

GREEN INNOVATION IN TEXTILE DYE WASTEWATER TREATMENT: A REVIEW EXPLORING SAWDUST, ORANGE PEEL, MANGO LEAVES, AND USED TEA LEAVES

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Abstract: This review examines the effectiveness of sawdust, orange peel, mango leaves, and used tea leaves in treating textile dye wastewater. The textile industry is one of the most polluting sectors of the economy, accounting for around 80% of all dyestuff production. A significant amount of fresh water is required for each stage of the handloom process, including bleaching, dying, and screen printing, depending on the quality, size, demand, and output of the plant. This adversely affects not just particular species or populations but also the natural biological ecosystems. Though there are several unique and fine in-plant sewage and water treatment techniques, they seem tedious and are costly. In recent times, many approaches have been introduced which are cost-effective, simple, and easy to apply with efficient removal of wide range of dyes. Furthermore, the integration of green innovations in wastewater treatment processes can show synergistic effects, enhancing the overall treatment efficiency. The huge postconsumption disposal and reusability makes them even more promising than the others.

Keywords- Wastewater, Green innovations, Bio-adsorbent, Adsorption, Industrial effluents, Biomass.

I. INTRODUCTION

Textile industry is ever emerging among the industrial segment across the globe. Since the primitive era, the usage of natural dyes is obtained from flora and fauna for colouring of textile Fiber. Large-scale applications of dyes can be found in a variety of industries, including textile, leather tanning, paper, wood staining, agricultural, biological and chemical research, pharmaceuticals, medications, cosmetics, hair colouring, photochemical cell manufacturing ^{1,2}.

Industrialization played an important role in the development and emergence of industrial segment. Any manufacturing set-up requires high water and energy consumption. And textile industry falls under such complicated industrial sector where large volume, high quality water is utilized and consequently generates large volumes of wastewater. The majority of the water is used for wet operations including finishing, sizing, and dying. As consequence, these operations produce enormous amounts of wastewater, which are frequently found to have bright colours, significant concentrations of organic chemicals, and wide compositional differences^{3,4}.

Because dyeing operations require a lot of water, the textile sector produces more contaminants in liquid effluents than any other business. Seventy percent of aromatic amines fall into the category of azo dyes, which are dyes with one or more azo groups among their many classes^{5,6}. Ten to fifteen percent of synthetic dyes are projected to be released into industrial waste due to flaws and imperfections in the dyeing process, which poses severe environmental problems globally^{7,8}. Textile mills are among the few rapidly developing and essential pilot plants. For instance, the creation of an integrated yarn production system for denim, along with the dyeing and finishing processes, necessitates an annual capacity of 20,000 tons of cotton fiber, which produces around 45 million denim pieces. However, in every industrial setup, there is a wastewater treatment plant that is actively engaged in sludge operations for wastewater produced from the factory's other facilities as well as from the sizing, dyeing, and finishing processes. This wastewater is then discharged to the sewage system in accordance with established standard protocols³. The rationale behind the significance of in-plant control technology in the textile sector has been presented in a few research. It outlines the philosophies behind achieving notable cuts in wastewater generation, raw material and energy consumption, water use, and other areas. It also proposes a cleaner production method to address the textile industry's current environmental problems^{9,10}.

II. TYPES OF DYES

In the history of 4000 years, the first organic colorant was blue dye called indigo were used to wrap around the mummies in Egyptian civilization. Natural dye was pre-dominant in olden times, covered all sort of dyes derived from natural sources like plants, animal or minerals. Years later, synthetic dyes came into existence around 1870s and for the first time the colorants (natural and synthetic) were classified along with the coloration process^{3,7}. However, modern day classification is broad, categorizing colours based on its colour index, chemical structure, method of preparation, ionic strength, properties and uses, health aid and safety^{11,12}. Among the industries working with dyes, textile industry deals according to the requirement of coloration of fiber, understanding the colour and its constitution to develop a new dye. Classification of dye based on the requirement of textile industry are:

2.1 Direct Dyes

These are mostly used for dying cotton-based fiber for their ease of application, wide-range of shade and low cost. They are further classified based on their applicability such as chromophore (like azo, stilbene, pthalocyanine, formazan, anthraquinone, thiazole, etc.) and fastness properties. Though are easy to apply but has moderate performances.

