Investigating best available technique for CO₂ chemical absorption: solvent selection based on empirical surrogate model and exergy loss

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Received: 30 January 2021 / Accepted: 13 June 2021 / Published online: 12 July 2021
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Abstract
The carbon dioxide concentration in the atmosphere has reached extremely high levels, generating environmental concerns. Unfortunately, despite the climate change, CO₂ is not included nowadays as a key environmental issue in Best Available Technique (BAT) reference documents (BREF). Industrially, the widespread industrial technology to capture CO₂ is the chemical absorption using aqueous monoethanolamine (MEA) at 30%wt, which is the basis of comparison for novel alternative techniques in the literature and seems a suitable candidate to be proposed as Best Available Technique. Nevertheless, there is an intense research to find alternative solvents that decrease the energy consumption for carbon capture and many solvents are claimed in the literature to outperform MEA. A novel empirical surrogate model and exergy balances are used to confirm that MEA is still the best candidate to be proposed as Best Available Technique. The surrogate model proposed in this study properly regresses the CO₂ gas liquid equilibrium data. The regressed parameters of the model are tabulated in this study for many aqueous alkanolamines and their mixtures, being the basis for computationally inexpensive chemical absorption column design. The surrogate model parameter considering the temperature is related with the chemical absorption energy and the consumed energy for solvent recovery. The obtained results show that none of the considered alkanolamine outperforms MEA in all the considered aspects, i.e. energy and solvent flowrate. MEA minimum flowrate is 15.62 mol solvent/mol gas and its heat of absorption regression parameter is − 27,745 J/mol. The proposed mathematical method is useful as a fast assessment for other novel alternatives that will be proposed in the future, providing energetically more efficient and cleaner technologies for CO₂ capture.

Graphic abstract

Keywords Minimum solvent flow rate · Energy parameter · Surrogate model · Aqueous alkanolamines · Post-combustion capture

Abbreviations
MEA Monoethanolamine
AMP 2-Amino-2-methyl-1-propanol
DEA Diethanolamine
Coal-fired power plants are the largest source of carbon dioxide emissions into the atmosphere and have a large operation window compared to other alkanolamines (Fig. 1). Nevertheless, in USA the physical absorption is the most relevant technique, using propylene carbonate or proprietary solvents such as Selexol, Rectisol. (Yamada 2021). Physical absorption avoids the energy consumption related to the reaction enthalpy during solvent regeneration, but the required solvent flow rate becomes impractical for low CO2 partial pressures such as for power plants’ flue gas (Fig. 2). According to Burnard (2020a), two main barriers must be overcome to spread the use of carbon capture: the high capital expenditure and lack of revenue generation for the low commercial value of CO2. Policy incentives and regulatory actions must be put in place urgently to overcome these two barriers.

The implementation and improvement of any industrial process requires a reference process considered as the Best Available Technique and in fact the chemical absorption with aqueous MEA has been taking this role for low partial pressure CO2 meanwhile for low partial pressure of CO2 MEA or proprietary aqueous amine solutions such as Cansolv, KS-1, etc. (Yamada 2021) are used. Mendez-Alvarez et al. (2016) pointed out that MEA and MDEA have a large operation window compared to other alkanolamines (Fig. 1).

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The implementation and improvement of any industrial process requires a reference process considered as the Best Available Technique and in fact the chemical absorption with aqueous MEA has been taking this role for low partial pressure CO2 capture for the last half century. European Best Practice Guidelines for CO2 Capture Technologies indicate that the 30% wt MEA solvent scrubbing process used in a number of large-carbon capture facilities is the chemical absorption: 90% of the carbon captured in large-carbon capture facilities in operation corresponds to the reaction enthalpy during solvent regeneration, but the required solvent flow rate becomes impractical for low CO2 partial pressures such as for power plants’ flue gas (Fig. 2). According to Burnard (2020a), two main barriers must be overcome to spread the use of carbon capture: the high capital expenditure and lack of revenue generation for the low commercial value of CO2. Policy incentives and regulatory actions must be put in place urgently to overcome these two barriers.

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industrial applications is considered the benchmark technology (CESAR 2018). However, despite climate change, CO₂ pollution is currently not included as a key environmental issue in the BREF documents (European Commission 2017).

The chemical absorption consists of capturing the gas by cycles of chemical absorption–desorption. Unfortunately, this process comes with a high recycling solvent flow rate and energy demand. Many reports point out that a MEA alkanolamine system to capture 90% CO₂ from a power plant flue gas can consume 30% of the power produced, which is translated into a CO₂ capture cost of 40–100 €/t CO₂ (Merkel et al. 2010). A more favourable scenario is obtained with an energy return of 85% when combined heat and power plants are retrofitted with post-combustion carbon capture (Morales-Mora et al. 2019) or the carbon capture unit also produces electricity (Li et al. 2018). An option to avoid the solvent recovery costs is to use the collected ammonia as fertilizer, e.g. Bonet-Ruiz et al. (2015) or He et al. (2017). Despite the drawbacks and improvements proposed, chemical absorption with alkanolamines has the greatest potential to reduce emissions (Figueroa et al. 2008).

