

## Supporting Information for Publication

### A rational design for reconciling high permittivity and breakdown strength in layered PVDF composites from TaB<sub>2</sub>@Ta<sub>2</sub>O<sub>5</sub> nanofiller induced Schottky barrier effect

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Fig. S1 exhibited the XRD result of the uncoated TaB<sub>2</sub> core. By comparison with the result in Fig. 2, the sharp peak at 28 degrees should come from the TaB<sub>2</sub> core. That meant some of the used TaB<sub>2</sub> particles were not nano-sized.

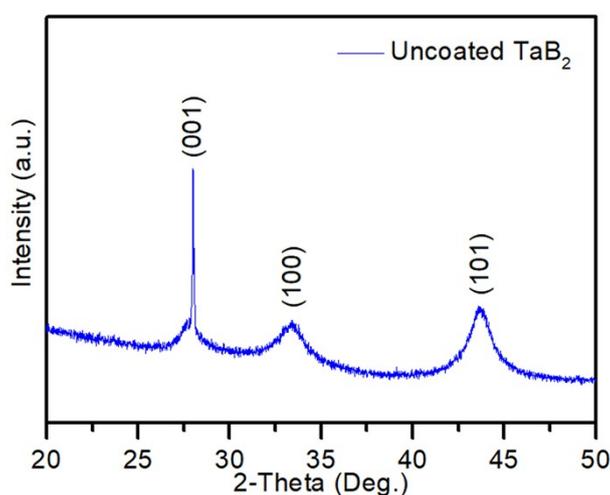
Fig. S2 exhibited the SEM, high-resolution TEM and electron diffraction pattern results of the core-shell filler.

Fig. S3 showed the permittivity results of some as-prepared samples as a function of testing frequency at room temperature. As discussed in the main text, those samples with gradually enhanced interface effects could be further verified based on the elevation of the frequency dependence of the permittivity (especially at the low frequency scope).

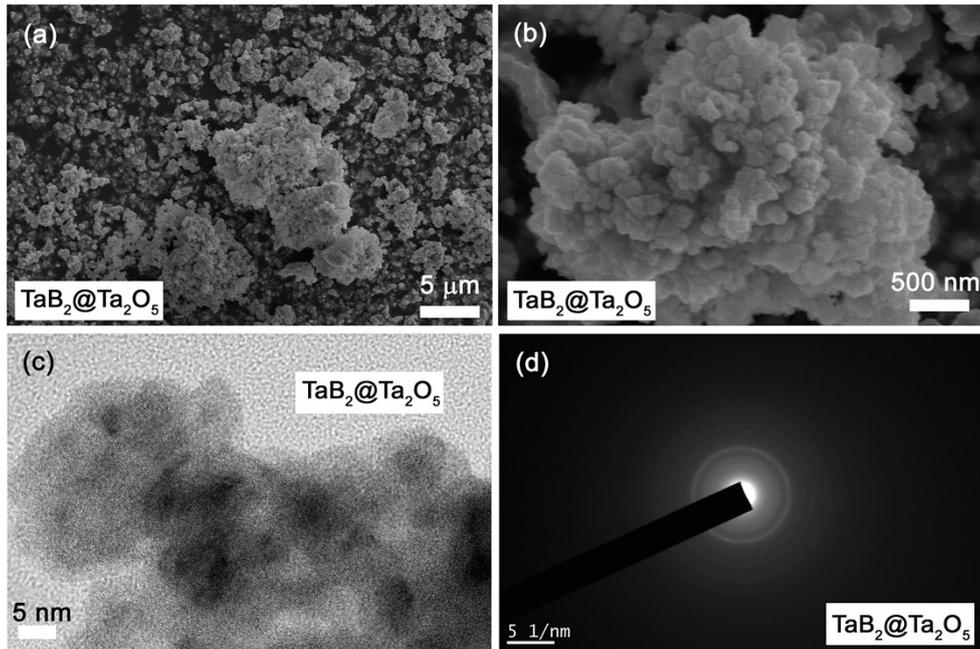
Fig. S4 showed the tested permittivity results of the homogeneous counterparts at 1 kHz. By comparison with the layered composites, the rather small differences about the permittivity results of both systems were confirmed. That suggested an insignificant effect of the layered structure on the permittivity of the entire composite.

Fig. S5 supplied the cross-sectional SEM result of the CS-H sample. The sandwich structure of the composite and the good filler dispersion in all the layers could be confirmed.

