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Research Article

Treatment of phenolic wastewaters by mass transfer using crab eye surface modified adsorbent

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Abstract

In this study, the abatement of phenol from water was determined by processed crab eye surface modified adsorbent (Indian plant: *Abrus precatorios*) as a source material. Removal of phenol from surface modified adsorbent was operated in intermittent process. Reduction of phenol depends upon different operational conditions such as pH of the solution, adsorbent load, concentration of phenol and period interaction for sorption. It was observed that maximum pH 8 and dilution 0.45 g/L were favourable for the removal of phenol from water. Application of kinetics studies showed that pseudo-second order fitted excellently with the data of phenol removal. Data trend shows removal of phenol was fitted best with Langmuir isotherm model as compared to Freundlich model. The change in enthalpy revealed phenol adsorption by the crab eye activated carbon was an exothermic process.

Keywords: Crab eye activated carbon; Adsorption; Isothermal parameters; kinetics; mass transfer.

1. Introduction

Credit of phenols in water system is received from many sources (Ahmaruzzaman, 2008). Phenol is catalogued serious pollutants due to their obnoxious nature (Kilic et al., 2011). Phenol discharge into water body have adversely affect the environment and ecology (Radhika and Palanivelu, 2006). They have bio-accumulative and toxic effect causing irritation, liver disorders, damage to central nervous system and can lead to death in humans, also they mutations and cancer (Rawat et al., 2021). Regulating and controlling act of international organization set the discharging limit of phenol in wastewater bodies (WHO, 1996), physical, chemical and biochemical treatment of contaminated water (Kumar et al., 2011). Deterioration of wastewater by various pollutants can be reduced by the use of surface modified adsorbents. This technique is still considered as alternative to the conventional wastewater treatment technologies due to several reasons (Mohanty et al., 2005; Wang et al., 2007; Lin and Juang, 2009). A number of adsorbents were derived from the wastes biomasses have been reported for the removal of phenol from water such as activated carbon prepared using aerial roots of banyan tree (Nirmala et al., 2019), guava tree bark (Mandal et al., 2020), pod shells of *Acacia tortilis* (Malakootian et al., 2018), cork bio-adsorbent (Mallek et al., 2018), powder of pine cones (Kumar et al., 2018) and palm oil shells biosorbent (Sahu et al., 2021). There are so many lacunae to use surface modified material as adsorbents (Ahmaruzzaman, 2008; Babel and Kurniawan, 2003; Kalderis et al., 2008; Tan et al., 2009; Streat et al., 1995).

The surface modifications of adsorbents have been done by different methods including modification using different chemicals like iron chloride (Cui et al., 2019; Verma and Singh, 2022), zirconium oxychloride (Bhan and Singh, 2022), potassium hydroxide (Sriramoju et al., 2020), pyromellitic dianhydride (Yu et al., 2010) and HNO_3 (Ranasinghe et al., 2018). Crab eye surface modified adsorbent can be used in the treatment of wastewater and agricultural remnant, industrial waste, sludge, slag and other residues were used as economically convenient material (Mubarik et al., 2012).

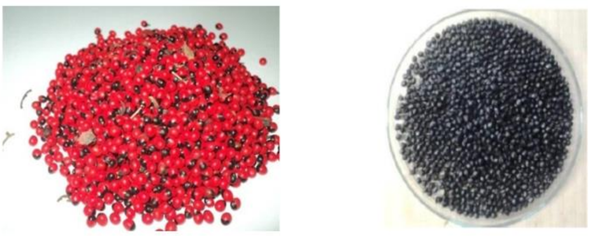
1. Materials and methods

2.1 Preparation of surface modified adsorbent from crab eye seeds

After processing of crab eye modified adsorbent bigger attention on high uptake capacity, cost, carbon content and utility are basic criteria (Rodriguez-Reinoso, 1997). Raw material was collected from National Institute of Technology Rourkela, India. Before treating crab eye washing was done in water and then crab eye was charred (Fig. 1b) at 623 K for 120 min. Amending the properties of crab eye surface modified adsorbent was retreated with sodium hydroxide charred. Crab eye was cleaned with double distilled water dried in oven at 105 ^oC for 10 h, after that it was preserve. Modified activated charred was named as crab eye surface modified adsorbent (CESMA). The characterization of crab eye surface modified adsorbent was done using standard methods (ISI. 1989; Eaton et al., 2005; Vogel, 1969).

