	www.c2	asopisma.pan.pl	PAN www.jou	rnals.pan.pl ————	
A R C H I V E S	O F	МЕТА	LLURGY	A N D	M A T E R I A L S
Volume 59			2014		Issue A

DOI: 10.2478/amm-2014-0253

A. WINIOWSKI*, M. RÓŻAŃSKI*

BRAZING OF ALLOY BASED ON Fe3Al ALUMINIDE

LUTOWANIE TWARDE STOPU NA OSNOWIE ALUMINIDKU Fe₃AI

Alloys based on the Fe₃Al intermetallic phase belong to a new generation of metallic materials intended for operation at higher temperatures and having properties something between those of metals and ceramic materials. They are characterised by relatively high oxidation resistance, high corrosion resistance, high-temperature creep resistance, high electrical resistivity, high abrasion resistance as well as resistance to erosion and cavitation. Although the material costs of these alloys are relatively low, they belong to materials which are very difficult to join by means of welding methods. For this reason, joining such materials remains an important and current research and technological problem. One of the methods used for joining such materials is brazing. This work shows the results of technological tests concerned with vacuum brazing an alloy based on the Fe₃Al (Fe86Al14) phase using silver (Ag72Cu28) and copper-nickel (Cu90Ni10, Cu95Ni5) filler metals as well as presents the results of tests on the mechanical and structural properties of obtained joints.

Keywords: vacuum brazing, Fe₃Al based alloy, brazing parameters, mechanical properties of brazed joints, structural properties of brazed joints

Stopy na osnowie fazy międzymetalicznej typu Fe₃Al należą do nowszej generacji materiałów metalicznych, przeznaczonych do pracy w podwyższonych temperaturach, o właściwościach pośrednich pomiędzy metalami a materiałami ceramicznymi. Charakteryzują się one przede wszystkim stosunkowo wysoką odpornością na utlenianie i korozję, żarowytrzymałością, wysoką rezystywnością elektryczną, a także wysoką odpornością na ścieranie, erozję i kawitację Stopy te, o stosunkowo niskich kosztach materiałowych, należą jednak do materiałów bardzo trudno spajalnych metodami spawalniczymi, dlatego zagadnienie ich łączenia wciąż stanowi wciąż ważny i aktualny problem badawczy oraz technologiczny. Jedną z metod łączenia takich materiałów jest lutowanie twarde. W niniejszej pracy przedstawiono wyniki badań technologicznych z zakresu lutowania próżniowego stopu na osnowie fazy Fe₃Al (Fe86Al14), spoiwami: srebrnym (Ag72Cu28) i miedziano-niklowymi (Cu90Ni10, Cu95Ni5), a także wyniki badań właściwości wytrzymałościowych i strukturalnych uzyskanych połączeń.

1. Introduction

Iron and aluminium create a phase equilibrium system with numerous intermetallic phases, yet only the FeAl and Fe₃Al phases constitute the usable base of engineering materials as they meet the von Mises yield criterion, characteristic of structures being the derivatives of regular body-centred crystal lattice designated as A2. Alloys based on the FeAl and Fe₃Al intermetallic phases are characterised by advantageous utility properties such as excellent resistance to oxidation, carburising and sulfuration, good resistance to corrosion in seawater and melted salts, high electric resistivity (increasing along with temperature) as well as high resistance to abrasion, erosion and cavitation. Owing to a relatively high Al content these alloys are characterised by low density (lower than in other iron alloys) and a low price. However, their disadvantages include low plasticity and susceptibility to brittle cracking at the room temperature in the atmosphere of air and steam. The characteristic feature of both these intermetallic phases is the

abnormality of tensile strength in the function of temperature. It increases in the $300^{\circ}C \div 600^{\circ}C$ range and next significantly falls. The plasticity and the brittle crack resistance of the alloys based on the FeAl and Fe₃Al phases can be increased by the appropriate modification of their chemical composition and microstructure as well as by decreasing the degree of the atomic order and using protective coatings. These alloys find applications in the power generation sector (elements of boilers and burners, gas filters, heat exchangers, pipes, screens etc.) as well as in chemical, food, shipbuilding and automotive industries (heat exchangers, heating elements, tools, pipes, containers, tanks, valve seats, rings, elements of exhaust systems) etc. [1÷3].

Methods used for making inseparable joints of alloys based on the FeAl and Fe₃Al phases include TIG welding without a filler metal or TIG and MIG welding with the use of specialist nickel filler metals, electron welding, laser welding, friction welding, diffusion welding and brazing $[2\div11]$.

