

Cast in situ composites of Ni₃Al / MeC type

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Abstract

In this work a new method SHSB (Self- Propagating High-Temperature Synthesis in Bath) has been used to produce of five new composites i.e. Ni₃Al / TiC, Ni₃Al / WC, Ni₃Al / Zr, Ni₃Al / NbC and Ni₃Al / TaC (in the amount of carbides of 5% volume fraction.) The composites fabricated by the „in situ” process, with the Ni₃Al compound used as a matrix material and the reinforcement composed of Ti, W, Zr, Nb or Ta carbide particles were made. Fabrication of composites was carried out in Balzers vacuum furnace and conducted the process in the atmosphere of argon at a negative gas pressure of 0,5 MPa. After melting down of aluminium and completion of the exothermic reaction between mixing powder of carbon, aluminium, titanium or tungsten, zirconium, niobium and tantalum, boron in the form of Al-B 3% master alloy was introduced to alloy melt. From thus fabricated composites, the specimens were prepared for metallographic examinations, and scanning topographic analysis. It have been shown that the size of TiC and TaC particles was comprised in a range of up to 10 µm. The NbC and ZrC carbides were characterised by the dimensions of up to 20 µm, while WC carbides were the largest (up to 80 µm.) In all the examined composites, the X-ray microanalysis revealed total absence of reaction products at the matrix-reinforcing particle interface. The SHSB process eliminated the fundamental problem - reactivity of the matrix-particle system, observed in the ex situ methods .

Keywords: Composite in situ, Intermetallic phase, SHSB Process, Exothermic reaction, Carbides of TiC, WC, ZrC, NbC, TaC.

1.Introduction

The past few years have faced a dynamic development of the research techniques of metal matrix composites [1-4] which, given a number of very attractive properties, are successfully applied by numerous sectors of industry, to mention as an example the automotive industry and aviation [3]. Among various MMCs, very popular have become the composite materials based on light metal alloys, like aluminium, titanium or magnesium, ensuring high relative strength. As a reinforcement, carbides (TiC, SiC, ZrC), oxides (Al₂O₃, TiO₂, MgO, ZrO₂), nitrides (BN, TiN, ZrN), borides (TiB₂, ZrB₂, SiB₂), silicides (MoSi₂), or fine particles of various intermetallics (Ni₃Al, NiAl, Fe₃Al, FeAl, Ti₃Al, TiAl) are commonly used [5].

A new and original solution consists in using the intermetallic Ni₃Al compound as a matrix material. What decided about the

choice of this particular material was a very interesting complex of physico-chemical and mechanical properties it can offer. Numerous studies are available that describe *in extenso* the technology by which the intermetallic materials are produced, specifying also their properties – mechanical and anticorrosive [6,7]. The Ni₃Al intermetallic is characterised by high resistance to oxidation and stability of mechanical properties over a wide range of temperatures. Its tensile strength is increasing in function of temperature and, depending on the size of the grains, reaches its maximum within a range of 650⁰ – 850⁰ C. Contrary to numerous nickel-based superalloys, the intermetallics are characterised by satisfactory compression strength in the temperature range of 650⁰ – 1100⁰ C, accompanied by high fatigue strength, combined with homogeneity and absence of chemical segregations when used as a matrix material. The Ni₃Al compound is also characterised by high creep resistance and

resistance to mechanical wear. Against the background of all these undeniable advantages, a definite drawback of the Ni₃Al intermetallic is its poor ductility. Even so, however, this inconvenience can be overcome by application of various microalloying additives, boron in particular [8]. On the other hand, the choice of metal carbides for the reinforcing phase has been justified by their quite specific properties – physical, chemical, and mechanical. Metal carbides form a group of compounds characterised by the high melting point, hardness, Young modulus, friction wear resistance and corrosion resistance at both ambient and high temperatures. Therefore, transforming the high-strength Ni₃Al compound into a composite reinforced with the creep-resistant and hard carbides enhances even more its excellent mechanical and tribological properties. The fabrication of Ni₃Al / MeC composites by the synthesis of TiC, WC, ZrC, NbC, TaC carbides in the SHSB process, i.e. directly in the Ni₃Al melt, opens the way to better control of the volume fraction of a reinforcing phase, ensuring - moreover - the thermodynamic stability of material and absence of chemical reactions at the matrix-reinforcing particle interface, with the additional advantage of a uniform dispersion of the composite particles. From the group of carbides presented in Table 1 [9], the authors have chosen those for which it has been possible to effectively initiate the exothermic reaction of synthesis [10].

