Supplementary Information:

Green synthesis of polyureas from CO$_2$ and diamines with a functional ionic liquid as the catalyst

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Hexyltributylphosphonium triazole (P$_{4,4,4,6}$Triz) and hexyltributylphosphonium aminotriazole (P$_{4,4,4,6}$ATriz) were prepared by the neutralization of hexyltributylphosphonium hydroxide (P$_{4,4,4,6}$OH) and 1,2,4-triazole or 3-amino-1,2,4-triazole according to literature methods.$^{26-27}$ Typically, a solution of P$_{4,4,4,6}$OH in ethanol was first prepared from P$_{4,4,4,6}$Br using the anion-exchange resin method. Second, equimolar 1,2,4-triazole or 3-amino-1,2,4-triazole was added to the P$_{4,4,4,6}$OH solution in ethanol. Next, the mixture was stirred at room temperature for 12 h. Subsequently, ethanol and water were removed by distillation at 60°C under reduced pressure. The obtained ILs were dried under high vacuum at 80°C for 8 h. The structures of these ILs were confirmed by NMR spectroscopy. $^1$H NMR and $^{13}$C NMR spectra were recorded on a Bruker AMX FT 400 MHz NMR spectrometer (Fig. S1).

P$_{4,4,4,6}$Triz $^1$H NMR (CDCl$_3$) 0.95 (m, 12H, CH$_3$), 1.30–1.52 (m, 20H, CH$_2$), 2.29–2.38 (m, 8H, PCH$_2$), 8.07 ppm (s, 2H, Triz C$_2$ and C$_5$);
$[^{13}\text{C}] \text{NMR (CDCl}_3\text{)}$ 13.3, 13.8, 18.5, 19.0, 21.7, 22.2, 23.7, 23.9, 30.3, 30.9, 148.8, 152.5 ppm.

$[^1\text{H}] \text{NMR (CDCl}_3\text{)}$ 0.94 (m, 12H, CH$_3$), 1.29–1.50 (m, 20H, CH$_2$), 2.21–2.29 (m, 8H, PCH$_2$), 7.34 (s, 2H, Triz-NH$_2$), 7.49 ppm (s, H, Triz C5);
P$_{4,4,4,6}$ATriz $^{13}$C NMR (CDCl$_3$) 13.4, 13.8, 18.5, 19.1, 21.7, 22.2, 23.6, 23.9, 30.4, 30.9, 148.6, 158.9 ppm.

**Fig. S1** NMR spectrum of the P$_{4,4,4,6}$Triz and P$_{4,4,4,6}$ATriz.

Fig. S2 CP/MAS $^{13}$C NMR spectrum of the solid products of diamines reaction with CO$_2$. 
Fig. S3 TGA traces of P$_{4,4,4,6}$Triz; (B) TGA traces of P$_{4,4,4,6}$ATriz.

Fig. S4 FT-IR spectra of P$_{4,4,4,6}$ATriz before and after use.

Fig. S5 Optimized structures of anion–2CO$_2$ complexes at the B3LYP/6-31+G (d, p) level.
Fig. S6 FT-IR spectra of P\textsubscript{4,4,4,6} Triz before and after absorption of CO\textsubscript{2}.