

Article

A Bio-Sourced Low-Temperature Cofired Ceramic: First Results

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Abstract

This study presents an initial effort to develop Low-Temperature Cofired Ceramics (LTCC) using local bio-sourced materials: Saint Jacques shells and slate tiles. Chemical analysis confirms that the Saint Jacques shells provide CaO and CaCO₃, while slate supplies the needed SiO₂ and Al₂O₃. The constituents, processed from a formulation targeting 70 wt% slate and 20 wt% shell fragments are crushed and ball-milled, mixed with 10 wt% boron trioxide (B₂O₃), and calcinated at 700 °C for two hours to remove organics, followed by a second milling. An aqueous slurry is then prepared and manually tape-cast to form tapes that are processed through standard LTCC process steps. Initial green-state mechanical tests showed elongation values up to ~7.8% and tensile break forces in the range of ~0.5–1.0 N, with lamination performed successfully using standard isostatic conditions. Cofiring yielded limited lateral shrinkage (~6%) but substantial vertical shrinkage (27%). Two-line method measurements indicate a relative permittivity of approximately 4.3 with a comparatively high loss tangent of 0.03, suggesting a vitreous phase and/or porous, inhomogeneous microstructure. A final resonator prototype is fabricated, yielding somewhat encouraging results for the feasibility of this bio-sourced LTCC route while highlighting the need to reduce dielectric losses in future work.

Keywords: Low-Temperature Cofired Ceramics (LTCC); aqueous tape casting; bio-sourced; environmentally friendly; sustainable electronics

1. Introduction

Low-Temperature Cofired Ceramics (LTCC) is a well-known family of materials used in microelectronic devices across various domains, including telecommunications, space technology, healthcare, microwave technology, radar systems, sensors, and microfluidic systems.

LTCC tapes are created by a process called tape casting, where a slurry is created by mixing ceramic powder with solvents, dispersants, binders and plasticizers. In order to maintain a high quality and high volume production, the solvents are most of the time fast-evaporating, such as methyl-ethyl-ketone, xylenes, toluene or acetone, and the binders are vinyl-based, acrylics or other products issued from the petrochemical industry [1]. Environmental and health concerns have raised an interest in aqueous tape casting as proposed in several scientific publications such as [2] treating Al₂O₃-BBSZ materials, [3] where BaTiO₃ films are cast for multilayered capacitors, and in [4] where Borosilicate glass combined with Al₂O₃ is proposed for LTCC. The solvent used in aqueous tape casting is, as the name suggests, water which is non-hazardous to health and environment. As such, a change from solvent-based to aqueous tape casting would contribute to goal 12: responsible consumption and production, which is one of the 17 sustainable development goals adopted by all United Nations member states in 2015.



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In 2023, Zhou et al. presented work on environmentally friendly LTCC [5]. This work discusses the production of a powder made from recycled soda-lime glass combined with naturally abundant volcanic ash. The soda-lime glass reduces the sintering temperature, and the volcanic ash doped with 2.5 wt% of Ba²⁺ enables a material with a low dielectric constant of 4.86 at 1 MHz, a loss tangent of 6.32×10^{-3} , a TCE of 8.72 ppm/°C, and a thermal conductivity of 1.04 W/(m·K). In this study, cylindrical pellets were created by uniaxial lamination and no tape casting was performed. The material composition of waste glass and volcanic ashes is given in their publication; see Table 1.

Table 1. Chemical compositions of waste glass and volcanic ash (wt%), from [5].

Waste Material	SiO ₂	Al ₂ O ₃	CaO	MgO	Na ₂ O	K ₂ O	Fe ₂ O ₃	Other
Waste glass	72.63	2.09	9.49	1.89	11.26	0.83	0.78	0.58
Volcanic ash	63.18	20.12	3.58	1.39	3.6	3.27	3.64	1.22

Soda-lime glass can also be produced from other waste products, such as foundry sands and oyster shells as proposed by [6]. In [6], the characteristics of these two materials collected from a metal casting foundry and from oyster production (pearls and food) were analyzed. Two main compounds needed to produce soda-lime glass were found: silicon and calcium.

One well-known commercially available LTCC tape is the A6M-E from Vibrantz Technologies, Houston, TX, USA [7]. This material has a relative permittivity of approximately 5.7 ± 0.15 and a loss tangent inferior of 0.0015 at 10 GHz [8]. An analysis of the main constituents of an earlier version of this material called A-6, (Ferro Corp, Cleveland, Ohio) can be found in [9], where they are given to be calcium oxide (CaO), silicon dioxide (SiO₂) and boron trioxide (B₂O₃). From [10] A6, DuPont 951, DuPont 943 Heraeus CT800, Heraeus CT700 and Motorola T2000 (commercially known as Heraeus CT2000) after-firing atomic% composition from Energy-Dispersive X-ray spectroscopy (EDX) analysis is presented. These data are given in Table 2.

Table 2. Chemical compositions of well-known commercial LTCC materials after firing (at%) [10].

Elements	951	943	A6	CT800	CT700	T2000/ CT2000
Al	42	76	1.5	41	31	52
Si	47	4	48	39	42	28
Ca	7	9	47	2	5.5	8.5
K	1	0.5		1	1.0	
Mg			1.5	3.5	3.5	
Zn				0.5	0.5	2.5
Ti		7.5		5.5	6.5	6.0
Co	0.5	0.5		0.5	1.0	
Ba				3.5	5.0	0.5

This information gives us the starting point for our creation of bio-sourced ceramic powder to be used with aqueous tape casting. Hence, the main constituents in commercially available LTCC materials are SiO₂ and Al₂O₃ or CaO (or Ca₂O₃).

2. Materials and Methods

Inspired by the information in [5,6] and by the composition presented for the LTCC tape in [9,10], we aim at creating our own LTCC material based on bio-sourced materials available locally. As well as oyster shells, other seashells are known to be similar in

composition, containing mainly calcium oxide (CaO) and calcium carbonate (CaCO₃). This led us to analyze Saint Jacques shells that are abundantly available at the French Brittany coast. Samples from the external shell as well as the internal mother-of-pearl were tested (Figure 1). The samples were prepared by removing all organic materials, washing and air drying them.

