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Brazing of alloys based on intermetallic phase Ni_3Al

A. Winiowski*, M. Różański

Department of Welding Technology, Institute of Welding, Gliwice, Poland

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Abstract

Alloys based on an intermetallic phase type Ni_3Al-γ' belong to a new generation of metallic materials intended for use at higher temperatures and are characterized by properties intermediate between those typical of metals and those characteristic of ceramics. The features of the alloys in question include a high melting point and high-temperature creep resistance, resistance to chemical and gaseous corrosion (oxidation), high resistance to abrasion, recrystallization and creeping as well as relatively low density. The alloys are difficult to weld, and therefore constitute a significant research and technological challenge. One of the methods applied in joining such materials as well as joining them with stainless steels and nickel alloys is brazing. This paper presents results of technological tests focused on the brazing of an alloy based on the phase Ni_3Al-γ' (also with stainless steel) with copper (Cu) and silver filler materials (Ag72Cu28). In addition, the results of mechanical and structural tests related to the previously mentioned joints are discussed.

Key words: vacuum brazing, intermetallic-based alloy, mechanical properties of brazed joints, structural properties of brazed joints, brazing parameters

1. Introduction

The phase Ni_3Al is a secondary solid solution formed as a result of a peritectic transformation; the density of the phase being 7.4 kg dm^{-3}, Young's modulus – 200 GPa, unit elongation at room temperature – from a few up to 50 %, and the maximum temperature of the application of alloys based on the phase: 800–1150 ℃. Characteristics of the phase and alloys based thereon also include high creep strength (increasing with temperature), very good abrasion resistance at temperatures in excess of 600 ℃, resistance to oxidation and carburizing in oxidizing and reducing atmospheres, as well as resistance to thermal fatigue. The alloys are difficult to weld, which can be attributed to their significant aluminium content, a tendency of intensive formation of a very stable aluminium oxide (Al_2O_3) on their surface, low ductility at lower temperatures, tendency to welding cracking (crystallization, liquation and their combinations), as well as cracking at higher temperatures, e.g. during heat treatment [1–3].

Methods, which are used in the production of permanent joints of the aforesaid alloys, include TIG arc welding without filler material or TIG and MIG welding with specialist nickel filler material, electron beam welding, laser welding, friction welding, diffusion welding and brazing [4–15].

Available scientific and technical publications contain relatively little information on the brazing of such alloys. Test results presented in such literature of the subject are often fragmentary, do not contain information about the properties of joints and indicate that the issue is still at an experimental stage [12–15]. An advantage of the brazing of alloys based on the phases Ni-Al is connected with the fact that their low ductility at room temperature (making welding and friction welding difficult) is, in the case under discussion, no impediment to joining and obtaining good quality. In addition, the presence of a continuous film of stable oxide Al_2O_3 does not pose a major technological challenge as, for instance, in the case of diffusion welding.

Publications [12–15] present examples of the brazing of an alloy based on the intermetallic phase NiAl-

*Corresponding author: tel.: 48 32 3358235; fax: 48 32 231 4652; e-mail address: andrzej.winiowski@is.gliwice.pl

β and producing joints of the said alloy with nickel (99.5 % Ni) and superalloy MM-247, with the use of nickel filler material NiSi4.5B3.2 (grade BNi-3 acc. to AWS), copper filler material (99.8 % Cu) and aluminium filler material (Al), respectively. In the case of the alloy based on the phase Ni_3Al-γ', related tests involved the use of aluminium filler material only [14]. The studies presented in publications were concerned with brazing within a limited range of time and temperature parameters and related tests referred to the structural mechanisms of the formation of brazed joints. The authors did not present the results of the tests of the mechanical properties of brazed joints.

Recently also Gliwice-based Instytut Spawalnictwa has been involved in tests concerned with brazing alloys based on intermetallic phases Ni-Al [17].

The impact of technological and material conditions of the brazing of an alloy based on the phase Ni_3Al-γ' in similar joints, and additionally in joints with chromium-nickel stainless steel grade X6CrNiTi18-10 on the mechanical and structural properties of these joints is discussed in this paper. Joining of materials having diversified physical and chemical properties and obtaining joints characterised by good operation properties constitutes always a significant problem with reference to research and technology [8–10, 12–15].

The tests involved the use of copper brazing filler material (grade B-Cu100-1085) and, as an innovative solution, silver brazing filler material (grade B-Ag72Cu-780).

