APPLICATION OF NANOSTRUCTURED INTERLAYERS IN JOINTS OF DIFFICULT-TO-WELD ALUMINIUM-BASE MATERIALS (Review)

D.A. ISHCHENKO

E.O. Paton Electric Welding Institute, NASU, Kiev, Ukraine

The paper deals with the technologies of application of nanostructured interlayers in the form of foils or coatings to improve making permanent joints of difficult-to-weld aluminium-base materials in the processes of diffusion and resistance welding, and well as welding with heating due to exothermal reaction of self-propagating high-temperature synthesis.

Keywords: diffusion welding, resistance welding, selfpropagating high-temperature synthesis, aluminium alloys, nanostructure, fillers, interlayers, coatings, foils, powders, plastic deformation, exothermal reaction

Aluminium and aluminium alloys come second after steel in terms of production and application. Owing to a unique combination of a complex of physico-mechanical, corrosion, and technological properties, aluminium-based alloys are successfully applied in different industries and in construction. Scope of aluminium alloy application in military equipment, in automotive, railway and water transportation, electrical engineering, in manufacture of cryogenic and chemical apparatuses, in agricultural and food machinery construction is considerable. In addition, high-strength aluminium alloys are the main structural material in flying vehicles, including aerospace engineering products (up to 80 % of volume by weight). Wider acceptance of such materials in manufacture of critical products is promoted by intensive current studies of weldability and development of effective measures on improvement of strength and reliability of welded joints, in particular, to prevent hot cracking and pores in welds.

In order to improve the processes of solid-state welding and the properties of permanent joints of difficult-to-weld materials, as well as alloys of different alloying systems, an effective technology was developed of application of nanostructured interlayers between the surfaces of items to be welded. Such interlayers are single- or multilayer coatings, foils or mixtures of ultradispersed powders. In diffusion welding with application of such materials high-strength welded joints with a dispersed microstructure are produced. Plastic deformation is localized in a thin interlayer that allows welding to be performed with application of modes with lower pressure, duration and temperature, i.e. the initial structure of welded materials is preserved.

Self-propagating high-temperature synthesis (SHS) of intermetallic compounds, applied for pressure welding, consists in gas-free combustion of metals

© D.A. ISHCHENKO, 2011

(charge components) in the gap between the materials being welded. This process is activated when ultradispersed film or powder interlayers, consisting of metals capable of entering into an instant exothermal reaction, are used as charge. SHS technology provides sound formation of joints owing to a high degree of heating localization in the welding zone.

Process of resistance welding with application of nanostructured interlayers is also improved, owing to shortening of welding time and increased localization of heating with prevention of material softening.

The purpose of this review is analysis of current developments of the methods of solid-phase permanent joining of materials based on aluminium-alloys with application of nanostructured interlayers in the form of foils or coatings, as well as with application of ultradispersed fillers, that provide improvement of the dispersed structure of the permanent joint zone with high strength properties.

Producing permanent joints with application of nanostructured interlayers in explosion welding. Pre-clad plates are often used as initial blanks in manufacture of laminated plates from high-strength aluminium alloys, and their initial thickness in the pack can vary significantly, depending on the final plate thickness and required ratio of the layers, thus affecting the quality of layer joining and being manifested in instability of properties at static and dynamic testing. In [1] a comparative assessment is made of the quality of the joint made by explosion welding, at cladding of high-strength alloys of Al-Zn-Mg system $(\Sigma Mg + Zn \ge 9 \%)$. It is shown that both strength and impact toughness of the joint increase with increase of the relative deformation rate ε at cladding. The most intensive growth of strength at increase of deformation rate up to 50 % and then a smoother increase up to values equal to the strength of AD1 interlayer are observed (Figure 1). Impact toughness reaches maximum values at higher deformation rates $(\varepsilon = 80 \%)$, being a characteristic more sensitive to defects of oxide film type. Heat treatment has a more significant influence on the joint quality, particularly, on the ductile properties (Figure 2). At increase of



quenching temperature and soaking time, the joint impact toughness decreases. Fracture mode becomes more brittle, particularly after recrystallization annealing at 550 °C temperature and $\varepsilon < 66$ %. This influence decreases at increase of deformation rate.

