Universitat Politècnica de Catalunya

Doctoral Thesis

Ph.D. Program in Environmental Engineering

Study of a hybrid system: Moving Bed Biofilm Reactor - Membrane Bioreactor (MBBR-MBR) in the treatment and reuse of textile industrial effluents

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ABSTRACT

As one of the oldest and most complex sectors within the manufacturing industries, textile industry consumes large quantities of water and produces large volumes of wastewater during its production. Textile wastewater often shows high color concentration, containing a large range of organic chemicals, with high chemical oxygen demand as well as hard-degradation materials. Therefore, understanding and developing effective textile industrial wastewater treatment technologies is environmentally important. Different treatments such as biological or physico-chemical processes have been studied to treat textile wastewater.

Membrane Bioreactor (MBR) technology has been widely applied in textile wastewater treatment and Moving Bed Biofilm Reactor (MBBR) is a relatively novel technology to treat this type of wastewater. Both of them have some deficiencies and limitations during application. Thus, A MBBR-MBR hybrid system could be an attractive solution to the shortcomings of each treatment process.

In this thesis, a hybrid MBBR-MBR system has been designed and applied for the treatment of textile wastewater. Additionally, the feasibility of reusing the treated water in new dyeing processes has been studied.

The first step of the thesis work has been the comparative study of the treatment of textile wastewater by three treatment processes, conventional activated sludge (CAS), MBR and MBBR, working under the same operating conditions. The performance of each process has been investigated and compared in technical, economic and environmental perspectives. The results showed that technically, MBR was the most efficient technology, of which the chemical oxygen demand (COD), total suspended solids (TSS), and color removal efficiency were 91%, 99.4%, and 80%, respectively, with a hydraulic retention time (HRT) of 1.3 days. MBBR, on the other hand, had a similar COD removal performance compared with CAS (82% vs. 83%) with halved HRT (1 day vs. 2 days) and 73% of TSS removed, while CAS had 66%. Economically, MBBR was a more attractive option for an industrial-scale plant since it saved 68.4% of the Capital Expenditures (CAPEX) and had the same Operational Expenditures (OPEX) as MBR. The MBBR system also had lower environmental impacts compared with CAS and MBR processes, since it reduced the consumption of electricity and decolorizing agent with respect to CAS. According to the results of economic and Life Cycle Assessment (LCA) analyses, the water treated by the MBBR system was reused to make new dyeings and the quality of new dyed fabrics was within the acceptable limits of the textile industry.

Combined with the theory and experimental results, a hybrid MBBR-MBR reactor was designed and applied in textile wastewater treatment. The MBBR-MBR system achieved reducing the HRT to 1 day, which is very promising in textile industry comparing with conventional biological treatment. The removal efficiency of COD reached 93%, which is almost the maximum for a biological process treating this type of wastewater, as well as the color removal performance, which achieved 85%. Additionally, 99% of the TSS were removed due to the filtration. Furthermore, new dyeing processes reusing the treated water were performed. Quality of the new dyed fabrics with treated water were compared with the reference fabrics. Color differences between new dyed fabrics and reference fabrics were found within the general requirement of textile industry ($DE_{CMC(2:1)} < 1$). The reuse of treated water in new dyeing processes is beneficial both for the industry and for the environment since the textile sector is an intensive water consumer in their dyeing and finishing processes.

Additionally, based on the experimental results in the pilot plant, an economic study and LCA analysis were carried out to evaluate the economic and environmental feasibility of the implementation of the hybrid MBBR-MBR on an industrial scale. Economically, MBBR-MBR had lower CAPEX and OPEX than CAS process due to lower effluent discharge tax and the decolorizing agent saved. The result of Net Present Value (NPV) and the Internal Rate of Return (IRR) of 18% suggested that MBBR-MBR is financially applicable for the implantation into industrial scale. The MBBR-MBR system also had lower environmental impacts compared with CAS process in the LCA study, especially in some categories, such as the Climate change, Human Health, Marine eutrophication, and ecotoxicity categories, thanks to the high quality of the effluent treated by MBBR-MBR and the avoiding of using extra decolorizing agent, a compound based on a quaternary amine.

RESUMEN

Como uno de los sectores más antiguos y complejos dentro de las industrias manufactureras, la industria textil consume grandes cantidades de agua y produce grandes volúmenes de aguas residuales durante su producción. Las aguas residuales textiles presentan a menudo una elevada coloración, contienen una amplia gama de productos químicos orgánicos, con una alta demanda química de oxígeno, así como materiales con poco degrabilidad. Por lo tanto, comprender y desarrollar tecnologías de tratamiento de aguas residuales industriales textiles eficaces es muy importante ambientalmente. Se han desarrollado diferentes tratamientos, tanto procesos biológicos como físico-químicos para la depuración de aguas residuales textiles.

La tecnología de Bioreactor de membranas (conocida por su acrónimo en inglés como MBR) se ha aplicado ampliamente en el tratamiento de aguas residuales textiles, mientras el Reactor de biofilm de lecho móvil (conocido por sus iniciales en inglés como MBBR) es una tecnología relativamente nueva para tratar este tipo de aguas residuales. Ambos muestran algunas deficiencias y limitaciones durante su aplicación. Por lo cual, un sistema híbrido MBBR-MBR podría ser una solución atractiva a los inconvenientes de cada uno de estos dos procesos individuales.

En esta tesis se ha diseñado y aplicado un sistema híbrido MBBR-MBR para el tratamiento de aguas residuales textiles. Adicionalmente, se ha estudiado la viabilidad de reutilizar el agua tratada en nuevos procesos de tintura.

El primer paso del trabajo desarrollado en la tesis ha sido el estudio comparativo del tratamiento de aguas residuales textiles mediante tres procesos de tratamiento, fangos activados convencionales (CAS), MBR y MBBR, trabajando en las mismas condiciones de funcionamiento. El rendimiento de cada proceso ha sido investigado y comparado desde una perspectiva técnica, económica y ambiental. Los resultados mostraron que, técnicamente, el MBR era la tecnología más eficiente, con valores de eliminación de demanda química de oxígeno (COD), sólidos en suspendidos totales (TSS) y de color de 91%, 99,4% y 80%, respectivamente, a un tiempo de retención hidráulica (HRT) de 1,3 días. MBBR, por otro lado, tuvo un rendimiento de eliminación de COD similar en comparación con CAS (82% frente a 83%), sin embargo reducía a la mitad el HRT (1 día frente a 2 días) y eliminaba un 73% de TSS , mientras que CAS tenía el 66%. Económicamente, MBBR era una opción más atractiva para una planta a escala industrial, ya que ahorraba 68,4% de los gastos de capital (conocidos por sus iniciales en inglés como CAPEX) y

tenía los mismos gastos operativos (conocidos por sus iniciales en inglés como OPEX) que MBR. El sistema MBBR también tuvo menores impactos ambientales en comparación con los procesos CAS y MBR, ya que redujo el consumo de agente decolorante con respecto a CAS y de electricidad con respecto a MBR. Según los resultados de los análisis económicos y de Análisis de Ciclo de Vida (conocido por sus iniciales en inglés como LCA), el agua tratada por el sistema MBBR se reutilizó para realizar nuevas tinturas comprobando que la calidad de los nuevos tejidos teñidos se encontraba dentro de los límites aceptables de la industria textil.

Combinado con la teoría y los resultados experimentales, se diseñó y operó un reactor híbrido MBBR-MBR para el tratamiento de aguas residuales textiles. El sistema MBBR-MBR logró reducir la HRT a 1 día, lo que es muy prometedor en la industria textil, en comparación con el tratamiento biológico convencional. La eficiencia de remoción de COD alcanzó el 93%, que es cercano al máximo para un proceso biológico de tratamiento de este tipo de aguas residuales. Así como el desempeño de remoción de color, que alcanzó el 85%. Además, se eliminó el 99% de los TSS debido a la filtración. A continuación, se realizaron nuevos procesos de tintura reutilizando el agua tratada. La calidad de los nuevos tejidos teñidos con agua tratada se comparó con los tejidos de referencia. Las diferencias de color entre los tejidos nuevos teñidos y los tejidos de referencia se encontraron dentro del requisito general de la industria textil ($DE_{CMC\ (2:\ 1)}$ <1). La reutilización del agua tratada en nuevos procesos de tintura es beneficiosa tanto para la industria como para el medio ambiente ya que el sector textil es un consumidor intensivo de agua en sus procesos de tintura y acabado.

Adicionalmente, con los resultados experimentales en la planta piloto, se realizó un estudio económico y un análisis de LCA para evaluar la viabilidad económica y ambiental de la implementación del sistema híbrido MBBR-MBR a escala industrial. Económicamente, MBBR-MBR tuvo los gastos de CAPEX y OPEX más bajos que CAS debido a las menores tasas de vertido de aguas residuales industriales y al ahorro de agente decolorante. El resultado del Valor Actual Neto (conocido por sus iniciales en inglés como VPN) y la Tasa Interna de Retorno (conocida por sus iniciales en inglés como TIR) del 18% sugirió que MBBR-MBR es financieramente viable para la implantación a escala industrial. El sistema MBBR-MBR también tuvo menores impactos ambientales en comparación con el proceso CAS en el análisis del ciclo de vida (LCA), especialmente en categorías, como cambio climático, salud humana, eutrofización marina y ecotoxicidad, gracias a la alta calidad del efluente tratado con MBBR-MBR y a evitar el uso de agente decolorante, que es un compuesto sintetizado a base de una amina cuaternaria.

RESUM

Com un dels sectors més antics i complexos dins de les indústries manufactureres, la indústria tèxtil consumeix grans quantitats d'aigua i produeix grans volums d'aigües residuals durant la seva producció. Les aigües residuals tèxtils presenten sovint una elevada coloració, contenen una àmplia gamma de productes químics orgànics, amb una alta demanda química d'oxigen, així com materials poc degrabilidables. Per tant, comprendre i desenvolupar tecnologies de tractament d'aigües residuals industrials tèxtils eficaces és molt important ambientalment. S'han desenvolupat diferents tractaments, tant processos biològics com físic-químics per al tractament d'aigües residuals tèxtils.

La tecnologia de Bioreactor de membrana (coneguda pel seu acrònim en anglès, MBR) s'ha aplicat àmpliament en el tractament d'aigües residuals tèxtils, mentre que el Reactor de biofilm de llit mòbil (conegut amb les seves inicials en anglès com MBBR) és una tecnologia relativament nova per tractar aquest tipus d'aigües residuals. Tots dos mostren certes deficiències i limitacions durant la seva aplicació. Per tant, un sistema híbrid MBBR-MBR podria ser una solució atractiva als inconvcenients de cadascun d'aquests dos processos individuals.

En aquesta tesi s'ha dissenyat i operatt un sistema híbrid MBBR-MBR per al tractament d'aigües residuals tèxtils. Addicionalment, s'ha estudiat la viabilitat de reutilitzar l'aigua tractada en nous processos de tintura.

El primer pas del treball desenvolupat a la tesi ha estat l'estudi comparatiu del tractament d'aigües residuals tèxtils mitjançant tres processos de tractament, fangs activats convencionals (CAS), MBR i MBBR, treballant en les mateixes condicions de funcionament. El rendiment de cada procés ha estat investigat i comparat des de les perspectives tècnica, econòmica i ambiental. Els resultats van mostrar que, tècnicament, MBR era la tecnologia més eficient, amb valors d'eliminació de la demanda química d'oxigen (COD), de sòlids suspesos totals (TSS) i de color de 91%, 99,4% i 80 %, respectivament, amb un temps de retenció hidràulica (HRT) de 1,3 dies. MBBR, d'altra banda, va tenir un rendiment d'eliminació de COD similar en comparació amb CAS (82% enfront de 83%), per contra aconseguia reduir a la meitat el HRT (1 dia enfront de 2 dies) i eliminava el 73% de TSS , mentre que CAS tenia el 66%. Econòmicament, MBBR era una opció més atractiva per a una planta a escala industrial, ja que estalviava el 68,4% de les despeses de capital (conegut amb les seves inicials en anglès com CAPEX) i tenia les mateixes despeses operatives (conegut amb les seves inicials en anglès com OPEX) que MBR. El sistema MBBR també va tenir menors impactes ambientals en comparació amb els processos CAS i MBR, ja que

reduia el consum d’agent decolorant respecte a CAS i el consum d'electricitatrespecte a MBR i. Segons els resultats de les anàlisis econòmiques i de Anàlisi de Cicle de Vida (conegut amb les seves inicials en anglès com LCA), l'aigua tractada pel sistema MBBR es va seleccionar per a reutilitzar en noves tintures, comprovant quela qualitat dels nous teixits tenyits es trobava dins dels límits acceptables de la indústria tèxtil.

Combinat amb la teoria i els resultats experimentals, es va dissenyar i operar un reactor híbrid MBBR-MBR per al tractament d'aigües residuals tèxtils. El sistema MBBR-MBR va aconseguir reduir la HRT a 1 dia, el que és molt prometedor en la indústria tèxtil en comparació amb el tractament biològic convencional. L'eficiència de remoció de COD va arribar al 93%, que és gairebé el màxim per a un procés biològic de tractament d'aquest tipus d'aigües residuals, així com el rendiment d’eliminació de color, que va arribar al 85%. A més, es va eliminar el 99% dels TSS gràcies a la filtració. A continuació, es van realitzar nous processos de tintura reutilitzant l'aigua tractada. La qualitat dels nous teixits tenyits amb aigua tractada es va comparar amb els teixits de referència. Les diferències de color entre els teixits nous tenyits i els teixits de referència es van trobar dins del requisit general de la indústria tèxtil ($DE_{CMC\ (2:1)}<1$). La reutilització d'aigua tractada en nous processos de tintura és beneficiosa tant per a la indústria com per al medi ambient ja que el sector tèxtil és un consumidor intensiu d'aigua en els seus processos de tintura i acabat.

Addicionalment, amb els resultats experimentals a la planta pilot, es va realitzar un estudi econòmic i una anàlisi de LCA per avaluar la viabilitat econòmica i ambiental de la implementació del sistema'híbrid MBBR-MBR a escala industrial. Econòmicament, MBBR-MBR va tenir despeses de CAPEX i OPEX més baixes que CAS degut a les menors taxes d’abocament d’aigües residuals industrialsi a l'estalvi d'agent decolorant. El resultat del valor actual net (conegut amb les seves inicials en anglès com VPN) i la Taxa Interna de Retorn (conegut amb les seves inicials en anglès com TIR) del 18% va suggerir que MBBR-MBR és financerament aplicable per a la implantació a escala industrial. El sistema MBBR-MBR també va tenir menors impactes ambientals en comparació amb el procés CAS en LCA, especialment en categories, com canvi climàtic, salut humana, eutrofització marina i ecotoxicitat, gràcies a l'alta qualitat de l'efluent tractat amb MBBR-MBR i al fet d’evitar l'ús de l’agent decolorant , que és un compost sintetitzat a partir d'una amina quaternària.

ACRONYMS

AOP	Advanced Oxidation Processes
BOD	Biochemical Oxygen Demand
CAPEX	Capital Expenditures
CAS	Conventional Activated Sludge
CIE	International Commission on Illumination
COD	Chemical Oxygen Demand
EPA	Environmental Protection Agency
EU	European Union
GT	General Tax
HRT	Hydraulic Retention Time
IRR	Internal Rate of Return
INTEXTER	Institut d’investigació Textile i Coperació Industrial
IRR	Internal Rate of Return
LCA	Life Cycle Assessment
MBR	Membrane Bioreactor
MBBR	Moving Bed Biofilm Reactor
NPV	Net Present Value
OM	Organic Material
OPEX	Operational Expenditures
SBR	Sequencing Batch Reactor
SRT	Sludge Retention Time
ST	Specific Tax
TKN	Total Kjeldahl Nitrogen
TP	Total Phosphorus
TSS	Total Suspended Solid
UN	United Nations
VSS	Volatile Suspended Solids

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CHAPTER I. INTRODUCTION

Water scarcity is a global concern. Textile industry, as one of the most water consuming and wastewater producing industries, has the responsibility to develop sustainable and effective wastewater treatment processes.

In this chapter, an introduction of current pressure on water resources and the role of textile industry in it are presented. Additionally, current technologies that have been applied in textile wastewater treatment are introduced.

With the motivation of finding an applicable treatment for wastewater pollution problem of textile industry, firstly, the thesis is focused on two biological technologies, Moving Bed Biofilm Reactor (MBBR) and Membrane Bioreactor (MBR), that have certain advantages over the conventional activated sludge (CAS) treatment in the textile industry. Moreover, a hybrid Moving Bed Biofilm Reactor- Membrane Bioreactor (MBBR-MBR), is designed, constructed, and studied for textile wastewater treatment. Therefore, a review of the state of art of the application of MBR, MBBR and MBBR-MBR in treating and reusing textile wastewater is given.

1.1. Pressure on water resources

Water is one of the most important resources on earth. Without water, there would be no life. The United Nations' (UN) latest World Water Development Report 2019 released that fast-growing water demand and climate change will continue increasing the global pressure on water resources [1.1]. According to the Projected Change in Water Stress of Aqueduct Water Stress Projections (shown in Figure I-1), rapid increases in water stress appeared across many regions including the Mediterranean, Central Asia, and the southwest of North America, within 30 years [1.2].

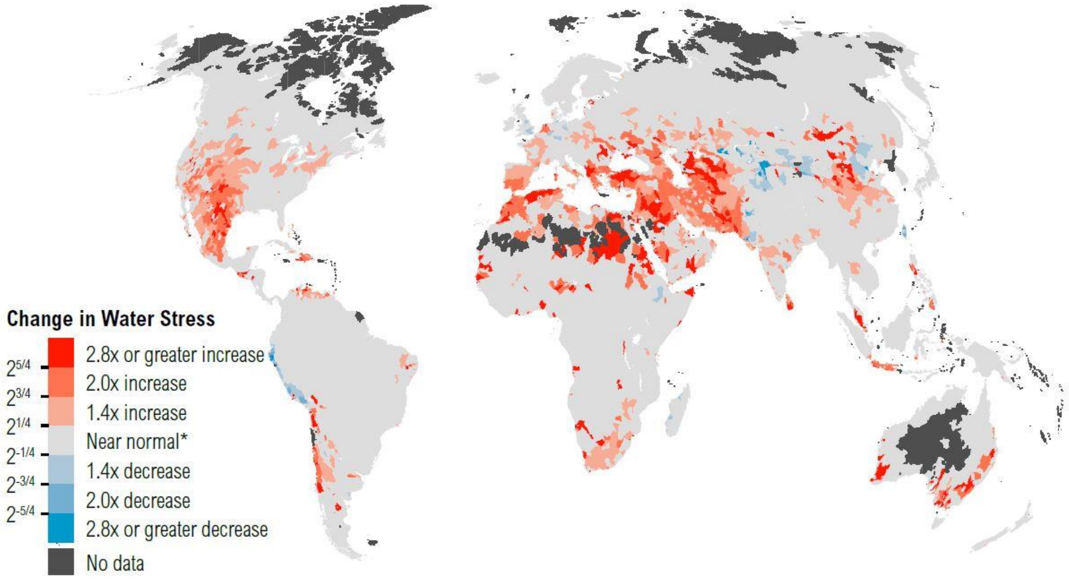

Figure I-1 Changes in Water Stress from 1950–2010 to 2030–50

Agriculture, production of energy, industrial uses and human consumption are the four main sources causing the growing water demand [1.1]. Agriculture consumes about 69 per cent of the water used globally each year, whereas industry is the second-largest water consumer, accounting for about 19 per cent of global water withdrawals. Although agriculture is still the largest water-consumer at present, the demand for industrial water will grow rapidly in the future due to the economic growth [1.3]. The projected increase in global annual water demand from 2005 to 2030, by region and sector is shown in Figure I-2 [1.4]. As can be seen from the figure, the global demand for water, especially in China and India, will continue to rise over the next two decades. Among them, although water demand for municipal and domestic use due to the population growth will account for a large proportion, the continued increase in industrial water demand due to rapid industrial growth should not be taken lightly.

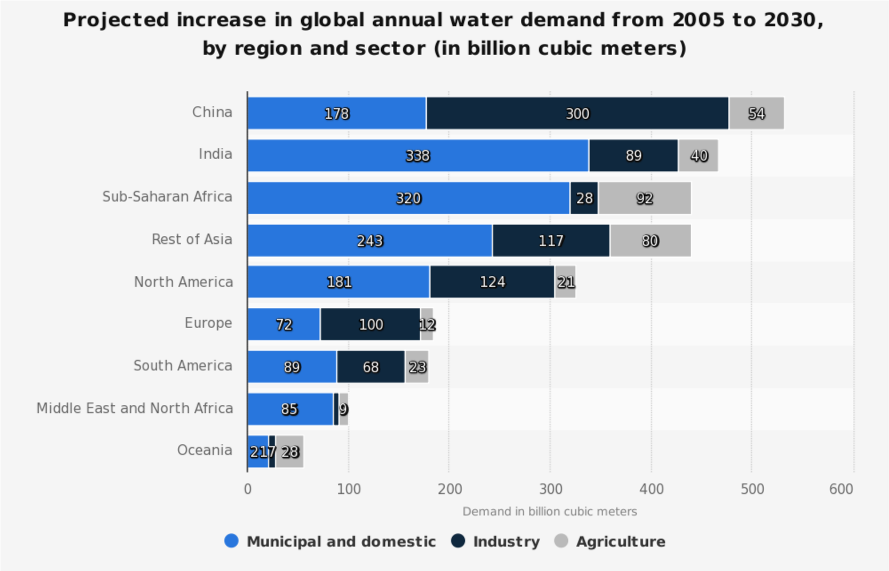

Figure I-2 Projected increase in global annual water demand from 2005 to 2030, by region and sector (Source: https://www.statista.com/statistics/278066/global-water-demand-by-region/, accessed on April 10, 2021)

The distribution of water consumption among the different sectors usually varies depending on the level of development and economic structure of each region. In the case of Spain, water use by sectors are presented in Figure I-3 [1.5]. It can be observed that agricultural activity is the one that consumes the most water (67 %), followed by domestic (14%) and energy (14%)，and finally industrial (5 %).

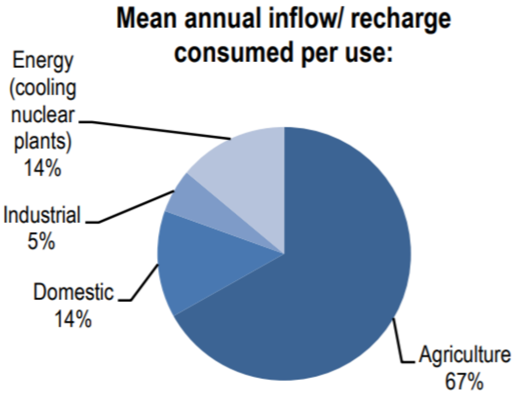

Figure I-3 Water use by sectors in Spain, 2015

(Source: OECD, https://www.oecd.org/spain/Water-Resources-Allocation-Spain.pdf, accessed on April 10, 2021)

Furthermore, water consumption varies depending on the type of industry. In relation to Spain, the latest data found on the distribution of water consumption in the different industrial sectors are from 2009 and is represented in Table I-1, showing the volumes of water captured by different industrial sectors according to the National Classification Code of Economic [1.6]. It can be observed that the textile industry is the fourth sector in water consumption, only surpassed by the metallurgy, paper and food industries.

Table I-1 Volumes of water captured by different industrial sectors in Spain
(Source: http://hispagua.cedex.es/datos/industria, accessed on April 10, 2021)

Type of industrial activity	**Volume of water captured (Hm^3)**
Food, Drink and tobacco	128
Textile	120
Wood	24
Paper	259
Chemical industry	-
Plastic	-
Metallurgy	239
Machinery and Mechanical equipment	16
Electrical, Electronic and Optical equipment	-
Coke plants, Oil refining	-

Textile industry is one of the oldest and most complex sectors within the manufacturing industries and is a leading consumer of water which ranks among tops water consuming industries [1.7]. Along with the large scale of water consumption, great amount of wastewater is produced. Therefore, the adaptation of environmental-friendly technologies remains a challenge to the textile industry. In addition to the use of appropriate treatment technologies, water reuse is also essential for textile industry. Water generated after the wastewater treatment is always crucial for the industries like textile. Treated water with good quality is able to be reused for mainstream manufacturing or production process, which is indeed going to be cost-effective and eco-friendly.

1.2. Wastewater from textile industry

The textile industry is water-intensive during its wet processes, such as sizing, desizing, mercerizing, scouring, bleaching, dyeing, printing, and finishing. The water demands of different wet operations are presented in Table I-2 [1.8].

