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Reassessment of the Binary Aluminum-Titanium Phase Diagram

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All available literature on the constitution of Ti-Al is reviewed. Based on a critical evaluation of these data the phase diagram for this system is assessed.

Keywords binary system, critical evaluation, phase diagram

1. Introduction

Münster, Sagel, and Zwicker stated 50 years ago: "Man kann sich unschwer Phasendiagramme konstruieren, welche den mitgeteilten Ergebnissen Rechnung tragen. Dieselben würden jedoch ohne Heranziehen weiterer experimenteller Daten einen mehr oder weniger spekulativen Charakter besitzen" [1956Mue]. ("One can easily construct phase diagrams, which account for the reported data, but such diagrams would have a speculative character unless further experimental data are added.") This referred to the already observed fact that because of a considerable interest in intermetallic materials based on titanium aluminides, the Al-Ti system has been studied very frequently, but the resulting representations of the phase diagram differ quite a lot from each other. Some of these differences may be attributed to experimental difficulties.

The most thorough assessment of the Al-Ti system was carried out by Murray in 1987 [1987Mur]. This assessment has been used in the standard reference book for binary alloy phase diagrams [1990Mur] and has been updated twice by Okamoto [1993Oka, 2000Oka]. Because [1987Mur] and the consecutive versions of that phase diagram are most frequently used as the standard reference for the Al-Ti system, the present assessment also makes extensive reference to it. The original phase diagram from [1987Mur] is shown in Fig. 1. It is noted that many phase boundaries are given as broken lines, usually due to lack or inconsistency of the data. Furthermore, phase equilibria among the phases αTi, βTi, and Ti_3Al are shown in a thermodynamically improbable way, as data were extremely scattered. Though the diagram by [1987Mur] was intended

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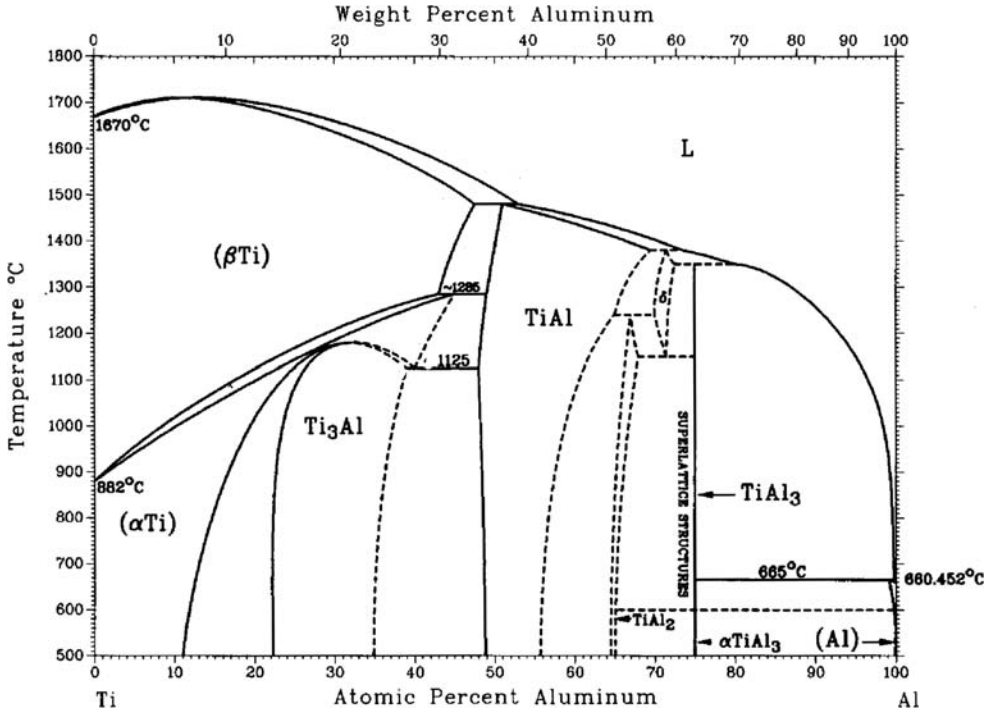

Fig. 1 Ti-Al system according to the assessment of J.L. Murray [1987Mur]

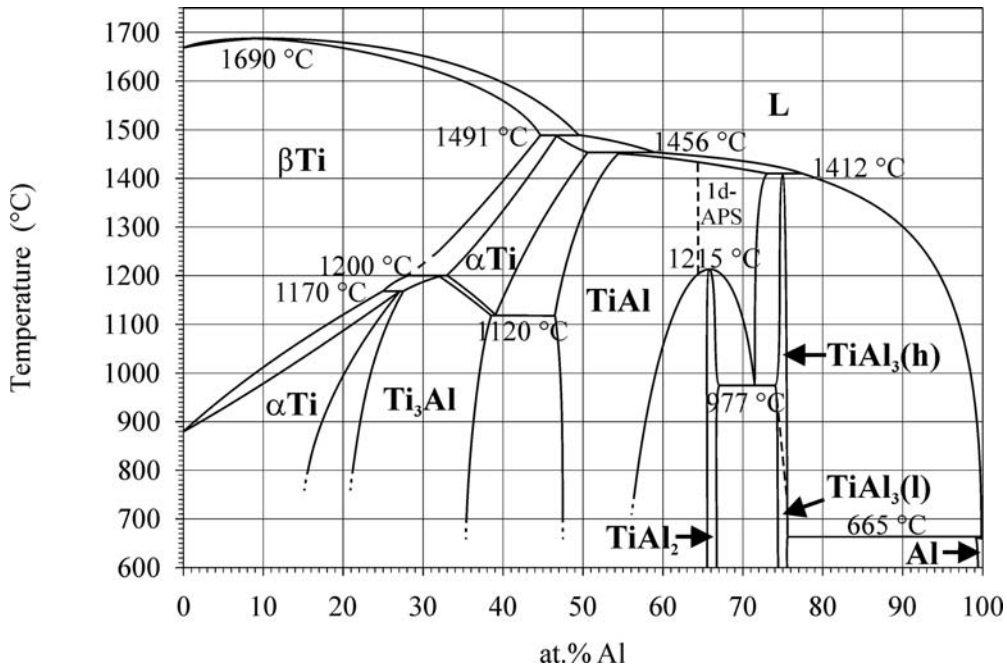

Fig. 2 Ti-Al system according to the current assessment

to show the deficiencies and areas where more work would be needed before phase equilibria were considered settled, later representations of this diagram often showed solid instead of broken lines, thus giving the impression that these phase equilibria had been fixed.

The current assessment critically reevaluates older data as well as reviews data that have become available since [1987Mur]. The phase diagram has been modified according to the current state of knowledge (Fig. 2). All these modifications are explicitly addressed in detail. Each invariant reaction, the accepted temperature, and compositions of the phases involved are presented as subtitles and argued in the following subsection. Crystallographic data for all solid phases are given in Table 1. The vast number of papers consulted for the present assessment is listed according to their information content in three categories: experimental data on the phase diagram, thermodynamic data and modeling of the phase diagram, and theoretical calculation of the phase diagram. To keep the present paper concise and legible, the authors quoted in the text only those references that contributed to their conclusions regarding the phase diagram. However, all papers consulted were retained in the list of references to give the reader the possibility of checking the authors' judgement.

2. Ti-Rich Section: Melting and Solidification

The Ti-rich part of the system contains three invariant reactions involving the liquid phase, i.e., L ↔ βTi, L + βTi ↔ αTi, and L + αTi ↔ TiAl (Fig. 3), as proposed originally by [1951Ogd], instead of the two invariant reactions according to [1987Mur], who accepted [1952Bum]. The three invariant reactions previously mentioned are accepted here as they were confirmed and refined by later research:

2.1 L ↔ βTi at 1690 ± 10 °C and 8.5 ± 3.5 at.% Al

These values are primarily based on [1951Ogd], who found melting points of βTi alloys to increase up to a content of 8.5 at.% Al. The maximum temperature observed was 1710 °C. However, because the melting point for pure Ti reported by [1951Ogd] is 20 K above the accepted value of 1670 °C [1990Mur], a correction of –20 K is applied to their temperatures. The occurrence of a melting point maximum is weakly supported by the data of [1956Kor] but not by [1952Bum]. Although generally accepting the diagram of [1952Bum] for the Ti-rich section, [1987Mur] adopted the melting point maximum for βTi. However, additional experimental confirmation seems desirable.