Table 1. Carbides used as a reinforcement of MMCs [6]

MeC	Density [g/cm ³]	Melting point [°C]	Microhardness. [μHV]	Heat of synthesis [kJ/mole]
TiC	4.93	3250	2850	183
SiC	3.21	2800;	3340	66
ZrC	6.73	3530	2920	184
VC	5.36	2830	2094	101
NbC	7.56	3760	2000	130
TaC	14.30	3880	1600	141
WC	15.60	2870	2080	35

2. Methods of research

The specific characteristics of a melting process of the intermetallic Ni₃Al compound result from a large difference in the melting points of aluminium and nickel, and from the occurrence of exothermic reactions during formation of this phase. In view of a rapid temperature increase up to about 1660°C, immediately after the occurrence of exothermic reaction between Al and Ni in the atmosphere of air, aluminium undergoes oxidation, followed by slag formation, which makes exact control of the alloy chemical composition very difficult. Therefore, the authors decided that the process of the Ni-Al alloy fabrication should be carried out in vacuum. Consequently, they used a Balzers vacuum furnace, and conducted the process in the atmosphere of argon at a negative gas pressure of 0,5 MPa. After melting down of aluminium and completion of the exothermic reaction, boron in the form of Al-B 3% master alloy was introduced to alloy melt. As a next step, applying the SHSB process, in the melt of the base Ni-Al-B alloy, during the successive stages of the process, the

carbides of titanium, tungsten, zirconium, niobium and tantalum (in the amount of 5% volume fraction) were produced. The adopted procedure was, for example, the following: titanium and carbon in the form of Ti-C-Al powder were pressed at a pressure of 500 MPa to form a briquette, which was next introduced to the base alloy melt at a temperature of 1430°C. As soon as the briquette entered into contact with the surface of the melt, the exothermic reaction of the titanium carbide synthesis started. After the lapse of 5 minutes, the ready Ni₃Al + TiC suspension was cast into ceramic mould. Similar method was used for the synthesis of the remaining carbides, i.e. WC, Zr, NbC and TaC. From thus fabricated composites, the specimens were prepared for metallographic examinations, scanning analysis and structural examinations.

3. The results of investigations

In the study, the results of the investigations carried out on the composites of Ni₃Al / TiC, Ni₃Al / WC, Ni₃Al / Zr, Ni₃Al / NbC and Ni₃Al / TaC are presented.

3.1. Ni₃Al / TiC composite

The microstructure of the fabricated Ni₃Al/TiC composite is shown in Figure 1, while the scanning topographic image of the specimen is shown in Figure 2.

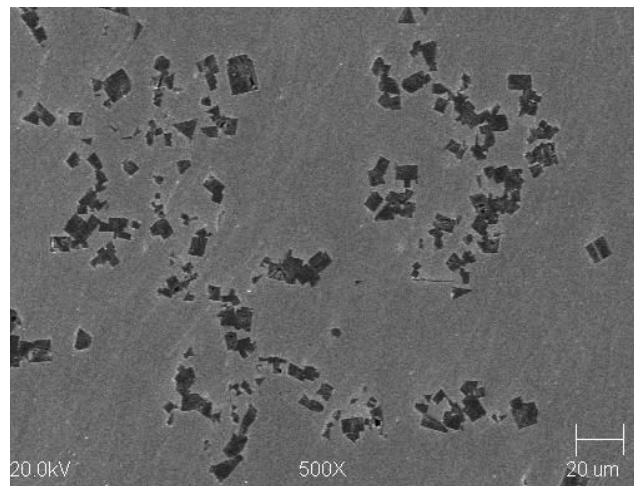


Fig. 1 Microstructure of Ni₃Al /TiC composite, 500x

A microphotograph of the Ni₃Al /TiC composite (Fig.1) shows titanium carbide precipitates of dimensions of up to 10 μm, distributed in the Ni₃Al matrix. The scanning image also confirms this fact (Fig.2). In the Ni₃Al/TiC composite, at selected points, local chemical analysis was made; its results are given in Table 2. No reactions have been observed between the Ni₃Al matrix and the TiC.

