

Removal of Phenol From Aqueous Solution Using Tamarind Seed Powder As Adsorbent

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Abstract: Removal of phenol using tamarind seed powder (TSP) through adsorption process from aqueous solution was studied. Batch process and adsorption isotherm were carried out under varying experimental conditions of contact time, initial phenol concentration, temperature, adsorbent dose, particle size and pH. A series of batch experiments were carried out for various concentrations of phenol (1.0-10.0ppm) in a rotary shaker at agitation speed of 120 rpm. The single system adsorption process showed that the time required to reach equilibrium condition is about one hour under different initial concentrations. The adsorption of phenol increases with the increase of pH up to pH 8.0 and then starts to decrease. The suitability of the Freundlich and Langmuir adsorption models to the equilibrium data were investigated for each phenol-adsorbent system. Results show that, the equilibrium data for the phenol-tamarind seed powder systems fitted the Langmuir model ($R^2=0.972$) best within the concentration range studied. Experimental results also show that, about 72% phenol was removed. Adsorbed TSP could be regenerated by desorption with the help of 1.0 M NaOH solution and recovered up to 94.90% of adsorbed phenol. Results demonstrate that the tamarind seed an indigenous agricultural waste, can be used as an efficient adsorbent for the removal of phenol and phenolic compounds from water and/or waste water.

Keywords: Adsorption, phenol, tamarind seed powder (TSP).

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I. Introduction

The phenolic pollution is commonly observed in the chemical and pharmaceutical industries like petrochemical industries, petroleum refineries, coal gasification operations, liquefaction process, and resin manufacturing industries, dye synthesis units, pulp and paper mills and drug formulation industries. It is a highly corrosive and nerve poisoning agent. Harmful side effects, such as mouthsour, diarrhea, excretion of dark urine and impaired vision are seen on consumption of these [1]. These are considered as carcinogenic and dangerous even when they are present at trace levels in the environment [2]. Phenol has been designated as a priority pollutant by the US Environmental Protection Agency (EPA) and the National Pollutant Release Inventory (NPRI) of Canada. International regulatory bodies have set strict discharge limits for phenols for a sustainable environment [3]. Due to the high toxicity of phenols, they are subjected to specific regulations. The Environmental Protection Agency (EPA) calls for lowering phenol content in potable and mineral waters to 0.5 ppb, while the limits for wastewater discharge are 0.5 ppm for surface waters and 1 ppm for the sewerage system. Removal of phenolics from waste water before discharging it to another point or place has become a challenge for the chemist of the world. There are different methods for the separation of phenols such as steam distillation, separation by extraction, separation by adsorption, separation by membrane, destruction of phenol by wet-air oxidation, electrochemical oxidation, biochemical abatement [4]. Various methods such as liquid-liquid extraction, three phase liquid system, pervaporation, nano-filtration, adsorption, cloud point extraction method, three phase electrode system, electrocoagulation, photodecomposition, enzymatic degradation using polyphenol oxidases and peroxidases and microbial degradation were applied for the removal of phenol from water [5]. Polymerization method shows 90% phenol removal efficiency. Photo reactor using titanium dioxide nanoparticles showed phenol removal efficiency of 90% [6] and solar photo catalytic reactor using FeSO_4 as a catalyst enhanced 20-30% phenol degradation [7]. Chitosan and chitosan impregnated granular activated carbon were applied for the removal of phenol from aqueous solution and found the efficiency were 78.4% and 90.49% respectively [8]. Sewage Sludge Based Adsorbent was used for the removal of phenol but it was reported that the process is highly influenced by the initial phenol concentration and phenol specification which is directly related to solution's pH [9].

Adsorption is considered as one of the appropriate techniques for removal of phenolics from water because the technique is easy to design and operate. The technique produces no toxic wastes. The spent sorbent

can serve as a source fuel to produce power[10]. Adsorption of phenol from aqueous solution was investigated using sodium zeolite[11], pomegranate peel ash [12], solidified landfilled sewage sludge [13], rice straw (untreated, physically treated and thermally treated)[14], banana leaf ash [15] and column packed with activated carbon[16] as an adsorbent. The results indicate that, adsorption capacity of the adsorbent was considerably affected by initial phenol concentration, pH, contact time, and adsorbent dosage. Many problems associated with the above mentioned methods have been reported in the literature such as high cost, low efficiency and generation of toxic products. Some of them are sophisticated and complicated. In this study an agricultural waste and very low cost adsorbent tamarind seed powder has been applied to remove phenol from aqueous solution. The main aim of this study is to explore the possibility using tamarind seed powder in removing phenol from aqueous solution by adsorption.

