Review Paper (NS-1)

ADSORPTION OF PHENOL FROM WASTEWATER USING LOCALLY AVAILABLE ADSORBENTS

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Received November 25, 2011 *Accepted Feb 5, 2012*

ABSTRACT

Phenol and its derivative compounds have received considerable attention from environmentalists, due to their undesirable effects in the water environment, where they end up as a result of improper disposal methods. As a result, concern about environmental protection has increased over the years from a global viewpoint. So research has been conducted all around with the common goal of reducing their concentrations to allowable limits or converting them to non-toxic, non-hazardous forms that may be released easily into the environment. Adsorption process has been proven one of the best water treatment technologies and activated carbon is undoubtedly considered as effective adsorbent for the removal of phenol from water. However, widespread use of commercial activated carbon is sometimes restricted due to its higher costs. Investigations have been made to develop inexpensive adsorbents utilizing numerous agricultural waste materials. By using these waste materials as locally available adsorbents is attractive due to their contribution in the reduction of costs for waste disposal, therefore contributing to environmental protection. In this review, the technical feasibility of locally available adsorbents prepared by utilizing different types of waste materials from literature has been compiled and their adsorption capacities for phenol removal as available in the literature are presented.

Key Word : Adsorption, Locally available adsorbents, Phenol and its derivatives, Wastewater treatment, Waste utilization, Adsorption capacities.

INTRODUCTION

Increasing stringent legislation on the purity of drinking water has created a growing interest in the treatment of wastewater and polluted effluents. Phenol and substituted phenols are one of the important categories of aquatic pollutants, which are considered as toxic, hazardous and priority pollutants[1]. The main sources of phenol which are released into the aquatic environment are the wastewater from industries such as coke ovens in steel plants, petroleum refineries, resin, petrochemical and fertilizer, pharmaceutical, chemical and dye industries [2]. Several treatment methods have been applied to remove phenolic compounds from aqueous solutions, such as biological treatment using live and dead organisms, catalytic wet oxidation and adsorption technology using activated carbons prepared from various precursors. Other methods include air stripping, incineration, ion exchange and solvent extraction.

Adsorption on activated carbon is currently being used and is most effective in treating high strength and low volume of phenolic wastewaters. Activated carbon is the most widely used for the removal of a variety of organics from wastewater. Activated carbons can be produced from any carbonaceous solid precursor: Both naturally occurring and synthetic. The selection of the precursor essentially determines the range of adsorptive and physical properties that can be attained in the activated carbon products. But the disadvantage associated with it is its high cost due to the use of non-renewable and relatively expensive starting material such as coal, which is

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a major economic consideration. This has prompted a growing research interest in the production of activated carbons from locally available materials especially for application concerning wastewater treatment[3]. Several agricultural byproducts have been found to be suitable precursors for production of activated carbon. Because the biomass obtained from these materials are cheaper, renewable and abundantly available and have high carbon and low ash contents[4].

AIMS AND OBJECTIVES

The main goal of this review is to provide a summary of information concerning the use of locally available materials as adsorbents for the removal of phenolic compounds. A comparative overview of some locally available adsorbents based on recent publications is therefore presented in this paper and their removal performances are reported[5].

Agricultural waste materials

Agricultural waste materials are those obtained as the by-products from the forestry and agricultural industries. These are generally inexpensive, renewable source of activated carbons and often present a disposal problem. The agricultural waste materials are organic compounds comprised of cellulose, hemicelluloses, lignin, lipids, proteins, simple sugars, water, hydrocarbons, and starch, containing a variety of functional groups. Therefore it is having potential sorption capacity for various organic pollutants.

Agricultural waste is a rich source for activated carbon production due to its low ash content and reasonable hardness[5], therefore, conversion of agricultural wastes into low-cost adsorbents is a promising alternative to solve environmental problems such as disposal of waste and also to reduce the preparation costs[6].

