Supporting Information

Designing of Ferroelectric/Linear Dielectric Bilayer Films: An Effective Way to Improve the Energy Storage Performances of the Polymer-Based Capacitors

Chen Chen, †, ‡ Jiwen Xing, † Yang Cui, †, ‡ Changhai Zhang, †, ‡ Yu Feng, †, ‡ Yongquan Zhang, †, ‡ Tiandong Zhang, *, †, ‡ Qingguo Chi, *, †, ‡ Xuan Wang, †, ‡ Qingquan Lei‡

†School of Electrical & Electronic Engineering, Harbin University of Science and Technology, Harbin 150080, PR China
‡Key Laboratory of Engineering Dielectrics and Its Application, Ministry of Education, Harbin University of Science and Technology, Harbin 150080, PR China
*Corresponding Author: Tiandong Zhang and Qingguo Chi. E-mail: tdzhang@hrbust.edu.cn and qgchi@hotmail.com
SUPPLEMENTAL RESULTS

The microscopic morphology of the film can be described by Figure S1. It can be found that the individual films and bilayer films exhibit uniform and dense structure, especially, no obvious interface separation can be observed for the bilayer films.

![Figure S1. Cross sectional images of polymer films. (a) Individual PI films. (b) Individual P(VDF-TrFE-CFE) films. (c) P(VDF-TrFE-CFE)/PI (17 vol.%), bilayer films. (d) P(VDF-TrFE-CFE)/PI (33 vol.%), bilayer films. (e) P(VDF-TrFE-CFE)/PI (67 vol.%), bilayer films. (f) P(VDF-TrFE-CFE)/PI (83 vol.%), bilayer films.](image)

The polarization behaviors are studied by measuring the $D$-$E$ loops, as shown in Figure S2. With the increase of PI volume ratio, Excitingly, compared to that of individual P(VDF-TrFE-CFE) films, the residual electric displacement for constructed P(VDF-TrFE-CFE)/PI bilayer films reduces significantly. It can be found that the maximum electric displacement decreases gradually with the increase of PI volume ratio, which is consist with the results of dielectric constant.
Figure S2. D-E loops of P(VDF-TrFE-CFE)/PI bilayer films. (a) P(VDF-TrFE-CFE)/PI (17 vol.%) bilayer films. (b) P(VDF-TrFE-CFE)/PI (33 vol.%) bilayer films. (c) P(VDF-TrFE-CFE)/PI (67 vol.%) bilayer films. (d) P(VDF-TrFE-CFE)/PI (83 vol.%) bilayer films.

According to the series capacitor model, the dielectric constant of the bilayer nanocomposites is calculated by the equation (R1):

$$\frac{1}{\varepsilon} = \frac{x_1}{\varepsilon_1} + \frac{x_2}{\varepsilon_2}$$  \hspace{1cm} (S1)

where $\varepsilon$ is the dielectric constant of the P(VDF-TrFE-CFE)/PI bilayer films, $\varepsilon_1$ and $x_1$ are the dielectric constant and volume ratio of the P(VDF-TrFE-CFE) layer, and $\varepsilon_2$ and $x_2$ are the dielectric constant and volume ratio of the linear PI. In equation (S1), $\varepsilon$ is determined by using $\varepsilon_1 ($~9.27) and $\varepsilon_2 ($~3.58) from the experimental data, while $x_1$ and $x_2$ are from the following equations:
\[ x_1 = \frac{d_1}{d_1 + d_2} \]  \hspace{1cm} (S2)

\[ x_2 = \frac{d_2}{d_1 + d_2} \]  \hspace{1cm} (S3)

where \( d_1 \) and \( d_2 \) are the film thicknesses of the P(VDF-TrFE-CFE) layer and the PI layer, respectively. According to the results, the dielectric constant of the P(VDF-TrFE-CFE)/PI bilayer film is located between the single-layer PI film and the single-layer P (VDF-TrFE-CFE) film.

As shown in Figure S4, by the simulation method of COMSOL Multiphysics, the space charges tend to accumulate at the interface between P(VDF-TrFE-CFE) and PI layers in the bilayer films due to the large mismatch of the electrical resistivity or dielectric constant in adjacent layers.

**Figure S4.** The space charge density simulation for bilayer films. (a) P(VDF-TrFE-CFE)/PI(17 vol.%) bilayer films. (b) P(VDF-TrFE-CFE)/PI(33 vol.%) bilayer films. (c)P(VDF-TrFE-CFE)/PI(67 vol.%) bilayer films. (d) P(VDF-TrFE-CFE)/PI (83 vol.%) bilayer films.