The purpose of the aforesaid tests was to develop material and technological conditions of brazing an alloy based on the phase Ni_3Al-γ' and brazing the alloy with chromium-nickel stainless steel grade X6CrNiTi18-10, ensuring the most advantageous operational properties of brazed joints.

2. Parent and brazing filler metals

The following parent metals were used during the investigation:

– alloy based on Ni_3Al-γ' intermetallic phase – a rod of a diameter of 12 mm, with the chemical composition according to the analysis: 86.72 % Ni, 13.28 % Al, produced in University of Science and Technology (AGH) in Cracow;

– stainless steel – 28 mm diameter rod, grade X6CrNiTi18-10 according to PN-EN 10088-1, with the chemical composition according to the analysis: 0.017 % C; 18.09 % Cr; 9.64 % Ni; 0.78 % Si; 1.37 % Mn; 0.20 % Ti.

The brazing filler metals used in the tests were:

– silver brazing alloy, grade B-Ag72Cu-780 (Ag 272) wg PN-EN ISO 17672:2010, in the form of a band being 0.05 mm thick;

– copper brazing filler metal (pure copper), grade B-Cu100-1085 (Cu102) wg PN-EN ISO 17672:2010, in the form of a band being 0.05 mm thick.

3. Selection of brazing parameters and production of test joints

The shear strength tests and structural tests of similar brazed joints of the alloy based on the phase Ni_3Al-γ' (Ni87Al13) and of dissimilar joints of the alloy with stainless steel grade X6CrNiTi18-10 involved the use of cylindrical butt brazed test joints. In case of the alloy based on the phase Ni_3Al-γ', the dimensions of the elements were ø $13 \times 15\ mm^2$, whereas in case of the steel X6CrNiTi18-10 – ø $25 \times 15\ mm^2$. According to previous experience [15], the cylindrical shape of the test piece favoured economical use of base metals and filler materials and, supported by appropriate instrumentation, made it possible to obtain shear stress in the brazed joint during strength tests.

After chemical etching and placing with brazing filler material profiles (3 profiles per one joint), the test pieces were vacuum-brazed in a vacuum furnace TORVAC S 16; the range being 10^{-3}–10^{-5} mbar. The temperature of the samples during brazing was controlled by means of a contact thermocouple type Pt-PtRh13 (an element of the furnace).

The selection of brazing time and temperature depended on the type of brazing filler material applied in the process and was established on the basis of information contained in available reference publications and after the analysis of related phase equilibrium systems. The method used in the production of test joints was that of diffusion brazing. Due to the foregoing, brazing temperatures significantly exceeded the melting points of brazing filler materials and hold times were relatively long. The decision to apply high brazing temperatures was also supported by conditions required for alloys in relation to thermal dissociation of aluminium-containing oxides in the alloy Ni87Al13.

In case of the test pieces made of the alloy Ni87Al13, brazing temperatures and hold times were as follows:

– in case of the copper filler material (pure copper, grade B-Cu100-1085, melting point 1085 ℃) – temperature 1150 ℃, hold time 60–180 min;

– in case of the silver filler material (grade B-Ag72Cu-780, melting point 780 ℃) – temperature 1000–1150 ℃, hold time 30–180 min.

The test pieces made of the alloy Ni87Al13 with the stainless steel grade X6CrNiTi18-10 were brazed using the copper filler material B-Cu100-1085, at a temperature of 1150 ℃, for a hold time of 180 min. In case of the brazing with the silver filler material B-Ag72Cu-780 the temperature was 1000 ℃ and the hold time was 30 min.

Table 1. Shear strength of brazed joints

No	Brazing materials	Filler metal	Brazing parameters		Shear strength of brazed joint (MPa)	
			Temperature (℃)	Holding time (min)	Mean value, R_t	Standard deviation
1	Ni_3Al+Ni_3Al	B-Cu100-1085	1150	180	110	17.9
2		B-Ag72Cu-780	1000	30	165	8.2
3			1000	60	147	10.3
4			1000	180	138	12.7
5			1150	60	5	0.9
6			1150	120	3	0.3
7	Ni_3Al+X6CrNiTi18-8	B-Ag72Cu-780	1000	30	122	6.4
8		B-Cu100-1085	1150	60	113	8.6

4. Shear strength of brazed test joints

Mechanical properties of brazed cylindrical test pieces made of the alloy Ni87Al13 as well as of the alloy and the stainless steel X6CrNiTi18-10 were tested with a testing machine Instron 4210. The test pieces underwent shearing in specially designed clamps, which enabled exposing the test pieces to shear strength only, without bending. Previous tests conducted at Instytut Spawalnictwa and information contained in reference publications indicate that the aforesaid shear strength test best reflects the sensitivity of the joints of materials (which, together with filler material, form brittle intermetallic phases) to shear stresses generated in the braze.

