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FLUXLESS BRAZING OF ALUMINIUM ALLOYS BY BRAZING FILLER METAL OF Al–Ge SYSTEM

O.M. Sabadash, S.V. Maksymova

E.O. Paton Electric Welding Institute of the NASU
11 Kazymyr Malevych Str., 03150, Kyiv, Ukraine

ABSTRACT

The paper gives the results of investigations on fluxless brazing of AD1M, AMts (Al–Mn), AD31 (Al–Mg–Si) aluminium alloys (Al–Mg–Si) with application of Al–25Ge–5Si–5Cu–1.5Mn–0.15Ti brazing filler metal at the temperature of 550±5 °C in the atmosphere of high-purity nitrogen. The results of high-temperature differential thermal analysis were used to determine the solidus and liquidus temperatures of the brazing filler metal. Thermal effects on the derived thermal curve are indicative of the presence of three phases, which correlates with the results of X-ray microprobe analysis. It is shown that the brazing filler metal structure in the initial state is formed by two solid solutions, based on α-Al and β-GeSi and eutectics. Mechanical testing revealed that the short-term strength of the brazed joints is higher than that of the base metal, and fracture occurs in AD1M alloy. Shear strength of brazed joint of AMts alloy is τ_t = 82 MPa. Application of steplike cooling of AD31 alloy brazed joint with soaking at the temperature of 500 °C promotes increase of shear strength from 84 to 102 MPa.

KEYWORDS: fluxless brazing, aluminium alloy, germanium brazing filler metal, nitrogen, brazed joint, shear strength

INTRODUCTION

Brazing with aluminium brazing filler metals is used for simultaneous joining of parts and assemblies of multilayer thin-walled (≤ 1mm thickness) structure from aluminium alloys of 1000 (Al), 3000 (Al–Mn), 6000 (Al–Mg–Si) series with the aim to provide corresponding technical characteristics (strength, precision size, temperature and heat conductivity) in operation.

The main problem of manufacture of brazed structures is violation of integrity of aluminium thin-walled parts due to interaction with brazing filler metal under conditions of high-temperature heating. Existing technological processes of brazing of separate assemblies, in particular, slot antennas from aluminium alloy AMTs (A3003) with brazing filler metal of Al–Si system by means of deepening in a melt from chloride compounds [1, 2] and wave guides in dry furnace air using brazing filler metal Al–Si in form of powder mixture with flux of KCl–LiCl [3, 4] system are ecologically dangerous because of significant evaporations of toxic compounds and require considerable expenses for utilization of flux and its corrosion-active residues.

Development of brazing sheets of base metal with thin surface layer of brazing filler metals of Al–Si [5] and Al–Si–Mg [5–7] systems promoted development of furnace brazing of heat exchangers from aluminium alloys of 1000 and 3000 series in controlled medium (vacuum, purified nitrogen). Formation of joint of Al-alloy at brazing temperature 600–620 °C in vacuum differs by Si diffusion in multilayer material and balance of rates of Mg evaporation in open and closed zones of radiator [8–10]. A disadvantage of this method is impossibility of application of corrosion-resistant coatings that contain anode protector-zinc directed on electrochemical protection of base metal by decrease of potential [10, 11].

There is information on application of aluminium brazing sheets with modified layer “brazing filler metal Al–Si–coating” [12] at furnace brazing of structures of aluminium alloys of 1000 and 3000 series in temperature interval 600–620 °C in purified nitrogen (p_{O_2} < 700 Pa, T_{dew} = –50 °C). In “brazing filler metal–Al–Si–coating-base metal” system the successful formation of brazed joint depends on alloying. Additives of lithium [13] and bismuth [14] reduce surface tension of brazing filler metal at the interface with base metal; multilayer electrochemical coatings from nickel [15], zinc, tin [16] in combination with non-corrosive flux KF–AlF_3, which actively cleans the surface, provide high capillary properties of brazing filler metal [17, 18] and improve wetting of base metal with brazing filler metal. Microadditives of Bi and Mg in Al–10Si brazing filler metal promote quality filling of a gap in atmosphere of nitrogen of ultra-high purity (p_{O_2} = 10^{-25} Pa) [19].

