Fixed-bed column adsorption study: a comprehensive review

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Abstract

Present paper involved the review of fixed-bed column studies for removal of various contaminants from synthetic wastewater. Basic concept of adsorption, its types (i.e., chemisorption and physisorption) and its mechanism, adsorbents and adsorbates were included. Comparison of batch and column adsorption study is mentioned. Complete study of breakthrough curve for designing adsorptive column is interpreted. This paper explicates the detailed explanation of various process parameters and isotherm models for column study. Fixed-bed adsorption studies using various adsorbates, i.e., metal, ion, dye and other hazardous materials, are reviewed, in which adsorption of chromium metal is most exploitable. Conclusion and some challenges for utilization in real world are also exposed.

Keywords Adsorption · Fixed-bed column · Adsorbent · Adsorbate · Process parameters · Isotherm models

Introduction

The "adsorption" was suggested by Bois-Reymond but given into the world by Kayser, which is defined as an increasing concentration of a specific compound at the surface of interface of the two phases. These specific compounds are transporting from one phase to another and thereafter adhered into surface. It is considered to be a complex phenomenon and depends mostly on the surface chemistry or nature of the sorbent, sorbate and the system conditions in between the two phases. It is the most inexpensive and efficient process for treatment of water or wastewater; therefore, it has been widely used for the removal of solutes from solutions and harmful chemicals from environment. It required less investment in terms of the initial cost and land, simple design, no other toxic effect and superior removal of organic waste constituent, compared to the other conventional treatment in water pollution control (Dabrowski 2001; Selim et al. 2014).

In adsorption process, there is higher concentration of materials at the surface or interface between the two phases, it is called interphase accumulation. The substance which is being adsorbed on the surface of another substance is called adsorbate. The substance, present in bulk, on the surface of which adsorption is taking place is called adsorbent. The interface may be liquid–liquid, liquid–solid, gas–liquid or gas–solid. Of these types of adsorption, only liquid–solid adsorption is widely used in water and wastewater treatment. Following four steps are considered, in which solute (adsorbate) is moved toward the interface layer and attached into adsorbent. (1) Advective transport: solute particles are moved from bulk solutions onto immobile film layer by means of advective flow or axial dispersion or diffusion, (2) film transfer: solute particle is penetrated and attached in immobile water film layer, (3) mass transfer: attachment of solute particle onto the surface of the adsorbent and finally (4) intraparticle diffusion: Movement of solute into the pores of adsorbent (Vasanth et al. 2004).

Mainly two types of adsorption are occurred. Physical sorption is occurred due to weak Van der Waals attraction forces. This sorption is reversible in nature with low enthalpy values, about 20 kJ/mol. Here, wear attractive forces are available between adsorbed molecules and the solid surface weak in nature. Therefore, adsorbed molecules are liberated to travel over the surface, as these molecules are not stuck to a particle side on the adsorbent surface. The electrostatic forces include dipole–dipole interactions, dispersion interactions and hydrogen bonding available among the adsorbate–adsorbent in physical sorption. When there is a net separation of positive and negative charges within a molecule, it is said to have a dipole moment. Whereas,

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chemical bonding between sorbate and sorbent molecule takes place in chemisorption. Therefore, this sorption is irreversible in nature and has high enthalpy of sorption than physical sorption 200 kJ/mol. Stronger electrostatic forces such as covalent or electrostatic chemical bond play a vital role in attraction between sorbent and sorbate. This bond is shorter in bond length and has higher bond energy. The ranges of energy for each reaction are: (1) Van der Waals force ($4 < DH < 10$ kJ/mol), (2) hydrophobic force (< 5 kJ/mol), (3) dipole force ($2 < DH < 29$ kJ/mol), (4) hydrogen bond ($2 < DH < 40$ kJ/mol), (5) coordination exchange ($40 < DH < 60$ kJ/mol) and (6) chemical bond ($DH > 60$ kJ/mol) (Montgomery 1985; Sawyer et al. 1994; Atkins 1994; Ghaly et al. 2016).

The adsorbent is broadly divided into three classes: (1) Synthetic adsorbent: Various porous materials are synthesized in laboratory using different processes, which have high adsorption capacities. Disadvantage is that this process of manufacturing is comparatively costly. (2) Natural adsorbent: Natural materials like plant root, leaf and agricultural waste are dried, crushed, sieved, again washed with distilled water and used as adsorbent for treatment of real as well as synthetic wastewater. This process is cheap, but adsorption capacity is comparatively low. (3) Semi-synthetic adsorbent: Natural materials undergo chemical as well as physical activation to develop highly porous surface. The major advantages of this adsorbent include: low cost, high efficiency, minimization of chemical or biological sludge, no additional nutrient requirement and regeneration of absorbent and possibility of metal recovery. Industrial adsorbent is also classified into three types according to their constitution: (1) oxygen-containing adsorbent, (2) carbon-based adsorbent and (3) polymer-based adsorbent (Sameera et al. 2011; Kratochvil and Volesky 1998; Kumar et al. 2005). The properties of the adsorbent are identified by different analytical techniques such as Fourier transform infrared spectroscopy (FT-IR), scanning electron microcopy (SEM), X-ray diffraction (XRD), porosity, pore diameter, pore volume and surface area analysis. FT-IR technique determines the chemical composition by investigating the function group. SEM investigates the morphology of adsorbent. XRD provides information on the crystallographic structure of the material (Sathasivam and Haris 2010; Esparza et al. 2011; Ahmad and Kumar 2011, Abdel Rahman et al. 2018).

An adsorbate is any substance that has undergone adsorption on the surface. In environmental chemistry, adsorbate is considered as pollutant or compounds contributing to the pollution, which adhered in porous adsorbent and easily removed. The various types of water pollutants can be classified into following major categories: (1) organic pollutant, which includes oxygen demanding waste, oil, sewage and agricultural waste, synthetic organic waste, disease causing wastes, (2) inorganic pollutant, which contains inorganic salts, mineral acids, finely divided metal compounds, trace metals, etc. (3) sediments, which are soil and minerals particles that are washed away from land by flood waters (Yi et al. 2008), (4) thermal pollution, in which higher temperature is considered as pollutant and (5) radioactive pollutants are pollutants that have a radiological hazard, its sources might be natural, accidental release of radio contaminant, historical releases due to military tests and/or historical discharge (Abdel Rahman et al. 2014). Each pollutant has different adverse effect. These pollutants are hazardous into mankind, aquatic life and other ecological constitutions (Sharma and Sanghi 2012).

