Review on Research for Removal of Phenol from Wastewater

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***Abstract* -** Phenol is a major pollutant in the wastewater because of its presence in the effluent of major processing and refining plants. It has severe effect on human being, both short term and long term. Various methods are used for removal of the phenol from wastewater such as adsorption, photodecomposition, volatilization and other various biological and non-biological methods. In the present study attempt is done to present the survey of the research on the phenol removal by various methods. The methods such as Polymerization, electrocoagulation, extraction, photodecomposition, advanced oxidation and ion exchange were used effectively by various investigators. These methods are reported to be efficient for the phenol removal. Suitable method for phenol removal can be selected based on availability of the material, extent of separation required and properties of phenolic effluent.

***Index Terms* -** phenol, effluent, concentration, enzyme, adsorbent

I. Introduction

Developing green and sustainable technology for the effluent treatment is very important research area in this era of industrial and social development. Many researchers have carried out the research in this field. One of the important pollutants from this point of view is phenol. It has its presence in the effluent from major chemical and pharmaceutical industries such as petrochemical industries, petroleum refineries, coal gasification operations, liquefaction process, resin manufacturing industries, dye synthesis units, pulp and paper mills and pharmaceutical industries. It is a highly corrosive and nerve poisoning agent. Phenol causes harmful side effects such as sour mouth, diarrhea, impaired vision, excretion of dark urine. It is also toxic for fishes.

The toxic levels usually range between the concentrations of 10-24 mg/L for human and the toxicity level for fish between 9-25 mg/l. Lethal blood concentration of phenol is around 150-mg/100 ml. Various treatment processes used for the removal and/or recovery of phenols are hot gas or steam stripping, adsorption, ion exchange solvent extraction, oxidation, phase transfer catalysis and biological treatment processes. Phenolic waste water is treated using activated carbon in the fixed bed/ moving bed/ fluidized bed.

II. VARIOUS METHODS FOR PHENOL REMOVAL

A.Polymerization

Stanisavljević and Nedić have carried out a research on phenol removal by polymerization. They carried out the phenol removal in a reaction step which was polymerization of phenol in presence of an enzyme horseradish peroxidise (HRP)[1]. They used hydrogen peroxide along with the peroxidase enzyme. The native enzyme (E) is oxidized by peroxide (H_2O_2) to an active intermediate enzymatic form. This accepts an aromatic compound into its active site and carries out its oxidation. A free radical is produced and released into solution leaving the enzyme in the compound state. This compound oxidizes a second aromatic molecule, releasing another free radical product and returning the enzyme to its native state, thereby completing the cycle. The Free radicals formed during the cycle diffuse from the enzyme into the bulk solution where they react to form polyaromatic products. These polymers are water-insoluble and may be removed by solid liquid operations. It was observed that phenol conversion in all experimented conditions was greater than 90%. The high efficiency observed is in accordance with conditions optimized to guarantee 90% polymerization using purified HRP. There is a compromise between the reduction in variable costs by the use of fewer enzymes and the increase in capital investment at the time of building the treatment facility. The effect of horseradish peroxidase (HRP) and H_2O_2 concentrations on the removal efficiency of phenol, defined as the percentage of phenol removed from solution as a function of time, has been investigated by Vasudevan and Li[2]. They found that the phenol is almost completely precipitated within 10 minutes. . The removal efficiency increases with an increase in the concentration of HRP, but an increase in the time of treatment cannot be used to offset the reduction in removal efficiency at low concentrations of the enzyme, because of inactivation of the enzyme. Nicell et. al. have carried out the search on phenol polymerization and precipitation using polyethylene glycol as additive [3]. They observed that the presence of polyethylene glycol enhance enzyme performance for the treatment of synthetic wastewaters containing a range of phenol concentrations between 0·5 and 16 millimolar (47 to 1500 mg/l) at neutral pH. Results demonstrated a significant improvement in catalyst lifetime in the presence polyethylene glycol in both batch and continuous stirred tank reactor configurations. Free and immobilized horseradish peroxidase was used for phenol polymerization by Pradip et. al. [4]. They used Horse Radish roots for the extraction of peroxidase enzyme in the laboratory. Buffer of pH 6.8 was used to soak the roots in order to avoid the enzyme inactivation during crushing and extraction. They observed that phenol polymerization of 84% was achieved at 100 mg/l using free HRP enzyme and phenol degradation of 62% was observed using Immobilized HRP enzyme bed reactor. They also observed that as the concentration of phenol increased, there was reduction in phenol polymerization efficiency. Effect of reaction condition on phenol polymerization was studied by Masuda et.al. [5]. They investigated the quantitative relationships between removal efficiency of phenol and reaction conditions using coprinus cinereus peroxidase. At an adequate enzyme dose, they observed that the most effective ratio of hydrogen peroxide to phenol was nearly 1/1 (mol/mol). They also observed that 12.2 U of the enzyme was needed to remove 1 mg of phenol.

