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## Advancement of Cellular Ceramics Made of Silicon Carbide for Burner Applications

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**Abstract.** Lower emissions of CO and NO<sub>x</sub> as well as a higher power density were observed in combustion processes performed in porous media like ceramic foams. Only a few materials are applicable for porous burners. Open-celled ceramic foams made of silicon carbide are of particular interest because of their outstanding properties. Two different SiC materials have been investigated, silicon-infiltrated silicon carbide (SiSiC) and pressureless sintered silicon carbide (SSiC). The oxidation behaviour of both has been characterized by furnace oxidation and burner tests up to 500 h operating time. Up to a temperature of 1200 °C SiSiC exhibited a good oxidation resistance in combustion gases by forming a protective layer of silica. High inner porosity up to 30% in the ceramic struts was found in the SSiC material. Caused by inner oxidation processes the pure material SSiC allows only short time applications with a temperature limit of 1550 °C in combustion gases. An increase of the lifetime of the SSiC foams was obtained by development of a new SSiC with an inner porosity of less than 12%. The result was a considerable reduction of the inner oxidation processes in the SSiC struts.

**Keywords.** porous burner; ceramic foam; silicon carbide

### 1. Introduction

The development of burner technologies has been driven by the demand of higher radiant heating rates on the one hand and tightened environmental constraints, which long for minimal pollutants emission, on the other hand. Recently, several investigations have shown the advantages of the porous burner technology. The porous burner offer low emissions of CO, NO<sub>x</sub> and HC, high energy efficiency and high power density [1]. The advantage of the combustion taking place in a porous media is the preheating of the fuel/air mixtures. This raises the flame temperature, reduces emissions and stabilizes the flame [2]. Therefore a cellular material with a high thermal conductivity is required, which transports the heat from the reaction zone upstream to the incoming gases.

Porous burners have been investigated for several different applications. Although they are of great economical and ecological interest for domestic gas heaters, they are still in an early stage of development. Similar porous structures are in use as afterburner in solid oxide fuel cell systems [3]. The most wanted features are wide modulation ranges and the possibility to feed the burner with gas mixtures of different calorific value. Porous burners are also integrated in industrial drying processes. IR-emitter plates in paper machines, for example, can be made of ceramic foam of siliconized silicon

carbide. The combustion of gases with lean or ultra-lean fuel/oxidant ratios can be improved with cellular structures in combination with catalysts. The presence of a catalyst, in form of a washcoat or placed in the entire matrix, promotes the conversion of climate-destructive methane of landfills, for example.

However, the combustion inside a porous structure is connected to special requirements. The porous structures must resist thermo mechanical stress caused by the high gradients in temperature, especially during start and modulation of the burner [4]. Depending on the fuel type, fuel/oxidant ratio and the pressure, very different corrosion effects can occur [5]. Hot wet atmospheres up to 17% water vapour and velocities of  $1 \text{ ms}^{-1}$  should be regarded at least. This entails very high requirements on the corrosion resistance, because most burners are used for long term applications, e. g. for more than 1000 h, in some cases up to 10 000 h. Additionally, according to the Stefan-Boltzmann-Law they should be characterized by high emissivity at highest operating temperatures (about  $1200 \text{ }^\circ\text{C}$ ), since the enhancement of the heat radiation obeys with the fourth power on temperature and a linear dependence on material emissivity. Furthermore, simple components made of cheap materials are needed for domestic or automotive mass application.

## 2. Silicon carbide for porous burners

### 2.1. Materials for high-temperature applications

With reference to the requirements mentioned above, the number of materials suitable for application in combustion environments is very small. Some important materials for high-temperature applications are listed in table 1.

