

MODELING OF ADSORPTION ISOTHERM CONSTANTS USING REGRESSION ANALYSIS & NEURAL NETWORKS

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SYNOPSIS

This paper deals with modeling the suitability of novel adsorbents in terms of the isotherm constants and temperature. Two different models viz., Linear Regression Model (LRM) and Model based on Normalized Sum of Squares (NMSS) are used. A database comprising of 120 data points (DB-1) pertaining to Freundlich constants is compiled from literature to find out the trends of n and k on Temperature (T). The accuracy of the prediction is compared using Standard Deviation (SD) based on expected values. The results are not satisfactory but indicated that the SD is lower for MNSS than that for LRM. The database is sorted out into three other data sets viz., having n values in the range 0.9 to 10 (comprising 94 data points), two clustered datasets for removal of Cr^{+6} & Pb^{+2} comprising of 12 and 14 data points respectively and fitted using both models. The SD values are comparatively lesser than those obtained with DB-1, but still not encouraging indicating wide spread in the database.

A functional relation is proposed for k in terms of n , Langmuir constants, and T . It is found that 39 data points are following both isotherms. A three-layer Back Propagation Network (BPN) is developed to predict k with 33 data points for training and the rest for testing. The results show that, a SD of 1.8676 is observed for three neurons in hidden layer at a learning rate of 0.7. This SD value again is not in the range of acceptable limit though better than the other regression techniques. This study hence throws light on the limitations of the experimental data reported in literature on adsorption isotherms.

1. INTRODUCTION

Physical modeling for any Engineering application is usually based on proposing empirical relations with large amount of experimental data and the relevant non-dimensional parameters using Regression techniques (Babu and Ramakrishna, 2002). Langmuir and Freundlich isotherms are used for fitting the experimental data in adsorption studies to understand the extent and degree of favorability of adsorption (Treybal, 1981). Each of these isotherms has two constants in their general forms (Singh and Srivastava, 2001; Patnaik and Das, 1995; Singh and Rawat, 1993; Ramu et al., 1992) given respectively by equations (1) and (2), indicating:

- Adsorption capacity: b of Langmuir equation and k of Freundlich equation
- Energy of adsorption: a of Langmuir equation
- Intensity of adsorption: $(1/n)$ of Freundlich equation

$$q_e = \frac{b(aC_e)}{(1+aC_e)} \quad (1)$$

$$q_e = k(C_e)^{1/n} \quad (2)$$

The Freundlich constant, n also indicates the degree of favorability of adsorption (Treybal, 1981). Both the isotherms depend upon temperature (T). The Freundlich constant, n should have values lying in the range of 1 to 10 for classification as *favorable* adsorption (Rao and Bhole, 2001; Raji et al., 1997).

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The constants a and k are used to estimate the enthalpy of adsorption (Singh and Srivastava, 2001). From the enthalpy of adsorption, the spontaneity (Patnaik and Das, 1995) and nature of adsorption as to whether it is exothermic or endothermic is predicted (Singh and Srivastava, 2001; Ajmal et al., 1998; Manju and Anirudhan, 1997; Raji and Anirudhan, 1997; Raji et al., 1997). A smaller value of $(1/n)$ indicates a stronger bond between adsorbate and adsorbent (Ramu et al., 1992), while a higher value for k indicates rate of adsorbate removal is high (Ajmal et al., 1998; Ramu et al., 1992). Hence it should be noted that the Isotherm constants are important in understanding the adsorption mechanism and their subsequent application for prediction of some important design parameters. They are dependent on temperature since temperature is an important parameter in estimating the thermodynamic parameters. A number of novel adsorbents are being developed by several investigators as an alternate to activated carbon (Babu and Ramakrishna, 2001; Rai et al., 1998; De and De, 1994). The present paper is focused on studying and modeling the suitability of the novel or non-conventional adsorbents in terms of the isotherm constants and temperature reported in literature.

