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SiSiC Heat Exchangers for Recuperative Gas Burners with Highly Structured Surface Elements

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Heat exchangers are used in gas burners as recuperators. Their efficiency is proportional to their surface which is usually limited by the burner length and diameter. Ceramic recuperators used nowadays in industrial burners are tubular. We studied and developed a new generation of ceramic heat exchanger with the final goal of increasing the efficiency or, at similar efficiencies, of reducing their size and weight. A commercial SiSiC heat exchanger component was used to guarantee safety and robustness. The use of structured textile geometries followed by their conversion into a ceramic is the main innovation of this work.

Introduction

Industrial production is a major consumer of energy. Energy consumption in Europe in the industry sector represents about 30% of the overall fuel consumption in all sectors. One can assess that ca. 60% of the industrial fuel consumption is spent for industrial process heating, and furthermore, more than 80% of the process heat is covered by fossil fuels (Source: German Federal Ministry of Economics and Technology). An efficiency increase in process heating of only a few percent would result in a significant reduction in energy consumption and carbon footprint.

Industrial branches of high energy consumption can be found in the metal industry (steel and alloys), glass industry, and ceramics production and processing. Because most of the relevant processes are operated at a high temperature, heat recovery becomes essential in terms of efficiency. The most common way to recover heat is by preheating the air or other gasses with hot effluent gasses. The so-called recuperative or regenerative heat exchanger systems,¹ which may be integrated in the burner assemblies, are commonly used for this purpose.

The use of burners equipped with recuperators results in significant efficiency increase. However, their efficiency is proportional to the heat exchanger surface which is limited by the overall volume and burner length restrictions and by the required heat exchanger and its material limitations, which will be discussed later on.

The highest efficiency can be reached using regenerative burners. However, the complexity of regenerative burner systems is higher, due to the discontinuous alternately mode of operation utilizing several regenerator matrices.

In all cases, it can be presumed that a 100 K decrease in the flue gas temperature after heat recovery results in an estimated reduction in fuel consumption of about 5% (Fig. 1).

Heat exchangers used nowadays are mostly made of high temperature alloy steel with corresponding temperature limitations. The use of ceramic materials brings the following advantages.

- operation and heat recovery at the higher temperatures,
- higher corrosion resistance in many harsh environments.

On the other hand, they present the following drawbacks:

- lower amount of heat can be recovered in comparison with the metal recuperator, due to manufacturing restrictions,
- fragile mechanical behavior, which implies that components are always over-dimensioned,
- restrictions due to dust transported with the gas flows,
- heat recovery devices should be directly integrated into the thermal processing facility, to

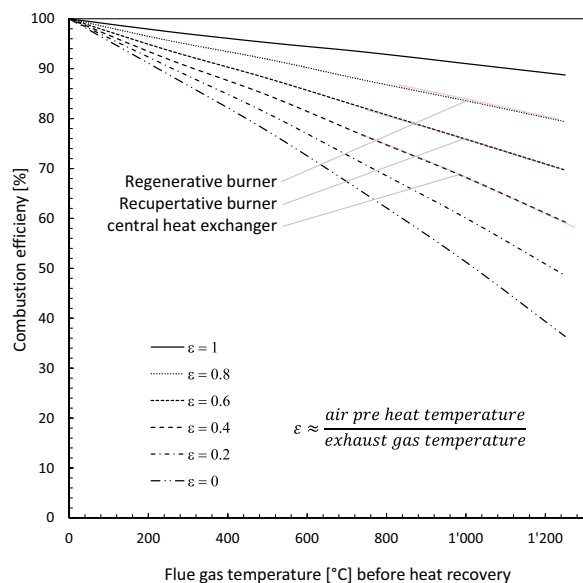


Fig. 1. Combustion efficiency with respect to the exhaust gases temperature before heat recovery; Experimental results for different types of heat recovery ².

minimize thermal losses and utilize the energy in an immediate manner.

Despite the obvious benefits of ceramic heat exchanger components for heat recovery at high temperature applications, the penetration of such technologies in industrial furnaces is restricted by comparatively higher prices and larger size of such components. Relatively simpler ceramic heat exchanger geometries with large dimensions are used up to now, due to several manufacturing and operational obstructions. Increasing their efficiency will lead to decrease their dimension overcoming the above-mentioned economical problems.

