Copper – Manganese – Silicon

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Introduction

The Cu rich part of the ternary system Cu-Mn-Si has been extensively studied. All the experimental data on this system have been reported in Table 1.
Two ternary phases τ_1 and τ_2 have been observed and agreement is noticed concerning their compositions. Several isothermal sections have been determined experimentally in the Cu rich part of the ternary system. [1930Smi, 1942Smi, 1945Smi] investigated alloys containing up to 10 mass% Mn and 10 mass% Si and presented some partial isothermal sections ranging from 450 to 800°C. More recently, [1974Muk] studied Mn rich alloys containing up to 25 mass% Si and 70 mass% Cu and reported a partial isothermal section at 700°C. General agreements have been observed. However, the data of [1930Smi] considerably deviate from those of the other studies at 800 and 700°C, if the manganese content is higher than 2 mass%. This has been noticed by [2003Mie] who carried out a complete thermodynamic description in the Cu rich part of the ternary system using ThermoCalc software. The calculations are in quite good agreement with the experimental data reported previously. Nevertheless, some calculated phase equilibria involving two ternary compounds τ_1 and τ_2 are not in agreement with those measured experimentally by [1974Muk].
In the review made by [1979Cha], a liquidus surface has been reported which is inferred from the studies of the Cu rich region by [1930Smi, 1942Smi, 1958Tur] as well the sections Cu-Mn_5Si_3 [1953Dre, 1962Smi], Cu-MnSi [1962Smi] and Cu-$MnSi_2$ [1962Smi]. The calculated liquidus surface presented by [2003Mie] in the Cu rich part (up to 30 mass% Mn and 10 mass% Si) correlates well with the experimental data mentioned previously.
The quasibinary system Cu-Mn_5Si_3, reported as a eutectic type, has been well established and experimental data of [1953Dre] and [1962Smi] are in agreement. The Cu-MnSi section was also considered as quasibinary by [1962Smi]. Others isopleths have also been reported at Mn:Si = 1.96 [1962Smi], at 2 and 4 mass% Si [1930Smi, 1958Tur], and at 2 and 4 mass% Mn [1930Smi, 1958Tur]. The calculated isopleths [2003Mie] are in quite good agreement with those reported experimentally.
As [1979Cha], [1969Gue, 1961Die] undertook a review of the system Cu-Mn-Si.

Binary Systems

The binary systems Cu-Mn and Cu-Si are taken from [2005Tur] and [2002Leb], respectively. The phase diagram Mn-Si is accepted from [1991Oka].

Solid Phases

All the crystallographic data for the unary, binary and ternary phases are reported in Table 2.
Two ternary compounds have been reported in the literature [1973Muk, 1974Muk]. τ_1 crystallizes in the $MgZn_2$ type structure and exhibits a very small phase region around the mean composition $Mn_{50.2}Cu_{32.5}Si_{17.3}$. Its crystallographic parameters have been reported in Table 2. τ_2 has an approximate composition of $Mn_{60.5}Cu_{28.3}Si_{11.2}$. These compounds exist at 700°C [1974Muk] but no information can be found concerning their temperature domain of existence.

Quasibinary Systems

The quasibinary system Cu-Mn_5Si_3 is of eutectic type and the eutectic point is located at 800°C and at about 24 mass% Mn_5Si_3 [1953Dre, 1962Smi] as shown in Fig. 1. The Cu-MnSi system is also quasibinary with the eutectic point located at 21 mass% MnSi and 775°C [1962Smi]. The diagram shown in Fig. 2 and is a compilation of [1962Smi] and [2003Mie].

Invariant Equilibria

A tentative reaction scheme is reported in Fig. 3. The invariant equilibria are reported in Table 3. The location of the ternary four-phase reactions have been taken from [1979Cha]. Two three-phase reactions have been also reported and were deduced from the quasibinary sections Cu-MnSi and Cu-Mn_5Si_3.

Liquidus, Solidus and Solvus Surfaces

The liquidus surface reported in Fig. 4 is a compilation of the experimental works done by [1930Smi, 1942Smi, 1958Tur]. The liquidus surface is drawn to be consistent with the binary systems and with the experimental polythermal sections available in the literature [1953Dre, 1930Smi, 1962Smi] as well as with the calculated liquidus curves suggested by [2003Mie]. The extrapolated curves are indicated as dotted lines in the drawing. Lack of data is observed near the Mn rich part. Consequently, the phase equilibria could not be determined and were symbolized as a question mark on the drawing.

Isothermal Sections

At 700°C [1974Muk] investigated the phase equilibria and found some solubility ranges for the binary phases inside the ternary system. The solubility of Cu in the (αMn), (βMn) and R phases is small. The region limits of the ν, βMn_3Si and Mn_5Si_3 inside the ternary were not determined accurately. Figure 6 shows the partial isothermal section at 700°C which is a compilation of the experimental work of [1974Muk] and the thermodynamic calculations by [2003Mie]. The phase equilibria involving the two ternary phases have been taken from the experimental work [1974Muk]. These equilibria were not reproduced by the thermodynamic calculations [2003Mie]. Other isothermal sections were measured and calculated from 450 to 800°C. Some of them are reported in Figs. 5, 7 and 8. General good agreement was observed between the experimental data [1930Smi, 1942Smi, 1945Smi, 1953Dre, 1958Tur, 1962Smi] and the calculated curves [2003Mie]. However, the solubility range of the (γMn,Cu) measured by [1930Smi] has not been in agreement with those of the other studies at 800 and 700°C if the Mn content is higher than 2 mass%. The data of the latter studies have been retained in this assessment.

