

Copper – Tin – Titanium

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Introduction

Phase equilibria data concerning the Cu–Sn–Ti system are mainly based on the experimental work of [1999Kob, 2001Nak1]. The first extensive investigation of the Cu–Sn–Ti ternary system is concerned with the determination of the isothermal section at 397°C [1999Kob]. In that work 123 alloys were prepared by arc melting of commercial purity elements (Ti: 99.93 mass%; Cu: 99.96 mass%, Sn: 99.99 mass%) in argon atmosphere. The phase equilibria of annealed alloy samples were investigated by XRD and electrical resistivity measurements. [2001Nak1] determined the liquidus surface of the Cu–Sn–Ti ternary system. By arc melting under purified argon gas, Cu–Sn–Ti ingots were prepared from pure elements (Cu, Sn and Ti with 99.9 mass% purity), and all samples were annealed at 800°C for 168 h. The liquidus surface of the Cu–Sn–Ti ternary system was then established by determining the primary crystallizing phases as well as the phase reactions involving the liquid phase. A partial reaction scheme was also proposed by [2001Nak1]. More recent experimental works about the Cu–Sn–Ti ternary system are summarized in Table 1.

Binary Systems

The three binary systems are characterized by a series of peritectic reactions and by a number of stable and metastable intermetallic phases. Assessments of the Cu–Sn system by [2005Fra], of the Cu–Ti system by [2002Ans] and of the Sn–Ti system by [2005Liu], are accepted in the present work.

They are based on previous assessment from [1996Shi, 2000Moo] for Cu–Sn, [1996Kum, 2002Can] for Cu–Ti and [1987Mur, 1998Kup] for Sn–Ti.

Solid Phases

The crystallographic data of the Cu–Sn–Ti phases and their temperature ranges of stability are listed in Table 2. All the Cu–Ti intermetallic phases are included in the review by [2002Ans] except the Ti_3Cu phase because the literature data was only up to 1999 in his review. In accordance with [2002Can] the differential thermal analysis (DTA) of the Cu–80 at.% alloy, Ti_3Cu is stable below 905°C. This result confirmed those reported by [1951Kla] which would lead to a peritectoid $\text{Ti}_2\text{Cu} + (\beta\text{Ti}) \rightleftharpoons \text{Ti}_3\text{Cu}$ and an eutectoid reaction $(\beta\text{Ti}) \rightleftharpoons (\alpha\text{Ti}) + \text{Ti}_3\text{Cu}$ instead of an eutectoid reaction $(\beta\text{Ti}) \rightleftharpoons (\alpha\text{Ti}) + \text{Ti}_2\text{Cu}$ indicated in [2002Ans]. The Sn–Ti phase diagram [1987Mur] shows four intermetallic phases, Ti_3Sn , Ti_2Sn , Ti_5Sn_3 and Ti_6Sn_5 . Recently, a new stable phase, Ti_2Sn_3 , was reported by [1998Kup] and its structure was determined by XRD and Mössbauer spectroscopy [2000Kue, 2003Obr]. $\text{Ti}_{6-x}\text{Cu}_x\text{Sn}_5$ ($0.0 < x < 0.1$) was treated as a solid solution according to the experimental data of [1999Kob], which is an extension of the binary phase Ti_6Sn_5 into the ternary. Four ternary phases, namely $\text{Ti}_5\text{CuSn}_3(\tau_1)$, $\text{TiCuSn}(\tau_2)$, $\text{TiCu}_2\text{Sn}(\tau_3)$ and $\text{Ti}_3\text{CuSn}(\tau_4)$ have been identified in this system. $\text{TiCuSn}(\tau_2)$ was identified as a stoichiometric phase with LiGaGe structure type and $P6_3mc$ space group [1999Kob], but [2000Wei] found that $\text{TiCuSn}(\tau_2)$ crystallizes with the space group $P6_3/mmc$ and is isostructural to InNi_2 . $\text{TiCu}_2\text{Sn}(\tau_3)$ was initially reported as having CsCl type structure by [1962Hei], which is the only ternary intermetallic phase included in the [V–C2]. However, [1967Hof] found that $\text{TiCu}_2\text{Sn}(\tau_3)$ has $L2_1\text{-MnCu}_2\text{Al}$ type of structure, and this was confirmed by [1999Kob]. The peritectoid formation of the $\text{TiCu}_2\text{Sn}(\tau_3)$ phase can be attributed to the difficulty to obtain this phase in the experimental work reported by [2001Nak1]. Recently, [2005Hua] reported a new ternary compound in the Cu–Sn–Ti system, $\text{Ti}_3\text{CuSn}(\tau_4)$. The alloy, Cu–23Ti–17Sn (at.%), was prepared by mixing and milling the appropriate quantity of Cu powder, Sn powder and TiH_2 powder. The samples were annealed at 900°C for 10 h. It was found that $\text{Ti}_3\text{CuSn}(\tau_4)$ is thermodynamically stable at 900°C and its crystal structure is of the Ni_3Sn_2 type (space group of $P6_3/mmc$). The type of reactions by which the τ_3 and τ_4 phases form is unknown and suggested for future studies.