Table I-2 Water requirements of different wet operations in textile industry

Process	**Requirements in litres/1000 kg products**
Sizing	500-8,200
Desizing	2,500-21,000
Scouring	20,000-45,000
Bleaching	2,500-25,000
Mercerizing	17,000-32,000
Dyeing	10,000-300,000
Printing	8,000-16,000

Furthermore, the volume of water used is only one part of the industry's environmental concerns. Textile wastewater is a mixture of many different compounds consisting of fiber and lint. The major pollutants are organic matters which come from the pre-treatment process, as well as additives and dyes from dyeing and printing processes. Of the total wastewater volume, pre-treatment wastewater accounts for about 45% and dyeing/printing process wastewater accounts for about 50% - 55%, while finishing process produces little. Table I-3 shows the typical wastewater generated during each textile textiles' manufacturing step.

Table I-3 Summary of wastewater generation during textiles' manufacturing

Process	**Wastewater**
Slashing/sizing	Biochemical Oxygen Demand (BOD); Chemical Oxygen Demand (COD); metals; cleaning waste, size
Desizing	BOD from water-soluble sizes; synthetic size; lubricants; biocides; antistatic compounds
Scouring	Disinfectants and insecticide residues; NaOH; detergents, fats; oils; pectin; wax; knitting lubricants; spinfinishes; sent solvents
Bleaching	Hydrogen peroxide, sodium silicate or organic stabilizer, high pH
Mercerizing	High pH; NaOH
Dyeing	Metals; salts; surfactants; toxics; organic proccessing assistants; cationic materials; color; BOD; COD; sulfide; acidity/alkalinity; spent solvents
Pringting	Suspended solids; urea; solvents; color; metals; heat; BOD; foam
Finishing	BOD; COD; suspended solids; toxics; spent solvents

The characteristics of textile wastewater can vary largely between processes and materials. Due to the wide range of processes, textile wastewater contains a complex mixture of chemicals and there is not a typical effluent. However, in general, major pollutants in textile wastewaters are high total suspended solids (TSS), COD, heat, color, and other soluble substances [1.9]. The typical characteriscs of textile wastewater are presented in Table I-4 [1.10]**.**

Table I-4 The typical characteristics of the textile wastewater for different types of effluents

Parameters	**Industrial activities**					
	Yarn	**Flock**	**Cotton woven fabric**	**Cotton knitted fabric**	**Wool fabric**	**Printing**
pH	5-12	6-10	8-12	6-11	6-8	7-10
SS (mg/L)	50-150	50-150	50-300	50-150	100-150	200-600
COD (mg/L)	500-900	4000-7000	1000-3000	800-1300	300-1000	2000-4000
BOD_5 (mg/L)	150-350	1200-2200	300-1000	200-450	100-400	500-1500
Color (units Pt-Co)	300-1000	300-1000	300-3000	100-1000	200-1500	1000-6000
Toxicity (equitox/m3)	3-10	3-10	4-15	4-10	5-25	variable
Volume ratio (L eff./Kg fiber)	10-80		100-300	80-120	100-300	variable

1.3. Standards for textile wastewater discharge in Spain and in the autonomous community of Catalonia

As the wastewater from industries is harmful to the environment and living beings, strict requirements for the emission of wastewater have been made. It varies according to local situation in different regions due to the difference in the raw materials, products, dyes, technology and equipment.

There are two standards for industrial effluents in Spain. One is for discharge to a water purifying plant which depends on the local legislation and the other is for discharge directly to natural rivers. The limits of discharge to rivers are more strict than to a treatment plant since the wastewater is discharged directly into the natural environment. As the experiments of this thesis was performed in Catalonia, the catalan standard for industrial effluent discharge to a water purifying plant is reported in the Annex II of Decreto 130/2003 [1.11], showing in Table I-5.

Table I-5 Discharge limits of industrial effluents in Catalonia

Parameters	**Limit of Value**	**Units**
T (ºC)	40	ºC
pH (interval)	6-10	pH
SS	750	mg/L
DBO_5	750	mg/L O_2
COD	1500	mg/L O_2
Oils and fats	250	mg/L
Chloride	2500	mg/L Cl^-
Conductivity	6000	µS/cm
Sulfur dioxide	15	mg/L SO_4^{2-}
Sulphates	1000	mg/L SO_4
Total Sulphates	1	mg/L S^{2-}
Dissolved Sulphates	0.3	mg/L S^{2-}
Total Phosphorus	50	mg/L P
Nitrates	100	mg/L NO^{3-}
Ammonium	60	mg/L NH_4^+
Organic and ammonia nitrogen	90	mg/L N

The Spanish national standard of industrial effluent discharge to rivers is reported in the Annex IV of Real Decreto 849/1986 [1.12]. In the case of the standard of industrial effluent discharge to rivers in Catalonia, the Table 3 of Table I-6 is applied, the most restrictive. The limit value for each parametre is shown in Table I-6.

Table I-6 Discharge limits of industrial effluentes in Spain

Parametre/Unit	**Limit of Value**		
	Table 1	**Table 2**	**Table 3**
pH (interval)	Between 5.5 and 9.5		
SS (mg/L)	300	150	80
DBO_5 (mg/L) O_2	300	60	40
DQO (mg/L) O_2	500	200	160
Color	inestimable in solution		
	1/40	1/30	1/20
Toxic metal	3	3	3
Cyanide (mg/L)	1	0.5	0.5
Chloride (mg/L)	2000	2000	2000
Sulfide (mg/L)	2	1	1
Sulphite (mg/L)	2	1	1
Sulfate (mg/L)	2000	2000	2000
Fluoride (mg/L)	12	8	6
Total phosphorus (mg/L)	20	20	10
Ammonia (mg/L)	50	50	15
Nitrite nitrogen (mg/L)	20	12	10
Oils and fats (mg/L)	40	25	20
Phenol (mg/L)	1	0.5	0.5
Aldehydes (mg/L)	2	1	1
Detergents (mg/L)	6	3	2
Pesticides (mg/L)	0.05	0.05	0.05

1.4. Textile wastewater treatment processes

In response to such the complicated problems of textile wastewater, textile industries must have their own purification system in order to obey the discharge limits presented in Section 1.3. In generall, there are three main stages of the wastewater treatment process, known as primary, secondary and tertiary treatment.

Primary treatment includes the physical processes of screening, comminution, grit removal, and sedimentation to remove floating or settleable materials. Secondary treatment removes the soluble organic matter that escapes from the primary treatment. Most secondary treatment of textile industry are grouped into two large groups: physical-chemical and biological. The most common physical-chemical treatment is coagulation-floculation and the most applied biological method is Conventional Activated Sludge (CAS) system. Although in some cases, combined treatments are necessary depending on the pollutants contained in the effluent. In many instances, the secondary treatment is not enough to bring textile wastewater up to color discharge standards and a tertiary treatment will be required. The following sections will focus on presenting the frequently used secondary and tertiary treatments for textile wastewater treatment.

1.4.1. Secondary physicochemical treatment

Physicochemical wastewater treatment has been widely used in the sewage treatment plant in the past as the secondary treatment of textile wastewater. The most common physicochemical method is the traditional coagulation-flocculation techniques. Chemical coagulation-flocculation process changes the physical state of dissolved solids and suspended matter by adding synthetic or natural coagulant and flocculant [1.13] and facilitates their removal by sedimentation. This method is able to provide an efficient color removal in colloidal suspension of dyes with the optimum coagulant dose, however, ineffective decolorization would occur with most of soluble dyes in the wastewater [1.14]. Besides, coagulation-flocculation has distinct disadvantages such as the insufficient removal of COD, the large production of sludge, and the increase of pH and salinity. The residue generated requires an additional treatment to be destroyed which would increase the operation cost.

1.4.2. Secondary biological wastewater treatment

The coagulation-flocculation method could achieve satisfactory color removal efficiency, but with some drawbacks. One of the main disadvantages of coagulation-flocculation is the insufficient COD removal, therefore nowadays textile industries generally use biological treatment method to treat their effluent. In general, the biological treatments are cheaper and more environmental-friendly than physicochemical ones and can remove dissolved matters in a way similar to the self-depuration but in a further and more efficient way than flocculation.

The most applied conventional biological treatment is the CAS system as is well-known. Subsequently, advanced biological treatments have been developed, such as the introduction of membranes for replacing the decanter by filtration or the addition of carriers in the biological reactor, known as MBBR, for improving the treating capacity.

1.4.2.1. Conventional Activated Sludge system

One of the most applied biological methods in treating textile wastewater is the conventional activated sludge (CAS) process [1.15]. The main objective of the CAS process is to remove the organic compounds [1.16]. CAS system has disadvantages such as long Hydraulic Retention Time (HRT), problems with sludge settling, requirement of large space [1.17] and has poor efficiency of color removal due to the low biodegradability of dyes used in various processes of production [1.18]. Hence, a tertiary physicochemical method is usually required for a better treating performance [1.19, 1.20], which will increase the cost of the process.

1.4.2.2. Membrane Bioreactor

In the past two decades, noticeable progress has been achieved on the Membrane Bioreactor (MBR) technology in industrial wastewater treatment. MBR is the combination of conventional biological wastewater treatment and membrane filtration. MBR differ from conventional biological wastewater treatment in the separation of activated sludge and treated wastewater [1.21]. The influent is fed into the aerated bioreactor where the organic components are oxidized by the activated sludge. The aqueous activated sludge solution then passes through a micro or ultrafiltration membrane filtration unit, separating the water from the sludge. The latter returns to the bioreactor, while permeate is discharged or reused as particle-free effluent. The membrane configurations used in MBR are mainly tubular, hollow fiber, and flat sheet (plate or frame). The membrane materials can be classified into three major categories: polymeric, metallic and inorganic (ceramic) and there are two configurations of MBR systems: external and submerged. Membrane processes can be classified according to the pore size and retained species (Table I-7) [1.22].

Table I-7 Classification of membranes according to pore size and retained species.

Membranes	**Pore size (μm)**	**Retained species**
Microfiltration	> 0.01	Large colloids, bacteria
Ultrafitration	0.005 – 0.01	Macro molecules, proteins
Nanofiltration	0.001 – 0.005	Multivalent salts
Reverse osmosis (RO)	< 0.001	Monovalent salts

Due to use of membranes in MBR, it is possible to retain all suspended solids and microbial flocks. Consequently, a longer solid retention time can be achieved in MBR systems which is not possible in conventional wastewater treatment systems. The main advantages of MBR systems in front of conventional biological treatment are [1.23, 1.24]:

- Enhanced effluent quality
- Efficient disinfection capability
- Lower space requirement
- Greater volumetric loading
- Minimal sludge production
- Shorter start-up time
- Low operating and maintenance manpower requirement

1.4.2.3. Moving Bed Biofilm Reactor

Recently, biofilm systems have drawn much attention in treating different types of industrial wastewater due to several advantages comparing with conventional biological treatment, such as space-saving [1.25, 1.26]. Among them, Moving Bed Biofilm Reactor (MBBR) has been applied in textile wastewater treatment in the last few years. The basic principle of MBBR technology is the biofilm method, which not only has the advantages of activated sludge method, but also overcomes the disadvantages of traditional sewage treatment methods and the defects of fixed biofilm method. MBBR uses carriers with the specific gravity similar to water, which can move freely with the flow of water. As the air oxidizes the suspended biological filler in the reactor, a layer of biofilm will be formed on the outer and inner surfaces of the filler. This system changes the living environment of microorganisms from the original liquid-gas two-phase environment to solid-liquid-gas three-phase environment, which provides good conditions for microbial activities. One of the highlights of MBBR is a smaller volume of the biological plant or a larger treating capacity in the same reactor volume due to the biofilm attached to carriers. Besides the great amount of biomass fixed on carriers, the concentration of biomass in suspension could be higher than a CAS process [1.27].

Both advanced biological systems, MBR and MBBR, have been developed in industrial wastewater treatment, but they have not been explored completely in textile wastewater treatment, especially MBBR. In recent years, a novel combined technology named as Moving Bed Biofilm Reactor-Membrane Bioreactor (MBBR-MBR) has been applied in treating urban wastewater and previous studies showed its superior capacity to the MBR for the treatment of municipal wastewater [1.28, 1.29]. The MBBR-MBR has many advantages over the MBR process such as less sludge production, higher organic loading capacity, better oxygen transfer and more benefits than MBBR system such as higher biological reaction rates through the accumulation of high concentrations of active biomass on the carries and better sludge decantation [1.30]. With all the benefits MBBR-MBR could have, the hybrid system could be an attractive solution for dyeing wastewater treatment.

1.4.3. Tertiary wastewater treatment

Since the conventional treatment methods could not achieve the sufficient color removal, there is the need for efficient tertiary treatment process. The most common tertiary treatments are:

- **Coagulation-flocculation** techniques, is observed as one of the most practised techniques used as tertiary treatment to remove color. Regardless of the generation of considerable amount of sludge, it is still widerly applied in developed and in developing countries [1.14]. However, the main limitation of this process is the generation of a residue which requires an additional treatment and the increase of wastewater conductivity due to the use of inorganic salts as coagulant [1.31, 1.32].
- **Adsorption** is another most popular technology to treat dye contained wastewater as tertiary treatment. The effectiveness of color removal is based on the dye-adsorbent interactions, surface area of adsorbent, particle size, temperature, pH and contact time [1.31, 1.33]. Activated carbon is the most commonly used adsorbent and can be very effective for many dyes [1.34]. However, the cost of carbon is relatively high and the number of reuses of the adsorbent is limited because the large molecules of dyes are rather well adsorbed, but desorption is extremely difficult and pyrolysis is required [1.32]. The limitations of this

technology are the disposal of used adsorbents, excessive maintenance costs which make it hard to apply in field scale applications [1.14].

- A**dvanced Oxidation Process (AOP)** is a family of methods that use oxidising agents such as ozone (O_3), hydrogen peroxide (H_2O_2) and permanganate (MnO_4) to change the chemical composition of dyes. AOP includes Fenton (H_2O_2/Fe^{2+}) and Photo-Fenton (UV/ H_2O_2/Fe^{2+}) reactions [1.35, 1.36], systems based on H_2O_2/ UV light [1.37], heterogeneous photocatalysis (UV/ TiO_2) [1.38, 1.39] and ozonation. These methods have, as a common disadvantage, their high cost of energy and reagents. Most of the AOP for textile wastewaters are highly expensive and its effectiveness varies widely with the type of constituents present in the textile wastewaters.

To sum up, tertiary treatment is necessary in many cases of textile wastewater treatment when the secondary treatment has not achieved the color removal requirements, but most of the physicochemical tertiary treatments are either expensive or generating new pollutant along with the treatment.

1.5. State of art of MBR, MBBR and MBBR-MBR applied in textile wastewater treatment and reuse

The increasingly restrictive environmental regulations are forcing textile industries to treat their effluents with more efficient systems. As mentioned in the last section, although conventional activated sludge system is the most applied technology in industrial-scale wastewater treatment for textile industry recently, other biological treatments have been studied to find a more sustainable solution for treating textile effluent. This section presents a literature review of aerobic and anaerobic Membrane Bioreactor (MBR) system that has been used in textile wastewater treatment and in some cases, the combination of MBR with other methods. Secondly, the application of Moving Bed Biofilm Reactor (MBBR) system as a relatively new technology applied in this sector is reviewed. Thirdly, the development of the novel hybrid MBBR-MBR process is investigated, and finally, the situation of the reuse possibilities after each of these treatments is studied.

1.5.1. Application of aerobic MBRs

Aerobic MBR combines the Conventional Activated Sludge (CAS) process with membrane filtration. Compared to CAS, MBR has become an attractive wastewater treatment technology due to its very high-quality treated water. A comparative study between aerobic MBR and CAS treating textile effluent showed that the rate of reduction of COD was 89-92% with MBR while 54-70% with CAS and the color removal rate was 72-73% with MBR (UF) and only 28% with CAS [1.40]. Another comparative study of aerobic MBR and CAS treating textile wastewater evaluated the working performance and kinetic coefficients of both systems under the similar conditions [1.15]. Their results showed that the high values of the maximum specific substrate utilization rate (k) in MBR process prove that the biomass employed more efficiently the organic matter than an CAS process. High values of the half-velocity constant (K) demonstrate that the MBR accept higher concentrations than CAS. As well as low true yield coefficient (Y) in MBR show a lower sludge production than CAS. These results can show that the MBR process is more attractive to treat textile wastewater than a CAS, due to the less production of sludge, accepting high organic concentrations and higher substrate utilization rate.

The research of the first case of aerobic MBR applied in textile wastewater treatment in Bangladesh showed that the performance of the MBR system with 90% of COD removed was significantly better than that of the CAS process with a low removal rate of 40-50% [1.41]. Chamam et al. (2007) made a comparison of treating textile effluent by bio-sorption and membrane bioreactor. The effluent in this study contained *Cassulfon* CMR which is a sulphonic textile dye mainly used to color “jeans” and the results confirmed the remarkably high potential of MBR to treat such dye effluents. The permeate quality was always free of suspended solids or turbidity [1.42]. Another comparative study of MBR and sequencing batch reactor (SBR) for dyeing wastewater treatment showed that the removal efficiencies of the MBR system for color, COD, BOD, and SS were 54, 79, 99 and 100%, respectively, all higher than the corresponding parameters for the SBR process [1.43].

Most of the previous studies confirmed that aerobic MBR is efficient for reducing the organic compounds from textile wastewater, but not as effective for eliminating color. A submerged hollow fiber aerobic MBR was studied by Huang et al. (2009) showing that the system was capable to treat dyeing wastewater up to 400 L/d and the removal ratio of COD reached 90%

and 60-75% for color removal [1.44]. The main mechanism of color removal was adsorption of dye molecules onto biomass. Therefore, the sludge growth was important to maintain a maximum color removal efficiency [1.45]. Yigit et al. (2009) investigated a pilot-scale aerobic MBR system for the treatment of textile wastewater from wet processes of a denim producing industry. Remarkably high removal efficiencies were obtained for various parameters and the treated wastewater had high potential for reuse in the textile industry [1.46].

It should be mentioned that the operation settings are important for the performance of aerobic MBR and depends on the characteristics of the membrane and specific treatment. The hydraulic retention time (HRT) which determines the treating capacity varies according to specific cases, however, in industrial scale treatment, the HRT usually tends to be around 2 days. Friha et al. (2015) reported the performance of aerobic MBR in treating raw textile wastewater and the efficiency of the MBR in reducing cytotoxicity. High removal efficiencies were achieved for COD, color and SS and the cytotoxicity was significantly reduced by MBR when operating at HRT of 2 days [1.47]. Another study of Konsowa et al. (2013) also found out that with the increase of HRT in the aerobic submerged MBR, the removal rate of COD and dye were improved. With HRT of 2 days, dye removal efficiency was achieved to 95% [1.48]. The long solid retention time (SRT) in MBR is also a benefit comparing with CAS. Longer SRT helps to increase sludge concentration and thus reduce the organic load, which has the advantages of small footprint and low sludge production. Some previous studies suggested that higher sludge concentration resulted in less fouling at longer SRT and lower Food-to-Mass (F/M) ratio [1.49]. The study of Innocenti et al. (2002) reported that the maximum nitrification was occurred when the SRT was changed from short to longer. This is likely due to the time allowed for slow growing microorganisms to exist at high SRTs in the MBR process [1.50]. The suction and backwash time settings is also important in the treating process. Schoeberl et al. (2005) observed that suction time was most important to have the largest effect on resistance increase followed by aeration intensity and backflush time during the optimization of operational parameter for an aerobic MBR treating dye house wastewater. The results of their study showed that COD and color removal from textile wastewater was 89–94% and 65–91%, respectively [1.51].

The results of aerobic MBRs applied in textile wastewater treatment are summarized in Table I-8.

Table I-8 Results of aerobic MBRs applied in textile wastewater treatment

MBR type	**Sample**	**Influent COD (mg/L)**	**COD removal (%)**	**Color removal (%)**	**References**
Aerobic MBR	Textile wastewater		89-92	70	[1.40]
Aerobic MBR	Textile mill		90		[1.41]
Aerobic MBR	Textile wastewater		79	54	[1.43]
Aerobic MBR	Dyeing wastewater	600-1200	85-92	60-75	[1.44]
Aerobic External MBR	Wastewater from a polyester finishing factory	1380-6033	76-90	46-98.5	[1.45]
Aerobic MBR	Denim producing textile wastewater	686 - 2278	97	> 97	[1.46]
Aerobic MBR	Textile wastewater	1463-3089	>90	97	[1.47]
Aerobic MBR	Textile wastewater with direct fast red dye-CI 81		87.7-96.3		[1.48]
Aerobic MBR	Dyehouse wastewater		89-94	65-91	[1.51]

The review on previous studies of aerobic MBRs applied in textile wastewater treatment informed that the aerobic MBR technology is able to treat textile wastewaters with the COD value varying from 600 to 6000 mg/L. Due to the variability of wastewater characteristics, although aerobic MBRs are effective in COD removal, the removal rates are also variable with a value range of 76-96%. Color removal by aerobic MBRs, on the other hand, is less effective. Although certain studies achieved high color removal rates, the color removal efficiency of most studies were variable and insufficient with a value range of 46-97. The value ranges obtained by this review are similar to those observed by other previous reviews [1.52].

1.5.2. Application of anaerobic MBRs

In recent years, in order to improve the color removal performance of MBR, anaerobic digestion has been studied and applied for textile wastewater treatment. Although, few studies were made only using anaerobic MBR because membrane fouling problem of anaerobic MBR is more serious than aerobic MBR. Lin et al. (2013) noted in their review study that the treatment of textile wastewater using solo anaerobic MBR barely has been reported [1.53]. The more common application of anaerobic MBR is its combination with other processes, which are presented in the next Section 1.5.3.

1.5.3. Application of MBR combining other advanced treatment technologies

In the recent past, a significant number of research studies have been carried out to improve the decolorization and fouling control of treating textile wastewater using MBR together with different combinations of physical, chemical and physic–chemical treatment techniques.

In order to achieve a high removal efficiency of organic compounds and color, several studies chose to use an anaerobic tank followed by an MBR unit. For example, Fan et al. (2000) studied a treatment system for dye wastewater from a woolen mill. It was composed with an anaerobic tank and a MBR unit. The average removal of COD, BOD, color and turbidity was 82%, 96%, 71%, 99%, respectively [1.54]. Zheng et al. (2003) reported the performance of a pilot-scale anaerobic tank followed by an MBR on treating wastewater from a Woolen Mill with the initial concentration range 179-358 mg/L of COD. The quality of treated water was excellent and met with the reuse water standard with the similar removal rate of the previous study [1.55]. Zheng and Liu (2004) carried out a study of a combined process of an anaerobic reactor and a MBR. The results showed that the removal rates of COD, BOD_5, color and turbidity were 80.3%, 95%, 59% and 99.3%, respectively [1.56]. In the study of You and Teng (2009), an anaerobic SBR plus aerobic MBR was tested for dyeing wastewater treatment containing an azo dye, Reactive Black 5. Nearly 97% of COD removal and 83% of true color removal was achieved using the anaerobic SBR and the aerobic MBR, respectively [1.57].

Normally, textile industries use two technologies for their production: dyeing or printing. In the wastewater from dyeing processes, the presence of nitrogen is limited. However, printing processes with urea generate wastewater with high amount of nitrogen in the subsequent equipment washing. Anoxic/oxic MBRs are a common option for the elimination of nitrogen.

The study of Chung et al. (2004) was dedicated in getting a better insight of denitrification/nitrification MBR process and optimum operational conditions to treat textile wastewater with high organic and nitrogen contents. Two experimental units, oxic MBR and anoxic/oxic MBR were operated. The results showed that anoxic/oxic MBR (COD removal rate 91.9%) outperformed oxic MBR (COD removal rate 81.5%) in removal efficiency of various parameters and in terms of nitrogen control [1.58]. Sun et al. (2015) studied the performance of an anaerobic-anoxic-aerobic MBR at removing organic compounds and nitrogen for treatment of textile wastewater [1.59]. Tian et al. (2015) carried out a study of the performance of hybrid anoxic/oxic MBR in simultaneous organic carbon and nitrogen removal from fiber wastewater. The results obtained when HRT > 32 h showed that the average removal efficiency of COD, NH^{4+}–N and TN in the hybrid A/O MBR could reach 56.5, 86.6 and 45.9 %, respectively. After supplementing alkalinity, the removal efficiency of NH^{4+}–N and TN reached 86.9% and 60.5%, respectively [1.60].

There are other studies that obtained higher organic matter removal and better membrane fouling control by combining MBR with coagulant. In the study of Baêta 2012, a submerged anaerobic MBR combined with PAC was applied in textile wastewater treatment which obtained the median removal efficiency of COD and color with 90% and 94%, respectively [1.61]. In a study carried out by Yan et al. (2009) a pilot-scale hybrid coagulation-MBR was investigated for real textile wastewater treatment. Poly-aluminum chloride (PAC) was used in the process. The hybrid system achieved much higher organic matter removal than that of MBR [1.62]. Teli et al. (2012) reported MBR fouling control and permeated quality enhanced by PAC. The pilot plant operated in two steps: 7.5 months without flux enhancer and 3 months with the addition of PAC. The addition of PAC showed a significant decrease of the filtration resistance due to cake layer formation and an increase of color and anionic surfactants removal rate [1.63]. Thanh et al. (2012) studied the fouling control of a submerged MBR treating dyeing wastewater by using Powder-Activated Carbon and Alum. The results demonstrated that the addition of activated carbon and alum into the MBR system improved the COD and color removal efficiency, and the fouling was also well controlled after the addition [1.64].

