

Supplementary Information for

**Electron scattering by Coulomb interactions of coupling
agents for high insulating aramid films**

Sidi Fan,¹ Xinghan Du,¹ Wenqi Zhang,¹ Kaixuan Sun¹, Rui Yang,¹ Fangcheng Lv,¹ Qing Xie,¹
and Xiang Yu^{1,2*}

¹*School of Electrical and Electronic Engineering, North China Electric Power University, Beijing
102206, China*

²*State Key Laboratory of Alternate Electrical Power System with Renewable Energy Sources,
North China Electric Power University, Beijing 102206, China*

*Corresponding author. E-mail: xiangyu@ncepu.edu.cn

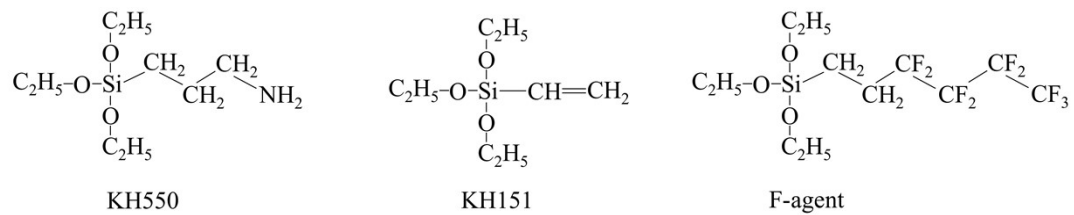


Figure S1. Chemical structures of KH550, KH151, and F-agent.

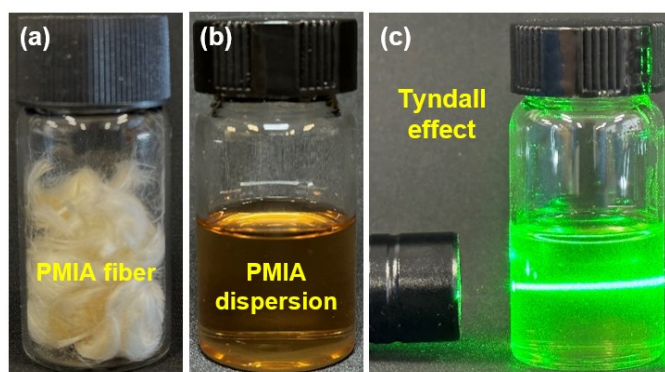


Figure S2. (a) Raw PMIA fibers. (b) PMIA dispersion. (c) Tyndall effect of PMIA dispersion.

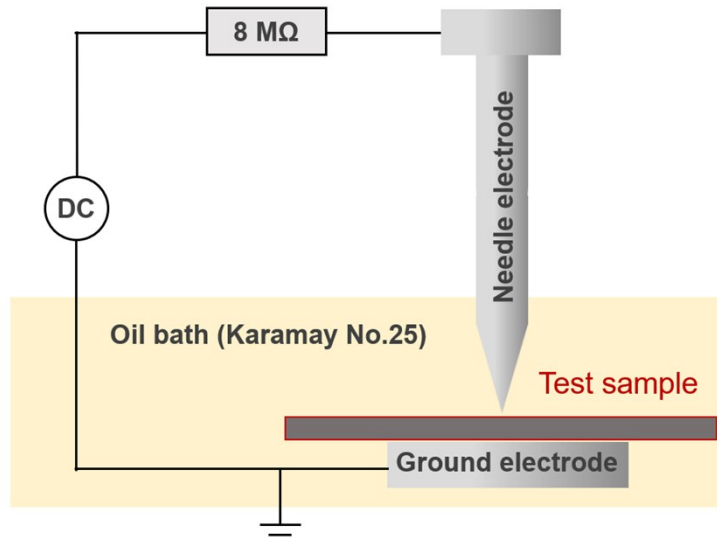


Figure S3. The test platform of breakdown strength.

