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High temperature compressive mechanical behavior of joined biomorphic silicon carbide ceramics

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Abstract

Silicon carbide-based, environmentally conscious, biomorphic ceramics were fabricated by the reactive infiltration of molten silicon into porous, carbonaceous preforms derived from pyrolysis of African Bubinga wood. The bulk microstructure and high temperature mechanical properties of these ceramics were studied. These biomorphic ceramics mimic the fibrous microstructure of the wood resulting in high strength and anisotropy. The compressive strength parallel to fiber direction, which is the growth direction of the tree, was 750 MPa at 1100 °C and 300 MPa at 1350 °C. The compressive strength perpendicular to fiber direction was 215 MPa at 1100 °C and 120 MPa at 1350 °C. These materials were joined using the ARCJoinT approach. The microstructure of the joints was studied by scanning electron microscopy and the high temperature strength was measured in compression, with the joint oriented 45° to the compression axis. The joined specimens had strengths from 615 MPa at 1100 °C to 250 MPa at 1350 °C when the fibers were parallel to the compression axis (forming 45° with the joint plane), which are about 20% lower than the strength of the bulk material in the same orientation. The strengths ranged from 373 MPa at 1100 °C to 175 MPa at 1350 °C when the fibers were forming 45° with the compression axis (perpendicular to the joint plane), which are lower than the average strength of the bulk material compressed axially and in the perpendicular direction.

Keywords: Biomorphic; High temperature; Joining; SiC; Strength

1. Introduction

Environmentally conscious ceramics (Ecoceramics) are a new class of materials, which can be produced with renewable resources (wood) and wood wastes (wood sawdust). The wood sawdusts are generated in abundant quantities by sawmills. On the other hand, natural woods of various types are available throughout the world. Wood has been known to be one of the best and most intricate engineering materials created by nature and known to mankind.[1–4] The environmentally conscious, biomorphic ceramic materials, fabricated via the pyrolysis and infiltration of natural wood-derived preforms, have tailorable properties with numerous potential applications. The experimental studies conducted to date on the development of materials based on biologically derived structures indicate that these materials behave like ceramic materials manufactured by conventional approaches. These structures have been shown to be quite useful in producing porous or dense materials having various microstructures and compositions.[5–15]

Microstructure and mechanical properties of a wide variety of wood specimens have been investigated and reported in other publications.[10–15] The African Bubinga wood is from the Leguminosae family of woods and has other common names as Essingang (Cameroon), Ovang, Kevazingo (Gabon), and Waka (Zaire).[16] The wood species of this family are found in equatorial Africa from Nigeria through Cameroon to the Congo region. It is found near rivers and lake shores and swampy inundated forests. The tree height is approximately 40–50 m and typically trunk diameters are 1–2 m. It is heartwood pink or red brown wood with a fine and even texture, with straight or interlocked grains, and with a density of 0.65–0.78 gm/cm^3.[16]

The ARCJoinT approach[17] used here is unique in terms of producing joints with tailorable microstructure and properties. Fabrication of joints by this approach is

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attractive since the thermo mechanical properties of the joint interlayer can be tailored to be very close to those of the silicon carbide based materials. In addition, high temperature fixturing is not needed to hold the parts at the infiltration temperature. A wide variety of silicon carbide-based ceramics (reaction bonded, sintered, CVD) and fiber-reinforced composites have been joined using this approach.[17–27] The high temperature mechanical properties of joints in these systems have been studied by tensile, flexure, and compression tests. However, these joints are expected to encounter various types of stress conditions and mixed stress modes.

In the present work, SiC-based biomorphic ceramics have been fabricated by the molten silicon infiltration of porous carbonaceous preforms derived from the pyrolysis of African Bubinga wood. These ceramics have also been joined using the ARCJoinT approach.[17] It is much more economical to build up complex shapes by joining together geometrically simple shapes. However, the joints must have good mechanical strength and environmental stability comparable to the bulk materials. In addition, the joining technique should be practical and reliable.

