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COMPOSITES THEORY AND PRACTICE

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APPLICATION OF LENS METHOD IN Fe40Al + n-Al_2O_3 COMPOSITE MATERIALS FABRICATION

The present study examines the impast of nanoceramics on the structure and selected mechanical properties of intermetallic alloys. In order to determine the grain size in the different variants of materials, with LENS fabricated Fe40Al, an equivalent mean diameter was determined. Resistance against oxidation of the material with and without the addition of nanoceramics was also determined. Observations of the microstructure and phasal analysis did not reveal the presence of nanoceramics in the bulk of the material. However, it was found that the addition of nano-oxide ceramics, increases the grain size and a 4-fold increased heat resistance compared to the reference material. For the alloy without the addition of the oxide nanoceramics, a relative deformation of 2% at a lower yield than the 2% composite Fe40Al vol. n-Al_2O_3 was reported. An attempt to explain the situation it was based on research using the Thermal Imager, which is equipped with a LENS MR-7. It allowed permanent registration of the temperature distribution and weld puddle determining a number of thermodynamic dependences. It additionally drew attention to the fact that during the manufacturing process, differences between the width of the liquid metal mesh for the alloys with and without nanoceramics was observed. It indirectly proves the existence of ceramics on the surface of powder particles at the time of melting the base material, and probably there where an increase in the width of the weld puddle was observed.

Keywords: **intermetallic phase Fe40Al, oxide dispersion strengthennig, LENS (Laser Engineered Net Shaping), Al_2O_3 nanoceramics**

WYKORZYSTANIE METODY LENS DO WYTWARZANIA MATERIAŁÓW KOMPOZYTOWYCH Fe40Al+n-Al_2O_3

W opracowaniu przeanalizowano wpływ nanoceramiki na strukturę i wybrane właściwości mechaniczne stopów na osnowie intermetalicznej fazy Fe40Al wytworzonych techniką LENS. W celu określenia wielkości ziarna w poszczególnych wariantach materiałowych wyznaczono średnią średnicę ekwiwalentną. Określono odporność na utlenianie materiału bez i z dodatkiem nanoceramiki. Obserwacje mikrostruktury oraz analiza fazowa nie ujawniły występowania nanoceramiki w objętości materiału. Stwierdzono jednak, że dodatek nanometrycznej ceramiki tlenkowej wpływa na wzrost wielkości ziaren oraz 4-krotnie podwyższa żaroodporność w stosunku do materiału odniesienia. Dla stopu bez dodatku nanoceramiki tlenkowej odnotowano odkształcenie względne na poziomie 2% przy niższej granicy plastyczności niż kompozyt Fe40Al+2%obj.n-Al_2O_3. Próbę wyjaśnienia zaistniałej sytuacji podjęto w oparciu o badania z wykorzystaniem Thermal Imagera, w który wyposażony jest LENS MR-7. Umożliwia on permanentną rejestrację rozkładu temperatury jeziorka ciekłego metalu i wyznaczania szeregu zależności termodynamicznych. Uwagę zwrócił fakt, że w trakcie procesu wytwarzania zauważono różnice pomiędzy szerokością oczka ciekłego metalu dla stopów z nanoceramiką i bez. Świadczy on pośrednio o istnieniu ceramiki na powierzchni cząstek proszku w momencie przetapiania materiału bazowego i prawdopodobnie tam, gdzie się ona znajduje, obserwuje się wzrost szerokości jeziorka ciekłego metalu.

Słowa kluczowe: **faza międzymetaliczna Fe40Al, stopy ODS (oxide dispersion-strengthened), LENS (Laser Engineered Net Shaping), nanoceramika Al_2O_3**

INTRODUCTION

The ordered intermetallic phases of Ti-Al, Ni-Al and Fe-Al systems, Al_2O_3, Y_2O_3 ZrO_2 reinforced type oxides SiC, TiC and WC carbides are currently under intensive study of their matrix materials. This is particular of composites, which include such steps as: FeAl, Fe_3Al, NiAl, Ni_3Al and TiAl [1]. They are characterized by relatively low density, high melting point and a linear coefficient of thermal expansion comparable to the phases forming the composite matrix [2].

