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Waste Water Treatment-Bed of Coal Fly Ash for Dyes and Pigments Industry

Syed Farman Ali Shah¹, Aziza Aftab¹, Noorullah Soomro¹, Mir Shah Nawaz¹ and Kambiz Vafai²

¹Mehran University of Engineering and Technology, Jamshoro, Pakistan, ²University of California Riverside, Ca, 92521, USA University of California, University of California, Riverside, Department of Mechanical Engineering, A363 Bourns Hall, Riverside, CA 92521-0425

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Abstract

The highly porous power plant waste ashes have been utilized to treat toxic effluent of a dyes manufacturing plant. An attempt has been made for the first time in Pakistan, to generate an effective and economically sound treatment facility for the toxic effluent of a dyes manufacturing plant. This is an indigenous bed which could replace expensive treatment facilities, such as reverse osmosis (RO), granulated activated carbon (GAC) bed, etc. The treatment efficiency was improved by coupling coagulants with fly ash adsorbent bed. The ash was collected from coal fired boilers of power plant at Lakhra Power Generation Company, Jamshoro, Pakistan. The use of this ash resolved the disposal and environmental issues by treating wastewater of chemical, dyes and pigment industry. The treatment bed comprised of briquettes of coal fly ash coupled with commercial coagulant ferrous sulfate-lime reduced COD, color, turbidity and TSS of effluent remarkably. An adsorption capacity and chemical behavior of fly ash bed was also studied. In coagulation treatment, coagulant FeSO4-lime influenced reduction of COD, color, turbidity and TSS by 32%, 48%, 50% and 51%, respectively. The CFAB coupled with coagulant, resulted an excessive removal of color, TSS, COD, and turbidity by 88%, 92%, 67% and 89%, respectively.

Keyword: Environmental pollution; Dye industry effluent; Coal fly ash; Adsorption; Chemical Oxygen Demand (COD).

Introduction

Dyes and pigment manufacturing industries have become a real threat to the environmental waters because they discharge around 150 tons of toxic and lethal chemicals per annum into water streams [1, 2]. The production of more than 10,000 types of dyes has exceeded to $7x10^5$ tons annually [3]. Since, these effluents carry an elevated concentration of lethal dyes, chemicals, metals, organic materials, acids and un-reacted dyes; they mainly consume oxygen in wastewaters. These effluents are harmful to health as they contain an elevated levels of COD (30-22000 mg/L), color, pH (1-14), turbidity (10-1000 mg/L), alkaline substances, temperature, acids, toxic substances and heavy metals [4], TDS(40-8000 mg/L).This variation is due to the nature of manufactured dye products [5-7].

Different biological and chemical methods have been employed for the elimination of dyes,

^{*}Corresponding Author Email: farman.shah@faculty.muet.edu.pk

COD and color from industrial effluents [8]. Chemical coagulation, fenton reaction [9], ultrafiltration, nano-filteration [10], reverse osmosis (RO), ozonation, oxidation, adsorption, and electrochemical techniques [11] are among the variety of methodologies which are used for the treatment of wastewater. Application of membrane technology is found successful but it is costly and discouraged as it creates fouling [12]. Ozonation eliminates color but does not reduce COD. Chemical coagulation is one of the mainly utilized tools designed for reduction of COD, color, suspended solids and turbidity [13, 14]. Adsorption is found efficient for the removal of pollutants from gases and wastewater. It is simple, fast, cost effective, reusable and highly efficient process if a low cost adsorbent is used [3]. However, a hybrid methodology comprised of coagulation and adsorption is projected to be more cost effective, environmental friendly and optimized treatment [15, 16].

