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Can the addition of a dielectric improve the figure of merit of a tunable material?

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Abstract

The influence of addition of a low-loss linear dielectric material to a tunable ferroelectric material has been investigated in terms of the electrostatic consideration. The calculations of the dielectric loss and dielectric non-linearity of ferroelectric-dielectric composites have been performed by using three different models. On the basis of results obtained, the figure of merit of the composite material has been evaluated. No improvement of the figure of merit of composite material compared to the pure ferroelectric has been observed for the considered models.

Keywords: Composites; Ferroelectrics; Quality factor; Tunable microwave devices

1. Introduction

Perovskite ferroelectrics like $SrTiO_3$ (STO) and $(Ba_xSr_{1\text{-}x})TiO_3$ (BSTO) make a group of materials attractive for application in microwave tunable devices for wireless communication systems.[1] These materials are characterized by tunability n:

$$n = \frac{\varepsilon(0)}{\varepsilon(E_{\max})}, \tag{1}$$

where $\varepsilon(0)$ and $\varepsilon(E_{\max})$ are the dielectric constants of the material at zero and at maximum bias field, respectively, and by the loss tangent ($\tan\delta$). Unfortunately, the loss tangent of ferroelectrics is not negligibly small as it is common for low permittivity microwave dielectrics. The loss tangent and tunability are dependent on the absolute value of the dielectric permittivity. Typically, the higher the dielectric permittivity, the higher the tunability and loss tangent. To reduce the loss and dielectric permittivity, attempts have been recently made to make composite materials as a mechanical mixture of non-tunable low loss and low permittivity dielectrics with ferroelectrics.[2,3]

Mixing a tunable ferroelectric with a linear dielectric modifies the electrical properties of the material due to two effects: first, the effect of mutual doping of components of the composite, and, second, the effect of the electrical field redistribution between different components of the mixture. In this paper we address the latter effect, which can be treated in terms of the electrostatic consideration. Specifically, we present results of modeling the dielectric loss and dielectric non-linearity of ferroelectric–dielectric composites performed taking into account only the effect of the field redistribution. Finally, on the basis of results obtained, we calculate the figure of merit of such kind of composite materials. We compared the figure of merit of three types of composites and that of the pure ferroelectric to find no increase of the figure of merit in the case of composites.

2. Simulation procedure

The dielectric properties of the bi-component composites have been theoretically addressed for a long time.[4–6] Basically, in most of the works, three main models of the component distribution are considered: (i) the composite represents a layered structure with layers of two components connected in series (Fig. 1a) so that the external electric field E applied to the composite is perpendicular to the interphase boundaries; (ii) the composite represents a columnar structure of different phases connected in parallel (Fig. 1b) so that the external electric field E is parallel to the interphase boundaries; and (iii) in the composite, one phase is embedded into another as spheres (Fig. 1c).

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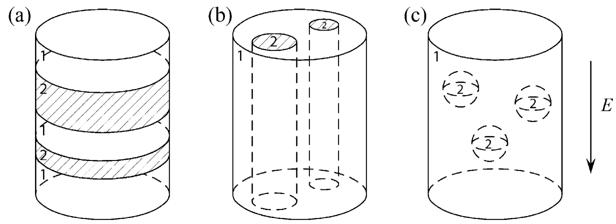

Fig. 1. Schematic of models of bi-component composite: (a) layered model, (b) columnar model, and (c) spherical inclusion model. 1, ferroelectric component; 2, dielectric component.

In this work, we will consider a bi-component ferroelectric–dielectric composite which consists of a high dielectric constant tunable ferroelectric material and of a low dielectric constant, low loss linear dielectric material. On the basis of models mentioned above we will evaluate the microwave performance of such kind of a composite material.

Let's consider first the layered model (Fig. 1a). In fact, this case can be presented as the in-series connection of several capacitors corresponding to different layers of the composite components. One can easily show that electrically this system is equivalent to the in-series connection of two capacitors corresponding to the two components of the mixture, so that the effective dielectric permittivity of the composite can be found as:

$$\frac{1}{\varepsilon_{\text{mix}}(q)} = \frac{1-q}{\varepsilon_{\text{f}}} + \frac{q}{\varepsilon_{\text{d}}}, \tag{2}$$

where q is the volume concentration of the dielectric phase in the composite, ε_{mix}, ε_{f}, and ε_{d} are the dielectric constants of the composite, ferroelectric and dielectric, respectively.