Batch, continuous moving bed, continuous fixed bed (upflow or downflow), continuous fluidize bed and pulsed bed are various types of technique by which the contact between adsorbate and adsorbent is mainly occurred in the adsorption system. Each method has merits and demerits, which are mentioned in Table 1. This table reveals that fixed-bed column is more preferable and industrially feasible for removal of various contaminations from synthetic as well as real wastewater. The performance of fixed-bed column is studied by breakthrough curves, i.e., a representation of the pollutant-effluent concentration versus time profile in a fixed-bed column. The mechanism of this adsorption is based on different phenomena, like axial dispersion, film diffusion resistance, intraparticle diffusion resistance (both pore and surface diffusion) and sorption equilibrium with the sorbent (Kafshgari et al. 2013; Miralles et al. 2010).

The relation between the nature of breakthrough curves and fixed-bed adsorption was as adequately expressed using mass transfer zone (MTZ) or primary sorption zone (PSZ). As per Fig. 1, feed water (wastewater) is inserted through the inlet of the column, the adsorbate is adsorbed most rapidly and effectively by the upper few layers of the fresh adsorbent during the initial stage of the operations. This is due to higher amount of adsorbent and small levels of adsorbate available at these upper layers, so that adsorbate is readily escaped in the lower strata of the bed and no adsorbate (pollutants) run off from the adsorbent at the first stage. So, primary adsorption zone or MTZ is attained near the top or influent end of the column. At this point, concentration of adsorbate (C) is zero, and thus, ratio of effluent and initial concentration (C/C_0) is zero. Thereafter, upper layer of adsorbent is gradually saturated, with feeding the polluted water (adsorbate) into the column, which becomes adsorbent less efficient progressively. Thus, the primary sorption zone also travels descending to fresher or un-adsorbed part of adsorbent in the column. Further, with movement of this zone, tendency is that more and more adsorbate comes out in the effluent as per points C_1/C_0, C_2/C_0, C_3/C_0 and C_4/C_0. The movement of this zone is mainly increasing with increasing initial concentration compared to linear velocity of the feed water. After some time (C_s), the column is completely

Table 1 Features and limitation of various sorption processes (Monash and Pugazhenthi 2010; Cavalcante Jr 2000; US EPA 1983)

| Particular	Batch sorption	Continuous fixed-bed sorption	Continuous moving bed sorption	Continuous fluidized bed sorption	Pulsed bed sorption
Introduction	Adsorbent and adsorbate are well mixed in diluted solution at constant volume in well-mixed system	Fixed-bed system consists of a adsorbent in which adsorbate is continuously flowed through a bed of adsorbent at constant rate	Continuous moving bed sorption is steady-state system, where both adsorbent and adsorbate are in motion, and bed of adsorbent section remains at constant, but not in equal condition	In this sorption, adsorbate is in contact with fluidized bed of adsorbent with sufficient or insufficient flow	In pulsed bed sorption, adsorbate is contacted with same adsorbent in bed, until desired results are not achieved
Features	Very easy and cheap technique Most of the researchers are using this technique to analyze feasibility of adsorbent—adsorbate system	Very easy and cheap technique Used for higher quantity of wastewater having higher pollution load Also, widely used for industrial purpose, because the adsorbate is continuously in contact with a given quantity of fresh adsorbent in fixed-bed column system	Complicated and very expensive technique As adsorbent is continuously replaced and fresh adsorbent is constant contact with adsorbate	Complicated and very expensive technique Used for higher quantity of wastewater having higher pollution load Also, applicable for industries because it allows rapid mixing of adsorbent—adsorbate and also, adsorbate is continuously flow automatically with controlled operation and easy handing	Very easy and cheap technique It is very easily controlled automatically operated system Also, it required lower dosage of adsorbent. This type has an advantage of better utilization of adsorbent because the adsorbents were kept for regeneration as soon as the adsorbent gets saturated
Disadvantages	Used for small quantity of wastewater having minimum pollution load; therefore, this operation is that it is scarcely found in the majority of practical (industrial) applications Adsorbent is removed from the system by simple filtration method	The problems associated with this sorption are adsorbent attrition, feed channeling, and non-uniform flow of adsorbent particles Forceful interaction is conducted in continuous fixed-bed systems to reduce space and time. As a result, it is difficult to carry out a priori design and optimization of fixed-bed columns without a quantitative approach	The large amount of adsorbent is required to complete sorption Continuous regeneration of adsorbent and adsorbent storage is essential	Flow of adsorbate is not measured with large deviation from plug flow and bubbling or feed channeling, which leads to insufficient contact of adsorbent—adsorbate The rapid mixing of adsorbent—adsorbate system leads to non-uniform residence time	Used for small quantity of wastewater having minimum pollution load especially lower suspected solid Adsorbent is not unfilled in normal operations
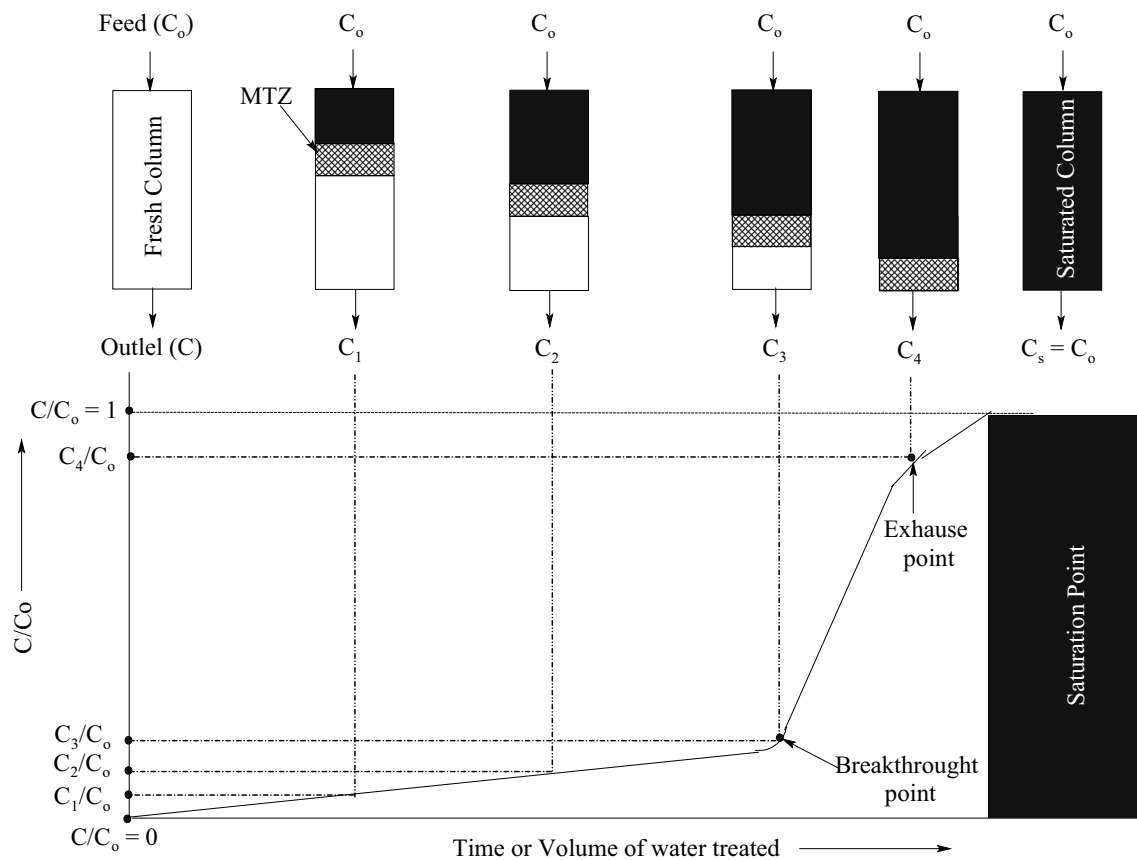
Fig. 1 Representation of breakthrough curve by movement of MTZ

