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Supplementary Information

Intermolecular interactions upon carbon dioxide capture in deep-eutectic solvents

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Spectroscopic characterization of deep eutectic solvents (DESs) and their precursors

MEA.Cl

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 3.0 (*t*, CH_2-NH_3^+), 3.69 (*t*, CH_2-OH); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 49.2 and 50.8.

HMIM.Cl

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 2.08, (*s*, N-H), 3.79 (*s*, CH_3-N), 7.29 (*d*, H-C5), 7.30 (*d*, H-C4) and 8.48 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 35.33, 119.9, 122.84 and 135.17.

[MEA.Cl][EDA] = 1:1

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 2.74 (*s*, CH_2 of EDA), 2.76 (*t*, CH_2-OH) and 3.57 (*t*, CH_2-NH_3^+); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 41.07, 41.96 and 61.37.

[MEA.Cl][EDA] = 1:1 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 2.75 (*s*, NH_2-CH_2), 2.80 (CH_2-NHCOO), 2.85 (*t*, CH_2-NHCOO), 3.05 (*t*, CH_2-NH_3^+) and 3.55 (*t*, CH_2-OH); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 38.92, 39.60, 40.18, 41.26, 58.09 and 164.35.

[MEA.Cl][EDA] = 1:2

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 2.68 (*s*, CH_2 of EDA), 2.71 (*t*, CH_2-OH) and 3.55 (*t*, CH_2-NH_3^+); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 41.77, 42.16 and 62.20.

[MEA.Cl][EDA] = 1:2 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^oC): δ_H (ppm): 2.71 (*s*, NH_2), 2.76 (CH_2-NHCOO), 2.80 (*t*, CH_2-NHCOO), 3.03 (*t*, CH_2-NH_3^+) and 3.52 (*t*, CH_2-OH); ^{13}C NMR (400 MHz, D_2O, 25 ^oC): δ_c (ppm): 39.25, 39.79, 40.18, 41.33, 58.59 and 164.35.

[MEA.Cl][EDA] = 1:3

^{1}H NMR (400 MHz, D_2O, 25 ^oC): δ_H (ppm): 2.65 (*s*, CH_2 of EDA), 2.69 (*t*, CH_2-OH) and 3.55 (*t*, CH_2-NH_3^+); ^{13}C NMR (400 MHz, D_2O, 25 ^oC): δ_c (ppm): 42.12, 42.24 and 62.52.

[MEA.Cl][EDA] = 1:3 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^oC): δ_H (ppm): 2.75 (*s*, NH_2-CH_2), 2.81 (CH_2-NHCOO), 2.85 (*t*, CH_2-NHCOO), 3.07 (*t*, CH_2-NH_3^+) and 3.56 (*t*, CH_2-OH);^{13}C NMR (400 MHz, D_2O, 25 ^oC): δ_c (ppm): 39.09, 39.72, 40.18, 41.29, 58.40 and 164.39.

[MEA.Cl][EDA] = 1:4

^{1}H NMR (400 MHz, D_2O, 25 ^oC): δ_H (ppm): 2.64 (*s*, CH_2 of EDA), 2.68 (*t*, CH_2-OH) and 3.54 (*t*, CH_2-NH_3^+); ^{13}C NMR (400 MHz, D_2O, 25 ^oC): δ_c (ppm): 42.26 and 62.65.

[MEA.Cl][EDA] = 1:4 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^oC): δ_H (ppm): 2.72 (*s*, NH_2), 2.78 (CH_2-NHCOO), 2.82 (*t*, CH_2-NHCOO), 3.04 (*t*, CH_2-NH_3^+) and 3.53 (*t*, CH_2-OH); ^{13}C NMR (400 MHz, D_2O, 25 ^oC): δ_c (ppm): 39.13, 39.73, 40.15, 41.28, 58.45 and 164.36.

[MEA.Cl][DETA] = 1:4

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 2.58 (*m*, CH_2-NH and CH_2-NH_3^+), 2.67 (*t*, CH_2-NH_2) and 3.53 (CH_2-OH); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 39.67, 42.24, 50.17 and 62.64.

[MEA.Cl][DETA] = 1:4 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 2.60 (*m*, CH_2-$NHCOO^-$), 2.71 (*t*, CH_2-NH), 2.97 (CH_2-NH_3^+) and 3.50 (CH_2-OH); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 38.90, 39.81, 47.85, 60.18 and 164.45.

[MEA.Cl][TEPA] = 1:4

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 2.38 (*m*, CH_2-NH_3^+), 2.48 (*t*, CH_2-NH- terminal), 2.56 (*t*, CH_2-NH- central), 2.61 (*t*, -CH_2-NH- terminal), 2.64 (*t*, CH_2-NH_2) and 3.51 (*t*, CH_2-OH); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 37.67, 39.68, 47.48, 50.30, 55.61 and 62.67.