Aluminium brazing filler metals of Al–(6.8–13) Si system melt at high temperature 577–613 °C (Figure 1, *a*) [20, 21] close to solidus temperature of alloys of 5000 (systems Al–Mg, alloy 5052 has T_s = 568 °C) and 6000 (Al–Mg–Si, alloy A6061, T_s = 600 °C) series [22] which has negative effect on base metal integrity and microstructure of α-solid solution. For reduction of negative effect of temperature on base metal it is necessary to use elements decreasing melt-

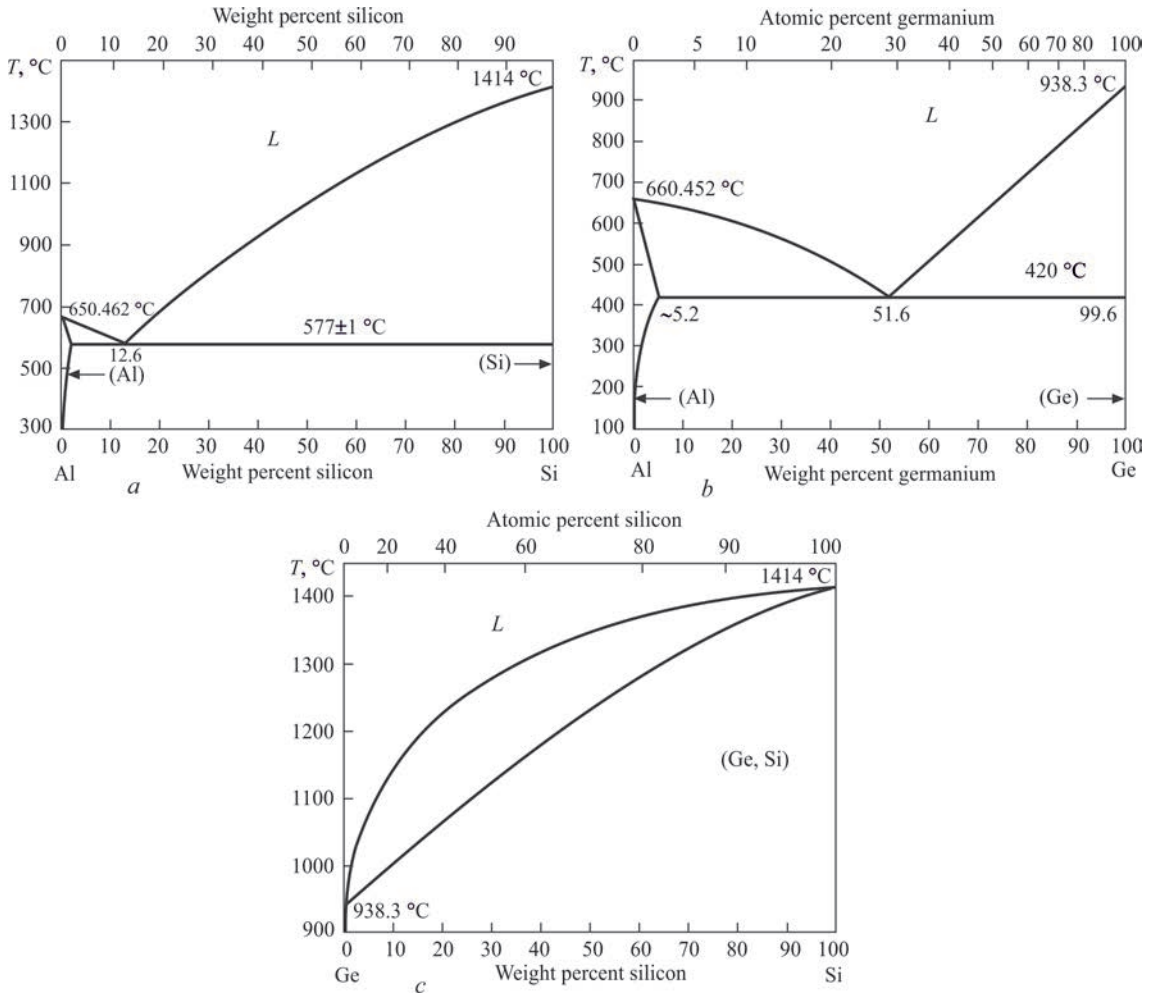

Figure 1. Binary diagrams of state of metallic systems: Al–Si (*a*); Al–Ge (*b*); Ge–Si (*c*) [20]

ing temperature of aluminium alloy to 450–550 °C (Figure 1, *b*).