Temperature – Composition Sections

Partial polythermal sections have been reported in the literature: from 1 to 4 mass% Si [1958Tur], from 1 to 12 mass% Mn [1930Smi, 1958Tur]. [2003Mie] calculated isopleths which are in quite good agreement with measured data deduced from experimental isothermal sections and isopleths. Nevertheless, the calculated isopleth at Mn:Si = 1.96 in the Cu rich part presents large discrepancies with the isothermal section measured at 700°C by [1974Muk]. The three-phase equilibrium (γMn,Cu) +τ_1 + τ_2 suggested by [2003Mie] was not observed by [1974Muk]. This latter study measured a very narrow (γMn,Cu) - βMn_3Si region splitting the (γMn,Cu) +τ_1 + τ_2 region into two separate triangles, (γMn,Cu) + τ_1 + βMn_3Si and (γMn,Cu) + τ_2 + βMn_3Si. Since these last three-phase equilibria have not been mentioned in the work [2003Mie], the isopleth Cu-$Mn_{1.96}Si$ was not retained in this assessment. Some isopleths are shown in Figs. 9, 10 and 11.

Notes on Materials Properties and Applications

Experimental data on materials properties are reported in Table 4.
[1930Voc] studied the mechanical and physical properties of Cu-Mn-Si alloys in the cast, drawn and rolled conditions with a view to developing and extending their uses. It was concluded that Cu alloys containing between 2 and 5 mass% Si and 0 - 5 mass% Mn have been the most useful alloys combining good strength and hardness with reasonable malleability. The mechanical properties of Cu rich alloys containing 1-2 mass% Mn and up to 2.5 mass% Si were investigated also by [1952Dre]. These alloys are found to be amenable to age hardening. In quenched alloys with 1.2 mass% Mn, a rise in silicon concentration from 2.5 to 4.4 mass% first increases and then reduces the effect of stress rise under relaxation test conditions at 200 and 300°C [1968Gay]. The hardness is found to be non-monotonic function by [1973Fri] with tow peaks,

low temperature at 250-300°C and high temperature at 500-550°C. An increase of the maximum of the hardening is observed in alloys Cu-Mn_5Si_3 as the Mn_5Si_3 content increases [1953Dre] leading to a value of about 177 kg·mm^{-2} at 12.5 mass% of Mn_5Si_3. [1967Mil] observed that the alloys Cu-3 at.% Si-1 at.% Mn with high reduction (80%) after annealing (30 min at 450°C) have a hardness (59 kg·mm^{-2}) higher than those which were only annealed (40.7 kg·mm^{-2}).

Miscellaneous

[1988Pra] investigated the microstructure in the cold-worked state of Cu-Mn-Si alloys in the fcc phase of Cu and studied the effects of Mn and Si. It was concluded that the presence, even in small amounts of Mn and Si, has a pronounced effect on the stacking fault probability of deformed Cu-Mn-Si alloys. [1948Ray] examined the effect of the compound Mn_2Si on the solid solubility isotherms of Cu-Mn-Si alloys and showed that the (γMn,Cu) / (γMn,Cu) + Mn_2Si obey to the relation $d\log K / d\theta = -Q / (R\theta^2)$ with $Q = 7.6 \cdot 10^3$ cal., where θ is the absolute temperature, Q is analogous to the heat of formation, K is a constant deduced from the solid solubility isothermls.
[1965Bra] developed a technique to prevent cracking in the welding of highly restrained 95.8 at.% Cu - 3.1 at.% Si - 1.1 at.% Mn alloys by controlling segregation in the weld. It has been observed that centerline cracking is associated with the dendritic type of structure and becomes arrested at the point where the structure changed from dendritic to cellular. Beyond a certain reduction (about 35%), the deformation by slip of Cu-3 at.% Si - 1 at.% Mn is accompanied by significant macroscopic shearing [1967Mil]. Annealing does not produce any restoration but a recrystallization alone which is done in two enough distinct stages for high reductions (> 50%). The use of high reductions makes it possible to obtain recrystallized states with very fine grains.
[1969Vas1] studied copper alloys containing about 3.48 mass% Si and 1.14 mass% Mn at liquid nitrogen temperature. The faulting parameters α (an average of $\alpha_{111\text{-}200}$ and $\alpha_{200\text{-}220}$) and β were determined at various annealing times after filling (1 and 3 h, 1 and 4 d). It was observed that the value of the parameter α was about 35% more at liquid nitrogen temperature than at room temperature estimated as 0.022 by [1969Vas2]. Using variance method on copper alloys containing 6.7 at.% Si and 1.3 at.% Mn, [1970Vas] found an apparent particle size and strain of 98 Å and 0.0018, respectively which are closer to those given by Fourier analysis than by integral breadth measurements.

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Table 1: Investigations of the Cu-Mn-Si Phase Relations, Structures and Thermodynamics

Reference	Method/Experimental Technique	Temperature/Composition/Phase Range Studied
[1930Smi]	Thermal analysis	450 - 1100°C / Cu rich alloys up to 10 mass% Mn and 8 mass% Si
[1942Smi]	Thermal analysis	450-800°C / Cu rich alloys up to 10 mass% Mn and 8 mass% Si
[1945Smi]	Microstructural analysis	500 - 800°C / Cu based alloys with up to 8 mass% Si and Mn
[1953Dre]	Thermal analysis and microstructure	400 - 100°C / Cu based alloys with 0 to 50 mass% Mn_5Si_3
[1958Tur]	Thermal analysis	400 - 900°C / Cu based alloys with 1-12 mass% Mn and 1-4 mass% Si
[1962Smi]	Thermal analysis and X-ray diffraction	500 - 1200°C / Cu-MnSi, Cu-Mn_5Si_3, from Cu to 8 mass%MnSi2
[1973Muk]	Metallography and X-ray diffraction	25°C / Cu based alloys with 11.5 to 25 at.% Si and 33 to 62.5 at.% Mn
[1974Muk]	X-ray diffraction	700°C / Cu based alloys with 60 to 10 at.% Si and 30 to 100 at.% Mn

