

TUNABILITY OF GRANULAR FERROELECTRIC DIELECTRIC COMPOSITES

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Abstract—Electrical tunability of a composite consisting of small ferroelectric spheres randomly dispersed into a dielectric background is studied. A new method to calculate the effective permittivity of such a nonlinear composite is introduced. The method is based on the Bruggeman effective medium theory and a specific model for the nonlinear permittivity of the ferrite. The resulting tunability (defined as a measure of the change in the permittivity due to the bias field) is a continuous function of the volume fraction of the ferroelectric material phase in the composite. As an example, SrTiO₃ is studied with two different nontunable background materials.

1. INTRODUCTION

Tunability describes the extent in which the permittivity of the material can be controlled using external biasing electric field. It is defined as the ratio between the maximum and minimum permittivities of the tunable material. Tunable materials are typically ceramics with very high relative permittivity values, up to thousands. In [1], the high permittivity of a ferroelectric ceramic is applied to create chiral particles. On the other hand, high permittivity can be disadvantageous: it can increase cross talking in integrated circuits and to reduce signal speed [2, 3]. One possibility to avoid high permittivities is to construct composites where nontunable low-permittivity dielectrics form the other component phase [4, 5]. Such materials can be made, for example, by compressing ceramic powders of dielectrics and tunable ferroelectrics.

Composites can be also useful in the design of tunable materials with modified mechanical properties. For some applications such as for low-temperature co-firing ceramic material (LTCC) technology [6, 7]

certain mechanical properties for tape materials are required. Tunable ferroelectric polymers, such as poly(vinylidene fluoride) (PVDF) [8, 9] are typically lossy materials with long switching time which limits their applicability at microwave frequencies. On the other hand, a composite consisting of ferroelectric inclusions in a polymer matrix can provide a flexible sheet with tunable permittivity and at the same time maintain the tunability properties of the ceramic. Tunable composites can be applied in the design of, for example, dual band antennas [10], tunable electromagnetic band-gap (EBG) structures [11], or tunable split ring resonators [12].

In Fig. 1 an illustration of an example of a tunable composite material is presented. There black inclusions of tunable material are randomly dispersed into a white nontunable background. To highlight the possibility that the composite can be completely filled with black inclusions, the boundaries between white inclusions are marked. The illustration corresponds, for example, to a composite that is constructed by compressing two ceramic powders. However, in the case of ceramic-polymer composite, there would be no boundaries between white inclusions. Although a crude model, the illustration in Fig. 1 seems plausible geometry for our approach, in light of the study of a BST/MgO-composite in [13].

Although there are several studies highlighting the promising experimental results with tunable composites, the theoretical background of such composites has not been widely analyzed. The studies [14–16], which base on earlier nonlinearity models for dielectrics [17–19] are based on a Bruggeman type of effective medium theory. There the tunability of the composite material has been modeled for low volume filling ratio of nontunable spherical inclusions in a tunable background. The main problem is that the tunability of the composite cannot be modeled with all volume fractions of tunable material. In particular, the tunability as a function of volume fraction of tunable phase has a singularity which is not in full agreement with experimental studies.

The present study provides a new approach to this problem of modeling nonlinear composites. Here, the effective medium approach is similar to that in these earlier studies, but the effect of the nonlinearity is taken into account in a different way. The result is a stable function without any singularities which can be applied without restrictions to the volume fraction of tunable material.

The new approach is based on the Bruggeman effective medium theory and a nonlinear model for the permittivity of the ferroelectric. The nonlinear model presented in [20] is used for the permittivity tunable material phase in the composite, due to its good agreement

with measurements. This nonlinearity will result in the tunability of the composite through the homogenization algorithm. The resulting tunability of the composite is a function of the volume fraction of inclusions, the temperature, the average size of inclusions, and the biasing electric field. The resulting tunability and permittivity are nonresonating continuous functions. The method can be applied with other models for the tunable material phase as well.

For simplicity, in the analysis to follow, tunable inclusions are modeled as spheres in order to reduce the amount of free parameters in characterizing the mixture. However, the method can as well be applied to ellipsoidal geometries. In such a case, the internal electric field that excites the particles is dependent on their geometrical shape parameters.

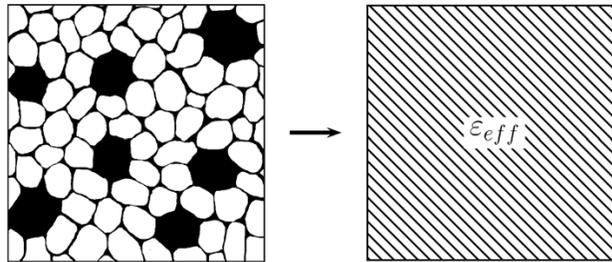


Figure 1. Homogenization of a two phase mixture of black inclusions with permittivity ε_i in background with ε_e . The effective permittivity ε_{eff} is dependent on the external bias field.

2. THEORY

This section describes how the effective permittivity of a composite consisting of tunable and nontunable materials can be determined and calculated with knowledge of the properties of the materials that it is composed of. The complex properties of the nonlinearity of the tunable permittivity will be transformed into the nonlinear properties of the composite. To model the permittivity of the tunable material we use the model presented in [20] which is outlined in Appendix A. It is a nonlinear function of the electric field inside the tunable phase. However, the analysis to follow is general in the sense that instead of the model presented in Appendix A, any other model describing the permittivity of a tunable material as a function of the electric field can be used. Also a model for the permittivity of a polycrystal dielectric ceramic is presented in Appendix A. The Bruggeman effective medium

theory is used to calculate the field in the tunable material as well as the effective permittivity of the composite. The tunability of the composite can be calculated from the effective permittivity.