2.2 Acid Dyes

They are combination of non-metallic and metallic dyes. The non-metallics are applicable for fibers like nylon, wool, silk with pH range of 3-7 called wet-fastness, whereas dry-fastness ranges from pH of 5-6. The overall dyeing strength if moderate to good, due to their lower molecular mass and high ion-ion electrostatic forces between dye and fiber. However, metallic dyes differ from non-metallic as they are based on mordant metals (usually chromium) being incorporated within dye molecule. They are highly recommended for nylon fabric.

2.3 Basic Dyes

These dyes are mostly applied to nylon, polyester and its modified substrate, paper, acrylics, etc. They are water-soluble, produces coloured cations and gets attracted to substrate carrying negative charge in the solution¹³.

2.4 Disperse Dyes

These dyes possess non-ionic character, are either sparingly soluble or insoluble in water. For fabrics like polyester, nylon, cellulose acetate or acrylic, disperse dyes are favourably used. They require carrier, so that the dye compound can diffuse easily into the substrate. However, now carriers are not used, instead dye are now being introduced in the boiling water at around 130°C.

2.5 Vat Dyes

These dyes are insoluble in water but when the carbonyl group which enable the dye to get reduced under basic condition, the dye becomes more water-soluble (i.e., A state called leuco compound). Thus, in leuco compound form it gets absorbed by the cellulose followed by further oxidation to get back the original insoluble vat dye within the fiber.

2.6 Reactive Dves

These dye forms covalent linkage, leading to its permanent attachment with the fibre and cannot be removed any further, becoming a part of the fibre. Therefore, reactive dyes give superiative colour fastness to wash¹⁴.

Types of dye Solubility (in water) **Fibers** Chemical type **Example** Acid Soluble Nylon, wool, silk, Santhraguinone, Congo red; Methyl triphenylmethane, (orange & red); Orange paper azine. xanthene, nitro, nitroso (I, II); Acid (blue, black, violet, yellow) **Basic** Soluble Paper, modified Cyanine, hemicyanine, azo, Methylene blue; Basic azine, xanthenes, acridine, red; Basic (brown and nylon, polyacrylonitrile, oxazine blue); Crystal violet; polyester, inks Aniline; Brilliant green Martius yellow; Direct Soluble Direct Cotton, rayon, paper, azo, anthraneuione, styryl, (anionic dyes) leather, nylon nitro, benzodifurano (black, orange, blue, violet, red) Disperse Insoluble (non-ionic polyester, polyamide, azo, anthraquione Disperse (blue, red 17, and acetate red, orange, yellow, dyes) styryl brown) Soluble Reactive (red. blue. Reactive Cotton, wool. silk, anthraquione nylon phthalocyanine black 5, yellow, black); Remazol (blue, yellow, red) Vat Insoluble Anthraquione Indigo, Vat green 6, Cotton, rayon and indigoids Benzanthrone, Vat

Table 1. Dyes used for different fibres

blue, Vat green

| Solvent | Insoluble | Plastics, | gasoline, | Azo, | triphenylmethane, | Solvent (1 | Red 24, Red |
|---------|-----------|---------------|-----------|---------------|-------------------|------------|-------------|
| | | varnishes, | lacquers, | anthraquinone | | 26, Red | 164, Yellow |
| | | stains, waxes | | | | 124, Blue | 35) |

