MICROSTRUCTURE OF Fe-Al BINARY COLD SPRAY THICK DEPOSIT AFTER ANNEALING

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Abstract

Examined FeAl deposits were produced by low-pressure cold spray technique using heated air (300 °C, 1.5 MPa) as working gas. Although metal powders were mixed in ratio 60/40 at.% Fe/Al, the final chemical composition of the 8-10 mm thick deposits changed strongly favouring aluminum (approximately 80 at.% Al).

Samples consisting of the deposited material were annealed isothermally in argon protective atmosphere. All annealing processes ran at temperatures from 250 °C to 750 °C for two hours. The overall change of material character during annealing was evaluated: changes in the character of fracture behavior, local micro hardness of the evolving phases and residual powder particles. Microstructure and chemistry of the newly formed phases was evaluated using analytical electron microscopy.

First occurrence of intermetallic phases was observed at 550 °C. The evolved phases incurred by diffusion mechanism and included non-equilibrium and stable phases close to Al_3Fe, FeAl, Al_5Fe_2 and Al_2Fe. The general shape of the samples annealed at temperatures exceeding aluminium melting temperature was preserved due to a rapid formation of intermetallic skeleton. The growth of external dimensions and associated porosity evolution occurred during the treatment. In this paper, overall feasibility of the application of the presented technique for technically applicable materials and the aforementioned growth of external dimensions with porosity evolution are discussed.

Keywords: Cold spray, reaction synthesis, intermetallics, diffusion, metallic foam

1. INTRODUCTION

Intermetallic phases typically exhibit strictly different crystallographic arrangement than the metallic elements form which they are constituted. Intermetallics also usually have high strength and lower ductility - characteristics that are connected to the crystal lattice type. Aluminides generally form very interesting group of materials that are considered to have high potential as material for high temperature application with good strength/density ratio. Iron aluminides - FeAl and Fe_3Al phases are attractive for their properties and also because the raw materials are relatively cheap. [1, 2]

Manufacturing of aluminides usually is limited to precise casting or casting and machining. In this study, the Cold Spray technique is used for deposition of arbitrarily shaped samples of material. [4-7]

Cold Spray is a technique using gas that expands in Laval-type jet to super-sonic speeds as carrier medium for accelerating individual particles of the deposited powder which weld locally upon impact on the substrate or previously deposited particles. The fact that the particles are not melted during flight and at impact and that oxidation of the particles surface is minimal establishes this method not only as surface engineering method but also for bulk material deposition method, where high volumes of material ca be deposited [8].

2. EXPERIMENTAL SETUP

Cold Spray technique was used to deposit the analysed material. A powder previously mixed form pure Fe and Al powders were used as feedstock. The elemental powders were mixed in the 60 at.% Fe : 40 at.% Al ratio. Air was used as the carrier gas, temperature 300 °C and pressure 1.5 MPa. Samples in the form of bars were deposited by 20 consecutive passes of the nozzle. The resulting material had thickness of approx. 1 cm (**Fig. 1**).

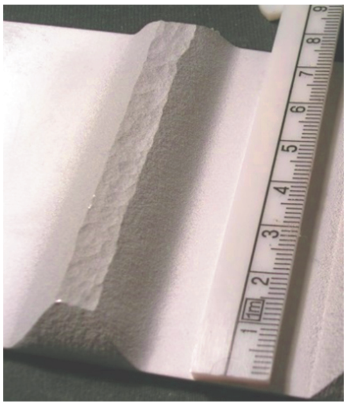

Fig. 1 Overall appearance of the deposited material

This material was cut using low speed saw to samples 5x6x7 mm big which were then used for annealing experiments. The annealing was done in tube furnace under flowing Ar (0.5 l/min, purity 4N8) protective atmosphere. Temperature as measured directly at the sample which was placed into 100 g heavy stainless steel holder that could be easily introduced and retracted from the hot zone of the furnace. This setup roved to provide both good control of the sample temperature and homogenous temperature field for the treated sample. Before annealing the substrate, which was commercially pure aluminium sheet was removed, so that only the deposited bi-metallic CS deposit was annealed.

Following temperatures were used: 250, 350, 450, 550, 650, 700 and 750 °C. The samples were heated fast to the desired temperature; 2 hours were used for the isothermal annealing. After two hours, the samples were moved out from the hot zone, but still left in the protective Ar atmosphere until cooled to 100 °C. Only then were the samples left to cool on air.

Annealed samples were fractured and one piece of the material was used to prepare a metallographic sample by standard grinding and polishing techniques. No etching was used, but colloid silica (OPS from Struers) was used for the last polishing step, which provided also visible microstructure.

