TREATMENT METHODOLOGY WITH AMMONIA RECOVERY OF DYES AND PIGMENT MANUFACTURING INDUSTRIAL WASTE-WATER- A REVIEW

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ABSTRACT - *Conventional primary-secondary biological treatment has only a limited capacity for removing organic and inorganic nitrogenous compounds from dye's and pigment manufacturing industrial waste water having high NH_3–N of 5712mg/L and COD of 42000mg/L. This type of waste water required some advanced treatment methodology. There also have a specific opportunity to remove nitrogen from waste water. This may be desorption or striping of free molecular ammonia at high PH. And recovered ammonia can be used for N-Based fertilizer production like ammonium sulphate and ammonium chloride. Flocculation in chemical treatments by NAOH or Ca(OH)2, increasing the PH has provided a most favourable condition for ammonia stripping. And then coagulation is done by chemicals with Ferrous Sulphate, Ferric Chloride, Polly-Aluminium Chloride and Fenton's Reagent and find an optimal and economic treatment method. This paper reviews the possibility of an economic and feasible methodology for treatment of dyes and pigment manufacturing industrial waste water and paper reviews the possible option for recovery of some useful components from waste water during chemical treatments and process efficiency by varying operating parameters like PH, Temp and dosing amount and finding a best way of treatment scheme with recovery of ammonia. Main pollution parameters are TDS, COD and Ammonia.*

KEY WORDS:- **Ammonia, Stripping, chemical oxygen demand (COD), Total dissolve solids (TDS), Iron iii Chloride, Coagulation-Flocculation, ammonia recovery, PH and Temperature.**

1. INTRODUCTION

Dyes and pigments are substances which when applied to a substrate Lead to selective reflection or transmission of incident daylight [1-2]. Substances which create the sensation of blackness or whiteness are also regarded as dyes or pigments. Colorants, not defined by this definition of dyes and pigments are called dyestuffs. Synthetic organic dyestuffs and pigments exhibit an extremely wide variety of physical, chemical and biological properties, making review of the Eco toxicological behaviour of the several thousand commercially available products difficult, production process are difficult to control the release of manufacturing material during dye manufacturing [3-15]. In general, the most losses of material during manufacturing process are intermediates that do not react and by-products such as incomplete dye or pigment molecules. The intermediates and by-products from the dye manufacture are present either in the product or in the solid waste streams regulated under RCRA (Conservation and Recovery Act), which include wastewater, solid residue, and the vapour and dust emissions. The losses products discharged in the plant effluent and some is discharged on any waste water treatment solids. The dyes and pigment present in water is undesirable. So a very small amount of theses colouring agents are very highly visible and may be toxic to the aquatic environment. It is well known that dye effluents be toxic and/or carcinogenic to mammalian animals [16-35]. The toxic behaviour waste streams are also described by many author's that it effects the ecosystem but also affects the human life that's by it necessary to treat before discharging or not to be re-used in process due to high COD ,TDS ,TSS,NH_3-N and complex mixtures of by products and intermediates have undesirable effects. So be needed a practical solution which is compatible environmentally as well as economic aspects. However, the preparation of the first synthetic dyestuff, Mauveine (13), by Perkin in 1986 gave birth to the new many other important sectors of the modern chemical industry. In comparison it is seen that synthetic dyes are better than natural Colorant in stability, fastness and many properties. Now due to their importance and there application's they are irreplaceable in our life (traffic lights and signs, drug identification, control systems) [3].

Here we seen the data given below in the table 1 and table 2 that shows highest per cent of aromatic amines, aromatic amines, nitrogen based row material is used and there for highest amount of ammonical nitrogen and aniline based waste is generated during dyes and pigment manufacturing .where aniline, ammonia and N-based organic waste is very toxic and hazardous in nature .there also several other by product like dyes and dyes intermediates colour compound generated waste produced during manufacturing process.

2. PROCESS APPROACH

The purpose of this review has to reduce the COD, TSS and neutralization of waste to dispose it in a stable form, Removal of colour and recovery of use full component like ammonia present as a waste during treatment procedure. There are several methods available to treat waste water such as coagulation-flocculation, biological, tertiary by sand charcoal filter, adsorption methods, adsorption by charcoal, electrochemical treatment, electrocoagulation, ozonation, evaporation followed by incineration process, photo catalysed hydrogen peroxide oxidation treatment process and some specific advanced treatment processes are available now these days. Microbial conversion of Nitrogen based organic waste present in waste water to nitrate followed by microbial conversion of nitrates to nitrogen gas referred as biological nitrification-DE nitrification. But at industrial level on large capacity financial aspects, treatment effectiveness and environmental concerns describe the feasibility and effectiveness of process. Evaporation followed by incineration is last option if none of the method works on toxic waste water. High COD disabled the evaporation and toxic behaviour disabled the biological treatment. So here an advance treatability study is required which also economic and environmentally feasible.