saturated or exhausted and thereafter, adsorption does not occur. At this point, the ratio of C/C_0 is 1 (one). In most of the cases of the sorption by column method operation of water and wastewater, breakthrough curves exhibit a characteristic 'S' shape but with varying degree of steepness (Chowdhury et al. 2013; Shafeeyan et al. 2014; Hasanzadeh et al. 2016).

As Fig. 2 shows, initially sorbent is regarded to be exhausted easily, breakthrough point is selected arbitrarily at lower value of break point concentration (C_b) for the effluent concentration and exhaustion point concentration (C_x) closely imminent influent concentration of adsorbate. Here V_b and V_x are the volume of effluent corresponding to break point concentration (C_b) and exhaustion point concentration (C_x), respectively. The primary sorption zone (PSZ) is the portion between exhaustion point (C_x) and breakthrough point concentration of adsorbate (C_b). If PSZ is assumed to have a constant length or depth (δ), some important parameters such as total time taken for the primary sorption zone to establish itself (t_x), time required for the exchange zone to move the length of its own height up/down the column (t_δ), rate at which the exchange zone is moving up or down through the bed (U_z), fraction of adsorbate present in the
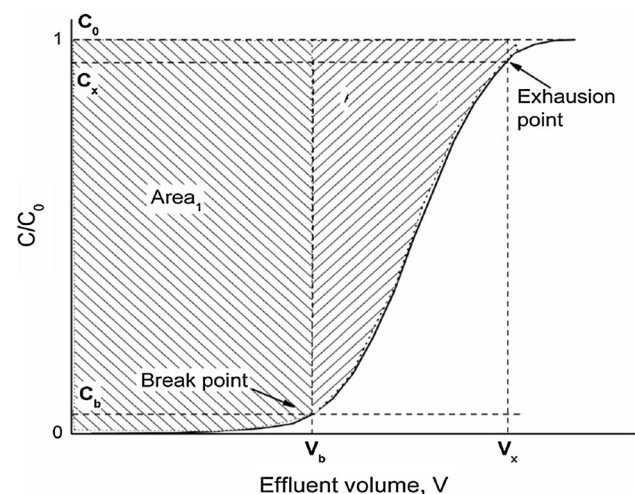

Fig. 2 Ideal breakthrough curve

adsorption zone (F) and percentage of the total column saturated at breakthrough are calculated using simple equations. These parameters play vital role for column designing (Gupta and Ali 2012; Crittenden and Thomas 1998).

Process parameter for column study

Most of the adsorption studies were conducted on synthetic wastewater as adsorbate, in which metal or dye solution is prepared and treated with adsorbent. Effect of various process parameters like the initial adsorbate concentration, flow rate of adsorbate in column, bed height of column, pH of adsorbate, particle size of adsorbent and temperature of system were performed and breakthrough and exhaust points were measured. All these parameters are importance for evaluating the efficiency of adsorbent in a continuous treatment process of effluents on the pilot or industrial scale (Yang et al. 2015). Table 2 shows the effect of process parameters on breakthrough and exhaust point with its features and explanation. Out of these parameters, initial adsorbate concentration, bed height and flow rate are most feasible parameters, as most of researchers are recently working on these parameters and utilized to remove various types of pollutants like dyes, metal, hazardous waste, etc. using natural and synthetic adsorbents.

Adsorption models for column study

Various practical features such as sorbent capacity, operating life span, regeneration time and prediction of the time necessary play a vital role during the operation of column using adsorption dynamics acquaintance and modeling. Also, these models provide detailed conclusions about the mechanism of the process. The adsorption column is subjected to axial dispersion, external film resistance and intraparticle diffusion resistance. So, the mathematical correlations for adsorption in fixed-bed columns are based on the assumption of axial dispersion, external mass transfer, intraparticle diffusion and nonlinear isotherms. A number of mathematical models have been developed for the evaluation of efficiency and applicability of the column models for large-scale operations. The Thomas, bed depth service time, the Adams and Bohart model, Yoon–Nelson, Clark, Wolborska and modified dose–response model are most commonly used to analyze the column behavior of adsorbent–adsorbate system. Most general and widely used for column studies is Thomas model (TM). Maximum solid-phase concentration of adsorbate on adsorbent and rate constant is determined using data obtained from column continuous studies by Thomas adsorption model. The Thomas model is proposed on assumption of Langmuir kinetics of adsorption–desorption that rate driving forces follow second-order reversible reaction kinetics and also no axial dispersion. The bed depth service time (BDST) model is based on the Bohart and Adams quasi-chemical

rate law. Rational of this model is that equilibrium is not immediate in bed, and therefore, the rate of the sorption process is directly proportional to the fraction of sorption capacity still remaining on the media. This model is provided by the relationship between bed depth and service time in terms of process concentrations and adsorption parameters. This model is based on the hypothesis that the adsorption rate is maintained by the surface reaction between adsorbate and the unused capacity of the adsorbent. The values of breakthrough time obtained for various bed heights used in this study were introduced into the BDST model. Therefore, sorbent quantity is being preferably used, instead of the bed height (Wan Ngah et al. 2012).