2. PROBLEM FORMULATION

In order to understand the functional dependency of these two constants on temperature, a database comprising of 120 data points pertaining to the Freundlich constants is collected from literature (Bhole et al., 2002; Rao and Bhole, 2002; Rao et al., 2002; Singh and Srivastava, 2001; Das and Patnaik, 2001; Singh et al., 1993; Ansari et al., 2000; Verma and Shukla, 2000; Prasad et al., 2000; Chandrasekhar and Chakravarthy, 2000; Rengaraj et al., 1999; Singh and Srivastava, 1999; Ajmal et al., 1998; Mall and Upadhyay, 1998; Mahadevaswamy et al., 1998; Anirudhan and Sreedhar, 1998; Raji and Anirudhan, 1997; Manju and Anirudhan, 1997; Raji et al., 1997; Mall and Kumar, 1997; Raji and Anirudhan, 1996; Srichand et al., 1994; Himesh and Mahadevaswamy, 1994; Ramu et al., 1992; Khagesan et al., 1991; Periasamy et al., 1991; Grover and Narayanaswamy, 1982; Khanna and Malhotra, 1977) and examined. It is found that, huge variations exist in the database of adsorption isotherm constants reported for several adsorbates and adsorbents. The range of the values is very wide ranging from, for example k , as low as 1×10^{-7} to as high as 727.05. Interestingly, satisfactory results are being reported in literature with respect to either of the two isotherms or both. The upper and lower limits of the Freundlich constant, k could not be found in the literature. Though it is reported in literature (Rao and Bhole, 2001; Raji et al., 1997) that the value of n in the range of 1 to 10 indicates favorable adsorption, few data points with values less than 1 are also reported. Under such circumstances, the database on Freundlich isotherm constants, i.e., adsorption capacity (k) and n is examined with the following objectives:

- To find the percentage of data points related to non-conventional/novel and commercial activated carbons
- To determine the Freundlich constant n reported in the range of 1 to 10
- To find the existence of any trend between k & T and n & T since k and n are *Isotherm* constants
- To understand the relationship among the Freundlich and Langmuir constants

The database is compiled and analyzed with the following assumptions:

- Unless otherwise mentioned in the literature, the room temperature is assumed to be 30 °C.
- While sorting the database on Freundlich constant (n), the lower limit is considered as 0.9 instead of 1, assuming a slight experimental error or graphical error in plotting the data in the determination of n value. Considering this assumption, the number of 'extra' data points that lie in the range of 0.9 & 1 for each of the databases are as follows:
 - Three data points in DB-2 i.e., a database comprising 94 data points
 - One data point in DB-4 i.e., a database comprising 14 data points exclusively for Pb^{+2}

3. RESULTS AND DISCUSSION

3.1. Linear Regression Model (LRM)

The database comprising of 120 data points (DB-1) are first sorted out with respect to the reported values of Freundlich constants, k and n . The Linear Regression technique is used to develop empirical Model (LRM) for prediction of k and n with respect to T . Standard Deviation (SD) based on the expected value (Babu, 1993) is chosen to identify the accuracy of the above Linear Regression Model (LRM).

It is observed that,

- The Freundlich constant, n is reported to have values in the range of 0.9 to 10 for 94 data points (comprising 87 data points of 43 different novel adsorbents) out of 120 literature data (comprising 110 data points of 51 different novel adsorbents) (see Fig. 1).
- There is a huge variation in the database (see Table-1) wherein the reported value of k ranges from 727.05 units to a very low value of 1×10^{-7} units. The reported values of n and T are varying from 25 to 0.06 and 70°C to 20°C respectively.
- The SD value of k vs. T is 104.84 while that of n vs. T is 3.46.

The database of 94 data points where the reported values of n lie in the range of 0.9 to 10 (DB-2) are separated from the initial database of 120 data points. The LRM is applied to DB-2. It is observed that:

- The reported value of k ranges from 727.05 units to a very low value of 3×10^{-6} units. The reported value of n and T are varying from 7.8125 to 0.91 and 70°C to 20°C respectively (see Table-1).
- The SD value of k vs. T is 86.558 while that of n vs. T is 1.267.
- The SD value of DB-2 for both the above empirical relations is relatively lower than that for DB-1 indicating the relative reduction in the variations in the values between DB-1 and DB-2 reported in literature.

