SUPPORTING INFORMATION

SEM images of the cross-sections of PEO/salt filled with clay-CNT hybrids are shown in **Supplementary Figures 1a-c** and **Figures 2a-c** (UV etched)**.** The samples were fractured in liquid nitrogen. In Figures 1a-c, the images show that clay-CNT hybrids are embedded in the polymer matrix and appear to be wrapped very well by the polymer. This indicates that nanofillers can restrict the crystallization of PEO as further supported by the X-ray diffraction. After UV etching (Figures 2a-c), more carbon nanotubes are exposed. The X-ray diffractions of the clay and clay-CNT hybrid are shown in **Supplementary Figure 3**. For pristine clay, the peak at around $2\theta=7.2^{o}$ corresponding to 001 plane reflection indicates the ordered stacked-layer structures of clay. Another peak at $2\theta=19.6^{o}$ is assigned to the 100 and 020 plane reflections [18]. The peak at $2\theta=28^{o}$ is attributed to the presence of salts. After growing CNTs into the clay, the characteristic peaks of clay disappear indicating that the growth of CNT among clay layers can lead to the disruption of the ordered structure of clay. **Supplementary Figure 4** shows the XRD patterns of PEO electrolyte and its composites with clay and clay-CNT hybrid. The broadened characteristic peak of clay around $2\theta=6^{o}$ is observed in both PEO electrolyte composites with 5 and 10% clay due to the formation of intercalated structure of clay [19]. None of characteristic peaks of clay is observed in PEO electrolyte composite with various clay-CNT hybrid contents. The strong characteristic peaks of PEO appear between 15 and 20^{o}, which represent the existence of crystalline regions [20]. With increasing nanofiller content (clay and clay-CNT) in the polymer electrolyte, the crystalline peaks become broader and weaker, suggesting nanofillers suppress the crystallization of PEO.

FTIR spectra of pure PEO with Li salt and various nanocomposites are shown in **Supplementary Figures 5-10**. The impedance spectroscopy of pure PEO without salt (control sample) and PEO filled with 10% clay-CNT (without salt) are shown in **Supplementary Figures 11 and 12,** respectively, showing resistance above 10^{8}ohm.

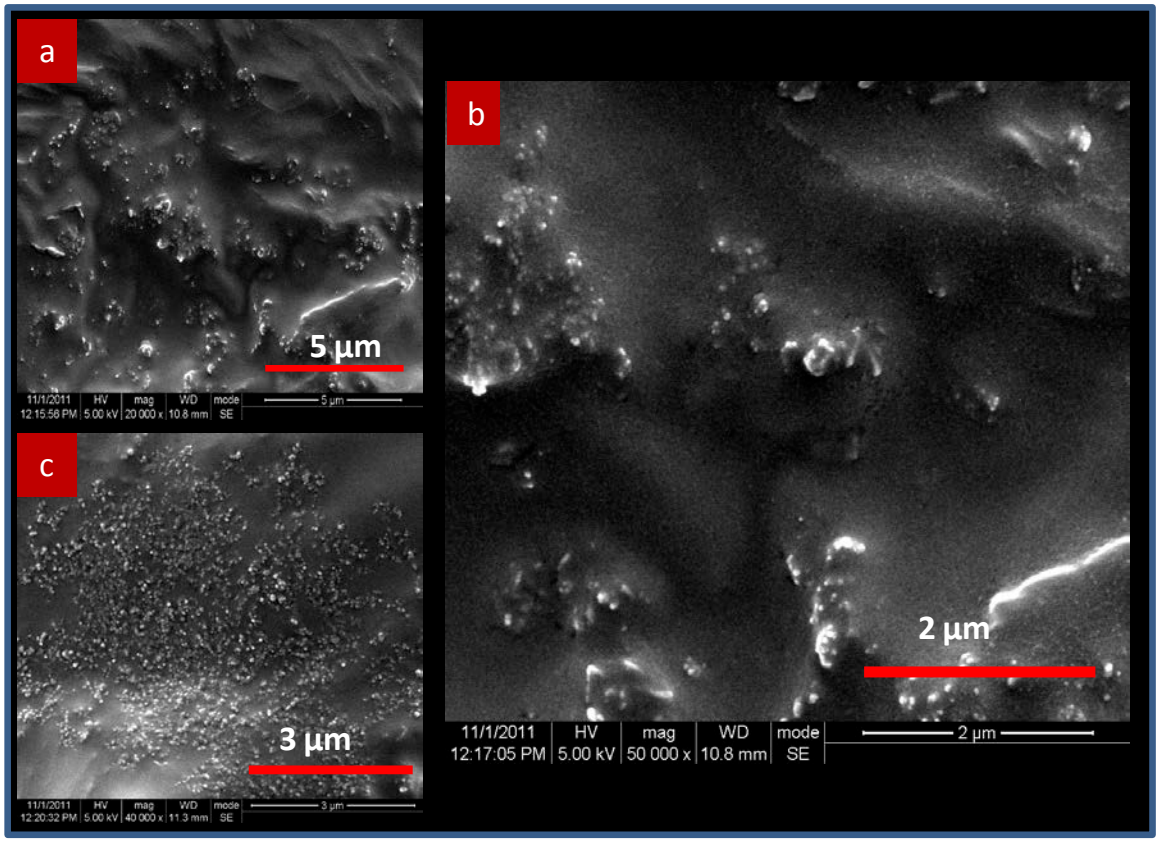
The molecular weights of the PEO (average 100,000 Mw) were measured before and after sonication using Waters 150-c GPC (Gel Permeation Chromatography) equipped with 410 differential refractometer at 25^{o}C. Deionized water was used as eluent with a