**Table 1.** Properties of important high-temperature bulk materials

		$\text{Al}_2\text{O}_3$	Cordierite	$\text{ZrO}_2$	SiC	Fe-Cr-Al
Density	$\text{gcm}^{-3}$	3.98	2	6.1	3.1–3.2	7.3
$\text{TC}^{\text{I}}$	$\text{Wm}^{-1}\text{K}^{-1}$	5–30	1–3	2–5	80–160	13.5–17.7
Specific heat capacity	$\text{Jg}^{-1}\text{K}^{-1}$	0.9–1.0	0.8–1.2	0.5–0.6	0.7–0.8	0.5–0.8
$\text{CTE}^{\text{II}}$	$10^{-6} \text{ K}^{-1}$	8	2–6	10–13	4.7–5.2	10.1–13.2
$\text{TSP}^{\text{III}} \text{R1 } \sigma/\text{E}*\alpha$	K	100	650	400	230	
$\text{TSP}^{\text{III}} \text{R2 } \sigma*\lambda/\text{E}*\alpha$	$10^{-3} \text{ Wm}^{-1}$	3	2	1.5	28	
$\text{TAL}^{\text{IV}}$	$^\circ\text{C}$	1900	1300	1800	1600	1330
Corrosion resistance		+	-	o	++	+
Price		o	+	--	-	-

<sup>I</sup> thermal conductivity 20–1000  $^\circ\text{C}$ , <sup>II</sup> linear coefficient of thermal expansion 20–1000  $^\circ\text{C}$ , <sup>III</sup> thermal shock parameter, <sup>IV</sup> thermal application limit (in air, short time)

For high application temperatures up to  $1900 \text{ }^\circ\text{C}$   $\text{Al}_2\text{O}_3$  was used in porous burners [6], restricted by the low thermal shock resistance. In contrast due to the excellent thermal shock resistance, cordierite was used in spite of the application temperature being limited at  $1300 \text{ }^\circ\text{C}$  [7]. Several investigations have been done on  $\text{ZrO}_2$  [8]. The properties of pure  $\text{ZrO}_2$  were found to be very interesting, but the phase change from tetragonal to monoclinic at  $1170 \text{ }^\circ\text{C}$  is connected with a change in volume (5–8%). To prevent this the tetragonal phase  $\text{ZrO}_2$  can be stabilized by sintering additives like yttrium, for example, accepting a lowering of the bearable temperature.

For applications requiring high thermal shock resistance and lower temperatures (up to  $1330 \text{ }^\circ\text{C}$ ) Fe-Cr-Al alloys are very interesting. Furthermore, composite materials and mullite were investigated as candidates for porous burners.

However, with the specified requirements silicon carbide was found to be the best compromise between application temperature, corrosion resistance, thermal conductivity and thermal shock

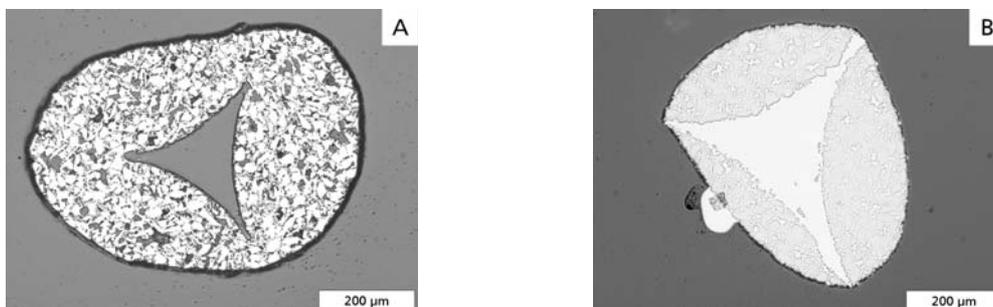
resistance. Additionally the low CTE and the high emissivity and strength at high temperatures are reasons for the usage of SiC for high temperature applications in combustion environments.

## 2.2. Types of silicon carbide

For high-temperature applications open-celled SiC can be made of pressureless sintered (SSiC), reaction-bonded silicon infiltrated (SiSiC) and recrystallized (RSiC) silicon carbide. According to the different sintering mechanisms and bonding types, various properties can be achieved. The cellular structure itself can be realized by replication techniques. Foams, for example, are created by coating polymeric templates with ceramic slurry, drying, pyrolysing and afterwards sintering or siliconizing. Additionally non-isotropic components can be manufactured with technical textiles, e.g. woven and knitted structures. In all cases flowable slurries are required to achieve a homogenous coating. On this account the solids content in the slurries is restricted. This affects the shrinkage and densification. While bulk SSiC, for example, can reach up to 99% of the theoretical density, ceramic foams of SSiC commonly have an inner porosity of about 30%. The amount of inner porosity of the recrystallized SiC is even higher. Although the high amount of porosity is connected with inner oxidation, a very good chemical stability is obtained in case of the pure or nearly pure materials like RSiC and SSiC recommending these materials in high-temperature applications.

