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Deep Neural Network for Prediction of Adsorbent Selectivity on Hydrogen Purification

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Abstract. With emergence of new materials, more and more materials are available for adsorption and separation processes. The adsorption selectivity of adsorbent to adsorbate is one of the important indicators in choosing materials. Because the adsorption experiment of the mixture is time-consuming and difficult, the selectivity of the adsorbent is generally calculated by the ideal adsorbed solution theory (IAST). Taking the CO_2/H_2 gas mixture as an example, this paper proposes a new adsorption selectivity calculation method based on a deep neural network (DNN) with 5 hidden layers, which takes the molar fraction of CO_2, adsorption pressure and Langmuir adsorption isotherm parameters as the inputs of DNN. Combining the DNN and the NIST/ARPA-E database to quickly and accurately calculate the adsorption selectivity, the hydrogen purification and carbon dioxide storage materials can be quickly screened.

Keywords: Hydrogen purification · Ideal adsorbed solution theory · Langmuir isotherm · Selectivity · Deep neural network

1 Introduction

Integrated gasification combined cycle (IGCC) is an advanced power system that combines coal gasification technology and an efficient combined cycle. IGCC will produce a lot of CO_2 and H_2. It has a good development prospect to capture CO_2 efficiently and obtain high-purity H_2 by pressure swing adsorption (PSA) technology [1]. The commonly used commercial adsorbents are activated carbon (AC) and zeolite. However,

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with the emergence of new materials, metal-organic frameworks (MOFs) are increasingly proven to be effective as adsorbents to separate CO_2 and H_2. Therefore, quickly and accurately selecting the appropriate adsorbent as the separation of CO_2 and H_2 becomes particularly important.

At present, the selectivity of the adsorbent is one of the important indicators to evaluate the performance of the adsorbent. Adsorption selectivity is the thermodynamic characteristic of the equilibrium-based separations system, which determines the separation efficiency [2]. The method commonly used to solve selectivity is the ideal adsorbed solution theory (IAST). The main practical advantage of IAST is that the adsorption selectivity of adsorbent can be estimated simply from the adsorption isotherm of a single gas, so no special equipment is required for the separation measurement of mixed gas [3]. However, the solution of IAST requires integral calculation from different adsorption isotherm models to solve the spreading pressure. Different adsorption isotherm models will obtain different analytical models, and the solution of selectivity is also relatively complex [4].

Deep neural network (DNN) has good advantages in processing non-linear characteristics and has been increasingly used in the simulation, prediction and optimization of gas adsorption and separation. In this paper, a DNN model is used to predict the separation selectivity. The inputs of DNN are CO_2 molar fraction, adsorption pressure and Langmuir adsorption isotherm parameters, and the output of DNN is separation selectivity. The training data of DNN comes from the selectivity by IAST calculations.

2 Mathematical Model

2.1 Langmuir Model for Calculating Adsorption Amount

Langmuir adsorption model is the most widely used isotherm model. The basic Langmuir adsorption model has simple from:

$$n = \frac{n_s b p}{1 + bp} \tag{1}$$

where n represents the equilibrium adsorption amount, p represents the equilibrium adsorption pressure, and n_s and b are saturation capacity and affinity, which are obtained by fitting the experimental values.

The extended Langmuir is used for the adsorption of multi-component gas:

$$n_i = \frac{n_{si} b_i p_i}{1 + \sum_i b_i p_i} \tag{2}$$

where i is the gas component.

2.2 Ideal Adsorbed Solution Theory (IAST) for Calculating Adsorption Amount

The ideal adsorbed solution theory (IAST) assumes that the adsorbed mixture is an ideal solution at constant spreading pressure and temperature. From the IAST, the spreading

pressure π is given by [5],

$$\boldsymbol{\pi_i^0\left(p_i^0\right) = \frac{RT}{A}\int_0^{p_i^0} nd\ln p} \tag{3}$$

where a is the adsorbent specific surface area, $\boldsymbol{R}$ is the molar gas constant. $\boldsymbol{p_i^0}$ is the gas pressure of component i that corresponds to the spreading pressure π.

The spreading pressure for a Langmuir isotherm via Eq. (3) is

$$\frac{A}{RT}\pi_i^0\left(p_i^0\right) = n_s \log\left(1 + bp_i^0\right) \tag{4}$$

At a constant temperature T, the spreading pressure of a single component is the same.

For binary adsorption of component 1 and 2, combining Eqs. (3) and (4),we can get,

$$n_{s1}\log\left(1 + b_1p_1^0\right) = n_{s2}\log\left(1 + b_2p_2^0\right) \tag{5}$$

The IAST requires

$$y_1p_t = x_1p_1^0,\ (1 - y_1)p_t = (1 - x_1)p_2^0 \tag{6}$$

where y_i and x_i are the molar fractions of component i in gas phase and adsorbed phase, respectively, and p_t is the total gas pressure.

