

## Simulation Of Multilayered Packed Bed Adsorption Column For Tetracycline Removal From Aqueous Environment

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### ABSTRACT

The widespread use of tetracyclines in humans and animals, which ultimately enter the aquatic environment, has become a major environmental and public health problem because of their persistence, toxicity, and role in antimicrobial resistance. Because of the difficulty to completely remove these emerging contaminants, conventional treatment technologies are not adequate, and advanced and efficient treatment technologies are required. In this work, the simulation and optimization of a multilayered packed bed adsorption column have been explained for adsorption of tetracycline from the aqueous environment in Aspen ADSIM V10 software. The proposed multilayer configuration is a novel approach that combines different adsorbent materials in a series to make use of the complementarity of the adsorption properties, in order to increase the removal of contaminants and to improve the use of the beds. Considering the different operational parameters such as influent concentration, flow rate, bed height, and the arrangement of the adsorbent layer, a dynamic adsorption model is developed to explore the mass transfer behavior, adsorption kinetic characteristics, and breakthrough characteristics. Thorough breakthrough curve studies are carried out to assess column performance and to determine important design parameters that will affect the efficiency of the adsorption process. In addition, process optimization is carried out to find the optimum multilayer configuration for the best removal of tetracycline while minimizing the use of adsorbent and the operating costs. The simulation framework allows for predictive evaluation of the system performance without going through extensive experimental trials, which is a cost-effective method to perform the design and scale up of adsorption processes. This proposed multilayered adsorption system is expected to show better performance than the traditional packed bed system, and will be a new and sustainable solution to the treatment of water contaminated with antibiotics. The results of this work will help develop new technologies for cleanup of emerging pollutants via scalable adsorption processes and contribute to the development of simulation based environmental process

engineering.

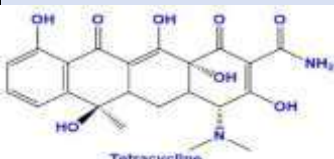
**Keywords:** Adsorption, Aspen ADSIM, Breakthrough Analysis, Dynamic Simulation, Multilayered Packed Bed, Process Optimization, Tetracycline, Water Treatment.

## INTRODUCTION

Water is an essential, renewable resource that is important for life, agriculture, and economic growth. However, it is problematic to depollute and irreplaceable in most of its uses. Although freshwater ecosystems cover only a small percentage of the Earth's surface, they play a significant role in supporting biodiversity and human requirements. Adsorption is the most commonly used method because of its cost effectiveness, ease of process, and effectiveness. A variety of absorbents have been discovered, including activated alumina, clay, carbon, iron based materials, and bio based absorbents. Among them, activated alumina is reorganized by the US EPA and WHO as one of the best technologies for impurity removal. Despite this acknowledgement, significant marketable applications remain limited because most investigations have been conducted at the laboratory scale. Scaling up is tested by high costs, time consumption and compound optimization of operating parameters. To address this gap, the existing study suggests a simulation based tactic using Aspen ADSIM V10. Water is an ordinary renewable reserve that can be managed by storage, transfer, or reprocessing. Water plays a vital role in supporting life, agriculture, economic growth, and overall comfort. However, it is difficult to clean once polluted, expensive to move, and rare for most uses. Water sources such as rivers, lakes, and groundwater are crucial for many activities such as farming, hydropower, industry, and recreation. Although freshwater covers only 0.5% of the Earth's surface and rivers hold only 0.01% of global water, they are still vital [1]. The requirement for water has been increasing steadily, straining water quality owing to industrialization, urbanization, and farming. In developing countries, such as Nigeria, poor sanitation and waste disposal exacerbate the problem. In the Sabon Birni Local Government Area, the main source of drinking water, groundwater, is at risk due to latrines, agricultural runoff, and unmanaged waste [2]. Water is critical for drinking, agriculture, and industry, but pollution from natural and human actions poses global health hazards. Approximately 80% of sewage is discharged untreated, contributing to more than 50 diseases and producing 80% of illnesses and 50% of child deaths worldwide. This connects water pollution to diseases such as diarrhea, skin conditions, and cancer, with impacts varying by region, age, and gender [3]. Pharmaceuticals develop impurities due to their persistence in marine environments; however, incomplete facts exist on their lasting environmental and health risks. This study reviewed research (2006–2022) on pharmaceutical occurrence in water across 14 MENA countries, identifying 159 dynamic compounds, with antibiotics being the most common (0.1–99,636 ng/L). Risk calculations revealed that 39 pharmaceuticals pose ecological risks and 8 pose potential human health risks. Compounds such as 17 $\beta$  estradiol, diclofenac, and carbamazepine exhibited particularly high environmental risk (RQ > 1000). Hormones showed the highest health risk via drinking water and fish

