

## Aluminium – Cobalt – Titanium

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### Literature Data

This evaluation updates and modifies in parts the thoroughly made critical evaluation by [1991Sch] in the same MSIT Ternary Evaluation Program. The Al–Co–Ti system was first investigated by [1966Mar], who prepared 110 alloys, melted from iodide titanium (99.97%), cobalt (99.9%) and aluminum (99.997%) under helium in an arc furnace with W-electrode, using a water cooled copper mould. The whole ternary system was determined by thermal, X-ray, and dilatometric analysis together with hardness and specific electrical resistivity measurements (only for Ti-rich alloys). The samples were annealed at 800°C and 600°C for 1 month in evacuated silica ampoules containing Ti-chips. The authors found two ternary phases:  $\text{TiCo}_2\text{Al}$  and another one called  $\text{Ti}_2\text{CoAl}_2$ , reported with a small range of homogeneity. The samples annealed at 600°C had compositions identical to those annealed at 800°C. The phase  $\text{TiCo}_2\text{Al}$  is a Heusler phase [1962Mar, 1963Gla, 1967Hof], the phase  $\text{Ti}_2\text{CoAl}_2$  is of the  $\text{Th}_6\text{Mn}_{23}$  type [1969Mar], the latter paper gives an approximate composition  $\text{TiCoAl}_2$ . A Japanese group [1967Tsu, 1968Tsu] studied the equilibria in the Ti-rich corner (more than 80 mass% Ti) between 1100 and 600°C. Commercially pure Ti-sponge (99.8%), Co (99.54%) and Al (99.99%) were melted in an argon arc furnace. Alloys with more than 85 mass% Ti were hot rolled between 1000 and 800°C to eliminate the as-cast structure. The specimens for microscopic examination were heat treated in argon filled silica capsules and subsequently quenched in water. Annealing times: 8 h at 1100°C, 1 day at 1050°C or 1 week at 1000°C. The specimens quenched from below 950°C were cooled in stages from 1000°C to the annealing temperature, being held for 1 week at 1000°C and then 1 week at 950°C, then 2 weeks at 900°C and 950°C and 1 month at 800, 750, 700, 650 and 600°C. For X-ray diffraction the specimens were heated in vacuum for 1 h to the required temperature and quenched [1967Tsu]. Eleven isothermal sections and nine isopleths of the Ti-rich corner with more than 80 mass% Ti were constructed [1968Tsu]. The paper [1967Tsu] is a short version of [1968Tsu] with five isothermal and four vertical sections. In two further papers [1969Tsu1, 1969Tsu2] the investigations were extended to 70 mass% Al+Co. In [1972Tsu] these two papers are combined in an English translation containing the same diagram and micrographs. From 111 alloys, most of them chemically analyzed, the liquidus surface as well as 6 isothermal sections were constructed, covering the partial system with less than 50 mass% Al and more than 30 mass% Ti. The authors reported a continuous solid solution  $(\text{Ti,Al})\text{Co}$  between  $\text{TiCo}$  and  $\text{AlCo}$  although the reported alloys cover only part of this range. The three phase fields  $\text{L}+(\text{Ti,Al})\text{Co}+\tau_2$  and  $\text{L}+\tau_2+\text{TiAl}$  have maxima with the reactions  $\text{L}+(\text{Ti,Al})\text{Co}\rightleftharpoons\tau_2$  and  $\text{L}\rightleftharpoons\tau_2+\text{TiAl}$  respectively. Phase equilibria in the Al–Ti portion were determined by [2000Kai]. The alloys prepared by arc melting were equilibrated at 1300°C for 1 day, 1200°C for 7 days and 1000°C 7 days in a evacuated quartz tube back-filled with Ar with Ti filings as getters. The equilibrium compositions were determined by EPMA with standard calibration method. The phase equilibria in the Co–Ti portion were investigated by [2001Ish]. Twenty six alloys were prepared with pure elements by arc melting under an Ar atmosphere. The alloys were sealed in a quartz tube with a titanium getter and equilibrated at 1100°C for 7 days, 1000°C for 14 days and 900°C for 21 days. The equilibrium compositions were determined by energy dispersive spectroscopy (EDS) using standard calibration method. The phase  $\tau_2$ , contrary to [1966Mar], is assumed to have a large range of homogeneity by Ti to Al exchange. Six invariant four-phase reactions were found. By X-ray diffraction it is not possible to distinguish between the hexagonal ( $\alpha\text{Ti}$ ) phase and  $\text{Ti}_3\text{Al}$  which is ordered. Therefore no ordering reflections were observed by [1972Tsu]. By thermal analysis, X-ray diffraction reflections and metallography of 16 as-cast alloys [1979Sei] constructed the whole liquidus surface. It was stated that the results of [1967Tsu] and [1972Tsu] were used to construct the phase diagram, but the lines of double saturation in this diagram differ significantly from those of [1972Tsu], although the alloys reported are not at all sufficient to prove these differences. The Al-rich part was not assessed but estimated to be similar to the Al–Fe–Ti system, assessed in the same paper and to match the known binary systems. Three ternary phases  $\tau_1\text{-TiCo}_2\text{Al}$ ,  $\tau_2\text{-Ti}_{1-x}\text{CoAl}_{2-x}$  and  $\tau_3\text{-Ti}_8\text{Al}_{22}\text{Co}_3$  were found by [1979Sei].

