Temperature dependence of the complex effective piezoelectric coefficient of ferroelectric 0-3 composites

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Temperature dependence of the complex effective piezoelectric coefficient $d_{31}^{\ast}$ for a ferroelectric 0-3 composite of small ceramic volume fraction has been studied. Theoretical predictions are based on our previously derived explicit expression of $d_{31}$ for a dilute dispersion of spherical particles in a continuous matrix [C. K. Wong, Y. M. Poon, and F. G. Shin, Ferroelectrics 264, 39 (2001); J. Appl. Phys. 90, 4690 (2001)]. Comparison is made with the well-known Furukawa’s model and their experimental measurements on a lead zirconate titanate (PZT)/epoxy composite with 13 vol % PZT [T. Furukawa, K. Fujino, and E. Fukada, Jpn. J. Appl. Phys. 15, 2119 (1976)], covering a wide temperature range from $-140$ to $+140$ °C. The real part and the imaginary part of the effective piezoelectric coefficient for the composite are investigated separately. Predictions for the real part of $d_{31}^{\ast}$ agree well with the observed values for temperatures larger than $60$ °C, but are larger than the observed values for lower temperatures, while predictions for the imaginary part of $d_{31}^{\ast}$ give fairly good agreement with the experimental data throughout the temperature range. © 2002 American Institute of Physics. [DOI: 10.1063/1.1503388]

I. INTRODUCTION

In previous articles, we have derived some relatively simple explicit expressions for the effective piezoelectric $d$ coefficients for ferroelectric 0-3 composites of small to medium-high volume fraction of the dispersed phase. The theoretical predictions have been compared with experimental results from published works, including the experimental results of Furukawa et al., who also presented a model for the effective piezoelectric coefficients of ferroelectric 0-3 composites. Piezoelectric composites have attracted much interest recently. They can be tailored to suit specific applications, and are commonly used in sensors and actuators. The piezoelectric properties of ferroelectric 0-3 composites have been studied by many workers theoretically and experimentally. Studies on the temperature dependence of piezoelectricity are also of great practical importance, since a sensor may operate over a wide temperature range. In addition, such studies can lead to a better understanding of these materials so that we can use them effectively in developing applications. As the temperature dependence of these effective piezoelectric coefficients for the composites involve relaxation, it is essential to consider the imaginary part of loss tangents of these coefficients. However, there are very few works concerning the imaginary part of these effective piezoelectric coefficients and their temperature dependence.

Lushcheikin gave the temperature dependence of the piezoelectric properties of a series of ceramic-polymer composites, but he did not consider the imaginary part of the corresponding dielectric, elastic, and piezoelectric properties. Similarly, Rittenmyer et al. have not given the imaginary part for their measurement on piezoelectric coefficients for lead titanate/polychloroprene rubber 0-3 composites. On the other hand, Furukawa et al. gave experimental values of the $d_{31}$ coefficient at $50$ °C for 0-3 PZT/epoxy composites of small volume fraction of inclusions, as well as the temperature profile of the complex piezoelectric $d_{31}^{\ast}$ coefficient for the PZT/epoxy composite with $13$ vol % PZT. However, they have not compared the measured temperature dependence of the complex piezoelectric $d_{31}^{\ast}$ coefficient with theoretical predictions based on their model.

This article examines the applicability of Furukawa’s model and our model to the theoretical prediction of the temperature dependence of the effective complex $d_{31}^{\ast}$ values for PZT/epoxy composites. Unlike our previous articles, in which real values are used for the physical properties, here complex valued properties are considered. This treatment becomes more significant for lossy materials. Experimental results from Furukawa et al. cited in the last paragraph are used for comparison, along with the prediction based on Furukawa’s model. The experimental data were taken in a temperature range from $-140$ to $+140$ °C at $10$ Hz. Both models show a fairly good agreement with the experimental data.

