Silver – Copper – Titanium

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Introduction

The first evaluation within the ongoing MSIT Evaluation Programs was made by [1988Kub], which is updated by the present work. The ternary Ag-Cu-Ti system has been investigated by [1969Ere1, 1969Ere2, 1970Ere1, 1970Ere2] using thermal and metallographic analyses supported by microhardness and X-ray measurements. This is the only systematic experimental study of equilibria in this system up to now. The system was later reassessed by [1977Cha] and reviewed in [1979Dri].
The system is characterized by the existence of a wide miscibility gap in the liquid state, formation of which is in agreement with criteria expressed in [1991But] and thermodynamic calculations of [1995Pau1, 1995Pau2, 1994Gub]. Thermodynamics of Ti oxidation was studied in Ag-Cu-Ti melts. Influence of Ti addition on segregation in Ag rich alloys [1981Duk, 1981Pav] and ageing of Cu rich alloys [1980Bzo] were studied in the ternary Ag-Cu-Ti system. Phase equilibria in the solid state are characterized by the formation of a continuous solid solution Ti_2(Ag,Cu).
The equilibria in the ternary Ag-Cu-Ti system with participation of Ti_2Cu_3 and $TiCu_2$ phases are doubtful due to the discrepancies with regard to the formation and transformation of these phases in the binary Cu-Ti system [2002Ans].

Binary Systems

All three binary systems have been reevaluated recently [2002Rom, 2002Ans, 2002Li] and are accepted here. Respective changes were applied to the here evaluated ternary Ag-Cu-Ti system to fit the phase diagrams of boundary binary systems. Only in the Cu-Ti system uncertainty in the concentration range 55-70 at.% Cu exists. The phase diagram of the binary Cu-Ti system was studied originally by Eremenko *et al.* and refined during the investigation of ternary Ag-Cu-Ti system [1970Ere2]. According to the accepted phase diagram by Eremenko *et al.* and [2002Ans] the Ti_2Cu phase is a congruently melting compound contrary to the peritectic formation assessed in [Mas2]. Such formation of Ti_2Cu leads to the existence of a eutectic in the Ti rich region of the Cu-Ti system. Main discrepancies in the Cu-Ti system refer to the Ti_2Cu_3 phase which according to the accepted phase diagram forms through a peritectoid reaction $Ti_3Cu_4 + TiCu_2 \rightleftharpoons Ti_2Cu_3$ at 875°C and is stable at room temperature, opposite to a peritectic formation $Ti_3Cu_4 + L \rightleftharpoons Ti_2Cu_3$ at 890°C and doubtful eutectoid decomposition at 800°C determined by [1970Ere2]. Such formation of the Ti_2Cu_3 phase affects equilibria of the high-temperature $TiCu_2$ compound which according to the accepted phase diagram forms at higher temperature through a peritectic reaction $Ti_3Cu_4 + L \rightleftharpoons TiCu_2$ at 890°C.

Solid Phases

No ternary compounds form in the Ag-Cu-Ti system. A regular solid solution exists between isotypic Ti_2Ag and Ti_2Cu [1969Ere1, 1970Ere1, 1970Ere2]. The dependence of cell parameters in this solid solution obeys Vegad's law [1970Ere1], see Table 1. Crystallographic data of the phases of the Ag-Ti and Cu-Ti systems are listed in Table 1 (the notation of phases adopted originally by Eremenko *et al.* added).

Invariant Equilibria

Invariant equilibria reported by [1970Ere1, 1970Ere2] were modified to fit the accepted phase transformations in the binary systems and are presented in Table 2 and Figs. 1a and 1b. Parts of the reaction scheme (Figs. 1a, 1b) have not been experimentally observed and therefore the scheme should be considered as tentative.

