

Fixed Bed Removal of Heavy Metal- a Review

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Abstract:

Removal of heavy metals from the wastewater is major area of research in environmental engineering. Adsorption is very effective method for heavy metal removal. The use of fixed bed for the heavy metal removal by adsorption is important part of the investigations carried out by the researchers. This review paper presents the research carried out for heavy metal removal in fixed bed by using various adsorbents with respect to the affecting parameters, the models used to explain the metal uptake and the adsorption efficiency. Fixed bed studies revealed that the break through time depends on the flow rates and initial metal ion concentrations. The studies carried out by various researchers also explains the appearance of break through curves by using various models such as Adam-Bohart, Thomas and Yoon-Nelson models. Attempts have been made to correlate the batch data to the column data by few researchers. Fixed bed adsorption was found to be very efficient and applicable method for heavy metal removal.

Keywords:

adsorption, models, kinetics, removal, isotherms.

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Introduction

The sources of heavy metals are variety of industrial activities and waste sources. Residual metals in the environment pose a threat to human health.. It can have serious detrimental effects on aquatic ecosystems also. The industrial effluents contain different derivatives of heavy metals such as Cd, Pb, Ni, Cr, As, Cu, Fe etc. Various methods to remove heavy metals include chemical precipitation, activated carbon adsorption, ion-exchange, chemical reduction, flocculation, filtration, evaporation, solvent extraction, biosorption, reverse osmosis, electro dialysis. These methods have their own limitations in terms of cost effectiveness, applicability in certain concentration range and operating difficulties. The adsorption is widely studied method for heavy metal removal because of advantages such as applicability, flexibility in terms of choice of adsorbent and high removal efficiencies. The batch experiments for various heavy metals are reported by investigators. The actual practical approach involves the studies in continuous operations. For this fixed bed has various advantages such ease of operation, flexibility, cost effectiveness and design. The present review aims at studying the fixed bed experiments and investigations with respect to the adsorbent used, parameters affecting the fixed bed operations and the efficiency of removal.

Fixed Beds For Heavy Metal Removal

Fixed bed column study for the removal of lead from aquatic environment by sodium carbonate treated rice husk(NCRH)was carried out by Kumar and Acharya[1].They reported the results of the study on the performance of low-cost adsorbent such as NCRH in removing lead. It was found that the adsorbent materials adopted were an efficient media for the removal of lead in continuous mode using fixed bed column. They conducted the fixed bed column experiment in a column having a diameter of 2 cm with 10 mg/l Pb(II) solution at a bed depth of 10 cm maintaining a constant flow rate of 10 ml/min. According to their investigation,NCRH was found to be efficient media for the removal of Pb(II) from aquatic environment. The column with 2 cm diameter, and bed depth 10 cm could treat 2.22 liters of Pb(II)at breakthrough, at initial concentration 10 mg/l.It was observed that about 3.74 g of NCRH was required per liter of Pb(II) treatment. Height of adsorption zone was found to be 10.32 cm and the rate at which the adsorption zone was moving through the bed was 1.38 cm/h. The values of adsorption rate constant (K) and adsorption capacity constant (No) were obtained as 0.0524 l/mg.h and 1712 mg/l respectively. Nwabanne and Igbokwe, P. K. investigated adsorption performance of packed bed column for the removal of lead (ii) using oil palm fiber[2].. They studied the influence of important parameters like inlet ion concentration, flow rate and bed height on the breakthrough curves and adsorption performance. It was observed that with increase in the bed height the throughput

