Activated Sludge: Conventional Dye Treatment Technique

Rudy Laksmono Widajatno, Edwan Kardena, Nur Novilina Arifianingsih, and Qomarudin Helmy

Abstract Bio-decolorization is a biological method intended to remove dyestuffs contained in textile wastewater. The biological method has three mechanisms that can be carried out at the decolorization stage, namely biodegradation, bioaccumulation, and biosorption. These three methods have been shown to have the potential to remove dyes from textile wastewater. Biodegradation is an environmentally friendly waste treatment at a lower cost than physical and chemical methods. The use of microorganisms in degrading synthetic dyes is to break the cyclic chain or the double bond of its chemical structure. Microorganisms used to degrade bacterial dyes will produce enzymes and change the chemical structure of the pollutant to be simpler so that the level of toxicity is low. The enzymes produced by the bacteria will then be used to degrade the dye. The biodegradation process carried out on the dye will also change the chemical structure of the chromophore or ausochrome groups. The reductive enzymes produced by several specific bacterial strains function to break the N = N double bond which will be replaced by two molecules of NH2. The product produced in the reduction process is two aromatic aminos which will cause

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no absorption of light in the visible spectrum which indicates that the reduction process and azo dye decolorization process occur.

1 Introduction

1.1 Textile Industry Process Overview

The textile dyeing and refinement industry is a water-intensive industry, one that uses large amounts of water to meet process needs. Specific water requirements vary from 25 to 180 L/kg-product. The use of water in the cotton dyeing process is more than that of synthetic fabrics. In the wet dyeing and refinement facility, about 72% of the total water used is process water which will then become wastewater. Details of water use in wet dyeing facilities are shown in Fig. 1.

Wastewater produced by the textile industry is a mixture of various compounds, consisting of fibers, biodegradable organic compounds, surfactants, salts such as sodium chloride and sulfate, alkalis that contribute to high pH, oils and fats, hydrocarbons, harmful heavy metals, recalcitrant compounds (difficult to degrade chemically) and persistent compounds (difficult to degrade biologically) from aromatic and heterocyclic compounds, and volatile compounds [51].

The World Bank estimates that around 17–20% of industrial wastewater comes from dyeing and fabric finishing facilities. The typical components of textile wastewater are shown in Fig. 2.

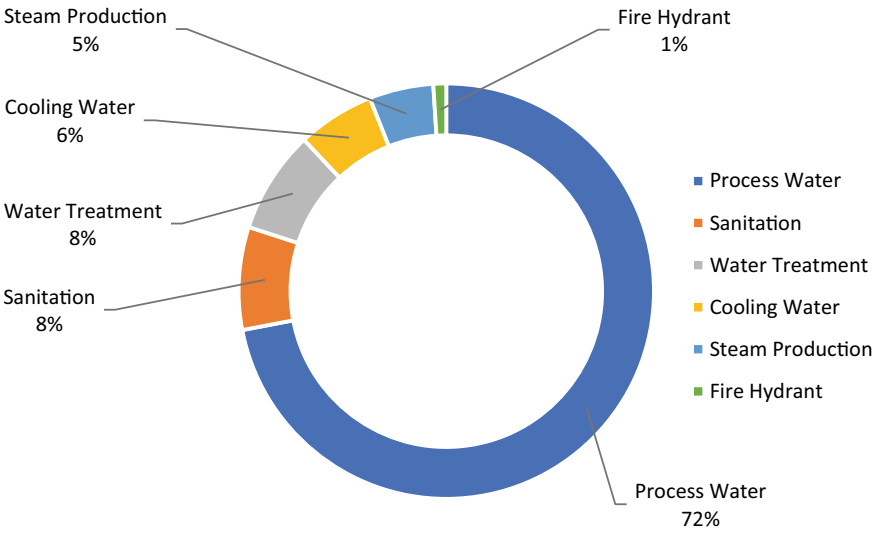

Fig. 1 Typical use of water in textile industry wet dyeing facilities

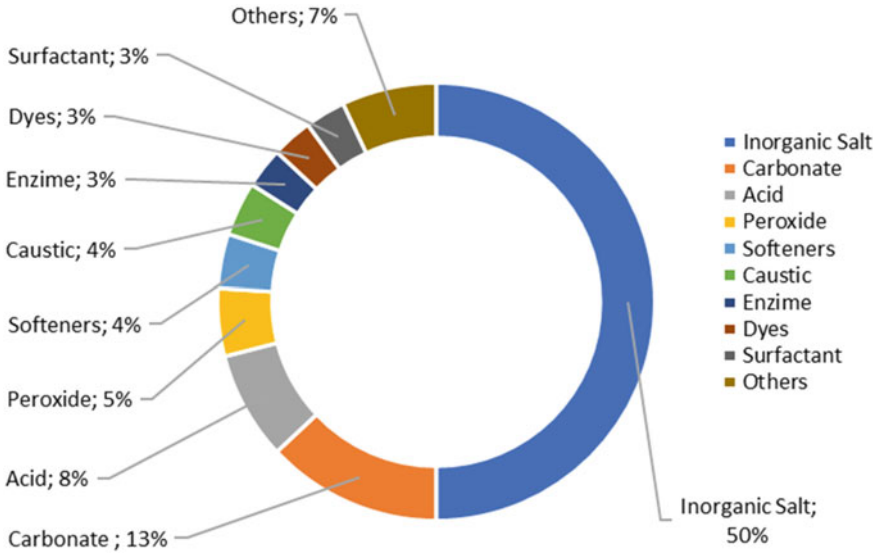

Fig. 2 Typical types of contaminants in textile wastewater

The main pollutants in the textile industry wastewater are classified into three types, floating materials, suspended solids, and dissolved solids, as shown in Fig. 3.

Textile wastewater is generally alkaline with a high content of organic compounds (700–2,000 mg/L BOD). Textile wastewater contains suspended solids, surfactants, and other organic compounds, including phenols and halogenated organic

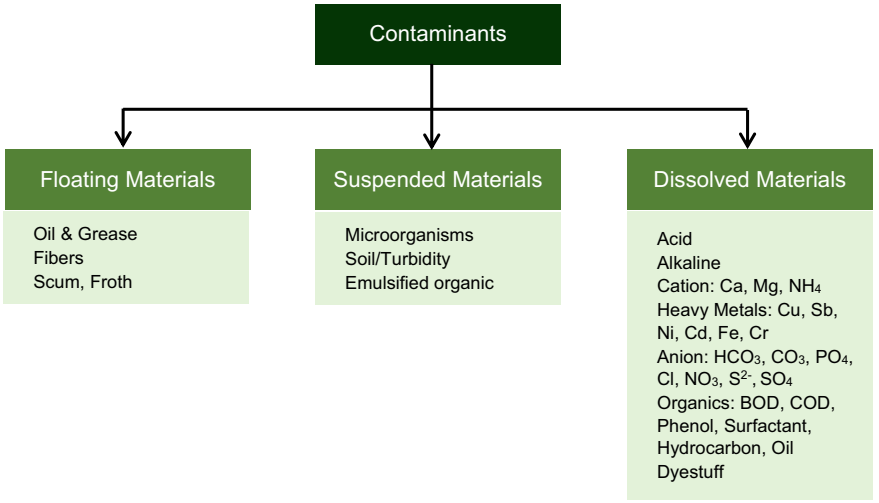

Fig. 3 Typical classification of major pollutant types in textile industry wastewater

compounds, and some contain metals such as lead, zinc, chromium, nickel, and copper. The textile industry also frequently uses a variety of flame retardant chemicals and other persistent and harmful organic compounds, such as brominated and chlorinated compounds, arsenic, and mercury, which can enter the textile industry wastewater stream. Typically, final processing steps such as bleaching, scouring, and dyeing are major sources of pollutant generation because they use large amounts of water and chemicals.

In general, wastewater sources from the textile industry are tabulated in Table 1, which are categorized by pollutant level (typical concentration), namely low, medium, and high strenght. Liquid waste with light category.

Adapted from IMI (Indonesian Ministry of Industry) 2020.

1.2 Wastewater Treatment Using Activated Sludge Process

Nature has the ability to neutralize pollution that occurs if the amount is small enough, but if it is in large quantities and loads it can have a negative impact on the environment because it can lead to changes in the environmental balance so that the waste is said to have polluted the environment. This can be prevented by treating industrial waste before being discharged into water bodies. There are many choices of technologies and techniques in wastewater management with certain characteristics. Determining the right technology for an industry is influenced by many factors such as waste characteristics, quantity, available technology, environmental impact, available land, available human resources, financial availability, ease of installation, operation and maintenance, and so on. The levels and stages of wastewater treatment can vary, depending on the specific characteristics of the wastewater generated by an industry [8]. Typical stages of the wastewater treatment process are as follows:

- Pre-Treatment. Pre-Treatment aims to remove large solids such as gravel and debris left over from the process. In addition, this stage also aims to equalize the flow rate and composition of the wastewater. This stage is generally carried out physically.
- Primary Treatment. Primary processing aims to remove settleable materials, both organic and inorganic materials by gravity, and remove floating materials through skimming methods. Primary treatment can reduce about 25–30% organics, 30–50% suspended solids, and about 60–80% of the total fat/oil contained in wastewater. Some organic nitrogen and phosphorus content as well as heavy metals attached to solids can be separated during this primary treatment. This stage is generally carried out physically and chemically.
- Secondary Treatment. Secondary treatment generally involves the removal of dissolved biological material and colloidal organic matter using an aerobic biological treatment process. The combination of secondary and primary treatment is able to remove BOD and SS levels up to 80–90% of the initial level as well as some heavy metals. This stage is generally carried out biologically and chemically.

