Copper – Nickel – Zirconium

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Introduction

A summary of experimental studies of phase equilibria is given in Table 1.
[1968Fed] investigated the phase equilibria of Cu corner with an emphasis on the solubility of Zr in (Ni,Cu). The solubility limits were determined at 600, 700, 800 and 900°C by metallographic technique and microhardness measurements. They also determined two isopleths at 0.2 and 0.6 mass% Ni within 0 to 0.49 mass% Zr.
Based on the results of thermal analysis of a number of alloys [1968Tak] constructed the liquidus surface of Zr rich alloys in the composition range of 20-40 mass% Cu and 15-30 mass% Ni. [1968Vya] investigated the phase equilibria of Zr corner involving $ZrCu_2$ and $ZrNi_2$ phases, and reported a partial liquidus surface, four partial isothermal sections at 800, 850, 950 and 1300°C, and two isopleths at constant ratios of Cu:Ni = 4:1 and 1:1. The phase equilibria were determined by dilatometry, thermal analysis, optical microscopy and hardness measurements.
The solid solubilities of binary intermetallics in the ternary regime have been investigated along the following composition sections: Zr_2Cu-Zr_2Ni [1964Ram, 1972Hav1], $Zr_{10}Cu_7$-$Zr_{10}Ni_7$ [1981Gli], and Zr_5Cu-Zr_5Ni [1986For]. Besides, investigations of the compounds [1964Ram, 1972Hav1, 1972Hav2, 1981Gli, 1986For, 2000Kov], formation of Zr rich metallic glasses by rapid solidification and their crystallization kinetics and mechanism [1992Gro, 2002Wan, 2003Liu, 2003Wan1, 2003Wan2, 2003Wan3, 2003Wan4, 2004Wan] were also reported.
Phase relations in the Cu-Ni-Zr have been reviewed by [1979Dri, 2000Gup].

Binary Systems

The Cu-Ni binary system is accepted from [2002Leb]. The Cu-Zr binary phase diagram is accepted from [2006Sem]. The Ni-Zr binary phase diagram is accepted from [1984Nas] in which the assessed temperature 1170°C for invariant reaction L ⇌ (Ni) + $ZrNi_5$ is replaced by 1196°C obtained in recent experiments [2001Miu].

Solid Phases

The unary, binary and ternary phases of the Cu-Ni-Zr system are presented in Table 2.
The solubilities of Zr in (Ni,Cu) as a function of temperature [1968Fed] are shown in Fig. 1.
Both [1964Ram] and [1972Hav1] reported an incomplete solubility between Zr_2Cu and Zr_2Ni. However, [1972Hav1] reported that as-cast $Zr_2Ni_{1-x}Cu_x$ alloys with $x \le 0.4$ have $C16$ ($CuAl_2$ type) structure, alloys with $x \ge 0.6$ have $C11_b$ ($MoSi_2$ type) structure, and a two-phase mixture in between. On the other hand, after annealing the as-cast alloys at 700°C for 19 h, [1964Ram] found that the solubility of Cu in Zr_2Ni is at least about 8 at.% and that of Ni in Zr_2Cu is negligible.
According to X-ray diffraction investigation of as-cast alloys $Cu_{10}Zr_7$ and $Ni_{10}Zr_7$ form a continuous solid solution [1981Gli]. However, it is not known if it would be the same after annealing. Even though Zr_7Cu_{10} and Zr_7Ni_{10} are isostructural, [2000Gup] pointed out that a deviation from an isomorphous-type pseudobinary (complete solid solubility) is expected along Zr_7Cu_{10}-Zr_7Ni_{10} section, as Zr_7Cu_{10} melts congruently while Zr_7Ni_{10} forms by a peritectic reaction. In as-cast alloys, both *a* and *b* lattice constants show a negative deviation while the *c* lattice constant shows a positive deviation from the ideal behavior.
[1986For] studied $Zr(Ni_{1-x}Cu_x)_5$ alloys after annealing them at 840°C. The lattice parameters of $Zr(Ni_{1-x}Cu_x)_5$ alloys are listed in Table 2. According to [1986For], in the composition limit of $0 \le x \le 0.3$ the alloys have cubic $AuBe_5$-type structure, while those with $0.35 \le x \le 1$ have Zr_2Cu_9 type tetragonal structure which is a long-period superstructure derived from $AuBe_5$ type of structure.

[2000Kov] reported about the ternary phase Zr_2NiCu of Heusler type that undergoes martensitic transformation to two monoclinic structures.

Invariant Equilibria

Two studies, [1968Tak] and [1968Vya], reported a ternary eutectic reaction L ⇌ (βZr)+$ZrCu_2$(r)+Zr_2Ni in the Zr corner; however, the reported invariant temperatures and concentrations of liquid phase are different. The data of [1968Vya] are preferable because of the large number of experiments performed. A partial reaction scheme of the system is shown in Fig. 2 based on the results of [1968Vya].
Characteristics of two invariant reactions are listed in Table 3. They are taken from [1968Vya], except for the compositions of Zr_2Cu(r) and Zr_2Ni, which are taken after [1964Ram].

Liquidus, Solidus and Solvus Surfaces

Figure 3 shows the liquidus surface in the composition range of Zr_2Cu-Zr_2Ni-Zr after [1968Vya].

Isothermal Sections

Figures 4, 5, 6 and 7 show the partial isothermal section [1968Vya] at 1300°C, 950°C, 850°C and 800°C, respectively. [1968Vya] constructed these partial isothermal sections using the results of isoplethal sections, thus, the location of phase boundaries at 950°C should be considered as only approximate. At other temperatures, position of the isoplethal sections enables to draw boundaries between the phase fields with reasonable accuracy. [1968Vya] did not report the solubility of Cu in Zr_2Ni at these temperatures, which is expected to be significant based on the results of other studies [1964Ram, 1972Hav1]. In accordance with this, the isothermal sections [1968Vya] are corrected to take into account solubility of Cu in Zr_2Ni [1964Ram].