Scientists, namely Bohart and Adams, investigated the equation for relationship between C_t/C_o and time in a continuous system, which is known as Adam–Bohart model (ABM). Basically, innovative studies were carried out by Adam and Bohart using gas–charcoal adsorption system, and thereafter, its equation can be useful for other continuous adsorption system. This model proposed that rate of adsorption depends upon concentration of the sorbing species and residual capacity of adsorption (Dorado et al. 2014). Yoon–Nelson model (YNM) is simple theoretical assumption, which does not concentrated upon properties of adsorbate, type of adsorbent and any physical features of the adsorption bed. This model is given probable statement that decreasing rate of adsorption is directly proportional to adsorbate adsorption and breakthrough on the adsorbent. Scientist, namely Clark, proposed model for breakthrough curves, which based suggestions that (1) column adsorption is mass-transfer concept with combination of Freundlich isotherm and (2) behavior of flow in column is of piston type. By using the laws of mass transfer and by neglecting the phenomenon of dispersion, Clark solved the system of equations of mass transfer. This model is called Clark model (CM). Wolborska mentioned the relationship that describes the concentration distribution in a bed for the low-concentration range of the breakthrough curve, which is referred as Wolborska model (WM). Another simplified numerical model used to describe fixed-bed column adsorption data is the modified dose response model (MDRM). This model basically diminishes the error resulting from the use of the Thomas model, particularly at lower or higher time periods of the breakthrough curve (Lee et al. 2015; Biswas and Mishra 2015).

Adsorption capacity of each model for different process parameters such as initial concentration of adsorbate, bed height, flow rate, etc. is calculated and mentioned by various scientists for designing the column. Table 3 depicts the equation, plot and parameters of each model. It also mentioned the variation, i.e., increasing or decreasing in column adsorption model parameters with respect to increasing operation parameters. Very important

Table 2 Effect of process parameter on breakthrough and exhaustion point

Process parameter	Features	Explanation
Initial adsorbate concentration (IAC)	Breakthrough and exhaustion points are occurred earlier with increasing influent concentration. And thereafter breakpoint time decreased with increasing the inlet concentration	Initially, adsorption was rapid because of the availability of large number of vacant sites. And thereafter, increasing initial adsorbate concentration results in a greater driving force to overcome mass-transfer resistance in the liquid phase and the sites are exhausted quickly, so the volume of effluent treated also decreases (Moyo et al. 2017; Saravanan et al. 2018)
Flow rate of adsorbate (FRA)	Breakthrough points generally occur faster with higher flow rate. Saturation of breakthrough time is increased significantly with a decrease in the flow rate	The rate of mass transfer gets increased, i.e., the amount of adsorbate adsorbed onto unit bed height (mass-transfer zone) gets increased with increasing flow rate leading to faster saturation (Lopez-Cervantes et al. 2017). And lower flow rate, adsorbate has more time to contact with adsorbent that resulted in higher removal of adsorbate in column (Ahmad and Hameed 2009; Sheng et al. 2018)
Bed height of column (BHC)	Breakthrough and exhaustion times are slower with increasing bed depth. Also, it was found that the volume of effluent treated increased with increasing the bed depth	This was attributed to an increase in the surface area and the number of binding sites available for adsorption. The time for interaction of adsorbate and adsorbent also increased with increasing amount of adsorbent (Fathi et al. 2014; Teutscherova et al. 2018)
pH of adsorbate (pH)	In some case, highest removals are found at acidic pH and maximum removals of some adsorbate are found at alkaline pH	It depended upon the nature of adsorbent and adsorbate (Banerjee and Chattopadhyaya 2013; Ahmed and Hameed 2018)
Particle size of adsorbent (PSA)	Breakthrough and exhaustion times are slower with increasing particle size of adsorbent. Maximum particle size is favored to get better adsorption capacity. But, moderate flow rate is preferred for industrial applications	Adsorption is a surface phenomenon and the extent of adsorption is expected to be proportional to the specific surface. However, very small particle size is not studied to avoid problem associated with solid–liquid separation. Further, smaller particles develop high-pressure drop in the fixed-bed column adsorbent (Unger et al. 2008; Zou et al. 2013)
Temperature (T)	Breakthrough and exhaustion times are slower with increasing temperature of system. But, adsorption capacity decreases with the increasing temperature	It might be due to that high operating temperature favored adsorbate diffusing faster into the adsorbent, giving a low breakthrough and exhaust time. Further, less adsorbate was required to satisfy the maximum adsorption capacity of adsorbent at high adsorption temperatures, indicating an exothermic process. For industrial applications, room temperature adsorption is preferred to reduce heating operation setup cost (Girish and Murty 2014; Ye et al. 2018)

Table 3 Details of column adsorption models and its parameters variation in column adsorption model w.r.t. increasing operation parameters (Lee et al. 2015; Biswas and Mishra 2015)

Column adsorption model	Linear equation	Plot	Parameter of model	Initial conc.	Flow rate	Bed height	Temperature
Thomas model (TM)	$\ln\left[\left(\frac{C_O}{C_t}\right) - 1\right] = \frac{k_{TH}q_o m}{Q} - \frac{k_{TH}C_O V_{eff}}{Q}$	$\ln\left[\left(C_o/C_t\right) - 1\right]$ versus time	k_{TH}	Decreased	Increased	Decreased	Decreased
			q_0	Increased	Decreased	Increased	Increased
Bed depth service time model (BDST)	$t = \frac{N_O}{C_0 F}Z - \frac{1}{k_{BDST}C_0}\ln\left(\frac{C_0}{C_t} - 1\right)$	Bed height versus time	N_o	Increased	Increased	N.A.	Increased
			k_{BDST}	Increased	Increased	N.A.	Increased
Adam and Bohart model (ABM)	$\ln\left[\left(\frac{C_O}{C_t}\right) - 1\right] = K_{AB}N_O\frac{Z}{u} - K_{AB}C_t t$	$\ln\left[\left(C_o/C_t\right) - 1\right]$ versus time	k_{AB}	Increased	Increased	Decreased	Decreased
			N_o	Decreased	Decreased	Increased	Increased
Yoon–Nelson model (YNM)	$\ln\left(\frac{C_t}{C_o - C_t}\right) = k_{YN}t - \tau k_{YN}$ $q_{OYN} = \frac{q_{(total)}}{X} = \frac{C_0 Q\tau}{1000X} = \frac{\frac{1}{2}C_O[(Q/1000)X2\tau]}{X}$	$\ln\left(\frac{C_t}{C_0 - C_t}\right)$ versus time	k_{YN}	Increased	Increased	Decreased	N.A.
			T	Decreased	Decreased	Increased	N.A.
Clark model (CM)	$\frac{C_t}{C_O} = \frac{1}{(1+Ae^{-rt})^{1/(n-1)}}$	C_t/C_0 versus time	A	Decreased	Decreased	Increased	Increased
			R	Increased	Increased	Decreased	Decreased
Wolborska model (WM)	$\ln\left(\frac{C_t}{C_O}\right) = \frac{\beta C_O t}{N_O} - \frac{\beta Z}{U}$	$\ln(C_t/C_0)$ versus time	B	Decreased	Increased	Decreased	Decreased
			N_o	Increased	Decreased	Increased	Increased
Modified dose response model (MDRM)	$\ln\left(\frac{C_t}{C_0 - C_t}\right) = a\ln\left(C_0 Qt\right) - a\ln\left(q_{mdr}m\right)$	$\ln\left(\frac{C_t}{C_0 - C_t}\right)$ versus $\ln\left(C_0 Q_t\right)$	A	Decreased	Increased	Increased	N.A.
			B	Increased	Increased	Increased	N.A.