Table 1 Identification of potential sources of textile industry wastewater generation

Potential sources	Characteristics
Low strenght	
Storm water	If the housekeeping of the drainage is not proper, there is the potential for small amounts of suspended solids, oil, and grease to go along with the water flow. Rainwater should be harvested for industrial process water
Leaks in heat exchangers, boilers, cooling towers, and steam traps	The amount of dissolved solids is low, in contrast to the condensate which sometimes has a high pH. Ideally, wastewater is separated prior to treatment
Medium strenght	
Machinary clean-up and maintenance	Oil & grease, surfactant
Boiler blowdown and maintenance	High temperature, High dissolved solid
Cooling tower blowdown	High suspended and dissolved solid
Filter and softeners backwash	High suspended and dissolved solid, salts
High strenght	
Sizing: Process for removing fine fibers that arise on the surface of the fabric so that the fabric becomes smooth, even, and clean. Sizing is a very important process for textile materials and must be carried out before mercerization, dyeing, and printing processes	High in organics content (COD/BOD) from excess starch, cellulose, wax, polyvinyl alcohol, and wetting agents. Typical water requirements are 0.5–8,5 L/kg with an average of 4.5 L/kg product
Desizing: Process for removing sizing agents which are mainly starch-based and can inhibit further textile processing	High in organics (COD/BOD), suspended and dissolved solids. Wastewater contains starch, cellulose, polyvinyl alcohol, surfactants, persulfate, and complexing agents. Typical water requirements are 2.5–21 L/kg with an average of 11.5 L/kg product
Scouring: Process for removing various impurities and contaminants from fibers using hot water and detergent	High in organics (COD/BOD) and pH. Wastewater contains surfactants, complexing agents, metals ion, and detergents. Typical water requirements are 20–45 L/kg with an average of 32.5 L/kg product
Bleaching: Process for bleaching textile materials generally uses chlorine-based bleach (sodium hypochlorite and sodium chlorite) or hydrogen peroxide	High in suspended and dissolved solids, fibers, and alkalinity, but low in organics contents. Wastewater contains chlorine, peroxide, sulfite, phosphate, enzime, fibers, and surfactants. Typical water requirements are 24–48 L/kg with an average of 35 L/kg product

(continued)

Table 1 (continued)

Potential sources	Characteristics
Mercerizing: Process for treating cellulose fibers using a caustic alkali solution (or ammonia solution) to increase the strength and affinity of the material for dyes	Low in organics (COD/BOD), dissolved solid, and oil&grease, but high in pH. Wastewater contains caustic, ammonia, wax, sulfate, antifoam, and complexing agents. Typical water requirements are 17–32 L/kg with an average of 24.5 L/kg
Dyeing: Process for giving color to textiles. In addition to dyes, various chemical additives are often used to increase the efficiency of the process. Liquor ratio (LR) is the mass ratio between the total dry matter and the total liquid/solution used. This value varies from 3:1 to 50:1 (for dyes with low affinity and low process efficiency)	High in organics, color, and dissolved solid, but low in suspended solid. Wastewater contains dyes, reducing agents, wetting agents, urea, solvents, phenolic compounds, aromatic hydrocarbons, sulfonic acid, formaldehyde, metals ion
Padding: Process where dyes and auxiliaries can penetrate into the textile material with the help of padder rollers	Wastewater contains polyacrilamide, foaming agents, polyacrylate, polymers, and sulfate
Printing: Process for giving a motif to textile using dyes or other materials, usually in the form of paste or ink	High in organics, color, oil and grease, suspended solids, and slightly alkaline. Low volume of wastewater that contains dyes, starch, binder, thickeners, reducing agents, polymers, polyacrylate, mineral oils, isopropanol, melamine derivative, and volatile organics compounds
Finishing: Process for producing a textile product to suit its intended purpose or end use. At this stage, the quality of the fabric will be increased through one or various stages of the refinement process	Low volume of wastewater that contains organics, and solvents
Coating/repellent: The process of finishing textile fabrics with various coating materials	Low volume of wastewater that contains resins, fluorocarbon, polysiloxane, aluminum, zinc, and chromium

- Tertiary Treatment. The tertiary/advanced process aims to remove suspended solids or dissolved solids remaining after going through primary and secondary processing. Advanced processing independently or separately is carried out to remove nutrients such as nitrogen, phosphorus, and organic materials that are difficult to handle (refractory organics), and heavy metals. This stage is generally carried out physically, chemically, and biologically.

If necessary, before being discharged into the environment, the effluent is disinfected first. The disinfection process aims to reduce or kill pathogenic microorganisms present in wastewater so that it does not pollute the environment. In addition, the

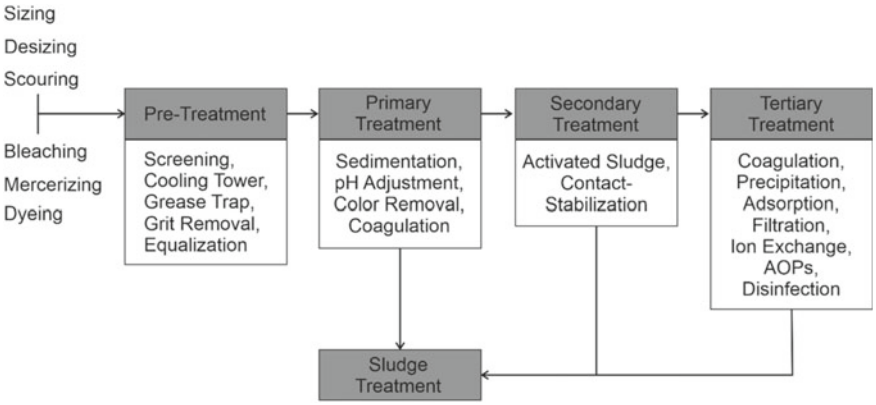

Fig. 4 Process technology options flow chart for textile wastewater treatment

wastewater treatment process generally produces sludge. The level of pollutants in the sludge will be removed or lowered to the quality standard so that it can be utilized or discharged into the environment. In general, the various technology options that can be used to treat textile wastewater are shown in Fig. 4.

Textile industry wastewater contains two elements that have the potential to pollute the environment, namely a relatively strong color and other organic carbon. In particular, the color content of textile wastewater is difficult to remove given the complex chemical structure of dyestuffs. The process of dyeing fabrics in the textile industry generally uses synthetic dyes so that wastewater discharged into the environment still contains residual dye because not all of it is absorbed by the fabric. In the textile dyeing process, about 10–15% of the dye used will be wasted into the environment as liquid waste, so the textile industry wastewater containing color must be treated first before being discharged into the environment. The textile industry wastewater treatment technology that is widely applied to date is a combination of chemical and biological processes. The chemical process commonly used is the coagulation–flocculation process using ferrous sulfate (FeSO4) and lime Ca(OH)2. With the addition of these two chemicals, a CaSO4 precipitate will be formed which will adsorb the dye. In addition, in the process of flocculation and deposition, this precipitate will also adsorb and trap other organic substances. The remaining organic substances that are still contained in the wastewater will be further processed using biological processes. In this case, the biological process commonly used is the activated sludge process or trickling filter (biofiltration). These chemical and biological treatment methods require high operating costs, especially for the procurement of chemicals and the generation of chemical sludge which is a new problem. Another treatment method that has been tried is to do physical processing using an adsorption process with powdered or granular activated carbon to remove color and organic matter at once, but this process also requires expensive costs because it has to replace saturated activated carbon. Activated carbon as an adsorbent has a limited operational time because it will experience saturation. In the adsorption process with activated

carbon, the use of granular activated carbon is more economical than powdered activated carbon, because granular activated carbon can be regenerated to restore its adsorption ability. On the other hand, the use of powdered activated carbon is more competitive than granular activated carbon because powdered activated carbon has a larger surface area so that the adsorption capacity is greater [65]. Activated carbon regeneration is an effort to extend the life of activated carbon by removing compounds that have been absorbed by activated carbon, thereby restoring the adsorption capacity of activated carbon to be reused. Regeneration efforts that are often carried out to date are chemical and physical regeneration. However, in a combined process between activated carbon and microorganisms, it is suspected that a bioregeneration process of activated carbon can occur so that it will extend the life of the activated carbon in addition to increasing the performance of this system.

Biological wastewater treatment processes with suspended culture systems have been widely used worldwide for domestic and industrial wastewater treatment. This process is principally an aerobic process in which organic compounds are oxidized to carbon dioxide, water, and new cell biomass. Oxygen supply is usually by blowing air mechanically through a blower or aerator. The most widely used suspended culture wastewater treatment system is the activated sludge system. Activated sludge is a complex biological mass produced when organic waste is treated aerobically. Sludge will contain a wide variety of heterotrophic microorganisms including bacteria, protozoa, and higher life forms. In other words, activated sludge is a mixture of sludge and microorganisms that have the ability to treat waste. Since the activated sludge system was first demonstrated by Edward Arden and William T. Lockett at the Davyhulme Sewage Works in Manchester, United Kingdom in 1914 [28], various modifications of the activated sludge system have been developed. But basically, it has two basic concepts, namely the biochemical stage in the aeration tank and the physical stage in the settling tank. The suspension liquid in the aeration tank in the wastewater treatment process with an activated sludge system is referred to as mixed liquor suspended solids (MLSS), which is a mixture of wastewater with microbial biomass and other suspended solids. MLSS is the total amount of suspended solids in the form of organic and mineral materials, including microorganisms.

