Lasers in Manufacturing Conference 2021

# Laser-based manufacturing of ceramic matrix composites

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**Abstract**

A novel fabrication route for ceramic matrix composites (CMCs) was investigated. This was based on laser-induced melting of a Y-Si-O matrix between SiC fibers. Scanning electron microscopy studies showed that melting and solidification of the matrix is possible without damaging the fibers. Thus, the feasibility of the process in principle was demonstrated. Due to the strong inhomogeneities in the microstructure, samples were prepared for mechanical characterization using FAST/SPS. Bending and tensile tests were performed on them at room temperature and at 1000 °C in air atmosphere. The mechanical behavior was damage tolerant and showed the dependencies between strength and fiber orientation typical for CMC.

Keywords: Ceramic matrix composites; laser-based fabrication; mechanical properties

## 1. Introduction

In the field of high-temperature materials, ceramic fiber composites (CMCs) represent a comparatively young class of materials. They consist of ceramic fibers embedded in a ceramic matrix. The so-called pull-out effect of the fibers gives these materials damage-tolerant properties. This behavior strongly distinguishes them from monolithic ceramics with their typically brittle fracture behavior [1].

CMCs are typically fabricated by sintering a powder-based matrix, infiltrating with polymers and pyrolyzing them (PIP), chemical vapor infiltration (CVI), or liquid silicon infiltration (LSI) [1–6]. These processes are based on the infiltration of fiber bundles or prepregs through a matrix material, which occurs before or during an furnace-based heat treatment. A generative build-up of bodies cannot be done with these processes.

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Furthermore, the joining of CMCs would be advantageous in some applications. In this regard, current solutions are based on soldering, but this results in brittle fracture behavior in the joining zone [7].

In this article, a laser-based manufacturing process is presented, which for the first time enables the fabrication of ceramic fiber composite structures without the use of furnace technology [8]. The process principle is based on the fact that ceramic fibers are desized as bundles and coated with a powder suspension. After drying, the coated fiber bundle is deposited in a defined manner and heated locally by means of a laser so that the matrix material melts and then solidifies again shortly afterwards. To assess the feasibility, microstructural images were evaluated and mechanical characterizations were carried out.

## 2. Experimental

Ceramic fiber of silicon carbide (Tyranno SA3, Ube Ind., Tokyo, Japan) was used. To prepare an yttrium silicate matrix, $Y_2O_3$ (Grade C, Höganäs AB, Höganäs, Sweden) and $SiO_2$ (Zandosil 30, Heraeus, Hanau, Germany) were mixed in a molar ratio of 1:2 via attritor grinding in isopropanol and homogenized. After drying, an aqueous suspension was prepared from this powder mixture. The fiber bundles were continuously coated with the suspension, dried and fed to the laser unit (Figure 1).

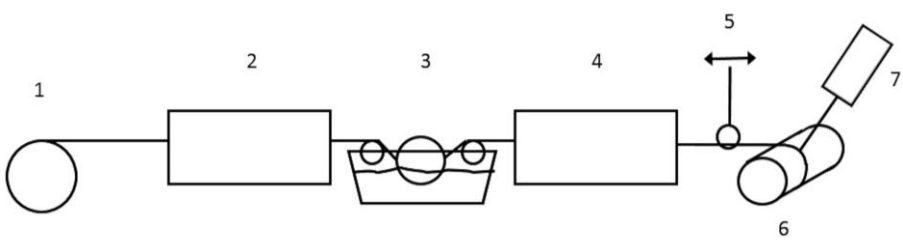


Fig. 1. Schematic drawing of the manufacturing process; 1: Fiber-roll, 2: Desizing, 3: Suspension ccoating, 4: Drying, 5: Fiber placement, 6: Rotating mandrel, 7: Laser [8]

The laser unit consists of a $CO_2$ laser (Coherent Diamond J-3 10.6 400 W OEM laser) and a computer-controlled laydown mechanism (Axial Ingenieurgesellschaft GmbH, Dresden, Germany). The complete laser unit is enclosed and flushed with argon so that the oxygen content is kept below 0.4%. The choice of materials and laser are based on investigations described in [9]. Microstructural investigations were performed on these samples using a high-resolution electron beam microscope (NVision40, Carl Zeiss AG, Oberkochen, Germany) and electron dispersive X-ray analysis (EDS).