3. CONVENTIONAL METHODS AND PRIVIOUS WORKS

The conventional techniques adopted to treat the wastewater are physical, chemical and biological methods.

1. Physical – Sedimentation (Clarification), screening, aeration, Filtration, Flotation and skimming, degasification, Equalization.

2. Chemical – Chlorination, Ozonation, Neutralization, Coagulation, Adsorption, Ion exchange.

3. Biological a. Aerobic – Activated sludge treatment methods, Trickling filtration, oxidation, ponds, lagoons, aerobic digestion b. Anaerobic – Anaerobic digestion, septic tanks, Lagoons The treatment process can be divided as:

I. Primary treatment process – Removal of suspended solids, oil, grit etc.

II. Secondary treatment process – Use of microorganisms in either aerobic or anaerobic condition for the reduction of the BOD and removal of colour and oil.

III. Tertiary treatment process – Use of electro dialysis, ion exchange and reverse osmosis for the final removal and purification of the wastewater.

The dyes and textile industry effluents which are treated with various physical and chemical treatments such as coagulation, flocculation, ozonation and biological treatment and nitrogen, phosphorous, organics and metal traces were removed and sludge is discharged. Then the main problem is of sludge disposal for which space is needed. According to A E Ghaly et al. [36] volume-5 1000182, disadvantages of the biological processes are the presence of the toxic heavy metals which are harmful to the growth of microorganisms, most of the dye stuff used is non -biodegradable in nature and the time requirement for the treatment is more.

3.1PRIMARY TREATMENT PROCESS

By Das S 2000 [37] the first step is the removal of suspended solids, excessive quantities of oil, grease and gritty materials. The effluent is first screened for coarse suspended materials such as yarns, lint, pieces of fabrics and rags using bar screens and fine screens.

This paper reviews the work on dyes and pigment manufacturing industrial waste water producing the following dyes products and row materials given in table 1 and table 2

Table no 1.

Sr no	Name of the dyes and pigment manufacturing industrial product	Qt. MT/month
A	Powder dyes (solid)	
1	Basic dyes such as	
	Rhodamine	
	Malachite green/brilliant green	
	Methyl violet	
	Auramine	
2	Solvent dyes such as	75
	Rhodamine base	
	Victoria blue base	
	Bismark brown base	
	Orange base	
	Solvent black	
B	Liquid dyes such as	
	Rhodamine liquid	
	Malachite green liquid	
	Victoria blue liquid	120
	Bismark brown /chrysiodine liquid	
	Methyl violet/ crystal violet liquid	
	Basic yellow liquid	
	Total	195
C	By product	6
	Aniline derivatives	
	Ammonia	
	N-Based organic compounds	
	Dyes and colour complex	
	Organic acids	
	Phenol derivatives	
	Benzene derivatives	

Table no 2

sr	Name of row material	Physical state	QT MT/month
1	Pthalic anhydride	Solid	10.00
2	Diethyl meta amino phenol	Solid	8.00
3	Dimethyl aniline/Mona methyl aniline	Liquid	20.00
4	Formaldehyde		5.00
5	Benzeldehyde	Solid	5.00
6	Meta phenyline diamine meta tolludine diamine	Solid	3.00
7	PANA	Solid	1.00
8	Sodium nitrite	Solid	1.00
9	Caustic soda	Solid	5.00
10	Hydro chloric acid	liquid	8.00
11	Sulphuric acid	Liquid	5.00
12	Acetic acid	Liquid	15.00
13	catalyst and surfactants	Solid	0.75

3.2COAGULATION/FLUCULATION

3.2.1COAGULATION

Neutralization of negative charge by higher valance cat-ion salt (Al+3, Fe+3) results in destabilisation of colloidal present in waste water. Coagulant dose are used to reduce the electrostatic repulsive forces by the addition of counter charged ions. Coagulants used are alum, Bentonite Clay, Poly aluminium Chloride (PAC), Poly aluminium Hydrochloride, Aluminium Chloride, aluminium Chlorohydrate, aluminium Sulphate, Ferric Chloride, Ferrous Sulphate Monohydrate, hypochlorite, $H_2 O_2$ /Fe2+ catalysed, ammonium sulphate, ammonium per sulphate, $FeCl_3$, aluminium sulphates. According to literature “Coagulants for Optimal Waste water Treatment from bekart environmental” [38] sulphate and chlorides of iron and aluminium are some most commonly used chemical coagulants used for coagulation processes.

