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Electromechanical characterization of piezoelectric PVDF polymer films for tactile sensors in robotics applications

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ABSTRACT

In this paper an experimental setup and experimental procedures which allow a fast and complete characterization of piezoelectric films continuously over the 1 Hz–1 kHz frequency range are reported. Some results related to PVDF electro-mechanical characterization are also presented. This work is intended as the first step for the electro-mechanical design of innovative integrated transductions systems for tactile sensors in robotic applications. The article concludes with a summary and a discussion of future investigations.

1. Introduction

Tactile sensing can be defined as the operation of perceiving and recognizing the properties of a contact event occurring at the surface of a deformable medium [1,2]. Some of the sensing organs in human skin work by converting mechanical and thermal inputs into electric signals. To mimic the complex behaviour of human skin in a humanoid robot a multimodal system would be required, which employs different kinds of transducers. Films of some polymers, such as polyvinylidene fluoride (PVDF) and its copolymers, opportunely conditioned, exhibit piezoelectric and pyroelectric properties [3–5] with a fast dynamic response. Moreover, they can be reduced to tiny compliant strips for embedding in a continuous elastic layer. Used as isolated sensors/actuators or assembled into more complex multimodal devices, they offer challenging solutions in many application fields including robotics. For these reasons they can be regarded as interesting candidates for tactile sensors [6].

PVDF is synthesized by addition polymerization of the $CH_2{=}CF_2$ monomer. When produced as the homopolymer (i.e. from 100% $CH_2{=}CF_2$ monomer), the majority of the PVDF chains have a regular structure of alternating CH_2 and CF_2 groups.

The crystalline phase of interest for PVDF ferroelectricity is the polar β phase. This structure can be obtained either by mechanical stretching of PVDF films [7] or by using the PVDF trifluoroethylene copolymer [P(VDF-TrFE)] which has the tendency to crystallize directly in the polar β-phase. The piezoelectric effect originates from induced polarization. The dipoles in a semi-crystalline polymer such as PVDF must be reoriented through the application of a strong electric field at elevated temperature [8–12]. The temperature is then lowered in the presence of the electric field, so that the domains are locked in the polarized state. The material piezoelectric effect is directly related to the degree of polarization achieved.

As discussed above, such piezoelectric films can be integrated in multimodal sensors, mimicking the behaviour of the human skin. In order to model these complex mechanical and electrical structures using Finite Element (FE) and multiphysics simulators as design tools, the basic PVDF "building block" must be known in all its properties. Thus, the first step towards the integration of different transducers in the same system consists in the electro-mechanical characterization of the single transducer.

With regard to the practical use of the robotic skin, it is important to emphasize the fact that it must work in dynamic environments. In particular, the frequencies of interest for this application range from 0 up to about 1 kHz [1]. PVDF thin films well cover the whole range (though not responding to static stimuli). Therefore, the knowledge of their dynamic

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viscoelastic–piezoelectric–dielectric response in this range and the temperature dependence must be investigated. The long time response of these polymers, which has been already investigated by Vinogradov et al. in creep tests [17], is of course another important feature to implement in the simulation models.

Linear electro-elastic constitutive equations are commonly used to describe the coupling of dielectric, elastic, and piezoelectric properties in piezoelectric materials [13].

In the frequency domain ($\hat{f}(\omega)$ means the Fourier Transform of any function $f(t)$) such equations are:

$$\begin{bmatrix} \hat{S} \\ \hat{D} \end{bmatrix} = \begin{bmatrix} \hat{s} & \hat{d}^T \\ \hat{d} & \hat{\varepsilon} \end{bmatrix} \begin{bmatrix} \hat{T} \\ \hat{\exists} \end{bmatrix} \tag{1}$$

where strain $\hat{S}$ and stress $\hat{T}$ are represented by 1×6 column vectors, while the electric displacement $\hat{D}$ and the electric field $\hat{\exists}$ are expressed by 1×3 column vectors. $\hat{s}$ is the 6×6 *compliance* matrix, $\hat{\varepsilon}$ the 3×3 *permittivity* matrix, both assumed to be symmetric, and $\hat{d}$ the *piezoelectric* 3×6 matrix.

Extensive research about static and dynamic viscoelastic and piezoelectric properties of PVDF is authored by research groups at Montana State University [14–18]. Electromechanical energy losses in different frequency ranges, creep and electromechanical relaxation of these viscoelastic materials have been studied by these and by other authors [16,17,19]. The whole set of elastic, dielectric and piezoelectric complex coefficients is retrieved [19] over the 200–600 kHz frequency range. Some P(VDF-TrFE) elastic, dielectric and piezoelectric constants as a function of frequency have also been evaluated in the MHz range from impedance data using five different methods [20]. In both cases, the frequency range is well above the relevant frequencies for the present application.