Ceramic recuperators used nowadays in industrial burners are tubular. Common recuperator lengths are in the order of 0.5 m and hence longer than the thickness of a typical furnace wall. Heat enhancement with structured surfaces is limited by the associated ceramic production technologies. Given these technological obstructions and the dimensional limitations, the level of heat recovery is restricted. State of the art recuperative burners show an air preheating and flue gas temperature level after the recuperator in the range of 500–700°C resulting in heat losses through the flue gas of about 25–35%.²

Within the CEREXPRO project (§ 5), we studied and developed a new generation of ceramic heat exchangers for high temperature heat recovery having the goal of

increasing the efficiency or, due their size and weight reduction at similar efficiencies, of lowering their price. The use of structured geometries typical of the textile industry followed by their conversion into a ceramic is the main innovation of this project. Although the principle is not new, what makes it innovating is the unusual combination of such different production fields which in turn leads to a high design flexibility of the heat exchanger.³

To facilitate the development, this approach was applied on an existing robust SiSiC heat exchanger component successfully applied in industrial furnaces. This approach guaranteed application safety and robustness. Due to the improved efficiency, we proved that a significant size reduction or, alternatively, an increase in the heat recovery level can be achieved because of the higher heat transfer rate of the heat exchange elements.

This paper is divided into four sections. The thermo fluid dynamic behavior of the structured surface is studied in section 'Design'. Section 'Processing' deals with the textile structure development and production and its conversion process from textile-derived materials into a thermal shock resistant ceramic (e.g., silicon infiltrated silicon carbide – SiSiC). Finally, section 'Prototype' presents a prototype of SiSiC heat exchanger for recuperative gas burners with SiSiC structured surface elements manufacturing and field testing.

Design

The design of the highly structured SiSiC surface elements must meet the requirements of robustness and adaptability to an existing heat exchanger. It was also taken into account what textile and ceramic state of the art technology could provide. An arc (or loop) of 180° was selected as the basic element for the structured surface. The aim was to reach a higher heat transfer coefficient by increasing the transferring surface and also by utilizing the bigger dispersion and thermal boundary layer structures due to the resulting turbulent flow characteristics. Figure 2 shows different possible arrangements of such loops on a flat surface.

Numerical simulations using commercial CFD packages were performed for geometries in Fig. 2 to calculate the flow characteristics and analyze the increase in heat transfer and pressure drop, that is, friction factor coefficient. To reduce computation time, a planar arrangement was selected as reference geometry. To simulate the fluid flow and heat transfer process, a

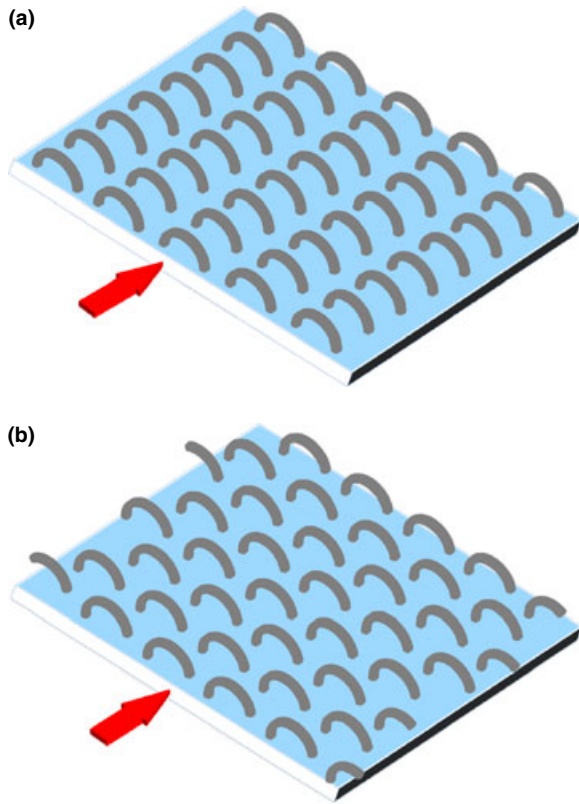


Fig. 2. Selected loops design (Radius $R = 8$ mm and diameter $D = 1$ mm): (a) Geometry 01 (3600 Loops/m²). Distance between Loops: 12 mm in flow direction and 24 mm perpendicular to the flow. (b) Geometry 02 (7200 Loops/m²). Distance between Loops: 8 mm in flow direction and 18 mm perpendicular to the flow.

typical repeating cell was defined. This cell was used to represent the entire structure by geometrical symmetry to further decrease the computational efforts.

The governing equations were solved using finite volume methods assuming a forced convection situation. Boundary conditions of the cubic cell were as follows: constant temperature at the bottom surface (blue in Fig. 2), which must be lower than the inlet temperature of the air flow (red arrow in Fig. 2) to generate a heat flux from the hot air to the cooled heat exchanger surface. A heat transfer coefficient of approximately $\alpha = 82$ W/m²/K of the flat solution was used as benchmark for evaluation of the new structured surfaces, while the operating conditions were kept constant.

