

User-friendly graphical user interface software for ideal adsorbed solution theory calculations

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Abstract—User-friendly graphic user interface software named IAST++, which was developed, can fit the adsorption data to various isotherm models and use the ideal adsorbed solution theory (IAST) to obtain mixture isotherm data. In the *Modeler* module of the IAST++, automatic model suggestion functionality exists, and users can readily visualize the fitting data via the built-in visualization module. In the *IAST* module of the IAST++, users can import an arbitrary number of pure isotherm data and have the option of selecting desired system variables (e.g., system pressure and gas composition or total uptake and adsorbed phase composition) to obtain the mixture isotherm data. The results of the IAST calculations are shown in both raw data form and visualization module, making IAST++ attractive software for both experimental and computational researchers.

Keywords: IAST, GUI, Adsorption Isotherms, Software, Parameter Estimation

INTRODUCTION

Porous materials contain pores on the order of few Angstroms that allow for adsorption of gas molecules. Due to their large surface area, porous materials such as zeolites, metal-organic frameworks (MOFs), activated carbon, and polymers have been investigated and deployed in many industrial applications [1,2]. For pure component gas molecules, it is relatively easy to conduct experiments under specific conditions (e.g., given temperature, given pressure) to obtain reliable data. In contrast, to traverse through a large space of varying mixture conditions to obtain a complete picture of the mixture isotherm adsorption properties would require a significant number of experiments to be conducted, which can be expensive and cumbersome. Unsurprisingly, this has led to a dearth of available mixture adsorption isotherm data in published literature because the experiments of estimating mixture adsorption are more difficult and complex than that of single component adsorption [3–6]. That said, given that most practical systems such as gas separation system of power plants involve mixtures, it becomes important to develop methods that can lead to an accurate depiction of mixture data.

Fortunately, Myers and Prausnitz developed an ideal adsorbed solution theory (IAST) [7], which provides a solid theoretical foundation to obtain any mixture isotherm data from individual pure component adsorption data. As such, IAST is widely used by scientists to obtain various mixture isotherms for many different systems [8–10]. IAST assumes that the gas phase is an ideal gas mixture and the adsorbed phase is an ideal solution in which the activity coefficient of each phase equals to unity. Therefore, the result of

IAST can be inaccurate for the system in which these assumptions become invalid. To remedy some of these issues, some researchers have modified IAST to overcome those limitations [11–19], but for this particular work, we concentrated on the basic IAST.

From a technical point of view, there are some main obstacles that make the coding of the IAST a difficult problem. First, prior to the actual IAST calculation, users must fit the adsorption data to a suitable isotherm model to obtain a functional form for the appropriate pure component isotherm curves. This is cumbersome because users do not know a priori, which isotherm model is appropriate for given adsorption data, and thus they would need to test all available isotherm models until they find a suitable isotherm model. Second, users have to solve the system of non-linear equations to get IAST calculation results, which can be very time-consuming and arduous.

To facilitate usage of IAST, other researchers have created various software that performs these functionalities. ISOFIT [20] offers a model fit functionality for given adsorption data, thus alleviating the first concern mentioned in the previous paragraph. And recently, the python library for IAST called pyIAST [21] was developed and released by Simon et al., thereby providing both fitting and the IAST calculation modules. However, pyIAST still relies on using command line interface/script files, thus making it difficult to operate for novices who might possess minimal knowledge regarding these platforms.

To reduce the barriers to entry to using the IAST software, we have developed new software, which we named IAST++ (where the name comes from its written language of C++). The IAST++ is graphic user interface (GUI) software that provides both the model-fitting module and the IAST solution module as well as basic visualization tools. The software was written with the novice user in mind as many of the experimentalists who might not want to bother with command line interface can use the software with-

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out any prior training or knowledge of scripting languages. As far as we know, no other software offers these kinds of functionality within the GUI platform.

The paper is divided into three sections (including the Introduction): In Section 2, we explain the structure behind our IAST++ software and provide the details behind each of the functionalities. In Section 3, we analyze the two case studies that demonstrate that our IAST software leads to accurate depiction of the mixture isotherm data.

IAST++ SOFTWARE

Our IAST++ software consists mainly of two modules: (1) *Modeler* and (2) *IAST*. The two modules were constructed separately such that independent operations are possible.

