PECULIARITIES OF INTERGRANULAR MASS TRANSFER OF GALLIUM IN ALUMINIUM ALLOY DURING SOLID PHASE ACTIVATION OF SURFACES BEING JOINED

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Peculiarities of formation of a dissimilar aluminium-steel diffusion joint in solid-state activation of the surfaces, being joined, by gallium are considered. The effect of products of gallium-aluminium interaction on a local change in mechanical properties within the zone of a permanent joint is shown.

Keywords: *diffusion welding, aluminium, aluminium alloys, reactive diffusion joint, intergranular diffusion, gallium, low-temperature activation of diffusion, indention, Berkovich indenter*

Sometimes, when producing the permanent steel-aluminium units of radio engineering purpose, it is required to reduce the temperature of technological processes down to 140 °C. One of the variants for joining steel with aluminium in a solid phase is brazing with application of pressure [1]. To prevent the formation of brittle intermetallic compounds at the interface of materials being joined the appropriate intermediate inserts of the third metal are used which are a certain diffusion barrier between materials being welded [2]. The activation of adhesive processes of the surfaces being joined is performed by deposition of layer of molten gallium with a tight pressing and consequent heating of the whole unit. Such technology of joining should provide formation of a strong joint without melting of parts and chemical refining of the surfaces being joined from oxide film.

High physical-chemical activity of gallium turns this technological task into a sufficiently complicated process which depends on the number of both positive and negative factors. Gallium is good to wet most metals, therefore they are easily subjected to «adhesion». The temperature interval of liquid state of gallium varies between 30–2204 °C [3]. Metallic «adhesives», based on gallium, impart thermal and electric conductivity, and also high rigidity to the weld. As a result of joining parts into assemblies without fusion of base metal the level of its buckling and oxidation sharply decreases. The strength at equal separation of joints produced using gallium «adhesives» is 26–30 MPa at the temperature of 20 °C. With increase of temperature the diffusion of gallium is sharply increased, and with its decrease the «adhesive» is solidified and facilitates joining of parts during seizure, even if they are not wet by the adhesive [4].

In the equilibrium diagram (Figure 1) [5] it is seen that gallium and aluminium form simple eutectic system in the point where gallium content is equal to 99.2 wt.% at the temperature of 29.7741 °C. The solubility of gallium in solid aluminium at eutectic temperature is approximately 15–20 %, the solubility of aluminium in gallium is negligible [6]. The exclusive feature of gallium is capability of its melts to overcooling: when reducing its temperature by 10–30° below the point of melting it remains in liquid state. The reason of such abnormality consists in peculiarities of molecules constitution and bonds between them under different aggregate conditions.

The joining of materials using molten gallium is a three-stage process: formation of a physical contact (wetting of surfaces being joined with molten gallium); activation of contact surfaces (removal of oxide film); volumetric development of interaction between materials (pressing of their surfaces and start of diffusion).

The mechanism of diffusion of gallium in aluminium is mainly connected with relation of atomic radii of a diffusant and components of solvent: approximation of size of atom of gallium to the size of atom of aluminium promotes the diffusion of gallium in vacancies of aluminium [7]. The rate of migration of atoms of gallium along the boundaries of grains of aluminium at the temperature of 27 °C is almost the same as the diffusion of gallium in liquid aluminium ($D = 1\cdot10^{-5}$ cm^2/s). The energy of interface is low and it is about 0.06–0.30 N/m. Therefore, in the

