

# Removal of Malathion from Wastewater by Coagulation-Adsorption Integrated Method

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**Abstract-** Commonly available poly -aluminium chloride is used as a coagulant and drumstick peel powder, bagasse fly ash are used as adsorbent for the removal of malathion in the present investigation. The efficiency of the coagulation –adsorption integrated method is tested for the determination of malathion using batch experiments under controlled conditions as a function of initial pH, contact time, initial concentration and the optimization amount of coagulant and adsorbent dose. These operation conditions applied on wastewater characterisation parameter namely COD, BOD, Turbidity and TSS. Malathion [1, 2-Di (ethoxycarbonyl) ethyl O, O-dimethyl phosphorodithioate; CAS 121-75-5] is most widely used insecticide in the world for the agriculture areas. Due to its low vapour pressure, apparent biodegradability and extensive use, malathion has led to the contamination of terrestrial ecosystems and has been detected in ground and surface waters in many countries beyond permissible limits. Malathion has caused a setback to the environment and also has increased potential risk to human health as it is an endocrine disruptor chemical, hence is posing a challenge to remove the malathion from wastewater. Currently, coagulation-adsorption integrated method is an effective technique can be used to treat malathion wastewater. Integrated process is environmental eco-friendly, low cost, low energy requirement and high efficiency for pollution uptake. Coagulation-adsorption integrated method for synthetic wastewater by using drumstick peel powder and bagasse fly ash malathion removal with respect to COD, BOD, TSS, Turbidity. They were 84.05%, 97.17%, 100%, 98.53% and 76.81%, 95%, 96.66%, 96% respectively. Same processes has been done for malathion industrial wastewater by using drumstick peel powder and bagasse fly ash gave very efficient removals with respect to COD, BOD, TSS, Turbidity. They were 98.53%, 98.11%, 98.88%, 98.96% and 94.63%, 96.22%, 97.77%, 97.72% respectively, under optimum conditions.

**Key word-** Adsorption, Bagasse fly ash, Chemical Coagulation, Drumstick peel powder, Malathion, Wastewater.

## I. INTRODUCTION

Clean water accessibility is a crucial problem in urban and suburb areas, especially for clean water which comply with

health requirement for daily uses [1]. Industrial wastewater generally contains high concentration of pollutants such as organic material, heavy metals and toxic compounds. Such wastewater causes environmental and health hazards and, consequently, must be properly treated before final disposal [2].

Pesticide/Agrochemical manufacturing industry wastewater poses pollution problems due to the toxic components, high chemical oxygen demand (COD), biochemical oxygen demand (BOD); high Total dissolved solids (TDS) and high alkaline pH in the range of 12-14. The most important portion of contamination due to this wastewater is observed in agricultural areas and in surface waters that come from agricultural areas. Major quality of pesticide pollution is released during pesticide manufacturing. Pesticide, usually have direct adverse effects on the living organisms [3].

Malathion which is a broad spectrum, highly toxic organophosphorous insecticide, registered in 1956, is abundantly used in agriculture, industrial and public health field to kill insects, ants, aphids, bagworm, beetle, cotton leaf worm, mosquito, lice etc. by inhibiting Cholinesterase enzyme. Malathion is rapidly and effectively absorbed by all routes including the gastrointestinal tract, skin, mucous membrane and lungs and causes blurred vision, excessive salivation, headache, giddiness, nausea and vomiting due to active metabolite malaoxon (metabolic product from malathion) which is 61 times more toxic than malathion. It affects nervous system, immune system, adrenal glands, liver and also carcinogenic in nature. Apart from this it is toxic to birds, fishes other aquatic invertebrates and honey bees too [4].

In the state of Maharashtra in Parbhani district, Godavari river water is found to contain 0.0823 mg/l of Malathion, which is alarmingly high and needs treatment [5-19].

The development and implementation of innovative water and wastewater treatment technology lends itself to providing operational and cost benefits for various client needs.

By using development and implementation innovative water and wastewater treatment solutions, municipalities can cope with evolving regulation, aging infrastructure and growing population. Industrial plants can meet the strictest regulatory requirements for wastewater discharge to the environment or to municipal collection systems, and address typical challenges such as variable inflow, seasonal peaks and high organic loads. Development and implementation innovative water and wastewater treatment solutions may also facilitate additional in-plant reuse of treated effluent with up to Zero Liquid Discharge (ZLD) and efficient energy recovery opportunities [20-22].

In this paper, the treatment of real wastewaters using chemical and adsorption techniques was conducted to develop an integrated low cost treatment process for the removal of TSS, Turbidity, BOD and malathion contaminants as well as for the reduction of COD.

