A critical review on recent developments in the low-cost adsorption of dyes from wastewater

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ABSTRACT

Water is one of the most important components in the environment. This is essential for all forms of life and leisurely plays a vital role in the world economy. The discharge of large amount of dye wastewater from different industries such as textile, leather, pulp, rubber pharmaceuticals, food processing, electroplating cosmetics, plastic, paper industries, etc. to the aquatic system constitutes the major hazards to the living environment. Hence, the rapid removal of these dyes from wastewater before their discharge is an important necessity of day to day life as well as for environmental safety. Several traditional treatment methods were available for the removal of dyes from water/wastewater such as chemical coagulation, filtration, flocculation, ozonation, oxidation, photocatalytic degradation, ion exchange, biodegradation, electrolysis and adsorption. Among all these treatment methods, adsorption process using activated carbon is one of the most important, effective and reliable method for the removal of dyes from aquatic system. However, the widespread application of activated carbon is restricted because of its high cost. Therefore, the attention has moved to select the low-cost and efficient adsorbents which are alternative to the existing activated carbon. Some of the natural materials, agricultural wastes, industrial wastes and biosorbents have been reported as an effective low-cost adsorbent for the removal of dyes from aquatic system by many researchers. The current review paper explains the detailed survey on the dye removal methods, and scope for the improvement can be done on the removal of dyes from industrial wastewater.

Keywords: Dyes; Methods; Removal; Adsorption; Wastewater; Toxicity

1. Introduction

Water is an essential component for living environment. Water pollution by various toxic pollutants has become one of the most important serious issues worldwide. The rapid growth of industrialization, anthropogenic activities, unplanned urbanization, unskilled utilization of natural water resources and tremendous increase in population lead to release of several toxic contaminants in to the environment [1–4]. In addition, on a total of 75% of underground water only 3% of water is found to be pure, in which only 1% is available for drinking and domestic purposes [5]. The provision for the pure water which is basically utilized for domestic and survival purposes is shrinking. This may be due to the

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fresh water pollution. The water and soil resources are getting contaminated due to the release of several noxious compounds such as industrial effluents, chemicals, agricultural runoff, domestic wastes and municipal waste. The wastewater not only decreases the available fresh water resources but also increases the probability of millions of waterborne disorders [6,7]. The important sources of water pollution include industrial chemicals, fertilizers, pesticides, radioactive wastes, solid waste, oil sludges and commercial waste. Apart from the anthropogenic sources some of the natural sources also cause water pollution but most of the frequent water pollution was caused by human activities [8]. Some of the organic compounds available are solvents, pesticides, polychlorinated biphenyls, dioxins and dyes [9]. Among the different pollutants generated from the different industrial applications, dye is considered to be one of the most hazardous pollutants due to their environmental impacts especially for their toxicity to the living organisms. Dyes are mainly discharged from the different industries such as textile, dye production, paper, food, printing, cosmetic, plastic and leather [10–16]. The release of these pollutants into the hydrosphere provides a significant threat to the living environment due to their visibility even at very low concentrations, reduces the sunlight penetration and their toxicity, potential mutagenecity and carcinogenicity [17–23].

1.1. Industrial dye materials

The dyes are coloured organic materials which are utilized by several industries such as textile, leather, paper, plastic, pharmaceuticals, cosmetics, food, etc. and applied for hair, fur, oil refinery products and grease [7,24–26]. Commonly, the dyes utilized in the garments manufacturing sectors are acidic, basic, reactive, specific, azo, vat, caustic, disperse and sulphur dyes [27]. The derivatives of the azo dyes are the major class of dyes used in the industrial applications [28]. Generally, the dyes can be classified into three categories: (i) cationic, (ii) anionic and (iii) non-ionic dyes. The anionic dyes are of direct, acidic and reactive dyes [29]. The cationic dyes possess several chemical structures based on their substituted aromatic groups and they are mostly applied in silk, wool, acrylic and nylon dyeing [30]. These dyes are mainly dependent on the positive ion. They are also known as basic dyes which are generally zinc chloride or hypochloride complexes [31]. They are positively charged, water soluble and provide coloured cations in the aqueous solution. The cationic groups were seen in different types of dyes including azo, anthraquinone, methane, diazo and tri-arylcarnbenium, phthalocyanine, several polycarboxyclic and solvent dyes. The anthraquinone dyes are weak and expensive. In addition, they have great properties, strong and low cost. Generally, the cationic dyes are visible and have brilliance and intensity of colours [29]. These cationic dyes are considered as harmful colourants causing serious health hazards such as cancer, genetic disorder, skin irritation and dermatitis [32].

The anionic dyes are mostly dependent on the negative ion [31]. These dyes belong to different types which show distinctive difference in their structure but they possess common feature such as ionic groups and water solubilizing properties. These dyes also include direct, reactive and acidic dyes [33]. The reactive groups of the reactive dyes interact with the wool and cotton and form strong covalent bonding with them. The release of reactive dyes into the ecosystem is not advisable because of their low degree of fixation due to the hydrolysis of reactive groups in water [34]. The acid dyes are applied with wool, silk, acrylic, polypropylene and polyamide fibres. They have good water soluble property. In addition, these are harmful to the humans because of the toxic organic sulphonic acids [35].

1.2. Hazards of industrial dye materials

The industrial dyes are considered hazardous to the environment because many dyes are toxic to living organisms either directly or through their absorption and reflection of sunlight entering the water which interferes with the growth of aquatic animals. These dyes not only provide harmful effects to the living environment but also damage the quality of the fresh water sources. The acidic, basic, reactive, azo, diazo, disperse, anthraquinone based and metal complex based dyes are used in the textile industries [36]. The higher intake of these dyes causes vomiting, shock, heart disorders, Heinz body formation, cyanosis, jaundice, quadriplegia and tissue necrosis in humans [37]. The availability of pigments in the aquatic system also leads to water coloring, oxygen reduction, thereby damaging plants and animals [38]. The most widely used basic or cationic dye is Malachite Green (MG), which was widely used as medical disinfectant, bio-cide and colouring agent to wool, silk, leather, jute, paper, cotton and acrylic materials. MG dye causes carcinogenic, mutagenic and teratogenic effects to mammalian cell. This provides potential harmful effects on kidneys, livers, gills, intestines and gonads in organisms [39–42]. Another most important basic dye is Methylene Blue (MB), which was widely used as a colouring agent for wool, cotton and silk. This MB dye is also utilized as a staining agent for making certain body fluids and tissues earlier to view during surgery and diagnostic assessments. This MB dye can also be utilized for the medical treatment of methaemoglobinemia and cyanide poisoning. In spite of several useful applications, this MB dye has a number of negative effects to the living environment which includes irritation of mouth, throat, oesophagus and stomach with symptoms of nausea, abdominal discomfort, vomiting and diarrhoea [22,43–45]. The skin contact to this MB dye can cause mechanical irritation which results in redness and itching. The Metanil Yellow dye is a tumour causing compound leading to cancer [46] and also cause enzymatic disorder in the human beings [47]. This dye generally does not provide mutagenic effect but it is capable of latering the gene expression [48]. During oral intake, this dye causes toxic methaemoglobinemia [49] and cyanosis [50] in human beings and the skin contact causes allergic dermatitis [51]. The oral consumption of dyes in animals creates testicular lesions [52].

