

# Progress of Dye Wastewater Treatment

## Methods



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**Abstract:** Wastewater treatment is an important aspect of the dyestuffs industry, as the process of dye production generates a large amount of wastewater containing various organic and inorganic pollutants. To protect the environment, effective wastewater treatment methods, such as chemical oxidation and biodegradation, are needed to remove pollutants from the wastewater and to meet the discharge standards. This paper reviews the research progress of dye wastewater treatment methods, including flocculation and precipitation, oxidation, electrochemistry, biodegradation, adsorption, etc. The different treatment methods are introduced and their current status and advantages and disadvantages are reviewed. Chemical methods have short treatment time and good effect, but the process is complicated and not easy to operate, and the intermediate products produced may have greater toxicity; biological methods have the advantages of environmental protection and mild conditions, but the treatment time is generally longer, and the poor biochemistry of many dyestuffs restricts the use of biological methods; physical methods have been widely used because of their simple operation and adaptability, among which adsorption is commonly used in the decoloration treatment of wastewater. Undoubtedly these researches are of great significance in recognizing the nature and characteristics of pollutants and mastering the principles and methodological features of environmental treatment technologies.

**Keywords:** Dye Wastewater; Treatment Method; Physical Method; Chemical Method; Biological Method

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## 1 Introduction

The dyestuff industry is one of the industries in the chemical industry with serious environmental pollution. At present, China's annual production of dyestuffs exceeds  $9 \times 10^5$  t, ranking first in the world and accounting for 45% of the world's total production [1]. According to statistics, about 1-2% of dyestuffs in the production process of dyestuffs and about 1-10% of dyestuffs in the dyeing process will be discharged into the environment with wastewater, which poses a serious threat to the environment. Even if the concentration of dyes in the water body is very low, it can equally cause strong visual impact and aesthetic damage, affecting the normal functioning of the receiving water body. Due to the absorption of sunlight by dyes in the water body, the transmission of light

through the water is weakened, which can inhibit the photosynthesis of aquatic organisms in the water body and reduce the diversity of aquatic organisms as a whole. Dyes, as a class of structurally stable organic compounds, are resistant to acid, alkali, light, and microorganisms, and have a long retention period in the environment. Therefore, the negative effect of dyes on the environment lies not only in their chromaticity and COD but also in the potential hazards to human health and the growth and development of plants and animals.

There are many methods for the treatment of dye wastewater, mainly including physical, chemical, and biological methods, such as flocculation and precipitation, oxidation, electrochemistry, biodegradation, adsorption,

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and so on [2, 3]. Each of these methods has its advantages and disadvantages [4]: the chemical method has a short treatment time and good effect, but the process is complicated and not easy to operate, and the intermediate products produced may have greater toxicity [5]; the biological method has the advantages of environmental protection and mild conditions [6–8], but the general treatment time is longer, and the poor biochemistry of many dyestuffs restricts the use of the biological method [9]; The physical method is widely used because of its simplicity and adaptability, of which the adsorption method is commonly used for the decolorization of wastewater [3, 10]. Undoubtedly, these researches are of great significance in recognizing the nature and characteristics of pollutants and mastering the principles and methodological features of environmental treatment technologies.

## 2 Overview of Research on the Treatment of Dye-Containing Wastewater

### 2.1 Classification of Dyes, Characteristics, and Hazards of Dye Wastewater

Dyestuff is an important fine chemical product that is closely related to our human beings' food, clothing, housing, and transportation. Not only do the textiles used by people need dyeing substances, but also leather, paper, wood, soap, paint, resin, beverage, and food need dyeing substances, and human beings cannot survive without dyestuff. There are many kinds of dyestuffs with complex chemical compositions, and there are two main bases for their categorization [11–13]. First, according to its application classification, it is divided into direct dyes, sulfide dyes, ice dyes, mordant dyes, oxidation dyes, reduction dyes, reactive dyes, acid dyes, disperse dyes, cationic dyes, fluorescent whiteners, etc. [14]. Secondly, according to the classification of chemical structure, it can be divided into azo dyes, anthraquinone dyes, stilbene dyes, indigo dyes, triphenylmethane dyes, styryl dyes, triaryl methane dyes, lactone dyes, etc., of which azo dyes have the largest production, followed by anthraquinone dyes. Azo dyes have complete chromatography and mature technology, and their molecules generally contain one or more azo

