IMPACT OF CONDITIONS OF DIRECTIONAL CRYSTALLISATION BY BRIDGMAN METHOD ON PHYSICAL AND METALLURGICAL CHARACTERISTICS OF Ni_3Al

Malcharcziková J.[1], Kursa M.[1], Beljajev I. V.[2]
[1]Department of Non-ferrous metals, Refining and Recycling, Faculty of Metallurgy and Materials Engineering, VŠB -Technical University of Ostrava, 17. listopadu 15/2172, CZ 708 33 Ostrava, Czech Republic;
[2]MAGNETON, Vladimir, Russia
e-mail: jitka.malcharczikova@vsb.cz, miroslav.kursa@vsb.cz

VLIV PODMÍNEK SMĚROVÉ KRYSTALIZACE BRIDGMANOVOU METODOU NA FYZIKÁLNĚ-METALURGICKÉ CHARAKTERISTIKY Ni_3Al

Malcharcziková J.[1], Kursa M.[1], Beljajev I. V[2].
[1]Katedra neželezných kovů, rafinace a recyklace, Fakulta metalurgie a materiálového inženýrství,
VŠB – Technická univerzita Ostrava, 17. listopadu 15/2172, 708 33 Ostrava, Česká Republika;
2MAGNETON, Vladimir, Rusko
e-mail: jitka.malcharczikova@vsb.cz, miroslav.kursa@vsb.cz

Abstrakt

Pro měření byly použity vzorky slitin Ni-Al o různém chemickém složení. Vzorky byly připraveny odlitím ve vakuové indukční peci. Odlitky ve formě válečků o průměru 10 mm a délce 95 mm byly dále směrově krystalizovány Bridgmanovou metodou s vertikálním uspořádáním na zařízení ve Vladimiru v Rusku. Odlitky byly umístěny v elektrokorundových trubicích se specifikovaným vrcholovým úhlem. Byly použity různé rychlosti směrové krystalizace pro ověření nejvhodnějších podmínek pro získání homogenní a monokrystalické struktury. Vhodné vzorky byly upraveny soustružením na tahové tyče a ty byly následně použity pro tahové zkoušky s měřením akustických emisí. Pro tyto zkoušky byly použity krátké tahové tyče s kruhovým průřezem o délce 55 mm s průměrem střední části tyče 5 mm. Při všech měřeních byla pozorována intenzivní akustická emise s charakteristickou závislostí na čase a korelací s jednotlivými stádii deformace. U všech měření je možno rozlišit několik oblastí na tahovém diagramu a charakteristiky akustické emise tomu odpovídají. Detailně byly prostudovány vzniklé lomové plochy. Na příčném průřezu byla určena orientace zrn a mikroanalýza chemického složení. Pro přesné určení chemického složení byla provedena energiově disperzní mikroanalýza a to jednak pomocí zpětně odražených elektronů i pomocí sekundárních elektronů. Pro určení orientace získaných zrn byla použita rentgenová difrakce metodou zpětného odrazu. Na příčných i podélných průřezech bylo provedeno metalografické vyhodnocení. Některé vzniklé struktury jsou velmi homogenní, což potvrdila i rentgenová analýza.

Abstract

Samples of Ni-Al alloys of various chemical composition were used for measurement. The samples were prepared by casting in vacuum induction furnace. Cast rollers with diameter 10 mm and length 95 mm were afterwards directionally crystallised by Bridgman method with

vertical arrangement on special equipment in Vladimir (Russia). The cast pieces were inserted into electro-corundum tubes with specified apex angle. Various rates of directional crystallisation were used for verification of the most suitable conditions for obtaining of homogenous and mono-crystalline structure. Suitable samples were adjusted by turning to tensile rods, which were then used for tensile tests with measurement of acoustic emissions. For these tests there were used short tensile rods of round section with length 55 mm and with diameter of central part of the rod 5 mm. During all measurements there was monitored intensive acoustic emission with characteristic dependence on time and with correlation to individual stages of deformation. In all measurements it is possible to discern several areas on tensile diagram and characteristics of acoustic emission correspond with it. The formed fracture surfaces were investigated in detail. Grain orientation and micro-analysis of chemical composition were determined on cross section. Energy dispersive micro-analysis was used for precise determination of chemical composition, both with back-scattered electrons and with secondary electrons. X-ray diffraction with reflection method was used for determination of orientation of obtained grains. Metallographic evaluation was made on cross-sections and longitudinal sections. Some of the formed structures are highly homogenous, which was confirmed also by X-ray analysis.