In this paper, the microstructure and compressive mechanical properties of reaction formed joints in biomorphic SiC are presented. The high temperature compressive strength of these joints (scarf butt geometry) has been measured up to 1400 °C in air. The test specimen geometry used requires a small amount of material and the stress and strain can be directly measured. The compressive strength of joints has been compared to that of bulk material.

2. Experimental procedure

The biomorphic silicon carbide ceramics used in this study were fabricated by the reactive infiltration of molten silicon into a porous preform of carbonized wood (African Bubinga). The infiltration was done in vacuum following the conventional procedure for fabrication of reaction formed SiC. The final product was a cellular structure of SiC with elongated "channels" and fiber-like structures along the axial direction of the original wood.

Experimental details of the joining process have been given in other publications.[17–24] The joining steps include the application of a carbonaceous mixture in the joint area and curing at 110–120 °C for 10–20 min. Silicon in the paste form is applied in the joint region and heated up to 1425 °C for 5–10 min. The molten silicon reacts with carbon to form silicon carbide with controllable amounts of silicon. Two orientations were chosen to join the biomorphic SiC: with the axial direction of the original wood perpendicular to the joint plane (joints type I), and with the axial direction of the original wood forming 45° with the joint plane (joints type II).

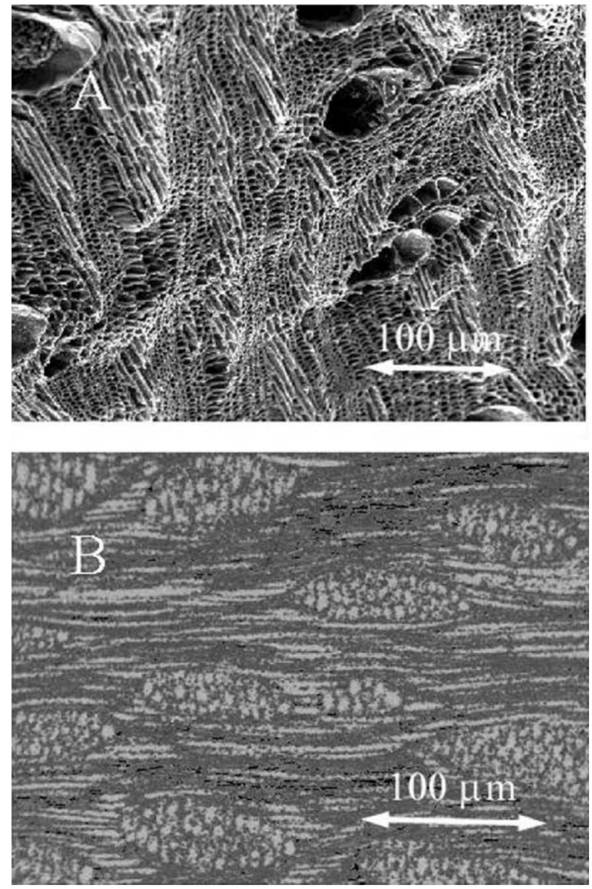

Fig. 1. SEM micrographs of: (A) porous carbon preform; and (B) the as-fabricated biomorphic SiC (silicon regions have white contrast and silicon carbide regions have gray contrast).

As-fabricated and joined samples were cut and polished for metallographic studies. Microstructural characterization was carried out using scanning electron microscopy (SEM).[1] Parallelepipeds of $3 \times 3 \times 5$ mm were tested in compression in air at a constant strain rate of 2×10^{-5} s^{-1} in the 1100 °C to 1400 °C temperature range. Testing was performed using a screw driven Instron universal testing machine model 1185 with a furnace mounted on its frame. The as-fabricated biomorphic SiC was tested parallel and perpendicularly to the axial direction. The joints (type I and II) were tested using samples with the joint forming an angle of 45° with the compression axis.

