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Case Report

# Sequential batch reactors for aerobic and anaerobic dye removal: A mini-review

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# ABSTRACT

One of the most challenging aspects of environmental conservation is the treatment of dye wastewater. Thus, this mini-review discussed the issues and major advances in the application and performance assessment of the aerobic, anaerobic, anaerobic-aerobic, aerobic-anaerobic, and anoxic-aerobic REACT-operated SBR systems with regards to bio-decolorization and COD removal rates. In SBRs run in aerobic modes, it is possible to cultivate aerobic granular sludge for color removal. To be precise, SBR has a higher COD removal efficiency with a lower color removal efficiency. Under anaerobic conditions, lesser COD removal was observed for several dyes studied in this chapter. In an alternating anaerobic-aerobic SBR system, color removal tends to occur during the anaerobic phase, while the aerobic phase is required to further reduce the effluent COD concentration. However, molecular oxygen drastically reduced color removal in SBR during dye wastewater treatment. This chapter discusses the aerobic-SBR treatment process for dye removal. The discussion focused on dye wastewater treatment using aerobic granules, granular activated carbon, adsorbents, biocarrier white rot fungi, varying dye mixtures, dye concentrations and SBR operational parameters. Adsorbents, membranes, biocarriers, exported microbial cultures, and various operational conditions have also been used in sequential anaerobic/aerobic batch reactors to enhance system performance. This chapter assessed the different treatment mechanisms and dynamics and concluded that combining two treatment methods significantly yields better color, DOC, and BOD<sub>5</sub> removal than a single biological or chemical treatment.

1. Introduction

The textile industry is responsible for more than fifty-four per cent of the dye effluents discharged into the environment. Annually, about 2.8  $\times$  10<sup>5</sup> tons of textile wastewater containing a large number of different synthetic dye categories are released into the environment [1]. Apart from the textile industry, synthetic dyes are also widely used in cosmetic, paper and printing, drug, garment firms, leather, and food processing industries [2]. Dyes are synthetic aromatic organic compounds that are utilized to color a variety of substances. They are essentially chemicals that adhere to the substrate and add color to its

feature. The color of the dye varies depending on the chromophore group. It's usually a complicated structure with a substrate of relatively high biotoxicity [3]. Dyes can be classified based on their type and characteristics. Chromophores and auxochromes, which are usually refractory heterocyclic and aromatic molecules, are found in dye structures. Auxochromes and chromophores, which are usually refractory heterocyclic and aromatic molecules, are found in dye structures [4].

Dyes can generally be divided into anionic, cationic, and non-ionic categories [5]. Additionally, dyes can be categorized according to their chemical properties, molecular structure, and solubility. Tables 1 and 2 highlight the structures, chemical formula, wavelength, and color

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#### Table 1

Different types of dyes' chemical composition and index properties as modified from [9-12]:

Dye specific name	chemical formula	Molecular weight (g/ mol)	Wavelength, $\lambda_{max}$ (nm)	Color Index No. (C. I. No.)
Direct Black 22	$C_{44}H_{32}N_{13}Na_3O_{11}S_3\\$	1083.97	470	35435
Reactive Red 159	$\mathrm{C}_{32}\mathrm{H}_{18}\mathrm{ClF}_{2}\mathrm{LiN}_{6}\mathrm{Na}_{2}\mathrm{O}_{11}\mathrm{S}_{3}$	885.08	514	-
Methyl Orange	$C_{14}H_{14}N_3NaO_3S$	327.33	460	13025
Methylene Blue	C <sub>16</sub> H <sub>18</sub> ClN <sub>3</sub> S.3H <sub>2</sub> O	319.85	660	52015
Acid Orange 7	$C_{16}H_{11}N_2Na_4S$	350.3	480	-
Acid Orange 10	$C_{16}H_{12}N_2O_7S_2{\cdot}2Na$	452.38	475	16230
Acid Red	$C_{20}H_{11}N_2Na_3O_{10}S_3\\$	604.5	506	16255
Acid Blue 9 Basic Blue 41	$\begin{array}{l} C_{37}H_{34}N_2Na_2O_9S_3\\ C_{20}H_{26}N_4O_6S_2 \end{array}$	792.84 482.57	- 609	42090 105
Basic Red 46 (BR46)	C <sub>18</sub> H <sub>21</sub> BrN <sub>6</sub>	401.3	530	825
Reactive Black 5	$C_{26}H_{21}N_5Na_4O_{19}S_6$	991.82	597	306452
Reactive Red 141	$C_{52}H_{34}O_{26}S_8C_{l2}N_{14}$	1774.19	-	-
Reactive Red 195	C <sub>31</sub> H <sub>19</sub> ClN <sub>7</sub> O <sub>19</sub> S <sub>6</sub> 5Na	1024.24	543	-
Direct Red	$\rm C_{35}H_{25}N_7Na_2O_{10}S_2$	813.72	505	29160
Vat Black 25	$C_{45}H_{20}N_2O_5$	668.65	675	69525
Vat Yellow 1	$C_{28}H_{12}N_2O_2$	408.41	587	70600
Acid Red 14	$C_{20}H_{12}N_2Na_2O_7S_2$	502.431	517	14720
Acid Yellow 9	$C_{12}H_{11}N_3NaO_6S_2 \\$	380.352	387	-
Remazol Brilliant Violet 5R	$C_{20}H_{16}N_3Na_3O_{15}S_4\\$	735.58	560	18097
Reactive Yellow 15	$C_{20}H_{20}N_4Na_2O_{11}S_3\\$	634.57	416	-
Reactive Red 120	$C_{44}H_{24}C_{12}N_{14}Na_6O_{20}S_6$	1469.98	512	292775
Reactive Orange 16	$C_{20}H_{17}N_3Na_2O_{11}S_3\\$	617.54	494	17757

index number of different dyes. Direct, reactive, azo, acidic, and basic dyes are the most prevalent types [6]. The chemical dyes that are most frequently employed on an industrial scale are Sulfur, azo anthraquinone, indigoid, triphenylmethyl, and variants of phthalocyanines [7,8].

Soluble dyes comprise acid, basic, direct, mordant, and reactive dyes, whereas insoluble dyes comprise azo, disperse, Sulfur, and vat dyes (see Table 3).