2.1. Homogenization

The effective permittivity ε_{eff} of a mixture is a macroscopic quantity describing the average permittivity over a large sample of composite material. It can be defined for time-varying fields with sufficiently low frequencies, such that the wavelength is considerably smaller than the average inhomogeneities in the composite. There are several theories to describe the effective permittivity of a randomly heterogeneous mixture. In this study, a Bruggeman effective medium theory is chosen because of its many appealing properties. It averages the permittivity around each inclusion and the electric field inside the sphere depends on the volume fraction of the spheres in the mixture.

As a result, the Bruggeman effective medium theory for spheres has a percolation threshold at volume filling ratio $1/3$. The percolation threshold is the volume filling ratio where the effective permittivity as a function of volume fraction of inclusions changes rapidly. For this amount of filling, randomly dispersed spheres become connected [21]. Another advantage of the Bruggeman mixing theory is that it gives a reasonable estimate for the effective permittivity of the mixture with all volume fractions of inclusions. This is a consequence of the particular symmetry property of the Bruggeman equation: it treats both phases in equal fashion. The equation reads [22]

$$f \frac{\varepsilon_f - \varepsilon_{\text{eff}}}{\varepsilon_f + 2\varepsilon_{\text{eff}}} + (1 - f) \frac{\varepsilon_d - \varepsilon_{\text{eff}}}{\varepsilon_d + 2\varepsilon_{\text{eff}}} = 0 \quad (1)$$

where f is the volume filling ratio of ferroelectric material in the composite, ε_d is the permittivity of the dielectric phase and ε_f is the permittivity of the ferroelectric material. Permittivities of materials consist of real and imaginary part: $\varepsilon_d = \varepsilon_{d,r}(1 - j \tan \delta_d)$ and $\varepsilon_f = \varepsilon_{f,r}(1 - j \tan \delta_f)$ where $\varepsilon_{d,r}$ and $\varepsilon_{f,r}$ are the real permittivities and $\tan \delta_d$ and $\tan \delta_f$ are the loss tangents of materials. For tunable composites, at least one of the phases has a nonlinear dielectric response. Here, the permittivity of the ferroelectric depends on the internal electric field according to the model presented in Appendix A.

The first term in Eq. (1) represents the polarizability of a ferroelectric sphere with permittivity ε_f in the background with permittivity ε_{eff} weighted with the volume fraction of ferroelectric material. The second term represents the polarizability of a dielectric sphere with permittivity ε_d in the dielectric background weighted with

the volume fraction of material ϵ_d . With small volume fractions, the ferroelectric material is dispersed as spheres into the dielectric background and with large volume fraction of ferroelectric material, dielectric spheres are randomly distributed into the ferroelectric background.

The classical Bruggeman effective medium theory is derived for linear dielectrics. Here the Bruggeman model is extended for nonlinear spheres by assuming that the internal field depends linearly on the external field, but the permittivity of the spheres depends nonlinearly on the same internal field E_i . The internal field is determined by the applied external biasing field (which is much stronger in amplitude than the time-dependent signal field), and because of the spherical inclusion shape, it reads

$$E_i = \left| \frac{3\epsilon_{\text{eff}}}{2\epsilon_{\text{eff}} + \epsilon_f} \right| E_B \quad (2)$$

where E_B is the applied biasing field to the whole sample. By following the logic behind the Bruggeman effective medium theory, the surrounding space around the ferroelectric sphere is homogenized to have the effective permittivity ϵ_{eff} . With very small volume filling ratios of ferroelectric material, the internal field of the ferroelectric approaches the internal field of an isolated sphere in the background material ϵ_d . With very large volume filling ratios the internal field approaches the average biasing field over the whole sample. Between these two extremes, the internal field is a continuous function if ϵ_f is continuous.

Effective permittivity of a ferroelectric–dielectric composite can be solved with Eq. (1) for the effective permittivity, Eq. (2) for the internal field and Eq. (A1) for the permittivity of the ferroelectric. Fortunately, this set of three non-linear equations is relative straightforward to solve. It can be solved iteratively with initial value $\epsilon_{\text{eff}} = \epsilon_d$ in $f = 0$. Then the volume fraction f is increased and the corresponding effective permittivity is solved recursively using the previous solution as an initial value. For simplification, the loss tangent is assumed to be constant.

The electric field inside the sphere is constant, as it is in a thin sheet of ferroelectric material between capacitor plates. Therefore it is assumed that in the model for the ferroelectric (A1), the height of the sheet is replaced with the average diameter of spheres.

2.2. Tunability and the Figure of Merit

The tunability of the composite is a very central parameter for nonlinear materials. It is defined as the ratio of the maximum to the minimum permittivity:

$$n = \varepsilon_{\text{eff}}(0)/\varepsilon_{\text{eff}}(E_b) \quad (3)$$

where $\varepsilon_{\text{eff}}(0)$ is the permittivity without the biasing field and $\varepsilon_{\text{eff}}(E_b)$ is the permittivity with the biasing electric field. The permittivity with the biasing field is smaller than without the biasing field. Both of these are functions of the volume fraction of ferroelectric inclusions. In addition to large tunability, it is usually desirable to have low losses. A figure of merit for a tunable material which measures both quantities at the same time is defined as [23]

$$K = \frac{(n - 1)^2}{n \tan \delta_0 \tan \delta_E} \quad (4)$$

where $\tan \delta_E$ is the loss tangent of the composite with the biasing field and $\tan \delta_0$ is the loss tangent without the biasing field. This is so-called Quality Factor of a Tunable Component (QFTC), which is now applied to a tunable composite.