Brazing filler metals with reduced temperature of melting are based on triple alloys of Al–Si–Me system with high content of metal-depressants (Me = Cu, Ge, Zn). In area reach with aluminium, Al–Cu–Si system contains combination of phases Al_2Cu and (Si) which are in equilibrium with solid solution based on α-aluminium [23–25]. Triple eutectic Al–5Si–27Cu melts at 525 °C [23].

Volume and morphology of crystals of brittle phase Al_2Cu as a constituent of weld microstructure which is present in intergranular layer of α-solid solution significantly effects a level of shear strength (40–60 MPa) of joint of alloy A6061 [26] brazed with Al–9.6Si–20Cu brazing filler metal (in vacuum at 570 °C temperature). Addition of nickel to brazing filler metal Al–9.6Si–20Cu narrows the temperature interval of melting of 73Al–20Cu–2Ni–5Si brazing filler metal, promotes enrichment of Al-solid solution with particles of phases Al_2Cu, $Al_6Cu_3Ni_6$ [27, 28], δ-$Al_3CuNi(Al_3Ni_2)$ [29] and increase of shear strength.

Fluxless brazing in purified nitrogen using Al-sheet with double-layer coating Al–Si and Cu–Ni, which at contact melting generates brazing filler metal 73Al–20Cu–2Ni–5Si, with simultaneous compression of parts at brazing temperature 540°C rises (~ by 50 %) level of strength of brazed joints of alloys A3003, A6013, A7475 [27, 28]. However, application of compression is not always acceptable that is promoted by geometry and technological peculiarities of brazed assemblies. Flux brazing of alloy A6063 using 73Al–20Cu–2Ni–5Si brazing filler metal and chloride flux with next heat treatment (530 °C/3 h, 160 °C/3 h) promotes increase of strength (by 28 %) of joint in comparison with that using Al–6.5Si–20Cu brazing filler metal [29]. It should be noted that brazing filler metals of Al–Si–Cu system with high content of copper exceed allowable difference of potential of corrosion in relation to potential of aluminium alloys that intensifies corrosion failure of brazed joint in aggressive medium [27, 28, 30] and requires additional protective means.

Alloys of Al–Ge–Si system, studied experimentally [31] and by calculation methods [32, 33] are promising (from point of view of melting temperature) for brazing of aluminium materials. In the alloys of triple system Al–Ge–Si the areas of binary eutectic reac-

Table 1. Composition (wt.%) and melting temperature (T, °C) of aluminium alloys

Alloy	Si	Fe	Cu	Mn	Mg	Cr	Zn	Ti	T_s	T_l
AD1M	0.3	0.3	0.3	0.03	0.05	0.0	0.1	0.15	643	657
AMts	0.6	0.7	0.20	1.0–1.5	–	0.0	0.1	–	643	654
AD31	0.2–0.6	0.5	0.1	0.1	0.45–0.9	0.1	0.2	0.15	616	654

tions L (liquid)↔(Al)+(Si) and L↔(Al)+(Ge) join with the area L↔(Al)+(SiGe) that results in decrease of melting temperature from 578 °C (~12.7 wt.% of Si) to 424 °C (~ 53 wt.% of Ge) [31].