The composition of liquid was determined for four invariant transformations (Table 2) and critical points of liquid immiscibility in the ternary system K_1 (64Ag-2Cu-34Ti (at.%)) and K_2 (30Ag-61Cu-9Ti (at.%)). Due to a wide immiscibility region in the ternary system most transformations are located close to the boundary of the binary Cu-Ti system in a narrow 5 at.% Ag region (Figs. 2a and 2b) and should be considered approximate. The decomposition of Ti_2Cu_3 phase in ternary system assumed by Eremenko *et al*. to be of the eutectoid type and to take place at 803°C was omitted. The invariant transformation U_6 ($Ti_3Cu_4 + TiCu_2 \rightleftharpoons L + Ti_2Cu_3$) was added to the reaction scheme proposed by [1970Ere2] at 875-851°C (Figs. 1a, 1b, 2a, 2b) in order to be consistent with the accepted binary Cu-Ti system.

Liquidus Surface

The liquidus surface based on thermal, metallographic and X-ray analyses undertaken by [1970Ere1] is presented in Figs. 2a and 2b. It was adjusted to be consistent with the binaries. The liquid immiscibility was determined to exist at temperatures above 850°C in the ranges of compositions 5-94.5 at.% Ag, 2-68 at.% Cu and 1.5-65 at.% Ti [1969Ere2]. In the region of existence of the congruent melting compound TiCu, the liquidus surface is raised with the estimated temperature of the maximum critical points L´, L´´ to be 970°C.

Isothermal Sections

Five isothermal sections at 1300, 1005, 960, 900 and 700°C are studied in the ternary Ag-Cu-Ti system [1969Ere1, 1970Ere2] using X-ray and metallographic analysis. The isothermal section at 1300°C is characterized by a liquid miscibility gap (Fig. 3). The isothermal section at 700°C (Fig. 4) presents the ternary Ag-Cu-Ti system in the solid state. All solid phases except (αTi) are in equilibrium with (Ag) at this temperature. Isotypic Ti_2Cu and Ti_2Ag form a series of solid solutions, but the cross section at 66.7 at.% Ti is not quasibinary due to equilibrium of TiAg and (βTi) at temperatures above 940°C. Crystallographic data of phases in equilibria at 700°C were determined [1970Ere1]. The solubility of a third component in binary phases was determined approximately to be 5 at.% Ag in TiCu, 2 at.% Cu in TiAg, and less than 2 at.% in others [1969Ere1]. The equilibrium of Ti_2Cu_3 phase and (Ag) omitted by [1969Ere1] added in the assessment of [1977Cha] is accepted here as the only probable. The phase boundaries, homogeneity ranges, *etc*. in the isothermal sections of Ag-Cu-Ti system presented in Figs. 3 and 4 were corrected to fit the accepted binary systems.

Temperature – Composition Sections

Three polythermal sections of ternary Ag-Cu-Ti system at 5 at.% Ag, 60 at.% Ag and TiAg-Cu, based on thermal analysis, have been presented by [1970Ere2]. Due to serious discrepancies with the accepted binary systems they are not presented here.

Thermodynamics

[1990Pak] determined the activity of Ti in Ag-Cu-Ti melts at 1000°C by measuring the oxygen potential, p_{O2}, in equilibrium with Ti in the melts and titanium oxide. This titanium oxide phase was determined by X-ray diffraction and was identified as Ti_2O. In order to determine the activity the free energy of formation of this phase had to be estimated, which results in an uncertainty of the activities of a factor 1.8. The results (Table 3) indicate a positive deviation from the ideal solution behavior. Table 4 shows the effect of Ag content on Ti activity in Ag-Cu melts at 1000°C.

The activities of Ti in Ag-28Cu melts at 1000°C also have been measured by an oxygen sensor [2002Ron] and are shown in Table 5. Contrary to the result of [1990Pak], a negative deviation from the ideal solution behavior was found. The difference might be due to the assumption of [1990Pak] that the reaction layer is Ti_2O, while [2002Ron] defines the equilibrium titanium oxide phase as TiO, which is according to the phase diagram. [2002Ron] also measured the effect of silver (Table 6) and copper (Table 7) content on the activity. Ag increases the Ti activity coefficient significantly.