II. Materials And Methods

2.1 Collection of the tamarind seeds

Tamarind seeds were collected from Sahib Bazaar, the central city market of Rajshahi city in Bangladesh. The dimension of the seeds was approximately $3 \times 2.5 \times 2$ cm³.

2.2 Preparation of tamarind seeds

After collection, the tamarind seeds were treated to make them ready for use. The seeds were washed and cleaned. They were first sun dried, then oven dried at 80°C and ground to powder. The particles of 30, 45 and 50 mesh were separated by sieving through standard test sieve. The powders were stored in three different stoppered reagent bottles.

2.3 Reagents and solution

All reagents utilized were AR grade. All plastic and glassware utilized were pre-washed with detergent water solution and washed with tap water and rinsed with double distilled water. All the solutions were prepared in double distilled water. The stock standard solution of 1000 ppm was prepared by dissolving 1.00 g of phenol in 1000 ml of double distilled water. The solution should be stable for at least 30 days and kept in a refrigerator. The working standard solutions of 20 ppm were prepared by appropriate dilution of the stock standard solution with double distilled water and used within 1 day.

The potassium ferricyanide solution (8% w/v) was prepared by dissolving 8.00 g of potassium ferricyanide in double distilled water in a 100 mL volumetric flask. 4-aminoantipyrine solution (2% w/v) was prepared by dissolving 0.20 g of it in 10 mL double distilled water. For the preparation of buffer solution, 3.0905 g of boric acid was dissolved in 500 mL and 2.00 g of NaOH was dissolved in another 500 mL double distilled water in two separate beakers. The probe of a calibrated pH meter was inserted into boric acid solution and NaOH solution was added gradually to it to adjust pH at 10.0.

2.4 Instruments/Equipment

Computer interfaced UV-Visible spectrophotometer (PG instruments, Model: T-60, UK) were used. Solutions were applied in a 3.0 mL quartz cell. Crusher, shaker and standard test sieves (HMK-BS1-200 standard test sieve, China) were used in the work.

2.5 Preparation of a standard calibration curve

A standard calibration curve was prepared for various phenol concentration (0.5, 1.0, 2.0, 3.0, 4.0, 5.0, 6.0 and 8.0 ppm). The solutions were prepared taking different aliquots of above standard 20 ppm phenol solution with the addition of 0.5 mL potassium ferricyanide solution, 5 mL buffer solution (pH=10), 0.4 mL 4-aminoantipyrine solution and double distilled water to make 25 mL final solution. Then absorbance was taken after 40 minutes at 500 nm and the results were used to plot a standard curve[17].

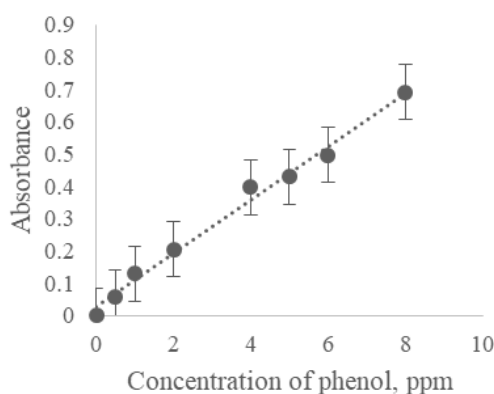


Fig 1: Standard calibration curve

For the removal of phenol from aqueous solution, a particular amount of tamarind seed powder was added to the solution and it was shaken for a particular period. After that (after adsorption) the phenol content of the both solutions (before and after adsorption) was measured against its absorbance. The effects of various parameters such as pH, contact time, initial phenol concentration, particle size and dose of adsorbent were studied. Batch experiments were also carried out for adsorption isotherm and desorption of phenol from tamarind seed powder.

III. Results And Discussion

Adsorption of phenol on tamarind seed powder was studied in aqueous solution optimizing various physicochemical parameters such as pH effect, contact time, temperature, initial concentration, particle size and dose of adsorbent.

3.1 Effect of contact time

Time is a very important parameter for the adsorption of phenol on adsorbent. The experiment was carried out at 30°C, particle size of 50 mesh, 2.5 g of adsorbent in each 25 ml of phenol solution of initial concentration 5 ppm.

