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REVIEW ARTICLE

Effects of textile dyes on health and the environment and bioremediation potential of living organisms

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Abstract The water is an essential resource for life on the planet and for human development. The textile industry is one of the anthropogenic activities that most consume water and pollute water bodies. Therefore, the present work aims to undertake a review on the main effects of the release of industrial dyes and the essential bioremediation mechanisms. The textile dyes significantly compromise the aesthetic quality of water bodies, increase biochemical and chemical oxygen demand (BOD and COD), impair photosynthesis, inhibit plant growth, enter the food chain, provide recalcitrance and bioaccumulation, and may promote toxicity, mutagenicity and carcinogenicity. In spite of this, the bioremediation of textile dyes, that is, the transformation or mineralization of these contaminants by the enzymatic action of plant, bacteria, extremophiles and fungi biomasses is fully possible. Another option is the adsorption. Despite some disadvantages, the bioremediation is essentially positive and can be progressively enhanced by modern biotechnological techniques that are related to the generation of more degrading and more resistant engineered organisms. This is a sustainable solution that provides a fundamental and innovative contribution to conventional physicochemical treatments. The resources of environmental biotechnology can, therefore, be used as tangible technological solutions for the treatment of textile dye effluents and are related to the ethical imperative of ensuring the minimum necessary for a quality life for the humankind.

Introduction

The textile industry is spread globally, generating around 1 trillion dollars, contributes with 7% of the total world exports and employs around 35 million workers around the world (Desore & Narula, 2018).

Despite its undeniable importance, this industrial sector is one of the biggest global polluters and it consumes high amounts of fuels and chemicals (Bhatia, 2017). The special emphasis is placed on the enormous use of drinking water in various operations of its production chain, such as washing, bleaching, dyeing, among others (Hossain, Das, Islam, Al Mamun, & Khan, 2018).

The textile industry is responsible for an extensive list of environmental impacts (Muthu, 2017). The air pollution

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produced involves, for example, the release of particulate matter and dust, oxides of nitrogen and sulfur and volatile organic compounds. The scraps of textile fabrics and yarns and discarded packagings constitute the primary solid waste. The textile sludge, on the other hand, reveals problems related to surplus volumes and unwanted composition, often presenting high loads of organic matter, micronutrients, heavy metal cations and pathogenic microorganisms (Bhatia, 2017).

The main damages caused by the textile industry to the environment, however, are those resulting from the discharge of untreated effluents into the water bodies (Bhatia, 2017), which normally constitute 80% of the total emissions produced by this industry (Wang, 2016). In the composition of most of the residual waters of the textile industry there are relatively high levels of biochemical oxygen demand (BOD) and chemical oxygen demand (COD) (Setiadi, Andriani, & Erlania, 2006). The greater emphasis should be attributed to the large amount of non-biodegradable organic compounds, especially textile dyes (Orts, del Río, Molina, Bonastre, & Cases, 2018).

The dyes are soluble organic compounds (Mahapatra, 2016), especially those classified as reactive, direct, basic and acids. They exhibit high solubility in water making it difficult to remove them by conventional methods (Hassan & Carr, 2018). One of its properties is the ability to impart color to a given substrate (Shamey & Zhao, 2014) because of the presence of chromophoric groups in its molecular structures. However, the property of fixing the color to the material is related to the auxotrophic groups, which are polar and can bind to polar groups of textile fibers (Wardman, 2017).

The color associated with textile dyes not only causes aesthetic damage to the water bodies (Setiadi et al., 2006), but also prevents the penetration of light through water (Hassan & Carr, 2018), which leads to a reduction in the rate of photosynthesis (Imran et al., 2015) and dissolved oxygen levels affecting the entire aquatic biota (Hassan & Carr, 2018). The textile dyes also act as toxic, mutagenic and carcinogenic agents (Aquino, Rocha-Filho, Ruotolo, Bocchi, & Biaggio, 2014; Khatri, Nidheesh, Singh, & Kumar, 2018), persist as environmental pollutants and cross entire food chains providing biomagnification (Sandhya, 2010), such that organisms at higher trophic levels show higher levels of contamination compared to their prey (Newman, 2015). In this sense, special mention should be made to azo-type textile dyes which, around 15–50%, do not bind to the fabric, during the dyeing process, and are released into wastewater which is commonly used, in developing countries, for the purpose of irrigation in agriculture (Rehman et al., 2018). The use of these azo compounds is very negative to soil microbial communities (Imran et al., 2015) and to germination and growth of plants (Rehman et al., 2018).

It is therefore essential to use treatment strategies (Orts et al., 2018), aiming to ensure the sustainability of the environment to future generations (Jordão, Puppim, & Broega, 2018) through physical, chemical and biological technologies or a combination of them (Setiadi et al., 2006). It is observed that the physical-chemical means, although successful, involves the inconveniences generated by the sludge disposal and the high costs with electricity, inputs or operation (Imran et al., 2015). The biodegradation is proposed as the most economical resource for the treatment of textile effluents (Paździor et al., 2017) when using enzymes secreted extracellularly by microorganisms (Doble & Kumar, 2005), such as azoredutases, laccases and the peroxidases (Imran et al., 2015) in order to transform or even mineralize textile dyes (Pajot, Figueroa, Spencer, & Fariña, 2008). In addition, there is the bioabsorption that can act when the dye is very toxic to the growth of the microorganism, through absorption, deposition and ion exchange, using dead bacteria, yeasts and fungi (Doble & Kumar, 2005). In this sense, the present work aims to investigate the main consequences to the health and environment caused by textile dyes and the potential of living organisms that can be used in the bioremediation processes of these pollutants.

Problems related textile dyes' environmental contamination

The textile dyes, along with a large number of industrial pollutants, are highly toxic and potentially carcinogenic (Sharma, Dangi, & Shukla, 2018), so that they are related to environmental degradation and various diseases in animals and humans (Khan & Malik, 2018).

The tendency to be recalcitrant in aerobic environments, especially in conventional treatment plants, is responsible for bioaccumulating the dyes in sediments and soil and transporting them to public water supply systems (Vikrant et al., 2018). Despite the environmental recalcitrance of the majority, they can be partially degraded or transformed in the presence of anoxic sediments, as occurs in the reduction of the azo-type compounds causing dangerous aromatic amines (Ito, Adachi, Yamanashi, & Shimada, 2016). Another possibility involves combining dyes with intermediate synthetic compounds or their degradation products to generate other mutagenic and carcinogenic substances (Vikrant et al., 2018).