The selection of the Y-Si-O matrix is based on the following aspects:

- In thermodynamic equilibrium, yttrium silicates are formed, which have sufficient mechanical stability at and excellent corrosion stability at room and high temperature [9–11]
- The coefficient of thermal expansion is close to that of SiC fibers and there are no chemical interactions between the components [12,13]

For strength determination, specimens were prepared by means of FAST/SPS (Field Assisted Sintering Technology / Spark Plasma Sintering). For this purpose, the fiber bundle was coated as described above and wound onto a mandrel without drying. However, no laser treatment was carried out. The winding was cut open, released from the mandrel and pressed into a flat scrim. After subsequent drying, eight such scrims were stacked alternately at 90° or 60° and placed in an 80 mm diameter carbon mold. This was followed by heat

treatment at 1400 °C with a heating and cooling rate of 200 K/min. Tensile test specimens were produced from the pressed plates (Figure 2). The orientation of the fibers to the tensile axis was varied, resulting in orientations of 0°/90°, +30°/-30° and +60°/-60°. In comparison, bending rods with 4 mm x 2 mm x 23 mm and 2.5 mm x 2 mm x 23 mm were produced and tested in a 4-point bending test at room temperature and 1000 °C in air. Each measured value is based on 4 to 6 individual measured values.

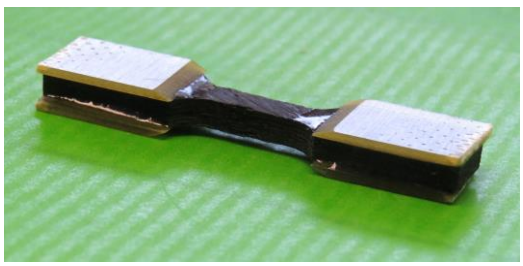

Fig. 2. Tensile test specimen with glass fiber reinforced polymer pads glued to the shoulders to reduce stress gradients from clamping

## 3. Results and discussion

Laser treatment was performed with a power of 10 W to 20 W and a constant take-off speed of 800 mm/min. The effective width of the laser port was varied by varying the deflection of the 1.1 mm wide beam so that the energy density ranged from 0.71 J/mm² to 1.36 J/mm². On the basis of the microstructural images (Figure 3, A and B) can be seen that the melting behavior is comparatively inhomogeneous. The light areas are completely melted matrix. The gray areas are matrix in the powder state. Gray circular areas are orthogonally cut fibers. Images C and D represent magnifications from section B. In image C, overheating occurred so that, on the one hand, Si-O species evaporated and recondensed as SiO2 and, on the other hand, the fiber material was dissolved by the molten matrix. This strong fiber-matrix bonding is unfavorable for the mechanical behavior of CMCs. Figure D shows an area where the matrix was melted, but no overheating occurred, so that no strong bond between fibers and matrix was formed. This can be seen from the fact that solidification-induced matrix cracks do not propagate into the fiber material but are deflected or stopped at the interface.

The reason for these different microstructural formations is most likely the inhomogeneous coating of the fibers with matrix powder. If there is sufficiant matrix powder on the roving, the energy of the laser is used for the melting enthalpy of the material. If there is little matrix powder, the energy input leads to overheating since less volume is available for phase transformation solid/liquid.

At the same time, there are still areas which have not been melted. The process-related one-sided coating of the fiber bundle resulted in the adherent matrix material being partially present on the underside of the deposited bundle. The superficial coupling of the laser and the high absorption of the SiC fiber material then led to insufficient through-heating. However, increasing the laser energy resulted in the fiber bundle breaking off due to excessive thermal stress on the fibers, or their degradation by the melt.

It is expected that an adjustment of the parameters (homogeneous coating of the roving, feed rate, energy density of the laser, etc.) will allow a significant improvement of the microstructure. At the time of writing this article, it was not yet possible to carry out corresponding tests due to the lack of availability of the equipment.