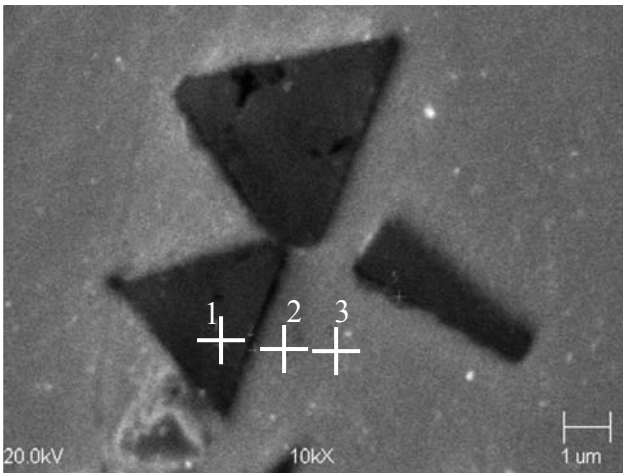


Fig.2 Scanning image of Ni₃Al /TiC composite, 10000x

Table 2. The results of chemical analysis of the TaC particles and matrix in Ni₃Al/TiC composite

Measuring point	C content wt%	Al content wt%	Ni content wt%	Ti content wt%
1	22,0	0,3	1,7	78,0
3	0	13,3	86,6	0,1
4	0	12,0	87,8	0,2
Stoichiometry of TiC: Ti = 79,9 %, C = 19,1 %				

3.2. Ni₃Al / WC composite

The fabricated composite was subjected to preliminary metallographic examinations. Its microstructure is shown in Figure 3.

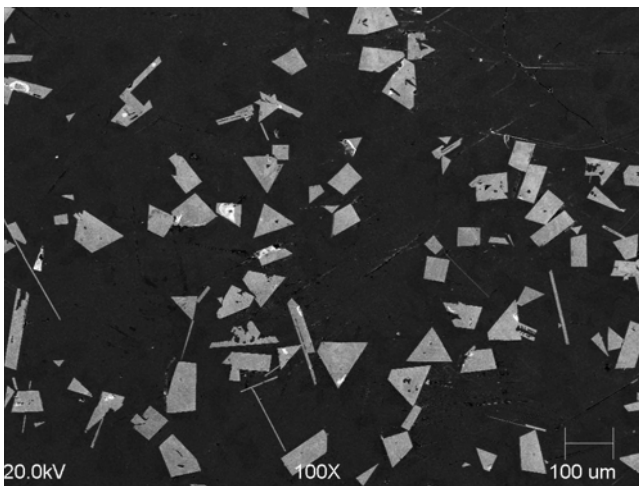


Fig.3. Scanning image of Ni₃Al/WC composite, 100x

The particles of WC carbide of dimensions reaching 20 to 80 μm are distributed in the the Ni₃Al composite matrix. Next, the

Ni₃Al/WC composite matrix-reinforcing particle interface was subjected to scanning analysis and X-ray microanalysis, as illustrated in Figure 4. No reactions have been observed to take place between the Ni₃Al composite matrix and the WC reinforcing phase. Local chemical analysis carried out in microregions of the phase boundary has confirmed this fact, indicating the existence of the Ni₃Al phase and tungsten carbide WC only (Table 3).

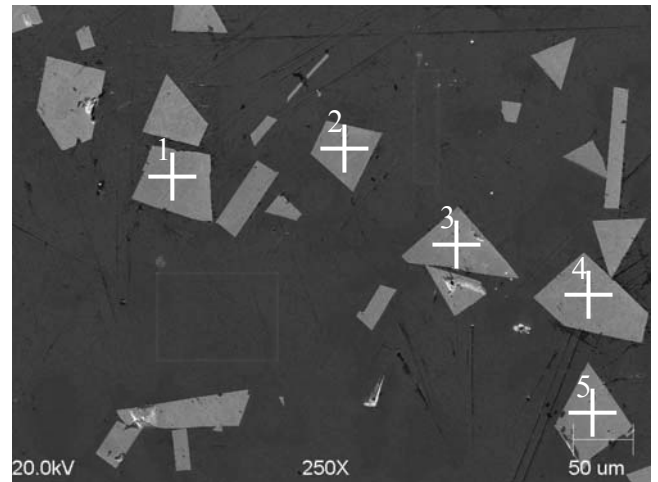


Fig.4 Scanning image of Ni₃Al /WC composite, 250x

Table 3. Chemical analysis in microregions of the Ni₃Al/WC composite matrix-reinforcing particle interface

Measuring point	C content wt%	Al content wt%	Ni content wt%	W content wt%
1	16,1	0,2	0,0	83,7
2	19,9	0,1	0,1	79,9
3	22,1	0,1	0,2	77,6
4	19,9	0,2	0,2	79,6
5	19,9	0,2	0,1	79,8
Stoichiometry of WC: W = 93,9 %, C = 6,1 %				

3.3. Ni₃Al / ZrC composite

As in the composite described above, also in this case, attention was focussed on the scanning analysis and X-ray microanalysis of the matrix-reinforcing particle interface. The microstructure in a sample of Ni₃Al/ZrC composite is shown in Figure 5, while the results of an X-ray microanalysis carried out for the Ni₃Al matrix-reinforcing particle interface are shown in Fig. 6.

