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Supplementary Informations

Enhanced Energy Storage Density of All-Organic Fluoropolymer Composite Dielectric via Introducing Crosslinked Structure

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Synthesis of unsaturated PVDF

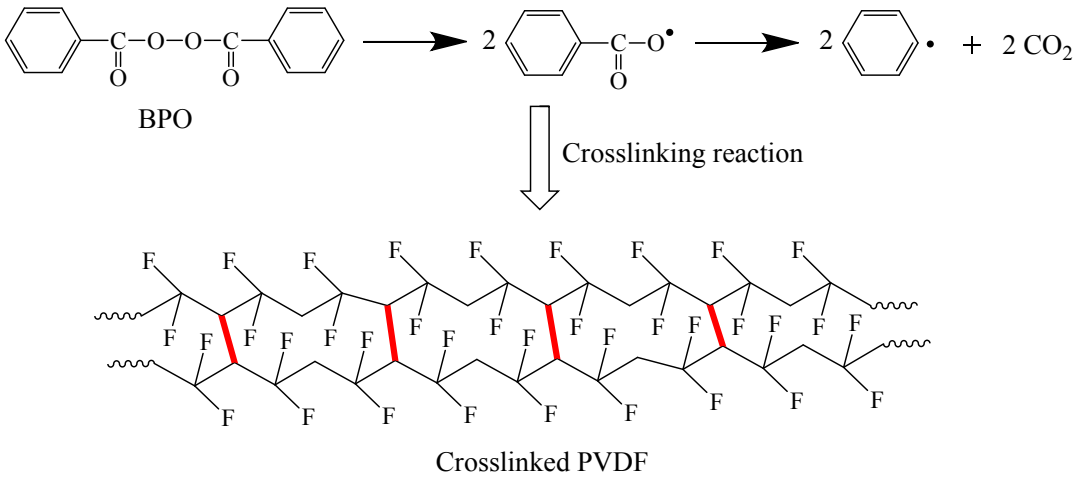
The unsaturated PVDF was synthetized via a nucleophilic substitution reaction. First, 5 g copolymer P(VDF-CTFE) were dissolved in NMP for 0.5 h at room temperature and formed a transparent and homogeneous solution. Second, 13.4 mmol of TEA were added into the P(VDF-CTFE) solution to ensure complete dispersion. The mixture was further stirred vigorously for 24 h at 50 ^{o}C to yield a homogenous mixture of unsaturated PVDF and residual TEA. Afterwards, the mixture was precipitated in dilute hydrochloric acid solution to remove the residual TEA. The product was dissolved in acetone followed by precipitation in methanol three times and dried at 50 ^{o}C for 12 h in a vacuum oven.[30]

30 S.B. Tan, J.J. Li, G.X. Gao, H.Y. Li and Z.C. Zhang, *J. Mater. Chem.*, 2012, **22**, 18496-18504.

Fabrication and crosslinking of unsaturated PVDF

A typical crosslinking reaction was conducted as followings. First, 0.75 g PVDF containing unsaturation powders were dispersed in DMF and stirred for 5 h to completely dissolve at room temperature. Then, 37.5 mg BPO was slowly added into the PVDF containing unsaturation solution. The polymer solution was cast on a glass plate and dried at 90 ^{o}C for 6 h. After that, the films were heated in a vacuum oven at 160 ^{o}C for 12 h to allow the thermal crosslinking reaction. The prepared samples were finally treated at 180 ^{o}C under vacuum for 24 h to remove residual BPO and crosslinking byproducts.[30]

The decomposition of initiator



Synthesis of PVDF terpolymer

The PVDF terpolymer was prepared via a hydrogenation process. 5 g P(VDF-CTFE) and 0.25 g AIBN were dispersed in 150 mL purified THF at room temperature for 0.5 h and degassed three times with dry nitrogen-vacuum cycles. Next, nBu$_3$SnH was injected into the reaction system using a syringe, and the reaction was kept stirring at 60 °C for 24 h. The mixture was slowly poured into water to precipitate a polymer, and extracted with *n*-hexane in a Soxhlet extractor for 12 hours to remove residual nBu$_3$SnH. After drying in a vacuum oven at 80 °C for 24 h, the PVDF terpolymer was synthetized [31].