B. Electro coagulation

Abdelvahab et.al. have carried out the research on electrochemical removal of phenol from oil refinery waste [6]. They explored the possibility of using electro coagulation for phenol removal. They used the cell with horizontally oriented aluminum cathode and a horizontal aluminium screen anode. They studied the phenol removal with respect to various parameters such as pH, operating time, current density, initial phenol concentration and addition of NaCl. According to them removal of phenol during electro coagulation was due to combined effect of sweep coagulation and adsorption. The results showed that, at high current density and solution pH of 7, remarkable removal of 97% of phenol after 2hour can be achieved. Also they observed that the rate of electro coagulation increases with decrease in phenol concentration and the maximum removal rate was attained at 30 mg /l phenol concentration. The study showed that, electro coagulation of phenol using aluminum electrodes is a promising process. Effect of the variables like pH, operating time, current density, initial phenol concentration and addition of NaCl on phenol removal by coagulation was studied by Zazouli and Taghavi [7]. They observed maximum phenol removal percentage at a pH value of 7. They also observed that increasing the concentration of phenol led to decrease in the removal efficiency. They also observed that the removal rate of phenol increased with increasing current density, and the highest current density (25 mA/cm2) had the maximum removal efficiency. Ashtoukhy et. al. have carried out the research on treatment of petrochemical wastewater containing phenolic compounds by electro coagulation using a fixed bed electrochemical reactor[8]. They explored the possibility of using electro coagulation to remove phenolic compounds from oil refinery waste effluent using an electrochemical reactor with a fixed bed anode made of randomly oriented aluminium rasching rings packed in a perforated plastic basket located above the horizontal cathode.

C. Extraction

The extraction of phenol from simulated sebacic acid waste water was tried by Rao et. al.[9]. They used 1-hexanol, 1-heptanol and 1 octanol as solvent for phenol removal. It was observed that 1 octanol showed lesser phenol removal efficiency compared to other two solvents. Xu et. al. have carried out investigation on extraction of phenol in wastewater with annular centrifugal contactors [10]. They carried out the experimental study on treating the wastewater containing phenol with QH-1extractant (the amine mixture) and annular centrifugal contactors. They observed that the extraction rate of the three-stage cascade was more than 99%. When 15% NaOH was used for stripping of phenol in QH-1(the amine mixture), the stripping efficiency of the three-stage cascade was also more than 99% under the experimental conditions.

D. Photodecomposition

Investigation of the photodecomposition of phenol in near-UV-irradiated aqueous TiO_2 suspensions was carried out by Ilisz et. al.[11]. They investigated The effects of charge-trapping species on the kinetics of phenol decomposition. They observed that the heterogeneous degradation of phenol followed apparently zero-order kinetics up to 70% conversion. The results of the experiments in the presence of Ag^+ indicated that the phototransformation of phenol can proceed via direct electron transfer, neither dissolved O_2 nor its reduced forms play a significant role in the degradation mechanism. Akbal and Onar have studied photocatalytic degradation of phenol [12]. They carried out the investigation to study photocatalytic degradation of phenol in the presence of UV irradiated TiO_2 catalyst and H_2O_2. They concluded that photocatalytic degradation can be effective method for phenol removal.

