## **Electronic Supporting Information**

for

## Thermal Stability and Swelling Behaviors of Nanoscale Ionic Materials (NIMs) with Tuned Structure

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- Figure S1. Schematic of the in-situ measurements of CO₂ capture capacity as well as thermally- and CO₂induced swelling of NIMs. (1) FT-IR spectrometer (Nicolet 6700, Thermo Fisher Scientific Inc.); (2) ATR optics and high pressure fluid cell (Golden Gate<sup>TM</sup>, Speca Ltd., UK); (3) Gas inlet; (4) Digital pressure gauge; (5) Gas cylinder; (6) Data acquisition system; (7) Temperature controller; (8) Vent; (9) Diamond crystal; (10) Mirrors; (11) Infrared light source.
- **Figure S2.** Thermal stability of Ionized PEGs with various molecular weights and the corresponding PEGs determined using a TGA, in oxygen environment with a ramping rate 5 °C/min.
- **Figure S3.** Thermal stability of NIMs with various chain lengths and the corresponding PEGs determined using a TGA, in oxygen environment with a ramping rate 5 °C/min.
- **Figure S4.** Thermal stability of NIMs with various chain lengths and the corresponding Ionized PEGs determined using a TGA, in oxygen environment with a ramping rate 5 °C/min.
- **Figure S5.** Thermal stabilities of NIMs with the same core fraction but different chain lengths determined using a TGA, in oxygen environment with a ramping rate 5 °C/min.

Figure S6. CO<sub>2</sub> capture capacity in NIMs with various chain lengths at 60 °C and  $P_{CO2} = 4 - 55$  atm.

Figure S7. Raman spectra in the  $v_a(CH_2)$  and  $v_s(CH_2)$  regions for PEGs and NIM-I-PEGs.



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**Figure S4.** Thermal stability of NIMs with various chain lengths and the corresponding Ionized PEGs determined using a TGA, in oxygen environment with a ramping rate 5 °C/min.



**Figure S5.** Thermal stabilities of NIMs with the same core fraction but different chain lengths determined using a TGA, in oxygen environment with a ramping rate 5 °C/min.



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