digestion, whereas ciprofloxacin and norfloxacin presented the highest antimicrobial resistance risk. Fate modeling training was planned for the most polluted MENA river but could not continue due to inadequate geospatial information, highlighting the requirement for improved monitoring and regulatory action in data scarce areas [4]. The general practice of medicines in healthcare, agriculture, and 12 activities has led to increased global environmental contamination and rising concerns about antibiotic resistance, mainly in developing countries [5]. A study on the Buriganga River reported seasonal variations in the occurrence of 29 antibiotics, with higher concentrations detected specifically during the dry period. Furthermore, several antibiotics exceeded resistance thresholds, indicating a genuine hazard to microbial ecosystems and public health. These findings underscore the need for stringent regulations and improved wastewater management to moderate antibiotic pollution in marine environments [6]. Tetracyclines have played a vigorous role in antimicrobial therapy for over 70 years because of their broad spectrum efficiency against various pathogens. This review highlights the pharmacological characteristics, clinical applications, and tests of traditional and modern tetracyclines, including doxycycline, minocycline, tigecycline, and metacycline. Despite the emergence of resistance, newer derivatives have better therapeutic potential and broader clinical use, maintaining the relevance of tetracyclines in managing both common and emerging infections [7]. Although very few studies have been conducted on the occurrence of antibiotics in water and wastewater in Pakistan, the fate and transport of different antibiotics have been detected in ground and underground water. Zainab et al., (2021) performed a fate and transport of 23 antibiotics in groundwater of three major cities including Islamabad, Lahore, Peshawar, Quetta, Faisalabad, Gujrat, and Karachi. The obtained results revealed that among the 23 antibiotics, ciprofloxacin and tetracycline were detected at the highest concentrations [8]. Tetracyclines are broad spectrum antibiotics that contain four hydrocarbon rings. This was previously reported in medicine in the 1940 s [9]. An organization the Drug Administration (FDA) and U.S. Food and accepted the usage of TCs in 1950 to encourage physical health and used an additive in animal food [10]. This type of antibiotic consists of four key elements: chlortetracycline (CTC), oxytetracycline (OTC), doxycycline (DC), and TC itself [11]. Such composites participate in a parallel chemical structure, resulting in a hydro naphthalene fundamental completed by four unified rings [12]. The chemical structures of TC, CTC, OTC, and DC are associated with their significant physicochemical properties. In particular, such mixtures display high water solubility [13]. The table 1 shows the properties and structures of different classes of tetracycline.

**Table 1: Properties and Structures [22].**

| Forms of Tetracycline | Molecular Formula                                             | Molecular Mass | Water solubility at 25°C(mg/L) | Chemical Structure                                                                                        |
|-----------------------|---------------------------------------------------------------|----------------|--------------------------------|-----------------------------------------------------------------------------------------------------------|
| TC                    | C <sub>22</sub> H <sub>24</sub> N <sub>2</sub> O <sub>8</sub> | 444.4          | 231                            |  <p>Tetracycline</p> |

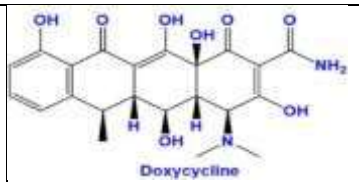
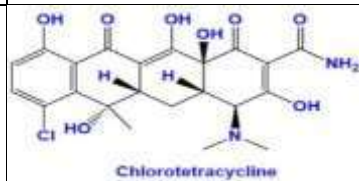
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|     |                           |       |     |                                                                                                           |
|-----|---------------------------|-------|-----|-----------------------------------------------------------------------------------------------------------|
| OTC | <chem>C22H23ClN2O8</chem> | 478.8 | 630 | <br>Doxycycline        |
| CTC | <chem>C22H24N2O8</chem>   | 444.4 | 630 | <br>Oxytetracycline    |
| DC  | <chem>C22H24N2O9</chem>   | 460.4 | 313 | <br>Chlorotetracycline |

Tetracyclines in humans and animals are the chief suppliers of ecological effluence. As distinguished by Xie et al., tetracycline antibiotics cannot be fully absorbed and metabolized inside the body, with approximately 75% of the antibiotic being expelled in an innovative arrangement [14]. TC pollution is mostly considered a source of water, followed by significant biological interruptions in the environment. The primary factors related to TC effluence in marine systems include the misuse of antibiotics in human and veterinary medicine. [15]. Few degradation products of TCs exhibit biological activity, posing hazards to living organisms unprotected from contaminated wastewater comprising the remains. Lengthy contact with unnecessary quantities of TCs is related to health threats, toxic belongings, and bacterial resistance. Such conditions may involve gastrointestinal disturbances, renal impairment, intracranial hypertension, and dermatological manifestations, such as rosacea and perioral dermatitis. [16]. These features show serious concerns about food safety and environmental wellbeing, which can be the reason for allergic reactions, antibiotic resistance, and toxicity in living things, as illustrated in Figure 1.



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Figure 1: Health risk associated with TCs [22].

The overall adsorption process can occur in two different ways: physical or chemical adsorption. The binding forces responsible for physical adsorption include size discriminating adsorption,  $\pi\pi$  interactions, filling of pores, and van der Waals forces, in which pollutants adhere to the surface of the adsorbent. Pollutant adsorption on the surface of an adsorbent is often referred to as physisorption. [17]. The process is reversible because pollutants do not interact chemically with the available vacant sites on the surface of the adsorbent; therefore, adsorbed pollutants can be easily removed from the adsorbent surface. Chemisorption is the adsorption process in which absorbed pollutants form a chemical bond by exchanging surface charges. The bond formation between the adsorbate and adsorbent material might be due to electron pairing, Lewis acid interactions, oxidation or reduction, and ion or lagend exchange. [18]. The core mechanism of pollutant adsorption onto an adsorbent through physical and chemical processes is shown in Figure 2.