**Alternative aqueous alkanolamines to MEA**

Alkanolamines are compounds that have both hydroxyl (-OH) and amino (-NH₂) groups. The basicity of the amino group allows the chemical capture of CO₂ due to its acidic character. Alkanolamines are the most common chemical absorbers for CO₂ capture. MEA chemical absorption is the dominant technology to industrially capture CO₂ from power plants exhaust gases up to now, but novel amine absorbents are expected to provide more efficient processes. There is an intense research to find alternative solvents that decrease the energy consumption of aqueous MEA solvent recovery and being less prone to degradation, e.g. Alaba et al. (2017). Besides of alternative aqueous alkanolamines, there are many other alternatives assessed in the literature such as alkanolamines in non-aqueous solvents, ionic liquids, amine functionalized deep eutectic solvents, enhanced with solid catalysts. Salting out effect suggests that physical absorption is not the only mechanism through which CO₂ is absorbed after full theoretical amine loading (Wanderley et al. 2018). However, the enhanced processes compare their advantages versus the aqueous alkanolamines, which are the most mature for treating flue gas. As the literature available is very extensive, this study focuses only on the aqueous alkanolamines because these are the basis of comparison for any other solvent used, and other alternatives will be assessed in future work.
There are many experimental CO₂ equilibrium data available for aqueous alkanolamines (Table 1) and some of their mixtures (Table 2). These data are not only useful to know the load of CO₂ on the alkanolamine but also to estimate the enthalpy of absorption using the simplified Gibbs–Helmholtz equation (Mathias 2016). Substituting the CO₂ fugacity by its partial pressure and assuming a low CO₂ fraction in the gas phase, then the enthalpy of absorption is estimated based on the variation of the CO₂ partial pressure with the inverse of the temperature (Eq. 1). A desorption parameter related with the desorption column heat duty is particularly useful for the ranking of alternative alkanolamines (Zhang et al. 2020a, b).

\[
\frac{d(\ln(P_{CO₂}))}{d(\frac{1}{T})} = \frac{\Delta H_{abs}}{R}
\]  

In the literature, there are many studies that claim certain aqueous alkanolamines are better than MEA. Besides the commercially available alkanolamines, the computer-aided molecular design is a powerful tool to propose novel solvents suitable for CO₂ capture (Papadopoulos et al. 2016). There is a large number of proposed solvents for CO₂ capture in the literature, for many of them there is some experimental gas liquid equilibrium or calorimetric data, for some of them there are pilot plant experiments but very few have been tested at larger capacity units and lengthier campaigns. Pilot plant and industrial operating data are not always published in the available literature although many studies are available.

Morgan et al. (2018) perform a study for a 10 t CO₂/h pilot plant, and Feron et al. (2017) run a pilot plant for 1000 h, and both provide a review of previous studies about MEA pilot plants. In pilot plant experiments, Akram et al. (2020) determined that using 40% MEA, the reboiler duty drops by up to 14% compared to the commonly used 30% MEA concentration. Enhanced packings, e.g. (Flechsig et al. 2018), and flexible operation of MEA columns, e.g. (Cormos et al. 2019), are also under study. In the literature, DEA (secondary amine) or MDEA (tertiary amine) are claimed to have a higher absorption capacity, higher resistance to corrosion and lower exergy lost than MEA (Chowdhury et al. 2018), and flexible operation of MEA columns, e.g. (Cormos et al. 2019), are also under study. In the literature, DEA (secondary amine) or MDEA (tertiary amine) are claimed to have a higher absorption capacity, higher resistance to corrosion and lower exergy lost than MEA (Chowdhury et al. 2018). The corrosion resistance and degradation to harmful compounds, e.g. nitrosamines (Mazari et al. 2019), are out the scope of the present study.

Although most of the attention at pilot scale is devoted to MEA, there are also some promising pilot plant studies about DEA/PZ for CO₂ rich gases, e.g. Dubois et al. (2017) or Laribi et al. (2019), or about MEA/DEA at ratio 4:1, Idem et al. (2006). Śpiewak et al. (2015) claim the use of AMP/Pz based on process development unit runs; later on, pilot plant runs were performed with MEA solutions as a recognized baseline solvent suitable for future comparative purposes (Stec et al. 2016) and nowadays the pilot plant study focuses on the methanation of the amine-absorbed CO₂ (Chwola et al. 2020). Nwaoha et al. (2019) successfully substituted the toxic PZ by 1,5-diamino-2-methylpentane (DA2MP) in pilot plant experiments for the industrially used MDEA/PZ mixture.

Many other aqueous alkanolamines have been proposed based on laboratory experiments. For example, Barzaghi et al. (2019) determine experimentally, using pure CO₂, that AMP and its mixtures reach higher loads than MEA and the enthalpy of absorption is smaller according to simplified Gibbs–Helmholtz equation. Many studies identify alkanolamines with a lower absorption enthalpy than MEA, e.g. for DEEA and DMEA are −47.0 and −48.6 kJ/mol, respectively (Xiao et al. 2016) and for 1DMA2P and MEA are −31.7 and −84.3 kJ/mol, respectively (Liu et al. 2017). It is also possible to combine several types of alkanolamines, the purpose of the combination being to take advantage of each compound, e.g. (Xiao et al. 2021). Gómez-Diaz et al. (2021) prove that DMEA/MEA mixture load does not depend on its ratio and DMEA has a faster reaction rate. Therefore, although the MEA is recognized as a proven suitable solvent for CO₂ capture at large scale, many promising alternatives are claimed in the literature.