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However, two major differences between these two models are noted. First, the constituents are assumed to be incompressible and there is no contribution from \(d_{33}^R\) of the inclusion phase in Furukawa’s model. In our model, the compressibility (or Poisson’s ratio) of the matrix is shown to play a significant role in the real part of the composite’s \(d_{31}\) constant and thus the assumption of incompressibility may limit the usage of their model for the prediction at different temperatures. Second, we have also found that the inclusion’s \(d_{33}^R\), which appears in our model, can have significant effects on the prediction of the imaginary part of \(d_{31}\) of the composite.

II. EFFECTIVE PIEZOELECTRIC COEFFICIENTS OF A COMPOSITE IN DILUTE LIMIT

In a previous article,\(^1\) we have derived closed-form expressions for the effective piezoelectric coefficients \(d_{33}, d_{31},\) and \(d_{13}\) for ferroelectric 0-3 composites. In our derivation, we have assumed that the inclusions are spherical, and both constituents are dielectrically and elastically isotropic. In this article, we are interested in the effective piezoelectric \(d_{31}\) coefficient only, since we are to compare the theoretical values with the complex \(d_{31}\) values of the PZT/epoxy composite measured by Furukawa et al.\(^3\) In the present case, the composite contains a dilute (13 vol %) suspension of electroactive inclusions in a nonpiezoelectric matrix, thus \(d_{31m} = 0\) and

\[
d_{31} = \phi L_{E}(L_{E}^{L} + L_{T}^{L})d_{31i} + L_{T}^{L}d_{33i}\tag{1}
\]

where

\[
L_{E} = \frac{3e_m}{(1 - \phi)e_i + (2 + \phi)e_m},
\tag{2}
\]

\[
L_{T}^{L} = \frac{I}{1 - \phi(3 - J)} - \frac{J}{1 - \phi(1 - 3J)},
\tag{3}
\]

\[
L_{T}^{L} = \frac{I}{1 - \phi(3 - J)} + \frac{2J}{1 - \phi(1 - 3J)},
\tag{4}
\]

and

\[
I = \frac{(1 - \nu_m)Y_i}{2(1 - 2\nu_i)Y_m + (1 + \nu_m)Y_i},
\tag{5}
\]

\[
J = \frac{5(1 + \nu_m)(1 - \nu_m)Y_i}{(1 + \nu_i)(7 - 5\nu_m)Y_m + 2(1 + \nu_m)(4 - 5\nu_m)Y_i},
\tag{6}
\]

where \(e, Y, \nu\), and \(\phi\) are permittivity, Young’s modulus, Poisson’s ratio, and volume fraction, respectively, and subscripts \(i\) and \(m\) denote inclusion and matrix, respectively.

Using an additional assumption that both phases of the composite are incompressible, Furukawa and coworkers gave an expression for the piezoelectric \(d\) coefficient\(^3\)

\[
d_{31} = \phi L_{E}L_{T}d_{31i},
\tag{7}
\]

where \(L_{E}\) is the same as Eq. (2) and

\[
L_{T} = \frac{5Y_i}{3Y_m + 2Y_i - 3\phi(Y_m - Y_i)}.
\tag{8}
\]

In Eq. (8), all \(Y\)'s can be replaced by shear modulus.

It is noted that our expression for the effective \(d_{31}\) constant [Eq. (1)] depends not only on \(d_{31i}\), but also \(d_{33i}\), which is absent in Furukawa’s model. In addition, our \(L_{T}\)’s [Eqs. (3) and (4)] cannot be reduced to that of Furukawa’s [Eq. (8)] by substituting \(\nu_i = \nu_m = 0.5\).