constant flow rate of 1 mL/min, and the instrument was calibrated using PEO standards. The sonication power is 25 W/L (100W, volume capacity is 4 L).

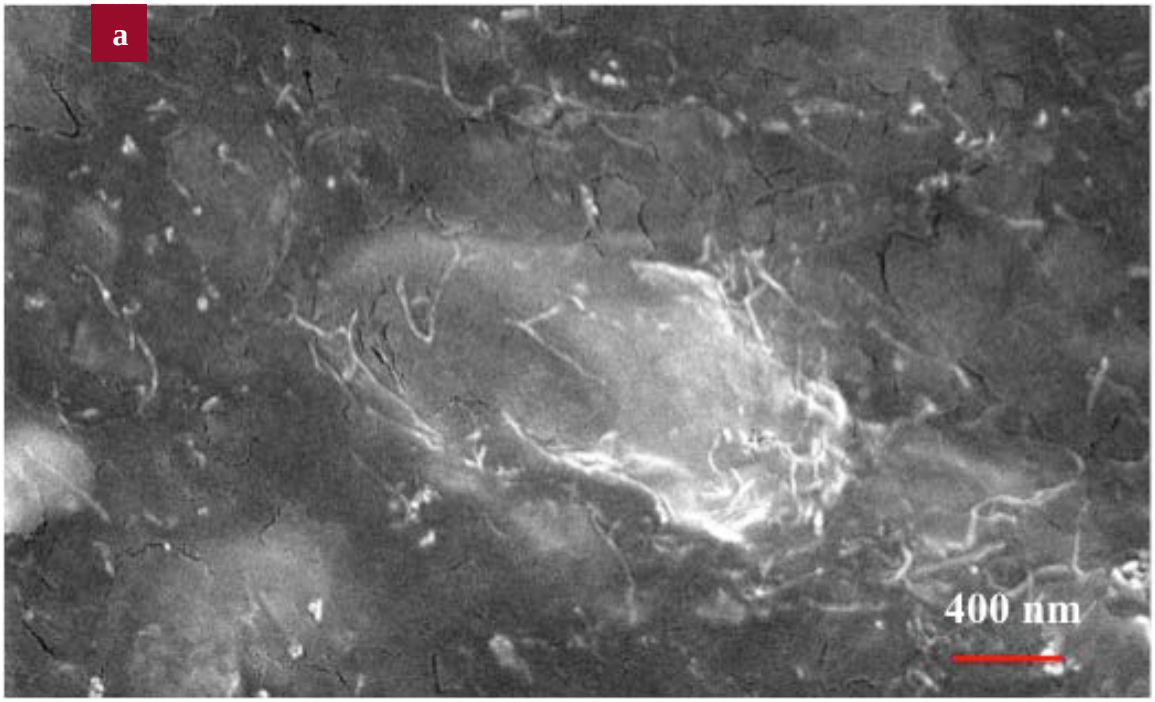
The GPC results are shown in **Supplementary Figure 13** and **Table 1.** The molecular weight of PEO appears to decrease slightly after sonication. Complex impedance tests on PEO before and after sonication reveal negligible "plasticizing effect" on ion conductivity of PEO.

