Joining of Ceramic and Metal Parts

Monolithic ceramics, composites or metals, which cannot be manufactured in one piece must be joined. By using joining technologies material hybrids are also possible. Ceramic-to-metal joinings expand the application spectrum enormously. By joining of simple serial parts complex geometries for specific applications can be realized.

Fig. 1
Kicker tube of a particle accelerator, consisting of externally copper-plated Al_2O_3 with brazed metal flanges on the front edge [3]

Fig. 2
Vacuum insulators with insulating tubes made of Al_2O_3 welded with ISO-KF or CF flanges [3]

The joining of components can be performed in different ways by using material adhesives as well as by form-closure. Each joining requires low dimensional tolerances, which can only be accomplished by machining. Mostly, non-permanent connections by interlocking metal to ceramic subcomponents are used. However, to obtain a permanent joining especially at high temperatures ceramics adhesives or solders are required. In regarding industrial applications, appropriate joining adhesives and cost efficient techniques must be selected [1, 2]. Fig. 1–2 show Al_2O_3 ceramic tubes joint to metal flanges. The parts were brazed and welded for their use in vacuum and activator technology [3].

Keywords
joining, ceramic-to-metal joining, ceramic-to-ceramic joining

Ceramic-based materials and suitable solders

Ceramic materials used for joining have to fulfil several requirements. Typical properties needed in severe environments are high chemical and mechanical as well as thermal stability at high temperatures of about 1000 °C [1]. With regard to permanent joining there is a high need to find out the most appropriate joining method and joining material. Both must be adapted to the base ceramic materials. The most common methods to join ceramics were evaluated in a literature review on scientific articles from the last ten years. A summary is presented in Fig. 3. It was concluded that joining is preferably performed in oxide furnaces using non-oxide ceramics. Metallic as well as glass solders are most often used for joining. Due to the high process stability and low costs, high temperature (HT)-furnaces are most commonly used. High temperature stable adhesives like solders based on glasses, glass-ceramics or metals are most appropriate.

In order to minimize thermal stresses during the joining process, the Coefficient of Thermal Expansion (CTE) must be considered. The CTE of the solder must be adapted to the CTE of the ceramic. In order to avoid microcracks, only small CTE tolerances $<\pm 0{,}5 \cdot 10^{-6}\ K^{-1}$ are tolerated. With ceramic-to-metal joinings, the inter-

C. Gadelmeier, J. Schmidt
Fraunhofer Institute for Silicate Research
Center for High Temperature Materials and Design HTL
95448 Bayreuth
Germany

www.htl.fraunhofer.de

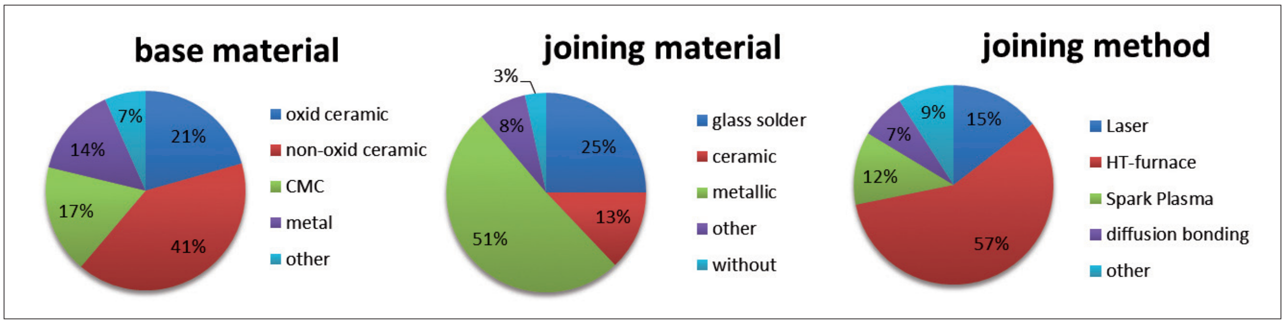

Fig. 3
Frequency of base materials, joining materials and joining methods reported in literature published between 2010 and 2016

mediate joint is a complex and sophisticated multilayer system.