The most conflicting data about ternary intermetallic phases concern the $\text{Ti}_5\text{CuSn}_3(\tau_1)$ phase. Based on XRD [1999Kob] determined the change in unit cell volume of $\text{Ti}_5\text{Cu}_x\text{Sn}_3$ as a function of the mole fraction of Cu. [1999Kob] concluded that $\text{Ti}_5\text{Cu}_x\text{Sn}_3$ is an extended solid solution of Cu in Ti_5Sn_3 and it is isostructural to Mn_5Si_3 . However, [2000Sch] reported that the Ti_5CuSn_3 phase is a ternary intermetallic phase, crystallizing with a space group of $P6_3/mcm$ and the structure that can be described either as a filled $D8_8\text{-Mn}_5\text{Si}_3$ type or as a Ti_5Ga_4 type. At the same time, [2000Wei] confirmed that Ti_5CuSn_3 is a ternary stoichiometric compound by full profile Rietveld analysis of the X-ray powder diffractogram. [2001Ham, 2005Hua] also found that Ti_5CuSn_3 is related to the Ti_5Ga_4 type structure and that it has only a narrow homogeneity range. Therefore, Ti_5CuSn_3 is treated as a ternary stoichiometric compound in this assessment.

Quasibinary Systems

[1999Ham] proposed a quasibinary section between (Cu) and the Ti_5CuSn_3 compound in this system, which was confirmed by [2001Nak1]. Quasibinary eutectic involving (Cu) and Ti_5CuSn_3 is located at 878°C . The eutectic liquid composition is close to Cu-9Sn-15Ti (at.%). The diagram is given in Fig. 1 according to the experimental data of [1999Ham, 2001Nak1].

Invariant Equilibria

The data concerning invariant equilibria are given in Table 3. They are mainly based on the experimental data of [2001Nak1]. A partial reaction scheme is given in Fig. 2, adapted from [2001Nak1]. Due to the limited experimental data, only the phase reactions involving the liquid phases are included. Ti_2Sn_3 was also included with appropriate modification to the eutectic reaction E_2 .

Liquidus Surface

The projection of the liquidus surface, Fig. 3, is taken from [2001Nak1] with modifications applied to the binary edges and to some of the invariant reactions according to the critically assessed binary systems. However, the liquidus surface (Fig. 3) and the reaction scheme (Fig. 2) proposed by [2001Nak1] do not show equilibrium of the τ_4 phase with the liquid phase. Such an equilibrium was proposed by [2005Hua] at 900°C , but the nature of the phase formation is yet unknown. This reveals an inconsistency which requires further experimental investigations. Due to the highly reactive nature of the element Ti, it is difficult to avoid oxidation and side reactions between sample and crucible, especially at high temperatures completely. To reduce possible reactions between the sample and the crucible [2001Nak1] employed differential scanning calorimetry with a heating rate of $10^\circ\text{C}\cdot\text{min}^{-1}$ when they recorded the phase transitions. Also care has been taken the measurements of the liquidus and/or solidus of Cu–Sn–Ti system may not be very accurate.

Isothermal Sections

The isothermal sections at 400 and 900°C are given in the Figs. 4 and 5, respectively. These were constructed on the basis of the experimental data obtained by [1999Kob, 2001Ham, 2005Hua] and amended to agree with the accepted binary systems and stability regimes of the ternary intermetallic phases. According to [2005Hua] τ_4 exists in equilibrium with the liquid phase, however this does not agree with the earlier investigations of the liquidus surface [2001Nak1], therefore the three-phase equilibrium $\text{Ti}_3\text{CuSn}+\text{Ti}_5\text{CuSn}_3+\text{L}$ shown by dashed lines in Fig. 5 seems to be questionable. The position of this three-phase region at 900°C was refined based on the data of [2005Hua] and the proposed quasibinary system (Cu)– τ_1 .

Notes on Materials Properties and Applications

Alloys of the Cu–Sn–Ti system belong to the group of high strength age hardening alloys [1961Saa, 1987Kiz]. In recent years, Cu–Sn–Ti alloys are widely used as brazing filler metals for diamond abrasive

tools, because Cu-Sn-Ti alloys wet diamond well because of their high adhesion activity with regard to diamond; they produce strong joints of diamond tools, exhibit a low melting temperature, and have a satisfactory wear resistance [1975Kiz, 1999Ham, 2001Nak2, 2004Kha]. These are some reasons for the increasing interest in Cu-Sn-Ti alloys.

