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Global and Localized Temperature Stability in Low Temperature Cofired Ceramic (LTCC)

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Outline

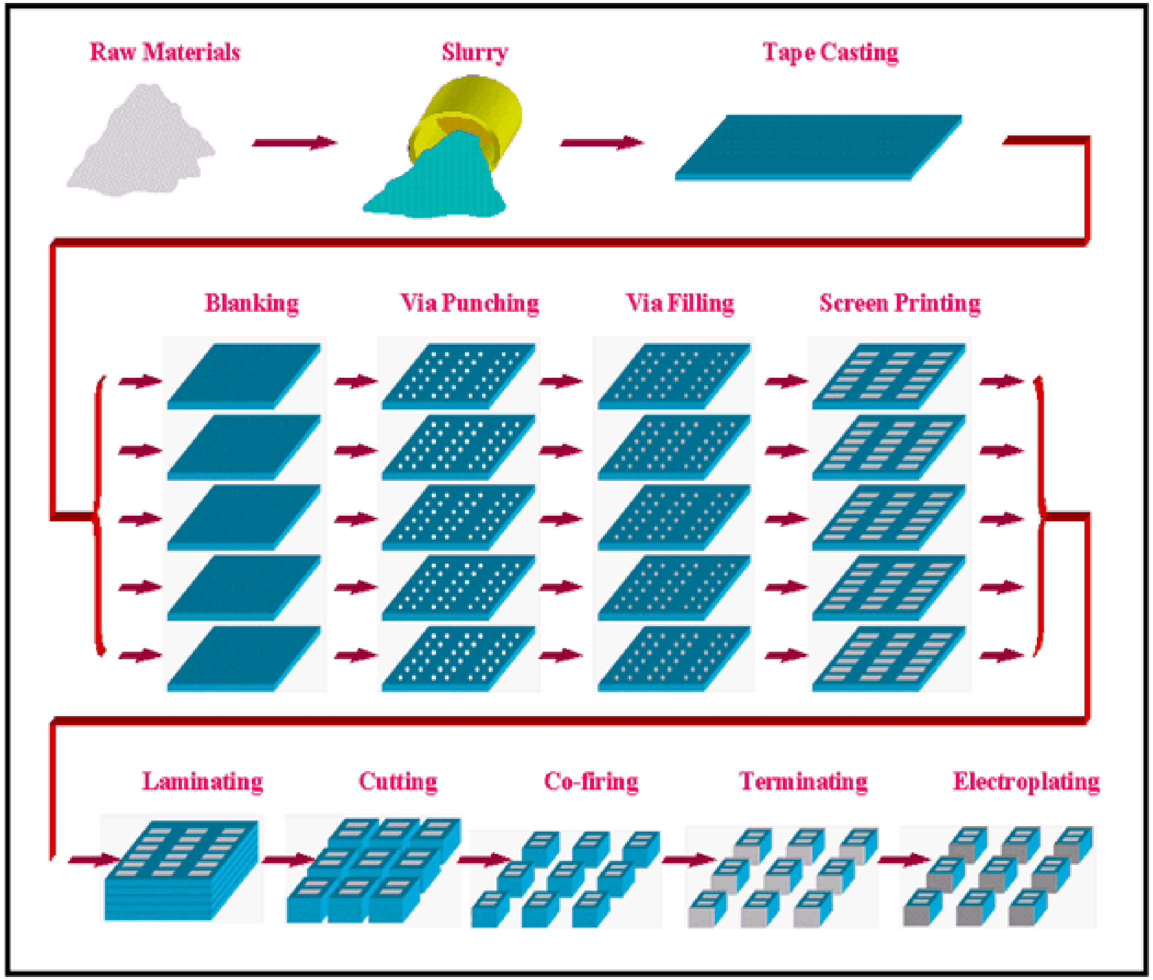
- Introduction
  - LTCC technology
  - Temperature coefficient of resonant frequency, τ_f

$$\tau_f = \frac{1}{f_r}\frac{\Delta f}{\Delta T}$$

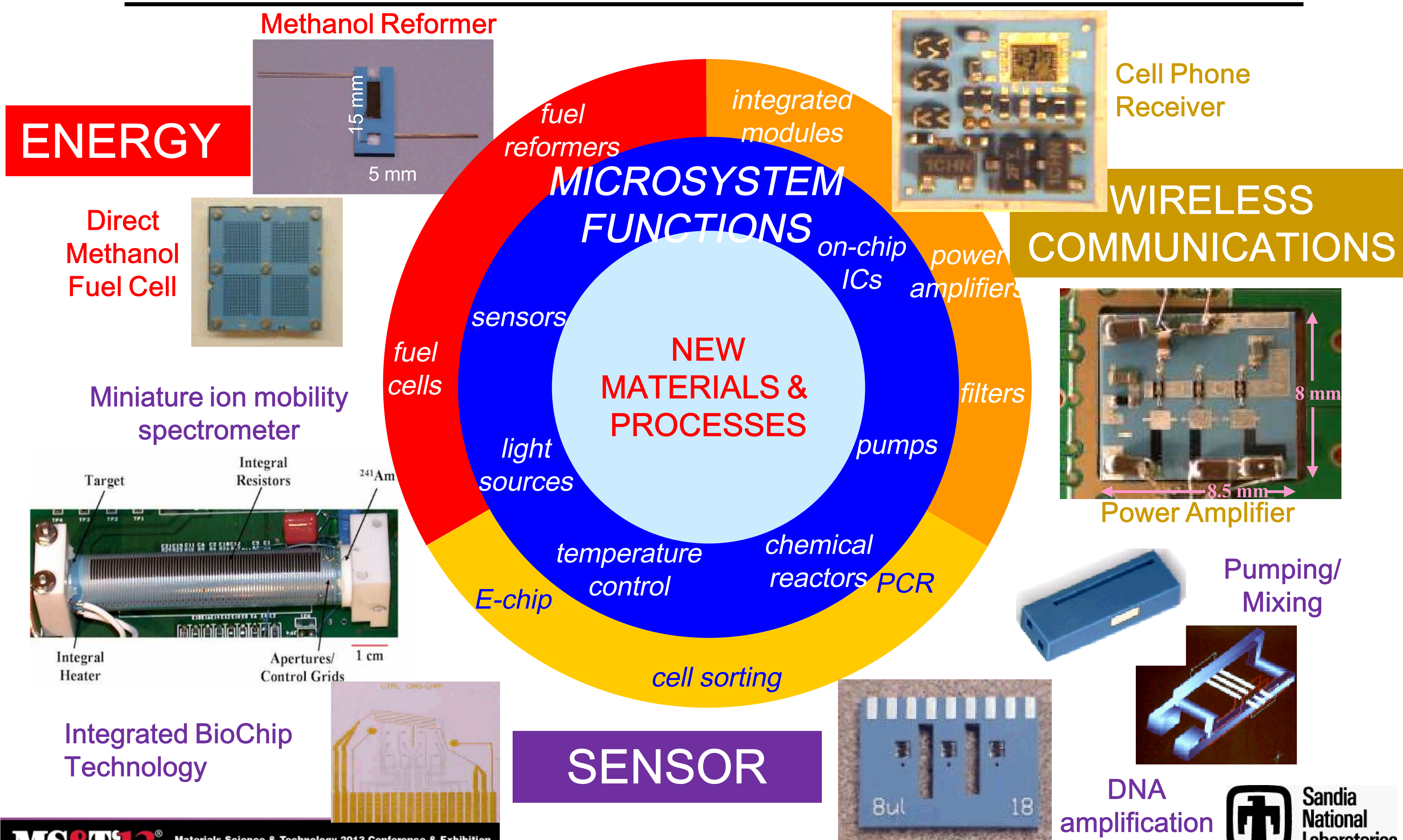
- Global temperature stability
  - $0\tau_f$ LTCC host dielectric and compensation mechanisms
  - Densification and crystallization in crystallizable glass-ceramics (T2000 and DuPont new 9k7)
- Localized temperature stability (SNL EC LDRD, Will Hsieh, 5353)
  - Development of τ_f compensating dielectrics
  - Simulation and experiment
  - Energy filling factor and effective τ_f compensation
  - Demo of localized $0\tau_f$: temperature stable S-band filters
- Summary

What is LTCC?

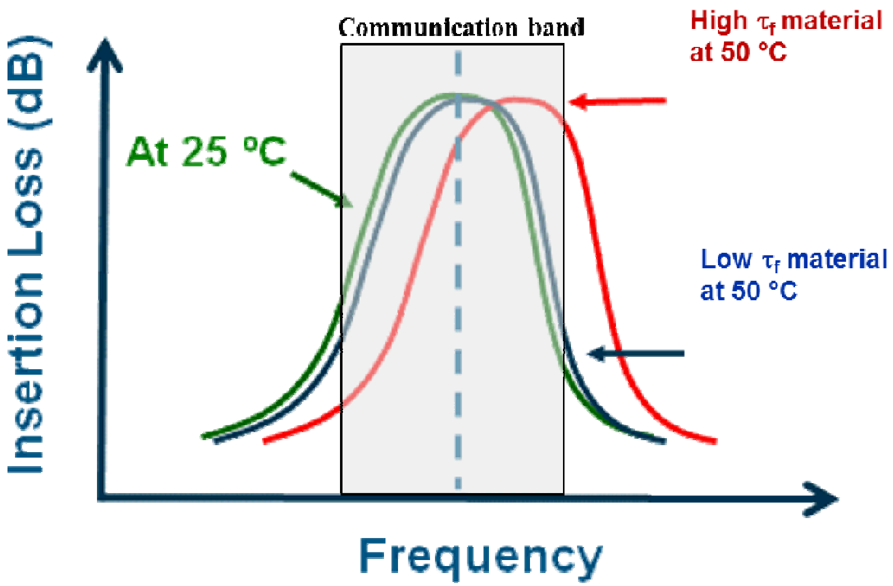
- All glass or glass-ceramic tape at sintering temp ≤ 950 °C, typical 850 °C. Ag/Au cofireable
- Multilayer → 3D circuitry, passive integration, micro feature formation
- Parallel process → high overall yield
- Established manufacture infrastructure



LTCC Microsystem



Why τ_f is Important?



Example:

GSM 900 MHz
Up-/down-link BW = 25 MHz
For a τ_f = -50 ppm/°C materials,

Δf ~ 6 MHz (-50 to 80 °C)

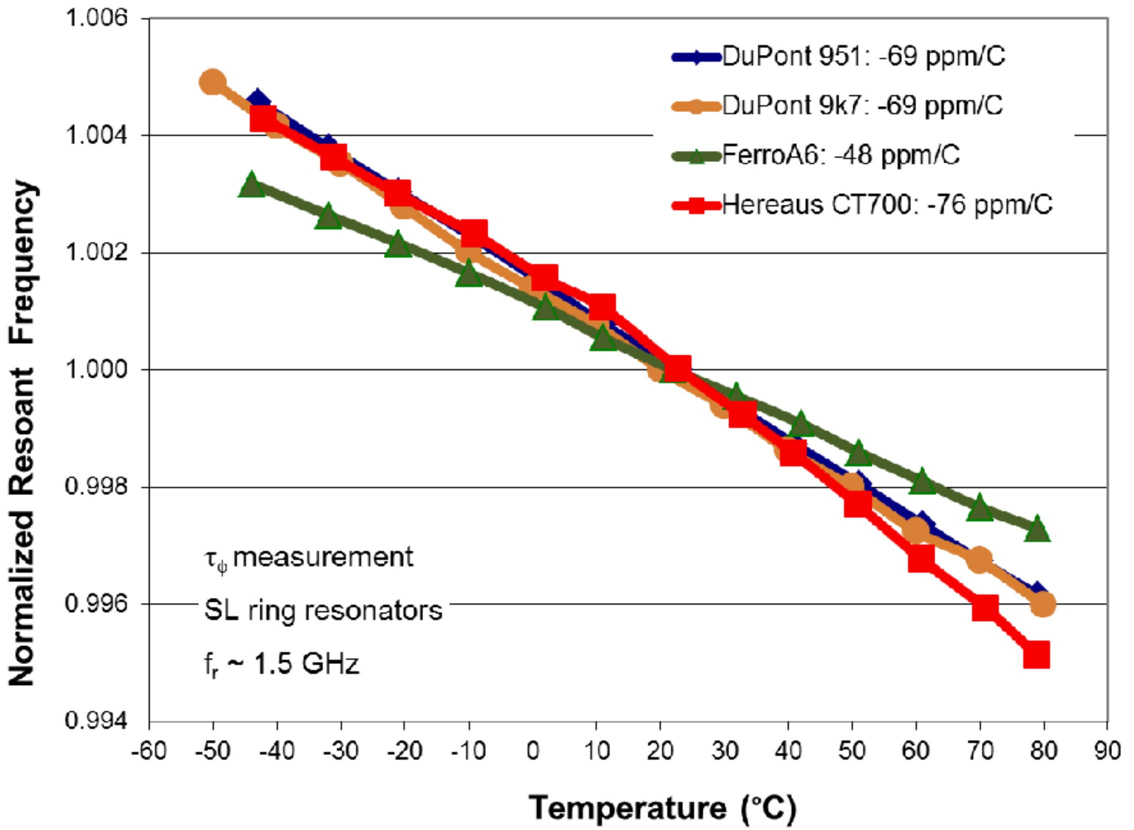
Shannon-Hartley

$$C = B \log_2(1+S/N)$$

C: channel capacity
B: bandwidth
S/N: signal/noise ratio

- Critical dielectric properties for RF/MW applications: ε, Q and τ_f
- Low τ_f is needed:
  - For filtering and frequency generation circuits
  - Efficient use of wireless bandwidth
  - To eliminate costly temperature compensation mechanisms

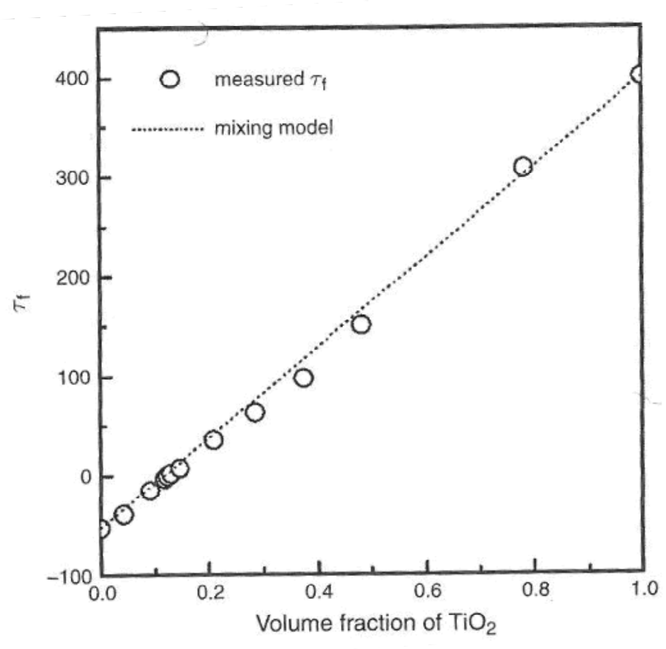
τ_f of Commercial LTCCs



- τ_f = -48 to -80 ppm/°C
- Designed primarily for packaging, not RF applications

τ_f Compensation

- Solid solution of dielectric ceramics with <u>opposite</u> τ_f
  - Microwave dielectrics
  - → Global $0\tau_f$ LTCC
- Stack of alternating layers of <u>opposite</u> τ_f materials
  - Sequential sintering of Al_2O_3/TiO_2
  - Adhesively bonded structure
  - → Localized $0\tau_f$ LTCC



$(1-x)CaWO_4 - x\,TiO_2$
S. H. Yoon et al 2007

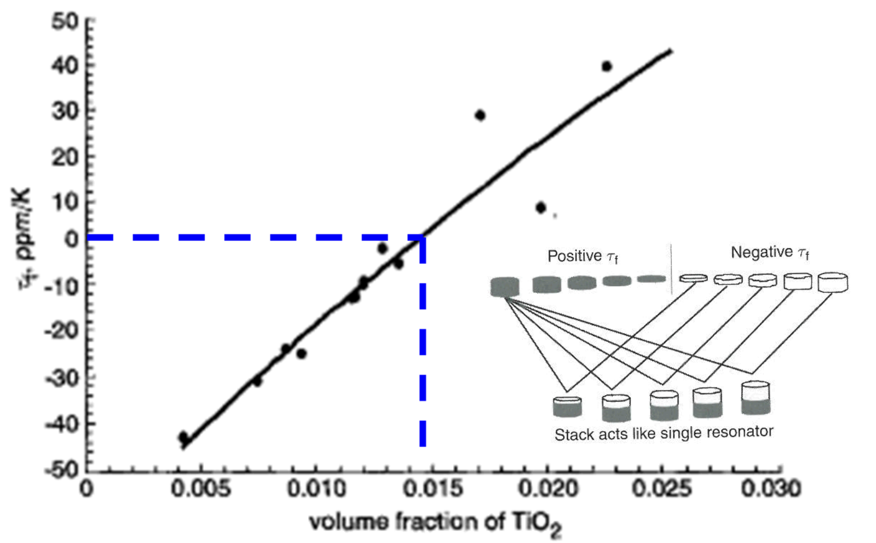


Fig. 4 τ_f *against volume fraction of* TiO_2
● measured
— predicted

Al_2O_3-TiO_2-Al_2O_3-TiO_2 …
N. Alford, 2000.

