Laser Beam Joining of Non-Oxidic Ceramics for Ultra High Temperature Resistant Joints

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ABSTRACT

The excellent technical properties of silicon carbide (SiC) and silicon nitride (Si_3N_4) ceramics, such as resistance to extreme temperatures, oxidation, mechanical wear, aggressive chemical substances and radioactive radiation and also its high thermal conductivity and good temperature-shock resistance, make these ceramics ideally suited for use in the field of nuclear technology.

However, their practical use has been limited so far because of the unavailability of effective joining techniques for these ceramics, especially for high temperature applications.

A new joining technology *(CERALINK@)* has been developed in a network project which allowed high temperature resistant and vacuum-tight joining of SiC or Si_3N_4 ceramics.
A power laser is used as heat source, which makes it possible to join ceramic components in free atmosphere in combination with a pure oxidic braze filler. As no furnace is necessary, there are no limitations on the component dimensions by the furnace-geometry.
During the joining process, the heated area can be limited to the seam area so that this technology can also be used to encapsulate materials with a low melting point. The seam has a high mechanical strength, it is resistant to a wide range of chemicals and radiation and it is also vacuum-tight.
The temperature resistance can be varied by variation of the braze filler composition – usually between 1,400 °C and >1,600 °C. Beside the optimum filler it is also important to select the suitable laser wavelength. The paper will demonstrate the influence of different wave lengths, i. e. various laser types, on the seam quality. Examples are chosen to illustrate the strengths and limitations of the new technology.

INTRODUCTION

The safe and long-time stable encapsulation of radioactive material under high-temperature conditions presupposes suitable materials for encapsulation. High-performance ceramics such as silicon carbide (SiC) and silicon nitride (Si_3N_4) can meet a wide range of requirements that result from such conditions. They have excellent material properties as regards thermal resistance, corrosion resistance, long-time stability of tightness and radioactive radiation resistance. So the disintegration temperature of SiC is > 2300 °C and of Si_3N_4 is > 2170 °C. Since these ceramics have no melting phase (which is positive for their field of application), they cannot be joined by conventional welding processes. Numerous research projects, mainly of the 1980s and 1990s, were concerned with this problem. To achieve a thermal resistance of 1000°C, metallic active fillers (such as [Ura 1999], [Tur 1993], [Tam 1996]) and non-metallic glass and ceramics fillers (such as [Lem 1995], [Hes 1993], [Lee 1998], [Wie 1990]) were developed and special processes such as reactive joining were investigated and used ([Sin 1998], [Tza 1994], [Nak 1996]).
The fields of application have remained very limited because the required process variables such as process atmosphere (vacuum or protective gas), process pressure (partly up to 200 MPa), process time (often hours due to the use of furnaces) and the related costs were a hindrance to their efficient use. Further restrictions result from the use of furnaces as energy source since these generally heat the whole component, i.e. it is impossible to produce mechanically strong, corrosion and high temperature resistant joints with locally restricted energy input if adjacent low melting components (to be encapsulated) are present.

THE JOINING PROCESS

If parts made of these ceramics are to be bonded, the absence of a melting phase makes it necessary to introduce a suitable braze filler to the area to be joined and to fuse the braze filler by means of a suitable heat source.