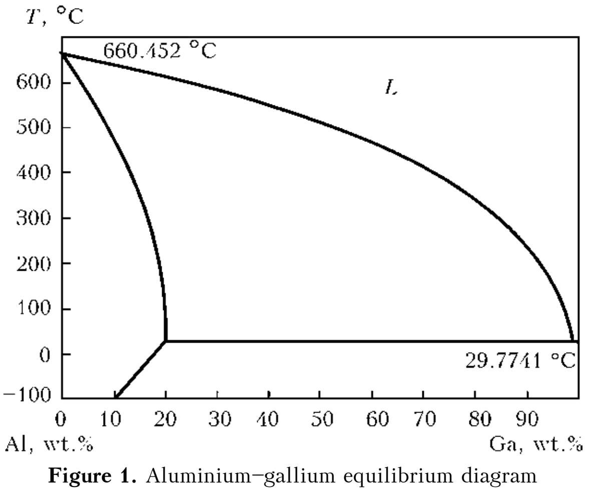

Figure 1. Aluminium–gallium equilibrium diagram

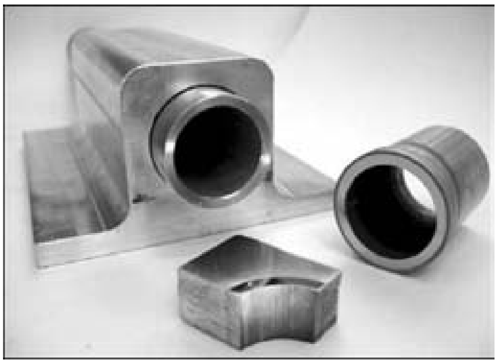

Figure 2. Appearance of the diffusion-welded parts

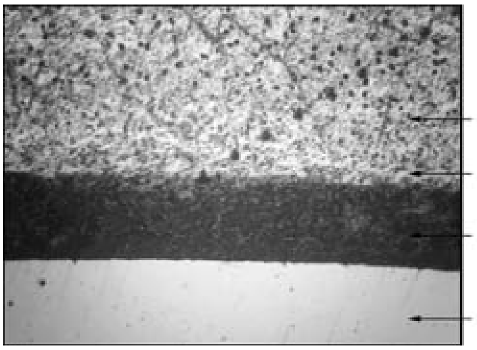

Figure 3. Microstructure (×300) of the zone of joining with dents of Berkovich indenter

presence of diffusant in liquid state at high temperature the conditions are created for grain-boundary phase transition of wetting.

At the initial stage, when the level of diffusion is negligible, the formation of homogeneous solid solution takes place due to diffusion along the grain boundaries. Then, the intermetallic compound is formed having somewhat decreased mechanical properties [8]. The tendency towards considerable decrease in strength is caused by the presence of Rehbinder effect [9, 10].

The unit of a product (Figure 2) represents by a design the joining of stainless steel 10Kh18N9T pipes of 30 mm diameter with a flange of aluminium alloy 6063 of Al–Mg–Si alloying system. To provide dissimilar joining of aluminium-steel on the outer surface of a pipe by a gas-dynamic method the layer of commercial aluminium AD1 (99.3 wt.% Al) of 300 μm thickness was deposited. The activation of adhesion processes of the surfaces being joined was performed by deposition of layer of molten gallium of 10–20 μm thickness with a tight pressing and further heating of the whole unit up to 140 °C during 2 h.

The investigation of depth of intergranular mass transfer of gallium in the process of its diffusion to aluminium was carried out each 1 h and after 60 days. The specimens were stored at the temperature of 30 °C.

Fractographic investigations of microstructure, determination of quantitative element composition and mapping of distribution of elements were performed using methods of scanning electron microscopy (SEM) and X-ray diffraction microanalysis on the basis of an analytic complex consisting of scanning electron microscope JSM-35CF (JEOL, Japan) and X-ray spectrometer with dispersion in energy of X-ray quanta of the model INCA Energy-350 (Oxford Instruments, Great Britain).

For micromechanical tests the nanoindenter «Micron-gamma» was applied designed for determination of mechanical properties using methods of continuous indentions of three-facet diamond Berkovich indenter, scanning by indenter, as well as metallographic and topographic methods [11].

The investigation of structure of specimens showed that as a result of diffusion in the sprayed layer and on the side of aluminium alloy the vast regions with changed (coarsed) structure were formed (Figure 3). The diffusion of gallium into the steel was negligible. The map of distribution of elements (Figure 4) evidences the mass transfer of gallium towards the side of aluminium alloy for the depth of up to 3 mm. The diffusion of gallium to aluminium alloy passes along the intergranular network with formation of distinctly marked boundary chains. During heating the liquid gallium wetted completely the grain boundaries, along which it was segregated, that gave it possibility to move quickly along the intergranular space to vast distances into the volume of material. The qualitative characteristic of degree of proceeding of grain boundary diffusion at each definite boundary is the width of near-boundary bands, where content of gallium reaches 85 wt.% (Figure 5, *a*). Gallium formed «islands» in the most thermodynamically beneficial places, such as clusters of eutectic mass of helium of high concentration (up to 97 wt.%) in corners of grains (spectrum 2, 4, Figure 5, *b*).

