Fluorinated Polydopamine Shell Decorated Fillers in
Polytetrafluoroethylene Composite for Achieving Highly
Reduced Coefficient of Thermal Expansion

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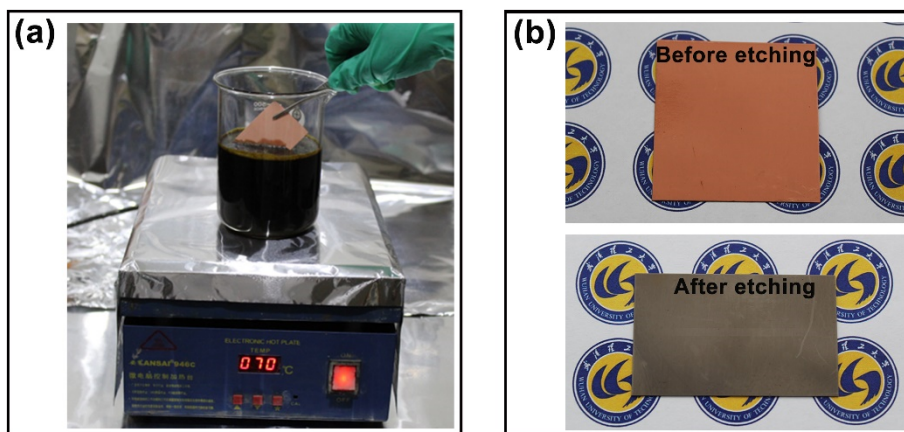† These authors contributed equally to this work.

Figure S1. Photograph of etching process of CCL (a) and photographs of the composite
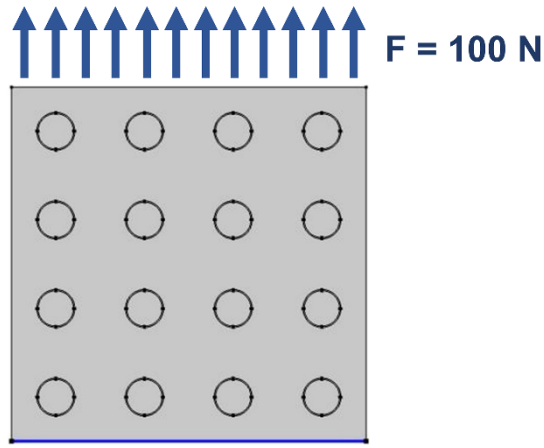
before and after etching (b).

Figure S2. Boundary conditions of the finite element simulation.