bonds with phenyl or naphthalenyl groups, and phenyl or naphthalenyl groups with substituent groups such as  $-\text{NH}_2$ ,  $-\text{Cl}$ ,  $-\text{CH}_3$ ,  $-\text{NO}_2$ ,  $-\text{SO}_3\text{H}$  and  $-\text{OH}$ . Most of the azo dyes themselves are not toxic, but their potential hazards should not be ignored. In 1895, after the discovery of bladder cancer patients among workers engaged in the production of magenta, caused people to pay great attention to the toxicity of azo dyes, carcinogenic effects and mutagenic activity have been studied [15]. The results showed that azo dyes containing p-phenylenediamine or benzidine in the molecular structure are decomposed into aromatic amines by the azoreductase of intestinal bacteria after entering the human body [13]. Azo dyes are not easily decomposed and mineralized in an aerobic state, but in an anaerobic state, they are easily decomposed and reduced by microorganisms to aromatic amines and other intermediate products. They are even less susceptible to decomposition and mineralization in the environment, and some are mutagenic, carcinogenic, or otherwise toxic, affecting the health of those exposed. The categories of dyestuffs regularly produced are 12, with nearly 600 varieties, and are increasing at the rate of 20 to 30 new varieties per year. However, this is accompanied by the discharge of approximately 1.16 billion tons of wastewater from dyestuffs each year.

Dyestuff wastewater mainly comes from the dyestuff and dyestuff intermediates production industry and dyestuff use industry, which consists of mother liquor of crystallization of all kinds of products and intermediates, auxiliary agents, materials lost in the process of production, and sewage water that washes the ground. Different varieties of dyestuffs, different production processes, and different dyeing processes produce different amounts and compositions of wastewater. Its water quality characteristics can be summarized as:

- (1) High chromaticity, low thousands, high hundreds of thousands. Printing and dyeing wastewater contains dyes and other colored pollutants, the color is very dark. It is generally believed that printing and dyeing wastewater with color will hinder the transmission of daylight in the water, and is not conducive to the photosynthesis of aquatic plants, the result is to reduce the bait of aquatic animals, in the process of degradation reduces the dissolved oxygen in the water, and is not conducive to the growth of aquatic animals, especially the wastewater with a large amount of suspended solids, which is more serious. It is believed that fish avoid colored water. Printing

and dyeing wastewater in the sulfuric acid or sulfate, in the soil reduction state can be converted to sulfide, the result of which produces a large amount of hydrogen sulfide, causing plant root rot. Secondly, it will also adversely affect the growth of soil microorganisms; soil acidification promotes the loss of salts and deteriorates soil properties. Due to phenomena such as soil crusting and condensation, the growth and development of the crop root system will be limited, and crop yields will be reduced. Therefore, printing and dyeing wastewater not only causes serious pollution to natural water bodies and soil but also directly jeopardizes human health. The treatment of printing and dyeing wastewater can not only reduce or avoid environmental pollution but also reuse the treated water, saving water resources. The removal of COD from wastewater is correlated with decolorization, but the problem of decolorization is difficult. Therefore the decolorization technology of this type of wastewater is an important topic for environmental workers.

(2) Small BOD<sub>5</sub>/COD<sub>Cr</sub> value and poor biochemistry.

Due to the complexity and long flow of the dye production process. The yield of the produced product is very low, only 40~50%. While producing products, the dyestuff industry also produces a large amount of waste liquid containing unreacted raw materials, by-products, and waste products. The COD of the wastewater reaches 1000~100,000 mg/L. What is more unfavorable is that most of the organic components in the wastewater of dyestuffs are based on aromatic hydrocarbons and heterocyclic compounds, with chromophores and polar groups, and the dyestuffs have been developed towards the direction of anti-photolysis, anti-oxidation, and anti-biochemistry in recent years, which makes the toxicity of the wastewater larger, the ratio of BOD/COD lower, and the biochemistry poor.