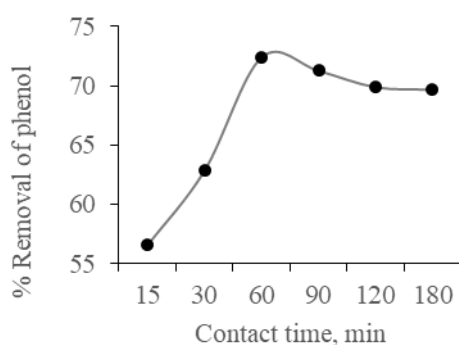


Fig 2: Effect of contact time on removal of phenol

The percent of removal of phenol increases with contact time until equilibrium. The result shows that the removal of phenol occurs quickly. The plot represents a continuous adsorption leading to saturation suggesting possible monolayer coverage of phenol on tamarind seed powder surface. It takes around 60 min for maximum (72.4%) adsorption.

3.2 Effect of initial pH

The pH of aqueous solution is an important parameter in the adsorption process. The removal of phenol from waste water is highly dependent on pH of the solution. pH affects the surface charge of the adsorbent and degree of ionization and specification of the adsorbed species[18].

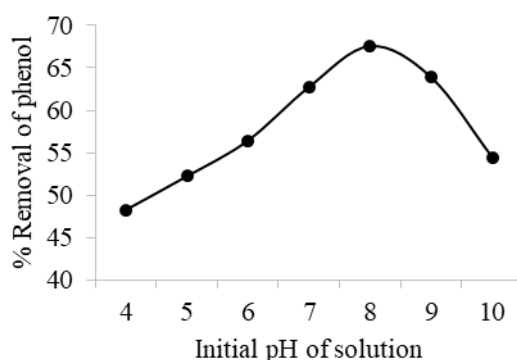


Fig 3: Effect of initial pH on removal of phenol

The fig 3 implies that adsorption of phenol increases with pH value from pH 4.0 to 8.0 and decreases thereafter. Thus optimum pH here is 8.0. This behavior can be explained by considering the nature of the adsorbent at different pH in phenol adsorption. Adsorption of phenol upto pH 8.0 suggests that the negatively charged phenolate ions bind to the positively charged functional groups of the surface of TSP and as of activated

carbon. At this pH, positive functional groups are exposed on adsorbent. But at pH above 8.0 it reverses. This occurs based on the relationship[19]:

$$P_o = P_t / [1 + 10^{(pK_a - pH)}]$$

Where, P_o is the concentration of ionized phenol species, P_t is the concentration of phenol taken, pH is the equilibrium pH and pK_a is 10.0. Phenol-TSP adsorption system shows similar results as phenol-activated carbon system in this respect. Similar results have been reported in the literature during adsorption of phenol on coir pith as adsorbent[20] and rice straw as adsorbents[14].

3.3 Effect of adsorbent dose

Different doses of adsorbent were introduced in different solutions of phenol of same volume and same initial concentration. The removal of phenol was found as follows.

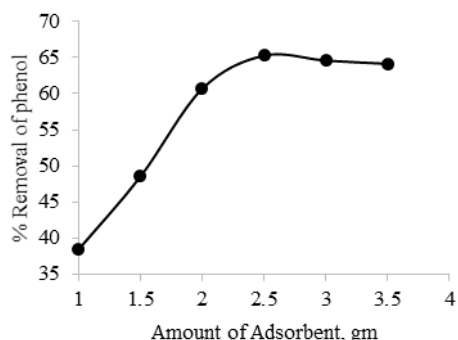


Fig 4: Effect of adsorbent dose variation on removal of phenol

The amount of adsorption increases with the dose of adsorbent and after a certain dose (2.5 g) of adsorbent, adsorption does not increase. The increase of adsorption seems that, there are still active sites on the surface of the adsorbent that are unsaturated [12]. If the dose is more than the optimum amount (2.5 g) in the process unit, stoichiometrically, the dose is more than the equivalent amount of phenol in the solution. For this reason, adsorption does not increase with the increase of adsorbent dose.

3.4 Effect of particle size

Same amount of adsorbent of different particle sizes were introduced in different solutions of phenol of same volume and same initial concentration. The removal of phenol was measured after same time. The result was as follows.

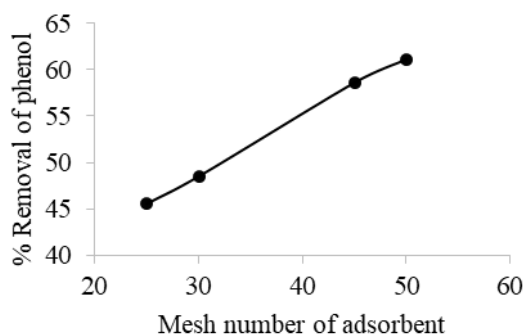


Fig 5: Effect of particle size of adsorbent on removal of phenol

Particle size adsorbent is very important for the adsorption. Adsorption of phenol is increased with decrease of particle size of adsorbent. This is due to the increase of surface area of the adsorbent which increases more available active sites.

