A NOVEL PIEZOELECTRIC PAINT SENSOR FOR NON DESTRUCTIVE TESTING

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ABSTRACT

Piezoelectric paint is a composite piezoelectric material which is comprised of tiny piezoelectric particles in polymer matrix. It is flexible and could be easily applied on a curved structural surface when compared with brittle piezoelectric ceramic sensors. The exploration history of the piezoelectric composite can be traced back to 1980s, when the research group of Newham reported the preparation of piezoelectric paint from a water-based suspension of electroceramics and polymer. This was followed by the research works performed by Egusa, who explored the health-monitoring capabilities of the piezoelectric paint. Recently Zhang reported the piezoelectric paint fabrication and extended the application to in situ fatigue crack detection and acoustic emission.

This work investigates a cost-effective method to produce the piezoelectric paint, the tape casting method, which is feasible in lab environments. The piezoelectric paint is produced and its microstructure is investigated under a scanning electron microscope. Two important physical properties, the modulus of elasticity and sensitivity are measured through experimental tests. Finally the piezoelectric charge constant $d_{31}$ is calculated from experimental data, the $d_{31}$ is an important value when applying the piezoelectric paint as vibration sensors or acoustic emission sensors in non-destructive testing.

\textit{Keywords:} Piezoelectric composite, Piezoelectric properties, Non-destructive testing
INTRODUCTION

Piezoelectricity refers to the phenomenon existing in certain materials, which generates an electric field when the material is subjected to a mechanical stress, and conversely, induces a mechanical strain in response to an applied electric field. The former is called direct piezoelectric effect and the later converse piezoelectric effect. Started from 1880 of its undoubted discovery, the piezoelectricity has grown to be a significant commercial importance in many fields. The piezoelectric sensors, which utilize the direct piezoelectric effect, have been widely used in non-destructive testing for flaws and cracks detection.

The traditional piezoelectric sensors used in non-destructive testing is piezoelectric ceramic sensors, this involves adhering the ceramic sensors on the structural surface and investigating the electronic signals received from sensors. Due to the physical properties of the ceramic sensors, such as hard, brittle, they can only withstand very small bending moment, thus the host structural surface must be flat and smooth, in order to ensure a robust and seamless adhesion and a confident measuring result. However, in many cases the structural surfaces are curved, these are usually seen in water pipelines, welding joints, etc. In these cases, the conformability of the ceramic sensor and the host structure is very poor, and extra treatments are required. Fig. 1 shows an adhered piezoelectric ceramic sensor on a steel tube, in order to make a strong conformability, the tube is pre-treated to make a flat contacting area for the sensor. This in fact brings change to the host structure and potential damages are also introduced in.

![Fig. 1: A piezoelectric ceramic sensor adhered on a steel tube](image)

In order to compensate the conformability problems of piezoelectric ceramic sensors, another group of piezoelectric composite, the piezoelectric paint has been explored. As indicated by the name, the piezoelectric paint is flexible, distributable and conformable. It is a composite piezoelectric material which is comprised of tiny piezoelectric particles and polymer matrix. The exploration history of the piezoelectric composite can be traced back to the 1980s, when the research group of Newnham and coworkers reported the preparation of piezoelectric paint [1, 2]. In these works, they prepared the composite paint from a water-based suspension of...
electroceramics and polymer, sprayed it on a brass plate and poled under 10-15kV/mm. They also measured the average value of the piezoelectric charge constant $d_{33}$ by a Piezo $d_{33}$-meter. Followed by the research works performed by Egusa and Iwasawa in the 1990s [3, 4], they prepared the piezoelectric paint by using PZT ceramic powder as pigment and epoxy resin as binder, spread the mixture on an aluminum beam surface with a paint brush and dried in air at room temperature, a screen mask was used to print electrodes on the paint surface and applied the thin film as vibration sensors. Later Egusa et al. also explored the health-monitoring capabilities of the piezoelectric paint [5]. Based on these works, improvements were done by the research group of Hale, who combined fine milled PZT powder with acrylic lacquer to make the piezoelectric paint, they used air brushes to spray the paint on a surface [6], and also characterized the piezoelectric paint through tensile test, dynamic test and environmental test [7]. Followed Zhang et al. reported the piezoelectric paint fabrication and extended the application to in situ fatigue crack detection [8] and acoustic emission [9]. Lately M. Dietze and M. Es-Souni reported the fabrication of PZT-PVDF-TrEF composites by screen-printing method on indium-tin-oxide-coated (ITO) glass substrate, and characterized the film by SEM and X-ray diffraction [10].