Approximately 750 million tons of coal fly ash is produced worldwide as waste from coal fuel power plants by combustion of pulverized lignite coal [17, 18]. The generated fly ash is collected from coal fired boilers through the electrostatic precipitators. That waste creates grave disposal and environmental problems [19]. The existing annual production of coal fly ash is approximately 650 million tons, globally. Thar coal Pakistan has 3400 million tons of coal assets which are 6th biggest assets of coal in the world. Lakhra coal assets are likely to be about 1350 million tones and are used for electricity generation. In FBC power plant at Khanote, the production rate of bottom ash and fly ash is 17500 and 56800 m^3/hr , respectively [20]. Coal fly ash is acidic in nature, abrasive and grey in color. It has definite porosity of 0.48%, surface area in the range of 2600 -8000 cm^2/g , particle size of 130-980 μ m, pore volume of 0.033 cm³/g, bulk density of 700-1000 kg/m³ and specific gravity of 2.5-2.7 [21]. Many researchers have focused on utilization of coal fly ash for manufacturing of potassium fertilizer, zeolite, cement, flue gas adsorbent concrete, raw material for road structure and bricks making, reactive dyes, adsorption of phenolic compounds, organic compounds, heavy metals and organic pollutants from industrial flue gases and effluents. It was acknowledged from effluents analysis of dyes manufacturing plant, that industrial effluents contain these high concentration of organic pollutants such as BOD, dyes, COD, phenolic compounds, color, TDS, turbidity and suspended solids. It was observed that industrial FeSO₄ coagulation treatment technique was unproductive for appropriate treatment of industrial effluents of dyes manufacturing plant due to effluents mixture complexity. The quality parameters of effluent did not match to National environmental quality standards (NEQS). Such untreated effluents lead to unfavorable effects on human health, marine animals, underground water resources and soil fertility. Shah et al. [15] used a hybrid process of coagulation and adsorption for the reduction of color, COD, TSS and turbidity using fly ash of sugarcane bagasse. This method was effective but the regeneration of fly ash was expensive. Thakur & Parmar [22] studied the adsorptive removal of cadmium from synthetic wastewater using a 2 cm diameter fly ash column through three column adsorption experiments. The granulated fly ash was studied as adsorbent for the reactive brilliant orange X-GN, direct red 12B organic and dyesethylene blue in wastewater solution. Hsu et al. [23] has carried out the de-coloration of two acid dyes (acid black 1 and blue 193) and two reactive dyes (reactive red 23 and blue 171) from the aqueous solution using fly ash. They optimized the parameters of process such as contact time, pH, adsorption temperature and dye concentration. Mario et al. [24] studied the removal of composite dyes from wastewater by using fly ash as an adsorbent.

Materials and methods Sample collection of wastewater

Effluent sample of 5 liters was collected from pigment and dyes manufacturing plants effluent release point of Archroma Pakistan Limited Jamshoro. Laboratory scale testing of collected wastewater sample was performed for COD, TSS, EC, color, TDS, salinity, temperature and pH using standard laboratory protocols [14, 25]. The sequence adopted for the experimental work is shown in (Fig. 1).



Figure 1. Schematic diagram of adsorption process using CFA bed

Adsorbent bed preparation using coal fly ash Pulverizing and screening

Coal fly ash was pulverized through cooled disc-pulverizer (MFC No.8375, Type 1025 W, Yoshida Seisakusho Co. Ltd Japan) at a speed of 550 rpm to obtain improved mechanical properties [26]. Pulverized fly ash samples were screened through RO-Tap type sieve shaker (A-87205, Heiko Seisakusho, Japan) at a speed of 280 rpm. Fine powdered and porous fly ash samples were collected (Aperture 25 μ m, mesh 200, wires diameter: 25 μ m) from testing sieve (IIDA Manufacturing Co. Ltd, Japan).

Washing and pretreatment

Porous and fine ash was washed, treated and dried for the preparation of briquettes. Whatman filter paper 42 (Whatman Manufacturing Co. Ltd, U.K) and vacuum filter (125 μ m) was employed for washing and filtration of raw fly ash. These pre-washed samples of fly ash were further chemically treated with 10% concentrated sulfuric acid [27] for 12 hours and then washed finally with distilled water to eliminate the surplus amount of acid through filtration process. Pre-treated fly ash samples were dried at room temperature for 2 hours.

Preparation of briquettes

The pre-treated fly ash samples were further proceeded to prepare briquettes by using Briquetting Machine (20-1320 Simplimet® Mounting Press, Buehler Ltd, USA). Prepared briquettes of fly ash were dried in an oven (Model # Lo-201, The Grieve Corporation, USA) for 24 hours at 110 ⁰C to increase their adsorption capacity and then they were finally cooled at room temperature.

Design and construction of the bed

The bed was designed for the placement of prepared briquettes and to allow proper effluent flow through the bed in order to get maximum treatment and removal results. Specifications of briquettes are as under:

Weight of briquette	=	05 gm
Diameter of briquette	=	2 cm
Density of briquette	=	1.31
gm/cm ³		

The specifications of the bed structures are as follow:

Length of bed	=	45.72 cm
Width of bed	=	30.48 cm
Height of bed	=	45.72 cm
Length of inner pipe	=	121.92 cm
Diameter of inner pipe	=	2.54 cm

Treatment of industrial effluent

Effluent treatment applying coagulation

Effluent samples were treated by means of coagulation using Ferrous sulfate-lime coagulant. To carry out the coagulation, different parameters were maintained such as speed of agitation (145 rpm), mixing period (5 minutes), settling time (1 hour) and temperature (25 °C). After settling of effluent flakes and before their analysis, coagulated sample was filtered with Whatman filter paper 42 (125 μ m).

