THE REALIZATION OF MICRO-REACTORS IN LTCC TECHNOLOGY FOR HYDROGEN PRODUCTION

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Key words: LTCC technology, micro-reactor, buried channels, steam reforming

Abstract: One of the possibilities for achieving portable power systems is a low-temperature fuel-cell system integrated with a fuel processor. A fuel processor is needed to make the hydrogen from liquid fuels (mainly methanol), as the required fuel for PEM (polymer-electrolyte membrane) fuel cells. LTCC (Low Temperature Co-fired Ceramics) technology was used to prepare prototypes of fuel processors for low-temperature fuel cells. The planar integrated LTCC structure with buried cavities and channels including two evaporators (fuel and water for steam reforming), the mixing chambers and the reformer was designed and successfully realized. The length of the buried channels is nearly 2 m. The carbon-based thick-film pastes were evaluated for sacrificial layers and were used to form the buried channels. Platinum-based heaters and temperature sensors were integrated onto the top of the structure. The fuel-processor micro-channels were covered with the required catalyst, using either the sol-gel or wash-coating techniques. A laboratory experimental environment for testing was also provided. Preliminary results on the steam reforming of the liquid fuel (methanol) and the water are presented.

Realizacija keramičnih mikro reaktorjev v tehnologiji LTCC za izdelavo vodika

Kjučne besede: LTCC tehnologija, mikro rektorji, pokopani kanal, parni reforming

Izvleček: Nizko temperaturne gorivne celice s polimerno membrano (PEM – Proton Exchange Membrane), integrirane s procesorjem goriva, so ena od možnosti za prenosne vire električne energije. Procesor goriva potrebujemo, da s pomočjo kanalizirane reakcije pridobimo vodik, ki je gorivo za gorivne celice, iz tekočih goriv, v glavnem metanola in vode. Prototipi keramičnih mikro reaktorjev so bili realizirani s pomočjo LTCC tehnologije (Low Temperature Co-fired Ceramics – keramika z nizko temperature žganja). Planarne integrirane LTCC strukture s pokopanimi votlinami in kanali so bile načrtane in izdelane. Te 3D strukture vsebujejo dva uparjalnika, za vodo in gorivi, mešalne komore in reformer za reakcijo med tekočim gorivom in vodo. Dolžina pokopanih kanalov je okrog 2m. Debeloplastne paste na osnovi ogljika so bile uporabljene za izdelavo pokopanih kanalov. Grelci in temperaturno senzorji na osnovi Pt debeloplastnih prevodnikov so bili integrirani na površini. Notranje površine mikro kanalov procesorskega dela so bile pokrite s katalizatorjem, ki je bil nanesen s sol-gel tehnologijo. Razvili smo eksperimentalno laboratorijsko okolje za testiranje. Predstavljeni so preliminarni rezultati kataliziranih reakcij med metanolom in vodo.

1. Introduction

2.1. LTCC ceramics

Advanced ceramic micro-systems are in many cases created with multilayer ceramic modules that integrate screen-printed thick-film electronic components and sub-circuits as well as 3D buried structures, for example, cavities or channels. Low Temperature Co-fired Ceramics (LTCC) technology is considered as one of the more suitable technologies for the fabrication of ceramic micro-systems. These LTCC materials are sintered at the low temperatures typically used for thick-film processing, i.e., around 850°C. To sinter to a dense and non-porous structure at these, rather low, temperatures, LTCC materials have to contain some (or a great deal of) low-melting-point glass phase. Unfired LTCC tapes are a mixture of glass and ceramic particles, for example, alumina, and an organic phase. During firing, first the organics burn out at around 450°C, leaving a mixture of glass and ceramic particles. At higher temperatures the glass phase melts and the material sinters to a dense and non-porous structure. The whole process is schematically shown in Fig. 1. During the high-temperature processing some part of the glass crystallizes ([1-5]). This increases the viscosity of the remaining glass phase and the glass does not soften during subsequent re-firings.

The microstructures of the green and the fired LTCC material (Du Pont 951) are shown in Figs. 2.a and 2.b, respectively ([4,6]). The unfired material is a mixture of darker

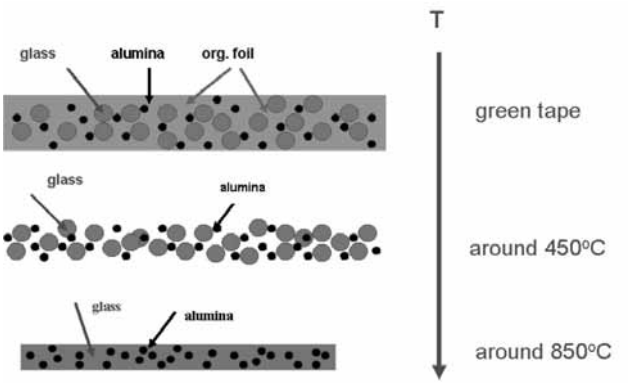


Fig. 1. Cross-sections of the LTCC tape at different temperatures during firing (schematic).

