

Potential Application of PVDF/CoFe₂O₄ Nanocomposites as Self-powered Piezoelectric Plantar Pressure Sensors

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Abstract— This paper discusses the use of flexible nanogenerators as plantar pressure sensors, specifically focusing on the development of a sensor based on PVDF-TrFE/CoFe₂O₄ nanocomposites with a multilayer-graphene/gold top electrode (MGGE). The study highlights the importance of biomechanical sensors in healthcare, particularly in detecting changes in gait patterns that may arise from non-musculoskeletal irregularities in the body. The paper provides an overview of the different types of foot sensor technologies used for pressure monitoring, with a focus on piezoelectric force sensors. The use of PVDF-TrFE, a type of piezoelectric polymer, is discussed, along with techniques for enhancing its piezoelectric response. The results of the study demonstrate improved piezoelectric capabilities of the proposed nanogenerator, with a final piezoelectric coefficient d_{33} of 34 pm/V, which is in excellent agreement with the average value obtained by Piezoresponse Force Microscopy (PFM) $d_{33} = (33.99 \pm 5.12)$ pm/V. The proposed sensor has the potential to be used in the diagnosis and treatment of motor-related issues in diabetes, podiatry, and rehabilitation.

Keywords—PVDF-TrFE nanocomposites; CoFe₂O₄; magnetic poling; piezoelectric effect; piezoresponse force microscopy (PFM), Plantar Pressure Sensors.

I. INTRODUCTION (HEADING 1)

In the last years a lot of interest was attracted to the biomechanical sensors. Biomechanics refers to the study of how the human body, particularly the foot, moves in the presence of gravity. The forward motion of the body resulting from the movement of the lower limbs is known as gait. Various kinetic and kinematic factors of individual body segments significantly impact human gait. The healthcare industry places considerable importance on detecting changes in gait patterns that may arise from non-musculoskeletal irregularities in the body. Investigating the distribution of plantar pressure is a crucial area of research to address motor-related issues in diabetes, podiatry, and rehabilitation. Compared to analyzing gait patterns, foot sensor technologies are simpler to use and implement. Moreover, foot sensors have been demonstrated to be effective in both diagnosis and treatment [1].

The development of smart insoles, which were originally created for plantar pressure monitoring, can be traced back to the late 20th century when the Massachusetts Institute of Technology (MIT) Media Laboratory designed piezoelectric-based sneakers that generated energy while walking [2-3]. Numerous studies have since attempted to develop smart insoles for foot pressure monitoring, utilizing techniques such as Micro Electromechanical Systems (MEMS) [4], Force

Sensitive Resistor (FSR) [5], Piezoelectric Force Sensors (PFS) [6], Capacitive Sensor (CS) [7], Air Pressure Sensor (APS) [8], Opto-pressure Sensors (OPS) [9], and Polymer Optical Fibers (POF). Particularly appealing are piezoelectric force sensors, which can function independently of an external power source. Pedotti et al. [10], Razian and Pepper [11], and Kärki et al. [12] have all employed piezoelectric materials in plantar pressure monitoring.

During the last few decades, the possibility to convert the vibrational mechanical energy into electric energy, thanks to the development of innovative energy harvesting devices based on the piezoelectric effect, has recently attracted the interest of the scientific community. Integration of the energy harvesting function within the sensor element is a promising alternative. This allows for a more compact and user-friendly system structure, as the device functions as both the sensor and the energy harvester.

As an example of piezoelectric, piezoceramics with high piezoelectric coefficients (d_{33}) have been preferred due to their high conversion efficiency. However, these materials are toxic, brittle, and harmful to the environment. In order to address these shortcomings, several research groups have turned to investigating the piezoelectric properties of polymers, nanostructures, and nanocomposites, all of which have demonstrated interesting piezoelectric properties. These materials are suitable for the fabrication of flexible nanogenerators that can be used as a plantar pressure sensor [13-19].

Poly(vinylidene fluoride-co-trifluoroethylene) (PVDF-TrFE), a type of piezoelectric polymer, has received considerable attention due to its exceptional properties including excellent piezoelectric and ferroelectric properties, high chemical resistance, high thermal stability, large polarization, short switching time, and mechanical flexibility. Such properties make it suitable for various advanced applications, ranging from sensing to energy harvesting. In order to achieve a high piezoelectric coefficient (d_{33}), it is essential to align the PVDF-TrFE dipoles along a preferential direction, which is typically achieved through electrical poling. However, this technique is not cost-effective and not practical for industrial use since it requires applying a high voltage to each device. As a result, alternative techniques such as mechanical stretching, spin-coating, quenching, and the addition of external additives such as metal nanocomposites, ceramic fillers, and graphene nanoplatelets have been studied to enhance the piezoelectric response of the polymer. Recent research has also shown that introducing ferromagnetic nanoparticles into PVDF-TrFE/CoFe₂O₄ nanocomposites can significantly improve d_{33} by applying a DC magnetic poling [20].

