COEXTRUSION OF POLYMERIC PIEZOELECTRIC FILAMENTS

R.S.Martins and J.M.Nóbrega, IPC/I3N – Institute for Polymers and Composites, Univ. of Minho, Portugal
R.Gonçalves, M.P.Silva, and S.Lanceros-Mendez, Centro/Dep. de Física, Univ. of Minho, Portugal
J.G.Rocha, Dep.t of Industrial Electronics, Univ. of Minho, Portugal
H.Carvalho, Centre for Textile Science and Technology, Univ. of Minho, Portugal

Abstract

The difficulties related to the development of industrial scalable production methodologies have limited the number of applications currently available in the field of interactive/electronic textiles, which are far below the anticipated a few years ago.

In these areas the integration of piezoelectric materials, that possess sensing/actuating capabilities, such as poly(vinylidene fluoride), PVDF, and can be processed using conventional processing techniques, is very promising and has encouraged a large number of research works. However, until now, most of the developed production methodologies are difficult to adapt to the industrial scale.

This work reports recent developments achieved in the framework of a research project, on the production of piezoelectric filament by coextrusion of PVDF and an electrically conductive thermoplastic. The developed production methodology is based on a conventional coextrusion line, for which a coextrusion die was designed to produce a multilayer filament, which comprises an inner layer of an electrically conductive Polypropylene grade and a middle layer of PVDF and is coated with an electrical conductive ink. The sensing capabilities of the produced filaments are also characterized.

Introduction

The exceptional pyro- and piezoelectric properties of Poly(vinylidene fluoride) (PVDF) motivated the interest of both industrial and academic communities, and its use in several applications that benefit from its sensor or actuator characteristics [1-3]. The PVDF properties depend both on its degree of crystallinity and orientation of its crystalline phase, which are strongly dependent on the processing conditions employed during production [1, 3-12].

PVDF is a polymorphic material that presents at least four crystalline phases, known as α, β, γ and δ. The α-PVDF, which is non-polar, is obtained during cooling of the melt at high or moderate cooling speeds [1, 3]. From the point of view of its electrical activity, the β-PVDF is the most effective, and can be obtained through the stretching of the α-PVDF at 80ºC using a stretch ratio (R) between 3 and 5 [11, 12]. In order to optimize its electroactive properties, β-PVDF is subjected to a poling process, by the application of a strong electrical field [1-3], which further orients the crystallite dipolar moments. The obtained electrically active polymer, can generate an electrical potential when subjected to a mechanical excitation, or a mechanical action is produced when it is subjected to an electric field. These characteristics motivated the use of PVDF in sensors and actuators applications.

A polymeric piezoelectric device comprises at least one piezoelectric layer and two electrically conductive layers, one at each side of the central layer, which are used as electrodes for the connection of electronic conditioning/drive equipment.

In conventional processes piezoelectric PVDF based devices are produced in the form of films, starting from the extrusion of a PVDF layer that is subsequently stretched, poled and, finally, coated by metallization 13.

Some research works available in the literature focus on the development of filament shaped PVDF sensors. Walter et al. [14,15] have studied extensively the phase transitions of extruded PVDF monofilaments, in which a composite part of simple PVDF filaments and epoxy resin was poled with a linear electric field, in a direction perpendicular to the fibers. The produced composite was shown to exhibit piezoelectric activity. In the comprehensive work done by Lund et al. and Ferreira et al. [16] for two-layered filaments, it was shown that the electroactive phase content is not affected by the conductive inner core. Additionally it was concluded that the β-phase content depends only on the processing temperature and stretch ratio, as happens for the single PVDF filaments. Sequential processing methods for the production of piezoelectric cables have been described by Mazurek et al [17,18]. They show that the cooling at -30ºC of PDVF, after stretching, increase the β-phase content, and the coating required for the placement of the outer electrode promotes a significant reduction.

In this work, the development of a new filament form is proposed, which opens up a whole new range of applications, especially the ones that involve its use in e-textile applications.

In this new process, the electrode (electrical conductive) and PVDF layers are arranged in a coaxial manner, as illustrated in Figure 1.

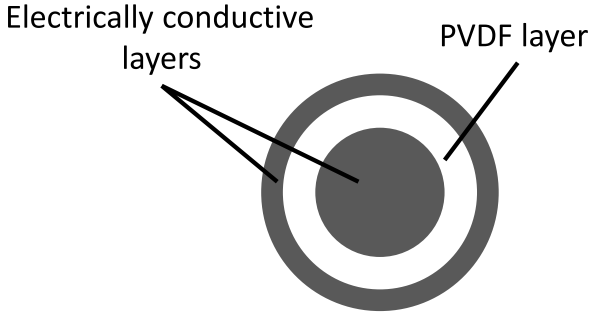

Figure 1. Cross section of the piezoelectric filament.