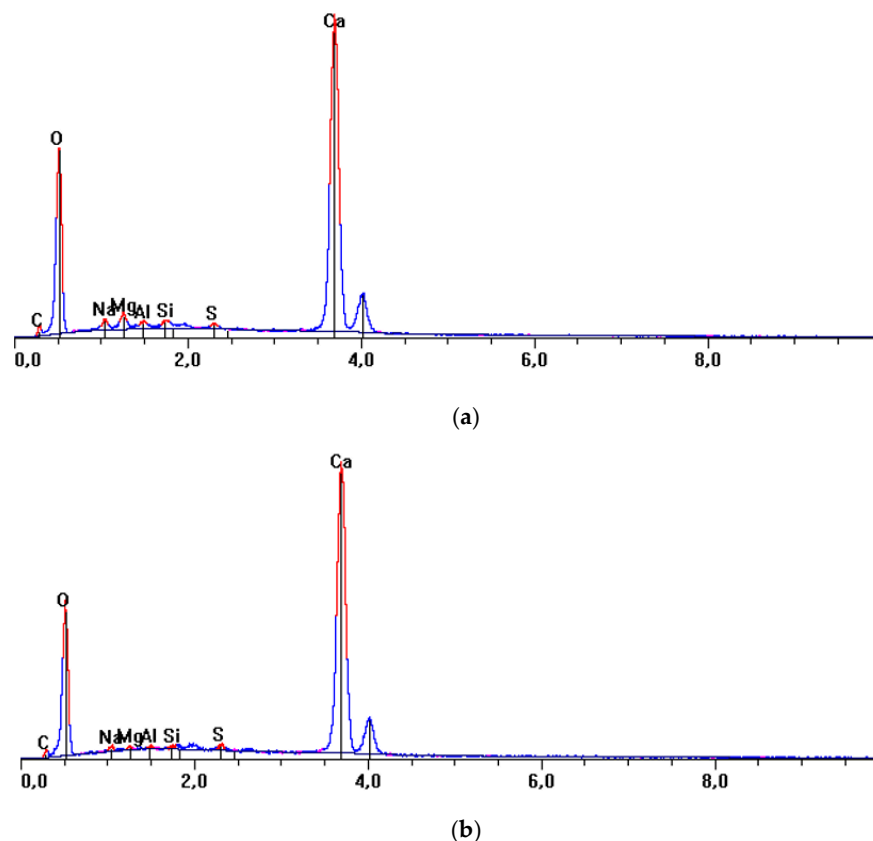


Figure 1. EDX material analysis of Saint Jacques shell exterior part (collected at the French Brittany coast): (a) external shell, (b) mother of pearl. The x-axis unit is keV.

The elemental analysis, (SEM-EDX from Princeton Gamma-Tech Inc., Rocky Hill, NJ, USA) confirms the expected composition of the material. The two samples of the Saint Jacques shell contain the same chemical components (Table 3) even though there is some variation in weight percentage. While both samples have a high content of calcium oxide (CaO) and calcium carbonate (CaCO₃), the outer part of the shell contains higher amounts of sodium oxide (Na₂O), magnesium oxide (MgO) and aluminum oxide (Al₂O₃) than the inner mother-of-pearl sample, which has a higher content of carbon dioxide (CO₂). Upon comparison with the constituents of Table 1, it is evident that the Saint Jacques shell can serve as a bio-source to provide some of the materials needed, particularly calcium oxide (CaO) and calcium carbonate (CaCO₃). From [5], we know that introducing small amounts (2.5 wt%) of Mg, Ca, Ba or B₂O₃ into the material lowers the dielectric constant and the dielectric losses, which is why the presence of MgO and CaCO₃ in the Saint Jacques shells is advantageous.

Table 3. Chemical compositions of Saint Jacques shells (wt%).

Shell Part	Ca	O	Na	Mg	Al	Si	C	S
External shell	14.04	81.88	0.82	0.74	0.34	0.32	1.66	0.18
Mother-of-pearl	9.21	85.10	0.27	0.09	0.07	0.08	5.01	0.17

To advance further, following [9,10], silicone dioxide (SiO_2) is also required. It is a well-known material used in glass and in ceramic materials, where it forms the vitreous phase. Although the Saint Jacques shell contains some SiO_2 , the quantity is insufficient for the creation of our LTCC material.

However, SiO_2 is abundant in quartz, sand and agate. Our second bio-sourced candidate, slate, is made up of quartz, mica and feldspathoid. This material is used for roof tiles throughout the French Brittany region. The local quarries have been closed for some years, so slate tiles are now imported, mainly from Spain and Portugal. Nevertheless, slate tiles need to be replaced every 50–80 years, so there is an abundant supply of waste slate due to the renovation and isolation of buildings. The elemental composition of slate has been obtained from EDX analysis (Figure 2 and Table 4).

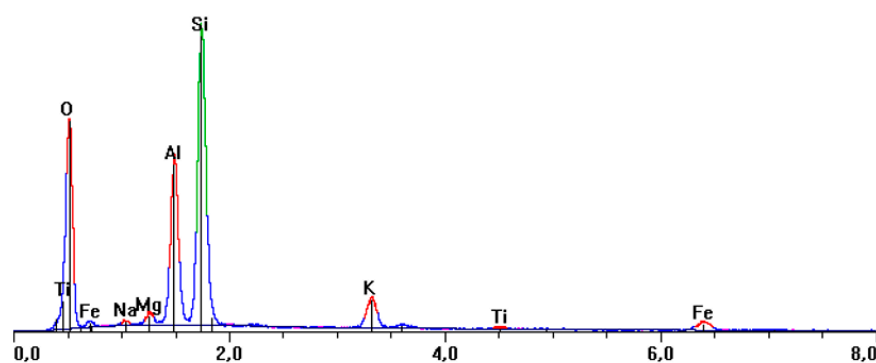


Figure 2. EDX analysis of slate material. The x-axis unit is keV.

Table 4. Chemical composition of slate material (wt%).

Si	Al	K	Fe	Na	Ti	Mg	O
24.81	11.63	3.41	2.14	0.60	0.34	0.95	56.11

Clearly, the slate mainly contains silicon dioxide (SiO_2) and aluminum oxide (Al_2O_3), as well as smaller amounts of potassium oxide (K_2O), iron oxide (Fe_2O_3 or FeO), magnesium oxide (MgO), sodium oxide (Na_2O) and titanium oxide (TiO_2). It is the main constituent, SiO_2 , that we need to form the ceramic material. The relatively high presence of Al_2O_3 is also positive. The same goes for titanium oxide and the presence of magnesium should also be beneficial (as it lowers the dielectric constant and losses).

The third main constituent from [9] is the boron trioxide (B_2O_3). This element was not detected in [10]. Nevertheless, this compound can be useful for LTCC compositions as it lowers the sintering temperature. This same effect is achieved by calcium trioxide when it releases carbon dioxide (CO_2) and transforms into calcium oxide (CaO) during firing. Boron trioxide occurs naturally in the form of borax, colemanite and ulexite. As it is not a material that can easily be found in bio-sourced or waste materials, we decided to buy this compound (di-Bore trioxide $\geq 97.5\%$, Thermo Fisher Scientific, Waltham, MA, USA).