Temperature – Composition Sections

Figures 8 and 9 show the isopleths [1968Fed] at constant Ni contents of 0.2 mass% and 0.6 mass%, respectively. In the original publication, [1968Fed] showed the presence of two-phase fields of (Cu)+$ZrCu_3$; however, according to the accepted binary Cu-Zr [2006Sem] the latter phase most likely to be $ZrCu_5$. Figures 10 and 11 show the isopleths [1968Vya] at constant ratios x_{Cu}:x_{Ni} of 4:1 and 1:1, respectively.

Thermodynamics

[1989Sid] measured the heat of mixing of five Ni rich liquid alloys at 1600°C using a high-temperature calorimeter. Later, [2000Wit] measured the heat of mixing of liquid alloys over a wide composition range at 1293 and 1295°C using a high-temperature isoperibolic calorimeter. They used 99.999% Cu, 99.98% Ni and 99.8% Zr to prepare the alloys. Figure 12 shows the isoenthalpic contours at 1565 ± 5 K [2000Wit]. Both [1999Zho] and [2000Wit] proposed that a large and negative heat of mixing may imply a tendency for chemical short range ordering in the liquid phase. [1999Zho] calculated the composition and temperature dependence of heat of mixing, using a generalized associate solution model [2000Kru] with the assumption of presence of two binary associates only, Zr_1Cu_2 and Zr_1Ni_2, in the liquid phase. They obtained a good agreement between the calculated and experimental values.

Notes on Materials Properties and Applications

The superconducting critical temperature, magnetic susceptibility and thermoelectric power of $Zr_2Ni_{1-x}Cu_x$ (*tI*12), $0 \le x \le 0.17$, were measured by [1972Hav2]. Both thermoelectric power and the critical temperature decreases monotonically as Ni is substituted by Cu [1972Hav2].
The intermetallic compound Zr_2NiCu is a promising candidate to develop high-temperature shape memory materials. [2000Kov] reported that the shape recovery of Zr_2NiCu is 90% at a strain level of 0.35%.

Miscellaneous

The tracer diffusivity of Cu in amorphous NiZr at 300°C was reported to be $2.6 \cdot 10^{-21}$ $m^2 \cdot s^{-1}$ [1986Hah]. Metallic glasses in the composition range of 50 to 70 at.% Zr and up to 25 at.% Cu have been obtained by rapid solidification [1992Gro, 2002Wan, 2003Liu, 2003Wan1, 2003Wan2, 2003Wan3, 2003Wan4, 2004Wan]. [2003Liu] found that the crystallization of $Cu_{20}Ni_{10}Zr_{70}$ amorphous alloy took place in two stages during continuous heating in a differential scanning calorimeter. Nucleation and growth of Zr_2Cu was followed by that of Zr_2Ni. The details of crystallization kinetics and associated kinetic parameters, and microstructure after crystallization have also been discussed [1992Gro, 2002Wan, 2003Liu, 2003Wan1, 2003Wan2, 2003Wan3, 2003Wan4, 2004Wan].

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

Reference	Method/Experimental Technique	Temperature/Composition/Phase Range Studied
[1964Ram]	XRD	700°C; $Zr_2(Ni,Cu)$
[1968Fed]	Thermal analysis, metallography, hardness	< 1100°C; up to 0.6 mass% Ni and 0.6 mass% Zr
[1968Tak]	Thermal analysis, soldering characteristics	< 1000; Cu: 10-30 mass%, Ni: 10-30 mass%, Zr: balance
[1968Vya]	Dilatometry, thermal analysis, XRD, optical microscopy, hardness	< 1560°C; Zr_2Cu-Zr_2Ni-Zr
[1972Hav1]	XRD	$Zr_8Ni_{4-x}Cu_x$, $0 \le x \le 1.5$

Reference	Method/Experimental Technique	Temperature/Composition/Phase Range Studied
[1972Hav2]	Superconductivity, thermo-electric power, magnetic susceptibility	$Zr_8Ni_{4-x}Cu_x$, 0 ≤ x ≤ 1.5
[1981Gli]	XRD	$Zr_7(Ni,Cu)_{10}$
[1986For]	XRD	840°C, $Zr(Ni_{1-x}Cu_x)_5$, 0 ≤ x ≤ 1
[1986Hah]	Secondary ion mass spectroscopy	300°C, ZrNi
[1989Sid]	High-temperature calorimetry	1600°C; $Zr_xNi_{1-x}Cu_x$, 0.015 ≤ x ≤ 0.075
[1992Gro]	Rapid solidification, DSC, XRD, TEM	≤ 550°C; $Zr_{50}Ni_{50-x}Cu_x$, 0 ≤ x ≤ 25
[2000Kov]	XRD, Resistivity, mechanical testing	250-850°C; Zr_2NiCu
[2000Wit]	Calorimetry	1293-1295°C; $Zr_{1-x-y}Ni_yCu_x$, 0.0102 ≤ x ≤ 0.9887, 0.0041 ≤ y ≤ 0.6335
[2002Wan]	Rapid solidification, DSC, TEM, XRD	≤ 600°C; $Ni_{10}Cu_{20}$
[2003Liu]	Rapid solidification, DSC, TEM, XRD	≤ 600°C; $Zr_{70}Ni_{10}Cu_{20}$
[2003Wan1]	Rapid solidification, DSC, XRD	≤ 600°C; $Zr_{70}Ni_xCu_{30-x}$, 5 ≤ x ≤ 15
[2003Wan2]	Rapid solidification, DSC, TEM, XRD	≤ 600°C; $Zr_{70}Ni_{10}Cu_{20}$
[2003Wan3]	Rapid solidification, DSC, TEM, XRD	≤ 600°C; $Zr_{70}Ni_{10}Cu_{20}$
[2003Wan4]	Rapid solidification, DSC, TEM, XRD	≤ 600°C; $Zr_{70}Ni_{10}Cu_{20}$
[2004Wan]	Rapid solidification, DSC, TEM, XRD	≤ 600°C; $Zr_{70}Ni_{10}Cu_{20}$