## II. MATERIALS AND METHODS

### A. Reagent and solution

All the reagents used in this study are of analytical grade. All glass wares used were of Borosil. Distilled water was used for making the synthetic samples. Agrochemical (Malathion) industrial wastewater samples were collected. Technical grade malathion of 50 % (E.C.) pure. Acetone, Magnesium Sulphate, Alkali iodide azide, Starch indicator, Sodium thiosulphate pentahydrate,  $K_2Cr_2O_7$ , Mercury sulphate, Ferroin indicator, Ammonium iron sulphate hexahydrate. Standard base of 0.1N NaOH and acid 0.1N  $H_2SO_4$  solutions were used for pH adjustment.

### B. Preparation of Coagulants, coagulant aid and malathion synthetic wastewater.

Stock solution of malathion (50% E.C.), coagulants of poly aluminium chloride (P.A.C.) and poly electrolyte (P.E.) of coagulant aid was made according to standard methods. Distilled water was used for making the synthetic samples. Working samples were made by adding required stock solution to the predetermined quantity of distilled water. The experiments were performed with a stock solution of 1000 ppm concentration of malathion and poly aluminium chloride. But to prepare a stock solution of 100ppm concentration of polyelectrolytes.

### C. Preparation of adsorbent

The Bagasse fly ash (BFA) collected from Vasantdada Sahakari Sakhar Karkhana (Vithewadi, Nasik, Maharashtra, India) is used as a natural adsorbent. The drumsticks are collected from market. Remove peels kept in sunlight 2-3 days for drying. Dry peels crush in mixer grinder to create fine powder is used as a natural adsorbent. It is thoroughly washed with hot water (70°C), dried, and sieved by using standard sieves (IS 437 1979). The fractional sieve analysis of the particles of BFA and DPP were done and found as the average particle size of 425  $\mu$ m and 150  $\mu$ m (equivalent U.S. standard test sieves) was used for the adsorption studies. The physico-chemical characteristics of DPP and BFA were determined using the standard procedures.

## III. EXPERIMENTAL PROCEDURE

Synthetic Waste-water and Industrial Waste-water samples were subjected to an integrated treatment scheme involving chemical coagulation followed by adsorption onto raw drumstick peel powder (DPP) and bagasse fly ash (BFA) in order to remove the residual malathion. Chemical treatment comprised adjustment of wastewater to neutral pH followed by addition of either poly aluminium chloride solutions using varying doses of 500-900ppm while rapid mixing at 110 rpm for 1 minute using jar test apparatus. A flocculation step was also conducted by adding 0.5ppm poly electrolyte to some samples with gentle stirring at 5 rpm for 20 minutes followed by settling then filtration. The purpose of the additional adsorption step is to improve the removal efficiencies of malathion contaminants from the wastewater samples that were chemically treated at the optimum coagulation doses. The adsorption step is allowed to stirrer at a speed 170 rpm for 180 minutes at fixed temperature of 25°C in the batch process. Series of experiments are using operational parameter conducted to effect of coagulant and adsorbent dose, initial malathion concentration, effect of pH & contact time. After the agitation being stopped, the suspensions were allowed to settle. The samples are filtered to remove fine particles and the concentration of malathion in solution is analysed by using HACH spectrophotometer (TSS), turbidity meter, COD reflux condenser, BOD incubator the results of physicochemical characterisation as shown in table 1.

Table 1:- Wastewater characterisation:

Test	Synthetic waste-water	Industrial waste-water
Turbidity	32 NTU	79 NTU
Total suspended solid	30 mg/l	90 mg/l
Total dissolved solid	62 ppm	425 ppm
pH	6.8	5.88
Conductivity	126.1 $\mu$ s	845 $\mu$ s
Colour	134 Ptco	516 Ptco
Alkalinity	37.20 mg/l	341.7 mg/l
Hardness	1 mg/l	8.5mg/l
C.O.D.	1104 mg/l	1640 mg/l
B.O.D.	350 mg/l	530mg/l
Calcium	0.6 mg/l	5 mg/l
Phosphate	0.08 mg/l	0.26 mg/l
Iron	0.041 mg/l	2.31 mg/l
Molybdenum	4.2 mg/l	10 mg/l
Potassium	0.1 mg/l	1.4 mg/l
Barium	7 mg/l	20 mg/l
Nitraver	6.86 mg/l	18.3 mg/l
Sulphate	0.1 mg/l	72 mg/l
Monochlor	0.3 mg/l	1.8 mg/l

