Microstructure and microwave properties of low temperature sintered BST ($Ba_{0.6}Sr_{0.4}TiO_3$) thick-films and their applicability to co-firing processes

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Outline

- **Introduction**

  - $Ba_{1-x}Sr_xTiO_3$
  - Microwave components

- **Low temperature sintered BST thick-films**

  - Experimental route
  - Additive system
  - Microstructure and phase content

- **Dielectric characterisation**

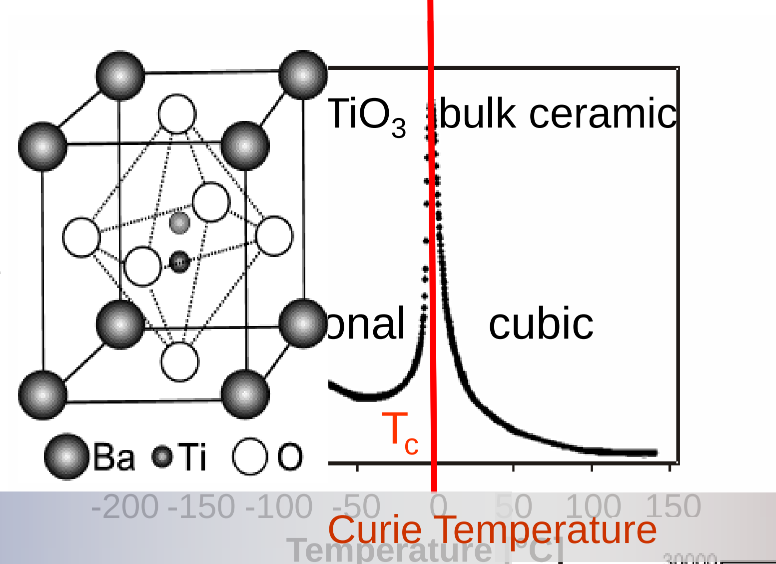
  - Co-Firing of MIM structures
  - CPW vs. MIM

- **Summary and Outlook**

System $Ba_{1-x}Sr_xTiO_3$ (BST)

Ferroelectric Phase | Paraelectric Phase

Tetragonal
Crystal Structure



Cubic
Crystal Structure

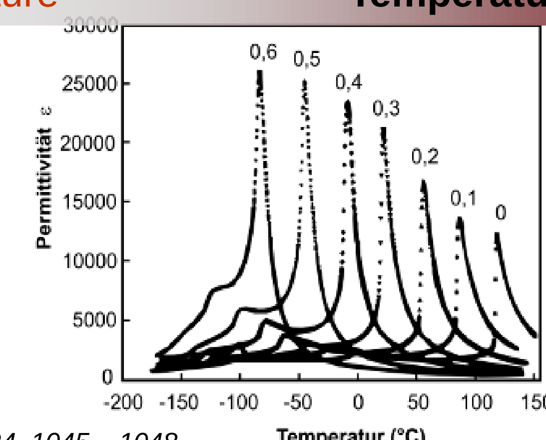
T_c

Curie Temperature

Temperatur

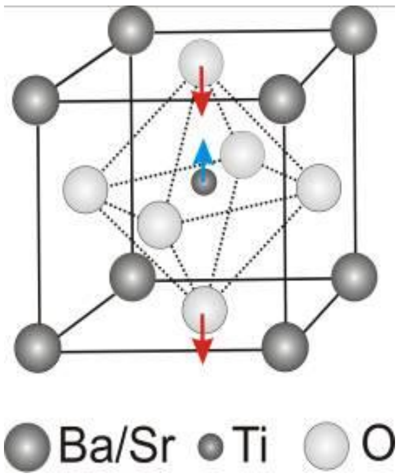
<u>For microwave applications:</u>

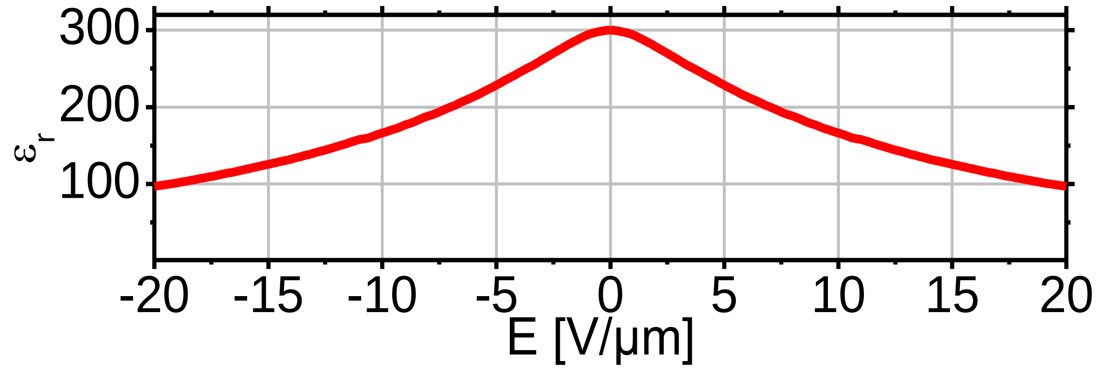
- usage of paraelectric phase
- application at RT → $Ba_{0.6}Sr_{0.4}TiO_3$