N.A. Data not available

parameters of Thomas mode, i.e., q_{TH} increased with the increase in initial concentration of adsorbent, bed height and temperature and corresponding K_{TH} values decreased. Also, q_{TH} decreased with increase in flow rate and corresponding K_{TH} values increased. Further, correlation coefficient value (r^2) from straight line graph of all models is calculated and mentioned in most of the research papers. The coefficient of determination is useful because it gives the proportion of the variance (fluctuation) of one variable that is predictable from the other variables. It is a measure that allows us to determine how certain one can be in making predictions from a certain model. This value plays important role for any adsorption isotherm. If coefficient value is closer to unity (1), then it indicates the most suitable isotherm model (Zhanga et al. 2011).

Adsorptive column

Variety of adsorbents and adsorbates are studied using different process parameters and isotherm models in recent years. In this review paper, column adsorption study is isolating using different adsorbates as follows.

Adsorption of metal and ion

Earth's crust is constituent of metal and other parts, but random human activities have significantly changed their geochemical cycles and biochemical balance. This results in accumulation of metals in plant parts having secondary metabolites, which is responsible for a particular pharmacological activity. Prolonged exposure to heavy metals such as cadmium, copper, lead, nickel and zinc can cause deleterious health effects in humans (Singh et al. 2011). Various scientists have being tried to remove metals and its ions using adsorptive column treatment. Synthetic adsorbents, namely polyacrylonitrile–potassium cobalt hexacyanoferrates and polyacrylonitrile–potassium nickel hexacyanoferrates, were synthesized, and adsorption of cesium was investigated. The effects of liquid flow rate, bed height and presence of other cations on the adsorption of cesium were performed. The bed depth service time (BDST) model and the Thomas model were used to analyze the experimental data, and the model parameters were evaluated (Du et al. 2014). Ali had synthesized economical adsorbent, i.e., carbon nanotube using Ni/MgO metal oxide for microwave exposure for thermal disintegration at 550 °C. This microwave-assisted nanotube had undergone column studies for removal of arsenite and

arsenate using process variables like initial concentration, flow rate and bed height. The data were analyzed using Tomas and Adam bohart models, and maximum removals were found to be 13.5 and 14.0 mg/g for arsenite and arsenate, respectively (Ali 2018). Novel 3D yttrium-based graphene oxide–sodium alginate hydrogel was prepared by sol–gel process for removal of fluoride via continuous filtration. Data were analyzed by Thomas model, and maximum uptake capacity was achieved to be 4.00 mg/g (He et al. 2018). Vertical column experiments using sugarcane bagasse were conducted for removal of manganese(II), and the highest removal efficiency was found to be 51.95% (Zaini et al. 2018). New chelating cellulose-based adsorbent, i.e., N-methyl-D-glucamine (NMDG)-type functional group attached to a novel boron selective chelating fiber, was prepared, characterized and utilized for boron removal. Yoon–Nelson, Thomas and modified dose response model were evaluated using data of various flow rates. Maximum boron adsorption capacity related to Thomas model was obtained up to 22.06 mg/g (Recepoglu et al. 2018). Freitas and their co-scientists were experimented for binary adsorption of silver and copper onto bentonite (Verde-lodo clay), in which first flow rate was optimized. Thereafter, effects of initial concentration and molar fraction were investigated using this optimum flow rate (Freitas et al. 2018).

Comparison studies of unmodified and modified jordanian kaolinite clay using humic acid were accomplished for removal of heavy metals such as lead(II), cadmium(II) and zinc(II). Various process variables for batch (contact time, adsorbent dose, initial metal ion concentration, pH and temperature) as well as column (initial concentration, flow rate and bed height) were evaluated. Results indicated that modified clay was more dominant than unmodified clay; and adsorption of the metal ions by both modified kaolinite clay followed the order: Pb > Cd > Zn (Al-Essa and Khalili 2018). Color from real textile effluent was removed in fixed-bed column of modified zeolite (SMZ), in which surface of natural zeolite was modified with a quaternary amine surfactant hexadecyltrimethylammonium bromide (HTAB). Breakthrough curves of different flow rate (0.015–0.075 l/min) and bed height (12.5–50 cm) at original as well as diluted wastewater (ratio of 25, 50 and 75%) were plotted, and breakthrough and exhaust points were calculated for each and every parameter. Also, experiments for regeneration of SMZ using NaCl and NaOH solution were carried out. Data were analyzed by BDST isotherm (Ozdemir et al. 2009). Different experiments of packed bed column were demonstrated for adsorption of hexavalent chromium from its synthetic solution (Rangabhashiyam and Selvaraju 2015a) and electroplating industries effluent (Rangabhashiyam et al. 2016) using chemically modified swietenia mahagoni shell. Also, caryota urens inflorescence waste biomass was utilized as

adsorbent for adsorption of hexavalent chromium (Rangabhashiyam and Selvaraju 2015b).

