Ni_3Al-Mo MULTILAYER COMPOSITES: MANUFACTURE, STRUCTURE, PROPERTIES

Povarova K.B.[1], Burmistrov V.I.[1], Antonova A.V.[1], Skachkov O.A.[2], Kiiko M.V.[3]

[1] Baikov Institute of Metallurgy and Material Science, Russian Academy of Sciences, 119911 Moscow, Russia
[2] Bardyn Scientific-Technical Centre Materials and Technology, Russian Federation. 107005 Moscow, Russia
[3] Institute of solid state physic, Russian Academy of Sciences, 142432 Chernogolovka, Russia

ABSTRACT

The experimental samples of multilayer composite material (MLCM) consisting of 25 layers Ni_3Al and 28 layers Mo were obtained by the following technology: hot isostatic pressing (HIP), hot rolling (HR), and cold rolling (CR) to a total degree of reduction of 95.8%. After HR, the layered structure is retained, but the layers become thinner. After CR, the Mo layers in the longitudinal direction become "wavy", remaining of virtually uniform thickness, whereas the Ni_3Al alloy layers become "extruded" between the Mo layers, forming regular structure consisting of alternating thickenings and reductions across the rolling direction. Annealing upon HIP and further CR to ε = 98.2% lead to the formation of the zones with disrupted layering. This is caused by different resistances of the Ni_3Al and more refractory Mo layers to room-temperature deformation. The cohesion between the layers is strong, and the interphase interface is free from any intermediate phases. The ultimate bending strength of the plates 2.3 mm thick is 1000, 256, 170, and 100 MPa at 20, 1000, 1100 and 1200°C, respectively. The samples 2.5 and 0.22 mm thick can be bent at 20°C by an angle of ~55 and ~60°, respectively.

INTRODUCTION

The idea of the design of composite material (CM) with the nickel aluminide matrix reinforced by the fibers of refractory high-temperature metal such as Mo or W has long ago attracted the attention of the developers of high-temperature materials [1]. Mo (density ρ = 10.2 g/cm^3, melting point T_m = 2620°C) and more refractory and heavy W (ρ = 19.4 g/cm^3, T_m = 3410°C) need the protection from oxidation, whereas lighter Ni_3Al and NiAl (ρ = 7.3 and 6.0 g/cm^3, respectively) which are less refractory (T_m = 1395 and 1630°C, respectively) are very oxidation-resistant and do not need any protective coating. Several examples of the development of such CM are known [1]. In [2, 3], the W/Ni_3Al and W/NiAl composites were prepared by the methods of powder metallurgy according to the following scheme: spray deposition of the Ni_xAl_y matrix powders, which were obtained by coreduction with calcium hydride (RCH), onto a mount with tungsten wire. These CM are free from disadvantages typical of the CM of the Ni superalloy/W type: no wide W_xNi_y reaction zone is formed, and no nickel-induced recrystallization of the deformed tungsten fiber is observed, i.e., at least two disadvantages leading to the degradation of W/Ni composites are virtually got over. This made it possible to obtain high-temperature oxidation-resistant 20W-80NiAl CM with a 100-hour strength σ_{100} = 180-200 MPa at 1200°C. The 40W-60Ni_3Al composite obtained by deformation of W bars put into the Ni_3Al powder matrix prepared from RCH powders also demonstrated a high heat resistance. It is known that the natural eutectic CM NiAl/9Mo, NiAl/34(Cr, Mo) with a high creep strength (σ = 90-160 MPa) at 1300 K at a creep rate $\varepsilon = 10^{-6}$ cm^{-1} and a satisfactory room-temperature fracture toughness K_{1c} = 15-21 MPa $m^{1/2}$ can be obtained by the method of direct solidification [4]. The assortment of eutectic CM is limited by the method of their production.