Jeon, J., *J. Eur. Ceram. Soc., 2004, 24, 1045 – 1048*

$Ba_{0.6}Sr_{0.4}TiO_3$ as tuneable dielectric material





BST shows a non-linear dependancy of the permittivity on a static E-field

Displacement of Ti^{4+}-ion through an external electric field

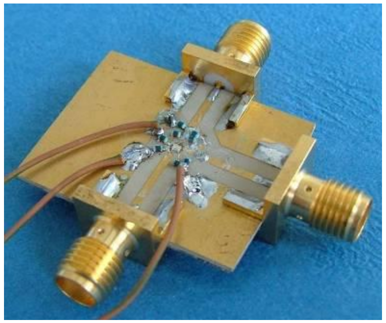
- *no power consumption*
- *high linearity*
- *fast tuning speed*

Dielectric tunability

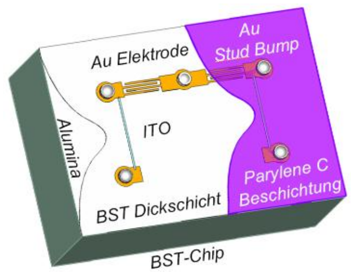
$$\tau_\varepsilon(E) = \frac{\varepsilon_r(E=0) - \varepsilon_r(E)}{\varepsilon_r(E=0)}$$

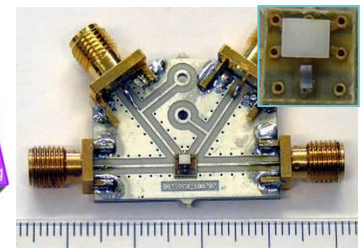
Microwave components based on BST thick-films

Tunable filter



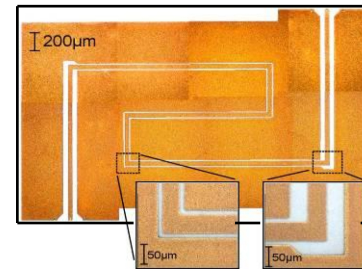
Tunable matching network



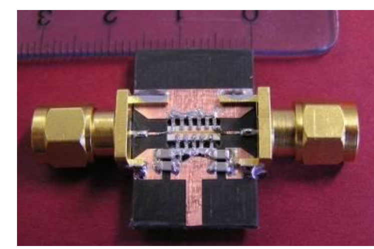


Phase shifter

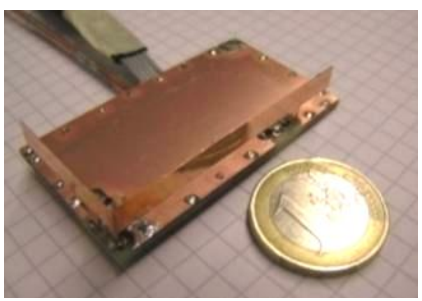
CPW based phase shifter



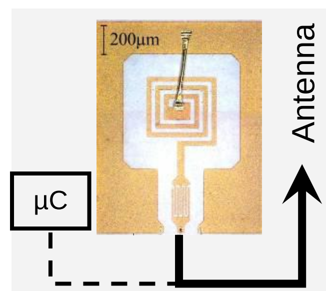
Left-handed phase shifter



Multiband antenna



RF-ID modulator



Microwave Engineering, Technical University Darmstadt

http://www.mwe.tu-darmstadt.de/de/fachgebiete/mikrowellentechnik/forschung/ferroelectrics/ferroelectrics.html

Motivation

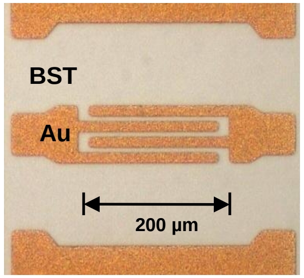
Restriction: High sintering temperature of BST thick film (~1200°C)

- co-firing with silver/gold electrodes not possible (melting point of silver = 962°C)
- co-fired MIM devices only feasible by using high temperature fireable electrodes (e.g. Pt)
- not compatible with LTCC technology (firing range 865-900°C)

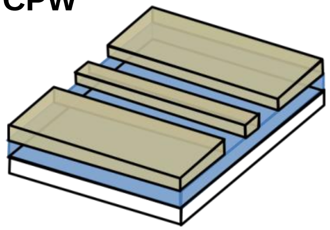
BST varactors

- so far: fabrication of electrodes **after** firing of BST thick-films via fotolithography
- coplanar varactors (IDC, CPW)