She indicated a field of primary crystallization of the Heusler-*cF16* type phase  $\text{TiCo}_2\text{Al}$ , which implies that there is no continuous solid solution between  $\text{CoAl}$  and  $\text{TiCo}$  in contradiction to [1969Tsu1, 1969Tsu2, 1972Tsu] and [1986Zas]. Several works [1962Mar, 1963Gla, 1967Mar, 1967Hof, 1973Web, 1984End, 1993Nak] contributed to the crystal structure and the lattice parameter of the Heusler phase  $\text{TiCo}_2\text{Al}$ . The phase equilibria in the  $\text{TiCo-TiCo}_2\text{Al-CoAl}$  pseudo-binary section ( $\text{Co} = 50 \text{ at.}\%$ ) were determined by [2002Ish1] using the diffusion couple method. The  $\text{TiCo/TiCo}_2\text{Al}$  and  $\text{TiCo}_2\text{Al/CoAl}$  couples were equilibrated for 2 days at  $1300^\circ\text{C}$ , 14 days for  $1200^\circ\text{C}$  and 21 days at  $1100^\circ\text{C}$ . From the concentration-penetration profiles obtained by EDS analysis, it was confirmed in the  $\text{TiCo-CoAl}$  pseudo-binary section that a continuous ordering from the  $\text{CsCl (B2)}$  type to Heusler ( $\text{L2}_1$ ) phase exist on both the  $\text{TiCo}$  and  $\text{CoAl}$  sides. The phase equilibria in the  $\text{TiCo-TiCo}_2\text{Al}$  section ( $\text{Co} = 52 \text{ at.}\%$ ) were also investigated by [2003Kaw] correlating microstructures and mechanical properties. Their alloys were prepared by arc melting and homogenized in a vacuum at  $1200^\circ\text{C}$  for 2 days, then annealed at  $700 - \sim 900^\circ\text{C}$ . Transmission electron microscopic observation was carried out to detect the anti-phase domain structure introduced during the ordering reaction in the as-quenched alloys. These studies also confirmed the continuous ordering reaction in the  $\text{TiCo-TiCo}_2\text{Al}$  section.

A detailed refinement of the crystal structure of the  $\tau_2$  phase has been performed by [2003Gry] employing X-ray single crystal- and neutron powder diffraction as well as electron diffraction.

### Binary Systems

The binary systems  $\text{Al-Co}$  and  $\text{Co-Ti}$  compiled by [Mas] are used as boundary systems. The  $\text{Al-Ti}$  system [1989Pri] is based on the critical assessment of Murray [1987Mur], but corrected with the results [1989McC] for the range of 40 to 55 at.%  $\text{Al}$  and [1989Kal] for the range of 65 to 75 at.%  $\text{Al}$ . This phase diagram is shown in Fig. 1.

### Pseudobinary Systems

The  $\text{TiCo-CoAl}$  section is reported by [2002Ish1] and [2003Kaw] to be a pseudobinary one and shown in Figs. 12 and 13 by dashed lines.

### Solid Phases

The reported binary phases and the ternary phases are represented in Table 1. The solid solubility of  $\text{Co}$  in ( $\text{Ti}$ ) and  $\text{Ti}_3\text{Al}$  is less than 5 at.%  $\text{Co}$  at  $800^\circ\text{C}$  [1966Mar]. There are conflicting assumptions on the solid solution between  $\text{TiCo}$  and  $\text{CoAl}$  and the observation of the ternary Heusler phase  $\text{TiCo}_2\text{Al}$ , which is an ordered form of the  $\text{CsCl}$ -solid solution " $\text{Co(Ti,Al)}$ ". Complete solid solubility is claimed by [1972Tsu], although the experimental points cover only the  $\text{Co-rich}$  part. An enthalpy vs concentration curve of the  $\text{CoAl-TiCo}$  section which shows no interruption by a two phase field was reported by [1986Zas]. Since the Heusler phase is a superstructure of the  $\text{CsCl}$  type, the distinction between both phases was possibly not well established. The ternary phase  $\tau_3$  [1979Sei] was not found by [1966Mar] and is outside the ranges investigated by other authors. But since similar phases exist in  $\text{Al-Ni-Ti}$  and  $\text{Al-Cu-Ti}$  [1965Ram], its existence is very probable. The solvus of the ( $\beta\text{Ti}$ ) phase was determined by [1967Tsu], it is shown in Fig. 2. The refinement of the crystal structure of the  $\tau_2$  phase gives as formula:  $\text{Ti}_{27.5}\text{Co}_{23.4}\text{Al}_{49.1}$ , its structure type has been determined as  $\text{Mg}_6\text{Cu}_{16}\text{Si}_7$ , a filled variant of the  $\text{Th}_6\text{Mn}_{23}$  type.

### Invariant Equilibria

The partial reaction scheme after [1972Tsu], corrected to the accepted binaries, is given in Fig. 3. A reaction scheme given by [1979Sei] is partially in contradiction to that of [1972Tsu] and to the accepted  $\text{Al-Ti}$  binary system. In the reaction scheme in Fig. 3 a continuous solid solution ( $\text{Ti,Al}$ ) $\text{Co}$  is assumed, from which the Heusler phase  $\text{TiCo}_2\text{Al}$  may form at lower temperatures.

### Liquidus Surface

The liquidus surfaces given by [1972Tsu] and [1979Sei] disagree in many details. That of [1972Tsu] is based on many more alloys and therefore is preferred in the construction of the liquidus surface in Fig. 4. The remaining parts given by [1979Sei] are based on so few alloys that they can be taken only as very tentative. Furthermore the phases  $\text{TiCo}_3$  and the two different modifications of  $\text{TiCo}_2$  are neglected in [1979Sei].

### Isothermal Sections

The isothermal sections of 1300, 1200, 1100, 1000, 900, 800 and 600°C in the Ti-rich part by [1972Tsu], the Al–Ti corner by [2000Kai] and the Co–Ti by [2001Ish] are integrated in Figs. 5, 6, 7, 8, 9, 10 and 11, respectively. Because of the discrepancy between [1967Tsu, 1972Tsu, 1979Sei] and [1966Mar], the isothermal section of [1966Mar] at 800°C is not shown. For the same reason, the isothermal section at 1000°C by [1972Tsu] was replaced by one constructed on the basis of recent data [2000Kai, 2001Ish].

### Temperature – Composition Sections

Nine vertical sections were constructed by [1968Tsu] in the Ti-corner.

### Thermodynamics

Enthalpies of formation by solution calorimetry in liquid Al for five alloys of the section  $\text{Ti}_{1-x}\text{CoAl}_x$  were determined by [1986Zas]. The entropies of alloys of the  $\text{Ti}_{1-x}\text{CoAl}_x$  section were determined by [1987Kra] using low temperature (78 to 273 K) heat capacity measurements.

### Notes on Materials Properties and Applications

Magnetic measurements on the Heusler phase  $\text{TiCo}_2\text{Al}$  were made by [1973Web, 1983Bus, 1984End]. For alloy concentrations of  $\text{Ti}_{1-x}\text{CoAl}_x$  with  $x > 0.6$  the ferromagnetic behavior changes to paramagnetic. The Curie temperature for the  $\text{TiCo}_2\text{Al}$  compound is determined as 134 K in [1983Bus] on a sample annealed at 527°C for 14 days.

