Brazed metal-ceramic components for space applications

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Brazed metal-ceramic components play an important role in the manufacturing of specialized scientific instruments for space applications, where a compact, lightweight, and robust construction is always needed. Soldering and brazing are the best joining techniques if dissimilar materials are involved. For the current ESA mission JUICE, an optimized ion source main structure consisting of a stack of concentric metal electrodes and ceramic rings was to be brazed in vacuum. In the improved design one-piece electrodes manufactured from a molybdenum-copper composite were used having the same coefficient of thermal expansion as alumina. Different coating systems were evaluated for the composite material. The effectiveness of TiAlN as diffusion barrier for gold could be demonstrated in a first brazing test between the coated Mo85Cu15 and metallized Al_2O_3 parts joined with Incusil® ABA^{TM}. Several main structures satisfying all technical requirements were successfully brazed.

1 Introduction

Brazed metal-ceramic components play an important role in the manufacturing of specialized scientific instruments such as the time-of-flight mass spectrometers developed by University of Bern, which were successfully used in different space missions (Rosetta [1-3], BepiColombo [4] and Luna [5]). A compact, lightweight and robust construction is always needed for space applications, so that soldering and brazing (which add only little extra weight) are the best joining techniques if dissimilar materials are involved.

Direct brazing of bulk materials is not a promising option: the use of coatings with specified chemical composition combined with dedicated heat treatments is the key. Metal-ceramic joining with pre-coated parts generally results in better wettability (as realized for MCP detector [6]), better leak tightness, mechanical performance and also longevity.

In May 2022, the European Space Agency (ESA) will launch the JUICE mission (Jupiter icy moons explorer) to study Jupiter and its three icy moons Europe, Ganymede and Callisto [7]. Mass spectrometry of the exospheres of the Galilean moons at high relative velocity will be performed [8]. The Neutral gas and Ion mass spectrometer (NIM), which is part of the Particle Environment Package (PEP) [9], is developed and manufactured by the Physics Institute of University of Bern together with Swiss industry and Empa. The NIM instrument is able to measure in different operation modes (neutral, ion and thermal mode) as was demonstrated with a prototype [10]. For such measurements a more complex ion source is needed compared to former missions.

The JUICE spacecraft will be exposed to high radiation during its three-year long science phase at Jupiter. Shielding will be used only partially to mitigate the high radiation dosages. The design of the NIM instrument (Fig. 1) is thus based on thick-film resistor technology, which is more or less directly exposed to radiation. The radiation hardness of thick-film resistors was successfully tested with 18 MeV protons [11].

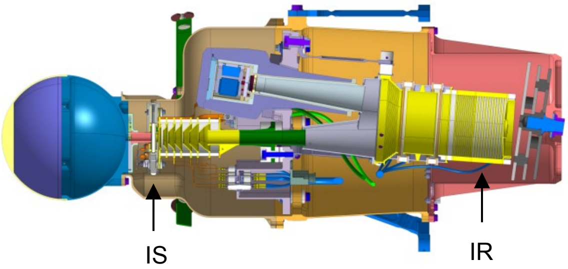


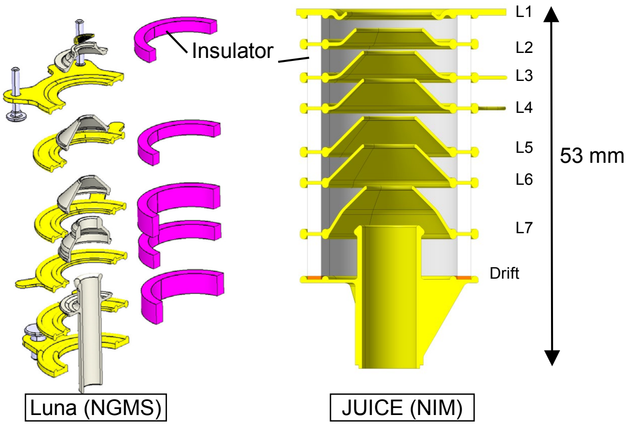
Figure 1. Schematic drawing of the NIM instrument (length 364 mm), showing the ion source (IS) and integrated reflectron (IR), for which thick-film technology is used.

