Supporting Information

Binder-Free, Thin-film Ceramic Coated Separators for Improved Safety of Lithium-Ion Batteries

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Table S1. Weights and volume of the separators used in this study

Separator used	Weight (g/m^2)	Thickness (µm)	Volume (cm^3)
PE	9.96	12	0.002712
EB-PVD coated Al_2O_3-PE	11.73	12.2	0.002716
commercial Al_2O_3-PE	15.27	20	0.004520
Al_2O_3-PI	19.25	22	0.004972

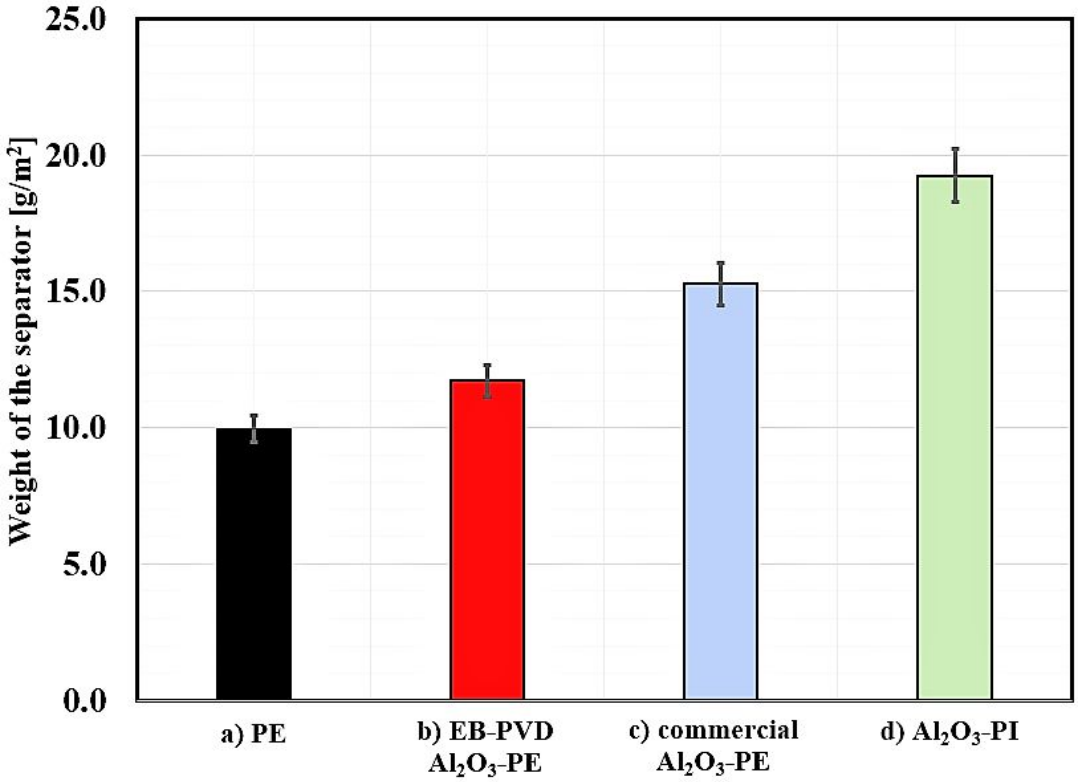
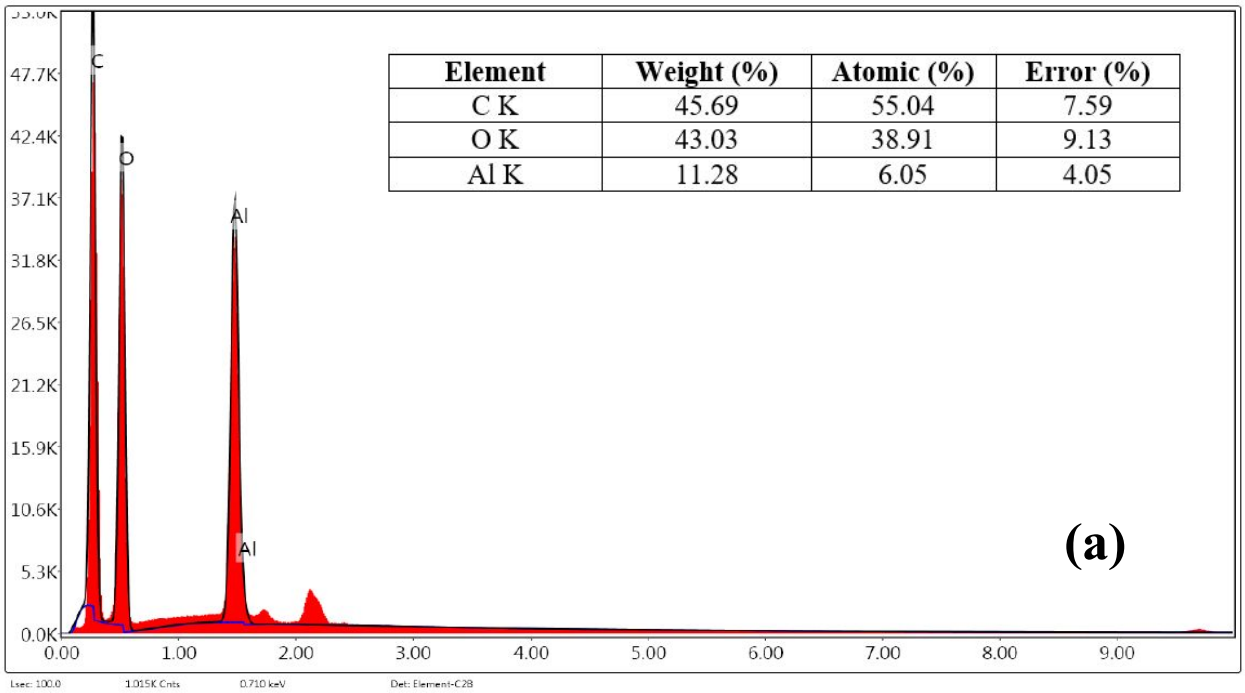
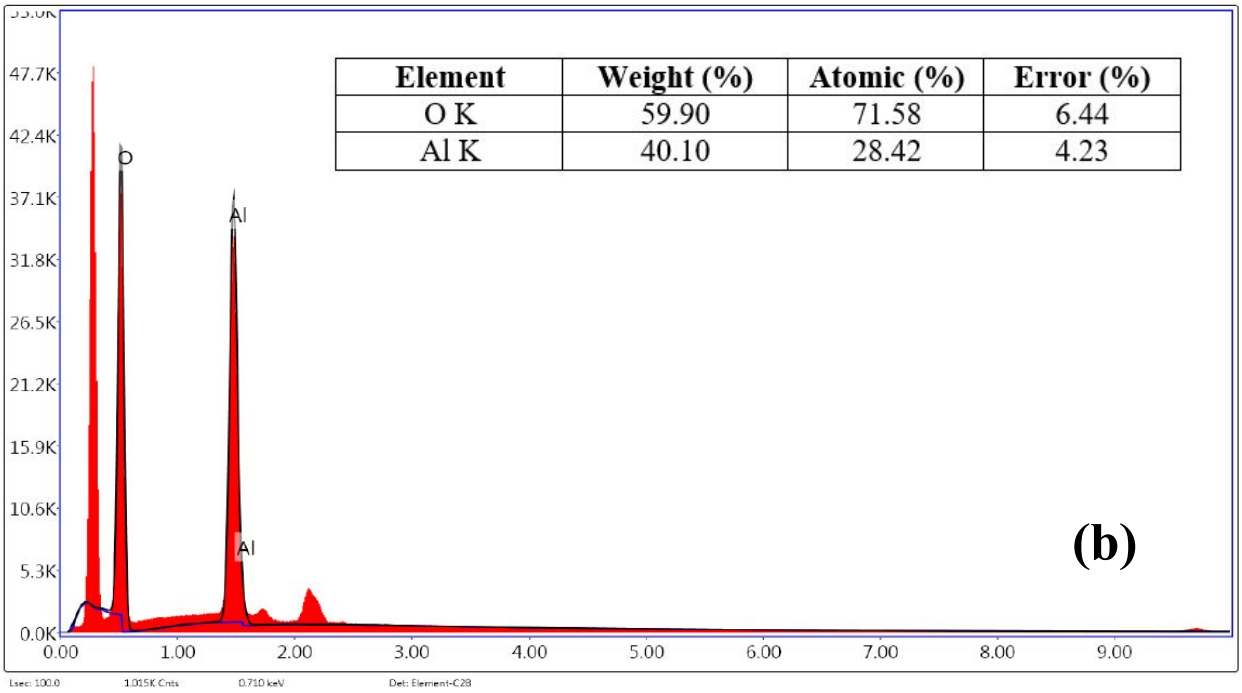


