FLEXIBLE LOW TEMPERATURE PIEZOELECTRIC FILMS FOR HARVESTING FROM TEXTILES

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Abstract: This paper reports new flexible low temperature PZT-polymer composites used for energy harvesting from textiles. The materials are screen-printable which gives the advantage of low-cost and suitability for mass-production. The PZT powder content was varied in three polymer binders and the resulting composites were denoted ECS-PolyPZT 1, 2 and 3. ECS-PolyPZT 2 with 45% PZT was found to be the most flexible material that achieved an initial d_{33} of 22 pC/N. Following optimization of the poling parameters, the d_{33} improved to 27 pC/N. This material is suitable for energy harvesting from fabrics enabling applications to be explored in the future.

Keywords: Piezoelectric, flexible, low temperature, fabric, kapton, energy harvesting

INTRODUCTION

When a piezoelectric materials is strained an electric charges is generated [1] and this feature is widely exploited in sensors and energy harvesters. Piezoelectric energy harvesters are the most widely researched form of energy harvester with considerable effort spent on improving piezoelectric material properties and harvester performance. There is some research on flexible piezoelectric harvesters concerned with a human-motion applications for powering portable micro-power devices but these use complex fabrication techniques [2-4]. This paper reports flexible piezoelectric materials that can be screen printed. Techniques such as screen-printing thick films are straightforward processes suitable for large mass production and low cost and are compatible with fabrics.

The motivation for this work is to develop a piezoelectric material that can be printed onto flexible substrates and fabrics. This requires printed films to be flexible and suitable for processing at low temperatures. Higher processing temperatures will limit the range of fabrics that can be used.

Piezoelectric materials are typically ceramics (e.g. lead zirconate titanate (PZT)), polymers (e.g. PVDF) or composites (i.e. ceramic piezoelectric particles in a polymer matrix [5]). Only piezoelectric polymers and composites provide sufficient flexibility and suitably low temperature curing for fabric applications and of these, composites are lower cost than single-phase piezoelectric polymers.

The electrical power generated by a piezoelectric energy harvester will depend upon the level of piezoelectric activity of the material (e.g. d_{33}) and the design of the harvester. Piezoelectric polymers such as PVDF provides a d_{33} between 20-30 pC/N [6].

Screen-printed low-temperatures piezoelectric composites have been reported [7-9] but previous works were simply concerned with the level of piezoelectric activity and did not investigate their flexibility.

This paper presents the optimization of screen-printable, low temperature piezoelectric composite materials with respect to piezoelectric activity and flexibility. The piezoelectric, dielectric and mechanical properties are investigated for the proposed materials.

EXPERIMENTAL

Materials Formulation

The proposed materials are mixtures of PZT ceramic powder (Pz29, Ferroperm Piezoceramics) mixed with a range of polymer materials. Two different particle of PZT powder (2 and 0.8 μm) were first mixed with a powder ratio of 4:1 [10]. The blended PZT powder was then mixed with one of the polymers binder.

Three different polymers were evaluated and the piezoelectric composites have been denoted ECS-PolyPZT 1, 2 and 3. The ratio (by weight) of polymer binder to PZT ceramic powder was varied and the piezoelectric and mechanical properties determined. The weight percentages of PZT powder investigated in ECS-PolyPZT 1 and 3 were 60, 70, 80 and 90%. The PZT weight percentages for ECS-PolyPZT 2 were 30, 45, 60 and 70% and could not be increased further because the paste became unprintable. In total, 12 piezoelectric composites were formulated.

The pastes were triple roll milled in order to disperse the PZT powder homogenously within the polymer. The result is a high quality smooth paste that can easily be screen-printed.

Screen-Printing of the Materials

A capacitor structure with the piezoelectric material sandwiched between two electrodes was screen-printed using a DEK 248 screen-printer on both Alumina and Kapton polyimide 300 HN substrates, thicknesses 635 and 75µm respectively, to investigate the piezoelectric properties and flexibility. Top electrodes were not printed on the Kapton samples in order to investigate flexibility as discussed later.