Process optimization and modification based on rigorous simulations show that the energy consumption of a reference MEA chemical absorption of CO₂ can be decreased; both for MEA or aqueous alkanolamines mixtures are attained similar regeneration reductions between 5 and 40% (Hosseini-Ardali et al. 2020). There are many simulation results that point out some aqueous alkanolamine mixtures outperforming MEA, not only from the energy point of view but also due to their higher stability, e.g. MDEA/PZ. Among the aqueous alkanolamines, some of them present the advantage to produce a phase split of the rich solvent stream (Papadopoulos et al. 2019). The phase poor in CO₂ is recycled directly to the absorption column, decreasing the flow rate of rich CO₂ phase fed to the distillation column. Examples of solvents with phase change behaviour are N,N-dimethylcyclohexylamine (DMCA), methylecyclohexylamine (MCA) and 2-amino-2-methyl-1-propanol (AMP) (Tzirakis et al. 2019), or mixtures such as MAPA/DEEA 3-(methylamino)propylamine/2-(diethylamino)ethanol. The computer-aided molecular design allows the screening and proposal of novel solvents suitable for CO₂ absorption (Papadopoulos et al. 2020a). A novel very promising solvent proposed by computer-aided molecular design is the S1N (N1-cyclohexylpropane-1,3-diamine), for which laboratory experiments corroborate its suitability when mixed with other alkanolamines, e.g. DMCA, (Papadopoulos et al. 2020b). The phase split solvent is proved to be advisable from economic (Zarogiannis et al. 2020a), environmental
| Alkanolamine | Temperature, K | CO₂ partial pressure, kPa | Amine concentration, % | CO₂ loading, α | References |
|-------------|---------------|---------------------------|-----------------------|--------------|-----------|
| AMP         | 298–328       | 0.41–1.449                | 23.5–46.0             | 0.19–1.1     | Dash et al. (2011) |
|             | 313–353       | 3.90–363.6                | 30                    | 0.28–0.9     | Seo and Hong (1996) |
|             | 313.2         | 1.25–144                  | 28                    | 0.4–0.9      | Roberts and Mather (1988) |
|             | 313.2         | 0.89–151.9                | 28                    | 0.4–0.9      | Yang et al. (2010) |
|             | 313, 343      | 0.16–5,279                | 18.8                  | 0.03–1.65    | Teng and Mather (1990) |
|             | 313–393       | 6–983.5                   | 30                    | 0–0.97       | Tong et al. (2012) |
|             | 303–328       | 0.31–1,472                | 40, 50                | 0.24–1.04    | Dash et al. (2011) |
|             | 293–353       | 1.59–94                   | 18.76, 28.14          | 0.13–0.94    | Tontiwachwuthikul et al. (1991) |
| AEEA        | 303–323       | 1.11–794.67               | 15                    | 0.060–1.407  | Guo et al. (2013) |
| DEA         | 313–353       | 4.85–357.30               | 30                    | 0.40–0.73    | Seo and Hong (1996) |
|             | 298–348       | 4.85–357.30               | 47.78                 | 0.0–1.09     | Sidi-Boumedine et al. (2004) |
|             | 323–366       | 0.4–3,798.00              | 25                    | 0.10–1.13    | Barreau et al. (2006) |
|             | 323           | 7.00–3,370.00             | 19.2                  | 0.45–1.13    | Jong et al. (1972) |
|             | 338.5–366.9   | 32.00–767.00              | 25                    | 0.40–0.79    | Lawson and Garst (1976) |
| DGA         | 323–373       | 1.58–4,720                | 60                    | 0.13–0.62    | Martin et al. (1978) |
| DIPA        | 313–343       | 107–4,064                 | 45                    | 0.52–1.05    | Hagtalab et al. (2014) |
|             | 313–343       | 91.20–3,826.6             | 30                    | 0.89–1.14    | Hagtalab and Talavaki (2017) |
|             | 313–373       | 2.7–5,888                 | 33.63                 | 0.07–1.11    | Isaacs et al. (1977) |
| MDEA        | 298–373       | 0.78–140.40               | 50                    | 0.01–0.49    | Park and Sandall (2001) |
|             | 298–348       | 2.70–559.50               | 48.88, 25.73          | 0.0–1.30     | Sidi-Boumedine et al. (2004) |
|             | 297.7         | 0.02–1.64                 | 23.63                 | 0.02–0.26    | Lemoine et al. (2000) |
|             | 313           | 0.18–92.80                | 22.9                  | 0.04–0.84    | Austgen et al. (1991) |
|             | 313           | 0.28–8.90                 | 22.9                  | 0.06–0.80    | Chung et al. (2010) |
|             | 323           | 6.00–434.00               | 50                    | 0.10–0.89    | Dicko et al. (2010) |
| MEA         | 303–323       | 0.90–355.90               | 48.88, 25.73          | 0.35–1.16    | Kumar and Kundu (2012) |
|             | 303–353       | 0.50–65                   | 12.00–15.00           | 0.017–0.577  | Tzirakis et al. (2019) |
| PZ          | 313–343       | 0.03–40.00                | 4.7                   | 0.16–0.96    | Bishnoi and Rochelle (2000) |
|             | 313           | 451–3948                  | 14.7                  | 0.32–0.72    | Hagtalab et al. (2014) |
|             | 354–464.8     | 28–2,583                  | 29.80–40.59           | 0.23–0.45    | Xu and Rochelle (2011) |
|             | 313–343       | 29–40,200                 | 4.7                   | 0.16–0.96    | Bishnoi and Rochelle (2000) |
|             | 313           | 5,800–7,500               | 15–60                 | 0.34–0.86    | Nguyen et al. (2010) |
| TEA         | 313–353       | 1.43–153.40               | 26.5                  | 0.03–0.53    | Chung et al. (2010) |
| AMMONIA     | 335–395       | 0.01–1,000                | 20.4                  | 1           | Lu et al. (2017) |
| AMP + PZ    | 293.15–323.15 | 0.127–140.4              | AMP: 8.9–3/PZ: 0.87–8 | 0.1511–0.9405 | Jahanagiri and Hassakeniadeh (2019) |
| DEA + AMP   | 313.15–373.15 | 22–2,838                 | DEA: 20–25/AMP: 5–10  | 0.3370–1.2   | Murrieta-Guevara et al. (1998) |
| DEA + MDEA  | 313.15–393.15 | 0.4–2,833.6              | DEA: 10–32.5/MDEA: 10–35 | 0.038–1.119  | Murrieta-Guevara et al. (1998) |
| DIPA + AEEA | 313.15–343.15 | 105–3,819.7              | DIPA: 20.25/AEEA: 5–10 | 0.5837–1.251 | Hagtalab and Talavaki (2017) |
| MDEA + MEA  | 313.15–373.15 | 1.12–2,080               | MDEA: 12–24/MEA: 6–18 | 0.1880–1.015 | Li and Shan (1992) |
| MDEA + PZ   | 313–375.15    | 0.033–95.78              | MDEA: 22.6–47.6/PZ: 0.4–21.3 | 0.027–0.37  | Ghalib et al. (2017) |
| MEA + DAP   | 315.15–333.15 | 13.24–215.46             | MEA: 10–12.5/DAP: 2.5–5 | 0.220–0.711  | Khodadadi et al. (2019) |
| DIPA + AMP + PZ | 313.15–343.15 | 112.9–3,709.7             | DIPA: 24–36/AMP: 7–13/PZ: 2–8 | 0.5020–1.091 | Hagtalab and Talavaki (2017) |
(Shavalieva et al. 2021) and operational (resilient to disturbances) (Zarogiannis et al. 2020b) points of view.