E. Biological Methods

Marrot et.al have carried out the research on biodegradation of high phenol concentration by activated sludge in an immersed membrane bioreactor.[13]. They investigated the effect of adaption of mixed culture on phenol degradation. They found that biological treatment was economical and practical for removal of phenol. High concentrations of phenol are inhibitory for growth. Biological and enzymettric treatments were used for phenol removal by Bevilaqua et. al[14]. The systems studied were conventional batch aerobic biological followed or preceded by enzymatic treatment. They employed the Tyrosinase as enzyme.. They observed that biological treatment effectively degrades phenol upto concentration of 420 mg/l. Enzymatic polishing of biotreated effluent removed up to 75% of the remaining phenol in a four-hour reaction with 46 $U.mL^{-1}$ of tyrosinase and 50 $mg.L^{-1}$ of chitosan (used as coagulant). The research on detection of phenol degrading bacteria and pseudomonas putida in activated sludge by polymerase chain reaction was carried out by Movahedyan[15]. According to results in this study, the best phenol-degrading bacteria that can utilize 500 – 600 mg/l phenol completely after 48 hours incubation belong to Pseudomonas Putida strains. It is clear that use of isolated bacteria can lead to considerable decrease of treatment time as well as promotion of phenol removal rate. Tziotzios et. al. have reported research on biological phenol removal[16]. They carried out research to study the efficiency of packed bed reactor on phenol biodegradation and effect of specific area of specific material on biodegradation efficiency. They used the indogeneous bacteria from olive pulp. The maximum phenol removal rate was observed to be 12.65 gram per litre per day, when filled with gravel support material under draw fill operation. Same operation with plastic packings showed maximum phenol removal of 4.3 grams per litre per day. A review on recent advances in biodegradation of phenol was carried out by Basha et al[17]. Comparative study of free and immobilized growth was carried out by Pishgar et. al[18]. They observed that the culture was able to degrade the phenol upto 700 mg/l. The immobilized cells were able to remove phenol at concentration of 100 to700 mg/l in a slightly shorter time period. The biodegradation rate of phenol improved when immobilized cells were applied. Kinetic Studies for an aerobic packed bed biofilm reactor for treatment of organic wastewater with and without phenol was carried out by Dey and Mukharjee[19]. They carried out studies on removal efficiency of COD and phenol in a mixture of carbohydrate and phenol.The aerobic bioreactor with the glass beads was used for the purpose. They concluded that the performance of the bioreactor decreased marginally under 50 mgL^{-1} phenol charging along with other carbohydrate in the influent wastewater.

At this condition, phenol removal rate of 89% was achieved. Almsi et. al. have carried out the work related to anaerobic wastewater stabilization pond for phenol removal[20]. The phenol removal efficiency was found to 90 percent. According to the study, anaerobic ponds with petroleum wastewater is attractive alternative than many other methods. Removal of phenol by using rotating biological contactors was tried by Pradip et.al.[21]. They examined the impact of process variables, viz concentration, rotational speed and percentage submergence of the disc on phenol removal.

F. Electro-Fenton (EF-Fere) method

An improved Electro-Fenton (EF-Fere) method using H_2O_2 amendments and electrogenerated ferrous ions was investigated to treat phenol-containing wastewater by Jiang et.al.[22]. The degradation process of phenol was carried out in an EF-Fere system, which was composed of a power source, a cylindrical electrolytic cell and a H_2O_2 dosing system. The electrolysis was controlled by an electrochemical working station. For the phenol degradation experiments conducted in the EF-Fere electrolytic system, the maximum COD removal efficiency of phenol-containing wastewater is achieved at the condition of 800 mg/L initial ferric ions concentration, 1.0A electric current with continuous H_2O_2 addition mode. Furthermore, SnO_2 film anode and UV irradiation in the EF-Fere system are beneficial to COD removal efficiency.

G. Advanced oxidation processes

Rubalcaba et. al. have used advanced oxidation processes coupled to a biological treatment for phenol remediation[23]. Results showed promising research ways for the development of efficient coupled processes for the treatment of wastewater containing toxic or biologically non-degradable compounds. Similar research has been carried out by Esplugas et. al. [24]. Though Fenton reagent was most effective for degradation of phenol,. lower costs were obtained with ozonation. In the ozone combinations, the best results were achieved with single ozonation. Phenol degradation in presence of chlorides and sulphates was carried out by Siedlecka et. al.[25]. They studied the degradation of three representatives of phenolic compounds in presence of chlorides and sulphates, namely phenol, 2 chlorophenol and 2 nitrophenol. The presence of anions influenced the degradation rates. Relatively low degradation rates were observed for 2 chlorophenol. The biodegradability of phenol was increased by chloride while that of other two derivatives was increased by presence of sulphates.

