



A critical review of textile industry wastewater: green technologies for the removal of indigo dyes

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Abstract

The denim textile industry represents an important productive sector. It generates wastewater with low biodegradability due to the presence of persistent pollutants, which can produce toxic and carcinogenic compounds; therefore, wastewater treatment reduces risks to aquatic life and public health. This paper presents a review of 172 papers regarding textile industry wastewater treatment for the removal of contaminants, especially indigo dyes used in the denim industry, in the context of green technologies. The physicochemical characteristics of textile wastewater, its environmental and health impacts, and the permissible limit regulations in different countries were reviewed. Biological, physicochemical and advanced oxidation processes for the removal of indigo dyes were reviewed. The goal of this study was to analyze the characteristics of green technologies; however, the research does not clearly demonstrate an effect on energy consumption savings, carbon footprint decreases, and/or waste generation. Advanced oxidation processes showed the highest color removal efficiency (95 and 97% in synthetic or real wastewater, respectively). Photocatalysis and Fenton reactions were the most efficient processes. None of the revised works presented results regarding upscaling for industrial application, and the results should be discussed in terms of the guidelines and maximum permissible limits established by international legislation. New technologies need to be developed and evaluated in a sustainable context with real wastewater.

Keywords Advanced oxidation processes · Biological processes · Dye removal · Industrial wastewater · Physicochemical treatment

Introduction

The textile industry represents economic activity for the manufacture of yarns, fibers, fabrics and other materials to obtain derivative products, such as clothing (Hassaan and Nemr 2017). This industry employs 35 million people worldwide and contributes 7% to total exports (Desore and

Narula 2018); therefore, it represents an important economic sector. Denim is one of the most important components of the textile industry because it is extensively used. It is a fabric made from colored cotton threads, usually of a blue shade (Chavan 2015; Fidan, Aydoğan and Uzal 2021). Color is the most important physicochemical characteristic, and it is obtained as part of the garment washing process in the textile finish (Periyasamy and Militky 2017; Madhav et al. 2018). The textile industry is considered a major source of wastewater due to the release of pollutants, such as non-biodegradable dyes (Naga Babu et al. 2019). The presence of metals such as chromium, copper, zinc, and manganese is common in textile processes to fix dyes through chemical complexation (Paul 2015; Uddin and Sayem 2020). Synthetic dyes, such as indigo blue (IB) and indigo carmine (IC), used in denim are recalcitrant dyes that are very difficult to remove from wastewater and chemically and photolytically stable (Quintero and Cardona 2010). The discharge of textile wastewater into the

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environment can cause skin and eye irritation, corneal and conjunctiva lesions, dermatitis, and cancer in humans with constant and prolonged contact (Khadhri et al. 2019).

The processes used for textile dyeing require high water consumption, and therefore the generation of wastewater (Paździor, Bilińska and Ledakowicz, 2019), so the removal of contaminants is necessary. Biological, physicochemical, and advanced oxidation process (AOP) treatments are generally evaluated in the removal of dyes and other pollutants in real or synthetic wastewaters (Legerská et al. 2016; Sivakami et al. 2018; Ramos et al. 2020). The color removal efficiencies of biological processes were as high as 100% in synthetic wastewater (Paz et al. 2017), and these efficiencies were lower in textile wastewater (78.4%) (Shoukat, Khan and Jamal, 2019). Balcik-Canbolat et al. (2017) obtained 100% color removal by nanofiltration and reverse osmosis processes in textile wastewater, and Yu et al. (2018) removed 100% of color by membrane filtration in synthetic water. Photoelectrocatalytic processes achieved 100% color removal in synthetic water (Oriol et al. 2019), and a Photo-Fenton process achieved over 98% color removal in wastewater (Lebron et al. 2021).

Different green technologies (GTs) have been proposed for textile wastewater treatment and the removal of IB and IC dyes from a sustainable approach (Nambela, Haule and Mgani, 2020). GTs mitigate the negative consequences of conventional treatment processes (Wang et al. 2019); however, this assertion must be clearly demonstrated (Gallego-Schmid and Tarpani 2019). Therefore, it is important to define the characteristics that a GT should meet for textile wastewater treatment. Treatment technologies must be developed to comply with international and national legislation. Governments have established maximum permissible limits for a group of pollutants in textile wastewater (U.S. Environmental Protection Agency 1982; Environmental Protection Administration Republic of China 2019; Comisión Técnica del Agua del Estado de México 2020a).

This review compiled 172 references extracted from the databases ScienceDirect, SpringerLink, PubMed, PubChem, NCBI, Taylor and Francis, Google Scholar, MedlinePlus, HubMed, and EBSCO. Recent technologies (2016–2022) for the removal of synthetic dyes, water and wastewater from the denim textile industry were analyzed. The objective of this review was to analyze the

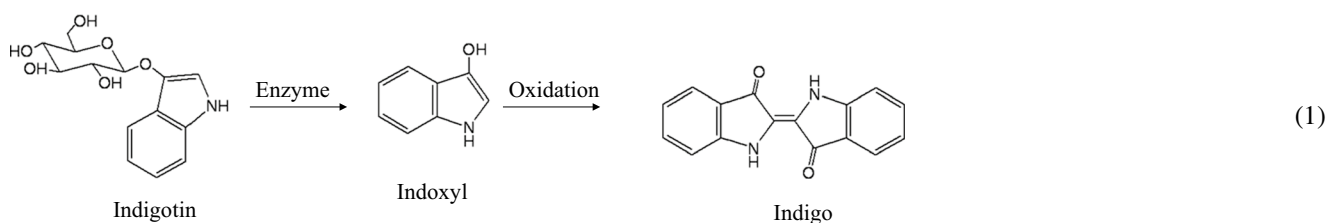
physicochemical characteristics of denim wastewater, its environmental and health impacts and the permissible limits in different countries. The characteristics of GTs and the advantages and disadvantages of biological, physical–chemical, and AOP treatments in terms of sustainability were discussed to establish the most efficient methodologies for denim wastewater treatment.

Dye characteristics

Indigo dyes

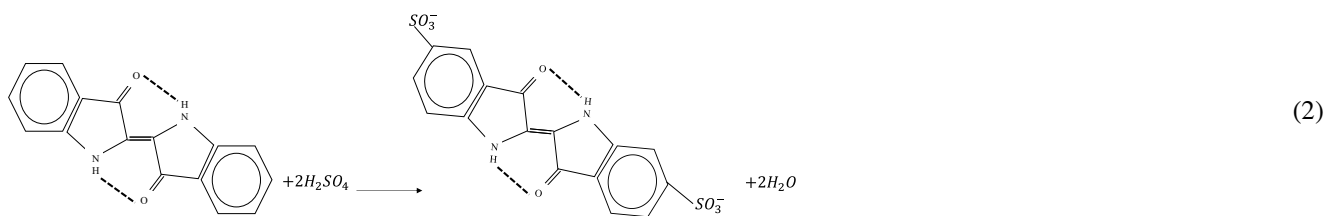
IB dyes are organic compounds of blue color obtained from plants by the biosynthesis of the secondary metabolites of the species *Indigofera tinctoria*, *Indigofera suffruticosa*, *Strobilanthes flaccidifolius*, *Polygonum tinctoriu*, and marine mollusks (*Murex brandaris*, *Murex trunculus*, and *Purpura hematoma*) which are obtained as natural extracts (Mendoza-Avila, Chauhan and Vazquez-Duhalt, 2020). Indigo dyes absorb electromagnetic radiation with wavelengths between 420 and 450 nm, from a blue to violet color. This dye is part of the ketonic group (C=O) (Ramos et al. 2020), is soluble in water, and is more soluble in polar organic solvents (Chavan 2015). Approximately 50,000 tons of dye is produced annually. Unfortunately, the extraction methods are not environmentally friendly (Mendoza-Avila, Chauhan and Vazquez-Duhalt, 2020). Recently studies have reported that dye enzymatic synthesis has a low environmental impact on microbial species such as *E. coli* and *Bacillus megaterium* (Hsu et al. 2018; Mendoza-Avila, Chauhan and Vazquez-Duhalt, 2020). IB dye has a CAS number of 482-89-3, and IUPAC name of 3H-indol-3-yl, 2-(1,3-dihydro-3-oxo-2H-indol-2-ylidene)-1,2-dihydro, and a chemical formula of C₁₆H₁₀N₂O₂ (Chavan 2015).

In the extraction process, indigo dye is obtained in its indigotin form, which is a precursor, and a fermentation step is necessary to obtain indoxyl and subsequently indigo (Eq. 1). Currently, in addition to natural fermentation, there are other methods for producing indigo, such as electrochemical reduction, chemical reduction, electrocatalytic hydrogenation, and catalytic hydrogenation (Uddin and Sayem 2020).



The growing demand for dyes after the industrial revolution generated interest in the artificial synthesis of these compounds. Different routes have been applied to produce them on a large scale using chemicals and environmentally unfriendly substances (Chavan 2015). Further GT research to improve the productivity, purity, and processing of natural indigo must be conducted (Mendoza-Avila, Chauhan and Vazquez-Duhalt, 2020; Nambela, Haule and Mgani, 2020; Uddin and Sayem 2020).

IC is a derivative of indigotin and is also used in the textile, food, cosmetics, and medical industries (Legerská et al. 2016). The chemical structure is observed in Eq. 2, and indigotin is sulfonated by incorporating sulfur into the aromatic ring (Costa et al. 2018). IC dye is also known as acid blue 74, has a CAS number of 860-22-0, an IUPAC name of disodium 5,5'-(2-(1,3-dihydro-3-oxo-2H-indazol-2-ylidene)-1,2-dihydro-3H-indol3-one) disulfonate, and a chemical formula of $C_{16}H_8N_2Na_2O_8S_2$ (No et al. 2021).



Uses in the industry

Indigotin is an anionic dye that is soluble in water and is used as a colorant in foods, cosmetics, clothing, and pharmaceuticals (Harrache et al. 2019). Its maximum permissible limits in food are in the range of 50 and 500 mg/kg. (Fig. 1).

The common use of indigo dyes is for denim dyeing, and approximately 57% of the dye produced is used in the textile industry and other industries, such as the dyeing industry (21%), the paper and pulp industry (10%), the tanning and paint industry (8%), and the dye manufacturing industry (7%) (Chowdhury et al. 2020). Figure 2 shows the uses of indigo dye and the environmental impact of denim laundering.

Dye production is a process in which a wide variety of chemical agents are employed, resulting in the production of environmentally unfriendly constituents (Mendoza-Avila, Chauhan and Vazquez-Duhalt, 2020) such as hydrochloric acid, sodium hydroxide, hypochlorite, potassium dichromate, urea, acetic acid, sulfur dyes, formic acid, and fluorocarbons (Arputharaj et al. 2016). The textile industry produces wastewater with a heterogeneous composition and abundance. These effluents represent a danger to the environment. Approximately 50 to 2600 L of water is utilized to

make 1 kg of fabric (Madhav et al. 2018; Bento et al. 2020). Textile wastewater is the major cause of environmental degradation and human illnesses (Sivaram et al. 2018).

Although the environmental impact of the textile industry is negative, this sector has contributed to the strengthening of the economy. The global textile and apparel market was valued at \$1.9 trillion in 2019, and this value is expected to reach \$3.3 trillion by 2030. The textile industry has contributed to economic recovery after the COVID-19 pandemic (Hajjar 2020). Due to the importance of this sector, it should be a priority for authorities and industry to generate and implement strategies to mitigate its environmental impact.

Physicochemical characteristics of textile wastewater

Textile industries are considered major sources of wastewater that release non-biodegradable compounds into natural

water sources (Naga Babu et al. 2019). Indigo effluents are characterized by a dark blue color because the dye is not fixed to the fiber in the fabric conditioning or washing stage (Ramos et al. 2020). A large portion of the chemicals and dyes do not adhere to the fiber during the dyeing process, and the excess is discharged into the environment as wastewater. Textile materials can absorb approximately 80% of dyes and chemicals (Chowdhury et al. 2020). Figure 3 shows the different stages of the textile production process and the main pollutants that are present in textile wastewater (Madhav et al. 2018; Bento et al. 2020).

In the desizing stage, various chemicals are used and provide mainly biological oxygen demand (BOD) such as starch, which is released from the tissue. Scouring is the stage where other non-cellulosic components (fats, oils, herbicides) of the fabric are removed by alkaline washing using detergents, resulting in wastewater with a high pH. In the mercerizing stage, the fabrics are washed with caustic soda to promote color retention and improve fabric strength, which increases the pH of the wastewater (Periyasamy and Militky 2017). The bleaching stage produces white fabric for the generation of pale and bright shades by applying hypochlorite, peracetic acid, hydrogen peroxide, or enzymes (Arputharaj et al. 2016; Roy Choudhury 2017). In the dyeing stage, different heavy metals can be added to the wastewater, since they are used as auxiliaries in the dyeing process to



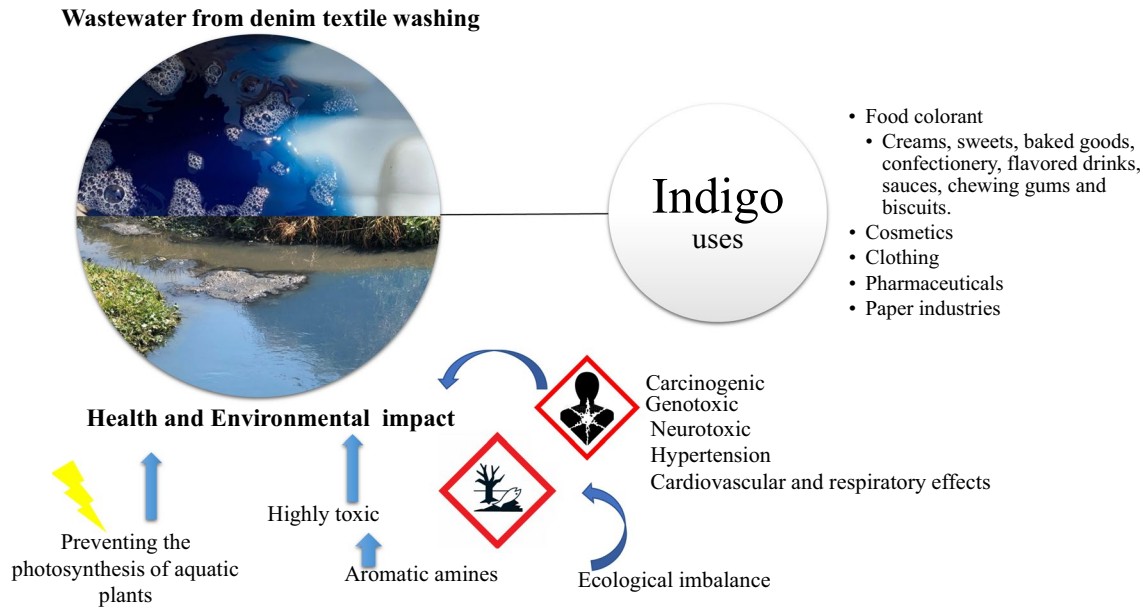


Fig. 1 Indigo dye uses and environmental and health impacts of denim textile wastewater

improve the adsorption of the dye on the fibers by complexation reactions (Periyasamy and Militky 2017; Madhav et al. 2018). This stage is considered to contribute the highest level of contamination since a large part of the dye is not retained by the fabric (20–30%), so color, BOD, and COD are increased considerably. In the printing stage, urea, other dyes, or metals are added. The washing steps include pre-wash bleaching, antichlor wash stone/acid/enzyme wash, and softener, which mainly add enzymes, surfactants, detergents, adsorbable organic halogens, sodium/disodium thiosulfate, acids, and BOD. The finishing stage, aimed at softening the fabrics, involves the use of agents designed for crosslinking, softening, and waterproofing (Madhav et al. 2018).

