One-Pot Simultaneous ARGET ATRP Strategy on Widening Long-Range

Ion Channels to Facilitate Ion Conductivity for Alkaline Anion Exchange

Membrane Fuel Cell

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Fig. S1. ¹H NMR spectra of VBC-Dim.



Fig. S2. FTIR spectra of PPO, BrPPO and PPO-PImIL.



Fig. S3. ¹H NMR spectra of BrPPO.



Fig. S4. ¹H NMR spectra of PPO-PImIL.



Fig. S5 Semilogarithmic kinetic plots of ARGET ATRP under different BrPPO/Br@CNTs feeding ratios.



Fig. S6. TEM image of BrPPO/Br@CNTs-4wt%.

| Sample | VBC-Dim/BrPPO ^a (mole ratio%) | %PImIL content in grafted CNTs ^b | %Br reacted ^c | Graft density ^d | Graft length ^e | IEC (mmol g ⁻¹) | | | |
|---------------------|---------------------------------------------|---------------------------------------------|--------------------------|----------------------------|---------------------------|-----------------------------|------------------|--------------------|-------------------|
| | | | | | | PImIL@CNTs (exp) | PPO-PImIL (theo) | Hybrid AEMs (theo) | Hybrid AEMs (exp) |
| BrPPO/Br@CNTs-1 wt% | 55.6 | 49.8 | 32 | 5.8 | 9.6 | 2.01 | 2.08 | 2.32 | 2.18 |
| BrPPO/Br@CNTs-2 wt% | 54.8 | 48.2 | 31 | 5.6 | 9.8 | 1.94 | 2.06 | 2.27 | 2.24 |
| BrPPO/Br@CNTs-4 wt% | 55.2 | 47.3 | 29 | 5.2 | 10.6 | 1.91 | 2.07 | 2.31 | 2.29 |
| BrPPO/Br@CNTs-8 wt% | 53.3 | 45.9 | 28 | 5.0 | 10.7 | 1.85 | 2.04 | 2.13 | 1.88 |

Tab. S1. Chemical compositions and IECs of PPO-PImIL, PImIL@CNTs and hybrid AEMs under different BrPPO/Br@CNTs feeding ratios

^{*a*} Based on ¹H NMR. ^{*b*} Based on TGA. ^{*c*} Based on ¹H NMR. ^{*d*} Number of PImIL per 100 units in aromatic backbone, calculated from the mol% of -CH₂Br in BrPPO 18%) multiplied by the % of Br reacted. ^{*e*} Average number of VBC-Dim units in each graft chain, calculated from the VBC-Dim/BrPPO mole ratio divided by graft density.

The graft amounts of VBC-Dim are estimated from the integral ratio of the aromatic protons of BrPPO backbone (denoted 'a', 'm' and 'n' in Figure S4) to the aromatic

protons of ionic side chains (denoted 'e' and 'f' in Figure S4) by equation S1. Where E, F, A, M and N represent the integrals of 'e', 'f', 'a', 'm' and 'n' peaks, respectively.

$$\frac{\text{VBC-Dim}}{\text{BrPPO}} = \frac{E+F}{2(A+M+N)} \times 100\%$$
(S1)

The reacted Br sites (mol%) during ARGET ATRP are calculated from integrals of peak 'P' and 'l' by equation S2. Where P and L represent the integrals of 'p' and 'l' peaks,

respectively.

%Br reacted=
$$\frac{2P}{2P+L} \times 100\%$$
 (S2)

| Comula | Weight loss (%) | | | | | | |
|---------------------|-----------------|----------------|----------------|--|--|--|--|
| Sample | 1 M NaOH | 2 M NaOH | 4 M NaOH | | | | |
| ImPPO | 4.9±0.15 | 5.2±0.21 | 5.8 ± 0.22 | | | | |
| BrPPO/Br@CNTs-1 wt% | 4.2 ± 0.08 | 4.4 ± 0.10 | 4.9 ± 0.16 | | | | |
| BrPPO/Br@CNTs-2 wt% | 3.3 ± 0.11 | 3.6 ± 0.13 | 4.0 ± 0.09 | | | | |
| BrPPO/Br@CNTs-4 wt% | 1.8 ± 0.12 | 2.0 ± 0.14 | 2.3 ± 0.17 | | | | |
| BrPPO/Br@CNTs-8 wt% | 2.1 ± 0.18 | 2.3 ± 0.11 | 2.7 ± 0.24 | | | | |

Tab. S2. Alkaline stability of AEMs at 80 $^{\circ}\mathrm{C}$ after 24 h