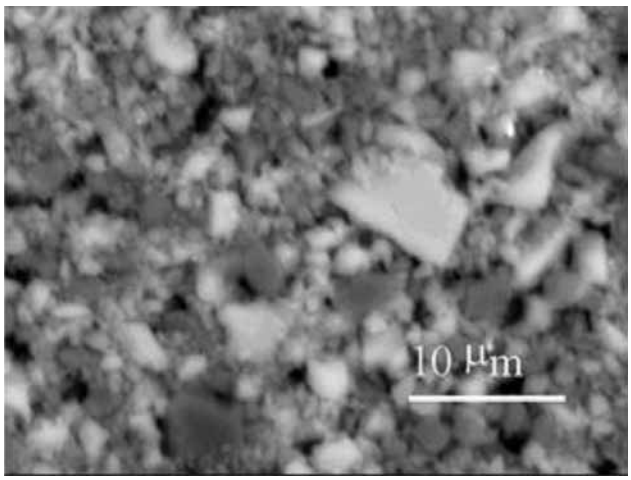


Fig. 2.a: Microstructure of a green LTCC tape. The darker grains are alumina and the lighter grains are glass particles

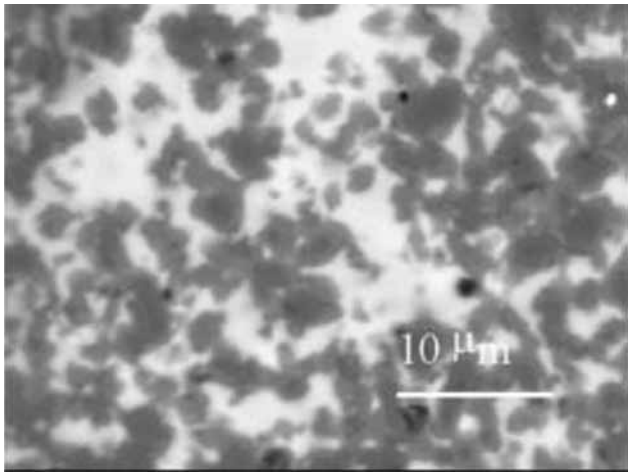


Fig. 2.b: Microstructure of a CC tape fired at 850°C. The material is densely sintered. The darker grains of alumina are embedded in the lighter glass matrix.

alumina and lighter glass particles. After firing the material is densely sintered with dark alumina grains in the glass matrix.

The whole process flow of the LTCC technology is schematically shown in Fig. 3. The green tapes are cut into the required dimensions. Vias are punched and filled with a conductor material. After this the conducting layers are screen printed. The substrates are then visually examined and put together into multilayer "packets. These "packets" are laminated under a pressure at temperatures around 80°C and fired at relatively low temperatures of around 850°C, which are typically used for thick-film processing,

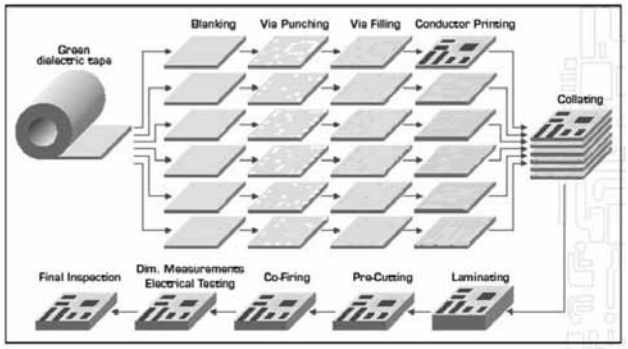


Fig. 3. The LTCC process flow.

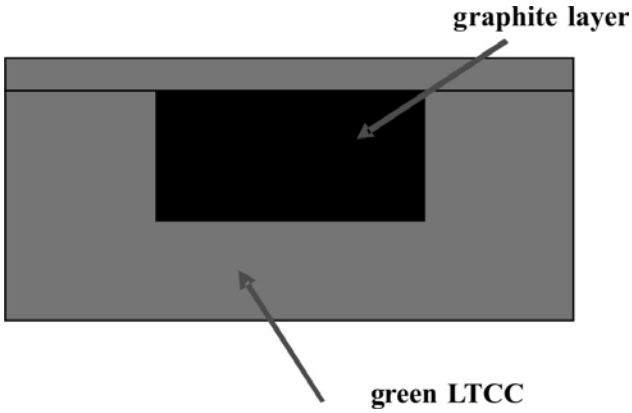


Fig. 4.a: Unfired LTCC structure. Graphite layer is laminated within the structure (schematic).

