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Supplementary Information



Figure S1. <sup>1</sup>H NMR, in CDCl<sub>3</sub>, confirmed the structure and purity of the 1,4-butyl(bis-carbonylimidazolide) (1,4-BBCI).



Figure S2. <sup>1</sup>H NMR, in CDCl<sub>3</sub>, confirmed the structure and purity of the 1,4-cyclohexanedimethanol(bis-carbonylimidazolide) (1,4-CHDMBCI).



Figure S3.  $CO_2$  generation occurred rapidly once monomers were in the melt-state and increased exponentially upon gelation.



Scheme S1. Ethyl acetate facilitated high conversion of 1,4-butanediol, **b**, and 1,4-cyclohexanedimethanol, **a**, to 1,4-butyl(bis-carbonylimidazolide) (1,4-BBCI), **1b**, and 1,4-cyclohexanedimethyl(bis-carbonylimidazolide) (1,4-CHDMBCI), **1a**, respectively.



Figure S4. ATR-IR confirmed presence of urethane linkages and alkene moieties of PU foams.



**Figure S5**. TGA temperature ramp experiment with imidazole small molecule (dashed line) and BBCI-MDA:T-403 0:100 PU foam before (dotted line) and after (solid line) solvent extraction.



Figure S6. Select TMA temperature ramp of BBCI-TADE:T-403 90:10 provided CTE from slope and  $T_g$  from onset point.



Figure S7. DSC analysis indicated that the addition of MDA increased the  $T_g$  of T-403-based PU foams.



**Figure S8**. The incorporation of T-403 reduced the  $T_g$  for TADE-based PU foams according to DSC analysis.



Figure S9. Select DSC trace of BBCI-MDA:T-403 0:100 PU foam with an observed  $T_g$  of -3 °C.



**Figure S10**. Introduction of surfactant (0.8 wt % Dabco<sup>®</sup> DC193) into BBCI-MDA:T-403 0:100 produced more open cells than without surfactant.



**Figure S11**. Introduction of surfactant (0.8 wt % Dabco<sup>®</sup> DC193) into BBCI-TADE:T-403 100:0 produced a more homogeneous distribution of cells than without surfactant.