Wastewater treatment with conventional activated sludge process generally consists of a primary settling basin, aeration basin, and a secondary settling basin, followed by a chlorination tank to kill pathogenic bacteria. In general, the treatment process is as follows, wastewater originating from pollutant sources is accommodated in an equalization tank. This equalization tank serves to regulate the discharge of wastewater which is equipped with a coarse screen to separate large impurities. The wastewater is then pumped to the primary settling tank. This primary settling basin serves to reduce suspended solids by about 30–50%, and organic content by about 25–30%. The runoff water from the primary settling basin is channeled into the aeration tank by gravity. In this aeration tank, the wastewater is aerated with air so that the existing microorganisms will decompose the organic substances present in the wastewater. The energy obtained from the decomposition of organic substances is used by microorganisms for their growth process. Thus, in the aeration tank, biomass will grow and develop in large enough quantities. This biomass or microorganism

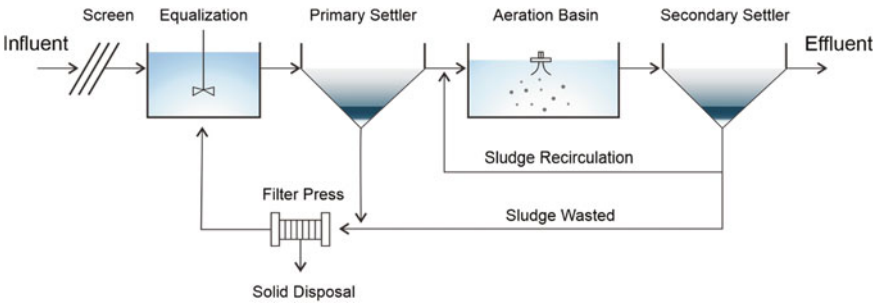

Fig. 5 Typical scheme of conventional activated sludge system

will continuously decompose the pollutant compounds present in the wastewater. From the aeration tank, the water flows into the secondary settling basin. In this tank, the activated sludge, which is a mass of microorganisms, is deposited and pumped back to the inlet of the aeration tank by a mud circulation pump. Overflow from the secondary settling basin is channeled to the chlorination bath. In this chlorine contactor tank, wastewater is contacted with chlorine compounds to kill pathogenic microorganisms. The suspended solids that settle in the secondary settling basin will be recirculated back to the aeration basin, and some of the sludge at a certain time will be disposed of as wasted sludge to control the age of the sludge in the activated sludge system (Tchobanouglous et al. 2004). Schematic of wastewater treatment process with standard or conventional activated sludge system can be seen in Fig. 5.

The biomass is separated in the secondary sedimentation tank so that it flocculates and settles. This causes bacteria, protozoa, filamentous microbe, and other microorganisms to form macroscopic flocs which will eventually settle. The attachment of these microorganisms is aided by the polysaccharide matrix produced by these microbes. An illustration of the formation of activated sludge floc can be seen in Fig. 6.

1.3 Operational Variables in Activated Sludge Process

The operational variables commonly used in wastewater treatment processes with activated sludge systems are as follows:

Hydraulic Retention time (HRT)*:* The hydraulic retention time is the average time required for wastewater to enter the aeration tank or tank [40]. For the activated sludge process, the value is inversely proportional to the dilution rate (D).

$$HRT = \frac{V}{Q} = \frac{1}{D}$$

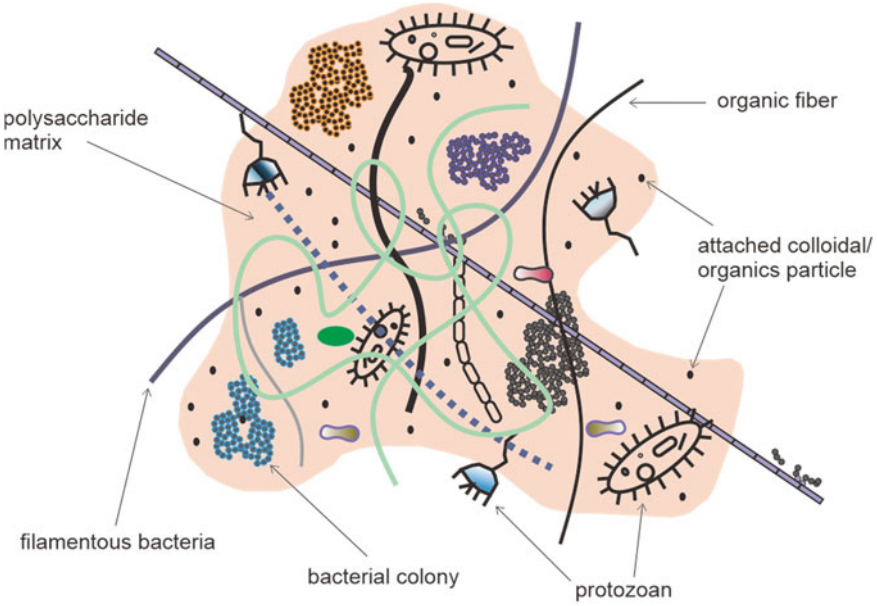

Fig. 6 Illustration of floc forming structure in activated sludge

where V is the volume of the reactor or aeration tank (m^3), Q is the discharge of wastewater entering the aeration tank (m^3/hour), and D is the dilution rate ($hour^{-1}$).

BOD Loading Rate or Volumetric Loading Rate: BOD load is the total mass of BOD in the influent wastewater divided by the reactor volume.

$$BOD\ Load = \frac{Q \times S_0}{V} \frac{kg}{m^3}.day$$

where Q is the incoming wastewater discharge (m^3/day), S_0 is the BOD concentration in the incoming wastewater (kg/m^3), and V is the reactor volume (m^3). For the conventional activated sludge process, the BOD load is generally in the range of 0.3–0.8 $^{kg}/m^3$.day, while for the extended aeration activated sludge process the BOD load is generally in the range of 0.15–0.25 $^{kg}/m^3$.day.

Microbial Degrader Concentration: The liquid suspension in the aeration tank in the wastewater treatment process with an activated sludge system are referred to as mixed liquor, which is a mixture of wastewater with microbial biomass and other suspended solids. Mixed liquor suspended solid (MLSS) is the total amount of suspended solids in the form of organic and mineral materials, including microorganisms. The MLSS was determined by filtering the mixed liquor with filter paper, then the filter was dried at a temperature of 105 °C, and the weight of the solids in the sample was weighed. The portion of organic matter in MLSS is represented by Mixed Liquor Volatile Suspended Solid (MLVSS), which contains both living and

dead microbes, non-microbial organic matter, and cell debris. MLVSS is measured by continuously heating a dried filter sample of MLSS to a temperature of 600 °C, and the value is close to 65–75% of the MLSS.

Food-to-Microorganism (F/M) Ratio: This parameter indicates the amount of organic matter (BOD) to be removed divided by the total mass of microorganisms in the aeration tank. The value of the F/M ratio is generally expressed in kilogram of BOD per kilogram of MLSS per day [44]. F/M can be calculated using the following formula:

$$\frac{F}{M} = \frac{Q(S_0 - S)}{MLSS \times V}$$

where Q is the effluent flow rate m^3 per day, S_0 is the BOD concentration in the wastewater entering the reactor area (kg/m^3), S is the BOD concentration in the effluent (kg/m^3), MLSS in (kg/m^3), and V is the volume of the reactor or aeration tank (m^3). The F/M ratio can be controlled by adjusting the circulation rate of activated sludge from the circulating final settling basin to the aeration basin and/or adjusting the wasted sludge. The higher the activated sludge circulation rate, tipically the higher the F/M ratio. For wastewater treatment with conventional activated sludge systems, the F/M ratio is 0.2–0.5 kg BOD per kg MLSS per day, but can be as high as 1.5 if pure oxygen is used (Hammer and [23, 26]. A low F/M ratio indicates that the number of microorganisms in the aeration tank is too much compared to organic compounds as food. This condition can cause starvation of healthy forming floc microbe, which is followed by several issues such as the formation of bulking, pin floc which triggers an increase in suspended solids content in the effluent.

Sludge Age: Sludge age is often called the mean cell residence time. This parameter indicates the average residence time of microorganisms in the activated sludge system. If HRT requires retention in hours, the residence time of microbial cells in an aeration tank can be days. This parameter is inversely proportional to the rate of microbial growth [67]. The age of the sludge can be calculated by the following formula:

$$Sludge\ Age = \frac{MLSS \times V}{SS_e \times Q_e + SS_w \times Q_w}$$

where V is the volume of the aeration basin (L), SS_e is the suspended solids in the effluent (mg/l), SS_w is the suspended solids in the sewage sludge (mg/l), Q_e is the effluent rate of waste (m^3/day), and Q_w is the rate of influent waste (m^3/day). Sludge age can vary between 5 and 15 days for conventional activated sludge systems. In winter, it can be longer than in summer. Important parameters controlling activated sludge operation are organic load, oxygen supply, and control and operation of the final settling basin. This final settling basin has two functions, for clarification and thickening of the sludge [7, 14, 58].

Sludge Volume Index (SVI): The conventional way to observe the settleability of a sludge is to determine the Sludge Volume Index. To determined SVI, take a mixture of sludge and wastewater (mixed liquor) from the aeration tank and put in a 1 L conical cylinder and left for 30 min, and record the volume of the sludge formed.

SVI is an indication of the volume occupied by 1 g of sludge. SVI can be calculated using the following formula:

$$SVI = \frac{SV}{MLSS} \times \frac{1000\,mg}{gram}$$

where SV is the volume of sludge deposited in a conical cylinder after 30 min of settling (ml), MLSS is mixed liquor suspended solid (mg/l). In wastewater treatment units with conventional activated sludge systems with MLSS < 3500 mg/l, typical SVI values are in the range of 50–150 ml/gr [54]. Considering that there are many operational parameters in the activated sludge process that must be controlled, the wastewater treatment process using the activated sludge process is quite complicated and requires sufficient operator expertise.

