an isothermal section at 400°C has been determined [2005Liu]. They prepared 152 alloys using 99.9% Cu, 99.8% La and 99.99% Ni. The ternary alloys were prepared either by arc melting or by induction melting in an argon atmosphere. Subsequently, the alloys were sealed in evacuated quartz tubes and homogenized in the temperature range of 420 to 600°C for up to 400 h followed by cooling to 400°C at $9°C \cdot h^{-1}$, and kept at this temperature for 400 h followed by quenching into liquid nitrogen. The phase equilibria were established by means of X-ray diffraction, electron probe microanalysis and differential thermal analysis.

Binary Systems

The Cu-La binary phase diagram is accepted from [1993Cha1, 2006Gry]. The Cu-Ni binary system is accepted from [1993Cha2, 2002Leb]. The La-Ni binary system is accepted from [1991Oka].

Solid Phases

Both $LaCu_5$ and $LaNi_5$ are isotypic, and they form a continuous solid solution $La(Ni,Cu)_5$. The crystal chemistry of hydride forming multicomponent compounds $La(Ni,Cu)_5$ and $La(Ni,Cu)_6$ has received significant attention due to their applications in nickel metal hydride (NiMH) batteries. Random substitution of only Ni-sites by Cu gives rise to stoichiometric compound $LaNi_{5-x}Cu_x$ with $0 \leq x \leq 5$, while random substitution of La by dumbbell pairs of (Ni/Cu) atoms gives rise to non-stoichiometric compound $LaNi_{6-x}Cu_x$ with $0 \leq x \leq 6$ [1992Coe, 1999Lat]. The lattice parameters of stoichiometric $LaNi_{5-x}Cu_x$ have been measured several times [1978Dwi, 1982Lia, 1982Pas1, 1982Pas2, 1984End, 1984Pas, 1992Mal, 1996Luo, 1999Lat], and in general they show a good agreement. Along the stoichiometric composition of $LaNi_{5-x}Cu_x$, both *a*- and *c*-parameters increase linearly as Ni is substituted by Cu giving a constant value of *c*/*a* ratio [1992Mal]. However, earlier experimental data of [1978Dwi, 1982Lia] show a negative deviation of both *a*- and *c*-parameters from ideal behavior. The lattice parameters of non-stoichiometric $LaNi_{6-x}Cu_x$ have also been measured [1992Coe, 1996Luo, 1997Sri, 1999Lat]. In $LaNi_{6-x}Cu_x$, $0 \leq x \leq 3$, the *a*-parameter shows a negative deviation while the *c*-parameter shows a positive deviation from ideal behavior [1992Coe], and these have been attributed to the orientation of dumbbell pairs of (Ni/Cu) atoms along the *c*-axis.

At 400°C, LaCu and $LaCu_2$ dissolve about 4 and 15 at.% Ni, respectively [2005Liu]. The solubility of Cu in La_2Ni_7 and $LaNi_3$ is about 2 at.%, in La_7Ni_{16} and La_2Ni_3 is about 3 at.%, and in LaNi is about 5 at.% [2005Liu]. Recently, a ternary phase $Cu_{85}La_{10}Ni_5$ with cubic symmetry has been reported [2005Liu].

The crystallographic details of all solid phases are listed in Table 2.

Isothermal Sections

The isothermal section at 400°C is shown in Fig. 1 [2005Liu]. However, they failed to detect phase boundaries involving La_3Ni, hence, they are shown dashed.

Temperature – Composition Sections

The vertical section $LaCu_5$-$LaNi_5$ was determined by [1982Lia], it is shown in Fig. 2. It is almost a quasibinary section except for the incongruent melting of $LaCu_5$. Dash solidus and liquidus lines at 70-83.33 at.% Cu was drawn by us because [1982Lia] did not take into account that in this part must be present a three-phase region La_5(Ni,Cu)+$LaCu_6$+L and a two-phase region $LaCu_6$+L since $LaCu_5$ forms by a peritectic reaction L+$LaCu_6$⇌$LaCu_5$.