Pressure drop, Nusselt number, and heat transfer coefficient were calculated at different Reynolds numbers. Figure 3 shows the ratio of the expected performance in comparison with the reference conventional

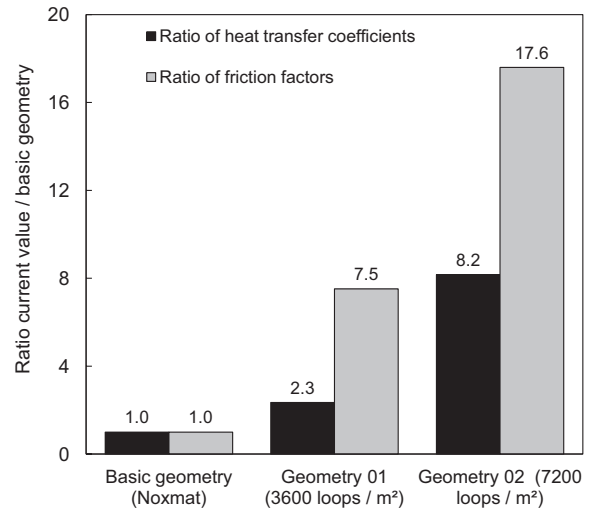


Fig. 3. Friction factor and heat exchange efficiency calculated for the different layouts.

recuperator burner at nominal conditions average (Reynolds number 5200).

These results indicate that the targeted recuperator design will result in a significantly higher heat recovery and slightly higher pressure losses, given the same burner size. On the other hand, keeping the same heat recovery, the burner length, and the pressure losses can be significantly reduced.

Processing

Textile Structure Assembly

Textile refers to any material made of interlacing fibers. Fabric refers to any material made through weaving, knitting, spreading, crocheting, or bonding that may be used in production of further goods In textile industry.⁴ Three processes were analyzed to realize a suitable structure as well as several fiber materials.

Nonwoven An assembly of textile fibers held together by mechanical interlocking in a random web or mat, by fusing of the fibers (in the case of thermoplastic fibers), or by bonding with a cementing medium. Initially, the fibers may be oriented in one direction or may be deposited in a random manner. Nonwoven was immediately abandoned because the proposed design could not be obtained with this technology.

- Weaving** Weaving is made by interlacing two yarns of similar materials so that they cross each other at right angles to produce woven fabric. The warp yarns run lengthwise in the fabric, and the filling threads (weft) run from side to side. Trials were performed, and samples produced on rapier weaving looms that were adapted to match the specification of the design (Fig. 2). Two weave drawings have been set corresponding to the different densities of loops, and thread count for base fabric has been determined according to the diameter of loops. A rapier loom was adapted to reproduce a sort of velvety texture (Fig. 4a). Velvet process uses several warp beams to balance the difference of absorbed warp length between base fabrics and loops and performs a weft thread or rails to maintain the loops height.
- Knitting** Knitting is a way to make a fabric by interlocking series of loops of one or more yarns. Two different types of knitting technologies exist: weft and warp knitting. Weft Knitting is a common type of knitting, in which one continuous thread runs crosswise in the fabric making all of the loops in one course. Different trials showed the interest to use in the building of the structure an additional yarn with thermo soluble properties (Fig. 4b).

The best performing material, both for weaving and ceramization, was a multifilament yarn made of polyethylene terephthalate (PET) fibers. Textile structure assembly had many objectives to comply with:

- structure geometry should comply with the design constraints,
- being the textile structure a scaffold on which deposit the ceramic layer, the thermo-physical behavior of its material under pyrolysis should be tuned properly,
- elastic modulus as well as yarn thickness and sizing should be sufficient to keep their geometry while replicating,

