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Response of PZT-rod Periodicity in piezoelectric polymer structure based on density of the composite and volume percent PZT in composite

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ABSTRACT: *Response of PZT-rod periodicity in piezoelectric polymer structure to the density of the composite and volume percent PZT in composite was evaluated, using derived empirical model;* $\mathscr{S} = \beta\rho^{-e} + N\gamma^{-b} - \eta$. *The validity of the model is strongly rooted on the core model structure;* $\mathscr{S} - N\gamma^{-b} \approx \beta\rho^{-e} - \eta$, *both sides of which are correspondingly near equal. The model-predicted results agree with previous research on the inverse relationship between the PZT rod periodicity and density of composite & volume fraction of PZT rod in composite. The correlations between the periodicity of PZT-rod in polymer structure and volume percent of PZT in composite & density of composite were all > 0.99. The standard error incurred in predicting the model based PZT rod periodicity, relative to the actual results was < 0.023%, implying over 99% model confidence level. The periodicity of PZT rod per unit volume fraction of PZT rod in composite and per unit density of the composite were -0.042 & – 0.043 mm/% and -0.533 & – 0.546 mm/ (gm/cc), using experimental and model-predicted results respectively. The overall maximum deviation of the model-predicted periodicity of PZT rod from actual results was 2.56%. The derived model will predict the periodicity of PZT rod, within the actual results range, on substituting into the model, values of the density of composite and volume fraction of PZT rod in composite, providing the boundary conditions are considered.*

KEYWORDS*: PZT-rod periodicity, piezoelectricity, polymer structure, density, PZT volume percent, composite*

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I. INTRODUCTION

Research has been conducted on some designed and fabricated composite materials, optimized for a special application. The applications range from electronic devices to mechanical structures. During composite material design, prime importance is given to the way in which the proper choice of component phases is interconnected to maximize the merit of the expected outcome.

It has been reported by several authors in (Newnham et al., 1978; Klicker et al., 1981; Klicker et al., 1982; Newnham et al., 1980; Rittenmyer et al., 1980; Safari et al., 1982; H. Banno and S. Saito, 1983) that piezoelectric ceramic-polymer composite transducers are a family of an important new class of materials. These reports indicate that current researches on piezoelectric ceramic-polymer composite materials focus on hydrophone applications in the low-frequency (less than 40 KHz) range, where the dimensions of the transducer are much smaller than the acoustic wavelength.

Until now, there has not been any indication of studies, geared towards the usefulness of these composites at higher frequencies (1-10 MHz) for medical diagnostic and nondestructive testing applications. At these frequencies, the acoustic wavelength is comparable to the scale of the composite microstructure. The acoustic impedance, bandwidth, and radiation patterns of the composite transducer can be controlled in a manner so sophisticated that it is impossible in single phase materials. The systematic investigations (McGrath et al., 1983) of the composite materials made from piezoelectric lead zirconate titanate (PZT) ceramics and piezoelectrically inactive polymer have focused on the understanding of electromechanical properties of the composite materials in resonant configurations. The knowledge of the high-frequency dynamic behavior of the composite was then used to evaluate the composite materials for ultrasonic transducer applications, with an emphasis on medical diagnostic applications.

The PZT rod-polymer composites have been prepared (McGrath et al., 1983) with 5 to 30 volume percent PZT, using 0.28 mm and 0.45 mm rods. In a disc of the PZT rod-polymer composite material, there are three principal types of resonance: the planar mode, the thickness mode, and various lateral modes caused by the regular periodicity of the PZT rod in the composite. These resonance modes have been studied with the following techniques: electrical impedance measurement as a function of frequency and laser probe dilatometry of the dynamic displacement, as function of frequency and position in the composite lattice. The observed resonance behavior was found to be a result of lateral interactions in the composite through the epoxy medium. It has been shown (McGrath et al., 1983) that the periodicity of PZT rod in a polymer structure, which also affects the highlighted resonance behavior, is the distance between the centers of neighboring PZT rods.