Light microscopy and electron microscopy were used to evaluate both fracture surfaces and the polished metallographic samples microstructure (Zeiss Z1M light microscope, Zeiss UltraPlus FEG-SEM). For chemical analysis, local and mapping EDS analyses were used (OXFORD EDS mounted on the UltraPlus). XRD analysis was used for phase occurrence determination (Philips Xpert).

Micro-hardness measurements of the individual grains and microstructure constituents were used to provide also mechanical property despite the small size of the samples. Vickers indenter and F_1 = 0.09807 N or F_2 = 0.1961 N were used as the indenting forces for 10 seconds.

3. EXPERIMENTAL RESULTS

The as sprayed material shows strong change in the average composition when comparing to the original feedstock composition, this has changed form 60/40 at.% Fe/Al to 17/83 at.% Fe/Al. The deposition efficiency of iron in this case was much lower than that of aluminium (**Fig. 2**).

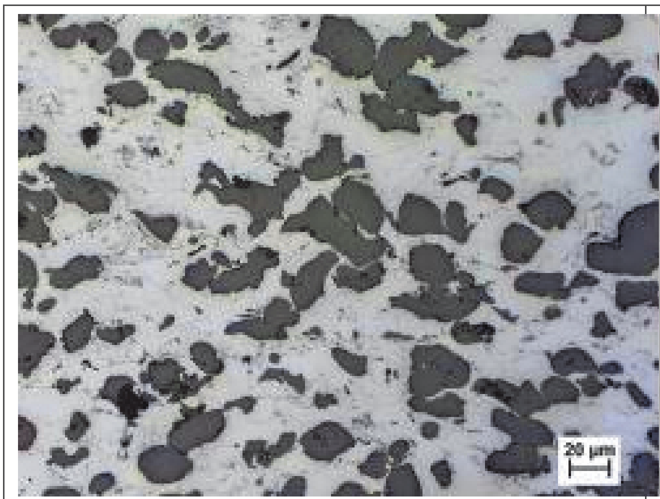

Fig. 2 Microstructure of the as deposited material dark phase Fe, light phase Al

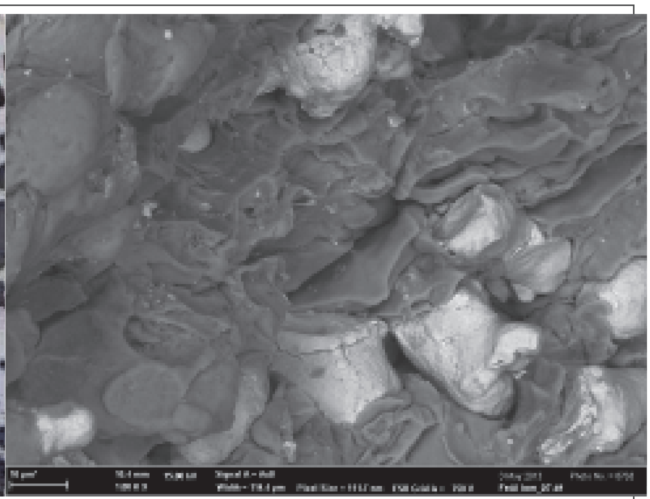

Fig. 3 Fracture of the as deposited material

The fracture surface of the as-sprayed material shows nicely that aluminium, although severely deformed by the deposition process, still possess some plasticity and produces ductile fracture, whereas the iron particles do not break at all and the fracture surfaces contain delaminated iron particles where the iron particles and the aluminium matrix have separated (**Fig. 3**).

3.1 Annealing at 250 - 500 °C

No new phase has been identified at temperatures 250, 350, 450 and 500 °C. Annealing at these temperatures caused changes in the micro hardness in individual iron and aluminium particles. This can be seen in **Table 1**, where all values are decreasing from the initial as-sprayed level. This is in agreement with expectations when we consider that we anneal material that has been plastically deformed and now is gradually recovering when annealed. One exception in the monotonous decrease is the case of aluminium particles hardness after annealing at 550 °C. The value is higher than the hardness value for 450°C. We expect this is due to diffusion of iron atoms into the aluminium. This creates solid solution strengthened by substitution mechanism. The fracture surface character is unchanged in all samples with lower annealing temperatures. The fracture surface is again composed of ductile fracture of the aluminium and delamination of the iron particles.

