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*Research Article*

# Predictability of Dielectric Constant of Polyvinylidiene Fluoride- $BaTiO_3$ (PVDF/BT) Composite based on Input Concentration of $BaTiO_3$ and its Associated Electric field Frequency

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**Special Issue**
*A Themed Issue in Honour of Professor Clement Uche Atuanya on His retirement.*

This themed issue pays tribute to Professor Clement Uche Atuanya in recognition of his illustrious career in Metallurgical and Materials Engineering as he retires from Nnamdi Azikiwe University, Awka. We celebrate his enduring legacy of dedication to advancing knowledge and his impact on academia and beyond through this collection of writings.

Edited by
Chinonso Hubert Achebe PhD.
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# Predictability of Dielectric Constant of Polyvinylidiene Fluoride- $BaTiO_3$ (PVDF/BT) Composite based on Input Concentration of $BaTiO_3$ and its Associated Electric field Frequency

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**Abstract**
The dielectric constant of polyvinylidene fluoride-$BaTiO_3$ (PVDF/BT) composite was predicted based on the input concentration of $BaTiO_3$ (filler) ɤ and associated electric field frequency Ғ. This followed the development of a model equation relating the dielectric constant, $BaTiO_3$ input concentration, and electric field frequency. The PVDF/BT composite was produced using a step-by-step process, involving hot-pressing the input materials such that the ceramics component is wrapped by the polymer. The validity of the derived model expressed as; $K = ße^{\phi ɤ} + ƀF^{ɱ} - N$ was rooted in the core expression $K - ße^{\phi ɤ} \approx ƀF^{ɱ} - N$, in that both sides of the expression are correspondingly near-equal, where ß, ϕ, ƀ, ɱ and N are the equalizing constants. According to the results of both experiments and model predictions, the dielectric constant increases with increasing filler loading and electric field frequency, as reported previously. The evaluated results demonstrated that the correlations between the dielectric constant, filler loading, and electric field frequency are 0.9982, 0.9955, and 0.9982, respectively, using model-predicted and experimental results, showing the responses of the composite`s dielectric constant to the highlighted influencing factors. The overall standard error incurred in predicting the dielectric constant of PVDF-BT composite is < 0.18%, for every change in the electric field frequency & $BaTiO_3$ input concentration. The PVDF-BT composite dielectric constants per unit electric field frequency and $BaTiO_3$ input concentration are $7.77 \times 10^{-6}$, $7.58 \times 10^{-6}$& $6.84 \times 10^{-6}$ $(Hz)^{-1}$ and 1.84, 1.80 & 1.62 $(wt\%)^{-1}$ as evaluated from experimental, derived model and regression model-predicted results. The highest deviation of the model-predicted dielectric constant (from experimental or real measurements) was less than 1.9%. This resulted in over 98% operational confidence in the resulting model and over 0.98 response coefficients for the dielectric constant's dependence on electric field frequency and $BaTiO_3$ input concentration.

**Keywords:** Dielectric Constant, Polyvinylidene Fluoride ($BaTiO_3$), Composite, Electric Field Frequency

## 1. Introduction

Following the rapid rise of the global economy and energy consumption, studies (Xie et al. 2019; Yang et al.2020; Kausar, 2017; Poonam et al. 2019) have created a high level of awareness about the necessity for the development of environmentally friendly and economically effective energy conversion/storage systems. Dielectric polymer materials have found widespread application in domains such as pulsed power systems (Zhou et al. 2017; Morales-Masis et al. 2019), sensors (Charaya et al 2019; Kim et al.2019), artificial muscle (Miryakili et al. 2018; Chen et al. 2019) and optoelectronics (Pan et al. 2017; Wang et al. 2017) as well as energy storage devices Li et al. 2021; Hu et al 2020). The relative dielectric constant (or permittivity) is also referred to as the dielectric constant. It is the ratio of a substance's permittivity to that of the vacuum. Scientists (Zhou and Jiang, 2019; Martin et al. 2014) have described the use of high permittivity polymers in energy storage and flexible sensing systems, citing their superior dielectric characteristics and flexibility. According to (Lukacs et al. 2022) permittivity, dielectric loss, and dielectric

breakdown strength (DBS) are related to the polarization types within the polymer, and the three metrics characterize the polymers' dielectric properties. It is commonly recognized that the permittivity values of many polymers are insufficient for their use in high-energy equipment. Despite their low dielectric loss and high DBS, they continue to be employed as dielectric and insulating materials in the electrical sector. This highlights the need for substantial research into modified polymers (Lallart et al. 2010) and polymer composites (Wang and Zhu, 2011) to achieve optimal dielectric characteristics.

Researchers have established that inorganic nanoparticles, such as ceramics with insulating characteristics (Bi et al. 2018; Hu et al. 2016) metals (Feng et al. 2016; Wang et al. 2018) or carbon particles with electrical conductivity (Yang et al. 2016; Dag et al. 2016; Zakaria et al 2018), can be combined with polymer matrix to generate polymer nanocomposites. The use of nanofillers such as graphene oxide (Ning et al. 2017; Zhou et al. 2015), Zn nanoparticles (Zhou et al. 2015), and $BaTiO_3$ (BT) ceramic nanofillers (Gao et al. 2021) has greatly enhanced the permittivity of composite materials. This was due to the success of numerous studies that focused on the influence of the composite's dielectric property on nanofillers and other significant properties such as nanofiller dispersibility in the polymer matrix, size, content, and form.