Shear strength tests were carried out for three test pieces and for each variant of time and temperature parameters of brazing time.

The results of the tests, after statistical processing, are presented in Table 1.

In case of joints made with the copper filler material B-Cu100-1085, the strength was tested only in relation to a hold time of 180 min and amounted to 110 MPa. The test pieces made with shorter hold times (60 and 120 min) revealed significant lacks of wetting of surfaces being joined and could be separated without applying a greater force.

In case of joints made with the silver filler material B-Ag72Cu-780, the highest shear strength (165 MPa) was obtained for the lowest brazing parameters (1000 ℃/30 min). The strength decreased with increasing temperature and hold time, and was particularly low at a temperature of 1150 ℃ (3–5 MPa).

In case of dissimilar joints, i.e. the alloy Ni87Al13 + steel X6CrNiTi18-10, brazed with copper filler material B-Cu100-1085, it was possible to observe that after applying a temperature of 1150 ℃ and a hold time of 60–180 min, the results of shear tests were quite similar (91–113 MPa) and tended to decrease with increasing hold time.

The shear strength of this type of dissimilar joints brazed with the silver copper material B-Ag72Cu-780 was tested for joints made with the use of parameters ensuring the highest strength for the alloy Ni87Al13 (1000 ℃/30 min); the strength amounted to 122 MPa and was lower than that of the joints made of the alloy Ni87Al13 (165 MPa).

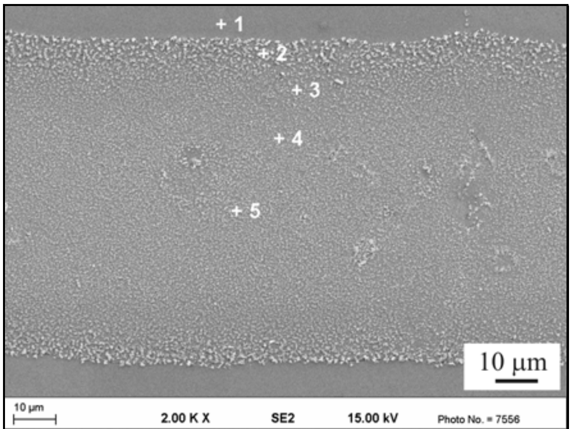
5. Metallographic and structural tests of brazed test joints

The initial identification of the structure of similar joints made of the alloy Ni87Al13 and of dissimilar joints made of the same alloy and the stainless steel grade X6CrNiTi18-10 was carried out using a Leica-manufactured light microscope MeF4A [17]. The identification, through metallographic tests, involved the determination of the morphological structure (i.e. complexity) of the joints, their appearance, shape, arrangement of phases and colouring.

In order to examine the morphology and structure closer as well as to analyse the chemical composition of individual phase components in related brazed joints, it was necessary to carry out additional structural examination by means of a scanning microscope LEO GEMINI 1525 with energy dispersion (EDS) micro-analyser ROENTEC.

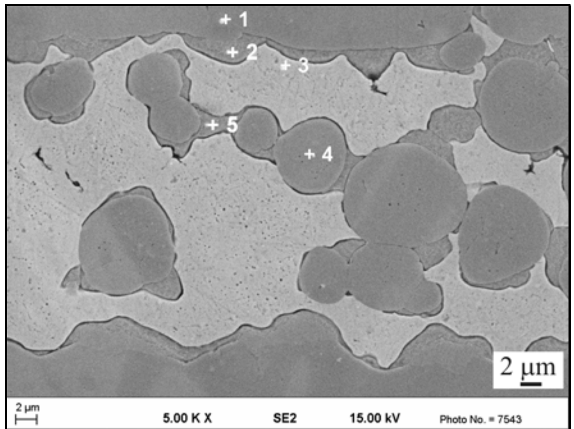
Figure 1 presents the structure of the joint based on the phase Ni_3Al (along with the results of EDS analysis), produced with the copper filler material Cu100-1085, at a temperature of 1150 ℃, with a hold time of 180 min. A braze obtained in the process composed of solid solutions of copper and nickel (with an addition of 5–7 % Al), with a nickel content of 21–43 %, decreasing towards the middle of the joint.