Figure S1. Weights of a) PE, b) EB-PVD coated Al_2O_3-PE (Al_2O_3, 100 nm), c) commercial Al_2O_3-PE (Al_2O_3, 1-2 µm), and d) Al_2O_3-PI separator, respectively



Element	Weight (%)	Atomic (%)	Error (%)
C K	45.69	55.04	7.59
O K	43.03	38.91	9.13
Al K	11.28	6.05	4.05



Element	Weight (%)	Atomic (%)	Error (%)
O K	59.90	71.58	6.44
Al K	40.10	28.42	4.23

Figure S2. Energy Dispersive Spectroscopy (EDS) elemental analysis of EB-PVD coated Al_2O_3-PE (Al_2O_3, 100 nm) separator a) with carbon (C) and, b) without C. Elemental C comes from the

polymer part of the separator. Aluminium and oxygen comes from the ceramic coating on the separator.

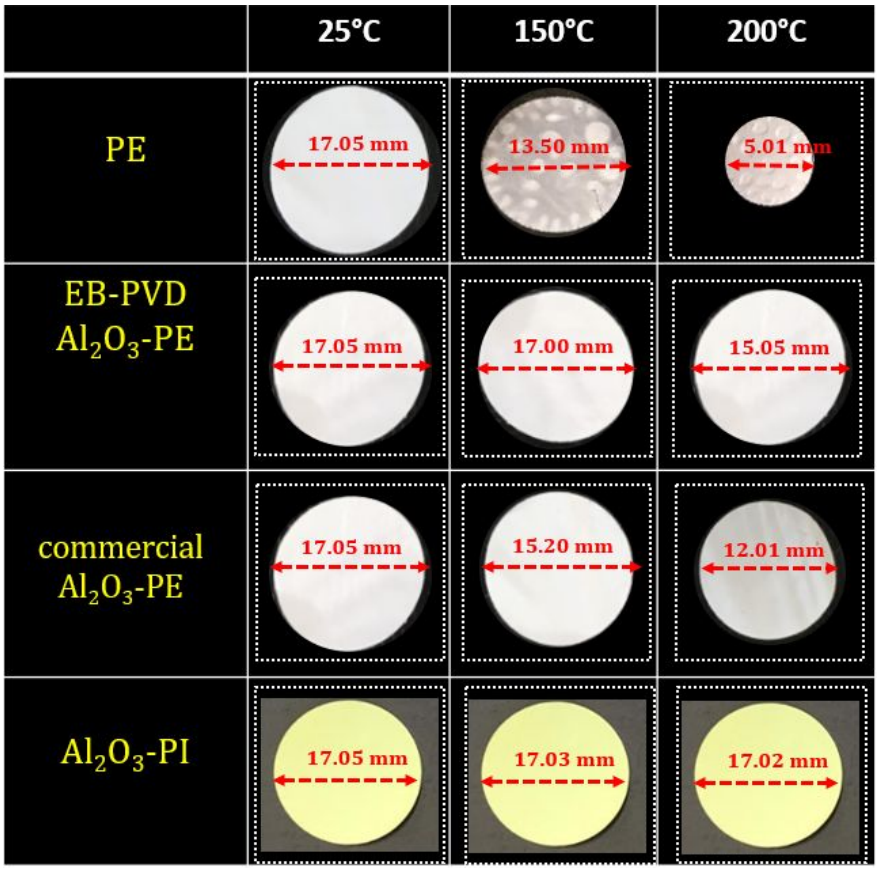


Figure S3. Thermal shrinkage measurement data comparing pristine PE, EB-PVD coated Al_2O_3-PE (Al_2O_3, 100 nm), commercial Al_2O_3-PE (Al_2O_3, 1-2 μm), and Al_2O_3-PI separator before and after annealing at 150 °C and 200 °C.