The main objective of the introduction of nanoparticles into an oxide ceramics matrix (Al_2O_3, Y_2O_3, ZrO_2) in ODS (Oxide Dispersion Strengthened) alloys is the increase in high-temperature properties of the base material, mainly resistance to creep, oxidation and recrystallization temperature [3]. These types of alloys combine the properties of intermetallic phases (high specific strength, relatively low density) with the advantages of ceramics (heat resistance, wear resistance) [4]. These materials due to the presence of a stable dispersion of oxides, evenly distributed in the matrix, have a high strength at elevated temperatures. ODS alloys have a fine grained structure (of the order of several micrometers), which is not desirable from the standpoint of resistance to creep at high temperatures. However, as a result of "controlled" secondary recrystallization, elongated grains were produced, significantly increasing the creep resistance of such materials [5].

The authors of [6] presented the results of studies in which it was observed that the component that significantly improves the properties of the high temperature composite material tested on the intermetallic phase matrix of the FeAl systems are Al_2O_3 particles with a nanometric structure. There was also an increase in the tribological properties, including erosive wear resistance of the test material with a decreasing mean particle diameter acting as the strengthening phase. It was also noticed that the introduction of this type of alloys to engineering practice, however, faces significant technological barriers. Currently, materials for the matrix phase of the Fe-Al system are more often made from composite powders containing chemically and thermally stable oxides with a nanometric structure, using the laser technique, incremental LENS (Laser Engineered Net Shaping). Numerous studies have shown that the properties of materials obtained by this method are often better than conventional plastics derived techniques [7, 8]. The main problem at the stage of preparing the charge, is the appropriate introduction of nano-ceramic oxide powder to a volume intermetallic alloy bulk in such a way as to achieve the desired morphology of composite particles, which will enable implementation of the LENS process [9]. The authors of [9] presented a sample selection of technological parameters of the manufacturing process of composite powders of intermetallic phase FeAl and intermetallic nanoceramic doped Al_2O_3 by low-energy milling in a Fritsch planetary mill, so as to obtain a minimum change in the base powder particle size, and a uniform distribution on the surface of the ceramics. The aim of this study was to use the LENS method to produce composite materials of this type to analyse their structure and the influence of the addition of a nanoceramic on the microstructure and mechanical properties.

MATERIALS AND METHODS

Incremental Fe40Al + 0.05% Zr + 50 ppm B with adimensional fraction from 44 to 150 μm, and nano-Al_2O_3 ceramics with an average grain diameter of 35 nm were used to generate the laser sampling technique.

Composite powders Fe40Al+0.05% Zr+50 ppm B + n-Al_2O_3 were obtained by low-energy milling in a planetary Fritsch ball mill. The milling of the powders was conducted in an environmental chamber (the atmosphere). The primary objective of the process was to obtain uniform distribution of the ceramic (anchor) on the surface of the base powder particles, while maintaining the morphology required in the process of shaping by means of the laser incremental LENS technique, i.e, spherical shape, and granulation in the range 44÷150 μm. Based on the analysis of literature and the autors own previous studies [5.10], a 2% by volume of nanometric aluminium oxide was addend to the base Fe40Al powder. The prepared batch was then placed in a cylinder made of tungsten carbide with a capacity of 20 ml and milled in the presence of 4 steel balls with a diameter of 10 mm (grinding media were chosen based on manufacturer's table) with a speed of 200 rev/min for 5, 10 and 15 minutes.

The structure of the composite powders obtained by each of the different technological options were analyzed in detail. It was found that the optimal time of low-energy milling of a metallic-ceramic feedstock, with the assumed speed, is about 10 minutes. It was found that after the milling time, a uniform distribution on the surface of the powder nanoceramics base (Fig. 1) was obtained and the highest volume fraction (approximately 90%) of the desired particle size range of 44÷150 microns (Fig. 2), with a minimal change in particle shape was recorder. In addition, there was no effect of grinding on the chemical composition of the base powder (the process did not cause pollution).