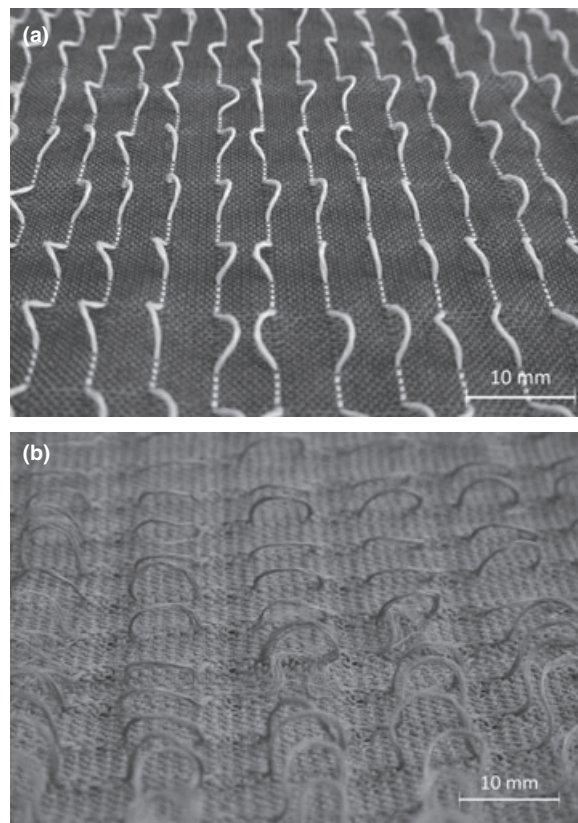


Fig. 4. Textile structures (Geometry 1) obtained by (a) weaving and (b) knitting.

- another extremely important requirement is that, as discussed later, the base fabric in contact with the SiSiC tube walls should be as loose and light as possible to minimize the formation of porosity onto the SiSiC tube (Fig. 5). Porosity is detrimental because it reduces heat transfer and acts as a crack initiator during thermal cycling.

The base fabric was assembled using PET yarns made of 120 filaments with a dtex 668 (the tex expresses the mass in grams per 1000 meters of a yarn). The loop structure was obtained with PET yarns made of 660 filaments with dtex 2004. As fibers selection was also dictated by the ceramization requirements, further details will be given in the next section. Indeed, there were opposite needs for the two technologies, for example, carbon fibers are interesting because they can be converted to SiC during LSI, but due to their rigidity, they are difficult to weave.



Fig. 5. 3D rendering, obtained by X-ray computed tomography, of highly packed base fabric, impregnated and processed, showing high internal porosity.

Textile Ceramization

In combustion environments, silicon carbide ceramics are, for their outstanding thermal and mechanical properties, among the best materials.⁵ In fact, they are commonly applied in high temperature furnaces because, if passive oxidation conditions are met,⁵ they withstand operating conditions for long time.⁶

Three processes were envisaged to fabricate highly structured SiSiC surface elements:

- Chemical vapor deposition (CVD)
- Polymer impregnation and pyrolysis (PIP)
- Liquid silicon infiltration (LSI)

Several process tests were performed, at the end, because of its industrial maturity and also because the commercial SiSiC tube is made with a similar technology (proven to be the best performer in a combustion chamber environment), LSI was adopted as manufacturing procedure. A thin (800 mPas) but, stable proprietary slurry made of α -SiC powders (fine and coarse), a plastic binder and an organic solvent were prepared. The textile structure was dipped into the slurry and hanged to drain the excess liquid. As the solvent was partially evaporated and the structure still tacky, it was placed on the SiSiC substrate as per Fig. 6a and further dried. The plate was then placed into a furnace and slowly pyrolyzed up to 1000°C, under inert atmosphere. The textile structure experienced a minor shrinking because of the mass loss

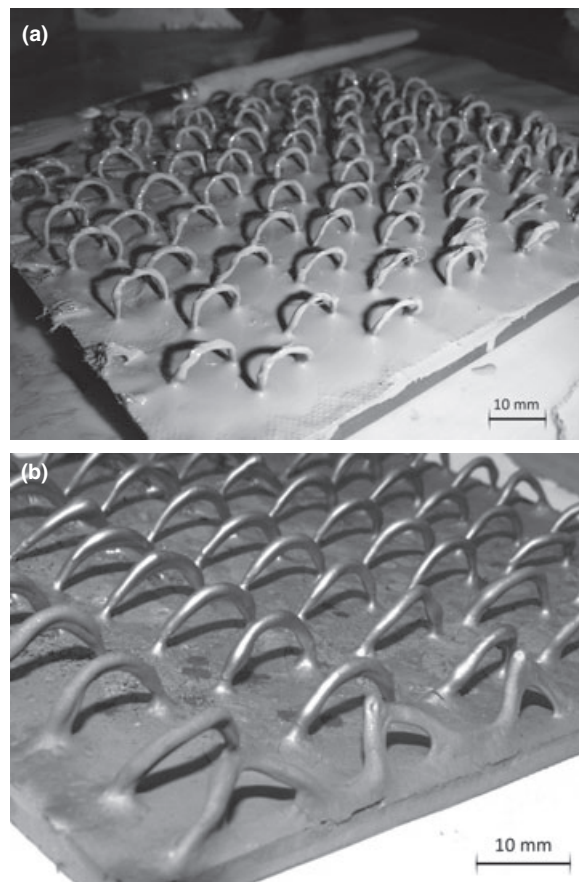


Fig. 6. (a) Flat sample preparation: Replicated structures (GEO2) are placed onto a SiSiC plate, (b) Final aspect of the plate (GEO1) after pyrolysis and LSI.