The consequences of the xenobiotic and recalcitrant nature of the dyes end up being impacting to the structure and functioning of the ecosystems (Rawat, Mishra, & Sharma, 2016). Long-term exposures, in particular, bring profound unfoldings, for example, to aquatic biota and to human health (Ito et al., 2016), as is the case with complexed metal dyes. This category of dyes is widely used by the textile industry, given their resistance, and exhibits half-lives of 2–13 years (Copaciu et al., 2013) presenting in their composition nickel (Brock, Groteklaes, & Mischke, 2000), copper, cobalt and, above all, chromium (Christie, 2001). Once released in the aquatic environment, the heavy metal cations can be assimilated by the fish gills, because they present negative charges, allowing their accumulation in certain tissues (Vargas, Paulino, & Nozaki, 2009). Thus, through the food chain, they can reach the human organs causing a series of pathologies (Khan & Malik, 2018). Moreover, oxidative stress, provided by chromium of textile dyes, is another problem associated with recalcitrant character, offering a considerable damage to the growth and development of plants, especially to photosynthesis and CO_2 assimilation (Copaciu et al., 2013).

Although dyes have been known to mankind since ancient times (Vankar, 2016), it was not until the late nineteenth century that synthetic forms began to be manufactured, along with intermediate compounds, causing high

incidence of bladder cancer, especially, benzidine and 2-naphthylamine (Christie, 2007). In general, the diseases provided by textile dyes comprise from dermatitis to disorders of the central nervous system (Khan & Malik, 2018) or may be related to the substitution of enzymatic cofactors that result in the inactivation of the enzymatic activities themselves (Copaciu et al., 2013).

The acute toxicity to textile dyes is caused by oral ingestion and inhalation, especially by exposure to dust (Clark, 2011), triggering irritations to the skin and eyes (Christie, 2007). The workers who produce or handle reactive dyes may have contact dermatitis, allergic conjunctivitis, rhinitis, occupational asthma or other allergic reactions (Hunger, 2003). The latter are the result of the formation of a conjugate between human serum albumin and the reactive dye, which acts as an antigen (Christie, 2007) producing immunoglobulin E (IgE) antibodies, which combine with histamine (Hunger, 2003).

The genotoxicity of textile dyes (Thakur, 2006) is the greatest potential long-term hazard to human health (Christie, 2007). Demonstrating, for example, the strong genotoxic effects of textile dyes, Tiwari, Tripathi, and Gaur (2016)) point to the existence of studies made in *Allium cepa* root cells exhibiting chromosomal aberrations.

Some dyes reveal mutagenic potentiality (Hunger, 2003). One of them is Azure-B, widely used in the textile industry, which is able to intercalate with the helical structure of DNA (Christie, 2007; Haq & Raj, 2018) and duplex RNA (Khan & Kumar, 2016), as it can be partitioned to the lipid membrane of the cells (Li, Zhang, Tang, Zhang, & Mao, 2014). It is noteworthy that this dye can reveal cytotoxic effects by acting as a notable reversible inhibitor of monoamine oxidase A (MAO-A), according to *in vitro* tests (Petzer, Harvey, Wegener, & Petzer, 2012), which is an intracellular enzyme of the nervous system central (Factor and Weiner, 2007) that plays an important role in human behavior (Di Giovanni et al., 2008). Its potential for enzyme inhibition also concerns glutathione reductase (Paul & Kumar, 2013) which plays an essential role in cellular redox homeostasis (Couto, Wood, & Barber, 2016).

The Disperse Red 1 dye is also used by the textile industry and exhibits mutagenic potential (Chequer et al., 2009). When used *in vitro* in human lymphocyte and human hepatoma (HepG2) cells, hepatocyte imitative cells *in vivo* (Will, McDuffie, Jeffy, & Olaharski, 2016), it is capable of increasing the frequency of micronuclei (Fernandes, Bustos-Obregon, & Salvadori, 2015), which are indicative of mutagenic activity at the chromosome level (Duarte & Rai, 2016). The assays performed on *Salmonella* spp. (Vacchi et al., 2016) suggests that it is responsible for the formation of DNA adducts (Chequer et al., 2011) which, in the case of humans, constitute a mutagenic event that is key to the characterization of cancer (Hsu & Stedeford, 2010). In its turn, the Disperse Orange 1 dye exhibits similar mutagenic behavior (Chequer et al., 2009) inducing DNA damage, found in the *Salmonella* spp. assays, involving base pair substitution and frameshift mutations that alter the reading frame. In addition, it has a cytotoxic effect, with apoptosis, in contact with HepG2 cells (Ferraz, Grando, & Oliveira, 2011).

The carcinogenesis comprises multiple stages favored initially by mutagenic factors (Hunger, 2003). The textile dyes may offer carcinogenicity, especially those of the azo and nitro type, and its effects manifest themselves over time (Mondal, Purkait, & De, 2018).

The Sudan I dye (Solvent Yellow 14) is the family of azo-lipophilic compounds widely used in various industrial segments, including textiles (Petrakis, Cagliani, Tarantilis, Polissiou, & Consonni, 2017). Although illegal, its use is still recurrent in foods, such as paprika (Di Anibal, Marsal, Callao, & Ruisánchez, 2012). Once present in the bodies of animals or humans, it is enzymatically transformed, through the action of the intestinal flora, into carcinogenic aromatic amines (Piątkowska, Jedziniak, Olejnik, Żmudzki, & Posyniak, 2018). Especially in the case of azo dyes, carcinogenicity can be produced by both the dye itself and its own metabolized compounds (Christie, 2007). In rats, the presence of Sudan I dye is confirmed by neoplastic liver nodules (National Toxicology Program, 1982).

The Basic Red 9 dye, used in the textile, leather, paper and ink industries (Duman, Tunc, & Polat, 2015), offers carcinogenic potential in humans (Lacasse & Baumann, 2012) and high environmental toxicity (Foguel et al., 2015). It breaks down, under anaerobic conditions, into carcinogenic aromatic amines and their disposal in water bodies has the potential for allergic dermatitis, skin irritation, mutations and cancer itself (Sivarajasekar & Baskar, 2014). The latter, according to the tests performed on rats, may comprise local sarcomas and tumors in the liver, bladder (Pohanish, 2017), mammary glands and hematopoietic system (National Toxicology Program, 1986).

