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Brazing transparent YAG to Ti6Al4V: reactivity and characterization

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ABSTRACT

The YAG/filler/Ti6Al4V system has been studied for the first time with the aim of producing brazed optical windows. Different fillers (AgCuTi, AgCu, Ag) and temperatures have been taken into account and the interfacial zones of the samples have been evaluated in terms of morphology and compositions. A thin and continuous metal-ceramic layer containing Ti is formed in contact with the YAG ensuring the adhesion between all the joined materials. The best joining results have been obtained at 850 °C, where we have observed the formation of an interfacial CuTi layer when AgCuTi and Ag have been used. No intermetallic compound formation was observed using the pure Ag at 970 °C and 1050 °C. The phase formation was interpreted using the CALPHAD method by means of recalculated Ag-Cu-Ti and Ag-Al-Ti isothermal sections. Microhardness tests have been conducted to evaluate the performances of the joints in terms of mechanical properties.

1. Introduction

This investigation aims to define a brazing process that leads to reliable joints between a transparent ceramic (Yttrium-Aluminium garnet-YAG) and a structural metal by defining and characterizing a filler alloy, which is capable of promoting interactions and good adhesion between the two different materials to be joined.

Brazing is a flexible and simple process that allows materials to be joined using a metallic interlayer, which has a relatively low melting temperature and promotes the bonding by wetting and reacting with the metal and the ceramic part. In the field of marine applications, new ceramic materials are requested to be used as transparent windows (e.g. for laser sensors). For this reason, the capability of the ceramic part to maintain its transparency after a joining process and during its immersion in the marine water represents a necessary requirement. Many other fields need these particular materials; atomic physics and frequency metrology applications, for instance, require nonmagnetic optical windows for ultra-high vacuum chambers; in this context sapphire and titanium joints were obtained by Ag-Cu-Ti brazes [1]. Jacobs et al. have analyzed the silica/PbAg/304 stainless steel interfaces, a material potentially required for various plasma diagnostic systems in the ITER vacuum vessel [2].

Gustarov has investigated how an ITER simulated environment can influence the mechanical stability and the optical characteristics of prototype windows assemblies in which Si_3N_4, silica and sapphire are the transmission elements bonded or brazed to a stainless steel or Ti ferrule. They found that the bonds at ceramic-metal interfaces remained leak-tight after irradiation and thermal cycling [3].

A thorough investigation on transparent and gas-tight optical windows in low-temperature co-fired ceramics (LTCC) was conducted three years ago [4]. Thin glasses (borosilicate glasses and fused silica) were bonded by thermo-compression in an LTCC substrate in order to obtain materials used as optical analysis systems, for innovative applications such as observing chemiluminescent reactions. Laser welding techniques for glass–glass and glass-metal joining have recently been considered, including hermetic sealing applications [5].

In this paper, transparent ceramic windows made of sintered YAG ($Y_3Al_5O_{12}$) coupled to Ti6Al4V supports have been studied as potential candidates in the marine applications mentioned before. This is due to their transparency, good mechanical properties and to the outstanding corrosion resistance of the individual components. Indeed, Ti6Al4V exhibits an exceptional erosion-corrosion resistance [6–9], a high fatigue strength in air [10–12], as well as in chloride environments [13] and a low coefficient of thermal expansion. This improves the interface compatibility with the ceramic counterpart. Generally, the filler alloys used for joining dissimilar materials contain an active element such as Ta, Nb, Zr, V or especially Ti [14–17] that promotes wetting of the ceramic

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surface and chemical bonding with the metallic member of the joint by adsorbing or reacting with the ceramic oxide.

In this work, a comparison was performed between joints produced using filler alloys with or without an active element in order to capture the differences in terms of interface reactivity and to find some possible suggestions to be adopted in the potential industrial process.

Different types of metal-ceramic joints YAG/filler/Ti6Al4V (filler = AgCuTi, AgCu, Ag) have been produced under inert atmosphere conditions (Ar), at high temperatures (between 850 °C and 1050 °C), and subsequently assessed in terms of interfacial reactivity by wetting tests and EDS analysis, and in terms of mechanical properties by micro-hardness test.

The wettability and the reactivity of the YAG substrate by the filler alloys AgCuTi, AgCu and pure Ag were previously analyzed by this Group and the results reported in [18]. These results showed excellent wettability and interfacial reactivity for the YAG/AgCuTi systems tested at both 850 °C and 950 °C. Indeed, the contact angles for these pairs were significantly below the limit of 90° and two different continuous metal-ceramic layers without defects and pores were observed along the whole contact zone. The microchemistry and the microstructure of new interfacial products represent a crucial point that has to be carefully controlled in order to obtain the desired mechanical performances. Indeed, excessive reactions can create undesirable brittle intermetallic phases compromising the technological quality of the joint. In addition, it is also necessary to consider that the production process of the filler alloys suggests limiting the concentration of the active metals in the brazes for economic (and sometimes, strategic) reasons. In these particular systems, like in several other metal/ceramic ones, the presence of Ti is necessary to wet the YAG surface. We were able to demonstrate that the formation of two metal-ceramic phases at the interfacial zone allows a new interfacial layer to be obtained in contact with the alloy drop and also well wetted by it [18].

