CRITICAL ANALYSIS OF THE Ti-Al PHASE DIAGRAMS

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În lucrare este prezentată analiza critică termodinamică şi cristalochimică a celei mai importante diagrame binare de faze ale aliajelor de titan, diagrama Ti-Al. Au fost studiate diferite variante ale diagramei de echilibru şi s-a propus schema şi fluxul diagramei Ti-Al. Sunt caracterizate compoziţional şi structural toate fazele intermediare din diagrama de echilibru Ti-Al.

In this paper the authors performed a critical thermodynamic and crystallo-chemical analysis of the most important binary phase diagram of Ti alloys, Ti-Al diagram. Different variants of the diagrams are critically analyzed and we suggest a schema and a flow diagram of Ti-Al diagram. The composition and structure of all intermediate phases of the Ti-Al equillibrium diagram are characterized.

Keywords: Ti-Al binary phase diagram, assessment

Introduction

Ti-Al binary phase diagram (BPD) is the most important phase diagram of Ti alloys. Aluminium is as essential for alloying titanium as carbon is for iron. Aluminium is the most abundant metal in the earth's crust (8.8 %), and it has found large applications due to its low density (2.71 g/cm^3) and high corrosion resistance. Titanium is the seventh most abundant metal in the earth's crust (0.63 %) and the fourth most used material in the industry, after iron, aluminium and magnesium. Titanium is a very important metal, with large applications in aerospace industry, naval industry, automobile industry, medical engineering, fuel cells, chemical industry etc. There are many experimental and theoretical works on Ti-Al BPD, concerning the stable and metastable phases, the phase equilibria and the accuracy of the published papers.

In this paper we analyse the most cited Ti-Al BPD in order to point out the controversies between different authors and we propose a schema of Ti-Al built on mediation of existing data.

1. Critical analysis of some experimental and calculated Ti-Al BPD

The most controversial area of Ti-Al BPD ranges between 55 and 77 at. %

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Al, and 900 to 1450 ^{o}C.

In Fig. 1 it is presented the Ti-Al BPD, published in 1990 [1]. With dashed lines are indicated the estimated transformation curves. There are four intermetallic compounds with variable composition ($AlTi_3$, AlTi, Al_2Ti, δ), and one with constant composition (Al_3Ti).

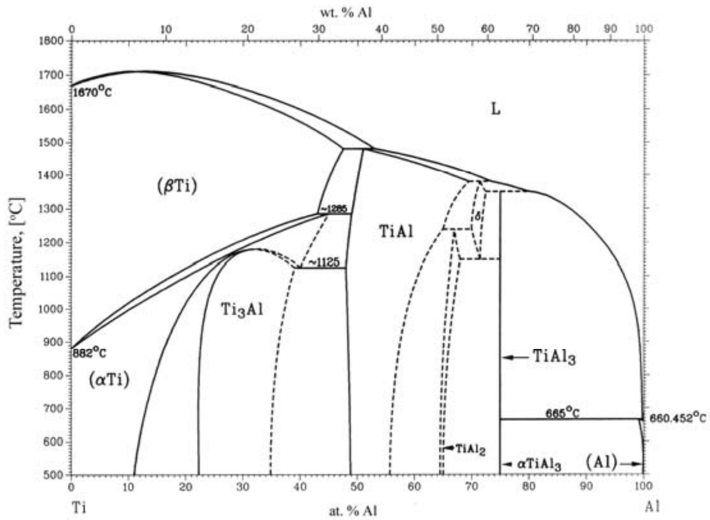


Fig. 1. The first variant of Ti-Al BPD [1].

At T≅1285°C and x_{Al}≅49 at. % the peritectoid transformation $(\beta_{Ti})_{0.43} + (AlTi)_{0.45} \leftrightarrow (\alpha_{Ti})_{0.49}$ occurs, but it was canceled in the latest works (they suggest the peritectic transformation $L + \beta_{Ti} \leftrightarrow \alpha_{Ti}$).

The liquidus and solidus curves of $L \leftrightarrow \beta_{Ti}$ show a maximum that in other works doesn't occur. The transformations of the intermetallic compounds are:

- peritectic transformations:

$$L_{0,53} + (\beta_{Ti})_{0.51} \xleftrightarrow{1480\,°C} (AlTi)_{0.475}$$

$$L_{0.735} + (AlTi)_{0.695} \xleftrightarrow{1380\,C} \delta_{0.715}$$

$$L_{0.8} + \delta_{0.725} \xleftrightarrow{1350\,°C} (Al_3Ti)_{0.75}$$

- peritectoid transformation: $(AlTi)_{0.65} + \delta_{0.725} \xleftrightarrow{1240\,°C} (Al_2Ti)_{0.67}$
- order-disorder transformation: $(\alpha_{Ti})_{0.309} \xleftrightarrow{1180\,°C} (AlTi_3)_{0.309}$

In Fig. 2 it is presented an experimental Ti-Al BPD [2].

