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Laser surface nanostructuring for reliable Si_3N_4/Si_3N_4 and Si_3N_4/Invar joined components

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ABSTRACT

IR pulsed laser radiation in air was applied to Si_3N_4 and Invar to obtain reliable Si_3N_4/Si_3N_4 and Si_3N_4/Invar adhesive bonded components. The laser pre-treatment produced a homogeneous nanostructured oxide layer on the surfaces, which effectively increased the adhesion at the adhesive/adherends interface and led to cohesive failure in the joining material. The mechanical strength of Si_3N_4/ Si_3N_4 and Si_3N_4/Invar joined components was measured, with and without laser nanostructuring, before and after thermal cycling from room temperature to 50 K, and it resulted that the exposure to extremely low temperatures did not affect the mechanical integrity of the joints. It was also demonstrated that this laser pre-treatment did not alter the mechanical properties of the ceramic substrate.

1. Introduction

Future large optical instruments in multisatellite missions will require ultra-high stable structures (some square meters), and a near to zero coefficient of thermal expansion (CTE) is necessary for both in-orbit and ground-and-orbit operations [1]. Silicon nitride has a low CTE over a wide range of temperatures and exhibits high stiffness and dimensional stability under moisture [2]. Moreover, Invar® is an Fe-Ni alloy with a 36% nickel content that exhibits the lowest expansion of known metals and alloys from the lowest temperatures up to approximately 200 °C [3]. For these reasons, silicon nitride and Invar alloy have been selected as key materials for highly stable and/or controlled expansion components in optical and structural aerospace systems.

One of the most critical issues for the use of ceramic-based components is that just a few joining methods are presently available and most of them require the use of a high temperature and pressure. Moreover, a low-temperature pressure-less joining process in air can facilitate the fabrication and repair of large onsite structures under a wide variety of field assembly conditions. In that sense, the application of adhesive bonding is increasing for similar or dissimilar joints [4] and adhesive bonding offers many advantages over mechanical fastening for the assembling of primary structures for aerospace applications, but requires robust materials and processing methodologies.

Surface pre-treatments are widely recognized as one of the key steps to producing robust and reliable bonds [5,6]. Generally, before similar or dissimilar adhesive bonding, ceramics must be accurately polished by means of an expensive lapping process, which significantly reduces the flexibility of the component design. Moreover, the adhesion on metals, such as Invar, can be achieved through the use of a primer [7], surface roughening obtained through sandblasting or chemical etching, or a UV laser surface treatment [8]. Previous studies, carried out by Airbus Defence and Space GmbH, have demonstrated that laser radiation can be used as an alternative and effective new technology for the surface pre-treatment of metals and several ceramics before bonding [9,10]. When the laser treatment of the surface leads to the formation of a nanostructured layer, an increase in the bonding performance has always been achieved, except for silicon carbide, where the formation of a graphite layer has been found to be responsible for the non-sufficient adhesion properties of the laser modified SiC surfaces [11]. The main advantages of the laser surface pre-treatment have been discussed in Suess et al. [11]; currently, this is considered the best process to obtain highly reproducible surface quality and reliable joints without employing environmentally harmful and toxic substances, thus meeting the EU REACh (Registration, Evaluation, Authorisation and Restriction of Chemicals) regulations [12].

The laser treatment of Si_3N_4 ceramics has also been proposed as an alternative to conventional surface finishing and machining processes [13]. CO_2 laser surface processing has been studied to reduce the grinding-induced defects on the surface of Si_3N_4 and it has been demonstrated that the flexural strength of Si_3N_4 increases slightly after the laser treatment, due to the improvement of the surface and sub-surface integrity induced by laser treatments [14]. On the contrary,

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Shulka et al. [15] have observed a reduction of the surface hardness when a laser treatment has been performed on as-sintered silicon nitride.

In the past, the formation of conical structures on Si_3N_4 surfaces by means of laser ablation before adhesive bonding was proposed by Man et al. [16]. The presence of the micro-cones provides a higher adhesion surface area and mechanical locking sites, thus improving the adhesion strength. Nevertheless, in this case, the effect of this micro-patterning on the mechanical strength of the ceramic materials was not investigated.

In the present paper, IR laser radiation in air was applied to Si_3N_4 and Invar to obtain reliable Si_3N_4/ Si_3N_4 and Si_3N_4/Invar adhesive bonded components. The laser pre-treatment produced a homogeneous nanostructured oxide layer on the surfaces, which effectively increased the adhesion at the adhesive/adherends interface and led to the cohesive failure in the adhesive. The mechanical strength of the Si_3N_4/ Si_3N_4 and Si_3N_4/Invar joined components was measured, with and without laser nanostructuring, before and after thermal cycling from room temperature to 50 K. Additionally, it was demonstrated that this laser pre-treatment did not affect the mechanical properties of the ceramic substrate.

2. Materials and methods

The silicon nitride used in this study was SN-GP produced by FCT Ingenieurkeramik GmbH (Germany). It is a polycrystalline β-Si_3N_4 obtained by gas pressure sintering using 3–10 wt% of sintering additives (RE_2O_3 / Al_2O_3) [17]. Its CTE is 1.9×10^{-6} °C^{-1} between room temperature and 250 °C. The chemical composition and properties of SN-GP are reported in [18]. As a reference pre-treatment for the adhesive bonding, Si3N4 surfaces were lapped to obtain a flatness of about 0.5 µm.

The Invar® M93 FeNi36 alloy was produced by Aperam Imphy (France). This 36% nickel-iron alloy exhibits a CTE of 1.5×10^{-6} °C^{-1} between –180 and 100 °C [19].