Thermodynamics

The heat of mixing of liquid alloys over a wide composition range was measured at 850°C in a solution calorimeter using the isoperibolic procedure [1999Zho]. In addition, [1999Zho] also measured the specific heat of liquid $Cu_{25}Ni_{50}La_{25}$ alloy in the temperature range of 817 to 937°C. They used elements of 99.99% purity to prepare the alloys. [1999Zho] used a generalized associate solution model [2000Kru] to analyze the experimental data. They assumed the presence of a ternary associate, $Cu_1La_1Ni_1$, in addition to the binary associates Cu_2La_1 and Ni_2La_1 to calculate the composition and temperature dependence of heat of mixing and specific heat of liquid alloys. Their results are shown in Figs. 3 and 4. The specific heat data suggest a temperature dependent heat of mixing of liquid alloys.
The standard heat of formation of $LaNi_{5-x}Cu_x$ intermetallic with $CaCu_5$ structure was determined by an isoperibolic calorimetric method [1982Pas1, 1982Pas2, 1984Pas], and their results are listed in Table 3. These results were also reviewed by [1985Per]. [1996Gon] extended Miedema's model to calculate the enthalpy of formation of $LaNi_4Cu$, and obtained a value of -126 kJ·mol^{-1} which agrees reasonably well with the experimental value of -143.16 kJ·mol^{-1} [1982Pas1, 1982Pas2].
The Gibbs energy of formation of $La(Ni_{1-x}Cu_x)_5$ hydrides was reported by [1978Shi]. With the knowledge of heat of formation of $LaNi_{5-x}Cu_x$, [1982Pas1, 1982Pas2, 1984Pas] calculated their enthalpies of hydrogenation. Also, the enthalpies of hydrogen absorption of $LaNi_{x-1}Cu$ intermetallics with $CaCu_5$ structure were measured using calorimetry [1996Luo]. In addition, they derived the entropy changes associated with hydrogen absorption at 25°C. [1999Shu] extended Miedema's model to calculate the enthalpy of hydrogenation of $LaNi_3Cu_2$ and $LaNi_4Cu$, and obtained a reasonably good agreement with the experimental data.

Notes on Materials Properties and Applications

The Cu-La-Ni alloys have been known as promising candidates for hydrogen storage materials with applications in the field of rechargeable nickel/metal hydride batteries. A series of studies in this system are related to hydrogen absorption/desorption behavior of stoichiometric $LaNi_{5-x}Cu_x$ with $0 \le x \le 5$ and non-stoichiometric $LaNi_{6-x}Cu_x$ with $0 \le x \le 6$ phases [1974Mal, 1978Shi, 1980Bel, 1982Lia, 1983Sem, 1984End, 1985Kon, 1985Per, 1996Luo, 1997Lat, 2000Nak, 2002Jou]. A summary of experimental investigation of properties is given in Table 4. At 40°C, the hydrogen absorption capacity of $La(Ni_{5-x}Cu_x)_5$ is less than that of $LaNi_5$ [1974Mal, 1978Shi].
The effect of hydrogen cycling on the lattice distortion [2000Nak] and defect generation [2002Jou] has been discussed in detail.
[1985Kon] reported specific magnetic susceptibility of $LaNi_{5-x}Cu_x$ intermetallics with $CaCu_5$ structure.

Miscellaneous

[1984Zhe] has discussed the direct synthesis method of $LaNi_{5-x}Cu_x$ intermetallic with $CaCu_5$ structure, starting with the powders of Cu, Ni and La_2O_3 by a reduction-diffusion process at 825°C.

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Table 1: Investigations of the Cu-La-Ni Phase Relations, Structures and Thermodynamics