The Crystal Violet dye, a member of the cationic triphenylmethane group, shows a very intense color (Ali, Shehata, & Ramadan, 2016) and is responsible for mitotic poisoning, which is associated with abnormal accumulation of metaphases (Mani & Bharagava, 2016) as well as the *in vitro* clastogenic effects observed in Chinese hamster ovules (Azmi, Sani, & Banerjee, 1998), which induce chromosomal damage (Newman, 2015). This powerful carcinogenic agent (Bharagava, Mani, Mulla, & Saratale, 2018) promotes fish tumors (Mani & Bharagava, 2016), as well as hepatocarcinoma, reticular cell sarcoma in various organs, such as the vagina, uterus, ovary and bladder (Littlefield, Blackwell, Hewitt, & Gaylor, 1985), hardened gland adenoma and ovarian atrophy in rats. In humans, it is capable of generating chemical cystitis, irritation of the skin and digestive tract, respiratory and renal failure, among others (Mani & Bharagava, 2016).

Despite the prohibited agreements (Christie, 2007), of the 4000 dyes that have been investigated for their toxicity, more than 100 of them with potential for the formation of carcinogenic amines are still available on the market (Lacasse & Baumann, 2012). In several regions of the globe, such as in India, export demands associated with cheap labor determine the existence of small-scale textile factories that clandestinely release toxic dyes into water bodies (Asthana & Shukla, 2014).

In face of this complex context, which presents deep unfolding to the ecosystems and to the human being, there are possibilities represented by bioremediation technologies which are directly related to the commitment of sustainable development. In other words, the bioremediation technologies foment the economic growth in harmony with the environment and ensure the quality of life.

Phytoremediation

The phytoremediation can be understood as the ability of plants to degrade, extract, transform and detoxify (Bharathiraja, Jayamuthunagai, Praveenkumar, & Iyyappan, 2018), through their enzymes and associated microflora, the contaminants of the air, soil, sediments, surface water and groundwater (Tahir, Sohail, & Khan, 2017).

This remarkable phytoremediative capacity is related above all to the genetic adaptation that has allowed the plants to become, over time, autotrophic bioreactors capable of alleviating environmental stress (Bharathiraja et al., 2018; Tahir, Yasmin, & Khan, 2016). It is undoubtedly a true green technology provided by nature itself (Rane, Khandare, Watharkar, & Govindwar, 2017).

The tables S1, S2, S3 and S4 (in Supplementary materials) provide examples of plants that are able to promote phytoremediation of textile dyes employing the principles of rhizofiltration, phytoextraction or phytoaccumulation, phytodegradation and rhizodegradation.The hydroponics systems, in turn, do not use the soil (Schwitzguébel et al., 2011) and considerably help plant metabolism for the extraction and degradation of textile dyes (Muthusamy, Govindaraj, & Rajendran, 2018). The examples are shown in table S5. They include *Nasturtium officinale* (Brassicaceae), which biodegrades Basic Red 46 (Torbati, Khataee, & Movafeghi, 2014), as well as *Pennisetum purpureum* (Poaceae), which discolour Poly R-478 (Paquin, Sun, Tang, & Li, 2006).

The role of plants in phytoremediation, as well as their metabolism and tolerance to toxicity, can best be understood through contributions from the pilot scale studies of hairy root, callus and cell suspension cultures (Rane et al., 2017).

The hairy root cultures provide the basis for the production of secondary metabolites and can be used for the research of different plant species with the ability to tolerate, accumulate or remove environmental pollutants (Lokhande, Kudale, Nikalje, Desai, & Suprasanna, 2015). Since hairy root cultures can be considered as true phytochemical factories (Ono & Tian, 2011), the production of secondary metabolites becomes stably increased using *Agrobacterium rhizogenes* (Tu, 2017) plasmid Ri. It is the biotransformation made not only for the synthesis of compounds of industrial and pharmaceutical interests, but also for the degradation and bioremediation of toxic substances (Ono & Tian, 2011).

The biotransformation of hairy root cultures of *Tagetes patula* (Asteraceae), mediated by *A. rhizogenes*, stimulates the enzymatic production of lignin peroxidases, Mn peroxidases, tyrosinases and azoredutases that allow the biodegradation of the Red 198 textile dye. As can be seen in table S5, the hairy root cultures of *T. patula* also demonstrate the possibility of biodegradation for textile dyes Golden Yellow HER, Methyl Orange, Orange M2RL, Navy Blue HE2R and Reactive Red M5B (Patil, Desai, Govindwar, Jadhav, & Bapat, 2009). The other example is in the phytodegradation of Reactive Green 19A-HE4BD textile dye by hairy root cultures of *Sesuvium portulacastrum* (Aizoaceae) biotransformed by *A. rhizogenes* (Lokhande et al., 2015).

The callus and cell suspension cultures are very useful tools when speaking about phytoremediation (Macek, Pavlikova, & Mackova, 2004). In this sense, according to table S5, the callus cultures of *Tecoma stans* (Bignoniaceae), protected by calcium alginate entrapment, biodegrade the Malachite Green textile dye via the enzyme peroxidase (Rani & Abraham, 2016). The cell suspension cultures of *Blumea malcolmii* (Asteraceae) biodegrade a wide variety of textile dyes belonging to distinct structural groups, such as Malachite Green, Remazol Red, Red HE8B, Methyl Orange, Red Reactive 2, Red HE 7B, Golden Yellow HER and Scarlet GDR (Kagalkar, Jadhav, Bapat, & Govindwar, 2011).

One of the major challenges of phytotechnology involves the transposition of laboratory-controlled experiments, such as hairy root, callus or cell suspension cultures, to a much larger extent, where systemic behavior and efficiency can be highly variable and dependent on environmental factors (Rane et al., 2017). Despite this, large-scale actions, such as ponds or phytoreactors, have been made based on the synergism between plants and rhizospheric microorganisms (Khandare & Govindwar, 2015; Tahir et al., 2016; Watharkar et al., 2018). The main advantage of this consortium is that the diverse cultures of plants, bacteria and fungi obtain a more efficient biodegradation for textile dyes (Chandanshive et al., 2017). According to table S6, the biodegradations of the dyes Brown 5R and Congo Red illustrate the consortia formed respectively between Convolvulaceas *Ipomea hederifolia* and *Ipomea aquatica* (Rane et al., 2016) and *Typha angustifolia* (Typhaceae) and *Paspalum scrobiculatum* (Poaceae) (Chandanshive et al., 2017).

