Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Quantification alkalinity of Deep Eutectic Solvents based on (H_-) and NMR

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Table S1. H_- values for hydrazine aqueous solution measured in this work at room temperature and reported in the literature.

Hydrazine (wt %)	H_- (reported)	H_- (measured)
15	11.93	11.85
20	12.29	12.17
25	12.72	12.56
30	13.15	12.94

Machine learning details:

Herein, the Pearson correlation coefficient p were applied to evaluate the relevance between two features, which can be expressed as:

$$p = \frac{\sum_i (x_i - \bar{x})(y_i - \bar{y})}{\sqrt{\sum_i (x_i - \bar{x})^2} \sqrt{\sum_i (y_i - y)^2}}$$

where x and y are two features and $\bar{x}$ and $\bar{y}$ are the mean values for all x and y. p value range from -1 to 1, and an absolute value close to 1 means a high linear relationship with two features.

Additionally, the coefficient of determination (R^2) and mean squared error (MSE) were applied for all algorithms to evaluated the accuracy of ML models. R^2 reflects the prediction performance of a model and a high value close to 1 is required for an ideal model. MSE measures the difference between true and predicted values for all entities and a small value to zero is desired. Here, R^2 can be expressed as followed:

$$R^2 = 1 - \frac{\sum_{i=1}^{n} (y_{pred} - y_{true})^2}{\sum_{i=1}^{n} (y_{pred} - \bar{y}_{pred})^2}$$

while MSE can be expressed:

$$MSE = \frac{\sum_{i=1}^{n} (y_{pred} - y_{true})^2}{n}$$

where y_{pred} and y_{true} represent the predicted and true values, and $\bar{y}_{pred}$ stands for the average values of all y_{pred}.

Table S2. The key hyperparameters for each algorithm in our ML models applied for grid search (hyperparameters not mentioned were kept at their default values).

Algorithms	Hyperparameters
Random Forest Regression	n_estimators = [5, 10, 20, 50, 70, 100] max_depth = [5, 6, 7, 9, 10, 20] max_features = [0.6, 0.7, 1]
XGBoost Regression	n_estimators = [5, 10, 20, 50, 70, 100, 200] max_depth = [5, 6, 7, 8]

	max_delta_step = [1, 3, 5, 7]
Artificial Neural Network	-

Table S3. The best hyperparameters for each algorithm in our ML models determined by ten-fold cross-validation (hyperparameters not mentioned were kept at their default values).

Algorithms	Hyperparameters
Random Forest Regression	n_estimators = 5 max_depth = 9 max_features = 0.7
XGBoost Regression	n_estimators = 20 max_depth = 5 max_delta_step = 1
Artificial Neural Network	-

Table S4. The mean squared error (MSE) and coefficient of determination (R^2) for different algorithm models.

	RF	XGB	ANN
MSE	$1.864*10^{-5}$	$1.929*10^{-5}$	$5.401*10^{-5}$
R^2	0.977	0.976	0.934

Table S5. The synthetic guanidine DESs components and abbreviations.

Name	Components	Abbreviation
DES1	Choline chloride/Tetramethylguanidine/ethylene glycol	ChCl/TMG/EG
DES2	Choline chloride/Tetramethylguanidine/ Butyleneglycol	ChCl/TMG/BDO
DES3	Choline chloride/Tetramethylguanidine/ Poly(ethylene glycol)	ChCl/TMG/PEG
DES4	Choline chloride/ Ditolylguanidine /ethylene glycol	ChCl/DTG/EG
DES5	Choline chloride/ Ditolylguanidine / Butyleneglycol	ChCl/DTG/BDO
DES6	Choline chloride/ Ditolylguanidine / Poly(ethylene glycol)	ChCl/DTG/PEG
DES7	Choline chloride/ 1,3-Diphenylguanidine /ethylene glycol	ChCl/DPG/EG
DES8	Choline chloride/ 1,3-Diphenylguanidine / Butyleneglycol	ChCl/DPG/BDO
DES9	Choline chloride/ 1,3-Diphenylguanidine / Poly(ethylene glycol)	ChCl/DPG/PEG