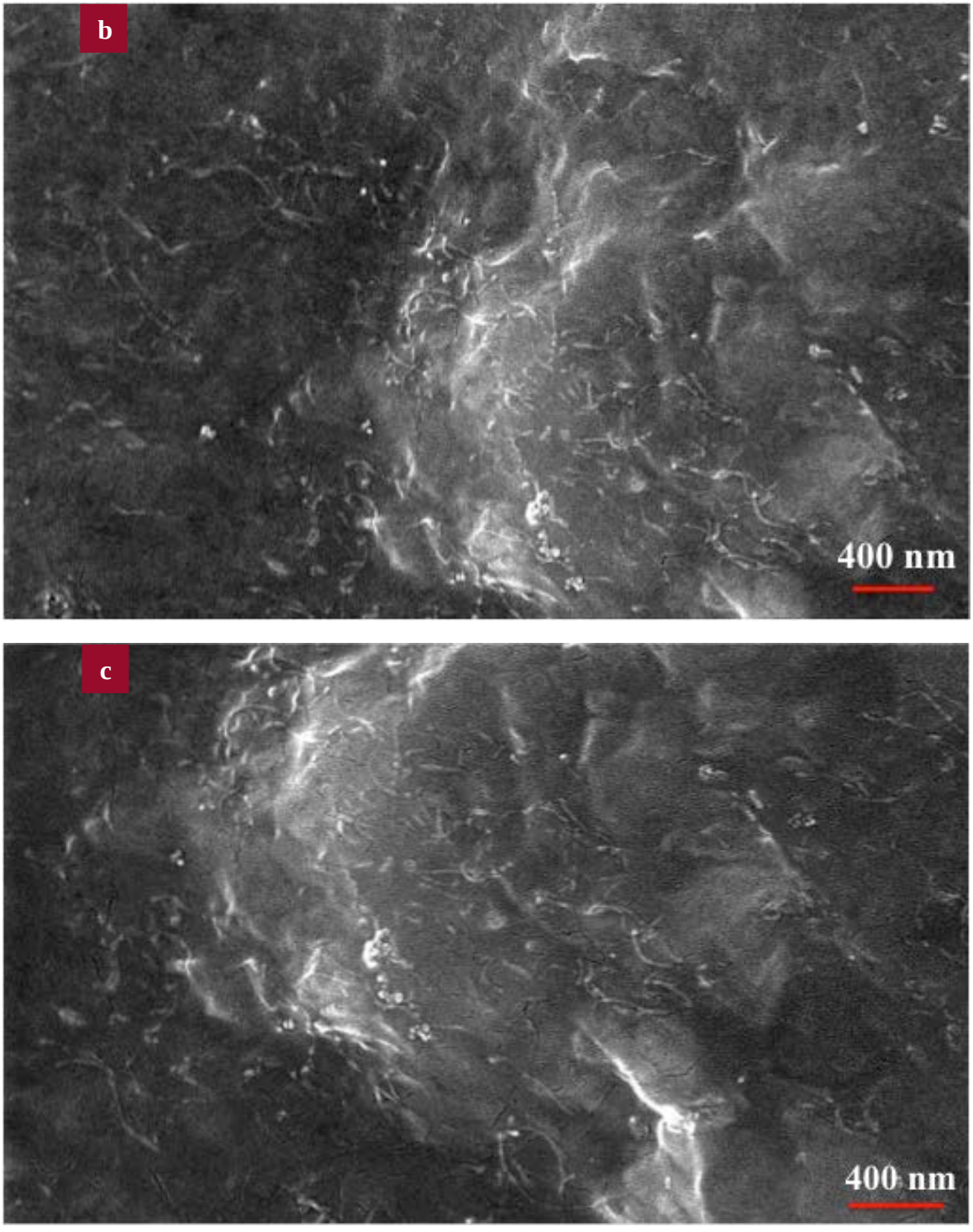
Supplementary Table 1. Molecular weights and the corresponding ion conductivities of PEO before and after sonication

