Peculiarities of the structural-phase state of a brazing zone of dissimilar metal joints

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Annotation. The present work is devoted to the study of the structural-phase state of the brazed joints of dissimilar materials obtained by rapidly quenched filler metals before and after tests in the working conditions. The constructive elements of the International Thermonuclear Experimental Reactor (ITER) working chamber were selected: mock-ups of the first wall (a beryllium cladding joined with the heat-removing bronze by high-temperature brazing) and the divertor (a tungsten cladding with built-up copper, brazing of copper with bronze). An analysis of the investigated samples and the data on the structural-phase state of the brazed joints and its evolution as a result of a cyclic thermal impact was carried out by metallography and electron microscopy.

1. Introduction
At present, materials with various physical and chemical properties (for example, ceramics and metal, alloys and metals with very different coefficients of thermal expansion, fusible and refractory metals, etc.) are used in many constructive elements. Brazed constructions often work under extreme conditions, which make them more demanding to thermal-, wear- and corrosion-resistance, mechanical strength, and radiation resistance. Damages can occur both at the unsatisfactory quality of the brazed seam in the initial state and at its non-compliance to the acceptance criteria of the existing standards, as well as in the event of defects in the course of operation. Amorphous and nanostructured brazing filler metals, possessing high characteristics of diffusion and capillary activity and a high homogeneity of the chemical composition, make it possible to solve the complex tasks of creating original and workable constructions from various materials. In particular, the cladding of the divertor (tungsten) and the first wall (beryllium) of the International Project ITER (International Thermonuclear Experimental Reactor) assumes brazing of the above-mentioned elements with bronze as this part of the construction is in direct contact with plasma [1]. As the right choice of the brazing filler metal, the knowledge of regularities of the structure- and phase-formation during the brazing, as well as an understanding of the processes occurring during the operation of the brazed joint are the necessary conditions to ensure a long-term operation of the product, an object of the present work was to study the evolution of the structural-phase state of the brazed seam during its operation by methods of the modern metallography.

2. Experimental part
Brazing filler metals based on copper (STEMET 1101 and 1108) were used in the work. Their compositions and characteristics are shown in Table 1. They were in the form of both rapidly-quenched amorphous and nanostructured ribbons, as well in the form of powders obtained by grinding
of a preliminarily embrittled amorphous ribbon. Ribbon-type brazing filler metals were prepared at the installation Crystal-702 that makes it possible to produce such alloys in the form of a ribbon (with a thickness of 25 - 100 μm and a width of 1.5 - 50 mm) by an ultrafast solidification of a planar jet of the melt on a rotating copper disc. The achievable cooling rate of the melt at the installation was regulated in the range of $10^4 - 10^6$ °C/sec. The maximum quantity of the ribbon obtained for one technological cycle was 0.8-1.0 kg for copper alloys.

Table 1. The composition and characteristics of the brazing filler metals.

| Brand of the filler metal | Nominal composition in (wt. %) | $T_{\text{melting}}$ (°C) | $T_{\text{braze}}$ (°C) | Brazing conditions | Brazed materials |
|--------------------------|--------------------------------|--------------------------|-------------------------|--------------------|-----------------|
| STEMET 1101              | 8.8 Ni, 7.0 P, 3.5 Sn, Cu - the rest | 615, 660 | 650, 670 | Gas flame, electrocontact, and induction brazing. | Copper and its alloys. Beryllium - copper alloys. |
| STEMET 1108              | 12 Sn, 5 In, 2 Ni, Cu - the rest | 750 | 870 | Gas flame, induction, electrocontact, and furnace brazing. | Copper, copper alloys, and steels |
|                          | 1 P                                  | 870 | 950 | Brazing in a vacuum or inert atmosphere. | Beryllium - copper alloys, and tungsten. |

3. Objects of the investigation

1. Sample 1. An element of the ITER first wall mock-up
The first wall is a multi-layer construction, of which the water-cooled bronze structure is covered with brazed beryllium tiles with a size of 16x16 mm and a thickness of 8 mm. The operation regime of the ITER reactor is thermocycling. Thermal stresses occur on the "beryllium-bronze" border during the heating cycle. Therefore, thermocycling experiments are the main workability criterion of the beryllium coating. To braze the beryllium tiles, a rapidly quenched amorphous ribbon-type non-silver filler metal STEMET 1101 is used [2]. The brazing of the mock-up is carried out either by an induction method or by heating with an electron beam. To preserve the desired mechanical properties of the temperature-sensitive bronze in each of the above-mentioned methods, it is necessary to minimize the time of heating / cooling of the CuCrZr alloy in the temperature range of 450–700–450 °C (figure 1).