[MEA.Cl][TEPA] = 1:4 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 2.35 (*m*, CH_2-NH_3^+), 2.50 (*t*, CH_2-NH- terminal), 2.59 (*t*, -CH_2-NH- terminal), 2.69 (*t*, CH_2-NH- central), 2.99 (*t*, CH_2-NHCOO) and 3.50 (*t*, CH_2-OH); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 37.32, 39.07, 41.82, 47.05, 51.73, 60.82 and 164.47.

[MEA.Cl][PEHA] = 1:4

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 2.41 (*m*, CH_2-NH_3^+), 2.56 (*t*, -CH_2-NH- central), 2.60 (*m*, -CH_2-NH- terminal) and 3.51 (*t*, CH_2-OH); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 37.66, 39.67, 42.19, 43.86, 45.43, 47.57, 50.30, 51.86, 52.97, 56.45, 59.38 and 62.68.

[MEA.Cl][PEHA] = 1:4 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^oC): δ_H (ppm): 2.33 (*m*, -CH_2-NH-CH_2-), 2.46 (*t*, CH_2-$NHCOO^-$), 2.56 (*m*, -NH-CH_2-CH_2-NH), 2.65 (*m*, -NH-CH_2-CH_2-NH) and 3.47 (*t*, CH_2-OH); ^{13}C NMR (400 MHz, D_2O, 25 ^oC): δ_c (ppm): 37.40, 39.15, 41.89, 43.72, 44.75, 47.24, 48.92, 51.82, 53.79, 56.21, 58.09, 61.09 and 164.40.

[HMIM.Cl][EDA] = 1:1

^{1}H NMR (400 MHz, D_2O, 25 ^oC): δ_H (ppm): 2.86 (*s*, CH_2-NH_2 of EDA), 3.58 (*s*, N-CH_3), 6.87 (*s*, H-C5), 6.98 (*s*, H-C4) and 7.49 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^oC): δ_c (ppm): 33.02, 39.76, 121.35, 127.36 and 138.47.

[HMIM.Cl][EDA] = 1:1 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^oC): δ_H (ppm): 2.90 (*t*, CH_2-NHCOO), 3.12 (-NH-CH_2), 3.449 (*s*, N-CH_3), 6.83 (*s*, H-C5), 6.91 (*s*, H-C4) and 7.52 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^oC): δ_c (ppm): 33.28, 38.85, 40.24, 121.46, 126.29, 137.88 and 164.41.

[HMIM.Cl][EDA] = 1:2

^{1}H NMR (400 MHz, D_2O, 25 ^oC): δ_H (ppm): 2.68 (s, CH_2-NH_2 of EDA), 3.57 (*s*, N-CH_3), 6.87 (*s*, H-C5), 6.98 (*s*, H-C4) and 7.48 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^oC): δ_c (ppm): 33.03, 41.50, 121.34, 127.52 and 138.45.

[HMIM.Cl][EDA] = 1:2 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^oC): δ_H (ppm): 2.75 (*t*, CH_2-NHCOO), 2.98 ((-NH-CH_2), 3.33 (*s*, N-CH_3), 6.67 (*s*, H-C5), 6.75 (*s*, H-C4) and 7.32 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^oC): δ_c (ppm): 33.04, 38.57, 38.82, 40.06, 121.25, 126.60, 137.91 and 164.23.

[HMIM.Cl][EDA] = 1:3

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 2.66 (*s*, CH_2-NH_2 of EDA), 3.60 (*s*, N-CH_3), 6.91 (*s*, H-C5), 7.02 (*s*, H-C4) and 7.52 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 33.09, 42.0, 121.37, 127.59 and 138.42.

[HMIM.Cl][EDA] = 1:3 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 2.62 (*s*, CH_2-NH), 2.67(*t*, CH_2-NHCOO), 2.93 (*t*, CH_2-NH), 3.28 (*s*, N-CH_3), 6.61 (*s*, H-C5), 6.70 (*s*, H-C4) and 7.22 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 32.86, 38.94, 39.54, 40.05, 121.12, 127.14, 138.10 and 164.18.

[HMIM.Cl][EDA] = 1:4

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 2.65 (*s*, CH_2-NH_2 of EDA), 3.61 (*s*, N-CH_3), 6.90 (*s*, H-C5), 7.01 (*s*, H-C4) and 7.51 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 33.03, 41.91, 121.35, 127.55 and 138.52.

[HMIM.Cl][EDA] = 1:4 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 2.79 (*s*, CH_2-NH), 2.86 (*t*, CH_2-NHCOO), 3.11 (*t*, CH_2-NH), 3.51 (*s*, N-CH_3), 6.81 (*s*, H-C5), 6.91 (*s*, H-C4) and 7.43 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 32.98, 38.99, 39.71, 40.23, 121.30, 127.31, 138.4 and 164.44.

[HMIM.Cl][DETA] = 1:4

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 2.59 (*t*, CH_2-NH-CH_2), 2.67 (*t*, CH_2-NH_2), 3.60 (*s*, N-CH_3^+), 6.89 (*s*, H-C5), 7.01 (*s*, H-C4) and 7.51 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 33.03, 39.56, 49.85, 121.35, 127.57 and 138.48.