This study proposes a fast method that, based solely on experimental data of vapour–liquid equilibrium of CO₂ gas, compares alternatives with MEA in terms of required flow-rate and energy consumption. This method has been applied to several alkanolamines whose experimental gas liquid equilibrium data are available in the literature (DEA, TEA, DIPA, MDEA, DGA, AEEA, PZ, AMP, NH₃) and some of their mixtures (Table 1). Table 1 compiles the operating conditions of temperature, CO₂ partial pressure, amine concentration and CO₂ load in which each experimental study has been performed. Some aqueous alkanolamines have been extensively studied in the literature by many authors and for some others there is only a single study. The aqueous alkanolamines for which there are not enough experimental points according to the number of model parameters to be properly regressed have not been considered. Although an exhaustive literature review retrieving a vast number of experimental data points has been performed in this study, some interesting alkanolamines may have been missed. However, the proposed method is fast and simple enough to be applied to them in further studies.

**Method**

**Standard model equations for alkanolamines gas liquid equilibrium**

Assessment of alkanolamine absorbents capture performance is crucial to decrease the carbon footprint of many processes. There are several activity coefficients models useful for electrolyte mixtures such as Pitzer or ElectNRTL that are implemented in commercial simulation software, e.g. Aspen Plus®. Aspen Plus® allows also to use the MSE or Aqueous models based on Debye–Hückel model from the software specific for electrolytes OLI®. MSE model implemented in OLI® allows very accurate calculations for electrolyte systems but is not easy to generate a novel alkanolamine at user level. The activity models such as ElectNRTL require to determine many tens of unknown parameters before modelling. Figure 3 shows that the absorption operation window without solids or vapours using the parameters readily implemented in ElectNRTL of Aspen Plus® appreciably differs from the operation window calculated using MSE of OLI® (Mendez-Alvarez et al. 2016). The activity coefficients complex models are not suitable when limited experimental data are available, then the regressed parameters have little physical relevance, then this drawback is overcome using simplified shortcut methods including the simplified Gibbs–Helmholtz equation (Jiang et al. 2018). OLI® software is used in the present study to solve the exergy balances.

The present study proposes a simple and effective surrogate model that with a small number of parameters and based solely on few CO₂ gas liquid equilibrium experimental data are able to estimate an energy parameter and minimum solvent flow rate required for each solvent. The proposed method is intended to simplify the assessment of CO₂ capture performance, but the regressed CO₂ vapour liquid equilibrium model also is useful for the absorption column design.

**Surrogate model equations for gas liquid equilibrium**

Plesu et al. (2018) proposed an equation able to accurately represent the CO₂ gas-liquid equilibrium on aqueous MEA available in the literature, in the CO₂ load range between 0.2 and 0.4 (Eq. 2). The equation neglects the alkanolamine mass
percentage and a value close to the calorimetric determined enthalpy of absorption for MEA for the temperature dependent term is obtained, i.e. $\sim -88 \text{kJ/mol} \times -84 \text{kJ/mol}$. The equation has a poor regression of the experimental data at high temperatures and loads, i.e. when the CO$_2$ partial pressure is higher and deviates from ideality.

$$\ln P_{\text{CO}_2} = 32.95 + 14.96 (\alpha) - 88.081 \left( \frac{1}{RT} \right)$$  \hspace{1cm} (2)

This empirical correlation that relates the molar composition and the temperature with the CO$_2$ partial pressure is computationally inexpensive due to its simplicity, but the main drawback is its low dimensionality which translates into its limited validity range (Carranza-Abaid et al. 2020). In this study, the dimensionality and validity range of the equation is extended. The extension of the previous equation to further alkanolamines including mixtures implies the addition of two parameters to the previous equation, which depend on the alkanolamine mass percentage (Eq. 3). A broad range of CO$_2$ gas liquid equilibrium data is regressed. The model assumes that the dependence of the CO$_2$ partial pressure on the inverse of the temperature does not depend on the composition. This rough approach is applied for sake of simplicity. The absorption energy term of the equation loses its physical meaning and does not correspond to the experimental calorimetric absorption energy. Nevertheless, it is expected to be an averaged value of the enthalpy of absorption providing an insight of the energy consumption for the rich solvent recovery.

$$\text{LN } P_{\text{CO}_2} = \text{Intersection} + X_1 \cdot (\alpha) + \Delta H_{\text{abs}} \cdot \left( \frac{1}{RT} \right) + S1 + S2$$  \hspace{1cm} (3)

$$S1 = \sum_{m=0}^{\text{no alk}} X_n \cdot (\% \text{wt Alkanolamine})$$  \hspace{1cm} (4)

$$S2 = \sum_{m=0}^{\text{no alk}} X_m \cdot (\% \text{wt Alkanolamine} \cdot (\alpha))$$  \hspace{1cm} (5)