The nickel-aluminide based CM reinforced by molybdenum is the promising material, which combines the heat resistance of molybdenum (at a density below that of tungsten) with the high oxidation resistance of nickel aluminide. It seems to be very attractive to use the Ni_3Al aluminide as the intermetallic (IM) component of CM because it is more ductile than monoaluminide. The relationship between the heat-expansion coefficients of the Ni_3Al and Mo pair (12.3-14.1 10^{-6}, K^{-1} and 7.5-8.2 10^{-6}, K^{-1} respectively) is better than that of the Ni_3Al and W pair. This should increase the CM stability upon thermocycling.

No information about the manufacture of the Ni_3Al-Mo type multilayer composite material (MLCM) is available. The production of the new type of high-temperature oxidation-resistant MLCM with enhanced plasticity is necessary for the production of the panels of complex profile, honeycomb panels, and other power structural parts of modern flight vehicles requiring the materials, which can operate at temperatures above the service temperatures of conventional nickel superalloys [5].

Therefore, the aim of the present work was to study the specific features of the manufacture of the Ni_3Al/Mo MLCM and to estimate some of its mechanical properties. It was assumed that the interaction between the MLCM components during the diffusion welding upon hot consolidation and hot rolling (HR) should according to the ternary phase diagram of the Ni_3Al-Mo system, which was analyzed in [6]. The Mo solubility in γ'-Ni_3Al is 5-7 at %, where Mo substitutes both Al (predominantly) and Ni positions in the ordered Ni_3Al ($L1_2$ type) lattice; the solubilities of Ni and Al in Mo do not exceed 1.8 and ~5 at.%, respectively; no intermediate IM, which can weaken the interphase interface, are formed in MLCM. Both components of MLCM have the reserves of technological effectiveness and low-temperature plasticity.

2. EXPERIMENTAL

2.1. Initial materials.

The Ni_3Al-based alloy ribbon 100 μm thick was obtained by two methods: (1) the ribbon up to 200 μm thick obtained by melt quenching in rotating rolls was subjected to cold rolling (CR) [7, 8] and (2) the plates obtained by roll compacting of the Ni_3Al powder obtained by the method of RCH were subjected to CR after vacuum sintering. The mechanical properties and the technological characteristics of the ribbons obtained by both methods made it possible to recommend them for the production of MLCM. The Mo ribbon 200 μm thick was prepared by conventional industrial technology.

2.2. Manufacture of MLCM billets

The ribbons were cut into cards of 25 x 85 mm in size, then alternating 28 Mo and 25 Ni_3Al cards were collected into a packet with double Mo cards in the middle section. The packet was put into a steel capsule prepared from a sheet 2 mm thick. The capsule was degassed and hermetically sealed in a vacuum system equipped with a cathode-ray gun in a vacuum of 10^{-4} - 10^{-5} mm Hg. The capsule with the card packet was subjected to hot isothermal pressing (HIP) in a gasostat at 1200°C at a pressure of 150 MPa for 2.5 h.

2.3. Rolling

The pressed billet with the steel shell was rolled in a hot mill at a temperature of 1050-950°C at a reduction of 2-0.8 mm per pass to a thickness of 2.3 mm (the total degree of reduction ε = 80.8%). Samples for the bending tests and metallographic examination were cut from the rolled billets. The remaining part of the billet was rolled in a "quartos" cold mill from 2.3 to 0.5 mm (the total degree of deformation ε = 95.8%) without intermediate annealing. The sample 0.5 mm thick was annealed upon HIP in a gasostat at 1200°C at a pressure of 150 MPa for 2.5 h, then the sample was rolled in the "quartos" cold mill from 0.5 to 0.22 mm without intermediate annealing (the total degree of deformation ε = 98.2%).
The microhardness was measured with a PMT-3 tester at a load of 100 g. The bending tests of the samples 2.3 mm thick were performed according to the scheme of three-point bend at room temperature (RT) with an IR-5047-50-11 machine at a loading rate of 1 mm/min and at temperatures of 1000, 1100, and 1200°C in vacuum with a unit NIKIMP-32-02.

