DEVELOPMENT OF AEROBIC GRANULAR SLUDGE FOR REMOVAL OF POLLUTANTS FROM TEXTILE WASTEWATER

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DEVELOPMENT OF AEROBIC GRANULAR SLUDGE FOR REMOVAL OF POLLUTANTS FROM TEXTILE WASTEWATER

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CERTIFICATION

The Thesis titled "Development of Aerobic Granular Sludge for Removal of Pollutants from Textile Wastewater" submitted by Fatema-Tuz-Zohra, Student No: 1018042508, Session: October, 2018, has been accepted as satisfactory in partial fulfillment of the requirements for the degree of MASTER OF SCIENCE IN ENVIRONMENTAL ENGINEERING on 17th June, 2023.

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DECLARATION

I hereby declare that the research contained in this thesis was conducted by myself and that I have not provided this work to any other parties for any other reason (apart from publishing).

June, 2023

Fatema-Tuz-Zohra

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ABSTRACT

The textile industry is renowned for its significant contribution to global economic growth. However, the textile manufacturing processes generate vast quantities of wastewater containing various pollutants that pose substantial environmental and health risks. Traditional wastewater treatment methods struggle to efficiently remove the complex and diverse contaminants like color, organic pollutant and suspended particles found in textile wastewater. Therefore, alternative treatment technologies, such as aerobic granular sludge (AGS), have gained increasing attention due to their potential for enhanced pollutant removal and process stability in the field of textile sectors.

This study presents a comprehensive investigation of the application of aerobic granular sludge for the treatment of textile wastewater. The primary objective of this study is to assess the performance and feasibility of AGS systems to achieve efficient and sustainable textile wastewater treatment. In this study, wastewater from a knit dyeing textile industry was collected to use as raw textile wastewater (RTWW) for feeding in the AGS reactor. The wastewater quality of RTWW was analyzed in the environmental laboratory and found average data for color ~3300 Pt-Co @ 455 nm, COD ~1000 mg/L and TSS ~120 mg/L. The geometry for the AGS reactor was selected at H/D of 5.0 and operational philosophy for AGS reactor was followed by steps of sequential batch reactor (SBR). The AGS reactor was initially fed with 1000 mL of RTWW and 1000 mL of activated sludge as seed. The activated sludge was collected from the aeration tank of a textile industry effluent treatment plant where the MLSS was 4600 mg/L at the starting of the reactor operation.

In this study, total fifty-eight cycles were conducted in the AGS reactor including granulation phase, reactivation phase after idle period and An-Ae SBR operation phase. Hydrodynamic shear force at 2.4 cm/s ~ 3.6 cm/s and short settling time at 5 minutes ~ 3 minutes were considered as selection pressures for development of AGS from seed flocculant sludge. This investigation found the successful growth of AGS in the batch reactor, with granule sizes ranging from 1 mm to 3 mm. The SEM analysis was also performed in this study to examine the morphology of the granules, where it was evident that granular sludge and flocculant sludge differed greatly from one another.

The effluent treated by AGS was examined in BUET environmental laboratory to assess the potential of AGS for treating real textile wastewater. The tests were done in different

conditions of operation including granulation phase, reactivation phase after idle period and An-Ae SBR operation phase. The results were observed more than 70% removal of color, COD and more than 95% removal of TSS from RTWW. The AGS capability to treat RTWW was also examined after resuming from the idle period lasted for two months. The results of color, COD and TSS removal were found as good as earlier of idle phase. In the study, the elimination of contaminants from textile wastewater using the AGS granules was also demonstrated by the FTIR spectra analysis.

TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

LIST OF ABBREVIATIONS

ACF	Activated Carbon Filter
ADMI	American Dye Manufacturing Index
AGS	Aerobic Granular Sludge
An-Ae	Anaerobic Aerobic
APHA	American Public Health Association
ASP	Activated Sludge Process
BOD	Biochemical Oxygen Demand
C	Celcius
CAS	Conventional Activated Sludge Process
CETP	Central Effluent Treament Plant
cm	Centimeter
cm/s	Centimeter Per Second
COD	Chemical Oxygen Demand
DO	Dissolved Oxygen
DoE	Department Of Environment
EC	Electrocoagulation
ECR	Environment Conservation Rules
EPS	Extracellular Polymeric Substances
ETP	Effluent Treament Plant
F/M	Food To Microorganism Ratio
FTIR	Fourier-Transform Infrared Spectroscopy
GDP	Gross Domestic Product
H/D	Height To Diameter Ratio
HLR	Hydraulic Loading Rate
HMI	Human Machine Interface
HR	High Range
HRT	Hydraulic Retention Time
LR	Low Range
MBR	Membrane Bioreactor
mg/L	Milligram Per Liter
MGF	Multi Grade Filter
ml	Milliliter
MLSS	Mixed Liquor Suspended Solid
mm	Millimeter
OLR	Organic Loading Rate
pcs	Pieces
PLC	Programmable Logic Controller
rpm	Rotation Per Minute

RTWW	Raw/Real Textile Waste Water
s	Second
SBBGR	Sequencing Batch Biofilter Granular Reactor
SBR	Sequential Batch Reactor
SCADA	Supervisory Control And Data Acquisition
SEM	Scanning Electron Microscopy
SOUR	Specific Oxygen Uptake Rate
SS	Suspended Solid
STWW	Synthetic Textile Waste Water
TDS	Total Dissolved Solid
TSS	Total Suspended Solid
TWW	Textile Waste Water
UASB	Upflow Anaerobic Sludge Blanket
VER	Volume Exchange Ratio
WW	Waste Water
WWTP	Waste Water Treatment Plant

Chapter 1

INTRODUCTION

1.1 General

Textile industries is one of the largest water users in Bangladesh. It is the largest exporting industry in Bangladesh, having experienced phenomenal growth in last few decades (Hasan et al., 2016). The sector creates about 4.2 million employment opportunities and contributes significantly to national GDP (Gross Domestic Product) being the world's second largest exporter of clothing after China (Islam et al., 2013). The garment sector now contributes about 77% of the country's foreign exchange earnings and 50% of its industrial work force. Its exporting incomes were US $ 17.9 billion in the year 2010-11, US $ 19.0 billion in the year 2011-12 and US $ 21.5 billion in the year 2012-13 (www.bgmea.com.bd), the share of textile sector is almost 80%. Bangladesh has earned "Brand Bangladesh" in readymade garment products exporting in the world, gratitude goes to the Multi-Fiber Agreement (MFA), and the Generalized System of Preferences (GSP) of the European Union, that conferred significant quota benefits to the country.

The dark side of this highly revenue generated sector is environmental pollution. The main environmental problems associated with textile industry are typically those associated with water body pollution caused by the discharge of untreated effluents (Dey and Islam, 2015). Textile industries consume high volumes of water per unit fabric for processing, which cause depletion of ground water levels at a high rate. In addition, in many cases textile effluents are discharged into rivers or wetlands without proper treatment. One of the studies found that in 2016, textile industries in Bangladesh produced about 1.80 million metric tons of fabric, which generated around 217 million m^3 of wastewater (2016) containing a wide range of pollutants (Hossain et al., 2018). Another study showed that average sized textiles mills consume water about 200 L per kg of fabric processed per day (Wang et al., 2011) and (Kant, 2012). Also, according to the World Bank estimation, textile dyeing and finishing treatment given to a fabric generates around 17 to 20 percent of industrial waste water (Kant, 2012; Holkar et al., 2016).

The textile effluent contains high amounts of polluting agents causing damage to the environment and human health including suspended and dissolved solid, biological oxygen demand (BOD), chemical oxygen demand (COD), chemicals, odor and color (Dey and Islam, 2015).Therefore, before discharge of textile wastewater into river, many treatment processes including chemical, bio-chemical, physical, hybrid treatment processes have been developed to treat it in an economic and efficient way. Though these technologies are verified to be highly effectual for the treatment of textile wastewater (Kumar and Bhat, 2012), only biological technologies are matter of attraction to researchers due to its sustainability for environment. All pollutants cannot be found to be removed efficiently from textile waste water by biological method, especially complex compound of dye materials and other inorganic components. The textile industry accounts for the largest consumption of dyestuffs at nearly 70 percent.

1.2 Background and Statement of Problem

In light of treatment technology for the textile wastewater different types of method are widely used including physio chemical, chemical and biological technology. Wastewater treatment plants utilizing biological methods, rather than chemical methods claim that their preference is due to low production of inorganic sludge, low operating costs and complete mineralization/stabilization of dye in biological method (Navin et al., 2018). This biological technology is used to remove COD, BOD_5, suspended solids and inorganic dyes from wastewater generated by textile industries. Synthetic dyes are designed to have high durability, which gives them high stability in water and a recalcitrant nature, resistant to biodegradation. Therefore, conventional biological wastewater treatment processes fail in efficiently removing textile dyes from wastewaters, potentially leading to their long term persistence in natural water bodies (Santos et al., 2007). In recent years, the ability of biodegradation for textile dyeing wastewater and dyestuffs involving both anaerobic and aerobic processes has been widely reported in the literature (Muda et al., 2010; Toorisaka et al., 2005; Isik et al., 2008 and Franciscon et al., 2009).

Bacterial azo dye biodegradation generally proceeds in two stages: (Dey and Islam, 2015) the anaerobic phase, responsible for color removal through reductive cleavage of the azo bond, resulting in the formation of generally colorless, but potentially hazardous, aromatic amines; the aerobic phase involving further degradation of aromatic amines (Van der and Villaverde, 2005). Azo dyes account for more than 70% by weight of all dyestuffs used worldwide (Rawat et al., 2016) and represent the largest class of synthetic colorants applied in textile processing (approximately 80% by weight), consequently being the most commonly released into the environment (Saratale et al., 2011; Singh et al., 2015). Although azo dyes are generally considered as xenobiotic compounds and recalcitrant to aerobic biodegradation (namely during treatment with conventional activated sludge, CAS), several microorganisms are able to transform azo dyes into colorless breakdown products (partial biodegradation), or even to completely mineralize these metabolites (total biodegradation), under certain environmental conditions (Pearce et al., 2003; Stolz 2001). Since complete dye degradation requires both anaerobic and aerobic conditions, several studies have been focused on treatment systems which utilized two different reactors to fulfill both conditions (Toorisaka et al., 2005; Moosvi and Madamwar, 2007; Isik et al., 2008). The operation of such a system is rather complicated as the anaerobic microorganisms in the anaerobic tank need to be separated before the wastewater can be pumped to the aerobic tank. To simplify the system, some studies were done on integrated anaerobic aerobic bioreactors, where aerobic and anaerobic conditions are combined in a single reactor can generally enhance the overall degradation efficiency, are cost effective and have reduced footprints (Franca et al., 2020). This kind of reactor is generally applied in sequential batch reactors (SBR) and previous studies have investigated the suitability of sequencing batch reactors (SBR) for textile wastewater treatment (Fu et al., 2001; Pasukphun and Vinitnantharat, 2003; Sirianuntapiboon et al., 2006) as well.

Therefore, researchers developed microbial granular sludge that can survive and function in both anaerobic and aerobic conditions and hence requires only one reactor. Potential strict anaerobic microorganisms can survive easily since oxygen only penetrates partially in the granules during aerated phase of the process (Muda et al., 2010). Biogranulation technology for wastewater treatment includes anaerobic and aerobic granulation processes. Both aerobic and anaerobic granulations involve cell-to-cell interactions that contain biological, physical and chemical phenomena. Through self-immobilization of microorganisms, granules are formed which consist of various bacterial species. These bacteria are expected to play different roles in degrading wastewater containing various organic chemicals and removing of nutrients. As compared to conventional activated sludge flocs, granular sludge has regular, denser and stronger microbial structure and good settling ability. These characteristics result in high biomass retention and withstand high-strength wastewater and shock loadings. Instead of the same characteristics of the formed granules for both anaerobic and aerobic granulation, these two technologies of granulation are not same in terms of process feasibility. The limitations of anaerobic granulation technology include the need for a long start-up period, a relatively high operation temperature and unsuitability for low strength organic wastewater. Anaerobic granulation technology is not suitable for the removal of nutrients from wastewater as well. In order to overcome those weaknesses, research has been devoted to the development of aerobic granulation technology.

Aerobic granulation has been observed in sequencing batch reactors (SBRs) (Liu et al., 2002; Beun et al., 1999). It has been utilized in treating high-strength wastewaters containing organics, nitrogen and phosphorus, and toxic substances (Jiang et al., 2002; Moy et al., 2002). However, the granulation process of aerobic sludge has not been well studied. Owing to achieve environment friendly biological technology for removing dyes and recalcitrant pollutants from textile wastewater, few researchers recently worked on aerobic granular sludge (AGS) technology. The aerobic granular sludge (AGS) technology, considered the next generation of wastewater treatment, represents a potential solution due to the high biomass retention capacity, anaerobic/anoxic/ aerobic microenvironments within granules and enhanced tolerance towards high organic loads and toxic compounds (Franca et al., 2020). Moreover, AGS technology seems highly promising to remove textile dyes from wastewaters because (Franca et al., 2020):

a. Integrated anaerobic-aerobic condition in a single reactor generally enhance the overall degradation efficiency.
b. Cost effective due to not having separate tanks for aerobic and anaerobic reaction.
c. Reduce footprint than other conventional biological process.

Most of the studies on the suitability of AGS technology for textile wastewater treatment were done by synthetic wastewater only. Though, few works were found on raw textile wastewater (RTWW), some additions were done to provide nutrients to biomass or specific sludge were used as seed to acclimatize AGS. Therefore, needs to do more work on the operational condition, AGS stability, resistance of AGS to prolonged idle periods and its reactivation performance to achieve desired removal efficiency in dye and organic load removal from RTWW.

1.3 Objectives with Specific Aims

1. Development of Aerobic Granular Sludge (AGS) in sequential batch reactor (SBR) targeting for a compact solution to treat tough wastewater of textile industry.

2. Determination of the AGS reactor efficiency for the removal of color, organic load (COD) and total suspended solid (TSS) from raw textile waste water.
3. Evaluation of AGS resistance to prolonged idle periods and its reactivation performance, owing to the discontinuous production processes and irregular wastewater discharges typical of the wet processing textile industry.

1.4 Scope of the Study

The scope of this study includes the establishment of a cost effective and operation friendly fully biological technology for treatment of highly polluted textile wastewater. The main focus of this study is only on the development of process and technology for AGS in the field of RTWW reviewing consecutive research works from last few years. This study may have the limitation to meet local government standard for wastewater parameters to discharge in the environment. In this case this work may be useful in future research works for further studies on AGS technology and its feasibility to treat highly polluted industrial wastewater like textile, tannery or others.

1.5 Thesis Outline

The thesis study consists of following sections:

Chapter one: This chapter describes importance of study on textile wastewater, the background of the study, previous works on similar research like aerobic granular sludge and its progress, objectives and limitation of the study within its scope.

Chapter two: The chapter two represents a brief of textile processing, contribution of water pollution from each step of textile process, the impact of pollution on environment caused by textile wastewater, proven treatment technologies for textile wastewater and their pros-cons, recent works on development of treatment technology for textile wastewater and latest development on AGS work for removing pollutants of textile wastewater.

Chapter three: This chapter contains process methodology, the experimental works details and preparation work for conducting this study. The experiment includes all the steps from the cultivation of aerobic granular sludge (AGS) to water parameter analysis for checking the efficiency of AGS to treat RTWW.

Chapter four: This chapter will explain the results and findings from entire experimental time. Related data and reports will be explained in this section and accordingly expected outcomes from the experiment will be analyzed.

Chapter five: This chapter will conclude the full study including results of the work and will recommend future provision of work on this subject.

Chapter 2
LITERATURE REVIEW

2.1 Introduction

This chapter will discuss on details of textile process for generating industrial wastewater, environmental impact for TWW pollution, different technologies for treatment of TWW, AGS technology application for treatment of industrial WW, advantages of AGS technology over ASP system and recent works on AGS for treatment of TWW.

2.2 Textile Process and Wastewater Generation

The textile industries are distinguished by the use of raw materials which determines the volume of water required for the process as well as wastewater generated (Dey and Islam, 2015). The major raw material for textile processing is grey fabric and most of materials consist of cotton and blended fabric (ShohidullhMiah, 2012).Generally such kind of textile processes include pretreatment, dyeing/printing, finishing and other technologies. Pre-treatment includes desizing, scouring, washing, and other processes. Dyeing mainly aims at dissolving the dye in water, which will be transferred to the fabric to produce colored fabric under certain conditions. Printing is a branch of dyeing which generally is defined as 'localized dyeing' (Praveen et al., 2018). In addition, in different circumstances, the singeing, mercerized, base reduction, and other processes should have been done before dyeing/printing (Praveen et al., 2018). All of these processes are the major source of water consumption as well as wastewater generation. One kg of fabric requires 50 to 2600 L of water on average ((Madhav et al. 2018; Bento et al. 2020). However, finishing and garment processing require comparatively less water consumption than the other processes, below is the processing steps for typical textile dyeing and printing industry showing the involved pollutants for each step in Figure 2.1 (Hannan et al., 2011).

During each stage different type of chemicals are used such as strong acids, strong alkalis, inorganic chlorinated compounds, hypochlorite of sodium, organic compound such as dye stuff, bleaching agent, finishing chemicals, starch, thickening agent, surface active chemicals, wetting and dispensing agents and salts of metals (Dey and Islam, 2015). The qualitative analysis of the pollutants contribution for each step is described in below Table 2.1 (EPA, 1998).

2.2.1 Effluent Characteristics from Textile Industries

Farjana et al., 2014 reported the characterization of untreated Textile effluent as, BOD_5 96-242 mg/L, COD 225-800 mg/L, TDS 228-2040 mg/L, TSS 15-110 mg/L and color 382-205 (Pt-Co Unit). In another study it was found that at Savar the pH varies 7.3-11.2, TDS 460-5981 mg/L, DO 0-6 mg/L, COD 118-2304 mg/L and BOD 60-461 mg/L whereas our national standard is for pH 6-9, TDS 2100 mg/L, DO 4.5-8 mg/L, COD 200 mg/L and BOD_5 50 mg/L respectively (Dey and Islam, 2015). Not only in Savar but also the scenario of the effluent characteristics in greater Dhaka are almost same. 33% of industries are located in the Dhaka district and 32% are in Narayanganj. There were 298 polluting textile mills listed by DoE in 1986, which is now 365 in number (Monroy et al., 2013).

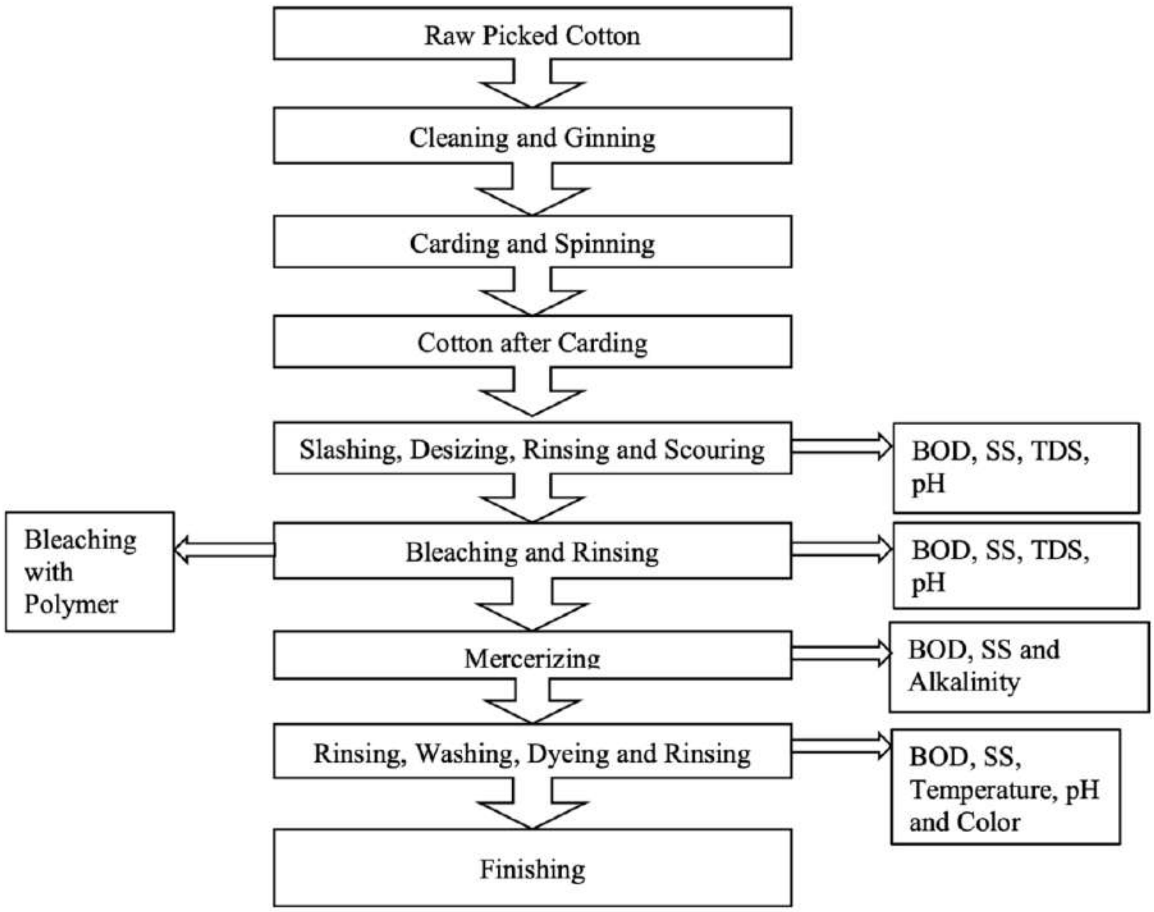


Figure 2.1: Different steps of typical dyeing and printing textile industry (Hannan et al., 2011).

2.2.2 Types of Dyes in Textile Industries

The most significant physicochemical characteristic, color, is obtained during the textile's washing process.

A colored substance that may transmit its distinct colors is referred to as a dye or dyestuff. When applied to materials, a good dye exhibits good fastness to washing, light, and heat. Dyes contain chromophores and auxochromes that are accountable for their substantiveness and color. The majority of textile dyes are synthetic chemical compounds with an aromatic structure, and it is always due to these synthetic dyes that light penetration is reduced and photosynthesis in aquatic ecosystems is disrupted.

Table 2.1: Qualitative details in each step of textile processing (EPA, 1998)

Process	Effluent Composition	Pollutant Nature
Sizing	Starch, waxes, Carboxymethyl Cellulose (CMC), Polyvinyl Alcohol (PVA), wetting agents.	High in BOD, COD
Desizing	Starch, CMC, PVA, fats, waxes, pectins	High in BOD, COD, SS, dissolved solids (DS)
Bleaching	Sodium Hypochlorite, Cl2, NaOH, H2O2, acids, Surfactants, NaSiO3, Sodium Phosphate, short cotton fibre	High alkalinity, high SS
Mercerizing	Sodium Hydroxide, cotton wax	High pH, low BOD, high DS
Dyeing	Dyestuffs Urea, reducing agents, oxidizing agents, Acetic acid, detergents,	strongly colored, high BOD, DS, low SS, heavy
Printing	Pastes, urea, starches, gums, oils, binders, acids, Thickeners, cross-linkers, reducing agents, alkali	Highly colored, high BOD Oily, appearance, SS, slightly alkaline

All aromatic compounds absorb electromagnetic energy, but only those that do so in the visible light spectrum (between 350 and 700 nm) exhibit color. Dyes comprise auxochromes, electron-withdrawing or electron-donating substituents that produce or amplify the color of the chromophore by changing the total energy of the electron system, as well as chromophores, delocalized electron systems with conjugated double bonds. Usual chromophores are -C=C-, -C=N-, -C=O, -N=N-, $-NO_2$ and quinoid rings, usual auxochromes are $-NH_3$, -COOH, $-SO_3H$ and -OH. Based on chemical structure or chromophore, 20-30 different groups of dyes can be discerned. Azo (monoazo, disazo, triazo, polyazo), anthraquinone, phthalocyanine and triarylmethane dyes are quantitatively the most important groups. Other groups are diarylmethane, indigoid, azine, oxazine, thiazine, xanthene, nitro, nitroso, methine, thiazole, indamine, indophenol, lactone, aminoketone and hydroxyketone dyes and dyes of undetermined structure (stilbene and sulphur dyes).

Dyes can be classified in to different types depends on various parameters as following Figure 2.2.

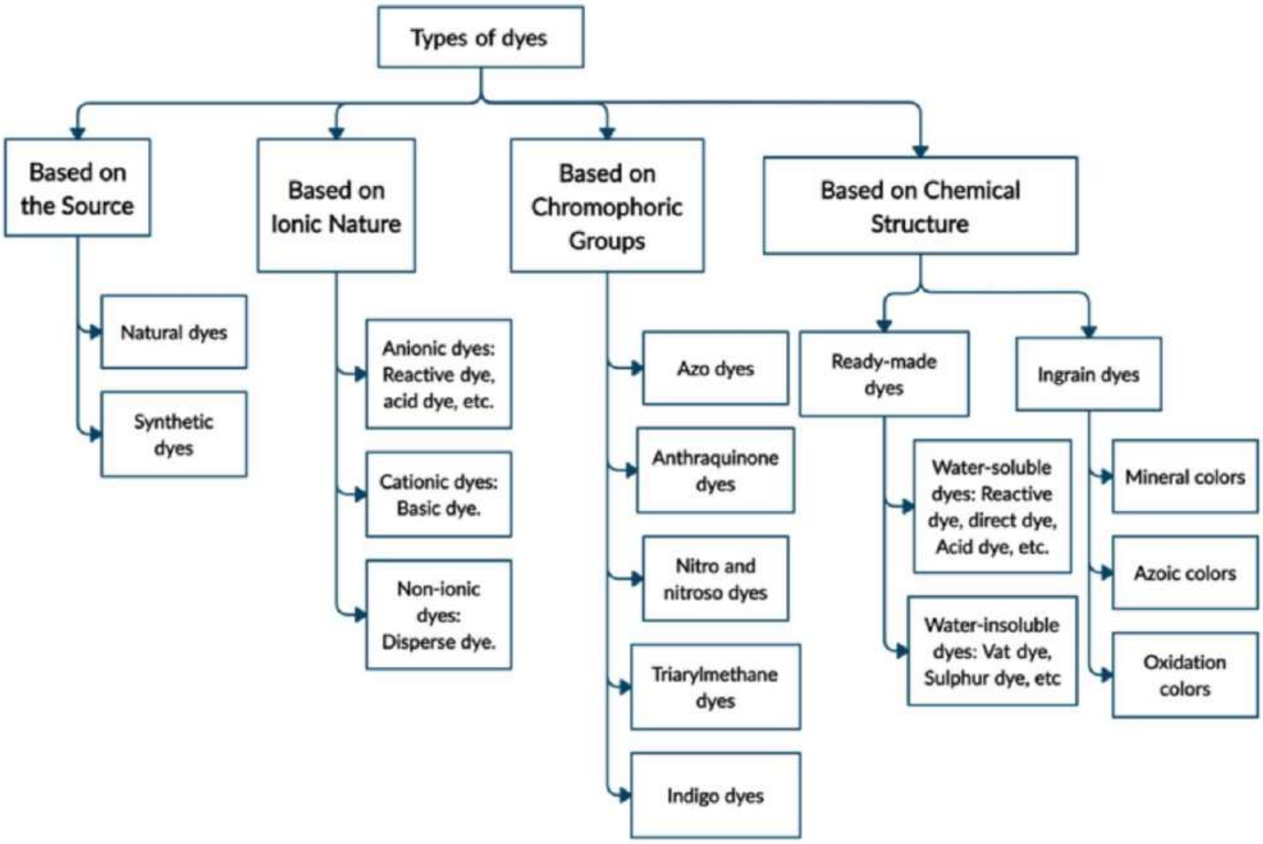


Figure 2.2: Classification of dyes (Dutta et al., 2022)

(a) Natural Dye: Until 1856, all textiles used throughout the world were natural dyes. That is, these dyes were obtained directly from the natural environment in one way or another. The vast majority of the natural dyes came from either plants or animals. For example, a major dye discovered by the Aztec or Mayan Indians was 'cochineal'.

Another natural animal dye which had a tremendous impact in the ancient world was 'Tyrian Purple'. This dye was discovered near the ancient city of Tyre, which is in Lebanon. It was discovered that when a sea mollusk native to the Tyre coastal region was crushed and refined, an extremely bright purple dye was produced. Today, the Tyrian Purple sea mollusk is extinct, but the dye can be made synthetically.

The most well known and most used dye of all time is 'indigo'. This dye is obtained from the leaves, stems and beans of a variety of plants, including one variety known as the indigo plant.

(b) Synthetic Dye: The first artificial dye, a purple called "mauveine," was discovered in 1856 by Sir William Henry Perkin of England. In addition to revolutionizing the textile dying industry, his discovery sparked the growth of the synthetic organic chemistry sector. Natural dyes don't produce colors with as much intensity or longevity as contemporary synthetic dyes. Additionally, these dyes can be challenging to get and labor-intensive to use on textile fibers.

(c) Direct Dye: These dyes, known as direct dyes, can be added straight to the fabric because they have a high affinity. With azo as their primary chromophoric group, this dye is primarily a sodium salt of sulphonic acid or carboxylic acid (Figure 2.3). They are anionic and soluble in water. It is common to color cellulosic fibers; protein fibers are also used (Benkhaya et al., 2017; Muntasir, 2020). They rely on dye to fiber associations like hydrogen-bonding and dipole moments to help with color fastness after dyeing rather than reacting with the fiber structure. In general they exhibit good, light fastness. However, they have poor wash fastness and poor wet crockfastness.