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Table 1: Investigations of the Cu-Sn-Ti Phase Relations and Structures

Reference	Method/Experimental Technique	Temperature/Composition/Phase Range Studied
[1975Kiz]	EPMA ¹ , XRD ²	0 to 30 mass% Sn, 0-20 mass% Ti
[1999Kob]	XRD, electrical resistivity	397°C, 123 samples in whole composition region
[1999Ham]	XRD, electron microscopy	600 to 950°C, quasibinary between (Cu) and Ti ₅ CuSn ₃
[2000Sch]	XRD, EPMA	800°C, 10Cu-30Sn-60Ti (mass%), 11.1Cu-33.3Sn-66.7 Ti (mass%), and 15Cu-30Sn-55Ti (mass%), crystal structure of Ti ₅ CuSn ₃
[2000Wei]	Full profile analysis of the XRD	45Cu-30Sn-25Ti (mass%), TiCuSn
[2001Nak1]	EPMA, XRD, DTA/DSC	800°C, 7.5 to 60 at.% Sn, 5 to 90 at.%Ti liquidus surface
[2001Nak2]	EPMA, XRD, DTA/DSC ³	Cu, 15 at.% Sn, 0 to 25 at.% Ti, liquidus and solidus
[2001Ham]	EPMA, XRD	900°C, composition range: Cu-Ti ₃ Sn and Cu-Ti ₆ Sn ₅
[2002She]	Crystal structure, XRD	800°C, TiCuSn
[2005Hua]	EPMA, XRD, TEM ⁴	900°C, Cu-17 at.%Sn-23 at.%Ti, Ti ₃ CuSn

¹ EPMA: Electron Probe Micro Analysis;² XRD: X-ray diffraction;³ DTA/DSC: Differential Thermal Analysis and Differential Scanning Calorimetry.⁴ TEM: Transmission Electron Microscopy**Table 2:** Crystallographic Data of Solid Phases

Phase/ Temperature Range [°C]	Pearson Symbol/ Space Group/ Prototype	Lattice Parameters [pm]	Comments/References
(Cu) < 1084.62	<i>cF4</i> <i>Fm$\bar{3}m$</i> Cu	<i>a</i> = 361.46	at 25°C [Mas2] melting point [1994Sub]
(βSn) 231.9681 - 13	<i>tI4</i> <i>I4₁/amd</i> βSn	<i>a</i> = 583.18 <i>c</i> = 318.18	at 25°C [Mas2]
(αSn) < 13	<i>cF8</i> <i>Fd$\bar{3}m$</i> C (diamond)	<i>a</i> = 648.92	[Mas2]
(βTi) 1670 - 882	<i>cI2</i> <i>Im$\bar{3}m$</i> W	<i>a</i> = 330.65	[Mas2]

Phase/ Temperature Range [°C]	Pearson Symbol/ Space Group/ Prototype	Lattice Parameters [pm]	Comments/References
(α Ti) < 882	<i>hP2</i> <i>P6₃mmc</i> Mg	$a = 295.06$ $c = 468.35$	at 25°C [Mas2]
β (CuSn)	<i>cI2</i> <i>Im$\bar{3}m$</i> W	$a = 302.6$	[2005Fra, Mas2]
γ (CuSn) 757 - 520	<i>cF16</i> <i>Fm$\bar{3}m$</i> AlFe ₃	$a = 611.7$	Cu ₃ Sn; [2005Fra, Mas2, V-C2]
Cu ₄₁ Sn ₁₁ 590 - 350	<i>cF416</i> <i>F$\bar{4}3m$</i> Cu ₄₁ Sn ₁₁	$a = 1798.0$	[2005Fra, Mas2, V-C2]
Cu ₁₀ Sn ₃ 640 - 582	<i>hP26</i> <i>P6₃</i> Cu ₁₀ Sn ₃	$a = 733.0$ $c = 786.4$	[2005Fra, Mas2, V-C2]
Cu ₃ Sn < 678	<i>oC80</i> <i>Cmcm</i> Cu ₃ Sn	$a = 552.9$ $b = 4775.6$ $c = 432.3$	[2005Fra, Mas2, V-C2]
η Cu ₆ Sn ₅ 416 - 198	<i>hP4</i> <i>P6₃/mmc</i> NiAs	$a = 419.2$ $c = 503.7$	[2005Fra, Mas2, V-C2]
η' Cu ₆ Sn ₅ < 198	<i>hR22</i>	$a = 864.8$ $c = 1067.5$	[2005Fra, V-C]
Ti ₂ Cu < 1012	<i>tI6</i> <i>I4/mmm</i> MoSi ₂	$a = 295.3$ $c = 1073.4$	[Mas2, V-C2, 1994Ali]
TiCu < 982	<i>tP4</i> <i>P4/nmm</i> TiCu	$a = 310.8$ to 311.8 $c = 588.7$ to 592.1	48 to 52 at.% Cu [Mas2, V-C2]
Ti ₃ Cu ₄ < 925	<i>tI14</i> <i>I4/mmm</i> Ti ₃ Cu ₄	$a = 313.0$ $c = 1994$	[Mas2, V-C2]
Ti ₂ Cu ₃ < 875	<i>tP10</i> <i>P4/nmm</i> Ti ₂ Cu ₃	$a = 313$ $c = 1395$	[Mas2, V-C2]
TiCu ₂ 890 - 870	<i>oC12</i> <i>Amm2</i> VAu ₂	$a = 436.3$ $b = 797.7$ $c = 447.8$	[Mas2, V-C2]
β TiCu ₄ 885 - ~400	<i>oP20</i> <i>Pnma</i> ZrAu ₄	$a = 452.5$ $b = 434.1$ $c = 1295.3$	~78 to ~80 at.% Cu [Mas2, V-C2]