3. STRUCTURE OF MLCM

The samples of alloys and MLCM for the examination of their structure and phase composition were ground and polished by conventional techniques and then chemically etched in an aqueous solution of acids, 2% HNO_3 + 2% HF. The microstructure was examined with a "Neophot-32" optical microscope and an "LEO-430" scanning electron microscope (SEM). The distribution of elements in the structure constituents was determined by X-ray electron microprobe analysis in the SEM with a microanalyzer.
The characteristic microstructures of the MLCM 2.3, 0.5, and 0.22 mm thick in the longitudinal and transverse directions are given in Fig. 1. In the LCM 2.3 mm thick obtained by HIP and HR, the Ni_3Al layer thickness is ~20-25 μm, and the Mo layer thickness is ~44-48 μm (Figs. 1a and 1b). Thickness of the layers in the rolling direction and the transverse direction of each material is uniform, and the relationship between the thicknesses remains Mo/Ni_3Al~2/1. This shows that both materials of the CM have close characteristics of resistance to deformation upon HR.
A principally different structure is formed upon CR of the ribbon from a thickness of 2.3 mm to 0.5 mm, i.e., from ε = 80.8 to ε = 95.8%. The longitudinal samples (CR direction) show that the Mo layers become of a "wavy" structure with the sites of the joining of similar layers and the formation of "cells" (Fig. 1c). In this case, the thickness of the molybdenum layer does not substantially change, whereas the Ni_3Al alloy layers "are extruded" between the Mo layers, forming a regular structure of alternating thickenings and reductions across the rolling direction of (in parallel to the generatrix of the roll). For the samples representing the transverse direction, this manifests itself in a substantial variation in thickness of the layers, especially of the Ni_3Al ones (Fig. 1d). The MLCM regions with double molybdenum plates are seen in the microstructure.
Annealing upon HIP and further CR to 0.22 mm of the plates with the wavy structure lead to the formation of the zones with destroyed layering of material and the "curled" structure in the longitudinal and transverse directions (Figs. 1e and 1f). This can be caused by different resistances to room-temperature deformation of the layers of Ni_3Al and more refractory Mo. The regions with the "curled" structure interstratify with the regions of layered structure throughout the entire area of the plates.