Dyeing is a phase of the textile process that involves the use of compounds such as formaldehyde, dye, surfactant, salt, and metals [16]. Due to the semi-continuous nature of dyeing and finishing processes, the dyes and chemicals employed frequently change from day to day, and occasionally even within the same day [17]. The dyeing industry uses a lot of water [18]. Shortfalls in the dyeing process, on the other hand, result in up to fifty percent of the dyestuff directly producing huge quantities of dye wastewater full of various types of pollutants [19]. Nonetheless, it also contain value-added products shown in Fig. 1 that can be recovered from the wastewater [3,20].

Several kinds of chemicals are being used in the dye industry. This

includes: acetic acid, vat dyes, auxiliary chemicals, soaps, sulfur, nitrates, and naphthol compounds, and heavy metals ( $Mg^{2+}$ ,  $Al^{3+}$ ,  $Ni^{2+}$ ,  $Hg^{2+}$ ,  $Cu^{2+}$ ,  $Cr^{6+}$ ,  $Fe^{2+}$ ,  $Mn^{2+}$ ,  $Pb^{2+}$ ) which significantly contribute to the high toxicity of dye wastewater [21]. The dye wastewater is primarily made up of a large number of complex substances with high contents of organic dyes, silica, organic pollutants (wax) and heavy metals. Discharging untreated dye wastewater and their metabolite products resulting from the dyeing process may lead to noxious, genotoxic, mutagenic, allergenic and carcinogenic effects on living things [22]. In humans, it can also result in cancer, mutation, hyperbilirubinemia, acute renal failure, and hemolytic anaemia. They can persist in the environment for a longer length of time. As a result, their improper discharge is unacceptable.

In surface and groundwater systems, dye wastewater is the prominent source of aquatic contamination that adversely affects the aquatic environment and negatively impact the drinking water quality [23–25]. This, in turn, can have an influence on food chains due to pollutant trophic transfer and bio-accumulation [26]. It may cause (i) poor order (ii) increase in temperature and turbidity, (iii) decrease in dissolved oxygen, (iv) change in pH and color [27–30].

Change in color and turbidity prevent sunlight from diffusing into water, thereby reducing oxygen bioavailability and photosynthetic activity [3]. Highly colored dye wastewaters make environment unpleasant aesthetically [31]. In recent times, attention has been focused on wastewater discharge onto or into the ground as one of the methods of effluent disposal. It is noted that plants have the major uptake of nutrients (nitrate, ammonia, phosphorus) and soil has the capability to absorb toxic inorganic and organic substances and microbes [32–36], hence making discharging into/onto ground a harmless act to some extent [37].

However, soil and vegetation have a certain limit to absorb the pollutants from wastewater [38–40]. As time passes, they will get saturated with pollutants and eventually contaminate the groundwater. Therefore, it becomes crucial to design an effective dye wastewater treatment process to handle issues with insufficient water supplies and the requirement for environmental conservation [41]. Therefore, it is anticipated that the information in this chapter will increase readers' basic literacy, guide researchers, and be included in subsequent lab tests and system efficiencies for dye removal. The chapter described the physical and chemical dye removal processes, biological treatment processes with emphasis on sequential Batch Reactors (SBRs). It further evaluates the (i) aerobic sequencing batch reactor for dye removal and (iii) dye removal under sequential anaerobic-aerobic conditions.

# 2. Dye treatment

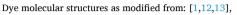
It is common knowledge that the treatment of dye wastewater is one of the environmental profession's most challenging tasks. Due to their synthetic origins and complicated structure, dye wastewater presents a complex treatment challenge, particularly in color removal [42], molecular weight of dye, concentration, pH, temperature, soluble salts, electron acceptors, and heavy metals have been reported as factors that affect dye treatment [3]. The necessity to develop novel treatment techniques that would effectively manage wastewater and conserve the ecosystem has been prompted by the growing public concern over water scarcity and the strict international environmental standards.

Therefore, in order to create a final effluent that can be released or utilized again, it is necessary to select the suitable treatments [43]. The methods used for dye removal are classified into three types: biological, physical, and chemical [44]. Various techniques within the aforementioned categories have been utilized for decolorization of industrial effluents containing dyes [45]. To reap the advantages of various treatment methods, integrated treatment systems comprised of combining two or more biological methods operating under various aerobic and anaerobic conditions, or combining physicochemical and biological methods, are often used for dye removal [46].

## 2.1. Physical and chemical treatment processes

The chemical and physical treatment techniques such as: coagulation, precipitation, electrochemical coagulation, flocculation, irradiation, electrolysis, flotation, reverse osmosis, oxidation and advanced oxidation, ozonation, pyrolyzed petrified sediment, membrane

# Table 2



filtration, advanced oxidation processes (photocatalysis, hydrodynamic cavitation, Fenton, and photo-fenton), sonochemical degradation, ultrafiltration, or adsorption [47–49], have been used for decolorization of dye wastewater [50]. Some of the merits of the physical treatments process include but not limited to simple design, low cost of maintenance and operations, absence or fewer chemical requirements and absence of inhibitory effect due to the presence of toxic substances [51–53]. However, because they scarcely convert the dye from its liquid