volume increases because of increase in the active sites. They analyzed the kinetic data using Thomas and Yoon and Nelson kinetic models. The kinetic data were well described by both models. The maximum adsorption capacity, calculated from both models, increased with increase in flow rate and initial ion concentration but decreased with increase in bed height. The comparison of the experimental breakthrough curve to the breakthrough profile obtained from Yoon and Nelson method showed a satisfactory fit for activated carbon derived from oil palm empty fruit bunch. Sadi and Fazaeli carried out research on lead removal in a fixed bed using nanostructure γ -alumina[3]. They compared Thomas, Yoon-Nelson, and Adams-Bohart models with experimental results. The model parameters were evaluated by linear regression analysis for Pb^{2+} adsorption in different bed heights, initial concentrations, and flow rates. The obtained experimental data was in good agreement with Thomas and Yoon-Nelson models, but in the case of Adams-Bohart model, low correlation coefficient was observed. Rice husk based activated carbon was used for copper removal from effluent by Khan et.al[4]. They investigated the effect of initial concentration, feed flow rate and bed height on the breakthrough characteristics of the bed. It was concluded that the fixed-bed adsorption system performed better with lower Cu (II) inlet concentration, lower feed flow rate and higher rice husk activated carbon (RHAC) bed height. The column experimental data fitted well to the Thomas and Yoon-Nelson models. Comparative study of copper (ii) removal on iron oxide, aluminum oxide and activated carbon by continuous down flow method was done by Salmani et.al[5]. They drawn

the experimental breakthrough curves from the adsorption data of packed bed columns. It was observed that uptake capacity of Cu (II) was highest for activated carbon (16.24 mg g⁻¹) followed by aluminum oxide (5.60 mg g⁻¹) and iron oxide (5.41 mg g⁻¹). It was observed that the theoretical adsorption capacities obtained by Thomas model were in good agreement with experimental capacities determined by adsorption data. It was concluded that the packed bed column with continuous down flow method can be successfully applied for the removal of heavy metals from contaminated waters in practical use.

Costodes et. al studied removal of lead (II) ions from synthetic and real effluents using immobilized *Pinus Sylvester's* sawdust in a fixed bed[6]. The investigation revealed that the experimental data closely fitted the bed depth service time model at 10% of the breakthrough curve. They used the data from the bed depth service time model on the mini-column. The performance of the pilot plant column accurately agreed with that obtained from the mini-column. The experiments carried out in a dynamic reactor allowed to bring out the influence of various parameters on the efficiency of the *P. sylvestris* sawdust. Suganthi used tamarind seeds adsorbent for removal of metals in a fixed bed[7]. He studied the removal of copper and cadmium in a fixed bed. He also assessed the effect of pH, flow rate and bed height on the adsorption of metal ions from aqueous solutions. It was observed that breakthrough capacities using wastewater containing cadmium and copper ions and phosphorylated tamarind seed carbon were superior to commercial activated carbon in the removal of metal ions.

Wheat straw as an adsorbent was used for cadmium removal in fixed and batch experiments by Muhamad et.al[8]. They carried out investigation in the flow rate range of 0.3 to 1 lpm and varying bed height from 0.5 to 2 meter. The results obtained agreed to the bed depth service time (BDST) model well. For estimations of the parameters that are necessary for the design of a fixed bed adsorber in practical applications, the experimental data were fitted to the Thomas, Adams-Bohart, and Yan models. It was observed that the Thomas model appeared to better describe the experimental results. Yelebi et.al. designed and developed a mathematical model for fixed bed adsorption column and solved it numerically by implicit backward Euler finite difference method[9]. The parametric study carried out on the model revealed that smaller bed porosity reduces the solute residence time in the bed and consequently increases the adsorption rate and that decrease in particle diameter decreases the breakthrough time. It was also observed that the increase in flow rate increases the adsorbate concentration ratio more rapidly. They concluded that the developed model was suitable and applicable to study the fixed bed adsorption column performance under isothermal conditions. Experimental investigations and theoretical modeling aspects in column studies for removal of Cr(VI) from aqueous solutions using activated tamarind seeds was investigated by Gupta and Babu[10]. It was observed during the investigations that as the flow rate increased from 10 ml/min to 20 ml/min, the breakthrough time decreased from 210 to 80 minutes. With increase in adsorbent mass, the breakthrough time delayed. The breakthrough times are obtained as 110,