[HMIM.Cl][DETA] = 1:4 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^oC): δ_H (ppm): 2.57 (*m*, CH_2-NHCOO), 2.68 (*t*, CH_2-NH-CH_2), 3.43 (*s*, N-CH_3^+), 6.73 (*s*, H-C5), 6.85 (*s*, H-C4) and 7.35 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^oC): δ_c (ppm): 32.95, 38.82, 47.45, 121.25, 127.38, 138.28 and 164.37.

[HMIM.Cl][TEPA] = 1:4

^{1}H NMR (400 MHz, D_2O, 25 ^oC): δ_H (ppm): 2.60 (*m*, CH_2-NH-CH_2 and CH_2-NH_2), 3.59 (*s*, N-CH_3^+), 6.87 (*s*, H-C5), 6.99 (*s*, H-C4) and 7.49 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^oC): δ_c (ppm): 33.02, 39.57, 47.42, 49.97, 55.08, 121.33, 127.57 and 138.44.

[HMIM.Cl][TEPA] = 1:4 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^oC): δ_H (ppm): 2.56 (*m*, CH_2-NH-CH_2 and CH_2-NHCOO), 3.49 (*s*, N-CH_3^+), 6.79 (*s*, H-C5), 6.91 (*s*, H-C4) and 7.41 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^oC): δ_c (ppm): 33.0, 39.06, 47.01, 48.65, 51.73, 121.28, 127.49, 138.30 and 164.38.

[HMIM.Cl][PEHA] = 1:4

^{1}H NMR (400 MHz, D_2O, 25 ^oC): δ_H (ppm): 2.59 (*m*, CH_2-NH-CH_2 and CH_2-NH_2), 3.58 (*s*, N-CH_3^+), 6.87 (*s*, H-C5), 6.99 (*s*, H-C4) and 7.49 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^oC): δ_c (ppm): 32.99, 33.02, 37.65, 39.61, 45.47, 47.54, 50.06, 51.88, 52.85, 56.44, 59.19, 121.32, 127.58 and 138.43.

[HMIM.Cl][PEHA] = 1:4 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^oC): δ_H (ppm): 2.54 (*m*, CH_2-NH-CH_2 and CH_2-NHCOO), 3.50 (*s*, N-CH_3^+), 6.79 (*s*, H-C5), 6.92 (*s*, H-C4) and 7.42 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^oC): δ_c

(ppm): 33.01, 37.44, 39.21, 43.75, 45.41, 47.27, 49.06, 51.84, 53.89, 56.24, 58.25, 121.28, 127.53, 138.27 and 164.35.

[MEA.Cl][AP] = 1:1

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 1.71 (*m*, -CH_2- of AP), 2.72 (*t,* CH_2-NH_3^+), 2.82 (*t,* CH_2-NH_2 of AP), 3.55 (*t*, CH_2-OH of AP) and 3.58 (*t*, CH_2-OH of MEA.Cl); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 31.15, 37.35, 42.02, 59.18 and 61.66.

[MEA.Cl][AP] = 1:1 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 1.41 (*m,* CH_2-$NHCOO^-$), 1.63 (*m*, -CH_2- of AP), 2.84 (*t,* CH_2-NH_3^+), 3.32 (*t*, CH_2-OH of AP), 3.44 (*t*, CH_2-OH of MEA.Cl); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 29.0, 37.14, 41.20, 58.78, 61.17, 161.32 and 164.50.

[MEA.Cl][AP] = 1:2

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 1.64 (*m*, -CH_2- of AP), 2.65 (*t,* CH_2-NH_3^+ of AP), 2.70 (*t,* CH_2-NH_2), 3.50 (*t*, CH_2-OH of AP) and 3.55 (*t*, CH_2-OH of MEA.Cl); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 32.54, 37.42, 42.24, 59.31 and 62.46.

[MEA.Cl][AP] = 1:2 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 1.38 (*m,* CH_2-$NHCOO^-$), 1.60 (*m*, -CH_2- of AP), 2.79 (*t,* CH_2-NH_3^+), 3.31 (*t*, CH_2-OH of AP), 3.41 (*t*, CH_2-OH of MEA.Cl); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 29.03, 31.97, 37.08, 58.73, 61.14, 161.57 and 164.52.

[MEA.Cl][AP] = 1:3

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 1.62 (*m*, -CH_2- of AP), 2.64 (*t,* CH_2-NH_3^+ of AP), 2.67 (*t,* CH_2-NH_2), 3.50 (*t*, CH_2-OH of AP) and 3.55 (*t*, CH_2-OH of MEA.Cl); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 33.03, 37.45, 42.29, 59.36 and 62.66.

