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An Experimental Study of Treatment on Tannery Waste Using Trickling Filter

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Abstract- The major public concern over tanneries has been traditionally about odour and water pollution from untreated discharges from tannery industries. This project involves design and fabrication of a biological unit i.e., trickling filter for degradation of organic, inorganic matters and potentially toxic heavy metals. The tannery effluent was collected in and around trichy and physiochemical and biological characterization of the untreated effluent was done. The trickling filter was operated for 45 days continuously to ensure the attachment of bacterial culture to the support media and development of biofilm layer. Bio sorption of heavy metals by metabolically inactive non-living biomass of microbial or plant origin is an innovative and alternative technology for removal of these pollutants from aqueous solution. . Thus, *Bacillus subtilis* is inoculated for the purpose. Initial characteristics of the tannery waste water was performed by analyzing parameters like BOD, COD, TSS, TDS, Turbidity, Carbonates, Bicarbonates, nitrite, fluorides, magnesium, oil and grease and Heavy metal analysis by Atomic Absorption spectroscopy. After the run time, a final characteristic of treated waste helps to analyze the difference between untreated and treated effluent which resulted about 50% reduction in final characteristics study. The Scanning Electron Microscopic (SEM) results indicated a high metal adsorption capacity of charcoal which proved to be an excellent adsorbent. The treated water was subjected to seed germination studies. Hence potentially toxic effluent will be treated for its permissible discharge limits and water obtained may be used as a coolant. The purpose of this project is to review the available information on various attributes of utilization of tannery effluent and explores the possibility of exploiting them for heavy metal remediation and to reduce the toxicity of tannery effluent.

Keywords: Trickling filter, Biosorption, Biofilm layer, *Bacillus subtilis*, Atomic Absorption spectroscopy, Scanning Electron Microscope.

I. INTRODUCTION

A. GENERAL

In general, tannery wastewaters have a high content of organic substances that vary according to their chemicals usage. It also contains enormous amount of chemical substances including toxic heavy metals which are highly harmful to environment. It may contain heavy metals such as lead, copper, iron, chromium, sodium, cadmium, zinc, and nickel. These chemical substances are responsible for contaminating ground water and soil pollution. The presence of heavy metals in the environment is a major problem because of their toxicity, bio- accumulation tendency, threat to human life and the environment. Heavy metals are toxic in nature; their presence even at low level may cases major health issues. It is prime important to treat the effluent by removing the harmful chemical substances, before being disposed into the environment.

B. BIOLOGICAL TREATMENT

Biological treatment arouses great interest because of their lower impact on environment. Then, chemical treatment which produces large amount of secondary waste products due to various reagent added. Acclimatisation refers to exposure of wastewater with the seed or microbial population. In such cases, industrial wastewater do not contain sufficient nutrients, we need to add in to the reactor. In certain cases, special type of micro-organisms is found to cause better biological oxidation if commonly available micro-organism may fail to achieve oxidation. These micro-organism may produces their enzymes which on interact with chemical compound and oxidise it.

C. TRICKLING FILTER

Trickling filter is also called as percolating filter or sprinkling filter. It consists of tanks of coarser filter media, over which the waste is allowed to sprinkle or trickle down, by mean of spray nozzles or rotary distributors. The percolation waste is collected at the bottom of the tank. The decomposition of the organic matter and the resultant purification of waste are brought about by a population of micro-organisms. Micro-organisms and bacteria, which are naturally present in waste and inoculated one, get attached to the filter media. Organic matter from the waste influent is also adsorbed on the biological film, which is formed by the micro-organisms around the filter media. In the outer portions of this film of biological mass or slime layer, the organic matter is degraded by the aerobic bacteria. As the micro-organisms grow, the thickness of the slime layer increases, and the diffused oxygen is consumed by the upper portion of the slime layer, thereby creating an anaerobic environment near the surface of the media particles. As the slime layer increases, the adsorbed organic matter is metabolized before it can reach the micro-organisms near the media face. This creates shortage of external source of organic carbon near the media face, due to which, the micro-organisms near the medium face enter into an endogenous phase of growth and lose their ability to cling to the media surface. The break up or detachment of the biomass from the slime layer is known as sloughing.