H. Adsorption and Ion exchange

Experiments were conducted to examine the liquid-phase adsorption of phenol from water by silica gel, activated alumina, activated carbon by Roostaei and Tezel [26]. Experiments were carried out for the analysis of adsorption equilibrium capacities and kinetics. They found it to be a promising method for phenol removal. The research on phenol removal from aqueous solution by adsorption and ion exchange mechanisms onto polymeric resins was done by Caetano et.al. [27]. They evaluated the removal of phenol from aqueous solution by using a nonfunctionalized hyper-cross-linked polymer macronet and two ion exchange resins. The nonfunctionalized resin reported the maximum loading adsorption under acidic conditions, where the molecular phenol form predominates. Ion exchange resins showed the maximum removal in alkaline medium. Desorption of nonfunctionalized resin was achieved by using the solution (50% v/v) of methanol/water with a recovery close to 90%. In the case of the ion exchange resins the desorption process was performed at different pHs. Qadir and Rhan have investigated the removal of phenol by using adsorption [28]. The treatment of waste with active carbon is considered to be an effective method for the removal of phenol from waste solution because of its large surface area. The experimentation on adsorption isotherms for phenol removal on activated carbon was carie out by Maarof et.al[29]. The adsorption isotherm parameters for the Langmuir and Freundlich models were determined using the adsorption data. It was found that both the Langmuir and the Freundlich isotherms described well the adsorption behavior of phenol on Norit Granular Activated Carbon) NAC 010, while the Freundlich isotherm described very well the adsorption of phenol on NAC 1240. Jadhav and Vanjara have carried out the research on adsorption of phenol sawdust, polymerized saw dust and sawdust carbon[30]. They studied the influence of the parameters like concentration, agitation speed, amount of adsorbent and the pH on adsorption capacity. Hycinthe as adsorbent was used for phenol removal by Uddin et.al[31]. They carried out the batch kinetic and isotherm studies under varying experimental conditions of contact time, phenol concentration, adsorbent dosage and pH. The infered that the adsorption of phenol decreased with increasing pH. The results also showed that kinetic data followed closely to the pseudo-second-order model. Activated carbons prepared from date stones was used for removal of phenolic compunds from aqueous solutions by adsoption by Dhidan[32]. He used activated carbon prepared from date stones by chemical activation with ferric chloride (FAC) as an adsorbent to remove phenolic compounds such as phenol (Ph) and p-nitro phenol (PNPh) from aqueous solutions. He achieved the maximum phenol removal of 98% at pH value of 5 and 90 minutes of contact time. Kadhim and Al-Seroury have carried out research on characterization the removal of phenol from aqueous solution in fluidized bed column by rice husk adsorbent.[33].They found that the pretreatment of rice husk increase the specific surface area and changed the functional groups , therefore leads to increase in the capacity of adsorption. Kulkarni et. al. have investigated the phenol removal from the effluent by using activated carbon in batch and fluidized bed experimentation[34]. In batch studies they studied the effects of various parameters like adsorbent dose, pH, particle size on rate of adsorption . In case of fluidized bed, the effect of various parameters like concentration, fluid flow rate and adsorbent particle size were studied. The % decrease in phenol concentration increases with increase in adsorbent dose. With a reduction in particle size, initially steep increase in % removal of phenol is observed and it becomes more significant for finer particles.In case of fludized beds, Increase in fluid flow rate gives better adsorption in case of activated carbon. In the present study particle size of 0.420 mm is found more beneficial.Girish and Murti have studied the potential of various low cost adsorbents for phenol removal[35]. This review indicated that these agricultur-al materials have equivalent or even more adsorption capacity to activated carbon. Lua and Jia have used oil palm shell activated carbon in a fixed bed adsorber for phenol removal.[36]. The used the adsorption using the activated carbon derived from oil palm shells for the phenol, which they found to be very effective.