^{*} DEPARTMENT OF WELDING TECHNOLOGY IN INSTYTUT SPAWALNICTWA (INSTITUTE OF WELDING) – GLIWICE, POLAND

www.czasopisma.pan.pl

PAN POLSKA AKADEMIA NAUK

However, these alloys are very difficult to weld due to the following factors:

- high aluminium content and susceptibility to the intense formation of very durable aluminium oxide (Al₂O₃) on the surface,
- low ductility both at low and high temperatures,
- susceptibility to the formation of welding cracks,
- required heat resistance and high-temperature creep resistance of joints.

The available scientific and technical publications contain relatively little and fragmentary information about brazing alloys based on the Fe-Al types of phases, which indicates that the problem is still at the experimental stage. The advantage resulting from using brazing for joining alloys based on the Fe-Al phases is the fact that their low ductility at the room temperature, usually making welding and friction welding difficult, in this case does limit the use of this joining process or prevents obtaining of good quality joints. The presence of the film of the durable oxide Al₂O₃ is not the major technological problem either, as is the case with, for instance, diffusion welding. The studies presented were concerned with brazing (diffusion brazing) of alloys based on the Fe₃Al phase using filler metals (interlayers) made of copper, aluminium, nickel (BNi-2 according to AWS) and Au-Cu alloys. The tests were mostly related to the structural analysis of the mechanisms of the formation of brazed joints $[8 \div 11]$.

Recently, tests on brazing of alloys based on the Fe₃Al intermetallic phase have also been carried out at Instytut Spawalnictwa in Gliwice [18]. These tests are the continuation of research on brazing alloys based on nickel and titanium aluminides [12, 13, 17]. This article presents the results of the aforesaid tests focused on the effect of the material-technological conditions of brazing of alloys based on Fe₃Al by means of silver and copper-nickel filler metals on the mechanical and structural properties of joints.

The tests aimed at developing brazing technological conditions making it possible to obtain the most advantageous operational properties of brazed joints.

2. Base and filler metals used in tests

The alloy used in the tests was based on the Fe₃Al phase (Fe86Al14 % wt.) and had the form of cast cylindrical samples \emptyset 13×15 mm (made at the Department of Engineering and Cast Alloys and Composites of the Faculty of Foundry Engineering at the AGH University of Science and Technology in Kraków).

The filler metals (interlayers between joined parts) used for making brazed joints were silver filler metal grade B-Ag72Cu-780 (Ag 272) according to PN-EN ISO 17672 (72.45% wt. Ag, remainder Cu) in the form of 0.05 mm thick band [19] and copper- nickel filler metal grades B-Cu90Ni-1140/1100 (Ni – 10.55% wt., Cu – remainder) and B-Cu95Ni-1120/1085 (Ni – 5.37% wt., Cu – remainder) in the form of 0.1 mm thick band.

3. Selection of brazing parameters and making of test joints

The shear strength tests and the structural tests of the brazed joints made of the alloy based on the Fe₃Al (Fe86Al14) phase involved the use of butt-brazed cylindrical test joints. Previous experimentation acc. methodology worked out at Instytut Spawalnictwa [12÷16] has revealed that the cylindrical type of the sample favours the economical use of materials (base and filler metals) and when supported by appropriate fixtures enables obtaining shear stresses in the brazed joint during strength tests.

After chemical etching and matching with brazing metal shapes (in the number ensuring obtaining 0.10 mm thick layer) all the samples were brazed in a TORVAC S 16 furnace in the vacuum range $10^{-3} \div 10^{-5}$ mbar. The temperature of the samples during brazing was monitored with a Pt-PtRh13 contact thermocouple being part of the vacuum furnace.

The adjustment of temperature and brazing time depended on the type of a brazing metal and its melting point and was determined on the basis of available reference publications $[1\div3]$ and after the analysis of appropriate phase equilibrium systems Fe-Al, Fe-Cu, Cu-Al, Ni-Al. The samples were subjected to diffusion brazing. For this reason the brazing temperatures significantly exceeded the melting points of the brazing metals and the times at which the joints were held at such temperatures were relatively long.