(3) The water quality and quantity varies greatly. The dyestuff industry is generally characterized by small batch and multi-species, and the printing and dyeing industry is also characterized by frequent change of varieties, mostly intermittent operation and intermittent discharge of wastewater, with a wide range of changes in the quality and quantity of water and a complex composition, which contains not only nitro, hydroxyl, amine, halogen, alkyl, sulphonic acid,

carboxylic acid, and a mixture of the above-substituted aromatic hydrocarbons, thickly cyclic aromatic hydrocarbons, heterocyclic aromatic hydrocarbons, aliphatic hydrocarbons, and unsaturated aliphatic hydrocarbons compounds. In addition, there are a large number of inorganic salts such as NaCl, Na<sub>2</sub>SO<sub>4</sub>, and many heavy metal ions as well as compounds of non-metallic elements N, P, S. The compounds are also available in the form of a variety of other substances such as alkanes and sulfonic acids.

The removal of COD<sub>Cr</sub> is correlated with decolorization, but decolorization is much more difficult. In short, the complex composition, high concentration, high COD<sub>Cr</sub> value, high chromaticity, and difficult-to-degrade wastewater generated from dyestuff production is a difficult problem in industrial wastewater treatment. Exploring effective and practical treatment methods has become a research topic at home and abroad today. In the actual use of dyes, azo dyes account for more than half of the dosage, so the study of degradation and decolorization of azo dyes wastewater has attracted much attention.

## 2.2 Dye-Containing Wastewater Treatment

China's textile printing and dyeing wastewater treatment began in the early sixties, the earliest put into use is the Baoding Chemical Fiber Factory viscose wastewater treatment project, the Beijing vinylon Factory acid aldehyde wastewater treatment project, followed by the Anhui Textile Printing and Dyeing General Factory dyeing wastewater treatment project is also put into operation. Since the early seventies, China's textile printing and dyeing wastewater treatment of scientific research gradually, the end of the seventies to the early eighties, China has a considerable number of textile printing and dyeing plant completed the wastewater treatment project. In the "Environmental Protection Law" and other laws and regulations promulgated, China's textile printing and dyeing water research and management work further promoted; foreign textile printing and dyeing wastewater treatment in the fifties that began in the development of the industry at that time, resulting in environmental hazards, many countries began to invest in research. From the fifties to the sixties, the main single-type technology research; to the seventies, mainly focusing on comprehensive research, that is, the study of regional governance methods, the

various governance techniques, and technical and economic indicators linked to the management of printing and dyeing wastewater and the overall governance of the region or the city is linked to becoming a component of it, to avoid duplication of governance. From the end of the 1970s to the present, more attention has been paid to the requirements of energy conservation, rational utilization of resources, and improvement of environmental quality.

## 2.3 Treatment of Dye-Containing Wastewater

Effective degradation and treatment technology for dyes is an important prerequisite for the treatment of dye wastewater. In view of the fact that most dyestuffs are chemically stable and difficult to degrade, scientists from all countries have attached great importance to the research on the degradation and treatment of dyestuffs and dyestuff wastewater. With the progress of science and technology as well as the continuous development of pollution control technology, human beings have found many effective methods to treat dye wastewater, which are summarized as the physical-chemical method, biological method, and physical-chemical-biological joint method [16–19].

### 2.3.1 Physical Method

#### (1) Adsorption

Adsorption method for removing pollutants from wastewater is a process in which one substance attaches to the surface of another substance [10, 18, 20–22]. It can occur between gas-solid, gas-liquid, and liquid-solid phases [23, 24], using the surface activity of the adsorbent, the molecular state of the pollutants adsorbed and concentrated on its surface, to achieve the purpose of purifying wastewater. Adsorption is a typical physicochemical treatment technology, which is commonly used in the environmental protection field for deep treatment and pre-treatment. Activated carbon is usually considered as the best adsorbent [25]. Other studies have shown that activated carbon can only adsorb water-soluble dyes, not suspended solids and insoluble dyes; the adsorption performance of activated carbon on different kinds of dyes is in the following general order: basic dyes > direct dyes > acid dyes > sulfurized dyes. It should be noted that due to the high price of activated carbon and the expensive regeneration cost, it is generally used in the treatment of

low concentration dye wastewater or deep treatment.