Reference	Method/Experimental Technique	Temperature/Composition/Phase Range Studied
[1978Dwi]	X-ray diffraction	700°C; $LaNi_{5-x}Cu_x$, $0 \le x \le 5$
[1978Shi]	Thermal analysis, optical metallography	<1050°C; 2-22.5 mass% Ni and 5-40 mass% Cu
[1982Lia]	Thermal analysis, XRD	800-1350°C; $LaNi_{5-x}Cu_x$, $0 \le x \le 5$
[1982Pas1, [1982Pas2]	Calorimetry, XRD	800°C, $LaNi_4Cu$
[1984End]	XRD	$LaNi_{5-x}Cu_x$, $0 \le x \le 5$
[1984Pas]	Calorimetry, XRD	800°C; $LaNi_5$-$LaCu_5$
[1984Zhe]	XRD	825°C; $LaNi_{5-x}Cu_x$, $0 \le x \le 5$
[1992Mal]	XRD	750-950°C, $LaNi_5$-$LaCu_5$
[1992Coe]	TEM, XRD	$LaNi_{6-x}Cu_x$, $0 \le x \le 3$
[1996Luo]	XRD	1050-1200°C; $LaNi_4Cu$ and Non-stoichiometric $La(Ni,Co)_z$, $5 \le z \le 6$
[1997Lat]	XRD	$LaNi_4Cu$ and $LaNi_5Cu$
[1997Sri]	XRD	Non-stoichiometric $La(Ni,Cu)_z$, $5 \le z \le 6$
[1999Lat]	XRD and Neutron diffraction	$LaNi_4Cu$ and non-stoichiometric $La(Ni,Cu)_z$, $5 \le z \le 6$
[1999Zho]	Calorimetry	850°C; 23.2-74.2 at.% La, 4.9-61.1 at.% Ni; liquid phase
[2000Nak]	XRD	1100°C, $LaNi_{4.5}Cu_{0.5}$
[2002Jou]	XRD	$LaNi_4Cu$
[2005Liu]	DTA, EPMA, XRD	400°C

Table 2: Crystallographic Data of Solid Phases

Phase/ Temperature Range [°C]	Pearson Symbol/ Space Group/ Prototype	Lattice Parameters [pm]	Comments/References
γ, (Ni,Cu)	*cF*4 $Fm\bar{3}m$		
(Ni) < 1455	Cu	a = 352.32	pure Ni at 25°C [Mas2]
(Cu) < 1084.62		a = 361.46	pure Cu at 25°C [Mas2]
(γLa) 918 - 865	*cI*2 $Im\bar{3}m$ W	a = 426	pure La [Mas2]

Phase/ Temperature Range [°C]	Pearson Symbol/ Space Group/ Prototype	Lattice Parameters [pm]	Comments/References
(βLa) 865-310	*cF*4 $Fm\bar{3}m$ Cu	*a* = 530.3	pure La [Mas2]
(αLa) < 310	*hP*4 $P6_3/mmc$ αLa	*a* = 377 *c* = 1217.1	pure La at 25°C [Mas2]
βCu_6La 905-218	*oP*28 *Pnma* β$CeCu_6$	*a* = 816.5 *b* = 514.8 *c* = 1023	[1993Cha1]
αCu_6La < 218	*mP*28 $P2_1/C$ αCu_6La	*a* = 514.67 *b* = 1021.03 *c* = 814.55 β = 91.51°	at 27°C [2006Gry]
Cu_2La < 830	*hP*3 *P6/mmm* AlB_2	*a* = 434.5 *c* = 381.9	[1993Cha1]
CuLa < 525	*oP*8 *Pnma* FeB	*a* = 754.3 *b* = 461.6 *c* = 572.4	[1993Cha1]
La_3Ni < 532	*oP*16 *Pnma* Fe_3C	*a* = 722 *b* = 1024 *c* = 660	[V-C2]
La_7Ni_3 < 530	*hP*20 $P6_3/mc$ Fe_3Th_7	*a* = 1014 *c* = 638.3	[V-C2]
LaNi < 715	*oC*8 *Cmcm* CrB	*a* = 390.7 *b* = 1081 *c* = 439.6	[V-C2]
La_2Ni_3 < 688	*oC*20 *Cmca* La_2Ni_3	*a* = 511.38 *b* = 973.15 *c* = 790.75	[V-C2]
La_7Ni_{16} < 714	*tI*46 $I\bar{4}2m$ La_7Ni_{16}	*a* = 735.5 *c* = 1451.1	[V-C2]
βLa_2Ni_7 1014 - 977	*hR*54 $R\bar{3}m$ Er_2Co_7	*a* = 505.6 *c* = 3698.1	[V-C2]
αLa_2Ni_7 < 977	*hP*36 $P6_3/mmc$ Ce_2Ni_7	*a* = 505.8 *c* = 2471	[V-C2]