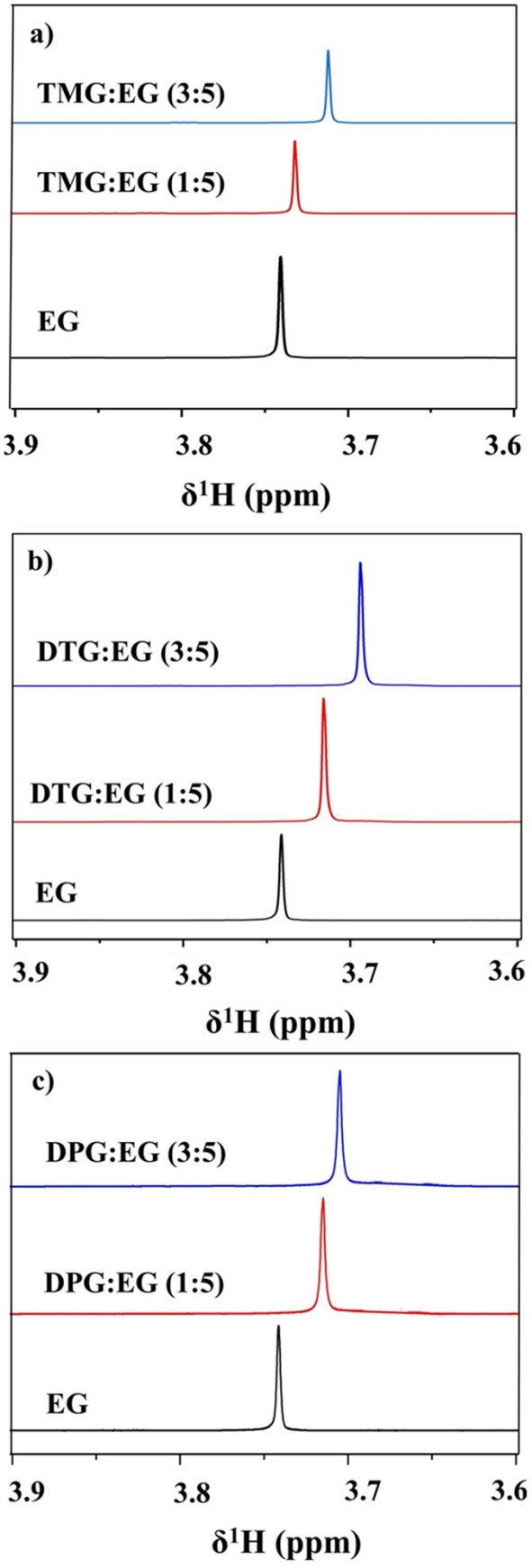

Fig. S1. ^{1}H NMR spectra of guanidine-based system.