[1994Gub] calculated two isothermal sections at 1300°C and 800°C. The calculation at 1300°C shows a three phase equilibrium between (βTi) and two liquids which is not found in the experimental diagram. The calculation at 800°C is incomplete and lacks several equilibria. [1995Pau1] calculated two isothermal sections at 950 and 1000°C based on the measurements of [1970Ere2] and own optical and SEM/EDX/EPMA observation of diffusion couples of Ti and Ag-Cu (AgCu25%, AgCu40%, AgCu50% and AgCu75% - compositions given in at.%). The calculation is more complete than the one from [1994Gub], but some three-phase equilibria are wrong or lacking at both temperatures. This could be caused by the use of wrong or inaccurate temperatures for the binary reactions. In all calculations, the miscibility gap is smaller than the experimental one, which is probably due to the use of a regular solution model.

Notes on Materials Properties and Applications

Ag-Cu-Ti alloys are used as an interactive brazing material for joining ceramics [1990Pak, 1995Pau2, 1994Gub, 1998Nak].

Miscellaneous

[1981Duk] found that the addition of Ti to Ag-Cu alloys resulted in a non-equilibrium ternary eutectic and the formation of the intermetallic compound $TiCu_4$. [1981Pav] investigated the mechanisms of precipitation in Ag-Cu alloys with small additions of titanium.

A vast amount of research was done on the use of AgCuTi as a brazing material. Several authors, [1992Kur], [1993Sue], [1993Kat], [1995Hon] [1997Hao], [1998Pau], [2001Jan], [2003Shi], [2003Bai], [2004Shi] investigated the reaction mechanism and the interfacial morphology. Others did research on the wetting and spreading behavior [1999Lop], [1999Ich], [2001Jan], [2001Sci], [2001Abe], [2001Pal], [2002Iwa], [2003Nov], [2004Muo], and the oxidation behavior [1999Lee].

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Table 1: Crystallographic Data of Solid Phases

Phase/ Temperature Range [°C]	Pearson Symbol/ Space Group/ Prototype	Lattice Parameters [pm]	Comments/References
(Ag) < 961.93	*cF*4 $Fm\bar{3}m$ Cu	*a* = 408.57	pure Ag at 25°C [Mas2] dissolves 5 at.% Ti at 959°C dissolves 14 at.% Cu at 780°C
(Cu) < 1084.87	*cF*4 $Fm\bar{3}m$ Cu	*a* = 361.46	pure Cu at 25°C [Mas2] dissolves 8 at.% Ti at 885°C dissolves 0.8 at.% Ti at 450°C dissolves 5 at.% Ag at 780°C
(βTi) 1670 - 790	*cI*2 $Im\bar{3}m$ W	*a* = 330.65	pure Ti(h) at 25°C [Mas2] dissolves 13.5 at.% Cu at 1005°C dissolves 15.5 at.% Ag at 1020°C
(αTi) < 882	*hP*2 $P6_3/mmc$ Mg	*a* = 295.06 *c* = 468.35	pure Ti(r) at 25°C [Mas2] dissolves 1.6 at.% Cu at 790°C dissolves 4.7 at.% Ag at 855°C
η, TiAg < 1020	*tP*4 *P*4/*nmm* γTiCu	*a* = 290.3 *c* = 574	48 to 50 at.% Ag [Mas2, V-C2]
δ, TiCu < 982	*tP*4 *P*4/*nmm* γTiCu	*a* = 310.8 to 311.8 *c* = 588.7 to 592.1	48 to 52 at.% Cu [Mas2, V-C2]
ε, Ti_3Cu_4 < 925	*tI*14 *I*4/*mmm* Ti_3Cu_4	*a* = 313.0 *c* = 1994	[Mas2, V-C2]
θ, Ti_2Cu_3 < 875	*tP*10 *P*4/*nmm* Ti_2Cu_3	*a* = 313 *c* = 1395	[Mas2, V-C2]
λ, $TiCu_2$ 890 - 870	*oC*12 *Amm*2 VAu_2	*a* = 436.3 *b* = 797.7 *c* = 447.8	[Mas2, V-C2]
$\beta TiCu_4$ 885 - ~400	*oP*20 *Pnma* $ZrAu_4$	*a* = 452.5 *b* = 434.1 *c* = 1295.3	~78 to ~80.9 at.% Cu [Mas2, V-C2]
$\alpha TiCu_4$ ≲ 500	*tI*10 *I*4/*m* $MoNi_4$		~78 to ~80.9 at.% Cu [Mas2]
$Ti_2Ag_{1-x}Cu_x$	*tI*6 *I*4/*mmm* $MoSi_2$	*a* = 295.5 to 294.3 *c* = 1185 to 1077	*x* = 0 to 1 [1970Ere2] linear dependence
Ti_2Ag < 940		*a* = 295.2 *c* = 1185	[Mas2, V-C2]
Ti_2Cu < 1012		*a* = 295.3 *c* = 1073.4	[Mas2, V-C2]