As natural indigo production is currently deficient, synthetic indigo has been developed, which contains toxic chemicals such as aniline and N-methylaniline residues. Additionally, denim is often dyed with sulfur, reactive pigments, and direct dyes that are non-biodegradable and toxic to aquatic organisms and other organisms. Most of these dyes contain heavy metals, such as chromium, copper, zinc, and manganese. In general, the coloring process has never achieved more than 85% efficiency, so the remaining color remains in the wastewater (Periyasamy and Militky 2017).

Table 1 presents the physicochemical characteristics of denim wastewater compared to the maximum permissible limits of the Environmental Protection Agency of Taiwan, China (Environmental Protection Administration Republic

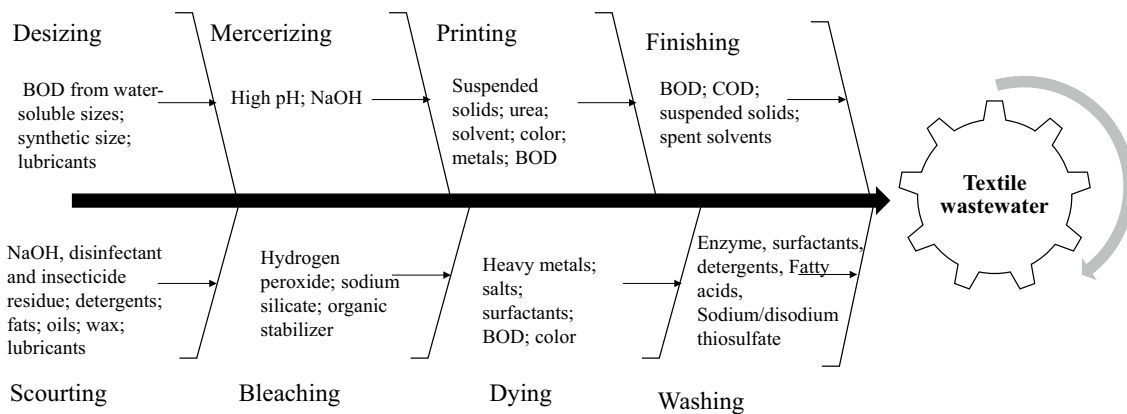


Fig. 2 Textile production process and the main pollutants that are present in textile wastewater

of China 2019). The minimum BOD value was 9.8 mg/L, which generates an effluent with a very low biodegradability index (0.009 BI) (Metcalf & Eddy Inc. 2014). In contrast, other authors obtained a BI of 0.69, and these differences depend on the process stage of textile garments (Tomei et al. 2016).

Table 1 shows the maximum values of 3000 Pt–Co units (Periyasamy and Militky 2017; Madhav et al. 2018; Yogalakshmi et al. 2020). In the bleaching stage, hypochlorite or peroxide are used (Arputharaj et al. 2016; Roy Choudhury 2017), and these oxidants could improve the BI of the dye, generating assimilable by-products. It has been observed that some oxidation treatments increase the biodegradability of dyes (Castañeda Juárez et al. 2020).

Reports of heavy metals in wastewater are limited (Table 1). Due to the characteristics of textile processes (Fig. 2), metals are required for many dyes to become fixed in fabric by complexation reactions (Madhav et al. 2018). Chromium, copper, zinc, and manganese are commonly associated with textile processes (Periyasamy and Militky 2017; Madhav et al. 2018; Velusamy et al. 2021). Therefore, their quantification in textile wastewater should be a priority; however, the studies shown in Table 1 do not determine most of the related parameters (N.D.); this demonstrates an inconsistency and a breach of the international guidelines.

Permissible limits of contaminants in textile wastewater

Table 2 shows the maximum permissible limits established by different countries for textile wastewater discharge. BOD₅ is generally regulated within a range of 20–150 mg/L. COD is generally regulated between 8 and 250 mg/L. The pH is regulated within a range of between 6 and 9 units. Additionally, the presence of heavy metals is possible due to the staining process of fabrics (Periyasamy and Militky 2017; Madhav et al. 2018). The color is regulated within 50 to 550 Pt–Co units. This parameter is not regulated in some countries, which indicated an inconsistency in legislation.

China's textile industry is the largest in the world in terms of global production and exports, in addition to being the largest exporter, with a share of 39% (Agarwal et al. 2017). In countries where the textile industry is an important economic activity, there are specific regulations for the resulting wastewater (China, Republic Taiwan, Bangladesh) (MEP 2012; Environmental Protection Administration Republic of China 2019).

When comparing the physicochemical characteristics of raw wastewater from the textile industry (Table 1), the results reveal that most of the parameters are not within the permissible limits (Table 2). The permissible limits of these parameters are dissimilar among different legislations, and

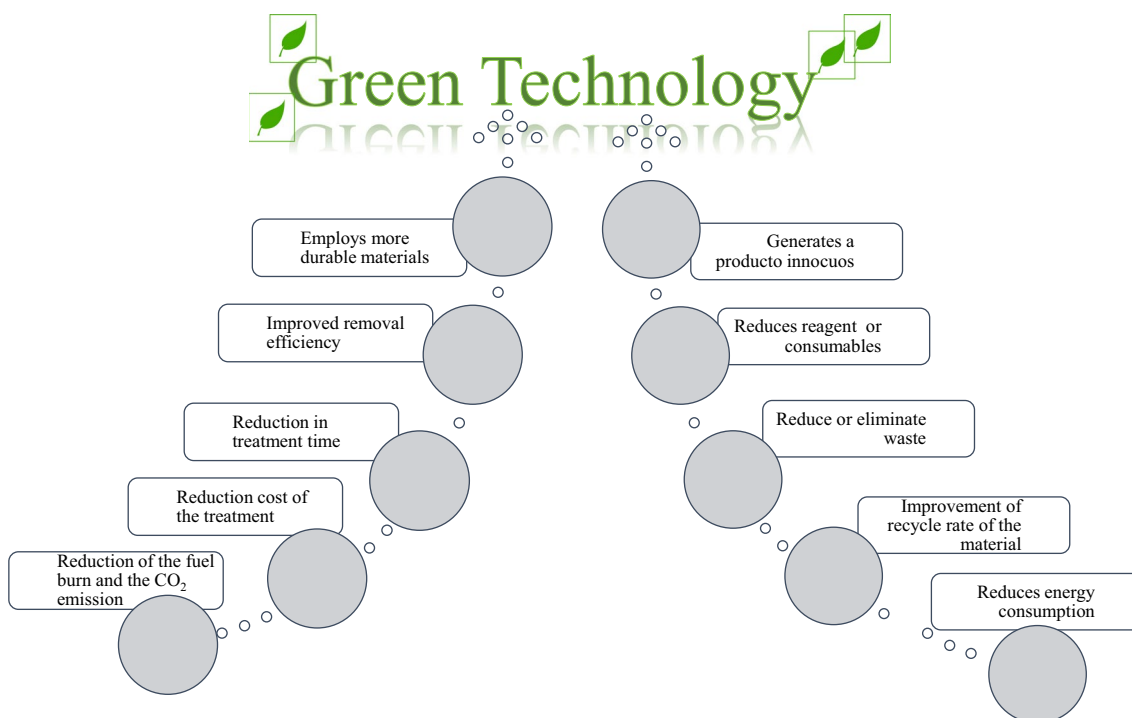


Fig. 3 GT desirable characteristics for wastewater treatment



Table 1 Physicochemical characteristics of raw wastewater from the textile industry

	References														
	Taiwan. Effluent Standards Dated	(Ramos et al. 2020)	(Hen-daoui et al. 2018)	(Belkacem et al. 2008)	(Almazán-Sánchez et al. 2016)	(Yaseen and Scholz 2019)	(Tomei et al. 2016)	(Thangaraj et al. 2017)	(Bhatia et al. 2018)	(Sánchez-Sánchez et al. 2018)	(Castañeda Juárez et al. 2020)	(Ćurić, Dolar and Karadakić, 2021)	(Xue et al. 2019)	(Yurt-sever, Sahinkaya and Cinar, 2020)	(Mubashar et al. 2020)
Country	Brazil	Tunisia	Algeria	Mexico	India	Italy	India	India	India	Mexico	Mexico	Croatia	Chinese	Turkey	Pakistan
pH	6–9	12.2	8.7	6.8	8.6–9.2	9.0	9.3	7.4–8.6	7.4–8.6	5.2	6.4	7.28	7.1	8–9.5	ND
BOD (mg/L)	ND	ND	310	91.9	1206–1750	9.8	532	170–361	1400	1400	151.9	395	ND	ND	ND
COD (mg/L)	ND	2080	620	344	3880–4400	1017	3874	317.6–8106	2022	734.1	734.1	1338	112	700–1250	755–871
BI	NA	ND	0.5	0.26	0.32–0.39	0.009	0.13	0.53–0.04	0.6923	0.2069	0.2069	0.2952	ND	ND	ND
Color (Pt-Co)	1180–1576	1750	> 200	330	ND	0.66*	ND	ND	3000	590	590	ND	ND	500–1250	ND
TSS (mg/L)	30	ND	ND	ND	550–650	ND	826	86–143.5	ND	405	405	93	ND	ND	ND
Turbidity (NTU)	NA	ND	765.5	104.6	12.5–16.6	ND	ND	ND	735	178	178	90	ND	ND	ND
Conductivity (µS/cm)	NA	12,372	3.1X10 ⁶	471.3	ND	ND	5.6X10 ⁵	ND	2.8X10 ³	ND	ND	ND	ND	4–5.5 X10 ³	ND
Fats and oils	10.0	ND	ND	ND	ND	ND	ND	ND	ND	26.8	26.8	ND	ND	50–200	ND
Phenols (mg/L)	1.0	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Sulfides (mg/L)	1.0	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Cyanides (mg/L)	1.0	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Cu (mg/L)	3.0	ND	ND	ND	ND	ND	3.1	ND	ND	ND	ND	ND	ND	ND	1.33–1.88
Cr total (mg/L)	2	ND	ND	ND	ND	ND	2.3	ND	ND	ND	ND	ND	ND	ND	1.32–2.02



the dye concentration is not regulated. Textile wastewater can have a dye concentration of 64.6 mg/L (De León-Condés et al. 2019), perhaps due to the diversity of dyes and reagents that are used in industry (Chavan 2015; Gallego-Schmid and Tarpani 2019; Paździor, Bilińska and Ledakowicz, 2019; Singh et al. 2019).

The objective of the regulations should be focused on promoting sustainability in the industry, applying the best available control technology, applying environmentally friendly processes to protect public health, and reducing the pollutant indicators to comply with final discharge standards.

Health and environmental implications of indigo dye

Indigo is a highly persistent recalcitrant dye, and its removal from wastewater is problematic because it is chemically and photolytically stable with a high solubility (10 g/L). The release of indigo dye into the environment can cause corneal and conjunctival damage, skin irritation, dermatitis, and cancer (Khadhri et al. 2019). It is employed for staining/dyeing in the textile industry and other industries, such as the cosmetics, food, leather, printing, and paper industries (Naga Babu et al. 2019). According to the legislation of the European Union and the USA, indigo dye is classified as a carcinogenic, genotoxic, and neurotoxic substance, and it can cause hypertension and cardiovascular and respiratory effects, as shown in Fig. 1 (Kekes and Tzia 2020). Furthermore, it irritates the respiratory and gastrointestinal tract leading to nausea, diarrhea, and vomiting and is therefore regarded as a highly toxic pollutant (Naga Babu et al. 2019).

Ingesting dyes can cause extreme sweating, confusion, methemoglobinemia and mouth burns. Water toxicity in mice is > 32,000 mg/kg body weight. Dyes may cause organ damage after prolonged or repeated exposure and are not readily biodegradable. Acute water toxicity in *Leuciscus idus* is > 10,000 mg/L (LC₅₀), and the average toxicity is > 500 mg/L (EC₅₀) in *Daphnia magna* (ROTH 2019).

Dye effluents mixed with natural source water produce an unpleasant odor due to low light penetration, and effluent turbidity tends to form a visible layer on the water surface due to lower density (0.8 kg/m³) of the effluent compared to the density of water. This blocks the penetration of sunlight needed by underwater living organisms, affecting photosynthesis and respiration processes (Oliveira et al. 2019).

Dyes are often transformed into toxic aromatic amines that absorb and reflect sunlight, affecting the photosynthesis of aquatic plants (Almazán-Sánchez et al. 2016). In water

bodies, dyes can generate problems with the absorption of light necessary for biomass, and their accumulation in lakes can cause ecological imbalances (Shinde, Patil, G.-D. Kim, et al. 2020). Eventually, the contamination of rivers with dye will reach the oceans, altering these ecosystems as well (Singh et al. 2019).

Green technologies and life cycle assessment for wastewater treatment

GTs are processes that allow us to obtain and use natural resources without negative effects on ecosystems, facilitating a balance between anthropogenic activities and nature (Guo et al. 2020; Schuelke-Leech 2021). GTs can also be defined as processes that allow the mitigation of the negative consequences of conventional production procedures, according to the application of guidelines to preserve the environment. Green finance has been demonstrated to significantly improve the relationship between economic development and the environment (Zhou et al. 2020).

The development and application of GT have increased in recent years, particularly in the areas of wastewater treatment (Wang et al. 2019). There is a diverse conceptualization of the characteristics that a GT should have this conceptualization is still focused on economic guidelines, ignoring social and environmental factors that contrast with and contradict its definition (Guo et al. 2020).

