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Study on the Implementation of sintered LTCC and Graphite as a sacrificial material for the fabrication of Microcombustors

NSF Summer Undergraduate Fellowship in Sensor Technologies
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ABSTRACT

The micro-combustor is a compact, sub-millimeter device that burns hydrocarbon fuels homogeneously as a source of power. It efficiently converts heat generated by combustion into electric power, and has the potential to replace batteries in portable applications that require long-term power. The possible benefits of these devices include their ability to provide greater energy and power density, higher temperatures and greater efficiency as a heat source. Also this technology can have many applications such as military portable systems, consumer portable system, and chemical control reaction systems.

The problem we are addressing is the fabrication of a gas fuel micro-combustor for a compact, portable electric power generator using thermoelectric elements. This device has been fabricated [1] using a competing technology, which is more complex, time consuming and therefore more expensive.

The materials to be used for the construction of this device are fundamentally Low Temperature Co-Fired Ceramic (LTCC) and Graphite. The fabrication of this device will rely essentially on a thermal process (sintering of the LTCC tapes). The instruments that will be use for the fabrication / characterization include: a furnace for sintering the ceramics, a heated press for the ceramics lamination, and a thermal laser and a numerically controlled milling machine for the patterning and machining of the tape.

In order to obtain a sense of the flow behavior within the device, numerous simulations have been made using a commercial program call FEMLAB. This program will take into

consideration a diversity of parameters to measure such as the speed, pressure, fluid Reynolds Number, among others.

The main objective of this project is to complete the fabrication of a small combustor that contains fundamentally three inputs, one output and a combustion area. In one of its inputs a combustible gas (hydrogen) is injected, and oxygen from the air as an oxidizer flows through the other two inputs. The gases are mixed in the combustion area. A flame is initiated in the combustion area to burn the fuel / oxidizer mixture.

It is hoped that the combustor fabrication will be completed as designed. The parameters that characterize its combustion and power are expected to be consistent with its application as an electrical generator by means of the thermoelectric effect.

We are using FEMLAB, a commercially available numerical analysis software package for the simulation process.
As means to practice the lamination process, where the LTCC and graphite are bonded by means of uni-axial forces and heat, an experiment was carried out using pencil leads (0.5mm and 0.7mm in diameter) as source of graphite. Three parameters, force, temperature and time, were used to control the pencil lead lamination to LTCC. The parameters were 1,100 -1,300 pounds, 100º to 200ºF, and 15 to 20 minutes, respectively. By sintering the LTCC (after the lamination with pencil leads), micro-channels were formed, taking the characteristics of graphite leads morphology. In other words, graphite serves as a sacrificial material in the formation of channels or conduits

Contents

1. Introduction

When the air is mixed with a combustible and ignited to form a flame producing high temperatures, the process is known as combustion. The combustion is a chemical reaction in which a fuel (element or component) is combined with an oxidizer (generally oxygen in form of gaseous O_2), giving off heat and producing an oxide. Frequently used types of element for the combustion are the carbon and hydrogen. The combustion process happens as often in living beings as in devices used as sources of energy.

When this process happens inside a device, is known as a combustor. Combustors are commonly seen in mechanical motors such as in cars, airplanes, boats, etc. As one knows these are made to move and climb, as they are designed for the displacement of great weights that require enormous amounts of energy, which implies the consumption of great amounts of fuel. Nevertheless components exist that do not require large amounts of energy. These in their majority are electronics systems, which are designed to consume energy supplied by means of batteries and electricity.

To make a combustor at a small scale, one that will work for devices requiring lower energy levels, it has been proposed that one must construct a combustor of proportionally smaller dimension. This is known as a micro-combustor. The micro-combustor is a compact, millimeter length device that burns hydrocarbon fuels homogeneously as a source of power.

The main objective of this project is to complete the fabrication of a micro-combustor out of LTCC tapes, which contains fundamentally three inputs, one output and a combustion area. In one of its inputs a combustible gas (probably hydrogen) is injected, and air flows through the other two inputs. The gases are mixed in the combustion area. A flame is initiated in the combustion area to burn the fuel / oxidizer mixture by means of a capacitor discharge or a piezoelectric element.

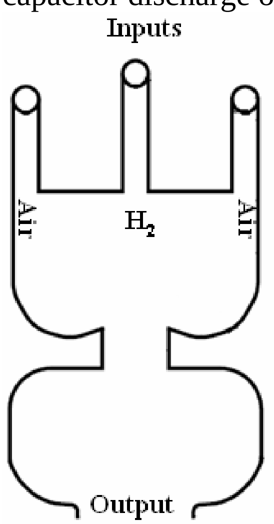
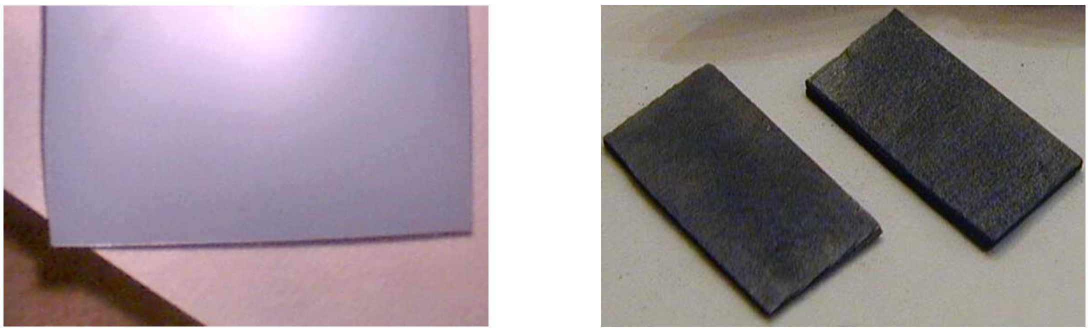


Fig1. Two-dimensional micro-combustor

The materials to be used for the construction of this device are Low Temperature Co-Fired Ceramic (LTCC) tape and Graphite. The LTCC represent an important alternative to be used as substrates for machining in the meso and micro scale. They provide several advantages including: electronic circuits can be integrated because of their hybrid nature, tapes of different compositions can be formulated to obtain desired layer properties (e.g. magnetic permeability), possibility of fabrication of hybrid structures consisting of ceramics, silicon, metals and/or some other suitable materials, layer count can be high, possibility of self-packaging, fabrication techniques are relatively simple, inexpensive and environmentally benign. Graphite is one of the two allotropic phases of carbon. This material has uses in many applications such as: electrodes, pistons, pencils, washers and diverse applications in engineering. It is of black color metallic, refractory brightness and it is easily worn away by means of heat (gasify).



a) b)

Fig2. a)LTCC and b)Graphite

The fabrication of this device will rely essentially on a thermal process (sintering of the LTCC tapes). The instruments that will be use for the fabrication / characterization include: a furnace for sintering the ceramics, a heated press for lamination of the ceramics, and a thermal laser combined with a numerically controlled milling machine for the patterning of the tape and machining of the graphite block.