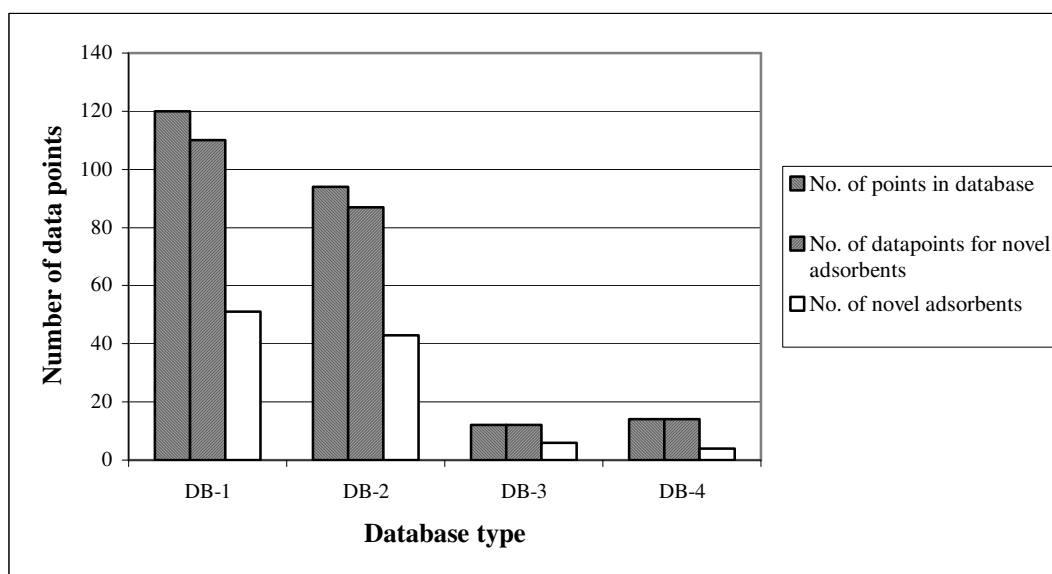


Fig. 1 Distribution of data points in the databases

Table-1 Range of Freundlich isotherm constants and temperature in the database

S.No	Database type	Range	k	n	T	Total data points
1	DB-1	Max	727.05	25	70	120
		Min	1×10^{-7}	0.06	20	
2	DB-2 #	Max	727.05	7.813	70	94
		Min	3×10^{-6}	0.91	20	
3	DB-3	Max	52.72	4.655	60	12
		Min	1×10^{-7}	1.01	20	
4	DB-4	Max	13.90	5.49	70	14
		Min	0.06	0.91	25	

The database for which the value of n lying in the range of 0.9 to 10

Since, it is evident from the above observations that, the variations in the database is due to spreading in the range of data points, the DB-2 is screened for clustering based on the type of adsorbate. Two datasets for removal of Cr^{+6} (DB-3) & Pb^{+2} (DB-4) comprising of 12 and 14 data points each are identified and subjected to regression using LRM. The results for each of the database are as follows:

- **For DB-3**
 - The Freundlich constant, n is reported to have values in the range of 1 to 10 for all the 12 data points comprising of 6 different novel adsorbents.
 - The reported value of k ranges from 52.72 units to a very low value of $1e-7$ units. The reported value of n and T are varying from 4.655 to 1.012 and 60 to 20 °C respectively (see Table-1).
 - The SD value of k vs. T is 5.7169 while that of n vs. T is 0.4048.
- **For DB-4**
 - The Freundlich constant, n is reported to have values in the range of 0.9 to 10 for all the 14 data points comprising of 4 different novel adsorbents (see Table-1 and Fig. 1).
 - The reported value of k ranges from 13.895 units to a very low value of 0.06 units. The reported value of n and T are varying from 5.49 to 0.91 and 70 to 25 °C respectively.
 - The SD value of k vs. T is 1.08464 while that of n vs. T is 0.3629.

A close examination of above observations indicates that, the error in previous cases i.e., DB-1 to DB-4 is due to wide spread in the range of data points since the SD and the range of three variables considered in the present study are showing decreasing trends.