1.4 Modification of Activated Sludge Process

The development of the industrial sector will have positive and negative impacts, where one of the negative impacts that arise is the increase in the amount of waste produced. This waste will disrupt the flow process in the industry so that the waste must be removed from the industrial process and generally the waste will be discharged into the environment. However, before being disposed of, the waste must be treated properly first so that it does not have bad consequences such as damage to the surrounding environmental ecosystem due to pollution or disturbance to public welfare. The problem is that the waste generated from the production of an item will have different treatment methods and technology so that the problem of treating this waste is a complex problem. In this sub-chapter, several modifications to the process and operation unit of the activated sludge WWTP will be discussed. The wastewater treatment process that contains color pollutants that are widely used in Indonesia currently is the activated sludge process. The problem faced in the current WWTPs is that the processed water often does not meet the quality standards to comply with the new regulations in 2019 [30]. The parameter that often exceeds the quality standard is the color parameter. Several factors that are often encountered include too short in hydraulic residence time, very large fluctuations in waste discharge, poor aeration process, and operational errors due to inadequate knowledge of the operator about the process. To overcome the problems mentioned above, technological innovation is needed to improve the efficiency of wastewater treatment, especially the conventional activated sludge process. One example to improve the activated sludge process is by adding activated carbon to the aeration tank. By adding activated carbon to the aeration basin, microorganisms will multiply on the surface of the activated carbon thus increasing the density of microorganisms that decompose organic matter and dye in the aeration basin.

Granular Activated Sludge (GAS): In biological wastewater treatment systems, microorganisms can exist as biofilm and biogranule. A biofilm consists of microorganisms attached to an inert substrate, while a biogranule is a microorganism that is completely self-immobilized [36]. Biogranules cannot occur in the natural environment. Strong selective pressure is the requirement to trigger biogranulation in reactors [37]. Observations of this phenomenon were first made in the sludge of anaerobic sewage treatment systems in 1980 [35]. The first granular sludge to be detected in an aerobic system was found in 1997 in the Sequencing Batch Reactor (SBR) [45]. About two decades after the discovery, aerobic granular sludge has been studied for applications such as treatment of domestic and high organic load wastewaters, bioremediation/biotransformation of toxic aromatic pollutants (including phenol, toluene, pyridine), treatment of industrial effluents (textile, dairy, brewery), adsorption of heavy metals, recovery of high value-added products, and others [53]. In comparison to conventional activated sludge systems, GAS has several advantages, including good settling ability, high biomass retention, tolerance to toxicity, high tolerability, and higher extracellular polymer substances (EPS) production [1, 46, 53, 63]. EPS is known as bacterium secreted sticky material containing proteins, polysaccharides, humic acids, and lipids that initiate the granulation process [55]. Aerobic granulation is affected by several operational parameters, including substrate composition, organic loading rate (OLR), hydrodynamic shear force, feast-famine regime, feeding strategy, dissolved oxygen concentration, reactor configuration, solids retention time, sequential batch reactor (SBR) cycle time, settling time, and volume exchange ratio [53].

The formation mechanism of aerobic particles consists of four stages [48]. Firstly, intercellular contact with microorganisms. Secondly, under the pressure of external disturbances, the microbes react similar to Quorum sensing and gather with each other to form initial aggregates. Thirdly, microorganisms unite with each other and enhance adhesion through the production of EPS. This EPS is an extracellular polymer, which is one of the important physiological capabilities of microorganisms. It can break down molecules in the environment (in vitro) into small molecules and then take them into the body. Finally, gradually form particles through hydrodynamic shearing force (by aeration bubbles), and switch between aerobic and anaerobic cycles in a single reactor tank, as the oxygen concentration gradient decreases with the depth of the particles, so obvious stratification is gradually formed. Generally, the hydrodynamic shear caused due to bubble aeration induces extracellular polymeric substances (EPS) production, cell surface hydrophobicity, and trigger cell–cell interactions contributing to initiation of granule formation. The gel forming exopolysaccharide components of the EPS can play a role in the structural stability. Segregated distribution of microorganisms in the granular activated sludge illustrated in Fig. 7.

After the mature particles are formed, the reactions in different layers are not the same. The reaction that can be achieved by the internal anaerobic and external aerobic composition is the removal of carbon, nitrogen, and phosphorus. The outermost aerobic part is decomposed by heterogeneous bacteria for COD and nitrifying bacteria for digestion and phosphorus uptake. The excessive anoxic layer in the middle can carry out the denitrification reaction The innermost anaerobic

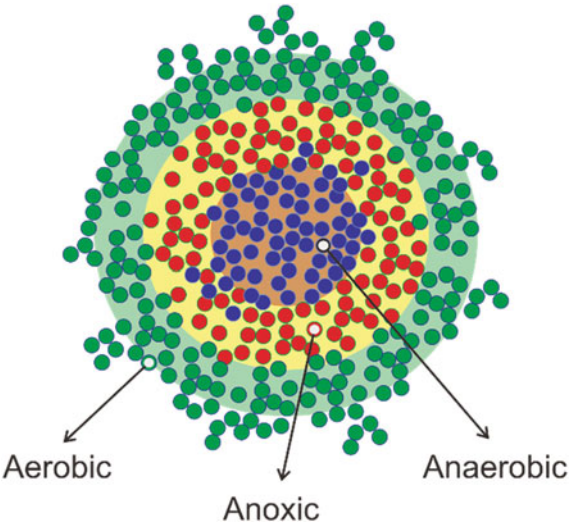

Fig. 7 Graphical representation on segregated distribution of microorganism in the individual granular activated sludge. Dyestuff in the wastewater was biotransformed to amines in the anaerobic zone, which were subsequently degraded in the anoxic–aerobic zone [9, 11, 41, 48, 71]

layer releases phosphorus. Anaerobic phosphorus release aerobic phosphorus uptake. Aerobic granular sludge (AGS) is a novel microbial community which allows simultaneous removal of carbon, nitrogen, phosphorus, and other pollutants in a single sludge system [18]. The biological dye removal process consists of 3 steps, firstly, anaerobic reduction occurs to cleave the azo dye molecule on its chromophore (-N = N-) to form colorless aromatic amine intermediates. The second is the cleavage of the aromatic ring of amine compounds into simpler aliphatic ones, and the third is mineralization in the next aerobic phase to form carbon dioxide, water, and ammonia. The granular activated sludge is then settled in the clarifier tank and due to its heavier characteristics than conventional flocs, and has a much faster settling speed (>12 m/hour), the sludge deposition process can be carried out in a smaller clarifier tank. It also allows for higher concentrations of biomass in the aeration tank, leading to more efficient degradation of wastewater [71].

Biological Activated Carbon (BAC): What is meant by biologically activated carbon is the biological process of activated sludge with a modification of the process in the form of adding a certain amount of activated carbon into a biological reactor containing microorganism culture. This combination treatment has been successfully developed by DUPONT as a PACT™ (Powder Activated Carbon Treatment) process [17, 27, 56]. This is done with the aim of improving the quality of the WWTP effluent, considering that the addition of activated carbon into the biological reactor will improve the system's performance and is more economical than the physical process with activated carbon adsorption which is carried out separately after the biological process [70]. The combined activated sludge and activated carbon system has better performance due to the following phenomena (as shown in Figs. 8, 9,10).

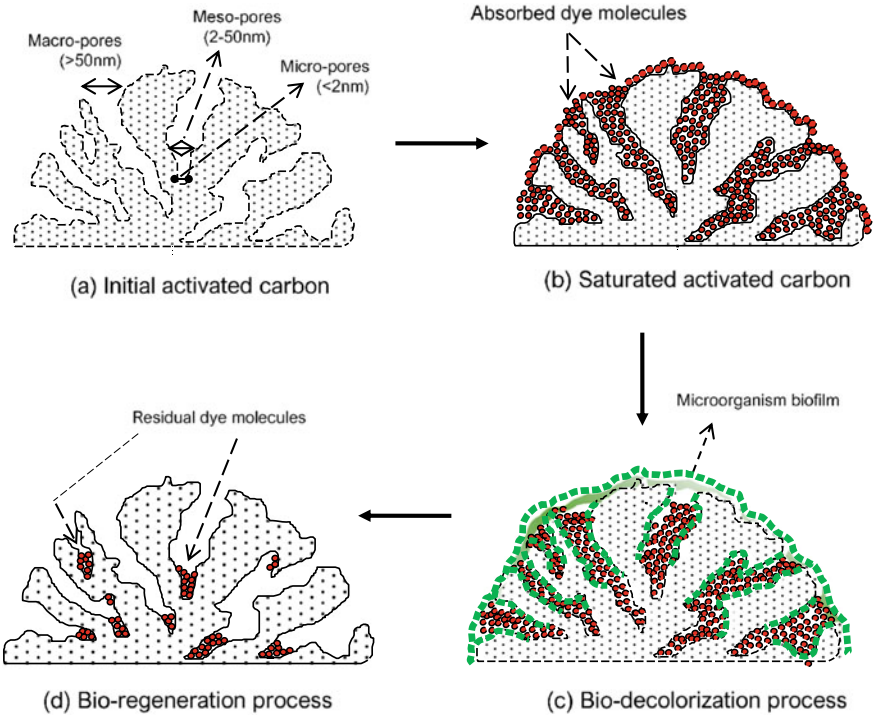

Fig. 8 The artistic scheme of the combined activated sludge and activated carbon system in the biodecolorization process of dye compounds, **a** initial activated carbon, **b** saturated activated carbon after absorbing dyes molecules, **c** breakdown of dyes molecules by microorganisms activity, and **d** bioregeneration process of activated carbon. [68, 69] Adapted from

a. Activated Carbon Adsorption. Activated carbon adsorbs dissolved substances in wastewater, including non-biodegradable substances. These non-biodegradable molecules are easier to adsorb, so their levels in wastewater are reduced.
b. Interaction Effect. Activated carbon increases microbial activity, because the surface of activated carbon becomes a matrix for microbial growth so that the rate of substrate removal increases. In addition, the absorption of the substrate on the surface of the activated carbon causes the substrate concentration to decrease. By decreasing the concentration of this substrate, the process of substrate biodegradation by microbes becomes faster.
c. Contact Optimization. Substrates that are adsorbed by activated carbon and the presence of microbes attached to the surface of activated carbon will increase the contact between the substrate and its degrading microbes.
d. Biological Regeneration Process. The presence of a substrate that is absorbed by activated carbon will be utilized by microorganisms, as a result, activated carbon becomes active again for further adsorption processes.