To investigate the temperature dependence of \(d_{31}\) based on Eqs. (1) and (7), all physical properties involved must be allowed to take on complex values. The complex permittivity, Young’s modulus, Poisson’s ratio, and piezoelectric coefficients may be written as\(^12\)

\[
\begin{align*}
e^* &= e' - i e'', \\
Y^* &= Y' + i Y'', \\
\nu^* &= \nu' + i \nu'', \\
d_{31}^* &= d_{31}^i - id_{33}^i, \\
d_{33}^* &= d_{33}^i - id_{33}^i
\end{align*}
\tag{9}
\]

where the superscript asterisk indicates a complex-valued property consisting of a real part, labeled as a single-primed character, and the double-primed quantity represents its imaginary part. Suppose all quantities other than \(\phi\) in Eqs. (1)–(6) are complex, Eq. (1) becomes

\[
d_{31}^\phi = \phi L_{E}^\phi(L_{E}^{L} + L_{T}^{L})d_{31i} + L_{T}^{L}d_{33i}\tag{10}
\]

and we define

\[
\begin{align*}
L_{E}^\phi &= L_{E}^i + iL_{E}^v = \frac{3\epsilon_m^*}{(1 - \phi)e_i^* + (2 + \phi)\epsilon_m^*}, \\
L_{T}^{L} &= L_{T}^i + iL_{T}^v = \frac{I^*}{1 - \phi(3J^*)^*} - \frac{J^*}{1 - \phi(1 - 3J^*)^*}, \\
L_{T}^{L} &= L_{T}^i + iL_{T}^v = \frac{I^*}{1 - \phi(3J^*)} + \frac{J^*}{1 - \phi(1 - 3J^*)},
\end{align*}
\tag{11}
\]

where \(I^*\) and \(J^*\) are given by Eqs. (5) and (6), respectively, with complex-valued properties. Substituting Eqs. (11) into Eq. (10) and using Eq. (9), one can obtain

\[
d_{31}^\phi = \phi \sum_{i=1}^{8} A_i, \tag{12}
\]

\[
d_{33}^\phi = \phi \sum_{i=1}^{8} B_i, \tag{13}
\]

where

\[
\begin{align*}
A_1 &= L_{E}^i(L_{E}^{L} + L_{T}^{L})d_{31i}, \\
A_2 &= -L_{E}^i(L_{E}^{L} + L_{T}^{L})d_{33i}, \\
A_3 &= L_{E}^i(L_{E}^{L} + L_{T}^{L})d_{31i}, \\
A_4 &= L_{E}^i(L_{E}^{L} + L_{T}^{L})d_{33i}, \\
B_1 &= L_{E}^i(L_{E}^{L} + L_{T}^{L})d_{31i}, \\
B_2 &= L_{E}^i(L_{E}^{L} + L_{T}^{L})d_{33i},
\end{align*}
\tag{14}
\]

\[
\begin{align*}
A_5 &= L_{E}^i(L_{E}^{L} + L_{T}^{L})d_{33i}, \\
A_6 &= -L_{E}^i(L_{E}^{L} + L_{T}^{L})d_{31i}, \\
A_7 &= L_{E}^i(L_{E}^{L} + L_{T}^{L})d_{33i}, \\
A_8 &= L_{E}^i(L_{E}^{L} + L_{T}^{L})d_{33i},
\end{align*}
\tag{15}
\]

Similarly for Furukawa’s model, Eqs. (7) and (8) become
before poling was given in the article of Furukawa et al.\(^3\) and their loss tangents for PZT 400 and PZT 5H in their experimental results have been reported by Furukawa et al.\(^3\) Thus, in this work the percentage changes of \(d_{33}^{\prime}\) from \(-140\) °C to room temperature are assumed to be the same as the results of Zhang et al. of \(d_{33}^{\prime}\). For \(d_{33}^{\prime}\), at a higher-temperature range, the percentage changes of \(d_{33}\) measured from room temperature to 100 °C for PZT 802 given by Cheng\(^18\) is used, and the whole temperature profile of \(d_{33}^{\prime}\) is scaled in such a way that \(d_{33}^{\prime} = 400\) pC/N at 50 °C, a value we have used before.\(^1,2\) For temperatures greater than 100 °C, the variation of \(d_{33}^{\prime}\) with temperature is assumed to be the same as that of \(d_{33}^{\prime}\), which has been measured by Furukawa et al.\(^3\). The whole temperature profile of \(d_{33}^{\prime}\) is then scaled in such a way that the loss tangent at \(-140\) °C to room temperature agrees with the measurement of Wang et al.\(^23\) for PZT 400. Wang et al. have reported the \(d_{33}\) values and their loss tangents for PZT 400 and PZT 5H in their article. The \(d_{33}\) values are 253 and 590 pC/N for PZT 400 but only two sets of after-poling values (corresponding to two temperature values) were given. We assume that the ratio of the before-poling value to the after-poling value is uniform throughout the temperature range, and is the same for the real and the imaginary parts. This ratio is taken to be the average of the two ratios (1.256 and 1.295) associated with the two temperatures given by Furukawa et al. Thus, a factor of 1.28 is multiplied to the before-poling values of \(\varepsilon_i\) in the computation.