Ceramic solders are commercially available in the market and were developed on customers' demand (Fig. 4). Suppliers are companies like Schott, Ferro and Corning. Organic adhesives can be used up to 300 °C and anorganic adhesives up to 2950 °C, respectively [4]. Glass solders can only be used up to 1050 °C in maximum. These joints are characterized by mainly amorphous – only partially crystalline – phases. If the cristallinity is higher than 90 %, the temperature stability can be increased up to 1600 °C. However, such material systems are still under development. The mechanical stability can be further increased through covalent bonding to the base ceramics and through the reduction of porosity.

The major disadvantage of available glass solders is their high silica content. The high-temperature stability of the joint is reduced noticeably by the formation of amorphous glass phases within the microstructure. Additionally, the pore volume content and the pore size within the joint increases leading to a remarkable reduction in strength [5].

The joining of ceramics to metals can be performed by metallizing ceramic surfaces or by using reactive metal brazes. Metal alloys can be used under controlled vacuum or in inert conditions, like argon atmosphere to avoid chemical reactions with nitrogen or oxygen [6–8]. Preferentially, the brazing process is performed by using titanium or silver containing alloys. Some eutectic alloys reveal high-temperature stability up to 1600 °C. Furthermore, ternary systems, e.g. AgCuTi were developed to perform the so called reactive-air-brazing (RAB) in air. It is suitable up

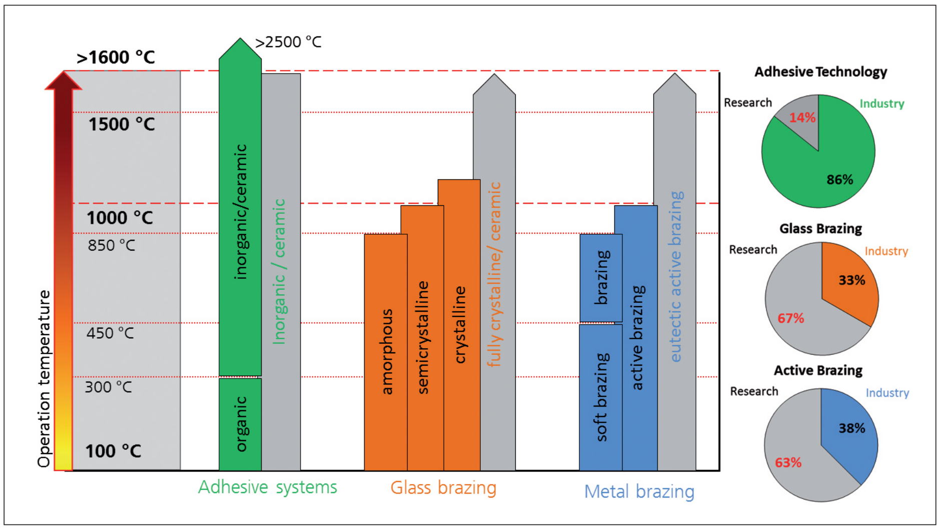

Fig. 4
Comparison of joining processes with respect to commercial availability and applicability at high temperatures