IDC

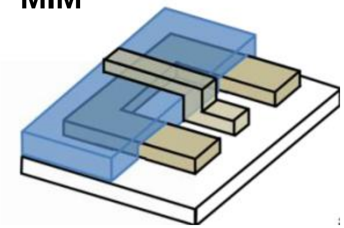


CPW



BST Ag Al_2O_3

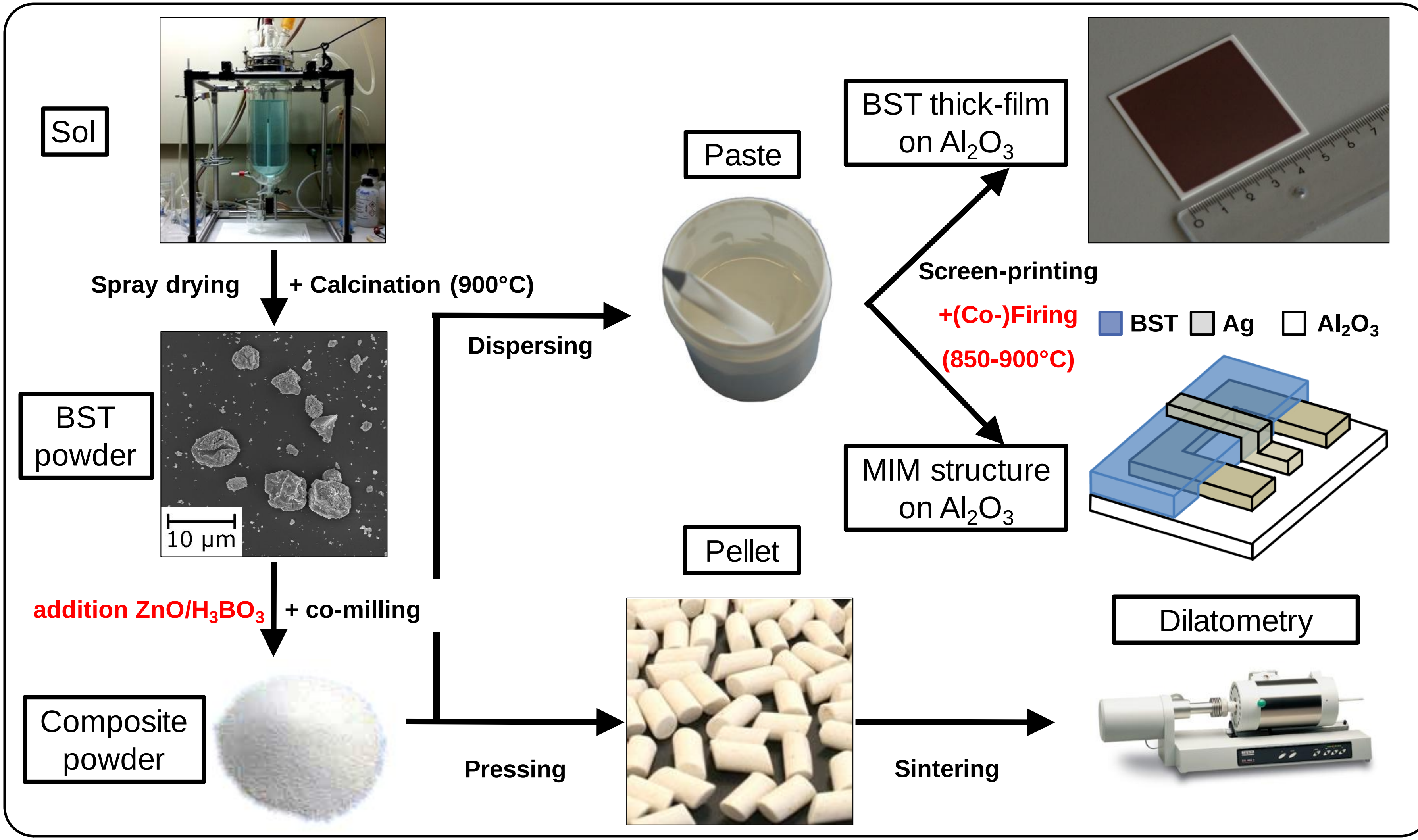
MIM



Objectives

→ development of low-temperature sintered BST-thick films
→ fabrication of co-fired MIM devices

Experimental route



Choice of additive system

Requirement additive system

- lowering of sintering temperature of BST
- no or limited formation of secondary phases
- low permittivity and dielectric loss

→ $ZnO-B_2O_3$ (molar ratio1:1)

(ε_r = 6.9, tan δ = 9.4×10^{-3} (16 GHz), Surendran 2004)