The removal of these toxic dyes from wastewater becomes globally essential activity because even a small concentration of pigment in water is noxious and highly conspicuous. As the elimination of these toxic dyes from aqueous solution is considered an environmental challenge, the government legislation needs the industrial wastewater to be treated, therefore there is a constant requirement to have an effective
process that can potentially remove toxic dyes. Due to the increased consciousness provided to the significance of effluent treatment plant, the research scientists are insisted to formulate potential, eco-friendly and low-cost technologies which are capable of eliminating toxic contaminants present in the wastewater and also to defend the populace from dreadful health disorders. This is the reason why it is essential to remove pigments and colourants from the aqueous solution. A variety of treatment technologies such as coagulation, filtration, oxidation, advanced oxidation process, electrochemical, biological treatment, ion exchange and adsorption have been employed for the removal of dyes from wastewater.

2. Dye removal methods

Dyes are one of the most vital classes of organic macromolecules. These dyes occupy an important place in our survival and are utilized in different industrial sectors that include textile, plastics, paints, leather, drugs, waxes, greases, cosmetics, optics, dye-sensitized solar cells, sensors, fur, hair, etc. The source, colour, molecular structure and colour index are some of the properties based on which the dyes are classified. Basically, they are distinguished depending on the chromophores available in these chemicals. In earlier days, the selection of dyes and its applications are not given a higher impact based on its effect on the environment because the industries failed to calculate the chemical composition of the dyes used in it. In 1980s, people started to provide much attention to the dye waste because of their health concern which is mainly based on aesthetic condition. Over this decade, more details on the deleterious consequences of dyes applications have been reviewed. In addition, the dye utilizing industries, government and other environmental organizations have enunciated certain permissible limits for the discharge of toxic colourants into the aqueous system and they take effective measures for the treatment of industrial dyes in the wastewater. In the beginning, the wastewater treatment was performed using some physical treatments including equalization and sedimentation to maintain the pH, total suspended solids and total dissolved solids of the discharged dye wastewater [53,54].

After that the secondary wastewater treatments such as filter beds were applied for the degradation of dye wastewater. In recent times, the introduction of the activated sludge processes was applied in the dye effluent treatment. Generally, the industrial effluent treatment includes preliminary, primary, secondary and tertiary treatment technologies. The various treatment technologies involved in the elimination of toxic dyes are depicted in Table 1. The preliminary treatment includes screening and gravity settling chamber to remove the large size and settleable solids from the wastewater, respectively. The primary treatment includes physical or chemical treatment to remove the suspended solids from the wastewater. The secondary treatment is the biological treatment to remove the biologically degradable organics in the wastewater. The important biological treatment techniques include attached and suspended growth systems. The waste sludge from the biological treatment units was further treated with the help of either by aerobic or anaerobic decompositions. The tertiary treatment is the physical–chemical treatment which includes adsorption, membrane, ion exchange, chemical oxidation and stripping.

The above-mentioned methods are highly expensive than the other biological treatment methods but these methods are used in the removal of inorganic pollutants which are not removed easily by the biological methods [55]. However, these methods are generally applied in series with the biological treatment but sometimes they are also used as a separate treatment process. The industrial dye wastewater is also treated in more or similar procedures but no single treatment methodologies were applied for all types of dye wastewater. Though various treatment techniques are available for dye removal, some of them are not more suitable due to their drawbacks [56]. The different dye removal methods were shown in Fig. 1.

2.1. Chemical coagulation/flocculation

The dye wastewater treatment with chemical coagulation/flocculation process is one of the effective methods to remove colour from the dye wastewater [57–59]. This method is a low cost, simple, reliable and low energy consuming process as compared with the other methods. This treatment process is a recognized method which effectively removes the colloidal, suspended and soluble solids through induced aggregation of both micro- and macro-particles into larger particles and followed by sedimentation. The conventional chemical coagulants used in the process include alum, ferric sulphate, ferric chloride, polyaluminium chloride, synthetic organic polymers, etc. [60–63].

Even though these types of coagulants have broad applications, they have also provided some shortcomings including relatively high costs, toxic effects to human beings and large volume of sludge production, which affects considerably the pH of the aquatic system [64–67]. Particularly, the alum, a common chemical coagulant which was worldwide applied in water and wastewater treatment, was reported.
to produce a large volume of sludge. These sludges react with natural alkalinity present in the water which leads to reduction in the pH [68]. In general, this process is economically viable for the removal of dyes from wastewater but sometimes becomes costly because of the cost of chemical coagulants. The major drawback of this treatment method is that the final sludge product is in large quantity as well as the removal of dye is pH dependent [69]. This treatment process is not suitable for highly soluble dyes such as reactive, azo, acid and basic dyes [70]. The synthetic polymer coagulants have also been reported to provide numerous environmental problems to the human beings. Some of the derivatives of these coagulants are non-biodegradable and their monomers are producing neurotoxic and carcinogenic effects [71].