2.2 L + βTi ↔ αTi at 1491 °C and 49.5, 44.6, 46.7 at.% Al

These values are primarily based on [1999Jun], who investigated the high-temperature phase equilibria by the directional solidification and quenching technique with additional differential thermal analysis. These results are strongly supported by the data of [1989Mis, 1991Mis, 1991Per, 1993Per] (1490 °C; 49.5, 46.5, 47.5 at.% Al) and [1990Hal] (~1486 °C; 47.8, 44.5, 46.2 at.% Al; citing however as reference [1989Hua]). [2000Kov] gave 1490 °C and 48, ~43.5, 46 at.% Al, while [1961Enc, 1965Far = 1973Wil = 1988McC, 1989Hua][1] reported 1472 °C and 49.5-50, 44.4-44.7, 46.1-46.7 at.% Al, respectively. [1993And] found 1517 °C and [1992And] reported 49.6 at.% Al for the liquid phase. [1996Tre = 2004Bul] gave 1520 °C, later corrected to 1505 ± 15 °C [2005Tre]. The

[1]Throughout this text the equal symbol (=) in a reference indicates that the information originally reported in the first reference is reprinted/quoted/accepted in the later work.

Table 1 Crystal structures of the solid phases

Phase, (Strukturbericht designation), temperature range, °C	Pearson symbol, space group, prototype	Lattice parameters, nm		Comments
Al, (*A*l), <660.452	*cF*4, *Fm*$\bar{3}$*m*, Cu	a = 0.40496	[1990Mas]	…
βTi, (*A*2), 1670-882	*cI*2, Im$\bar{3}$*m*, W	a = 0.33065	[1990Mas]	…
αTi, (*A*3), 1491-1120 and <1170	*hP*2, P6$_3$/*mmc*, Mg	a = 0.29506	[1990Mas]	a,c vs. Al-content: [1951Ogd], [1952Bum], [1925Ros], [1966Gro], [1987Mur]
		c = 0.46835		
Ti_3Al, α_2 ($D0_{19}$), <1200	*hP*8, *P*6$_3$/*mmc*, Ni_3Sn [1961Gol]	a = 0.5765	[1967Bla] at 25 at.% Al	a,c vs. Al-content: [1987Mur], [2000Bra1] a,c vs. temp.: [2000Bra1]
		c = 0.4625		
TiAl, γ ($L1_0$), <1456	*tP*4, *P*4/*mmm*, AuCu [1952Duw]	a = 0.4000, c = 0.4075	[1995Bra] at 50 at.% Al	a,c vs. Al-content: [1952Bum], [1987Hua], [1988Vuj], [1993Pfu], [1994Bra2], [1995Bra], [1996Men] a,c vs. temp: [1995Bra], [1999Bit]
$TiAl_2$, <1215	*tI*24, *I*4$_1$/*amd*, $HfGa_2$ [1962Poe]	a = 0.3971	[1989Mab]	a,c vs. temp.: [2000Bra2]
		$c/6$ = 0.4052		
$TiAl_3$(h), ($D0_{22}$), 1387-?	*tI*8, *I*4/*mmm*, $TiAl_3$(h) [1931Fin]	a = 0.3849	[1973Loo1], [2001Bra]	…
		$c/2$ = 0.4305		
$TiAl_3$(l), Ti_8Al_{24}	*tI*32, *I*4/*mmm*, $TiAl_3$(l)	a = 0.3877	[1973Loo1], [2001Bra]	…
		$c/8$ = 0.4229		
Phases not included in the equilibrium diagram; (m) metastable				
$Ti_{1-x}Al_{1+x}$ (?)	*tP*4, *P*4/*mmm*, AuCu [1994Bra1]	a = 0.4030	[1994Bra1]	In as-cast alloys; c/a is inverse to c/a of TiAl
		c = 0.3955		
	*oP*4, *Pmmm*, $Ti_{1-x}Al_{1+x}$ [1990Sch]	a = 0.40262	[1990Sch]	In as-cast alloys
		b = 0.39617		
		c = 0.40262		
Ti_3Al_5(m)	*tP*32, *P*4/*mbm*, Ti_3Ga_5 [1928Mii]	a = 1.1293	[2001Bra]	Precipitated in TiAl
		c = 0.4038		
$TiAl_2$(m)	*oC*12, *Cmmm*, $ZrGa_2$ [1980Mii]	$a/3$ = 0.40315	[1990Sch]	In as-cast alloys
		b = 0.39591		
		c = 0.40315		
1d-APS (Ti_5Al_{11}, Ti_2Al_5)	Tetragonal ordered superstructures of AuCu	a = 0.39230	[1990Sch]	For Ti_5Al_{11} (*I*4/*mmm*, *tI*16, $ZrAl_3$ [1965Ram])
		$c/4$ = 0.41337		
		a = 0.39053	[1990Sch]	For Ti_2Al_5 (*P*4/*mmm*, *tP*28, Ti_2Al_5 [1986Mii])
		$c/7$ = 0.41703		
$TiAl_3$(m)	*cP*4, *Pm*$\bar{3}$*m*, $AuCu_3$	a = 0.3967	[1990Sri]	By mechanical alloying
		a = 0.3972	[1994Bra2]	By splat cooling

original estimate by [1951Ogd] was 1630 °C. This reaction was not accepted by [1987Mur].

2.3 L + αTi ↔ TiAl at 1456 °C and 59, 50.5, 54.5 at.% Al

The coexistence of αTi and TiAl was observed in situ up to the highest temperature measured (1450 °C [1988McC, 1989McC], 1350 °C [1990Shu], 1380 °C by [1998Has]) using high-temperature x-ray diffraction (XRD). The value for the incongruent melting temperature of TiAl is primarily based on [1991And = 1993And], who found a break in the slope of melting points versus Al-content at 1482 °C. Because [1991And = 1993And] observed the temperature for the reaction L + βTi ↔ αTi at 26 K above the accepted value (see above), a correction of –26 K was applied yielding 1456 °C for L + αTi ↔ TiAl. Analogously, [1951Ogd] observed melting of TiAl at 1480 °C. Because their data for pure Ti were 6 K too high at ~900 °C (αTi ↔ βTi was observed at 888 °C versus 882 °C, the accepted value [1990Mur]) and 20 K too high at ~1700 °C (βTi ↔ L was observed at 1690 °C versus 1670 °C, the accepted value [1990Mur]), a correction of –16 K was applied yielding 1464 °C. Strong support comes from [1965Far = 1973Wil = 1988McC = 1990Shu] who reported ~1452 °C. The temperatures reported by [1989Hua, 1990Hal] (~1442 °C), [1989Kal] (1435 °C), and [1991Mis = 1993Per] (1422 °C) are significantly lower, but the value of ~1485 [1996Tre = 2004Bul] (later modified to 1475 ± 10 °C [2005Tre]) is substantially higher. Incidentally, the values given by [1952Bum = 1954McP] or [1960Sat1] (1460 °C) fit nicely, although the former two assumed TiAl to form from L + βTi, which was accepted by [1987Mur].

The composition of the liquid phase (59 at.% Al) is assessed to give a best fit of the liquidus data of [1990Sch], [1991And = 1993And] (as above corrected by –26 K), and [2000Ste = 2002Pal]. Values of 56.7 at.% Al were given in

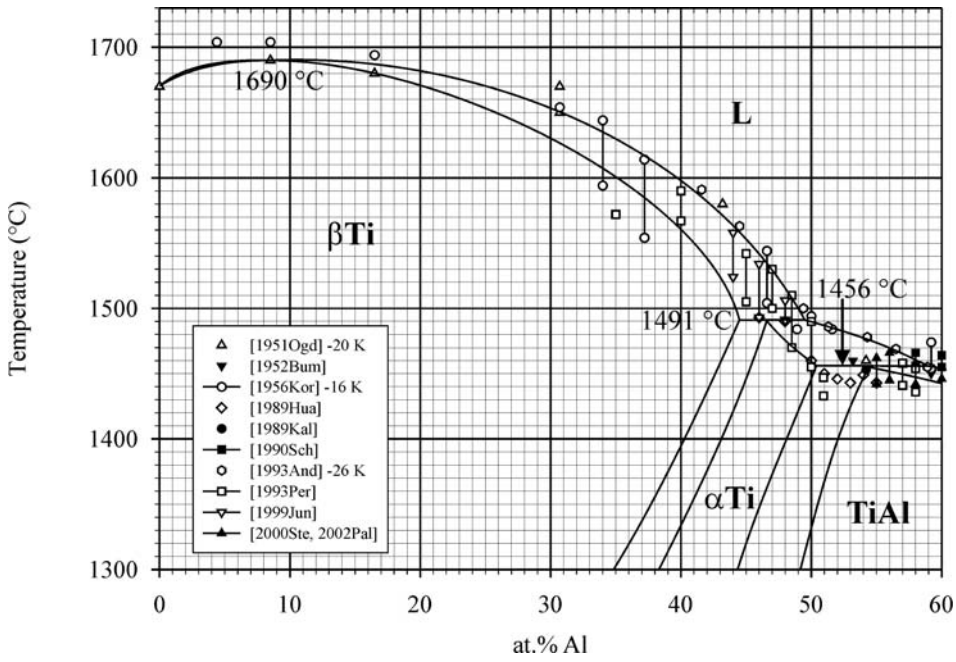

Fig. 3 Liquidus and solidus of the Ti-rich part of the Ti-Al system; the data by [1951Ogd, 1956Kor, 1993And] have been corrected by −20, −16, and −26 K, respectively, for reasons detailed in the text