Treatment of industrial effluent through CFAB

Effluent samples were passed through the designed CFAB in order to remove pollutants at optimized parameters such as flow rate, retention time, etc. After adsorption process through CFAB, samples of treated wastewater effluent were filtered by the use of vacuum filtration system with Whatman filter paper (125 μ m) (Whatman manufacturing Co. Ltd. U.K). Quality of treated effluent samples was tested as per standard protocols of laboratory. Then comparison of single coagulation method and fly ash bed were performed to check adsorption capacity [28].

Adsorption percent and capacity of fly ash adsorbent for the removal of effluent pollutants

The effluent pollutants adsorption study was carried out by use of fly ash adsorbents contact time and flow rate. On fly ash briquettes, the effluent pollutants adsorption (%) was calculated as follows:

Adsorption % =
$$(100/Cf)^*(Ci-Cf)$$
 (1)

Where C_f (mg/L) is after adsorption final concentration of effluent pollutant and Ci (mg/L) is

before adsorption initial concentration of pollutants in effluent.

In similar manner the adsorption potential (mg/g) may be calculated as following:

$$Qe = V/m (Ci-Ce)$$
(2)

Where V is the effluent volume (mL) and m is the fly ash adsorbent mass (g), Ce and Ci are the equilibrium and initial pollutants concentration (mg/L) in effluent of dyes plant and Qe is the adsorption potential (mg/g) [29].



Figure 2. 3-D view of coal fly ash bed (COMSOL mph

Results and Discussion *Treatment of industrial effluent by coagulant method*

Effluent from the designated dye and pigment industrial plant was analyzed before and after the treatment with adsorbent bed. After coagulation, the levels of color, COD, turbidity and TSS were higher than the levels set by NEQS (Table 1) and the efficiency of 30-50% was recorded. It may be due to the non-partition of elementary bonding of the pollutants with wastewater.

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Table 1. Pre- and Post- coagulation treatment results of dyes and pigments.

Parameter	Pre-treatment	Post-treatment
Color (PtCo)	35600	15300
COD (mg/L)	2590	1760
Turbidity (FTU)	6500	3250
TSS (mg/L)	90	35
TDS (mg/L)	7090	7170
Salinity (ppt)	7.3	7.4
pH	8.5	8.5

Treatment of dyes plant effluent by coagulation method

It was determined from the prior results that effluents of composite nature and dyes were possibly refractory type to permit the elimination of contaminants by the given treatment. Hence, an advanced experimental work was conducted in order to increase the effluent treatment efficiency. Effluent samples of dyes were treated through coagulation process by means of FeSO₄-lime as coagulant. It was found from this treatment analysis that values of electrical conductivity, TDS and salinity were increased in FeSO₄-lime treatment. The decrease in pH during the process pointed out chemical reaction between the sample contents and the coagulant. The coagulant FeSO₄lime influenced elimination of COD by 32%, color by 57%, turbidity by 50% and TSS by 61% (Fig.3(a)).

Parallel treatment was given to the samples of effluent having composite nature after using coagulation process using FeSO₄-lime coagulant. The results achieved for the decrease in salinity, TDS and electrical conductivity were improved. Hence, parallel treatment enhanced the removal efficiency obtained by treating the samples with coagulant FeSO₄-lime. The addition of FeSO₄-lime to the effluent caused decrease in the amounts COD, color, turbidity and TSS (Fig. 3(a&b). These results implied that coagulant FeSO₄-lime was the better coagulant. Observed increase in the concentration of EC, salinity and TDS could be linked with the increase in the concentration of salts, the reaction of which with the reactive parts enclosed in the dyes effluents was possibly the essential reason behind the decline of color and COD. Due to disposal effluents composite nature the coagulation method resulted in minor decline in the concentration of organic pollutants.

Treatment of industrial dyes effluent by designed CFA bed

Material composition and analysis of fly ash

Adsorption is a physical method, the degree and rate of adsorption is definite for specified adsorbent that is subjective to its physical and chemical properties such as composition, surface area and pore size [3]. The fly ashes showed suitable chemical composition. Oxides of iron, silica and alumina in fly ashes are good coagulants for the decrease in concentration of toxins in effluent. Due to porosity, adsorption capacity and high surface area fly ashes worked as adsorbents like activated carbon.

