### **Supporting Information**

# Evaluation of hybrid amine and alcohol solvent with ionexchange resin catalysts for energy-efficient CO<sub>2</sub> capture

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 different catalysts in the rich amine solvent.

#### Section S1. Materials

Monoethanolamine (99%), 2-(Methylamino)ethanol(99%), 2-(Ethylamino)ethanol (99%), 2-(Diethylamino)ethanol (99%). N.Ndimethylethanolamine (98%) and N-Methyldiethanolamine (99%), Amberlyst-15 resin, and Amberlite IR-120 resin were obtained from Shanghai Aladdin Industrial Corporation, China. Amberlite-732 resin, and Amberlyst-35 resin were purchased from Shanghai Macklin Chemical Reagent Co. Ltd., China. HCl (AR, 36.0 - 38.0%), methanol (AR, 99%) and ethanol (AR, 99%) were obtained from Sinopharm Chemical Reagent Co. Ltd., China. CO<sub>2</sub> (99.9%), and N<sub>2</sub> (99.99%) were acquired from Changsha Rizhen Gas Co. Ltd., China.

#### Section S2. Resin activation.

The different types of commercial ion exchange resins were activated by the impregnation method at room temperature in 2 M HCl for 24 h. After that, the ion exchange resins were washed with deionized water until the filtrate was neutral. Then, the ion exchange resins were washed with ethanol and dried at 60 °C for 24 h to give the final, activated catalysts used for this study.

#### Section S3. Catalyst characterization

#### Section S3.1. Nitrogen physisorption

The  $N_2$  physisorption technique was used to measure the surface area, pore volume and pore size of the catalysts and was performed on the NOVA 2000e nitrogen adsorption desorption instrument. Before each analysis, the prepared catalyst sample was degassed at 120 °C for 8 h using helium. The pore volume and size were obtained by using the Barrett-Joyner-Halenda (BJH) method. The specific surface area was calculated by the Brunauer-Emmett-Teller (BET) theory.

#### Section S3.2. Total acid sites analysis

The Bronsted acid sites of the catalyst samples were measured by acid-base titration method as reported by Bozkurt et al <sup>1</sup>. Approximately 0.1 g catalyst was put into 20 mL of 2.0 M NaCl solution for 24 h. Then, the liquid was titrated with 0.01 M NaOH. The acid sites (mmol  $H^+/g_{cat}$ ) were calculated in Eq S1 as follows:

$$The acid capacity = \frac{molarity of NaOH\left(\frac{mmol}{mL}\right) * volume of NaOH spent(mL)}{catalyst(g)}$$
(S1)

#### Section S4. Experimental apparatus

The experimental apparatus for  $CO_2$  desorption and absorption mainly consisted of a round bottom four-neck flask as the batch reactor, a mass flowmeter (Beijing Seven-star Electronics Co., Ltd, China), a heating mantle (Henan Shengbo instrument Co. Ltd., China), an infrared  $CO_2$  analyzer (GS10, Ennix, Germany), and an electricity meter (Zhejiang Tepsung electric Co.Ltd.) as presented in Figure S1.



# Figure S1. Schematic diagram of the experimental apparatus for CO<sub>2</sub> desorption and absorption.

Briefly, in the CO<sub>2</sub> desorption experiment, 500 mL (in sections 3.2.1, 3.2.2, 3.2.3) or 300 mL (in sections 3.2.4, 3.2.5, 3.4) CO<sub>2</sub>-rich amine solution with or without catalysts or ethanol solvent were added into the round bottom flask. The desorption temperature was raised from 25 to 90 °C and held at 90  $\pm$  0.1 °C. Also, the solution was stirred at 1200 rpm. The CO<sub>2</sub> released from the rich amine solution was cooled, dried, and mixed with 1 L/min N<sub>2</sub> carrier. Meanwhile, the CO<sub>2</sub> concentration was measured every ten seconds by an infrared CO<sub>2</sub> analyzer during the desorption experiment. The end of each desorption test was realized as the point where the concentration of CO<sub>2</sub> in the carrier gas was lower than 0.2%. The heat input for each desorption test was recorded by an electricity meter. However, this obtained value of the heat duty is higher than the actual value due to the simplified apparatus so that for this work the relative heat duty was used to compare the desorption performance.