Table 2: Crystallographic Data of Solid Phases

Phase/ Temperature Range [°C]	Pearson Symbol/ Space Group / Prototype	Lattice Parameters [pm]	Comments/References
(Ni,Cu)	*cF*4 $Fm\bar{3}m$		
(Ni) ≤ 1455	Cu	a = 352.32	pure Ni at 25°C [V-C2]
(Cu) ≤ 1084.62		a = 361.46	pure Cu at 25°C [V-C2]
(βZr)(h) 1855 - 863	*cI*2 $Im\bar{3}m$ W	a = 356.8	dissolves up to 5.7 at.% Cu [2006Sem] and 2.92 at.% Ni [1984Nas], pure βZr [V-C2]
(αZr)(l) < 863	*hP*2 $P6_3/mmc$ Mg	a = 323.2 c = 514.7	dissolves up to ~0.2 at.% Cu [2006Sem] and 0.2 at.% Ni [1984Nas], pure αZr at 25°C [V-C2]

Phase/ Temperature Range [°C]	Pearson Symbol/ Space Group / Prototype	Lattice Parameters [pm]	Comments/References
$Zr(Ni_{1-x}Cu_x)_5$	*cF*24 $F\bar{4}3m$ $AuBe_5$	 *a* = 671.2 *a* = 673.6	0 ≤ *x* ≤ 0.30 [1986For] at *x* = 0.05 [1986For] at *x* = 0.3 [1986For]
$ZrNi_5$ < 1300		*a* = 670.64 to 670.72	at *x* = 0 [1984Nas]
$ZrCu_5$ < 1032		*a* = 687.0	at *x* = 1 [2006Sem]
$Zr(Ni_{1-x}Cu_x)_5$	*t*** Zr_2Cu_9 (long period superstructure derived from $AuBe_5$ type)	*a* = 674.5 *c* = 675.2	at *x* = 0.35 [1986For]
$Zr_{14}Cu_{51}$ ≤ 1112	*hP*68 *P*6/*m* $Gd_{14}Ag_{51}$	*a* = 1124.44 *c* = 828.15	[2006Sem]
Zr_3Cu_8 ≤ 1028	*oP*44 *Pnma* Hf_3Cu_8	*a* = 786.93 *b* = 851.47 *c* = 646.0	[2006Sem]
$Zr_{24}Cu_{13}$ 960 - 915	*o**37	*a* = 1119.0 *b* = 791.2 *c* = 998.48	[2006Sem]
$Zr_7(Ni_{1-x}Cu_x)_{10}$	*oC*68 *C*2*ca* Zr_7Ni_{10}	 *a* = 1246.8 *b* = 923.2 *c* = 931.5	0 ≤ *x* ≤ 1 in cast state [1981Gli] at *x* = 0.5 [1981Gli]
Zr_7Cu_{10} < 935		*a* = 1267.29 *b* = 931.63 *c* = 934/66	at *x* = 1, [2006Sem]
Zr_7Ni_{10} < 1160		*a* = 1238.6 to 1249.7 *b* = 915.6 to 921.0 *c* = 921.1 to 932.5	at *x* = 0 and 41.1 to 43.5 at.% Zr [1984Nas]
ZrCu(h) 960 - 725	*cP*2 $Pm\bar{3}m$ CsCl	*a* = 326.6	at 49.9 at.% and 25°C [2006Sem]

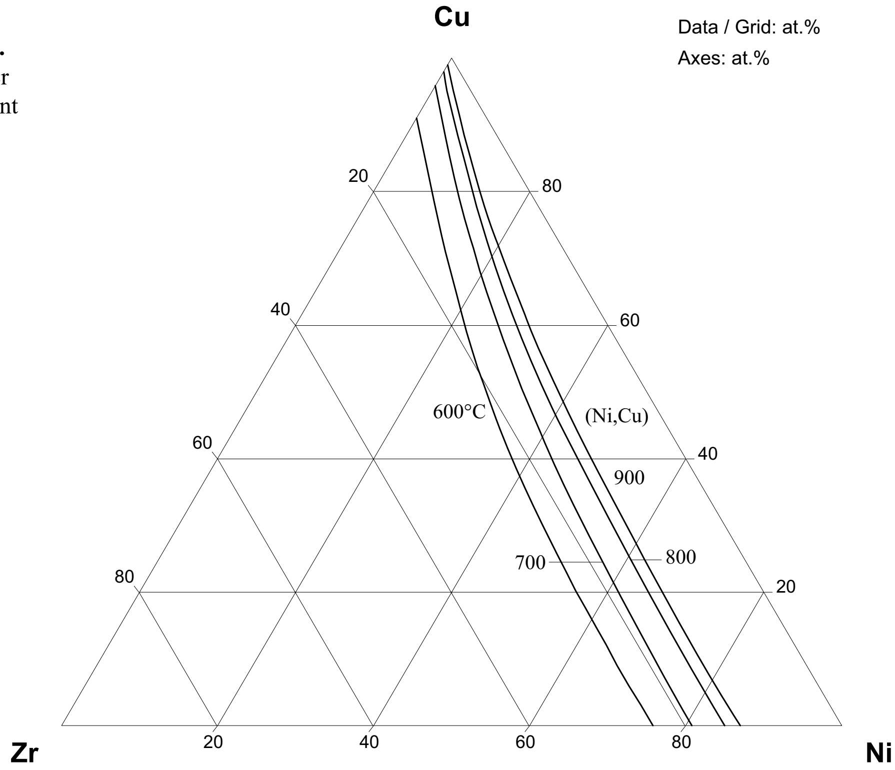
Phase/ Temperature Range [°C]	Pearson Symbol/ Space Group / Prototype	Lattice Parameters [pm]	Comments/References
ZrCu(l) < 140	*mP*4 $P2_1/m$ TiNi	*a* = 328.2 *b* = 414.8 *c* = 524.9 β = 103.7°	martensite, [2000Kov]
	*mC*16 *Cm*	*a* = 633.7 *b* = 856.3 *c* = 534.5 β = 105.6°	martensite, [2000Kov]
Zr_2Cu(h) 1025 - 950	*tI*6 *I*4/*mmm* $MoSi_2$	*a* = 322.04 *c* = 1183.2	[2006Sem]
Zr_2Cu(r) < 950	*tP*150	*a* = 1592.4 *c* = 1132.8	[2006Sem]
$ZrNi_5$ ≤ 1300	*cF*24 $F\bar{4}3m$ $AuBe_5$	*a* = 670.64 to 670.72	15 to 18 at.% Zr [1984Nas]
Zr_2Ni_7 ≤ 1440	*mC*36 *C*2/*m* Zr_2Ni_7	*a* = 469.8 *b* = 823.5 *c* = 1219.3 β = 95.83°	[1984Nas]
$ZrNi_3$ ≤ 920	*hP*8 $P6_3/mmc$ Ni_3Sn	*a* = 530.9 *c* = 430.3	24.5 to 26.0 at.% Zr [1984Nas]
Zr_8Ni_{21} < 1180	*aP*29 $P\bar{1}$ Hf_8Ni_{21}	*a* = 647.21 *b* = 806.45 *c* = 858.75 α = 75.18° β = 68.00° γ = 75.20°	[1984Nas]
Zr_9Ni_{11} 978 - 1170	*tI*40 *I*4/*m* Zr_9Pt_{11}	*a* = 988.0 *c* = 661.0	[1984Nas]
ZrNi ≤ 1260	*oC*8 *Cmcm* CrB	*a* = 326.8 *b* = 993.0 *c* = 410.21	[1984Nas]