Investigation according to Gururaja et al. (1985) has shown that among all the different composites, only those with PZT rods embedded in spurred epoxy matrix with regular periodicity (1-3 connectivity), appeared to be very promising on the ultrasonic transducer application The research revealed that PZT polymer composites have shown several remarkable advantages over conventional piezoceramic materials for ultrasonic applications. Findings from the investigation further revealed that 70-90% PZT in most composites is replaced by a low-density polymer, bringing in a better acoustic impedance matching to the human body.

Previous researches (McGrath et al., 1983; Gururaja et al., 1985) show that the volume fraction of PZT rod in composite, density of the composite and periodicity of the PZT rod in the composite are related. Until now, no existing mathematical expression has established this relationship. This unknown stance therefore prompted the need for the present work to fill in the gap.

The present work aims at mathematically deriving the dependence of PZT rod periodicity in the composite on the PZT rod volume fraction in composite and density of the composite. It is strongly believed that the model if derived will be able to predict the PZT rod periodicity, within the actual range, on just substituting into the model, the PZT rod volume fraction and density, providing the boundary conditions are considered.

II. MATERIALS AND METHODS

A. Materials and Methods

a. Sample Preparation

PZT powder of grain size between 1 and 3µm was used in this work to make rods of 1 mm diameter. The powder was supplied by the Shanghai Institute of Ceramics, having properties similar to the Vernitron (Morgan Matroc Ltd) PZT-4 composition. The rods were extruded through a home-made die and sintered at 1300°C for about 6 hours. To measure the piezoelectric properties of the ceramic component, a rod sample of length 0.78 mm was cut from a long-fired rod (Kwok et al., 1995).

A series of holes of 1 mm diameter were drilled in already cast epoxy sheets, used as the matrix, in a square pattern with center-to-center periodicity noted. The fired PZT rods were inserted into these holes, to glue the PZT rods to the polymer. After curing the composites, they were cut into disks of about 0.7 mm thick and 12 mm in diameter. The PZT rod volume percentage in the composite was 5%. The experiment was repeated for a PZT percent volume range 7-30%, and the density of composite and associated periodicity of the PZT rod in the composite evaluated (Kwok et al., 1995).

b. **Composite Density Evaluation**

The various densities of the PZT rod and epoxy resin were measured using the Digital Density Meter, after which the density of the PZT rod-epoxy resin composite (gm/cc) was calculated using the expression (J. Pattar and D. Ramesh, 2023);

$$\rho_c = V_r \times \rho_r + (1-V_r) \times \rho_m \qquad (1)$$

Where

ρ_c = Density of composite

V_r = Volume fraction of Reinforcement

ρ_r = Density of Reinforcement

ρ_m = Density of matrix

B. Model Derivation

Table 1: Variation of PZT rod periodicity in polymer structure with the density of the composite and volume percent PZT ceramic in composite

(ȣ)	(ρ)	(ɤ)
1.78	1.11	5
1.57	1.32	7
1.26	1.64	10
1.19	1.77	12
1.08	1.97	15
0.90	2.29	20
0.82	2.69	25
0.73	3.08	30

Computational analysis of the actual results shown in Table 1, gave rise to Table 2 which indicate that;

ȣ - Nɤ^-ƀ ≈ ẞρ^-ɵ - ɱ (2)

ȣ = ẞρ^-ɵ + Nɤ^-ƀ - ɱ (3)

The derived expression in (3) is an empirical model which predicts the periodicity of PZT-rod in piezoelectric ceramic-epoxy structure based on the density of the composite and volume percent PZT in composite. The model is a sum of two power functions. Comparative analysis of 1 and Table 2 indicates that ȣ, ɤ and ρ are periodicity of PZT-rod in polymer structure (mm), density of composite (gm/cc) and volume concentration of PZT in composite (%) respectively. The equalizing constants; N, ƀ, ẞ, ɵ and ɱ are 2.0292, 0.4991, 0.9862, 0.8962 and 0.001 respectively. They were generated using software (Nwoye, 2008).