The field dependence of the composite dielectric constant $\varepsilon_{\text{mix}}(q, E)$ originates from that of the dielectric constant $\varepsilon_{\text{f}}(E_{\text{f}})$ of the ferroelectric component, where E_{f} is the electric field applied to the ferroelectric. This dependence can be found from Landau's theory.[7] It is known that the dielectric constant of the ferroelectric materials like STO and BSTO can be presented in the form:

$$\varepsilon_{\text{f}}^{-1}(E_{\text{f}}) = \varepsilon_0\alpha + 3\beta\varepsilon_0 P_{\text{f}}^2(E_{\text{f}}) \tag{3}$$

where $\varepsilon_0 = 8.85 \times 10^{-12}$ F/m is the permittivity of free space,$\varepsilon_{\text{f}}(0) = 1/\varepsilon_0\alpha$ is the dielectric constant of the ferroelectric in the absence of external electric field, α and β are coefficients of the free energy expansion in Landau's theory. $P_{\text{f}}(E_{\text{f}})$ is the dc field induced polarization which can be found by solving the following equation:[7]

$$\beta P_{\text{f}}^3(E_{\text{f}}) + \alpha P_{\text{f}}(E_{\text{f}}) - E_{\text{f}} = 0. \tag{4}$$

One should note that a variation of the volume concentration q of the dielectric material leads to a redistribution of electric field between the components of the composite.

To find the dielectric constant of the composite material as a function of the applied field E and concentration q one should append Eqs. (2)–(4) with the condition of continuity of electrical displacement:

$$\varepsilon_0\varepsilon_{\text{b}}E_{\text{f}}(q) + P_{\text{f}}(E_{\text{f}}) = \varepsilon_0\varepsilon_{\text{d}}E_{\text{d}}(q), \tag{5}$$

where ε_{b} is the background dielectric constant, and the equation for the voltage drops across the composite components:

$$E_{\text{f}}(q)(1-q) + E_{\text{d}}(q)q = E. \tag{6}$$

Using Eqs. (2)–(6) the solution of the problem can be presented in the form:

$$\beta P_{\text{f}}^3(E_{\text{f}}) + \alpha^* P_{\text{f}}(E_{\text{f}}) - E_{\text{f}}^* = 0, \tag{7}$$

$$\begin{cases} \alpha^* = \alpha + \dfrac{q}{\varepsilon_0\varepsilon_{\text{b}}q + \varepsilon_0\varepsilon_{\text{d}}(1-q)} \\ E_{\text{f}}^* = E\dfrac{\varepsilon_0\varepsilon_{\text{d}}}{\varepsilon_0\varepsilon_{\text{b}}q + \varepsilon_0\varepsilon_{\text{d}}(1-q)} \end{cases}, \tag{8}$$

$$\varepsilon_{\text{mix}}^{-1}(q, E) = \varepsilon_0\alpha^* + 3\beta\varepsilon_0 P_{\text{f}}^2 - \frac{q^2\left[\alpha\varepsilon_0(\varepsilon_{\text{b}} - \varepsilon_{\text{d}}) - \dfrac{\varepsilon_{\text{b}}}{\varepsilon_{\text{d}}} + 1\right]}{\varepsilon_{\text{b}}q + \varepsilon_{\text{d}}(1-q)} - \frac{q\alpha\varepsilon_0\varepsilon_{\text{d}}}{\varepsilon_{\text{b}}q + \varepsilon_{\text{d}}(1-q)} - 3q\beta\varepsilon_0 P_{\text{f}}^2. \tag{9}$$

It is seen that, in the limit of small concentration of the dielectric material in the mixture ($q < < 1$) and when $\varepsilon_{\text{f}}/\varepsilon_{\text{d}} >> 1$, the last three terms in the Eq. (9) can be neglected and the renormalized electric field E^* in Eq. (8) becomes equal to the electric field E. Thus, Eqs. (7) and (9) become identical to the equation for the permittivity and polarization of the pure ferroelectric material [Eqs. (3) and (4)], but with the coefficient α replaced by α*.

One can easily see that, in the case of the layered model, the behavior of the composite is equal to that of the pure ferroelectric whose coefficient α is increased, which corresponds to a decrease of the Curie–Weiss temperature of the material.