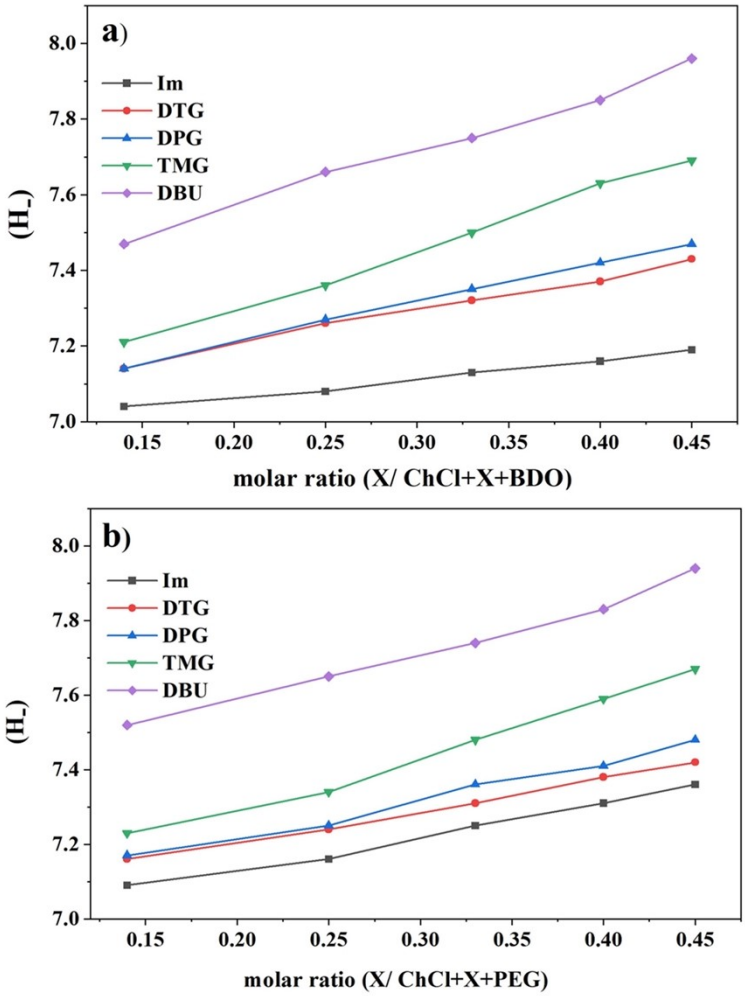

Fig. S2. a, b) The H- of various DESs when changing the molar ratio of components, X represents the alkaline component.