Phase/ Temperature Range [°C]	Pearson Symbol/ Space Group / Prototype	Lattice Parameters [pm]	Comments/References
$Zr_2Ni_{1-x}Cu_x$	*tI*12 *I*4/*mcm* $CuAl_2$	 *a* = 655.0 *c* = 524.0	0 ≤ *x* ≤ 0.24 at 700°C [1964Ram] at *x* = 0.24 and 25°C [1964Ram]
Zr_2Ni ≤ 1120		*a* = 647.7 to 648.3 *c* = 524.1 to 526.7	at *x* = 0, [V-C2]
* Zr_2NiCu ?	*cF*16 $Fm\overline{3}m$ $MnCu_2Al$	*a* = 645.0	[2000Kov]
	*mP*4 $P2_1/m$ TiNi	*a* = 333.6 *b* = 419.3 *c* = 523.6 β = 103.1°	martensite, [2000Kov]
	*mC*16 *Cm*	*a* = 623.4 *b* = 897.3 *c* = 530.1 β = 103.8°	martensite, [2000Kov]

Table 3: Invariant Equilibria

Reaction	*T* [°C]	Type	Phase	Composition (at.%)		
				Cu	Ni	Zr
L ⇌ (βZr) + Zr_2Cu(r) + Zr_2Ni	~920	E_1	L	15	11	74
			(βZr)	2.5	1.7	95.8
			Zr_2Cu(r)	33	-	67
			Zr_2Ni	~8	~25	67
(βZr) ⇌ (αZr) + Zr_2Cu + Zr_2Ni	805	E_2	(βZr)	~2.5	~2.5	~95
			(αZr)	~0.1	~0.1	~99.8
			Zr_2Cu	33	-	67
			Zr_2Ni	~8	~25	67

Fig. 1: Cu-Ni-Zr. Solid solutions of Zr in (Ni,Cu) at different temperatures



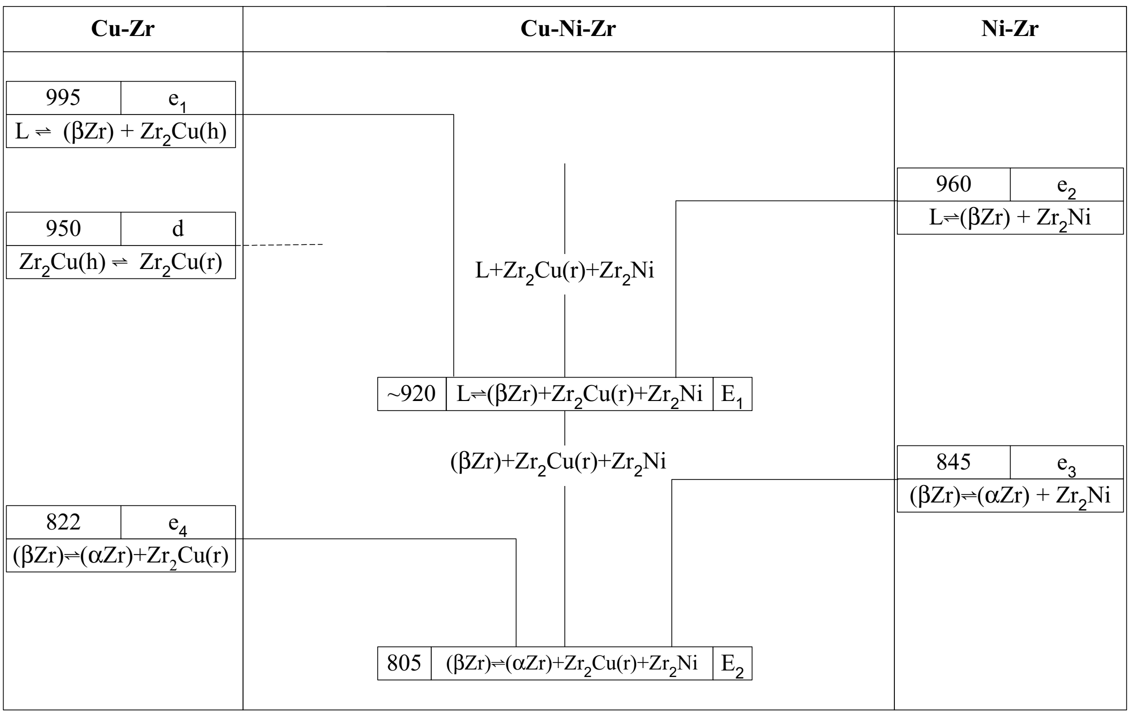


Fig. 2: Cu-Ni-Zr. Partial reaction scheme for the solidification of Zr rich alloys