D. ECO-FRIENDLY ADSORBENTS

Adsorption is the process of adhesion of atoms, ions, or molecules from a gas, liquid, or dissolved solid to a surface. This process creates a film of the adsorbate on the surface of adsorbent. Some adsorbent have a potential of adsorbing the chemical hazardous compound. Eco friendly adsorbents refers to adsorbents which does not supposed to cause any damages, hardships, discomfort or hazards to the environment. So that it will not disturb biological process of environment. It is different from the non-biodegradable adsorbents because which affects the environment for thousands of year without getting deformation. If it will be a biodegradable adsorbent, it will be decomposed. So ecofriendly adsorbent is only safe for environment. Biodegradable adsorbent used in this projects are as follows

- Coconut shells
- Tamarind seeds
- River pebbles
- Charcoals

E. MICROBIAL INOCULATION

Acclimatisation means exposure of effluent with the seed or microbial population. Tannery waste may normally contain of some bacteria. In addition to these bacteria, we have inoculated *Bacillus subtilis* into it. Bacteria may protect themselves from toxic substances in the environment by transforming toxic compound through oxidation and reduction. The processes by which microorganisms interact with toxic metals enabling their removal are bioaccumulation, biosorption and enzymatic reduction. Thus the treated waste may be disposed with proper disinfection.

II. RELATED WORK

E.Dermou *et al*. [14] have constructed a pilot scale trickling filter and tested for biological chromium (VI) removal from industrial wastewater. Indigenous bacteria from industrial sludge was enriched and used as inoculums for the filter. *Bacillus sp.* was used for transformation of Cr (VI) into Cr (III). The processes by which microorganisms interact with toxic metals enabling their removal are bioaccumulation, biosorption and enzymatic reduction. Three different operating modes were used to investigate the optimal performance and efficiency of the filter, i.e. batch, continues and SBR with recirculation. The latter one was found to achieve removal rate up to 530g Cr (VI)/m^2 d. The low operating cost combined with the high hexavalent chromium reduction rates indicates that this technology may offer a feasible solution to a very serious environmental problem.

M.A.A. Jahan *et al*. [3] have attempted to characterize physiochemical parameters of tannery effluent and investigate the efficacy, and applicability of the biological treatment utilizing aquatic plants macrophytes and algae.

Igwe, J. C. and Abia A.A. [11] suggested the usage of biosorbents such as agricultural by-products, bacteria and fungi as a low cost material instead of using conventional by-products for removing heavy metals.

Haimanot HABTE LEMJI *et al.* [2]

This investigation was to confirm a trickling filter filled with gravel as an alternate biological process over conventional high cost treatment process with regret to nutrient reduction from brewery wastewater. Steady state evaluation of the trickling filter aerobic and anaerobic biofilm system for nutrient removal was made. In trickling filter which are packed bed reactors, the effluent flows downward thus trickling on the surface area of the packed bed parties whereas the organic and nutrients are assimilated by the biomass growing on the packed bed media. This is the basic principle underlying the high water treatability of this biological treatment method. Trickling filter also encourages oxygenation and removal of carbon dioxide from the waste water which is important in the case of using the treating effluent. Filter material consists of lumps of rock approximately sized from 16-32 and 32-64mm (River stone) which were placed over the hollow base on a supporting layer sized from 80-100mm was used as the bio filter. The result obtained reveal that the bioreactor's average efficiency range from 65.46 to 86.59% and from 10.45 to 5.66% for total nitrogen and total phosphorous.