To calculate the microwave performance of our composite material we still need to calculate the loss tangent of the mixture. For that let's use the equation for the energy dissipation in dielectric media. For our bi-component media one can write:

$$W_f + W_d = W_{mix}, \tag{10}$$

where W_f, W_d, and W_{mix} are the energy dissipated in ferroelectric part of the mixture, in dielectric part of the mixture, and the total dissipated energy in whole composite, respectively.

It is known that energy dissipated in dielectric media is proportional to the imaginary part of the complex dielectric constant of the media ε'':[8]

$$W = \frac{\omega}{2}\varepsilon''\langle\tilde{E}\rangle^2 V, \tag{11}$$

where ω is the circular frequency, $\langle\tilde{E}\rangle$ is the average amplitude of ac field applied to the media, and V is the volume of this dielectric media.

Combining Eqs. (11) and (10), and taking into account that the volumes of the dielectric layers in our model (Fig. 1a) are directly proportional to the concentration of the components, we find:

$$\frac{\omega}{2}\varepsilon''_f\tilde{E}_f^2(1-q)V + \frac{\omega}{2}\varepsilon''_d\tilde{E}_d^2 qV = \frac{\omega}{2}\varepsilon''_{mix}\langle\tilde{E}\rangle^2 V \tag{12}$$

where the $\tilde{E}_f$ and $\tilde{E}_d$ are the amplitudes of ac fields in ferroelectric and dielectric, respectively.

By rewriting Eq. (12) through the loss tangent $\tan\delta = \varepsilon''/\varepsilon'$, and taking into account the continuity of electrical displacements [Eq. (5)] we obtain the equation for the loss tangent of the composite:

$$\tan\delta_{mix}(q, E) = \frac{\tan\delta_f(E_f)\varepsilon_d(1-q) + \tan\delta_d\varepsilon_f(E_f)q}{(1-q)\varepsilon_d + \varepsilon_f(E_f)q}. \tag{13}$$

To find the loss tangent of the composite under applied dc electric field, we should specify first the field dependence of the loss tangent of the ferroelectric component. For the moment, the theory of the dc field dependence of the dielectric loss is developed only for the case of the intrinsic loss mechanisms.[9,10] It predicts an increasing field dependence of the loss tangent as typical behavior in the case of weak and moderate field, which was experimentally documented for high quality single crystals.[1] As for material of lower quality like ceramics and films, to which the present paper is addressed, in these, the dc field dependence of the loss is clearly controlled by extrinsic mechanisms. In this case, according to experimental results, the typical trend is an increase of the loss tangent with increasing dielectric permittivity of the material (see e.g. Ref. 11). To model this situation we assume that the loss tangent of the ferroelectric material $\tan\delta_f(E)$ is proportional to its dielectric permittivity $\varepsilon_f(E)$. In other word for the loss tangent of the ferroelectric component we set:

$$\tan\delta_f(E) = \tan\delta_f(0)\frac{\varepsilon_f(E)}{\varepsilon_f(0)}. \tag{14}$$

For the loss tangent of the dielectric part of the mixture $\tan\delta_d$, we assume that it does not depend on electric field.

Finally, by using Eq. (13), (3) and taking into account Eqs. (7)–(9) we can calculate the tunability [Eq. (1)] and loss tangent of the layered composite.

Let's now consider the second model: the columnar structure shown in Fig. 1b. Clearly, electrically, this system is equivalent to the parallel connection of two capacitors corresponding to the two components of the mixture and effective dielectric permittivity of the composite can be presented as:

$$\varepsilon_{mix}(q) = \varepsilon_f(1-q) + \varepsilon_d q. \tag{15}$$

In the case of parallel connection of the composite components, the electric field is the same in all parts of the composite, being equal to the applied field E. That means that to find the field dependence of the composite dielectric constant we can simply use Eq. (15) where ε_f is given by Eqs. (3) and (4) with electric field $E_f = E$.

To calculate loss tangent we can also use the same formula as before [Eq. (12)] and taking into account that the electric field in all parts of the composite is the same ($\langle\tilde{E}\rangle = \tilde{E}_f = \tilde{E}_d$) for the imaginary part of dielectric constant of the mixture one can write:

$$\varepsilon''_{mix}(q, E) = \varepsilon''_f(E)(1-q) + \varepsilon''_d q. \tag{16}$$

By rewriting Eq. (16) through the loss tangent we obtain:

$$\tan\delta_{mix}(q, E) = \frac{\tan\delta_f(E)\varepsilon_f(E)(1-q) + \tan\delta_d\varepsilon_d q}{(1-q)\varepsilon_f(E) + \varepsilon_d q}. \tag{17}$$

Under electric field applied again we assume that loss tangent of the ferroelectric part is defined by Eq. (14), and loss tangent of the dielectric part $\tan\delta_d$ does not depend on electric field.