This paper proposes the use of flexible nanogenerators as a plantar pressure sensor, based on PVDF-TrFE/CoFe₂O₄ nanocomposites with a multilayer-graphene/gold top electrode (MGGE). The sensor was electromechanically evaluated by applying perpendicular force with a commercial mini-shaker and monitoring the voltage produced by the piezoelectric effect using a digital oscilloscope. The nanogenerator demonstrates improved piezoelectric capabilities, with a final piezoelectric coefficient d_{33} of 34 pm/V, which is in excellent agreement with the average value obtained by Piezoresponse Force Microscopy (PFM) $d_{33} = (33.99 \pm 5.12)$ pm/V.

II. RESULTS AND DISCUSSIONS

A. Fabrication of PVDF/CoFe₂O₄ Nanogenerator

Spin coating process was used to fabricate the PVDF-TrFE/CoFe₂O₄ nanocomposite thin films [20]. Using a tip sonicate, a solution containing 1% CoFe₂O₄ nanoparticles and DMF was sonicated. The resultant DMF/CoFe₂O₄ solution was added in the desired proportion to Piezotech FC Ink L, a PVDF-TrFE ink. The solution was then sonicated for 30 minutes in a bath before being spin-coated onto PET/ITO. The resultant film was cured in an oven at 120°C for two hours. In order to achieve magnetic poling, a DC magnetic field was delivered to the nanostructured thin film using a ferromagnetic core while maintaining the film temperature at 65 °C. As soon as the active PVDF-TrFE/CoFe₂O₄ layer was formed, a multilayer-graphene paper that had been sputter-coated with 20 nm of Cr and 60 nm of Au was employed as the device's top electrode [21]. The graphene multilayer paper was coated with Cr/Au to increase electrode conductivity [16]. Figure 1 depicts a schematic illustration and a photograph of the produced nanogenerator.

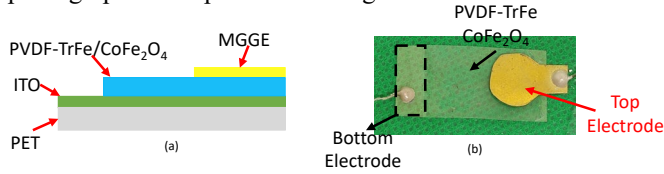


Figure 1 Schematic representation (a) and a picture (b) of PVDF-TrFE/CoFe₂O₄ nanogenerator.

B. Morphological Characterization

The morphology of the synthesized PVDF-TrFE/CoFe₂O₄ nanocomposite was examined using a Field Emission Scanning Electron Microscope (FE-SEM). The samples were sputter coated with a uniform Cr coating of 20 nm thickness to avoid charging during SEM imaging. Figure 2 depicts the nanocomposites' homogenous morphology, which is comprised of spherulitic structures with an average diameter of 1 μm.

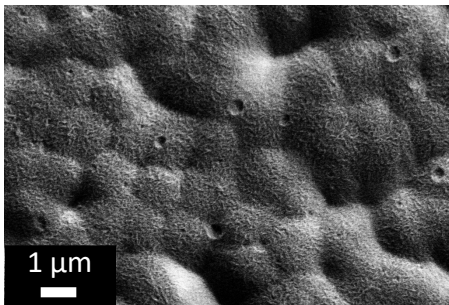


Figure 2 FE-SEM image of PVDF-TrFE loaded with 5 wt.% of CoFe₂O₄.

C. FT-IR Characterization

Five samples were analyzed using FT-IR to see if the presence of CoFe₂O₄ nanoparticles and subsequent magnetic poling affected the β -phase fraction. Nanocomposite samples poled in a DC magnetic field of 50 mT, 110 mT, and for 60 min. and 120 min. are shown in Figure 3 along with their corresponding Fourier transform infrared (FT-IR) spectra.