## IV. RESULTS & DISCUSSION

### A. Effect of coagulant- adsorbent dose

The effect of coagulant dose was studied by varying dose in the range of 50 ppm -800ppm at fixed synthetic waste-water Malathion concentration of 500ppm and 480 ppm fixed industrial waste-water followed by effect of adsorbent dose was studied by varying dose in the range of 0.2gm -1.2gm

for DPP & 0.5gm -5gm for BFA at same fixed Malathion concentration of 500ppm. Coagulant-Adsorbent dosage is an important parameter because this factor determines the capacity of coagulant- adsorbent for a given initial concentration of the coagulant- adsorbent. The effect of polyaluminum chloride with polyelectrolytes followed by  $m_{DPP}$  &  $m_{BFA}$  on the uptake of Malathion by DPP & BFA for initial concentration 500ppm is shown in fig. 1. The Malathion removal is observed to increase rapidly with an increase in  $m$  up to about polyaluminum chloride 500ppm with polyelectrolytes fixed dose 0.5 ppm, 0.2 g/500ml DPP & 0.5g/500ml BFA. An increase in the coagulation-adsorption with the coagulant-adsorbent dosage can be attributed to the availability of more coagulation-adsorption sites and greater surface area for contact. Thus, the optimum dose polyaluminum chloride 800ppm with polyelectrolytes 0.5ppm,  $m_{DPP}$  &  $m_{BFA}$  for synthetic waste-water Malathion concentration of 500ppm and malathion industrial waste-water may be taken as 0.2g/500ml & 1 g/500ml. An Integrated method for synthetic waste water by using drumstick peel powder and bagasse fly ash adsorption step then filtered sample of water to test in laboratory results malathion removal with respect to COD, BOD, TSS, Turbidity. They were 73.91%, 91.42%, 93.33%, 93.50% and 63.04%, 88.57%, 90%, 86.62% respectively. Same processes apply for malathion industrial waste water (480ppm) by using drumstick peel powder and bagasse fly ash shown the results malathion removals with respect to COD, BOD, TSS, Turbidity. They were 94.63%, 92.45%, 91.11%, 87.34% and 90.73%, 90.56%, 86.66%, 85.82% respectively. The effect of coagulant-adsorbent dose on removal of Malathion as shown in fig.1, 2, 3 & fig .4.

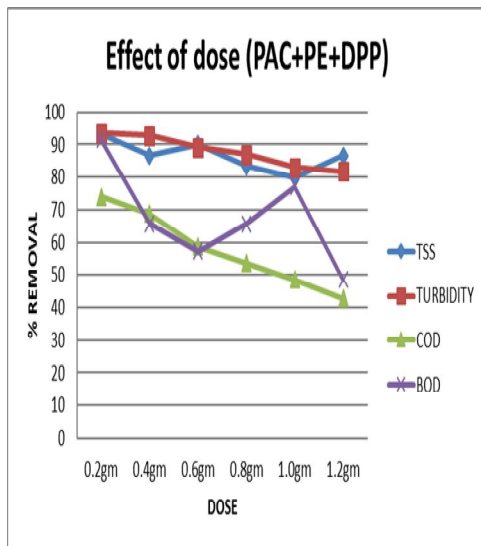


Figure 1.: Effect of dose on the removal of malathion from synthetic waste water by using drumstick peel powder (t = 41 min.+3 hr., PAC Dose = 800ppm,PE Dose=0.5ppm , malathion initial concentration =500ppm at 298 K).

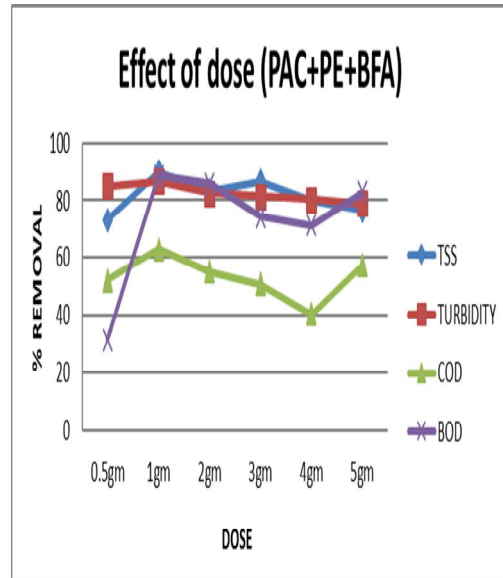


Figure 2.: Effect of dose on the removal of malathion from synthetic waste water by using bagasse fly ash (t = 41 min.+3 hr., PAC Dose =800ppm,PE Dose=0.5ppm , malathion initial concentration =500ppm at 298 K).

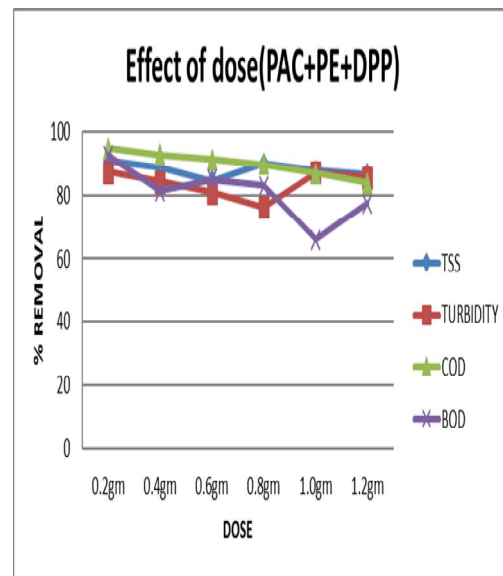


Figure 3.: Effect of dose on the removal of malathion from industrial waste water by using drumstick peel powder (t = 41 min.+3 hr., PAC Dose =800ppm,PE Dose=0.5ppm , malathion initial concentration =480ppm at 298 K).