Sacrificial material (graphite) insert.

The materials for the fabrication of the structure in figure 1 were graphite and LTCC. Even though the combustor is fundamentally two dimensional, it has a cavity where the fuel – oxidizer pre-mixed flame will be ignited. To form this cavity, a graphite insert (a sacrificial material, as it will disappear during high temperature sintering of the LTCC tape) will be used to keep the cavity from collapsing during sintering.

For graphite, a computer numerical control (CNC) was used. The CNC is a versatile system that allows controlled motion of the tools and parts through a computer programs that use numeric data. The numerical data that was used for the CNC in this work are the result of exporting data from a commercial program called Autocad 2000 i. This program is a useful tool in the field of engineering since it allows for an excellent quality draft of the architecture for any design. The detailed structure of the micro-combustor of a symmetrical form was designed using such CAD software. After exporting the data of the design to the CNC machine, a grinder was used as a tool to define the structure in graphite. The following figure schematically illustrates the process of definition of the structure morphology with the CNC.

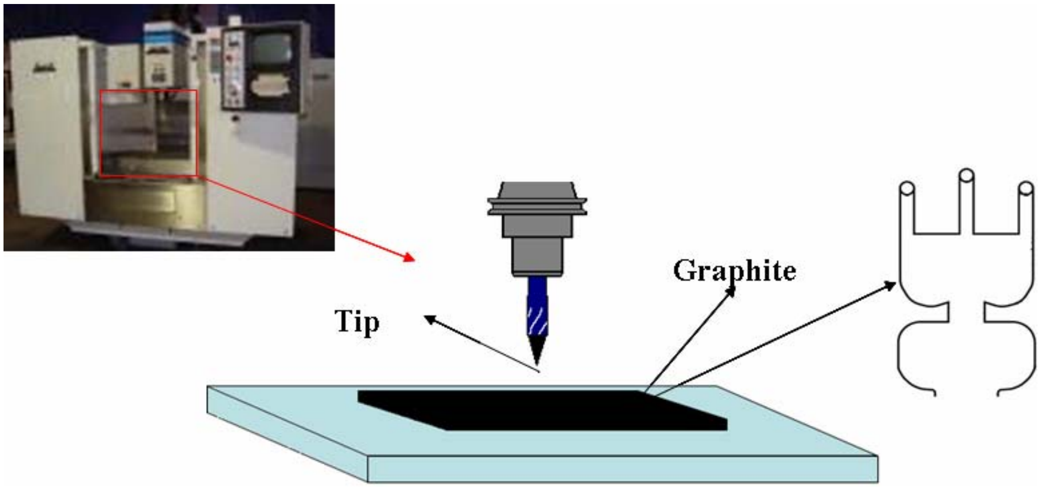


Fig.3 Schematic of Process with the CNC for the Microcombustor

When finished, the CNC machined graphite insert has the follow aspect.

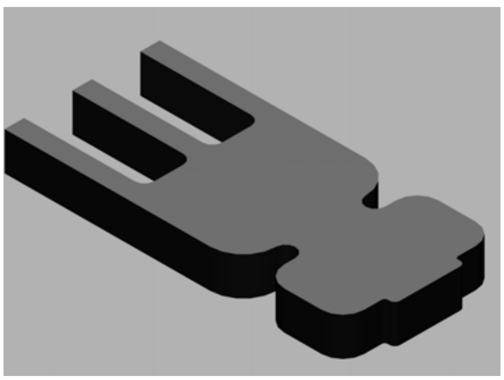


Fig.4 Scheme of the Graphite after of the CNC process

The LTCC tape combustor structure.

To define the combustor structure in LTCC tape a thermal laser was used to transfer the device pattern from an Autocad file (DXF file) to the ceramic tape. The laser system consists of a platform where the LTCC sample lies and a moving laser head shinning IR (10.6 μm) on the tape. A large piece of LTCC tape was placed in the laser platform and multiple units of the same pattern were serially machined.

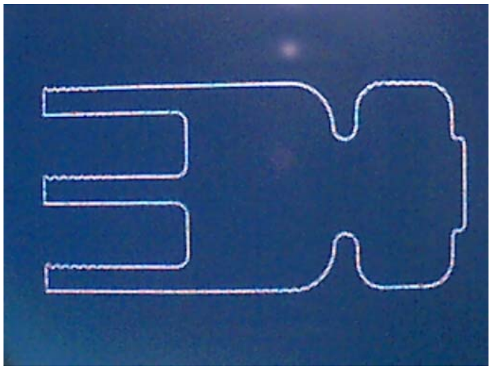


Fig.5 LTCC micro-combustor pattern

Once the pattern was cut into the LTCC tape by the laser, one remove the material outside of the pattern outline and utilize the resulting units for lamination.

The resulting cavity in the LTCC tape unit, served as the combustor cavity. Multiple tape units were laminated and the resulting cavity filled with the graphite sacrificial insert. Lamination is the method or process utilized to bond all the LTCC tape sheets as to construct a monolithic 3-D structure upon heating under a stress. When laminating LTCC tapes, it is important to keep all the sheets consistently with the same side up, that is, the LTCC tapes are fabricated over a Mylar sheet. To facilitate the release of the LTCC from the Mylar, a lubricant is utilized. It is important that the “shinny” side (side facing the Mylar) is always up or down.