during pyrolysis, and some cracks could be observed. Cracks minimization was worth a long optimization work in which textile architecture of the base fabric and amount of slurry employed were carefully adjusted. After pyrolysis, the object was placed in contact with silicon flakes and LSI was performed at 1500°C under a 10^{-2} (mbar) residual pressure. The furnace was quickly brought to the process temperature, held at that temperature for 1 h, and left cooling naturally. A final flat sample is shown in Fig. 6b

A great effort was devoted to the selection of proper fibers, further called fugitive (Fig. 7a) or permanent (Fig. 7b), meaning that in one case, they would produce, respectively, a hollow or full space within the loop.

Examples of fugitive fibers are polypropylene (PP) and PET. On the other end, permanent fibers are those containing already a ceramic phase (e.g., C, SiC).

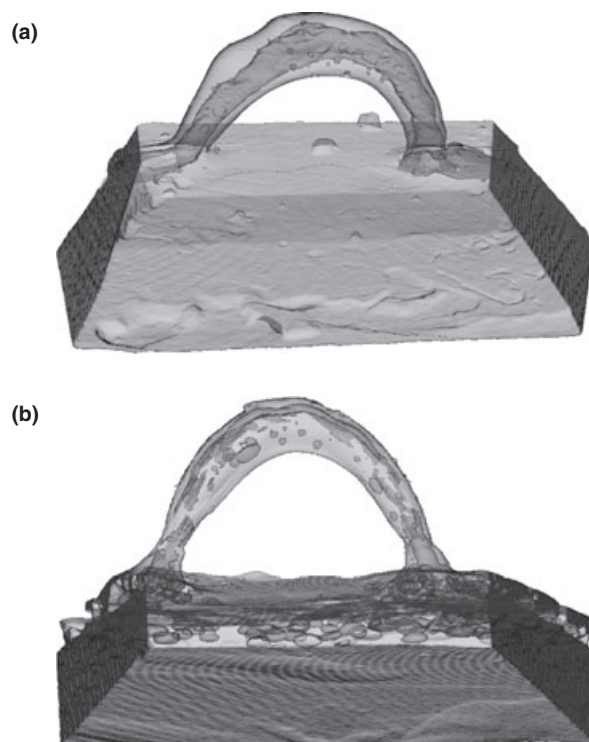


Fig. 7. 3D rendering, obtained by X-ray computed tomography, of SiSiC loops. (a) Hollow dark gray region inside the loop formerly occupied by the fugitive yarn, (b) full, that is, permanent yarn.

Fugitive, PET fibers were selected because of their:

- low cost,
- easiness to be weaved,
- similar behavior to the slurry (weight loss, shrinking) during pyrolysis resulting in less cracks formation.

Their main drawback is due to their low rigidity which could not guaranty to keep the loop shape during processing.

Ceramic Characterization

To characterize the skeleton material of the loops, another manufacturing technique was used to obtain samples with suitable dimensions. Green bulk plates comprised of the same constituent materials were produced by hot pressing, pyrolyzed, and silicon infiltrated. This solution allowed to produce monolithic pieces complying with the geometries necessary for characterization.

Table I. Material Properties Measured on the SiSiC Material

Property	Average value	RSD%
Young's modulus (GPa)	264	6
Vickers micro hardness Load 500 (g) HV	1729	11
MOR (3-point flexural stress) (MPa)	203	21
Thermal conductivity (W/m/K)		
193°C	85.08	
501°C	59.66	
1004°C	41.68	
1504°C	37.49	
Coefficient of thermal expansion (per °C)		
100°C	7×10^{-6}	
300°C	4.5×10^{-6}	
1400°C	5×10^{-6}	

Elastic modulus was measured using the impulse excitation technique with a Mk5 apparatus (Grindo Sonic Leuven B). Thermal conductivity λ was calculated according to the relationship: $\lambda = \alpha c_p \rho$. Thermal diffusivity α together with specific heat capacity c_p was measured with a Flashline 4010 (Anter, New Castle, DE). Measurements of the coefficient of thermal expansion as a function of the temperature, with a fixed heating rate of 5°C/min and from RT to 1500°C, were performed with a Setsys Evolution TMA (Setaram F). Mechanical properties under bending were measured with a Sintech 10 D universal testing machine (MTS, Eden Prairie, MN). Microhardness was measured with a microdurometer (Leitz Wetzlar D). Samples' microstructure was analyzed with Zeiss-Evo-50 SEM (Table I).

