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## Modelling of adsorption process in industrial wastewater treatment – A review

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Industrial wastewaters contain a multitude of environmentally toxic and harmful pollutants, which when discharged unchecked can cause irreparable damage to the biota. Adsorption has been an exciting method of treatment for the removal of toxic components from industrial effluent. An understanding of isotherm modelling will further the appreciation of the adsorption process behaviour. Sorption rate, thermodynamics, stability, spontaneity and capacity, are all parameters that can be estimated by mathematical modelling using isotherms. Several models exist to simulate and predict adsorption behaviour, each attempting to simplistically represent the underlying process, building on certain assumptions. This paper reviews existing adsorption isotherms and the advancements made by authors in this field, whether it be non-linearization, multi-component accommodation or parameter replacement. A researcher may apply the model that best suits his or her experimental requirements.

Keywords: Isotherm, mathematical modelling, advanced treatment, adsorption.

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### Introduction

The current market from the local to the global scale is filled with consumerist products. Invisible to the eye is the manufacture mechanism which consumes its unique set of raw materials and spouts out toxic by-products. The multitude of by-products from innumerable industries are to be treated by intricately designed processes so as to reduce them to a more acceptable and eco-friendly form. One of the processes that achieves this is adsorption and the process design is governed by an isotherm model. Even with the advent of modern programming power, the need for aesthetic models has not diminished. Researchers even today attempt to incorporate as many adsorption process parameters as possible into their models in the most efficient and humble ways possible. The objective of this paper is to review all the prominent classical and modern approaches to isotherm modelling and were amalgamated in the previous that the current century.

### Linearized verses non-linear mathematical models

The broad standard in modelling is to use linear regression first to determine whether it can fit the particular type of curve for data available. Linear regression models can fit limited curve shapes, but mathematical simplicity yields pa-

rameters more easily. If linear regression fails, then the necessity of nonlinear regression arises. Linear regression trumps over non-linear modelling largely due to its simplicity of understanding, ease of application and presentation of wider statistics for model and data assessment. Non-linearization is much more versatile. It can model a wide variety of curve profiles, but requires more effort in independent parameter determination and best-fit analysis. Due to such high flexibility in model formation, hypothesis testing has to be performed on each parameter individually. The regression coefficient and  $p$ -values are also not possible to be found.

A nonlinear programming was executed up to a particular number of iterations within limits of the experiment<sup>1</sup>. Upon convergence, this led to simplification of the procedure and acceptable parametric values. Statistical predictors are great tools associated with a regression study. They help in developing an optimal process equation for the sorption isotherm. A study was conducted among five commonly used statistical indicators<sup>2</sup>. The best fit isotherm determination in case of linear regressions was presented best by Student's T-test and the  $\chi^2$ . In case of non-linearity, four out of the five functions (except Student's T-test) gave agreeable estimations. Error analysis was used as another indicator for comparing performance of best fit by linear and non-linear regression.

$R^2$  coefficient was best suited for linearization and error functions like HYBRID and sum of squared errors, SSE represented non-linear systems well.

In another study, performance of multiple regression models<sup>4</sup> was appraised by means of median percentage error (MPE) and mean absolute relative error (MARE). In case of repetitions greater than three, orthogonal distance regression method gave results synonymous with the weighted least squares model, a much simpler procedure. A study involving the utilisation of activated carbon for removal of basic dyes demonstrated that the non-linear method was better at acquiring the isotherm parameters<sup>5</sup>.

Linearizing a non-linear isotherm equation leads to creation of inherent errors. The linear approach considers constant error distribution at each value of equilibrium pollutant concentration in solution. This alters the shape of error distribution. Regression on non-linear data would evade such inaccuracies and provide more accurate parametric values. Realizing this, linear model parameter estimation was forsaken in a study and only non-linear isotherm and kinetics modelling was done<sup>6</sup>. Both isotherm seemed to predict results of chemisorption very well alongside Langmuir model.