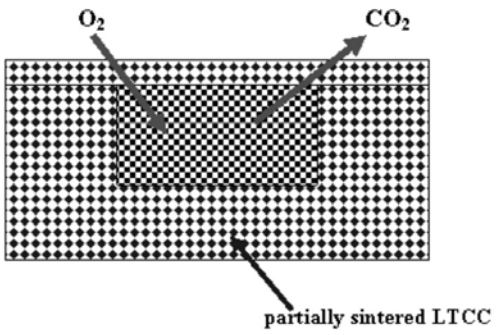


Fig. 4.b: Partially sintered LTCC structure with open porosity. The oxygen and carbon dioxide are diffusing through the porous structure (schematic).

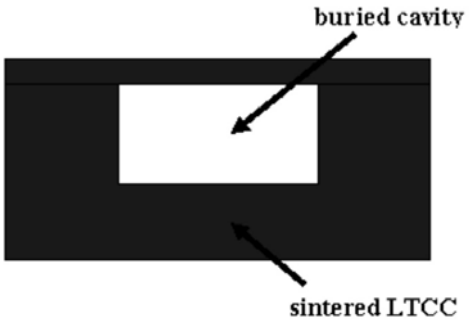


Fig.. 4.c: LTCC material is fully densified (schematic).

2.2. Sacrificial materials

LTCC technology is inherently efficient for producing 3D structures, e.g., buried cavities or channels, for different applications, ranging from pressure sensors to "laboratory-on-chip" systems ([7,8]). However, as mentioned above, LTCC tapes contain a relatively high concentration of low-melting-point glass, enabling the materials to sinter densely at relatively low firing temperatures. During the firing the top parts of the structures over the buried cavities tend to deform; generally they tend to sag. One of the possible solutions is to use so-called sacrificial carbon-based layers within the 3D LTCC structures ([9-11]). These sacrificial layers are either screen-printed (pastes) or inserted (tapes) into the green LTCC structures before the lamination. During the lamination and the earlier stages of the firing they must support the desired 3D structure and prevent deformations. Later, the sacrificial layers ought to burn out. The temperatures for the oxidation of the carbon material must be a little higher than the temperature for the onset of sintering of the LTCC 3D structures. This means that while the LTCC tapes start to sinter they are still porous enough so that the oxygen from the air can diffuse to the inside of the LTCC structures and the CO_2 can diffuse to the outside. All the carbon most be oxidized before the LTCC layers are fully densified. This is shown very schematically in Figure 4.a (unfired structure), 4.b (porously sintered LTCC structure) and 4.c (densely sintered LTCC structure), respectively.

A typical sintering/densification curve for the LTCC material (951, Du Pont) is shown in Figure 5 ([12]). The material starts to sinter at temperatures around 700°C and is more or less fully densified at 800°C. However, note that the sintering curve is a "dynamic" one – the temperature is increasing. However, if the samples were to be held at some temperature the material will still shrink, albeit at a slower rate. Suitable temperatures for the oxidation of the sacrificial layers would be a little over 700°C.

2.3. Micro-reactors for hydrogen production

Small, compact fuel cells that operate on liquid fuels offer lightweight alternatives to batteries, thus allowing the greater portability of electronic devices, e.g., laptop computers.

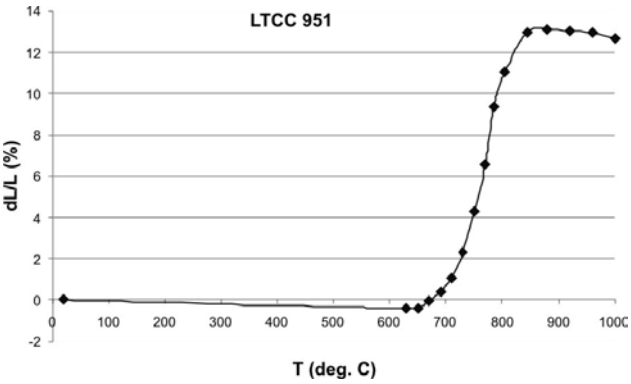


Fig. 5: Sintering curve of a typical LTCC material ([12])

These fuel cells are mainly based on low-temperature (under 100°C) PEM electrolytes (Proton Exchange polymer Membranes or Polymer Electrolyte Membranes). The required fuel for these fuel cells is pure hydrogen. To supply this fuel, water and a liquid fuel are steam reformed into hydrogen-rich gases with the help of appropriate catalysts.