1. Module 1: Modeler

In the *Modeler*, the parameters of different isotherm models are obtained by minimizing the weighted sum of squared error (WSSE) given by

$$WSSE = \sum_i [w_i(m_i - m_{i,exp})]^2, \quad w_i = \frac{1}{1 + m_{i,exp}} \quad (1)$$

where w_i is the weighting factor, m_i is the uptake amount calculated from the model for the i^{th} pressure value and $m_{i,exp}$ is the i^{th} experimental uptake datum. The weighting factor was used to prevent overfitting at just the high uptake values. All the parameters were set to one initially, and the parameters ranged from 0 to 10^5 , where negative numbers were disallowed to prevent unphysi-

cal negative isotherm parameters. The code was designed such that the users can override the default parameter values and range of allowed parameters.

The *Modeler* supports seven different isotherm models selected from available literature [21–23]. The names and the equations for each model are given as follow

$$\text{Linear (Henry): } m(P) = kP, \quad (2)$$

$$\text{Langmuir: } m(P) = q \frac{kP}{1 + kP}, \quad (3)$$

$$\text{Dual-Site Langmuir (DSL): } m(P) = q_1 \frac{k_1 P}{1 + k_1 P} + q_2 \frac{k_2 P}{1 + k_2 P}, \quad (4)$$

$$\text{Langmuir-Freundlich (LF): } m(P) = q \frac{(kP)^n}{1 + (kP)^n}, \quad (5)$$

$$\text{Dual-Site Langmuir-Freundlich (DSLFL):} \\ m(P) = q_1 \frac{(k_1 P)^{n_1}}{1 + (k_1 P)^{n_1}} + q_2 \frac{(k_2 P)^{n_2}}{1 + (k_2 P)^{n_2}}, \quad (6)$$

$$\text{Brunauer-Emmett-Teller (BET):} \\ m(P) = q \frac{k_1 P}{(1 - k_2 P)(1 + (k_1 - k_2)P)}, \quad (7)$$

$$\text{Quadratic: } m(P) = q \frac{k_1 P + 2k_2 P^2}{1 + k_1 P + k_2 P^2}, \quad (8)$$

If it is the case that none of these isotherm models can accurately describe the input data, users can also use the linear interpolator, which simply interpolates all the data points with segments.

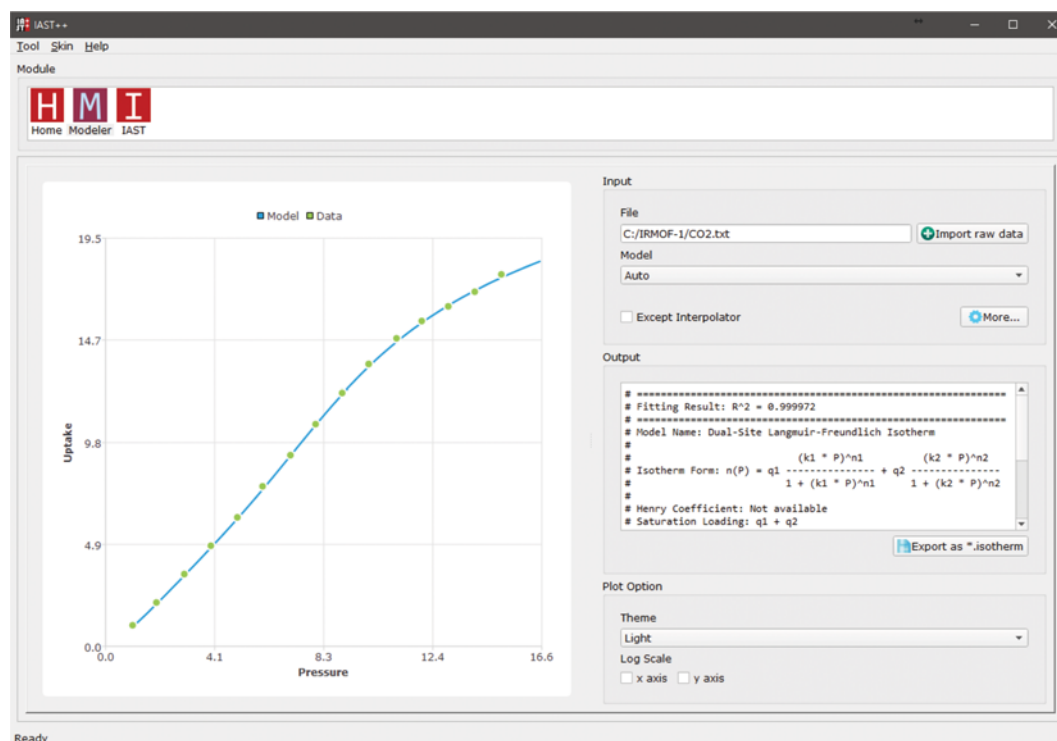


Fig. 1. A snapshot of the Modeler in IAST++. Users can see the fitting result via the visualization plot in left panel side and can obtain the model form and parameters at the 'Output' on the right panel side. Users can save the fit result in '*.isotherm' format and can use these files for IAST and uptake calculation.