The combination of these materials with ceramic substrates is a key technological element for the realization of a lightweight instrument that is compliant to both the performance and the instrument qualification requirements. Throughout the last two decades, many skills for metal-ceramic brazing and coatings for space applications were developed and optimized at Empa. While some of these procedures could be re-used for the current JUICE mission, others had to be modified with regard to increased demands on radiation hardness and performance of the NIM instrument. For example, manufacturing of the ion source body (ISB) of the NIM instrument involved a combination of dedicated coating, heat treatment and brazing steps. The ISB is a high-accuracy brazed construction consisting of a stack of concentric metal electrodes and insulating ceramic rings. The concentric electrodes have to be gold-plated mainly for best electrical conductivity of the surfaces. In former space projects vacuum brazing was performed for niobium and alumina components (Table 1 and Fig. 2) having similar coefficients of thermal expansion

($\Delta\alpha < 10^{-6}$/K). Because machining of complex niobium parts is challenging and the surface quality is insufficient for the ion-optical application, the electrodes were manufactured from two different materials (molybdenum or titanium alloy), which were then welded into the niobium brazing rings before brazing. In contrast, for the current JUICE mission the electrodes were machined as one-piece from the composite material molybdenum-copper (Mo85Cu15), which has a coefficient of thermal expansion similar to alumina. This not only required adjustments of formerly developed brazing processes, but also the application of a new coating system.

Table 1. Material combinations of the ion source body of previous space missions and the current JUICE mission

Mission Instrument (Launch)		Electrode (+coating)	segments	Insulator	Filler metal for brazing
Rosetta RTOF (02.03.2004)		Nb ring Mo *welded* (Au)	5	Al_2O_3 97.6% AL300 MET Wesgo (ring)	Incusil ABA Wesgo (foil)
BepiColombo STROFIO (16.10.2018)		Ti6Al4V (Ni/Au)	3	Si_3N_4 (spacer)	(clamped)
Luna NGMS *(2022/2023)*		Nb ring Ti6Al4V *welded* (TiAlN / Au)	5	Al_2O_3 97.6% AL300 MET Wesgo (ring)	Incusil ABA Wesgo (foil)
JUICE (NIM) *(2022/2023)*		Mo85Cu15 (TiAlN / Au)	7	Al_2O_3 97.6% AL300 MET Wesgo (ring)	Incusil ABA Wesgo (foil)
		Ti6Al4V (TiAlN / Au)	1		



Two-part electrodes
(Nb + Ti6Al4V)

One-piece electrodes
(7 x Mo85Cu15 / 1 x Ti6Al4V)

Figure 2. Design of the ion source main structure (body) for the previous Rosetta and Luna mission (left) and the current JUICE mission (right). Compared to the former design used for Rosetta and Luna no welding step is necessary for manufacturing of the electrodes: In the improved design of the current mission, the electrodes (lenses L1 – L7) are machined in one piece from Mo85Cu15 and can directly be brazed to the alumina insulators because of similar coefficients of thermal expansion.

2 Materials and experimental procedure

2.1 Molybdenum-copper composite components and coating systems

Pure molybdenum would be an optimal electrode material with regard to its high electrical conductivity and chemical inertness towards gold coatings. However, the CTE mismatch ($\Delta\alpha_{Mo/Al2O3} > 2\cdot10^{-6}$/K) and thus the thermal stresses induced by the Mo/alumina brazing would be higher compared to the previously used Nb/alumina brazing. For the Mo-Cu-composite material, the coefficient of thermal expansion can be designed via the ratio of Mo and Cu to match that of alumina. There are two approaches possible for the production of Mo-Cu composites [12]: manufacturing of laminated copper/molybdenum/copper (CMC) sheet and production of Mo-Cu composites from powders by either solid-state sintering or liquid infiltration. In the present case, the electrodes were fabricated from porous molybdenum that was vacuum-infiltrated with molten copper. The electrodes were machined from bars of Mo85Cu15 with Ø40 mm x 250 mm (lens elements L2 – L7, Figs. 2 and 3) and with Ø52 mm x 100 mm (lens L1), respectively. The bars were purchased from AT & M Refractory Materials & Ceramics Branch, Beijing (www.atmcn.com).