3.2.2 FLUCCULATION

Flocculation is the agglomeration of destabilizing particles in to a large size particle known as flocks .which can be remove by sedimentation or floatation. Flocculation process enhance simply by adding neutral electrolytes like NACL and KCL in small amount that are able to reduce the zeta potential of suspended partials to zero and sufficient to induce flocculation of weakly charged, water insoluble, organic non-electrolytes such as steroids. In case of more highly charged, insoluble polymers and polyelectrolyte species, such as Ca-salts and alums or sulphates, citrates and phosphates are usually required to achieve flock formation depending on particle charge, positive or negative out of these we use Polly Electrolyte.

Mechanism: There are two major forces acting on colloids

1. Electrostatic repulsion

2. Intermolecular or van der Waals attraction force.

Some common Flocculants are NAOH, $NAHCO_3$, $CA(OH)_3$, lime, acryl amide, acrylic acid etc.

Harsha p. shrivastava et al. 2011[39] and Deepa chandran 2016 2229-5518 [40] studied and carried out various experimentation on master composite effluent neutralised with hydrated lime. Various types of flocculants and coagulants were tried on the effluent and separate out the sludge formed sludge formed % reduction of COD, TDS, EC and Colour was measured. Can-Zeng Liang et al. 2014 [41] performed experiments for the treatment of highly concentrated multiple dyes wastewater (MDW, 1000 ppm), poly aluminium chloride (PAC) and polydiallyldimethyl ammonium chloride (PDDA) were found to be the most effective coagulant and flocculent. The CF process can achieve about 90% of dye removal at the optimal dosage of PAC/PDDA=400/200 ppm, and the MDW with pH>3 is favourable for the coagulation flocculation treatment. Tak-Hyun Kim et al. 2003 [42] work on the artificial addition of electrolyte solution and to decrease the pollutant loading efficiently on the post electrochemical oxidation process in order to improve the performance of organics removal by PAC and FeCl3, Cl-based chemical coagulants, was found as successful step of electrochemical oxidation. And it seems that PAC and FeCl3 were able to achieve sufficient removal efficiency of organics as well as to exclude the artificial addition of a supporting electrolyte and chloride reactant. D. georjius et al. 2003 [43] and Sheng H. Lin 1996 [44] experimented on Coagulation/Flocculation by Ferrous Sulphate and lime. All the experiments were carried and simulated an actual industrial wastewater treatment plant. Treatment with lime alone proved to be very effective in removing the colour (70–90%) and part of the COD (50–60%) from the textile and dyes industrial wastewater. Moreover, the treatment with ferrous sulphate at maintained pH in the range 9.0±0.5 using lime was equally effective. S. Sadri Mmoghaddam et al. 2010 [45] analyse the results on experimentation that the decrease of initial pH was always beneficial for dye removal and no re-stabilization phenomenon was occurred even at the used maximum FCS (FERRIC CHLORIDE SLUGE) dosage. Also it seems that iron hydroxides of the FCS could neutralize the negative charges on dye molecules. Therefore, the sweep flocculation and/or the charge neutralization might play key roles in the enhancement of dye removal. The optimum initial pH, FCS dosage and initial dye concentration were found to be 3.5; 236.68 mg dried FCS/L and 65.91 mg/L, respectively. Dye removal of 96.53% is observed which confirms close to RSM (REFERENCE SURFACE METHODOLOGY) results.

3.3 ELECTROCOAGULATION

As water passes through the electrocoagulation cell which consists of electrode multiple reactions take place simultaneously. First a metal ion is driven into the water. At electrode surface water dissociate into hydrogen gas and hydroxyl groups. Meanwhile, freely flow electrons destabilize surface charges on suspended solids and emulsified oils. As the reaction proceeds further, large flocks form that entrain suspended solids, heavy metals, emulsified oils and other contaminants. Finally, the flocks are removed from the water in downstream solids separation and filtration process steps. Gianluca Ciardelli and Nicola ranieri 2001 [46] studied and analyse that electrocoagulation process efficient for removing colour (80–100%) and chemical oxygen demand (70–90%).