The phytoremediation of textile dyes is environmentally correct, economically efficient, aesthetically pleasing and, being a large autotrophic biomass system, requires little provision of nutrients (Kabra, Khandare, Kurade, & Govindwar, 2011; Kabra, Khandare, Waghmode, & Govindwar, 2011; Rane et al., 2015). However, it depends on the soil properties and the plant's own absorption and degradation capacity (Tahir et al., 2017).

Bioremediation by microorganisms

Bioremediation by microorganisms provides the removal, reduction or destruction of harmful compounds by microorganisms such as bacteria, algae, filamentous fungi and yeasts in soil, water, sludge, waste or effluent (Das & Dash, 2017; Saxena, 2015). One of the strategies involved is *in situ* bioremediation by means of biostimulation or bioaugmentation consisting of, respectively, the introduction of nutrients to favor local microorganisms or the inoculation of exogenous microorganisms (Kasai, 2011; Saxena, 2015). The other strategies are *ex situ* bioremediation by composting and landfarming, by bioreactors, by hybrid crops and by the genetic improvement of lineages (Das & Dash, 2014).

From environmental samples, such as the textile effluents themselves, it is possible to select and isolate wild microorganisms that are capable of biodegrading dyes (Kandelbauer & Guebitz, 2005). Another possibility involves identifying, isolating, cloning and transferring genes that encode degradative enzymes that increase the biodegradation capacity of native species. The so-called super-degrading microorganisms (Pereira & Alves, 2011)

appear as hybrid or engineered strains (Kandelbauer & Guebitz, 2005). An example is in the bacterium *Escherichia coli* that starts to load the azoredutase enzyme gene, capable of biodegrading azo textile dyes, transferred from the wild variety *Pseudomonas luteola* (Chang, Kuo, Chao, Ho, & Lin, 2000). The gene encoding the CotA lacase enzyme, from *Bacillus subtilis* (Gupta, Garg, Capalash, Gupta, & Sharma, 2015), can also be transferred to *E. coli* allowing it to discolor the azo and anthraquinone dyestuffs (Pereira & Alves, 2011).

The bacteria can biodegrade textile dyes under aerobic or anaerobic conditions (Telke, Kadam, & Govindwar, 2015). In the case of azo textile dyes, bacterial biodegradation comprises the reductive cleavage of azo bonds (—N=N—) by the azoreductase enzymes (Saratale, Saratale, Chang, & Govindwar, 2011). This biodegradation also counts on the participation of other enzymes, such as peroxidase, laccase, tyrosinase, NADH-DCIP reductase and MG reductase (Telke et al., 2015).

The bacterial biomasses are good biosorbent materials for the bioremediation of textile dyes (Tan, Li, Lu, & Chen, 2010), which serve as sources of carbon or nitrogen (Roy et al., 2018). The mechanisms involved in the interaction between living and dead cell biomass and textile dyes are complex. They include, for example, adsorption, where the interaction takes place between the molecules of the textile dyes and the chemical groups, possessing electric charges, which are present on the bacterial cell surface (Srinivasan & Viraraghavan, 2010).

The main disadvantages of bacterial biosorption are the adsorption capacity and the final disposal of the biomass, as well as the pre-treatment and the characteristics of the dyes and their effluents (Srinivasan & Viraraghavan, 2010). However, there are the low operating costs and the possibility of using hybrid adsorbent systems with great efficiency for textile dyes (Wawrzkiewicz, Bartczak, & Jesionowski, 2017).

As shown in table S7, the bacterial biodegradation of textile dyes can be carried out using pure cultures (Saratale et al., 2011). For example, after the bioaugmentation in activated sludge, *Aeromonas punctata* and *Shewanella putrefaciens* can each degrade azo textile dyes Acid Red 88, Reactive Black 5, Direct Red 81 and Disperse Orange 3 (Khalid, Arshad, & Crowley, 2008). The azo dye Navitan Fast Blue S5R, of great importance to the textile and tannery industries, can in turn be degraded aerobically, in the presence of glucose, by *Pseudomonas aeruginosa* (Nachiyar & Rajkumar, 2003). Due to sucrose and yeast extract, *Aeromonas hydrophila* can degrade the Crystal Violet dye by laccase enzymes and, especially, lignin peroxidase (Bharagava et al., 2018). In addition, since the bacteria can be biogenerators of gold and silver nanoparticles (Anthony, Murugan, Jeyaraj, & Gurunathan, 2013), the biodegradation of the Direct Black 22 and Reactive Yellow 186 dyes can be performed by gold nanoparticles, synthesized from cultures of *Acinetobacter* sp. SW30, together with sodium borohydride ($NaHB_4$). These nanoparticles, obtained by biogenesis, act under anaerobic conditions as an electron transfer system between the hydride ions donors and the textile dye receptors, which enables their rapid degradation (Wadhwani, Shedbalkar, Nadhe, Singh, & Chopade, 2018).

The co-cultures, or hybrid bacterial cultures, can lead to a higher level of biodegradation, especially because textile dye molecules are attacked in different positions (Chandra, 2016). The sulfated textile dye Green HE4BD is degraded by *Proteus vulgaris* and *Micrococcus glutamicus* at a much higher level (Table S8). There is the formation of smaller molecular weight intermediates (naphthalene moieties) by the consecutive action of oxidoreductive enzymes present in the co-culture (Saratale, Saratale, Chang, & Govindwar, 2010). Similarly, the consortium between *Ochrobactrum* sp., *Pseudomonas aeruginosa* and *Providencia vermicola*, treating a textile effluent of diverse composition and red violet color, achieves better results of biodegradation. This is possible due to the action of oxidoreductase enzymatic mechanism of laccase, NADH 2,6-dichlorophenolindophenol (NADH-DCIP) reductase and azoreductase activity (Vijayalakshmidevi & Muthukumar, 2015), as occurs between *Bacillus* sp. and *Staphylococcus epidermidis* for the biodegradation of Congo Red dye through oxidase and catalase enzymatic activities (Ayed, Achour, Khelifi, Cheref, & Bakhrouf, 2010).

The effectiveness of co-cultures (Table S8) is also expressed in the communities of sulfate-reducing bacteria present in microbial fuel cells. In them, the biodegradation of the Red Acid 114 dye, the removal of sulfates and even the obtaining of bioelectricity can occur (Miran, Jang, Nawaz, Shahzad, & Lee, 2018). The reason involved is that the textile industry effluent, containing dyes, can be used as a substrate for the microbial fuel cell and its biodegradation is capable of producing electrical energy through organic oxidation (Dutta & Sahai, 2018).