Fig. 3: Cu-Ni-Zr.
Partial liquidus surface

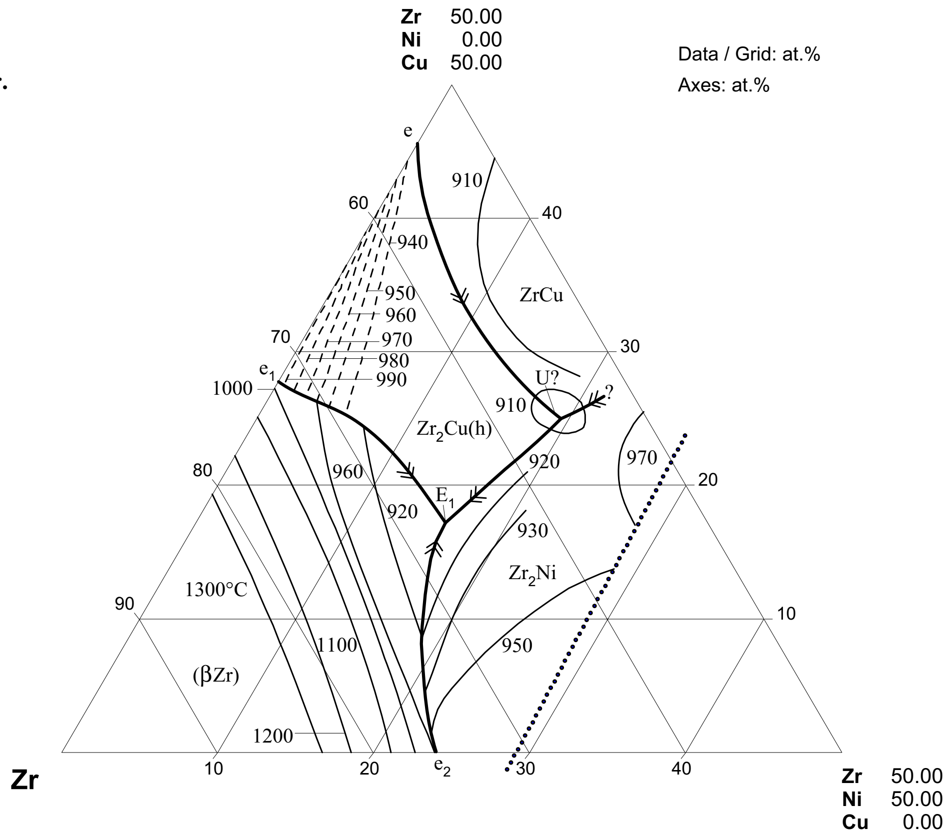


Fig. 4: Cu-Ni-Zr.
Partial isothermal section at 1300°C

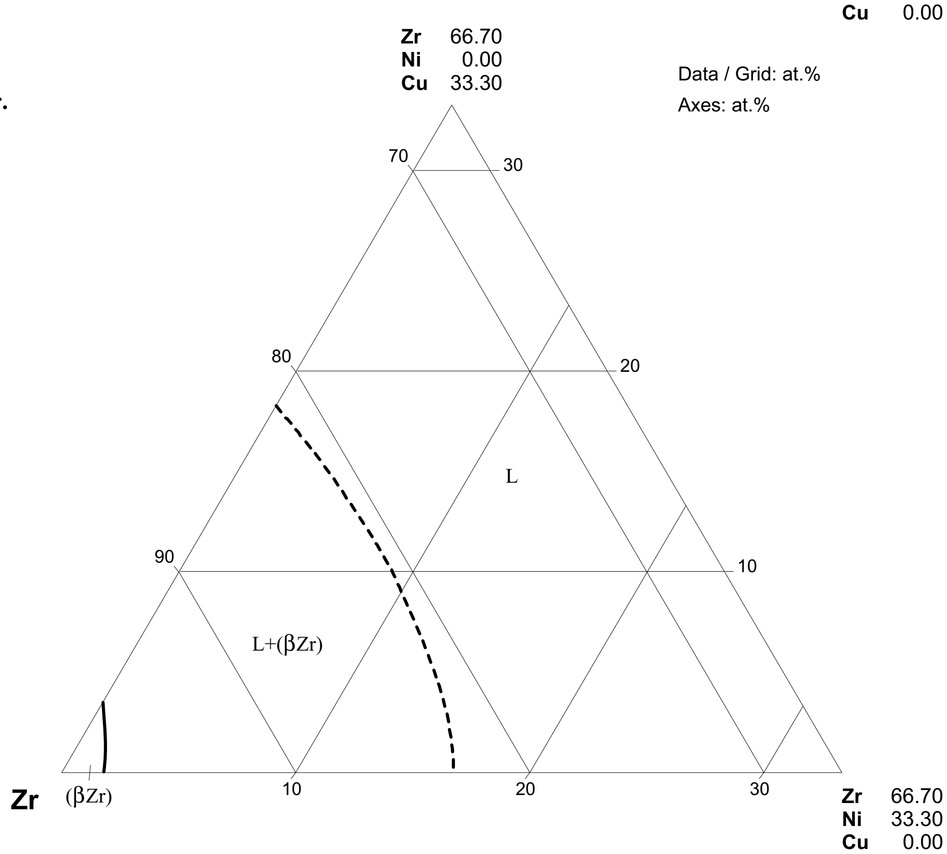


Fig. 5: Cu-Ni-Zr.
Partial isothermal section at 950°C

