Supplementary Materials

Aqueous 2-Ethyl-4-methylimidazole Solution for Efficient CO_2 Separation and Purification

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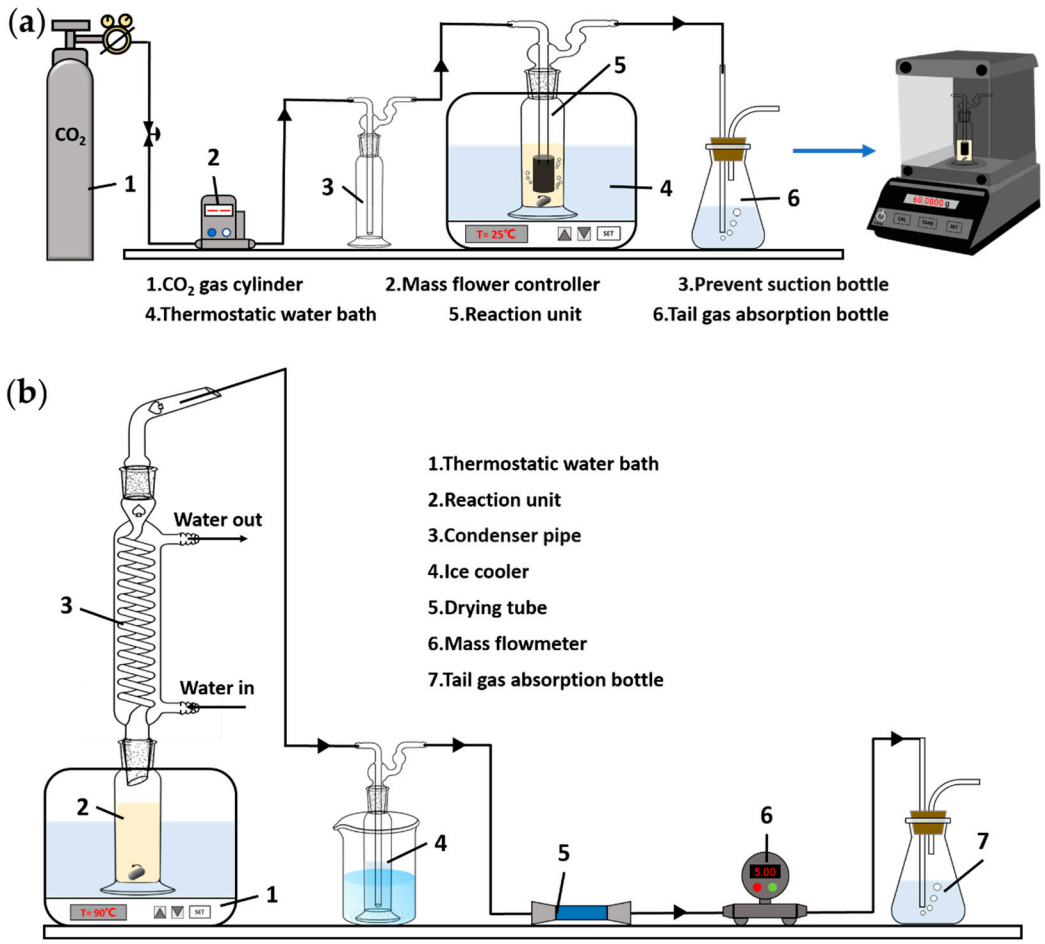


Figure S1. Schematic of experimental setups. (**a**) CO_2 absorption setup. (**b**) CO_2 desorption setup.