M. Swathi *et al*. [4]

The ground water has been largely polluted due to the growth of tannery industries in India. The present study aimed at studying the effect of tannery effluent which

contained chromium and other pollutants beyond non permissible limits by using cactus powder as an adsorbent.
This paper deals with the removal of chromium and other major pollutants like COD, iron, BOD, TSS, TS, hardness, chloride, ammonia, nitrate, sulphide and sulphate from the tannery wastewater. From the experimental results, it was found that chromium concentration in wastewater was reduced to the level of 0.87 mg/l, COD was removed up to 70%, sulphate removal was found to be at the level of 90 %, BOD was reduced to the level of 70% and iron reduction was at the level of 98% and other pollutants were reduced up to the level of acceptance. Thus it has been proved that the cactus powder can be used as an adsorbent for the treatment of tannery wastewater.

III. METHODOLOGY

A. SAMPLE COLLECTION

Tannery effluent was collected in around trichy. It was collected in can covered with plastic bag where the effluent was collected as a whole from various factories nearer to that area for aeration and treatment

Fig.1 Sample Collection

B. PILOT SCALE TRICKLING FILTER

pilot scale or lab scale trickling filter is the miniature model of actual trickling filter. It is constructed to smaller scale for laboratory purposes. We have fabricated a pilot scale trickling filter with diameter 38.12 cm (1.25 ft) and depth of 30.5 cm (1.00 ft) perforated at the bottom. It consist of high density plastic container as a filter media tank as well the collection tank. The collection tank contains various type of adsorbent materials in the order of coconut shells, tamarind seeds, river pebbles, and finally charcoal. This pilot filter height is typically of a full-scale industrial filter. It is also fabricated with rotary distributor which is used to distribute the wastewater uniformly on the top of the filter media. The rotary distributor consisted of ultra-polyvinyl chloride (UPVC) with perforated rotating arm for uniform and effective distribution of wastewater. The motor which is attached to the rotary distributor causes rotation. The waste is collected in collection tank which is pumped to the rotary distributor by submersible water pump (3ft height). Bearing is provided in between the delivery pipe and rotary distributor in order to avoid twisting effects. Hence the pump and rotary motor should run simultaneously throughout duration of running. It consist of packing material as beds, then effluent flows downward in trickling filter on the surface area of the packed bed materials whereas the biofilm layer is formed by growing of biomass. This operation runtime was continued for 45 days with intermediate tests.

Fig.2 Pilot Scale Trickling Filter

C. ADSORBENT MATERIAL

Adsorption refers to adhesion of molecules on the surface of adsorbent. This process leads to formation of adsorbate layer on the surface of the adsorbent. It adsorbs some chemical compound which is harmful to dispose in environment.

i. COCONUT SHELLS

Coconut has a scientific name of *Cocus indica*. Coconut shells are rigid shell of coconut tree which is inside the fibrous mass of coconut. Coconut shells are first collected and broken into smaller pieces. These pieces are packed in to trickling filter as adsorbents as well as the material which enhances biofilm layer. It is packed at top of the trickling filter as a first layer of adsorbent.

Fig.3 Coconut Shells

ii. TAMARIND SEEDS

Tamarind tree (*Tamarindus indica)* is a leguminous tree in the family Fabaceae indigenous to tropical Africa. It seeds are locally available and abundant in nature. It is usually rigid and consists of absorbing capacities. In our project we provide tamarind seeds as another packing material. It is packed next to top layer of filter tank.

Fig.4 Tamarind Seeds

iii. RIVER PEBBLES

River pebbles are rigid and round in shape, which are collected from sand filter station. During the operation of trickling filter, biomass grows and forms a layer called biofilm layer. Pebbles provide the surface area for the formation of biofilm layer. It is provided in between second top layer and bottom layer.

Fig.5 River Pebbles

iv. CHARCOAL

Charcoal is a lightweight, black residue, consisting of carbon and any remaining ash, obtained by removing water and other volatile constituents from animal and vegetation substances. It is produced by slow pyrolysis. We use the charcoal of *Prosopis juliflora* (Seemai karuvelam) which is harmful for environment and available abundantly in Tamil Nadu. It evacuate ground water table and leads to water scarcity for drinking as well as agriculture. It is packed as the bottom layer of trickling filter.