Keywords: Ni-Al based intermetallic compounds, tensile tests, acoustic emission, grain orientation

1. Introduction

Ni-Al based intermetallic compounds are very interesting from the physical-metallurgical viewpoint. Their application in demanding environments, particularly at increased and high temperatures under oxidation atmosphere is of great importance. Directional crystallisation gives a possibility of influencing the structure and properties of the final product. This process makes it possible to control formation of shrinks, to influence grain size and manner of their growth [1]. Directional crystallisation by Bridgman method is one of the simplest and most frequently used methods. The tube with charge in its bottom part is situated into thermal field of furnace. During slow passage of the tube with melted metal through steep temperature gradient there occurs progressive directional crystallisation of molten metal and thus growth of crystal [2].

2. Preparation of samples

Cast pieces were used for experimental purposes. The cast pieces were prepared in vacuum induction furnace LEYBOLD - type IS3/1. Prior to the melting there were performed 2 vacuum treatments, namely below the value of 0.04 mbar with use of two-stage pumping by rotary and Roots pumps. Several basic samples were cast with composition of 25, 24.5 and 24 at. % Al in the form of rollers with length of 95 mm and diameter of 10 mm. Melting was realised in a corundum crucible and casting was made into graphite ingot moulds.

3. Directional crystallisation by bridgman method with use of crucibles with specified apex angle

The series of samples was used for directional crystallisation by Bridgman method on the equipment in Vladimir in Russia. Special corundum crucibles with an apex angle 60-70°

were used for these samples. The Table 1 gives composition of the used samples and speed of directional crystallisation. The Figure 1 shows a detail of the tip of the sample 139.2 after directional crystallisation.

Table 1 Composition of samples and used speed of directional crystallisation

Sample No.	Speed of directional crystallisation [mm/hour]	Ni contents [at.%]	Al contents [at.%]
139.1	108	75	25
139.2	60	75	25
139.3	18	75	25
141.1	108	76.5	24.5
141.2	60	76.5	24.5
141.3	18	76.5	24.5
143.1	105	76	24
143.2	18	76	24
143.3	60	76	24

Fig.1 Detail of the tip of the sample

3.1 Metallographic evaluation of structures, micro-analysis of chemical composition

The Figures 2, 4 and 6 show micro-structures of on cross sections of the samples Nos. 139.1, 139.2 and 139.3 in central part of the sample. This structure is highly homogenous, which was confirmed also by X-ray analysis. The Figures 3, 5 and 7 show macro-structures of these samples.