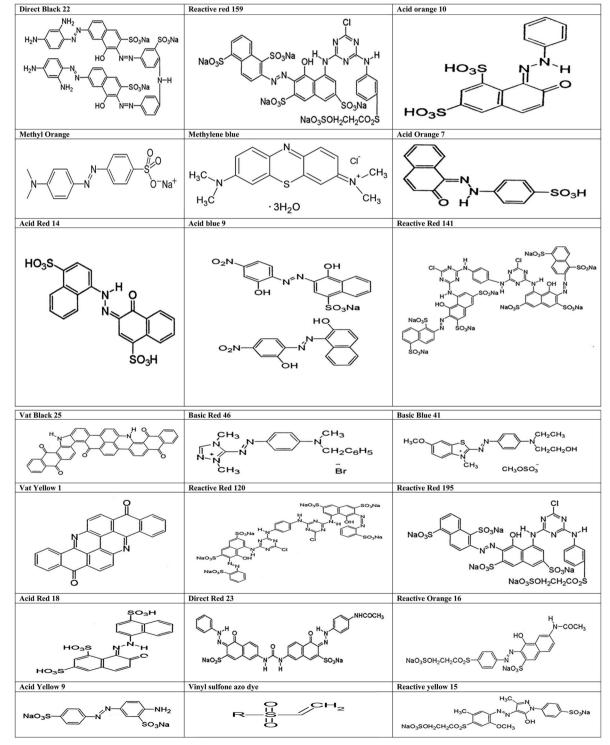


Table 3

I	/arious c	iye	types	and	their	properties	as	modified	from	[	14	,1	.5	]:
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Dye type	Properties
Acid dyes	Improved wool, silk, polypropylene, and polyamide fibers are utilized in conjunction with organic sulfonic acids and a few other types of fibers
Basic dyes	Acrylic fibers are colored using basic dyes, which have basic groups like -NH2 and -NR2. They are usually used in conjunction with a mordant and are soluble in water
Direct dyes	inexpensive and soluble in H <sub>2</sub> O
Disperse dyes	contain nitro, azo, and anthraquinone groups; they are water- insoluble and have substantial affinity for nylon, cellulose, and acrylic fibers
Vat dyes	Most frequently used for dying natural cellulosic fibers, insoluble in water, good rapidity.
Reactive dyes	used most frequently to color cellulosic fibers made of natural materials
Azo dyes	covers 60–70 % of total dyes produced and utilized in industries.
Anthraquinone dyes	used for mummies wrapping. Examples: Reactive Blue 4, Disperse Blue 73 dyes
Indigo dyes	organic compound with a distinctive blue color, unique in dyeing denim jackets and jeans, represent an ancient organic color
Sulfur dyes	It can be produced by heating phenols, nitro compounds, or aromatic amines with alkali polysulfides and is used for coloring celluloid fibers. It is insoluble in water.
Phthalocyanines	architectural flexibility, high stability, better spectroscopic individualities, and low coordination



Fig. 1. Value-added products from dye industry wastewater.

to solid state, these physicochemical treatment techniques are classified as non-destructive [54–56]. Although the chemical treatment processes tend to be more efficient than the physical process, requires less space and usually compact set up and treats diverse wastewater contaminants [57].

They are also facing several technical and economic limitations such as: hazardous sludge generation, operational problems, costly, energy intensive, adsorbent regeneration, waste and sludge handling problems, low performance, insufficient versatility and usage, interference by other wastewater constituents, membrane fouling and production of toxic by-products [58–61]. Other drawbacks related to the chemical treatment processes are the generation of toxic or hazardous metabolites and by-products during treatment [62,63], pH dependent, low stability, and demand for large amount of chemicals [64,65], Whilst the physical treatment process is further limited it application and dependent on high temperatures, initial concentration of chemical oxygen demand (COD), biological oxygen demand (BOD), color, heavy metals and pH of the dye wastewater [57,66].

#### 2.2. Biological treatment processes

Dye decolorization through biological treatment techniques has gained momentum as they are the most efficient, sustainable, renewable, reliable, environmental-friendly, financially appealing and selective methods with low energy, and as well less sludge generation, that can be applied to a broad spectrum of dyes in industries of varying scales [9]. They produce less harmful wastes for disposal and ensure complete mineralization of organic pollutants. More so, minimum chemical and water usage is required [11,67]. Less sludge is produced in biological treatment systems via techniques such the activated sludge process, pure culture of the decolorizer, oxidation ponds, aerated lagoons, and sequencing batch reactors [68–70].

In wastewater containing dye, a variety of bacteria, algae, fungus, actinomycetes, plant powders, and baker's yeast may be able to remove color and organic materials [71,72]. Bacterium is the most extensively investigated and adopted for azo dye decolorization because of its widespread high growth rate and adaptability. Enzymes like oxidoreductase, laccase, azoreductase, and peroxidase could also be employed [73]. Adding electron donors such as glucose or acetate ion appears to stimulate azo bond reduction cleavage [74]. According to Ref. [75], added colors to dyestuffs Contaminated water is a type of refractory organic substance that bacteria in conventional biological treatment systems can either solubilize or use as sources of energy and carbon. However, a high influent color concentration can result in a partial suppression of the color-degrading bacteria. In biological systems, colorant elimination occurs either by adsorption, degradation, or a combination of both. The decolorization of various groups of bacteria is affected by aerobic and anaerobic operational conditions, as shown in Fig. 2.

Aerobic systems are frequently used for dye degradation as a component of numerous biological processes. Unfortunately, most times the treatment is insufficient. Interestingly, since many dyes are vulnerable to reductive alteration in anaerobic conditions, anaerobic systems can handle significant volumes of organic waste. Therefore, both color degradation and dye molecule disintegration can be accomplished via a hybrid aerobic and anaerobic approach [13]. In the hybrid environment, dye removal is often accomplished through bond cleavage, which causes the formation of hazardous, colorless aromatic amines as well as metabolite biodegradation [76,77]. Several researchers focused on using SBR for dye wastewater treatment because it is a periodic discontinuous process that promotes the mineralization of toxic compounds. Regardless of any inflow condition, the exposure time, frequency, and amplitude of the respective concentration can be set.

# 3. Sequential batch reactors (SBRs)

SBRs are an activated sludge process that is batch-operated and in which the different conditions are satisfied at different times in a single reactor, basin, or tank. SBR which proves to be a compacted system needs a long sludge age to treat wastewater [78]. Other than time orientation, another advantage of SBR is the operational flexibility [79]. The various treatment phases are carried out in a cycle at predetermined and programmable intervals [80]. Aeration and clarification are included between the five (5) processes of filling, reaction, settling, drawing, and idling (see Fig. 3). The SBR system is developed based on the influent load, biomass mass and settleability, reactor capacity, aeration system, and the percentages of the overall cycle specified for

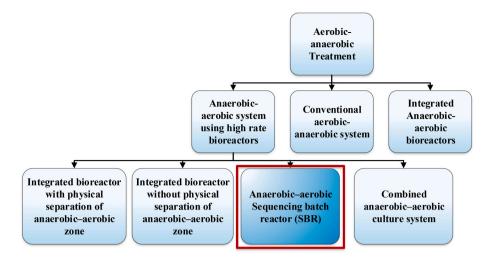


Fig. 2. Biological conditions for dye treatments.