Mock-ups are installed on the stand Cepheus-M and covered with a water-cooled mask.

![A heating-cooling diagram](image_url)
The mock-ups are supplied by cooling water and the beryllium surface, seen from under the masks, is exposed from above to a cyclic heat flow that is simulated by an electron beam. The main parameters of the experiment expected in the ITER reactor are shown in Table 2.

Table 2. The main parameters expected in the ITER reactor.

| Parameter                                      | Value                      |
|------------------------------------------------|----------------------------|
| The absorbed heat power density                | 0.5 – 5.9 MW/m²             |
| The water pressure at the inlet into the mock-up | 2 ±5% MPa                  |
| The water temperature at the inlet into the mock-up | 70 ± 5 °C                  |
| The water consumption through the mock-up      | 0.42±5% kg/s               |
| The water speed                                | 2 ±5% m/s                  |
| The duration of a pulse / pause during the thermocycling experiments | 15/15 s                  |
| The heat loading area                          | 75 cm² (the entire surface of the coating) |

A fragment of the ITER first wall mock-up is shown in Figure 2.

Figure 2. An element of the ITER first wall mock-up.

2. Sample 2. An element of the ITER divertor
The divertor consists of two main parts, a support construction made from stainless steel and a water-cooled part facing to the plasma that is coated by erosion- and heat-resistant tungsten.

Table 3. Working conditions of the protective elements of the ITER divertor from tungsten (the average heat flow is 0.3 MW/m²).

| Action type                                      | Vertical target | Dome | Liner |
|--------------------------------------------------|-----------------|------|-------|
| The number of cycles (without a replacement)     | ~3000           |      |       |
| The maximum heat flow, MW/m²                     | 5               | 3    | 0.2–1 |
| The maximum damage degree, dpa                   | ~0.6            | ~0.7 | ~0.1–0.2 |
| The maximum generation of helium, appm           | < 0.35          | < 0.4| < 0.1 |
| The working temperature, °C                      | 200–600         | 500–1200 | 500–1200 |
| The energy at the plasma disruption, MJ/m²       | 1–5             | 10   | 1     |
| The number (N) / duration of plasma disruptions, ms | 500/1…2        | 500/1…2 | 500/1…2 |
Figure 3 shows a fragment of the ITER divertor. The tungsten is coated by a copper layer with a thickness of 3 mm; the copper is brazed to the bronze that is welded to a heat-removing steel base. For brazing, a filler metal STEMET 1108 was used [2].

Working conditions of the protective elements of the divertor made from tungsten (the average heat flow is 0.3 MW/m²) are presented in Table 3.

4. Metallographic analysis
A metallographic analysis was carried out at the starting stage of the experiment to obtain primary information about the microstructure of the seam and the heat-affected zone, to confirm or refute the presence of structural-phase changes and their qualitative estimation, and to analyze the presence defects in the brazed seam. The images were obtained by a metallographic microscope METAM PB-21 using the Video Test "Structure"-5.2 software package. To reveal the grain boundaries, chemical etching (at the etchant of FeCl₃ + HCl + H₂O) was carried.

4.1. Sample 1. The joint of beryllium with bronze
According to the present investigation, it can be said that the brazed joint of beryllium with bronze has a high quality: pores, cracks and faulty brazed areas are absent; the joint width is the same in all areas; the joint structure is fine and presumably of eutectic composition. The structural-phase changes in the sample after operation in relation to the initial one were not detected (Figure 4).

Figure 4. Microstructure of the brazed seam and the heat-affected zone of the joint of beryllium with bronze (the marker is 100 μm): a) the sample before the tests and after the chemical etching; b) the sample after the tests and after the chemical etching.
4.2. Sample 2. The joint of copper with bronze

It is necessary to note a high content of defects (faulty brazed areas) of up to 100 μm in length in the brazed seam of both the samples. They are mainly observed at the edges of the brazed joint. The quantity of defects and their size is much less in the central zone of the brazed seam (Figure 5).

![Images of the brazed seam and heat-affected zone](image)

Figure 5. Microstructure of the brazed seam and the heat-affected zone of the joint of copper with bronze (the marker is 100 μm): a) the sample before the tests and before the chemical etching (edge); b) the sample before the tests and after the chemical etching (edge); c) the sample after the tests and before the chemical etching (edge); d) the sample after the tests and after the chemical etching (edge); e) the sample after the tests and before the chemical etching (center); f) the sample after the tests and after the chemical etching (center).

