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Mining the intrinsic trends of CO_2 solubility in blended solutions

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ABSTRACT

CO_2 solubility in trisodium phosphate (TSP) and its mixed solutions is a crucial information for CO_2 absorption and utilization. However, with limited experimental data and large variations of experimental conditions, intrinsic trends of CO_2 solubility under a specific set of conditions are difficult to be determined without comprehensive experiments. To address this, here, a machine learning based data-mining is proven a powerful method to explore the intrinsic trends of CO_2 solubility trained from 299 data groups extracted from previous experimental literatures. A generalized machine learning input representation method was applied, for the first time, by quantifying the types and concentrations of the blended solutions. With a general regression neural network (GRNN) as the algorithm, we found that the intrinsic trends of CO_2 solubility could be well-fitted with a limited amount of experimental data, having the average root mean square error (RMSE) lower than 0.038 mol CO_2/mol solution. More importantly, it is shown that with a generalized input representation, machine learning can mine the relationships between CO_2 solubility and various experimental conditions, which could help to better understand the intrinsic trends of CO_2 solubility in blended solutions.

1. Introduction

Currently, the increasing pollution not only causes serious problems such as global warming but also could threaten to end human life on our planet. Greenhous gas (GHG) is a main factor that causes the global warming issues. Carbon dioxide (CO_2) emission, mainly emitted from the combustion and utilization of conventional fossil fuels, accounts for approximately more than half of the GHG emissions and is the main greenhouse gas for climate change [1]. The utilization of other cleaner energy, like ethanol [2,3] and clean fuel cells [4], could also increase the CO_2 emission during recent years. According to the recent report by Energy Information Administration (EIA), the worldwide energy-related CO_2 emissions will still increase by a growth rate of 0.6% per year in the year range of 2015 and 2040 compared to a higher growth rate of 1.3% per year between 1990 and 2015 [5]. Although the increasing rate is decreased, it is still crucial to deal with the CO_2 emissions.

During the past years, plenty of researchers have studied the different CO_2 capture techniques in response to dealing with the growing CO_2 emissions [6–11]. Three major CO_2 separation methods from gas mixture are pre-combustion capture, oxy-fuel combustion and post-combustion capture. CO_2 absorption is proved to be an effective and mature technique in post-combustion methods, which has been widely used in various industrial processes [12–14]. Generally, an excellent absorbent type owns a high reaction constant with CO_2 and a high CO_2 solubility. Through former researches and accumulations, a variety of liquid solutions are used as the CO_2 absorption solutions such as water [8], NaOH [15], K_2CO_3 [16], amines [17,18], ionic liquids (ILs) [19], and amino acid salt (AAS) solution [20]. However, these absorbents have limitations in easy to corrosion, low CO_2 absorption capacity, and high energy consumption during the CO_2 regeneration process. Thus, it is motivated to identify a novel solvent which could improve the performance of CO_2 absorption. A blend of two different solvents was proved to be an effective way to obtain the desired performance [8,16].

Piperazine (PZ), amines, and inorganic salts are the commonly used additives for the single solutions which could significantly improve the reaction rate between gas and liquid phases [21–23]. As an inorganic solvent, trisodium phosphate (TSP) with chemical formula Na_3PO_4 is able to absorb the acidic gases. Fig. 1 depicts the 2D and 3D chemical structures of TSP. It is nonvolatile, less corrosive and highly alkaline even at very low concentrations, which is a feasible absorbent for absorbing CO_2. The overall reaction between TSP and CO_2 are demonstrated as follows [24]:

$$CO_2\ (g) + Na_3PO_4{\cdot}12H_2O \leftrightarrow Na_2HPO_4{\cdot}7H_2O\ (s) + NaHCO_3\ (s) + 4H_2O. \quad (1)$$

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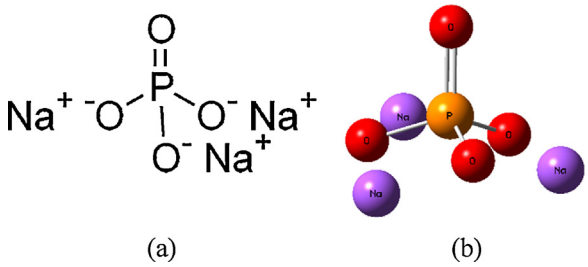

Fig. 1. (a) 2D and (b) 3D chemical structures of trisodium phosphate. Orange, red, and purple spheres represent P, O, and Na, respectively (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

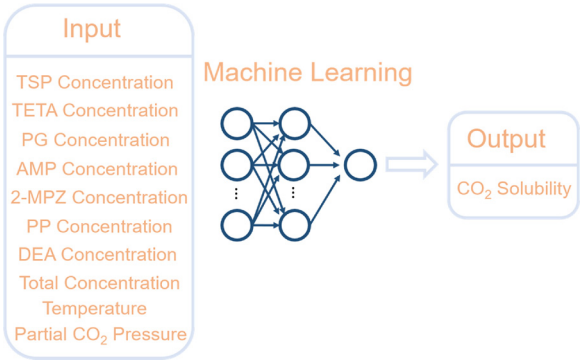

Fig. 2. Schematic diagram of the proposed machine learning method in this study.