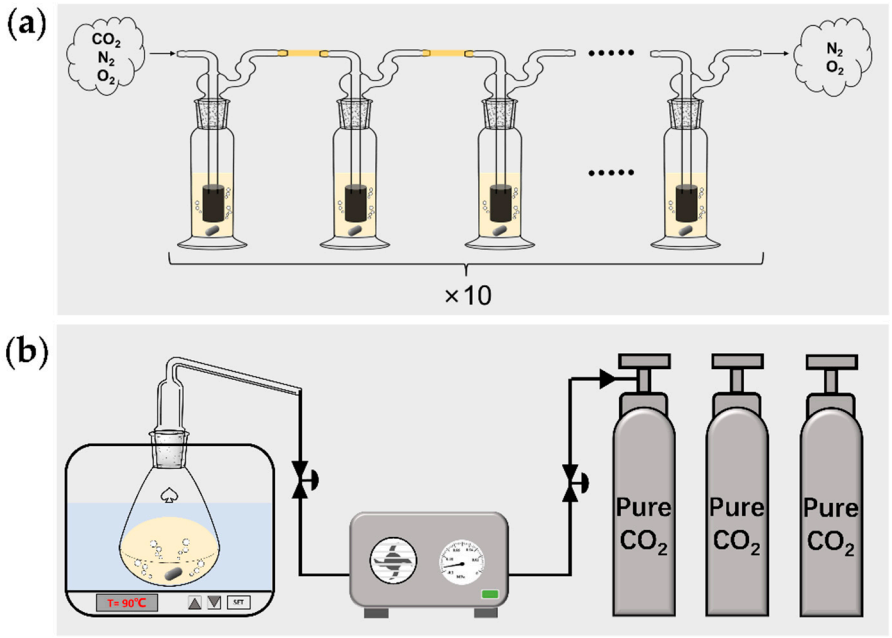


Figure S2. (**a**) Schematic of the bubbling absorption setup, which consists of 10 bubbling reactors connected in series. (**b**) Schematic of desorption and compression setup.

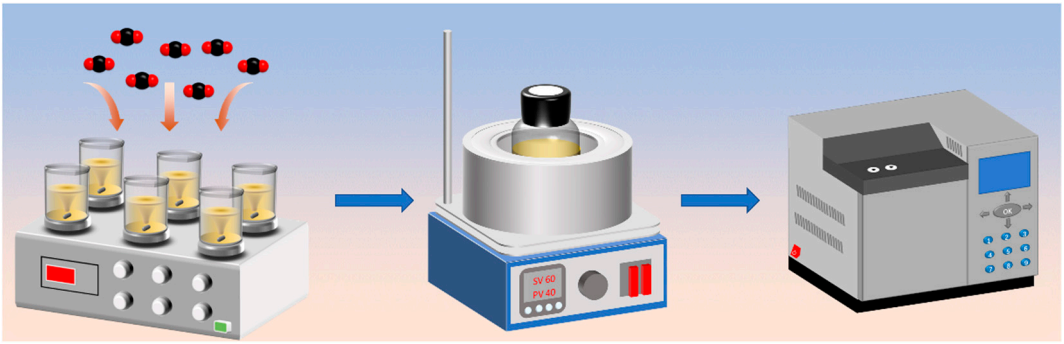


Figure S3. Schematic of imidazole aqueous solution direct air capture and heat concentration of CO_2.

Table S1 Basic properties of ten solid imidazoles.

Compounds	Molecular formula	Molecular weight	Density	Melting point	Boiling point (at 760 mmHg)	Vapour pressure (at 25℃)	pKa (at 25℃)
Imidazole	$C_3H_4N_2$	68.08	1.01 g/mL	90-91 ℃	257 ℃	0.024 mmHg	6.95
2-Methylimidazole	$C_4H_6N_2$	82.10	1.06 g/mL	142-145 ℃	267 ℃	0.014 mmHg	7.85
4-Methylimidazole	$C_4H_6N_2$	82.10	1.02 g/mL	44-47 ℃	263 ℃	0.017 mmHg	7.69
2-Ethylimidazole	$C_5H_8N_2$	96.13	1.02 g/mL	78-80 ℃	268 ℃	0.013 mmHg	7.99
2-Propylimidazole	$C_6H_{10}N_2$	110.16	0.99 g/mL	56-62 ℃	286 ℃	0.005 mmHg	8.02
2-Buthylimidazole	$C_7H_{12}N_2$	124.18	0.98 g/mL	52 ℃	279 ℃	0.007 mmHg	8.00
2-Isopropylimidazole	$C_6H_{10}N_2$	110.16	0.99 g/mL	129-131 ℃	258 ℃	0.0227 mmHg	7.97
1,2-Dimethylimidazole	$C_5H_8N_2$	96.13	1.08 g/mL	29-30 ℃	206 ℃	0.348 mmHg	8.21
2,4-Dimethylimidazole	$C_5H_8N_2$	96.13	0.97 g/mL	85-87 ℃	274 ℃	0.010 mmHg	8.50
2-Ethyl-4-methylimidazole	$C_6H_{10}N_2$	110.16	0.98 g/mL	47-54 ℃	276 ℃	0.008 mmHg	8.68

Table S2 Summary of CO_2 capture capacity of various imidazole aqueous solutions.

