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INVESTIGATION OF INTERACTION OF Ni_3Al-BASED ALLOY WITH INTERLAYERS OF DIFFERENT ALLOYING SYSTEMS FOR TLP-BONDING

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Fusion welding of cast high-temperature nickel alloys with high content of strengthening disperse phases is problematic. Even more serious problem is welding of intermetallic-based materials. Therefore, different methods of brazing are widely used for such materials joining. TLP-bonding (Transient Liquid Phase Bonding) is a term which has the most often application abroad. Considering that brazing filler materials have lower melting temperature than base metal, the concentration of depressants (reducing brazing filler metal melting temperature) in a brazed weld shall be reduced to the minimum in order to increase working temperature of brazed joints in brazing process. The depressants of high-temperature brazing filler metals are divided on several groups. Interaction of Ni_3Al-based alloy with brazing filler metals containing silicon, boron, zirconium and hafnium was investigated in the work. SBM-3 brazing filler metal of Ni–Cr–Co–Al–Ti–Ta–Re–W–Mo–Hf–B system was developed based on investigations results. 17 Ref. , 7 Figures.

Keywords: *brazing, nickel alloys, strengthening phase, depressants, brazing filler metal development, melting temperature*

The efficiency of gas turbines significantly depends on the temperature of working gas. Therefore, for the manufacture of guides and working blades, materials with higher heat resistance are developed. Such are the new alloys based on intermetallics, in particular, based on the intermetallic Ni_3A1. The promising methods of their joining are welding in the solid state, for example, friction welding, diffusion welding in vacuum with fused or nonfused interlayers or brazing [1, 2]. Brazing is a more universal method of joining, but its main problem is providing strength properties of brazed joints, which are close to those of the base metal. Good results are provided by TLP-bonding technology.

Ni_3A1intermetallic-based alloy is a structural material for gas turbines of a new generation [3, 4]. Ni_3A1 phase has a face-centered cubic lattice, in which chromium, molybdenum and tungsten are limitedly soluble, and the solubility of elements in this series decreases. The metals like titanium, tantalum and niobium dissolve in the γ'-phase, substituting aluminum and strengthening it. Cobalt has a high solubility in nickel, substituting nickel in the γ-solid solution. The ordered structure of the γ'-phase provides its high stability and operation of the alloy up to 1200 °C. The intermetallic, alloyed with small amounts of tantalum and chromium, has a high resistance in oxidizing environment at the temperatures up to 1100–1200 °C.

When joining different structures based on intermetallic Ni_3A1, operating at high temperatures and increasing the efficiency of gas turbines, joining technologies are required having both similar and dissimilar combinations with other high-temperature alloys. The solution to this problem is certainly relevant.

Problem statement. Aircraft materials science is successfully developed and intermetallic materials based on Ni_3A1have been known since the end of the last century [5, 6]. Investigations of methods of their joining began to be actively carried out only recently, and for the first time the possibility of applying existing brazing filler metals for brazing of high-temperature nickel alloys was considered in the works. These brazing filler metals can be divided into three groups [2]:

1) nickel-based alloyed brazing filler metals using silicon and boron as depressants, which in most cases are introduced together to reduce brazing temperature and concentration of each of them;

2) brazing filler metals based on alloyed nickel with the use of elements of the IV and V groups of the Periodic Table as depressants;

3) brazing filler metals of the system Ni–Pd.

In Ukraine and abroad, brazing filler metals of the first group, for example, VPr11, VPr11-40N, BNi1, BNi2, BNi5, VPr42, NS12, NS12A, STEMET 1301, STEMET 1311 are the most widely known. Complex alloyed brazing filler metals containing silicon and boron are VPr24, VPr27 and oth.

In works [7–9], complex alloyed brazing filler metals VPr36, VPr37 and VPr44 were investigated for brazing of high-temperature nickel alloys ZhS32, ZhS36 and EP975 alloy with VKNA-4U alloy based on the intermetallic Ni_3A1. The results obtained are little described, but the mentioned brazing filler metal systems show the possibility of increasing the high-temperature strength of brazed joints of alloys based on the intermetallic Ni_3A1.

The aim of the work is to study the interaction of the alloy based on the intermetallic Ni_3A1 with brazing filler metals of different systems and to increase the strength of joints, produced by TLP-Bonding technology, to the level of 70–80 % of the strength of the base metal.