Root	Wt %	At %
O K	02.97	07.27
AlK	34.80	49.98
ZrL	00.60	00.07
FeK	61.63	42.68

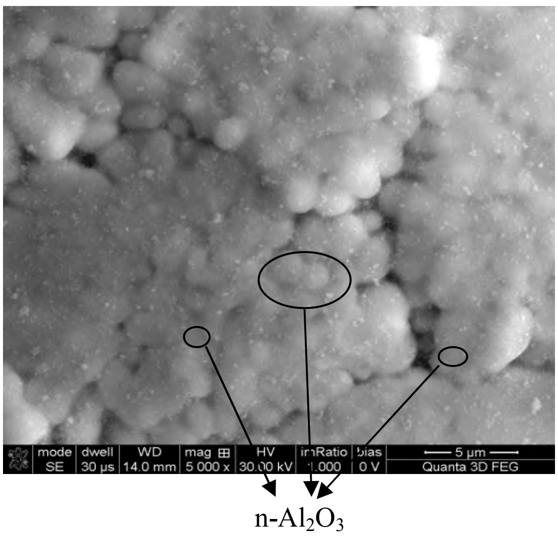

Fig. 1. Morphology and composition of Fe40Al+0.05%Zr +50 ppm. B+ +2% vol. Al_2O_3 powder after low-energy milling for 10 min. at 200 rpm

Rys. 1. Morfologia proszku Fe40Al+0,05%Zr+50 ppmB+2% obj. Al_2O_3 po procesie niskoenergetycznego mielenia przy parametrach 10 min i 200 obr/min

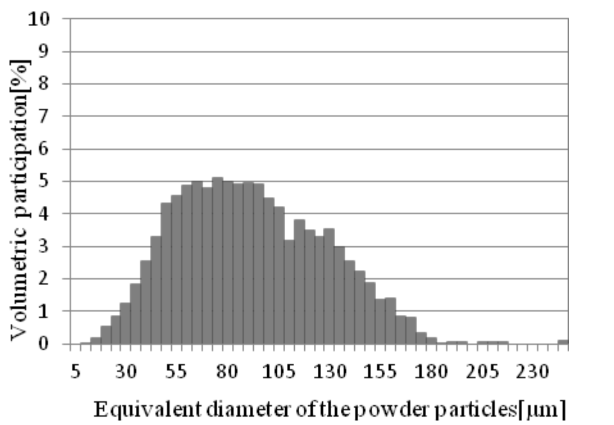

Fig. 2. Histogram of particle size distribution of Fe40Al+0.05% Zr+ 50 ppm B with 2% vol. Al_2O_3 after milling at 200 rpm for 10 min. during analysis of IPS U

Rys. 2. Histogram rozkładu średnicy cząstek proszku Fe40Al+0,05% Zr+ 50 ppm B z dodatkiem 2% obj. Al_2O_3 po mieleniu z prędkością 200 obr/min w czasie 10 min na podstawie analizy IPS U

The research material was applied to the sandblasted surface of a substrate made of Armco iron. The manufacturing process was carried out by controlling the laser power, "focus position" to the ground, and the powder feed rate at a constant, regulated flow. For the principal research, the selected composite material with the lowest relative porosity (equal to 1.42%) and reference material (without the addition of nanoceramics) were made using the same technological parameters of the LENS process, i.e. 300 W laser power, powder flow rate of 0.1 rpm and a scan order of 3.9 mm/s. The samples intended for testing the mechanical properties were polished on a grinding-polishing Mintech Presi Z65, and then polished using diamond slurry. In order to confirm the structural chan ges, stereological analysis was performed using a computerized image analyzer, NIS-Elements BR 3.0, on micrographs taken with an optical microscope Nikon DS-U1. Metallographic studies were performed using a scanning electron microscope FEI Quanta 3D FEG. The phase composition of alloys with and without the addition of nanoceramics was identified by X-ray diffraction. TG Experiments were carried out with a Setar in TG. The samples were heated at a rate of 50°C/min, and then isothermally annealed for 24 hours at 800°C in an atmosphere of synthetic air. The analysis of the mechanical properties was carried out by Vickers hardness tests and a static tensile test, realized in a universal testing machine Instron type 8,501, cut using wire EDM. The samples cut using EDM wire were tested in the state immediately after the formation by means of the LENS technique, and after annealing at 400°C for 100 h in order to reduce the amount of thermal vacancies.