The characteristic of the electrical resistivity of the  $\text{Ti}_{32}\text{Co}_{22}\text{Al}_{46}$  compound is typical metallic and the temperature dependence follows the Bloch–Grüneisen relation with a Debye temperature of ~300 K [2003Gry]. The residual resistivity for the  $\text{Ti}_{32}\text{Co}_{22}\text{Al}_{46}$  compound is 0.97 mΩ·cm [2003Gry].

The  $\text{Ti}_{47}\text{Co}_{28}\text{Al}_{25}$  compound can absorb up to 0.8 wt.% hydrogen (8.57 mg) [2003Gry].

### Miscellaneous

The stability of the Heusler phase  $\text{TiCo}_2\text{Al}$  up to the liquidus surface (1750°C) is described by [1979Sei]. The phase stability of the  $\text{L}_{21}$  phase in the TiCo– $\text{TiCo}_2\text{Al}$ –CoAl quasibinary section was reported by [2002Ish2]. There, the (metastable) critical temperature of  $\text{B}_2/\text{L}_{21}$  order-disorder transition of stoichiometric  $\text{TiCo}_2\text{Al}$  was given as 1827°C and the two tri-critical temperatures of the  $\text{B}_2+\text{L}_{21}$  decompositions were estimated as about 1127°C. The critical compositions of the continuous ordering evaluated by [2002Ish2] are shown in Fig. 12 superimposed on the stable melting equilibria (dashed lines). Also shown are the assumed limits of both  $\text{B}_2+\text{L}_{21}$  two-phase fields [2002Ish2]. [2003Kaw] describes the ordering temperature vs composition for continuous ordering on the TiCo rich side of the section at 52 at.% Co from ~1300 down to 600°C but does not recognize the separation into two phases  $\text{B}_2+\text{L}_{21}$ , see Fig. 13.

### Note added in press

Phase equilibria at 950°C has been reported by [2000Din] based on EPMA, quantitative X-ray diffraction and optical microscopy data for arc-melted samples annealed at 950°C for 240 hours. Note: In this work the existence of the  $\tau_3$  phase has been confirmed in as-cast and annealed samples.

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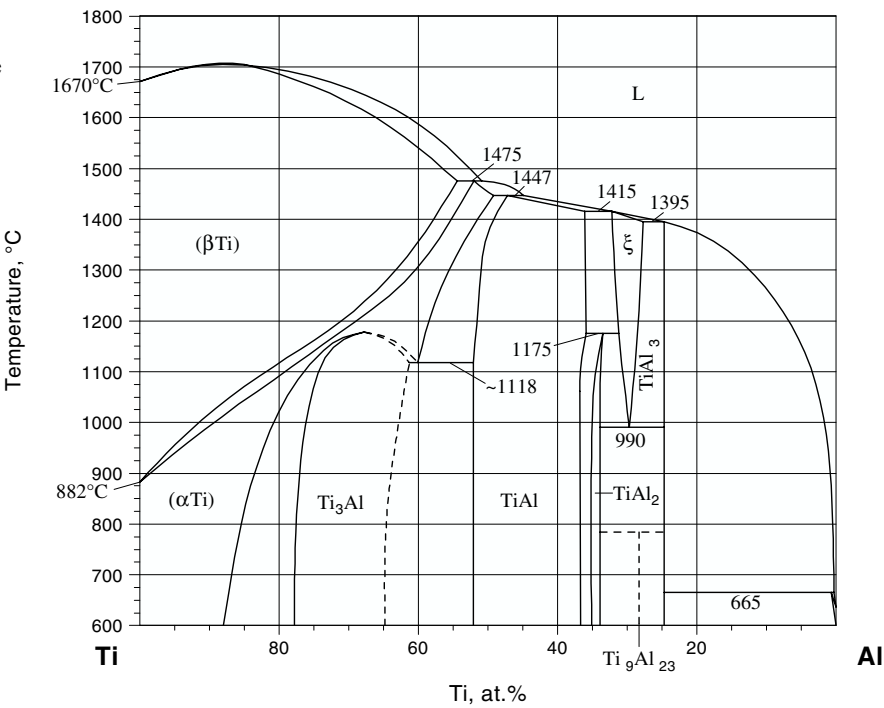
**Table 1:** Crystallographic Data of Solid Phases

Phase / Temperature Range [°C]	Pearson Symbol/ Space Group/ Prototype	Lattice Parameters [pm]	Comments/References
(Al) < 660	<i>cF4</i> <i>Fm<math>\bar{3}m</math></i> Cu	$a = 404.88$	24°C [V-C]
( $\gamma$ Co)(h) 1495-422	<i>cF4</i> <i>Fm<math>\bar{3}m</math></i> Cu	$a = 354.46$	[V-C]
( $\epsilon$ Co)(r) < 422	<i>hP2</i> <i>P6<math>_3</math>/mmc</i> Mg	$a = 250.71$ $c = 406.95$	[V-C]
( $\beta$ Ti)(h) 1670-882	<i>cI2</i> <i>Im<math>\bar{3}m</math></i> W	$a = 330.65$	[V-C]
( $\alpha$ Ti)(r) < 882	<i>hP2</i> <i>P6<math>_3</math>/mmc</i> Mg	$a = 295.08$ $c = 468.55$	[V-C]
Ti <sub>2</sub> Co < 1058	<i>cF96</i> <i>Fd<math>\bar{3}m</math></i> Ti <sub>2</sub> Ni	$a = 1130$	[V-C]
TiCo <sub>2</sub> (c) < 1235	<i>cF24</i> <sup>a)</sup> <i>Fd<math>\bar{3}m</math></i> MgCu <sub>2</sub>	$a = 669.2$	[V-C], homogeneity range 66.5 to 67 at.% Co [Mas]
TiCo <sub>2</sub> (h)	<i>hP24</i> <sup>a)</sup> <i>P6<math>_3</math>/mmc</i> MgNi <sub>2</sub>	$a = 473$ $c = 1541$	[V-C], homogeneity range 68.75 to 72 at.% Co [Mas]
TiCo <sub>3</sub> $\leq 1190$	<i>cP4</i> <i>Pm<math>\bar{3}m</math></i> CuAu <sub>3</sub>	$a = 361.4$	[V-C]
Co <sub>2</sub> Al <sub>5</sub> < 1172	<i>hP28</i> <i>P6<math>_3</math>/mmc</i> Co <sub>2</sub> Al <sub>5</sub>	$a = 767.15$ $c = 760.85$	[V-C]
Co <sub>4</sub> Al <sub>13</sub> $\leq 1100$	<i>mC100</i> <i>Cm</i> Co <sub>4</sub> Al <sub>13</sub>	$a = 1518.3$ $b = 812.2$ $c = 1234.0$ $\beta = 107.9^\circ$	[V-C]
Co <sub>2</sub> Al <sub>9</sub> < 944	<i>mP22</i> <i>P2<math>_1</math>/a</i> Co <sub>2</sub> Al <sub>9</sub>	$a = 855.6$ $b = 629.0$ $c = 621.3$ $\beta = 94.76^\circ$	[V-C]
TiAl <sub>3</sub> < 1395	<i>tI8</i> <i>I4/mmm</i> TiAl <sub>3</sub>	$a = 384.9$ $c = 861$	[1989Kal]