For the CO<sub>2</sub> absorption experiment, a magnetic stirring rate of 1200 rpm with temperature set at 40 °C was used. The simulated flue gas (15 vol% CO<sub>2</sub>, and 85 vol% N<sub>2</sub>) with a total flowrate of 0.5 L/min was bubbled into the lean amine solution. The CO<sub>2</sub> concentration in the outlet gas was also recorded by an infrared CO<sub>2</sub> analyzer as aforementioned.

During the absorption and desorption processes, the amount of  $CO_2$  absorbed or desorbed was determined by two quantitative methods. The gas phase measurement

was obtained by the infrared  $CO_2$  analyzer. The liquid phase measurement was made with Chittick apparatus <sup>2</sup>. The mass balance was verified by comparing the results from these two methods, and the average absolute relative deviations between gas and liquid phase measurements were within 5%.

#### Section S5. Analysis

The CO<sub>2</sub> reaction rate (r, mol/(s\*L)) was measured through the CO<sub>2</sub> concentration by the infrared CO<sub>2</sub> analyzer in Eq S2, and the quantity of CO<sub>2</sub> desorbed ( ${}^{N_{CO_2,gas}}$ , mmol) from the gas method was calculated in Eq S3 as follows:

$$r = \left| \frac{1}{22.4 \times V} \left( v_{CO_2}^{in} - \frac{X_{CO_2}^{out}}{1 - X_{CO_2}^{out}} \times v_{N_2}^{in} \right) \right|$$

$$N_{CO_2,gas} = V \int_0^t r dt$$
(S2)
(S2)

Here, V(L) represents the volume of amine solution,  $v_{CO_2}^{in}, v_{N_2}^{in}$  (mol/s) represent the gas flowrates of CO<sub>2</sub> and N<sub>2</sub> from the mass flowmeter, respectively, and  $X_{CO_2}^{out}$  (%) represents the CO<sub>2</sub> concentration of the outlet gas.

The quantity of CO<sub>2</sub> desorbed ( ${}^{N_{CO_2, liquid}}$ , mmol) from the liquid method was calculated with Eq S4, and the cyclic capacity (mol/mol) was defined in Eq S5 as follows.

$$N_{CO_2, liquid} = (\alpha_{rich} - \alpha_{lean}) \times C \times V$$
(S4)

The cyclic capacity = 
$$\alpha_{rich} - \alpha_{lean}$$
 (S5)

Here,  $\alpha_{rich}$ ,  $\alpha_{lean}$  (mol CO<sub>2</sub>/mol amine) represent the CO<sub>2</sub> loadings of rich and lean amine solution, respectively. *C* (mol/L) represents the concentration of the amine. The heat duty (*HD*, kJ/mol) of amine regeneration was obtained from Eq S6, and the relative heat duty (*RHD*, %) was defined in Eq S7 for a fair comparison as follows.

$$HD = \frac{E_{electricity}}{N_{CO_2,gas}}$$
(S6)

$$RHD = \frac{HD_i}{HD_{baseline}} \times 100\%$$
(S7)

Here,  $E_{electricity}$  (kJ) represents the energy consumption for amine regeneration recorded by the electricity meter,  $HD_i$  (kJ) represents the HD of different amine systems with catalysts or ethanol solvent, and  $HD_{baseline}$  (kJ) represents the HD of the amine regeneration blank run.

#### Section S6. Influence of catalyst types at 80°C

To obtain a more convective conclusion on the catalyst performance for solvent regeneration, the catalytic activities of the studied catalysts were investigated at additional temperature of 80 °C, which is lower than the typical catalytic solvent regeneration temperature of 90 °C. As a result, the CO<sub>2</sub> desorption rates of CO<sub>2</sub>-rich MEA solution with and without cation exchange resin catalysts at 80 °C are displayed in Figure S2. The peak CO<sub>2</sub> desorption rate of CO<sub>2</sub>-rich MEA solution without catalyst was  $4.614 \times 10^{-5}$  mol/(s\*L) at 1260 s. The maximum desorption rate of Amberlite IR-120 catalyst was  $4.963 \times 10^{-5}$  mol/(s\*L) at 1260 s. This showed a slight improvement compared with the blank run. The Amberlyst-35 slightly improved the CO<sub>2</sub> desorption rate and achieved its maximum of  $5.266 \times 10^{-5}$  mol/(s\*L) at 1180 s. Moreover, the peak