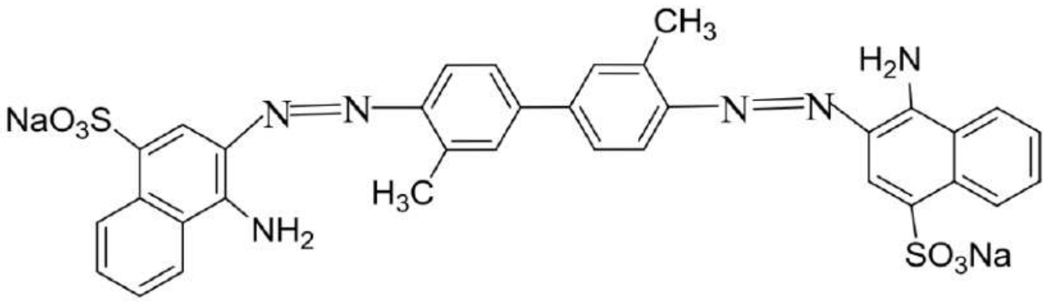


Figure 2.3: Example of direct dye (C.I. Direct Red 2)

(d) Reactive Dye: Reactive dyes contain reactive groups with halogens, which combine to form a covalent link and become a part of the fiber structure. It is typically used to dye cotton items, although it can also be used to color protein and polyamide-based products (Benkhaya et al., 2017; Muntasir, 2020). It is anionic in nature, soluble in water, and exhibits strong wash fastness, which means it contributes less to the volume of wastewater production than other dyes. Although they differ in how they react to sunlight, most reactive dyes offer acceptable lightfastness. The principal chemical classes of reactive dyes are azo (including metalized azo), triphendixazine, phthalocyanine, formazan, and anthraquinone, vinyl sulphone etc (Figure 2.4).

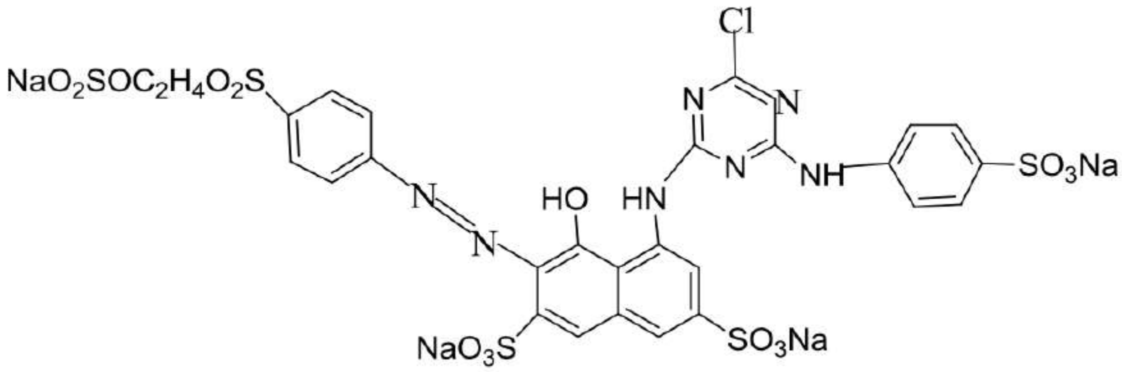


Figure 2.4: Example of reactive dye (C.I. Reactive Red 198)

(e) Disperse Dye: Disperse dyes are scarcely soluble dyes that penetrate synthetic fibres. These dyes are used to color thermoplastic hydrophobic textiles. It is mainly used for dyeing acetate, triacetate, nylon, acrylic, polyester-related goods (Benkhaya et al., 2017; Muntasir, 2020). They are usually small azo or nitro compounds (yellow to red), anthraquinones (blue and green) or metal complex azo compounds (all colors) (Figure 2.5).

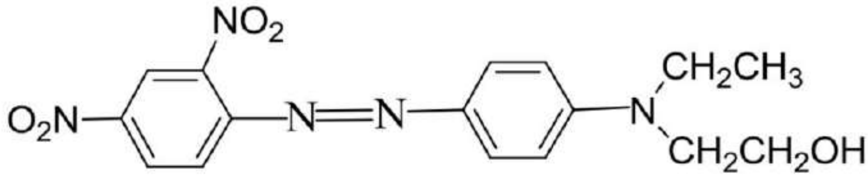


Figure 2.5: Example of disperse dye (C.I. Disperse Red 8)

(f) Acid Dye: Acid dyes primarily consist of carboxylic, or sulphuric acid salts, (Figure 2.6) which are extremely soluble in water and anionic. They primarily form ionic connections but can also produce van-der-Waals and H-bonds (Dutta et al., 2022). Additionally, acid dyes are applied in an acidic bath; they can color protein and polyamide fibers since these fibers are particularly responsive to the dye (Dutta et al., 2022) which includes nylon, silk or wool. Acid dyes are azo chromophoric systems (the most important group), anthraquinone, triphenylmethane or copper phthalocyanine, which are soluble in water by the introduction of one to four sulphonate groups (Benkhaya et al., 2017).

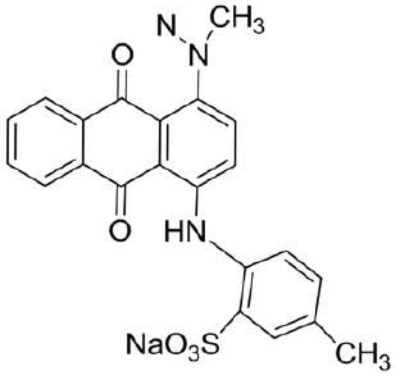


Figure 2.6: Example of acid dye (C.I. Acid Blue 25)

(g) Basic Dye: Basic dyes can dissolve in water and form colored cations in solution, which are electrostatically drawn to substrates with a negative charge (Benkhaya et al., 2017). These water-soluble cationic dyes are used on modified polyesters, nylons, and polyacrylonitrile for paper. They were initially used for silk, wool, and cotton that had been tannified. Basic dyes have a wide color spectrum and a variety of colorfastness characteristics. Many of these dyes generate fluorescent (neon) colors or exceptionally vivid hues. These ultra-bright dyes typically exhibit low lightfastness. The principal chemical classes are diazahemicyanine, triarylmethane, cyanine, hemicyanine, thiazine, oxazine, and acridine (Figure 2.7).

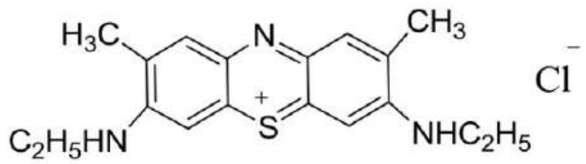


Figure 2.7: Example of basic dye (Basic Blue 24)

(h) Vat Dye: Vat dyes, which are water-insoluble dyes, are especially and frequently used to color cellulose fibers. Vat dyes is commonly employed for dying cotton-based products and they are mostly utilized to color denim or jeans (Dutta et al., 2022).

The dyeing process is based on the reduced (leuco) vat dyes' solubility. Leuco vat dyes that are soluble impregnate the cloth after being reduced with sodium dithionite. The dye is then returned to its insoluble state by oxidation. The dyes are high wash fastness and usually very lightfast. With proper after washing, they exhibit good to excellent wet and dry crock fastness.

These dyes typically consist of a keto group and Preston has claimed that other vat dye classes (Figure 2.8), such as indigoid and thioindigoid, anthraquinone (indanthrone, flavanthrone, pyranthone, acylaminoanthraquinone, anthrimide, dibenzathrone, and carbazole), are provided by indigo derivatives, most of which are halogenated (especially bromo substituents) (Preston, 1986). Indigo, also known as indigotin, is the most significant naturally occurring vat dye and is present in many indigofera species as its glucoside, indicant (Benkhaya et al., 2017). Vat refers to the vats that were used for the reduction of indigo plants through fermentation. Today, synthetic indigo is the dye mostly used by the textile industry. When extremely high light- and wet-fastness qualities are required, vat dyes are used.

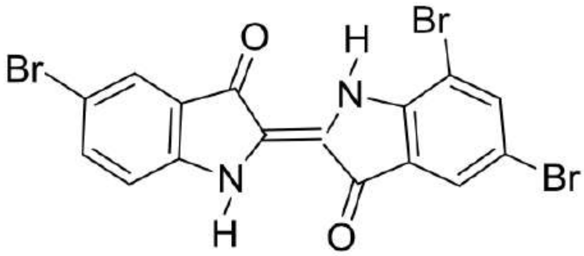


Figure 2.8: Example of vat dye (Vat Blue 5)

(i) Azoic Dye: These were mainly mono or bi-azo water-insoluble coloring substances that need coupling components to produce colors (Dutta et al., 2022). Azoic have been reported to the largest class of synthetic dyes in different studies (Lucas et al., 2007; Benkhaya et al., 2017). Azo dyes have a variety of structural elements, but the presence of the azo linkage, or N=N (Figure 2.9), is their most significant structural characteristic. It is possible for this linkage to occur more than once, hence mono azo dyes only have one azo linkage, compared to two in diazo and three in triazo, respectively (Benkhaya et al., 2017). Cotton, nylon, and polyester-related materials are commonly used with these dyes (Benkhaya et al., 2017).

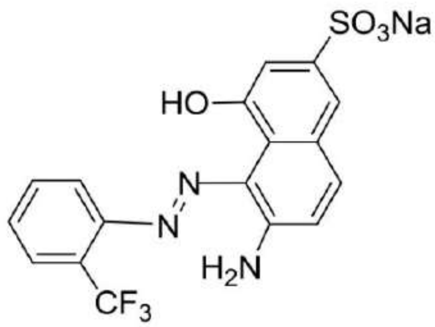


Figure 2.9: Example of azoic dye

(j) Sulfur Dye: These colors are applied to cotton using sodium sulfide as the reducing agent in an alkaline reducing bath. However, they are not soluble in water, so they need reducing agents to make them soluble (Dutta et al., 2022). This set of dyes is somewhat modest in terms of numbers. This class is significant from an economic standpoint because the dying is inexpensive and has strong washfastness characteristics.

These colors are made mostly through thionation of different aromatic intermediates (Figure 2.10), and the majority are exceedingly complicated in structure and largely unexplored. The sulfur dye used to produce black and brown cotton fabrics is analogous to vat dyes.

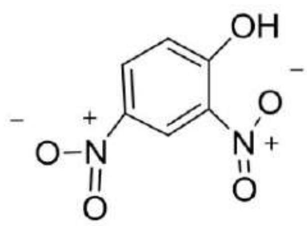


Figure 2.10: Example of sulfur dye (Sulfur Black)

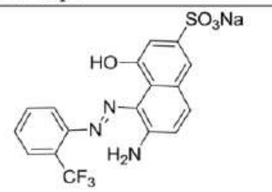
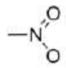
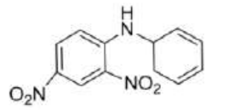
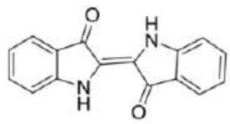
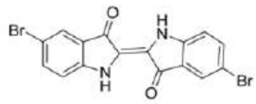
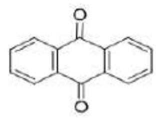
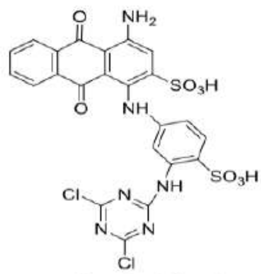
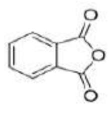
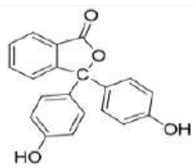
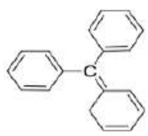
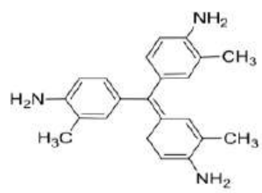
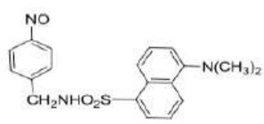
Wastewater is the main entry point for dyes into the environment. Data on dye consumption and the level of fixing of the various dye classes should be taken into account when determining the proportional contribution of the various dye classes in the wastewater of textile-processing businesses. Table 2.2 shows the percentage of contribution of different dyes in textile effluent.

Table 2.2: Estimated degree of fixation for different dye/fibre combination (EWA 2005)

Dye Class	**Fiber Type**	**Degree of Fixation, %**	**Loss to Effluent, %**
Acid	Wool, silk, nylon	80~95	05~20
Basic	Acrylic, silk	95~100	0~ 05
Direct	Cellulose	70~95	05~30
Metal Complex	Wool	92~98	02~08
Reactive	Cellulose	50~90	10~50
Sulphur	Cellulose	60~90	10~40
Vat	Cellulose	80~95	05~20
Disperse	Polyester, nylon	90~100	0~10
Modified Basic	Acrylic	95~98	02~05

The chromophore list for different types of dyes is summarized in Table 2.3 (Marzec, 2014).

Table 2.3: Different chromophores for various types of dyes

Class	Chromophore	Exemple
Azo dyes	—N=N—	 Acid Red 337
Nitro dyes		 Disperse yellow 14
Indigoid dyes		 C.I. Vat Blue 35
Anthraquinone dyes		 Reactive Blue 4
Phthalein dyes		 Phenolphtaleine
Triphenyl methyl dyes		 Basic violet 2
Nitroso dyes	—N=O	 Fluorescent-labeled nitroso compound (DNSBA-NO)

2.3 Environmental Impact of Textile Industries Wastewater

The textile industry generally faces difficulty in meeting wastewater discharge limits, particularly, with regard to dissolved solids, pH, BOD, COD, sometimes, heavy metals and color of effluent (Chen et al., 2005). Industrial emission and waste effluent generated from factories are associated with heavy disease burden and this could be part of the reasons for the current shorter life expectancy, 61.4 years both for male and female in the country (WHO) when compared to the developed nations. Some heavy metals contained in these effluents (either in free form in the effluents or adsorbed in the suspended solids) from the industries have been found to be carcinogenic (Tamburlini et al., 2002) while other chemicals equally present are poisonous depending on the dose and exposure duration.

An azo dye made from the aromatic amine 1,4-diamine benzene can cause rhabdomyolysis, severe tubular necrosis, lifelong blindness, contact dermatitis, chemosis, lacrimation, exopthamlmosis, vomiting, gastritis, hypertension, and vertigo. Upon consumption, there is respiratory discomfort as well as oedema of the face, throat, pharynx, tongue, and larynx (LGC, 1999). Water can enhance aromatic amines, making it easier for them to enter the body through the skin and some other accessible places, like the mouth. Since more dye can be consumed in a shorter amount of time, ingesting it is a quicker and potentially riskier method of absorption. After being broken down by liver enzymes, water-soluble azo dyes become dangerous. When humans are exposed to wastewater, all those things take place (Hassaan and Nemr, 2017). These chemicals are not only poisonous to humans but also found toxic to aquatic life and potential sources of food contamination (Dey and Islam, 2015).

High TSS and TDS detected could be attributed to the high color (from the various dyestuffs being used in the textile mills) and they are the major sources of heavy metals. Increased heavy metal concentrations in river sediments increases suspended solids concentrations (Dey and Islam, 2015). TSS generally contains fine clay, plankton, organic and inorganic compounds, colloidal substances, and other microorganisms (Bruggen et al., 2001). Unprocessed industrial, municipal, and agricultural wastage increases the TSS value (Ahmed et al., 2016). Suspended particles can choke fish gills and kill them. A protensive edible freshwater fish called Mastacembelus armatus that was exposed to textile effluent may have caused changes to the way that ions are regulated in tissues including the liver, kidneys, and muscles raising the concentration of potassium, calcium, and magnesium ions while maintaining the quantity of sodium and chloride ions (Karthikeyan et al., 2006). The freshwater crab Spiralothelphusa hydrodrome, a vital source of food for individuals in southern India, has been affected by the effluent from the textile dye industry, which has resulted in a loss of nutritional content, including lipids, carbohydrates, and protein (Sekar et al., 2009). Like Catla, dye effluent has a significant impact on feed uptake and food conversion rates (Dutta et al., 2022).

Dyes of textile effluent also decrease the capacity of algae to make food and oxygen (Mazumder, 2011). The growth of Spirulina platensis is inhibited and the nutritional value of the water is decreased when the concentration of color in the water is raised. The chain in the bodies of water is impacted by the Ramazol Red Brilliant dye; as a result, it leads to an unbalanced environment (de Sousa et al., 2012). Additionally, S. Quadricauda, a freshwater microalga, can have its development and biomass output markedly reduced by the use of indigo dye, as well as have their morphological structure altered (Chia and Musa, 2014).

Various dyes in wastewater severely affect photosynthetic function in plant (Praveen et al., 2018). They also have an impact on aquatic life due to low light penetration and oxygen consumption. Color water causes scarcity in the light which is essential for the development of the aquatic organisms. As result, it leads to an imbalance in the environment (Praveen et al., 2018). They may also be lethal to certain forms of marine life due to the occurrence of component metals and chlorine. Dyes are also detected to hinder with certain municipal wastewater treatment operations such as ultraviolet decontamination etc. (Mazumder, 2011).

Therefore, untreated textile effluent can contaminate groundwater and waterbodies, reduce dissolved oxygen in water and affect aquatic ecosystems which may indirectly cause climate change.

2.4 Technologies for Textile Industries Wastewater Treatment

Organic compounds, suspended particles, and dyes are just a few of the pollutants found in textile effluent that can be harmful to the environment and people's health. Therefore, to minimize the environmental impact of textile wastewater generation, textile manufacturers can adopt more sustainable production practices, such as using water-efficient technologies, reducing chemical use, and implementing wastewater treatment systems. Various treatment technologies have been developed to treat textile wastewater.

Here are some of the most common textile wastewater treatment technologies:

a. Physical treatment: Physical treatment methods include screening, sedimentation, flotation, and filtration. These processes remove suspended solids and some organic compounds.

b. Chemical treatment: Chemical treatment methods involve the use of chemicals to remove pollutants from the wastewater. Examples of chemical treatment methods include coagulation, flocculation, and oxidation.

c. Biological treatment: Biological treatment methods use microorganisms to degrade organic pollutants in the wastewater. Examples of biological treatment methods include activated sludge process, anaerobic digestion, and constructed wetlands.

d. Advanced oxidation processes: Advanced oxidation processes (AOPs) involve the use of powerful oxidizing agents such as ozone, hydrogen peroxide, and UV light to degrade pollutants in the wastewater.

e. Membrane filtration: Membrane filtration is a technology that uses membranes to filter out pollutants from the wastewater. Examples of membrane filtration include ultrafiltration, nanofiltration, and reverse osmosis.

f. Electrochemical treatment: Electrochemical treatment involves the use of an electric current to degrade pollutants in the wastewater. This technology is effective in treating wastewater with high concentrations of dyes.

These treatment technologies can be used individually or in combination, depending on the characteristics of the textile wastewater and the desired treatment outcome.

2.4.1 Activated Sludge Process for TWW

Activated Sludge Process (ASP), which is a common method used for treating wastewater in textile industries. In the ASP process, microorganisms are used to break down the organic matter in the wastewater, converting it into less harmful substances.
The process typically involves four stages:

a) Primary Treatment: In this stage, large solids are removed from the wastewater through sedimentation or filtration (Metcalf and Eddy, 2003).

b) Aeration Tank: In the aeration tank, wastewater is mixed with activated sludge, which is a mixture of microorganisms, oxygen, and nutrients. The mixture is aerated to provide oxygen to the microorganisms and facilitate their growth and activity (Metcalf and Eddy, 2003).

c) Secondary Clarification: In this stage, the mixture of wastewater and activated sludge is allowed to settle, and the clarified water is separated from the sludge.

d) Sludge Treatment: The sludge that is separated from the clarified water is then treated through various methods, such as anaerobic digestion or aerobic stabilization, to reduce its volume and make it less harmful before it is disposed of.

The ASP process is effective in removing pollutants from textile wastewater, including organic matter, nitrogen, and phosphorus. It is also relatively simple and cost-effective to operate, making it a popular choice for textile industries.

However, the ASP process also has some limitations, such as high energy requirements for aeration and the potential for the formation of odorous compounds. The space required for this technology cost a lot to end user and higher capital cost is a common negative side of this process for textile industries.

Therefore, it is important to carefully design and operate the ASP system to ensure effective and efficient wastewater treatment. The general process flow has been shown in Figure 2.11.

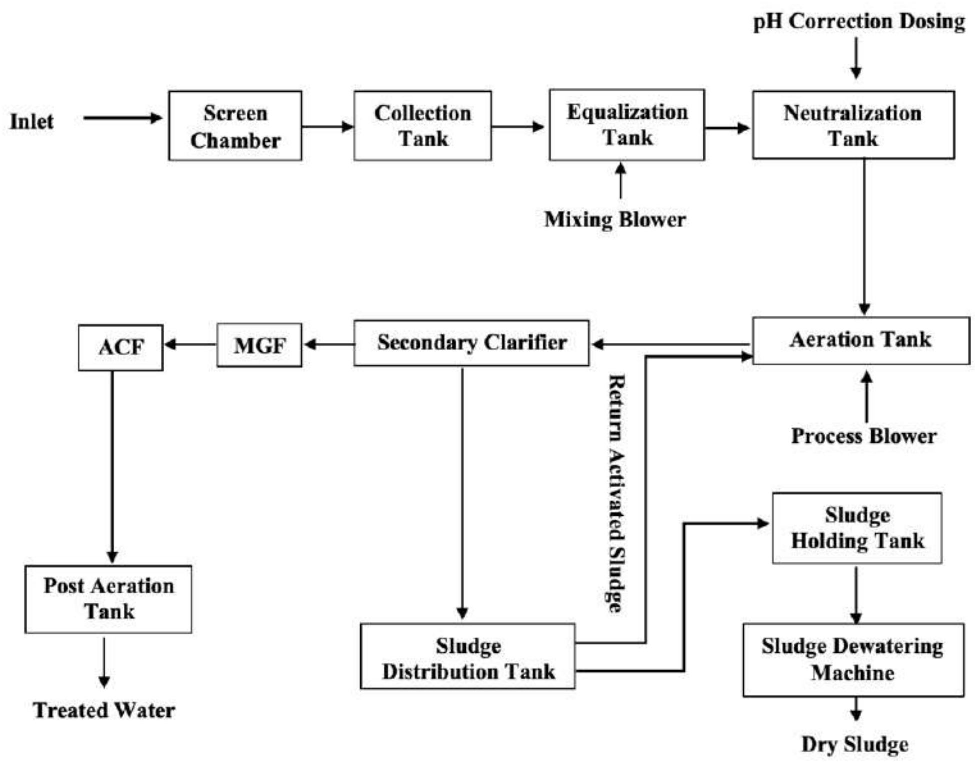


Figure 2.11: Process flow diagram for activated sludge process ETP (Pandey and Singh, 2014)

2.4.2 Bio-chemical Process for TWW

Bio-chemical technology for effluent treatment plant (ETP) is a combination of both biological and chemical process for degradation of textile pollutants (Tripathy and De, 2006). Generally, this system is applied to those wastewaters which are tough to treat completely by biological system.

Since, the wastewater from textile industries typically contains a variety of pollutants, including organic and inorganic compounds, dyes, solvents, and heavy metals, so lot of textile industries use this system to treat their effluent.

This process methodology is classified follows:

a) Preliminary treatment: Preliminary treatment consists of removal of floating materials like dead animals, wood pieces, tree branches heavy settable inorganic solid, fat, oil and grease from the influent.

Screens: Screens are used for removal of large floating and sub-merged material such as plastic, paper pieces, rubber, etc. from effluent. Screens are classified in various ways:

i) Based on method of cleaning- such as mechanical or manual.
ii) Depending upon shape- such as disc, drum, band, etc.
iii) Based on the size of opening- such as coarse, medium and fine screens.

b) Primary treatment : Primary treatment involves the physical separation of suspended and colloidal material from waste water stream. It is accomplished in equalization cum neutralization tank (Babu et al., 2007). It also involves chemical coagulation and flocculation to enhance removal efficiencies. This is usually accomplished in plane sedimentation tank also known as primary clarifier. Typically, primary treatment can remove 25-35% of the BOD and 60-80% of the TSS before the water goes to secondary treatment.

c) Secondary treatment : Secondary or biological treatment involves removal of organic matter and residual suspended material which is generally accomplished by using biological unit processes (Metcalf and Eddy, 2003).

Aerobic process- Suspended growth:
Suspended growth processes are commonly known as activated sludge process. Aerobic suspended growth systems are of two types - those which employ sludge recirculation (conventional activated sludge process) and those which do not have sludge recirculation.

d) Tertiary treatment: Tertiary treatment involves removal of residual pollutants to some extent which are not removed in primary and secondary treatment. This includes removal of organic materials, soluble inorganic materials etc. In this case, multi graded filter and activated carbon filter will be employed to ensure national standards for final discharged water (Sultana et al., 2013).

However, Bio-chemical process based effluent treatment plant (ETP) produces lot of chemical sludge and operation cost is also higher due to higher chemical consumption in plant. Therefore, Textile industries should prioritize the adoption of sustainable and environmentally friendly practices to minimize the negative impacts of ETPs on the environment and human health.

The general process flow has been shown in Figure 2.12.

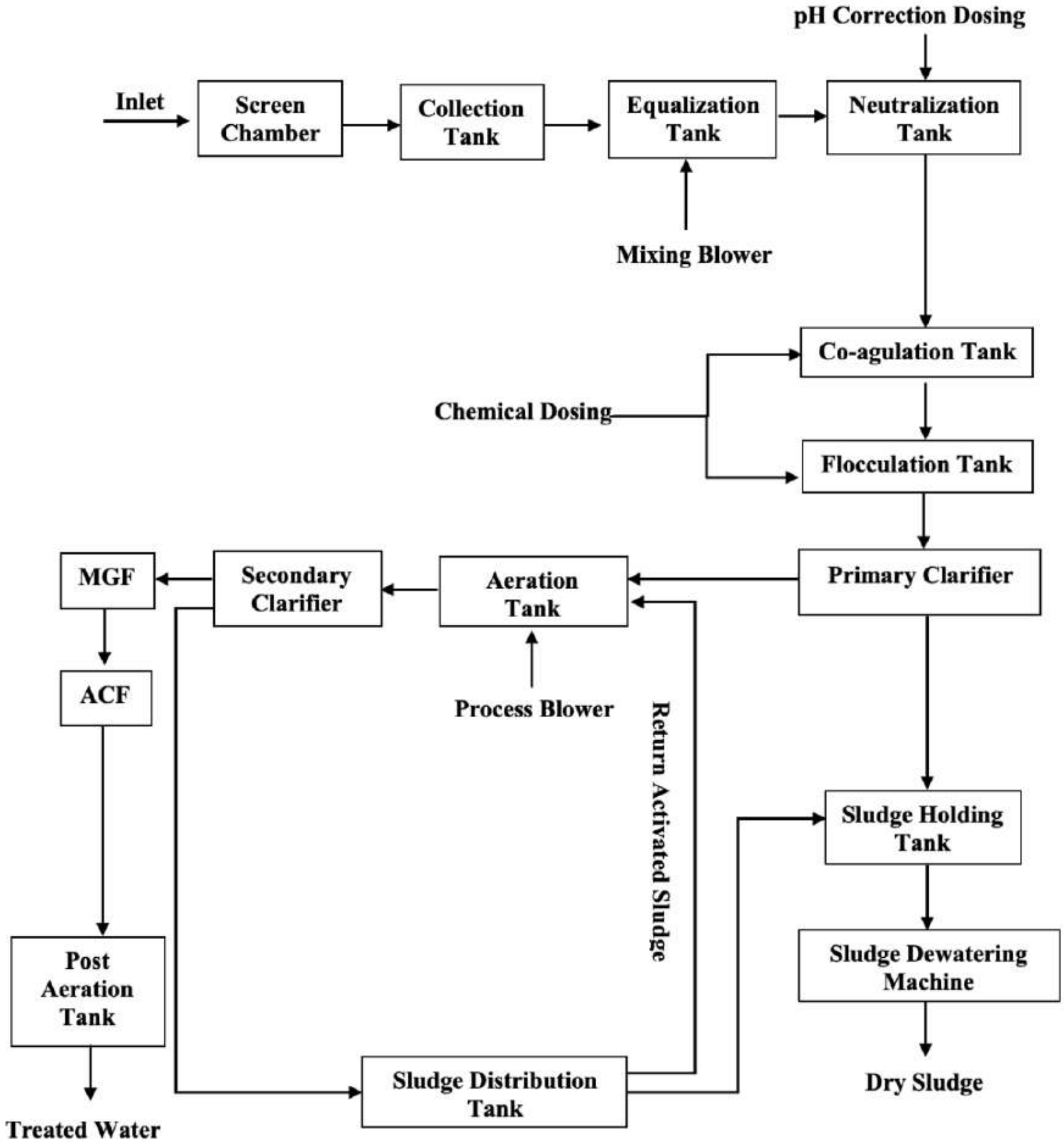


Figure 2.12: Process flow diagram for Bio-Chemical ETP (Tripathy and De, 2006)

2.4.3 Membrane Bio-reactor Process for TWW

MBR (Membrane Bioreactor) based ETP (Effluent Treatment Plant) is a type of wastewater treatment system that combines biological treatment and membrane filtration technology to produce high-quality treated water (You et al., 2008). The microorganisms break down the organic matter in the wastewater, while the membrane filters remove suspended solids, bacteria, and other contaminants.

In the textile industry, MBR ETPs are often used to treat the wastewater generated during various textile manufacturing processes. MBR ETPs have several advantages over conventional wastewater treatment systems. They require less space, produce higher-quality treated water, and can be operated at a lower cost. The general process flow of MBR system has been shown in Figure 2.13.

