PIEZOELECTRIC ACTUATION OF A DIRECT WRITE ELECTROSPUN PVDF FIBER

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ABSTRACT
Piezoelectric actuation of a doubly clamped suspended poly (vinylidene fluoride) (PVDF) fiber has been demonstrated. The PVDF fiber is directly deposited onto a collector using near-field electrospinning with *in situ* electrical poling process. The piezoelectric effect can be further enlarged by polarizing the fiber under a voltage of 2kV for two hours at a temperature of 80°C. A displacement of 2μm on the center of a suspended PVDF fiber, corresponding to a strain of 0.004%, as a result of piezoelectric effect has been detected under an applied electric field of 1.6V/μm. We believe this technology could enable new potential actuation applications for NEMS/MEMS.

INTRODUCTION
PVDF is a highly non-reactive, flexible, inexpensive, and leading polymer with good piezoelectric property [1]. It is very attractive in performing the energy conversion function between electric and mechanical domains and hence can be utilized as electromechanical actuators, artificial muscles, motion sensors and energy harvester. The actuators based on PVDF film has been widely developed and studied [2, 3]. However, there are few reports on the actuation of a single PVDF fiber. The main advantage of the actuators based on PVDF fibers is its scalability. In this work, we demonstrated a piezoelectric actuator based on a single fiber, which could be potentially scaled down to nanometers or scale up to fiber networks for various potential applications.

PVDF is a semicrystalline polymer consisting of 4 crystalline phases α, β, γ and δ. Among these, non-polar α phase is the most common and usually found in commercially available powders and films. It has random orientation of dipole moments, and thus the dipole moments cancel each other out. β phase has all the dipole moments pointing to the same direction, such that it is responsible for the piezoelectric properties of the polymer as the piezoelectric behavior is based upon the dipole orientation within crystalline phase of PVDF. Figure 1 shows the basic concept of the near-field electrospinning (NFES) [4, 5, 6] process with PVDF fibers to cause *in situ* electrical poling. During this process, the high electric field aligns the dipole moments in the solution to have the same direction [7]. Therefore, electrospinning of PVDF from its solution can transform non-polar α phase in the crystalline into polar β phase, introducing the piezoelectric effect of electrospun PVDF fibers.

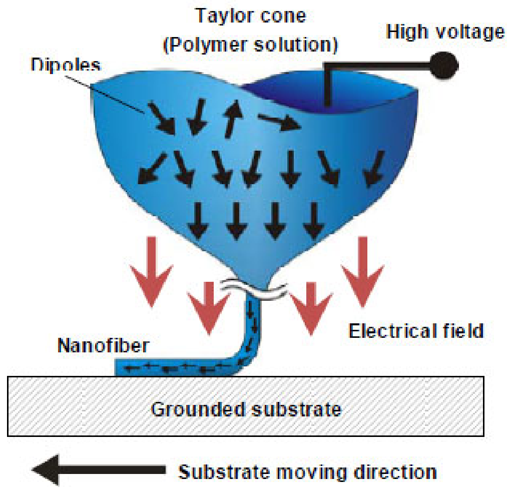

Figure 1. *Schematic diagram of the electrospinning process for in situ poling of PVDF fibers to gain its piezoelectric property. The polarity of the fiber is determined as shown.*

DESIGN AND FABRICATION
Control capability of electrospinning is a key issue to make functional devices and to tailor their performance. NEFS has capability to control the deposition position of PVDF fibers in contrast to random deposition of conventional electrospinning. In our demonstrations, the PVDF fiber with a diameter of 550 nm is deposited continuously and the complex patterns are assembled by controlling the movement of the collector using programmable motion stage (Newport, Inc), shown in Figure 2 (a)-(d). Controllability of the fiber deposition enables us to attain arrays of aligned PVDF devices.

In the preparation of PVDF solution, we use dimethyl sulfoxide (DMSO) as solvent for PVDF powder (Mw=534000), and add acetone and surfactant

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into the solvent to improve the evaporation rate and reduce the surface tension of the solution, respectively.

During NEFS, the diameters of fibers could be controlled by adjusting various electrospinning parameters, such as PVDF concentration, voltage, needle-to-collector distance and motion stage translational speed. Figure 2(e) shows the controllability of fiber diameters based on motion stage translational speed. Increasing the stage translational speed could produce thinner PVDF fibers. However, higher speed (more than 90mm/s) would terminate the electrospinning process, making fiber discontinuous.