The textile industry plays a critical role in the world economy (Koszevska 2018); however, it relies upon activities that are still not sustainable. Worldwide, it has a serious impact on the environment due to the consumption of enormous amounts of resources, and it generates 5% of the world's waste (Fischer and Pascucci 2017). This industry discharges large quantities of wastewater with various pollutants (Almazán-Sánchez et al. 2016; Thangaraj et al. 2017; Castañeda Juárez et al. 2020); therefore, it is important to develop of GTs focused on reducing or eliminating the negative environmental impact of industry on water resources. Fig. 3 presents the desirable characteristics that a GT should have to preclude negative effects on wastewater treatment.

A GT for denim wastewater treatment should preferably employ more durable materials, which can reduce consumption and mitigate environmental effects in the manufacturing process. Additionally, it should improve pollutant removal according to international regulations through synergistic effects that enhance efficiency or minimize operating times or reagent consumption.

This GT favor the decrease of fuel consumption and CO₂ emissions (Shao et al. 2021) the reduction in production time and costs is more related to the reduction of consumerism and not to an economic guideline (Fig. 3). Moreover, a GT should not be excluded because of the costs associated with its application. Consumerism has affected the natural environment, increasing the demand and overexploitation of resources and, ignoring negative effects on the environment (Ambec, Stefan; De Donder, 2018).

Some technologies commonly used in wastewater treatment may generate wastes, such as sludge (Wang et al. 2017). GTs seek to reduce or eliminate the generation of these wastes, decreasing the use of inputs that could also damage the natural environment. A GT is characterized by proposing new uses and applications for these wastes, improving their qualities, and reducing their generation or toxicities (Romero-Pareja et al. 2017; Wang et al. 2017), generating harmless (innocuous) products.

Reducing or eliminating energy consumption is a very clear goal in the development of a GT to minimize the demand, energy production, and environmental impacts associated with electricity generation related to climate change (Cao et al. 2016). Therefore, a GT should promote the use of renewable energies, such as solar, biofuel, hydroelectric, or wind energy, in treatment plant operation (Qazi et al. 2019). The recovery of materials is related to the use of inputs or parts that are taken as waste to be reintegrated into the process, and reduces the impacts associated with the emission of greenhouse gases (Fortuna and Diyamandoglu 2017).

One tool for substantiating the impacts of a wastewater treatment process is life cycle analysis (LCA), which considers raw materials, production, transport, consumption, and disposal (Gallego-Schmid and Tarpani 2019; Muthu 2020). This tool makes it possible to determine the inputs and outputs of a GT to reduce its negative effects on the natural environment (Fig. 2). Therefore, GTs should be evaluated within the framework of an LCA (Moni et al. 2020); however, a limited number of studies employ LCAs as an integral part of their research.

On the other hand, laboratory-scale processes, reported in the majority of studies, do not have the same complexity, as industrial-scale processes, and the equipment used is not equivalent to reactors and machinery, so laboratory-scale data may represent overestimated environmental impacts (Moni et al. 2020). The application of an LCA on a laboratory-scale process may not offer real estimated values that

can be extrapolated to the industrial-scale. The application of LCAs by the scientific community in the treatment of textile industry wastewater is limited (Gallego-Schmid and Tarpani 2019), and these few studies lack support and evidence about the GT studied.

Treatments for Indigo removal and denim wastewater

Biological treatment

Biological treatments consist of the degradation of dyes or contaminants by metabolic or adsorption routes, such as bacteria, yeasts, fungi, algae, plants, enzymes, or any material derived from a raw material of natural origin (Paz et al. 2017; Shoukat, Khan and Jamal, 2019). The treatment, operating conditions, and removal percentage by biological treatments of IB dye in synthetic and real wastewater are presented in Table 3.

In general, these treatments are more economical, and environmentally friendly (Paz et al. 2017; Bento et al. 2020), probably due to the low consumption of energy and reagents. Their disadvantages are the sensibility of microorganisms to low biodegradability, low tolerance to extreme pH changes, persistent pollutants, lower adaptability of the processes, larger surfaces for the installation of equipment, and longer residence time (54 h) for dye removal (Gürses, Güneş and Şahin, 2021) (Table 3).

The use of plants and microorganisms in the removal of dyes from textile wastewater is a more attractive and economical option than physical–chemical methods (Singh et al. 2019). However, dye degradability depends on the characteristics of the dye and the wastewater (Shoukat, Khan and Jamal, 2019); thus, these systems must be adapted and conditioned to the particular characteristics of the wastewater to be treated.

Most of the treatments in Table 3 are from recent years and show color removal efficiencies in synthetic wastewater of up to 73–100% (Bankole et al. 2017; Paz et al. 2017), even though it has been reported that the dyes are not biodegradable (Chavan 2015). On the other hand, for real wastewater, the efficiencies achieved are in the range of 60–78% (Shoukat, Khan and Jamal, 2019; Hevira et al. 2020). This is due to the diversity of contaminants that textile effluent contains, such as detergents, hypochlorite,

Table 3 Indigo removal by biological treatments in synthetic and textile wastewater

Country	Technology	Operating conditions	Result	References
Portugal	Enzymatic degradation	Wastewater: synthetic Enzyme activity: 1000 U/L pH: 4.5 Temperature: 25 °C Dye C ₀ : 25 mg/L Time: 24 h Ionic liquid (IL): 75 mM of [N ₁₀₁₁₁]Br and [C ₁₀ mim]Cl	Color removal: 93%	(Bento et al. 2020)
Brazil	Bioelectrochemical remediation (immobilization of enzyme at the anodic surface)	Wastewater: synthetic Cathode: commercial platinum Anode: activated carbon fibers felt The distance anode–cathode: 1 cm Volt: 10 V I: ~ 10 mA j: 33.75 mA/cm ² Volume: 10 mL Temperature: 28 °C Electrolyte support: NaCl at 50 mM Time: 60 min Dye C ₀ : 0.001% w/v (38 μM) pH: 7.46 Catalytic activity: 280 U/mL	Color removal: 83.6%	(Garcia et al. 2017)
Nigeria	Bioremediation	Wastewater: synthetic Dye C ₀ : 10 mg/L pH: 2 Temperature: 30 °C Cell biomass: 2 g Time: 5 days Yeast: <i>Diutina rugosa</i>	Color removal: 99.9%	(Bankole et al. 2017)
Spain	Bioremediation	Wastewater: synthetic Bacteria: <i>Bacillus aryabhatai</i> DC100 Dye C ₀ : 180 mg/L Volume: 50 mL Inoculum: 1 mL pH: 8.8 Temperature: 37 °C Agitation: 304.1 rpm Salt concentration: 19.2 g/L Media culture: Nutrient Broth (NB), Luria–Bertani Broth (LB) and Trypticasein	Color removal: 100%	(Paz et al. 2017)
Pakistan	Anaerobic-SBR/ aerobic-SBR treatment	Wastewater: industry Total HRT: 54 h HRT in Anaerobic: 48 h HRT in Aerobic: 6 h SRT anaerobic: infinite SRT aerobic: 20 h pH: 6.8–7.2 Operational volume: 6 L -COD ₀ : 2200–2800 mg/L Total nitrogen: 250–300 mg/L	Color removal: 78.4% COD removal: 99.5% TKN removal: 99.3%	(Shoukat, Khan and Jamal, 2019)

Table 3 (continued)

Country	Technology	Operating conditions	Result	References
Mexico	Electrooxidation– <i>Salix baby-lonica</i>	Wastewater: textile industry Electrodes: boron-doped diamond pH: 5.23 Time: 300 min j: 3.5 mA/cm ² COD ₀ : 2022 mg/L BOD ₀ : 1400 mg/L Color initial: 3000 U Pt–Co Turbidity initial: 735 NTU Photoperiod: 12 h light, 12 h dark Time contact with plants: 15 days Temperature: 19–22 °C	Color removal: 60.8% COD removal: 42.9% BOD removal: 83.3% Turbidity: 26.5%	(Sánchez-Sánchez et al. 2018)
Turkey	Decolorization by <i>Pseudomonas aeruginosa</i> and crude laccase enzyme	Wastewater: synthetic <i>Pseudomonas aeruginosa</i> Temperature: 30 °C Stirring speed: 150 rpm Time: 24 h Volume: 50 mL Inoculum: 1 mL Media culture: 20 mL Luria broth (LB) Enzyme Dose: 54 U/mL pH: 3.0 Time: 300 s Temperature: 30 °C Dye C ₀ : 100 mg/L	Color removal by <i>Pseudomonas aeruginosa</i> : Dye C ₀ 50 mg/L: 92% Dye C ₀ 100 mg/L: 91% Color removal by laccase: 57.2%	(Boran et al. 2019)
Mexico	Bacterial and fungal strains	Wastewater: synthetic Dye C ₀ : 50 mg/L <i>Bacillus</i> BT5: 100 rpm, pH: 7, 37 °C, 384 h <i>Bacillus</i> BT9: 100 rpm, pH: 7, 37 °C, 216 h <i>Lysinibacillus</i> BT32: 100 rpm, pH: 7, 37 °C, 216 h <i>Lactobacillus</i> BT20: 100 rpm, pH: 7, 37 °C, 264 h <i>Aspergillus</i> HIT: 100 rpm; pH 7.0; 28 °C; 48 h	<i>Bacillus</i> BT5 color removal: 36% <i>Bacillus</i> BT9 color removal: 59% <i>Lysinibacillus</i> BT32 color removal: 29% <i>Lactobacillus</i> BT20 color removal: 92% <i>Aspergillus</i> HIT color removal: 96%	(Valdez-Vazquez et al. 2020)
China	Recombinant laccase enzyme by <i>Bacillus subtilis</i> cjp3	Wastewater: synthetic pH: 9.0 Time: 4 h Temperature: 20–80 °C Expression host: <i>Escherichia Coli</i> Dye C ₀ : 25 mg/L Shaking conditions: 150 rpm	Maximum activity: 7320 U/L Color removal: 86%	(Qiao et al. 2017)
Poland	Laccase-like multicopper oxidases by <i>Myrothecium roridum</i>	Wastewater: synthetic Time: 3 h Dye C ₀ : 200 mg/L	Activity: 1 U/L Color removal: 95%	(Jasińska et al. 2018)



Table 3 (continued)

Country	Technology	Operating conditions	Result	References
Brazil	Microfiltration–membrane bioreactor	Wastewater: industry Dye C_0 : 0.74 g/L Conductivity initial: 3275 $\mu\text{S}/\text{cm}$ COD initial: 3050 mg/L Ammonia initial: 10.2 mg/L Pore diameter: 0.4 μm Filtration area: 1 m^2 N° Module: 1 Pressure: 1 bar Flow rate: 2.4 L/min Volume biological tank: 6 L A pore size of 0.5 μm , Support media: membrane area of 0.044 m^2 , packing density of 500 m^2/m^3 HRT: 8 h	Microfiltration: Color removal: 100% COD removal: 65% Conductivity: 25% Bioreactor: COD removal: 73% Ammonia removal: 100%	(Couto et al. 2018)

[N₁₀₁₁₁]Br: decyltrimethylammonium bromide

[C_{10mim}]Cl: 1-decyl-3-methylimidazolium chloride

j Current density, I Current intensity, V Voltage; TKN Total Kjeldahl Nitrogen, HRT Hydraulic Retention Time, SRT Solid Retention Time,

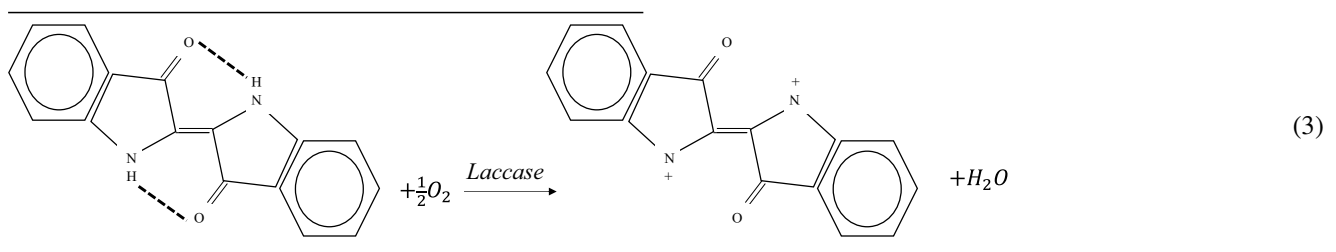
hydrogen peroxide, acidifying agents, disinfectants, insecticides, and heavy metals that inhibit metabolic pathways (Chavan 2015; Gallego-Schmid and Tarpani 2019; Paździor, Bilińska and Ledakowicz, 2019; Singh et al. 2019).

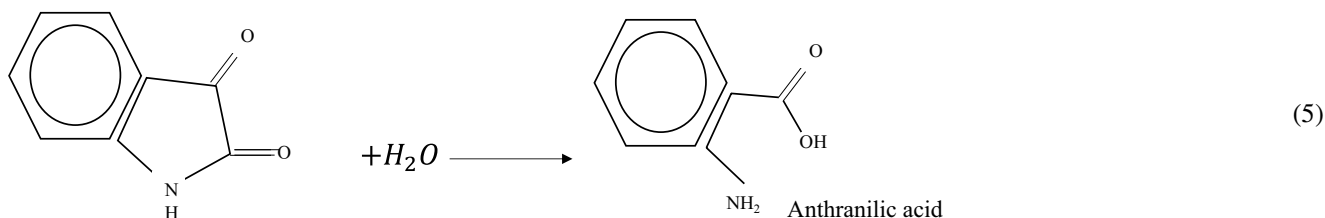
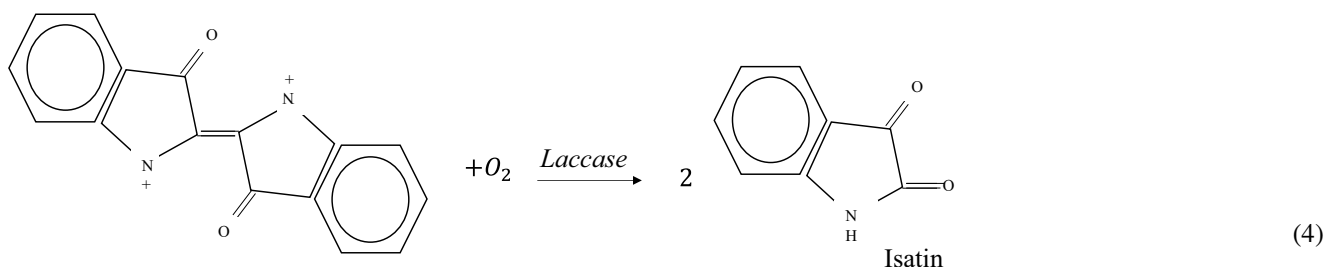
Bento et al. (2020) explained that real wastewater contains enzyme inhibitors (chlorine and pH), which reduce enzyme activity; for this reason, enzymatic processes should be evaluated under real conditions. An efficient application of biological treatments cannot always be successful in the treatment of denim wastewater due to the labile nature of the enzymes, the inhibition of microorganisms, and the competition between the different pollutants (Bento et al. 2020; Hevira et al. 2020).

