Development of A Low Temperature Co-Fired Ceramic Fuel Processor for the Micro-scale Solid Oxide Fuel Cell System

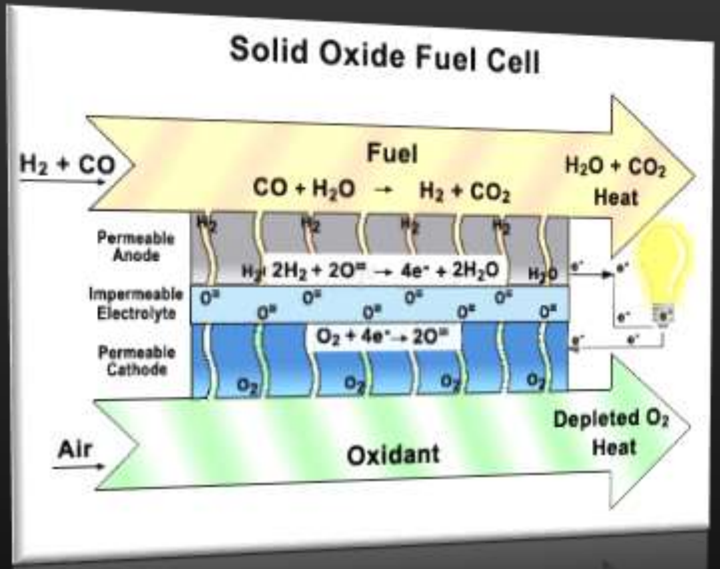
B. Jiang[1,2], A. J. Santis-Alvarez[3], P. Muralt[1], D. Poulikakos[3], T. Maeder[2]

[1]Laboratory of Ceramics, EPFL Lausanne, CH-1015 Lausanne, Switzerland
[2]Laboratory of Micro-engineering for Manufacturing, EPFL Lausanne, CH-1015 Lausanne, Switzerland
[3]Laboratory of Thermodynamics in Emerging Technologies, ETH Zurich, CH-8092 Zurich, Switzerland

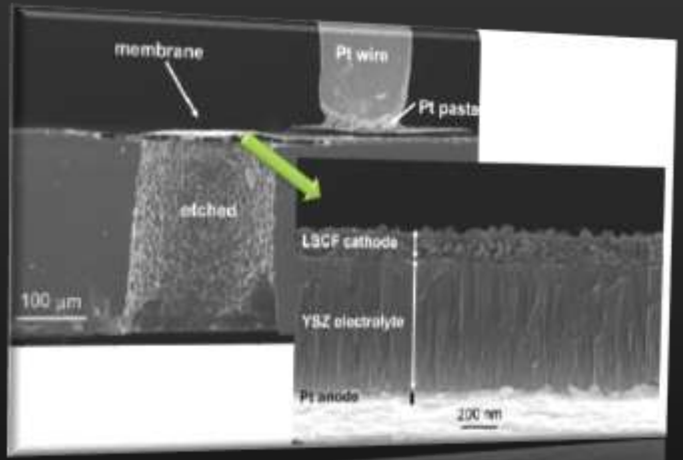
Outline

- Introduction
  - Concept of the micro-scale solid oxide fuel cell system (µ-SOFCs)
  - Concept of the µ-SOFC fuel processor
  - Low temperature co-fired ceramic (LTCC) technology
- Design of LTCC fuel processor
- Fabrication process
- Fluidic, thermal and thermally self-sustain characterization
- Conclusion
- Outlook

What are μ-SOFCs?