The bacterial biodegradation, especially that of azo dyes, generally exhibits a high degree of removal and mineralization of the harmful compounds, as well as being economically viable, producing little sludge and being faster than that performed by fungi. However, it is necessary to constantly monitor the effective toxicity of the obtained compounds and to control the various physico-chemical parameters involved, such as the agitation and oxygen levels, the type and concentration of substrate offered, temperature, pH and concentration of the textile dye (Saratale et al., 2011).

Although its microbiological composition varies according to biotic and abiotic factors, activated sludge is an ecosystem composed of bacteria, fungi and cyanobacteria, as well as protozoa and metazoa microfauna (Wanner, 1998). Its use in bioreactors is one of the processes most used by the textile industry (Dias, Sampaio, & Bezerra, 2007).

A good example of the functioning of bioreactors involves the use of activated sludge from a textile effluent treatment plant for the biodegradation of the Reactive Black 5 textile dye. After the development of adequate biomass, with glucose as a co-substrate, the dye solution ($100\,mg.L^{-1}$) was fed into a fermenter vessel of 4,2 L capacity. Under pH = 6,0 and 30 °C, the content was stirred at 10 rpm continuously throughout 2 days. The effluent was then collected into the aerobic reactor. The pH was maintained between 6.0 and 7.0 and the aerobic experiment was realized during 2 days at room temperature. The anaerobic phase achieved more than 90% decolorization due to the reduction of azo bonds releasing aromatic amines to be partially removed by the aerobic treatment (Mohanty, Dafale, & Rao, 2006).

The anaerobic and aerobic phase bioreactor used for the treatment of textile effluents can be improved. Since

various molecular and functional groups, arranged along polymeric chains, can form different interactions with the dye molecules (Panic et al., 2013), the use of polymers can make their absorption viable. This increases the degradation efficiency of the anaerobic and aerobic treatment steps, as well as assists in reducing the toxicity to biomass (Tomei, Angelucci, & Daugulis, 2016).

Although useful for the removal of various textile dyes, activated sludge bioreactors cannot be used alone to fully meet the biodegradation needs of these compounds (Christie, 2007). Moreover, they produce large amounts of sludge, are very sensitive to the composition of the effluent, offer operational difficulties and exhibit little efficiency to the reactive textile dyes (Dias et al., 2007).

The extremophiles are organisms that develop in conditions considered inhospitable, according to most eukaryotes, because of the high physical-chemical stress involved (Plath, Tobler, & Riesch, 2015). A good part of its representatives is the result of the Archaea domain, but there are also those belonging to the Bacteria and Eukarya domains (Horikoshi & Bull, 2011). The extremophiles and their extremozymes are one of the most attractive bioremediation tools, especially in aggressive industrial processes, such as those in the textile industry, which comprise stressful conditions of pH, temperature, salinity and toxicity (Amoozegar, Mehrshad, & Akhoondi, 2015).

In textile dyeing, various salts are used providing significant difficulties for biodegradable microbial communities. The reason is that high saline concentrations can cause plasmolysis (Meng, Liu, Zhou, Fu, & Wang, 2012), that is, lead the cytoplasm to lose water and contract, such that the plasma membrane gradually separates from the cell wall (King et al., 2001). As an effective alternative, halotolerant and halophile microorganisms, or yet, extremophiles living in environments with high salt concentrations (Babu, Chandel, & Singh, 2015), can be used to biodegrade azo textile dyes in media with high salt content (Meng et al., 2012). In this sense, even in the presence of the metallic cations Mg^{2+}, Ca^{2+} and Pb^{2+}, the bacterial community formed by *Bacillus*, *Sedimentibacter*, *Pseudomonas*, *Clostridiales* and *Streptomyces* exhibits good performance in the discoloration, under high concentrations, of the Reactive Brilliant Red X-3B dye and the complete discoloration occurs under high concentrations of sodium chloride (NaCl) (Tan, Qu, Zhou, Ma, & Li, 2009). The bacterium *Shewanella aquimarina*, in turn, discolored the Acid Red 27 dye in NaCl containing medium (Meng et al., 2012). In addition, *Schefersomyces spartinae* yeast can biodegrade, aerobically and with high salinity, the Acid Scarlet 3R dye through azo reduction, deamination and desulfurization (Tan, He, Song, Fu, & Shi, 2016). Another yeast, *Pichia occidentalis*, can also reductively discolor, deminer and mineralize, aerobically and with high salinity, the Acid Red B dye (Song, Shao, Ning, & Tan, 2017).

The thermophiles, in turn, can withstand and develop at temperatures ranging from 45 °C to 122 °C and are an important alternative for the discoloration of azo textile dyes (Table S10) (Amoozegar et al., 2015). For example, bacterial strains identified by the 16S rRNA sequencing as homologous to the species *Anoxybacillus pushchinoensis*, *Anoxybacillus kamchatkensis* and *Anoxybacillus flavithermus*, can discolor, at a temperature of 65 °C, in a bench scale bioreactor, the Reactive Black 5 dye with a two-fold higher efficiency rate compared to bottle cultures (Deive et al., 2010). The bacterium *Anoxybacillus rupiensis*, in turn, is able to decolorize the dark red textile effluent, with pH 10,5, obtained from a dyeing plant in Aurangabad, India, by means of flasks incubated at 60 °C (Gursahani & Gupta, 2011).

However, those organisms that can live stably in two or more extreme environments are considered as polyextremophiles (Table S10) (Cockell, 2015). An example comprises the bacterial strains, identified by the 16S rDNA sequencing, as belonging to the genus *Bacillus* sp., especially the bacterium *Bacillus pallidus*, that are able to degrade the effluent in a bioreactor of a textile drain with temperatures of 60–65 °C and pH between 9,3 and 10 (Paar et al., 2001).

The extremophiles and polyextremophiles become an important source of research, especially for bioremediation, in the textile industry. Given the difficulties of their production, according to conventional culture protocols, their genes of interest can be identified, isolated, cloned and genetically transferred to be expressed by organisms for which the methods of production are already well established and standardized (Christie, 2007).

The fungi produce a large variety of intra and extracellular enzymes with high biodegradability (Saratale et al., 2011). The biotransformation of compounds, residues and wastewater by fungi characterizes mycoremediation (Singh, 2006), that is, the fungal biomass is used to biodegrade industrial pollutants in soil and water (Cotter, 2014).

The fungal treatment of textile dyes is an economical and effective alternative to discoloration (Singh, 2006). The main mechanisms involved are biosorption, bioaccumulation and biodegradation (Kaushik & Malik, 2015).