Materials and procedure of investigations. For investigations, an experimental high alloy based on intermetallic $Ni_3A1$with the following content of basic elements (wt.%) was melted: 0.085 C; 6.6 Cr; 11.63 Co; 4.67 W; 1.49 Mo; 5.61 Si; 7.44 Ta; 1.6 Hf; 2.0 Ti; Ni is the rest. Microstructure of the alloy is shown in Figure 1. In the initial heat-treated state, the alloy has a uniform $\gamma + \gamma'$ structure with a high share of γ'-phase (68–72 vol.%).

The sizes and shape of specimens for investigations and mechanical tests were determined according to the accepted standard procedures (ISO 783–89) and the data given in work [10].

The base of the studied brazing filler metals was nickel alloyed with the same elements as the base material of the system Ni–Co–Cr–A1–Ti–Ta–W–Mo. For this purpose, the available computer programs for calculating the content of phases and critical temperatures were used. To reduce the melting temperature, as a depressant, boron and silicon and similarly zirconium and hafnium were together or separately introduced to the alloy.

The wetting quality of the high-temperature alloy was evaluated by wetting angle at different temperatures. The spreading of brazing filler metal was determined by specific spreading area. After that, the specimens were cut at the center of a drop of brazing filler metal and wetting angle was determined from a photo of macrosection.

To investigate the flow of brazing filler metal into the gap, wedge-shaped specimens with the sizes of 20×12×3 mm (lower) and 20×6×3 mm (upper) were used, which were exposed one on the other along the length with a zero gap on a one side and a gap of 0.3 or 0.6 mm on the other.

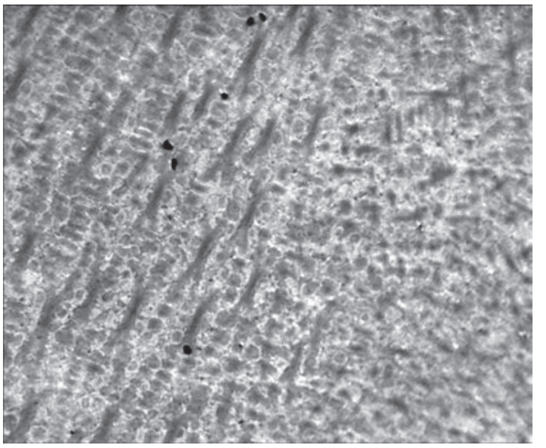

Figure 1. Microstructure (×45) of melted alloy based on Ni_3Al (magnific. by 1.5 times)

The formation of joints of butt welds was investigated on cylindrical specimens with a diameter of 13 mm and a length of 35 mm.

Structural investigations were carried out using optical metallography and scanning electron microscopy. The chemical composition was determined by local X-ray spectrum microanalysis by individual points and by area. The structure of high-temperature alloys was detected by chemical etching in a solution consisting of 10 g of chlorine iron, 30 ml of hydrochloric acid and 120 ml of alcohol or Marble's reagent: 100 ml of HCl, 20 g of $CuSO_4$, 100 ml of H_2O with addition of H_2SO_4 (0–20 ml). Murakami's reagent was used to differentiate between carbides and σ-phase: 10 g of red blood salt, 10 g of potassium hydroxide or 7 g of sodium hydroxide.

Differential thermal analysis was carried out in thermal analyzer HTDTA-8M with simultaneous measurement of temperature of the investigated specimen and the reference under heating and cooling in electric resistance furnace. The rate of heating and cooling was automatically maintained constant.

Investigations on determination of melting and crystallization temperatures were carried out in the atmosphere of high-purity helium. The rate of heating and cooling was 0.8 °C/s. Specimens with the mass of 1 g were placed in the crucibles of yttrium oxide Y_2O_3. The temperatures of phase transformations were determined using a calibration curve constructed on the melting points Al, Cu, Fe, and Pt. The cooling curves were used to qualitatively control the number of phase transitions and to determine the temperatures of the start of crystallization. The error of the results was ±7 °C.

Determination of mechanical characteristics was carried out during static short-term and long-term tensile tests of cylindrical specimens.

At the first stage, as applied to the melted alloy based on Ni_3A1, according to the results of the analysis of sources [7–9], for investigations and correction of composition brazing filler metal VPr36 was selected. To reduce the temperature of brazing, correction of the composition of brazing filler metal VPr36 was also carried out by the authors of works [11, 12], who introduced silicon into brazing filler metals for this purpose, using brazing filler metal NS12, containing 12 % of Si. For alloy based on Ni_3A1, a higher melting point of brazing filler metal is required, and to increase heat resistance of joints, the silicon content in the base metal is strictly limited. Therefore, silicon was not used in our investigations. In addition, in brazing filler metal it was necessary to use alloying by analogy with high-temperature alloys of a new generation, which was taken into account in this work.

