Polymer Dielectric Films Exhibiting Superior High-Temperature Capacitive Performance by Utilizing an Inorganic Insulation Interlayer

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Finite-element simulations

In order to qualitatively understand the charge carrier behaviors in nanocomposites, a unipolar electron injection and transport model is established. Here, we describe electrons injected from the metal electrode into the dielectric by Schottky thermionic emission, which is expressed as

$$J_e = A T^2 \exp\left(-\frac{\varphi_i}{k_B T}\right) \exp\left(\frac{eE}{4\pi\varepsilon_0 \varepsilon_r}\right)$$

(S1)

where \(J\) is the current density, \(A\) is the Richardson constant, \(T\) is the temperature, \(\varphi_i\) is the injection potential barrier, \(k_B\) is the Boltzmann constant, \(e\) is the elementary charge, \(E\) is the electric field, \(\varepsilon_0\) is the vacuum permittivity, \(\varepsilon_r\) is the relative permittivity, respectively.

The behavior of charge within the films is governed by the following equations:

Poisson’s equation,

$$\frac{\partial \psi(r,t)}{\partial \tau} - \frac{\rho(r,t)}{\varepsilon_0 \varepsilon_r}$$

(2)

Current continuity equation,

$$\frac{\partial n(r,t)}{\partial \tau} + \frac{\partial J(r,t)}{\partial \tau} = s$$

(3)
Charge transport equation,

\[ J(r, t) = \mu(r, t)n(r, t)E(r, t) \]  \hspace{1cm} (4)

where \( \psi \) the electrical potential, \( \rho \) the total charge density (here equal \( n(r, t) \)), \( s \) the source term (\( s=0 \) in this simulation), \( \mu \) the mobility of charge.

**Table 1**

Definition of parameters used in this simulation.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Values</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>Charge mobility of PEI</td>
<td>( 1 \times 10^{-10} )</td>
<td>cm(^2)/(V\cdot s)</td>
</tr>
<tr>
<td>Charge mobility of MgO</td>
<td>( 1 \times 10^{-13} )</td>
<td>cm(^2)/(V\cdot s)</td>
</tr>
<tr>
<td>Dielectric constant of PEI</td>
<td>3.15</td>
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</tr>
<tr>
<td>Dielectric constant of MgO</td>
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<td>Electric field</td>
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<td>MV/m</td>
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<td>Temperature</td>
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<td>K</td>
</tr>
<tr>
<td>Schottky barrier</td>
<td>1.55</td>
<td>eV</td>
</tr>
</tbody>
</table>
Figure S1 Cross-sectional structure of SEM images of (a) PBP films, (b) PEI/BN films, (c) PEI/MgO films.

Figure S2 SEM-EDX mapping of PMP films.
Figure S3 Representative low-magnification of SEM image of PMP films.

Figure S4 XRD patterns of (a) MgO powder. (b) PEI/MgO films. (c) PMP films. (d) BN powder. (e) PEI/BN films. (f) PBP films.
Figure S5 FTIR spectra of (a) PEI/MgO films. (b) PMP films. (c) PEI/BN films. (d) PBP films.

Figure S6 Frequency-dependent and temperature-dependent dielectric properties measured at 150°C and 1 kHz, respectively. (a) (b) PEI/MgO films. (c) (d) PEI/BN films. (e) (f) PBP films.
Figure S7 Electronic structure of (a1) PEI, (a2) MgO, (a3) PEI and MgO. Band diagrams at PEI/MgO interface. (b) PEI and MgO interlayer before contact. (c) PEI and MgO interlayer after contact. Herein, $E_g$ is bandgap, $E_F$ is Fermi levels, $E_{\text{HOMO}}$ stands for the highest occupied molecular orbital of the dielectric, $E_{\text{cutoff}}$ represents the energy of secondary electron cutoff. $\phi_p$ is the work function, $E_{\text{A}}$ is the electron affinity, $E_{\text{vac}}$ is the vacuum energy level. $\phi_e$ is the potential barrier for electrons, and $\phi_h$ is the potential barrier for holes.
Figure S8 Weibull distribution of (a) PEI/BN films. (b) PBP films. (c) PEI/MgO films measured at 150°C. (d) Comparison of breakdown strength of resultant films in this work measured at 150°C.
Figure S9 Hysteresis loops measured at 150°C of (a) PEI/MgO-3 film. (d) PEI/BN-3 film. (g) PBP-5 film. Discharged energy density and efficiency of (b) PEI/MgO-3 film. (e) PEI/BN-3 film. (h) PBP-5 film at various temperatures. Discharged energy density and efficiency of (c) PEI/MgO films. (f) PEI/BN films. (i) PBP films measured at 150°C.

Figure S10 Cross-sectional structure of SEM images of PMP films after 50000 charge/discharge cycles.