Brazing filler metals of Al–Ge–Si system [34, 35] with high content of Ge (~ 25 %) are too expensive for most of the applications. A joint of alloy A6061 brazed with Al–12Si–(25, 35, 45)Ge–(≤ 1)Mg–(≤ 1)Cu brazing filler metals in vacuum at increase of temperature from 550 to 575 °C and long-term holding (~ 60 min) reaches strength making ~ 90 % of base metal strength [35]. As for alloy A5052 (Al–Si–2.2–2.8 % Mg), strength limit of joint dramatically decreases (~ 40 %) due to growth of available layer of brittle compound Mg_2Ge in a reaction zone at interaction with Mg (from base metal) and Ge from the brazing filler material. Partial replacement of Ge by Zn in the brazing filler metal Al–9.5Si–10Ge–15Zn–0.75Sr and addition of Sr promoting refinement of β-(GeSi) contribute to increase of strength (by ~ 7 %) in the joint of alloy A6061 [36] in comparison with joint produced using the brazing filler metal Al–9.5Si–10Ge–15Zn [37] at brazing temperature 580 °C.

Acceptable melting temperature (525–565 °C) is typical for hypoeutectic alloys of Al–Si system containing zinc (10–50 wt.%) [38]. In the triple alloy Al–Si–Zn at silicon content > 1.6 wt.% it solidifies in form of acicular phase [39]. Microadditives of 0.09Sr [40], 0.06Ce and 0.08 wt.% Ti [41] refine primary crystals of (Si) phase in the brazing filler metal Al–6.5Si–42Zn. In flame flux brazing of the aluminium alloy A6061 with Al–6.5Si–42Zn–009Sr brazing filler metal at 580 °C temperature and using forced cooling of a joint in water the strength can reach ~ 90 % of strength limit of base metal [40]. A disadvantage of the brazing filler metals of Al–Si–Zn system is possibility to dissolve base metal (at T = = 433 °C ~ 70 wt.% of Zn [23] dissolves in Al) that can promote failure of brazed thin-walled elements of multilayer aluminium structure. Thus, development of the light-alloy brazing filler metals and methods of brazing at temperature lower than 550 °C is currently relevant and being in demand in manufacture of separate structures and brazed assemblies from aluminium alloys.

The aim of present work is investigation of formation of joints of aluminium alloys AD00, AMts, AD31 using light-alloy germanium brazing filler metal (T_1 = 500 °C) under conditions of fluxless brazing in high-purity nitrogen medium.

MATERIALS AND INVESTIGATION PROCEDURE

The samples for investigations were made from aluminium alloys AD1, AMts and profile of alloy AD31 of 1 mm thick (Table 1).

A brazing filler metal of Al–Ge–Si–Cu system with Mn and Ti additives was made using induction melting in pure argon medium of metals (99.95Al, 99.95Cu, Ge polycrystalline) and addition alloys (Al–12Si, Al–2Ti, Al–10Mn) at temperature 700 °C in a crucible from fine-graphite of MPG-7 grade. Obtained alloy was poured in a copper mould at 570–580 °C temperature. Temperatures of solidus (T_s) and liquidus (T_l) of the brazing filler metal was determined by thermal differential analysis (TDA) using VDTA-8M3 unit under heating-cooling conditions (V = 40 °C/min) in helium medium. Preliminary an assembly (alloy of 1 g weight, thermal couple KhA of D = 0.1 mm, alundum crucible) was calibrated at temperature of pure metals (Al, Zn, Sn) solidification. The aluminium samples were cleaned before brazing: degreasing in solution 15 % NaOH, R = 50–55 °C, t = 60 s; etching in solution 20 vol.% of HNO_3, 2 vol.% HF, t = 60 s, rinsing between the operations in hot (T = 60–65 °C) and cold (T = 8–22 °C) water of double distillation. Brazing of the samples (Figure 2) with lap size 1 mm was carried out at 550±5 °C temperature and holding $t \leq 2.5$ min in flow nitrogen (99.999 vol.% N_2, 0.0005 vol.% O_2, 0.0007 vol.% H_2O) using a brazing paste (powder of brazing filler metal, coupling liquid — laprol 6003-2B-18).