Table 1 Microhardness values of Fe and Al particles

Temperature	**Fe particles**	**Al particles**
(°C)	HV 0.025	HV 0.01
0	596	56
250	512	47
350	434	35
450	373	34
550	281	42

3.2 Annealing at 550 - 750 °C

At 550 °C new phases were formed around the iron particles by mutual diffusion (**Fig. 4**). By local chemical analysis compositions of the level $Al_{70}Fe_{30}$ were measured (**Fig. 5**). Using XRD Al_5Fe_2 peaks were identified in the diffraction spectrum. Only minor part of the iron particles have reacted after two hours of annealing so still both original iron and aluminium form the most of the sample.

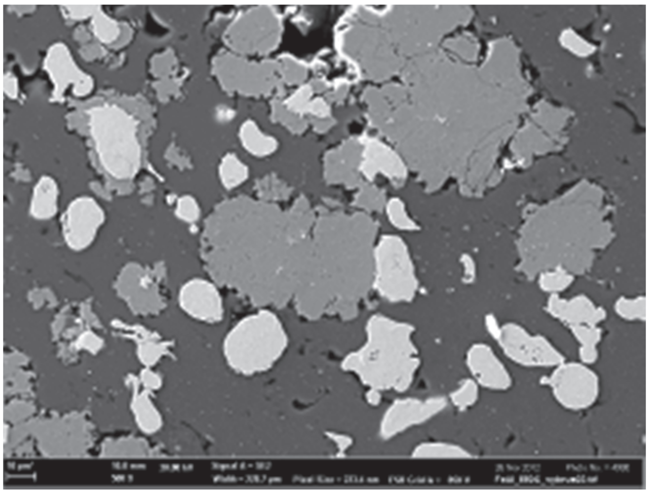

Fig. 4 Microstructure of the material after 550 °C
Darkest Al, grey *$Al_{70}Fe_{30}$*, lightest Fe

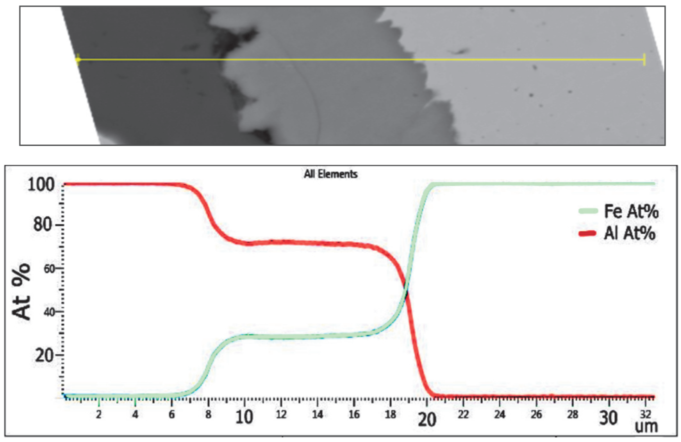

Fig. 5 Analysis of the intermetallics formed at 550 °C

The next annealing temperature - 650 °C is very close to eutectic temperature of the Fe-Al system, which is 655 °C and also quite close to melting temperature of pure aluminium, which is 660 °C. Therefore some local melting could be anticipated. Extreme pores evolved in the material. Instead of bulk material with some inherent porosity, the material changed its character into open foam-like structure. The outer shape of the sample was nevertheless maintained, although the macroscopic dimensions of the sample increased. The microstructure consisted mainly of lamellar mixture of two phases with average inter-lamellar distance of 0.36 µm (**Fig. 6**). The chemical composition of the mixture was determined to be $Al_{60}Fe_{40}$ so the two phases forming the lamellas can be estimated as FeAl and $FeAl_2$ (**Fig. 7**). Some areas with homogeneous structure were identified to be Al_5Fe_2 and also some isolated small remnants of iron could be seen in the microstructure. The fracture surface has changed according to the new character of the material. The fracture surface has brittle character (**Fig. 8**).