2.3 Selectivity Calculated by IAST and Extended Langmuir Equation

Adsorption selectivity in a binary mixture of components 1 and 2 is defined as

$$S_{12} = \left(\frac{x_1}{y_1}\right)/\left(\frac{x_2}{y_2}\right) = \left(\frac{x_1}{x_2}\right)\left(\frac{y_2}{y_1}\right) = \left(\frac{n_1}{n_2}\right)\left(\frac{y_2}{y_1}\right) \tag{7}$$

The adsorption selectivity can be solved by combining Eqs. (5) and (6). If the extended Langmuir equation is used to solve the adsorption selectivity, combining Eqs. (2), (7) becomes

$$S_{12} = \left(\frac{n_{s1}b_1p_1}{1 + \sum_i b_ip_i}/\frac{n_{s2}b_2p_2}{1 + \sum_i b_ip_i}\right)\left(\frac{y_2}{y_1}\right) = \frac{n_{s1}b_1}{n_{s2}b_2} \tag{8}$$

where p_i is calculated from the total pressure p_t by multiplying the mole fraction y_i. In this paper, component 1 represents CO_2, and component 2 represents H_2. So here S_{12} represents the selectivity of an adsorbent for CO_2 compared to H_2. Higher S_{12} is good for hydrogen purification.

2.4 Deep Neural Network (DNN) for Selectivity

Artificial neural network (ANN) has strong nonlinear processing ability, which includes an input layer, hidden layers and an output layer. A deep neural network (DNN) is a neural network with multiple hidden layers. The more the number of hidden layers, the better the nonlinear fitting characteristics of the ANN, and the better it is to deal with complex problems. In this paper, the number of hidden layers is set to 5, and the number of neurons in each hidden layer is 10. The structure of the DNN in this paper is shown in Fig. 1. The inputs of DNN are CO_2 molar fraction, adsorption pressure and Langmuir adsorption isotherm parameters. And the output of DNN is separation selectivity. The chosen method of training data generation is via Latin hypercube sampling (LHS). The significant advantage of LHS is that a small number of samples can represent the entire sample space. The upper and lower boundaries of the LHS are shown in Table 1. The total number of sample points is set to 6000, and the adsorption selectivity values of the selected sample points are calculated by IAST. Due to the large difference between the input parameter values, the calculated selectivity difference is also large. The adsorption selectivity values are set between 3 and 1500. Finally, 3602 sample points are selected to train DNN.

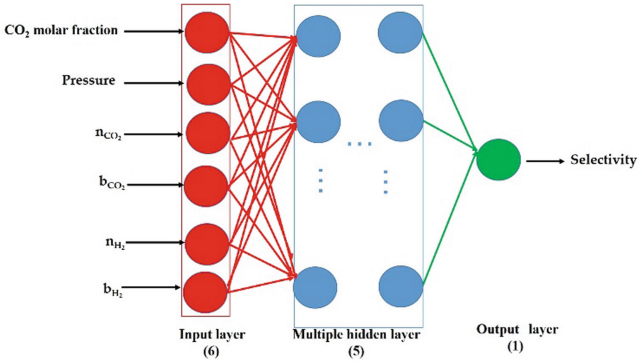

Fig. 1. DNN structure for predicting separation selectivity.

Table 1. Lower (lb) and upper (ub) boundaries of Latin hypercube sampling (LHS).

Condition	CO_2 molar fraction	Pressure (bar)	n_{CO_2} (mol/kg)	b_{CO_2} (bar^{-1})	n_{H_2} (mol/kg)	b_{H_2} (bar^{-1})
lb	0.1	0.1	5	0.1	0.5	0
ub	0.9	30	20	1.5	6	0.6

3 Results and Discussion

The NIST/ARPA-E database of Novel and Emerging Adsorbent Materials is a free, web-based catalog of adsorbent materials and measured adsorption properties of numerous materials, thus making it a treasure trove for data-driven analysis [6, 7]. As shown in Fig. 2, the Isotherm Visualization Tool of the NIST/ARPA-E database can quickly obtain the adsorption isotherm and Langmuir fitting parameters by searching the adsorbent material and gas species. The adsorption of CO_2 and H_2 on activated carbon and CuBTC is randomly selected in the NIST/ARPA-E database. Therefore, by combining NIST/ARPA-E Database and DNN model, the selectivity of different adsorbents to CO_2 and H_2 can be quickly calculated so as to achieve the purpose of fast screening materials.

The algorithm of DNN is completed by Matlab software. The correlation coefficients of the training set, validation set, test set and whole data set of the DNN are 0.99998, 0.9998, 0.99688 and 0.99948, respectively, indicating that the DNN model can well predict the selectivity. In order to further verify the accuracy of the DNN model, the selectivity of activated carbon and CuBTC under different pressures and CO_2 molar fractions is calculated by extended Langmuir, IAST and DNN. As shown in Fig. 3, by comparison, the selectivity values calculated by the IAST agree well with the values calculated by the DNN model, which further verifies the robustness of the DNN model. In Fig. 3 (a), for an 80:20 H_2/CO_2 mixture, CuBTC selectivity under different adsorption pressures is greater than activated carbon. In Fig. 3 (b), at 10 atm, the selectivity of CuBTC is also greater than that of activated carbon at different mole fractions of carbon dioxide. Therefore, for ideal H_2 purification and CO_2 capture materials, CuBTC is more suitable than activated carbon. Compared with activated carbon, CuBTC has a larger surface area and higher pore volume. There are also related articles about the application of CuBTC in PSA hydrogen purification. For the 81:19 H_2/CO_2 mixture, the purity 99.99+ of hydrogen can be obtained by a 4-step PSA cycle with CuBTC as an adsorbent [8].

