A R C H I V E S O F M E T A L L U R G Y A N D M A T E R I A L S
Volume 54 2009 Issue 4

T. DUREJKO*, P. JÓŹWIK*, Z. BOJAR*

JOINING OF Ni_3Al MICROCRYSTALLINE FOILS BY SHS REACTION

ŁĄCZENIE MIKROKRYSTALICZNYCH FOLII NA OSNOWIE FAZY Ni_3Al METODĄ REAKCJI SHS

An attempt of obtaining Ni3Al/FeAl/Ni3Al joints via sintering of Ni3Al foil with a reactive spacer is described. The method is based on sintering technically pure iron and aluminium powders or Fe and Al P/M (Powder Metallurgy) compacts. For sintering the Ni_3Al foils in two material conditions were selected – immediately after rolling and after soaking at the temperature of 1000 C for up to two hours.

A presintering (stage one) was carried out at the temperature of 620°C under cyclic variable load which initiated volumetric reaction SHS (Self-Propagating High Temperature Synthesis). The second stage (basic sintering) relied on free soaking of obtained joints at the temperature of 1200°C in a protective argon atmosphere. This stage was carried out in two variants: the first variant – with fast heating of the Ni_3Al/FeAl/Ni_3Al system and maintaining in the temperature for 15 minutes and next slow cooling in air, and the second one - heating and cooling of the Ni_3Al/FeAl/Ni_3Al system in a furnace (total time – 1hour).

The following parameters of obtained samples were analysed: metallographic analysis using scanning microscope, grain size, chemical composition (point and linear analysis), and microhardness.

In the first stage (presintering) an occurence of the SHS reaction was confirmed. Metallography studies revealed a zonal structure of the samples. Depending on applied variants of heat treatment (in the basic sintering), from two up to eight transient zones with different chemical composition appear in the joint structure. The first variant including rapid heating and cooling during basic sintering causes increase in hardness in transient zones (up to the level of 360 HV), occurrence of a hard (473±56 HV), non-visible in microscopic observation FeAl zone, and an appearance of local cracks on boundaries between the Ni_3Al phase and a zone directly adjacent to it. For the second variant of the basic sintering microhardness in Ni_3Al and transient zones is comparable while for FeAl grains amounts to 320±8 HV.

First recognizing tests on disruption of obtained joints were done. They confirmed high quality of joints produced in the variant II. Additionally, a successful attempt to bond Ni_3Al strips with a Fe-Al reactive spacers using high-current pulses was performed.

W pracy przedstawiono próbę otrzymywania złącz Ni_3Al/FeAl/Ni_3Al metodą spiekania folii Ni_3Al z przekładką reaktywną w postaci mieszaniny czystych technicznie proszków żelaza i aluminium lub wypraski z proszków Fe i Al. Do spiekania wytypowano folie Ni_3Al w dwóch stanach materiałowych – bezpośrednio po walcowaniu oraz walcowaniu i wygrzewaniu w temperaturze 1000°C i czasie do dwóch godzin.

Spiekanie wstępne (etap pierwszy) przeprowadzono w temperaturze 620°C pod obciążeniem cyklicznie zmiennym inicjując objętościowo reakcję SHS (Self-Propagating High Temperature Synthesis). Etap drugi (spiekanie zasadnicze) polegał na swobodnym wygrzewaniu utrzymanych złącz w temperaturze 1250°C i atmosferze ochronnej argonu. Etap ten zrealizowano w dwóch wariantach: pierwszy – z szybkim grzaniem układu Ni_3Al/FeAl/Ni_3Al i wytrzymaniem w temperaturze przez 15 minut a następnie studzeniem w powietrzu i drugi – z grzaniem i studzeniem układu Ni_3Al/FeAl/Ni_3Al razem z piecem (łączny czas 1h).

Uzyskane próbki poddano analizie metalograficznej na mikroskopie skaningowym, analizie wielkości ziarna, składu chemicznego (punktowo oraz liniowo) oraz pomiarom mikrotwardości.