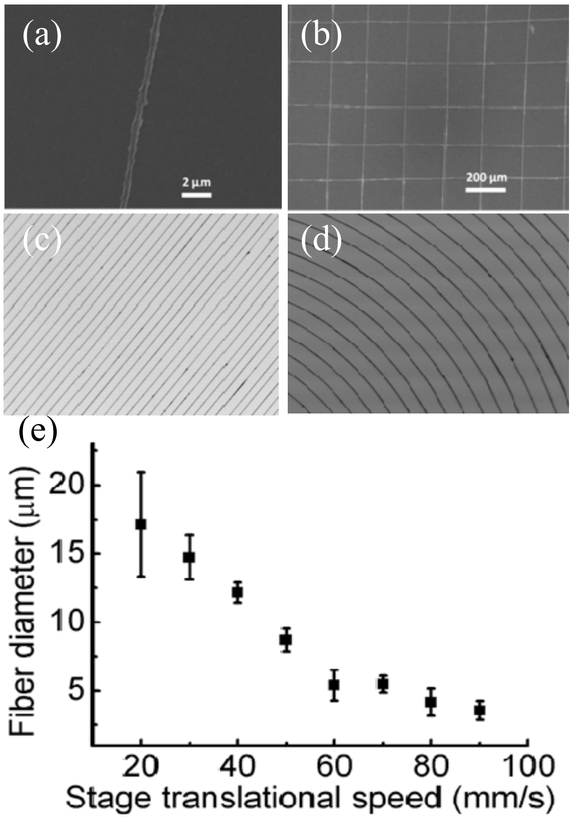

Figure 2. *Experiments showing controllability of NEFS for PVDF fibers* ***(a)*** *A single electrospun PVDF nanofiber with diameter of 550nm.* ***(b)*** *A grid pattern with controlled 200µm spacing* ***(c)*** *Parallel fibers with controlled 100µm spacing.* ***(d)*** *Arc pattern with controlled 100µm spacing.* ***(e)*** *Dependence of electrospun PVDF fiber diameters on the stage translational speed. Other parameters are as follows: PVDF concentration (20wt%), solvent (DMSO: acetone) weight ratio (5:5), surfactant concentration (5wt%), needle-to-collector distance (1.5mm) and electrospinning voltage (0.92 kV).*

Figure 3 (a) illustrates the actuation scheme based on doubly clamped piezoelectric PVDF fiber. The aluminum electrodes are put on the glass substrate with a spacing of 500µm to 1mm to serve as the collector for NFES process and the input contacts. PVDF fiber is written directly across two grounded aluminum electrodes using NFES and then the two ends of the suspended fiber are fixed by conductive epoxy. Figure 3(b) shows the SEM image of the suspended PVDF fiber with diameter of 2.6µm and length of 500µm.

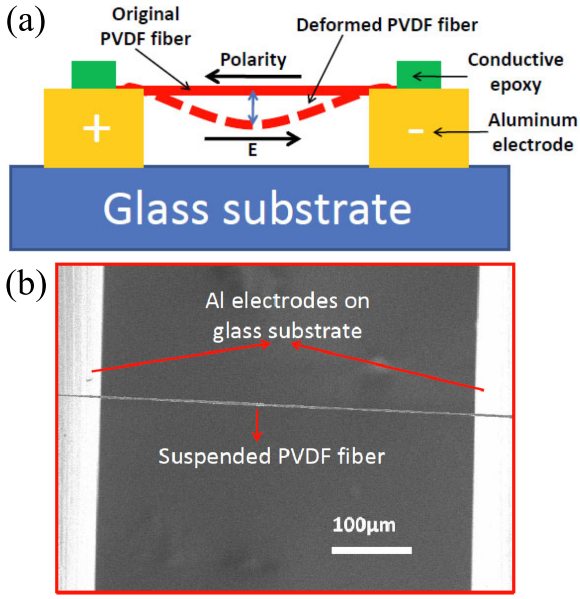

Figure 3. ***(a)*** *Cross section view of the piezoelectric PVDF fiber actuator. When applying an electrical field that is opposite to the polarity along the fiber length, the suspended fiber elongates and buckles down.* ***(b)*** *SEM image of a suspended electrospun PVDF fiber (2.6µm in diameter and 500µm in length).*