115 and 210 min for 15, 20 and 25 g of activated tamarind seeds. As the initial concentration increased from 100 mg/l to 200 mg/l, the breakthrough time changed from 210 mg/l to 45 mg/l. Pore diffusion model was found suitable for explaining the break through curve. Tan et.al carried out continuous packed bed biosorption of copper by immobilized seaweed biomass[11]. Inactivated biomass of the brown seaweed *Sargassum baccharia* immobilized onto polyvinyl alcohol (PVA) gel beads was used for copper removal. They packed the PVA-immobilized biomass beads in a glass column with a diameter of 1.6 cm. Two continuous-flow column tests were performed to determine breakthrough curves at different bed lengths (11 and 13cm). It was found during the investigation that the dynamic behavior of the biosorption column was not entirely predictable on the basis of a simplified two parameter packed bed model. The breakthrough curves obtained in this study exhibited a very broad trailing edge due to slow intraparticle diffusion within the pores of the PVA-immobilized biomass beads. A review on adsorption of metal ions on tea factory waste was done by Wasewar[12]. Insoluble cell walls of tea leaves are largely made up of cellulose and hemicelluloses, lignin, condensed tannins and structural proteins. Disposal of the exhausted adsorbent loaded with heavy metal ions creates another environmental problem such as acid rains. This problem may be overcome to some extent by using one of the elimination methods (e.g. elution, incineration and pyrolysis). Tea waste was found to be very efficient for heavy metal removal. The metal Laden tea waste can be incinerated effectively. Gaikwad has carried out research on copper ion removal from

acid mine drainage by using *Psidium Guava* leaves powder[13]. He studied the effect of bed height, initial concentration and flow rates on the removal efficiency. He observed that the an increase in flow rate decreases the volume treated until the breakthrough point and therefore decreases the service time of the bed. It was concluded that copper can be removed from aqueous solutions very effectively by means of the column exchange process on natural *Psidium Guava* leaves powder. Horse chesnut, oak valonia, peduncle of oak valonia, pericarp of oak valonia, seed of oak valonia, diatomite, brownish and beige sepiolites were used as adsorbents by Murathan et.al[14]. They used fixed bed system for the adsorption. Adsorption capacities of horse chesnut, oak valonia, eduncle of oak valonia, pericarp of oak valonia, seed of oak valonia, diatomite, brownish and beige sepiolites were found. The adsorption capacities estimated in terms of maximum zinc adsorbed per gram of adsorbent were 12.25 mg, 22.75 mg, 87.75 mg, 49.14 mg, 3.34 mg, 5.81 mg, 6.55 mg and 7.15 mg respectively. Chitosan coated bentonite (CHB) in fixed bed for removal of nickel from waste water was used by Futalan et.al[15]. They studied effect of bed depth and initial concentration on service time and shape of breakthrough curve. It was observed during the investigation that the breakthrough curves become steeper and the bed service time becomes shorter with decreasing bed depth, and increasing flow rate and initial concentration. The maximum removal of Ni(II) removal of about 88% was achieved. It was concluded the lowcost CHB is a feasible and effective adsorbent in removing Ni(II) ions from aqueous solution in a fixed bed system. Sulaymon et.al studied the

biosorption of Pb (II), Cd (II), and Hg(II) from simulated aqueous solutions using baker's yeast biomass[16]. It was observed that the Cd (II), Pb(II), and Hg(II) uptake process followed the pseudo-second order rate model with R^2 values of 0.963, 0.979, and 0.960 respectively. Also Langmuir isotherm provided best fitted the solute uptake. A two parameters model for the modeling of breakthrough curves was used. It was observed that an increase in the initial concentration of each adsorbate makes the breakthrough curves much steeper. The reason may be increase in driving force for mass transfer with the increase of adsorbates concentrations. The increase in the flow rate for each solute decreases the breakthrough time due to the decrease in the contact time between the adsorbate and the adsorbent along the adsorption bed.

Medvidovic et.al. studied column performance in lead removal from aqueous solutions by fixed bed of natural zeolite-clinoptilolite[17]. Their investigation was aimed at removal of lead ions from aqueous solutions using the column method with more successive service and regeneration cycles. The highest effectiveness of the column performance has been attained for the zeolite particle size of 0.6–0.8mm, at the initial concentration of 212.5mgPb/L and the flow rate of 2mL/min. It was also observed that the effluent volume (BV) at the breakthrough point decreases with the increase of lead concentration in the initial solution, at the same flow. They performed eight service and regeneration cycles. For this 70–75BV of the sodium nitrate solution was This is a volume 4–5 times smaller than the lead solution volume per service cycle. The lead concentration in the regenerate is