Comparison of batch and column treatment for removal of nickel(II) and copper(II) using chemically modified *Cucurbita moschata* was exploited, which indicated column treatment is more feasible than batch process (Khan and Rao 2017). Series of synthetic solutions of cadmium(II), copper(II), lead(II) and zinc(II) were prepared and tried to remove using chemically modified multi-metal-binding biosorbent (MMBB) in packed bed column. The breakthrough curves for influent flow rate, initial metal concentration and bed depth were prepared, and data were explored using Thomas, Yoon–Nelson and modified dose response. Breakthrough and exhaust points, MTZ, t_b, t_{sat}, C_p and t_p were calculated. The highest metal adsorption capacities of modified MMBB at the exhaustion times were 38.25, 63.37, 108.12 and 35.23 mg/g for Cd, Cu, Pb and Zn, respectively. Desorption study by HCl and applicability of biosorbent tested using semi-simulated wastewater were also conducted (Abdolali et al. 2017). Column adsorption studies on nickel and cobalt removal from aqueous solution using native and biochar form of *tectona grandis* were performed. Breakthrough curves were plotted for process variable bed height, flow rate and inlet metal ion concentration. Data were applied to Adam–Bohart, Thomas and Yoon–Nelson, in which Thomas model was found to be in good agreement with higher R^2 and closer experimental and theoretical uptake capacity values (Vilvanathan and Shanthakumar 2017). Remaining reviews are mentioned in Table 4, which represents the recently published papers of metal and ion adsorbate, its adsorbent, operation parameters, investigated column adsorption models and respective maximum adsorptive capacity related to Thomas model and corresponding reference.

Adsorption of dye

Dyes usually have a synthetic origin and complex aromatic molecular structures which make them more stable and more difficult to biodegrade. Degradation of dyes is typically a slow process. The removal of color is needed to be considered in the disposal of textile wastewater due to aesthetic deterioration as well as the obstruction of penetration of dissolved oxygen and sunlight into water bodies, which seriously affects aquatic life. Besides, the dye precursors and degradation products are proven carcinogenic and mutagenic in nature. Consumption of dye-polluted water can cause allergy reactions, dermatitis, skin irritation, cancer and mutation both in babies and matures (Patel and Vashi 2013). Lopez-Cervantes and team members had prepared biosorbent chitosan–glutaraldehyde from shrimp shells for the removal of the textile dye Direct Blue 71 from an aqueous solution. This bioadsorbent was analyzed using

Table 4 Details of column adsorption studies of metal and ions

Adsorbate	Adsorbent	Operation parameters	Column isotherm investigated	Thomas maximum adsorption capacity	References
Fluoride	Kanuma mud	Initial concentration, flow rate and bed height	TM and BDST	0.585 mg/g	Chen et al. (2011)
Cadmium(II)	Syzygium cumini L leaf powder	pH, initial concentration, flow rate and bed height	TM, BDST, ABM and YNM	29.08 mg/g	Rao et al. (2011)
Copper(II), lead(II) and cadmium(II)	Functionalized SBA-1mesoporous silica with polyamidoamine	Flow rate and bed height	TM and BDST	1.6, 1.3 and 1.0 mmol/g	Shahbazi et al. (2013)
Hexavalent chromium (Cr^{+6})	Modified corn stalk	pH, influent concentration, flow rate and bed height	TM, ABM and YNM	152,323.70 mg/g	Chen et al. (2012)
Copper(II)	Chitosan–zeolite composite	Bed height	BDST, CM	41.14 g/L	Wan Ngah et al. (2012)
Uranium(VI)	Grapefruit peel (GFP)	Initial concentration, flow rate, bed height and particle size of GFP	TM, BDST, YNM and CM	104.1 mg/g	Zou et al. (2013)
Copper(II)	kenaf (Hibiscus cannabinus, L) fibers	Flow rate and bed height	TM and BDST	47.27 mg/g	Hasfalina et al. (2012)
Chromium(VI)	Orthophosphoric acid-activated lignin	pH, initial concentration, flow rate, bed height and ionic strength	TM, BDST, ABM and MDRM	0.889 mmol/g	Albadarin et al. (2012)
Cesium(I) and strontium(II)	Montmorillonite–iron oxide composite	Initial concentration and flow rate	TM	4.42 and 15.28 mg/g	Ararem et al. (2013)
Chromium(VI)	Leonardite	Initial concentration and flow rate	TM, BDST, YNM, WM, CM and MDRM	127.53 mg/l (BDST)	Dorado et al. (2014)
Cadmium(II) and lead(II)	Dead calcareous skeletons	Initial concentration, flow rate and bed height	TM, BDST and YNM	66.16 and 75.18 mg/g	Lim and Aris (2014)
Copper(II)	Polyaniline-coated sawdust	Initial concentration, flow rate and bed height	TM, BDST and YNM	58.23 mg/g	Liu and Sun (2012)
Bromate	Fe(II)–Al(III)-layered double hydroxide	Initial concentration, flow rate and bed height	TM and BDST	71.01 μmol/g	Yang et al. (2015)
Copper(II)	Surface-modified eucalyptus globulus seeds	Initial concentration, flow rate and bed height	TM, BDST and YNM	300.5 mg/g	Senthil Kumar et al. (2015)
Flouride	Activated alumina	Initial concentration, flow rate and bed height	TM, YNM and ABM	11.01 mg/g	Ghorai and Pant (2004)
Phosphate	Zirconium-loaded soyabean residue (okara)	pH, initial concentration, flow rate, bed height and particle size	TM, BDST and ABM	12.21 mg/g	Nguyen et al. (2015)
Chromium(VI)	Alkaline anion exchange fiber	Initial concentration, flow rate, bed height, pH and temperature	TM, ABM, YNM and CM	210.2 mg/g	Wang, Li and Zeng (2015)
Copper(II) and nickel(II)	Magnetized sawdust (Fe$_3$O$_4$–SD)	Initial concentration, flow rate and bed height	TM, ABM and YNM	43.45 and 33.08 mg/g	Kapur and Mondal (2016)

Table 4 (continued)