2.2. Filtration

Filtration methods such as microfiltration, ultrafiltration, nanofiltration and reverse osmosis are important treatment methods for drinking water and wastewater applications [72–78]. These treatment methods have been studied for the removal of colour from dye wastewater [79,80]. They simultaneously remove BOD and COD of the wastewater. Each and every filtration technique is most preferable for particular types of water/wastewater treatment facilities. Generally, the microfiltration technique is not preferred in wastewater treatment because of its large pore size [81]. The ultrafiltration and nanofiltration techniques are effectively utilized for the removal of dyes from different industrial dye wastewater. The application of these treatment techniques was limited to the textile dye wastewater treatment because of the frequent clogging of dye molecules over the membrane pores. The selection of the membrane type and porosity of the filter mainly depends on the composition of the wastewater and optimum temperature required for the process; Chen et al. [82] investigated the dye removal using filtration process. The results from this study showed that the filtration membrane has given a good antifouling capacity while treatment time. The major drawback of this filtration technology includes high investment cost, generation of secondary waste streams and frequent membrane fouling [83].

2.3. Oxidation

Oxidation method is a type of treatment method for the removal of dyes from wastewater by utilizing oxidizing agents. In general, the two types such as chemical oxidation and UV-assisted oxidation using chlorine, ozone, Fenton’s reagent, hydrogen peroxide (H₂O₂) and potassium permanganate (KMnO₄) have been employed for the removal of dyes from industrial wastewater, particularly those wastewater collected from primary wastewater treatment unit, that is, sedimentation process. These treatment methods are most commonly used methods for the treatment of dye wastewater because they required only less concentration and less time to react. These methods are used to partially or completely decompose the dye molecules. But, the complete degradation of dye compounds to carbon dioxide and water was studied theoretically. It is important to note that the catalyst and pH plays a vital role in the oxidation process for the removal of dyes from wastewater.

Generally, chlorine is most widely used as a disinfectant for water treatment because of its strong oxidizing power. This can be applied in the form of calcium hypochlorite and sodium hypochlorite. It is also widely used for the reduction of colour such as pulp and textile bleaching [10]. The water...
soluble dyes such as acid, reactive, direct and metal complex dyes are easily decolourized by hypochlorite but the water insoluble dyes include disperse and vat dyes are resistant to decolourization [10,84]. Generally, the reactive dyes need longer reaction time for its decolourization but the metal complexed dye wastewater remains halfway coloured even after the reaction time period was extended. The presence of amino or substituted amino groups on the naphthalene ring of the dye molecules is most vulnerable to chlorine and decolourize more easily than other dye molecules [85]. The oxidation of dye molecules can be improved by altering the pH of the system and also by providing the suitable catalysts. For example, the degradation of metal complex dyes liberates heavy metals such as copper, iron, nickel and chromium. These discharged metals are said to possess catalytic effects for improving the decolourization of the dye molecules. The use of chlorine gas is an inexpensive treatment technique for decolourizing the dye molecules but it leads to provide unavoidable side reactions. These side reactions generate organochlorine compounds, which include toxic trihalomethane that increases the absorbable organic halogens concentration of the treated water. The metals liberated after the degradation of metal complex dyes can cause corrosion in metallic vessels.

Ozone was seemed to be a potential agent for decolourizing the textile dye wastewater [86–93]. The ozonation for the removal of colour (reactive dyes) from the dye wastewater can be achieved in shorter contact time and it is an effective process for the harmful compounds discharged from the conventional reactive dyes [94]. The decolourization of reactive dye was studied and the ozonation effectively removed colour and chemical oxygen demand [88,95]. However, in some cases the ozonation does not influence the removal of chemical oxygen demand significantly [94,96]. The oxidation potential of ozone was observed as 1.5 times that of chlorine [97]. This treatment does not liberate the chlorinated disinfection by-products which are carcinogenic. Therefore, ozone technology was found to have high potential because of its high technical feasibility. However, the wide application of ozone in water and industrial effluent treatment was limited due to its high cost operation. This may be due to the mass transfer limitations stemming from the low solubility of the ozone and improper selection of the contactors without consideration of the operative reaction system. Ozone is normally generated in bulk quantities by high voltage electrical discharge across a thin gap of dry air or oxygen. The electrical discharge process can produce in addition to ozone, other short-lived species such as OH radicals, OH +, ion, atomic hydrogen, electrons and oxygen atomic radicals [98–100]. The liberation of these species mainly depends on the reactor configuration and the voltage used. These reactive species can improve the rate of oxidation if they can react with the pollutants in the fluid before they decay [97].

Hydrogen peroxide is an extremely pale blue colour liquid and it gets converted to colourless liquid in a dilute solution. In addition, it is slightly more viscous than water. This hydrogen peroxide has potential oxidizing properties [10]. This is a powerful bleaching agent for bleaching paper besides other applications. In the year 1994, nearly 50% of the world’s production of hydrogen peroxide was applied for paper and pulp bleaching [101]. This is also used for producing the peroxide enzymes, which are utilized for decolourization of dyes [102]. But this process depends on the pH and generates waste.

Fenton's reagent, a mixture of an iron catalyst and hydrogen peroxide, was applied to decolourize the coloured wastewater [103–107]. This reagent is stronger than hydrogen peroxide agent. Normally, this Fenton's reagent is effective in decolourization of both soluble and insoluble dye molecules. Some of the dyes including vat and disperse dyes are highly resistant against Fenton's reagent. The dyes such as remazol brilliant blue B, indanthrene blue GCD, sirrus supra blue BBR, helixazin blue BGT and irgalan blue FGL have been studied and reported that these dyes were significantly decolourized by Fenton's reagent [10]. It is also identified that the colour is not only removed but also observed that the total organic carbon, chemical oxygen demand (except for reactive dyes) and toxicity have been reduced. This treatment technique also applied for high suspended solids concentration and is suitable for wastewater treatment when the municipality allows the release of Fenton's sludge into sewage. In the aspect of biological point of view, the quality of the sludge is not only enhanced but the removal of phosphate was also observed. The major drawbacks of this treatment technique is that it is generally effective within the narrow pH range of less than 3.5 which involves the sludge production and required longer reaction time [108]. Advanced oxidation processes (AOPs), which include the generation of exceedingly responsive hydroxyl radical (HO·), have developed as outstanding water and wastewater treatment innovation for the decomposition or mineralization of a various scope of natural contaminants. A class of AOP comprises of photoactivated forms. The photoactivated responses are described by the free radical instrument started by the association of photons of a legitimate vitality level with the impetus photocatalytic oxidation). The present audit means to give a complete investigation on the system of UV-TiO2 photocatalytic oxidation prepare, photocatalyst material, light sources and the sorts of photoreactors [109]. The proficiency of the framework is likewise influenced by the method of TiO2 application as immobilized on surface or as suspension.