The relative fraction of the β phase, $F(\beta)$ can be evaluated, as reported in [13]–[15], [17], [18], [22], using the well-known equation:

$$F(\beta) = \frac{A_{\beta}}{(K_{\beta}/K_{\alpha})A_{\alpha} + A_{\beta}} \quad (1)$$

where A_{α} and A_{β} are the absorbance at the wavelengths (763 and 840 cm^{-1}) associated to the main peaks of the α - and β -phases, respectively. The ratio between the absorption coefficients of the β - and α -phases is $K_{\beta} / K_{\alpha} \sim 1.3$.

As it can be observed the spectra of neat PVDF-TrFE presented two clear peaks at 763 cm^{-1} and 840 cm^{-1} that can be used to estimate the $F(\beta)$. From the data reported in Figure 3 we obtained a $F(\beta)$ value equal to 81.06 %, a value higher than the value obtained for pure PVDF [15] and in agreement with values reported for PVDF-TrFE in [23]. However, as reported in [20], when CoFe₂O₄ nanoparticles are added to the PVDF-TrFE, the FT-IR spectra show a broadband shoulder close to the α -phase peak located at 763 cm^{-1} , making difficult the evaluation of the relative fraction of β -phase by using eq. 1. We believe that the presence of this shoulder is due to the interaction between the polymer and the nanoparticles. This hypothesis was supported by the presence of a broad peak at 1740 cm^{-1} in the spectra of all PVDF-TrFE/CoFe₂O₄ nanocomposite samples, which, as reported in [24], can be attributed to carbonyl groups (C=O). We speculate that this absorption band originates from the formation of bonds between O atoms of the CoFe₂O₄ nanoparticles and C atoms of PVDF-TrFE.

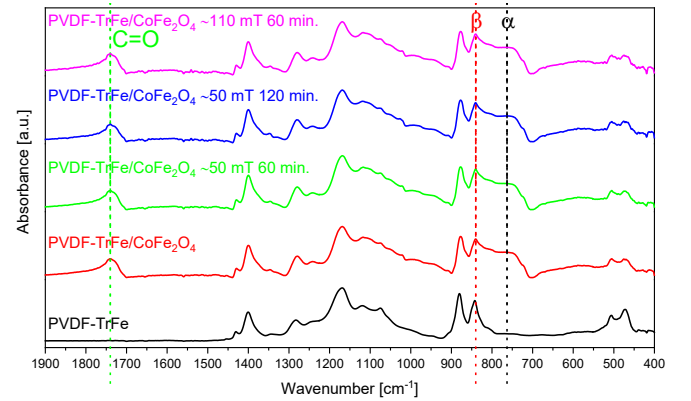


Figure 3 FT-IR spectra of the produced samples: i) neat PVDF-TrFE; ii) PVDF-TrFE filled with CoFe₂O₄ at 5 wt.%; iii) the PVDF-TrFE filled with CoFe₂O₄ at 5 wt.% and poled for 60 min. with a magnetic field of 50 mT; iv) the PVDF-TrFE filled with CoFe₂O₄ at 5 wt.% and poled for 120 min. with a magnetic field of 50 mT; v) the PVDF-TrFE filled with the CoFe₂O₄ at 5 wt.% and poled for 60 min with a magnetic field of 110 mT.

Nevertheless, as reported in [20], deconvolution of the shoulder from the absorption band related to the α -phase centered at 763 cm^{-1} , led an estimation of $F(\beta) = (82 \pm 3)$ % for the different samples. This estimation and the observation that the intensity of the β -peaks remains unaffected by the

introduction of CoFe_2O_4 nanoparticles as well as by the successive magnetic poling processes, provided evidences that the relative fraction of the β -phase remains nearly constant upon the introduction of the nanoparticles and is unaffected by the magnetic poling. We, then, believe that the increase of the d_{33} is due to the strong interaction between the CoFe_2O_4 nanoparticles and the magnetic field and their dragging effect over the polymeric chains of the PVDF-TrFE, resulting in their alignment along the direction of the applied DC magnetic field. As a result, we can infer that the primary effect of the DC magnetic poling is to increase the orientation of the β -phase domains along the magnetic field direction, since the β -phase content remains nearly constant.

D. PFM Characterization

The piezoelectric properties of the PVDF-TrFE/ CoFe_2O_4 nanocomposites were quantitatively evaluated by PFM measurements. An average piezoelectric coefficient (d_{33}) is determined by measuring three different regions of each sample and using the procedure developed in our previous works [14], [15], [25]. In Figure 4 were reported the d_{33} values for different applied DC magnetic field intensities, ranging from 0 mT to 220 mT. As can be seen, the piezoelectric coefficient increases when the magnetic field is increased and reaches a maximum value of (18.31 ± 1.06) pm/V for 1 h of the application of the magnetic field at 50 mT. Also shown in Figure 4 is the d_{33} value of a sample magnetically poled with a field of 50 mT at room temperature (RT). The value, although higher than that of the un-poled sample, is substantially lower than that of the sample poled with the same magnetic field at 65°C .