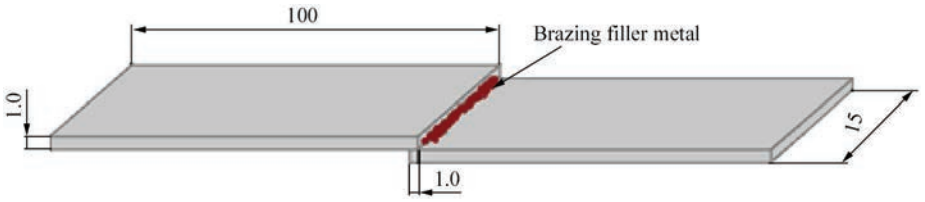

Figure 2. Sketch of lap joint

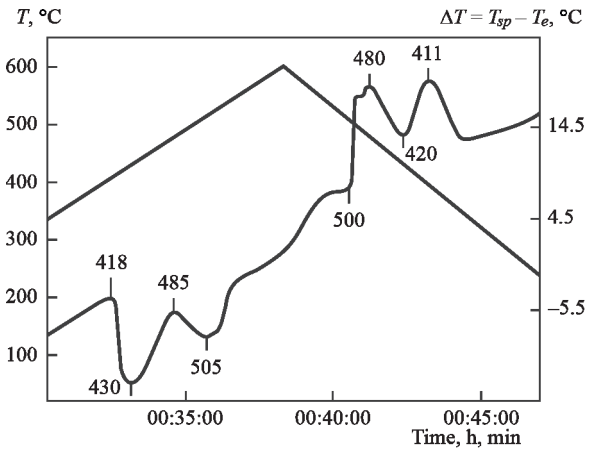

Figure 3. DTA thermogram of brazing filler metal Al–25Ge–5Si–5Cu–1.0Mn–0.15Ti

After heating turn down there was used a step-by-step cooling mode with holding at liquidus temperature of the brazing filler metal (500 °C) during 15 min.

Microstructure of the brazing filler metal in initial condition and in brazed joints of aluminium alloys was examined using scanning electron microscope JSM-840 with X-ray microprobe analyser Camebax SX50. Tensile tests were used in order to determine shear strength of the brazed joints at room temperature with set speed (V = 1 mm/s) of tensile machine grip travel.

INVESTIGATION RESULTS AND DISCUSSION

The results of high-temperature differential thermal analysis showed the temperatures of solidus and liquidus of brazing filler metal Al–25Ge–5Si–5Cu–1.0Mn–0.15Ti (Figure 3). Heat effects on the obtained thermal curve indicate presence of three phases, temperature interval of melting of which lies in T = 418–505 °C range.

It is necessary to note that a heat effect from eutectic component is weakly expressed that is caused by its amount. Microstructure investigations of Al–25Ge–5Si–5Cu–1.0Mn–0.15Ti brazing filler metal showed that the crystals of β-solid solution Si_xGe_y are in equilibrium with α-Al solid solution (grey colour) in a cast state. The solid solutions form separate areas with eutectic component which is a mesh from lamellar crystals of white colour precipitated on the boundaries of grains of solid solution based on al-

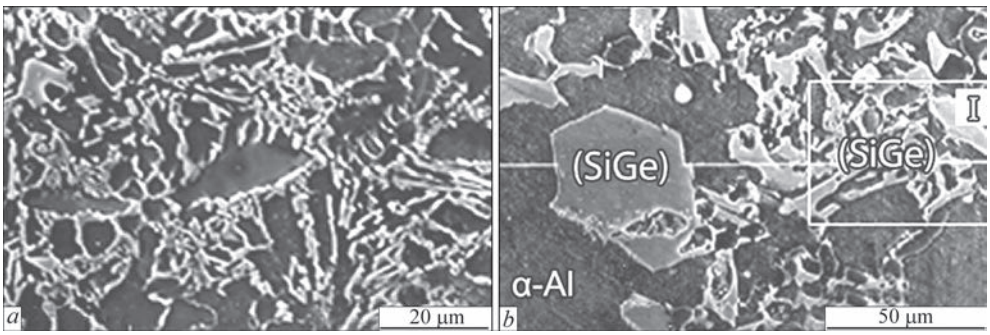

Figure 4. Microstructure of brazing filler metal 63.6Al–25Ge–5Si–5Cu–1.5Mn–0.15Ti: *a* — in cast state; *b* — in state of annealing by mode (400 °C/15 min)