Langmuir isotherm, when linearized in different forms yields varied Langmuir constants. Graphs of  $C_e/q_e$  against  $C_e$ ,  $1/q_e$  against  $1/C_e$ ,  $q_e$  against  $q_e/C_e$ , and  $q_e/C_e$  against  $q_e$  were plotted<sup>3</sup>. It was understood that each linearized form presents different variation in errors, specific to the corresponding mode of linearization. In other words, the reorientation of a non-linear isotherm model into a linear isotherm model seems to indirectly modify the error functions, thus influencing error variance. A general trial-and-error technique, applicable to computer based linear regression was developed to determine the coefficient  $R^2$  for a series of values of a constant  $A$ .

### Modelling of adsorption

Modelling using various methods have been performed by experimenters across the globe. Henry's law along with Langmuir and Freundlich models are considered as pillars of classical isotherm modelling. Many further models have been developed on tracks of thoughts based on mechanistic and empirical approaches. Compilation of sequentially arranged modular mathematical steps constitute a mechanistic model. These models characteristically have a perceptible and physical facet<sup>49</sup>. Extensive parameter observation

and data recording are what characterise an empirical model. A demerit to this approach is that an empirical model allows you to predict the product yield sans the understanding of process and parameter interactions. A suggestion to tackle this situation and attain superlative yields is to develop mechanistic models based on empirical modifications, after extensive data collection and analysis.

Researchers have long since tried to model adsorption capacities of different sorbents<sup>50</sup>. Condensed information of different models has been listed in Table 1.

Species attempted to be removed from wastewaters include single and multiple component heavy metals, single and polycyclic aromatics, long chain organics, volatile compounds as also multi component acid dyes and other toxic pollutants that escape biological effluent treatment.

The Langmuir standard was applied to multiple component mixtures. It calculates the sorptive capacity,  $q_i$  (mg/g) of  $i$ -th component in a mixture of  $N$  components at equilibrium using the equilibrium concentration  $C_{ei}$  (mg/L). Maximum sorption capacity,  $q_{mi}$  (mg/g) and Langmuir coefficient,  $k_i$  are determined by fitting the single component adsorption equilibrium data. To account for errors generated from such an approximation, a correction  $n_i$  was applied. It depends on the characteristics of the  $i$ -th component as also the multi-component system characteristics<sup>12</sup>. We have,

$$q_i = \frac{q_{mi} k_i \left( \frac{C_{ei}}{n_i} \right)}{1 + \sum_{j=1}^N k_j \left( \frac{C_{ej}}{n_j} \right)} \quad (1)$$

The Freundlich standard was applied to binary systems. Assuming  $k_{fi}$  and  $n_i$  to be Freundlich sorption constants ( $i = 1, 2$ ) for each component and  $x_i, y_i, z_i$  to be model parameters, we have,

$$q_i = \frac{k_{fi} C_{ei}^{n_i + x_i}}{C_{ei}^{x_i} + y_i C_{ei}^{z_i}} \quad (2)$$

Adsorption of pesticide species onto forest soil was performed and found to display complex isotherm curves. These isotherm curves are either single step or multiple step<sup>7</sup>. High performance liquid chromatograph was used to determine the individual pesticide loadings in solution. A non-linear statistical model was used in data analysis. A higher step in a