From [10], a typical LTCC powder contains silica (SiO_2), alumina (Al_2O_3), calcium carbonate (CaCO_3), and calcium oxide (CaO), as well as additives. Each component plays a specific role in the formulation: SiO_2 allows the formation of the necessary glass phase for LTCC while at the same time fragilizing the material's mechanical characteristics. Al_2O_3 is a good electrical insulator that increases mechanical strength and improves thermal stability, CaCO_3 acts as a fluxing agent that lowers the sintering temperature as it decomposes into CaO , which also promotes densification and improves mechanical strength. Additives, such as B_2O_3 , lower the sintering temperature and relative permittivity while improving

mechanical resistance. These oxides have different relative permittivities (ϵ_r), as presented in Table 5.

Table 5. Relative permittivity, ϵ_r , at 1 MHz of oxide compounds [11].

Compound	SiO ₂	Al ₂ O ₃	CaO	B ₂ O ₃	MgO
Relative permittivity	3.79–4.50	10.1	11.8	4.5–5.5	9.65

Currently, we are not targeting a specific value for the relative permittivity. The purpose of this first experiment is to prove that the slate and Saint Jacques shells can be used to create an LTCC material. Based on previous information, our goal is to create a material with the following composition: 60 wt% SiO₂, 10 wt% Al₂O₃, 20 wt% CaO, and 10 wt% B₂O₃. This should be possible by using a mixture of 70 wt% slate, 20 wt% Saint Jacques shell fragments, and 10 wt% B₂O₃ powder. Using the percentages with the dielectric constants in Table 1, we arrive at an estimated low frequency relative permittivity of 3.63.

2.1. Powder Preparation

Fragments prepared by manual breaking of the slate and Saint Jacques shell were first dry-milled separately in a planetary ball mill (Retsch PM100, Retsch GmbH, Haan, Germany) for ten minutes at 400 rpm using ten 15 mm zirconia balls in an 80 mL zirconia bowl. The powders obtained were mixed together with the boron oxide at the intended wt% to form 9 g of powder. The powder was then calcinated at 700 °C for two hours to remove any organic residues from the bio-sourced ingredients. The mixed powder was dry-milled once more in the planetary ball mill with 250 pieces of 5-mm zirconia balls for three minutes at 300 rpm.

2.2. Slurry Preparation and Tape Casting

The slurry was prepared according to the following procedure: First, 9.0 g of powder is placed in a pot. Then, 4.5 g of deionized water, 4.4 g of dispersant 2% (wt), 8.0 g of binder solution 30% (wt), and 0.5 g of foam master are added. The last three products are from Fiaxell SOFC Technologies in Lausanne, Switzerland. Twenty-five zirconia balls were added, and the pot was sealed and rolled at 80 rpm on a Fisherbrand roller bench (from Fisher Scientific) for 24 h. After rolling, the zirconia balls were removed, the pot was resealed, and the slurry was kept at 5 rpm for another two hours to remove any bubbles. Then, the slurry was passed through a nylon cloth (150 µm mesh) to filter out agglomerates and any remaining bubbles. Once filtered, the slurry was cast onto a polypropylene (PP) sheet placed on a tape caster bench glass surface. To improve tape release, the PP sheet's surface was treated in advance with a 50:50 wt% rapeseed oil–isopropanol mixture. The slurry viscosity was 2.9 Pa.s. The tape casting was done manually with a doctor blade set to a height of 0.7 mm and a speed of approximately 150 mm/s. Drying took place during 24 h at room temperature without any additional heating.

3. Results

Several analyses were performed to judge the material's performance in the context of a standard LTCC manufacturing process. Layer thickness, dried density, elongation and tensile break force (TA1, Lloyd Material tester from Amtek, Largo, FL, USA), visual inspection (50X Moticam optical microscope from Motic, Universal City, TX, USA) and surface profiling (White light optical profilometer) were completed as initial tests followed by a complete LTCC prototype production covering laser cutting, screen printing, uniaxial lamination, isostatic lamination and sintering.

3.1. Initial Tests

The layer thickness was measured with a regular micrometer to be $180 \pm 20 \mu\text{m}$. The dried density was found to be 1442 kg/m^3 . Elongation measured at 72 h, 144 h and 268 h were 6.5%, 7.8% and to 7.55%, respectively, while tensile break force, tested at the same occasions, were found to be 0.87 N, 0.51 N and 0.98 N. The elongation values are quite acceptable when compared to commercially available tapes as we have found that the A6M-E elongates to 5%. However, the tensile break force for the A6M-E material is found to be 2.32 N, which is more than double our values.

The visual inspection using a 50X microscope shows that the tape is not homogenous (Figure 3). Evidently the ball milling procedure needs to be improved to make the particles smaller and allow for a homogenous tape and ceramic material.

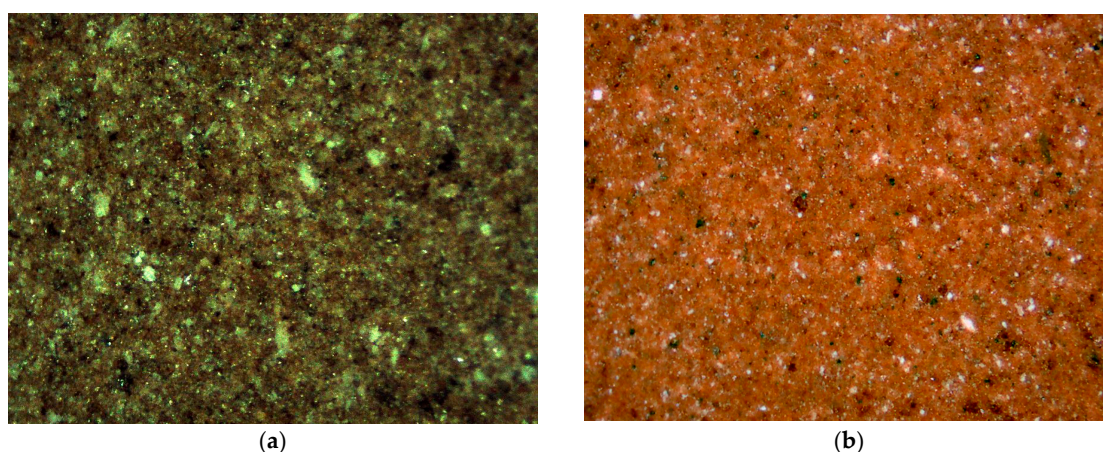


Figure 3. Visual inspection, with 50X magnification, of the material (a) after 24 h of drying and (b) after sintering. The color change is mainly due to light setting in the microscope.

The red/brown color is not usually what you expect for a ceramic material based on SiO_2 , Al_2O_3 and CaO . Most likely the iron oxide contained in the slate generates this color. This agrees with [5], where the volcanic ash contained Fe_2O_3 and all the presented samples have a brown color.