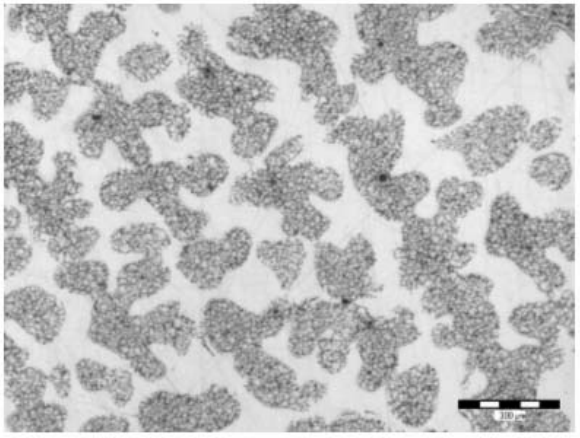

Fig.2 Sample No. 139.1, cross-section, Z=50x

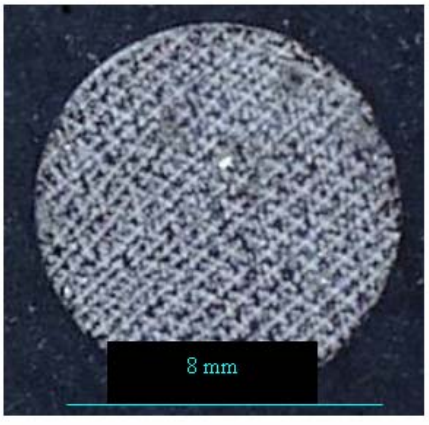

Fig.3 Sample No. 139.1

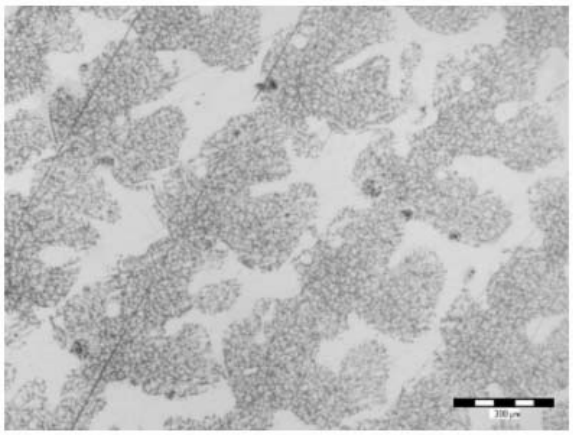

Fig.4 Sample No. 139.2, cross section, Z=50x

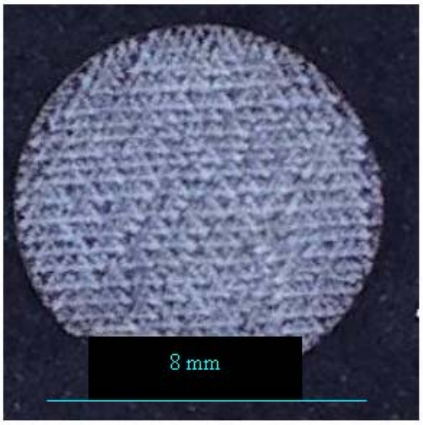

Fig.5 Sample No. 139.2

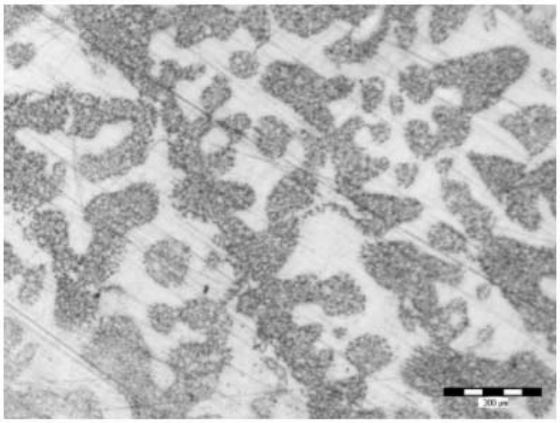

Fig.6 Sample No. 139.3, cross section, Z=50x

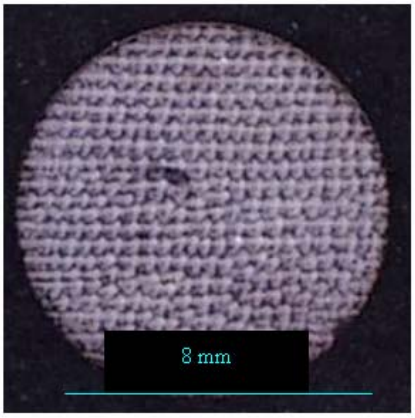

Fig.7 Sample No. 139.3

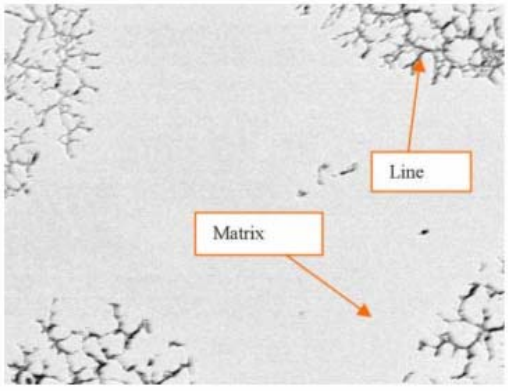

Fig.8 EDX, photographed in back-scattered electrons, 139.1 S, detail of matrix, magn. 940x

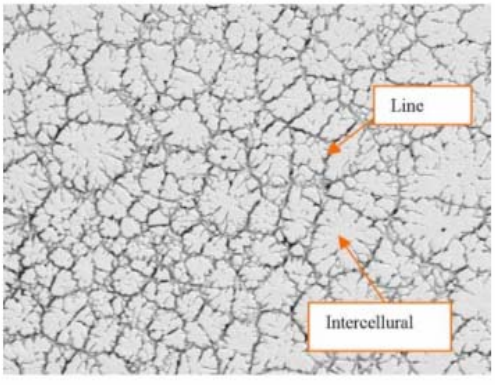

Fig.9 EDX, photographed in back-scattered electrons, 139.1 S, detail of mesh, magn. 940x

Energy dispersive micro-analysis was used for precise determination of chemical composition, both with back-scattered electrons and with secondary electrons. The measurement was focused always on type-identical places, i.e. on bright part marked as "matrix" (Fig. 8) and on part of mesh (Fig. 9). This is again composed of brighter phase ("intercellular"), which is interlaced with dark lines ("line").

Table 2 Average values of chemical composition determined by EDX

sample measuring point	aver. Al [at.%]	aver. Ni [at.%]	deviation Al	variance Al
139.1S matrix	26.580	73.420	0.2779	0.65
139.1S intercellular	24.658	75.342	0.3341	0.96
139.1S line	17.036	82.964	0.4781	1.23
139.2S matrix	25.328	74.672	0.2252	0.52
139.2S intercellular	23.978	76.022	0.1134	0.32
139.2S line	16.552	83.448	0.1533	0.46
139.3S matrix	25.532	74.468	0.2263	0.66
139.3S intercellular	23.972	76.028	0.1694	0.48
139.3S line	16.738	83.262	0.1825	0.52
143.1S grain	25.373	74.628	0.2592	0.73
143.2S grain	25.494	74.506	0.2548	0.67

