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Materials for Freeform Fabrication of GHz Tunable Dielectric Photonic Crystals

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ABSTRACT

Photonic crystals are of interest for GHz transmission applications, including rapid switching, GHz filters, and phased-array technology. 3D fabrication by Robocasting enables moldless printing of high solid loading slurries into structures such as the "woodpile" structures used to fabricate dielectric photonic band gap crystals. In this work, tunable dielectric materials were developed and printed into woodpile structures via solid freeform fabrication (SFF) toward demonstration of tunable photonic crystals. Barium strontium titanate ceramics possess interesting electrical properties including high permittivity, low loss, and high tunability. This paper discusses the processing route and dielectric characterization of $(\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3)\text{:MgO}$ ceramic composites, toward fabrication of tunable dielectric photonic band gap crystals.

INTRODUCTION

The material system of barium strontium titanate (BST) has been widely studied for low loss, tunable dielectric applications. Such applications include phased array antennas, photonic crystal waveguides, and photonic crystal filters. Phased array antennas enable beam sweeping without mechanically oscillating the antenna by altering the capacitance, and hence phase, of individual array elements and the vector of the array wavefront. Photonic crystal filters are 3D lattice structures that forbid the propagation of electromagnetic waves at distinct wavelengths or frequencies, thus creating a photonic band gap (PBG).^{1,2} The unique ability of PBGs to create directions of zero effective photonic density of states enables waveguiding, switching, and filtering of a variety of electromagnetic radiation frequencies, from optical to GHz. Introduction of defects into these crystals enables photonic crystal waveguides, which allow propagation of light around 90° bends for the designed wavelength. To date, passive PBGs made of non-tunable materials have generally been produced, while introduction of tunable materials could enable high speed PBG switching and reconfigurable waveguide architectures. Developing a direct-write, tunable 3D PBG filter has been the focus of this research study, as a first step toward waveguides and more complicated devices.

Three methods were combined in this work: (1) fabrication of PBGs by the 3D SFF method of Robocasting³⁻⁷, (2) modeling of PBG transmission and reflection as a function of dielectric permittivity and loss, and (3) development of novel tunable dielectric materials. The combination of these three efforts is aimed at enabling 3D fabrication of active photonic band gap structures, which would enable selective transmission or reflection of frequencies by

electrically tuning the PBG lattice. Examples of a static Al_2O_3 PBG lattice robocast by Smay et al. and its GHz transmission properties are shown in Figure 1.^{4,5}

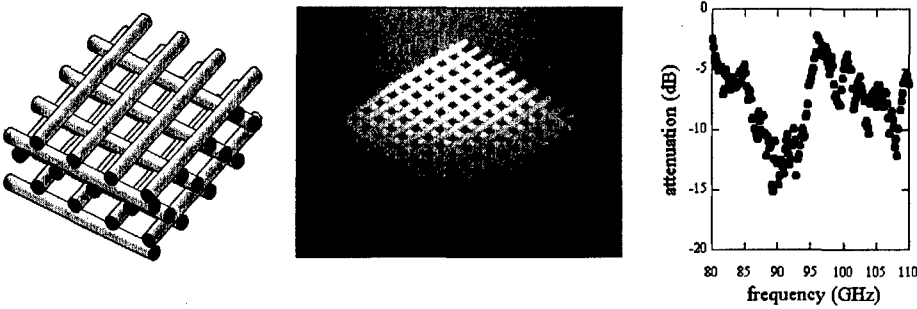


Figure 1. (left) Representation of a face centered tetragonal dielectric woodpile PBG structure, which mimics diamond cubic symmetry, (center) a robocast alumina PBG structure with $300\mu\text{m}$ rods, and (right) GHz transmission measurements of the robocast PBG structure. (Smay et al.^{4,5})

While a large stop band is demonstrated for such Al_2O_3 PBG structures, the ability to selectively tune this behavior from fully transmitting to fully reflecting has not been developed. A means of tuning these materials is use of near-ferroelectric materials, which rearrange domains on application of a DC voltage. For single crystal barium titanate (BaTiO_3), the theoretical maximum tuning of relative permittivity is 97% ($\epsilon_r = 4700 \rightarrow 130$). We are developing similar materials based on $(\text{Ba}_{1-x}\text{Sr}_x)\text{TiO}_3$ (BST), which show a high degree of AC permittivity tuning at low DC biases, as shown in Figure 2.