As natural minerals have good basic properties such as surface adsorption, mineral pore filtration, and interlayer ion exchange, and are abundant, cheap, and easy to obtain, they are widely used in the treatment of environmental pollution and have formed an important class of materials-environmental mineral materials. The research conducted by Hang et al [17] showed that the decolorization rate of calcium-type montmorillonite produced in Jiaozhou, Shandong Province, for disperse dyes and basic dyes was above 90%, which was better than that of the flocculation method. In addition, oblique hair zeolite treated with acid and alkali and then activated can effectively treat dye wastewater with a decolorization rate of nearly 100%.

The use of industrial wastes such as coal, fly ash, and slag as adsorbents is a good way of comprehensive utilization of wastes. Fang Jinwu et al [26] studied the decolorization ability of coal ash from different coal mines and different particle sizes on dye wastewater. The results show that the best decolorization effect is the coal dust with particle size less than 80  $\mu\text{m}$ , and the decolorization rate reaches 80-100% for reactive dye wastewater when the dosage of coal dust is 50  $\text{kg}/\text{m}^3$ . Sawdust is first hydrolyzed by weak acid and then made into adsorbent after coking, which can adsorb many kinds of dye molecules.

The use of synthetic inorganic adsorbents to remove dyes has also been reported. For example, magnesium oxide and calcium hydroxide are mixed into particles and burned at 400-1000  $^{\circ}\text{C}$  to produce a kind of adsorbent that is easy to regenerate and can be recycled. Magnesium oxide has a good decolorizing effect on acid dyes, direct dyes, dispersed dyes, reactive dyes, and so on. It should be pointed out that the use of adsorption for decolorization if the waste residue after adsorption can not be reasonably regenerated or disposed of, will cause secondary pollution, which is the choice of adsorption decolorization must be considered.

#### (2) Coagulation and sedimentation method

The coagulation and sedimentation method is a more stable and mature method for treating dye wastewater. The commonly accepted mechanisms are bridging action, compression double layer, net trapping, and electric neutralization. The coagulant's characteristics determine its settlement performance, while many environmental factors including temperature, pH, and Eh (redox potential of the platinum electrode relative to standard hydrogen electrode) may promote or inhibit the settlement function. In recent

years, IPF (inorganic polymer flocculant) has become a hot spot for studying the behavior and mechanism of coagulation and flocculation [22, 27]. Compared with ordinary coagulants, IPF can form more effective flocculation of the form  $Al^{3+}$ . The main research direction of coagulation is to develop effective coagulants, especially organic and inorganic composite coagulants. Zhang Kaisong et al [28] developed an inorganic-organic composite coagulant, and the effect of dye wastewater treatment than polymerized aluminum chloride (PAC) is more obvious. Wu Dunhu et al [29] on the use of boron mud composite coagulant treatment of dye wastewater research results show that: when the dose of 0.3 ~ 0.6 g/L, pH value of 4.0 ~ 11.5, the decolorization rate of more than 92%, better than PAC.

### (3) Membrane separation method

Membrane separation technology is applied to dye wastewater, mainly through the separation, concentration, and recovery of pollutants in wastewater to achieve the purpose of wastewater treatment. In the treatment of dye wastewater, the application of more ultrafiltration and reverse osmosis. Fan Lili et al [30] used an integrated reverse osmosis device to study the dye wastewater provided by Fuyang Dyeing Factory, and under an operating pressure of 15 MPa, the effluent conductivity, COD mass concentration, and chromaticity were 23  $\mu\text{s}/\text{cm}$ , 10.8 mg/L, and 7 times, which were in line with the national level of discharge standards. Membrane separation technology does not need to add chemical reagents and does not produce new chemicals in the treatment process, to avoid secondary pollution, the process is simple and easy to operate and can be recovered from the wastewater dyes, and recycling. However, the biggest disadvantage of membrane separation technology is that the membrane flux will decrease with the extension of the treatment process, the replacement frequency is faster, and the membrane cleaning needs a certain cost, the material of the membrane, such as anti-acid and alkali, corrosion resistance, etc., will also greatly affect the treatment effect [31–33]. In addition, the price of the membrane is expensive, which makes the cost of membrane separation technology to treat dyestuff wastewater too high [34, 35], and greatly limits the application and promotion of membrane separation technology in the dyestuff wastewater treatment industry.