III. ENVIRONMENTAL IMPACT OF DYE DISPOSAL

Wastewater treatment is getting more and more difficult, particularly for the textile industry. Most of the textile industry runs wastewater treatment facilities, yet data shows that these facilities don't live up to regulatory criteria. When textile effluents are added to agricultural soil, even for a little period of time, the soil's organic matter, calcium, sodium, potassium, magnesium, nitrogen, and phosphorus content deteriorates in comparison to soil that is normally water irrigated¹⁵. Due to the high demand for cotton, silk and polyester not only nationally but also globally, its integrated facility consumes approximately 80 % of overall production of 130,000 tonnes of dyestuff. However, environmental pollution and contamination of local area has been recognized and they are clearly visible^{16,10}. Colorant are the first easily observable contaminate in soil and nearby water bodies (initially around 1ppm)^{9,6}. The flaw in dyeing procedures has estimated the release of around 10-15% of the synthetic dyes (an industrial waste not only restricted to commercial dyes but also heavy metals like arsenic, chromium, lead, cadmium, mercury), consequential environmental and health hazards not only to micro-flora but to others including human¹⁷. Significantly:

- i) Surface level contamination of water can lead to disruption of ecosystem. The dye tends to accumulate in food chain, contributes to fatality of aquatic and soil toxicity, hence, biological degradation¹⁸.
- ii) Sulphonated by-products and their repeated unregulated dump cause negative aesthetic effect leads to soil toxicity. Whereas lack of penetration of light, steadily decreasing photosynthetic activity, lack of available oxygen to sustain life cause aquatic life to decline¹⁹.
- iii) Some dyes have aromatic amine structure due to the formation of carbonium ions and nitrenium from acyloxy amines, are capable of binding to DNA and RNA, causing mutation and tumour formation. Another aromatic compound named benzidine forms reactive intermediates due to metabolic transformation, initiating carcinogenesis⁸.

Table 2. Textile waste-water pollution regulations imposed by different countries

| Parameters | Standards | | | | | |
|------------------------------------|-----------|-------|---------|-----------|-------------|-------|
| | INDIA | USA | CANADA | THAILAND | PHILIPPINES | CHINA |
| pН | 5.5-9 | 6-9 | 6.5-8.5 | 5-9 | 6-9 | 6-9 |
| Biological Oxygen Demand (mg/L) | 30 | 50 | 50 | 20-60 | 30-200 | 60 |
| Chemical Oxygen Demand (mg/L) | 250 | 80 | 80 | 120-400 | 200-300 | 200 |
| Total Dissolved Solids (mg/L) | 2000 | 2000 | 2000 | 2000-5000 | 1200 | - |
| Sulphide (µg/L) | 2000 | 200 | 200 | earc | - JOUP | 1000 |
| Free chlorine (µg/L) | 1000 | 1000 | 1000 | | 1000 | |
| Copper (µg/L) | 3000 | ≤1000 | ≤1000 | 1000 | 1000 | 2000 |
| Mercury (μg/L) | 0.01 | 0.05 | 0.026 | 5 | 5 | - |
| Zinc (µg/L) | 5000 | 10000 | 30 | - | 5000-10000 | 5000 |
| Iron (µg/L) | 3000 | 20000 | 300 | - | 1000-20000 | - |
| Manganese (µg/L) | 2000 | 5 | 5 | 5000 | 1000-5000 | 2000 |

(Source: [Ref 3, 20])

IV. METHODS OF REMOVING DYE

The primary objective of any industry is to develop an efficient and cost-effective solution to eliminate the die-out of wastewater from textiles in order to preserve habitats in and near water bodies. Effluents discharged from the industries are a mixture of not only dyes but metals and other pollutants. Such organic and inorganic contaminants cause fatal haemorrhage, nausea, skin irritation and dermatitis^{21,22}. Furthermore, secondary hazardous compounds that are detrimental to the environment can be produced by synthetic dyes during their breakdown. Therefore, this is an important problem whose solution can improve the quality of aquatic ecosystems by effectively removing these harmful colours from industrial wastewater before it is discharged into the environment^{22,23}. Therefore, to avoid high long term accumulated risk, essential procedure are done before discharging the effluent, which are easy and efficient. They are diluted and recycled off-site to reduce toxicity and effective as well as efficient to remove colours. The methods are categorized into:

Table 3. Methods used for dye removal

| Method | Туре | Merit |
|------------|---|---|
| Physical | Adsorption, sedimentation process, reverse osmosis, microfiltration, ultrafiltration, | Effective at low volumes for a variety of colorants, economical, removal efficiency, low-cost intensive |
| | nanofiltration | regeneration process, low loss of adsorbent with good sorption for a particular colorant ²⁴ . |
| Chemical | Oxidation process (H ₂ O ₂ , Cl, Fenton's reagent, KMnO ₄ and O ₃), ozonation process, photo catalysis, advanced oxidation process | Efficient and useful for water soluble or insoluble colorants, which can decolorize and remove a wide variety of waste with brief storage time as well as reduced capital involvement ²⁵ . |
| Biological | Anaerobic, aerobic or both anaerobic—aerobic bacteria and fungi such as Phanerochaetechrysosporium, Escherichia coli, Pseudomonas aerogenosia, Bacillus, Klebsiella | In addition to COD removal, which is resistant to a wide range of complex colourants, colour removal is facilitated. (Biogas produced is used for steam generation, upstanding removal efficiency at low volumes and concentrations. The removal of a specific colourant is desirable. ^{26,27}) |

V. GREEN TECHNOLOGIES IN REMOVING DYE

Green Technology for textile dye wastewater treatment refers to environmentally friendly methods and processes used to treat wastewater generated from textile dyeing processes. These methods aim to minimize environmental impact, reduce energy consumption, and promote sustainability. Some actively used Green Technologies are Enzymatic degradation²⁸, Bio electrochemical remediation²⁹, Bioremediation with Yeast Diutina rugosa³⁰, Electrooxidation—Salix babylonica³¹, Decolorization by Pseudomonas aeruginosa and crude laccase enzyme³², Bacterial and fungal strains³², Microfiltration—membrane³³, Nanofltration (NF) and reverse osmosis (RO)³⁴, Coagulation³⁵ and electrocoagulation³⁶, with overall colour removal efficiency ranging from 73-100%³⁷. The main disadvantages of these processes are heavy energy consumptions that increases operational cost, their negligible recovery and emission of greenhouse gas emissions.

Therefore, out of many greener approaches that has been put forth to treat textile wastewater that causes least unfavourable effects than other conventional treatment methods is adsorption using bio-adsorbent as it's the most effective way to remove synthetic colours from aqueous effluents^{38,39,40,41}. It does so by transferring the dyes from the water effluent to a solid phase, which reduces the effluent volume to the least^{40,42}. Following this, the adsorbent can be regenerated or kept dry and away from the environment^{42,43}. The most commonly used adsorbent for removing dye from aqueous solutions is activated carbon due to its superior adsorption capabilities^{16,44}. Unfortunately, the high cost of using activated carbon widely around the globe to remove dye from industrial effluents, limits its broad application for wastewater treatment^{45,46}. Therefore finding the alternative, inexpensive bio-adsorbents for the removal of dye is becoming more and more popular. Examples of these are agricultural biomasses⁴⁷, orange seeds⁴⁸, neem sawdust⁴⁹, sugarcane bagasse⁵⁰, cactus peel⁵¹ with removal efficiency ranging from 75-97.8%. While these bio-adsorbents are often cheaper than activated carbon as they are readily available from the natural resources, their overall cost-effectiveness may vary. Factors like availability, preparation methods, and required dosage can affect the cost-benefit balance.