desorption rate of CO<sub>2</sub>-rich solution with Amberlite-732 and Amberlyst-15 catalysts were  $5.566 \times 10^{-5}$  and  $5.571 \times 10^{-5}$  mol/(s\*L) at 1250 and 1200 s, respectively. It can be concluded that all the catalysts reached the maximum desorption rate earlier than the blank run, and the value of the peak CO<sub>2</sub> desorption rate was higher than for the MEA solution without catalysts. Moreover, the Amberlyst-15 catalyst best accelerated the proton transfer and best enhanced the CO<sub>2</sub> desorption rate.

In addition to the CO<sub>2</sub> desorption rate, the CO<sub>2</sub> desorption amount is improved in the first 1200 s at 80 °C. Only 12.0 mmol CO<sub>2</sub> was desorbed from CO<sub>2</sub>-rich MEA solution without catalyst, while 13.8 mmol CO2 was released when the Amberlite IR-120 was used. The Amberlyst-35 slightly enhanced the CO<sub>2</sub> desorbed amount to 14.5 mmol, and the quantities of CO<sub>2</sub> desorption attributed to the use of Amberlite-732 was 15.0 mmol. The best performance was exhibited by Amberlyst-15, which led to a  $CO_2$ desorption amount of 16.3 mmol, 36% higher than that for CO<sub>2</sub>-rich MEA solution without catalyst. Therefore, all the ion-exchange resin catalysts are capable of desorbing greater amounts of CO<sub>2</sub> compared to the blank run, thus reducing the heat duty of MEA solution regeneration because the heat duty is related to the quantity of CO<sub>2</sub> desorbed per unit of energy consumption. The Dowex D001, Amberlite IR-120, and Amberlyst-35 slightly reduced the heat duty by around 7.7, 12.7, and 17.2% compared with the noncatalytic MEA solution system, respectively. Also, the Amberlite-732 decreased the heat duty by around 20.0%. It is noteworthy that the Amberlyst-15 catalyst minimized the heat duty up to 26.2%, demonstrating its superior

MEA regeneration efficiency. In terms of the relative heat duty, the catalytic performance trend was seen to be Amberlyst-15 > Amberlite-732 > Amberlyst-35 > Amberlite IR-120 > Dowex D001 > no catalyst, which is consistent with the trend measured at 90°C.



**Figure S2.** The CO<sub>2</sub> desorption rate curves with different catalysts at 80 °C.

#### Section S7. Influence of initial CO<sub>2</sub> loading

According to the VLE model of the MEA-CO<sub>2</sub>-H<sub>2</sub>O system, the mole fraction of  $HCO_3^-$  increases with increasing CO<sub>2</sub> loading, while the mole fraction of CO<sub>3</sub><sup>2-</sup> is negligible. Figure S2a presents the desorption rate curves for catalytic and noncatalytic MEA systems with initial CO<sub>2</sub> loading of 0.53, 0.51, and 0.49 mol CO<sub>2</sub>/mol MEA. The amount of CO<sub>2</sub> desorbed in 1200 s with and without catalyst is compared in Figure S2b. As can be seen, with a decrease in initial CO<sub>2</sub> loading, the quantities of CO<sub>2</sub> desorbed within 1200 s tended to decrease linearly. As shown in Figure S2b, the MEA blank run could desorb 92.00, 57.00, 35.00 mmol CO<sub>2</sub> with the initial CO<sub>2</sub> loading of 0.53, 0.51, and 0.49 mol CO<sub>2</sub>/mol MEA, respectively. The Amberlyst-15 catalyst with CO<sub>2</sub> initial loading of 0.53 and 0.51 mol CO<sub>2</sub>/mol MEA increased the quantities of CO<sub>2</sub> released

by 21.47%, 26.75%. The highest increment by 39.29% in the desorbed amount of  $CO_2$  was presented for the initial  $CO_2$  loading of 0.49 mol  $CO_2$ /mol MEA with 2 wt.% Amberlyst-15.