When choosing depressants, the investigations were taken into account considered in work [13], in which the prospective use of such depressants as zirconium and hafnium in brazing filler metals was established.

Therefore, at the second stage of investigations in brazing filler metals for brazing intermetallic alloy, zirconium and hafnium were used as depressants. These elements form unlimited solutions between themselves and replace each other in the intermetallics of nickel in any ratios. The temperature of brazing using brazing filler metals with hafnium was 1225–1230 °C, and using brazing filler metals with zirconium was 1200–1210 °C.

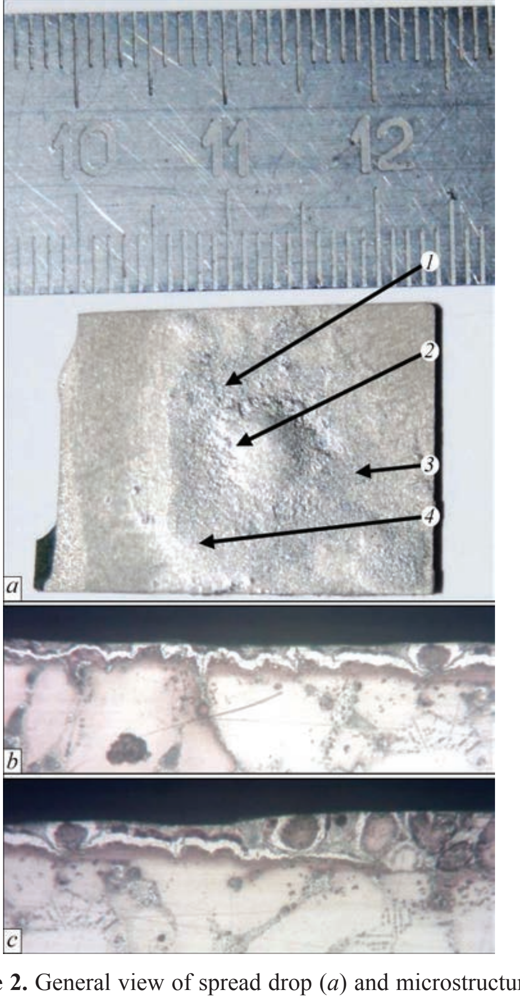

Figure 2. General view of spread drop (*a*) and microstructure of near-surface layer of brazing filler metal alloy in the halo of drop (*b*) and closer to its center (*c*) brazing filler metal with zirconium (see description *1–4* in the text)

The spreading of brazing filler metal with zirconium over the intermellic alloy is shown in Figure 2, where it is seen that spreading area of brazing filler metal consists of the central zone of brazing filler metal at the points *1* and *2*, the peripheral zone at the point *3* and the halo of a drop at the point *4*. The chemical composition of the metal in these zones is significantly different. In the central zone, the composition varies in height of the drop, but contains elements of the base of brazing filler metal. Further, in the peripheral zone and in the halo of a drop, the concentration of zirconium grows and an eutectic layer appears, which repeats micro roughness of the surface and envelops separate particles of brazing filler metal. With the introduction of 2.5 % of Zr in certain areas of the surface, its concentration increases to 11–12 wt.%. When introducing 5.0 % of Zr into brazing filler metal, in some areas the concentration of zirconium reaches 21 %, the concentration of niobium (up to 30 %) and tungsten (up to 14 %) sharply increase, the concentration of nickel (21–25 %), aluminum and titanium decreases, which testifies the formation of intermetallics.

An analogue of zirconium is hafnium, which, unlike zirconium, has a low diffusion mobility of atoms. Both elements have a low solubility in nickel and similar state diagrams with nickel. A low solubility of hafnium and zirconium in nickel along with the formation of eutectics broaden the temperature range of melting and crystallization of brazing filler metals, which is bad for alloys based on Ni_3A1intended for operation at temperatures up to 1200 °C. Brazing filler metals with zirconium and hafnium have a good wetting of the alloy based on Ni_3A1 at a vacuum of 10^{-3} Pa, as well as an uneven distribution in the brazed weld. The wetting angles, depending on the composition of brazing filler metal and temperature, vary from 13 to 5 degrees. Taking into account a high affinity of zirconium and hafnium to oxygen during brazing, it is necessary to strictly control the pressure in the working vacuum chamber (not more than $3\cdot10^{-3}$ Pa) and the leakage value, not more than $3\cdot10^{-5}$ Pa·m^2·s^{-1}.