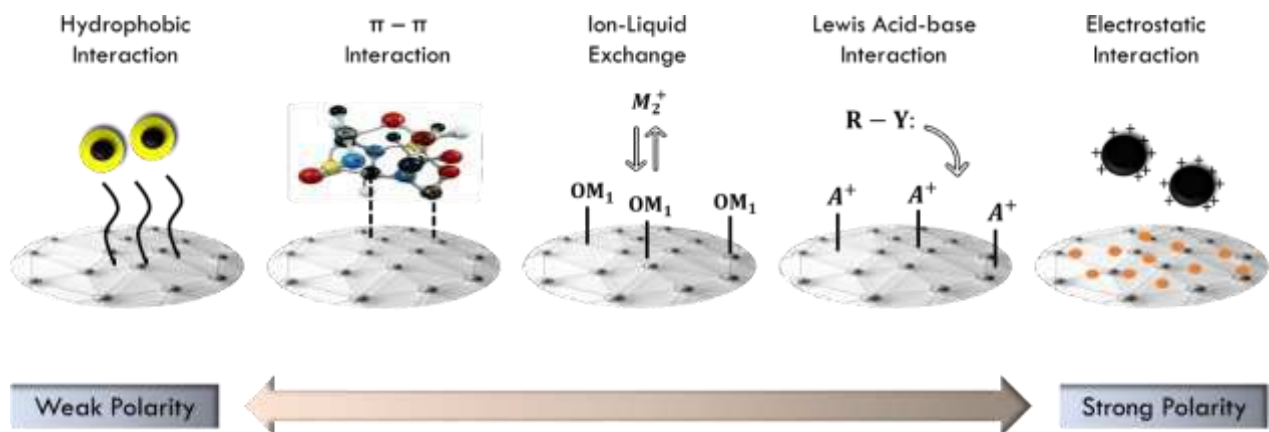


Figure 2: Main Sorption Mechanism of pollutants on to a 2DMxene based nanomaterials [28].

Bentonite (BN) and its acid-treated form (BA1) have been explored for their ability to eliminate tetracycline hydrochloride (TC) from aqueous solutions because of their maximum surface area, cation exchange capacity, and cost-effectiveness. Complete characterization (XRF, XRD, SEM/TEM, XPS, TG/DTG, and gas adsorption isotherms) revealed that BA1 is a mesoporous material with a surface area of  $>165 \text{ m}^2/\text{g}$  [19]. Existing wastewater treatment methods are insufficient for removing these antibiotics, resulting in the contamination of oceans, surface water, groundwater, and even intake water. This study frame worked the sources, impacts, and spread of tetracycline pollution and reviewed conventional treatment techniques, including adsorption, biological, physical, and chemical processes [20]. The table 2 shows the adsorbents material used for TCs removal.

Table 2. Adsorbents material

| Adsorbent Materials | Co<br>mg/l | Exp. Conditions |              |            | qm<br>mg/g | References |
|---------------------|------------|-----------------|--------------|------------|------------|------------|
|                     |            | pH              | dose<br>mg/l | Temp<br>oC |            |            |
|                     |            |                 |              |            |            |            |

|                          |     |      |     |    |         |      |
|--------------------------|-----|------|-----|----|---------|------|
| GACox                    | 100 | 4    | 400 | 25 | 714     | [21] |
| Zeolites                 | 100 | 3    | 50  | 50 | 244.63  | [22] |
| Chitosan                 | 200 | 8.5  | 100 | 30 | 1795.01 | [23] |
| Palm leaves biomass      | 20  | 5.86 | 60  | 25 | 132     | [24] |
| Corn straw               | 80  | 7    | 100 | 25 | 1368    | [25] |
| Wheat straw              | 80  | 7    | 100 | 25 | 1438    | [26] |
| Sunflower seed husk      | 50  | 6    | 100 | 40 | 429.3   | [27] |
| GO/gC3N4Fe3O4            | 50  | 3    | 30  | 25 | 120     | [28] |
| Fly ash                  | 40  | 4    | 90  | 30 | 30.72   | [29] |
| Palm leaf                | 50  | 7    | 90  | 45 | 13.71   | [30] |
| Pear tree wood           | 500 | 7    | 500 | 50 | 930.3   | [31] |
| Magnetic Graphene oxide  | 50  | 45   | 66  | 40 | 106.6   | [32] |
| Deedleaf derived biochar | 100 | 6    | 400 | 25 | 336.7   | [33] |
| MnFe2O4/rGO              | 10  | 3.3  | 50  | 25 | 41      | [34] |

GACox= Surface Oxidized granular activated carbon; MGO=Magnetic Graphene oxide; DD = diethylenetriaminepentaacetic acid; MSCG = Fe3O4@SiO2–chitosan/GO; NSC = nitrogen/Sulphur carbon dots; MSPP = Magnetic starch polyurethane polymer

## MATERIAL AND METHODS

### Overview:

The present research proposes a simulation model to assess and improve the performance of singlelayered and multilayered packed bed adsorption columns for the removal of tetracycline from aqueous environments. The methodology combines the adsorption

modelling, dynamic breakthrough analysis and process optimization by Aspen ADSIM V10. First, physicochemical and adsorption parameters of tetracycline and some low-cost adsorbents are extracted from literature and fed into simulation environment. The choices of the candidate adsorbents are the Activated Alumina, Natural Zeolite and China Clay because the materials are available and economically feasible, have adsorption capability and complementary surface characteristics. A dynamic packed bed adsorption model is developed in Aspen ADSIM V10 to simulate the phenomena of mass transfer, adsorption equilibrium and contaminant transport in the column. Two configurations are examined: a single layer adsorption column and an innovative multilayered adsorption column with sequential layers of adsorbent to capture the contaminant as much as possible across the column.