Recently, research has been focused on the preparation of activated carbons from agricultural by-products such as coconut shell, husk, shells, rice husk seed coat, stone, kernels and peels of various agricultural products, palm oil empty fruit product, miscellaneous agricultural wastes like bark, wooden, roots and other products like straw, fibers, bagasse, hulls.[7] A summary of some relevant published data with some of the latest important results and giving a source of up-to-date literature on the adsorption properties, preparation methods of some alternative adsorbents used for phenol removal is presented in **Table 1**, and the results are discussed here.

Coconut *(Cocos nucifera)* is one of the major crops which generates huge amount of solid waste in the form of shell, husk and fibers. So it forms the precursor for production of activated carbon. Activated carbon was prepared from coconut shell through KOH-CO_2 physiochemical activation at 850^0C by Mohd Din et al.[5] The adsorption isotherms were conformed by both Langmuir and Freundlich isotherm models and adsorption system followed the pseudo second order kinetic model. Adsorption capacity of 205.8mg /g was obtained.

Coconut shell has been converted to activated carbon through chemical activation with KOH by Hu and Srinivasan[8] . The properties of the carbon produced were dependent on the impregnation ratio and the activation temperature. The removal capabilities were found to be 206, 267 and 257 mg/ g for phenol, 4-chlorophenol and 4-nitrophenol respectively.

Coconut husk based activated carbon were explored as adsorbents for the removal of 2,4,6 trichlorophenol (TCP) from aqueous solution by Hameed et al[9] . It was found that the adsorption increased with increase in agitation time and initial concentration while acidic pH was favorable for the adsorption of TCP. The maximum adsorption capacity was 716.10 mg/g.

Coconut husk was used to remove 2,4,6-trichlorophenol under optimized conditions by Hameed et al.[10] The effect of activation temperature, activation time and KOH to char impregnation ratio were studied. The adsorption capacity was found to be 191.73 mg/g.

Namasivayam and Kavitha[11] utilized coir pith carbon has an adsorbent for understanding the mechanism of the phenol removal. It was found that the adsorption capacities of 48.31, 19.12 and 3.66 mg/g were obtained for phenol, 2,4-dichlorophenol and p-chlorophenol. The FTIR studies shows that the participation of the specific functional groups in adsorption interaction, while SEM studies visualized the formation of the adsorbed white layer on the phenol surface.

The applicability of shells, seed coat, stone and kernels of various agricultural products as adsorbents for the removal of toxic pollutants from water has been investigated. The feasibility of activated carbon from almond shell, hazelnut shell, walnut shell and apricot stone for the removal of phenol has been investigated by Ayguun et al.[12] and found that the adsorption capacity of 70.4, 100, 145 and 126 mg/g was obtained for almond shell, walnut shell, hazelnut shell and apricot stone respectively. It was found that the impregnating agents and activating agents had influence on phenol removal.

The effectiveness of the almond shell carbon for the treatment of pentachlorophenol from water were performed by Santos et al.[13] and a saturation adsorption capacity of 9.6 mg/g was obtained under continuous flow experiments. The nature of sorption on almond shells carbon was understood by focusing on the structural and chemical characterization of the carbons.

Date stones were converted into a carbonaceous adsorbent and used for the removal of phenol from wastewater by Alhamed[14]. Packed bed studies were performed using four different activated carbon particle sizes of 1.47, 0.8, 0.45 and 0.225mm and the initial rate of adsorption predicted were from the pseudo-second order model. The adsorption capacity of 90.4 mg/g was obtained at higher initial pollutant concentration. It was proposed that the adsorption decreased with increasing particle diameter as a result of higher interfacial area provided by particles with smaller diameter.

Naas et al.[15] investigated date pit from agricultural solid wastes to prepare effective adsorbent for the removal of phenol from refinery and synthetically prepared aqueous solution wastewater. The kinetics data were best fitted by the pseudo-second order model. The adsorption capacities were found to be 16.64 and 12.6 mg/g for refinery and aqueous solutions respectively. Various chemical and thermal methods were tested for the regeneration of saturated activated carbon and ethanol found to be the most effective with more than 86% regeneration efficiency.

In the recent work published by Mazzoco et al.[16] black stone cherries has been used as raw material in the preparation of granular activated carbon by chemical activation with phosphoric acid for the sorption of phenol from dilute aqueous solutions. The optimum carbonization temperatures were found between 500 and 550^{0}C with an activation ratio of 1.0 g of phosphoric acid/g raw material. The maximum adsorption of 133.33mg of phenol/g GAC was obtained.