Figures 2 and 3 (along with the results of EDS analysis) present the structures of the joints of the alloy



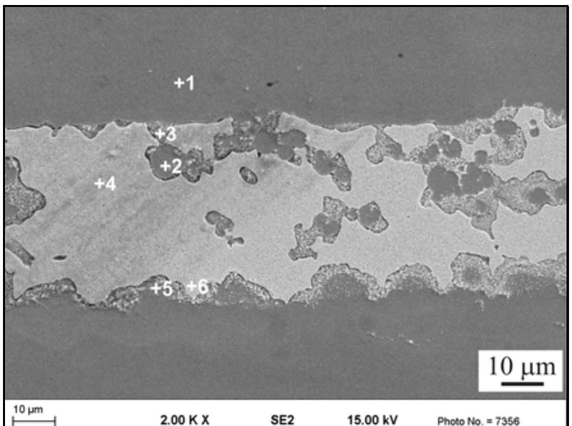
Point denotation	Chemical composition, % wt.		
	Ni	Al	Cu
1	71.43	14.57	14.00
2	43.14	7.35	49.50
3	35.14	8.69	56.17
4	30.38	6.53	63.09
5	21.26	5.04	73.70

Fig. 1. Microstructure (SEM) and results of energy dispersive spectrometry (EDS) of joint made of Ni_3Al alloy with B-Cu100-1085 copper filler metal at 1150 °C for 180 min.



Point denotation	Chemical composition, % wt.			
	Ni	Al	Cu	Ag
1	79.58	20.42	—	—
2	17.64	5.84	69.24	7.27
3	2.26	1.45	4.54	91.75
4	64.88	15.51	16.37	3.25
5	17.73	4.92	75.85	1.50

Fig. 3. Microstructure (SEM) and results of energy dispersive spectrometry (EDS) of joint made of Ni_3Al alloy with B-Ag72Cu-780 silver filler metal at 1000 °C for 180 min.



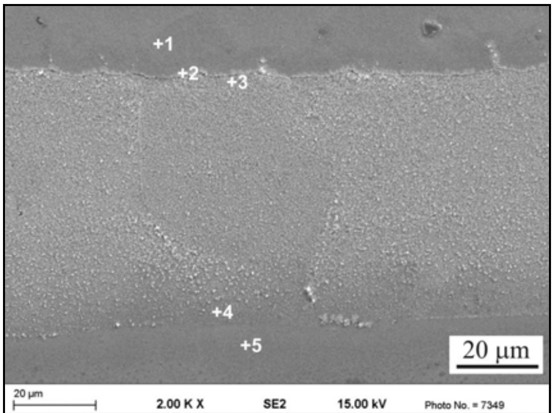
Point denotation	Chemical composition, % wt.			
	Ni	Al	Cu	Ag
1	82.28	17.72	—	—
2	65.94	15.45	18.82	—
3	16.86	3.18	62.85	17.11
4	—	0.05	15.39	84.56
5 (1)	50.95	7.82	39.44	1.79
6 (2)	0.27	0.18	18.88	80.67

Fig. 2. Microstructure (SEM) and results of energy dispersive spectrometry (EDS) of joint made of Ni_3Al alloy with B-Ag72Cu-780 silver filler metal at 1000 °C for 30 min.

being tested, brazed with the silver filler material of a near-eutectic composition (grade B-Ag72Cu-780). In the said structures, the middle part of a braze is the eutectic structure of molten brazing metal, composed of the solid solutions Ag-Cu and Cu-Ag. The diffusion zones of the joints are composed of globular precipitates of copper-enriched solid solution based on the phase Ni_3Al as well as of solid solutions Cu-Ni. The joints produced at higher brazing temperatures and with longer hold times fill the whole braze [16]. The eutectic base of the structures of the aforesaid joints is strongly depleted in the solid solution Cu-Ag based on copper; this being due to the fact that most of the said chemical element intensively passes to the solution based on the phase Ni_3Al. The amount of such brittle precipitates, increasing with an increasing brazing temperature and hold time, decreases the shear strength of the joints (which is demonstrated in the test results – Table 1) but, undoubtedly, increases their high-temperature creep resistance. Such a dramatic deterioration of the mechanical properties of the joints brazed at a temperature of 1150 ℃ (Table 1) can also be attributed to imperfections present in the silver phase (solid solution Ag-Cu) such as voids, probably caused by the evaporation of this phase at a relatively high temperature.