Piezoelectric Filament Production

As shown in Figure 2, the production of the piezoelectric filament starts with the coextrusion of the inner (electrode) and middle (PVDF) layers. For this work and electrically conductive polypropylene (Premix 1396) and an extrusion grade PVDF (Solef Ta-1010) were used for the inner and middle layers, respectively.

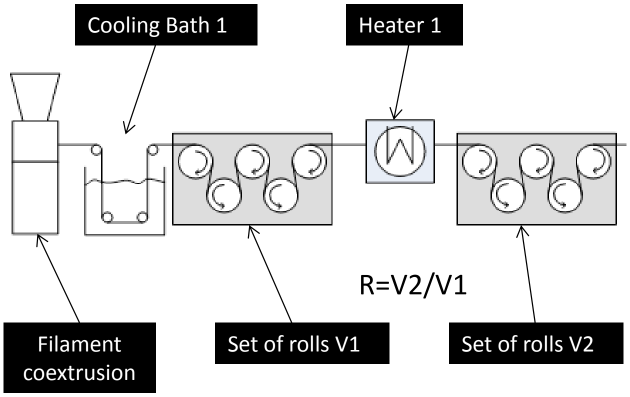

Figure 2. Schematic view of the extrusion line used to produce the two layered coextruded filaments.

The extrusion die used to produce the coextruded filaments is illustrated in Figure 3. As shown, the inner (Mi) and outer (Mo) material layers are combined to produce a two layer filament with a coaxial arrangement. To allow slight adjustments during production, the axial position of the part P1 can be controlled. As can be concluded from Figure 3, the position of part P1 directly affects the relative restriction of the two layer flows, and thus their relative thickness.

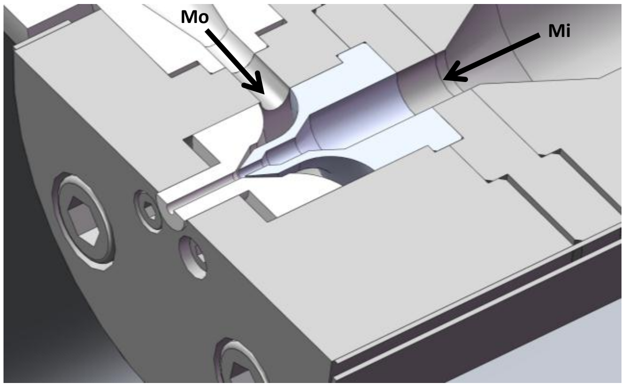

Figure 3. Cut view of the extrusion die developed to produce the coextruded two layer filaments.

The Set of rolls V1 (Figure 3), allow to control the linear velocity of the filament at the die exit and, consequently, its diameter. After leaving the die flow channel, the filament is cooled in a water bath to room temperature and is then heated in Heater 1 (Figure 3), which is a forced convection air temperature controlled oven. Inside Heater 1, when it reaches a temperature at which is deformable, the filament is stretched, due to the pulling force promoted by the set of rolls V2 (Figure 3), that have a linear velocity higher than the one at set of rolls V1. The draw ratio is determined by the relation between the linear velocities V2 and V1, respectively the set velocity for roll systems 2 and 1, and is given by R=V2/V1. It is important to mention that the cooling and heating steps are required to assure a better control of the temperature at which the filament draw takes place. At the end of the extrusion line the continuous produced filament is wound.

Subsequently the filament is coated with a silver conducting ink that forms its third (outer) layer. The two conductive layers are then connected directly to a voltage source and subjected to high voltage.. The voltage source’s protection automatically stops upon a sudden increase of current that resulted either from material breakdown or localized defects.

Piezoelectric Filament Characterization

For preliminary tests the filament in rest was manually compressed with a plastic part on a direction transversal to the filament axis.

Two different experimental set-ups were used to evaluate the electromechanical response of the produced piezoelectric filaments, in a more controlled manner.

In one set-up the filament is attached to a vibration generator (Figure 4), using a set of acrylic parts built for this purpose.