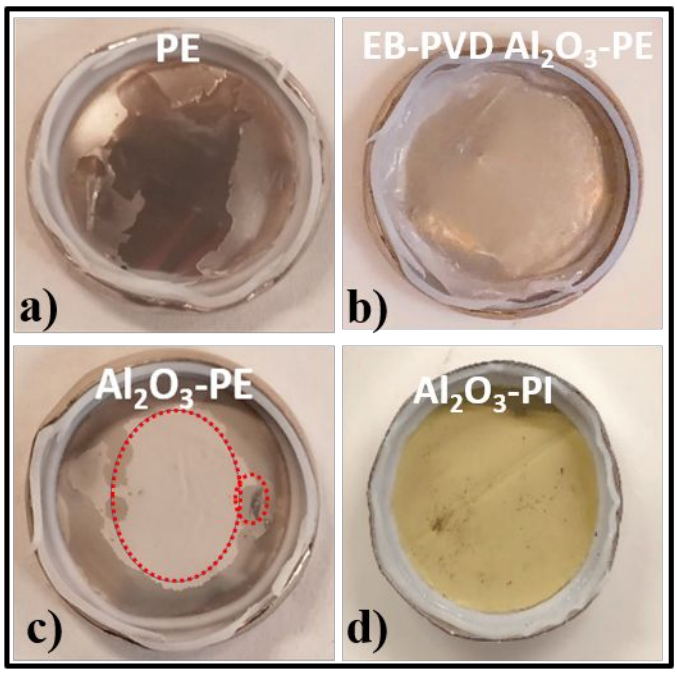


Figure S4. Postmortem optical images of a) PE, b) EB-PVD coated Al_2O_3-PE (Al_2O_3, 100 nm), c) commercial Al_2O_3-PE (Al_2O_3, 1-2 μm), d) Al_2O_3-PI separators after in-situ impedance measurement till 190 °C.

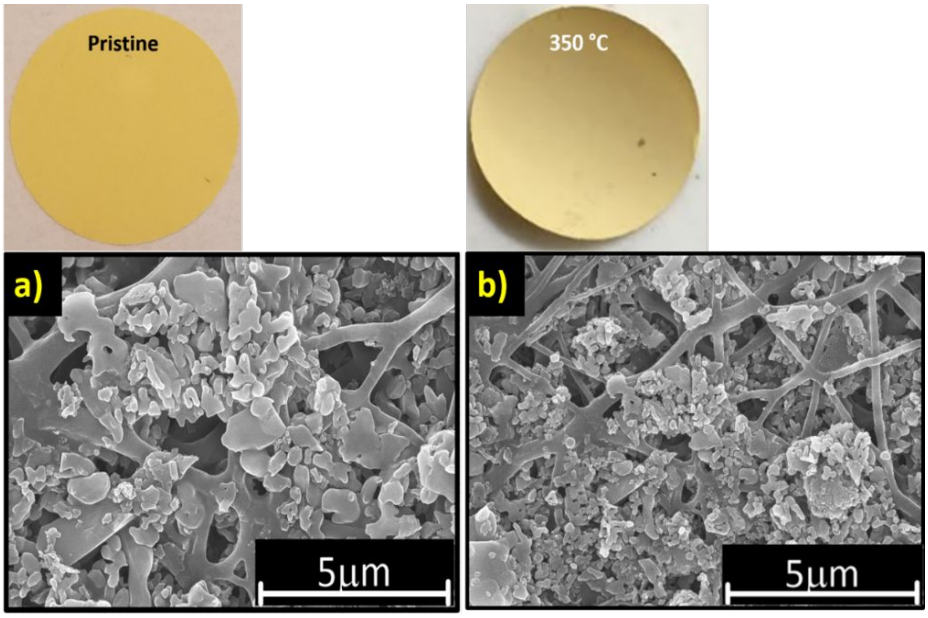


Figure S5. a) SEM morphology and optical images of Al_2O_3-PI separators a) Non-annealed, b) After annealing in argon for 1 h at 350 °C.