Fig.6 Charcoals

D. CULTURING AND INOCULATION OF *Bacillus subtilis*

Bacillus subtilis is also known as the hay bacillus or grass bacillus. It is a Gram-positive, catalase-positive bacterium, found in soil and gastrointestinal tract of ruminants and humans. A member of genus *Bacillus, Bacillus subtilis* is rod-shaped, and can form a tough, protective endospore, allowing it to tolerate extreme environmental condition. It was cultured with the culture medium of nutrient broth in a conical flask, and kept in temperature of 26º C in Incubator for 24 hours. Then it is inoculated into the effluent.

Phylum	:	Firmicutes
Class	:	Bacilli
Order	:	Bacillales
Family	:	Baillaceae
Genus	:	Bacillus
Species	:	Bacillus subtilis

Fig.7 *Bacillius subtilis* its Culture medium

E. DISINFECTION AND DISPOSAL OF WASTE

Finally treated water contains inoculated bacteria. Hence it may be harmful to environment and human beings if infected. Treated water was properly disinfected with chlorination and UV treatment and disposed in to waste land.

F. ATOMIC ABSORPTION SPECTROSCOPY (AAS)

Atomic absorption spectroscopy (AAS) is a spectro analytical procedure for the quantitative determination of chemical elements employing the absorption of optical

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radiation (light) by free atoms in the gaseous state. In analytical chemistry the technique is used for determining the concentration of a particular element in a sample to be analyzed. Atomic absorption spectroscopy (AAS) is a widely used technique for determining a large number of metals. In the most common implementation of AAS an aqueous sample containing the metal analyzes is aspirated into an air-acetylene flame, causing evaporation of the solvent and vaporization of the free metal atoms. This process is called atomization. This test conducted in National College in Trichy

Fig.8 Atomic Absorption Spectroscopy

G. SCANNING ELECTRON MICROSCOPE (SEM)

Scanning Electron Microscope (SEM) was used to study the internal structure of adsorbents. It was tested to find out whether adsorbents adsorbed the heavy metals. It also conducted in National College in Trichy

Fig.9 Scanning Electron Microscope (SEM)

IV. RESULT AND DISCUSSION

A. INITIAL CHARACTERIZATION OF TANNERY EFFLUENT

Table 1: Initial Characterization of sample

S.No	Parameter	Result	Permissible limits
1.	Turbidity NTU	256	5
2.	Total solids (mg/l)	2486	500-1000
3.	Total dissolved solids(mg/l)	2355	2100
4.	Total Suspended Solids	393	600
5.	pH	8.59	5.5-9
6.	Electrical conductivity (dsm^{-1})	3.68	1.5
7.	BOD (mg/l)	1985	30
8.	COD (mg/l)	1325	250
9.	Bi Carbonate (mg/l)	546	200
10.	Chloride (mg/l)	1486	600
11.	Sulphate (mg/l)	209	12
12.	Phosphate (mg/l)	0.158	5.0
13.	Sodium (mg/l)	1119	350
14.	Oil & Grease (mg/l)	0.239	10
15.	Colour	>1hue	Colourless
16.	Odour	Unpleasant	Odourless

Sample of tannery effluent was collected and tested for initial characteristics which are tabulated above. It had higher values in various parameters, so that these effluents must be properly treated before it being disposed.

B. ATOMIC ABSORPTION SPECTROSCOPY (AAS)

i. CHROMIUM (Cr)

Sample ID	Signal Abs	Rsd %	Conc mg/L	Corrected Conc mg/L
Cr Blank	-0.001	48.9	0.0000	
Cr Standard 1	0.057	0.7	5.0000	
Cr Standard 2	0.117	0.9	10.0000	
Cr Standard 3	0.182	1.6	15.0000	
Cr Sample Blank	0.068	14.0	5.9102	0.0000
Cr Tannery waste	0.155	9.9	13.0050	7.0947

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ii. COPPER (Cu)