The spherical inclusion model consists of a random distribution of spheres formed of a low loss and low dielectric permittivity material in a ferroelectric host medium (Fig. 1c). The well-known electrostatic solution of the problem for a dielectric sphere embedded into a dielectric medium gives the electric field distribution in the sphere and in the medium. This solution also enables calculation of the effective dielectric permittivity of a two-component material in the limit of low concentration of the spherical inclusions ($q << 1$).[4] For the dielectric–ferroelectric bi-component system one can write for dielectric constant of the mixture ε_{mix} and electric field in the ferroelectric matrix $\vec{E}_f$:[12]

$$\varepsilon_{mix}(q) = \varepsilon_f + 3q\varepsilon_f\frac{\varepsilon_d - \varepsilon_f}{\varepsilon_d + 2\varepsilon_f}, \tag{18}$$

$$\vec{E}_f(\vec{r}) = \vec{E} + \sum_i^N \frac{\varepsilon_d - \varepsilon_f}{\varepsilon_d + 2\varepsilon_f}\frac{R^3}{(\vec{r} - \vec{r}_i)^3}\vec{G}\vec{E}, \tag{19}$$

where R is the radius of the dielectric inclusions, N is the total number of the dielectric spheres distributed in the composite, and $\vec{r}_i$ is the radius-vectors pointing to the center of the ith spherical inclusion.

In the limit where $\varepsilon_d << \varepsilon_f$ these expressions can be reduced to:

$$\varepsilon_{mix}(q) = \varepsilon_f\left(1 - \frac{3}{2}q\right), \quad (20)$$

$$\vec{E}_f(\vec{r}) = \vec{E} - \sum_i^N \frac{R^3}{(\vec{r} - \vec{r}_i)^3}\overleftrightarrow{G}\vec{E}. \quad (21)$$

This solution takes into account a non-uniform electric filed distribution in the host material but does not take into account its dielectric nonlinearity. This dependence makes the solution of the problem more complicated. However, in the cases of small relative tunability, $(n-1) << 1$ (a typical situation in bulk ceramics), one can show that when calculating the dielectric nonlinearity of the composite, the second term in Eq. (21) can be neglected so that in the ferroelectric matrix one can set $\vec{E}_f \approx \vec{E}$. Thus, to calculate the effective dielectric permittivity at non-zero dc electric field we can still use Eq. (20), where $\varepsilon_f = \varepsilon_f(E)$.

The loss tangent of the composite in the spherical inclusion model can be calculated on the same lines as in the previous case. A difference in calculation is that now the ac field seen by the ferroelectric is inhomogeneous [see Eq. (21)]. The energy dissipated in the ferroelectric reads:

$$W_f = \frac{\omega\varepsilon_f''}{2}\int_{V_f}\tilde{E}_f^2 dV. \quad (22)$$

Taking into account the non-uniform electric filed distribution in the host material (Eq. (21)) one can calculate the energy dissipated in the ferroelectric by using Eq. (22) and integrating the electric field in ferroelectric matrix over the volume of ferroelectric $\tilde{E}_f$ over the volume of ferroelectric V_f. Then, neglecting the loss in the dielectric inclusions compared to that in the matrix and using Eq. (12), in the limit of small concentration of dielectric inclusions q one can obtain the imaginary part for the permittivity of this composite:

$$\varepsilon''_{mix}(q, E) \approx \varepsilon''_f(E)\left(1 - \frac{3}{2}q\right). \quad (23)$$

Then, finally, the loss tangent of the composite in spherical inclusion model can be presented in the form:

$$\tan\delta_{mix}(q, E) \approx \tan\delta_f(E). \quad (24)$$

One can see that, for the spherical inclusion model in the limit of small concentration of dielectric inclusions q the behavior of loss tangent is similar to that in the columnar model and one can expect no gain in dielectric loss of tunable composite materials by adding the low loss dielectric into the mixture. In the same time, in according to Eq. (20) one can expect a strong decrease of relative dielectric permittivity of the composite at the same level of additives in the mixture.