Phase / Temperature Range [°C]	Pearson Symbol/ Space Group/ Prototype	Lattice Parameters [pm]	Comments/References
Ti <sub>9</sub> Al <sub>23</sub> ≤ 780		$a = 384.3$ $c = 3346.4$	superstructure of TiAl <sub>3</sub> [1989Kal]
ξ, TiAl <sub>2.4</sub> (h) 1415-990	<i>tI</i> 16 <i>I4/mmm</i> ZrAl <sub>3</sub>	$a = 391.7$ $c = 1652.4$	[1989Kal]
TiAl <sub>2</sub> < 1175	<i>tI</i> 24 <i>I41/amd</i> HfGa <sub>2</sub>	$a = 397.6$ $c = 2436$	[1989Kal]
TiAl < 1447	<i>tP</i> 4 <i>P4/mmm</i> CuAu	$a = 401.1$ $c = 406.9$ $a = 398.8$ $c = 408.1$	at 46 at.% Al [1989Kal] at 62 at.% Al
Ti <sub>3</sub> Al ≤ 1180	<i>hP</i> 8 <i>P6<sub>3</sub>/mmc</i> Ni <sub>3</sub> Sn	$a = 578.2$ $c = 462.9$	[V-C]
(Ti <sub>1-x</sub> Al <sub>x</sub> )Co TiCo < 1325 CoAl < 1648	<i>cP</i> 2 <i>Pm</i> $\bar{3}m$ CsCl	$a = 299.5$ $a = 286.11$	$0 \leq x \leq 1$ [V-C] [V-C]
*τ <sub>1</sub> , TiCo <sub>2</sub> Al	<i>cF</i> 16 <i>Fm</i> $\bar{3}m$ BiF <sub>3</sub>	$a = 584.7$ $a = 584.8$	[1962Mar], [1979Sei] [V-C]
*τ <sub>2</sub> , Ti <sub>1+x</sub> CoAl <sub>2-x</sub>	<i>cF</i> 116 <i>Fm</i> $\bar{3}m$ Th <sub>6</sub> Mn <sub>23</sub>  Mg <sub>6</sub> Cu <sub>16</sub> Si <sub>7</sub>	$a = 1193$ $a = 1193.56 \pm 0.03$	$0 \leq x \leq 1$ [1972Tsu] TiCoAl <sub>2</sub> [1969Mar] Ti <sub>27.5</sub> Co <sub>23.4</sub> Al <sub>49.1</sub> [2003Gry]; a filled variant of the Th <sub>6</sub> Mn <sub>23</sub> -type
*τ <sub>3</sub> , ≈Ti <sub>8</sub> Co <sub>3</sub> Al <sub>22</sub>	<i>cF</i> 4	$a = 395$	Cu <sub>3</sub> Au-like [1979Sei]

<sup>a)</sup> Possibly only one of the two TiCo<sub>2</sub> based Laves phases is a stable phase [Mas]