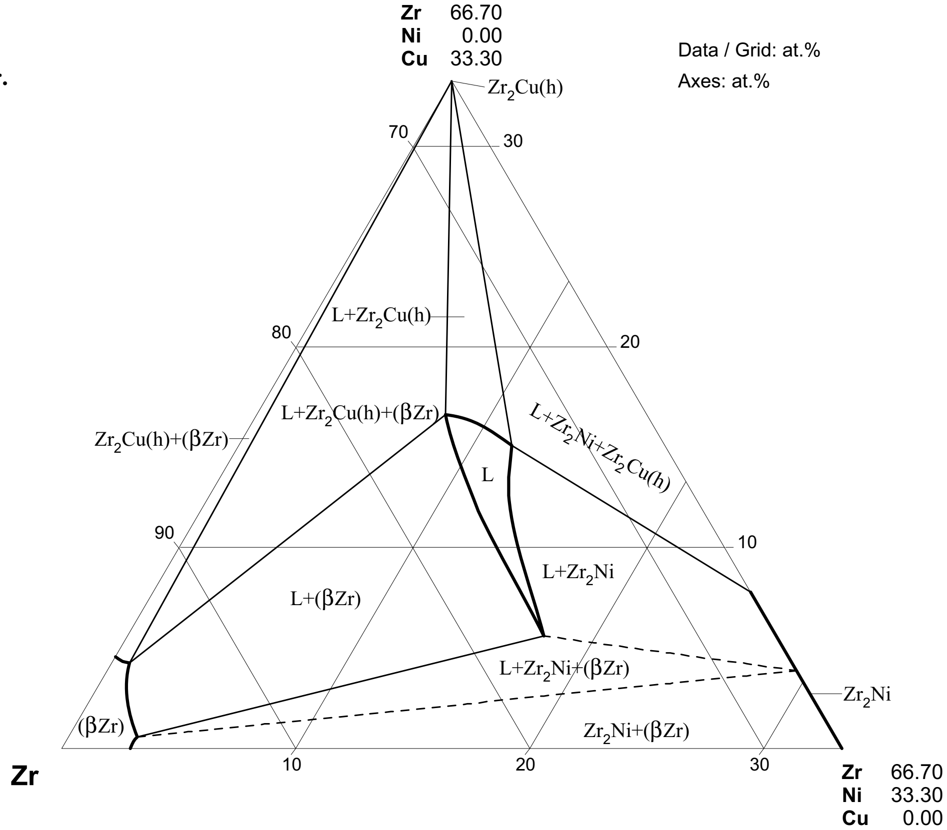


Fig. 6: Cu-Ni-Zr.
Partial isothermal section at 850°C

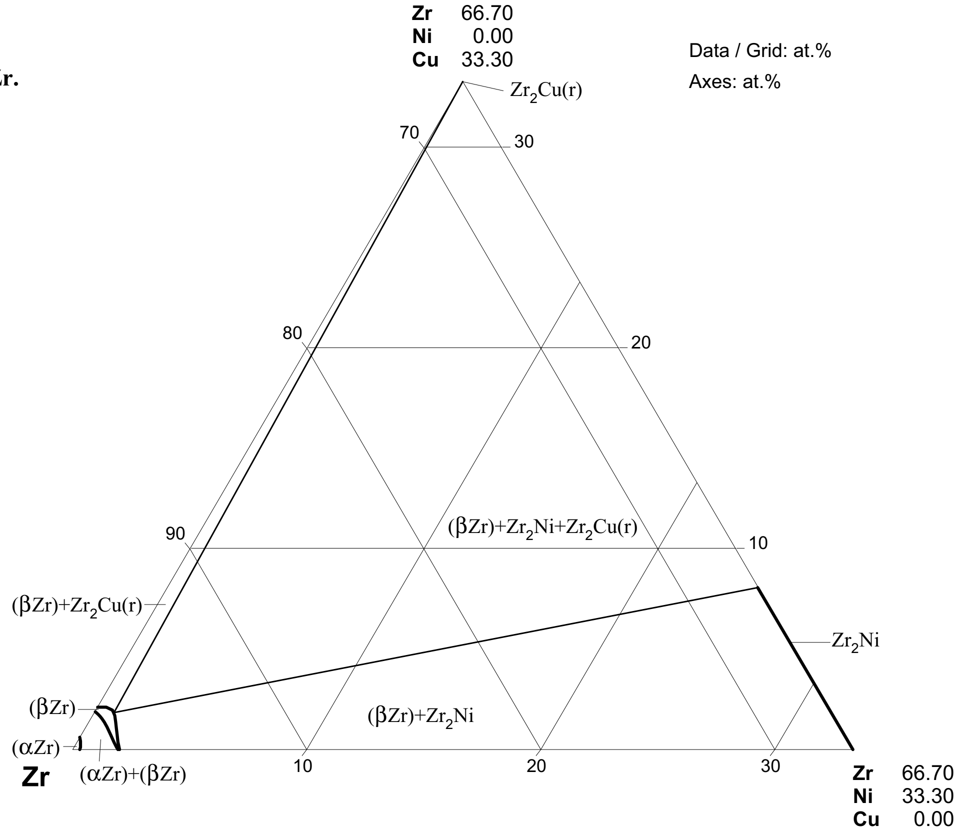


Fig. 7: Cu-Ni-Zr.
Partial isothermal section at 800°C

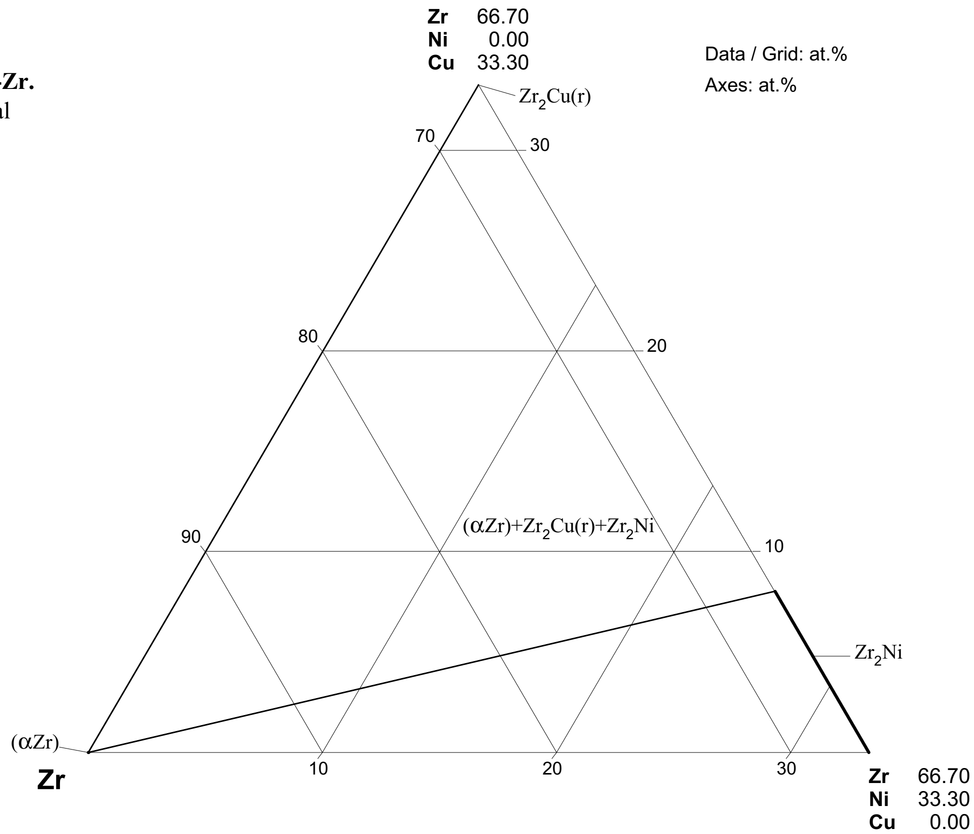