Due to their superior dielectric characteristics, barium titanate/polyvinylidene fluoride (BT/PVDF) nanocomposites are among the most extensively researched composite systems. The size of the BT fillers has been demonstrated to have a substantial effect on the dielectric responses of nanocomposites (Bi et al. 2017). Furthermore, increasing the input volume fraction of fillers significantly increased the nanocomposites' breakdown strength and dielectric characteristics. The dielectric characteristics of polymers are largely dependent on the frequency of the external electric field. While dielectric loss is produced by energy loss from charge movement or dipole reorientation, which is also affected by the frequency of the external electric field, permittivity is directly tied to frequency-dependent polarization types. Further research (Qiu et al. 2022) has shown that for polymer dielectrics, dielectric properties are affected by not only external factors (such as frequency of electric field and temperature), but also controllable internal factors (such as interfacial morphology of the fillers and the filler loading level).

The size and surface of $BaTiO_3$ ceramic powder have been demonstrated (Fu et al. 2015) to play an important impact on the dielectric characteristics of the composite during the preparation process. This comes after the discovery that $BaTiO_3$/PVDF composite is a common hybrid ceramic-polymer system. A related research (Lukacs et al 2022; Lallart et al. 2010)] shows that the dielectric and piezoelectric properties of the composite can be effectively improved by increasing the ceramic content and decreasing the grain size, and however, deteriorate when the content and size exceed a certain value. Based on the foregoing, the electrical properties of the composites cannot be simply improved by increasing the ceramic content and reducing the ceramic size.

The current study intends to predict the dielectric constant of a $BaTiO_3$/PVDF composite based on the $BaTiO_3$ input concentration and associated electric field frequency. This is based on prior research (Bi et al. 2017) indicating the dielectric constant of the composite increases with an increase in both the input concentration of $BaTiO_3$ and the electric field frequency. However, there is no current mathematical or empirical statement that connects the highlighted variables. The empirical model that will be developed during the project will evaluate the dielectric constant using these process parameters. It is expected that the resulting model will predict the dielectric constant within the experimental range simply by substituting values of the input concentration of $BaTiO_3$ and electric field frequency into the model if the boundary conditions are taken into account.

## 2.0 Material and methods

The $BaTiO_3$ (white powder) used in this work was obtained from Shandong Guoci Functional Materials Co., Ltd., China. The melting point and relative density are 1625°C and 6.017 g/cm$^3$, respectively. The particle size of $BaTiO_3$ used was between 0.01 μm and 200 μm. The carbon, oxygen, barium, and titanium contents are 31.58%, 30.61%, 28.11% and 8.85%, respectively. Elements such as Si, Al and Co are also present as impurities.

### 2.1 Preparation Process of Composites

2.1.1 Preparation Process

The preparation process in the previous research (Wang et al. 2023) was used. The process is all about hot-pressing the input materials, as a way of integrating the much needed heating and pressing, to ensure a dense solid harnessed as the composites. Fig. 1 gives a diagram of the hot-press process, where the piezoelectric ceramic powder

($BaTiO_3$), fully mixed with the polymer matrix (PVDF), was heated and melted, enabling the ceramic powder to get wrapped by the polymer. This process enabled the ceramic materials to combine without sintering ceramic growth, and still possess certain mechanical strength. This shows that composite materials can reduce the brittleness of ceramics and significantly improve the overall mechanical toughness.

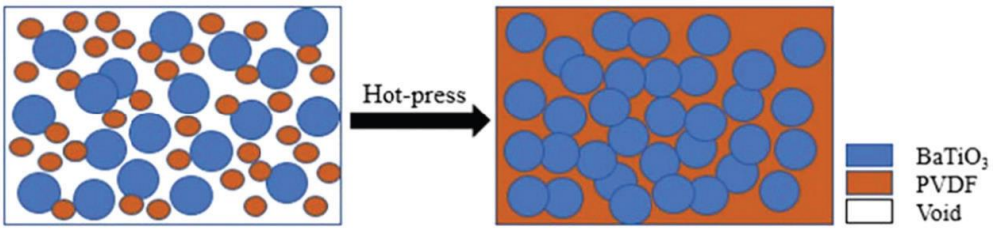


Fig. 1: The schematic diagram of the preparation process

2.1.2 $BaTiO_3$/PVDF Composites

The procedure involving the preparation of the composite is shown in Fig. 2. At 950°C and 60°C, for 3 h each, the $BaTiO_3$ powder & PVDF polymer were placed in a muffle furnace and oven respectively. $BaTiO_3$&PVDF were also poured into a beaker with ethyl alcohol, and then put into an ultrasonic washer to accelerate the mixing process for 30 min. This was done after drying and cooling to room temperature, following initial heating. The dried mixture was put into a grinding machine to grind into powder after another round of heating 90°C in an oven. The powder was heated again to 180°C in heating equipment and then pressed at 25 MPa for 30 min using a hot-press forming machine. This resulted into a cylindrical composite of 13-mm diameter and 1-mm thickness. The materials were thereafter evenly coated on the top and bottom using TYD-110Y conductive silver fluid, provided by UV Tech. Material, Ltd. (Guangzhou, China). The dried and coated composites were polarized at 3kV/mm for 30 min using a polarization device, following treatment in an oil bath at 120°C. The polarized composites were then placed in a dried container to discharge for 24 h (Wang et al. 2023).