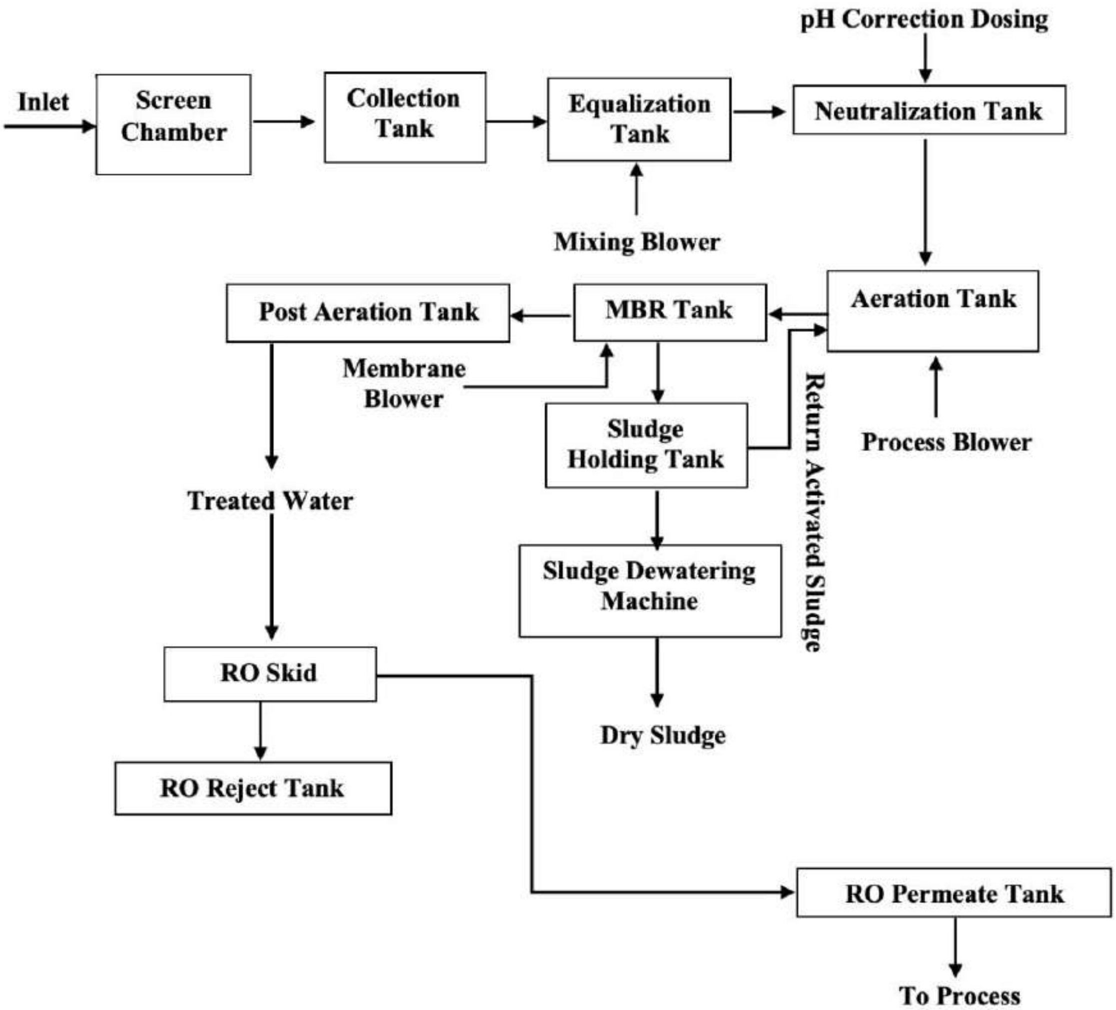


Figure 2.13: Process flow diagram for MBR ETP (You et al., 2008).

The major steps of MBR based ETP are as following:

a) Pre- Treatment by Screening: Physical separation by manual screening of 6 mm and 4mm of big sized impurities like cloth, plastics, wood logs, paper, stone etc.

b) Primary Treatment in Equalization Tank: After screening inlet wastewater is sent to equalization tank to make it equal or homogenous form by providing air from air blower through coarse type diffuser. Neutralization is also done in a small portion of this tank where removal of excess acidity or alkalinity from wastewater by chemical dosing.

c) Secondary Screening (applicable for Membrane Technology only): Equalized raw wastewater is passed through ≤1 mm punch hole internally fed rotary drum screen or center flow traveling band screen or fine brush screen to remove fine particles like hair, sand, glass etc. which is dangerous for membrane fiber.

d) Secondary Treatment in Aerobic Reactor/Tank: In this method, effluent is aerated in a reaction tank consist of microbial population in suspension form. Aerobic bacteria degrade effluent into CO_2 and H_2O for which oxygen is supplied through mechanical aeration or by diffused aeration system. The bacterial flora grows and remains suspended in the form of a floc called activated sludge. A part of sludge is recycled for the same tank to provide an effective microbial population for a fresh treatment cycle.

e) Tertiary Treatment by Membrane Separation in Membrane Tank: Membrane filtration is process of removing particulates and bacterial impurities that could not be removed in earlier treatment, from water by passing it through a porous medium. It is used to remove colloidal and other impurities which impart turbidity to water. Since the MLSS wastewater from aerobic tank is directly pass-through membrane so it is considered as part of aeration tank and this combined system is called Membrane Bio-Reactor/MBR.

i. Membrane filtration provides a positive barrier to suspended bio-solids that they cannot escape the system unlike gravity settling in activated sludge process.

ii. Due to the above aspect of MBR, aeration tank size in the MBR system can be one-third to one-fourth the size of the aeration tank in an activated sludge system. Further, instead of gravity settling based clarifier, a much more compact tank is needed to house the membrane cassettes in case of submerged MBR and skid mounted membrane modules in case of non-submerged, external MBR system.

iii. Thus, MBR system requires only 40-60% of the space required for activated sludge system, therefore significantly reducing the concrete work and overall foot-print.

iv. Due to membrane filtration (micro/ultrafiltration), the treated effluent quality in case of MBR system is far superior compared to conventional activated sludge.

f) Collection of Treated Water in Permeate Tank: The produced treated water from membrane filtration is accumulated to Back flush tank, which will always filled up and to be over flowed to permeate tank. In permeate tank post aeration mechanism to increase the DO for discharging water. The collection of MBR treated water and other cleaning operations of the membrane fiber by air or chemical is fully performed with automatic process control in SCADA/HMI.

g) Sludge Treatment by Dewatering Process: Sludge wasting is an integral process in MBR treatment system. The excess waste sludge comes from MBR tanks to this sludge holding tank. Then sludge is made dry after removing excess liquid from wet sludge by using sludge dewatering machine. The produced sludge from dewatering generally contains 80~88 % water.

Despite of several advantages, MBR system effluent treatment plant has some contradictory sides as well. MBR ETPs are typically more expensive to construct than traditional wastewater treatment plants, additional blower for membrane operation causes higher energy requirement thus increase the operation cost and lot of automation sometimes makes the system clumsy rather than make it easy. Therefore, textile industries still cannot accept that MBR based ETP as a sustainable and cost-effective solution for them.

2.4.4 Electro-coagulation Process for TWW

Electrocoagulation is a process that uses an electrical current to treat wastewater by destabilizing and coagulating contaminants. The process involves passing an electrical current through the wastewater, which causes the formation of metal hydroxide flocs (Butler et al., 2011). These flocs attract and trap the contaminants, allowing them to be easily separated from the water. The process is efficient, easy to operate, and requires little maintenance.

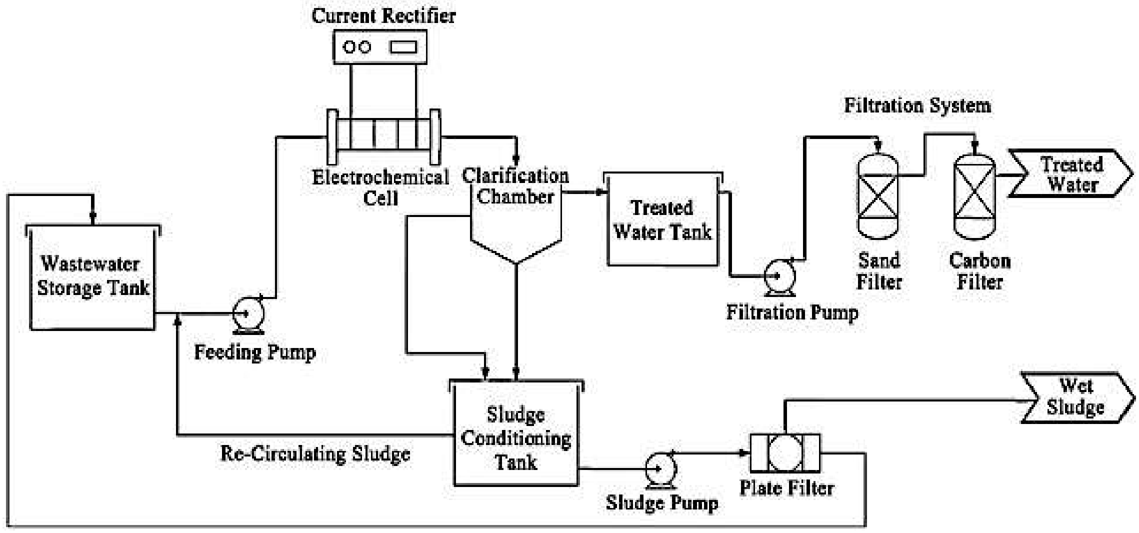


Figure 2.14: Process flow diagram for electrocoagulation (EC) system ETP (Meas et al., 2010).

The major steps involved in an electrocoagulation (EC) (Figure 2.14) effluent treatment plant (ETP) are as follows:

a) Pretreatment: The first step in the EC ETP process is to pretreat the wastewater to remove large particles and debris, such as hair, lint, and fiber. This can be done through screening or sedimentation.

b) Electrocoagulation: The pretreated wastewater is then fed into an electrocoagulation reactor where an electrical current is applied. The reactor contains metal electrodes, which generate metal hydroxide flocs that attract and coagulate contaminants in the wastewater.

c) Flocculation: The flocs formed in the electrocoagulation reactor are then allowed to settle or are passed through a flocculation tank where the flocs grow in size by attracting other coagulated particles.

d) Separation: The settled flocs or the flocs formed in the flocculation tank are then separated from the water by sedimentation, filtration, or centrifugation.

e) Discharge or reuse: The treated wastewater can then be discharged into a nearby water body or reused for non-potable purposes, such as irrigation or industrial processes.

The disadvantages of EC system also cannot be avoided for the ETP users of textile industries. Electrocoagulation-based ETP consumes a significant amount of energy, making it expensive to operate. Electrocoagulation requires regular maintenance to ensure that it operates at peak efficiency. The electrodes used in the process need to be cleaned and replaced periodically, which can be time-consuming and costly. In these circumstances, the basic drawbacks of EC system need to be considered before adopting it as a wastewater treatment technology for textile industries.

2.5 Aerobic Granulation Technology for Wastewater Treatment

Aerobic granulation technology is a process used in wastewater treatment plants to remove organic matter and other pollutants from wastewater. It involves the formation of dense, compact aggregates of microorganisms, called aerobic granules, that can efficiently degrade contaminants. The process starts by introducing wastewater into a reactor where microorganisms are present. The microorganisms form small aggregates that gradually grow larger and denser, forming aerobic granules.

The aerobic granule was defined as follows: "Granules making up aerobic granular activated sludge are to be understood as aggregates of microbial origin, which do not coagulate under reduced hydrodynamic shear and which settle significantly faster than activated sludge flocs" (Morgenroth et al., 1997). Due to its distinct characteristics, including great settleability, high biomass retention, and high resistance against variations in pollution load (Tay et al., 2002), as well as environmental variables like temperature and pH, granular sludge has emerged as a prominent wastewater treatment. It has been successfully applied in various wastewater treatment plants worldwide including both municipal and industrial wastewater and is considered a promising technology for future wastewater treatment.

2.5.1 History of Aerobic Granulation

The concept of aerobic granulation was first introduced in the late 1990s (Heijnen and Loosdrecht, 1998; Morgenroth et al., 1997), and since then, it has been studied and developed extensively. The initial studies focused on the formation and characteristics of aerobic granules, and their potential application for wastewater treatment. The first full-scale applications treating sewage started to appear from 2005 under the trade name Nereda® (Giesen et al., 2013).

AGS wastewater treatment systems have proliferated recently, helping to uncover and fully explore the technology's benefits as well as develop answers to pressing technical problems and newly emerging operational challenges (Campo et al., 2021). By mid-2021, there were about 90 AGS wastewater treatment facilities, more than doubling from the previous year (Sepúlveda et al.,2019). The facilities ranged in size from 100 to 600,000 m3, and their main functions were to fully biodegrade organic materials or to remove nitrogen and phosphorus from household sewage and mixed household-industrial sewage (Hamza et al., 2022). AGS-deploying firms' data analysis revealed that 13 new full-scale wastewater treatment plants were being built and put into operation in 2020–2021. The number of full-scale AGS installations is provided in below Figure 2.15 (Hamza et al., 2022).

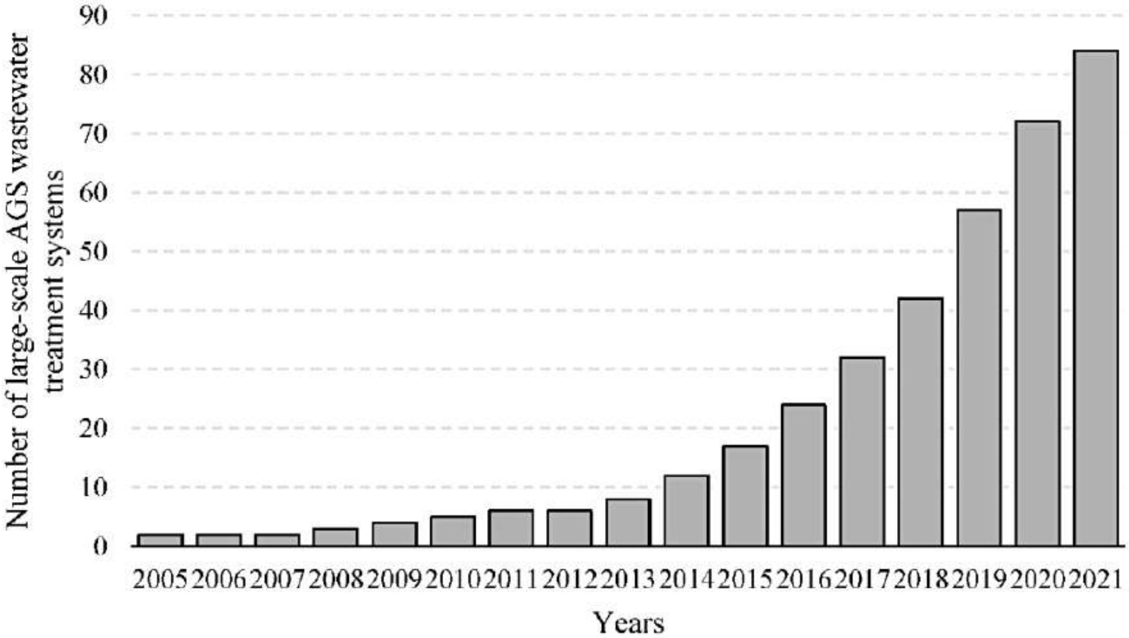


Figure 2.15: Worldwide number of large-scale AGS wastewater treatment systems from 2005 to 2021 (Hamza et al., 2022).

2.5.2 Granulation Models

Based on the thermodynamics, some physico-chemical model mechanisms for granulation have been developed; those include inert nuclei, model, selection pressure model, multivalent positive ion-bonding model, local dehydration and hydrophobic interaction model and surface tension model.

a) Inert Nuclei Model: When there are inert microparticles present in the reactor, bacteria may adhere to the particle surfaces and create an early biofilm, or embryonic granules. Under specific operating conditions, the associated bacteria can multiply and help the mature granules develop further. The inert nuclei concept, which is depicted in below Figure 2.16, contends that the presence of nuclei or micro-size bio-carriers for bacterial attachment is a necessary prerequisite for granulation (Liu et al., 2003).

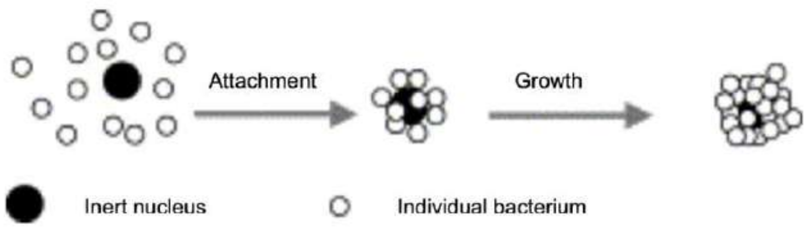


Figure 2.16: Scheme for inert nuclei model (Liu et al., 2003).

b) Selection pressure model: This model shows that the system would continue to contain heavy components while washing off lighter, scattered muck. Microbial aggregation in the reactor might be a useful defense mechanism against strong selection forces. Gao et al., 2010 found in their study that when the hydraulic selection pressure was very low, no granulation was seen. Their result also showed that high hydraulic stress and quick settling time improved granulation.

c) Multi-valence positive ion-bonding model: Bacteria have negative charged surfaces under usual pH condition; a basic idea to expedite granulation process is to reduce electrostatic repulsion between negatively charged bacteria by introducing multi-valence positive ion, such as calcium, ferric, aluminum or magnesium ions, into seed sludge (Yu et al., 2001).

The multi-valence positive ions may promote sludge granulation by bonding with extracellular polymers (EPS) (Figure 2.17). The high affinity between EPS and calcium create the bridge between EPS and link cells, EPS together to form an initial three dimension structure of microbial community, in which bacteria can further grow.

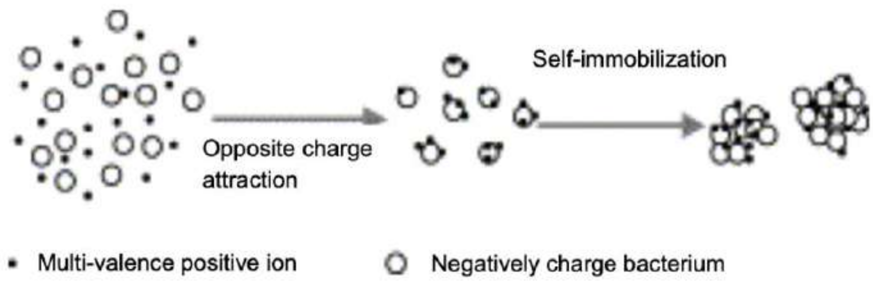


Figure 2.17: Schematic presents multi-valence positive ion-bonding model (Liu et al, 2003).

d) EPS Bonding Model: EPS could change the surface negative charges of bacteria, and thereby bridge two neighbor cells physically to each other as well as with other inert particulate matter.

Complex connections between EPS and cells create a net-like structure with plenty of water that shields cells from dehydration (Sheng et al., 2010) and creates a barrier of protection for the bacterial community, which helps to long-term stability (Jiang and Liu, 2012). EPS plays a significant part in aerobic granulation for microbial communication or the aggregation-disaggregation theory. Granule development together with the densification of bacterial aggregates is explained by an increase in cohesion in the initial phenomena caused by EPS. The ability of an aggregate to resist shear and elongation forces that cause erosion and/or breakup is known as cohesion. (Wan et al., 2011). Due to particular interactions of the EPS that make up the granule matrix, as the cohesion rises, the dense clustering of microorganisms expands.

e) Local dehydration and hydrophobic interaction mode: The outer surfaces of bacteria are hydrated under typical culture pH conditions. This water layer on bacteria's surfaces would stop one bacterium from approaching another. Strong hydration repulsion serves as the primary force keeping cells apart under physiological conditions, therefore local dehydration of surfaces that are close to one another would be necessary for cell-to-cell aggregation. Strongly hydrophobic surfaces will produce irreversible adherence. The hydrophobicity of cell surfaces increases, promoting cell-to-cell contact and acting as a catalyst for cell self-separation from liquid phase (Liu et al., 2003).

f) Surface Tension Model: Microbial granulation is a process to create a new interface, granule-liquid disrupting pre-existing individual bacteria-liquid interface. If the surface free energy of bacteria is lower than the surface free energy of the liquid, the free energy of aggregation decreases, and aggregation is encouraged as the surface energy of the inner carrier decreases. The opposite trend would occur if the surface energy of bacteria is higher than the liquid. Hydrophilic cell aggregation is promoted at low liquid surface tension, whereas hydrophobic cell aggregation is inhibited (Liu et al., 2003).

g) Cell-to-cell communication model: Intercellular communication and multicellular coordination have been known as an effective way for bacteria to achieve an organized spatial structure. Quorum sensing has been proven to be a major example of social behavior in bacteria, as signal exchange among individual cells allows the entire population to determine the best strategy to interact with the environment (Liu et al., 2003).

2.5.3 Anaerobic Granular Sludge vs Aerobic Granular Sludge

Aerobic granular sludge and anaerobic granular sludge are two types of microbial aggregates used in wastewater treatment processes. While both types of sludge have similarities, there are also significant differences in terms of their operational conditions and treatment capabilities.

A. Oxygen Requirement:

Aerobic Granular Sludge: Aerobic granular sludge requires oxygen for the microbial community to thrive. It is typically used in aerobic processes, such as sequencing batch reactors (SBRs) or aerobic granular reactors, where oxygen is supplied to support the growth of aerobic bacteria.

Anaerobic Granular Sludge: Anaerobic granular sludge, as the name suggests, operates under oxygen-free conditions. It is used in anaerobic processes, such as upflow anaerobic sludge blanket (UASB) reactors or anaerobic digestion systems. Anaerobic bacteria in these systems convert organic matter into biogas (methane and carbon dioxide) in the absence of oxygen.

B. Microbial Composition:

Aerobic Granular Sludge: The microbial composition of aerobic granular sludge primarily consists of aerobic bacteria, which require oxygen to survive. These bacteria are responsible for the oxidation of organic pollutants present in the wastewater.

Anaerobic Granular Sludge: Anaerobic granular sludge is composed of anaerobic microorganisms, including bacteria and archaea. These microorganisms perform anaerobic digestion, breaking down complex organic compounds into simpler substances like methane and carbon dioxide.

C. Application:

Aerobic Granular Sludge: Aerobic granular sludge systems are commonly used for the removal of organic matter, nutrients (such as nitrogen and phosphorus), and some recalcitrant compounds from wastewater. They are effective in treating high-strength wastewaters, such as those from food processing or pharmaceutical industries.

Anaerobic Granular Sludge: Anaerobic granular sludge systems are utilized for the treatment of organic-rich wastewaters, including municipal sewage, agricultural waste, and industrial effluents. They are particularly useful for energy recovery through the production of biogas and reducing sludge volume.

D. Sludge Characteristics:

Aerobic Granular Sludge: Aerobic granular sludge has a dense and compact structure, with good settling properties. It forms granules with a diameter typically ranging from 0.5 to 3 mm.

Anaerobic Granular Sludge: Anaerobic granular sludge also has a granular structure, but the granules are generally larger and have a diameter ranging from 1 to 6 mm. They exhibit good settling characteristics and are less prone to washout compared to other types of anaerobic sludge.

Both aerobic and anaerobic granular sludge systems have their own advantages and applications, depending on the wastewater composition and treatment objectives. The choice between them depends on factors such as the type of pollutants to be removed, energy recovery potential, and treatment efficiency requirements.

2.5.4 Operational Advantages of Aerobic Granular Sludge

Aerobic granular sludge offers several operational advantages for the treatment of highly polluted industrial wastewater:

A. High Pollutant Removal Efficiency: The compact and dense structure of granules allows for a high biomass concentration and efficient removal of pollutants.

B. Short Hydraulic Retention Time (HRT): The process typically operates with shorter HRTs compared to conventional wastewater treatment systems, leading to a smaller reactor footprint.

C. Reduced Sludge Production: The granules have a slower biomass growth rate compared to suspended biomass systems, resulting in reduced sludge production.

D. Resistance to Toxic Substances: The granules exhibit increased tolerance to toxic substances present in industrial wastewater due to the diverse microbial community and robustness of the granule structure.

2.6 Activated Sludge Process (ASP) vs Aerobic Granulation Sludge (AGS)

Aerobic granulation technology has many advantages over conventional activated sludge process of wastewater treatment. It can reduce the cost and energy requirement for wastewater treatment.

The activated sludge process, invented in 1914 in England by Arden and Lockett, is the most popular biological wastewater treatment technique. Through the years, numerous advancements have been made to this procedure. An efficient solid-liquid separation is key to the success of this technology. The efficiency of activated sludge treatment systems is mainly determined by two factors, first of all the metabolic capability of the growing microorganisms and secondly the settling properties of the activated sludge aggregates, usually flocs. A crucial phase in the treatment of activated sludge is the clarifier, where sludge separation takes place. By using adhesion forces in the clarifier, sludge flocs must coagulate into big, dense floc aggregates in order to accomplish pretty excellent settling of the biomass. Basically, there are two main types of settling problems: (Dey and Islam, 2015) bulking sludge due to the excess growth of filamentous organisms and poor flocculation or floc formation properties of the microorganisms.

Activated sludge flocs grow typically in a range of 30 to 1800 µm in diameter (Hilligardt and Hoffmann, 1997) but have a density that is only slightly higher than water density. Additionally, flocs and floc aggregates make it difficult for themselves to settle, which results in a relatively slow overall sedimentation velocity. The loss of suspended particulates into the effluent is frequently the result of inadequate separation qualities. If the sludge compacts poorly, recycling of the solids is hampered, thereby the treatment efficiency is reduced. If flow conditions in the tank cannot be further improved, hydraulic loading of the system must be reduced or the settling time must be extended in cases with poor sludge settling qualities. To prevent the discharge of organic matter into the effluent and the violation of COD discharge limitations, it is frequently expensive to construct and run adequate settling tanks.

Aerobic granular sludge (AGS) technology was developed and proposed as means of more compact wastewater treatment (Morgenroth et al., 1997). Aerobic granules are denser, more compact, and more spherical than activated sludge flocs (Liu and Tay, 2004; Beun et al., 1999). As a result, there is a high settling velocity that may enable for high biomass concentrations to contribute to compact reactors (Ni et al., 2009). AGS is most frequently produced in sequencing batch reactors (SBRs), where granule formation is favored by the operation philosophy of SBR and the choice of relatively slow-growing bacteria (Kreuk and Loosdrecht, 2004; Liu et al., 2004a).

In the study of Bengtsson et al., 2018, they performed a comparative experiment on municipal wastewater treatment between processes based on AGS and activated sludge. They found that a process based on AGS, including pre- treatment, post-polishing and sludge treatment, can be significantly more compact than a process based on activated sludge with a 40-50 % smaller foot-print, even at the same water depth. The compact nature of the AGS alternative was due to the high sludge concentration that can be applied and that no separate clarifier is needed. The estimated electricity demand was lower for the treatment process based on AGS than other option, namely 23 % lower than activated sludge.

In comparison to activated sludge and currently available compact solutions, procedures based on AGS appear to be more advantageous in terms of footprint and electricity demand.

2.7 Process for Aerobic Granulation

Generally, some multiple steps are required for aerobic granulation process where physio-chemical and biological factors are involved (Liu and Tay, 2002). The steps of the different processes are showed as following:

I. **Physical Process:** The first step is physical movement of microorganisms to initiate bacterium to bacterium contact. This physical movement actually is caused due to some physical forces like hydrodynamic force, diffusion mass transfer, gravity, thermodynamic effects, and cell mobility.

II. **Physio-Chemical Process:** The next step is to keep the stability of cell to cell contact as much as possible. This stability can be resulted from some attractive forces which are physical forces (e.g., Van der Waals forces, opposite charge attraction, thermodynamically driven reduction of the surface free energy, surface tension, hydrophobicity, filamentous bacteria that can bridge individual cells), chemical forces, and biochemical forces including cell surface dehydration, cell membrane fusion, signaling, and collective action in bacterial community.

III. **Microbial Process:** The third step is to make mature aggregation of the formed cell to cell contact. This maturation can be done through production of extracellular polymer, growth of cellular clusters, metabolic change, environment-induced genetic effects that facilitate the cell–cell interaction and result in a highly organized microbial structure.

IV. **Shear Force based Process:** The fourth step is to form a stable three-dimensional structure of microbial granules. For microbial cells to aggregate, a number of conditions have to be fulfilled. The outer shape and size of granules would result from the interactive strength/pattern between granules and hydrodynamic shear force, micro- bial species and substrate loading rate, and so on. Shear force has been demonstrated to play an important role, and also influences the structure and metabolism of aerobic granules (Tay et al., 2001b; Yang et al., 2002).

2.8 Favorable Environmental Factors for Aerobic Granulation

It has been noted that a variety of species, including methanogens, acidifying bacteria, nitrifying bacteria, denitrifying bacteria, and aerobic activated sludge, can contribute to biogranulation. In the study of nitrifying granulation, Tay et al. (2002) discovered that even for the identified nitrifying bacteria, nitrifying granules only developed under intense selection pressure. These seem to suggest that aerobic granulation is a microbial phenomena brought on by environmental changes through altering the microbial surface characteristics and metabolic processes of microbes. (Tay et al., 2001b; Qin et al., 2004; Wang et al., 2005). As a result, aerobic granulation ought to be species-independent and potentially indurated rather than constitutive. (Liu et al., 2005a).

There are few environmental factors which are liable to influence the formation of aerobic granulation. According to Liu and Tay (2004), a variety of factors, such as substrate composition, organic loading, hydrodynamic shear force, feast-famine regime, feeding strategy, reactor configuration, solids retention time, cycle time, settling time, and exchange ratio, would affect how aerobic granules formed in sequencing batch reactors (SBR) and their characteristics. To the creation mechanism of aerobic granules, however, only variables linked to selection pressures on the sludge particles would be relevant (Liu et al., 2005a). In SBR, the settling time and the volume exchange ratio had been recognized as the two main selection forces. (McSwain et al., 2004; Qin et al., 2004; Hu et al., 2005; Liu et al., 2005a; Wang et al., 2005). Therefore, aerobic granulation would be unsuccessful if sufficient settling time or exchange ratio is not controlled during operation of SBR. The importance of the major environmental factors is described in the following sections:

a) <u>Substrate Composition:</u>

The composition of the substrate is one of the key factors that can influence the development and stability of aerobic granules. A study by Wu et al. (2014) investigated the effects of substrate composition on the formation and characteristics of aerobic granules in a sequencing batch reactor (SBR) where they used three different substrates: acetate, glucose, and a mixture of acetate and propionate. The results showed that the substrate composition had a significant impact on the formation and properties of aerobic granules.

In another study, AGS reactors operating with solely soluble carbon (acetate and hydrolyzed peptone) and those operating with both soluble and particulate carbon (particulate starch) were contrasted by Wagner et al. (2015). The anaerobic phase required to be prolonged in the presence of particles to allow for substrate hydrolysis. The granules became less stable with a significant portion of floccular sludge due to constant access to soluble substrate from the hydrolysis, which led to a rise in the concentration of suspended particles in the effluent.

Granule stability was found to be improved with the presence of divalent cations Ca^{2+} and Mg^{2+} by forming bridges between EPS molecules (Li et al. 2009; Ren et al. 2008). It was shown that wastewater streams with calcium ion concentrations between 40 and 100 mg/L accelerated the rate of sludge aggregation (Zeeuw and Lettinga, 1980). Higher calcium concentrations caused aggregates to develop, which settled three to four times more quickly than those at lower concentrations (Mahoney, 1987).

b) <u>Organic Loading:</u>

Organic loading rate (OLR) is an important parameter that can significantly impact the performance of aerobic granulation. OLR refers to the amount of organic matter (measured as chemical oxygen demand or COD) that is applied to the reactor per unit of time per unit of reactor volume. When the OLR is too low, there may not be enough organic matter to support the growth and formation of the microbial granules. Conversely, when the OLR is too high, the excess organic matter can lead to the accumulation of toxic by-products, such as volatile fatty acids, which can inhibit the formation of the granules. An influent COD of around 600 mg/L is effective for granule formation (Jungles et al., 2014). Therefore, aerobic granulation technology is more appropriate for the treatment of high-strength industrial wastewater. For the treatment of low-strength domestic wastewater, it will be necessary to increase its COD by the addition of external carbon sources such as volatile fatty acids.