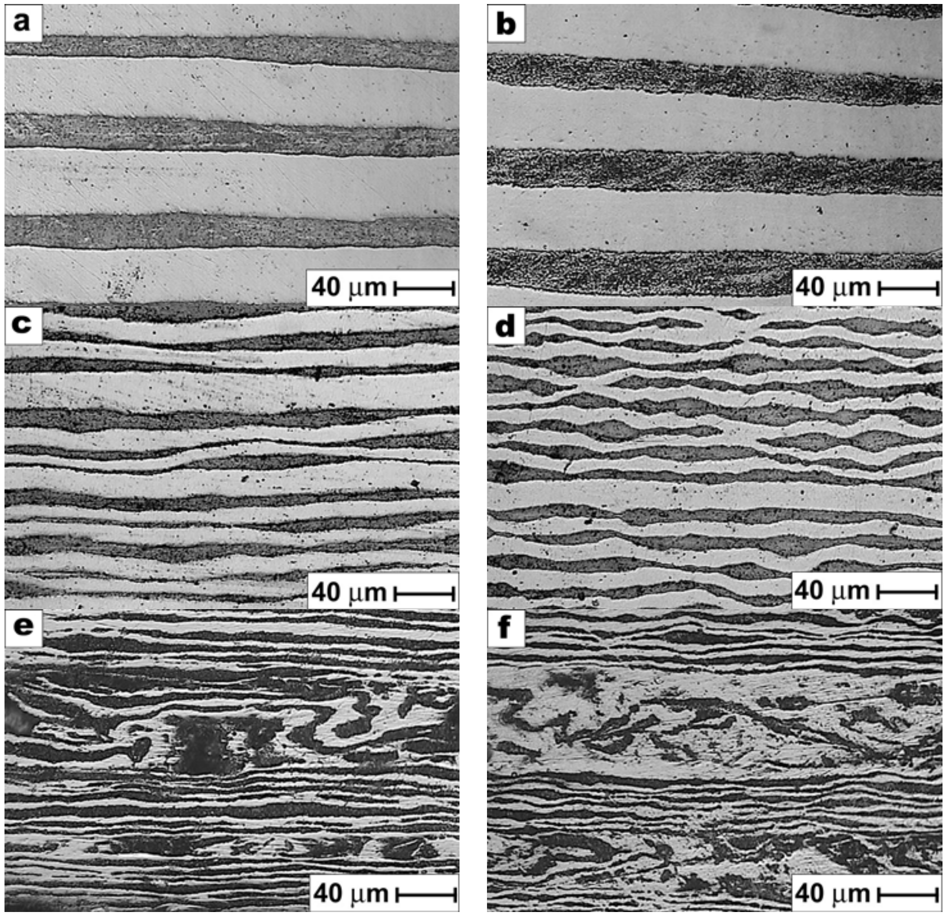

Fig. 1. Characteristic microstructures of the LCM (a, b) 2.3 mm, (c, d) 0.5 mm, and (e, f) 0.22 mm thick in the (a, b, e) transverse and (b, d, f) longitudinal directions.

The microstructure analysis shows a strong cohesion between the layers in MLCM of any investigated thickness and the absence of any intermediate phases at the interphase interfaces. This confirms the correctness of the selection of MLCM components, which according to the Ni-Al-Mo ternary diagram are in equilibrium with each other. Therefore, the interaction between the MLCM components during the diffusion welding upon HIP and HR occurs in accordance with the ternary phase diagram of the Ni_3Al -Mo system [5].
The distribution of elements in MLCM 2.3 mm thick according to the data of X-ray electron probe analysis is given in Fig. 2.

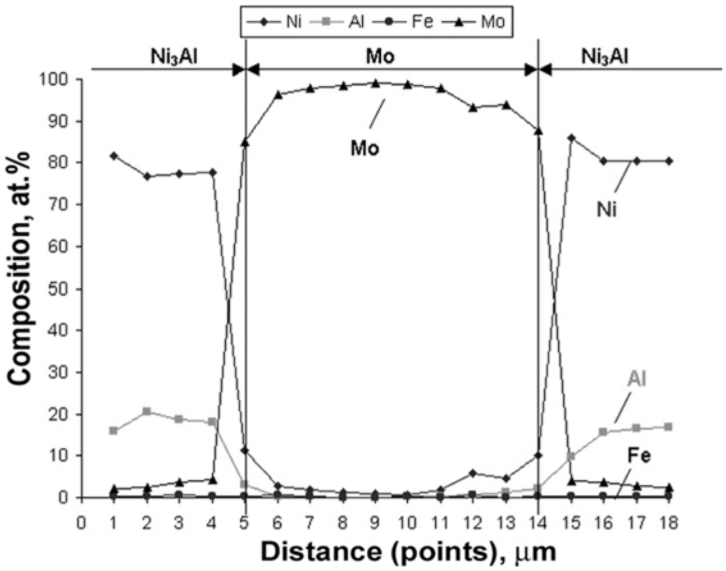

Fig. 2. Distribution of elements in MLCM 2.3 mm thick according to the data of X-ray electron-probe microanalysis (distance between the points ~ 5 μm).

It is seen that intermediate phases are absent, and the diffusion processes occurring at the interphase interface upon HIP and HR lead to the formation of the solid solutions of Ni and Al in Mo and Mo in Ni_3Al. The alloy based on Ni_3Al contains up to 0.75 at % Fe. The diffusion zone at the Mo and Ni_3Al sides does not exceed 10 μm.