Completing the available information motivates the effort in developing methods and tools to characterize the electromechanical behaviour of PVDF films in the above cited frequency range.

The aim of this work is accordingly twofold: first, to illustrate the experimental setup and the experimental procedures which allow a fast and complete characterization of piezoelectric films continuously over the 1 Hz–1 kHz frequency range, secondly to contribute developing a set of tools and data repository for use in the electromechanical design of integrated sensing systems. An exhaustive characterization of PVDF films is however beyond the scopes of this preliminary work.

2. Materials and methods

2.1. PVDF films

Commercial metallized PVDF sheets (190 mm × 280 mm in size) from Measurement Specialties Inc. [http://www.meas-spec.com/default.aspx] have been purchased. PVDF samples have been cut from those sheets in rectangular geometries of various sizes, according to the type of test.

Purchased sheets have been already stretched and poled. Rolls of piezo film by Measurement Specialties Inc. are produced in a clean room environment. The process begins with the melt extrusion of the polymer resin pellets into sheet form, followed by a stretching step that reduces the sheet to about one-fifth of its extruded thickness. Stretching at temperatures well below the melting point of the polymer causes chain packing of the molecules into parallel crystal planes ("beta phase"). Even if the temperature of the thermal treatment cannot be given because the details of film processing are considered proprietary and confidential, the company ensures that its default process yields films which are thermally stable to 60–70 °C. To obtain high levels of piezoelectric activity, the beta phase polymer is poled by application of very high electric fields (of the order of 100 V/μm) to align the crystallites to the poling field. In such conditions, the piezoelectric behaviour exhibits a material symmetry in the orthorhombic crystal system (C_{2V} class), corresponding to that of the so-called *orthotropic* materials. That means that with reference to the symmetry axes the dielectric matrix $\hat{\varepsilon}$ is diagonal (3 elements), the compliance matrix $\hat{s}$ is formed by 9 independent elements and the piezoelectric matrix $\hat{d}$ has only 5 elements different from zero. The samples are oriented with axis 1 along the stretching direction, axis 2 in the in-plane orthogonal direction and axis 3 along the through-thickness direction.

Among the different thickness choices, 110 μm thick films have been chosen for higher robustness. Thin sputtered metallization has been preferred to silver ink, for a higher signal to noise ratio. The standard sputtered metallization is composed of 700 Å of copper covered with 100 Å of nickel, which has good conductivity and is resistant to oxidation.

2.2. Experimental method

2.2.1. Test equipment

The experimental setup (Fig. 1(a)) for measurements of viscoelastic moduli consists of a rigid frame with a lower fixed plate to which an electro-mechanical shaker (Brüel&Kjaer, Minishaker Type 4810 with Power Amplifier Type 2706) is assembled. An accelerometer (PCB 355B03) is fixed to the moving head of the shaker. The upper head of the frame can be positioned by means of a regulation screw at various distances as required by the type of test and the sample size. Regulation of this screw also provides the amount of preload on the sample (in a range from 0 to 6 N). A piezoelectric force transducer (PCB 208M116) is fixed to this head of the frame. It is also used to read the preload.

Pairs of grips of different shapes can be mounted between the force transducer and the accelerometer to hold the samples. The loading chain is formed, accordingly, by: shaker, accelerometer, sample, force transducer. All these elements have to be accurately aligned before any test.

The test is controlled by a computer in a completely automatic way. A swept sine signal is fed into the shaker. Each train – at a given frequency – is made of a fixed number of 20 waves. The settable parameters are start and end frequencies, number of steps, amplitude. Frequency spacing and total duration of the test are thus determined by the frequency range and the number of steps. The output signals of accelerometer (response) and force transducer (stimulus) are continuously acquired (signal conditioned by PCB Sensor Signal Conditioner 482C54) and processed in frequency to give the response function for each frequency step. The complex moduli are calculated from the response function and the geometrical parameters of the sample. Even though the apparatus can run from 1 Hz to 10 kHz, there are practical limits on the effective frequency range, depending partly on the used sensors and partly on the sample properties. At very low frequencies, depending on the stiffness of the sample, the sensitivity of the accelerometer drops and noise is introduced. On the other hand, it is important to care that the fundamental frequency of the sample is sufficiently high with respect to the measurement range to avoid interference. Other sources of interference may be produced by undesired vibration modes of the sample (e.g. flexural modes in a tensile test). Normally the range between 10 and 1000 Hz could be explored without difficulties. In the post-processing section of the program all frequency spectra were filtered by a numerical Savitzky-Golay filter. The program permits to display a stress-strain plot of each test, which may be useful to verify the linearity of the material behaviour. Both the vibration amplitude and the value of the applied force are determined by the response of the mechanical chain and change