We have also investigated the effect of the application time of the DC magnetic field on d_{33} . Figure 4 (b) shows the measured d_{33} values of samples exposed to the magnetic field for increasing periods of time. The d_{33} values tend to saturate after 90 min of magnetic field poling, reaching, in the case of the samples poled with a magnetic field of 50 mT, an average value as high as 34 pm/V. We note that such a value is comparable to or even higher than those reported for electrically poled PVDF-TrFE samples [26]. This renders the process of magnetic poling of PVDF-TrFE/ CoFe_2O_4 nanocomposites particularly attractive for applications where electrical poling may have limitations.

We interpret the d_{33} increase induced by DC magnetic poling as due to the increased alignment of the β phase domains along the magnetic field direction, rather than to an increase of the β phase content, as proposed in [26]. In particular, as suggested by the FT-IR analysis, we speculate that the formation of a carbonyl group ($\text{C}=\text{O}$) is related to the chemical bonding that can take place between PVDF-TrFE and CoFe_2O_4 nanoparticles. The application of the DC magnetic field induce the ferromagnetic nanoparticles to orient themselves along the direction of the applied field, dragging the polymeric chains of PVDF-TrFE in the same direction. As also reported in [26], we observed that the d_{33} reaches a maximum value when a relatively high magnetic field strengths is applied ($B \sim 50$ mT). Application of higher magnetic fields up to 220 mT produced a decrease in the d_{33} values after magnetic poling. As already pointed out, when the DC magnetic poling is performed at RT the value of d_{33} does not increase as much as in the case in which the

temperature is kept at 65°C (see Figure 4). This indicates that also the temperature plays a relevant role in the orientation of the β phase. Indeed, by increasing the temperature the polymeric nanocomposite becomes softer, facilitating the orientation of β phase along the applied magnetic field direction [23].

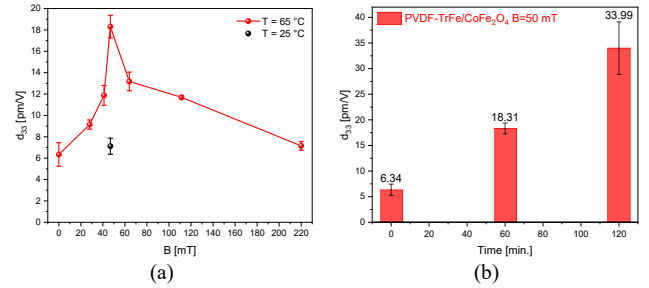


Figure 4 (a) d_{33} value of polymeric nanocomposites as a function of the amplitude of the applied DC magnetic field. The black point represents the value of d_{33} when the DC magnetic poling was performed at RT; (b) value of d_{33} of the nanocomposites with 5 wt % of CoFe_2O_4 as a function of the application time of the DC magnetic field.

E. Electromechanical Characterization

The optimized PVDF-TrFE/ CoFe_2O_4 nanocomposites were used as active layers to fabricate flexible nanogenerators suitable to convert the vibrational mechanical energy associated with the wearer person into an electrical signal, realizing a biomechanical sensor for healthcare, able to detect changes in gait patterns that may arise from non-musculoskeletal irregularities in the body. Within this aim, a multilayer-graphene gold paper is used as top electrode, as sketched in Fig.1. In Fig. 5 the response of the flexible nanogenerators, based on PVDF-TrFE/ CoFe_2O_4 nanocomposites poled with 50 mT for 1h and for 2h, are reported when a sinusoidal force of 0.25 N at the frequency of 110 Hz was applied through a commercial mini-shaker. We have chosen this experimental condition we believe to represent an extreme and challenging working condition, even though the expected frequency is much lower and the force due to wearer load is generally one order of magnitude higher. Moreover, the selected force and frequency allow us to compare the obtained piezoelectric response to the one found in [27], [28], where a method similar to the normal load was used to estimate the piezoelectric coefficient d_{33} . We calculated the current collected from a RC circuit using an external resistance of 1 M Ω :

$$d_{33} = \frac{V_{out}}{2\pi f R |F|} \quad (2)$$

where V_{out} is the amplitude of the voltage acquired by a digital oscilloscope, R is the value of the load resistor and F is the magnitude of the applied force. A value of 19 pm/V and of 34 pm/V were evaluated, for the samples poled with 50 mT for 1h and for 2h, respectively. These values are in good agreement with the values (18.31 ± 1.06) pm/V and (33.99 ± 5.12) pm/V measured through PFM on the PVDF-TrFE/ CoFe_2O_4 films before depositing the top electrode. From the data reported in Fig. 5 we have then evaluated the power density:

$$PD = \frac{P}{A * t} \quad (3)$$

where A is the area solicited by our shaker (0.53 cm^2), t is the thickness of our films (around 10 μm) and $P = V^2/R$ is the power across the external resistance. As can be observed in