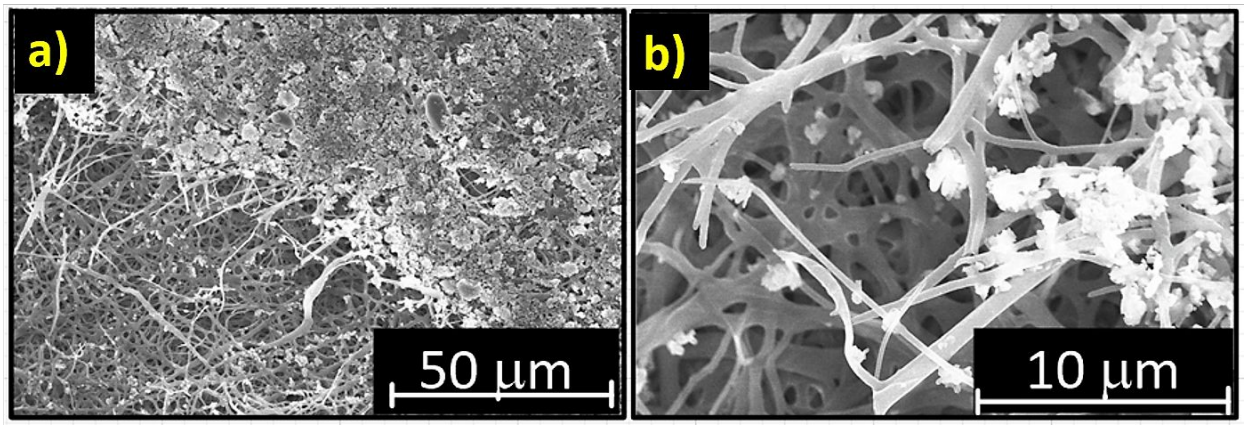


Figure S6. Morphology of the junction area (a) and tape side (b) after peeling test of the Al_2O_3-PI separator.

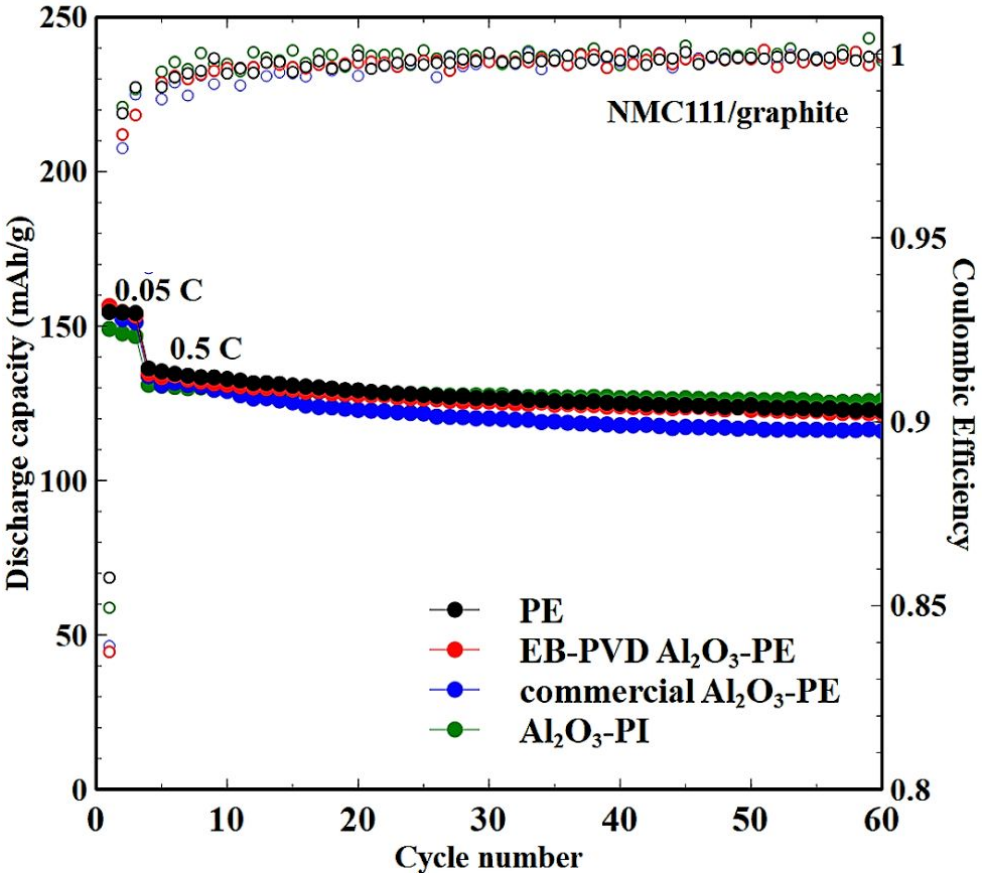


Figure S7. Electrochemical performance of NMC111/graphite full cells at C/20 (0.05C – Formation cycles) and C/2 (0.5C - long term cycles) using PE, EB-PVD coated Al_2O_3-PE (Al_2O_3,

100 nm), commercial Al_2O_3-PE (Al_2O_3, 1-2 µm), and Al_2O_3 coated PI separators measured at room temperature (22 °C).