## 2.3.2 Chemical Method

### (1) Catalytic oxidation method

The catalytic oxidation method accelerates the decomposition of the oxidant in the system through the catalytic

effect and makes it react with the organic matter in the water quickly, resulting in the oxidative degradation of organic pollutants in a relatively short period of time [36, 37]. In response to the problem that the effect of using advanced chemical oxidation and aerobic biological treatment is not ideal for dispersed dye wastewater, Zhou Jian et al [38] used catalytic oxidation to treat dye wastewater that could not meet the standard after internal electrolysis, not only the daily treatment of anthraquinone series of dispersed dyes up to 2,500 t, but also reduced the chromaticity and COD value of the dyes wastewater that did not meet the standard after internal electrolysis, greatly reducing the operating costs. Arslan [39] used  $Fe^{2+}$  catalyzed ozone oxidation for the treatment of dispersed dye wastewater, and the conclusion of the study pointed out that when ozone (applied dose of 2300 mg/L) oxidation was used alone, it only had a certain degradation effect under the condition of pH=3, and the decolorization rate was only 77%, and the removal rate of COD was only 11%; however, the treatment of anthraquinone series dispersed dye wastewater by the combination of  $Fe^{2+}$  flocculation, ozone oxidation, and  $Fe^{2+}$  catalyzed ozone oxidation was not only a good treatment but also reduced the chromaticity and COD value after internal electrolysis treatment, greatly reducing the operating cost. However, with the combination of  $Fe^{2+}$  flocculation, ozone oxidation and  $Fe^{2+}$  catalytic ozone oxidation, the dosage of  $Fe^{2+}$  was 0.09-18 mmol/L, and the pH value of dye wastewater was 3-13, the decolorization rate reached 97%, and the removal of COD was increased to 54%.

### (2) Fenton reagent method

$Fe^{2+}$  or  $Fe^{3+}$  as a catalyst, in the presence of  $H_2O_2$  produced by the strong oxidation, can make many organic molecules oxidized, and the reaction system does not require high temperature and high pressure, the reaction conditions are not harsh, the reaction equipment is also relatively simple, the scope of application is wider [40]. In recent years, some scholars have introduced ultraviolet light (UV), oxalate, etc. into the Fenton method, making the oxidation capacity of the Fenton method greatly improved, and the treatment effect more significant. the shortcomings of the Fenton method are: oxidation capacity is relatively weak, the effluent contains a large number of iron ions, and coloration. In recent years, the immobilization technology of iron ions, the Fenton oxidation method has become an important direction.

### (3) Photo-oxidation

Photo-oxidation is the use of photochemical reactions

to degrade pollutants, including no catalyst and catalyst participation in 2 kinds, the former is also known as photochemical oxidation, and the latter is also known as photocatalytic oxidation [19, 41]. Photodegradation usually refers to the role of organic matter in the light, gradually oxidized into low molecular intermediate products, and ultimately generate  $\text{CO}_2$ ,  $\text{H}_2\text{O}$  and some other ions, such as  $\text{PO}_4^{3-}$ ,  $\text{NO}_3^-$ ,  $\text{Cl}^-$ , and so on. The photodegradation process of organic matter can be divided into direct photodegradation and indirect photodegradation. Direct photodegradation refers to the absorption of light energy by organic molecules after further chemical reactions. Indirect photodegradation is the existence of certain substances in the surrounding environment to absorb light energy to form an excited state, and then induce organic pollutants to produce a series of oxidative degradation reactions, which is more effective in dealing with difficult biodegradation of organic pollutants in the environment.

#### (4) Ozone oxidation method

Ozone oxidation ability is very strong, in addition, to disperse dyes, it can destroy the organic dyes color or color-assisting groups and has a certain decolorizing effect [42, 43]. Due to the low solubility of ozone in water, how to more effectively improve the solubility of ozone in aqueous solution has become a hot spot and the key to the study of ozone oxidation technology. In addition, the use of ozone will produce some by-products, in particular, we should pay attention to the carbonyl compounds of formaldehyde, acetaldehyde, and other aldehydes, because these substances have acute and chronic toxicity and certain carcinogenic, teratogenic, mutagenic, easy to lead to secondary pollution, in addition, the cost of the ozone generator is relatively high, and therefore the use of a separate is not enough to be economical.

