Experimental and Numerical Studies of Dielectric Properties of BaTiO3-platinum Composites at Microwave Frequencies

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ABSTRACT
The conductor loading method has been used for creating a lossy dielectric material with a desired loss tangent. This method may also be applicable for developing high dielectric constant materials. In this paper, we will present the experimental results of the dielectric properties of $BaTiO_3$ and $BaTiO_3$-platinum $(BaTiO_3\text{-Pt})$ composites. These composite materials were designed to increase the real part of the effective dielectric constant at high frequency. Three different platinum volume fractions were used, 3, 5 and 10%, to make $BaTiO_3$-Pt composites, in addition to a pure $BaTiO_3$ material. To characterize the $BaTiO_3$-Pt composites, microwave frequency measurements were conducted using the waveguide transmission technique. The experimental and numerical results verify that it is possible to increase the dielectric constant using the conductor loading method.

Index Terms — **Microwave dielectric properties, conductor loading, barium tetratitanate.**

1 INTRODUCTION

HIGH relative dielectric constant (ε_r) materials such as barium-tetratitanate ($\varepsilon_r = 37$ at 6 GHz) are often used for wireless devices as a substrate of miniature-sized antennas and dielectric resonators [1]. The typical antenna has a dimension such as patch length and line length of approximately $\lambda/2$ to $\lambda/4$ where λ is the effective wavelength due to the substrate material or structure. Because most wireless systems operate at a lower microwave region, one technique to reduce the physical dimension is to fabricate antennas on a high ε_r substrate or resonator. The typical high-ε_r substrate materials are titania ($\varepsilon_r = 96$ at 6 GHz) based composites and ε_r ranges

from 30 to 100. These dielectric materials also have relatively low loss characteristics at microwave frequencies. Although some materials such as strontium titanate ($SrTiO_3$) have very high ε_r (2000 to 4500 within a limited temperature range), they have limited use as an antenna substrate due to the requirement of a precise temperature control.

It is known that when dielectric materials are loaded with conducting particles, both the real and imaginary parts of ε_r will increase as the fractional volume of inclusion increases [3, 4]. This technique has been used for designing lossy dielectric materials such as beryllia-silicon carbide (BeO-SiC) and AlN-SiC for microwave tube applications [2]. The undoped beryllia (BeO) has a dielectric constant close to 6.4 and small loss tangent at the GHz region. When BeO is doped with SiC, the resulting composite materials show much higher real and imaginary parts of ε_r. The BeO-SiC composite is designed to absorb electromagnetic waves, and one of the requirements is high loss tangent which can be controlled by the amount of SiC.

The conductor loading method may also be applicable for creating high-ε_r materials. However, the main problem is that when the low-loss host material is loaded with conducting particles, both real and imaginary parts of ε_r tend to increase. The rate of increase depends on the volume fraction, size and shape, and conductivity of inclusions. Extensive analytical and numerical studies on the dielectric properties of composite materials have been conducted in the past, but there is no accurate model to predict the complex dielectric constant [4-7]. Moreover, there is very little published experimental data to compare with calculations. In this paper, we will present the experimental results of the dielectric properties of conductor-loaded composite materials.

2 MATERIAL PROCESSING

As we stated previously, our objective is to create a material with a very high dielectric constant using the conductor loading technique. Because the expected increase is 10 to 100% depending on the amount of inclusion and the volume fraction of the conductor cannot be too high because it will introduce conductor loss, the host material must have a high dielectric constant with a low-loss tangent. Baruim titanite oxide ($BaTiO_3$) which has an $Re(\varepsilon_r)$ of 90-110 and loss tangent of 0.001 is a good candidate, and this was chosen as a host material [10]. The conductivity of the inclusion material must be sufficiently high to minimize the conductor loss at microwave frequencies. In addition, conducting particles must withstand the high-temperature and high-pressure processing. Although silver, gold and copper are excellent conductors, they don't meet the other requirements. We chose platinum as the inclusion material. The conductivity of platinum [$\sigma = 9.52 \times 10^6$ (S/m)] is less than that of gold [$\sigma = 4.098 \times 10^7$
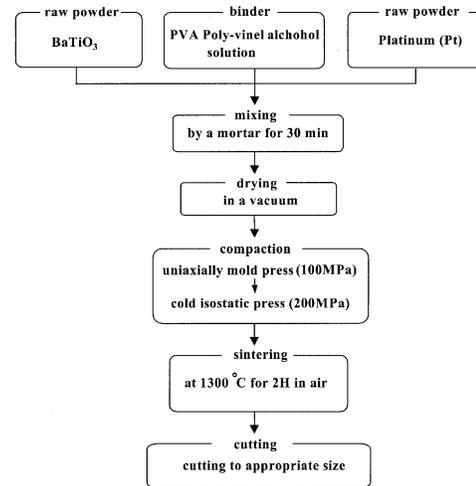