The observed particles of zirconium carbides are several times smaller than the particles of WC carbides. Also in this case, no chemical reactions were observed at the matrix-reinforcing particle interface. In the Ni₃Al/ZrC composite, at selected points of the composite, local chemical analysis was made; its results are given in Table 4.

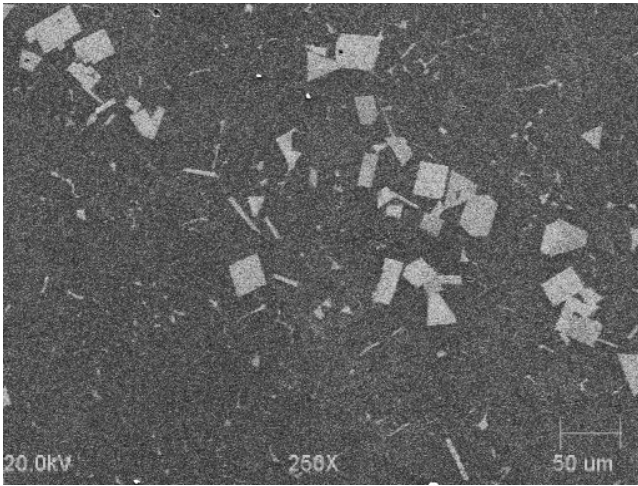


Fig.5. Scanning image of Ni_3Al/ZrC composite, 100x

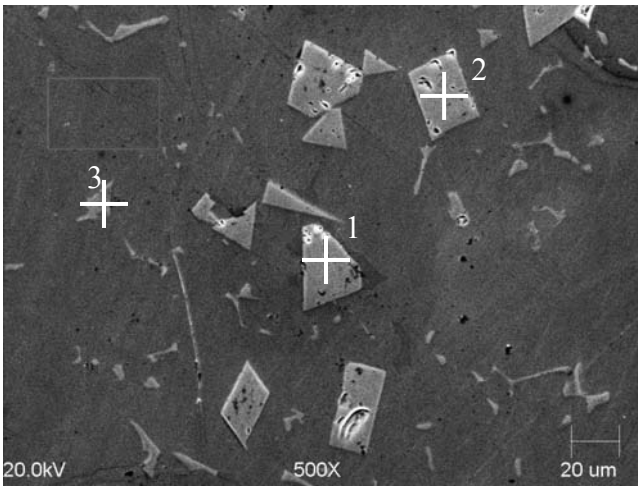


Fig 6. Scanning topographic image of Ni_3Al/ZrC composite, 500x

Table 4. Chemical analysis in microregions of the Ni_3Al/ZrC composite matrix-reinforcing particle interface

Measuring point	C content wt%	Al content wt%	Ni content wt%	Zr content wt%
1	11,9	0,1	1,6	88,1
2	12,3	0,1	1,6	87,7
3	12,1	0,2	3,5	87,9
Stoichiometry of ZrC: Zr = 88,3%, C = 11,7 %				

3.4. Ni_3Al/NbC composite

The metallographic examinations of this composite have indicated that the NbC niobium carbides occur in two forms, viz as single particles and, more frequently, as large agglomerations (Fig. 7). Their size is comprised in a range of 10 – 20 μm .

The X-ray microanalysis reveal the two main phases, i.e. Ni_3Al and NbC (Fig.8,) confirmed further by the results of local chemical analysis (Table 5).