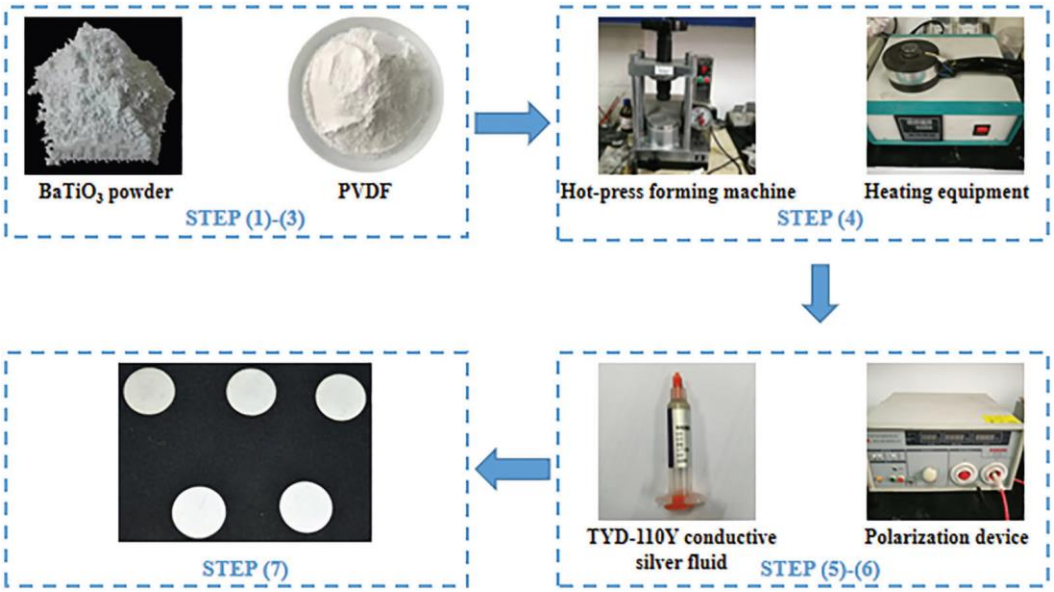


Fig. 2: The flowchart of composites preparation (Wang et al. 2023)

**2.2 Testing Methods**

2.2.1 Electrical Conductivity

A very important indicator used to characterize a piezoelectric material's ability to conduct electric current is the electrical conductivity (σ). This parameter is the ratio of current density to electric field strength. The σ values of the $BaTiO_3$/PVDF composites were measured using an HP 4294A precision impedance analyzer (Wang et al. 2023)

2.2.2 Dielectric Properties

The polarization degree of the piezoelectric materials characterizes by the relative dielectric constant, which is the ratio of the permittivity of piezoelectric materials to the vacuum. The constant can also represent the amount of charge that can be stored in the piezoelectric materials. In this study, the capacitance of piezoelectric materials was measured using a Hioki LCR Meter IM3536 with a high-speed measurement of 1 m/s, and high-precision measurement of ±0.05% representative value. The relative dielectric constant $\varepsilon_r$ or K was then calculated using the following equation (Wang et al. 2023). Where *C* is the capacitance of the sample, t is the thickness of the sample, A is the area of the sample, and $\varepsilon_0$ is the dielectric constant of vacuum.

$$\varepsilon_r = {C_t}/{e_0 A} \quad (1)$$

Where

$$\varepsilon_r = \varepsilon_m / \varepsilon_0 \quad (2)$$

and

$\varepsilon_m$ = Permittivity of the material

**3.0 Results and Discussions**

Table 1 shows variation of the dielectric constant of PVDF-BT composite with the input concentration of $BaTiO_3$ and its associated electric field frequency. The table shows that the dielectric constant of the composite increases with increase in both the input concentration of $BaTiO_3$ and electric field frequency, in line with past report [31]. The dielectric constants recorded in Table 1 (actual values) were evaluated by substituting the values of A, t and *C* from experiment into equation (1), while $\varepsilon_0$ is known as 8.85418782 x $10^{12}$ Farad/meter.