The fungal biosorption, performed by living or dead cells, plays an important role in the discoloration of textile dyes (Srinivasan & Viraraghavan, 2010). The phosphate and carboxyl groups, which originate from glucuronic acid, are probably responsible for the negative charges, whereas the amino groups, originating from chitosan, create positive charges on the fungal cell wall (Naja & Volesky, 2011). These functional groups allow the binding or biosorption of the textile dye molecules to the surface of the fungal cell wall in a process that can be complete in a few hours (Kaushik & Malik, 2015), in such a way that the interactions involved are essentially of physical-chemical order (Srinivasan & Viraraghavan, 2010). In this sense, table S11 shows that the biomasses of dead cells of *Aspergillus niger* and *Trichoderma* sp., at pH = 2, are able to bioadsorver the Orange G dye (Sivasamy & Sundarabal, 2011). The dead cell biomass of *Aspergillus niger*, obtained by autoclaving at 121 °C, can also bioabsorber, at the pH of 3 to 7, the Direct Blue 199 dye which contains, in its structure, copper cations (Xiong, Meng, & Zheng, 2010). In particular, autoclaving can break the fungal structure, expose binding sites and maximize biosorption, or yet, it increases the interaction between fungal cell walls and textile dyes (Sen, Raut, Bandyopadhyay, & Raut, 2016). Other examples can also be found with the biosorption of Methyl Orange, by *Aspergillus flavus* (Takey et al., 2014), and Methylene Blue, by *Aspergillus fumigatus* (Table S11) (Kabbout & Taha, 2014).

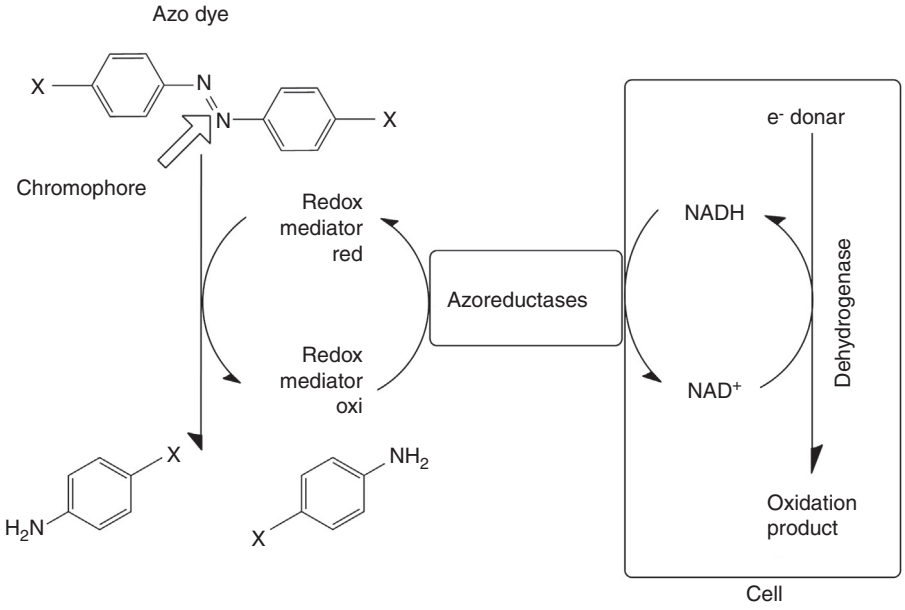

Figure 1 Proposed mechanism of azoreductases for azo dyes degradation (adapted from Khan, Bhawana, & Fulekar, 2013).

The biodegradation, in turn, is the main mechanism used by fungi for the discoloration of textile dyes (Sen et al., 2016). It is mediated by enzymes (Kaushik & Malik, 2015) which mainly comprise azoredutases, lignin peroxidase, Mn peroxidase and laccases (Saratale et al., 2011). The white rot fungi, for example, are able to biodegrade the textile dyes (Kandelbauer & Guebitz, 2005) by means of peroxidases and laccases. For this, they can be used in different configurations of bioreactors, such as fixed film, fixed bed, fluidized bed and rotating biological contactors (Doble & Kumar, 2005). Despite showing good results (Table S12), such as occurs in the biodegradation of Reactive Red 2 and Reactive Blue 4 dyes, by *Trametes versicolor* (Nilsson, Möller, Mattiasson, Rubindamayugi, & Welander, 2006), the bioreactors with white rot fungi may exhibit problems. These are related, in particular, to excessive fungal growth, capable of clogging the continuous reactor, to bacterial contamination inhibiting fungal enzymatic activity (Sen et al., 2016) and to the long hydraulic retention time for discoloration of the textile dye (Saratale et al., 2011). In addition to fungal bioreactors of white rot, it is observed that the consortium between the Convolvulaceae, already mentioned, *I. hederifolia* and *Cladosporium cladosporioides*, an endophytic fungus, allows the construction of a column rizoreator, considered as a bioreactor of upstream flow, to efficiently biodegrade, via tyrosinase, peroxidase, laccase and riboflavin reductase, the Navy Blue HE2R dye (Patil et al., 2016).

The endophytic fungi deserve to be mentioned because they live in the tissues of plants, without causing visible symptoms, and provide their protection, especially against pathogens and herbivores (Sim, Chen, & Ting, 2019). Another important aspect is that they are potential textile dyes degraders (Krishnamurthy & Naik, 2017). In this case, endophytic biodegradation occurs because of the role played, in particular, by the laccase enzyme. Other enzymes, in turn, exhibit undeniable contribution, such as lignin peroxidase, Mn peroxidase, reductase and tyrosinase (Sim et al., 2019).

The table S13, as shown below, provides examples of endophytic fungi and their bioremediation potential for textile dyes.

The mycoremediation is therefore a natural, safe and low-cost process (Jain, Yadav, Nigam, & Sharma, 2017). The isolation and sequencing of gene coding for biodegradable enzymes is very promising, as well as the viability of bioengineered fungi (Singh, 2006). Despite this, it still faces the influence of restrictive factors, namely nutrients, pH, temperature, oxygen levels and the very nature of textile effluents (Sen et al., 2016).

Due to the extensive and undeniable importance of microorganisms for the dyes and their effluents, it is essential to elucidate the essential role of the main related enzymes, such as azoredutases, laccases and peroxidases.