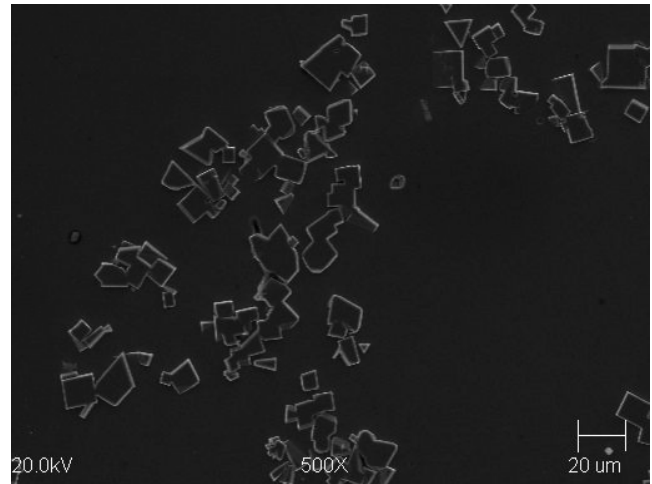


Fig.7. Scanning image of Ni_3Al/NbC composite, 500x

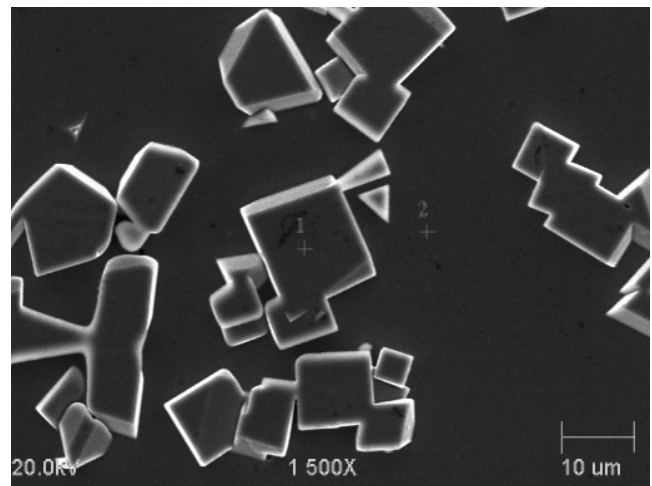


Fig.8 Scanning topographic image of Ni_3Al/NbC composite, 1500x

Table 5. Chemical analysis in microregions of the Ni_3Al/NbC composite matrix-reinforcing particle interface

Measuring point	C content wt%	Al content wt%	Ni content wt%	Nb content wt%
1	33,2	0,0	0,1	66,8
2	0,1	13,5	86,3	0,0
Stoichiometry of NbC: Nb = 88,5 % , C = 11,5 %				

3.5. Ni₃Al / TaC composite

The metallographic examinations of this composite have indicated that the NbC niobium carbides occur also in two forms, viz as single particles and, more frequently, as large agglomerations (Fig. 9). The size of the precipitates was comprised in a range of 4 – 10 μm (Fig.11).

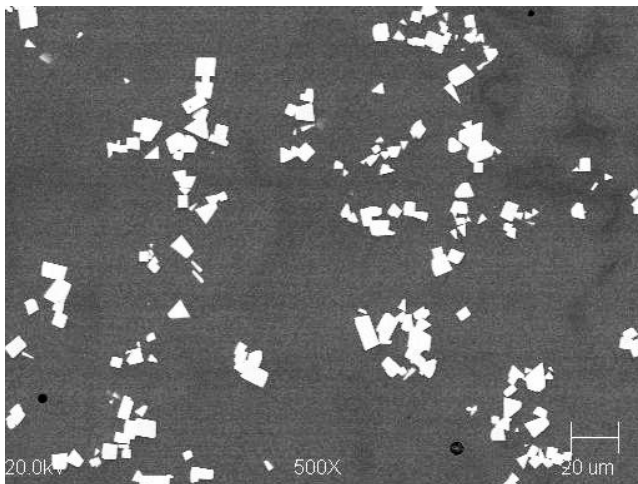


Fig.9 Scanning image of Ni₃Al/TaC composite, 500x

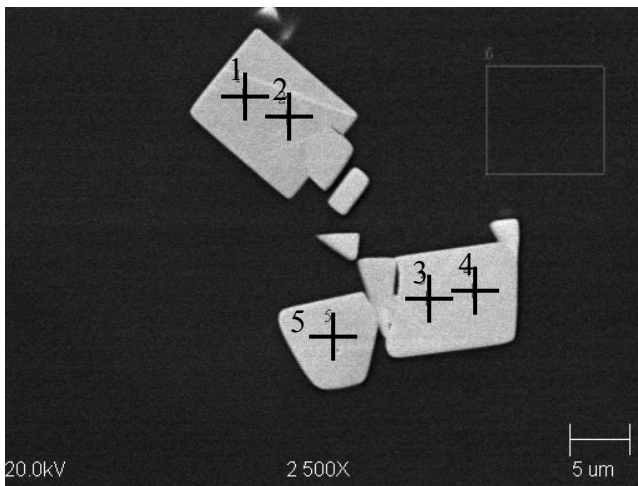


Fig.10 Scanning image of Ni₃Al /TaC composite with marked points of chemical analysis, 2500x

The chemical composition were made by the EDX method at points indicated in Fig. 10; the obtained results are compared in Table 6.