Table 1: Variation of the dielectric constant of PVDF-BT composite with input concentration of $BaTiO_3$ and its associated electric field frequency

| (*K* ) | (ɤ ) wt% | (Ғ) x$10^3$ Hz |
|---|---|---|
| 10.01 | 0 | 0.1 |
| 11.10 | 0.89 | 0.5 |
| 12.50 | 2.00 | 1 |
| 13.57 | 2.80 | 5 |
| 14.85 | 3.80 | 10 |
| 16.08 | 4.34 | 50 |
| 17.61 | 5.00 | 100 |
| 19.25 | 5.89 | 500 |
| 21.30 | 7.00 | 1000 |

3.1 Model formulation

Computational analysis of the variables in Table 1 gave Table 2 which shows that;

$$K - ße^{ϕɤ} \approx ϧF^{ɱ} - N \quad (3)$$

$$K = ße^{ϕɤ} + ϧF^{ɱ} - N \quad (4)$$

The derived model in the expression (4) predicts the dielectric constant of a nano polymer; polyvinylidiene fluoride-$BaTiO_3$ (PVDF/BT) composite, based on the input concentration of $BaTiO_3$ and its associated electric field

frequency. Table 1 and Table 2 shows that *K*, ɤ and Ғ are the dielectric constant, input concentration of $BaTiO_3$ (wt%) and $BaTiO_3$ associated electric field frequency (Hz) respectively. The equalizing constants; ß,ϕ, Ƀ, ɱ and N are 5.012, 0.1091, 3.4825, 0.0795 and 0.001 respectively. The interactive input of these constants cancels out the units and places *K* as dimensionless.

3.2 Model validity

The validity of the derived model in (2) is strongly rooted in the core model structure expressed in (1), both sides of which are correspondingly almost equal. This is confirmed in Table 2 (computed from experimental results in Table 1) following the evaluated numerical values of both sides of the structure components.

Table 2: Variation of K - $ße^{\phi ɤ}$with $ƁҒ^{ɱ}$ - N

| *K* - $ße^{\phi ɤ}$ | $ƁҒ^{ɱ}$ - N |
|---|---|
| 4.9980 | 5.0211 |
| 5.5768 | 5.7068 |
| 6.2661 | 6.0300 |
| 6.7672 | 6.8533 |
| 7.2628 | 7.2416 |
| 8.0327 | 8.2302 |
| 8.9618 | 8.6965 |
| 9.7202 | 9.8837 |
| 10.5432 | 10.4437 |

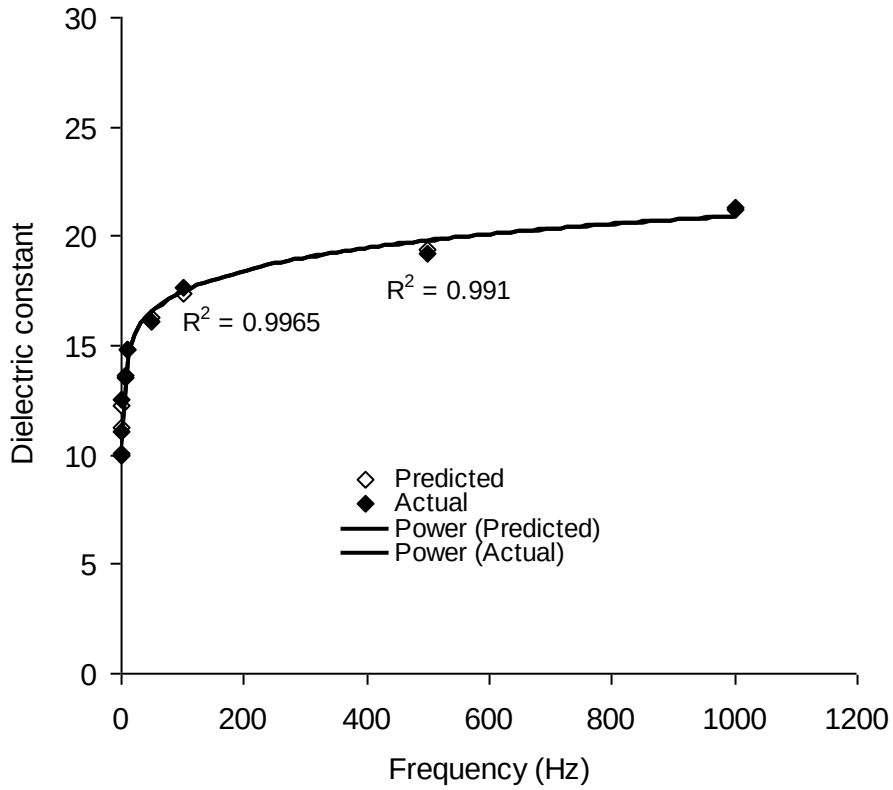

Fig. 3: Coefficient of determination between dielectric constant and electric field frequency as evaluated from actual results and derived model

The correlations between the dielectric constant of PVDF-BT and $BaTiO_3$ associated frequency & its input concentration were calculated using model-predicted and experimental results. These are 0.9982 and 0.9955 & 0.9982 and 0.9990; evaluated from the coefficients of determination $R^2$ shown in Fig. 3 & Fig. 4 respectively. The evaluated correlations are indicative of close fitted trend lines and show the responses of the composite`s dielectric constant to the highlighted influencing factors.