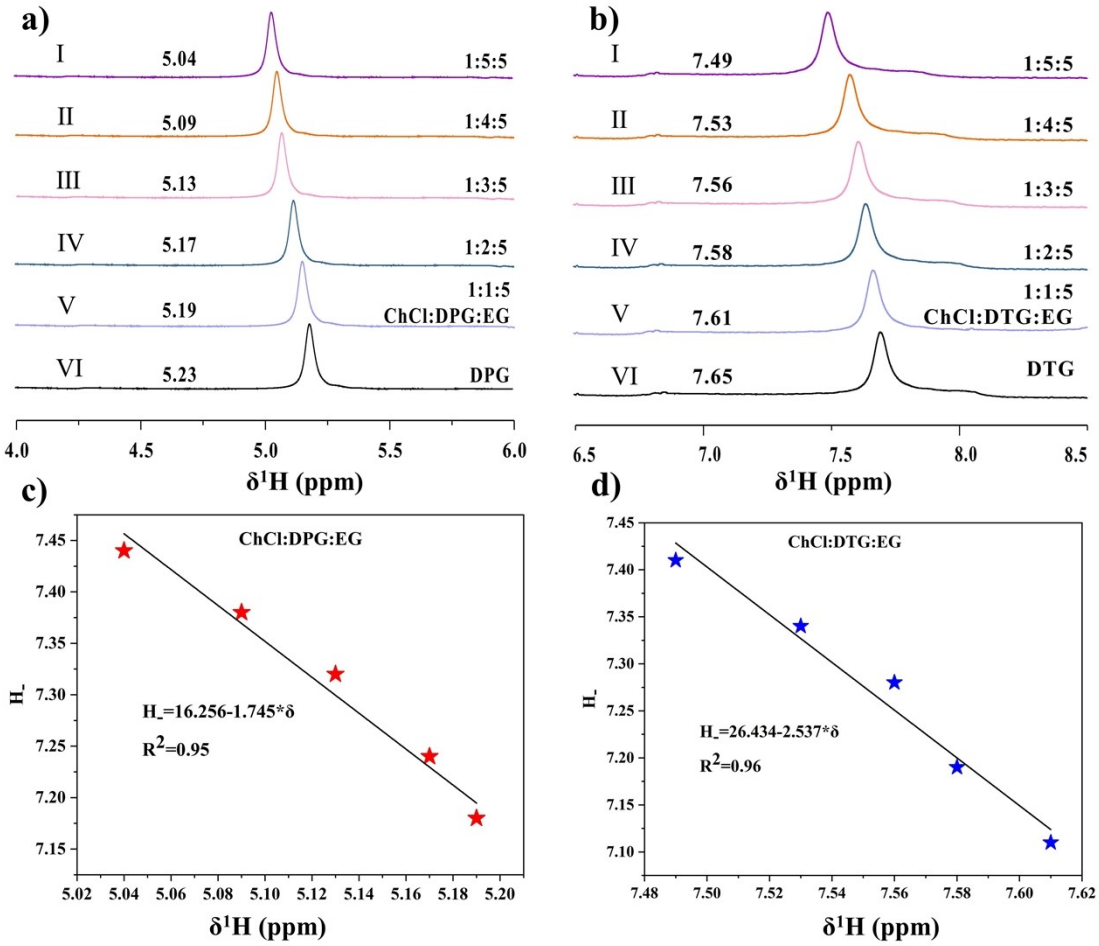

Fig. S3. a, b) Chemical shift of specific N-H protons of DPG and DTG molecules. c, d) the linear relationship between the chemical shift and H$_{-}$.

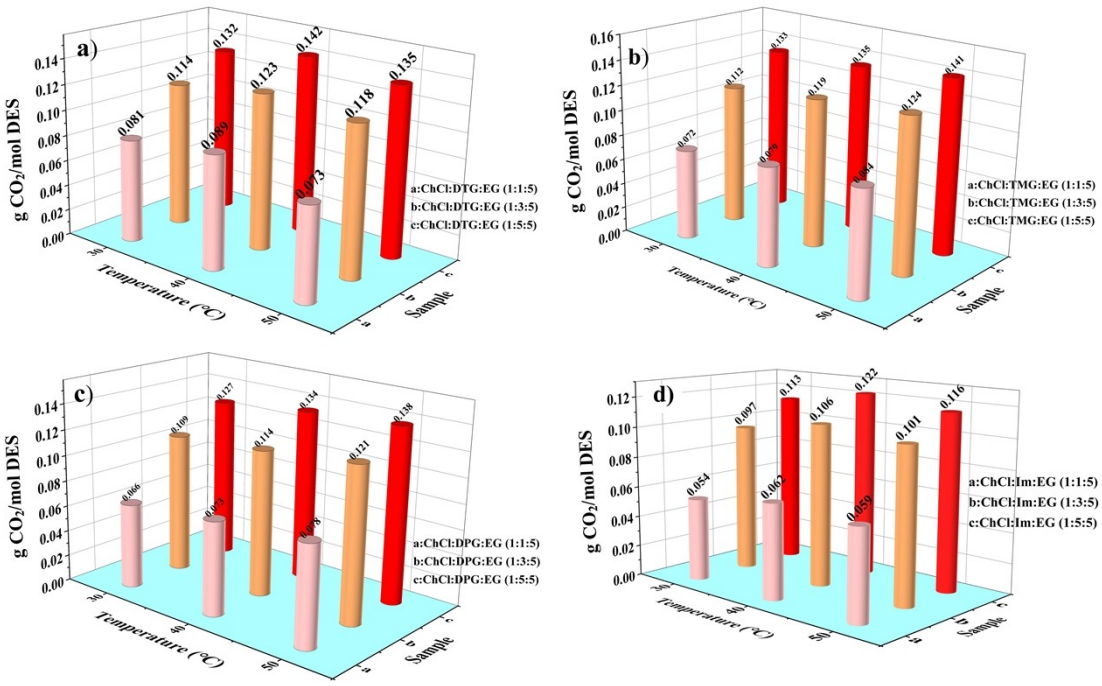

Fig. S4. CO_2 capture capacity of DESs with different molar ratio of components under different temperature (30 40 and 50 ℃).