2.2 Preparing of stock solutions

The standard stock solution was prepared by dissolving 1g of phenol in 1L distilled water. Required dilutions were done from standard stock solution. Further study was performed with the prepared standard stock solution.



(a) Raw crab eye (b) Charred carb eye

Fig. 1. Photograph of (a) raw crab eye and (b) charred crab eye

2.3 Surface characterization of crab eye activated adsorbent

The surface of materials remains neutral at particular pH (pH_{PZC}), and it possess negative surface charge, when pH of the surrounding solution is more than the value of pH_{PZC}; however, possess positive surface charge at pH less than the value of pH_{PZC} (Liu et al., 2012; Dabrowski et al., 2005; Yanhui et al., 2012). The pH_{PZC} of crab eye surface modified adsorbent (6.30) was determined as per the method presented by Saeed et al. (2009). Acidic and basic surface characterizing was done by Boehm titration method (Boehm, 2002).

2.4 Adsorption studies

Uptake capacity of phenol was determined by the Eq. 1.

$$q_{Ph} = \frac{\%R \times C_0}{100 \times m} \qquad (1)$$

C_0 represents the dilution factor of phenol, m represents the mass of crab eye activated surface modified material and R represents the removal percentage of phenol, it was determined between time intervals of 25 to 350 min surface modified crab eye was utilized from 0.20 to 6.1 g/L for the determination of optimum concentration of the adsorbent required. The pH study for phenol adsorption was carried out in the range of 2 to 12. The particular pH is set by adding hydrogen chloride and sodium hydroxide. Thermal effect of reduction of phenol was done in the range of 308-338K.

2. Results and discussion

3.1 Effect of Crab Eye surface modified adsorbent dosage

To find out the maximum dose of crab eye surface modified adsorbent, the dosage was taken 0.20 to 6.0 g/L and concentration of phenol was 100 mg/L (Fig. 2). The adsorption capacity was observed decreasing as the quantity of adsorbing materials was increased. The point where two curves cutting each other was determined as the optimum dose, maximum removal was found at highest dose 0.6 g/l. Above meeting point, there is a decreasing trend in removal of phenol due to limited availability of binding site. Surface modified adsorbent processed from various raw materials reported in other studies (Kilic et al., 2011; Tan et al., 2009; Dutta et al., 2001).

3.2 Effect of operating pH

The pH is an important parameter and plays paramount role in the eradication of pollutants from the wastewater. Removal of phenol by **surface modified adsorbent (**0.5 g/L) was examined from 2 to 12 range of pH as shown in Fig. 3. The removal of phenol at pH ≤ 8 was maximum and became constant. An increase in pH > 8, removal of phenol was decreased significantly.

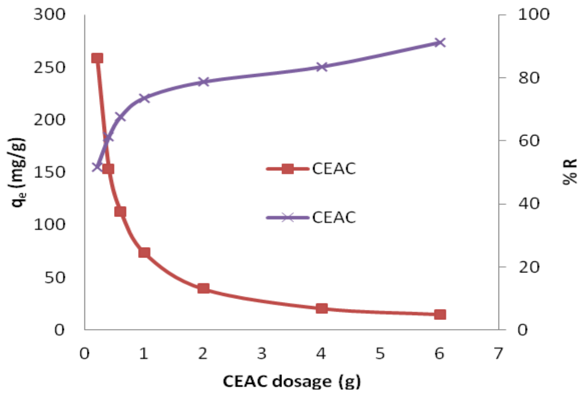


Fig. 2. Differing amount of crab eye surface modified adsorbent in for phenol adsorption (initial phenol concentration=100 mg/L, operating pH=6, operating temperature=308 K, contact time=6.50 h)