[1965Far = 1973Wil = 1988McC = 1989McC = 1990Shu], 56 at.% Al in [2000Kov], or 54.5-55 at.% Al by [1989Hua = 1990Hal]. The composition for αTi was found by intersecting best fits for the αTi/αTi + L phase boundary [1999Jun] and for the αTi/αTi + TiAl phase boundary [1989McC, 1994Kai, 1996Din, 1998Vee] with the isotherm at the assessed temperature of 1456 °C. This value is in full agreement with 51 ± 1 at.% Al, which has been measured within a directionally solidified two-phase αTi + TiAl alloy [1993Sno]. The accepted value of 50.5 at.% Al is within 1 at.% of the values given by [1965Far = 1973Wil = 1988McC = 1989McC = 1990Shu, 1989Hua = 1990Hal, 1991Mis = 1993Per, 2000Kov]. The composition of TiAl (53.5 at.% Al) is taken from [1965Far = 1973Wil = 1988McC = 1989McC = 1990Shu]. It is corroborated by the value of 54 ± 1 at.% Al, measured within a directionally solidified alloy [1993Sno] and fits with the extrapolation of the αTi + TiAl/TiAl phase boundary [1994Kai, 1996Din] (Fig. 3). The values by [1989Hua = 1990Hal], [2000Kov], and [1960Sat1] are respectively 1, 1.5, and 2 at.% Al below the assessed value.

3. Ti-Rich Section: Solid-State Reactions

The data existing at the time when [1987Mur] assessed the Al-Ti system did not suggest whether the αTi/Ti_3Al and αTi/βTi phase fields intersect.

As conclusively shown by in situ neutron diffraction experiments [1988Wat], the phase Ti_3Al is formed in the solid state by the reaction βTi + αTi ↔ Ti_3Al [1960Sat1, 1988Wat, 1994Kai], but not by αTi + TiAl ↔ Ti_3Al [1965Kor, 1980Mar, 1980Ouc, 1982Col1] or αTi ↔ Ti_3Al [1966Cro, 1985Shu, 1998Vee, 2000Kov, 2000Ohn] (as assumed by [1987Mur]). As a consequence, two other reactions occur: βTi + Ti_3Al ↔ αTi and αTi ↔ Ti_3Al + TiAl (Fig. 4 and 5).

3.1 βTi + αTi ↔ Ti_3Al at 1200 ± 10 °C and ~28, 33, 32 at.% Al

These values are primarily based on extrapolation of phase boundary data for βTi + αTi/αTi [1990Shu, 1994Kai] as well as αTi/αTi + Ti_3Al/Ti_3Al [1963Cla, 1967Bla, 1990Shu, 1994Kai], and are strongly supported by the results obtained by [1988Wat] (~1200 °C, ~29/35/30 at.% Al). The microstructure data of [2002Suz] are not conclusive, however, because they support either an incongruent formation of Ti_3Al (at 1196 °C and 28.7 / 32 / 31.3 at.% Al) or a congruent formation (see below). The original temperature estimated by [1960Sat1] was 1300-1400 °C.

The accepted incongruent formation of Ti_3Al from βTi + αTi contradicts reports assuming a congruent formation {[1966Cro] at ~875 °C and 25 at.% Al; [1985Shu] at 1169 °C and 30 ± 2 at.% Al (as read from the diagram, which does not obey the phase rule); [1998Vee] at 1178 °C and ~32-33 at.% Al (as read from diagram, which does not obey the phase rule); [2000Kov] at 1180 °C and ~33at.% Al; [2000Ohn] at 1185 °C and ~31 at.% Al; [2002Suz] at 1192 °C (in Table 2) or 1194 °C (read from Fig. 10) and 31.5 at.% Al; or [1996Tre = 2004Bul, 2005Tre] at ~1200 °C and 32 at.% Al}. The supposition by [2002Suz] of a correlation between oxygen content and αTi/Ti_3Al transition temperature does not hold upon looking in detail at the oxygen content data of [1985Shu]. Rather, oxygen contents seem to be less a factor than the mesh size within the existing data grit, as well as the accuracy limits of the experimental methods used.

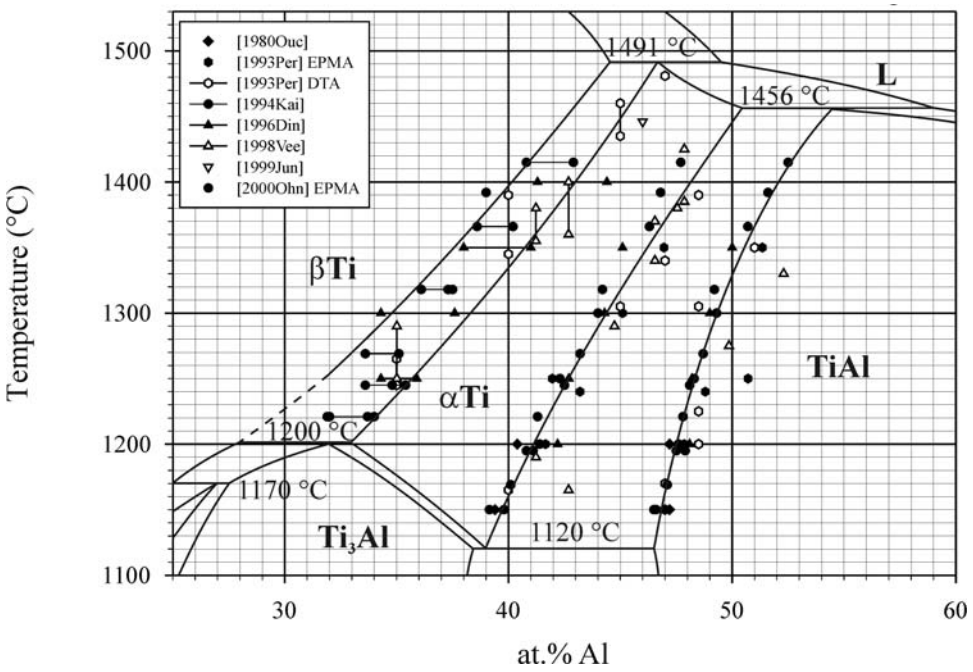

Fig. 4 Phase equilibria between the phases βTi, αTi, and TiAl at high temperatures. The scatter of data for βTi + αTi may be caused by B2-type ordering in βTi as discussed in Sec. 4.1. EPMA data for βTi + αTi from [1993Per] have been omitted as these data have not been considered by the author. Black symbols denote compositions established by EPMA; open symbols denote temperatures determined by DTA.

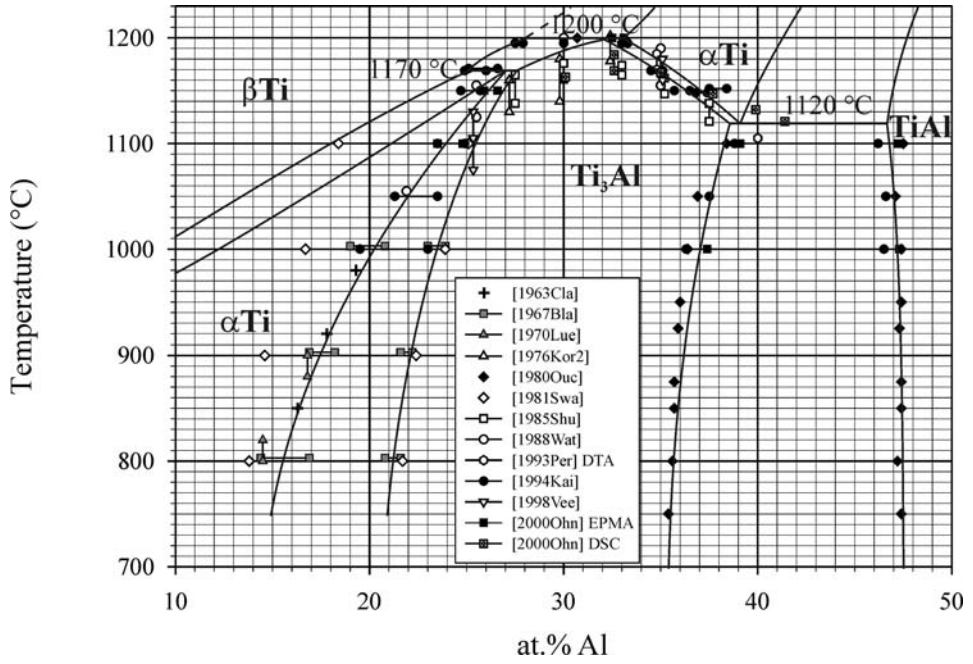

Fig. 5 Phase equilibria involving Ti_3Al. The scatter of the data for the αTi/αTi + Ti_3Al phase boundary may be caused by coherency stresses and short range ordering (SRO) in αTi as discussed in Sec. 4.2. Open symbols denote transformation temperatures, and black symbols denote compositions determined by EPMA. DSC: temperatures determined by differential scanning calorimetry.