#### (5) Ultrasonic oxidation method

With the in-depth study of ultrasonic chemistry, ultrasonic oxidation is considered a clean and good prospect for the application of the method, becoming an effective technology to deal with water pollution. The high temperature and pressure formed by the acoustic cavitation effect under the action of ultrasound prompts the dissociation of water vapor and other gases inside the cavitation bubbles to produce free radicals, which triggers the ultrasonic chemical reaction. Ince et al. [44] showed that: pH has an important effect on the degradation of dyes, and the degree of degradation increases with the decrease in pH; the smaller the molecular mass, the simpler the structure, and the smaller the structure, and the simpler the molecular

mass. The smaller the molecular mass, the simpler the structure, and the easier the degradation of dye molecules with anodyne substituents in the proximity of hydroxyl substituents. Tezcanli-Gtiyer et al. [45] found that the hydroxyl radicals attacked the dye's chromophore first, and the decoloration of dyes was faster than the destruction of the aromatic ring.

#### (6) Electrochemical method

Electrochemical treatment technology has progressed rapidly in recent years, and the synergistic effect of oxidation, photocatalytic oxidation, or catalytic oxidation has been added to the original basis, and the limitations of microelectrolysis technology have been better solved. At present, the electrochemical method is mainly applied in the removal of biotoxic organic pollution compounds, and one of the most attractive features of this method is that it can exert an electrocatalytic performance that is unique to the electrochemical method, and it can selectively degrade the organic pollutants to a specific degree. In addition, the electrochemical method has good synergy with other treatment methods and can be used in combination to achieve the desired treatment effect. However, the use of electrochemical methods for the complete degradation of organic pollutants in water is too expensive and requires a large amount of energy.

### 2.3.3 Biological Method

The biological treatment method is to separate and oxidatively degrade dyes through the flocculation and adsorption function of biological organisms and biodegradation. Bio-flocculation and bio-adsorption do not chemically change the dyes. The biodegradation process, on the other hand, uses microbial enzymes and other effects to oxidize or reduce the dye molecules, destroying the chromophore and unsaturated bonds of the dyes, and through a series of oxidation, reduction, hydrolysis, and chemosynthesis, the dye molecules are eventually degraded into simple inorganic substances or converted into nutrients or protoplasm needed by various microorganisms themselves. There are three kinds of biological treatment methods: aerobic treatment, anaerobic treatment, and anaerobic-aerobic combined treatment. For the traditional biological treatment of textiles, dye wastewater in the organic dyes cannot play an effective role in the treatment of the actual situation, some scholars in recent years, have focused on research and development of anaerobic and aerobic technology, and have achieved unexpected results. Some studies show that with the simulta-

neous application of aerobic and anaerobic methods, through the realization of complementary advantages, a lot of aerobic biological methods cannot oxidative degradation or degradation of a limited degree of organic dyes, through the anaerobic method can be realized to different degrees of degradation. As one of the practical water pollution treatment technologies, the development and research of microbial treatment of dye wastewater has a history of many years. The mechanism of microbial decolorization and degradation is very complex and diverse, and many degradation processes and reaction mechanisms are still unclear and need to be explored continuously. In the early 1980s, immobilized microbial technology became a hot spot for research on organic industrial wastewater treatment at home and abroad. This technology is to immobilize dye-degrading microorganisms on the surface of specific carriers to improve microbial degradation efficiency. The microorganisms used for immobilization have various ways such as single and mixed. Relevant studies point out that mixed bacteria decolorize and degrade better. With the development of immobilized decolorizing bacteria carrier technology, decolorizing degradation reaction time is also greatly reduced. Bio-enhancement technology adds microorganisms with specific functions in the biological treatment system to improve the treatment performance of the original treatment system for the removal of difficult-to-degrade organic matter. The main ways to implement the bio-enhancement technology are the addition of highly efficient degradation of microorganisms; the addition of genetically engineered bacteria (GEM); the optimization of the nutrient supply of the existing treatment system, through the addition of substrates or substrate-like substances to stimulate the growth of microorganisms or improve their vitality. Membrane bioreactors are also a new wastewater treatment technology developed in recent years.