Adsorbate	Adsorbent	Operation parameters	Column isotherm investigated	Thomas maximum adsorption capacity	References
Nickel(II) and chromium(II)	TiO$_2$ agglomerated nanoparticles	Initial concentration, flow rate and bed height	TM, BDST and ABM	33.18 and 12.94 mg/g	Debnath et al. (2010)
Cadmium(II) and lead(II)	Grape stalk wastes (GSW)	Initial concentration and particle size of GSW	TM	31.53 and 49.40 mg/g	Miralles et al. (2010)
Manganese(II)	Granular-activated carbon from agrowaste of mangostene fruit peel	Initial concentration, flow rate and bed height	TM, ABM and YNM	7257.32 mg/g	Chowdhury et al. (2013)
Chromium(II)	Pistachio shell	Initial concentration, flow rate, bed height, pH, effluent concentration and temperature	TM, ABM and YNM	27.95 mg/g	Banerjee et al. (2018)
Copper(II)	Amino-functionalized ramie stalk	Initial concentration flow rate and bed height	TM, ABM, YNM and BDST	0.528 mmol/g	Wang et al. (2018)
Lead(II) and cadmium(II)	PAAC nanocomposite	Initial concentration, flow rate, bed height, pH and temperature	TM	36.20 and 37.25 mg/g	Zendehdel and Mohammadi (2018)
Fluoride	Magnesia–pullulan composite (MgOP)	Initial concentration, flow rate, bed height, pH, temperature and other existing anions	TM and YNM	16.6 mg/g	Ye et al. (2018)
Arsenate	Chitosan	Initial concentration, flow rate, bed height, bed diameter and flow direction	TM, ABM and YNM	51.2 mg/g	Brion-Roby et al. (2018)
Copper(II), cobalt(II) and nickel(II)	Sugarcane bagasse	Initial concentration, flow rate and bed height	TM and ABM	1.060, 0.800 and 1.029 mmol/g	Xavier et al. (2018)
Cyanide	Blast furnace granulated slag	Initial concentration, flow rate, pH and bed height	–	91.6% (Removal efficiency)	Rout et al. 2018
Fluoride	Magnesium–hydroxyapatite pellets	Initial concentration, flow rate, pH, bed height, particle size and particle shape	TM and ABM	45.5 mg/g	Mondal et al. (2018)
Copper(II), magnesium(II) and nickel(II)	Yersiniabactin, immobilized to XAD16 resin	Flow rate and pH	TM and MDRM	0.12, 0.2 and 0.1 mg/g	Moscatello et al. (2018)
Chromium(VI)	Ionic liquid functionalized cellulose (ILFC)	pH	TM and YNM	181.8 mg/g	Zhen and Long (2018)
Chromium(VI)	Co-immobilized activated carbon and Bacillus subtilis	Initial concentration, flow rate and bed height	TM	11.7 mg/g	Sukumar et al. (2017)
Chromium(VI), copper(II) and zinc(II)	Activated Neem bark	Initial concentration, flow rate and bed height	TM and YNM	53.95, 12.45 and 23.54 mg/g	Maheshwari and Gupta (2016)

Table 4 (continued)

Adsorbate	Adsorbent	Operation parameters	Column isotherm investigated	Thomas maximum adsorption capacity	References
Chromium(VI)	Polypyrrole/Fe_3O_4 nanocomposite	Initial concentration, flow rate, composition of nanocomposite and bed height	TM, ABM and YNM	258.36 mg/g	Bhaumik et al. (2013)
Copper(II)	Tetraethylenepentamine-modified sugarcane bagasse	Initial concentration, flow rate and bed height	TM and YNM	0.26 mmol/g	Chen et al. (2017)
Copper(II) and nickel(II)	Magnetized sawdust (Fe_3O_4–SD)	Initial concentration, flow rate and bed height	TM, ABM, YNM and BDST	31.89 and 23.59 mg/g	Kapur and Mondal (2016)
Copper(II) and nickel(II)	Natural and immobilized marine algae *Sargassum* sp.	Initial concentration, flow rate and bed height	TM	2.06 and 1.69 mmol/g	Barquilha et al. (2017)

scanning electron microscopy, X-ray diffraction and nuclear magnetic resonance spectroscopy. The effect of various process parameters such as bed height, inlet Direct Blue 71 concentration, flow rate was performed. Column isotherms Adams–Bohart, Thomas and bed depth service time mathematical models were utilized, in which bed depth service time model showed good agreement with the experimental data and the high values of correlation coefficients. Maximum dye removal capacity was found to be 343.59 mg/g (Lopez-Cervantes et al. 2017). Iranian Luffa cylindrica and NaOH-modified Luffa cylindrica as a natural lignocellulosic adsorbent were prepared and investigated for biosorption of methylene blue (MB) using a fixed-bed column. The response surface methodology based on central composite design was used to evaluate the interactive effects of three major operating parameters like inlet dye concentration, Luffa dosage and feed flow rate on the dye removal percentage (response variable). The breakthrough curves were predicted by the Adams–Bohart and Thomas models using nonlinear regression analysis, in which maximum adsorption capacities of methylene blue dye were achieved to be 21.4 and 46.58 mg/g for Luffa and NaOH-modified Luffa, respectively. Higher capacity of NaOH-modified Luffa is attributed to the intensification of the negatively charged surface of the base-modified adsorbent with hydroxyl groups. Desorption studies were also performed with HCl (Baharlouei and Sirousazar 2018). Glass beads coated with chitosan were used for food azo dyes adsorption in a fixed-bed column, and maximum capacity of the adsorption column was found at range of 13.5–108.7 mg/g (Vieira et al. 2014). Other dye removal using column adsorption studies is depicted in Table 5.

Miscellaneous adsorbate

Miscellaneous adsorbates like benzaldehyde, salicylic acid, levofloxacin, etc. are also being removed by column adsorption treatment. Meng et al. studied the column adsorptive removal of salicylic acid on the surface of wollastonite-based imprinted polymer (WMIP). Effect of initial concentration of salicylic acid, column bed height, flow rate and temperature is performed, and data were analyzed by Thomas and Adam and Bohart models (Meng et al. 2013). Feasibility of fixed-bed column filled with activated charcoal prepared from coconut husks for removal of benzaldehyde from its aqueous solution is conducted. Various parameters such as inlet concentration, feed flow rate, bed depth and column inner diameter were evaluated (Canteli et al. 2014). Four types of magnesium (Mg)-impregnated biochars were prepared via thermal pyrolysis of wood chips pretreated with $MgSO_4$ and characterized it with various sophisticated instruments. Batch as well as continuous fixed column experiments was carried out in order to remove antibiotics,