3. Results of simulation

In this section we present results of modeling the dielectric response of the composite material representing the mechanical binary mixture of the following components:

The performance of tunable microwave components is convenient to characterize by using the so-called Quality Factor of a Tunable Component (QFTC) offered by Vendik:[13]

$$K = \frac{(n-1)^2}{n\tan\delta(0)\tan\delta(E_{max})}, \quad (25)$$

where n is a tunability defined above, $\tan\delta(0)$ and $\tan\delta(E_{max})$ are the loss tangents at zero and at maximum bias field, respectively.

We have calculated this quality factor according to three models described above for dielectric–ferroelectric composite whose characteristics are presented in Table 1.

Fig. 2 represents the calculated values of the composite dielectric constant (ε_{mix}) and electrical tunability (n_{mix}) as functions of concentration of the linear dielectric material in the mixture q (Fig. 2a) and dielectric constant of the mixture ε_{mix} (Fig. 2b), respectively.

The loss tangent of the composite material $\tan\delta_{mix}$ as a function of dielectric constant of the mixture ε_{mix} is presented at Fig. 3.

The calculated Quality Factor of a Tunable Component (QFTC) [Eq. (25)] is presented in Fig. 4.

From Fig. 2a it is seen that according to the considered models the addition of a dielectric material to the mixture leads to the decreasing of the dielectric constant of the composite. At the same time, the tunability of the ferroelectric–dielectric composite also decreases (Fig. 2b): stronger in the case of layered model;

Table 1
Characteristics of the composite components

Material:	Dielectric constant ε		Loss tangent ($\tan\delta$) at $E=0$
	$E=0$	$E=7.3$ kV/cm	
Ferroelectric ($\beta = 7.37 \times 10^9 JC^{-4}m^{-5}$)	3300	2640	1×10^{-2}
Linear dielectric	8.4	8.4	1×10^{-4}

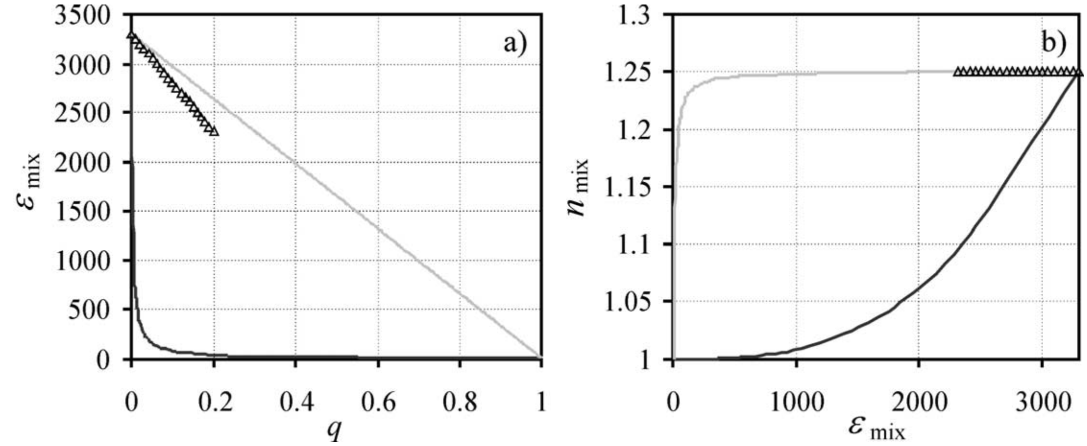

Fig. 2. (a) Calculated dielectric constant of the composite ε_{mix} as a function of concentration q; (b) tunability of the composite n_{mix} as functions of its dielectric constant ε_{mix}. Black lines, layered model; gray lines, columnar model; white triangles, spherical inclusion model.

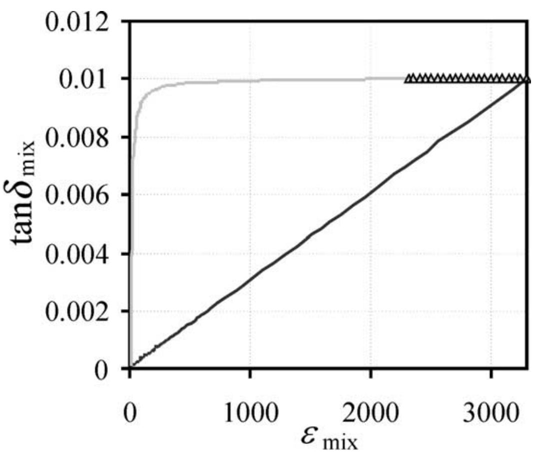

Fig. 3. Calculated loss tangent of composite, $\tan\delta_{mix}$, as a function of its dielectric constant ε_{mix}. Black lines, layered model; gray lines, columnar model; white triangles, spherical inclusion model.