Other authors combined MBR with the use of specific microorganisms, mainly fungi, to enhance the treatment efficiency. In this respect, Hai et al. (2008) reported the excellent fouling prevention capacity of a fungi MBR. White-rot fungi *C. versicolor* was used for this study. The system showed stable performance of the MLSS concentration (up to 25 g/L) [1.65]. They also used the system to study the decolorization capacity of pure fungus as well as MBR-sludge. After the addition of powdered activated, excellent stable dye removal and stable enzymatic activity was observed [1.66]. Another research studied the key factors for fungal decolorization in MBR under non-sterile environment. The MBR obtained a 93% removal efficiency of azo dye (Acid Orange II). Results demonstrated the adverse effect of bacterial contamination on fungal activity [1.67]. Taylor et al. (2017) investigated the reactive dye removal ability of mixed filamentous fungal strains with submerged MBR in non-sterile conditions. They had the conclusion of using mixed fungal strains in the MBR system is efficient for removing reactive dyes with the removal efficiency of color and COD were 90.71% and 90%, respectively [1.68].

The combination of MBR and oxidation or advanced oxidation is aimed to eliminate the color from dye-contained water. Brik et al. (2004) reported that three oxidation treatments were tested to improve the efficiency of color removal of MBR: ozonation, chlorination and hydrogen peroxide oxidation. The result showed that ozonation was the most efficiency method that by using only 38 mg/L within 20 minutes, it was possible to achieve the reuse recommendation with a satisfactory color removal of 93% [1.69]. Feng et al. (2020) investigated the performance of MBR combined with Fenton oxidation for the treatment of dyeing wastewater. They used ferrous sulphate ($FeSO_4 \cdot 7H_2O$), H_2O_2 (30%, W/W) and diethylene glycol as reagents for Fenton oxidation process. The results showed that the removal of TOC and color was 88% and 90%, respectively [1.70].

The textile wastewater, especially that from spent cotton reactive dyebaths, contains high salt content [1.71]. Several studies investigated the possibility of recover the salt from wastewater for new productions and most of these studies used MBR followed by NF or RO. Grilli et al. (2011) evaluated the treatability of textile wastewaters in a bench-scale experimental system including an anaerobic biofilter, an anoxic reactor and an aerobic MBR followed by a NF membrane. The results showed a good COD (90-95%) removal in the MBR system because of the presence of the anaerobic biofilter and an effective color removal (70%) was obtained [1.72]. Moreover, salt was also separated from effluent by the NF membrane [1.73]. Jager et al. (2012) analyzed a pilot-scale dual-stage MBR which was consisted of two phases: the side-stream UF-MBR followed by NF and RO. During the study, the UF-MBR treatment system and RO polishing step showed average overall COD removals of 75 and 90.1%, and color removals of 28.6 and 97.2%, respectively [1.74].

The results of MBR combining other advanced technologies applied in textile wastewater treatment are summarized in Table I-9.

Table I-9 Results of MBR combining other advanced technologies applied in textile wastewater treatment

MBR type	**Sample**	**Influent COD (mg/L)**	**COD removal (%)**	**Color removal (%)**	**References**
Anaerobic + aerobic MBR	Woolen mill	54-473	82	71	[1.54]
Anaerobic + aerobic MBR	Woolen mill	179-358	92.4	74	[1.55]
Anaerobic SBR +aerobic MBR	Synthetic dyeing water	128-321	80.3	59	[1.56]
Anaerobic-anoxic-aerobic MBR	Textile wastewater	657-944	85		[1.59]
Anaerobic-biofilm + anoxic-aerobic MBR + NF	Textile wastewater		90-95	70-90	[1.72]
UF-MBR+NF/RO	Textile wastewater	5815	75 (UF) 86 (NF) 90 (RO)	28.6 (UF) 98 (NF) 97 (RO)	[1.73]
Oxidation treatments + MBR	Textile mill	4000-6200	>80	50-90	[1.69]
Fenton oxidation + MBR	Dyeing wastewater	1100-1300		69.5	[1.70]
Coagulation + MBR	93% Dyeing wastewater	393-534	90.7	83.7	[1.62]
Coagulation + MBR	65% textile wastewater	284	81	68	[1.63]

1.5.4. Status of MBBRs applied in textile wastewater treatment

Moving Bed Biofilm Reactor (MBBR) has been applied in many cases of industrial wastewater treatment, but it is a relatively novel technology for treating textile dyeing wastewater. Combining the advantages of suspended growth and biofilm system, the MBBR has been developed more efficient than CAS process to treat textile wastewater because a large quantity of biomass can be maintained in the reactor by using carriers. However, MBBR has the same sludge decantation problem as CAS. From the consulted bibliography, it is clear that MBBR applied to treat textile wastewater requires the addition of coagulant, this is common in MBBR technology to improve sludge decantation, but in addition to textiles, it is essential to achieve good levels of decolorization, as observed in the work done by Shin et al. (2006). They studied a combined process consisted of a MBBR and chemical coagulation for textile wastewater treatment. The MBBR system had anaerobic-aerobic-aerobic in series followed by chemical coagulation with $FeCl_2$. After the MBBR process, 85% of COD and 70% of color were removed. After the coagulation, 95% of COD and 97% of color were removed [1.75]. Park et al. (2010) reported an anaerobic-anaerobic-aerobic MBBRs treating textile dyeing wastewater. Polyurethane-activated carbon (PU-AC) foam were filled with 20% for biological treatment. After an eight-day operation, 86% of the total COD was removed [1.27]. Other studies combined MBBR with oxidation in order to improve the removal of color. For example, Castro et al. (2017) studied the combination of ozonation and MBBR for treating textile wastewater with Reactive Orange 16 azo dye. They eliminated 93% of the COD and 97% of the color, respectively [1.25]. Francis and Sosamony (2016) studied the performance of MBBR on pre-treated textile wastewater by Fenton oxidation. After the oxidation, 86% COD removal was achieved [1.76]. Gong (2016) investigated a four-stage lab-scale treatment system (anaerobic–aerobic MBBR–ozonation–aerobic MBBR) in series for textile wastewater treatment. Although the results showed great removal efficiencies of COD and color were 94.3%, and 96.3%, respectively, energy consuming, large space, and maintenance of four stages of treatment should be taken into account when such complex system wants to be applied in industrial-scale treatment [1.77].

1.5.5. Status of MBR-MBBR applied in textile wastewater treatment

The advantages and disadvantages of the MBBR and MBR systems are presented in their status of art. In conclusion, among the main advantages of the MBR systems, one of them is the ability to work at much higher biomass than activated sludge processes, which results in lower volume biological reactors. However, we find that the reduction of reactor volume is limited because the biomass concentration in practice has a limit. How could we solve this problem? MBBR is just happens to be the solution with its main advantage. MBBR can withstand large organic load thanks to the biomass fixed on the carriers. Moreover, MBBR can reduce the volume of the biological reactor, or to treat a larger organic load in the same reactor volume. However, MBBR often requires the addition of a certain amount of coagulants for a better sludge decantation. It is interesting to note that MBR can solve the poor decantation of MBBR. Both MBR and MBBR have been studied in textile wastewater treatment individually, MBR-MBBR hybrid system could be an attractive solution for dyeing water purification because of the high efficiency and low consumption of energy and space.

Pervissian et al. (2012) did an assessment of the performance of MBBR-MF system for treatment of industrial wastewater. The total COD removed was 97% and the fouling of the membrane was reduced with the MBBR [1.78]. To our best knowledge, there was only one study about MBBR-membrane filtration applied in textile wastewater treatment, in which a combined anaerobic-aerobic MBBR-MF was investigated for the treatment of azo dye reactive brilliant red X-3B. The COD and color removal rate achieved at 85% and 90%, respectively. The color reductions mainly occurred in anaerobic conditions [1.79].

1.5.6. Reuse of treated water by MBR or MBBR in new textile processes

What can be done to reduce the water footprint of textiles? In addition to an effective technology that can helps the wastewater meet the discharge standard, the feasibility of reuse the treated water in new productive processes is also valuable.

Some researchers have performed water reuse experiments after MBR treatment, such as the Malpei et al. (2003). They concluded that after the treatment of MBR, the textile treated effluent was suitable for reuse in some operations of the dyeing cycle such as the first washing [1.80]. Brik et al. (2004) found that it was possible to reuse the treated water by MBR combined with ozonation in new textile processes with a satisfactory color removal rate [1.69]. Sert et al. (2017) suggested that the use of a nanofiltration (NF) after the MBR process is a good option to treat and reuse of MBR effluent [1.81]. In the study of Cinperi et al. (2019), pilot-scale plants employing membrane bioreactor (MBR), nanofiltration (NF) and brackish water reverse osmosis (BWRO) processes at different test conditions was investigated. Their results showed that the reuse of MBR+NF+UV and MBR+RO+ UV effluents achieve the reuse requirement for new dyeing process [1.82]. These previous studies of reuse after MBR treatment required the combination of other technologies: ozonation or other membrane technologies such as NF or RO, mainly to remove color to meet reuse criteria.

MBBR also requires combination with other techniques to be able to reuse the treated water. Gong (2016) investigated a four-stage lab-scale treatment system (anaerobic MBBR-aerobic MBBR-ozonation-aerobic MBBR in series) and the final effluent could meet the reuse requirements of textile industry [1.77].

1.6. Scope of the thesis

An overview of the previous literature on Membrane Bioreactor (MBR) and Moving Bed Biofilm Reactor (MBBR) applied in textile wastewater treatment is given in the state of art.

MBR was found a well-developed technology in the treatment of textile effluent, it was effective in removal of organic compounds of textile wastewater, but in many cases, the combination with other technologies is needed to eliminate color from the water.

MBBR process used in textile wastewater treatment showed that they can operate with high concentrations of biomass but with the need of extra coagulation for better decantation.

In the process of literature searching, we found that only few studies have investigated the combination of MBR and MBBR technologies. MBBR-MBR can work at high organic loading rates because MBBR can remove most biodegradable contaminants and the particulate components can be separated by MBR. Furthermore, MBBR-MBR system will reduce the space and energy consumption comparing with MBR. Therefore, development of the application of MBBR-MBR should be attractive to textile wastewater treatment as a reliable and effective method.

The present thesis will focus on treating real textile wastewater from a local textile finishing industry. The textile industry has CAS plant for its wastewater treatment which allows us to compare the applicability of different methods. Moreover, as this textile industry works with both synthetic fiber (polyester) and natural fiber (cotton), the characteristics of its wastewater are representative, and the conclusions obtained from treating its wastewater could be applicable to the vast majority of textile industries. In the first stage of the thesis, laboratory-scale pilot-plants of CAS, MBR and MBBR will be investigated and compared in the wastewater treatment to verify the feasibility of applying the combined MBBR-MBR system.

Afterwards, the MBBR-MBR pilot-plant will be designed, constructed, and optimized for the textile wastewater treatment. New dyeings made with the treated water will be performed to verify the treating efficiency and quality. Additionally, based on the experimental results obtained by applying the MBBR-MBR pilot plant, an economic study and LCA analysis were carried out to evaluate the economic and environmental feasibility of the implementation of the hybrid MBBR-MBR on an industrial scale.

The state of the art of this thesis has been published as a review in Desalination and Water Treatment:

Yang, X., Crespi, M., & López-Grimau, V. (2018). A review on the present situation of wastewater treatment in textile industry with membrane bioreactor and moving bed biofilm reactor. Desalination and Water Treatment, 103, 315–322. https://doi.org/10.5004/dwt.2018.21962

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CHAPTER II. ASSUMPTIONS AND OBJECTIVES OF THE THESIS

According to many years of operation practice, although Conventional Activated Sludge (CAS) process in treating industrial wastewater is relatively mature, it still has many shortcomings and deficiencies, such as large aeration tank volume which results in high cost of infrastructure, low adaptability to changes of organic load, and poor efficiency in color removal in the case of textile wastewater. In view of the above factors, alternative treatments have been investigated for industrial wastewater treatment such as Moving Bed Biofilm Reactor (MBBR) and Membrane Bioreactor (MBR).

MBBR treatment makes up for many shortcomings of CAS method, such as its good stability, strong ability to withstand the impact of organic and hydraulic loads, high removal rate of organic matter, and small volume of the reactor. As the majority of biomass is fixed on the plastic carriers added, and the biomass concentration in suspension is maintained at values similar to activated sludge reactor. However, one of the main problems of the MBBR is the worsening of the decantation of the biomass in comparison to an activated sludge system. This often requires the addition of coagulants if a well clarified effluent is required.

MBR treatment has strong treatment capacity, high solid-liquid separation efficiency, good effluent water quality, small footprint. Compared to CAS, the sludge production of MBR is reduced, as longer sludge ages are achievable. Moreover, the sludge age and hydraulic retention time are independent. Among the main advantages of the MBR system over CAS treatment, one of them is the ability to work at much higher biomass than CAS, which results in lower volume biological reactors. However, we find that the reduction of reactor volume is limited because the biomass concentration in practice has a limit in practical applications to avoid affecting the oxygen transfer coefficient "α".

The application of MBBR and MBR technologies in textile wastewater presents an added limitation derived from the characteristics of this type of water. In the Chapter I, it has been shown that both technologies require the application of a tertiary treatment, especially to eliminate color, either by the addition of coagulant or other advanced treatments. Especially if the objective is to reuse the water, these tertiary treatments are essential.

An MBBR-MBR hybrid system can overcome both general and particular limitations for textile wastewater. MBBR-MBR can work at high organic loading rates because MBBR remove the majority of biodegradable contaminants and the particulate components can be separated by MBR. Furthermore, MBBR-MBR system will reduce the hydraulic retention time (HRT) and energy consumption comparing with MBR. Therefore, development of the application of MBBR-MBR should be attractive to industrial wastewater treatment as a reliable and effective method.

Therefore, based on these considerations, the general objective of this thesis is to design a hybrid MBBR-MBR system for treatment of textile wastewater and optimize its working conditions for a high treatment efficiency to achieve the reuse of treated water in new dyeing processes.

Based on this general objective, the specific objectives are as follows:

A. Make a comparative study of the treating efficiency of MBBR and MBR relative to CAS for textile wastewater.
    - Optimize the treating performance of MBBR and MBR to reduce the HRT compared to that commonly required by CAS, by monitoring the removal efficiency of COD, color and SS.
    - Use a small amount or avoid using decolorizing agent after the optimization of MBBR and MBR.
    - Evaluate the economic and environmental feasibility of the implementation MBR and MBBR technologies on an industrial scale plant and to be able to select the method of textile wastewater treatment with lower investment and operating costs, and lower environmental impact related to energy and materials consumption.
    - Reuse the treated water from the treatment selected in the above evaluation in new dyeing processes.

B. Study of the hybrid MBBR-MBR system for treatment of textile wastewater from a local textile industry.
    - Design and set up the hybrid MBBR-MBR system considering a correct movement of carriers and the installation of the membrane.
    - Optimize the hybrid MBBR-MBR system by monitoring the treating performance and removal efficiency of COD, color and SS to avoid the addition of decolorizing agent.

- Study of reuse of treated water by the hybrid MBBR-MBR system in new dyeing processes.
- Economic study and LCA analysis to evaluate the economic and environmental feasibility of the implementation of the hybrid MBBR-MBR on an industrial scale.

CHAPTER III. MATERIALS AND METHODS

As mentioned in the previous chapter of objectives, the first stage of the thesis is to make a comparative study of Conventional Activated Sludge (CAS), Moving Bed Biofilm Reactor (MBBR) and Membrane Bioreactor (MBR) in textile wastewater treatment. The next stage of the thesis is the study of the hybrid MBBR-MBR system. Descriptions of each pilot plants are presented below.

3.1. Characterization of pilot plants

3.1.1. CAS pilot plant

The pilot plant of CAS was designed by the Environmental Pollution Control laboratory of Institut d'Investigació Textile i Cooperació (INTEXTER), which is made by glass shown in Figure III-1. The components of the plant are described below:

- Aerated reactor, capacity of 5 L
- Decantation tank, capacity of 4 L
- Mammoth pump of recirculation
- Peristaltic pump of water inlet
- Air diffusers

Figure III-1 CAS pilot plant

3.1.2. MBBR pilot plant

The MBBR pilot plant is the same one of CAS, to which the carriers are added. The pilot plant is shown in Figure III-2.

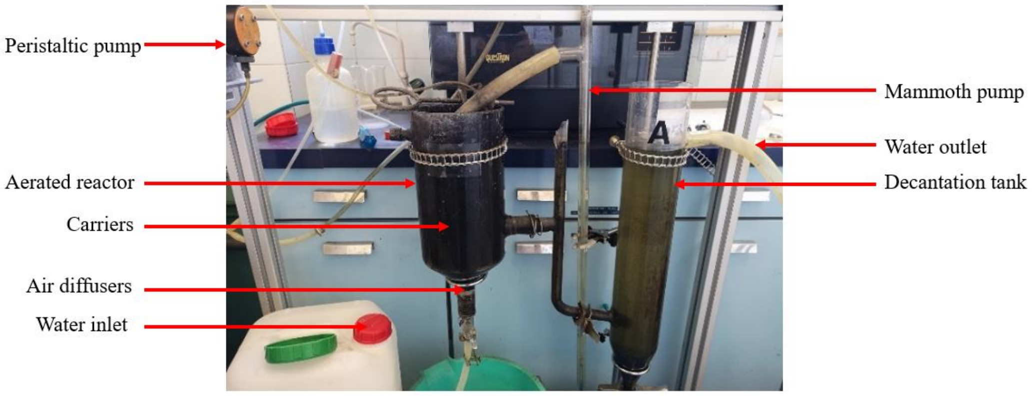

Figure III-2 MBBR pilot plant

BIOFILL C-2 plastic carriers (Figure III-3) were used in the MBBR study (BIO-FIL, Barcelona, Spain). The main specifications and operation characteristics of carriers are shown in Figure III-3 and Table III-1.

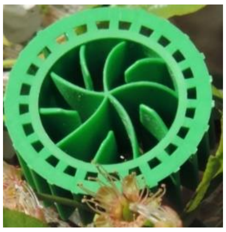

Figure III-3 BIOFILL C-2 plastic carriers (BIO-FIL, Barcelona, Spain)

Table III-1 BIOFILL type C-2 carriers characteristics

Specific Surface	590 m^2/m^3
Piece diameter	25 mm
Free volume	90%
Weight per piece	2.1 g
Density	< 1 kg/m^3

3.1.3. MBR pilot plant

The pilot plant of MBR used in this study was designed by the laboratory, composed of an aerobic reactor with a submerged ultrafiltration membrane. Figure III-4 shows the MBR pilot plant. The components of the plant are described below:

- Aerated reactor, capacity of 20 L
- Decantation tank, capacity of 4 L
- Centrifugal pump of water inlet
- Two peristaltic pumps of filtration and backwash
- Flowmeter
- Pressure gauge of the membrane
- Membrane

Figure III-4 MBR pilot plant

The influent was pumped directly from a raw wastewater tank, mixed completely with aeration in the reactor. There was an air inlet in the membrane module to prevent membrane fouling. The period of filtration and backwashing was set at 15 minutes and 30 seconds. The membrane of ultrafiltration de PVDF: ZeeWeed-1 (GE Power &Water, Canada) applied in the MBR pilot plant is shown in Figure III-5. The main characteristics of ZW-1 is described in Table III-2.

Figure III-5 ZW-1 membrane

Table III-2 characteristics of ZW-1

Model	ZW-1, submersible module
Configuration	Outside/in hollow fiber
Membrane surface	0.05 m^2
Pore size	0.04 µm
Maximum TMP	0.62 bar
Typical operating TMP	0.1-0.5 bar
Maximum TMP back wash	0.55 bar
Operating pH range	5-9

3.1.4. Hybrid MBBR-MBR system

The main objective of the thesis is the evaluation of the feasibility of a hybrid MBBR-MBR system for treatment of textile wastewater. An MBBR-MBR plant has been designed and built, shown in Figure III-6. Later in the Chapter V, all the information on the design and the components of the pilot plant is presented. The components are briefly described below:

- The hybrid reactor, capacity of 147 L (the MBBR tank of 79 L and the membrane tank of 31 L)
- Peristaltic pump of water inlet
- Permeate and backwash peristaltic pump
- Level sensor (controls the feed pump)
- DO sensor

- Thermostat
- Membrane
- Membrane pressure sensor

Figure III-6 Pilot plant of hybrid MBBR-MBR system

Biomass grew as suspended flocs and as a biofilm in the MBBR. The biomass which grew as a biofilm was developed on carriers which moved freely in the water volume by aeration. BIOFILL C-2 plastic carriers used in MBBR-MBR study are the same as previous MBBR study. Characteristics of carriers can be seen in Table III-1.

A MOTIMO BT01 hollow fiber flat plat membrane (MOTIMO Membrane Technology Co., Ltd., China) with a membrane filtration area of 1 m^2 was immersed into the membrane zone (Figure III-7). Details of the membrane are included in Table III-3.

Figure III-7 MOTIMO BT01 hollow fiber flat plat membrane

Table III-3 MOTIMO BT01 characteristics

Model	MOTIMO BT01
Configuration	hollow fiber flat plat membrane
Membrane surface	1 m^2
Pore size	0.03 µm
Maximum TMP	80 kPa
Operating TMP	10 - 60 kPa
Operating pH range	1 - 13

3.2. Analytical techniques

This section details the methodology followed for the effluent characterization and the equipment used in each determination are described.

3.2.1. Effluent characterization

The control of the performance of the pilot plant is carried out with the following analyses in the laboratory of Control of the Environmental Pollution of INTEXTER. The effluents will be characterized at the entrance, in the bioreactor and at the exit of various treatments in order to determine their efficiency. The parameters are determined following the Standard Methods 23rd edition [3.1]. The frequency of the analyses is shown in the Table III-4.

Table III-4 Frequency of the analyses

Parameters	**Influent**	**Biological Reactor**	**Effluent**
Weekly Frequency			
pH	3	3	3
Temperature	3	3	3
Conductivity	3	-	3
SS	3	3	3
Turbidity	3	-	3
Color	3	-	3
COD	3	-	3
BOD_5	1	-	1
TKN	1	-	1
P_{total}	1	-	1

The equipment and methods used in the determination of each parameter are listed in below.

- pH is determined by using a pH meter (CRISON GLP 21) following the method 4500-H^+B (Figure III-8).

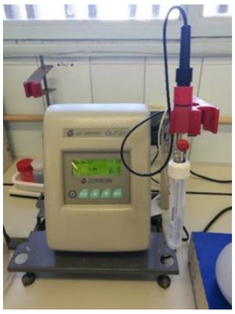

Figure III-8 pH meter (CRISON GLP 21)

- Temperature is determined by using a portable thermometer (Delta OHM HD 2107.1) following the method 2500 (Figure III-9).

c

Figure III-9 Thermometer (Delta OHM HD 2107.1)

- Conductivity is measured by using a conductivity meter (CRISON GLP 31) following the method 2510B. (Figure III-10)

Figure III-10 Conductivity meter (CRISON GLP31)

- Suspended Solid (SS) is analyzed following the method 2540D and the equipment is presented in Figure III-11.

Figure III-11 Equipment for the determination of SS

- Turbidity is determined by using a turbidity meter (La Motte 2020) following the method 2130B (Figure III-13).

Figure III-12 Turbidity meter (La Motte 2020)

- Color is evaluated by visual comparison of the sample with solutions of known concentrations of color following the method 2120B (Figure III-13).

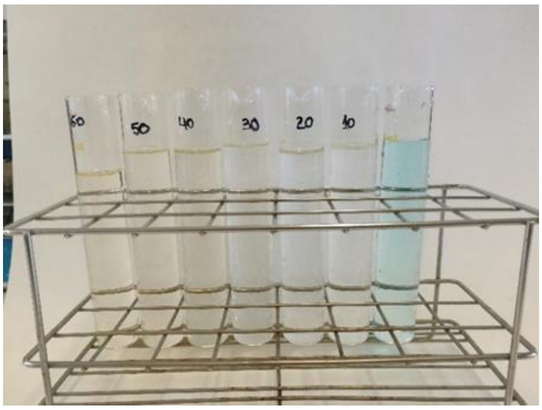

Figure III-13 Determination of color by visual comparison

- COD values are obtained by using the method 5220C (Valuation with Mohr’s salt).
- BOD_5 values are evaluated following the method 5210B (5-Day BOD Test).
- Nitrogen is determined analytically using the Kjendahl method (Metcalf y Eddy, 2003). The equipment used is a distillation unit (Büchi B-324), shown in Figure III-14a, and digester (Büchi 424), shown in Figure III-14b, following the methods 4500.