Silicon infiltrated silicon carbide (SiSiC) is fabricated as a shrinkage-free but dense material, where the coarse primary particles are bonded by the reaction of silicon metal melt with carbon to secondary  $\beta$ -SiC. Remaining pores are filled by silicon. The metallic Si causes very high mechanical strength but low corrosion resistance. The porous burner application is limited at about 1350 °C (short time).

Based on the mentioned characteristics of the SiC-types, only SSiC and SiSiC were investigated in the following furnace and burner rig tests. The typical microstructure of the two materials is illustrated in the cross-sections of two struts in figure 1. The porosity was detected by light microscopy.



**Figure 1.** Cross-section of ceramic foam struts made of SSiC (A) and SiSiC (B).

## 3. Theoretical oxidation behaviour of SiC and experimental methods

The oxidation behaviour of the SiC itself depends on the intrinsic properties of the material and the environment it is used in. As a nonoxide ceramic SiC will always show a reaction with oxygen or water vapour in the hot and wet combustion atmosphere. Basically three reaction types have been identified. The formation of a protective silica layer on the surface, which serves as an effective oxygen diffusion barrier, leads to a parabolic oxidation kinetic [9]. With increased thickness of the layer the diffusion paths of the oxygen become longer and the reaction with the substrate is decelerated. In contrast to this passive oxidation a continuously linear weight loss can occur at very high temperatures, high water vapour and high gas velocities. In this case no protective silica layer exists, as only volatile reaction products are formed. The third reaction type is the so-called para-linear kinetic. A silica layer is formed, but depending on the environmental conditions (temperature, water vapour, gas velocity) it is continuously degraded by the formation of volatile silicon hydroxides ( $\text{Si(OH)}_4$ ) [10]. In burner application the material should show parabolic or at least para-linear oxidation behaviour.

The long-term oxidation stability of cellular ceramics is closely related to the amount of inner oxidation. Especially under thermo-cyclic conditions the different coefficients of thermal expansion of SiC and the oxide inside the strut, causes crack formation and accelerate the degradation of the mechanical foam stability. The attack by hot and wet atmospheres at the outer surface and inside the struts, strongly limits the lifetime. To counteract this effect the goal of the material development is to realise open-celled foam with homogeneously coated struts and a closed inner porosity of about 5%. By variation of the slurry's composition, only with regard of temporary organic binder and powder mixtures, a new generation of SSiC-foams was developed. Instead of 30% inner porosity a microstructure with less than 12% inner porosity was achieved.

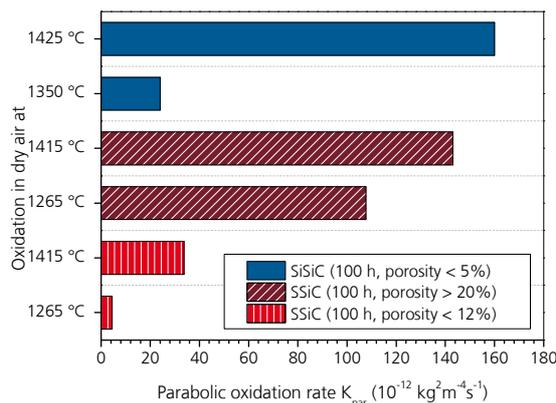
To investigate the oxidation behaviour of the SiC foams, tests in muffle furnace and two different burner rigs were performed. The samples with 10–20 pores per inch (ppi) and 90% cell porosity were characterized by weight change and analysis of the microstructure. The burner rig used samples with diameter of 135 mm and a high of 15 mm. Gas velocities of 2.4–3.5 ms<sup>-1</sup> could be adjusted. In the lifetime test rig cylindrical samples of 65 mm diameter and a high of 40 mm have been used, with gas velocities of 1.7–2.3 ms<sup>-1</sup>. The parabolic oxidation rate was determined using the Deal-Grove model [9] as displayed in equation (1). Herein *t* is the time of oxidation, *m* is the calculated slop of the linear fit of the weight gain per *t*<sup>-1/2</sup> and *s* the surface area of the sample.