3.4 OZONATION

Ozone acts as a powerful oxidizing agent due to the reaction,

$$O_3 \longrightarrow O_2 + [O]$$

The nascent oxygen formed due to decomposition of ozone is responsible for the oxidation of a number of substances. The combination of ozone oxidation followed by biological treatment has been installed full-scale at a large German industrial chemical complex. Rip G. Rice 1996 [47] experimented on Ozone coupled with ultraviolet radiation and/or hydrogen peroxide (advanced oxidation) is being utilized to destroy organic contaminants in ground waters at munitions manufacturing plants and at Superfund sites (hazardous wastes).

3.5 FENTONS TREATMENT

In Fenton's method oxidation is carried out by hydrogen peroxide in presence of Fe^{2+} catalyst at optimum PH of 4-4.5 gives maximum efficiency of 98% COD reduction. Further development is done by photo catalysed Fenton process which carried out in presence of light/UV Light results in maximum efficiency of COD reduction of industrial waste water. Putri F. Khamaruddin et al. 2011 [48] work on amines treatment by Fenton's reagent and it was found to be a suitable chemical treatment method. Change the perimeter like temperature and Ph. Thus for this study, the experimental ranges for temperature and pH: 30°C ≤ T ≤ 60°C and 2 ≤ pH ≤ 4. The molar ratio of reagents, H2O2:Fe2+ used was 95. Amit Kumar Tiwari and Vijay Kumar Upadhyay 2013 [49] experimented treatment of high COD waste water by Fenton process successfully. theoretically calculated H2O2 doses are higher and are neither cost effective nor shows good reduction in COD value when used alone so efficiency increased by adding ferrous for up to 90% COD reduction OF waste effluent.

4 SECONDARY TREATMENTS

Biological treatment processes is applicable only if nature of waste is organic in nature which can be converted to a simple non-toxic and stable By-products by biological degradation process using bio-culture and urea for bacterial growth. Ibrahim M. Banat et al. 1996 [50] has made attention to words the ability of microorganisms to carry out dye manufacturing industrial waste water after primary treatment for further waste reduction in BOD and de-colorization. Microbial de-colorization and degradation of dyes is seen as a cost-effective method for removing these pollutants from the environment. Work has revealed the existence of a wide variety of microorganisms capable of decolorizing an equally wide range of dye. Harpreet Singh Rai et al. 2005 [51] work on Biological removal of dyes from effluents of textile and dyestuff manufacturing industry. It offers some distinct advantages over the commonly used chemicals and physicochemical methods. They also show possible mineralization of the dyes to harmless inorganic compounds like carbon dioxide and water, and formation of a lesser quantity of relatively harmless sludge. Secondary treatment removes 85 to 95 % of BOD and TSS and minor portions of nitrogen, Phosphorus and heavy metals. But biological treatments are rarely applicable in dyes manufacturing industrial waste water due to the toxic nature and bacterial growth is not possible.

5 TERTIARY TTREATEMENT

Tertiary treatment and its purposes best described by B Ramesh Babu et al. 11(2007) [52] purpose of tertiary treatment is to provide a final treatment stage to raise the effluent quality Before it is discharged to the environment or recycled to the process . Raise the treated water quality to such a level to make it suitable for reuse. This step removes different types of pollutants such as organic matter, SS, nutrients, pathogens, and heavy metals. It includes sedimentation, membrane processes, filtration, ion exchange, activated carbon adsorption, electro dialysis, nitrification and di-nitrification etc. further the membrane processes can be classified into sub-processes such as electro dialysis (ED) or electro dialysis reversal (EDR), microfiltration (MF), ultrafiltration (UF), Nano filtration (NF), reverse osmosis (RO) some of them described below.

ADSORPTION AND SAND FILTERATION

Adsorption is physiochemical method used in wastewater treatment, which can mix the wastewater and the porous material powder or granules, such as activated carbon and clay, or let the wastewater through its filter bed composed of granular materials. Commonly used adsorbents are activated carbon, silicon polymers and kaolin. Different adsorbents have selective adsorption of dyes. But, so far, activated carbon is still the best adsorbent of dye wastewater. It can be removed 92.17% and COD can be reduced 91.15% after primary or secondary treatment. We use activated carbon in sand charcoal filter. Evangelos

Diamadopoulos and Christos Vlachos 1996 [53] studied and founded the effectiveness of sand carbon filtration in ferric chloride coagulation process. They find that the filtration by sand carbon filter does not required any external electrolytes if we using PAC or Ferric Chlorite as a coagulant and gives better results.