The accepted incongruent formation of Ti_3Al from βTi + αTi rules out an incongruent formation of Ti_3Al from βTi + TiAl as proposed by [1965Kor] (1250 °C), [1980Ouc] (1215 °C), [1980Mar] (~1150 °C), or [1982Col1] (~1135 °C).

3.2 αTi ↔ Ti_3Al + TiAl at 1120 ± 10 °C and 39, 38.5, 46.5 at.% Al

The temperature is primarily based on [1992Hel, 1994Kai] who reported 1120 ± 5 °C and 1120 ± 10 °C,

Table 2 Thermodynamic data

Phase	Composition	Property	Value	Technique used	Reference
All phases	Ti_xAl_{1-x} ($0.1 < x < 0.95$)	Activities	…	Vapor pressure at 840 < T °C < 1378	[1996Eck]
Liquid	Ti_xAl_{1-x} ($0 < x < 1$)	ΔH (integral enthalpy of mixing)	ΔH (mixing) reaches min. of –30 kJ/mol of atoms at $x = 0.47$	Calorimetry at 1727 °C	[1974Esi, 1975Esi]
	Ti_xAl_{1-x} ($0 < x < 0.13$)	ΔH (integral enthalpy of mixing)	…	Calorimetry at 1500 °C	[1981Bat]
	Ti_xAl_{1-x} ($0.0018 < x < 0.1783$)	ΔH (integral enthalpy of mixing)	…	Calorimetry at 1448 °C	[1985Bat]
	Ti_xAl_{1-x} ($x \rightarrow 1$)	…	ΔH_{Al} – 98 kJ/mol	Calorimetry at 1727 °C	[1974Esi, 1975Esi]
	Ti_xAl_{1-x} ($x \rightarrow 0$)	…	ΔH_{Ti} – 114 kJ/mole	Calorimetry at 1727 °C	[1974Esi, 1975Esi]
	Ti_xAl_{1-x} ($x \rightarrow 0$)	(ΔH_{Ti}, ΔH_{Al}: partial enthalpies of solution at infinite dilution)	ΔH_{Ti} – 151.9 ± 2.2 kJ/mole	Calorimetry at 745 °C	[1997Tur]
βTi	Ti_xAl_{1-x} ($0.6 < x < 1.0$)	Activities	$RT \ln a_{Ti} = RT \ln x_{Ti} + x_{Al}^2$ (–51.9 ± 3.3) kJ/mol of atoms $RT \ln a_{Al} = RT \ln x_{Al} + x_{Ti}^2$ (–51.9 ± 3.3) + 6.7 ± 2.9) kJ/mol of atoms	Vapor pressure at 1507 °C	[1971Hoc]
αTi	Ti_xAl_{1-x} ($0.8763 < x < 0.9778$)	emf at 650, 741, 768, and 788 °C	…	…	[2003Gas]
	$Ti_{95}Al_{05}$	γ (el. spec. heat coeff.)	3.5 mJK^{-2}/mol of atoms	Adiabatic calorimetry	[1970Col, 1973Col]
	$Ti_{92}Al_8$	emf	464.6 – 0.177T mV	emf at 687 °C	[1970Sam, 1971Sam]
	$Ti_{91.5}Al_{8.5}$	$\Delta H°$ (formation)	–9.5 ± 1.0 kJ/mol of atoms	Direct synthesis calorimetry	[1955Kub]
	$Ti_{85}Al_{15}$	γ (el. spec. heat coeff.)	1.9 mJK^{-2}/mol of atoms	Adiabatic calorimetry	[1970Col, 1973Col]
	$Ti_{55}Al_{45}$	Activities of vs. T (°C) (Al: $1000 < T < 1340$, Ti: $1250 < T < 1420$	…	Vapor pressure	[1999Jac, 2002Jac]
Ti_3Al	$Ti_{80}Al_{20}$	γ (el. spec. heat coeff.)	3.7 mJK^{-2}/mol of atoms	Adiabatic calorimetry	[1970Col, 1973Col]
	$Ti_{76}Al_{24}$	$\Delta H°$ (formation)	–23.9 ± 1.0 kJ/mol of atoms	Direct synthesis calorimetry	[1955Kub]
	$Ti_{75}Al_{25}$	$\Delta H°$ (formation)	–25.3 ± 1.0 kJ/mol of atoms	Direct synthesis calorimetry	[1960Kub]
	$Ti_{75}Al_{25}$	$\Delta H°$ (formation)	–20.3 ± 1.9 kJ/mol of atoms	Direct synthesis calorimetry	[2004Rzy]
	$Ti_{75}Al_{25}$	$\Delta H°$ (formation)	–29.9 ± 3.3 kJ/mol of atoms	Solution calorimetry	[2004Rzy]
	$Ti_{75}Al_{25}$	emf	305.7 – 0.066T mV	emf at 687 °C	[1970Sam, 1971Sam]
	$Ti_{75}Al_{25}$	γ (el. spec. heat coeff.)	1.9 mJK^{-2}/mol of atoms	Adiabatic calorimetry	[1969Ho]
	$Ti_{73}Al_{27}$	emf	364.0 – 0.135T mV	emf at 687 °C	[1970Sam, 1971Sam]
	$Ti_{72}Al_{28}$	γ (el. spec. heat coeff.)	2.2 mJK^{-2}/mol of atoms	Adiabatic calorimetry	[1969Ho]
	$Ti_{71}Al_{29}$	emf	328.0 – 0.099T mV	emf at 687 °C	[1970Sam, 1971Sam]
	$Ti_{70}Al_{30}$	γ (el spec. heat coeff.)	2.5 mJK^{-2}/mol of atoms	Adiabatic calorimetry	[1969Ho]
	$Ti_{69}Al_{31}$	$\Delta H°$ (formation)	–29.4 ± 1.0 kJ/mol of atoms	Direct synthesis calorimetry	[1955Kub]
	$Ti_{69}Al_{31}$	emf	273.23 – 0.0747T mV	emf at $547 < T$ °C < 627	[2001Red]
	$Ti_{67}Al_{33}$	emf	228.6 – 0.032T mV	emf at 687 °C	[1970Sam, 1971Sam]
	$Ti_{64}Al_{36}$	$\Delta H°$ (formation)	–30.9 ± 1.0 kJ/mol of atoms	Direct synthesis calorimetry	[1955Kub]
	$Ti_{65}Al_{35}$	$\Delta H°$ (formation)	–30.0 ± 1.0 kJ/mol of atoms	Direct synthesis calorimetry	[1960Kub]
TiAl	$Ti_{52}Al_{48}$	emf	219.69 – 0.0914T mV	emf at $547 < T$ °C < 627	[2001Red]
	$Ti_{51}Al_{49}$	$\Delta H°$ (formation)	–40.0 ± 1.0 kJ/mol of atoms	Direct synthesis calorimetry	[1955Kub]

(continued)

Table 2 Thermodynamic data (continued)

