STRUCTURAL AND CONSTITUTIONAL STABILITY OF INTERMETALLIC Ni-Al ALLOY AFTER ISOTHERMAL ANNEALING

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Metallurgically prepared intermetallic Ni-Al alloy was remelted by an unidirectional solidification method and then submitted to the isothermal annealing at the temperature of 1373 K for technically significant times up to 500 hours. The constitutional stability and the structure evaluation of the alloy are given for the casting as well as the unidirectional state.

ŠTRUKTÚRNA A KONŠTITUČNÁ STABILITA INTERMETALICKEJ ZLIATINY Ni-Al PRI IZOTERMICKOM ŽÍHANÍ

Na vzorkách intermetalickej zliatiny Ni-Al, pripravenej metalurgickou cestou a následne pretavenej metódou usmernenej kryštalizácie, sme aplikovali izotermické žíhanie pri teplote 1373 K pre technicky významné časy až do 500 hodín. Uvádzame štruktúrne hodnotenie a konštitučnú stabilitu zliatiny pre liaty a pre usmernený stav.

1. Introduction

The reason why intermetallic nickel base phases with aluminium addition (predominantly NiAl and Ni_3Al aluminides) are of a great research interest is their occurrence almost in all superalloys. On the other hand, they can be found as matrix phases in composite materials, as well. These intermetallics are interesting for the high temperature application due to their high strength, stiffness, creep properties and fatigue resistance. Because of a quite high concentration of aluminium that influences the above mentioned properties, these alloys have also a good corrosion resistance and sufficient high temperature oxidation resistance.

The morphology of binary Ni-Al intermetallic compounds varies not only with nickel or aluminium content, but with heat treatment, as well. According to the literature [1, 2], Ni-rich NiAl phase with cubic B2 structure transforms to martensitic NiAl with tetragonal $L1_0$ structure. At defined conditions [3], the martensitic NiAl can transform to Ni_5Al_3 or Ni_2Al phases.

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The aim of this work was to evaluate the microstructure of intermetallic Ni-Al compounds after isothermal annealing at 1373 K up to 500 hours. Since the directional solidification significantly influences the toughness of this type of intermetallics [4], the microstructure was studied for both the as-cast and directionally solidified samples.

2. Experimental procedures

Intermetallic Ni-Al compounds were metallurgically prepared by vacuum induction melting. The charge of high purity metals was weighted in stochiometric ratio 3 : 1 of nickel to aluminium, respectively, taking into account possible overburning of aluminium during melting. The intermetallic compounds were cast into cylindrical alumina moulds with the inside diameter of 12×10^{-3} m. Directional solidification was performed at constant growth rate of 2.5×10^{-6} m s^{-1} and constant temperature gradient at the solid/liquid interface $G_L = 11 \times 10^3$ K m^{-1}.

The as-cast and directionally solidified specimens were submitted to isothermal annealing at the temperature of 1373 K in air. A semi-automatic annealing equipment with accuracy of ± 2K/24 hours was used. The temperature of the furnace was measured using a thermocouple. The specimens were annealed during 10, 50, 100 and 500 hours with consequent evaluation of microstructure. For structural observation the samples were chemically etched in 20 ml H_2O + 20 ml HNO_3 + + 10 ml HCl solution.

Optical and electron microscopy was used to study the microstructure. The phase analysis was performed by transmission electron microscopy (TEM). Quantitative microanalysis of the elements in coexisting phases was performed by the electron microscope JEM 100 C with an energy-dispersive spectrometer KEVEX DELTA 4.

3. Results and discussion

According to the binary phase diagram shown in Figure 1, Ni_3Al phase is stable in a very narrow area [5]. Hence, some intermetallic phases with high melting temperature can be formed during the melting of charge. Table 1 summarizes the crystal structures of Ni-rich intermetallic phases that may be formed in metallurgically prepared ingots.

Figure 2a shows that metallurgically prepared as-cast Ni-Al alloy was not monophase. Its microstructure consisted of thin needles with composition corresponding to Ni_3Al. They were distributed in the interdendritic region of the matrix. The matrix as a whole showed the ratio of nickel to aluminium being greater than stochiometric 1 : 1, but lower than 5 : 3. Some defects like cavities and contractions were observed in the as-cast samples. These defects were the products of casting and fast cooling.