The synergistic effects of a combination of several technologies can improve dye removal. Recently, a bioelectrochemical treatment with enzymes immobilized on the anode was evaluated (Table 3) (Garcia et al. 2017). The electrooxidation-*Salix babylonica* combination achieved a color removal efficiency of 60.8%. However, COD removal

was low (42.9%), and the plants obtained more biomass and photosynthetic pigments (Sánchez-Sánchez et al. 2018). These combined effects improve efficiencies, so a single technology may not be the solution, and the combination or coupling of treatments results in improved efficiency and reduced costs.

Biological treatments degrade dyes due to intra- and extracellular enzymes (Quintero and Cardona 2010). Boran et al. (2019) evaluated dye degradation using the enzyme laccase, and a removal of 57.2% in synthetic water (Table 3). The oxidation by this enzyme under aerobic conditions is described in Eq. 3–5. In the first stage, the separation of H-bridge bonds occurs, followed by the incorporation of oxygen, which causes the separation of aromatic rings through biohydroxylation, forming carboxylic acids used in metabolism (Quintero and Cardona 2010; Legerská et al. 2016). This enzyme only requires molecular oxygen as a co-substrate, and one of its important functions is to increase the susceptibility of dyes to spontaneous nucleophilic attack (e.g., H_2O) with the addition of oxygen atoms (Quintero and Cardona 2010).



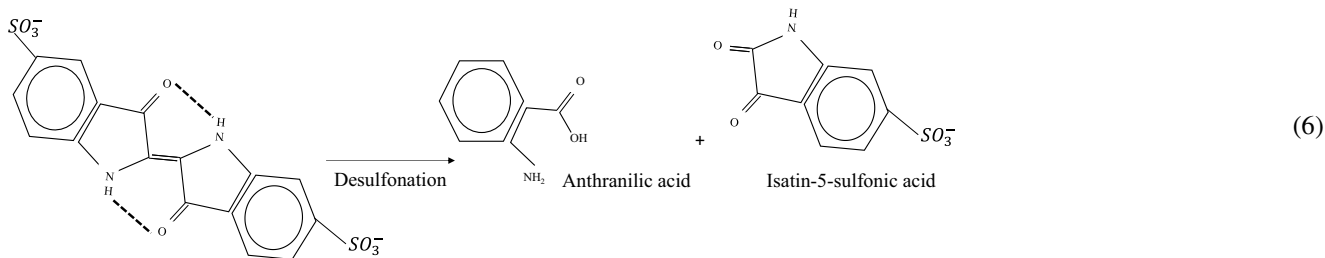


Laccase synthesis is possible for some species of fungi and bacteria, such as *Scytalidium thermophilum*, *T. hirsuta*, *Escherichia coli*, *Bacillus megaterium*, and *Sclerotium rolfsii*, and it has been observed that fungal enzymes are more efficient in the removal of IC (Eq. 6) dye, although the final products in both cases are anthranilic acid; this difference is due to the higher redox potential of fungal enzymes compared to that of bacterial enzymes (Legerská et al. 2016). *Aspergillus HIT* showed a color removal of 96% in 48 h, which was significantly faster than *Bacillus*, *Lysinibacillus*, and *Lactobacillus* (Table 3) (Valdez-Vazquez et al. 2020).

Physical-chemistry treatment

Adsorption

This method consists of matter transfer from the liquid phase of denim wastewater to a solid phase or surface. Materials such as activated carbon, zeolites, sweepers, activated alumina, lignite coke, and bentonite are usually used (Cossu et al. 2018). Adsorption is an attractive method due to the low costs associated with its operation, its ease of operation, its effectiveness over a wide



Under anaerobic conditions, dye degradation can occur; however, azo dyes generally produce aromatic amines that are more toxic to the environment, and some of them are easily metabolized under aerobic conditions, so combined anaerobic–aerobic treatments are more recommended (Gürses, Güneş and Şahin, 2021).

pH range, and its high performance in color removal. The adsorption mechanism usually occurs at temperatures between 25 and 40 °C (Table 4), and it is possible to recover the adsorbent material for reuse after a certain number of cycles (Kekes and Tzia 2020). Organic wastes are commonly used to obtain adsorbent materials.



Table 4 Operating conditions of adsorption technologies for indigo dye removal

Country	Adsorbent	Optimal operation conditions	Result	References
Tunisia	Activated carbon	Wastewater: synthetic Langmuir model is the most appropriate Flow rate of 0.5 mL/min Dye C_0 : 100 mg/L Temperature: 15 to 40 °C pH: 7.8	Color removal: 76.5% Reusability over three cycles Adsorption capacity: 50.1 IC mg/g adsorbent	(Khadhri et al. 2019)
Greece	Chitosan and Chitosan/ β -Cyclodextrin beads	Wastewater: synthetic Fitted to Langmuir and Freundlich models pH: 4 Volume operation: 200 ml Temperature: 25 °C	Color removal: 98.7% Adsorption capacity: 500–1000 mg IC/g adsorbent	(Kekes and Tzia 2020)
Mexico	Iron-Modified clay	Wastewater: textile industry Elovich model and isotherms data to Freundlich model Time: 72 h pH: 6.8 Volume operation batch: 0.015 L Modification of the material Current intensity: 0.5 A Time: 130 min pH: 7 Electrodes: Fe-Cu	Adsorption color capacity: 43.9 U Pt Co/g Adsorption TOC capacity: 2.6 mg/g Adsorption COD capacity: 9.2 mg/g COD removal: 54.9% TOC removal: 46.9% Color removal: 65.2% Reusability over four cycles	(Almazán-Sánchez et al. 2016)
India	Red mud, hazardous aluminum industry waste	Wastewater: synthetic Volume operation batch: 100 mL Mixture: 300 rpm Dye C_0 : 100 mg/L Temperature: 30 °C Time: 100 min pH: 3–4	Color removal: 94% Reusability over five cycles Adsorption capacity: 62.6 IC mg/g	(Naga Babu et al. 2019)
India	Calcium hydroxide	Wastewater: synthetic Fitted to Langmuir model Time: 50 min pH: 12 Dose adsorbent: 0.1 g/ 100 mL Dye C_0 : 5 mg/ 100 mL Temperature: 330 K	Color removal: 97.3%	(Ramesh et al. 2017)
USA	Hemoglobin/Iron oxide	Wastewater: synthetic Langmuir isotherm models Pseudo-first and second-order model pH: 4–5 Dye C_0 : 50 mg/L Time: 6 h	Reusability over four cycles Adsorption capacity: 104.2 mg/g	(Essandoh and Garcia 2018)
Algeria	Activated carbon	Wastewater: synthetic pH: 5.2 Webber-Morris diffusion model Dye C_0 : 60 ppm Stirring speed: 400 rpm	Adsorption capacity: 17.4 mg/g	(Harrache et al. 2019)

Table 4 (continued)

Country	Adsorbent	Optimal operation conditions	Result	References
India	Nanoporous magnesium aluminophosphate	Wastewater: synthetic Time: 20 min Dye C_0 : 100–500 mg/L Temperature: 30 °C Dose adsorbent: 0.5 g/100 mg/L of IC Dose adsorbent: 2.5 g/500 mg/L of IC	Color removal at $C_0 = 100$ mg/L: 75–80% Color removal at $C_0 = 500$ mg/L: 30–35%	(Sivakami et al. 2018)
Indonesia	Biosorption	Wastewater: synthetic and industry Biosorbent: <i>Terminalia Catappa shell</i> pH: 2 Time: 30 min Dye C_0 : 600 mg/L Temperature: 25 °C	Adsorption capacity: 26.7 mg/g Reusability over tree cycles Color removal in synthetic wastewater: 64–73% Color removal in industry wastewater: 60.1%	(Hevira et al. 2020)
France	Calcined and Uncalcined ZnAl-r Anionic	Wastewater: synthetic Dye C_0 : 80–250 mg/L Time: 3 h pH: 4–10 Zn/Al molar ratios of 3 and 4	Adsorption capacity: CZnAl-4 = 520.8 mg/g, CZnAl-3 = 358.4 mg/g, ZnAl-3 = 67.25 mg/g, ZnAl-4 = 21.65 mg/g	(Bessaha et al. 2017)
Italy	Chitosan hydrogels	Wastewater: synthetic pH: neutral Stirring speed: 150 rpm Dye C_0 : 20–1000 mg/L	Adsorption capacity: 118 mg/g Reusability tree cycles	(Salzano de Luna et al. 2017)
Mexico	Activated carbon and polyelectrolyte poly	Wastewater: synthetic Dye C_0 : 300 and 1500 mg/dm ³ Ratio masa adsorbent/volume: 5 mg/dm ³ Wastewater: Industry Color initial: 1500 U Pt–Co COD initial: ≈1400 mg/dm ³	Adsorption capacity: 222 and 416 mg/g Color final: ≈400 U Pt–Co COD final: ≈700 mg/dm ³	(Herrera-González et al. 2019)
Brazil	Modified sugarcane bagasse	Wastewater: industry Particle size: 0.7 mm Dosage: 0.6 g Dye C_0 : 149 U Pt–Co Volume: 50 mL Stirring speed: 150 rpm pH: 2–3 Time: 24 h	Color removal: 100%	(Leon et al. 2020)

Some authors have denoted these materials as biosorbents (Hevira et al. 2020), although the mechanism for the removal of contaminants is the same in all cases.

Table 4 shows recent studies using activated carbon derived from waste organic matter, modified materials to improve adsorption, and nanomaterials. Usually, a dye is used as a model to evaluate the adsorption capacity of a material, and color removal efficiencies between 64 and 98% in synthetic water have been reported (Table 4). In denim wastewater, the efficiencies are lower (65–73%) because of a great variety of contaminants that compete with the active sites of the adsorbent. Metal ions bind more easily to these sites, reducing the dye removal efficiency (Hevira et al. 2020).

Chitosan is a non-toxic and biodegradable material, that is very attractive for dye removal, with an adsorption capacity of 500–1000 mg IC/g adsorbent (Table 4) and a removal efficiency of 98.7%. This biosorbent is synthesized by crustaceans or microorganisms. Its adsorption process is exothermic, spontaneous, and favorable at low temperatures (Kekes and Tzia 2020). However, its removal processes in denim wastewater and other contaminants, such as heavy metals, must be evaluated.

The color removal mechanism is described in Fig. 4. Depending on the adsorbent, hydrophobic-hydrophobic interactions between the hydrogen bonds and the dyes or the adsorbent surface are possible. The amine, imine, and hydroxyl groups on the adsorbent form hydrogen bonds

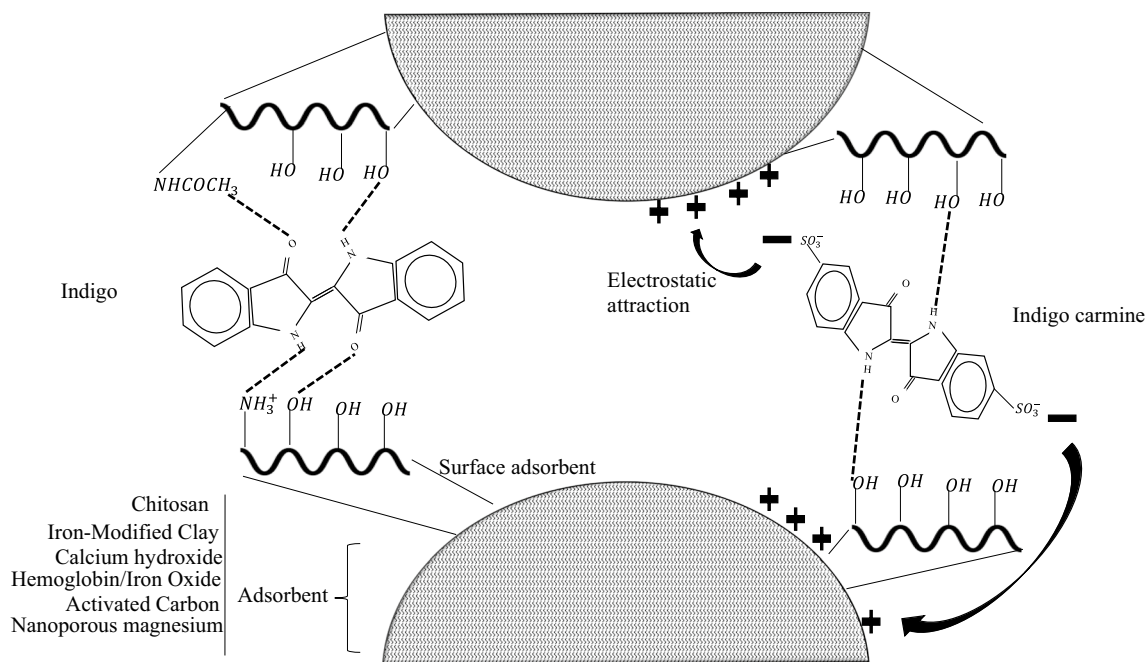


Fig. 4 Mechanisms of adsorption of indigo dyes

with the azo, hydroxyl, and amine groups of the dyes (Li et al. 2012; Salzano de Luna et al. 2017; Chowdhury et al. 2020). Additionally, for the IC dye, $-SO_3^-$ groups are electrostatically negative. Depending on the adsorbent and the point of zero charge the electrostatic interactions between the $-SO_3^-$ group of the dye and the surface of the adsorbent could retain the dye (Hevira et al. 2020).

Filtration

Filtration is a physical process characterized by the use of a membrane or filter media at different pore size ranges (Cossu et al. 2018). The wastewater passes through the filter media and sometimes it is necessary to operate at high pressures, which implies high electrical energy consumption. Different filtration processes, such as microfiltration, ultrafiltration, nanofiltration, and reverse osmosis, have been evaluated in the recovery of wastewater from the textile industry (Balcik-Canbolat et al. 2017). The color removal efficiencies can range 94 to 100% (Table 5), and these processes are highly effective and environmentally friendly (Han et al. 2017).