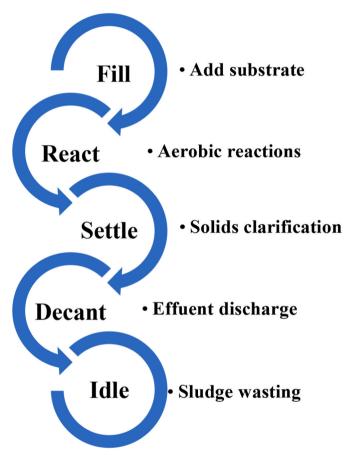


Fig. 3. SBR operational cycle.

the several treatment phases. SBRs are outfitted with diffusers, inlet and exit valves, oxygen providing kits, and mechanical sludge outtake devices in order to efficiently regulate the system [81]. Sludge settles once aeration is stopped and the supernatant liquor is removed by a drainage system. Filling and reaction times vary depending on the aeration and mixing procedures used to remove carbon, nitrogen, and phosphorus. Sludge wasting can occur close to the end of reaction stage, as well as during settling, decanting, or idling.

For better COD and dye degradation, the sludge activation process known as SBR has been enhanced to combine both aerobic and anaerobic phases in a single unit. It acts as an alternative to traditional wastewater treatment systems for suspended growth [82,83]. It is an amended activated sludge system utilized to resolve the low-density and bulking sludge obstacles occasioned by the clarifier's high volume, with the added benefit of operating in either oxic or anoxic conditions. However, sludge age alongside various physical and chemical conditions affects the performance of such a system [84]. Practical solutions to the operational issues with the activated sludge process have been achieved using SBRs. The periodic operation, which subject organisms to frequent oxygen and substrate gradients, creates selective pressures that can lead to the selection of particular cultures that are able to break down xenobiotic chemicals and endure harsh and unpredictable environments [85].

The conventional SBR, which first oxidizes ammonia to nitrite before going on to oxidize nitrite to nitrate and produce nitrogen gas, is an essential step in the nitrification-denitrification process [76]. SBR has a long history of application in the treatment of textile waste, especially for the removal of azo colors since it enables the maintenance of fungal activity for an extended period of time and may produce better dye decolorization outcomes than batch cultivation [86]. Several types of SBRs under different operating conditions are investigated in treating textile wastewater. These include aerobic SBR, anaerobic SBR, and anaerobic-aerobic SBR. According to Ref. [18], SBRs are very successful at removing organic matter, nutrients, and dyes from textile industry wastewater.

# 4. Evaluation of aerobic and anaerobic SBR conditions for dye removal

# 4.1. Aerobic sequencing batch reactor for dye removal

Due to their simplicity of use and relatively low operational and capital expenses, aerobic methods are preferred for the treatment of dye wastewater. Aerated lagoons, activated sludge, and biofilm processes are among aerobic biological approaches for treating dye wastewater [87]. An activated sludge system is a kind of aerobic treatment process in which oxygen is introduced into the reactor via an aeration tank, supported by a settling tank that would help enable biological flocs to settle, trying to separate biological sludge from clear treated water [88]. Typically, an SBR reactor's bottom fine-bubble diffuser is where aeration is performed by pumping air through it. It was discovered that the initial substrate to sludge concentration ratio and response time were crucial factors in organic matter biodegradation. SBR, complete mix, and extended aeration are the three commonly used systems for activated sludge. Azo dye decolorization is possible under low activity aerobic conditions because, oxygen is a more efficient electron acceptor than azo dyes [89].

Enzymes produced by microorganisms in wastewater (mono- and dioxygenase), catalyze the integration of oxygen from  $O_2$  into the aromatic ring of organic compounds prior to ring fission [90]. An aerobic treatment has the disadvantage of having high operating cost and complexity in mineralizing azo dye reduction ingredients. This is due to insufficient microbial population competent of metabolizing such compounds [22]. However, it is outweighed by higher reactor stability which are less sensitive to fluctuations in effluents and sustainability. According to literature, aerobic granules (AGS) have been widely used for dye color and organic matter removal from dye wastewater. It has been disclosed that dye-decolorizing AGS created under microaerophilic conditions and at low DO  $\leq 2$  mg/L have merits such as: convenient and steady microbial structure, good settling attributes, high biomass concentration and resistance to shock loads, better solid–effluent separation, as well as denser and stronger aggregate structure [16].

Pollutants in textile wastewater include suspended solids, heavy metals, heat, basicity, organic and inorganic matter. For textile effluent that had been anaerobically treated and contained the pigments MX-8B, MX-5B, and MX-2R, aerobic SBR was examined as a final step. The system was also capable of reducing influent dye concentrations, effluent COD and SS concentrations to 5, 100 and 20 mg/L, respectively [91]. Biological biodegradation of textile wastewater using an SBR system yielded 80-95 % COD removal over a reaction time of 22-25 hours. The maximum BOD removal at 91 %, was achieved for a  $S_0/X_0$ ratio of 0.16 on a BOD/MLSS basis. Removal rates were ranged from 8 to 48 and 0.4 to 28 for dissolved and suspended solids [92]. The combination of three (3) white rot fungi (Pleurotus floridanus, Ganoderma lucidum and Trametes pubescens) has been proven to be good in terms of textile wastewater decolorization in SBR [83]. Studies have revealed that the GAC-SBR system is more effective for treating textile wastewater than the customary SBR system. The GAC-SBR system could be used to treat textile wastewater comprising up to 160 mg/L of direct red 23 or direct blue 201 with no consequences on COD and BOD5 removal efficiencies. However, due to the adverse effects of dye at high concentrations, it had an effect on denitrifying bacteria and dye removal efficiency. To reduce the efficiency of the GAC-SBR system, direct red 23 was more impactful than direct blue 201 [75].