In material composition of fly ashes the percentage of oxides of aluminum and iron was high. The chemical behavior of fly ashes proved it valuable for the industrial effluent treatment. Coal fly ashes have high porosity and surface area that result in greater adsorption capacity for pollutants [28]. The adsorption was affected largely by the particle size when fly ash was used. The smaller the particle size the greater the specific surface area, which promotes external surface adsorption. From fly ash analysis, it was found that chemical and physical treatment improved the porosity, surface area and adsorption capacity of fly ashes. Adsorption capacity was largely due to their structural uniqueness and porous texture which gave them a large surface area and their chemical nature can be simply customized by chemical treatment sequentially [30]. Effective treatment of effluent by means of adsorption depends upon chemical structure, adsorption capacity, surface area, particle size and porosity of fly ashes.

Treatment of dyes plant effluent by designed coal fly ash (CFA) bed

The adsorption process by using Coal fly ash (CFA) bed resulted in greater reduction of pollutants as compared to conventional methods. Various experiments were performed at different flow rates i.e 1 L/m, 2 L/m, 5 L/m and 10 L/m. The maximum reduction of 66.66% in COD was achieved at flow rate of 1 L/m and 62% at 2 L/m. The designed bed revealed better results at flow rate of 1 L/m because the adsorption capacity of bed depends on the size of bed and the retention time of effluent. Thus, the effluent was passed through the prepared bed at the flow rate of 1 L/m and COD, color, turbidity, TSS and TDS were reduced to 67%, 89%, 88% and 92%, respectively. The Results were in agreement with the results of Shah et al. [15].

The analysis chemical oxygen of demand (COD) is commonly employed to ultimately measure the total organic compounds present in water. Thus, it is an important tool for the measurement of quality of water. COD is expressed in ppm (parts per million) and/or milligrams per liter (mg/L), which represents the mass of oxygen consumed per liter of solution. The COD test is repeatedly performed to monitor the efficiency of water treatment plant. This check is based on the truth that a powerful oxidizing agent in acidic conditions can completely oxidize more or less any organic compound to carbon dioxide. The COD is the quantity of oxygen used to chemically oxidize organic water contaminants to inorganic end products. The results of CFAB are shown in Fig. 3(a).

Color is one of the major water quality parameters. Color is mainly caused by dissolved organic matter, e.g. fulvic and humic acids (organic disintegration products from vegetation). It may also be the result of impurities coming from minerals such as manganese and iron. The intensity of color may be a good indication of the organic content present in water. If color is due to the presence of organic matter, there is always a relation between the level of organic content and color intensity. Hence the decrease of organic matter in the wastewater is a significant parameter to be managed in industrial waste and to reduce the damage to the environment.

The coal fly ash bed showed very impressive results in reduction of color in wastewater treatment. Fig 3(a) shows the results obtained by passing the wastewater over designed bed.

Another important parameter of wastewater is turbidity, which is the evaluation of

water clearness and estimation of suspended material present in water. The suspended material decreases the passage way of light to the water. Suspended materials include algae, soil particles (sand, silt and clay), plankton, organic and microbes. Color of water can also be affected by turbidity. Higher turbidity value increases temperature of water because more heat can be absorbed by suspended particles. This sequentially decreases the concentration of dissolved oxygen (DO) because hot water holds a smaller amount of DO than cold water. Greater turbidity also decreases the amount of light penetrating in the water, which minimizes the production of DO and photosynthesis. Turbidity test is appropriate for evaluating the wastewater treatment plants cleaning efficiency. The turbidity results of CFA are given in Fig.3(a).

TSS are solid particles suspended in the water, including inorganic and organic material. These may include industrial wastes, plankton and silt. Absorption of light may be reduced due to higher values of suspended solids. TSS can include a wide variety of materials, such as decaying plant and animal matter, silt, sewage and industrial wastes. Suspended solids high concentrations can cause numerous troubles for aquatic life and stream health. The designed CFA bed has greater TSS reduction efficiency. TSS removal is shown in Fig. 3(b).