Compounds	Concentration	CO_2 capture capacity	Ref.
Imidazole	3 M	0.19 mol/mol	1
1-Methylimidazole	3 M	0.13 mol/mol	1
2-Methylimidazole	3 M	0.42 mol/mol	1
4-Methylimidazole	3 M	0.31 mol/mol	1
1,2-Dimethylimidazole	3 M	0.35 mol/mol	1
Imidazole	30 wt%	0.21 mol/mol	2
1-Methylimidazole	30 wt%	0.12 mol/mol	2
2-Methylimidazole	30 wt%	0.36 mol/mol	2
4-Methylimidazole	30 wt%	0.29 mol/mol	2
2-Ethylimidazole	30 wt%	0.29 mol/mol	2
1,2-Dimethylimidazole	30 wt%	0.36 mol/mol	2
2-Ethyl-4-methylimidazole	30 wt%	0.47 mol/mol	2
Imidazole	6.67 wt%	0.45 mol/mol	This work
2-Methylimidazole	6.67 wt%	0.80 mol/mol	This work
4-Methylimidazole	6.67 wt%	0.67 mol/mol	This work
2-Ethylimidazole	6.67 wt%	0.79 mol/mol	This work
2-Propylimidazole	6.67 wt%	0.78 mol/mol	This work
2-Buthylimidazole	6.67 wt%	0.74 mol/mol	This work
2-Isopropylimidazole	6.67 wt%	0.75 mol/mol	This work
1,2-Dimethylimidazole	6.67 wt%	0.82 mol/mol	This work
2,4-Dimethylimidazole	6.67 wt%	0.85 mol/mol	This work
2-Ethyl-4-methylimidazole	3.33 wt%	1.01 mol/mol	This work
2-Ethyl-4-methylimidazole	6.67 wt%	0.88 mol/mol	This work
2-Ethyl-4-methylimidazole	13.33 wt%	0.80 mol/mol	This work
2-Ethyl-4-methylimidazole	20.00 wt%	0.69 mol/mol	This work

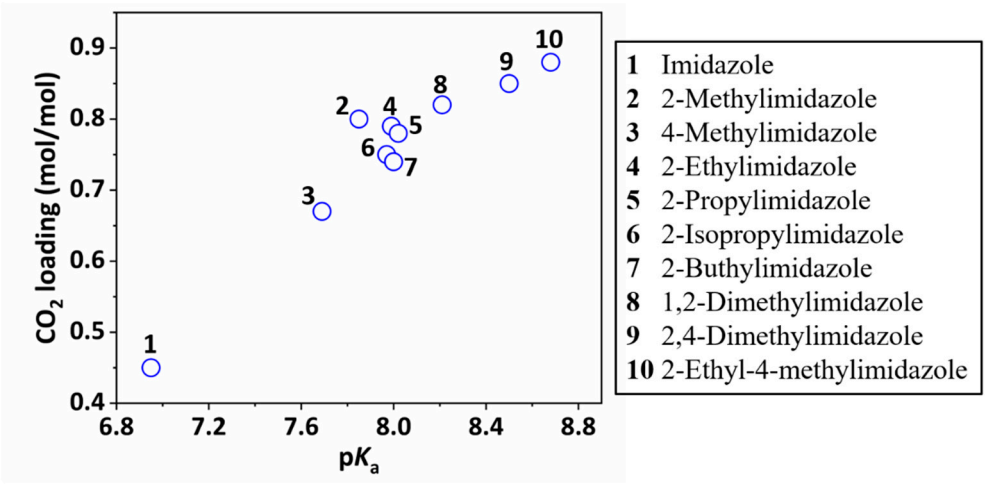
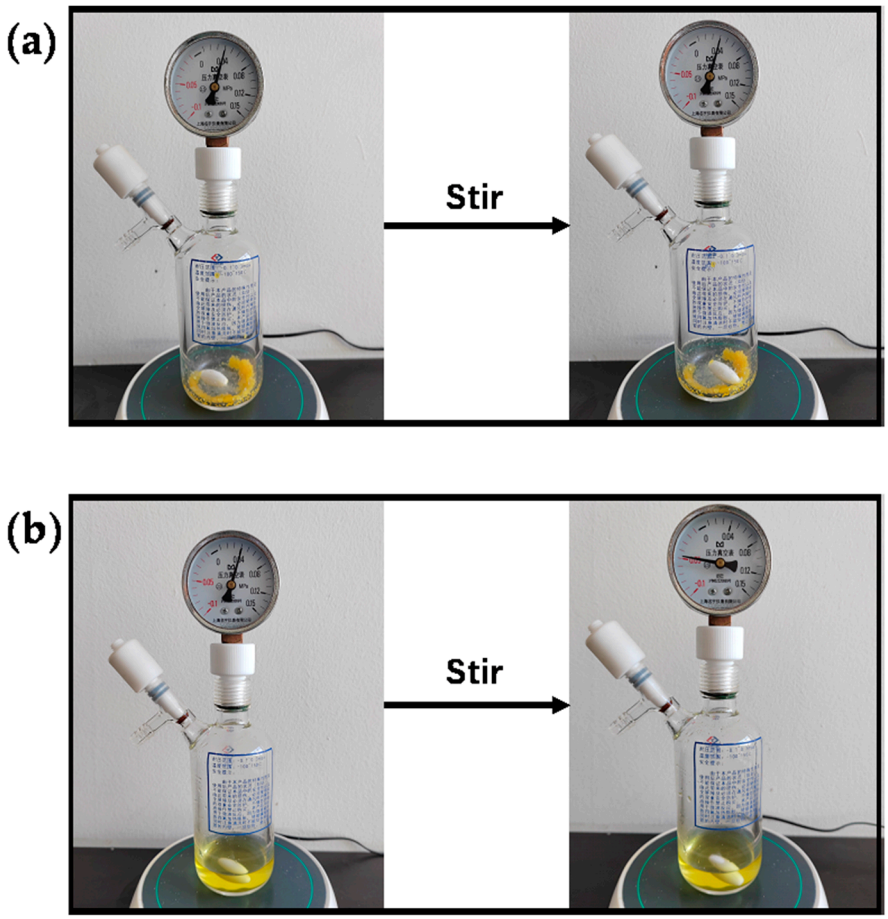


