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Original
A plasma pre-treatment to improve adhesion on SiC and Si3N4 ceramics / Casalegno, Valentina; Ferraris, Monica; Perero, Sergio; Suess, Manuela; Wilhelmi, Christian; Pedroni, Matteo; Vassallo, Espedito; Salvo, Milena. - In: MATERIALS LETTERS. - ISSN 0167-577X. - STAMPA. - 272:(2020), p. 127855. [10.1016/j.matlet.2020.127855]

Availability:
This version is available at: 11583/2817349 since: 2020-04-28T15:48:19Z

Publisher:
Elsevier

Published
DOI:10.1016/j.matlet.2020.127855

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http://dx.doi.org/10.1016/j.matlet.2020.127855

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16 April 2024

A Plasma pre-treatment to improve adhesion on SiC and Si_3N_4 ceramics

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Abstract

The design of interfaces, coupled with suitable joining materials and joining technologies, is a key parameter in component manufacturing and surface pre-treatments are crucial steps for the production of robust and reliable bonds. In this work, plasma pre-treatments on SiC and Si_3N_4 are studied and discussed. The pre-treated joined samples have shown a higher mechanical strength than the reference value (lapped surfaces) and the increased adhesion at the ceramic/adhesive interface have led to cohesive failure of the adhesive.

Keywords: Ceramics; Surfaces; Joining

1. INTRODUCTION

Because of their outstanding thermomechanical properties and stability over certain temperature ranges e.g. from ambient down to nearly zero K, SiC and Si_3N_4 ceramics have attracted a great deal of interest in the satellite instrument field [1-3]. The availability of highly reliable ceramic joints, combined with structural robustness and extremely lightweight designs, is still a challenging engineering task. In this context, the use of adhesive bonding is increasing for similar and dissimilar joints; it offers many advantages, compared to mechanical fastening (e.g. more uniform stress distribution, reduced weight and cost [4]), but requires effective processing methodologies. Surface pre-treatments are often used to functionalize a surface prior to gluing, and they may result in bonds which show a marked improvement in adhesion [5]. This surface functionalization relies on a combination of a few or of all of the following effects: fine surface cleaning, modification of the surface topography, changes in the crystalline structure of the surface layer, and

activation of the chemical groups [6]. Other studies 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 [7-12]. A review on the current status of metal and ceramic surface treatments can be found in ref. 13.

In previous works, the authors have shown and discussed the relationship between the surface morphology and the adhesion properties of laser pre-treated SiC and Si_3N_4 [9, 10]: for SiC, the formation of a graphite nano-layer resulted in unsatisfactory joints properties, while robust and reliable joints (with cohesive failure) were obtained with laser pre-treated Si_3N_4. The aim of this work is to show that joined samples with similar mechanical properties can be achieved with an alternative method for both ceramics, SiC and Si_3N_4.

In the present work, SiC and Si_3N_4 substrates have been treated by means of a dry plasma reactive ion etching process (RIE) in order to increase the mechanical strength of their adhesive joints. The process has been realized in a low density capacitively-coupled plasma reactor in a CF_4/H_2 gas mixture. The mechanical properties of plasma pre-treated joints have been measured and compared with the results obtained from different pre-treatments (lapping and laser treatment), already discussed in refs. 9, 10.

2. EXPERIMENTAL

The silicon carbide used in this study was Boostec®SiC produced by Mersen (France). It is a polycrystalline α-SiC (> 98.5 wt% SiC) obtained by means of pressureless sintering. 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 means of gas pressure sintering, using 3-10 wt% of sintering additives (RE_2O_3/Al_2O_3). As a reference pre-treatment, the SiC and Si_3N_4 surfaces were lapped to obtain a flatness < 5 µm.

The plasma treatment on SiC and Si_3N_4 was carried out by means of an RF plasma system [14]. The following optimized parameters were used: for SiC, power 200 W; pressure 10 Pa; CF_4 flow 18 sccm and H_2 gas flow 2 sccm; treatment time 30 min; for Si_3N_4, power 300 W; pressure 20 Pa, CF_4 flow 69 sccm and H_2 rate 6.9 sccm; treatment time 30 min.

An Invar® M93 FeNi36 alloy, produced by Aperam Imphy (France), was used for the dissimilar joint test. Laser surface nanostructuring was performed on the as-received Invar in air with a nanosecond pulsed

Nd:YVO4 laser (λ= 1064 nm), using a PowerLine E Air 25 laser system from Rofin (Germany); average supply power of 25 W and pulse frequency up to 200 kHz [9, 10].