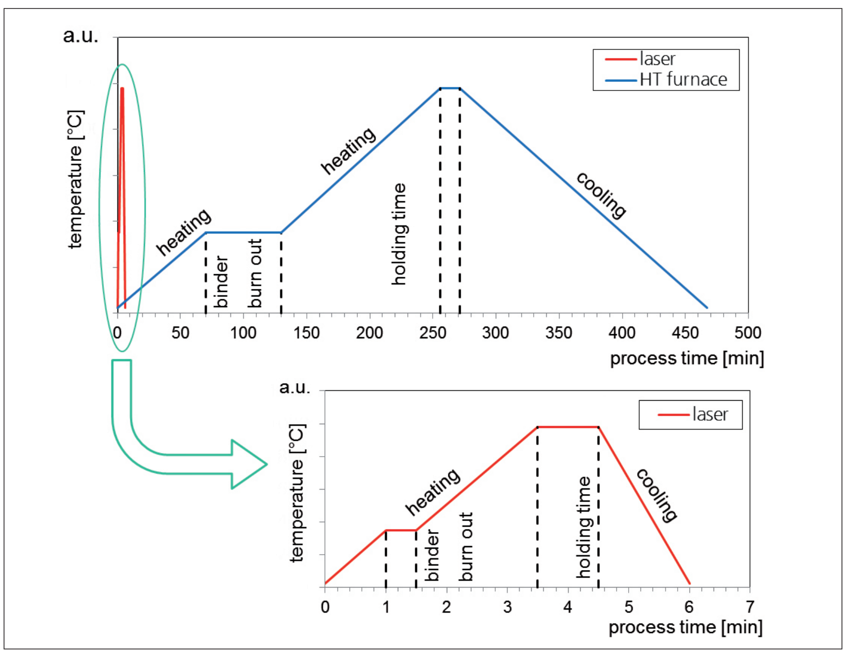

Fig. 5
Comparison of the process times between high-temperature furnace joining and laser joining

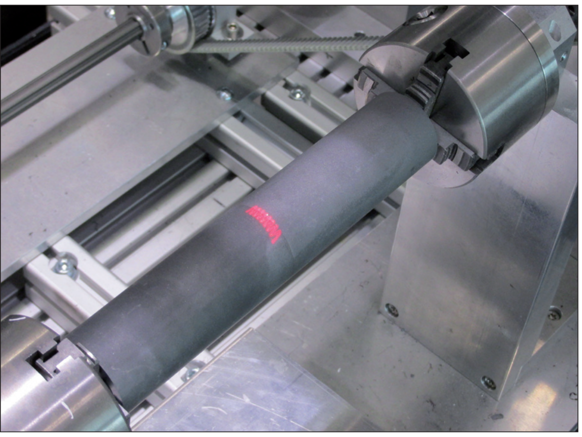

Fig. 6
Rotating joining device to perform a tube-to-tube laser joining process at Fraunhofer Center HTL

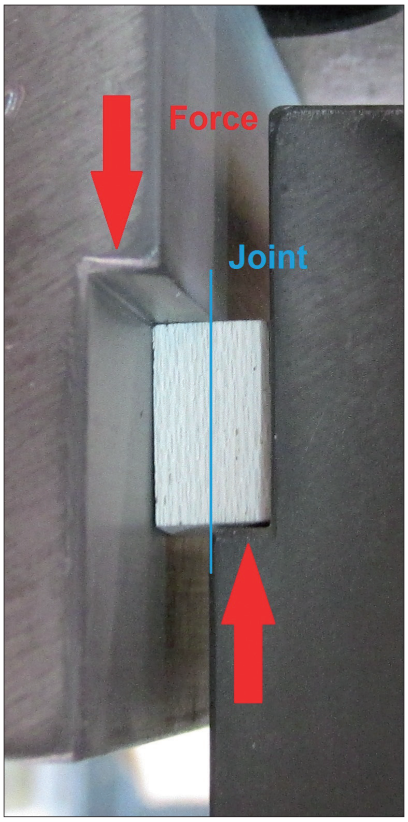

Fig. 7
Joined O-CMC samples (10 mm × 10 mm × 6 mm) prior to shear testing according to ASTM D 905