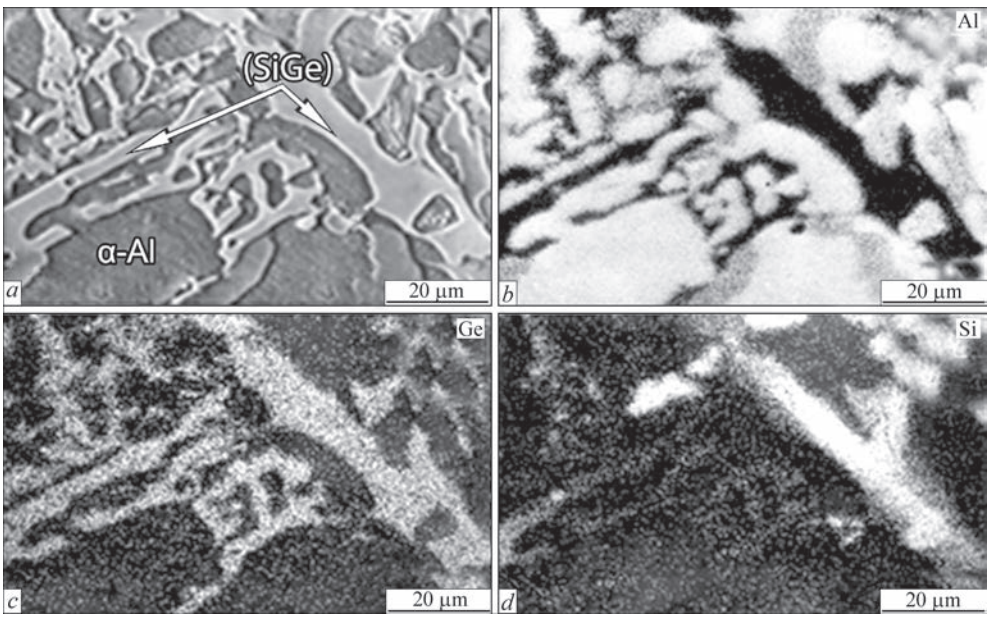

Figure 5. Electron image (*a*) of microstructure of brazing filler metal and maps of distributions of elements Al (*b*), Ge(c), Si (*d*)

uminium (Figure 4, *a*). Obtained data are well correlated with corresponding binary diagrams of state of metallic systems (Figure 1, *b*, *c*). Besides separate particles of germanium-based phase of ~ 40 μm size were found in the brazing filler metal: 6.65Al–87.65Ge–4.8Si–0.73Cu–0.17Mn–0.2Ti

The alloys of Al–Ge and Al–Si systems are characterized by minimum melting temperature in eutectic formation and the alloys of Si–Ge system differ by presence of the continuous solid solutions of alternating concentration (Figure 1, *c*) and higher melting temperature [20].

Content of aluminium, germanium and silicon in the brazing filler metal indicates the area of triple diagram Al–Si–Ge [31] with reaction L↔(Al)+(Si_xGe_y) where the next constituents exist in equilibrium, namely liquid phase, aluminium solid solution, β-phase (Si_xGe_y). Cooling (4 K/min) of the alloys of Al–Ge–Si system promotes formation of lamellar crystals of Si_xGe_y phase with different weight relationship of Si/Ge [31].

