Dielectric Hysteresis Behaviors of Polyvinylidene Fluoride-based Multilayered Dielectrics Controlled by Confined Distribution of Conductive Particles

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I. Sample Preparation

The PVDF (FR-907) with a density of 1.78 g/cm$^3$ was provided by Shanghai 3F Co. Ltd. (China). The conductive carbon black (E900), with a density of 1.80 g/cm$^3$ and average particle size of 100-150nm, was offered by Chengdu ZhengHao Particular Carbon Black Co. Ltd. (China).

The PVDF/CB (85/15, wt%wt%) pellets (denoted as cPVDF) were prepared using a twin-screw extruder (Nanjing Giant Co. Ltd, China) prior to the layer-multiplying coextrusion. Subsequently, the dried cPVDF pellets and pure PVDF pellets were co-extruded from two extruders forming a 2-layer melt and then flowed through an assembly of LMEs. By applying 1, 3, and 5 LMEs, 4-, 16-, and 64-layer PVDF/cPVDF composites were obtained in this work, as illustrated in Fig. S1. By controlling the extruding speed of each extruder, the thickness ratio of PVDF and cPVDF layers was about 1:1, irrespective of the number of layers. Thus, the CB content in each multilayered specimen was about 7 wt% (the CB loading in cPVDF layers was maintained at 15 wt%). The thickness of each extrudate was maintained at 1 mm.

For comparison, neat PVDF was prepared by using only one extruder of above coextrusion system combining 5 LMEs, in order to keep a similar processing history to 64-layer system. The thickness of the extrudate was 1 mm.
**II. Characterizations**

**Polarized Optical Microscope**

The polarized optical microscope observation was performed using an Olympus BX51 (Japan) polarizing microscope equipped with an attached CCD camera. The transmission mode was applied for observing the multilayered structure, while the reflection mode was applied for the surface observation after the breakdown test.

**Dielectric measurement**

Dielectric properties of each specimen in the thickness direction were measured using an Agilent
4294A impedance analyzer (US). The typical applied ac voltage was 1 V, and the test frequency range was $10^2$–$10^7$ Hz. Prior to the tests, the contact faces of the specimen were coated with conductive silver paste for eliminating contact resistance. The dielectric permittivity ($\varepsilon$) and loss factor ($\tan \delta$) were obtained from the tests.

**Breakdown strength Test**

The electrical breakdown test was carried out on a high voltage tester, CS2671A (Nanjing Chang Sheng Instrument Co. Ltd). Each sample was placed between two sphere-like electrodes and immersed in insulating silicone oil. During the test, the applied DC voltage rose rapidly until the samples broke down with the warning current value preset at 5 mA. At least five data points for each sample were collected, and the average value was calculated.

**D–E hysteresis test**

D–E hysteresis loops of the unpoled multilayered cylindrical specimens were measured at 10 Hz with a ferroelectric instrument (Radiant Technologies, Inc. Albuquerque, NM, US.). The tests were carried out under the applied voltage of 1500V, the preset voltage would maintain for one second before the tests.