In this study, the PTFE-based composite was established as a two-dimensional square model with a side length of 14 μm. According to the volume percentage of fillers, a 4 x 4 array of SiO_2 spheres with a radius (r). The r was established in the middle of the model, and the shell thickness (d) was established on the outside of the sphere. Correspondingly, the values of r and d were determined. The r is approximately 0.75 μm, and d is 0.02 μm determined by the TEM results (Figure 1). The PTFE-based composite was set as a linear elastic material. The boundary conditions were set beneath the model conditions. And a force of 100 N in the Y direction was applied to the upper section, as shown in Figure S2. By setting the boundary conditions, the stress distribution of the established two-dimensional model could be calculated under the applied force.
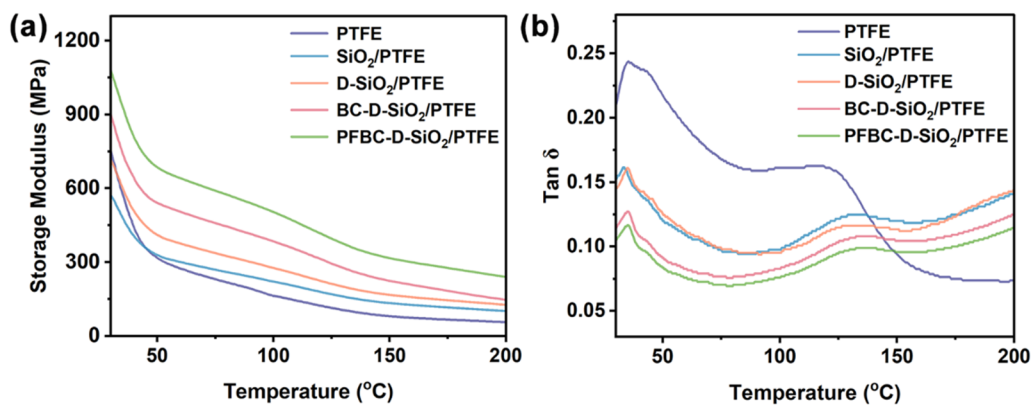
Figure S3. Dynamic mechanical analysis for storage modulus curves (a) and Tan δ curves (b) of neat PTFE, SiO$_2$/PTFE, D-SiO$_2$/PTFE, BC-D-SiO$_2$/PTFE, and PFBC-D-SiO$_2$/PTFE exfoliated composite coatings with 30 vol% of filler loading content.
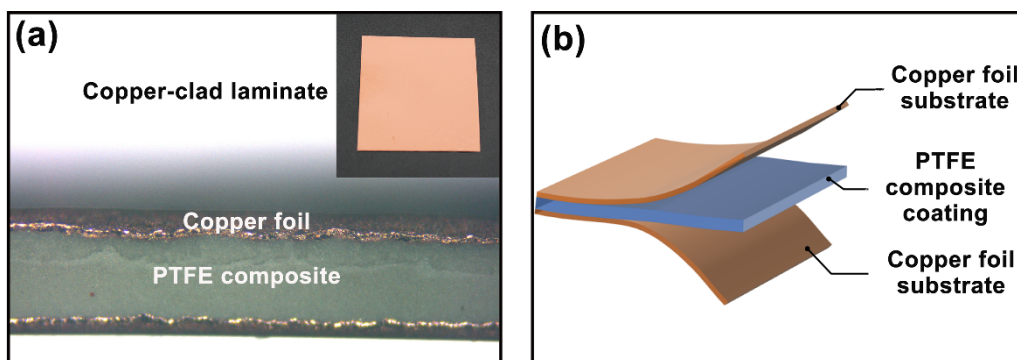
Figure S4. (a) Cross-sectional images of CCL (inset is the optical image of the surface of CCL) and (b) schematic illustration of CCL shows its sandwich structure.
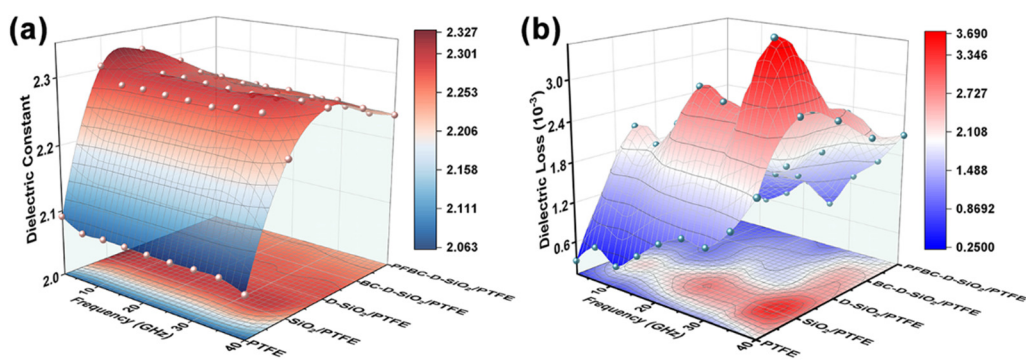
Figure S5. Frequency dependence of dielectric constant (a) and loss (b) of neat PTFE, the exfoliated SiO$_2$/PTFE, D-SiO$_2$/PTFE, BC-D-SiO$_2$/PTFE, and PFBC-D-SiO$_2$/PTFE composite coatings with 30 vol% of filler loading content.
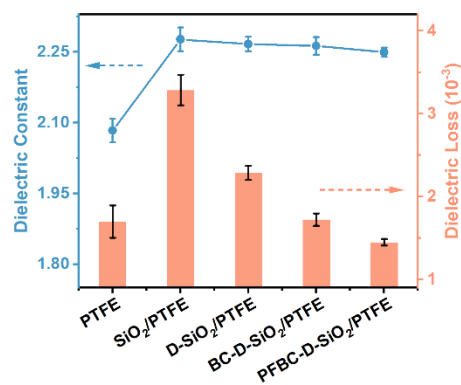
Figure S6. Dielectric properties (@ 30 GHz) of neat PTFE, SiO_2/PTFE, D-SiO_2/PTFE, BC-D-SiO_2/PTFE, and PFBC-D-SiO_2/PTFE exfoliated composite coatings with 30 vol% of filler loading content.
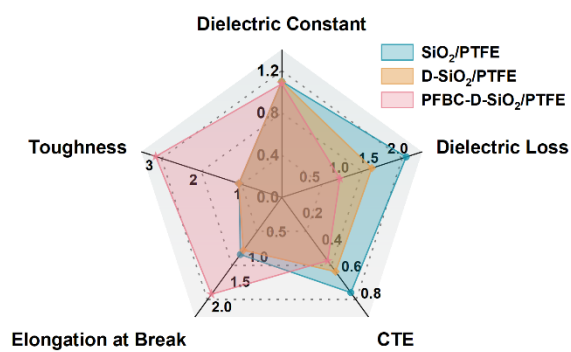
Figure S7. In comparison to the dielectric constant (D_k), dielectric loss (D_f), CTE, elongation at break, and toughness of neat PTFE, D-SiO$_2$/PTFE, and PFBC-D-SiO$_2$/PTFE exfoliated composite coatings with 30 vol% of filler loading contents. (Parameters of neat PTFE: D_k = 2.08, D_f = 0.00171, CTE = 160 ppm/°C, elongation at break = 92.36 %, and toughness = 5.33 MJ/m^3.)