The decrease in removal of phenol with an increasing pH is dependent of the pH_{ZPC} of the adsorbent. The modified crab eye adsorbent surface becomes negatively charged at higher pH. Interaction of negative surface and ionic phenol molecule adsorption of phenol got reduced (Nadavala et al., 2009; Kumar and Min, 2011; Altenor et al., 2009). Hence, pH 8 was found ideal pH to removal of phenol using surface modified crab eye adsorbent**.**

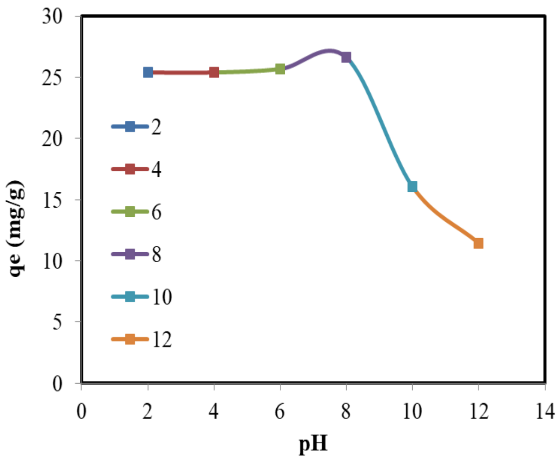


Fig. 3. Effect of operating pH on the phenol uptake capacity and its equilibrium (initial phenol concentration=100 mg/L, adsorbent dose=0.5 g/L, operating temperature=308 K, contact time=5.30 h)

3.2 Effect of interaction time

To ascertain interaction time for maximum uptake capacity onto crab eye surface modified adsorbent. Experiments were run at different time intervals from 30 min to 360 min at pH 8 using 0.5 g/L adsorbent concentration and 100 mg/L concentration of phenol (Fig. 4). Uptake capacity of crab eye surface modified adsorbent was initially very zips, it was observed that ~30 mg/g removal occurred in the initial 30 min. After that, removal rate slowed down. This is occurs due to the covering of remaining surface sites by phenol molecules. The equilibrium point was obtained at 120 min resulting in removal of phenol ~42 mg/g, which denotes the available surface sites saturation. Therefore, 120 min was observed to be the ideal contact time to remove of phenol crab eye surface modified adsorbent.

3.4 Diffusion Study

The removal rate can be limited by pollutant diffusion through pores or surface or by the diffusion through external liquid boundary layer i.e., film diffusion.

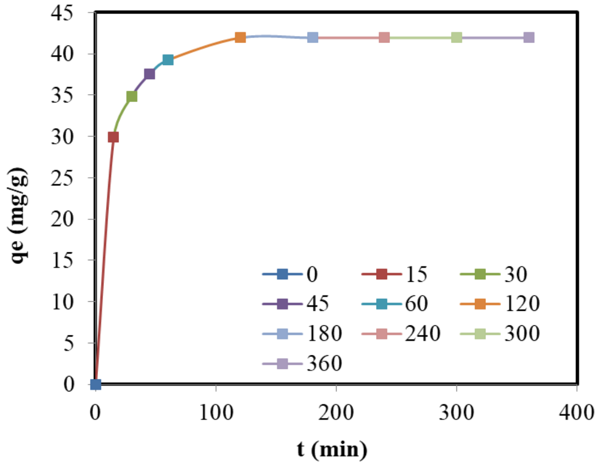


Fig. 4. Interaction and reduction of phenol onto CESMA (initial phenol concentration=100 mg/L, adsorbent dose=0.9 g/L, operating temperature=308 K, pH=8)

The kinetic data of phenol adsorption attained in the this study was analyzed by application of model proposed by Boyd et al. (1947)

$$F(t) = 1 - \left(6/\pi^2\right)\exp(-B_t) \quad (2)$$

$$\text{and } F(t) = \frac{q_t}{q_e} \quad (3)$$

q_t and q_e are the uptake capacity (mg/g)at different time intervals and at equilibrium time. F(t) represents the fractional amount of adsorbed phenol and Bt represents the F(t) function.