### Data Collection and Analysis:

Data were gathered from journal articles, experimental investigations, adsorption databases, and simulation based studies focusing on antibiotic remediation using low-cost and environmentally sustainable adsorbents. Particular emphasis was placed on adsorption equilibrium characteristics, kinetic parameters, mass transfer coefficients, adsorbent properties, and fixed bed column performance indicators. The collected information was systematically analyzed to identify suitable adsorbents for multilayer column configuration, including Activated Alumina, Natural Zeolite, and China Clay. Key parameters such as adsorption capacity, surface area, porosity, particle size, bed

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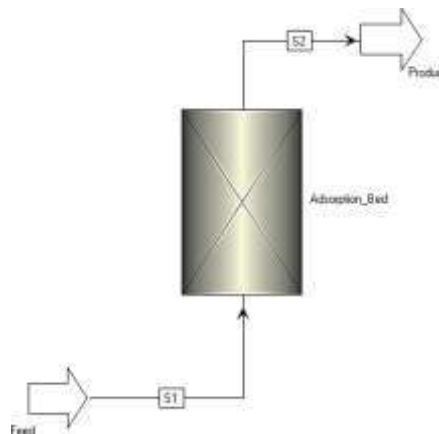
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density, influent concentration, pH, and flow rate were extracted and standardized for simulation purposes.

## Aspen ADSIM Model Development

Aspen ADSIM V10 was employed to develop and simulate the adsorption system to study the removal of tetracycline (TC) from aqueous solutions under dynamic packed bed operating conditions. A simulation framework was developed to assess and compare the adsorption performance of single and multiple layer packed bed adsorption columns.

The single layer adsorption model is a simplified model which contains a feed (S1), an adsorption bed and a product (S2) as shown in Figure 3-5. The influent containing tetracycline is introduced into the adsorption column via stream S1 and does not reach the treated effluent (S2) until after it has flowed through the adsorbent bed where the contaminant is removed. The Multilayer configuration is made up of 3 adsorption



layers in sequence: Activated Alumina, Natural Zeolite and China Clay. The different layers give different adsorption properties and offer extra adsorption sites for the capturing of tetracyclines. The sequential arrangement

Figure 3: Aspen ADSIM model of the single layer packed bed adsorption column for tetracycline.

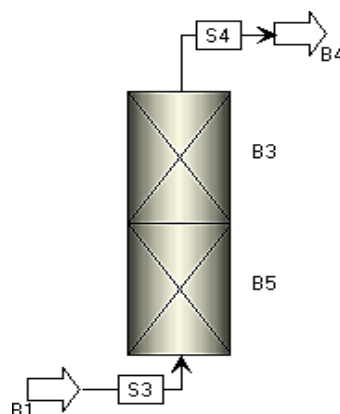


Figure 4: Aspen ADSIM model of the Duallayer packed bed adsorption column for tetracycline.

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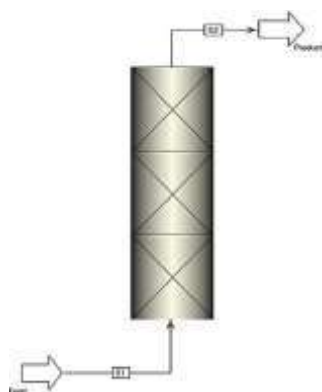


Figure 5: Aspen ADSIM model of the multilayer packed bed adsorption column for tetracycline.

### Selection of Adsorbents:

The selection of adsorbents was carried out through a systematic evaluation of materials reported in the literature for the effective removal of tetracycline and other pharmaceutical contaminants from aqueous solutions. Activated Alumina, Natural Zeolite, and China Clay were identified as suitable adsorbents due to their low cost, widespread availability, environmental compatibility, and demonstrated adsorption performance. These materials possess distinct physicochemical characteristics that contribute to different adsorption mechanisms, enabling enhanced contaminant capture when utilized in a multilayer arrangement. Critical adsorbent properties, including specific surface area, pore volume, porosity, particle size distribution, bulk density, and adsorption capacity, were collected from published studies and incorporated into the simulation model.

### Theoretical Assumptions

The dynamic simulation of the packed bed adsorption column was developed based on the following assumptions to simplify the modeling process while maintaining reliable prediction accuracy:

The adsorption column was discretized using the Upwind Difference Scheme (UDS1) with 200 computational nodes to accurately represent axial concentration gradients along the column length.

Mass transfer within the adsorption system was represented using a lumped mass transfer model, accounting for both external film diffusion resistance and intraparticle diffusion resistance as the controlling transport mechanisms.

Adsorption equilibrium between the aqueous phase and adsorbent surface was assumed to follow the Langmuir Isotherm Model, which describes monolayer adsorption on homogeneous adsorption sites.

Temperature and pressure were maintained constant throughout the simulation, and thermal effects on adsorption performance were neglected.