Phase/ Temperature Range [°C]	Pearson Symbol/ Space Group/ Prototype	Lattice Parameters [pm]	Comments/References
αTiCu_4 ≤ 500	<i>tI10</i> <i>I4/m</i> MoNi ₄	-	~78 to ~80 at.% Cu [Mas2]
Ti ₃ Cu < 905	<i>tP4</i> <i>P4/mmm</i> <i>oP*</i> <i>Pbam</i>	<i>a</i> = 415.8 <i>c</i> = 359.4	[V-C2] [2002Oka]
Ti ₃ Sn < 1676	<i>hP8</i> <i>P6₃/mmc</i> Ni ₃ Sn	<i>a</i> = 591.6 <i>c</i> = 476.4	[Mas2], [2003Obr]
Ti ₂ Sn < 1550	<i>hP6</i> <i>P6₃/mmc</i> InNi ₂	<i>a</i> = 465.3 <i>c</i> = 570	[Mas2], [2003Obr]
Ti ₅ Sn ₃ < 1506	<i>hP16</i> <i>P6₃/mcm</i> Mn ₅ Si ₃	<i>a</i> = 804.9 <i>c</i> = 545.4	[Mas2], [2003Obr]
Ti ₆ Sn ₅ (l) < 686	<i>oI44</i> <i>Immm</i> Nb ₆ Sn ₅	<i>a</i> = 1693 <i>b</i> = 914.4 <i>c</i> = 573.5	[Mas2], [2003Obr]
Ti ₂ Sn ₃ < 767	<i>oC*</i> <i>Cmca</i> V ₂ GaSn ₂ <i>oC*</i> <i>Cmca</i> ?	<i>a</i> = 596.74 <i>b</i> = 1995 <i>c</i> = 701.3 <i>a</i> = 596 <i>b</i> = 1994 <i>c</i> = 702	[2000Kue] [2003Obr]
Ti _{6-x} Cu _x Sn ₅ 1488 - 686	<i>hP22</i> <i>P6₃/mmc</i> Ti ₆ Sn ₅	<i>a</i> = 917.4 to 922.3 <i>c</i> = 563.6 to 569.1	0.0 $\leq x \leq$ 0.1 [1999Kob]
* τ_1 , Ti ₅ CuSn ₃	<i>hP16</i> <i>P6₃/mcm</i> Mn ₅ Si ₃ or Ti ₅ Ga ₄	<i>a</i> = 816.2 <i>c</i> = 557.4	[2000Sch]
* τ_2 , TiCuSn	<i>hP6</i> <i>P6₃mc</i> LiGaGe or <i>hP6</i> <i>P6₃/mmc</i> InNi ₂	<i>a</i> = 439.72 <i>c</i> = 601.68 <i>a</i> = 439.555 <i>c</i> = 601.505	[1999Kob] [2000Wei]