The presence of structural changes, such as an increase in the width of the diffusion zone and formation of grains of a new phase, was revealed in the sample exposed to thermal cycling tests.
5. Electron-microscopic investigations, an analysis of state diagrams
Images of the brazed seam microstructure for each of the prepared sections of the brazed joints were obtained at various magnifications by an electron microscope Carl Zeiss EVO-50XVP with EDS Inca x-act in the regime of reflected and secondary electrons. To estimate the distribution of elements, a chemical analysis of the brazing filler metal was carried out after operation of the product; maps of the distribution of elements were made as well as [3].

5.1. An element of the ITER first wall mock-up. Sample 1
The seam microstructure of the sample before the tests (Figure 6) consists of three phases that are equally distributed throughout the whole volume of the seam. Its width varies in a small range and is of the order of 40 ± 2 μm. The beryllium contains implanted particles of the abrasive material, the relief is strong enough, and a step is clearly visible at the junction of the brazing filler metal and beryllium. Large pores and cracks are absent, small pores (less than 10 μm²) are extremely rare. The sample does not contain significant changes after operation (Figure 7), the microstructure is also three-phase, and only a slight increase of the dispersion is visible owing to the thermal treatment. Additional detailed investigations were not carried out due to the lack of structural-phase changes. An analysis of the phase diagrams made it possible to determine phases that are most likely present in the brazed seam structure (Figure 8).

Figure 6. Brazing of beryllium (bottom) with bronze (top). The sample before the operation.

Figure 7. Brazing of beryllium (bottom) with bronze (top). The sample after the operation.

Figure 8. Brazing of beryllium (bottom) with bronze (top). The sample before the operation. 1) α-solid solution based on copper; 2) eutectic of ε and η phases; 3) phase with a maximum content of phosphorus - (37–39) % Mn, (34–36) % Ni, ≈ 24% P).

5.2. An element of the ITER divertor. Sample 2
There is a great number of small pores in the brazed seam both before and after its operation (Figures 9 and 10) which are rounded and elongated along its line, as well as larger pores and faulty brazed areas that reach a length of 40 μm (the area is of the order of 400 μm²) are also present. The seam width of the initial sample varies from 20 to 50 μm, its boundaries are slightly visible due to the fact that the materials of the brazing filler metals and the joined elements have an equal metal base...
(copper), and the seam is non-uniform in phase composition. Its structure is coarse two-phase. The first phase, heavier, is presumably of the eutectic composition with a grain size of 100–700 μm and it is mainly located at the central zone of the seam. The second one is a copper-based solid solution. The brazed seam of the sample after the tests has a more ordered microstructure and a relatively equal width. The dispersion of its structure is significantly increased and there is also a great number of pores and cracks. When a larger magnification is used, it is noticeable that the eutectic phase is distributed in the form of a grid. It is necessary to point out the presence of chemical compounds (phosphides) absent in the initial sample. They are distributed chaotically, have a very various form and are found in the form of both globules (≈ 10 μm) and thin elongated needles (1×50 μm).

![Figure 9. Brazing of copper with bronze. The sample before the operation.](image1)

![Figure 10. Brazing of copper with bronze. The sample after the operation.](image2)

As shown by metallographic analysis, elongated tangential cracks, formed most likely in the process of crystallization of the brazing filler metal, are present in the seams of the edge zones of both the samples. However, the electron image shows that these cracks end at the grain boundaries and do not penetrate into the base material. There is even no growth of the structure imperfection in the product after its operation, which indicates the ability of the brazed joint to withstand the planned loads.

6. Conclusion

Based on the experimental results obtained, the following conclusions can be made:

1. As a result of the tests of details exposed to thermal loads, the brazed joints can experience structural-phase transformations and preserve the initial microstructure without any changes.

2. The seam of the joint of beryllium with bronze, obtained by a copper brazing filler metal STEMET 1101 and possessing a high quality has undergone the minimum structural-phase changes. A small increase of the structure dispersion should be interpreted as the result of the heat treatment without a phase recrystallization.

3. It is necessary to point out the low quality of the initial brazed joint obtained using the brazing filler metal STEMET 1108 (brazing of copper with bronze). Numerous defects in the form of faulty brazed areas, pores and cracks are present in the brazed seam, which to some extent is quite strange as the brazed materials have similar physicochemical properties and, as well as the brazing filler metal, have the same metal base, i.e. copper. The differences in the microstructure of the samples before and after the tests (an increase in the number and the size of defects, the seam width and the dispersion, the allocation of intermetallics and a eutectoid grid) indicate an intensive diffusion of elements of the brazing filler metal and the brazed materials. The formation of chemical compounds of the Mn-Ni-P system having a high brittleness is a negative factor, which in turn may negatively affect the mechanical properties of the brazed joint and lead to failure of the constructive element.
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