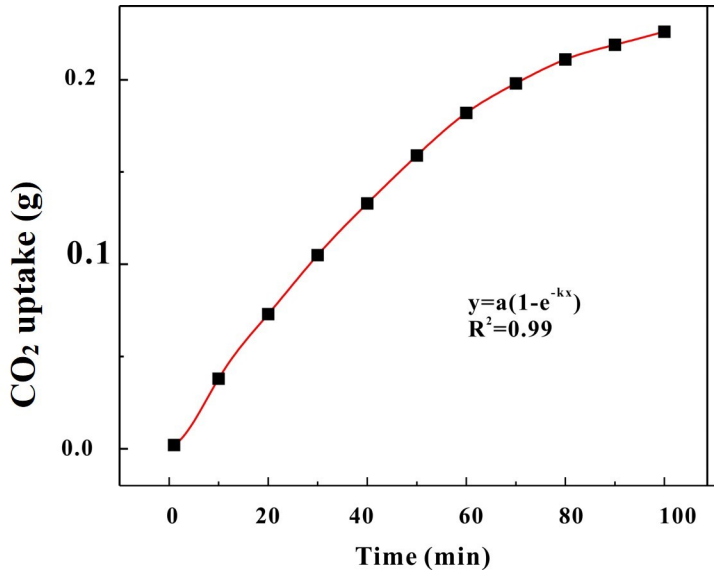

Fig. S5. CO_2 capture mass growth model.

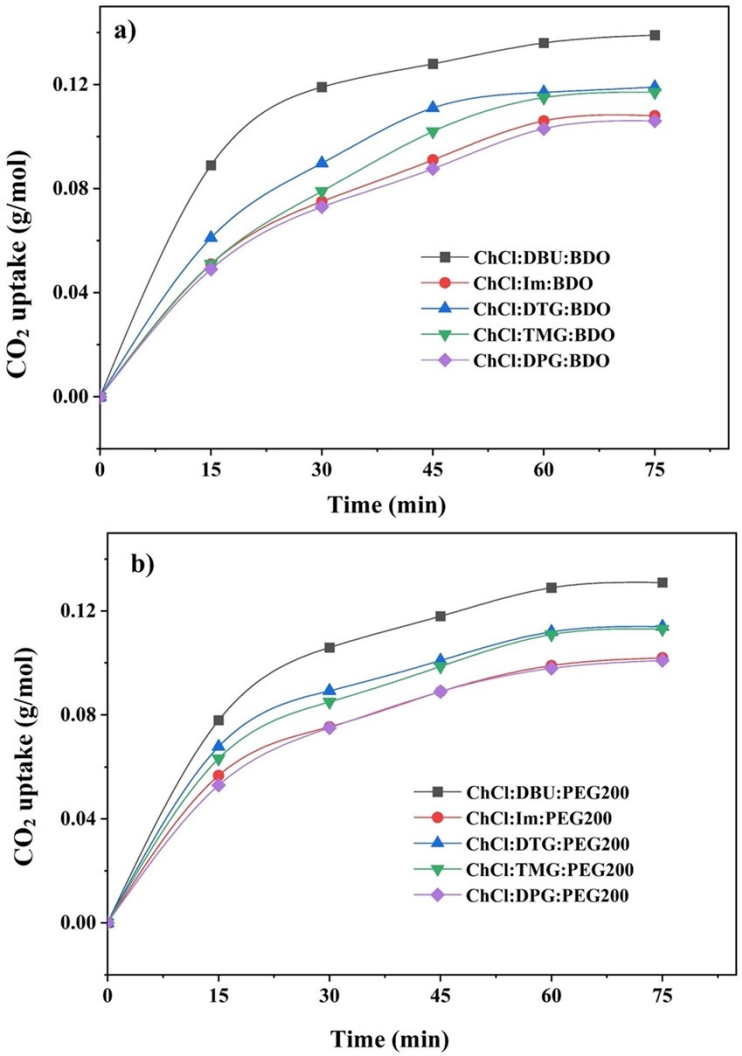

Fig. S6. Change the alcohol base composition, the CO_2 capture curves of various DESs with a molar ratio of control components of (1:3:5) at 40 ℃. a), b) are BDO and PEG 200 , respectively.