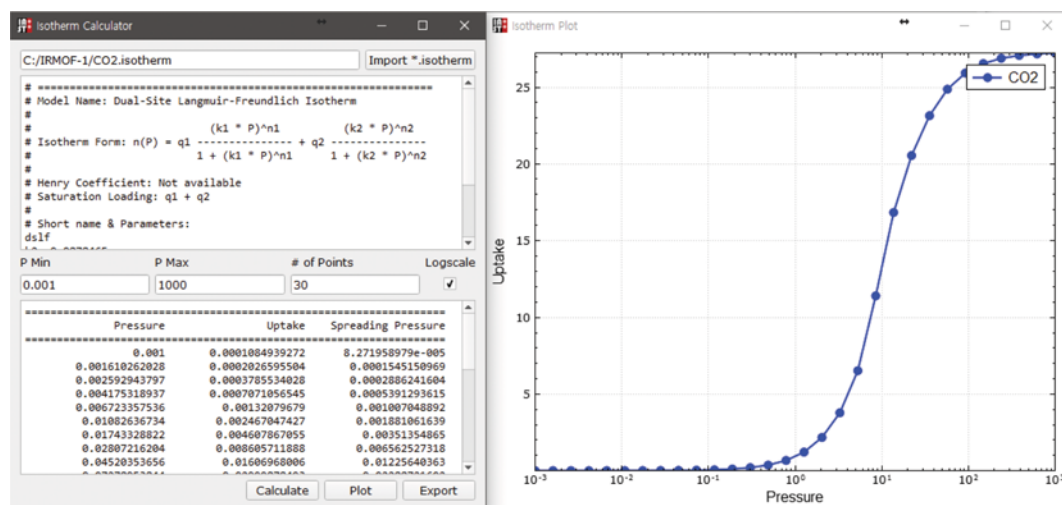


Fig. 2. A snapshot of Isotherm Calculator in IAST++. Users can calculate the adsorption isotherm data from the '*.isotherm' file.

Upon importing the raw adsorption data onto the software framework, the *Modeler* makes comparisons between all of the available models and suggests the most appropriate model for the given adsorption data. There are many ways to determine which one is the best fit, and in IAST++, we use the following equation:

$$F = N_D \times WSSE \quad (9)$$

where N_D is the number of parameters of each isotherm model. The *Modeler* selects the model that has minimum F value. This implies that if the $WSSE$ of all models are similar, the simpler model is suggested. Additionally, if the R^2 , coefficient of determination, is less than 0.9999, the interpolator isotherm is suggested. Our built-in visualization option allows the users to check the fitting results in the same window as the *Modeler* (snapshot of the *Modeler* is shown in Fig. 1). Additionally, IAST++ offers a feature which we call the 'Isotherm Calculator' in which users can calculate uptakes for any pressure value using the result from the *Modeler* (See Fig. 2).

2. Module 2: IAST

In a system of N mixture gas components within the adsorbent

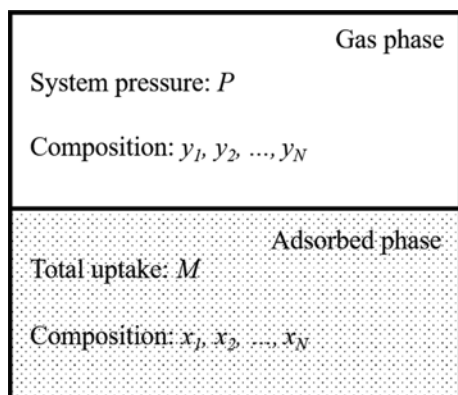


Fig. 3. Illustration of gas mixture adsorbed system. We can get a complete set of thermodynamic information if we set N independent variables in the figure because the total degrees of freedom is N .

at a fixed temperature, there are N degrees of freedom [7] (an illustration of adsorbed gas mixture system is shown in Fig. 3). Therefore, to determine the thermodynamic state of the system, N independent variables must be selected, as shown in Fig. 3. Because the composition of each phase is dependent (the sum of the composition of each phase is unity) on one another, each composition cannot be independently selected. For example, one cannot select y_1, y_2, \dots, y_N but can select $y_1, y_2, \dots, y_{(N-1)}, x_N$. In general, it is natural to select one extensive variable and $N-1$ intensive variables. The IAST module offers three kinds of variable sets and the list of calculation modes is summarized in Table 1.

