# All-organic Composites for Ferroelectric Polymer/Cellulose with High Energy Density and Efficiency by Hydrogen Bonds

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**Characterization**

FT-IR (Avatar-360, Thermo Nicolet) and XRD (Smartlab, Rigaku) were used to detect the crystallographic transition of PVDF phase in PVDF/MG composites. The wavenumber range of FTIR was from 650 $cm^{-1}$ to 4000 $cm^{-1}$. The scanning speed of XRD was 5°/min and the scanning range was from 5° to 50°.

The behavior of melting and crystallization for PVDF/MG composites were tested by DSC (Perkin-Elmer). The range of temperature is from 30°C to 240°C with a heating and cooling rate of 10 °C/min.

The morphology characterizations and energy dispersive spectrometer (EDS) characterizations were conducted on a scanning electron microscopy (Hitachi S4800).

The permittivity of the composite films was tested with an LCR instrument (Keysight Technologies) from 100 to $10^6$ Hz at 1 V.

The breakdown strength, energy capacity, efficiency and leakage current of the composite films were tested with a ferroelectric test system (CPE1901, Polyk) and electrostatic meters (6517B, Keithley) at $10^3$ Hz. The films were vaporized with a diameter of 3mm (area 7.068 $mm^2$) platinum electrodes on both sides before test.

The thermally stimulated discharge current (TSDC) tests were performed by a Keithley 6517B amperemeter equipped with a Quatro-Cryosystem temperature control system. First, all samples were applied 1000V DC poling voltage for 30 min at 100 °C, then quickly cooled the samples to − 60 °C under the premise of maintaining the electric field. After keeping the electric field at − 60 °C for 10 min, the sample is short-circuited.

Finally, the sample is heated to 140 °C at a heating rate of 3 °C/min and the current is recorded.

Density functional theory (DFT) calculations were carried out with the Materials Studio 2020 software at the Dmol3 module and Forcite code. All repeat units were optimized before the calculations. The molecular model of GMA, MMA, and VDF only contains one repeat unit and the molecular model of VDF + MMA and VDF + GMA contains one VDF unit and one MMA or GMA unit. The binding energy between VDF and MMA or GMA was obtained by Forcite code calculation and analysis. The binding energy (Eint) was calculated by the following formula,

$$E_{int} = E_{complex} - (E_{OH} + E_{GMA\ or\ VDF})$$

where Ecomplex, EVDF, and EMMA or GMA represent the total energies of complexes, VDF, and MMA or GMA, respectively.