## Mathematical Modelling

The precise mathematical formulation of the packed bed adsorption column will be derived through differential mass balance analysis of tetracycline (TC) between the fluid and solid phases. By defining a control volume of height  $\Delta z$  and cross-sectional area  $A$ , the transport of TC through the column will be characterized considering axial dispersion, interphase mass transfer, and the effect of bed porosity  $\epsilon$ . Also, volume of the solid  $V_s$  will be used through the Equation (1).

$$V_s = (1 - \epsilon)\Delta z \quad \text{--- (i)}$$

The four terms related to accumulation, convection, axial dispersion and adsorption were added to the governing equation as shown in Figure 6. Overall mass balance for packed bed system is given by Equation (2).

$$\begin{aligned} & \frac{\partial C}{\partial t} \\ & \frac{\partial Q}{\partial C} \\ & \frac{\partial C}{\partial z} \\ & (u_o \epsilon C)_z - (u_o \epsilon C)_{z+\Delta z} - (\epsilon E_z \frac{\partial C}{\partial z})_{z+\Delta z} + (\epsilon E_z \frac{\partial C}{\partial z})_z \\ & - \rho \Delta z \frac{\partial Q}{\partial t} = \epsilon \Delta z \frac{\partial C}{\partial t} \quad \text{--- (ii)} \end{aligned}$$

Where,  $E_z$  is the coefficient of the axial dispersal expressed in  $m^2/s$ ,  $u_0$  describes interstitial velocity expressed in the unit  $m/s$ ,  $z$  signifies the position of the axial adsorption bed,  $\rho_s$  is the adsorbent bed column density expressed in  $kg/m^3$ , the overall process time in minute is described by  $t$ ,  $Q$  describes the adsorption capacity of the pollutant and expressed in  $mg/g$ , and  $C$  is the concentration of TC in the water ( $mg/L$ ).

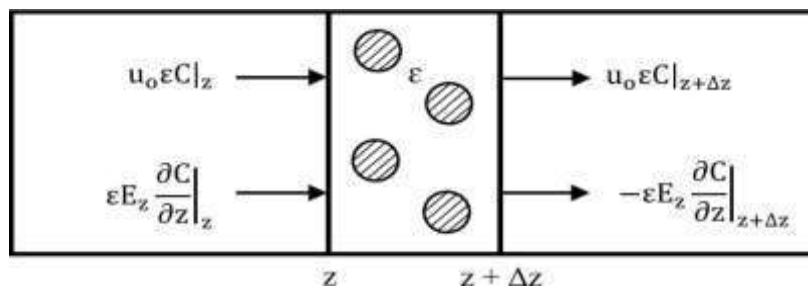


**Figure 6: Mass balance of control volume[59]**

## Determination of Adsorption Rate

Kinetic mass transmission coefficient is sufficiently predictable to convey the LDF guess spread from force for contaminants is a rectilinear purpose of the solid phase loading. LDF prototypical is regularly considered in the procedure because it is mathematically unpretentious. The rate of adsorption expressed as  $dQ/dt$  and LDF model based on the rate of adsorption can be expressed through the Equation (3).

$$\frac{\partial Q}{\partial t} = k(Q^* - Q) \quad \text{---(iii)}$$

LDF

As kLDF describes the coefficient of mass transfer (1/s). The linear driving force constant (kLDF) accounts for dual mass transfer resistances, namely the external film diffusion resistance characterized by the mass transfer coefficient  $k_f$  (m/s), and the intraparticle surface diffusion resistance represented by  $k_s$  (m<sup>2</sup>/s). The kLDF,  $k_f$ , and  $k_s$  can be calculated through the equation described in Table 3.

## Adsorption Equilibrium Isotherm

At equilibrium conditions, the interaction between tetracycline (TC) molecules and the surface of the selected of the adsorbent materials was described using adsorption equilibrium isotherms. These models allow the quantitative relationship of the amount of tetracycline adsorbed onto the adsorbent, with the concentration of the residual tetracycline in the aqueous phase, at a fixed temperature. Equilibrium behavior is fundamental for understanding the adsorption mechanism and is crucial to the accurate prediction of the performance of packed bed adsorption systems. In this study, the adsorption isotherm models most commonly used were taken into account: Langmuir and Freundlich isotherms.

## Numerical Method

This software considers decoding expressions of the ordinary differential equation (ODEs), and partial differential equation (PDEs), and expression of algebraic terms for the packed bed adsorption column proto typical. Firstly, Eq. 3 to 5 is spatially discretized among nearly 200 nodes in the axial track by means of the limited difference scheme, which is generally called as UDS1, also stated in Section 2.1, also transforms the PDEs to ODEs and then ultimately to algebraic equations. The terms of the UDS1 for altering the differential equations expressed as DAEs and PAEs can be articulated as the Eq. 11 and 12 [57].

$$\frac{\partial T}{\partial z} = T_i - T_{i-1} \quad \text{---(XI)}$$

$$\frac{\partial^2 T_i}{\partial z^2} =$$

$$\frac{\partial^2 T}{\partial z^2} = \frac{T_{i+1} - 2T_i + T_{i-1}}{\Delta z^2} \quad \text{---(XII)}$$

Although (T) and (i) indicate the variable and place. Resultant scheme of DAEs is resolute instantaneously, w.r.t space and period.