Sample	Mw (Da)	Mn (Da)	PDI (Mw/Mn)	Ion Conductivity (S/cm)
Before sonication	104139	64412	1.62	5.58×10^{-7}
After sonication	87187	52522	1.66	6.87×10^{-7}

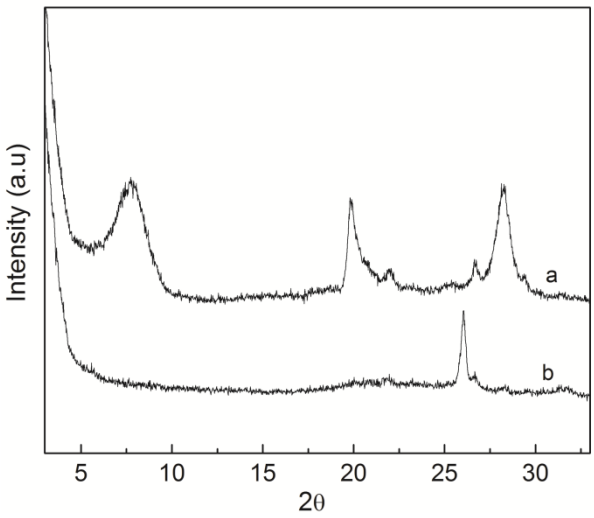


Supplementary Figure 1. SEM of PEO/salt with clay-CNT hybrid fillers (a)-(b) 5% and (c) 10% filled.

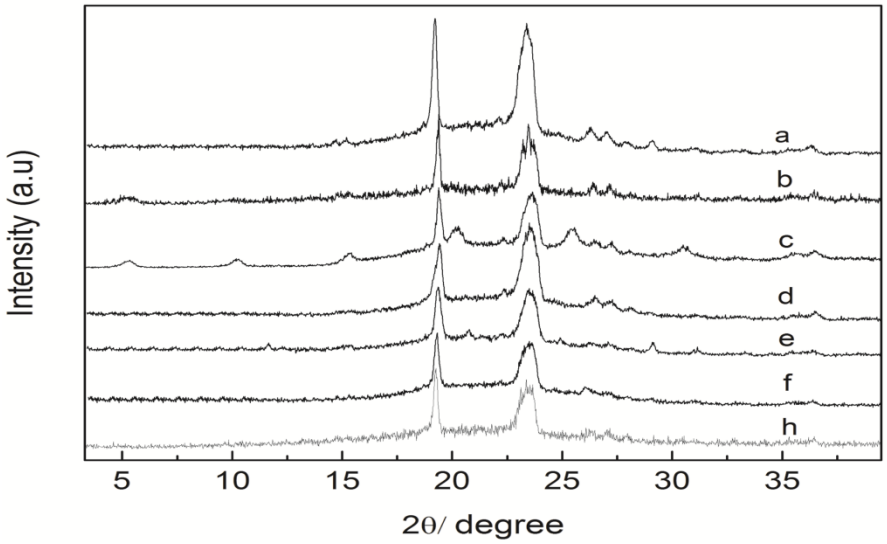




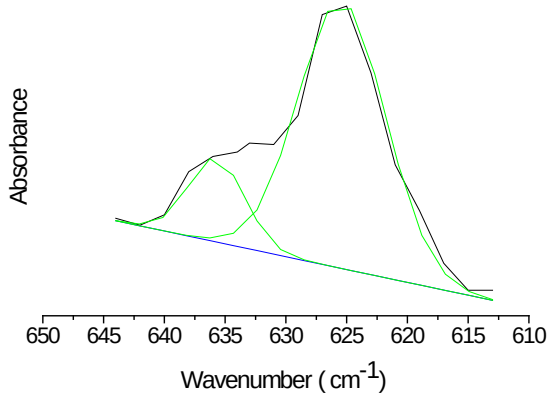
Supplementary Figure 2. SEM of UV etched PEO/salt with clay-CNT hybrid fillers (a) 5% and (b-c) 10%. The UV lamp profile: 500 W power and 254 nm wavelength.