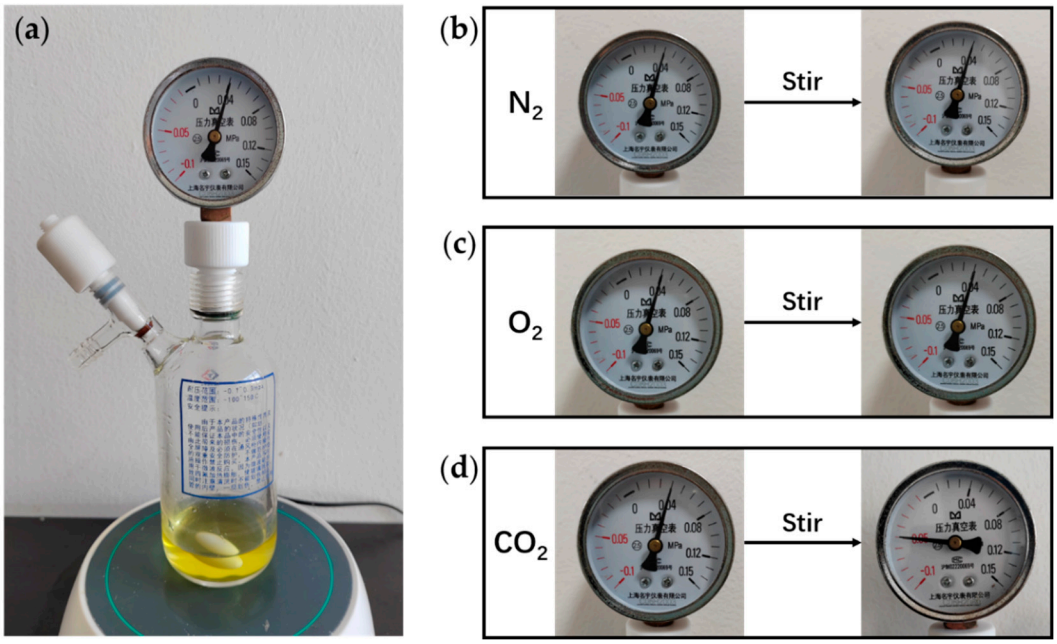
Figure S4. Relationship between molar absorption of CO_2 and p*K*a.



*压力真空表: pressure vacuum gauge

Figure S5. Study on CO_2 absorption capacity of 2-ethyl-4-methylimidazole by using anhydrous solid and aqueous solution. (**a**) Photographs showing the absorption of CO_2 by pure 2-ethyl-4-methylimidazole solid. (**b**) Photographs showing the absorption of CO_2 by 2-ethyl-4-methylimidazole aqueous solution.

The pressure gauge can indicate the change of gas pressure in the pressure-resistant reaction bottle. Before the reaction, the bottle was vacuumized, filled with CO_2 gas to a pressure of 0.04 MPa, and then stirred for absorption. The experiment shows that the pointer of the bottle containing pure 2-ethyl-4-methylimidazole solid keeps a reading of 0.04 MPa, while the pointer of the bottle containing 2-ethyl-4-methylimidazole aqueous solution drops from 0.04 MPa to about -0.05 MPa.