$$K_{par} = m^2 s^{-2} t \quad (1)$$

## 4. Results and Discussion

### 4.1. Results of the furnace tests

The results of the furnace oxidation are displayed in figure 2. During oxidation in dry air both, SiSiC and SSiC foams, showed parabolic oxidation behaviour with formation of a protective silica layer. The lowering of the amount of inner porosity by the new developed SSiC foams reduced the surface area available for oxidation. This was the reason for the significant decrease of the oxidation rate. In comparison, a bulk SSiC reported by Costello [11] oxidized in dry air at 1400 °C showed an oxidation rate of about 4.7 x 10<sup>-12</sup> kg<sup>2</sup>m<sup>-4</sup>s<sup>-1</sup>, which is very close to the results of the SSiC foam. The lower the amount of open inner porosity, the more the strut material behaved like nearly dense bulk SSiC.



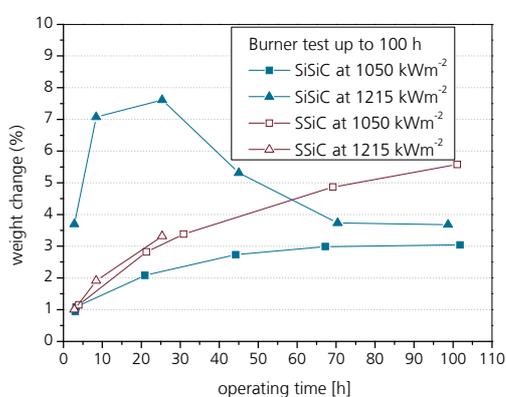
**Figure 2.** Comparison of the parabolic oxidation rates of SiSiC and SSiC with two different inner porosities.

While the oxidation rate of SSiC continuously increased with higher temperature in dry air, the oxidation rate of the SiSiC at temperatures above 1400 °C raised excessively. The reason for this was the partial melting of the free silicon out of the SiSiC structure, turning the dense material into a porous one.

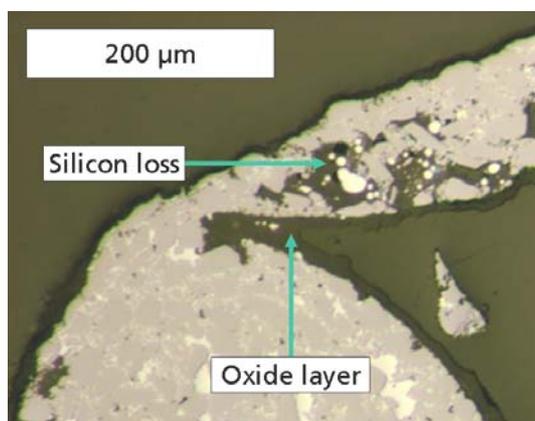
#### 4.2. Results of the burner rig test

SiSiC (inner porosity < 5%) and conventional SSiC (inner porosity 30%) samples were investigated at  $1050 \text{ kWm}^{-2}$  and  $1215 \text{ kWm}^{-2}$ . The results are demonstrated in figure 3. For low power density the SiSiC displayed the smallest weight gain with a parabolic oxidation kinetic. The amount of weight gain was found to be approximately half as high as for SSiC under the same conditions. However, for  $1215 \text{ kWm}^{-2}$  the oxidation behaviour changed into a para-linear kinetic, connected with a significant degradation of the micro structure as shown in figure 4.

The oxidation rate of the SSiC was higher than that of SiSiC. However, the parabolic oxidation rate was found to be stable even at the higher power density. The lifetime of the SSiC foam with about 30% inner porosity was shortened by the inner oxidation, which led to mechanical failure after less than 2000 h operating time.



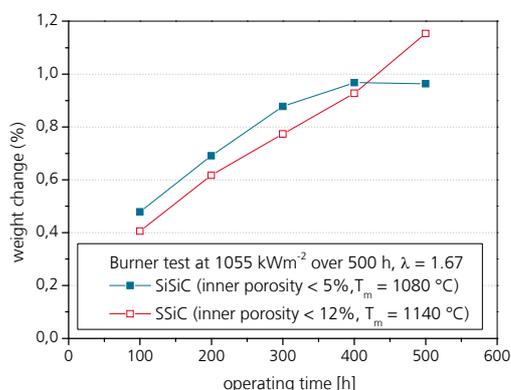
**Figure 3.** Weight change during burner rig test of SiSiC and SSiC foam samples at  $1050 \text{ kWm}^{-2}$  and  $1215 \text{ kWm}^{-2}$  with  $T_{\text{max}}$  lower than  $1200 \text{ }^{\circ}\text{C}$ .