→mixing with BST via co-milling

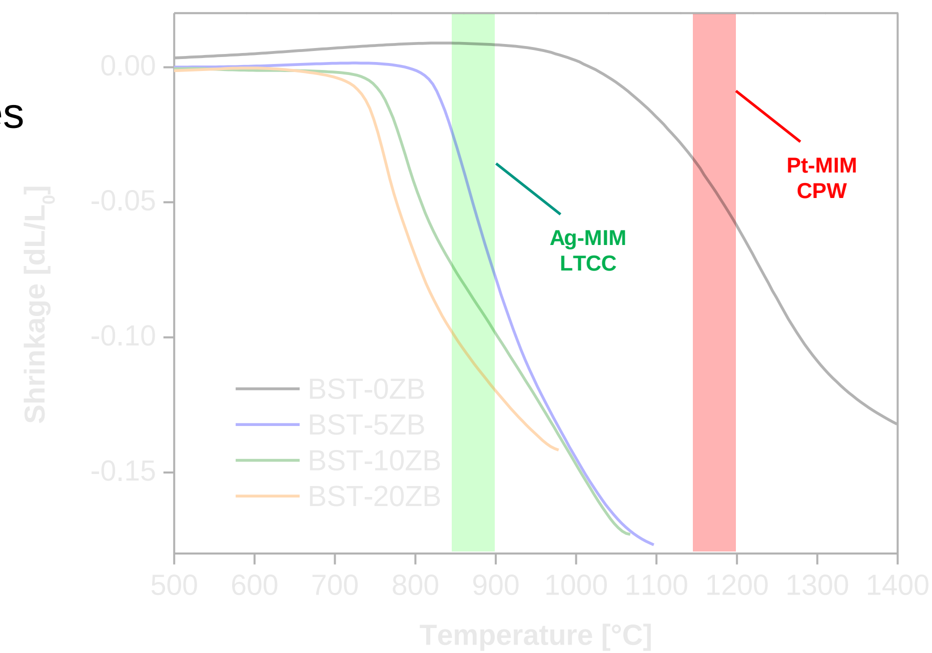
Samples

1.) **BST-5ZB** (5 vol.% $ZnO-H_3BO_3$)

2.) **BST-10ZB** (10 vol.% additive)

3.) **BST-20ZB** (20 vol.% additive)

Dilatometry

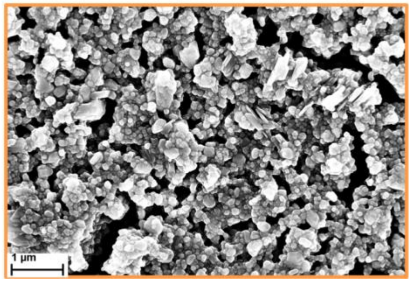


→ Reducing of sintering temperature

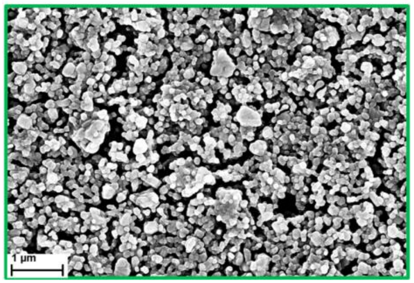
(further details: Kohler et al., IJAC 2013, doi: 10.1111/ijac.12116)

Microstructure & phase content of thick films

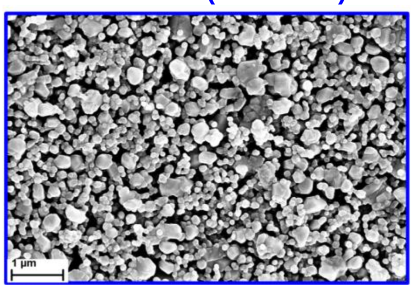
BST-20ZB (Porosity P = 34.8%)



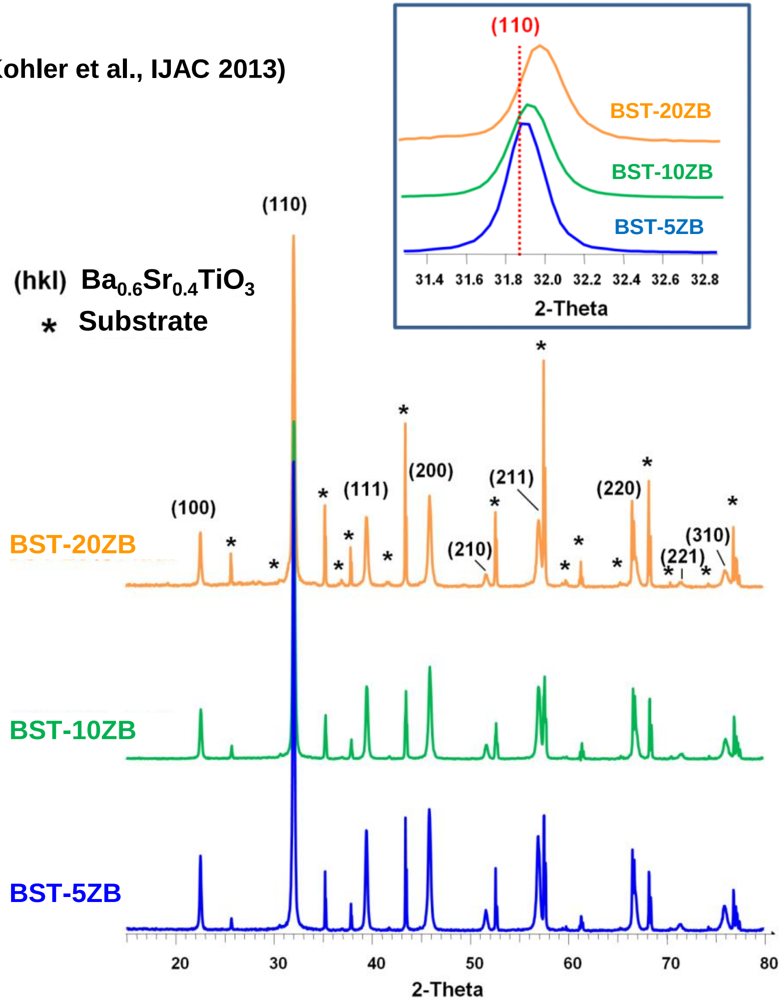
BST-10ZB (P = 41.5%)