*压力真空表: pressure vacuum gauge

Figure S6. Study on selective gas absorption. (**a**) Photograph of reaction experiment setup. (**b-d**) Photographs showing that the reading of the pressure gauge pointer changes under different gas feeds. (**b**) N_2, (**c**) O_2, (**d**) CO_2.

Add 2-ethyl-4-methylimidazole aqueous solution into a pressure resistant reaction bottle, and vacuum the gas before reaction. Inject high-purity N_2, O_2 and CO_2 gas into the bottle to make the pressure in the bottle reach about 0.04 Mpa, and then stir for absorption. It can be seen that the pointer reading of the bottle filled with N_2 and O_2 is maintained at 0.04 Mpa.

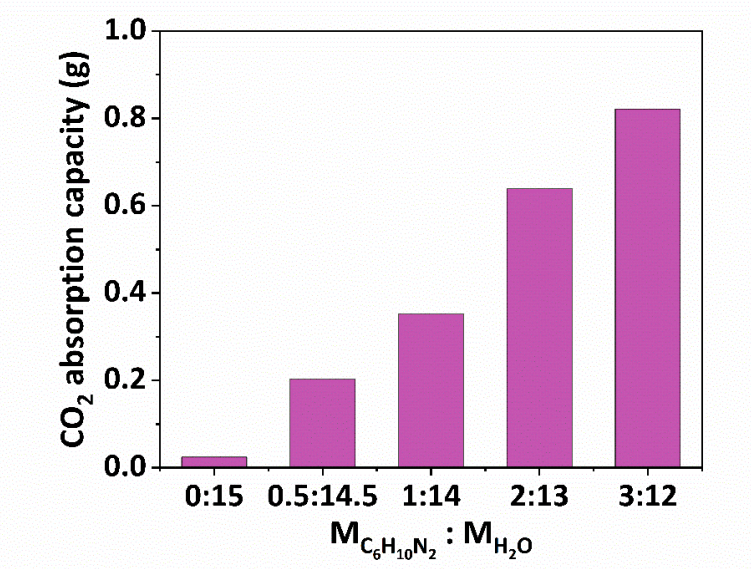


Figure S7. CO_2 absorption capacity of 2-ethyl-4-methylimidazole solution with different mass fractions.

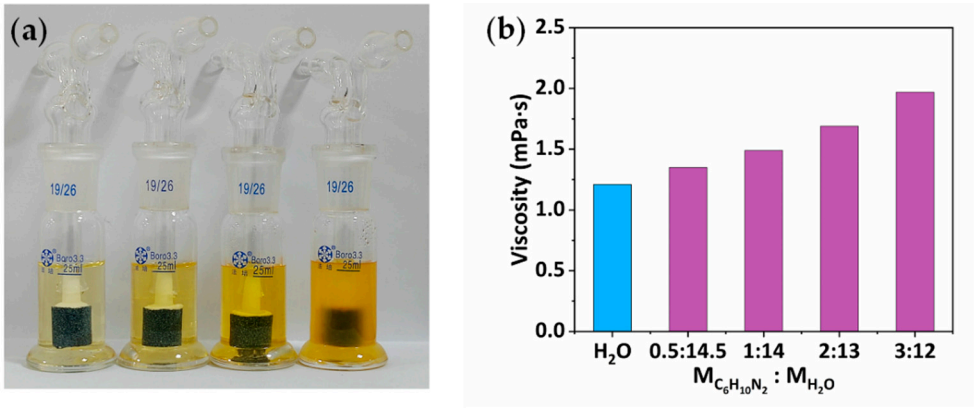


Figure S8. (**a**) Photograph of 2-ethyl-4-methylimidazole solution with different mass fractions. (**b**) Viscosities of various 2-ethyl-4-methylimidazole solutions.

Table S3 Comparison of 2-ethyl-4-methylimidazole and amine absorbents.