3.2. LTCC Prototyping

LTCC fabrication is usually made by slitting and blanking, via and cavity creation, via filling, layer screen printing, stacking, lamination and cofiring. To analyze our material, we have followed the same procedure, first by making an empty device (no via holes or cavities, no printing) and then by preparing a real functional device.

3.2.1. Ceramic-Only Prototype

The first device was prepared by cutting layers of the tape into $50.8 \text{ mm} \times 50.8 \text{ mm}$ squares with alignment holes present in the corners for stacking. The micromachine equipment is a fibered laser with a 1064 nm wavelength (LEM2 from Laser Cheval, Marnay, France). The successful parameters were pulse duration 20 ns, power 60%, galvanometric speed 100 mm/s and frequency 20 kHz repeating 12 times to cut through the $180 \mu\text{m}$ thick tape.

A stack with eleven layers was prepared for isostatic lamination in the PTC LT04001 (Pacific Trinetics Corporation, San Jose, CA, USA). This sample underwent a lamination at 70°C , 21 MPa with a 5 min soak time followed by 5 min of lamination. As can be seen in Figure 4, the result is positive as the individual layers cannot be perceived.

Finally, the sample was fired using the same profile as is proposed for the A6M-E material with a sintering peak at 850°C for 15 min, see Figure 5. The shrinkage was 6% in the X- and Y-direction and 27% in the Z-direction.

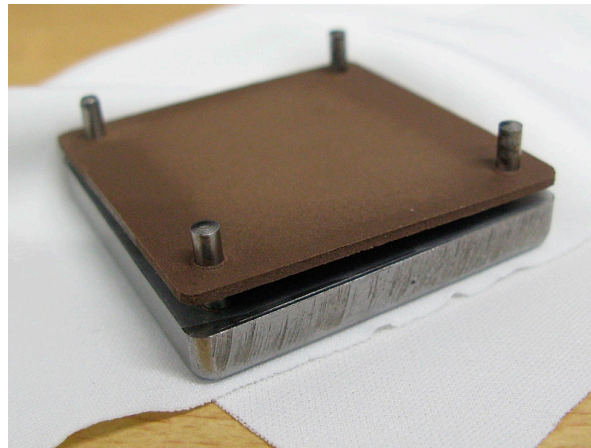


Figure 4. Eleven layers of the bio-sourced tapes after lamination using an isostatic press from PTC.

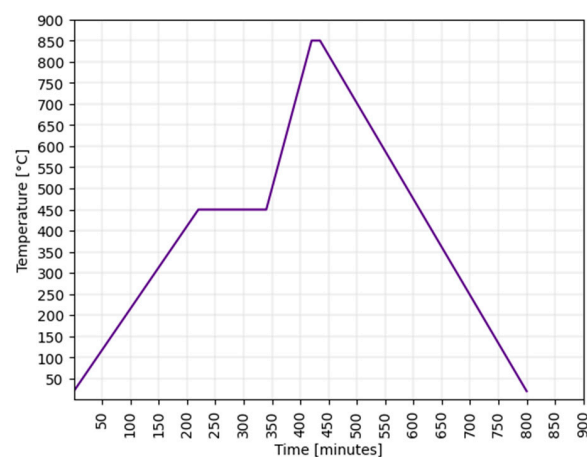


Figure 5. Firing profile for the bio-sourced LTCC tape used with programmable static furnace, P330, from Nabertherm (Nabertherm GmbH, Lilienthal/Bremen, Germany).

The surface profiling, performed using an in-house built white light optical profilometer, indicates that the surface is not smooth as a height variation of 40.7 to 70.1 μm is found (Figure 6).



Figure 6. Surface profiling over a 223 μm long trace. The green lines indicate the maximum and minimum overall values for the test, the blue line indicates the profile at each measured point.

3.2.2. Relative Permittivity Analysis

As discussed before, the relative permittivity is an important characteristic for materials intended for microwave devices. Given our available fabrication and measurement equipment, we have chosen to use the “two microstrip line method” as proposed by [12]. However, we have used it with Coplanar non-grounded Waveguides (CPWs) instead of microstrip lines to make the fabrication easier. The verification of the validity of this model for CPW transmission lines is presented in Appendix A. All that is needed is the creation of two different-length CPWs on the substrate material.

The lines are created by laser ablation on a top layer screen-printed silver with dimensions that should give a line impedance of 50 ohms. L1 is 536 μm , and L2 is 184 μm long,

the central line width, w , is $133\ \mu\text{m}$, and the gaps, s , are $37\ \mu\text{m}$. These measures all have tolerances of $\pm 10\ \mu\text{m}$. The two lines are presented in Figure 7. Their creation is achieved with the use of the fibered laser (LEM2 from Laser Cheval, Marnay, France).

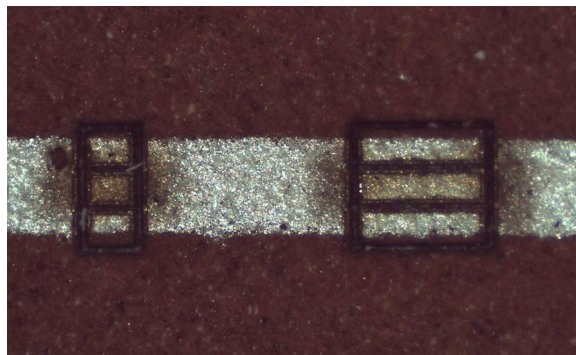


Figure 7. The two CPW lines for the relative permittivity test purpose. The shorter line, L2 (as defined by [12]) is at the left, and the longer line, L1, at the right.

The two lines are measured using a 10 MHz to 67 GHz VNA (ZVA 67 GHz, from Rohde & Schwartz, GmbH, Munchen, Germany), a probe station (WL170 from Signatone Corp, Gilroy, CA, USA) with $150\ \mu\text{m}$ pitch GSG Picoprobes (67A-GSG-150-EDP from GGB Industries Inc., Naples, FL, USA). Before measuring these devices, a short, open, load and thru (SOLT) calibration was performed with the help of a CS5-calibration kit (GGB Industries Inc. Naples, FL, USA), with input power of 0 dBm and an averaging factor equal to 10 by 4001 points. Due to dispersion not being taken into account by the calibration procedure, the CS5 substrate will not allow correct measurement at frequencies below 5 GHz [13].