**Table 1.** Information on isotherm models

Model	Variables	Assumptions	Might fail at	Examples
1. Henry	1	1. Low pollution loads 2. Minimised interparticle interactions	1. Intermediate to high pollutant loads	10
2. Dubinin-Radushkevich	2	1. Heterogeneous surface 2. Intermediate and high loading 3. Temperature dependent 4. Gaussian energy distribution	1. Empirical 2. Low pollutant loads	20, 21
3. Elovich	2	1. Kinetics based model 2. Multi-layer adsorption	1. Kinetic data might not show regularity	19
4. Flory-Huggins	2	1. Extent of surface coverage 2. Expresses feasibility of adsorption	1. Mostly valid for polymeric compounds	16
5. Freundlich	2	1. Multiple layer 2. Heterogeneous 3. Physisorption	1. Explaining chemisorption 2. Single layer 3. Varying pollutant load	13, 14
6. Halsey	2	1. Multi-layer adsorption 2. Large distance between adsorbent and pollutant 3. Heterogeneous, Type II	1. High pollutant loads more preferred 2. Fails Henry's law at low sorbent coverage	17, 22
7. Jovanovich	2	1. Langmuir isotherm assumptions 2. Adsorbate-adsorbent mechanical interactions 3. Predicts retention capacity	1. Explaining physisorption 2. Lateral interparticle interaction 3. Hits saturation at high concentrations	17, 18
8. Langmuir	2	1. Single layer 2. Homogeneous 3. Chemisorption	1. Large number of layers 2. Heterogeneous systems 3. Interparticle interactions	11, 12
9. Temkin	2	1. Interparticle interactions 2. Chemisorption 3. Linear change in heat of adsorption	1. Higher and lower ion dilutions 2. Ignores arrangement of adsorbate layer	14, 15
10. Aranovich	3	1. Based on BET 2. Broad concentration range	1. Assumes adsorbent is flat and uniform 2. Non-extensive free energy calculation	29
11. Brunauer-Emmett-Teller (BET)	3	1. Mono and multiple layer 2. Surface adsorption area 3. Homogeneous 4. Higher adsorbed layers have identical adsorption energy, equalling the fusion heat	1. Extremities of the loading spectrum 2. Interaction amid adsorbed species 3. Adsorbent-adsorbate interactions 4. Heterogeneity of surface	22, 29
12. Fowler-Guggenheim	3	1. Inter-adsorbate interaction 2. Linear change in adsorption enthalpy with concentration 3. Exo- and endothermicity explained	1. Very high surface coverage is not explained	28
13. Frenkel-Halsey-Hill	3	1. Multiple layer assumed 2. Holds true to potential theory 3. Adsorbent-pollutant separation distance 3. Inter-layer spacing and interaction 4. Inter-adsorbate interaction	1. Lower concentrations	16

Table-1 (contd.)