3. RESULTS

In this section two tunable composites are studied. The ferroelectric phase is SrTiO₃ in both cases. The other phase in the first case is polymer (PTFE), and in the other it is another ceramic (rutile). PTFE (also known as Teflon) was chosen because of its low losses and high breakdown voltage, reported as high as 2000 kV/cm [24] at cryogenic temperatures. Rutile has also small losses but its permittivity is higher than that of PTFE. Both composites are studied at temperature 77 K, where SrTiO₃ possesses ferroelectric behavior. Models and parameters for the used materials are presented in Appendix A.

3.1. SrTiO₃/PTFE-polymer Composite

SrTiO₃/PTFE-polymer composite consists of ceramic and polymer phases. If there are significant amounts of polymer in the mixture, the elastic properties of the composite are very different from the ferroelectric and the composite may be made mechanically flexible. In addition, since the permittivity of PTFE is low ($\varepsilon_d = 2.1$), we would be able to produce a composite with effective permittivity much smaller

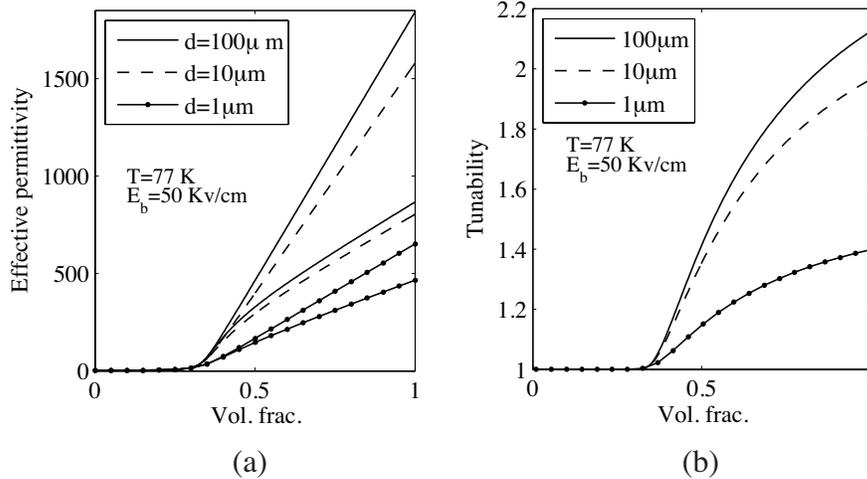


Figure 2. Effective permittivity (a) and tunability (b) of the composite as a function of volume fraction of SrTiO_3 in a PTFE-polymer matrix. In (a), the upper curves corresponds to the effective permittivity without the biasing field and the lower curves with the biasing field.

than that of SrTiO_3 . The dielectric properties and tunability of the composite are calculated using the principles of the previous chapter.

The effective permittivity as a function of volume fraction of SrTiO_3 in the composite is presented in Fig. 2(a) both for the biasing electric field strength $E_B = 50 \text{ kV/cm}$ and without the biasing field. Three different diameter sizes for the ferroelectric powder are shown. The upper curve for each inclusion size represents the effective permittivity without the biasing field. For high volume filling ratios of ferroelectric effective permittivity increases as the inclusion size increases. This is because of the size dependent model for the permittivity of the ferroelectric. The effective permittivity of the mixture with $f = 1$ gives the same value as the ferroelectric model for the same inclusion size. For low volume filling ratios the effective permittivity seems to be virtually the same as for the PTFE polymer. The permittivity starts to increase with the volume filling ratio of $1/3$, which is the percolation threshold. The permittivity of SrTiO_3 spheres is much larger than that of the PTFE. Therefore the electric field inside disconnected spheres is virtually zero, and hence the external biasing field does not change the permittivity of ferroelectric spheres.

In Fig. 2(b) the tunability for the same composites as in Fig. 2(a)

are presented with the same biasing electric field. Both the tunability and the effective permittivity depend on the size of ceramic inclusions. This is a result of the size dependence of the model for SrTiO_3 , where the permittivity is reduced for the small inclusions. The physical reason is that the transition layer where the polarization of atoms changes from minimum to the maximum value is not negligible for small inclusions.

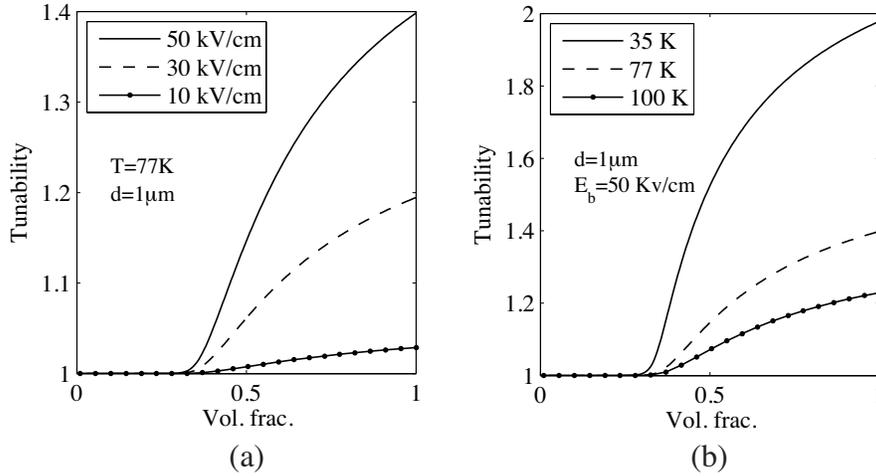


Figure 3. Tunability of the composite as a function of volume fraction of SrTiO_3 in a PTFE-polymer matrix for varying biasing fields (a) and temperatures (b).

In Fig. 3(a) the diameter for inclusions is kept constant $d = 1 \mu\text{m}$, but the tunability is shown for three different biasing fields. In Fig. 3(b), the tunability is shown with $E_B = 50 \text{ kV/cm}$ for three different temperatures. The curves show that the $\text{SrTiO}_3/\text{PTFE}$ composite is not tunable at volume filling ratios smaller than the percolation threshold of $f = 0.33$ with any parameter combinations for the temperature, biasing fields or the diameter for the ferroelectric spheres.