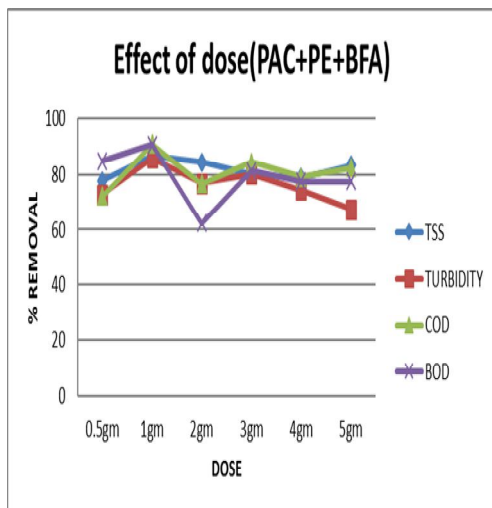


Figure 4.: Effect of dose on the removal of malathion from industrial waste water by using bagasse fly ash (t = 41 min.+3 hr., PAC Dose =800ppm,PE Dose=0.5ppm , malathion initial concentration =480ppm at 298 K).

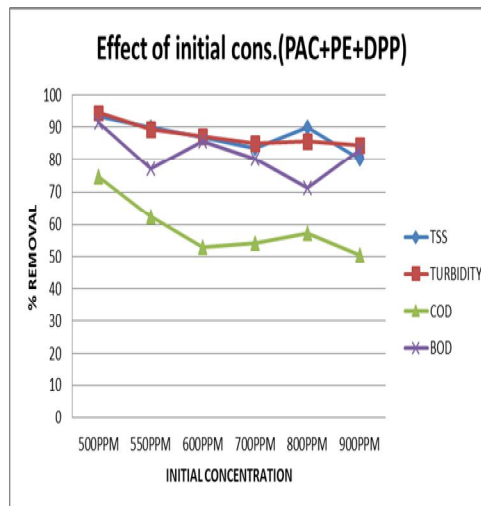


Figure 5.: Effect of initial concentration on the removal of malathion from synthetic waste water by using drumstick peel powder (t = 41 min.+3 hr., DPP Dose = 0.2 gm, PAC Dose =800ppm,PE Dose =0.5ppm at 298 K).

**B. Effect of initial concentration**

The effect of malathion initial concentration (500ppm -900 ppm) synthetic wastewater and (480ppm-900ppm) industrial wastewater at fixed temperature 298 K on the equilibrium uptake of malathion by the polyaluminum chloride 800ppm with polyelectrolytes fixed dose 0.5ppm, DPP at  $m_{DPP} = 0.2g/500ml$  & BFA at  $m=1g/500ml$  and  $t = 41 \text{ min} + 3hr.$  was studied and a plot of the removal of malathion and the sorptive uptake of malathion by coagulation-adsorption versus malathion initial concentration with at fixed temperature An Integrated method for synthetic waste water by using drumstick peel powder and bagasse fly ash adsorption step then filtered sample of water to test in laboratory results malathion removal with respect to COD, BOD, TSS, Turbidity. They were 74.63%, 91.42%, 93.33%, 94.53% and 68.11%, 88.57%, 90%, 87.56% respectively. Same processes apply for malathion industrial waste water by using drumstick peel powder and bagasse fly ash shown the results malathion removals with respect to COD, BOD, TSS, Turbidity. They were 95.60%, 92.45%, 92.22%, 88.53% and 92.19%, 92.45%, 88.88%, 86.83% respectively as shown in fig.5,6,7 & fig .8

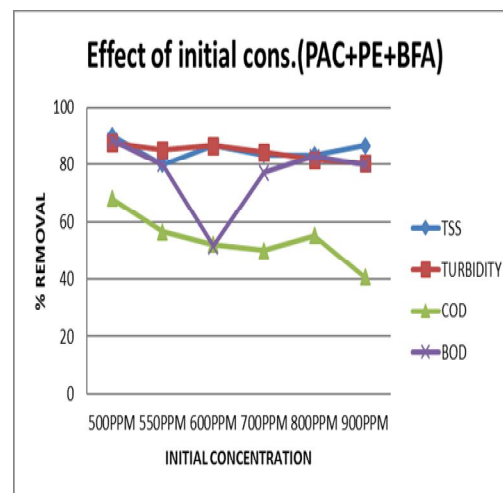


Figure 6.: Effect of initial concentrations on the removal of malathion from synthetic waste water by using bagasse fly ash (t = 41 min.+3 hr., BFA Dose = 1 gm, PAC Dose =800 ppm,PE Dose=0.5ppm at 298 K).