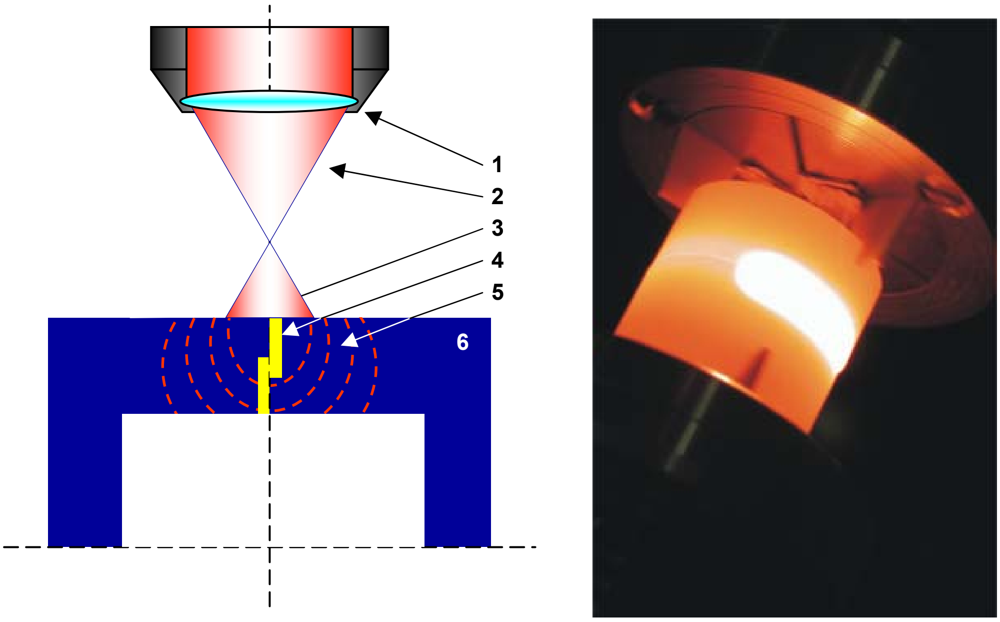


Figure 1 Laser beam joining process
Left side: sectional drawing (with: 1 = Laser optics, 2 = Laser beam, 3 = Heated surface, 4 = Joining seam with filler, 5 = Temperature field, 6 = SiC half capsule)
Right side: dimmed picture of joining process

A purely oxidic filler consisting of aluminium oxide (Al_2O_3), yttrium oxide (Y_2O_3) and silicon dioxide (SiO_2) has proved to be optimally suited for the joining process.

The oxide ratios can be altered to set the softening and melting temperatures of the synthesized braze filler. The maximum temperature range for application of this compound can be varied from 1400 °C to >1600 °C.

If the joined components are to be used in a reactor core, the relatively high absorption cross-section for neutrons of yttrium can present a problem. In this case, yttrium oxide can be replaced by another oxide, which has a lower absorption coefficient.

The coefficient of expansion of the braze fillers is selected so that it is close to the coefficient of expansion of the ceramics to be brazed. This is easier for SiC than for Si_3N_4. Since in the filler system used both the coefficient of expansion and the melting point can be determined by the percentage of SiO_2, both aspects should be considered in the composition of the filler. The different coefficients of expansion of the ceramics SiC and Si_3N_4 also have the effect that the joining of SiC parts with Si_3N_4 is extremely problematic.
Wetting of both ceramics by the oxidic braze filler is optimal. This provides a decisive advantage over the conventional methods: pre-treatments such as etching, metallization or activation are not required. The composition of the braze filler eliminates the need of a protective gas or vacuum even during the joining process - the examinations were conducted in free atmosphere.

The energy source used to fuse the braze filler is a laser. Extensive examinations conducted with CO_2 lasers, Nd:YAG lasers and diode lasers dealt with the question which laser wavelengths, i.e. laser types, are best suited for joining.
Systems applied were: a CO_2 laser (wavelength: 10.6 µm) with a maximum cw laser beam power of 2000 W, a Nd:YAG laser (wavelength: 1.06 µm) with a maximum cw power of 1,000 W and diode lasers (wavelength: 0.808 µm and 0.940 µm) with a maximum cw power of more than 3,000 W.
The defocused laser beam was directed towards the seam area. For this reason and due to the high thermal conductivity in combination with a small thermal expansion coefficient of both ceramics (SiC: $\lambda > 70 Wm^{-1}K^{-1}$, $\alpha > 4.5 \cdot 10^{-6}K^{-1}$; Si_3N_4: $\lambda > 40 Wm^{-1}K^{-1}$,$\alpha > 3.0 \cdot 10^{-6}K^{-1}$) laser processing is possible without preheating [Rei 2001]. A laser-supported radiation pyrometer, which can measure the surface temperature and the emissivity coefficient of the ceramic material, was employed to control the temperature against the power of the laser. Cylinder-shaped capsules of LPSSiC, SSiC and Si_3N_4 were used for the study.

As a result of the different absorption of the wavelengths used both in the brazing filler and the material, the filler melted at different depths. At a temperature of 1,500°C, about 89% of the wavelength of 1.06µm (Nd:YAG) and about 59% of the wavelength of 10.6µm (CO_2) were absorbed by the SiC. The absorption of the filler is not sufficiently known, however, its properties can be compared with those of glass. Oxidic glass materials absorb less

than 15% of a wavelength of 1.06µm and more than 80% of a wavelength of 10.6µm [Zha 1996], [Bla 1989], [Tsa 1997].
It was found out experimentally that the penetration of the CO_2 laser radiation in combination with the thermal conduction is about 33% less than with the Nd:YAG laser in case of silicon carbide. The interaction of the diode laser beam with the ceramic material is quite similar to the Nd:YAG laser radiation.

Furthermore, through treatment by Nd:YAG radiation the ceramic surface underwent commutations (Fig. 2). The laser-affected zones were characteristically marked by a thin foamy glass phase at the surface. When the CO_2 laser was used, no significant changes were detectable on the SiC surface (Fig. 3). The Si_3N_4 surface was coated with a white, some micrometers thin SiO_x layer (Fig. 4).