Theoretically, if there is no external stress applied at the piezoelectric PVDF fiber, the magnitude of the strain ε along the fiber axis is proportional to the applied electric field E and the piezoelectric coefficient d_{33}, as:

$$\varepsilon = d_{33}E \qquad (1)$$

PVDF has negative d_{33} so that positive electric field (in the same direction with the polarity) generates compression strain along the fiber axis, consequently causing no deformation of fiber because the two ends of the fiber are fixed. Negative electric field (in the direction opposite to the polarity) generates tensile strain, which could cause the fiber to buckle. The central point displacement of the fiber under negative electric field can be detected based on microscope focus/defocus technique and the generated strain ε along the fiber axis can be calculated as [8]

$$\varepsilon = \frac{\Delta l}{l_0} \approx \frac{1}{2l_0}\int_0^{l_0}\left(\frac{dD}{dx}\right)^2 dx = 2.44\left(\frac{D_c}{l_0}\right)^2 \qquad (2)$$

Where l_0 is the original length of the fiber, Δl is the change in fiber length and D_c is the displacement on the fiber center. Tiny Δl can be amplified by this configuration because it has quadratic dependence on D_c. As an example, with $\Delta l = 0.02\mu m$ and $l_0 = 0.5mm$, D_c is on the order of $2\mu m$, which can be detected using microscope focus/defocus technique.

EXPERIMENTAL RESULTS

Experimental results show out-of-plane displacement on the fiber center when the external electric field is applied along the fiber length and an optical microscope records and measures the out-of-plane displacements on the fiber center. Figure 4 shows the out-of-plane movement at the central part of an electrospun PVDF fiber, which is measured by refocusing the lens of the microscope on the same spot and recording the vertical migration distance of the lens that is equivalent to the displacement of the fiber.

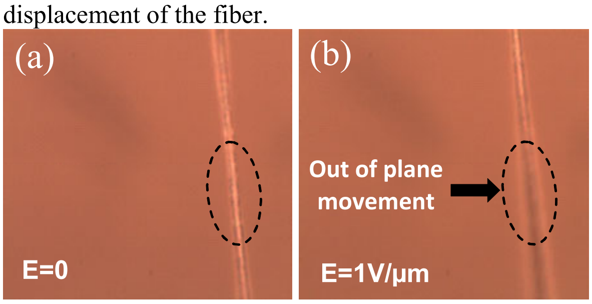

Figure 4. *Optical images of the central part of an electrospun PVDF fiber demonstrating the out-of-plane movement* ***(a)*** *before and* ***(b)*** *after applying an external electrical field of 1V/μm along the fiber length.*

Figure 5 plots the relationship between the central point displacement of the electrospun PVDF fibers and the applied electric field. Line (a) in Figure 5 is from the electrospun PVDF fiber with in-situ poling, indicating the displacements in responding to positive and negative electric fields (positive and negative directions are defined as relative to the direction of the original electrospun fiber polarity) are in the same direction with small variation in magnitude. This does not agree with the previous theoretical prediction on piezoelectric responses. The reason is explained with the fact that electrostatic attraction could occur between the space charges trapped in the fiber and the electrodes, as illustrated in Figure 6(a). Under high voltage, the charge carriers within the fiber are forced to migrate a small distance inside the fiber and then become trapped with a built-in potential [9, 10]. As a result, the fiber is deformed due to the electrostatic attraction force and the central point moves downwards as shown in Figure 6(a). Moreover, it is found that larger displacement is recorded under negative electric field in Figure 5(a) and this implies the existence of piezoelectric effect in the system. Clearly, the asymmetric responses come from the contributions of both piezoelectric and electrostatic effects depicted in Figure 6(b). When applying +E, total deformation is equal to combined downward electrostatic deformation and upward piezoelectric deformation. When applying –E, total deformation is equal to combined downward electrostatic deformation and downward piezoelectric deformation. The difference between the displacement magnitudes under positive and negative electric fields is twice the effects of the piezoelectric responses of the PVDF fiber under the same magnitude of applied field.

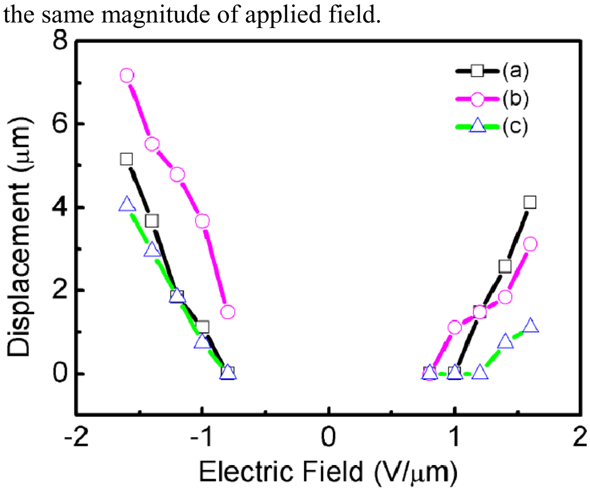

Figure 5. *Experimental results showing the relationship between the central point displacement and applied external electric field for a prototype electrospun PVDF fiber: (a) without repolarization (b) with repolarization (c) with repolarization and double-layer electrodes.*