3.5 Effect of initial concentration of phenol

Same amount of tamarind seed powder of same particle size were introduced in phenol solutions of different initial concentrations. The removal of phenol was measured after same time.

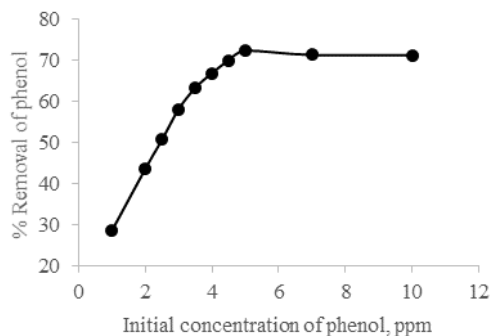


Fig 6: Effect of Initial concentration of phenol on removal of phenol

Adsorption of phenol increases with increase in initial concentration of phenol and attains a plateau after a certain initial concentration. This is caused by the increase of mass transfer driving force and therefore the rate at which phenol molecules pass from the solution to the particle surface. In this study, the maximum 72.0% of phenol was removed at initial concentration of 5.0 ppm.

3.6 Effect of temperature

Temperature has a great effect on adsorption process. The effect of temperature on adsorption of phenol on tamarind seed powder was investigated by varying temperature from 30 to 50 °C.

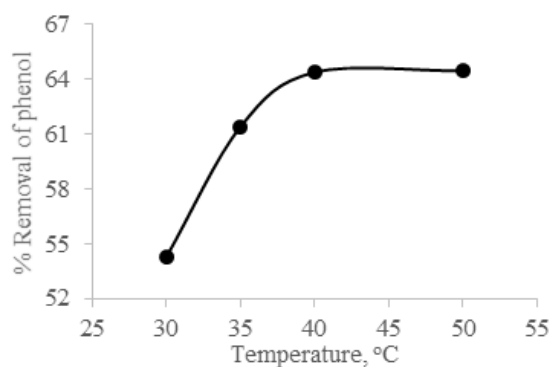


Fig. 7: Effect of temperature on removal of phenol

From Fig. 7, it was observed that, adsorption favors at above room temperature. This indicates that the adsorption of phenol on tamarind seed powder is an endothermic process. This might be due to the increase in the number of pores on the adsorbent surface. The high temperature reduces the thickness of outer surface of the adsorbent and increase the kinetic energy of phenol molecules. As a result phenol molecules are easily adsorbed on the adsorbent surface [18][21].

3.7 Adsorption isotherm

In this work, the adsorption isotherm was also studied. The Freundlich and Langmuir isotherm were studied in the way used by various authors [22, 23, 24] which provide the basis for the design of adsorption systems. The Langmuir and Freundlich models are the most widely used isotherm equation.

$$\frac{1}{q_e} = \left(\frac{1}{bq_m}\right) \frac{1}{C_e} + \frac{1}{q_m} \dots\dots\dots (1) \quad \text{Langmuir adsorption}$$

Where, q_e is the amount of suspended solids adsorbed in mg/g, C_e is the equilibrium concentration (mg/L), b is the adsorption equilibrium constant (L/mg) which is related to the energy of adsorption [25], and q_m (mg/g) is the maximum adsorption capacity.

Freundlich isotherm model is mathematically expressed as shown in equations (2) and (3), respectively [26, 27, 28]:

$$q_e = K_f \cdot C_e^{1/n} \dots\dots\dots (2) \quad \text{non-linear form Freundlich isotherm}$$

$$\log q_e = \log K_f + 1/n \log C_e \dots\dots\dots (3) \quad \text{linearized form Freundlich isotherm}$$

The regression values (R^2) clearly show that adsorption process fitted better with the Langmuir isotherm model with value of $R^2 = 0.9719$ as compared to the Freundlich isotherm model with R^2 value of 0.8935. The best fit of equilibrium data in the Langmuir isotherm expression predicted the monolayer coverage

of phenol onto TSP. The maximum adsorption capacity of TSP was found to be 24.153 mg/L whereas that of sodium zeolite was found to be 13.051 mg/L[11].