(a) (b)

Figure III-14 Distillation unit (Büchi B-324) and digester (Büchi K-424)

- Phosphorus is determined by using the method 4500-P E (Ascorbic Acid Method) with the spectrophotometer (SHIMADZU UV 2401PC). (Figure III-15)

Figure III-15 Spectrophotometer (SHIMADZU UV 2401PC).

- Microscopic observations of the sludge were performed with optical microscope (Nikon Eclipse 50i), see in Figure III-16, in order to evaluate the presence of specific microorganisms and its relationship with the condition of the sludge.

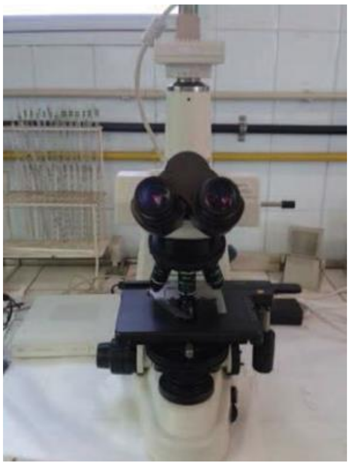

Figure III-16 Microscope (Nikon Eclipse 50i)

3.2.2. Dyeing tests using treated water

The objective of the dyeing tests is to evaluate the feasibility of reusing the treated water in new cotton dyeing processes.

The dyeing tests using treated water were performed with a laboratory Ti-Color dyeing machine (Prato, Italy) (Figure III-17a) equipped with 10 stainless steel tubular containers of 100 mL. The tests were performed under the following conditions according to a previous study [3.2]: 10 g cotton fabric, dye concentration of 3% o.w.f (overweight of fiber), liquor ratio 1:10 (1 g fiber/0.01 L dye bath). Three commercial reactive dyes supplied by Dystar were used in the water reuse study: Procion Yellow HEXL; Procion Crimson HEXL; Procion Navy HEXL. In addition to the required amount of dye, 60 g/L NaCl and 26 g/L Na_2CO_3 were added. The dyeing procedure is shown in (Figure III-17b). After the dyeing process, washing procedures were performed to remove the dye that was not fixed onto the fabrics. Nine washing steps with softened tap water were carried out with the following conditions:

1st–3rd: Cleaning with softened tap water at 50 °C for 10 min;

4th: Soap cleaning with 2 g/LCOTEMOLL TLTR at 95 °C for 15 min;

5th: Cleaning with softened tap water at 50 °C for 10 min;

6th: Soap cleaning with 2 g/L COTEMOLL TLTR at 95 °C for 15 min;

7th–9th: Cleaning with softened tap water at 50 °C for 10 min.

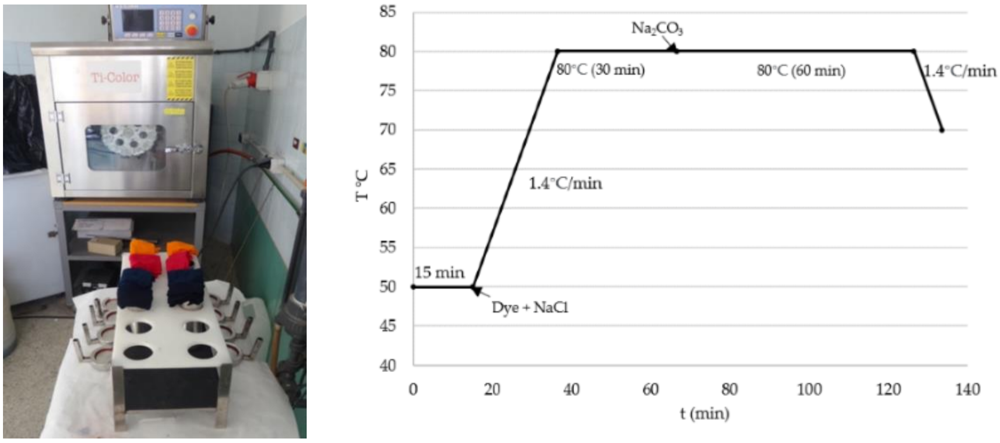

Figure III-17 (a) Ti-Color equipment (b) dyeing procedure

MINOLTA CM 3600d spectrophotometer were used to determine the color differences between dyes carried out in the different reuse studies of the permeate (Figure III-18).

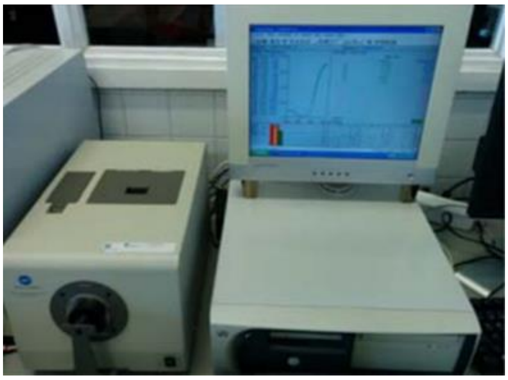

Figure III-18 MINOLTA CM 3600d spectrophotometer

The quality of dyed fabrics with reused water was determined according to Standard UNE-EN ISO 105-J03 by color differences with respect to reference dyeings performed with softened tap water [3.3].

Color differences (DE) were calculated with respect to standard dyeings carried out with decalcified tap water. The measurements of DE with respect to the standard dyeing were performed in triplicate, taking three points of the dyed fabric. Results which caused a coefficient of variation higher than 5% were discarded.

DE are determined by evaluation of the three cylindrical coordinates which describes color space: L* (luminosity), C* (chroma) and hº (hue angle from 0º to 360º). DL*, DC* and DH* are the differences of each parameter versus the reference dyeings [3.4]:

DL*> 0: lighter; DL*< 0: darker

DC*> 0: brighter; DC*< 0: duller

DH*, difference in hue

These values allow the calculation of color differences ($DE_{CMC(2:1)}$) as following equation:

$$DE_{CMC(2:1)} = [(DL^* /2SL)^2 + (DC^* /SC)^2 + (DH^* /SH)^2]^{1/2} \qquad \text{(III-1)}$$

where S_L, S_C and S_H were calculated from the chromatic coordinates corresponding to reference dyeings (L_R, C_R and h_R) as follows:

$S_L = 0.040975L_R/(1 + 0.01765L_R)$

If $L_R < 16$, $S_L = 0.511$

$S_C = [0.0638C_R/(1 + 0.0131C_R)] + 0.638$

$S_H= S_C(T_f+1-f)$

$f= \{(C_R)^4/[(C_R)^4+1900]\}^{1/2}$

$T= 0.36 + |0.4\cdot\cos(35+h_R)|$ if $h_R \geq 345°$ or $h_R \leq 164°$

$T= 0.56 + |0.2\cdot\cos(168+h_R)|$ if $164°< h_R < 345°$

$DE_{CMC(2:1}$ equation is simplified as follows:

$$DE_{CMC(2:1)} = [(DL_{cmc})^2+(DC_{cmc})^2+(DH_{cmc})^2]^{1/2} \tag{III-2}$$

In textile industry, one unit ($DE_{CMC\ (2:1)} \leq 1$) is the acceptance limit for color differences of quality control [3.5].

The color space can be also determined by rectangular coordinates: L*, a* and b*. Where a* indicates color tone from green (a*< 0) to red (a*> 0), and b* represents color tone from blue (b*< 0) to yellow (b*> 0). Color differences are calculated with these coordinates according to the equation DE_{CIELab}:

$$DE_{CIELab} = [(DL^*)^2+(Da^*)^2+(Db^*)^2]^{1/2} \tag{III-3}$$

Da* and Db* allows to identify tone changes with respect to the reference dyeings:

Da*> 0: redder; Da*< 0: greener

Db*> 0: yellower; Db*< 0: bluer

Color space in cylindrical (L*, C* and hº) and rectangular (L*, a* and b*) chromatic coordinates is presented in Figure III-19 [3.6]:

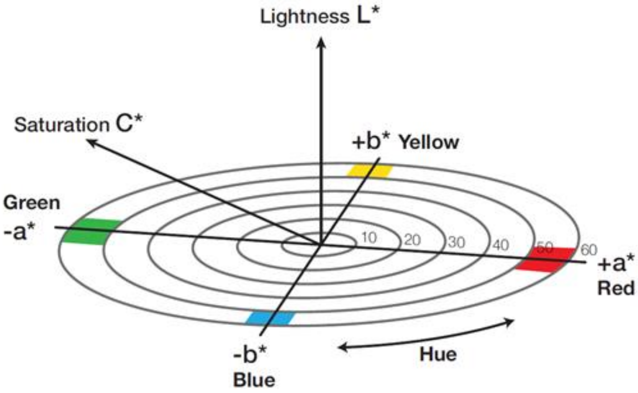

Figure III-19 Color space in cylindrical (L*, C* and hº) and rectangular (L*, a* and b*) chromatic coordinates. (Source: X-Rite)

In order to visually show the color differences of dyed fabrics with the treated water, photos of the dyed fabrics are shown. Because the photo may be affected by different lighting and camera device, color of the dyed fabrics is presented by a color convertor using color coordinates of CIELAB (L*, a* and b*). CIE here stands for International Commission on Illumination (International Commission on Illumination), which is an international authority on lighting, color, etc. CIELAB has a particularly good feature which is device independent. The color converter used is a free tool of NIX Color Sensor [3.7].

3.3. Economic assessment

The economic assessment, that accounts for both capital costs (CAPEX) and operating costs (CAPEX + OPEX), was performed by considering individual cost contributions to each treatment process (CAS, MBBR and MBR). CAPEX is the initial capital costs of construction and equipment and OPEX is the ongoing daily operating costs, such as energy consumption and equipment maintenance.

Additionally, the financial feasibility analysis was conducted by examining the Net Present Value (NPV) and the Internal Rate of Return (IRR). The calculation of NPV and IRR is taken into consideration of the investment payback period as 15 years. NPV is the sum of the present value of the net income obtained by folding the income and cost flow back to the beginning of the period. NPV is calculated using the following Equation III-4:

$$NPV = \sum_{t=1}^{n} \frac{s}{(1+i)^t} \qquad \text{III-4}$$

in which “s” is the profit or loss in the year (cash flow), “i” is the interest rate considered, “t” is the number of the year [3.8].

IRR is the interest rate when the cumulative NPV is zero. This IRR means the rate of the largest currency devaluation that the project can withstand. It is also calculated using the Equation III-4.

3.4. Environmental assessment

To compare the environmental impact of different treatment processes, Life Cycle Assessment (LCA) was performed according to ISO 14040 standards [3.9]. SimaPro 7.3.3 software was used for the LCA study, following the ReCiPe V1.06 midpoint (problem-oriented approach) and endpoint (damage-orientd approach).

The midpoint method is a characterization method that provides indicators for comparison of environmental interventions at a level of cause-effect chain between emissions/resource consumption [3.10] and the midpoint indicators can identify issues of specific environmental concern. The endpoint method is a characterization method that provides indicators at the level of Areas of Protection (natural environment's ecosystems, human health, resource availability) [3.10] and the endpoint indicators can be very helpful in decision support [3.11, 3.12].

The midpoint impact categories considered are:

- Climate change (CC)
- Ozone depletion (OD)
- Terrestrial acidification (TA)
- Freshwater eutrophication (FE)
- Marine eutrophication (ME)
- Human toxicity (HT)
- Photochemical oxidant formation (POF)
- Particulate matter formation (PMF)
- Terrestrial ecotoxicity (TET)
- Freshwater ecotoxicity (FET)
- Marine ecotoxicity (MET)
- Ionising radiation (IR)
- Agricultural land occupation (ALO)
- Urban land occupation (ULO)
- Natural land transformation (NLT)
- Water depletion (WD)
- Mineral resource depletion (MRD)
- Fossil depletion (FD)

The endpoint impact categories considered are:

- Damage to Human Health
- Damage to Ecosystems diversity
- Damage to Resource availability

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CHAPTER IV. TREATMENT OF TEXTILE WASTEWATER BY CAS, MBR AND MBBR: A COMPARATIVE STUDY FROM TECHNICAL, ECONOMIC AND ENVIRONMENTAL PERSPERCTIVES

This chapter shows the results of the treatment of textile wastewater by three treatment processes: Conventional Activated Sludge (CAS), Membrane Bioreactor (MBR) and Moving Bed Biofilm Reactor (MBBR), working under the same operating conditions. The objective of this chapter is to compare the treating efficiency of each treatment in COD, color and SS removal, and also to compare their economic and environmental feasibility.

4.1. Introduction

As one of the largest industries worldwide, the textile industry produces significant amounts of wastewater. Textile wastewater is generated in different steps during production, such as destarching, mercerization, dyeing, and washing [4.1], and is known to contain considerable amounts of organic compounds which provide color to the effluent [4.2]. In recent years, more strict regulations of effluent discharge have been applied in the textile industry, in order to reduce dye residues in the effluent before discharge into natural streams [4.3]. Consequently, finding suitable technologies to obtain an effective treatment of textile wastewater and to reuse its effluent in new production processes is essential for the industry's sustainable development.

One of the most applied biological methods in treating textile wastewater is the conventional activated sludge (CAS) process [4.4, 4.5]. The main objective of the CAS process is to remove organic compounds [4.6]. The CAS system has disadvantages such as high hydraulic retention time (HRT), problems with sludge settling, requirement of large space [4.7], and poor color removal efficiency due to the low biodegradability of dyes which can only be partially adsorbed on biomass [4.8–4.10]. Hence, a tertiary physicochemical method is usually required to give a better treatment performance [4.3, 4.11], which will increase the cost of the process.

In the past two decades, noticeable progress has been achieved with membrane bioreactor (MBR) technology in industrial wastewater treatment. MBRs separate the sludge by filtration, which differs from conventional CAS treatment [4.12]. MBRs can reduce land space and sludge production with a high biomass concentration in the reactor and are able to treat influent with wide fluctuations of quality [4.13–4.16]. In the case study of MBR applied in textile wastewater treatment in Bangladesh, the performance of the MBR system was better than that of the CAS [4.17]. Another study reported that high removal efficiencies were achieved for chemical oxygen demand (COD), color, and total suspended solids (TSS), and the cytotoxicity was significantly reduced by MBR when operating at an HRT of 2 days [4.18].

Recently, biofilm systems have drawn much attention in treating different types of industrial wastewater due to their several advantages compared with conventional biological treatment, including saving space [4.19, 4.20]. Among them, the moving bed biofilm reactor (MBBR) also has been applied in textile wastewater treatment in the last few years. One of the highlights of MBBR is a smaller volume of the biological plant or a larger treating capacity in the same reactor volume due to the biofilm being attached to carriers. Besides the great amount of biomass fixed on carriers, the concentration of biomass in suspension could be higher than that in the CAS process. In a previous study of textile effluent treatment [4.21], the pilot-scale plant of MBBR removed 86% of COD and 50% of color, respectively.

In addition to the selection of suitable wastewater treatment from a technical point of view, the increased demand for sustainability of industries has led to the use of life cycle assessment (LCA) as a tool to evaluate the feasibility of technologies [4.22]. Previous studies have estimated the environmental impacts generated by one or combined units of treatment plants for textile wastewater. Nakhate et al. evaluated the environmental footprints of a textile wastewater treatment plant and found out that consumption of electricity dominated in most of the environmental burden [4.23]. Cetinkaya and Bilgili compared, in another study, the environmental impacts caused by two desalination systems, and they found that using LCA could assess the environmentally friendlier treatment system for textile wastewater [4.24].

The aim of the current experimental study was to compare the efficiency of the CAS system, MBR process, and MBBR system in treating real textile wastewater. CAS is the current treatment process of the textile industry which provides the wastewater for our study. In order to improve the treating efficiency based on the existing CAS treatment, we have chosen MBBR and MBR to compare the technical, environmental, and economic feasibility. Parameters such as chemical oxygen demand (COD), total suspended solids (TSS), and color were determined to verify that MBR and MBBR have a better efficiency than CAS process. Special attention was paid to color removal, as color is one of the main problems in textile wastewater treatment.

Based on the experimental results in the pilot plant, an economic study and LCA were carried out to compare the economic and environmental feasibility of implementation of these technologies on an industrial scale and also to select the method of textile wastewater treatment with lower investment, operating costs, and environmental impact related to energy and materials consumption.

Water treated with the most viable method was reused to make new dyes because water reuse in the textile industry, a large water consumer, is one of the main factors to achieve sustainable development.

4.2. Methodology

4.2.1. Pilot Plant Description and Analysis

Three pilot plants were investigated for textile wastewater treatment in this study. The flow diagram of each plant is shown in Figure IV-1 and the images of three pilot plants have been demonstrated in Figure III-1, Figure III-2, Figure III-3 of CHAPTER III. Among them, the plant for the CAS process and the plant for MBR were operated in parallel. The pilot plant for MBBR was the same as for the CAS operation, but without the recirculation of sludge. The three treatments were operated with a controlled temperature of 25 °C. The textile wastewater was obtained from a local textile industry, Acabats del Bages, S.A. (Monistrol de Montserrat, Spain). The characteristics of the wastewater are shown in Table IV-1, including pH, COD, color, biochemical oxygen demand (BOD), TSS, total nitrogen (TN), and total phosphorous (TP). The duration of experiments for three pilot plants was 96 days.

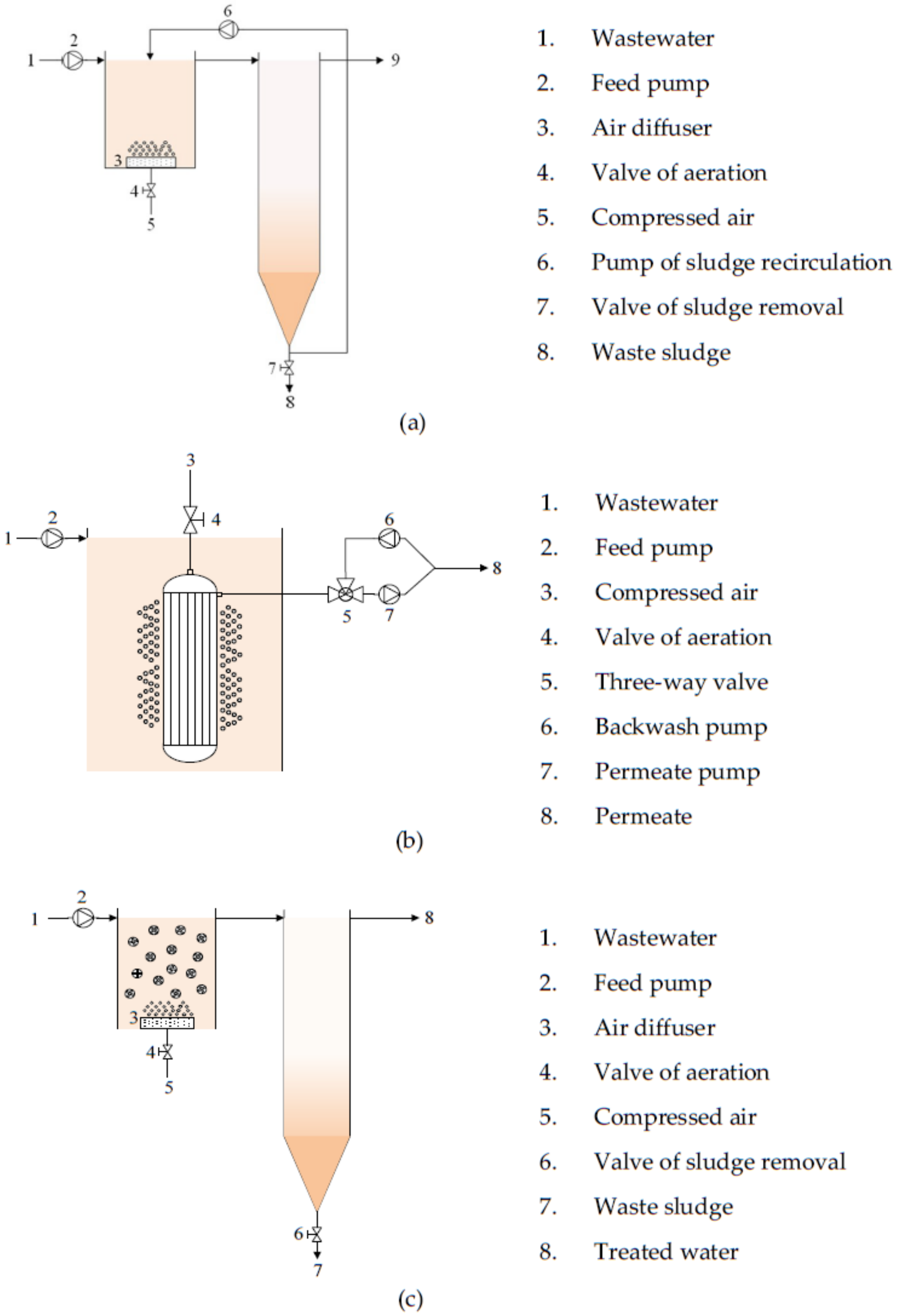


Figure IV-1 Flow diagrams of (**a**) Conventional Activated Sludge (CAS); (**b**) Membrane Bioreactor (MBR); (**c**) Moving Bed Biofilm Reactor (MBBR)

Table IV-1 Characteristics of textile wastewater

Parameters	Average
pH	8.6
COD mg/L	2000
Color Pt-co /L	700
BOD mg/L	400
TSS mg/L	940
TN mg/L	54
TP mg/L	11

It should be noted that the pH of wastewater returned to 8.6 in the reactor due to the buffering effect caused by the presence of carbonates, usual in textile wastewater. It was unnecessary and unattainable on an industrial scale to adjust the pH. Therefore, in the economic and LCA study, we did not take into account the amount of acid on the industrial scale.

The CAS pilot plant was composed of an aerobic reactor (volume 4 L) connected to a decantation tank. The flow rate in the CAS plant was 2 L/d, and the HRT was set to 2 days as the HRT of the current CAS plant of the textile industry.

The MBR used in this study was a pilot plant, composed of an aerobic reactor with a submerged ultrafiltration membrane. A Polyvinylidene fluoride (PVDF) hollow fiber membrane module ZeeWeed-1 (ZW-1) (GE Power & Water, Canada) was used. The membrane characteristics are shown in Table III-2 of Chapter III. The aerobic reactor had a working volume of 20 L. The influent was pumped directly from a raw wastewater tank, mixed completely with aeration in the reactor. There was an air inlet in the membrane module to prevent membrane fouling. The period of filtration and backwashing was set at 15 minutes and 30 seconds for the laboratory-scale reactor according to previous study with the membrane module [4.25].

As mentioned before, the MBBR pilot plant was the same one as in the CAS process. The aerobic reactor was filled with the carriers at a filling ratio of 30% (v/v). The plastic BIOFILL C-2 carriers used in this study were provided by BIO-FIL (Barcelona, Spain). The main specifications and operation characteristics of carriers are shown in Table III-1 of Chapter III. MBBR operation was inoculated with aerobic sludge collected from the wastewater treatment plant of the same textile industry. The start-up period lasted 3 weeks so biofilm could grow on the carriers.

In the initial phase, both MBR and MBBR were operated with 2 days of HRT, as was the CAS system. In order to assess a larger treating capacity and efficiency, the flow rate was increased gradually during the experiments. The flow rate in the MBR plant was fixed at 15 L/d and the HRT was 1.3 days, whereas the flow rate in the MBBR plant was 4 L/d and the HRT was fixed at 1 day. In the phase after the flow rates were stable, the concentration of dissolved oxygen (DO) in CAS was 2.1 mg/L, similar to the DO level in the MBBR reactor of 2.2 mg/L. MBR had a lower DO concentration of 1.8 mg/L.

4.2.2. Economic Analysis

The economic assessment of capital expenditures (CAPEX) and operational expenditures (OPEX) for three treatment schemes is determined in the results section.

4.2.3. Environmental Impact Analysis

To compare the environmental impact of three treatment processes, life cycle assessment (LCA) was performed according to standard ISO 14040 [4.26]. Simapro was used as the LCA software. The database used was Ecoinvent 3.1. ReCiPe, midpoint and endpoint approach, and Hierarchist perspective were considered as the methodology to calculate environmental impact. The selected functional unit was “1 m^3 of treated effluent”. The data used in this study were taken from the experimental results.

4.2.4. Dyeing Tests Using Treated Water

The dyeing tests using treated water were performed with a laboratory Ti-Color dyeing machine (Prato, Italy) (Figure III-17a) following the methods presented in Section 3.2.2 of CHAPTER III [4.27]. Three commercial reactive dyes supplied by Dystar were used in the water reuse study: Procion Yellow HEXL, Procion Crimson HEXL, and Procion Navy HEXL. The dyeing procedure is shown in Figure III-17b and nine washing steps are described in in Section 3.2.2 of CHAPTER III.

4.2.5. Analytical Methods

During this study, the control of the three pilot plants was carried out with analyses by characterizing the water at the entrance, in the bioreactor, and at the exit to determine the working efficiency. COD, TSS, TN, TP, color, pH, conductivity, and turbidity were determined following the Standard Methods 23rd edition [4.28].