After measurement, the effective permittivity, ϵ_{eff} , is first obtained following the steps indicated in [12]. The resulting values are presented in Figure 8. Smoothing with a moving average of 10 was applied for better readability. The ϵ_{eff} is found to have a mean value of 2.46 and a standard deviation of 0.20, calculating from 5 GHz and upwards. From this value, an ϵ_r of 4.3 can be estimated using the LineCalc tool from Advanced Design Systems (641.update2.1 29 April 2026, ADS2026 Update2.1, Keysight Technologies, Santa Rosa, CA, USA). In comparison with the relative permittivity estimated from the weight percent ratio to be 3.63, the results seem quite plausible, even though somewhat higher. From the tolerance and roughness analysis performed in Appendix A, this value should be correct within a 10% limit. Nevertheless, the method used gives a first indication, which is valuable.

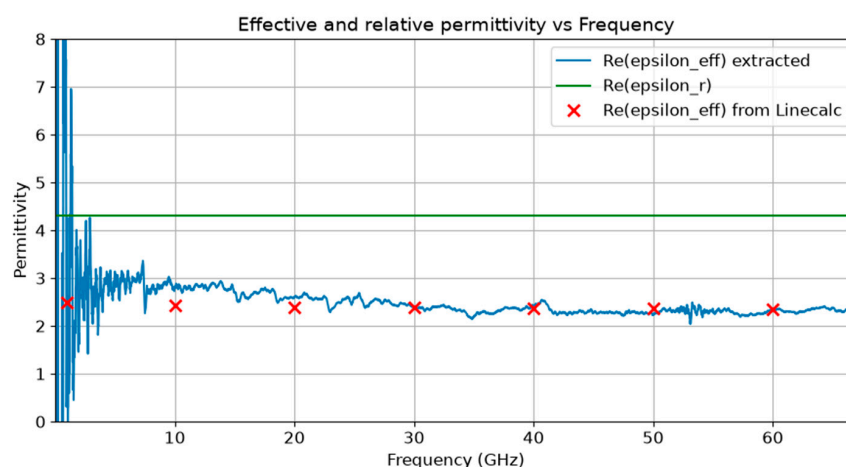


Figure 8. Effective and estimated relative permittivity extracted from S-parameter measurements using the two-line method. A smoothing by a moving average of 10 has been used. The green line indicates the relative permittivity value used for LineCalc and the red crosses the related ϵ_{eff} .

3.2.3. Full Prototype

A second prototype including a microwave ring resonator for test was also fabricated based on tape created from two slurry preparations using basically the same recipe. In this run, added fabrication procedures such as via hole cutting, via filling and screen-printing were performed. The same laser (LEM2 from Laser Cheval, Marnay, France) was used with parameters as set previously. An M2H screen printer from EKRA (Asys group, Dornstadt, Germany) was used together with 325 meshed inox screens (with 18 μm diameter wires in the mesh) for via hole filling and screen printing of the motives. The paste used was Ferro CN33-493 for the via holes and Ferro CN33-498G for the ring resonator and the ground plane.

The via screen printing was performed on the top fourth layer only. The ground plane was positioned on top of the third layer and the ring resonator structure on the top fourth layer, with each layer being 180 μm thick before firing and 130 μm after firing.

After stacking (Figure 9), a lamination nonstick film was placed on the tape before closing the fixture with the top plate. The unit, placed in a vacuum-sealed bag, was isostatically laminated before firing.

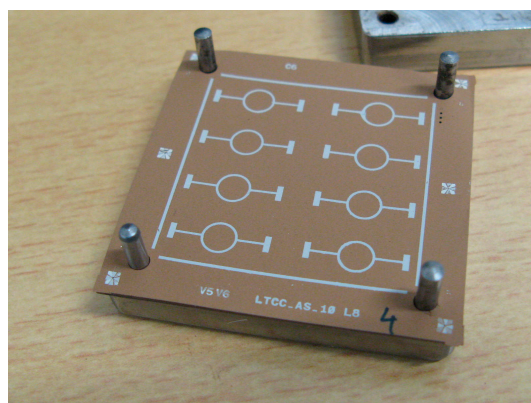


Figure 9. After stacking layer by layer to produce the resonator structure, we finally show the top layer with resonator structures screen-printed with Ag paste.

Before measurement, laser ablation was carried out to create the openings in the coplanar probe pads and the coupling gap between the ring and the microstrip transmission lines; see Figure 10. The created gaps are 30 μm wide, which is too small for screen printing and why this operation is necessary to create the resonators. The laser ablation not only opened up the metal but also created a certain groove in the ceramic structure.

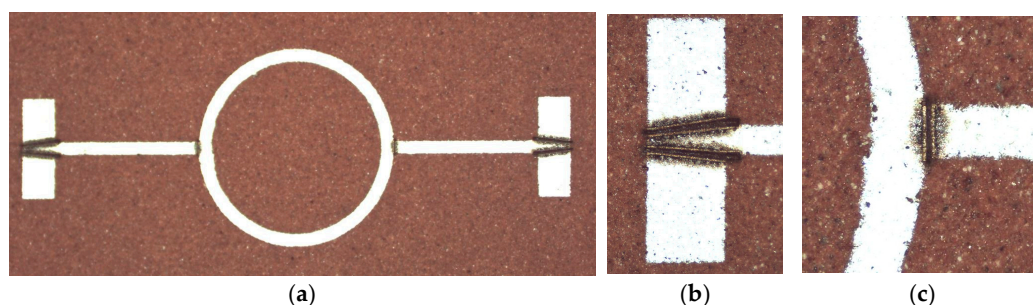


Figure 10. Final resonator after laser ablation: (a) complete device, (b) probe pad and (c) coupling gap.

S-parameter measurements were performed using the 67 GHz VNA (Rohde & Schwartz, GmbH, Munchen, Germany) together with 150 μm pitch GSG probes (67A-GSG-150-EDP from GGB Industries Inc. Naples, FL, USA). SOLT calibration was performed with the help of a CS5-calibration kit (GGB Industries Inc. Naples, FL, USA), using 0 dBm

input power and a 10 MHz step over the 0–30 GHz frequency range, with an averaging factor of 4.

The results of the S21 transmission parameter are presented in Figure 11 together with retro-simulation results. Retro-simulation means creating an electromagnetic simulation correcting dimensional values for what is obtained after firing and tuning relative permittivity and loss tangent until a descent match between simulation results and the measured ones is obtained. The resonant peaks are not very clear from the measurements but are good enough to match quite well with the simulation results where the relative permittivity is set to 4.0. From the retro-simulation, we can also estimate the loss tangent to be approximately 0.03. This value is high compared to commercial LTCC materials and excludes it in its present state from being used for any industrial product.