## Simulation Procedure

A simulation procedure was developed and executed with the Aspen ADSIM V10 software, which was used to model the dynamic adsorption of tetracycline (TC) in single and multilayered packed bed adsorption columns. Tetracycline was not found in the Aspen property database, so it was first defined as a user specified component based on its physicochemical properties that were published in the literature. The adsorption performance of each of the three materials (Activated Alumina, Natural Zeolite, China Clay) were separately analyzed by simulations. These adsorbents were then combined into a multilayered packed bed system to explore the combined effect of successive layers of adsorbents. Figure 7. Proposed Aspen ADSIM Based Simulation Framework for Tetracycline Removal in Single Layered and Multi Layers Packed Bed Adsorption Column.

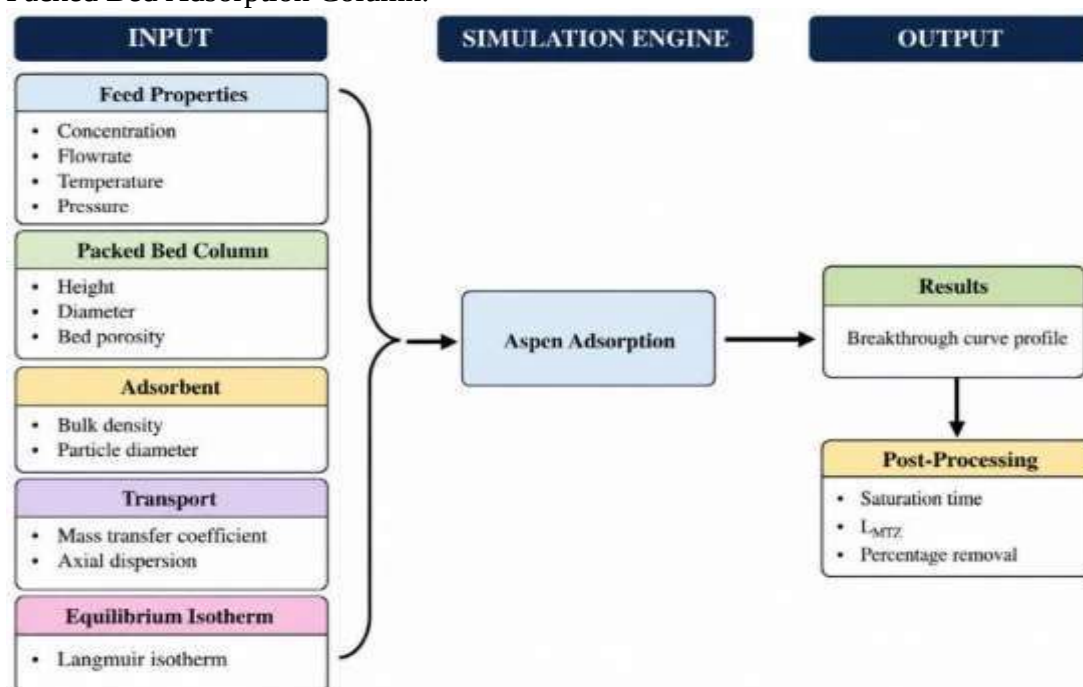


**Figure 7: Proposed Aspen ADSIM simulation framework for tetracycline adsorption**

## RESULTS AND DISCUSSION

### Breakthrough Curve Analysis of Tetracycline Adsorption

The adsorption characteristics of the tetracycline (TC) was studied in packed bed adsorption columns under dynamic condition both at single and multilayer adsorption layer. The adsorption characteristics of tetracycline (TC) was examined in packed bed

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adsorption columns under dynamic condition (single and multilayer adsorption layer). The simulated breakthrough curves of all the adsorption systems (China Clay, Natural Zeolite, Activated Alumina, Dual Layer (AA + NZ) and Triple Layer (AA + NZ + CC)) are shown in Figures 8-12.

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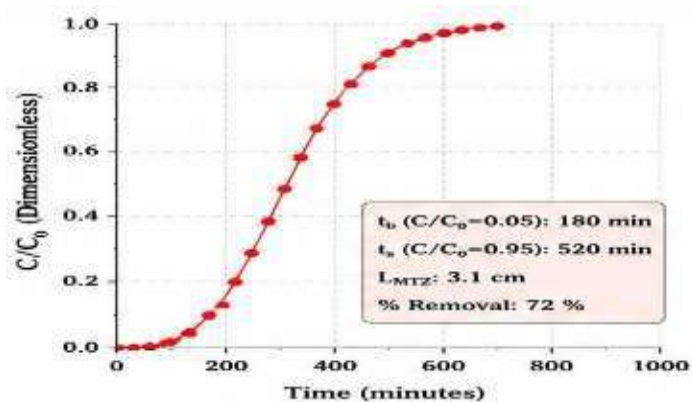