Principle of SOFC



Silicon base μSOFC membrane

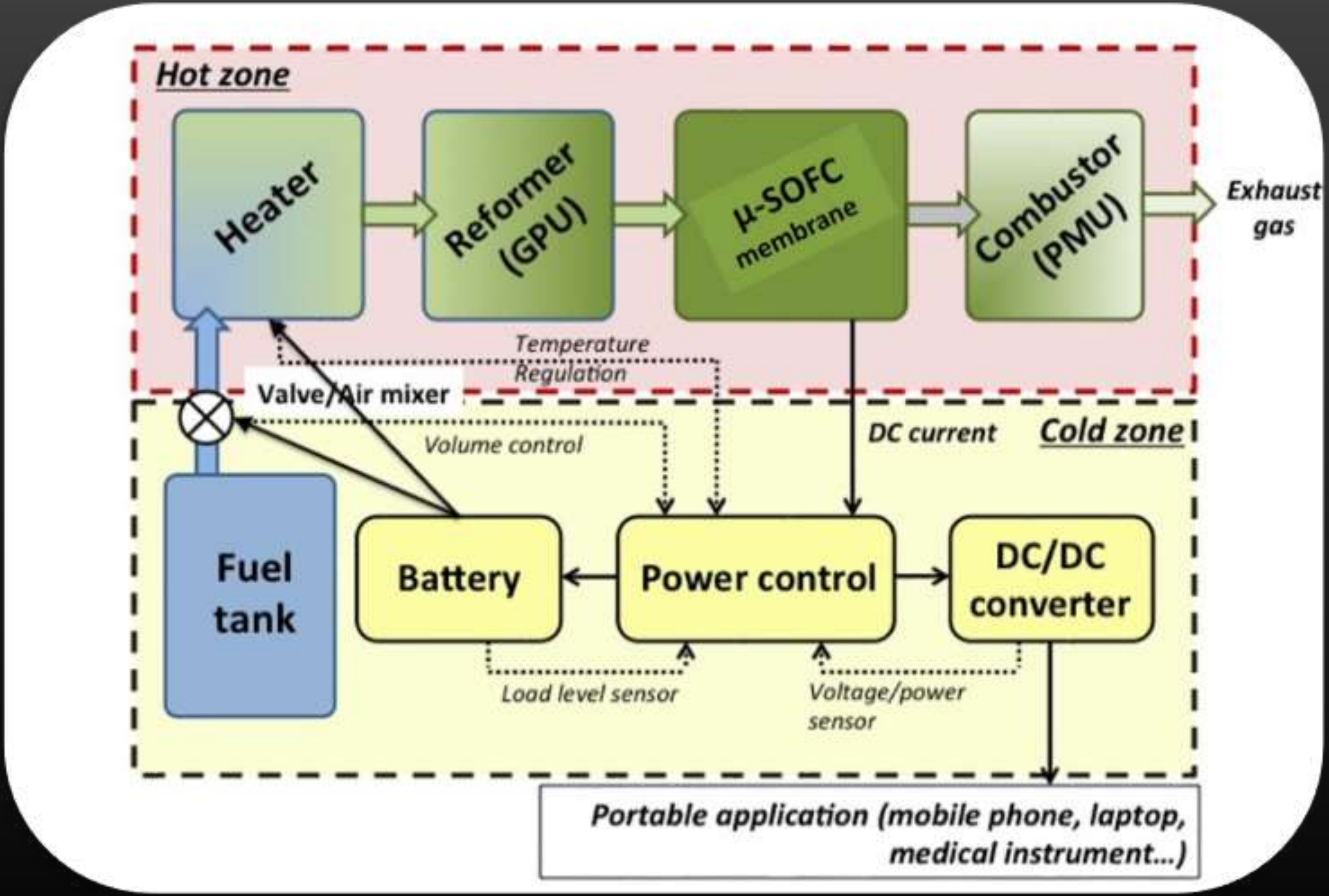
2008 Bieberle, J. Power Sources

1st commercialized μ-SOFC product

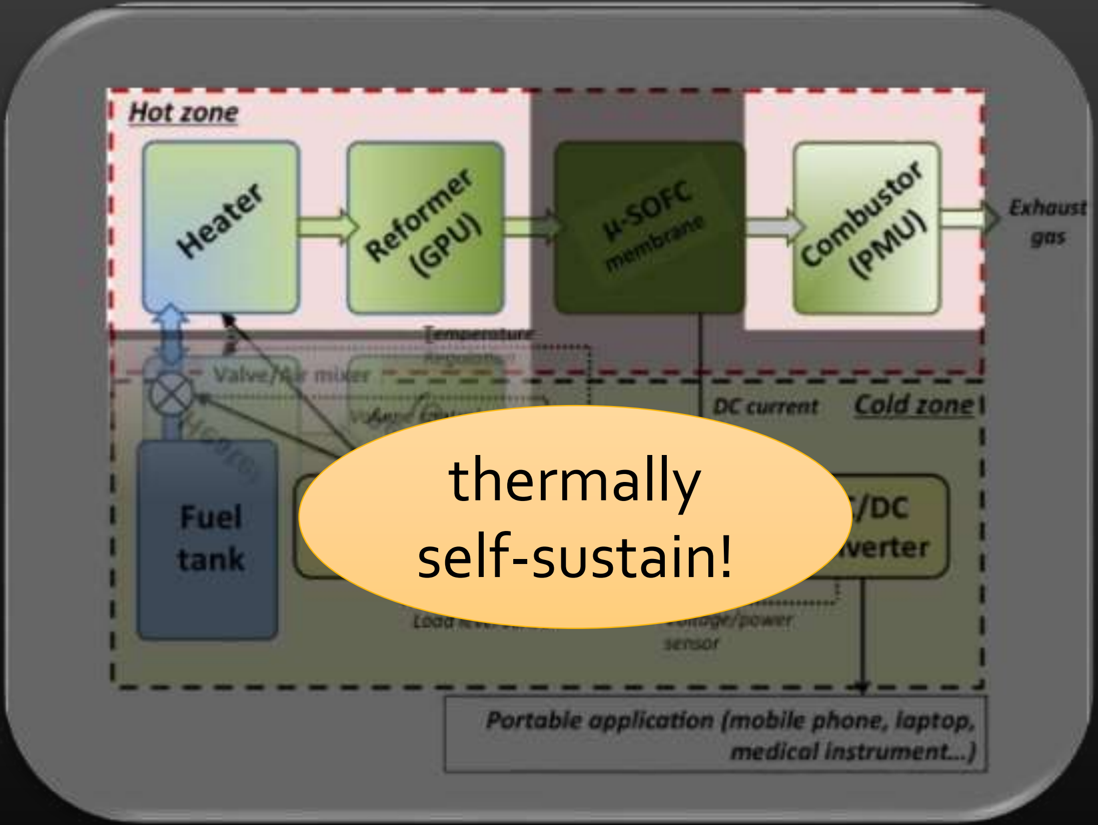
Lilliputian Systems, Inc.

What is a μ-SOFC fuel processor?

Concept of μ-SOFCs system

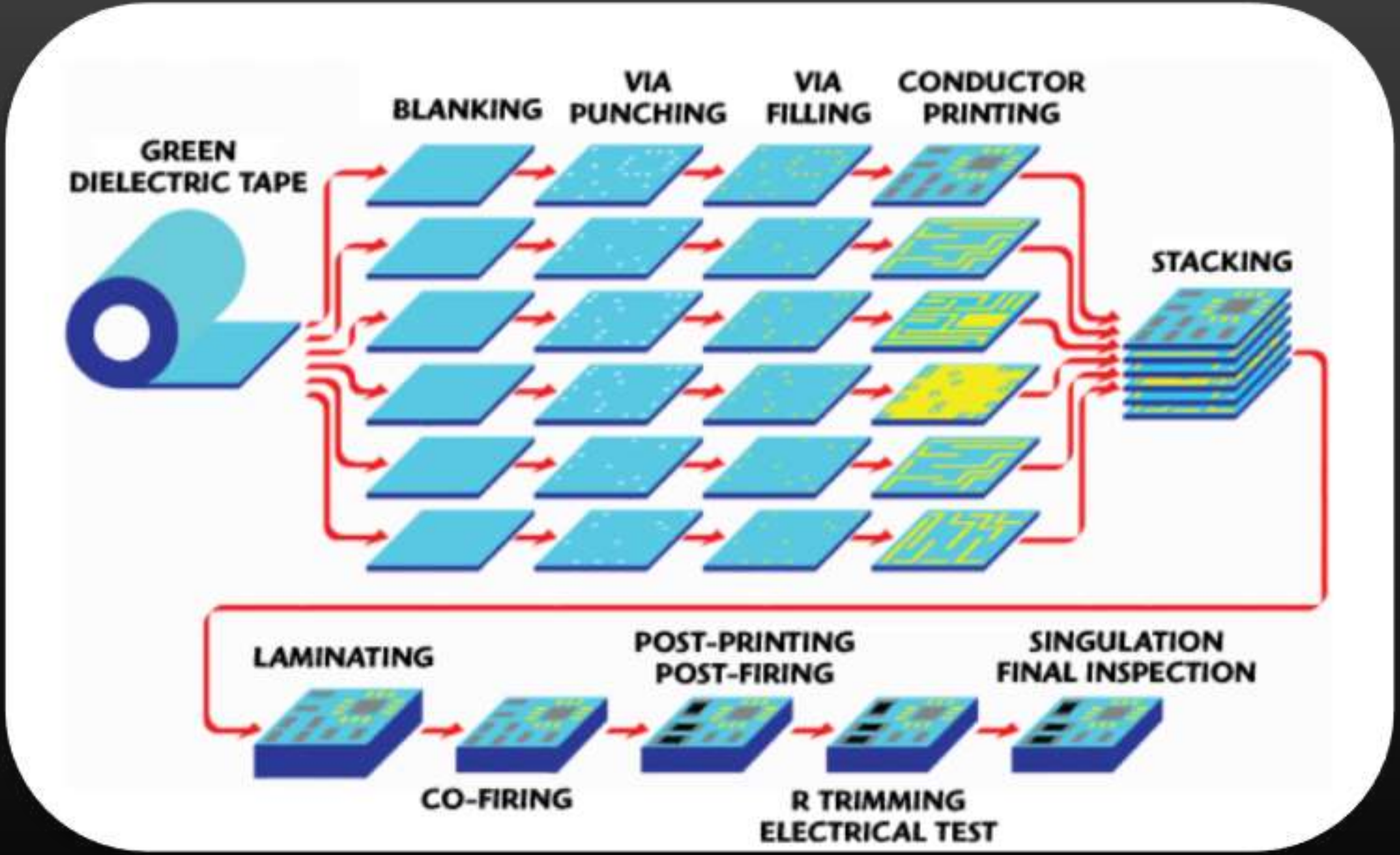


What is a μ-SOFC fuel processor?



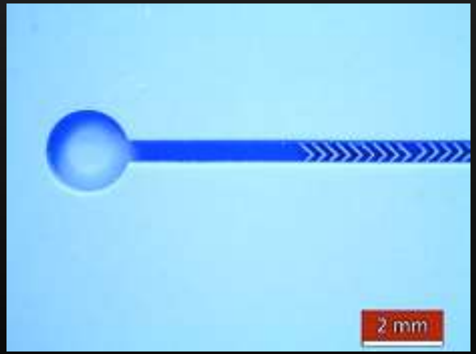
- The fuel processor is a vital component in the whole μ-SOFC system
  - Heater – providing initial thermal energy for trigging the chemical reaction
  - reformer – converting hydrocarbon fuel mainly into hydrogen and carbon monoxide with a partial oxidation reaction route
  - combustor – totally oxidizing exhaust gas from the μ-SOFC membranes
  - thermally independent operation for minimizing the power input to the system

What is LTCC technology?