Na etapie spiekania wstępnego potwierdzono występowanie reakcji SHS. Badania metalograficzne ujawniły strefową budowę próbek. W zależności od zastosowanych wariantów obróbki cieplnej (spiekania zasadniczego) w strukturze złącza występuje od dwóch do ośmiu stref przejściowych o różnym składzie chemicznym. Wariant pierwszy obejmujący gwałtowne grzanie i chłodzenie podczas etapu spiekania zasadniczego powoduje wzrost twardości w strefach przejściowych (do poziomu 360 HV), występowanie twardej, niewidocznej podczas obserwacji mikroskopowych strefy FeAl (473±56 HV) oraz pojawienie się lokalnych pęknięć na granicach pomiędzy fazą Ni_3Al i strefą bezpośrednio do niej przyległą. W przypadku spiekania

* FACULTY OF ADVANCED MATERIALS AND TECHNOLOGY, MILITARY UNIVERSITY OF TECHNOLOGY, KALISKIEGO 2 STREET, 00-908 WARSAW, POLAND

zasadniczego według wariantu drugiego mikrotwardość w Ni_3Al i strefach przejściowych jest porównywalna, natomiast w ziarnach FeAl wynosi 320±8 HV.

Przeprowadzono rozpoznawcze badania rozrywania uzyskanych złącz, które potwierdziły wysoką jakość połączenia otrzymanego zgodnie z wariantem II. Dodatkowo przeprowadzono również udaną próbę łączenia taśm Ni_3Al z przekładką reaktywną Fe-Al poprzez impulsy wysokoprądowe.

1. Introduction

Alloys based on ordered intermetallic phases, commonly called intermetals, like Ni_3Al alloys, belong to this modern material group [1]. The Ni_3Al intermetallics present, as compared to nowadays utilized nickel-based superalloys, abnormal increase with temperature in yield point, relatively excellent oxidation and corrosion resistances [2]. These materials, in a form of foils, found or will find in the nearest future application as elements for car catalysts, jet aircraft engines, or electromechanical microsystems (heat micro-exchangers, micro-reactors, micro-servo-motors etc.) [3].

The major obstacle to application of intermetals in industry is their low plasticity and a tendency to brittle cracking, in particular in room temperature [2]. Despite presenting by alloys based on the intermetallic Ni_3Al phase with boron addition a certain plasticity (as shown by C.T Liu and V.K. Sikka, [4]), this is not sufficient to provide an obtaining a material – foil of thickness below 800 μm via cold rolling.

Using a method of controlled cold plastic strain, developed by the authors, including a selection of a single draft and a plastic strain velocity, strips with thickness below 100 μm were obtained [5]. Cold rolled strips have, despite high strengthening, good plasticity at bending and noticeable one at tension. Tests results show that the material obtained with this method of working is promising for structural high-temperature applications, mainly because of its high specific strength. Properly profiled foils can be used as functional and structural elements like laminates or honeycomb structures. However, due to potentially high brittleness and structure stability (low diffusion coefficient) of the above discussed material, bonding of thin foils looks rather difficult. Traditional, thermal methods of bonding are hardly useful due to a very low thickness of bonded elements. On the other hand, advanced bonding technologies, like laser micro-welding need a very complex instrumentation and can be, first of all, applied in laboratory conditions. On the base of our experience in powder metallurgy applied to FeAl sinters manufacturing [6], an idea of bonding of Ni_3Al strips with a reactive spacer in a form of a mixture (with previously determined granulation and stoichiometric composition) of technically pure iron and aluminum powders appeared. Properly selected parameters of welding process enabled exothermic SHS (Self-Propagating High Temperature Synthesis) reaction and diffusive bonding of the individual system elements to be initiated in the Fe-Al powder volume. One should have in mind, however, that the FeAl phase is commonly recognized as a brittle phase and can create a weak link in the obtained construction [7,8]. This problem was solved by technology. Heavy investigations of intermetallic FeAl sinters showed that their plasticization can be obtained by a cyclically variable load applied to the presintering stage [9]. During cyclically variable loading an intense mechanical fragmentation of primary oxidation layers (mainly Al_2O_3 on aluminum particles aurface) which causes increase in a area of metallic contact of iron and aluminum particles (i.e. increase in energy released in the SHS reaction) as well as a homogeneous deposition of oxidation phase in the sinter volume. Thus, the developed technology can be used not only in the process of thermal bonding of thin foils made of the Ni_3Al alloy, but also for manufacturing of intermetallic gradient FeAl/Ni_3Al systems. A full controlling of a strengthening level of the Ni_3Al foil and of a level of oxidation phase and FeAl matrix refinement enables practically unlimited amount of gradient structure to be created. A number of information present in a scientific literature devoted to bonding of thin Ni_3Al foils is minimal and is predominantly limited to utilization of the SHS energy during nickel – aluminum reaction (lower energy than in Fe-Al reaction).