Balsora and Mondal [24,25] carried out the CO_2 absorption experiments using TSP and diethanolamine (DEA) + TSP solutions. The solubility of CO_2 in the single and mixed solutions was obtained at 303–333 K. It was indicated that TSP was an excellent promoter to the base fluids for CO_2 capture. TSP was also used as a promoter into the monoethanolamine (MEA) solutions [26]. On the other hand, previous studies [27,28] also reported the CO_2 capture performance for the blended solutions using TSP as the base solution. Compared with MEA solutions, TSP-based solutions showed better absorption performance and had a smaller corrosion rate, which was favorable for the CO_2 capture process. Although the experimental data of CO_2 solubility in TSP and TSP-blended solutions have been reported, the available results were rather limited. Meanwhile, CO_2 absorption experiments were always of high cost, difficult control, and long period. Thus, prediction should be a more efficient way to obtain CO_2 solubility data. To predict the CO_2 solubility in blended solutions, some conventional methods like equation of states (EOS) and molecular dynamics (MD) simulations are theoretically available. However, many EOS requires to specify and acquire the complicated physical and thermodynamic parameters for each specific solution [29], while MD requires huge computational cost as well as a precise empirical energy potential for each CO_2-blended solution system. These drawbacks significantly hinder their applications for CO_2 solubility prediction. Some other fast fitting method (*e.g.*, polynomial fitting) are limited by a specific mathematical form, which may cause some problems like under- and over-fitting, and even exponential explosion. To avoid the potential issues above and provide an ultra-fast solution of CO_2 solubility prediction, in our recent study, it was found that machine learning (more specifically, artificial neural networks (ANNs)) could be a powerful technique to predict the CO_2 solubility and its thermal properties in solution [30], outperforming other statistical methods like multiple linear regression. However, there are more challenging problems that were still unsolved: **i**) how to predict the CO_2 solubility with different solution components and compositions? and **ii**) Can we understand the trends of CO_2 solubility under different experimental conditions? To address these problems, here, a generalized machine learning representation is firstly proposed for adopting different types and concentrations of blended solutions. TSP and the blends of TSP (DEA, triethylenetetramine (TETA), 2-methylpiperazine (2-MPZ), 2-amino-2-methyl-1-propanol (AMP), potassium glycinate (PG), and potassium prolinate (PP)), were examined numerically for the CO_2 solubility. The chemical reaction kinetics between CO_2 and the above solutions were given in Refs. [31–33]. Based on 299 experimental data groups extracted from literature, we show that with a kernel-based ANN model, such a generalized machine learning representation could help to precisely capture the non-linear relationship between experimental conditions and CO_2 solubility. With a well-trained ANN model, the trends of the CO_2 solubility with the ratio between TSP and its blended solutions were fitted and discussed, showing that the data-mining method could help to better understand and predict the intrinsic trends of CO_2 solubility under various experimental conditions.

2. Methodology and modeling

2.1. Modeling

Machine learning is a powerful technique for mining the intrinsic relationships between the independent and dependent variables that are hard to be discovered physically [34,35]. In this study, the target is to predict the CO_2 solubility under different solution components and concentrations, temperature, and partial CO_2 pressure during experiments. Here, we develop a generalized machine learning input representation, which include the concentrations of TSP, TETA, PG, AMP, MPZ, PP, DEA, total concentration, temperature, and partial CO_2 pressure. This representation could help to identify both the type and concentration of the blended solution. If the specific blend component does not exist in the data group, its concentration is artificially set as zero in the input. The corresponding CO_2 solubility is then set as the output. The schematic diagram of the algorithmic architecture is shown in Fig. 2.

In this paper, general regression neural network (GRNN) is applied

Table 1
Data range of CO_2 solubility in TSP and TSP-blended solutions.

TSP concentration range /M	Additive concentration range /M	No. of data	CO_2 partial pressure range /kPa	Temperature range /K	Solubility α /(mol CO_2/mol solution)	Ref.
1.0-2.5	0	30	5.92-48.59	313-323	0.631-1.204	[28]
1.5-2.0	0.5-1.0 (TETA)	45	2.05-48.13	303-323	0.943-1.354	[28]
1.5-2.0	0.5-1.0 (PG)	45	2.99-48.29	303-323	0.706-1.061	[28]
1.5-2.0	0.5-1.0 (AMP)	45	2.38-49.01	303-323	0.667-0.959	[28]
1.5-2.0	0.5-1.0 (2-MPZ)	45	2.18-49.18	303-323	0.881-1.229	[28]
1.5-2.0	0.5-1.0 (PP)	45	2.24-49.02	303-323	0.643-1.084	[28]
1.0-2.0	0	13	10.13-20.27	303-333	0.502-0.96	[24]
0.9-1.96 (DEA)[a]	0.02-0.4 (TSP)	31	10.133-20.265	303-333	0.62-0.869	[25]

[a] In this case, DEA was used as the base solution and TSP is used as the additive.