Figure 8: Proposed Aspen ADSIM simulation framework for China Clay

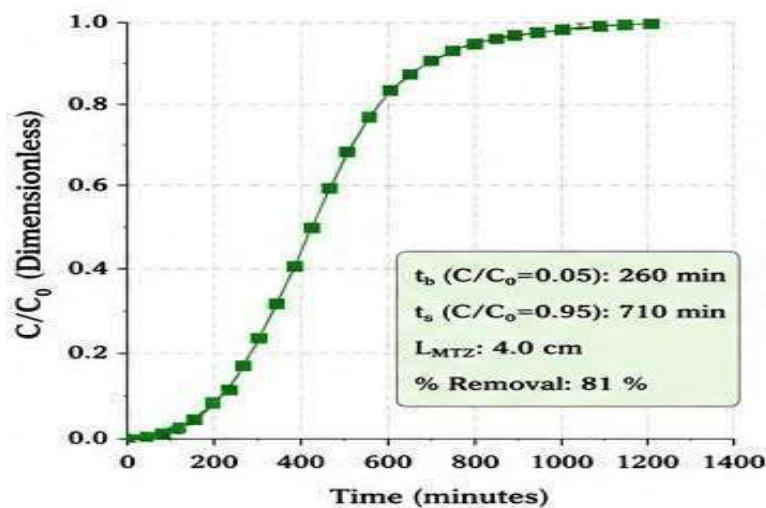


Figure 9: Proposed Aspen

ADSIM simulation framework for Natural Zeolite

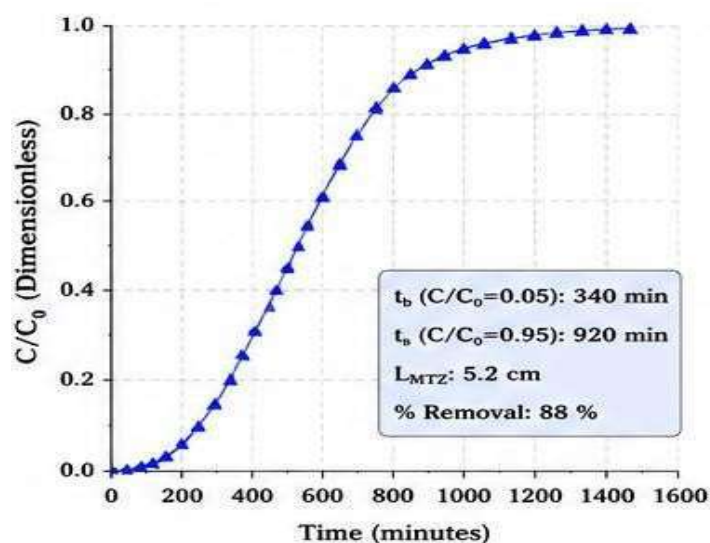


Figure 10: Proposed Aspen ADSIM simulation framework for Activated Alumina

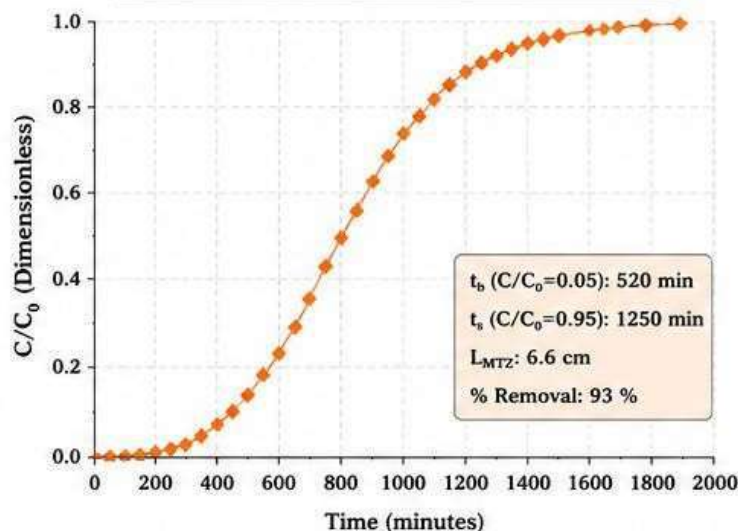


Figure 11: Proposed Aspen ADSIM simulation framework for Dual Layer

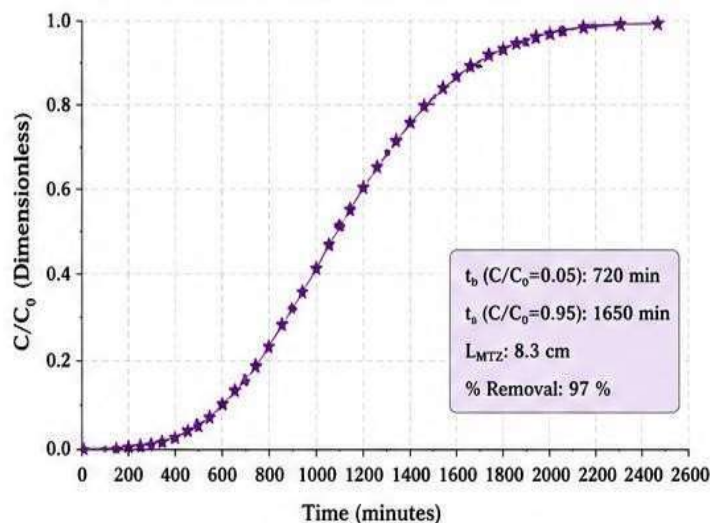


Figure 12: Proposed Aspen ADSIM simulation framework for Triple Layer

The effect of the adsorbent configuration on breakthrough behavior was revealed by the simulation results. The single layer systems showed the lowest levels of performance, with China Clay showing breakthrough time of ~180 min and saturation after 520 min, while Natural Zeolite showed breakthrough time of 260 min and saturation after 710 min. Activated Alumina exhibited the best adsorption properties of the adsorbents tested and the breakthrough time was about 340 min, and the saturation time for the column was 920 min. The higher surface activity and the stronger adsorption affinity of tetracycline molecules on the improved Activated Alumina is the reason for the improvement in its performance. The breakthrough profiles have the typical sigmoidal shape typical of fixed bed adsorption systems. In the initial stages, the concentration in the effluent was almost negligible as there were plenty of active adsorption sites in the column.