**Fig. 1: Al-Co-Ti.**  
Accepted Al-Ti phase  
diagram



**Fig. 2: Al-Co-Ti.**  
Solvus lines of the  
(βTi) phase field  
[1967Tsu]

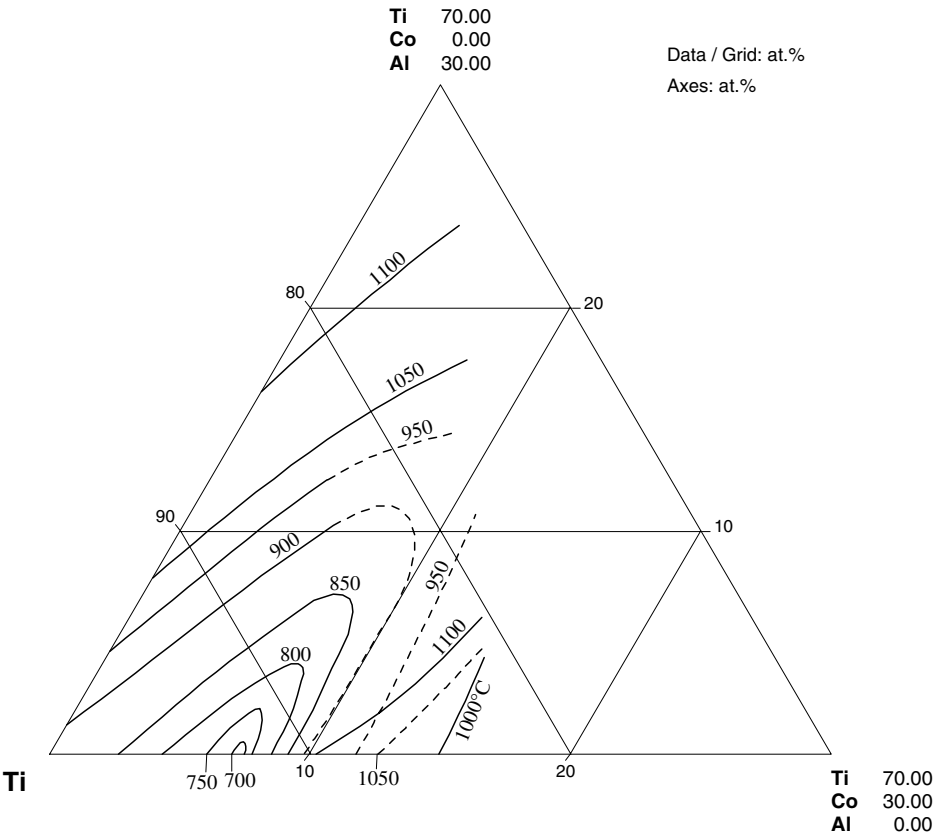
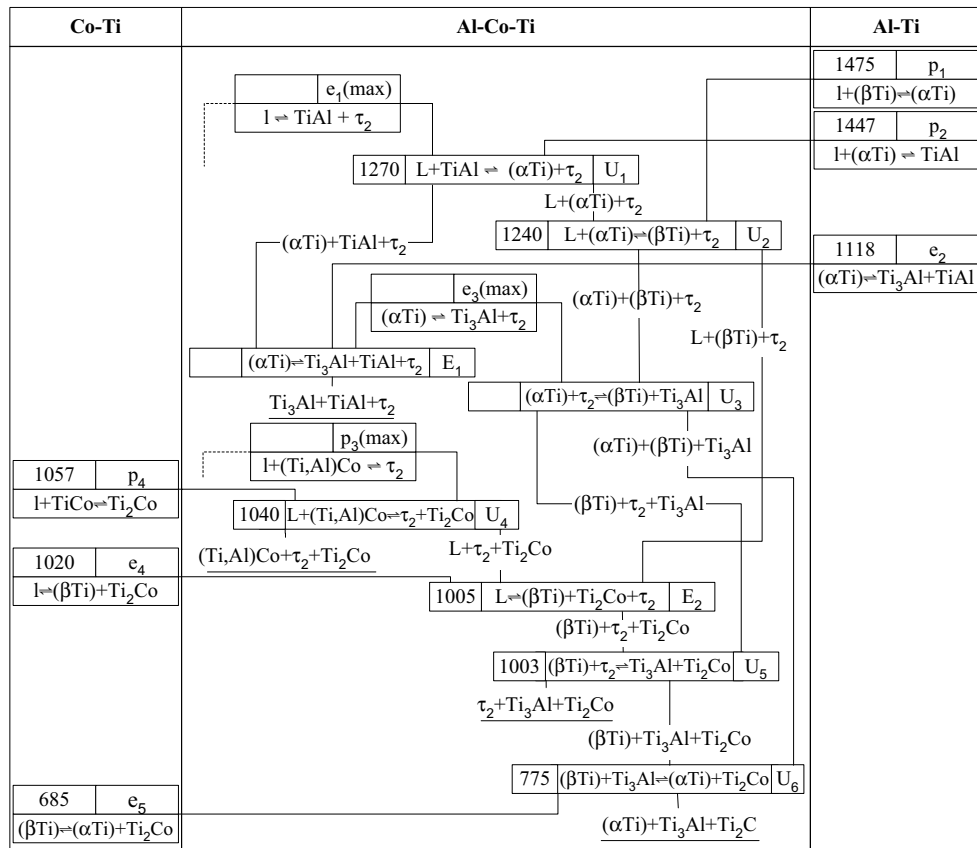
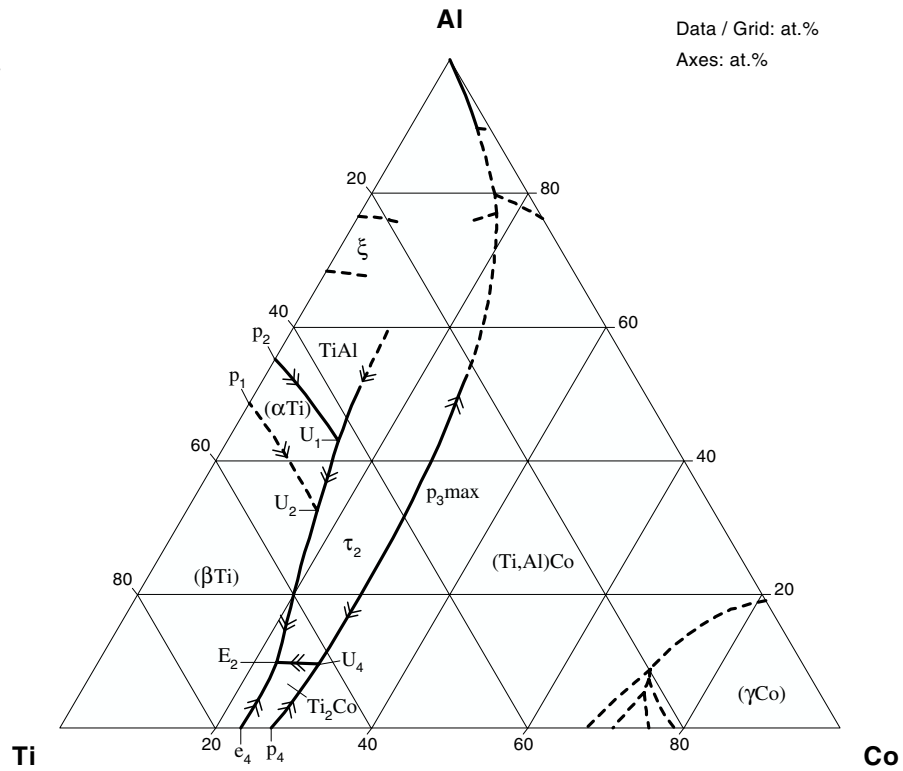
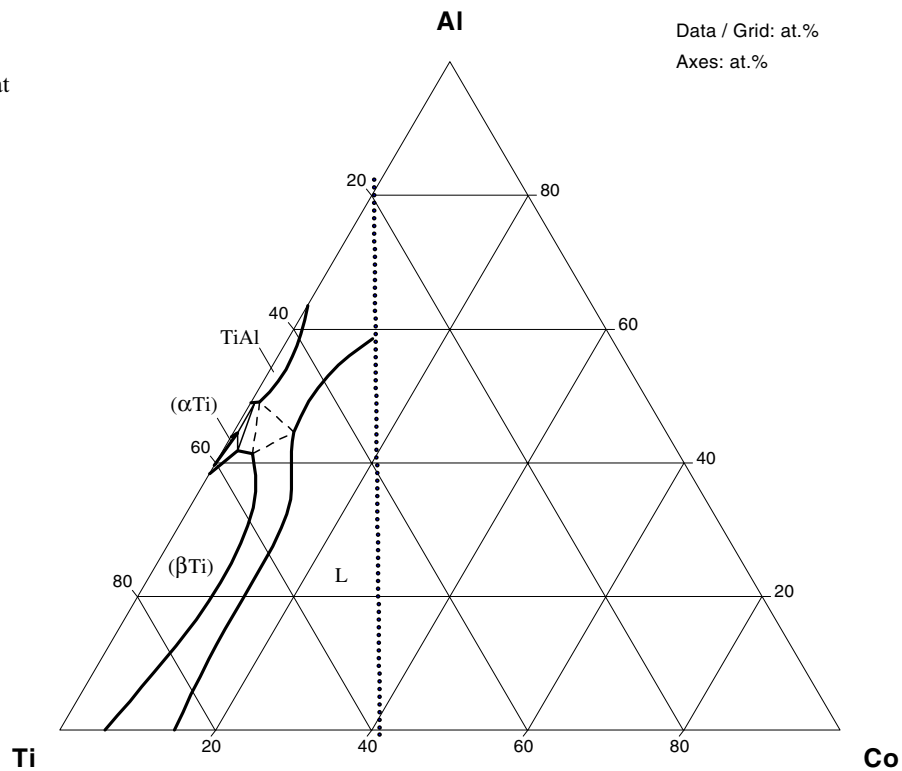