SHS process for permanent joining of materials and producing intermetallic coatings. During metal joining using SHS, heating of the joint area occurs at the expense of an exothermal reaction in the charge, placed before welding between the surfaces to be joined [2]. SHS-product participates in formation of weld material to a greater or smaller degree. Distinction is made between two variants of the process realization. In the first case (SHS-brazing) the materials to be joined and charge layer placed into the gap between them are cold in the initial condition or uniformly preheated. Short-time local heating initiates a combustion wave in this layer that heats the surfaces being joined and melts the SHS product. After that the welding area is compressed, bringing the material surfaces as close as possible to each other and partially removing the SHS-product from the gap. In the second case (SHS-welding) electrically-conducting charge is used and the process is initiated at the expense of passing current through it and the materials being welded, the exothermal reaction running simultaneously in the entire volume of the charge.

In SHS-process the initiation temperature in thin bilayer films depends on the heating rate and ratio of each layer thicknesses. Gas-free combustion is realized at initiation temperatures which are by 300–350 °C lower than on powders (for instance, for Al/Fe and Al/Co it is in the range of 250–400 °C, and for Al/Ni it is 200–300 °C) [3]. SHS mechanism is similar to the process of explosive crystallization. At the initial stage, the solid-phase reactions arising on the contact surface of film condensates can be gas-free combustion. Reaction in thin films can be implemented also on powder surface, if the second reagent is in the liquid phase. High cooling rates after passing of SHS wave in two-layer films lead to stabilization of high-temperature and metastable phases.



Figure 1. Dependence of the strength of layer adhesion, σ_{tear} (1) and impact toughness a_n (2) on the degree of deformation [1]

In the filed of permanent joining, the interest to multilayer foils based on elements forming intermetallics is also due to their application as fillers and as sources of local heating of metal at realization of SHS reaction (gas-free combustion) in the welding gap. In [4] evaluation of the intensity of heat evolution during SHS process in laminated Ni/Al foils placed between two copper foils being joined, was performed. It is shown that depending on chemical composition, thickness and initial microstructure characteristics of laminated foils, the intensity of heat evolution can vary in a broad range from 70 up to 400 W/cm². The velocity of propagation of gas-free combustion wave also depends on the thickness of component layers: at their reduction to the nanometric scale the velocity of propagation of SHS reaction wave through the foil can reach 10 m/s. The thermogram (Figure 3) shows that after initiation of SHS reaction the pack temperature quickly rises (in 0.1 s) up to the value dependent on the amount of the evolving heat, weight of sample and copper foil, which were selected so that heating of the entire pack did not exceed the copper melting temperature. This temperature, as a rule, was in the range of 500–700 °C.

Work [5] is devoted to investigation of the mechanisms of diffusion, formation and stability of new phase nuclei in the reactions corresponding to SHS in Ni–Al system. At comparison of the quantitative and qualitative change of phase composition for different content of aluminium particles (30 and 50 %) embed-



Figure 2. Dependence of joint impact toughness on quenching temperature ($\tau = 3$ h) (a) and soaking time (T = 470 °C) (b) [1]: $t-4 - \varepsilon = 50$, 60, 70 and 80 %, respectively



Figure 3. Thermograph of SHS reaction in a sample of Ni–Al system (61.2 wt.% Al) [4]

ded into the nickel matrix, prevalence of Ni_3Al phase for 30 % Al and 50 % NiAl is demonstrated. The rate of dissolution of embedded aluminium particles in the first case, when the system was heated pulse-like in an incremental series of successive time intervals, and the final material structure was memorized, is lower after the quenching process, than in the second case, when the system dynamic structure, which was obtained at pulsed heating was memorized, and it was again heated pulselike during a new time interval.

In [6–8] the micro- and nanostructures of multilayer films, obtained by the method of magnetron sputtering and consisting of alternating layers of titanium and aluminium in a broad range of layer thickness values, were studied. Microscopic features of propagation of gas-free combustion waves in them are considered. In the process of gas-free combustion in multilayer films of Al–Ti system the most probable mechanism of self-propagating reaction is aluminium diffusion into β -Ti at the temperature close to that of transition of α -Ti into β -Ti. SHS results in formation of intermetallic compounds of titanium and aluminium, which are highly textured poreless polycrystalline materials, which have two boundary systems normal to each other: between the layers and intergranular. Reagent layers are solid and rather even, and their mixing along the boundaries is slight (Figure 4). Columnar grains are oriented normal to the foil plane. They become almost indiscernible with decrease of layer thickness. However, the granular structure is preserved, and sometimes becomes more pronounced (Figure 5). As the coefficient of aluminium diffusion in titanium is quite small, intergranular boundaries can be the paths for anomalously fast diffusion, as they are oriented parallel to the diffusion flow. Compared to combustion of powder mixtures, «spreading» of SHS wave in the foil is more uniform On the other hand, the intergranular boundaries can slow down the heat flow directed from the hot products to the unburnt part of the sample, i.e. along the foil.