4. PROPERTIES OF MLCM

4.1. Microhardness

The material upon HR and CR undergoes work hardening, which increases the hardness of the layers (table). This effect is more pronounced for Ni_3Al with the ordered fcc ($L1_2$) structure and less pronounced for bcc Mo. Annealing of the MLCM 0.5 mm thick at 1200°C for 2.5 h upon HIP leads to the softening of Ni_3Al by ~ 30%, whereas molybdenum is softened insignificantly.

Table

Microhardness of the MLCM

Processing, plate thickness, total degree of reduction	Microhardness, H_{μ}, kg/mm^2		
	Ni_3Al layer	Mo layer	Ni_3Al-Mo LCM
Initial material	350 ± 10	270 ± 20	-
HIP+HR, 2.3 mm, ε=80.8%	500 ± 20	300 ± 20	-
HIP+HR+CR, 0.5 mm, ε=95.8%	630 ± 40	350 ± 40	-
HIP+HR+CR+HIP 0.5 mm	430 ± 40	330 ± 40	360 ± 40
HIP+HR+CR+HIP+CR, 0.22 mm, ε=98.2%	-	-	420 ± 40

4.2. Bending test

The characteristic "stress-strain" diagram recorded upon RT bending is given in Fig. 3, and the bending angles for LCM of different thicknesses are given in the table.

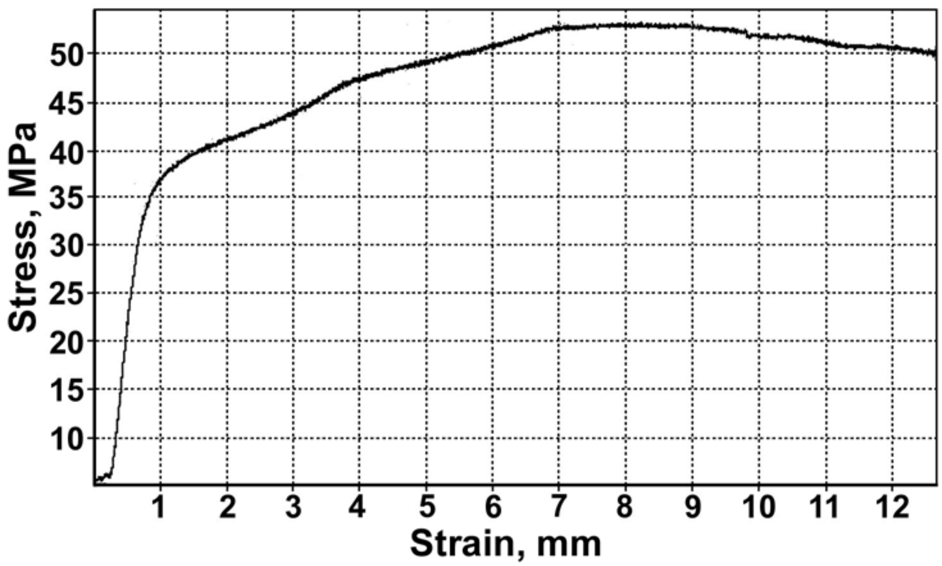

Fig. 3. Characteristic "stress-strain" diagram recorded upon three-point bending at RT.

The LCM plates 2.3 mm thick obtained by the regime “HIP + HR to $\varepsilon = 80.8\%$” can be bent without failure to an angle of 55 deg around the mount of 2.5 mm in diameter. The ultimate bending strength at RT is 1000 ±100 MPa. Annealing upon HIP of the cold-rolled plates 0.5 mm thick leads to the release of the stresses induced by CR, but decreases the ability to the concentrated plastic deformation upon bending, since it promotes the pinning of dislocations at least in one MLCM structure constituent such as molybdenum layers. The subsequent CR deformation to a thickness of 0.22 mm increases the number of mobile dislocations in the molybdenum LCM component, and the ability to the concentrated plastic deformation by bending is restored. The above data show that the obtained material has the reserve of plasticity at RT and can be used for the manufacture of the articles of complex shape after the proper selection of heat treatment conditions.