Sample ID	Signal Abs	Rsd %	Conc mg/L	Corrected Conc mg/L
Cu Blank	-0.000	45.9	0.0000	
Cu Standard 1	0.036	0.8	1.0000	
Cu Standard 2	0.108	0.5	3.0000	
Cu Standard 3	0.179	0.1	5.0000	
Cu Sample Blank	0.000	>99	0.0020	0.0000
Cu Tannery waste	0.001	5.7	0.0230	0.0210

iii. LEAD (Pb)

Sample ID	Signal Abs	Rsd %	Conc mg/L	Corrected Conc mg/L
Pb Blank	-0.001	17.9	0.0000	
Pb Standard 1	0.054	1.6	5.0000	
Pb Standard 2	0.107	1.2	10.0000	
Pb Standard 3	0.162	0.3	15.0000	
Pb Sample Blank	-0.007	7.0	-0.4580 C	0.0000 C
Pb Tannery waste	-0.008	75.6	-0.5802 C	-0.1221 C
Pb Tannery waste	-0.005	11.7	-0.2931 C	0.1650 C

As the tannery effluent contains chromium (13.005 mg/l), copper (0.21 mg/l), and lead (0.165 mg/l) which are higher than permissible limits (2 mg/l), (0.1mg/l), (0.1 mg/l) respectively. These harmful heavy metals must be treated properly to ensure environmental safety.

C. INTERMEDIATE TESTS

Table 2: Intermediate tests

S.No	Parameter	Result	Permissible limits
PHYSICAL PARAMETER			
1.	Colour	>1hue	Colourless
2.	Odour	Agreeable	Odourless
3.	Turbidity (NTU)	217	50 – 100
4.	Temperature	34 ºC	45 ºC
5.	Total solids(mg/l)	2045	500-1000
6.	Total dissolved solids(mg/l)	2022	2100
7.	pH	7.34	5.5 – 9
8.	Electrical conductivity	3.16	1.5
9.	BOD(mg/l)	1584	100
10.	COD(mg/l)	897	250
11.	Oil & Greases(mg/l)	0.18	20
12.	Phenolic Compound(mg/l)	0.36	5.0
ANIONS			
13.	Bi Carbonate(mg/l)	186	200
14.	Chloride(mg/l)	1383	600
15.	Sulphate(mg/l)	189	12
16.	Phosphate(mg/l)	0.142	5.0
17.	Fluoride(mg/l)	5.39	15
CATIONS			
18.	Calcium(mg/l)	239	200
19.	Magnesium(mg/l)	142	3
20.	Sodium(mg/l)	356	350
21.	Potassium(mg/l)	0.22	2
HEAVY METALS			
22.	Zinc(mg/l)	0.180	15
23.	Copper(mg/l)	0.019	3.0
24.	Chromium(mg/l)	8.150	2.0
25.	lead(mg/l)	0.130	1.0

D. SCANNING ELECTRON MICROSCOPE

For evaluating the characteristics of adsorbent elements, SEM (Scanning electron microscope) and EDX (Energy-dispersive X-ray spectroscopy) are useful analytical equipment. Scanning Electron Microscope images of charcoal clearly states its adsorbent capacity. In this SEM images, internal anatomy of charcoal is indicated. It also visualised the formation of bio-film layer on the surface of adsorbents. In addition to bio-film layer, it also represents

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Adsorbed materials evidently. Thus it proved to be a better adsorbent. EDX (Energy-Dispersive X-ray spectroscopy) results clearly indicate that charcoal consists of mainly Ca, Cl, C and K small amounts of, Mg, Na, and Si.