RESULTS AND DISCUSSION

Analysis of the microstructure of the obtained samples showed that they are characterized by a heterogeneous size and shape of grains. The volume of the material can be divided into three distinctive zones which are conventionally named areas: at the edge, in the volume and the substrate (Fig. 3). The difference in the mean grain diameter is mainly due to the directional heat removal during the process of applying successive layers of the built mass. The largest equivalent diameter, for both the reference material and composite were recorded at ground level (Table 1). It is worth noting that the grain size for the alloy with the addition of nanoceramics oxide is approximately 2-fold higher, and for an area in the volume of up to 3-fold, compared to "pure" Fe40Al. The calculated values of the aspect ratio α (ratio of maximum diameter to equivalent) showed that in the zone at the base, its value fluctuates around 0.4, which shows columnar, highly elongated grains (Fig. 3c). The other two areas of the microstructure of the samples is similar to uniaxial character.

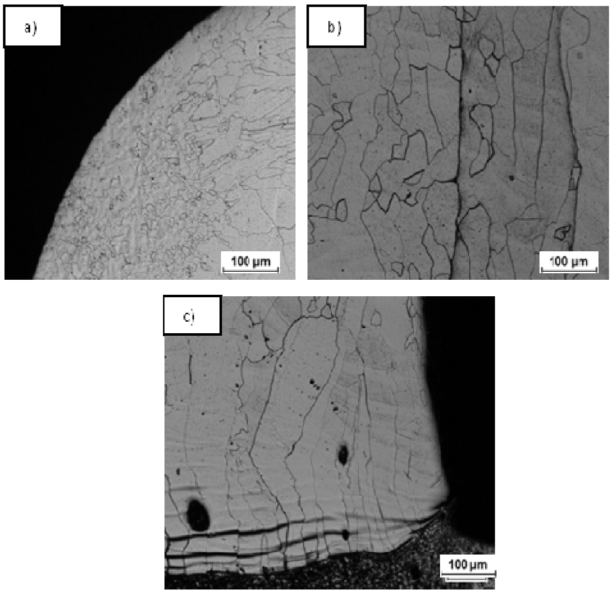

Fig. 3. Microstructure of alloy Fe40Al 2% vol. and Al_2O_3: a) at edges, b) in volume, c) base

Rys. 3. Mikrostruktura stopu Fe40Al+0,05% Zr+50 ppm B+2%obj. Al_2O_3: a) przy krawędzi, b) w objętości, c) przy podłożu

TABLE 1. **Value of grain equivalent diameter for alloy without and with addition of Fe40Al nanoceramics**

TABELA 1. **Wartość średnicy ekwiwalentnej ziarna dla stopu Fe40Al bez i z dodatkiem nanoceramiki**

Analyzed areas	Alloy Fe40Al	Fe40Al + n-Al_2O_3 alloy
at edge	16.56±4..49	36.80±7.08
in volume	12.41±2.21	48.54±4.74
at base	22.69±6.26	53.53±4.22

Despite the high heterogeneity of morphological structures in the analyzed material, no fluctuations in chemical composition were observed. The homogeneity in terms of phase, in turn, was confirmed by X-ray diffraction experiments. On the diffractogram in both cases, reflection from the plane (100) originating from orderly intermetallic phases was observed. There was also a lack of strengthening of the X-ray crystallographic planes for oxide ceramics (Fig. 4), suggesting a strong texturation of the investigated alloys.