http://www.microwavejournal.com

LTCC flow sensor



LTCC microreactor

Design of a LTCC fuel processor

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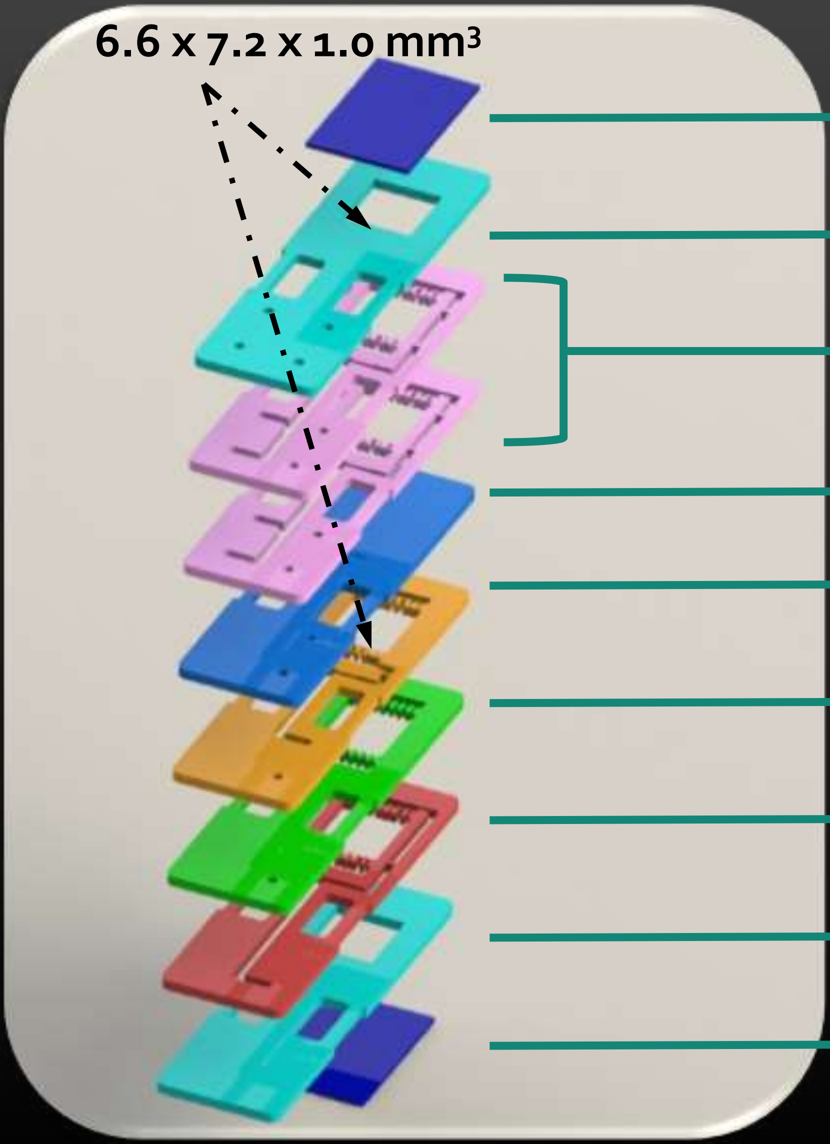
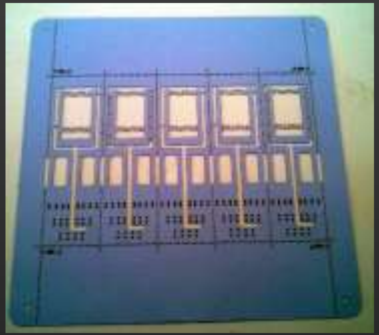
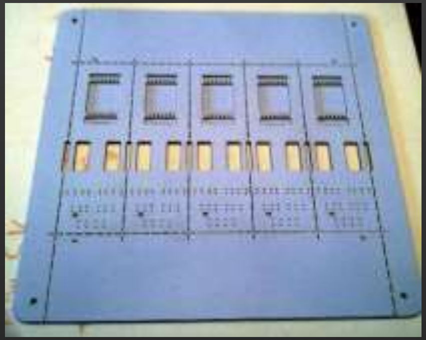
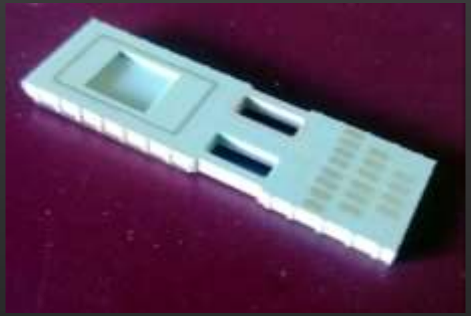


- Cap
- Top cover layer
- Packed bed 1 – fuel/air mixture inlet — reforming
- Thick-film heater — Heating
- Packed bed 2 – air fluidic channel
- Packed bed 2 – separation layer
- Packed bed 2 – exhaust gas outlet
  — combustion (Packed bed 2 – separation layer and exhaust gas outlet)
- Bottom cover layer with electrical connection pads
- Cap

Fabrication of LTCC fuel processors





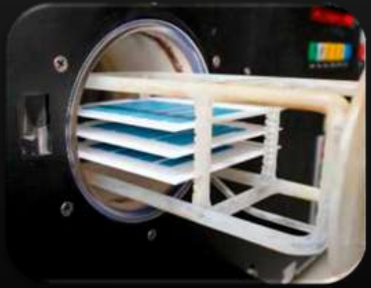


Laser ablation
(a Nd-YAG laser, LS9000)

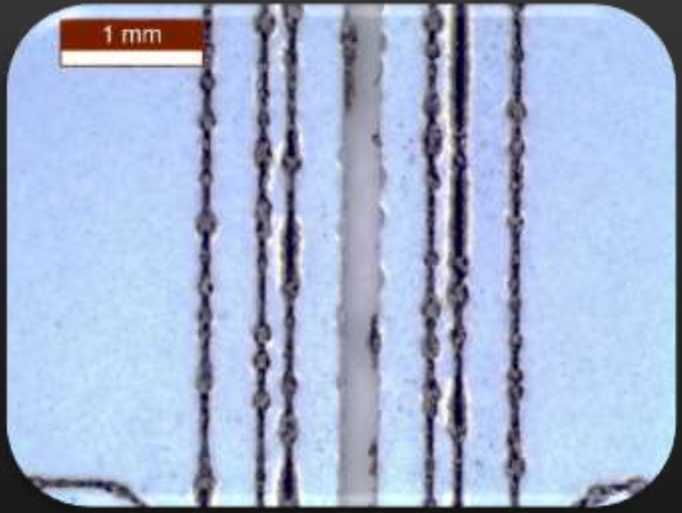
Screen printing
(Aurel C900)

Uniaxial lamination
(2 – 20 MPa
25°C – 70°C)

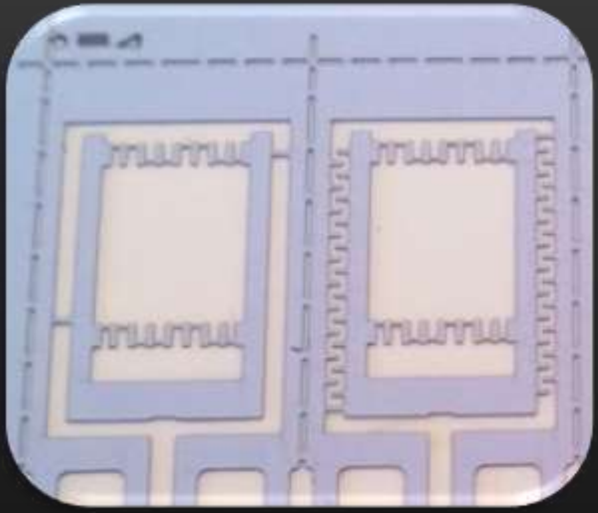
Sintering
(ATV PEO-601 lamp furnace
10 hours – 875°C – Air)



Fabrication of LTCC fuel processors



Bad



Good

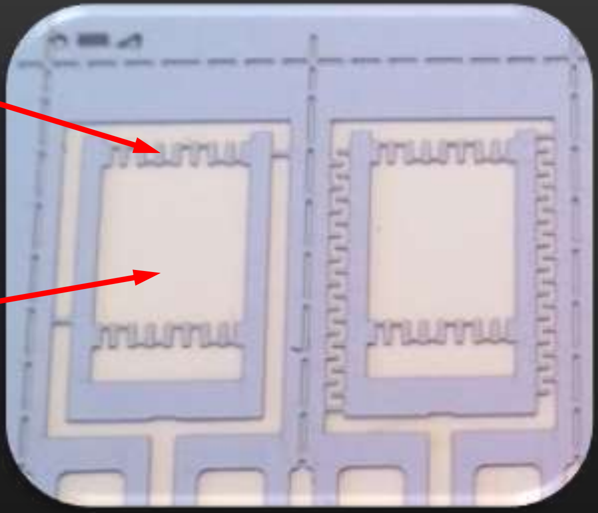
Channel and cavity formation

- Laser cutting parameters
  a. I - diode current (A)
  b. f - frequency of the acoustic optical switch (kHz)
  c. v - beam deflection velocity (mm/s)
- Inadequate laser parameters may result in channel sidewall bulging or heavy burnt of the cutting interface to destroy the fine fluidic structure on LTCC

Fabrication of LTCC fuel processors

Strips for the catalyst barrier (200 µm)

Cavities for the catalytic packed bed (6.6 x 7.2 mm^2)



Good

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Fabrication of LTCC fuel processors