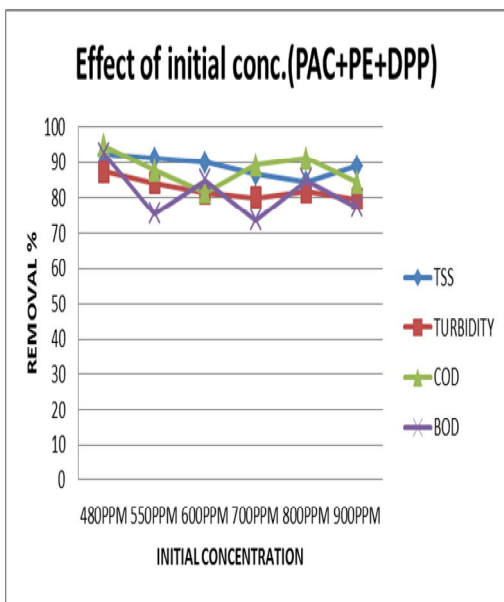


Figure 7.: Effect of initial concentrations on the removal of malathion from industrial waste water by using drumstick peel powder (t = 41 min.+3 hr., DPP Dose = 0.2 gm, PAC Dose =800ppm, PE Dose =0.5ppm at 298 K).

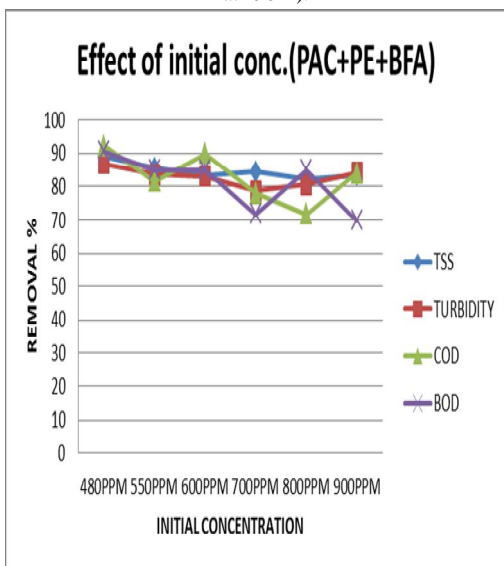


Figure 8.: Effect of initial concentrations on the removal of malathion from industrial waste water by using bagasse fly ash (t = 41 min.+3 hr., BFA Dose = 1 gm, PAC Dose =800ppm, PE Dose =0.5ppm at 298 K).

**C.Effect of initial pH**

The pH value can affect the structural stability of malathion and, therefore, its concentrations. Effects of initial pH range 2-12, on the removal of malathion at fixed 298 K for malathion initial concentration 500ppm (synthetic wastewater) & 480ppm(industrial wastewater), polyaluminum chloride 800ppm with polyelectrolytes fixed dose 0.5ppm,  $m_{DPP} = 0.2 \text{ g}/500\text{ml}$  &  $m_{BFA} = 1\text{g}/500\text{ml}$ . Equilibrium was attained in  $t = 41 \text{ min.} + 3 \text{ h.}$  The maximum affinity to malathion was found at  $\text{pH}_0 \sim 12$ . So, based on the experimental observations, further experiments were performed at  $\text{pH}_0 \sim 12$ . An Integrated method for synthetic waste water by using drumstick peel powder and bagasse fly ash adsorption step then filtered sample of water to test in

laboratory results malathion removal with respect to COD, BOD, TSS, Turbidity. They were 77.08%, 94.28%, 96.66%, 95.06% and 70%, 91.42%, 96.66%, 94% respectively. Same processes apply for malathion industrial waste water by using drumstick peel powder and bagasse fly ash shown the results malathion removals with respect to COD, BOD, TSS, Turbidity. They were 97.07%, 94.33%, 97.77%, 98.46% and 93.65%, 92.45%, 95.55%, 97.05% respectively as shown in fig.9,10,11 & 12.

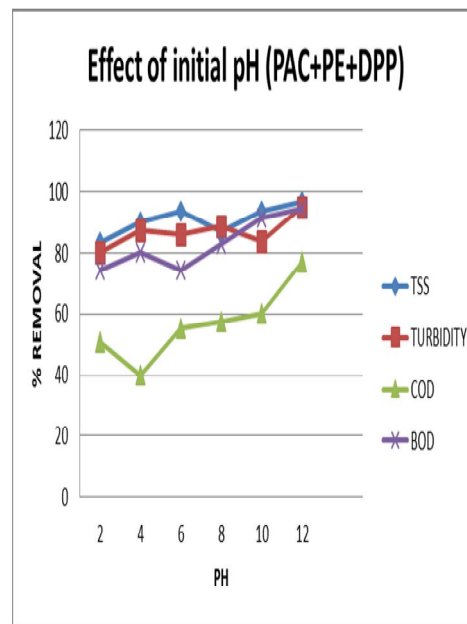


Figure 9.: Effect of initial pH on the removal of malathion from synthetic waste water by using drumstick peel powder (t = 41 min.+3 hr., DPP Dose = 0.2 gm, malathion initial concentration =500ppm, PAC Dose =800ppm, PE Dose =0.5ppm at 298 K)