In a more recent article by the same authors, resting and autoclaved bio-sludge from a sewage treatment plant were used as adsorbents for dye and organic matter removal, with the former demonstrating greater adsorption capacity than the latter. The system demonstrated a low direct dye removal efficiency with raw textile wastewater at an organic loading of  $0.083 \text{ kgBOD}_5/\text{m}^3$ -d. When 0.89 g/L was added to the textile wastewater, the removal efficiencies of dyes, COD, BOD5, and TKN increased [93]. Adsorption of Basic Blue 41 (BB41) and Basic Red 46 (BR46) onto resting and thermally treated bio-sludge was accomplished using a GAC-SBR system. According to the study, BR46 was absorbed onto the bio-sludge at a greater efficiency than BB41. Resting bio-sludge had a 22 % higher color adsorption efficiency than thermally treated bio-sludge. The GAC-SBR system after supplementing glucose had the highest color (80 %) and COD (97 %) removal efficiencies 80 % and 97 % respectively after 5 days of HRT [94].

Aerobic granules for the treatment of methylene blue (MB)contaminated water were efficiently produced in an SBR system using activated sludge as inoculum with glucose, acetate, and soluble starch as carbon sources in the influent mixture. After 87 days of cultivation, stable granules primarily composed of bacterial microcolonies with distinctly identified shapes and diameters ranging from 2 to 4 mm were acquired. The granules demonstrated to be viable and efficient for degrading MB-containing wastewater [71]. One other study on MB found that combining SBR and Fenton oxidation improved COD and color removal efficiencies over a single SBR-stage treatment. It was better at improving the biodegradability of chemical oxidants while reducing their toxicity [95]. While still on aerobic granules, Reactive yellow 15 (YD) dye decolorization, as well as COD and ammonium removal were attained in a bioreactor that operated under microaerophilic conditions for about 80 days. It is worth noting that they were significantly removed during the anoxic period in the presence of nitrifying and azo dye decolorizing bacteria [96].

For bio-decolorization of Acid Orange 7 (AO7) dyes, a longer anoxicreact period is preferable, although the shortest anoxic-react period achieves the maximum COD elimination rate. As shown in Table 3, AO7 bio-decolorization and COD removal in an anoxic–aerobic REACT operated SBR without co-substrate performed better [97]. Inducing an anoxic condition aided in the initial reduction of Acid black 210, which resulted in dye mineralization during the subsequent aerobic process. Surprisingly, neither of the organic loading rates tested inhibited reactor performance. Forced short-term unsteady state conditions in SBR operation, along with recurrent microbe exposure to specified process conditions, produced effective system performance [46].

COD removal efficiency fluctuated during two weeks of anoxicaerobic REACT acclimatization. However, both Reactive Orange 16 (RO16) biodecolorization and COD removal deteriorated by  $\sim 40$  % due to successive RO16 shocks. During the anoxic-REACT period at higher carrier filling ratios (10% (v/v)), the difference in COD removal and decolorization rates improved by 0.16 and 0.30 %, respectively. As a consequence of the actively metabolic system, the microbial strength in trying to remove RO16 molecules increased from 0.015 to 0.0304. It was extremely important that the attached-growth biomass system was present [98].

In an aerobic SBR system with an indigenous bacterial consortium treating Reactive Orange M2R (ROM2R), COD, BOD and color reduction stabilized at around 57 %, 43 %, and 70 % respectively. Under batch culture systems, *Pseudomonas* sp. Was the key bacterial population essential for the dye bioremediation [99]. The decolorization performance of Vinyl sulfone azo dye (RB5) by *Trametes versicolor* immobilized on polyurethane foam in SBR was investigated in a study by activity [100]. Manganese, Laccase, and lignin peroxidase activities closely related to the decolorization process were produced in a sustained increment throughout the SBR culture. Changes in the dye's chemical structure, which are attributed to enzyme activity, were found to be the cause of dye decolorization, as evidenced by variations in the absorption spectrum.

The volumetric dye loading rate can have an effect on dye removal efficiency. This was demonstrated in an SBR system operating under aerobic conditions varying 3–20 g dye/m<sup>3</sup>/d concentrations of Blue Bezaktiv S-GLD 150 (BB150) [90]. At volumetric dye loading rates below 15 g dye/m<sup>3</sup>/d, the results showed that microbial consortia had the best cleansing ability, with the highest decolorization rates and COD removal rates. Notwithstanding, decolorization and COD removal, lowered at 20 g dye/m<sup>3</sup>/d. The impacts of dye concentration on the decolorization of reactive brilliant red K-2G and KE-3B were examined. The microbial community demonstrated high color removal efficiency for 400 mg/L and 1100 mg/L concentrations of KE-3B and K-2G, respectively [101].

Photo-treated reactive red 141 solutions were then biodegraded in an aerobic SBR under different HRT conditions. Assuming complete biodegradability, low residual dissolved organic carbon values were found, which were comparable to those acquired in a control reactor. The aerobic biological treatment of Direct Blue 85 (DB) simulated textile wastewater showed a significant organic carbon removal with limited decolorization efficiency. But, simultaneous SBR operation with waste sludge, a significant enhancement in the reactor performance was obtained in terms of TOC, COD, BOD<sub>5</sub>, color and dye removals [76].

# 4.2. Anaerobic sequencing batch reactor (ASBR) for dye removal

An anaerobic dye treatment system is a process characterized by low energy requirements where microorganisms conduct their metabolism only in the lack of oxygen and produces methane and carbon dioxide as byproducts [102]. Transferring electrons from the oxidized substrate to electron acceptors is a biological process requiring a complicated electron transport chain mediated by dehydrogenase, menaquinone, and cytochrome pools [103]. It is likewise a non-specific extracellular process where dye acquires decreasing equivalents from chemical or biological origins [10].

In the absence of substrate which azo dye tries to act as an electron acceptor but there is very little dissolved oxygen, aromatic amines may function as electron donors [104]. High organic load wastewater is treatable by anaerobic microbes. They may greatly reduce color intensity significantly better than aerobic microorganisms and are vulnerable to the reductive reduction of many dyes [1,13]. A study by Ref. [103], reported that dye removal through anaerobic respiration was enhanced when alternate terminal electron acceptors such as: humic acids, metal oxides, di-methyl sulfoxide, sulphides, fumarate,  $NO_2^-$ , sulfur,  $NO_3^-$ , and thiosulphate are present.