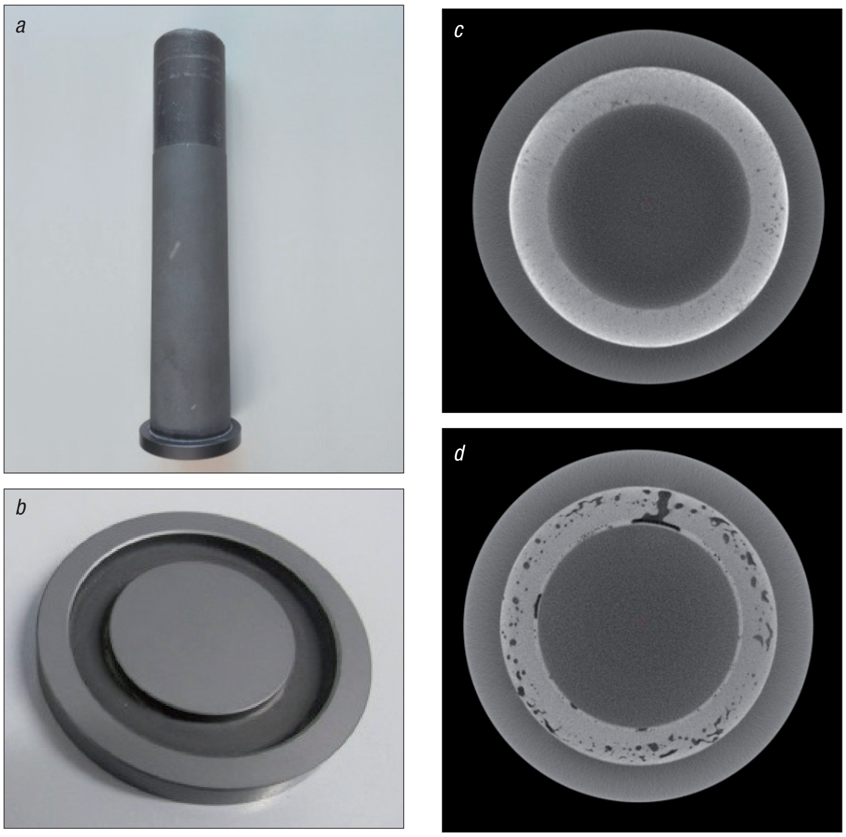

Fig. 8a–d
Tube-to-flange joints for SiSiC parts (a, b) with a new developed glass ceramic brazing (c), and commercial glass brazing (d), the latter showing large porosity

to a maximum operation temperature of 1000 °C [9].

Technologies to obtain ceramic-to-ceramic joinings

Today, most industrial joining processes for ceramic components are realized in high-temperature furnaces. The process times range from several hours to one day, depending on the heating and cooling cycles. Commercial glass brazing processes usually take between 8–24 h [1]. Using batch processes in large furnaces it is possible to process a high number of parts under adjustable gas atmospheres. Due to closely tolerance process parameters, furnace processes prove to be very reliable. The disadvantage is that it is not possible to intervene in the joining process during the heating cycle.

As an alternative, joining with laser heating can be used in order to achieve short cycle times within minutes (Fig. 5) [5, 10]. Laser heating enables local heating only of the joining zone. Furthermore, complex heating patterns can be realized using high-speed laser scanners for movement of the laser beam and CNC tables for movement of the parts. Thus, this process is very well suited for a small number of parts. It allows quick and direct corrections of the joining set-up and can be combined with measuring techniques for quality control. However, the process is complicated due to a high number of parameters, such as laser power, focal length, aperture and scan parameters. The complexity of the laser joining is caused by the interaction between the laser beam and the processing material. It was shown that a 3D-simulation of the temperature profile enables an optimization of the process parameters [11]. Fig. 6 shows a rotating CNC device coupled with a CO_2 laser system for the joining of ceramic tubes.

In addition to these joining methods, ceramics can also be joined by Spark Plasma Sintering (SPS). SPS is a heating process derived from conventional hot pressing. The energy input takes place via a high current, which acts directly onto the component and allows local volume heating. The advantages are a short processing time due to rapid heating as well as limited deformation of the joined parts [12]. Due to the uniaxial orientation of the pressing tool, however, only simple geometries can be realized in contrast to the laser and furnace technology.

Tab. 1
Applications for components of the joining technology [16]

Area of Application	Requirements for Basic Materials	Example	Materials Combination
Electrical industry	Electrically conducting/isolating	Circuit boards	Ceramic-ceramic Ceramic-metal
Power generation	High-temperature stable, good thermal properties, long-term stable, chemically stable, gas tight	Heat exchangers for heat recovery, compound-pipe	Ceramic-ceramic Ceramic-metal CMC-metal
Gas turbine and aviation	Resistant to thermal shock, good mechanical properties, low fatigue, gas tight	Flame tubes, guide and rotating vanes	CMC-CMC CMC-metal
Thermal engineering	Long-term stable, high- temperature stable, resistant to thermal shock, insulating, distortion-free	Carrier structures, insulating elements, ducts, hot gas valves	Ceramic-ceramic CMC-ceramic CMC-CMC CMC-metal
Metal industry	Long-term stable, high- temperature stable, resistant to thermal shock	Continuous process devices, Furnace fans	CMC-CMC, CMC-metal
Medical engineering	Bioinert, long term stable	Dental prostheses	Ceramic-metal

Testing and characterization of ceramic joints

For an evaluation of the component behaviour in the later application, it is necessary to determine mechanical strength values of the joined parts. Depending on the combination of ceramic, CMC or metal, different test standards are available. Suitable standards are DVS 3101-1 for the determination of the tensile strength and DVS 3101-2 for the four point bending strength. The investigation of ceramic-ceramic and CMC-CMC joined components showed that shear and flexural strength tests according to ASTM D 905 and ASTM C 1341-00 are very suitable for their characterization [13, 14]. Fig. 7 shows a joined O-CMC sample in a shear test set-up according to ASTM D 905. However, no tests are known yet with respect to cyclic loading or at high temperatures above 1000 °C.