SEM image in Fig. 8 shows a central void typical in fugitive loops. The microstructure of the as-produced loop shows only two phases: dark gray zones and light gray zone. XRD analysis (Fig. 8b) confirmed that these phases are, respectively, silicon carbide and silicon.

Microscopic characterization confirmed that the base material of the loops is very similar to that of the SiSiC ERBISIC foams (Erbicol SA, Balerna CH). Indeed, this material applied by replica on reticulated foams has proven to withstand long lasting severe oxidative environments typical of the porous burners technology.⁷

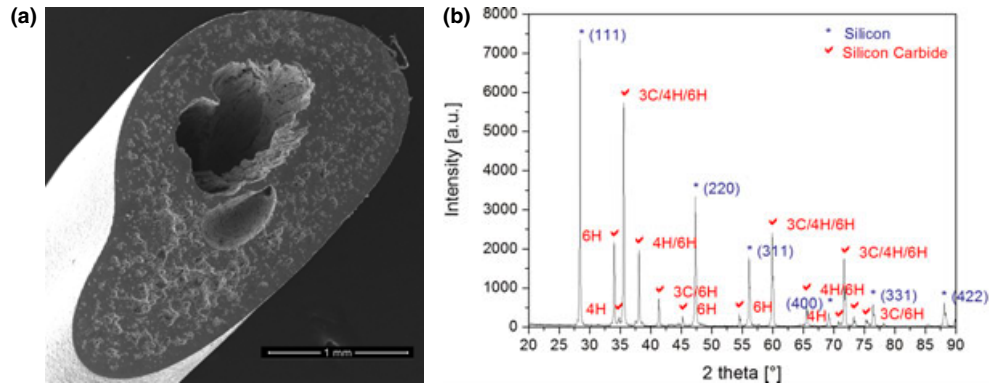


Fig. 8. (a) SEM image of a loop cross-section produce by replicating a PET yarn (hollow space) as per the process described in the previous section, loop cross-section is not round due to slurry draining before solidification (b) X-ray diffractometry analysis of the loop material.

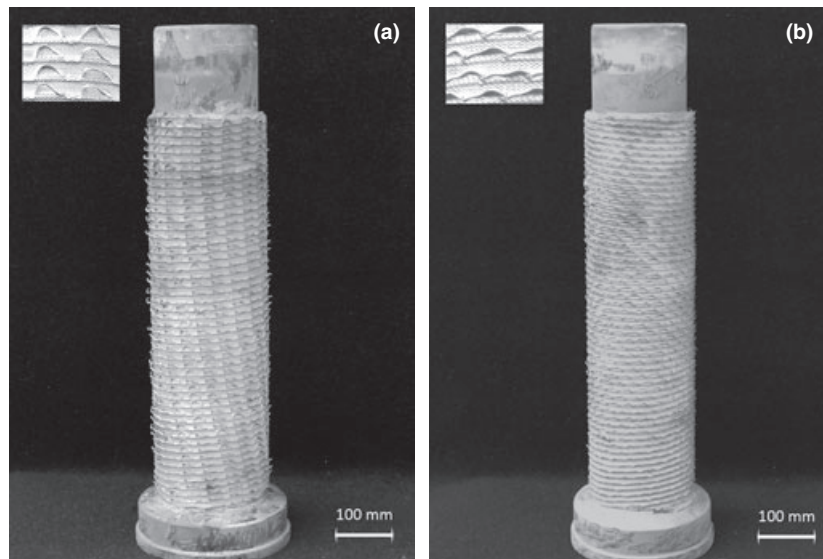


Fig. 9. Two SiSiC heat exchangers with different loops distribution. (a) Geometry 01, 3600 Loops/m². Loop: $R = 8 \text{ mm}/D = 1 \text{ mm}$ and (b) Geometry 02, 7200 Loops/m². Loop: $R = 8 \text{ mm}/D = 1 \text{ mm}$.

Properties were utilized to refine system design. They also confirmed to be quite similar to other commercial SiSiC products.

Prototype

Manufacturing

The manufacturing method developed and optimized in section 'Processing' was adopted to fabricate two complete heat exchanger units with the two

reference geometries. Textiles and SiSiC tubes (Schunk Ingenieurkeramik Willich-Münchheide, D) were combined in the ceramization process, and silicon infiltrated to obtain the final article. Textile structure placing inside and outside the tube walls was optimized, given the small radiuses of curvature of the tube.

The entire procedure was optimized on cylindrical SiSiC tubes produced by Schunk. It was then applied on a real recuperator.