The samples made of the Fe86Al14 alloy were brazed at the following brazing temperatures and hold times:

- in the case of copper-nickel filler metals grade B-Cu90Ni-1100/1140 (melting point of 1100÷1140°C) and B-Cu95Ni-1085/1120 (melting point of 1085÷ 1120°C) – the brazing temperature amounted to 1200°C, the hold time was 30÷180 min.
- in the case of silver filler metal B-Ag72Cu-780 (brazing metal Ag72Cu28 having a eutectic-like composition, melting point of 780°C) the brazing temperature amounted to 1000÷1150°C, the hold time was 30÷300 min.

In all of the joints the brazing gaps were properly filled with the filler metal. The samples brazed with the silver filler metal were characterised by better wettability than the ones brazed with the copper-nickel filler metals. This was manifested by the fact that the side walls of the joined cylindrical elements were, to a considerable degree, whitened with the filler metal in the area adjacent to the braze.

4. Shear strength of brazed joints

The mechanical properties of the brazed cylindrical samples were determined acc. methodology worked out at Instytut Spawalnictwa [$12\div15$] with a testing machine manufactured by the Instron company (model 4210) by subjecting them to shearing in special holders designed in such a manner that the samples were subjected to shear forces only, without bending. The shear strength tests involved the use of three samples and were carried out for each variant of temperature–time brazing process parameters.

Test results after the statical processing are presented in Table 1.

1488

www.czasopisma.pan.pl



1489



1

Brazing	Parameters of brazing	Shear strength of brazed joints MPa
Shear strength of l	prazed joints of Fe80	TABLE 6Al14 alloy

No	filler metal	of brazing		MPa	
		Temp., °C	Time, min	Average	Stand. dev.
1		1200	30	168.6	87.0
2	B-Cu95Ni-1120/1085		60	202.2	4.4
3			180	181.8	13.2
4		1200	30	210.7	3.9
5	B-Cu90Ni-1140/1100	1200	60	233.3	43.4
6		1200	180	199.1	18.6
7		1000	30	99.2	4.9
8	D A 720 780		180	94.6	21.1
9	B-Ag72Cu-780		300	131.1	18.3
10		1150	60	95.6	22.2
11			120	130.1	20.1

In the case of copper-nickel filler metals the highest shear strength values of 233 MPa (B-Cu90Ni-1140/1100) and 202 MPa (B-Cu95Ni-1120/1085) were obtained for the joints made at 1200°C and with the hold time of 60 min. Further elongation and reduction of a hold time decreased the aforesaid property. The joints made using the B-Ag72Cu-780 brazing metal were characterised by the highest shear strength of 131 MPa for the brazing temperature of 1000°C and the hold time amounting to 300 min. Similar strength (130 MPa) was obtained by the joints brazed at 1150°C with the hold time of 120 min. Slightly lower shear strength was observed in the case of the joints brazed with the aforesaid brazing metal at 1100°C with the hold time of 30min (99 MPa) as well as at the temperatures of 1100 and 1150°C and the hold times of 180 min and 60 min (95 MPa) respectively.

5. Structural tests of brazed joints

The initial identification of the structures made during the technological tests of the Fe86Al14 alloy brazed joints were carried out by means of metallographic tests (acc. PN-EN 12797:2002) on a light microscope (type MeF4M) manufactured by Leica [20].

In the case of the copper-nickel filler metals (grade B-Cu90Ni-1100/1140 and B-Cu95Ni-1085/1120) the brazed joints of the Fe86Al14 alloy made at a temperature below 1200°C revealed numerous voids or the complete lack of a joint representing metallic continuity. Only the joints brazed at 1200°C were characterised by good quality (without any significant imperfections). As showed additionally EDS analyses, the structures of the brazes obtained were composed of the mixture of solid solutions based on copper with a diversified aluminium content: $14 \div 16\%$ (wt.) – darker precipitates and $7 \div 9\%$ (wt.) – brighter precipitates, enriched with nickel and iron (Fig. 1, 2 b). In these brazes it was possible to observe the globular particles of the material being joined (the alloy based on the Fe₃Al phase passed into the braze) with a high

copper content (even up to 20%) and nickel (Fig. 1, 2,a,b). For relatively longer hold times (approximately 180 min) at the brazing temperatures it was possible to observe the areas of the braze with the entire diffusion of the filler metal into the material being joined (Fig. 1, 2c). These joints also revealed the following imperfections:

- numerous voids (Fig. 4d), being probably the result of the Kirkendall effect;

- penetration of the filler metal between the grains of the material being joined (this penetration was already observed for the hold time of 60 min - Fig. 4b).