Ma et al., 2013 showed in their study that the majority of the biomass in the inner core of granules would experience substrate scarcity due to the diffusion restriction when the COD concentration of wastewater declines, which lowers the food to microbe ratio (F/M). Thus, the inner section of the granules' activities may be severely repressed. As a result, part of the granules broke down into minute debris, which negatively impacted the biomass settling process and washed out of the reactor. Smaller granules were also found in this low OLR condition and these researchers obtained 750 μm size of aerobic granule after long cultivation time.

When the COD concentration was high, the F/M ratio increased, more aerobic granules were produced, and the reactor experienced high biomass retention. (Peyong et al., 2012; López-Palau et al., 2009). According to D. Gao et al. (2011), increased OLRs boosted granulation rates and found that since the start-up date, the granulation rates have dramatically increased. According to their findings, the initial production of aerobic granules may be favored by a relative high specific loading rate of about 0.4 g COD/(g SS. day), and the growth of bigger granules may be stimulated by a higher loading rate than lower rate.

c) Hydrodynamic Shear Force:

High shearing force is one of the main factors for the production of aerobic granules, and hydraulic shear force has been reported to have a considerable impact on the creation of aerobic granules (Liu et al., 2010). The majority of AGS laboratory-scale reactors are built as bubble columns, where shear forces are produced by the aeration rate, which is measured as up-flow superficial velocity and typically falls between 1-2 cms^{-1}. Granules did not form at surface air velocities less than 1.2 cms^{-1} according to past research (Beun et al., 1999; Tay et al., 2001a). Other studies revealed that aerobic granules were not able to grow properly at surface air gas velocities as low as 0.008 cms^{-1}, but were instead seen at velocities as high as 0.025 cms^{-1} (Gao et al., 2010). It was discovered that shear force had a favorable influence on polysaccharide, SOUR, cell surface hydrophobicity, and granule specific gravity.

i. **Shear force induced cell surface hydrophobicity:** Granular sludge has a hydrophobicity that is significantly higher than bio-flocs. In fact, there is strong evidence to show that the hydrophobicity of the cell surface is an important affinity force in the self-immobilization and attachment of cells (Marshall and Gruickshank, 1973; Pringle and Fletcher, 1983). Therefore, it would seem that hydrophobicity might initiate cell to cell interaction and could perhaps be the primary factor in the beginning of granulation. Zhu et al., 2012 found that in the higher hydraulic shear condition, the sludge surface hydrophobicity was also higher where they used the surface gas velocity at the rate of 3.6 cm/s.

ii. **Shear force induced extracellular polymer substances (EPS):** Extracellular polymeric substances (EPS) are sticky materials secreted by cells, and may play an important role in cell adhesion phenomena, formation of matrix structure, microbial physiology, and improvement of long-term stability of granules (Schmidt and Ahring, 1994; Tay et al., 2001c; Liu et al., 2004b; McSwain et al., 2005).

The main components of EPS are proteins and polysaccharides, with smaller amounts of nucleic acids, humic acids, lectins, lipids, and other polymers. A multi-species community of microorganisms is often embedded in an EPS matrix in technological systems such as membrane bioreactors and wastewater treatment processes primarily because they can better acquire substrates and nutrients, maintain hydration, and protect themselves from environmental stresses and predation (Wingender et al., 1999). Biodegradation of organic compounds present in municipal and industrial effluents is most commonly accomplished using the activated sludge process where microorganisms exist as large aggregates held together by EPS (Raszka et al., 2006).

A stable granular structure is more likely to emerge when polysaccharide synthesis is encouraged by shear force (Tay et al., 2001a).

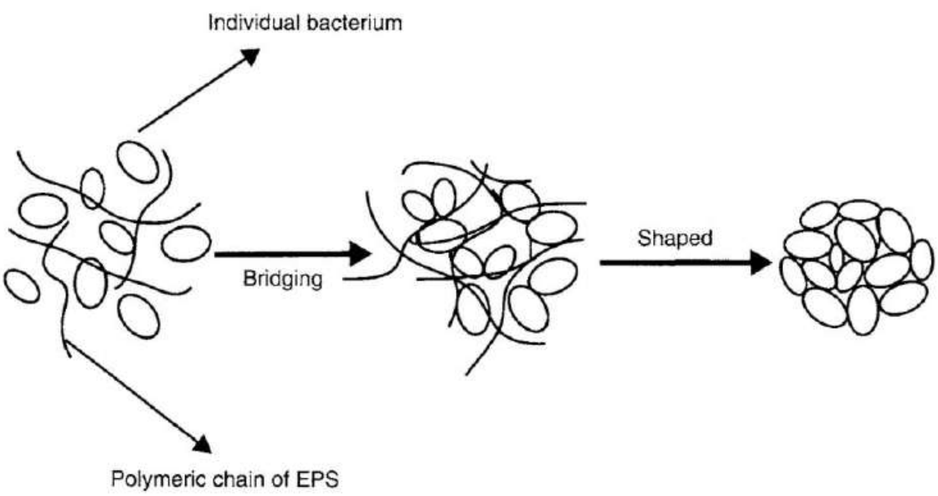


Figure 2.18: Cell to cell stable structure formation by EPS polymeric chain (Liu et al., 2004b).

According to one theory (Liu et al., 2004b), EPS in granules acts as a physical link between adjacent bacterial cells as well as other inert particulate matter (Figure 2.18), allowing them to settle out as aggregates. In their study also by using scanning and transmission electron microscopy, EPS has been found in many kinds of biogranules. EPS may offer extensive surface area for bacterial attachment during the biogranulation process. In addition, extracellular polysaccharide matrices that surround aggregated bacteria may offer places where organic and inorganic materials might be drawn (Yu et al., 2001; Sponza, 2002; Liu et al., 2004b). Evidence suggests that bio granule development is not the result of a random aggregation of suspended microorganisms but rather a micro-biological evolution (Mamouni et al., 1995; Fang, 2000; Tay et al., 2001c).

d) <u>Feast-Famine Regime:</u>

Many researchers investigated that feast-famine regime during SBR cycle operation would be a vital influential factor of aerobic granulation. Each cycle of SBR operation comprises two parts that make up the aeration period: a degradation phase in which the substrate is depleted and follows an aerobic starvation phase that occurs after the external substrate is no longer available (Liu et al., 2007). Under starvation period, bacteria reportedly became more hydrophobic, which probably made adhesion or aggregation easier (Bossier and Verstraete, 1996).

According to previous studies, 3h to 8h cycle time was generally applied for cultivation of aerobic granules, and long starvation phase was identified (Tay et al., 2001a; Liu and Tay, 2006). On the other hand, some researchers (Gao et al., 2011; Liu and Tay, 2008) found that granules are more likely to form under short starvation period, while granules cultivated under long starvation period are reported more stable (Liu and Tay, 2007a). Recently, it was reported that cycle time of 3 h with 140- min starvation led a faster granulation than cycle time of 12 h with 470-min starvation under the same organic loading rate OLR (Wang et al., 2005).

These findings suggest that the rapid granulation in SBR is favored by short starving times. More particular, a brief starvation time might be used to speed up the creation of granules at the start of aerobic granulation, whereas a longer starvation period could improve granule stability (Liu et al., 2016). Starvation would also change the morphology and extracellular polymeric substances (EPS) characteristics of mature aerobic granules (Pijuan et al., 2009).

e) <u>Feeding Strategy:</u>

The four processes of feeding, aeration, settling, and discharge make up the functioning of almost all aerobic granular sludge SBR systems. The average feeding session lasts only a few minutes. As a result, there are less substrate gradients in the liquid phase, which promotes the development of non-filamentous bacteria. There is evidence that adding substrate at various aerobic feeding times can produce significant substrate gradients in SBR and, as a result, enhance sludge's propensity to settle (Am et al., 2003; McSwain et al., 2004). Few researchers claimed that feeding habits foster the growth of non-filamentous bacteria that have high substrate uptake rates during times of high substrate concentration and the ability to store reserve materials during times of starvation (Liu and Liu, 2006; Martins et al., 2004).

f) <u>Reactor Configuration:</u>

Reactor height to the diameter (H/D) ratio is another crucial parameter for aerobic granule formation. According to Awang et al. (2016), a rise in the H/D ratio proportionally accelerates the microorganisms' maximal specific growth rate in an SBR (Awang and Shaaban, 2016). A cylindrical reactor with a height to diameter (internal) ratio of 50 cm/17.6 cm was employed by Linlin et al. (2005) (Linlin et al., 2005). Similarly, Val del Rio et al (2012) have used a SBR for aerobic granule development which had a height to internal diameter ratio of 465 mm/85 mm (Val del Rio et al.,2012).

Since de Kreuk and van Loosdrecht (2004) developed the anaerobic feeding through a stagnant granule bed in a column-type reactor, most studies performed have used this process configuration. In practice, very high height to diameter (H/D) ratio is unpractical and it is difficult to get plug-flow with the risk of getting inhomogeneous accumulation of carbon in the granule bed (Winkler et al., 2011). In previous research, when an AGS reactor was changed from H/D ratio of 9 to 2 after 250 days, the feeding was changed from plug-flow feeding through stagnant granule bed to fast feeding with mixing (Rocktäschel et al., 2013).

g) Cycle Time:

The unique feature of a SBR is its cyclic operation, which leads to the periodical biodegradation phase followed by the aerobic starvation phase in every cycle (Liu and Tay, 2006). The cycle time represents the frequency of solid discharge through the effluent withdrawal or the so-called washout frequency, and it is interrelated to the hydraulic retention time (HRT). Microorganisms are either flushed away or trapped in the reactor through the development of granular sludge when the hydraulic selection pressure in terms of HRT is high (Wang et al., 2006). Faster granulation and more frequent biomass washout are caused by shorter cycle times with larger cycle numbers, which suggests that there is a stronger hydraulic selection pressure at play during that time.

h) Settling Time:

The settling time acts as a major hydraulic selection pressure on sludge. In an SBR, wastewater is treated in successive cycles, each of which lasts several hours. At the end of each cycle, the biomass is settled before the effluent is withdrawn. A short settling time preferentially selects for the growth of rapidly settling bio-particles and the bio-particles with poor settle ability is washed out. Therefore, short settling time has frequently been used in many researches to improve the aerobic granulation in SBRs.

With a long settling time, poorly settling sludge flocs cannot be withdrawn effectively and they may in turn out competence granule-forming bioparticles on nutrients for growth. As a result, in an SBR run at a prolonged settling time, aerobic granulation cannot be accomplished. This shows that an important consideration in the granulation of activated sludge is the settling time (Ni et al., 2008).

The inoculated activated sludge was nearly completely washed away and biomass with improved settling qualities was chosen as the settling period was gradually decreased from 10 to 5 min (Wang et al., 2014). In some studies, the very short settling time often setup in the range 2-5 min. A large amount of biomass washed out, but the filamentous bacteria disappeared after only a short time, the aerobic granules appeared more compact and achieved high performance for processing of N and P removal (Lotti et al., 2014).

Liu et al. (2005a) proposed that there would be a minimum settling velocity, (Vs)min, for bioparticles to be retained in the reactor; and it can be defined as follows:

$$(Vs)_{min} = \frac{L}{Settling\ Time} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \text{(2.1)}$$

Where, L= discharge port in meter

Vs= settling velocity in m/hr.

Equation (2.1) suggests that only bioparticles with a settling velocity larger than (Vs)min can be retained in the system, whereas those with a settling velocity less than (Vs)min could be removed from the reactor. As a result, the heaviest, most spherical aggregates are the ones that settle quickly, whereas the smallest, lightest, and irregularly shaped aggregates take the longest to settle. It seems possible to choose bioparticles based on their settling velocity.

When (Vs)min is smaller than 3.8 m h^{-1}, the suspended bio flocs are dominant in the system. Thus, if the SBR is operated at a (Vs)min below 3.8 m h^{-1}, suspended sludge could not be effectively withdrawn from the reactor. Successful aerobic granulation was also reported at the respective settling velocities of 10.0 m h^{-1} and 16.2 m h^{-1} (Beun et al., 2002). The growth rates of aerobic granules were found to be much lower than that of suspended activated sludge (Yang et al., 2004; Liu et al., 2005b). It should be a reasonable consideration that suspended sludge could easily outcompete aerobic granules due to its faster growth. Such out competition in turn would repress aerobic granulation and eventually leads to the disappearance of the aerobic granular sludge blanket in SBR if suspended sludge is not effectively withdrawn.

i) <u>Exchange Ratio:</u>

The hydraulic loading rate (HLR) and influent substrate concentration both affect the organic loading rate (OLR) for an SBR system. As a result, using a low concentration wastewater as the substrate and using a typical HLR will result in an OLR that is insufficient to prevent the growth of sludge during granulation. However, a rising OLR caused by an increased HLR will make up for the low substrate concentration, which will then work in favor of the granulation process.

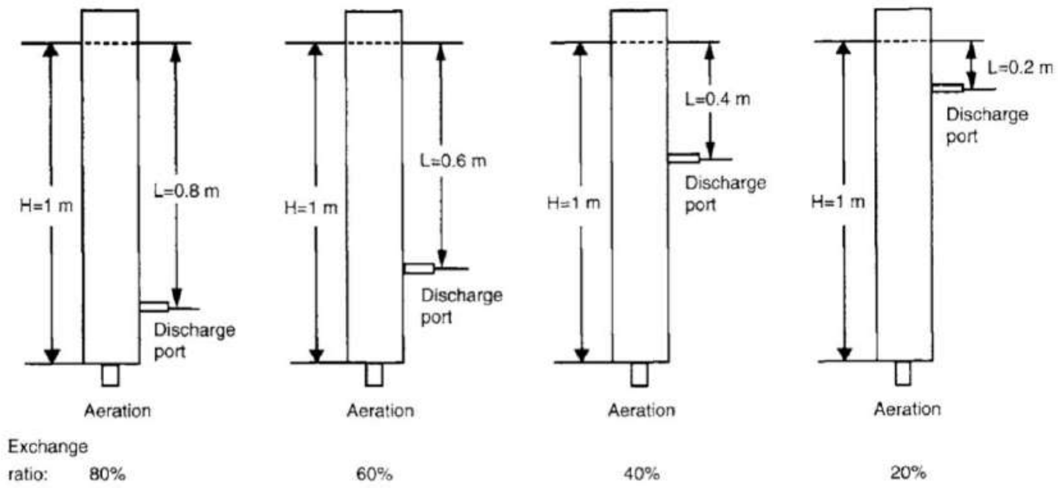


Figure 2.19: Concept of volume exchange ratio (VER) in SBR (Liu et al., 2005a).

The HLR of an SBR is governed by its volume exchange ratio (VER) (Figure 2.19) and cycle period. In the work of Ni et al., (2008), the cycle period was set at 3h because of the low total COD level in the wastewater. In this case, keeping a high level of VER was the only way to elevate its HLR. The VER of the pilot-scale SBR was gradually increased from 50 to 70%. The liquid volume exchange ratio of 50% is commonly used for each cycle of the SBR used for aerobic granule formation (Lochmatter et al., 2014).

2.9 AGS in Sequential Batch Reactor

Aerobic granules are usually developed during wastewater treatment using sequencing batch reactors (SBR) (Etterer and Wilderer, 2001). SBRs have a well defined operational cycle, with each cycle having a potential duration of 3-6 hours.. Each cycle is further divided into definite filling period, aeration or mixing period, settling period and effluent drawing period (Figueroa et al., 2008). The features of SBR are its operation cycle and its phases. Therefore, they are directly related to the mechanism behind granule formation. For instance, short settling period of SBR is of great importance for stable granule formation. Qin et al (2004) investigated the effect of settling time on granule formation. Settling time of 5, 10, 15 and 20 minutes were evaluated. It was observed that a settling time of 5 minute was most effective for granule development (Qin et al., 2004). Since most of the environmental factors liable for granulation are only possible to maintain in SBR reactor, the majority of studies found successful to develop aerobic granulation in SBR.

The SBR consists of a self-contained treatment system incorporating equalization, aeration, anoxic reaction, and clarification within one basin.There are some major steps in a SBR tank which are as following (EPA, 1999; Ardern and Lockett, 1914 and EPA, 1993):

1. Fill – The fill operation consists of adding the waste and substrate for microbial activity. The fill cycle can be controlled by float switches to a designated volume or by timers for multireactor systems. A simple and commonly applied mode to control the fill cycle is based on reactor volume, resulting in fill times inversely related to influent flow rates. The fill phase can include many phases of operation and is subject to various modes of control, termed static fill, mixed fill, and react fill. Static fill involves the introduction of waste influent with no mixing or aeration. This type of fill method is most common in plants requiring nutrient control. In such applications, the static fill will be accompanied by a mixed fill stage such that the microorganisms are exposed to sufficient substrate, while maintaining anoxic or anaerobic conditions. Both mixing and aeration are provided in the react fill stage. The system may alternate among static fill, mixed fill, and react fill throughout the fill cycle.

2. React – The purpose of the react stage is to complete reactions initiated during fill. The react stage may comprise mixing or aeration, or both. As was the case in the fill cycle, desired processes may require alternating cycles of aeration. The length of the react phase may be controlled by timers, by liquid level controls in a multitank system, or when the desired degree of treatment has been attained, verified by monitoring reactor contents. Depending upon the amount and timing of aeration during fill, there may or may not be a dedicated react phase.

3. Settle – Liquid solid separation occurs during the settle phase, analogous to the operation of a conventional final clarifier. Settling in an SBR can demonstrate higher efficiencies than a continuous-flow settler, since total quiescence is achieved in an SBR.

4. Draw – Clarified effluent is decanted in the draw phase. Decanting can be achieved by various apparatus, the most common being floating or adjustable weirs. The decanting capability is one of the operational and equipment limitations of SBR

technology. Adaptation or development of equipment compatible with a fluctuating liquid level is required.

5. Idle – The final phase is termed as the idle phase and is only used in multibasin applications. The time spent in the idle phase will depend on the time required for the preceding basin to complete its fill cycle. Biosolids wastage will typically be performed during the idle phase.

From previous literatures it is found that aerobic granules were successfully cultivated in few pilots scale SBRs and the results of those works shown that reasonable organic loading rate, high H/D ratio, sequencing batch operation and settling time could be necessary factors for effective AGS formation (Sirianuntapiboon et al., 2006).

2.10 Removal of TWW Pollutants by AGS

Microorganisms are protected from inhibitory chemicals by the AGS tight structure. Due to this property, industrial wastewater including poisonous azo-dyes, inhibitory organic compounds, high strength wastewaters from dairy, breweries, live-stock effluents, and toxic heavy metals can be successfully treated by AGS technology (Maszenan et al., 2011; Nancharaiah and Kiran, 2018).

AGS contains qualities that could perhaps encourage the full biomineralization of azo dyes. In particular, the presence of anoxic-anaerobic and aerobic zones within the granules (Winkler et al., 2013) and their improved resilience to large organic loads and harmful recalcitrant chemicals (Franca et al., 2018) may be useful for treating TWW. According to de Kreuk et al. (2005), AGS are frequently cultured in aerobic SBRs where their granular structure generates gradients in DO and substrate concentration along the radial direction. This stratification results in layers of various microenvironments (aerobic, anoxic, and anaerobic) within each AG and enables the co-existence of various types of microorganisms and metabolisms in the same tank.

Manavi et al. (2017) proposed that the channels in AGS could be used to transport organic substrates and dyes into the anaerobic core region of the granules, where (facultative) anaerobic bacteria would reduce azo dyes using the reducing equivalents produced by the oxidation of the organic compounds.

The removal of pollutants by aerobic granular sludge occurs through a combination of physical, chemical, and biological processes. Here are some key mechanisms involved in pollutant removal:

i. **Adsorption:** Aerobic granular sludge has a high surface area, allowing it to adsorb pollutants onto its surface. The granules can adsorb organic compounds, heavy metals, and other contaminants present in wastewater.

ii. **Biodegradation:** The microbial communities within the granular sludge are responsible for the biodegradation of organic pollutants. These microorganisms, such as bacteria and fungi, break down complex organic molecules into simpler forms through enzymatic reactions.

iii. **Filtration and Sedimentation**: The granular structure of the sludge provides a physical barrier that can filter suspended solids, colloidal particles, and other particulate matter from the wastewater. Additionally, the granules settle faster due to their size and density, facilitating the removal of settled solids during the sedimentation process.

iv. **Diffusion Limitation:** The compact structure of the granules creates diffusion limitations within their interior, resulting in the establishment of anoxic or anaerobic microenvironments. This enables the degradation of pollutants that require low oxygen or are recalcitrant to aerobic degradation.

v. **Biofilm Formation:** The microbial aggregates within the granular sludge form a biofilm on the surface of the granules. This biofilm enhances the microbial activity and provides protection against toxic substances, enabling more efficient pollutant removal

2.11 AGS Technology for Synthetic TWW vs Raw TWW

AGS system has been used to treat various wastewater and pollutants, including phenol effluent (Carucci et al., 2009), municipal wastewater (Kreuk and Loosdrecht, 2006), dairy effluent (Arrojo et al., 2004), wastewater from soybean production (Su and Yu, 2005), and wastewater rich in nitrogen and phosphorus (Cassidy and Belia, 2005). Owing to high treatability of tough pollutants by AGS in SBR, previous studies have investigated the suitability of sequencing batch reactors (SBRs) for textile wastewater treatment (Fu et al., 2001; Lourenço et al., 2001; Pasukphun and Vinitnantharat, 2003; Vives et al., 2003; Sirianuntapiboon et al., 2006).

Muda et al. were the first to describe the emergence of AGS employing mixed sludge (sewage, textile mill sludge, and anaerobic granules) as seed in dye-containing synthetic wastewater (Muda et al.,2010). The carbon source of the synthetic wastewater was mainly consist of glucose, ethanol and sodium acetate and for mixed dyes different synthetic dyes were used like Sumifix Black EXA, Sumifix Navy Blue EXF and Synozol Red K-4B. An initial COD of 1270 mg/L, 1020 ADMI (American Dye Manufacturing Index), and an average ammonia content of 38 mg/L were obtained from the mixture. This study showed the effectiveness of intermittent anaerobic and aerobic conditions for treatment of synthetic textile wastewater in SBR. Each cycle of the SBR was 6 h where 5 min filling, 340 min reaction, 5 min settling, 5 min decanting and 5 min idle was maintained. The reaction phase began with an anaerobic phase lasting 40 minutes, was followed by an aerobic phase lasting 130 minutes, a second anaerobic phase lasting another 40 minutes, and a second aerobic phase lasting 130 minutes. Muda et al. also followed the volume exchange ratio as 50 % during the decanting phase. At the conclusion of the study, researchers found that their constructed granule in a sequential batch reactor system effectively removed 94%, 95%, and 62% of the COD, ammonia, and color, respectively. The granules were found to have an average particle diameter of 2.3 ~ 1.0 mm and a maximum size of 4 mm at the final stage of the experiment (day 66).

Muda et al. (2012) again researched on aerobic granular sludge evaluating the operating conditions of the SBR reactor for better treatment of synthetic textile wastewater than their previous work (Muda et al., 2012). In this study they used same seed sludge and feed wastewater for reactor as they did in the work of Muda et al. (2010). The reactor was run in eight-hour cycles that included a 5-minute fill, a 460-minute reaction, a 5-minute settle, a 5-minute decant, and a 5-minute idle period. The reaction mode consists of 230 min of anaerobic and 230 min of aerobic phases. At the end of this study for 72 days COD removal percentage was 93% and color was 56% only. It is important to know that at initial stage the COD and color removal was 76 % and 36 % which increased in the later stage of the experiment due to changes in operating condition. Muda et al. (2012) found the lower removal of color at initial stage of experiment because of low HRT of 8 hours which is not enough to provide higher removal. As the reaction phase was divided into anaerobic and aerobic phases, the actual HRT required for color removal to occur (i.e. anaerobic phase) was only about 4 hours, which may be too short for the dye degradation to occur. Longer HRT, especially on the anaerobic phase is normally required for better color removal (Isik and Sponza, 2008).

Later a lot studies have been done on synthetic textile wastewater treatment by AGS technology in SBR reactor; the below Table 2.4 shows summary of the those few works:

Table 2.4: List of recent AGS works on Synthetic TWW

Reactor Type	Seed Sludge for Acclimatization of AGS	WW type	Inlet COD, mg/L	COD removal, %	Color removal, %	Reference
Intermittent Anaerobic-Aerobic SBR	• Mixed sewage and textile mill sludge and • Anaerobic sludge (from UASB)	Synthetic WW (Mixed dyes)	1270	94	62	Muda. et al., 2010
Anaerobic-Aerobic (An-Ae) SBR	• Mixed sewage and textile mill sludge and • Anaerobic sludge (from UASB)	Synthetic WW (Mixed dyes)	1240	93	56	Muda. et al., 2012
Intermittent Anaerobic-Aerobic SBR	• Mixed sewage and textile mill sludge and • Anaerobic sludge (from UASB)	Synthetic WW (Mixed dyes)	1270	94	87	Muda. et al., 2011
Anaerobic-Aerobic SBR	Conventional Activated Sludge (CAS) (from municipal WWTP)	Synthetic WW (Single dye)	700	85	30~55	Moghaddam and Moghaddam, 2016

Continuing Table 2.4: List of recent AGS works on Synthetic TWW

Reactor Type	Seed Sludge for Acclimatization of AGS	WW type	Inlet COD, mg/L	COD removal, %	Color removal, %	Reference
Anaerobic-Aerobic SBR	AGS (from municipal WWTP)	Synthetic WW (Single dye)	1000 ~ 3000	80	>90	Franca et al., 2015
Aerobic SBR	CAS and Micro mycelium pellets from a white rot fungus	Synthetic WW (Single dye)	Not available	94	>96	Hailei et al., 2010
Aerobic SBR (granulation) and Batch Static-Shaking Reactor (decolorization)	Bacterial cultures from a dye-contaminated area	Synthetic WW (Single dye)	Not available	90 (after 72 h static)	48 ~ 72 (after 48 h)	Chaudhari et al., 2017
Aerobic SBR (granulation) and Batch Static Reactor (decolorization)	Isolates from soil/sludge contaminated with textile dye industrial wastewater	Synthetic WW (Single dye)	64000	56	100 (after 8 ~ 12 hours)	Kolekar et al., 2012

Continuing Table 2.4: List of recent AGS works on Synthetic TWW

Reactor Type	Seed Sludge for Acclimatization of AGS	WW type	Inlet COD, mg/L	COD removal, %	Color removal, %	Reference
Aerobic SBR (granulation) and Batch Microaerophilic Reactor (decolorization)	• CAS with synthetic wastewater containing acetate (for the granulation experiment) or • AGS acclimatized to the dye in 72 h cycle for 19 days (decolorization experiment)	Synthetic WW (Single dye)	16 (TOC)	79 ~ 95	89 ~ 100	Sarvajith et al., 2018
i. Anaerobic-Aerobic SBR ii. Intermittent Anaerobic-Aerobic SBR	Conventional Activated Sludge (CAS) (from municipal WWTP)	Synthetic WW (Single dye)	1000	80	80 ~ 85	Mata et al., 2015
Anaerobic-Aerobic SBR with Static vs Plug Flow Feeding	Stored AGS, previously acclimatized to Acid Red 14 (Franca et al., 2015)	Synthetic WW (Single dye)	1000 ~ 3000	68 ~ 90	66 ~91	Franca et al., 2017
Anoxic-aerobic SBR	CAS from a municipal WWTP, acclimatized to Methylene Blue	Synthetic WW (Single dye)	500	93	56	Ma et al., 2011

Table 2.5: List of recent AGS Works on Raw TWW

Reactor Type	Seed Sludge for Acclimatization of AGS	WW type	Inlet COD, mg/L	COD removal, %	Color removal, %	Reference
Intermittent Anaerobic-Aerobic SBR	• Sterilized sludge from a textile WWTP and • Acclimatized mixed bacterial culture (Alcaligenes sp., Bacillus sp., Acinetobacter sp. and Stenotrophomonas sp.) added in every cycle filling stage	Sterilized raw Textile Wastewater	200 ~3000	80	90	Ibrahim et al., 2010
Anaerobic-Aerobic SBR	• Sterilized sludge from a textile WWTP and • Acclimatized mixed bacterial culture (Alcaligenes sp., Bacillus sp., Acinetobacter sp. and Stenotrophomonas sp.) added in every cycle filling stage	Sterilized raw Textile Wastewater	800 ~ 1000	46	61	Kee et al., 2014

Continuing Table 2.5: List of Recent AGS Works on Raw TWW

Reactor Type	Seed Sludge for Acclimatization of AGS	WW type	Inlet COD, mg/L	COD removal, %	Color removal, %	Reference
Anaerobic-Aerobic SBR	Conventional Activated Sludge (CAS) (from municipal WWTP)	• Synthetic WW (granulation period) • Synthetic WW-7% + raw Textile WW-93% (decolorization period)	1200 (initial)	68	73	Manavi et al., 2017
Sequencing Batch Biofilm Granular Reactor	Conventional Activated Sludge (CAS) (from municipal WWTP)	Mixed Raw Municipal (30%) + Raw Textile Wastewater (70%)	249 ± 65	82.1 ± 3.6	33.9 % ~ 52.6 %	Lotito et al., 2014
Sequencing Batch Biofilm Granular Reactor	Conventional Activated Sludge (CAS) (from municipal WWTP)	Raw Textile Wastewater	688 ± 280	55 ~78	0 ~ 60	Lotito et al., 2012

Real TWW's composition is more complex and variable than that of manufactured TWW. Real TWW treatment with AGS has only received a few investigations, and this application has not yet been thoroughly investigated. The list of works on real TWW is shown on Table 2.5.

Lotito et al. investigated the effectiveness of a sequencing batch biofilter granular reactor (SBBGR) for the pre-treatment of real TWW and the treatment of mixed municipal-textile wastewater (Lotito et al. 2012; Lotito et al. 2014). In the study of Lotito et al., 2012 a PLC controlled SBBGR was introduced for the reactor of RTWW treatment. They ran the reactor (Fig.11) in cycles with a filling phase lasting a few minutes (depending on the volume of the influent), a contact phase (aerobic degradation), during which the filled wastewater was continually aerated and recycled through the reactor bed, and a final drawing phase lasting 15 minutes.