Although the fabrication and application of piezoelectric paint have been continuously reported, a systematic characterization of it is still incomplete. This work describes the fabrication of the piezoelectric paint, followed by its microstructure characterization. To fully understand the piezoelectric properties, its Young’s modulus and sensitivity are also investigated by various experimental methods, from these experimental data the piezoelectric charge constant $d_{33}$ could be deduced.

## PIEZOELECTRIC PAINT PREPARATION

The previous published works have showed that the piezoelectric paint is mainly composed of piezoelectric ceramic powder and polymer resin. The former is as a pigment and the later is as a binder. The piezoelectric ceramic powder used in this study is a commercial product. The powder mainly includes PZT added with ZrO2, PbO, TiO2, etc. After twice spray pelletization, the powder particle size is controlled within 1µm. The ceramic powder is mixed with plasticizer, defoamer, and solvent, the mixture is ball milled for 2 hours, which gives enough energy to separate the ceramic powder into the polymer matrix. Experimental tries have shown that the ball milling time for at least one hour and a half is necessary, less than this the ceramic powder cannot be completely and evenly distributed in the solvent, which leads to precipitation and air void in the paint. The weight percentage of ceramic powder is chosen up to 60%, more than this the slurry is too thick to be casted on the substrate and leads to a fabrication failure.

The prepared piezoelectric slurry is tape casted on a substrate. Generally, the tape casting method is a popular and important ceramic form technique which is widely used in industries to produce thin sheets of flexible tape. It produces thin films inexpensively and therefore is widely accepted due to its suitability, simplicity and low implementation cost when compared with the other available methods, such as dry pressing, slip casting, or injection molding. The tape casting method has been adopted to manufacture a variety of multi-layer ceramic package products, as well as non-ceramic, polymer and metal applications [11]. Based on these experiences of tape casting which have been developed for years, a lab applicator was design to perform this technique. The casted paint is set aside for complete drying for more than 24 hours and peeled.
off from the substrate. Afterwards a conductive paint is applied on both sides of the piezoelectric paint for polarization. The polarization process is done in an oil bath with an electric field up to 5kV/mm, although it is suggested to polarize the piezoelectric composites in alleviated temperature above 80°C, this is not applicable for the piezoelectric paint, since the high temperature accelerates the polymer aging. Fig. 2 (b) shows a piezoelectric paint sample, and Fig. 2 (b) demonstrates the high flexibility of the piezoelectric paint, as could be seen the piezoelectric paint conforms well on the steel tube without any damage.

Fig. 2: (a) A piezoelectric paint sample (b) the piezoelectric paint conforms on a steel tube

PIEZOELECTRIC PAINT MICROSTRUCTURE

As the quality of the casted sample almost always determines whether it could be polarized or not, the microstructure of the prepared piezoelectric paint is investigated under a scanning electronic microscope. Fig. 3(a) shows a smooth and homogenous piezoelectric paint and Fig. 3 (b) is its zoom. As could be seen, in this sample the piezoelectric ceramic particles are evenly distributed in the polymer matrix, and there is no air void or flaw, in this case, the sample could be successfully polarized under high electric field. Contrarily, casting failure or unevenly mixed slurry leads to severe air void and structural fissure. The dark parts in Fig. 3(c) are the air void, Fig. 3(d) is the close up of the air void. The flaws cover large area of the piezoelectric surface, this failure results in dielectric break down under polarization and are greatly avoided.