6 ADVANCE TREATMENTS

A advance treatment by M. I. Aguilar et al. 2005 [54] physicochemical treatment (coagulation–flocculation) was applied to a slaughterhouse wastewater, using anionic polyacrylamide as coagulant aid to improve the settling velocity of the flocks in Ferric Sulphate, Aluminium Sulphate and PAC treatment. By Sheng H. Lin and Ming L. Chen 1997 [55] on the enhancement of efficiency of electrochemical method, addition of a small amount of hydrogen peroxide is found to be highly beneficial. Idil Arslan-Alaton et al.2009 [56] analysed Degradation by electro-Fenton method gives 60-80% at near 3 PH and iron catalysed provided with suitable electrodes. Treatment by photo Fenton like advance treatment methods gives 98% colour, 78% COD and 59% TOC removal.

6.1CATALYTIC WET AIR OXYDATION PROCESSES

L. Oliviero et al. 2003 volume-40 [57] described the treatment of toxic nitrogen-containing compounds are best oxidised by Wet Air Oxidation (WAO) processes. The Catalytic Wet Air Oxidation (CWAO) of these nitrogenous compounds, mainly produced in chemical and pharmaceutical industries with oxidation of aniline because it was so toxic in nature, often chosen as a best method applied in dye manufacturing industries. Discus the selectivity towards organic by-products (specially, azo, nitroso and nitro compounds, phenolic compounds and carboxylic acids) as well as towards several inorganic forms of nitrogen (NH_4^+, N_2, NO_2^-, NO_3^-) here Ammonia is one of the most refractory by-products formed during catalytic WAO process by the nitrogen-containing organic pollutants and is itself a pollutant. Catalyst used are generally iron aluminium high valance ion and The oxidation of aniline was proposed by catalytic oxidation in presence of wet air the effectiveness enhancement was recorded by L. Oliviero et al. 2003 volume-81 2003 [58].

6.2 TRETMENT PROCSSES WITH AMMONIA RECOVERY

From environmental aspect purification of ammonia containing wastewater is expected. High efficiency ammonia desorption can be done from the water by air on proper temperature and PH. After the desorption process, ammonia can be recovered and used in another technology. M. Orvos et al. vol 2 2008 [59] verified the desorption of ammonia from waste at different condition of temperature and PH high PH ammonia present in free molecular state and can be recovered /removed easily from high ammonia containing dyes and pigment manufacturing industrial waste water. Steam and air stripping Recovery of ammonia: desorption of ammonia from industrial, municipal waste water streams and biogas plant contains high conc. of ammonium nitrogen is stripped with air by providing alkaline medium.

Absorption of ammonia from waste gases: from waste gases ammonia gas is absorbed because of its high solubility at low PH and efficiency enhanced by lowering the saturation temp.

Biological conversion: biological conversion by some nitrifying plant and bacterial conversion into direct nitrogen and nitrogen based bio fertilisers is possible at low concentration because at high conc. ammonia shows toxic behaviour and it is impossible condition for bacterial growth. In case of dyes manufacturing industries there also many other toxic compounds like aniline are present.

Some previous research works on ammonia recovery are: by I. Çelen & M. Türker "Recovery of Ammonia as Struvite from Anaerobic Digester Effluents" in 2010 [60]. By Lorenzo Liberti et al 1979 ammonia recovered from secondary effluents using selective ion exchange method with production of a slow-release fertilizer as $MgNH_4PO_4\text{-}6H_2O$ [61]. Precipitation by magnesium ammonium phosphate is done by M. Altinbas et al 2002 [32]. Zeng L et al 2008 recovered ammonia from anaerobically digested cattle manure by steam stripping and evolution is performed at different PH [63]. H. W. H. Menkveld et al. 2017 Recovered ammonium from digestate as fertilizer by stripping of ammonia and formed ammonium sulphate [64]. Wendong Tao and Anayo T. Ukwuani 2015 recovered ammonia through thermal stripping at PH 9 & 11 and acid absorption of ammonia to form ammonium sulphate fertilizers [65]. Sibel Başakçilardan-Kabakci et al. 2007 study on Recovery of Ammonia from Human Urine by Stripping with air and Absorption sulphuric acid [66]. Now these days control of nitrogen is of increasing importance. Many of waste water contain ammonia and untreated wasted in stabilisation process.

7 CONCLUSIONS

1. Chemical stabilisation of waste with Ammonia recovery option is beneficial by point of economic feasibility of process.
2. Iron third chloride and PAC is best coagulating agent for dyes manufacturing industries waste water.
3. Chemical stabilisation of this type waste water is most feasible way of treatment
4. Stripped ammonia can be recovered as ammonium salt like ammonium chloride or ammonium sulphate by absorbing stripped ammonia directly in to acid which are most useful conventional fertilisers for agricultural purpose.

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