Candidates for τ_f Adjustment

$$\tau_f = -\tfrac{1}{2}\tau_\varepsilon - \alpha$$

device (τ_f) **material** (τ_ε, α)

α= coefficient of thermal expansion (CTE)

α: 3-10 ppm/°C → τ_ε dominates τ_f

τ_ε is opposite in sign to τ_f

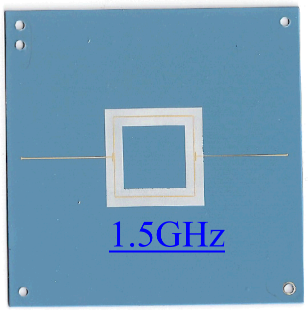
Global $0\tau_f$ → TiO_2

Localized $0\tau_f$ → $SrTiO_3$, V-glass

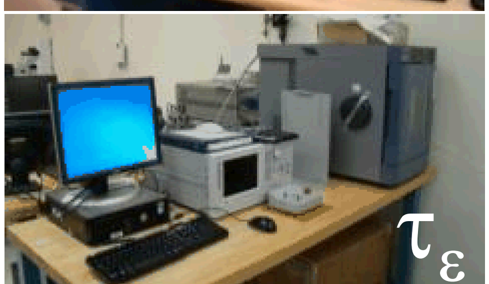
Materials	Density (g/cm^3)	ε	τ_ε (ppm/°C)	τ_f (ppm/°C)	Sintering Temp (°C)
TiO_2	4.23	85	-750	370	~ 1200
$CaTiO_3$	3.98	180	-1850	920	~ 1400
$SrTiO_3$	5.13	300	-3000	1500	~ 1550
Al_2O_3	4.00	9.6	105	-60	~ 1600
V-glass	2.77	7.3	N/A	N/A	Tg= 625°C

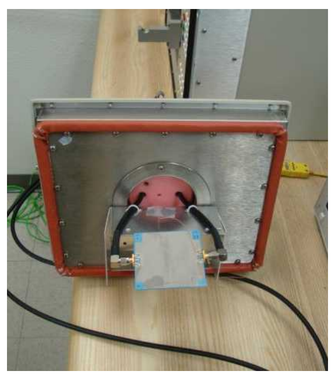
Characterization of τ_f and τ_ε

- Test vehicle; 4 layer LTCC panel



- Test setup





Test method

- Resonant frequency, f_r, from reflection coefficient, S11
- Temperature range -50 to 80°C

$$\tau_f = \frac{1}{f_r}\frac{\Delta f}{\Delta T}$$

$$\tau_f = -\tfrac{1}{2}\tau_\varepsilon - \alpha$$

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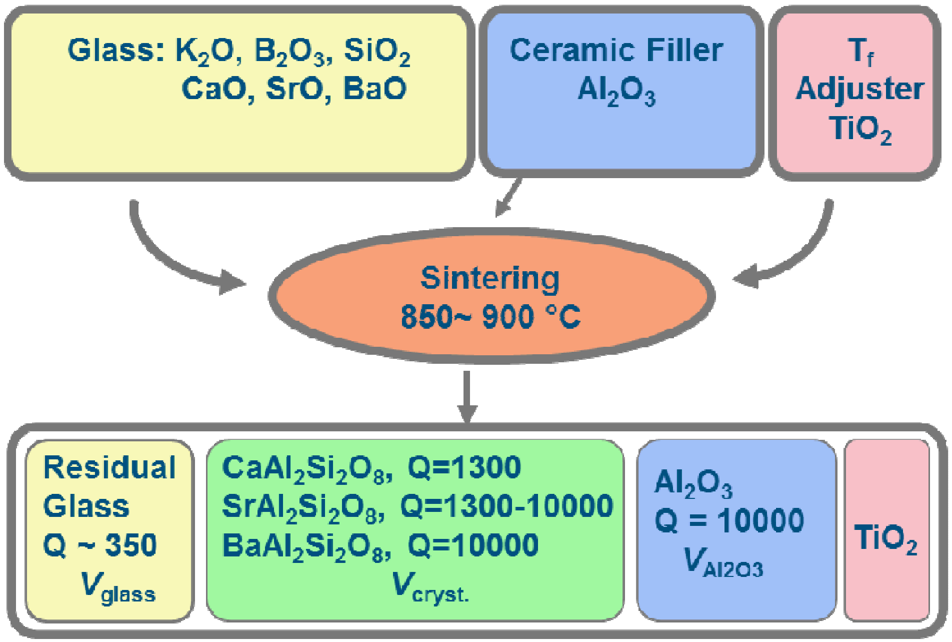
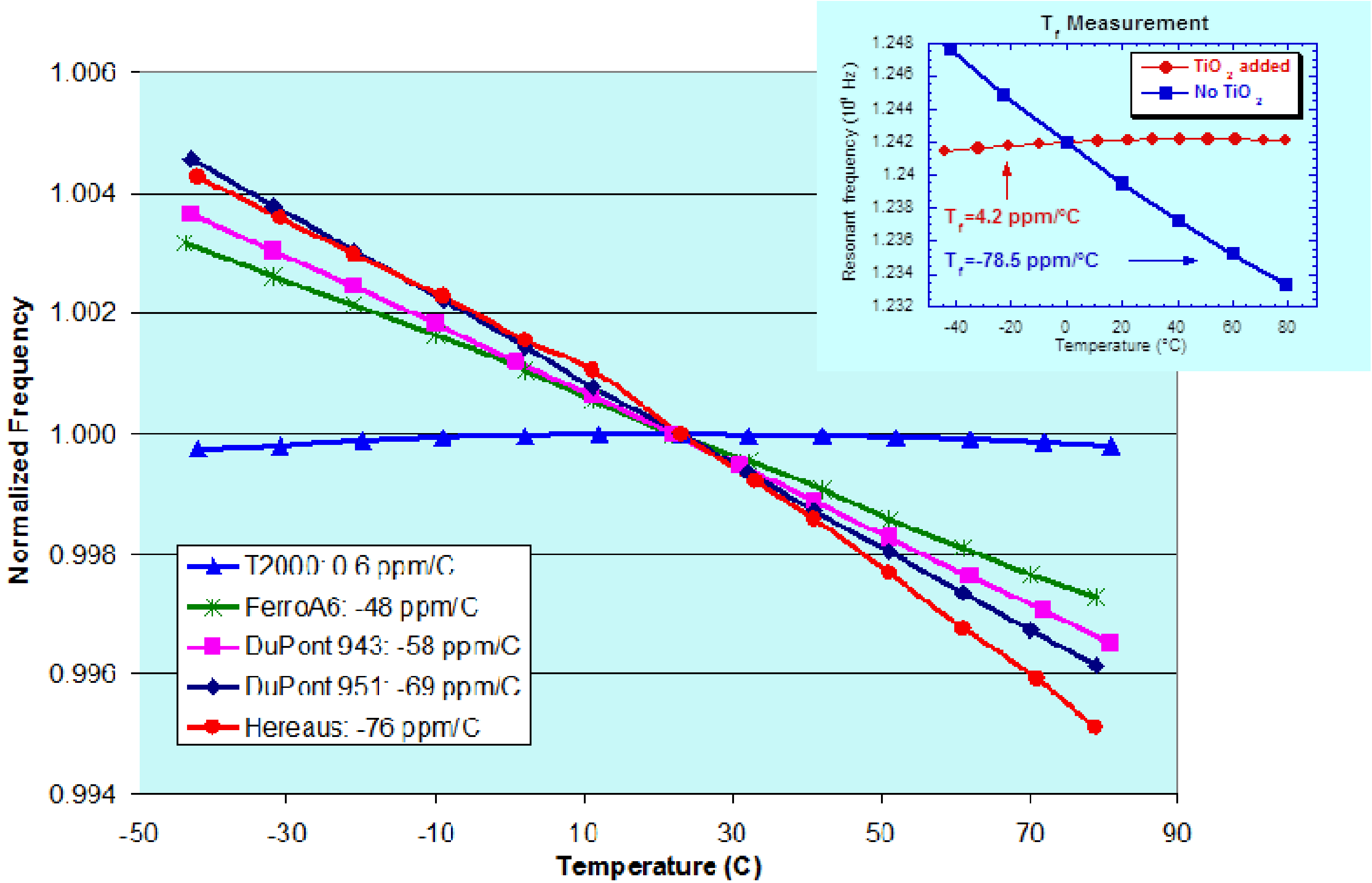
$0\tau_f$ T2000 LTCC

- Glass design
  - K_2O, B_2O_3 and SiO_2, melting temp. and viscosity
  - Loaded with alkaline earth metals
- 6.2wt% TiO_2 added to compensate for τ_f



Global $0\tau_f$

T2000 Temperature Stability



Steve Dai, e al, J. Am. Ceram. Soc. 85[4], 828-32 (2002)

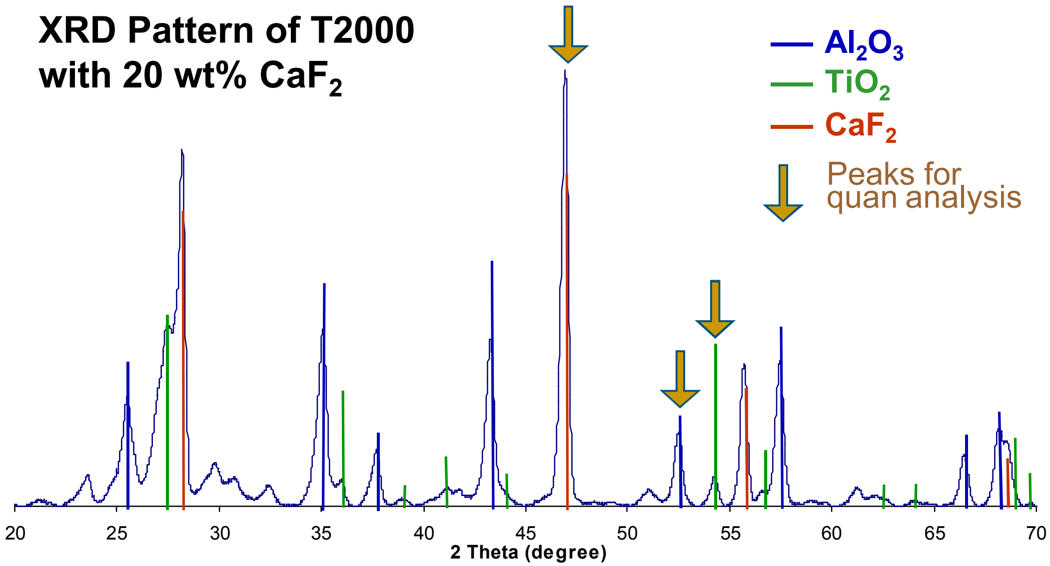
Mystery of τ_f Compensation

	NGK LTCC dielectric (US Patent 5,556,585)	T2000 LTCC
τ_f w/o TiO_2 (ppm/°C)	-50	-79
TiO_2 for $0\tau_f$ (wt%)	15	***> 20?***
Glass ceramic category	Non-reactive	reactive

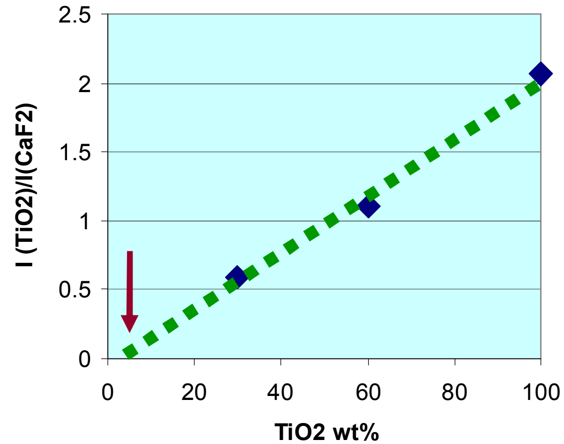
- **τ_f compensation in non-reactive LTCC ~ wt/vol of TiO_2**
- **In T2000 actual TiO_2 = 6.2 wt% for $0\tau_f$, << needed for a non-reactive LTCC**
- **Mechanism of τ_f adjustment by TiO_2 in reactive glass-ceramic LTCC has not been studied**

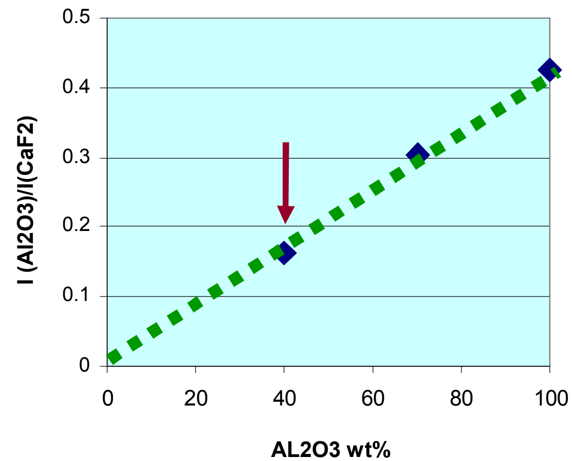
XRD Quantitative Analysis of TiO_2 in T2000

XRD Pattern of T2000 with 20 wt% CaF_2



Calibration Curves





T2000 Quantitative Analysis: (wt%)

	W_{TiO2}	W_{Al2O3}	W_{CaF2}
Ref 1	0	100	20
Ref 2	30	70	20
Ref 3	60	40	20
Ref 4	100	0	20
Sintered T2000	3.5	40.3	20
Unfired T2000	6.2	49.1	20

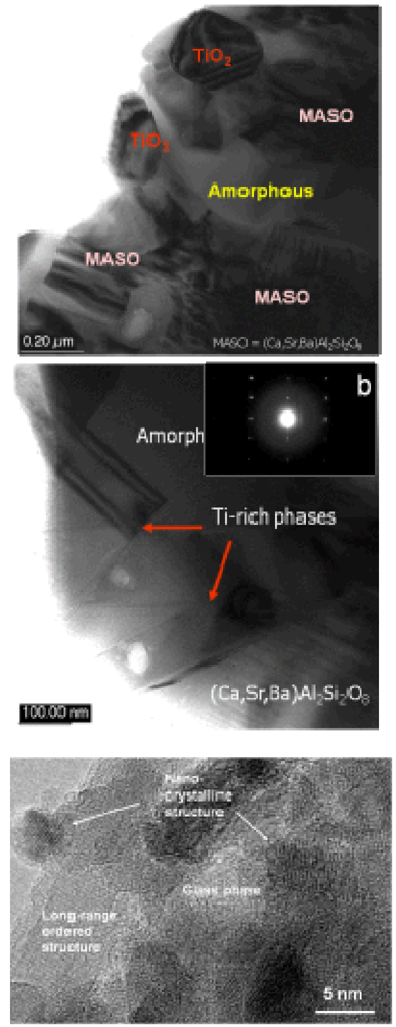
Three Forms of Ti Existence by TEM

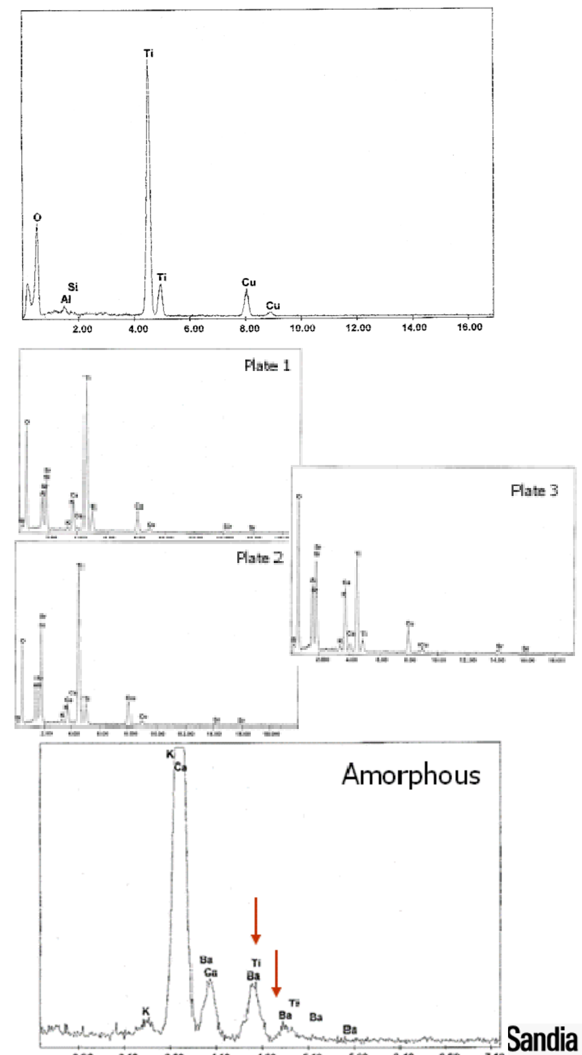
- **TiO_2 particles**

- **Plate-like Ti rich crystalline phase**
  - **Ti content fluctuates from plate to plate**
  - **Nucleation and growth**

- **Ti in amorphous phase**
  - **Dissolved into glass**
  - **Possible nano-crystalline titanates**