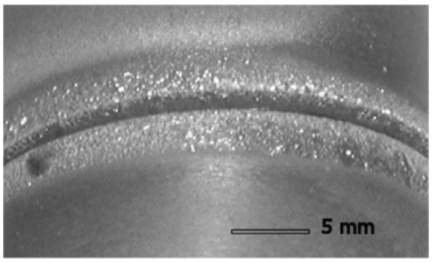


Figure 2
View of the Nd:YAG laser-brazed SiC capsule

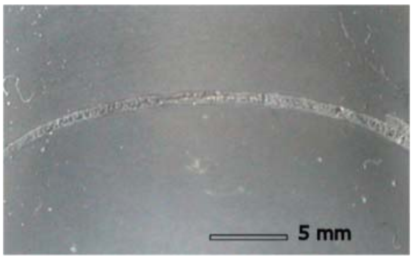


Figure 3
View of the CO_2 laser-brazed SiC capsule

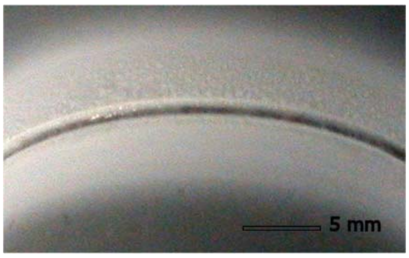


Figure 4
View of the CO_2 laser-brazed Si_3N_4 capsule

The structure of the re-solidified filler was formed irrespective of the absorption properties of both wavelengths. The brazing filler melted throughout the whole seam. The solidified phase looked homogeneous. The formation of (unwanted) pores depended on the gap width and on the composition of the filler. In a gap width smaller than 50µm no pores were observed. The helium leakage of the joints is $< 2\cdot10^{-8}$ mbar·l/s.

Generally all investigated types of lasers are applicable. Since high-power diode lasers (> 1,000 W) with acceptable focus diameter are now commercially available, this type of laser is favoured for further work. Other advantages of this laser type are its high efficiency und its very compact design. Fig. 5 shows a diode laser with 3,100 W continuous beam power.

Figure 5

Diode laser with 3,000 W beam power and flexible optical fibre cable [Rofin 2004]

(Height x Width x Length = 22x26x56 cm)

Then the aim was to examine by elemental analysis of the seam area which interaction occur in the composition of both the ceramics and the braze filler, using a laser spectrometer. Figure 6 shows two linescans over a line segment of 770µm taken at right angles to the seam. The capsule surfaces were cleaned from deposits before measurements were taken.

It should be pointed out that the moderate change in the elemental composition that occurs in the zone of transition between ceramics / braze filler / ceramics must be attributed to the focus diameter of the laser spectrometer which corresponds to about the seam width.