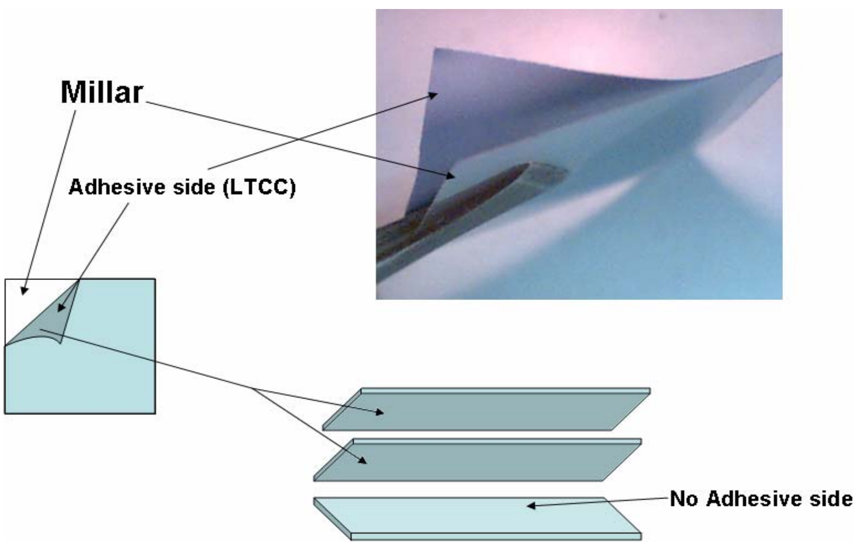


Fig.6 Consistent orientation of LTCC sheets before lamination.

The LTCC sheets at both ends of the laminate (top and bottom) are protected from the hot plattens in the hydraulic press by Mylar sheets. During lamination, the stress and time are controlled for best results. In our case we laminated at 1000 psi for 20 minutes at a platten temperature of 80 C.

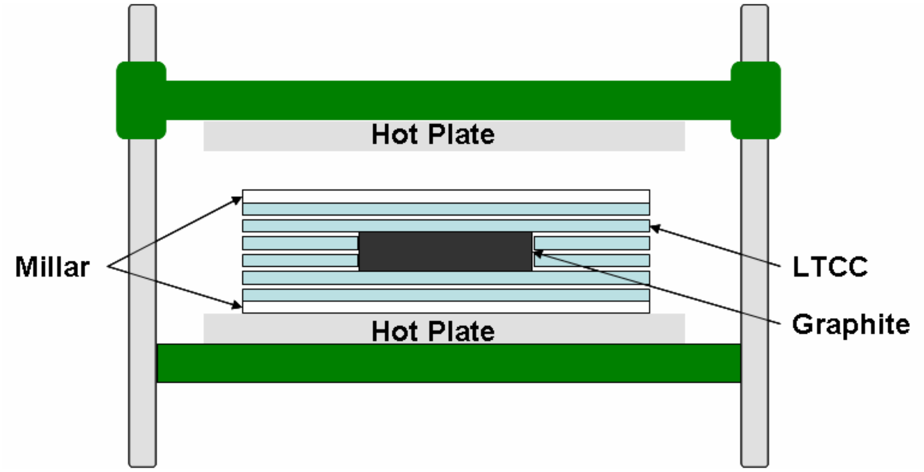


Fig.7 Scheme of the Lamination Process for the combustor

LTCC sintering

Sintering of the LTCC is the process in which green tape changes from a compliant plastic to a rigid solid and from a clear blue color to a darker hue. Sintering occurs after the structure has been laminated and it consists in placing the laminated structure in a furnace (LTCC and Graphite) and heat treat the composite to 850 C following a programmed sequence of heating steps. The structures to be sintered are placed on a alumina substrate as to avoid deformations conformal to the furnace substrate.

Fig9. Alumina substrate.

After the structure has been sintered one should observe that the graphite has disappeared and the perforations or cavities in the LTCC structure are clean.

The picture below is an example of a practice run, where we laminated LTCC tapes and used as a sacrificial material several pencil leads (graphite). The morphology is obviously that of a cylindrical channel, as can be observed.

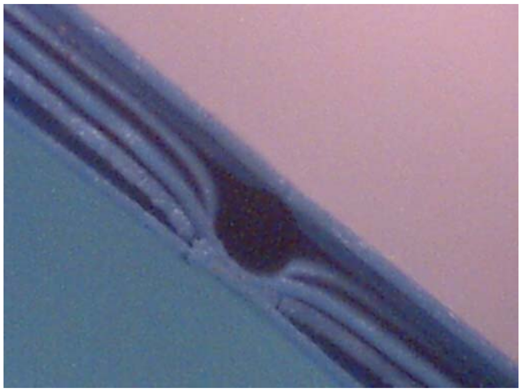


Fig10. LTCC sintering example using a pencil lead as sacrificial material.

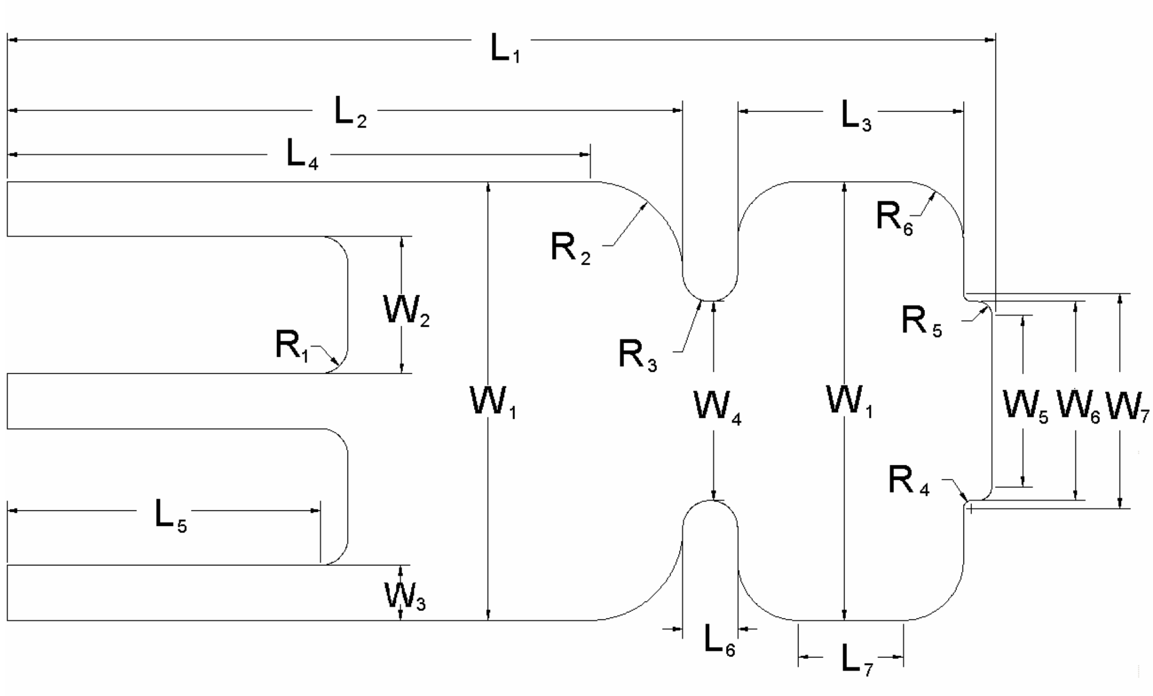
In order to obtain a preview of the flow behavior within the device, numerous simulations have been made using a commercial program call FEMLAB. This program will take into consideration a diversity of measurable parameters such as the speed, pressure, fluid Reynolds Number, and others. All simulations are two dimesional in natrue and they explore different configurations, morphologies, and conditions on which the combustor might be operated. The bulk of the simulations were concerned with the fluid mechanics. We used the Navier-Stokes formulation, preserving non-linearities and for convective and diffusive transport, the k.ε pertubatory approach. We were concerned with the effect of combustor geometry and volumetric flow rate of both fuel and oxidizer on the mixing. We would like to have a “pre-mixed” flame with a homogeneous composition near stoichiometry. All the simulations provided us with some insight into what was important concerning geometry (by indicating “dead zones” for mixing and particular configurations leading to mixing)