In this proposed model equation, the partial pressure of CO$_2$ ($P_{\text{CO}_2}$) is expressed in Pa, the heat absorption in J/mol ($\Delta H_{\text{abs}}$), the gas constant ($R$) is 8.314 m$^3$-Pa/(K-mol), temperature ($T$) is measured in Kelvin, $X_i$ are the regressed parameters of the surrogate model, %wt Alkanolamine is the amine mass percentage and “$\alpha$” is the CO$_2$ load (expressed as the ratio of mols CO$_2$ absorbed divided by mols alkanolamines necessary for the absorption). The equation is not only providing insights on the energy consumption according to the $\Delta H_{\text{abs}}$ term but also on the solvent required according to the load term (Eq. 6).

$$\frac{d(\ln P_{\text{CO}_2})}{d\alpha} = X1 + \sum (X_m \cdot \% \text{wt Alkanolamine})$$  \hspace{1cm} (6)

**Mass and exergy balances**

CO$_2$ absorption process scheme is composed mainly of two main sections: the CO$_2$ absorption and the solvent recovery (Fig. 4). The main unit of the process is the absorption column where the spontaneous absorption of CO$_2$ towards the equilibrium takes place. Therefore, in this section there is no appreciable energy consumption and usually operates at a rather low temperature around 40°C to promote the CO$_2$ absorption. The energy is mainly consumed in the solvent recovery section. The solvent recovery is not spontaneous, and the main energy of the process is consumed at a distillation column reboiler to reverse the CO$_2$ absorption. The flowsheet is completed with other unit operations such as heat exchangers for heat recovery and a decanter in case of phase split. Nevertheless, for sake of simplicity, the present study focuses only on the minimum flow rate of solvent to the absorption column.

The main streams considered in the mass balances are pointed up in accordance with Fig. 4. The carbon is captured from the exhaust gases (stream 1) collecting a treated gas stream (stream 2) at the top of the absorption column. An aqueous stream containing the alkanolamine is fed at the top of the absorption column, generating a stream of solvent enriched in CO$_2$ (stream 5). The CO$_2$-rich solvent is fed to a distillation column whose purpose is to separate pure CO$_2$ collected by the distillate (stream 6) and the CO$_2$ lean solvent stream (stream 4) collected at the bottoms and recycled to the absorption column. The input exhaust gas stream generated from the combustion with an air excess is composed in volume by 79% N$_2$, x % CO$_2$ and (21-x) % O$_2$, where in this study a x = 10% vol CO$_2$ is assumed.

The minimum solvent flow rate is calculated according to the method proposed by Sala et al (2014) assuming...
an infinite number of stages. Assuming infinite number of equilibrium stages for the absorption column, at absorption column top, CO₂ concentration in the treated exhaust gases output (stream 3) is in equilibrium with the CO₂ concentration present in the lean solvent output stream from the distillation column (stream 4). On the other hand, at absorption column bottoms, the feed exhaust gas to the absorption column is in equilibrium with the concentration of CO₂ present in CO₂ rich solvent. Contrary to Sala et al (2014) where the equilibrium was retrieved from a model, in this study, both CO₂ equilibrium information is retrieved from literature experimental equilibrium data. N₂ and O₂ are not absorbed, and therefore, their flow rates at the input (stream 1) and output (stream 2) gas streams are the same.

For a given concentration of solvent in water, all flow rates and compositions of the streams are calculated according to the mass balances described in more detail in the following section. The entropy and enthalpy of these streams required to calculate the exergy balances are determined with OLI® software.

**Mass balances**

The mass balances are solved assuming a calculation basis of 100 mol/s for the exhaust gas feed to the system with a 5 to 25% in volume CO₂ and a recovery of 70% of CO₂. Most of the studies use a recovery of 90%, but this is an artificial cap (Burnard 2020b). The process is operated at 1 atm, and all the streams are at this pressure.

According to the Dalton Law, the partial pressure of CO₂ at the exhaust gas is calculated. The CO₂ load (relationship between the composition of CO₂ in a determined stream and total alkanolamine—Eq. 7) in the liquid stream (stream 5) in equilibrium with the input exhaust gas is retrieved from experimental equilibrium data.

\[
\alpha_5 = \frac{w_{5,\text{CO}_2}}{w_{\text{ALK}}^T} = X1
\]  

A similar procedure is applied to the top of the absorption column. Establishing a CO₂ recovery of 70%, the amount of CO₂ that leaves the system can be assessed. The air is not absorbed. According to the Dalton Law, the CO₂ partial pressure at the gas phase at the top of the column is also calculated. According to the experimental data of CO₂ equilibrium, the load in the liquid phase at the top of the column is determined (Eq. 8).

\[
\alpha_4 = \frac{w_{4,\text{CO}_2}}{w_{\text{ALK}}^T} = X2
\]

The alkanolamine flow rate in the lean and rich solvent streams is the same and therefore with the previous CO₂ load expressions and the CO₂ mass balance, the molar minimum flowrate of alkanolamine is calculated (Eq. 9). Notice that the minimum alkanolamine flowrate depends on the difference of load values between the top and bottom of the adsorption column. The mass percentage of alkanolamine in the aqueous solution provides the water flow rate (Eq. 10).

\[
w_{\text{ALK}}^T = \frac{w_{5,\text{CO}_2} - w_{4,\text{CO}_2}}{\alpha_5 - \alpha_4} = \frac{w_{6,\text{CO}_2}}{\alpha_5 - \alpha_4}
\]

\[
w_{\text{water}}^T = w_{\text{ALK}}^T \cdot \frac{MW(\text{ALK})}{MW(\text{water})} \cdot \frac{(100 - x) \text{ g water}}{\text{g ALK}}
\]