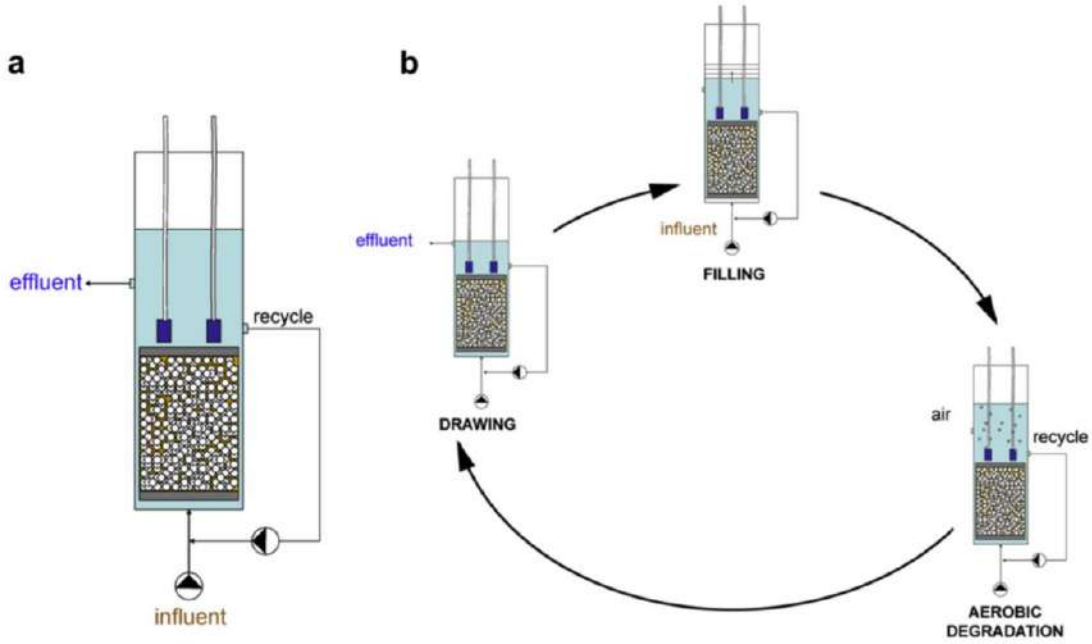


Figure 2.20: SBBGR sketch and cycle: a) two clearly distinct parts can be individuated: the microbial bed at the bottom and the liquid phase at the top; b) each cycle consists in a filling phase followed by the contact period (aerobic degradation) and a final drawing phase (Lotito et al. 2012).

After the operation of 200 days the COD and color removal was not found to the level to meet the local discharge limit. Hence, the researchers suggested this reactor as an effective pre-treatment for dyeing textile wastewater before discharge into municipal sewer system. However, the AGS properties and granulation process were not deeply investigated in their studies.

AGS development in actual TWW was examined by Ibrahim et al. and Kee et al. (Kee et al., 2015; Ibrahim et al., 2010). The real TWW was sterilized before to feeding, and four bacteria that degrade color were utilized to create the AGS, so its use appears to be challenging. AGS created in synthetic media decomposed after prolonged exposure to actual TWW, according to Manavi et al. (Manavi et al., 2017). Microbial community analysis was not used in any of these two investigations.

In reality, there aren't many researches looking into how the microbial community and nitrifying bacteria activity alter when the sludge is granulated in a real TWW treatment. AGS in TWW also takes a lot of time to form, which could limit its use. According to earlier research, the production of AGS in synthetic TWW took 21–87 days while it took real TWW between 6–300 days (Kee et al., 2015; Ibrahim et al., 2010; Ma et al., 2011; Bashiri et al. 2018; Kolekar et al., 2012).

Chapter 3

METHODOLOGY

3.1 Introduction

This chapter depicts the experimental method of this study extensively. The following section will cover details of raw waste water sample collection and its analysis, SBR reactor set-up for AGS, granulation phase, sample collection during granulation and its analysis, decolorization phase of the experiment and AGS performance check, idle phase and AGS performance check after its reactivation from idle period.

3.2 Raw TWW Sample Collection

A textile knit dyeing factory was selected for collecting sample to conduct this research work. The factory name is "Fakhruddin Textile Mills Ltd." and it is located at Kewa, Ghorgaria, Master Bari, Gazipur (Figure 3.1). Fakhruddin Textile Mills Ltd. is the pioneer of synthetic fabric dyeing in Bangladesh since it is established in 2002.

Table 3.1: Production units and capacity of FTML

Production Units	Capacity
Knitting	25 tons/day
Dyeing	30 tons/day
Seamless Knitting	100,000/month
Seamless Dyeing	5000 pcs/day
Printing	40,000 pcs/day
Embroidery	3000 million stitch/day
Washing	8000 pcs (pigment dyeing)/day 15,000 pcs (acid wash, stone wash, special wash etc.)/day

Since the focus of this research work is on the tough textile wastewater like a dyeing plant so the waste water collected from this factory would be quite suitable for this study. The location of this factory was quite away from Dhaka city and to keep the collected sample intact, air-conditioned transport was used from site to BUET lab.

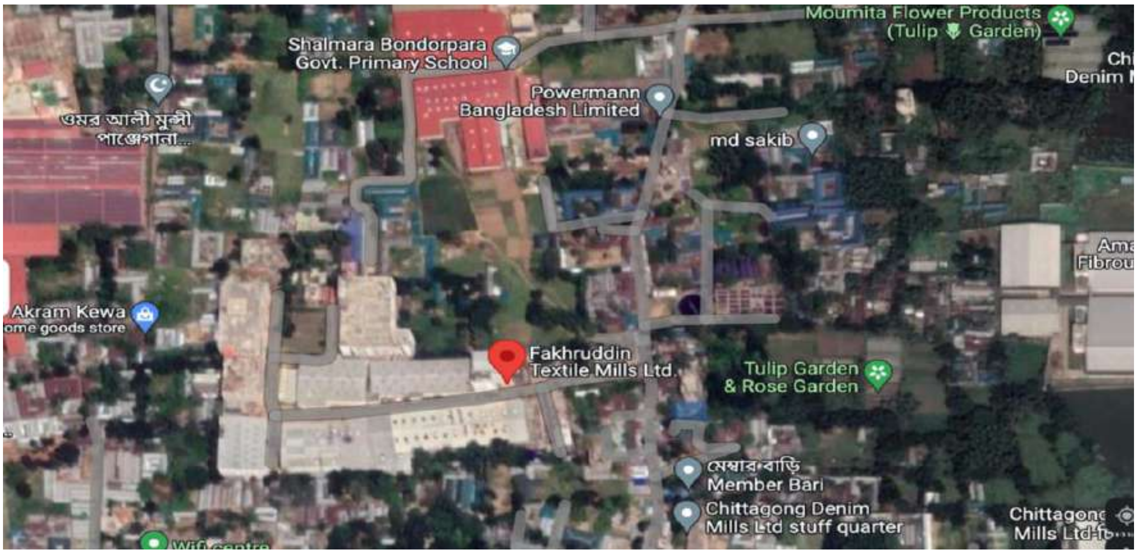


Figure 3.1: Dyeing plant selected for sample collection

This textile plant already has a biological ETP which is based on activated sludge process and sample wastewater was collected from their ETP inlet (Figure 3.2) which is exactly from neutralization tank. This helped to avoid the pH correction step in the lab before starting the experiment (Figure 3.3 b).

Figure 3.2: ETP Inlet of Fakhruddin Textiles

Figure 3.3: a) Collected sample; b) Sample collection point (Neutralization Tank of ETP)

The research experiment was conducted in the environmental engineering laboratory in department of Civil Engineering, BUET, Dhaka, Bangladesh.

3.3 Raw TWW Characterization

The raw textile waste water was characterized by determination of pH, COD, TDS, TSS, Color and ammonia.

(a) Chemical Oxygen Demand (COD):

APHA standard method 5220 D (APHA, 2012) was followed to determine the COD of collected sample raw textile wastewater. There are two ranges of reagent vial (HACH) for analyzing COD of wastewater; low range (LR< 150 mg/L) and high range (HR>150 mg/L). Only HR vial was selected for the sample RTWW because it was already informed at time of collection from source that COD is greater than 150 mg/L. Sample RTWW was added (2 ml) to vial tube and it was shaken vigorously for well mixing and reaction with potassium dichromate vial tube. The tube with sample and reagent was then put in COD reactor (Hach 45600) and heated to 150 °C for 2 hours (Figure 3.4 a). Finally, after 2 hours the tube was cooled at room temperature and then placed in spectrophotometer (DR 6000, HACH) for measuring the COD value of sample (Figure 3.4 b). Spectrophotometer was calibrated with blank HR vial before testing with sample WW vial.

(a) (b)

Figure 3.4: Apparatus used for COD test; a) COD Reactor; b) Spectrophotometer (HACH-DR 6000)

(b) Total Suspended Solid (TSS):

Total suspended solid (TSS) was measured for the raw wastewater according to APHA standard method 2540 D (APHA, 2012). Gravimetric method was applied to determine TSS of the sample TWW. 100 mL sample was taken for drying in oven at 105 deg C for 24 hours. Then calculating the weight before and after drying, TSS data was found for the sample TWW. The apparatus using in this test are shown in Figure 3.5 a, b and c.

(a) (b) (c)

Figure 3.5: TSS and TDS measuring a) Oven; b) Weight Machine and c) Desiccator

(c) Total Dissolved Solid (TDS):

Total dissolved solid (TDS) of sample wastewater was determined by gravimetric method following the APHA standard method 2540 C (APHA, 2012). The procedures were almost same as for TSS measurement. Here, 100 mL sample TWW was to be filtered first prior (Figure 3.6) to keep in oven for drying the sample. Finally, TDS was calculated from weight difference between empty beaker and beaker with dried sample.

Figure 3.6: Gravity filtration for TDS measurement

(d) Determination of Color:

Color was one of the important parameters for textile dyeing wastewater. As the visual color of the raw sample was quite dark so dilution was required before final sampling for color measurement. Raw sample was diluted ten times and also filtered by micron filter paper before taking reading for color measurement. Spectrophotometer (DR 6000, HACH) was used to collect the color measurement of raw TWW and all readings were taken at 455 nm wavelength. Distill water was used as blank sample to calibrate the spectrophotometer at the same wavelength at which sample raw TWW was to be tested.

(e) Determination of pH:

pH data of raw TWW was taken by portable pH meter (Multi 3500i Handheld Multimeter, WTW) in laboratory.

(f) Determination of Ammonia:

Spectrophotometer (DR 6000, HACH) was the instrument to measure ammonia in water. Sample water was reacted with three types ammonia reagents. Finishing the reaction within few minutes the ammonia reading was taken by putting diluted sample. Calibration was also done with blank reacted sample following raw water testing.

3.4 Design of Reactor for AGS

Reactor configuration and its operating conditions are major factors for cultivation of aerobic granular sludge. Reactor is to be designed based on the operation philosophy of sequential batch reactor (SBR).

A graduated measuring cylinder was used to consider as the SBR tank for this experiment. Different studies showed variety of height/diameter (H/D) ranges for reactor configurations to form AGS; ranges of H/D were 2.5, 4, 8, 16, 24 and even 30 in one of the studies (Kong et al., 2009). In this study, H/D was selected as 5.0 and volume of the cylinder was 2000 mL. This H/D was a constant variable during the entire experimental work because 2000 mL size cylinder from local market has fixed diameter and length, here the height was 42.16 cm and diameter was 8.38 cm. The following materials are needed to start up the SBR operation:

a) Seed sludge
b) Sample raw textile waste water
c) Air blower or aerator (SB-348A) (Figure 3.8)
d) Air diffuser (Figure 3.8)
e) Thermometer (Figure 3.9)
f) Beaker-500 mL (Figure 3.7)
g) Glass rod
h) Heating Mantle (Gerhardt) (Figure 3.9)
i) Measuring Cylinder as SBR Reactor – 2 L volume (Figure 3.7)

Figure 3.7: RTWW in beaker and measuring cylinder as SBR Reactor

Figure 3.8: Air Blower/Aerator and Air Diffusers (Ceramic Type-Stone shaped)

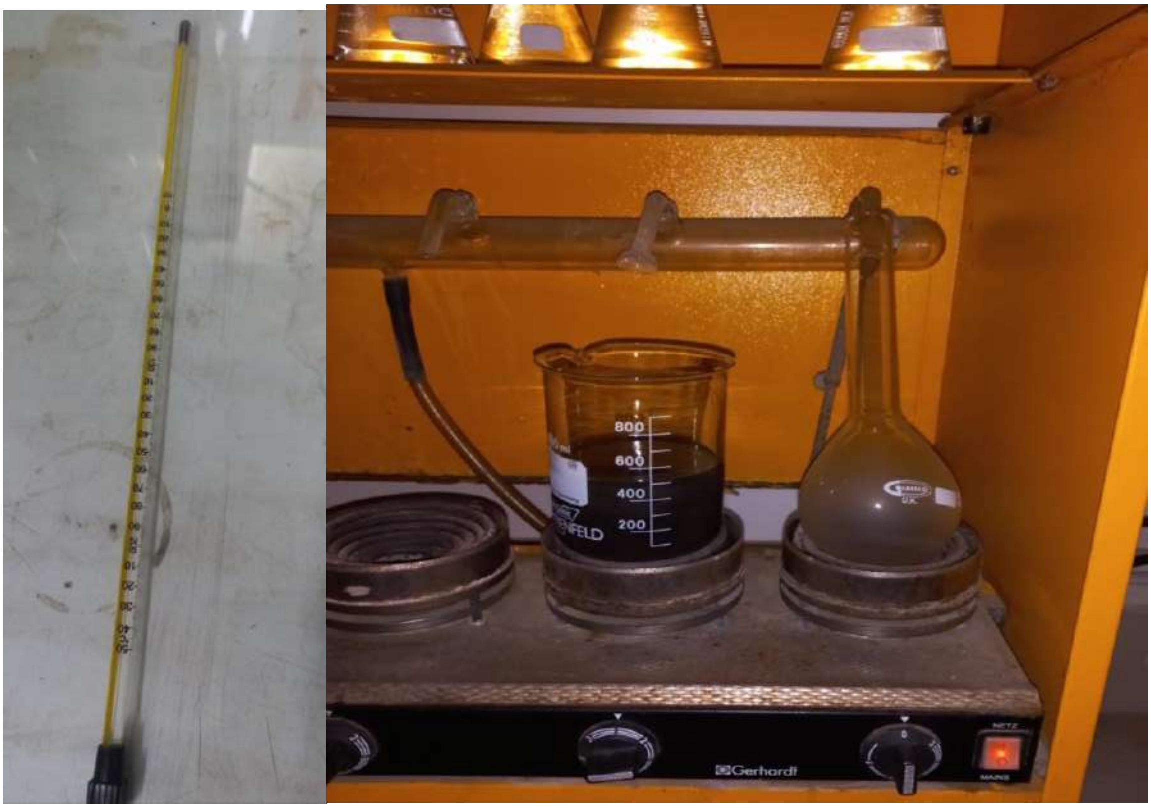


Figure 3.9: Thermometer and heating mantle used for the experiment

The RTWW sample was preserved in refrigerator to intact the quality. Every time before feeding in the reactor, the RTWW has to be heated (Heater- Gerhardt) to rise the temperature from 7^0C to 25^0C~30^0C. Temperature maintaining is necessary to culture any kind of microorganisms in the biological reactor.

Activated sludge from (Fakhruddin Textile Mill) dyeing textile factory was taken to use it as seed sludge for initiating bacteria acclimatization in this SBR. The MLSS (mixed liquor suspended solid) of this seed sludge was 4600 mg/L initially which was measured in laboratory by gravimetric method. The SBR was inoculated with 1000 mL seed sludge and was fed with 1000 mL RTWW for commencing the reactor operation of this study. In this study, no chemical and nutrients were added in the seed sludge to culture desired microorganism.

3.5 Aerobic Granulation Stage in SBR

This study used aerobic mode of operation to culture bacteria for aerobic granular sludge (AGS). Aerobic granules are self-immobilized aggregates of microorganisms which are able to remove pollutants like COD, TSS, Color from textile wastewater. The mechanism of microorganism aggregation is self-motivated process without any carrier support, instead some selection pressure needs to be applied for occurring the self-accumulation between bacteria to bacteria.

After the reactor set up for SBR, granulation stage was started by operating SBR cycles. Each cycle is considered 4~5 hours where 95% time will be covered by aeration phase and 5 % is anaerobic and settling phase (Figure 3.10). Due to lack of automatic system the cycles were run manually maintaining the available working time in the BUET-Environmental Laboratory. The operation was done batchwise where each cycle was conducted in each day and taken all the experimental data accordingly.

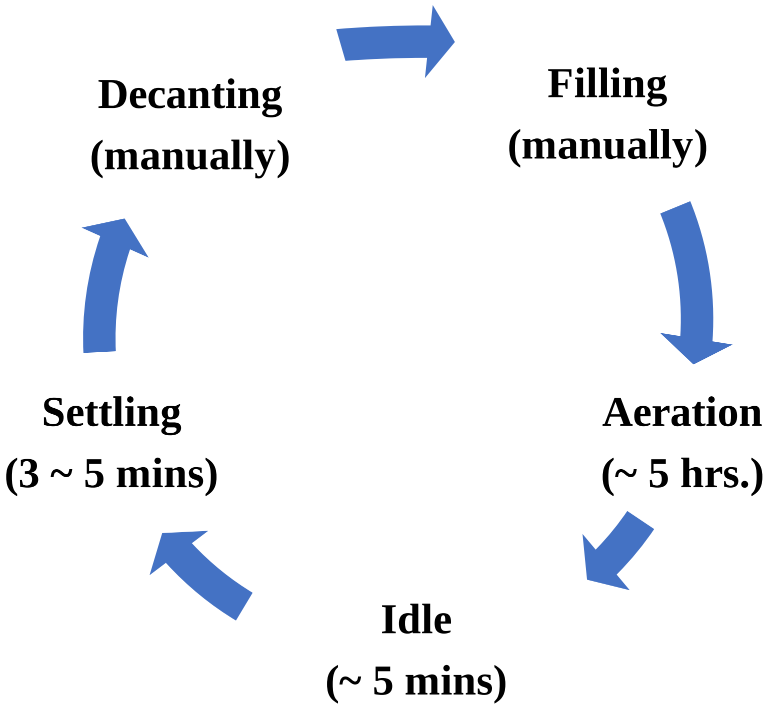


Figure 3.10: Aerobic SBR cycle steps during granulation phase of the study

The steps of the SBR cycle operation for granulation phase is as following:

1. At first, feeding of RTWW was done without the help of any pump. The RTWW was poured from the beaker to the top of the cylinder by hand. No fixed time of feeding was possible to maintain as this was manual. The feeding rate was followed according to the volume exchange ratio (VER) which was to be maintained during granulation phase.

2. Then, reaction started with aeration in the SBR. In SBR, the aeration was done with aerator (SOBO, Model: SB-348A) and diffusers which was ceramic made stone type. Each aerator has two outputs and port diameter each output is 4 mm. The capacity of aerator is 2 x 4 L/min and pressure is 2 x 0.2 bar. The diffusers were put at the bottom of the cylinder for proper mixing and diffusion of oxygen throughout the reactor (Figure 3.11). The aeration step was quite longer around four to five hours when the SBR cycles were operated for granulation period only.

3. After finishing the aeration, idle condition was to be continued for few minutes. Idle condition means no feeding and no action is done for reaction. No aeration and mechanical stirring were done here, a glass rod was used to stir the mixture slowly for keeping the idle phase and restricting the sludge to be settled down.

4. Following the idle step, settling is to be occurred for few minutes (Figure 3.12 and 3.13). During this step no aeration or mixing was done. Here, settling time was very short in order to enhance the environment for fast growing microbial aggregates so that they outcompete the slow growing microbes in the reactor. This situation is called the selection pressure for granulation stage.

5. Final step is decanting which is done manually by gravity force (Figure 3.14). Decanting volume of RTWW was followed according to the VER during granulation period. There are four discharge point at the level of 500 ml, 1000 ml, 1500 ml and bottom of the cylinder.

Figure 3.11: SBR Cycle aeration-reaction step during granulation phase of the study

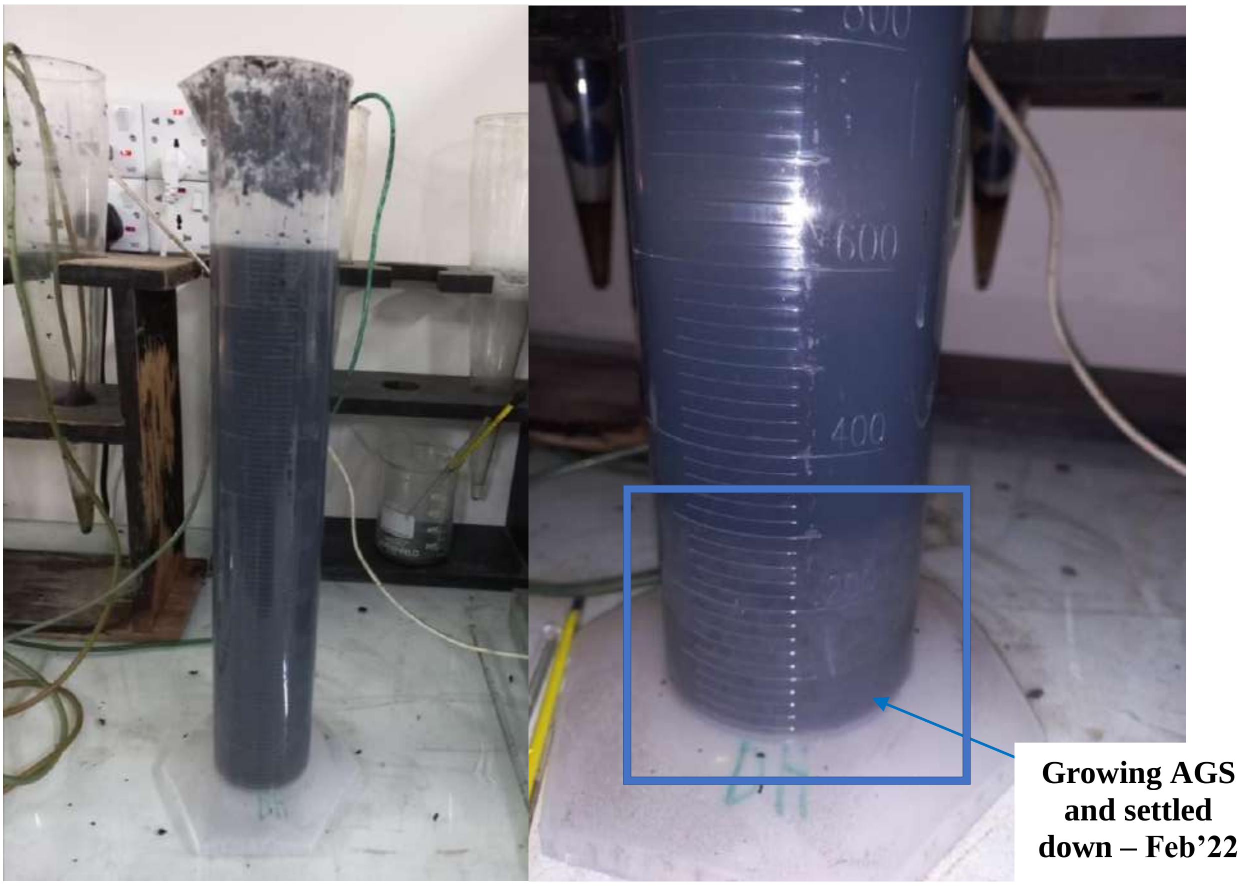


Figure 3.12: SBR cycle settling step during granulation phase of the study

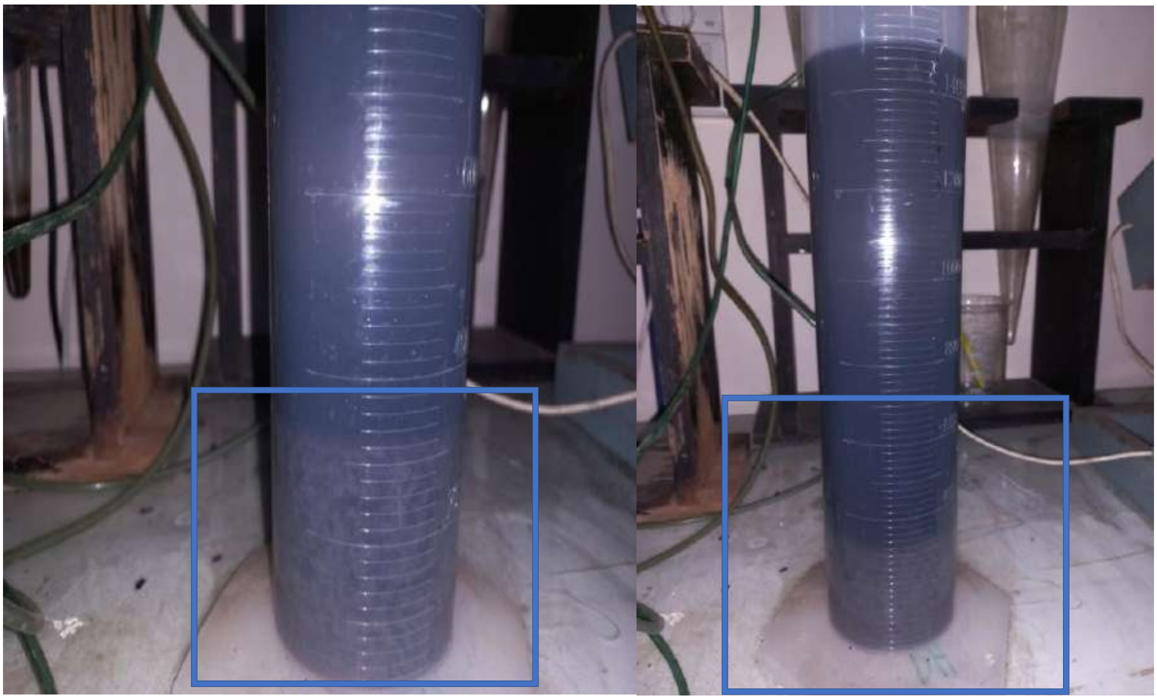


Figure 3.13: Development of AGS settled bottom of reactor during granulation phase of the study (March’22)

Figure 3.14: SBR cycle decanting step during granulation phase of the study

During the bacteria acclimatization period reaction time has to be increased, settling time has to be decreased and aeration flow has to be increased gradually whereas volume exchange was kept as 25%. These operational conditions enhanced the high hydrodynamic shear forces and short settling time which has impact to grow dense microbial aggregates for AGS formation. Total twenty-six (26) number of cycles were conducted for granulation stage. The different operating conditions for different cycles are summarized in below Table 3.2.

Figure 3.13 shows the developing AGS during the experiment of granulation, aggregated biomass was visible at the time of granulation operation cycles.

Waste water analysis were done for COD, TSS and color during granulation process to observe progress on cultivation.

Table 3.2: Operating conditions for granulation

Cycle Serial Number	Total Duration of each cycle, hrs.	Aeration Step, hrs.	Idle Step, mins.	Settling Step, mins.	Aeration Rate, L/min	HRT, hrs.	VER, %	Time of Experiment
Cycle-1	4.42	4	20	5	8	17.68	25	28.12.2021
Cycle-2	4.42	4	20	5	8	17.68	25	29.12.2021
Cycle-3	4.25	4	10	5	8	17.00	25	02.01.2022
Cycle-4	4.25	4	10	5	8	17.00	25	03.01.2022
Cycle-5	4.90	4.75	5	4	8	19.60	25	04.01.2022
Cycle-6	4.90	4.75	5	4	8	19.60	25	05.01.2022
Cycle-7	4.90	4.75	5	4	8	19.60	25	09.01.2022
Cycle-8	4.88	4.75	5	3	12	19.52	25	11.01.2022
Cycle-9	4.88	4.75	5	3	12	19.52	25	17.01.2022
Cycle-10	4.88	4.75	5	3	12	19.52	25	18.01.2022
Cycle-11	5.13	5	5	3	12	20.52	25	23.01.2022
Cycle-12	4.88	4.75	5	3	12	19.52	25	25.01.2022
Cycle-13	4.88	4.75	5	3	12	19.52	25	29.01.2022
Cycle-14	4.88	4.75	5	3	12	19.52	25	06.02.2022
Cycle-15	5.13	5	5	3	12	20.52	25	07.02.2022
Cycle-16	5.13	5	5	3	12	20.52	25	08.02.2022
Cycle-17	5.13	5	5	3	12	20.52	25	13.02.2022
Cycle-18	5.13	5	5	3	12	20.52	25	14.02.2022
Cycle-19	5.13	5	5	3	12	20.52	25	15.02.2022
Cycle-20	5.13	5	5	3	12	20.52	25	28.02.2022
Cycle-21	5.13	5	5	3	12	20.52	25	06.03.2022
Cycle-22	5.13	5	5	3	12	20.52	25	07.03.2022
Cycle-23	5.13	5	5	3	12	20.52	25	08.03.2022
Cycle-24	5.13	5	5	3	12	20.52	25	13.03.2022
Cycle-25	5.13	5	5	3	12	20.52	25	14.03.2022
Cycle-26	5.13	5	5	3	12	20.52	25	15.03.2022

3.6 An-Ae SBR Operation for Treatment of RTWW

When granulation stage ends and it is apparent that granular sludge is developing, the treatment experiment of RTWW is to be performed in the reactor. RTWW was fed to reactor filled of AGS without any addition of nutrient. The SBR operation for treatment of RTWW was almost same as it was done in granulation stage. The procedures for all steps of SBR operation is following the same as in granulation period only the operating conditions are different (Figure 3.15).