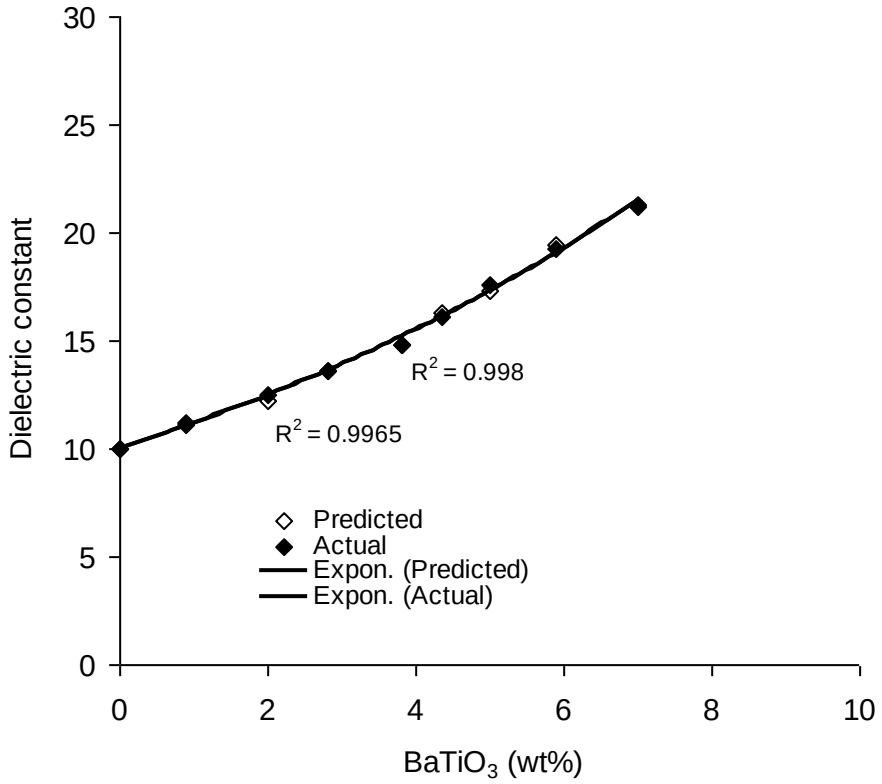


Fig.4: Coefficient of determination between dielectric constant and input concentration of $BaTiO_3$ as evaluated from actual results and derived model.

Analysis of the model-predicted results relative to the experimental shows that the overall standard error incurred in predicting of the dielectric constant of PVDF-BT composite is < 0.18%, for every change in the $BaTiO_3$ associated frequency & its input concentration. This gives a model confidence level above 99%.

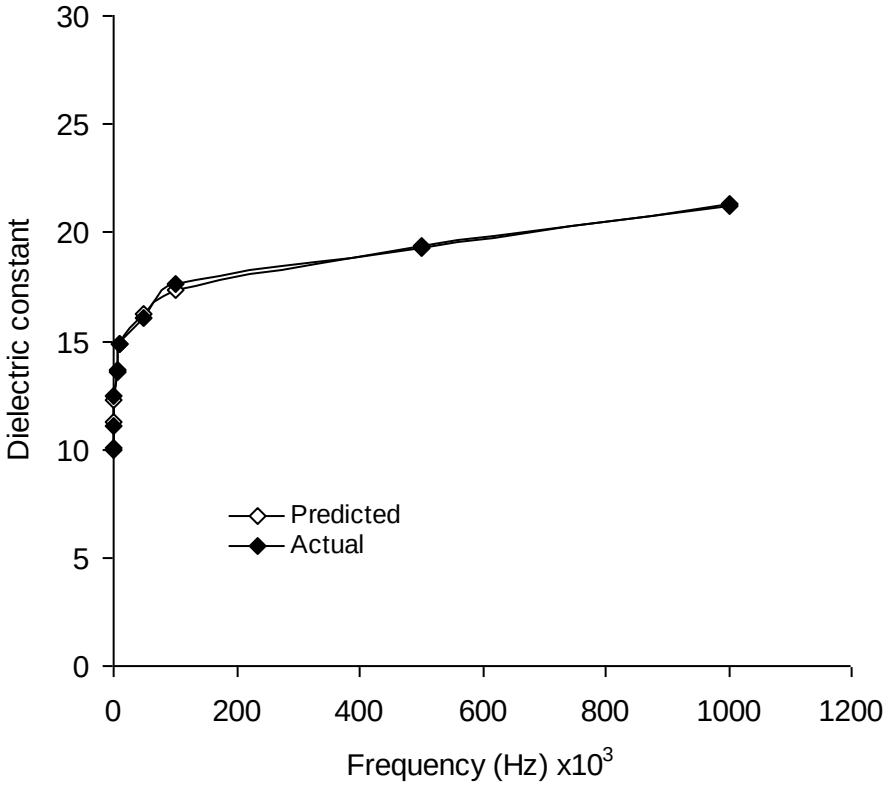


Fig. 5: Comparison of the dielectric constants of PVDT-BT composite (relative to electric field frequency) as evaluated from actual results and derived model.

Comparative analysis of Fig. 5 and Fig. 6 shows closely aligned and fitted curves of experimental and model-predicted results, represented by dielectric constants of PVDT-BT composite relative to $BaTiO_3$ associated frequency and its input concentration respectively. These curves are characterized by similar spread & trend of results point distribution and very close corresponding result sets. Furthermore, the curves from these figures are positive in nature, indicating a direct relationship between the highlighted dielectric constant and $BaTiO_3$ associated frequency & input concentration respectively. It is therefore expected that these highlighted relationships will prompt positive slopes.