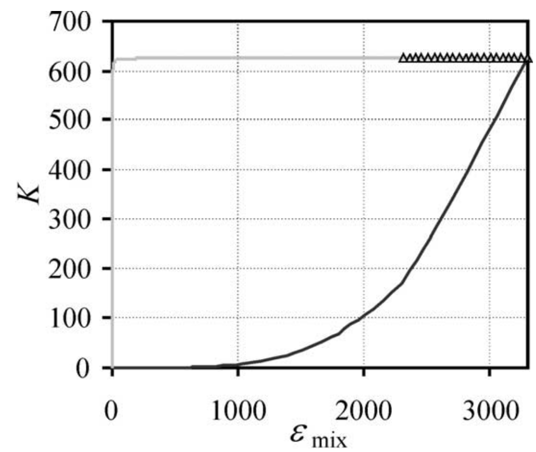

Fig. 4. Calculated quality factor of composite, K, as a function of its dielectric constant ε_{mix}. Black lines, layered model; gray lines, columnar model; white triangles, spherical inclusion model.

does not change in a wide range of the dielectric concentration q and then sharply decreases in the case of columnar model; and remains constant in spherical inclusion models (within the limits of the model applicability).

The behavior of loss tangent of the composite is different in different models (Fig. 3): in the layered model, the loss tangent linearly decreases with the dielectric constant of the composite; in the case of the columnar model, the loss tangent does not change in a wide range of the dielectric constant of the mixture and then substantially decreases; and finally, in the third model, in the limit of low concentrations of the inclusions q the behavior of loss tangent is similar to that in the columnar model (see Eq. (17) and Eq. (24)). Thus, for small q, in the columnar and spherical inclusion models the loss tangent of the composite does not depend on the amount of dielectric inclusions in the mixture whereas the relative dielectric permittivity of the composite substantially decreases at the same level of concentration of dielectric in the mixture q.

From Fig. 4 we can see that, the quality factor K decreases with decreasing dielectric constant of the composite (increasing concentration of the dielectric q) for the layered and columnar models. For the spherical inclusion model the quality factor K does not change in the limit of low concentrations of the dielectric inclusions q as well as in the columnar model. Thus, according to our simulation no improvement of the quality factor K is expected due the addition of a low-loss dielectric to a tunable ferroelectric material.

4. Conclusions

The impact of addition of a low-loss linear dielectric material on a tunable ferroelectric material has been investigated in terms of the electrostatic consideration.

Three different models (layered, columnar, and spherical inclusion models) have been used for calculation of the dielectric response of the dielectric–ferroelectric composite material. It has been shown that according to considered models the addition of a dielectric material to the mixture leads to a decrease of the dielectric constant of the composite in three considered models.

In the limit of small concentrations of the dielectric additives q, it has been found that the tunability n of composite material decreases with amount of additives q in the layered model and almost does not change in the columnar and spherical inclusion models which show a similar to each other behavior. The loss tangent of composite has shown a behavior similar that of tunability: a decreasing concentration dependence in the case of the layered model and virtually no concentration dependence in the case of the columnar and spherical inclusion models. The evaluation of Vendik's quality factor of a tunable component, Eq. (25), has shown that it does not increase with introduction of a low-loss dielectric in a tunable ferroelectric material. One should also note that in the spherical inclusion model which is most closer to real composite materials, at the level of additives q where one can already observe a strong drop of the dielectric constant of the mixture, the tunability, loss tangent, and consequently Vendik's quality factor K of the tunable composite remains almost constant. Thus, we see that, at least due to the effect of the field redistribution considered in this paper, one cannot improve the performance of ferroelectric material in a tunable composite. However, one cannot exclude a possible advantage of using the tunable components based on composite materials with the same quality factor K as pure ferroelectric material but with a lower value of dielectric constant an increase of K caused by an addition of a dielectric in the ferroelectric, which is due to a kind of doping effect, e.g. due to a doping induced reduction of the loss in the ferroelectric matrix. All these facts enable us to conclude that any, from the point of view of Vendik's quality factor K, processing of ferroelectric/dielectric composite for applications in tunable ferroelectric devices should have no advantage compared to a simple doping of the material.

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