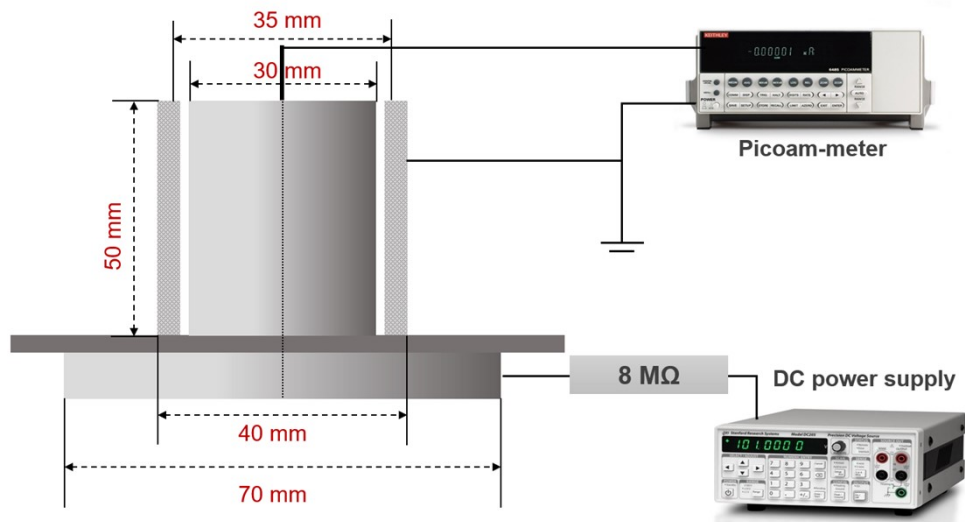


Figure S4. The test platform of volume conductivity.

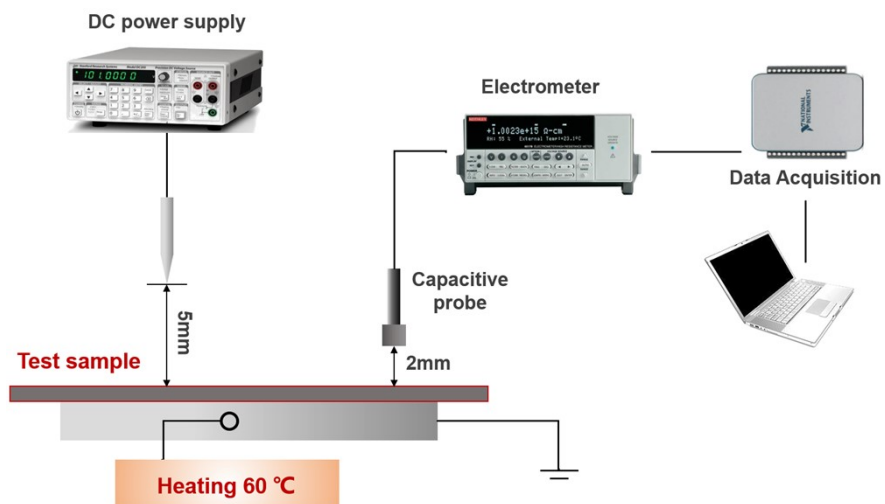


Figure S5. The test platform of isothermal surface potential decay (ISPD).

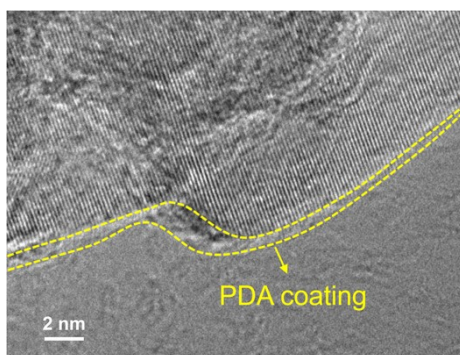


Figure S6. High-resolution TEM image of ND@PDA.

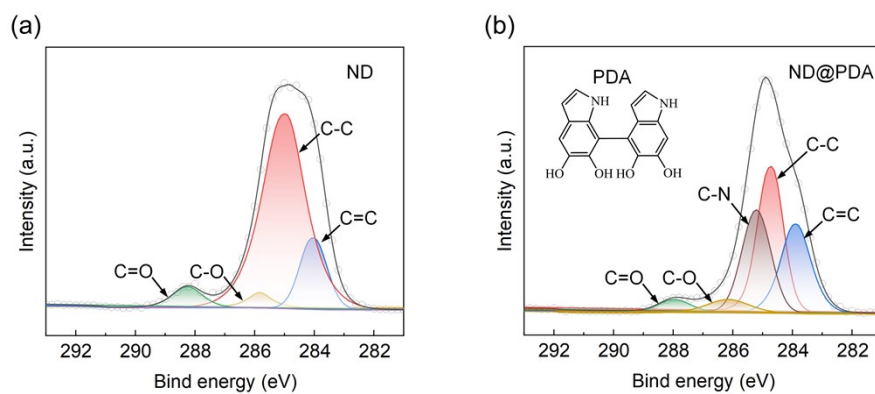


Figure S7. (a) The XPS spectra of deconvoluted C 1s peaks for ND and (b) ND@PDA.