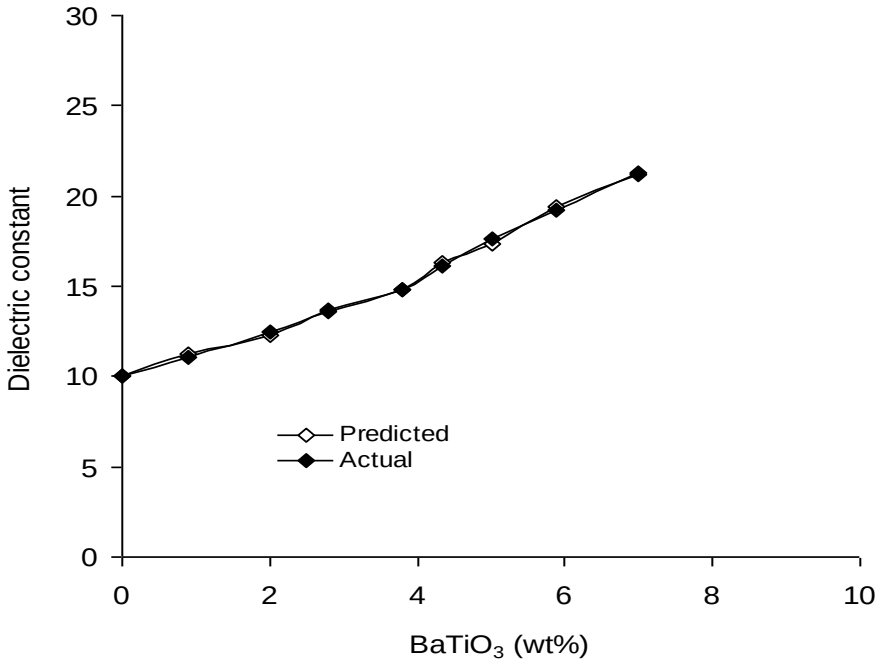


Fig. 6: Comparison of the dielectric constants of PVDT-BT composite (relative to input concentration of $BaTiO_3$) as evaluated from actual results and derived model.

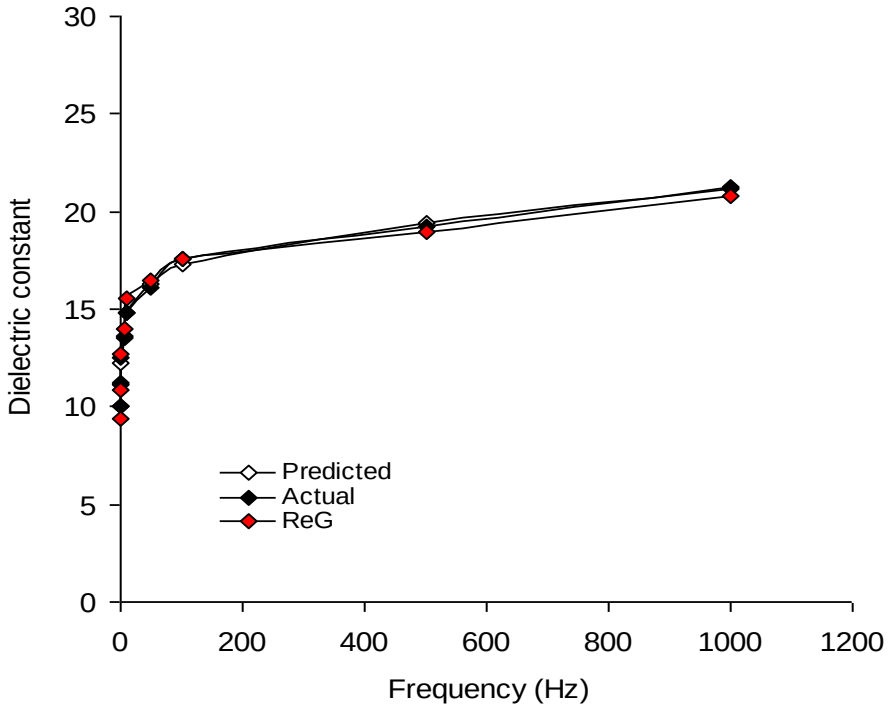


Fig. 7: Comparison of the dielectric constants of PVDT-BT composite (relative to electric field frequency) as evaluated from actual results, derived model and regression model-predicted results.

Each of Fig. 7 and Fig. 8 show three closely aligned and fitted set of curves, designating dielectric constant of PVDT-BT composite relative to $BaTiO_3$ associated frequency and its input concentration respectively. These curves

represent the experimental, derived model-predicted and regression model-predicted results. These curves share same characteristics with those from Fig.5 and Fig. 6 respectively, except for the extra curve referred to as regression curve. The extra curve gives the standard computer-aided (model) prediction, based on the trend from the experimental results. The essence of plotting the regression results alongside those from experiment and model-prediction is to ascertain the validity of the derived model through revelation of the level of discrepancies from the first two sets of results already shown. Curves from Fig. 7 and Fig. 8 are also expected to give positive slopes, following the direct relationships between the dielectric constant and $BaTiO_3$ associated frequency & input concentration respectively.