Table 5 Details of column adsorption studies of dye

Dye adsorbate	Adsorbent	Operation parameters	Column isotherm investigated	Thomas maximum adsorption capacity	References
Malachite green (MG)	NaOH-modified rice husk	pH, initial concentration, flow rate and bed height	TM, BDST, ABM and YNM	101.31 mg/g	Chowdhury and Saha (2013a)
Methylene blue	Waste watermelon rind	Initial concentration, flow rate and bed height	TM, BDST and ABM	113.5 mg/g	Lakshmipathy and Sarada (2016)
Acid yellow 17	Tamarind seed powder	Initial concentration, flow rate, pH and bed height	TM, YHM, BDST and ABM	978.5 mg/g	Patel and Vashi (2012)
Methylene blue	Pine cone	Initial concentration, flow rate and bed height	TM, BDST and YNM	55.68 mg/g	Yagub et al. (2015)
Methylene blue	NaOH-modified rice husk	Flow rate and bed height	TM, BDST and YNM	101.3 mg/g	Chowdhury, and Saha (2013b)
Malachite green (MG)	NaOH-modified rice husk	pH, initial concentration, flow rate and bed height	TM, BDST, ABM and YNM	101.31 mg/g	Chowdhury and Saha (2013a)
Allura red AC, tartrazine and sunset yellow FCF	Glass bead-coated chitosan	pH and bed height	TM, BDST and YNM	29.8, 75.1 and 65.6 mg/g	Vieira et al. (2014)
Methyl blue	Biochar and Kaolin	Initial concentration, flow rate and bed height	TM, BDST and YNM	20.06 mg/g	Dawood et al. (2018)

levofloxacin. Effect of different flow rate, initial concentration and bed depth was analyzed (Zhao et al. 2018). Xu and team member prepared carbon nanotube (CNT) and utilized as an adsorbent for removal of 2-naphthol. Process variables (flow rate, initial concentration and bed depth) and column isotherms (Thomas, Yoon–Nelson and BDST) were also analyzed. The breakthrough and exhaust point was calculated. The equilibrium adsorption amount of 2-naphthol on the CNT-based composite adsorbent varies from 122.7 mg/kg to 286.6 mg/kg in this experimental region (Xu et al. 2017). Peng and co-scientists had fabricated, characterized and utilized another adsorbents, amine functionalized magnetic-activated charcoal derived from bamboo wastes (AFM-BAC) and activated charcoal from bamboo wastes (BAC) for adsorption of fluoroquinolone antibiotics ciprofloxacin (CIP) and norfloxacin (NOR) through batch and column method. The saturated adsorption capacities of BAC and AFM-BAC were 172.5 mg/g and 293.2 mg/g for CIP and 193.4 mg/g and 315.7 mg/g for NOR, respectively (Peng et al. 2017).

Atenolol was removed using granular charcoal by Sancho (Sancho et al. 2012) and Sotelo (Sotelo et al. 2012), and their adsorption capacities were 51.10 and 44.36 mg/g, respectively. Comparison of batch and column adsorption studies was performed using activated carbon for removal of pharmaceutical product diclofenac. Initial pollutant concentration, weight of adsorbent and volumetric feed flow rate were analyzed. Breakthrough time, the time when

5% of initial concentration is detected in the effluent, was at higher initial concentration and lower flow rate. Fractional bed utilization increased with the increase in the initial concentration and flow rate, but decreased with higher amount of activated carbon. Breakthrough curves experimental data were fitted using Thomas, Bohart–Adams and Yan analytical models. Yan model showed the highest average of the determination coefficients ($R^2 = 0.9842$) of all experiments, while the amounts adsorbed by the packed column were better predicted by Thomas equation (Franco et al. 2018). The adsorption of ranitidine hydrochloride (RH) onto microwave-irradiated Aegle marmelos Correa fruit shell was also investigated in a fixed-bed column (Sivarajasekar et al. 2018). The removal of total organic carbon from real industrial waste water using polyethylenimine-functionalized pyroxene nanoparticles (PEI-PY) embedded into diatomite by Hethnawi et al. (2017). Removal of acetaminophen from synthetic wastewater in a fixed-bed column adsorption using low-cost coconut shell waste pretreated with NaOH, HNO$_3$, ozone and/or chitosan was performed, and results of maximum adsorption capacity were: ozone-treated GAC (20.88 mg/g) > chitosan-coated GAC (16.67 mg/g) > HNO$_3$-treated GAC (11.09 mg/g) > NaOH-treated GAC (7.57 mg/g) > as-received GAC (2.84 mg/g). This reveals that the ozone-treated GAC is more preferable adsorbent than other investigated adsorbents (Yanyana et al. 2018).

Challenges for utilization

1. As industry is always demanded for low cost, lower discharge, environmental friendly, easily available material usage and least spacious for effluent treatment plant, and most of plant consists of biological treatment as a tertiary treatment due to its vast feasibility; the main disadvantages of any adsorption are that the high price of treatment and difficult regeneration. It also produced solid waste of exhausted adsorbent.

2. Column adsorption studies are considered as better adsorption due to reasonable advantages, but challenge for column adsorption is that as fluid is passed through the fixed bed of solid adsorbents, initially transfer of adsorbate from the feed fluid occurs at the bed entrance. As feed fluid is continuously passed toward the column, MTZ progressively move through the bed once the adsorbent in a region becomes saturated with the adsorbate molecules. After particle time duration, the adsorbent particles upstream or downstream of the MTZ do not participate in the mass-transfer processes, and thus, adsorption process of removing the adsorbate (pollutants) is congested. Thereafter adsorbent must be replaced or regenerated. Fixed-bed column adsorption has facing other problems of poor temperature controller, undesirable heat gradients, un-wanted chemical reactions, channeling and difficult to clean.

3. For proper industrial prospective, series of column should be attached for better adsorption results. Other factors such as column containing multiple adsorbents, numerous adsorbate system and also their appropriate ratio are to be considered.

4. All the experiments are being accomplished using synthetic wastewater of metal, dye and other contaminations including pharmaceutical products in the continuous fixed-bed column studies by various researchers. But, real industrial like textile, dyeing, electroplating, tanning, paper, etc. effluent must be considered for the removal of components contributing the COD, BOD, color and other parameters. Furthermore, regeneration studies and desorption step modeling must be conducted.

Conclusion

From various literature surveys, we concluded that fixed-bed column studies for removal of various contaminations from synthetic wastewater are still in the very infancy. This review paper comprised of adsorption, its types and mechanism, types of adsorbent, adsorbate and adsorption study. Column study is compared with other adsorption studies in tabulated form, which revealed that column study is better, easy, simple, economical and feasible for industrial for removal of various contaminations including dye, metal and other hazardous waste. Breakthrough curves and its parameters are interpreted to design column by various figures. Numerous process parameters are known to have important influence on this phenomenon: initial concentration of adsorbate, flow rate, bed height, pH, particle size of adsorbent and temperature. Detail description of column isotherms models, i.e., Thomas model, bed depth service time, the Adams and Bohart model, Yoon–Nelson, Clark, Wolborska and modified dose–response model, are stated to understand the adsorption system. We have reviewed recent development of different adsorbents in the application of contaminant (adsorbate), such as metal, ion, dye and other pollutants removals using fixed-bed column study concerning to operation parameters, investigated isotherms. Finally, challenges for utilization of the fixed-bed column adsorption study are demonstrated, showing the gaps between pilot and industrial scales.

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