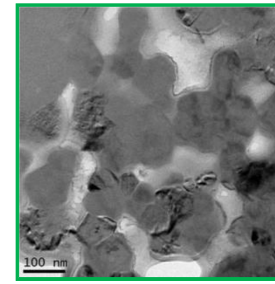
BST-5ZB (P = 45.0%)



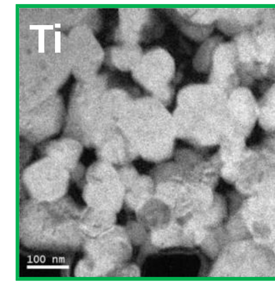
(Kohler et al., IJAC 2013)



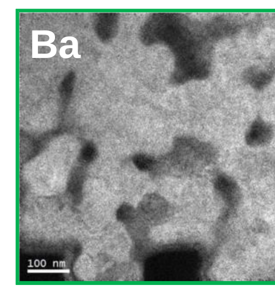
TEM BST-10ZB



Distribution Titanium



Distribution Barium



<u>Higher additive amount</u>

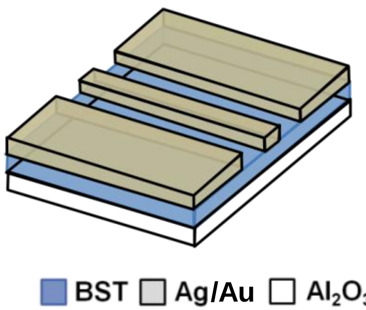
→ lower grain sizes
→ clustering of particles

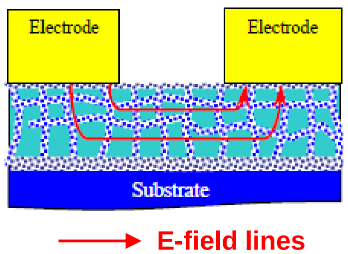
→ no significant formation of (crystalline) secondary phases
→ shift of BST main (110) reflex (**$Ba_{0.6}Sr_{0.4}TiO_3$→ $Ba_{0.5}Sr_{0.5}TiO_3$**)

→ dissolution of barium in amorphous phase

Dielectric characterization – CPW vs. MIM

Concept coplanar waveguides (CPW)



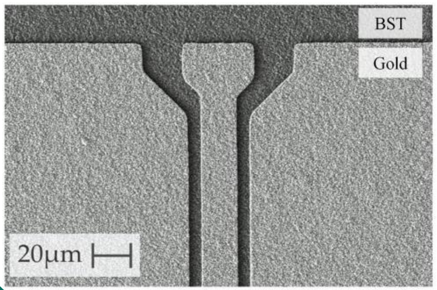


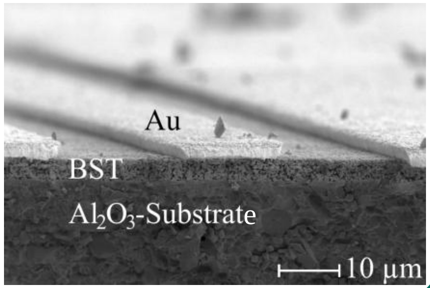
(Gevorgian et al., 2009)

Fabrication

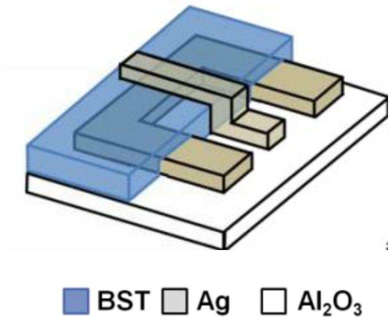
1. Screen-printing BST-ZB paste on Al_2O_3
2. Drying and firing (**900°C/1h**) of thick-films
3. Fotolithography of gold electrodes

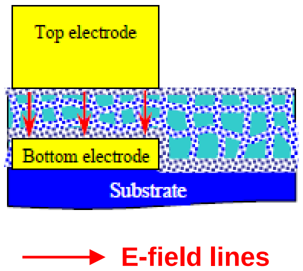
Realized Structures





Concept Metal-Insulator-Metal (MIM) structure



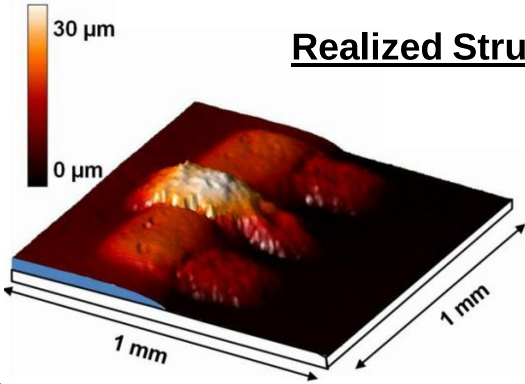


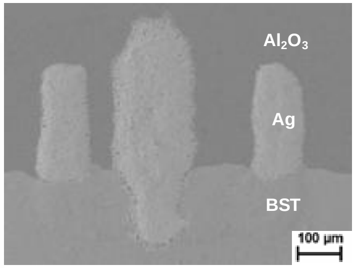
(Gevorgian et al., 2009)