Three intermetallic compounds with variable composition ($AlTi_3$, AlTi, Al_3Ti) and two with constant composition (Al_2Ti, Al_5Ti_2) can be identified.

The phase transformations are:

- melting:

$$L \xleftrightarrow{1670\,°C} \beta_{Ti}$$

$$L \xleftrightarrow{660.452\,°C} Al$$

- polymorphic transformation:

$$\beta_{Ti} \xleftrightarrow{882\,°C} \alpha_{Ti}$$

- five peritectic transformations:

$$L_{0.494} + (\beta_{Ti})_{0.448} \xleftrightarrow{1490\,°C} (\alpha_{Ti})_{0.473}$$

$$L_{0.551} + (\alpha_{Ti})_{0.514} \xleftrightarrow{1462.8\,°C} (AlTi)_{0.55}$$

$$L_{0.725} + (AlTi)_{0.665} \xleftrightarrow{1415.9\,°C} (Al_5Ti_2)_{0.714}$$

$$L_{0.791} + (Al_5Ti_2)_{0.714} \xleftrightarrow{1392.9\,°C} (Al_3Ti)_{0.75}$$

$$L_{0.999} + (Al_3Ti)_{0.75} \xleftrightarrow{664.2\,°C} Al_{0.994}$$

- peritectoid transformation:

$$(AlTi)_{0.645} + (Al_5Ti_2)_{0.714} \xleftrightarrow{1199.4\,°C} (Al_2Ti)_{0.667}$$

- two eutectoid transformation:

$$(\alpha_{Ti})_{0.396} \xleftrightarrow{1118.5\,°C} (AlTi_3)_{0.382} + (AlTi)_{0.467}$$

$$(Al_5Ti_2)_{0.714} \xleftrightarrow{990\,°C} (Al_2Ti)_{0.667} + (Al_3Ti)_{0.742}$$

- order-disorder transformation: $(\alpha_{Ti})_{0.309} \xleftrightarrow{1164\,°C} (AlTi_3)_{0.309}$

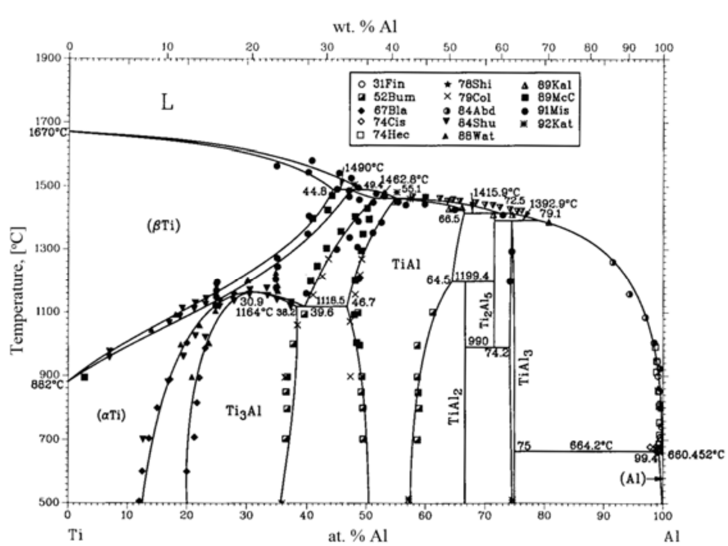


Fig. 2. The second variant of Ti-Al BPD [2].

In Fig. 3 the authors try to offer a new perspective of the most controversial area (indicated here as ξ domain, and detailed in Fig. 4). With dashed lines are indicated the hypothetic transformation curves.

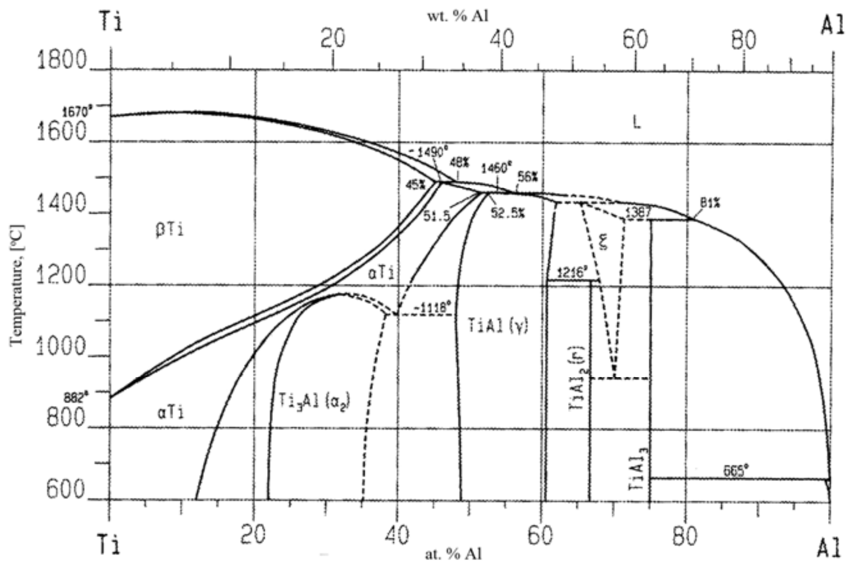


Fig. 3. The third variant of Ti-Al BPD [3].