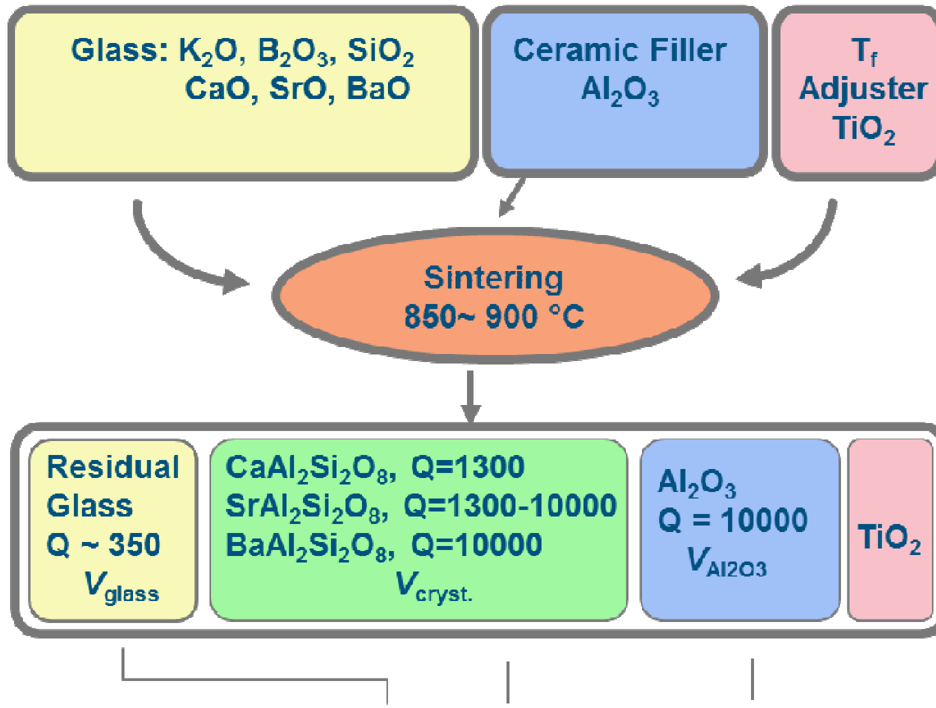
All contribute to $0\tau_f$ T2000 dielectric





Crystallizable High Q LTCC

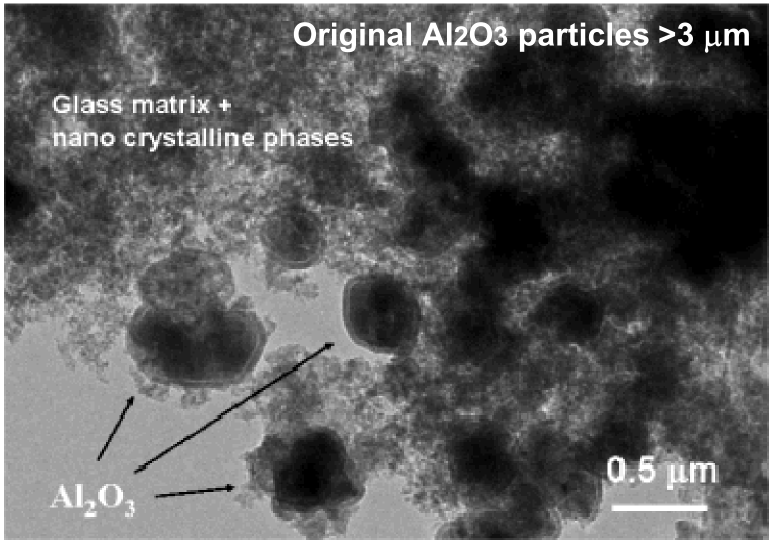
- Glass design
  - K_2O, B_2O_3 and SiO_2, melting temp. and viscosity
  - Loaded with alkaline earth metals
- Sintered composite
  - Formation of high Q anorthite crystalline phases
  - Self limiting process
  - Measured Q: 1050 – 1200
  - Calculated Q: ~ 1100

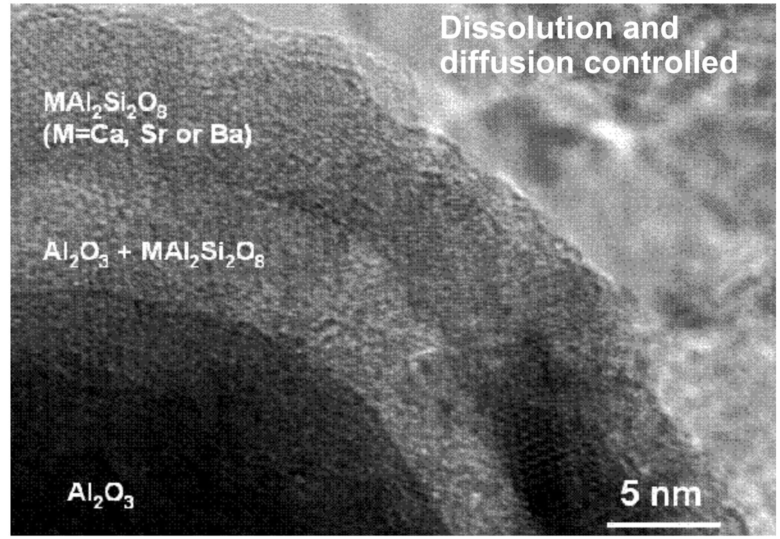


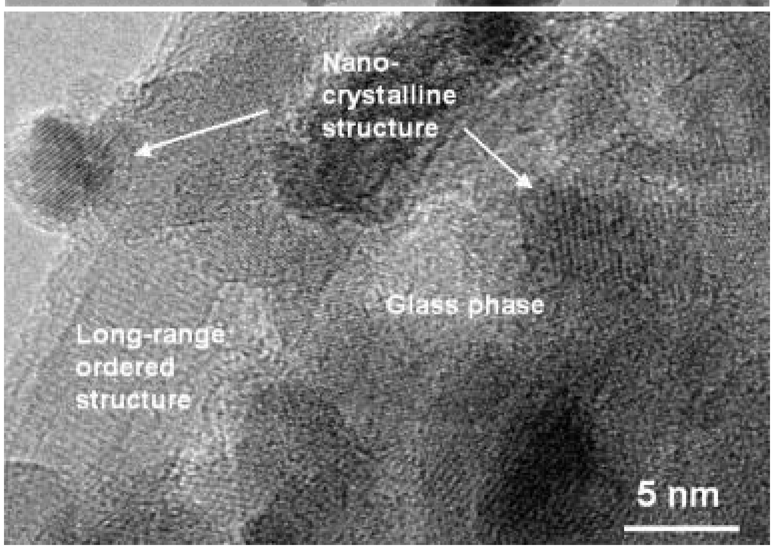
$$\frac{1}{Q} = \sum \frac{V_i}{Q_i} \approx \frac{V_{glass}}{350} + \frac{V_{cryst.}}{1500} + \frac{V_{Al_2O_3}}{10000}$$

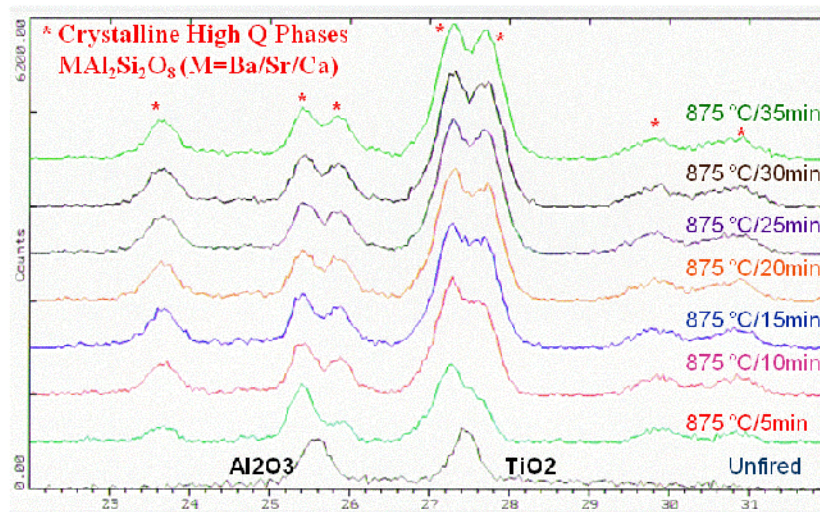
Jau-Ho Jean, Yu-Ching Fang, Steve Dai and David Wilcox, *J. Am. Ceram. Soc.* **84**[6], 1354-60 (2001).

Formation of High Q Phases in Reactive T2000

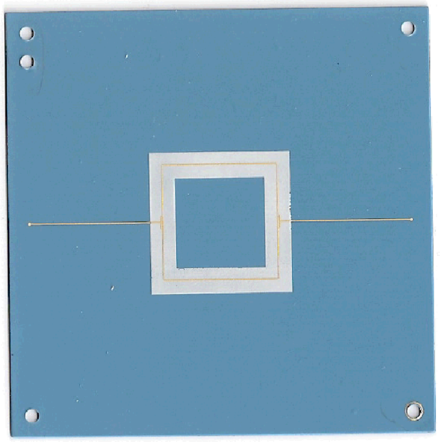








Localized $0\tau_f$ in Existing LTCC?



951 LTCC
- Non-crystallizable glass + Al_2O_3 filler
- τ_f = -69 ppm/°C
- Not designed for RF/MW
- Widely used

Localized $0\tau_f$
- Add-on solution
- Locality = only in needed area/volume

Challenges

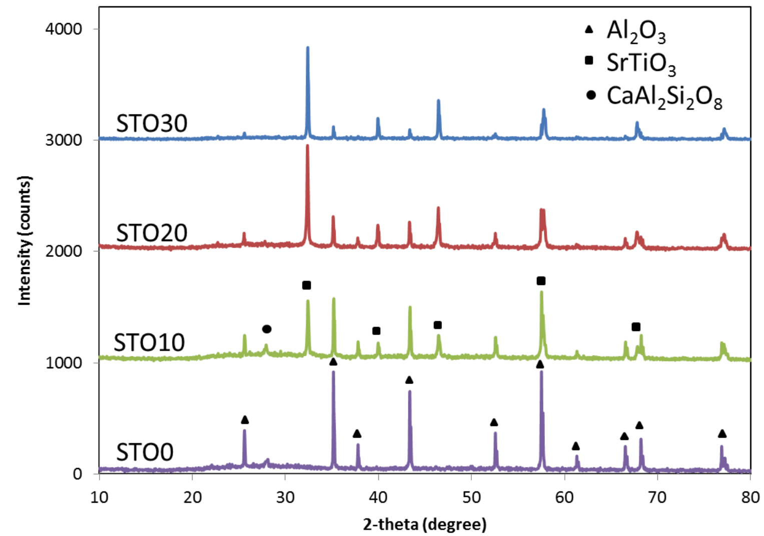
- **Developing τ_f compensating materials**
- **Cofiring of τ_f compensating materials in a multilayer LTCC**
- **Effect of thickness and placement of τ_f compensating material**

τ_f Adjuster Formulation, Property and Microstructure

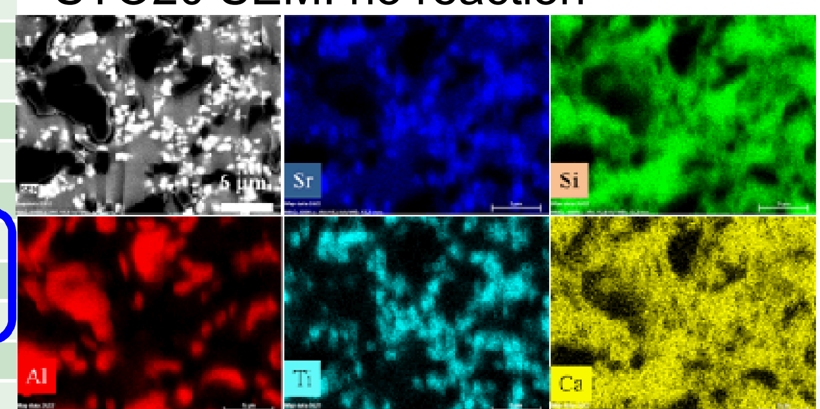
Composition	V-glass (wt%)	Al_2O_3 (wt%)	TiO_2 (wt%)	$CaTiO_3$ (wt%)	$SrTiO_3$ (wt%)	Total wt%
CMB base	55	45				100
CMB TO15	55	30	15			100
CMB TO30	55	15	30			100
CMB CTO10	55	35		10		100
CMB CTO20	55	25		20		100
CMB STO10	55	35			10	100
CMB STO20	55	25			20	100
CMB STO25	55	20			25	100
CMB STO30	55	15			30	100
CMA STO20	60	20			20	100
CMC STO20	49.5	30.5			20	100

Sample	Archimedes bulk density (g/cc)	ε (1 MHz at RT)	τ_ε (ppm/°C)	Estimated τ_f (ppm/°C)
951 LTCC	3.10	7.88	112	-59
CMB base	3.19	7.79	190	-99
CMB TO10	N/A			
CMB TO30	2.77			
CMB CTO10	3.20	9.72	83	-45
CMB CTO20	3.15	12.0	-58	26
CMB STO10	3.21	9.57	10	-9
CMB STO20	3.30	12.22	-240	117
CMB STO25	3.31	13.88	-379	186
CMB STO30	3.35	15.76	-525	259
CMA STO20	3.23	11.96	-241	117
CMC STO20	3.35	12.27	-192	93

STO XRD: no/minimal reaction



STO20 SEM: no reaction



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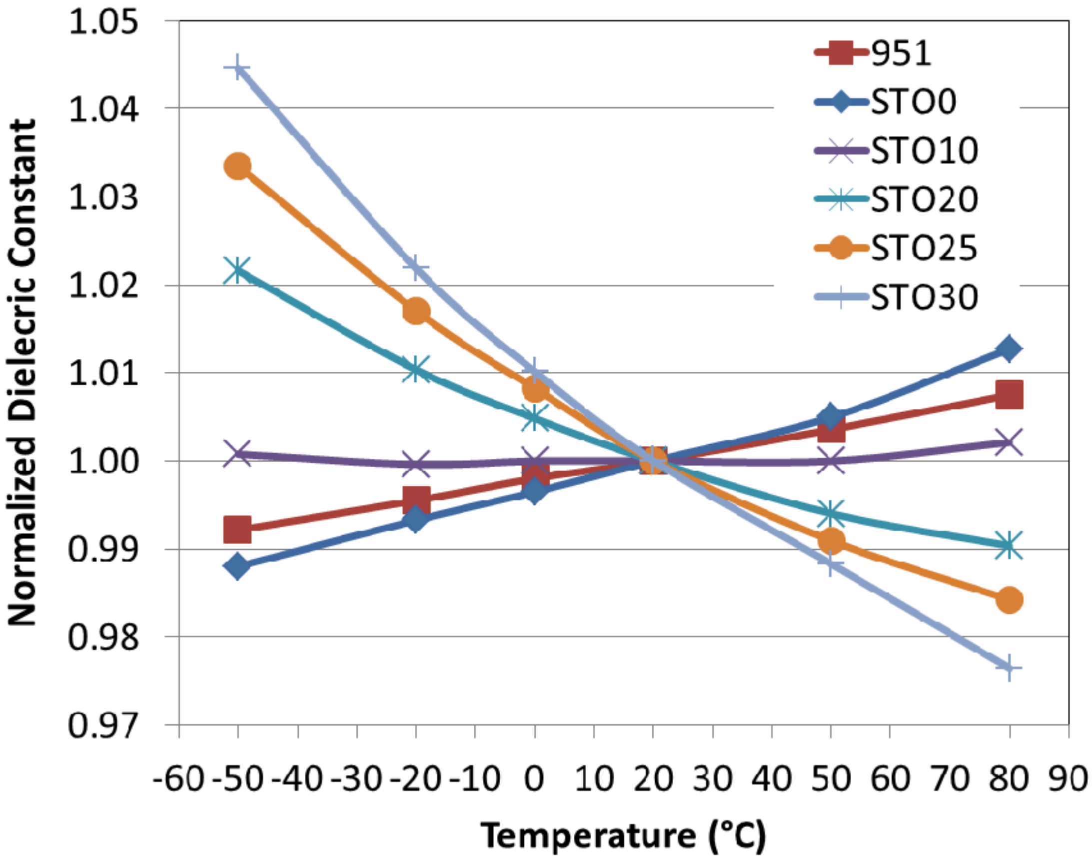
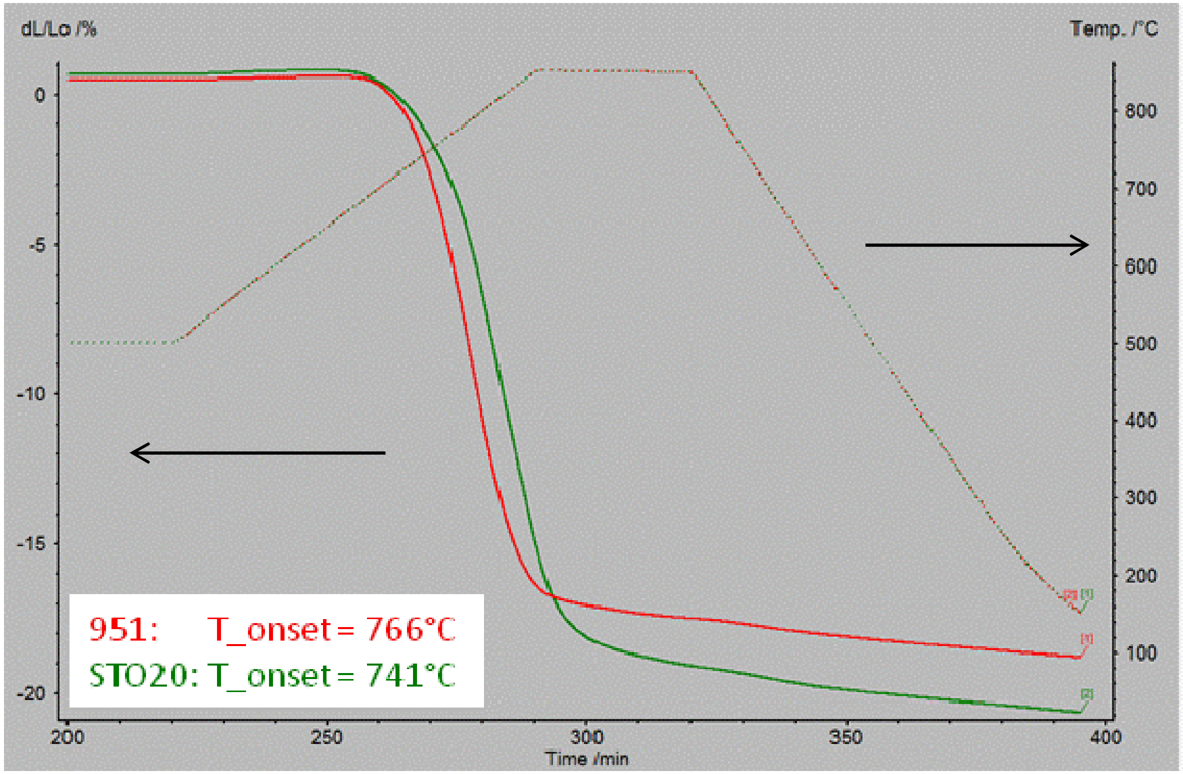
τ_ε of Compensating Materials