III. RESULTS AND DISCUSSION

A. Boundary and Initial Conditions

Consider cylindrically shaped PZT rod the composite, interacting with the matrix, which is epoxy. The wave velocity of is assumed to be affected by the volume fraction and density of PZT rod in the composite. The considered range of the volume fraction of PZT in composite, range of density of the composite and PZT rod periodicity are 5-30%, 1.11 – 3.08 cc/gm and 0.73 – 1.78 mm respectively.

Table 2: Variation of ꞩ - Nɤ$^{-ƀ}$ with ßρ$^{-ɵ}$ - ɱ

ꞩ - Nɤ-ƀ	ßρ-ɵ - ɱ
0.8711	0.8971
0.8017	0.7679
0.6169	0.6320
0.6030	0.5902
0.5548	0.5361
0.4451	0.4683
0.4129	0.4053
0.3585	0.3589

B. Model Validity

In view of the derived model in equation (3), the validity is strongly pivoted on the core model structure in equation (2), expressed as ꞩ - Nɤ$^{-ƀ}$ ≈ ßρ$^{-ɵ}$-ɱ, both sides of which have close corresponding point values. Computational analysis of experimental results in Table 1 gave Table 2, being the confirmed mathematical outcome of the model structure, following closeness of the corresponding structure component values. Furthermore, the derived model was validated by comparing the derived model-predicted results with experimental, by carrying out deviational analysis and statistical analysis, involving evaluation of the correlations and standard errors.

C. Statistical analysis

a. Correlation

The correlation between the periodicity of PZT-rod in polymer structure and volume percent of PZT in composite & density of composite were calculated as 0.9994 and 0.9979, using model-predicted and experimental results respectively. This was based on the coefficient of determinants on the actual and model-predicted results, shown in Figs. 1 and 2, evaluated using equation (4).

$$R = \sqrt{R^2} \tag{4}$$

b. Standard Error (STEYX)

Statistical analysis of generated results indicates that the overall standard error on predicting the periodicities of PZT-rod in polymer structure (relative to the experimental results) is 0.023%, for every change in the volume percent of PZT in composites and density of composite. This translates into a model confidence level above 99%.

c. Graphical Analysis

Graphical analysis of Fig.1 and Fig. 2 show closely fitted and aligned curves of periodicities of PZT-rods in composites, relative to volume percent of PZT and its density in composite respectively.

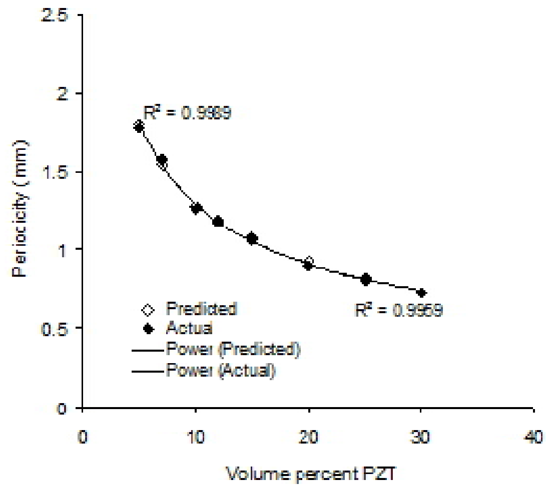

Fig.1: Comparison of the periodicities of PZT-rod in polymer structure (relative to the volume percent PZT in composite) from the actual and model prediction.