The solutions to the IAST entail solving a system of non-linear equations that consist of spreading pressure defined as

$$\pi_i = \frac{RT}{A} \int_0^{p_i^o} \frac{m_i^o(P)}{P} dP, \quad (10)$$

where π is the spreading pressure, i is the index of species, R is the gas constant, T is the temperature, A is the surface area, p_i^o is the particular pressure, m_i^o is the adsorption isotherm of pure component i and P is the pressure. Because T , R and A is same for all species, we can define the reduced form of spreading pressure given by

$$\pi_i^* = \frac{\pi_i A}{RT} = \int_0^{p_i^o} \frac{m_i^o(P)}{P} dP. \quad (11)$$

In addition, because particular pressure p_i^o satisfies following equation,

$$P y_i = x_i p_i^o, \quad (12)$$

Table 1. The list of calculation modes of the IAST module. The definition of variables is shown in Fig. 3

Mode	Input variables	Output variables
1	$P, y_1, y_2, \dots, y_{(N-1)}$	$M, x_1, x_2, \dots, x_{(N-1)}$
2	$P, x_1, x_2, \dots, x_{(N-1)}$	$M, y_1, y_2, \dots, y_{(N-1)}$
3	$M, x_1, x_2, \dots, x_{(N-1)}$	$P, y_1, y_2, \dots, y_{(N-1)}$

Table 2. Summary of the forms of spreading pressure, initial guesses, and constraints

Mode	Spreading pressure	Initial guess	Constraint
1	$\pi_i(x_i) = \int_0^{p_{y_i/x_i}} \frac{m_i^o(P)}{P} dP$	$x_i = \frac{m_i^o(P_{y_i})}{\sum_i m_i^o(P_{y_i})}$	$\sum_i x_i = 1$
2	$\pi_i(y_i) = \int_0^{p_{y_i/x_i}} \frac{m_i^o(P)}{P} dP$	$y_i = x_i$	$\sum_i y_i = 1$
3	$\pi_i(p_i^o) = \int_0^{p_i^o} \frac{m_i^o(P)}{P} dP$	Solution of $Mx_i = m_i^o(p_i^o)$	$\sum_i \frac{Mx_i}{m_i^o(p_i^o)} = 1$

the spreading pressure can be expressed as a function of y_i , x_i or p_i^o . The thermodynamic state of the system is obtained by solving the following system of equations,

$$\pi_1^* = \pi_2^* = \pi_3^* = \dots = \pi_n^* \quad (13)$$

The IAST module solves the above equations with various mathematical function forms of the spreading pressure based on mode options. The form and initial guesses of each mode are summarized in Table 2. Although a specific algorithm called FastIAS [24] can be used to solve these equations, we found FastIAS to be quite unstable because it uses the Newton-Raphson method that is highly sensitive to initial conditions. Therefore, the system of equations is solved by minimizing the square of the each equation and constraints using the downhill simplex (Nelder-Mead) method [25]. In general, minimizing the square of the equation is not an appropriate way to solve the equation, but we found that in most of our cases, the solution scheme is stable while providing reasonable accuracy and performance.

CASE STUDIES

In this section, two case studies are provided to verify the accuracy of the IAST++. In Section 3-1, a comparison between IAST++ and pyIAST [21] is made for sanity purposes. In Section 3-2, the five mixture isotherm data for linear alkanes in IRMOF-1 are