The quality of dyed fabrics with reused water was determined according to Standard UNE-EN ISO 105-J03 by color differences with respect to reference dyeings performed with softened tap water [4.29]. Total color differences ($DE_{CMC(2:1)}$) were calculated from lightness (DL^*), chroma (DC^*), and Hue (DH^*) using Equation III-2 presented in CHAPTER III.

A spectrophotometer, MINOLTA CM 3600d (Osaka, Japan), was used for these measurements according to Standard illuminant D65/10°.

Generally, the color difference of one unit ($DE_{CMC\,(2:1)} \leq 1$) is the acceptable limit in the textile industry.

4.3. Results and Discussion

4.3.1. Treating Efficiency

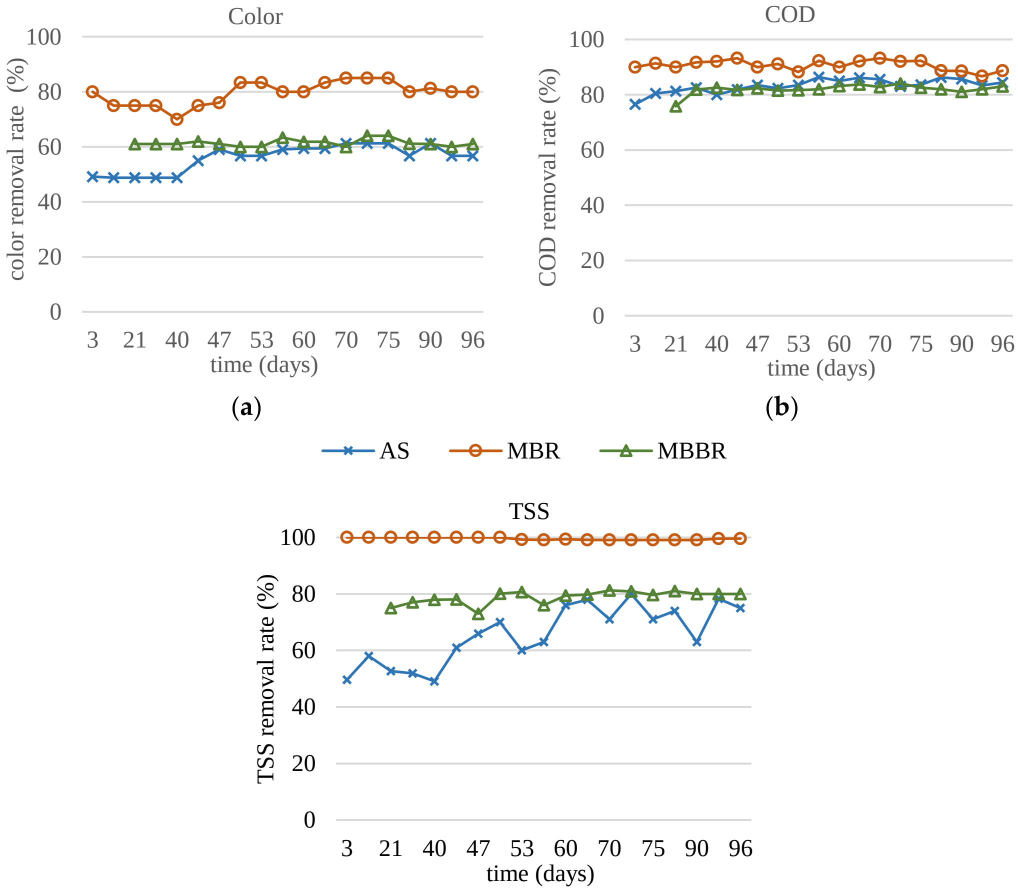
During the experiments, the average biomass concentrations in the reactor of CAS, MBR, and MBBR were 3 g/L, 2.3 g/L, and 3.5 g/L, respectively. As the textile wastewater had rather low contents of TN (54 mg/L) and TP (11 mg/L), over 90% removal of TN and TP was obtained after MBR and MBBR treatment, whereas CAS eliminated 88% of TN and TP.

As mentioned in Section 4.2.1, the initial HRT for CAS, MBR, and MBBR was 2 days, whereas the initial organic loading rate (OLR) was the same for the three treatments at 1 kg COD/ (m^3 d). The HRT of MBR and MBBR was gradually reduced to evaluate if the treating efficiency could be maintained while the treating capacity was increased.

Color in the influent varied between 400 and 1500 mg Pt-co/L. The removal rates of color obtained by the three treatment systems are shown in Figure IV-2a. The average color removal efficiency was 55% in the CAS process and was 80% in the MBR system, while in the MBBR system the color removal achieved 61%. MBR was significantly more efficient at removing color than the CAS process under the same operating conditions. MBBR had a higher color-removing performance than the CAS process, while the HRT (2 days) of CAS was twice the HRT (1 day) of MBBR. In order to meet discharge standards, decolorizing agent was added to the effluent from the CAS and MBBR processes. After adding 200 ppm of decolorizing agent, the color content reached the discharge standard in CAS, while the amount of decolorizing agent needed for MBBR was 100 ppm. In conventional biological treatment, the addition of various adsorbents and chemicals into the activated sludge system to improve the color removal efficiency is a common method, which will increase the cost and will generate secondary contaminates [4.30, 4.31].

COD of the influent remained at about 2000 mg/L. The average COD effluent of the CAS process was 350 mg/L, and the average efficiency of COD removal was 83%. The average COD value of the effluent from MBR was 170 mg/L, and the COD removal rate was 91%. The removal rates of COD in the three pilot plants are illustrated in Figure IV-2b. Furthermore, the CAS process worked with an HRT of 2 days, while the HRT of MBR worked only within 1.3 days. This demonstrated the efficiency and stability of the biological process of MBR. Similar results of COD removal in the MBR system and CAS process were also observed previously [4.17, 4.32], indicating that after MBR treatment, a better COD removal efficiency can be obtained from the conventional AS process. The average COD value of the effluent from MBBR was 179 mg/L, and the COD removal rate was 82%. Although the removal rates of COD of the CAS and MBBR processes were similar, HRT of MBBR was half of the HRT of the CAS process. The average OLRs for the CAS system, MBR, and MBBR were 1 kg COD/ (m^3 d), 1.5 kg COD/ (m^3 d), and 2 kg COD/ (m^3 d), respectively.

The TSS removal rates in the CAS system, MBR, and MBBRs are shown in Figure IV-2c. During the parallel experiments of CAS and MBR systems, the average value of TSS in the influent was 940 mg/L. The average TSS removal efficiency in the CAS process was 66%, while in MBR system the TSS removal achieved 99.6%. From the perspective of TSS removal, membrane filtration is an attractive method because of the total retention of suspended matter and significant retention of colloidal matter [4.33]. The results showed the advantage of the MBR process in TSS reduction with respect to the CAS process. The MBR process reached high TSS elimination without the necessity to add a tertiary treatment. MBBR achieved an average TSS removal rate of 78%, which was better than that of the CAS system.



(**a**) (**b**)

(**c**)

Figure IV-2 Removal rates of color (**a**), COD (**b**), and TSS (**c**) in activated sludge system, MBR, and MBBR

4.3.2. Microscopic observation of the sludge

In the study, sludge in three pilot plants was taken from the same textile industry. Due to this, there was almost no stage needed for sludge adaption. The sludge was in stable conditions during the operation.

The presence of ciliate protozoa in activated sludge is of great importance in the process, as they directly contribute to the clarification of the effluent through two activities, which are flocculation and predation, the latter being the most important [4.34]. Several studies have shown experimentally that the presence of ciliates in wastewater treatment plants improves the quality of the effluent. When there is a high number of ciliates, the effluent from the treatment plant has less turbidity and lower BOD [4.35–4.37]. The ciliates present in the mixed liquor can be classified into two broad categories according to their relationship with the floccules: ciliates associated with the floccules (pedunculates and crawlers) and ciliates not associated with the floccules (free – swimmers).

Free-swimming ciliates were observed in both pilot plants during the whole experimental, as It is shown in Figure IV-3.

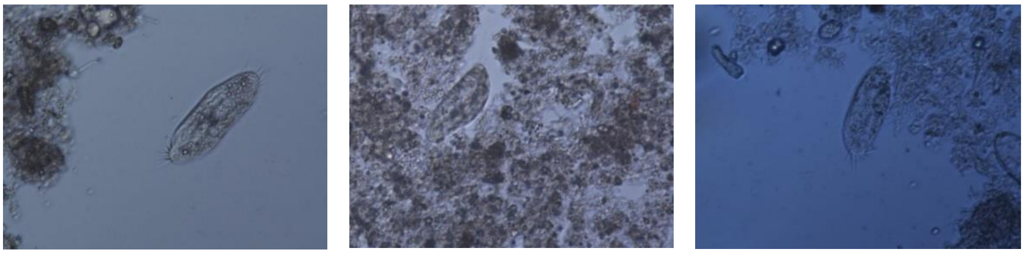

(a) (b) (c)

Figure IV-3. Free-swimming ciliates in (a) AS sludge; (b) MBR sludge; (c) MBBR sludge

Vorticella pedunculated ciliate can be found in media with a certain amount of organic matter and develops in activated sludge systems when its operation is stable indicating good performance and appropriate aeration. The longer the peduncle and the larger the crown, the better the level of purification. A great amount of Vorticella was found in the sludge of both reactors (see in Figure IV-4).

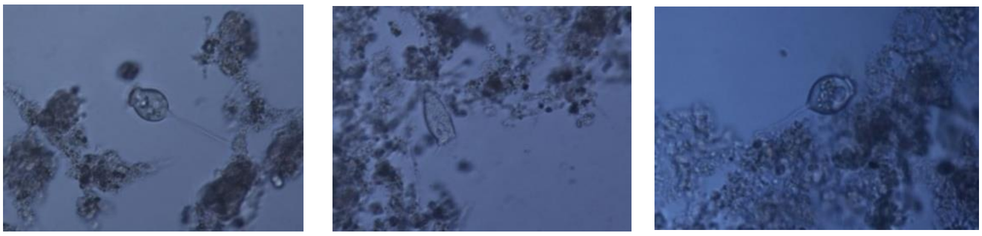
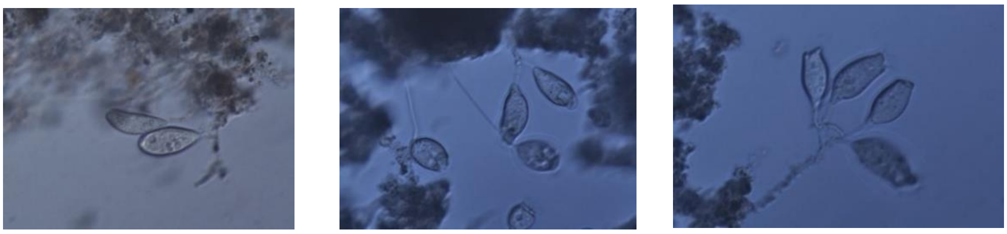

(a) (b) (c)

Figure IV-4 Pedunculated Ciliates (Vorticella) in (a) AS sludge; (b) MBR sludge; (c) MBBR sludge

Epistylis are very common in industrial wastewater treatment systems and play an important role in effluent clarification. In addition, Epistylis species of wastewater treatment plants can be used as performance bio-indicators of a great variety of parameters and processes [4.38, 4.39]. In all the systems, Epistylis were observed (see in Figure IV-5).



(a) (b) (c)

Figure IV-5 Pedunculated Ciliates (Epistylis) in (a) AS sludge; (b) MBR sludge; (c) MBBR sludge

Rotifers are multicellular organisms. They have different shapes and sizes and have a more complex structure than protozoa. The presence of rotifers in activated sludge means a stable sludge with plenty of oxygen in good situation. They contribute to the clarification of the effluent. Some species contribute to floc formation by mucus secretion [4.40]. Rotifers were found in MBBR sludge in the final period of the operation when the system was well stabilized (see in Figure IV-6).

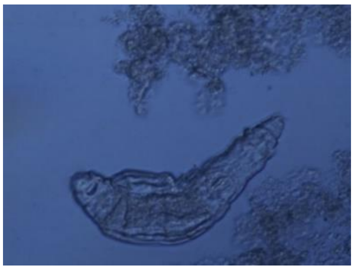


Figure IV-6 Rotifer in MBBR sludge

4.3.3. Economic Study of the Three Treatment Processes

The local textile industry produced 222,700 m^3 of wastewater per year with 11 months under operation. The wastewater treatment method of the industry is conventional activated sludge (CAS). The daily treatment flow is 920 m^3/d. The current HRT of the CAS plant is 2 days.

4.3.3.1. Capital Expenditures (CAPEX)

The CAPEX of the CAS system was considered to be the reference (0) in the economic study. The CAPEX of MBR and MBBR treatments were added directly to the CAPEX of the CAS system.

For the MBR full-scale system, the membrane and the installation of the membrane (366,153 €) have been considered for the CAPEX estimation according to the CAPEX calculation from a study of the cost of a small MBR (100–2500 m^3/d flow capacity) [4.41].

For the MBBR full-scale system, the cost of carrier medias (115,500 €) has been considered for the CAPEX estimation according to the suppliers' information.

4.3.3.2. Operational Expenditures (OPEX)

Consumption of energy, decolorizing agent data, and environmental tax of wastewater discharge and sludge management and were gathered in order to estimate operational expenditures (OPEX) of the three treatment plants.

Additionally, the cost of membrane replacement represented 2.4% of the energy cost [4.42], and the average lifetime of the UF membrane was taken as 10 years. The maintenance and repair costs represented 19.5% of the energy cost [4.42]. MBR could withstand higher concentrations of biomass with much longer sludge retention time (SRT) than in conventional AS, which allows much less sludge production in the MBR system and consequently, lowers the frequency of sludge disposal [4.13, 4.43]. During the experimental study of MBR, sludge concentration did not exceed the withstanding limit of the membrane. The sludge generation of MBR was estimated according the increasing rate of the biomass concentration and the concentration limit for the membrane.

The detailed OPEX calculation of each treatment plant is demonstrated in the following tables. (Table IV-2, Table IV-3, Table IV-4).

Table IV-2 CAS operational cost for treating 1 m^3 wastewater

Concept						**Total Price €/m^3**	**Reference**
(a) Consumption	**Unit**	**Amount**	**Unit**	**Unit price**	**Convert to €/m^3**	**0.55**	
Electricity	kW/m^3	0.96	€/kw	0.187	0.17952		[4.44]
Decolorizing agent	kg/m^3	0.2	€/kg	1.85	0.37		[4.45]
(b) Environmental tax	**Unit**	**Amount**	**Unit**	**Unit price**		**0.86**	
Sludge generation	kg/m^3	0.83	€/kg	0.158	0.013114		[4.46]
Wastewater discharge							[4.47]
OM[1]	kg/m^3	0.23	€/kg	1.0023	0.230529		
TSS	kg/m^3	0.32	€/kg	0.5011	0.160352		
N	kg/m^3	0.008	€/kg	0.761	0.006088		
P	kg/m^3	0.003	€/kg	1.5222	0.0045666		
Conductivity	S/cm	0.00598	€/Sm^3/cm	8.0198	0.0479584		
summation					0.449494		
ST[2] = 1.5 × SUM					0.67424101		
GT[3]					0.163		
Total price						**1.41**	

[1] OM: organic material; [2] ST: specific tax; [3] GT: general tax.

Table IV-3 MBR operational cost for treating 1 m^3 wastewater

Concept						**Total Price €/m^3**	**Reference**
(a) Consumption	**Unit**	**Amount**	**Unit**	**Unit price**	**Convert to €/m^3**	**0.51**	
Electricity	kW/m^3	2.72	€/kw	0.187	0.50864		[4.44]
Decolorizing agent	kg/m^3	0	€/kg	1.85	0		[4.45]
(b) Environmental tax	**Unit**	**Amount**	**Unit**	**Unit price**		**0.43**	
Sludge generation	kg/m^3	0. 023	€/kg	0.0158	0.0003634		[4.46]
Wastewater discharge							[4.47]
OM	kg/m^3	0.11	€/kg	1.0023	0.110253		
TSS	kg/m^3	0.04	€/kg	0.5011	0.020044		
N	kg/m^3	0.004	€/kg	0.761	0.003044		
P	kg/m^3	0.002	€/kg	1.5222	0.0030444		
Conductivity	S/cm	0.00533	€/Sm^3/cm	8.0198	0.04274553		
summation					0.17913093		
ST = 1.5 × SUM					0.2686964		
GT					0.163		
(c) Membrane replacement						**0.01**	[4.42]
(d) Maintenance and repair						**0.10**	[4.42]
Total price						**1.05**	

Table IV-4 MBBR operational cost for treating 1 m^3 wastewater

Concept						**Total Price €/m^3**	**Reference**
a) Consumption	**Unit**	**Amount**	**Unit**	**Unit price**	**Convert to €/m^3**	**0.27**	
Electricity	kW/m^3	0.48	€/kw	0.187	0.08976		[4.44]
Decolorizing agent	kg/m^3	0.1	€/kg	1.85	0.185		[4.45]
b) Environmental tax	**Unit**	**Amount**	**Unit**	**Unit price**		**0.78**	
Sludge generation	kg/m^3	0.29	€/kg	0.158	0.004582		[4.46]
Wastewater discharge							[4.47]
OM	kg/m^3	0.23	€/kg	1.0023	0.230529		
TSS	kg/m^3	0.24	€/kg	0.5011	0.120264		
N	kg/m^3	0.009	€/kg	0.761	0.006849		
P	kg/m^3	0.002	€/kg	1.5222	0.0030444		
Conductivity	S/cm	0.00595	€/Sm^3/cm	8.0198	0.04771781		
summation					0.40840421		
ST = 1.5 × SUM					0.61260632		
GT					0.163		
Total price						**1.05**	

In terms of the consumption part, MBR had the highest cost (0.51 €/m^3) of electricity consumption because it required more electricity to operate and to maintain the membrane filtration. However, CAS had the highest cost in the total consumption, with a value of 0.55 €/m^3, among the three treatments due to the larger amount of decolorizing agent used. This was not necessary for MBR because MBR achieved the color removal requirement and was used less in MBBR since MBBR had a better color removal performance. The reason that MBBR consumed half the electricity of the CAS system is that the HRT of MBBR was 1 day while in CAS it was 2 days, which means that MBBR with doubled treating capacity could save 50% of the electricity expense.

In regard to environmental tax, it can be observed that MBR had the lowest expense (0.43 €/m^3) since it had a better performance with organic compounds and TSS. MBBR, with half the HRT and more efficient treatment behavior, would pay a lower environmental tax (0.78 €/m^3) than the CAS system (0.85 €/m^3)

As mentioned in 4.3.3.1, the CAPEX for MBR was 366,153 €, and for MBBR it was 115,500 €, in order to improve the existing AS treatment of the studied textile industry. The only investment of MBBR in CAPEX is the carriers, and the maintenance of carriers is more convenient and economical than maintaining the membrane. Even though the OPEX of MBR and MBBR are at the same value, MBBR had the advantage of low energy consumption and competitive treatment performance. Taken together, the results of CAPEX and OPEX show that MBBR is a more attractive option for the textile industry economically.

4.3.4. LCA study results

The LCA study begins with the analysis of the inventory results of three treatments and then moves on to the environmental impact assessment.

4.3.4.1. Inventory results

The inventory results of each treating process are shown in Table IV-5. All data are related to the functional unit (1 m^3 treated water). The impact of sludge generation was not taken into account in Simapro software; therefore, the impact of sludge generation could not be quantified in the LCA study. Nevertheless, sludge generated in the three treatments was quantified and is presented in Table IV-5.

Table IV-5 Inventory analysis of three processes

	Amount						**Unit/FU**	**Ecoinvent Unit Process**
Processes Included in LCA	**AS**		**MBR**		**MBBR**			
	Input	**Output**	**Input**	**Output**	**Input**	**Output**		
COD	2	0.35	2	0.17	2	0.34	Kg	
TSS	0.94	0.32	0.94	0.04	0.94	0.24	Kg	
N	0.055	0.008	0.055	0.004	0.055	0.009	Kg	
P	0.008	0.003	0.008	0.002	0.008	0.002	Kg	
Color	700	315	700	140	700	267	g Pt-co	
Conductivity	6.46	5.98	6.46	5.33	6.46	5.95	mS/cm	
Wastewater	1	0.959	1	1	1	0.959	m^3	
Sludge		0.83		0.023		0.29	Kg	
decolorizing agent	0.2		0		0.1		Kg	DTPA, diethylenetriamine pentaacetic acid, at plant/RER U
Electricity	0.96		2.72		0.48		Kwh	Electricity, medium voltage, production ES, at grid/ES U

4.3.4.2. Environmental impact assessment

The environmental impact of each treatment process according to the LCA results using endpoint approach is discussed, and then the three studied treatments are compared with respect to their total environmental impact.

CAS system

The results of the environmental impact assessment are presented in points (mPt) so that different categories could be compared. Firstly, the impact of the CAS treatment process was evaluated. As shown in Table IV-6, the CAS process had the lowest impact on Ecosystems, while it had a major impact on Resources, followed by Human health.

Table IV-6 Environmental impact of CAS

	Human Health (mPt)	**Ecosystems (mPt)**	**Resources (mPt)**
Electricity (kWh/m^3)	22.8	1.9	31.8
Decolorizing agent (kg/m^3)	34.4	3.4	81.2
TOTAL	57.2	5.3	113.0

The decolorizing agent represents 60%–70% of the environmental impact of the CAS system, having the most significant impact for all the categories.

The impact of the decolorizing agent on the detailed categories with relation to Human health, Ecosystem, and Resources is shown in Figure IV-7. The decolorizing agent had an impact on Human health mainly because of the effect on Climate change human health as well as Particulate matter formation categories, while Terrestrial ecotoxicity and Climate change ecosystems categories had major impacts on Ecosystems. Apart from that, the Fossil depletion category had the major responsibility for impacting Resources, while the Metal depletion category had almost no impact.

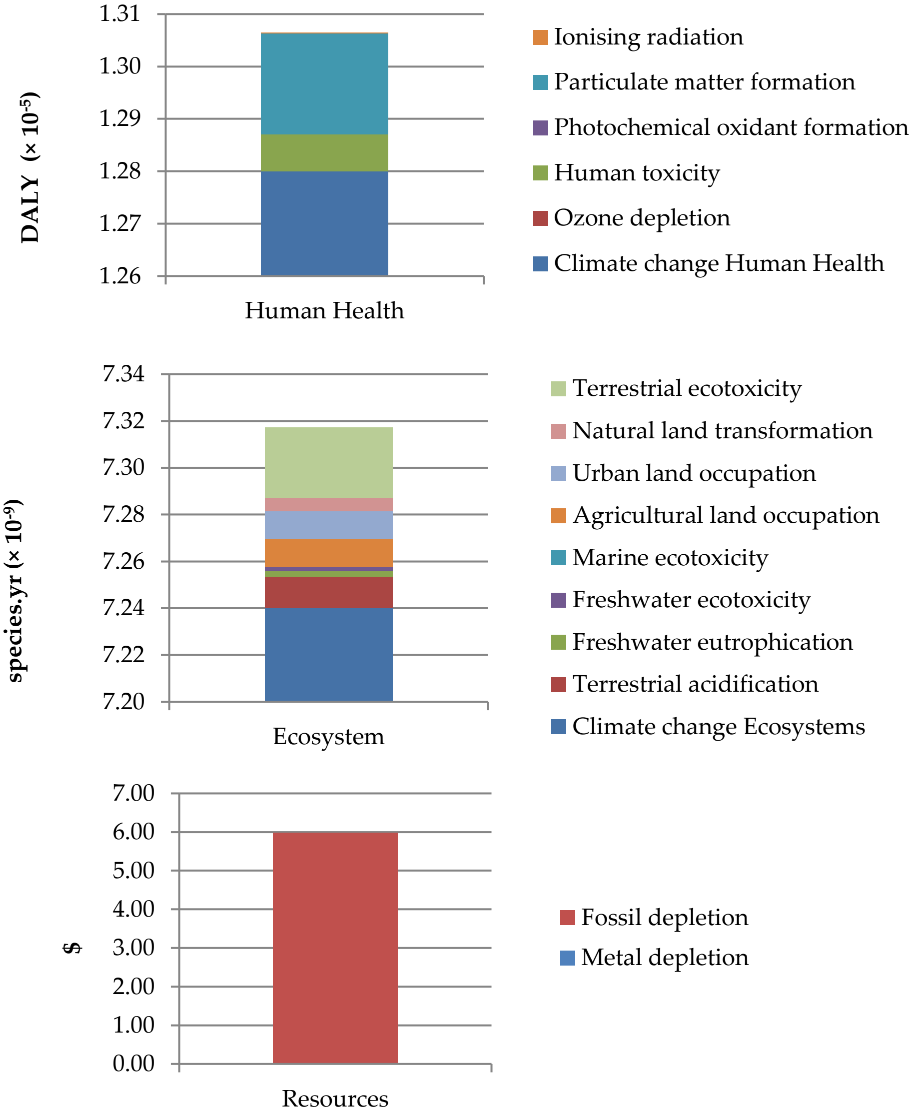


Figure IV-7 Analysis of the effect of the decolorizing agent on the impacted CAS categories

MBR Treatment

In the MBR treatment, as can be seen in Table IV-7, there was no consumption of decolorizing agent since the system removed most of the color in the effluents. The consumption of electricity during treatment represents the total environmental impact. The results show that the impact on Ecosystem was much lower, while the major impacts were on Resources and Human Health.