Table 2: Crystallographic Data of Solid Phases

Phase/ Temperature Range [°C]	Pearson Symbol/ Space Group/ Prototype	Lattice Parameters [pm]	Comments/References
(γMn,Cu)	*cF*4 $Fm\bar{3}m$ Cu	a = 362.63 a = 375	at x = 0.04 [V-C2] at x = 0.80 [V-C2]
(Cu) < 1084.62		a = 361.46 a = 360.82	at x = 0 [Mas2] at x = 0 [V-C2] melting point [2005Tur]
(γMn) 1138 - 707		a = 386	at x = 1 [Mas2]
(δMn) 1246 - 1138	*cI*2 $Im\bar{3}m$ W	a = 308.0	[Mas2]
(βMn) 1087 - 707	*cP*20 $P4_132$ βMn	a = 631.52	[Mas2]

Phase/ Temperature Range [°C]	Pearson Symbol/ Space Group/ Prototype	Lattice Parameters [pm]	Comments/References
(αMn) < 707	*cI*58 $I\bar{4}3m$ αMn	a = 891.26	at 25°C [Mas2]
(Si) < 1414	*cF*8 $Fd\bar{3}m$ C-diamond	a = 543.06	0 to 0.003 at.% Cu [2002Leb]
γ_3 ≤ 700	*c***	-	[2005Tur]
$MnCu_3$ (γ_2) ≤ 450	*c***	-	[2005Tur]
$MnCu_5$ (γ_1) ≤ 410	*c***	-	[2005Tur]
R, Mn_6Si < 880	*hR*159 $R\bar{3}$ $Co_5Cr_2Mo_3$	a = 1087.1 ± 0.5 c = 1918.0 ± 0.9	12 to 15.75 at.% Si [1991Oka] [V-C2]
ν, Mn_4Si < 1060	*oI*186 *Immm* Mn_4Si	a = 1699.2 ± 0.4 b = 2863.4 ± 0.7 c = 465.6 ± 0.1	16.2 to 18.75 at.% Si [1991Oka] [V-C2]
βMn_3Si 1075.3 - 677	*cF*16 $Fm\bar{3}m$ BiF_3	a = 572.2	24 to 25.6 at.% Si [1991Oka] [V-C2]
αMn_3Si < 677	-	-	25 to 25.6 at.% Si [1991Oka]
Mn_5Si_2 < 850	*tP*56 $P4_12_12$ Mn_5Si_2	a = 890.97 ± 0.2 c = 871.53 ± 0.3	28.6 at.% Si [1991Oka] [V-C2]
Mn_5Si_3 < 1283.2	*hP*16 $P6_3/mcm$ Mn_5Si_3	a = 691 c = 468.4	37.5 at.% Si [1991Oka] [V-C2]
MnSi < 1269.6	*cP*8 $P2_13$ FeSi	a = 455.8	40.5 to 50.2 at.% Si [1991Oka] [V-C2]
$Mn_{11}Si_{19}$ < 1150.2	*tP*120 $P\bar{4}n2$ $Mn_{11}Si_{19}$	a = 552 c = 4820	63.3 at.% Si [1991Oka]
κ, Cu_7Si 842 - 552	*hP*2 $P6_3/mmc$ Mg	a = 256.06 c = 418.46	11.05 to 14.5 at.% Si at 12.75 at.% Si [2002Leb]
β, ~Cu_6Si 853 - 787	*cI*2 $Im\bar{3}m$ W	a = 285.4	14.2 to 16.2 at.% Si at 14.9 at.% Si [2002Leb]

Phase/ Temperature Range [°C]	Pearson Symbol/ Space Group/ Prototype	Lattice Parameters [pm]	Comments/References
δ, Cu_5Si(h) 824 - 711	*t***	*a* = 881.5 *c* = 790.3	17.6 to 19.6 at.% Si Sample was annealed at 700°C [2002Leb]
γ, Cu_5Si(r) < 729	*cP*20 $P4_132$ βMn	*a* = 619.8	17.15 to 17.6 at.% Si [2002Leb]
ε, $Cu_{15}Si_4$ < 800	*cI*76 $I\bar{4}3d$ $Cu_{15}Si_4$	*a* = 961.5	21.2 at.% Si [2002Leb]
η, $Cu_3Si(h_2)$ 859 - 558	*hR** $R\bar{3}m$ or *t***	*a* = 247 α = 109.74° *a* = 726.7 *c* = 789.2	23.4 to 24.9 at.% Si [2002Leb] [V-C2]
η', $Cu_3Si(h_1)$ 620 - 647	*hR** $R\bar{3}$	*a* = 472 α = 95.72°	23.2 to 25.2 at.% Si [2002Leb]
η'', Cu_3Si(r) < 570	*o***	*a* = 7676 *b* = 700 *c* = 2194	23.3 to 24.9 at.% Si [2002Leb]
* τ_1, $Mn_{50.2}Cu_{32.5}Si_{17.3}$	*hP*12 $P6_3/mmc$ $MgZn_2$	*a* = 483 *c* = 783.4	[1974Muk]
* τ_2, $Mn_{60.5}Cu_{28.3}Si_{11.2}$	-	-	[1974Muk]

Table 3: Invariant Equilibria

Reaction	*T* [°C]	Type	Phase	Composition (at.%)		
				Cu	Mn	Si
L + $Mn_{11}Si_{19}$ ⇌ (Si) + MnSi	< 1144	U_1	L	8.01	30.65	61.34
			$Mn_{11}Si_{19}$	0	36.66	63.34
			(Si)	0	0	100
			MnSi	0	50	50
L + δ ⇌ β + η	< 820	U_2	-	-	-	-
L + (Si) ⇌ η + MnSi	< 802	U_3	(Si)	0	0	100
			MnSi	0	50	50
l ⇌ (γMn,Cu) + Mn_5Si_3	800	e_6	l	69.10	19.31	11.59
			Mn_5Si_3	0	62.5	37.5
L + η ⇌ β + MnSi	< 800	U_4	MnSi	0	50	50
l ⇌ (γMn,Cu) + MnSi	775	e_7	l	71.08	14.46	14.46
			MnSi	0	50	50
L ⇌ Mn_5Si_3 + MnSi + (γMn,Cu)	< 775	E_1	L	70.85	16.82	12.33
			Mn_5Si_3	0	62.5	37.5
			MnSi	0	50	50
L ⇌ β + MnSi + (γMn,Cu)	760	E_2	L	74.43	9.34	16.23
			MnSi	0	50	50