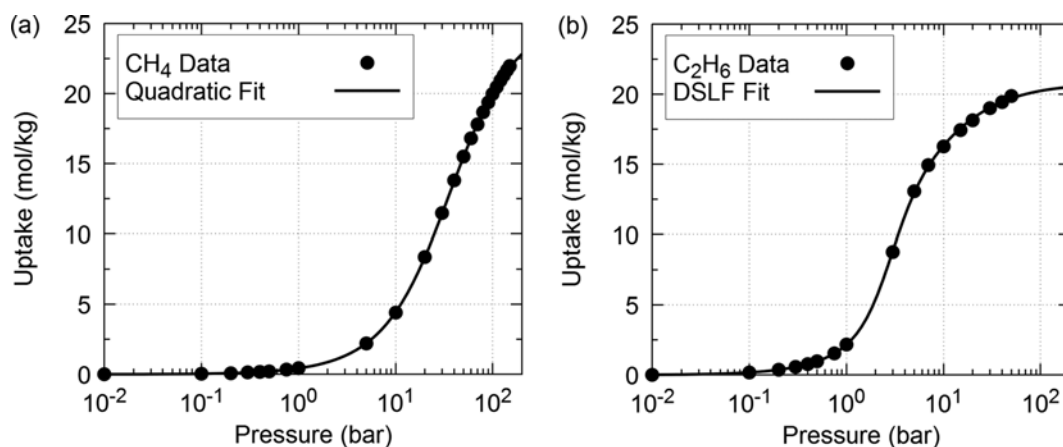
Table 3. The IAST results calculated by IAST++ and pyIAST at T= 298 K. The results of both of IAST++ and pyIAST are shown in the single column because the results of the two software are the same

Pressure (bar)	y_{ethane}	IAST++ and pyIAST	
		Uptake (mol/kg)	x_{ethane}
5	0.05	2.69	0.20
5	0.15	3.79	0.46
5	0.25	5.01	0.63
5	0.35	6.29	0.74
5	0.45	7.63	0.82
10	0.05	5.47	0.22
10	0.15	7.89	0.50
10	0.25	9.93	0.66
10	0.35	11.80	0.76
10	0.45	13.22	0.83
15	0.05	8.09	0.23
15	0.15	11.00	0.51
15	0.25	13.22	0.67
15	0.35	14.63	0.77
15	0.45	15.43	0.84
20	0.05	10.22	0.24
20	0.15	13.27	0.52
20	0.25	15.03	0.68
20	0.35	15.98	0.77
20	0.45	16.62	0.84
25	0.05	12.12	0.24
25	0.15	14.80	0.52
25	0.25	16.12	0.68
25	0.35	16.87	0.77
25	0.45	17.36	0.83

compared with previously published data.

1. Methane and Ethane Mixture in IRMOF-1

In this section, IAST calculations for methane and ethane mix-

**Fig. 4. The model fit result of methane and ethane in IRMOF-1 at 298 K obtained from the *Modeler* in IAST++. The model fits are well described by the adsorption data for both of methane and ethane.**

ture in IRMOF-1 are performed with both pyIAST and IAST++. The selection of this particular system stems from the fact that the same pure component isotherms were analyzed in the pyIAST paper [21]. Similar to the previous work, the interpolator isotherm model is used for both methane and ethane isotherm curves. In Table 3, the IAST calculation results from both pyIAST and IAST++ are listed, and it can be seen that the results from each software are exactly the same.

Next, the interpolator isotherm and model isotherm were compared to see how the selection in the isotherm model affected the IAST calculations. First, the *Modeler* found the quadratic model to be the most appropriate model for methane and Dual-Site Langmuir-Freundlich model, for ethane. The results for the fit and the parameters associated with the aforementioned models can be seen from Fig. 4 and Table 4. Before proceeding, we also checked that auto-suggestion was reasonable by obtaining parameters for all available models. The result of the model fit is shown in Fig. 5, for methane and Fig. 6, for ethane. Fig. 5(a) and Fig. 5(b) represent the same data in different y-axis scale. In Fig. 5, we did not plot the result of DSL and BET because these models converge to Langmuir isotherm and did not plot the result of DSLF because

Table 4. The model fit result of CH₄ and C₂H₆ in IRMOF-1 obtained from *Modeler* in the IAST++

Model	Quadratic			
	q (mol kg ⁻¹)	k ₁ (bar ⁻¹)	k ₂ (bar ⁻²)	R ²
CH ₄	12.8486	0.033182	0.000774	0.999986
Model	Dual-Site Langmuir-Freundlich			
	q ₁ (mol kg ⁻¹)	k ₁ (bar ⁻¹)	n ₁	R ²
C ₂ H ₆	8.47533	0.350368	2.89112	0.999964
	q ₂ (mol kg ⁻¹)	k ₂ (bar ⁻¹)	n ₂	
	12.3083	0.18429	1.05401	

this model converges to LF isotherm. This means that the additional parameters do not improve the accuracy of the model for given adsorption data. The quadratic model well describes the saturation (Fig. 5(a)) as well as the Henry region (Fig. 5(b)). In Fig. 6, we do not plot the result of DSL and BET due to the same reason of Fig. 5. The DSLF model well describes the saturation (Fig. 6(a)) as well as the Henry region (Fig. 6(b)). Also, users can use the qua-

