



Review Paper

Removal of Phenol from Wastewater in Packed Bed and Fluidised Bed Columns: A Review

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Abstract

Water contaminated by phenol is becoming a major problem in water supplies as these organic compounds cause hazards for human health and environment. Different types of columns have been used for water cleanup of organic pollutants from wastewater. Many studies report that adsorption using contacting devices are a very efficient method for phenol removal from contaminated water. This paper reviews recent research in the packed bed and fluidised bed columns used for the adsorption and biodegradation of phenol, along with emphasis on their configuration and design, mechanism of operation and the future research and development requirements.

Keywords: Phenol, wastewater, adsorption, packed bed, fluidised bed.

Introduction

The US Environmental Protection Agency (EPA) designated phenols as priority pollutants because of its toxicity and adverse effects upon environment¹. The wastewaters discharged from pesticide, paint, solvent, pharmaceuticals, paper and pulp industries and also from water disinfecting process will contribute to main source of phenol². The different treatment methods to eliminate phenolic compounds from aqueous solutions like biological treatment methods, pervaporation, membrane based solvent extraction process, extraction, distillation, adsorption on activated carbon and advance oxidation processes were reviewed by Girish and Ramachandra Murty³. Adsorption method using activated carbon is found to be efficient in treating of highly concentrated phenolic wastewaters. Activated carbon is extensively used for the removing a variety of organics from wastewater. The production of activated carbon can be done from either naturally occurring or synthetic carbonaceous material. The adsorptive and physical properties that are acquired in the final activated carbon depends on the selection of precursor material⁴. Girish and Ramachandra Murty addressed a major economic factor that the activated carbon produced from expensive precursor material like coal is costly and non renewable in nature⁵. This has motivated a budding research interest for producing activated carbons from numerous agricultural waste material for wastewater treatment as reported by Girish and Ramachandra Murty⁶.

The previous studies on pollutant removal were limited to batch studies because of the inability to treat bulk quantity of material. So continuous studies using packed column was favoured over the batch studies over time as it had more advantages. This review provides an overview of recent work done in packed bed

and fluidised bed columns for removing phenol and other pollutants from wastewater using agricultural waste as adsorbents.

Packed bed and fluidised bed columns for wastewater treatment

The phenol removal in packed bed columns: Packed bed columns are contacting device in which wastewater containing pollutants is passed through a column containing porous adsorbent and the pollutant gets adsorbed onto the surface of the adsorbent. It is preferred for its ease of operation, high yields, high liquid residence times and can be scaling up from a laboratory step⁷. It boosts the interfacial area while decreasing the mass transfer resistance between gas and liquid phases inside the column⁸. It is also favoured for the removal of pollutants present in very low concentration in large quantity of wastewater⁹. The various works with packed bed reactors for the removing phenol and other pollutants from wastewater are compared. The different operating conditions were studied and various models developed for the adsorption are reported.

The packed bed filled with activated carbon of various particle sizes 1.47, 0.8 and 0.45 mm, made from waste dates stone were utilised by Alhamed for the breakthrough studies of phenol adsorption from aqueous solution¹⁰. The variation of bed height of 5, 10, 15, 20 and 25 cm, initial phenol concentration of 50, 100 and 150 ppm, and flowrate of 23.3–141.5 ml/min were studied for the phenol removal. The breakthrough curves obtained from the experiments were analysed using an axial dispersion model and equivalent length of unused bed model (LUB).

The phenol adsorption from aqueous solution in a packed bed containing activated carbon made from *P. pinaster* bark was tested by Vazquez et al.⁷. From the experiments, the equivalent length of unused bed was investigated using the LUB design approach. The results elucidated that the length of unused bed increases with decreasing flow rate.