Table S1. Elemental contents of four types of ND fillers from the XPS results.

Fillers	Atomic content (%)				
	C	N	O	Si	F
Pure ND	89.22	0.64	10.14	/	/
ND@KH550	83.57	2.84	11.38	2.21	/
ND@KH151	83.44	1.88	12.75	1.93	/
ND@F-agent	78.32	1.91	11.23	2.05	6.49

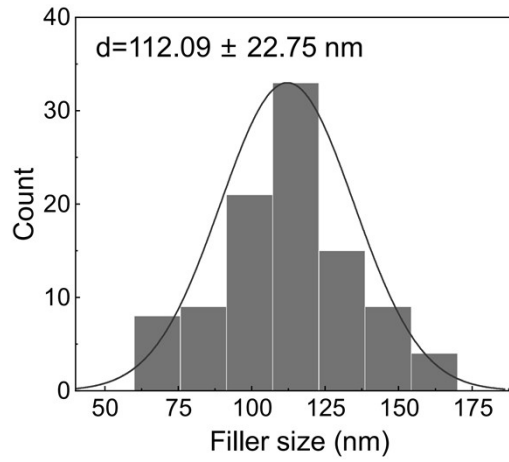


Figure S8. The size distribution of ND particles.

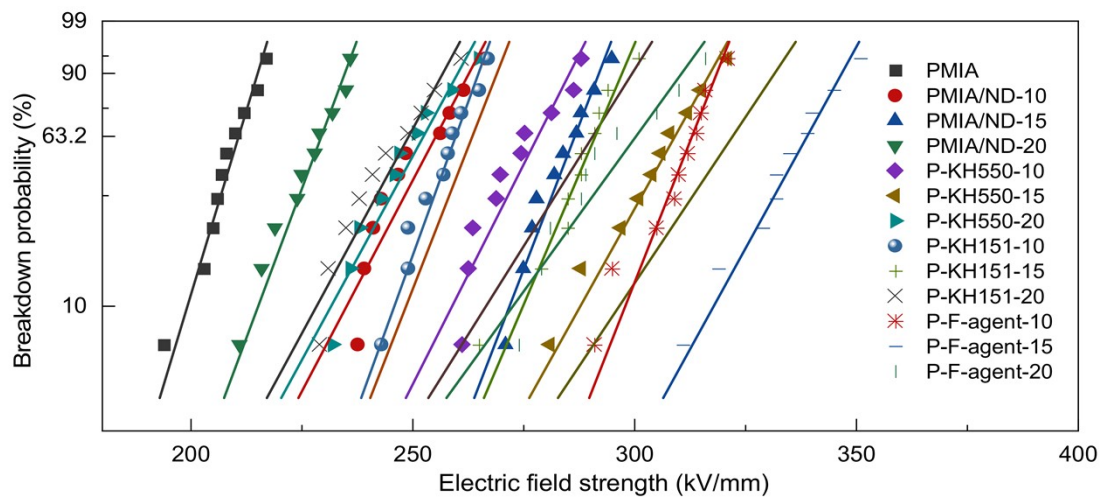


Figure S9. The Weibull distribution plots of E_b obtained from various films at doping ratios of 10 wt%, 15 wt%, and 20 wt%.