Phase	Composition	Property	Value	Technique used	Reference
	$Ti_{50}Al_{50}$	$\Delta H°$ (formation)	−36.5 ± 1.0 kJ/mol of atoms	Direct synthesis calorimetry	[1960Kub]
	$Ti_{50}Al_{50}$	$\Delta H°$ (formation)	−36.2 ± 1.0 kJ/mol of atoms	Direct synthesis calorimetry	[1960Kub]
	$Ti_{50}Al_{50}$	$\Delta H°$ (formation)	−35.1 ± 0.5 kJ/mol of atoms	Direct synthesis calorimetry	[2003Nas]
	$Ti_{50}Al_{50}$	$\Delta H°$ (formation)	−37.1 ± 1.0 kJ/mol of atoms	Direct synthesis calorimetry	[2004Rzy]
	$Ti_{50}Al_{50}$	$\Delta H°$ (formation)	−41.9 ± 3.1 kJ/mol of atoms	Direct synthesis calorimetry	[2004Rzy]
	$Ti_{50}Al_{50}$	emf	149.3 − 0.074T mV	emf at 687 °C	[1970Sam, 1971Sam]
	$Ti_{46}Al_{54}$	$\Delta H°$ (formation)	−40.0 ± 1.0 kJ/mol of atoms	Direct synthesis calorimetry	[1955Kub]
	$Ti_{45}Al_{55}$	$\Delta H°$ (formation)	−37.5 ± 1.0 kJ/mol of atoms	Direct synthesis calorimetry	[1960Kub]
	$Ti_{42}Al_{58}$	$\Delta H°$ (formation)	−40.3 ± 0.8 kJ/mol of atoms	Direct synthesis calorimetry	[1955Kub]
	$Ti_{40}Al_{60}$	$\Delta H°$ (formation)	−38.8 ± 1.0 kJ/mol of atoms	Direct synthesis calorimetry	[1960Kub]
	$Ti_{38}Al_{62}$	Activities vs. T (°C) (Al: 900 < T < 1200, Ti: 1250 < T < 1350	…	Vapor pressure	[1999Jac]
$TiAl_2$	$Ti_{35}Al_{65}$	emf	217.85 − 0.126T mV	emf at 547 < T °C < 627	[2001Red]
	$Ti_{33}Al_{67}$	$\Delta H°$ (formation)	−37.1 ± 0.9 kJ/mol of atoms	Direct synthesis calorimetry	[2003Nas]
	$Ti_{33}Al_{67}$	$\Delta H°$ (formation)	−38.6 ± 2.6 kJ/mol of atoms	Solution calorimetry	[2004Rzy]
$TiAl_3$	$Ti_{26}Al_{74}$	emf	193.42 − 0.1055T mV	emf at 547 < T °C < 627	[2001Red]
	$Ti_{25}Al_{75}$	$\Delta H°$ (formation)	−36.9 ± 1.0 kJ/mol of atoms	Direct synthesis calorimetry	[1955Kub]
	$Ti_{25}Al_{75}$	$\Delta H°$ (formation)	−35.6 ± 1.0 kJ/mol of atoms	Direct synthesis calorimetry	[1960Kub]
	$Ti_{25}Al_{75}$	γ (el. spec. heat coeff.)	1.1 mJK^{-2}/mol of atoms	Electrical resistivity	[1974Dun]
	$Ti_{25}Al_{75}$	θ_D (Debye Temp.)	444 K	Electrical resistivity	[1974Dun]
	$Ti_{25}Al_{75}$	C_{p298}	22.6 JK^{-1}/mol of atoms	Adiabatic calorimetry	[1974Stu]
	$Ti_{25}Al_{75}$	ΔH_{298} (formation)	−36.9 ± 1.3 kJ/mol of atoms	Direct synthesis calorimetry	[1974Stu]
	$Ti_{25}Al_{75}$	$\Delta H°$ (formation)	−36.6 ± 1.2 kJ/mol of atoms	Direct synthesis calorimetry	[1994Mes]
	$Ti_{25}Al_{75}$	$\Delta H°$ (formation)	−39.2 ± 1.8 kJ/mol of atoms	Direct synthesis calorimetry	[2003Nas]
	$Ti_{25}Al_{75}$	$\Delta H°$ (formation)	−37.8 ± 2.3 kJ/mol of atoms	Solution calorimetry	[2004Rzy]

respectively. This value is strongly supported by [1993Bar] (1110 < T < 1125 °C), [1985Shu, 1989Mis = 1991Mis = 1993Per] (1110 °C), [1989McC] (showing ~1113 °C, but citing [1988Jon], who in turn cites [1987Mur] giving 1125 °C!), [1979Muk] (~1130 °C), [1990Shu] (1125 °C, citing [1989McC]!), [1993Ram, 1999Vee = 2003Vee] (1125 °C) [2000Kov] (~1118 °C), or [1996Tre = 2004Bul] (~1125 °C). The value given by [1960Sat1] was 1050 °C.

The compositions for αTi (39 at.% Al) and Ti_3Al (38.5 at.% Al) are accepted from [1992Hel, 1994Kai] and strongly supported by [1960Sat1, 1991Mis = 1993Per] (39 and 37.5 at.% Al), [1993Bar] (39.6 and 38.5 at.% Al), [1985Shu] (39.5 at.% Al for αTi), and [1990Shu] (40 and 39 at.% Al). The value accepted for TiAl (46.5 at.% Al) is a compromise between the data of [1992Hel, 1994Kai] (45.8 at.% Al), [1991Mis = 1993Per, 1989McC = 1990Shu] (48 at.% Al), and [1993Bar] (48.5at.% Al).

The accepted eutectoid decomposition of αTi into Ti_3Al + TiAl also rules out an incongruent formation of Ti_3Al from βTi + TiAl (see also above).

3.3 βTi + Ti_3Al ↔ α Ti at 1170 ± 10 °C and 25, ~27.5, 27 at.% Al

These values are primarily based on the extrapolation of the data for the phase boundaries βTi + αTi/αTi [1990Shu, 1994Kai] as well as αTi/αTi + Ti_3Al/Ti_3Al [1963Cla, 1967Bla, 1990Shu, 1994Kai]. They are strongly supported by the data of [1961Enc] (as corrected by [1965Far = 1973Wil]: 1172 ± 8 °C and 23.9, 28, 26 at.% Al), [1973Sas] (1180 °C and 18.7, 26.0, ~30.7 at.% Al; given as 1100 °C and 15.7, 25.3, 21 at.% Al in earlier versions [1966Tsu, 1967Tsu]); [1988Wat] (~1150 °C and 24, 26, 25 at.% Al) and [1994Kai] (1160 ± 10 °C and 23.5, 26, 25 at.% Al). Other data agree on the occurrence of such a reaction, but report deviating temperatures as well as compositions ([1963Cla] 1100 °C and 15.5 / 25.3 / 23.5 at.% Al; [1965Kor, 1976Kor2] 1080 ± 20 °C and 15 / 22.5 / 17.5 at.% Al; as well as [1980Ouc] 1115 °C and 24 at.% Al for αTi).

There is a considerable scatter in the data from the available literature concerning the phase boundaries shown in Fig. 4. For plotting the figure, preference has been given to those data where the compositions of the coexisting phases have been directly determined in quenched samples by electron probe microanalysis (EPMA), though two sets of such data were disregarded. EPMA data for βTi + αTi from [1993Per] have been omitted as these data have not been considered by the author himself. EPMA data from [1993Bar] for αTi/αTi + TiAl are in good agreement with the respective phase boundary shown in Fig. 4, but their data for coexisting TiAl are markedly shifted toward higher

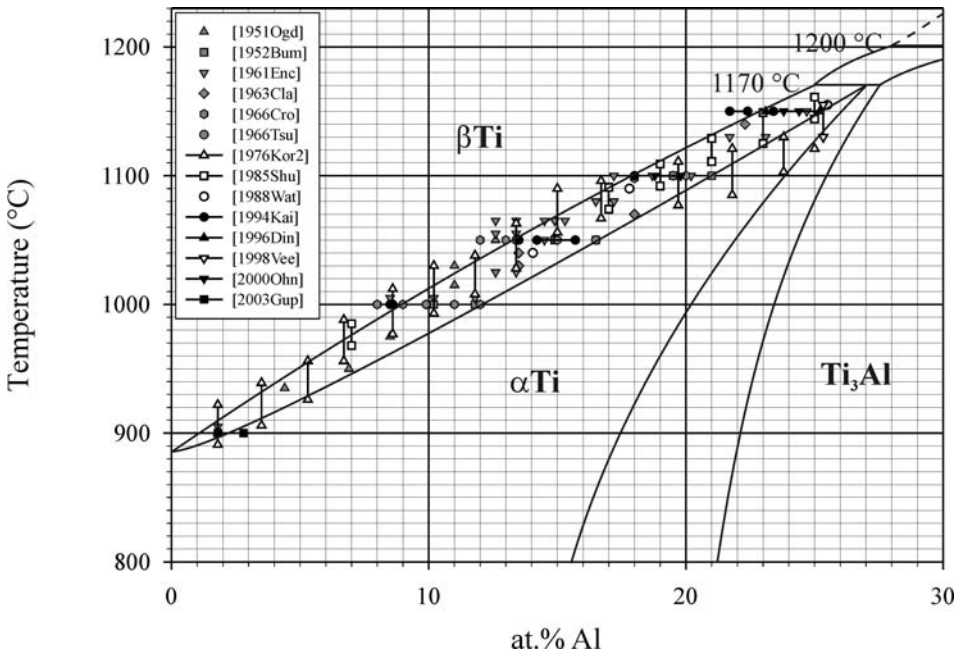

Fig. 6 Phase equilibria between βTi and αTi. Grey symbols denote 2-phase alloys, open symbols denote upper and lower transformation temperatures [1976Kor2, 1985Shu, 1998Vee] or transformation temperature [1988Wat], and black symbols denote compositions determined by EPMA

Al-contents when compared with other investigations. As there is no apparent reason for this shift, the data of [1993Bar] have not been included in Fig. 4.

