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Simultaneous treatment of phenol and cyanide containing aqueous solution by adsorption, biotreatment and simultaneous adsorption and biotreatment (SAB) process

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ABSTRACT

This study presents a comparative study of adsorption and biotreatment process alone with simultaneous adsorption and biotreatment (SAB) process. Adsorption studies were conducted with coconut husk activated carbon (CHAC), as adsorbent. SAB and biodegradation studies were carried out with immobilized and suspended cultures of *Serratia odorifera* MTCC 5700, respectively. Phenol and cyanide solution were used for all studies with initial concentration of 300 mg/L and 30 mg/L, respectively, initial pH of 8 and temperature of 30 °C. The optimum adsorbent dose was found 30 g/L for adsorption process and SAB process. In SAB process, the removal efficiency was found more than adsorption and biodegradation process and achieved removal efficiency of 89.88% for phenol and 93.06% for cyanide. Three mono components and six multicomponent equilibrium isotherm models were considered. Non-linear regression technique was used for estimation of model parameters. Among tested multicomponent isotherm models Extended Freundlich model found to fit well with the experimental data. The kinetics of microbial growth and biotreatment of phenol and cyanide were also investigated. Haldane's and Monod model have been used to interpret experimental data on phenol and cyanide biotreatment. It was established that simultaneous adsorption and biotreatment process is more effective than biotreatment and adsorption process alone.

1. Introduction

The elimination of pollutants, from wastewater has attracted great consideration because of their antagonistic effects on human health and environmental. The most significant pollution sources comprising phenol, cyanide and their compounds are the effluent generated from the coke, iron–steel, mining, petroleum refining, paint, pesticide, solvent, electroplating, wood preserving chemicals, pharmaceuticals, and explosives manufacturing industries [1–8]. Cyanide can cause thyroid effect, skin irritation, and breathing problem and phenol affects liver kidneys, vascular system and convulsions etc [2,4]. Phenol and cyanide comprising wastewater cannot be discharged without eliminating these toxic elements. Environmental protection agencies of many countries have proposed an acceptable limit for phenol and cyanide in effluent. US environmental protection agency (US EPA) and minimal national standard (MINAS) of central pollution control board (CPCB) limit for cyanide in effluent as 0.2 mg/L and for phenol as 0.5 mg/L [2,9]. Therefore, it is necessary the removal/recovery of both compounds for letting down the concentrations to below under regulatory limits.

Various treatment technologies have been proposed for elimination of phenol and cyanide. These methods include such as adsorption, ion exchange, leaching, oxidization, precipitation, biodegradation, membrane separation incineration, and solvent extraction etc [10,11]. However, the limitations of these methods are high maintenance and operational costs, complicated process involved in the treatment and generation of poisonous sludge during operation. Relatively, adsorption process is found to be a better alternative in wastewater treatment because of simplicity of design and ease of operation [12–16]. A large variety of adsorbents have been examined for their capability to eliminate various types of pollutants from wastewater [1,2,12–17]. Mostly granular activated carbon is used as adsorbent for wastewater treatment due to its great adsorption capacity for removal of phenol and cyanide [1,4,17]. However, because of high cost, there has been an increasing interest in utilizing natural occurring adsorbent for removal of pollutants from wastewater. The interest is increasing

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Nomenclature

q_e	Specific uptake of adsorbent at equilibrium (mg/g)
Q_o	Langmuir model constant (mg/g)
b	Langmuir model constant
$Q_{e,i}$	Amount of ith component adsorbed per gram of adsorbent at equilibrium (mg/g)
$Q_{o,i}$	Modified Langmuir model constant for *i*th component (mg/g)
$C_{e,i}$	Concentration of ith component in the binary mixture at equilibrium (mg/L)
K_F	Freundlich model constant (mg/g)
n	Freundlich model constant
$K_{F,i}$	Extended Freundlich model constant (mg/g)
K_{RP}	Redlich–Peterson model constant (L/g)
a_{RP}	Redlich–Peterson model constant (L/mg)
β	Redlich–Peterson model constant
$\eta_{RP,i}$	Multicomponent Redlich–Peterson model constant (L/g)
$a_{RP,i}$	Multicomponent Redlich–Peterson model constant (L/mg)
β_j	Multicomponent Redlich–Peterson model constant
x_i, y_i, z_i	Constant in modified Redlich–Peterson model
MPSD	Marquardt's percent standard deviation
N	Number of observations in the experimental Isotherm
P	Number of parameter in regression model
$Q_{e,i}^{exp}$	Experimental value of Q_e (mg/g)
$Q_{e,i}^{cal}$	Predicted value of Q_e (mg/g)

in phenol and cyanide removal by adsorbents due to their easy availability and lower cost. Numerous agricultural solid waste have been used to develop activated carbons such as bamboo, rice husk, oil palm shell, coconut shell, almond shell and coir pith etc. [18–21]. Coconut husk is a low cost, abundant natural material, easily available for production of activated carbon. However, some studies are available in the literature on the application of coconut husk activated carbon (CHAC) for the treatment of pollutants [19,21–23].