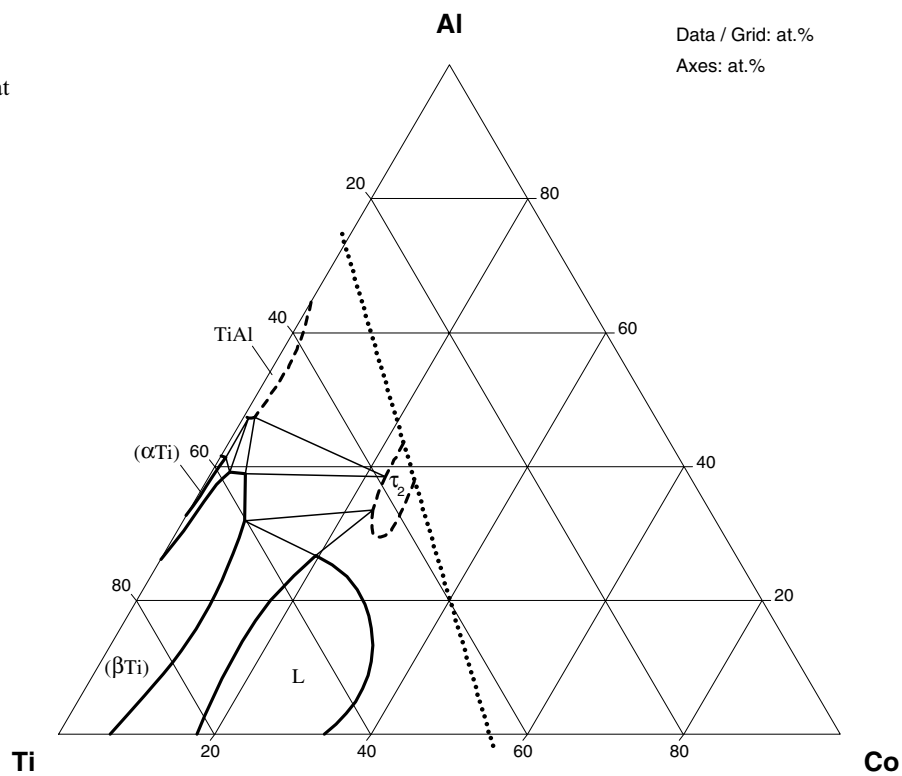
Fig. 3: Al-Co-Ti. Reaction scheme

Fig. 4: Al-Co-Ti.  
Liquidus surface

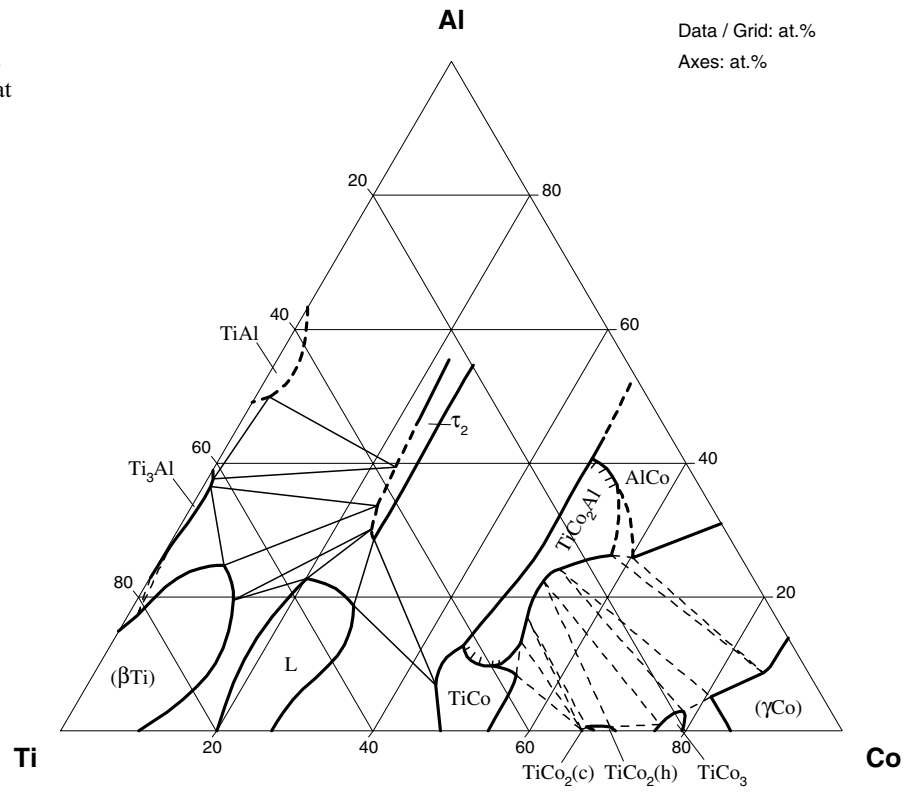
**Fig. 5: Al-Co-Ti.**  
Isothermal section at  
1300°C