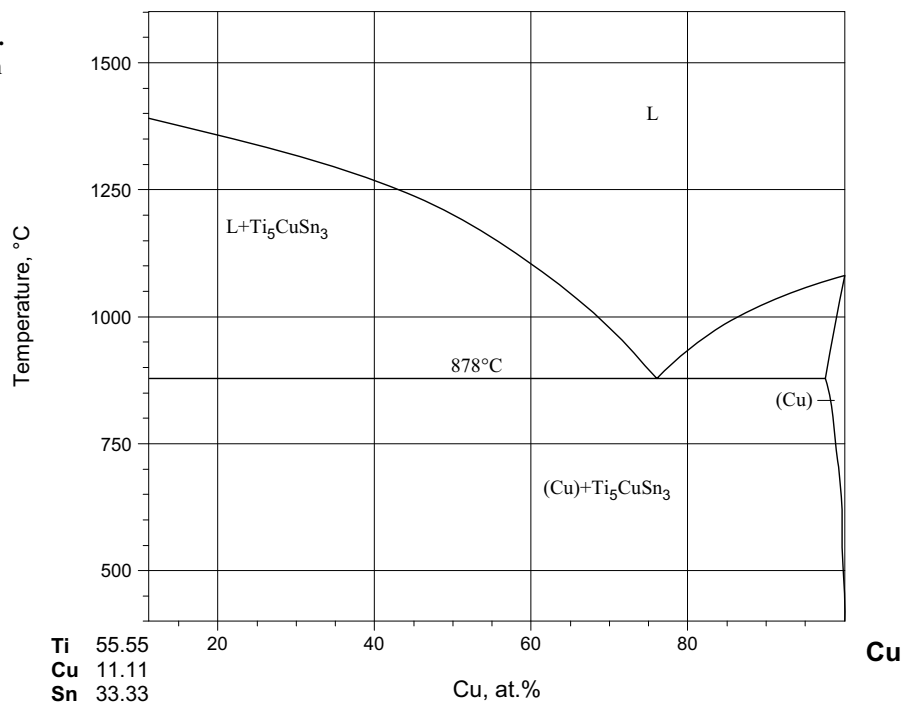
Phase/ Temperature Range [°C]	Pearson Symbol/ Space Group/ Prototype	Lattice Parameters [pm]	Comments/References
* τ_3 , $\text{Ti}_{1-x}\text{Cu}_{2+x}\text{Sn}$	$cP2$ $Pm\bar{3}m$ CsCl or cF^* $Fm\bar{3}m$ MnCu_2Al	$a = 592$ $a = 590.4$ to 592.6 $c = 522.9$	[1962Hei] $0.0 \leq x \leq 0.3$ [1999Kob]
* τ_4 , Ti_3CuSn	$hP6$ $P6_3/mmc$ Ni_3Sn_2	$a = 463.6$ $c = 522.9$	[2005Hua]

Table 3: Invariant Equilibria

Reaction	T [°C]	Type	Phase	Composition (at.%)		
				Cu	Sn	Ti
$\text{L} + \text{Ti}_5\text{Sn}_3 + \text{Ti}_2\text{Sn} \rightleftharpoons \tau_1$	> 1300	P_1	L Ti_5Sn_3 Ti_2Sn τ_1	~11.1 0.0 0.0 11.1	~33.3 37.5 33.3 33.3	~55.6 62.5 66.7 55.6
$\text{L} + \text{Ti}_2\text{Sn} \rightleftharpoons \tau_1 + \text{Ti}_3\text{Sn}$?	U_1	-	-	-	-
$\text{L} + \text{Ti}_5\text{Sn}_3 \rightleftharpoons \tau_1 + \text{Ti}_6\text{Sn}_5$?	U_2	-	-	-	-
$\text{L} + (\beta\text{Ti}) \rightleftharpoons \text{Ti}_3\text{Sn} + \text{Ti}_2\text{Cu}$	< 989	U_3	-	-	-	-
$\text{L} + \text{Ti}_2\text{Cu} \rightleftharpoons \text{Ti}_3\text{Sn} + \text{TiCu}$	959	U_4	-	-	-	-
$\text{L} + \text{Ti}_3\text{Sn} \rightleftharpoons \tau_1 + \text{TiCu}$	859	U_5	-	-	-	-
$\text{L} + \tau_1 + \text{Ti}_6\text{Sn}_5 \rightleftharpoons \tau_2$	827	P_2	-	-	-	-
$\text{L} + \text{TiCu} \rightleftharpoons \tau_1 + \text{Ti}_3\text{Cu}_4$?	U_6	-	-	-	-
$\text{L} + \text{Ti}_3\text{Cu}_4 \rightleftharpoons \tau_1 + \text{TiCu}_2$?	U_7	-	-	-	-
$\text{l} \rightleftharpoons (\text{Cu}) + \tau_1$	878	e_6 (max)	l	76	9	15
$\text{L} + (\text{Cu}) \rightleftharpoons \tau_1 + \text{TiCu}_4$	860	U_8	-	-	-	-
$\text{L} + \tau_1 + \text{Ti}_6\text{Sn}_5 \rightleftharpoons \tau_2$	827	P_2	-	-	-	-
$\text{L} \rightleftharpoons \tau_1 + \text{TiCu}_2 + \text{TiCu}_4$?	E_1	-	-	-	-
$\text{L} + (\text{Cu}) \rightleftharpoons \beta(\text{CuSn}) + \tau_1$	772	U_9	L (Cu) $\beta(\text{CuSn})$ τ_1	77.7 96.7 - 11.1	17.6 1.3 - 33.3	4.7 3.0 - 55.6
$\text{L} + \beta(\text{CuSn}) \rightleftharpoons \tau_1 + \gamma(\text{CuSn})$	748	U_{10}	-	-	-	-
$\text{L} + \tau_1 \rightleftharpoons \gamma(\text{CuSn}) + \tau_2$	732	U_{11}	-	-	-	-
$\text{L} + \gamma(\text{CuSn}) + \tau_2 \rightleftharpoons \text{Cu}_3\text{Sn}$	687	P_3	-	-	-	-