Table IV-7 Environmental impact of MBR

	Human Health (mPt)	**Ecosystems (mPt)**	**Resources (mPt)**
Electricity (kWh/m^3)	64.6	5.4	90.1
Decolorizing agent(kg/m^3)	0	0	0
TOTAL	64.6	5.4	90.1

Figure IV-8 shows the impact of electricity consumption for MBR treatment on the detailed categories related to Human Health, Ecosystem, and Resources. Climate change human health and Particulate matter formation categories were the main factors that had an impact on Human health of electricity consumption. In the meantime, the impact on Ecosystems mainly was due to Agricultural land occupation and Climate change ecosystem, while Terrestrial ecotoxicity, Natural land transformation, Urban land occupation, and Terrestrial acidification had minor impacts on the Ecosystem category. Furthermore, the major impact on Resources came from Fossil depletion category, while the Metal depletion category had almost no impact.

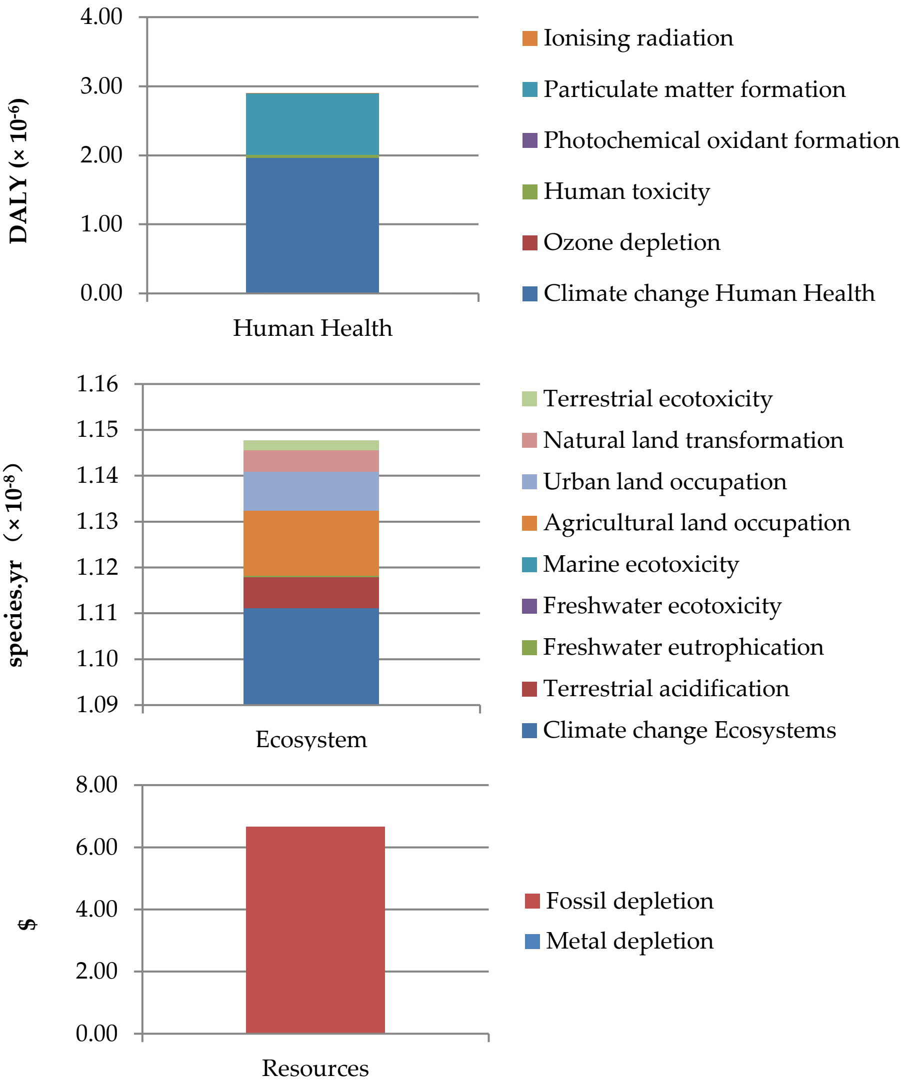


Figure IV-8 Analysis of the effect of electricity consumption on the impacted MBR categories

MBBR Treatment

As shown in Table IV-8, MBBR treatment, like AS and MBR treatments, also had a major impact on Resources, while the impact on Ecosystem was the lowest. The environmental impact of the consumption of decolorizing agent was mainly presented in Resources.

Table IV-8 Environmental impact of MBBR

	Human Health (mPt)	**Ecosystems (mPt)**	**Resources (mPt)**
Electricity (kWh/m^3)	11.4	0.9	15.9
Decolorizing agent(kg/m^3)	17.2	1.7	40.6
TOTAL	28.6	2.6	56.5

Comparison of the Three Treatments

The environmental impacts of three treatments are compared in Table IV-9 to evaluate which treatment had lower environmental impacts.

As shown, the MBBR system had the lowest impact on all three categories. Although decolorizing agent was used in the final step of AS and MBBR to obtain a well-clarified effluent and due to the filtration, decolorizing agent was not needed for MBR, the consumption of electricity had more significant environmental impacts on Human Health and Ecosystems.

Table IV-9 Environmental impacts of the three processes

	Human Health (mPt)	**Ecosystems (mPt)**	**Resources (mPt)**
AS	57.2	5.3	113.0
MBR	64.6	5.4	90.1
MBBR	28.6	2.6	56.5

In addition to the endpoint methods, which are helpful for decision-making because results can be compared in points, midpoint analysis was also performed to help identify issues of specific environmental concerns [4.48]. The results of midpoint assessment are shown in Table IV-10. It can be observed clearly that MBBR was environmentally advantageous since most of its impacts were the lowest in most of the categories, except in Climate change Human Health, Marine eutrophication, and Freshwater ecotoxicity, which were the three impact categories associated with the use of decolorizing agent. In MBBR operation, impacts on several categories could be reduced more than 70% more than those generated in MBR, and these categories were Particulate matter formation, Terrestrial acidification, Agricultural land occupation, Urban land occupation, Natural land transformation, and Urban land occupation. The CAS system had high environmental impacts, especially on Climate change Human Health, Marine eutrophication, and Freshwater ecotoxicity, due to the amount of decolorizing agent used in the treatment.

Table IV-10 Comparison of three processes: midpoint analysis

Impact Category	**Unit**	**CAS**	**MBR**	**MBBR**
Climate change Human Health	kg CO_2-eq	1.29	0.19	0.65
Ozone depletion	kg CFC-11 eq	1.39×10^{-7}	7.03×10^{-8}	6.94×10^{-8}
Human toxicity	kg 1.4-DB eq	0.12	6.10×10^{-2}	6.06×10^{-2}
Photochemical oxidant formation	kg NMVOC	3.89×10^{-3}	5.58×10^{-3}	1.95×10^{-3}
Particulate matter formation	kg PM10 eq	1.95×10^{-3}	3.41×10^{-3}	9.73×10^{-4}
Ionising radiation	kg U235 eq	0.16	0.26	0.08
Terrestrial acidification	kg SO_2-eq	6.46×10^{-3}	1.17×10^{-2}	3.23×10^{-3}
Freshwater eutrophication	kg P-eq	7.84×10^{-5}	6.77×10^{-5}	3.92×10^{-5}
Marine eutrophication	kg N-eq	2.24×10^{-3}	4.27×10^{-4}	1.12×10^{-3}
Terrestrial ecotoxicity	kg 1.4- DB eq	2.96×10^{-4}	1.69×10^{-4}	1.48×10^{-4}
Freshwater ecotoxicity	kg 1.4- DB eq	7.40×10^{-3}	2.48×10^{-4}	3.70×10^{-3}
Marine ecotoxicity	kg 1.4- DB eq	1.04×10^{-3}	5.19×10^{-4}	5.18×10^{-4}
Agricultural land occupation	m^2 year	5.51×10^{-3}	1.27×10^{-2}	2.75×10^{-3}
Urban land occupation	m^2 year	2.16×10^{-3}	4.34×10^{-3}	1.08×10^{-3}
Natural land transformation	m^2 year	1.25×10^{-5}	2.67×10^{-5}	6.27×10^{-6}
Water depletion	m^3	1.12×10^{-2}	8.10×10^{-3}	5.60×10^{-3}
Metal depletion	kg 1Fe eq	2.02×10^{-3}	2.08×10^{-3}	1.01×10^{-3}
Fossil depletion	kg oil eq	5.17×10^{-7}	4.14×10^{-7}	2.59×10^{-7}

4.4. Reuse of the Treated Effluent

Considering the previous results of economic and LCA analyses, MBBR treatment was selected as the most feasible method to be applied at industrial scale. At this point, the possibility of reusing the treated wastewater in a new dyeing processes was determined. MBBR was selected to check if the removal results of COD, SST, and color were sufficient to make new dyes without their quality being affected by the presence of organic matter residues, suspended solids, and residual dyes.

The treated water from the MBBR process was reused for a new dyeing process. Three reactive dyes—Procion Yellow HEXL, Procion Crimson HEXL, and Procion Navy HEXL—were used in the water reuse study. The color differences with respect to a reference dyeing are shown in Table IV-11. $DE_{CMC(2:1)}$ values of all three dyes were lower than 1, which is the acceptable limit for the textile industry. The results imply the feasibility of MBBR treatment to obtain a water reuse proportion up to 100% in the new dye baths. It should be considered that in practical textile production, there is 30% water loss due to evaporation or water fixed into the textile products. Therefore, the wastewater generated accounts for 70% of freshwater consumed by the industry. Although all the treated water was reused, it is not equal to 100% of the total water consumed by the industry. If we wanted to reuse all treated water, this would be 70% of the water consumed.

Table IV-11 Color differences between fabrics dyed with the treated effluent and a reference dyeing

Reactive Dyes	**100% Effluent Reused**
	$\mathbf{DE_{CMC(2:1)}}$
Procion Yellow HEXL	0.55 ± 0.08
Procion Crimson HEXL	0.76 ± 0.07
Procion Navy HEXL	0.42 ± 0.01

A comparison of the cotton fabrics dyed with the three dyes studied is shown inFigure IV-9 Comparison of cotton fabrics made with the three dyes studied Figure IV-9.

Figure IV-9 Comparison of cotton fabrics made with the three dyes studied

In order to present the color differences more precisely, color converted with CIELAB of each fabric is shown in Figure IV-10. The example of the color convertor of Procion Crimson HEXL is demonstrated in Figure IV-11.

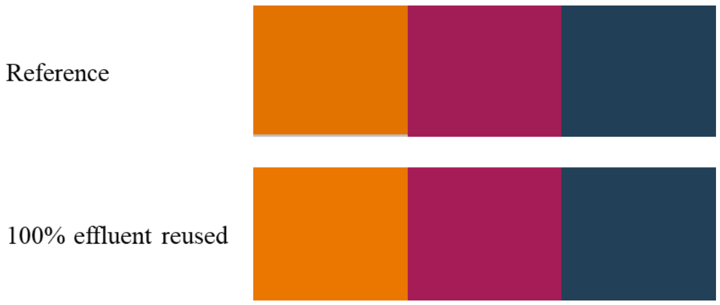
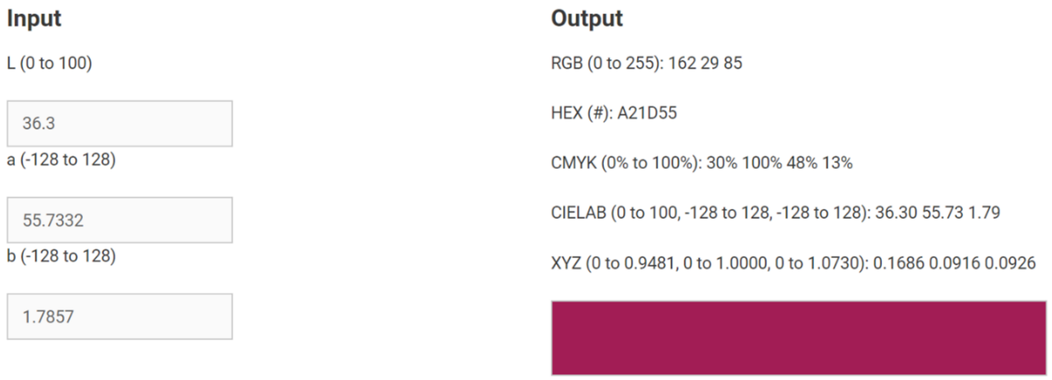


Figure IV-10 Comparison of color of cotton fabrics using CIELAB color space.



Input

L (0 to 100)

36.3

a (-128 to 128)

55.7332

b (-128 to 128)

1.7857

Output

RGB (0 to 255): 162 29 85

HEX (#): A21D55

CMYK (0% to 100%): 30% 100% 48% 13%

CIELAB (0 to 100, -128 to 128, -128 to 128): 36.30 55.73 1.79

XYZ (0 to 0.9481, 0 to 1.0000, 0 to 1.0730): 0.1686 0.0916 0.0926

Figure IV-11 Example of color converting process of Procion Crimson HEXL

4.5. Conclusions

After carrying out the comparative study in three pilot plants with CAS, MBR, and MBBR technologies, MBBR showed that it was a better alternative than CAS, with a comparable COD removal rate to CAS and a more efficient color reduction, while the treating capacity was doubled. Although the MBR was the most efficient technology for organic compounds and color removal, the economic and LCA study suggested that MBBR is a more attractive option for textile wastewater treatment at an industrial-scale plant. MBBR had the same OPEX as MBR, both lower than that of the CAS system, but MBBR had lower investment costs and lower CAPEX, which was 68% less than the CAPEX of MBR. MBBR also largely reduced the environmental impacts on different categories with respect to CAS and MBR processes in general. MBBR reduces the environmental impact as compared with the AS, since it reduced the consumption of electricity and decolorizing agent with respect to AS. MBR had a higher electrical consumption but avoided the consumption of decolorizing agent.

Finally, new dyes made with treated water from MBBR met the quality standard for the textile industry ($DE_{CMC(2:1)} \leq 1$). The presence of organic matter residues, suspended solids, and residual dyes in the effluent of MBBR did not affect the dyeing quality. Reuse of wastewater up to 100% is very promising in the textile industry as it is a considerable water-consuming industry worldwide.

According to the results obtained in this study, an alternative method of textile wastewater treatment for the future could be the combination of MBBR and MBR. The hybrid system MBBR-MBR is able to work efficiently at high organic loading rates with a low HRT, obtaining good performance in COD, SST and color removal, which will reduce space and energy consumption and also avoiding the application of tertiary treatments.

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CHAPTER V. DESIGN OF THE HYBRID SYSTEM: MBBR-MBR

In this chapter, the design motivation, the design considerations, and components description of the hybrid system, Moving Bed Biofilm Reactor – Membrane Bioreactor (MBBR-MBR), is presented.

This study is co-funded by ACCIÓ (Generalitat de Catalunya) within the REGIREU Project (COMRDI16-1-0062).

5.1. Motivation of design a hybrid MBBR-MBR system

As mentioned in the Chapter II, the main advantage of Membrane Bioreactor (MBR) system is that it can treat wastewater with high biomass concentration, which is higher than the limit of Conventional Activated Sludge (CAS) process. Therefore, the volume of the bioreactor can be reduced relatively, but to a limited extent, because in practice the concentration of biomass is limited due to its impact on the oxygen transfer rate which will affect the membrane performance. On the other hand, Moving Bed Biofilm Reactor (MBBR) is able to work with larger organic loads thanks to the biofilm attached on its carriers. However, one of the main problems of MBBR is that sludge decantation is not efficient that in most cases, a certain amount of coagulant is needed to meet the effluent standard.

The comparative study on CAS, MBR and MBBR treating textile wastewater in Chapter IV also demonstrated the above conclusions. MBBR plant showed that it had a better treating performance with doubled treating capacity than CAS in COD removal and color reduction. The economic and LCA study added to the evidence that MBBR is a more attractive option for textile wastewater treatment at an industrial-scale plant. The deficiency of MBBR is that the effluent needed to be decolorized using decolorizing agent to meet the discharge standard which increased the environmental impact. MBR plant had a great efficiency in the treatment of textile wastewater on removing organic compounds and suspended solids, furthermore, the treated water from MBR met the reusing criteria in new dyeing processes avoided the consumption of decolorizing agent. The main disadvantage of MBR is the relatively higher energy consumption.

Combining the theoretical and practical conclusions, a hybrid MBBR-MBR system can solve the decantation problem of MBBR in the treatment of wastewater by the filtration of MBR, and at the same time take advantage of that MBBR can treat high-concentration biomass. Additionally, biofilm attached on carriers of MBBR could maintain the oxygen transfer coefficient within a high range, thereby reducing the energy consumed by biological aeration.

5.2. Design considerations of the hybrid MBBR-MBR system

The main consideration in the hybrid MBBR-MBR design is to put the two technologies, MBBR and MBR in one reactor. The reactor is divided into two part for each process. Specific considerations for the reactor are described as follows:

- Firstly, the length and height of the reactor are determined by the size of the membrane and the installation of the membrane. As mentioned in Section 3.1.4 of Chapter III, A MOTIMO BT01 hollow fiber flat plat membrane (**错误!未找到引用源。**) with a membrane filtration area of 1 m^2 is used for the MBR process. The length and height of the membrane are 400 mm and 600 mm, respectively.
- The width of the reactor is set in order to maintain a correct movement of MBBR carriers by aeration.
- The MBBR part and MBR part are separated partially to prevent the movement of carriers from damaging the membrane.
- The MBBR part and MBR part are connected by a gap of 1 cm between the bottom of the wall to the bottom of the reactor (shown in Figure V-1), which allows the water flow to pass smoothly while preventing the passage of carriers with a diameter of 10 mm.

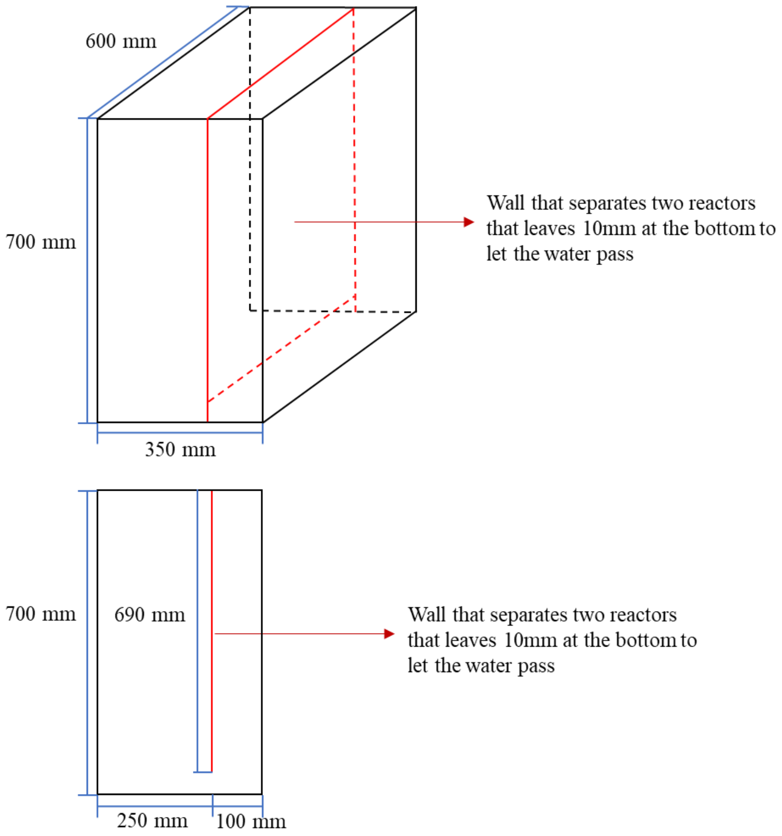


Figure V-1 Dimension description of the MBBR-MBR reactor and its reactor and side section

5.3. Description of the components of the hybrid MBBR-MBR system

The vessel of the pilot plant of the hybrid MBBR-MBR system was fabricated by the provider of the carriers BIO-FIL according to the design information. The reactor is shown in Figure III-6 of Chapter III. The total volume of the hybrid MBBR-MBR reactor was 147 L, and the effective volume was 110 L. The reactor was divided into two parts: the MBBR part (79 L) and the membrane tank (31 L).

The Figure V-2 presents the components of the pilot plant. Furthermore, each of the components and their function within the general process are described.

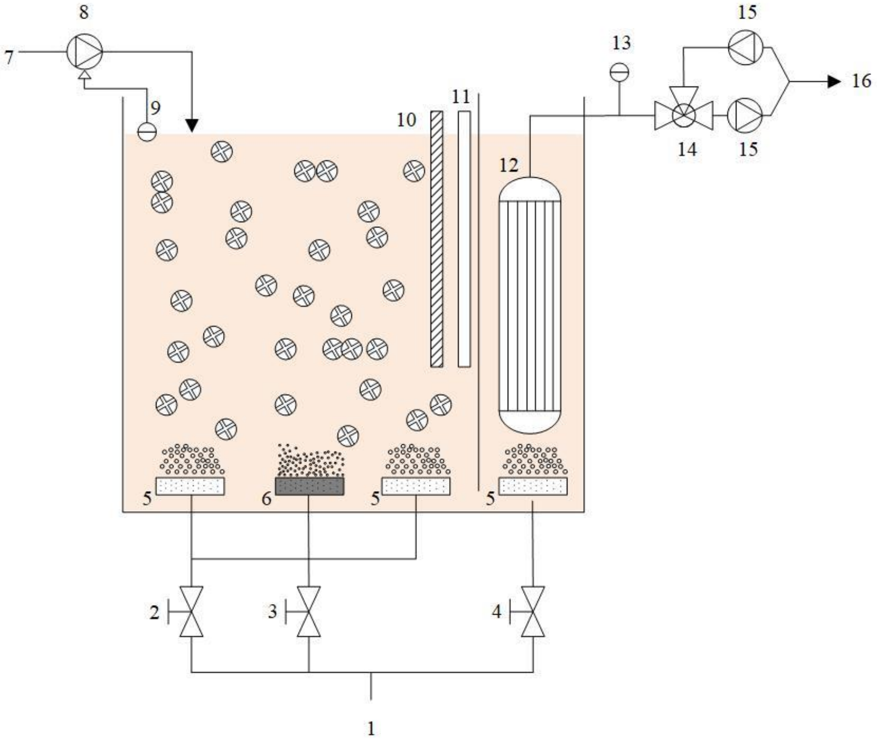


Figure V-2 Diagram of the hybrid MBBR-MBR pilot plant

1. Compressed air:

   Compressed air is supplied by the compressed air supply system of the laboratory.
2. Needle valve:

   It controls the air flow of thick bubble air diffusers for the circulation movement of carriers.
3. Valve controlled by DO (Labview):

   When the DO measured by the DO sensor is below 2 mg/L, the valve will open to let air in through fine bubble diffuser to raise DO. When the DO exceeds 4 mg/L, the valve will close to save energy.
4. Time controlled valve (Labview):

   It controls the thick bubble air diffuser for membrane scouring.
5. Thick bubble air diffuser (for mixing carriers and for air scouring the membrane):

   It provides power for the circulation movement of the carriers in the MBBR part.

   It keeps the membrane from fouling by air scouring.

   It should be noted that thick bubbles do not affect the DO concentration.
6. Fine bubble diffuser：

   For oxygen diffusion to adjust the DO concentration in the reactor.

7. Wastewater inlet

   The wastewater from a local textile industry is transported and stored regularly in a bulk container of 1 m^3.
8. Feed pump:

   ProMinent Dosing Pump Beta® BT4a for the wastewater inlet
9. Level sensor:

   Controls the feed pump to adjust the flowrate.
10. DO sensor:

    It monitors the stability of oxygen concentration in the reactor.
11. Thermostat:

    It keeps the temperature in the reactor stable at 25 °C.
12. Membrane:

    MOTIMO BT01 hollow fiber flat plat membrane with 1 m^2 of filtration area is placed.
13. Membrane pressure sensor:

    It monitors the membrane pressure during filtration to observe if membrane fouling occurs.
14. Three-way valve

    For the filtration and the backwash of the membrane.
15. Permeate and backwash pump controlled by Labview:

    Gilson MINIPULS 3 Peristaltic Pumps which allows the filtration suction and backwash according to the settled filtration and backwash period.
16. Permeate (treated water)

    The backwash water is taken from the permeate.