The bottom and top electrodes were printed using low temperature silver polymer paste ELX 30 (from Electra Polymers). The curing temperature of the materials was determined by the polymer as it is processed at lower temperature levels compared to PZT. All the materials have been cured in box oven at conditions shown in Table 1. All the curing conditions are suitable for fabric applications.

Table 1: Curing conditions of the printed materials

	Curing temperature (^{o}C)	Curing time (min)
ECS-PolyPZT 1	130	10
ECS-PolyPZT 2	125	10
ECS-PolyPZT 3	130	10
ELX 30 Silver/polymer	125	5

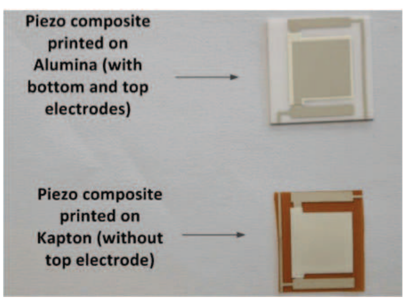


Fig. 1: The capacitive devices after printing on both Alumina (top) and Kapton (bottom)

Poling the Devices

The piezoelectric film needs to be poled to activate piezoelectric properties of the material. The poling process can be achieved by applying an electric field across the piezoelectric film at elevated temperatures for a certain time. The electric field aligns the dipoles and the heating provides the energy for the dipoles to realign.

For poling the printed devices, direct-contact method was chosen which involves applying an electric voltage across the piezoelectric film with the aid of the bottom and top electrodes. The electric field can be calculated according to the thickness of the film by the following equation $E = v/d$, where E, V and d are the electric field, voltage applied across the electrodes and the thickness of the piezoelectric film, respectively.

RESULTS AND COMPARISONS

Permittivity and PZT Content

A higher permittivity is beneficial because it allows for greater poling fields. Fig. 2 shows the permittivity of all composites at different weight percentages of PZT powder. This shows that for ECS-PolyPZT 2 the permittivity falls beyond 45% PZT. This is because the porosity films increases with increasing concentrations of PZT and the trapped voids reduce the measured permittivity. Fig. 3(a) shows the film cross section of ECS-PolyPZT 2 45% PZT which clearly has fewer voids than the 70% version in Fig. 3(b). The permittivity of ECS-PolyPZT 1 and 3 were found to be less sensitive to variations in PZT concentration with small reductions being evident beyond 80 and 70% respectively.

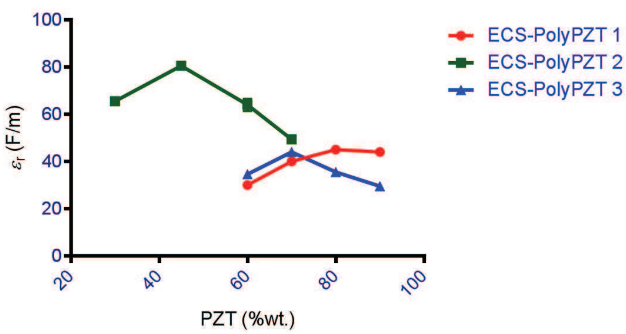
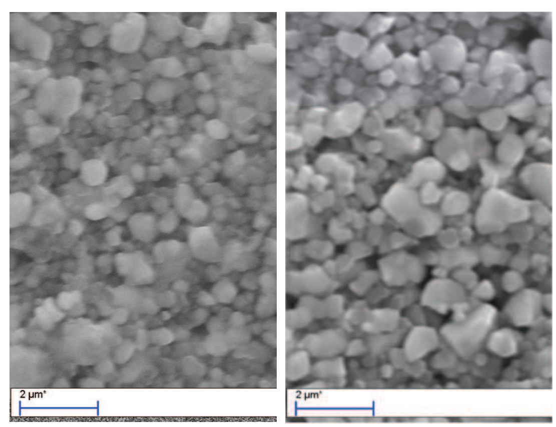