## Comparison of Single Layer and Multilayer Systems:

A comparative analysis of all the investigated adsorption configuration points out the advantages of the multilayer packed bed systems. Although each adsorbent performed reasonably well in the adsorption process, the adsorption capacities of each adsorbent were restricted by the finite number of adsorption sites available. As these sites filled up, breakthrough happened very quickly, lowering the general effectiveness of treatment. The triple layer system provided about four times longer breakthrough time than the China Clay column and over two times as long a service life as the Activated Alumina system. As a result, the results from the simulation also validate the effectiveness of the multilayer adsorption technology option for overcoming the limitations of single adsorbent systems in order to achieve better fixed bed adsorption performance.

## Result Analysis:

The Aspen ADSIM V10 simulations were able to accurately predict the dynamic adsorption characteristics of tetracycline in single layer and multilayer packed bed adsorption columns. The breakthrough curves generated showed that the column performance, breakthrough and saturation time, and overall removal efficiency of tetracycline was clearly affected by the adsorbent configuration. Activated Alumina was found to show maximum adsorption among the single layer adsorption systems, followed by Natural Zeolite and China Clay. The breakthrough time of the Activated Alumina column was about 340 min, and the exhaustion time was 920 min, suggesting that the adsorption affinity of tetracycline molecules on the Activated Alumina column is stronger. By comparison, China Clay had the lowest adsorption capacity and the first breakthrough occurred around 180 min and was followed by saturation after 520 min. The intermediate performance was seen with Natural Zeolite having a breakthrough time of 260 min and exhaustion time of 710 min. The use of multilayer adsorption had a great impact on the performance of the column. In the dual layer configuration (Activated Alumina/ Natural Zeolite) the breakthrough time was increased to about 520 min and the time to column exhaustion to 1250 min. This improvement shows the effectiveness of the use of different pore structure and adsorption properties adsorbents. The first adsorption layer was able to remove the most tetracycline, and the second layer was able to remove the tetracycline that was not removed by the first adsorption layer. The triple layer of Activated Alumina, Natural Zeolite and China Clay produced the best adsorption capacity. The simulation forecasted the breakthrough time of about 720 min and the saturation time of 1650 min. The predicted removal efficiencies of tetracyclines for China Clay, Natural Zeolite, Activated Alumina, dual layer and triple layer systems were about 97%. The table 3 shows the comparison of the proposed multilayer adsorption column with reported tetracycline adsorption studies.

**Table 3. adsorption studies**

| Adsorbent/System | Study Type | Pollutant | Removal Efficiency (%) | Key Findings |
|------------------|------------|-----------|------------------------|--------------|
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| <b>Multilayer Packed Bed Column (Activated Alumina + Natural Zeolite + China Clay)</b> | <b>Aspen ADSIM Simulation</b> | <b>Tetracycline</b> | <b>97</b> | <b>Longest breakthrough time (720 min), highest bed utilization, extended saturation time (1650 min), and superior adsorption.</b> |
|----------------------------------------------------------------------------------------|-------------------------------|---------------------|-----------|------------------------------------------------------------------------------------------------------------------------------------|

## CONCLUSION

In this study, a successful development and evaluation of a multilayered packed bed adsorption column for removal of tetracycline (TC) from aqueous environment were developed using Aspen ADSIM V10. China Clay, Natural Zeolite, Activated Alumina, dual layer (Activated Alumina–Natural Zeolite) and triple layer (Activated Alumina–Natural Zeolite–China Clay) configurations were tested using dynamic simulations. The breakthrough curves obtained were useful to identify the adsorption performance, the efficiency of the bed utilisation, the mass transfer characteristics and the column service life. The simulation results showed that the column's adsorption performance increased substantially with the increase in complexity of the column. The adsorption efficiency of single layer systems was in the sequence: Activated Alumina, China Clay. The adsorption efficiency of single layer systems was: Activated Alumina > China Clay. The use of several layers of adsorbents significantly boosted the efficiency of tetracycline removal due to the sequential adsorption and utilization of the adsorption sites. The proposed triple layer adsorption column had the longest breakthrough time of 720 min, the highest saturation time of 1650 min and the maximum amount of tetracycline removed was about 97%, which is better than all of the other adsorption column configurations investigated. The high performance of the multilayer adsorption system was attributed to the complementary adsorption property of the various materials used: Activated Alumina, Natural Zeolite and China Clay. The results indicate that the multilayer packed bed adsorption technology is a potential effective and sustainable method for cleaning the water contaminated with antibiotics. The suggested simulation technology is a cost-effective way to test the performance of the adsorption column before experimental testing and industrial use. Experimental testing of the developed model, optimization of the sequence of the adsorbents, experimental testing of competitive adsorption in multi contaminant systems, and further improving the adsorption efficiency and operational sustainability through the inclusion of artificial intelligence based optimization techniques in the future can be considered.

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