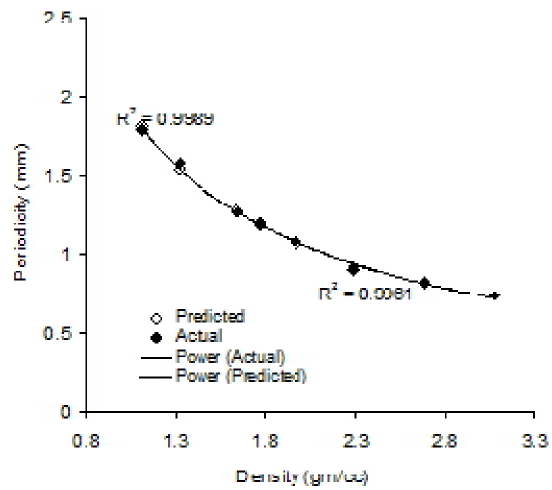

Fig. 2: Comparison of the periodicities of PZT-rod in polymer structure (relative to the density of composite) from actual and model-prediction

These curves represent experimental and model-predicted results. Each set of curves from the figures, excellently show similar trend & spread of results point distribution, and well fitted corresponding point values.

Curves from the figures indicate negative orientation, and will likely prompt negative slopes, baring the inverse variation of periodicities of PZT-rods in composites with volume percent of PZT in composite and density of composite respectively.

c. Deviational Analysis

The functionality and acceptability of a derived model largely depends on the deviation of model-predicted result from corresponding experimental value. Fig. 3 indicates sets of points on the curves, each of which gives the percent deviation between the experimental and the corresponding model-predicted results. The figure gives 2.56% as the overall maximum deviation of model-predicted periodicity of PZT-rod in polymer structure (from corresponding experimental results). This translates into over 97% operational model confidence level. The figure also shows that the least and highest deviations of the model-predicted periodicity of PZT-rod in polymer structure are 2.56 and 0.0

% respectively. Furthermore, these deviations correspond to periodicities of PZT-rod in polymer structure: 0.923 and 0.73 mm, volume percents PZT: 20 and 30% & densities of composite: 2.29 and 3.08 gm/cc respectively. The overall model confidence level therefore places between 93-99%, following evaluations from standard deviation, correlations and maximum deviation. Introduction of correction factors to the model-predicted results are necessary, to overcome the deviation and realize the experiment results.

The deviation Dv, of model-predicted PZT rod periodicity from the corresponding actual result was given by

$$Dv = \left(\frac{\mathcal{S}_m - \mathcal{S}_E}{\mathcal{S}_E} \right) x \ 100 \qquad (5)$$

Where

$\mathcal{S}_m$ and $\mathcal{S}_E$ are periodicities of PZT rod in polymer structure evaluated from actual and model-predicted results respectively

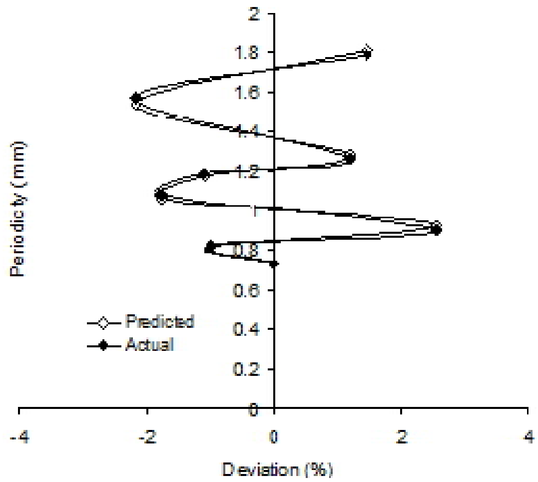

Fig. 3: Variation of model-predicted periodicity of PZT rod in composite with its corresponding deviation from experimental results

It is instructive to state that the model results deviation from corresponding experimental values prompted from, non consideration during model formulation, the effects of surface properties of the PZT rod and physicochemical interaction between the PZT rod and the epoxy matrix, all of which play vital roles during the polymer reinforcement process with PZT rod. This necessitated the introduction of correction factor, to bring the model-predicted PZT rod periodicity to those of the corresponding experimental values.