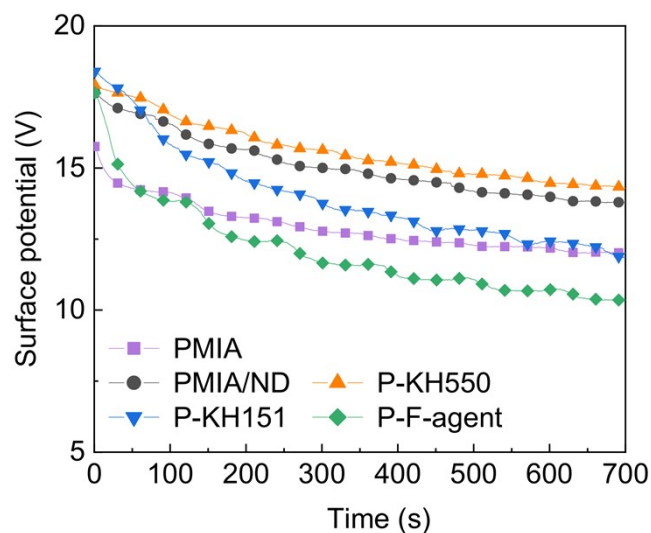


Figure S10. The ISPD curves of various films at a doping ratio of 15 wt%.

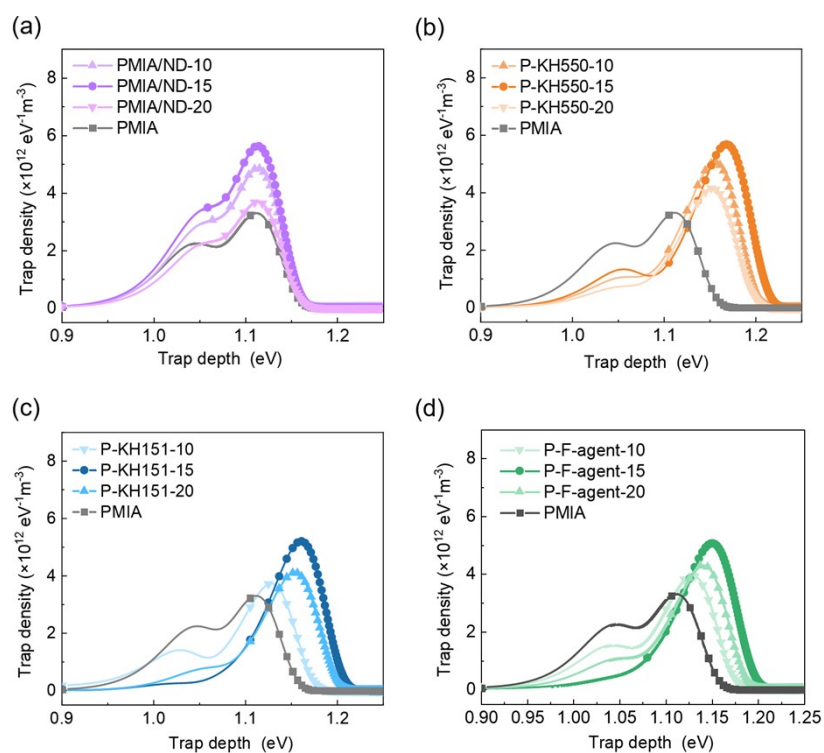


Figure S11. The trap depth-density curves of deep traps in various films.

Note 1. Band structures of PMIA and ND

Utilizing the Density Functional Theory (DFT) method, the energy band structures of PMIA and ND are analyzed based on the DMol³ module in *Materials Studio* software. The molecular models of PMIA and ND are shown in Figure S11, while the detailed calculation settings are outlined as follows.

Firstly, a Geometry Optimization procedure is conducted to refine the energy configurations of the molecular models. During this process, the PBE-GGA is used as the exchange-correlation functional. The accuracy of each parameter is set as described in Table S2. Additionally, the OTFG ultrasoft option is selected for the pseudopotential. Following the completion of the structural optimization, a single-point energy calculation is performed to analyze the energy band structure characteristics of each material.

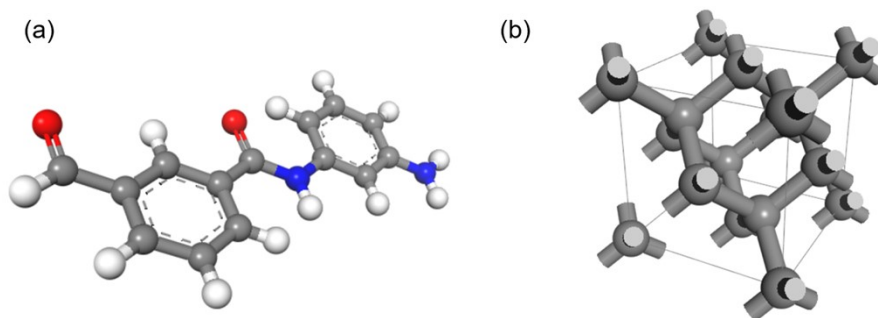


Figure S12. (a) Molecular structures of PMIA and (b) ND.