2. Materials and experimental procedure

In order to make Ni_3Al/FeAl/Ni_3Al joints, the Ni_3Al foils of 30×20×0.5 mm size in two conditions (three pairs per condition) were prepared. The first condition included foils immediately after rolling (strengthened condition – grain size about 5 μm) (Fig.1a), the second one – a condition after rolling and a heat treatment at 1000°C in two hours (grain size about 5 μm) (Fig. 1b).

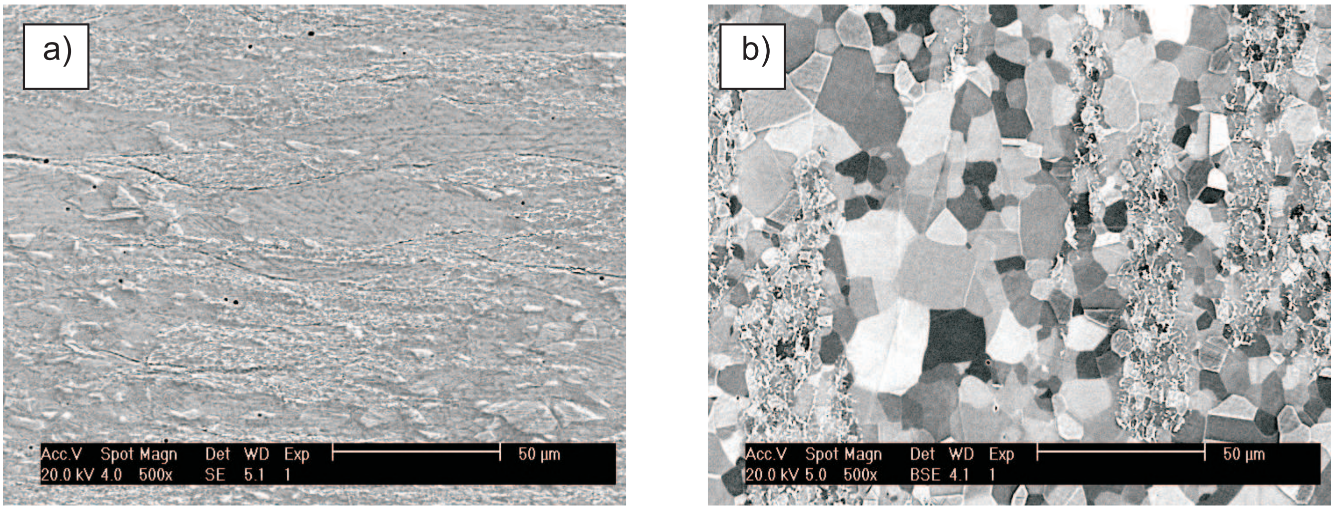

Fig. 1. Microstructure of Ni_3Al foils, (a) after rolling, (b) after rolling and a heat treatment at 1000°C in two hours

The sheet metal plates were polished on the junction side using abrasive paper with gradation values of 200, 600, and 1000 gradation, respectively. In order to protect Fe-Al powder mixture against pouring from among foils, an electro-erosion method was used to notch grooves of 0.4 mm in width and 0.2 mm in depth along the symmetry axis of the longer side (first junction). In the first system a mixture of technically pure Fe and Al powders of 38÷40 µm granulation and 60 to 40 % at. ratio of iron to aluminum was used as a reactive spacer. For the second system a reactive spacer was made up of a P/M (Powder Metallurgy) compact consisting of a Fe-Al powder mixture (matrix compacting under 300 MPa for 15 minutes) of the same granulation and composition as for the first case. After presintering samples underwent basic sintering in two variants. In the first variant joint was soaked in the temperature of 1250°C without protective atmosphere for 15 minutes. After fixed a sample was taken out from a furnace and cooled in air. In the second variant a sample was freely sintered at 1250°C in argon atmosphere and soaked and cooled in a furnace (total time 1 hour).