Figure 1. Flow chart of processing procedure.

(S/m)], but the material is suited for the high temperature processing.

Baruim titanite oxide ($BaTiO_3$) ceramics and $BaTiO_3$ platinum (Pt) composites are fabricated by sintering powder compacts of $BaTiO_3$ and platinum. Predetermined amounts of $BaTiO_3$ powders were mixed with platinum powders in a mortar with a pestle. The fabrication procedures (flow chart) of the porous piezoelectric materials are shown in Figure 1. The starting material ($BaTiO_3$ powder, platinum, and PVA binder (poly-vinyl alcohol) are mixed by a mortar for 30 minutes. The mixture is then dried in a vacuum. Two stages of powder compaction are carried out, uniaxial pressing under 100 MPa to form the green samples and cold isostatic pressing under 200 Mpa. The samples are then sintered at 1300° C for 2 h in air. The samples are then machined to the required dimensions for experimental investigation.

The sintered density of the composites is then determined. After sintering at 1300° C for 2 h, the powder compact is sintered to near full density. The $BaTiO_3$ platinum composites processed have 0, 3, 5, and 10 volume percent of platinum. The microstructures of the composites are observed using a scanning electron microscope (SEM) as shown in Figure 2. The white pores are the spaces that were occupied by the platinum particles. The size distribution of platinum particles is quite large as shown in Figure 2. Finally, the sample is cut into an X-band dimension and polished (Figure 3).

3 WAVEGUIDE DIELECTRIC CONSTANT MEASUREMENT TECHNIQUE

There are several different techniques to measure the dielectric constant of materials at microwave frequencies. If a sample can fit inside a waveguide or coaxial line, the most commonly used method is the Nicolson-Ross method
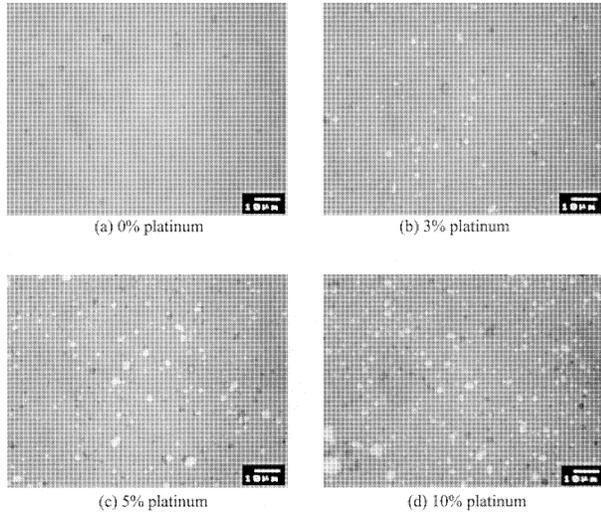
Figure 2. Micro-structural view of $BaTiO_3$ platinum composites.

and its variations which improves the stability for the sample length as it becomes close to $\lambda/2$ [8, 9]. Recently, we have developed a robust waveguide method to characterize lossy dielectric materials at X- (8.2−12.4 GHz) and S (2.6−3.9 GHz) bands [3]. The same setup and processing method will be used for this project.