The phenol containing wastewater from petrochemical, leather and polymer industries were treated with microorganism *Pseudomonas pictorum* (NCIM 2077) immobilized on alginate and activated carbon-alginate beads in a upward packed bed column by Murugesan and Sheeja¹¹. The estimation of mass transfer coefficient for the phenol degradation was found. The evaluation of components pH, colour, odour, total dissolved solid, total suspended solid, phenol content, biochemical oxygen demand and chemical oxygen demand were carried out. A model was developed for the mass transfer of phenol from the bulk liquid to the surface of the immobilized beads.

Tziotzios et al. investigated the degradation of phenol using indigenous bacteria from olive pulp in two packed reactors supported with calcite gravel and plastic tubes¹². The draw-fill operating mode were examined for both the reactor. But in the reactor packed with plastic tubes the draw-fill operating mode with recirculation was also investigated. It was found that the maximum phenol removal rate of 25.3 g phenol/d and 14.03 g phenol/d were achieved for the reactor packed with calcite gravel and plastic tubes respectively for a phenol feed concentration of 500 mg/l. It was concluded that by increasing the phenol concentration to about 2750 mg/l, the bacterial growth was inhibited.

According to Sahoo et al. a packed bed column with cross flow air circulation at different ports used for the biodegradation of p-nitrophenol (PNP) by *Arthrobacter chlorophenolicus* A6 improved the hydrodynamic behaviour inside the column¹³. The total PNP removal accomplished in the column was 99.9% removal for a feed loading rate of 2787 mg/l d for an influent concentration of 1400mg/l and 18 h hydraulic residence time.

A bioreactor having internal loop air circulation and having honeycomb-like ceramic as the carrier and immobilized with bacteria, *Achromobacter* sp. was proposed by Quan et al. for degrading 2,4-dichlorophenol (2,4-DCP) and phenol in both batch as well as continuous mode¹⁴. It was examined that when the reactor was fed with the mixed proportion of phenol and 2,4-DCP and operated in batch mode, phenol inhibited the biodegradation of 2,4-DCP, resulting in the decrease of 2,4-DCP removal rate from 100 to 87.9%. It was also reported that the phenol present in the feed altered the major carbon source change from 2,4-DCP to phenol.

The packed bed made of activated carbon processed from oil-palm shells were explored for phenol removal from aqueous solution by Lua and Jia¹⁵. The effects of flow rate, initial concentration and bed height of packed bed on the phenol

removal were studied. It was found that by increasing phenol flow rate and initial concentration in packed bed resulted in steep breakthrough curves and short breakthrough times. It was also reported that with a smaller bed height, the ratio of the effluent to the influent adsorbate concentration reached unity faster than that for a higher bed height.

The phenol biosorption from aqueous solution on immobilized dried activated sludge was investigated in a continuously operated packed bed column by Aksu and Gonen¹⁶. The influence of flow rates and inlet phenol concentrations on the breakthrough curves for phenol sorption were analysed. It was reported that the phenol sorption increased with decreasing the flow rate and with increasing inlet phenol concentration.

According to Tan et al. the activated carbon obtained from oil palm shell was used as adsorbent in a packed column for removing trichloro-phenol from aqueous solutions¹⁷. The effect of the initial concentration, feed flow rate and bed height on the removal rate was evaluated. The problem of regeneration of adsorbent was overcome by desorption using ethanol solvent in recovering the spent activated carbon.

Nava et al. improved the phenol sorption from aqueous solution in a packed bed having surfactant modified zeolite rich tuff as adsorbent¹⁸. It was illustrated that the adsorption was influenced with the amounts of surfactant in the zeolitic-rich tuff, the kind of surfactant and the initial pH of the phenol solutions. It was clarified that the phenol sorption behaviour was due to the modification of the hydrophilic character of the zeolitic tuff surface.

According to the work published by Adak and Pal, the experiments were performed for phenol removal in a packed column of 2 cm diameter containing surfactant-modified alumina¹⁹. The adsorption behaviour was described for phenol removal in packed bed having bed heights of 10, 20 and 30 cm. The depth of exchange zone, breakthrough time and adsorption capacity on phenol removal were evaluated.