Samples obtained after basic sintering were cut (using electro-erosion machine) in the place of junction (perpendicularly to the longer flank, next mechanically grinded on the grinding – polishing machine STRUERS PLANOPOL 3 using abrasive papers of 100÷2000 gradation, and polished on polishing disks with a diamond slurry of 6, 3, 1, and 0.25 µm granulations. Microstructure of investigated joints after basic sintering was revealed by chemical etching with a reagent composed of the 33% CH_3COOH + 33% HNO_3 + 33% H_20 + 1% HF (FeAl regions) and with Marble reagent (Ni_3Al regions).

An assessment of a size and shape of matrix grains (etched microsections, detektor BSE (Back Scattering Electron)) was based on analysis of 500 samples (on average) for each technology variant.

Quantitative investigations for chemical composition of individual zones of the Ni_3Al/FeAl/Ni_3Al joints were carried out using point and linear EDS (Energy-Dispersive Spectroscopy) analysis.

For microhardness tests a Vicker's metod was used (50 G load for 10 s, 10 measurements were performed in all regions of occurrence of each phase).

3. Results and discussion

Analysis of a displacement of a bottom stamp in function of time/cycle number was made. It was stated that for suggested conditions of presintering (load type and level, temperature) of prepared Ni_3Al/FeAl/Ni_3Al joints an initiation of the volumetric SHS reaction is possible. Rapid drop on a displacement curve shows the occurence of exothermic reaction and appearance of a mixture of phases with high aluminium content in the reactive spacer structure (at fixed thermal conditions) (Fig. 2). A drop value and an instant of its appearance does not depend on spacer type (a mixture of Fe and Al powders or P/M compact of technically pure Fe and Al powders) and is comparable to all investigated samples. After SHS reaction a condensation of reaction products occurs.

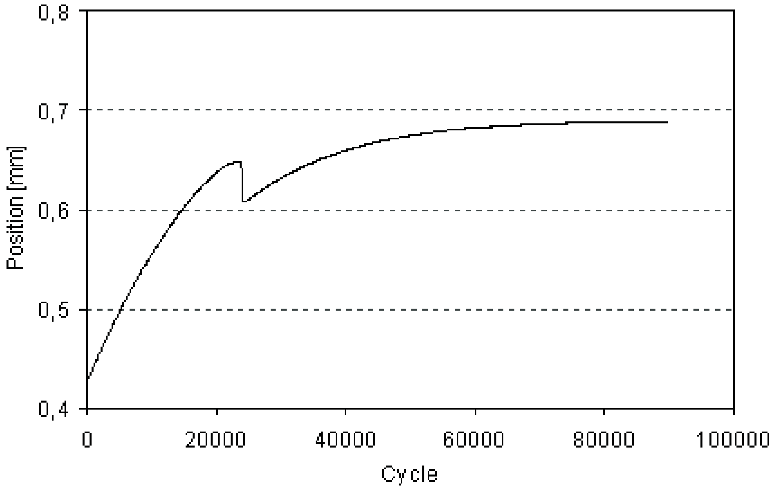

Fig. 2. Influence of SHS reaction on lower stamp position during presintering process

Analysis of a surface of non-etched metallographic microsections showed that the suggested method of the Ni_3Al/FeAl/Ni_3Al systems bonding anbles a diffusive junction to be set among reactive spacer and intermetallic foils. For the first variant (spacer as a Fe and Al powder mixture) an occurence of two transient zones of the width of 30 μm each was revealed (Fig. 3b). The basic sintering caused a reconstruction of structural components of the spacer into grains of a FeAl phase (7,7±3 μm). Non-uniform in size Al_2O_3 oxides are placed in the grain boundaries. In grains the FeAl phase grains a nickel content up to 6% at. was observed.

In transient zones nickel content gradually increases (towards foil interior), reaching a value characteristic for Ni_3Al phase at the distance of about 100 μm from the axis of symmetry. It is worth noting that a presence of iron was also observed in transient zones.