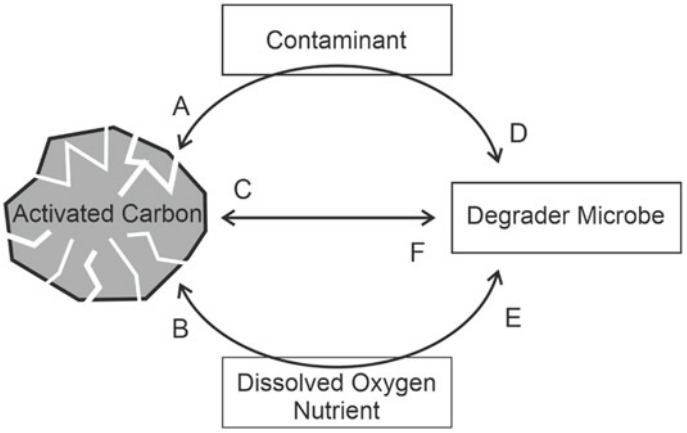

Fig. 9 Simplified relationship of four factors in biological activated carbon. Activated carbon adsorbs both dissolved oxygen and contaminants (A-B), degrader microbe utilizing contaminant and nutrient to grow on the surface of activated carbon (C-D-E), and by the decomposition of contaminants initially adsorbed by activated carbon, promote the regeneration process of the saturated activated carbon (F)

e. Microbial Flocs Core. Carbon particles have a certain density so that they can function as ballast elements and as a core for flocculation of biomass, as a result, the deposition of activated sludge flocs increases.

It can be explained that the transport of microorganisms during the bioregeneration process enters the pore structure through the macropores, mesopores, and micropores, it is possible because the size of microorganisms is smaller than the macro- and meso-pore diameters, but in the micropore structure, microorganisms cannot enter because of their size. larger microorganisms. From the test results of activated carbon that has been regenerated, the efficiency of color removal, COD, and absorption ability tends to decrease over time from the initial activated carbon [68]. The process of using activated carbon coated with a biofilm of microorganisms for water and wastewater treatment processes was developed in the 1970s. [52] examining the bioregeneration of activated carbon used to purify textile wastewater. For this case, it was operated for 11 months and it was stated that the results were good, but the mathematical bioregeneration process was not reported and the mechanism was not mentioned. Little attention has been paid to the development of biologically activated carbon regeneration research as an alternative to traditional methods of activated carbon regeneration. The experimental practice was carried out with three different treatments, namely activated carbon which was added to the biological system, the second was fine sand which was added to the activated sludge system, and the third was control in the form of activated sludge without adding other materials. In these three treatments, the growth of microorganisms was observed with the results in the first treatment, the number of microorganisms in activated sludge reached 15×10^5 colonies/cc, the second treatment resulted in 25×10^2

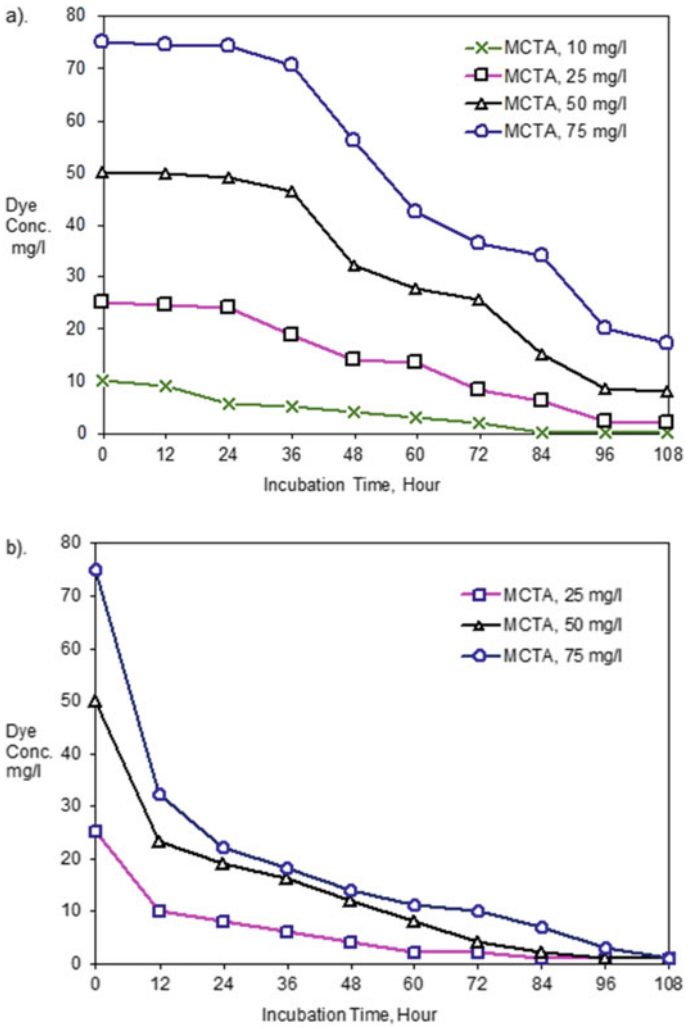

Fig. 10 Biodegradation assay of monochlorotriazine reactive red (MCTA-RR) dye for 108-h incubation time using Pseudomonas rudinensis. The presence of microorganism activity in the system caused a decrease in the concentration of MCTA dye (a), while a faster decolorization was observed in the BAC system where the color removal process was caused by both adsorption of activated carbon and due to biodegradation action of microorganisms. Adapted from [68]

colonies/cc, and the third resulted in 5×10^2 colonies/cc. Biodegradation of organic pollutants occurring in the BAC column overgrown with a large amount of aerobic biomass will result in a longer operating time than unadulterated carbon and this makes for a low cost treatment. Although the BAC process has been widely used, but the mechanism is not widely known especially the relationship between biodegradation and carbon sequestration, these two can be promoted individually or they occur

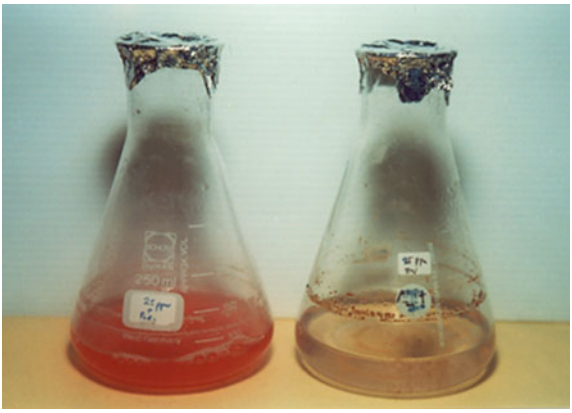

Fig. 11 Visual image of biodecolorization process of 25 mg/l monochlorotriazine reactive red dye using *Pseudomonas rudinensis* (left: control reactor; right: after 108-h incubation time)

simultaneously in the biological column of activated carbon [49, 50, 66]. The biological activated carbon process was developed based on the activated carbon process, which uses the synergistic effect of the pollutant biodegradation process by attached growth microbes and the adsorption process on activated carbon. Activated carbon is known to have a high specific surface area and a highly developed pore structure, so it is characterized by its great effect in absorbing dyestuff and organic matter in textile wastewater. In the process of Biological Activated Carbon technology, both granular and powdered activated carbon is used as a carrier, by growing or immobilizing microorganisms under proper temperature and nutritional conditions on the activated carbon surface and finally forming BAC, which can exert adsorption and biodegradable roles simultaneously. Biological activated carbon technology consists of the interaction of activated carbon particles, microorganisms, contaminants, and dissolved oxygen, in the mixed liquor in the aeration tank. Figure 9 illustrates a simplified relationship that shows how the four factors interact with each other. The relationship between activated carbon and contaminants is only the adsorption effect of activated carbon, and the reaction depends on the nature of the activated carbon and contaminants. While activated carbon can adsorb DO and microorganisms adsorbed on the surface of activated carbon, DO administration will degrade contaminants. In summary, with the interaction of these four factors, the goal of removing both dyes and contaminants from textile wastewater can be achieved using a biologically activated carbon process.

Generally, the widely adopted textile wastewater treatment is a sequential anaerobic–aerobic biological treatment process. This treatment method has proven to be the most successful yet economical in decolorizing the dye content in textile wastewater due to the breakdown of azo bonds (—N = N—) in an anaerobic process into intermediate compounds, before entering the aerobic process which then continues the mineralization of the intermediate compound into carbon dioxide and water. Currently, biological treatment for textile wastewater using microorganisms has been widely studied compared to physical and chemical treatment due to its environmental friendliness, reproducible efficiency treatment and economical implementation. BAC

can be categorized as a biofilm which is best described as a community of microorganisms attached to the activated carbon surface as a carrier. BAC can be developed by single or multispecies microorganisms which have the ability to form on both living and nonliving surfaces. The growth of microorganisms in the form of biofilms protects them from adverse environmental conditions and acts as a shield against environmental stresses, enabling communication and exchange of genetic material and nutrient availability and persistence in different metabolic states. Biofilms can have very long biomass residence times when treatment requires slow-growing organisms with poor biomass yield or when wastewater concentrations are too low to sustain activated sludge floc growth [47]. The decolorization process in this biofilm system can be similar to the decolorization mechanism in a granular activated sludge system.