Only few standardized test methods are known to evaluate joints by Non-Destructive Testing (NDT) methods. There is a high need to investigate the materials phases and flaws such as inclusions, pores and cracks. E.g., the dye penetrant test according to DIN EN 571-1 is suitable for fast optical quality control on cracks.

Computed Tomography (CT) is used as a powerful but costly test method, which makes it possible to investigate the microstructure and defects within the joint of complex components. Furthermore, other imaging methods like ultrasound or thermography are suitable for the qualification of the joint. Fig. 8 shows the result of CT analyses of joined SiSiC tubes with flanges as sub-components performed with different glass brazings.

For ceramic heat exchangers or hot gas lines, a high gas tightness is required. Since hot process gases often are corrosive, the joints also require chemical resistance. Only form fits and adhesive bonds provide sufficient mechanical stability and sealing properties in such severe environments. The gas leak tightness test according to DIN EN 1779 is suitable for the qualification of gas tight joints at ambient temperature. According to [15], several glass brazing systems were used to join ceramic capsules made of SSiC cohesively by using the laser technology. The focus of this application is on heat recovery processes in power generation.

The high temperature testing of the leak tightness at Fraunhofer Center HTL was qualified with tube-to-flange bonds shown in Fig. 8a. For that purpose the joint is placed in a horizontal tube furnace, operating at temperatures >1000 °C, whereas the opposite end of the tube is cooled and coupled to the gas testing system.

Potential applications of joined ceramics

The use of joining processes in ceramic industry has a wide range of advantages. The range of applications can be increased to large and complex dimensions. Additionally, the realization of joint layers either with electrically conductive or insulating properties increase the application spectrum. The joining of different materials such as ceramic-to-metals or to Ceramic Matrix Composites (CMC) can also be realized. Tab. 1 shows possible applications of joining technology in several branches. In the electrical industry ceramic connections based on metal brazings have already been established. In the metal industry and in the field of gas turbines joined ceramic components are less frequently used.

The use and transport of hot process gases is crucial for energy saving processes

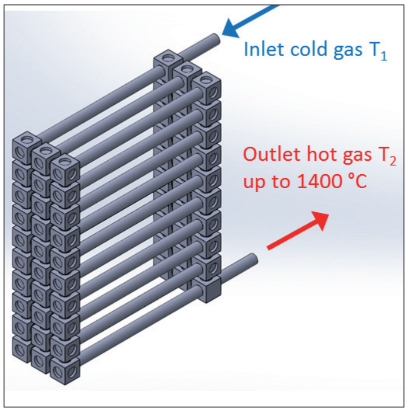

Fig. 9
Sketch of a high-temperature heat exchanger made of SSiC for the heat recovery at temperatures up to 1400 °C

Type	Oxide ceramics		Nonoxide ceramics
	Monolithic ceramics	CMC	Metal
Type of brazing	Glass / ceramic brazing	HT adhesive bonding	Active brazing
Joining geometry	Adhesive bond	Form closure	Force closure
Procedure	Screen process	Brush	Dosage
Joining method	Laser technology	High temperature furnace	Other joining method
Analysis	Analytical method / test technique CT, ultrasound, thermography, terahertz, high and room temperature mechanical testing, and gas tightness		