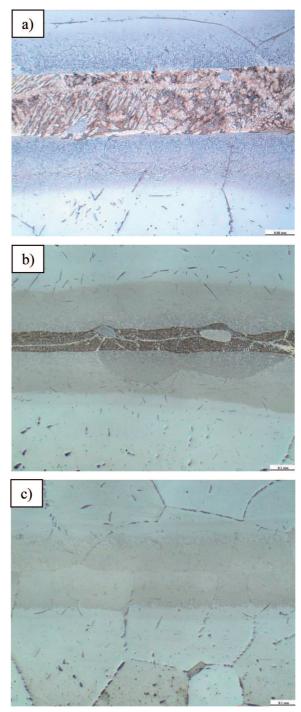


Fig. 1. Structure of Fe86Al14 alloy joints brazed with B-Cu90Ni-1100/1140 filler metal in the following brazing conditions: a) $1200^{\circ}C/60$ min, b), c) $1200^{\circ}C/180$ min (etch. 10 ml HNO₃+ 20 ml HCl + 30 ml glicerol, magn. 400-1000x)

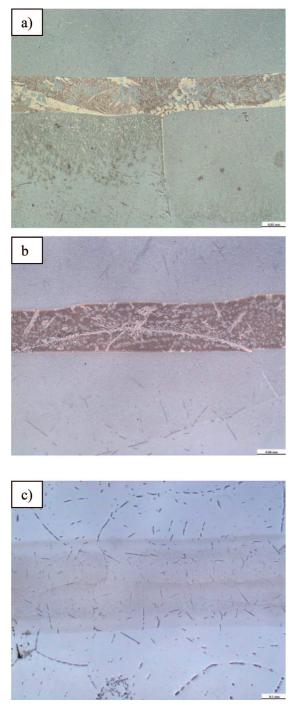


Fig. 2. Structure of Fe86Al14 alloy joints brazed with B-Cu95Ni-1085/1120 filler metal in the following brazing conditions: a) $1200^{\circ}C/60$ min, b), c) $1200^{\circ}C/180$ min (etch. 10 ml HNO₃+ 20 ml HCl + 30 ml glicerol, magn. 400-1000x)

In the case of the Fe86Al14 alloy joints brazed with the silver filler metal (grade B-Ag72Cu-780) the brazes obtained were characterised by the structure composed of solid solutions based on copper (dark precipitates) and silver (bright precipitates), similar to the eutectic structure of the filler metal (Fig. 3). The phases which were present in these joints were solid solutions which, apart from silver and copper, contained also the components making up the composition of the material being joined, i.e. aluminium and iron. Additionally EDS analyses showed, that the content of aluminium in the solid solution based on silver amounted to $0.46 \div 1.93\%$ (wt.), whereas

in the solid solution based on copper it was $8.48 \div 20.56\%$ (wt.) and increased along with the process temperature and the hold time at that temperature. The joints made with shorter hold times revealed cracks between the solid solution precipitates.

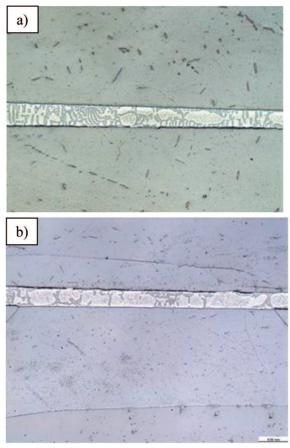


Fig. 3. Structures of Fe86Al14 alloy joints brazed with B-Ag72Cu-780 filler metal in the following brazing conditions: a) 1000° C/300 min, b) 1150° C/120 min (etch. 10 ml HNO₃+ 20 ml HCl + 30 ml glicerol, magn. 400x)

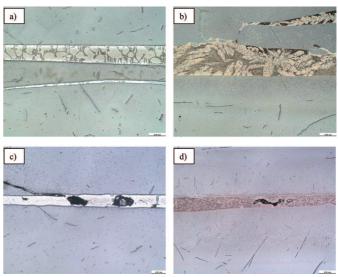


Fig. 4. The examples of brazing imperfections in Fe86Al14 alloy joints brazed with silver filler metal B-Ag72Cu-780 in temperature 1150°C and time 120 min (a, c) and brazed with Cu-Ni filler metal B-Cu95Ni-1085/1120 in temperature 1200°C and time 60 (b) i 120 (d) min (etch. 10 ml HNO₃+ 20 ml HCl + 30 ml glicerol, magn. 400-1000x)

www.czasopisma.pan.pl

In turn, the joints of the highest strength, made with the parameters of 1000°C/300 min and 1150°C/120 min revealed the capilary penetration of the brazing metal between the enlarge grains of the alloy as well as few voids being probably, as in the case of the joints brazed with copper-nickel filler metals, the result of the Kirkendall effect.