Table S2. The accuracy of each parameter.

Parameters	Accuracy
Energy convergence	5×10^{-6} eV/atom
Atom maximum force convergence	0.01 eV/Å
Displacement convergence	5×10^{-6} Å
Cutoff energy	400 eV
Self-Consistent Field (SCF) convergence	5×10^{-7} eV/atom

Note 2. The calculation of electrostatic potential distributions

The electrostatic potential distributions of each coupling agent are calculated based on the DFT method using Gaussian 16.0. During the calculations, the B3LYP hybrid functional and the 6-31G(d) basis set are employed. Subsequent quantitative analysis of the specific surface potential is displayed through the *Multwfn* program, with the following parameters: a grid spacing of 0.25 bohr and an electron density isosurface of 0.001 e/bohr³.

Note 3. Study of PMIA/ND composite models based on Molecular Dynamics

Composite models of PMIA/ND using various coupling agents are constructed in *Materials Studio* software. The interfacial interactions between PMIA/ND or PMIA/modified ND are then explored. The dynamic processes are calculated using the *COMPASS III* force field and further analyzed using the Forcite Plus toolbox.

The composite models encompass two distinct layers:

(1) Construction of the PMIA layer. A PMIA monomer molecule is constructed according to its chemical formula. Subsequently, an amorphous cell is constructed, consisting of 6 molecular chains with a polymerization degree of 10. This initial structure undergoes geometric optimization, employing ultra-fine calculation accuracy. To ensure sufficient relaxation of the molecular chains, dynamics calculations of 500 ps are conducted under both NVT and NPT ensemble conditions. The temperature is set at 298 K, while the pressure is set at 1.01×10^{-4} GPa, with a time step of 1.0 fs. These dynamic relaxations adhere to the Nose temperature control method and Berendsen pressure control methods.

(2) Construction of the ND layer. ND cells are sourced from the crystal database in MS. The (1 1 1) plane of ND is selected for cleavage, and the exposed C atoms are saturated with the -OH groups. By constructing a supercell task, the dimensions of the ND surface are adjusted to match those of the PMIA layer.

(3) Construction of the modified ND layer. Three types of coupling agents are grafted onto the saturated ND surface, with a specific grafting quantity of 3 for each coupling agent.

Utilizing the *Build Layers* module, the two crystal surfaces are integrated into an interface model (ND or modified ND at the bottom and PMIA atop). A vacuum layer with a thickness of 150 Å is set above the PMIA layer to confine its interactions only with the underlying ND surface. To enhance the accuracy of the interface structure, an additional round of geometric optimization is performed, followed by 500 ps of NVT dynamics. Upon the completion of the NVT dynamics calculation, the trajectory files from the final 100 ps are extracted for comprehensive analyses, focusing on the binding energy, MSD, and RDF of various composite models.

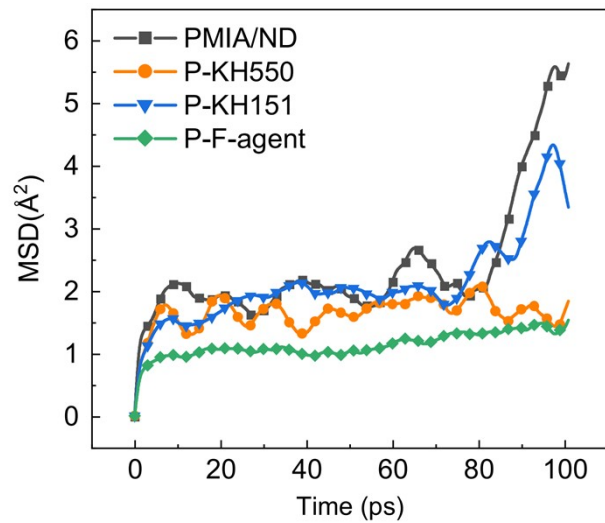


Figure S13. The MSD curves of PMIA molecular chains in various model.