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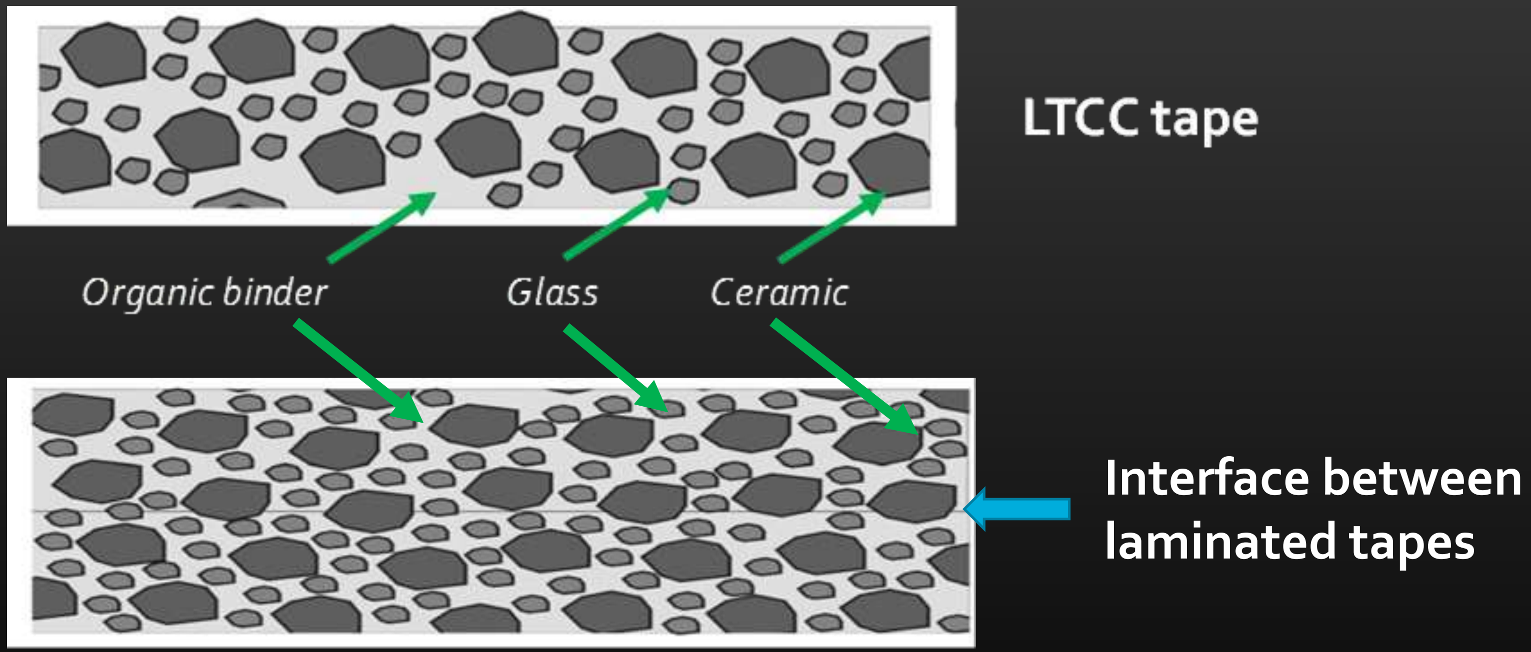
Lamination

Uniaxial lamination

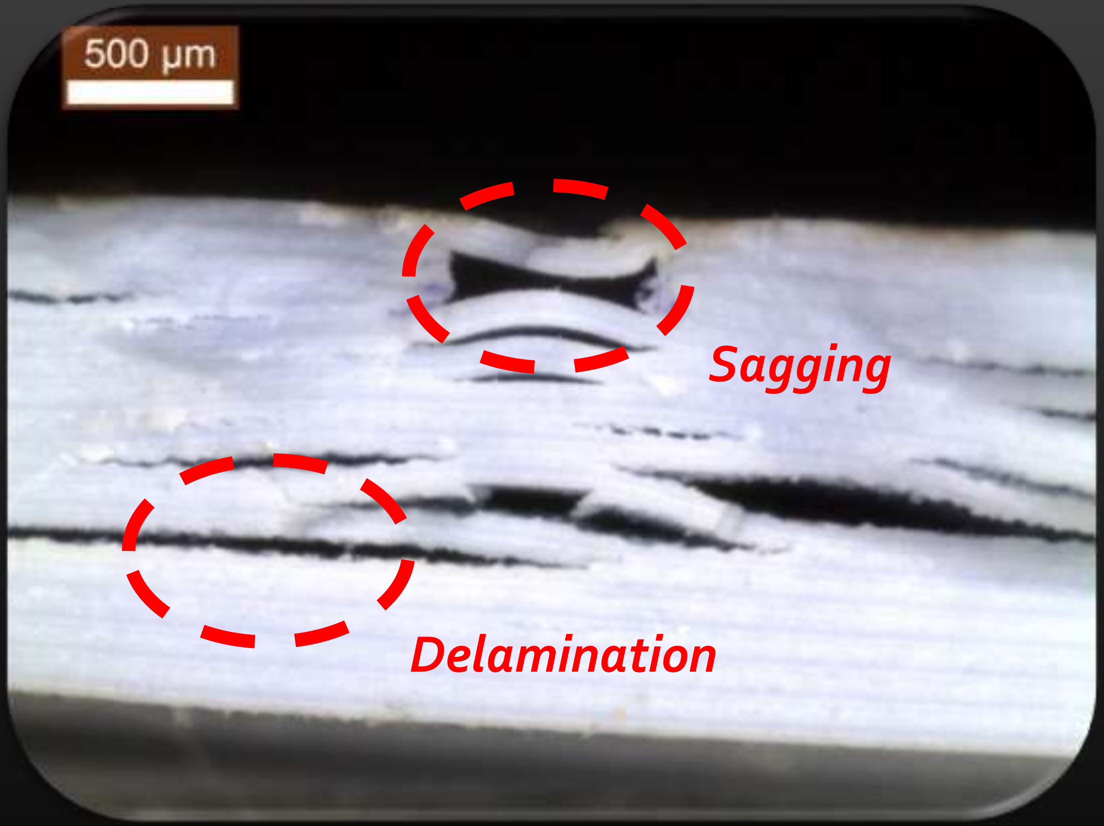


D.Jurków and L. Golonka, J.of the European Ceramic Society, 2012

Fabrication of LTCC fuel processors

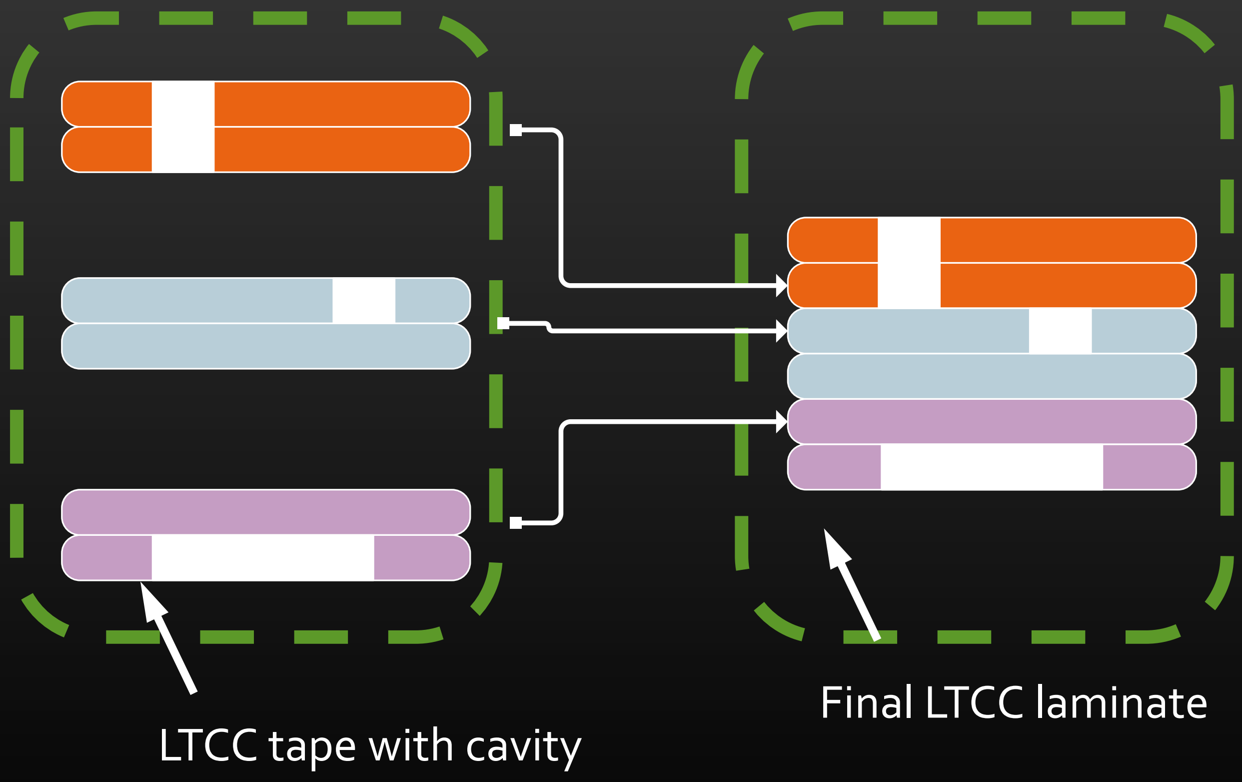
Lamination

- improper lamination causes sagging of fluidic channel and delamination of fired samples
- Progressive lamination provides a good solution to the fine, complex fluidic structure formation in development of the LTCC fuel processor



