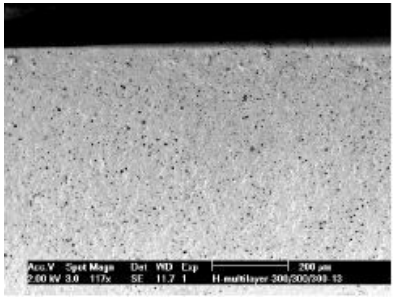
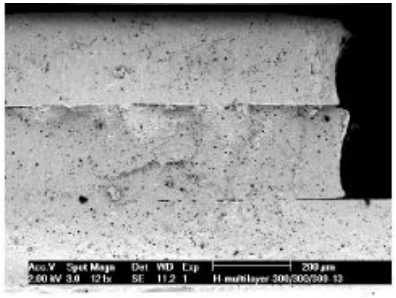
ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

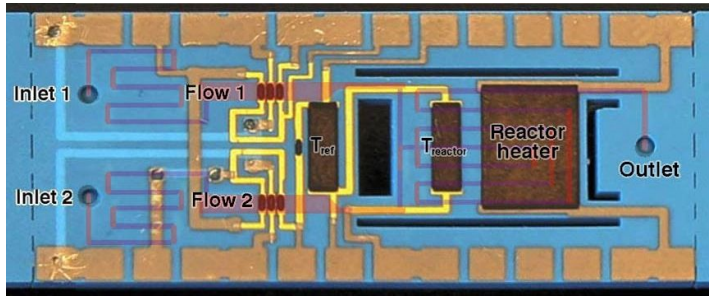
Lamination of LTCC at low pressure and moderate temperature using screen-printed adhesives

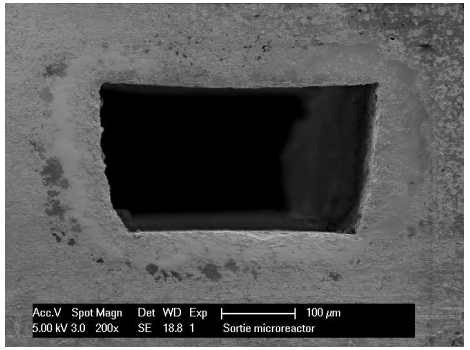
Thomas Maeder, Bo Jiang, Fabrizio Vecchio, Caroline Jacq, Peter Ryser and Paul Muralt

École Polytechnique Fédérale de Lausanne (EPFL), Switzerland









Outline

ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

1. Introduction - lamination of LTCC
2. Paste formulation - hot-melt adhesives
3. Application to LTCC test structure
4. Conclusions & outlook

Outline

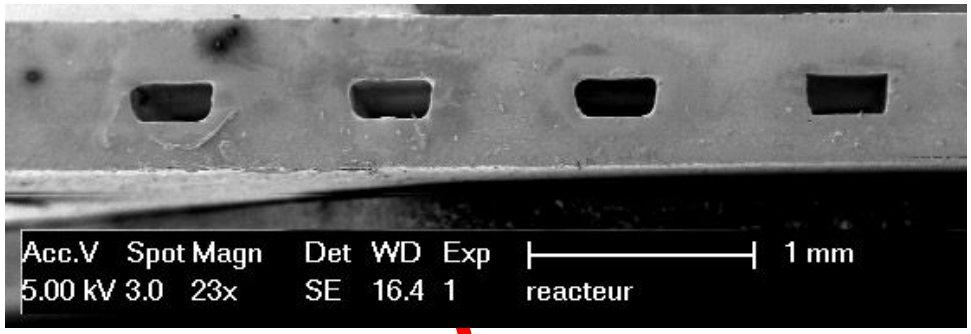
ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

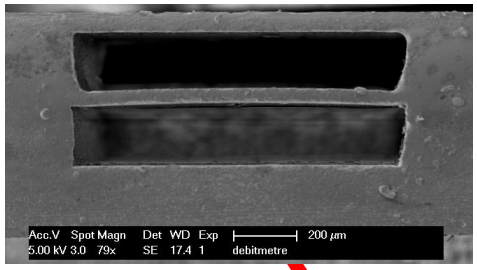
1. **Introduction - lamination of LTCC**
2. Paste formulation - hot-melt adhesives
3. Application to LTCC test structure
4. Conclusions & outlook