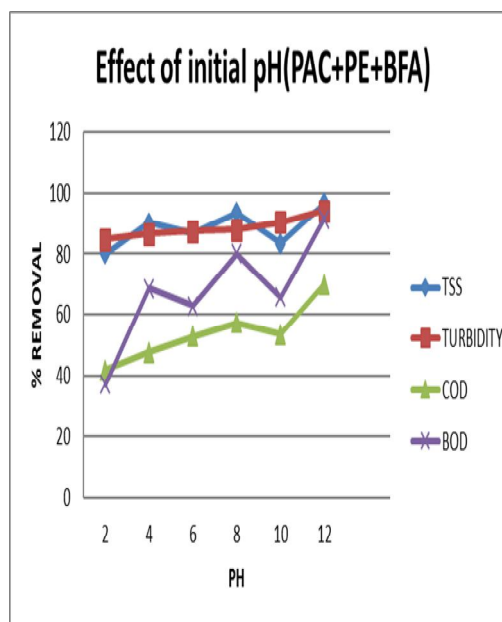


Figure 10.: Effect of initial pH on the removal of malathion from synthetic waste water by using bagasse fly ash (t = 41 min.+3 hr., BFA Dose = 1 gm, malathion initial concentration =500ppm, PAC Dose =800ppm, PE Dose =0.5ppm at 298 K).

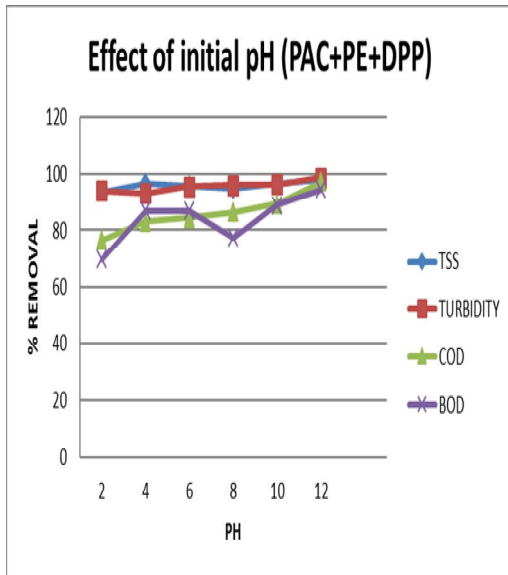


Figure 11.: Effect of initial pH on the removal of malathion from industrial waste water by using drumstick peel powder (t = 41 min.+3 hr., DPP Dose = 0.2 gm, malathion initial concentration =480ppm, PAC Dose =800ppm, PE Dose =0.5ppm at 298 K).

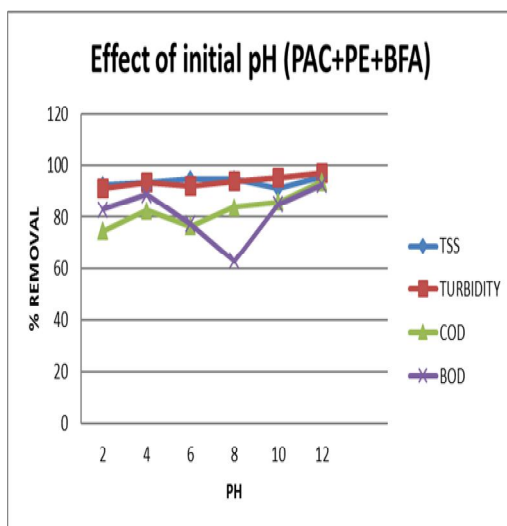


Figure 12.: Effect of initial pH on the removal of malathion from industrial waste water by using bagasse fly ash (t = 41 min.+3 hr., BFA Dose = 1 gm, malathion initial concentration =480ppm, PAC Dose =800ppm, PE Dose =0.5ppm at 298 K).

**D. Effect of contact time**

The effect of contact time on the removal of malathion by polyaluminum chloride 800ppm with polyelectrolytes fixed dose 0.5ppm, DPP for  $m_{DPP} = 0.2g/500ml$  & BFA for  $m_{BFA} = 1 g/500ml$  and malathion initial concentration 500ppm (synthetic wastewater) & 480ppm (industrial wastewater). Initially the coagulation-adsorption rate of malathion onto DPP & BFA is found to be instantaneous because of the availability of more coagulation-adsorption sites. The coagulation process was done then steady state adsorption is assumed after 30 min., 60min., 90min., 120min., 150min. & 180 min. and further experiments were carried out at t=3 h(180 min.). An Integrated method for

synthetic waste water by using drumstick peel powder and bagasse fly ash adsorption step then filtered sample of water to test in laboratory results malathion removal with respect to COD, BOD, TSS, Turbidity. They were 84.05%, 97.17%, 100%, 98.53% and 76.81%, 95%, 96.66%, 96% respectively. Same processes apply for malathion industrial waste water by using drumstick peel powder and bagasse fly ash shown the results malathion removals with respect to COD, BOD, TSS, Turbidity. They were 98.53%, 98.11%, 98.88%, 98.96% and 94.63%, 96.22%, 97.77%, 97.72% respectively as shown in fig.13, 14, 15 & 16.