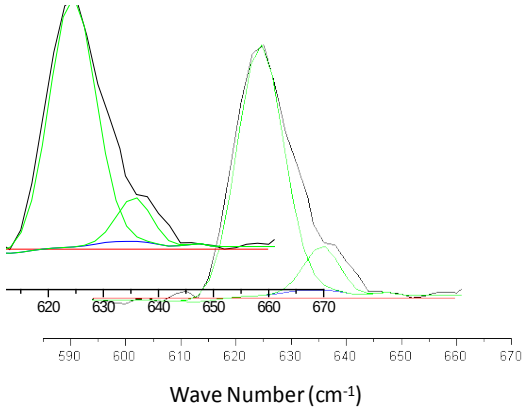
Supplementary Figure 3. XRD pattern of sodium montmorillonite clay (a) and clay-CNT hybrid (b)



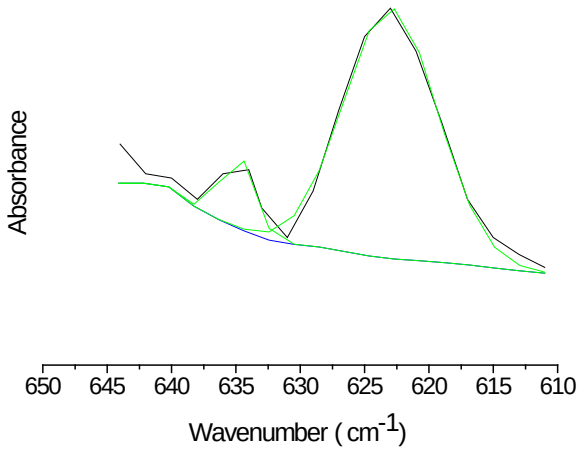
Supplementary Figure 4. XRD pattern of PEO/$LiClO_4$ electrolyte (a) and PEO/$LiClO_4$ nanocomposite electrolytes with 5% clay (b), 10% clay (c), and (d-f) 2, 5, 10,15% clay-CNT hybrids



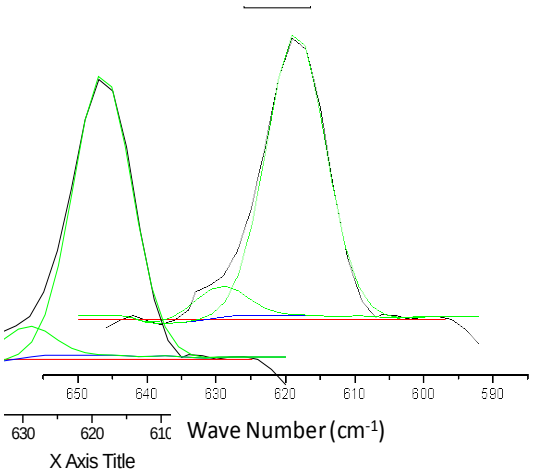
Supplementary Figure 5. FTIR of PEO/salt, peak area ratio: 623 (82.2%), 634 (17.8%)



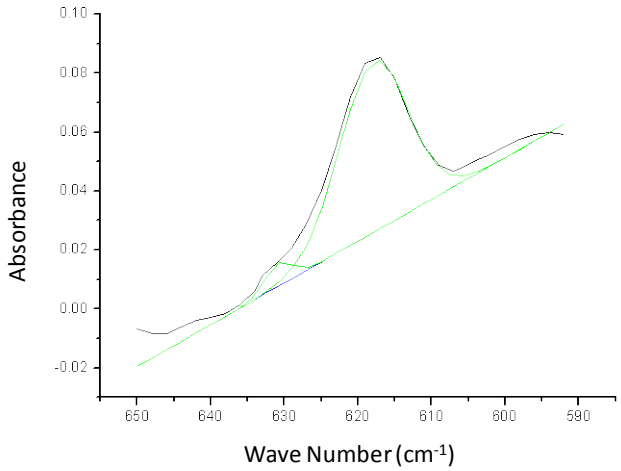
Supplementary Figure 6. FTIR of PEO/salt with 5% clay, 623 (88.4%), 635 (11.6%)