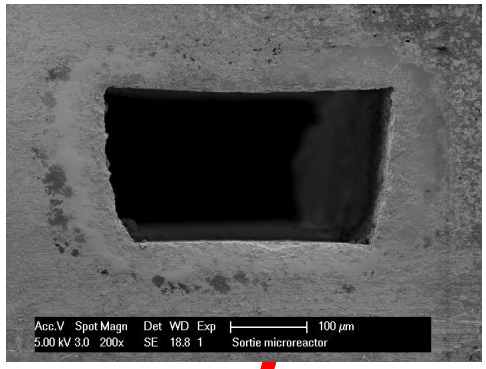
Ceramic structuration: microreactor

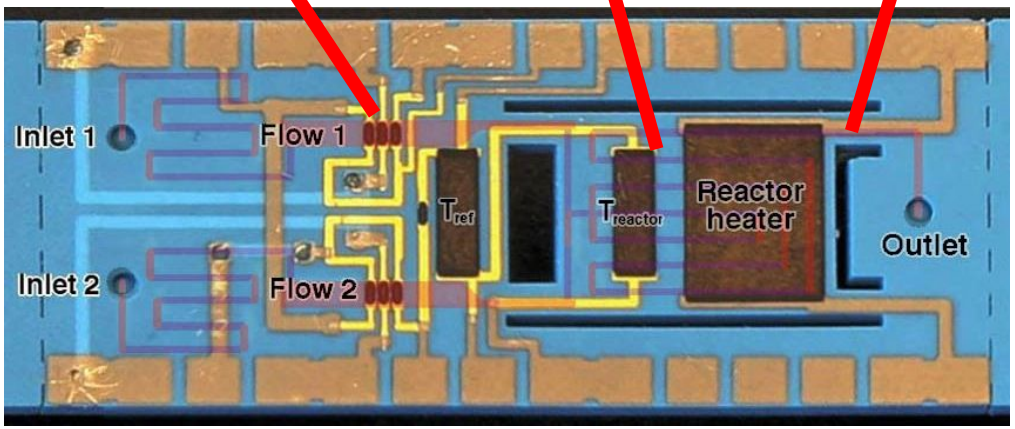
ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

- Complex fluidic circuit (2 layers + membranes)
- Flow sensors
- Heaters
- Channels in support posts
- …

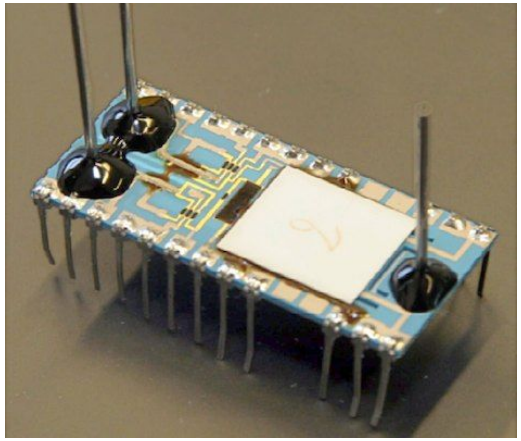








LTCC module + fluidic circuit

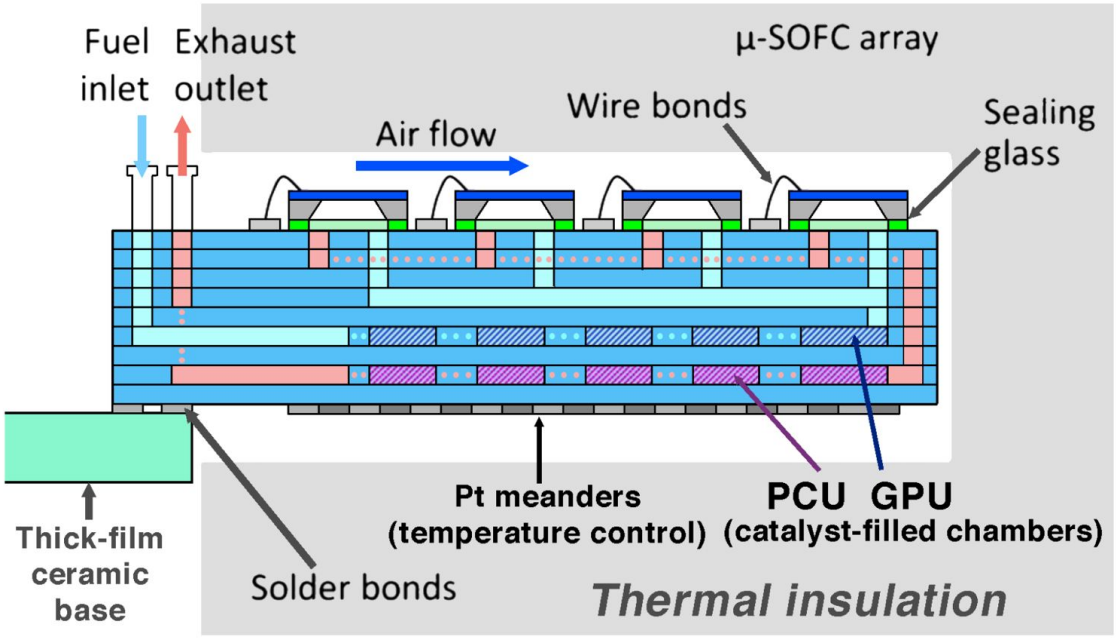


Complete device

Ceramic structuration: *μ*-SOFC module

ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

- Very complex:
  - Reformer (GPU)
  - Post-combustor (PCU)
  - Heat exchangers?
  - + all transfer piping
- Active filler material (catalyts)
- Many layers