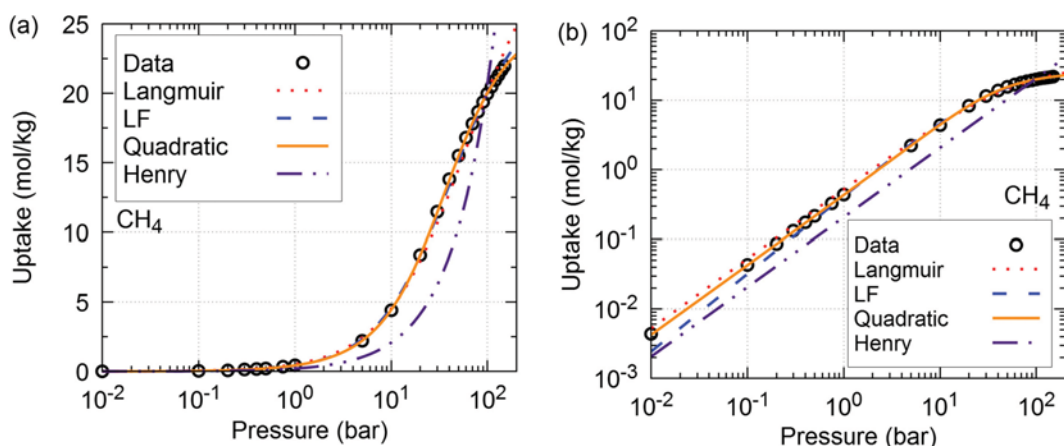


Fig. 5. The model comparison plots for CH₄ adsorption data at 298 K in different axis scale. (a) Linear scale of the y-axis, (b) log scale of the y-axis. The quadratic is the best model for given adsorption data.

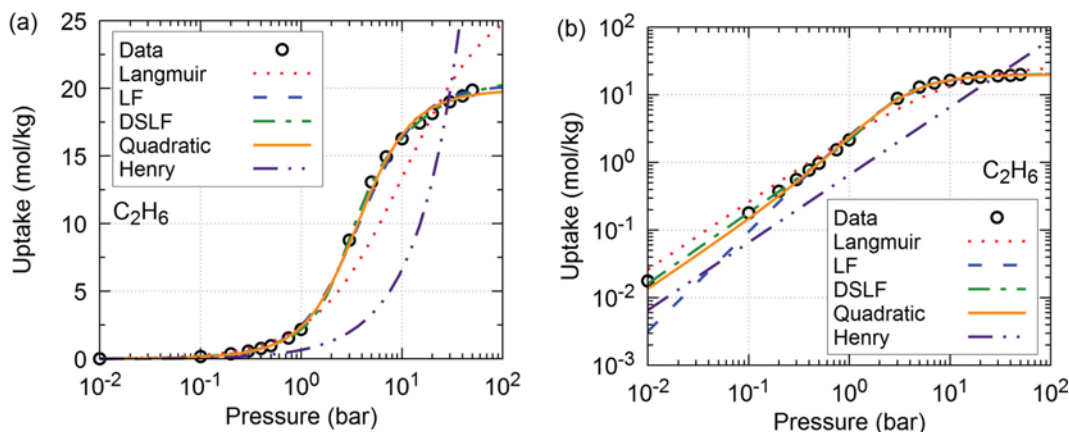


Fig. 6. The model comparison plots for C₂H₆ adsorption data at 298 K in different axis scale. (a) Linear scale of the y-axis, (b) log scale of the y-axis. The DSLF is the best model for the given adsorption data.

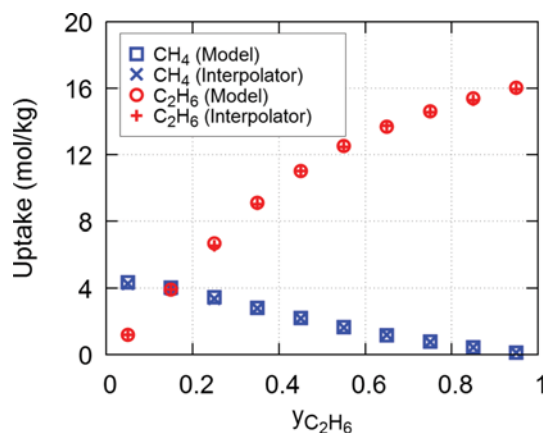


Fig. 7. IAST calculation result in model isotherms listed in Table 4 (square and circle) and linear interpolator (cross and x mark). The result of each model is similar because we have a sufficient range of the pure component isotherms.

dratic model for ethane because the model quite well describes the adsorption data as shown in Fig. 6.