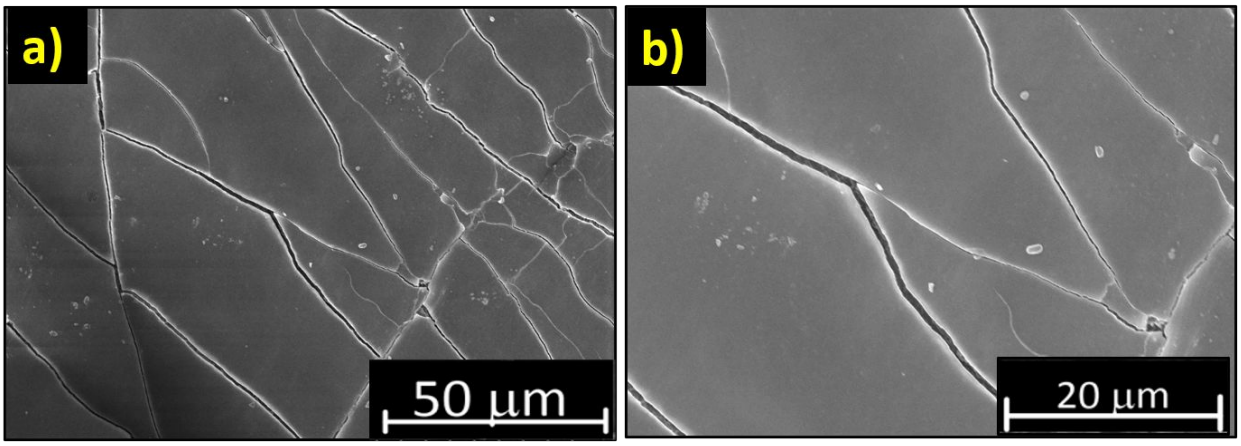


Figure S8. Separator degradation evaluation after cycling. SEM images of EB-PVD coated Al_2O_3-PE (Al_2O_3, 100 nm) separator cycled in NMC111/Graphite cells at magnifications a)1.00k, and b) 2.00k.

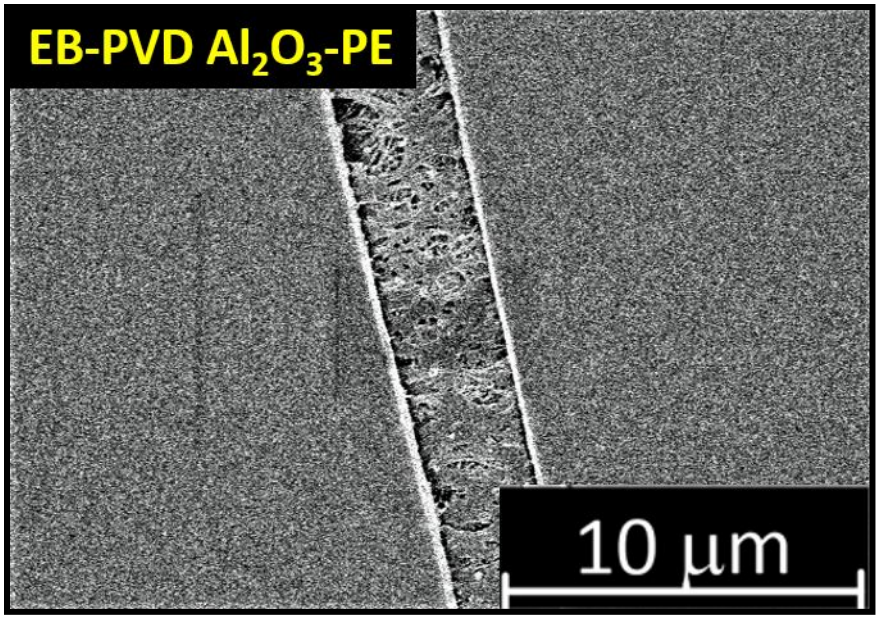


Figure S9. SEM images of EB-PVD coated Al_2O_3-PE (Al_2O_3, 100 nm) separator at a higher magnification. It has a dense ceramic coating. In addition to the cracks on the ceramic coating, a lot of pores are present in the polymer membrane that provides channels for Li^+ transport and electrolyte continuity.