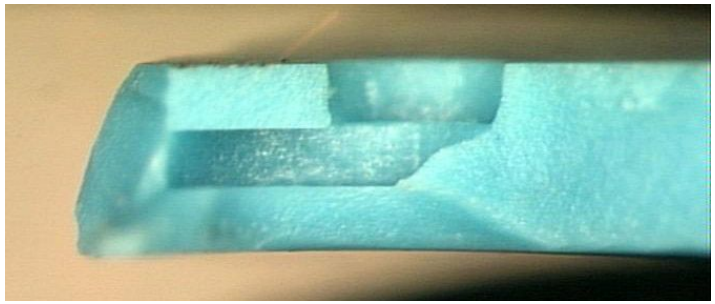
LTCC *μ*-SOFC module

PCU = post-combustor
GPU = reformer

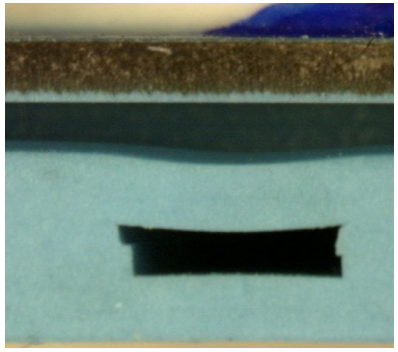
Ceramic structuration: gas channels

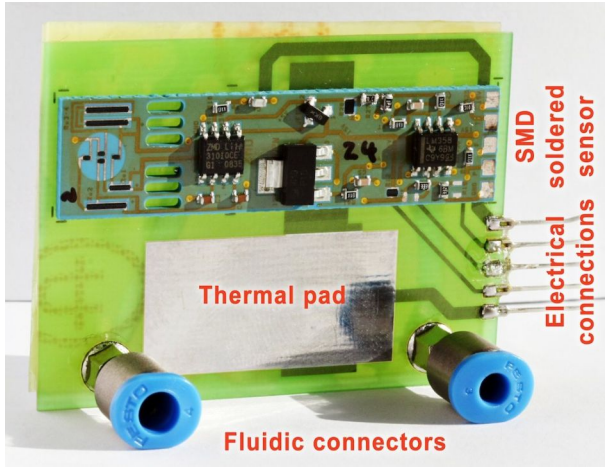
ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

- Complex circuits
- Multisensors
  - Pressure
  - Flow
  - Temperature

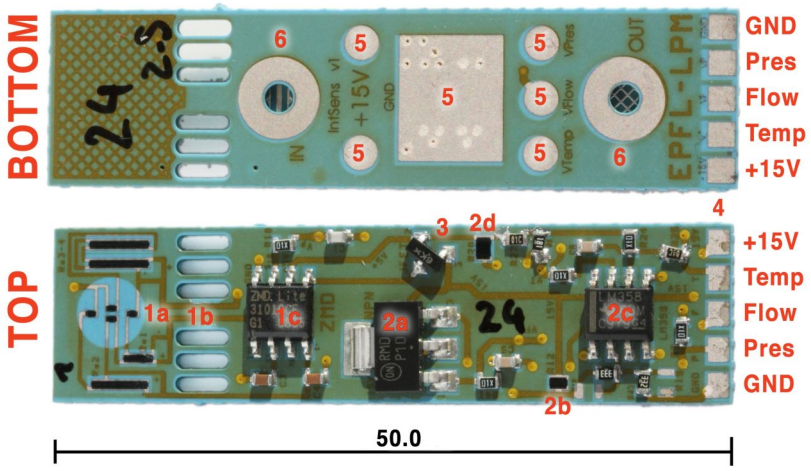


Process gas channels & integrated modules





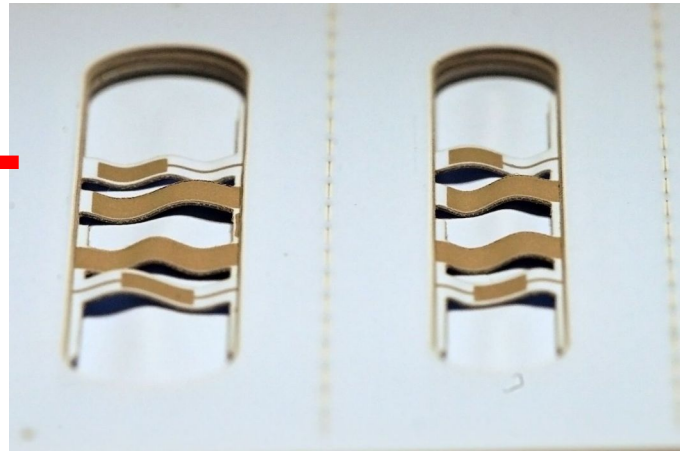
Gas multisensor (p, Q, T)