The results of X-ray microanalysis determined that annealing promotes the following phases in the brazing filler metal, namely solid solution based on

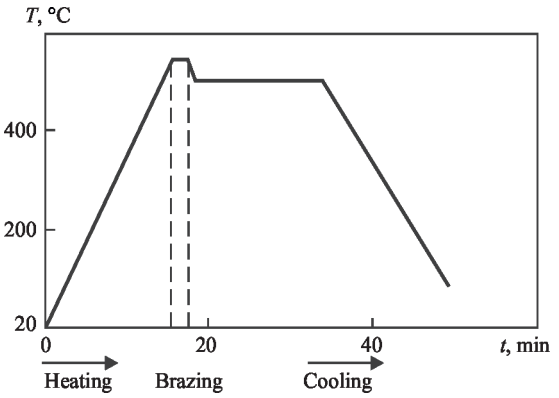

Figure 6. Thermal cycle of brazing by brazing filler metal of Al–Ge–Si–Cu system of aluminium alloys in nitrogen of high purity

aluminium 93Al–4.6Ge–0.1Si–1.4Cu–0.07Mn–0.01Ti(α-Al), crystals of phase Si_xGe_y that contain 0.40Al–42.85Ge–56.58Si–0.17Cu (Mn, Ti traces), eutectic component and phase based on germanium 39.44Al–59.05Ge–0.67Si–0.84Cu (Mn, Ti traces) (Figure 4, *b*). After annealing it is possible to observe increase of sizes of the grains of Si_xGe_y phase in comparison with initial condition. They are characterized by alternating concentration of component elements. Work [42] indicates that in the alloys of Al–18Si–*x*Ge system at Ge content up to 60 % the grains of prima-

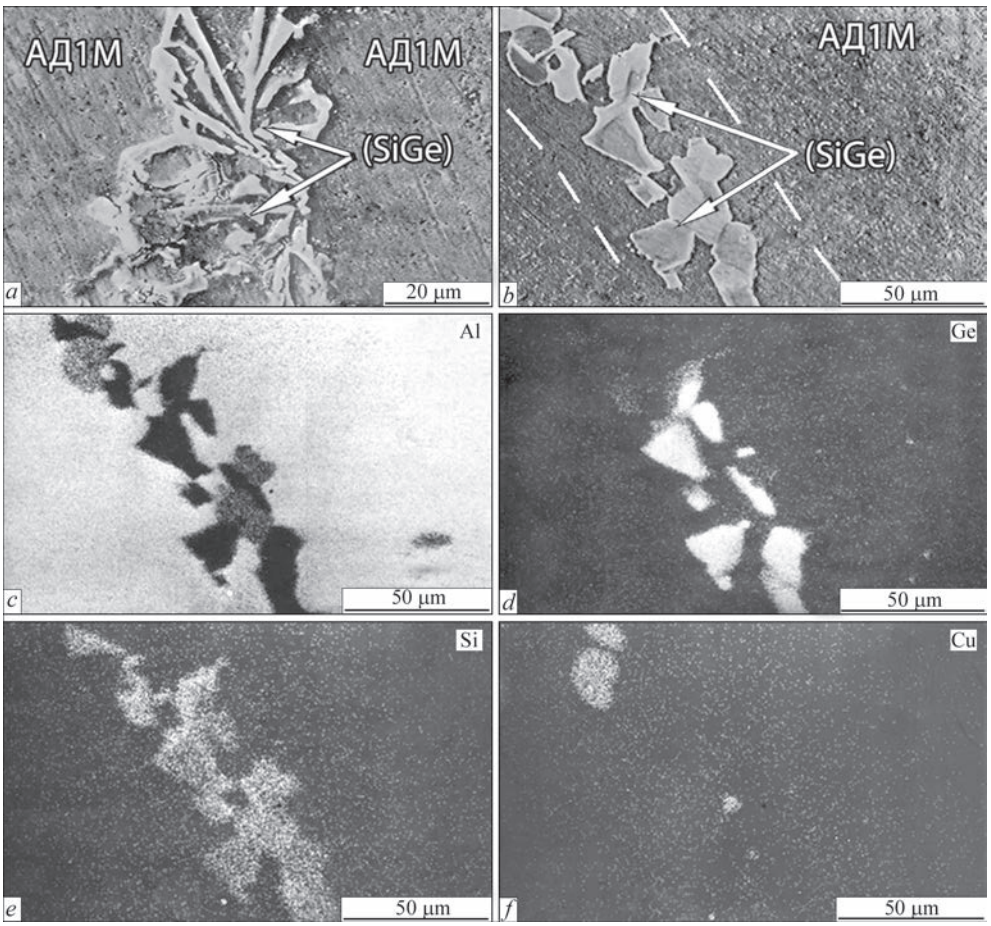

Figure 7. Electron image of microstructure of brazed weld of alloy AD1M in initial condition after brazing (*a*), after step-by-step cooling (*b*) and maps of distribution of chemical elements: aluminium (*c*), germanium (*d*), silicon (*e*) and copper (*f*) in application of Al–25Ge–5Si–5Cu–1Mn–0.15Ti brazing filler metal