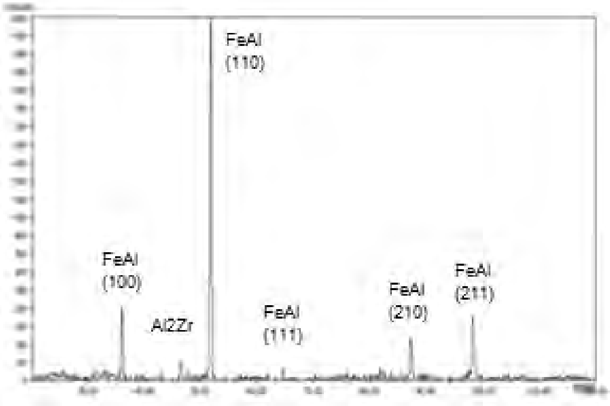

Fig. 4. Results of X-ray phase analysis of alloy 2% vol. Al_2O_3+Fe40Al

Rys. 4. Wyniki rentgenowskiej analizy fazowej stopu Fe40Al+ +2% obj. Al_2O_3

On the diffractogram obtained for the composite material reflections originating from the oxide Al_2O_3 ceramics (Fig. 4) were observed. This was confirmed by the results of structural studies, there was no presence of ceramic particles in the intermetallic matrix either. This may indicate a too low "sensitivity" of the applied research methodologies. On the Rother hand, the peak was observed coming from the Lavesa Al_2Zr stage. The appearance of this phase is the result of the admixture of Zr in the starting powder Fe40Al+0.05% Zr+ 50 ppm B. Confirmation of this was the analysis of the chemical composition of the particles and impurities, where we observed a marked increase in thev compactness of Zr.

Due to the lack of direct confirmation of the presence of particles in the volume of alloy nanoceramics, we decided on an indirect method to assess its possible impact, i.e. the evaluation of heat resistance. Both feeds, without and with the addition of n-Al_2O_3, were therefore tested for resistance to oxidation, using a calorimeter working in the thermogravimetric mode. The kinetics of isothermal annealing at 800°C in air atmosphere for 24 hours was analyzed on the basis of weight change per unit area of the sample as a function of time. For each of the tested alloys weight gain was recorded. It was observed that with increasing temperature at the surface of the test materials an Al_2O_3 passive oxide layer appears and its thickness increases with annealing time, until they reach full stability in the surrounding conditions. The change in weight of the composite material reaches a plateau after about 3 hours of annealing, while Fe40Al after about 5, and the increase in its mass per unit area is 4-fold higher than for the alloy doped with nanoceramics (Fig. 5).

The cross-sectional study of hardness by Vickers samples showed that the analyzed parameter stands at 350 ± 15 HV10 for the composite, which is lower by about 50 units in relation to the reference material (Fig. 6). This difference can be partly explained by strengthening of the alloy Fe40Al derived from the grain boundaries (approximately 3-fold less than the equivalent diameter of grains in relation to the composite Fe40Al+2% vol. n-Al_2O_3). The high level of hardness recorded for the tested materials is a result of high internal stresses resulting from the specifics of LENS technology and saturation of the alloys with thermal vacancies. All the performed imprints were in the volume of the grain.

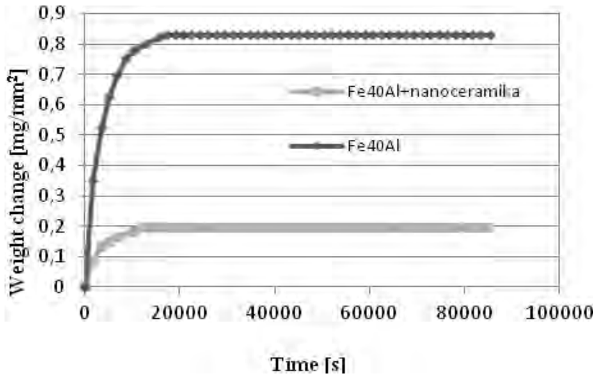

Fig. 5. Thermogravimetric curves (800°C, 24 h) for matrix alloy phase nanoceramics Fe40Al without addition of Al_2O_3 and addition of 2% vol. nanoceramics Al_2O_3

Rys. 5. Krzywe termograwimetryczne (800°C, 24 h) dla stopu na osnowie fazy Fe40Al bez dodatku nanoceramiki Al_2O_3 i z dodatkiem 2% obj. nanoceramiki Al_2O_3

Fig. 6. Hardness for selected technological options

Rys. 6. Twardość badanych materiałów dla wybranych wariantów technologicznych

In order to determine the mechanical properties of the obtained materials, the prepared samples were subjected to a static tensile test. The test was carried out for alloys without the addition of nanoceramics and done immediately after formation with the LENS technique and subsequent 100h-long relaxing heat treatment at 400°C. On the basis of the tensile curves, we can define the basic strength parameters such as: strength $R_{0.2}$, tensile strength Rm and deformation Ac.