Fig. 10
Available joining technology at the Fraunhofer Center HTL

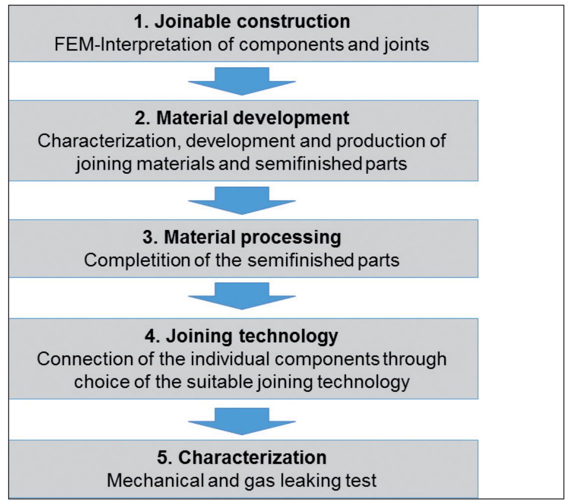

Fig. 11
Steps for the development of joining processes for ceramics and CMC

in the future. Energy efficiency can be improved in power generation and thermal processing by the use of high-temperature heat exchangers, hot gas fans and hot gas lines. The economic substitution of metals by ceramics or CMC requires an appropriate design and improved high-temperature properties. In order to increase the efficiency through higher process temperatures, the use of hot gas resistant ceramic materials with the aid of joining techniques is becoming increasingly important [16].
The use of ceramic heat exchangers is an innovative technology that can contribute to energy-efficient production in heat treatment processes at temperatures >1200 °C. However, the technology is only feasible by the use of gas-tight ceramic components and cohesive joining processes. Fig. 9 shows a schematic sketch of a joined, non-oxide ceramic tube type heat exchanger for temperatures up to 1400 °C. According to the current state-of-the-art, heat exchangers made of SSiC components are used for chemical engineering in corrosive environments at T<900 °C. [17]

Development of the joining technology within the project EnerTherm

Efficient production technologies are characterized by low energy costs and low emissions. Particularly, during chemical, ceramic and metal processing, there is a great demand to save energy due to the high-temperature production processes. There is an increasing demand to improve process stability and to decrease the number of rejected parts. Within the project EnerTherm “Energy Efficiency of Thermal Processes” funded by the Bavarian Ministry of Economic Affairs (BMWi), ceramic components are developed for high-temperature furnaces. In order to significantly improve the energy efficiency of existing and new production plants, lightweight components made of monolithic ceramics or CMC are required.
Possible target components are high-temperature heat exchangers, high-temperature fans, carrier structures or high-temperature measuring devices. Various joining technologies are investigated for the production of geometrically complex ceramic structures. Fig. 10 gives an overview of the applied techniques and methods for joining ceramic materials at the Fraunhofer Center HTL. With the help of this research project, a reproducible joining process for oxide and non-oxide ceramics and CMC could be established, comprising five individual steps (Fig. 11).
The focus was on monolithic ceramics such as SiSiC or SSiC for the use as high-temperature heat exchangers and composite ceramics based on Oxide Ceramic Matrix Composite (O-CMC) for the construction of high temperature lightweight structures [18]. A glass-ceramic brazing

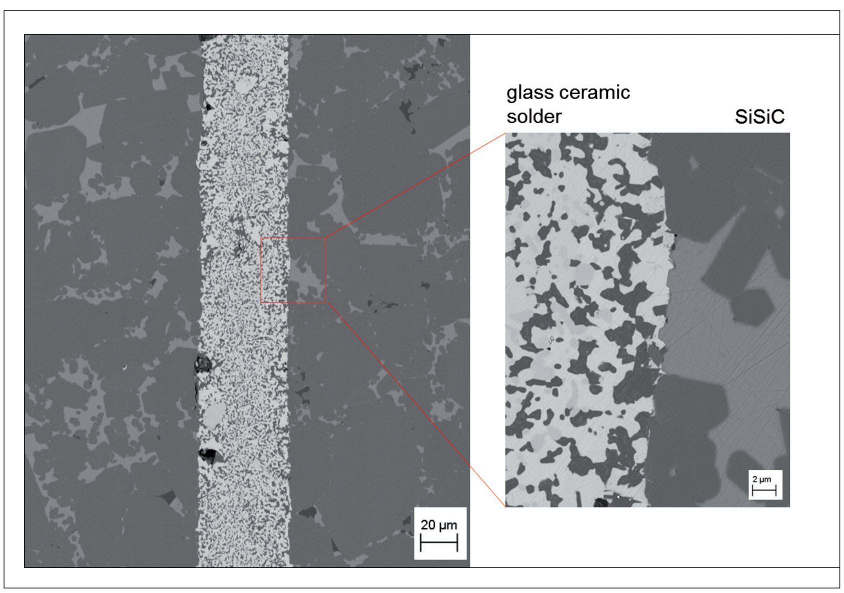

Fig. 12
Joints of SiSiC with a glass-ceramic solder performed by furnace-joining, microstructure recorded with SEM