- τ_ε = slope of f_r vs T, positive → negative as STO increases
- STO30 has the largest τ_ε →positive τ_f

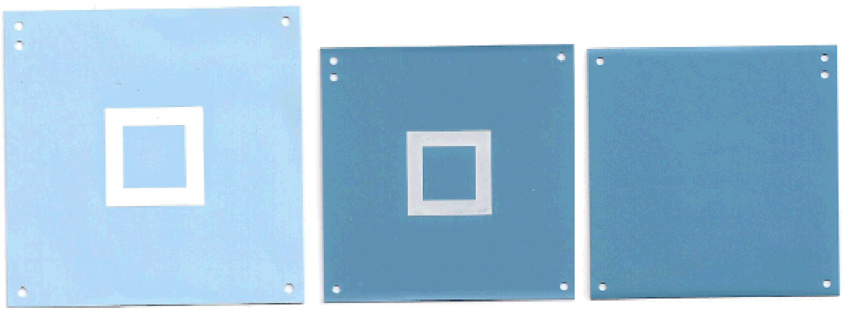
STO20 Cofireability - dilatometry

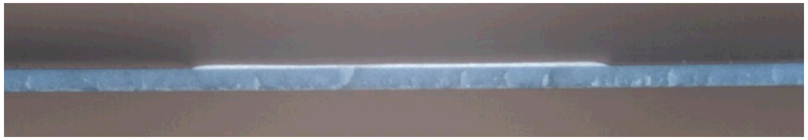


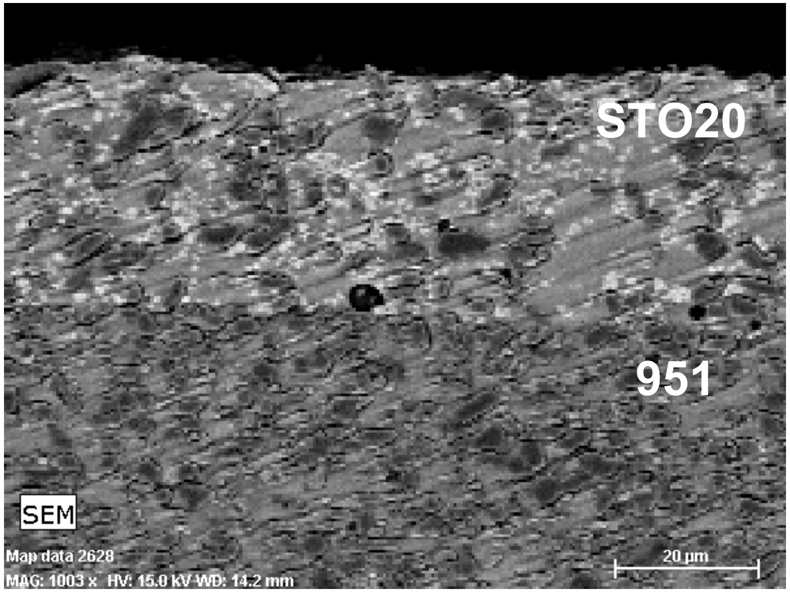
- **The temperatures for the onset of STO20 and 951 shrinkage are closely matched**

STO20 Cofireability – printed layer on 951

Green | Sintered front | Sintered backside

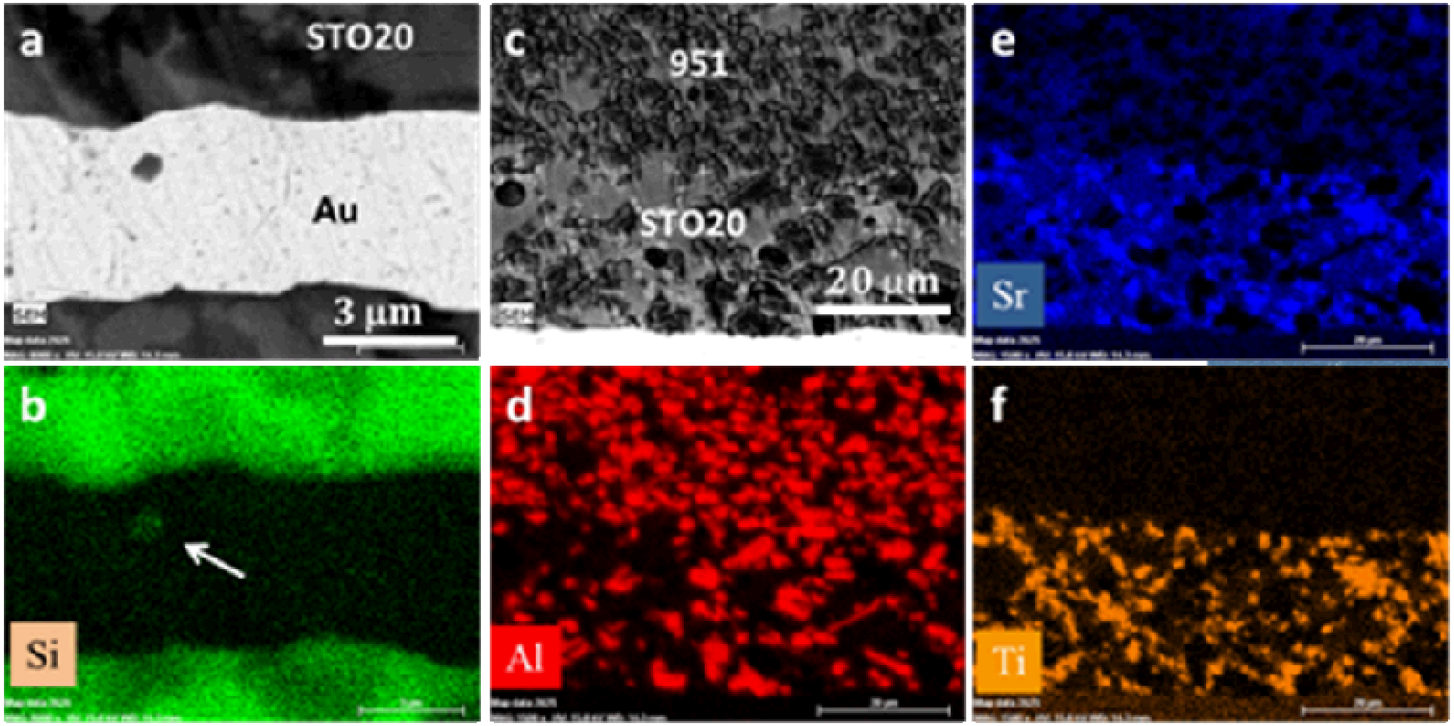


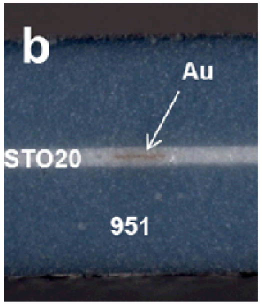




- **No deformation of cofired printed STO layer on a 10 mil 951 tape**
- **Both optical and SEM images show a clean STO20/951 interface, suggesting no or minimal reaction or inter-diffusion**

Cofireability - microstructure





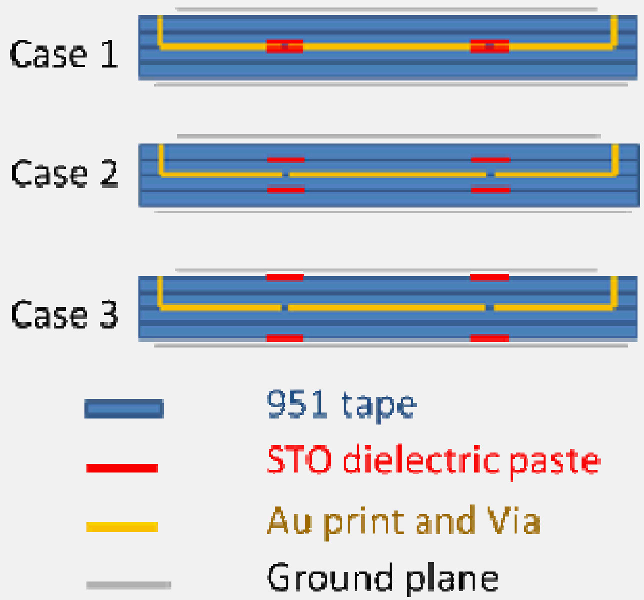
- STO20 layers in a 4-layer 951 panel also sintered flat
- No inter-diffusion from SEM pictures: preserve STO20 properties

3 Variables

- STO content:
  - 20, 25 and 30 wt%

- STO thickness:
  - Simulation: 5 to 50 μm
  - Experiment: 20 and 40 μm

- STO placement
  - Case 1: next to SL
  - Case 2: one LTCC tape away from SL
  - Case 3: two tape layers away from SL, just under surface ground planes



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ε_{eff} of Heterogeneous Multilayer Dielectric

$$\frac{1}{C} = \frac{1}{\pi}\int_0^{\infty} \frac{[\tilde{f}(\beta)/Q]^2}{\beta}\frac{1}{Y}\,d\beta$$

$\beta = 2\pi/\lambda_g$: propagation constant, Q: SL charge

$$\varepsilon_{eff} = \frac{C}{C_o}$$

$$\frac{\tilde{f}(\beta)}{Q} = \frac{8}{5}\frac{\sin(\beta w/2)}{\beta w/2} + \frac{12}{5(\beta w/2)^2}\left[\cos(\beta w/2) - \frac{2\sin(\beta w/2)}{\beta w/2} + \left(\frac{\sin(\beta w/4)}{\beta w/4}\right)^2\right]$$

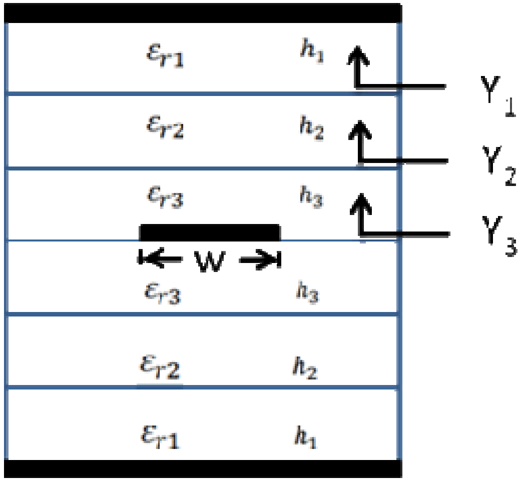
Fourier transform of the charge density distribution

$$Y_1 = \varepsilon_o\varepsilon_{r1}\coth(\beta h_1)$$

$$Y_2 = \varepsilon_o\varepsilon_{r2}\frac{Y_1 + \varepsilon_o\varepsilon_{r2}\tanh(\beta h_2)}{\varepsilon_o\varepsilon_{r2} + Y_1\tanh(\beta h_2)}$$

$$Y_3 = \varepsilon_o\varepsilon_{r3}\frac{Y_2 + \varepsilon_o\varepsilon_{r3}\tanh(\beta h_3)}{\varepsilon_o\varepsilon_{r3} + Y_2\tanh(\beta h_3)}$$

Y_1, Y_2, Y_3 are transverse transmission line admittance



$$Y = 2\varepsilon_o\varepsilon_{r3}\frac{Y_2 + \varepsilon_o\varepsilon_{r3}\tanh(\beta h_3)}{\varepsilon_o\varepsilon_{r3} + Y_2\tanh(\beta h_3)}$$

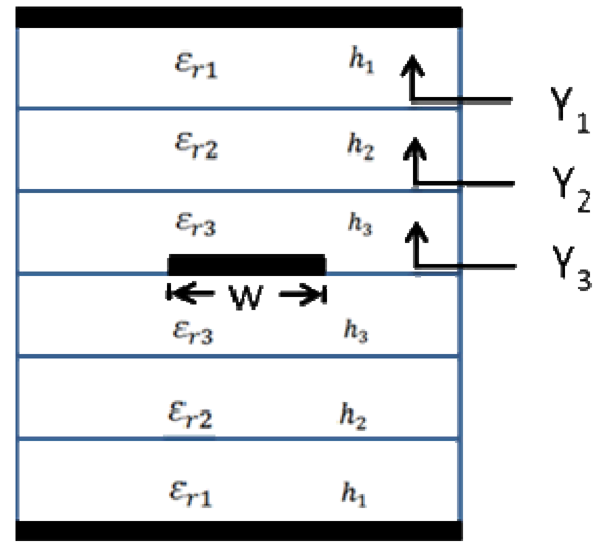
ε_{eff} and f_r

$$\frac{1}{C_o} = \frac{1}{\pi}\int_0^{\infty} \frac{\left[\frac{\tilde{f}(\beta)}{Q}\right]^2}{\beta} \frac{1}{Y_o} d\beta$$

$$\varepsilon_{eff} = \frac{C}{C_o}$$

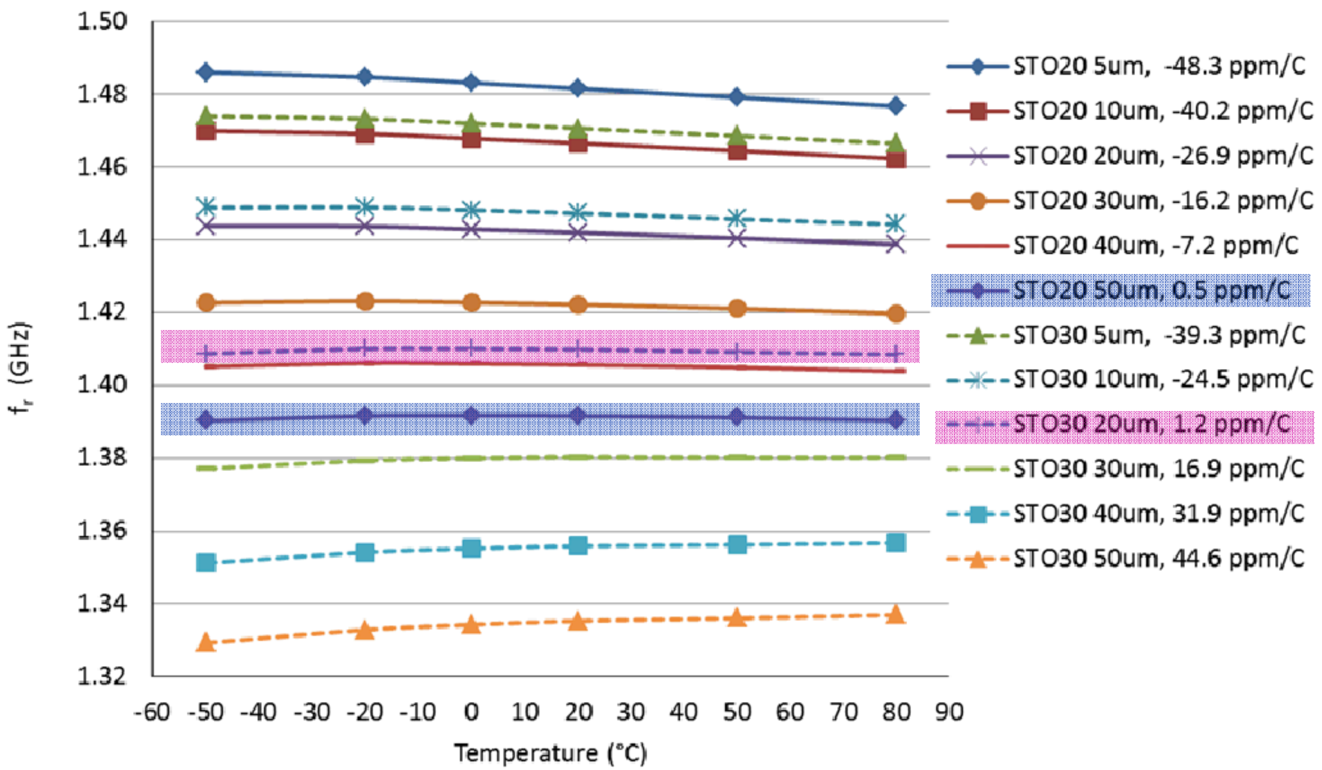
$$Y_{30} = \varepsilon_o coth[\beta(h_1 + h_2 + h_3)]$$

$$Y_o = 2Y_{30} = 2\varepsilon_o coth[\beta(h_1 + h_2 + h_3)]$$



Method of Momentum EM simulation → f_r

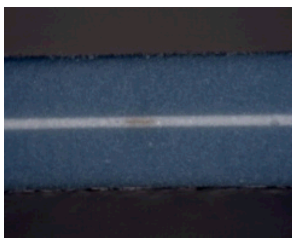
Effect of Composition and Thickness for Case 1



