Phase Inversion in PVDF Films with Enhanced Piezoresponse Through Spin-Coating and Quenching

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In the last ten years, piezoelectric polymer thin films have attracted a lot of interest in the production of flexible nanogenerators, sensors, and actuators. Poly (vinylidene fluoride) (PVDF) is one of the most interesting piezoelectric polymers for a wide range of advanced applications. In order to increase the electroactive response of PVDF, the CH2–CF2 dipoles must be aligned and oriented along a preferential direction. Dipoles orientation is usually obtained by electrical poling. However, this technique has some limitations in terms of cost-effectiveness and practical implementation. In the present work, poly(vinylidene fluoride) (PVDF) films were produced by spin-coating, followed by quenching under different conditions, in order to investigate the dominant mechanism of the β-phase formation. The influence of the polymer/solvent mass ratio of the solution, the rotational speed of the spin-coater and the crystallization temperature of the film on both the β-phase content and the piezoelectric coefficient (d33) were investigated. This study demonstrates that the highest values of d33 are obtained when thinner films, produced with a lower concentration of polymer in the solvent (i.e., 20 wt.%), go through quenching in water, at room temperature. Whereas, in the case of higher polymer concentration (i.e., 30 wt.%), the best value of d33 (~30 pm/V) was obtained through quenching in liquid nitrogen. We believe that in the former case, phase inversion is mainly originated by electrostatic interaction of PVDF with the polar molecules of water, due to the low viscosity of the polymer solution. On the contrary, in the latter case, due to higher viscosity of the solution, mechanical stretching induced on the polymer during spin-coating deposition is the main factor inducing self-alignment of the β -phase. These findings open up a new way to realize highly efficient devices for energy harvesting and wearable sensors.