[MEA.Cl][AP] = 1:3 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 1.46 (*m,* CH_2-$NHCOO^-$), 1.68 (*m*, -CH_2- of AP), 2.87 (*t,* CH_2-NH_3^+), 3.41 (*t*, CH_2-OH of AP), 3.49 (*t*, CH_2-OH of MEA.Cl); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 29.07, 32.0, 37.13, 58.79, 59.16 and 164.64.

[MEA.Cl][AP] = 1:4

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 1.62 (*m*, -CH_2- of AP), 2.64 (*t,* CH_2-NH_3^+ of AP), 2.66 (*t,* CH_2-NH_2), 3.51 (*t*, CH_2-OH of AP) and 3.56 (*t*, CH_2-OH of MEA.Cl); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 33.20, 37.43, 42.28, 59.40 and 62.76.

[MEA.Cl][AP] = 1:4 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 1.27 (*m,* CH_2-$NHCOO^-$), 1.48 (*m*, -CH_2- of AP), 2.67 (*t,* CH_2-NH_3^+), 3.21 (*t*, CH_2-OH of AP) and 3.29 (*t*, CH_2-OH of MEA.Cl); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 28.95, 31.91, 36.94, 58.59, 58.99 and 164.35.

[HMIM.Cl][AP] = 1:1

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 1.71 (*m*, -CH_2-), 2.90 (*t,* CH_2-NH_2), 3.48 (*s*, N-CH_3), 3.53 (*t*, CH_2-OH), 6.79 (*s*, H-C5), 6.88 (*s*, H-C4) and 7.40 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 29.36, 32.98, 37.29, 58.91, 121.27, 127.36 and 138.34.

[HMIM.Cl][AP] = 1:1 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^oC): δ_H (ppm): 1.65 (*m*, -CH_2-), 2.84 (*t,* CH_2-$NHCOO^-$), 3.37 (*s*, N-CH_3), 3.44 (*t*, CH_2-OH), 6.70 (*s*, H-C5), 6.79 (*s*, H-C4) and 7.33 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^oC): δ_c (ppm): 29.02, 33.0, 37.22, 58.79, 121.22, 127.0, 138.03 and 164.55.

[HMIM.Cl][AP] = 1:2

^{1}H NMR (400 MHz, D_2O, 25 ^oC): δ_H (ppm): 1.64 (*m*, -CH_2-), 2.73 (*t,* CH_2-NH_2), 3.52 (*s*, N-CH_3), 3.53 (*t*, CH_2-OH), 6.84 (*s*, H-C5), 6.94 (*s*, H-C4) and 7.44 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^oC): δ_c (ppm): 31.75, 32.98, 37.36, 59.21, 121.30, 127.49 and 138.43.

[HMIM.Cl][AP] = 1:2 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^oC): δ_H (ppm): 1.66 (*m*, -CH_2-), 2.85 (*m,* CH_2-$NHCOO^-$), 3.43 (*s*, N-CH_3), 3.47 (*t*, CH_2-OH), 6.74 (*s*, H-C5), 6.84 (*s*, H-C4) and 7.36 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^oC): δ_c (ppm): 29.07, 32.95, 37.17, 58.79, 121.24, 127.18, 138.22, 161.02 and164.61.

[HMIM.Cl][AP] = 1:3

^{1}H NMR (400 MHz, D_2O, 25 ^oC): δ_H (ppm): 1.61 (*m*, -CH_2-), 2.66 (*t,* CH_2-NH_2), 3.52 (*s*, N-CH_3), 3.55 (*t*, CH_2-OH), 6.84 (*s*, H-C5), 6.95 (*s*, H-C4) and 7.46 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^oC): δ_c (ppm): 32.61, 32.99, 37.40, 59.30, 121.30, 127.51 and 138.41.

[HMIM.Cl][AP] = 1:3 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^oC): δ_H (ppm): 1.59 (*m*, -CH_2-), 2.78 (*m,* CH_2-$NHCOO^-$), 3.36 (*s*, N-CH_3), 3.39 (*t*, CH_2-OH), 6.67 (*s*, H-C5), 6.78 (*s*, H-C4) and 7.30 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^oC): δ_c (ppm): 29.05, 32.0, 37.08, 58.72, 121.18, 127.12, 138.12, 161.08 and 164.51.

[HMIM.Cl][AP] = 1:4

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 1.59 (*m*, -CH_2-), 2.63 (*t,* CH_2-NH_2), 3.52 (*t*, CH_2-OH), 3.56 (*s*, N-CH_3), 6.85 (*s*, H-C5), 6.96 (*s*, H-C4) and 7.47 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 29.03, 33.03, 37.43, 59.34, 121.30, 127.53 and 138.41.

[HMIM.Cl][AP] = 1:4 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 1.54 (*m*, -CH_2-), 2.73 (*m,* CH_2-$NHCOO^-$), 3.31 (*s*, N-CH_3), 3.34 (*t*, CH_2-OH), 6.63 (*s*, H-C5), 6.73 (*s*, H-C4) and 7.25 (*s*, H-C2); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 29.02, 31.97, 37.01, 58.66, 121.14, 127.07, 138.05, 161.07 and 164.43.