To evaluate different coatings and brazing procedures, as well as the mechanical performance of the base material and the braze joints, several additional tests were performed. For these tests, samples were produced by electric discharge machining (EDM):

- Ø40 mm x 1 mm for coating experiments
- small substrates for brazing and shear tests (20 mm x 10 mm / 4 mm x 4 mm; thickness 1 mm)

Tensile specimens according to DIN 50125 – A 8 x 40 were manufactured from the Ø40 rod by EDM and turning.

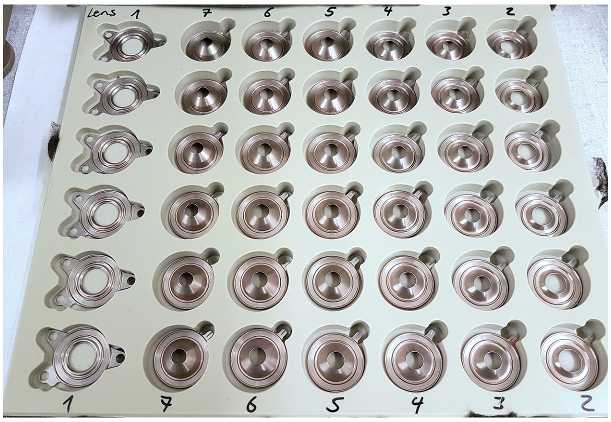


Figure 3. Electrodes for the NIM ion source machined from Mo85Cu15.

Different types of coating systems were evaluated for application onto the Mo85Cu15 substrates (see Fig. 4, option 1 – 4). Due to the required specification of gold on the top of the electrodes, the lens elements L1 – L7 had to be coated with a diffusion barrier. Direct gold plating (e.g. 2 µm Au-PVD with an adhesion layer; Fig. 4, option 1) is not possible due to the high chemical affinity between Cu and Au, even though the composite contains 85 wt.-% molybdenum.

In the present case, a TiAlN-based coating (Balinit® Futura Nano) was used as diffusion barrier. This coating can be applied for copper, low-carbon steels, titanium alloys (cf. electrodes for Luna NGMS, Table 1) and molybdenum. TiAlN thin films are used as protective layer in various fields as these films have a high thermal stability and offer higher hardness, oxidation and wear resistance in comparison with TiN. The violet-grey coating shows a microhardness of 3300 (HV 0.05) and can be employed up to 900 °C [13, 14]. Tungsten was used as an additional interlayer between TiAlN and the final Cr/Au metallization, selected due to preliminary coating tests (Fig. 4).

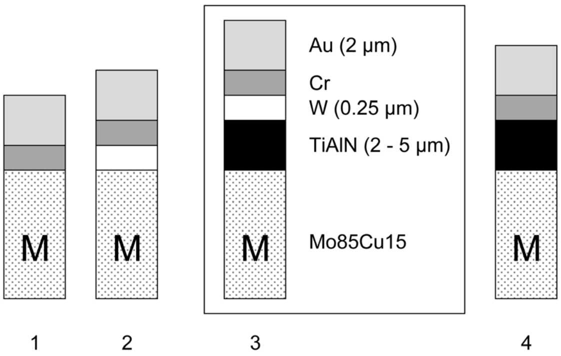


Figure 4. Different coating systems evaluated for the ion source electrodes (M) with the final option 3 (TiAlN + W + Cr/Au).

2.2 Alumina components

For the alumina components of the ISB, 97.6% Al_2O_3 rings metallized with Mo/Mn (13 µm - 38 µm; Wesgo AL 300 metallization) with an additional Ni plating (2.5 µm – 6 µm) were used.

2.3 Heat treatments and brazing

Conventional cleaning procedures were applied during subsequent processing steps. After cleaning, all Mo85Cu15 parts (lenses and test specimens) were annealed in vacuum furnaces (Cambridge Vacuum Engineering, model 1218H; SCHMETZ type E80/1H) at 850 °C/2 h. After application of the TiAlN diffusion barrier coating, the components were heat-treated at 500 °C/3 h. After the final gold coating (W + Cr/Au), a dummy brazing process was applied (750 °C/10 min., $p < 5 \cdot 10^{-4}$ Pa) to test the adhesion and the barrier stability for gold.