In Fig. 4(a) losses for the composite with different inclusion sizes are presented. Above percolation threshold, losses of the composite are close to the losses of the pure ferroelectric material. In Figs. 4(b)–5(b), all parameters are the same as in Figs. 2(b)–3(b), but now the figure of merit is displayed instead of the tunability. Figure of merit decreases more rapidly than the tunability when volume fraction of polymer increases, because losses of the composite remain close to the losses of the ferroelectric.

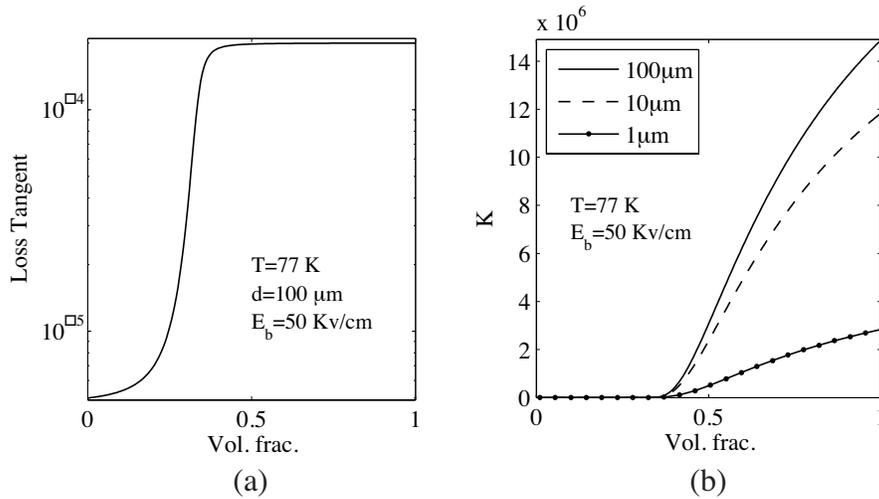


Figure 4. Loss tangent (a) and the figure of merit (b) of the composite as a function of volume fraction of SrTiO_3 in a PTFE-polymer matrix for different diameters for SrTiO_3 inclusions.

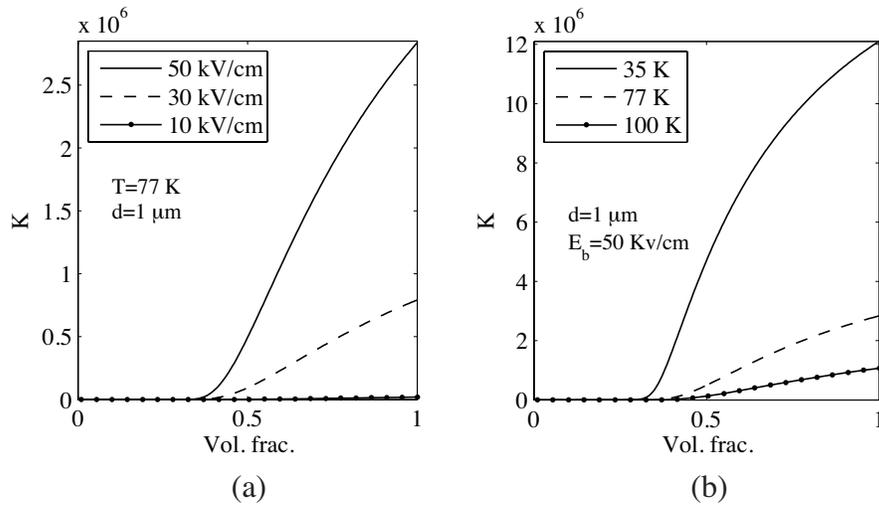


Figure 5. The figure of merit of the composite as a function of volume fraction of SrTiO_3 in a PTFE-polymer matrix for varying biasing electric field (a) and temperature (b).

3.2. SrTiO₃/rutile Composite

In this section, the same analysis is performed for the ferroelectric-rutile composites as in the previous section for PTFE-polymer composites. Rutile (TiO₂) is a ceramic material which causes restrictions with the mechanical properties of the composite as compared to the polymer matrix. However, rutile has a much higher permittivity than PTFE, which helps to achieve tunability at lower volume fractions of the ferroelectric phase. This is due to the fact that the field inside the ferroelectric inclusions becomes higher (see Eq. (2)). The low volume fraction load helps to reduce the overall effective permittivity while maintaining at the same time a reasonable tunability. Furthermore, rutile has smaller losses than the PTFE polymer.

In Fig. 6(a) the effective permittivity for the SrTiO₃/rutile composite is presented with the same parameters as for PTFE in Fig. 2(a). The electrical contrast between permittivities of rutile and ferroelectric is smaller than for PTFE and ferroelectric. Therefore the electric field can better penetrate to the ferroelectric material and the permittivity increases more gradually than for the PTFE composite.