Despite the great development of the biological method, with the decrease of the biochemical degree of dye wastewater and the limitation that microorganisms have harsh requirements on nutrients, pH value, temperature, and other conditions, it is difficult to adapt the biological method to the actual situation of large fluctuation of dye wastewater water quality, many kinds of dyes and high toxicity in the practical application of treating dye wastewater. Such as the high efficiency of microorganisms and immobilization and other bio-enhanced technologies. Many experts and scholars have devoted themselves to the screening of efficient degrading bacteria and the construction of genetically engineered bacteria to re-

alize the use of abundant resources available in nature to serve mankind, but the practice has shown that the newly developed high-efficiency bacteria applied to the treatment of dye wastewater may not be able to fully achieve the expected strengthening effect. In addition, microorganisms have their own safety problems, and the flow of highly efficient bacteria and genetically engineered bacteria into the natural environment may pose a threat to the natural environment and the ecological balance, therefore, the application of these biological methods must be subjected to rigorous environmental safety inspection and assessment in advance. Meanwhile, the degradation mechanism of dyes by microorganisms and the metabolic mechanism of microorganisms need to be further studied and explored.

### 2.3.4 Combined Physical-Chemical-Biological Approach

Single physical and chemical methods and biological methods for treating dye wastewater have their advantages, but they also have limitations [46]. Therefore, many scholars and engineers began to try to combine the physical-chemical and biological methods and achieved very good treatment results by complementing each other's advantages and taking advantage of each other's shortcomings. For the water quality characteristics of towel factory wastewater with small water volume, high content of organic pollutants, a wide range of concentration fluctuation, large alkalinity, deep chromaticity, and poor biochemistry, Xu Yudong [47] used the anaerobic folded plate reaction tank - biological contact oxidation tank - coagulation and sedimentation - sand filtration tank joint treatment process for dyeing Xu Yudong [47] used the combined treatment process of anaerobic folding flow plate reaction tank - biological contact oxidation tank - coagulation precipitation - sand filter tank to treat the dye wastewater, and the effluent water quality could reach the first level of industry discharge standard. Lu et al [48] based on the traditional dye wastewater treatment process, the use of hydrolysis acidification - contact oxidation method combined technology for the treatment of dye wastewater, the results show that the combined treatment method process structure is simple, the treatment effect on the wastewater is obvious, the effluent water quality is stable. Khattri et al [49] applied the principle of combining physicochemical and biological treatment methods, the use of biological adsorbent for the treatment of Crys-

talline violet, methylene blue, peacock green, wakadamy-cin B and other dyes wastewater were treated with good results. Walker et al [50] immobilized a kind of *Pseudomonas aeruginosa* on granular activated carbon to treat the wastewater containing acid dyes the combined treatment effect was greatly improved compared to the use of single activated carbon.

### 3 Conclusion and Outlook

This paper reviews the research progress of dye wastewater treatment methods, including flocculation and precipitation, oxidation, electrochemistry, biodegradation, and adsorption. The different treatment methods are introduced and their current status, advantages, and disadvantages are reviewed. Chemical methods have short treatment time and good effect, but the process is complicated and not easy to operate, and the intermediate products produced may have greater toxicity; biological methods have the advantages of environmental protection and mild conditions, but the treatment time is generally longer, and the poor biochemistry of many dyestuffs restricts the use of biological methods; physical methods have been widely used because of their simple operation and adaptability, among which adsorption is commonly used in the decoloration treatment of wastewater. Undoubtedly these researches are of great significance in recognizing the nature and characteristics of pollutants and mastering the principles and methodological features of environmental treatment technologies.

As the current technology is applied to the actual dyestuff wastewater treatment is difficult to meet the needs of dyestuff enterprises in both technical and economic aspects. Therefore, many environmental protection scientists are committed to the research and development of new dye wastewater treatment technologies. In recent years, the more active research on new technologies for dyestuff wastewater treatment includes supercritical water oxidation technology, high-temperature deep oxidation technology, low-temperature plasma chemical technology, extraction technology, and so on. Dye wastewater is still one of the more difficult industrial wastewater to treat, and it is necessary to consider both the advancement of the treatment technology and the feasibility of the infrastructure investment and operating costs. Therefore, the techno-economic analysis of these emerging technologies will make their industrialization and application break through the bottleneck faster.

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