Supporting Information

Cost and economic analysis of amine solutions

The cost of the amine solution (*CA*, \$/min) in the biogas upgrading process is calculated by the price of the amines (*PA*, \$/dm^3) and the cycling rate of amines (*CCR*, dm^3/min) in the absorption unit, and the volatilization, corrosion and degradation of amines are not considered in the process. Chen et al. (2022) found that the cycling rate of amines can be calculated from the CO$_2$ flow rate (FR_{CO2}, mol CO$_2$/min) and the CO$_2$ absorption capacity of amines (A_{abs}, mol CO$_2$/dm^3), as shown in Eq. (S1).

$$CCR = \frac{FR_{CO_2}}{A_{abs}} \quad , \tag{S1}$$

In addition, Nwaoha et al. (2016) found that the CO$_2$ flow rate can be calculated from the simulated biogas flow rate (FR_{biogas}, mol/min), as shown in Eqs. (S2) and (S3).

$$FR_{biogas} = \frac{P_{abs}V_{biogas}}{RT_{abs}} \quad , \tag{S2}$$

$$FR_{CO_2} = FR_{biogas} \times x_{CO_2} \quad , \tag{S3}$$

where P_{abs} is the pressure during biogas upgrading (101.325 kPa), V_{biogas} is the flow rate of the simulated biogas (580 mL/min), R is the universal gas constant (8.314 kJ/kmol·K), T_{abs} is the temperature during biogas upgrading (298.15 K), and x_{CO2} is the molar ratio of CO$_2$ in the simulated biogas.

The cost price of the individual amine (*PI*, \$/kg) was obtained from the supplier's chemical catalog. And the price of amine per unit volume (*PA*, \$/dm^3) was calculated by Eq. (S4).

$$PA = \sum_{1}^{5}\left(PI_i \times n_i \times m_i\right) \quad , \tag{S4}$$

where n_i is the molar concentration of the amine in the solution (mol/dm^3), m_i is the molar mass of the amine (kg/mol), i refers to the five amines used in this paper.

The cost of the amine solution can then be calculated using Eq. (S5).

$$CA = PA \times CCR \quad , \tag{S5}$$

The CO$_2$ desorption heat cost of the amine solution can be calculated from the cycling rate of amines, regeneration rate, CO$_2$ desorption heat and energy cost, as shown in Eq. (S6). According to the US Energy Information Administration (EIA) announced that the industrial electricity consumption (*IEC*) in 2021 is 0.0726$ per kWh.

$$DC = A_{\text{abs}} \times CCR \times RR \times DH_{\text{CO}_2} \times IEC \quad , \tag{S6}$$

where *DC* is the cost of amine desorption ($/min), *RR* is the regeneration efficiency of amines (%), DH_{CO2} is the heat of CO_2 desorption of amine (kJ/mol CO_2).

In addition, the rich amine solution can continue to enter the absorption tower reaction after regeneration from the desorption unit, so it has a certain regeneration gain (*RG*, $/min), as shown in Eq. (S7).

$$RG = CA \times RR \quad , \tag{S7}$$

The final total relative energy consumption (*TRC*, $/min) is given by Eq. (S8).

$$TRC = DC + CA - RG \quad , \tag{S8}$$

Thermodynamic analysis

The total energy loss of aqueous amines (Q_{ene}) mainly includes the heat of CO_2 desorption (Q_{des}), the latent heat of vaporization of water (Q_{lat}) and the sensible heat of solvent (Q_{sen}), which can be calculated by the following equation (Eqs. (S9)–(S12)):

$$Q_{\text{ene}} = Q_{\text{des}} + Q_{\text{lat}} + Q_{\text{sen}} \quad , \tag{S9}$$

$$Q_{des} = m_{amine} \int_{\text{abs}}^{\text{des}} \Delta h_{\text{des}}(\alpha) d\alpha \quad , \tag{S10}$$

$$Q_{sen} = m_{amine}(C_{amine} + R_{solvent} C_{solvent} + \alpha_{des} C_{CO_2})(T_{des} - T_{abs}) \quad , \tag{S11}$$

$$P_{CO_2} = P \quad , \tag{S12}$$

where m_{amine} is the amount of amine required to capture 1 kg CO_2 (mol), α is the CO_2 loading (mol CO_2/mol amine), Δh_{des} is the CO_2 desorption enthalpy of the solvent (kJ/mol), R_{solvent} is the molar ratio of the remaining components of the fresh solvent to the amine, C_{amine}, C_{solvent} and C_{CO2} are the specific heat capacities of amine solvent and CO_2, respectively (kJ/(mol·K)), α_{des} is the CO_2 loading of the amine solution into the desorption column (mol CO_2/mol amine), m_{water} is the amount of water vapor produced per 1 kg of CO_2 desorbed (mol), and λ is the latent heat of vaporization of water (kJ/mol), which at 363.15K is 41 kJ/mol.

The method for calculating the heat of CO_2 desorption based on the Gibbs-Helmholtz equation (Eq. (S13)) was described in detail in a previous study and was mainly implemented by the reaction apparatus shown in Fig. 1.

$$\frac{d(\ln P_{CO_2})}{d(\frac{1}{T_{abs}})} = -\frac{\Delta h_{des}}{R}, \tag{S13}$$

where *R* is the ideal gas constant, P_{CO2} is the partial pressure of CO_2, and T_{abs} is the absorption temperature of the amine solution.

It was worth noting that the vaporized mass of water is not directly available, so it could be solved by Eqs. (S14)–(S16) (Oexmann et al., 2012; Conway et al., 2014; Nwaoha et al., 2017).

$$Q_{lat} = \lambda \frac{P_{H_2O\text{-}des}}{P_{CO_2\text{-}des}}, \tag{S14}$$

$$P_{H_2O\text{-}des} = x_{H_2O\text{-}des} P_{Saturation}, \tag{S15}$$

$$P_{CO_2\text{-}des} = P_{Atmospheric} - P_{Saturation}, \tag{S16}$$

where P_{H20} and P_{CO2} are the partial pressures of water and CO_2 in the desorption process (kPa), respectively, $P_{Saturation}$ is the saturation pressure of water in the desorption process (kPa), which can be obtained using the Antoine equation, and $X_{H2O\text{-}des}$ is the molar fraction of water in the desorption solution.

The energy consumption analysis is an important indicator used by amines for biogas upgrading and includes three main components: the heat of desorption of CO_2, the sensible heat of the solvent and the latent heat of vaporization of water. Where the CO_2 desorption heat (Q_{des}) was solved by the Gibbs-Helmholtz equation, the trend and data of sample absorption with P_{CO2} and T_{abs} were shown in Fig. 5 and the fitted curve was shown in Fig. 6, and the corresponding desorption heat can be found by Eq. (S13). The sensible heat (Q_{sen}) of the mixed solution during the heating process could be found by Eq. (S8), and the latent heat of vaporization of water (Q_{lat}) could be obtained by Eqs. (S14)–(S16), and the relevant data of MEA were obtained from the studies of Ye et al. (2019) and Kim et al. (2015).

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