Phase/ Temperature Range [°C]	Pearson Symbol/ Space Group/ Prototype	Lattice Parameters [pm]	Comments/References
$La(Ni,Cu)_5$	*hP*6 *P*6/*mmm* $CaCu_6$	*a* = 513.0 *c* = 408.7	$LaNiCu_4$, at 25°C [1992Mal]
		a = 509.6 *c* = 405.0	$LaNi_2Cu_3$, at 25°C [1992Mal]
		a = 506.6 *c* = 402.5	$LaNi_3Cu_2$, at 25°C [1992Mal]
		a = 503.3 *c* = 400.7	$LaNi_{3.98}Cu_{0.96}$, at 25°C [1982Pas1]
		a = 503.94 *c* = 400.52	$LaNi_4Cu$, at 25°C [1996Luo]
		a = 503.79 *c* = 400.64	$LaNi_{4.02}Cu_{0.99}$, at 25°C [1999Lat]
		a = 502.8 *c* = 399.5	$LaNi_{4.75}Cu_{0.25}$, at 25°C [1984End]
		a = 502.72 *c* = 399.46	$LaNi_{4.5}Cu_{0.5}$, at 25°C [2000Nak]
		a = 502.52 *c* = 401.27	$LaNi_{4.2}Cu$, at 25°C [1996Luo]
		a = 501.12 *c* = 402.04	$LaNi_{4.36}Cu_{0.99}$, at 25°C [1999Lat]
		a = 501.02 *c* = 402.04	$LaNi_{4.4}Cu$, at 25°C [1996Luo]
		a = 498.64 *c* = 404.81	$LaNi_{4.5}Cu_{1.49}$, at 25°C [1999Lat]
		a = 499.84 *c* = 402.84	$LaNi_{4.6}Cu$, at 25°C [1996Luo]
		a = 498.68 *c* = 403.47	$LaNi_{4.8}Cu$, at 25°C [1996Luo]
		a = 498.61 *c* = 403.24	$LaNi_{4.84}Cu_{0.95}$, at 25°C [1999Lat]
		a = 498.75 *c* = 403.88	$LaNi_5Cu$, at 25°C [1996Luo]
Cu_5La < 805		*a* = 518.7 *c* = 410.9	[1993Cha1]
$LaNi_5$ < 1350		*a* = 501.8 *c* = 397.8	[V-C2]

Phase/ Temperature Range [°C]	Pearson Symbol/ Space Group/ Prototype	Lattice Parameters [pm]	Comments/References
* τ_1, $Cu_{85}La_{10}Ni_5$	*cF** *Fm*3*c*	*a* = 1158.0	[2005Liu]

Table 3: Standard Heat of Formation of $La(Ni,Cu)_5$ with $CaCu_5$ Structure

Phase	Temperature [°C]	Heat of formation [kJ·mol^{-1}]
$LaCu_5$	298.15	–75.77
$LaNiCu_4$	298.15	–93.35
$LaNi_2Cu_3$	298.15	–107.58
$LaNi_3Cu_2$	298.15	–128.51
$LaNi_4Cu$	298.15	–143.16
$LaNi_5$	298.15	–161.16