Concerning the piezoelectric coefficient \(d_{33}^{\prime}\), no experimental results have been reported by Furukawa et al.\(^3\) and the results reported by other researchers must be used. It should be noted that, for a given temperature, the dielectric, elastic, and piezoelectric properties vary with the composition of PZT,\(^14,15\) as well as the addition of dopants.\(^16,17\) Zhang et al.\(^16\) have measured the \(\varepsilon_i\), \(d_{31}\), and \(d_{33}\) for four types of PZT from 4.2–300 K. The magnitude and profile of their experimental \(\varepsilon_i\) and \(d_{31}\) for their Navy-type II PZT are very similar to the corresponding results of PZT given by Furukawa et al.\(^3\) Thus, in this work the percentage changes of \(d_{33}^{\prime}\) from \(-140\) °C to room temperature are assumed to be the same as the results of Zhang et al. of \(d_{33}^{\prime}\). For \(d_{33}^{\prime}\), at a higher-temperature range, the percentage changes of \(d_{33}\) measured from room temperature to 100 °C for PZT 802 given by Cheng\(^18\) is used, and the whole temperature profile of \(d_{33}^{\prime}\) is scaled in such a way that \(d_{33}^{\prime} = 400\) pC/N at 50 °C, a value we have used before.\(^1,2\) For temperatures greater than 100 °C, the variation of \(d_{33}^{\prime}\) with temperature is assumed to be the same as that of \(d_{33}^{\prime}\). Concerning the \(d_{33}^{\prime}\), although there have been some experimental works on the temperature dependence of \(d_{33}\) and \(d_{33}^{\prime}\) for PZT,\(^3,14,22\) very few have reported the loss tangent or the imaginary part of \(d_{33}\) for PZT. These experimental results reveal that the temperature profile of \(d_{33}^{\prime}\) is quite similar to that of \(d_{33}^{\prime}\). We assume the temperature profile of the loss tangent of \(d_{33}\) to be similar to that of \(d_{33}^{\prime}\), which has been measured by Furukawa et al.\(^3\)

\[d_{31}^{\prime} = \phi L_E^{\prime} L_T^{\prime} d_{31}^{\prime}, \]
\[L_E^{\prime} = L_E^{\prime} + iL_T^{\prime}, \]
\[L_T^{\prime} = \frac{5 Y_i^*}{3 Y_m^* + 2 Y_i^* - 3 \phi(Y_i^* - Y_m^*)}, \]
\[d_{31}^{\prime} = \phi \sum_{i=1}^{n} C_i, \]
\[d_{33}^{\prime} = \phi \sum_{i=1}^{n} D_i, \]

where

\[C_1 = L_E^{\prime} L_T^{\prime} d_{31}^{\prime}, C_2 = -L_E^{\prime} L_T^{\prime} d_{31}^{\prime}, C_3 = L_E^{\prime} L_T^{\prime} d_{31}^{\prime}, C_4 = L_E^{\prime} L_T^{\prime} d_{31}^{\prime}, \]
\[D_1 = L_E^{\prime} L_T^{\prime} d_{33}^{\prime}, D_2 = -L_E^{\prime} L_T^{\prime} d_{33}^{\prime}, D_3 = -L_E^{\prime} L_T^{\prime} d_{33}^{\prime}, D_4 = -L_E^{\prime} L_T^{\prime} d_{33}^{\prime}. \]