3. Results and discussion

The microstructure of the porous carbon preform and as-fabricated materials is shown in Fig. 1A and B respectively. The axial direction of the original wood goes from

[1] Electron Microscopy Centre, University of Seville.

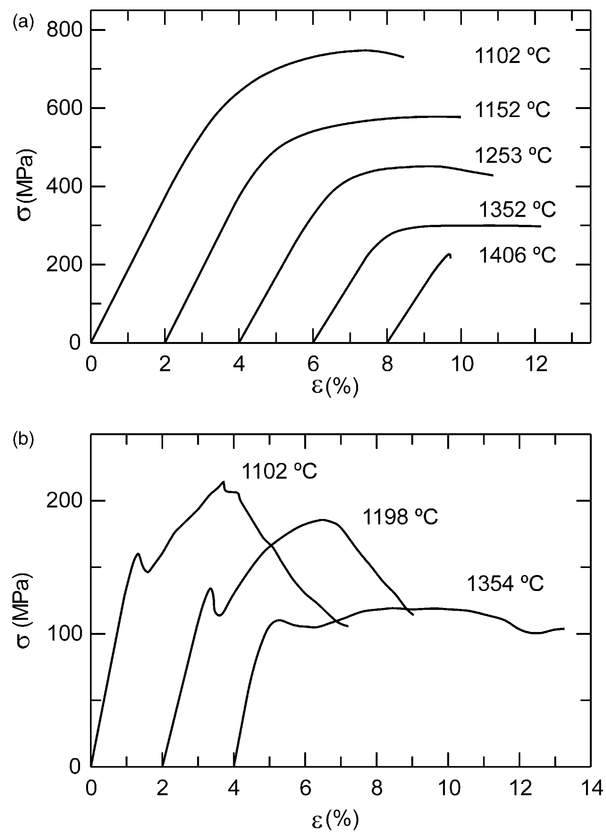

Fig. 2. Plot of stress versus strain as a function of temperature for the as-fabricated biomorphic SiC. (a) Compression parallel to the axial direction, (b) compression perpendicular to the axial direction. The curves are displaced on strain axis for clarity.

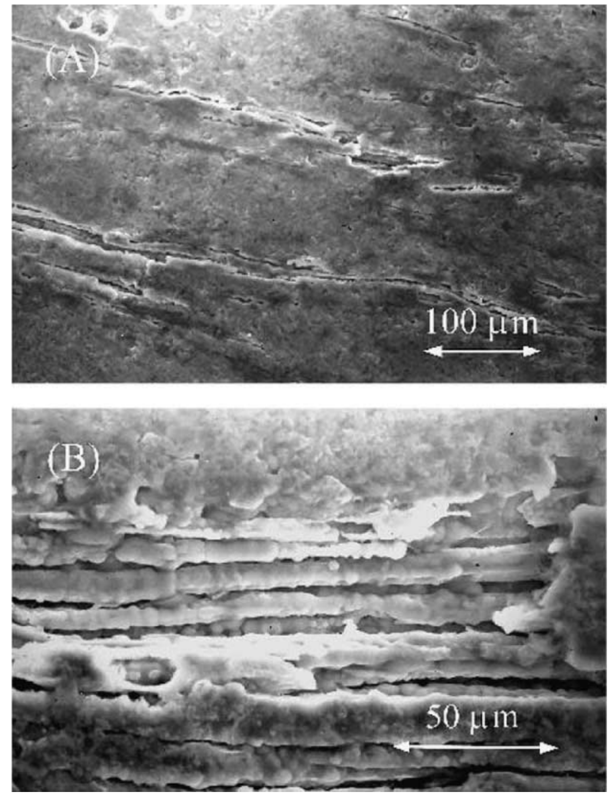

Fig. 3. SEM micrograph of the as-fabricated biomorphic SiC compressed axially (left-right direction in the micrographs). (A) Cracks parallel to the compression direction. (B) Detail of the fiber-like structures.

left to right on the micrograph (Fig. 1B). The white areas are silicon and the gray areas silicon carbide. The unreacted silicon filled up the former salvia channels of the wood. Fig. 1B also shows clearly other families of channels perpendicular to the micrographs, which corresponds to the radial ''rays'' of the original wood.

