

Supplementary Information

**Highly Insulating Thermoplastic Nanocomposites based on a Polyolefin Ternary Blend
for High-Voltage Direct Current Power Cables**

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Table S1. Formulations of all materials and σ_{DC} at 70 °C and 30 kV mm⁻¹; ^asingle measurement, a relative error of $\Delta x = 10\%$ is assumed; ^bmean of two measurements of two separately compounded samples, error calculated according to $\Delta x = (x_{max} - x_{min})/2$; ^cdata from ref. [Ouyang et al., *High Volt.*, 2022, 7, 251-259].

| | LDPE (wt%) | PP (wt%) | SEBS (wt%) | Al ₂ O ₃ (wt%) | σ_{DC} 30 kV mm ⁻¹ (10^{-15} S m ⁻¹) 70 °C |
|---|---------------|-------------|---------------|---|--|
| LDPE | 100 | - | - | - | 43.0 ± 4.3^a |
| LDPE:Al ₂ O ₃ | 98.7 | - | - | 1.3 | 9.6 ± 1.5^b |
| LDPE:Al ₂ O ₃ | 97.7 | - | - | 2.3 | 3.7 ± 0.6^b |
| PP | - | 100 | - | - | 1.4 ± 0.1^c |
| PP:LDPE | 75 | 25 | - | - | 20 ± 2.0^a |
| PP:LDPE:Al ₂ O ₃ | 72.8 | 25 | - | 2.2 | 2.5 ± 0.4^b |
| SEBS | - | - | 100 | - | 10.1 ± 1.0^a |
| SEBS:PP:LDPE | 71 | 24 | 5 | - | 11.6 ± 1.2^a |
| SEBS:PP:LDPE | 42 | 38 | 20 | - | 4.3 ± 0.4^a |
| SEBS:PP:LDPE:Al ₂ O ₃ | 68.9 | 24 | 5 | 2.1 | 2.89 ± 0.4^b |
| SEBS:PP:LDPE:Al ₂ O ₃ | 40.7 | 38 | 20 | 1.3 | 2.6 ± 0.4^b |