In summary, Eqs. (12) and (13) are used for the prediction of effective piezoelectric coefficients \(d_{31}^{\prime}\) and \(d_{33}^{\prime}\), with \(L_E^{\prime}\) and \(L_T^{\prime}\)’s given by Eqs. (11). For Furukawa’s model, Eqs. (18) and (19) are to be used accordingly for \(d_{31}^{\prime}\) and \(d_{33}^{\prime}\), with \(L_E^{\prime}\) and \(L_T^{\prime}\) given by Eqs. (11) and (17), respectively.

### III. COMPARISON WITH EXPERIMENTAL DATA

Our theoretical prediction and the prediction based on Furukawa’s model of complex piezoelectric coefficient \(d_{31}^{\prime} = d_{31}^{\prime} + id_{31}^{\prime}\) are compared with the experimental data of Furukawa et al.\(^3\) for a PZT/epoxy composite for the temperature range between \(-140\) and \(+140\) °C (Figs. 1 and 2), with frequency at 10 Hz. The composite sample has 13% of PZT by volume. As insufficient data on dielectric/elastic/piezoelectric properties of the constituent materials (PZT and epoxy) were provided in their articles, typical values have been adopted in our calculation, as will be explained in the following paragraphs (Secs. III A and III B).

#### A. Temperature dependence of complex \(\varepsilon_i\) and \(d_{33}^{\prime}\)

Temperature dependence of the permittivity \(\varepsilon_i^*\) of PZT before poling was given in the article of Furukawa et al.,\(^3\)

![FIG. 1. Comparison with experimental data of Furukawa et al. for the effective \(d_{31}^{\prime}\) (real part) of a PZT/epoxy composite measured at 10 Hz.](image1)

![FIG. 2. Comparison with experimental data of Furukawa et al. for the effective \(d_{31}^{\prime}\) (imaginary part) of a PZT/epoxy composite measured at 10 Hz.](image2)
and PZT 5H, respectively. We use the loss tangent for PZT 400 (rather than that of PZT 5H) because its room temperature $d_{33}$ value is comparable to the corresponding value (221.5 pC/N) measured by Cheng for PZT 802. Based on the above assumptions, the temperature profile of the complex $d_{33}$ is shown in Fig. 3.

B. Temperature dependence of mechanical properties of constituents

Furukawa et al. have measured the temperature dependence of the complex Young’s modulus for PZT and epoxy, but they have not measured the Poisson’s ratios. Poisson’s ratio for the PZT $\nu_i$ has been assumed to be 0.3, and its variation with temperature is assumed to be small. Moreover, as the Poisson’s ratio for the epoxy $\nu_m$ may vary drastically with temperature, two extreme values, 0.3 and 0.5, have been plotted in Figs. 1 and 2 for comparison. We note that Furukawa et al. have used $\nu_i = \nu_m = 0.5$, regardless of the temperature variation. Nevertheless, Fig. 1 shows that the band enclosed by our predictions may not be narrow enough to confirm that the Poisson’s ratio for the constituents are temperature independent. According to the measurement of Tcharkhtchi et al. for epoxy (which is also diglycidylether of bisphenol A, as Furukawa et al.), the bulk modulus is free of viscoelastic effects. From their results of bulk modulus and the measurements of Furukawa et al. of Young’s modulus, we find that the imaginary part of the Poisson’s ratio for the epoxy is a very small value (about 0.002) and it does not affect the $d''_{31}$ and $d''_{i}$ values of the composite significantly. Concerning the imaginary part of the Poisson’s ratio for the PZT, we expect that its effect on the prediction of the effective $d''_{31}$ should be smaller than that of epoxy. It is because the complex Young’s modulus of the PZT has been shown to be nearly independent of temperature, and the present composite sample contains quite a small volume fraction of PZT. Therefore, we have assumed the imaginary part of the Poisson’s ratio for the constituents to be zero, which is the same as the treatment used in Menard’s text. It should be noted that some types of PZT may not behave like the sample from Furukawa et al. (negligible temperature dependence for Young’s modulus). In such a case, the above treatment might not be appropriate.