The sample is placed inside the X-band waveguide, and the scattering parameters (S parameters) of the sample are obtained with the vector network analyzer (NWA) as shown in Fig. 3. This is the layered structure problem in which the material properties ε_r and μ_r are included in the characteristic impedance Z_s and propagation constant γ. S_{11} (reflection) and S_{21} (transmission) of the incident wave can be expressed with the following equations [1, 2].

$$\Gamma_1 = \frac{Z_S - Z_0}{Z_S + Z_0}$$

$$\Gamma_2 = \frac{Z_0 - Z_S}{Z_0 + Z_S} = -\Gamma_1$$

$$\Gamma_3 = \frac{Z_0 - Z_S}{Z_0 + Z_S} = \Gamma_2 = -\Gamma_1$$

$$T_{21} = 1 + \Gamma_1$$

$$T_{12} = 1 + \Gamma_2 = 1 - \Gamma_1$$
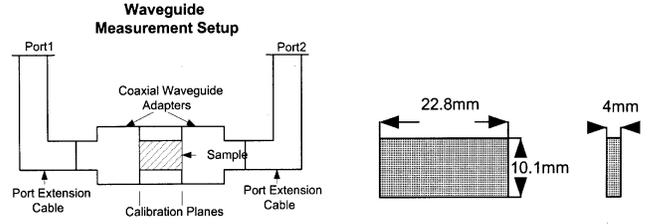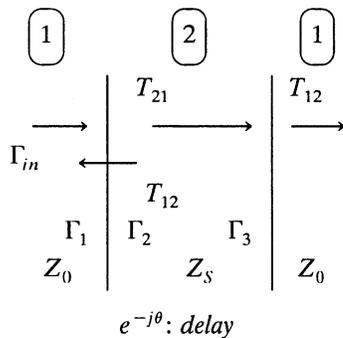

$e^{-j\theta}$: *delay*

Figure 3. Left: Permittivity measurement setup with the waveguide sample holder; Right: Processed X-band waveguide sample size.

The total reflection S_{11} can be expressed as a summation of all reflections.

$$S_{11}(\omega) = \Gamma_1 + T_{12}T_{21}\Gamma_3 e^{-2j\theta} \sum_{n=0}^{\infty} \Gamma_2^n \Gamma_3^n e^{-2jn\theta} \quad (1)$$

Since $\sum_{n=0}^{\infty} x^n = \dfrac{1}{1-x}$ for $|x| < 1$, we get

$$S_{11}(\omega) = \Gamma_1 + \frac{T_{12}T_{21}\Gamma_3 e^{-2j\theta}}{1 - \Gamma_2\Gamma_3 e^{-2j\theta}} = \frac{\Gamma_1(1 - e^{-2j\theta})}{1 - \Gamma_1^2 e^{-2j\theta}} \quad (2)$$
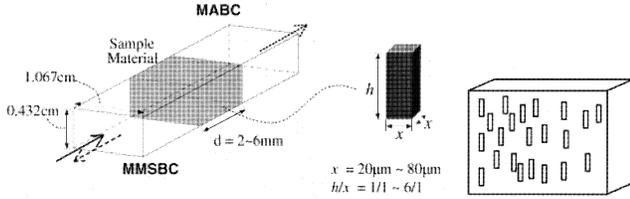
Similarly, we can find S_{21} as

$$S_{21}(\omega) = T_{21}T_{12}e^{-j\theta}\frac{1}{1 - \Gamma_3\Gamma_2 e^{-2j\theta}} = \frac{(1 - \Gamma_1^2)e^{-j\theta}}{1 - \Gamma_1^2 e^{-2j\theta}} \quad (3)$$

where the phase shift θ is given by $\theta = \gamma L$ and L is the sample thickness. The wave number in the direction of wave propagation, γ, is given by

$$\gamma = \sqrt{\frac{\omega^2 \mu_r \varepsilon_r}{c_0^2} - \left(\frac{2\pi}{\lambda_c}\right)^2} \quad (4)$$

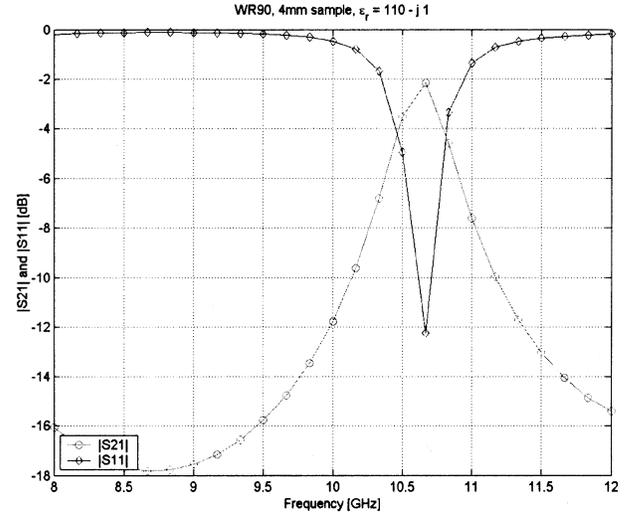where c_o is a speed of light and λ_c is the cutoff wavelength of X band waveguide ($\lambda_c = 4.573$ cm).