Combustor architecture

Below is a detailed description of the combustor morphology and configuration. There basically two competing forms, differing in length.

2. AutoCAD2000i based architecture diagrams.

Below is a detailed scheme of the combustor architecture. All the dimensions are in the SI system.



Number	Dimensions (millimiter)					
	L	W	R	1.5L	1.5W	1.5R
1	18.0641	8.0304	0.5	27.9615	12.0456	0.75
2	12.3539	2.5063	1.7	18.5309	3.7595	2.55
3	4.1274	1.0074	0.5	6.1911	1.5111	0.75
4	10.6538	3.6379	0.15	15.9807	5.4569	2.25
5	5.7291	3.1378	0.25	8.5937	4.7067	.375
6	0.9991	3.6378	1.1013	1.4987	5.4567	1.5195
7	1.9248	3.9378		2.8872	5.9067	

Some experimental details

Materials and tools

The tools and materials utilized in the fabrication of these devices were:

1. Green tape type 951AT from Dupont, Delaware, PA, U.S.A (approximately 100 µm thick).
2. Isotemp Programmable Forced-Draft Furnace (Fisher Scientific)
3. Heated press (Carver Model C)
4. Graphite (Arrow Springs, CA,U.S.A)
5. commercially available Alumina substrates
6. Computer Numerical Control (CNC) from Fadal
7. X-660 Laser Platform (Universal Laser Systems, Scottsdale, AR, U.S.A., 60 W CO_2 laser, wave length of 10.6 µm)

Computer Numerical Control (CNC) milling machine processing

Using a band saw a piece of graphite of 2mm thickness was obtained. A face mill tool in the CNC was used to smooth the graphite facing the LTCC tapes. The FACE Mill is a 2 in diameter cylinder with 3 carbide tips capable of polishing the graphite to less than 2 mils.

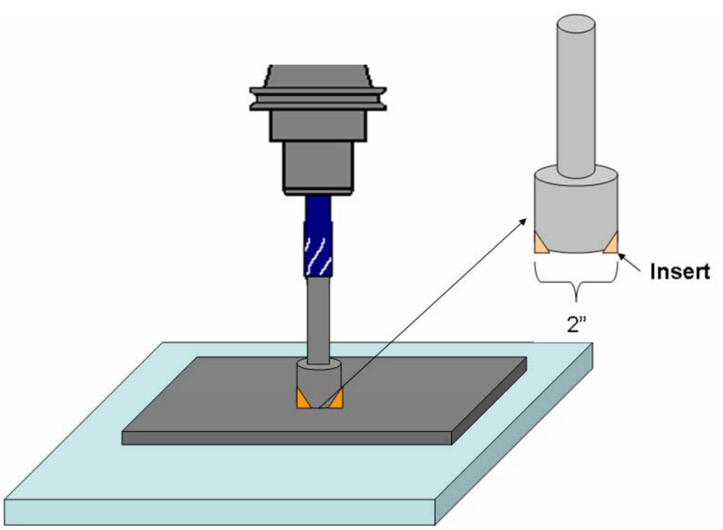


Fig11. Smoothing the graphite faces.

Fig13. Finished graphite insert

Some further details of the LTCC processing

Before attempting the fabrication of the combustor, we calculated the number of LTCC tape sheets needed for the laminated structure. The DuPont 951 LTCC is about 4 mils thick (around 100 μm) and for one of the possible combustor insert thickness (2mm), at least 20 layers were needed. The schematic below depict some of the process details.