A number of industries have employed different anaerobic technologies because of the following advantages: flexibility, stable performance, lower sludge production, operational simplicity, high biogas yield, efficient effluent quality control, higher reaction rate at the start of reaction, smaller area requirements, resistance to shock loading and facilitation of further pollutants degradation [11,68,105]. However, it has the disadvantage of longer start up time and acclimatization, and potential production of odorous and corrosive gas. Incomplete biodegradation of dyes [89]. Also, operating an anaerobic reactor needs agitation to enhance substrate transfer to the microorganisms present in granulated biomass for anaerobic degradation [106].

Endogenous sludge lysis offered the required quantity of reducing equivalents for orange II degradation in a nutrient-free and co-substrate SBR system working in an anaerobic mode. Increasing co-substrate concentrations improved Orange II decolorization rates but hampered COD removal. As can be seen in Table 3, high decolorization efficiency was achieved in the system when 600 mg/L of Orange II was added [102]. When the concentration of Methyl orange (MO) in an ASBR was increased, the microbial community configuration drastically changed. The most abundant microorganisms found in the anaerobic system were *Anaerolineaceae* and *Sulfuricurvum*.

A good correlation existed between community structure and degradation performance. A relatively modest amount of methane was created as a result of the successful competition of azo reduction over sulphate reduction and methanogenesis [68]. A 23-h reaction time in a fully anaerobic operated reactor treating tetra-azo dye Direct Black 22 (DB22) resulted in an effluent with high acute ecotoxicity as a result of aromatic amines build up [11]. Under anaerobic conditions, a study

# Table 4

Dye removal efficiencies in SBR system.

Dye type	Influent dye concentration (mg/L)	Operational temperature (°C)	Operational capacity of the reactor (L)	Cycle time (h)	HRT (d)	SRT (d)	Percentage removal (%)	Ref.
Aerobic condition								
Procion Red H-E7B	250	21–23	2	24	1–10		65 dyes 52.4 DOC	[ <mark>98</mark> ]
Vat Yellow 1	40		10	24	3	5–16	75.12 dyes; 70.61 COD;	[95]
Direct Red 23	40		10	24	3-7.5		76 dyes	[103]
Mixture of MX-8B, MX- 5B & MX-2R	25–100		3	6–12			58 COD	[91]
Direct Blue 201	40–160		10	24	7.5	28–31	94–99 dyes 94–97 TKN	[75]
Brill Blue KN-R	20–40		5.5	24	1.83	10	57 dyes 97.22 COD	[24]
Reactive Brilliant Red K- 2G	500-1100	30		12			≈100	[101]
Reactive Brilliant Red KE-3B	200-800							
Reactive Blue 19	40		7	24	1.83	10	71.7 dyes 93 COD	[24]
Methylene Blue	4–10		4	4	0.333		56 dyes, 93 COD	[71]
Blue Bezaktiv S-GLD 150 dye	3–20 g/L	$27\pm3$	8	24		30	88-97 dyes; 95–98 COD	[76]
Acid Orange 7	15–60	$26\pm1$	3	24		15	99 AO7 dyes 90-92 COD	[ <mark>93</mark> ]
Textile wastewater	-	$28\pm1$	3.5	24	5	10-20	71.3 color, 79.4 COD	[83]
Sirius Blue K-CFN (Direct Blue 85:DB)	50-85	25		24			60-69 Color, 79 TOC, 80 COD	[76]
Reactive yellow 15	10–50		3	24			89-100 dyes. 79-95 TOC. 92-100 NH4+N	[96]
Reactive orange 16	10–1000	24–26	5	24	0.7		97 COD, 100 dyes	[90]
Methylene blue	500	$30\pm2$	5.5	24	4	16	86.3 COD, 84 colors	[95]
Methyl orange	250–500	$30\pm2$	1.5	6–48	2		96.83 color, 88 NO <sub>3</sub> –N, 86 COD, 83 TOC	[103]
Anaerobic condition							00 100	
Acid Orange 7	125-625	$25\pm1$	2	24			98 Color 88 COD	[11]
Orange II	100-600		5	24			89 Color	[102]
Methyl Orange	25–500	35	2.5	24	4–14		>75 dyes >85 COD	[68]
Reactive Red 159	1000-8000	$30\pm2$	2	24	4–24	7–24	97.68 dyes	[1]
Tetra-Azo Dye Direct Black 22		$37\pm1$	5	24			81.4 dyes 76.4 COD	[11]

focused on using mixed culture of *Sphingomonas paucimobilis, Agrobacterium radiobacter, Aeromonas hydrophila* and *Bacillus* spp. for bacterial decolorization. Within 2 h and under static conditions, 91 % and 99 % decolorization efficiencies were achieved for Crystal Violet and Malachite Green, respectively [107]. The efficiency of An ASBR system treating Reactive Red 159 stabilized on the 30th day at long SRT and short HRT with a high dye concentration. The dye effluent concentration was considerably lowered to  $142.62 \pm 5.35$  mg/L with a decolorization rate of  $264.54 \pm 7.13$  mg/L/h under the optimal conditions listed in Table 4 [1].

## 4.3. Dye removal under sequential anaerobic-aerobic conditions

Anaerobic systems are sensitive to the reductive reduction of different colors and can manage water with large organic loads, whereas aerobic systems are best for treating wastewater with modest organic loads [1]. Dye removal has been confirmed to be less under aerobic conditions than in anaerobic and microaerophilic conditions. Micro-aerophilic conditions are prioritized to: (i) remove ammonium and dye concurrently, and (ii) use oxidative and reductive biological conversions for dye removal [96]. Ultimately, the combination of anaerobic and aerobic SBR processes with flocculent-activated sludge processes has been identified as being the most hopeful and satisfactory concept for dye removal using mixed bacterial cultures [10]. This is due to the fact that, toxic byproducts produced under anaerobic/anoxic conditions can be broken down aerobically [108].