The material properties μ_r and ε_r are included in the reflection coefficient Γ_1 and γ in equations (2) and (3). In principle, both μ_r and ε_r can be obtained from S_{11} and S_{21}. If the sample material is non-magnetic ($\mu = \mu_o$), the permittivity ε_r has two unknowns, a real and an imaginary part, so that two equations are generally needed to solve the problem. Either S_{11} or S_{21} (complex variables) can be used to calculate ε_r. However, most experimental data contain random and systematic errors, and we have found that the S_{11} method is susceptible to a small amount of error in experimental data. Because of this concern, the inversion based on S_{11} was not used. The most robust approach to estimate ε_r is to measure S_{21} of the same material with two different thicknesses. By combining two S_{21} measurements and solving for one ε_r, it is possible to decrease the measurement uncertainty and increase the accuracy. Unfortunately, we have only one sample thickness in this project and the data inversion was conducted with one S_{21} data.

Figure 4. Configuration used for the FDTD simulations. The dimension of conducting particles and distributions in a sample (random case) are also shown.
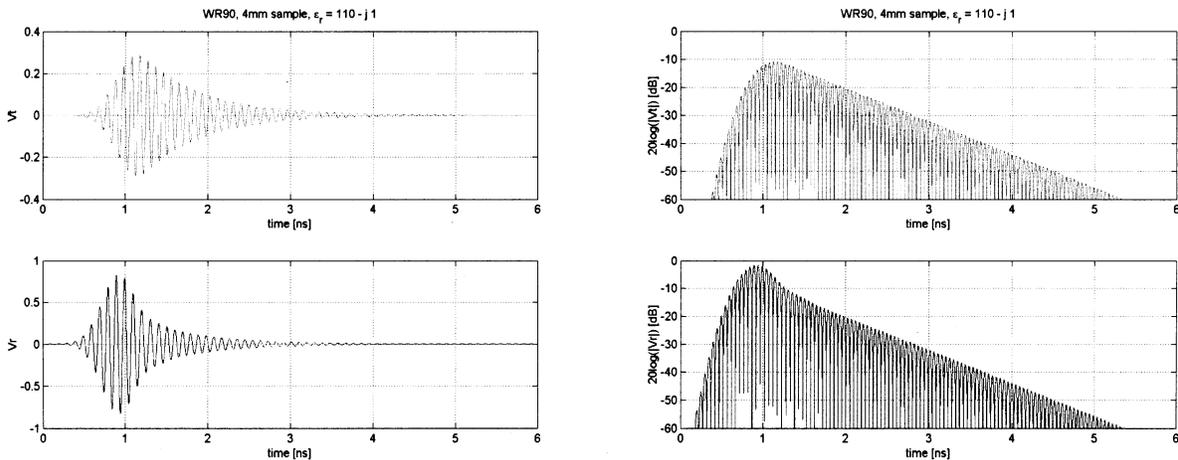
4   FDTD SIMULATIONS OF BaTiO$_3$ COMPOSITE AND INVERSION

The objectives of the numerical simulations are (1) to test whether the data inversion method works for BaTiO$_3$ composite and (2) to test the effects of the shapes and orientations of inclusions. Most of our previous experiences with dielectric constant measurement were either lossy (loss tangent = 0.3 to 0.5) or low-loss dielectric materials such as a Teflon composite. To test whether the X-band waveguide method is suited for BaTiO$_3$ composites, we conducted finite difference time domain (FDTD) simulations using homogeneous materials. The FDTD method has been widely used for high frequency applications and details can be found in many references [11]. The simulation structure is shown in Figure 4. A sample material is assumed to have the dielectric constant ε_r = 110-j1 and thickness = 4 mm. Figure 5 shows both S$_{11}$ and S$_{21}$ responses in time-domain. Unlike lossy materials or low-loss materials, the time-domain responses of a BaTiO$_3$ sample contain long tails. In most microwave frequency systems, small reflections from connectors and adapters are unavoidable. One of the nice features of modern NWA is a time-domain display which can be obtained from the original frequency-domain measurement. The location of large reflections can easily be identified in time-domain, and the time-domain gating function is usually employed to eliminate unwanted reflections. However, if a S_{21} time