[TBAB][AP] = 1:2

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 0.92 (*t*, CH_3), 1.38 (*m*, CH_2 of TBAB), 1.60 (*m*, CH_2 and CH_2-NH_2), 2.42 (*t*, CH_2-OH), 2.85 (*t*, CH_2 of AP), 3.29 (*t*, CH_2 of TBAB) and 3.69 (*t*, CH_2 of AP); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 13.64, 19.69, 24.10, 34.10, 40.95, 58.95 and 62.48.

[TBAB][AP] = 1:2 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 0.83 (*t*, CH_3), 1.25 (*m*, CH_2 of TBAB), 1.55 (*m*, CH_2 and CH_2-NHCOO), 2.91 (*t*, CH_2 of AP), 3.07 (*m*, CH_2 of TBAB), 3.50 (*t*, CH_2 of AP) and 3.57 (*t*, CH_2 of AP); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 12.91, 19.15, 23.13, 32.12, 37.27, 58.08, 58.97 and 164.71.

[TBAB][AP] = 1:3

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 0.86 (*t*, CH_3), 1.29 (*m*, CH_2 of TBAB), 1.59 (*m*, CH_2 and CH_2-NH_2), 2.59 (*t*, CH_2 of AP), 3.11 (*m*, CH_2 of TBAB) and 3.56 (*t*, CH_2-OH); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 12.86, 19.15, 23.14, 34.10, 37.43, 58.11 and 59.55.

[TBAB][AP] = 1:3 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 0.80 (*t*, CH_3), 1.22 (*m*, CH_2 of TBAB), 1.52 (*m*, CH_2 and CH_2-NHCOO), 2.92 (*t*, CH_2 of AP), 3.04 (*m*, CH_2 of TBAB) and 3.55 (*t*, CH_2-OH); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 12.87, 19.11, 23.09, 29.29, 32.08, 37.20, 58.87, 161.34 and 164.68.

[TBAB][AP] = 1:4

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 0.86 (*t*, CH_3), 1.28 (*m*, CH_2 of TBAB), 1.59 (*m*, CH_2 of TBAB), 2.58 (*t*, CH_2-NH_2 of AP), 3.11 (*m*, CH_2 of AP) and 3.56 (*t*, CH_2-OH); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 12.83, 19.14, 23.12, 34.08, 37.42, 58.10 and 59.55.

[TBAB][AP] = 1:4 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 0.75 (*t*, CH_3), 1.17 (*m*, CH_2 of TBAB), 1.47 (*m*, CH_2 of TBAB), 1.69 (*m*, CH_2 of TBAB), 2.87 (*t*, CH_2-NHCOO), 2.99 (*m*, CH_2 of AP) and 3.50 (*t*, CH_2-OH); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 12.89, 19.08, 23.06, 29.20, 32.08, 37.15, 58.81, 161.11 and 164.59.

[TBAB][AMP] = 1:3

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 0.98 (*t*, CH_3 of TBAB), 1.06 (*s*, CH_3 of AMP), 1.44 (*m*, CH_2 of TBAB), 1.65 (*m*, CH_2 of TBAB), 3.26 (*s*, CH_2 of AMP) and 3.34 (*m*, CH_2 of TBAB); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 13.69, 19.74, 24.17, 27.14, 50.59, 59.04 and 71.61.

[TBAB][AMP] = 1:3 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 0.84 (*t*, CH_3 of TBAB), 1.07 (*s*, CH_3 of AMP), 1.26 (*m*, CH_2 of TBAB), 1.54 (*m*, CH_2 of TBAB), 3.09 (*m*, CH_2 of TBAB) and 3.33 (*s*, CH_2-OH of AMP); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 12.80, 23.21, 52.37, 58.10, 68.83 and 163.15.

[TBAB][AMP] = 1:4

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 0.87 (*t*, CH_3 of TBAB), 0.96 (*s*, CH_3 of AMP), 1.29 (*m*, CH_2 of TBAB), 1.57 (*m*, CH_2 of TBAB), 3.12 (*m*, CH_2 of TBAB) and 3.24 (*s*, CH_2-OH of AMP); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 12.93, 19.18, 23.16, 25.12, 49.77, 58.12 and 71.27.

[TBAB][AMP] = 1:4 after CO_2 absorption

^{1}H NMR (400 MHz, D_2O, 25 ^{o}C): δ_H (ppm): 0.81 (*t*, CH_3 of TBAB), 1.07 (*s*, CH_3 of AMP), 1.23 (*m*, CH_2 of TBAB), 1.51 (*m*, CH_2 of TBAB), 3.06 (*m*, CH_2 of TBAB) and 3.33 (*s*, CH_2-OH of AMP); ^{13}C NMR (400 MHz, D_2O, 25 ^{o}C): δ_c (ppm): 12.82, 19.10, 22.78, 30.23, 53.03, 58.05, 68.20 and162.80.