2 Biological Dye Removal Process

The biological textile wastewater treatment process can be carried out under two conditions, namely aerobic conditions and anaerobic conditions or a combination of anaerobic and aerobic [72]. Aerobic biological processes are usually used for textile wastewater treatment with a low Biological Oxygen Demand (BOD) load, while anaerobic biological processes are used for wastewater treatment with very high BOD loads [32, 39]. Generally, biological wastewater treatment can be divided into three, namely biological processes with suspended culture, biological processes with attached culture and treatment processes using a lagoon or pond system. Biological process with suspended culture is a treatment system using the activity of microorganisms to decompose pollutant compounds present in the water and the microorganisms used are cultured in suspension in a reactor. Biological process with attached culture is a treatment system which the microorganisms used are cultured and attached the surface of the media. This process is also known as the microbiological film process or biofilm process. Examples of wastewater treatment technologies in this way include: trickling filters, submerged biofilters, rotary biological contact reactors, and others. The process of biological wastewater treatment with a lagoon or pond is to accommodate wastewater in a large pond with a long residence time so that with the activity of microorganisms that grow naturally, pollutant compounds present in the water will decompose [62]. The dye molecule is a combination of unsaturated organic substances with chromophores as color carriers and auxochromes as color binders with fibers. Unsaturated organic substances found in the formation of dyestuffs are aromatic compounds, including aromatic hydrocarbons and their derivatives, phenols and their derivatives, and nitrogen-containing hydrocarbon compounds. The chromophore group is the group that gives the molecule its color. Based on the source origin, dyes are divided into two, namely natural dyes and synthetic dyes. Natural dyes are natural dyes or dyes derived from plants, while synthetic dyes are aromatic hydrocarbon derivatives such as benzene, toluene, and naphthalene. The names and chemical structures of the chromophore are shown in Table 2. Based on the coloring

Table 2 Chemical structure of dye chromophore

Chemical bonding group	Chemical structure
Azo group	—N = N—
Ethylene Group	—C = C—
Carbonyl Group	—C = O
Carbon Group – Nitrogen	—C = NH; C = N—
Sulfur Carbon Group	—C = S; —C–S–S–C—
Nitroso	NO; —N – OH
Nitro	NO_2; NN–OOH

process, the dyestuffs are classified as reactive dyes, dispersion dyes, direct dyes, vat dyes, sulfur dyes, basic dyes, acid dyes, and solvent dyes.

Many studies have been carried out on the ability of microorganisms to degrade textile dyes because the dye contains organic nutrients that can be used for bacterial growth. Reports on the use of synthetic dye-degrading bacteria have been identified including: Alcaligenes eutrophus, Bacillus subtilis, Klebsiella pneumonia, Pseudomonas stutzeri, and sphingomonas sp. [61]. This dye decolorization research began with the discovery of the metabolism of mammals fed a mixture of azo dyes. The azo dyes that enter the digestive tract of these animals are reduced by the microflora in the digestive tract under anaerobic conditions. This reduced azo bond produces side products or intermediate compounds, namely amino azo benzene derivatives which are known to be carcinogens. Azo dyes reduction catalyzed by azo reductase enzymes in the liver is the same as azo reduction by microorganisms present in contamination under anaerobic conditions. From the results of these studies, further research has been developed for anaerobic digestion of dyestuffs. Furthermore, the biodegradation of dyes under anaerobic conditions is quite potential to remove textile dyes. The use of microorganisms in degrading synthetic dyes is to break the cyclic chain or the double bond of the chromophore compound. Microorganisms used to degrade bacterial dyes will produce enzymes and change the chemical structure of the pollutant to be simpler so that the level of toxicity is low. The enzymes produced by bacteria will then used to degrade dyes. The biodegradation process carried out on the dye will change the chemical structure of the chromophore or auxochrome groups through the process of reducing the N = N double bonds found in azo dyes replaced by HN-HN from NH_2.

The removal of the dyes from the textile and dyestuff manufacturing industry biologically can be broadly classified into three categories: aerobic treatment, anaerobic treatment, and a combination of both. The color removal process that occurs in textile wastewater treatment is the process of breaking the double bond in the

chemical structure of the dye. The result of breaking the dye chain is several chemical compounds as intermediate products including aromatic amine compounds. Aromatic amines are compounds that are very harmful to the environment because of their very toxic nature, so a series of intermediate product degradation processes are needed to become the final product in the form of minerals that are not harmful to the environment. The results of the research on breaking the chemical structure of color in both aerobic and anaerobic activated sludge systems are summarized in Table 3.

The textile wastewater treatment plant which is widely applied by the textile industry in Indonesia is the conventional activated sludge system. In this installation, the aerobic systems that dominate are contact aeration, step aeration, and contact stabilization. Biological treatment, aerobically or anaerobically, is generally considered the most cost- and technically effective way to remove major pollutants from complex or high-strength organic textile wastewater such as BOD, COD, and TSS. On the other hand, only the biological process treatment of the textile wastewater, may not be sufficient to meet the quality standards which over time and by increasing in environmental awareness demands tightening both the quality and quantity of wastewater to be discharged into the environment. As a case study that occurred in Indonesia, regulations regarding color parameters were not regulated before 2019, but starting in 2019, the nationally applied color parameter quality standard regulations state that the entire textile industry is required to treat color parameters with a value of no more than 200 platinum cobalt units (pt–co). Regulation of the Indonesian Ministry of Environment and Forestry Number 16, 2019 which regulates textile industry wastewater quality standards, as shown in Table 4 depicted the spirit in regulating, limiting, and improving environmental quality by tightening the concentration value of wastewater parameters.

To cope with the newly more stringent regulatory thresholds value, many textile industries have to modify or even rebuild their existing WWTPs by adapting the latest wastewater treatment technologies. The most widely applied WWTP modification is to add a decolorization unit with a coagulation flocculation process using ferrous sulfate ($FeSO_4$) and lime $Ca(OH)_2$. With the addition of these two chemicals, a $CaSO_4$ precipitate will be formed which will adsorb the dye. In addition, in the process of flocculation and deposition, this precipitate will also adsorb and trap other organic substances before being biodegraded further in the activated sludge unit. Another process modification that is generally carried out in existing WWTPs is to add an anaerobic decolorization unit since the chemical structure of the dye is a double bond in the chromophore group. Many studies suggest that the cleavage of the dye double bond will occur optimally under anaerobic conditions. However, dyestuffs can be biodegraded anaerobically although not completely, but only part of their chemical groups will be degraded. Therefore, adding an anaerobic unit will increase the efficiency of decolorization with further intermediate compound destruction taking place

Table 3 Dyes bio-decolorization in activated sludge system

Dyes	Biodecolorization treatment	Results	References
Aerobic			
Reactive Red (RR) (monochlorotriazine)	Degrader culture: Pseudomonas rudinensis	After 108 h of incubation periods, P. rudinensis able to completely decolorized RR with initial concentration of 10 mg dye/l; 92%, 84%, 77.3% color removal efficiency with initial concentration of 25, 50, 75 mg dye/l, respectively. Using biological activated carbon system with the same culture, higher color removal efficiency was observed with 96%, 98%, and 98.7% color removal with initial concentration of 25, 50, 75 mg dye/l, respectively	[68]
Acid Dyes (Acid Red-119)	Degrader culture: Bacillus thuringiensis SRDD	Bacillus thuringiensis exhibited 50–60% decolorization of 5000 ppm Acid Red-119 in 7 days of incubation. B. thuringiensis was also able to decolorized more than 98%, 92%, 95%, and 95% of C.I. Acid brown 14, C.I. Acid black 210, C.I. Acid violet 90, and C.I. Acid yellow 42 azo dyes at 100 ppm concentration in 24 h, respectively. When the developed isolate of B. thuringiensis was examined for bioremediation of actual azo dye contaminated waste it removed 70% color from the waste in 24 h	[13]
Reactive Red BS C.I. 111	Degrader culture: Pseudomonas aeruginosa NGKCTS	P. aeruginosa culture exhibited 91% decolorization of 300 ppm Reactive Red BS dye within 5.5 h over a wide variation of pH ranging from 5.0 to 10.5 and temperature from 30 to 40 ^{0}C under static conditions in the presence of either glucose, peptone, or yeast extract. The addition of 300 ppm of Reactive Red BS, in each step feeding, gave more than 90% decolorization within 2 h corresponding to 136 mg per liter per hour dye removal rate	[57]
Reactive Red 195 (Sulfonated Azo Dyes)	Degrader culture: Bacillus cereus M1 and M6	Bacillus cereus M1 and M6 were proved to decolorizing sulfonated azo dyes (Reactive Red 195) under aerobic conditions for more than 97% after 72 h of incubation. Carbon and nitrogen source used in this study is maltose and peptone	[43]

(continued)

Table 3 (continued)

Dyes	Biodecolorization treatment	Results	References
Reactive Red 198 (RR198)	Degrader culture: Aspergillus flavus	Results showed that bioremoval of RR198 by Aspergillus flavus under optimized conditions (initial dye concentration 50 ppm, pH 4, and 12 ml culture inoculum volume) increased to over 84.96% with increasing time until equilibrium was reached after a period of 24 h. A low pH was the most effective, which can be advantageous when applied to real wastewater which is generally acidic	[15]
Reactive Red 198 (RR198)	Degrader culture: Enterococcus faecalis and Klebsiella variicola	The removal efficiency of RR198 dye at an initial concentration of 10–25 mg/L was more than 98% within 72 h incubation period; however, removal efficiency was reduced to 55.62%, 25.82%, and 15.42% with initial concentrations of 50, 75, and 100 mg/L, respectively. The highest removal efficiency occurred at pH 8.0, reaching 99.26% after 72 h of incubation, occured after increasing the incubation temperature from 25 °C to 37 °C	[16]
Acid Orange 7	Degrader culture: Bacillus cereus (MTCC 9777) RMLAU1	The maximum decolorization of 68.5% from initial 100 mg dye/l was achieved at optimum pH 8.0 and 33 °C under static culture conditions during the 96-h incubation period. When using real textile wastewater, the maximum decolorization of 52.5% occurred when the system was supplemented with optimized exogenous carbon and nitrogen sources along with B. cereus augmentation. Sulfanific acid was identified as acid orange 7 dye degradation product, and other metabolic products indicated the presence of amino and hydroxyl functional groups. Researchers suggest that this strain may be suitable to be employed for in situ decolorization of textile industrial effluent under wide environmental conditions	[20]