C. Temperature dependence of effective $d_{31}$ constants

From Fig. 1, concerning the real part of $d_{31}$, the prediction based on Furukawa’s model lies in between our predictions for $\nu_m = 0.3$ and $\nu_m = 0.5$. At the low-temperature regime, $\nu_m$ tends to be smaller and therefore its actual value should be closer to 0.3 rather than 0.5. At the high-temperature regime, especially near 140 °C, where a primary dispersion is seen in the Young’s modulus for epoxy, $\nu_m$ tends to 0.5. Our previous articles have reported that our model is slightly closer to the experimental data (measured at 50 °C) than Furukawa’s model, with $\nu_m = 0.35$. For the region from room temperature to about 110 °C, which is the glass transition temperature for the epoxy, variation of $\nu_m$ is expected to be small and our predictions are in relatively good agreement with the experimental data. For the low-temperature region, both Furukawa’s model and our theory do not make a good agreement with the experimental data. Furukawa et al. have suggested that the predictions were greater than the experimental values due to imperfect poling of the inclusions. This argument does not seem to be applicable in this system because we have shown that theoretical predictions give good agreement at some temperature ranges, but they disagree with experimental data at some other temperature ranges. An imperfect poling should not influence only some temperature ranges (say, around −100 and 50 °C that Furukawa et al. reported); it should influence the whole range from −140 to 140 °C. However, we believe that there are likely other mechanisms not included in the present model which may have been significant at low temperatures, as the discrepancy between predictions and the experimental data tends to be larger there (Fig. 1). This may need further investigation.

On the other hand, Fig. 1 also reveals the importance of $\nu_m$ in $d''_{31}$ prediction. As $\nu_m$ is expected to vary with temperature, the actual prediction varies across the band enclosed by $\nu_m = 0.3$ and $\nu_m = 0.5$ and it is quite wide when compared to that in Fig. 2. Both real and imaginary parts of our $L'_T$’s vary with $\nu_m$, with $L''_T$’s showing the largest variation. We have found that $L''_T$ and $L''_m$ will increase in magnitude by about 250%–225% and 93%–91%, respectively, for $\nu_m$ changing from 0.3–0.5. However, $L'_T$ and $L''_m$ change at most by 0.7% and 15%, respectively. Figure 4 shows the relative contributions of the various terms (only $A_1$, $A_5$, and $C_1$ are shown; other $A$’s and $C$’s are very small) defined in Eqs. (14) and (20) to the resultant $d''_{31}$. It clearly reveals that $A_3$ varies quite substantially with $\nu_m$. No factor similar to $A_3$ appears in the model of Furukawa et al. since it is associated with $d_{33}$, which is not in their model. Actually, for the assumption of rigid inclusions ($Y_i \gg Y_m$), which should be applicable to Furukawa’s experimental data, Eqs. (5) and (6) may be approximated to
I'1\(n^m\)
\(\sim\)22

J'5\(2^1\(n^m\)
\(\sim\)23

without dependence on all other mechanical properties except \(n^m\). It shows that \(n^m\) is a significant parameter in such predictions of piezoelectric coefficients of 0–3 composites. Furukawa’s model assumes the constituents are incompressible. Their model may then be thought to be applicable only to high temperatures where \(n^m\) is close to 0.5. Indeed, the predictions given by Furukawa’s model are in excellent agreement with the experimental data beyond 110 °C (the glass transition temperature for the epoxy).