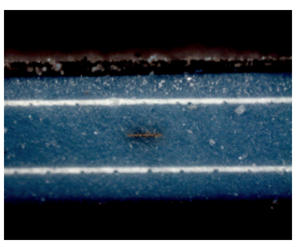
- f_r
  - STO20 shows 0 τ_f at 50 μm
  - STO30 shows 0 τ_f at 20 μm
- τ_f compensation
  - Scales to wt% of STO
  - Scales to thickness

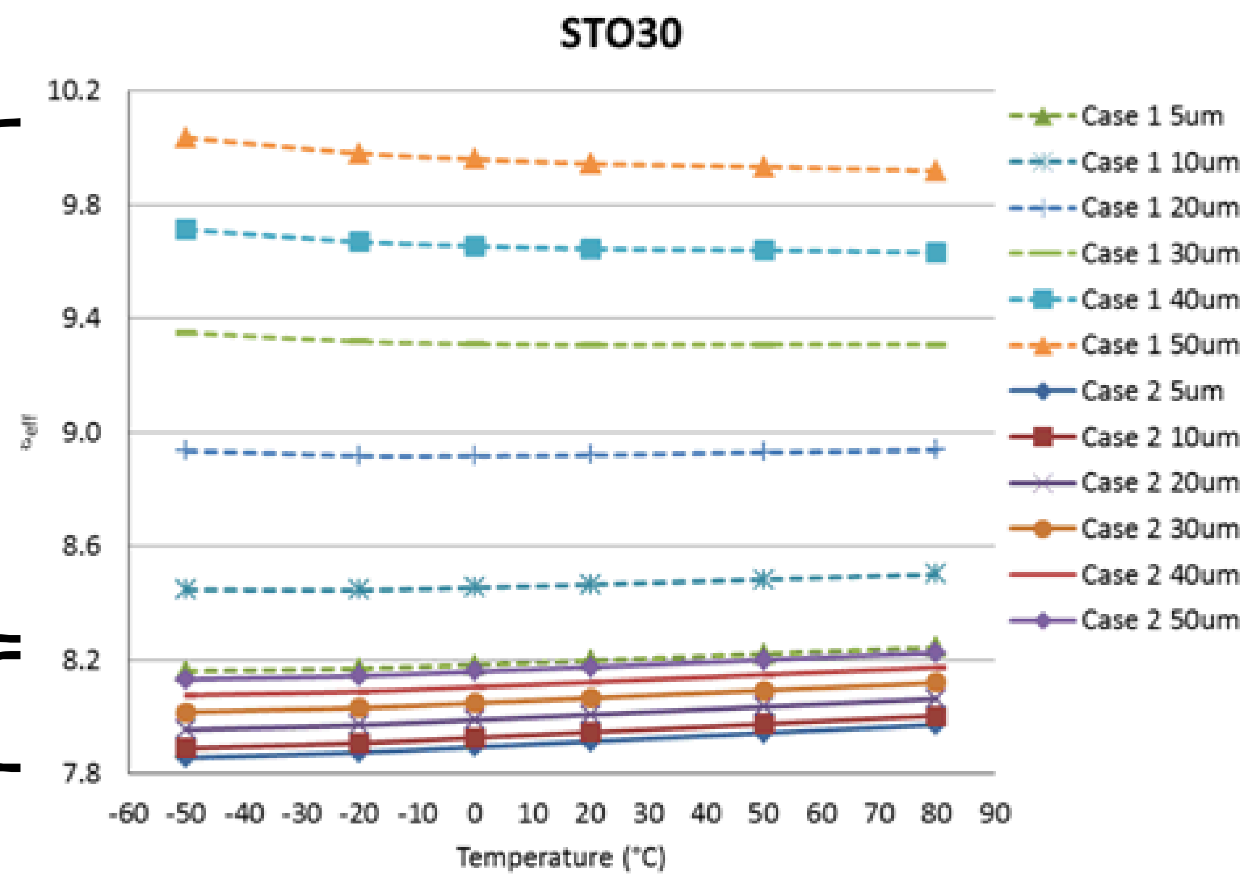
Effect of Configuration: ε_{eff}

Case 1



Case 2

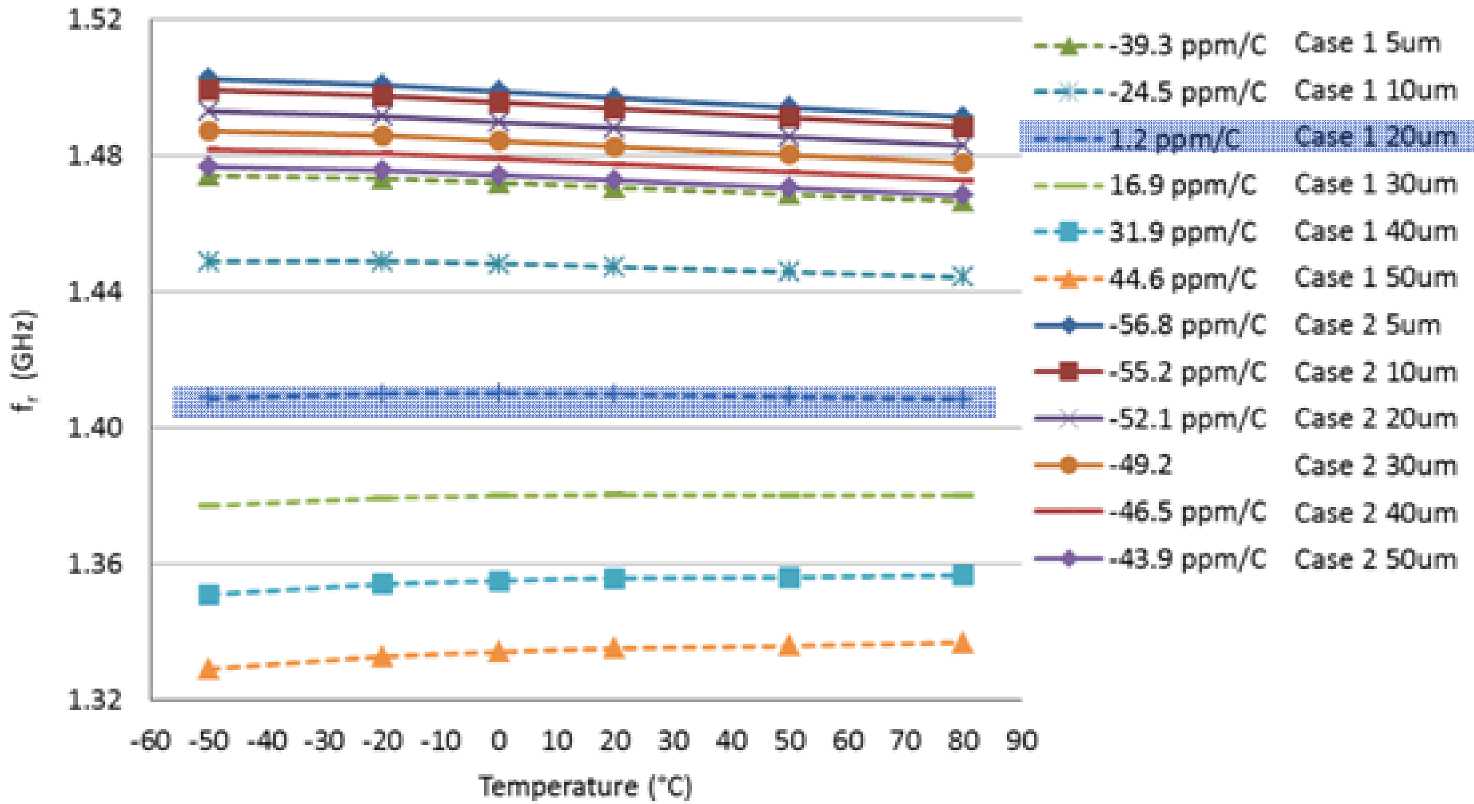




- **Case 1**
  - **Increases of ε_{eff} significant**
  - **A crossover in slope**
- **Case 2**
  - **Change of ε_{eff} minimal**
  - **SL sees much less STO**

Localized $0\tau_f$ Simulation

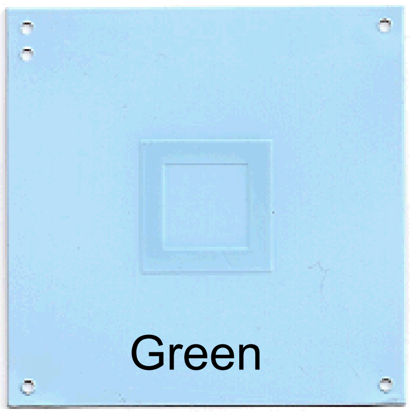
Effect of Configuration: f_r

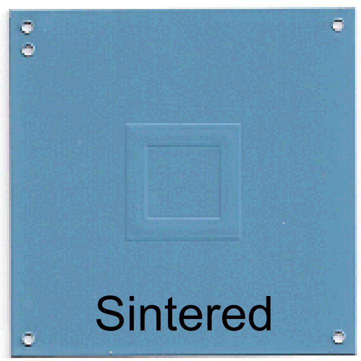


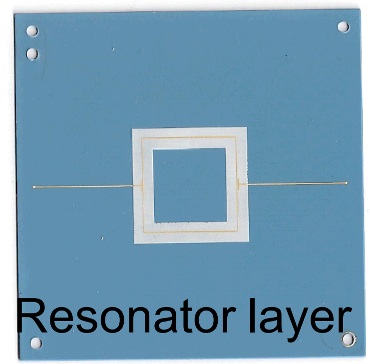
- f_r
  - STO30 shows 0 τ_f at 20 μm in case 1
  - Never reaches 0 τ_f in case 2

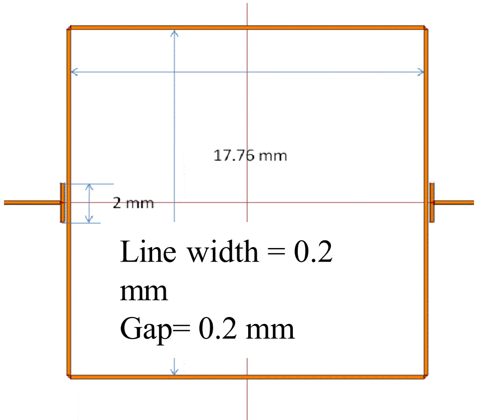
- τ_f compensation
  - Placement of STO is critical
  - Most effective when next to SL

SL Resonator Panels

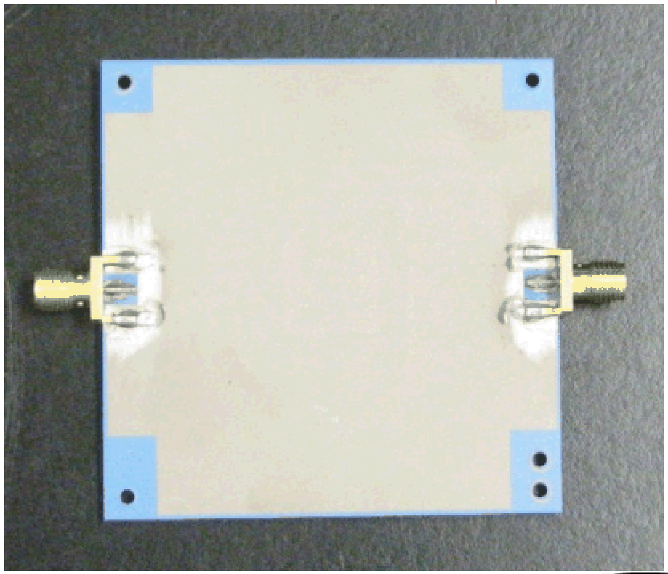




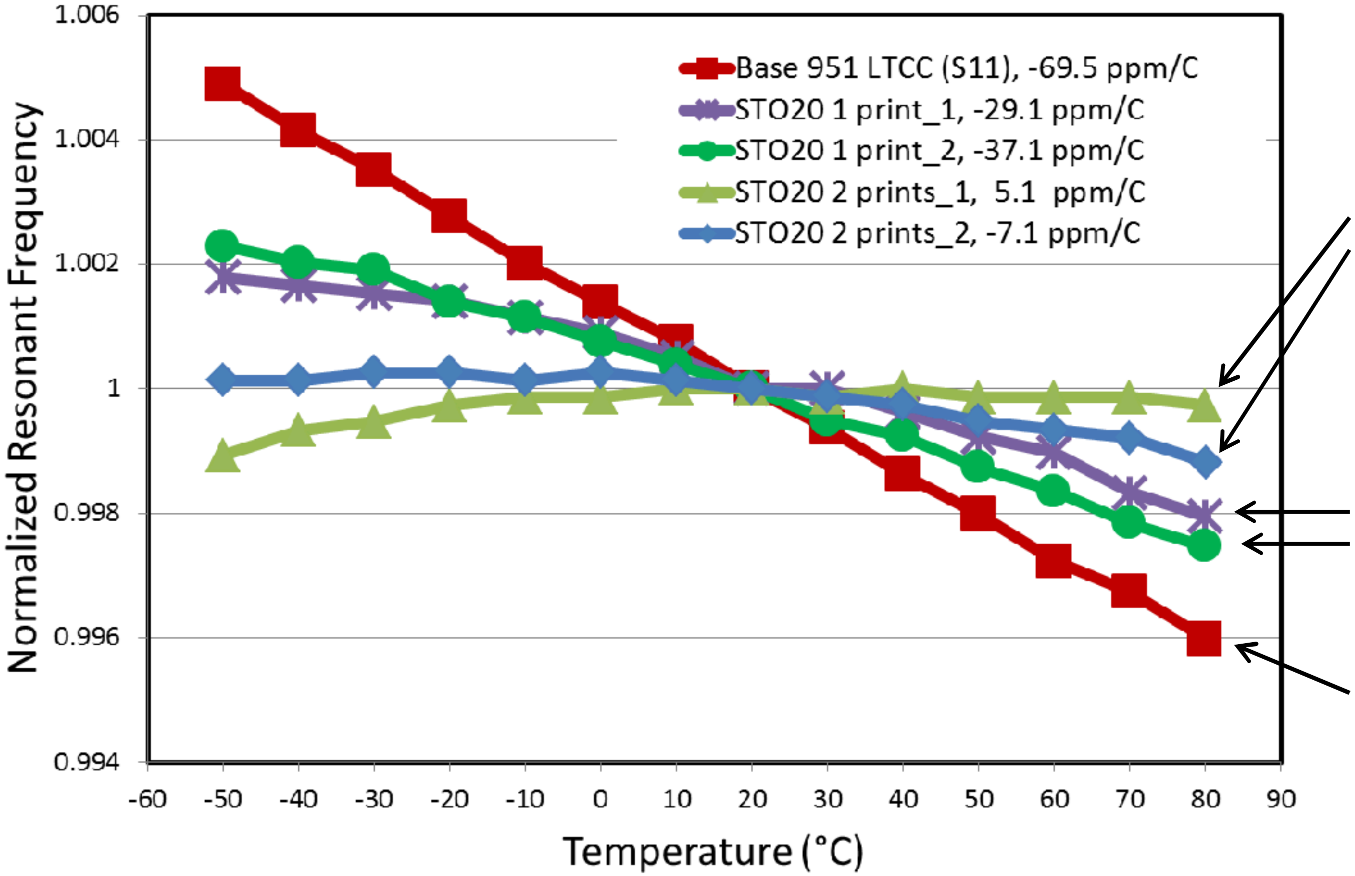


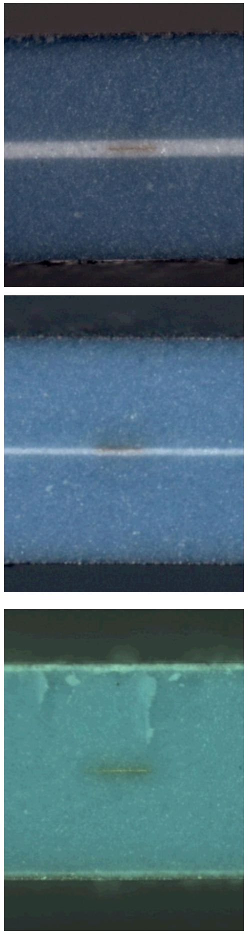


- **Standard LTCC fabricated with 951 matching conductors**
- **4 layers of 10 mil 951 tape. 50 Ω lines. f_r = 1.5 GHz**
- **No localized deformation, indicating no sintering mismatch**
- **Embedded STO shown as "embossed" feature on the non-contact side during lamination**



τ_f Results for STO20





- **A true, near zero or zero τ_f is achievable**
- **Variation of τ_f, possibly by varying the STO20 thickness**

Steve Dai, "Localized Temperature Stability in Multilayer LTCC", J of Microelec and Elect Packaging, 8, 1-6, 2011