There is also a maximum of $L \leftrightarrow \beta_{Ti}$ transformation curves. The intermetallic compounds with variable composition are $AlTi_3$ (α_2) and AlTi (γ), and the ones with constant composition are Al_2Ti and Al_3Ti. The Al_2Ti compound is considered stable up to 1216 °C, compared with 1199.4 °C, in other papers. Some experimental data that are also different from others are the peritectic transformations of Al_3Ti (1387 °C, versus 1392.5 °C in [2]) and AlTi (1460 °C, versus 1462.6 °C).

In Fig. 4 is presented the most controversial area of Ti-Al BPD.

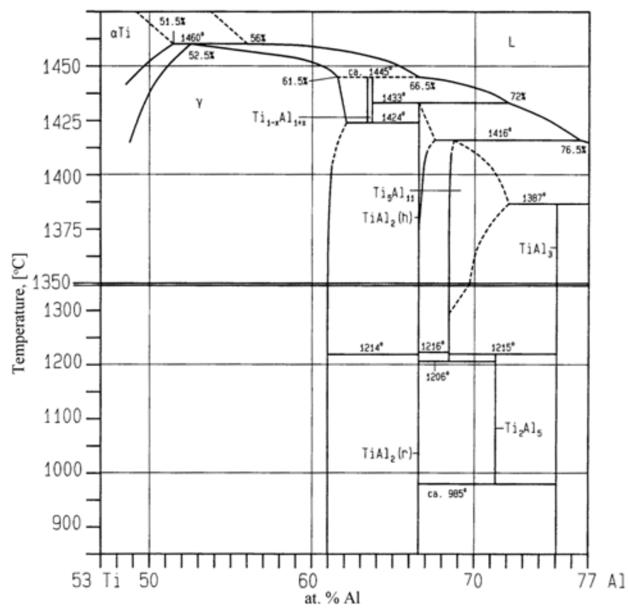


Fig. 4. The detalied ξ area of Ti-Al BPD.

The ξ domain is a particular point of view of the authors [3], but there are many mistakes in representing the transformations. The invariant transformations at 1214 °C and 1216 °C have only two points of intersection, and the ones at 1424 °C and 1445 °C have four points of intersection, instead of three, according with the thermodynamic rules. We consider that the metastable intermetallic compounds were introduced without a critical analyze.

The AlTi, $Al_{1+x}Ti_{1-x}$, Al_2Ti, $Al_{11}Ti_5$, Al_3Ti compounds are formed by peritectic transformations:

$$L_{0.56} + (\alpha_{Ti})_{0.515} \leftrightarrow \gamma_{0.525}$$
$$L_{0.665} + \gamma_{0.615} \leftrightarrow (Al_{1+x}Ti_{1-x})_{0.63}$$
$$L_{0.718} + (Al_{1+x}Ti_{1-x})_{0.645} \leftrightarrow (Al_2Ti)_{0.667}$$
$$L_{0.765} + (Al_2Ti)_{0.675} \leftrightarrow (Al_{11}Ti_5)_{0.691}$$
$$L_{0.791} + (Al_{11}Ti_5)_{0.72} \leftrightarrow (Al_3Ti)_{0.75}$$

The Al_5Ti_2 compound is formed by a peritectoid reaction:

$$(Al_{11}Ti_5)_{0.695} + (Al_3Ti)_{0.712} \leftrightarrow (Al_5Ti_2)_{0.712}$$

In Fig. 5 is accepted the existence of $Al_{23}Ti_9$ intermetallic compound. It is represented with dashed line, suggesting an estimation of its boundaries. The intermetallic compounds are: $AlTi_3$, AlTi, Al_2Ti, $Al_{11}Ti_5$, $Al_{23}Ti_9$, Al_3Ti and α-Al_3Ti. All but Al_3Ti have variable composition (Fig. 5.a, b).

In this BPD the maximum of $\beta_{Ti} \leftrightarrow L$ transformation is more emphasized.

The phase transformations are:
- order-disorder transformation: $AlTi_3 \xleftrightarrow{1177\,°C} \alpha_{Ti}$;
- polymorphic transformation: $\alpha_{Ti} \xleftrightarrow{882°C} \beta_{Ti}$;
- four peritectic transformations:

$$L + \alpha_{Ti} \xleftrightarrow{1457\,°C} AlTi;$$
$$L + AlTi \xleftrightarrow{1415\,°C} Al_{11}Ti_5;$$
$$L + Al_{11}Ti_5 \xleftrightarrow{1395\,°C} Al_3Ti;$$
$$L + Al_3Ti \xleftrightarrow{665\,°C} \varphi_{Al};$$