Fig. 8: Cu-Ni-Zr.
Isopleth at a constant Ni content of 0.2 mass%, plotted in at.%

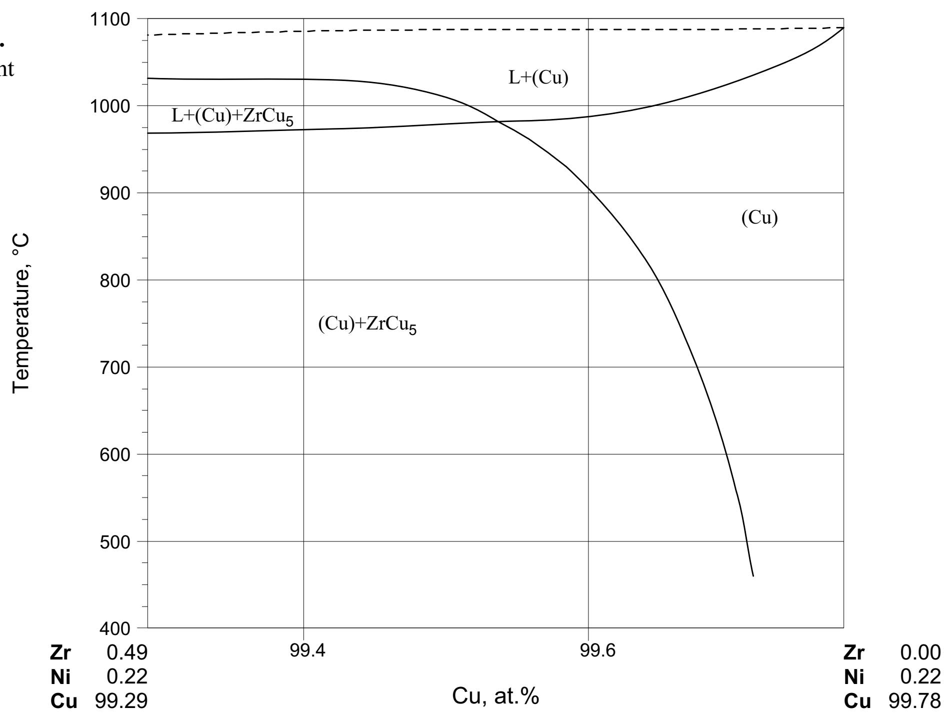


Fig. 9: Cu-Ni-Zr. Isopleth at a constant Ni content of 0.6 mass%, plotted in at.%

