Title:	In-situ Electric Field Facilitated Piezo- and Pyro-electric Nanogenerators.
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Abstract: Nanogenerators are one of the prominent means to harvest energy from mechanical vibrations, motions, and temperature fluctuations to convert it into electrical energy. Thus, the piezo- and pyroelectric devices are preferably endorsed and extensively explored to deliver clean, accessible, and sustainable energy alternatives for wearable electronics. However, to achieve these piezo- and pyroelectric properties in a flexible material, the electroactive phase nucleation within the crystalline region of semi-crystalline polyvinylidene fluoride (PVDF) is the primary concern. The post-poling process is the next essential requisite for the unidirectional molecular dipole orientation within the polymer. Noteworthy to mention that during this process, very high electric field strength of order ~ 10 2 MV/m is typically required, which causes dielectric fatigue and electrical breakdown. Hence, this high electric field treatment limits the traditional electroforming and poling processing and hinders the desired output of the device. This thesis presents an in-situ approach to nucleate the electroactive phases in polyvinylidene fluoride (PVDF) under a lower electric field (~ 0.1 MV/m), than the typically required field for the ex-situ process. The acquired electroactive PVDF has been utilized to fabricate piezo- and pyroelectric devices and demonstrated as self- powered sensors, including pressure mapping, breathing, and proximity sensor for practical applications. In this context, the δ-phase comprising PVDF nanoparticles have been fabricated during the in-situ process through an electrospray technique under electric field strength of ~ 0.1 MV/m, i.e., 10 3 times lower than the typical value required for δ-phase transformation. The kink propagation model, governed by the rotation of every secondary polymer chain unit along the main -c-c- chain axis, validates the δ-phase transformation in the electrospray process. The piezoelectric properties of δ-PVDF nanoparticles have been investigated through piezo response force microscopy, and the piezoelectric coefficient of ~ –11 pm/V was achieved. The δ-PVDF nanoparticle comprised piezoelectric nanogenerator (PNG) has shown a very prompt response as a self-powered pressure sensor, with higher pressure sensitivity of ~80 mV/kPa for realistic pressure mapping sensor applications. The electrode deposition over δ-PVDF nanoparticles was an utmost concern during the device preparation. To address this issue, we have adapted the in-situ vapor phase polymerization technique to deposit the electrodes over δ-PVDF nanoparticles and achieved an all-organic 1piezoelectric nanogenerator. This resulting all-organic stretchable, breathable, and flexible nanogenerator displayed an excellent electrical output with higher mechanical sensitivity due to the superior electrode compatibility between active materials and organic electrodes compared to metal electrodes. The obtained all-organic nanogenerator also exhibits excellent air permeability, enabling the advantage of wearable devices. Further, a machine learning algorithm was proposed to recognize different finger gestures efficiently to predict the source point with the highest possible accuracy of ~ 94%. Since the δ-PVDF nanoparticles are also expected to be pyroelectric in nature, a simultaneous temperature-dependent and pyroelectric study was also performed with an in-situ fabricated all-organic nanogenerator, and a prominent pyroelectric current and voltage response was recorded under different temperature stimuli as evidence of pyroelectricity in δ-PVDF nanoparticles. In PVDF, β-phase is another electroactive phase, which is successfully achieved in this study during the electrospinning process in the presence of external fillers for in-situ preparation of PVDF nanofibers. To improvise the pyroelectric response PVDF nanofibers based nanogenerators, we have introduced the MXene in PVDF nanofibers. The pyroelectric study for this composite system was performed under IR light. The PVDF-MXene nanofiber based pyroelectric nanogenerator has shown remarkable improvement in pyroelectric response, with an enhanced pyroelectric coefficient of 130 nK -1 m -2 for the composite as compared to the pristine counterpart due to the confinement effect of MXene in nanofibers and hydrogen bonding between the functional groups present in MXene and -KH 2 -, -KF 2 - groups, in PVDF. In subsequent studies, we have demonstrated that the fabricated pyroelectric nanogenerator could also be utilized as an excellent pyroelectric breathing sensor, proximity sensor, and IR data transmission receiver. Further, supervised machine learning algorithms are proposed to distinguish different types of breathing signals with ~ 98 % accuracy for real-life applications. Finally, to study the in-situ electrical poling feasibility in PVDF film-based system, we have performed a comparative study with the post-poling process. To validate this possibility, we have prepared solution casted PVDF films under corona discharge to induce the electroactive phase in PVDF under lower electric field (~1.5 MV/m), which is ~ 60 times lower than the field required in traditionally adopted post poling process. The effective poling at this electric field strength is also dependent on the poling duration that facilitates the presence of oxygen and additional carbon species as observed in XPS study. Konsequently, the UPS study confirms the effective changes in surface potential and the associated density of states (DOS). The optimum poling duration is observed between 30-50 min to identify the prominent changes in surface potential of PVDF film.

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