Brazing filler metals with zirconium for brazing the alloy ZhS6U were also used in work [14]. Two brazing filler metals of the system Ni–Co–Cr–Ti–Nb–A1–(Me)–Zr, containing 1 and 2 % of Zr were investigated. Their temperatures are: solidus — 1101 and liquidus 1231 °C with 2 % of Zr and respectively 1141 and 1259 °C with 1 % of Zr. For application brazing filler metal with 1 % of Zr is recommended. As to the brazing temperature, brazing filler metal is not suitable for brazing alloy based on Ni_3A1. Brazing filler metals with zirconium and hafnium are well studied in works [15–18] as-applied to high-temperature nickel alloys with a brazing temperature up to 1210 °C. An increase in brazing temperature can lead to a degradation of the structure of the base metal.

At the third stage, brazing filler metal with the use of boron as a depressant was investigated. These brazing filler metals are the most studied and widespread in industry. During the development of brazing filler metal two problems were solved: the choice of the base of brazing filler metal and the determination of the necessary concentration of boron to provide the necessary liquidus and solidus temperatures. Brazing filler metals with boron traditionally have a high manufacturability.

When choosing a base of brazing filler metal, general provisions of the development of high-temperature alloys were used based on the influence of each of the alloying elements, including alloys of a new generation, on mechanical properties, formation of strengthening phases as well as brittle phases, heat resistance, liquidus, solidus, solvus temperatures, temperature of evolution and number of phases, their composition and other. At the same time, aircraft materials, in particular, intermetallic Ni_3A1 and alloys for marine gas turbines, operating at lower temperatures, but under conditions of high-temperature salt corrosion (HSC), were considered. The operating conditions of marine turbines significantly affect the chromium content in alloys, reducing their heat resistance.

The found ways to increase heat resistance and resistance against HSC allowed creating alloys for marine turbines of a new generation with an increase in their operating temperature by 50–60 °C [19]. Accordingly, it is necessary to increase heat resistance of joints, as well as brazing temperature. To do that, it is necessary to create new brazing filler metals, using the same principles of alloying, which are used to create alloys.

Many other problems are common to aircraft and ship gas turbine building, which allows using the same calculation methods and computer programs. When choosing the base of brazing filler metals, calculations were also used, but with the same base systems of brazing filler metals, the concentrations of a number of elements in them are significantly different. For example, the resistance of alloys against the formation of σ-phase depends on the group of elements, including chromium. Therefore, at a high chromium content, it is necessary to reduce the concentrations of other elements, strengthening a solid solution, at the same number of electronic vacancies and for this purpose the alloy was doped with elements, which are the

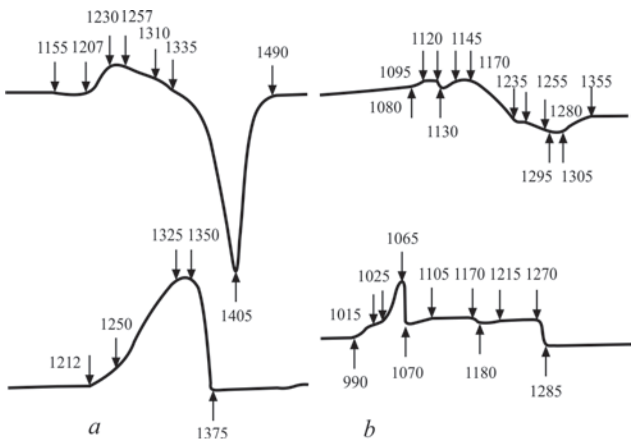

Figure 3. HTDTA curves for base of brazing filler metal (*a*) and after introduction of boron (*b*)

most effective strengtheners. Such alloying is used in high-temperature alloys of a new generation.

To calculate the second problem (choice of boron concentration), it is necessary to have a regression equation of the effect of boron on the liquidus and solidus temperature of the brazing filler metal base. There is not enough statistical data to do that. Therefore, the problem was solved experimentally. Several alloys were melted (brazing filler metal base), to which boron was added at three different concentrations. The produced specimens were subjected to HTDTA, investigations on wetting and spreading of brazing filler metal. In Figure 3, *a* the thermogram of one of the melted alloys is shown (brazing filler metal base), and in Figure 3, *b* — HTDTA results after addition of boron.

The spreading of one of the brazing filler metals in the alloy based on Ni_3A1 is shown in Figure 4.