Fig. 2: Permittivity versus percentage of PZT by weight in the composites



(a) *(b)*

Fig. 3: SEM images of (a) ECS-PolyPZT 2 (with 45% PZT powder by weight) and (b) ECS-PolyPZT 2 (with 70% PZT powder by weight) composites

Piezoelectric Coefficients and PZT Content

Piezoelectric properties were also investigated for different concentrations of PZT by measuring the d_{33} coefficient of 5 samples of each of the PZT percentages. Five d_{33} measurements were taken from each sample giving 25 measurements in total. The average of these 25 d_{33} measurements is plotted in the graph shown in Fig. 4. Alumina samples only were used for this study. All the devices were poled with an electric field E = 6 MV/m, temperature T = 160 °C and time t = 10 min. The d_{33} measurements were performed by using a piezometer (PM35, PiezoTest).

The results shown in Fig. 4 indicate that the maximum piezoelectric coefficients obtained for ECS-PolyPZT 1, 2 and 3 are at 90%, 60% and 90% respectively (around 17, 25 and 20 pC/N).

For ECS-PolyPZT 2, the maximum d_{33} was obtained at 60% PZT by weight. However, a drop in d_{33} is observed when the weight percentage of PZT reached 70%. This is because of the increased film porosity at this percentage.

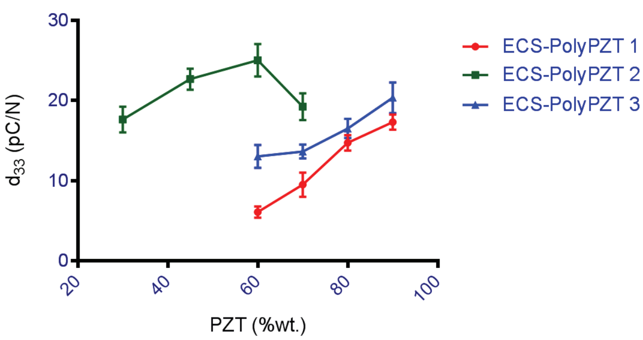


Fig. 4: Piezoelectric coefficients versus percentage of PZT by weight in the composites. Poling parameters were taken at fixed conditions (E = 6MV/m, T=160 °C and t= 10 min)

Flexibility and PZT Content

The flexibility of the Kapton devices was tested by bending the samples around different diameter formers and observing the minimum radius of curvature the film can withstand before failure. The films fail when cracks or wrinkles are observed on the film surface using a microscope. The most flexible formulations were found to be ECS-PolyPZT 2 with PZT weight percentages 30, 45, 60 and 70% and ECS-PolyPZT 1 (80% PZT). These films withstood bending without a former which is a radius of curvature of 75µm (the thickness of the Kapton). The most flexible ECS-PolyPZT 3 variant was found to be 70% PZT which survived a 1 mm radius of curvature.

The most flexible films were further tested by observing the number complete bending cycles without a former the materials can withstand (see Fig. 5). ECS-PolyPZT 2 inks with 30%, 45%, 60% and 70% PZT failed after complete bending 8, 4, 2 and 2 cycles, respectively. ECS-PolyPZT 1 80% PZT failed after 2 cycles.