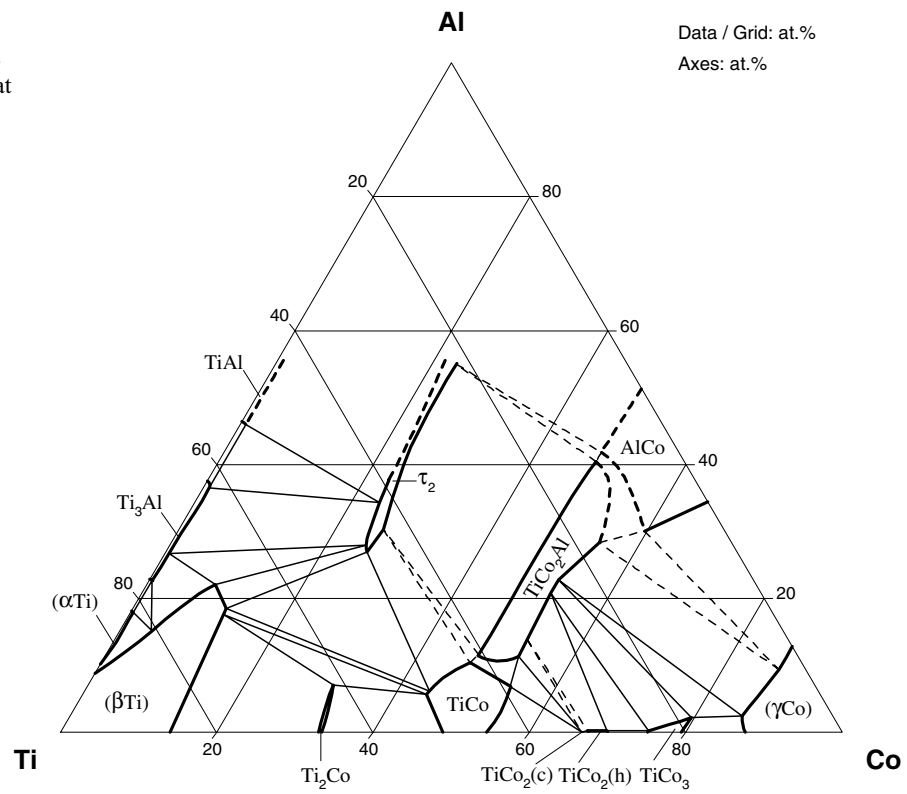
**Fig. 6: Al-Co-Ti.**  
Isothermal section at  
1200°C



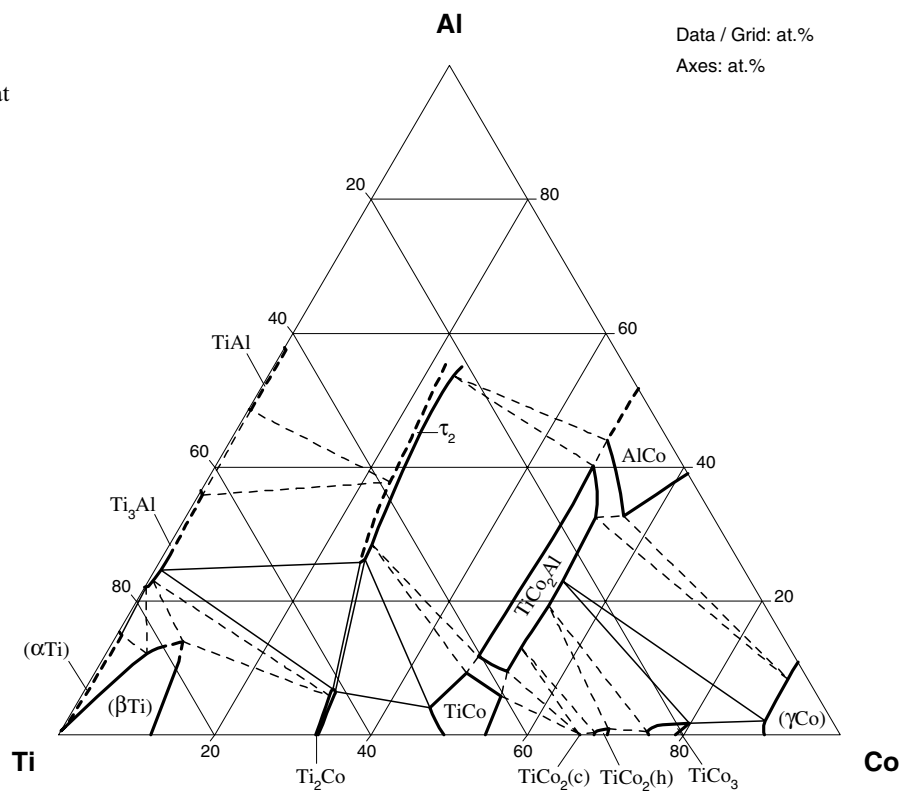
**Fig. 7: Al-Co-Ti.**  
Isothermal section at  
1100°C



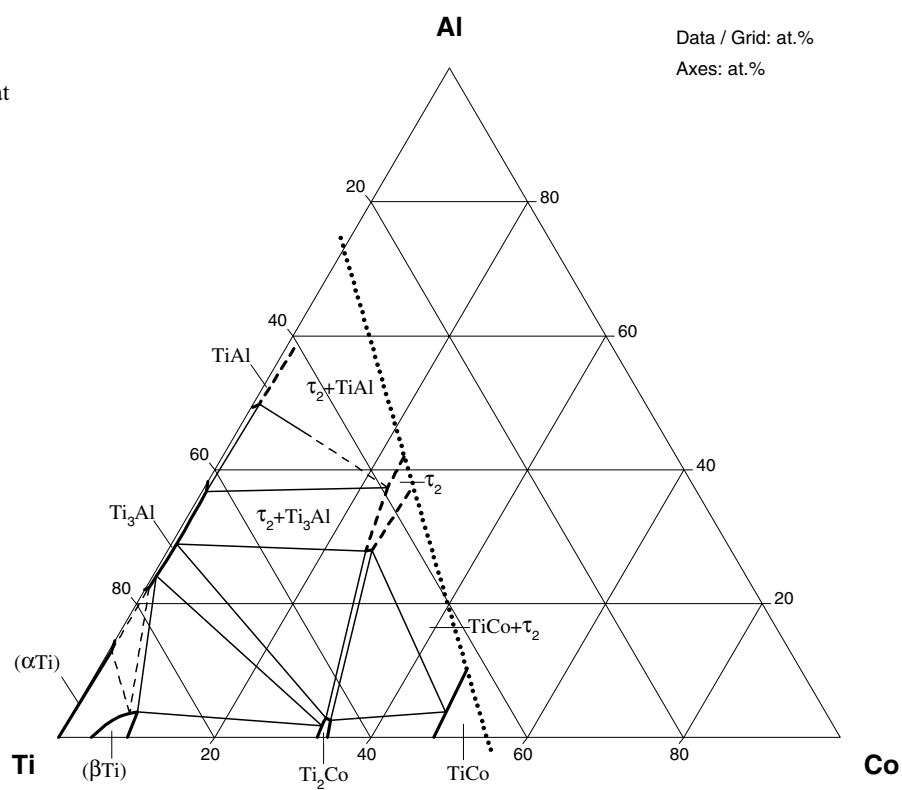
**Fig. 8: Al-Co-Ti.**  
Isothermal section at  
1000°C