When poling, electrodes have to be applied on both sides of the piezoelectric paint. This is realized by directly applying a conductive paint on the sample surface. The conductive paint is composed of µm-range silver particles and adhesive, it is very thin and dries out quickly. In order to confirm that the conductive paint is not going to infiltrate into the piezoelectric paint and spoil the physical properties of the piezoelectric paint, the cross section of the sample is also investigated. The sample in Fig. 4(a) is covered with the conductive paint, as could be seen the polymer matrix is very dense and there is no infiltration of the conductive paint into the sample. Fig. 4(b) indicates it more clearly, the bright particles are the piezoelectric ceramic, and in between them is the polymer matrix, no other substance exists.
Fig. 3: SEM micrographs of piezoelectric paint surfaces

Fig. 4: SEM micrographs of a piezoelectric paint cross section

PIEZOELECTRIC PAINT PROPERTIES

Mathematical model

In many engineering field applications which involve in-plane strain, e.g. acoustic emission, vibration, the piezoelectric charge constant $d_{33}$ is an important value to know, which indicates the induced polarization in direction 3 per unit stress in direction 1, or induced strain in direction...
1 per unit electric field applied in direction 3. Fig. 5 shows the coordinate system and the piezoelectric paint polarization direction.

![Fig. 5: Coordinate system and polarization direction](image)

To deduce $d_{31}$, first of all, an analytical model of the piezoelectric paint is considered. Assuming that the piezoelectric paint is under low mechanical stress, by which the material behaviour can be considered linear, and we assume the piezoelectric paint is isotropic in in-plane, because the piezoelectric paint is poled in the out-of-plane direction, the electric displacement could be rewritten as,

$$ D_3 = d_{31} \sigma_1 + d_{32} \sigma_2 $$  \hspace{1cm} (1)

where $\sigma_1$ and $\sigma_2$ are the two in-plane stresses. The in-plane isotropic assumption leads to,

$$ D_3 = d_{23}(\sigma_1 + \sigma_2) $$  \hspace{1cm} (2)

and consider again the stress and strain relationship for a two-dimensional isotropic material,

$$ \begin{bmatrix} \epsilon_1 \\ \epsilon_2 \end{bmatrix} = \frac{E}{1 - \nu^2} \begin{bmatrix} 1 & \nu \\ \nu & 1 \end{bmatrix} \begin{bmatrix} \sigma_1 \\ \sigma_2 \end{bmatrix} $$  \hspace{1cm} (3)

The strain sensitivity of the piezoelectric paint could be defined as a ratio of the electric displacement and the biaxial strains, which is the electric displacement per unit of strain,

$$ S = \frac{D_3}{\epsilon_1 + \epsilon_2} $$  \hspace{1cm} (4)

therefore,

$$ d_{21} = \frac{1 - \nu}{E} S $$  \hspace{1cm} (5)

If $E$ and $S$ is known, $d_{31}$ could be calculated from (5). The following subsections will describe the experimental measurements of $E$ and $S$.

Elastic modulus

The elastic modulus $E$ of the piezoelectric paint is measured through a tension testing. In this case five tensile specimens with weight percentage of 50% are prepared to ensure repeatability,
they are shaped according to DIN EN ISO 527-1 [13]. Since the sample is soft and thin, direct placement in the testing machine is difficult and might bring damages beforehand, to solve this problem, a second clamp is designed between the specimen and the testing machine grip. Fig. 6(a) shows that a load cell is applied in between the upper part of the second clamp and the tensile head, it is connected to a carrier-frequency amplifier Picas for signal conditioning. As the sample is stretched, the force exerted by the instrument and the displacement of the sample are measured, this data is used to construct a stress-strain curve, by which the elastic modulus could be calculated. Fig. 6(b) is the stress-strain curve of a sample which is converted from the load-elongation data. In Fig. 6(b) the initial portion of the curve is incompatible with Hooke's law, it could be explained that due to the paint is very flexible and soft, it is very difficult to adjust it in the machine with zero stress and strain, however as it elongates, the stress and strain show almost linearity, this is indicated by the straight line in Fig. 6(b), the slope of the linear portion is the elastic modulus. The averaged elastic modulus of the five samples is 27 ± 15 MPa. In fact it should be noticed that quantifying the elastic modulus accurately is almost impossible, however this result gives a reference for future calculations.