**Results**

Many of the main aqueous alkanolamines studied in the literature for CO₂ absorption are critically compared and ranked according to the minimum solvent flow rate required and an energetic parameter provided by the proposed model regression. The literature review indicates that the benchmark process used in the literature to compare any novel exhaust gases CO₂ capture technology is the chemical absorption with MEA. It is expected that the present study results help to decide if the MEA absorption is a proper candidate to be proposed as the Best Available Technique (BAT). When a process is proposed as BAT in the BREFs does not imply that there are no other more efficient or advantageous techniques. The BAT process defines a base line of comparison for any novel technique implemented that should be at least better than the BAT. It is expected that the present results provide some light on the carbon capture and help to include the CO₂ as contaminant in the BREFs.

**Surrogated model**

The surrogate model proposed (Eq. 3) correlates the CO₂ gas liquid experimental data in general with good correlation coefficients (Tables 3, 4 and 5). However, in some cases such as for TEA or MEA + DAP the correlation is worse. Table 4 presents the regressed parameters for single aqueous alkanolamines and Tables 5 and 6 for binary...
and ternary mixtures. For some alkanolamines mixtures, there are not enough experimental data at different conditions to fit all the model parameters. The Pareto diagram is presented in Fig. 5, showing a random scatter of points at both sides of the diagonal, proving that the model is able to regress properly the gas liquid equilibrium experimental data for all the alkanolamines. Although more parameters would be included in the model, small deviations from the model are unavoidable due to experimental errors.

### Regeneration stream flow rate

The minimum solvent stream flow rate determines the pumping energy and cost related to the equipment and piping diameters. According to the minimum solvent flow rate results, MEA is on a top position together with ammonia (Table 6). DGA is also at a top position, but it is important to notice that no experimental data for 30% alkanolamine were available for DGA and a 60% mass fraction is used in
this case (Table 3). NH₃ was also industrially used, but its volatility discouraged its use. Therefore, MEA is a suitable alkanolamine from the required flow rate point of view for diluted exhaust gases absorption. There are some alkanolamines that are able to reach a higher CO₂ load than MEA but in this case what is important is the CO₂ load difference corresponding to the gas liquid equilibrium when the partial pressure of CO₂ varies. The obtained results depend on the CO₂ gas concentration and recovery and thus, higher CO₂ gas concentrations or recoveries affect the calculated minimum flow rate and the alkanolamines ranking. The minimum flow rate of 30% MEA aqueous solution to capture the CO₂ from an exhaust gas with 25% vol CO₂ is lower than the flowrate of any other assessed alkanolamines to capture a 5% vol CO₂. The exception is the volatile NH₃ and the DGA, whose experimental data are only available for 60% DGA aqueous solution.

It is expected that a lower minimum solvent flowrate is attainable when the parameters associated with the load of the surrogated model become higher. Figure 6 shows that for 30% alkanolamine and higher concentrations, the lowest minimum solvent flowrate corresponds to the use of MEA. Although being the worst choice according to Table 6, TEA seems also a good option for solvents with high alkanolamine concentrations but due to its low correlation coefficient further experimental research of gas liquid equilibrium is required. The results show that PZ performs better when its mass percentage is low but notice also that PZ regressed parameters provide negative slope values for high % Alkanolamines, which is physically unfeasible.

**Energy consumption**

Two different approaches are applied in this section for exhaust gases with 10% CO₂ content to identify solvent alternatives outperforming MEA:

- the first approach is based on an exergy balance of the absorption column at 30 °C based on the previous mass balance results and the thermodynamic parameters retrieved from OLI® software;
- the second approach is based on the regression parameters of the proposed surrogate model.

The energy consumption of the carbon capture process by chemical absorption is expected to be related with the exergy lost in the absorption column. The previously mass balances solved for a minimum solvent flow rate are used to perform the exergy balance. The exergy balance on the absorption column depicts some alkanolamines showing better behaviour than MEA (Table 7), for instance the DEA. In case of DPTA, using the experimental equilibrium data where the process takes places spontaneously,
the value of the exergy is positive which is not in agreement with the fact that the process should be spontaneous. Therefore, thermodynamic parameters implemented in commercial simulation software must be used with caution. Nevertheless, again MEA is in a rather favourable result.

Another approach to determine some energy favourable alkanolamines is based on the regressed surrogated model. According to the surrogated model, for the energy the focus is placed on the temperature-dependent parameters as it is expected to provide some hints about the energy consumption, although its value does not correspond to the absorption enthalpy. For instance, MEA energy regressed parameter has a value of $-28 \text{kJ/mol}$ versus the $-84 \text{kJ/mol}$ enthalpy of absorption (Table 8). The energy regressed parameters are ranked in Table 9, showing that MEA has a lower more favourable value than the other alkanolamines, which is in agreement with the industrial practice. Only the PZ + AMP mixture would have a more favourable parameter. The Pareto diagram between the regressed and the experimental CO$_2$ gas liquid equilibrium for PZ + AMP mixture is shown in Fig. 7, indicating reasonable agreement between the experimental data with the regressed equation (Eqs. 11, 12 and 13).

Although AMP and PZ solvents show worse results at the level of exergy of absorption, when mixed together, they show the best results.
For this mixture, if we assume a total solvent concentration of 30% (23% AMP and 7% PZ), the regeneration flow is 40.15 mol solvent/mol gaseous input, a value much higher as for MEA solvent of 15.62 mol solvent/mol gaseous input (Table 6). Therefore, despite its low value at heat absorption levels, its minimum solvent flowrate is quite high.