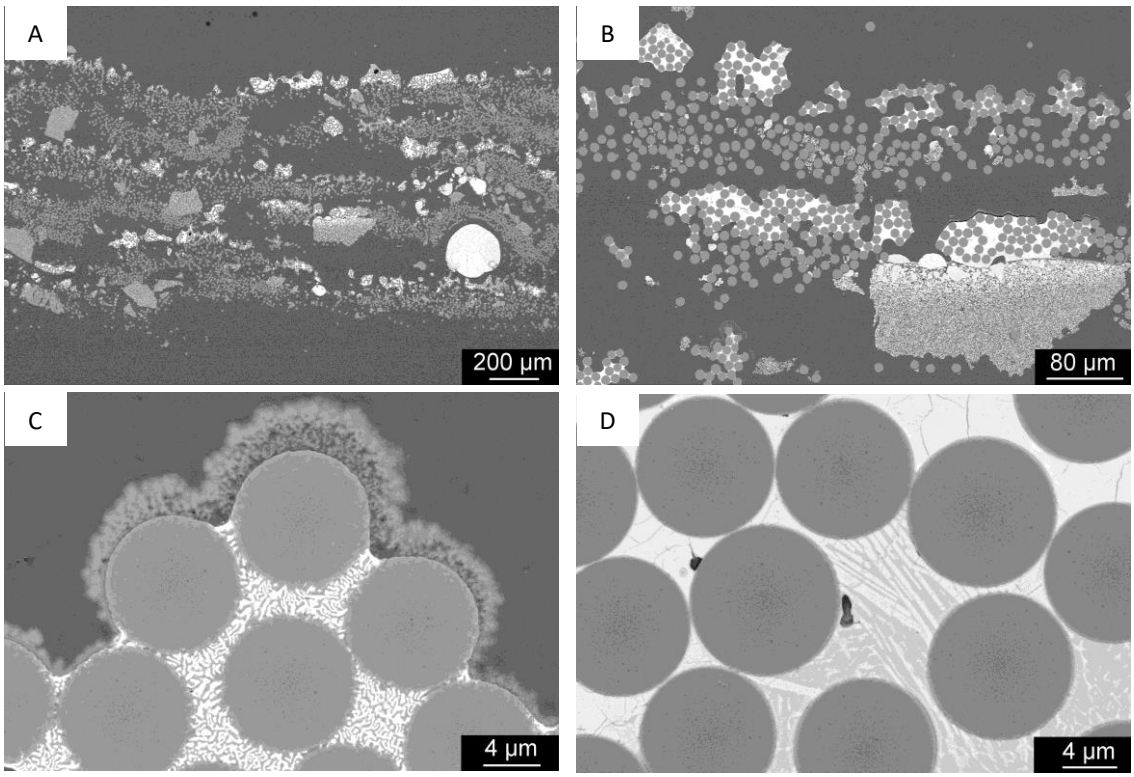


Fig. 3. SEM pictures of the microstructure after Laser-treatment; (A) complete cross-section, (B) magnified middle-section; (C) overheated region, (D) aimed microstructure

In order to nevertheless obtain mechanical characteristic values, sample material was produced using FAST/SPS technology. Due to the high heating rates that can be achieved, similar states of disequilibrium can be obtained as by laser-based melting. (Figure 4). The disadvantage of the process, which prevents a practical application for the material class, is the very limited variety of geometries. Only round plates with a maximum diameter of 80 mm can be produced. Nevertheless, it was possible to generate specimens for tensile tests. Comparing the right picture with Figure 3 D, it can be seen that the fibers are also embedded in a matrix, which was achieved by the molten phase, but remained sufficiently weakly bonded. In the left picture of Figure 4 however, it can be seen that the fiber content in the cross-section is significantly higher than in Figure 3 A. Due to the complete melting of the matrix and the applied pressure, a higher compaction was achieved than in the laser tests.