Batch adsorption studies were carried out for evaluating the suitability of plum kernels as an adsorbent for the removal of phenol from waste water by Juang et al.[17] The activation temperature and time tested were in the ranges 750-900^{0}C and 1-4 h, respectively and the adsorption capacity was 257.4mg/g.

The potential of nutshells of Sterculia alata, a forest waste to prepare activated carbons by zinc chloride activation to remove phenol from aqueous solution was evaluated Mohanty et al.[18] Experiments were carried out at different impregnation ratios, carbonization temperature and time. The maximum uptake of phenol was found at pH 3.5 and adsorption capacity was 2.8 mg/g.

Corn cob carbon nut shells were explored as adsorbents for the adsorption of phenolic compounds by Tseng and Tseng[19]. Activated carbons were prepared with KOH/char ratios from 0.5 to 6. Activated carbons with the KOH/char ratio from 0.5 to 2 show a cottony, twisted, and deformed surface of honeycomb holes with contracted walls while carbons with ratio between 3 and 6, exhibit regular honeycomb holes with thick and smooth walls are clearly visible. The adsorption capacities at impregnation ratio of 2 and 6 were found to be 232 and 340 for phenol and 362.8 and 485 for chlorophenol and 333.7 and 451.2 mg/g for 2,4 - dichlorophenol respectively.

Recently researchers investigated the applicability of peels of agricultural waste materials as the precursors for activated carbon. The potential of activated carbon from banana peel for the adsorption of phenolic compounds was tested by Mandi et al.[20] and an adsorption capacity of 689 mg/g), revealing that banana peel could be employed as a promising adsorbent. The adsorption process was very fast, and it reached equilibrium in 3 h of contact. Pomegranate peel was converted into a carbonaceous adsorbent and used for the removal of 2,4-dichlorophenol from water by Bhatnagar et al.[1] and the maximum adsorption was 65.7 mg/g of adsorbent.

The seeds of some agricultural byproducts have been explored as effective adsorbents. The adsorption of phenol on palm seed coat activated carbon was evaluated by Rengaraj et al.[21]. The adsorption capacity of palm seed coat activated carbon was 18.3 mg /g for the particle size of 0.5 mm. In comparison with commercial activated carbon palm seed coat carbon, has a lower ash content which indicates more carbon content.

Rubber seed coat as an adsorbent for the adsorption of phenol from aqueous solution and resin manufacturing industry wastewater was investigated by Rengaraj et al.[22]. The adsorption of phenol followed first order reversible kinetics. The adsorption capacities for rubber seed coat carbon are 56 mg/g for synthetic phenolic solution and 36 mg/g for live phenolic wastewater, respectively. Spent carbon can be recovered by desorption with sodium hydroxide.

Palm oil empty fruits and palm pith have been investigated as adsorbent by various researchers for the removal of phenolic compounds from water. The adsorption behavior of 2,4,6-trichlorophenol on oil palm empty fruit bunch-based activated carbon was investigated by Hameed et al.[23]. The optimum activation conditions such as activation temperature, time and impregnation ratio with KOH were found to be 814 ^{0}C, 1.9 h and 2.8 respectively. The adsorption capacity was 168.89 mg/g and the textural characterization results revealed that the activated carbon was mesoporous in nature.

Palm pith carbon has been used for the removal of 2,4- dichlorophenol from aqueous solution by Sathishkumar et al.[24]. The effect of various parameters like agitation time, adsorbent dose, pH and temperature were studied. It was found that the acidic pH was favorable for the adsorption of 2,4-DCP and the adsorption capacity of 19.16 mg/g for the particle size of 250-500 µm was reported.

Oil palm empty fruit bunch-based activated carbon has been utilized for the removal of 2,4,6-trichlorophenol and desorption studies were carried out by Hameed et al.[25] and desorption of 99% was achieved by ethanol desorption technique. The maximum monolayer adsorption capacity was 500 mg/g.