Substitution of eq. 1 in eq. 2, results in a simplified equation Eq (4):

$$B_t = -0.4977 - \ln(1-F) \quad (4)$$

As the Fig. 6 shows the straight lines of the linear fits in the graph plotted between B_t and t do not pass through origin which indicates that removal of phenol using CESMA was primarily occurred by the process of mass transfer where particle diffusion was rate limiting step (Kalavathy et al., 2005).

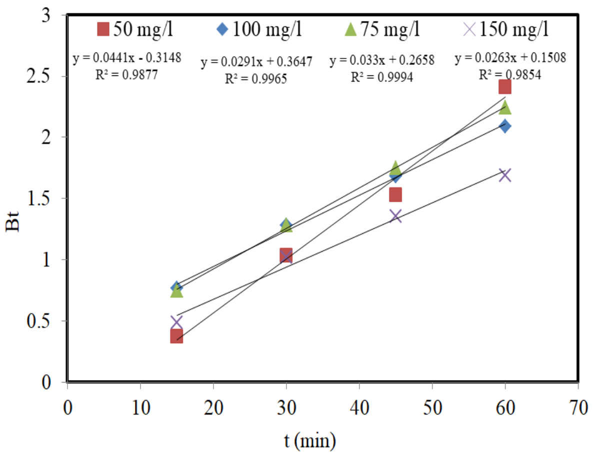


Fig. 5. Boyd graph for the removal of phenol by CESMA (initial phenol concentration=60-160 mg/L, adsorbent dose=0.5 g/L, operating temperature=308 K, contact time=2 h, pH=8)

The values of Bt were determined the diffusivity, De (cm^2/s) using Kumar et al. (2006):

$$B = \frac{\pi^2 D_e}{r^2} \quad (5)$$

The particle radius was calculated through sieve analysis. The coefficient of diffusion D_e was calculated by Eq. (4) for the phenol concentration from 60 to 160 mg/L. The average D_e value was observed to be 5.84 x 10^{-13} m^2/s for the adsorption of phenol onto crab eye surface modified adsorbent.

Following equation are used to determine the mass transfer coefficient:

$$\ln\left[\frac{C_t}{C_o} - \frac{1}{1+pK}\right] = \ln\left[\frac{pK}{1+pK}\right] - \left[\frac{1+pK}{pK}\right] K_1.Sv.t \quad (6)$$

$$Sv = \frac{6w}{d_p \rho_p (1-\varepsilon_p)} \quad (7)$$

Where, the initial concentration of phenol (mg/L) and phenol concentration after different time intervals are represented by C_0 and C_t respectively, p (g/L) corresponds to the mass removal K (L/g) constant for Langmuir isotherm, β_1 (Cm/s) corresponds to the coefficient of mass transfer and Sv (L/cm) corresponds to the sites available on surface of adsorbent per unit volume. Tending to zero the mass transfer is controlling. A plot of $\ln[(C_t/C_o) - (1/(1+mK))]$ versus t results in a straight line (Fig.7) gives the plot for mass transfer.