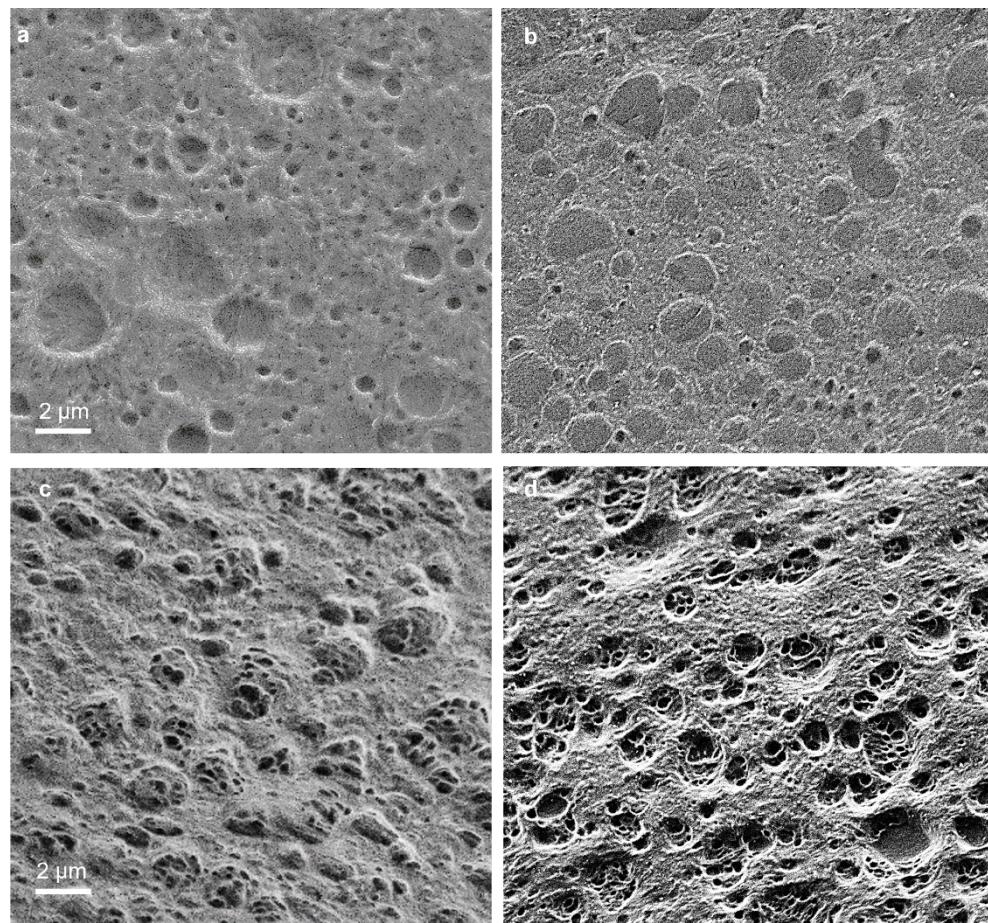


Figure S1. SEM micrographs of the cryofractured, etched and sputtered surfaces of (a): 25:57 PP:LDPE binary blend, (b): 25:72.8:2.2 PP:LDPE: Al_2O_3 binary blend nanocomposite, (c) 5:24:71 SEBS:PP:LDPE ternary blend , and (d) 5:24: 68.9:2.1 SEBS:PP:LDPE: Al_2O_3 ternary blend nanocomposite.

Table S2. Peak melting temperatures, T_m^{PP} and T_m^{PE} , and crystallization temperatures, T_c^{PP} and T_c^{PE} , of PP and LDPE extracted from first heating and cooling DSC thermograms as well as the crystallinity of PP and LDPE, X_c^{PP} and X_m^{PE} , calculated according to $X_c = \Delta H_f / \Delta H_f^0$ where ΔH_f is the enthalpy of fusion of the PP and LDPE fraction, respectively, and $\Delta H_{f,PP}^0 = 207 \text{ J g}^{-1}$ and $\Delta H_{f,PE}^0 = 293 \text{ J g}^{-1}$.

| | LDPE (wt%) | PP (wt%) | SEBS (wt%) | Al ₂ O ₃ (wt%) | T_m^{PP} (°C) | T_m^{PE} (°C) | T_c^{PP} (°C) | T_c^{PE} (°C) | X_c^{PP} (°C) | X_m^{PE} (°C) |
|---|---------------|-------------|---------------|---|--------------------|--------------------|--------------------|--------------------|--------------------|--------------------|
| LDPE | 100 | - | - | - | - | 112 | - | 96 | - | 42 |
| LDPE:Al ₂ O ₃ | 98.7 | - | - | 1.3 | - | 114 | - | 97 | - | 43 |
| PP | - | 100 | - | - | 167 | - | 110 | - | 38 | - |
| SEBS | - | - | 100 | - | - | - | - | - | - | - |
| SEBS:PP:LDPE | 42 | 38 | 20 | - | 167 | 115 | 112 | 96 | 39 | 39 |
| SEBS:PP:LDPE:Al ₂ O ₃ | 40.7 | 38 | 20 | 1.3 | 164 | 111 | 115 | 98 | 37 | 36 |