higher so that the solution of lead can be reused. The total of 850 mg of lead was removed per gram of zeolite. Nwabanne and Igbokwe carried out studies on the adsorption of lead (11) and copper (11) in a fixed bed column using activated carbon prepared from nipa palm nut [18]. It was observed that the rate constant for Thomas model increased with increase in flow rate and initial ion concentration but remained constant at varying bed height. The maximum adsorption capacity increased with increase in flow rate and initial ion concentration but decreased with increase in bed height. Also they observed that the time required for 50% breakthrough decreased with increase in flow rate, bed height and initial ion concentration. They concluded that the experimental breakthrough curve compared satisfactorily with the breakthrough profile calculated by Yoon and Nelson method. Vassilis studied ion exchange and adsorption fixed bed operations for wastewater treatment with emphasis on modeling fundamentals and hydraulics analysis [19]. The overall effectiveness of a fixed bed operation depended mainly on its hydraulic performance. Medvidovic et al. studied the fixed bed column for lead removal by natural aeolite [20]. They presented the prediction of breakthrough curves for the fixed bed column based on batch studies. There was very good agreement between experimental and calculated breakthrough curves. It indicated that the Mass transfer model is applicable for prediction of the breakthrough curve from batch experiments. Kafshgari et al. used marine algae *Cystoseria indica* pretreated with 0.1 M CaCl_2 solution in a packed bed column for molybdenum (VI) biosorption [21]. The column was a simple glass tube with inner

diameter of 1.5 cm and length of 10 cm. Two plastic sieves both with pore size of 0.5 mm were installed at the top and bottom of this column. The controlled-rate step shifted from external to internal mass transfer limitations, as the flow rate increased. They applied Thomas, Yoon-Nelson, Belter and Yan models to the experimental data obtained from the biosorption of Mo (VI) ion onto *C. indica*. Among these models, the Belter model appeared to describe the experimental results better. The maximum biosorption capacity of Ca-pretreated *C. indica* for Mo (VI) was found to be 18.32 mg/g at optimum flow rate of (1.4 mL/min). It was observed that the breakthrough and exhaustion time decreased from 17.14 hr to 9.05 hr and from 0.006 h to 0.002 hr respectively, with the increase of flow rate from 0.7 to 2.1 ml/min.

The studies on application of physical models and artificial neural network for prediction of breakthrough for adsorption of Fe(II) from aqueous phase by chitosan were done by Radnia et al. [22]. They carried out batch experiments at initial concentration range of 10-50 mg/L and temperature range of 20–40°C. Adsorption capacity of 28.7 mg/g and removal efficiency of 93% were obtained in batch experiments. They performed the fixed bed experiments in continuous up-flow mode and constant temperature of 25°C. It was observed that compared to physical models, simulation of dynamic behavior of the system using Back Propagation Artificial Neural Network (BP-ANN) demonstrated high coincidence between experimental and predicted breakthrough curves. The study was carried out by Kumar and Acharya on the performance of low-cost adsorbent such as NCRH in removing

copper[23]. They performed the fixed bed column experiment in a column having a diameter of 2 cm with 10 mg/l Cu(II) solution at a bed depth of 10 cm maintaining a constant flow rate of 10 ml/min. The result indicated that height of adsorption zone was 10.21 cm and the rate at which the adsorption zone was moving through the bed was 1.48 cm/h. They estimated the values of adsorption rate constant (K) and adsorption capacity constant (No) as 0.056 l/mg.h and 1623 mg/l respectively. Saw dust was used for heavy metal removal from mine waste by Velizar et.al[24]. Results on the batch and column adsorption of copper and some associated ions by employing linden and poplar sawdust as a low-cost adsorbent were presented. The adsorbent was found to be satisfactory for copper removal with highest adsorption of 80 percent. They verified results obtained in the batch mode through the column test by using the real mine water originating from an acid mine drainage (AMD) of the copper mine. The results were in agreement with each other. A research on heavy metals removal in fixed-bed column by the macro fungus *Pycnoporus sanguineus* was carried out by Zulfadhly et.al[25]. They investigated the ability of *Pycnoporus sanguineus* to adsorb heavy metals from aqueous solution in fixed-bed column. Design parameters such as column bed height, flow rate and initial concentration of solution were studied during investigation. Comparison was done between experimental breakthrough profiles and the simulated breakthrough profiles obtained from the mathematical model. They used bed depth service time model (BDST) to analyze the experimental data. Also they evaluated the BDST model parameters needed for the design of