Due to their unique microstructure, Ecoceramics have anisotropic mechanical behavior. The stress versus strain plots for the compression experiments on the bulk material are shown in Fig. 2a and b. The compressive strength in the axial direction ranges from 750 to 230 MPa for temperatures from 1100 to 1400 °C, with the stress–strain curve showing a flat region after plastic yield. When the compression is applied perpendicularly to the axial direction the strength is between 3 and 4 times lower, and the stress–strain curve is not purely monotonic, showing a region of hardening after the plastic yield.

The strength anisotropy can be noticed in the microstructure of the samples after deformation. When the compression is applied in the axial direction the samples do not have a significant change of shape (the deformation is barrel-like). Fig. 3A shows typical cracks parallel to the compression direction that can be seen on the sample surface. Fig. 3B shows a detail of a crack where it can be noticed that the silicon carbide has a fiber-like structure, and how the plastic deformation of the material is accompanied by the deformation and bending of these fibers. The origin of this microstructure is related to the formation of silicon carbide through the former channels of the wood, resulting in a very strong bonding of the silicon carbide within a channel and a weaker bonding between different channels. These silicon carbide ''fibers'' will enhance the strength and toughness of the material by crack bridging. The strength is considerably lower when the stress is applied perpendicularly to the axial direction, since it is controlled by the bonding between silicon carbide filaments. The continuous debonding between filaments cause the stress–strain curve to be rocky with hardening possibly associated with the crushing of the material between filaments. The resulting microstructure consists of sliding SiC fibers as indicated in Fig. 4A (fibers perpendicular to micrograph) and B (fibers parallel to micrograph). Details of the microstructure are shown in Fig. 4C and D. The microstructure observed explains the different plastic behavior: in one case (axial compression) the material deformation is controlled by pure plastic yielding/bending of the fibers, and, in the other case (traverse compression), the deformation is controlled by damage accumulation/debonding between the fibers.