The average values from these measurements are given in the Table 2. The measurements were made on cross section of central parts of samples. Al was determined and the rest was calculated.

Contents of nickel and aluminium in the area of bright phase marked as "matrix" correspond to inter-metallic phase Ni_3Al in all the measured samples. Composition of brighter areas in mesh also corresponds to the Ni_3Al phase, but aluminium contents is here reduced by 1 and even more at. %. Dark lines have substantially lower contents of aluminium. Higher contents of Al in solid solution of γ (Ni), which forms dark mesh, can be explained as result of comparatively high cooling rates with subsequent suppression of diffusion process in a solid phase [4]. Another possible explanation may be that analysing beam of electrons is so broad that it reaches also the area beyond the lines. In that case it would be possible that these lines are formed by solid solution of nickel. At crystallisation from the melt there did not occur any peritectic reaction. Particles of Ni_3Al drop out directly from the melt, therefore this homogenous structure, the character of which approaches single crystal, was created very easily.

3.2 X-Ray evaluation of structures

Cross section of central parts of the samples 139 and 143 were used for determination of grain orientation. X-ray diffraction by reflection method with use of Fe-radiation without filter was applied on homogenous samples 139 at 3 different places of cross section, on poly-crystalline sample 143 in various grains. The obtained Lauegrams were evaluated by

determination of angles between pairs of zones, to which correspond traces lying on intersecting hyperboles [3]. The results obtained for the samples 139.1, 139.2 and 139.3 are given ion the Table3.