The βTi + αTi two-phase field is shown in Fig. 6. Data for the phase boundaries have been reported from metallographic observations and x-ray diffraction (XRD) [1951Ogd, 1952Bum, 1960Sat1, 1961Enc, 1963Cla, 1966Cro, 1966Tsu], transmission electron microscopy (TEM) [1970Bla, 1973Sas], or by more direct methods such as the hydrogen pressure method [1955McQ], electrical resistivity measurements [1960Sat1, 1963Kor, 1998Vee], differential thermal analysis (DTA) [1970Jep, 1976Kor2, 1985Shu, 1986Sir], in situ neutron diffraction [1988Wat], or EPMA of coexisting phases in quenched samples [1980Ouc, 1994Kai, 1996Din, 2000Ohn, 2003Gup]. From the majority of the above results, it is clear that the βTi + αTi two-phase field is quite narrow. It has also conclusively been shown that the αTi + βTi two-phase field broadens by the addition of oxygen [1954Thy, 1956Sch]. Therefore, data from the direct methods that suggest a more extended two-phase area have not been taken into account. Furthermore, Fig. 6 shows only data of metallographic and XRD investigations in which the alloys have been identified as βTi + αTi.

4. Ti-Rich Section: Features not Adopted

4.1 B2-type Ordering of βTi

In Fig. 4, the phase boundary of βTi is drawn as a dashed line only between 1200 and 1250 °C. It would be the "obvious" extrapolation between the invariant reaction at 1200 °C and the data for T > 1250 °C, but it is not supported by experimental results [1993Per, 1994Kai, 1996Din, 2000Ohn], which require it to be shifted by up to 2 at.% towards more Al-rich compositions in the said temperature interval. As a possible explanation a B2-type ordering of βTi has been proposed [1994Kai] and seemed plausible from a theoretical point of view [2000Ohn]. On the other hand it has been pointed out that the occurrence of B2 order at a content of about 25 at.% of solute seems to be rather unlikely though the question will only be resolved by work on suitable ternary alloys [1998Mas].

4.2 Influence of Coherency Stresses, Oxygen and Short Range Ordering (SRO) on αTi/αTi + Ti_3Al

A related issue, the dependency of the αTi + Ti_3Al phase field on coherency stresses, is discussed by [1970Lue, 1988Wat, 1993Huh]. Oxygen, too, has a considerable influence on the αTi/αTi + Ti_3Al phase boundary in that it shifts the phase boundary to higher temperatures and lower Al contents [1968Cro, 1969Cro, 1970Bla, 1972Kor, 1976Lim, 1988Wat, 1996Ard]. This shift has also been attributed to a change of coherency by the addition of oxygen [1976Lim].

Furthermore, [1967Bla, 1969Bla, 1973Nam, 1988Wat] consider possible short range ordering (SRO) in αTi. Different results in the αTi /αTi + Ti_3Al phase boundary are obtained, whether a SRO sensitive analytical method is used or not [1988Wat]. The presence of SRO in αTi is supported by electrical resistively and magnetic measurements by [1956Mue, 1961Yao, 1963Cla, 1966Cro, 1973Nam, 1981Swa]. It has also been claimed recently that the presence of SRO has been verified by neutron diffraction [2001Nee, 2002Nee].

4.3 Ti_2Al

There have been repeated reports of a hexagonal Ti_2Al phase [1957Enc, 1983Loi, 1994He, 1999Yu]. These phases have been shown to be either nitrogen or carbon impurity stabilized [1986Kau, 1991Hua, 1996Cha]. In the case of oxygen contamination, a cubic βMn-type phase with a_0 = 0.691 nm labeled $Ti_3Al_2O_3$ [1996Bey, 1999Abe] or $Ti_5Al_3O_2$ [1995Zhe, 1997She] has been reported. This phase must be part of the homologous series of phases $Ti_{2n-1}Al_n$ postulated by [1998Men2].

5. Al-Rich Section: Melting and Solidification

In the Al-rich part, two invariant reactions involving the liquid phase are universally accepted (Fig. 7a).

5.1 L + $TiAl_3$(l) ↔ Al at 665 °C and 99.92, 75.5, 99.2 at.% Al

The temperature for this peritectic reaction has been well established [1931Fin, 1940Nis, 1948Fin, 1972Max, 1974Cis = 1974Ker, 1978Shi, 1984Abd, 1984Hor] and also the low solubility of Ti in the liquid at this temperature [1931Fin, 1934Boh, 1948Fin, 1974Hec, 1983Sch, 1984Abd] (Fig. 7b). The composition of Al is based on EPMA measurements [1984Abd, 1991Min], which are supported by resistivity measurements of [1984Hor]. It agrees with [1948Fin] and has been adopted by [1987Mur]. From measuring the microhardness, resistivity and lattice constants a higher Al-content of about 99.85 at.% Al had been deduced previously [1946Bue, 1983Kuz]. No measurements for the composition of $TiAl_3$ at the peritectic temperature exist.

It is noted that data for the Al-rich liquidus between about 80 and 90 at.% Al are available only from [1923Erc, 1926Man]. As pointed out later, undercooling is frequently encountered in Al-rich Al-Ti melts, and this may attribute for the difference between the temperature for the peritectic decomposition of $TiAl_3$ at 1412 °C (see consecutive section) and that estimated from extrapolation of the data by [1923Erc, 1926Man].

5.2 L + "A" ↔ $TiAl_3$(h) at 1412 ± 4 °C and ~77.5, ~73, 75 at.% Al

The phase "A" is given either as TiAl [1951Ogd, 1952Bum, 1956Kor, 1973Wil, 2001Ste2, 2002Pal], Ti_5Al_{11} [1965Ram, 1987Mur, 1989Kal, 1990Sch, 1993Hay, 2001Bra], Ti_2Al_5 [1973Loo2, 1992Kat, 1993Oka, 1997Zha, 2000Oka], LP_{HT}/1d-APS [1985Loi4, 1988Loi, 2001Ste2, 2002Pal], or γ_2 [2005Kai] for reasons described below.

The reaction temperature of 1412 ± 4 °C (Fig. 8) is based on the differential thermal analysis (DTA) data of [1956Kor] (1410 °C), [1990Sch] (1416 °C; in more recent experiments additional peaks at 1387 °C reported in [1990Sch] could not be reproduced [2005Sch]), [2000Ste] (1411 °C), [2002Pal] (1408 °C), and [2005Tre] (1410 ± 15 °C; modified from 1425 °C [1996Tre = 2004Bul]). The value given by [1989Kal] was a little lower (1395 °C). An even lower value of ~1340 °C was obtained by visual observations of incipient melting [1952Bum] (accepted by [1987Mur]).

The compositions for the phases participating in the formation of $TiAl_3$(h) are derived by extrapolation. Compared with previously assessed data ([1952Bum = 1973Wil]: 75.9/72.7/74.9 at.% Al; [1965Ram]: 76.5, 70, 74.9 at.% Al; [1987Mur]: ~80, ~72.5, 75 at.% Al; [1989Kal]: 76.7, 72.5, 75 at.% Al; [1990Sch = 1993Hay = 2000Kov = 2001Bra]: ~81, 71.1, 75 at.% Al; [2000Oka]: 79.1, 71.4, 75 at.% Al; [2005Tre]: 78.0, 71.5, 75 at.% Al) there is general agreement that $TiAl_3$(h) forms peritecticaly at the stoichiometric composition. There is also reasonable agreement about the composition of "A," especially when taking into account that this composition has been associated with a number of different phases. The composition for the liquid can only be estimated. Data from [1956Kor] (as given in the phase diagram but not in the table) and [1989Kal] show that the liquidus temperature at 80.6 at.% Al is already markedly below the peritectic temperature. Therefore the composition should be below 80 at.% Al.

6. Al-Rich Section: Solid State Reactions

In the Al-rich section [1987Mur] presented all phases and phase equilibria in the solid state by broken lines except $TiAl_3$, which was drawn as a single solid line. Substantially more data have become available by now for this part of the phase diagram and most of the issues are settled (Fig. 9).

For temperatures above 1215 °C, all recent investigations of the Al-rich phase equilibria showed the absence of extended two-phase fields up to about 72 at.% Al [1999Pal, 2001Bra, 2001Ste2, 2002Pal, 2004Hat2, 2005Kai]. All alloys ranging from 65 to 72 at.% Al quenched from these or lower temperatures showed the presence of 1d-APS (denoted as $Ti_5Al_{11}(DO_{23})$ [1965Ram, 1990Sch, 1994Bra2, 2001Bra], $\gamma_2(D0_{23}')$ [2005Kai], Ti_2Al_5 [1973Loo2, 1990Sch], one-dimensional antiphase domain structures (1d-APS) [1980Mii, 1981Mii, 1984Mii, 1986Mii, 2002Pal], or "long period structures (LP)" [1984Loi, 1985Loi2, 1985Loi3, 1985Loi4, 1988Loi]). It remains to be settled, whether there is a single-phase field of TiAl with a second-order transition to 1d-APS at 65 ± 1 at.% Al [2002Pal, 2005Kai], if small two-phase fields (≤1 at.%) exist [2001Bra], or if the 1d-APS are transient structures in precipitation reactions occurring upon cooling [1990Sch]. Below 1215 °C, the ordered phase $TiAl_2$ becomes stable. Below 977 °C, this phase coexists with $TiAl_3$(h).