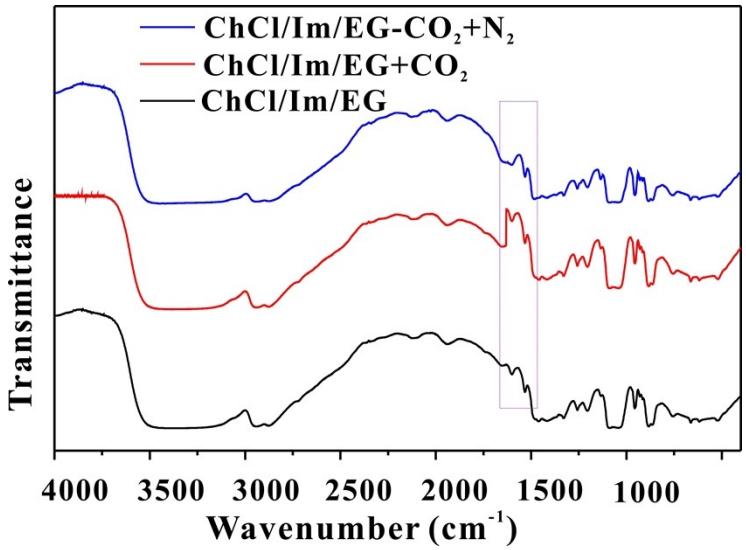

Fig. S7. FT-IR before and after CO_2 capture by Im-based DES.

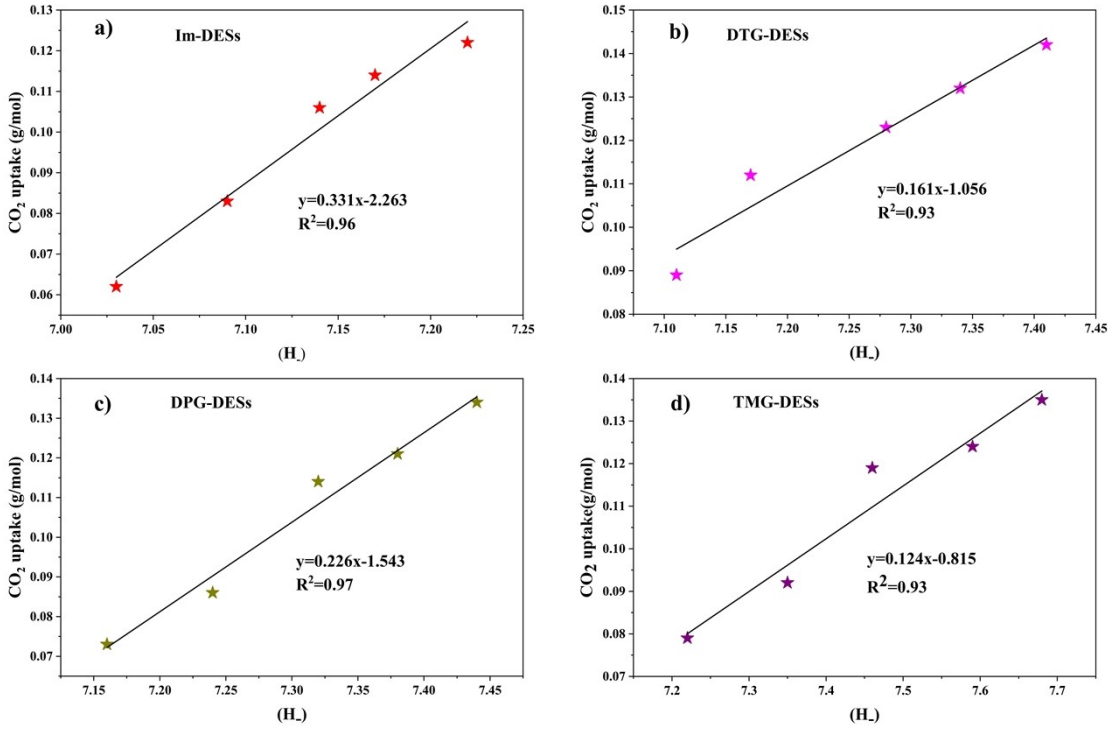

Fig. S8. The linear relationship between the H_- value of different DESs and the CO_2 capture amount, the reaction temperature is 40 ℃