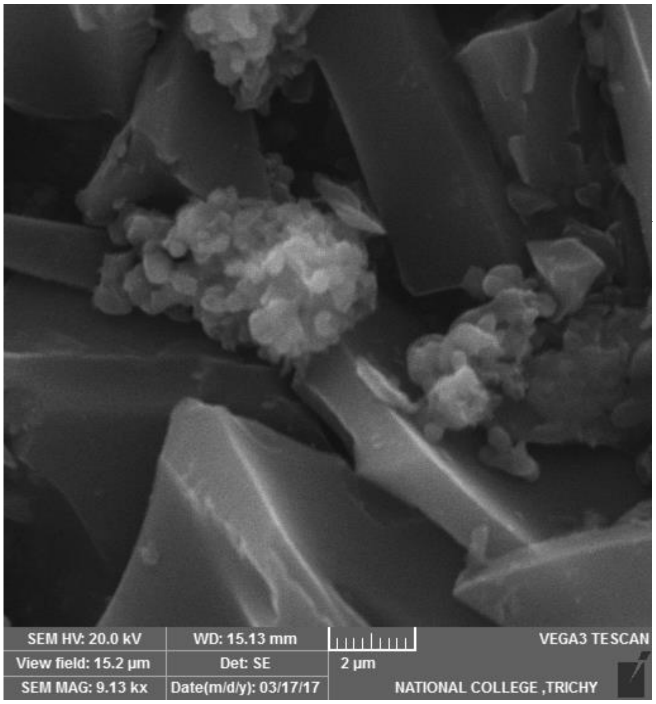


Fig 10. SEM image showing Adsorbed Elements in Charcoal

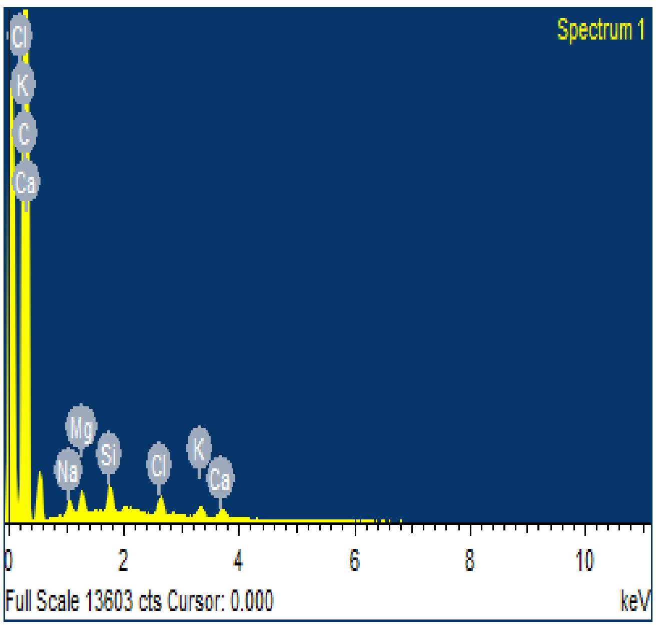


Fig 11. EDX (Energy-Dispersive X-ray spectroscopy)

E. FINAL TESTS

Table 3. Final tests

S.No	Parameter	Result	Permissible limits
PHYSICAL PARAMETER			
1.	Colour	>1hue	Colourless
2.	Odour	Agreeable	Odourless
3.	Turbidity (NTU)	85	50 – 100
4.	Temperature	34ºC	45 ºC
5.	Total solids(mg/l)	734	500-1000
6.	Total dissolved solids(mg/l)	604	2100
7.	pH	5.6	5.5 – 9
8.	Electrical conductivity	1.71	1.5
9.	BOD(mg/l)	732	100
10.	COD(mg/l)	415	250
11.	Oil & Greases(mg/l)	0.06	20
12.	Phenolic Compound(mg/l)	0.18	5.0
ANIONS			
13.	Bi Carbonate(mg/l)	128	200
14.	Chloride(mg/l)	392	600
15.	Sulphate(mg/l)	126	12
16.	Phosphate(mg/l)	0.13	5.50
17.	Fluoride(mg/l)	5.89	15
CATIONS			
18.	Calcium(mg/l)	148	200
19.	Magnesium(mg/l)	191	3
20.	Sodium(mg/l)	319	350
21.	Potassium(mg/l)	0.32	2