5.4. Description of the operation of the hybrid MBBR-MBR system

Raw wastewater is stored in a 1000L tank before the treatment. The wastewater enters the MBBR part through the inlet pump and is fully mixed with the carrier under the action of aeration mixing. There is a level sensor connected to the inlet pump to maintain the flowrate. Then the water enters the MBR reactor through the 1 cm gap below the middle partition of the reactor as shown in Figure V-1. Through membrane filtration, the treated water is sucked out by the outlet pump. The treated water, also called as permeate, is also used for backwashing of the membrane.

The central control system is the program of Labview and the computer with Labview is connected to the reactor to control the DO, temperature, time of filtration and backwash of the membrane.

- The DO sensor is connected with Labview to maintain the stability of oxygen concentration in the reactor by controlling the fine bubble valve. When the DO below 2 mg/L, the valve will open to let air in through fine bubble diffuser to raise DO. When the DO exceeds 4 mg/L, the valve will close to save energy.
- The Thermostat is connected with Labview to keep the temperature in the reactor stable at 25 °C.
- The permeate and backwash pump is connected with Labview to control the cycle time of filtration and backwash.

After setting up the parameters in Labview for the reactor operation, the treatment can run by itself with periodical inspections.

CHAPTER VI. STUDY OF A HYBRID SYSTEM: MOVING BED BIOREACTOR -MEMBRANE BIOREACTOR (MBBR-MBR) IN THE TREATMENT AND REUSE OF TEXTILES INDUSTRIAL EFFLUENTS

In this chapter, the pilot-plant of Moving Bed Biofilm Reactor coupled with Membrane Bioreactor (MBBR-MBR), which was designed in the Chapter V, was studied for the wastewater treatment of textile industry. Furthermore, the reuse feasibility of treated water was investigated.

This study is co-funded by ACCIÓ (Generalitat de Catalunya) within the REGIREU Project (COMRDI16-1-0062).

6.1. Introduction

Sustainable development has become an increasingly important factor for many industries. Among numerous pollution problems a textile industry may face, wastewater treatment has always been an essential part. In the fact of being one of the major water intensive industries (typically 200 - 400 L water consumed per Kg finished fabrics produced) [6.1], textile sector consequentially generates a considerable amount of wastewater of which characteristics highly depend on the applied dyes, auxiliary chemicals and the process itself [6.2]. The challenge for textile industry is to find effective and suitable wastewater treatments due to the increasing demand for strict effluent discharges [6.3], and to enable water reuse from an economic and environmental standpoint [6.4]. There is not a universal quality requirement of water reuse for the textile industry, because of the complexity during the production, such as the distinct demands of different fibers (natural fibers, synthetic fibers, etc.), diversity of the textile processes and the different requirements for the final products. Among all the processes, dyeing is the most delicate and complicated process in which to reuse water. Because the remains of organic matter, suspended solids and the presence of residual dye can harm the fixation of the new reactive dye and obtain tinctures with less color intensity and less saturation (both caused by lower dye exhaustion) or with changes of hue mainly caused by the residual color of water [6.5].

In the wide range of treating methods for textile wastewater, Membrane Bioreactor (MBR) has been increasingly put into use for industrial wastewater treatment including textile wastewater.

MBR is a biological treatment based on CAS process combined with a microfiltration or ultrafiltration membrane separation [6.6, 6.7]. This technology has a number of advantages associated to the filtration process, such as less space required, higher removal of organic pollutants than conventional biological process and better sludge performance [6.8, 6.9]. Previous studies have shown cases of textile wastewater treatment with MBR. Most of the studies showed that the removal rate of COD could reach 80-90%, however, MBR could not remove the color efficiently as dyes are not highly biodegradable [6.10–6.13]. Konsowa et al. (2013) studied the effectiveness of an MBR pilot plant for the treatment of dye wastewater. They found out that the removal efficiency of COD and dye matters improved with longer HRT [6.14]. Friha et al. (2015) investigated a submerged MBR for dye wastewater treatment and it resulted in high COD and color removal efficiency with HRT of 2 days [6.15]. MBR is able to treat wastewater efficiently at concentrations of biomass superior to an AS process. However, there is a limit of the biomass concentration in practical applications. In spite of the capacity of high biomass concentrations that the membranes can withstand without altering their performance, it is not usual to go above 8-10 g/L to avoid affecting the oxygen transfer coefficient [6.16].

Comparing to MBR, Moving Bed Biofilm Reactor (MBBR) has been applied to treat textile wastewater in recent years. MBBR is a biological treatment with carrier medias for the formation of attached biofilm. MBBR technology was first developed in the 1990s by Hallvard Ødegaard [6.17]. MBBR can withstand high organic load of the wastewater because of the biofilm fixed on the carriers. For this reason, the benefit of MBBR is space-saving or high treating capacity. Shin et al. (2006) investigated a pilot-scale MBBR system combined with coagulation for textile wastewater treatment. After the MBBR process, color removal rate was 70% and the overall color removal performance was improved after the coagulation with $FeCl_2$ [6.18]. Park et al. (2010) studied a pilot-scale MBBR process treating dye wastewater and the color and COD was removed 50% and 86%, respectively [6.19]. The organic load of MBBR could reach higher value than it of an AS reactor because a large amount of biomass is fixed on carriers [6.20, 6.21]. The worsening of the sludge decantation is the main problem of MBBR. In practical application, the addition of coagulant to obtain a well-clarified effluent is indispensable.

A hybrid MBBR-MBR system would solve the disadvantage of poor decanting of MBBR systems, by separating the biomass from the wastewater through the MBR membrane. On the other hand, the oxygen transfer coefficient will not be affected since the high concentration of biomass is fixed on MBBR carriers, which allows an energy saving in the aeration. Pervissian et al. (2012) studied the effectiveness of an MBBR-MF process in treating wastewater from a food industry. Their results showed that the membrane filtration minimized variable characteristics of the MBBR effluent caused by influent feed fluctuations [6.22]. To the best of our knowledge, two previous articles were published about biofilm-membrane filtration applied in textile wastewater treatment. The first one studied a combined anaerobic-aerobic MBBR-MF in treating textile wastewater with azo dye reactive brilliant red X-3B [6.23]. The color and COD in their influent were relatively low but their results obtained confirmed that the combination of MBBR with membrane is a feasible process for textile wastewater treatment. In the second study found, Spagni et al. (2010) evaluated an anaerobic/anoxic/aerobic system coupled to a microfiltration membrane for the treatment of textile wastewater. The decolorization mainly took place in the anaerobic biofilm tank which was filled up to vol. 51% of carriers [6.24]. To our best knowledge, no articles have been found of reusing textile water treated with MBBR-MBR.

In this study, a hybrid system: MBBR-MBR, was designed and built for treating textile wastewater from a Catalan textile industry. The study focused on the removal performance of organic compounds, suspended solids and color. Afterwards, the treated water was reused in dyeing processes to make new fabrics to evaluate the feasibility of reusing the treated effluent.

6.2. Materials and methods

6.2.1. Characteristics of textile wastewater

The textile wastewater used in this study was taken from the Catalan textile industry, Acabats del Bages (Monistrol de Montserrat, Spain). The wastewater was taken at the outlet of the homogenization tank. Characteristics of the wastewater: chemical oxygen demand (COD), biochemical oxygen demand (BOD), color, pH, conductivity, turbidity, total suspended solids (TSS), volatile suspended solids (VSS), total Kjeldahl nitrogen (TKN), total phosphorus (TP), are presented in Table VI-1.

Table VI-1 Wastewater characteristics

Parameters	**Unit**	**Value**
pH	—	8.44 ± 0.54
Conductivity	mS/cm	5.15 ± 0.47
Turbidity	NTU	306 ± 94
COD	mg/L	1496 ± 440
TSS	mg/L	337 ± 121
VSS	mg/L	267 ± 92
Color	mg Pt-co/L	700 ±234
TP	mg/L	10 ± 2
TKN	mg/L	55 ± 21

6.2.2. Reactor set-up

The detailed components of the hybrid MBBR-MBR pilot plant has been described in the Section 5.3 of Chapter V. Briefly, the hybrid MBBR-MBR reactor is designed considering a sufficient space for the correct movement of plastic carriers and the proper installation of the membrane. The diagram of the MBBR-MBR reactor is shown in Figure V-2 of Chapter V.

BIOFILL C-2 plastic carriers (BIO-FIL, Barcelona, Spain) were added to the MBBR tank with a filling ratio of 25 vol.%. The information of carriers is shown in Table III-1.

The total duration of the study was 222 days. During the start-up period, a long HRT (5 days) was set to allow the microbial adaptation and growth. The start-up stage was 30 days for the biofilm growth on carriers. Two operational periods were performed after the start-up stage. In the first period, MBBR was operated without the membrane, while in the second period the complete MBBR-MBR system was operated. In order to guarantee a stable status of the sludge, the flow rate was increased progressively from 1.8 L/h to 4.58 L/h during the first period. The membrane was installed on the day 114. A MOTIMO BT01 hollow fiber flat plat membrane (MOTIMO Membrane Technology Co., Ltd. (China)) with a membrane filtration area of 1 m^2 was immersed into the membrane zone. The characteristics of the membrane are included in Table III-3.

6.2.3. Reuse of treated water in new dyeing processes

New dyeing processes using the water treated by the MBBR-MBR reactor is made with a laboratory dyeing machine Ti-Color (Prato, Italy), shown in Figure III-17(a). The settings of dyeing and washing processes are set according to a previous study [6.25] and described in

Section 3.2.2 of Chapter III. The dyes used in the water reuse study were three commercial reactive dyes supplied by Dystar which were Procion Yellow HEXL, Procion Crimson HEXL and Procion Navy HEXL.

6.2.4. Analytical methods

Parameters of water characteristics: COD, TSS, VSS, TKN, TP, conductivity, turbidity, color and pH were determined according to the Standard Methods 23rd edition [6.26].

For the calculation of the concentration of biomass fixed on carriers, five carriers were sampled from the MBBR before the installation of membrane, after the installation of membrane and at the final of the studied period. For maintaining the percentage of carriers in the reactor, the same amount of new and marked carriers were put into the reactor after the sampled carriers were took out. The determination of biomass concentration fixed on carriers was carried out following the method reported in a previous study [6.27]. The carriers with attached biofilms were weighed and then were sonicated for 3 minutes. After that, centrifugation was performed in order to wash off the attached biomass from the carrier. Then the weight of clean carriers was measured, and the weight of biomass fixed to each carrier element was calculated. The total number of carriers was known and the total weight of fixed biomass per liter of reactor volume was calculated.

To determine the quality of dyed fabrics, the assessment of color reproducibility was performed with a spectrophotometer MINOLTA CM 3600d (Osaka, Japan) following the Standard UNE-EN ISO 105-J03 [6.28].

Color differences (DE) are determined by evaluation of the three cylindrical coordinates which describes color space: L* (luminosity), C* (chroma) and hº (hue angle from 0º to 360º). DL*, DC* and DH* are the differences of mentioned parameters versus their values of the reference dyeing [6.29]. The comprehensive description of the method has been presented in the section 1.8.6 of Chapter III.

In textile industry, $DE_{CMC\ (2:1)} \leq 1$ is the acceptance restriction of color differences for the control of dyeing quality [6.30].

6.3. Results and Discussion

6.3.1. HRT and MLSS

The operation of MBBR-MBR pilot plant was carried out for 222 days. After the start-up period, flow rate was increased from 1.8 L/h to 4.6 L/h in MBBR without membrane module in 42 days. The organic loading rate (OLR) in MBBR raised from 0.33 kg COD/ (m³ d) to 1.12 kg COD/ (m³ d) along with the increase of flow rate, and HRT was reduced from 2.5 days to 1 day. The attached and suspended biomass was adapted to the final flow rate gradually during the increase of flow rate. The membrane was installed when the flow rate was stable of 4.6 L/h and the OLR increased from 0.12 kg COD/ (m³ d) to 1.79 kg COD/ (m³ d). The MBBR-MBR pilot-plant worked during 125 days at HRT of 1 day, which is a short HRT comparing with a conventional MBBR-coagulation system, such as reported in a previous study [6.18]. MBBR-MBR plant also halved the HRT (2 days) of the CAS plant of the company, from where the textile wastewater was taken.

As shown in Figure VI-1, the mixed liquor suspended solids (MLSS) had a steady growth after the membrane installation at 114 days since the filtration blocked suspended matters.

The biomass attached to the carrier was 1.045 mg/carrier before the installation of membrane, 2.025 mg/carrier after the installation of membrane and 2.275 mg/carrier after the complete operation. That is, the biomass density was 0.11 g/L, 0.20 g/L and 0.23 g/L in the corresponding period.

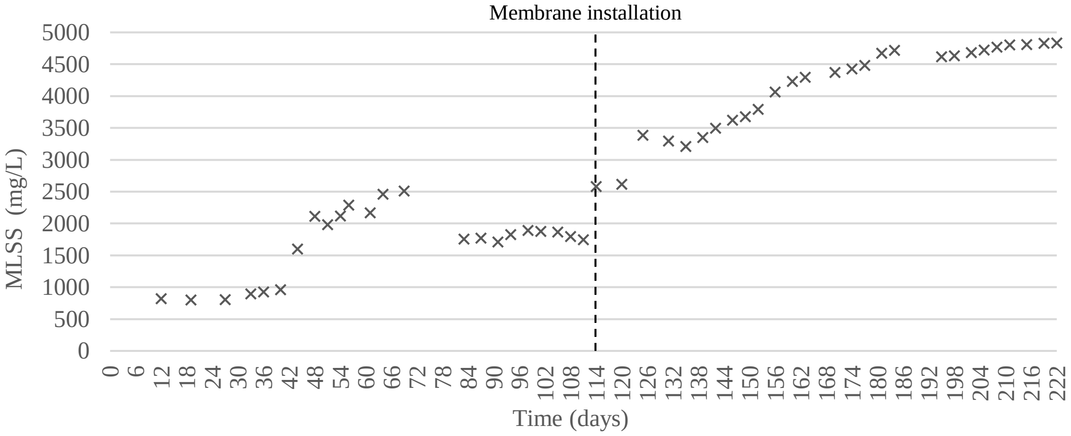


Figure VI-1 Concentration of MLSS in the reactor

6.3.2. Removal of COD

The average concentration of COD of the influent was 1500 mg/L during the study. After the membrane installation, the average COD removal achieved 93%±2%. The COD values of influent and effluent and COD removal rate during the complete operation are shown in Figure VI-2. The COD removal rate 78% of MBBR was higher than the COD removal of the conventional biological treatment [6.31]. The result showed that the removal efficiency of COD increased notably after the MBR installation. Due to the existence of non-biodegradable matters in textile wastewater, COD removal by simple biological methods is limited. Previous studies showed that normally the removal rate could reach 80 – 90% with CAS system [6.32-6.34]. Moreover, 93% of COD removal rate was obtained with the HRT of 1 day, which halved the HRT (2 days) of the CAS process of the textile industry from where the wastewater was taken. The reduction of HRT allows a significant space-saving or an increase in the treating capacity.

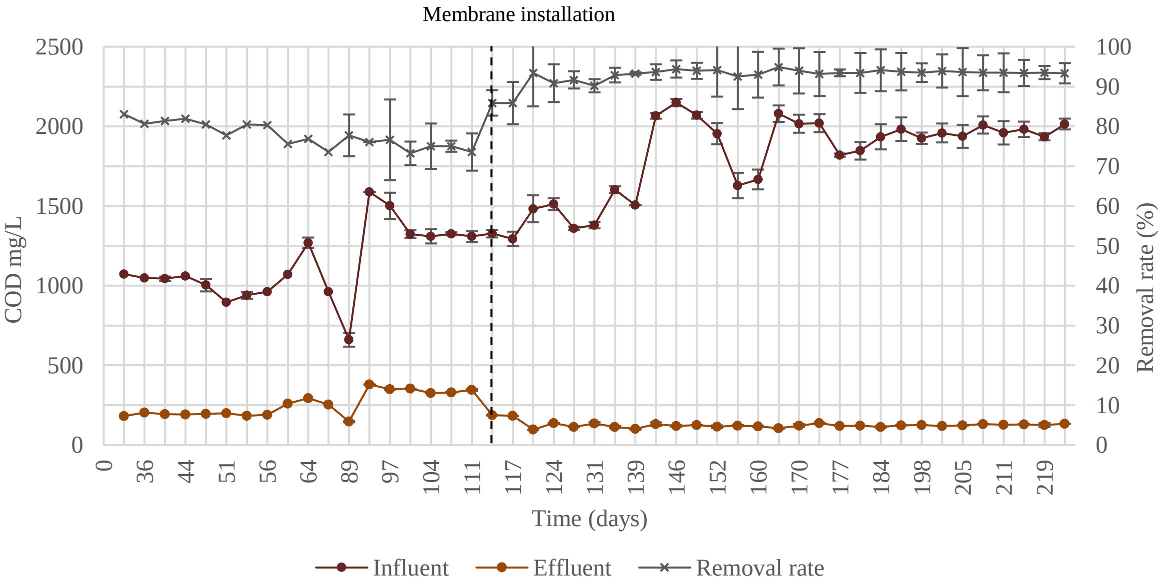


Figure VI-2 COD values of influent and effluent and COD removal rate

6.3.3. Color removal

CAS system is a common method in real textile industries for their wastewater treatment. CAS could accomplish the reduction of organic pollutants but could not meet the requirements of removing color from the wastewater. A tertiary treatment, such as chemical coagulation, is necessary for a satisfactory color removal. This tertiary treatment, however, increases the cost and generates secondary pollutants such as exhausted activated carbon or chemical sludge [6.35, 6.36]. During the experiments, color in the influent varied between 300-1000 mg Pt-co/L. The color removal performance of MBBR-MBR system is shown in Figure VI-3. The average color removal efficiency was 56%±7% during the MBBR operation and increased to 85%±2% after the membrane installed, which allowed the saving on the addition of decolorizing products that have significant environmental impact generally.

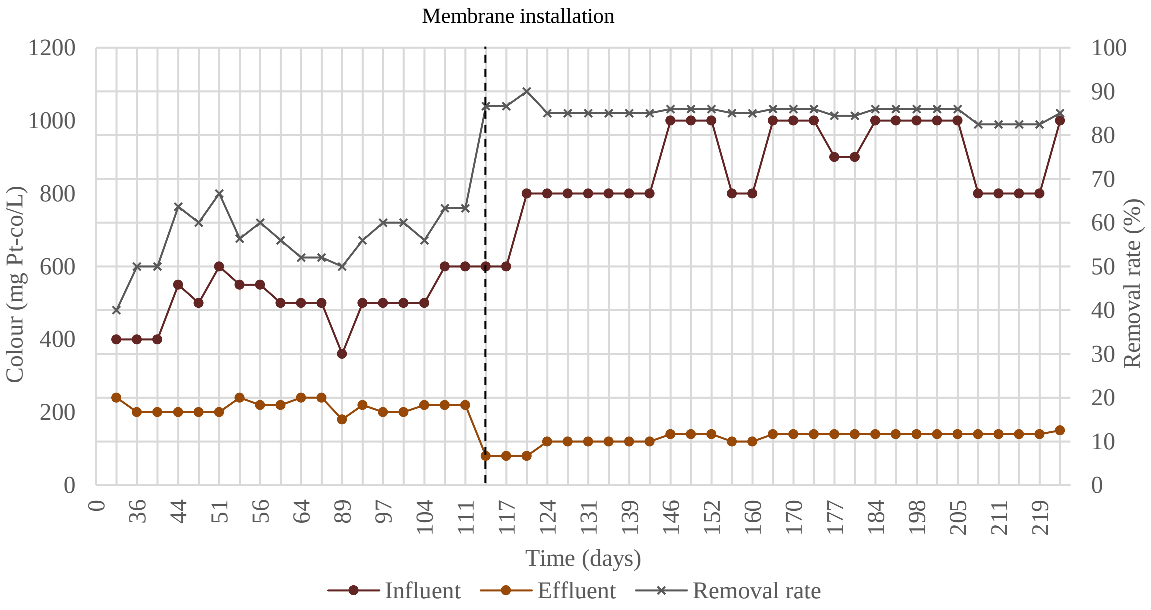


Figure VI-3 Color values of influent and effluent and color removal rate

6.3.4. TSS and turbidity removal

The TSS and turbidity removal rate is shown in Figure VI-4. TSS and turbidity removal rate followed the same tendency: the removal rate was fluctuating around 65% - 85% during the MBBR operation and increased to 99% of TSS and 100% of turbidity after the addition of the membrane. MBR process can result in a highly clarified effluent without the necessity to add coagulation product, which is usually required in a conventional MBBR. Generally, coagulation agents have a relatively high impact on the environment [6.37].

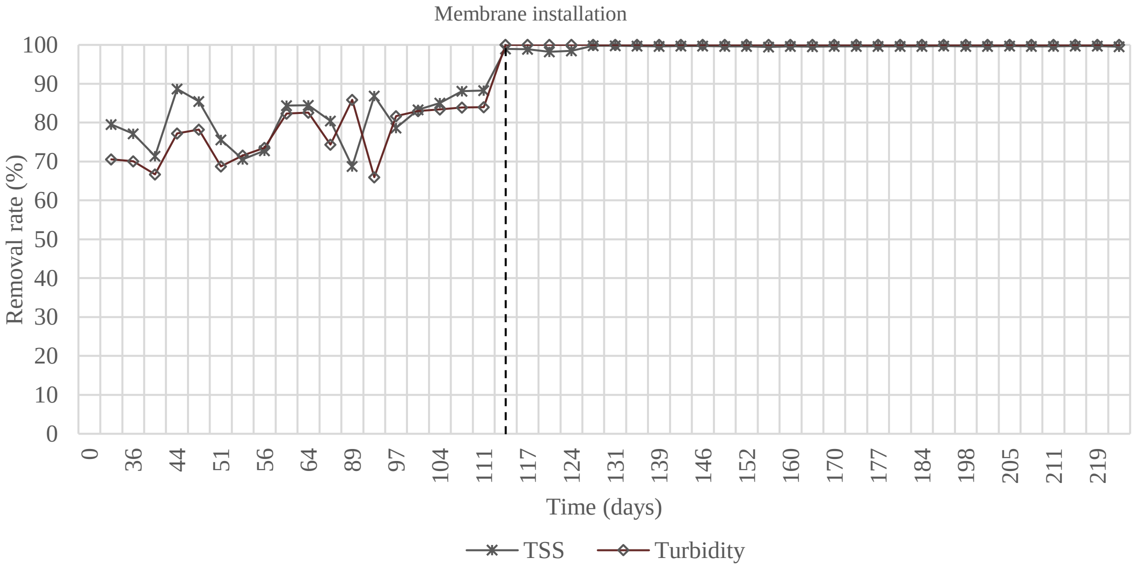


Figure VI-4 Removal rate of TSS and turbidity

6.3.5. Reuse of the treated effluent

Effluent obtained in the treatment was reused in a new dyeing process. A mixture of 70% of the effluent with 30% of softened tap water and a sample of 100% of treated effluent were reused in the dyeing process. Normally, the evaporation and the adsorption of water into the fiber causes 30% of the water lose during textile production. The quality of the dyes with 100% reused water (ideal reuse) was analyzed to evaluate the possibility to achieve a fully circular dyeing process, however, it would only represent 70% of the input water. 100% of softened tap water was used in the dyeing process as the reference sample. The color differences of dyeing processes with three reactive dyes comparing with a reference dyeing are shown in Table VI-2.

In the case of 70% effluent reused, $DE_{CMC(2:1)}$ values of all dyes were lower than 1, within the acceptance requirement of textile industry. The results demonstrate the feasibility of reuse the wastewater treated by the MBBR-MBR process with a proportion of 70% in the new dye baths.

In the case of 100% effluent reused, $DE_{CMC(2:1)}$ value of Procion Crimson HEXL and Procion Navy HEXL were below 1 within the acceptable range. $DE_{CMC(2:1)}$ value of Procion Yellow HEXL was 1.04, which is very close to the acceptance limit.

In order to further understand the dyeing performance, the effects on color differences of chromatic coordinates changes are studied (Table VI-2). The influence of each chromatic coordinates on dyeing results are:

- Yellow dyeing: The coordinates show that $DL_{cmc} > 0$ and $DC_{cmc} < 0$, which means that the color was lighter and slightly less saturated (duller) than the reference dyeing. Difference of hue (DH_{cmc}) are related to Da* and Db*. Yellow fabrics dyed with reused water were greener and slightly bluer.
- Crimson dyeing: As $DL_{cmc} < 0$ indicates that the color of new dyeings is darker than the reference. The cause could be that the slight residual color of the treated effluent increases the intensity of the red. When 100% effluent was reused, the $DC_{cmc} < 0$ means the color was slightly less saturated (duller). 70% effluent reused in new dyeing leads the Crimson fabrics (Red) without much change of hue, whereas in the case of 100% effluent reused, Crimson (Red) fabrics are greener and slightly bluer.
- Navy dyeing: As shown in the table, exceptionally low values of DL_{cmc}, DC_{cmc}, DH_{cmc} are observed. The slight residual color of the water does not affect the intensity, saturation and shade of the Navy (blue) fabric. The values of Da* and Db* show exceptionally low differences in tone.