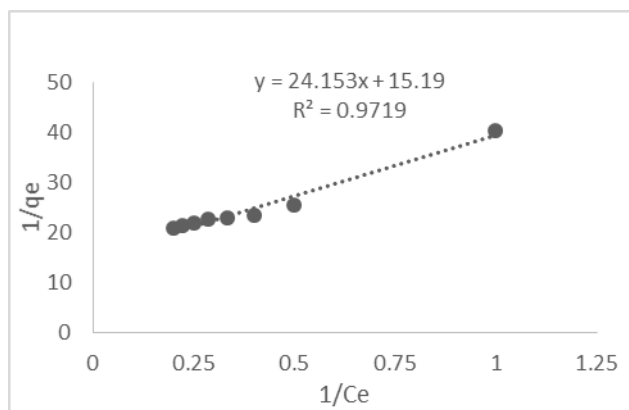
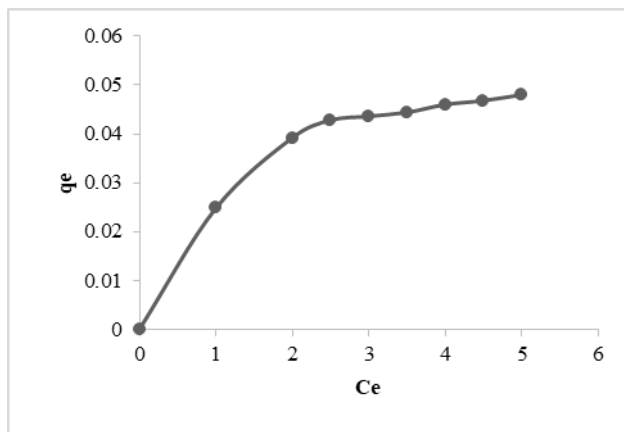


Fig 8: Langmuir isotherm

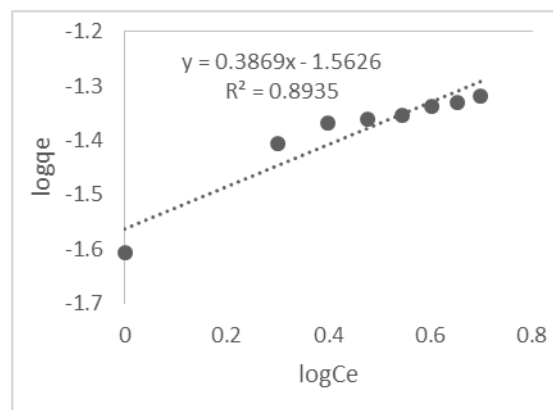


Fig 9: Freundlich isotherm

3.8 Regeneration of the adsorbent/Desorption

Regeneration of adsorbent is economically very important in industrial waste water treatment. Experiments were conducted for regenerating TSP using various desorbing agents (such as HCl, H₂SO₄, NaOH and KOH solutions). The percent of recovery of phenol using these reagents are shown in figure 10. In this case, the solution of NaOH showed the maximum recovery of phenol (94.90%) from the TSP. After regeneration, the TSP was washed with distilled water, dried in oven and reused in subsequent operations. The regenerated TSP lost 9.70% of its initial adsorption efficiency. It makes the operation more economical reducing the total cost.

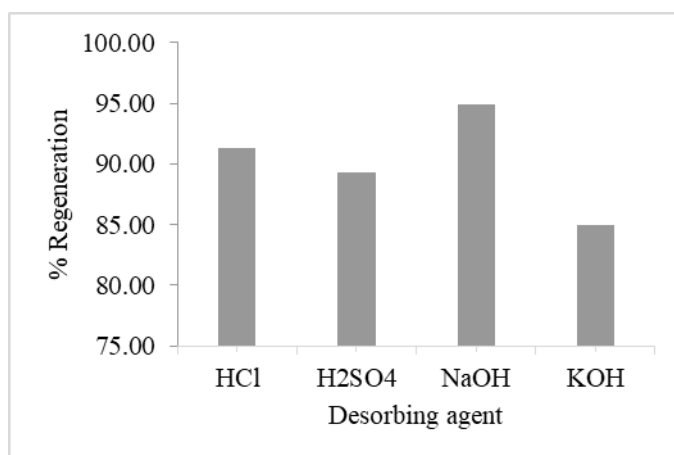


Figure 10: Percent of phenol recovered using various desorbing agents

IV. Conclusion

In the present work, TSP (Tamarind Seed Powder) was applied for the removal of phenol from waste water. The conclusion of this work can be pointed as follows.

- The TSP can be an effective adsorbent for the removal of phenol(s) from aqueous solutions.
- In the context of the present environmental situation, use of TSP, a new pathway might be opened in the whole range of contaminant removal from waste water.
- TSP as an adsorbent can be compared with well-recognized adsorbent activated carbon as well as others in economical respect.
- This adsorbent can be regenerated and reused.
- Moreover, TSP is an agricultural as well as industrial waste that has been directed as a wealth in the treatment of waste water.

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