Figure 6, we obtained an average value of $0.04 \mu\text{W}/\text{cm}^3$ and of $0.136 \mu\text{W}/\text{cm}^3$ for the samples poled with 50 mT for 1h and for 2h, respectively.

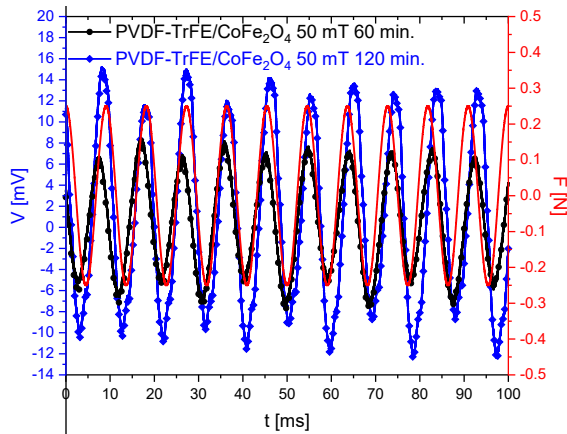


Figure 5 Electromechanical response of the PVDF-TrFE/CoFe₂O₄, poled with 50 mT for 1 h and 2 h (black and blue curve, respectively) when a load of 0.25 N at 110Hz was applied (red curve).

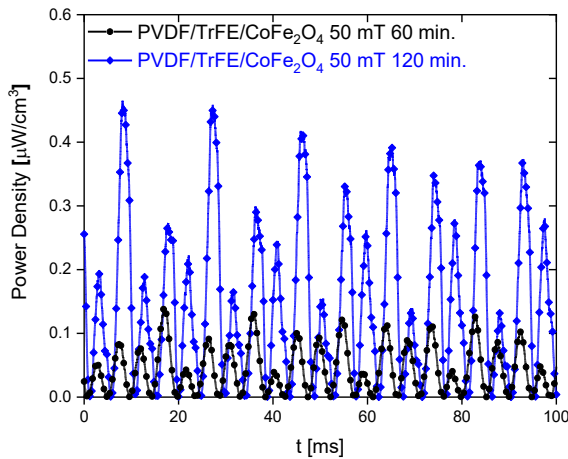


Figure 6 Calculated output power density across the external resistance for the sample poled with 50 mT for 1h (black line) and for 2h (blue line).

III. CONCLUSION

The goal of this research was to produce a sensor that would fit subject of plantar pressure sensor capable of recognizing human motion states for walking, running, and jumping. As a result of our efforts, high d_{33} piezoelectric polymeric nanocomposites have been successfully demonstrated as the basis for flexible nanogenerators that could be employed as a plantar pressure sensor. Nanogenerators were created by doping PVDF-TrFE with ferromagnetic CoFe₂O₄ nanoparticles and then applying DC magnetic poling. The highest d_{33} value (34 pm/V) was obtained introducing 5 wt% of CoFe₂O₄ nanoparticles and applying a DC magnetic field of 50 mT for 2 h. Furthermore, we adopted an innovative solution to fabricate a flexible top electrode based on multilayer-graphene/gold paper.

The developed combination of polymeric nanocomposite, including ferromagnetic CoFe₂O₄ nanoparticles, and the application of a DC magnetic poling provides an alternative route to obtain highly efficient piezoelectric materials with excellent d_{33} values. In particular, the magnetic poling appears to be easily implemented industrially, if compared to electrical poling, requiring the formation of top and bottom electrodes

and the individual poling of each device. The electromechanical measurements have demonstrated how the produced device can be particularly attractive in the fabrication of energy harvesting devices for wearable sensors in flexible electronics applications. In particular, we observed that from the fabricated prototype we are able to extract a maximum power density of $0.438 \mu\text{W}/\text{cm}^3$. PVDF-TrFE nanogenerators has been found to be a suitable material for the using plantar pressure measurements as it is deformable and light and does not therefore act as a foreign object.

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