The adsorbent from oil palm empty fruit bunch were found effective for the removal of phenol from water by Alam et al.[26] by physical activation with nitrogen gas at 900^{0}C to produce powder activated carbon. It was reported that the adsorption capacity was 4.868mg /g.

Porous carbons were prepared from oil palm empty fruit bunches by Ling et al.[27]. Carbons were impregnated with different KOH ratio and phenol removed came out to be 90.09, 91.74 and 89.29 mg/g for 30, 50 and 70% KOH respectively. The Scanning Electron Microscopy (SEM) study of the carbon powders revealed that the porous structures enhanced as the percentages of KOH increased.

The roots of the agricultural materials were found to be potential adsorbents for water treatment. The root of an Indian plant Hemidesmus indicus has been used as raw material in the preparation of powder activated carbon by chemical activation with sulphuric acid by Srihari and Das[28] for the sorption of phenol from aqueous solutions. The adsorption capacity was 16.10 mg/g and the adsorption data followed pseudo-first order, pseudo-second order model and pore diffusion model.

Beet pulp, a major by-product in sugar industry was converted into a carbonaceous adsorbent and used for the removal of phenol from wastewater by Dursun et al.[29]. It was produced by carbonisation in N_2 atmosphere at 600^{0}C for 1.5 h. The maximum phenol adsorption capacity was obtained as 89.5 mg/g at the temperature of 600^{0}C at pH of 6.0.

Different agricultural wastes like bark, wooden and timber wastes have also been investigated as adsorbents. Some of the important ones have been briefly summarized here. The feasibility of pine wood based activated carbon for the removal of phenol and chlorophenol has been investigated by Tseng et al.[30] and the effect of micro porosity of the carbons on adsorption capacity was explored. Activated carbons prepared at different activation times of 0.5, 1.5, 2.7, and 4.0 h were studied. The carbons prepared at longer activation times had a larger adsorption capacity because the number of pores increases with increasing activation times. The adsorption capacities came to be 232.6 and 240.6 for phenol and 367.7 and 415.2 mg/g for 3 chlorophenol at activation time of 2.7 and 4hours respectively.

Activated carbon was prepared from pine bark by Bras et al.[31] and investigated for the adsorption of pentachlorophenol. Pine bark carbon surface acidity is helpful in decreasing the pH of the solutions, promoting the increase of the hydrophobic species in solution and adsorption capacity was 8.6 mg/g.

Pinus pinaster bark was converted into a carbonaceous adsorbent and used for the removal of phenol from aqueous solution in a packed bed by Alvarez et al.[32]. It has been found that the inlet phenol concentration as well as by the flow rate influences the adsorption rate. Both the breakthrough time and the dynamic capacity of the column decreased when the flow rate was increased, for the higher inlet concentration, due to an insufficient residence time of the phenol solution in the column. The dynamic capacity of the bed from 7.5 to 0.4 min was 0.038 to 0.15 mg phenol/g bark, respectively.

The effectiveness of biomass material KOH activated saw dust in adsorbing phenol from aqueous solutions was studied by B.H. Hameed and A. A. Rahman[33]. A decrease in the pH of solutions led to a significant increase in the adsorption capacity. The pseudo first order and pseudo second order kinetic models were used to analyze the adsorption data obtained and the maximum adsorption capacity of carbon was 149.25 mg/g.

Saw dust has been used as raw material in the preparation of powder activated carbon by chemical activation with zinc chloride for the sorption of phenol from dilute aqueous solutions by Mohanty et al.[34] . It was indicated that ZnCl2 plays an important role during carbonization on the surface properties of carbon. The adsorption capacity was 2.82 mg/g.

Mohanty et al.[35] explored Tamarindus indica wood as adsorbents for removal of phenol from dilute aqueous solutions in a multistage bubble column adsorber. The carbonization time and temperature are the two most important factors that affect the quality of zinc chloride activated carbon. The optimum operating parameters for the performance of the multistage bubble column adsorber were found as 160 min of contact time, 1 g/l of activated carbon loading, and 0.0263 m/s of superficial gas velocity. The maximum phenol removal was upto 80 mg/g.