Fabrication

1. Screen-printing of silver electrodes as well as BST-ZB pastes
2. Drying and **co-firing (865°C/1h)** of multilayer

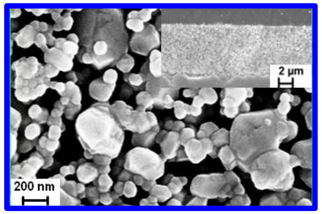
Realized Structures



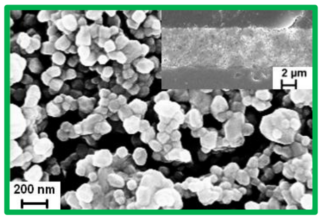


Dielectric characterization - CPW

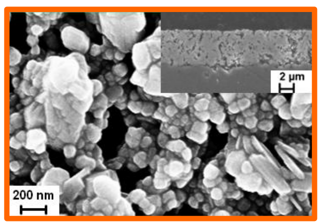
BST-5ZB

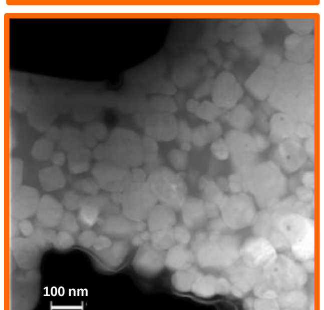


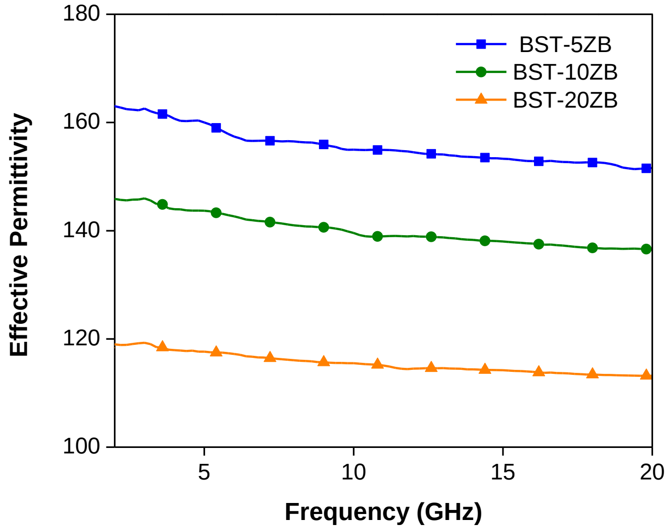
BST-10ZB

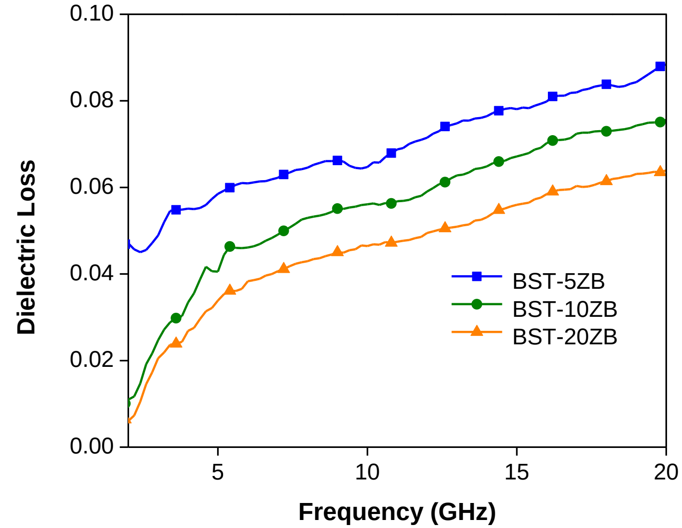


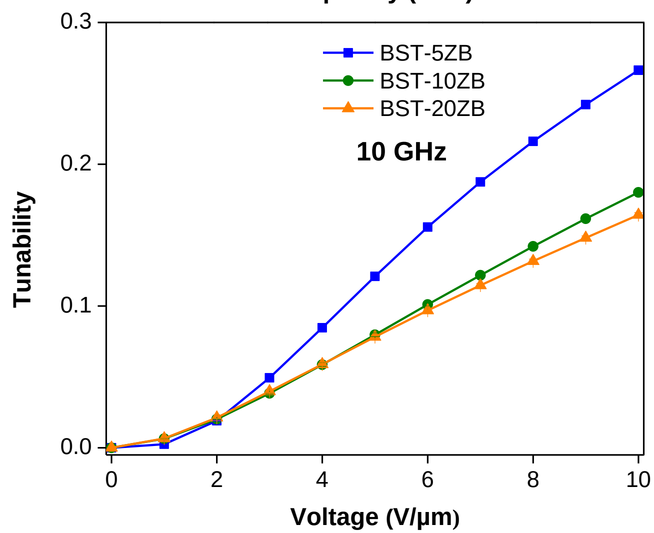