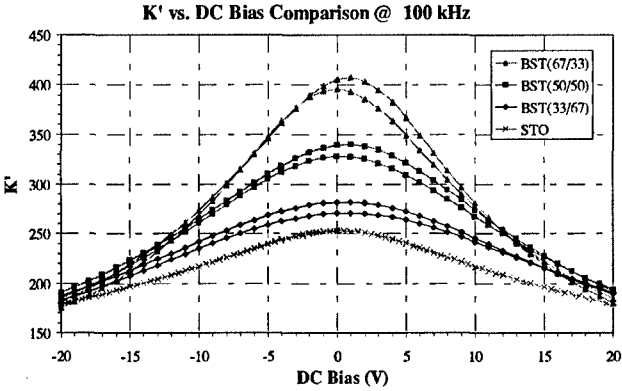


Figure 2. Dielectric constant vs. DC bias for $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ compositions from $x = 0$ to 0.67.

Materials choice for this application involves finding a compromise between high dielectric tunability and retaining a high Q ($= \epsilon_r' / \epsilon_r''$), ideally $Q = 200$ -1000 in the 1-100 GHz range. Significant modeling of PBG structures was conducted with CFD Research Corporation (Huntsville, AL), to understand the influence of permittivity, dielectric loss, and PBG geometry on transmission gap width, depth, and stability. The CFDRD electromagnetic modeling data, acquired with their multiphysics ACE+ modeling software, demonstrated the importance of proper choice of dielectric constant and loss for applications. In brief, the modeling predicted very low loss materials ($\epsilon''/\epsilon' < 0.01$) with a permittivity $\epsilon' < 100$ would be necessary for PBG applications. Materials with higher losses would be expected to be absorptive in the GHz range, while materials with higher permittivities would be expected to reflect, not transmit, GHz radiation outside the designed band gap region. In our present research, we have worked to counterbalance the desired low losses with the desired high dielectric constants (K) and high tunabilities of the material, where tunability is the percent change of the dielectric constant on application of a DC voltage bias (Figure 2). Again, this tunability is a means to shift the photonic band gap frequency, which would enable voltage-tunable filter and other active applications.

The objective of this materials research was to formulate, process, and fabricate a 3-D PBG lattice structure composed of BST mixed with a low permittivity, low loss nonferroelectric oxide, in this case magnesium oxide (MgO).⁸ The first objective was to determine the desired ratio of barium titanate (BT) to strontium titanate (ST) to produce a low loss dielectric ($\epsilon''/\epsilon' < 0.01$) with a reasonable amount of tunability (3-15%). The $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ ratio selected was $x = 33$, BST 33/67. MgO was mixed in a mol ratio of 1:3 BST to MgO to correspond to unit cell matching. The corresponding material system of BST and MgO resulted in loss values less than 1% and tunability of approximately 5%. We are presently fabricating and testing 3D PBG structures of this material.

EXPERIMENTAL DETAILS

$\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ powders were prepared by a solid state processing method. Commercial powders of BaTiO_3 and SrTiO_3 were milled together in ethanol with yttria stabilized zirconia (YSZ, diameter 10 mm) media for 24 hours on a U.S. Stoneware ball mill. The suspension was dried in an explosion-proof drying oven for 12 hours at 60°C. This powder was then dry milled for 4 hours to break up any agglomerates formed during the drying process, and calcined at 1100°C in air for 6 hours. After calcination, the BST powder was mixed with the MgO and ball milled in ethanol for an additional 24 hours. The suspension was again dried in an explosion proof oven for 12 hours at 60°C, and dry milled for an additional 4 hours. The BST-MgO powders were calcined at 1300°C in air for 6 hours. Upon completion of the final calcination step, the powders were sieved using a U.S. Standard Series No. 18 sieve.