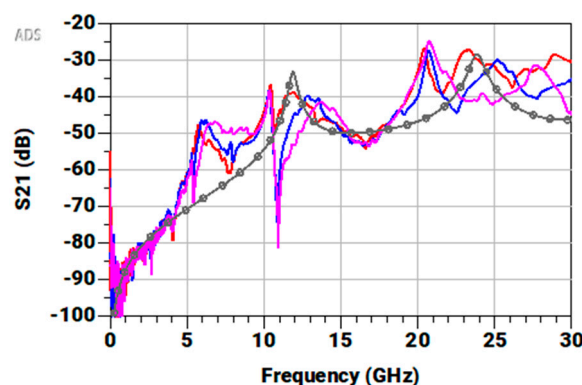


Figure 11. Measured insertion loss in dB for the three samples (red, blue and magenta) and a retro-simulation in grey (-o-).

4. Discussion

This work presents the very first results of a bio-sourced LTCC material created from slate and Saint Jacques shells. The material mix and fabrication procedure led to a ceramic material with some interesting and encouraging features. Slurry preparation and tape casting based on an aqueous solvent was successful, and the obtained tapes were easily processed with standard LTCC fabrication procedures. Compared to the A6M-E material, the tensile break force is low, while the elongation is higher. Even if the break force is low, the tapes were easily handled and required no additional care as compared to commercial materials. Lamination using a standard profile in an isostatic press worked very well. The surface roughness is indeed high, and the grains are visible at 50X visual inspection. This suggests that the grinding of the powder did not diminish the individual grains enough, something that should be better controlled in a future run. The shrinkage of only 6% in a lateral direction is less than the A6M-E material even though a vertical shrinkage of 27% is normal. The small lateral shrinkage may suggest that full densification did not take place during firing. An enlarged study of the sintering profile and thermogravimetric analysis would enhance the information to improve the densification.

Comparing it with the expected relative permittivity of 3.63 calculated from the compounds comprising the material, the relative permittivity value of 4.3 obtained using the two-line method falls within the same range. This value is lower than most available LTCC tape materials, which is advantageous for high-frequency devices given that the dimensions of transmission lines can be relaxed, as the guided wavelength scales with the inversed square root of the effective permittivity. On the other hand, the losses seem to be high, with a $\tan \delta$ of 0.03, which makes this material, in its present form, undesirable for high-frequency devices where dielectric loss should be minimized. Nevertheless, this fact does not exclude the possibility of using this material in other applications. The high losses

suggest that the material contains a large vitreous phase and is porous and inhomogeneous in structure. The B_2O_3 is known to form a vitreous phase, which will increase the relaxation losses, while CaO and MgO tend to modify the structure to create vacancies, interstitials and point defects that favor ionic mobility, which in turn increases the losses.

Even so, the outcome of this first attempt to create a bio-sourced LTCC material based on aqueous tape casting is quite positive, even if there are several improvements to be made.

On the positive side:

1. The chosen bio-sourced materials result in a ceramic material;
2. Aqueous tape casting has been developed with correct results;
3. Laser-cutting procedure work well;
4. Screen printing with commercial LTCC silver paste worked out fine;
5. The tape withstands isostatic lamination using standard parameters;
6. The tensile force and elongation values for the green tape are correct;
7. Sintering resulted in flat prototype samples with limited shrinkage;
8. A low relative permittivity is obtained.

On the negative side:

1. The material seems to be vitreous and porous;
2. The surface roughness is high;
3. The loss tangent is high.

From these results, it may be worth leaving out or at least diminishing the amount of boron trioxide in a future powder creation. The visual inspection and roughness measurement indicates that the grain size is large, which is why the planetary ball mill procedure should be prolonged and better supervised; possibly wet milling would enable a smaller grain size. To better understand the material, grain-size control, EDX final material composition, thermogravimetric analysis, flexibility, elongation, thermal conductivity and relative permittivity measurements at different frequencies may be useful for the next run. Different mixes of the two main components may also be helpful. Further efforts will also focus on developing a fully bio-sourced material including the slurry constituents.

5. Intellectual Protection

The findings in this work are protected by an Envelope Soleau, number DSO2026017417.

Author Contributions: Conceptualization, C.K.; methodology, C.K. and M.S.; validation, C.K. and M.S.; formal analysis, C.K.; investigation, C.K.; resources, C.K.; data curation, C.K.; writing—original draft preparation, C.K. and M.S.; writing—review and editing, C.K. and M.S.; visualization, C.K. and M.S.; supervision, C.K.; project administration, C.K.; funding acquisition, C.K. and M.S. All authors have read and agreed to the published version of the manuscript.

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Abbreviations

The following abbreviations are used in this manuscript:

LTCC	Low-Temperature Cofired Ceramics
EDX	Energy-Dispersive X-ray spectroscopy
CPW	Coplanar Waveguide transmission line (non-grounded)
SOLT	Short, Open, Load and Thru
GSG	Ground Signal Ground
ADS	Advanced Design System

Appendix A

Verification of the Two-Line Method for CPW Transmission Lines

To verify the eligibility of the two-line method for use with coplanar lines, we proceed in steps:

1. Implement the algorithm on simulated results from microstrip lines
2. Implement the algorithm on simulated results from coplanar waveguide lines
3. Implement the algorithm on measured results from coplanar waveguide lines
4. Validate or not the method

The simulation set up of step 1, using two microstrip lines and ADS is shown in Figure A1.

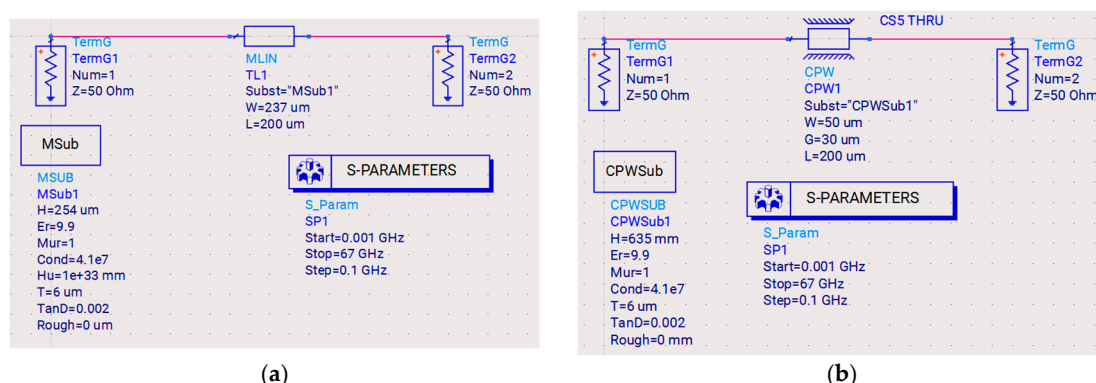


Figure A1. Simulation test bench for (a) the microstrip, (b) the coplanar waveguide line. Two lengths of each type were tested: 200 μm and 550 μm .