Besides, after nonlinear treatment by DNN, the process optimization design of adsorption and separation can be carried out. For example, Ref. [9] predicts and optimizes CO_2 adsorption capacity, selectivity and adsorption heat through DNN and multi-objective optimization algorithm. It is worth noting that the selectivity at a fixed temperature calculated by extended Langmuir is a constant value, and the selectivity value does not change with adsorption pressure and mole fraction. Obviously, the calculation error of extended Langmuir is large. Ref. [10] points out that the IAST gives the most accurate predictions than the extended Langmuir and LRC for three binary systems. Therefore, more and more researchers use IAST to develop corresponding application programming interfaces (APIs) for calculating selectivity, such as pyIAST, pyGAPS and GraphIAST [11].

Compared with IAST, another advantage is that the DNN does not need to solve the integral solutions of different isotherm models. Actually, the DNN proposed in this paper can calculate the selectivity of any two-component gas, such as CO_2/CH_4, CH_4/H_2, etc. When the input parameters of DNN exceed the lower and upper boundaries of training data, the predicted results may deviate from the IAST theoretical calculations. The robustness of the DNN can be enhanced by increasing the dimension of input parameters and adjusting the structure of the DNN, which is not the focus of this paper. The DNN

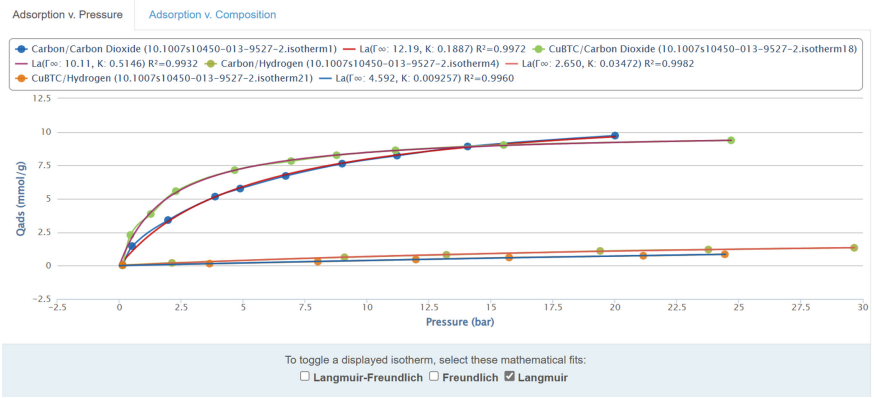

Fig. 2. Adsorption isotherms of CO_2 and H_2 on CuBTC and AC at 298K on NIST/ARPA-E (equilibrium data from Ref. [12]).

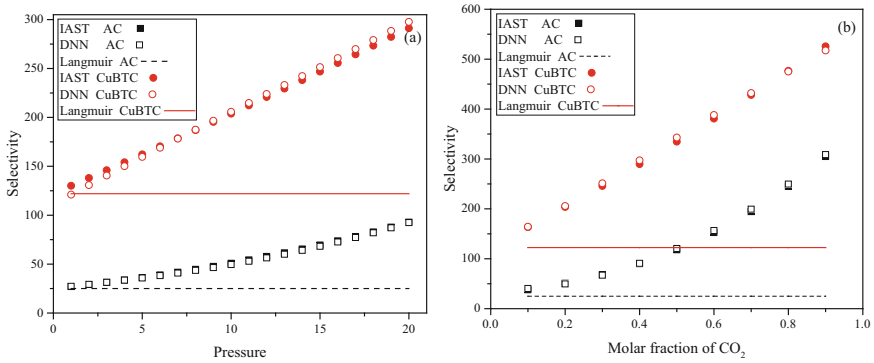

Fig. 3. Extended Langmuir, IAST and DNN calculated selectivities of activated carbon and CuBTC for a 20:80 CO_2/H_2 mixture at different pressure (a) and a 10 atm CO_2/H_2 mixture at a different molar fraction of CO_2 (b).

model is a new attempt at calculating selectivity and can improve the efficiency of adsorbent screening. In addition, the DNN model can also be extended to the calculation of adsorption selectivity of multi-component gases to optimize the design of layered beds with multi-adsorbent.

4 Conclusion

The deep neural network model constructed in this paper can quickly predict the adsorption selectivity of CO_2/H_2. The results show that the correlation coefficient of the DNN model for the whole data set can reach 0.99948. In addition, by combining with NIST/ARPA-E Database, this paper takes activated carbon and CuBTC as examples to compare the adsorption selectivity of the two adsorbents under different pressures and

CO_2 molar fraction at the same temperature. The results show that the adsorption selectivity of CuBTC is greater than that of activated carbon, so CuBTC is more suitable for H_2 purification and CO_2 storage than activated carbon.

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