- two eutectoid transformation:

$$\alpha_{Ti} \xleftrightarrow{1148\,°C} AlTi_3 + AlTi;$$
$$Al_{11}Ti_5 \xleftrightarrow{990\,°C} Al_2Ti + Al_3Ti;$$

- two peritectoid transformations:

$$Al_{11}Ti_5 + AlTi \xleftrightarrow{1175\,°C} Al_2Ti;$$
$$Al_3Ti + Al_2Ti \xleftrightarrow{777\,°C} Al_{23}Ti_9$$

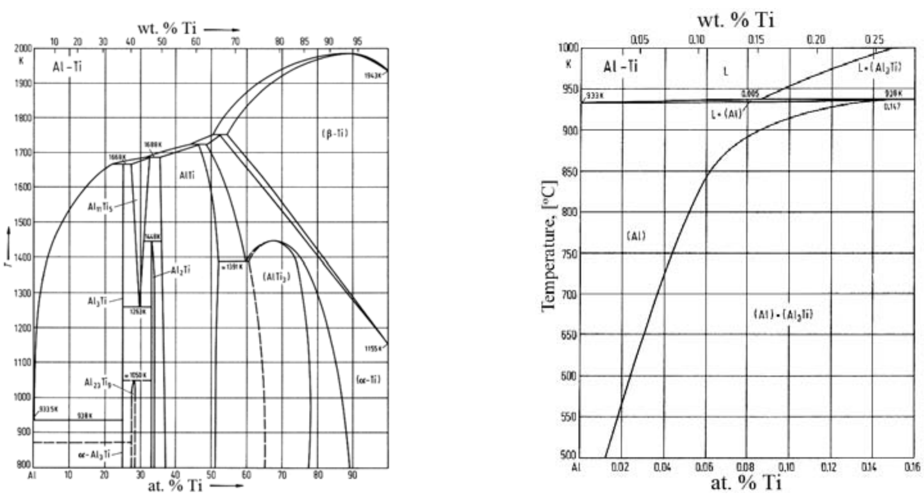


Fig. 5. a. The fourth variant of Al-Ti BPD.
b. Detail of the peritectic transformation $L+(Al_3Ti) \xleftrightarrow{665\,°C} (Al)$ [4].

In Fig. 6 a recent detail of the peritectic transformation $L+(Al_3Ti) \xleftrightarrow{665\,°C} (Al)$ is presented. Between Fig. 6 and Fig. 5.b there is a difference of 0.047 at. % Ti for the peritectic point.

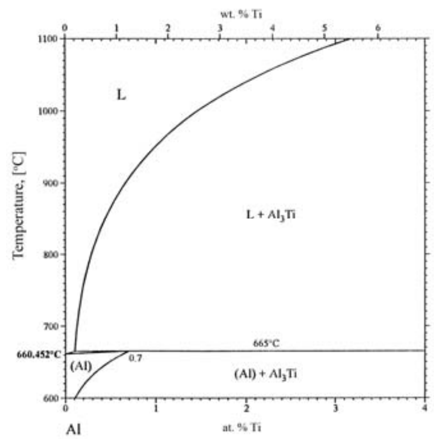


Fig. 6. The peritectic transformation $L+(Al_3Ti)\xleftrightarrow{665^{\circ}C}(Al)$ [5].

In Fig. 7 a calculated Ti-Al BPD is represented. The sublattice model was used for the thermodynamic calculation. The BPD is similar with the one from Fig. 5, but the $Al_{11}Ti_5$ compound is considered with constant composition and the $Al_{23}Ti_9$ compound as being metastable, hence it was not included in the calculations. The results are very close to the experimental ones.

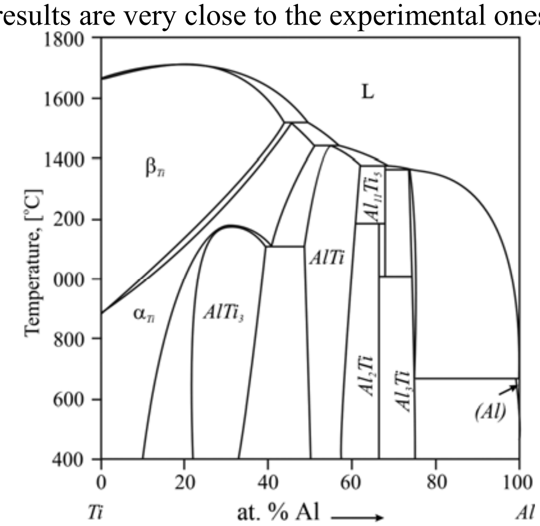


Fig. 7. The calculated Ti-Al BPD using the sublattice model.

2. Intermediate phases of Ti-Al BPD

The $AlTi_3$ and AlTi intermetallic compounds are largely accepted as having variable composition, with a wide homogeneity domain. The Al_2Ti and Al_5Ti_2 are accepted as having constant composition. The Al_3Ti is treated as an intermetallic compound with variable composition in some papers, and with constant composition in other papers. There are twelve intermetallic compounds recognized in the literature [6]. The $Al_{1+x}Ti_{1-x}$ (x=0.281) intermetallic compound can be assimilated with $Al_{16}Ti_9 \cong Al_{15}Ti_9 = Al_5Ti_3$, compound that satisfies most of the experimental results. Other four compounds do not appear in any of the presented BPD, though they occur between 0 and 50 at. % Al, hence we consider them as metastable phases.