Figure S3. The  $CO_2$  desorption performance with different initial  $CO_2$  loading. (a)

 $CO_2$  desorption rate curves. (b) Total amount of  $CO_2$  desorbed at 1200 s.



Figure S4. Raman spectra of MEA solution at various periods. (a) CO<sub>2</sub> desorption

without catalyst. (b)  $CO_2$  desorption with 2 wt.% Amberlyst-15.

#### Section S8. Influence of catalyst/MEA solvent ratio

As aforementioned, the Amberlyst-15 exhibited superior catalytic performance among the ion exchange resin catalysts. Of interest then, is the influence of catalyst/MEA solvent ratio on CO<sub>2</sub> desorption performance. To study this, the amounts of 1, 2, and 3 wt.% of catalyst were introduced into CO<sub>2</sub>-rich MEA solutions, and the desorption rate, the amount of CO<sub>2</sub> released, and the relative heat duty were compared and illustrated in Figure S4a-S4b. From Figure S4a, it is clear that an insignificant improvement was observed in the desorption rate with the catalyst/MEA solvent ratio of 1 wt.%. With the catalyst/MEA solvent ratio at 2 wt.% and at 3 wt.% there was obvious but similar enhancement of the desorption rate. As shown in Figure S4b, when the catalyst/MEA solvent ratio was increased from 1 wt.% to 2 wt.%, the amount of CO2 released increased, which could be attributed to the increase in the number of catalyst proton acid sites in the system. But when the catalyst/MEA solvent ratio was further increased up to 3 wt.%, the amount of CO<sub>2</sub> desorbed only increased slightly, which may be due to excess catalyst leading to transfer resistance.

The regeneration heat duty with different ratios is also presented in Figure S4b, and the 2 wt.% Amberlyst-15 showed lower relative heat duty of 78.89% than that of 1 wt.% Amberlyst-15 of 93.06%. While the 3 wt.% Amberlyst-15 presented the lowest heat duty of 75.75%, this is only slightly lower than for the 2 wt.%. Hence, the optimal ratio of catalyst/MEA solvent is 2 wt.% in this study, which is consistent with the results by Zhang et al <sup>3</sup>.



**Figure S5.** The CO<sub>2</sub> desorption performance with different catalyst/MEA solvent ratios. (a) CO<sub>2</sub> desorption rate curves. (b) Total amount of CO<sub>2</sub> desorbed and relative heat duty at 1200 s.

#### Section S9. Influence of methanol concentration

Figure S5a shows the desorption rate of 5 M MEA with different methanol concentrations, and it is clear that the methanol solvent significantly enhanced the desorption kinetics. In the MEA-water blank test run, the maximum CO<sub>2</sub> desorption rate only reached  $2.613 \times 10^{-4}$  mol/(s\*L) at 1120 s. Under the same condition, methanol-MEA-water blend solvent with 5, 10, 12.5, 15 wt.% methanol reached maximal release rate of CO<sub>2</sub> of  $3.803 \times 10^{-4}$  mol/(s\*L),  $5.097 \times 10^{-4}$ mol/(s\*L),  $5.748 \times 10^{-4}$ mol/(s\*L) and  $6.252 \times 10^{-4}$ mol/(s\*L), at 1130 s, 1010 s, 1100 s and 1050 s, respectively. In comparison with the blank MEA solvent, the maximum value of CO<sub>2</sub> desorption rate was notably improved with the increasing of methanol concentration. However, above the methanol concentration of 20 wt.%, only a slight improvement was observed in the maximum desorption rate in comparison with the concentration of 15 wt.% at  $6.252 \times 10^{-4}$ mol/(s\*L). Considering that too high a methanol concentration may cause a large

amount of volatilization in the absorption process, the optimal methanol concentration is 15 wt.%.