The potential of jute stick carbon to remove phenol from aqueous solution was evaluated by Mustafa et al.[36]. It was justified that the extent of removal was dependent on concentration of phenol, contact time, pH, and quantity of adsorbent. The Freundlich isotherm was found to be efficient for the adsorption system and removal was 8 mg/g.

Rice is one of the major staple food grown throughout the world and rice industry produces several by-products which are investigated. Rice husk and rice husk char have been used as potential pollutants for removal of phenol by Ahmaruzzaman and Sharma[37] and Langmuir model fits the adsorption data better than the Freundlich isotherm. The adsoption capacity was found to be 4.5 and 7.9mg/g for rice husk and rice husk char respectively.

The potential of rice husk ash for the adsorption of phenol and chlorophenol was tested by Shiundu et al.[38] and an adsorption capacity of 14.382 and 0.1532 mg/g were obtained for phenol and chlorophenol respectively. The adsorption mechanism was understood by color formation reactions.

The suitability of different miscellaneous agricultural wastes like straw, fibres, bagasse, hulls, pomace and other parts have also been explored as locally available adsorbents.

Olive pomace, a commonly produced agricultural byproduct, was examined as adsorbent for the removal of phenols from olive-mill wastewater by Stasinakis et al.[39]. Three different forms of olive pomace, dried olive pomace (OP-1), dried and solvent extracted olive pomace (OP-2) and dried, solvent extracted and incompletely combusted olive pomace (OP-3) were used for preliminary studies. OP-1 and OP-2 could not be used as sorbent materials due to their trend to release of polyphenolic compounds. It was found that the adsorption capacity was 2.59 and 11.40 mg/g from kinetic and isotherm studies for OP-3. Phenol removal efficiency was improved by increasing sorbent concentration and decreasing particle size of the sorbent material.

Cimino et al.[40] used raw carbons made from olive cake as raw material in the preparation of activated carbon by chemical activation with H_2SO_4, HCl and HNO_3 for the sorption of phenol from wastewater and 20.68 mg/g of maximum adsorption capacity was achieved. It is found that acidic treatment changes the surface properties. It was found that these different behavior between the various carbons tested may be because of activation treatments so that different surface groups are formed.

Barley straw and its ash was selected from to prepare effective adsorbent for phenol removal from wastewater by Maleki et al.[41] .The adsorption capacity was strongly dependent on the pH of the solution, Freundlich isotherm best describes the adsorption data and barley straw ash was found to have more adsorption capacity than barley straw. Experiments gave the adsorption capacity of 0.067 and 0.0314mg/g for barley straw and barley straw ash respectively.

Shorea robusta leaf has been used as raw material in the preparation of powder activated carbon by chemical activation with zinc chloride for the sorption of phenol from dilute aqueous solutions by Mishra and Bhattacharya.[42] The maximum adsorption capacity was 0.216 mg/g in the range of pH 6-8. The adsorption data satisfies Freundlich isotherm.

Hazelnut Bagasse carbon activated with zinc chloride was examined for the removal of phenol from water by Karabacakoglu et al.[43]. The results showed that the removal efficiency increased with increase in adsorbent dose and phenol concentration. Freundlich and Langmuir adsorption isotherms were applied to the experimental data. The adsorption capacities of 97.36, 91.32 and 99.27 mg/g were found at 25, 35 and 45^{0} C respectively.

Coq et al.[44] investigated jute fiber as an adsorbent for the adsorption of phenol. Jute fibers were activated by both methods (physical by carbon dioxide and chemical by phosphoric acid). Due to the compositions of jute fiber which contains cellulose, hemicellulose and lignin, the activated jute fibers present a higher specific surface area higher than 900 m^2/g. Adsorption capacity of 181 mg/g was obtained. Some of the agricultural waste materials have been found to be prospective

Table 1: Comparision of adsorption capacities of adsorbents for phenol and its derivatives removal from waste water