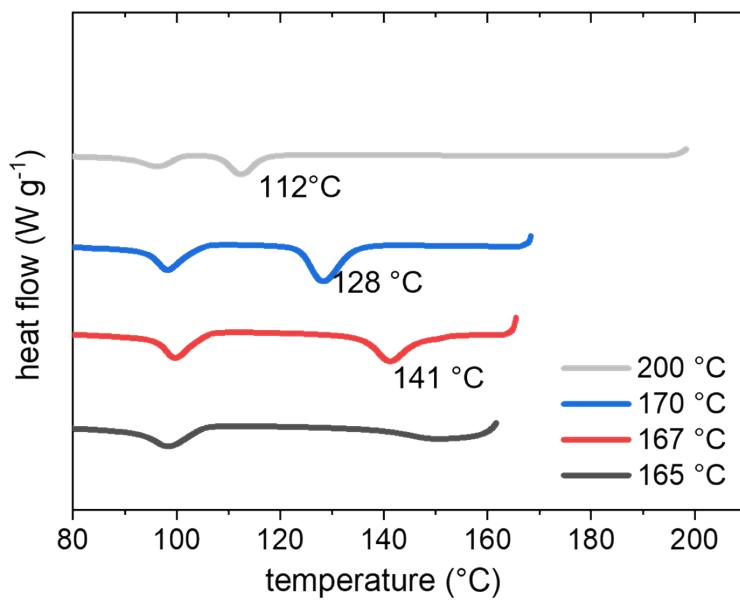


Figure S2. DSC cooling thermograms of 20:38:42 SEBS:PP:LDPE ternary blend after complete melting at 200°C (gray) and after partially melting at different self-seeding temperatures (blue, red and black), yielding a peak crystallization temperature of optimally nucleated PP of $T_c^{Max} \approx 141^{\circ}\text{C}$.

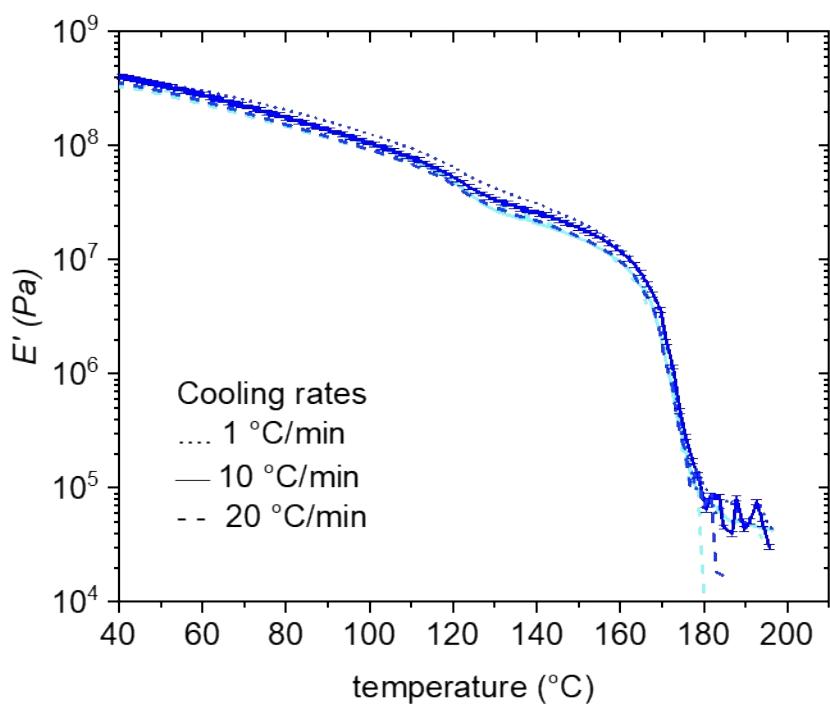


Figure S3. DMA heating thermograms showing the storage modulus E' of 20:38:42 SEBS:PP:LDPE ternary blends (sky blue) and 20:38:40.7:1.3 SEBS:PP:LDPE: Al_2O_3 ternary blend nanocomposites (blue), prepared with different cooling rates: -1 $^{\circ}\text{C min}^{-1}$ (dotted), -10 $^{\circ}\text{C min}^{-1}$ (solid) and -20 $^{\circ}\text{C min}^{-1}$ (dashed).