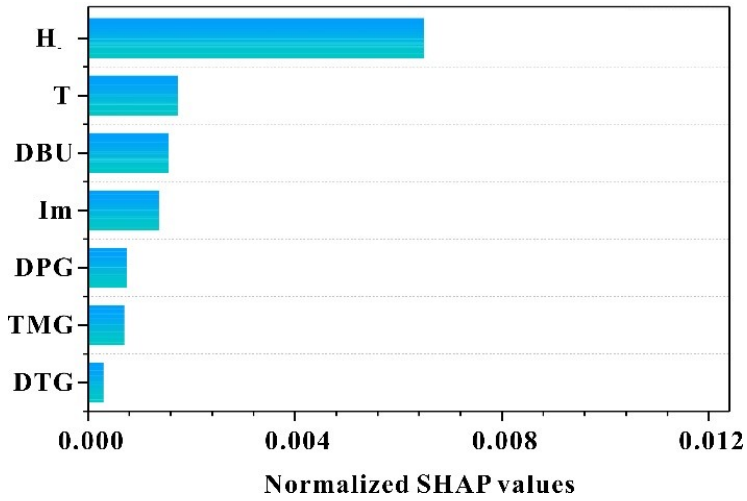

Fig. S9. SHAP feature importance for the testing data set of CO_2 capture in DESs.

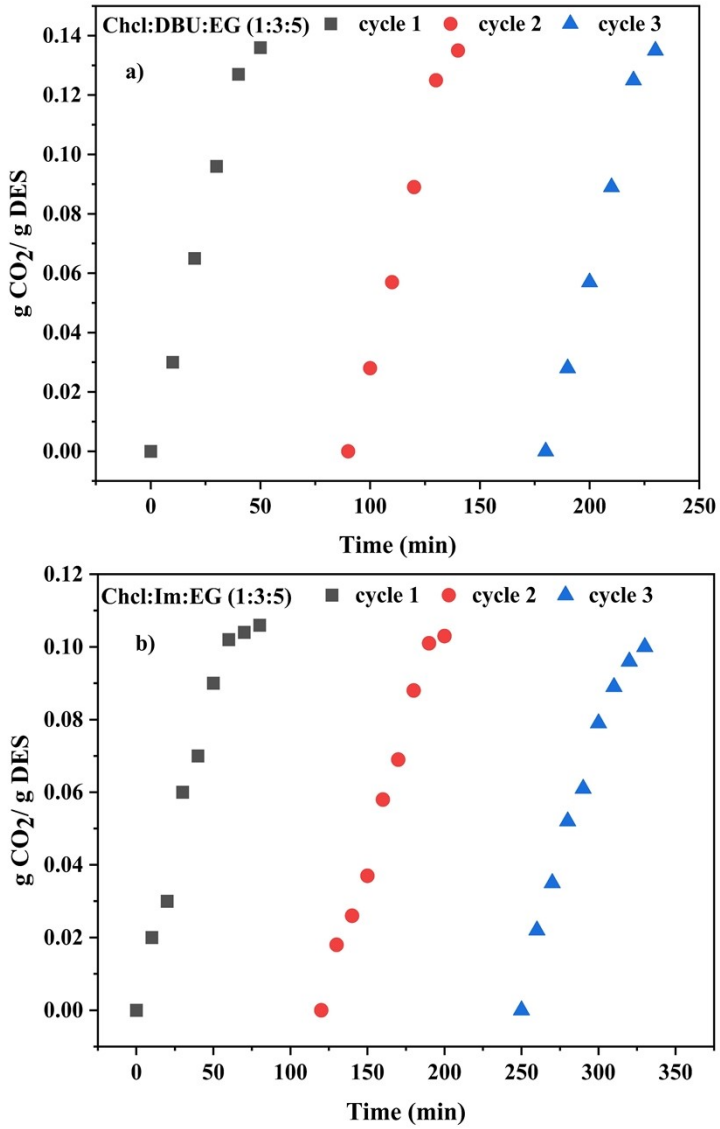

Fig. S10. CO_2 absorption/desorption cycles of two type DESs. a) the mole ratio is ChCl:DBU:EG (1:3:5), b) the mole ratio is ChCl:Im:EG (1:3:5). The desorption temperature is 80 ℃ under N_2 bubble condition.

