## **Supporting Information**

Artificially-engineered, bicontinuous anion-conducting/-repelling polymeric phases as a selective ion transport channel for rechargeable zinc-air battery separator membranes

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**Figure S1.** Assembly diagram of a Zn-air cell suggested in this work, wherein the major cell components were integrated in the following order: Zn anode electrodeposited on a nickel plate, separator membrane, and air cathode.

**Figure S2.** SEM images (upper row) and chemical stability in 6M KOH electrolyte solution (lower row) of PVA/PAA nanofiber mat as a function of PVA/PAA composition ratio: (a) PVA/PAA = 5/5 (w/w); (b) 7/3; (c) 9/1.

**Figure S3.** Effect of annealing environment (air vs. Ar, at 120 °C) on chemical degradation of Nafion-impregnated PVA/PAA nanomat: (a) photographs; (b) TGA profiles.

**Figure S4.** (a) SEM images of microporous PP separator (Celgard3501). (b) A photograph showing mechanical flexibility of Celgard3501.

**Figure S5.** Tensile properties of PBE membrane and Celgard3501: (a) stress-strain curve; (b) summary of major tensile characteristics of the membranes.

**Figure S6.** Nyquist plots used for estimating ionic conductivity of: (a) Celgard3501; (b) Nafion film; (c) PVA/PAA film; (d) PBE membrane.

**Figure S7.** (a) A diffusion cell used for measurement of OH<sup>-</sup> transference number. (b) Comparison of OH<sup>-</sup> transference number between different membranes.

**Figure S8.** Photographs showing the crossover of blue dye through PBE membrane and Celgard3501.

**Figure S9.** Voltammogram profiles (scan rate = 5 mV s<sup>-1</sup>) of Celgard3501 and PBE membrane, revealing no appreciable decomposition of any components in the PBE membrane takes place within the operating voltage range (vs.  $Pt/Pt^{2+}$ ).

**Figure S10.** Video clips showing the operation of hypnosis-spiral wheels connected to Zn-air cells incorporating: (a) Celgard3501; (b) PBE membrane.

**Figure S11.** Electrochemical activity of Zn-air cells incorporating the reused separator membranes. (a),(b) For the reused Celgard3501 (obtained after the cycle time of 900 min in

the 1<sup>st</sup> cell): (a) galvanostatic charge/discharge profiles of the 2<sup>nd</sup> cell; (b) SEM and EDS images of the air cathode surface after the 2<sup>nd</sup> cycling test. (c),(d) For the reused PBE membrane (obtained after the cycle time of 2500 min in the 1<sup>st</sup> cell): (c) galvanostatic charge/discharge profiles of the 2<sup>nd</sup> cell; (d) SEM and EDS images of the air cathode surface after the 2<sup>nd</sup> cycling test.



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b

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b



**Figure S4.** (a) SEM images of microporous PP separator (Celgard3501). (b) A photograph showing mechanical flexibility of Celgard3501.



b	Tensile Strength / MPa	Elongation at Break / %	Tensile Modulus / MPa
Celgard3501	81.3	75.9	751.4
PBE membrane	39.4	46.2	973.1

**Figure S5.** Tensile properties of PBE membrane and Celgard3501: (a) stress-strain curve; (b) summary of major tensile characteristics of the membranes.



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