The surrogated model including the S1 and S2 parameters has a great influence on the value of the energy regressed parameter, and therefore, the regressions are repeated using only the S1 parameter for single aqueous alkanolamines (Table 9). Without the S2 parameter then the regression correlation of alkanolamines mixtures is rather poor and therefore not performed. The model is not able to regress all the data, and therefore, the regression is performed only in the linearized range of experimental points that is indicated in the table. Although the regressed energy parameter for MEA is still far away from its heat of absorption, the ranking is still showing that MEA is very favourable. In the case of DGA, using S1 and S2 in the surrogated model leads to unfavourable results, when only S1 is considered then its energy and load regression parameters are more favourable than MEA. Therefore, further experimental gas liquid equilibrium studies on DGA would be interesting to find in which operating conditions DGA could be more advantageous than MEA.

The surrogate model provides a very good regression of experimental CO₂ gas liquid equilibrium, but the energy parameter has only physical meaning for systems at constant composition. However, the lower relative values for MEA in all the regressions indicate that MEA is a good choice, which is in agreement with its nowadays industrial use. Several regressions at different compositions are required for a more precise energy assessment.

Hence, considering both the exergy assessment and the surrogate model parameters, the obtained results indicate that MEA is a suitable choice as benchmark from the energy point of view, although some alkanolamines could outperform MEA.

### Alkanolamines screening

Table 10 indicates that MEA is a good choice to propose it as a BAT, which is in agreement with its nowadays industrial use. Although DGA could require a lower solvent flow rate, a higher energy consumption is expected. However; further, research is required on DGA to find the operating conditions in which this compound could be advantageous. On the other hand, although PZ + AMP mixture would provide an energetically favourable process, its high solvent flow rate discourages its use. Finally, simulation results pointing out that some alkanolamines can have a good performance, e.g. MDEA or DEA, should be taken with caution due to the uncertainties in the thermodynamic data implemented in the commercial simulation software. It is possible that some alternative solvents to MEA are advantageous, therefore further research on this field is required and nowadays

| Alkanolamine | α Operation range | Int | α | ΔHabs J/mol | % wt Alk | R² |
|--------------|------------------|-----|---|-------------|---------|----|
| AMP         | 0.2–0.9          | 27.09 | 7.72  | −62,227    | 0.4     | 0.93 |
| MEA         | 0.4–1.0          | 15.49 | 9.16  | −35,091    | 0.11    | 0.99 |
| MDEA        | 0.04–0.9         | 24.02 | 5.54  | −47,405    | 0.05    | 0.90 |
| DEA         | 0.2–0.9          | 21.71 | 9.56  | −46,658    | 0.04    | 0.97 |
| TEA         | 0.03–0.5         | 20.01 | 10.74 | −35,969    | 0.03    | 0.84 |
| PZ          | 0.1–1.0          | 21.29 | 9.16  | −65,295    | 0.92    | 0.99 |
| DIPA        | 0.1–1.1          | 22.34 | 8.23  | −46,068    | 0.05    | 0.98 |
| DGA         | 0.4–0.8          | 22.31 | 13.87 | −30,789    | −0.10   | 0.94 |

Fig. 7 Experimental data and multiple regression AMP + PZ

\[
\begin{align*}
\text{LN PCO}_2 &= 14.58 - 1.7 \cdot (\alpha) - 21558.32 \cdot \left( \frac{1}{RT} \right) + S1 + S2 \quad (11) \\
S1 &= -0.66 \cdot (\% \text{wt AMP}) - 0.24 \cdot (\% \text{wt PZ}) \quad (12) \\
S2 &= 0.77 \cdot (\% \text{wt AMP} \cdot (\alpha)) + 0.26 \cdot (\% \text{wt PZ} \cdot (\alpha)) \quad (13)
\end{align*}
\]
MEIA is suitable to be proposed as the best available technique.

**Alkanolamine losses in exhaust gas**

A final important point is that some alkanolamine amount can be lost in the exhaust gas phase. Unfortunately, the experimental data available for this situation are very scarce and mainly available for ammonia. This section shows that the previous expression is not only valid for CO2 gas liquid equilibrium but also for the alkanolamine gas liquid equilibrium. It provides the basis for the application of this model to perform calculations of absorption columns design.

The design of a physical absorption column for low concentration of absorbed compound (e.g. < 10% vol as for CO2 in exhaust gas combustions) can be easily performed assuming a constant gas flow rate (linear operating line, Eq. 14) and the Henry’s law to define the equilibrium line (Eq. 15). NRTL and UNIQUAC thermodynamic models without dissociation are also suitable to describe the physical equilibrium but not the chemical one (Rosa et al. 2021). Solving the integral of Eq. 16 between the mass fraction of the absorbed/desorbed compound at the exhaust gas input and output streams provides the number of transfer units (NTU) of the absorption column (Eq. 16).

Operating line: \( Y_i = Y_{i1} + \frac{L}{G} \cdot (X_i - X_{i0}) \) (14)

Henry’s law (physical absorption) : \( Y_i^* = H \cdot X_i \) (15)

\[
\text{NTU} = \frac{1}{\gamma_0} \int_{y_0}^{y} \frac{dy}{y - y^*}
\]

where \( Y_i \) is the mass fraction of compound i in the gas phase, \( X_i \) is the mass fraction of compound i in the liquid phase, \( L/G \) is the ratio of liquid and gas mass flow rates, \( Y_{i1} \) is the mass fraction of compound i in gas output and \( X_{i0} \) is the mass fraction of compound i in liquid input, \( Y_i^* \) mass fraction of compound i in the gas phase in gas liquid equilibrium with \( X_i \), \( H \) is the Henry’s constant characteristic for each compound at a fixed temperature.