Figure 6. Frequency-domain responses of BaTiO$_3$ sample inside the X-band waveguide. Upper line S_{11} and lower line S_{21}. FDTD simulation. $\varepsilon_r = 110$-$j1$ is used.

response has a long tail, it may be difficult to separate unwanted reflection from a desired response. The FDTD simulation provides the time-domain responses. To estimate the material properties, the time-domain response must be converted into the frequency-domain response and the inversion method such as genetic algorithm (GA) must be applied. Figure 6 shows the frequency-domain response of the same FDTD simulations. The correct value of ε_r was estimated from data shown in Figure 6 with equations (2) and (3). FDTD is a flexible method and, in principle, the different sizes and shapes of conducting particles can be included in a host BaTiO$_3$ in order to study their effects. In practice, however, the size of platinum particles cannot be too small due to the memory and CPU time requirements. Nevertheless, the FDTD simulations were conducted with two different particle shapes and the results will be discussed later.

Figure 5. Time-domain responses of BaTiO$_3$ sample inside the X-band waveguide. Top S$_{21}$ and bottom S$_{11}$. FDTD simulation. $\varepsilon_r = 110$-$j1$ is used.

5   EXPERIMENTAL RESULTS

Figure 7 shows the S_{21} responses of the BaTiO$_3$ sample without platinum particles in both time- and frequency-domains. NWA is properly calibrated to obtain S_{21} data shown in Figure 7. The general trend is very similar to that of the FDTD simulation shown in Figures 5 and 6. As expected the time-domain response shows a long tail, and it is difficult to determine the optimum gate position. To isolate a desired response from the reflection caused by the waveguide-to-coaxial adapter, we also conducted the measurement with a long X-band waveguide, but there was no significant difference.

Figure 8 shows the time-domain responses of the BaTiO$_3$ composite with Pt = 3% and Pt = 10%, respectively. The inclusion of platinum does not change the shape significantly indicating that the loss part is still small in these samples. The gate position was chosen to select a desired response only. For example, the second large peak shown in the Pt = 0 and 3% cases (left figure in Figures 7

and 8) is a spurious reflection, and it must be eliminated before processing data. It appears that the Pt = 10% case (Figure 8, right) shows a longer tail compared to the Pt = 0% case. This can be explained in that the Pt = 10% case has a higher dielectric constant than that of Pt = 0%. Therefore, the energy will be trapped within the material longer than that of the Pt = 0% case.