The separation mechanism occurs due to two factors: (1) The membrane is negatively charged on the surface and repels dye (also negatively charged) through electrostatic repulsion, and (2) the dye molecules form aggregates through hydrogen bonds that can be effectively rejected via size exclusion (Fig. 5) (Han et al. 2017; Thong et al. 2018).

The number of filter media layers significantly influences the removal efficiency; 10 layers of nylon nanofibers were necessary for the 100% removal efficiency of indigo dye and increased with the number of filter layers (Table 5) (Yu et al. 2018). A trend in the development of these technologies is related to the synthesis of new filter materials such as nylon, polyethersulfone, and polyamide, and the evaluation of their dye retention capacities (Han et al. 2017; Thong et al. 2018; Yu et al. 2018). The filtration process is simple to operate, but the consumption of electrical energy and membranes may raise questions when discussing the LCAs of these products.

Coagulation and electrocoagulation

In the coagulation process, colloidal particles are destabilized to enhance their agglomeration into larger particles and their removal by gravity (precipitation) or flotation (flocculation). Destabilization is achieved by the addition of chemical reagents (coagulants) to reduce repulsive forces by neutralizing electrical forces. The most common coagulant agents are Al and Fe salts, characterized by multivalent ions with opposing charges (Cossu et al. 2018).

Electrocoagulation is the generation of chemical agents in situ released from anode oxidation, and Fe, Mg, and Al are commonly used (Table 6), generating hydroxide ions and



Table 5 Filtration technologies for wastewater treatment in the textile industry

Country	Technology	Operating conditions	Result	References
Singapore	Nanofiltration	Wastewater: synthetic Filter: polyethersulfone Molecular weight: 1500 Da Pore diameter \approx 1.96 nm Dye C_0 : 50 g/10 L Time: 72 h pH: 8.69 Permeability media: 13.2 LMH/ bar	Color removal: 94.9%	(Thong et al. 2018)
China	Multi-layer nylon-6 (PA-6) nanofibrous membranes by electrostatic spinning	Wastewater: synthetic Pressure: 15 MPa 10 layer PA-6 nanofiber Specific surface area: 5.68 m ² /g Average pore size: 12.15 nm	Color removal: 100%	(Yu et al. 2018)
Turkey	Nanofiltration (NF) and reverse osmosis (RO)	Wastewater: industry Pretreatment: biological SBR NF and RO effective filtration area: 2.6 m ² Initial fluxes in NF: 85 L/m ² h Initial fluxes in RO: 40 L/m ² h Membrane: NF270 and BW30 Polyamide, pH = 2–11, Max. operating pressure = 41 COD initial: 516 mg/L Conductivity initial: 6720 μ S/cm TDS initial: 3580 mg/L Sulfate initial: 1910 mg/L Chloride initial: 698 mg/L Hardness initial: 103 mg/L CaCO ₃	COD final: < 5 mg/L Conductivity final: 23 μ S/cm TDS final: 11 mg/L Sulfate final: N.D Chloride final: 4.9 mg/L Hardness final: 5 mg/L CaCO ₃ Color removal: 100%	(Balcik-Canbolat et al. 2017)

*MPa: Megapascal, LMH/bar: The flux unit volume (L)/ area of the membrane (m²) time (h)/ pressure (bar)

N.D.: Not detected

hydrogen gas (Hendaoui et al. 2018). However, it has been reported that Fe electrodes produce coagulants that are more environmentally friendly than Al or Zn, which can be harmful at high concentrations (Donneys-Victoria et al. 2020). The coagulant destabilizes and agglomerates pollutants, forming larger particles which, depending on their density, may precipitate or flocculate (Fig. 6).

Table 6 shows the operating conditions and removal efficiencies of denim wastewater treatment. The effect of pH on electrocoagulation is important due to the generation of hydroxide ions, which increases the pH value up to 47% more than the initial pH value (Hendaoui et al. 2021). This parameter significantly influences in the speciation of hydroxides and the Z potential of colloids (Naje et al. 2017). Thus, in some works the final pH is increased to 11.17 (Table 6) (Donneys-Victoria et al. 2020). Table 2 presents the permissible pH limits by different legislations, between 6–9 and 5.5–10.

In general, temperature, stirring speed, power supply, electrode spacing, mode of operation (batch or continuous), conductivity, ion concentration, contaminant concentration, electrode material, number of electrodes, and cell design are factors that influence contaminant removal (Naje et al. 2017). In coagulation and electrocoagulation, the color removal efficiencies are in the range of 80–93.9%.

Recently the use of Mg²⁺ in wastewater has been proposed for dye coagulation. Ramos et al. (Ramos et al. 2020) proposed the use of wastewater from the saline industry (BW), in the physicochemical treatment of textile effluent containing IB dye (Table 6) with a removal efficiency of 80%. This proposal promotes the use of waste from another industry to treat wastewater from the textile industry; however, BW increases the levels of other pollutants.

The main disadvantage of coagulation and electrocoagulation is sludge production which requires subsequent treatment and neutralization, increasing the treatment costs (Hendaoui et al. 2021). In addition, the consumption of electrical

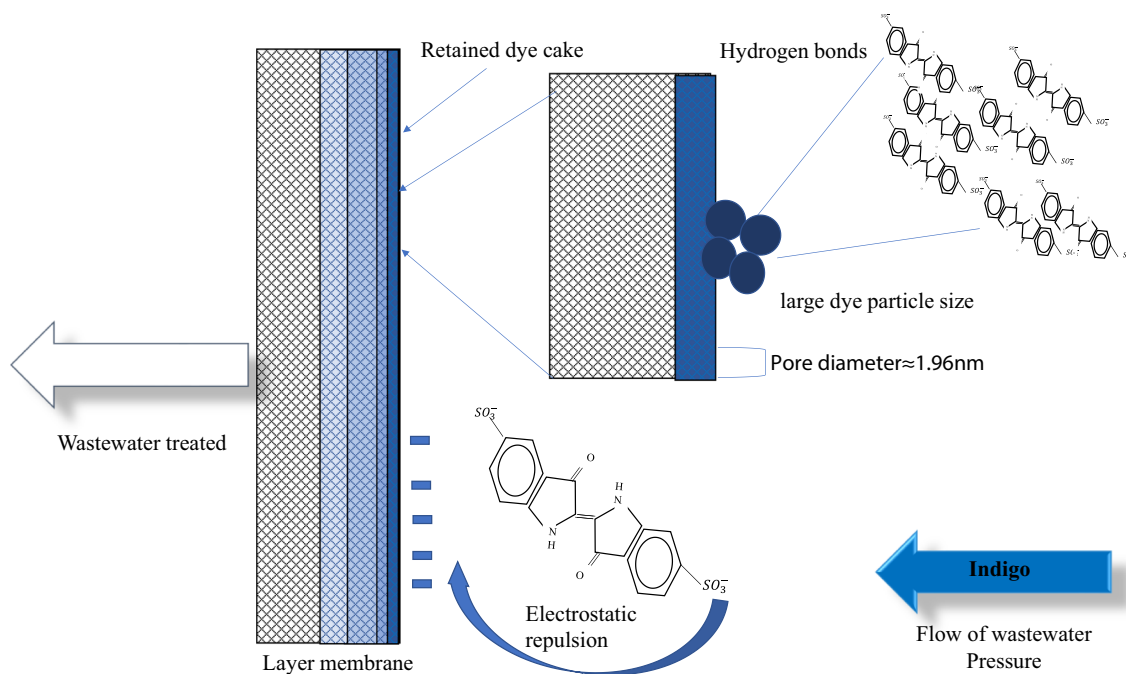


Fig. 5 Indigo dye filtering mechanism

energy is highly criticized because it represents the major limitation in large-scale applications.

Sludge management is a priority due to the final disposal and associated costs that increase when this sludge is generated with chemical agents that are not environmentally friendly. The use of Fe electrodes has been recommended; the resulting generated magnetite can be removed by magnetization. The volumetric rate of sludge formation was 0.2–0.3 L/g with aluminum electrodes and 0.08–0.09 L/g with Fe electrodes, generating more compact layers of sludge (Naje et al. 2017). Generally, the quantity, characteristics or disposal strategies, such as fertilization or soil recovery, are not reported (Table 6).

Advance oxidation processes (AOPs)

AOPs are a set of technologies based on the oxidation of organic pollutants by an oxidizing agent. During these reactions, one or more electrons are transferred from the oxidant to the pollutant, causing degradation or mineralization (Cossu et al. 2018; Rekhate and Srivastava 2020). The advantages of AOPs are the use of environmentally friendly reagents (Ramos et al. 2020), the reduction or generation of harmless sludge (Bello, Abdul Raman and Asghar, 2019a), the production of effluents with lower toxicities

(Mohammadi, Moussavi and Giannakis, 2022) and the use of other forms of energy, such as sunlight (Louhichi et al. 2022). AOPs should not be discriminated against by the costs associated with their application because their environmental and health benefits could be superior.

AOPs commonly used in wastewater treatment are classified as photocatalytic processes where the oxidizing agent is activated by ultraviolet radiation (e.g., $\text{H}_2\text{O}_2/\text{UV}$, $\text{O}_3/\text{H}_2\text{O}_2/\text{UV}$, UV/TiO_2 , $\text{H}_2\text{O}_2/\text{TiO}_2/\text{UV}$, $\text{O}_3/\text{TiO}_2/\text{UV}$) (Berkün Olgun et al. 2021; Pan et al. 2021, 2022). Processes based on Fenton reactions involve an oxidant (hydrogen peroxide) that is activated by a catalyst, usually Fe (e.g., $\text{Fe}^{2+}/\text{H}_2\text{O}_2$, $\text{Fe}^{2+}/\text{H}_2\text{O}_2/\text{UV}$, Electro-Fenton, photo-Fenton, Solar-Fenton) (Bello, Abdul Raman and Asghar, 2019b; Segura et al. 2021; Zhu et al. 2022). Processes based on ozone oxidation (e.g., O_3/UV , $\text{O}_3/\text{H}_2\text{O}_2$, $\text{O}_3/\text{Fe}^{2+}$, $\text{O}_3/\text{ultrasound}$) (Rekhate and Srivastava 2020; Li et al. 2021; Yacouba et al. 2021) and electrooxidation through imposed an electric potential involve the hydroxyl radicals being generated on an electrode surface (Asfaha, Tekile and Zewge, 2021; Ding et al. 2021; Ozyonar and Solmaz 2021; Song et al. 2022). Recently, peracids such as peracetic acid, and salts, such as peroxymonosulfate and persulfate, have been proposed as new oxidants in wastewater treatment (Chen and Liu 2021; Chu et al. 2021; Escobedo, Cho and Chang, 2021; Hao et al. 2021; Kiejza et al. 2021).



Table 6 Operating conditions of coagulation and electrocoagulation processes in the removal of indigo dye in textile wastewater treatment

Country	Technology	Operating conditions	Result	References
Brazil	Coagulation	Wastewater: synthetic 150 rpm for 3 min Slow mixing at 48 rpm for 10 min 30 min settling time 200 mg Mg^{2+} /L 100 mg Mg^{2+} /L corresponded to 7.0 L of BW/m^3	Turbidity: 95% removal Color: 80% removal	(Ramos et al. 2020)
Tunisia	Electrocoagulation	Wastewater: textile industry Electrodes: Fe–Fe pH: 7.5 Inlet flow rate: 2 L/min Voltage: 47 V Zeta potential: -4 mV Dye C_0 : 100 g/10 L Volume: 2 L Surface area: 2877.49 cm^2	Color: 93.9% Cost of 0.0927 USD/ m^3	(Hendaoui et al. 2021)
Colombia	Electrocoagulation	Wastewater: synthetic Electrodes: Mg–Al Surface: Mg = 76.5 cm^2 and Al = 46.6 cm^2 Batch mode Volume: 1 L Dye C_0 : 0.9 g/L Sodium chloride initial: 1 800 mg/L Interelectrode distance: 0.5 cm j: 0.34 A Temperature: 25 °C Electric charge supply: 0.86 Ah/L and 1.3 Ah/L pH: 11.17 Polarity change time: 2 min	NPOC: 90.4% Cl^- 69.1%	(Donneys-Victoria et al. 2020)
Tunisia	Electrocoagulation	Wastewater: textile industry pH: 7.2 Inlet flow rate: 1.1 L/min Voltage: 66 V Color initial: 1750 U Pt–Co COD initial: 2080 mg/L Conductivity initial: 12,372 $\mu S/Cm$ pH: initial 12.2 Volume: 2.4 L 2 Chamber contains a set of 28 iron parallel electrodes Electrodes surface area: 2844.7 cm^2 Electrodes: Fe–Fe	Color removal: 89.2% COD removal: 76.1% Conductivity removal: 29.76% Cost of 0.527 USD/ m^3	(Hendaoui et al. 2018)
Turkey	Electrocoagulation	Wastewater: synthetic pH: 5.8 j: 13.31 mA/ cm^2 Dye $_0$: 20 mg/L Time: 115.8 min Sludge: 0.928 kg/ m^3 Electrode consumption: 0.0305 kg/ m^3 Energy consumption: 7.461 kWh/ m^3 Operating cost: 0.79 US\$/ m^3	Dye removal: 82.5%	(Tanyol et al. 2021)
Turkey	Electrocoagulation	Wastewater: textile industry Electrodes: Al–Fe j: 5 mA/ cm^2 pH: 5 Area of each electrode: 20 cm^2	TOC: 2.5% removal COD: 18.6% removal, Turbidity: 83.5% removal TSS: 64.7% removal Color: 90.3–94.9% removal	(Bener et al. 2019)



Table 6 (continued)

Country	Technology	Operating conditions	Result	References
Tunisia	Electrocoagulation	Wastewater: synthetic Electrodes: Pb Electrode distance: 4 cm Vol.: 100 mL Electrolyte support: the amount of sodium nitrate (NaNO ₃): 10 g/L Current density: 0.5 A/m ²	Direct current (color removal): 98.8% Alternating current (color removal): 100%	(Othmani et al. 2017)
Brazil	Electrocoagulation	Wastewater: synthetic Dye ₀ : 0.002% (w/v) pH: 7.0 Electrolyte support: 0.05 mol NaCl Time: 80 min Current: 0.01 A Temperature: 28 °C Electrodes: steel, aluminum, bronze	Aluminum commercial electrode: color removal 84% Steel commercial electrode: color removal 90% Bronze commercial electrode: color removal 96%	(Oliveira et al. 2019)
Thailand	Electrocoagulation	Wastewater: textile industry pH: 4.0 Time: 60 min, j: 300 A/m ² Vol.: 500 mL Electrodes: Al Electrode area: 50 cm ² Electrode distance: 15 mm	COD removal: 71.9% Color removal: 96.3%	(Prayochmee, Weerayuttil and Khuanmar, 2021)
Turkey	Electrocoagulation	Wastewater: textile industry Electrodes: Fe-stainless steel pH: 8.27 Current: 0.8 A j: 20.78 mA/cm ² Conductivity: 4080 µs/cm Time: 15 min Distance: 10 mm	Color removal: 97.8%	(Kahraman and Şimşek 2020)

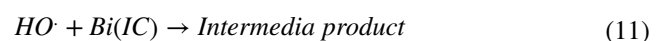
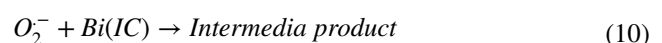
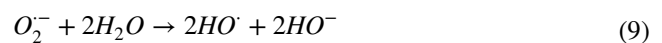
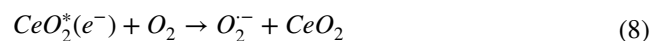
j: Current density

Photocatalytic methods

Table 7 presents the operating conditions and removal efficiencies of the removal of color and other contaminants by photocatalytic methods. In general, most of these treatments have been evaluated in synthetic water. The real wastewater matrix is more complex (Chavan 2015; Gallego-Schmid and Tarpani 2019; Paździor, Bilińska and Ledakowicz, 2019; Singh et al. 2019), and the corresponding color removal efficiencies are in the range of 82–100%.