Work [9] describes the modes of phase transformations at heterogeneous reaction of gas-free combustion in multilayer nanofilms of Al–Ti system. It is shown that interaction of elements runs as a following sequence: disordering of titanium crystalline structure with simultaneous increase of interplanar spaces and saturation of solid titanium by aluminuim atoms; ordering of the crystalline lattice with formation of α -Ti based solid solution and parallel formation of TiAl₃ phase; ordering of mixed titanium and aluminium atoms into the crystalline structure of the final product of TiAl alloy. In the combustion mode all the process stages run almost instantaneously — in less than 0.04 s. At application of both the modes inheritance



Figure 4. Microstructure of multilayer Al/Ti films as a result of SHS (scanning electron microscopy) [8]



Figure 5. Microstructure of Al/Ti film fracture at 95 nm layer thickness [8]



of initial layer texture by the intermediate and final phases occurs.

In [10-12] the advantages of combining the SHSprocess and mechanical impact at joining ultradispersed materials from AlNi and AlTi intermetallics are considered. Duration and mode of mechanical activation at dispersion in powder mixtures of nickel with aluminium and titanium with aluminuim, influence the process characteristics and composition of gas-free combustion products. Combustion for these compositions proceeds in a microheterogeneous mode, and heating rate is determined by the time of heating of the composite particles. In the case of thermal explosion mode, where combustion cannot be stopped immediately after reaching the inflammability limit, it can be slowed down at the expense of different duration of combustion after ignition, by changing the heating rate and using coarse nickel powder as in situ heat source. This indirect method successfully simulates the operation of wave hardening that is applicable only to the mode of plane wave propagation.

Resistance welding. Work [13] gives the results of investigation of the features of joint formation by resistance welding technology using Al/Ni and Al/Cu nanostructured foils as inserts between the parts of AD0, 1460 and AMg6 aluminium alloys to be joined. Such a technology is characterized by highly-concentrated heat evolution in the butt that reduces the welding time and prevents metal softening. When foil consisting of layers of aluminium and nickel is used, additional heat evolves in the contact zone that is due to running of exothermal reaction between the metals which is accompanied by formation of intermetallic phases. Development of an exothermal reaction depends on the heating rate in welding. With increase of the latter, the amount of flash increases under the impact of the compression force. Heating rate of 500-800 °C/s is optimum for producing sound joints. Foil fragments preserve their laminated structure, i.e. aluminium and nickel reaction runs locally (Figure 6). Application of aluminium-copper nanostructured foils allows a marked lowering of welding temperature (by 100–150 °C) owing to running of the process of formation of Al₂Cu eutectic in the foil. This is particularly important in welding thermally unstable aluminium alloys.

Proceeding from investigations of the mechanism of joint zone formation, authors of [14] developed methods to produce by resistance spot welding sound joints from steel and AMg6 and AMts aluminium alloys, using bimetal steel—aluminium plates, which were made by rolling or explosion welding. Here it is shown that in manufacture of the inserts, preference should be given to explosion welding, as it allows producing a sound joint with self-cleaning of the surface during slanting collision of sheet blanks. Using a bimetal transition piece, resistance spot welding of AMg6 alloy to St3 steel was performed. Regularities



Figure 6. Fragments of Al/Ni foil in the weld metal [13]

of the processes of melting, solidification, interdiffusion and chemical interaction of the components, as well as their change with increase of temperature and pressure allow determination of the optimum welding modes and producing a strong joint of the bimetal and base material.

Methods to produce nanostructured foils and coatings. Alongside the welding process parameters and postweld treatment, quality of joints made using nanostructured foils and coatings is also influenced by chemical and phase composition, size of ultradispersed particles and thickness of nanostructured foils or coatings. Therefore, features of the processes of their manufacture are also important. Nanostructured foils and coatings are made by the following methods: hardening by melt spinning [15–20]; detonation deposition [21, 22]; condensation [23]; deposition after thermal [24], magnetron [25], vacuum-arc [26] spraying; ion implantation [27, 28]; and electroplating.