The plasma treated SiC and Si_3N_4 were characterized using Field Emission Scanning Electron Microscopy (FESEM- ZEISS Supra 40) with an Energy Dispersive Spectroscopy (EDS- SW9100 EDAX) detector.

The plasma treated SiC and Si_3N_4 (25x25x5 mm^3) were bonded to obtain similar (SiC joined to SiC) and dissimilar (Si_3N_4 joined to Invar) joints using Hysol® EA9321 (Henkel Corporation, USA), cured at room temperature for at least 7 days, according to the supplier's specifications. The apparent shear strength of the joined samples was determined using a single lap offset (SLO) test, under compression, adapted from ASTM D905-08, at room temperature (SINTEC D/10 universal testing machine) [10]. The samples were bonded with a joining area of 12.5 x 25 mm^2 and a bonding gap thickness of 0.15 mm; cross-head speed of 1 mm/min. Three samples were tested for each joint.

3. RESULTS AND DISCUSSION

Figures 1a and 1b-c show the surface morphology of the as-fired and plasma treated SiC, respectively. The as-fired SiC (Figure 1a) shows a residual porosity that was estimated to be about 2% in volume [10]. The morphology of the SiC surface, after the plasma treatment, consisted of a homogeneous round-shaped microstructure (Figure 1b-d) without any visible pores. The EDS analysis on the plasma treated surface did not reveal the presence of any different elements from Si and C (C 53 at%; Si 47 at%).

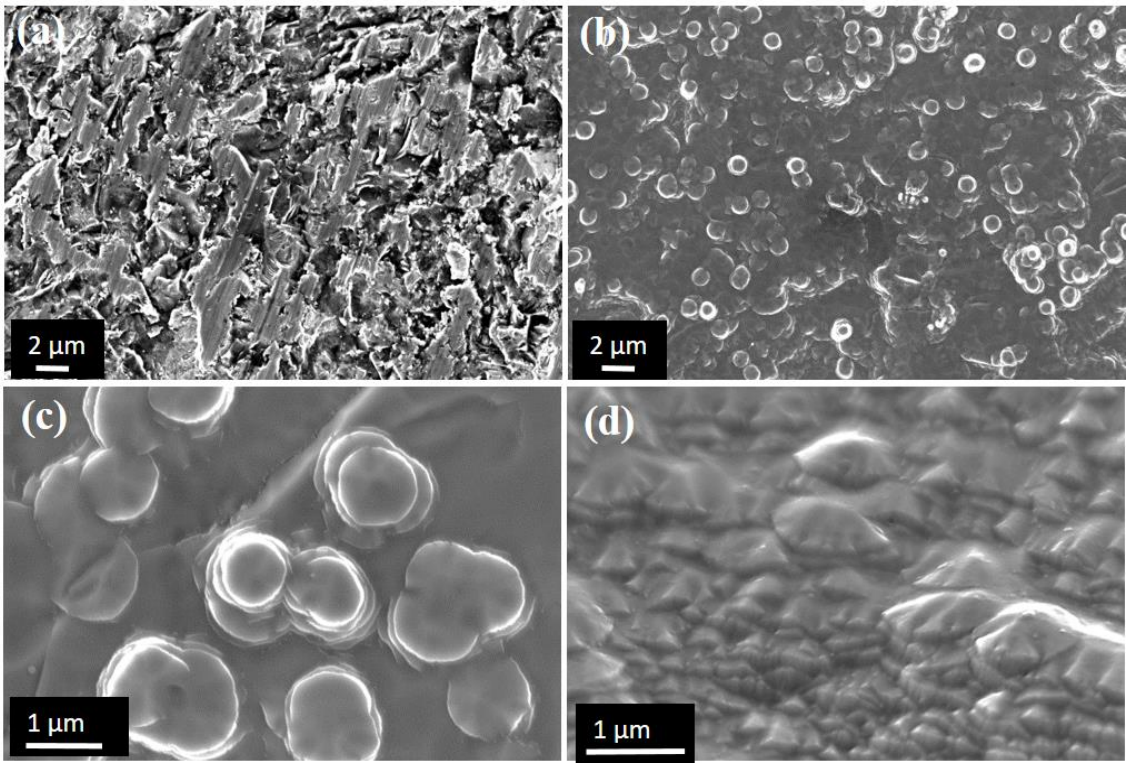

Figure 1. SEM micrographs, top view of (a) as-fired and (b, c) as-fired SiC after the plasma treatment. (d) tilted view of the plasma treated SiC surface.