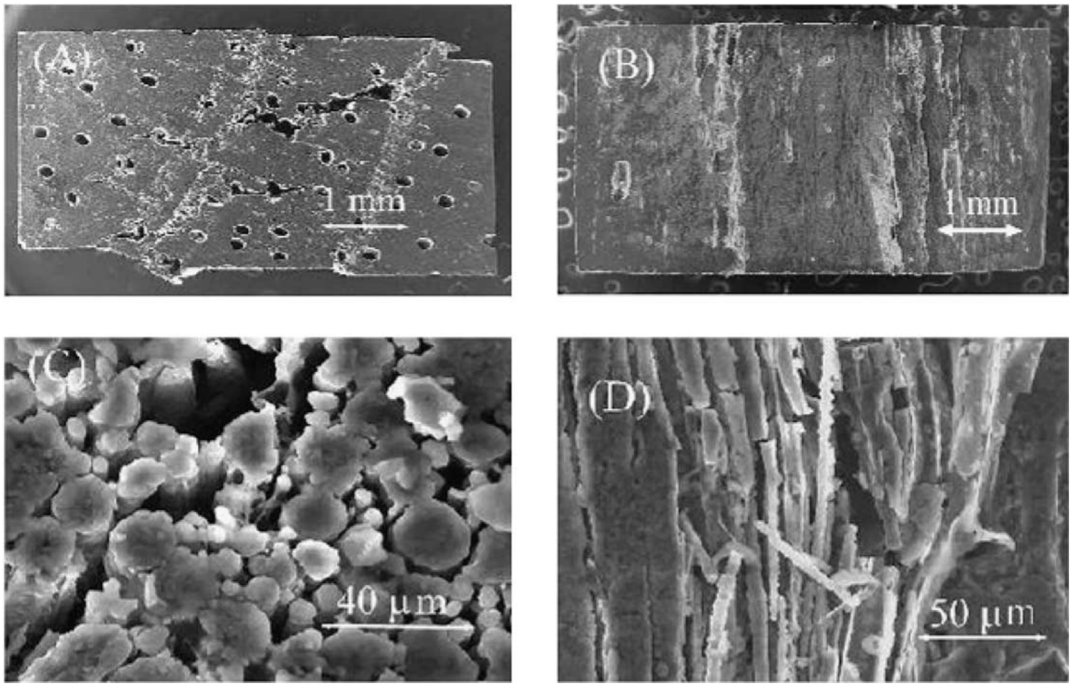

Fig. 4. SEM micrograph of the as-fabricated biomorphic SiC compressed perpendicularly to the axial direction (left-right direction in the micrographs): (A) and (B) general view of the sample faces perpendicular and parallel to the fibers; (C) and (D) details of (A) and (B) respectively.

The microstructure of the joint is shown in Fig. 5. The thickness of the joint ranges from 35 to 60 μm and it has a gray contrast indicating that it is silicon carbide rich. White silicon regions next to the bulk material are also observed. The origin of these regions is not well understood but could be associated with the use of too low pressure or could originate from the biomorphic SiC as the joining temperatures are over the melting point of silicon.

The stress–strain curves obtained in the constant strain rate compressive experiments of the joints are shown in Fig. 6. Type I joints (Fig. 6a, see drawing inserted) can undergo significant plastic deformation (the total strain prior to failure is between 6 and 10%). The strength (taken as the maximum stress held by the sample) decreases with temperature, although, it is about 120 MPa at 1399 °C, which is very close to the melting point of silicon (1410 °C). The axial direction of the bulk material in these joints is oriented at 45° to the compression axis, so the bulk strength in this orientation must have a value between the hard (axial) and soft (radial) orientations. We can estimate the strength of the bulk taking the average between these two values. As it can be observed in Fig. 7 the strength of the joint is under this average value indicating that the joint is probably weaker than the bulk material. Fig. 6b shows the stress–strain curve for type II joints (see drawing inserted on Fig. 6b). The strength of type II joints is higher than for the previous set of joints but lower than the bulk material in the same orientation (axial orientation, Fig. 7). The strain to failure of type II joints is around 2% for all temperatures, lower than for type I joints.

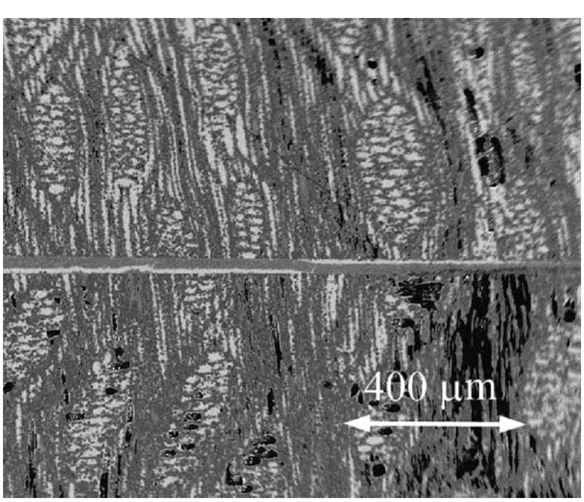

Fig. 5. SEM micrograph of an as-fabricated joint (type I).

After testing, samples were examined by SEM. The deformation in the samples with a joint occurred mainly by shear in the joint plane at all temperatures for both type of joints (Fig. 8). In a type I joint deformed at 1100 °C, open cracks can clearly be seen along the joint (Fig. 8A). A closer look at the fracture surface shows that the joint fails in one or two of the joint-bulk interfaces (Fig. 8B), in opposition to what has been observed for other materials [26–27] where the joint failed internally. At 1350°C, the type I joint does not show as brittle behavior as at 1100 °C (Fig. 8C) and the strain prior to