14. Harkins-Jura	3	<ol style="list-style-type: none"> <li>1. Multiple layer, Type II adsorption</li> <li>2. Heterogeneous</li> <li>3. Determine surface area</li> </ol>	1. Higher type isotherms	31
15. Hill	3	<ol style="list-style-type: none"> <li>1. Homogeneous</li> <li>2. Cooperative adsorption due to inter-species interaction</li> </ol>	<ol style="list-style-type: none"> <li>1. Heterogeneity</li> <li>2. Semi-empirical</li> </ol>	21
16. Hill-de Boer	3	<ol style="list-style-type: none"> <li>1. Inter-adsorbent and inter-adsorbate interactions</li> <li>2. Agility of the pollutant</li> </ol>	1. Concentration variation affects adsorption capacity	25
17. Jossens	3	<ol style="list-style-type: none"> <li>1. Adsorbate-adsorbent interactions</li> <li>2. Heterogeneous</li> <li>3. Temperature dependent</li> </ol>	1. Chemisorption is not specifically explained	28
18. Khan	3	<ol style="list-style-type: none"> <li>1. Bi-adsorbate systems</li> <li>2. Uptake from pure solutions of high or low dilutions</li> </ol>	1. Higher mixture components	27
19. Koble-Corrigan	3	<ol style="list-style-type: none"> <li>1. Heterogeneous</li> <li>2. High and low concentration</li> </ol>	<ol style="list-style-type: none"> <li>1. Partial empirical</li> <li>2. Trial and error optimization</li> </ol>	21
20. McMillan-Teller	3	<ol style="list-style-type: none"> <li>1. Surface tension effects</li> <li>2. Heterogeneity of substrate surface</li> </ol>	<ol style="list-style-type: none"> <li>1. Empirical</li> <li>2. Lower concentration</li> </ol>	30
21. Radk-Prausnitz	3	<ol style="list-style-type: none"> <li>1. Wide pollutant loading range</li> <li>2. Homogeneous and heterogeneous</li> </ol>	1. Suffers considerably during error analysis	20, 26
22. Redlich-Peterson	3	<ol style="list-style-type: none"> <li>1. Hybrid adsorption mechanism</li> <li>2. Homogenous and heterogeneous systems</li> <li>3. Broad pollutant loadings</li> </ol>	1. Interparticle interactions	16, 21
23. Sips	3	<ol style="list-style-type: none"> <li>1. Heterogeneous</li> <li>2. Dilution, temperature and pH dependence</li> </ol>	<ol style="list-style-type: none"> <li>1. Doesn't follow Henry's law at high dilutions</li> <li>2. Inter-adsorbate interaction</li> </ol>	16, 26
24. Toth	3	<ol style="list-style-type: none"> <li>1. High to low dilutions</li> <li>2. Heterogeneous</li> <li>3. Single and multiple layer</li> </ol>	1. Empirical	23, 24
25. Unilan	3	<ol style="list-style-type: none"> <li>1. Uniform energy distribution</li> <li>2. Heterogenous</li> <li>3. Defines reversibility of adsorption</li> </ol>	1. Empirical	18
26. Baudu	4	<ol style="list-style-type: none"> <li>1. Variation in Langmuir coefficients with loading</li> </ol>	1. High linearization inherent bias	28
27. Dubinin-Astakhov	4	<ol style="list-style-type: none"> <li>1. Heterogeneous</li> <li>2. Multiple layer</li> <li>3. Physisorption</li> <li>4. Temperature dependence</li> </ol>	<ol style="list-style-type: none"> <li>1. Semi-empirical</li> <li>2. Higher and lower ion dilutions</li> </ol>	20, 32
28. Dubinin-Serpinsky	4	<ol style="list-style-type: none"> <li>1. First layer adsorption occurs at primary sites. Hydration occurs leading to hydrogen bonding with and within the latter layers</li> <li>2. Activity of sites reduce as micropore filling increases</li> </ol>	1. Assumes that saturation concentration is achieved at equilibrium	32

Table-1 (contd.)

29. Guggenheim Anderson de-Boer (GAB)	4	1. Combines Langmuir and BET 2. Multiple layer, physisorption 3. Inter-layer interaction and potential difference between layers	1. An advanced model having same assumptions as base models.	17, 22
30. n-layer BET	4	1. Space for adsorption is finite 2. Only 'n' number of layers are adsorbable 3. Pollutant-substrate interaction 4. Energy of first layer adsorption is greater than fusion heat	1. As 'n' approaches infinity, it reverts back to BET	34
31. Weber-Van Vliet	4	1. Fits large number of experimental data systems with minimal least squared errors 2. Fitting of data by multiple non-linearity	1. Empirical	33
32. Anderson	4, 5	1. Adsorption area is lower in progressing layers 2. Finite adsorption layers 3. All concentrations of pollutant	1. May fail for highly efficient sorbents	35
33. Fritz-Schlunder	3, 4, 5	1. Fits variety of equilibrium data 2. All concentrations of pollutant 3. Very stable model and handles model variations well	1. Complex 2. Empirical	15, 28

multiple step isotherm was computed by summation of each previous step. Assuming,  $s$  represents the total number of steps, and  $b$  is the limit of the sorption mechanism concentration (mg/L),

$$q_i = \sum_{i=1}^s \left\{ \frac{q_{mi} k_i [(C_e - b_i) + |C_e - b_i|]}{2 + k_i [(C_e - b_i) + |C_e - b_i|]} \right\} \quad (3)$$

A unified Freundlich-Langmuir model was proposed to find  $q_e$  of an individual component in a mixture of  $N$  constituents by utilising the complete range of multiple-solute sorption data<sup>8</sup>. It is shown in eq. (4). Here  $j$  represents an additional pollutant in the mixture and  $N$  is the total number of pollutants.