Reaction	T [°C]	Type	Phase	Composition (at.%)		
				Cu	Sn	Ti
$L + \tau_2 \rightleftharpoons \text{Cu}_3\text{Sn} + \text{Ti}_6\text{Sn}_5$	572	U_{12}	-	-	-	-
$L + \text{Cu}_3\text{Sn} \rightleftharpoons \eta\text{Cu}_6\text{Sn}_5 + \text{Ti}_6\text{Sn}_5$	407	U_{13}	-	-	-	-
$L + \text{Ti}_2\text{Sn}_3 \rightleftharpoons \text{Ti}_6\text{Sn}_5 + (\text{Sn})$	230	U_{14}	-	-	-	-
$L \rightleftharpoons \eta\text{Cu}_6\text{Sn}_5 + \text{Ti}_6\text{Sn}_5 + (\text{Sn})$	~226	E_2	-	-	-	-

Fig. 1: Cu–Sn–Ti.
Quasibinary system
Cu–Ti₅CuSn₃(τ_1)



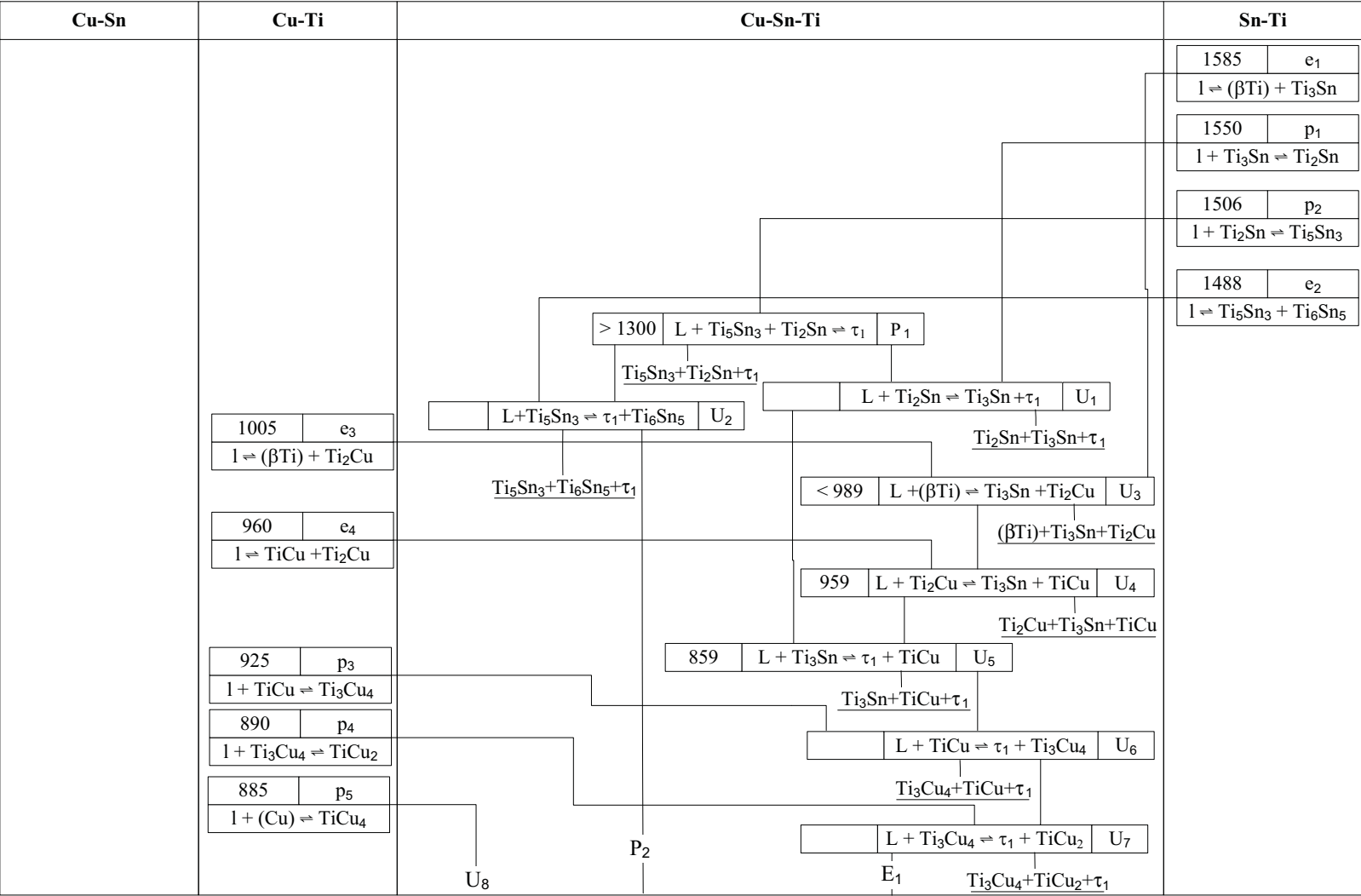


Fig. 2a: Cu-Sn-Ti. Reaction scheme, part 1