The integration of both processes results in anaerobic color removal by bond cleavage, which produces colorless and hazardous aromatic amines, and following aromatic amine mineralization via the aerobic treatment of non-specific enzymes via hydroxylation and aromatic ringfission [76,109]. Dye decolorization in an SBR working under alternating anaerobic-aerobic conditions may be mediated by two different methods. First, electrons from the oxidation of the carbon substrate are directly transferred to the azo bond. The second technique uses biological sulphate reduction to chemically reduce the azo dye in a bio-mediated manner. The redox potential (ORP) can be used as the only control variable to regulate both the anaerobic and aerobic end stages.

Remazol Brilliant Violet 5R (RBV 5R) decolorization could be accomplished through one of two mechanisms. Either by sulphate reduction or by direct electron transfer from the cell to the dye. In anaerobic mixed culture with glucose and sulphate, RBV 5R was decolored. Findings revealed that sulphate-reducing bacteria (SRBs) remarkably contributed to organic carbon removal and dye reduction. Regrettably, because aerobic conditions are detrimental to SRBs, alternate anaerobic–aerobic phases lead to significantly reduced anaerobic sulphate removal yields. Additionally, under aerobic conditions, anaerobic sulphides can be re-oxidized, increasing the sulphate accumulation at the beginning of succeeding anaerobic phases [109]. Under anaerobic conditions, mixed microbial cultures make it possible for the reductive decolorization of RBV 5R dye and the formation of benzene and naphthalene-based aromatic amines. The subsequent aerobic phase was reported to be beneficial in removing additional COD.

This indicates that the contribution of the aerobic phase to color removal is insignificant, despite the fact that it was capable of removing benzene-based aromatic amines [110]. The presence of oxygen has a negative impact on the azo reductase enzyme and color removal efficiencies. A very small amount of dissolved oxygen, on the other hand, has no inhibitory effect [104]. Under cyclic anaerobic conditions, an activated sludge bacterial consortium could decolorize Remazol Black B (RB-B) and RBV 5R. It was discovered that two distinct successive decolorization periods were observed due to different decolorization mechanisms. The interference of mass transfer limitations was experienced during diazo dye RB-B decolorization. Adsorption to biomass was not noticed for either dyes [111].

The initial AR18 dye and COD concentrations at the end of an alternating anaerobic–aerobic SBR with external feeding were

immensely decreased from 500 mg/L and 3270 mg/L to 10 mg/L and 95 mg/L, respectively. This shows that nearly complete AR18 decolorization has occurred [21]. AR18 decolorization was aided by AR18 infiltration into the interior layers of the granules as well as the capacity of anaerobic microbes within the granules to break down the dye by using the firmly bound EPS as a carbon and energy source. The aerobic granules, however, were unable to biodegrade anaerobically produced AR18 intermediates under aerobic conditions [77]. Similar to RBV 5R dyes, authors [82], reported that, Acid Red 18 and its degradation products made up 15 % of the COD still present in the treated effluent, the majority of AR18 dye removals took place in the anaerobic phases, with the aerobic phase contributions being minimal.

Alternate anaerobic-aerobic SBR process combined with UF treatment was used for Acid yellow 256 dye treatment. Results show high decolorization efficiency and COD reduction with *S. marcescens* and *K. oxytoca* as the predominant bacterial species. Increasing the aerobic cycle duration increases COD removal efficiency and lower decolorization rate [89]. Basic Red 46 (BR46) decolorization could approximately be fully achieved in SBR while COD removal gradually decreases with increasing dye concentration [108].

An anaerobic—aerobic SBRs, operating with aerobic granular sludge at varying hydrodynamic regimens was used for Acid Red 14 (AR14) dye biodegradation. The detection of the aromatic amine 4-amino-naphthalene-1-sulfonic acid, that was further aerobically decomposed, encompassing deamination and hydroxylation of the aromatic ring, proved the AR14 bio-decolorization via anaerobic azo bond reduction [112]. AR14 dye and its decomposition products had no negative impact on treatment performance, as organic load removal efficiency was greater than 80 %, with up to 77% having occurred in the anaerobic phase, as shown in Table 5.

This corresponds to an increase in the biomass of *Defluviicoccus*related glycogen accumulating organisms. The return of SRT values above 25 days increased biodecolorization yield while also promoting the complete bioconversion of the recognized aromatic amine during the aerobic reaction phase. Granule breakup took place only in the dye-free control SBR after long-term operation. This signifies that AR14 dye has a significant impact on granule stability [22]. Decolorization occurred via anaerobic azo bond reduction with aromatic amine creation, including the highly recalcitrant 4A1NS, when AR14-decolorizing bacterial strains from the Oerskovia genera were isolated from an anaerobic-aerobic SBR operating with a synthetic textile wastewater feed for a prolonged period of time.

Under anaerobic conditions, *Oerskovia paurometabola's* high color removal ability can reach 100 mg AR14/L removal in 24 hours [10]. To examine granules' ability to treat real dyeing wastewater, simulated dye wastewater containing various dye mixture was treated with an influent concentration gradually increased from 50 % to 100 %. After 90 days of operation, the toxic effect of the dye wastewater resulted in a 43.3 % reduction in the amount of specific oxygen uptake rate. The granules collapsed at 100 % influent dye concentration, resulting in a granular ratio reduction.

Biodegradable carbon sources play a significant role in achieving highly efficient Reactive Red 195 (RR 195) dye wastewater treatment. It has been suggested that in order to supply the electron equivalents needed for dye molecule oxidation, the influent COD/dye ratio should be carefully chosen. This is due to the fact that high influent color concentrations may cause partial inhibition of color degrading microorganisms [13]. Addition of external carbon source in an SBR system treating AO7 dye worsens COD removal efficiency. However, it enhanced the system's reductive condition. The anaerobic and aerobic microbes mixed in the bioreactor at various levels is thought to have contributed in the removal of both color and organic matter [113].

Typical of anaerobic SBR phases, TOC exhibits no remarkable change, because dye transformation into the respective amines occurs. Reactive Black 5 (RB5) dye contaminated water treatment with a GAC–biofilm SBR system resulted in a high removal of COD, RB5, and its

#### Table 5

Dye removal efficiencies under sequential anaerobic-aerobic conditions.