Table 2: Invariant Equilibria

Reaction	T [°C]	Type	Phase	Composition (at.%)*		
				Ag	Cu	Ti
L_1 + (βTi) ⇌ L_2 + Ti_2(Ag,Cu)	982	U_1	L_1 L_2	13 88	30 10	57 2
L + (βTi) ⇌ Ti_2(Ag,Cu) + TiAg	960	U_2	-	-	-	-
L_3 ⇌ L_4 + Ti_2(Ag,Cu) + TiCu	954	E_1	L_3 L_4	10 84	38 14	52 2
L + TiAg ⇌ Ti_2(Ag,Cu) + (Ag)	929	U_3	-	-	-	-
L + Ti_2(Ag,Cu) ⇌ TiCu + (Ag)	908	U_4	-	-	-	-
L_5 + TiCu ⇌ L_6 + Ti_3Cu_4	900	U_5	L_5 L_6	6 66	61 32	33 2
Ti_3Cu_4 + $TiCu_2$ ⇌ L + Ti_2Cu_3	875-851	U_6	-	-	-	-
L + TiCu ⇌ Ti_3Cu_4 + (Ag)	860	U_7	-	-	-	-
$TiCu_2$ ⇌ L + Ti_2Cu_3 + $TiCu_4$	851	E_2	L	5	72	23
L + Ti_3Cu_4 ⇌ Ti_2Cu_3 + (Ag)	843	U_8	-	-	-	-
L + Ti_2Cu_3 ⇌ $TiCu_4$ + (Ag)	808	U_9	-	-	-	-
L + $TiCu_4$ ⇌ (Cu) + (Ag)	783	U_{10}	-	-	-	-

* - the composition of L estimated from a figure of the liquidus surface [1970Ere1], agrees with those given in mass% in [1977Cha]

Table 3: Activity of Diluted Ti in Eutectic 72Ag-28Cu (mass%) Melts at 1000°C [1990Pak]

x_{Ti} (final mol fraction Ti in the melt)	$a_{Ti} \cdot 10$	$\log\gamma_{Ti}$
0.03087	1.13	0.563
0.03087	1.30	0.624
0.0190	0.753	0.598
0.00863	0.403	0.669
0.00432	0.264	0.786
0.0105	0.468	0.649
0.00414	0.213	0.712
0.0198	0.827	0.621

Table 4: Activity of Diluted Ti in Eutectic 72Ag-28Cu (mass%) Melts at 1000°C [1990Pak]

Alloys (composition in mass%)	x_{Ti} (final mol fraction Ti in the melt)	$a_{Ti} \cdot 10$	$\log\gamma_{Ti}$
Ag-1Ti	0.0016	1.27	1.90
Ag-10Cu-1Ti	0.0067	1.34	1.30
Ag-28Cu-1Ti	0.0105	0.468	0.649

Table 5: Activity of Diluted Ti in Eutectic Ag-Cu (mass%) Melts at 1000°C [2002Ron]

Ti (mass%)	Cu (mass%)	x_{Ti} (final fraction Ti in the melt) $\cdot 10^{-3}$	a_{Ti}	$\ln\gamma_{Ti}$
0.4	28.74	0.577	$4.00 \cdot 10^{-5}$	–2.67
0.6	28.4	1.374	$4.82 \cdot 10^{-4}$	–2.02
0.8	29.15	2.728	$5.09 \cdot 10^{-4}$	–1.68
1.0	29.95	4.64	$1.01 \cdot 10^{-3}$	–1.53
1.5	28.54	9.002	$3.47 \cdot 10^{-3}$	–0.95