In Table 1 there are summarized all the intermetallic compounds.

Table 1

Al_mTi_n intermetallic compounds (n_{af} – number of the atoms in the formula)

No.	Formula	*m/n*	x_{Al}	n_{af}
1.	Al_3Ti_{17}	0.176	0.150	20
2.	Al_6Ti_{19}	0.315	0.240	25
3.	$AlTi_3$	0.333	0.250	4
4.	$AlTi_2$	0.500	0.333	3
5.	Al_2Ti_3	0.666	0.400	5
6.	$AlTi$	1.000	0.500	2
7.	Al_5Ti_3	1.666	0.625	8
8.	Al_2Ti	2.000	0.666	3
9.	$Al_{11}Ti_5$	2.200	0.687	16
10.	Al_5Ti_2	2.500	0.714	7
11.	$Al_{23}Ti_9$	2.555	0.718	32
12.	Al_3Ti	3.000	0.750	4

The compositional characteristics of the Al_mTi_n intermetallic compounds are presented in Table 2.

Table 2

Compositional characteristics of the Al_mTi_n intermetallic compounds

No.	Formula of the compound	Stoichiometric composition, x_{Al}	Range of the homogeneity domain	Width of the homogeneity domain, Δx_{Al}	Degree of order, η_s	Melting character
1.	Al_3Ti_{17}	0.150	-	-	-	-
2.	Al_6Ti_{19}	0.240	-	-	-	-
3.	$AlTi_3$	0.250	0.2-0.382	0.182	0.818	-
4.	$AlTi_2$	0.333	-	-	-	-
5.	Al_2Ti_3	0.400	-	-	-	-
6.	$AlTi$	0.500	0.467-0.62	0.153	0.847	*Incongruent*
7.	Al_5Ti_3	0.625	0.628-0.645	0.017	0.983	*Incongruent*
8.	Al_2Ti	0.666	0.66-0.675	0.015	0.985	*Incongruent*
9.	$Al_{11}Ti_5$	0.687	0.685-0.72	0.035	0.965	*Incongruent*
10.	Al_5Ti_2	0.714	0.705-0.712	0.007	0.993	-
11.	$Al_{23}Ti_9$	0.718	0.73-0.76	0.03	0.97	-
12.	Al_3Ti	0.750	0.742-0.755	0.013	0.987	*Incongruent*

The characterization of the crystalline structure is presented in Table 3.

Table 3

Crystalline structure of the intermetallic compounds

No.	Intermetallic compound	Strukturbericht Symbol	Prototype	Pearson Symbol	Space Group	Lattice Parameters [Å]
1.	α_{Ti}	*A3*	*Mg*	*hP2*	$P6_3/mmc$	-
2.	β_{Ti}	*A2*	*W*	*cI2*	$Im\overline{3}m$	-
3.	Al_3Ti_{17}	-	*Mg*	*hP2 (?)*	$P6_3/mmc$	*a=2.925; c=4.667*
4.	Al_6Ti_{19}	-	*Mg*	*hP2 (?)*	$P6_3/mmc$	*a=2.900; c=4.645*
5.	$AlTi_3$	DO_{19}	Ni_3Sn	*hP8*	$P6_3/mmc$	*a=5.780; c=4.647*
6.	$AlTi_2$	-	Ni_3Sn	*hP8 (?)*	$P6_3/mmc$	*a=5.775; c=4.638*
7.	Al_2Ti_3	-	*Mg*	*hP2 (?)*	$P6_3/mmc$	*a=2.877; c=4.612*
8.	*AlTi*	$L1_o$	*AuCu*	*tP4*	*P4/mmm*	*a=4.001; c=4.071*
9.	Al_5Ti_3	-	-	*tP32*	*I4/mbm*	*a=4.0262; b=2.9617; c=4.0262*
10.	Al_2Ti	-	Ga_2Hf	*tI4(24)*	$I4_1/amd$	*a=3.976; c=24.36*
11.	$Al_{11}Ti_5$	-	Al_3Zr	*tI16*	*I4/mmm*	*a=3.917; c=16.524*
12.	Al_5Ti_2	-	Al_5Ti_2	*fI28*	-	*a=3.9053; c=2.9196*
13.	$Al_{23}Ti_9$	-	Al_3Ti	*(tetr.)*	-	*a=3.843; c=33.465*
14.	Al_3Ti	DO_{22}	Al_3Ti	*tI8*	*I4/mmm*	*a=3.846; c=8.594*
15.	φ_{Al}	*A1*	*Cu*	*cF4*	$Fm\overline{3}m$	*a=4.0496*

3. The schema of Ti-Al BPD

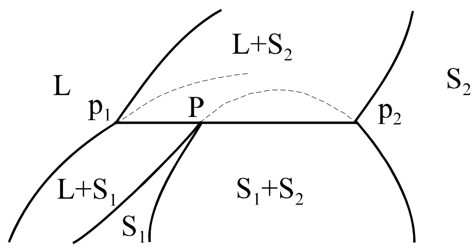
The proposed schema (Fig. 8) is a synthesis of all analyzed Ti-Al BPD, and it is a qualitative one, its purpose being to evidentiate all the curves, invariant transformation, and important points. The authors consider all the intermetallic compounds having variable composition.