Table 4: Investigations of the Cu-Mn-Si Materials Properties

Reference	Method/Experimental Technique	Type of Property
[1930Voc]	Tensile tests	Mechanical properties
[1942Smi]	Electrical conductivity measurements	Electrical conductivity
[1952Dre]	Mechanical measurements	Mechanical properties
[1953Dre]	Hardening measurement	Mechanical properties
[1961Die]	Hardening measurement	Mechanical properties
[1967Mil]	Hardening measurement	Mechanical properties
[1968Gay]	Stress relaxation measurement	Mechanical properties
[1973Fri]	Hardness measurement	Mechanical properties

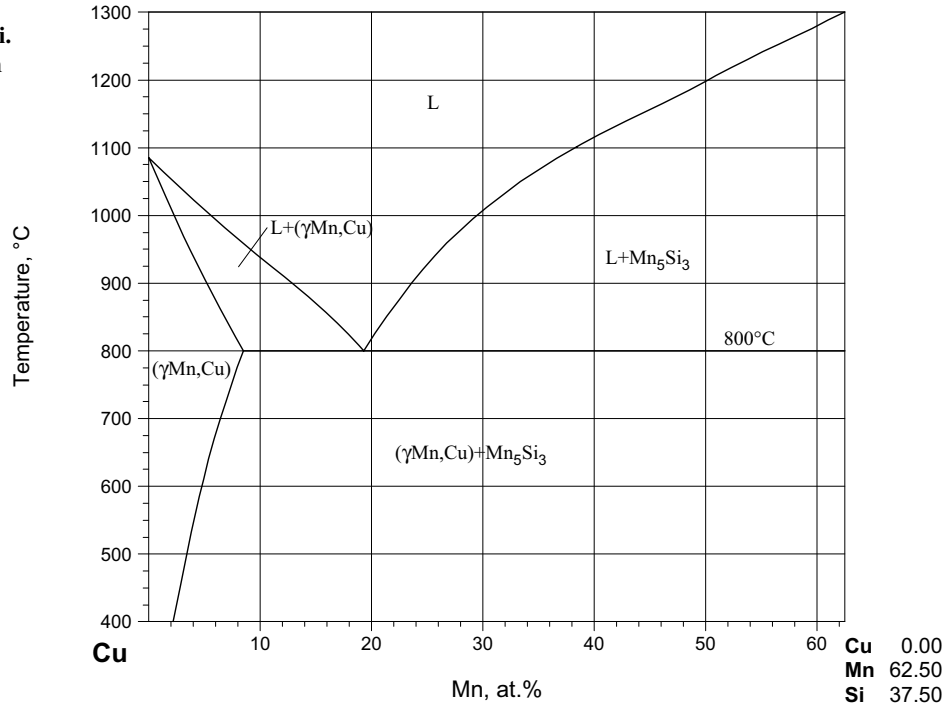

Fig. 1: Cu-Mn-Si. Quasibinary system Cu - Mn_5Si_3