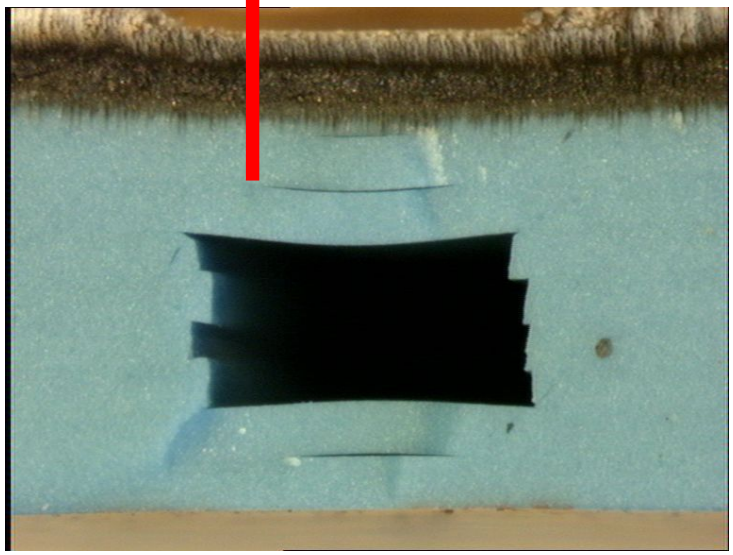


Ceramic structuration: lamination issues

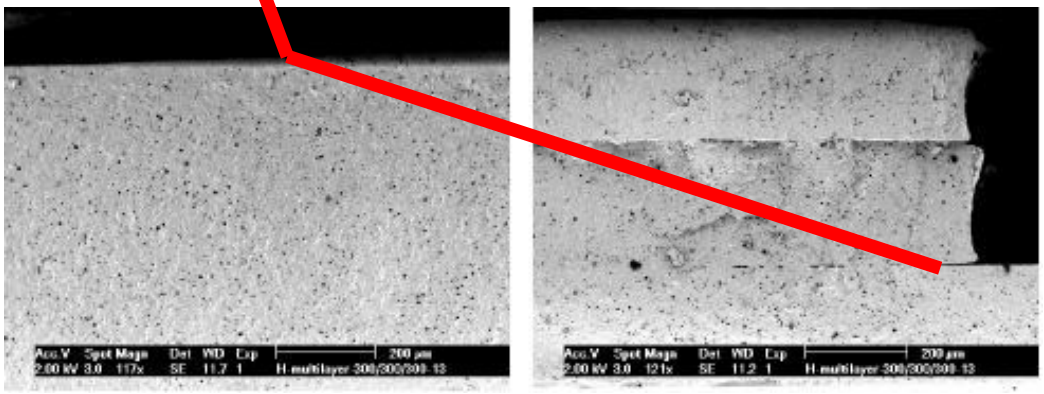
ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

- Standard: ≈70°C 20 MPa
- Deformation of intricate structures
- Lower pressure at edges
- Bonding above & below cavities





Multilayer above & below cavity



Bonding besides a cut-out zone

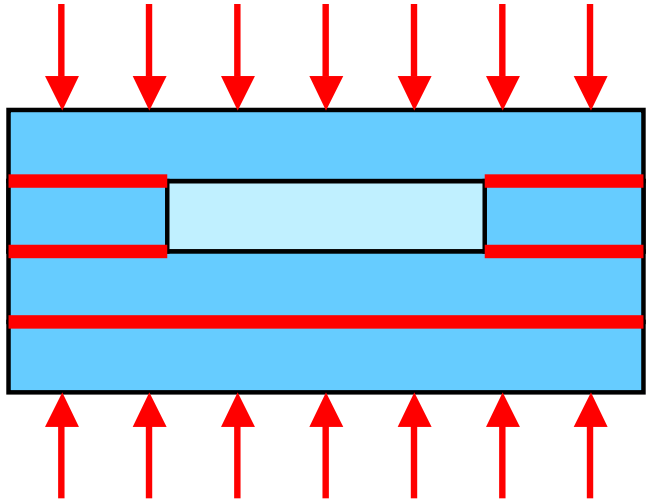
Basic problem: homogeneous LTCC behaviour

Mechanisms of lamination ≈ tape deformation

Lamination: proposed solutions

- Sacrificial fillers
  - Prevent crushing / deformation
  - Allow standard high-pressure/-temperature lamination
  - Somewhat cumbersome (fill complex features)
  - Ensure pressure transfer - good lamination

- Low-pressure lamination
  - Careful determination of minimal parameters
  - "Glues": solvents / thinners / honey / printing vehicles
  - Adhesive tapes



Outline

ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

1. Introduction - lamination of LTCC
2. **Paste formulation - hot-melt adhesives**
3. Application to LTCC test structure
4. Conclusions & outlook

Hot-melt formulation: requirements 12

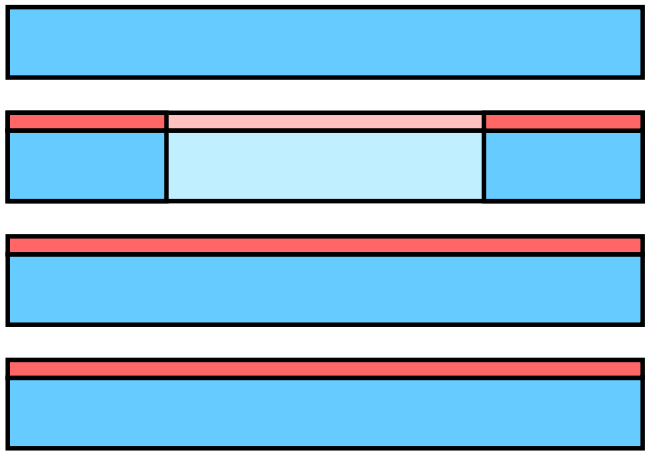
ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

1) Processing

- Screen-printable (infrastructure)
- High-solids (low solvent)
- Nonagressive towards LTCC tape
- Print over whole raw tape (generic)

2) After printing

- Low/no tack
  - No dust from laser, punching, …
  - Normal handling & storage possible
- Overprintable (?)
- Corrections possible during stacking/alignment