CONCLUSION

The published results given above are indicative of a high efficiency of application of nanostructured interlayers for joining difficult-to-weld aluminium alloys. Such interlayers can be elementary or multilayer coatings, foils or mixtures of ultradispersed powders. The produced welded joints are high-strength, with a dispersed microstructure.

Under the conditions of diffusion welding with application of foils or coatings with an ultradispersed structure, plastic deformation is localized in a thin interlayer. This allows applying smaller compression force and accelerating the process of welding without heating, thus promoting preservation of the initial structure of welded materials.



SCIENTIFIC AND TECHNICAL

SHS is characterized by high-temperature phase formations in the contact zone at anomalously fast reaction and diffusion. Here sound weld formation is ensured at comparatively soft temperature modes, owing to a high decree of heating localization in the welding zone.

Application of nanostructured multilayer foils in resistance welding causes an additional highly concentrated heat evolution in the joint zone. This is promoted by a local exothermal reaction between the interlayer metals, initiated by electric current, that allows preserving the structure and strength properties of the base material.

- Shlensky, A.G. (2008) Examination of joint quality in cladding of Al-Zn-Mg system alloys. *Tekhnologiya Mashinostroeniya*, 8, 20-23.
 Klubovich, V.V., Kulak, M.M., Samoletov, V.G. (2005) Producing permanent joints of materials using SHS. In: *Welding and related technologies*: Transact. Minsk.
- Myagkov, V.G., Zhigalov, V.S., Bykova, L.E. et al. (1998) 3 Self-propagating high-temperature synthesis and solid-phase reactions in two-layer thin films. *Zhurnal Tekhnich. Fiziki*, 68 (**10**), 58-62.
- Shishkin, A.E., Rogovchenko, D.S., Ustinov, A.I. (2009) Evaluation of heat evolution intensity in quick-propagating high-temperature synthesis in Ni/Al multilayer foils. Metal-lofizika i Nov. Tekhnologii, 31(9), 1179–1188.
- 5. Denisova, N.F., Starostenkov, M.D., Kholodova, N.B. (2005) Study of formation and stability of new phase nuclei in reactions corresponding to SHS in Ni-Al system. In: Proc. of 9th Int. Sci.-Techn. Conf. on Composites in National Economy (Barnaul, Nov. 2005). Barnaul: AltGTU, 100-105.
- Rogachev, A.S., Grigoryan, A.E., Illarionova, E.V. et al. (2004) Gas-free burning of bimetallic Ti/Al nanofilms. *Fizi* ka Goreniya i Vzryva, 40(2), 45–51.
- Grigoryan, A.E., Elistratov, N.G., Kovalev, D.Yu. et al. (2001) Autowave propagation of exothermic reactions in thin multilayer films of Ti-Al system. *Doklady RAN*, 381(**3**), 368–372.
- Grigoryan, A.E., Illarionova, E.V., Loginov, B.A. et al. (2006) Structural peculiarities of thin multilayer Ti/Al films for self-propagating high-temperature synthesis. Izvestiya Vuzov. Tsvet. Metallurgiya, 5, 31-36. Rogachev, A.S., Gashon, Zh.K., Grigoryan, A.E. et al.
- (2006) Formation of crystalline structure of products during heterogeneous reaction in multilayer bimetallic nanosystems. Izvestiya RAN. Physics Series, 70(4), 609–611.
- 10. Grigorieva, T.F., Korchagin, M.A., Barinova, A.P. et al. (2000) Self-propagating high-temperature synthesis and mechanical fusion in producing monophase highly-refined intermetallics. Materialovedenie, 5, 49-53.
- 11. Biswas, A., Roy, S.K. (2004) Comparison between the microstructural evolution of two modes of SHS of NiAl: key to a common reaction mechanism. *Acta Mater.*, 52(2), 257–270.
- Shkodich, N.F., Kochetov, N.A., Sachkova, N.V. (2006) On the influence of mechanical activation on SHS compositions of Ni-Al and Ti-Al. *Izvestiya Vuzov. Tsvet. Metallurgiya*, **5**, 44–50. 12.