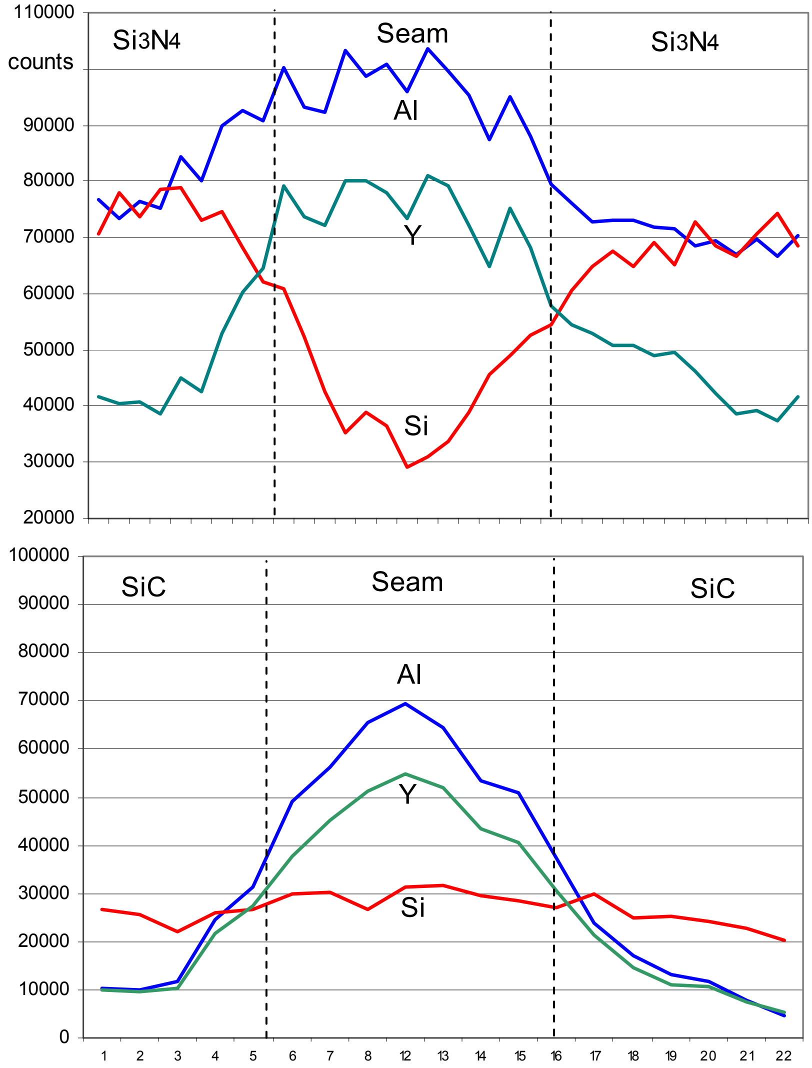


Figure 6
Linescan across the joining seam
Top: Si_3N_4 ceramic capsule
Bottom: SiC ceramic capsule
Shaded: braze filler area

It can be shown that the concentration of the elements yttrium and aluminium increases in the braze filler as expected. The decrease of the silicon concentration in the braze filler can also be expected since its proportion is cut by half as compared with the ceramics.
However, the measurement confirms the latter only for the Si_3N_4 seam. The concentration of silicon across the SiC seam remains unchanged along the line of measurement, i.e. the base material was not removed either. Any diffusion of silicon from the base material into the seam would lead to a reduction of the strength in these areas. Bending stress applied to brazed ceramic rods resulted in breakage even far off the heat-affected zone.
The 4-point bending strength of test bars (according to DIN 51110) was more than 70% of the strength of the virgin material.

APPLICATIONS

The applicability of the laser beam joining technology shall be demonstrated by two examples: encapsulation of radioactive material in cylindrical hollow SiC capsules as well as additional jacketing of spherical fuel elements with a SiC shell.

To demonstrate the first case, (inactive) pellets were encapsulated in SSiC. For this purpose two ceramic half capsules (Figure 7 – 1.) were joined gas-tight by means of the laser beam joining method. The aim was to create a long-term stable, high-temperature resistant, gas-tight and extremely corrosion resistant encapsulation for highly active material.

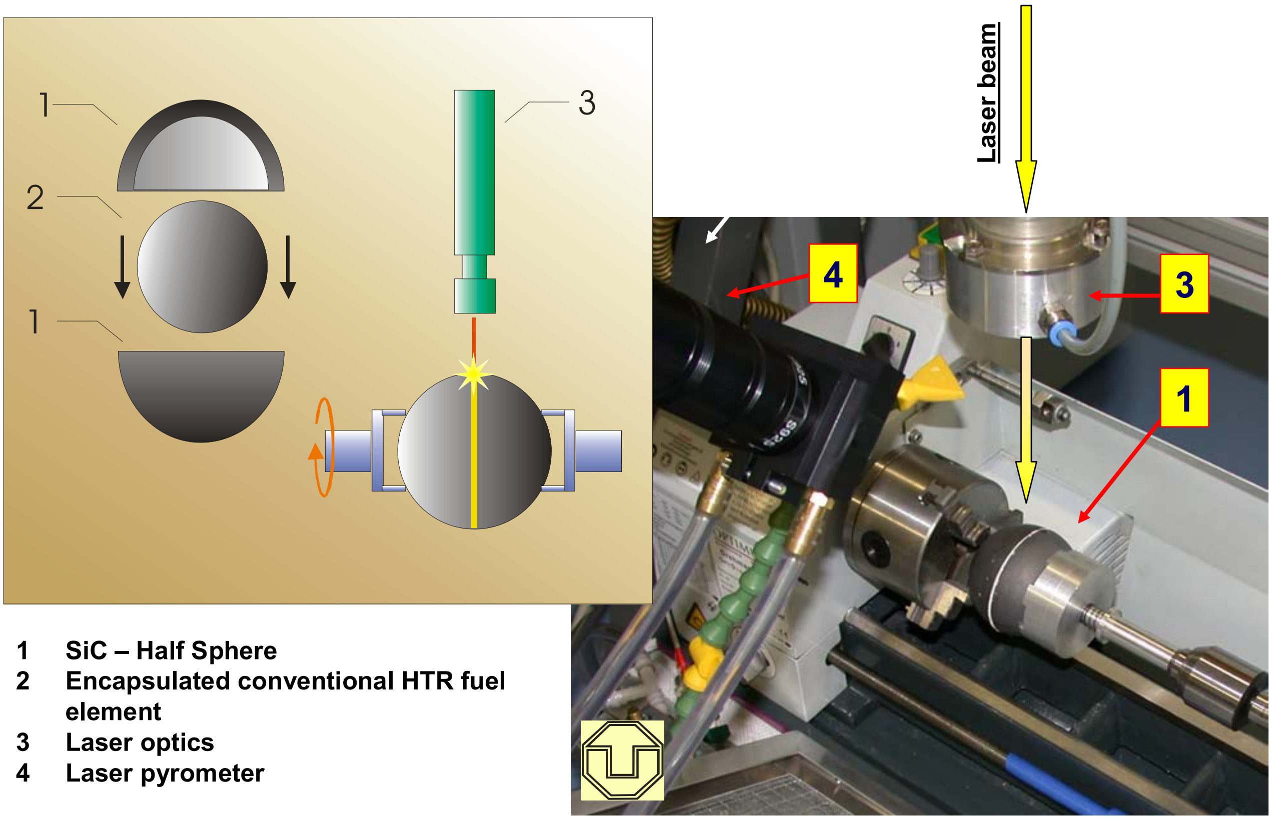
Although SiC and Si_3N_4 ceramics have extraordinary properties such as high-temperature and corrosion resistance which are unmatched by metallic materials, they are sensitive to shock loads, like all ceramic materials. When encapsulated radioactive substances are transported, such shocks might occur in transit the instant an accident happens. This leads to the requirement of further constructive measures in order to protect the capsule. The entire construction was built on the assumption that mechanical stress would occur first, followed by the extreme thermal stress. The test case defined for mechanical stress was a free fall from 38 m height (here: high-rise building – Figure 7) onto a concrete panel.