31 Y. Lu, J. Claude, Q. Zhang and Q. Wang, *Macromolecules*, 2006, **39**, 6962-6968.

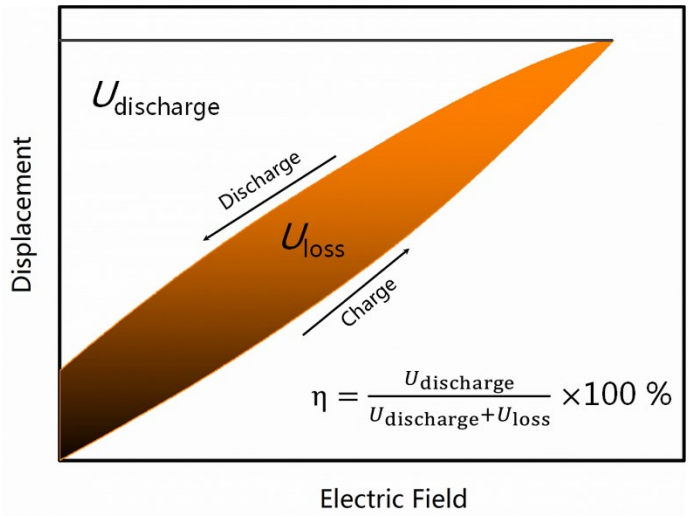


Fig. S1 *D-E* loop of a dielectric material. $U_{discharge}$ is the discharged energy density, and U_{loss} is the energy loss. The value of $U_{discharge}$ and U_{loss} can be obtained by integrating calculation. $U_{discharge}$ + U_{loss} is the totally charged energy density, and the discharge-charge efficiency (η) defines the content of $U_{discharge}$.

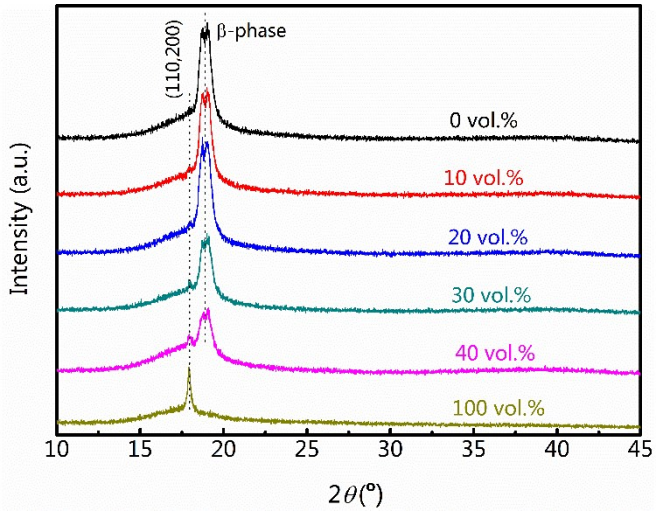


Fig. S2 XRD patterns of the composite films with different c-PVDF volumes.

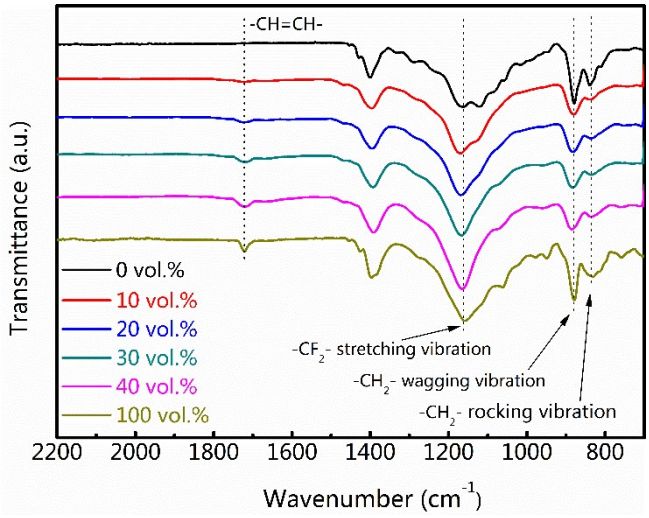


Fig. S3 FTIR results of PVDF-based polymers and polymer composites.