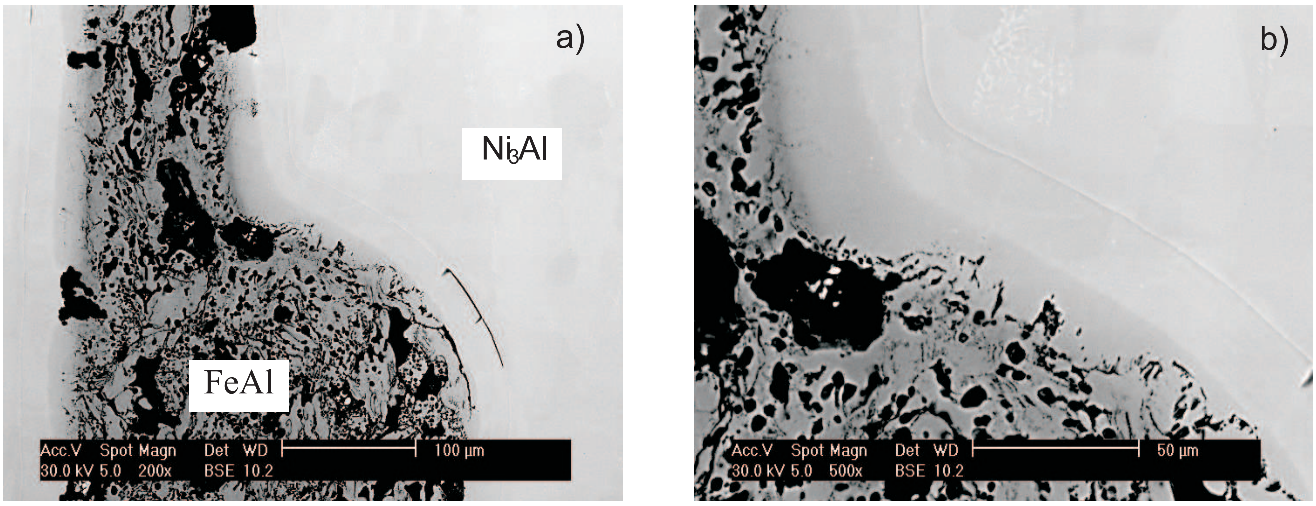

Fig. 3. Microstructure of Ni3Al/FeAl/Ni3Al joint (variant I), (a) local cracks along the boundary Ni3Al - first transient zone, (b) transient zones

"Hard" conditions of the basic sintering (rapid heating up to 1250°C and cooling in air) of the Ni_3Al/FeAl/Ni_3Al joint caused local cracks along the boundary Ni_3Al – first transient zone (Fig. 3a). The analysis of a microstructure of joined regions of Ni_3Al strips showed that during sintering a process of recrystallization occurred (material after deformation) in result of which an Ni_3Al grain size has been stabilized at the level of 40±5 μm.

In the second technology variant (spacer in the form of Fe-Al P/M compact) a sintered zone consists of uniform in size Al_2O_3 oxides and grains based on a matrix of FeAl phase (8.8±2 μm). In the joint structure eight transient zones can be distinguished (Fig. 4b). In the sintered zone volume an increased level nickel content (about 8% at.) was observed (as compared to variant I). This is mainly due to one-hour basic sintering (in the variant I basic sintering was carried out only 15 minutes). The total thickness of transient zones in the second variant is comparable to the thickness of a transient zones for the variant I. A detailed analysis of boundaries of individual transient zones did not revealed any cracks (Fig. 4a).