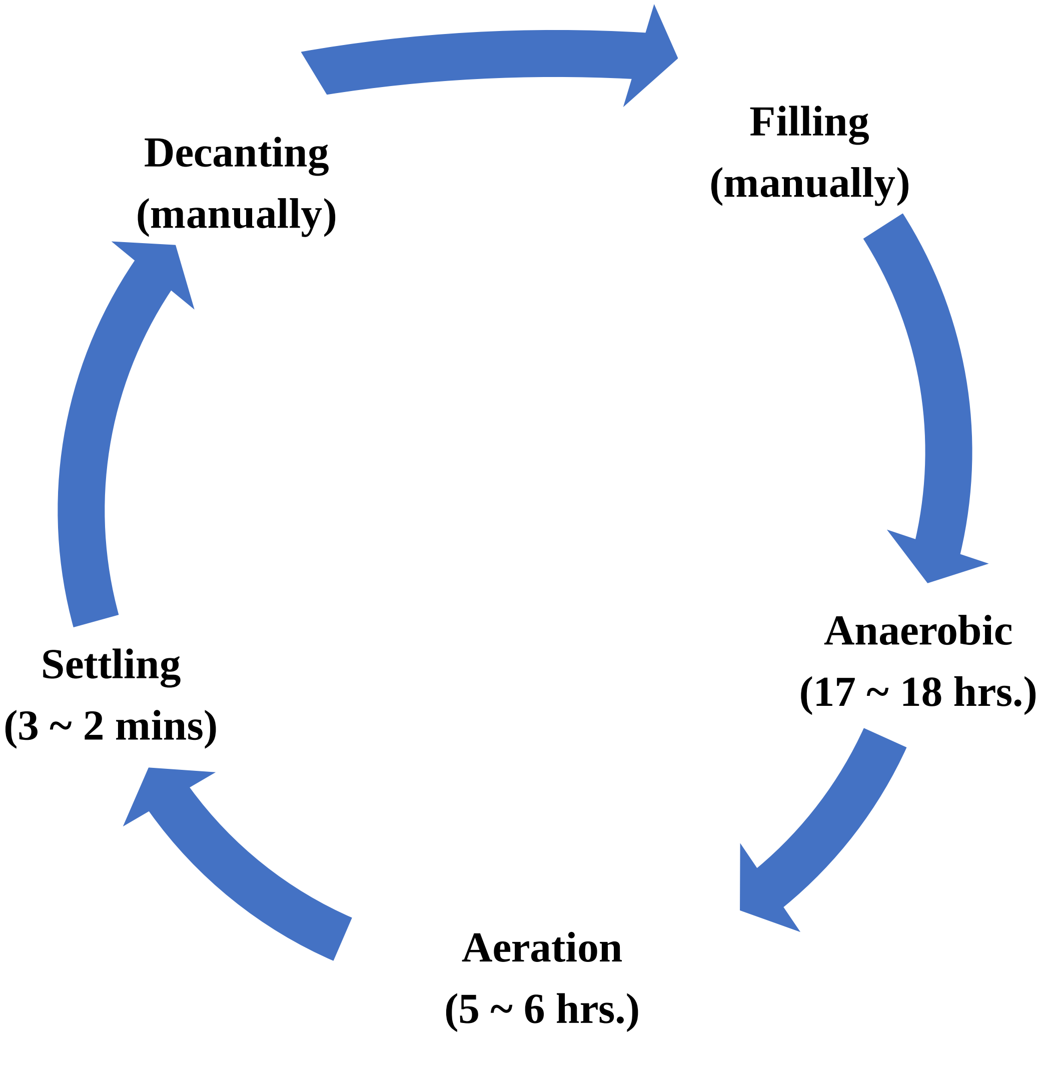


Figure 3.15: SBR cycle steps during An-Ae SBR operation for RTWW treatment

The steps of the SBR cycle operation for An-Ae SBR operation phase is as following:

1. At first, feeding of RTWW was done without the help of any pump. The RTWW was poured from the beaker to the top of the cylinder by hand. The feeding rate was followed according to the volume exchange ratio (VER) which was varied during this treatment phase.

2. After filling the reactor with RTWW, the reactor was operated in anaerobic phase. Anaerobic condition was maintained in absence of air supply including slow mixing of RTWW in the reactor (Figure 3.16). The anaerobic phase was kept longer for decolorization of RTWW and mixing was done by rotary laboratory shaker (DLAB, SK-L330-Pro). The rotation speed of the shaker was maintained between 150 ~170 rpm.

3. Then, reaction starts with aeration in the SBR. In SBR, the aeration was done with aerator (SOBO, Model: SB-348A) and diffusers which was ceramic made stone type. Each aerator has two outputs and each output port diameter is 4 mm. The capacity of aerator is 2 x 4 L/min and pressure is 2 x 0.2 bar. The diffusers were put at the bottom of the cylinder for proper mixing and diffusion of oxygen throughout the reactor. The aeration step was kept around four to five hours while the SBR cycles were operated for An-Ae SBR operational phase.

4. Following the aeration phase, settling is to be occurred for few minutes. During this step no aeration or mixing was done. Here, settling time was very short and fast settling of granular sludge was obtained at the bottom of reactor.

5. Final step decanting was done manually by gravity force. Decanting volume of RTWW was followed according to the VER (Table 3.3) during An-Ae SBR operational period.

Figure 3.16: SBR cycle anaerobic step during An-Ae SBR operation for treatment of RTWW

Each cycle of the SBR for this experiment was considered 22.5 ~23 hours where 70% is anaerobic phase and 30% is aerobic phase. During this experiment, operation conditions has to be changed like volume exchange ratio from 25% to 50% and aeration rate is increased or decreased to evaluate the reactor performance. No nutrient or additional seeds were added in this reactor for running the decolorization and treatment experiment of RTWW. After the fifth cycle of this SBR experiment, MLSS of the reactor was tested by gravimetric method and data was found as 10,648 mg/L. Therefore, it is apparent that the sludge developing in the reactor, gradually became denser and more aggregated from the initial stage when the biomass concentration was 4600 mg/L.

Figure 3.17: SBR cycle aeration steps during An-Ae SBR operation for RTWW treatment

The operating conditions of this stage is as following Table 3.3.

Table 3.3: Operating conditions during An-Ae SBR operation for treatment of RTWW

Cycle Serial Number	Total Duration of each cycle, hrs.	Anaerobic Step, hrs.	Aeration Step, hrs.	Settling Step, mins.	Shaking Speed of Anaerobic Step, rpm	Aeration Rate, L/min	HRT, hrs.	VER, %	Time of Experiment
Cycle-1	23.05	17	6	3	150	12	92.20	25	21.03.2022
Cycle-2	23.05	17	6	3	160	12	92.20	25	22.03.2022
Cycle-3	22.53	17.25	5.25	2	160	12	90.13	25	27.03.2022
Cycle-4	22.53	17.25	5.25	2	160	12	90.13	25	28.03.2022
Cycle-5	23.28	18	5.25	2	160	12	93.13	25	29.03.2022

Figure 3.18: SBR cycle decanting steps during An-Ae SBR operation for treatment of RTWW

3.7 Idle Period and Reactivation Stage for AGS

After finishing the few cycles of An-Ae SBR operation, formed granular sludge is to be kept idle at below 7°C for couple of months. AGS ability of RTWW treatment has to be tested using the similar operational conditions used in granulation stage. Meanwhile, AGS long term operational efficiency after idle period can be evaluated by performing the de-colorization and treatment experiment with RTWW.

The procedures for idle condition of formed granular sludge is as following:

a. Granular sludge was collected from the reactor after total 31 cycles of both granulation and de-colorization stages.

b. The sludge from reactor bottom was collected and settled for 5 mins ~ 8 mins (Figure 3.19 a).

c. Finally, 100 ml sludge was extracted and dense granular sludge were found.

d. This collected 100 ml sludge with 200 ml distilled water was placed in a 500 ml sized reagent glass bottle.

e. Then, the collected AGS with distill water was stored in refrigerator at 6.5 deg C for 60 days.

f. During storing time no aeration facility was maintained and no nutrient was added to the sludge.

g. After 60 days the 100 mL AGS was restored to the reactor where 1900 ml RTWW was fed to fill the 2000 ml reactor volume (Figure 3.19 b).

h. Reactivation of long-time stored AGS was accomplished by running the SBR reactor at the same operating conditions which was applied during granulation stage. The procedures for operation of SBR to reactivate the idle AGS was followed as same as granulation period (Section 3.5).

(a)

(b)

Figure 3.19: (a) AGS extraction before idle period and (b) AGS condition after idle period

The operating conditions of this reactivation stage is as following Table 3.4.

Table 3.4: Operating conditions for reactivation stage

Cycle Serial Number	Total Duration of each cycle, hrs.	Aeration Step, hrs.	Idle Step, mins.	Settling Step, mins.	Aeration Rate, L/min	HRT, hrs.	VER, %	Time of Experiment
Cycle-1	5.07	5	2	2	12	20.28	25	06.06.2022
Cycle-2	5.30	5.25	1	2	12	21.20	25	07.06.2022
Cycle-3	4.80	4.75	1	2	12	19.20	25	08.06.2022
Cycle-4	5.30	5.25	3	2	12	21.20	25	12.06.2022
Cycle-5	5.91	5.83	3	2	12	23.64	25	13.06.2022
Cycle-6	5.91	5.83	3	2	12	23.64	25	14.06.2022

Continuing Table 3.4: Operating conditions for reactivation stage

Cycle Serial Number	Total Duration of each cycle, hrs.	Aeration Step, hrs.	Idle Step, mins.	Settling Step, mins.	Aeration Rate, L/min	HRT, hrs.	VER, %	Time of Experiment
Cycle-7	5.41	5.33	3	2	12	10.82	50	19.06.2022
Cycle-8	5.58	5.50	3	2	12	11.16	50	20.06.2022
Cycle-9	5.58	5.50	3	2	12	11.16	50	21.06.2022
Cycle-10	5.60	5.50	3	3	8	12.73	44	02.07.2022
Cycle-11	5.10	5	3	3	8	11.59	44	03.07.2022
Cycle-12	5.10	5	3	3	12	11.59	44	04.07.2022
Cycle-13	5.30	5.25	3	2	12	12.05	44	23.07.2022
Cycle-14	5.58	5.50	3	2	12	11.16	50	24.07.2022
Cycle-15	5.58	5.50	3	2	12	11.16	50	25.07.2022
Cycle-16	5.58	5.50	3	2	12	11.16	50	26.07.2022
Cycle-17	5.58	5.50	3	2	12	11.16	50	06.08.2022
Cycle-18	5.58	5.50	3	2	12	11.16	50	07.08.2022
Cycle-19	5.58	5.50	3	2	12	11.16	50	08.08.2022
Cycle-20	5.58	5.50	3	2	12	11.16	50	20.08.2022
Cycle-21	5.58	5.50	3	2	12	11.16	50	21.08.2022

During the reactivation experiment the decanted RTWW was tested for checking the AGS ability to treat the RTWW. From experiment it was shown that after twenty-one cycles the results of AGS activity was found suitable for further treatment of wastewater.

3.8 An-Ae SBR Operation after Reactivation of AGS

When the test result of RTWW during reactivation test was found satisfactorily for AGS ability, the An-Ae SBR experiment for treatment of RTWW was performed for few more cycles. The operating conditions and procedures of this experiment were almost same as section 3.6, only few conditions were changed in this phase of experiment. The operation conditions of this stage are as described in Table 3.5.

Table 3.5: Operating conditions of An-Ae SBR operation for RTWW treatment

Cycle Serial Number	Total Duration of each cycle, hrs.	Anaerobic Step, hrs.	Aeration Step, hrs.	Settling Step, mins.	Shaking Speed of for Anaerobic Step, rpm	Aeration Rate, L/min	HRT, hrs.	VER, %	Time of Experiment
Cycle-1	23.53	18	5.5	2	170	8	53.48	44	22.08.2022
Cycle-2	23.70	17.67	6	2	170	8	53.87	44	23.08.2022
Cycle-3	23.78	18.25	5.5	2	160	16	47.57	50	28.08.2022
Cycle-4	23.78	18.25	5.5	2	170	16	47.57	50	29.08.2022
Cycle-5	24.03	18.50	5.5	2	170	12	48.07	50	30.08.2022
Cycle-6	23.53	18	5.5	2	170	12	47.07	50	10.09.2022

The temperature and pH were always kept within acceptable range for all above experiments. Temperature was maintained between 25^{o}C to 30^{o}C. The pH for any raw wastewater was neutralized up to 6.5 ~7.5 for SBR operation. Both temperature and pH maintaining is important for healthy microbial reaction and survival of them in sludge.

Chapter 4
RESULTS AND DISCUSSIONS

4.1 Introduction

In this study, the raw textile wastewater quality was analyzed for few numbers of samples required to conduct the experimental work. Result of granulation formation for AGS and effectiveness of AGS technology for real textile wastewater treatment will be discussed in detail in this section. FTIR and SEM analysis for the formed granules will be demonstrated also in the following discussion.

4.2 Raw Wastewater Characterization

The study mainly was conducted on raw textile wastewater from a knit dyeing textile plant since color removal and tough pollutant removal from textile factories are a burning question in Bangladesh. Also, other type wastewater including treated tannery water from Savar CETP and another raw textile wastewater from a denim dyeing textile factory were collected, for evaluation of AGS effectiveness. The analysis test for all sample wastewater was done in BUET Environmental Lab following the standard method described in section 3.3.

Table 4.1: List of raw textile dyeing wastewater characteristics

Sl No.	Date of Sample Collection	COD, mg/L	Color, Pt-Co	TSS, mg/L	TDS, mg/L	Type of WW	Source of WW
1	27.12.2021	1159	3620	170	3522	Knit Dyeing	ETP Inlet
2	23.01.2022	1233	4090	160	3860	Knit Dyeing	ETP Inlet
3	23.02.2022	1117	2930	107.3	4227.7	Knit Dyeing	ETP Inlet
4	21.03.2022	897	3230	72.7	3992.6	Knit Dyeing	ETP Inlet
5	29.06.2022	797	3130	106.6	4519.6	Knit Dyeing	ETP Inlet
6	04.08.2022	987	2900	122.3	3103.9	Knit Dyeing	ETP Inlet

Though Table 4.1 does not include data of BOD_5, first two days BOD_5 tests were done to check the biodegradability of the RTWW. BOD_5 for the sample collected on 27^{th} December, 2021 and 23rd January, 2022 was found 128 mg/L and 340 mg/L respectively. These results showed that BOD_5/COD was 0.1 and 0.3 respectively for those two days which indicated low biodegradability of the RTWW.

The standard limits of effluent discharge to surface water or environment are governed by Department of Environment (DoE) imposing the regulations in ECR'23 (latest published on 5th March, 2023). All textile industries must discharge their effluent maintaining the COD, BOD_5, TSS, TDS, Color limit according to this ECR'23 (Table 4.2 shows the list for the parameters). Among all of them, the most stringent limit recently imposed for the color value. In the previous version of ECR'97, there was no limit for color applied on textile sectors. Synthetic dyes are one of the harsh components for textile wastewater. In addition, without using chemical treatment it is very tough to remove those colors from the effluent of textile factories. Therefore, this study did an experiment to remove the tough pollutants including color from the raw textile waste water of a knit dyeing factory.

Table 4.2: Regulations for textile effluent in ECR'23

Sl No.	Parameters	Unit	Standard Limits (for discharge in surface water)
1	pH	--	6~9
2	Color	Pt-Co	150
3	Temperature	^{o} C	Not more than 5^{o} from surface water
4	Suspended Solids (SS)	mg/L	100
5	BOD_5 at 20^{o} C	mg/L	30
6	COD	mg/L	200
7	TDS	mg/L	2100 (not applicable for sea water discharge)
8	Oil and grease	mg/L	10

4.3 Formation of Aerobic Granulation Sludge (AGS)

AGS formation was initiated by introducing the acclimatization of bacteria with the seed sludge, which was activated sludge from a textile effluent treatment plant. As discussed in section 3.5, the required favorable conditions were applied to the SBR. Selection pressure was considered to form AGS in this research work, they are hydrodynamic shear force as superficial velocity and settling time.

Superficial gas velocity is an estimate of how quickly a fluid passes through an object (e.g., a pipe) or a porous medium (e.g., gravel). The formula to calculate this is as below:

$$u_s = \frac{Q}{A} \quad \text{.. (4.1)}$$

Where, u_s =is the superficial velocity of a given phase, in centimeters/second (cm/s)

Q =is the volume flow rate of the phase, in centimeters cubed/second (cm^3/s)

A= is the cross-sectional area of the pipe or porous medium, the fluid is flowing through in centimeters squared (cm^2)

In this work, cross sectional area of the reactor follows the below formula:

$$A = \pi r^2 \quad \text{..(4.2)}$$

The selection pressure data for this study is described in Table 4.3.

Table 4.3: List of selection pressures data during granulation experiment

Cycle Serial Number	Cross Sectional Area A, cm^2	Air Volume Q, L/min	Air Volume Q, cm^3/s	Superficial Air Velocity u_s, cm/s	Settling Step, mins.
Cycle-1	55.1	8	133.3	2.4	5
Cycle-2	55.1	8	133.3	2.4	5
Cycle-3	55.1	8	133.3	2.4	5
Cycle-4	55.1	8	133.3	2.4	5
Cycle-5	55.1	8	133.3	2.4	4
Cycle-6	55.1	8	133.3	2.4	4
Cycle-7	55.1	8	133.3	2.4	4
Cycle-8	55.1	12	200	3.6	3
Cycle-9	55.1	12	200	3.6	3
Cycle-10	55.1	12	200	3.6	3
Cycle-11	55.1	12	200	3.6	3

Continuing Table 4.3: List of selection pressures data during granulation experiment

Cycle Serial Number	Cross Sectional Area A, cm^2	Air Volume Q, L/min	Air Volume Q, cm^3/s	Superficial Air Velocity u_s, cm/s	Settling Step, mins.
Cycle-12	55.1	12	200	3.6	3
Cycle-13	55.1	12	200	3.6	3
Cycle-14	55.1	12	200	3.6	3
Cycle-15	55.1	12	200	3.6	3
Cycle-16	55.1	12	200	3.6	3
Cycle-17	55.1	12	200	3.6	3
Cycle-18	55.1	12	200	3.6	3
Cycle-19	55.1	12	200	3.6	3
Cycle-20	55.1	12	200	3.6	3
Cycle-21	55.1	12	200	3.6	3
Cycle-22	55.1	12	200	3.6	3
Cycle-23	55.1	12	200	3.6	3
Cycle-24	55.1	12	200	3.6	3
Cycle-25	55.1	12	200	3.6	3
Cycle-26	55.1	12	200	3.6	3

From literature survey, it was found that superficial air velocity for granulation should be more than 1.6 cm/s (Muda et al., 2010) and for this study it was kept 2.4 cm/s and 3.6 cm/s (Table 4.3) which was considered to create the suitable condition for developing aerobic granules in the reactor. The settling time was maintained according to the shortest possible time 5 minute to 3 minute for favoring the granulation from the seed sludge.

4.3.1 Analysis of RTWW Parameters During Granulation Stage

The RTWW from a knit dyeing textile plant was fed to the reactor during the granulation stage. The treated RTWW from the SBR operation was analyzed at the time of granulation experiment.

The sample of treated wastewater from SBR outlet was not collected for testing from first cycle. Since the selection pressures for developing AGS was applied to the SBR, the initiation of granulation from flocculant seed sludge needed some time for acclimatization in the reaction environment. After 10^{th} cycle of granulation operation, few samples of treated wastewater were collected for testing the effect of pollutant removal in the SBR during granulation stage. The testing data for treated RTWW by AGS is described in Table 4.4.

Table 4.4: RTWW pollutant removal percentage at SBR outlet during granulation experiment

Parameters / Sampling Days	COD % Removal	TSS % Removal	Color % Removal	TDS % Removal	Cycle Number for Sampling
1 (11^{th} Day)	74.46	64.71	79.14	7.16	Cycle-11
2 (16^{th} Day)	74.29	98.90	51.96	no removal	Cycle-16
3 (19^{th} Day)	73.32	99.10	50.12	no removal	Cycle-19
4 (23^{rd} Day)	71.89	99.30	32.08	no removal	Cycle-23

It is found from the RTWW water analysis report that COD removal percentage was within the range between 71 % ~ 75 %, TSS removal range was 65% ~ 99% and color removal was 32% ~ 79%. There was no progressive performance in terms of TDS removal for granulation stage of operation. Residual amount of WW parameters was measured in treated effluent where residual COD was 296 ~ 329 mg/L, with an average value of 312.5 ± 16.5 mg/L; residual TSS was 60 mg/L to less than 5 mg/L and residual color was 755 ~ 2040 Pt-Co at 455 nm wavelength, with an average of 2002.5 ± 37.5 Pt-Co at 455 nm wavelength (Table 4.5). Therefore, from the graphical representation in Figure 4.1 the treatment results of COD and TSS shows a uniformity to demonstrate the effectiveness of SBR operation during granulation stage. On the contrary, color removal efficiency is manifesting a descending trend at this stage of SBR operation.

Table 4.5: List of RTWW parameters at the outlet of AGS reactor during granulation stage

Parameters	COD In (mg/L)	COD Out (mg/L)	TSS In (mg/L)	TSS Out (mg/L)	Color In (Pt-Co)	Color Out (Pt-Co)	Cycle Number for Sampling
1 (sampling day-1)	1159	296	170	60	3620	755	Cycle-11
2 (sampling day-2)	1233	317	160	1.8	4090	1965	Cycle-16
3 (sampling day-3)	1233	329	160	1.4	4090	2040	Cycle-19
4 (sampling day-4)	1117	314	107.3	0.8	2930	1990	Cycle-23

In this stage of experiment, the data of TSS in the SBR treated WW shows the tendency of granular sludge formation in the reactor due to the higher settleability of suspended particles in reactor. At the starting of granulation period, both superficial air velocity and settling time were applied to the reactor as selection pressure for developing granular sludge from flocculant seed sludge. The settling time was kept 5 minutes from first cycle and gradually it was decreased to 3 minutes up to 26th cycle of granulation operation. As a result, rapidly settling bioparticles were favored by this short settling time, while bioparticles with a weak ability to settle were washed out. The significance of short settling time in granulation operation was also demonstrated in the study of Ni et al., 2008. Therefore, this factor enhanced the very high removal of suspended particle (TSS) from the wastewater throughout the operation cycles of granulation in SBR and consistent progress on higher removal efficiency was found from 11th cycle to 23rd cycle.

The higher HRT sustained and strong biological activity in the reactor system were to blame for the good COD removal efficiency even at the steepest and shortest settling time (Melesse et al., 2020). In this study during granulation period HRT was maintained 17 hours to 20 hours for maintaining required retention time to biodegradation of organic pollutants while in the previous study done by Melesse et al., 2020, HRT was kept 20 hours for good biomass retention in the reactor.

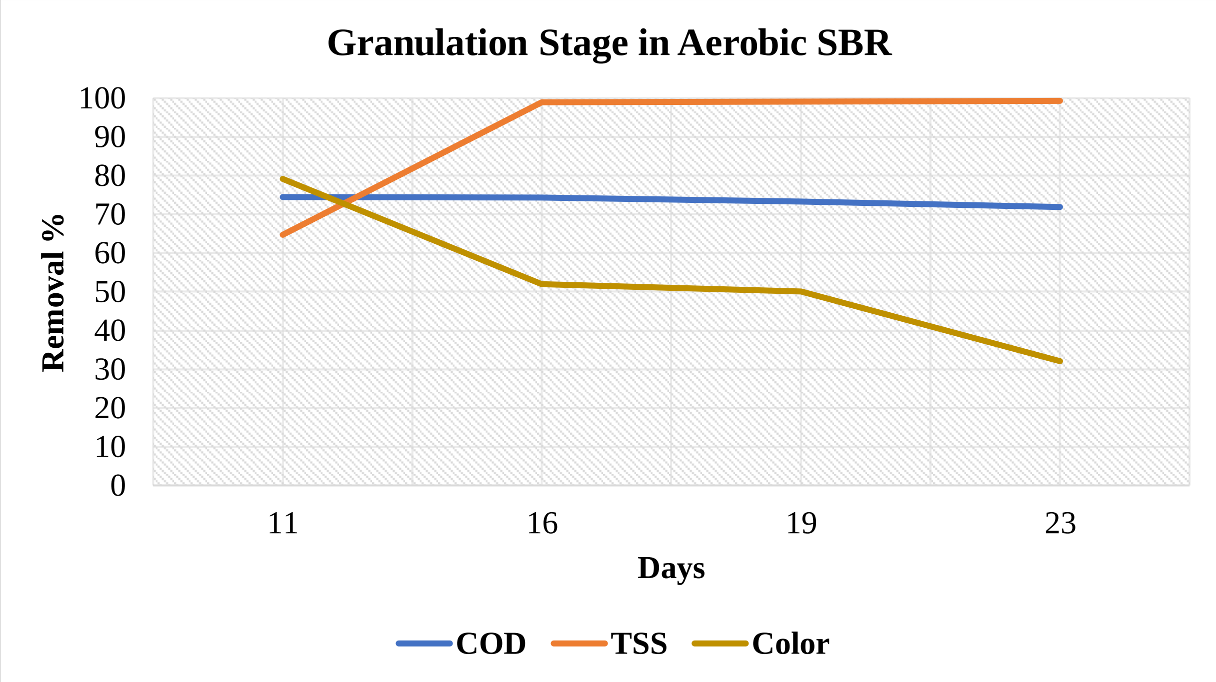


Figure 4.1: Graphical view for RTWW parameters at the outlet of SBR during granulation stage.

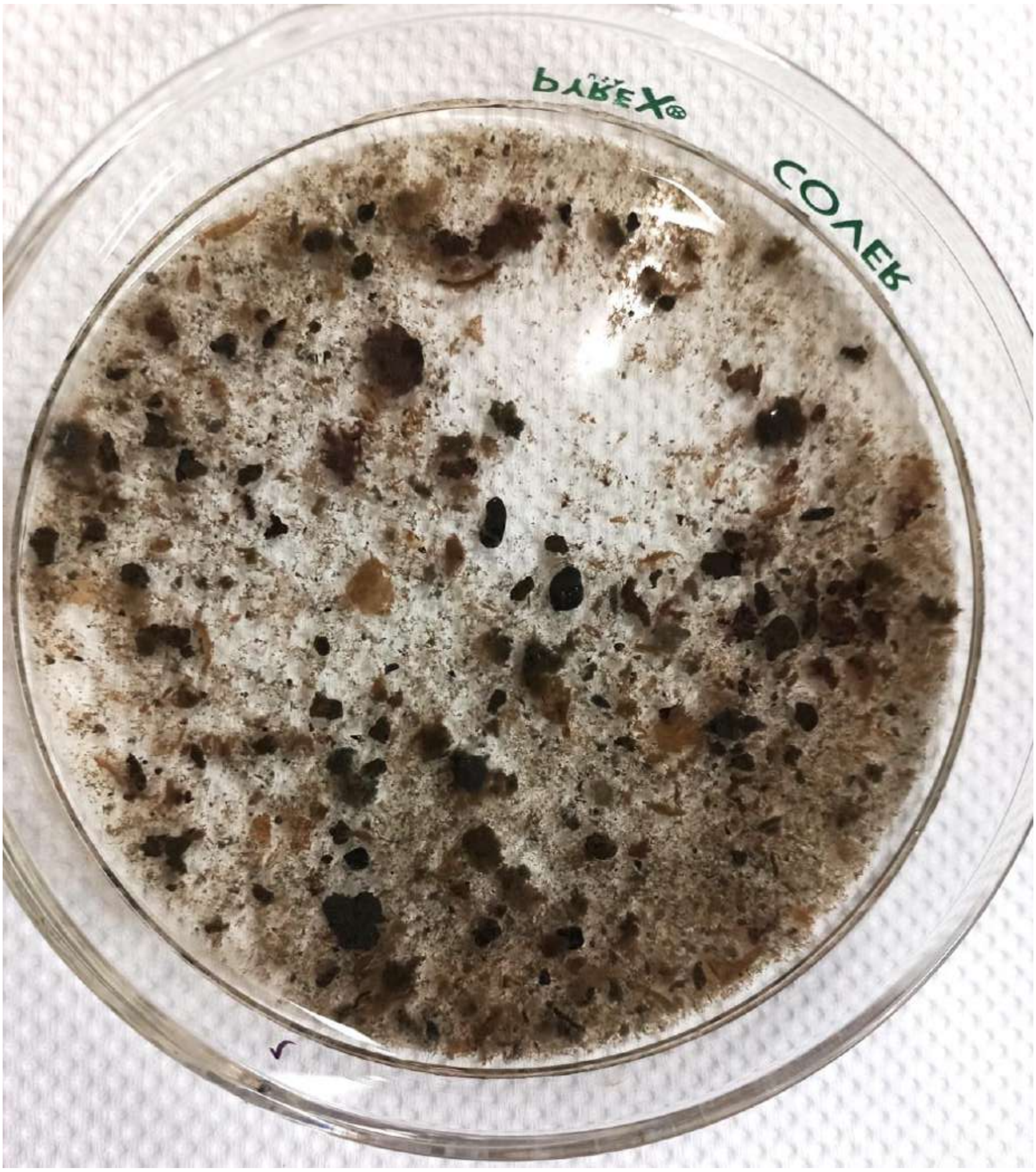
The average of color removal was 53.25% and this low percentage of color removal may be due to insufficient adaptation time. It takes more time to build up a suitable population of organisms that degrade the dyes in the reactor since dye compounds are resistant and challenging to decompose (Muda et al., 2010).

The absorption of color into the sludge biomass throughout the experiment may also influence the inconsistency of color removal. Few researchers have reported the absorption of color into the sludge biomass (Otero et al., 2003; Sirianuntapiboon and Srisornsak, 2007).