The laser surface nanostructuring was performed on Si_3N_4 with as-fired surface quality and Invar with a milled surface quality of Ra 0.35 µm. The treatment was carried out with the PowerLine E Air 25 laser system from Rofin (Germany) in air. This original designed laser system is a nanosecond pulsed Neodymium doped Yttrium-Orthovanadate laser (Nd:YVO4) with a wavelength λ of 1064 nm. The laser system has an average supply power of 25 W and allows a pulse frequency from continuous wave (cw) up to 200 kHz. During the present study, a wide parameter optimisation was executed over the entire power intensity spectrum of the laser system. The discussed parameters are representative of the achieved results.

Si_3N_4 ($25 \times 25 \times 5$ mm^3) with lapped, which is here referred to as "reference", and laser treated surface qualities were bonded to obtain similar Si_3N_4/ Si_3N_4 and dissimilar Si_3N_4/Invar joints using Hysol® EA9321 (Henkel Corporation, USA), a two-component thixotropic paste adhesive, which was cured at room temperature for at least 7 days, according to the supplier's specifications [20].

Laser structured Si_3N_4 and Invar, as well as joined specimens, were characterised using Scanning Electron Microscopy (SEM) with an Energy Dispersive Spectroscopy (EDS) detector (AURIGA FIB-SEM Instrument, Carl Zeiss Microscopy GmbH, Germany) and X-ray Photoelectron Spectroscopy (XPS) (Quantum 2000 Scanning ESCA Microprobe, Physical Electronics GmbH, Germany). The SEM analysis for the lateral view of some laser treated Si_3N_4 was performed on fractured surfaces, achieved by cutting with a diamond wire saw (Modell 6234, well Diamantdrahtsägen GmbH, Germany) and the sample was then broken up after cooling in liquid nitrogen.

The mechanical strength of Si_3N_4, after the laser treatment, was evaluated by means of the co-axial ring in ring test according to DIN EN1288-5 [21,22] on 6 samples. Furthermore, lapped and laser structured Si_3N_4 surfaces were quantitatively analysed by the confocal technique using a 3D non-contact profilometer (Sensofar S-neox). Quantitative measurements were performed using the software embedded in the system (SensoSCAN, Sensofar Metrology, Terrassa, Spain).

Some joined samples were cooled under vacuum in a cryocooler from room temperature to 50 K in 5 h, 1 h dwelling time at 50 K, then were heated up to room temperature in 20 h. This cryo-cycling is representative of the environmental conditions found in satellites. The samples were mounted, three at a time, in contact with the second stage of a cryogen free cryocooler. The temperature was measured with two thermometers, one fixed on the second stage of the cryocooler (cryostat temperature) and the other one on the top side of the middle sample (sample temperature). In order to improve the thermal contact a thin indium foil was placed between the lowest sample and the cryocooler stage and between the samples. The good thermal contact between the samples and the cryostat is proved by the very small temperature difference measured by means of the two thermometers. After the first thermal cycle, the samples were dismounted for a preliminary optical check and, then, submitted to further four consecutive thermal cycles in order to achieve five cryo-cycles.

The mechanical strength (apparent shear strength) of the Si_3N_4/ Si_3N_4 and Si_3N_4/Invar joined samples was determined using a single lap offset (SLO) test in compression at room temperature adapted from ASTM D905-08 (universal testing machine SINTEC D/10) [23] before and after cryo-cycling. The test configuration and the dimensions of the samples are reported in [11]; the samples were bonded with a joining area of 12.5×25 mm^2 and a bonding gap thickness of 0.15 mm (Fig. 1). For each joint 3 samples were tested. The load was applied by moving the cross-head at a speed of 1 mm/min. The maximum force was recorded and the apparent shear strength was calculated by dividing the maximum force by the joining area. All the fracture surfaces were macroscopically characterised.

3. Results and discussion

3.1. Laser surface nanostructuring of Si_3N_4 and Invar alloy

Fig. 2a and b-d show the surface morphology of as-fired and laser treated Si_3N_4, respectively.

As-fired Si_3N_4 top view (Fig. 2a) shows a microstructure with homogeneous nitride grain size higher than 2 µm in length; a residual porosity is observable among the grains.

The morphology of the as-fired Si_3N_4 surface, after laser treatment, consisted of a homogeneous open porous nanostructure with

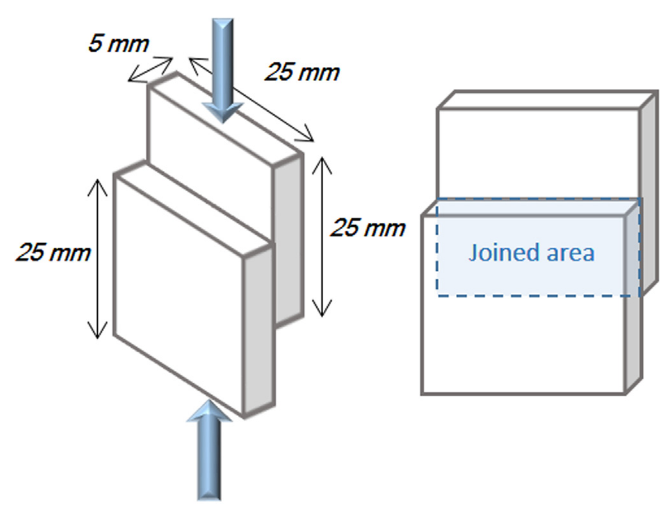

Fig. 1. Geometry of the joined specimens for the single lap offset test in compression.