Speciation of absorbed CO_2

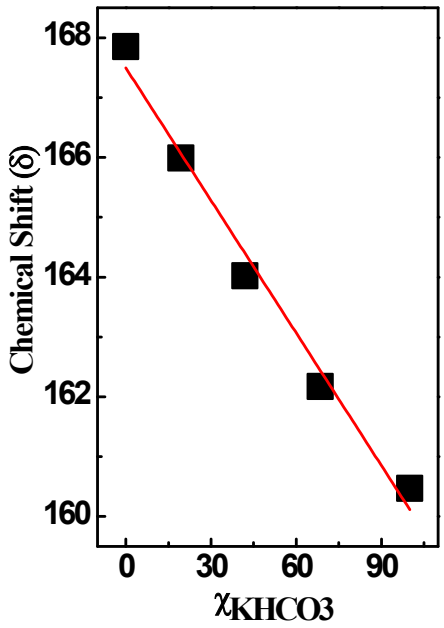

Fig. S1 Linear relation between chemical shift (δ) and mole fraction of $KHCO_3$.

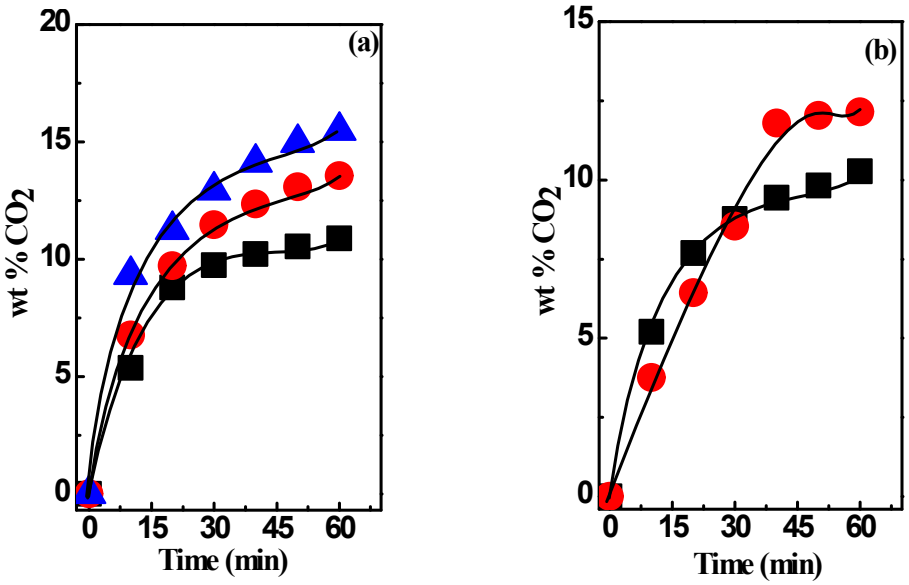

Figure. S2 (a) CO_2 absorption kinetics in TBAB AP- and (b) TBAB AMP-based DESs at 1:2 (■), 1:3 (●) and 1:4 (▲) mole ratios.

Dependence of CO2 uptake on viscosity

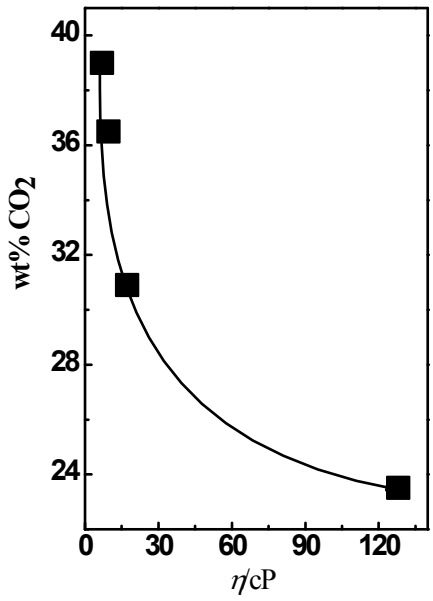

Fig. S3 Dependence of CO_2 wt% on viscosity (η) in [MEA.Cl][EDA] at 1:4.