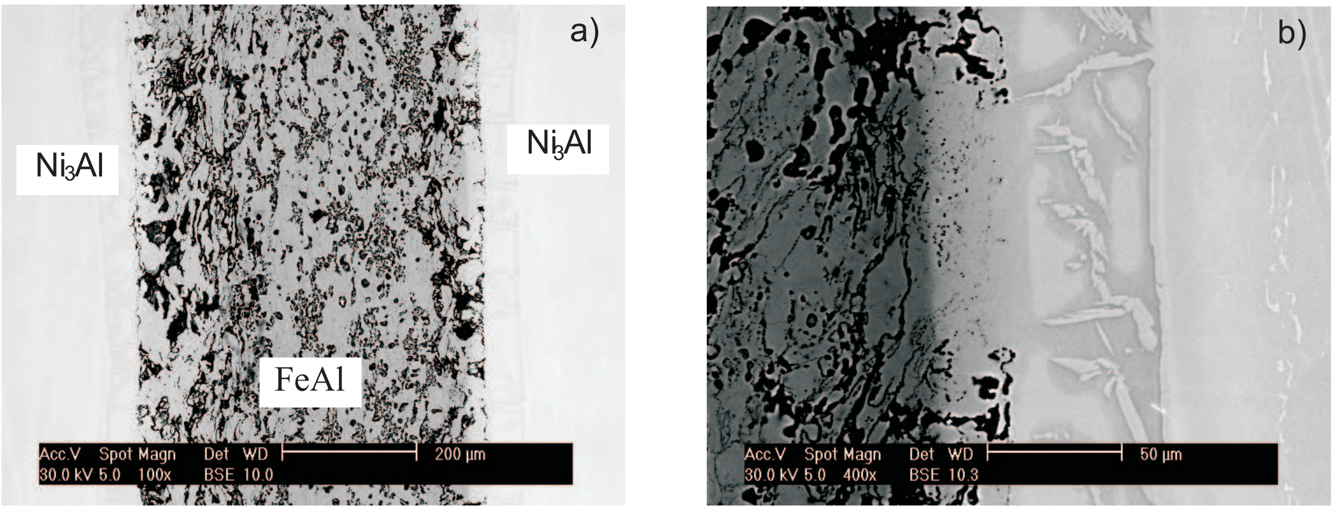

Fig. 4. Microstructure of Ni_3Al/FeAl/Ni_3Al joint (variant II), (a) overall view, (b) transient zones

A prolonged time of the basic sintering affected a growth of the Ni_3Al phase grains. Over eight-fold increase in grain equivalent diameter up to 42±4 m was found. A microhardness of the Ni_3Al foil after basic sintering, independently on technology variant used, amounts to 235±8 HV (Fig. 5a,b). In the first joint, an increase in microhardness is observed for transient zones region. Both first and second zone have elevated microhardness in relation to the intermetallic foil of about 130 HV. In the joint structure another zone appears which was not observed under microscope. Linear measurements of the microhardness showed that its thickness comes to 40 μm (in FeAl spacer depth) while microhardness holds the level of 473±56 HV ("central" zone a FeAl microhardness amounts to 338±25 HV) (Fig. 5a).

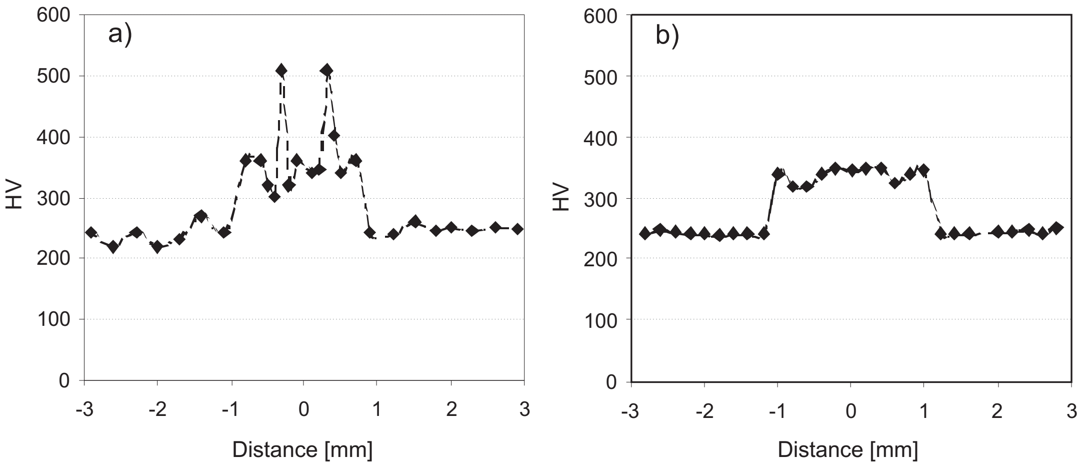

Fig. 5. Linear microhardness measurements of Ni_3Al/FeAl/Ni_3Al joint, (a) variant I, (b) variant II

The second joint is characterized by a stabilized microhardness in the region of transient zones, comparable to that of the Ni_3Al foil. Increase in hardness of the analyzed Ni_3Al/FeAl/Ni_3Al system was observed only in FeAl volume. In contrast to variant I, microhardness value of the FeAl intermetallic phase is highly uniform and amounts to 320±8 HV (Fig. 5b).

High quality of the joint made according to variant II was confirmed in a disruption test of the joined foils. Additionally, a successful test of joining of Ni_3Al foils throughout Fe-Al reactive spacer using high-current pulses was performed.