From the S-parameters and applying the algorithm given in [12], the obtained effective permittivity is plotted against frequency. The effective permittivity, ϵ_{eff} , is also calculated using the ADS LineCalc tool for the same substrate material at several frequencies (Figure A2). The accordance between calculated and algorithm retrieved ϵ_{eff} is excellent.

In step 2, the same exercise is carried out using the coplanar non-grounded model given in ADS, again verifying that the ϵ_{eff} calculated by the algorithm equals the one found by using LineCalc tool. These results are presented in Figure A3. The agreement between calculated and extracted ϵ_{eff} is again excellent.

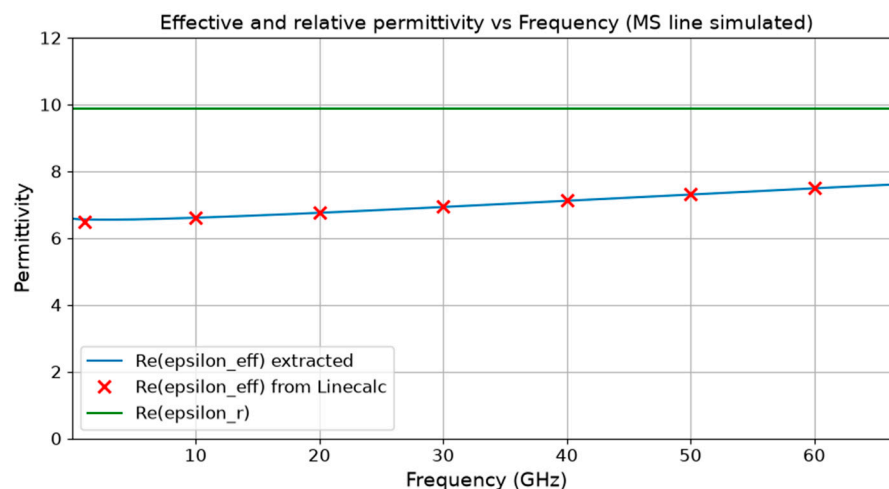


Figure A2. Results from use of the two-line algorithm using the ADS microstrip model. $\text{Re}(\epsilon_{\text{eff}})$ is the value given for simulation and for calculating the ϵ_{eff} from LineCalc.

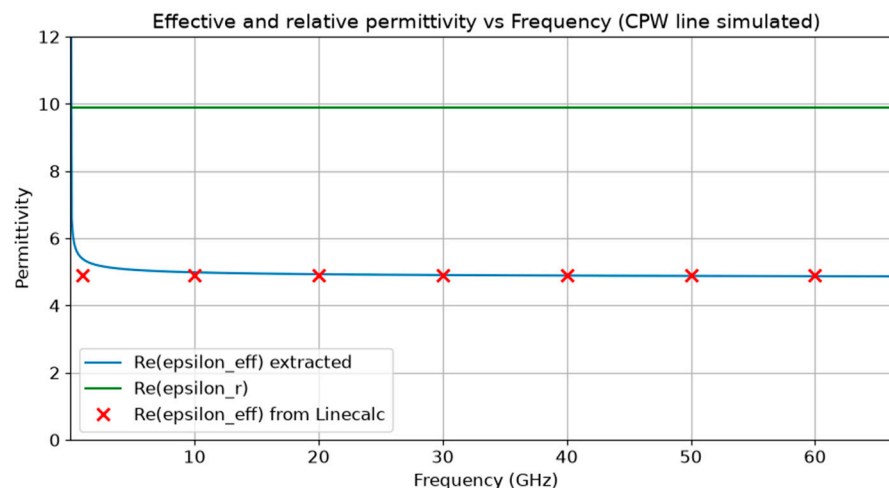


Figure A3. Results from use of the two-line algorithm using the ADS CPW model. The ϵ_r (green line) indicates the verification value used in the LineCalc tool for calculation of ϵ_{eff} (red crosses).

To carry out step 3, a CS5 substrate from Picoprobe is used as the test vehicle. We do not have precise information about the substrate material; however, it is described as alumina. The phase propagation speed, v_p , is reported as $0.442 \times c_0$ [13]. Using Equation (A1), we obtain an ϵ_{eff} of 4.91. LineCalc then yields an ϵ_r of 9.9 for the CS5 substrate, which is consistent with typical values for alumina:

$$v_p = 0.442c_0 = \frac{c_0}{\sqrt{\epsilon_{\text{eff}}}} \quad (\text{A1})$$

The CS5 substrate includes multiple CPW lines with well-defined dimensions. Measurement data taken using the lengths THRU and L3 are processed with the same verification algorithm. The results are shown in Figure A4. When compared with the simulated results in Figure A3 and with the value obtained from the calculation using Equation (A1), the agreement is good. For the simulated CPW line, we assumed an ϵ_r equal to 9.9 initially, and this assumption is supported by the measurement results obtained with the two-line method.

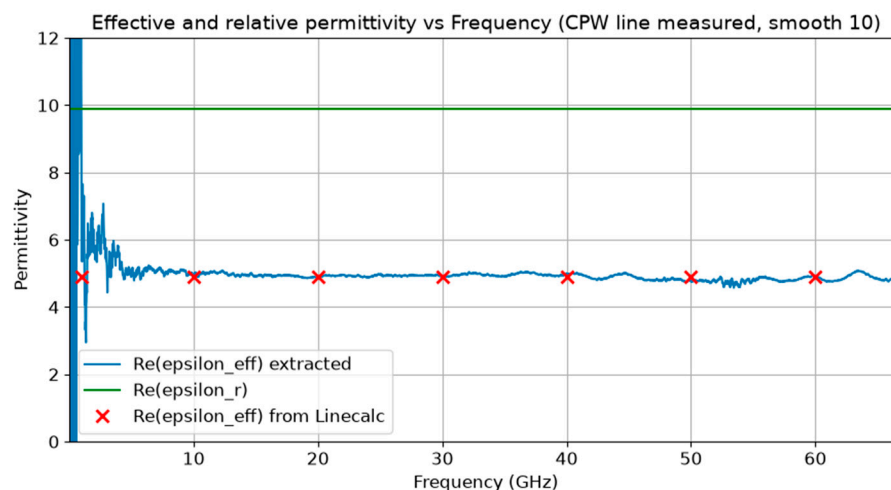


Figure A4. Results from probed measurements of the CS5 substrate (using lengths THRU and L3). The results have been smoothed by a moving average of 10.