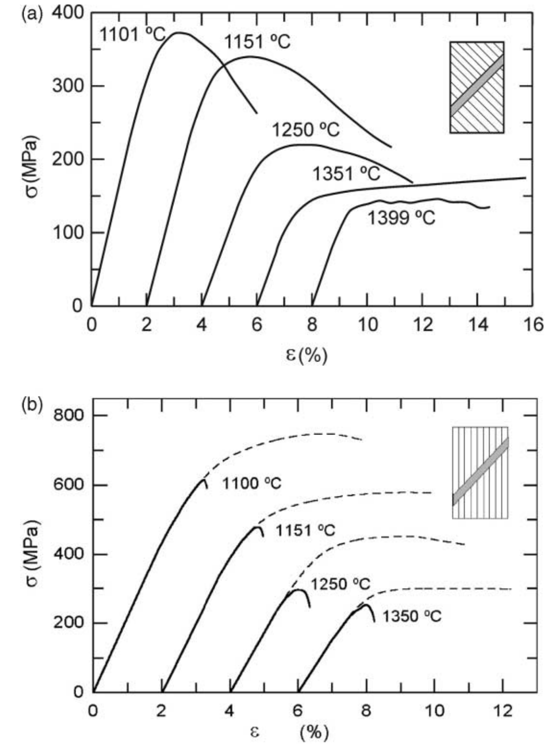

Fig. 6. Plot of stress versus strain as a function of temperature for the compressive experiments on the joints: (a) type I joints, (b) type II joints. The curves of bulk material compressed parallel to the axial direction are included in (b) for comparison (dashed line). A drawing of the joint geometry is included as an insert in the graphs. The curves are displaced on strain axis for clarity. See text for further discussion.

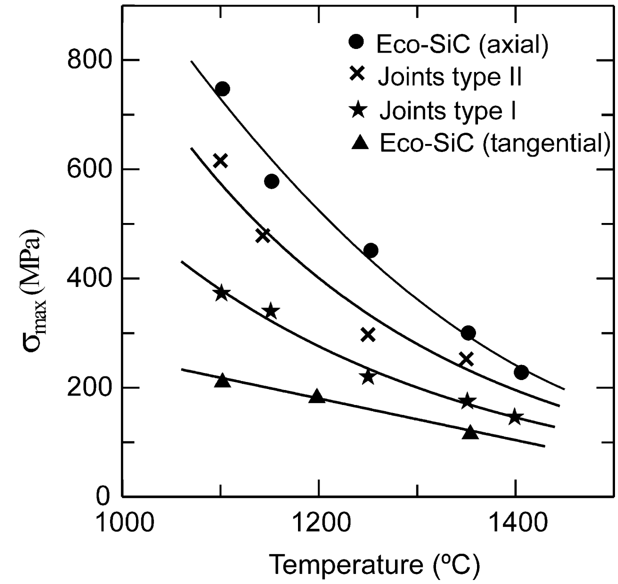

Fig. 7. Plot of the maximum strength as a function of temperature for the experiments of Figs. 2 and 6.

failure is almost double. Fig. 8D shows the detail of a type I joint deformed at 1350 °C where the silicon carbide grains and some silica formation can be observed. In deformed samples containing a type II joint, cracks were commonly observed in the bulk next to the joint region (Fig. 8E and F, type II joint deformed at 1350 °C).

A comparison of the compressive strength of joints in Ecoceramics and other types of SiC based materials is shown in Fig. 9. As discussed in previous compression studies of joints of sintered and reaction bonded SiC ceramics ,[26–27] the SiC grains in the joints are actually in contact and they can not move freely without undergoing some plastic deformation, so viscous flow of these grains cannot occur. The low load carrying ability of silicon at these temperatures is incompatible with a viscous flow model predicting the strengths measured in those studies. Likewise, similar mechanisms may also play a key role for the materials in this work. The strength of joints in sintered SiC (Fig. 9) is very high due to the higher effective strain rate in the joint (bulk sintered SiC does not deform plastically at these temperatures) and the lack of Si in the bulk material. In reaction formed silicon carbide (RFSC) the strength of bulk and joint is similar and the joined material behaves like the bulk.