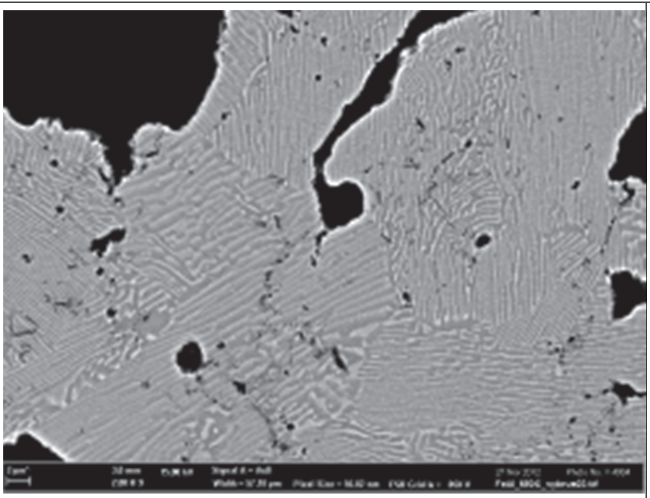

Fig. 6 Microstructure of material annealed at 650 °C

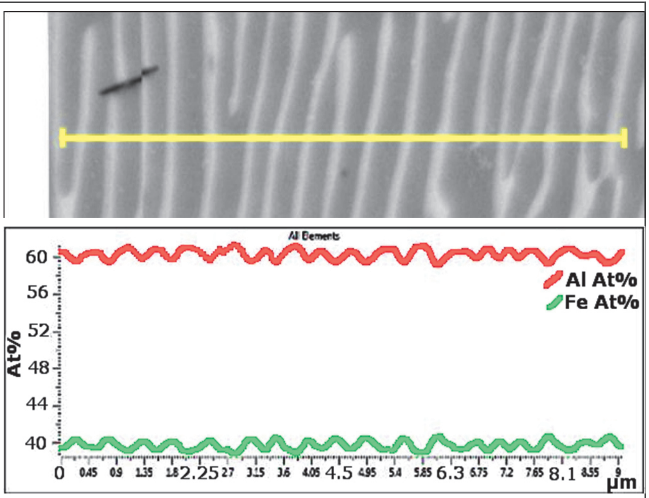

Fig. 7 Analysis of the intermetallics formed at 650 °C

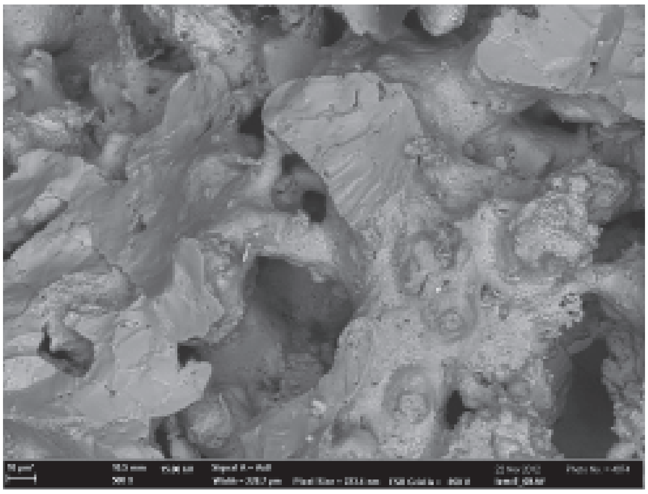

Fig. 8 Fracture of material annealed at 650 °C

At 700 and 750 °C the material is heated to temperatures exceeding both eutectic temperature and aluminium melting temperature by significant difference. In spite of this, the samples did not change into drop of liquid. The overall shape of the samples was kept, only the outer macroscopic dimensions grew bigger. The new microstructure was again formed by high percentage of open interconnected porosity and metallic material in majority consisting of lamellar microstructure. Together with the lamellar $FeAl/FeAl_2$ microstructure, homogeneous grains decorated by network of phase identified as Al_5Fe_2 were found in the microstructure. The average inter-lamellar distance is 0.38 μm for 700 °C and 0.34 μm for 750 °C. Some areas with higher iron content with small areas that represent the last undissolved iron particles were present in both samples annealed at the highest temperatures. Hence two hours of annealing at 700 and 750 °C were not sufficient to homogenize the chemical composition across the sample. The fracture surfaces again reveal high amounts of porosity and brittle character of fracture of the metallic material.

4. SUMMARY

The samples annealed at temperatures lower than 450 °C did not exhibit any measurable new phase formation. Only gradual change of local hardness of the aluminium and iron particles was observed, which can be attributed to deformation strengthening healing. First occurrence of intermetallic phases has been found after 550 °C annealing, which is in accordance with the previously published thermal analysis papers [9-12].

Distinctively lamellar microstructure has been created constituting of FeAl and $FeAl_2$ phases at temperatures higher than 650 °C, similar to [13]. Samples with the lamellar microstructure had open porosity character. The porosity can be attributed to Kirkendall solid state diffusion at lower temperatures [14] and to liquid-solid reaction of aluminium rich melt and the more iron rich intermetallics or pure iron particles.

The porous intermetallic material manufactured by this approach can be used further for aluminium matrix reinforcement by melt infiltration or simply as is for filtering or catalyst support for high temperatures taking advantage of the high temperature resistance of the intermetallic phases and the high specific surface of the open porosity.

CONCLUSIONS

Iron aluminides were created by annealing of elemental powders cold spray deposit. Uniform porosity materials were created at repeated attempts. Since the cold spray technique allows to form any shapes and sizes of the deposits, the material can be considered for technical applications rather than similar products achieved by pressing or free powder reactions.

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