A very schematic representation of a combination of the fuel processor and the PEM fuel cell is shown in Figure 6. A liquid fuel and water and/or air are fed into the fuel processor and the resulting hydrogen-rich gas is used as a fuel for the PEM fuel cell.

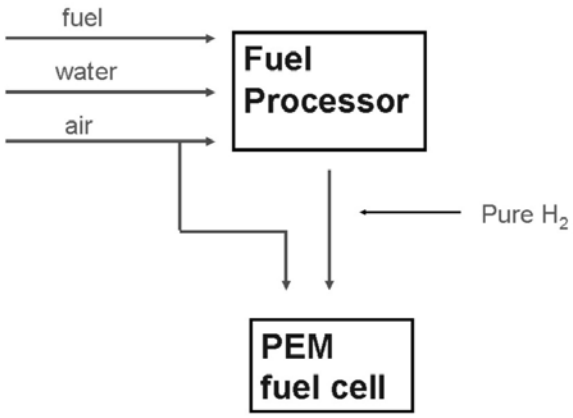


Fig. 6: Schematic presentation of a combination of the fuel processor and the PEM fuel cell. Liquid fuel and water and/or air are fed into the processor.

The ideal catalyst-assisted reaction would result, for example, for methanol and water:

$$CH_3OH + H_2O \rightarrow CO_2 + 3\,H_2 \qquad (1)$$

However, some side reactions also produce some small quantities of the undesirable (i.e., harmful) CO, which must be removed after the reforming. Therefore, complete fuel processors for the conversion of liquid fuels into (more or less) pure hydrogen consist basically of fuel vaporisers, fuel reformers and gas clean-up units (mainly to remove the excess carbon monoxide). A schematic outline of the fuel processor is shown in Figure 7. The two modules on the right are the water gas shift rector (WGS) and the preferential oxidation reactor (PrOx) for removing the carbon

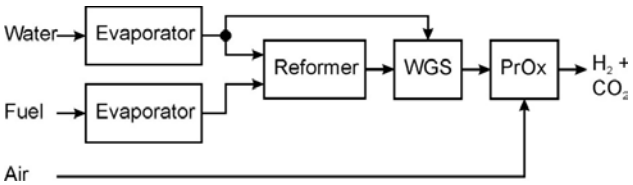


Fig. 7: Schematic outline of the fuel processor. WGS – water gas shift. PrOx – partial oxidation.

monoxide from the gas mixture by reactions with water and oxygen, respectively. The catalyst-assisted reactions are:

Reformer – fuel + H_2O + (O_2) → H_2 + CO_2 + CO (2)

WGS (water gas shift) – CO + H_2O → CO_2 + H_2 (3)

PrOx (preferential oxidation) – CO + O_2 → CO_2 (4)

Ceramic fuel processors based on LTCC technology for the catalyst-assisted production of hydrogen from liquid fuels (mainly methanol) for PEM (polymer-electrolyte membrane) fuel cells have been reported in the open literature. A. Kundu et al. ([13]) made an extensive overview of micro-processors with over 100 references. Most of the described devices are realized on single-crystal silicon wafers. However, some are made with LTCC or other ceramic materials. Koripella et al. ([14]) (Motorola) patented the compact design of an LTCC-based fuel processor with an integrated PEM fuel cell. Y. Shin et al. ([15]) reported simple, three-layered LTCC structures for micro-reformers and PrOX (preferential oxidation of the carbon monoxide) units.

In the paper the design and development of the planar ceramic micro-reactor will be described. The reactor was made using LTCC technology and sacrificial materials. Carbon-based materials for sacrificial layers will be briefly discussed. The planar LTCC structure includes two evaporators - fuel and water for steam reforming, mixing chambers and the reformer. Preliminary experiments were made with the water/methane steam reforming.

3. Experimental

3.1. Sacrificial layers

The evaluated carbon-based pastes for sacrificial layers were Harmonics (Carb-Paste-1) and Electro Science Labs. (4440). They were analyzed by thermogravimetric analysis (TGA). The heating rate was 5K/min. In most cases the firing profile of the LTCC tapes was simulated. This means firing first for 1 hour at 450°C (LTCC tapes organic binder burnout). However, the highest firing temperatures were in most cases 720°C (and not 850°C, which is the firing temperature of the LTCC material) for some hours to determine how long it takes to completely oxidize the carbon-based materials at temperatures when the sintering of the LTCC materials starts, while the ceramic tapes still "possess" the open porosity. In the case of the Harmonics thick-film paste some residue of a brownish powder (less than 1 wt.%) was left after the firing. This residue was analyzed using an X-ray diffraction (XRD) analyser and a scanning electron microscope (SEM) equipped with an energy-dispersive X-ray analyzer (EDS).