As the joints for RFSC and biomorphic silicon carbide are fabricated using the same process, the joining region is expected to have the same strength in both materials. The bulk RFSC from Ref. 27 has a higher strength than the bulk biomorphic SiC studied in this work. It would be expected then that the strength of the joints of biomorphic SiC were determined by that of the bulk biomorphic SiC, and that the bulk and joint behaved similarly, as occurs in RFSC. The joints of biomorphic SiC studied in this work, however, have lower strength than the bulk (to make this statement we compared the bulk material in the axial directions with type II joints).

Plotting together the compression experiments curves of the bulk biomorphic SiC in the axial directions and of the type II joints (Fig. 6B), it can be seen that both systems yield plastically in the same way, but the joints fail well into the plastic region of the bulk. Bulk and joint then have similar plastic behavior, probably controlled by the bulk strength as expected, but the joints have a lower failure strain. The reason for this early failure can be found in microstructural observations like the one shown in Fig. 8E and F, where damage originated in the bulk reach the joint (type II). Due to the particular microstructure of biomorphic SiC, the cracks originated during plastic deformation are aligned with the material fibers. In the case of type II joints these cracks form a 45° angle with the joint and propagate very easily into it, resulting in an earlier failure than the material without a joint. The damage of the bulk with the orientation used for type II joints originate at a higher applied stress than for the orientation