3.2. Model based on Normalized Sum-of-Squares (MNSS)

The above database is subjected to regression using another model based on Normalized Sum-of-Squares (MNSS), which indicates the weighted error in the database and is given by (Hanslip and Buffham, 2002).

$$\text{Sum of squares, } S^2 = \sum_{i=1}^N (M_i - M_{\text{model}})^2 \quad (3)$$

$$\text{Normalized sum of squares, } s = \frac{\sqrt{nS^2}}{\sum M_i} \quad (4)$$

In this approach, lesser value of s indicates a good fit.

3.3. Comparison of LRM & MNSS

The above four databases, DB-1 to DB-4, are subjected to MNSS and the results are given in Table-2. From Table-2, it is evident that the SD values are comparatively lesser than that in LRM. This indicates that,

- the data points are widely spread in the database and
- the deviations in the errors are canceling out leaving a small error in the database

Table-2 Comparative results of SD for MNSS and LRM

S.No.	Parameters	Model	Standard Deviation for			
			DB-1	DB-2	DB-3	DB-4
1	k vs. T	LRM	104.93	86.661	5.7168	1.0846
		MNSS	2.578	2.798	0.7914	0.3000
2	n vs. T	LRM	3.463	1.261	0.4048	0.3629
		MNSS	1.223	0.554	0.4514	0.4323

High values of SD are obtained for the trend k vs. T (see Table-2) for both DB-1 & DB-2 under LRM. Interestingly, the SD value for these two databases is drastically reduced using MNSS. This led to a close examination of k values in the databases. The k values are compiled in four clusters: (i) values greater than 100; (ii) values of k lying between 100 and 10 (both limits including); (iii) values of k less than 10 and up to 1 (including 1); and (iv) values of k less than 1. The results are plotted in Fig. 2 for all the databases. The value of k exceeds 100 for 11 data points and is less than 1 for 33 data points in DB-1 (see Fig. 2). In case of DB-2, the k value exceeds 100 for 5 data points and is less than 1 for 21 data points. Interestingly, the k values in DB-3 & DB-4 are less than 100. The spread in the k values is reduced from DB-1 to DB-4, which is evident from Fig. 2. This explains the reason for very high values of

SD in DB-1 & DB-2 using LRM. In the case of MNSS, the wide spread in the k values got nullified resulting in comparatively lower values of SD for DB-1 & DB-2.

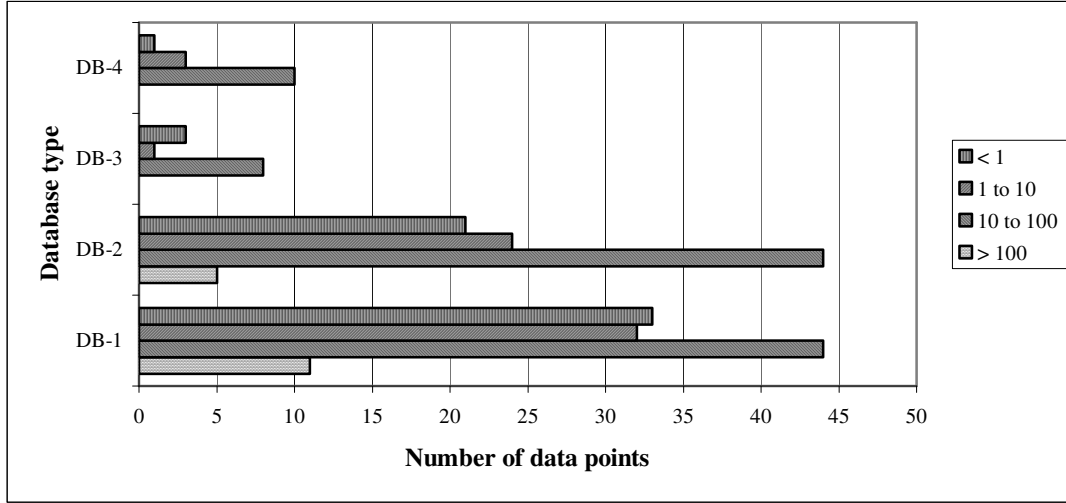


Fig. 2 Distribution of k values in the databases

3.4. Model based on Generalized Langmuir Equation

The adsorption isotherms may be described (Goyal et al., 1999) by general Langmuir equation given by