SL Resonators

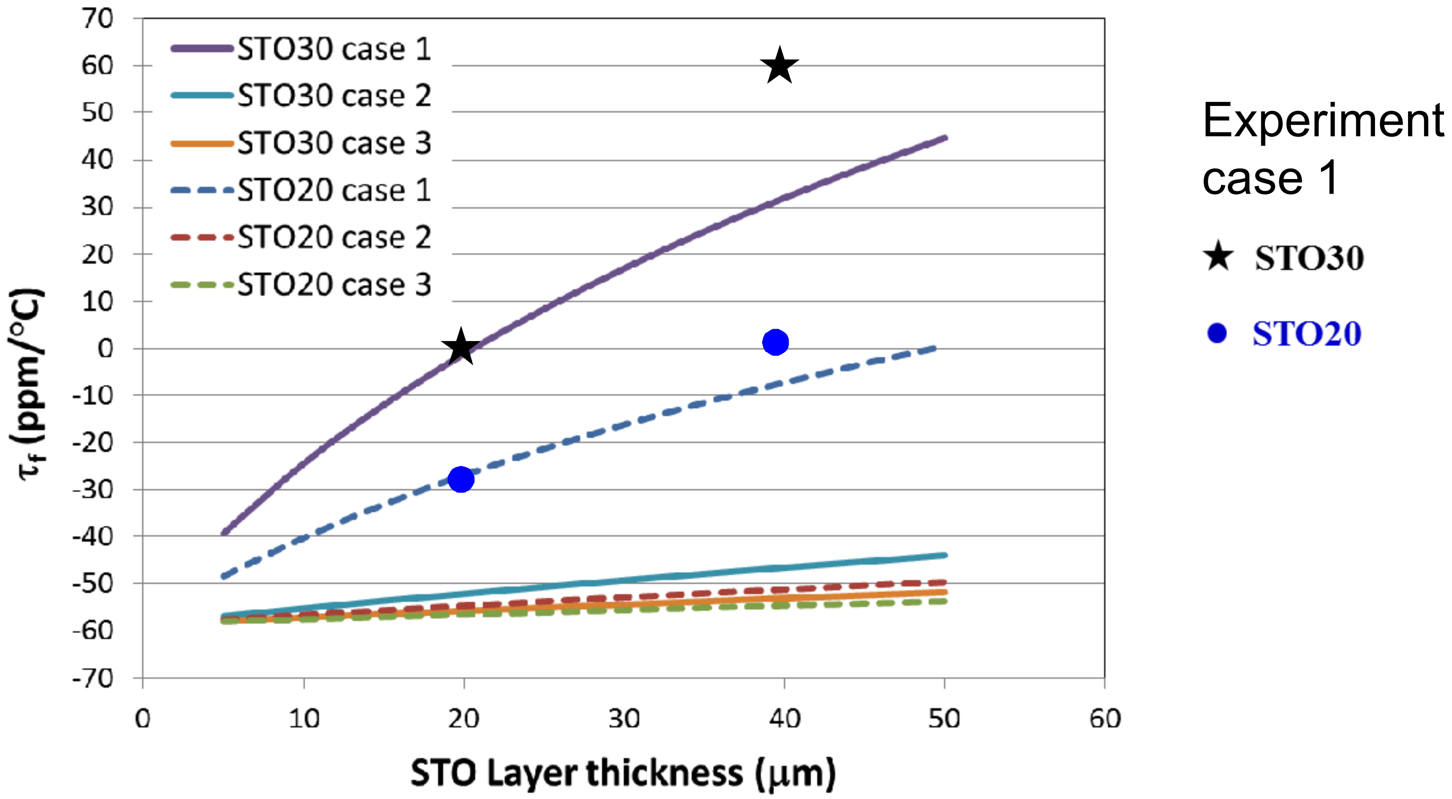
STO composition	Appx STO Thickness (μm)	τ_f (ppm/°C)		
		Case 1	Case 2	Case 3
STO20	20	-28.8	-67.5	-71.2
	40	**1.9**		
STO30	20	**1.0**	-61.7	-69.3
	40	60.0	-54.8	-62.3

- **Only in case 1 a ~ 0 τ_f is realized**
  - **STO20**
    - **τ_f = -28.8 ppm/°C at 20 μm**
    - **τ_f = +1.9 ppm/°C at 40 μm**
  - **STO30**
    - **$0\tau_f$ = +1.0 ppm/°C at 20 μm**
    - **τ_f over-corrected at 40 μm**

- **In cases 2 and 3, the τ_f is far from 0 ppm/°C**

- **Placing τ_f adjuster next to SL is essential for τ_f compensation**

Experiment Versus Simulation



- Good match for STOs at 20 μm
- Simulation underestimate at thicker STO

Sandia National Laboratories

τ_f of a Dielectric Composite

- **Why the STO is effective only in case 1 (= next to the SL)?**

- **τ_f expressed as**

$$\tau_f = \sum_i P_i \tau_{fi} \qquad\qquad \boldsymbol{P_i = E_i / E_{total}}$$

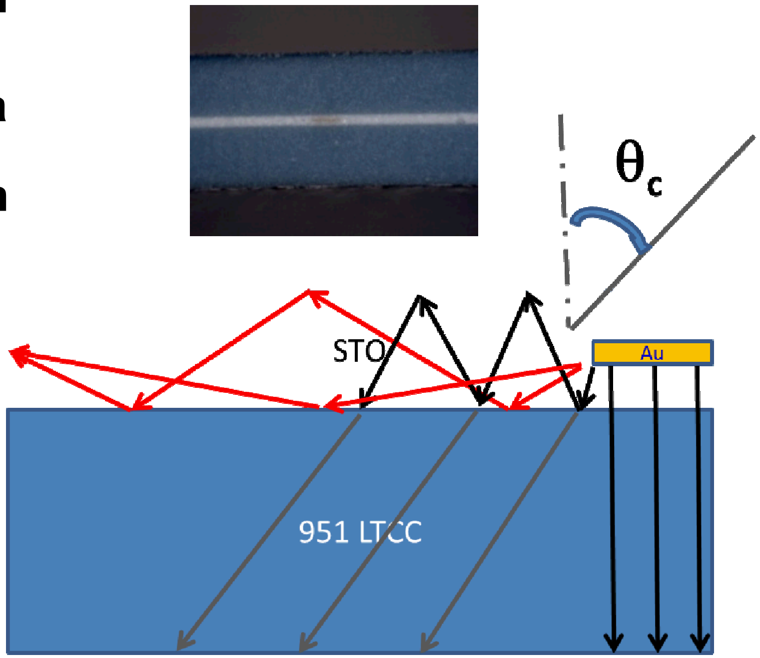
where P_i is defined as the energy filling factor, the fraction of total EM energy confined in dielectric phase *i*

- **P_i depends on dielectric constant, volume, geometry of dielectric *i*, as well as the EM field distribution inside the dielectric**

Energy Concentration in STO Layers Case 1

- **Low aspect ratio → fringing electrical field**
- **EM wave propagates high ε → low ε media**
- **Critical angle for total internal reflection in case 1**
  - ε_{STO30} = 15.76
  - ε_{LTCC} = 7.88
- **Critical angle (Snell's law)**

$$\theta_c = \text{asin}\left(\frac{n_{LCTT}}{n_{STO30}}\right) = \text{asin}\left(\frac{\sqrt{\varepsilon_{LTCC}}}{\sqrt{\varepsilon_{STO30}}}\right) = 45°$$



$\theta_c \geq 45°$, total internal reflection in STO

$\theta_c < 45°$, reflection/refraction at the STO-951 interfaces

Energy concentration in STO → effective τ_f compensation

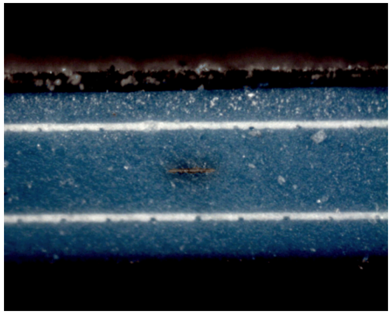
No Energy Concentration in Cases 2 and 3

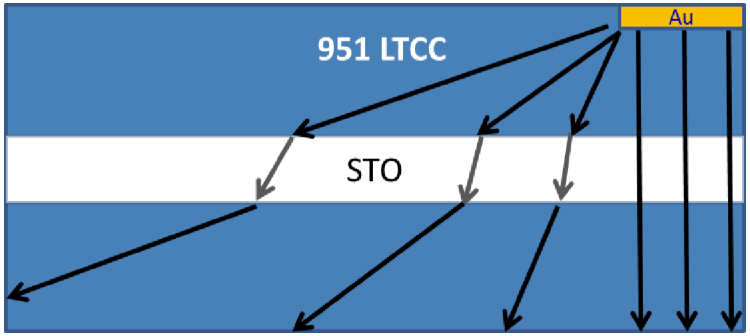
- EM wave propagates low ε → high ε → low ε media

- No total internal reflection

- Energy in STO scales to volume

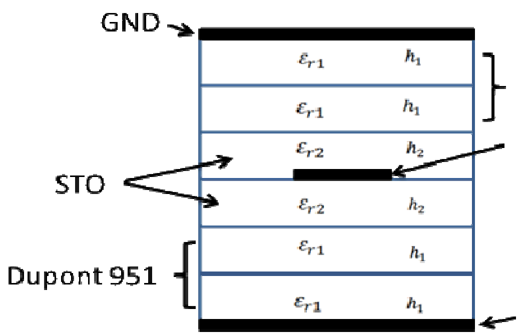
- Overall τ_f of SL resonator is dominated by 951 LTCC

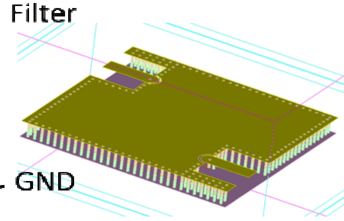




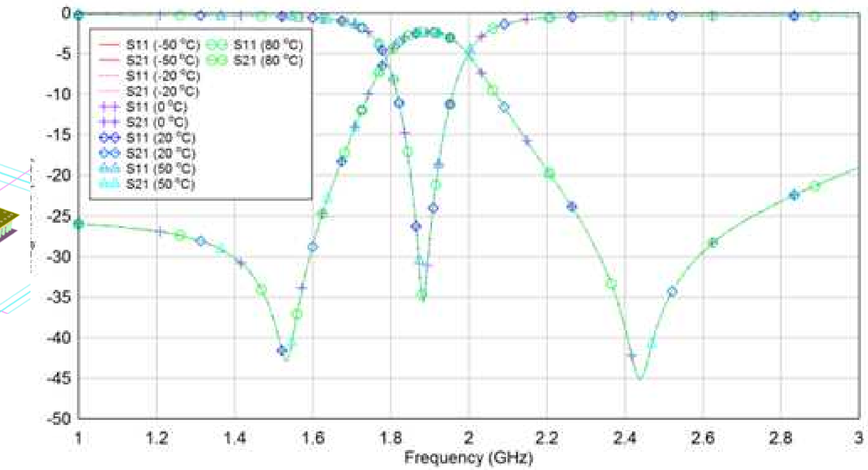
Steve Dai, "Localized Temperature Stability in Multilayer LTCC", submitted to J Am Ceram Soc

$0\tau_f$ Demo: design of a S-band filter

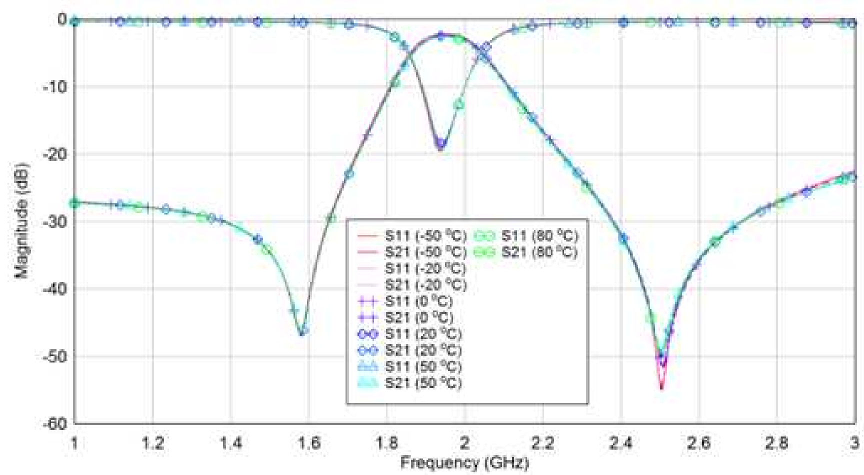




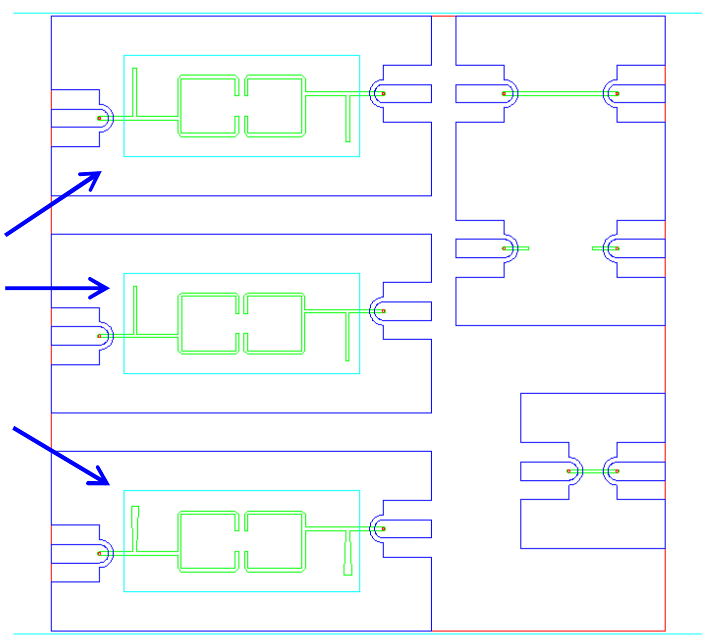
Simulation (20 µm STO30)



Measurement (τ_f = 1.8 ppm/°C)