Table 4: Investigations of the Cu-La-Ni Materials Properties

Reference	Method/Experimental Technique	Type of Property
[1974Mal]	Absorption/desorption	Hydrogen absorption in $LaNi_4Cu$
[1978Shi]	Absorption/desorption	Hydrogen absorption in $LaNi_{5-x}Cu_x$
[1980Bel]	Absorption/desorption	Hydrogen absorption in $LaNi_{4.9}Cu_{0.1}$
[1982Lia]	Absorption/desorption	Hydrogen absorption in $LaNi_{5-x}Cu_x$
[1983Sem]	Absorption/desorption	Hydrogen absorption in $LaNi_{5-x}Cu_x$
[1984End]	Absorption/desorption	Hydrogen absorption in $LaNi_{5-x}Cu_x$
[1985Kon]	Absorption/desorption	Catalytic and magnetic properties, and hydrogen absorption of $LaNi_{5-x}Cu_x$
[1996Luo]	Absorption/desorption	Hydrogen absorption in $LaNi_{5-x}Cu_x$
[1997Lat]	Absorption/desorption	Deuterium absorption in $LaNi_4Cu$, and $LaNi_5Cu$
[2000Nak]	Absorption/desorption	Hydrogen absorption in $LaNi_{4.5}Cu_{0.5}$
[2002Jou]	Absorption/desorption	Hydrogen absorption in $LaNi_4Cu$