$$q_i = q_{mi} k_i C_{ei} \left( \sum_{j=0}^N k_j C_{ej} \right)^{n_i} \quad (4)$$

Adsorption onto soils may be characterized by a distributed reactivity model (DRM). It helped portray inherent heterogeneities which in turn simplified understanding of irregularities in non-linear modelling and error reduction<sup>9</sup>. In eq. (5), while  $m_j$  as the summed mass fraction and  $k_D$  as the mean

partition coefficient for linear sorptions onto sorbents,  $m_{nl}$  represents the non-linear portion of the adsorbed mass. Typically, the pollutants vary in number from  $i = 1$  to  $N$ , with  $N = 2$  for binary systems.

$$q_i = m_j k_D C_e + \sum_{i=1}^N (m_{nl})_i k_f C_e^{n_i} \quad (5)$$

A multiple layer, heterogeneous adsorption system was represented graphically<sup>36</sup>. A visual representation yielded that multiple layers adsorb onto sites with high capacities before mono layer sorption begins on weakly sorptive sites.

Physisorption onto solids was described by utilising the merits of the density functional method<sup>37</sup>. The method holds true for both solids with high and low porosities. A comparison of model outputs with experimental values reveals high concordance. If  $\rho$  is the density variation from surface in outward direction  $z$  and  $\rho_0$  is the density variation in absence of wall interactions, then their difference gives us free surface  $q_e$  as in eq. (6),

$$q_e = \int (\rho - \rho_0) dz \quad (6)$$

A unified approach to represent different types of isotherm curves from type I to VI is given by eq. (7). Patch wise adsorption onto sites with variable energies was studied<sup>38</sup>. The sites with maximum frequency of adsorption energy are averaged as  $a_i$ , probability factor  $a_i$  and the standard deviation  $m_i$ . Adsorption equilibrium constant  $k$  is the inverse of  $p_s$ , the pressure at saturated sorption.

$$q_e = \sum_{i=1}^n a_i \left\{ \frac{\left( \frac{p}{p_s} e^{\varepsilon_i/RT} \right)^{RT/m_i}}{1 + \left( \frac{p}{p_s} e^{\varepsilon_i/RT} \right)^{RT/m_i}} \right\} \quad (7)$$

Majority of adsorption studies are equilibrium based. Mono component, non-equilibrium data was modelled by combining classical equations with intra-particle diffusion models<sup>39</sup>. In case of irregular and extended breakthroughs, this model reduced errors and improved predictions considerably. Evaluation of chromatographic column analysis was done by a parabolic dilution curve for intra-particle diffusion. The concepts equated to each other were,

$$q_n = \bar{q} + \frac{(C - C_s^*)B_i}{5} \quad (8)$$

$$q_n = \frac{q_m k C_s^*}{1 + k C_s^*} \quad (9)$$

We have,  $q_n$  as non-equilibrium sorption capacity,  $\bar{q}$  is the average sorption capacity, dependant on time and bed height.  $B_i$  is the Biot number,  $C_s$  is the pollutant concentration and  $C_s^*$  is the pollutant concentration under dynamic settings.

Certain aromatics like aniline were extracted from solution by using activated carbon. A blend of Joranovic method, building on Langmuir method was used in order to generate a model with very low error<sup>40</sup>. Studies were conducted on activated carbon and found that sorption increased with lower sorbent particle size, lower solubility of pollutant in water and higher molecular weight of the pollutant. If  $r$ ,  $p$  and  $z$  are isotherm constants, then the eq. (10) is,

$$q_e = \frac{[1 - e^{-p C_s^z}] r C_e}{1 + C_e} \quad (10)$$

Analysis of heavy metal sorption onto substrates was aided

by development of ion-exchange isotherms, as given by eq. (11). This sorption phenomenon is different from both physisorption and chemisorption<sup>41</sup>. The terms  $i$  and  $j$  denote the metal ion species in the bi-component mixture, whereas  $x$  and  $y$  are the equivalent fraction of a component in liquid and solid phase respectively.