Depending on the base of brazing filler metal and boron concentration, the wetting angle and specific spreading areas are changed. The main role is played by brazing temperature. All melted brazing filler metals wet the alloy well, but the temperature range of brazing filler metal application was determined by the wetting angle of not more than 10°. The optimum angle is 3–5°. The specific area of spreading during the

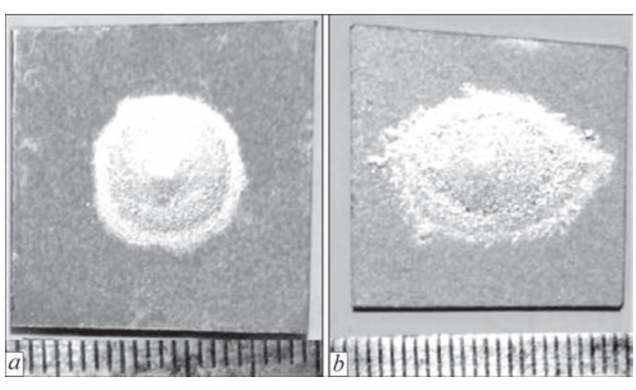

Figure 4. Spreading of batch weight of 100 mg of powder of brazing filler metal SBM-3 on alloy Ni_3A1 at temperatures of 1250 (*a*) and 1265 °C (*b*)

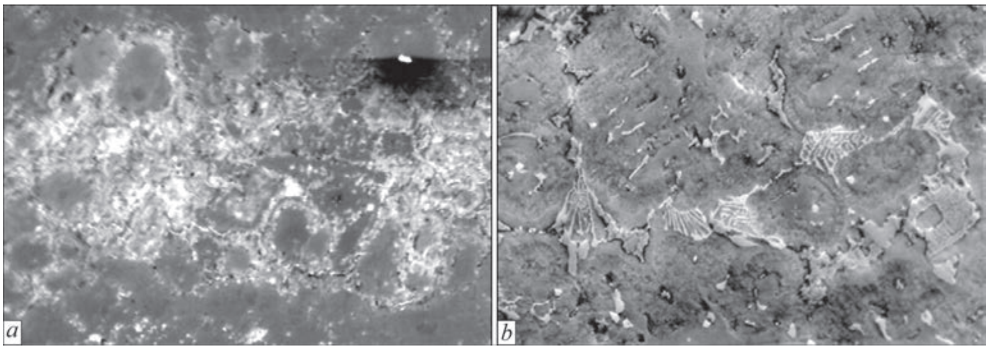

Figure 5. Microstructure of weld metal in brazing of wedge specimen with a gap of 361 (*a*) and 297 μm (*b*)

temperature of investigations of 1250–1265 °C was 1.7–2.0 mm^2/mg.

The formation of a brazed joint was estimated by the structure of the weld, its chemical composition (by area and phase), as well as by the maximum and minimum gaps, which are filled without defects at a temperature of brazing. This can be determined most accurately by brazing wedge-shaped specimens with a change in the gap from zero to 0.6 mm. The depth of dissolution of the base metal was also determined on such specimens. Regardless of the chemical composition of brazing filler metal, with an increase in the temperature of brazing the depth of dissolution of base metal increases, and it is as larger as wider the gap and the higher the boron concentration. The change in the concentration of boron also changes the number and composition of phases: γ, γ′, M_xC_y, M_3B. When analyzing by areas, the composition of the alloy varies little. The combined volume fraction of the carbide and boride phases increases already with increasing boron concentration from 1.0 to 1.2 wt.%, and the content of chromium, molybdenum and tungsten is almost half decreased.

The microstructure of the weld metal, brazed with a filler metal containing 2.5 % of Re, is shown in Figure 5.

According to the results of the analysis of the structure and chemical composition of melted specimens, brazing filler metal SBM-3 was developed, the microstructure of joints of which is shown in Figure 6.

Analyzing the results of investigations of phases and chemical composition of joints of the alloy based on Ni_3A1, produced by TLP-bonding method with brazing filler metal SBM-3, it should be noted that the matrix of brazing filler metal has a composition close to that of the base metal, plus individual elements, which is not in the base metal, but they are introduced into brazing filler metal. The structural composition of the weld metal and base metal are close. The joints structure has no continuous eutectic interlayers, carbide, boride or carboboride precipitations.