Orientation in the samples 139 is always the same at all 3 points of measurement. Structure of these samples approaches the structure of preferentially oriented poly-crystals. In the samples 143 there were selected some grains at random. In the sample No. 143.2 orientation of grain axis <511> was determined without deviation and with deviation smaller than 2°.

In the sample No. 143.3 there was determined grain orientation <221> with deviation smaller then 5°.

Table 3 Determination of crystal orientation

Sample No.	Orientation	Note
139.1 a	<221>	deviation < 5°
139.1 b	<221>	deviation < 5°
139.1 c	<221>	deviation < 5°
139.2 a	<311>	deviation 8°, general orientation
139.2 b	<311>	deviation 8°, general orientation
139.2 c	<311>	deviation 8°, general orientation
139.3 a	<111>	deviation 4°
139.3 b	<111>	deviation 4°
139.3 c	<111>	deviation 4°

3.3 Tensile tests with measurement of acoustic emissions

Tensile tests were made for assessment of influence of various parameters of directional crystallisation, such as chemical composition and speed of crystallisation, on mechanical properties. Short tensile rods or round section with length of 55 mm and diameter of central part of the rod 5 mm were used for these tests. Table 4 summarises the measured values for 5 chosen samples of three different chemical compositions and with different speeds of crystallisation. Acoustic emission was measured with use of commercial testing equipment DAKEL XEDO made by company DAKEL-ZD Rpety, Czech Republic. Two-threshold detection according to the ASTM standard was applied, which enables simple amplitude discrimination of signals.

During all measurements there was monitored intensive acoustic emission with characteristic dependence on time and correlation with individual stages of deformation. In all measurements it is possible to discern several areas on the tensile diagram and characteristics of acoustic emission correspond with it. At first there appears the area of low loads with low level of acoustic emission. With increasing load there appears characteristic increase in number of emission overshoots, which can be related to activation of macro-plastic deformation. It is then followed by the area of degradation with gradual decline of strain, which is characterised by intensive acoustic emission. Particularly strong signals appear at the moments of jump drop of strain. This is characteristic for the samples 139.1 and 143.3. Process of deformation with load of the samples 139.2 and 139.3 is uniform, which corresponds with the obtained type of homogenous structure. The samples 139 and 143 were used for determination of mechanical properties.

Table 4 Resultant values obtained at tensile tests

Sample No.	Al [at.%]	Speed of directional crystallisation [mm/hour]	$Rp_{0.2}$ [MPa]	R_m [MPa]	Ductility till R_m [%]
139.1	25	108	267	392	13.9
139.2	25	60	343	707	20.5
139.3	25	18	245	505	53.4
143.1	24	105	222	501	13.1
143.3	24	60	208	430	17.3

The table 4 gives the measured values of yield strength, strength and ductility till yield strength for the given samples.

In this case conditions of directional crystallisation have manifested themselves very distinctly. In the sample 139.1 there did not occur any significant change of already measured values for the samples of the same type [4]. However, in the samples 139.2 and 139.3 there has occurred very significant increase in strength, but especially unexpectedly high increase in ductility up to 53 % in the sample 139.3 with speed of directional crystallisation 18 mm/hour.

The Figures 10 to 13 show load diagrams for the samples 139.1, 139.2, 139.3 and 143.3 with record of acoustic emissions.