biosorption columns. for lead, copper and cadmium removal in the column. The experimental and simulated data was in agreement with each other. A research on biosorptive removal of heavy metal from industrial effluent by fixed-bed column of red microalgae was carried out by Ibrahim et.al[26]. They used three different species of nonliving red algal biomass *Laurancia obtusa*, *Geldiella acerosa* and *Hypnea* sp. By using these, three types of fixed-bed columns were built for the removal of toxic heavy metal ions such as Cu^{2+} , Zn^{2+} , Mn^{2+} and Ni^{2+} from industrial effluent. Algal column of *L. Obtusa* gave highest removal efficiency. It was concluded that red microalgae were very effective in removing heavy metals with removal efficiency of 94 percent. Ghorbani et.al. used polyaniline and its nanocomposite (PAN) containing nanometer-size Fe_3O_4 in fixed bed for the removal of anions and heavy metals from cotton textile waste water[27]. It was observed that PAN and its composites were able to remove the anions and heavy metals. Rivera investigated the heavy metal removal in a packed bed, anaerobic upflow reactor (Anflow)[28]. The anaerobic bioreactor was demonstrated for municipal wastewater at 19 cubic meter per day. It was observed that heavy metal removal in an Anflow bioreactor was similar to an ion exchange process. Here a saturation transient began at the bottom of the column and moved upward. Adsorption experiments were carried out by electric arc furnace slag (EAFS) in a fixed-bed column mode by Beh et.al.[29]. It was confirmed that the EAFS was effective in the removal of heavy metals via adsorption. The zinc was reduced from 4.02 mg/l to 0.01 mg/l, and iron from 23.3 to 0.08 mg/l. Henriques et. al. reported manganese removal by

using *Sargassum filipendulain* a batch fixed bed experiments[30]. They applied a mathematical model based on mass balances in the fluid and in the sorbent to represent the experimental fixed-bed column data. The isotherm data from batch experiments was utilized for breakthrough curve. It implied a significant mismatch in relation to the laboratory data. The fixed-bed column data and the corresponding simulated profile of the breakthrough curve were in agreement to the experimental results. Research on the batch and fixed-bed column performance of red mud adsorbents for Lead removal was carried out by Mobasherpour et.al[31]. It was observed that The breakthrough time and exhaustion time decreased with increasing flow rate, decreasing bed depth and increasing influent lead concentration. The bed depth service time model and the Thomas model were in well agreement with the experimental data. Batch sorption and fixed-bed experiments were conducted by Tian et.al.to examine the ability of functionalized multi-walled carbon nanotubes (CNTs) as filter media to remove two heavy metal ions(Pb^{2+} and Cu^{2+}) from infiltrating water[32]. Fixed-bed columns were packed with CNTs and natural quartz sand by three different packing: layered, mixed, and deposited. It was observed that the addition of CNTs in very small amount(0.006 %) increased the fixed-bed's filtering efficiency of Pb^{2+} and Cu^{2+} by 55–75% and 31–57%, respectively.

Conclusion

The removal of many heavy metal such as chromium, cadmium, copper, nickel, lead, mercury, iron can be carried out by using low cost adsorbents. The fixed bed

arrangement can be used for this purpose very efficiently. The research reveals that the maximum removal up to 90 to 99 percent can be achieved. The shape of break through curve, break even time and saturation time depends on the flow rate, initial concentration and bed depth. With increase in the flow rate and initial concentrations, the break through time decreases. With increase in mass of adsorbent, the breakthrough time is delayed. The prediction on the breakthrough times at various flow rate and initial concentrations by various models was in agreement with experimental results. It was also found that the batch data is very useful and can be used to predict break through characteristics. It can be concluded that fixed bed adsorption is very good alternative for heavy metal removal.

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