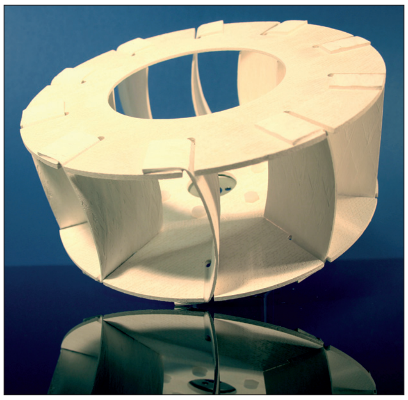

Fig. 13
Joined high-temperature impeller made of O-CMC for application temperatures up to 1200 °C (heigth = 100 mm, diameter = 300 mm, weight = 1,15 kg)

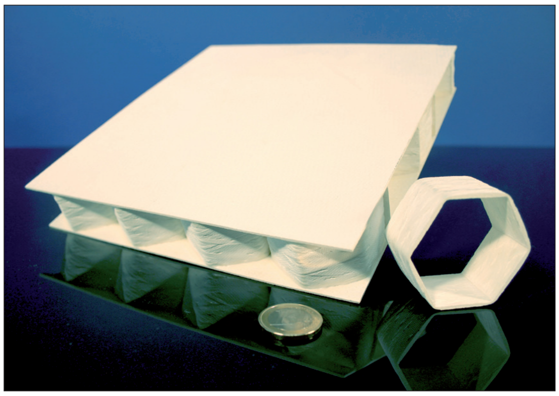

Fig. 14
Joined O-CMC high-temperature sandwich element with comb structure (height = 30 mm width = 200 mm)

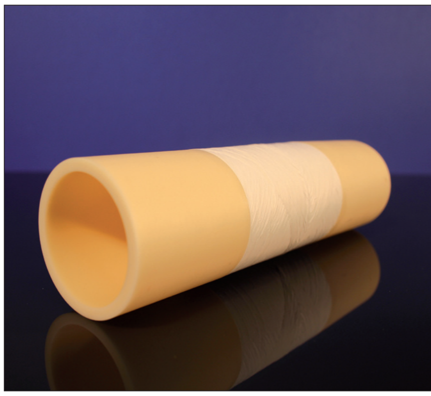

Fig. 15
Joined hybrid tube made of O-CMC and Al_2O_3 liners for application temperatures up to 1200 °C

was developed as joining material, which remains thermally stable up to 1400 °C due to its high crystallized fraction. Bending strength tests at room temperature and 1100 °C resulted in values between 50–100 % of the mechanical stability of the base ceramic materials.

Fig. 12 shows SEM micrographs of a Si-SiC joint with a glass-ceramic brazing. The size and number of defects in the joint determine the total strength of the composite. Only small amounts of pores and cracks can be observed in the SiSiC hybrid structure, which effects a high strength and good sealing of the joint. The low gas leakage was confirmed by investigations in a helium leak test according to DIN EN 1779 where a leakage rate of $1 \cdot 10^{-7}$ mbar·l/s was determined.

Due to the existing CMC manufacturing processes, only geometrically simple components can be produced preferentially by pressing, winding and lamination. Joining technologies are used for the realization of geometrically complex lightweight structures. Fig. 13–15 show examples of joined components such as a high-temperature impeller and a carrier structure, both made of O-CMC, and high-temperature tube made of O-CMC and Al_2O_3 liners.

Summary

Ceramic-to-ceramic joints increase the portfolio of ceramic applications. They are realized if complex shapes or materials combinations are required. Joining is mainly executed by using glass solders or metal brazings with good wetting behaviour and adapted coefficients of thermal expansion with regard to the base materials. The materials bonding is often completed by form or force closure in order to obtain high mechanical strength in the joint.