2.4. Electrochemical methods

Electrochemical treatment falls under tertiary treatment technique which also employed for the removal of colours from dye wastewater [110–112]. The main advantage of this treatment technology is its environmental compatibility due to the fact that its main reagent, that is, electron, is a clean reagent. Other advantages include its flexibility, high energy efficiency, amenability of automation and safety. The decolourization of the dye wastewater is attained either by electro-oxidation with non-soluble anodes or by electrocoagulation with soluble anodes [113–115]. The electrochemical treatment in dye removal is highly depending on the anode materials. Based on the anodic materials utilized, the efficiency will increase [116]. Some of the anodic materials include iron, boron-doped diamond electrode, etc., with various operating conditions have been employed for the electro-degradation of dye molecules [117–119]. The removal of colour from Direct Red 80 dye wastewater using three
different electrode materials including iron, polypyrrole doped with chromium and boron-doped diamond electrode. This treatment method is active in the removal of colour from soluble and insoluble dyes with the reduction of chemical oxygen demand. It was observed that the extent of colour and organic load removal mainly depends on the types of anodic materials and the applied voltage. However, the main disadvantages are high cost for electricity, sludge production and pollution because of chlorinated organic and heavy metals due to indirect oxidation.

2.5. Advanced oxidation processes

AOPs are extremely efficient and novel treatment methods for the rapid elimination of many organic and inorganic substances [120–122]. Recently, AOPs have appeared as potentially effective treatment techniques for converting the organic pollutants into non-toxic substances [123–126]. This process is involving in the simultaneous utilization of more than one oxidation processes because in certain cases the single oxidation system is not enough for the total decomposition of the dye molecules. These AOPs utilize strong oxidizing agents such as hydroxyl radicals generated in situ which causes the series of reactions thereafter to convert the macromolecules into smaller and less harmful substances [125,126]. In most cases, the macromolecules are completely mineralized into carbon dioxide and water. The important advantage of AOPs as compared with the other treatment techniques is its easiness to handle and the generation of significantly less residues. The AOP techniques include ultraviolet (UV) photolysis, Fenton’s reagent oxidation and sonolysis. Nidheesh et al. [127] conducted a study on advanced oxidation process in which they have explained about the anodic oxidation, electro-Fenton, peroxicoagulation, fered Fenton, anodic Fenton, photolelectro-Fenton, sonoelectro-Fenton, bioelectro-Fenton, etc. This study mentioned that complete oxidation of dyes is possible in the advanced oxidation process. This treatment technique has the capability of degrading the dye molecules at ambient temperature and pressure. These AOPs have an advantage over the biological wastewater treatment for waste effluents containing toxic or bioinhibitory contaminants. The different varieties of AOPs are used in chemical oxidation processes utilizing ozone, combined ozone and hydrogen peroxide, UV enhanced oxidation such as UV/hydrogen peroxide, UV/ozone, UV/air wet oxidation and catalytic wet air oxidation (in this case, air is used as the oxidant). 10%–20% of colour was removed while using UV alone but when it was utilized along with hydrogen peroxide nearly 90% of the colour gets removed [128].

The combination of Fenton reaction and UV is called as photo-Fenton reaction [129]. The combination of these two treatment technologies is available to enhance the elimination of colourants from the dye wastewater [130–135]. The photo-Fenton reaction is highly effective than Fenton reaction at an optimum condition [130]. Photocatalysis is another AOP for the removal of pollutants from the waste stream [136–142]. In this treatment technique, the light energy from a light source triggers an electron from the valence band of the catalyst to the conduction band with a series of chemical reaction which results in the generation of hydroxyl radicals. The generated hydroxyl radicals have higher oxidizing potential which can easily oxidize most of the organic compounds. The different chalcogenides, that is, oxides such as TiO₂, CeO₂, ZrO₂, ZnO, etc. or sulphides such as ZnS, CdS, etc. have been utilized as a photocatalyst for the removal of different varieties of dyes including direct, reactive, vat and disperse dyes. Sonolysis is another treatment technique which utilizes the ultrasonic waves to decolourize and degrade the dye molecules [143–147]. The mechanism of sonolysis generally depends on the formation of short-lived radical species generated in violent cavitation events. The major drawbacks of AOPs are the generation of some unexpected by-products, complete mineralization is not possible and the process depends on the pH. The limitation of the process varies depending on the types of AOPs implemented for the particular type of wastewater. For example, the important operating parameters for the removal of colour by UV/H₂O₂ are hydrogen peroxide concentration, residential time, pH, UV radiation intensity, chemical structure of dye molecules and dye bath additives. However, the AOPs have proven to be effective and outstanding treatment technologies for the removal of colour from effluents but they are quite expensive especially for small-scale industries of developing countries.

2.6. Biological methods

Biological treatment technique is the most common and widely adopted treatment techniques for the removal of colour from the dye wastewater [148–158]. A variety of species have been employed for the decolourization and mineralization of the different dye molecules. The biological method provides significant advantages such as comparatively inexpensive, less operating costs and non-toxicity of the completely mineralized end products. These biological methods are classified into three categories, that is, aerobic, anaerobic, and combined effect of aerobic and anaerobic treatments.