6. Conclusions

- The technological tests focused on the vacuum brazing of the Fe86Al14 alloy with the silver filler metal grade B-Ag72Cu-780 and copper-nickel filler metal grades B-Cu90Ni-1100/1140 and B-Cu95Ni-1085/1120 enabled obtaining good quality joints with the following brazing temperatures and times:
 - 1000°C/300 min and 1150°C/120 min for the silver brazing metal,
 - 1200°C/30÷180 min for the copper-nickel filler metals.
- The highest shear strength (131 MPa) of the joints made of the Fe86Al14 alloy brazed with the B-Ag72Cu-780 silver filler metal was obtained for the following temperature/time parameters: 1000°C/300min and 1150°C/120min.
- In the case of the joints made with the copper-nickel B-Cu95Ni-1120/1085 and B-Cu90Ni-1140/1100 the highest shear strength, i.e. 202 MPa and 233 MPa respectively, was obtained for the brazing parameters of 1200°C/60 min.
- 4. The structures of the joints made with the silver filler metal and copper-nickel metals at the brazing temperatures 1150 and 1200°C respectively, with longer hold times, contained few imperfections in the form of voids, characteristic of diffusion brazed joints and attributable to the Kirkendall effect.
- 5. The extension of welding time leads to the coagulation of dispersive precipitates having the chemical composition corresponding to the Fe_3Al phase and, as a result, to the reduction of the strength of the joints made using the B-Cu90Ni-1140/1100 filler metal.

REFERENCES

- Z. Bojara, W. Przetakiewicz, Materiały metalowe z udziałem faz międzymetalicznych. BEL Studio, Warszawa (2006) (in Polish).
- [2] R.W. M e s s l e r, Joining of Advanced Materials. Butterworth – Heineman, Stoneham (1993).
- [3] S.C. D e e v i, V.K. S i k k a, C.T. L i u, Progress in Materials Science **42**, 177-192 (1997).
- [4] J. Adamiec, P. Adamiec, Kongres JOIN-TEC 189 (2003).
- [5] J. A d a m i e c, A. G r a b o w s k i, A. L i s i e c k i, Materiały konferencyjne, 7STL, Szczecin (2002) (in Polish).
- [6] J. Adamiec, P. Adamiec, M. Więcek, Acta Metallurgica Slovaca 10, 745-748 (2004).
- [7] W. Włosiński, T. Chmielewski, M. Kucharczyk, Przegląd Spawalnictwa 1, 222-228, 2004 (in Polish).
- [8] Z. L i, R.K. S h i u e, S.K. W u, Intermetallics 18, 422-428 (2010).
- [9] R.K. Shiue, S.K. Wu, Y.L. Lee, Intermetallics **13**, 818-826 (2005).
- [10] R.K. Shiue, S.K. Wu, I-H Lee, Gold Bull 44, 49-56 (2011).
- [11] Y.L. Lee, R.K. Shiue, S.K. Wu, Intermetallics 11, 187-195 (2003).
- [12] A. Winiowski, M. Różański, Kovove Materialy 51, 2, 123 -128 (2013).
- [13] Z. Mirski, M. Różański, Archives of Civil and Mechanical Engineering 13, 4, 415-421 (2013).
- [14] A. Winiowski, Archives of Metallurgy and Materials 55, 4, 991-1000 (2010).
- [15] W. G a s i o r, A. W i n i o w s k i, Archives of Metallurgy and Materials 57, 4, 1087-1093 (2012).
- [16] A. Winiowski, M. Różański, Archives of Metallurgy and Materials 58, 4, 1007-1011 (2013).
- [17] A. Winiowski, M. Różański, Welding International 5, 331-335 (2011).
- [18] A. Winiowski, M. Różański, The Final Report of Statutory R&D Activity. Cc-53, Instytut Spawalnictwa, Gliwice (2010) (in Polish).
- [19] PN-EN ISO 17672:2010, Brazing Filler metals.
- [20] PN-EN 12797:2002, Brazing Destructive tests of brazed joints.

Received: 20 January 2014.