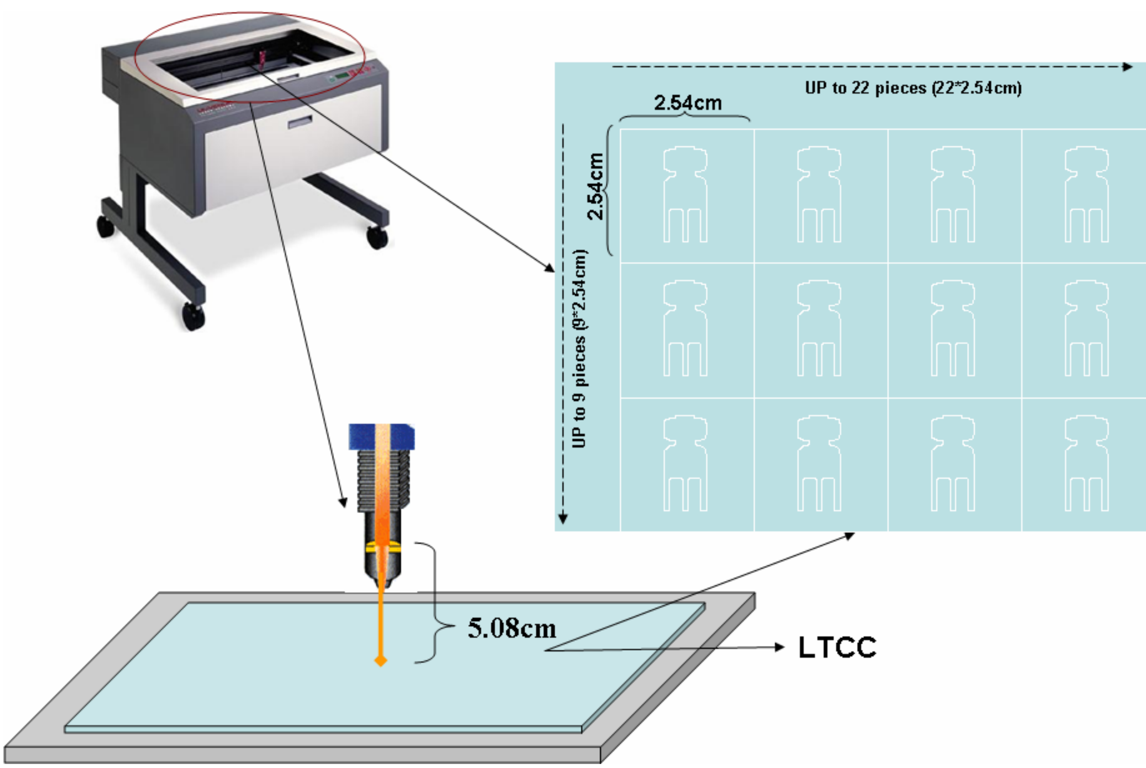


Fig14. Laser Process

The laser head velocity and power were the parameters best utilized for the LTCC tapes cutting. The cutting power was a strong function of where in the tape sheet you wanted to cut. If you wanted to cut far from the periphery (inside), then the power applied was 3% of the maximum power and the velocity 5 % of max. The outside (near the periphery) was 0.05 P_{max} at 0.05 V_{max} as depicted below.

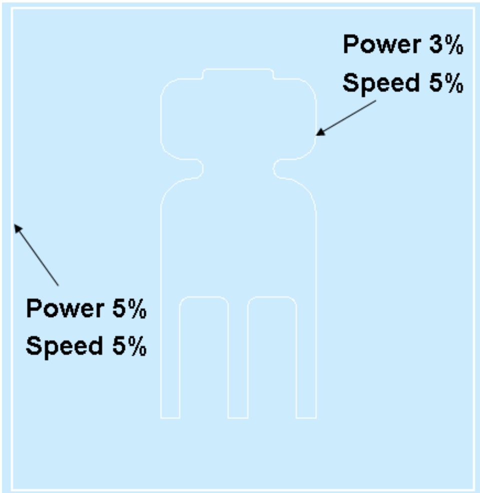


Fig15. Power (%) and Speed (%) for cut of LTCC

Some further details of the lamination Process

Again, twenty layers of LTCC were laminated around the graphite insert. Below one can note the near plastic behavior of the LTCC before firing (green tape), and stacked before lamination with the 20 LTCC layers.

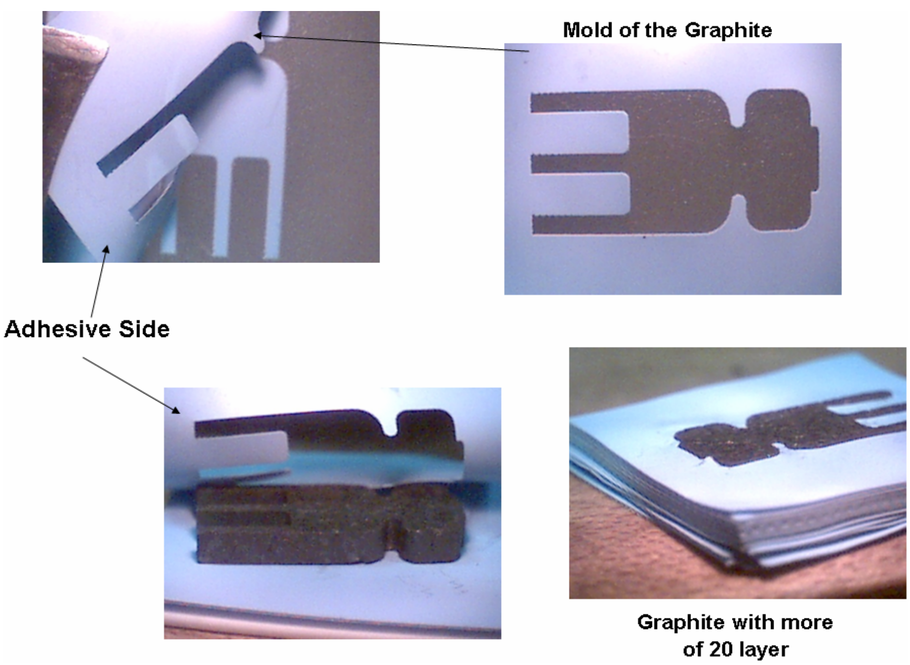


Fig16. Lamination process before pressing.

For the nominal dimensions structure a uniaxial pressure of 1000 to 1300 psi was utilized. For the 1.5 scale, the uniaxial pressure was incremented to 3300 psi. Both structures were exposed to temperatures in the range from 80 to 120C for a period ranging from 15 to 30 minutes.

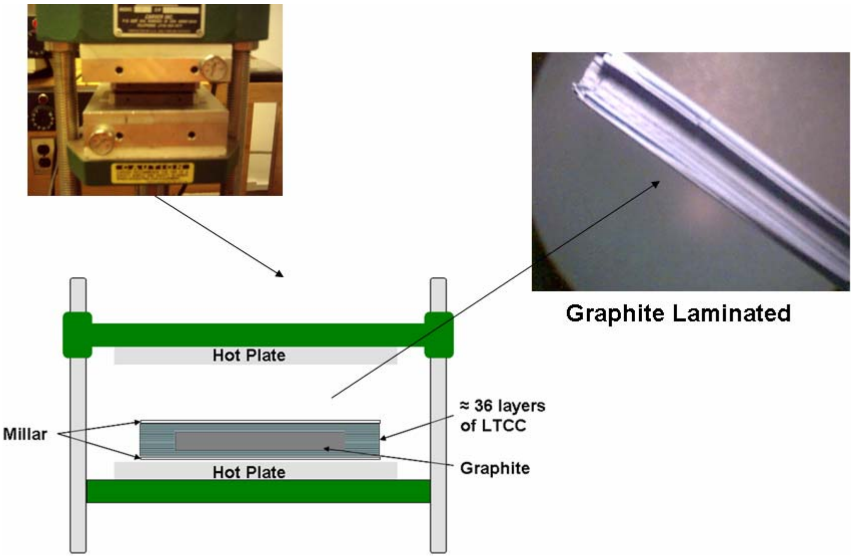


Fig.17 The lamination press

Sintering Process

The laminated structure was placed on a fully sintered Alumina substrate and heat treated as described below.