The complex dielectric constants are estimated from the measured frequency domain S_{21} data shown in Figure 9 for four different platinum concentrations. The accuracy of the estimation depends on the random and systematic noises in the original S_{21} data. Based on previous experience, we have found that if the magnitude of S_{21} is greater than 0.2, the error in estimation is small. Figure 10 shows the estimated ε_r of four Pt concentrations. If we estimate ε_r with more controlled data of $|S_{21}| > 0.4$, the average value of effective dielectric constants are $\varepsilon_r = 112$ for Pt = 0%; $\varepsilon_r = 121$ for Pt = 3%; $\varepsilon_r = 129$ for Pt = 5%; and $\varepsilon_r = 146$ for Pt = 10%. The effective dielectric constant is
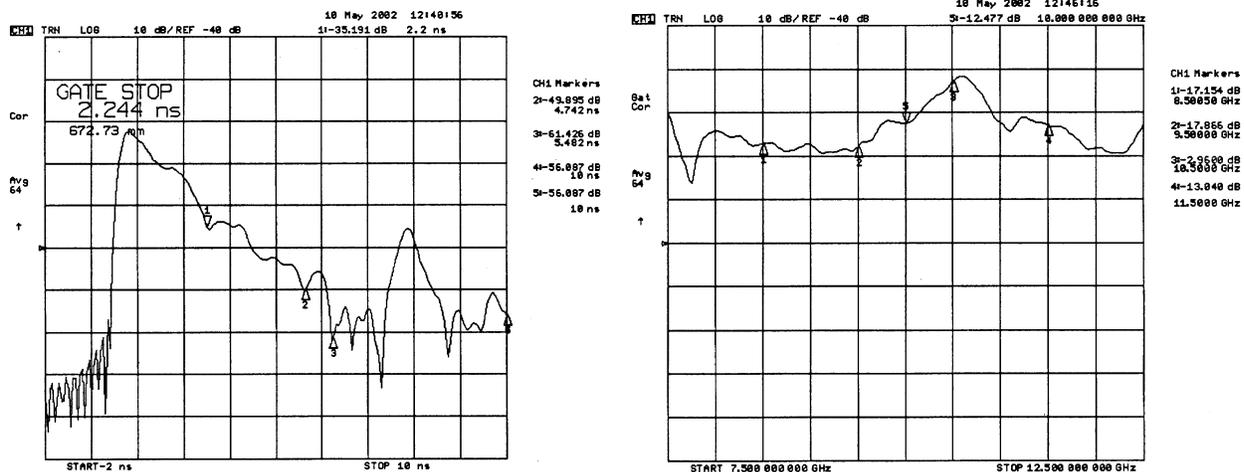

Figure 7. Time- and frequency-domain responses of BaTiO$_3$ sample (Pt = 0%). In frequency-domain, the gate width is set to -0.5 ns to 5.5 ns.
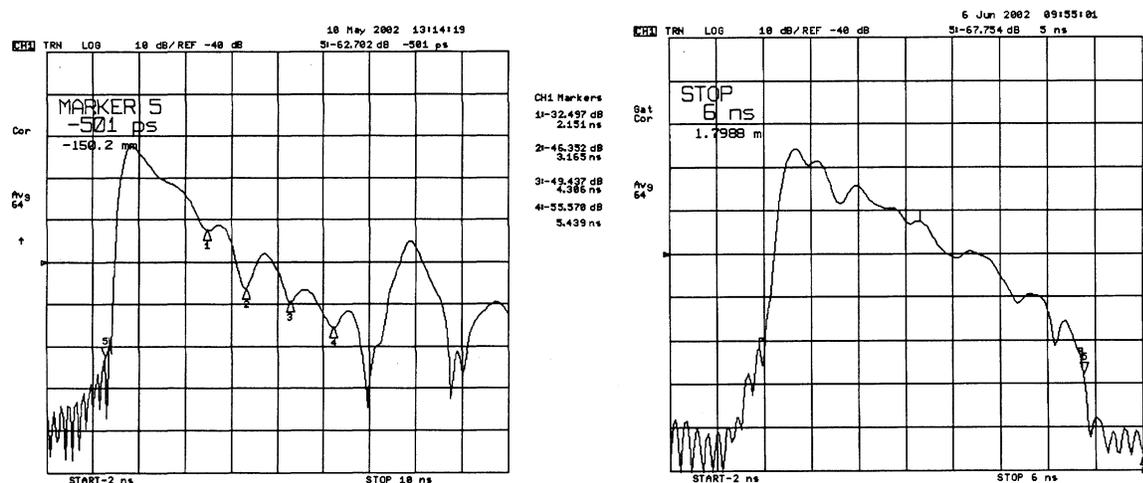

Figure 8. Time-domain responses of BaTiO$_3$ samples. Left, Pt = 3%; Right, Pt = 10%.

increased by 30% over this range. The waveguide transmission technique is not suited for measuring a small imaginary part accurately, and results of the imaginary part may contain significant error. However, the imaginary part does not increase much up to the platinum concentration of 10%. A better method to estimate the imaginary part is the cavity-based techniques.

In this experiment, we did not control the sizes and shapes of platinum particles, and most particles are found to be much smaller than 10 μm. To study the effects of particle sizes and shapes on the effective dielectric constant, we have conducted the FDTD simulations [12]. Figure 11 shows both real and imaginary parts of ε_r for four different aspect ratios. The size of base area (x by x) is fixed at 40 μm x 40 μm and the height (h) is changed from 40 to 240 μm. It is clear that by controlling the aspect ratio and orientation of particles, we may be able to increase the real part of the effective dielectric constant significantly. This has to be tested experimentally.