$$Y_i = \frac{1}{1 + \frac{x_j}{k_{ij} x_i}} \quad (11)$$

A sorption system not favouring a forward reaction was studied based on the local equilibrium theory which ignores the concentration gradient between solid and solution phase<sup>42</sup>. We have,  $e$  as the porosity inherent to the sorbent material,  $v$  as the velocity of liquid in interstices,  $dq_e/dC$  is the gradient of capacity with concentration and  $Z$  is the bed length. Sorption time available  $t$  is given by eq. (12),

$$t = \frac{Z}{v} \left[ 1 + \left( \frac{1-e}{e} \right) \left( \frac{dq_e}{dC} \right) \right] \quad (12)$$

### Effect of different parameters on isotherm modelling

Various processes affect how a sorbent-sorbate system behaves. Process analysis, simulation and optimization involve inclusion of the major processes and parameters that affect sorption. Due to model simplification, some factors are omitted from equations. Analysis of these variables is also of concern and have been studied as given below.

#### Effect of pH:

Dependence of sorption efficiency on pH was studied and found to vary in direct relation with affinity coefficient<sup>43</sup>. The analytical relation and the mono-component model proposed is given by eq. (13). Cationic competitiveness exists among charged species for sorption onto sites of the sorbent<sup>44</sup>. This has been seen to affect the pH of the solution and thus the sorption capacities as realized by a study conducted on cadmium and copper.

$$\log K = -0.95 \text{ pH} + 7.35 \quad (13)$$

$$q_n = \frac{q_m (C_e k \text{ pH})^n}{1 + (C_e k \text{ pH})^n} \quad (14)$$

#### Effect of moisture content:

Smaller moisture content on adsorbent was in direct correlation with elevated curves during modelling. An exponen-

tial and empirical relation exists between moisture content and adsorption capacity<sup>46</sup>. With  $M$  as moisture (%) and  $c$ ,  $d$  as empirical constants reliant on  $C_e$ , we may interpret the relation as,

$$q_e = cM^d \quad (15)$$

*Temperature:*

Temperature is a very important factor which governs both adsorption capacity and structural integrity of the adsorbent. A gas adsorption model was developed as given by eq. (16). This relation can be extended to an aqueous solution of pollutants, so as to relate temperature with adsorption extent<sup>46</sup>. Considering  $T$  to be temperature in °C,  $V$  as adsorbed pollutant content and  $x$ ,  $y$  as functions of temperature and concentration,

$$\frac{T}{V} = xT + y \quad (16)$$

*Site energy distribution:*

Distribution of energy over sorbent surface is assumed to be either homogeneous or heterogeneous. An extension of Toth model was derived, incorporating effects of energy distribution directly into the model<sup>47</sup>. An exponential quasi-Gaussian variation of site enthalpies was observed at temperatures as low as 77 K. The leftward broadening of the graph indicates that the statistical mean energy is higher than majority of the values. If  $E_{\max}$  is the maximum site energy and  $E_{\min}$  is the minimum, then,

$$E_{\max}^* = -\ln\left(\frac{C_{\max}}{C_e}\right)RT \quad (17)$$

A similar equation for  $E_{\min}$  is applicable. With  $C_{\max}$  (maximum concentration of pollutant allowable) and  $C_e$  (equilibrium concentration of pollutant in solution)<sup>48</sup>.

## Conclusions

Industrial pollution abatement is of prime significance. With that goal in mind, to suit each industrial need, several mathematical isotherm models for adsorption have been developed. Researchers have made alterations to previous models by changing experimental parameters affecting the process. Each isotherm model begins with certain assumptions and builds upon them to produce the desired outcome. Non-linearization in regression studies have proven to reduce errors and provide better concordance with experimental

data. Existing isotherm models, their assumptions and shortcomings have been discussed. Different adsorption modelling approaches have been mentioned. This should help the reader apply an appropriate model to study their system. Inclusion of intrinsic factors into modelling equations has also been discussed for better evaluation of their effects.

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