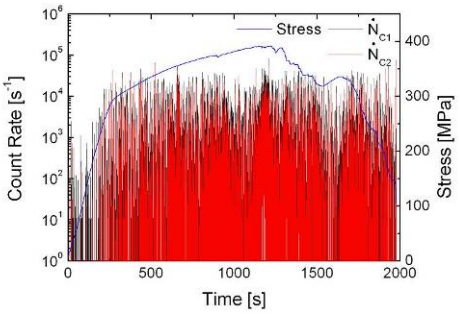

Fig.10 Load diagram for the sample No. 139.1

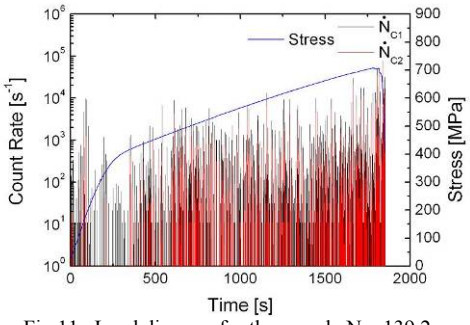

Fig.11 Load diagram for the sample No. 139.2

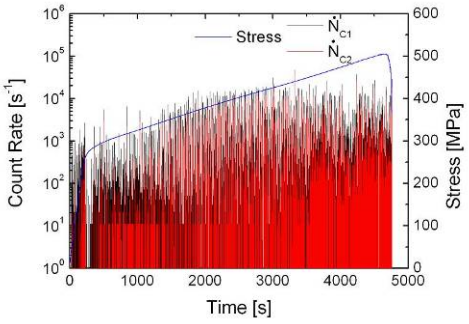

Fig.12 Load diagram for the sample No. 139.3

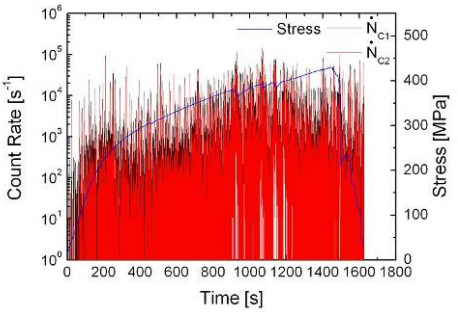

Fig.13 Load diagram for the sample No. 143.3

In the samples with higher ductility (marked as 139.2 and 139.3) there occurs a transverse fracture. The sample 139.1 has a very distinct structure on the fracture surface. The

sample 143.3 has – in spite of very high value of ductility – classically split grains of acicular shape. Typical de-cohesion of longitudinally directed crystals was observed here. The fracture itself is also of laminar type. Similar type of fracture was observed on the samples prepared at the VŠB-TU Ostrava [4]. Figures 14-17 show tensile rods with details of fracture surfaces.

Fig.14 Tensile rod of the sample 139.1 after tensile test, detail of fractures surface

Fig.15 Tensile rod of the sample 139.2 after tensile test, detail of fractures surface

Fig.16 Tensile rod of the sample 139.3 after tensile test, detail of fractures surface

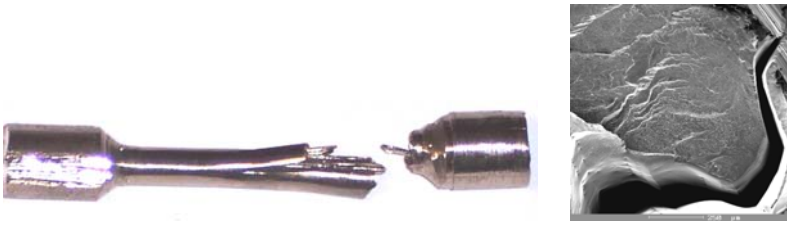

Fig.17 Tensile rod of the sample 143.3 after tensile test, detail of fractures surface

4. Evaluation

The work makes an evaluation of the samples prepared by the Bridgman method on the equipment for directional crystallisation. The samples prepared in Vladimir (Russia) show highly homogenous structure approaching the final required structure. Structures of the samples 139 have special character as there occur areas with reduced contents of aluminium. In these areas a line analysis was performed. It follows from these analyses that peritectic reaction did not take place during crystallisation from the melt. Very interesting results were obtained from

tensile tests with measurement of acoustic emissions. In the samples 139.2 and 139.3 there occurred surprising increase in ductility – up to the values of 20 and 53 %.

Future works with this experimental material will be oriented to the area of micro-structural evaluation, including determination of density of dislocations in individual types of samples and monitoring of their changes as a result of deformation at tensile tests.

Acknowledgement

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