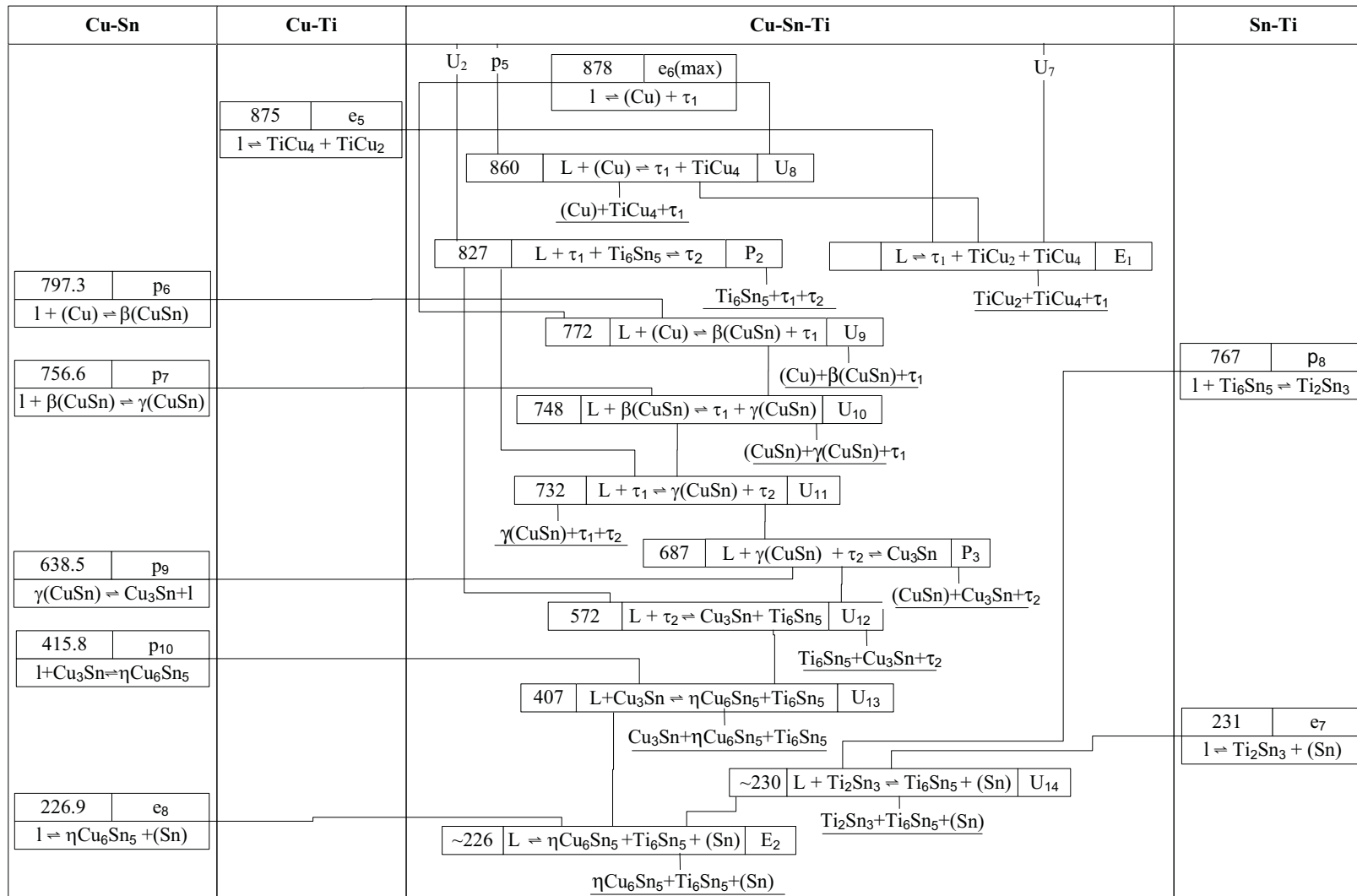


Fig. 2b: Cu-Sn-Ti. Reaction scheme, part 2

Fig. 3: Cu-Sn-Ti.
Liquidus surface
projection

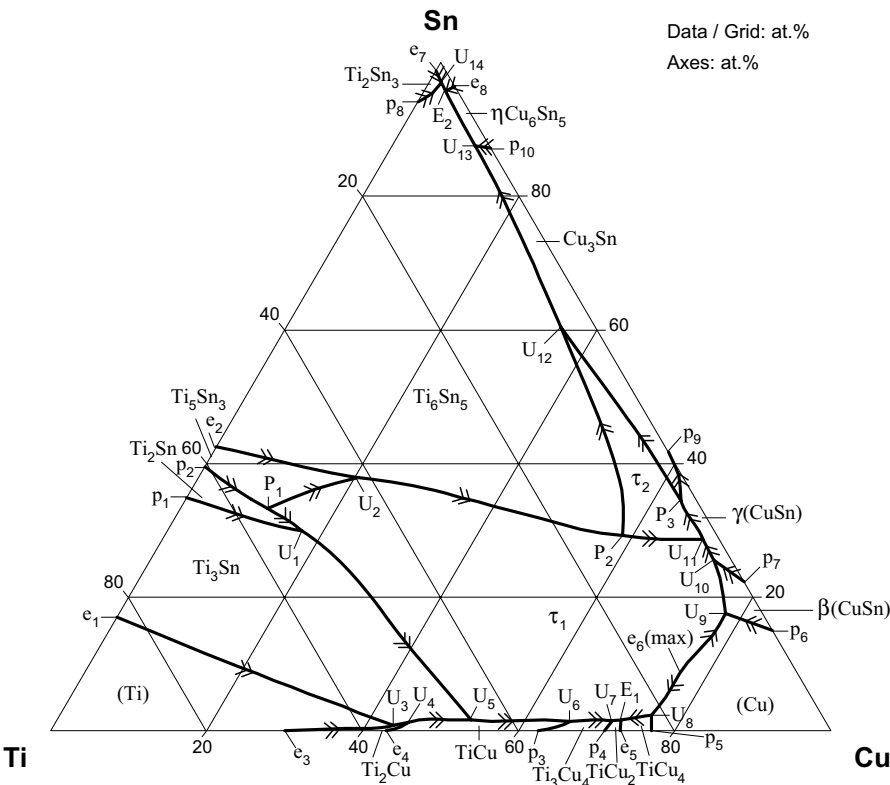
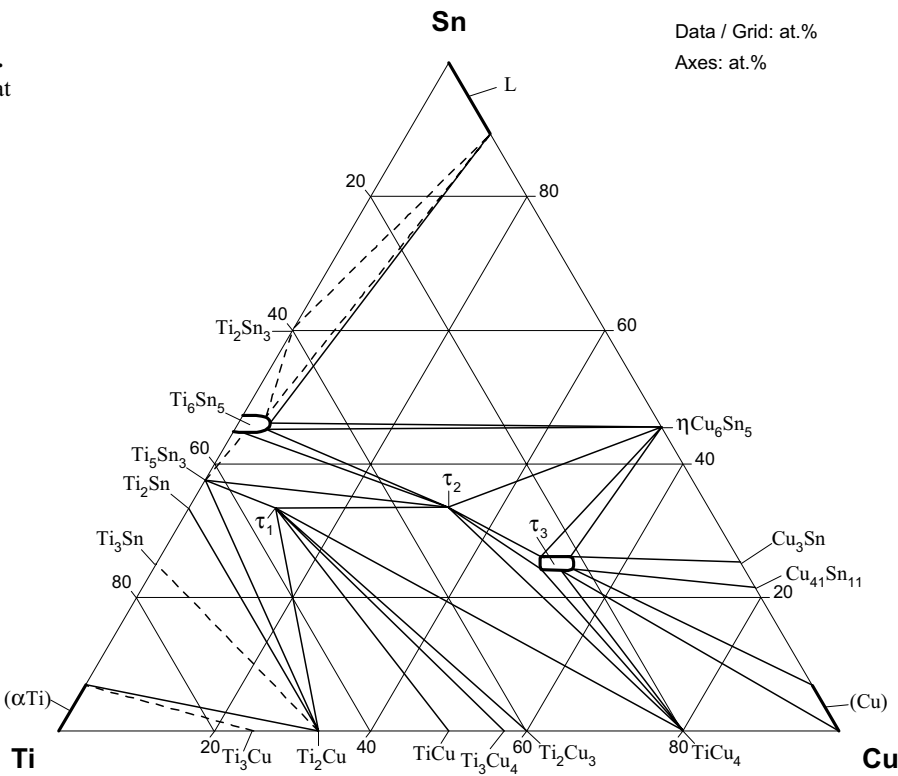


Fig. 4: Cu-Sn-Ti.
Isothermal section at
400°C



Data / Grid: at.%
Axes: at.%

