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Optimization study of adsorption parameters for removal of phenol on aluminum impregnated fly ash using response surface methodology

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ABSTRACT

The current study envisaged the efficacy of adsorptive removal of phenol using aluminum impregnated fly ash (AFA). Central composite rotary design (CCD) by response surface methodology (RSM) was used to optimize the adsorption process. The adsorption parameters such as adsorbent dose, pH, contact time and temperature on phenol removal were used as the independent variables in CCD and their effects were investigated. Based on CCD design, the quadratic models were developed correlating the adsorption variables to two responses, *i.e.* removal efficiency and adsorption capacity. Analysis of variance (ANOVA) was incorporated to judge the adequacy of model. At initial concentration (C_0) of 200 mg/l, the optimum conditions of 13.63 g/l of adsorbent dose, 6.79 h of contact time, 5.65 of pH and 34.44 °C of temperature gave 86.4% removal efficiency and 12.67 mg/g adsorption capacity. Model prediction was in good agreement with experimental results and AFA was found successful in removing phenol from its aqueous solution.

1. Introduction

The advent of industrialization and technologies have comforted people's life but also resulted in increased environmental problems due to the accumulation of waste discharged in water, air and soil. Among various harmful and toxic pollutants, concentration of phenol has increased in last few years due to its extreme toxicity and persistency in the environment. It has been listed as priority pollutants by the Ministry of Environment and Forests (MOEF), Government of India and Environmental protection agency, United States (USEPA) [1]. World Health Organization (WHO) has limited its concentration level to 1.0 mg/l in drinking water [2]. Toxic effects of phenol on humans include anorexia, fainting, vomiting, liver damage, kidney damage, headache, coma, paralysis and other serious mental problems [3]. Therefore, considering the environmental implications, their treatment is important for safe discharge.

Among various available techniques, adsorption using activated carbon is conventional method [4], however, low cost adsorbent such as bagasse [5], coconut shell [6], tea waste biomass [7], pressmud [8] and fly Ash (FA) [9] are practiced recently due to its attractively low cost. FA is an industrial by-product found in abundance having low regeneration losses and leaching properties. It is produced as waste thus scientists are trying to find its alternative use to deal with its disposal problem. It is found that if 42 million tones of fly ash were recycled for some use, it would reduce 27 500 acre ft of landfill space. Raw fly ash has low adsorption capacity and need to be pretreated and modified therefore, chemical modification by impregnation of metal ions on adsorbent can increase its adsorption capacity for removal of various pollutants. However, more research is required in this field and its industrial application is yet lagging.

Some of the work done have been carried on impregnation of adsorbent for removal of variety of pollutants. Mondal et al. studied the adsorption of arsenic on copper impregnated GAC (2.5%) showing specific uptake of 0.0299 mg/g [10]. Bhumica et al. studied the equilibrium isotherm and kinetic for co-adsorptive removal of phenol and cyanide on iron impregnated GAC [11]. Banerjee et al. showed the efficiency of iron and aluminum impregnated fly ash for removal of heavy metals, nickel and zinc [12]. However, very few work has been reported on Al impregnated fly ash for phenol removal.

The intention of the present research is the preparation of aluminum impregnated fly ash (AFA) to utilize its potential for removal of phenol from aqueous solution. Influence of adsorption parameters such as adsorbent dose, pH, contact time and temperature on phenol removal was investigated. A central composite design (CCD) by response surface methodology (RSM) using Design Expert software version 8.0.7.1 (STAT-EASE Inc., Minneapolis, USA) was selected to elucidate the simultaneous