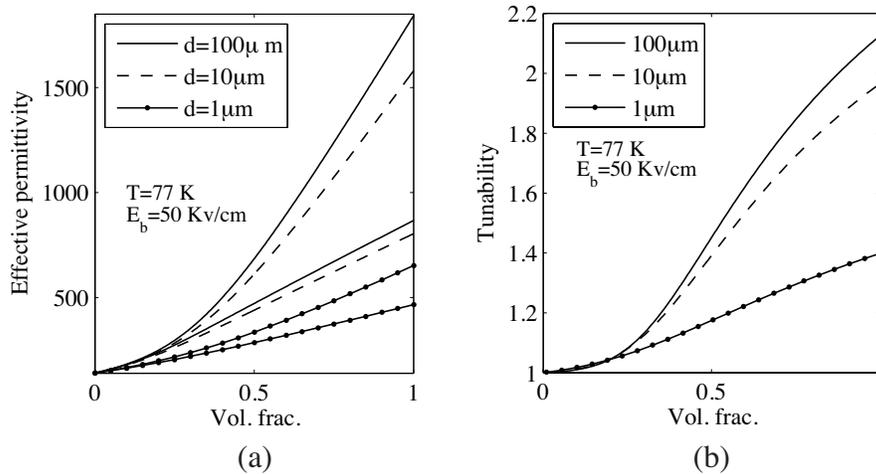


Figure 6. Effective permittivity and tunability (b) of the composite as a function of volume fraction of SrTiO₃ in the SrTiO₃/rutile composite. In (a), the upper curves corresponds to the effective permittivity without the biasing field and the lower curves with the biasing field.

The tunability is presented in Fig. 6(b)–Fig. 7(b). Unlike PTFE composite, the rutile composite is tunable with volume filling ratios

smaller than the percolation threshold $f = 0.33$. This is because smaller contrast makes the electric biasing field nonzero inside the tunable spheres.

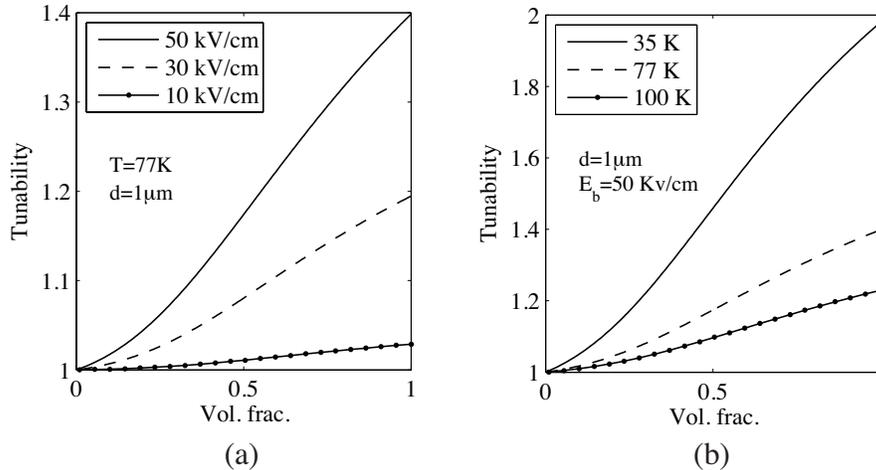


Figure 7. Tunability of the composite as a function of volume fraction of SrTiO₃ in the rutile/SrTiO₃ composite for varying biasing fields (a) and temperatures (b).

The figure of merit which is presented in Fig. 8(b)–Fig. 9(b) is also highly improved compared to the PTFE composite. This is the result of lower losses (Fig. 8(a)) and because of larger tunability than for the PTFE composite. With very low filling ratios of the tunable material in the composite, the figure of merit is very high in some cases. This happens when the losses of the composite are closer to the losses of the dielectric material, but there is some slight tunability in the composite.

4. DISCUSSION

In the previous section, two very different examples for SrTiO₃ composites were treated: the ferroelectric was mixed either with a PTFE polymer or with rutile ceramic. Tunability, effective permittivity, and the figure of merit as functions of the volume fraction of ferroelectric material were analyzed and illustrated.

The polymer composite allows the mechanical properties of the ferroelectric to be modified and the permittivity can be reduced from the high values of SrTiO₃. However, the volume filling ratio of the tunable material must be higher than the percolation threshold

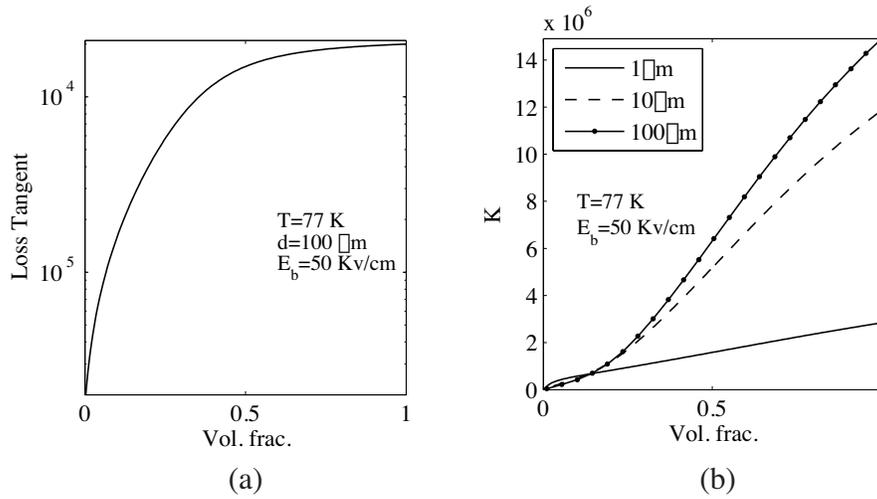


Figure 8. Loss tangent (a) and the figure of merit (b) of the composite as a function of volume fraction of SrTiO₃ in the SrTiO₃/rutile composite for different diameters for SrTiO₃ inclusions.