The first tubes were cut in shorter units. Textiles were impregnated in the ceramic slurry and cut to

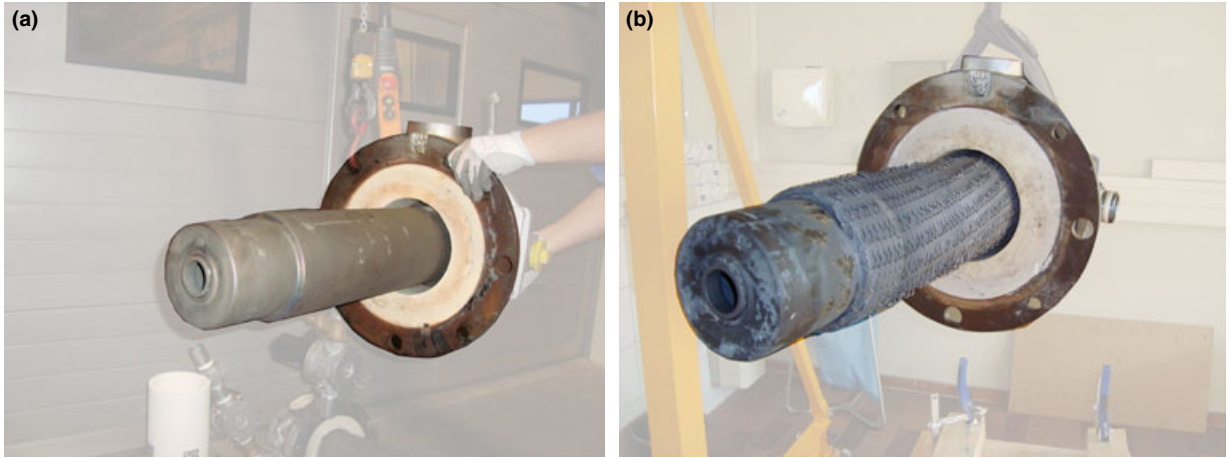


Fig. 10. burner with different recuperators (a) plain recuperator and (b) loop recuperator with Geometry 01, 3600 Loops/m². Loop: $R = 8 \text{ mm}/D = 1 \text{ mm}$.



Fig. 11. Overview of all measuring points (red for waste gas, yellow for natural gas, blue for combustion air).

dimension. These patches were placed inside and outside the tube sections. Several trials were performed to obtain tube sections complying with the design requirements. The pieces were pyrolyzed and infiltrated with molten silicon according to the ceramization process defined in section “Textile Ceramization”. Once validated the entire procedure, two complete recuperators for an 80 kW burner were produced. Results for the two different geometries are shown in Fig. 9.

Field Tests

The recuperators tested in this work at different power levels were as follows:

- plain recuperator without loops and with waste gas guiding tube (Fig. 10a)

- structured recuperator with loops (geometry 1) and with waste gas guiding tube (Fig. 10b)

Both recuperators were assembled in the same test burner and operated in indirect heating using a jacket tube $\varnothing 200 \text{ mm}$. To characterize the burner, many different measuring points were assembled at the burner and the jacket tube. Figure 11 shows the points where several parameters were acquired.

Via these measurements the following parameters could be determined:

- Average temperatures at inlets and outlets as well as in the jacket and flame tube
- temperature profiles throughout the recuperator
- waste gas mixture
- firing efficiency
- pressure drops

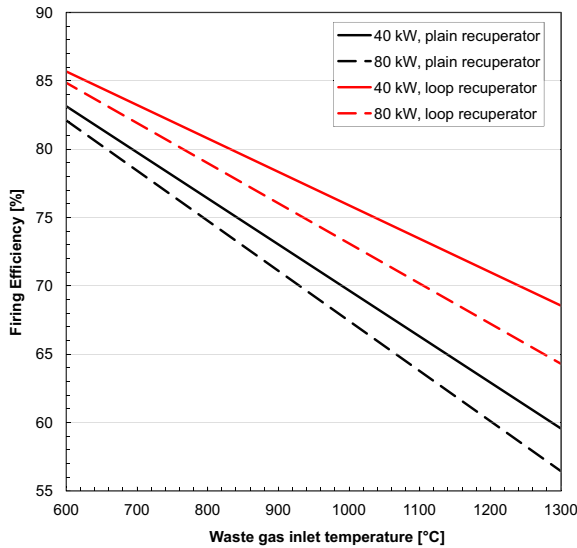


Fig. 12. Comparison of firing efficiency of plain and loop recuperator at different capacities.