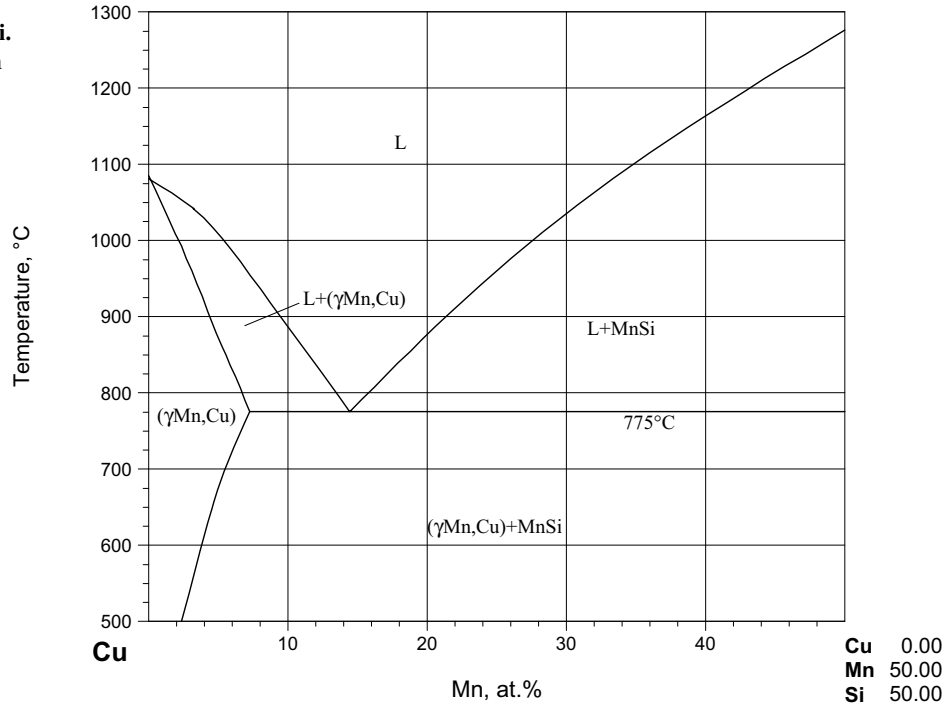

Fig. 2: Cu-Mn-Si. Quasibinary system Cu - MnSi

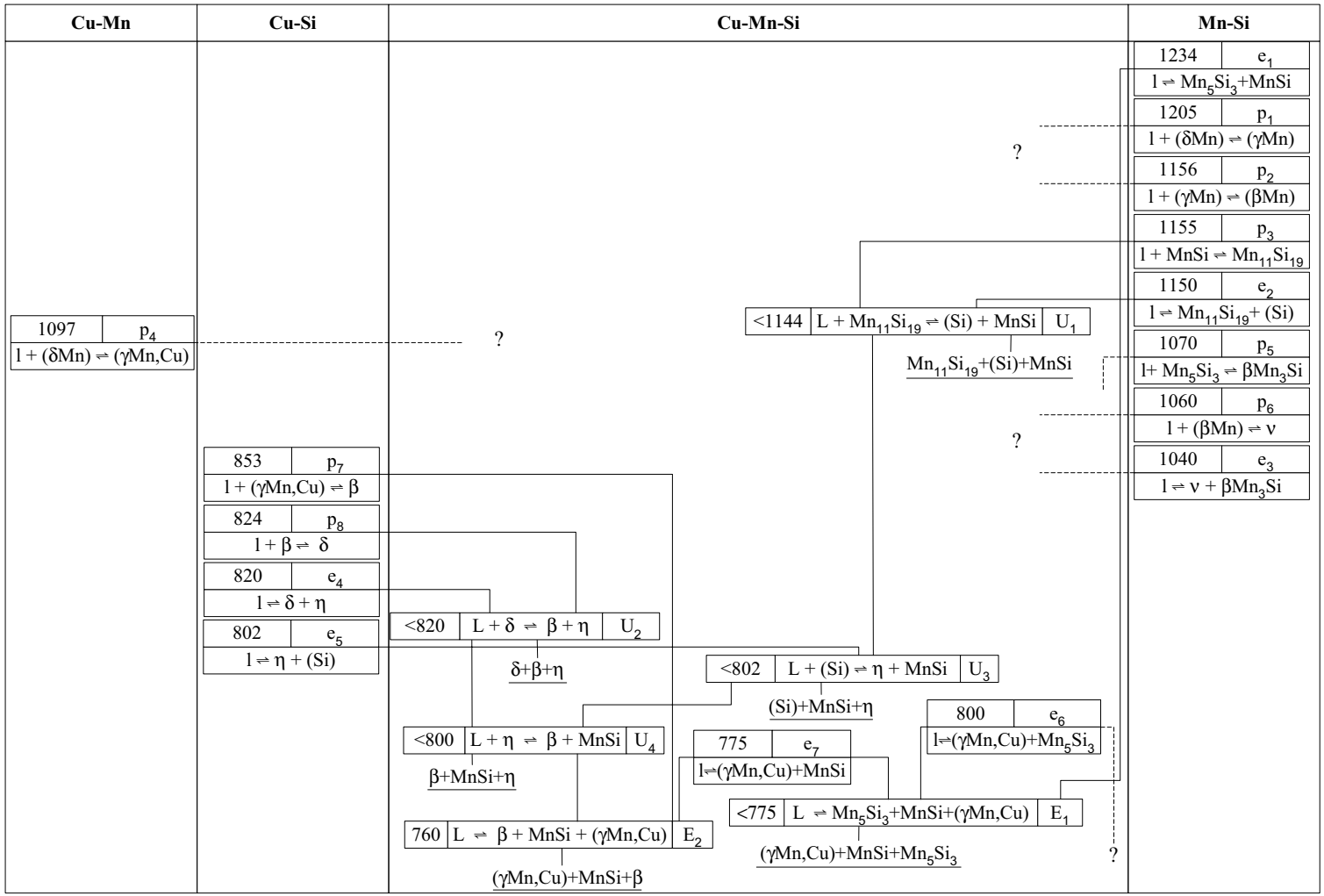

Fig. 3: Cu-Mn-Si. Reaction scheme

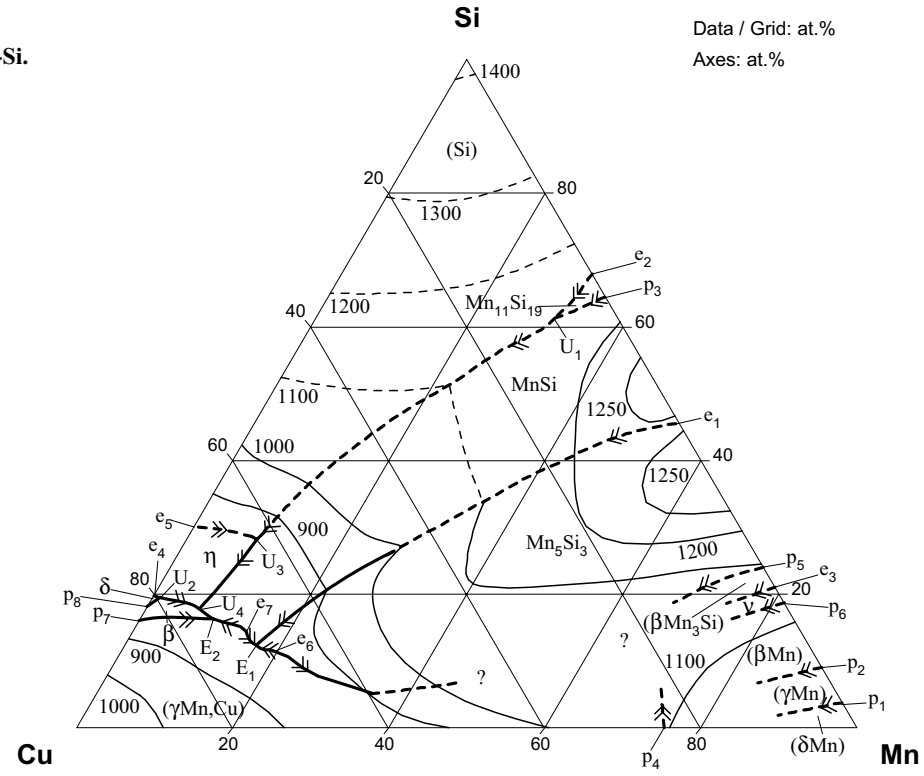

Fig. 4: Cu-Mn-Si. Liquidus surface

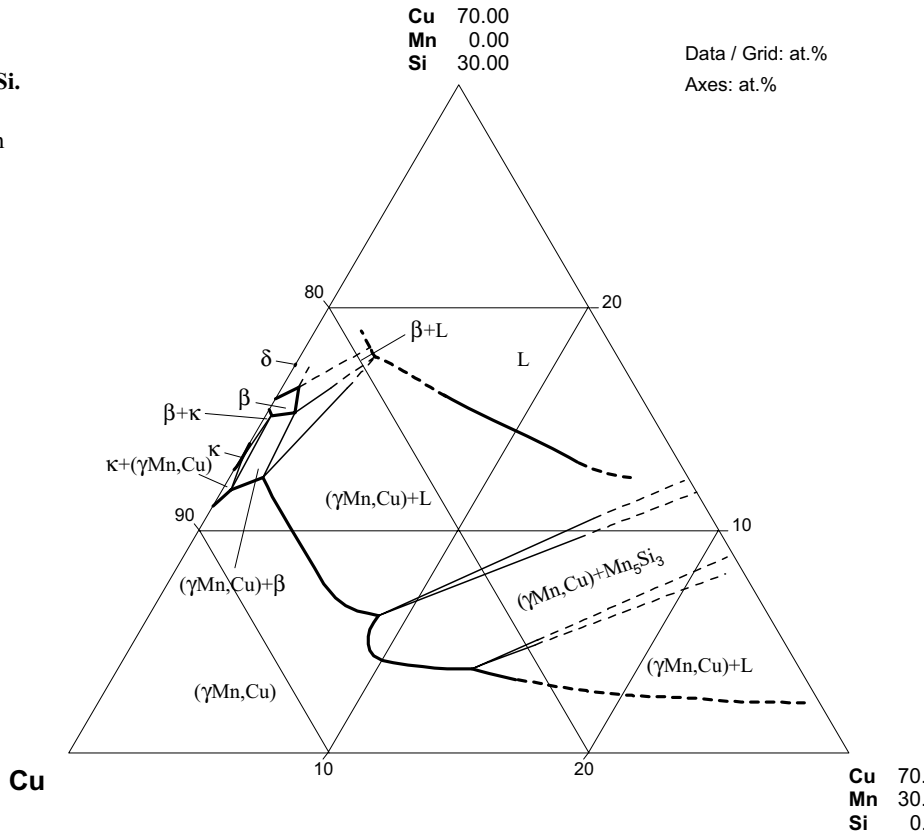

Fig. 5: Cu-Mn-Si. Partial isothermal section at 800°C in the Cu rich corner