Wetting experiments were performed with Wesgo-Incusil® ABA™ on uncoated and coated specimens. Incusil® ABA™ was selected due to the good experiences with this filler material in former space missions (Table 1). A preliminary brazing test (750 °C/10 min., $p < 5{\cdot}10^{-4}$ Pa) was performed with the coated flight parts (Fig. 5). The specimens for the additional shear tests were prepared following the same procedure. To estimate the effect of the various brazing treatments on the Mo85Cu15 composite material, some tensile specimens were annealed at 850 °C for 2 h.

Figure 5. First brazing test (AL 300 MET (Ø24 x Ø20 x 4) + Incusil ABA + Mo85Cu15 (TiAlN/W/Cr+Au)).

Finally, the ion source body (Fig. 2) – consisting of 31 parts – was brazed at 750 °C using a custom-designed brazing jig which guarantees an accurate mechanical alignment of the concentric electrodes (Fig. 6).

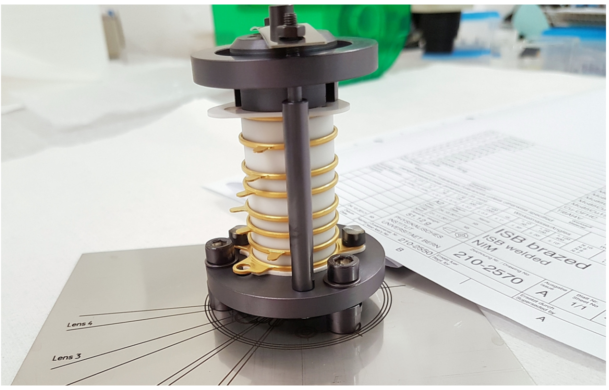


Figure 6. ISB assembled in the brazing jig just before the brazing step.

2.4 Characterization of the Mo85Cu15 composite and joints

The composite material and the joints manufactured in the first brazing tests were evaluated by scanning electron microscopy and energy-dispersive x-ray spectroscopy (SEM-EDX, Fei NanoSem230).

Shear testing according to EN 15340 (STM series 5 shear tester/ 4 kN, Walter+Bai AG) was done for small test specimens. Tensile tests were performed with a SHIMADZU AGS-X 100 kN tester.

3 Results and discussions

3.1 Microstructure

The cross-section of the Mo-Cu composite material used for the electrodes (Fig. 7) shows that the material consists of a matrix of relatively large molybdenum particles, with an average particle size of about 10 µm (see Fig. 7b) densely filled with copper. This microstructure indicates that liquid copper was introduced into a sintered Mo skeleton (or that a compacted Mo-Cu powder was heat-treated above the melting point of Cu). In the SEM cross-section image no voids were observed. It is not known if after the copper infiltration forging and/or heat treatment was performed to further densify the material. The copper is evenly distributed, which is advantageous for the mechanical properties of the material. However, the large open Cu surfaces and channels (up to 20 µm) can act like a sponge for the gold plating as applied to the electrode material. Hence, an effective diffusion barrier between copper and the gold plating is necessary (cf. Section 2.1).

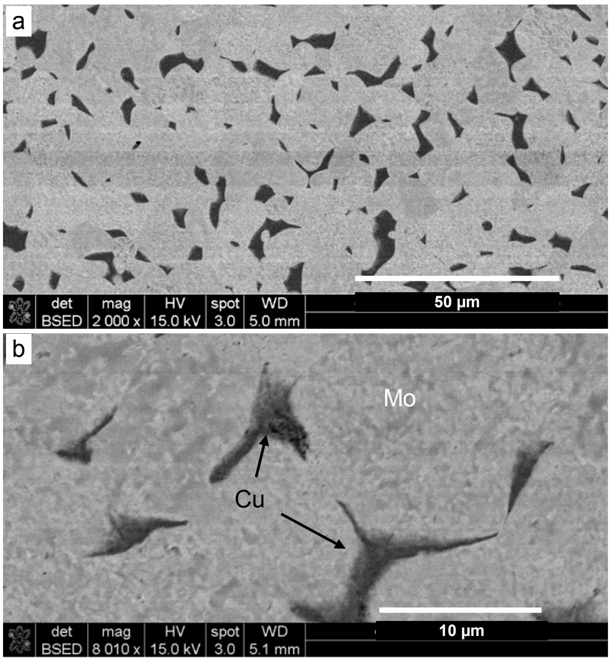


Figure 7. SEM cross-sectional image of the Mo85Cu15 composite: same material batch as used for NIM ion source lenses.