Based on the obtained results, it was observed that the addition of nanoceramics increases the yield strength and tensile strength, while depreciation of the deformation in relation to the variant without the admixture of n-Al_2O_3. For both samples Fe40Al recorded a total deformation of about 2% while that sample after annealing shows a greater reserve of strength by about 0.4%. In the case of alloy doped nanoceramics annealing has a beneficial effect on the value of deformation (an increase of approximately 0.2% compared to the option immediately after preparation) and increases the yield of both conventional as well as tensile strength (Fig. 7).

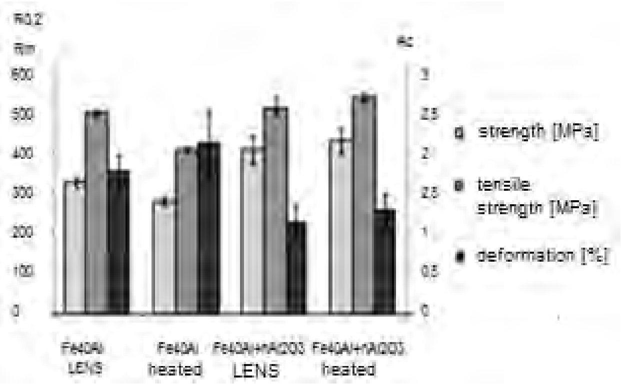

Fig. 7. Summary of parameter values obtained during static tensile test

Rys. 7. Zestawienie wartości parametrów uzyskanych podczas statycznej próby rozciągania

weld puddle during the manufacturing process showed differences between the width of the liquid metal mesh for the alloys with and without nanoceramics. In the case of the alloy with the addition nanoceramics, the weld puddle width clearly increases (Fig. 8), probably due to the complete dissolution of the ceramic particles in the lake of molten metal. Therefore attempts to use Al_2O_3 oxide with an increased grain size and assessment of the cooling rate with the use of thermal imager, which is an integral part of the LENS MR-7device technology are planned in future.

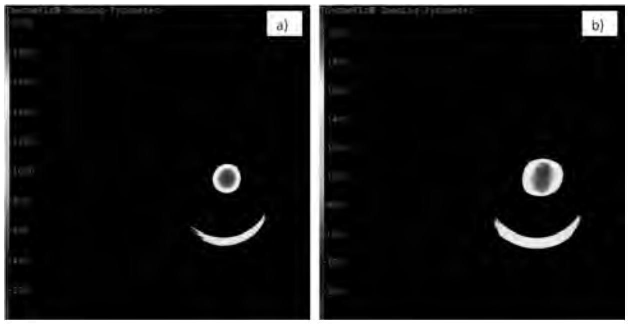

Fig. 8. Temperature distribution in lake of molten metal alloy: a) without nanoceramics, b) with nanoceramics [11]

Rys. 8. Rozkład temperatury w jeziorku ciekłego metalu stopu: a) bez nanoceramiki, b) z nanoceramiką [11]

CONCLUSIONS

Based on comprehensive analysis of the obtained results it was found that a low-energy milling process allows the formation of spherical composite Fe40Al--nAl_2O_3 powders. Properly selected grinding parameters allow us to gain uniform distribution on the surface of aluminum oxide powder particles Fe40Al+0.05% Zr+50 ppm B, with simultaneously, a minimal change in their morphology, which is important for LENS technology. Direct identification techniques, ie, microscopic observations and detailed study of the final Prasal composition in the material (produced by LENS) did not reveal the presence of Al_2O_3 particles in the volume of the nanoceramics received material. However, it was found, that in the nanoceramics doped material, the average grain diameter is about 3-fold greater than for the reference alloy. The most spectacular effect was observed in samples tested for heat resistance. Four times less weight gain per unit area for the alloy was observed. Mechanical tests have shown that the hardness of the obtained alloy is higher for the FeAl alloy, at a lower proof strength and greater relative strain of about 2% after relaxing heat treatment.

Based on the obtained results it was stated that soluble nanoceramics in the lake of the molten metal application process thereby increase the thermal capacity of the material, resulting in hyperplasia of the grains relative to the reference alloy. The temperature control

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