Ti-Al BPD has the following main characteristics:

- total solubility in liquid state;
- partial solubility in solid state;
- three intermetallic compounds with incogruent melting, formed by peritectic transformations (AlTi, $Al_{11}Ti_5$, Al_3Ti);
- one intermetallic compound formed by peritectoid transformation (Al_2Ti);
- one intermetallic compound formed by order-disorder transformation ($AlTi_3$);

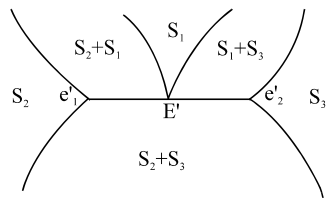
The phase transformations that take place in Ti-Al BPD are:

- five peritectic transformations:



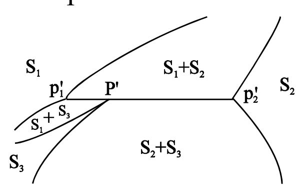
$$L_{p_2} + \left(\beta_{Ti}\right)_{p_1} \leftrightarrow \left(\alpha_{Ti}\right)_{P_1}$$
$$L_{p_4} + \left(\alpha_{Ti}\right)_{p_3} \leftrightarrow AlTi_{P_2}$$
$$L_{p_6} + AlTi_{P_5} \leftrightarrow (Al_{11}Ti_5)_{P_3}$$
$$L_{p_8} + (Al_{11}Ti_5)_{p_7} \leftrightarrow (Al_3Ti)_{P_4}$$
$$L_{p_{10}} + (Al_3Ti)_{p_9} \leftrightarrow (Al)_{P_5}$$

- two eutectoid transformations:



$$\left(\alpha_{Ti}\right)_{E'_1} \leftrightarrow (AlTi_3)_{e'_1} + (AlTi)_{e'_2}$$
$$(Al_{11}Ti_5)_{E'_2} \leftrightarrow (Al_2Ti)_{e'_3} + (Al_3Ti)_{e'_4}$$

- one peritectoid transformation:



$$(AlTi)_{p'_1} + (Al_{11}Ti_5)_{p'_2} \leftrightarrow (Al_2Ti)_{P'}$$

- one order-disorder transformation:

$$\left(\alpha_{Ti}\right)_{D'} \leftrightarrow (AlTi_3)_{D'}$$

- one polymorphic transformation:

$$Ti_{\alpha} \leftrightarrow Ti_{\beta}; \quad \alpha_{Ti} \leftrightarrow \beta_{Ti}$$

The coordinates of the intersection points of Ti-Al shema are presented in Table 4.

Table 4

The coordinates of the intersection points of the Ti-Al schema

Symbol	Signifiance	X_{Al}, [% at.]	T, [°C]	T, [K]
T_{Al}	Melting temperature of Al	*100*	*660.452*	*933.452*
p_{10}	Transition point	*99.90*	*665*	*938*
P_5	Peritectic point	*99.30*	*665*	*938*
p_8	Transition point	*77.46*	*1395*	*1668*
p_9	Lyotectic point	*75.00*	*665*	*938*
P_4	Peritectic point	*74.39*	*1395*	*1668*
p_7	Lyotectic point	*72.20*	*1395*	*1668*
p_6	Transition point	*67.80*	*1415*	*1688*
e'_4	Lyotectoid point	*74.65*	*990*	*1263*
E'_2	Eutectoid point	*69.76*	*990*	*1263*

P_3	Peritectic point	*67.30*	*1415*	*1688*
p_2'	Lyotectoid point	*68.63*	*1175*	*1448*
e_3'	Lyotectoid point	*66.70*	*990*	*1263*
P'	Peritectoid point	*66.50*	*1175*	*1448*
p_5	Lyotectic point	*64.20*	*1415*	*1688*
p_1'	Lyotectoid point	*63.81*	*1175*	*1448*
p_4	Transition point	*55.10*	*1462.8*	*1735.8*
p_2	Transition point	*49.4*	*1490*	*1763*
P_2	Peritectic point	*55.00*	*1462.8*	*1735.8*
p_3	Lyotectic point	*51.40*	*1462.8*	*1735.8*
P_1	Peritectic point	*47.30*	*1490*	*1763*
p_1	Lyotectic point	*44.80*	*1490*	*1763*
e_2'	Lyotectoid point	*46.70*	*1118.5*	*1391.5*
E_1'	Eutectoid point	*39.60*	*1118.5*	*1391.5*
e_1'	Lyotectoid point	*38.20*	*1118.5*	*1391.5*
D'	Order-disorder point	*30.90*	*1164*	*1437*
D	Melting temperature of β_{Ti}	*11.00*	*1710*	*1983*
T_{P1}	Polymorphic transformation temperature $Ti_\alpha \leftrightarrow Ti_\beta$	*0.00*	*882*	*1155*
T_{Ti}	Melting point of Ti	*0.00*	*1670*	*1943*