The basic steps of COSMOL Multiphysics simulations:

Step 1: Creating a new Comsol Multiphysics model to configure the basic settings of the model.

Model Wizard selects the space dimension as 2D, the selected physical field is the Electric Current (ec), Default study selection is Stationary.

Step 2: Building a geometric model.
Length unit is selected as micrometer (μm), rectangle 1 is newly created as PI layer of P(VDF-TrFE-CFE)/PI(50vol.%) bilayer films (The width and height of Rectangle 1 are 20 μm and 7.5 μm, respectively), rectangle 2 is newly created as P(VDF-TrFE-CFE) layer of P(VDF-TrFE-CFE)/PI(50vol.%) bilayer films (The width and height of Rectangle 2 are 20 μm and 7.5 μm, respectively).

Step 3: Configuring the material of the geometric model.

Empty material 1 and empty material 2 are added as P(VDF-TrFE-CFE) and PI, The dielectric constant and conductivity of P (VDF-TrFE-CFE) are set to 9.3 and 1×10^{-14} S/m, respectively(The area of rectangular 1 is selected), The dielectric constant and conductivity of PI are set to 3.6 and 1×10^{-12} S/m, respectively(The area of rectangular 2 is selected).

Step 4: Configuring the parameters of the current field.

The electric potential 1 is newly created and the value is set to 300V(The selected area is the bottom edge of rectangle 2), The electric potential 2 is newly created and the value is set to 0V as ground potential(The selected area is the top edge of rectangle 1).

Step 5: Configuring the grid.

The size of unit is chosen to be finer.

Step 6: Allocation.

Step 7: The expression of the potential 2D plot group was replaced with space charge density(ec.rhoq). Its unit is C/m^3.

Step 8: Exporting the simulation result.

The thickness of each film used in the breakdown strength measurement is shown in Table S1.

<table>
<thead>
<tr>
<th>PI</th>
<th>P(VDF-TrFE-CFE)/PI(17vol.%)</th>
<th>P(VDF-TrFE-CFE)/PI(13vol.%)</th>
<th>P(VDF-TrFE-CFE)/PI(15vol.%)</th>
<th>P(VDF-TrFE-CFE)/PI(14vol.%)</th>
<th>P(VDF-TrFE-CFE)/PI(12vol.%)</th>
<th>P(VDF-TrFE-CFE)/PI(17vol.%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>12μm</td>
<td>11μm</td>
<td>13μm</td>
<td>15μm</td>
<td>14μm</td>
<td>12μm</td>
<td>17μm</td>
</tr>
<tr>
<td>13μm</td>
<td>11μm</td>
<td>13μm</td>
<td>15μm</td>
<td>14μm</td>
<td>15μm</td>
<td>16μm</td>
</tr>
<tr>
<td>12μm</td>
<td>10μm</td>
<td>14μm</td>
<td>16μm</td>
<td>15μm</td>
<td>14μm</td>
<td>15μm</td>
</tr>
<tr>
<td>14μm</td>
<td>10μm</td>
<td>15μm</td>
<td>16μm</td>
<td>15μm</td>
<td>14μm</td>
<td>15μm</td>
</tr>
<tr>
<td>15μm</td>
<td>12μm</td>
<td>12μm</td>
<td>15μm</td>
<td>14μm</td>
<td>16μm</td>
<td>17μm</td>
</tr>
<tr>
<td>13μm</td>
<td>13μm</td>
<td>14μm</td>
<td>15μm</td>
<td>13μm</td>
<td>16μm</td>
<td>15μm</td>
</tr>
<tr>
<td>14μm</td>
<td>11μm</td>
<td>14μm</td>
<td>14μm</td>
<td>15μm</td>
<td>17μm</td>
<td>16μm</td>
</tr>
<tr>
<td>13μm</td>
<td>12μm</td>
<td>13μm</td>
<td>13μm</td>
<td>16μm</td>
<td>15μm</td>
<td>14μm</td>
</tr>
</tbody>
</table>

The scraper mentioned in the manuscript means a film applicator with adjustable scale function (Produced by Hongjuli Test Equipment Factory in Tianjin). Figure S5 gives a concrete schematic diagram, the distance between scraper and substrate can be changed by fastening the screws. Then the films with different thickness can be obtained by adjusting the distance between scraper and substrate.

![Figure S5 Schematic diagram of fabrication equipment.](image-url)