BST-20ZB









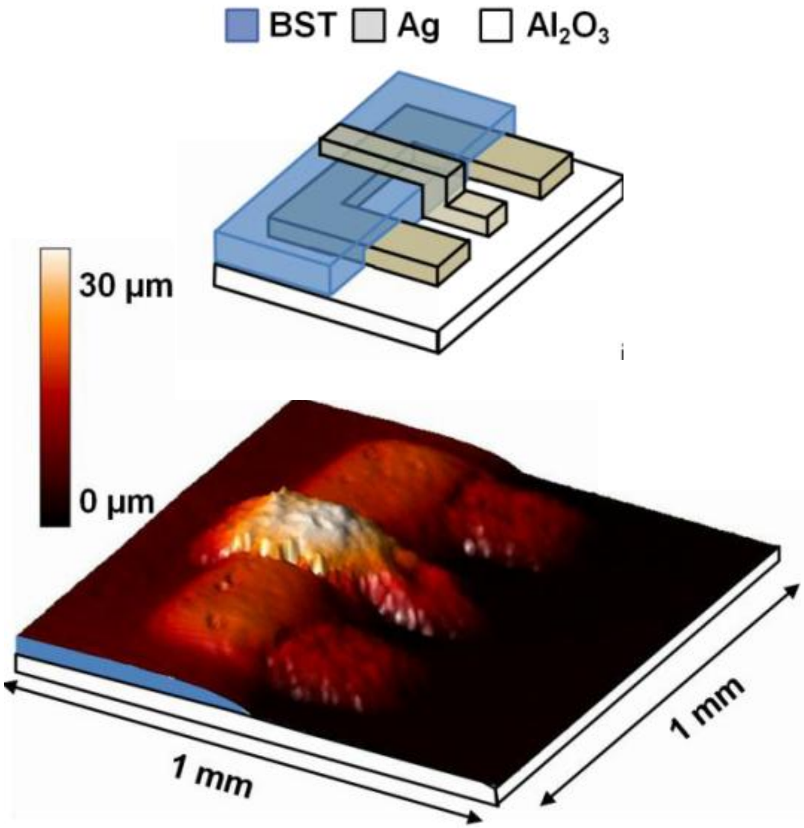


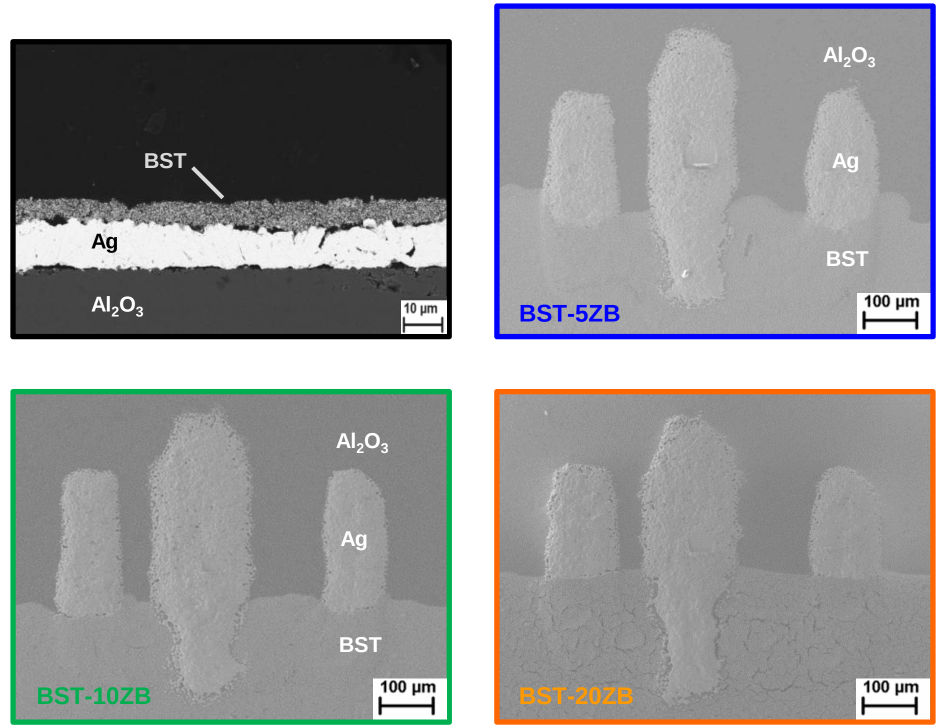
→ decrease of **ε**, **tan δ**, **τ** with increasing additive amount

<u>Due to microstructure</u>

- lower grain sizes
- clustering of particles
- higher content amorphous phase
- change in ratio of Ba:Sr

Fabricated MIM structures (co-fired at 865°C/1h)





+ good adhesion between layers
+ no reaction (proved by XRD, REM)
+ no infiltration of Ag in porous BST
+ dielectric characterization possible