The ultimate bending strength (loss of the bearing capacity) was measured upon the three-point bending tests to be 256, 173, and 100 MPa at 1000, 1100, and 1200°C, respectively.

CONCLUSIONS

The billets consisting of alternating 25 Ni_3Al cards and 28 Mo cards were obtained by the HIP method. The LCM plates 2.3 and 0.5 mm thick were obtained by HR and subsequent CR without intermediate annealing, and the plate 0.22 mm thick was obtained by CR after annealing upon HIP. CR leads to the formation of a wavy structure of the Mo layers and a regular "thickening-reduction" structure of the Ni_3Al layers across rolling direction. This is caused by different resistances of the Ni_3Al layers and more refractory Mo layers to room-temperature deformation. The interlayer bonding is strong, and the interphase interface is free from intermediate phases. Solid solutions are formed in the near-interface regions in accordance with the nature of physicochemical interaction between Ni_3Al and Mo, which are in equilibrium in the pseudo-binary section of the Ni-Al-Mo ternary phase diagram. The work hardening of Ni_3Al upon rolling is more intense than that of Mo. The ultimate bending strength of the plate 2.3 mm thick is 1000±100, 256, 173, and 100 MPa at 20, 1000, 1100, and 1200°C, respectively. The obtained material has a reserve of RT plasticity and can be used for the manufacture of the articles of complex shape in the case of the proper selection of heat treatment conditions.

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REFERENCES

1. Povarova K.B. and Bannykh O.A. Physicochemical approaches to the development of high-temperature and oxidation-resistant composite materials with metallic or intermetallic matrix reinforced by refractory metals and interstitial phases. Part 1// Materialovedenie. 2001, No. 11, pp. 24-33; Part 2// Materialovedenie. 2001, No. 12, pp. 23-29; Part 3// Materialovedenie. 2002, No. 1, pp. 20-29.
2. Vasil'yeva A.G., Ustinov L.M., et al., Composite material NiAl-W// Vestnik MVTU. 1991, No. 1, pp. 91-96
3. Bannykh O.A., Povarova K.B., Kut'enkov V.A., Fridman A.G., Golovkina T. E., and Zavarzina E.K. High-temperature composite material with metallic matrix// Metally.1992, No. 3, pp. 145-149.
4. Noebe R D, Walson W..S. Prospects for development of structural NiAl alloys// Structural Intermetallics of 1997. Ed. Nathal M V, Darolia R, Liu C T, Martin P L, Miracle D..B., Wagner R, Yamaguchi M., The Minerals, Metals and Materials Society. 1997, pp. 573-584
5. Povarova K.B., Antonova A.V., Burmistrov V.I., and Skachkov O.A. Ternary phase diagrams of Al -Ti (Ni, Ru) - Me as the basis for the selection of layered aluminide-base composites containing ductile component// Metally. 2005, No. 3, pp. 75-82
6. Povarova K.B., Kazanskaya N.K., Drozdov A.A., and Morozov A.E, Physicochemical regularities of the interaction of nickel aluminides with alloying elements. I. Formation of solid solutions based on nickel aluminides // Metally. 2006, No. 5, pp. 58-71
7. Malienko E.I., Povarova K.B., Skachkov O.A., Edneral A.F., D'yakonov D.L., and Larin V.V., Optimization of the mechanical properties of rapidly solidified ribbon from the alloys based on Ni_3A1 intermetallic compound // Metally. 1999, No. 2, pp. 73-78
8. Povarova K.B. Rapily solidified ribbon from the alloys based on Ni_3A1 intermetallic compound as a material for layered composites// Zagotovitel'nye Proizvodstva v Mashinistroenii, 2005, No. 4, pp. 51-56