3.2. Micro-reactor

The prototype LTCC reactor – a horizontally connected evaporator and reformer – was designed. It is shown schematically in Figure 8. The reactor is composed of two evaporators, two mixing chambers and a reformer. On the left-hand side are the buried cavities for the input of the fuel and the water. Two levels of buried channels (dimensions 400 μm x 50 μm) – the evaporator part – connect the entrance chambers and the two mixing chambers in the middle of the substrate. Three levels of channels lead from the mixing chambers into the "output" chamber, i.e., the reformer. The channels were made with the help of sacrificial layers. The chosen graphite-based thick-film paste was screen-printed onto the green LTCC foils. On the top of the structure two platinum heaters for separate green LTCC foils (951, Du Pont) were laser cut to the required dimensions. The graphite-based thick-film paste was screen-printed to form the buried channels. The 3D structures were made by laminating layers of 951 (Du Pont) LTCC tapes at 70°C and at a pressure of 200 bars. The prepared samples were fired first for 1 h at 450°C (burn-out of organics) and then fired at maximum temperatures of 850°C. On the top of the fired structures platinum heaters and temperature sensors, and palladium/silver connections were printed and fired at 850°C.

The CuO-CeO_2 catalyst containing 20 mol % CuO was directly prepared without the need for a calcination step, by co-precipitation from mixtures of cerium and copper aqueous nitrate solutions with ammonium hydroxide. The catalyst slurry was prepared by mixing the catalyst (6.5 wt. %), aluminium hydroxide sol and 2-propanol. The precursor was then injected into the micro-channels with the syringe

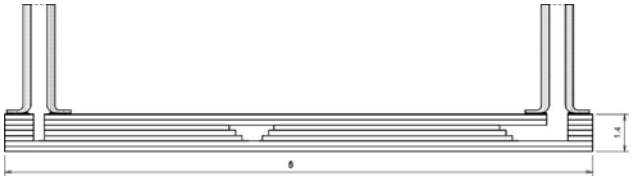


Fig. 8.a: Schematic longitudinal cross-section of the LTCC-based reactor comprising integrated evaporator and reformer. The evaporator is on the left-hand side and the reformer on the right-hand side. Buried cavities for the input of fuel and water on the left, mixing chambers in the middle and gas output cavity on the right are connected with buried channels

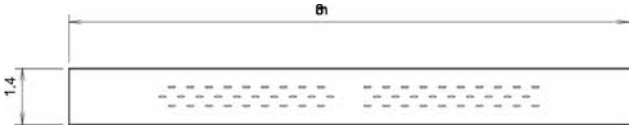


Fig. 8.b: Schematic transversal cross-section of the LTCC based reactor through three layers of buried channels on the right (reformer) part of the structure.

until it filled up the volume of all the micro-channels. The coating was then dried at 60°C for 12 h, and the process was repeated to achieve the desired weight of catalyst. After drying the samples were calcined in air at 450°C for 4h.

3.3. Integrated laboratory experimental environment

The experimental work (reforming) was performed within an integrated experimental environment, which supports various kinds of tests of the operation and efficiency of the reformer and its subsystems during the design and optimisation phase. The majority of the components of the integrated experimental environment are controlled via a computer, which simplifies the acquisition of the results and their documentation. The integrated experimental environment shown in Fig 9 contains the following subsystems:

- Temperature-profile measurement of the ceramic body surface is measured by a thermo-vision camera and is used to support the optimal thermal design of the ceramic structure.
- Temperature measurement and control is implemented independently for the evaporator and the reformer. The temperature is controlled using Pt sensors and heaters, which are integrated onto the reformer surface. Dedicated measurement and power-control electronic circuits are designed and provided.
- Flow rates of input reactants (water, methanol/diesel) must be carefully controlled and they have to be independent of the back pressure in the reformer. For the dosage, syringe-like cylinders are used and powered by step motors, allowing the adjustment of flow rates and quantities.
- Control software running on a personal computer is used for the data acquisition and control. Within the software the temperature evaluation and control is implemented, as well as the control of the flow rates and the quantities of the input reactants.

Preliminary experiments were made with methanol as the fuel and water for the steam reforming. The gas reaction products were bubbled through the water where the un-reacted methanol and water condensed in the water bath. The chemical analysis of the output products was performed using mass spectroscopy analyses.