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Nomenclature

AFA	aluminum impregnated FA
AL	aliased
ANOVA	analysis of variance
AR	analytical grade reagent
b_i	linear coefficient
b_0	intercept coefficient
b_{ii}	quadratic coefficient
b_{ij}	interaction term
C_e	equilibrium concentration of phenol in solution (mg/l)
C_0	initial concentration of phenol in solution (mg/l)
CCD	central composite design
cor total	totals of all information corrected for the mean
CV	coefficient of variation
DF	degree of freedom
F	number of factors
F	Fischer statistics
FA	Fly ash
M	adsorbent dose
n_c	center runs
N	number of experimental runs
pH_0	initial pH
prob > F	proportion of time or probability expected to obtain the stated F value
PRESS	predicted residual error sum of squares
q_e	adsorption capacity at equilibrium (mg/g)
R^2	correlation coefficient
RSM	response surface methodology
RE	Remark
SD	standard deviation
SEM	scanning electron microscopy
SU	suggested
t_i	time (h)
T	temperature (°C)
V	volume of adsorbate solution (l)
W	mass of the adsorbent (g)
x_i	coded value of the ith variable
X	function of the distance from the point to the design center
X_i	natural value of the ith variable
X_{max}	highest limits of the ith variable
X_{min}	lowest limits of the ith variable
X_1	first factor, adsorbent dose (mg/g)
X_2	second factor, contact time (h)
X_3	third factor, pH
X_4	fourth factor, temperature (°C)
XRD	X-ray diffraction
Y	variance of the predicted response
Y_i	predicted response
Y_1	first response, removal efficiency (%)
Y_2	second response, adsorption capacity (mg/g)

Greek symbol

$\pm\alpha$	axial points

effects of parameters on adsorption process. The multiple regression analysis technique in the RSM was utilized to develop the equations and empirical models correlating the removal efficiency and adsorption capacity to four adsorption variables. The analysis of variance (ANOVA) was included to judge the adequacy of model. The optimized conditions developed from model were validated experimentally and feasibility of AFA for phenol removal was also noted.

2. Materials and methods

2.1. Preparation of adsorbent

Coal FA was obtained from thermal power plant near New Delhi, India. The sample of FA was washed and treated with 0.5 M H_2SO_4 in 1:2 ratio (solid to liquid). The mixture was oven dried at 110 °C for 2 h to remove moisture and impurities. 100 g of dried sample is then treated with aluminum sulphate solution containing 3% Al^{3+} in 1:3 ratio of solid to liquid. The impregnation was performed at 70 °C and pH 5 (as the precipitation was observed above pH 5) until the complete evaporation was observed followed by drying at 110 °C for 24 h. The pH of the solution was maintained at 5 by adding solution of 1N HCl. The dried material was washed with Millipore water until the washing liquid was free from Al ions and then dried to constant weight. The final product was used to carry out the batch experimentation.

2.2. Batch adsorption studies

Phenol (C_6H_5OH) of analytical reagent grade (AR) from Himedia Laboratories Pvt. Ltd. (Mumbai, India) was used for preparing the synthetic solutions. Stock solution of 1000 mg/l of phenol was prepared by dissolving 1 g of pure phenol crystal in 1 l of Millipore water and stored in a brown colored glass bottle to avoid its photo-oxidation. The batch experimentation was performed in 150 ml conical flask with 50 ml working volume taking initial concentration (C_0) equal to 200 mg/l. The flasks were agitated in temperature controlled orbital shaker (Metrex MO-250, India) at 120 rpm. The pH of the solution is controlled using dilute aqueous solution of either HCl or NaOH. 1 ml of sample was withdrawn and filtered with standard Whatman filter paper (cat. no. 1001 125) and properly diluted for analysis of phenol. The concentration of phenol was estimated by UV/vis spectrophotometer (UV 210 A, Schimadzu, Japan) by determining its absorbance at 270 nm wavelength. The equilibrium adsorption uptake, q_e (mg/g) and percentage removal efficiency of phenol was determined using Eqs. (1) and (2):

$$q_e = \frac{(C_0 - C_e)}{w} V \quad (1)$$

$$\text{Removal efficiency } (\%) = \frac{(C_0 - C_e)}{C_0} 100 \quad (2)$$

where C_0 is the initial adsorbate concentration (mg/l), C_e is the equilibrium adsorbate concentration (mg/l), V is the volume of the solution (l) and w is the mass of the adsorbent (g).

2.3. Analytical methods

The morphology of FA and AFA were evaluated using LEO 435 VP Scanning electron microscope (SEM) at 15 kV and 1000 magnification. The pore diameter and specific surface area were measured by Brunauer–Emmett–Teller (BET) method and nitrogen adsorption isotherm using an ASAP 2010 Micromeritics instrument, using the software of Micromeritics. Nitrogen was used as