ry phase (Si_xGe_y) become sufficiently coarse and can have size of several hundred micrometers.

Mapping of microstructure of brazing filler metal (Figure 5, *a*) allowed determining separate phases which contain increased concentration of aluminium (Figure 5, *b*), germanium (Figure 5, *c*) and silicon (Figure 5, *d*) that correlate with quantity determination of composition of separate phases.

Heating of the aluminium samples to temperature 550±5 °C that exceeds liquidus temperature of the brazing filler metal by 50 °C (with 1.5–2 min holding) in high purity nitrogen medium using germanium brazing filler metal provides its melting and wetting of base metal. Cooling in keeping with thermal cycle (Figure 6) provokes formation of the brazed joints of aluminium alloys AD1M, AMts, AD31 with full penetration.

Using X-ray microanalysis there was examined a structure of the joint from AD1M alloy after brazing (Figure 7, *a*) and after step-by-step cooling mode (Figure 7, *b*). It was determined that the structure of brazed weld from alloy AD1M contains α-solid solution based on aluminium; lamellar crystals of β-phase (Si_xGe_y) and low melting eutectic component.

Obtained results of the investigation showed that composition of metal of the brazed weld has little difference from composition of the brazing filler metal in initial condition. Concentration of elements in α-Al solid solution 94.22Al–3.79Ge–0.9Si–85Cu–0.02Ti–0.26Mn virtually does not change. In eutectic (39.44Al–59.05Ge–7.11Si–0.84Cu) there is insignificant increase of silicon concentration from 0.67 to 7.11 % and change of morphology of crystals of (Si_xGe_y) phase from lamellar to faceted and increase of their size (Figure 7, *b*).

Mechanical tensile tests (at room temperature) allowed determining that strength of the overlap brazed samples from alloy AD1M is higher than the strength of base metal, failure takes place on AD1M base metal, but not on the brazed weld. Short-term shear strength of the joint from alloy AMts makes 82 MPa. And shear strength of the brazed joint from alloy AD31 after step-by-step cooling with holding at brazing filler metal liquidus temperature (500 °C/15 min) rises from 84 to 102 MPa.

CONCLUSIONS

There were determined the temperature-time modes (T_m = 550±5 °C, $t \leq 2$ min) of formation of quality joint from aluminium alloys AD1M, AMts and AD31 at fluxless brazing using brazing filler metal Al–25Ge–5Si–5Cu–1Mn–0.15Ti in high purity nitrogen.

X-ray microanalysis determined that after brazing the structure of brazed weld of joint from aluminium alloy (AD1M) contains two solid solutions, namely based on aluminium (α-Al) and based on β-phase (Si_xGe_y) which form eutectic component.

The results of mechanical tests (at room temperature) proved that strength of the brazed weld from alloy AD1M is higher than the strength of base metal (σ_t = 60MPa). Shear strength of the brazed joint from alloy AMts makes 82 MPa.

Application of step-by-step cooling for the brazed joint from alloy AD31 promotes increase of shear strength from 84 to 102 MPa.

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ORCID

O.M. Sabadash: 0000-0002-9582-8673,
S.V. Maksymova: 0000-0003-0158-5760

CONFLICT OF INTEREST

The Authors declare no conflict of interest

CORRESPONDING AUTHOR

S.V. Maksymova
E.O. Paton Electric Welding Institute of the NASU
11 Kazymyr Malevych Str., 03150, Kyiv, Ukraine.
E-mail: maksymova.svitlana15@ukr.net

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