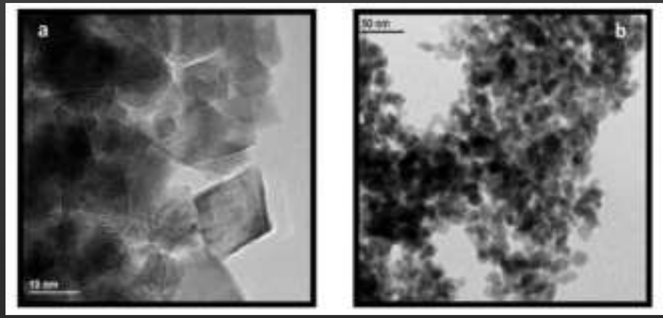
Fabrication of LTCC fuel processors

Progressive lamination procedure



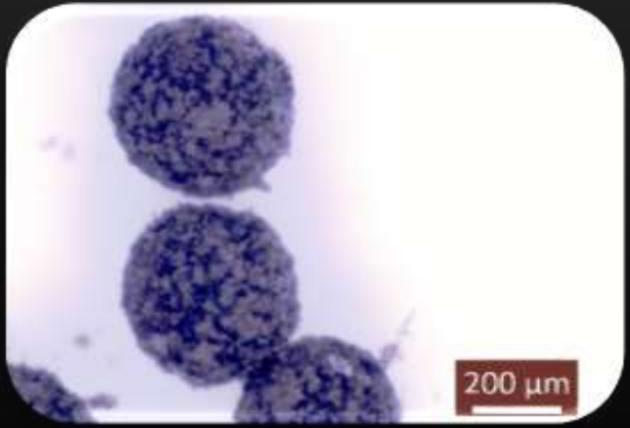
- For those tapes with same fluidic pattern are pre-laminated. The thickness of the fluidic structure was therefore built up by the lamination at room temperature with applied standard pressure (21 MPa)
- Each laminated functional layer were stacked and laminated together one by one with a applied low pressure (< 4MPa) at 70°C

Catalyst for the fuel reactions



Transmission electron micrographs of the as-prepared Rh/(Ce,Zr)O_2 catalyst particles

2008 Hotz, Chem. Eng. Sci.



10 wt% of Rh/(Ce,Zr)O_2 on YSZ beads support

Partial oxidation of propane

$C_3H_8 + 2O_2 \rightarrow 5H_2 + 4CO$

- Exothermic reaction
- High syngas (CO & H_2) production possible with Rh-metal catalyst

YSZ beads (diameter: 100 – 200 µm) as catalyst supports for building up the packed bed in the processor

👍 homogenous packed bed & low pressure drop
👍 no solvent needed for the filling
👎 low thermal conductivity – improper to obtain isothermal bed condition (alternative: high temperature alloy felt as catalytic bed)

Fluidic characterization

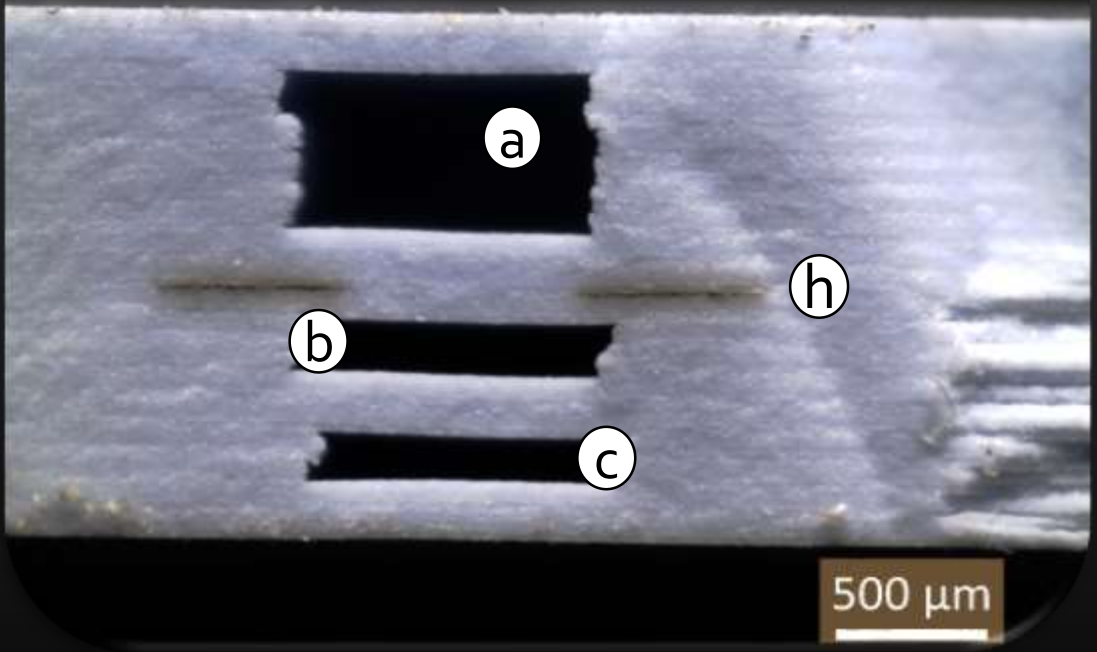
- Fluidic structure

a. Cross section of exhaust channel: 0.5 x 0.96 mm^2
b. Channel 1: 0.95 x 0.17 mm^2
c. Channel 2: 0.95 x 0.14 mm^2

- Heater (h)

a. screen printed thick-film platinum (CL11-6109 , < 50 milliohms per square, Heraeus Inc.)
a. Co-firing process
b. fired thickness ≈ 10 μm