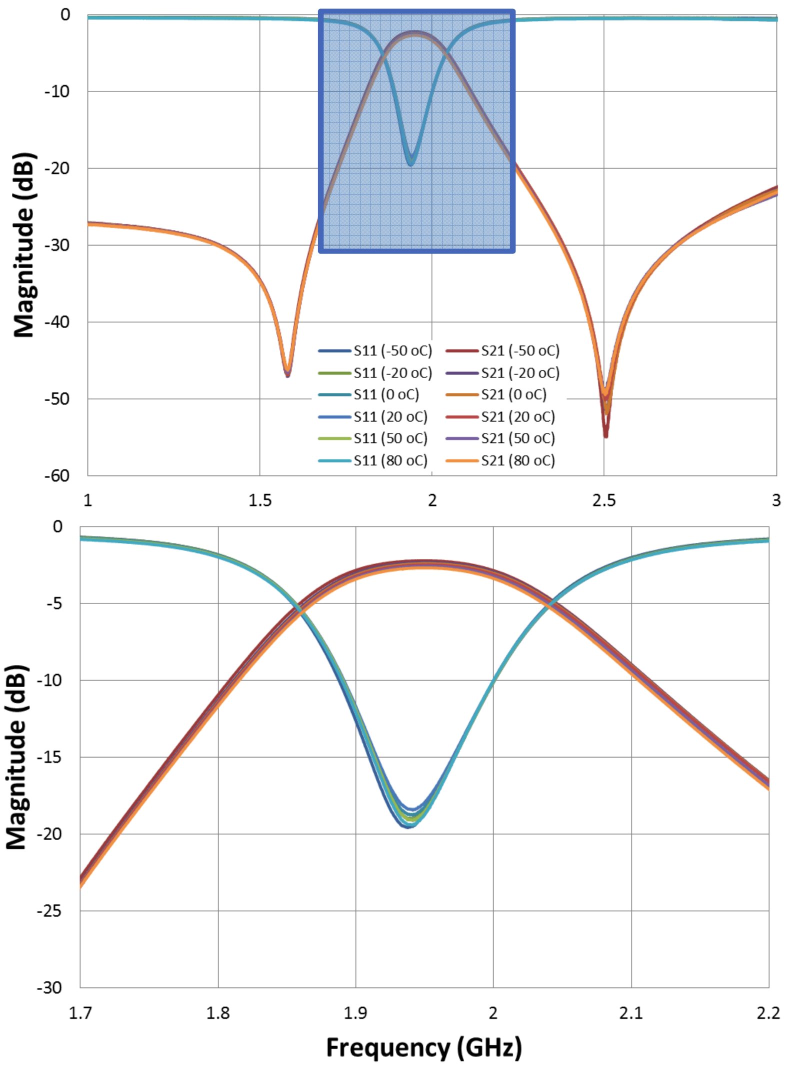


STO layers

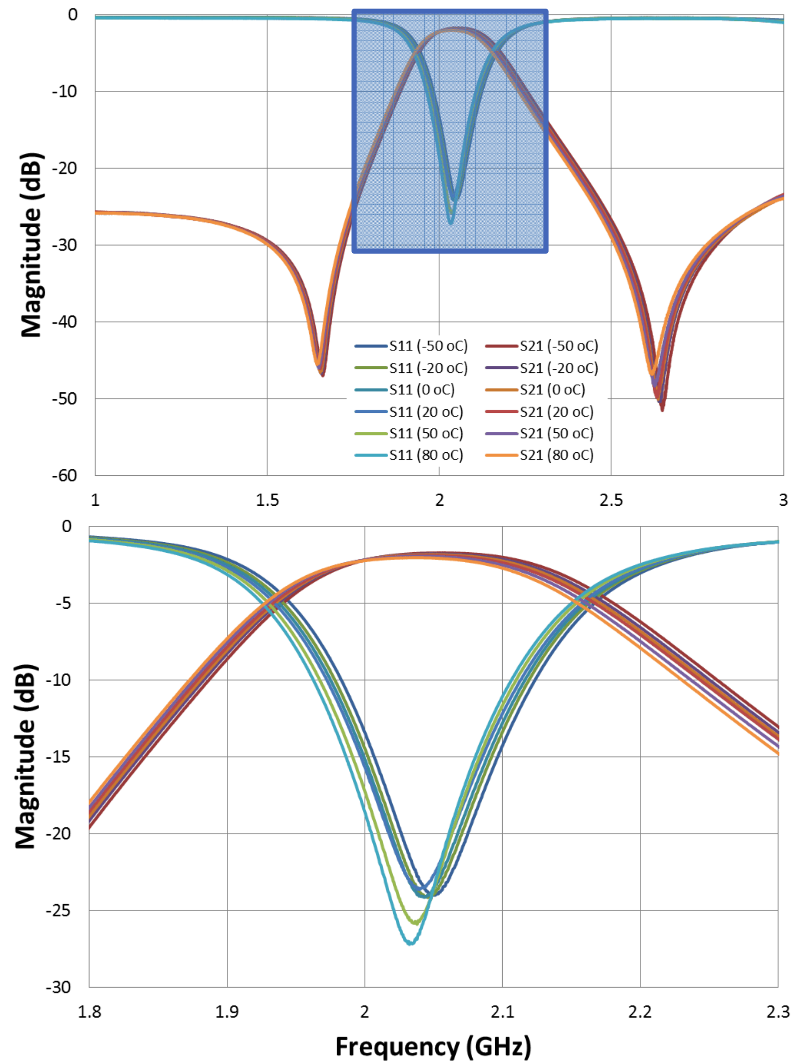


$0\tau_f$ Demo: filter measurement

With 20 μm STO30, τ_f = 1.8 ppm/°C



Without STO, τ_f = -71.2 ppm/°C



Summary on localized $0\tau_f$

- τ_f compensating materials compatible to existing 951 LTCC have been successfully developed (patent pending)
- SL resonators with a near $0\tau_f$ have been demonstrated by locally integrating a τ_f compensating material in a multilayer LTCC package
- Placing compensating materials next to the SL resonator is essential for τ_f compensation, owing to the EM energy concentration in the compensating dielectric layers
- S-band filter with $0\tau_f$ has been demonstrated

Acknowledgement

- The authors thank Adrian Wagner, Tom Chavez , Dennis De Smet and Shelley Williams for help in LTCC fabrication and measurement

- This work is supported by the Laboratory Directed Research and Development (LDRD) program at Sandia National Laboratories, a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000

Thank You!

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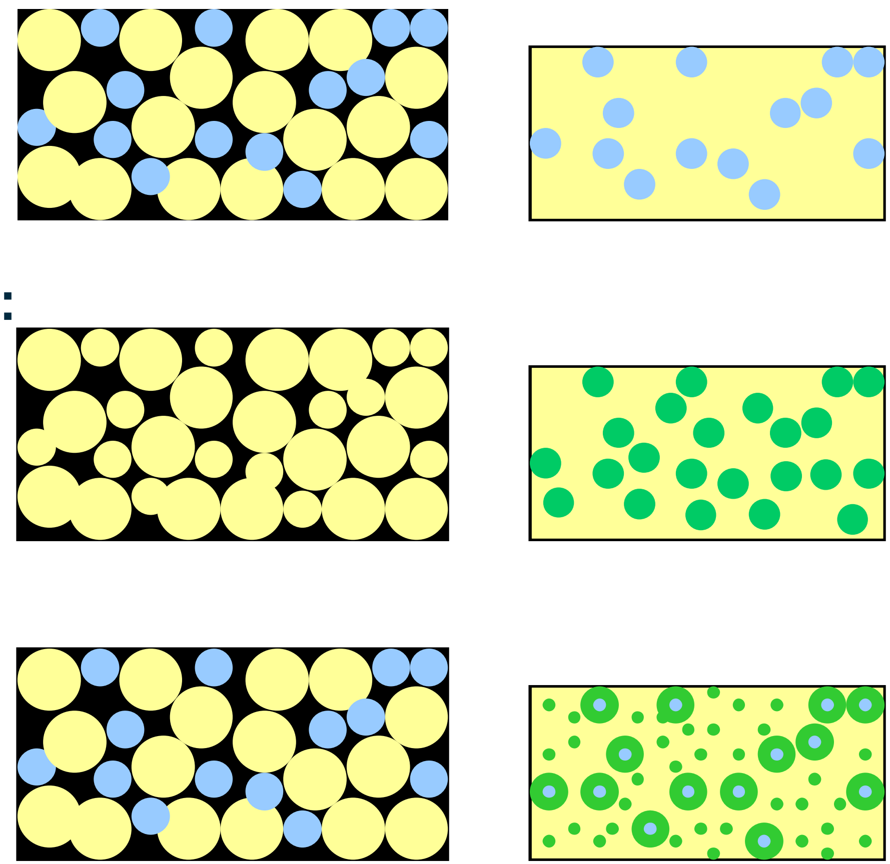
LTCC Dielectrics

Green | Sintered



Type 1, no crystallization:

- Unreactive, glass = low T sintering agent

DuPont 951

Type 2, self-derived crystallization:

- Re-crystallizeable, partial formation of crystalline high Q phases

Ferro A6

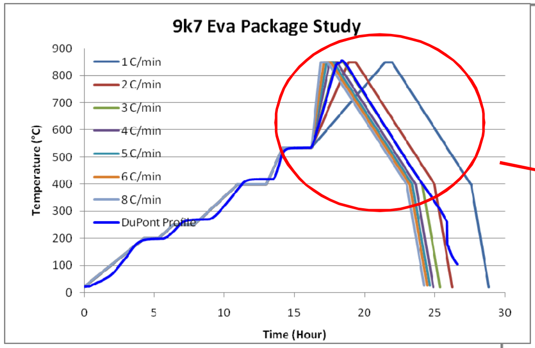
Type 3, reaction-derived crystallization:

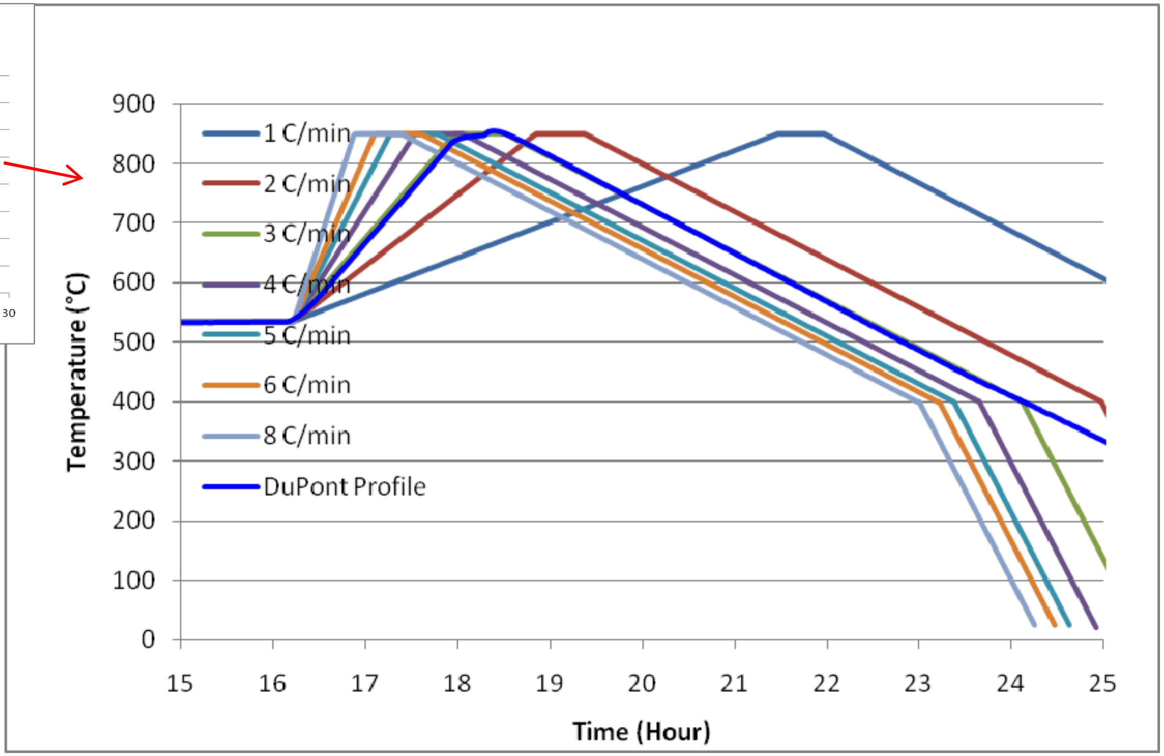
- Reactive, forming high Q crystalline phases

Motorola T2000

Organic binder | Glass | Ceramic filler (Al_2O_3) | Crystalline phase

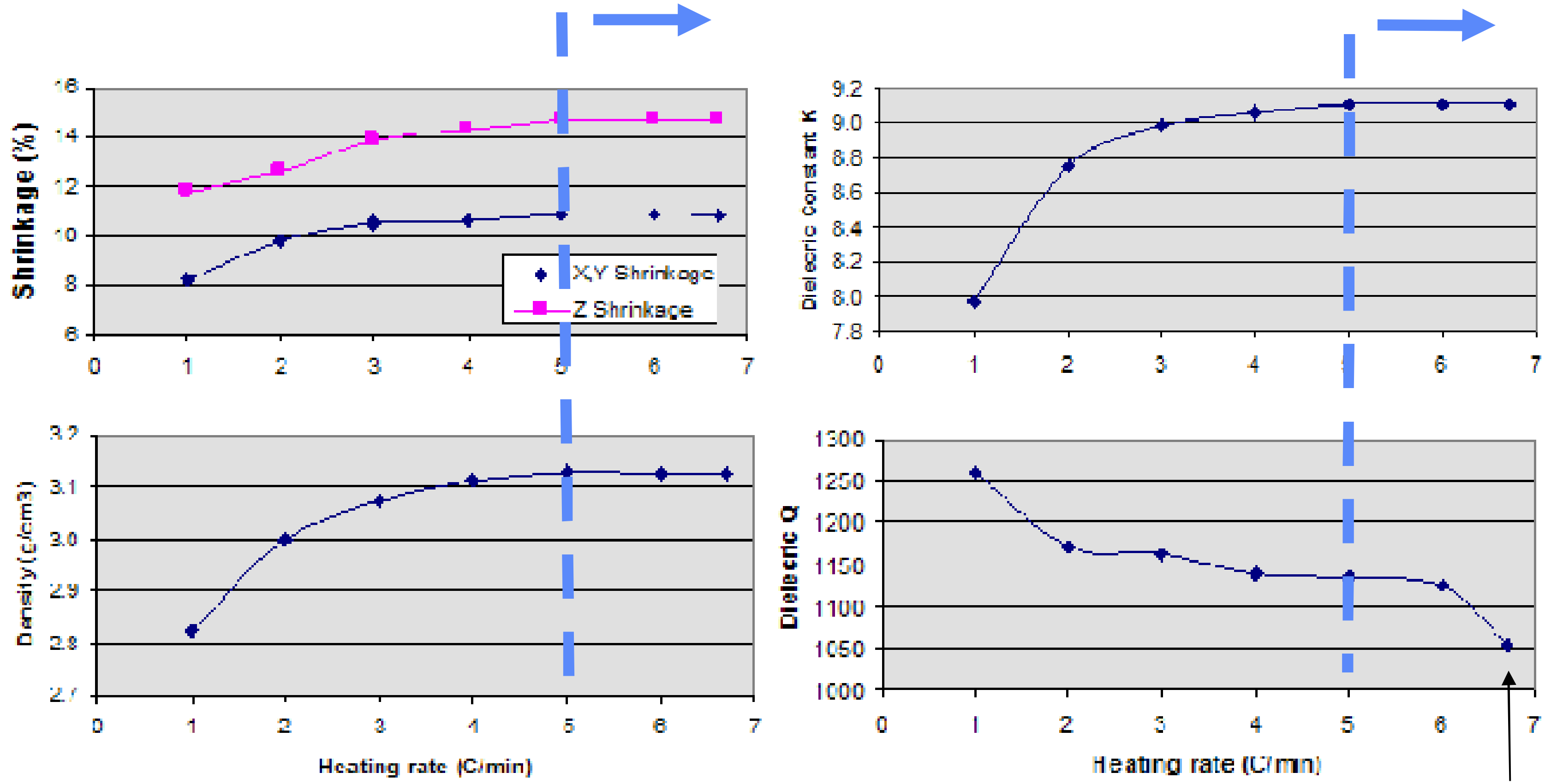
Heating Rate





- **Heating rate critical for crystallization versus densification**

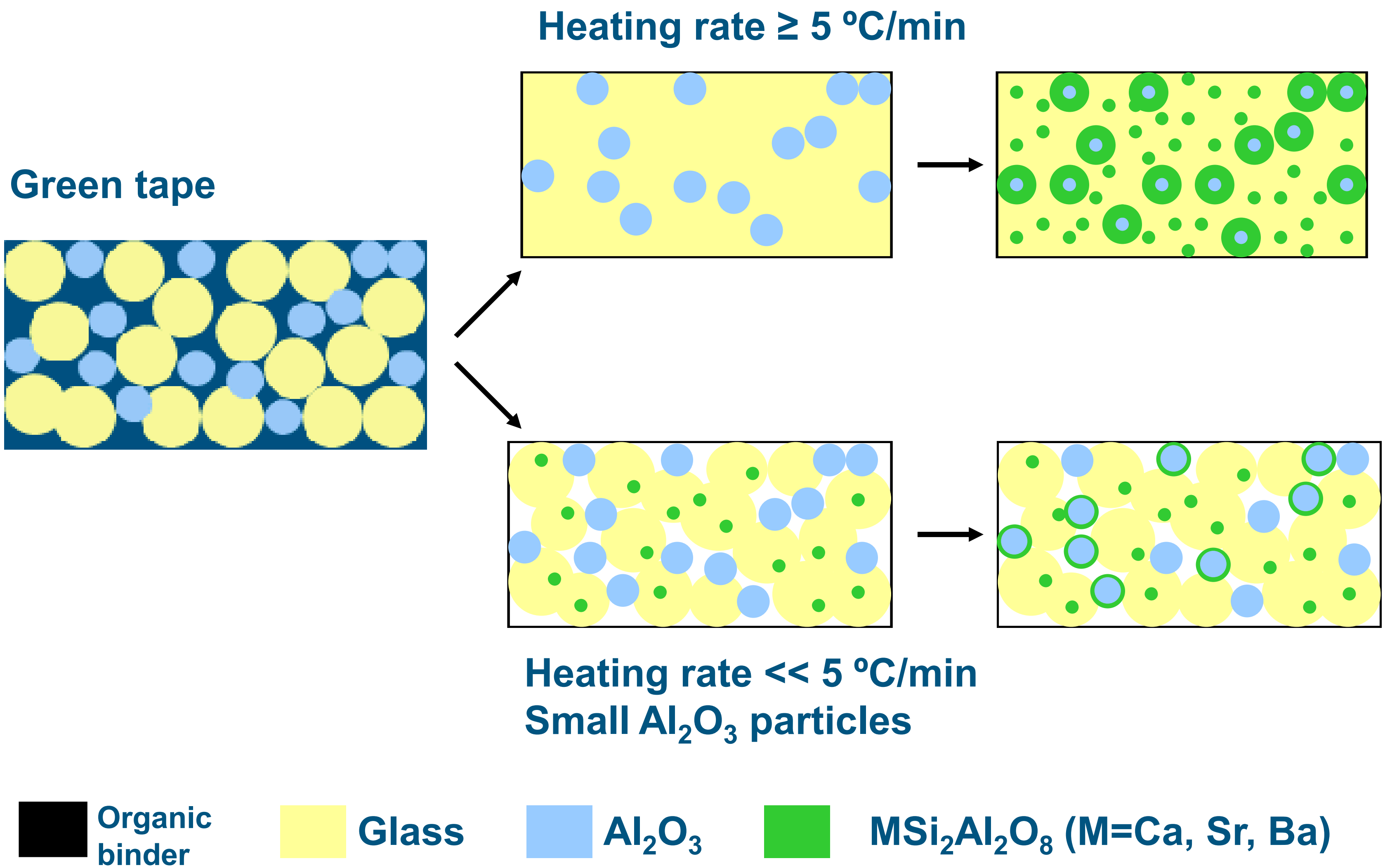
Effect of Heating Rate in 450~875 ºC



Heating rate 6.7 ºC/min too high, 30 min sintering time not reached

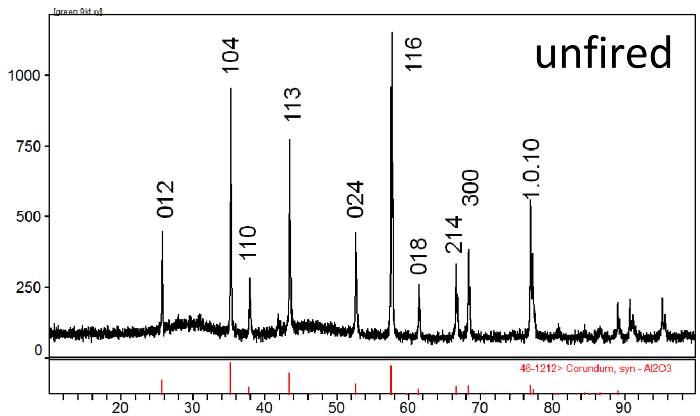
- Heating rate ≥ 5 ºC/min is necessary
- Sintering time is 30 min