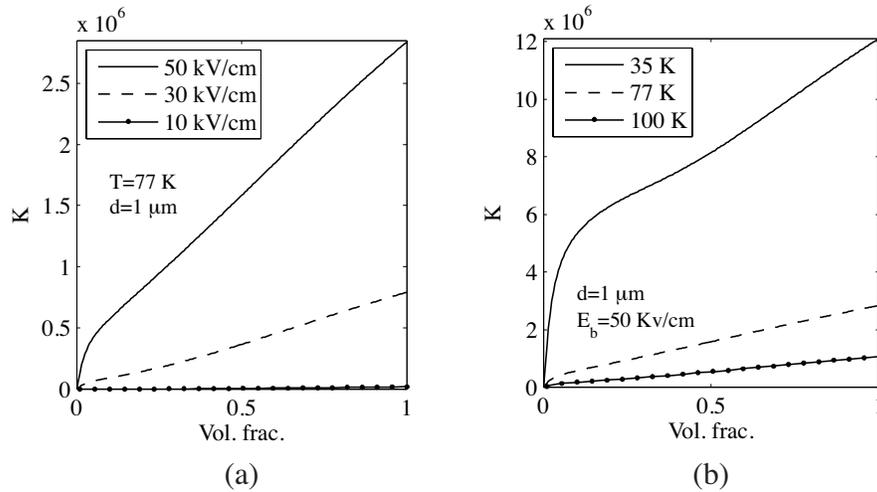


Figure 9. The figure of merit of the composite as a function of volume fraction of SrTiO₃ in the SrTiO₃/rutile composite for varying biasing electric field (a) and temperature (b).

because of the low permittivity of the polymer. If some other shape than spherical inclusions for the ferroelectric material are used, the percolation threshold will be reduced. Hence the material can be made tunable with smaller volume filling ratios of the ferroelectric phase.

On the other hand, the results showed that the rutile composite does not have such a strong percolation behavior for any of the parameters (effective permittivity, tunability, or figure of merit). This is attractive because of the small amount of ferroelectric needed while still achieving tunability. For example, the tunability of 1.1 at $T = 77\text{ K}$ and with inclusion size $10\ \mu\text{m}$ can be achieved with $f = 0.4$ volume filling ratio of SrTiO_3 in the PTFE polymer matrix or $f = 0.28$ in the rutile matrix. The effective permittivity of the mixture is 160 with PTFE and 310 with rutile. The figure of merit is $3 \cdot 10^5$ with PTFE and $2 \cdot 10^6$ with rutile. Although the effective permittivity is smaller with PTFE matrix, the figure of merit is better with rutile, thanks to the lower losses in rutile. Of course, a ceramic material with smaller permittivity and loss tangent than rutile would be even better.

It is worth noting that the analysis of the present paper can be used along with other models for the ferroelectric than the one in Appendix A. This is because Eqs. (1) and (2) remain valid, from which the effective permittivity can be solved, as long as the permittivities of tunable (ε_d) and nontunable (ε_f) phases are known. Also the behavior of the nontunable phase of the composite can be assumed to be temperature-dependent. Then the model can be applied for compensating the possible temperature dependence of the tunability. High-permittivity dielectrics might be especially suitable for temperature compensation, because already a small amount of ferroelectric can produce the effect of tunability and more freedom remains for optimization.

The effect of the inclusion size should be considered only indicative, because the model for the size-dependent permittivity of the tunable material is originally derived for thin sheets instead of granular inclusions. With the filling ratio of 100% of ferroelectric material, we would expect the permittivity and the tunability to be dependent more on the thickness of the whole ferroelectric sheet than the grain size. Accordingly, a more sophisticated model would replace the grain size d with a parameter describing the average thickness of ferroelectric phase across the sample. This would be a function, dependent on the volume filling ratio, with two limits: the grain size and the thickness of the composite layer. The interface effect on the ferroelectricity at the nanoscale have been studied in detail in [25].

The results in the present study differ from those in [14] and [26] where the tunability was not found to reduce when adding nontunable

material into the composite. There, also a strong resonance in the tunability was reported around the volume fraction of 40 percent of tunable material. In contrast, in the present study, adding nontunable material reduces the tunability. The tunability is also a continuous function without any nonphysical resonances. If nontunable spheres are added into a tunable material (high volume filling ratios in the figure), the electric field inside the spheres is enhanced, and compared with the average, the field outside the spheres (within the ceramic phase) is smaller. Hence the global tunability of the composite should be reduced.

This fact has also been confirmed by the experimental studies [4, 5] where it was found that addition of a small amount of nontunable dielectric MgO to a tunable BST or BSTO material did not increase the tunability. The addition MgO reduces the loss tangent and permittivity of the composite. It is interesting that the observed reduction in [4] was greater than the prediction in the present study. From practical viewpoint, this makes such tunable composites even more appealing.

5. CONCLUSIONS

A novel method to calculate the effective permittivity and tunability of a composite of dielectric and tunable materials is presented. The method is applicable for all mixing ratios, as long as one phase can be approximated as randomly dispersed spheres. The Bruggeman effective medium theory is combined with a model for the nonlinear permittivity of a ferroelectric material (SrTiO_3). In this study, the model presented in [14] was used, but any other model for the permittivity of a tunable material could be used as well. Because the model for the ferroelectric is a size-dependent, also the effective permittivity of the tunable composite depends on the size of tunable inclusions. Both the effective permittivity and the tunability decreases when the size of tunable inclusions decreases.

As examples, two different composites, SrTiO_3 /PTFE-polymer and SrTiO_3 / TiO_2 -rutile, were studied. If the permittivity of the nontunable dielectric phase is low (PTFE), the composite is tunable with volume filling ratios of the tunable material larger than the percolation threshold. When the dielectric phase has large permittivity (TiO_2), the tunability of the composite increases gradually as a function of volume filling ratio of tunable material in the composite. Hence, in that case the composite is tunable also below the percolation threshold. With high permittivity dielectric combined with ferroelectric phase, a tunable material with almost any effective

permittivity between the permittivities of tunable and nontunable phases can be achieved.

ACKNOWLEDGMENT

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APPENDIX A.