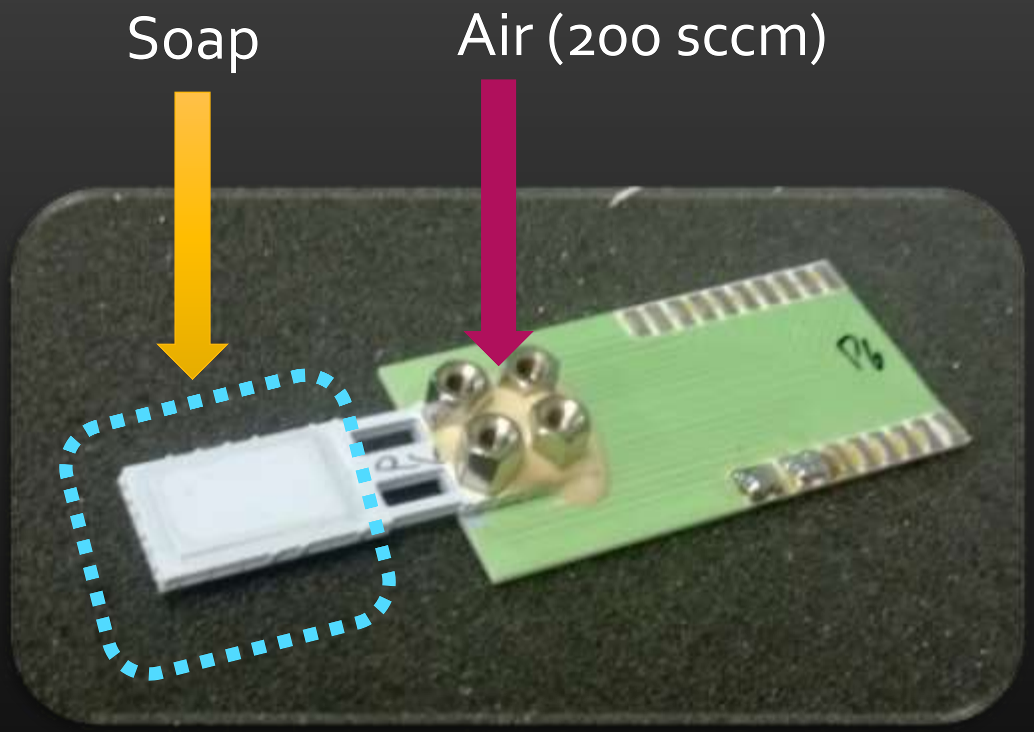
Cross-section of the LTCC processor



Fluidic characterization

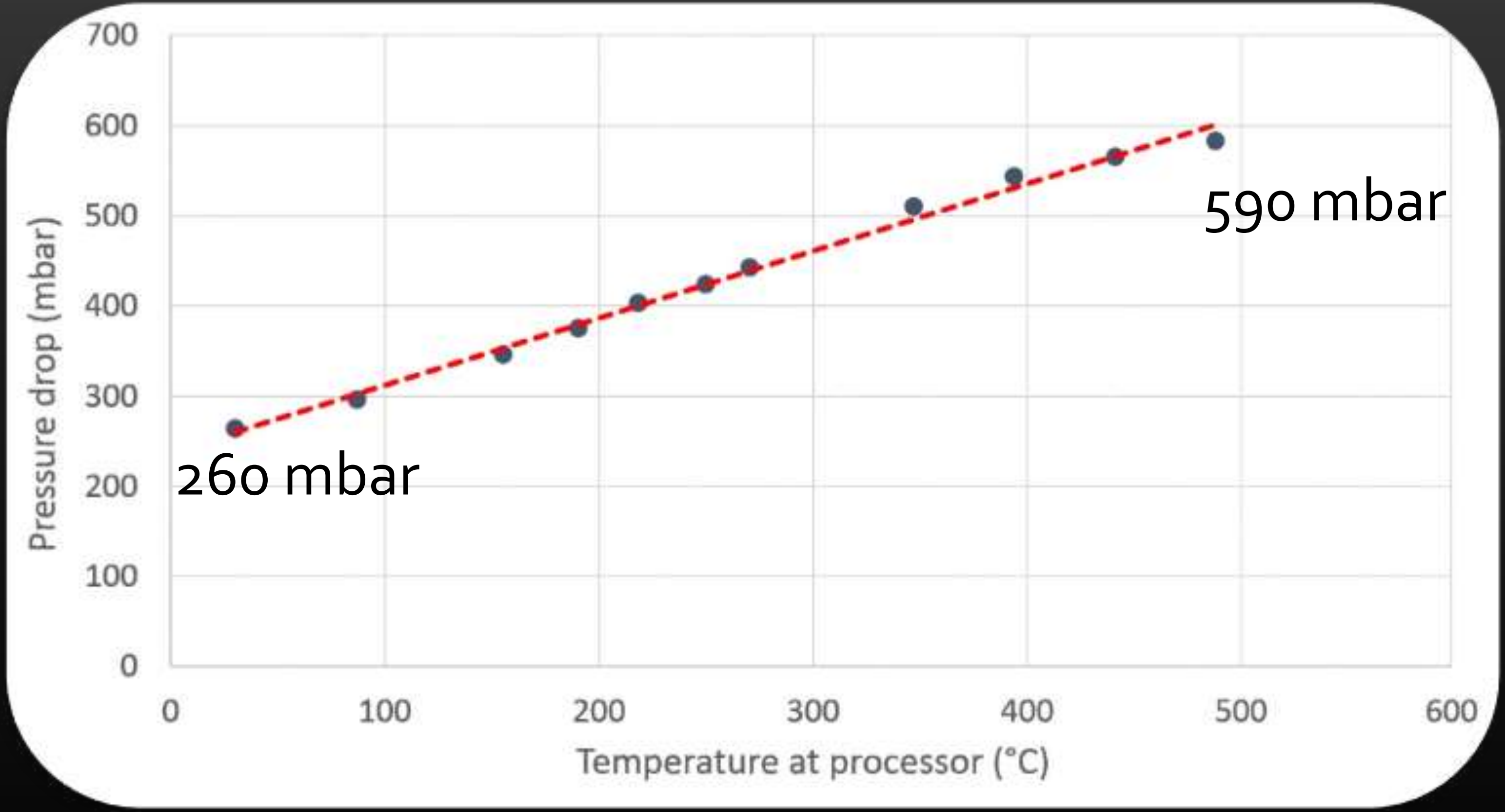
Hermeticity of as-prepared MSR

- Using ESL 4026 sealing glass for the processor packaging
- Simply using soap solution to check the leakage of sealed LTCC fuel processor under apply 200 sccm of air flow at room temperature
- No leaking was observed on as-prepared fuel processor



Fluidic characterization

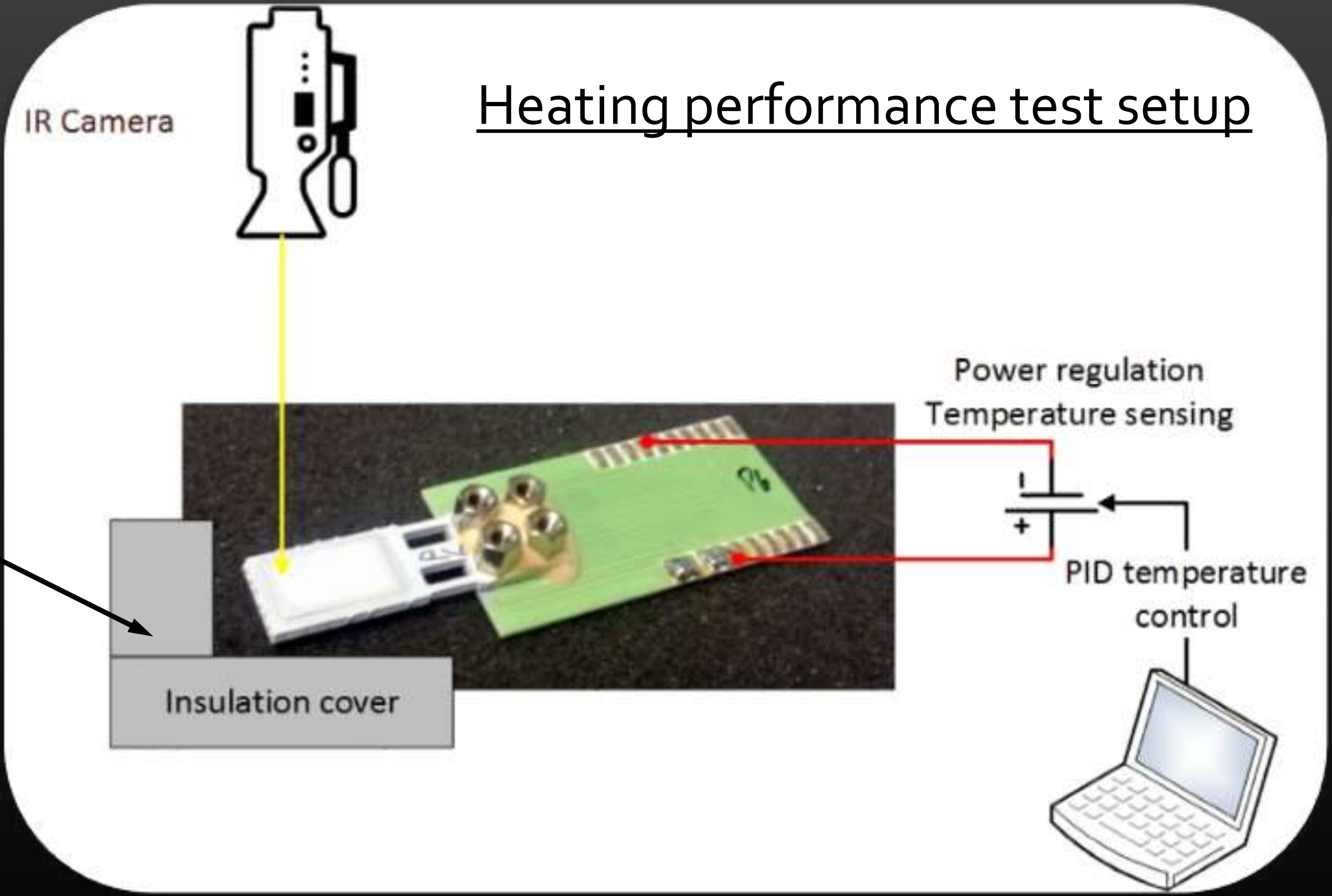
Pressure drop



- Flow rate of air: 200 sccm
- Moderate pressure drop across the processor
  590 mbar @ 490°C

Thermal characterization

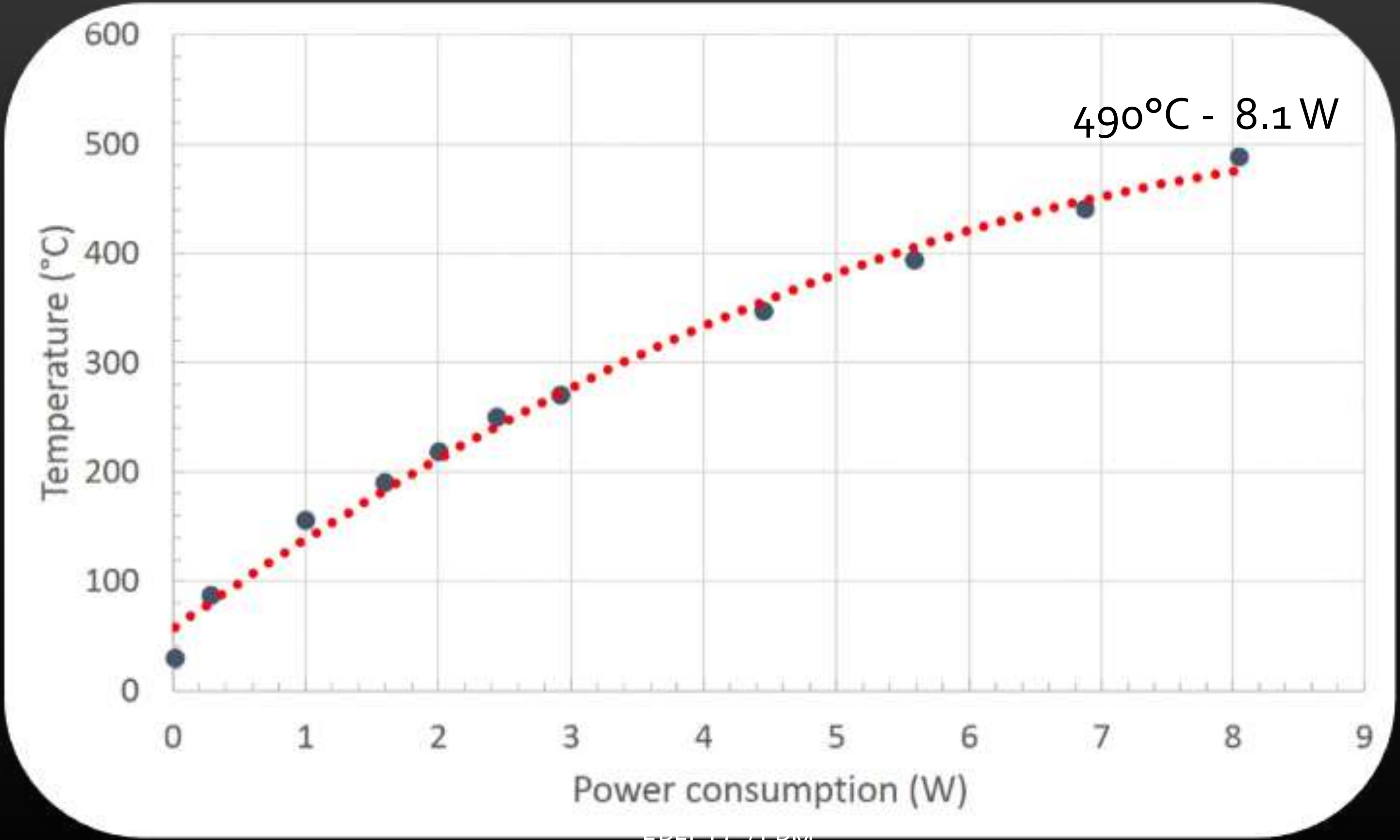
WDS© Ultra, thermal conductivity at 500° C: 0.027 W/m/K