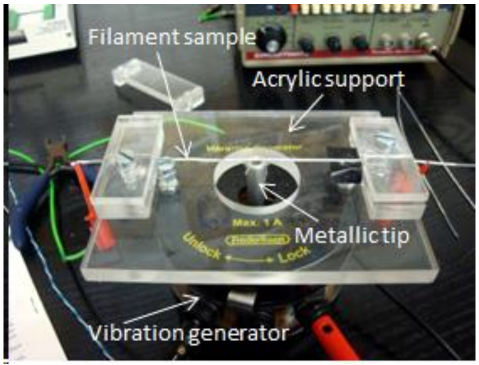

Figure 4. Electromechanical testing set-up based on a vibration generator

For the characterization test, the vibration generator is fed with a 0.5 Hz square wave, which results in an oscillating movement of the generator's metallic tip with an amplitude of circa 3 mm. The metallic tip displacement promotes the bending of the filament.

In the other characterization set-up the filaments were fixed to the grips of a universal testing machine, working in the tensile mode, and submitted to 20 loading-unloading cycles with an amplitude of 0.2 mm at a speed of 100 mm/min. (Figure 5).

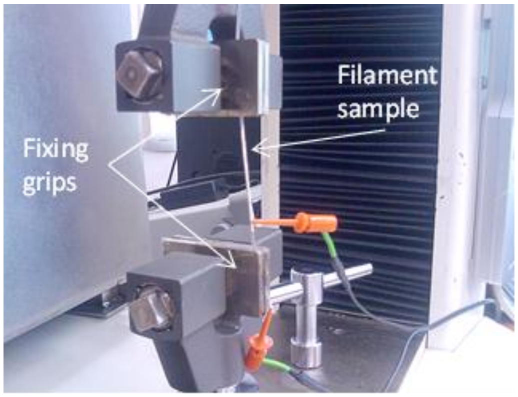

Figure 5. Stretching in universal testing machine

In order to measure the generated electrical signals the filaments were connected to a custom-built charge amplifier. The output signal of the amplifier was connected to a National Instruments NI-6259 data acquisition board, and custom-developed software based on Labview was used for signal acquisition, display and storage.

The acquired signals showed to be affected by some 50 Hz noise and its harmonics, which blurred the signals of smaller amplitude. Correct grounding and shielding of the conditioning electronics provided a significant reduction of this noise. A Butterworth 10th-order bandstop filter with a stopband between 45 and 55 Hz was implemented in the software. With this configuration even the signals with very small amplitude could be identified.

Results and Discussion

Figure 6 shows an example of a signal acquired when compressing the filament in the transversal direction and manually, with a plastic part. The response of the piezoelectric activity of the filament is evident.

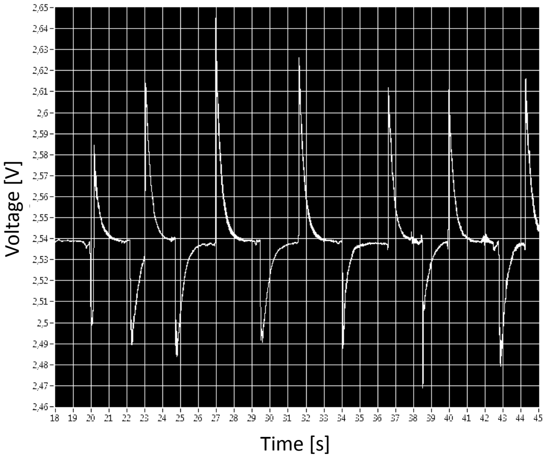

Figure 6. Electrical signal produced by the manual compression of the filament, with a plastic part, in the transversal direction

The measured signal is typical for a piezoelectric sensor coupled to a charge amplifier. Piezoelectric sensors generate signals only when subjected to dynamic stimuli, while static forces do not produce any signal. The charge amplifier picks up the signal generated during variations in a capacitor present in the amplifier's feedback loop, but the capacitor discharges through the feedback resistor with an adjustable time constant. This time constant can be chosen according to the application.

Figure 7 shows the signals acquired during the vibration generator test. The signal amplitude is in this case of about 25 mV.

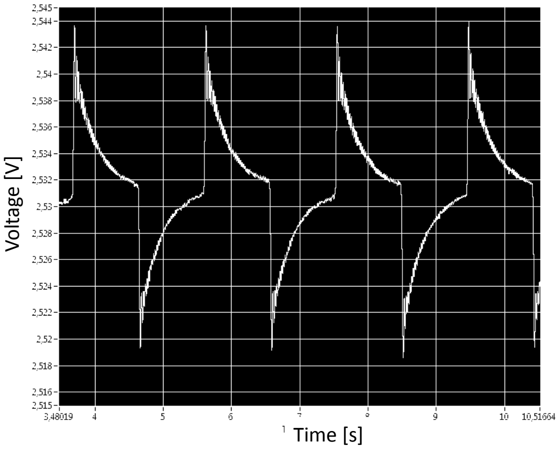

Figure 7. Electrical signal produced during the vibration generator test

The axial deformation induced during the universal testing machine test, produced the typical signal shown in Figure 8.