Figure 2 shows the imaginary part of \(d_{31}^*\) for the composite, comparing Furukawa’s model and our prediction. The profile of the experimental result looks very similar to the imaginary part of the permittivity of epoxy shown in the article by Furukawa et al.\(^3\) Below room temperature, both Furukawa’s model and our prediction give fairly good agreement to the experimental data. Concerning the temperature range from 50 to about 120 °C, our prediction based on \(n^m\)
\(=0.5\) allows a direct comparison with Furukawa’s model. As evident from Fig. 2, our predictions show significant advantage over Furukawa’s model. The predicted values assuming \(n^m=0.5\) are much closer to the experimental data than the predicted values for \(n^m=0.3\) in this temperature range. A feature in our expression [Eq. (10)] is that, other than \(d_{31}^*\), \(d_{33}^n\) plays an important role in the prediction, especially in the imaginary part. Figure 5 and 6 show the relative contributions of the various terms defined in Eqs. (15) to the resultant \(d_{31}^n\) for \(n^m=0.3\) and 0.5, respectively. Compared with Fig. 7, which shows relative contributions of the terms in Eqs. (21) from Furukawa’s model, they clearly reveal that \(B_5\) and \(B_8\) of Eqs. (15) vary sensitively with \(n^m\) (though the effective \(d_{33}^n\) may not be so), and they are both factors associated with \(d_{33}^n\) which is not included in Furukawa’s model. This \(d_{33}^n\) contribution is more significant in the higher-temperature region.

All in all, the temperature profile of the complex \(d_{31}^n\) constant for the composite is quite similar to the temperature profile of complex permittivity for the epoxy as shown in the article by Furukawa et al.\(^3\) However, the drastic variation of the composite \(d_{31}^n\) should not be mostly governed by the piezoelectric properties of PZT alone, but also the dielectric and elastic properties of the constituents. At such small volume fraction of PZT, the dielectric and elastic properties of

\[
I = \frac{1 - \nu_m}{1 + \nu_m},
\]
\[
J = \frac{5}{2} \frac{1 - \nu_m}{4 - 5 \nu_m},
\]

FIG. 4. Prediction for the temperature dependence of \(A_1\), \(A_5\), and \(C_1\) [Eqs. (14) and (20)]. Other A’s and C’s are not shown because they contribute to less than 0.1% to the overall values.

FIG. 5. Prediction for the temperature dependence of \(B_i\)’s [Eq. (15)] with \(n^m=0.5\).

FIG. 6. Prediction for the temperature dependence of \(B_i\)’s [Eq. (15)] with \(n^m=0.5\).

FIG. 7. Prediction for the temperature dependence of \(D_i\) to \(D_4\) [Eq. (21)].
the matrix phase are thought to make larger contributions to the composite $d_{31}^{\mu}$ than that of PZT's. Our predictions give slightly better agreement with experimental data over a broader temperature range. This merit is much more obvious in the imaginary part of the effective piezoelectric coefficient $d_{31}^{\mu}$.

**IV. CONCLUSIONS**

The temperature dependence of the complex effective piezoelectric coefficient $d_{31}^{\mu}$ for ferroelectric 0–3 composites has been studied theoretically at small ceramic volume fraction. Our theoretical prediction has been compared with Furukawa’s model for the experimental values of PZT/epoxy from published work, also by Furukawa et al. Fairly good agreement was obtained for temperatures larger than 60 °C for the prediction of the real part as well as the imaginary part of $d_{31}^{\mu}$. The comparison shows that the contribution of the $d_{33}^{\mu}$ term in Eq. (10) (which is absent in Furukawa’s model) can be significant in predicting the imaginary part of the effective $d_{31}^{\mu}$ coefficient. Moreover, the compressibility of the matrix is shown to be a key factor for the real-part prediction of the $d_{31}$ constant, especially for composites with rigid inclusions. As incompressibility for both constituents is not assumed in our model, our predictions are expected to be applicable to a wider temperature range. To conclude, temperature affects the piezoelectric properties of a composite in a complicated manner because the piezoelectric properties of a composite depend not only on the piezoelectric properties of the constituents, but also on their dielectric and elastic properties, although they contribute to varying degrees.

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17. Data sheet from Morgan Electro Ceramics Inc.