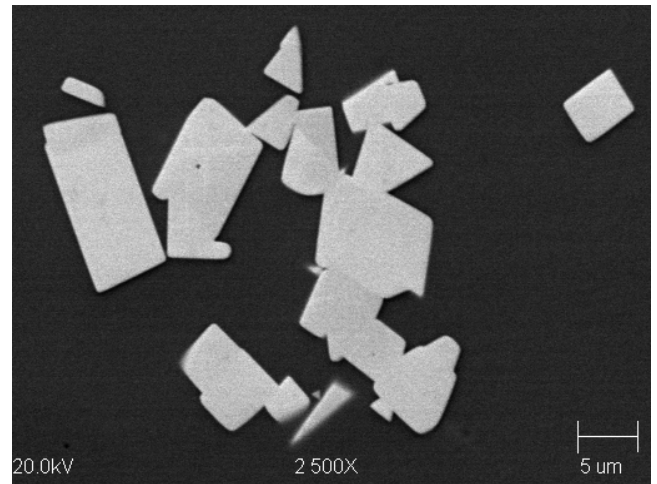


Fig.11 Scanning image of Ni₃Al /TaC composite, 2500x

Table 6. The results of chemical analysis of the TaC particles and matrix in Ni₃Al/TaC composite

Measuring point	C content wt%	Al content wt%	Ni content wt%	Ta content wt%
1	7,0	0,0	1,7	93,0
2	7,2	0,1	1,4	92,8
3	5,8	0,1	1,5	94,2
4	6,8	0,0	1,6	93,2
5	7,2	0,0	1,7	91,8
Stoichiometry of TaC: TaC = 93,8 %, C = 6,2 %				

Conclusions

1. The SHSB process has turned out to be quite an efficient tool in the synthesis of five composites, i.e. Ni₃Al / TiC, Ni₃Al / WC, Ni₃Al / Zr, Ni₃Al / NbC and Ni₃Al / TaC. Moreover the SHSB process eliminated the fundamental problem - reactivity of the matrix-particle system, observed in the ex situ methods .
2. The size of the TiC and TaC particles was comprised in a range of up to 10 μm. The NbC and ZrC carbides were characterised by the dimensions of up to 20 μm, while WC carbides were the largest (20 – 80 μm.).
3. In all the examined composites, it can be observed the absence of reaction products at the matrix-reinforcing particle interface (absence of oxides and other similar precipitates, no cracks present).

References

- [1] E. Fraś, A. Janas, A. Kolbus, E. Olejnik, Matrix- particle Interphase boundaries of the selected in situ and ex situ

- Composites MMCs ,Archives of Foundry, 2006, 6, 18, 297-304 (in Polish).
- [2] E. Fraś, A. Janas, A. Kolbus, H. Lopez, Synthesis of Composites in situ Al/ TiC by SHSB method, IV Scientific Conference. PMTK- Composites 2000, Jaszowiec 2000, 201-207 (in Polish).
- [3] A. Janas, Doctor's Thesis, AGH, Cracow,1998 (in Polish).
- [4] L. Froyen, In situ processing of metal matrix composites, Proceedings of the International Conference on Light Alloys and Composites, Zakopane,1999, 15-29.
- [5] A. Ibrahim, F. Mohamed, E. Lavernia, Particulate reinforced metal-matrix composites- a review, Journal of Materials Science 26, 1997, 1137-1156.
- [6] S.C. Huang, A.I. Taub, K.M. Chang, Acta Metallurgica 1984, 32, 1703.
- [7] S.C. Deevi, V.K. Sikka, Processing and Applications Intermetallics, 1996, 4, 357.
- [8] E. Fraś, A. Janas, A. Kolbus, E. Olejnik, Cast Ni₃Al / MeC (Me-W,Zr) composites in situ, Archives of Foundry,2006, 6, 18, 317-324 (in Polish).
- [9] S. Stolarz, High-melting Compounds and Phases, WŚ, Katowice, 1978 (in Polish).
- [10] A. Zuhair Munir, U. Anselmi-Tamburini, Self-propagating exothermic reactions, The synthesis of high-temperature materials by combustion in materials, Materials Science Reports, 1989, 3, 277.