The polarization of the electrospun PVDF fiber can be further strengthened by polarizing the fiber in the same direction as the original polarity. The prototype procedure is conducted under a voltage of 2kV at a relatively high temperature of 80°C for two hours. We can see obvious piezoelectric response enhancement for the repolarized PVDF fiber, as shown in Figure 5 (b).

The large electrostatic force is mainly because of the asymmetry of electrodes as shown in Fig. 6(a). Therefore, by adding one extra layer of top electrodes on the two original aluminum electrodes should reduce the electrostatic effects as explained in Figure 6(c). In the ideal case, the electrostatic force can be balanced and reduced to zero. However, in reality, it is difficult to eliminate the electrostatic force. Figure 5(c) shows the results for the device with double-layer electrodes, indicating the electrostatic force is further reduced (possibly close to zero when the electric field is less than 1.4V/μm as the displacement is close to zero under the positively applied electrical field). A displacement of 2μm resulting from converse piezoelectric effect is extracted under an electric field of 1.6V/μm.

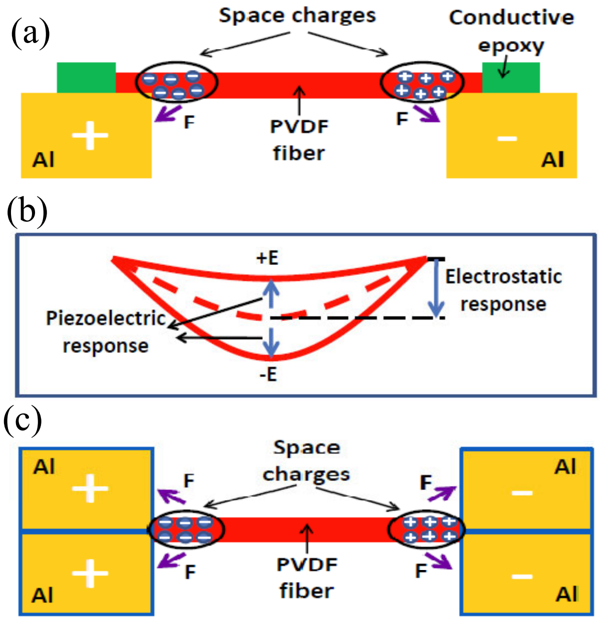

Figure 6. *(a) Side view for illustration of electrostatic effect, the electrostatic force between the trapped space charges in the fiber and electrodes deforms the fiber, thus its central point moves downwards. (b) Deformed PVDF fiber under +E or –E as a result of piezoelectric effect and electrostatic effect. When applying +E, the total deformation is equal to the combined downward electrostatic deformation and upward piezoelectric deformation; when applying –E, the total deformation is equal to the combined downward electrostatic deformation and downward piezoelectric deformation. (c) Side view showing balanced electrostatic force by adding aluminum electrodes on top of the original electrodes. The PVDF fiber in (a, c) were enlarged for illustration purpose.*

The piezoelectric strain is calculated according to equation (2) and a strain of 0.004% has been calculated from the repolarized electrospun PVDF fiber under the applied electric field of 1.6V/µm. Furthermore, from the linear data fit on the strain analyses, piezoelectric coefficient d_{33} is obtained as 39pm/V, which is a little larger than those reported for bulk PVDF [11, 12].

CONCLUSIONS

Piezoelectric response of a suspended PVDF fiber directly written by NEFS has been characterized in this work. The doubly clamped fiber configuration helps amplifying tiny fiber elongation to larger deformation at the center of the fiber for measurements. Results show that under an electric field of 1.6 V/µm, an out-of-plane displacement of 2µm is achieved which corresponds to a calculated piezoelectric coefficient d_{33} as 39pm/V. Furthermore, experiments show NEFS can deposit PVDF nanofibers with complex patterns. As such, the combination of good piezoelectric properties and controllable patterns of PVDF fibers could lead to structures such as arrayed actuators for applications in artificial muscles and switches.

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