Table 6: Effect of Silver Content on the Activity Coefficient of Ti in Eutectic Ag-Cu Melts Containing 1 mass% Titanium at 1000°C [2002Ron]

x_{Ti}	x_{Ag}	$a_{Ti} \cdot 10^{-4}$	γ_{Ti}
0.00142	0.0094	1.51	0.1065
0.00139	0.0198	3.50	0.2516
0.00135	0.0385	6.14	0.4550
0.001295	0.0405	13.1	1.018

Table 7: Effect of Copper Content on the Activity Coefficient of Ti in Eutectic Ag-Cu Melts Containing 1 mass% Titanium at 1000°C [2002Ron]

x_{Ti}	x_{Cu}	$a_{Ti} \cdot 10^{-4}$	γ_{Ti}
0.00126	0.00474	2.692	0.2136
0.00122	0.0219	0.952	0.0781
0.00117	0.0326	0.658	0.0563
0.00115	0.0355	0.379	0.0330

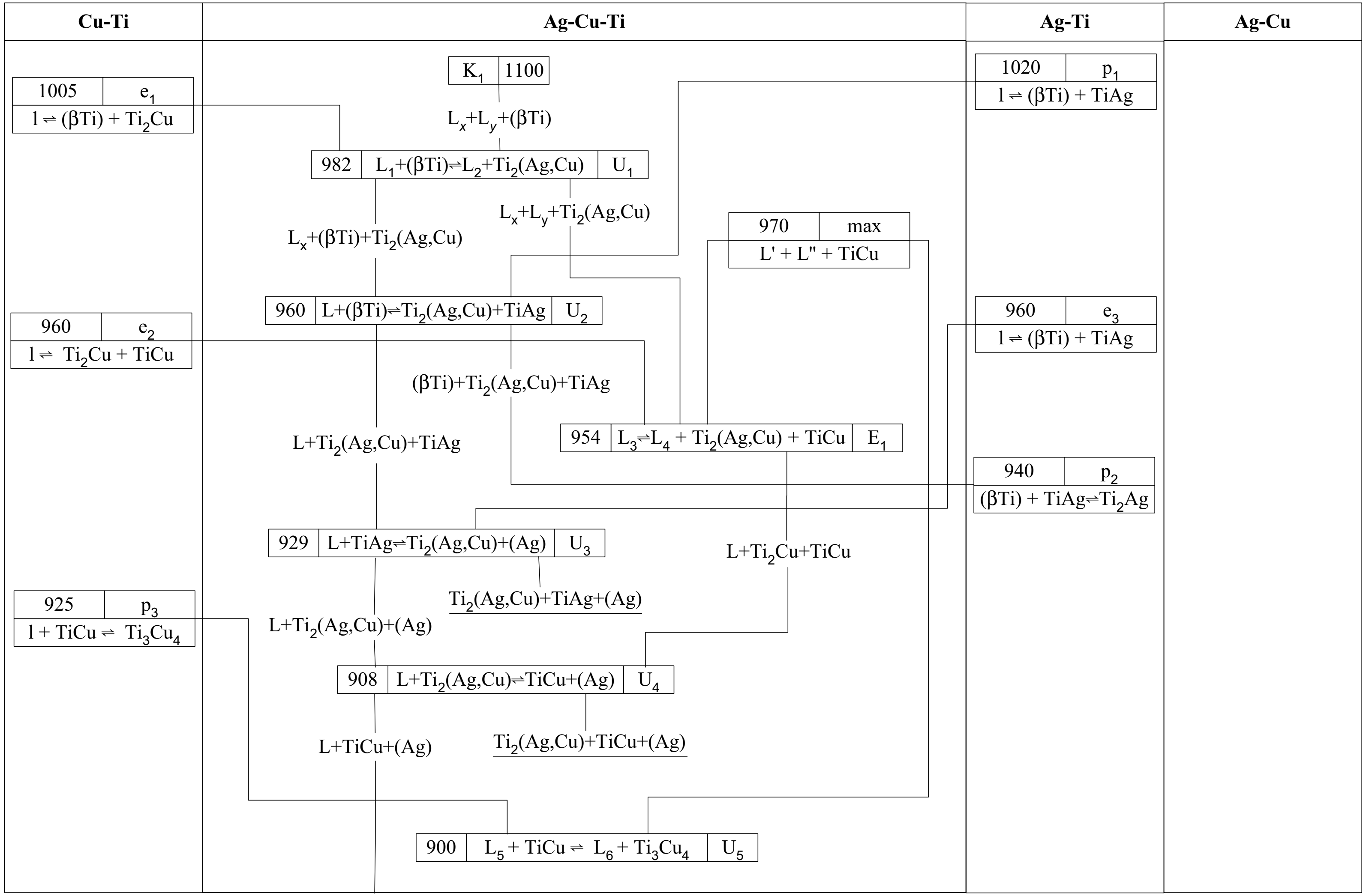

Fig. 1a: Ag-Cu-Ti. Reaction scheme

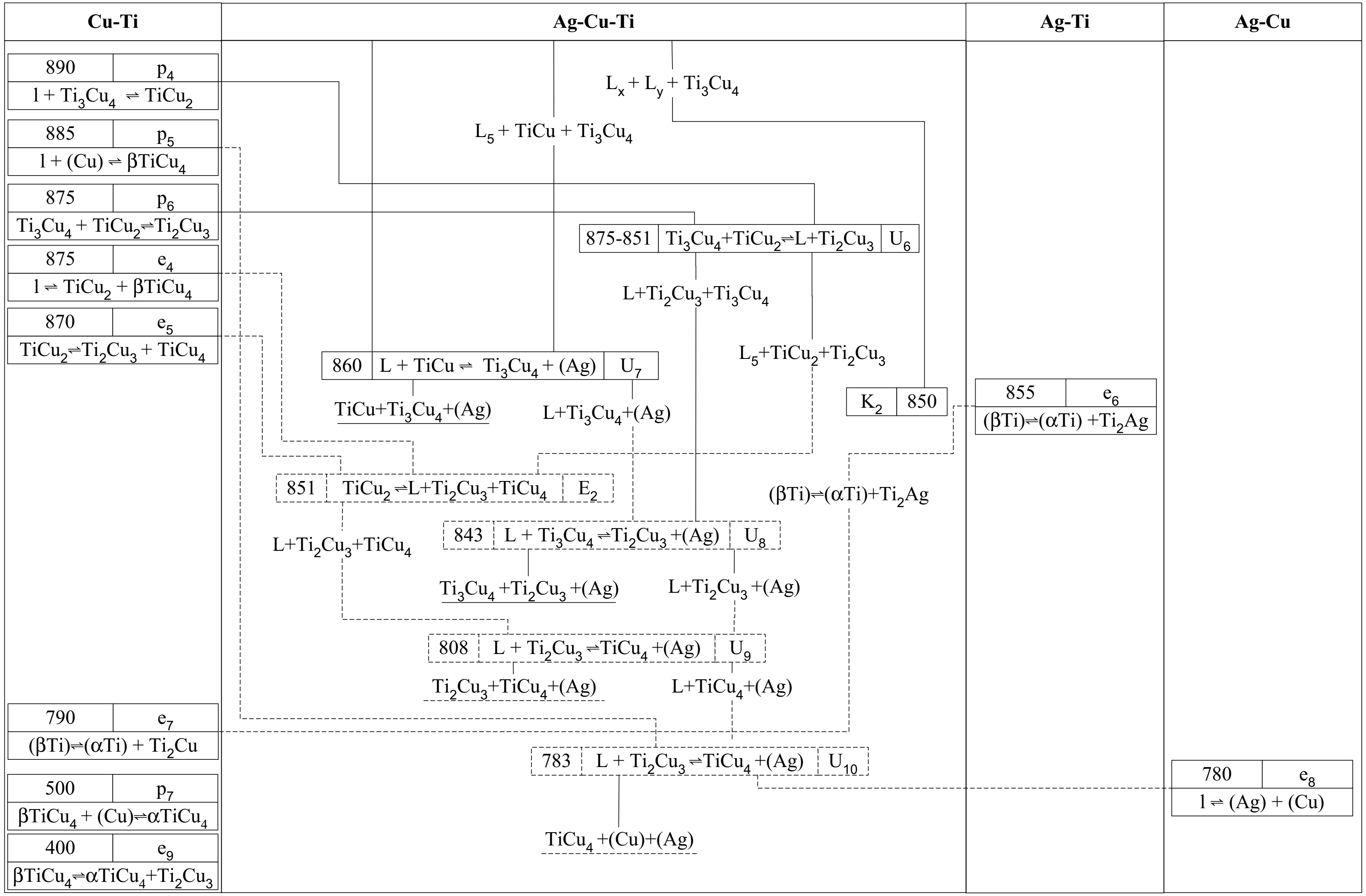

Fig. 1b: Ag-Cu-Ti. Reaction scheme (continued)