The aerobic treatments are carried out in the presence of oxygen. The aerobic microorganisms such as bacteria and fungi are the two most important organisms that have been utilized for the effective treatment of dye wastewater. At aerobic situations, the enzymes secreted by the bacteria available in the wastewater breakdown the organic compounds. The identification, isolation and application of the aerobic bacteria are important steps in the biological treatment of different types of dye wastewater [158,159]. The different types of triphenylmethane dyes include malachite green, brilliant green, crystal violet, magenta, pararosanilin and ethyl violet have been effectively removed by the strain Kurthia sp. [160]. However, it is important to note that the synthetic dyes are not uniformly vulnerable to decomposition by activated sludge process in a conventional aerobic process. An attempt has been implemented to produce specific aerobic bacterial strains for the decolourization of the specific dye structure [161]. The fungal strains seemed to be capable of decolourizing the azo and triphenylmethane dyes [158–163]. Among the different fungi strains, Phanerochaete chrysosporium has been studied exclusively for the past two decades because of its ability to decolourize the different varieties of dyes [158,159,164,165]. Further, the microorganisms including Cyathus bulleri, Rhyzopus oryzae,
Funalia trogii, Coriolus versicolor, Streptomyces sp., Laetiporus sulphureus, Trametes versicolor and other microorganisms have also been employed for the decolourization of dye wastewater [158,166–168]. The different operating parameters such as concentration of dye wastewater, initial pH and wastewater temperature affects the biological treatment of dye wastewater. The biological treatments are compatible for certain dyes and most of the dyes are recalcitrant to the biological breakdown or non-transformable under aerobic conditions [158,159,169]. The anaerobic treatments are carried out in the absence of oxygen. The anaerobic treatments have been applied for the decolourization of the different varieties of synthetic dyes [158,159,170–173]. In some cases, the anaerobic pre-treatment step is the cheapest and alternative to the aerobic system [171]. The expensive aeration and problems with the bulk sludge are avoided in the anaerobic system. However, the major drawbacks of this treatment techniques are biological oxygen demand removal is inadequate, incomplete mineralization of dyes, incomplete nutrient removal and sulphates are converted into sulphides [171]. The combined treatment of aerobic and anaerobic was implemented to effectively decolourize the textile industrial wastewater. The important advantage of this method is to provide complete mineralization which is frequently achieved due to the synergistic action of different organisms [158,174]. The reduction of azo bond was observed during the anaerobic conditions and the resulting colourless aromatic amines can be mineralized under aerobic conditions [175]. Hence, for the effective treatment of dye, the anaerobic decolourization followed by aerobic treatment was generally preferred [176]. Some of the major drawbacks of this biological treatment technologies are requirement of large land area, less flexibility in design and operation, prolonged time needed for decolourization–fermentation processes thereby making it unable to remove the dyes from wastewater on a continuous mode [158,177–179].

2.7. Ion exchange

Ion exchange is a reversible process where an ion from the solution is exchanged for a similarly charged ion attached to the solid resin [180]. This treatment was generally adopted for softening the salt water to provide drinking water [181]. Zeolites are one of the most important ion exchange materials used worldwide. This material is available naturally and also this was prepared synthetically. In 1934, Adams and Holmes found that the phenol–formaldehyde resin has cation exchange properties. This gives important information regarding the preparation of different types of resins including cationic and anionic ion exchange resins. Some of the polymeric materials holds an ion exchange mechanism which include polystyrene sulphonate, polystyrene phosphonate, polystyrene amidoxime, sulphonated phenolic resin, phenolic resin, polystyrene-based trimethyl benzyl ammonium, epoxy-polylamine and aminopolystyrene. The batch and fixed bed column studies have been employed for the elimination of contaminants from water.

The different experimental studies have been conducted for the removal of dyes from wastewater using the ion exchange resins [182–199]. The major drawbacks of ion exchange method are incapable of handling highly concentrated wastewater, matrix gets fouled by organics, non-selective and highly sensitive to pH of the wastewater.

2.8. Adsorption

Adsorption is a surface phenomenon and it is the process for the separation of mixtures on a laboratory and industrial scale which can be explained as the increase in concentration of a particular component at the interface between the two phases. The adsorption process has been effectively utilized for the removal of colour from wastewater [200–235].

The term ‘adsorption’ refers to process in which a material is concentrated over the solid surface from the liquid or gaseous surroundings. Adsorption on porous carbon was explained as early as 1550 B.C. in an ancient Egyptian papyrus and afterwards by Hippocrates and Pliny the Elder, which is importantly for medicinal applications. However, on scientific records the adsorption phenomenon was explained by Scheele in 1773 for gases exposed to carbon [200,236].

In 1785 it was continued by Lowitz for the reversible removal of colour and odorous compounds from water by wood charcoal. Larvitz in 1792 and Kehl in 1793 identified similar behaviour with vegetable and animal charcoals, respectively. However, for the first time in 1881, Kayser has introduced the term adsorption to differentiate the surface accumulation from intermolecular penetration. In addition, he explored that the adsorption process is a surface accumulation of material. Generally, it is important to differentiate the various types of adsorption process. If the attraction between the adsorbent and adsorbate is due to weak van der Waals forces, then this type of adsorption process is called as physical adsorption or physisorption. This type of adsorption process is a reversible process. Also, if the attractive forces are due to covalent bond then this type of adsorption process is called chemical adsorption or chemisorption. Because of the high bonding strength between the adsorbent and adsorbate in chemisorption process, it is very difficult to remove the adsorbed species from the solid surface. Generally, a good adsorbent should possess a porous structure. The time required to achieve the adsorption equilibrium should be as small as possible only then the dyes can be removed in lesser time.

Basically, adsorption of dyes mainly depends on the properties of the dyes and the surface properties of the adsorbent materials. Activated carbon (AC) is widely applied in the removal of dyes from wastewater. The commercial activated carbon produced from the different sources includes coal, coconut shells, charcoal, lignite, wood and sawdust. The two types of AC such as H-type and L-type have been produced. The H-type is the positive charge upon water and hydrophobic nature. The L-type is a negative charge upon water and hydrophilic in nature [202,237,238]. The activation of materials was carried out by partial oxidation for the purpose of developing pore structures in it. There are two types of activation which include physical and chemical activation.

Usually, the physical activation procedure needs high temperature and longer activation time as compared with the chemical activation. The AC requires a complete washing to remove the excess acids in the chemical activation method. AC can be in granular form, that is, granular activated
Activated carbon (AC) has been widely used as adsorbent for the removal of dyes from wastewater but its high cost poses an economical problem. Hence, there is a need to produce cheap and easily available materials, which can be utilized more economically on large-scale operations. Owing to the economical problems, the research interest into the development of alternative adsorbents to substitute the costly AC has intensified in recent years. The attention has focussed on different natural solid materials, which are able to remove the dyes from wastewater at cheaper cost. Cost is an essential factor for comparison and selection of the suitable adsorbents. An adsorbent can be measured as a cheap materials if it requires the modest processing and available abundantly in nature. This may also be explained as an agricultural by-product or waste material from the industries and requires additional disposal cost. Some of the waste products from agricultural operations and industries, natural materials and biomaterials have been identified as potentially economical alternative adsorbents. Most of these materials have been employed for the removal of dyes from wastewater.