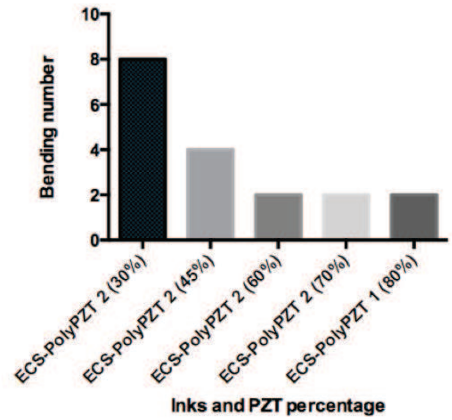


Fig. 5: Number (cycles) of complete bending test

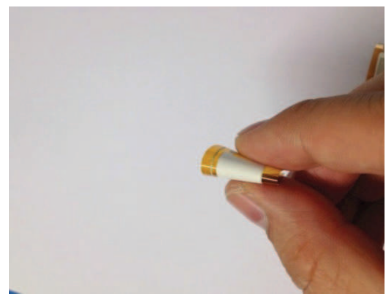


Fig. 6: Showing the flexibility of the printed materials

Optimum Material

The optimum material in terms of binder type and PZT percentage was found to be ECS-PolyPZT 2 with 45% PZT. This was found to have the best combination of piezoelectric activity and film flexibility. The Young's Modulus of this material was found to be 0.723 MPa.

Poling Parameters Optimization

The poling parameters were optimized for the optimum material ECS-PolyPZT 2 45% PZT. Four samples were used for poling optimization and five measurements have been taken from each. Initially, the poling temperature was investigated with the electric field and poling fixed at E= 4 MV/m and t = 10 min. The results in Fig. 7 show that the higher the poling temperature, the higher d_{33}. Beyond 225 °C the samples were found to become short circuited.

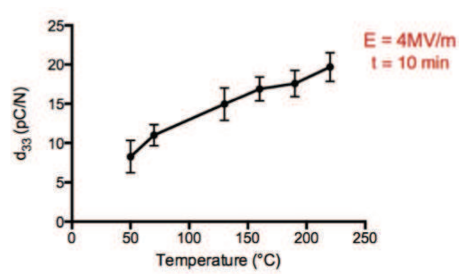


Fig. 7: Poling temperature optimization

Next the poling time was investigated with the electric field and temperature fixed at 4 MV/m and 160 °C, respectively. The optimum temperature (225 °C) is too hot for many fabrics and therefore 160 °C was used.

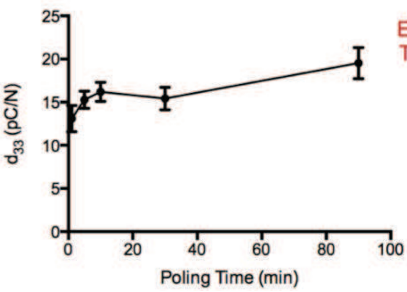


Fig. 8: Poling time optimization

The influence of poling time on d_{33} is shown in Fig. 8 with the maximum d_{33} being obtained after 90 minutes. However, the increase in d_{33} beyond 10 minutes was quite small and does not justify the time taken. Therefore the optimum poling time is 10 minutes.

Finally, the electric field was optimized with the temperature and time fixed at 160 ^{o}C and 10 minutes. The results in Fig. 9 show the highest average value of d_{33} was 27 pC/N with an electric field of 10 MV/m, which is an increase of 5 pC/N on the un-optimized value.

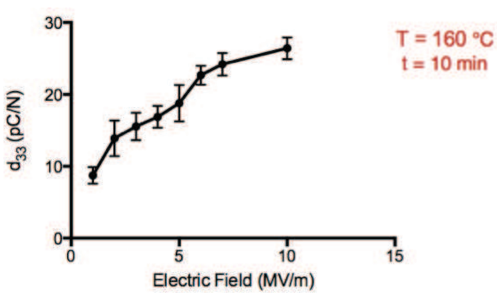


Fig. 9: Poling electric field optimization

CONCLUSION

Three types of polymer binder were used to form screen printable, flexible, low temperature PZT-polymer composites (denoted, ECS-PolyPZT 1, 2 and 3). The PZT powder content was varied and piezoelectric properties and film flexibility have been studied. Each composite has four formulations enabling 12 materials to be evaluated.

From the 12 materials, one has been identified as the most suitable material with an initial d_{33} of 22 pC/N and can withstand complete folding 8 times before failing. After poling optimization its d_{33} increased to 27 pC/N. This material is suitable for use on a wide range of fabrics and will now be used to develop fabric based energy harvesters.

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