The composite powders were uniaxially pressed at 100 MPa, then sintered at 1300°C in air for 6 hours. The sintered ceramic pellet dimensions were 13 mm in diameter and 1 mm or 3mm in thickness. The pellets were then polished and electroded by a DC sputtering unit. Dielectric constant, dielectric loss, and tunability measurements were carried out from 20 Hz to 1 MHz with an HP4284A Precision LCR meter. Dielectric measurements from 0.5-20 GHz were carried out on 7mm diameter, 3mm thickness cylindrical samples using an HP 8510 Network analyzer and HP 85071B software.

RESULTS AND DISCUSSION

Low frequency dielectric measurements, comparing BST pellets to BST-MgO pellets, indicated the addition of nonferroelectric MgO to BST significantly reduced the dielectric constant, as expected. A logarithmic dielectric mixing rule generally appeared appropriate for these composites. For example, BST 33/67 without MgO displayed a dielectric constant of 553, but when 3 moles of MgO were added for every 1 mol of BST, the dielectric constant decreased to 37 at 1 MHz. This addition of MgO allowed the dielectric constant to be tailored to near a goal of $\epsilon' = 40$, as was modeled to be appropriate for PBG transmission.

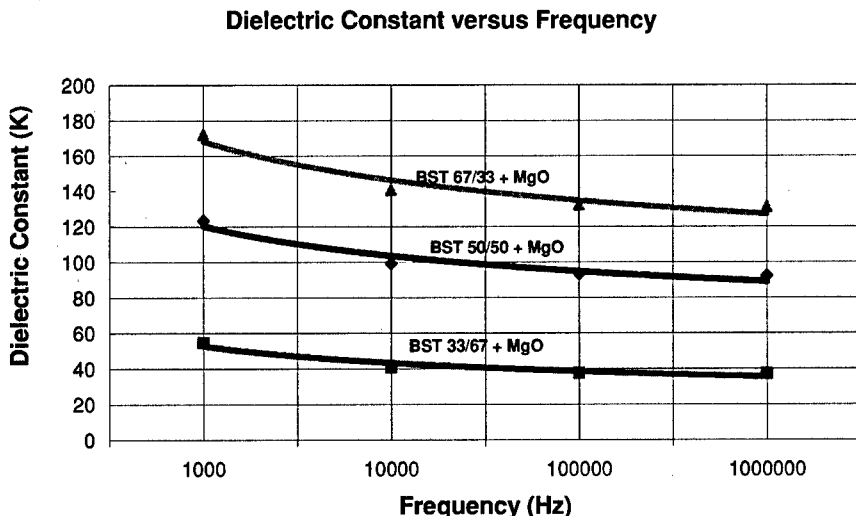


Figure 3. Variation of BST/MgO composite dielectric constants for different Ba:Sr ratios.

As one would expect, on increasing the barium content of the BST, the dielectric constant increased (Figures 2 and 3). This is attributed to the transition from paraelectric behavior to near-ferroelectric behavior as the overall Ba/Sr content nears 67/33. As can be seen in Figure 2, the capacitance-voltage hysteresis loops display an asymmetrical offset for samples of BST 50/50 and 67/33 composition, suggesting local polar regions undergoing domain switching. Such switching would have the effect of raising both permittivity and dielectric loss. For PBG applications, the dielectric loss has a crucial impact on the photonic band gap transmission. In particular, the PBG insertion loss (dB) increases, and the band gap sidewall slope (dB/GHz) and well depth (dB) decrease as dielectric loss increases. BST(33/67):MgO composites appear to display an acceptable combination of permittivity and low loss for tunable GHz applications. Q values above 500 were obtained for direct written composites, measured at 1 GHz (Figure 4).

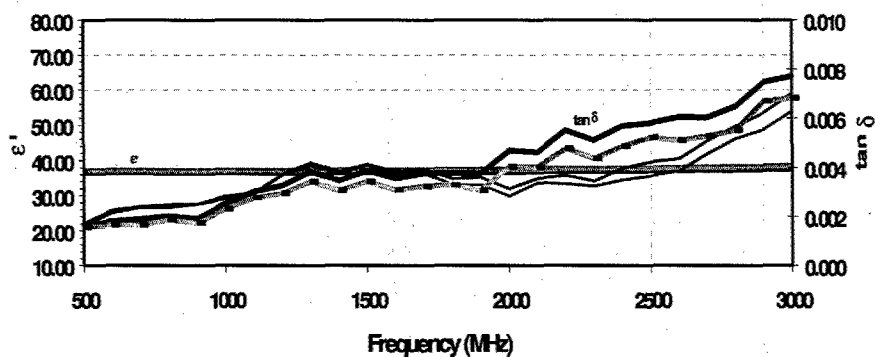


Figure 4. BST(33/67):MgO permittivity and $\tan \delta$ versus frequency for four samples.