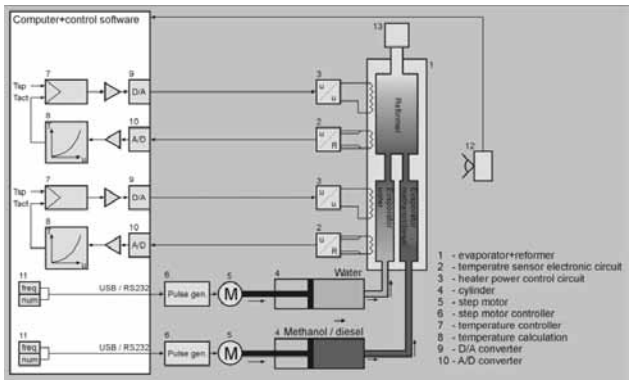


Fig. 9. The integrated experimental environment

4. Results and discussion

4.1. Carbon-based sacrificial layers

The TGA analyses of the carbon-based sacrificial materials are rather similar. The DTA analyses of the Harmonics and ESL (Electro Science Labs.) pastes are shown in Figures 10.a and 10.b, respectively. The weight loss is on the left "y" axis and the temperature is on the right. Samples were heated up to 450°C and held at this temperature for 1 hour to simulate the firing cycle of the LTCC green tapes. The temperature was increased to 720°C and the samples were then held at this temperature. The results for both pastes are similar. There is a noticeable weight decrease at 450°C, which is attributed to a burn-out of the organic vehicle in the pastes. The complete evaporation and burn-out of the added organic materials is mainly over before the 450°C "step" is reached and only a small (if any) further decrease of the samples' weight is observed during the one hour at 450°C. When the temperature reached 720°C a significant oxidation of the carbon-based materials starts. For both

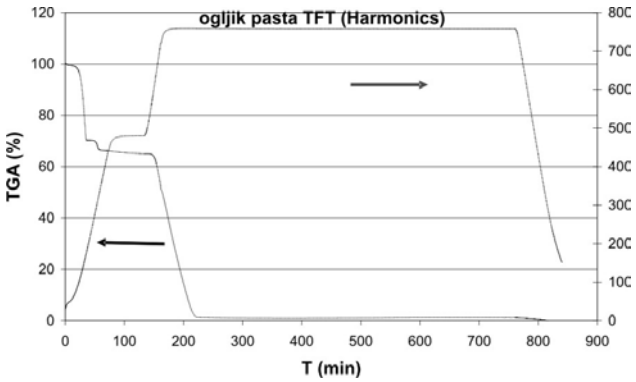


Fig. 10.a: TGA analysis (weight loss) of carbon-based sacrificial material (thick-film paste, Harmonics). The weight loss is on the left "y" axis and the temperature is on the right.

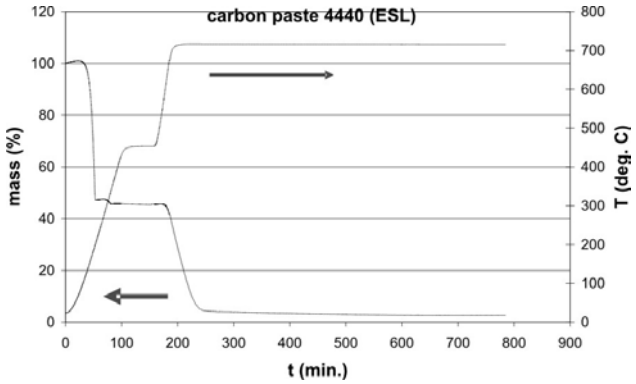
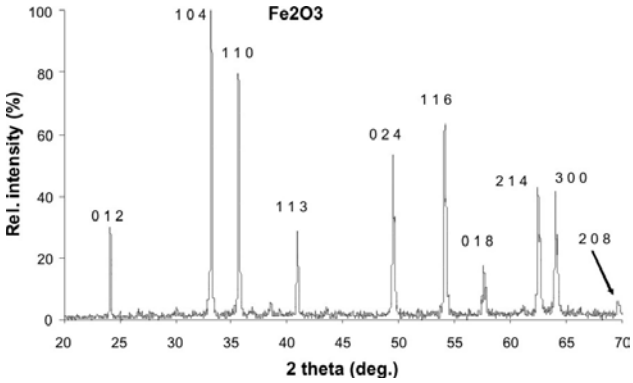


Fig.10.b: TGA analysis (weight loss) of carbon-based sacrificial material (thick-film paste, ESL, 4400). The weight loss is on the left "y" axis and the temperature on the right.