A.1. Permittivity of the Ferroelectric

The model for the relative permittivity of SrTiO₃, used in this study, has been taken from [14]. It is frequency-independent and reported to be applicable up to frequency about 100 GHz. However, when applied to a granular composite material, the high-frequency limit is also determined by the grain size. The wavelength should be much larger than the scale of inhomogeneities in the material in order to avoid enhancement of scattering losses.

The real part of the relative permittivity of a ferroelectric depends on the temperature and the biasing electric field inside the material [20]

$$\varepsilon_{f,r}(T, E_b) = \varepsilon_{00}([(\xi^2 + \eta^3)^{1/2} + \xi]^{2/3} + [(\xi^2 + \eta^3)^{1/2} - \xi]^{2/3} - \eta + a^2)^{-1} \quad (\text{A1})$$

where ε_{00} is the analog of the Curie-Weiss constant and parameter ξ is a function of biasing electric field, parameter η is a function of the temperature, and a is a parameter related to the thickness of the ferroelectric layer. The parameter ξ is defined

$$\xi = \sqrt{\xi_B^2 + \xi_S^2} \quad (\text{A2})$$

where ξ_S is the dispersion parameter of the material characterizing the density of defects of the material and ξ_B is the normalized biasing voltage

$$\xi_B = E_i/E_N \quad (\text{A3})$$

where E_i is the biasing electric field in the material and E_N is a normalization constant of the material. If the ferroelectric material is mixed with dielectric material, the bias field E_i that excites the particles is calculated with the Bruggeman effective medium theory, Eq. (2). The temperature parameter η is defined

$$\eta = (\Theta_F/T_C) \sqrt{1/16 + (T/\Theta_F)^2} - 1 \quad (\text{A4})$$

where T is the temperature of the ferroelectric, T_C is the Curie temperature, Θ_F is analog to the Debye temperature of sublattice oscillations that are responsible for the ferroelectric polarization.

Near the surface of the ferroelectric layer, the polarization changes gradually from the maximum value towards the minimum. This transition layer has an effect to the permittivity of a thin ferroelectric sheet. This can be taken into account with parameter a :

$$a = \sqrt{2\epsilon_{00}/\alpha h} \quad (\text{A5})$$

where the parameter α is related to the dispersion of the longitudinal and transverse optical type oscillations of the crystal lattice and h is the thickness of the ferroelectric layer.

In this study, spherical inclusions were considered. Therefore instead of the thickness of the layer, the diameter of the inclusions d was used in (A5): $h = d$. Although this is a rough estimate, it should be a better estimate than to use the model for an infinitely thick layer. According to [20], the reduction in the permittivity of thin material layer is caused by the different response of atoms in the surface layer to the electric field. In other words, the atoms near the surface have a weaker polarization than atoms deep in the material in the same excitation field. This transition layer should exist also for spherical inclusions. Since here the model of planar transition layer is used for spherical inclusions, the results concerning the effect of the inclusion size should be considered only indicative.

For SrTiO₃, the following modeling parameters were used: $T_m = 2$ K, $\xi_S = 0.018$, $T_c = 42$ K, $\Theta_F = 175$ K, $\epsilon_{00} = 2081$, $E_N = 19.3$ kV/cm and $\alpha = 2 \cdot 10^9$ 1/m [14]. The model for the permittivity of SrTiO₃ does not include losses. Therefore a constant loss tangent of $\tan\delta_f = 2 \cdot 10^{-4}$ was used [27].

A.2. Permittivities of Nontunable Dielectrics

The permittivity of the nontunable phase and loss tangent are assumed to be constant to reduce the amount of different parameters and to highlight the effect of the tunable phase. However, the model is valid for more complicated permittivities, such as frequency and temperature-dependent permittivity of the dielectric.

The permittivity of the rutile is calculated from the polycrystal model [28]

$$\epsilon_d(\text{TiO}_2) = \frac{1}{3}\epsilon_z + \frac{2}{3}\epsilon_t - \frac{2}{9} \frac{(\epsilon_z - \epsilon_t)^2}{\epsilon_z + 2\epsilon_t} \quad (\text{A6})$$

where ϵ_z and ϵ_t are the anisotropic permittivity values from [27]. The polycrystal model is a better assumption than a single-crystal model,

because the SrTiO₃/rutile composite is compressed using ceramic powders. Anisotropic permittivity values $\epsilon_t = 170$ and $\epsilon_z = 240$ [27] give $\epsilon_d(\text{TiO}_2) = 141$ for rutile. For the PTFE phase, the value of $\epsilon_d(\text{PTFE}) = 2.1$ [29] was used. At cryogenic temperatures the loss tangent of PTFE is $\tan\delta_d(\text{PTFE}) = 5 \cdot 10^{-6}$ [29], and the loss tangent of rutile is $\tan\delta_d(\text{TiO}_2) = 1.7 \cdot 10^{-6}$ [27].