Absorbent	Absorption gas composition	CO_2 capture capacity	Desorption temperature	Ref.
DBA + H_2O + ethanol	pure CO_2	0.82 mol/mol	100 ℃	3
DMCA + MCA + H_2O	pure CO_2	0.985 mol/mol	110 ℃	4
AMP + PZ + DME	pure CO_2	0.87 mol/mol	120 ℃	5
EMEA + DEEA	pure CO_2	0.677 mol/mol	110 ℃	6
MEA + sulfolane + H_2O	15% CO_2 85% N_2	0.518 mol/mol	120 ℃	7
[NH_2e-mim][BF_4] + MEA+ H_2O	pure CO_2	0.4554 mol/mol	120 ℃	8
AMP + PZ + MEA+ H_2O	15% CO_2 85% N_2	0.61 mol/mol	90 ℃	9
DEEA + AEEA + H_2O	12% CO_2 88% N_2	0.64 mol/mol	120 ℃	10
MEA + AMP + DMSO + PMDETA	pure CO_2	0.88 mol/mol	120 ℃	11
EMEA + DEEA + PX	pure CO_2	0.59 mol/mol	100 ℃	12
TETA + AMP + NMF	15% CO_2 85% N_2	0.86 mol/mol	120 ℃	13
2-Ethyl-4-methylimidazole + H_2O	pure CO_2	0.80 mol/mol	90 ℃	This work
2-Ethyl-4-methylimidazole + H_2O	15% CO_2 8% O_2 77% N_2	0.54 mol/mol	90 ℃	This work

DBA: dibutylamine, **DMCA**: *N*,*N*-dimethylcyclohexylamine, **MCA**: *N*-methylcyclohexyamine, **AMP**: 2-amino-2-methyl-1-propanol, **PZ**: piperazine, **DME**: dipropylene glycol dimethyl ether, **EMEA**: *N*-ethylmonoethanolamine, **DEEA**: *N*,*N*-diethylethanolamine, **MEA**: monoethanolamine, **[NH_2e-mim][BF_4]**: (1-(1-aminoethyl)-3-methylimidazole fluoroborate, **AEEA**: 2-((2-aminoethyl) amino) ethanol, **DMSO**: dimethyl sulfoxide, **TETA**: triethylene tetramine, **PX**: *p*-xylene, **PMDETA**: *N*,*N*,*N*′,*N*′′,*N*′′-pentamethyldiethylenetriamine, **NMF:** *N*-methylformamide.

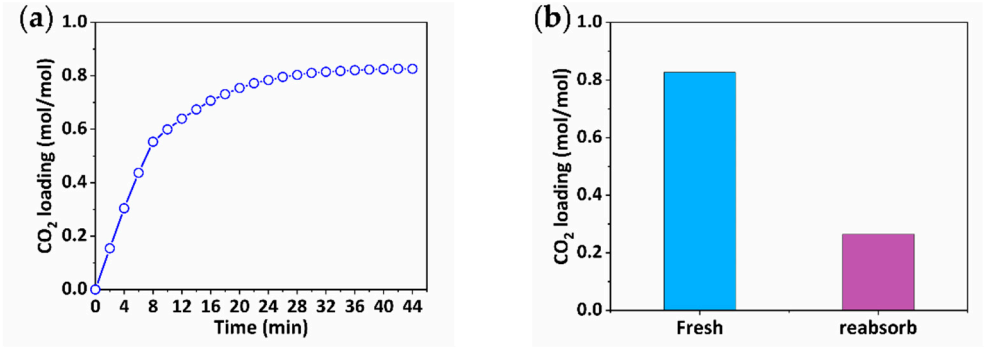


Figure S9. CO_2 absorption-desorption performance of ethanolamine. (**a**) Absorption capacity curve of ethanolamine with time (2 g MEA + 13 mL H_2O). (**b**) After desorption at 90 °C, the secondary reabsorption rate of MEA significantly decreases.

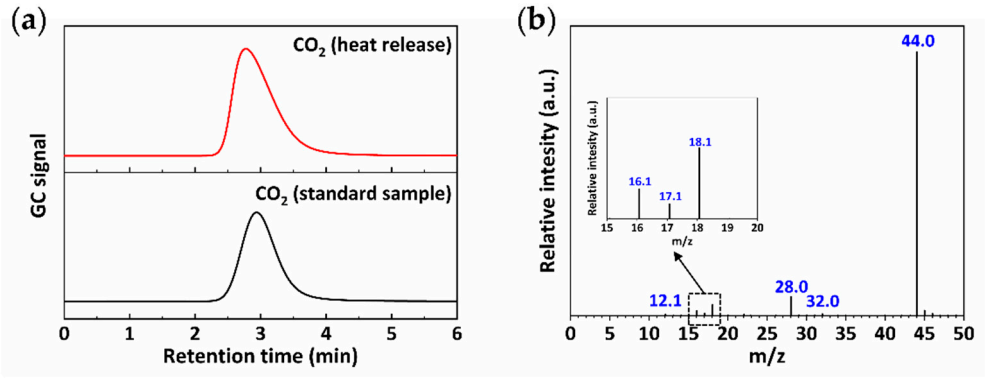


Figure S10. (**a**) Gas chromatography measurements of heat released gas. (**b**) Mass spectrum of heat released gas.