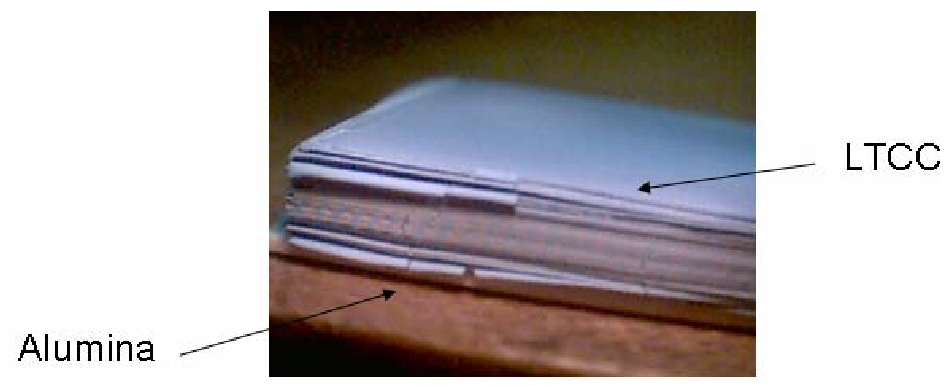


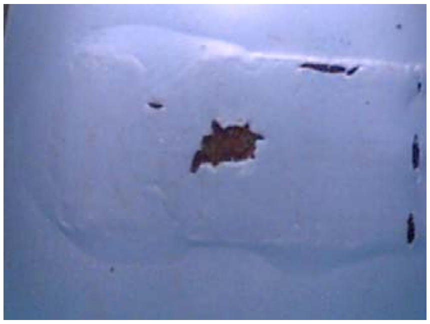
Fig18. Alumina and LTCC

The heating schedule as programmed in the resistance furnace:

- From room temperature to 300º C at a rate of 10º C/min.
- Kept at 300 C for 30 minutes
- Ramp from 300 C to 850º C at a rate of 10º C/min
- Kept at 850º C for 1 hour and 45 minutes.
- Turn furnace off and let it cool to room temperature.

Overall results

After sintering, the structures were examined for termination details. For example to what extent the sacrificial material has been utilized (oxidized, disappear)

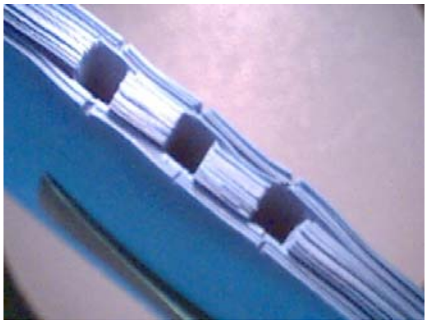


a) Simple Scale

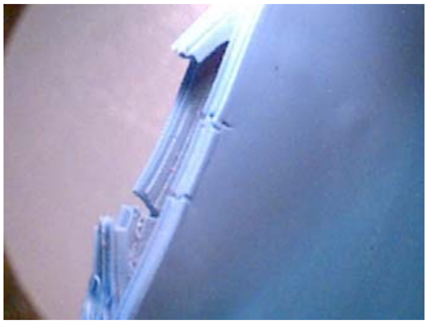
b) 1.5 Scale

Fig19. Cavity in the combustor surface to evaluate graphite gasification.

The gas inlets and outlets of the initial trial I participated yield the following results.



a)



b)

Fig20 Micro-combustor inlets (a) and outlet (b).

The top laminate inlet / outlet orifices after sintering looked, as depicted below. Both the lateral and top laminate I /O ports were considered marginally usable.

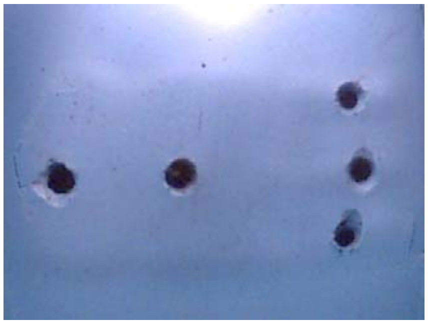
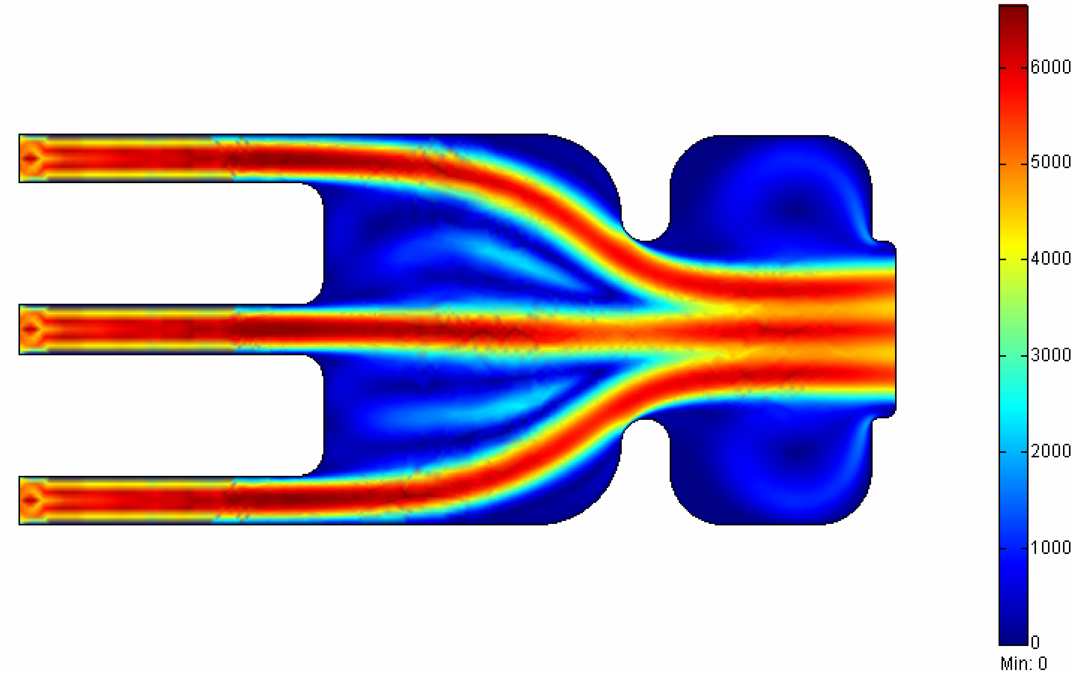
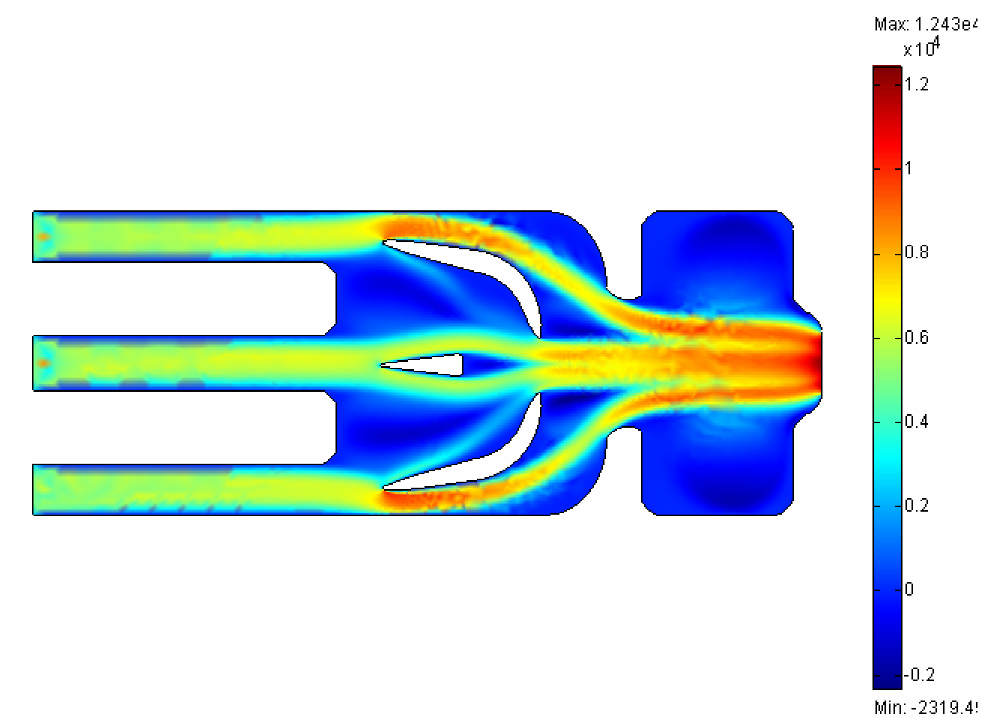