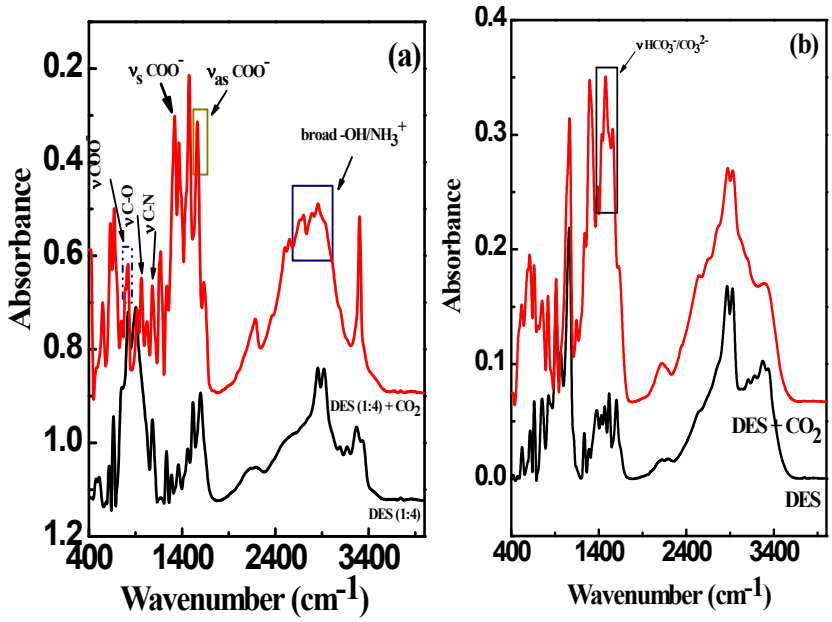

Fig. S4 FTIR-spectra of (a) [MEA.Cl][EDA] and [HMIM.Cl][EDA] at (1:4)**.**

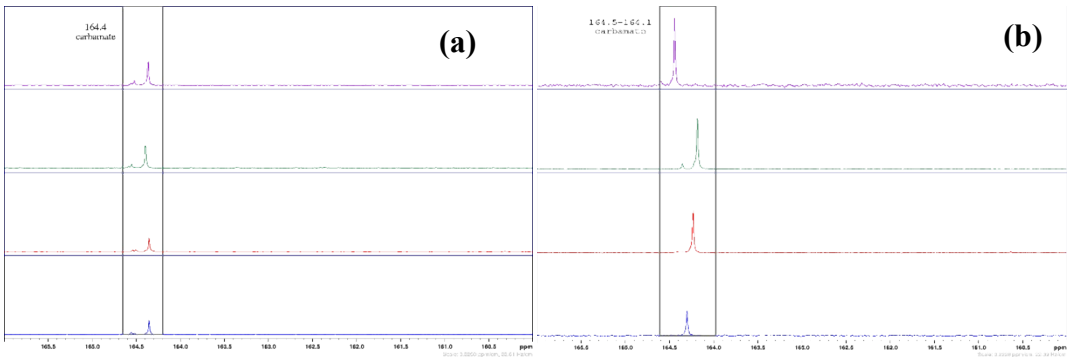

Fig. S5 ^{13}C NMR spectrum of (a) [MEA.Cl][EDA]- and (b) [HMIM.Cl][EDA]-class of DESs at 1:1 (-), 1:2 (-), 1:3 (-) and 1:4 (-) mole ratios.

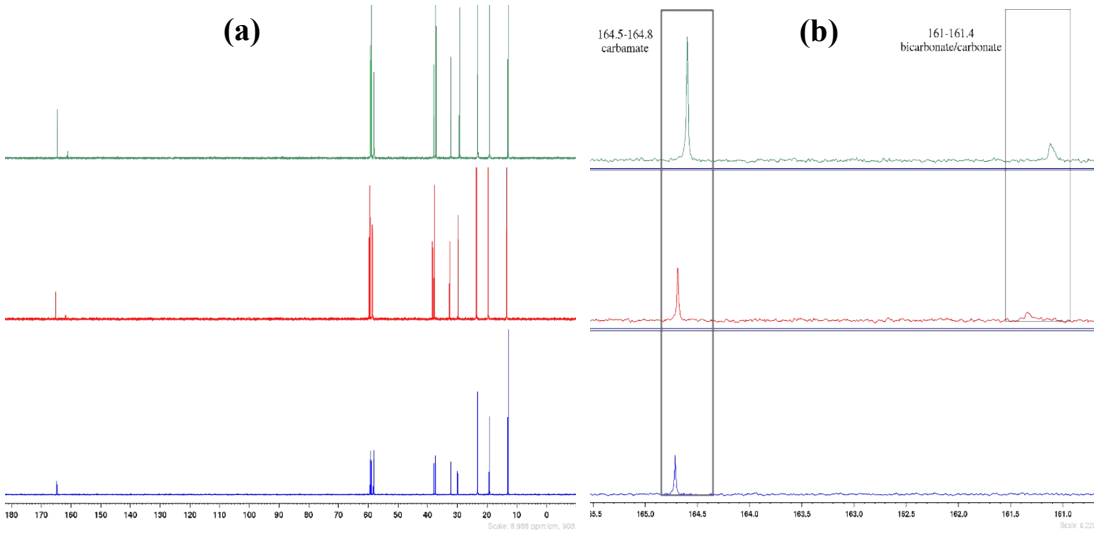

Fig. S6 ^{13}C NMR spectrum of [TBAB][AP]-based DESs at 1:2 (-), 1:3 (-) and 1:3 (-) mole ratios. Full range spectra is shown in (a) while the presence of carbamate and carbonate/bicarbonate is shown in (b).