Dye type	Influent dye concentration (mg/L)	Operational temperature (°C)	Operational capacity of the reactor (L)	Cycle time (h)	Anaerobic: Aerobic reaction time (h)	HRT (d)	SRT (d)	Percentage removal (%)	Ref.
Acid Orange 7	5000		1	24	10.5:10	1.667	15	90-99 dyes 80 COD	[117]
Acid Orange 7	50	$30\pm1$	8	24	27.35:9.65			85 dyes 91 TOC	[117]
Remazol Brilliant Violet 5R	100	25	6.5	6–48	12:11.9			92 dyes >75 COD	[10]
Remazol Brilliant Violet 5R	50–100	$25\pm1$	6.5	12	8:2.75		15	38 dyes 96 COD	[104]
Reactive Red 195	30–50	$36\pm2$	5	12	19:4		30–50	97 dyes	[13]
Reactive Black 5	10–250		1.8	24	16:4			98.97 dyes 94 COD	[104]
Acid Red 18	0–280	$28\pm2$	5.5	24	14:8.5	2.75	12	44 dyes >85 COD	[82]
Remazol Brilliant Violet 5R	100	$25\pm0.5$	6.5	12	6:6		15	83-89 dyes	[109]
Acid Red 18	1000		7.7	24		2.2		97 dyes 99 COD	
Reactive Red 198	20–50	$23\pm2$	10	12–24	16:4		15	76-98 dyes 81-94 COD	[116]
Acid Red 18	500	$22\pm2$	5.5	24	16:7	2	18	88-98 dyes. 55-91 COD	[21]
Acid Red 14	20–60	23	1.5	6	1.5:3.5		15–25	90 Color 77 COD	[22]
Acid Red 18	50–100		5	6	1.33:4.33	0.375		38 dyes >85 COD	[77]
Basic Red 46	5-500		10	8–24	16:6	1	20	65 dyes	[22]
Dye wastewater	50–100	$27\pm2$	4	6	1.25:4.57			42.6 dye 87 COD	[16, 82]
Acid Red 14	40		1.5	6	1.5:3.5	0.5		89 dyes	[108]
Reactive Blue 21	50	$30\pm1$	4	24	8:13	3	10	98 dyes. 98.5 COD	[21, 116]
Reactive Blue 21	0–100		4	21		3	10	98 dyes 98.5 COD	[16]
Acid Red 14	20-100	30	4	8				91 dyes	[10]
Acid Red 18	500		20	24		2	45	80	[118]

aromatic amines. As the concentration of the co-substrate added to the influent increased, the effectiveness of RB5 removal also improved [114]. In order to remove Reactive Red 198 (RR 198) dyes more effectively, the anaerobic contact periods and organic loading rate were increased. However, the efficiency of COD removal declined. The color removal was influenced by the COD concentration. 20 and 50 mg/L RR 198 concentration had no inhibitory effect on the activity of the microorganisms [115]. Reactive Blue 21 (RB21) molecules were not significantly affected by microorganisms at short anaerobic period of about 4 h due to phthalocyanine dyes molecular structure strength and high solubility. Dye and COD removal were significantly improved when anaerobic phase duration was increased to 16 hours [116].

# 5. Conclusion

Environmental contamination brought on by the processing of poisonous dyes by the dyeing industries is still a problem. Numerous techniques have been used throughout dye degradation, with varying degrees of success. Recently, scientists have been paying closer attention to the use of biological therapy methods. They discovered that, azo dyes molecular structures and their metabolites have a significant impact on the efficiency of aerobic, anaerobic, anaerobic-aerobic, aerobic-anaerobic, aerobic-anaerobic, and anoxic–aerobic REACT operated SBRs with regards to bio-decolorization and COD removal rates. Stability, satisfactory dynamics, microbial community structure, and microbial diversity are essential for stable SBR efficiency. To be precise, SBR has a higher COD removal efficiency with a lower color removal efficiency. In terms of color adsorption yield, activated sludge from domestic wastewater performs better than activated sludge from the textile sector. In SBRs run in aerobic modes, the cultivation of aerobic granular sludge for color removal is possible. However, molecular oxygen drastically reduced color removal in SBR during dye wastewater treatment.

This chapter discussed aerobic-SBR treatment process for dye removal. The discussion focused on dye wastewater treatment using aerobic granules, granular activated carbon, adsorbents (waste sludge), biocarrier (polyurethane foam) white rot fungi, varying dye mixtures, dye concentrations and SBR operational parameters (HRT, SRT, cycle time etc.). White rot fungi could efficiently decolorize azo dyes. Decolorization kinetics relies on the structural complexity of dye that is mostly reformed during the anaerobic stage due to the presence of various reducing agents. Under anaerobic conditions, lesser COD removal was observed for several dyes studied in this chapter. Using a periodic discontinuous operation, dye wastewater can be successfully treated. Color removal tends to occur during the anaerobic phase, as the aerobic phase is required to subsequently effluent COD effluents concentration. Adsorbents (waste sludge, GAC etc.), different exported microbial cultures, membranes, nanofiltration (NF) and reverse osmosis (RO), biocarriers, variation of operational conditions have also been used in sequential anaerobic/aerobic batch reactors to enhance system performance.

Chemical pretreatment of dye wastewater prior to SBR treatment leads to an improved effluent quality. It could be noted in this chapter that the photocatalysis and photo-Fenton reaction as pre-treatment methods have been victoriously used by several researchers to biocompatibilized reactive dye solutions. The dye solution could be decolorized, non-toxic, and much more biodegradable after partial oxidation operated under mild conditions. It is, therefore, essential to review and provide a sustainable and ecofriendly pathway for the treatment of dye-containing wastewater with the focus on process dynamics or mechanisms that prevents the negative effects of the treatment process on the natural receiving water courses, human health, and the environment. This review compares different techniques commonly used in the treatment of dye wastewater, with a focus on recent research on the process dynamics, merits and drawbacks of the approaches and the need to refocus attention on aerobic and anaerobic SBR systems which is promising for sustainable treatment of widespread toxic and emerging organic pollutants emanating from dye wastewater from the textile industries.

# Declaration of competing interest

The authors wish to declare that there is no conflict of interest or ethical approval required regarding the publication of this manuscript.

# Data availability

No data was used for the research described in the article.

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