Bioremediation by enzymes

The ideal objective of biodegradation is the complete degradation of a compound, also called mineralization, which occurs through its decomposition into water, carbon dioxide and/or an inorganic end product (Kaushik & Malik, 2015). In this search, the metabolic potential of the microorganisms can be used to degrade or transform organic contaminants to produce less aggressive compounds to the environment (Qu, Hong, & Zhao, 2018).

The azoredutases are key enzymes for the discoloration of textile dyes. They promote the reductive cleavage of the azo bonds and release the aromatic amines that will be degraded in CO_2 and H_2O (Imran et al., 2015).

It is believed that this reductive cleavage reaction can occur through direct mechanisms, where the azoredutase enzymes physically interact with the textile dye molecules transferring the electrons (Imran et al., 2015), or through indirect mechanisms, where the cooperation of coenzymes is required, such as nicotinamide adenine dinucleotide (NAD^+), nicotinamide adenine dinucleotide

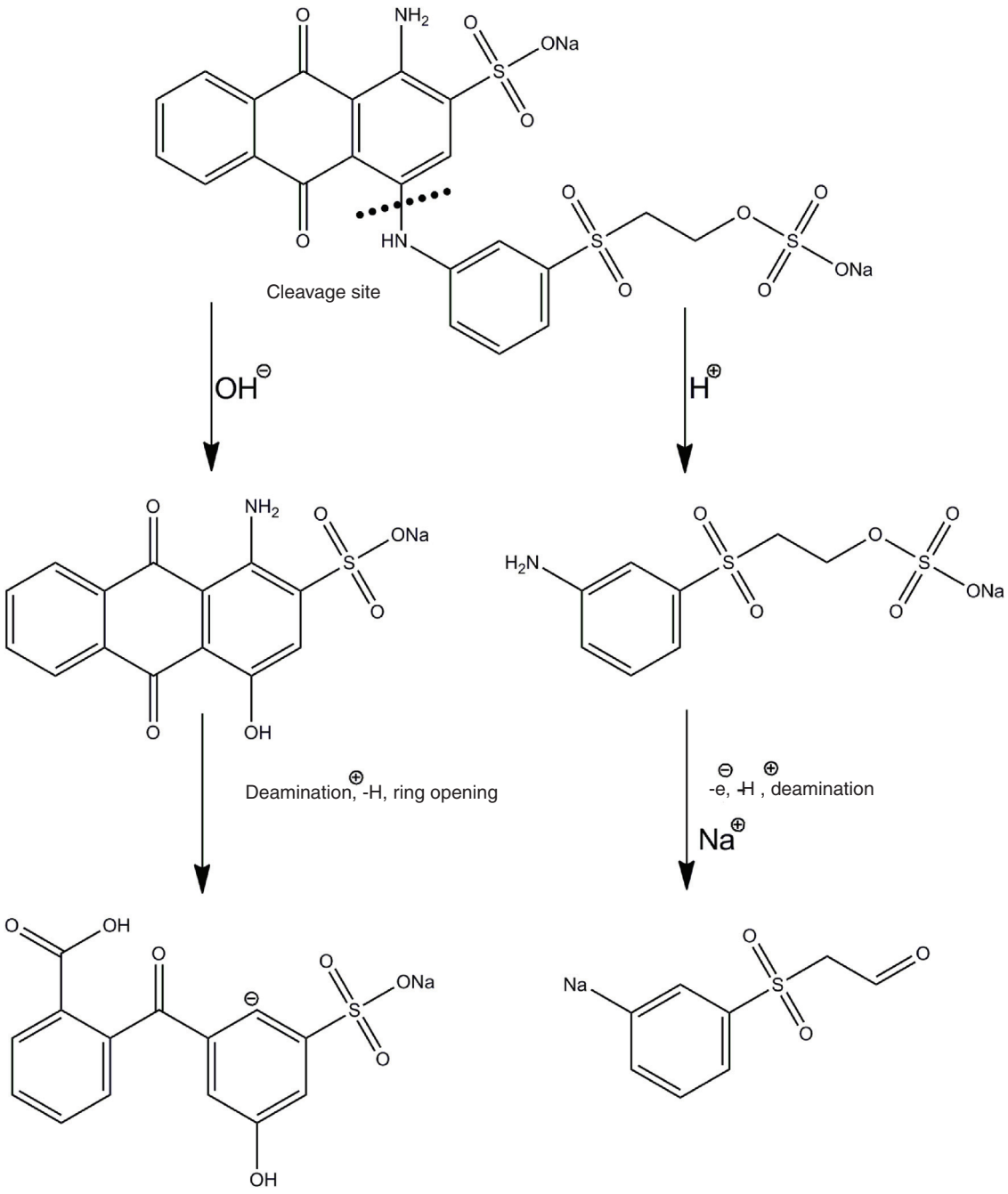

Figure 2 Degradation products of Remazol Brilliant Blue R observed by Osma et al. (2010) using a laccase from *Trametes pubescens*.

phosphate ($NADP^+$) and flavin and adenine dinucleotide (FAD) (Guo, Kang, Wang, & Yang, 2010; Sen et al., 2016). The azoredutase enzymes then transfer the electrons to these coenzymes which in turn carry them to the molecules of textile dyes promoting the breakdown of their azo bonds. Therefore, coenzymes are not a part of the enzymatic structure, but are intermediary carriers (Mcguire & Beerman, 2006; Vanmeter, Vanmeter, & Hubert, 2013), or redox mediators (Telke et al., 2015), which accelerate the rate of the electron transfer process responsible for reductive cleavage (Guo et al., 2010; Rather, Akhter, & Hassan, 2018). In their oxidized forms NAD^+, $NADP^+$ and FAD, coenzymes receive the electrons from the azoredutase enzymes and are reduced to NADH, NADPH and FADH. These reduced forms are then oxidized when they donate these same electrons to the molecules of the textile dyes and return to their original NAD^+, $NADP^+$ and FAD forms (Fig. 1). This indirect mechanism, however, requires the absence of oxygen (O_2), since it can compete with the molecules of the textile dyes, preferentially oxidizing the NADH, NADPH and FADH factors, which inhibits the process of electron transfer to the reductive cleavage (Sen et al., 2016).