Fig.21 Inlet / outlets on the top laminate of a sintered micro-combustor.

Numerical Simulations.

To obtain some insight that might help us in the design of the combustor, multiple numerical simulations using the commercial package FEMLAB were realized. The fluid mechanic simulations yield surface velocity (mm/s) on a 2-D structure. The inlet velocity (Reynolds number) was varied as well as some structural and configuration parameters.





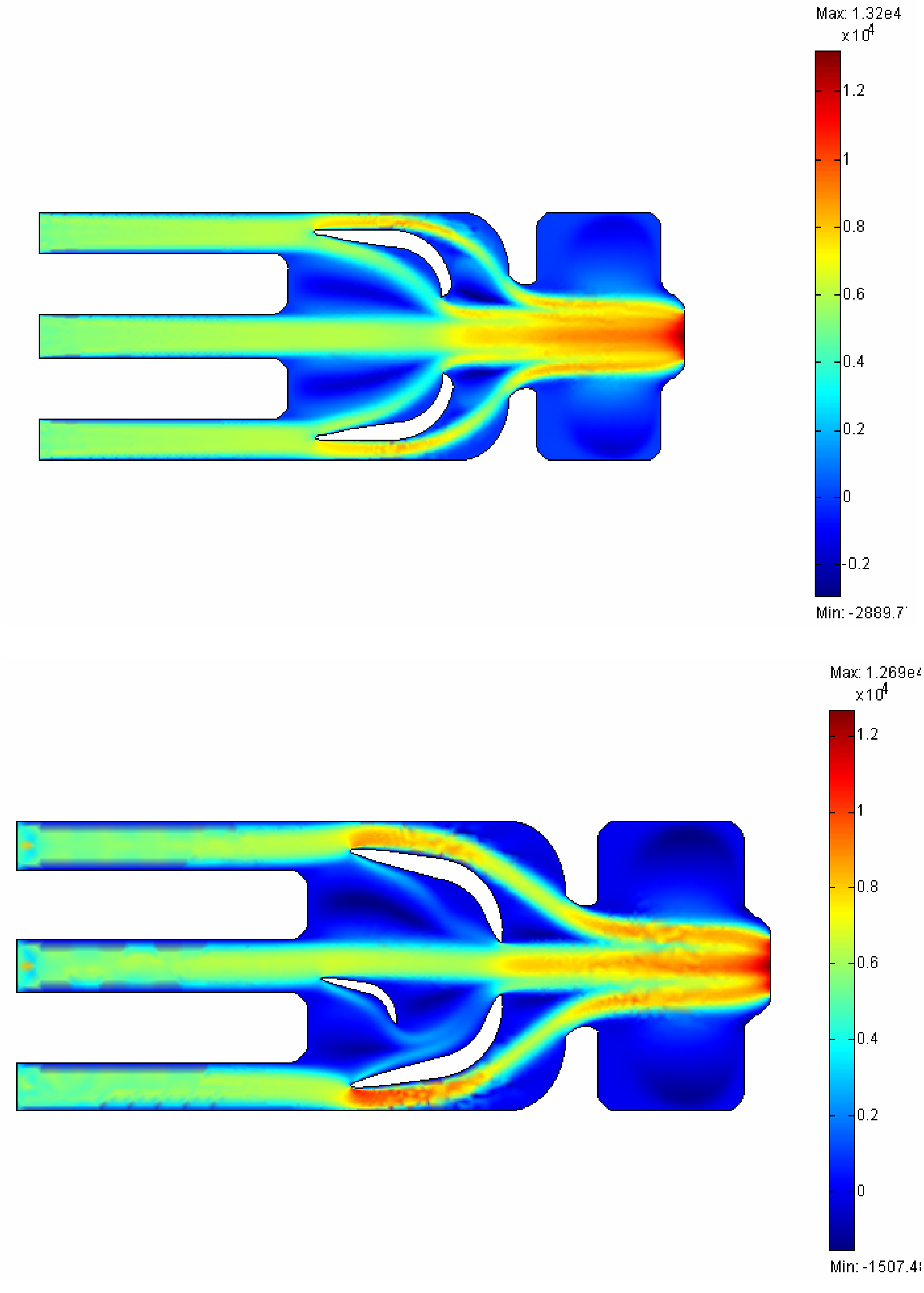


Fig22 Samples of simulation results for different flow rates and configurations of the combustor geometry. The false color map gives the surface velocity as depicted in the scale in the right of the figure.