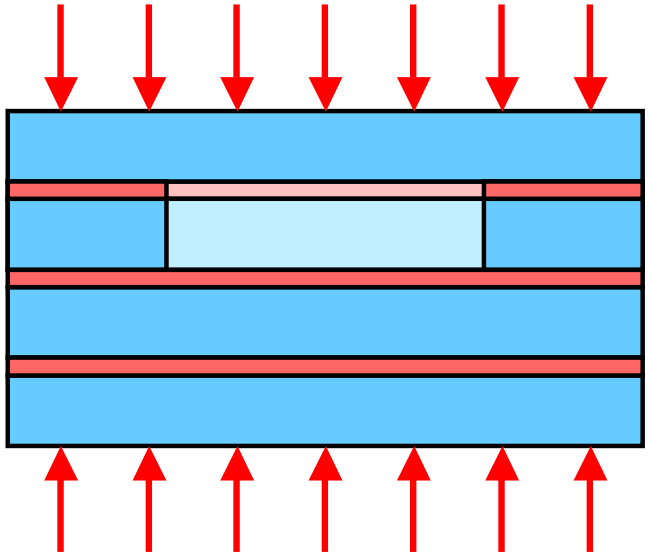


Hot-melt formulation: requirements 34

ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

3) Lamination

- Allow lamination at **moderate** temperature & **low** pressure
- Melt at moderate temperature
- Large decrease of viscosity / increase of tackiness

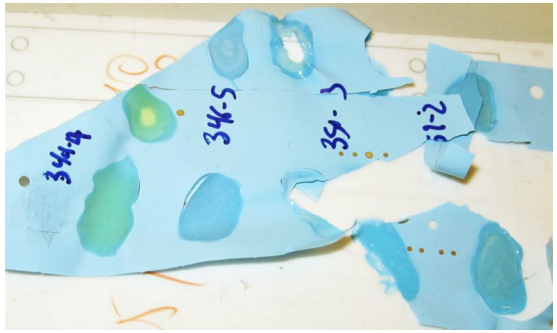


4) Firing

- Hold / "fuse" tapes together by capillary action
- Debind cleanly
  - Not too much organics
  - Binder burnout assorted with LTCC

Formulation

- **Tape binders (typ.):**
  - Acrylics: PMMA, PEMA, PBMA, …
  - Polyvinylbutyral (PVB)
  - > Sensitive to polar organic solvents



- **Screen emulsions (typ.):**
  - Polyvinylalcohol (PVA, PVOH)
  - > Sensitive to water only

- **Avoid tape dissolution:**
  - Low solvent amount: high-solids
  - Solubility parameter mismatch
  - Balance volatility: screenability vs. persistence - attack
    - In this respect, methods compatible with volatile solvent better (spray, inkjet, dipping, …)

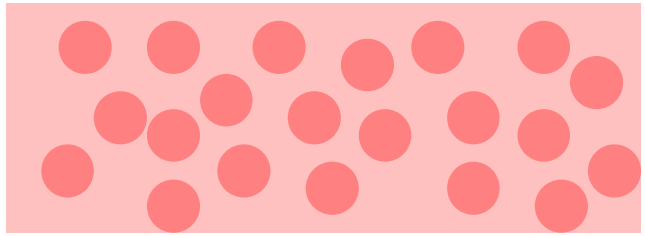
Formulation: tested systems

ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

2 alternatives:

- **Solvent-binder-plasticiser**
  - Formulate to dry "just non-tacky"
  - Upon reheating, plasticiser serves as solvent
  - Somewhat progressive solid-liquid transformation (homogeneous glass)

- **Solvent-binder-[plasticiser]-wax**
  - Wax crystallises out @RT: non-tacky
  - Upon reheating, wax acts as a solvent
  - More abrupt solid-liquid transition possible



Formulation: binders

ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

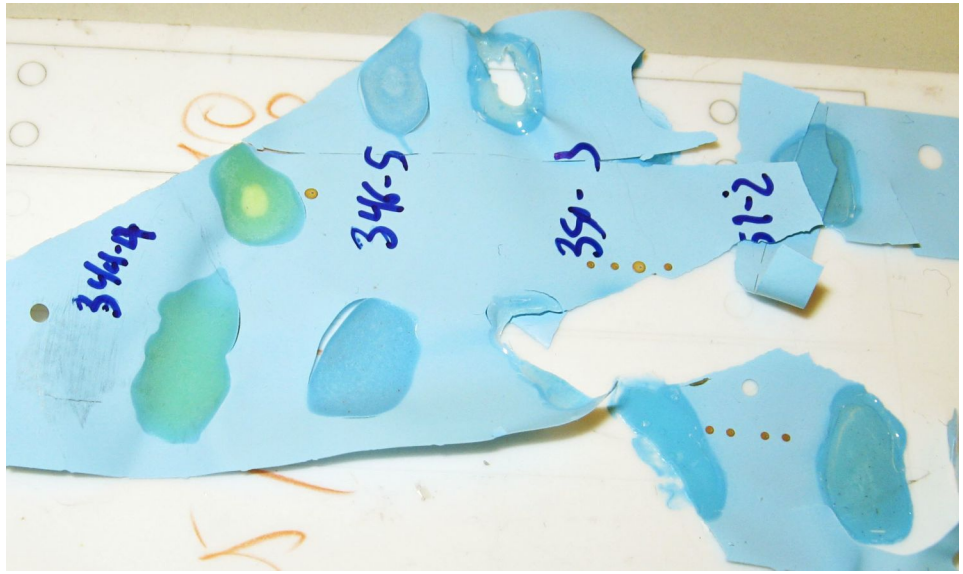
- "Common" binders selected
- PVB, PxMA or EC: used in tapes & pastes
- Hansen solubility parameters (give an idea…)
- Other binders possible (HPC, PVP, …)