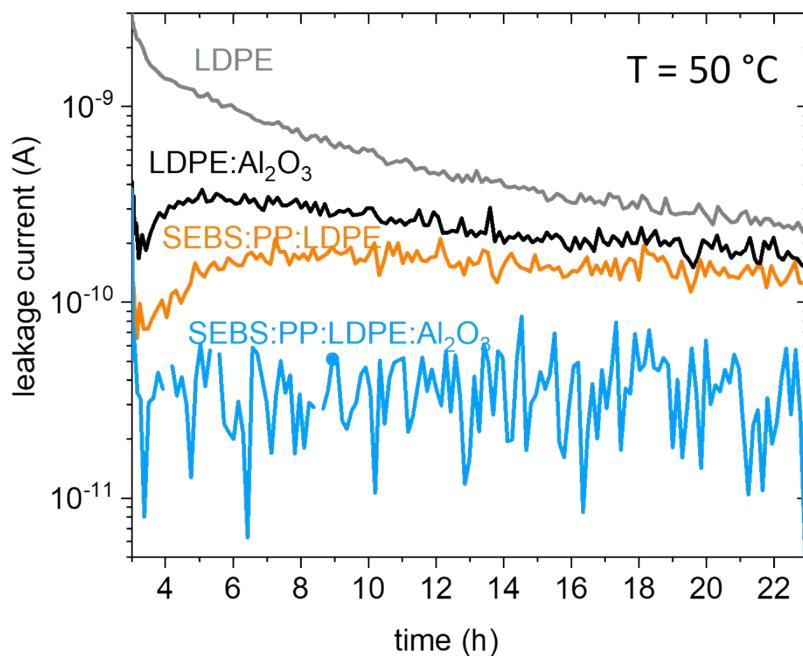


Figure S4. Leakage current at 50 °C and 30 kV mm⁻¹ as a function of time of neat LDPE (gray), 98.7:1.3 LDPE:Al₂O₃ (black), 20:38:42 SEBS:PP:LDPE (orange), and 20:38:40.7:1.3 SEBS:PP:LDPE:Al₂O₃ (blue); yielding σ_{DC} values of $3.9 \cdot 10^{-15}$ S m⁻¹, $2.5 \cdot 10^{-15}$ S m⁻¹, $1.6 \cdot 10^{-15}$ S m⁻¹ and $0.5 \cdot 10^{-15}$ S m⁻¹, respectively.

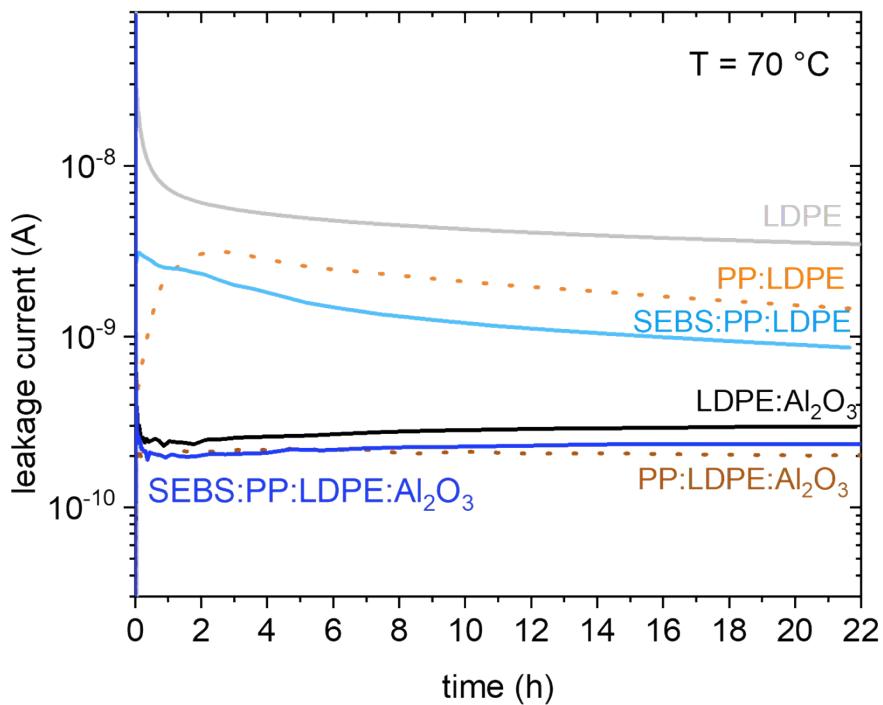


Figure S5. Leakage current at 70°C and 30 kV mm^{-1} as a function of time of neat LDPE (grey), 25:75 PP:LDPE binary blends (orange), 5:24:71 SEBS:PP:LDPE ternary blends (sky-blue), 97.7:2.3 LDPE: Al_2O_3 (black), 25:72.8:2.2 PP:LDPE: Al_2O_3 (brown) and 5:24:68.9:2.1 SEBS:PP:LDPE: Al_2O_3 (blue).