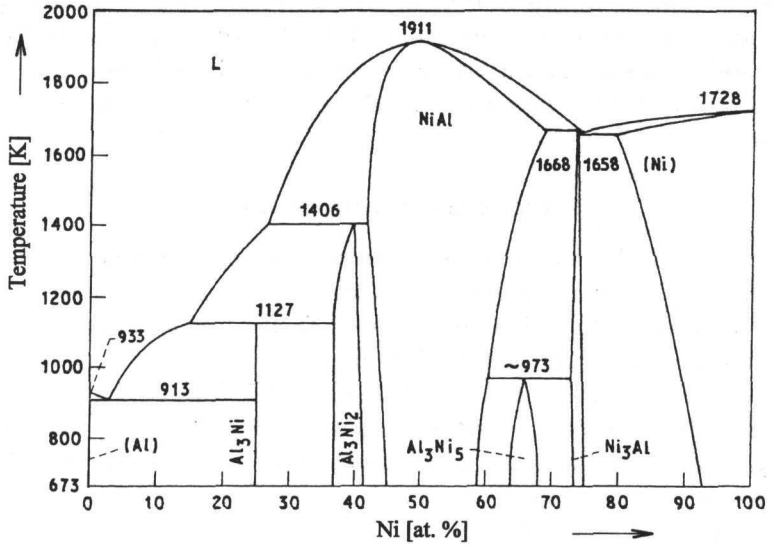

Fig. 1. Binary Ni-Al phase diagram.

Table 1. Summary of the intermetallic crystal structures in Ni-rich part of binary phase diagram

Phase	Lattice Parameter [nm]	Structure Type	Space Group
NiAl	$a = 0.287$	B_2 (CsCl)	Pm3m
NiAl Martensite	$a = 0.383$ $c = 0.320$	$L1_0$ (AuCu)	P4/mmm
Ni_3Al	$a = 0.358$	$L1_2$ (Cu_3Au)	Pm3m
Ni_5Al_3	$a = 0.753$ $b = 0.661$ $c = 0.376$	Pt_5Ag_3	Cmmm
Ni	$a = 0.352$	Al(Cu)	Fm3m

Figure 2b shows that the microstructure of Ni-Al alloy was morphologically different after directional solidification. The directional solidification caused coarsening of Ni_3Al needles and eliminated the casting defaults. This method controls the grain size in dependence on crystallisation growth rate, as well.

In order to determine coexisting phases in the structure of Ni-Al intermetallic compounds, TEM analysis was done. This method was applied to all specimens. Figure 3a shows a coarsened particle in a directionally solidified specimen. Figure 3b

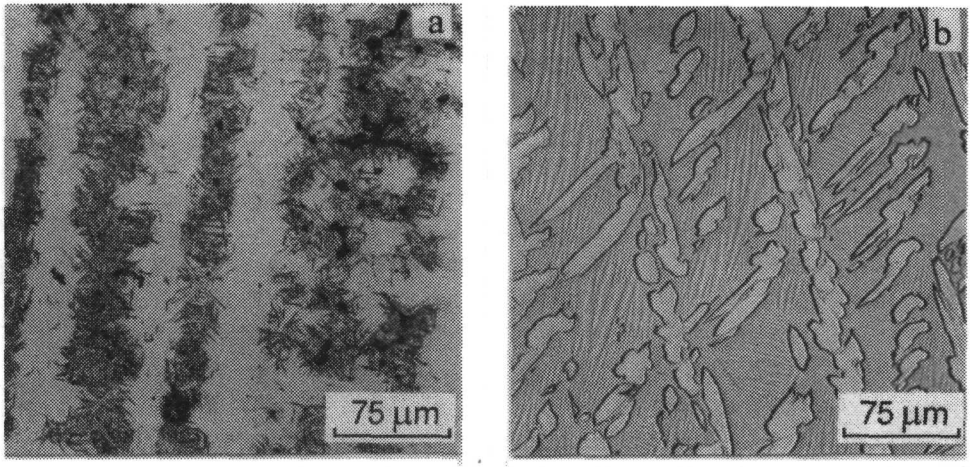

Fig. 2. Optical micrograph of cross section of a) as-cast Ni-Al alloy showing Ni_3Al needles distributed in Ni-Al matrix and dark cavities; b) directionally solidified state of Ni-Al alloy.