Nanoparticles as photocatalysts for denim wastewater treatment have been used successfully, (N et al. 2021), and the corresponding removal efficiencies range between 88 and 93.9% (Table 7) (Uma, Ananda and Nandaprakash, 2019; N et al. 2021). Cerium oxide and zinc oxide were applied in oxidation reactions with Ce nanoparticles, and electrons are excited and pass from the valence band to the conduction band by the effect of photons received from UV radiation, which generates an equal number

of positively charged holes. The electrons generated are transferred to the catalyst (CeO₂-NPs), and atmospheric O₂ reacts to form superoxide radicals (O₂⁻). In the same way, the holes in the valence band react with water (H₂O) to generate hydroxyl radicals (OH[·]) (N et al. 2021). These produced radicals react with the dye, followed by their molecular breaking until mineralization (Uma, Ananda and Nandaprakash, 2019; N et al. 2021) (Eq. 7–11).



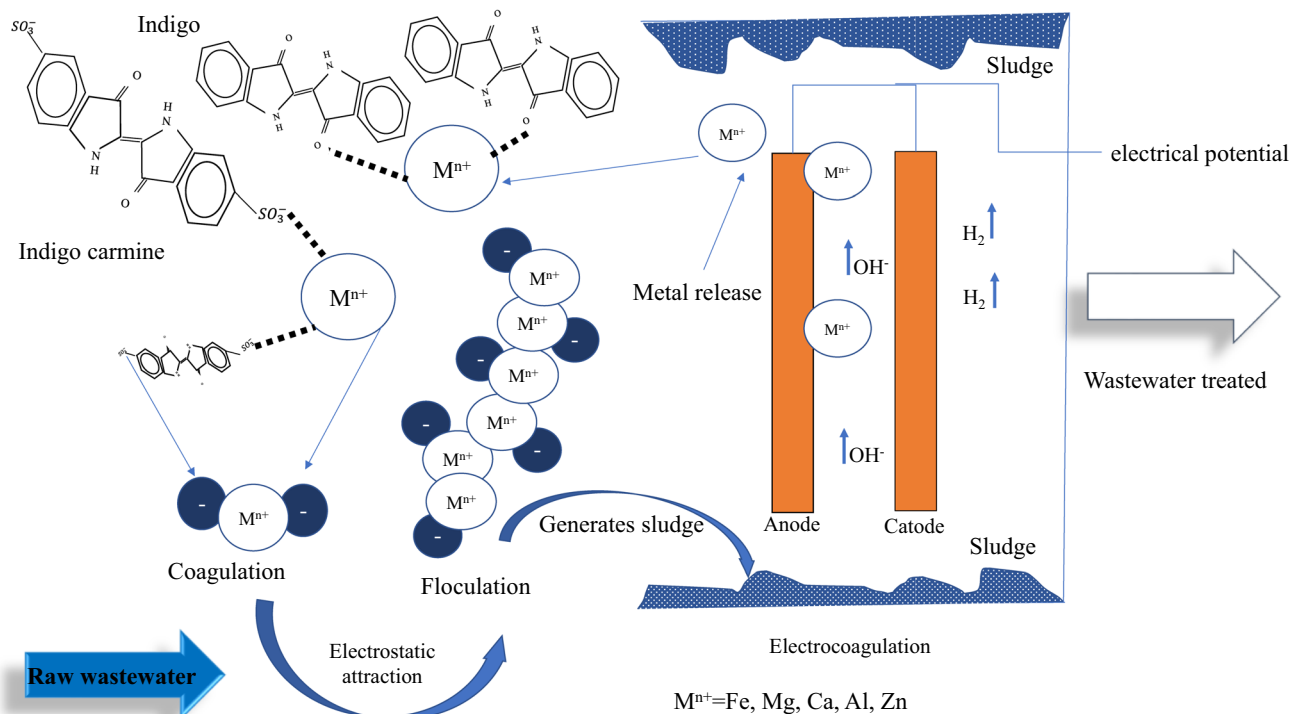
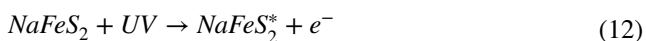


Fig. 6 Coagulation and electrocoagulation mechanism for IB and IC removal

Recently sodium iron disulfide (NaFeS_2) has been synthesized for the photocatalytic degradation of IC; it allows the simultaneous generation of radicals O_2^- and OH^\cdot (Eq. 9); superoxide is generated as indicated by Eqs. 12–13.



One of the advantages of these catalysts is their regeneration and reuse capacity (Uma, Ananda and Nandaprakash, 2019). However, they must be evaluated in real wastewater treatment to determine if there is an inhibitory effect or if the concentration of radicals generated is sufficient to obtain the same efficiencies. IB and IC dyes are generally used as models for the evaluation of synthesized catalysts for their easy application and determination in the laboratory.

Although photocatalysis is indicated as a GT in many works (Güy and Özacar 2018), there are no arguments to confirm this point, and few studies employ solar radiation as a natural source of UV light in denim wastewater. Short wavelength UV light radiation is used in the majority of research, and the UVA light range is 365–405 nm (Table 7); this energy can come from solar radiation.

Solar photocatalysis has been successfully employed in water treatment, disinfection, and the removal of pollutants (Spasiano et al. 2015; Malato et al. 2016). However, its application in denim wastewater treatment has been rarely studied (Malato et al. 2016). Further exploration and development of solar photocatalysis could overcome the disadvantages of using electrical energy to operate lamps and the high costs of UV equipment, (Badli et al. 2017; Gomes et al. 2019). (M., S. and Keerthi, 2019).

Electrooxidation

Electrooxidation consists of the generation of HO^\cdot through the electrolysis of water, by the action of the flow of electrons that are passed through a pair of electrodes, by which the generation of HO^\cdot occurs (Vences-Benitez et al. 2017; Martínez-Huitle and Brillas 2021). Table 8 presents recent studies on electrooxidation in textile wastewater treatment and dye degradation, and the color removal efficiencies range from 92.9 to 100% (Table 8). Boron-doped diamond (BDD) electrodes have been evaluated (Vences-Benitez et al. 2017). Although the COD removal efficiencies ranged from 41 to 80%, the generation of HO^\cdot occurred, as indicated in Eq. 14 (Vences-Benitez et al. 2017).



Table 7 Photocatalysis treatment, operating conditions, and removal efficiencies

Country	Technology	Operating conditions	Result	References
India	Cerium oxide nanoparticles (CeO ₂ -NPs)	Wastewater: synthetic Dye C ₀ : 20 ppm CeO ₂ -NPs: 30 mg UV-Visible light Time: 90 min	Color removal: 88%	(N et al. 2021)
India	Iron disulfide (NaFeS ₂)	Wastewater: synthetic UV light: 125 W Dye C ₀ : 20 ppm/250 mL Photocatalyst: 0.060 g Time: 40 min	Color removal: 99%	(Dileepkumar et al. 2020)
India	Ni-doped ZnO nanoparticles	Wastewater: synthetic Nanoparticles to be 78.3 nm UV light radiation ($\lambda = 325$ nm) Amount of catalyst/dye: 0.02 g Time: 40 min Dye C ₀ : 2X10 ⁻⁵ M pH: 8	COD removal: 93.9%	(Uma, Ananda and Nandaprakash, 2019)
Turkey	Tannin/ZnFe ₂ O ₄ /ZnO	Wastewater: synthetic UV light: 128 W ($\lambda = 365$ nm) UV visible light: 128 W Time: 90 min Dye C ₀ : 16 mg/L Volume: 100 mL Dose of catalyst: 50 mg/100 mL 30 min in the dark Five times recycling	Color removal: 82–99%	(Güy and Özacar 2018)
India	g-C ₃ N ₄ /MnV ₂ O ₆ Heterojunction Photocatalyst	Wastewater: synthetic Tungsten halogen lamp: 300 W Dye C ₀ : 10 mg/L Pyrex glass reactor Time: 60 min Dose of catalyst: 100 mg/100 mL	Color removal: 94% COD removal: 73%	(M., S. and Keerthi, 2019)
Brazil	Free-standing cellulose film containing manganese dioxide nanoparticles	Wastewater: synthetic Time: 25 min Dye C ₀ : 20 ppm Fluorescent light: 16 W pH: 2 Teen times recycling	Color removal: 90%	(Oliveira et al. 2020)
Spain	Photoelectrocatalytic	Wastewater: synthetic Photoanode: TiO ₂ nanotube Aire-diffusion cathode: carbon-PTFE H ₂ O ₂ : 5 mM pH: 3 Electrode's separation: 1 cm Volume: 150 mL Temperature: 25 °C j: 3 mA/cm ² LED lamp: 36 W UV ($\lambda_{max} = 365$ nm and 405 nm) Irradiance: 9 W/m ² Distance lamp: 2–2.5 cm Flow rate: 600 mL/min Supporting electrolyte: Na ₂ SO ₄ Dye C ₀ : 0.26 mM Time: 35 min	Color removal: 100% COT removal: 84.6%	(Oriol et al. 2019)

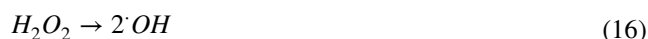


Table 7 (continued)

Country	Technology	Operating conditions	Result	References
Colombia	oligo-phenylenevinylenes applied (SiO ₂ -OPV) as Heterogeneous Photocatalysts	Wastewater: synthetic LED Lamp: 3 W (350- 450 nm) Dose of catalyst: 20 mg/3 mL Dye C ₀ : 50 μM Time: 20 min Temperature: 20 °C tirred: 100 rpm	Color removal: 93%	(Acelas and Sierra 2021)
India	Multi-doped ZnO Solar photocatalysis	Wastewater: synthetic Intensity of solar radiation: 1200–950 W/m ² Time radiation: 3 h Time treatment: 120 min Dye C ₀ : 15 mg/L pH: 9 Dose of catalyst: 2 mg/mL Four times recycling	Color removal: 98%	(Shinde, Patil, G. Do Kim, et al. 2020)
Brazil	TiO ₂	Wastewater: synthetic Dye C ₀ : 6 mg/L pH: 4 Time: 30 min Dose of catalyst: 12 mg/L In the presence of Tert-butanol (t-BuOH)	Color removal: 70–77%	(Neto et al. 2021)
South Africa	Nanocomposite (Nd–ZnO–GO)	Wastewater: synthetic Dye C ₀ : 20 mg/L Dose of catalyst: 100 mg/100 mL Time: 1 h Solar simulator light: λ = 420 nm Five times recycling	TOC removal: 76% Color removal: 95%	(Oppong et al. 2017)
Egypt	AgIO ₄ /ZnO nanoparticles	Wastewater: synthetic Natural sunlight Irradiations Radiation intensities: 1200 and 1450 W/m ² Temperature: 30–35 °C Reactor: quartz Vol: 150 mL Dose of catalyst: 15 wt % AgIO ₄	Color removal: 98%	(Abdel-Aziz, Ahmed and Abdel-Messih, 2020)
South Africa	Eu–TiO ₂ –GO Composite	Wastewater: synthetic Time: 60 min λ > 420 nm Lamp: Xe at 300 W Dye C ₀ : 20 mg/L Stirrer: 200 rpm	Color removal: 96% TOC removal: 76%	(Oppong et al. 2021)



The most influential factor is the acidic pH, which can favor the formation of a high concentration of H₂O₂, by increasing the concentration of hydronium ions as described in Eq. 15 and 16, and therefore improves pollutant degradation (Palma-Goyes et al. 2018). (Vences-Benitez et al. 2017; Castillo-Suárez et al. 2018).



The use of electrical energy for the formation of oxidants is highly criticized since its consumption is high, so this technology is considered unfriendly to the environment (Kaur et al. 2020). There is a trend toward the exploration of more efficient electrodes for the generation of radicals,

Table 8 Recent treatments of textile wastewater by an electrooxidation process

Country	Operating conditions	Result	References
Mexico	Wastewater: industrial Electrodes: BDD-BDD pH: 4.5 j: 6 mA/cm ² Volume: 100 mL Superficial anodic area: 307.5 cm ² Time operation: 780 min COD initial: 3160 mg/L BOD initial: 340.9 mg/L Color initial: 3260 Pt–Co U TOC initial: 1299 mg/L	COD removal: 44.1% TOC removal: 26.5% Color removal: 99.99% BOD final: 57.33 mg/L	(Vences-Benitez et al. 2017)
Mexico	Wastewater: synthetic Anode: Ir-Sn-Sb Cathode: zirconium Stirred: 150 rpm Electrolyte support: 0.25 mol/L NaCl pH: 2 j: 9.375 mA/cm ² Dye C ₀ : 20 mg/L Volume: 150 mL	Color removal: 100% COD removal: 77.5% TOC removal: 24%	(Palma-Goyes et al. 2018)
Finland	Wastewater: industrial Time operation: 143 min pH: 5.4 i: 1.41 A Anode electrode: Aluminum Cathode electrode: Ti/RuO ₂ Anode dimension: 100 × 85 × 1.5 mm Cost: US\$ 8.56 / kg of COD pH: 9.8 BOD initial: 196 mg/L COD initial: 1156 mg/L Color initial: 1410 Pt–Co U	BOD final: 41.5 mg/L COD final: 219.8 mg/L Color final: 71 Pt–Co U	(Kaur et al. 2020)
Turkey	Wastewater: synthetic textile Anode electrode: TiO ₂ Cathode electrode: Graphite Volume: 120 mL Graphite area = 59 mm × 57 mm TiO ₂ area: 49 mm × 122 mm Distance electrodes: 25 mm i: 0.3 A pH: 7 Electrolysis time: 120 min Voltage: 20.3 V	Color removal: 92.9%	(Bakaraki Turan et al. 2021)

*Boron-Doped Diamond: BDD

the use of renewable energy sources such as solar panels, for the power supply (Fadhil et al. 2021), and coupled treatments. Biorefractory compounds are oxidized to less toxic and biodegradable by-products (Zhang et al. 2020) reducing energy consumption.