3. Adsorbent from agricultural wastes

Agricultural wastes are lignocellulosic compounds which mainly consist of cellulose, lignin and hemicelluloses [280,281]. Lignin is a polymeric compound that consists of aromatics and has a complex three-dimensional structure. The phenylpropane units in the lignin compounds are bound together to produce a very complex matrix. It consists of different functional groups including hydroxyl, methoxy and carbonyl. The hemicellulose in the agricultural wastes is mostly soluble in alkali and more easily hydrolyzed. Also both hemicelluloses and cellulose consist of oxygen functional groups including hydroxyl, ether and carbonyl [282]. These functional groups play an essential part in the generation of adsorbents. Previously, some of the research scientists have utilized various precursors for the synthesis of ACs including waste plastics (polyethylene terephthalate and polyvinyl chloride), industrial wastes (polymeric residues, fly ash, pitch, etc.) and other wastes (sewage sludge, tires, etc.) [283]. Generally, it is an important task to produce AC with low cost for the commercial manufacturing industries.

Recently, some of the research groups have adopted agricultural wastes to produce AC because they contain high carbon content, low inorganics, require mechanical strength, low levels of ash content and low cost [281–285]. These materials are available in large amounts and are one of the most abundant renewable resources in the globe. These waste materials have small or no economical importance and often provide the disposal problem. Hence, there is an important requirement to valorize these low cost materials. The transformation of these waste materials into ACs would provide the significant economic value which helps to reduce the cost for the disposal of waste and most importantly which provides a potential low-cost alternative to the existing commercial ACs. These waste materials have been effectively utilized as raw materials for the preparation of AC with high adsorption capacity, required mechanical strength and low ash content [286,287]. Adsorption process utilizes the solid material as an adsorbent for the removal
of different types of contaminants from wastewater. Among the different materials proposed as an adsorbent, AC is the most accepted material for the removal of pollutants from wastewater. AC is an amorphous material which is having high degree of porosity and these pores have been produced during its manufacture. The high porosity leads to removal of large amount of pollutants from wastewater [288–294]. The important factors considered for the selection of agricultural adsorbents are its low cost, availability and organic composition which provide higher affinity towards the specific dye wastewater. After utilized for wastewater treatment, these materials can be disposed to the environment without any complicated and expensive regeneration process because of their low cost.

The different types of agricultural waste materials have been utilized for the preparation of AC which includes corn straw [295], wheat straw [295], rice straw [296,297], sawdust [298–300], corncob [301,302], bagasse [303,304], coketex, rice husks [298,306,307], tobacco husk [308], nut shells [309–311], soyabean oil cake [312], oil palm shell [313], oil palm fibre [314], vegetable residues [315], etc. Generally, there are two important steps in the synthesis of AC which include (i) carbonization of the materials below 1,000°C at an inert atmosphere and (ii) activation of the carbonized material (physical or chemical activation).

Physical activation is a method in which the forerunner is converted into AC utilizing gasses and is for the most part done in a two-stage handle [316]. Carbonization is the primary stage which involves the formation of a char (non-porous) by pyrolysis of the raw material at a temperature range of 400°C–850°C and sometimes it may be operated at 1,000°C in the nitrogen atmosphere. Activation is the second stage which includes making the contact of the char with an oxidizing gas, for example, carbon dioxide, steam, air or their blends, in the temperature of somewhere around 600°C and 900°C, which brings about the expulsion of the more complicated carbon and the arrangement of an all around created micro-pore structure. Generally, CO2 is the activation gas because it is costless, simple to handle and it encourages control of the activation process on account of the moderate reaction rate at temperatures around 800°C [316]. It is beneficial to note that the ACs prepared by physical activation did not have acceptable qualities to be utilized as adsorbents or as filters [284].

Chemical activation is a method, in which the carbonization and activation are happening simultaneously, with the precursor being blended with chemical activating agents as dehydrating agents and oxidants. This activation includes impregnation with chemicals, for example, H3PO4, ZnCl2, NaOH, KOH and K2CO3 followed by heating under a nitrogen stream at temperatures in the scope of 450°C–900°C, contingent upon the impregnant utilized. This activation method was carried out in a single step and at lower temperatures. This method is available to prepare the superior porous structure in the adsorbent with the environmental concerns of using chemical agents for activation could also be produced. In addition, part of the utilized chemicals was easily recovered [285,316,317]. A two-stage process was available for the preparation of AC which includes physical and chemical processes [318]. It is also identified that an additional one more treatment step is available, that is, steam pyrolysis [319–323]. This treatment can be available in two forms: (i) the raw agricultural waste is heated at moderate temperatures between 500°C and 700°C under a flow of pure steam and (ii) the raw agricultural waste is heated at 700°C–800°C under a flow of steam. Numerous agricultural wastes include bagasse, straw, nut shell, grape seeds, almond shells, coconut husk, corn stover, oat hulls, peanut cherry, stones, apricot stones and hulls have been experimented with this procedure [324–337].

The recent review on literature indicates that the agricultural waste showed comparatively adequate removal efficiency as compared with the commercial materials. Dye decolourization method is not specific and depends on many parameters. There are many agricultural adsorbents available and which can act as alternatives to the existing AC but complete replacement is not possible. Generally, the selection of agricultural wastes is based on its low cost, availability and organic composition which shows the strong affinity for some identified dye wastewater.

3.1. Adsorbent produced from lignin

Lignin is a natural polymer composed mainly of aromatic compounds. It is available along with hemicelluloses goes about as a binding agent of cellulose strands in the structures of plants. The role of lignin is to give structural quality, give fixing of water directing framework that connects roots with leaves, and ensure plants against debasement [338]. It is a macromolecule, which comprises of alkylphenols and has an unpredictable three-dimensional structure. Lignin is covalently connected with xylans on account of hardwoods and with galactoglucomannans in softwoods [339]. The essential chemical phenylpropane units of lignin (principally syringyl, guaiacyl and p-hydroxy phenol) are fortified together by an arrangement of linkages to frame an extremely complex matrix. It contains an assortment of functional groups, for example, hydroxyl, methoxyl and carbonyl, which bestow a high extremity to the lignin macromolecule [340–345].

Lignin, a waste material or by-product released from paper mills in substantial amounts is utilized as a part of its raw state and also altered state for evacuation of contaminants by researchers/scientists. A spherical sulphonic lignin adsorbent was produced from a bamboo pulp mill by-product. This adsorbent was examined for the expulsion and recuperation of cationic dyes that include cationic turquoise GB, cationic red GTL and cationic yellow X-5GL from fluid arrangements [346]. Some of the research works are also available to explain the utilization of lignin as an effective adsorbent for the removal of dyes from wastewater [347–350].