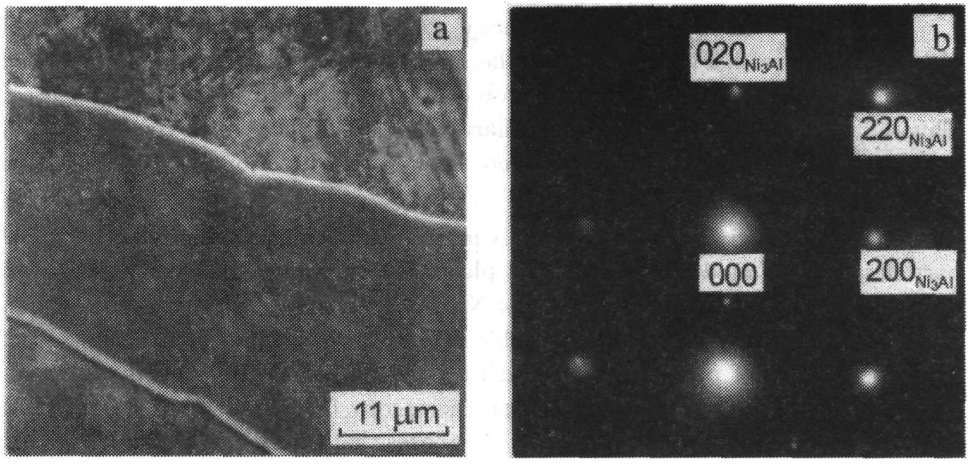

Fig. 3. a) TEM micrograph of Ni_3Al particle in NiAl matrix, directionally solidified specimen; b) selection electron diffraction pattern from Ni_3Al particle, $\langle 001 \rangle$ zone axis.

shows an electron diffraction pattern from this particle corresponding to the cubic face-centred lattice with $L1_2$ structure of Ni_3Al phase. Similar diffraction patterns of Ni_3Al phase were also taken from the as-cast specimens.

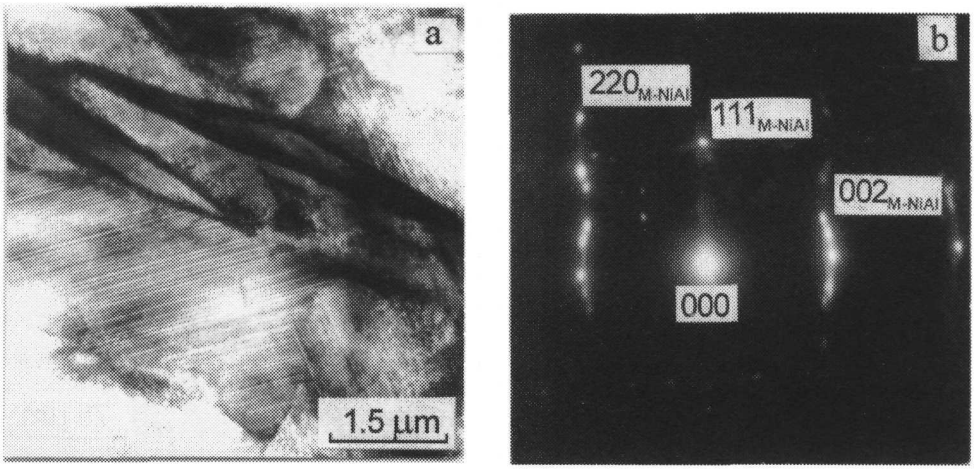

Fig. 4. a) TEM micrograph of martensitic NiAl lamellas in the matrix of directionally solidified specimen after 500 hours of annealing; b) electron diffraction pattern from selected area of martensitic NiAl matrix, ⟨110⟩ zone axis.

In both the as-cast and directionally solidified samples a martensitic NiAl phase was observed. Figure 4a shows the morphology of martensitic plates in the NiAl matrix. Figure 4b shows an electron diffraction pattern taken from this area. This diffraction reveals reflections of martensitic NiAl phase with tetragonal $L1_0$ crystal structure. Martensitic plates were finer in the directionally solidified than in the as-cast specimens.