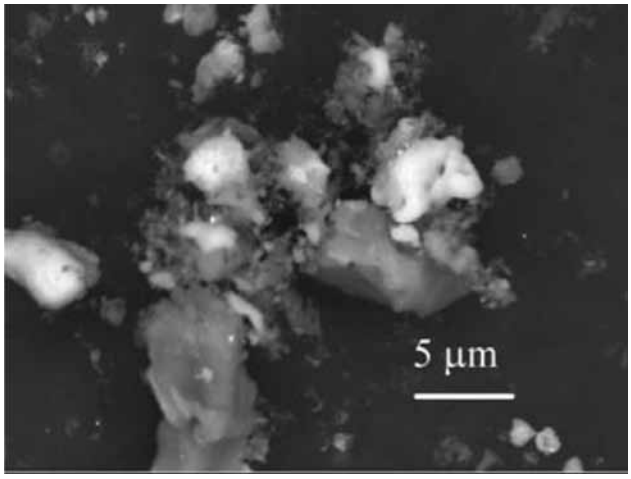


Fig. 11: The X-ray analysis of the brownish residue after the burn-out of some graphite-based sacrificial materials. The peaks of Fe_2O_3 are indexed.

the analyzed materials the oxidation is over in 100–150 min at 720°C. After that time no significant loss of weight can be detected.

As mentioned in the "Experimental" section, after the burn out the Harmonics thick-film paste leaves a small quantity (less than 1 wt.%) of brown residue. The X-ray analysis of the residue is shown in Figure 12.a and an SEM picture is presented in Figure 11.b. The X-ray analysis showed that this residue is an iron oxide (Fe_2O_3). This was confirmed by the EDS micro-analysis of the residual powder. The lighter particles observed in the microstructure in Fig. 12.b. are the iron oxide. Therefore the ESL 4440 which oxidises cleanly without any residue was used for sacrificial layers.

4.2. LTCC micro-reactor

The ceramic micro-reactor was prepared from eight layers of LTCC tapes. Carbon-based sacrificial layers were used to form the buried structures. The dimensions of the planar micro-reactor after firing are 70x30x1.45 mm^3. The surface of the horizontally integrated evaporator and the reformer realized in the LTCC technology reactor with the attached input and output "tubes" are shown in Figure 12.a. From left to right: the input openings for the fuel and water, the heater for both evaporators and the Pt-based resistive temperature sensor, the heater for the reformer

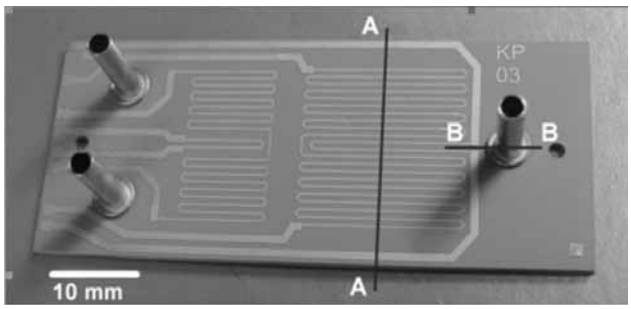


Fig. 12.a: The picture of the surface of the horizontally integrated evaporator and reformer realized in LTCC technology.

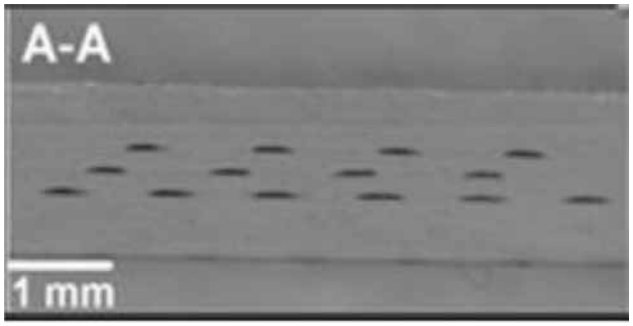


Fig. 12.b: The microstructural cross-section through the line A-A (see Fig. 7.b) is shown. Three layers of buried channels connect the mixing chambers and the output chamber.

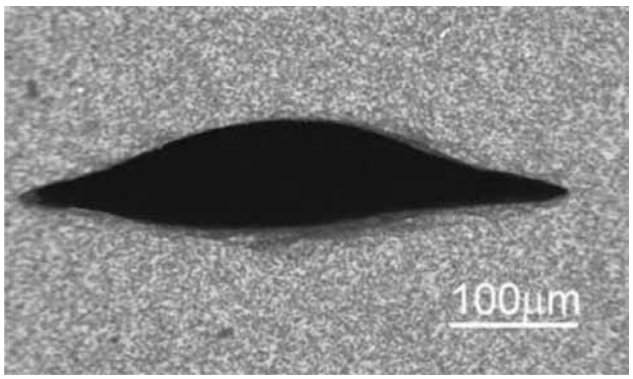


Fig.12.c: Cross-section of a channel.

