## Supporting Information

## On the Silica Surface Modification and Its Effect on Charge Trapping and Transport in PP Based Dielectric Nanocomposites

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In order to estimate the amount of injected and released space charge during polarization and depolarization, the areas under current versus time curves were calculated and compared. It is noteworthy that the short-circuit TSDC is a net sum of both, homo- and heteropolar discharge current components, and hence, the integrated charge represents the net charge. Figure 1a compares the amount of injected charges for each specimen during the isothermal polarization. There is a reduction in the injected charges upon introduction of silica nanoparticles, with this being also indicative of reduced conductivity for the silica nanocomposites. This reduction becomes more significant when the modified silica is incorporated into the system. This is due to the presence of the amine functional group on the silica surface which, due to the polarity, hinders further injection of space charges into the material. Figure S1a also well visualizes the decrease of space charge injection as an effect of interfacial crystallization (NC\_5 < NC\_6 & NC\_7 < NC\_8).



a.



Figure S1. a) Estimated amount of injected space charge during polarization; b) Percentage of

the released charge relative to the injected charge

Figure S1b demonstrates the values of relative released charges with regard to the amount of the injected charges. For the reference samples UP and NC\_0, less than 20% of the injected charges are released during depolarization, whereas in case of e.g. NC\_8 with a high APTES grafting density on silica, this value increases to ca. 55%. This indicates that for the samples with modified silica, lower space charge remains in the sample after depolarization. As mentioned before, NC\_5 and NC\_7 contain silica nanoparticles with a rougher APTES layer morphology due to the APTES island growth at lower reaction temperatures. This roughness can contribute to the formation of deeper traps at the filler-polymer interface, as can also be seen in the trap depth distribution graphs in Figure 9 (in the manuscript). This can be the reason for the lower amount of released charges for NC\_7 compared to NC\_8. This difference, however, is not observed in case of NC\_5 and NC\_6 with lower APTES grafting densities.