Binder	δ_d [MPa$^{0.5}$]	δ_p [MPa$^{0.5}$]	δ_h [MPa$^{0.5}$]	R [MPa$^{0.5}$]	Note
PVB (20% OH)	18.6	4.4	13.0	10.6	Solubility in polar solvant with H bonding (alcohols, acohol-ethers, alcohol-esters)
PMMA	18.6	10.5	7.5	8.6	*Solubility in esters, ketones, alcohol-ethers, acohol-esters*
EC	17.1	7.3	9.7	9.0	Intermediate btw. PMMA & PVB In practice very easy to solubilise in a wide range of modestly polar solvents

Formulation: solvents

ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

- **Classical - terpineol, texanol**
  - Excessive dissolution of tapes



- **Glycols - PG, BG, …**
  - Low attack
  - Not suitable for PVB / EC
  - Suitable for other binders
- **Fatty alcohols**
  - (Cyclo)hexanol: good compromise
  - + tailing solvent: lower volatility
  - Suitable for PVB / EC (esp. with co-solvents / plasticiser)

Formulation: plasticisers & waxes

ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

- **Mixing possible to tune properties**
  - Plasticiser-wax: plasticiser stays in resin, wax precipitates out
  - Wax-wax: increase total solubility in ink & tune consistency

Substance†	T_m [°C]	**Note**
Triacetin	L	Somewhat too volatile; T_b = 258°C
TBAC TEG-EH	L	Good plasticising action; T_b > 320°C
C18E2	38	Dissolves EC at ≈50°C; soft precipitates @RT
So-16	48	Dissolves EC at ≤60°C; soft precipitates @RT
Cetanol	49	Dissolves EC & PVB at ≤60°C; hard precipitates @RT

† **TBAC = tributylacetylcitrate - tested with EC only**
TEG-EH = triethylene glycol bis(2-ethylhexanoate) - tested with both EC & PVB
So-16 = sorbitan monopalmitate
C18E2 = steareth-2 (diethylene glycol stearyl ether)

Formulation: examples

ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

- **Optimise cold-warm contrast**
  - Use low-MW resin & high amount of additives ("fragile glass")
  - Use wax with strong cold-warm solubility dependence

Ink	**Homogeneous** #362-4	**Wax-precipitate** #363-2	**Wax-precipitate** #364-2
Solvent	Cyclohexanol	Cyclohexanol	Hexanol
Binder	EC	EC	EC
Plasticiser	TBAC	TBAC	-
Wax	-	Cetanol	C18E2 & cetanol
Ink (wet)	Clear	Clear	Clear
Ink (dry)	Clear	Milky	Milky
Solids	50%	55%	60%

Outline

ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

1. Introduction - lamination of LTCC
2. Paste formulation - hot-melt adhesives
3. **Application to LTCC test structure**
4. Conclusions & outlook

Lamination quality & distortion

ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

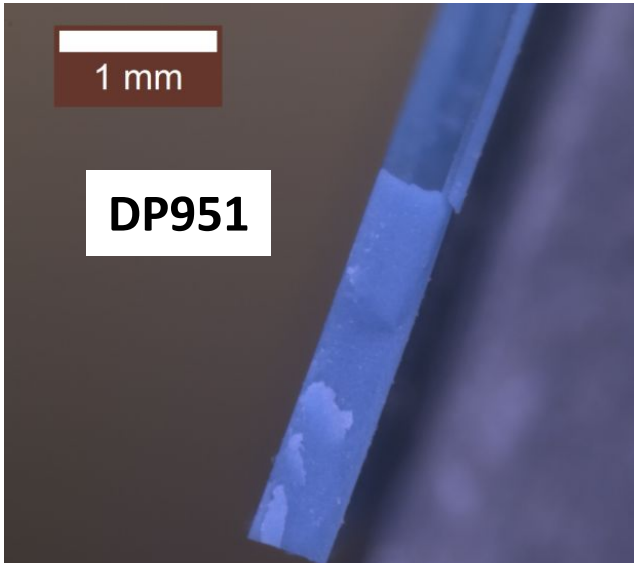
- Simple ø5 mm membranes
- Tapes: DP 951 or Her HL2000
- Good bonding & low distorsion @60°C

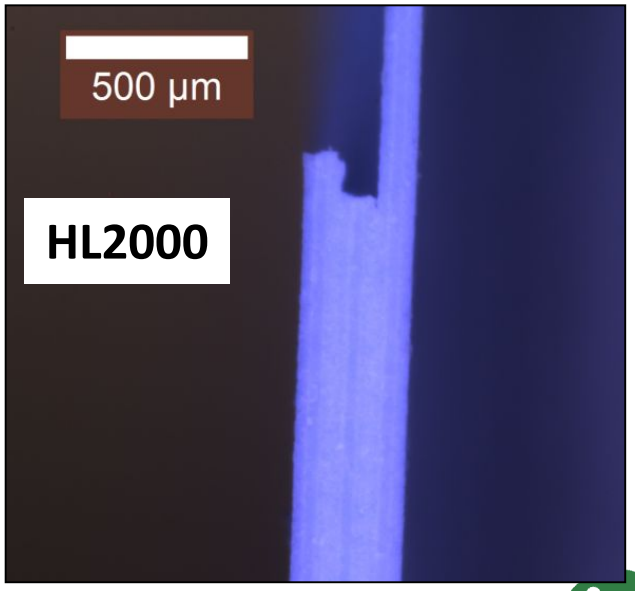
DP951

Membrane (µm)		1.94 MPa		3.88 MPa		7.75 MPa	
Glue	LTCC	fired quality		fired quality		fired quality	
#344-5	1st layer	7	dense	7	dense	4	dense
#346-6	1st layer	12	dense	6	dense	10	dense
#362-4	1st layer	6	dense	11	dense	7	dense
#363-2	1st layer	11	dense	6	dense	3	dense