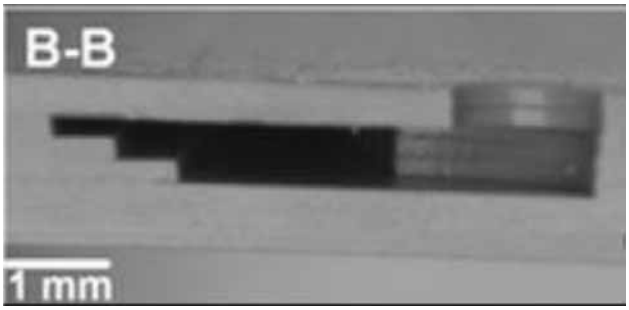


Fig. 12.d: The microstructure through the line B-B (see Fig. 7.b) represents the cross-section of the buried cavity at the output chamber.

and the second Pt-based resistive temperature sensor, and the output opening. In Figure 12.b the microstructural cross-section through the line A-A is shown. Three layers of buried channels connect the mixing chambers and the output chamber. The total length of the buried channels is around 2 m. A cross-section of one channel is shown in Figure 12.c. The microstructure (line B-B in Figure 13.b) presents the cross-section of the buried cavity at the outlet. The three-layered structure – three layers of channels – can be seen in Fig.12.d.

Within the experimental environment, which was described in the "Experimental" subsection, a number of experiments were performed. The first set of experiments was intended to test the thermal design of the ceramic body structure. We heated up the reactor structure to the working temperatures and observed the temperature profiles on the surface with a thermo-vision camera. Fig-13 shows an example of the temperature profile. The bottom region in the picture represents the area of the evaporator and the upper region is the reforming reactor.

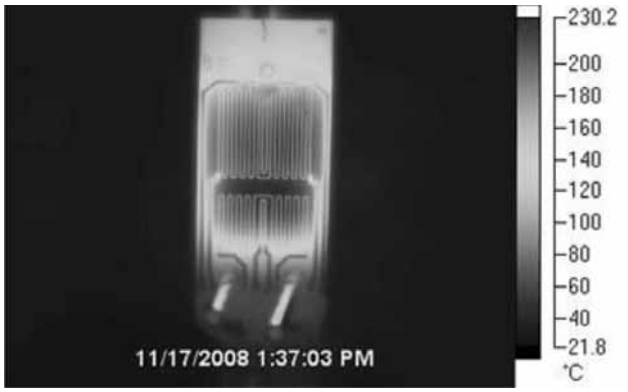


Fig. 13: An example of the temperature profile.

In the second set of experiments the operations of the reforming process were analysed and the preliminary analyses of the output products using the mass spectroscopy were performed. The preliminary results are shown in Table 1 where the concentrations of the output gases vs. the processing conditions are summarised. The table above contains seven different measurements. These measurements were conducted with the LTCC reformer at different temperatures and different flow rates of water and methanol. Generally, the produced hydrogen seems to be very pure. Note, however, that the molecular masses of N_2 and CO are the same i.e., 28, and therefore they cannot be distinguished using mass spectroscopy. At low temperatures (measurements 1 and 2) below the catalyst temperature window only very small amounts of hydrogen were produced. On the other hand, when the temperatures were within the required temperature window (measurements 3 and 4), the concentrations of hydrogen were increased significantly.

Table 1 – Mass spectroscopy results under various process conditions (temperatures., and flow rates of methanol and water)

No.	Temp. (°C)	Flow rate methanol (ml/h)	Flow rate water (ml/h)	H_2(%	CH_4(%)	N_2/CO(%)	O_2(%)	Ar(%)	CO_2(%)
1	200	22,6	34	5	0.6	69	23	0.9	1,6
2	200	11,3	34	7	0.6	68	23	1,1	1,1
3	270	11,3	34	35	0.5	45	15	0.6	3,9
4	370	11,3	23	55	0.2	34	7	0.3	3,8

5. Conclusions

The described ceramic micro-reactor was developed for the steam reforming of liquid fuels and water into hydrogen, which will fuel low-temperature fuel cells. The reactor was designed and made using LTCC technology. The planar LTCC structure with buried cavities and channels, including two evaporators - fuel and water for steam reforming, mixing chambers and the reformer was realized. Carbon-based sacrificial layers were evaluated and then used to form the buried structures. The total length of buried channels is around 2 m. Platinum-based heaters and temperature sensors were integrated onto the top of the structure. The CuO / CeO_2 catalyst was deposited within the channels of the structure. The reforming process was evaluated by analysing the output gas products using mass spectroscopy. These preliminary measurements validated the efficiency of the used catalyst.

The results confirm that Low Temperature Co-fired Ceramic (LTCC) technology has the potential to fulfil the process requirements meant for the conversion of fuels into a hydrogen-rich gas. Currently, the technology has demonstrated its suitability for the design and realisation of a planar micro-reactor with a reasonable yield of hydrogen.

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