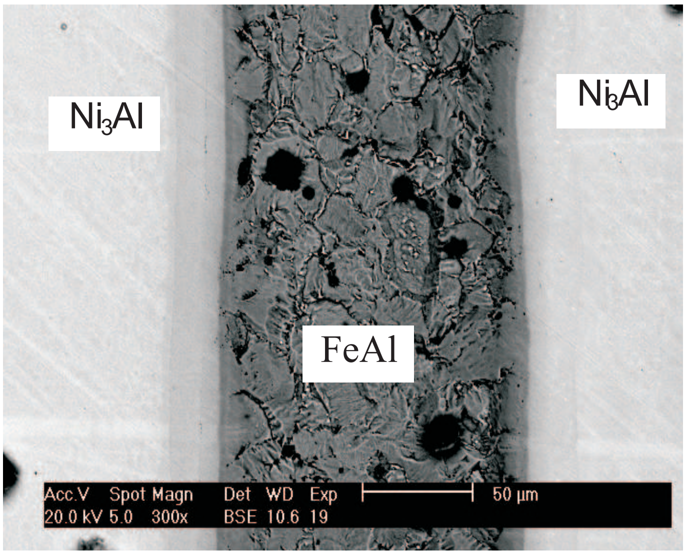

Fig. 6. Microstructure of Ni_3Al/FeAl/Ni_3Al joint after high-current pulses sintering

4. Summary

On the basis of executed investigations one can state that developed method of bonding Ni_3Al foils with a reactive sintering makes obtaining a diffusive coupling between joint elements possible. A fundamental stage of the suggested process is presintering. Properly selected thermal conditions as well as a level and type of load enable volumetric SHS reaction to be initiated (spacer type has no influence on the SHS reaction start energy amount released during reaction). Energy released in the reaction causes a loss of thermal stability of the Ni_3Al foil and an intense mutual diffusion structural components of the foil and reactive spacer. Basic sintering makes reconstruction of highly aluminium phases in the reactive spacer possible. Diffusion range for both technological variants comes to a level of 100 µm; in variant I two transient zones can distinguished while for variant II as much as eight transient zones. A too high number of transient zones can cause a decrease of a joint strength (boundaries of a phase separation create a structural notch). However, basic sintering conditions for the first variant lead to local cracks on a phase separation boundary. Microhardness level measured in the samples exhibits particularly large internal stresses in then joint number 1.

Sintering of Ni_3Al/FeAl/Ni_3Al systems based on high-current pulses is in initial stage. First results of structural investigations show a possibility for volumetric initiation of the SHS reaction and obtainment of qualitatively good joints (after full cycle of heat treatment).

Acknowledgements

This work was financially by The Polish Ministry of Science and Higher Education of Poland (PBG N507 100 31/2403), to whom we are gratefully acknowledged.

Special thanks belong to Professor Michalski from Warsaw Technical University for his preparation of the Ni3Al/FeAl/Ni_3Al joints via high-current sintering.

REFERENCES

[1] D. Trinh, M. Muller, Aluminides, 4H1609 Functional Materials, Project Report, KTH, 2002.

[2] S. C. Deevi, V. K. Sikka, C. T. Liu, Processing, properties, and applications of nickel and iron aluminides, Progress in Materials Science **42**, 177-192 (1997).

[3] S. M. Spearing, Materials issues in microelectromechanical systems (MEMS), Acta Materialia **48**, 179-196 (2000).

[4] C. T. Liu, V. K. Sikka, Nickel aluminides for structural use, Journal of Metals **38**, 19-21 (1986).

[5] P. J ó ź w i k, Z. B o j a r, Analysis of grain size effect on tensile properties of Ni_3Al – based intermetallic strips, Archives of Metallurgy and Materials **52**, 321-327 (2007).

[6] K. K a r c z e w s k i, S. J ó ź w i a k, Z. B o j a r, Mechanism of strength properties anomaly of Fe-Al sinters by compression tests at elevated temperature, Archives of metallurgy and materials **52**, 361-366 (2007).

[7] S. C. D e e v i, V. K. S i k k a, Nickel and iron aluminides: an overview on properties, processing, and application, Intermetallics **4**, 357 (1996).

[8] C. T. L i u, J. S t r i n g e r, J. N. M u n d y, L. L. H o r t o n, P. A n g e l i n i, Ordered intermetallic alloys: an assessment, Intermetallics **5**, 579 (1997).

[9] W. P r z e t a k i e w i c z, T. D u r e j k o, Z. B o j a r, J. B y s t r z y c k i, Sintering of Fe-Al Elemental powders under cyclic loading, Mass and Charge Transport in Inorganic Materials, 341-348 (2003).

Received: 10 March 2009.