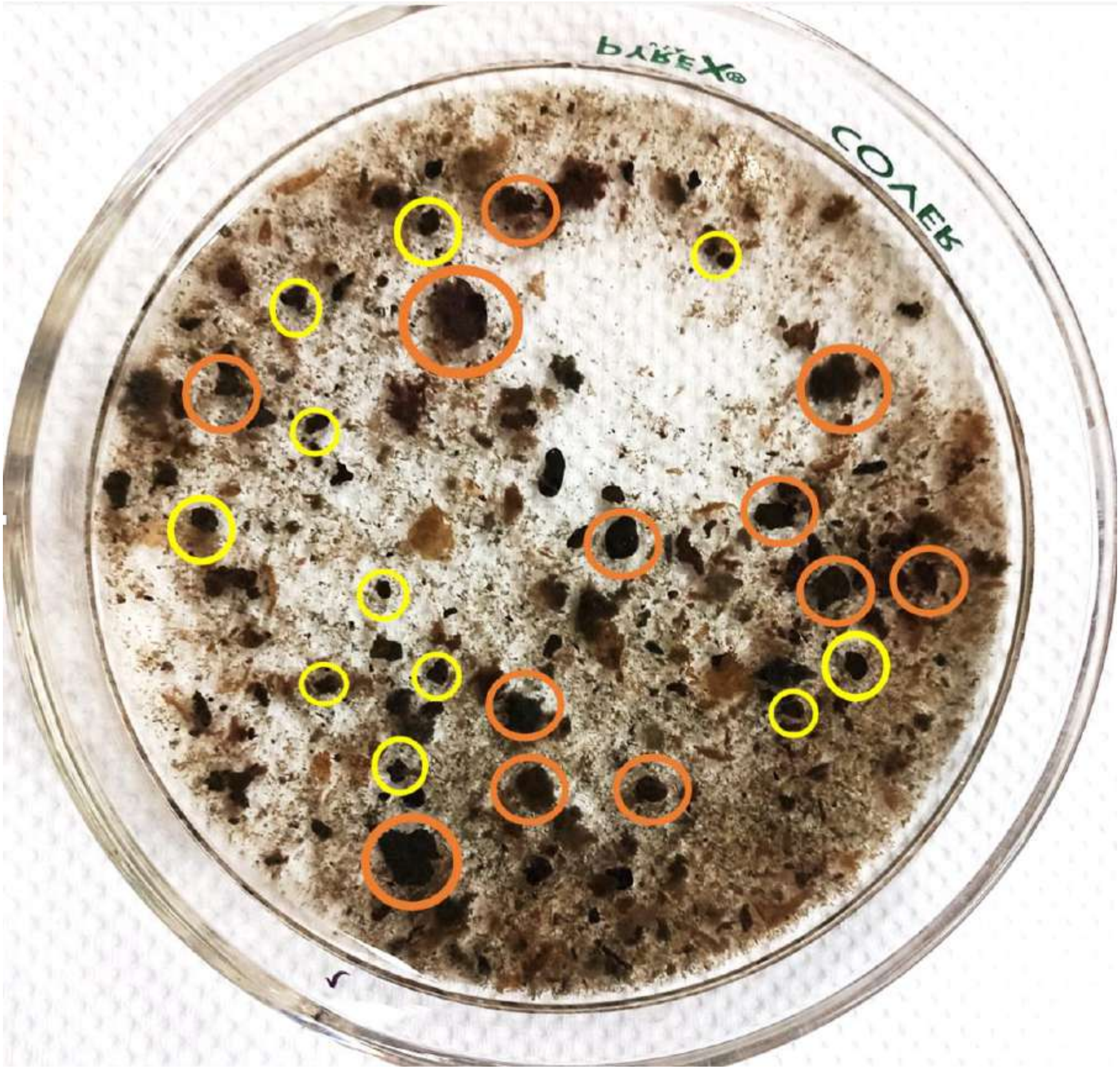
4.3.2 Status of Aerobic Granular Sludge (AGS) in SBR and Its Properties

After twenty-six number of cycles in batchwise operation for granulation phase, few cycles were done for conducting the decolorization test on treatment of RTWW. On the above section 4.3.1 the results of TSS are indicating good settling properties of sludge which developed in the reactor with times, initiated from the seed activated sludge. The good settling nature of activated sludge can be achieved only after a long waiting settling time in clarifier. Whereas, in this study the settling time was too short comparison with conventional activated sludge process. Therefore, it is apparent that granular sludge is developing in the reactor throughout batch operation cycles of granulation stage.

The granular sludge properties were again observed after carrying out the idle phase and reactivation phase cycles of SBR operation. At this stage visual evaluation was done with naked eye. The formed sludge was settled down for 2 minutes at the bottom of reactor. Then, it was collected to a glass petri dish to check the granules condition, its size and shape (Figure 4.2 a).



(a)



(b)

Figure 4.2: (a) and (b) are pictures taken for developed aerobic granules in the reactor

The cultivated aerobic granules were shaped quite circular, though the shape of all granules were not uniform. The size of granules was found from 1 mm to 3 mm diameters, in Figure 4.2 (b) orange marked granules have larger size diameter and yellow marked granules are categized as smaller size diameter. The below Table 4.6 shows granular size of AGS found in previous studies for textile wastewater.

Table 4.6: A brief list of AGS granular size found in different studies

Sl No.	Reactor System for AGS	WW Type	Granule Size	Reference
1	Aerobic SBR	RTWW	1 mm ~ 3 mm	Author of this Study
2	Aerobic SBR	STWW	1 mm ~ 2 mm	Kolekar et al., 2012
3	Aerobic SBR	STWW	1.2 mm ± 0.4	Sarvajith et al., 2018
4	An-Ae SBR	Sterilized RTWW	3.3 mm ± 0.9	Kee et al., 2014
5	An-Ae SBR	RTWW + STWW	0.3 mm~0.5 mm	Manavi et al., 2017
6	SBBGR	RTWW + Municipal WW	0.5 mm	Lotito et al., 2012

Microscopic view was analyzed as well for the aerobic granules to observe the structure of granules. The pictures taken during microscopic observation were shown as below in Figure 4.4 and 4.5.

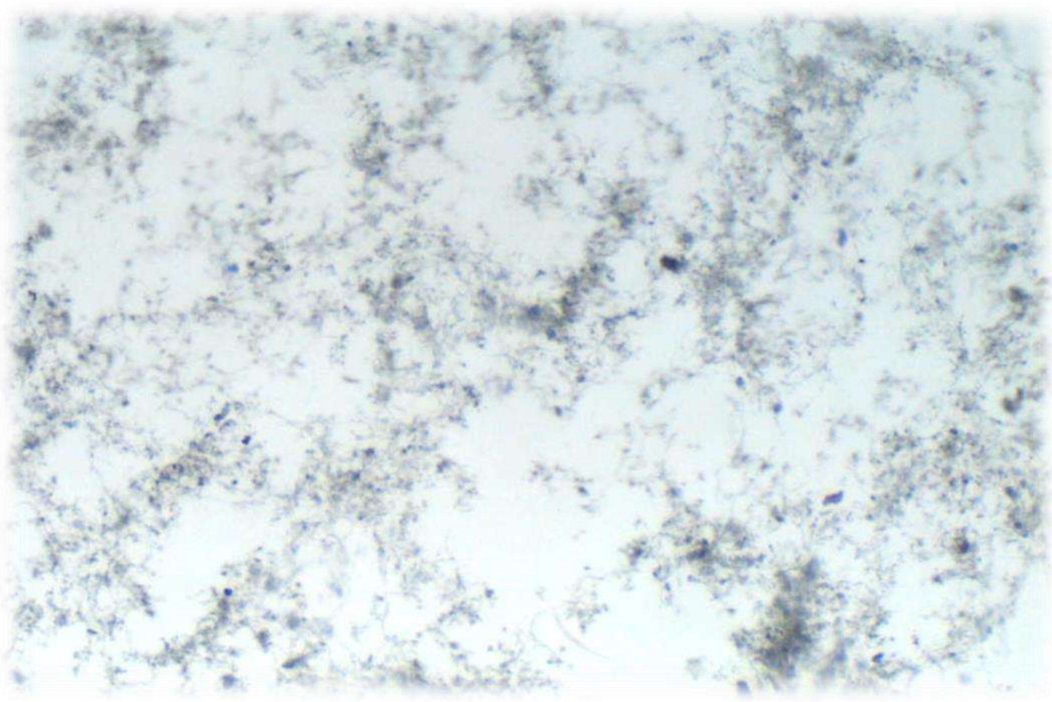


Figure 4.3: Microscopic view for seed activated sludge taken as 10 X magnifying resolution

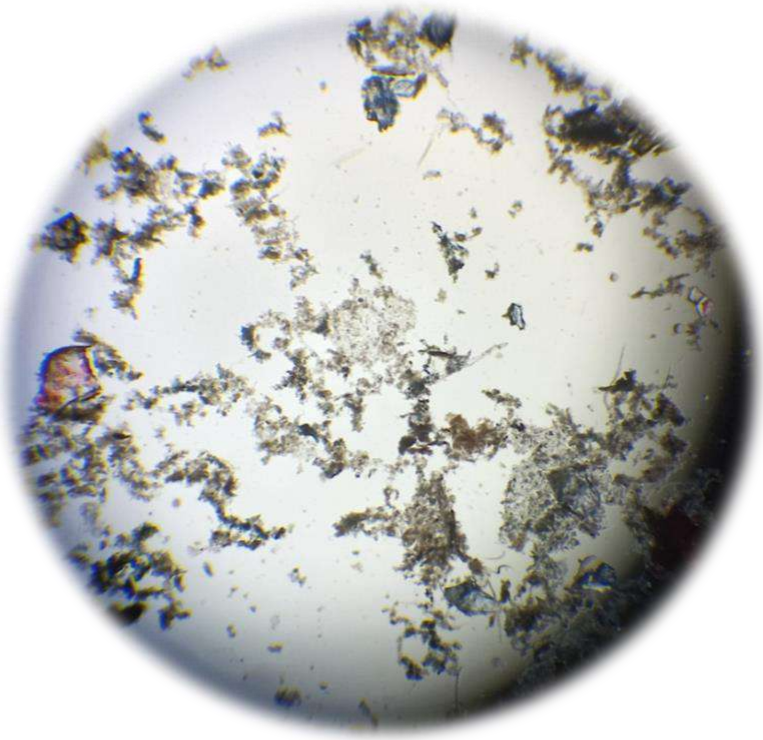


Figure 4.4: Microscopic view for aerobic granular sludge taken with 1 X magnification

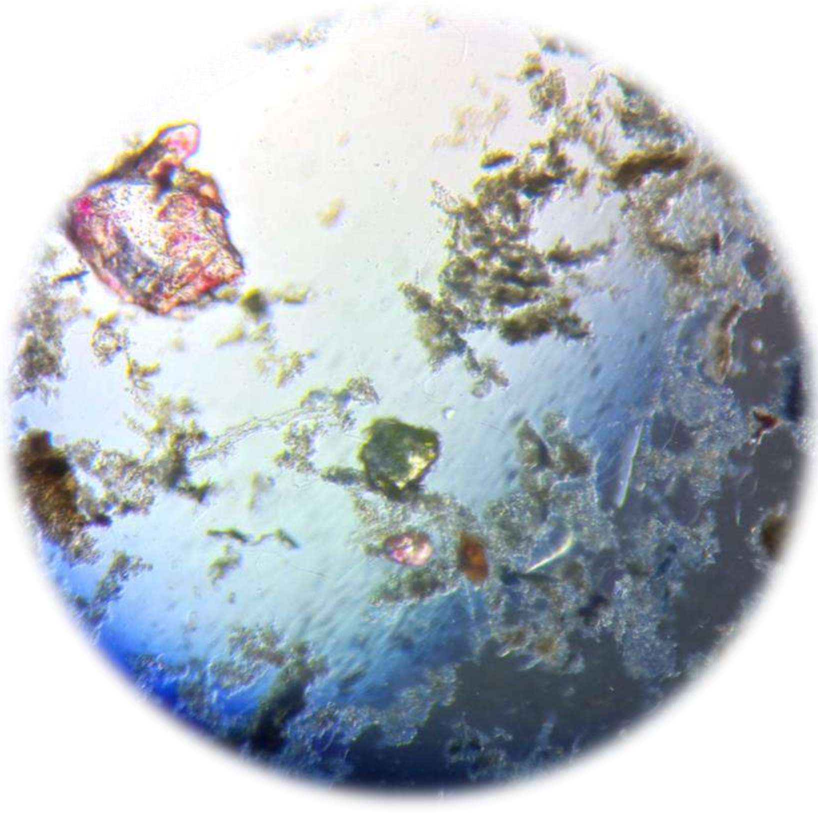


Figure 4.5: Microscopic view for aerobic granules taken with 10 X magnification

The above pictures demonstrated the comparison between the activated seed sludge and aerobic granular sludge formed in the reactor. At the inception of this study the seed sludge was in suspended floc form (Figure 4.3) and later on completing required cycles of SBR batch operation, aerobic granular sludge was appeared in reactor (Figure 4.4 and 4.5).

The reactor seed sludge was originally flocculant; however, it quickly changed to granular sludge while maintaining the necessary hydrodynamic pressure for granule formation. Gradually lowering of the settling time and increasing hydraulic shear stress were the parameters chosen for pressure application to stimulate the granulation process.

The hydrodynamic shear force was applied as superficial air velocity and it was increased from 2.4 cm/s to 3.6 cm/s during granulation stage of operation; also, after the idle period resumed, it was maintained as high as 3.6 cm/s at the time of reactivation stage of SBR operation. According to previous studies, filamentous bacteria can reproduce excessively in conditions of low hydraulic shear stress and AGS can only form when the apparent gas velocity is higher than 1.2 cm/s (Tay et al., 2001a). Additionally, increased hydraulic shear stress can cause cells to secrete more EPS, which in turn encourages the growth of sludge particles with a dense structure and an intact look (Tay et al., 2001a). The hydrophobicity of the sludge surface gradually increases under the influence of hydraulic shear stress, further enhancing the structural stability of AGS (Marshall and Gruickshank, 1973; Pringle and Fletcher, 1983; Zhu et al., 2012).

The granules size in this study was limited to 1 mm ~ 3 mm due to high shear stress was maintained for enhancing granulation. The growth of larger granules, which is possible in an anaerobic system, may not be possible due to the intense shearing force generated by mixing and aeration during the reaction phase (van Benthum et al., 1996; Kwok et al., 1998).

The study by Devlin et al. found that wastewater with low concentrations of COD (300 mg/L) formed stable granular sludge even at a low superficial gas velocity of 0.41 m/s, but when the COD concentration increased to 600 mg/L and above, the same hydraulic shear stress was unable to form AGS (Devlin et al., 2017). This suggests that the effect of hydraulic shear stress on sludge granulation and particle structural stability also depends on the substrate concentration. In this study, the substrate concentration in feed wastewater was greater than 700 mg/L as COD and the ranges of COD in SBR inlet were 797 mg/L ~1233 mg/L. Hence, this parameter of operation also augmented the effect of hydraulic shear stress for developing AGS in the reactor.

The settling time was decreased stepwise from cycles of granulation period to cycles of reactivation phase of operation. During these stages of operation, the settling time was gradually lowered from 5 minutes to 3 minutes at granulation operation phase and maintained 2 minutes at reactivation phase. The volume exchange ratio (VER) was also applied as driving force for acclimatization of AGS in the reactor. During granulation operation VER was 25% and at the time of reactivation experiment it was slowly increased from 44 % to 50%. Therefore, the wash out of poorly settling fluffy flocs was a result of the short settling time and high exchange ratio used in this study, which led to the selection of the faster-settling, denser biomass. The denser particles remained in the reactor eventually transformed into aerobic granules. The production of aerobic granules in SBR has been extensively reported to be influenced by settling time (Adav SS et al., 2008; Zhang Q et al., 2016), which supports the findings of the current study.

4.3.3 Analysis of RTWW Parameters During An-Ae SBR Operation Stage

RTWW from SBR outlet was analyzed in BUET environmental laboratory to evaluate the efficiency of aerobic granular sludge (AGS). The below report was got from one cycle of RTWW sample analysis:

i. COD removal 55.41 %
ii. TSS removal 80.41 %
iii. TDS no removal
iv. Color removal 28.02 %

The analysis report during An-Ae SBR operation phase showed less removal percentage for COD and color than in granulation stage. Therefore, few operation conditions were changed in further An-Ae SBR operation cycles (section 3.8) after conducting the reactivation operation from idle phase for getting better performance of AGS.

In this period, the reaction phases of operation were changed from granulation stage. Here, longer anaerobic reaction was conducted to achieve better performance on color removal. However, the removal of WW parameters from sample of this phase was not up to the mark. Few cycles conducted for this phase was not also good enough to evaluate the AGS performance for RTWW treatment, so more operations on this phase were done later to check the effectiveness of AGS for removing COD, TSS and color from RTWW.

4.4 Performance Analysis of AGS During Reactivation Phase

When idle phase was at end after sixty days of preservation at low temperature in anaerobic condition, reactivation phase started for further experiment of treatment capability on RTWW. The removal percentage of treated RTWW parameters during reactivation phase is shown in Table 4.7.

Table 4.7: RTWW pollutant removal percentage at SBR outlet during reactivation experiment

Parameters / Sampling Days	COD % Removal	TSS % Removal	Color % Removal	TDS % Removal	Cycle Number for Sampling
1 (6th Day)	53.40	no removal	28.17	4.39	Cycle-6
2 (9th Day)	51.17	no removal	38.39	2.08	Cycle-9
3 (12th Day)	52.07	99.50	60.38	3.23	Cycle-12
4 (16th Day)	63.24	98.78	68.53	2.66	Cycle-16
5 (19th Day)	57.04	93.46	72.07	no removal	Cycle-19
6 (21st Day)	61.80	99.97	75.52	no removal	Cycle-21

At this stage of experiment COD removal percentage was between 51 % ~ 63 %, TSS removal percentage was between 0 % ~ 99% and color removal percentage was between 28% ~75%. No significant removal percentage was found for TDS of RTWW treated by AGS.

The graphical picture (Figure 4.6) shows that a consistency is present for COD removal by AGS after resuming from idle period. TSS removal efficiency shows an ascending trend from zero to almost hundred percent day by day at the time of reactivation experiment. A progressive outcome was obtained in color removal by AGS during this stage of operation as well, where the efficiency increased from 28 % to maximum 75 % (Table 4.7).

The similar selection pressures were applied in the SBR after restoring the AGS from idle phase. The effect of hydraulic stress, hydrophobicity, shorter settling time and high substrate concentration were manifested on the AGS after15th cycle of this reactivation operation.

Therefore, from 16th cycle the steady pattern was found on removal of COD, TSS and color from RTWW by the reactivated AGS.

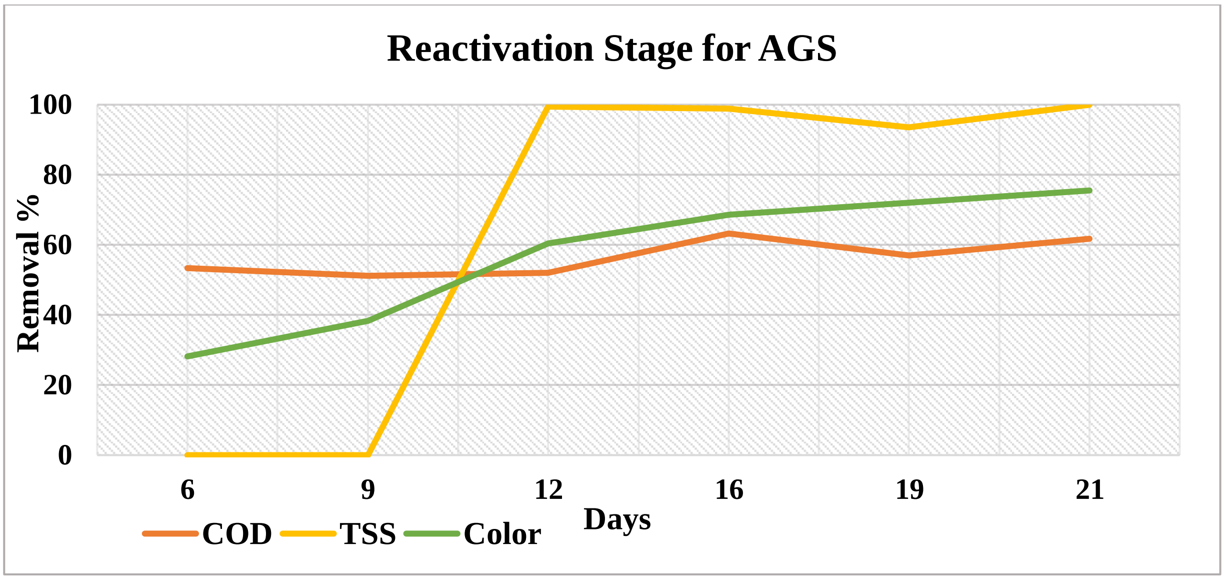


Figure 4.6: Graphical representation on RTWW parameters analysis at outlet of SBR for reactivation experimental period

The analysis report of treated RTWW during reactivation phase is shown in Table 4.8.

Table 4.8: List of RTWW parameters at the outlet of AGS reactor during reactivation stage

Parameters	COD In (mg/L)	COD Out (mg/L)	TSS In (mg/L)	TSS Out (mg/L)	Color In (Pt-Co)	Color Out (Pt-Co)	Cycle Number for Sampling
1 (sampling day-1)	897	418	72.7	~ 72	3230	2320	Cycle-6
2 (sampling day-2)	897	438	72.7	~ 72	3230	1990	Cycle-9
3 (sampling day-3)	797	382	106.6	0.5	3130	1240	Cycle-12
4 (sampling day-4)	797	293	106.6	1.3	3130	985	Cycle-16
5 (sampling day-5)	987	424	122.3	8	2900	810	Cycle-19
6 (sampling day-6)	987	377	122.3	0.04	2900	710	Cycle-21

Hence, the gradual improvement of TSS, Color and COD parameters (Table 4.7 and 4.8) throughout the time of reactivation experiment indicates that AGS kept in idle phase has recovered its ability to treat the RTWW in SBR. Afterwards, the An-Ae SBR operation for treatment of RTWW was conducted to evaluate the final performance analysis of AGS acclimatized in this study.

4.5 AGS Performance Evaluation for Final An-Ae SBR Treatment Test on RTWW

AGS ability to treat tough pollutants including Color, COD and TSS from raw textile wastewater has been investigated at the final stage of this study after resuming the AGS activity from idle condition. The operating conditions also has been modified (section 3.8) in this phase to get better performance of AGS than found in anaerobic-aerobic SBR operation before idle period. The treatment efficiency of AGS during final An-Ae SBR operation stage is listed in Table 4.9.

Table 4.9: RTWW pollutants removal efficiency by AGS during An-Ae SBR final operation

Parameters / Sampling Days	COD % Removal	TSS % Removal	Color % Removal	TDS % Removal	Cycle Number for Sampling
1 (1st Day)	70.92	98.92	75.00	0	Cycle-1
2 (2nd Day)	69.30	99.67	76.72	0.15	Cycle-2
3 (4th Day)	72.14	96.59	68.45	2.26	Cycle-4
4 (5th Day)	67.78	66.80	71.21	3.06	Cycle-5

After completing twenty-one cycles of reactivation in aerobic SBR, the expected results were found in the anaerobic aerobic SBR experiment for treatment of RTWW. The COD removal efficiency is 67 % to maximum 72 % and color removal data is 68% to maximum 77% (Table 4.9). TSS removal results are in quite high range from 67% to maximum 100%, this is due to the good settling property of the aerobic granular sludge (AGS) acclimatized in the reactor. TDS removal has no significant impact by this AGS technology. Actually, no biological technology can effectively remove TDS from wastewater, rather further advance technology like reverse osmosis or evaporation is always require following biological treatment scheme (Kannan et al., 2006).

Residual amount of WW parameters was measured in treated effluent where residual COD was 275 ~ 318 mg/L, with an average value of 296.5 ± 21.5 mg/L; residual TSS was 40 mg/L to less than 5 mg/L and residual color was 675 ~ 915 Pt-Co at 455 nm wavelength, with an average of 795 ± 120 Pt-Co at 455 nm wavelength (Table 4.10). The below graph (Figure 4.7) is showing the trend of COD, TSS and color removal from RTWW in the final anaerobic-aerobic SBR operation. The COD and color in raw textile dyeing wastewater are always considered as harsh pollutants, especially for color removal conventional biological system is found less effective in terms of process feasibility. In fact, activated sludge process is sensitive to several compounds (like most commercial dyes) which are toxic to microorganisms; this results in problems of sludge bulking, rising sludge and pin point floc formation, thereby greatly affecting treatment performance ((Delee et al., 1998; Ahn et al., 1999). As proved by the low TSS in the effluent, no such problems were observed in SBR of this experiment. The below Table 4.10 shows the RTWW characteristics after treated by AGS in final operation of An-Ae SBR.

Table 4.10: List of RTWW parameters at the outlet of AGS reactor during final An-Ae SBR operation

Parameters	COD In (mg/L)	COD Out (mg/L)	TSS In (mg/L)	TSS Out (mg/L)	Color In (Pt-Co)	Color Out (Pt-Co)	Cycle Number for Sampling
1 (sampling day-1)	987	287	122.3	1.5	2900	725	Cycle-1
2 (sampling day-2)	987	303	122.3	0.4	2900	675	Cycle-2
3 (sampling day-3)	987	275	122.3	4.1	2900	915	Cycle-4
4 (sampling day-4)	987	318	122.3	40.6	2900	835	Cycle-5

In this study, AGS technology shows consistence performance than other studies done earlier for Color and COD removal from real textile wastewater. The below Table 4.11 depicts the AGS performance data on COD and color removal done in previous studies for both RTWW and STWW.

Table 4.11: List of AGS efficiency for COD and Color removal (%), done in different studies

Sl No.	Reactor Type	WW Type	Color removal, %	COD removal, %	Reference Study
1	An-Ae SBR	RTWW	68 ~ 77	68 ~ 72	Author of this Study
2	An-Ae SBR	STWW	56	93	Muda et al., 2012
3	An-Ae SBR	STWW	30 ~ 55	85	Moghaddam and Moghaddam, 2016
4	An-Ae SBR	RTWW (sterilized)	61	46	Kee et al., 2014
5	An-Ae SBR	RTWW+ municipal WW	73	68	Manavi et al., 2017
6	SBBGR	RTWW+ municipal WW	33.9 % ~ 52.6 %	82.1 ± 3.6	Lotito et al., 2014
7	SBBGR	RTWW	0 ~ 60	55 ~78	Lotito et al., 2012

In previous studies of AGS, the removal percentage for color and COD was found in good range when synthetic textile wastewater was used. However, those studies found that use of raw textile wastewater resulted in poor removal efficiency for color and COD (Table 4.11). Therefore, in this study raw textile wastewater was applied for both AGS acclimatization stage and treatment stage where no specialized seed or mixture was used with RTWW to expedite the performance of AGS unlike the previous studies (Table 4.11). In this study, the removal of color and COD from RTWW was found quite higher ranges than the earlier researches.

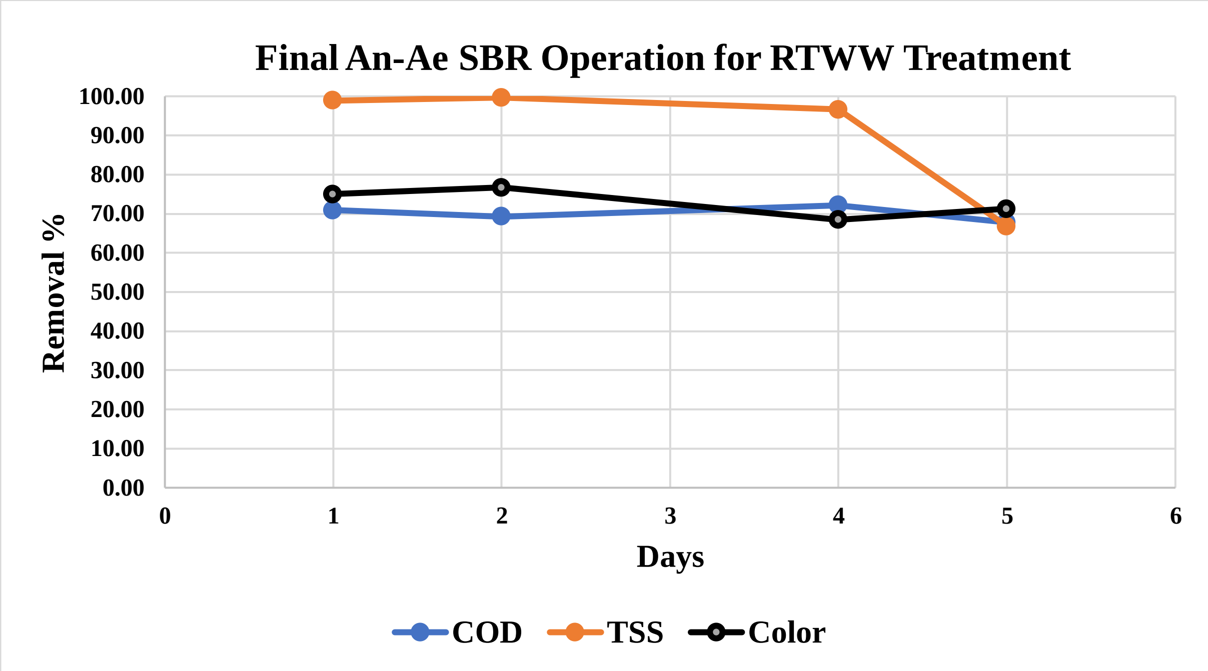


Figure 4.7: Graphical representation on RTWW pollutants removal in final experiment of An-Ae SBR.

In the literatures, the degradation and decolorization of dye during anaerobic conditions has been widely reported (Santos et al., 2007). In this study, the developed aerobic granular sludge in SBR has been proven capable of performing the degradation process during anaerobic and aerobic phases, based on the removal performance of the system. This shows that the microbial granular sludge contains aerobic, facultative, and anaerobic bacteria (Muda et al., 2010). Li and Liu (2005) claimed that the diffusion of oxygen into the interior of the granules became restricted after they reached a size larger than 0.5 mm. Given that the average size of the microbial granular sludge formed in this study was 2 ± 1 mm, this may indicate the presence of anaerobic microbes in the center part of the sludge. Thus, the inner layer of anaerobic zone stimulated the biodegradation of dyes with the help of anaerobic microbes present in the AGS. The outer layer of the granules contains aerobic microorganisms, which are primarily responsible for COD removal as they can readily access oxygen molecules.

Aerobic granular sludge systems compared to activated sludge systems, are characterized by a low biomass sludge production rate. The low sludge production rate is the result of the relatively high sludge retention achieved by aerobic granular sludge (Liu and Tay, 2007b). During the experiment of this study, MLSS of the granular sludge was found to achieve maximum 10648 mg/L which was initially 4600 mg/L in the seed flocculant sludge. This indicated high biomass retention in the AGS system. It is also found in the previous literatures that biomass concentration in the AGS system reactor is typically in the range of several grams per liter i.e. 10 to 20 g/ l, or higher (Liu and Liu, 2006). The high sludge retention time achieved due to high biomass concentration could have promoted the development of species able to degrade dyes. As known in previous research, long sludge ages allow additional time for bacteria to acclimate to potentially toxic and/or inhibitory compounds as well as to adjust to any carbon compound that is difficult to degrade (Fongsatitkul et al., 2008).