In order to verify the accuracy of the GC signal, standard CO_2 was tested, and the GC curve (black line) was recorded for comparison. As seen, the GC signal of the released gas is identical to that of standard CO_2, confirming that CO_2 is desorbed.

In a mass spectrometer, CO_2 molecules can be ionized or even split into charged fragments, such as $[CO_2]^+$, $[O_2]^+$, $[CO]^+$, $[O]^+$ and $[C]^+$. Fragment signal distribution is as follows: m/z=12.1, $[C]^+$; m/z=16.1, $[O]^+$; m/z = 28.0, $[CO]^+$; m/z = 32.0, $[O_2]^+$; m/z = 44.0, $[CO_2]^+$. At the same time, due to a small amount of H_2O in the released gas, there is m/z=16.1, $[O]^+$; m/z=17.1, $[OH]^+$; M/z=18.1, $[H_2O]^+$ weak segment signal.

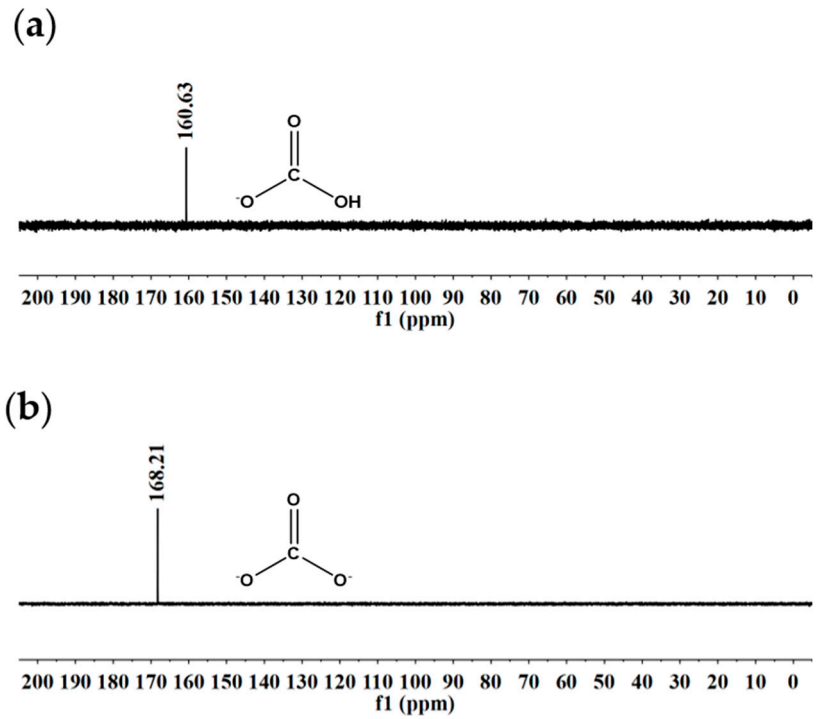


Figure S11. (**a**) ^{13}C NMR spectrum of $NaHCO_3$ aqueous solution. (**b**) ^{13}C NMR spectrum of $NaCO_3$ aqueous solution.

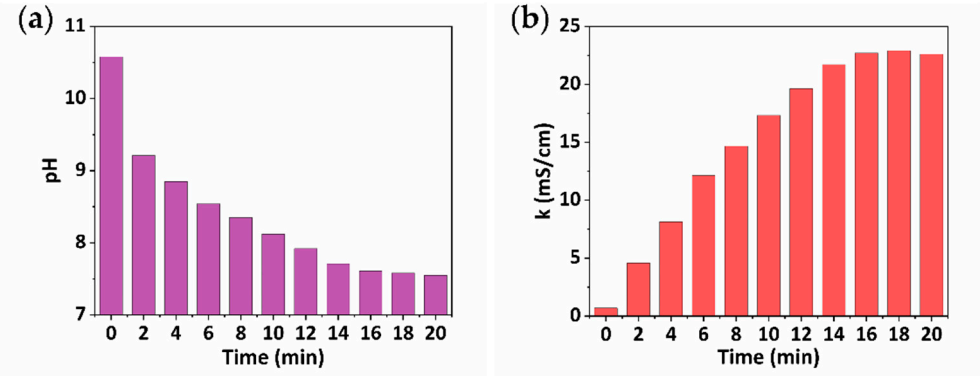


Figure S12. Changes of pH value and conductivity with time during CO_2 absorption by 2-ethyl-4-methylimidazole aqueous solution. (**a**) pH change. (**b**) Conductivity change.