The feasibility of upflow fixed-bed column containing activated carbon used for the removal of catechol from aqueous solution was explored by Richard et al.²⁰. The critical time required for the breakthrough of the required concentration was determined from Homogeneous Surface Diffusion Model (HSDM) which defines relation between adsorption equilibrium and mass-transfer kinetics.

The dye adsorption from water onto the surface of montmorillonite and montmorillonites activated with hydrochloric acid in packed bed was examined by Teng and Lin²¹. The effect of flow rate, initial dye concentration and bed length on the adsorption was evaluated. It was concluded that the low flow rate of feed increased the residence time of the feed solution and therefore the solute molecules have more time to penetrate into the pores of the adsorbent and thus increased

the adsorption. The activation with acid increased surface area because of the nature of ionic size in the acid.

In the work published by Vijayaraghavan et al., the crab shell carbon particles were utilised to remove nickel from aqueous solution in a up flow packed column²². The experiments were carried out with various bed heights and different flow rates. The studies for the regeneration of sorbent were performed with seven sorption-desorption cycles using 0.01M EDTA as elutant. It was also found that due to continuous use of crab shell carbon, the breakthrough curves became more flattened and breakthrough time decreased from 28.1 to 9.5 h as the cycles progressed from one to seven.

Ahmad and Hameed proposed chemically treated granular activated carbon (GAC) prepared from bamboo waste for the adsorption of Reactive black (RB5) from aqueous solutions in a packed column²³. The packed column studies were performed by varying feed inlet concentration, feed flow rate and bed height. The experimental data were examined by the Adam's-Bohart, Thomas and Yoon-Nelson models and the data were fitted best to the Thomas and Yoon-Nelson models.

A packed column containing olive pomace and charcoal was successfully utilised by Al-Asheh et al. for removing of Methylene Blue (MB) dye from aqueous solution²⁴. It was enlightened that the removal of dye increased with increasing bed height, decreasing feed flow rate and decreasing feed initial concentration. It was elucidated that the behaviour of the breakthrough curve were enhanced by packing the column with multiple layers of olive pomace carbon and charcoal.

The activated carbon prepared from rubberwood sawdust used for the removal of Bismark Brown dye in a packed column was examined by Prakash Kumar et al²⁵. The influence of bed height, flow rate and initial concentration of the solution on the adsorption were described. The bed depth service time (BDST) model was proposed by varying the flow rate and initial concentration. The adsorption capacities were reported to be 2000 and 1111 mg/g for steam treated carbon and chemical-steam treated carbons respectively.

The Phenol Removal in fluidised bed Columns

Fluidized column are contacting device in which a fluid (liquid or gas) is passed up through the porous material and the material is converted from a static solid state to a dynamic fluid state. The advantage of the fluidised column are that it makes it possible to attain phase homogeneity and enhances solid-fluid contact interfacial area which enable to handle high volumetric loading²⁶. Furthermore that it possess a high mass and heat transfer rates and non clogging of the bed²⁷. In addition it prevents the formation of hot spots even with highly exothermic reactions because of heat transfer between gas to particle and bed to wall. It thus lead to the enhancement of pore and surface area, which directly describe its adsorption capacity²⁸. The

recent research carried out to remove the phenol and other pollutants from wastewater is enumerable. And also the operational feasibility of the fluidised bed reactors is studied from the relevant works like regeneration and production of adsorbent.

A fluidised bed bioreactor with gas distributor for axial mixing was utilised by Ochieng et al. for the treatment of petroleum industry wastewater containing phenol²⁶. It was shown that low fluidisation velocity of 2.7cm/s was economical for the wastewater treatment.

The influence of the solution flow rate and pollutant initial concentration on the feasibility of fluidised bed used for the removal of phenol from wastewater were tested by McKay²⁹. It was found that the adsorption increased with the flow rate of solution and a model was developed to show the relationship between the external mass transfer coefficient and the liquid-phase Reynolds number.