Now a day biotreatment technique is economical and effective due to few byproduct production. A number of microorganisms are capable of degrading phenol and cyanide. Phenol and cyanide are utilized by microorganism as the sole source of carbon, energy and nitrogen. Cyanide is degraded by strains such as *Pseudomonas*, *Acinetobacter* and *Alcaligenes Bacillus*, and phenol is degraded by including *Pseudomonas* spp., *Alcaligenes* spp., *Spreptomyces setonii*, *Bacillus stearothermophilus*, and *Azotobacter* spp. [24–32]. Some fungi are also able to utilize the phenol and cyanide such as *Fusarium solani*, *Gloeocercospora sorghi*, *Trichosporon cutaneum*, *Candida tropicals*, *Fusarium* spp., *Aspergillus* spp., and *Graphium* spp. etc. [33–37].

Adsorption and biotreatment techniques are two methods mostly used for treatment of phenol and cyanide containing wastewater. Both techniques either used separately or combined for high removal performance of a process. The microorganisms immobilize on adsorbent surface enhance the removal efficiency of the process. This is owing to the biofilm creation on the surface of adsorbent where both process adsorption and biotreatment carried out simultaneously [38,39]. Some literatures were found using simultaneous adsorption and biotreatment for elimination of phenol and cyanide [40–42].

Hence, in this present investigation adsorption, biotreatment and simultaneous adsorption and biotreatment (SAB) of phenol and cyanide containing aqueous solution were carried out in a shake flask method. In this study, the batch experiments have been carried out with an initial concentration range 10–100 mg/L of cyanide and 100–1000 mg/L of phenol. The effect of process parameters such as adsorbent dose, contact time, pH, temperature and initial concentration on percentage removal were also carried out in batch experiment. The ability of microorganisms to grow at high phenol and cyanide concentration and to degrade both phenol and cyanide were calculated in biotreatment method.

2. Materials and methods

2.1. Adsorbate and adsorbent preparation

All AR grade chemicals were used without further treatment and supplied by Himedia Laboratories Pvt., Mumbai, India. To prepare stock solution containing 100 mg/L cyanide was prepared by dissolving 0.0189 g of NaCN in 1 L of millipore water (Q-H2O, Millipore Corp. with resistivity of 18.2 MX-cm). Stock solution comprising 1000 mg/L of phenol was prepared by dissolving 1 g of pure phenol crystal in 1 L of millipore water. All experimental solutions of phenol and cyanide were prepared by diluting the stock solution with appropriate amount of millipore water. Coconut husk obtained from local market of IITR roorkee, INDIA was cleaned and washed with millipore water. After washing, coconut husk was dried in an oven for 12 h and carbonized in muffle furnace at 300 °C for 2 h. The acid activation of carbonized coconut husk was carried out by 2N H_2S0_4 at atmospheric temperature with periodic stirring for about 24 h. The coconut husk activated carbon (CHAC) was washed with millipore water to remove residual chemicals and dried at 60 °C for 24 h in an oven to completely remove moisture. This CHAC was used throughout the study as an adsorbent. The surface characteristics of CHAC are as follows: BET surface area: 310 m^2/g, porosity: 0.51 mL/g, moisture content: 5.5%, bulk density: 1.20 g/mL and ash content: 2.2%.

2.2. Microorganism and growth medium

S. odorifera MTCC 5700 species was isolated from coke wastewater in laboratory by Agarwal and Balomajumder [42]. The growth medium for *S. odorifera* MTCC 5700 are as follows: Na_2HPO_4, KH_2PO_4, NaCl, NH_4Cl, $MgSO_4.7H_2O$, and Glucose.

2.3. Development of inoculum and experimentation

The acclimatization of *S. odorifera* MTCC 5700 in phenol and cyanide environment was featured as follows: The freeze dried culture was inoculated in the sterilized liquid growth medium. The experiments were carried out in a 250 mL round bottom flask with a working volume of 100 mL at 120 rpm in an incubator cum orbital shaker (Metrex, MO-250, India) [42]. The pH of growth medium was adjusted with 1N NaOH or HCl and incubated in shaker at 30 °C for 48 h. After 48 h, the medium in flask becomes turbid representing microorganism growth in the flask. Measured amount of phenol and cyanide were added into flask media with initial concentration of 50 mg/L for phenol and 5 mg/L for cyanide. The growth of *S. odorifera* MTCC 5700 initially inhibited and the growth of microorganisms starts after 8 h. The phenol and cyanide were continuously added with increments of 50 mg/L of phenol and 5 mg/L of cyanide till the phenol and cyanide concentration in the growth medium reached 1000 mg/L and 100 mg/L, respectively. The *S. odorifera* MTCC 5700 was familiarized to phenol and cyanide in the growth medium starting from a lower concentration range of 5–100 mg/L for cyanide and 50–1000 mg/L for phenol. The temperature of process was adjusted at 30 °C.