Given that the CS5 substrate contains a set of different lines, a follow-up simulation and measurement has been done using the THRU line as reference (length 200 μm) and the lines L3, L4, L5 and L6 with width lengths 550, 1000, 1500 and 6600 μm , respectively. As can be seen from Figure A5, showing simulation results, and Figure A6, from measurements, the results are in agreement. The larger the line-length difference (indicated as ΔL), the shorter the frequency range of valid results. This is expected since results are compromised once ΔL is longer than half the guided wavelength in the material. Thus, a better more wideband result is obtained with a smaller length difference between the two lines. At very low frequencies, the extracted ϵ_{eff} increases rapidly. This part of the model is impacted by the start frequency of the simulation/measurement as well as the frequency step. A smaller step size improves the results. As discussed in [13], coplanar lines cannot provide accurate measurements below 5 GHz because the calibration does not account for line dispersion. This dispersion arises from the CPW being quasi-TEM, with frequency-dependent distributed parameters due to dielectric and conductor dispersion, as well as field penetration effects.

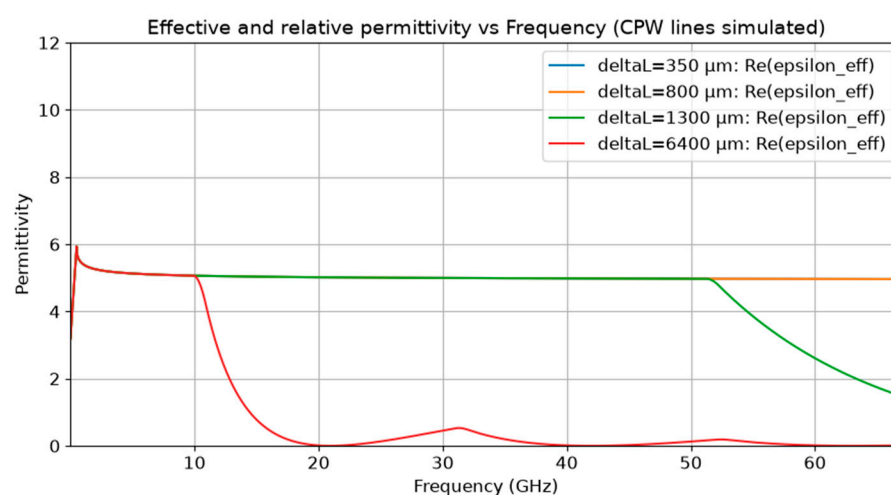


Figure A5. Extraction of ϵ_{eff} from simulation of different line lengths. The blue and yellow lines overlap, why the blue one, $\Delta L = 350 \mu\text{m}$ is not visible in the graph.

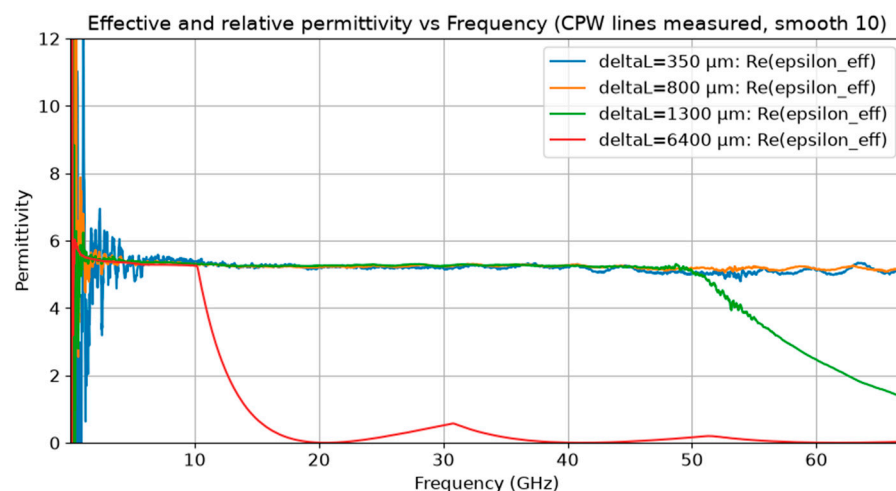


Figure A6. Extraction of ϵ_{eff} from measured results of CS5 substrate lines.

As was presented in [12], this method may give less accurate results for a lossy line compared to a lossless as simulations resulted in a variation in ϵ_{eff} of $\pm 3\%$ for a material having a loss tangent of 0.006 at 3 GHz. In this work, we have assumed a $\tan\delta$ of 0.002, which is normal for high-quality alumina substrate, and the returned result coincides well with the values calculated from LineCalc. As a last test, a new simulation of the CPW lines was performed, where the gap between the grounds and the central line was increased by 10 μm , i.e., increased by 33%, and the roughness was set to 70 μm . Using the same algorithm, we achieve results as presented in Figure A7. The extracted ϵ_{eff} (compared to Figure A3) has increased by approximately 0.4 or 8%. Indeed, the roughness value is chosen to be rather extreme. Also, the added gap width has impacted the line impedance from 50 Ω to 44 Ω . These changes affect the final result, but the outcomes remain within 10% of the expected value. Therefore, we can conclude that, even for an unknown material, the proposed method can determine the effective permittivity with a reasonable degree of certainty.

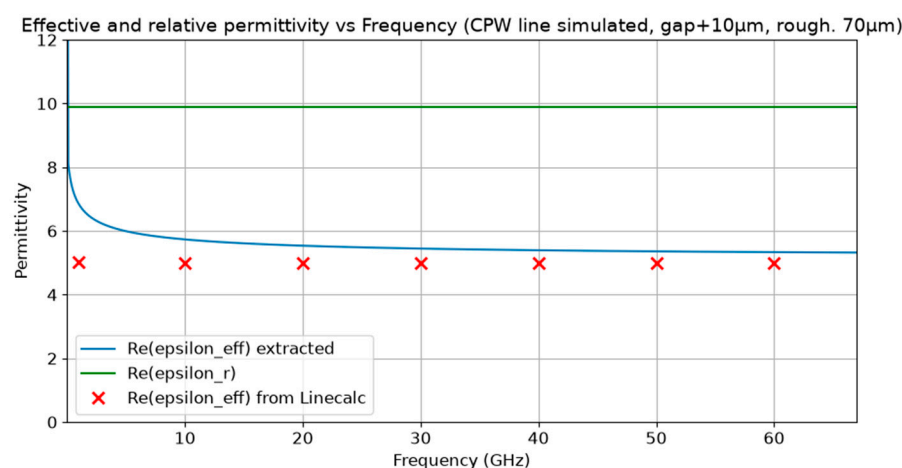


Figure A7. Extraction of ϵ_{eff} from simulated results of CPW lines where the gap has been increased by 33% and the surface roughness is defined to be 70 μm .

Comparing the results from measurement and simulation, the two-line method clearly provides rational results for microstrip line, as well as for coplanar waveguide lines, which concludes step 4. However, for the use of this method and coplanar lines, results based on measurement and SOLT calibration below 5 GHz should be omitted.

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