- quality of silver electrodes (edges, roughness)
- cracks in BST-20ZB thick-film

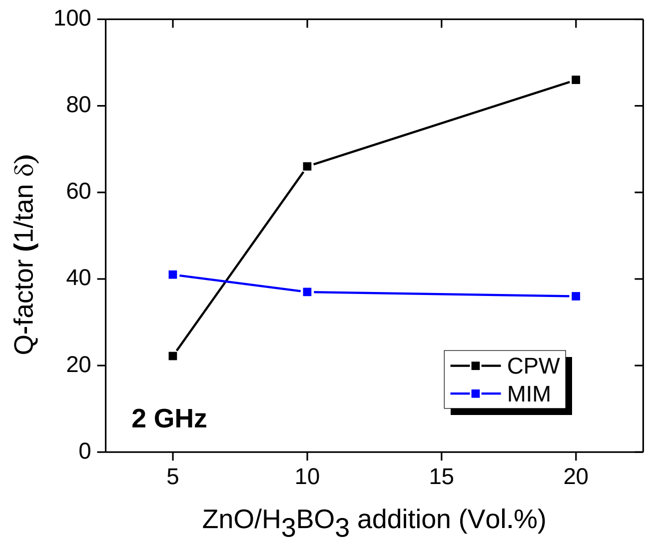
Dielectric characterization – CPW vs. MIM (2GHz)

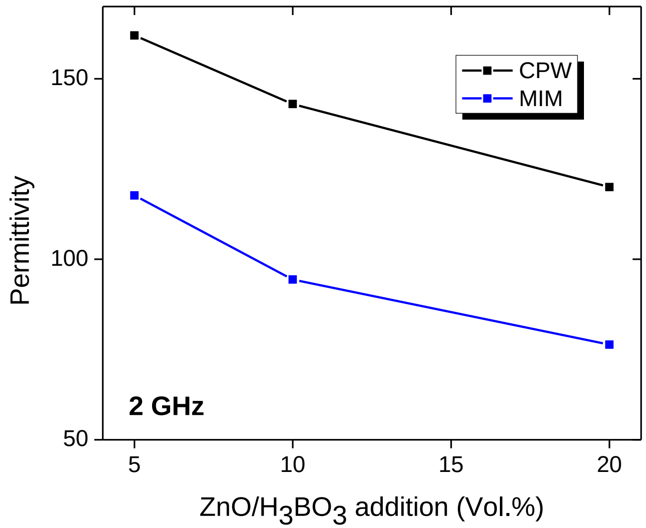
<u>**CPW vs. MIM**</u>

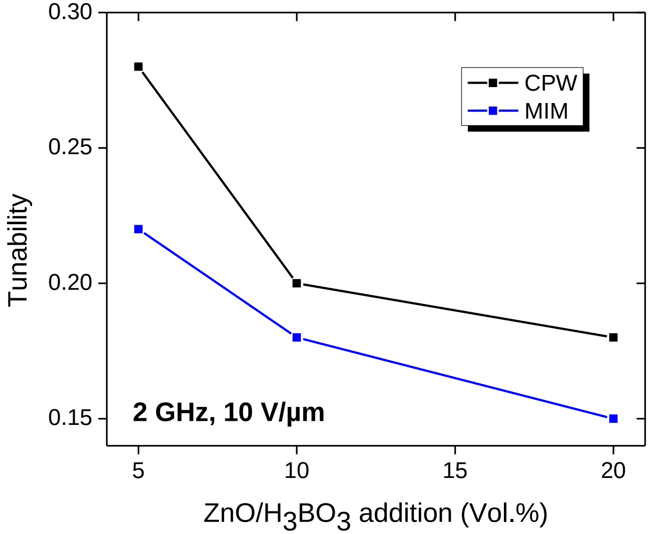
→ similar trends for permittivity and tunability

→ different for Q-values
(due to loss of rough electrodes for MIM)

→ values of tunability for MIM lower than
expected (printing quality has to be optimized)







Summary and Outlook

- **material system BST-ZnO/B_2O_3**

  - lowering of sintering temperature achieved

- **low temperature sintered BST thick films**

  - dependancy of additive amount on the microstructure and phase content

    → grain sizes

    → clustering of particles

    → content of amorphous phase

    → Ba-Sr ratio

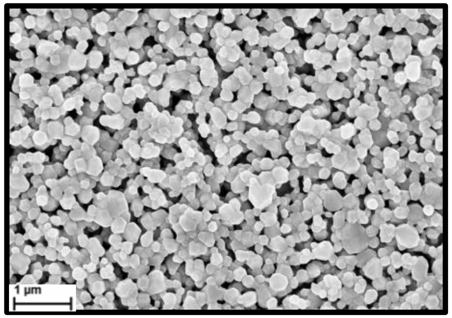
- **Co-firing of MIM structures**

  - good adhesion and compatibility of multilayers

- **Outlook**

  - optimizing printing quality → fully printed RF component

  - usage of CuF-coped BST with ZnO/B_2O_3



CuF BST-5ZB (τ = 45%, 80V, 2GHz)

Thank you for your kind attention!