The curves and the horizontal lines of the invariant transformations of Ti-Al schema are (Fig. 8):

- liquidus curve $T_{Ti}Dp_2p_4p_6p_8p_{10}T_{Al}$, formed of 6 curves, corresponding to the six phases that separate from the liquid state:
  - the liquidus of β_{Ti}, $T_{Ti}Dp_2$;
  - the liquidus of α_{Ti}, p_2p_4;
  - the liquidus of AlTi, p_4p_6;
  - the liquidus of $Al_{11}Ti_5$, p_6p_8;
  - the liquidus of Al_3Ti, p_8p_{10};
  - the liquidus of (AlTi), $p_{10}T_{Al}$;
  - solidus curve $T_{Ti}Dp_1P_1p_3P_2p_5P_3p_7P_4p_9P_5T_{Al}$;
- solvus curves:
  - $e_1'f_3$, normal solvus (AlTi phase precipitates);
  - $e_2'f_4$, normal solvus ($AlTi_3$ phase precipitates);
  - p_5p_1', normal solvus ($Al_{11}Ti_5$ phase precipitates);
  - $p_1'f_5$, normal solvus (Al_2Ti phase precipitates);
  - $P'e_3'$, retrograde solvus (AlTi phase is disolved);
  - $P'f_6$, retrograde solvus ($Al_{11}Ti_5$ phase is disolved);

- e'_3f_7, normal solvus (Al_3Ti phase precipitates);
- $P_3p'_2$, retrograde solvus (AlTi phase is disolved);
- $p_7E'_2$, normal solvus (Al_3Ti phase precipitates);
- e'_4f_8, normal solvus (Al_2Ti phase precipitates);
- p_9f_9, normal solvus ((Al) phase precipitates);
- P_5f_{10}, normal solvus (Al_3Ti phase precipitates);
- $p_3E'_1$, normal solvus (AlTi phase precipitates);
- $P_2e'_2$, retrograde solvus (α_{Ti} phase is disolved);

- two polymorphic curves $T_{p_1}p_1$ and $T_{p_1}P_1$, corresponding to the beginning and the ending of the Ti polymorphic transformation ($\alpha_{Ti} \leftrightarrow \beta_{Ti}$);

- ordinus curves:
  - $f_1D'E'_1$, superior ordinus;
  - $f_2D'e'_1$, inferior ordinus;

- horizontal lines:
  - the horizontal lines of peritectic transformation: $p_1P_1p_2$, $p_3P_2p_4$, $p_5P_3p_6$, $p_7P_4p_8$, $p_9P_5p_{10}$;
  - the horizontal lines of eutectoid transformations: $e'_1E'_1e'_2$, $e'_3E'_2e'_4$;
  - the horizontal line of peritectoid transformation: $p'_1P'p'_2$.

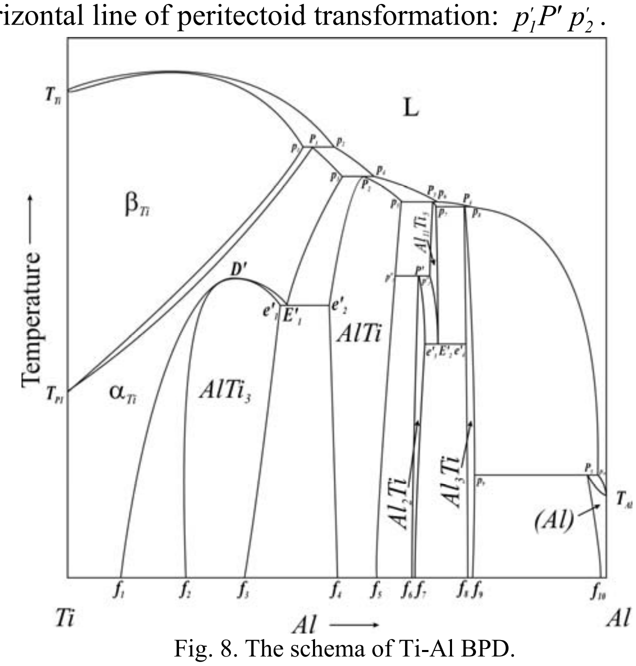


Fig. 8. The schema of Ti-Al BPD.

3. The flow diagram of Ti-Al BPD

The flow diagram of Ti-Al BPD was built based on the Ti-Al schema and according with Gibbs phase rule. The flow diagram has 8 rectangles that represent the triphasic equilibria, connected by lines that represent the biphasic equilibria.