In addition to the degradation mechanism, adsorption onto the biomass is another possible method for dye removal (Aksu, 2001; Crini, 2006). The high surface area, porosity, and strong settling capabilities of AGS make it an appealing material for the biosorption-based removal of colors and metals from wastewater (Adav et al., 2008). This phenomenon also depends on the chemistry of the specific dye and the microbial biomass (Robinson et al., 2001). For example, activated sludge removes medium to high portions of soluble basic and direct dyes, primarily by adsorption, whereas acid and reactive dyes are poorly adsorbed (Delee et al., 1998; Rozzi et al., 1998; Lotito et al., 2012). In the earlier studies it was also found that viable cells had a higher adsorption capacity than dead cells (Ledakowicz et al., 2001). In this study the AGS was acclimatized using knit dyeing wastewater, where different types of dyes are used like reactive dyes, disperse dyes, acid dyes and sulfur dyes etc. These types of dyes in the RTWW exhibited the possible potential for adsorption of dye components on the biomass of AGS in this study.

However, the removal of TSS was decreased after the 4^{th} Cycle of decolorization test. This kind of behavior may be occurred due to disintegration of AGS in the system which gradually results in poor settleability of particles. At this stage the biomass concentration was measured and it was found quite lower as 9400 mg/L than the previous data 10648 mg/L taken before starting the final An-Ae SBR operation. This indicates that the shear force provided by air mixing gradually diminishes the mechanical strength of the AGS rather than aggregation. This kind of phenomenon was also described in the earlier studies where hydraulic shear stress increased particle abrasiveness with time of SBR operation, resulting in the destruction of particle mechanical strength (Zhou et al., 2016).

In this study, final TSS at the outlet of AGS reactor (Table 4.10) complied with discharge standard of DoE. Though the removal efficiency of AGS for color and COD was found in satisfactorily range (Table 4.9), the final value of color and COD at the outlet (Table 4.10) of AGS reactor ultimately did not meet the local standard for effluent discharge (ECR, 23). In case of meeting COD and color limit of discharge, additional pretreatment should be introduced before starting the SBR operation with RTWW. Chemical coagulant and flocculant with primary settler (Tripathy and De, 2006) can be used to reduce 30~35% organic pollutant initially before feeding the RTWW in the AGS reactor, since AGS reactor of this study is capable of removing more than 70% organic pollutant. Decolorant chemical dosing also can be done in the pretreatment unit to reduce some portion of color earlier of the AGS reactor operation unit, so that final WW from AGS reactor can meet the local standard of color.

4.6 Aerobic SBR for Granulation vs An-Ae SBR for RTWW Treatment

In this study, the operation conditions during granulation are not same as the operation conditions during RTWW treatment experiment in An-Ae SBR. Granulation was done in aerobic SBR to favor the environmental conditions for developing AGS in the reactor (section 4.3.2 discussion) where SBR cycle time for reaction was required not more than 5 to 6 hours. In previous studies, SBR cycle time for reaction was also kept within 6 hours to develop AGS from seed sludge (Kee et al., 2014; Manavi et al., 2017; Kolekar et al., 2012; Sarvajith et al., 2018). This study found that the color removal percentage was not stable for higher removal efficiency in aerobic SBR operational conditions. The below Table 4.12 shows a comparison of color removal efficiency in aerobic SBR and An-Ae SBR.

Table 4.12: Comparison of color removal % in aerobic SBR and An-Ae SBR

Sl No.	Aerobic SBR Operation (During granulation phase) (Color removal %)	An-Ae SBR Final Operation (During treatment test) (Color removal %)	Sampling days
1	79.14	75	Sample-1
2	51.96	76.72	Sample-2
3	50.12	68.45	Sample-3
4	32.08	71.21	Sample-4

In Table 4.12 it is shown that with days of aerobic SBR operation, color removal was not consistent for more than 70% removal. Though, aerobic SBR found good color removal at starting days of operation, but later the color removal efficiency became poor as the operations continued in aerobic SBR. On the other hand, An-Ae SBR showed a consistent removal of around 70% color removal during the operational days of An-Ae SBR experiment.

Therefore, to get consistent higher removal efficiency for color, the treatment experiment was done in An-Ae SBR instead of in aerobic SBR. As discussed earlier in section 4.3.1, higher range of color removal needs longer reaction time in anaerobic conditions (Muda et al., 2010). In this study, the cycle time of An-Ae SBR was maintained for 24 hours inspired from earlier studies. Previous researchers also followed 24 hours of cycle time for the experiment of color removal from textile wastewater (Kee et al., 2014; Manavi et al., 2017; Sarvajith et al., 2018).

The reactivation stage of this study also followed the aerobic SBR conditions where it was found that after 16^{th} cycle of SBR operation color removal was above 70%. However, during reactivation experiment the consistency of this removal percentage for color in the aerobic SBR conditions was not tested in this study. As the intention of reactivation stage was to activate the AGS after idle period for testing AGS performance to treat RTWW in An-Ae SBR. Therefore, future studies can work on this subject to check the AGS ability after idle period for consistent color removal percentage in aerobic SBR.

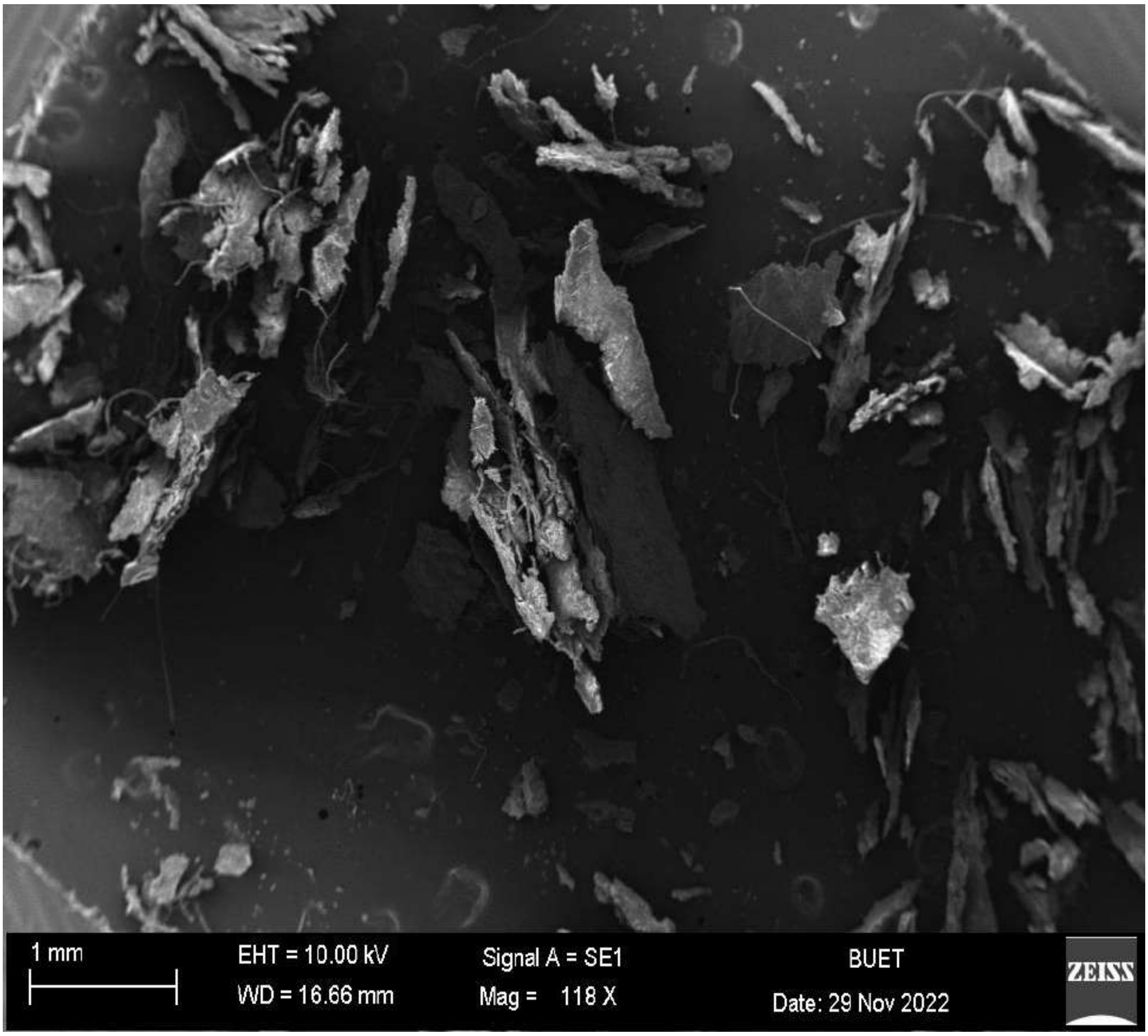
4.7 Physio-Chemical Characterization for AGS of this Study

4.7.1 SEM Analysis of AGS

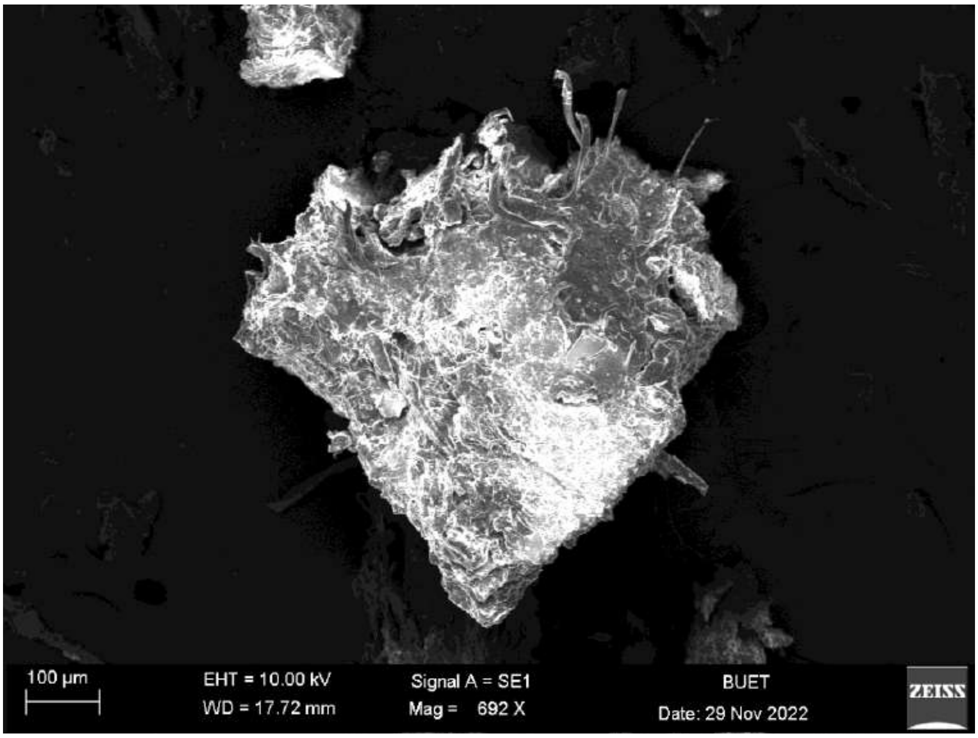
The AGS was tested for Scanning Electron Microscopy (SEM) analysis at the end of this experiment to understand the physical properties of it. In total fifty-eight cycles were run including granulation, An-Ae SBR, reactivation and final An-Ae SBR throughout this research. After fifty-eight (58) cycles the collected AGS was taken for SEM test.

The cultivated aerobic granular sludge (AGS) was collected from the reactor after settling of 10 minutes. The AGS sample was taken to beaker for dewatering and it was kept in room temperature for 25 days to dry this sludge. Before putting the sample in SEM equipment, freeze drying of AGS was also done for 24 hours. The SEM analysis test was done at the laboratory of Chemical Engineering Department of BUET.

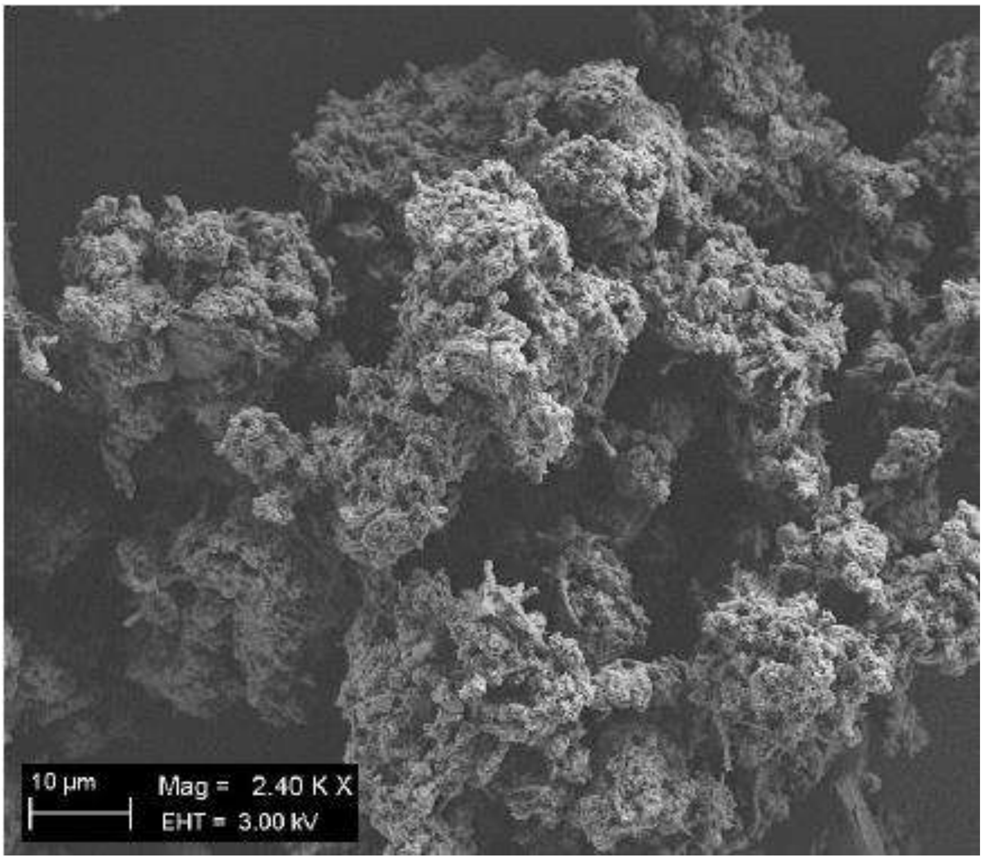
The SEM test pictures clearly shows the nature of granular structure in the taken sample. The physical structure of sludge is compact and has a regular shape almost in circular form. The appearance of the AGS is not matched with the fluffy and irregular shape of conventional activated sludge. Few pictures during this test have been attached as following and a SEM micrograph of activated sludge done by Neis et al., 2012 has been shown also to differentiate the morphology of AGS and activated sludge from conventional process of treatment.



(a)



(b)



(c)

Figure 4.8: (a) , (b) The surface of cultivated AGS of this study in different dimension of SEM analysis (Author) and (c) SEM picture of activated sludge (Neis et al., 2012).

4.7.2 EDX Analysis on AGS

Energy dispersive X ray (EDX or EDS) analysis is a widely employed technique by today's materials scientists and used together with a scanning electron microscope (SEM), an edx detector can generate more information about a sample than an SEM can alone. Using EDX, researchers can quickly generate information about the chemical composition of a sample, including what elements are present as well as their distribution and concentration.

With an SEM, a variety of signals offer up different information about a given sample. For example, backscattered electrons produce images with contrast that carry information about the differences in the atomic number, while secondary electrons produce topographic information about the sample. Yet when SEM is joined with an EDX detector, X-rays can also be used as a signal to produce chemical information.

Interestingly, EDX can be used for both qualitative and quantitative analysis, enabling users to identify both the type of elements that are present as well as the percentage of each element's concentration within the sample. And as with traditional SEM, the technique requires little to no sample preparation and is non-destructive, meaning that it doesn't damage the sample.

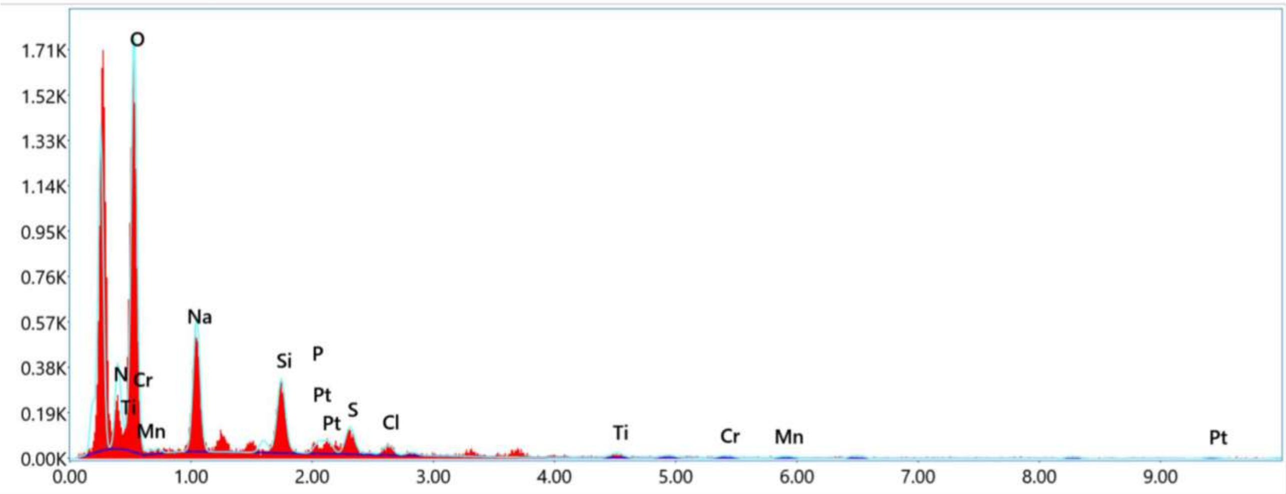


Figure 4.9: Spectrum of chemical composition analysis for AGS at BUET Chemical Engineering Lab using EDX technology with SEM

EDX analysis with SEM was done at the time for AGS of this experiment to study the chemical composition of this AGS. This was done at BUET Chemical Engineering Department Laboratory and the results of this test were shown in Figure 4.9 and Table 4.13. It is found from the EDX results that the AGS is mostly composed with oxygen molecules, in addition few heavy metals like chromium, titanium, platinum are also present in the sludge. AGS absorbed these heavy metals from different effluents fed in the reactor for many cycles of SBR operation.

4.7.3 FTIR Analysis on AGS

Fourier transform infrared spectroscopy (FTIR) was also performed at BUET Chemical Engineering Department Laboratory for evaluation of composition of the AGS.

FTIR analysis (Fourier Transform Infrared Spectroscopy) is a powerful analytical technique that is used to identify and quantify the presence of various chemical bonds in a given sample. The identification of organic, polymeric, and occasionally inorganic materials is accomplished using this infrared spectroscopy technique. The FTIR test relies on infrared light to scan materials and observe bond characteristics. When there are unknowns, FTIR analysis services can identify the compounds and the general sort of material being tested.

Table 4.13: The EDX analysis results for AGS in weight % and atomic % done with SEM at BUET

Smart Quant Results

Element	Weight %	Atomic %	Net Int.	Error %	R	A	F
N K	12.15	16.07	45.88	12.17	0.9128	0.2383	1.0000
O K	53.62	62.06	271.73	9.81	0.9178	0.2587	1.0000
NaK	17.04	13.72	102.17	9.43	0.9307	0.3908	1.0018
SiK	6.30	4.16	67.26	7.53	0.9418	0.6934	1.0046
P K	0.00	0.00	0.02	99.99	0.9451	0.7466	1.0068
S K	3.04	1.76	26.26	12.18	0.9483	0.8051	1.0078
ClK	1.40	0.73	10.65	19.29	0.9513	0.8425	1.0102
TiK	1.40	0.54	6.10	19.68	0.9654	0.9631	1.0412
CrK	1.05	0.37	3.31	36.89	0.9708	0.9774	1.0587
MnK	0.85	0.29	2.24	29.86	0.9734	0.9829	1.0708
PtM	3.15	0.30	11.07	13.63	0.9469	0.7790	1.0124

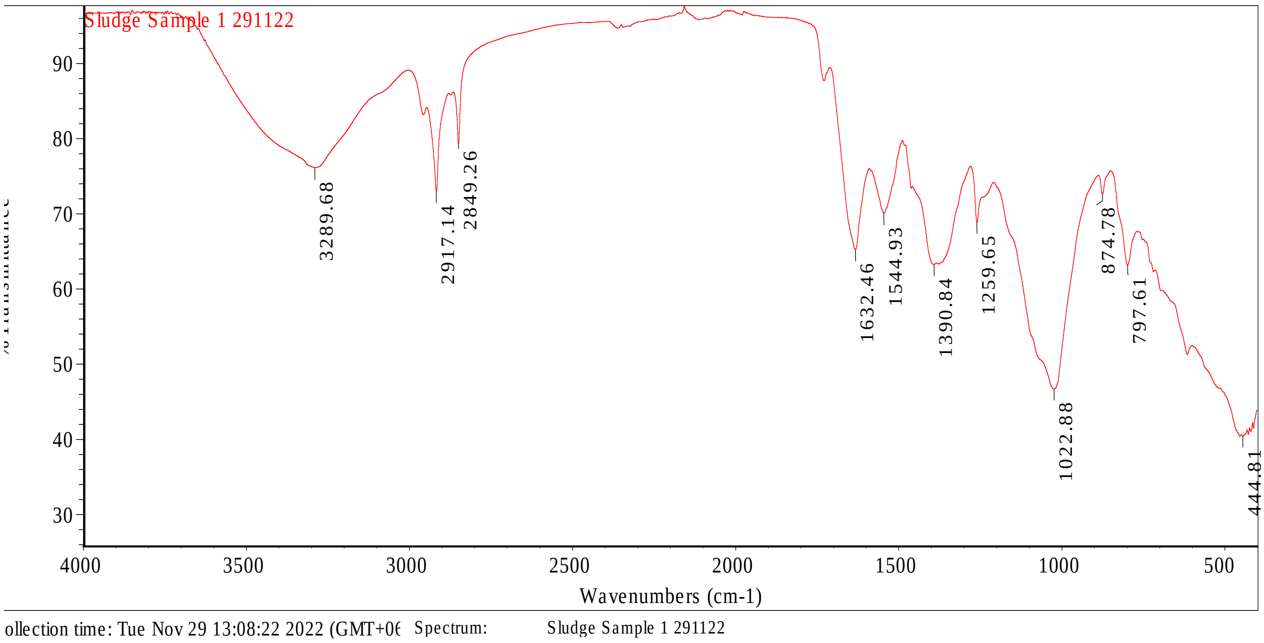


Figure 4.10: FTIR of AGS sludge taken the sample at the end of this study

The FTIR analysis was conducted between 400 cm^{-1} to 4000 cm^{-1} wavelengths for the collected AGS from this experimental reactor (Figure 4.10). Sharp and less intensity (more than 60% transmittance) peaks are obtained in the wavelengths of 2917 cm^{-1}, 2849 cm^{-1}, 1260 cm^{-1}, 875 cm^{-1} and 798 cm^{-1}. A broad band is found at 3290 cm^{-1} wavelength which is less intense and high intense little wide band is seen at 1023 cm^{-1}.

The highest peak at 1023 cm^{-1} depicts the presence of C-O stretching (Tibebe et al., 2022) of organic compounds which could be originated from dyes, surfactants, cellulose fibers, or other organic contaminants commonly found in textile effluents.

The wide and small peak at 3290 cm^{-1} is associated with compounds containing the stretching vibration of hydrogen-bonded hydroxyl groups O-H from carboxyl's, phenols or alcohols (Sun Y et al., 2014). Some textile dyes and finishing agents may contain hydroxyl groups that contribute to this peak.

Aliphatic compounds, such as alkanes and alkenes, typically display peaks in the region of 2800-3000 cm^{-1}. The small and sharp peaks in wavelength of 2917 cm^{-1} and 2849 cm^{-1} indicates the stretching vibrations of carbon-hydrogen (C-H) bonds in the aliphatic chains (Kaçan and Kütahyalı, 2012). The organic compounds in textile effluents and microorganisms are responsible to present these ranges band of peaks which indicates the existence of such compounds in the AGS.

The short and small peak at 1545 cm^{-1} indicates the NH groups resulted due to the vibration movements (Kac̦an and Kütahyalı, 2012). This could be the originated from both dye compounds of textile and tannery effluent as at the last stage of experiment tannery effluent was also tested on AGS. Since, NH groups can be found in textile effluent as well as in tannery effluent due to various organic pollutants, dyeing components and tanning materials.

A medium sized short peak at 1391 cm^{-1} is illustrated the presence of the phenolic OH groups (Kac̦an and Kütahyalı, 2012) from different dye component in textile industries. If the textile effluent contains protein-based materials such as wool or silk, the amide bands may be observed in the FTIR spectrum. Amide I band (~1650 cm^{-1}) corresponds to the C=O stretching vibrations and such kind of band is also found in Figure 4.11 at the wavelength of 1633 cm^{-1}.

According to the study of (Kalaivani and Srinivasan, 2019) for FTIR assessment on tannery sludge, they got a very sharp peak at 873 cm^{-1} which was assigned to Al-O stretching vibration, the band centered around 750 cm^{-1} shows the alkaline (Na, K) content in sample. Therefore, in this study of AGS FTIR, a very small less intense peak was observed at the wavelength of 875 cm^{-1} which represents the existence of Al-O stretching vibration in the AGS sample.

The above discussion on FTIR spectra analysis concludes that the AGS produced in this experimental study was able to reduce the pollutants from textile effluent where organic pollutants, suspended particles, dye residues and other unwanted matters were biologically degraded, adsorbed and diffused by AGS.

Chapter 5
CONCLUSION

5.1 Introduction

The study was conducted on textile dyeing effluent to introduce a cost effective, environment and operation friendly technology for treating and removing tough pollutants including recalcitrant color materials from real textile wastewater. This work manifested that AGS could be a potential solution for applying in textile sectors of Bangladesh in lieu of current physio-chemical and conventional biological technology.

Most of the studies on textile effluent for studying AGS was done on synthetic wastewater and only few works were found on real textile effluent using customized microorganisms, nutrients or seed sludge. However, this study was intended to explore the possibility of growing aerobic granular sludge (AGS) in a SBR where real textile effluent was applied with activated sludge as seed without any additional nutrients or customized microbial community.

In aspect of current technology for treating textile wastewater, this study specially focused on color removal from real textile wastewater. Presently most of the industries treat color by using chemical decolorant dosing which is costly and not sustainable for environment due to chemical residue sludge production. In addition, recent regulation of government published on ECR'23 imposes more stringent controlling on color effluent discharge from textile industries. This work demonstrated the AGS efficiency of color removal up to maximum 76%, therefore this accomplishment of AGS technology may be executed in future for textile dyeing effluent to remove color in a sustainable manner. In that case AGS technology for textile dyeing wastewater might need a post treatment of color removal with chemical dosing to meet the local regulations of discharge. This will enhance the sustainable technology for treatment of textile effluent, by reducing operating cost due to less chemical consumption for removing color from wastewater than the conventional system.

5.2 Conclusion of this Study

The short summary of the experimental findings is as following:

a) This study was successful to develop aerobic granular sludge (AGS) in SBR using real textile wastewater from a knit dyeing plant. In this study after 52 number cycles of operation (i.e., 52 days SBR operation, one cycle in a day) including granulation, reactivation and An-Ae SBR testing phase, aerobic granules were found within the size of 1 mm to 3 mm checked by visual inspection.

b) The morphology of AGS was obtained from the SEM analysis after total fifty eight cycles of SBR operation. The SEM image shows that AGS has dense, compact and regular shape which is different from loosely attached flocs and irregular shape of conventional activated sludge.

c) Good settling properties of the AGS were observed in all phases of operation in SBR-granulation, idle and reactivation and An-Ae SBR testing phase. The suspended solid (TSS) removal of more than 90% within short settling time (2 minutes ~ 3 minutes) showed the desired settling capability of AGS produced from the seed suspended sludge in the SBR.

d) At the time of granulation experimental phase, AGS performance was found at the desired level in terms of removing the organic pollutants (COD), suspended particles and color components. The AGS efficiency for reducing COD, TSS and color were found maximum 75%, 99% and 79% respectively.

e) AGS ability to treat raw/real textile waste water of a dyeing plant was also evaluated after resuming the SBR operation from 60 days inactivity of AGS in anaerobic environment. During the decolorization test after reactivation of AGS from idle phase, the treatment efficiency of AGS for COD removal was up to 72.1%, TSS removal was up to 99.7% and color removal was up to 76.7%. The results of COD, TSS and color represents that AGS was stable and strong enough to treat raw textile waste water even after keeping in idle condition without any nutrient for two months.

f) FTIR analysis explained the presence of chemical speciation of different synthetic dyes in the AGS. It concludes that the colour component from RTWW was adsorbed and degraded by AGS.

5.3 Recommendations

i. Though good removal efficiency was found by AGS in this experiment to remove color, COD and TSS, the standard guideline for discharging TWW was still under range in terms of color and COD. The results of this study only meet ECR’23 guideline for the parameter of TSS. Therefore, more studies are needed on operational conditions to get the optimum efficiency of AGS for removing COD and color in accordance with meeting local discharge guideline. However, current AGS technology of this experiment can also be applied in textile wastewater with addition of few pretreatment units like coagulation-flocculation chemical dosing including primary settler for complying discharge standard of DoE.

ii. In this experimental work microbial community analysis and AGS long term stability was not studied which is important factor to commercialize this technology in industrial market. Therefore, it is suggested to do more work on AGS to treat real textile wastewater for identifying the optimum morphology and reaction ability of AGS.

iii. Dedicated research work is necessary on operational conditions of SBR for AGS acclimatization. This segment of work is required for improving the AGS efficiency for removing organic matter, toxic materials, heavy metals, suspended particles and recalcitrant color from real textile wastewater. Since the operational conditions like aeration, hydraulic retention time (HRT), organic loading rate (OLR), pH, alkalinity and temperature have great impact on treatment capability of AGS for wastewater.

Therefore, it is important to consider these factors for future works to ensure optimal performance of aerobic granular sludge processes on treatment of real textile wastewater from a dyeing plant.

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