$$q_e = \frac{b(aC_e)^n}{(1+aC_e)^n} \quad (5)$$

The above equation reduces (Goyal et al., 1999) to conventional Langmuir and Freundlich isotherms respectively under specific conditions such that,

$$q_e = k(C_e)^n \quad (6)$$

$$\text{where, } k = b(a^n) \quad (7)$$

Here, k and n are Freundlich constants while a and b are Langmuir constants, while q_e and C_e are the amount of adsorbed (mg/g) and the adsorbate concentration at equilibrium (mg/L) respectively.

Starting from the generalized form of the equation (Eq. (5)), in this paper, the functional dependency of the parameter k with n , and the two Langmuir constants is studied. Since, these constants are dependent on temperature, a function is proposed such that, k depends on n , Langmuir constants, and T . From the entire database of 120 experimental data points collected from literature, the data reported to be following both the Isotherms is separated. It is found that 39 data points (comprising 34 data points pertaining to 10 novel adsorbents) are lying under this condition in the database.

3.5. Model based on Artificial Neural Networks: Back Propagation Network (BPN)

A three-layer Back Propagation Network (BPN) is developed and used to predict k with 33 data points for training (DB-5) and the rest for testing (DB-6). The range of the variables lying in the above two databases is given in Table-3.

The results show that, a SD of 1.8676 is observed for three neurons in hidden layer at a learning rate of 0.7. This SD value again is not in the range of acceptable limit though better than the other regression techniques. This throws light on the limitations that might have been faced in generating the experimental data on adsorption isotherms.

Table-3 Range of segregated data for BPN analysis

S.No.	Parameter	Maximum	Minimum
1	Langmuir constant, b	333.33	0.0092
2	Langmuir constant, a	2.58	0.00109
3	Freundlich constant, n	25	0.4428
4	Freundlich constant, k	727.05	0.06
5	Temperature, T	70	25

Babu and Ramakrishna (2002) opined that, the Physical Modeling based on affecting dimensional variables in conjunction with Regression Techniques does not always ensure good prediction unless and until all the affecting variables or affecting dimensionless groups are identified. The success of physical modeling is highly dependent on the thorough understanding and complete knowledge of the physical phenomenon that is taking place in the given system. The authors, in the above study, tested a multiple variable linear regression model to predict the removal of fluoride from contaminated water. The results from that study proved that Regression Techniques have the limitation in accurately predicting the output due to incomplete understanding of physical phenomena. Hence the authors used a three layer BPN of Artificial Neural Networks (ANNs) for Physical Modeling purposes. A SD of 0.26 is observed and is comparatively less than that obtained for linear regression model indicating that ANN can be used as a tool for successfully developing a physical model. Successful applications of ANN to adsorption studies are reported in literature (Basheer and Najjar, 1996).

4. SUMMARY & CONCLUSIONS

Langmuir and Freundlich isotherms are used for fitting the experimental data in adsorption studies to understand the extent and degree of favorability of adsorption. The two isotherms depend on temperature and they have two constants each in their general form. The present paper focused on studying and modeling the suitability of the novel or non-conventional adsorbents in terms of the isotherm constants and temperature reported in literature. Based on literature review, a function is proposed such that, k depends on n , Langmuir constants, and T .

The results of the study indicated that,

- There is a huge spread in the range of values for isotherm constants reported in literature.
- The Freundlich constant n lies in the range 0.9 to 10 for majority of the novel adsorbents.
- The range of the Freundlich constant, k values reported did not have any correlation with n .
- Empirical model between k & T and n & T did not give encouraging results for a heterogeneous data.
- Regression Techniques have the limitation in accurately predicting the output due to incomplete understanding of physical phenomena.
- ANN can be successfully used as a tool for developing a physical model.
- There is a need to have a closer look at adsorption as a process, the procedures followed and problems encountered in generating the experimental data reported in literature *in lieu* of its huge variations.

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