It is worth noting that the initial conductivity of the solution is only 691 μS/cm, the conductivity increases rapidly after CO_2 is introduced, and the conductivity after absorption saturation is 22.7 mS/cm, about 33 times of the initial conductivity value. It shows that there is only a small amount of OH^- in the reaction between 2-Ethyl-4-methylimidazole and water in the initial solution, but when CO_2 is introduced, a large number of bicarbonate ions form and the reaction balance moves to the right, generating more OH^- for CO_2 absorption.

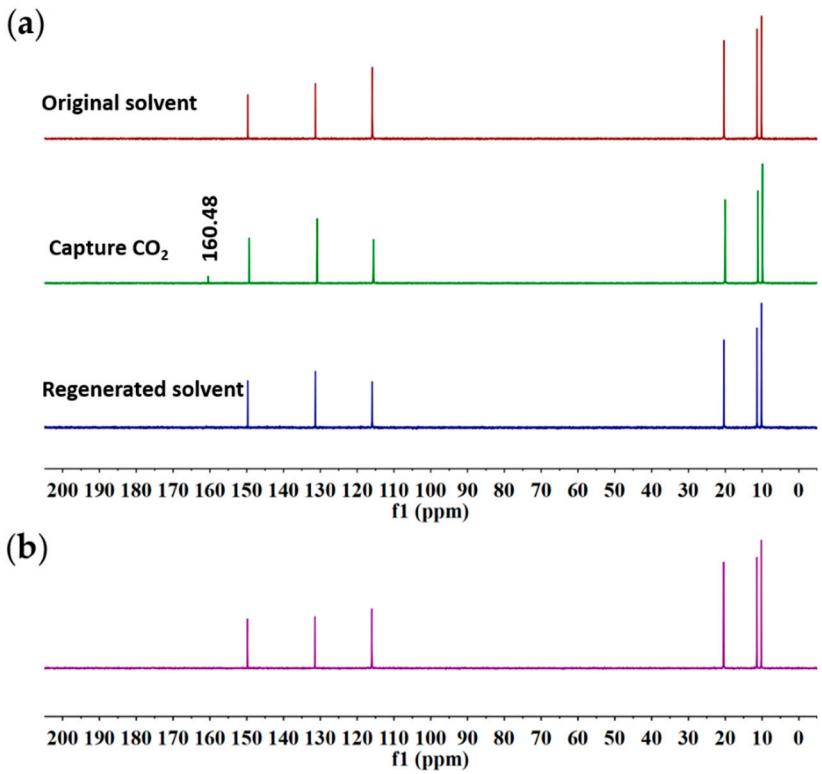


Figure S13. (**a**) ^{13}C NMR spectra of 2-ethyl-4-methylimidazole during absorption-desorption of CO_2 from the synthetic flue gas. (**b**) ^{13}C NMR spectrum of 2-ethyl-4-methylimidazole after 8 cycles.

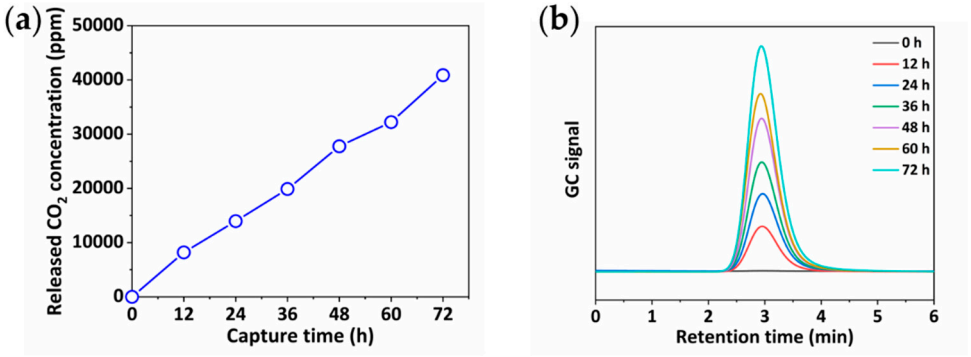


Figure S14. The capture of ambient air by 2-ethyl-4-methylimidazole. (**a**) Curve of CO_2 enrichment concentration with capture time. (**b**) Corresponds to the change of peak area detected by gas chromatography.

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