6   CONCLUSIONS

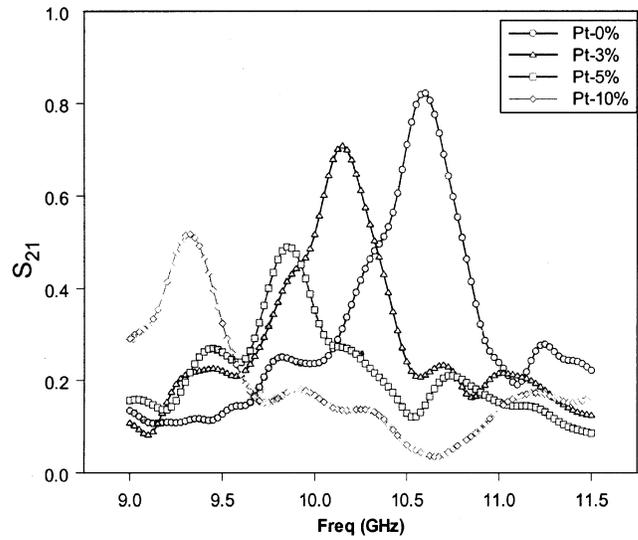Although our results are still preliminary, we have shown that a higher dielectric constant can be achieved

Figure 9. Magnitude of S_{21} for four different Pt concentrations. Pt = 0%, Pt = 3%, Pt = 5%, Pt = 10%.