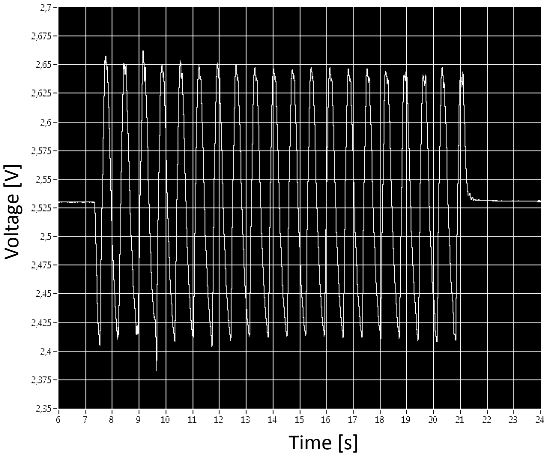

Figure 8. Electrical signal produced by mechanical excitation through the universal testing machine

In this case, the amplitude of the signals is of about 150 mV, an order of amplitude higher than those found with the vibration generator test. The deformation induced during bending produced by the vibration generator is small when compared to the one produced with the tensile stress test.

Conclusion

In this work a proof-of-concept has been developed for a coextruded filament sensor, which produces an electrical signal when stimulated by mechanical stimuli, behaving in the same way as standard piezoelectric film sensors.

Acknowledgments

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References

1. J. Lovinger, Developments in crystalline polymers, Elsevier applied science, London (1982).
2. S. Bauer, Journal of Applied Physics 80, p. 5531 (1996).
3. H. S. Nalwa, Ferroelectric Polymers: Chemistry, Physics and Applications Marcel Dekker, Inc, New York (1995)
4. Salimi and A. A. Yousefi, Polymer Testing 22, p. 699 (2003).
5. S. Lanceros-Mendez, J. F. Mano, A. M. Costa and V. H. Schmidt, Journal of Macromolecular Science, Part B: Physics 40 (2001), p. 517
6. K. Nakamura, D. Sawai, Y. Watanabe, D. Taguchi, Y. Takahashi, T. Furukawa and T. Kanamoto, Journal of Polymer Science Part B: Polymer Physics 41 (2003), p. 1701.
7. S. Lanceros-Mendez, M. V. Moreira, J. F. Mano, V. H. Schmidt and G. Bohannan, Ferroelectrics 273 (2002), p. 15
8. K. Matsushige, K. Nagata, S. Imada and T. Takemura, Polymer 21 (1980), p. 1391.
9. R. Gregorio and E. M. Ueno, Journal of Materials Science 34 (1999), p. 4489.
10. R. Gregorio Jr. and M. Cestari, Journal of Polymer Science Part B: Polymer Physics 32 (1994), p. 859.
11. J. Gomes, J. Serrado Nunes, V. Sencadas and S. Lanceros-Mendez, Smart Materials and Structures, 19 (6) (2010) 065010.
12. V. Sencadas, R. G. Jr. and S. Lanceros-Mendez, Journal of Macromolecular Science, Part B: Physics 48 (2009), p. 514
13. Piezo Film Sensors Technical Manual, Application Note, Measurement Specialties, Inc., available at http://www.meas-spec.com/product/t_resources.aspx?id=540# (2008), p.43
14. S. Walter, W. Steinmann, J. Schütte, G. Seide, T. Gries, G. Roth, P. Wierach and M. Sinapius, “Characterisation of piezoelectric PVDF monofilaments,” Materials Technology: Advanced Performance Materials, vol. 26, no. 3, (2011) pp. 140–145,
15. W. Steinmann, S. Walter, G. Seide, T. Gries, G. Roth, and M. Schubnell, “Structure, properties, and phase transitions of melt-spun poly(vinylidene fluoride) fibres,” Journal of Applied Polymer Science, vol. 120, no. 1, pp. 21-35 (2011)
16. A. Lund, B. Hagström, Melt Spinning of b-Phase Poly(vinylidene fluoride) Yarns With and Without a Conductive Core, Journal ofAppliedPolymer Science,Vol. 120, 1080–1089 (2011)
17. B. Mazurek, S. Rozecki, T. Kowalzcyk: The 6th International Conference On Properlies and Applications of Dielelectric Materials, China, Xi'an Jiaotong University, pp 1041- 1044 (2000)
18. B. Mazurek, S. Różecki, D. Kowalczyk, T. Janiczek, Influence of piezoelectric cable processing steps on PVDF beta phase content, Journal of Electrostatics, Volumes 51-52, Elsevier B.V. pp 180-185 (2001)