(continued)

Table 3 (continued)

Dyes	Biodecolorization treatment	Results	References
Coomassie brilliant blue, Bromcresol purple, Congo red, and Sarranine	Degrader culture: Bacillus amyloliquefaciens W36	The optimum decolorization conditions were that the strain Bacillus amyloliquefaciens W36 was grown in a medium using 1 g/L maltose as carbon source, and 1 g/L (NH4)2SO4 as nitrogen source, supplemented with 100 mg/L different dyes at pH 6.0, incubated at 30 °C, and stirred at 200 rpm for 96 h. The bacteria could aerobically decolorize dyes, such as Coomassie brilliant blue, Bromcresol purple, Congo red, and Sarranine with an efficiency of 96 h. 95.42%, 93.34%, 72.37%, and 61.7%, respectively	[38]
Triphenylmethane Brilliant Green, Fluorone Erythrosine, Triphenylmethane Crystal Violet, Azo Evans Blue, Fluorone Bengal Rose, Azo Congo Red	Degrader culture: Mixed culture of total 31 bacteria, isolated from WWTP aeration tank	Using Kimura medium, mixed bacterial culture was multiplied for 48 h and after that fed with various dyes with an initial concentration of 100 mg dye/l and incubated at temperature 26 ^{0}C for 144 h incubation period. Color removal efficiency was achieved for triphenylmethane brilliant green (85.7%), fluorone erythrosine (78.9%), triphenylmethane crystal violet (65.5%), azo Evans blue (64.4%), fluorone Bengal rose (61.0%), and azo Congo red (57.4%), respectively	[21]
Azo dye C.I. Procion Red H-3B	Degrader culture: Pseudomonas stutzeri	Maximum decolorization of Procion Red H-3B achieved at 96%, when P.stutzeri was grown in a medium contains 50 mg dye/l, 1% fructose, and 0.5% peptone as carbon and nitrogen source with pH 7.5 and 30 ^{0}C temperature for 24 h of incubation period	[4]
Disperse Red 54	Degrader culture: Brevibacillus laterosporus	Under optimized conditions (pH 7, 40 °C), Brevibacillus laterosporus led to 100% decolorization of DR54 (at 50 mg L(-1)) within 48 h. Yeast extract and peptone, supplemented in medium enhanced the decolorization efficiency of the bacterium. Researchers identified the final biodegradation product was N-(1λ(3)-chlorinin-2-yl)acetamide, and suggested that the metabolites obtained after biodegradation of DR54 were non-toxic as compared to the untreated dye	[34]

(continued)

Table 3 (continued)

Dyes	Biodecolorization treatment	Results	References
Methanil Yellow G (MY-G)	Degrader culture: Mixed bacterial consortium ZW1 (Halomonas, 49.8%; Marinobacter, 30.7%; and Clostridiisalibacter, 19.2%)	Mixed bacterial consortium ZW1 was enriched under saline (10% salinity), alkaline (pH 8.0), and temperature (40 °C) conditions to decolorize Methanil Yellow G. Addition of yeast extract in the medium led to 93.3% decolorization of 100 mg/L MY-G within 16 h of incubation period (compared with 1.12% for control)	[24]
Acid Blue 113 (di-Azo Dye)	Degrader culture: Pseudomonas stutzeri AK6	Biodecolorization of Acid Blue 113 dye, a commonly used textile di-azo dye, has been conducted using Pseudomonas stutzeri AK6 strain. The initial dye concentration of 300 ppm was decolorized up to 86.2% within 96 h of the incubation period	[33]
Anaerobic			
Acid Red 14	Degrader culture: Oerskovia paurometabola	Decolorization batch tests with 20–100 mg/l AR14 in a synthetic textile wastewater supplemented with yeast extract indicated that Oerskovia paurometabola has a high color removal capacity for a significant range of AR14 concentrations (91% after 24 h in static anaerobic culture). Further analysis confirmed that decolorization occurred through azo bond (-N = N-) cleavage under anaerobic conditions, the azo dye being completely reduced after 24 h of anaerobic incubation for the range of concentrations tested. Another interesting research finding, partial (up to 63%) removal of one of the resulting aromatic amines (4-amino-naphthalene-1-sulfonic acid) was occurred when subsequently subjected to aerobic conditions	[19]
Alizarin Yellow R (AYR)	Degrader culture: Activated sludge microflora	AYR decolorization under anaerobic–aerobic–anoxic SBR shown the optimum removal efficiency of 85.7% and 66.8% at initial AYR concentrations of 50 and 200 mg/l, conversely higher AYR concentration of 400 mg/l indicates inactivation of the activated sludge due to the insufficient support of electron donors in the anaerobic process. Further decolorized by-products p-phenylenediamine and 5-aminosalicylic were completely decomposed in the aerobic stage of the treatment applied for 50 and 200 mg/l of initial dye	[10]

(continued)

Table 3 (continued)

Dyes	Biodecolorization treatment	Results	References
Alizarin Yellow R (AYR)	Degrader culture: Activated sludge microflora	A four-compartment anaerobic baffled reactor (ABR) incorporated with membraneless biocatalyzed electrolysis system (BES) was examined for the treatment of azo dye AYR with initial concentration of 200 mg/l. The decolorization efficiency in the ABR-BES (8 h HRT, 0.5 V) was found higher than that in ABR-BES without electrolysis (95.1 ± 1.5% compared to 86.9 ± 6.3%), while higher power supply (0.7 V) give higher efficiency up to 96.4 ± 1.8% and VFAs removal	[12]
Acid Orange 7 (AO7)	Degrader culture: Activated sludge microflora	Two-stage anaerobic system (acidogenic and methanogenic phase) were used to investigate the removal of an azo dye AO7 (2.14 mM initial concentration), with starch (1 g/l) as the primary co-substrate. Research results discovered that under 5 days HRT, the methanogenic phase accounted for about 90% of the entire AO7 removal, and the obtained removal rate constant for AO7 was 2.93-fold higher of that in the acidogenic phase. Effluent from the acidogenic phase containing readily available electron donors was fed as the influent for the methanogenic phase, in which AO7 was preferred to be reduced	[29]
Acid Orange 7 (AO7)	Degrader culture: Non-adapted methanogenic granular sludge microflora	Fed-batch and continuous reactor under anaerobic conditions was used to investigated the decolorization of AO7 with loading rate of 1.7 mM/day (590 mg/l.day), and fed with glucose (2 g/l) as co-substrate able to remove 92% AO7. It was noticed that when the co-substrate was reduce (AO7 0.3 mM and glucose 0.25 g/l, AO7 removal efficiency was decreased significantly to 78%	[42]
Acid Orange 6 (AO6) Acid Orange 7 (AO7)	Degrader culture: Anaerobic sludge from a full scale UASB plant microflora	Sequential fixed-film anaerobic batch reactor (SFABR) was used to investigated decolorization of two azo dyes (AO6 and AO7), with varying initial dye concentration and co-substrate (0.5 g/l glucose). At 300 mg/l AO6 and AO7, more than 90% efficiency was achieved with removal rates were 168 mg/l.day and 176 mg/l.day, respectively. Degradation by-products identified as 4-aminobenzenesulfonate and Aminoresorcinol were discovered to be resistant to further degradation under anaerobic environment	[59]

(continued)

Table 3 (continued)

Dyes	Biodecolorization treatment	Results	References
Acid Orange 7 (AO7) Direct Red 254 (DR254)	Degrader culture: Mixed anaerobic bacterial consortia	The anaerobic treatment of a monoazo dye (AO7) and a diazo dye (DR254), was investigated in a methanogenic laboratory-scale Upflow Anaerobic Sludge Blanket (UASB), fed with acetate as primary carbon source. A color removal achieved more than 88% for both dyes at a HRT of 24 h, while at a HRT of 8 h, a more extensive reductive decolorization was observed for DR254 (82%) compare to AO7 (56%). Research results suggested that methanogenic cultures predominantly to perform azo bond cleavage	[6]
Reactive Orange 16 (RO16)	Degrader culture: Granular sludge from a full scale UASB plant microflora	Submerged anaerobic membrane bioreactors (SAMBRs) was used to investigated the decolorization of azo dyes containing textile wastewater and operated for almost four months with increasing RO16 concentration from 0.060 to 3.2 g/l. The results indicated that high removal efficiency of 99% was achieved by SAMBRs even fed with high concentration (3.2 g/l) of dye	[60]
Yellow Gold Remazol (YGR)	Degrader culture: Anaerobic sludge from a pilot scale UASB reactor microflora	Two continuous bench-scale treatment were used to investigated decolorization of Yellow Gold Remazol. Upflow Anaerobic Sludge Blanket followed by an activated sludge (UASB/AS) system and UASB followed by a shallow polishing pond (UASB/PP) were fed with 50 mg/l YGR and 350 mg/l pretreated residual yeast, as co-substrate. Results indicated that the UASB/PP system achieved the highest removal of YGR and chemical oxygen demand with efficiency of 23% and 85%, respectively	[2]
Reactive Red 2 (RR2)	Degrader culture: Mixed culture containing Desulfovibrio aminophilus, Thermoanaerobacter, Lactococcus raffinolactis, Ruminiclostridium and Rhodopirellula	An integrated hydrolysis/acidification (HA) and multiple anoxic/aerobic (AO) process was used to investigated the removal of RR2 and nitrogen from azo dye containing wastewater. The RR2 and nitrogen removal percentages of the HA and AO process with HRT of 12 h, treating initial concentration of 30 mg/l RR2 and 114 mg/l NH4Cl were 89.4% and 54.0%, respectively	[22]