Fig. 2a: Ag-Cu-Ti. Liquidus surface projection

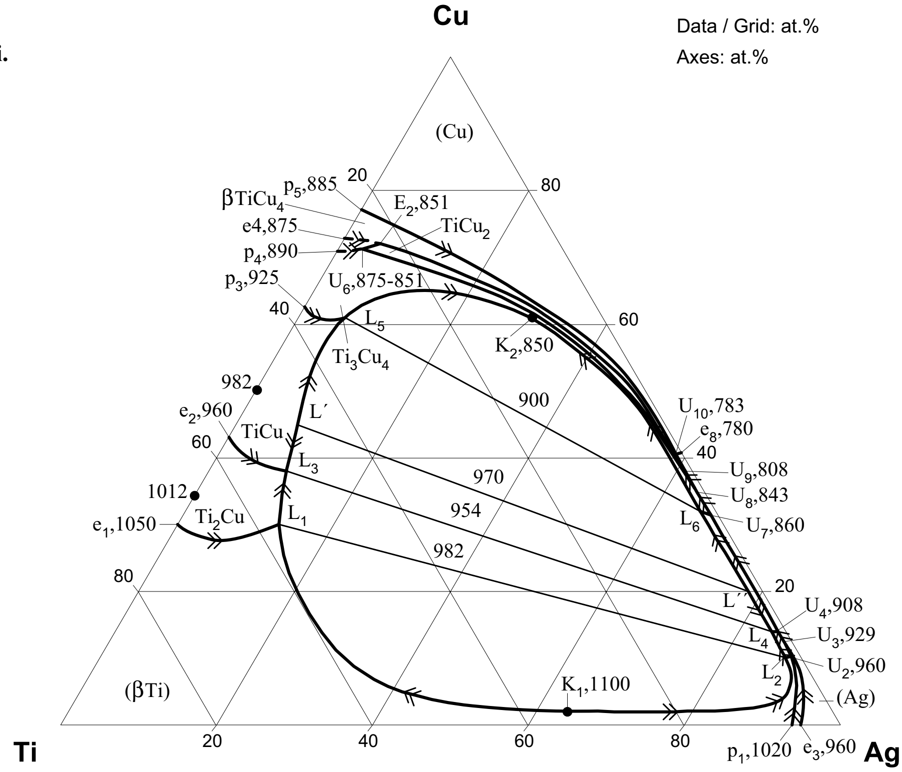

Fig. 2b: Ag-Cu-Ti. Liquidus surface; enlarged view of Fig. 2a in the Ag rich corner