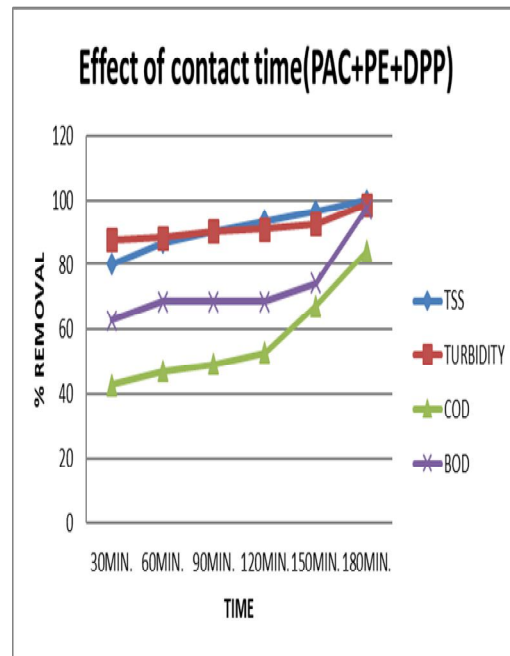


Figure 13.: Effect of contact time on the removal of malathion from synthetic waste water by using drumstick peel powder (t = 41 min.+3 hr., DPP Dose = 0.2 gm, malathion initial concentration =500ppm, pH =12, PAC Dose =800ppm, PE Dose =0.5ppm at 298 K).

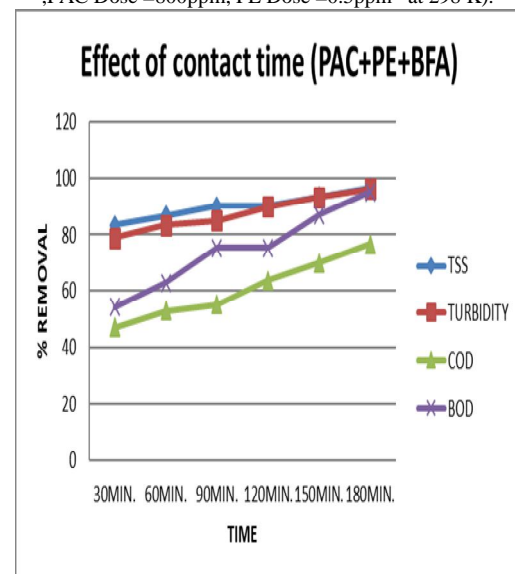


Figure 14.: Effect of contact time on the removal of malathion from synthetic waste water by using bagasse fly ash (t = 41 min.+3 hr., BFA Dose = 1 gm, malathion initial concentration =500ppm, pH =12, PAC Dose =800ppm, PE Dose =0.5ppm at 298 K).

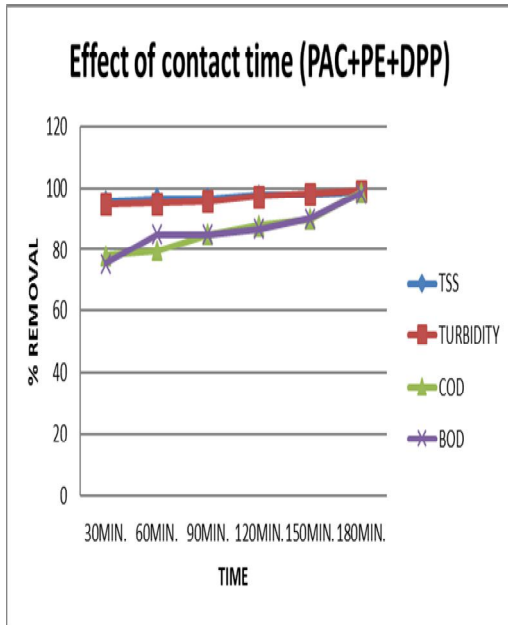


Figure 15.: Effect of contact time on the removal of malathion from industrial waste water by using drumstick peel powder (t = 41 min.+3 hr., DPP Dose = 0.2 gm, malathion initial concentration =480ppm,pH =12 ,PAC Dose =800ppm, PE Dose =0.5ppm at 298 K)