HL2000

Profilometer results		**1.94 MPa**		**3.88 MPa**		**7.75 MPa**	
Glue	LTCC	fired quality		fired quality		fired quality	
#346-6	1st layer	8	dense	10	dense	15	dense
#362-4	1st layer	9	dense	5	little crack	13	dense
#363-2	1st layer	10	little crack	13	dense	12	dense

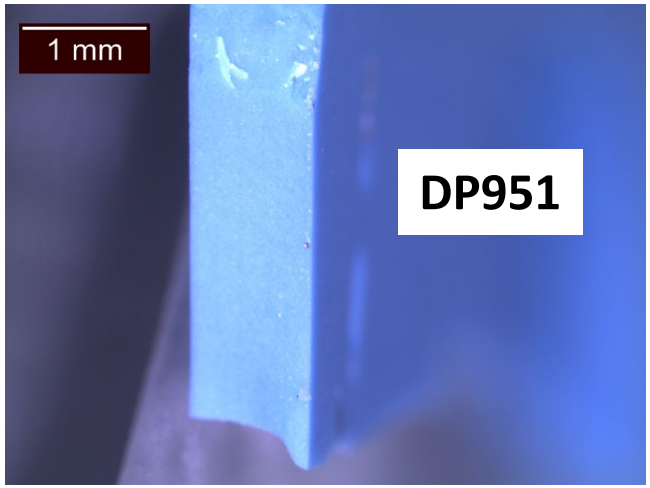


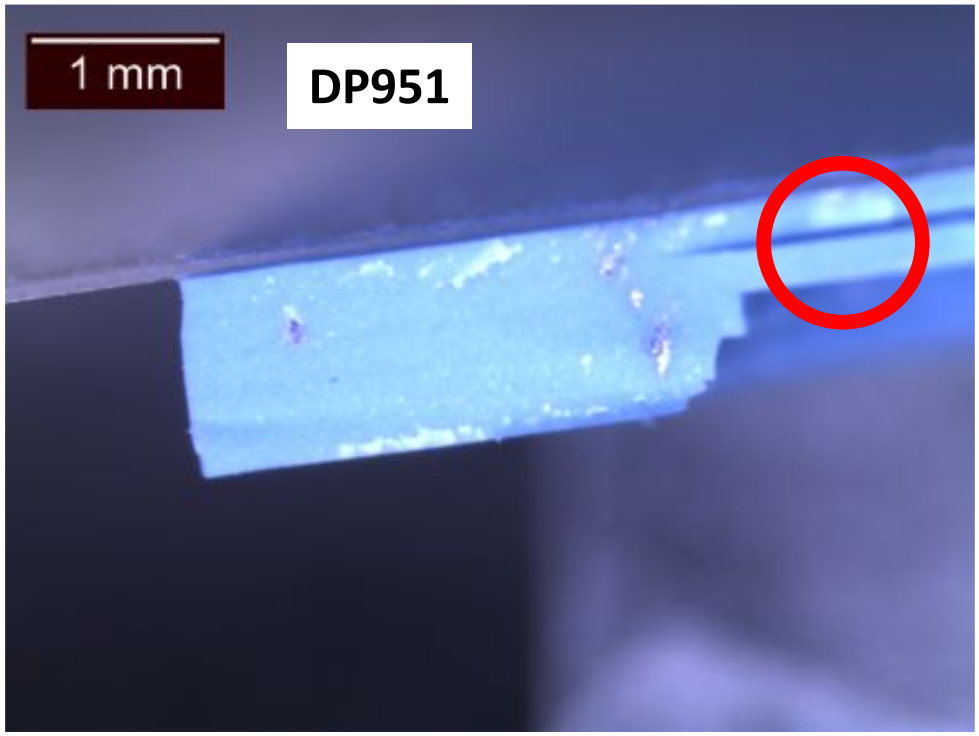


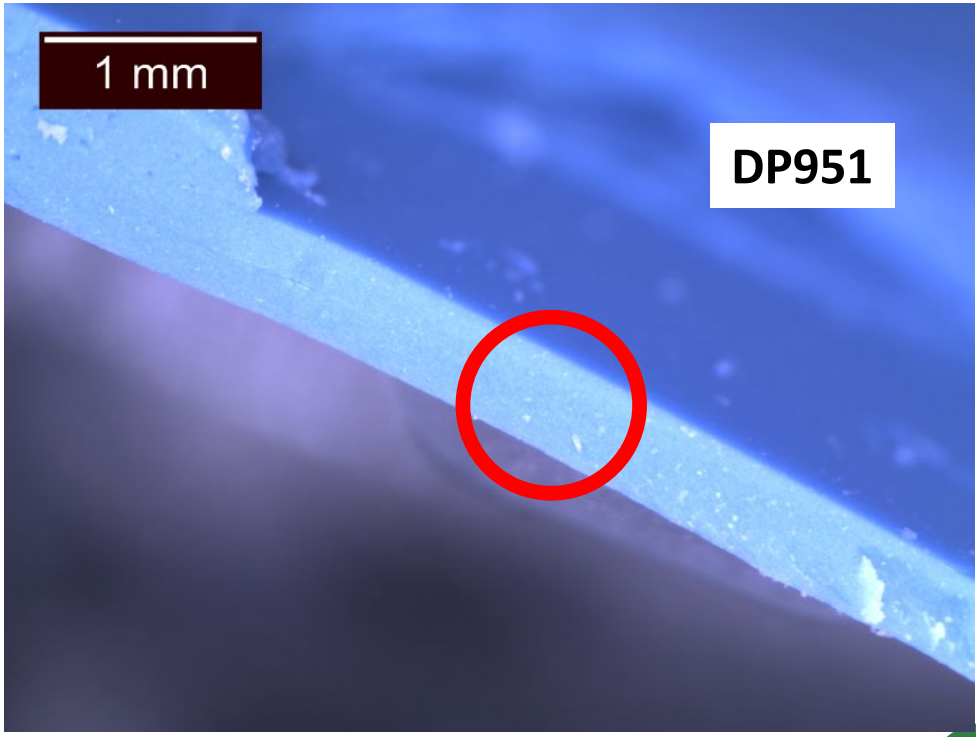
Lamination quality & distortion

ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

- Body mostly OK
- Double membrane layer difficult (no pressure)
  - More glue: lamination OK, but deforms more upon firing







Outline

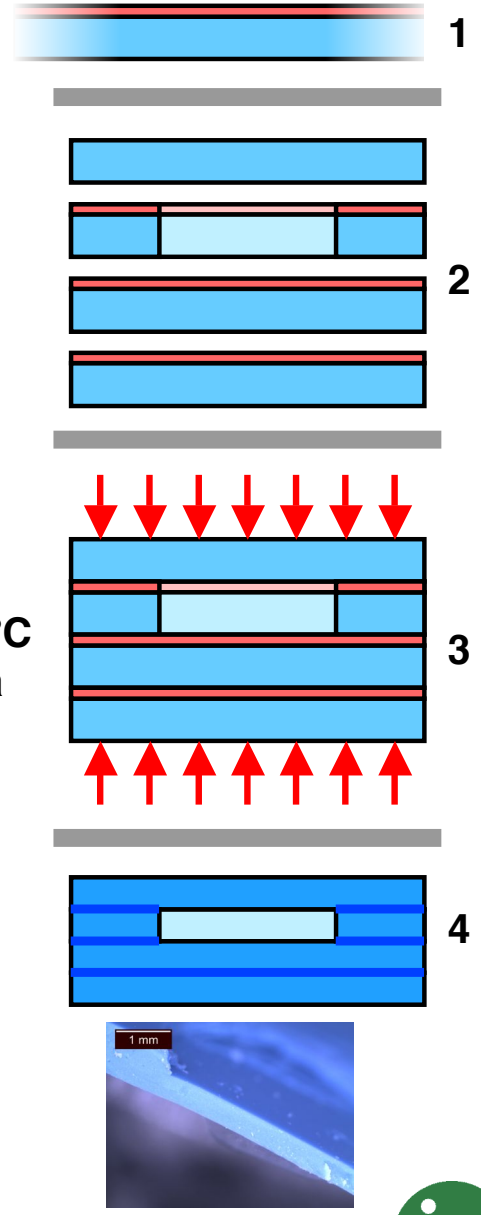
ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

1. Introduction - lamination of LTCC
2. Paste formulation - hot-melt adhesives
3. Application to LTCC test structure
4. **Conclusions & outlook**

Conclusions & outlook

- **Simple low-pressure lamination technique**
  - Generic prior application onto tape
  - Use of existing infrastructure (screen-printing), or other methods
  - Easy, predictable processing & handling
  - Low-pressure lamination at moderate temperature
  - Formulations using safe chemicals, low toxicity

- **Outlook**
  - Accurate study of what is going on…
    - Rheology
    - Phase transitions (waxes)
  - Further optimise formulations
  - Study "difficult" cases (layers atop cavities, …)



THANK YOU! Questions?

ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

Acknowledgements

- Swiss National Science Foundation
  - 'Sinergia' grant CRSI22-126830/1
- ONEBAT project partners
  - www.nonmet.mat.ethz.ch/research/onebat

FNSNF
FONDS NATIONAL SUISSE
SCHWEIZERISCHER NATIONALFONDS
FONDO NAZIONALE SVIZZERO
SWISS NATIONAL SCIENCE FOUNDATION

ONEBAT

zh aw Zürcher Hochschule für Angewandte Wissenschaften

csem

LC EPFL

LTNT

NTB INTERSTAATLICHE HOCHSCHULE FÜR TECHNIK BUCHS

ETH Eidgenössische Technische Hochschule Zürich
Swiss Federal Institute of Technology Zurich

Ceramics ETH

LPM LABORATOIRE DE PRODUCTION MICROTECHNIQUE, CH-1015 LAUSANNE