Figure 3(a) Effect of Packed Bed Treatment on Wastewater



Figure 3(b) Effect Of Packed Bed Treatment On Wastewater



Figure 4. Effect of flow rate on removal efficiency of CFAB in terms of COD



Figure 5. Color removal efficiency of CFAB at different flow rates



Figure 6. Effect of flow rate on efficiency of CFAB in term of turbidity reduction



Figure 7. Effect of flow rate on TSS removal efficiency of CFAB

Effect of Flow rate on removal efficiency of CFA bed

Effect of flow rate on COD reduction

Effluent samples of dyes were passed through designed coal fly ash bed at different flow rates and COD was compared. Highest reduction of 66.6% in COD was achieved at 1 L/m and 62% reduction at flow rate of 2 L/m. On increasing the flow rate the decrease in removal was observed. Thus, COD reduction rate increases with the increase in adsorbent contact time (Fig. 4). CFA Bed showed the most valuable results because CFA could capture organic compounds effectively due to high porosity. CFA reduced pollutants of effluent efficiently; its adsorption capacity depends upon the interaction of pollutants of effluent with the prepared adsorbent of fly ash. Presence of inorganic salts could boost the adsorption of organic species to carbon [31]. Coal fly ash bed showed less effect on pH reduction. However the range of pH was under NEQS.

Effect of flow rate on color reduction

Dyes wastewater was treated through coal fly ash bed at low variations. Highest reduction of 89% was resulted in color at flow rate of 1 L/m [32]. Color reduction rate depends upon adsorbent and effluent contact time. CFA bed showed valuable results for color decline (Fig.5). CFA bed proved most helpful system for color reduction due to high porosity. Higher rate of color reduction could be achieved due to specific surface area, chemical composition and porosity of fly ash [15]. Highly porous nature of fly ashes may possibly detain the effluent pollutants efficiently. Analysis of fly ash confirmed that it has higher porosity and can capture inorganic salts and organic pollutants. Coloring compounds of dyes were disintegrated and detained by fly ash pores in addition to fly ash elemental chemical behavior.

Effect of flow rate on turbidity reduction

At 1 L/m flow rate higher turbidity removal rates of 88% were observed. By increasing flow rate, adsorption of turbid material decreases (Fig. 6) Maximum adsorption rate achieved at lower flow rates and huge fall of effluent turbidity was observed. It was observed from experimental work that coal fly ash bed showed standardized performance in treatment of dyes wastewater [32]. Effluent's material causing turbidity was detained in the fly ash adsorbent pores. Maximum turbidity removal rate achieved due to high porosity and surface area.

Effect of flow rate on reduction of TSS

Highest total suspended solids removal of 92 % was achieved at flow rate of 1 L/m. Removal rate of TSS depends upon retention time of adsorbent [33]. The maximum contact of effluent with fly ashes briquettes yields maximum reduction of TSS (Fig.7). These adsorbent briquettes of fly ash were proficient for the TSS removal from wastewater of dyes due to chemical structure, surface area and high porosity of fly ash. Pores of fly ash briquettes capture the suspended solids and settle down. According to the adsorption capacity of adsorbent, equilibrium state can be achieved within specific contact time. Suspended solids interacted with pores of fly ash and were detained in pores of fly ash briquettes until equilibrium state achieved [28].

Effect of flow rate on adsorption (%) and adsorption capacity of CFAB

The high adsorption capacity of fly ash permitted the elimination of suspended or soluble organic pollutants and removal of color from dyes wastewater. Also found that fly ash has good capacity of settling, which makes it possible to eliminate suspended solids by acting as a settling aid [28]. Effluent sample of dyes was treated by means of adsorption process with briquettes of fly ash for the removal of effluent pollutants. The experimental study acknowledged that CFA adsorbent bed may possibly adsorb greatest proportion of pollutants such as color, COD, TSS and turbidity. It was concluded from coal fly ash adsorption study that it has greater affinity for pollutants of effluent. TSS removal through fly ash was found maximum. Dyes plant effluent was treated by means of adsorption process by the use of flv ash bed for the reduction of color, COD.TSS and turbidity. Adsorption capacity of CFA bed for COD, color, turbidity and TSS are shown below in (Fig. 8, 9, 10, 11), respectively.



Figure 8. Adsorption capacity (qe) of fly ash bed for reduction of COD of dyes effluent



Figure 9. Adsorption capacity (qe) of fly ash bed for the reduction of color of dyes effluent



Figure 10. Adsorption capacity (qe) of fly ash bed for reduction of turbidity from dyes effluent



Figure 11. Adsorption capacity (qe) of fly ash bed for removal of TSS from dyes effluent

Conclusion

CFA-Bed is an effective and inexpensive for the removal of color, COD, TSS and turbidity due to its high porosity and adsorption capacity. Simple adsorption or coagulation process is not the most effective for the treatment of dyes and pigments wastewater. Dyes and pigments waste water treatment efficiency was enhanced by the use of CFA-Bed and reduced the color, TSS, COD, and turbidity by 88%, 92%, 67% and89%, respectively. Problems of environmental pollution can also be minimized by utilization of CFA-Bed in dyes and textile industries waste water treatment.

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