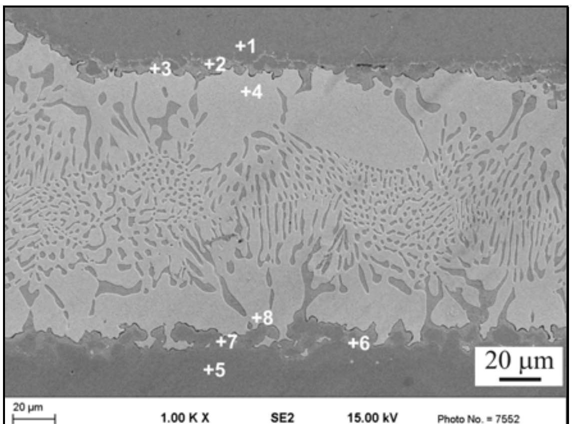
In case of the joints of the alloy Ni87Al13 with the stainless steel X6CrNiTi18-10 brazed with the copper filler material B-Cu100-1085, the structure of the braze was composed of the solid solution Cu-Ni, which was richer in copper from the steel side, whereas from the alloy Ni_3Al side it was richer in nickel (Fig. 4).

However, in case of the aforesaid joints brazed with the silver filler material B-Ag72Cu-780, in the diffusion zone from the steel side one can notice solid solutions based on copper with nickel, silver, aluminium, iron and solid solutions based on nickel with copper, aluminium, iron and silver as well as precipitates of phases Ni-Al (Fig. 5). From the side of the alloy Ni87Al13 one can observe the precipitates of solid solutions Cu-Ag and Cu-Ni. The middle part of a



Point denotation	Chemical composition, % wt.				
	Ni	Al	Cu	Fe	Cr
1	8.71	0.68	2.58	69.58	17.21
2	18.57	0.70	18.64	49.19	12.69
3	7.90	2.49	84.87	3.74	0.77
4	80.88	17.13	1.99	—	—
5	57.35	10.99	25.97	5.69	—

Fig. 4. Microstructure (SEM) and results of energy dispersive spectrometry (EDS) of dissimilar joint of Ni_3Al alloy (below) and X6CrNiTi18-10 steel (above), with B-Cu100-1085 copper filler metal at 1150 °C for 180 min.



Point denotation	Chemical composition, % wt.					
	Ni	Al	Cu	Ag	Fe	Cr
1	11.31	1.02	1.07	1.26	61.42	14.46
2	58.67	13.23	20.01	1.65	5.47	0.97
3	19.60	5.16	59.98	12.25	2.48	0.53
4	0.05	—	7.76	92.19	—	—
5	64.15	15.80	13.77	3.55	1.88	0.85
6	18.26	5.74	58.59	13.81	2.94	0.66
7	18.44	6.00	62.36	10.34	2.37	0.49
8	0.37	0.38	10.53	88.57	0.08	0.07

Fig. 5. Microstructure (SEM) and results of energy dispersive spectrometry (EDS) of dissimilar joint of Ni_3Al alloy (below) and X6CrNiTi18-10 steel (above), with B-Ag72Cu-780 silver filler metal at 1000 °C for 30 min.

braze is composed of the eutectic mixture Ag-Cu (the structure of molten brazing metal).

6. Conclusions

1. From among silver and copper filler materials selected through the analysis of reference publications and intended for the brazing of the alloy Ni87Al13 based on the intermetallic phase Ni_3Al-γ', the silver filler material grade B-Ag72Cu-780 and copper filler material grade B-Cu100-1085 can be applied in practice.

2. The maximum shear strength of the joints of the alloy Ni87Al13, vacuum-brazed with the silver filler material B-Ag72Cu-780, is 165 MPa or 100 MPa, if brazed with the copper filler material B-Cu100-1085.

3. Good quality and the highest shear strength of the joints of the alloy Ni87Al13, vacuum brazed with the silver filler material grade B-Ag72Cu-780 is ensured by a brazing temperature of 1000 ℃ and a hold time of 30 min; in case of the joints brazed with the copper filler material grade B-Cu100-1085 – by a temperature of 1150 ℃ and a hold time of 180 min.

4. Increasing a brazing temperature and extending a hold time during the brazing of the joints with the silver filler material B-Ag72Cu-780 resulted in decreasing strength and quality of the former; this being due to the formation of many brittle intermetallic phases, increasing the heat resistance of the joints.

5. The strength of the joints type: alloy Ni87Al13 – stainless steel X6CrNiTi18-10, brazed with the copper filler material B-Cu100-1085 and silver filler material BAg72Cu-780, while applying parameters ensuring the highest strength of the alloy Ni87Al13, amounted to 113 and 122 MPa, respectively.

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