By enhancements of the joints and the holistic technological approach within the last years, it is now possible to realize adhesive joints of monolithic ceramics and CMC. All joinings can be carried out by using laser technology or high-temperature furnaces. Both methods lead to comparable results regarding their mechanical properties. Joining with lasers offers the great advantage of having the high thermal load locally in the joining area and having short joining times within some minutes only.

At Fraunhofer Center HTL joining technology was demonstrated with ceramic components for heat technology. Basic investigations were conducted within the project EnerTherm.

The key was the development of adaptable glass-ceramic brazing systems. Due to their high cristallized fraction these joints allow application temperatures of up to 1400 °C. The ceramic to ceramic joints have approved high gas tightness, so that the joined components are suitable for hot gas atmospheres.

References

[1] Brevier Technische Keramik. Lauf 2003, 186–204

[2] Salmang, H.; Scholze, H: Keramik (Teil I). Berlin, Heidelberg, New York 1982, 947–950

[3] Sauerzapfe K.; Wampers, H.: FEA – a key technology for integrative design of ceramic-metal-assemblies. CERAMIC APPLICATIONS **2** (2014) [1] 28

[4] Luhn, R.; Jahn, S.: Hochtemperaturkleben keramischer Werkstoffe. DKG-Tagungsband Fügen von Keramik: Grundlagen – Verfahren – Anwendungen. Erlangen 2012, 35–36

[5] Lippmann, W.; Börner, F.D.; Hurtado, A.; Fügen von keramischen Bauteilen durch laserinduziertes Löten – Neue Simulationstools erschließen eine innovative Technologie. Abschlussbericht zum BMBF-Förderbericht Cerajoin, Förderkennzeichen 03X0507F, 2010

[6] Wielange, B.; Schüler, H.: Löten keramischer Werkstoffe, Handbuch Technische keramische Werkstoffe. J. Kriegsmann (ed.), Essen 1998, Kap. 3.9.2.0, p.1 ff.

[7] Jacques, E.; et al.: Preliminar results on joining of thin SiC/SiC composites by silicides compounds and local heating. Ceram. Mater. for Energy Applic. **32** (2011) [9] 73–83

[8] Halbig, M.C.; et al.: Characterization of silicon carbide joints fabricated using SiC

particulate-reinforced Ag-Cu-Ti alloys. Ceramics Int. **39** (2013) 4151–4162
[9] Kopp, N.; Bobzin, K.: Reactive air brazing – Löten von Keramik-Keramik- und Keramik-Metall-Verbunden an Luft. Aachen 2012
[10] Hubert, D.; et al.: Selective laser soldering by means of glass solder for high temperature applications. 9th Int. Conf. LÖT 2010
[11] Baber, J.; Raether, F.: Production of oxide ceramic coatings on glass by laser sintering. Glastechn. Ber. Glass Sci. Technol. **73** (2000) 211–215
[12] Rizzo, S.; et al.: Joining of C/SiC composites by spark plasma sintering technique. J. Europ. Ceram. Soc. **34** (2014) 903–913
[13] Ferraris, M.; Hinoki, T.; Katho, Y.: Torsional shear strength of silicon carbide components pressurelessly joined by a glass-ceramic. Int. J. of Appl. Ceram. Technol. **9** (2012) [4] 786–794
[14] Salvo, M.; et al.: Shear and bending strength of SiC/SiC joined by a modified commercial adhesive. Int. J. of Applied Ceram. Technol. **9** (2012) [4] 778–785
[15] Börner, F.D.; Lippmann, W.; Hurtado, A.: Laserfügen keramischer Wärmerohre zur Energiegewinnung aus einem allothermen Vergasungsprozess. cfi/Ber. DKG **90** (2013) [10] D19–D27
[16] Jäger, H.: Ceramic Composites 2050. Roadmap der Abteilung Ceramic Composites des Carbon Composites e.V., 2012
[17] Meschke, F.: Vollverschweißter Wärmeübertrager aus SiC-Keramik. DKG-Tagungsband Fügen von Keramik: Grundlagen – Verfahren – Anwendungen. Erlangen 2012, 8
[18] Gadelmeier, C.; et al.: Adhesive bonding of ceramics for complex ceramic parts in high temperatures furnaces. Mater. Sci. Forum, Vol. 825–826, (2015) 279–286