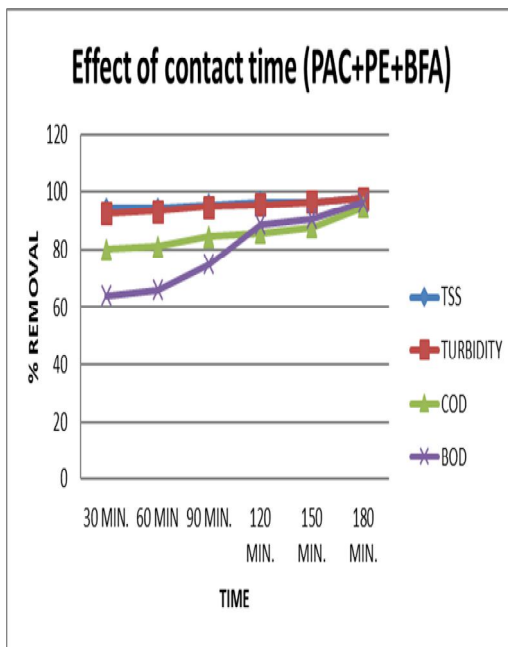


Figure 16.: Effect of contact time on the removal of malathion from industrial waste water by using bagasse fly ash (t = 41 min.+3 hr., BFA Dose = 1 gm, malathion initial concentration =480ppm, pH =12 , PAC Dose =800ppm, PE Dose =0.5ppm at 298 K).

**E. Comparative study:**

Fig .17 and Fig.18 shows the removal of Malathion under optimum condition for coagulation, adsorption and (coagulation-adsorption) integrated synthetic waste –water and industrial waste -water by using drumstick peel powder and bagasse fly ash. It is observed that in both waste-water sample drumstick peel powder more efficient than bagasse fly ash to removal of Malathion.

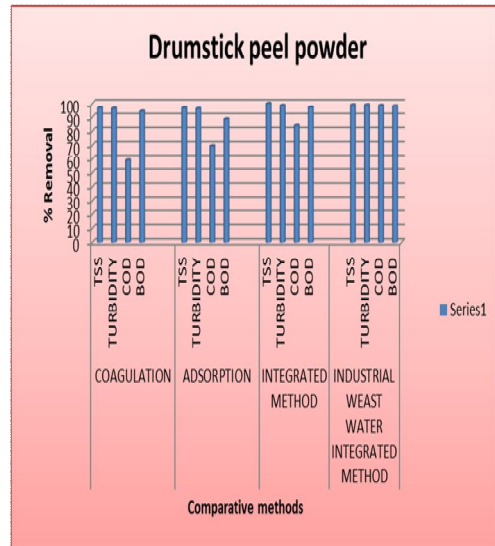


Figure 17.: Comparative studies of methods to using coagulant & DPP

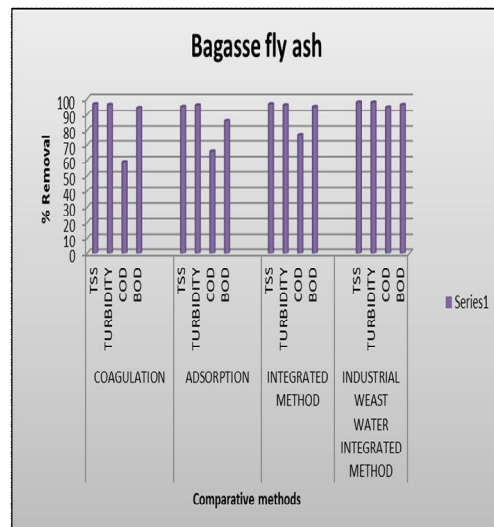


Figure 18.: Comparative studies of methods to using coagulant & BFA.

**V.CONCLUSION**

An integrated system involving chemical coagulation followed by adsorption was developed for the treatment of the synthetic waste water & industrial waste water. An Integrated method for synthetic waste water by using drumstick peel powder and bagasse fly ash adsorption step then filtered sample of water to test in laboratory results malathion removal with respect to COD, BOD, TSS, Turbidity. They were 84.05%, 97.17%, 100%, 98.53% and 76.81%, 95%, 96.66%, 96% respectively. Same processes applied for industrial waste water by using drumstick peel powder and bagasse fly ash shown the results malathion removals with respect to COD, BOD, TSS, Turbidity. They

were 98.53%, 98.11%, 98.88%, 98.96% and 94.63%, 96.22%, 97.77%, 97.72% respectively.

Higher (%) removal of Malathion was observed in low concentration range. The process of removal has been optimising for various parameter affecting the removal. Drumstick peel powder is more efficient than Bagasse fly ash in both synthetic and industrial waste water.

Integrated process is environmental eco-friendly and low cost, low energy requirement. The coagulation-adsorption integrated process is an interesting and growing technology in water purification treatment. The problems associated with the disposal of exhausted adsorbent can be solved either by its incineration or its disposal after proper treatment. The data generated can serve as base line data for designing treatment plant for the treatment of malathion containing effluents/waste-water.

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