6.1 TiAl/1d-APS ↔ $TiAl_2$ at 1215 °C and 65.7 at.% Al

There is general agreement that $TiAl_2$ with $HfGa_2$-type structure is stable up to 1215 °C [1990Sch, 1997Ben, 2001Bra, 2001Ste2, 2002Pal]. The value given by [1989Kal] is significantly lower (1175 °C). The original estimate was ~1250 °C [1965Ram]. It has conclusively been shown that $ZrGa_2$-type $TiAl_2$ is a metastable phase

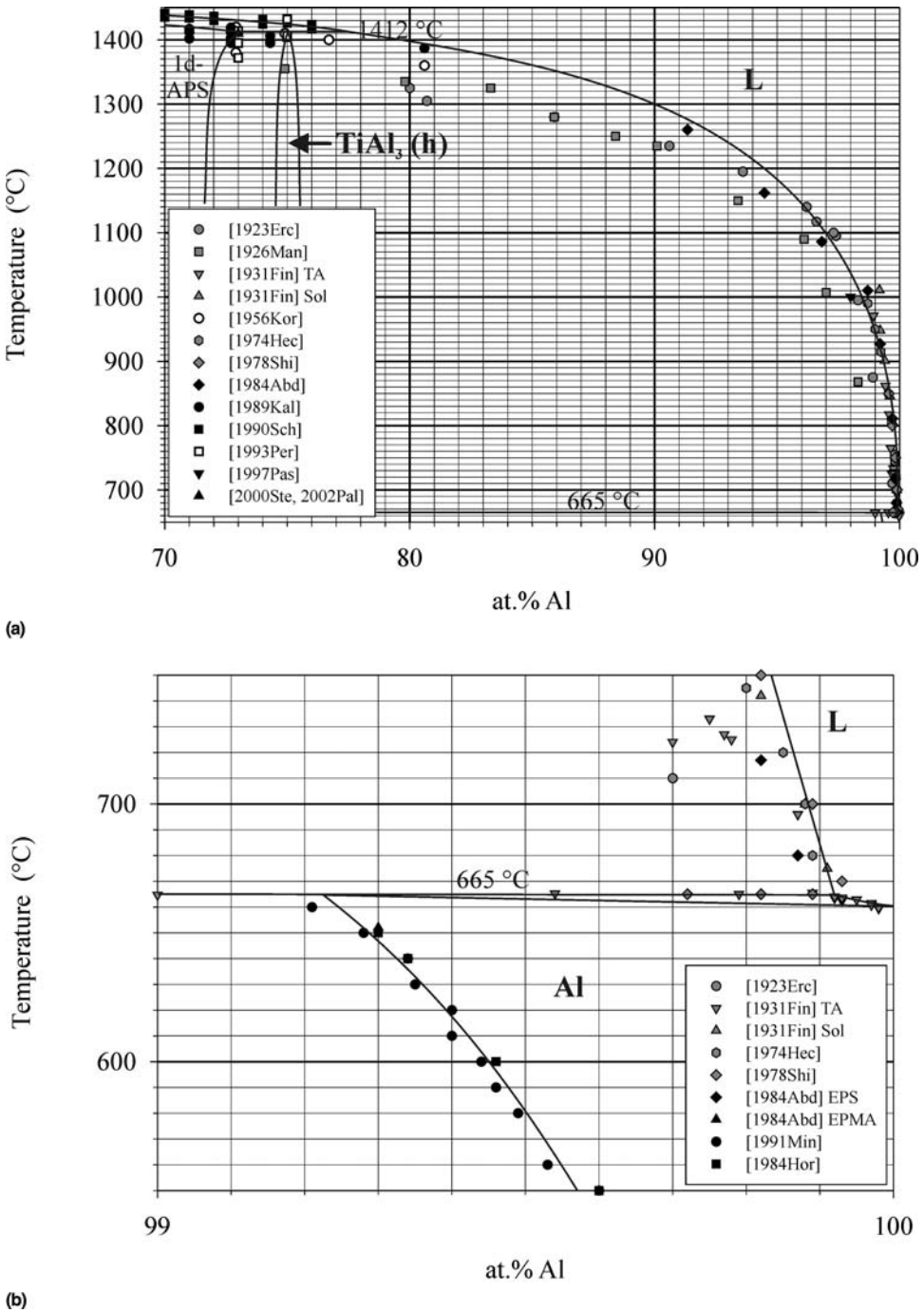

Fig. 7 (a) Liquidus of the Al-rich part of the Ti-Al system (TA: thermal analysis; Sol: determination of the solute by wet chemical analysis). (b) Solubility of Ti in solid and liquid Al (TA: thermal analysis; Sol: determination of the solute by wet chemical analysis; EPS: electromagnetic phase separation)

[2000Bra2, 2001Bra, 2001Ste1, 2001Zha], and it is therefore labeled $TiAl_2$(m) [2000Bra2]. The Al-content of the Al-rich phase boundary of TiAl increases steadily with temperatures up to 66-67 at.% Al at 1215 °C. At higher Al-contents, the Ti-rich phase boundary of the 1d-APS decreases to 977 °C at 72 at.% Al (Fig. 9) [1973Loo2, 1999Pal, 2001Bra, 2002Pal, 2005Kai]. Because of this shape of the TiAl phase limit, precipitation of $TiAl_2$ occurs upon cooling alloys from T > 1215 °C between 60 and 72 at.% Al. It seems that $TiAl_2$ forms by ordering from TiAl. Considering the crystal structures (Table 1) it is plausible that $TiAl_2$(m) ($ZrGa_2$-type) has no nucleation problems and thus will appear until the stable phase $TiAl_2$ ($HfGa_2$-type) has grown. As this is a very slow process, $TiAl_2$(m) can persist for considerable time [1999Nak, 2001Ste1, 2001Zha].

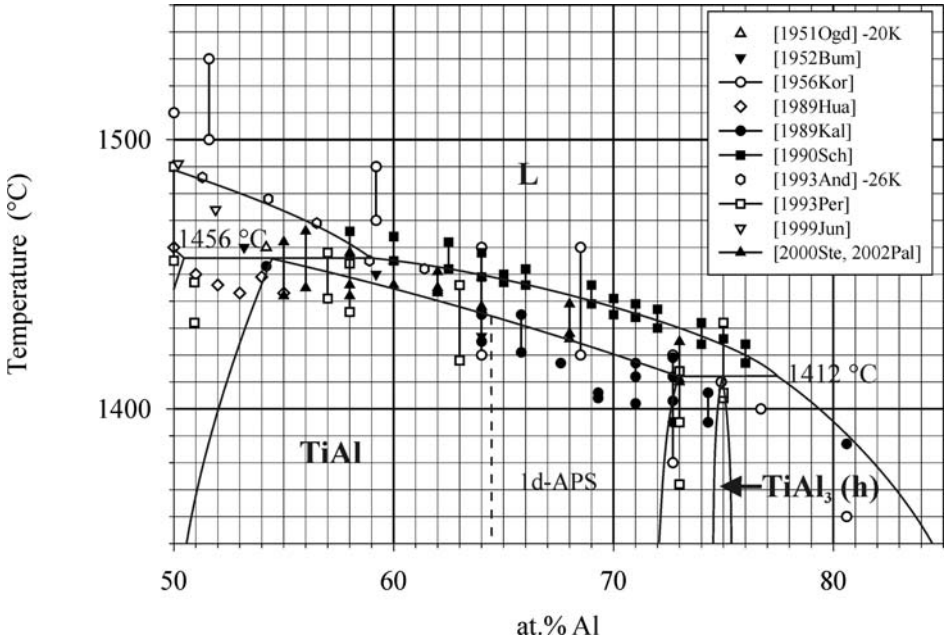

Fig. 8 Liquidus and solidus of the Al-rich part of the Ti-Al system. The data by [1993And] have been corrected by −26 K for reasons detailed in the text.