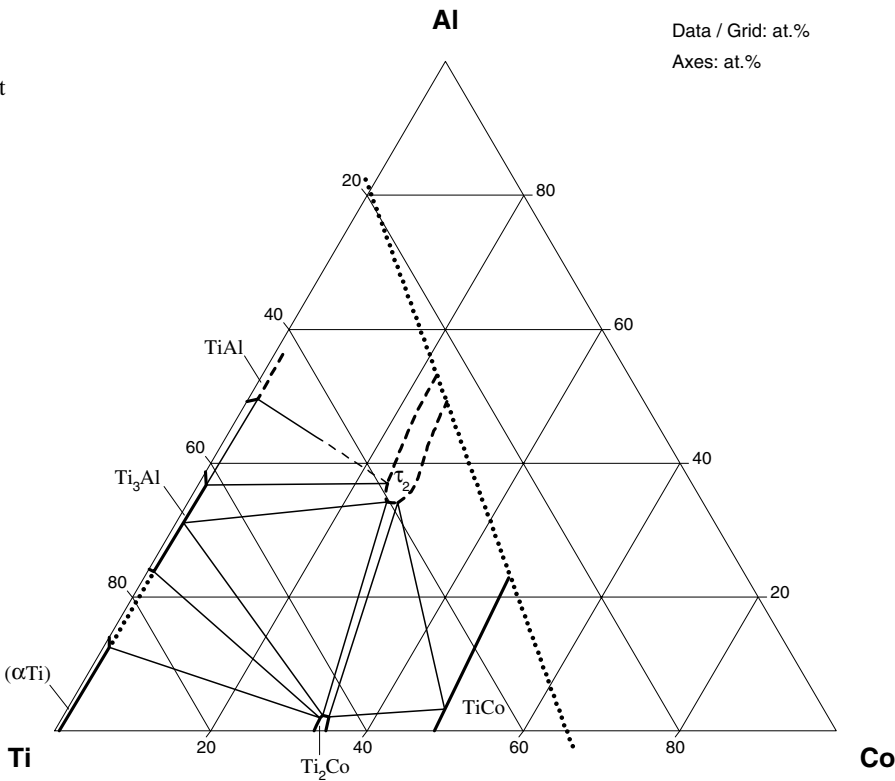
**Fig. 9: Al-Co-Ti.**  
Isothermal section at  
900°C



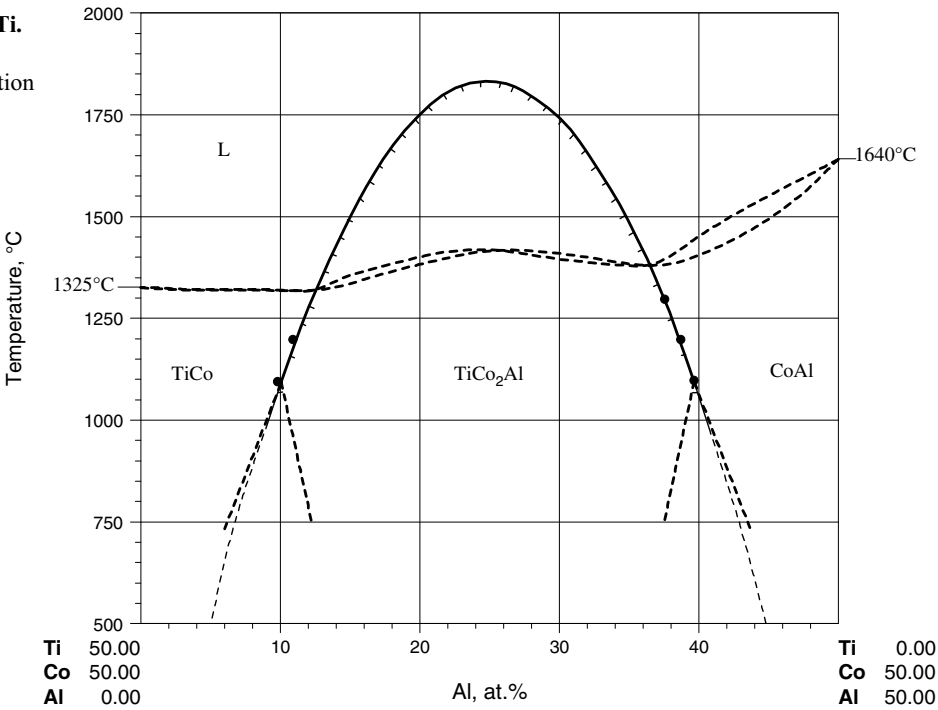
**Fig. 10: Al-Co-Ti.**  
Isothermal section at  
800°C [1972Tsu]



**Fig. 11: Al-Co-Ti.**  
Isothermal section at  
600°C [1972Tsu]



**Fig. 12: Al-Co-Ti.**  
TiCo - CoAl  
pseudobinary section  
[2002Ish1]



**Fig. 13: Al-Co-Ti.**  
Part of the  
TiCo - TiCo<sub>2</sub>Al  
pseudobinary section  
(Co = 52 at.%)  
[2003Kaw]