Thus, in order to avoid the use of the active element in the filler material with a consequent simplification of the process, YAG/Ag/Ti6Al4V and YAG/AgCu/Ti6Al4V pairs were also tested to demonstrate that the spontaneous Ti migration from Ti6Al4V bulk to the interfacial zone is sufficient to promote the wettability and the adhesion of the metal-ceramic components. Indeed, the Ti6Al4V counterpart could act as an active source of Ti for the brazing alloy.

2. Experimental part

2.1. Ceramic materials

The ceramic substrates were prepared by CNR-ISTEC following the experimental procedure of Esposito et al. [19,20], starting from extremely pure (≥99.99%) and sub-micrometric ceramic oxide powders of Al_2O_3 and of Y_2O_3 sintered at 1735 °C for 16 h under high vacuum conditions (10^{-4} Pa).

Before the brazing process, the substrates (transparency of about 80% [21]) were polished on diamond grinding discs to reach a final surface roughness (S_a) below the value of 1.00 μm, measured by an optical confocal-interferometric profilometer (Sensofar S-neox) on an area typically about 3 × 3 mm^2.

2.2. Metals and alloys

Foils of CB4-AgCuTi alloy (Degussa-Germany), AgCu eutectic (99.9% purity, Metalli Preziosi-Milano, Italy) and Ag (99.997%, Goodfellow, Cambridge, UK), used as filler materials in the joining tests, were laminated in order to reach a thickness of 100 μm and cleaned in an ethanol ultrasonic bath. The CB4-AgCuTi alloy has a density of 9.9 g cm^{-3}, a thermal expansion coefficient equal to 18 × 10^{-6}/K and a melting range of 780–805 °C [22]. It is considered as a medium-melting-point active filler (melting point between 700 °C and 1000 °C [23,24]).

Table 1
Compositions of AgCuTi and AgCu alloys utilized as filler (at%).

	Ag	Cu	Ti
AgCuTi	57.7	36.8	5.5
AgCu	60.2	39.8	–

The compositions of the tested alloys are reported in Table 1.

The metallic support Ti6Al4V (grade 5 Ti, Titalia, Milano, I), at a composition of 6 wt% Al and 4 wt% V (10 at% Al, 3.5 at% V), was cut to obtain parallelepipeds of 1 cm length and polished on diamond grinding discs to reach a final surface roughness S_a « 1 μm. The YAG/filler/Ti6Al4V sandwiches were brazed after assembling the pieces using a graphite clamp.

2.3. Procedure

The joining experiments were performed in a specially designed furnace that can reach 1600 °C [25]; the precision of the temperature readings can be estimated to be ±5 °C.

The oxygen partial pressure P_{O2} in the working atmosphere (Ar with less than 0.5 ppm gaseous impurities, flowing at 50 cm^3/min) was continuously monitored by solid state oxygen sensors at the chamber inlet and outlet. A P_{O2} of about 10^{-2} Pa (measured at the gas outlet) was measured and different temperatures were chosen (from 850 °C to 1050 °C), depending on the constitution of the filler. The samples in the sandwich configuration were then introduced into the preheated furnace by a magnetically operated push rod, as soon as temperature and P_{O2} reached their pre-set values. The joining samples were tested at different temperatures (YAG/AgCuTi/Ti6Al4V at 850 and 950 °C, YAG/AgCu/Ti6Al4V at 850 °C; YAG/Ag/Ti6Al4V at 970 and 1050 °C).

All the specimens were maintained at the selected temperature for 10 min, cooled at a rate of 5 °C/min and finally extracted from the furnace only when room temperature was reached.

The treated samples were then embedded in epoxy resin, cut for morphological observation after polishing down to 1 μm grit and cleaning in an ethanol ultrasonic bath.

Accurately polished micrographic sections of our samples were observed by scanning electron microscopy (SEM, model: LEO 1450 VP) with the aim to detect the morphological features, and analyzed by electron microprobe (EDS) (Oxford Instruments, 7353 model with Oxford-INCA software v. 4.07, type of detector: Si(Li)). EDS analyses were performed on at least five areas for each sample to determine the local composition. The following parameters were used: – working distance of 15 mm, – live time of 60 s, – acceleration voltage of 20 kV, – Co as standard element. By using these parameters, the detection limit is 0.1 wt%, while the precision is 1 wt%.

Microhardness of the joints was measured on cross-sectioned samples using a Vickers indenter, with a load of 0.01 kgf (0.098 N) and a dwell time of 12 s.

3. Results and discussion

One general result that should be underlined is that all the samples tested during the work maintain the same crucial and necessary required characteristics of an excellent grade of transparency.

In the following paragraphs the interfacial microstructure of the different types of the metal-ceramic joints will be accurately analyzed and presented.