3.2. Adsorbent produced from sawdust

Sawdust is a standout amongst the most engaging materials among agricultural waste materials, utilized for evacuating toxins, for example, colours, salts and overwhelming metals from water and wastewater [351]. The material comprises lignin, cellulose and hemicellulose, with polyphenolic groups assuming critical part to bind colours through various systems. By and large the adsorption happens by ion exchange, complexation and hydrogen bonding,
Different productive studies have been done on the expulsion of colours by sawdust [352–354]. Several researchers have used this saw dust in natural and its modified form for the removal of dyes from wastewater [355–363]. Many adsorption kinetics and isotherm models for the dye removal by sawdust have been reported by the researchers. Also the thermodynamics on the removal of dyes by the sawdust has been presented.

3.3. Adsorbent produced from rice husk

Rice husk is an agricultural waste which was obtained from the rice processing industry as a by-product. Generally, these industries are generating more than 100 million tonnes out of which 96% of the waste is being produced by the developing countries [286]. The usage of this wellspring of agricultural waste would take care of both a disposal issue and additionally access to a less expensive material for adsorption in water contaminations control framework [364–366]. The principle parts of rice husk are carbon and silica (15%–22% SiO2), it can possibly be utilized as an adsorbent [37,367,368]. Many researchers have been using rice husk as an effective adsorbent for the removal of dyes from wastewater [366,369–378].

3.4. Adsorbent produced from waste peels

Among a few agricultural wastes that have been experimented on as adsorbents for wastewater treatment, the “waste peels” from fruits and vegetables are of awesome significance since the vast majority of the peels are disposed of as waste and discover no application anywhere, which here and there posture genuine transfer issues [2]. Most of the waste from homes mainly consist of these peel wastes. Additionally these wastes were generated from the fruits and vegetable industries during the process of selecting, sorting and boiling processes. Generally, these wastes are disposed into the environment or fed to the domesticated animals. The fruits and vegetable wastes and its by-products are produced in large quantity during the industrial operations which provide an impact over the environment and these wastes should be properly handled. Then again, they are extremely rich in bioactive segments that are considered to beneficially affect wellbeing [379]. These waste peels are natural, eco-friendly and monetary wellsprings of adsorbents. These wastes can be used in the expulsion of colours from wastewater.

Some of the waste peels include citrus peel waste (grapefruit peel waste, pomelo peel waste, orange peel waste and lemon peels), banana peel waste, cassava peel waste, jackfruit peel waste, pomegranate peel waste and garlic peel wastes have been applied for the removal of different kinds of pollutants from aqueous solution [2,380–389]. Adsorption influencing parameters for the removal of dyes from wastewater have been studied by the researchers. Adsorption equilibrium, kinetics and thermodynamics for the removal of colour from wastewater have also been investigated.

3.5. Adsorbent produced from other agricultural wastes

The numerous endeavours to discover cheap and effortlessly accessible adsorbents to evacuate the toxic pollutants from wastewater, for example, the agricultural wastes whereas per their physical-chemical attributes and ease they might be great potential adsorbents [11,217]. Agricultural preparations are accessible in expansive amounts the world over. Because of this, the huge amounts of agricultural wastes are rejected. These agricultural wastes are not only utilized in the removal of toxic dye from wastewater but also employed for the removal of various organic and inorganic toxic pollutants from wastewater.

Lignocellulosic materials comprise of three principle basic parts which are lignin, cellulose and hemicelluloses. Lignocellulosic materials additionally contain extractive basic segments which have small molecular size. Distinctive adsorbents obtained from agricultural wastes have been utilized for colour expulsion from wastewater and numerous investigations of colour adsorption by agricultural wastes are available. Agricultural and industrial processing units discard a lot of untreated waste which may contaminate the land, water and air, and accordingly harm the environment.

Then again, ill-advised treatment of these wastes causes comparable issues. Along these lines, administrative control of contaminations ought to be established to forestall or to minimize the exchange of unsafe material to different territories. Hence inside the most recent couple of years numerous thoughts have been acquainted all together with appropriately discard these wastes, for example, serious use as adsorbents for poison expulsion particularly for colour evacuation where it demonstrated high adsorption limit. These are renewable, accessible in vast sums and low cost when contrasted with different materials utilized as adsorbents. These wastes are superior to different adsorbents in light of the fact that the agricultural wastes are generally utilized without or with at least handling (washing, drying, crushing) and in this way lessen generation costs by utilizing a shoddy crude material and disposing of vitality expenses connected with warm treatment.

Some of the agricultural wastes derived from low cost and readily available resources such as grass waste, rice straw, barley straw, guava leaves, papaya seeds, plant leaf powder, banana waste, pumpkin seed palm kernel fibre, castor seed shell, pineapple stem, pumpkin seed hull, coffee husks, coconut husk, dehydrated peanut hull, Parthenium hysterophorus, coconut bunch waste, hazelnut shells, peanut hull, soy meal hull, yellow passion fruit waste, fruit juice residue, Luffa cylindrica fibres, jute waste, cereal chaff, wheat straw, bagasse, rubber seed shell, wheat shells, apricot shell, walnut shell, almond shell, cashew nut shell, coconut shells, citric acid, cotton and gingelly seed shell, lentil shell, rice shell, pine sawdust, groundnut shell, pongam seed shell, olive stone, coir pith, cane pith, plum kernel, sunflower stalk, white rice husk, white ash, wood-derived biochar, pinewood, neem bark, sky fruit husk, carbon cloth, sunflower hull, wheat stem, etc. have also been successfully adopted for the removal of colour from wastewater [287,399–411].