The effectiveness of TiAlN as a diffusion barrier could be shown in the first brazing test between the coated Mo85Cu15 parts and metallized Al_2O_3 parts joined with Incusil® ABA^{TM} (Fig. 4: Mo-Cu coating configuration 3, Fig. 5: brazing configuration, and Fig. 8: SEM cross-sectional images). There are three regions of interest: ① the metallization zone of Mo85Cu15 (MET 1), ② the brazing filler zone, and ③ the metallization zone (MET 2) of Al_2O_3 (Fig. 8b). During joining (750 °C/10 min.) the gold metallization of the Mo85Cu15 composite dissolved into the brazing alloy: silver- and copper-rich regions with various gold content could be detected throughout the brazing zone (Fig. 8c). However, the underlying TiAlN and W coating layers remain at the original Mo85Cu15 surface, and no gold diffusion into Mo85Cu15 was observed, i.e. the TiAlN layer fulfills its purpose as an efficient diffusion barrier for Au (Fig. 9). In addition, Ni-Ti phases distributed throughout the entire brazing zone can be found (Figs. 8c and 8d). These phases are formed from Ni coming from the Wesgo AL 300 metallization of the Al_2O_3 and the small Ti content of the Incusil ABA brazing alloy. Towards the Al_2O_3 base material, the intertwined structure of Mo and glassy components of the Wesgo AL 300 metallization (MET 2) can be found, which realizes a good mechanical bonding between the metallic braze filler and the ceramic.

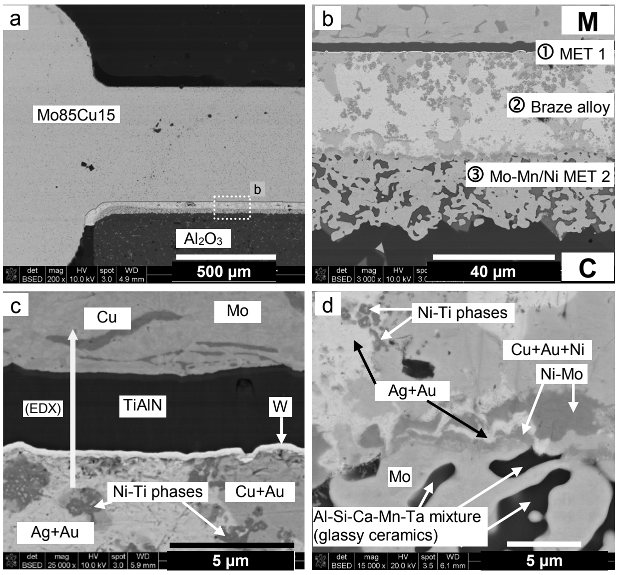


Figure 8. SEM cross-sectional images of the first brazing test sample.

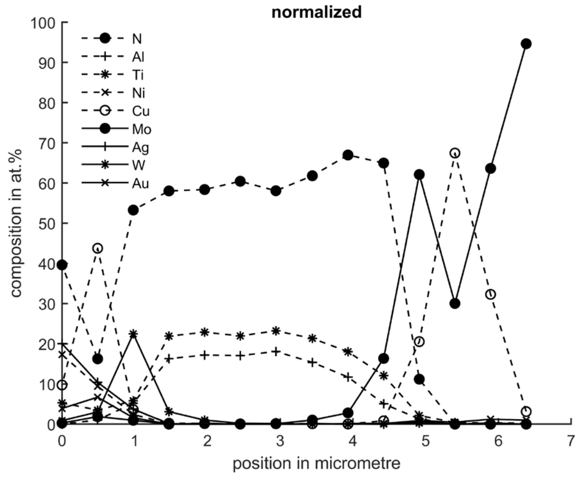


Figure 9. EDX linescan (*l* = 7 µm) from brazing alloy into Mo85Cu15 (Fig. 8c)

3.2 Mechanical tests

Annealing has a strong effect on the mechanical behaviour of the Mo85Cu15 composite material. After annealing, the material has an ultimate tensile strength (UTS) of 496 ± 5 MPa with a three times higher strain at failure compared to the as-received material (47 % versus 16 %, Table 2). The ductile behaviour of this composite could also be observed during shear testing of brazed substrates (deformation of sheared parts). The mean shear strength of uncoated metal-metal joints was 222 ± 14 MPa (*n* = 10). Shear tests for other material and coating combinations were also performed, as well (not shown here).