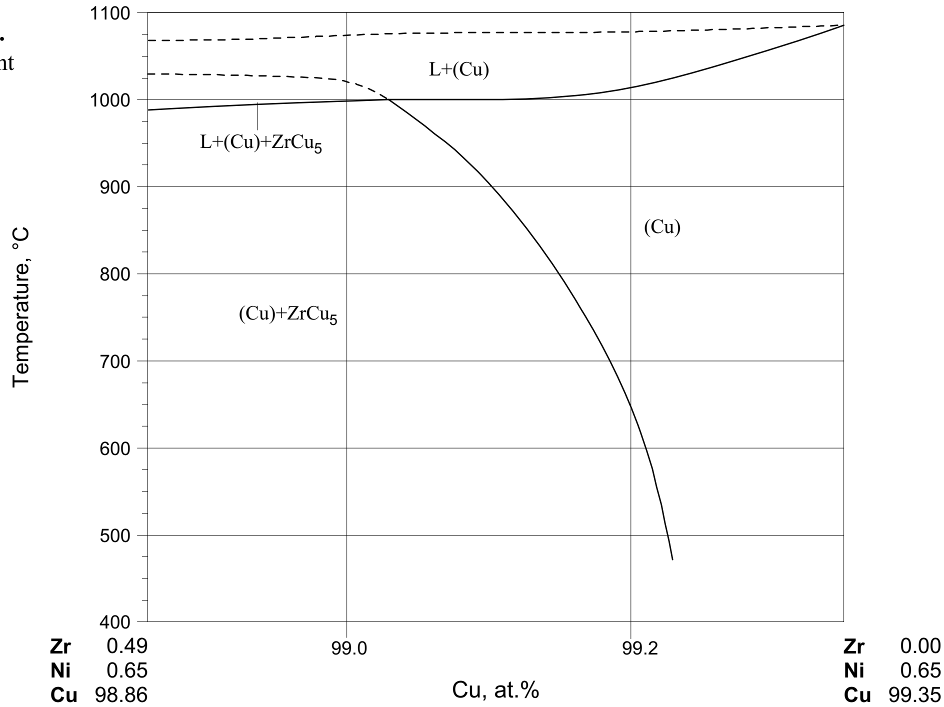


Fig. 10: Cu-Ni-Zr. Isopleth at a constant ratio of Cu:Ni=4:1

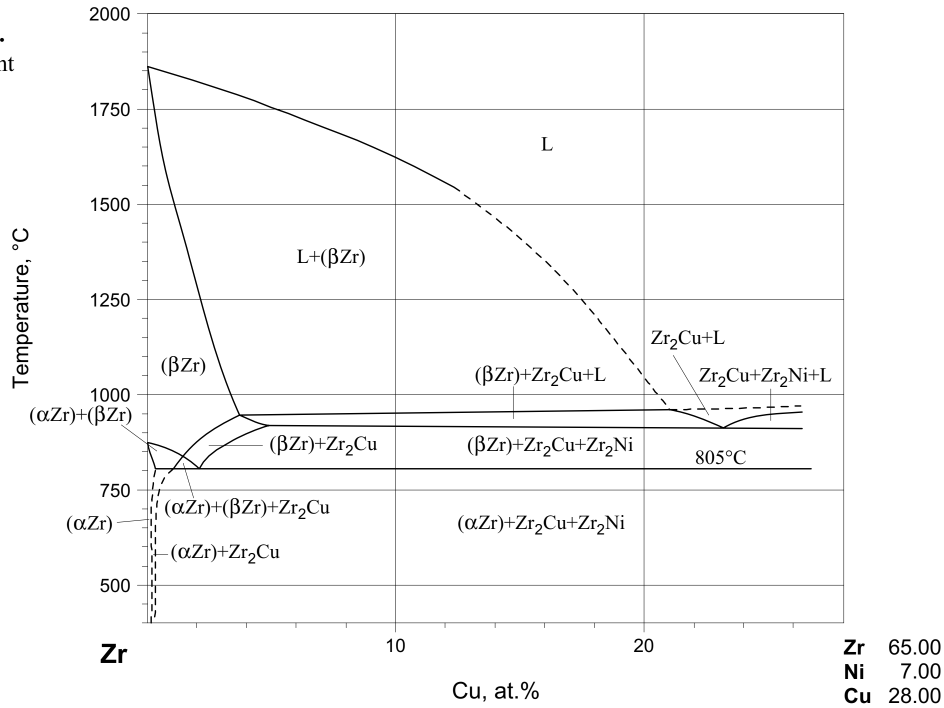


Fig. 11: Cu-Ni-Zr. Isopleth at a constant ratio of Cu:Ni=1:1

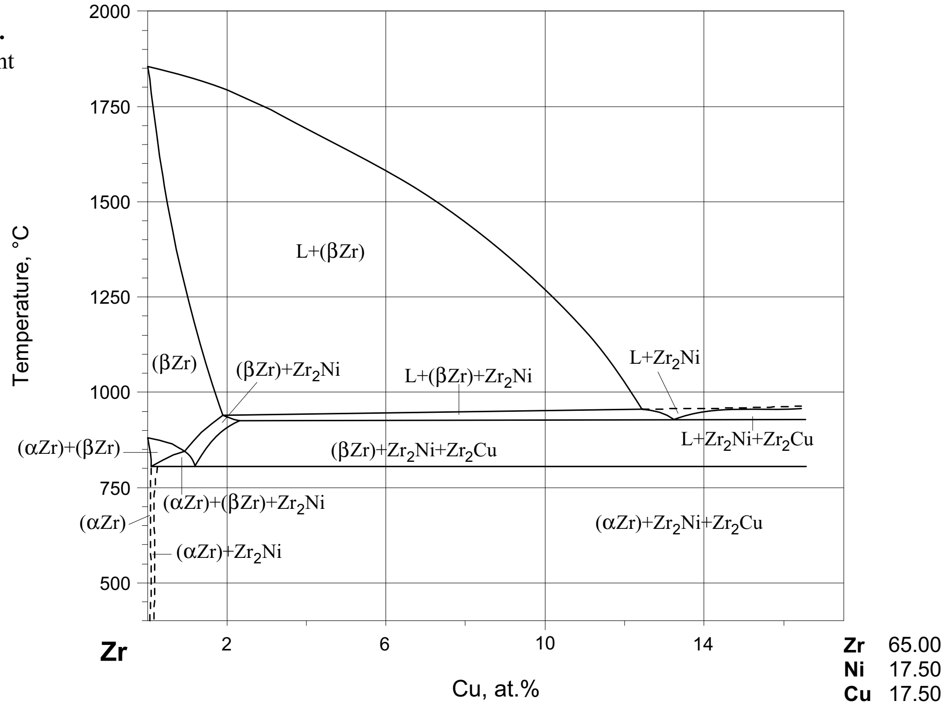


Fig. 12: Cu-Ni-Zr. Projection of isoenthalpic lines of liquid and undercooled liquid alloys at 1292°C, in kJ·mol[−1]