- 13. Kuchuk-Yatsenko, V.S., Shvets, V.I., Sakhatsky, A.G. et al. (2007) Specifics of resistance welding of aluminium alloys with nanostructural aluminium-nickel and aluminium-copper foils. Svarochn. Proizvodstvo, **9**, 12–14.
- 14. Kovalevsky, V.N., Demchenko, E.B., Lopatko, I.G. (2006) Application of nanocoatings in pressure welding of dissimilar materials. Svarka i Rodstv. Tekhnologii, Issue 8, 84–87.
- Shpak, A.P., Majboroda, V.P., Kunitsky, Yu.A. et al. (2004) Nanolayered fragments in aluminium alloys. *Nanosistemy*, *Nanomaterialy*, *Nanotekhnologii*, 2(2), 681–687. 15.
- 16. Neumerzhitskaya, E.Yu., Shepelevich, V.G. (2005) Structure, properties and thermal stability of rapidly-solidified foils of aluminium alloy with chromium, nickel and manganese. Perspect. Materialy, 4, 69–73.
- 17. Karpets, M.V., Firstov, S.O., Kulak, L.D. (2006) Specifics of phase formation in quickly-hardened Al-Fe-Cr alloys in the presence of quasicrystals. Fizyka i Khimiya Tv. Tila, 7(1), 147–151.
- 18. Gutko, E.S., Shepelevich, V.G. (2005) Investigation of rapidly-solidified foils of binary and ternary aluminium-base alloys containing zinc and magnesium. *Fizika i Khimiya* Obrab. Materialov, 4, 81-85.
- 19. Tashlykova-Bushkevich, I.I., Shepelevich, V.G. (2000) Elemental layer-by-layer analysis of component distribution in the volume of rapidly-solidified aluminium low alloys. Ibid., 4, 99-105.
- Tashlykova-Bushkevich, I.I., Gutko, E.S., Shepelevich, V.G. et al. (2008) Structural and phase analysis of rapidlysolidified Al-Fe alloys. Poverkhnost. Rentg., Sinkhr. i Nejtronnye Issledovaniya, 4, 69–75.
- 21. Oliker, V.E., Sirovatka, V.L., Timofeeva, I.I. et al. (2005) Influence of properties of titanium aluminides and detonation spraying conditions on phase and structure formation of coatings. Poroshk. Metallurgiya, **9**/**10**, 74-84. 22. Romankov, S.E., Kaloshkin, S.D., Pustov, L.Yu. (2006)
- Synthesis of titanium-aluminide coatings by the method of mechanical fusion and subsequent annealing on the surface of titanium and aluminium. *Fizika Metallov i Metallovede*nie, 101(1), 65-73.
- 23. Perekrestov, V.I., Kosminskaya, Yu.A., Kravchenko, S.N. (2003) Principles of structure formation of condensates of low-supersaturated Cu, Ti, Al and Cr vapors. Metallofizika i Nov. Tekhnologii, 25(6), 725-735.
- Buzhenets, E.I., Majboroda, V.P., Shpak, A.P. et al. (2004) Structural peculiarities of quasi-crystalline coatings of 24. Al₆₃Cu₂₅Fe₁₂ alloy. *Nanosistemy, Nanomaterialy, Nanotekh-*nologii, 2(**4**), 1323–1329.
- Hampshire, J., Kelly, P.J., Teer, D.G. (2003) Tribological properties of co-deposited aluminium-titanium alloy coa-tings. In: Proc. of 30th Int. Conf. on Metallurgical Coa-tings and Thin Films (Apr. 28-May 2, 2003, San Diego, Calif.), 392–398.
- Budilov, V., Kireev, R., Kamalov, Z. (2004) Intermetallic products formed by joint cold cathode vacuum arc sputte-ring of titanium and aluminium. In: *Proc. of 11th Int. Conf.* on Rapidly Quenched and Metastable Materials (Oxford, 25-30 Aug. 2002), 656-660.
- Kurzina, I.A., Bozhko, I.A., Kalashnikov, M.P. et al. (2005) Formation of surface layers containing intermetallic compounds of Ni-Al and Ti-Al systems at high-intensity ion implantation. Perspekt. Materialy, 1, 13-23.
- Kurzina, I.A., Bozhko, I.A., Kalashnikov, M.P. et al. (2004) 28.High-intensity implantation of aluminium ions into titanium. Metallofizika i Nov. Tekhnologii, 26(12), 1645-1660.

aton, IRNAL