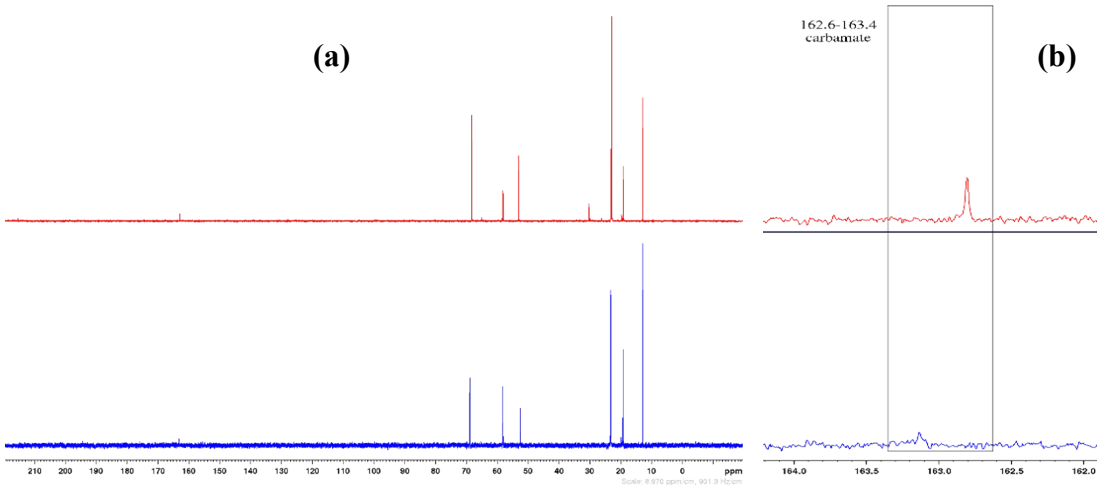

Fig. S7 ^{13}C NMR spectrum of [TBAB][AMP]-based DESs at 1:3 (-) and 1:4 (-) mole ratios. Full range spectra is shown in (a) while the presence of carbamate is shown in (b).

Recyclability Experiments

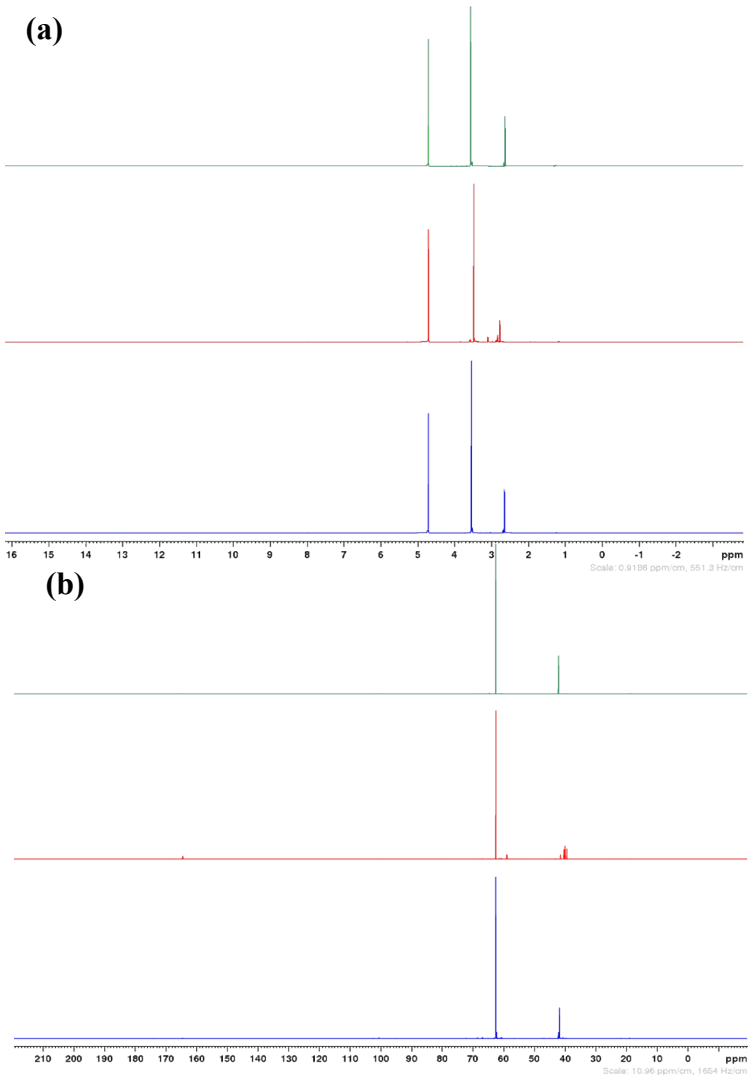

Fig. S8 ^{1}H (a) and ^{13}C (b) NMR spectra of 30 wt% [MEA.Cl][EDA]=1:3 + ethylene glycol before CO_2 absorption (–), after CO2 absorption in the 3rd cycle (–), and after CO_2 desorption in the 3rd cycle (–).