Fig. 1: Cu-La-Ni.
Isothermal section at 400°C

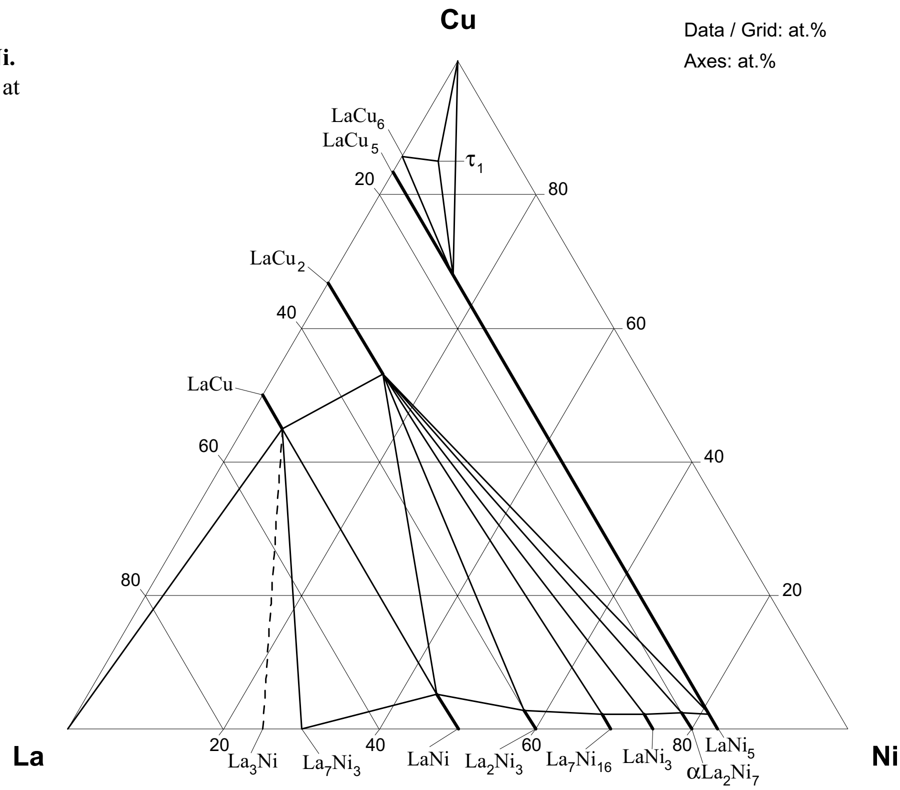

Fig. 2: Cu-La-Ni.
Vertical section $LaNi_5$ - $LaCu_5$

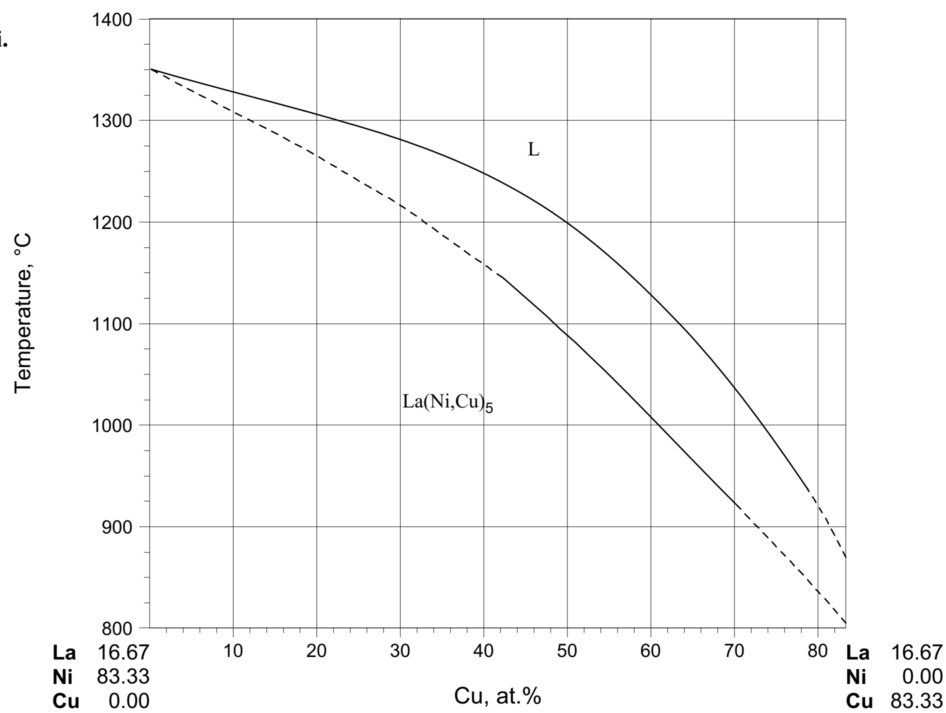

Fig. 3: Cu-La-Ni. Specific heat of liquid $Cu_{25}La_{50}Ni_{25}$ alloy. Points are experimental data, dashed line represents mechanical mixture of liquid and undercooled liquid components; solid line is calculated using an associated solution model.

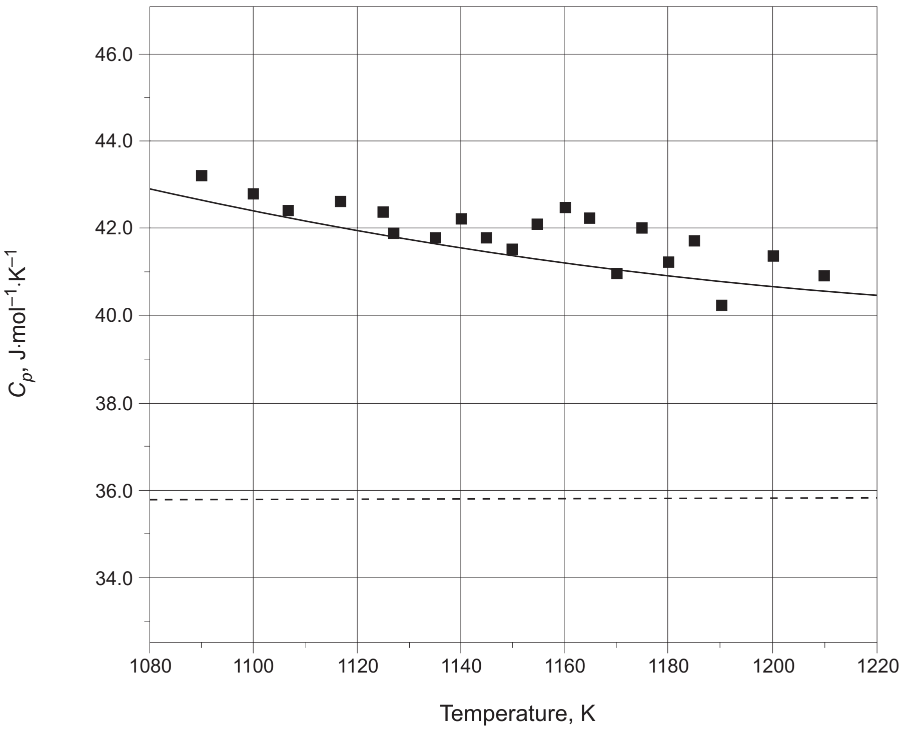

Fig. 4: Cu-La-Ni. Isocontours of enthalpy of mixing (in kJ·mol^{-1}) of liquid alloys at 850°C calculated using an associate solution model