The NiAl matrix was not completely martensitic. At close vicinity of the Ni_3Al particles a cubic modification of NiAl phase was identified. Figure 5a shows an interface between cubic and martensitic NiAl phases. Figure 5b shows an electron diffraction pattern taken from the cubic NiAl phase with B2 crystal structure. This coexistence of cubic and martensitic NiAl phase can be seen in the cross sections of Ni-Al compounds by optical microscopy (Figure 2). A detailed TEM analysis revealed neither Ni_5Al_3 nor Ni_2Al phases that might occur in Ni-rich NiAl intermetallic alloy. In accordance with work [3] a stable Ni_5Al_3 phase is created by ageing at temperatures lower than 973 K.

Annealed and unannealed samples consisted of Ni_3Al particles distributed in a NiAl matrix. Ni_3Al phase preferentially precipitated on the grain boundaries. However, the precipitation of Ni_3Al was also observed within the grains. The longer time of annealing the more coarse are the Ni_3Al particles. Thin needles of Ni_3Al phase of as-cast state coarsened with the annealing time. The longest time of annealing (500 hours) caused creation of large islands of Ni_3Al phase in the NiAl matrix. Contrary

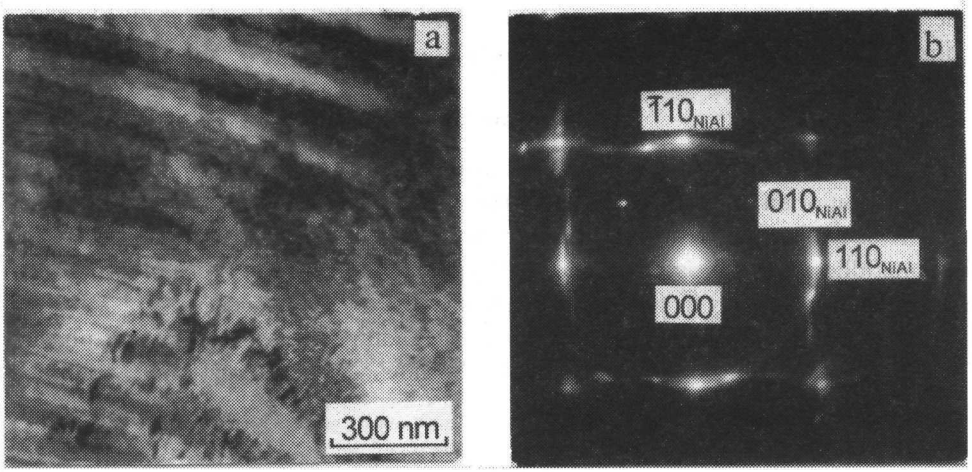

Fig. 5. a) TEM micrograph of martensitic/cubic NiAl interface in as-cast specimen after 500 hours of annealing; b) selected electron diffraction pattern from cubic NiAl phase, ⟨001⟩ zone axis.

Table 2. Quantitative microanalysis of elements in Ni-Al intermetallic alloy before and after annealing at 1373 K

Annealing at 1373 K	State	Element	NiAl matrix [wt.%]	Ni_3Al [wt.%]	k
0 hrs.	As-cast	Al	21.28	15.76	1.35
		Ni	78.72	84.24	0.93
	Directionally solidified	Al	20.46	15.05	1.36
		Ni	79.54	84.95	0.94
500 hrs.	As-cast	Al	21.82	15.82	1.38
		Ni	78.18	84.18	0.93
	Directionally solidified	Al	21.57	15.87	1.36
		Ni	78.43	84.13	0.93

to as-cast specimens, the evolution of Ni_3Al coarsening was different. During longer annealing time some of the Ni_3Al particles began to dissolve, while others – of the size above the critical – coarsened.

Table 2 summarises the results of quantitative microanalysis of the elements in coexisting phases in as-cast and directionally solidified Ni-Al compounds before and after annealing. The coefficient k represents a ratio of the element content in

the NiAl matrix to the element content in the Ni_3Al particles. From Table 2 the constitutional stability of investigated alloy is evident.

4. Conclusion

The method of unidirectional solidification does not change the origin phase composition of an alloy and eliminates the casting defects.

After isothermal treatment the morphology of metallurgically prepared Ni-Al intermetallic alloy consists of the Ni_3Al cubic phase of $L1_2$ type and the NiAl matrix that was found in two modifications: cubic and martensitic.

Isothermal annealing does not change the composition of coexisting phases. The Ni_3Al phase is prone to coarsening for both as-cast and directionally solidified state.

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