The laccase enzymes are also called phenoloxidases (Saratale et al., 2011). In the catalytic center (Baldrian,

2009) of each of them there are four copper atoms (Singh, 2006) distributed in three different sites, with type I copper conferring its intense blue color (Chandra, 2016). The laccases oxidize various phenols and their derivatives, such as ethers, aromatic amines and other non-phenolic compounds (Boulatov, 2006). The substrates oxidized by the laccases lose electrons which are transferred to O_2 (Kruus, Niku-Paavola, & Viikari, 2001) and the enzymatic electron transport system occurs from type I copper to types II and III (Chandra, 2016). The O_2 is then reduced to H_2O (Hofrichter & Ullrich, 2011) at the copper sites types II and III (Chandra, 2016), while the substrates usually give free radicals (Kruus et al., 2001) which may participate in other reactions, such as polymerization and hydration (Sen et al., 2016). For these reasons, the laccases can be used for the detoxification of several industrial effluents, among which the textile (Chandra, 2016) which produces high levels of polycyclic aromatic hydrocarbons (Ning et al., 2015) and sulfonated aromatic amines (Arslan-Alaton & Olmez-Hanci, 2010). Osma, Toca-Herrera, and Rodríguez-Couto (2010)) propose of transformation pathway of the antraquinonic dye Remazol Brilliant Blue R by immobilized laccase obtained from *Trametes pubescens*. The treatment provided subproducts with the chromophore group broken and molecules with less molecular weight, being expected to be less environmentally hazardous than parent dye molecule (Fig. 2).

The peroxidases, in turn, exhibit hemiproteins (El Enshasy et al., 2017), that is, they have a heme group, which is a complex between an iron (Fe^{3+}) cation and protoporphyrin IX (Husain, 2010), responsible by a myriad of functions, such as electron transfer and redox catalysis (Battistuzzi, Bellei, Bortolotti, & Sola, 2010). These enzymes, unlike laccases, use hydrogen peroxide (H_2O_2) as an electron-terminal receptor (Husain, 2010) which is then reduced to allow the oxidation of a wide variety of organic and inorganic substrates (Battistuzzi et al., 2010). McMullan et al. (2001) propose a pathway of peroxidases to degradation of sulfonated azo dyes, and demonstrate that the peroxidase activity promote an initial cleavage sites in the chromophore group (Fig. 3). Due to their low specificity of substrates, peroxidases find application in multiple areas, including being suitable for the treatment of effluents containing textile dyes (Mendes, Robalo, & Martins, 2015).

The most commonly studied peroxidases used for the discoloration of textile dyes comprise lignin peroxidase and Mn peroxidase (Husain, 2010). The lignin peroxidase has a high redox potential (Chandra, 2016) catalyzing, in the presence of H_2O_2, the oxidation of non-phenolic aromatic rings in lignin (Bajpai, Anand, & Bajpai, 2006) and many phenolic compounds for the generation of radical cations (Wertz, Deleu, Coppée, & Richel, 2018). These, in turn, are unstable molecules that can trigger, for example, the demethylation, the opening of aromatic rings and the dimerization of phenols (Rahi, Rahi, Pandey, & Rajak, 2009). This allows lignin peroxidase to mineralize recalcitrant aromatic compounds, including azo textile dyes (Imran et al., 2015). On the other hand, Mn peroxidase is oxidized by H_2O_2 (Yadav, Singh, Yadava, & Yadav, 2015) to form an intermediate compound which, in turn, oxidizes the Mn^{2+} cation to Mn^{3+} (Husain, 2010). The Mn^{3+} cation interacts with organic acids to form a complex capable of oxidizing various substrates, such as

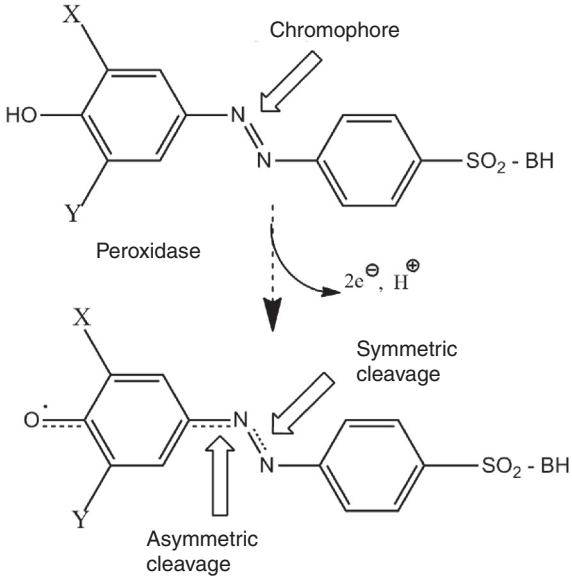

Figure 3 Activity of peroxidase to degradation of sulfonated azo dye proposed by McMullan et al. (2001). The peroxidase activity provides two cleavage sites in —N═N— chromophore group, that can produce a symmetric or asymmetric cut in the azo dye molecule.

lignin itself (Rahi et al., 2009), phenols (Yadav et al., 2015) and textile dyes (Husain, 2010).

One of the main disadvantages of this form of bioremediation is that the enzymes become susceptible to inactivation through the action of inhibitors found in the severe conditions of the polluted environment to be treated (Hochstrat et al., 2015). However, enzymatic bioremediation appears to be quite attractive and promising, especially as it is an effective alternative to conventional physicochemical treatments, to generate significantly reactive free radicals (Saratale et al., 2011) and to remove highly diluted or particularly recalcitrant pollutants (Illanes, 2008). Advances in molecular biology and genetic engineering, in turn, can provide important contributions through the expression of genes of interest in viable host microorganisms to obtain, with high productivity and low cost, more active and versatile enzymes able to treat effluents from the textile industry more efficiently (Sen et al., 2016). In addition, nanostructures, such as carbon nanotubes, because of their large specific surface area and their excellent mechanical and chemical properties, can serve as supports for the immobilization of proteins and enzymes, facilitating their operation and increasing their stability and amplitude of its pH (Oliveira, da Luz, Kasuya, Ladeira, & Junior, 2018).

Conclusions

The textile industries produce effluents with high levels of toxic and recalcitrant compounds, such as dyes, which generate disastrous effects on the environment and the human being. In order to mitigate or even eliminate the harmful consequences involved, plant, bacteria, extremophiles and fungi biomasses can be used to discolor, transform or mineralize the textile dyes. Despite the excellent results, all bioremediations offer

limitations to a greater or lesser degree. However, molecular biology, genetic engineering and nanotechnology, coupled with academic-scientific research, can overcome these constraints by focusing more on the more efficient and stable engineered enzyme-producing organisms. Given this, the environmental biotechnology is ethically and efficiently placed as the great tool to promote sustainable development to the present and to the future.

Conflicts of interest

The authors declare no conflicts of interest.

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.biori.2019.09.001.

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