4. Adsorbent from industrial wastes

Generally industrial exercises create enormous measure of strong waste materials as by-products. Some of the by-products are reused and the remaining are disposed as landfills. In this manner, the likelihood of reuse in adsorption
forms speak to a fascinating arrangement primarily in light of the fact that these industrial waste materials are accessible free of cost and cause significant disposal issue. As of late, various industrial wastes have been examined with or without treatment as adsorbents for the expulsion of poisons from wastewaters. Some of the industrial wastes include fertilizer industry wastes, fly ash, leather industry wastes, steel industry wastes, aluminium industry wastes and paper industry wastes [412]. Fly ash is a waste material generating from ignition operations. The primary employments of fly ash incorporate development of streets, blocks, concrete and so forth. The high concentration of silica and alumina in fly ash debris makes it decent contenders for use as a modest adsorbent for mass use [411,412]. The blast furnace slag, sludge and dust (steel industrial wastes) are the most experimented materials in the adsorption process. Red mud (aluminium industry waste) is a waste material framed amid the generation of alumina when the bauxite mineral is subjected to caustic leaching [413]. The poisonous quality and colloidal nature of red mud particles make a genuine contamination peril. Numerous propositions for the red mud use, for example, in the preparation of red mud blocks, as filler in black-top street development, as iron metal, and as a wellspring of different minerals [412]. After pre-treatment, the red mud is appropriate for the removal of Congo red from aqueous solution [413]. Rather, fertilizer industry additionally delivers various by-products in substantial concentrations which make genuine transfer issues and debase the encompassing environment. The industrial wastes have been successfully employed for the removal of dye pollutants from wastewater [414–435]. Because of the minimal effort and high accessibility of these wastes, it is not vital to have complicated regeneration operations. This type of low-cost adsorption methods have been attracted by many researchers/scientists. Be that as it may, the majority of the times the adsorption capacities of such adsorbents are not high, and thus, the study and investigation of more new adsorbents are still under advance. Diverse innovative absorbents used nowadays in the elimination of dyes and other toxic pollutants from aqueous solutions are depicted in Table 2. The objective of the present research is to create modest and viable adsorbents from agricultural waste, as another option to the current adsorbents.

5. Conclusion and future perspectives

This review article clarifies the various methodologies involved in the elimination of toxic dyes. This article mainly concentrates on the various economically feasible treatment technologies available for the treatment of dyes. Various methods such as filtration, coagulation, flocculation, oxidation, advanced oxidation process, electrochemical methods, ion exchange, biological methods and adsorption methods have been used in the treatment of industrial dye effluents. On analyzing all the above methodologies the physicochemical methods are seemed to be somewhat effective but they are not used often due to their complexity and high cost. The chemical methods involve some more chemicals in the process and create secondary pollution which cannot be treated easily. Eventhough, the biological methods are

<table>
<thead>
<tr>
<th>Dyes</th>
<th>Biosorbent</th>
<th>Adsorption capacity (mg g⁻¹)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chromium (VI)</td>
<td>Mixed waste tea</td>
<td>94.34</td>
<td>[436]</td>
</tr>
<tr>
<td></td>
<td>Coffee ground</td>
<td>87.72</td>
<td></td>
</tr>
<tr>
<td>Diamine Green B</td>
<td>Rice husk cellulose</td>
<td>207.15</td>
<td>[437]</td>
</tr>
<tr>
<td>Acid Black 24</td>
<td></td>
<td>268.88</td>
<td></td>
</tr>
<tr>
<td>Congo Red</td>
<td></td>
<td>580.09</td>
<td></td>
</tr>
<tr>
<td>2-Picoline</td>
<td>Almond shell and orange peels</td>
<td>166.7</td>
<td>[438]</td>
</tr>
<tr>
<td></td>
<td>activated carbon</td>
<td>288.57</td>
<td></td>
</tr>
<tr>
<td>Crystal violet</td>
<td>Peanut hull waste</td>
<td>100.6</td>
<td>[439]</td>
</tr>
<tr>
<td>Methylene blue</td>
<td>Cotton waste activated carbon</td>
<td>369.48</td>
<td>[440]</td>
</tr>
<tr>
<td>Eriochrome black T</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Rhodamine B</td>
<td>Chlorella pyrenoidosa</td>
<td>63.14</td>
<td>[441]</td>
</tr>
<tr>
<td>Methylene blue</td>
<td>Phragmites australis</td>
<td>41.2</td>
<td>[442]</td>
</tr>
<tr>
<td>Procion red H-E7B</td>
<td>Nigrospora biomass</td>
<td>188.79</td>
<td>[443]</td>
</tr>
<tr>
<td>Reactive blue BF-5G</td>
<td>Malt bagasse</td>
<td>42.58</td>
<td>[444]</td>
</tr>
<tr>
<td>Sulphur blue 15</td>
<td>Acidithiobacillus</td>
<td>1,428.6</td>
<td>[445]</td>
</tr>
<tr>
<td>Congo red</td>
<td>Aspergillus carbonarius</td>
<td>99.01</td>
<td>[446]</td>
</tr>
<tr>
<td></td>
<td>Penicillium glabrum</td>
<td>101.01</td>
<td></td>
</tr>
<tr>
<td>Methylene blue</td>
<td>Eucheuma spinosum</td>
<td>833.33</td>
<td>[447]</td>
</tr>
<tr>
<td>Reactive red 198</td>
<td>Nostoc linckia HA 46 biomass</td>
<td>93.5</td>
<td>[448]</td>
</tr>
<tr>
<td>Amaranth dye</td>
<td>Water hyacinth</td>
<td>70</td>
<td>[449]</td>
</tr>
<tr>
<td>BEZAKTIV Red S-Max</td>
<td>Phoenix dactylifera rachis</td>
<td>196.08</td>
<td>[450]</td>
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</table>
economically feasible, this process is not preferred as it is time consuming and skilled labour. In midst of all the treatment technologies used in the wastewater treatment, the adsorption process is seemed to be the efficient technique in the elimination of the industrial dyes. This technique is simple, feasible, effective, efficient and eco-friendly in dye removal. The adsorbents used in this process are prepared from simple sources such as saw dust, agricultural wastes, lignin, waste peels, activated carbon and industrial waste which are all easily affordable and available in nature. These adsorbents possess high adsorption capacity and it is easily rejuvenated using simple chemicals. More research works have to be carried in this sector to lower the harmful effects of this textile dyes. The different surface modification methods must be optimized to prepare the efficient adsorbent for the elimination of dyes from industrial wastewater. Most of the researches on dye removal using low cost methods are dealt with batch adsorption process and this information must be designed for the scale up studies. The life cycle analysis of the prepared material must be evaluated to check the repeatability of the adsorbent usage and its disposal scenario. The regeneration procedure for the spent adsorbent must be properly devised to predict the proper regenerating methods.

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