The chemical absorption is performed in an equivalent way, but using an expression derived from the proposed regression model instead of Henry’s law. For a fixed alkanolamine mass percentage and temperature, Eq. 3 leads to an exponential relation between the liquid and gas equilibrium fractions according to Eq. 17, where A and B are the parameters derived from the proposed model.

Chemical Absorption: \( Y_i^* = e^{A + B \cdot X_i} \) (17)

The pilot plant at Munmorah power station (test serie 5) is used as illustrative example for chemical absorption column design (Yu et al. 2011). The first step is to retrieve the CO2 and NH3 gas liquid experimental data available in the literature and regress the proposed equilibrium model for CO2 (Eq. 18) and NH3 (Eq. 19). The data for CO2 correlation are retrieved from Qi et al. (2015), Jilvero et al (2015), Kurz et al (1995), Otsuka et al (1960) and Verbrugge (1973); the NH3 gas liquid equilibrium is only available in the last two references. The correlation coefficients obtained for the regressed models are of 0.92 and 0.89 for CO2 and NH3, respectively.

\[
\ln \text{PCO}_2 = 26.97 + 9.53 \cdot (\alpha) + 61599.09 \cdot \left( \frac{1}{8.314 \cdot T} \right) + 0.04 \cdot \% \text{NH}_3
\]

\[
\ln \text{PNH}_3 = 20.50 - 4.49 \cdot (\alpha) - 29910.99 \cdot \left( \frac{1}{8.314 \cdot T} \right) + 0.16 \cdot \% \text{NH}_3
\]

The operating conditions assumed, based on the pilot plant data, are the following:

- Adiabatic absorption, operated at atmospheric pressure and constant temperature of 17.5 °C (pilot plant operates between 101–105 kPa and 15–20 °C)
- Gas inlet: 760 kg/h flow rate with 8.5% vol CO2 (37.74% CO2 removal)
- Gas outlet: assumed exhaust gas molar composition of 8.5% CO2, 10% O2, 78% N2 and 3.5% H2O and free of NH3 (pilot plant composition ranges between 8.5–12% CO2, 6.5–10% O2, 76–78% N2, 3–6% H2O, 190–280 ppm SO2, 200–330 ppm NO, < 10 pp NO2).
- Liquid inlet (rich solvent): 8,040 kg/h flow rate with 4.5% wt NH3 (pilot plant operates between 2–5% wt NH3).
• Liquid outlet (lean solvent): 4.5% wt NH₃ and 0.3 CO₂ load (pilot plant operates between 0.2–0.5 load)

Eqs. 18 and 19, for \( T = 290.15 \) K and \( \% \) NH₃ = 4.5% wt NH₃ in liquid phase, depend only on the load (\( \alpha \)) which, due to the constant \( \% \) NH₃, provides the mass fraction of CO₂ in the liquid phase. These equations lead to Eqs. 20 and 21, where the gas molar fraction is calculated by applying Dalton law:

\[
Y^{*}_{CO₂} = e^{1.66+0.82X_{CO₂}} \tag{20}
\]

\[
Y^{*}_{NH₃} = e^{8.83–0.39X_{CO₂}} \tag{21}
\]

The number of transfer units of the absorption column is calculated based on the CO₂ data, i.e. using the \( X_{CO₂} \) in the exhaust gas inlet and outlet (this last calculated from the CO₂ recovery) and the previous equations (Eqs. 14, 16 and 20). Assuming a gas inlet free of ammonia and with the number of transfer units of the column already calculated, the procedure is repeated to calculate the mass fraction of NH₃ at the gas outlet (Eqs. 14, 16 and 21). The calculated amount of NH₃ lost is of 3.67 kg/h which is in good agreement with the experimental value determined in the pilot plant that was around 3 kg/h.

**Conclusions**

A novel shortcut model that regresses a surrogate equation and requires a limited amount of gas liquid CO₂ experimental data is proven to be useful to compare and screening of alkanolamines. The minimum solvent flow rate is calculated by applying the CO₂ mass balance and assuming the liquid CO₂ gas liquid equilibrium is reached at both ends of the absorption column. Therefore, the load and the energy parameter of a novel surrogate model are used as key performance indicators related with the minimum solvent flowrate and energy consumption, whose values are compared to the aqueous MEA which is used as reference. MEA minimum flowrate is 15.62 mol solvent/mol gas and its heat of absorption regression parameter is \(-27,745 \) J/mol. Some alkanolamines perform better than MEA in some respects, for example DGA requires less solvent flow rate or PZ + AMP mixture has a more favourable energy parameter but requires higher solvent flow rate. MEA-based CO₂ capture processes are the leading technology for the CO₂ capture from fossil fuel combustion power plants. The literature review and regression results prove that MEA chemical absorption is a suitable benchmark for other solvents, sustaining its inclusion in BREF documents as best available technique and CO₂ should be included as a contaminant in BREF documents. As future work, the method should be applied to a larger number of solvents and identify alternative solvents to MEA, whose operating conditions of use are more advantageous.

**Supplementary Information** The online version contains supplementary material available at https://doi.org/10.1007/s10098-021-02143-7.

**Acknowledgements** Author Alexandra Elena Plesu Popescu is a Serra Húnter fellow. The authors acknowledge the valuable contribution of master students Alan Chavarria, Diana Medina, Enric Selfa, Toni Peiró and Jesús García from University of Barcelona (Master of Environmental Engineering) for their support to the bibliographic review, data mining and surrogate model development during their master thesis research.

**Funding** Open Access funding provided thanks to the CRUE-CSIC agreement with Springer Nature.

**Declarations**

**Conflict of interest** The authors declare no conflict of interest.

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