The different quantities of  $CO_2$  released over 1200 s is given in Figure S5c- S5d. The blended solvent with various concentrations of methanol not only improved the release kinetics but also increased the amount of  $CO_2$  desorbed. As illustrated in Figure S5d, only 42.45 mmol  $CO_2$  is released from the blank 300 mL of 5 M  $CO_2$ -rich MEA in 1200 s, while the rapid kinetics of the methanol/MEA/water solvent resulted in 58.95 to 116.40 mmol  $CO_2$  with methanol concentration in the range of 5-20 wt.%. In comparison with the 15 wt.% methanol solvent, insignificant improvement in  $CO_2$ desorption quantity was presented with the higher 5 wt.% methanol solvent addition, namely, 20 wt.%.

To further investigate the effect of methanol concentration on  $CO_2$  desorption, the  $CO_2$  loading curves as a function of time and the cyclic capacity are compared in Figure S5b and Figure S5e, respectively. Figure S5b summarizes the  $CO_2$  loading over 3600 s of desorption process. The  $CO_2$  loading at the end of the desorption step is 0.44 mol  $CO_2$ / mol MEA for the blank MEA solvent, which shows a poor cyclic capacity of 0.07 mol  $CO_2$ / mol MEA at the low temperature of 90 °C. However, methanol/MEA/water solvent presents an excellent regeneration ability and cyclic capacity under the same temperature. Among the different concentrations of methanol, the 20 wt.% methanol-MEA-water solvent shows a cyclic capacity as high as 0.26 mol  $CO_2$ / mol MEA, an improvement of 3.7 times compared with that of blank MEA solvent. It should be noted

that the cyclic capacity is already about 0.23 mol  $CO_2$ / mol MEA achieved with the concentration of methanol at 15 wt.%, only slightly less than the 20 wt.% methanol solvent. Overall, these results demonstrate that the addition of methanol could significantly enhance the cyclic capacity of MEA based solvent. In addition to improving the cyclic capacity to achieve practical industrial requirements at low temperature, the regeneration heat duty was also reduced because greater quantities of  $CO_2$  were released with the same reaction time.





Figure S6. Effect of methanol concentration on CO<sub>2</sub> desorption performance at 90 °C.
(a) CO<sub>2</sub> desorption rate curves. (b) CO<sub>2</sub> loading curves. (c) Desorbed CO<sub>2</sub> amount

curves. (d) Total amount of CO2 desorbed at 1200 s. (e) Cyclic capacity. (f) Relative



**Figure S7.** Comparison of cyclic capacity for the different alcohol-MEA systems at different regeneration time. (a) cyclic capacity of MeOH-MEA system. (b) cyclic

capacity of EtOH-MEA system.



Figure S8. Comparison of relative heat duty for methanol and ethanol addition system at 1200 s.

#### Section S10. Effect of catalyst and ethanol on CO<sub>2</sub> absorption performance

Evaluating the  $CO_2$  absorption process is an important part of understanding thermal amine-based  $CO_2$  capture technology. Thus, examining the effect of Amberlyst-15 catalyst and ethanol solvent on  $CO_2$  absorption performance is required. The  $CO_2$  absorption curves of  $CO_2$  loading as a function of time for blank, catalyst-MEA-water and catalyst-EtOH-MEA-water systems are presented in Figure S8. Note that there is no significant difference in catalyst-MEA-water and catalyst-EtOH-MEAwater systems compared with the blank run. Consequently, it can be concluded that the introduction of catalyst and ethanol solvent had no negative effect on the  $CO_2$ absorption process in terms of  $CO_2$  loading and absorption rate.



Figure S9. Influence of catalyst and ethanol solvent on CO<sub>2</sub> absorption performance.