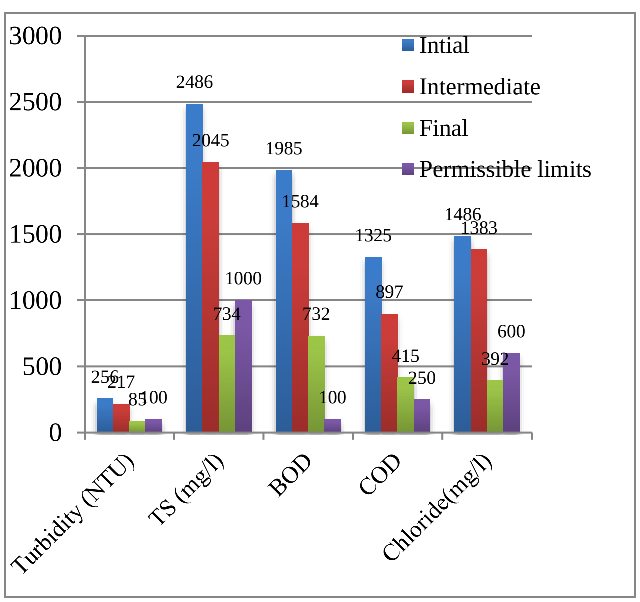
HEAVY METALS			
22.	Zinc(mg/l)	0.15	15
23.	Copper(mg/l)	0.016	3.0
24.	Chromium(mg/l)	0.59	2.0
25.	lead(mg/l)	0.12	2.0

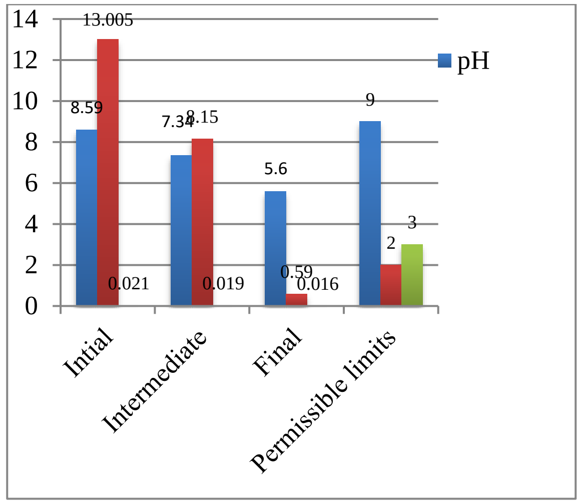
F. GERMINATON TEST

Germination test refers testing the growth phase of the seedlings. Germination study further indicates the unsuitability of the treated effluent is not recommended for agricultural purposes but can be used for other purposes like coolants in industries, etc. This aspect needs to be investigated further in order to promote it for agricultural purposes.

Fig 12. Germination tests

G. COMPARITIVE STUDIES





V. CONCLUSION

The results revealed that the waste water was effectively treated to 50% removal of all physical, chemical parameters. The Biological Oxygen Demand (BOD), Chemical Oxygen Demand (COD), and Chromium content of tannery effluent initially are 1985mg/l, 1325mg/l and 13.005mg/l respectively. The effect of co-ions on adsorption of Cr (VI) was studied in SEM-EDX, which showed the maximum adsorption of cations and anions. 45L of tannery effluent were treated to reach the chromium (VI) standard (0.1 mg L^{-1}) established by the Central Pollution Control Board (CPCB). The results revealed that the trivalent chromium is significantly adsorbed on activated carbon and the method could be used economically as an efficient technique for removal of Cr (III) and purification of tannery wastewaters. The BOD, COD, and Chromium content of treated effluent reduced to 732mg/l, 415mg/l and 0.59mg/l respectively. Germination study further indicates the unsuitability of the treated effluent is not recommended for agricultural purposes but can be used for other purposes like coolants in industries, etc. This aspect needs to be investigated further in order to promote large-scale use of non-conventional adsorbents. In spite of the scarcity of consistent cost information, the widespread uses of low-cost adsorbents in industries for wastewater treatment applications today are strongly recommended due to their local availability, technical feasibility, engineering applicability, and cost effectiveness. If low-cost adsorbents perform well in removing heavy metals and other parameters at low cost, they can be adopted and widely used in industries not only to minimize cost inefficiency, but also improve profitability.

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