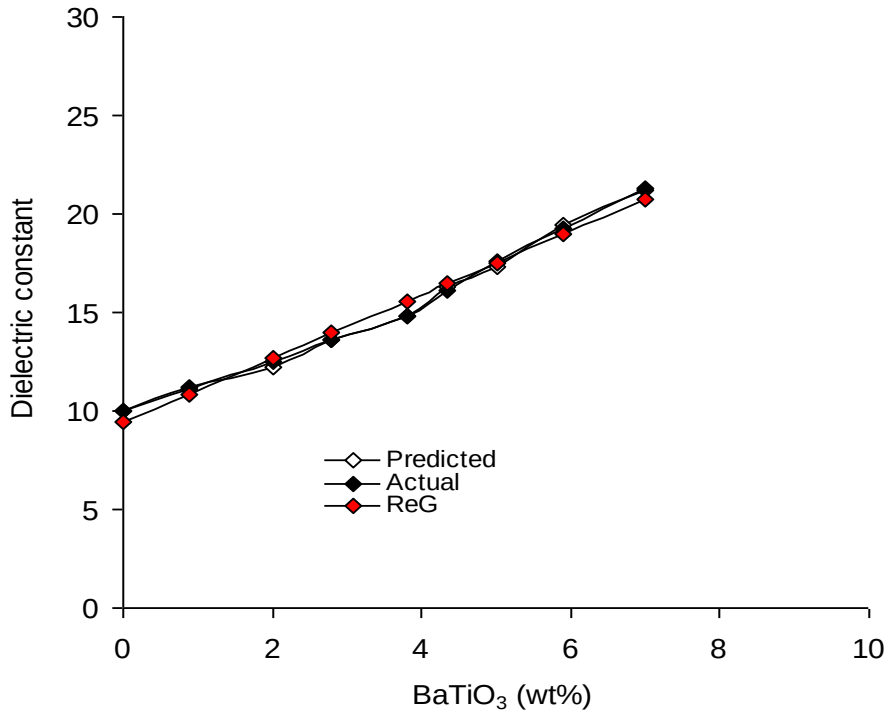


Fig.8: Comparison of the dielectric constants of PVDT-BT composite (relative to input concentration of $BaTiO_3$) as evaluated from actual results, derived model and regression model-predicted results.

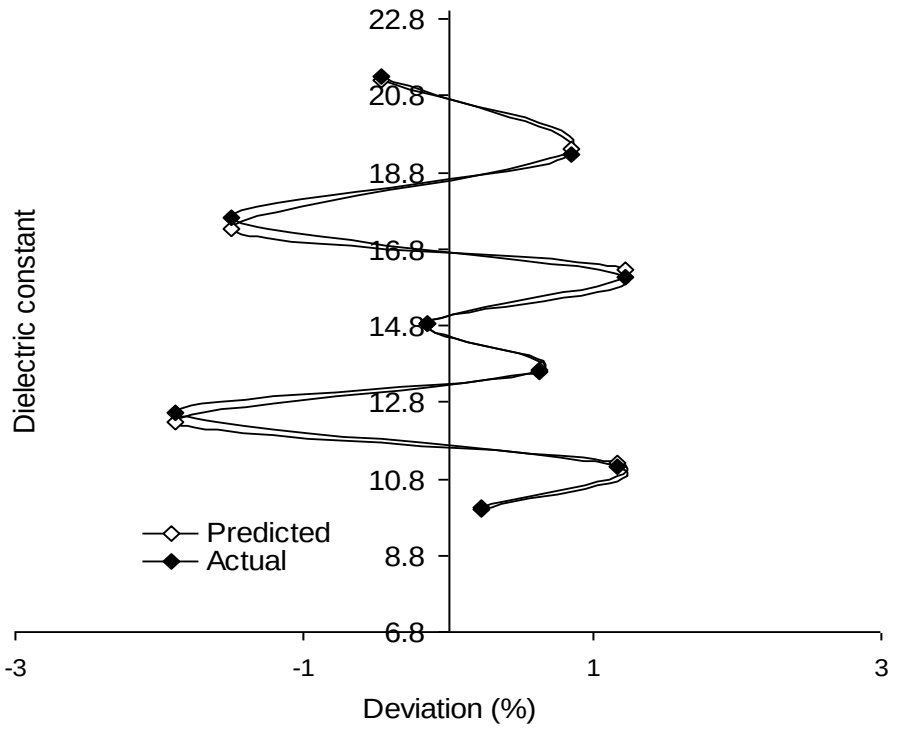


Fig.9: Variation of model-predicted dielectric constant of PVDF-BT composite with its corresponding deviation from experimental results.