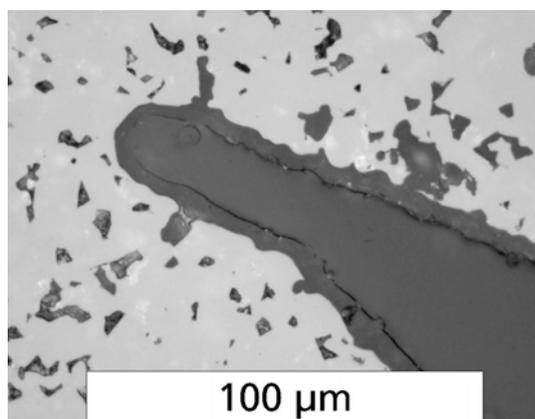
**Figure 4.** Cross-section of a SiSiC foam strut oxidized in burner rig over 100 h at  $1215 \text{ kWm}^{-2}$ .

#### 4.3. Lifetime burner tests with new developed SSiC foams

Some results of the burner tests are described in figure 5 and 6. During the furnace tests the benefit of the optimized lower inner porosity in the new developed SSiC foams under dry air was investigated.



**Figure 5.** Comparison of the weight change of SiSiC and new developed SSiC (porosity < 12%) during 500 h oxidation at power density  $1055 \text{ kWm}^{-2}$  and  $\lambda = 1.67$ .



**Figure 6.** Optical analysis of the microstructure of a SSiC strut cross-section from burner test (operating time 500 h, power density  $1055 \text{ kWm}^{-2}$ ,  $T_m = 1140 \text{ }^{\circ}\text{C}$ ).

In long time burner tests up to 500 h operating time, the new developed SSiC displayed similar weight gain and oxidation rate as the SiSiC (figure 5). In contrast, the weight gain of the conventional SSiC (30% inner porosity) in the first burner rig test, was found to be about twice as high as that of SiSiC. This different behaviour was attributed to the lower inner porosity in the microstructure of the new developed SSiC (< 12% inner porosity). In this case the oxidation was found to occur just on the outer and inner surface of the struts. The SiC was protected by the silica layer even after long operating times at elevated temperatures.

Similar to the short time burner rig test the temperature during the test did not reach more than 1200 °C. Under these conditions the SiSiC displayed good oxidation stability, since no melting of free Si occurred. However, the SSiC has a great advantage concerning the chemical resistance of the nearly pure material. Moreover it tolerates temperature peaks up to 1550 °C in combustion environment.

For long term stability in combustion environments a nearly closed inner porosity is required. In the analysis of the microstructure was found, that only a very small amount of inner oxidation occurred (figure 6). A homogenous and dense silica layer on the outer and inner strut surface was formed which is the prerequisite for parabolic oxidation behaviour and a long time stability. Based on these results the new developed SSiC foam is a promising material for high temperature application.

## 5. Summary and conclusion

The concept of combustion in porous media is connected to special requirements on the applied materials. It was pointed out that silicon carbide is the best choice for porous burner application. The differences of the two types, pressureless sintered and siliconized silicon carbide in the oxidation behaviour has been investigated with furnace and burner tests.

SiSiC showed good oxidation resistance at temperatures up to 1350 °C in dry air. The limitation of the material was given by the out-melting of free silicon. Although the melting point of pure silicon lies over 1400 °C impurities can lower this temperature significantly. The temperature peaks occurring during start or modulation of the porous burner entail the accelerated degradation of the material.

SSiC foams were found to be restricted by the amount of inner porosity. With the new developed SSiC foam material an inner porosity of less than 12% was achieved. By furnace test the influence of the inner porosity was proved. Promising properties of the SSiC material have been shown by comparison of the new SSiC foam with SiSiC foam in burner lifetime test for 500 h operating time.

On the basis of these results it was concluded that open-celled ceramic foam made of SSiC seems to be an excellent alternative for SiSiC foams, because of the good resistance against corrosion and high temperatures of the nearly pure material.

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