VI. MOST PROMISING BIO-ADSORBENTS

Some examples of such promising Bio-adsorbent (waste remover) are orange peel, saw dust, mango leaves, and used tea leaves^{9,52}. They are Leaf-based or wet-waste materials (raw or activated). Attracted special attention among the adsorbents tested, primarily because they are inexpensive, readily available in large quantities in nearly every part of the world, require minimal preparation, and are frequently reusable because they have no other uses⁷. Furthermore, adhering to the circular economy's tenets, employing biodegradable materials as absorbents to remove dyes from wastewater can drastically cut down on the quantity of waste produced^{53,1}

6.1 Orange Peel

Citrus being one the most used flavour in food industry right from beverages to food additives, from cosmetics to medicines. Though they can be reused for its pectin source, essential oil, polyphenol and carotenoid, but their disposal remains as concern and tagged as littering waste. Orange peel is largely composed of cellulose pectin; hemi-cellulose, lignin and other low molecular weight compounds including limestone. Apart from its multiuse, they are highly effective for removal of heavy metals like iron and its oxides, lead, cadmium, etc. Different studies suggested orange peel to be highly efficient in acidic pH^{54,55}.

6.2 Mango Leaves

One of the evergreen plants, it can produce up to two million tons of leaves a year, most of which are burned or thrown away. The ability of mango leaves to remove acid yellow-99 and azo dye, which are carcinogenic, from simulated wastewater, prevent water pollution, and understand the mechanisms underlying the phenomenon have all been thoroughly investigated ^{56,57}.

6.3 Sawdust

Identified as the most plentiful and overlooked by-product of the wood industry, it is made up of lignocellulosic material, which is a sustainable starting point for the production of activated carbon. It is made up of cellulose, hemicellulose, and lignin^{6,58}. The presence of functional groups like carboxyl, hydroxyl, phenol and amide, they adsorb a large variety of dye and can be modified with

acid or base to enhance the adsorption properties⁵⁹. There are physical and chemical methods of activating sawdust to become activated carbon and are highly efficient for removal of direct dyes⁶⁰.

6.4 Spent/Used tea leaves

Tea is one of the most popular drinks in the world, especially in Asia and the Middle East, where tons of waste are generated after tea is consumed. They are categorized as carbonous adsorbents, and because of their special physiochemical properties—such as a greater surface area and quicker adsorption kinetics—they can be used to remove pollutants from wastewater and serve as an affordable initial material for the synthesis of activated carbon^{61,4}. Tea leaves net dry matter is made up of carboxylate, aromatic, phenolic, hydroxyl, and oxyl groups, which contribute to their unique ion-exchange behaviour. It has the ability to remove metal from solutions and wastewaters as a result^{62,63,64}.

| Sr. no. | Adsorbent | pН | Adsorbent Dosage (g/L) | Temperature (activated compound) | Adsorption capacity (mg/g) | Removal Efficiency |
|------------|--------------------|------|------------------------------|--|----------------------------------|-----------------------|
| 1 | Used Tea leaves | 6 | 4 | 110 °C | 2.987 | 95% |
| 2 | Mango leaves | 8-10 | 3.8-4.3 | 98 °C | 1.998 | 93.80% |
| 3 | Orange peel | 3 | 5-6 | 100 °C | 2.001 | 92.75% |
| 4 | Sawdust | 2-10 | 4 | 60°C | 2.345 | 75% |

Table 4: Comparison of adsorbent based on their characteristics

VII. CONCLUSION

This review is an attempt to present a wide range of studies published in the literature so far based on treatment of textile dye wastewater using greener innovation and presents a promising solution to mitigate environmental pollution and address sustainability concerns within the textile industry. Greener innovation offers several advantages over traditional treatment methods, including lower energy consumption, reduced chemical usage, and decreased production of hazardous by-products. Additionally, these approaches often result in improved effluent quality and compliance with stringent environmental regulations, thereby promoting responsible industrial practices. Though, adsorptive removal of dyes is still a research area wide open and is constant development, may efforts have been made by researchers to contribute with a low-cost alternative that can be as efficient as that offered by classical remediation technologies and many more yet to be made. By embracing innovation and prioritizing environmental stewardship, the textile industry can play a vital role in advancing sustainable development goals and promoting a more sustainable future for all.

VIII. REFERENCES

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