Fenton oxidation

The Fenton reaction results in the generation of HO[·] and has been applied in the oxidation of persistent organic

compounds such as dyes. The combination of hydrogen peroxide with Fe²⁺ salts at optimum pH (2.8) generates radicals as shown in Eq. 17. Fe is the catalyst of the reaction, improving the rate of reaction by oxidizing from Fe²⁺ to Fe³⁺ (Cossu et al. 2018; Gonçalves et al. 2020).

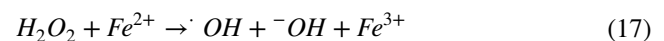


Table 9 presents the operating conditions for Fenton processes. The color removal efficiencies range from 98 to

Table 9 Operating conditions for Fenton reaction oxidation treatments of textile wastewater and dyes

Country	Technology	Operating conditions	Result	References
Mexico	Heterogeneous Fenton catalyst	Wastewater: textile industry Catalytic iron oxide nanoparticles: 2 g/L pH: 3.2 H ₂ O ₂ : 0.32 mM Time: 20 min Volume: 50 mL COD initial: 2360 mg/L Stirred: 60 rpm	Color removal: 99% COD removal: 83%	(De León-Condés et al. 2019)
Brazil	Photo-Fenton	Wastewater: synthetic Dye C ₀ : 50 mg/L Time: 60 min Fe ²⁺ : 4 mmol/L Ratio: H ₂ O ₂ /Fe ²⁺ : 6:1 Mercury vapor lamp: 90 W UV-C (λ = 254 nm) Surface irradiance: 130.2 mW/cm ² Lamp-reactor distance: 1.55 cm Volume reactor: 1.5 L Recirculation flow rate: 10 L/min	TOC removal: 75%	(Ramos et al. 2020)
Spain	Photo-electro-Fenton	Wastewater: synthetic Dye C ₀ : 0.26 mM Supporting Electrolyte: 0.050 M Na ₂ SO ₄ Fe ²⁺ : 0.50 mM pH: 3.0 UV LED: λ _{max} = 365 nm and 405 nm Irradiance: 29 W/m ² Distance lamp: 2.0–2.5 cm Temp: 25 °C Photoanode: TiO ₂ nanotube Aire-diffusion cathode: carbon-PTFE Time: 35 min j: 3 mA/cm ²	COD removal: 100%	(Oriol et al. 2019)
Brazil	Photo-Fenton	Wastewater: textile industry pH: 3.5 Ratio molar Fe:H ₂ O ₂ : 1:1.4 Quartz tube Vol: 280 mL Mercury-vapor lamp: 6 W Radiation: 254 nm Intensity of the mercury-vapor lamp: 1.03 × 10 ⁻⁶ Einstein/min Total Cost: 0.509 US\$/m ³	Color removal: > 98.5% COD removal: 69.8%	(Lebron et al. 2021)
Brazil	Photo-Fenton	Wastewater: synthetic Lamp: Master HPI-T 400 W Catalytic: 20 mg of Fe ₃ O ₄ /RGO Dye C ₀ : 10 mg/L H ₂ O ₂ : 30% Time: 5 min pH: 6	Color removal: 100%	(Gonçalves et al. 2020)

*NTs = nanotube arrays, Fe₃O₄/RGO: magnetite/reduced graphene oxide, PTFE: Polytetrafluoroethylene

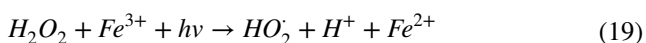
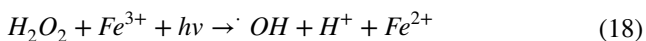
100%. Fenton reactions increase radical generation without the generation of residual peroxide by UV radiation, this process is known as a photo-Fenton processes. According to Eq. 18, UV radiation allows the regeneration of the

catalyst and favors the formation of a higher concentration of HO[•] (Eqs. 19–20) (Lebron et al. 2021), and UV radiation allows the maximum utilization of the H₂O₂ dose, limiting the occurrence of residuals (Castañeda-Juárez et al. 2020).

Table 10 Other wastewater treatment systems in the textile industry based on dye oxidation

Country	Technology	Operating conditions	Result	References
Mexico	Ferrate	Wastewater: industry Color initial: 20 and 30 mg/L Na ₂ FeO ₄ at a dose of 200 mg/L -200 mg/L of K ₂ FeO ₄ Time: 30 min	Color removal with Na ₂ FeO ₄ : 94–96% Color removal with K ₂ FeO ₄ : 92.5% Turbidity: 96% COD: 56.3% Sulfates: 24.8% Ammonia nitrogen: 50.5% Nitrates: 94.4% TSS: 42.6% Oil and grease: 100% pH final: 11.4 TDS: 11.2%	(Castañeda Juárez et al. 2020)
Colombia	Electrocoagulation and electrooxidation	Wastewater: industry USD: 1.47/m ³ pH: 4 Conductivity: 3.7 mS/cm j: 4.1 mA/cm ² Electrodes: Fe/Al, Gr/Ti	COD removal: 73% Turbidity removal: 95% Total solid removal: 54% TOC removal: 76% BOD ₅ : 37% Individual contribution COD removal: EC: 51% EO: 21%	(GilPavas et al. 2017)
Japan	Argon dielectric barrier discharge (plasma reactor)	Wastewater: synthetic Gap distance: 1.0 and 2.5 mm Excited: 3 kHz Power: 1.72 and 1.76 W Dye ₀ : 20 mg/L Production H ₂ O ₂ : 15–18 mg/L	Color removal: 99.3%	(Yonezawa et al. 2019)
Japan	Photolysis	Wastewater: synthetic Irradiance: 8.7 mW/cm ² Distance: 20 mm Dye ₀ : 3.33 × 10 ⁻⁴ mol/L λ: 147 nm Lamp: Hg Radiation dose: 50 J/cm ²	TOC removal: 49%	(Zukawa et al. 2019)
Brazil	Nonthermal plasma	Wastewater: synthetic Working plasma gases were O ₂ Flow rate: 0.5 L/min Frequency pulse: 100 Hz Energy pulse: 50 mJ Initial pH: 5.8 Conductivity: 1.0 μS/cm Vol: 100 mL Dye ₀ : 20 mg/L Time: 10 min	Color removal: 100%	(Crema et al. 2020)

EC Electrocoagulation, EO Electrooxidation

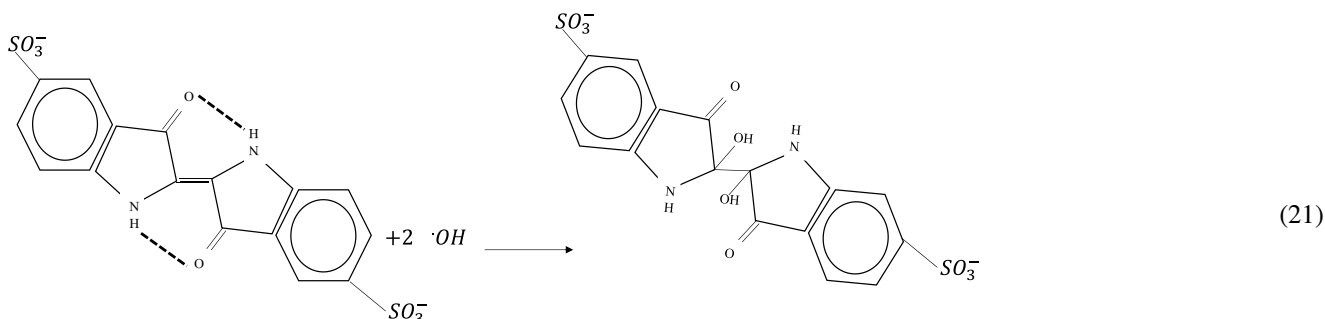


The acidic pH contributes to the stability and maintenance of the H₂O₂ oxidation potential, causing the concentration of active Fe²⁺ species to be high and H₂O₂ decomposition to be minimized (Ramos et al. 2020). The best-operating conditions for Fenton reactions are mostly at acidic pH

(Table 9), to maintain its oxidation potential ($E^\circ = 2.85$ V) (Crema et al. 2020). However, a neutral pH is desirable to avoid reagent consumption in acidification and changes in the chemical properties of the contaminants. An alternative for precluding these disadvantages is heterogeneous Fenton reactions; however, few works have discussed their use and application in denim wastewater treatment. Heterogeneous systems employ a catalyst that is easy to separate and recover, and no pH adjustments are required to obtain high removal efficiencies since they operate at a neutral pH (Dai et al. 2018; Gonçalves et al. 2020). The catalyst can be easily separated from the medium with a magnetic bar, and it

can be used at least one more time without losing its activity (Gonçalves et al. 2020).

The oxidation of the dyes by the action of hydroxyl radicals can occur by breaking the carbon–carbon double bond in the indigo structure by hydroxylation, which accompanies the breaking of carbonyl bonds. This generally occurs in the first few minutes of the reaction, within 3–35 min (Table 9). The main chromophore groups of the molecule are broken and the blue color of the dye dissipates rapidly (Ramos et al. 2020). The hydroxyl radical oxidation reactions are presented in Eq. 21.



Other treatment systems

Other treatment systems based on the generation of an oxidizing agent are presented in Table 10. The corresponding color removal efficiencies range from 92 to 100%. The combination of treatments such as electrocoagulation and electrooxidation had COD removal efficiencies of 73% (GilPavas et al. 2017). Photolysis was enhanced by the formation of hydroxyl radicals according to Eq. 22 with a TOC removal efficiency of 49% (Zukawa et al. 2019).

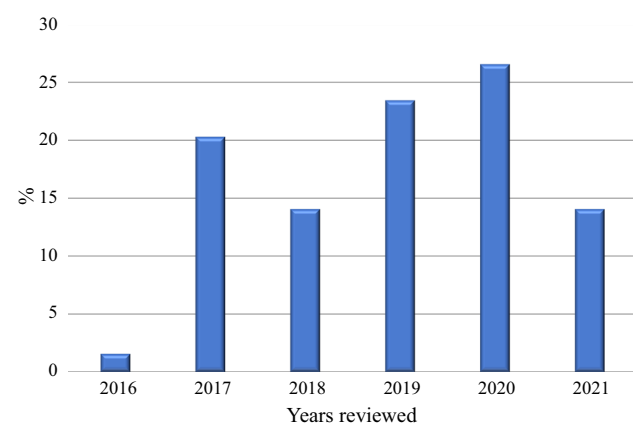
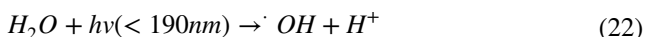


Fig. 7 Trend in the evaluation and development of technologies for the removal of indigo blue dye

The new generation of AOP, applying plasma discharge to improve the removal efficiency of IC dye, has also been used in the oxidation of persistent organic compounds generating hydroxyl radicals. Although there are many types of water treatment reactors based on plasma discharge, the most used are pulsed corona discharge excitation, the bubbling of in-water droplets, or those applied to water surfaces using a pulsed energy source (Yonezawa et al. 2019; Crema et al. 2020).

Excited argon gas has been employed in the generation of hydroxyl radicals in solution with IC dye, with a color removal efficiency of 99.3% (Yonezawa et al. 2019). Oxygen

gas proved to be more efficient than nitrogen in the removal of color in a nonthermal plasma process because it could favor peroxide formation (Crema et al. 2020). Reactions related to the formation of $\cdot\text{OH}$ radicals by argon dielectric barrier discharge produced on liquid surface are described in Eq. 23 and 24:



Critical analysis and future trends

Figure 7 shows the percentage of reviewed studies, from 2016 to 2021, in which technologies for the treatment or removal of indigo blue and indigo carmine were evaluated and developed. There is no clear trend in the development of strategies for mitigating the contamination of denim wastewater in contrast with the growth of this industry (Agarwal et al. 2017; Cercetare-dezvoltare et al. 2017; Oliveira et al. 2019; Hajjar 2020).

From 2018 to 2020, an increasing trend in publications was observed, and in 2021, a decrease occurred. It has been indicated that during the COVID-19 crisis, a massive flow of publications was oriented to this topic, generating more than 20,000 papers published from December 2019 to July 2020 (Harper et al. 2020). The decrease publications related to the removal of indigo blue in 2021 could have been affected by

the isolation and closure of laboratories. The pandemic crisis has had a secondary effect on scientific development, which can be evaluated in the medium or long term.

The regulations analyzed show a variety of parameters and maximum permissible limits for contaminants present in denim wastewater. This variability could be related to the particular characteristics of the processes, production stage, and final finishing of denim garment (Paul 2015; Periyasamy and Militky 2017). Therefore, is necessary to improve and apply the international regulations, evaluate the characteristics of wastewater based on the particularities of the region or process. Color is the most characteristic parameter for denim wastewater; however, it is not regulated for some countries (Table 2), which shows a contradiction and an inconsistency between the standards and the physicochemical characteristics for denim wastewater. Regulations play an important role in controlling pollution and, ensuring the protection of the environment and public health, which contributes to maintaining the economic growth of nations and a sustainable textile industry to ensure continued growth.

The methods of treatment and removal of pollutants from denim wastewater were classified into 3 major groups, (1) biological systems, (2) physicochemical treatments, and (3) advanced oxidation processes. The AOPs have been mostly used, and 41% of the studies on the removal of IB and, IC, and the treatment of textile wastewater were evaluated (Fig. 8a), followed by physicochemical (34%) and biological (25%) treatments. Dyes (IB and IC) from the denim industry have been mostly used as model contaminants for color removal.

Figure 8b shows that 69.8% of the reviewed studies used synthetic wastewater or aqueous solutions, and only 30.2% evaluated real wastewater. The matrix of components in industrial wastewater is more complex (Chavan 2015; Gallego-Schmid and Tarpani 2019; Paździor, Bilińska and Ledakowicz, 2019; Singh et al. 2019; Hevira et al. 2020). IB and IC are evaluated under ideal conditions, which limits technological development and application in industrial wastewater. A future trend should be to use real wastewater for evaluation. It is important to consider that new dyeing processes are frequently developed (Muthu 2018); therefore, the generation and adaptation of new sustainable technologies will be necessary.