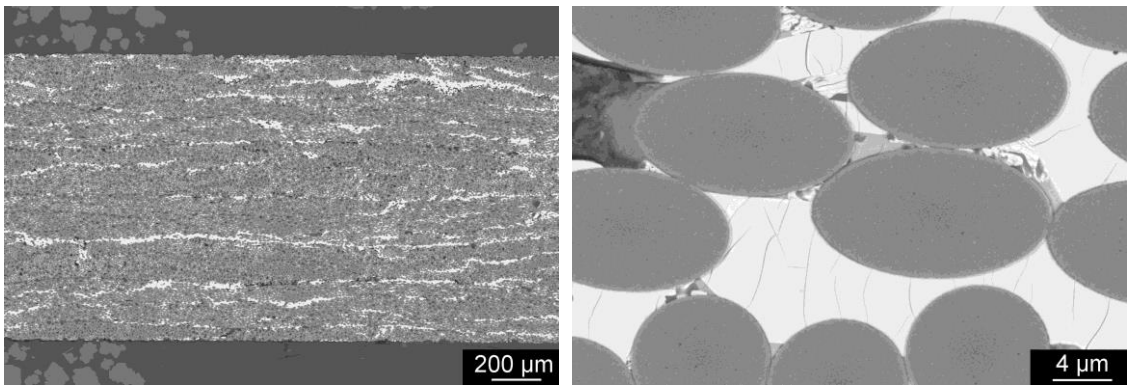


Fig. 4. Sample after FAST/SPS-treatment; (left) complete cross section, (right) higher magnification

Figure 5 shows the measured values of the tensile and flexural tests at room temperature. It can be seen that in the tensile and bending tests, the strength and stiffness are strongly dependent on the fiber orientation. As the orientation deviates from the loading axis, the mechanical properties decrease. This is in agreement with behavior of CMCs already reported in the literature [14]. The comparatively large differences in strength values between tensile and flexural tests are striking. Here, the magnitude of the values from the tensile tests is about 50% of the values from the flexural tests. This is due to several reasons. On the one hand, the effectively tested volumes of the specimens differ very significantly and, on the other hand, it can be assumed that several stress states are superimposed in the tensile test. These are caused by the clamping device and have a very significant effect on the measured values due to the small specimen size. For this reason, significantly larger specimens are required by the standard (ASTM C1275). However, these could not be produced using the methods described.

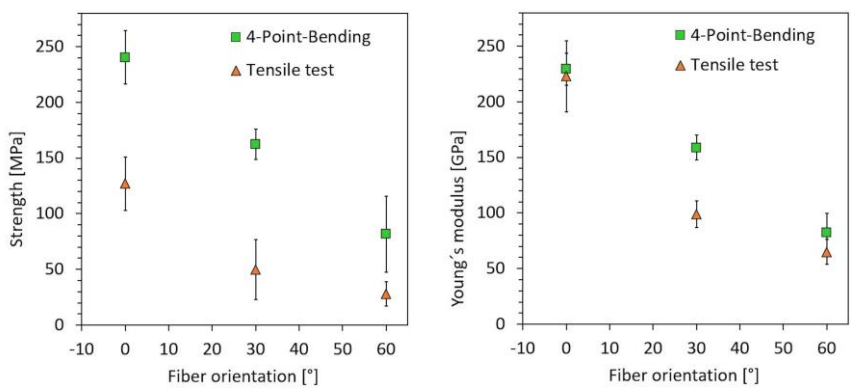


Fig. 5. Mechanical properties of FAST/SPS specimens depending on fiber orientation at room temperature; (left) Tensile strength, (right) Young´s modulus

The stiffnesses measured in tension and bending are in good agreement, with lower values in the tensile test at +30°/-30°. However, the exact cause for this behavior could not yet be determined. The measurement of flexural strength at 1000 °C showed no degradation of mechanical properties at 0°/90° and +30°/-30°

orientation. However, due to the necessary preload during heating, no values could be determined at +60°/-60°, as the stress reached in the specimen during this process exceeded the strength of the material.

Table 1. Bending strength depending on fiber orientation at room temperature and at 1000 °C in air atmosphere

| Orientation | Bending strength at room temperature (MPa) | Bending strength at 1000 °C (MPa) |
|---|---|---|
| 0°/90° | 240 (+-24) | 230 (+-103) |
| +30°/-30° | 163 (+-14) | 195 (+-44) |
| +60°/-60° | 82 (+-34) | n.d. |

From the fracture surfaces (Figure 6) it can be seen that the type of failure or crack propagation depends strongly on the orientation of the fibers. At 0°/90°, clear fiber pull-out can be seen in 0° orientation. In +60°/-60° orientation, failure is mainly characterized by interlaminar shearing. A direct fiber pull-out does not occur. Both effects, fiber pull-out and interlaminar shear, increase the surface area during crack propagation and thus enable the damage-tolerant failure behavior that characterizes CMCs.