with the conductor loading technique. The increase is approximately 30-35% for the Pt = 10% case. Unlike lossy dielectric materials such as BeO-SiC, we have found that
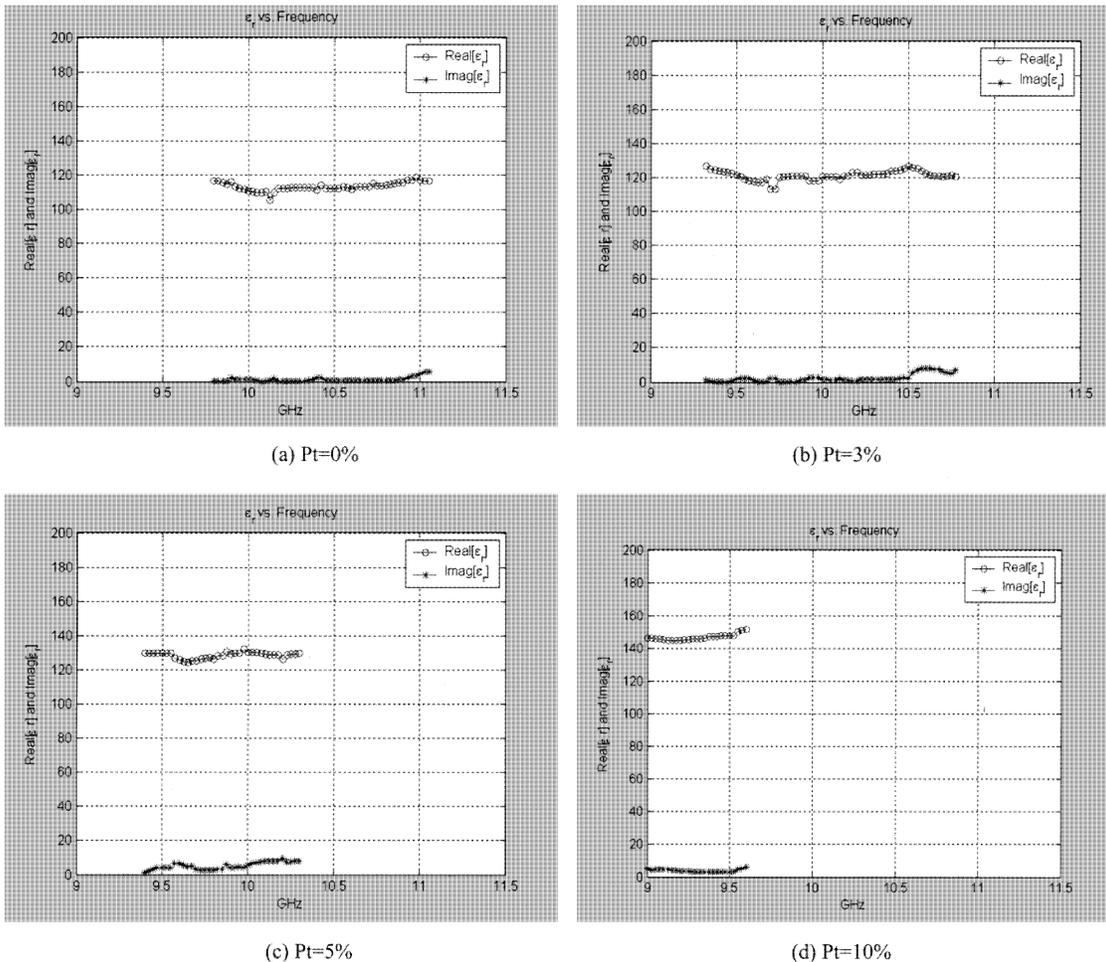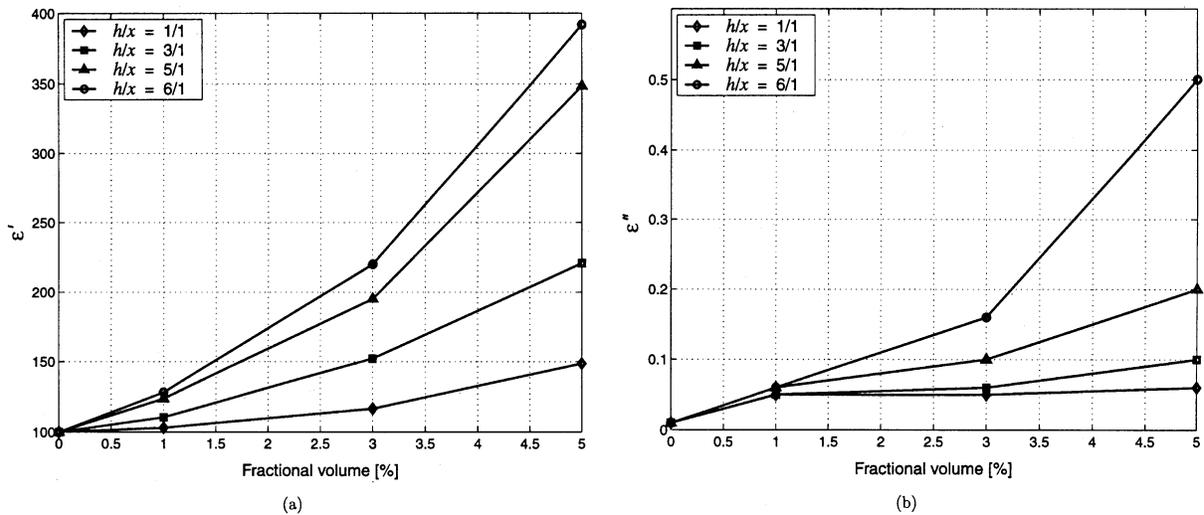
(a) Pt=0%   (b) Pt=3%

(c) Pt=5%   (d) Pt=10%

Figure 10. Estimated dielectric constant of BaTiO$_3$ composites for four different Pt concentrations.

Figure 11. Real (left) and imaginary (right) part of ε_r versus fractional volume of conducting particles. Particles are perfect conductor with the base size of $x = 40\,\mu m$. The host $= \varepsilon_r\ 100\text{-}j0.01$, frequency $= 2.2$ GHz. The particle height h is varied to show the effects of aspect ratio (see Figure 4).

the high ε_r and low loss materials pose problems in both measurements and data inversion process. The instability encountered in the inversion process may be due to the inaccurate data obtained from NWA. The gate position and width as well as the material placement are critical in this experiment. Although a silver paint was used for plugging small air gaps between the sample and waveguide, another source of error in measurements is these small air gaps. Finally, the FDTD numerical simulations show that a further enhancement of dielectric constant is feasible by controlling the particle aspect ratio and orientation.

ACKNOWLEDGMENTS

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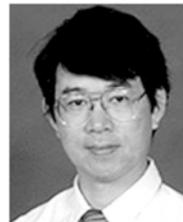
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