Brazed joints pass heat treatment, including high-temperature homogenization and staged cooling. The mode of heat treatment of the alloy includes heating up to 1180 °C with the exposure of 2 h → heating to 1265 °C with the exposure of 2 h and air cooling, heating up to 1050 °C with the exposure of 4 h and air cooling. After heat treatment, the boride eutectic is absent due to the formation of highly-dispersed borides and carboborides, which are characterized by the presence of active carbide and boride formers, which are clearly determined on the spectra during local X-ray spectral analysis. All such inclusions have a low aluminum content.

Mechanical tests of joints were carried out at operating temperatures with the determination of short-term and long-term strength on the basis of 50 and 100 h of tests. The test for the short-term strength of

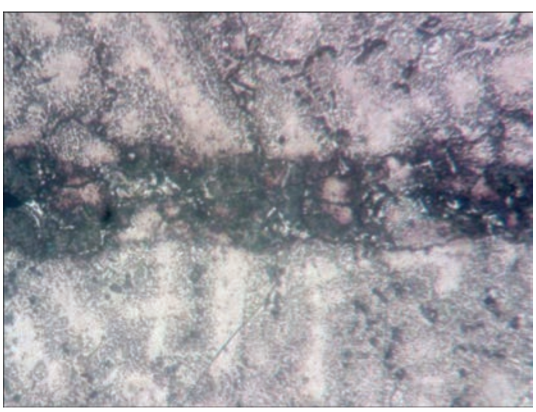

Figure 6. Microstructure (×250) of joint of the alloy based on intermetallics Ni_3A1 in brazing with a constant gap of 0.08 mm by brazing filler metal SBM-3

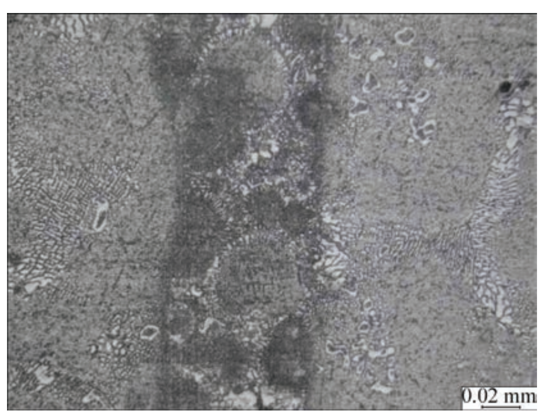

Figure 7. Microstructure of joint of alloy based on Ni_3Al with brazing filler metal SBM-3

the polycrystalline base metal at 900 °C showed a mean value: σ_t = 830 MPa, $\sigma_{0.2}$ = 720 MPa; at 1000 °C σ_t = 530 MPa; $\sigma_{0.2}$ = 460 MPa. The strength of joints was at the same level. The fracture of the specimen occurred on the base metal. The microstructure of a joint in the butt zone is shown in Figure 7.

The long-term strength of joints of Ni_3A1based alloy at a temperature of 900 °C on the basis of tests of 100 h amounted to σ_{100}^{900} = 280 MPa, which is equal to 81.9 % of the strength of base metal, based on 50 h of σ_{50}^{900} = 320 MPa, which is 85.6 % of the strength of base metal.

On the basis of the carried out investigations, brazing filler metal SBM-3M for brazing of high-temperature nickel alloys of marine gas turbines was also developed, a significant difference of which from brazing filler metal SBM-3 is a higher chromium content. Both brazing fillers metals are developed on the base of Ni–Co–Cr–A1–Ti–Ta–Re–W–Mo–B system.

Conclusions

1. The work proposes new approaches to the choice of alloying elements of brazing filler metals base, providing a solid solution and dispersion strengthening. In particular, the introduction of tantalum and rhenium into brazing filler metals, which are used for alloying of high-temperature alloys of turbines of a new generation and have a higher efficiency in increasing heat resistance and heat strength and reducing the concentration of molybdenum and tungsten or replacing them.

2. At a brazing temperature of 1250 °C, the wetting angles of high-temperature alloy based on Ni_3A1 intermetallics by brazing filler metal SBM-3 do not exceed 7°, as to the chemical composition and structure, the weld metal is close to the base metal, short-term strength at 900 °C is at the level of the base metal, and the long-term strength of TLP-joints on the basis of 50 and 100 h of tests is not lower than 80 % of the base metal.

3. The results of the work showed that the created brazing filler metal SBM-3 and the TLP-bonding technology correspond to the aims of investigations.

4. According to the results of work for experimental industrial use, brazing filler metals of Ni–Co–Cr–A1–Ti–Ta–Re–W–Mo–B system were melted with Hf and without it.

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