D. Periodicity of PZT-rod in polymer structure per unit volume percent PZT

Periodicity of PZT-rod in polymer structure per unit volume percent PZT $\mathcal{S}_\Upsilon$ mm/% was calculated from the expression:

$$\mathcal{S}_\Upsilon = \mathcal{S} / \Upsilon \qquad (6)$$

Re-written as

$$\mathcal{S}_\Upsilon = \Delta\mathcal{S} / \Delta\Upsilon \qquad (7)$$

Equation (7) is detailed as

$$\mathcal{S}_\Upsilon = \mathcal{S}_2 - \mathcal{S}_1 / \Upsilon_2 - \Upsilon_1 \qquad (8)$$

Where

$\Delta\mathcal{S}$ = Change in the periodicities of PZT-rod in polymer structure $\mathcal{S}_2, \mathcal{S}_1$ at two volume percents PZT Υ_2, Υ_1

Plotting points (5, 1.78) & (30, 0.73) and (5, 1.806) & (30, 0.73) as shown in Fig.1, designated as ($ɤ_1$,$ᶊ_1$) and ($ɤ_2$, $ᶊ_2$) for experimental and model-predicted results, and substituting them into equation (8) gives the slopes: -0.042 and – 0.043 mm/%, as their respective periodicity of PZT-rod in polymer structure per unit volume percent PZT.

E. Periodicity of PZT-rod in polymer structure per unit density of composite

Periodicity of PZT-rod in polymer structure per unit density of composite $ᶊ_\rho$ mm/(gm/cc) was calculated from the expression;

$$ᶊ_\rho = ᶊ/\rho \qquad (9)$$

Re-written as

$$ᶊ_\rho = \Delta ᶊ/\Delta\rho \qquad (10)$$

Equation (10), is detailed as

$$ᶊ_\rho = ᶊ_2 - ᶊ_1/\rho_2 - \rho_1 \qquad (11)$$

Where

$\Delta ᶊ$ = Change in the periodicities of PZT-rod in polymer structure $ᶊ_2$, $ᶊ_1$ at two densities of composite ρ_2, ρ_1

Similarly, on plotting points (1.11, 1.78) & (3.08, 0.73) and (1.11, 1.806) & (3.08, 0.73) shown in Fig. 2, designated as (ρ_1,$ᶊ_1$) and (ρ_2, $ᶊ_2$) for experimental and model-predicted results, and substituted into the (11), gives the slopes: -0.533 and – 0.546 mm/ (gm/cc), as their respective periodicity of PZT-rod in polymer structure per unit density of composite.

The negative signs preceding the respective periodicities of PZT-rod in polymer structure, per unit volume percent PZT and density of composite, just indicate that their respective curves as shown in Fig 1 and Fig. 2 are positioned in the negative plane. Their real values are the magnitudes of the respective slopes.

IV. CONCLUSION

Response of PZT-rod periodicity in piezoelectric polymer structure to the density of the composite and volume percent PZT in composite was evaluated, using a derived empirical model; $ᶊ = ß\rho^{-ɘ} + Nɤ^{-ƀ} - ɱ$. The validity of the model is strongly rooted on the core model structure; $ᶊ - Nɤ^{-ƀ} \approx ß\rho^{-ɘ} - ɱ$, both sides of which are correspondingly near equal. The model-predicted results agree with previous research on the inverse relationship between the PZT rod periodicity and density of composite & volume fraction of PZT rod in composite. The correlation between the periodicity of PZT-rod in polymer structure and volume percent of PZT in composite & density of composite were all > 0.99. The standard error incurred in predicting the model based PZT rod periodicity, relative to the actual results was < 0.023%, implying over 99% model confidence level. The periodicity of PZT rod per unit volume fraction of PZT rod in composite and per unit density of the composite were -0.042 & – 0.043 mm/% and -0.533 & – 0.546 mm/ (gm/cc), using experimental and model-predicted results respectively. The overall maximum deviation of the model-predicted periodicity of PZT rod from actual results was 2.56%. The derived model will predict the periodicity of PZT rod, within the actual results range, on substituting into the model, values of the density of composite and volume fraction of PZT rod in composite, providing the boundary conditions are considered.

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