Koran et al. demonstrated the feasibility of a fluidized-bed reactor packed with granular activated carbon (GAC) to treat synthetic wastewater containing pentachloro phenol (PCP) and four other polycyclic aromatic hydrocarbons (naphthalene, acenaphthene, pyrene, and benzo (b) fluoranthene) under anaerobic conditions³⁰. It was enumerated that the reactor achieved a removal efficiency upto 99.8% for PCP and removal efficiency of 86 and 93% for naphthalene and acenaphthene respectively. The concentration of pyrene and benzo (b) fluoranthene in the effluent were extremely low due to the high-adsorptive capacity of GAC for these compounds.

Vinod and Reddy studied the phenol degradation by using the microorganism *Psuedomonas putida* immobilized on solid particles in a fluidized bed bioreactor³¹. It was found that the phenol gets transferred from the bulk phase to the surface of the bio film and the mass transfer coefficient was found to have the value varied from 0.0726×10^{-5} to 0.2012×10^{-5} m/s.

The phenol biodegradation in a fluidized column having *Pseudomonas putida* cells immobilized within calcium alginate gel beads was reported by Gonzalez et al.³² The effect of the hydraulic residence time and organic loading rate on the removal efficiency of phenol was explored. From the results, it was concluded that the bioreactor showed high phenol degradation efficiency of about 90%.

Mungmart et al. correlated the phenol decomposition rates in a three-phase fluidized-bed reactors utilizing various combinations of advanced oxidation processes such as ozone alone, titanium dioxide deposited on silica beads, metal catalyst either nickel or cobalt impregnated on mesoporous carbon beads and ozone in combination with each catalyst³³. It was proved that the use of cobalt catalyst with the presence of ozone led to the best removal of aqueous phenol which decomposed within a period of 10 min, with rate constant value of 0.1944 min^{-1} .

The suitability of immobilized soybean peroxidase bound to glass supports with varying surface area were employed by Gomez et al. in fluidized bed reactor to study phenol degradation³⁴. It was reported that when derivatives immobilized on supports have the highest surface area 80% removal was achieved.

The combined effect of phenol decomposition with ozone and adsorption on activated carbon was evaluated in a three phase fluidised bed reactor by Charinpanitkul et al³⁵. The activated carbons (AC1 and AC2) with surface areas of 1106 and 1150 m²/g and average pore diameters of 2.3 and 1.7 nm, respectively, were used for the experiment. It was found that AC1 which had higher mesopore fraction could remove more intermediates than AC2. Also it was summarised that with the combined action of AC1 and ozone the phenol removal was up to 100%.

The degradation of phenol in fluidized bed using different combinations of advanced oxidation processes such as ozone, ozone/UV light, ozone/UV light/activated carbon, oxygen/UV light/Titanium dioxide and ozone/UV light/Titanium dioxide were correlated by Dong et al³⁶. It was found that the photocatalytic ozonation gave the maximum phenol conversion because of the combined effects of homogenous ozonation in the liquid phase and heterogeneous ozonation on the surface of the catalyst. It was also concluded that heterogeneous photolytic ozonation (in the presence of particles) was faster than homogeneous photolytic ozonation.

Conclusion

This study provides an bird eye view of the packed and fluidised bed columns used for treatment of wastewater containing phenol and also on the different operational conditions and their performance. However packed bed and fluidised bed columns received great requirement over the recent time due to their numerous advantages such as high operational simplicity, better yields, high liquid residence times, increasing the interfacial area between gas and liquid phases, low energy consumption and low chemical consumption. But still, columns require improvement to have better performance compared to existing designs. The bioreactor should address factors like large pollutant loadings, high temperature, handling water insoluble compounds and scaling up the reactor to industrial scale. Hence, to enhance the performance of the reactors for phenol adsorption, there is an indispensable requirement of novel efforts to be done in the reactor design.

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