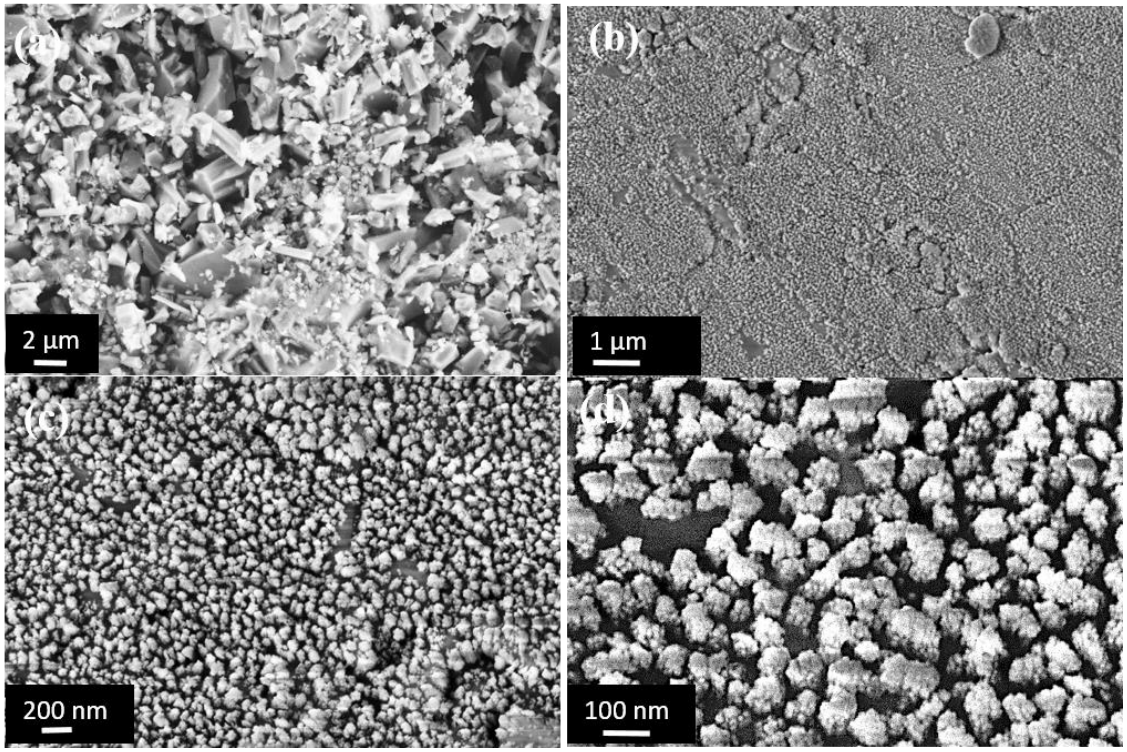

Figure 2. SEM micrographs, top view of (a) as-fired and (b-d) as-fired Si_3N_4 after the plasma treatment.

Figures 2a and 2b-d show the surface morphology of the as-fired and plasma treated Si_3N_4, respectively. Again in this case, the as-fired Si_3N_4 (Figure 2a) shows residual porosity among the grains, while the morphology of the plasma treated Si_3N_4 surface consists of homogeneously distributed agglomerates of spherical nanoparticles (Figure 2b-d). The EDS analysis (semi-quantitative analysis; EDS analysis precision of ±2% for the main components) on the as-fired Si_3N_4 surface showed the presence of 25.9 at% C, 26.9 at% N, 35.6 at% Si, 8.8 at% O, 2.1 at% Al and traces of Ti and Mg; Al, Mg, Ti and O are elements that are included in the sintering aids of Si_3N_4 and C can be attributed to adventitious C (carbon contamination originating from the environment). Furthermore, the plasma treated surface showed the presence of 53.5 at% N, 27.0 at% Si, 18.0 at% O and 1.5 at% Al: the absence of C the absence of C on the treated surface could demonstrate that the plasma treatment had a cleaning effect and led to a significant reduction in surface contamination; a similar effect was observed by the authors when Si_3N_4 surfaces were pre-treated by means of a pulsed laser in air [9].

During the plasma treatment, the formation of a non-thermal discharge supplies neutral chemically active etchants (F atoms), energetic ions and inhibitor precursor radicals (CF_2 and CF_3). The radicals deposited on the substrate are able to form fluorocarbon polymer films, which protect the substrate from further and excessive etching. However, a part of the protective layer is removed and perforated by a simultaneous, biased ion bombardment. The competition between polymer film deposition and its removal, as a result of the bombardment of energetic ions, causes local variations of the etch rate on the substrate surface, which in turn leads to the growth of random micro and nano-scaled features. The aim of this work has been to investigate whether the obtained surface morphology could create an interlocking structure at the ceramic/adhesive interface, thereby improving the mechanical properties of adhesively bonded samples, and whether it could lead to cohesive failure within the adhesive.