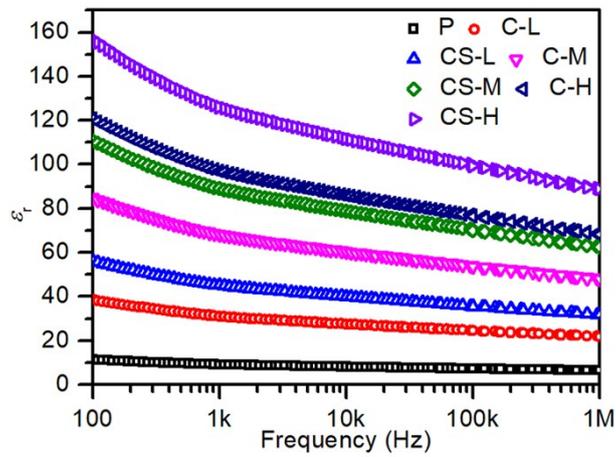
Fig. S6 showed the dc conductivity results of the even counterpart of CS-H as a function of electric field. Based on the results obtained in this figure and Fig. 8(a), the conductive ability of this even counterpart under the high applied fields was found to lie between C-H and CS-H samples, suggesting the obvious contribution of the interface barrier effect (from gradient sandwich structure) to low conductivity of the composite at high fields. Besides, the Ta<sub>2</sub>O<sub>5</sub> shell might play a more important role in achieving a low conductivity than the sandwich structure.



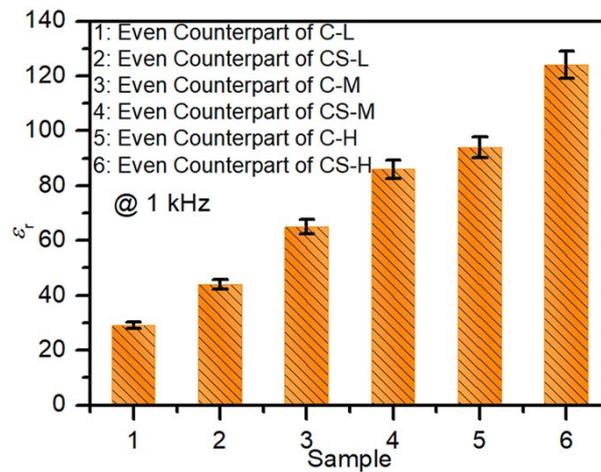
**Figure S1.** XRD result of the uncoated TaB<sub>2</sub> core.



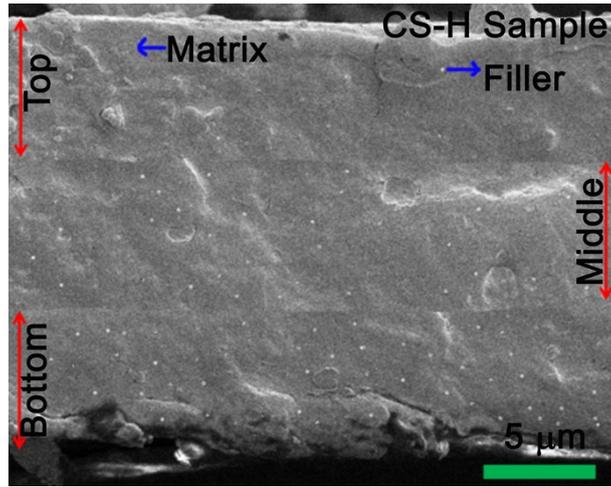
**Figure S2.** (a,b) SEM results, (c) high-resolution TEM result and (d) electron diffraction patterns of core-shell filler.



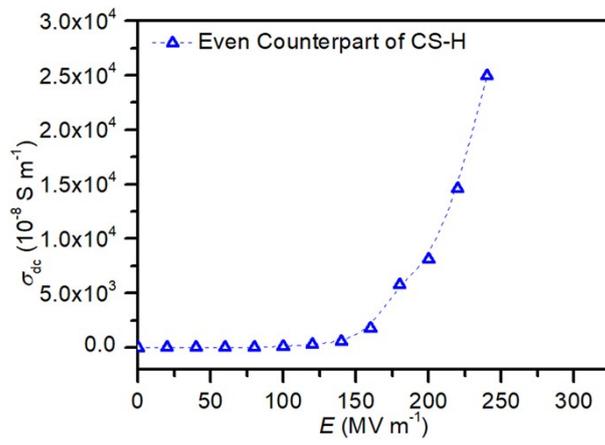
**Figure S3.** Permittivity results of some samples as a function of frequency at room temperature.



**Figure S4.** Permittivity results of the homogeneous counterparts at 1 kHz.



**Figure S5.** Cross-section SEM result of the CS-H sample.



**Figure S6.** DC conductivity results of the even counterpart of CS-H as a function of electric field.