Densification vs Crystallization – criticality of process parameter

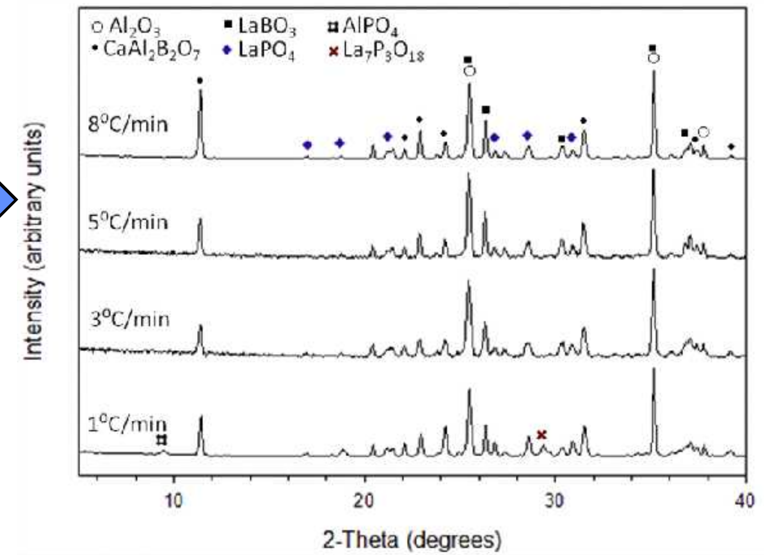


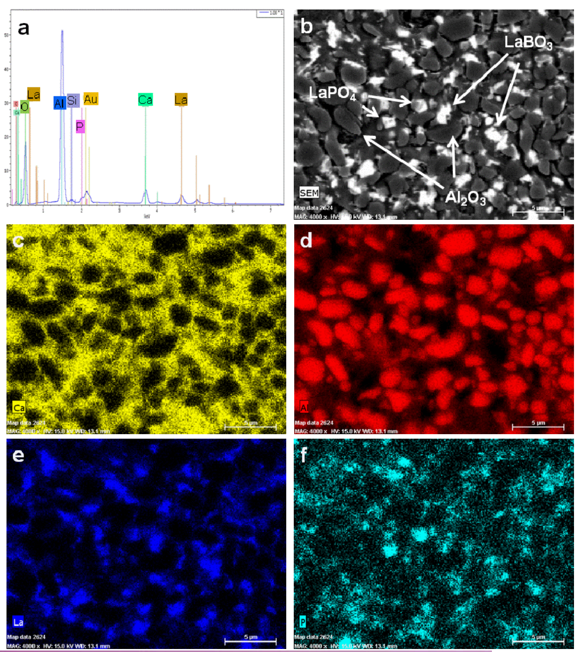
Crystallization in DuPont Low Loss 9k7 LTCC

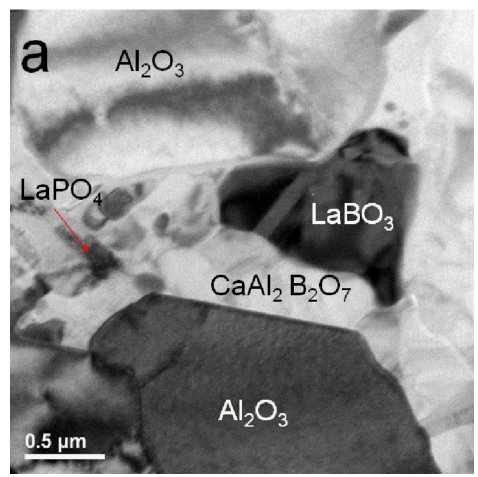
both self- and reaction- derived

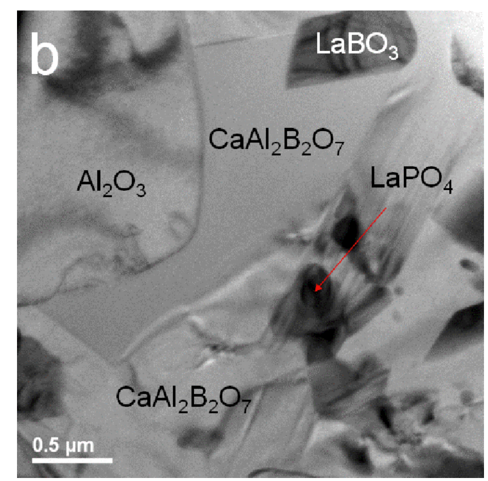


sintering

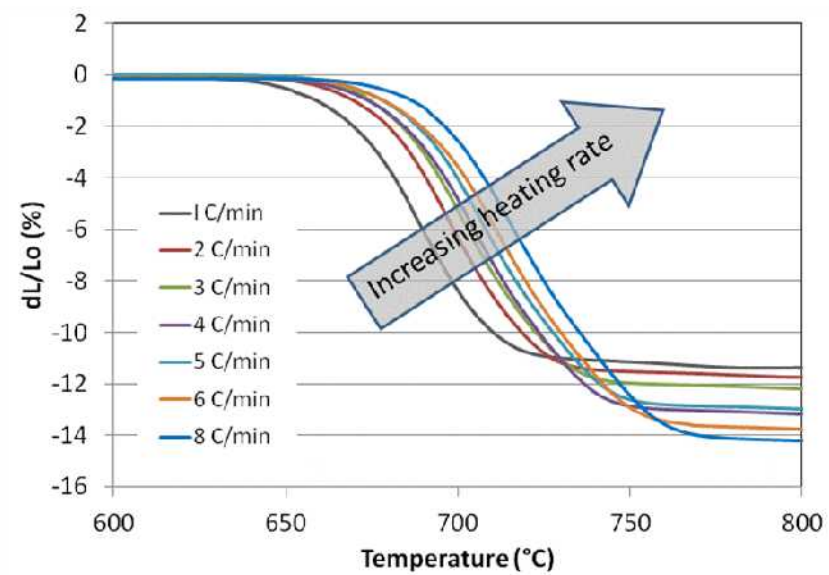


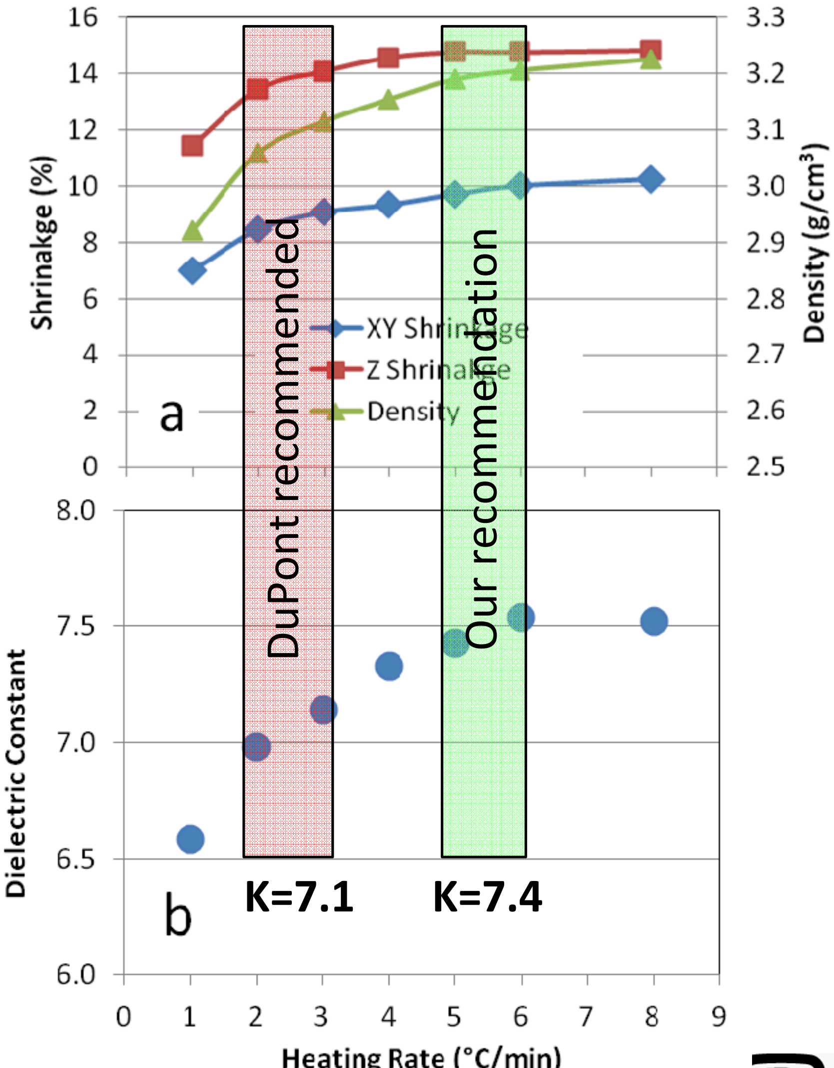






Densification and Process Window of 9k7





Steve Dai, "Densification and Crystallization in Crystallizable Low Temperature Co-fired Ceramics", J Mater Sci, 47, 4579–4584, 2012

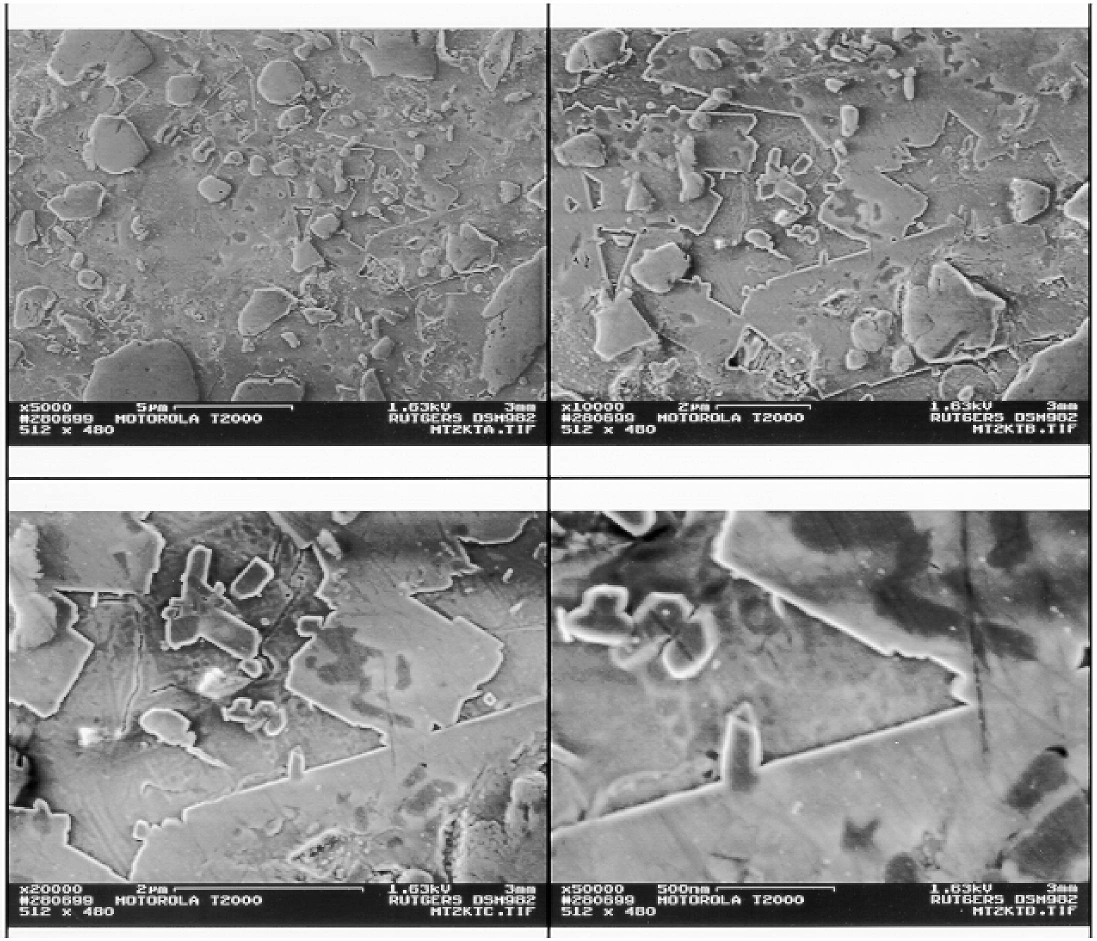
Data to KCP for proper process of 9k7 LTCC

Summary of Global $0\tau_f$

- $0\tau_f$ achieved by a minimal amount of TiO_2 with a combination of
  - Residual TiO_2 particles
  - Ti-rich crystalline phases (titanates)
  - Nano titanates and/or Ti in amorphous phase
  - The dissolved Ti is more effective in τ_f adjustment

- High Q is achieved
  - Engineered LTCC formulation
    - → self-derived and/or reaction derived crystallization
  - A heating rate allows densification followed by crystallization is essential for sintering

Morphology By FESEM



- Glass matrix
- Al_2O_3 particles, 0.5 μm to 3.0 μm
- Wedge-shaped crystalline phase

Properties of compensating materials

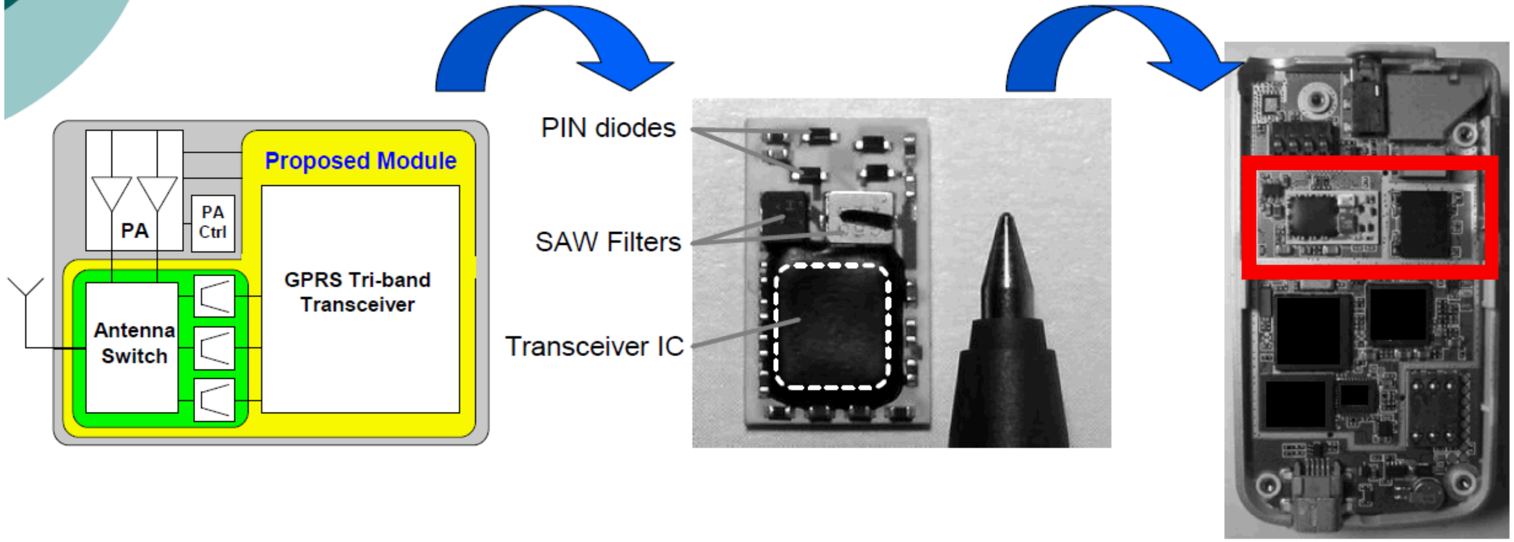
Composition		Temperature (°C)						τ_ε (ppm/°C)
		-50	-20	0	20	50	80	
Dielectric constant	951	7.82	7.84	7.86	7.88	7.91	7.94	118
	Base	7.70	7.74	7.76	7.79	7.83	7.89	190
	STO10	9.58	9.57	9.57	9.57	9.57	9.59	10
	STO20	12.49	12.35	12.28	12.22	12.15	12.10	-240
	STO25	14.34	14.12	13.99	13.88	13.75	13.66	-379
	STO30	16.46	16.10	15.92	15.76	15.57	15.39	-525

- τ_ε changes from positive to negative as STO increases
- STO20, STO25 and STO30 are promising for τ_f compensation. STO10 not considered
- Dielectric constant increases as STO increases → lower f_r

LTCC for RF and MW applications

Design Example:
RF Frontend Module in LTCC

- LTCC transceiver module for Tri-band GPRS mobile phone application:



•Y.-S. Lin, C.-C. Liu, K.-M. Li, and C. H. Chen., "Design of an LTCC tri-band transceiver module for GPRS mobile applications," IEEE Trans. Microwave Theory Tech., vol. 52, pp. 2718-2723, Dec. 2004..