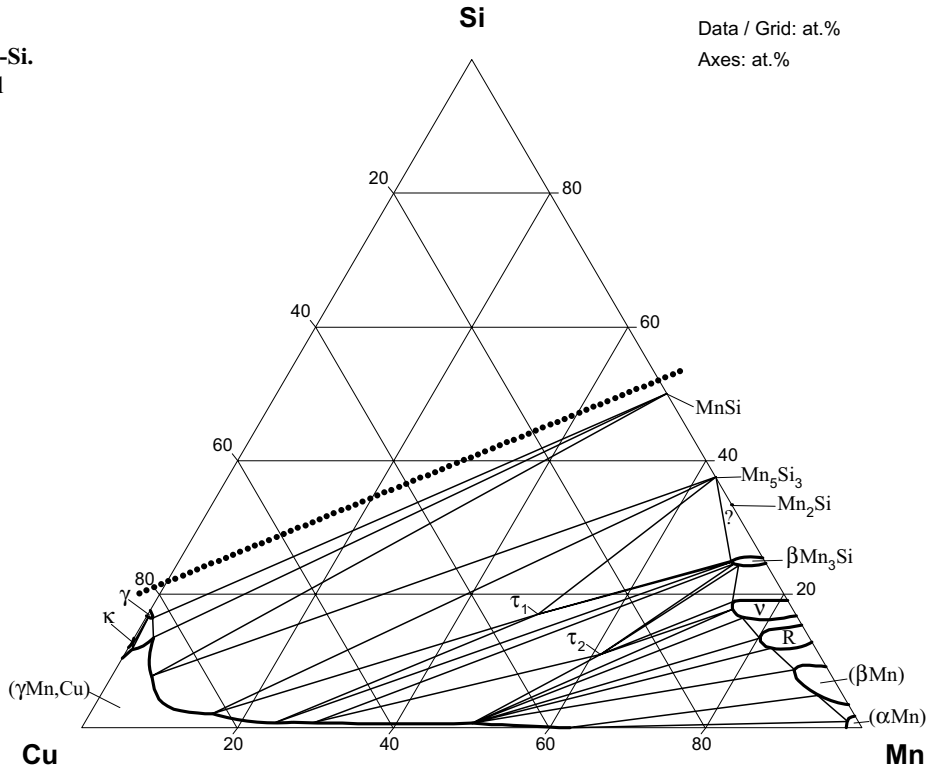

Fig. 6: Cu-Mn-Si. Partial isothermal section at 700°C

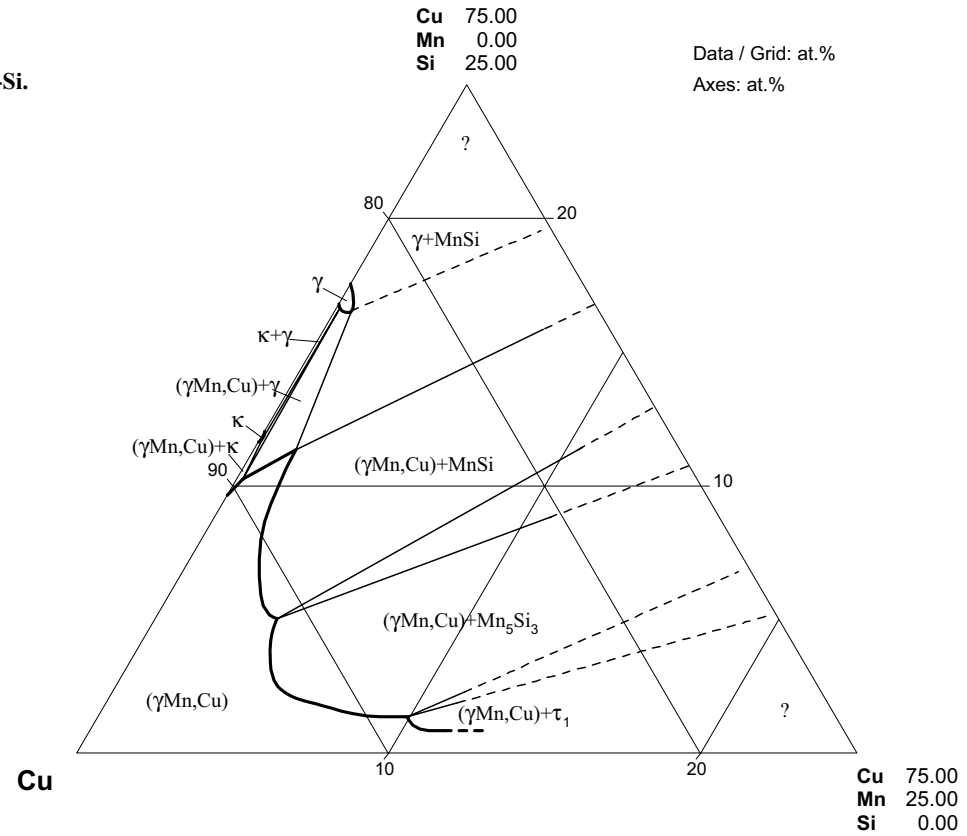

Fig. 7: Cu-Mn-Si. Partial isothermal section at 600°C

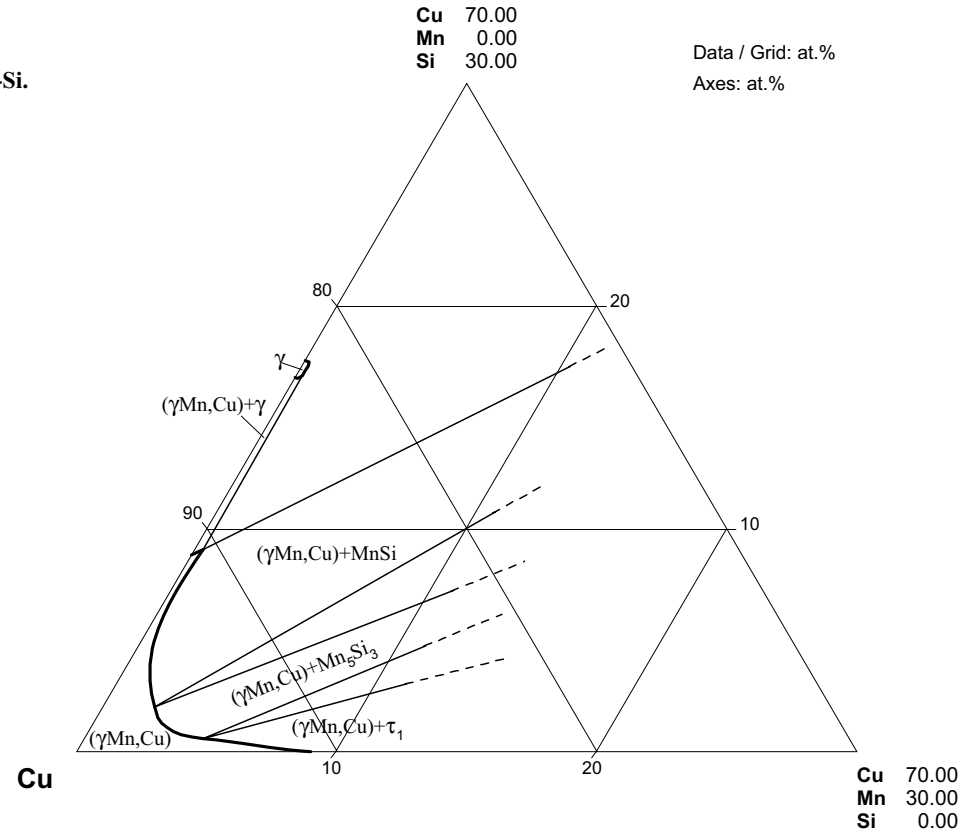

Fig. 8: Cu-Mn-Si. Partial isothermal section at 450°C