The single lap offset (SLO) test was used, under compression, to evaluate the adhesion properties of the SiC and Si_3N_4 surfaces plasma treated with Hysol® EA9321. These mechanical strength results were compared with those obtained from different pre-treatments (lapping and laser treatment) used in previous works [9, 10]: lapped SiC joined to lapped SiC, laser treated SiC joined to laser treated SiC, lapped Si_3N_4 joined to

laser treated Invar and laser treated Si_3N_4 joined to laser treated Invar joints; the apparent shear strength values of the joined samples are summarized in Table 1.

Table I. Apparent shear strength values of the joined samples.

Joints	Apparent shear strength (MPa)	Failure mode
Lapped SiC/SiC	41.6 ± 0.9[10]	Adhesive
Laser SiC/SiC	34.8 ± 3.4[10]	Adhesive
Plasma SiC/SiC	**44.3 ± 2.4**	**Cohesive**
Lapped Si_3N_4/Invar	38.9 ± 4.3[9]	Mainly adhesive on Si_3N_4
Laser Si_3N_4/Invar	42.7 ± 1.1[9]	Mainly Cohesive
Plasma Si_3N_4/Invar	**42.7± 1.9**	**Cohesive**

Both the lapped and laser treated SiC-SiC joints showed complete adhesive failure, which is generally caused by an inadequate surface quality of the ceramic substrate. Furthermore, as discussed in ref. 10, the laser treatment on SiC resulted in a lower level of adhesion at the adhesive/ceramic interface, due to the formation of a thin graphite layer on the SiC surface. On the contrary, the plasma treatment on the SiC surface effectively increased the adhesion at the interface: the plasma pre-treated SiC joined samples failed cohesively within the Hysol® EA9321 (Figure 3a) and showed a mechanical strength of 44.3 ± 2.4 MPa (Table I), which is higher than that measured in a previous work [10] for lapped or laser treated SiC joined and tested in the same way.

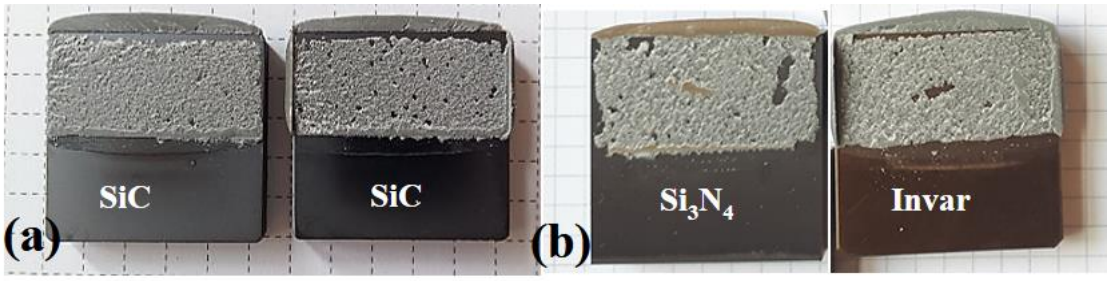

Figure 3. Fracture surfaces of (a) plasma treated SiC/Hysol® EA9321/plasma treated SiC (cohesive failure ~95%) and (b) plasma treated Si_3N_4/Hysol® EA9321/laser treated Invar joints (cohesive failure ~90%) after a single lap offset (SLO) test.

Furthermore, predominantly adhesive failure was observed for the lapped Si_3N_4 joined to Invar (Table I) at the Hysol® EA9321/Si_3N_4 interface. In ref. 9, the optimization of the Si_3N_4 surface, by means of a suitable laser treatment, led to the formation of an effective interlocking structure at the Hysol® EA9321/Si_3N_4 interface, which in turn increased the adhesion and resulted in cohesive failure within the adhesive (τ~ 42.7 ± 1.1 MPa, Table I). The same mechanical performances were obtained for the plasma treated joints, i.e. cohesive failure (Figure 3b) and apparent shear strength of 42.7± 1.9 MPa, respectively. Both the plasma and laser pre-treatments produced a homogeneous micro- and nanostructured layer on the Si_3N_4 surface, which may be infiltrated by the adhesive and thus lead to an improvement in the interfacial strength, an excellent bond strength, and cohesive failure of the adhesive as a result of lap-shear under compression.

4. CONCLUSIONS

The considered plasma treatment has been found to be a suitable, non-expensive and user-friendly process for the surface pre-treatment of SiC and Si_3N_4. The plasma treatment has proved to be effective in increasing adhesion at the ceramic/adhesive interface, offers the possibility of treating large and complex areas (~ 1 m^2) with a high degree of automation, and can also easily be used for complex-shaped geometries.

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