In Figure 6 the distinctions in dents of the indenter at constant minimal loading in indenting of aluminium alloy at the initial stage of diffusion are shown. It

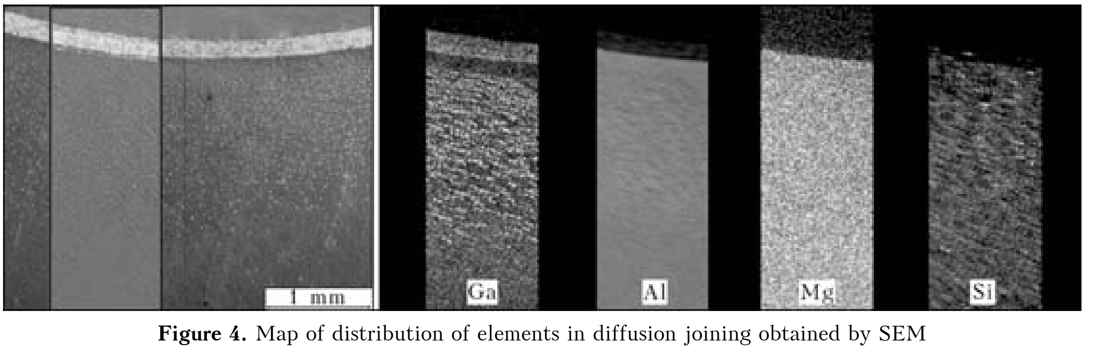

Figure 4. Map of distribution of elements in diffusion joining obtained by SEM

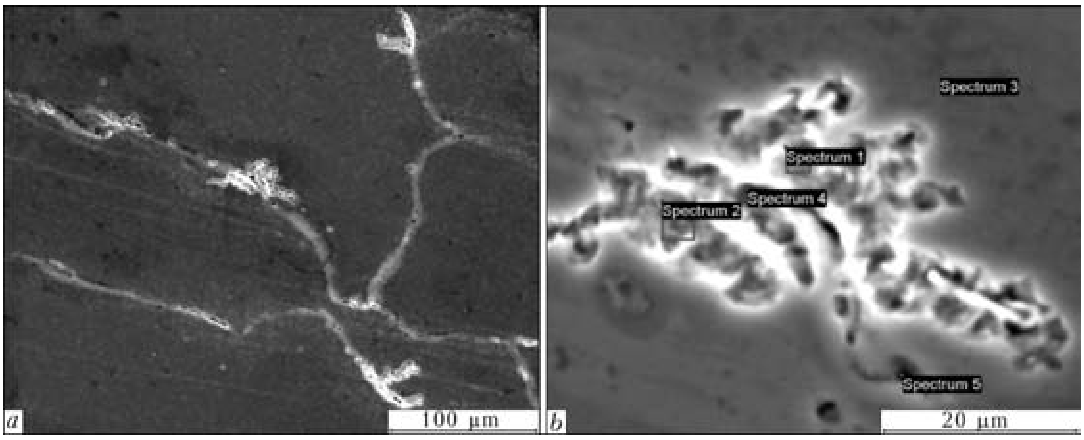

Figure 5. Pattern of intergranular clusters of eutectic mass of gallium in the aluminium alloy obtained by SEM (*a*, *b* see in the text)

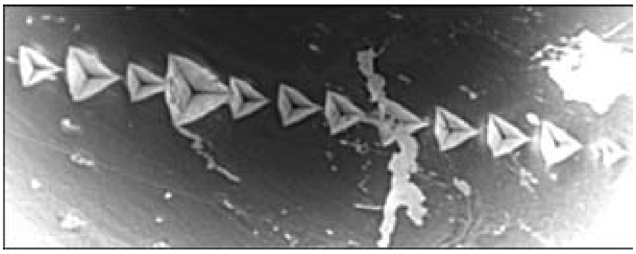

Figure 6. Dents (×600) of Berkovich indenter in the structure zones of aluminium alloy

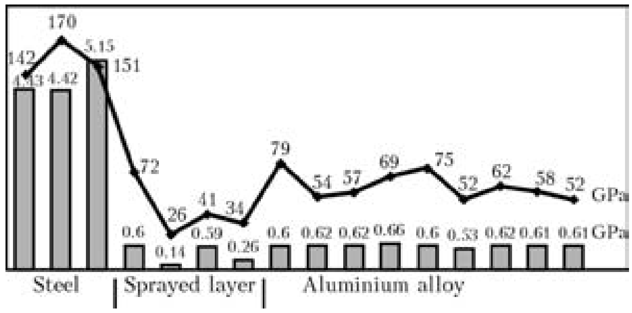

Figure 7. Diagrams of distribution of microhardness according to Meyer (*columns*) and elasticity modulus of Young (*curve*)

was produced at ultra-low (0.010–0.001 N) loading of indenter due to softening of material and impossibility of indenting at standard loading for aluminium (0.2 N) [12]. The large dents with separation of material are characteristic of intergranular boundary region filled with eutectic mass of gallium.