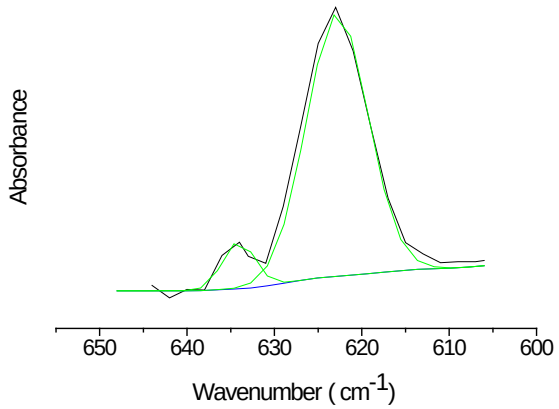
Supplementary Figure 7. FTIR of PEO/salt with 10% clay, 624 (90.6%), 635 (9.4%)



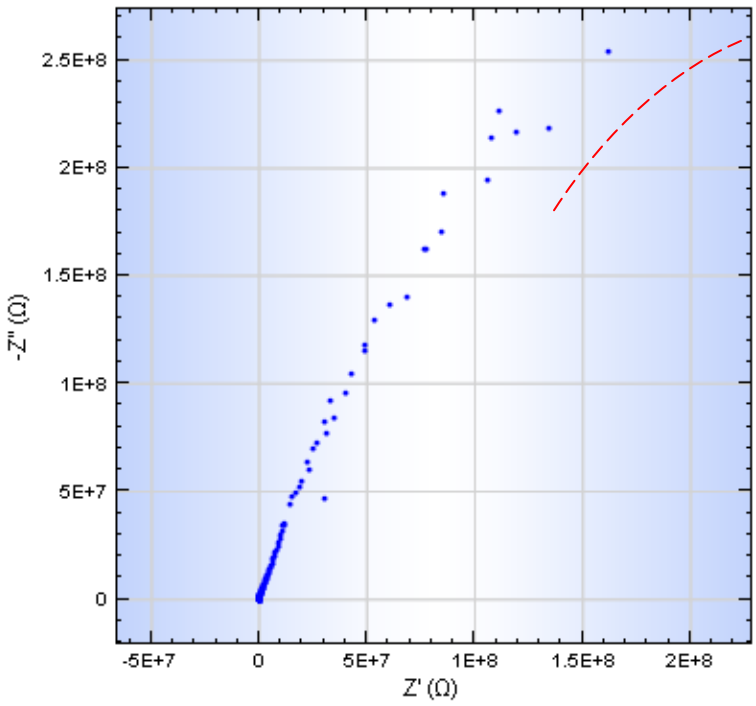
Supplementary Figure 8. FTIR of PEO/salt with 5% clay-CNT, 620 (91.03%), 630 (8.97%)