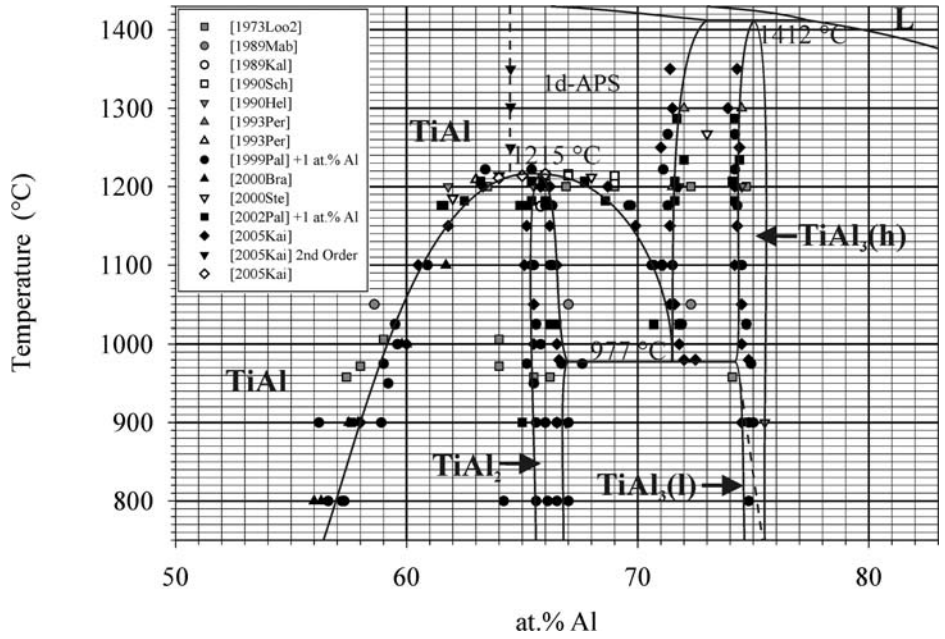

Fig. 9 Phase equilibria in the Al-rich part of the Ti-Al system. The data by [1999Pal, 2002Pal] have been corrected by +1 at.% Al because analyses in other Al-rich alloy systems have revealed such a shift, caused by a fault in the ZAF matrix correction procedure. Filled grey and black symbols denote compositions established by EPMA, and open symbols denote temperatures determined by DTA or DSC [2005Kai]. The possible existence of 1d-APS is discussed in Sec. 6.

6.2 TiAl/1d-APS ↔ $TiAl_2$ + $TiAl_3$(h) at 975 < T < 980 °C and 71.5, 67.0, 74.2 at.% Al

Below 1000 °C, TiAl/1d-APS decomposes by a eutectoid reaction into $TiAl_2$ + $TiAl_3$(h). From equilibrated samples, it has been shown that the reaction must take place above 975 °C [1999Pal, 2002Pal] and below 980 °C [2005Kai]. These values are in good agreement with the temperatures given by [1985Loi2, 1989Kal] (990 °C), [2001Bra] (995 °C), and [1990Sch] (970 < *T* < 1000 °C). The original estimate by [1965Ram] was ~950 °C. The value given by [1973Loo2] is considerably higher (1150 °C). This reaction has not been observed by DTA [1989Kal, 1990Sch, 2000Ste] due to its sluggishness [2002Pal].

The compositions for $TiAl_2$ and $TiAl_3$(h) at the invariant reaction are more or less fixed because both phases have only limited homogeneity ranges. The composition for TiAl/1d-APS is obtained by extrapolation of the $TiAl_2$ + 1d-APS/1d-APS and 1d-APS/1d-APS + $TiAl_3$(h) phase boundaries [1999Pal, 2001Bra, 2002Pal, 2005Kai]. This is in perfect agreement with all other reports [1965Ram, 1973Loo2, 1985Loi2, 1989Kal, 1990Sch].

6.3 $TiAl_3$(h) ↔ $TiAl_3$(l)

The occurrence of a low-temperature modification of $TiAl_3$ ($TiAl_3$(l), Ti_8Al_{24}) was first reported by [1973Loo1] and subsequently confirmed in [1983Rao, 2001Bra, 2002Pal]. Neither the homogeneity range nor the transformation temperatures have been fixed as data vary considerably. In the original description, a transition temperature of ~638 °C [1973Loo1] was given, which is in good agreement with $600 < T < 635$ °C [1983Rao]. In contrast, [2001Bra] reported ~950 °C for the Ti-rich phase boundary (74.5 at.% Al) and 735 °C on the Al-rich phase boundary (75 at.% Al).

7. Al-rich Section: Features not Adopted

7.1 $Ti_{1-x}Al_{1+x}$

A high-temperature phase having a very limited stability range labeled $Ti_{1-x}Al_{1+x}$ was first reported by [1990Sch]. The diffraction pattern of this phase is very similar to the pattern of TiAl in that it has identical peak positions. It differs only by reversed intensity ratios for pairs of peaks split due to deviation of TiAl from cubic symmetry [e.g., (200)/(002)]. Thus, in transmission electron microscopy (TEM) investigations in particular, $Ti_{1-x}Al_{1+x}$ might frequently have been mistaken for TiAl. [1994Bra1] confirmed the occurrence of that phase but proposed a different crystal structure and reported a much larger range of existence [2001Bra]. The existence of a phase with the latter structure would require the presence of a two-phase field between TiAl and $Ti_{1-x}Al_{1+x}$. Such a two phase field has not been found in diffusion couple studies between 1234 and 1350 °C [2002Pal, 2005Kai]. However, more data are needed to conclusively prove or disprove the existence of this phase.

7.2 Ti_3Al_5

The "phase" Ti_3Al_5 was first observed in Ti-63 at.% Al annealed at 700 °C [1982Mii]. Subsequently, it has frequently been observed in quenched samples and samples annealed at about 800 °C or below [e.g., 1996Nak2, 1999Sat, 2000Lei, 2001Koy, 2001Bra, 2004Doi]. TEM observations revealed that Ti_3Al_5 does not occur as a separate phase but forms islands or ordered domains within TiAl [e.g., 1985Loi1, 2002Nak]. On heating of Ti_3Al_5-containing samples, it is observed that the onset of the DTA signal due to the dissolution of Ti_3Al_5 depends strongly on the alloy composition [2000Ste, 2001Ste1]. No signal is observed on subsequent cooling or repeated annealing of the samples. Also, once the Ti_3Al_5 ordering has been removed by annealing above the transition temperature, it does not reappear after prolonged heating below the transition temperature [2001Ste1]. Ti_3Al_5 is therefore considered as a metastable state [2001Ste1, 2002Hay, 2002Inu] and should be labeled Ti_3Al_5(m). Additional ordered clusters have been reported [1985Loi1, 2002Hat, 2002Inu]. Ti_3Al_5 had already been considered by [1987Mur] as a nonequilibrium structure.

7.3 Ti_9Al_{23} and $TiAl_3$(m)

A phase Ti_9Al_{23} has been reported by [1965Ram]. It has a significantly smaller subcell volume than any of the binary Ti-Al phases [1965Ram, 1990Sch]. It has been shown that this phase is stabilized by silicon at the expense of $TiAl_3$ [1988Tak].

$TiAl_3$(m) is a metastable cubic compound that has been observed in rapidly solidified [1985Hor, 1993Maj, 1994Bra2] or mechanically alloyed [1991Sri] samples. Actual observed compositions are in excess of 75 at.% Al [1985Hor, 1993Maj], and the phase may be identical with other metastable (cubic) Al-rich phases that have been reported frequently [1973Oha, 1974Cis = 1974Ker, 1982Arn, 1983Sch].

8. Thermodynamic Data and Phase Diagram Modeling

Table 2 lists the experimental data available on thermodynamic properties. None of the CALPHAD type descriptions based on these available thermodynamic data [1970Kau, 1973Kau, 1978Kau, 1988Gro, 1988Mur, 1989Mis, 1992Kat, 1993Oeh, 1995Sau = 1996Sau = 1998Sau, 1997Zha, 2000Ohn, 2001Dan] matches the assessed phase diagram as a whole because all were modeled to fit earlier sets of the experimental data. There are, however, remarkable agreements between individual equilibria of the above assessed experimental phase diagram and the calculated ones.

9. Miscellaneous

Regarding topics not covered within this review, we would like to mention that diffusion data for Ti-rich alloys (including TiAl) have been comprehensively reviewed recently [2000Mis]. Diffusion data for the Al-rich phases were reported by [1973Loo1, 1973Loo2, 1980Lev, 1999Woe, 2005Kai]. Ab initio calculations (see Reference List C) certainly will play a prominent role in the future but are outside the scope of the current review.

Acknowledgments

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Short codes used in references: C, crystal structure; D, diffusivity data; E1, experimental equilibrium diagram, up to 56 at.% Ti, involving the liquid phase; E2, experimental equilibrium diagram, transus αTi/βTi; E3, experimental equilibrium diagram, formation of α_2Ti_3Al; E4, experimental equilibrium diagram, two phase regions αTi + α_2Ti_3Al, α/α_2Ti_3Al + γTiAl; E5, experimental equilibrium diagram, section 50-75 at.% Al; E6, experimental equilibrium diagram, solution of Ti in solid or liquid Al; E7, experimental thermodynamic data; M, thermodynamic modeling, equilibrium diagram; P, physical properties (thermal expansion, hardness etc.); T, calculated equilibrium diagram using first principle theories

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