![Fig. 6: Tensile test (a) test set up (b) measured stress-strain curve](image)

**Sensitivity**

As indicated in (5), the sensitivity of the piezoelectric paint could be deduced if the electric displacement and the biaxial strain could be measured simultaneously. The idea of the experimental measurement of sensitivity is from [13], which is realized by adhering the piezoelectric paint on an aluminum cross, on the same position but another side of the cross, a rosette strain gauge is applied, as shown in Fig. 7(a). The strain gauge is used to measure the principal strains $s_1$ and $s_2$. A test bed which is similar to the one described in [13] is constructed. The test bed consists of two 200mm*200mm*6mm aluminum plates, in between the plates there are eight steel bars which are placed as shown in Fig. 7(a). Fig. 7(b) illustrates the exact positioning of the aluminum cross and the steel bars. The set up ensures that the bending moment on the piezoelectric paint will be constant. Fig. 7(c) is the constructed test bed.

An impact is applied on the top aluminum plate by a hammer with rubber tip, the output signal of the piezoelectric paint goes into a charge amplifier, and the signals of the strain gauge
go to a four channel carrier-frequency amplifier, the outputs of the two amplifiers are measured by a handyscope HS4. Fig. 8 (a) and (b) shows the outputs of the strain gauge and the piezoelectric paint. From Fig. 8(a), the principle strains $s_1$ and $s_2$ could be calculated. It could be seen from Fig. 8(b) that the piezoelectric paint has a similar response as the strain gauge. The scatter plot in Fig. 8(c) shows the relationship between the electric displacement and sum of the principle strains. By making a linear regression, it indicates that the two shows linearity in the measuring range, therefore the slope represents the sensitivity $S$. In this case, the sensitivity is $1.96 \times 10^{-4} \text{C/(m}^2\text{)}$/m).

![Fig. 7: (a) Assembling schematic of the test bed (b) two dimensional illustration of the test bed (c) constructed test bed](image)

Piezoelectric charge constant $d_{31}$

As it has been predicted by (5), $d_{31}$ can be calculated as long as $E$ and $S$ is known. Therefore, by replacing $E$ and $S$ back in to (5), and assuming $\nu$ is 0.3, $d_{31}$ is estimated as $6.61 \pm 4.56 \text{pC/N}$. This result is close to the $d_{31}$ predicted by Li [14] (4.4pC/N), in which he used a percolation based mixed model to predict the piezoelectric paint constants. In addition, this value is comparable with the piezoelectric charge constant of the other piezoelectric polymers.

CONCLUSION

In this work, a new material, the piezoelectric paint, has been characterized. SEM is used to investigate the microstructure of the piezoelectric paint, to understand the separation of the piezoelectric particles in the polymer matrix. By deducing a relationship between the piezoelectric charge constant $d_{31}$, Young's modulus $E$ and sensitivity $S$, and measuring the latter two, the piezoelectric charge constant $d_{31}$ could be calculated. The result agrees well with a published result and gives more understanding of the piezoelectric paint, which is very useful when applying the it in non-destructive testing. However, it should be noticed that accurate characterization of the piezoelectric paint is still difficult, due to many inaccuracies in the manufacturing and measuring process.

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Fig. 8: (a) Strains measured by the rosette strain gauge (b) electric displacement (c) relationship between the sum of the principal strains and electric displacement

REFERENCES