Figure 7 SiC Encapsulated Pellets - Free Fall Tests (Height: 38 m from concrete panel)

To prevent the ceramic capsule from being destroyed by the internal impact (caused by the impingement of the enclosed pellets onto the inner capsule wall), the free space between the pellet and the capsule wall was filled with coarse SiC granules (Figure 7 – 2.). To protect the ceramic capsule against an external impact, it was surrounded by a thick jacket of polysiloxane (resistant to temperatures of more than 300 °C) after joining. Subsequently, the containers manufactured in this way underwent mechanical tests. The result found out was that all containers had passed the tests without damage, irrespective of the impact angle (Figure 7 – 4 and 5).

The second application test aimed at enclosing the spherical fuel elements of a high-temperature reactor (e.g. pebble bed modular reactor) with an additional spherical SiC shell in order to improve the resistance to corrosion and abrasion of the graphite spheres. Furthermore, this can also improve the suitability of the spherical fuel elements for final storage.

As shown in Figure 8, the graphite spheres were enclosed by two custom-fit SSiC half shells. Before joining, a thin layer of braze filler was applied to the specially shaped joining areas. The sphere halves prepared in this way were lightly pressed together in a rotating chuck and joined by means of a laser beam. The experiments conducted so far have shown that is principally possible to apply the laser technology for this purpose.



1 SiC – Half Sphere
2 Encapsulated conventional HTR fuel element
3 Laser optics
4 Laser pyrometer

Figure 8 SiC encapsulation of HTR fuel elements by laser beam joining

SUMMARY

An innovative technology for the ceramic joining of non-oxidic ceramics has been developed, taking silicon carbide and silicon nitrite as an example. This technology is based on an oxidic glass ceramic filler and a customized laser technology. Nd:YAG as well as CO_2 or diode lasers can be used as energy source. The filler composition ensures that the technically relevant properties of the ceramic materials are maintained, such as high-temperature resistance, corrosion and radiation resistance. The process has the advantage that it can be carried out in free atmosphere. The joints are free of cracks, gas-tight and have a high mechanical strength. The surfaces to be joined need no pre-treatment. The energy input restricted to the seam and processing in free atmosphere allows the brazing of objects of geometrically unlimited sizes. Process times are limited to a few minutes; the joining process itself is completed within the range of a few seconds. Another advantage of the selective heating of the braze filler is that temperature sensitive materials can be adjacent to the components to be joined without being destroyed. It is also possible to encapsulate low melting materials, if necessary with additional cooling. So the process can also be used for the safe encapsulation of radioactive materials.

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University of Technology Dresden, Laserinstitut Mittelsachsen e.V. an der Hochschule Mittweida (LIM), Germany and TKC - Technische Keramik GmbH, Meißen, Germany

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