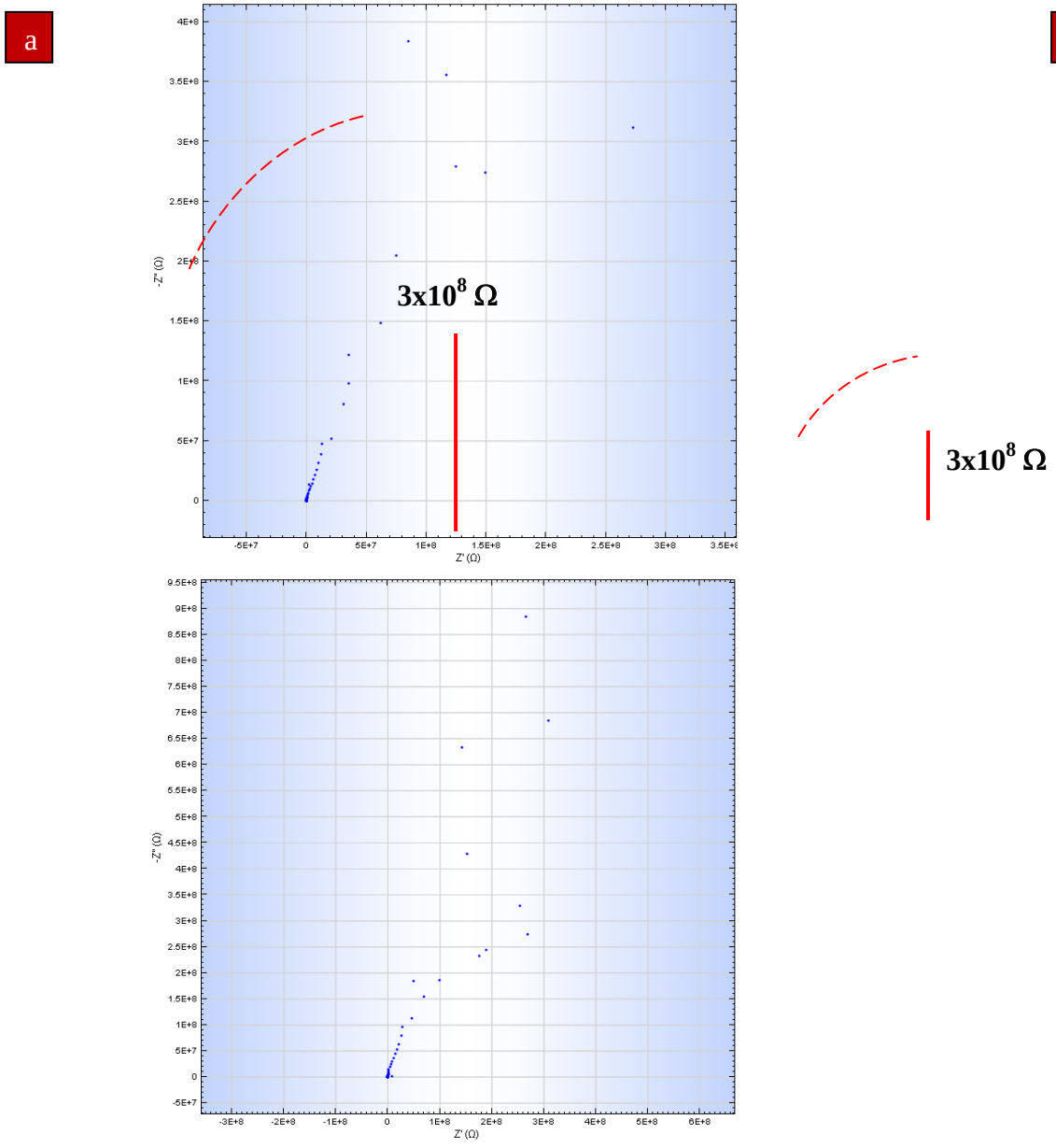
Supplementary Figure 9. FTIR of PEO/salt with 10% clay-CNT, 620 (95.7%), 630 (4.3%)



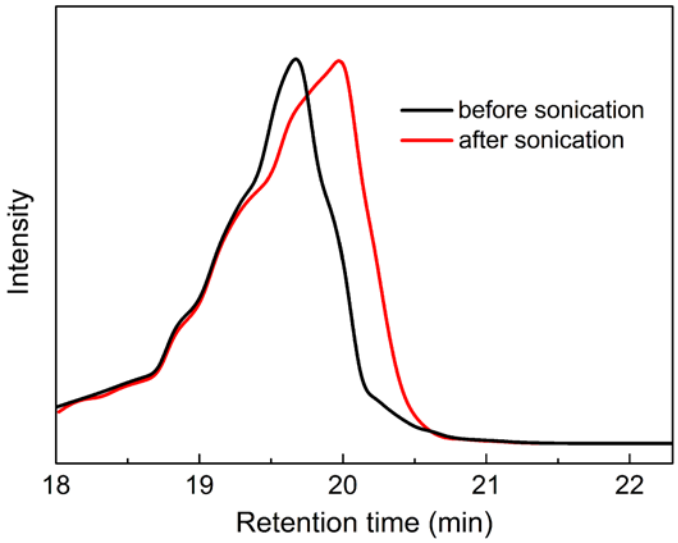
Supplementary Figure 10. FTIR of PEO with 15% clay-CNT, 623 (91.4%), 634 (8.6%)



Supplementary Figure 11. Impedance spectroscopy of pure PEO without salt (control sample)



Supplementary Figure 12. Impedance spectroscopy of PEO without Li salt and with (a) 5%, (b) 10% clay-CNT fillers



Supplementary Figure 13. GPC chromatogram of PEO solution before and after sonication