The jumpy change of microhardness and modulus of elasticity (Figure 7) characterizes the presence of meta-stable phases of gallium with magnesium and zinc [13–17]. The comparison of basic mechanical properties of materials being joined before the effect of gallium and properties of materials in the joint after diffusion shows the presence of general softening at the level of 10 %. At repeated heating of specimens the change in mechanical properties was not observed that proves the formation of solid solution Al_5Ga_2.

It follows from the above-described that the physical-chemical processes of interaction between the base metal (aluminium), components of aluminium alloy (magnesium and zinc) and gallium occur in the zone of formation of permanent joint. Gallium as activator of the surfaces being joined provided the removal of oxide layer at the surface (deoxidation). Its diffusion passed not only intergranually with an intensive growth of sizes of grains, but also volumetrically with a shifting of old boundaries of grains of aluminium and formation of new ones.

The distinctive feature of mass transfer of gallium in the aluminium is the formation of multistage transformation in a meta-stable phase with gallium. It resulted in formation of solid solution Al_5Ga_2 in the gap and adjacent volume of parts being joined.

1. Gab, I.I. (2008) Solid-phase brazing with pressure application. In: *Inorganic materials science*: Encyclopedia. Vol. 1, Book 2. Ed. by G.G. Gnesin, V.V. Skorokhod. Kiev: Naukova Dumka.
2. Ryabov, V.R., Ishchenko, A.Ya., Muravejnik, A.N. (1996) Current methods of welding of steel-aluminium pipes. *Avtomatich. Svarka*, **2**, 32–42.
3. *http*://en.wikipedia.org/wiki/Ga
4. Tikhomirova, O.I., Pikunov, M.V. (1969) Influence of shape and size of second component particles on properties of gallium solders. *Poroshk. Metallurgiya*, 84(**12**), 51–56.
5. *http*://www.crct.polymtl.ca/FACT/phase_diagram.php?file=Al-Ga. jpg&dir=SGTE
6. Mondolfo, L.F. (1979) *Structure and properties of aluminium alloys*. Moscow: Metallurgiya.
7. Kholyavko, V.V. (2006) *Formation of phase composition, structure and properties of quasi-crystalline alloys of Al–Cu–Fe system in reaction diffusion of gallium*: Syn. of Thesis for Cand. of Techn. Sci. Degree. Kiev.
8. Livanov, V.A. (1982) Action of gallium on aluminium and its alloys. *Tekhnologiya Lyog. Splavov*, **5**, 30–32.
9. *http*://ru.wikipedia.org/wiki/Effect_Rebindera
10. Likhtman, V.I., Shchukin, E.D., Rebinder, P.A. (1962) Physico-chemical mechanics of metals. In: *Adsorption phenomena in processes of deformation and fracture of metals*. Moscow: AN SSSR.
11. Ishchenko, A.Ya., Khokhlova, Yu.A. (2009) Evaluation of mechanical properties of microstructural constituents of welded joints. *The Paton Welding J.*, **1**, 34–37.
12. Khokhlova, Yu.A., Khokhlov, M.A. (2009) Nanoscale effect in diffusion joints with gallium. In: *Abstr. of Int. Conf. on Problems of Welding, Related Processes and Technologies* (Nikolaev, 14–17 Oct. 2009). Nikolaev: NUK, 111.
13. Larikov, L.N., Franchuk, V.I., Maksimenko, E.A. (1991) Substructural strengthening in aluminium and its alloys interacting with gallium. *Metallofizika*, 13(**10**), 3–10.
14. Larikov, L.N., Maksimenko, E.A., Franchuk, V.I. (1990) Structural changes in aluminium and its alloys during embrittlement by liquid gallium. *Ibid*., 12(**1**), 115.
15. Franchuk, V.I., Larikov, L.N. (1992) Change of orientation of crystallites in near-surface layers of polycrystalline aluminium in diffusion interaction with gallium. *Izvestiya RAN. Metally*, **6**, 105–110.
16. Larikov, L.N., Prokopenko, G.I., Franchuk, V.I. et al. (1990) Study of embrittlement of aluminium and AMg6 alloy in interaction with liquid gallium by acoustic emission method. *Fiziko-Khimich. Mekhanika Materialov*, **3**, 5–9.
17. Larikov, L.N., Franchuk, V.I., Tikhonovich, V.V. et al. (1991) Diffusion-induced migration of grain boundaries in Al–Ga system. *Metallofizika*, 13(**8**), 56–62.