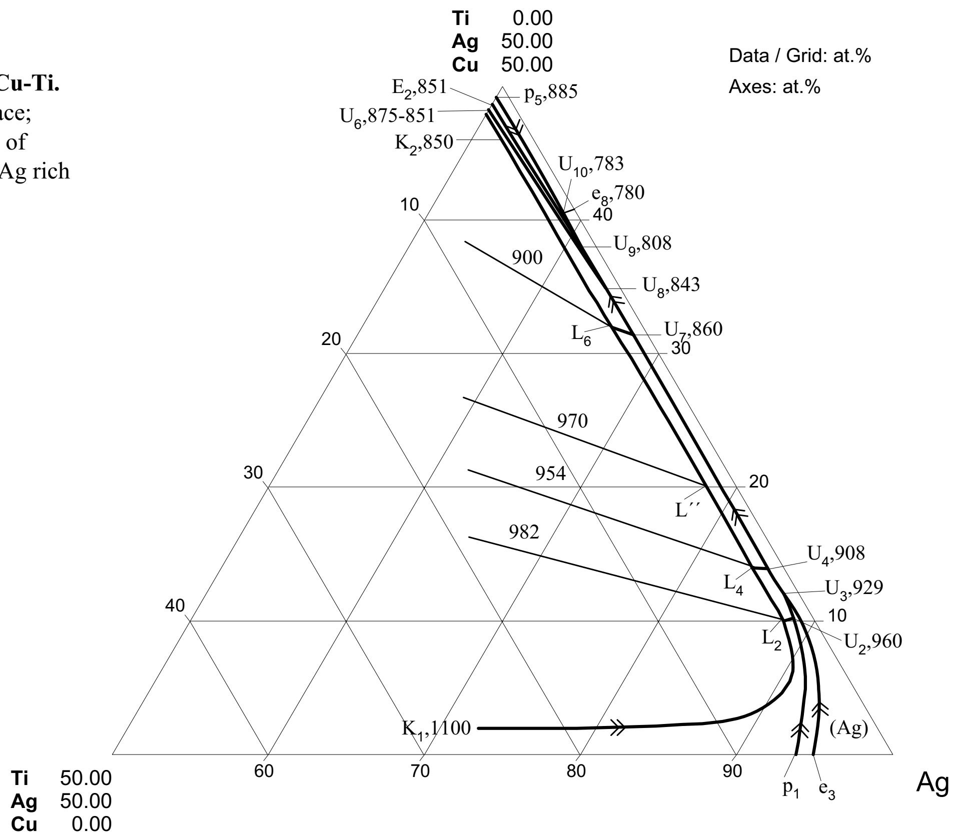

Fig. 3: Ag-Cu-Ti.
Isothermal section
at 1300°C

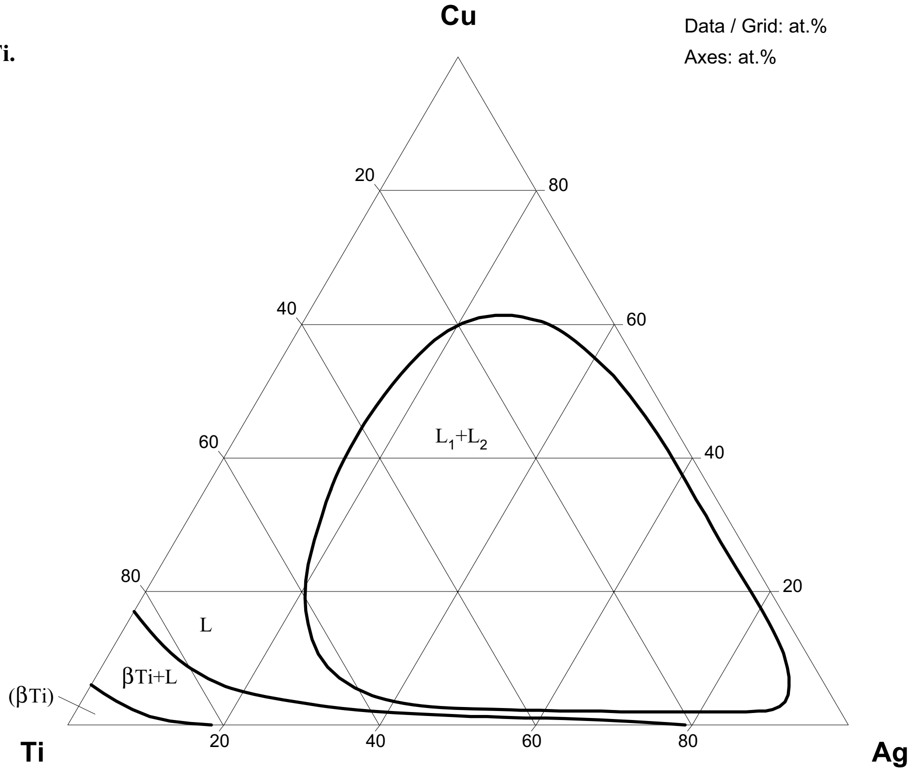

Fig. 4: Ag-Cu-Ti.
Isothermal section
at 700°C