(continued)

Table 3 (continued)

Dyes	Biodecolorization treatment	Results	References
Methyl Orange (MO)	Degrader culture: Denitrifying anaerobic methane oxidator microflora (Methanomethylovorans, Moranbacteria)	A methane-based hollow fiber membrane bioreactor (HfMBR) inoculated with an enriched anaerobic methane oxidation (AOM) culture was used to investigated the decolorization of MO at various initial concentration of 400–800 mg/l at temperature 35 °C, and HRT ranging from 12 to 48 h. Results showed the MO decolorization achieved ranging from 88 to 100% with maximum decolorization rates of 883 mg/l.day	[3]
Remazol Brilliant Blue R (RBBR)	Degrader culture: Mixed culture containing Proteobacteria, Spirochaetae, Aminicenantes, Bacteroidetes, Thermotogae, and Chloroflexi	An anaerobic dynamic membrane bioreactor (AnDMBR) was used to investigated the RBBR decolorization operated for a total of 120 days in three stages at mesophilic environment of 37 °C. Initial concentration of RBBR ranging from 0.5 g/l (stages I–II) to 1 g/l (stage III), with addition of 4.5 g/l glucose as co-substrate (stages I–II). The AnDMBR was run under hydraulic retention time (HRT) of 5 and 2.5 days, while organic loading rates in stages I–III were kept at 5.0, 5.0, and 1.0 g COD/l.day, respectively. Results showed satisfactory for soluble COD and RBBR removal efficiency of 98.5% and 97.5%, respectively	[5]
Reactive Yellow 15 (RY15)	Degrader culture: Mixed culture collected from Activated Sludge WWTP containing Bacillus, Pseudomonas, and E-Coli	Immobilized mixed cells using biocarrier of sodium alginate (SA), starch (St), and Gelatin (Ge) cross-linking with polyvinyl alcohol (PVA) were used to investigated decolorization of RY15 via a sequential anaerobic–aerobic process. Results showed complete decolorization (100%) of RY15 was occurred, and COD removal were 92% ± 6.8, 96% ± 3.5, and 100%, using PVA-SA, PVA-St, and PVA-Ge at RY15 initial concentrations of 10 mg/l, under the overloading rate (OLR) and Hydraulic retention time (HRT) of the aerobic bioreactor are 24.5 mg/l.hour and 41.37 h, respectively	[25]

Table 4 Textile industry wastewater quality standards in Indonesia [30]

Parameters	Unit	Threshold value		
Flowrate	m^3/day	≤ 100	100 < F < 1000	≥ 1000
BOD	mg/l	60	45	35
COD	mg/l	150	125	115
TSS	mg/l	50	40	30
Total phenol	mg/l	0.5	0.5	0.5
Total chromium	mg/l	1	1	1
Total ammonia	mg/l	8	8	8
Sulfide	mg/l	0.3	0.3	0.3
Oil & grease	mg/l	3	3	3
pH	–	6–9	6–9	6–9
Color	Pt–Co	200	200	200
Temperature	°C	Dev. 2°	Dev. 2°	Dev. 2°
Maximum flowrate	m^3/ton product	100	100	100

in the existing aerobic unit. Schematic process flow diagram of several suggested WWTPs modification is described in Figs. 12, 13.

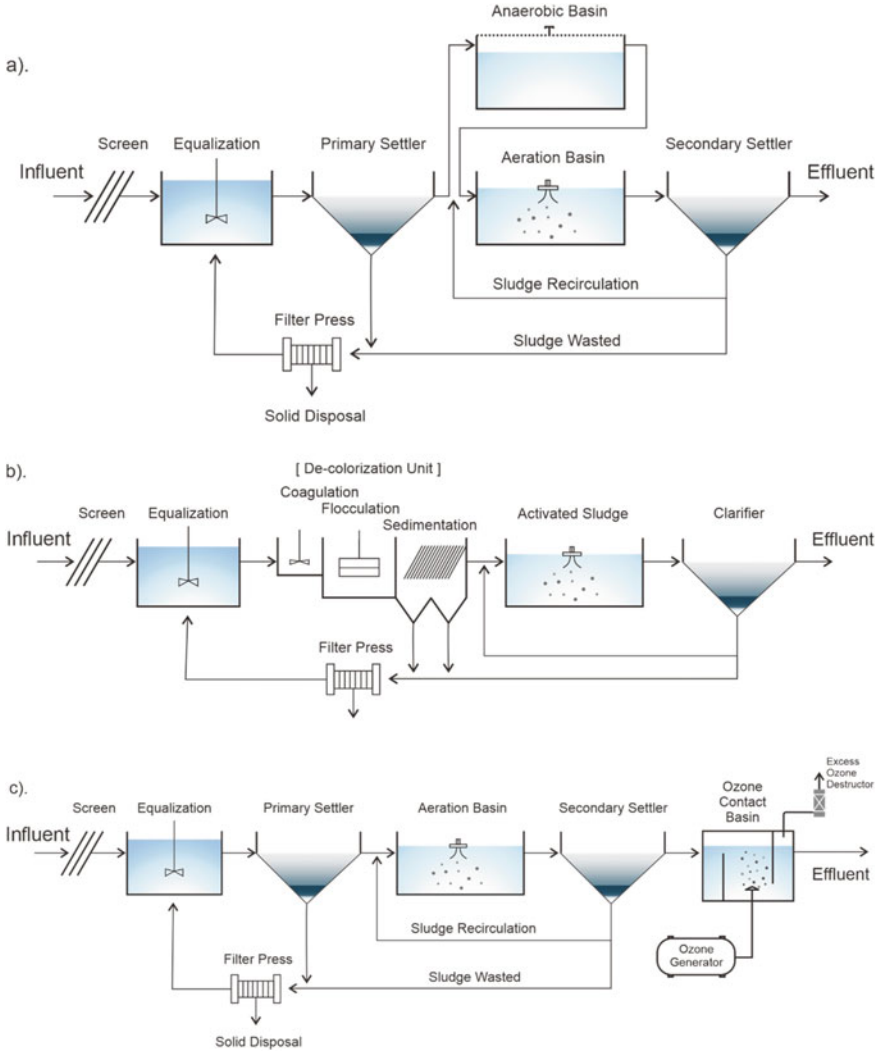

Fig. 12 Schematic process flow diagram of commonly practiced in the modification of existing WWTP to cope with newly stringent regulatory threshold value in textile wastewater by simply adding anaerobic unit before aeration basin (**a**), installing decolorization unit using coagulation-flocculation process (**b**), and installing ozone unit for color destruction as pre- and/or post-treatment (**c**)

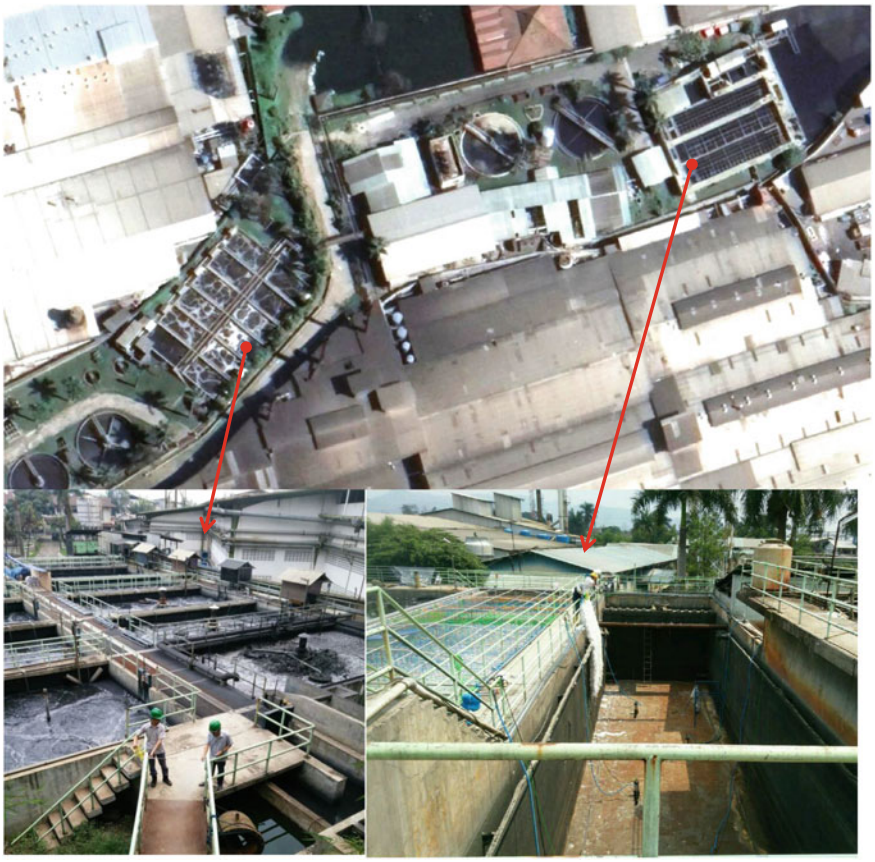

Fig. 13 WWTP Cisirung (aerial view, upper), an integrated communal wastewater treatment plant located in Bandung, West Java, Indonesia, processes up to 175 L per second wastewater collected from 22 nearby textile industries. To cope with newly stringent regulatory threshold value in textile wastewater, WWTP operator company made modification by simply adding anaerobic unit (lower right) before aeration basin (lower left)

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