Adsorbent	Solute	Adsorption capacity (mg/g)
Coconut shell	Phenol	205.8
Coconut shell	Phenol	206
Coconut shell	4- Chlorophenol	267
Coconut shell	4- Nitrophenol	251
Coconut husk	2,4,6-trichlorophenol	716.10
Coconut husk	2,4,6-trichlorophenol	191.73
Coir pith	Phenol	48.31
Coir pith	2,4 -dichlorophenol	19.12
Coir pith	p- nitrophenol	3.66
Almond shell	Phenol	70.4
Hazelnut shell	Phenol	100
Walnut shell	Phenol	145
Apricot stone	Phenol	126
Almond shell	Pentachlorophenol	9.6
Date stones	Phenol	90.4
Date pit	Refinery waste water	16.64
Date pit	Synthetic waste water	12.6
Black stone cherries	Phenol	133.33
Plum kernels	Phenol	257.4

Nutshells of *Sterculia alata*	Phenol	2.8
Corn cob	Phenol (KOH/char:2)	232
Corn cob	Phenol (KOH/char:6)	340
Corn cob	Chlorophenol (KOH/char:2)	362.8
Corn cob	CP (KOH/char:6)	485
Corn cob	2,4 - dichlorophenol (KOH/char:2)	333.7
Corn cob	DCP (KOH/char:6)	451.2
Banana peel	Phenol	689
Pomegranate peel	2,4-dichlorophenol	65.7
Palm seed coat	Phenol	18.3
Rubber seed coat	Phenolic solution	56
Rubber seed coat	live phenolic wastewater	36
Oil palm empty fruit bunch	2,4,6-trichlorophenol	168.89
Palm pith carbon	2,4- dichlorophenol	19.16
Oil palm empty fruit bunch	2,4,6-trichlorophenol	500
Oil palm empty fruit bunch	Phenol	4.868
Oil palm empty fruit bunch	Phenol (KOH 30 %)	90.09
Oil palm empty fruit bunch	(KOH 50 %)	91.74
Oil palm empty fruit bunch	(KOH 70 %)	89.29
Hemidesmus indicus roots	Phenol	16.10
Beet pulp	Phenol	89.5
Pine wood	Phenol (activation 2.7 hours)	232.6
Pine wood	(activation 4 hours)	240.6
Pine wood	3 chlorophenol (activation 2.7 hours)	367.7
Pine wood	(activation 4 hours)	415.2
Pine bark	Pentachlorophenol	8.6
Pinus pinaster bark	Phenol breakthrough time 7.5 min	0.038
Pinus pinaster bark	0.4 min	0.15
Saw dust (KOH)	Phenol	149.25
Saw dust ($ZnCl_2$)	Phenol	2.82
Tamarindus indica wood	Phenol	80
Jute stick	Phenol	8
Rice husk	Phenol	4.5
Rice husk char	Phenol	7.9
Rice husk char	phenol	14.382
Rice husk char	Chlorophenol	0.15322
Olive pomace	Phenol (kinetic studies)	2.59
Olive pomace	(isotherm studies)	11.40
Olive cake	Phenol	20.68
Barley straw	Phenol	0.067
Barley straw ash	Phenol	0.0314
Shorea robusta leaf	Phenol	0.216
Hazelnut Bagasse carbon	Phenol (25^0 C)	97.36
Hazelnut Bagasse carbon	35^0 C	91.32
Hazelnut Bagasse carbon	45^0 C	99.27
Jute fibre	Phenol	181

adsorbents for the removal of pollutants waste water. The prominent among them includes peanut hulls, grain sorghum, coffee bean husks, jackfruit peel, pistachio shell, cherry stones, and sugar beet.

RESULTS AND DISCUSSION

This paper reviewed the adsorption of phenol from wastewater locally available adsorbents which will promisingly replace conventional adsorbents. But the adsorbent has to be subjected to different treatment methods (physical and chemical) to increase the surface properties, adsorption capacity and life time of the adsorbent. This review indicates that these agricultural materials have equivalent or even more adsorption capacity to activated carbon. Comparing different adsorbent has high difficulty since different parameters are taken into consideration for evaluating the overall performance of the adsorbent[47]. Finally, if the adsorbent comes out with very good efficiency for the treatment of wastewater containing phenol and its compounds, then it can be successfully commercialized to industrial scale which will be beneficial to the society in future.

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