Table 3: Differential between experimentally determined and model-predicted dielectric constant $K_\varepsilon$ and $K_M$ respectively.

| $K_\varepsilon$ | $\Delta K = K_M - K_\varepsilon$ |
|---|---|
| 10.01 | 0.0231 |
| 11.10 | 0.1300 |
| 12.50 | -0.2361 |
| 13.57 | 0.0861 |
| 14.85 | -0.0212 |
| 16.08 | 0.1975 |
| 17.61 | -0.2653 |
| 19.25 | 0.1635 |
| 21.30 | -0.0995 |

The maximum deviation of model-predicted dielectric constant (from experimental or actual values) as shown in Fig. 9 is 1.89%. This gives over 98% operational model confidence levels. The figure also shows at each corresponding results set, the discrepancy (in percent) between the experimental and model-predicted results. The overall model confidence level therefore places between 98 and 99%, following evaluated correlations, standard error and maximum deviation. The least and highest deviations of the model-predicted dielectric constants are -0.14 and -1.89% respectively. These deviations correspond to the dielectric constants: 14.8288 & 12.2638, electric field frequencies: 10 & 1.0Hz and input concentrations of $BaTiO_3$: 3.8 & 2.0 wt% respectively. Table 3 shows that the evaluated maximum differential between experimentally determined dielectric constant and model prediction is –

0.2653, which is quite negligible. The positive and negative differentials are indicative of surplus and deficit in model-prediction respectively, relative to the corresponding experimental result.

3.3 Dielectric constant of PVDF-BT composite per unit frequency of electric field.
Dielectric constant of PVDF-BT composite per unit frequency of electric field $K_F$ $(Hz)^{-1}$ was calculated from the expression;

$$K_F = K / F \quad (5)$$

Re-written as

$$K_F = \Delta K / \Delta F \quad (6)$$

The expression (6), is detailed as

$$K_F = K_2 - K_1 / F_2 - F_1 \quad (7)$$

Where

$\Delta K$ = Change in the dielectric constant of PVDF-BT composite $K_2$, $K_1$at two electric field frequencies associating $BaTiO_3$ $F_2$, $F_1$.

Plotting points (5000, 13.57) & (1000000, 21.3), (5000, 13.6561) & (1000000, 21.2005) and (5000, 13.9674) & (1000000, 20.7720) as shown in Fig.7, designated as ($F_1$, $K_1$) and ($F_2$, $K_2$) for experimental, derived model and regression model-predicted results, and substituting them into the expression (7), gives the slopes: 7.77 x $10^{-6}$, 7.58 x $10^{-6}$ and 6.84 x $10^{-6}$ $(Hz)^{-1}$, as their respective PVDF-BT composite dielectric constant per unit electric field frequency associating $BaTiO_3$

3.4 Dielectric constant of PVDF-BT composite per unit input concentration of $BaTiO_3$
Dielectric constant of PVDF-BT composite per unit input concentration of $BaTiO_3$
$K_\gamma$ $(wt\%)^{-1}$ was calculated from the expression;

$$K_\gamma = K / \gamma \quad (8)$$

Re-written as

$$K_\gamma = \Delta K / \Delta \gamma \quad (9)$$

The expression (9), is detailed as

$$K_\gamma = K_2 - K_1 / \gamma_2 - \gamma_1 \quad (10)$$

Where

$\Delta K$ = Change in the dielectric constant of PVDF-BT composite $K_2$, $K_1$at two $BaTiO_3$ input concentrations $\gamma_2$, $\gamma_1$.

A plot of points (2.8, 13.57) & (7, 21.3), (2.8, 13.6561) & (7, 21.2005) and (2.8, 13.9674) & (7, 20.7720) as shown in Fig. 8, designated as ($\gamma_1$, $K_1$) and ($\gamma_2$, $K_2$) for experimental, derived model and regression model-predicted results, and then substituted into the expression (10), gives the slopes: 1.84, 1.80 and 1.62 $(wt\%)^{-1}$, as their respective PVDF-BT composite dielectric constant per unit $BaTiO_3$ input concentration.

## 4.0. Conclusion

The predictability of the dielectric constant of polyvinylidiene fluoride- $BaTiO_3$(PVDF/BT) composite based on input concentration of $BaTiO_3$ (filler) and its associated electric field frequency was established through derivation of a model expression relating the dielectric constant, $BaTiO_3$ input concentration and electric field frequency. The validity of the derived model expressed as; $K = ße^{ϕɤ} + ЂF^{ɱ} - N$ was rooted in the core expression $K - ße^{ϕɤ} \approx ЂF^{ɱ} - N$ where both side of the expression were correspondingly near-equal. Results generated from both experiment and model prediction indicates that the dielectric constant increases with increased the filler loading and electric field frequency. The correlations between the dielectric constant and filler loading & electric field frequency are 0.9982 and 0.9955 & 0.9982 and 0.9990 using model-predicted and experimental results. The overall standard error incurred in predicting of the dielectric constant of PVDF-BT composite is < 0.18%, for every change in the electric field frequency & $BaTiO_3$ input concentration. The PVDF-BT composite dielectric constants per unit electric field frequency and $BaTiO_3$ input concentration are 7.77 x $10^{-6}$, 7.58 x $10^{-6}$& 6.84 x $10^{-6}$ $(Hz)^{-1}$ and 1.84, 1.80 & 1.62 $(wt\%)^{-1}$ as evaluated from experimental, derived model and regression model-predicted results. The overall maximum deviation of the model-predicted dielectric constant (from experimental or actual results) was less than 1.9%.

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