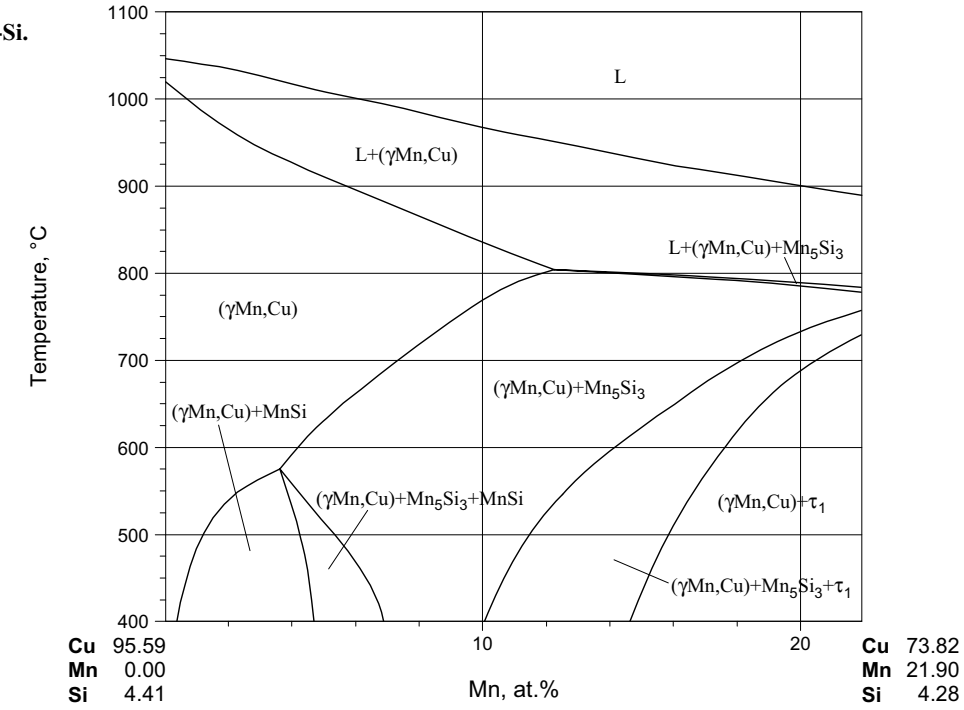

Fig. 9: Cu-Mn-Si. Partial isothermal section at 450°C

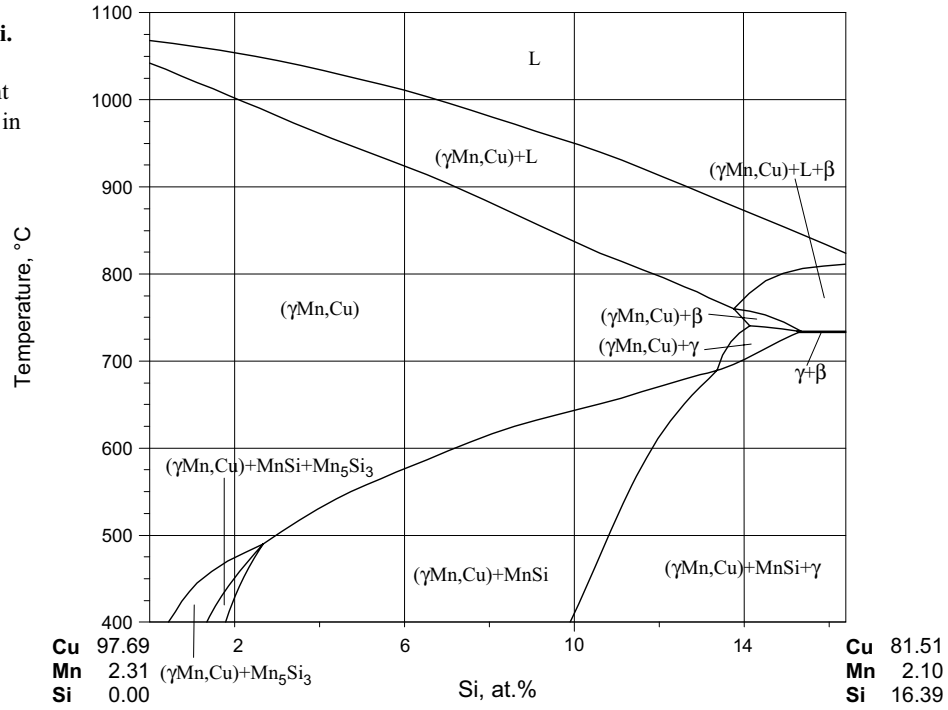

Fig. 10: Cu-Mn-Si. Isopleth at the constant Mn content of 2 mass%, plotted in at.%

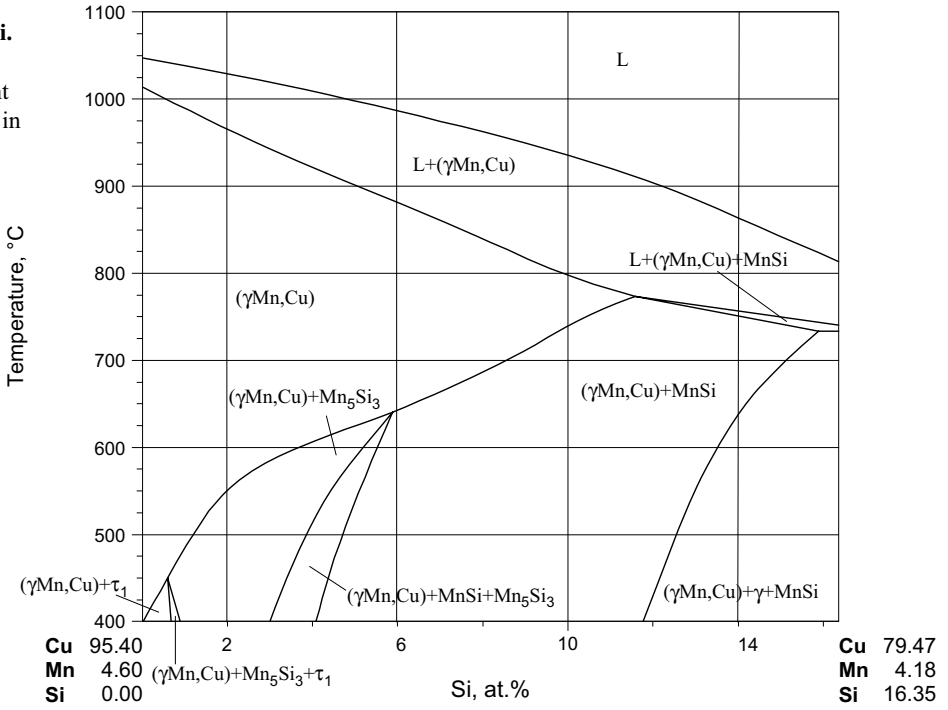

Fig. 11: Cu-Mn-Si. Isopleth at the constant Mn content of 4 mass%, plotted in at.%