The ability to change permittivity and dielectric constant has been predicted by modeling to enable photonic band gap to shifts to different frequencies (Figure 5). Initial results show good correlation between measured Al_2O_3 PBG properties and CFDR modeling results, and is being extended to BST PBG structures.

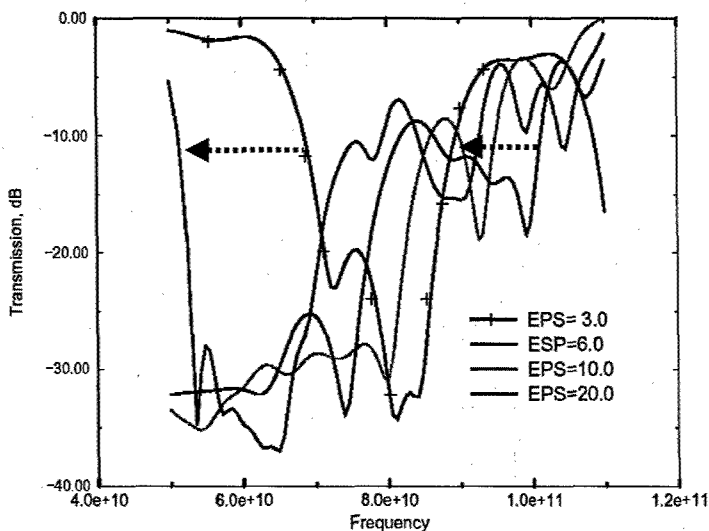


Figure 5. Effect of lattice permittivity on 40-120 GHz PBG transmission, for ϵ' (EPS) = 20 to 3.

CONCLUSION

In this work, we developed high Q, tunable dielectric compositions for direct write compatible, tunable PBG structures. Modeling results suggest a tunable dielectric with a $\epsilon' = 40$ and $Q \sim 1000$ would be a suitable material for this application. From initial studies incorporating (Ba,Sr)TiO₃ with a non-ferroelectric phase, one strong candidate for such photonic band gap filters is BST33/67 + MgO (1:3 molar ratio). By adding the MgO in a 1 to 3 molar ratio, a low dielectric constant ($\epsilon' = 37$) was achieved with low loss. BST 33/67 + MgO composites displayed lower loss values than BST 50/50 or 67/33 composites at high frequency (1 MHz - 100 GHz), which is our region of interest. From the GHz testing, BST33/67 + MgO displayed a high enough permittivity in the GHz region to provide the necessary photonic band gap shift desired. Initial testing of robocast PBGs of BST/MgO composites are underway.

ACKNOWLEDGMENTS

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REFERENCES

1. Special issue on Photonic Crystals, *Advanced Materials* **13**(6), 369-450, (2001).
2. K.M. Ho, C.T. Chan, C.M. Soukoulis, R. Biswas, M. Sigalas, *Solid State Commun.* **89**(5) 413 (1994).
3. J. Cesarano, R. Segalman, P. Calvert, *Ceram. Ind.* **148**(4), 94 (1998).
4. J.E. Smay, J. Cesarano, and J.A. Lewis, *Langmuir* **18**(14), 5429 (2002).
5. J.E. Smay, G.M. Gratson, R.F. Shepherd, J. Cesarano, and J.A. Lewis, *Adv. Mater.* **14**(18), 1279 (2002).
6. B.A. Tuttle, J.E. Smay, J. Cesarano, J.A. Voigt, T.W. Scofield, W.R. Olson, and J.A. Lewis, *J. Am. Ceram. Soc.* **84**(4) 872 (2001).
7. J.E. Smay, J. Cesarano, B.A. Tuttle, and J.A. Lewis, *J. Appl. Phys.* **92**(10), 6119 (2002).
8. L.C. Sengupta, E. Ngo, J. Synowczynski, *Integrated Ferroelectrics* **15**(1-4), 181 (1997).