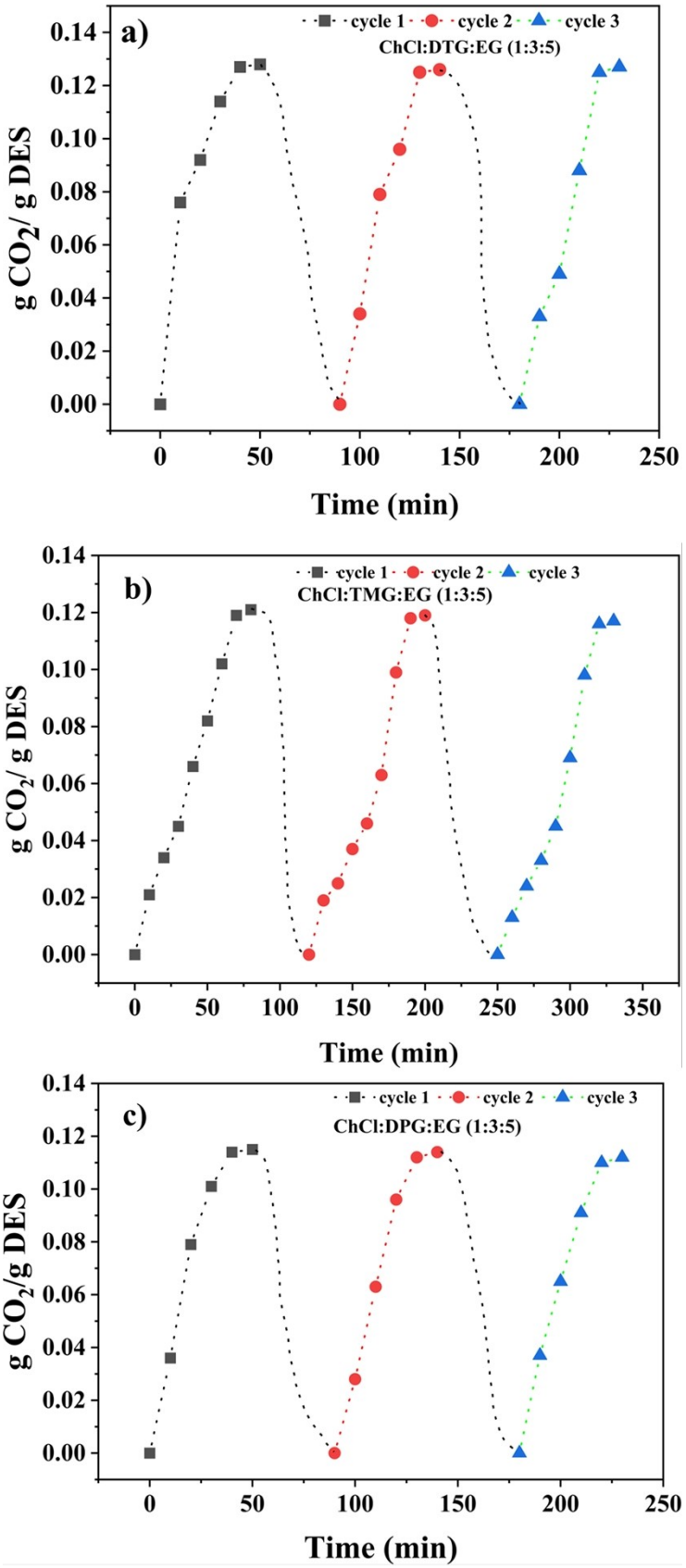

Fig. S11. CO_2 absorption/desorption cycles of three type DESs. a) the mole ratio is ChCl:DTG:EG (1:3:5), b) the mole ratio is ChCl:TMG:EG (1:3:5) and c) the mole ratio is ChCl:DPG:EG (1:3:5). The desorption temperature is 80 ℃ under N_2 bubble condition.

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