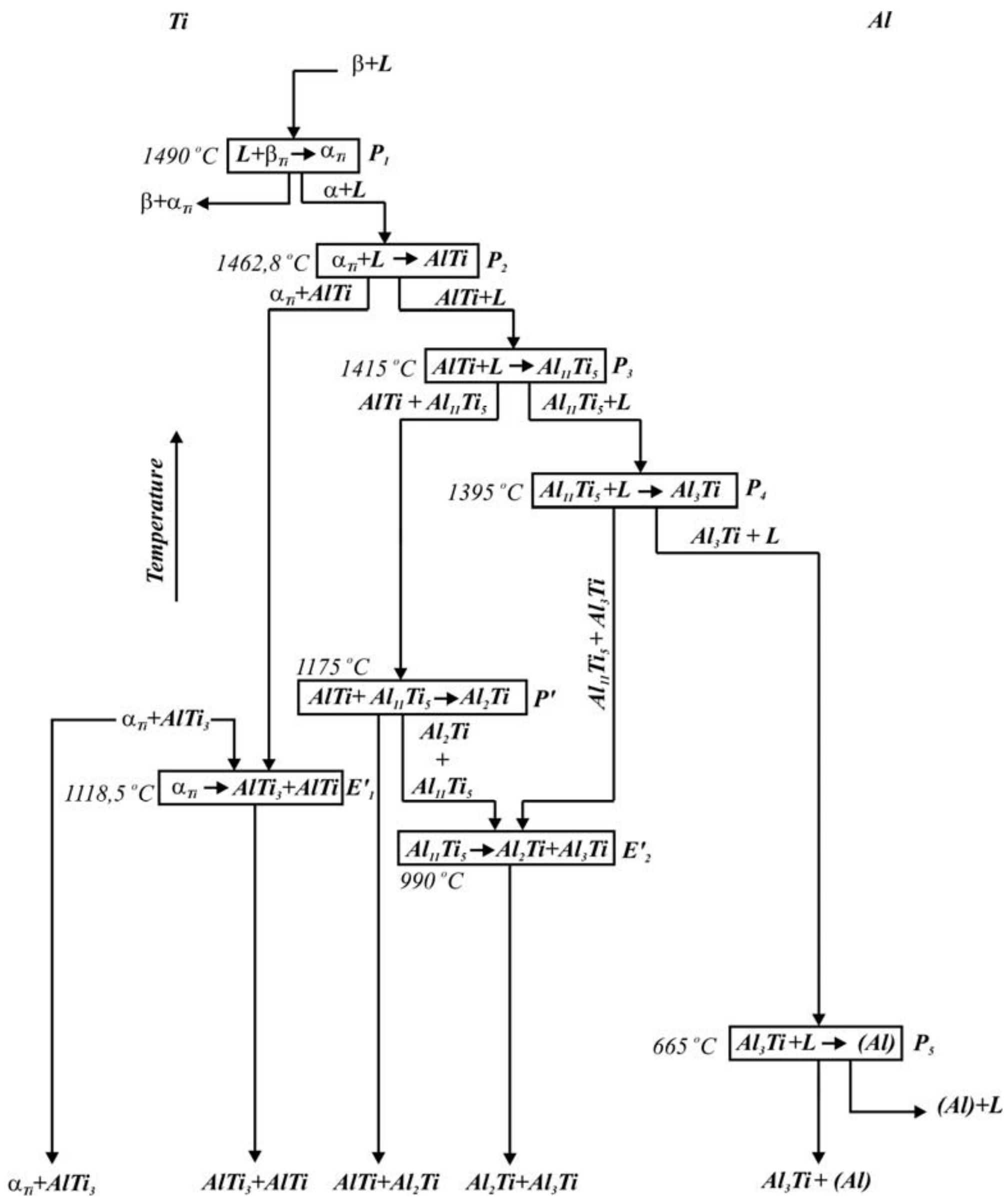


Fig. 9. The flow diagram of Ti-Al BPD.

Conclusions

1. Ti-Al BPD is the most important phase diagram of Ti alloys, and new assessments are necessary to be done, especially based on new and more accurate experimental works.
2. Though the Ti-Al BPD has been intensively studied we can not consider any of the published diagrams fully reliable. The published Ti-Ni BPDs should be critically analyzed, before using them for any practical or theoretical applications.
3. By our critical analysis of some Ti-Al BPD we suggest a general schema, considering all the intermetallic compounds with variable composition.
4. A synthesis of all important points and curves from the Al-Ni BPD was realized in our work, indicating also their coordinates (Table 4).
5. Based on our suggested Ti-Al schema we built the flow diagram (Fig. 9), that allows to establish the correct succession of biphasic and triphasic equilibria.
6. Ti-Al is also an essential part of the Ti-Ni-Al ternary phase diagram, and it shall be used for the ternary phase diagram assessment.
7. Through our assessment we pointed out that many published works regarding the Ti-Al BPD have many controversial results and even some unacceptable mistakes.

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