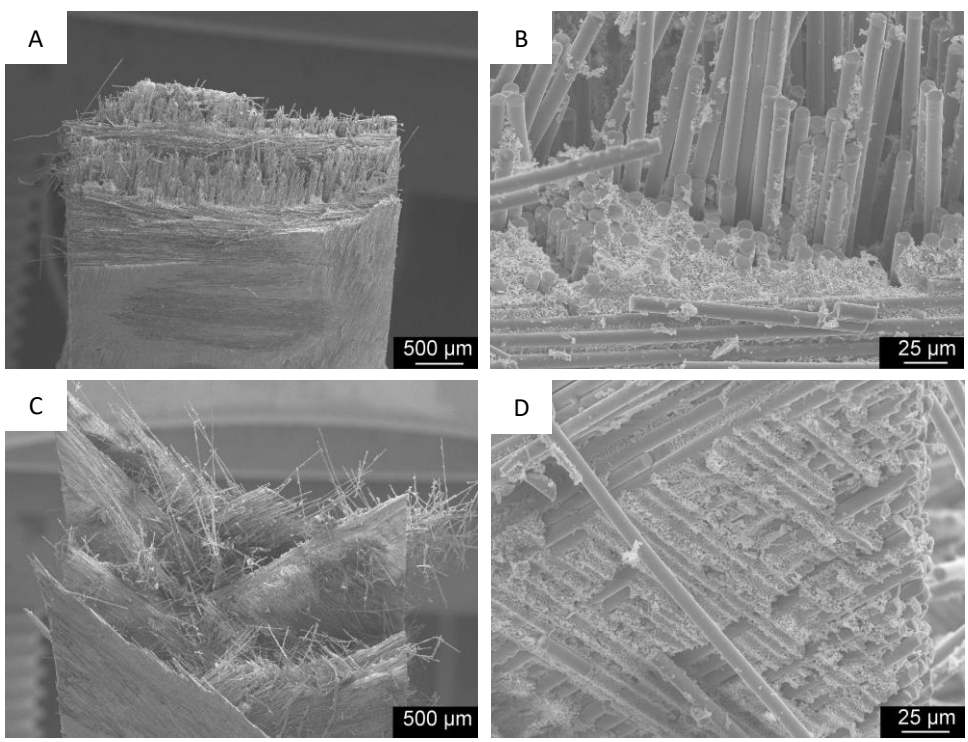


Fig. 6. SEM micrographs of the fracture surface after tensile test; (A) 0°/90°, (B) higher magnification of (A); (C) +60°/-60°; (D) higher magnification of (C)

## 4. Conclusion

In this article, a novel laser-based process for the fabrication of CMCs is presented. In this process, Y2O3 and SiO2 are locally melted between SiC fibers by laser and form the Y-Si-O-based matrix after solidification. Microstructural investigations have shown that the melting and solidification of the matrix is possible without damaging the fibers. A homogeneous microstructure could not be achieved so far due to the inhomogeneous loading of the fiber rovings with the matrix-forming powder.

For mechanical characterization, comparative samples were prepared by means of FAST/SPS and tested in bending and tensile tests. Due to the higher fiber volume fraction, no direct conclusions can be drawn from the material produced via FAST/SPS to the material produced by laser treatment. Nevertheless, the measured values allow an estimation of the potential of this novel material and process combination. In both cases the formation of a Y-Si-O matrix is carried out from the molten phase and, thus, the interactions between fibers and matrix are the same. The strength values were in acceptable ranges and the fracture behavior was clearly damage tolerant. This was also demonstrated at 1000 °C in an air atmosphere without any barrier coating.

The authors assume that for the laser process, a more homogeneous powder-loading of the roving and appropriately adjusted laser parameters, a homogeneous microstructure and promising material properties can be generated. This would enable applications in the field of additive manufacturing or joining by retaining damage tolerant properties.

## Acknowledgements

This work was conducted within a project funded by the German Federal Ministry for Education and Research (BMBF) with the number 03ZZ0222C.

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