Figure 8c shows the average color removal percentages regardless of the initial and treatment conditions. The studies reviewed in this research were classified by 3 treatment groups and the type of water used. In synthetic wastewater, AOPs had an average color removal efficiency of 95.3%, followed by biological treatments with 90.9% and physicochemical treatments with 84.2%. In real wastewater, AOPs had an average efficiency of 97.5%,

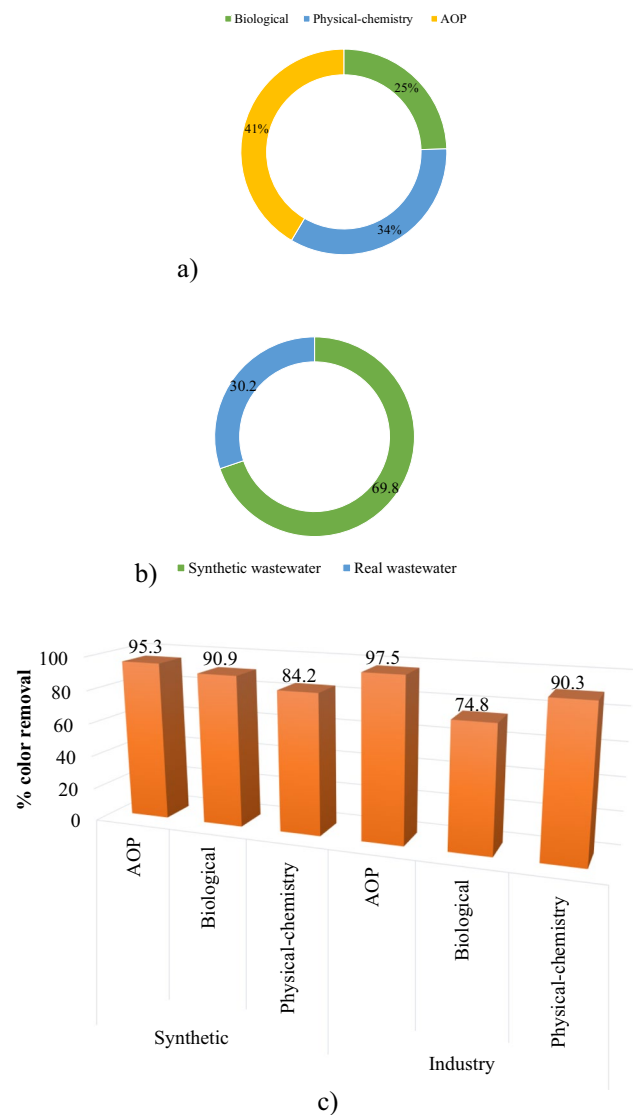


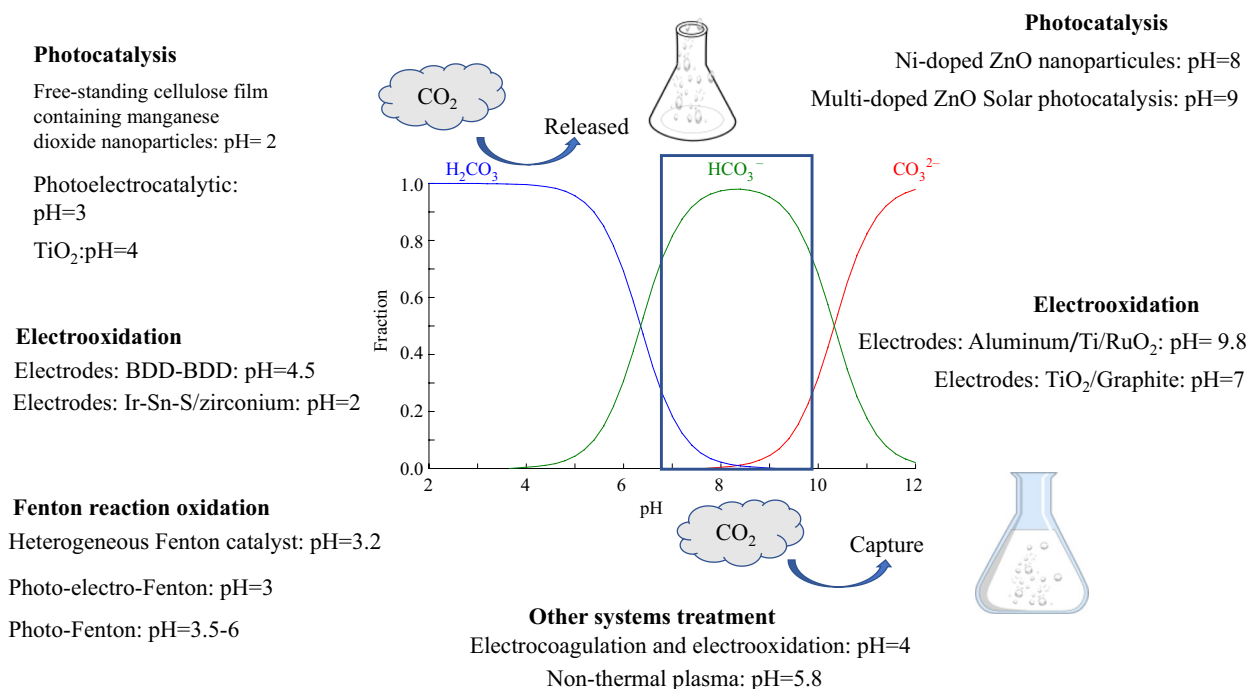
Fig. 8 Trends in **a** studies evaluating color removal according to treatment type, **b** water type and **c** removal efficiencies (%) of treated water vs. treatment systems

followed by physicochemical processes with 90.3% and biological treatments with 74.8%. Biological systems were less efficient when treating textile wastewater; this may be related to the toxic effects of the water components, such as the peroxide or residual hypochlorite used in the bleaching or washing of denim (Arputharaj et al. 2016; Roy Choudhury 2017) or the presence of heavy metals from dye fixation (Paul 2015; Uddin and Sayem 2020). In general, AOPs are the best strategy for color removal.

The textile industry must gradually ensure sustainable wastewater treatment to achieve the reuse or recovery of wastewater and reduce the consumption of reagents, carbon footprint, consumption of inputs, and energy

Characteristics	Technologies							
	Biological treatment	Adsorption	Filtration	Coagulation and Electrocoagulation	Photocatalytic	Electrooxidation	Fenton oxidation	Plasma reactor
Employs more durable materials	☆	★	☆	☆	★	★	★	☆
Improved removal efficiency	☆	★	★	★	★	★	★	★
Reduction in treatment time	☆	★	★	★	★	☆	★	★
Reduction cost of the treatment	★	☆	☆	☆	☆	☆	☆	☆
Reduction of the fuel burn and the CO ₂ emission	★	★	★	★	☆	☆	☆	☆
Generates a product innocuo	★	★	★	☆	★	★	★	★
Reduces reagent or consumables	★	☆	☆	☆	☆	★	☆	☆
Reduce or eliminate waste	★	☆	☆	☆	★	★	★	★
Improvement of recycle rate of the material	☆	☆	☆	☆	☆	☆	☆	☆
Reduces energy consumption	★	☆	☆	☆	★	☆	★	☆
Total	6	5	4	3	6	5	6	4

a)



b)

Fig. 9 a Characteristics of a GT and bevaluated technologies that allow CO₂ capture and carbon footprint reduction

consumption. One trend is the use of life cycle analyses to evaluate sustainable GT approaches, so a robust database must be developed to ensure adequate analysis. Of all GTs analyzed in this review, none were demonstrated to be totally environmentally friendly (Figs. 3 and Fig. 9a), so in this review the desirable characteristics of GTs were proposed (Campos-Guzmán et al. 2019; Moni et al. 2020).

AOPs are highly efficient in the removal of color and organic matter; however, the complete mineralization of contaminants represents an increase in the carbon footprint of the process contributing to global warming (Vidya Lekshmi, Yesodharan and Yesodharan, 2018; Oriol et al.

2019; Vorontsov 2019; Nguyen et al. 2020; Wang and Zhu 2020). Therefore, for the mineralization of organic matter into CO₂, some strategies for its capture (in the form of HCO₃⁻ and CO₃²⁻) are recommended, such as photocatalysis and electrooxidation (Shinde, Patil, G. Do Kim, et al. 2020) (Fig. 9b).

In this review, no reports of upscaling up or pilot projects for treating water from the textile industry were found. A suggested future trend is the application of GTs on an industrial scale to evaluate operating conditions, technical feasibility, and economic costs.



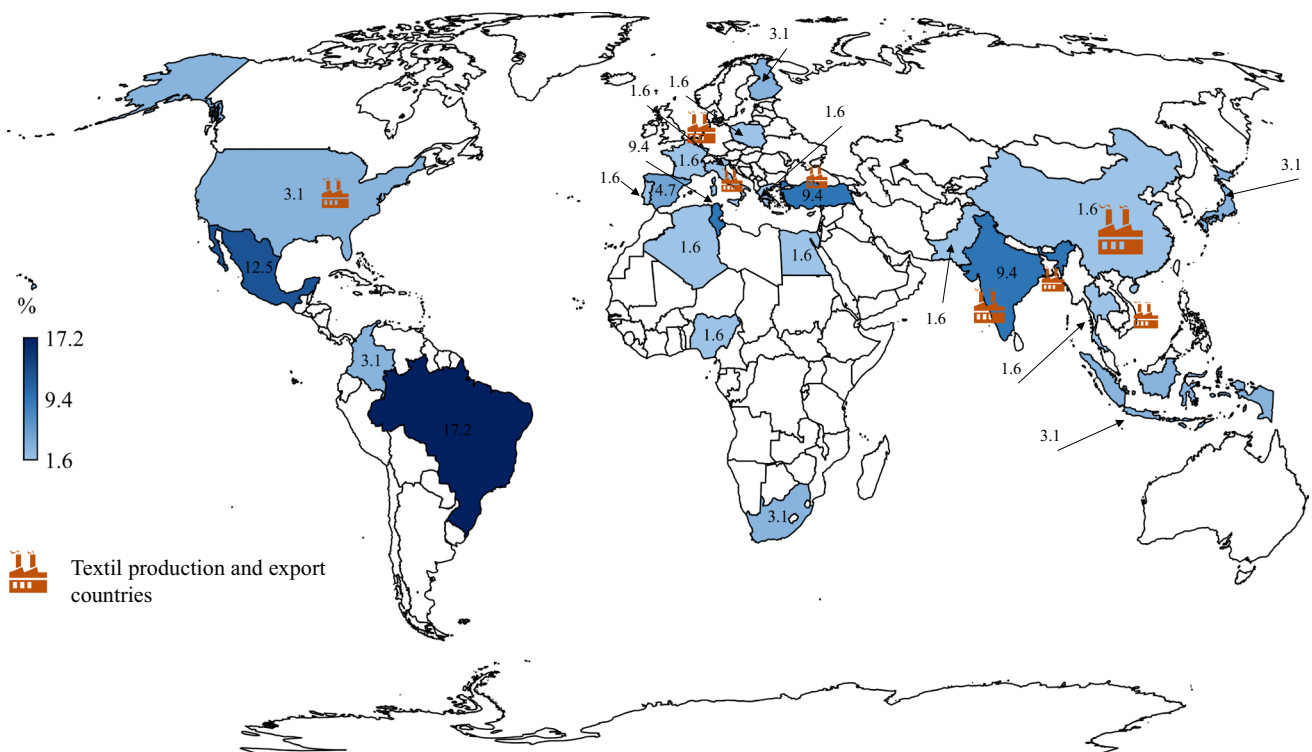


Fig. 10 Percentage of studies reported between 2016 and 2021 on denim wastewater treatment and countries with the largest textile production

Figure 10 presents a world map indicating the locations where textile wastewater treatment studies have been reported (Agarwal et al. 2017; Cercetare-dezvoltare et al. 2017). Of the papers reviewed, 17.2% were reported from Brazil, followed by Mexico (12.5%), India (9.4%), Turkey (9.4%), and Tunisia (9.4%). In contrast, the countries with the highest textile production and exports are China, Germany, Italy, the USA, India, Bangladesh, Vietnam, and Turkey. This is because there is no recent published information.

China is the largest exporter worldwide, ranging from cotton production to garments. Bangladesh is the second-largest exporter of clothing, and the textile industry is of great importance for the economic development of this country. India is the second-largest producer of cotton fiber and the second-largest exporter of textiles and garments in the world, after China (Agarwal et al. 2017).

Regulations play an important role in pollution mitigation; however, countries with specific regulations adapted to the particular conditions of their textile wastewater make less effort to generate new remediation technologies.

Conclusion

AOPs have been used mostly in the removal of dyes from denim wastewater (41%). These technologies have shown removal efficiencies of 95.3 and 97.5% in synthetic and real wastewater, respectively, from IB and IC dyes, so they could be the most suitable technologies for the removal of color and contaminants in textile effluents. The technologies that have only been tested in the removal of synthetic water must be promptly evaluated in the treatment of textile wastewater to identify the feasibility of their application at the industry scale.

GTs must have several desirable characteristics and demonstrate that they favor the reduction of energy consumption and the carbon footprint, reducing inputs, using clean energy, etc. It is necessary to promote the use of strategies to demonstrate the green profile of these technologies and avoid self-branding as a GT without supporting evidence.

The parameters and maximum permissible limits of pollutants in textile wastewater of different international legislations were found to be significantly inconsistent. Regulatory guidelines play an important role in the sanitation, assurance, protection, and reuse of water to preserve human



health and environmental protection, so they should be promoted and applied. Some inconsistencies were observed in the results of the treatments reviewed because they are not discussed in terms of the guidelines and maximum permissible limits established by the legislation.

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Authors contributions Luis Antonio Castillo-Suárez drafted the work or revised it critically for important intellectual content; conception or design of the work; literature search and data analysis. Ana Gabriela Sierra-Sánchez made substantial contributions to the work design; drafted the work or revised it critically for important intellectual content. Ivonne Linares-Hernández approved the version to be published; drafted the work or revised it critically for important intellectual content; critically revised the work; Conceptualization. Verónica Martínez-Miranda made substantial contributions to the work design; or the acquisition, analysis, or interpretation of data. Elia Alejandra Teutli-Sequeira made substantial contributions to the work design; drafted the work or revised it critically for important intellectual content.

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