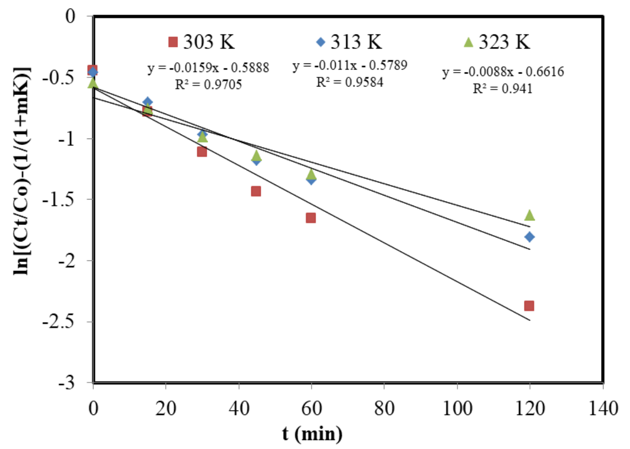


Fig. 6. Mass transfer graph, reducing phenol onto CESMA (initial phenol concentration=100 mg/L, adsorbent dose=0.5 g/L, contact time=2 h, pH=8)

β_1 values have been find out through various intercept line at C_0=100 mg/L, consequently values at various temperature 308, 318, 328 K were found to be 1.02×10^{-7}, 483×10^{-7}, 0.316×10^{-7} cm/s. Phenol transfer from liquid to surface modified adsorbent is appreciable . The coefficient of mass transfer has been reported (Mondal et al., 2010).

3.5 Equilibrium studies of phenol removal onto crab eye surface modified adsorbent

Freundlich, Langmuir, Redlich-Peterson and Tempkin isotherms goodness were tested (shown in Fig. 7) with varying temperature of 313-343K. The non-linear regression coefficient for

Langmuir isotherm was found highest among all the isotherms. After calculation of *n* value it was found that it is less than one. Prove the fitness of Langmuir isotherm.

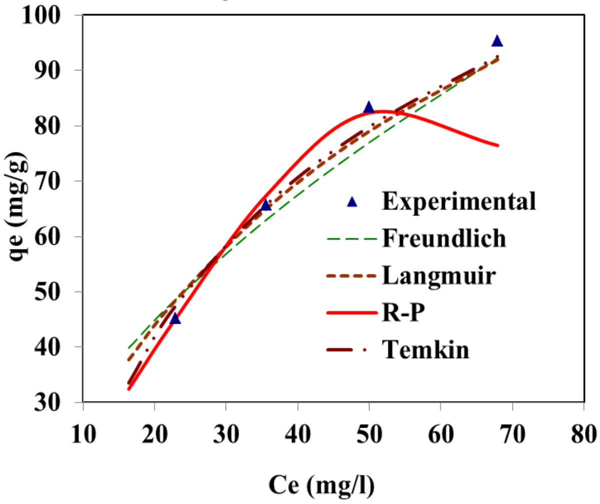


Fig. 7. Equilibrium isotherms plot for phenol removal using CESMA (initial phenol concentration=60-160 mg/L, adsorbent dose=0.45 g/L, operating temperature=308 K, contact time=2 h, pH=8)

3.6 Calculation of Thermodynamic Properties

Thermodynamics properties (ΔH^o), (ΔG^o), and entropy (ΔS^o) were calculated (Table 1) for the removal of phenol onto crab eye surface modified adsorbent by using standard equations (Parfitt and Rochester, 1983; Jain et al., 2004). The values of ΔG^o negative at varying temperatures indicating removal are favorable (Jain et al., 2004; Roig et al., 1993). Enthalpy (ΔH^o) shows that reduction of phenol is exothermic in nature (Longhinotti et al., 1998).

Table 1 Thermodynamic properties of phenol removal onto crab eye surface modified adsorbent (C_0=60-160mg/l, t=2h, m= 0.5g/l PH_0=8)

C_o (mgLl)	ΔH^o (kJ/Mol)	ΔS^o (kJ/Mol.K)	ΔG^o (kJ/Mol K)			
			308 K	318 K	328 K	338 K
50	-8.78	32.27	-18.56	-18.85	-21.13	-22.33
75	-12.37	18.65	-18.04	-18.13	-18.58	-18.50
100	-14.75	19.33	-17.05	-17.23	-18.47	-18.70
125	-12.83	21.11	-19..48	-19.79	-18.21	-17.20
150	-8.29	29.45	-17.84	-19.28	-17.44	-18.79

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