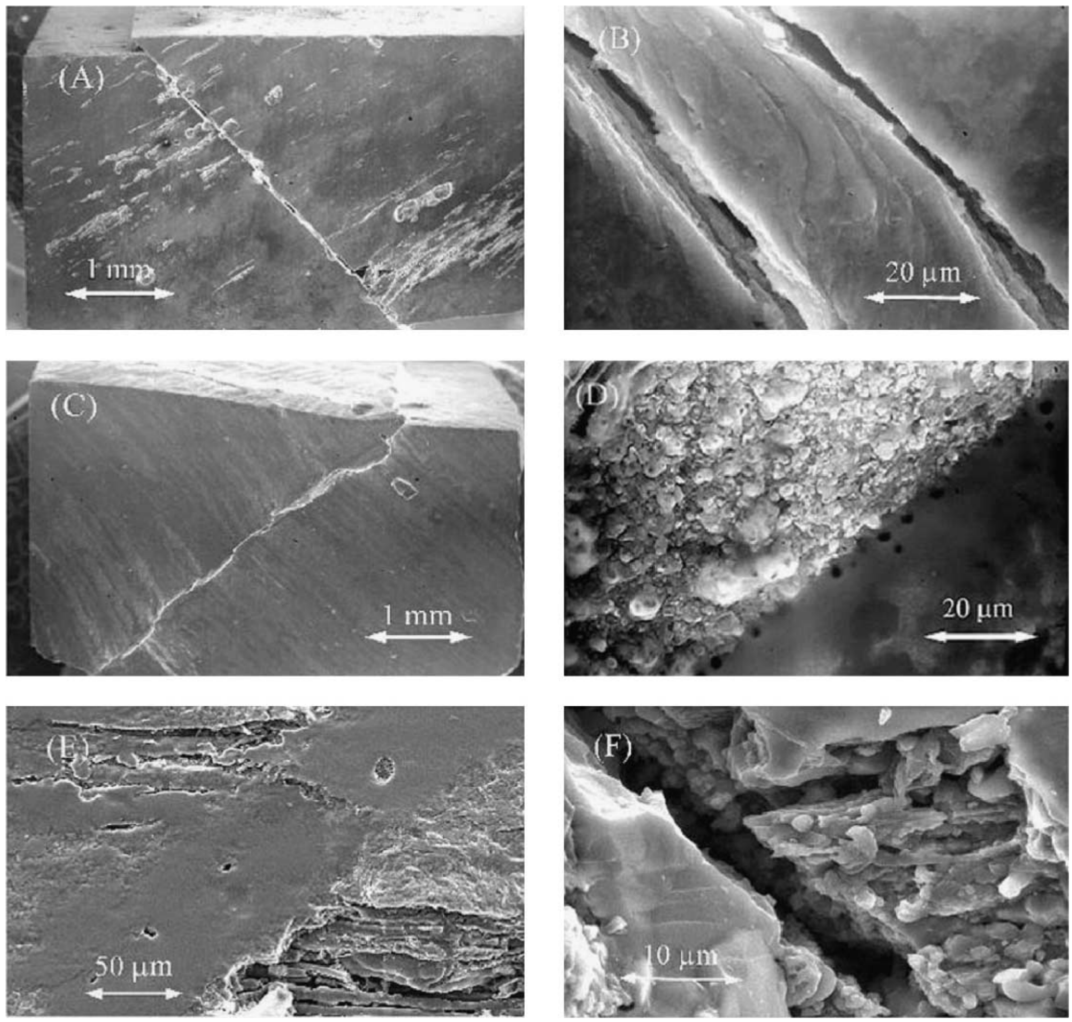

Fig. 8. SEM micrographs of joint after high temperature deformation. (A) Type I joint deformed at 1100 °C, (B) detail of (A), (C) type I joint deformed at 1350 °C, (D) detail of (C), (E) type II joint deformed at 1350 °C, and (F) detail of (E). See text for further discussion.

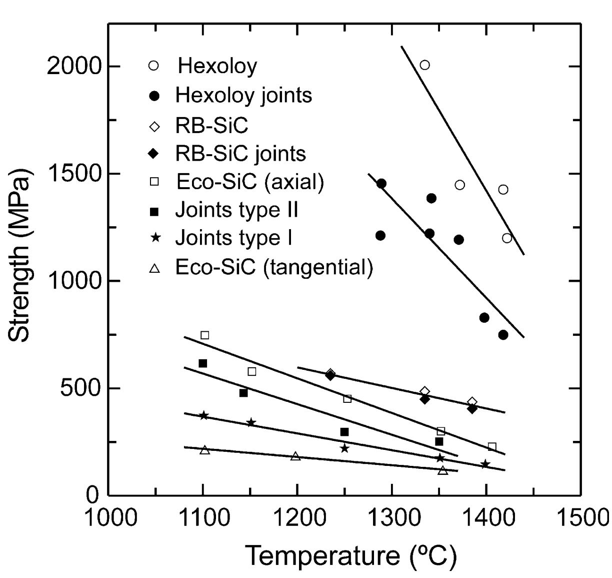

Fig. 9. Comparison of compressive strength of biomorphic SiC joints and joints of other silicon carbide based materials.[26,27].

used for type I joints, which could cause a faster propagation of the cracks in the joint–bulk interface and be responsible for the lower strain to failure of type II joints. The presence of silicon regions in the joints also makes the crack propagation into the joints easier and could also contribute to some strength degradation.

4. Conclusions

It has been demonstrated that SiC-based environment conscious ceramics can be fabricated from renewable natural resources. These ceramic materials have a microstructure that resembles the microstructure of the wood preform. They behave as a silicon carbide-based cellular solid, capable of reaching very high strengths.

In addition, the ARCJoinT approach can be used to produce joints with good strength in these materials. The ultimate strength of joined biomorphic SiC is controlled by two factors:

1. The strength of the bulk material, which depends on the relative orientation of the compression axis with the axial direction of biomorphic SiC. It determines the plastic yield of the joint.
2. The propagation of cracks through the joint–bulk interface, which depends on the relative orientation of the joint plane with the axial orientation of biomorphic SiC. It determines the strain to failure.

It is likely that regions of silicon next to the joint–bulk interface also play some role on the degradation of the joint strength. Therefore, it is necessary to determine the origin of the free silicon in the joints and reduce its presence. These findings will result in higher performance of joints.

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