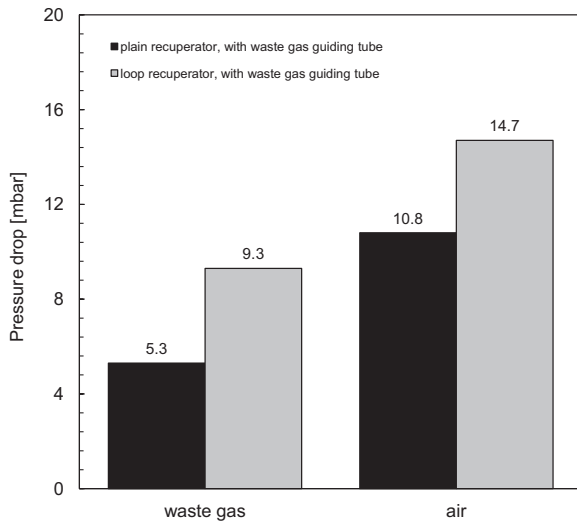


Fig. 13. Comparison of pressure drop for waste gas and air of plain and loop recuperator at 80 kW with an inlet temperature of 1000°C.

Using the loop recuperator with geometry 1, the firing efficiency was increased by 7–9% depending on the waste gas inlet temperature. Although tests have been not yet performed, we expect a further increase in the efficiency using of loop geometry 2 and an optimizing of the waste gas guiding tube. The firing efficiency at capacities of 40 and 80 kW with different

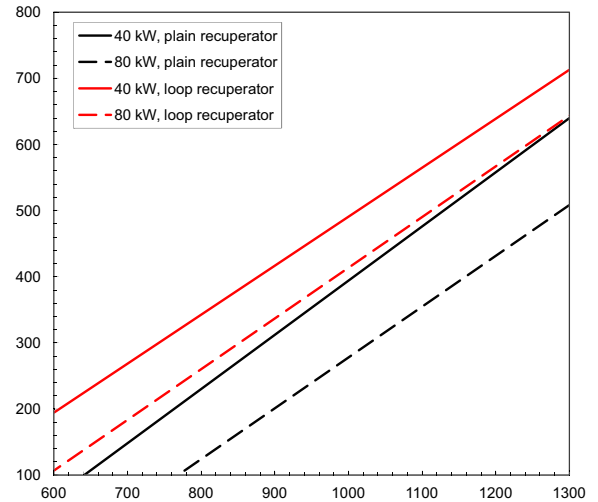


Fig. 14. Comparison of air preheating of plain and loop recuperator at different capacities.

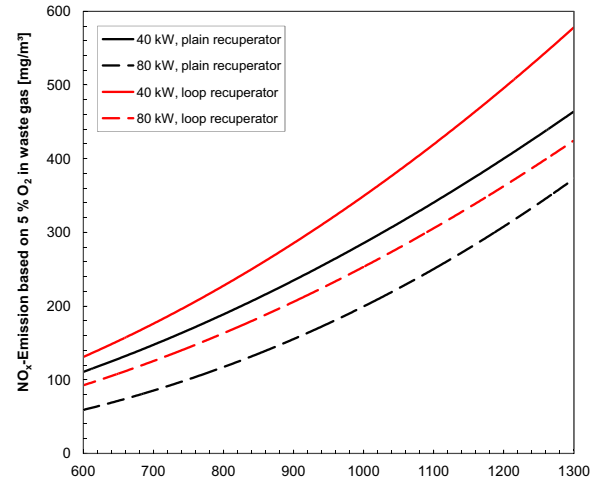


Fig. 15. Comparison of NO_x emissions in waste gas of plain and loop recuperator at different capacities.

waste gas inlet temperatures is shown in Fig. 12. The increase in the efficiency causes a higher pressure drop at the air and at the waste gas sides (see Fig. 13). The air preheating (Fig. 14) could be increased by 60–80°C with the use of the new recuperator geometry.

Certainly, the raising of efficiency and air preheating causes an increase in the NO_x emissions in the waste gasses. At 40 kW, they raise about 25%, at 80 kW about 13% (Fig. 15). With specific arrangements

concerning to the combustion system, a reduction in the emissions could be achieved. Due to the increased surface area of the tube with highly structured surface elements, oxidation phenomena will be enhanced. In any case, if passive oxidation conditions are met, they should not reduce the performance of the components, as previously demonstrated on SiSiC reticulated ceramics.⁷

As expected for this kind of materials, the Si-SiC loops are very fragile. After multiple assembling, many loops on the waste gas side were broken and lost. Therefore, a possibility for protecting the loops during the assembling has to be developed.

Further work is currently ongoing to study the overall behavior of these structures along the entire lifespan of the component, in particular, to study the deposition of dust and or soot onto the loops.

Acknowledgments

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