Table 2. Tensile tests of Mo85Cu15 (DIN 50125 – A 8 x 40).

Sample	State	UTS MPa	Strain at failure %
1_1	as-received	302	14.8
1_2		455	16.6
1_3		452	16.8
1_4	annealed 850 °C/2 h	499	46.9
1_5		498	46.3
1_6		490	47.3

3.3 Ion source body brazed and wired

In total three ion source bodies (ISB-1 to ISB-3) were successfully brazed between April and July 2018 (Fig. 10a). The 7 electrodes (Mo85Cu15) and one drift tube (Ti6Al4V) are separated by alumina insulators with thicknesses between 4 and 7 mm. Any brazing failure – e.g. small droplets, twisted or blocked electrodes – would severely deteriorate the performance of the ion source. Although 15 brazing foils (total thickness 0.75 mm) were used, the geometrical deviations were small (e.g. variations of 0.03 mm for the height, measured at three points at 120° apart). Several functional tests like partial discharge tests (ΔU_{max} = 10.5 kV) or thermal cycling were applied to test the performance of the ISB. The current-voltage characteristic was better in comparison to the former designs used for Rosetta and Luna (Table 1). This increase in performance can be attributed to the use of one-piece electrodes and the optimized material selection (the gold-plated Mo85Cu15 is more conductive than the gold-plated Ti6Al4V).

ISB-1 was used for the functional tests; the other two ion source bodies were wired by brazing with Incusil ABA paste (Fig. 10b). This technique is much better (and easier to perform) than welding of copper wires, although fusing to a brazed body can be risky. Also after the second brazing step the gold coating did not diffuse into the electrode material and the geometry of the body remained unchanged. The multilayer coating is probably more affected by the repeated brazing cycles, especially the tungsten interlayer with a much lower CTE compared to the brazing alloy.

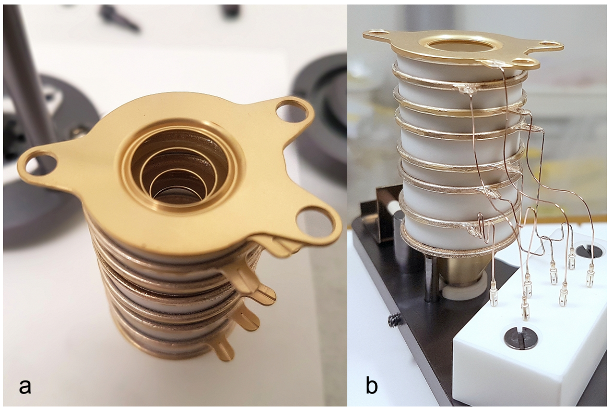


Figure 10. a) brazed ISB-1: m = 52 g / l = 53 mm / $Ø_{max}$ 50 mm; b) wired ISB-2: copper wires (Ø0.3 mm) were brazed to electrodes and plugs using the same filler metal (Incusil ABA).

4 Conclusion

The composite material Mo85Cu15 was for the first time used as electrode material for the ion source main structure of the NIM instrument of JUICE, consisting of a stack of concentric metal electrodes and alumina insulators. Uncoated material (metal electrodes and ceramic insulators) could be vacuum-brazed with the active brazing alloy Incusil® ABA™. However, due to the instrument specifications (gold on top of the electrodes and dielectric strength of the ceramic) metallized parts had to be used. TiAlN / W / Cr+Au proved to be a suitable coating system for Mo85Cu15 substrates: TiAlN successfully fulfilled its purpose as an efficient diffusion barrier for Au. The adhesion between W and TiAlN may be a weak point of the multilayer. Currently tensile tests are performed to disclose the effect of the W interlayer on the obtained joint strength.

Two ISB were manufactured and then wired by brazing, i.e. two subsequent brazing steps were performed. The multiple fusing steps were not detrimental to the mechanical accuracy of the whole ISB structure.

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