A research on mass transfer coefficients in a packed bed using tamarind nut shell activated carbon to remove phenol was done by Goud et. al[37]. They reported work on the preparation and characterization of activated carbon from tamarind nutshell, an agricultural waste byproduct, and its use in a packed bed for the removal of phenol. They found that that the sorption of phenol is dependent on both the flow rate and the particle size of the adsorbent, and that the breakpoint time and phenol removal yield decrease with increasing flow rate and particle size. For mass transfer coefficient, experimental values were in excellent agreement with the predicted values from the correlation. The experimentation for examining the application of adsorption packed-bed reactor model for phenol removal was done by Sorour et.al[38].They conducted the experiments to determine the Langmuir equilibrium coefficients and to determine the bulk sorbate solution concentration for different adsorption column depths and different time as well. They predicted a packed bed model which was in well agreement with the laboratory data. Batch and column studies for phenol removal on to onto nano iron oxide and alginate microspheres was carried out by Soni et.al[39]. They studied the adsorption behavior of o-nitrophenol from aqueous medium, using nano iron oxide loaded calcium alginate beads using equilibrium batch and column flow techniques. They reported the sorption capacity per unit bed volume and rate constant to be 578.4 mg/L and 1.18 L/mg/min, respectively. Kulkarni et.al. have used tamarind bean activated carbon for phenol removal in fluidized bed[40]. They studied the effect of the effect of various parameters like concentration, fluid flow rate and adsorbent particle size.They observed that It is observed that as the concentration increases the percent saturation of adsorbent increases. Also increase in fluid flow rate gives better adsorption.

I. Membrane based separation

Membrane-Based Separation of phenol/water mixtures using ionically and covalently cross-linked ethylene-methacrylic acid copolymers was tried succesfully by Mixa and Staudt[41]. They performed the Membrane-based separation of phenol/water mixtures with concentrations of phenol between 3 wt% and 8 wt% in the feed with nonmodified as well as cross-linked ethylene-methacrylic acid (E-MAA) copolymers with different amounts of methacrylic acid. They concluded that using nonmodified membranes with higher methacrylic acid monomer content in the polymer, lower fluxes and higher enrichment factors were observed. Also the Investigation of different cross-linked membranes showed that with high phenol concentration in the feed, ionic cross-linking seems to be very promising. Polyurethane urea as pervaporation membrane for the selective removal of phenol was tried by Gupta et.al[42]. They reacted the Hydroxyterminated polybutadiene (HTPB) with 2,4-toluene diisocyanate (TDI) followed by the addition of a diamine chain extender (prepared by the condensation reaction of 4,4′-diaminodiphenylsulfone and terepthalaldehyde) to prepare an imine containing polyurethane urea (PIUU).About 88 percent of phenol was seperated as a condensed permeate. Diaconu et. al have carried out research on use of membrane techniques for phenol separation[43].They presented a separation study of some phenolic compounds frequently encountered in the environment: m-nitrophenol, p-nitrophenol, m-cresol, p-cresol, using the bulk liquid membranes technique. They used chloroform as the organic solvent for the membrane and studied the operational parameters of the transport and also established the optimum separation conditions (the feed phase pH, the receiving phase pH, the time period of the transport). The best transport efficiencies were obtained for m-nitrophenol and p-nitrophenol. The transport efficiencies in the case of these phenolic derivatives were 92% for nitrophenol and 98% for p-nitrophenol.

III.CONCLUSION

For removal of phenol extensive research has been carried out to study the removal potential of each of the method. Many biological and nonbiological processes such as polymerization, electro coagulation, extraction, photodecomposition, advanced oxidation process and adsorption were employed various researchers for phenol removal. By using the enzyme horseradish peroxide, for phenol polymerization. At optimal conditions, phenol polymerization efficiency of about 90 percent was observed. The removal of phenol during electro coagulation was due to combined effect of sweep coagulation and adsorption. At high current density and solution pH of 7, remarkable removal of 97% of phenol after 2 hour was achieved.

Photoreactor for wastewater treatment using titanium dioxide nanoparticles is also promising alternative. Various suspended and attached growth processes were also used effectively and phenol removal efficiency of 90 % was achieved. By using electro-Fenton process the removal efficiency of 96% was obtained. Adsorption operation by various adsorbents is also attractive alternative. It has the advantage of wide range of adsorbents available. Many adsorbents were waste materials from agricultural and other industrial operations such as distilleries. Phenol removal ranging from 88 % to 95 % was observed using various adsorbent. Adsorption seems to be possibly most widely studied operation for phenol.

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