Table S1. A brief review of research works for solvent regeneration process with

Catalyst	Single/blend solvent	Desorption temperature (°C)	Main results	References
γ- Al <sub>2</sub> O <sub>3</sub> /HZSM-5, HZSM-5, HY, γ-Al <sub>2</sub> O <sub>3</sub>	MEA	50-105	Mixture catalysts presented better performance than single catalysts; Amount of desorbed $CO_2$ increased by 20.1-31.2%; Heat duty reduced by 16.9-23.7%.	(Liang et al., 2016) <sup>4</sup>
SAPO-34, SO <sub>4</sub> <sup>2-</sup> /TiO <sub>2</sub>	MEA	70-96	<ul> <li>SAPO-34 showed better performance than SO<sub>4</sub><sup>2-</sup>/TiO<sub>2</sub>;</li> <li>Amount of desorbed CO<sub>2</sub> increased by 14.1-28.2%;</li> <li>Heat duty reduced by 17.1-24.3%.</li> </ul>	(Zhang et al., 2017) <sup>5</sup>
HZSM-5, MCM-41, SO <sub>4</sub> <sup>2-</sup> /ZrO <sub>2</sub>	MEA	70-98	Performance, HZSM-5 > MCM-41 > $SO_4^{2-}/ZrO_2$ ; Amount of desorbed CO <sub>2</sub> increased by 10.6-29.4%; Heat duty reduced by 9.8-24.8%.	(Liu et al., 2017) <sup>6</sup>
Nanoparticles SiO <sub>2</sub> , TiO <sub>2</sub> , Al <sub>2</sub> O <sub>3</sub>	MEA	103	The $TiO_2$ shows best performance, and the use of $TiO_2$ nanoparticles saved desorption time by 42%.	(Wang et al., 2016) <sup>7</sup>
$V_2O_5$ , $MoO_3$ , $WO_3$ , $TiO_2$ , and $Cr_2O_3$ ,	MEA	35-86	Performance, $MoO_3 > V_2O_5 > Cr_2O_3 >$ $TiO_2 > WO_3$ ; Amount of desorbed $CO_2$ increased by 44-	(Bhatti et al., 2017) <sup>8</sup>

different catalysts in the rich amine solvent.

			94%;	
			Sensible heat reduced by 25-48%.	
			Amount of desorbed CO <sub>2</sub> increased by	( <b>7</b> hong at
SZMF ME	MEA	60-98	38.1-54.7%;	(Znang et)
			Heat duty reduced by 27.7-39.4%.	$a_{1}, 2019)^{5}$
SO <sub>4</sub> <sup>2–</sup> /ZrO <sub>2</sub> - HZSM-5			Amount and rate of CO <sub>2</sub> desorption	
	МЕА	98	increased by 40 and 37%;	(Xing et
	MLA		Energy consumption reduced by	al., 2020) <sup>10</sup>
			approximately 31%.	
SO <sub>4</sub> <sup>2-</sup> /ZIF-67- C@TiO <sub>2</sub>			Amount and rate of CO <sub>2</sub> desorption	
	MEA	88	increased by 64.5 and 153%;	(Xing et
			Energy consumption reduced by	al., 2021) <sup>11</sup>
			approximately 36%.	
γ-			The combination of "tri-solvent +	
		heterogeneous catalysts" was 0.3+2+2		
$Al_2O_3/HZSM-5$ ,	MEA-BEA-	90	mol/L MEA+BEA+AMP + blended $\gamma$ -	(Shi et al.,
HZSM-5, γ-	AMP	90	$Al_2O_3/H-ZSM-5 = 2:1$ , whose relative heat	$2021)^{12}$
$Al_2O_3$			duty (%) was 32.9% compared to 5.0 M	
			MEA as a benchmark.	
			$V_2O_5$ , $WO_3$ , and $TiO_2$ were found to be	
$V_2O_5$ , $WO_3$ ,	DGA- DEGMME	90	effective in decreasing the relative heat	(Bhatti et
and $TiO_2$ ,			duty by 23.5%, 14.6%, and 14.4%,	al., $2021$ ) <sup>13</sup>
			respectively.	
Fe/Ni@COF			The obtained nanomaterials achieve a	
		88	considerable improvement in $CO_2$	(Li et al., 2022) <sup>14</sup>
	MEA		desorption amount, representing a	
			substantial increase of 540% relative to	,
CeO <sub>2</sub> -MOF- M HPW			traditional thermal desorption.	
	MEA		The $CO_2$ desorption capacity and rate were	
		0.0	increased by 38.1% and 100%,	(Wei et al.,
		MEA 88	88	respectively, and the desorption energy
			consumption was reduced by 29.4% In	
			comparison with the un-catalytic process.	

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