Max. applied voltage: 24 V

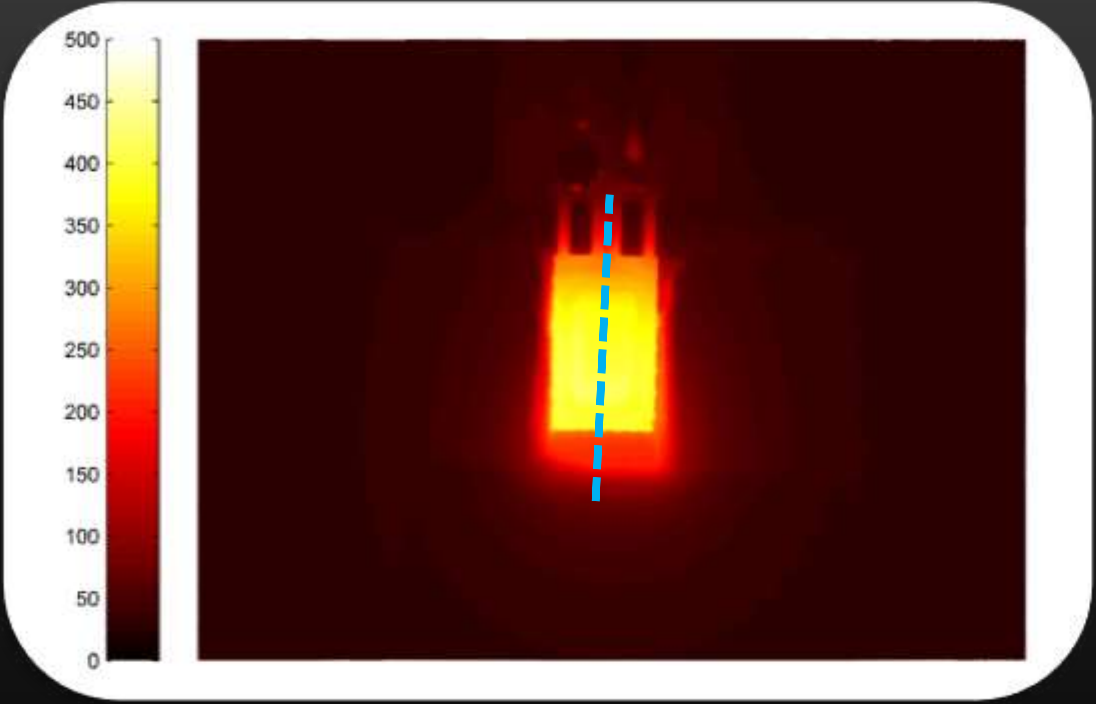
Thermal characterization

Power consumption vs. heating performance

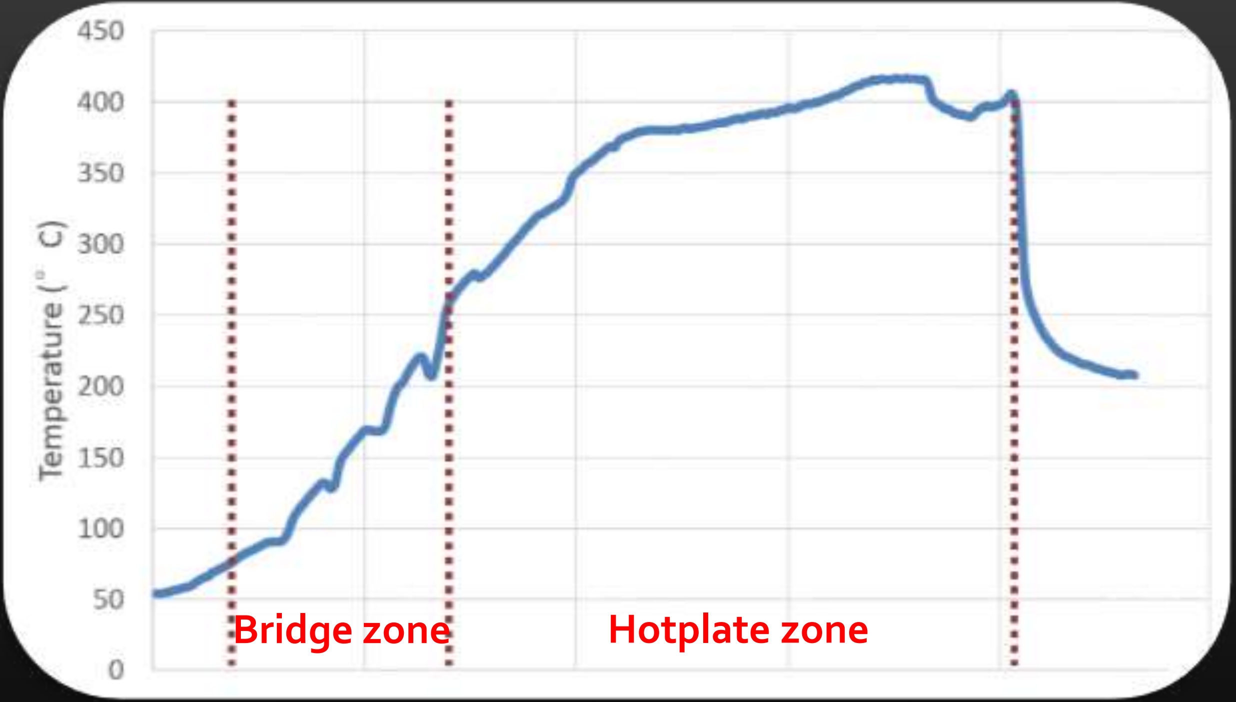


Thermal characterization

An IR Image of LTCC fuel processor running at 400°C of temperature set point



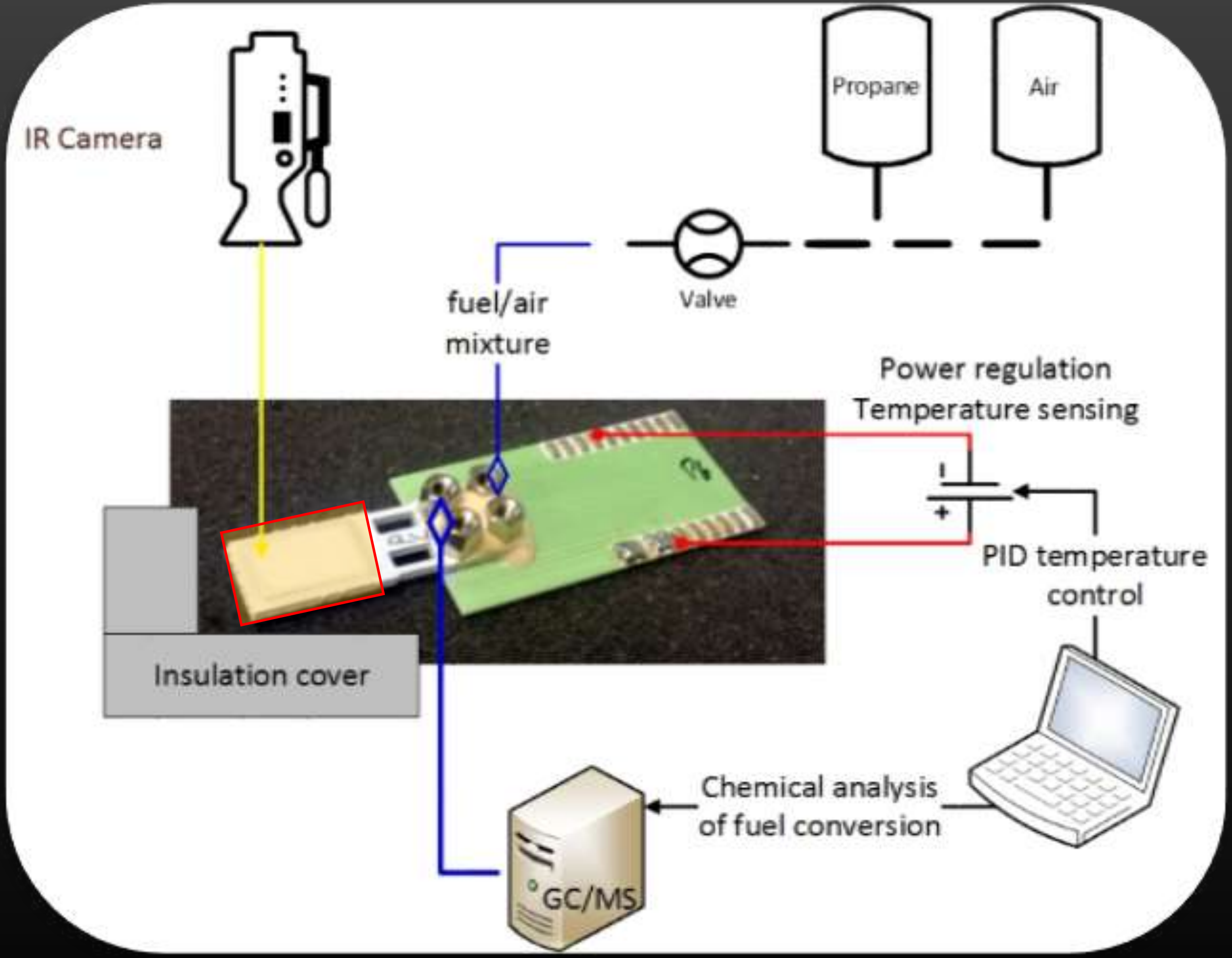
Temperature distribution profile



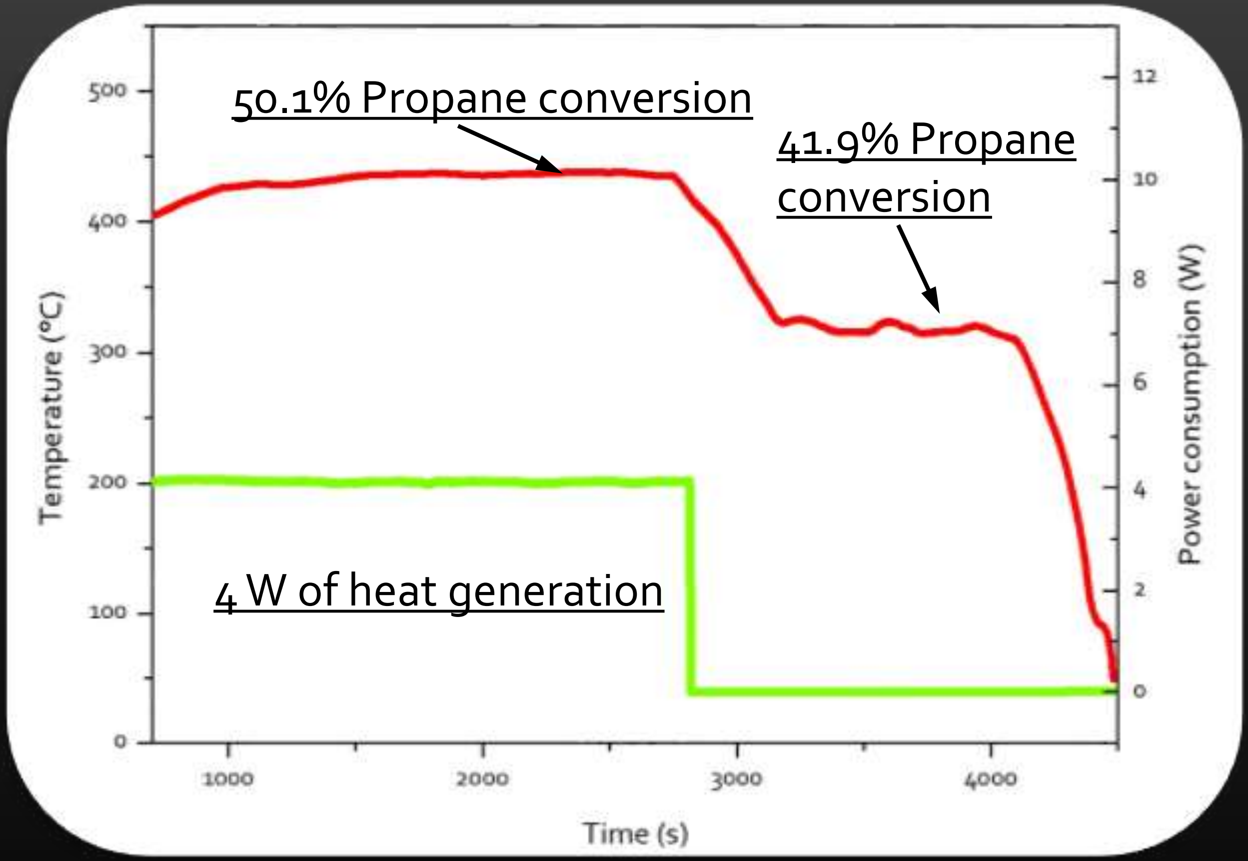
Bridge structure for efficient thermal isolation = convectional fluidic and electrical connection available

Self-sustain reaction characterization

Thermally self-sustain reaction testing setup



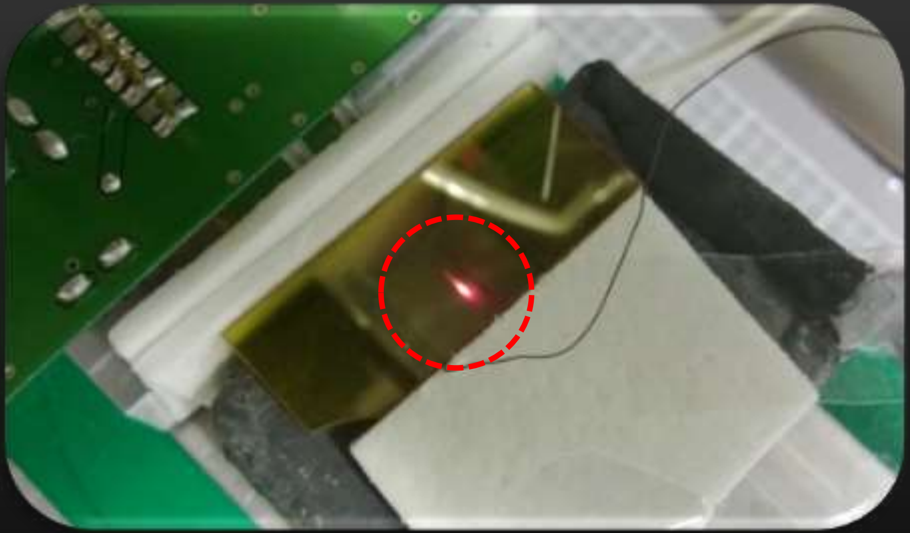
Self-sustain reaction characterization



Self-sustaining reaction

a. C/O ratio = 1
b. Flow rate = 200 sccm of air/propane mixture
c. Starting temperature = 400°C
d. Propane was able to partially oxidized in the LTCC fuel processor
e. The exothermic reaction makes the thermally independent reactor operation possible

Self-sustain reaction characterization



Problem:

a. unstable sealing caused the leakage after the exothermic reaction
b. the leakage may result in propane's low conversion

Conclusion

- A LTCC fuel processor was developed for the µ-SOFCs system
- The micro-scale fine structuration was realized in LTCC fuel processor through a novel progressive lamination
- The catalyst packed bed with YSZ beads could provide moderate pressure drop across the LTCC fuel processor
- The novel bridge structure efficiently isolates the heating zone of the processor
- 70% of heating area at hot zone could maintain relatively homogenous thermal distribution
- The LTCC fuel processor achieved self-sustaining operation through the exothermic partial oxidation of propane at a feeding flow rate of 200 sccm and C/O ratio = 1

Outlook

- Achieve reliable sealing for the LTCC fuel processor packaging
- Improve design of packed bed for avoiding hot spot
- Total oxidization process implementation in the LTCC fuel processor
- Thermal insulation enhancement

Thank you!