Conclusions

One of the first things we learned in processing the combustor is that the sacrificial material, in this case graphite, might be difficult to gasify if the process is not done carefully. A larger outlets for the gasification products (CO and CO_2), drilling holes in the graphite insert and increasing the sintering temperature and time. Since we did only two runs, it was difficult for us to find a processing window for the gasification. Another structural issue was that the number of layers in both the top and bottom has to be increased to avoid stress induced de-laminations and cracking.

Our experiments showed that having at least 8 LTCC tape layers in both top and bottom circumvent the stress induced mechanical failures.

The combustor edges easily deformed when sintered without lateral support, suggesting that perhaps we need a sacrificial mold in the outside of the combustor structure to avoid sagging and deformation.

No doubt that LTCC tapes with graphite as a sacrificial material form an inexpensive and easy to manipulate materials for combustor applications as compared to metals and nitrides as previously reported in the literature.

Recommendations

An inexpensive and convenient material as a lamination aid (glue) is honey. Common honey burns all its volatiles before 300C and possesses the viscosity necessary for lamination. It was used by us, in the formation of a cylindrical channel with a pencil lead as sacrificial material and works like a champ. One can see in the figures below how difficult it is to laminate around a cylindrical structure. Once honey was used to enhance the lamination, de-lamination problems were minimized.

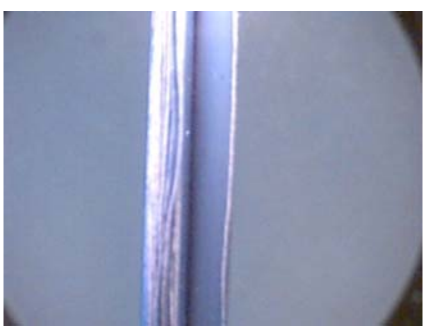


Fig23. Lamination failure (de-lamination) using pencil-lead as sacrificial material.

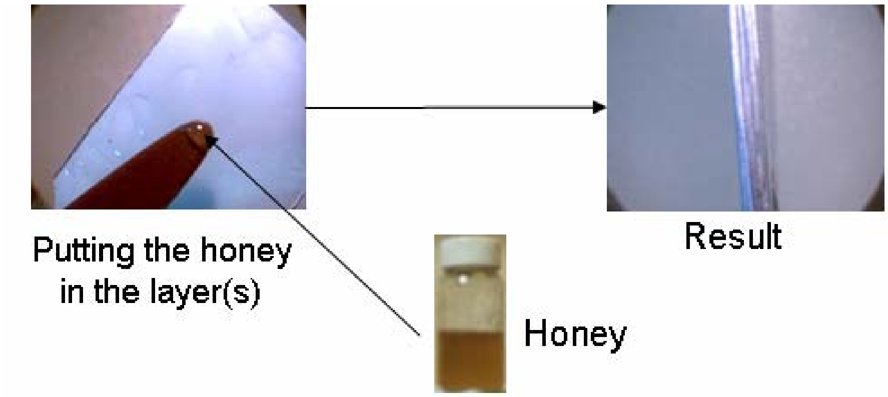


Fig24. Well laminated channel using pencil-lead as a sacrificial material.

Another significant improvement in preserving symmetry and surface smoothness is achieved by placing the laminated structure between two fully fired alumina sheets during sintering.

Another recommendation borne by the simulation results is to elongate the combustor outlet to avoid de-laminations and cracking, as suggested by the figure below.

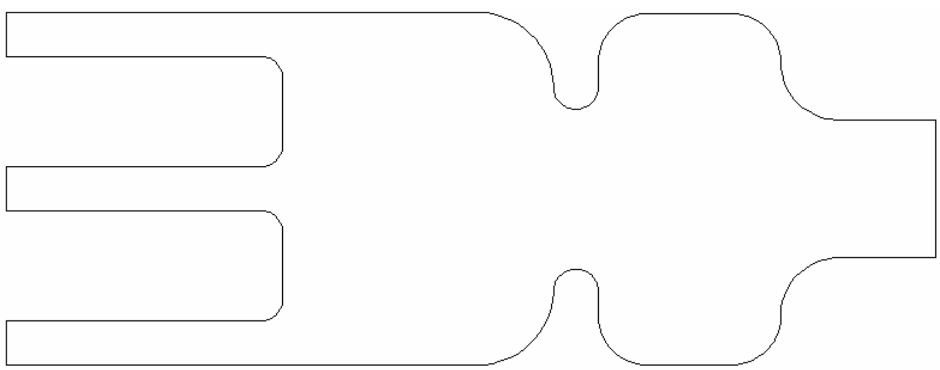


Fig. Proposed configuration and morphology to avoid thermal stresses damage during sintering and perhaps operation.

Acknowledgements

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Reference

[1] Lesley Millar, “Novel Microcombustor for Highly Efficient Generation of Electric Power from Fuel”, *Office of Technology Management*, University of Illinois at Urbana.

[2] Shuji Tanaka, Takashi Yamada, Shinya Sugimoto, Jing-Feng Li, and Masayoshi Esashi “Silicon nitride ceramic-based two-dimensional microcombustor”, *Journal of Micromechanics and Microengineering*, 13 (2003) 502–508.

[3] Lars Sitzki, Kevin Borer, Ewald Schuster and Paul D. Ronney, “Combustion in Microscale Heat-Recirculating Burners”, *The Third Asia-Pacific Conference on Combustion*, Seoul, Korea, June 24-27, 2001

[4] D. G. Norton, K. W. Voit, T. Brüggemann, and D. G. Vlachos, “Portable Power Generation Via Integrated Catalytic Microcombustion-Thermoelectric Devices”, Department of Chemical Engineering,University of Delaware.

[5] Miguel Perez Tolentino, Rogerio Furlan, Idalia Ramos and Jorge J. Santiago Aviles “Study of Laser Milling Of Sintered LTCC and Quartz Substrates for Microfluidic Applications”, *The National Conference On Undergraduate Research (NCUR) 2005*,Washington and Lee University Virginia Military Institute Lexington, Virginia, April 21– 23, 2005.

[6] M.R. Gongora-Rubio, P. Espinoza-Vallejos, L. Sola-Laguna, J.J. Santiago-Aviles, “Overview of low temperature co-fired ceramics tape technology for meso-system technology (MsST)”, *Sensors and Actuators A 89*, 2001, pp. 222-241.

[7] E.W. Simoes, I. Ramos, L. Garcia, R. Furlan, J.J. Santiago-Aviles, and M.T. Pereira, “Numerical modeling of a process for definition of features in low temperature co-fired ceramics”, *in Electrochemical Society Proceedings*, Volume 2002-8, Porto Alegre (Brazil), SBMicro 2002, September 9-14, 2002, pp. 244-252.