Finally, the IAST calculation was performed at $P=10$ bar and $T=298$ K for varying ethane composition using the best-fit models (Quadratic for methane and DSLF for ethane) and the interpolator isotherm (Fig. 7). The models and interpolator yield almost identical IAST calculation results as there exist a sufficiently large number of uptake data points across a large range of pressures. However, without having sufficient range of adsorption data, the use of the interpolator can be very harmful to the result of IAST calculation because the interpolator isotherm assumes that the last value of uptake is the saturation loading.

Note that users must use the uptake in the 'mol' based unit (e.g., mol/kg or mol/g) not 'mass' based unit (e.g., g/kg) because IAST is based on the molar quantities. Also, all isotherms must have the same units for pressure and uptake. If these rules are not observed, the final results will be significantly inaccurate.

2. Mixture Isotherm of Linear Alkanes in IRMOF-1

In this section, the mixture isotherms of linear alkanes in IRMOF-

1 are computed and compared with the data from the mixture grand canonical Monte Carlo (GCMC) simulation. The pure component isotherm data are taken from the GCMC simulation results by Jiang and Sandler [26]. In their reference paper, the unit of fugacity is used, but in this section, we treat the fugacity as pressure for simplification purposes.

The pure component isotherms of linear alkanes at $T=300$ K are shown in Fig. 8(a). For fitting purposes, the linear interpolator was used for all of the isotherms due to the presence of sufficient range of adsorption data. In Fig. 8(b), the result of IAST and GCMC simulation are shown with the gas component ratios of $C_1:C_2:C_3:C_4:C_5=5:4:3:2:1$. As shown in Fig. 8(b), there exists a good agreement between the results of IAST and GCMC simulations. The software snapshot of IAST++ performing this particular calculation is shown in Fig. 9.

CONCLUSION

IAST++ is an integrated software tool that allows users easy access to IAST. IAST++ offers various isotherm models from the simplest linear model to the practical isotherm model (e.g., DSLF). Also, there is auto-suggestion functionality in that users can get the best model for given data with reasonable matrix and can check the result of model fitting via plot in GUI. IAST++ also offers additional calculation mode (mode 3) that can predict the pressure and composition of the gas phase. We newly suggest the initial guess for the mode 3 that is stable for most of the conditions. There is no functionality in the existing libraries. Given that IAST is a method used by many experimentalists in the field that pertains to gas adsorption/absorption, the goal was to make very easy-to-use software geared towards novice users. As such, we included built-in visualization tools such that the users need not import their raw data onto another platform to view their results. Currently, we have a free version of the IAST++ software available on our website, with the links to a manual and a video tutorial. It is our hope that we can expand the functionality of this software in version 2.0 to satisfy the needs of primary experimentalists who are not yet well-versed in using the computational software.

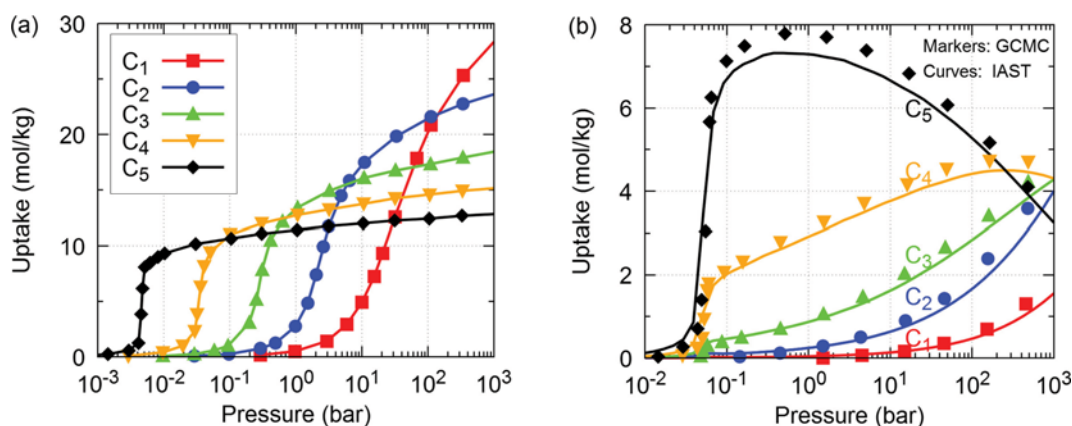


Fig. 8. (a) The pure component isotherms of linear alkanes in IRMOF-1 at 300 K calculated using GCMC simulation [26]. (b) The mixture isotherms of linear alkanes in IRMOF-1 at 300 K. The composition ratio of the gas phase is $C_1:C_2:C_3:C_4:C_5=5:4:3:2:1$. The markers indicate the result of GCMC simulation [26] and the curves indicate the result of IAST.