REFERENCES

1. Guérin, F., "Microwave chiral materials: a review of experimental studies and some results on composites with ferroelectric ceramic inclusions," *Progress In Electromagnetics Research*, PIER 9, 219–263, 1994.
2. Huang, C.-C., "Analysis of multiconduction transmission lines with nonlinear terminations in frequency domain," *J. of Electromagn. Waves and Appl.*, Vol. 19, No. 8, 1069–1083 2005.
3. Wang, X.-H. and B.-Z. Wang, "Generalized transmission line theory for parallel planar transmission lines," *J. of Electromagn. Waves and Appl.*, Vol. 19, No. 9., 1171–1181, 2005.
4. Sengupta, L. C. and S. Sengupta, "Breakthrough advantages in low loss, tunable dielectric materials," *Mat. Res. Innovat.*, Vol. 2, 278–282, 1999.
5. Chen, Y., X. Dong, J. Li, and Y. Wang, "Dielectric properties of Ba_{0.6}Sr_{0.4}TiO₃/Mg₂SiO₄/MgO composite ceramics," *J. Appl. Phys.*, Vol. 98, 064107, 2005.
6. Tummala, R., "Ceramic and glass-ceramic packaging in the 1990s," *J. Am. Cer. Soc.*, Vol. 74, 895–908, 1991.
7. Barnwell, P., W. Zhang, J. Lebowitz, K. Jones, N. MacDonald, C. Free, and Z. Tian, "An investigation of the properties of LTCC materials and compatible conductors for their use in wireless applications," *Proc. International Symposium on Microelectronics*, 659–664, Boston, MA, 2000.
8. Hakeem, N. A., H. I. Abdelkader, N. A. El-sheshtawi, and I. S. Eleshmawi, "Spectroscopic, thermal and electrical investigations of PVDF films filled with BiCl₃," *J. Appl. Pol. Sci.*, Vol. 102, 2125–2131, 2006.
9. Xu, H., J. Zhong, X. Liu, J. Chen, and D. Shen, "Ferroelectric and switching behavior of poly(vinylidene fluoride-trifluoroethylene) copolymer ultrathin films with polypyrrole interface," *Appl. Phys. Lett.*, Vol. 90, 092903, 2007.
10. Shynu, S. V., G. Augustin, C. K. Aanandan, P. Mohanan, and K. Vasudevan, "Design of compact reconfigurable dual

- frequency microstrip antennas using varactor diodes,” *Progress In Electromagnetics Research*, PIER 60, 197–205, 2006.
11. Zheng, Q.-R., B.-Q. Lin, Y.-Q. Fu, and N.-C. Yuan, “Characteristics and applications of a novel compact spiral electromagnetic band-gap (EBG) structure,” *J. of Electromagn. Waves and Appl.*, Vol. 21, No. 2, 199–213, 2007.
 12. Lee, S.-W., Y. Kuga, and A. Ishimaru, “Quasi-static analysis of metamaterials with small tunable stacked split ring resonators,” *Progress In Electromagnetics Research*, PIER 51, 219–229, 2005.
 13. Irvin, P., J. Levy, R. Guo, and A. Bhalla, “Three-dimensional polarization imaging of (Ba,Sr)TiO₃:MgO composites,” *Appl. Phys. Lett.*, Vol. 86, 042903, 2005.
 14. Astafiev, K. F., V. O. Sherman, A. K. Tagantsev, and N. Setter, “Can the addition of a dielectric improve the figure of merit of a tunable material?” *J. Eur. Cer. Soc.*, Vol. 23, 2381–2386, 2003.
 15. Mokry, P., A. K. Tagantsev, and N. Setter, “Size effect on permittivity in ferroelectric polydomain thin films,” *Phys. Rev. B*, Vol. 70, 172107, 2004.
 16. Sherman, V. O., A. K. Tagantsev, N. Setter, D. Iddles, and T. Price, “Ferroelectric-dielectric tunable composites,” *J. Appl. Phys.*, Vol. 99, 074104, 2006.
 17. Stround, D. and P. M. Hui, “Nonlinear susceptibilities of granular matter,” *Phys. Rev. B*, Vol. 37, 8719–8724, 1988.
 18. Stround, D. and V. E. Wood, “Decoupling approximation for the nonlinear-optical response of composite media,” *J. Opt. Soc. Am. B*, Vol. 6, 778–786, 1989.
 19. Bergman, D. J., “Nonlinear behavior and 1/f noise near a conductivity threshold: Effects of local microgeometry,” *Phys. Rev. B*, Vol. 39, 4589–4609, 1989.
 20. Vendik, O. G. and S. P. Zubko, “Modeling the dielectric response of incipient ferroelectrics,” *J. Appl. Phys.*, Vol. 82, 4475–4483, 1997.
 21. Garboczi, E. J., K. A. Snyder, and J. F. Douglas, “Geometrical percolation threshold of overlapping ellipsoids,” *Phys. Rev. E*, Vol. 52, 819–828, 1995.
 22. Sihvola, A., *Electromagnetic Mixing Formulas and Applications*, IEE Electromagnetic Waves Series 47, The Institution of Electrical Engineers, 1999.
 23. Vendik, I. B., O. G. Vendik, and E. L. Kollberg, “Commutation quality factor of two-state switchable devices,” *IEEE Trans. on Micr. Theor. and Techn.*, Vol. 48, 802–808, 2000.

24. Chowdhuri, P., T. Bement, C. Espinoza, and G. Weeks, "DC break down strength of dielectric materials at cryogenic temperatures," *Proc. of 7th IEEE/PES Transmission and Distribution Conference and Exposition*, 140–147, 1979.
25. Duan, C., R. F. Sabirianov, W. Mei, S. S. Jaswal, and E. Y. Tsymbal, "Interface effect on ferroelectricity at the nanoscale," *Nano Letters*, Vol. 6, 483–487, 2006.
26. Sherman, V. O., A. K. Tagantsev, and N. Setter, "Tunability and loss of the ferroelectric-dielectric composites," *IEEE International Ultrasonics, Ferroelectrics, and Frequency Control joint 50th Anniversary Conference*, 33–38, 2004.
27. Gallop, J. and L. Hao, "Single crystal microwave dielectrics at low temperature: losses and non-linearities," *J. Eur. Cer. Soc.*, Vol. 23, 2367–2373, 2003.
28. Herring, C., *J. Appl. Phys.*, Vol. 31, 1939, 1960.
29. Jacob, V., J. Mazierska, K. Leong, and J. Krupka, "Microwave properties of low-loss polymers at cryogenic temperatures," *IEEE Trans. Micr. Theor. Techn.*, Vol. 50, 474–480, 2002.