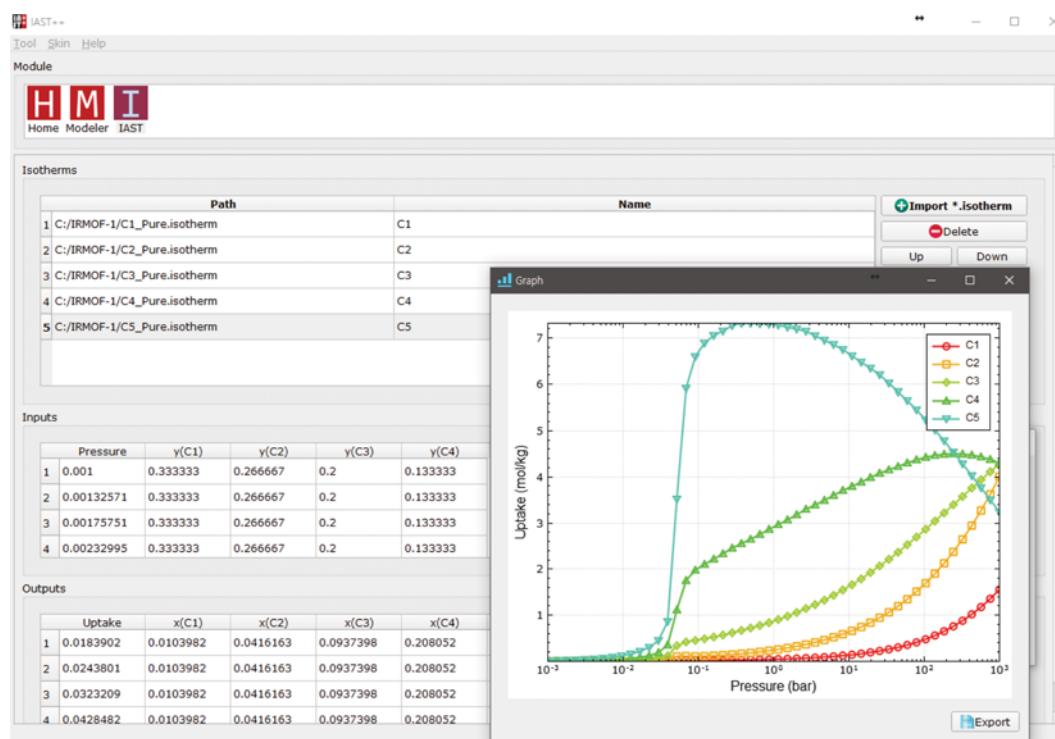


Fig. 9. A snapshot of the IAST module in IAST++. The inputs and outputs are given in table form on the left side. Users also can see the result of IAST calculation via the plot on the right panel side.

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NOMENCLATURE

A	: surface area
F	: $N_D \times \text{WSSE}$
k	: isotherm model parameter
k_1	: isotherm model parameter
k_2	: isotherm model parameter
M	: total amount of uptake of adsorbed phase
m	: uptake function
m_i	: uptake amount calculated from the model for the i^{th} pressure value
m_i^0	: adsorption isotherm of pure component i
N	: the number of species
N_D	: the number of parameters of each isotherm model
n	: isotherm model parameter
n_1	: isotherm model parameter
n_2	: isotherm model parameter
P	: pressure
p_i^0	: particular pressure of species i
q	: isotherm model parameter
q_1	: isotherm model parameter
q_2	: isotherm model parameter
R	: gas constant
R^2	: coefficient of determination

T	: temperature
WSSE	: weighted sum of squared error
w_i	: weighting factor of i^{th} pressure value
x_i	: adsorbed phase composition of species i
y_i	: gas phase composition of species i

Greek Letters

π_i	: spreading pressure of species i
π_i^*	: reduced spreading pressure of species i

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