In general, DH_{cmc} has the most effects on differences of color between fabrics dyed with treated water and reference fabric. Residual dyes remained in the effluent caused the fabrics slightly greener and bluer.

Table VI-2 Chromatic coordinates and color differences between fabrics dyed with the treated effluent and a reference dyeing

		DL_{cmc}	**DC_{cmc}**	**DH_{cmc}**	**Da***	**Db***	**$DE_{cmc(2:1)}$**
70% effluent reused	Procion Yellow HEXL	0.45	-0.16	0.78	-1.09	0.05	0.92
	Procion Crimson HEXL	-0.24	-0.02	0.33	-0.10	0.60	0.41
	Procion Navy HEXL	-0.15	0.02	0.14	0.13	-0.06	0.21
100% effluent reused	Procion Yellow HEXL	0.34	-0.38	0.90	-1.57	-0.48	1.04
	Procion Crimson HEXL	-0.29	-0.39	-0.37	-1.04	-0.72	0.61
	Procion Navy HEXL	0.38	0.16	-0.24	-0.29	-0.19	0.48

Figure VI-5 shows the visual comparison of the fabrics dyed using the treated water with respect to the reference.

Figure VI-5 Visual comparison of fabrics made with the three dyes

In order to present the color differences more precisely, color converted with CIELAB of each fabric is shown in Figure VI-6. The example of the color convertor of Procion Crimson HEXL is demonstrated in Figure VI-7.

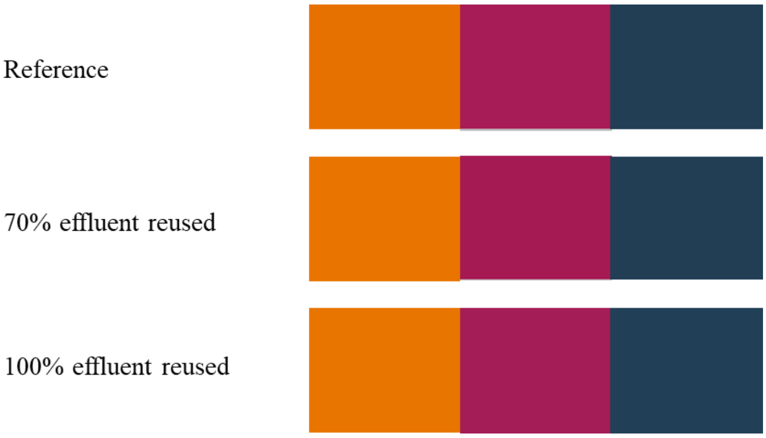
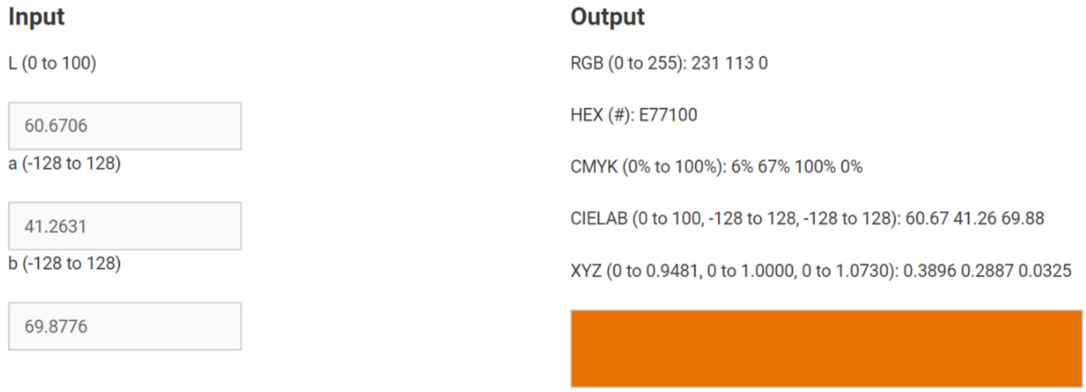


Figure VI-6 Comparison of color of cotton fabrics using CIELAB color space.



Input

L (0 to 100)

60.6706

a (-128 to 128)

41.2631

b (-128 to 128)

69.8776

Output

RGB (0 to 255): 231 113 0

HEX (#): E77100

CMYK (0% to 100%): 6% 67% 100% 0%

CIELAB (0 to 100, -128 to 128, -128 to 128): 60.67 41.26 69.88

XYZ (0 to 0.9481, 0 to 1.0000, 0 to 1.0730): 0.3896 0.2887 0.0325

Figure VI-7 Example of color converting process of Procion Crimson HEXL

6.4. Conclusions

The effectiveness of hybrid system MBBR-MBR in real textile wastewater treatment and the reuse of treated effluent was demonstrated in this study.

MBBR-MBR showed better performance than single MBBR. A better performance was observed after the membrane installation in COD removal (93%), color removal (85%), TSS (99%) with HRT of 1 day. The HRT reduction of 50% in practical cases is promising that can result in space and energy saving. COD removal of 93% is almost the maximum for a biological process in this type of wastewater because the water is not completely biodegradable. 85% of color removed without a tertiary treatment is an attractive option for textile wastewater treatment from the perspective of economic and environmental impact. The results show that the hybrid MBBR-MBR system is an effective process for textile wastewater treatment.

A mixture of 70% of treated water with 30% of softened tap water and a sample of 100% treated water was reused for the dyeing process. In the case of 70% treated water reused, the color differences $DE_{CMC(2:1)}$ with respect to the reference dyes of three monochrome dyes (Procion Yellow HEXL; Procion Crimson HEXL; Procion Navy HEXL) were inferior to 1, the limit value accepted for textile industry. In the case of 100% regenerated water reused, the color difference with respect to the reference of two dyes were below 1 and of dye Procion Yellow HEXL was 1.04, which is on the acceptance limit. Therefore, the MBBR-MBR treatment allows new dyeings to be made with 100% treated water. If the textile industry installs an MBBR-MBR plant to treat all the wastewater from production and wants to reuse all the treated water, it could reduce its consumption of tap water up to 70%.

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Yang, X., López-Grimau, V., Vilaseca, M., Crespi, M., Ribera-Pi, J., Calderer, M., & Martínez-Lladó, X. (2021). Reuse of textile wastewater treated by moving bed biofilm reactor coupled with membrane bioreactor. Coloration Technology, 00, 1–9. https://doi.org/10.1111/cote.12543

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CHAPTER VII. REDUCTION OF COST AND ENVIRONMENTAL IMPACT IN THE TREATMENT OF TEXTILE WASTEWATER USING THE HYBRID MBBR-MBR SYSTEM

In this chapter, the economic and environmental feasibility of applying the hybrid MBBR-MBR system on industrial scale was conducted using the experimental results of Chapter VI.

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7.1. INTRODUCTION

The printing and dyeing wastewater discharged from the textile industry during its production is one of the factors affecting the water environment. As the demand for water in the textile continues to increase, the discharge standards are becoming stricter, and the water bill continues to rise, how to treat textile wastewater with high efficiency and low consumption is an important task. For industrial-scale wastewater treatment, we must not only pay attention to whether the treating efficiency meets the discharge standard, but also combine the sustainable development strategy and investment analysis to find the most suitable treatment method.

The processing products of the textile industry are mainly cotton, wool, silk, chemical fiber, etc. Each production process needs a specific processing technology and corresponding sizing agents, dyes, and auxiliaries [7.1]. Due to the different raw materials processed, product varieties, different processing techniques and processing methods, the composition of this type of wastewater is very complex, and it is often characterized by variable pH, high concentrations of Chemical Oxygen Demand (COD), high turbidity, problems of color and limited biodegradability due to the dyes remained in the wastewater [7.2]. In response to such complex type of wastewater, researchers and industries have developed various treatment processes, such as physico-chemical (coagulation-flocculation, adsorption, advanced oxidation and filtration), biological technologies and combined treatment processes. Compared with physico-chemical methods, biological processes are more environmentally friendly because of the complete degradation of contaminants without producing secondary pollutants [7.3].

Among different biological treatments, Membrane Bioreactor (MBR), as a promising process combining biological treatment and membrane filtration, has been increasingly applied for industrial wastewater treatment, including the textile sector [7.4]. MBR process has shown several advantages over Conventional Activated Sludge (CAS) treatment, such as small footprint, stable effluent quality, high tolerance to high concentrations of organic matters, and lower sludge production [7.5, 7.6]. Due to the benefits of MBR over CAS reflected in better effluent quality, no additional chemical products needed and lower sludge production, the MBR process has proved in Life Cycle Assessment (LCA) studies to be the more eco-friendly option, especially in environmental impact global warming potential, abiotic depletion and acidification, etc. [7.7, 7.8]. In previous techno-economic research, MBR showed higher cost due to the large energy consumption, but the fact that MBR plant can reduce the Hydraulic Retention Time (HRT) compensates for the higher power expense [7.9, 7.10]. However, even MBR can efficiently treat wastewater with higher organic load than CAS process, the concentration of biomass is limited in practical applications to avoid affecting the oxygen transfer coefficient [7.11].

Another biological treatment that has been attracting more and more attention in textile wastewater treatment in recent years is Moving Bed Biofilm Reactor (MBBR), thanks to its ability to withstand way higher biomass concentration. The organic load of MBBR could reach higher than that of CAS processes because a large amount of biomass is fixed to the carriers [7.12]. Previous studies have shown that MBBR can effectively remove COD, but its ability to remove color is limited because of its incompleteness of sludge decantation [7.13, 7.14]. In practical application, it is essential to add coagulant to obtain a well-clarified effluent, and in many cases, extra decolorizing agent needs to be used for improving the removal of color [7.15]. From the economic and environmental perspective, these extra products that need to be added have a cost and result in environmental impacts. For example, most of the decolorizing agents used are quaternary ammonium salt [7.16], which have a high impact on the toxicity category [7.17].

A hybrid MBBR-MBR system will improve sludge decantation of the MBBR system by the MBR membrane filtration. On the other hand, since a part of biomass is fixed on the MBBR carriers, the oxygen transfer coefficient will not be affected, allowing energy saving for aeration. Two previous studies have been published on the application of biofilm-membrane filtration in textile wastewater treatment showing this treatment is viable for treating textile wastewater [7.18, 7.19]. With the effective treatment of MBBR-MBR, the effluent does not need to add coagulant or decolorizing agent, which will notably reduce the environmental impact. To the best of our knowledge, no articles have been found on the reuse of treated textile wastewater by MBBR-MBR. Economically, water reuse in textile industry will allow a particularly important saving not only in the water cost as a consumer but also in the environmental tax.

In this study, a hybrid system: MBBR-MBR, was designed and built for treating textile wastewater from a local textile industry. The study focused on the removal performance of organic compounds, suspended solids and color. Afterwards, the treated water was reused in dyeing processes to make new fabrics to evaluate the feasibility of reusing the treated effluent.

Based on the experimental results in the pilot plant, an economic study and LCA analysis were carried out to evaluate the economic and environmental feasibility of the implementation of the hybrid MBBR-MBR on an industrial scale, which to our knowledge, no such research has been done. The feasibility analysis of the MBBR-MBR system is based on the comparison with the results of the CAS system of the textile industry that provided us wastewater for this study.

7.2. Design and methodology

7.2.1. Characteristics of textile wastewater

The textile wastewater used in this study was taken from a Catalan textile industry. The wastewater was taken at the outlet of the homogenization tank. Characteristics of the wastewater: chemical oxygen demand (COD), biochemical oxygen demand (BOD), color, pH, conductivity, turbidity, Total Suspended Solids (TSS), volatile suspended solids (VSS), total Kjeldahl nitrogen (TKN), total phosphorus (TP), are presented in the Table VI-1 of Chapter VI.

7.2.2. Reactor set-up

The hybrid MBBR-MBR reactor is designed considering a sufficient space for the correct movement of plastic carriers and the proper installation of the membrane. The diagram of the MBBR-MBR reactor is shown in Figure V-2 of Chapter V. The total volume of the hybrid MBBR-MBR reactor was 147 L, and the effective volume was 110 L. The reactor was divided into two parts: the MBBR part (79 L) and the membrane tank (31 L). The temperature during the operation was maintained at 25 °C, controlled by a submerged thermostat. A peristaltic pump was used for the wastewater input. Diffusers were fixed on the bottom of the reactor, of which the fine bubble diffuser was for oxygen diffusion, and the two thick bubble diffusers were for mixing carriers and for air scouring the membrane. The characteristics of carries used in the study are presented in Figure III-3 of Chapter III.

The total duration of the study was 222 days. The membrane was installed after the growth of biofilm on the carriers of MBBR was stabled. The flow rate was increased progressively to maintain a stable status of the sludge. A MOTIMO BT01 hollow fiber flat plat membrane (MOTIMO Membrane Technology Co., Ltd. (China)). Details of the membrane are included in Figure III-7 of Chapter III.

7.2.3. Reuse of treated water in new dyeing processes

New dyeing processes using the MBBR-MBR reactor's treated water was performed with a laboratory dyeing machine Ti-Color (Prato, Italy). To increase the affinity between the dye and the fiber, a certain amount of salt is added during the dyeing process. The dyeing and washing processes were set according to the previous study [7.20] and was described in Section 3.2.2 of Chapter III. The dyes used in the water reuse study were three commercial reactive dyes supplied by Dystar, which were Procion Yellow HEXL, Procion Crimson HEXL and Procion Navy HEXL.

7.2.4. Analytical Methods

During this study, the determination of water quality was carried out by monitoring the COD, TSS, color, pH, conductivity and turbidity, following the Standard Methods 23rd edition [7.21].

The quality of dyed fabrics with reused water was determined according to Standard UNE-EN ISO 105-J03 by color differences with respect to reference dyeing performed with softened tap water [7.22]. Total color differences ($DE_{CMC(2:1)}$) were calculated from lightness (DL*), chroma (DC*), and Hue (DH*) using the following the Equation III-2 of Chapter III.

A spectrophotometer, MINOLTA CM 3600d (Osaka, Japan), was used for these measurements according to Standard illuminant D65/10°. Generally, the color difference of one unit ($DE_{CMC(2:1)} \leq 1$) is the acceptable limit in the textile industry [7.23].

7.2.5. Economic Analysis

The economic assessment of capital expenditures (CAPEX) and operational expenditures (OPEX) for the MBBR-MBR schemes is determined in the results section. The results are compared with the previous study of CAS system treating the same wastewater [7.24]. Additionally, the financial feasibility analysis was conducted by examining the Net Present Value (NPV) and the Internal Rate of Return (IRR). The calculation of NPV and IRR is taken into consideration of the investment payback period as 15 years. NPV is the sum of the present value of the net income obtained by folding the income and cost flow back to the beginning of the period. NPV is calculated using the following Equation III-4 of Chapter III.

IRR is the interest rate when the cumulative NPV is zero. This IRR means the rate of the largest currency devaluation that the project can withstand. It is also calculated using the Equation III-4.

7.2.6. Environmental Impact Analysis

To compare the environmental impact of the existing CAS system and the new MBBR-MBR process, life cycle assessment (LCA) is performed according to standard ISO 14040 [7.25]. Simapro is used as the LCA software. The database used is Ecoinvent 3.1. ReCiPe, midpoint and endpoint approach, and Hierarchist perspective are considered as the methodology to calculate environmental impact. The selected functional unit is “1 m^3 of treated effluent”. The data used in this study is taken from the experimental results.

7.3. Results and Discussion

7.3.1. Treating Efficiency

During the experiment, the average concentration of COD of the influent was 1500 mg/L. The MBBR-MBR treatment had an average COD removal of 93% with the HRT of 1 day. The removal rate maintained steady even when the COD values of influent fluctuated greatly. This is because MBBR has a strong ability to resist shock organic loading [7.26]. The hybrid MBBR-MBR process achieved to halve the HRT (2 days) of the CAS process of the textile industry from where the wastewater was taken. The reduction of HRT allows a significant space-saving or an increase in the treating capacity.

Color of the influent varied between 300-1000 mg Pt-co/L during the experiment. The average color removal efficiency was increased to 85% when the treatment was stable, which allowed the saving on the addition of decolorizing agent that generally result in significant environmental impact. TSS and turbidity removal rate was up to 99% and 100%, respectively. The MBR part of the hybrid system can lead to a highly clarified effluent without the necessity of adding coagulation product, which is usually required in a conventional MBBR. Generally, coagulation agents have a relatively high impact on the environment [7.27].

In the previous study of CAS treating the same wastewater, the average removal rate of COD, color and TSS was 83%, 55% and 66%, respectively [7.24]. The color removal of 55% was insufficient to comply with current legislation and decolorizing agent must be added. These experimental results of present study and previous study are used to calculate the economic costs and environmental impacts of the LCA study.

7.3.2. Reuse of the treated water

Treated water obtained after the MBBR-MBR process was reused in a new dyeing process. Normally, the amount of wastewater discharged by a textile industry discharges accounts for 70% of the total freshwater consumption [7.24]. 100% of softened tap water was used in the dyeing process as the reference sample. The color differences of dyeing processes with three reactive dyes were compared with the reference dyeing are shown in Table 4. As shown in the table, $DE_{CMC(2:1)}$ value of Procion Crimson HEXL and Procion Navy HEXL were below 1 within the acceptable range. $DE_{CMC(2:1)}$ value of Procion Yellow HEXL was 1.04, which is on the acceptance limit.

Table VII-1Chromatic coordinates and color differences between fabrics dyed with the treated effluent and the reference dyeing

100% effluent reused	**DL_{CMC}**	**DC_{CMC}**	**DH_{CMC}**	**$DE_{CMC(2:1)}$**
Procion Yellow HEXL	0.34	-0.38	0.90	1.04
Procion Crimson HEXL	-0.29	-0.39	-0.37	0.61
Procion Navy HEXL	0.38	0.16	-0.24	0.48

7.3.3. Economic Study of the hybrid system

The local textile industry from where the wastewater was taken produces 222,700 m^3 of wastewater per year with 11 months under operation. The wastewater treatment method of the industry is conventional activated sludge (CAS). The daily treatment flow is 920 m^3/d. The current HRT of the CAS plant is 2 days.

7.3.3.1. Capital Expenditures (CAPEX)

The CAPEX of the CAS system was considered to be the reference (0) in the economic study. The CAPEX of the MBR-MBBR treatment was added directly to the CAPEX of the CAS system.

For the MBR part, the membrane and the installation of the membrane (366,153 €) have been considered for the CAPEX estimation according to the CAPEX calculation from a study of the cost of a small MBR (100–2500 m^3/d flow capacity) [7.28]. For the MBBR part, the cost of carrier medias (96,250 €) has been considered for the CAPEX estimation according to the suppliers' information. So, the total CAPEX is 462,403 €.

7.3.3.2. Operational Expenditures (OPEX)

Consumption of energy, decolorizing agent data, and environmental tax of wastewater discharge and sludge production were gathered in order to estimate operational expenditures (OPEX) of the MBBR-MBR treatment.

Additionally, the cost of membrane replacement represented 2.4% of the energy cost [7.29], and the average lifetime of the UF membrane was taken as 10 years. The maintenance and repair costs represented 19.5% of the energy cost [7.29]. MBBR-MBR could withstand higher biomass concentrations with much longer sludge retention time (SRT) than CAS, which allows much less sludge production after the treatment and consequently lowers the frequency of sludge disposal [7.30]. During the experimental study of MBBR-MBR, sludge concentration did not exceed the withstanding limit of the membrane. The sludge generation was estimated according to the increasing rate of the biomass concentration and the concentration limit for the membrane.

The OPEX values of a CAS plant of our previous study [7.24] are listed for the comparison with MBBR-MBR. The detailed OPEX calculation of the existing CAS plant and the MBBR-MBR plant is demonstrated in the following Table VII-2 and Table VII-3, respectively.

Table VII-2 CAS operational cost for treating 1 m^3 wastewater

Concept						***Total Price €/m^3***	***Reference***
(a) Consumption	**Unit**	**Amount**	**Unit**	**Unit price**	**Convert to €/m^3**	**0.55**	
Electricity	kW/m^3	0.96	€/kw	0.187	0.17952		[7.31]
Decolorizing agent	kg/m^3	0.2	€/kg	1.85	0.37		[7.32]
(b) Environmental tax	**Unit**	**Amount**	**Unit**	**Unit price**		**0.86**	
Sludge generation	kg/m^3	0.83	€/kg	0.158	0.013114		[7.33]
Wastewater discharge							[7.34]
OM[1]	kg/m^3	0.23	€/kg	1.0023	0.230529		
TSS	kg/m^3	0.32	€/kg	0.5011	0.160352		
N	kg/m^3	0.008	€/kg	0.761	0.006088		
P	kg/m^3	0.003	€/kg	1.5222	0.0045666		
Conductivity	S/cm	0.00598	€/Sm^3/cm	8.0198	0.0479584		
summation					0.449494		
ST[2] *= 1.5 ×SUM*					0.67424101		
GT[3]					0.163		
Total price						**1.41**	

[1] OM: organic material; [2] ST: specific tax; [3] GT: general tax.

Table VII-3 MBBR-MBR operational cost for treating 1 m^3 wastewater

Concept						***Total Price €/m^3***	***Reference***
(a) Consumption	**Unit**	**Amount**	**Unit**	**Unit price**	**Convert to €/m^3**	**0.21**	
Electricity	kW/m^3	1.12	€/kw	0.187	0.20944		[7.31]
Decolorizing agent	kg/m^3	0	€/kg	1.85	0		[7.32]
(b) Environmental tax	**Unit**	**Amount**	**Unit**	**Unit price**		**0.35**	
Sludge generation	kg/m^3	0.023	€/kg	0.158	0.003634		[7.33]
Wastewater discharge							[7.34]
OM[1]	kg/m^3	0.11	€/kg	1.0023	0.110253		
TSS	kg/m^3	0.006	€/kg	0.5011	0.003006		
N	kg/m^3	0.007	€/kg	0.761	0.005327		
P	kg/m^3	0.001	€/kg	1.5222	0.001522		
Conductivity	S/cm	0.00482	€/Sm^3/cm	8.0198	0.038655		
summation					0.123742		
ST = 1.5 ×SUM					0.185613		
GT					0.163		
(c) Membrane replacement						**0.01**	
(d) Maintenance and repair						**0.04**	
Total price						**0.61**	

In terms of the consumption part, although MBBR-MBR had higher electricity consumption because it required more electricity to operate and to maintain the membrane filtration, CAS operation cost more in consumption due to the use of Decolorizing agent. This was not necessary for MBBR-MBR because it achieved the color removal requirement.

In regard to environmental tax, thanks to the great performance of MBBR-MBR treatment on organic compounds, color and TSS removal, it had a lower expense (0.35 €/m^3) than the expense of CAS (0.86 €/m^3).

7.3.3.3. Evaluation of the Economic Feasibility (NPV and IRR)

The CAPEX and OPEX of the MBBR-MBR system have been commented in the above sections. The values of expenditures refer to year zero and have been re-adjusted at a rate of 1.4% yearly in the following years. This rate is taken from the average value of the Spain's inflation target in the next five years [7.35]. Furthermore, due to water recovery and the reduction in the wastewater, discharge revenues and some costs being avoided could be achieved. By MBBR-MBR treatment, as it was demonstrated that water recovery could reach 70% approximately. The water recovery will allow the saving in water consume cost and the discharge tax. The textile industry with CAS treatment pays 0.56 € for each m^3 of water used [7.36], and also has been paying 0.86 € for each m^3 of wastewater discharged. Considering the daily treatment flow of 920 m^3/d, it was assumed that 644 m^3 of water could be recovered, and 644 m^3 of water was not discharged daily. Therefore, the avoid cost of water consumption is 360.64 € daily and the avoid cost of water discharge is 553.84 € daily.

With all these cost components, cash flow was assessed for 15 years of economic life in Table VII-4.

Table VII-4 Cash flow (€) assessment for membrane filtration alternative

Year	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**	**13**	**14**	**15**
Revenues															
Water recovery	0	87,275	88,497	89,736	90,992	92,266	93,558	94,868	96,196	97,542	98,908	100,293	101,697	103,121	104,564
Reduction in discharge	0	134,029	135,905	137,808	139,737	141,694	143,677	145,689	147,729	149,797	151,894	154,020	156,177	158,363	160,580
Total Revenue	0	221,304	224,402	227,544	230,730	233,960	237,235	240,556	243,924	247,339	250,802	254,313	257,874	261,484	265,145
Expenditures															
CAPEX	462,403	0	0	0	0	0	0	0	0	0	0	0	0	0	0
OPEX	0	135,847	137,749	139,677	141,633	143,616	145,626	147,665	149,732	151,829	153,954	156,110	158,295	160,511	162,758
Total	462,403	135,847	137,749	139,677	141,633	143,616	145,626	147,665	149,732	151,829	153,954	156,110	158,295	160,511	162,758
NET CASH FLOW	-462,403	85,457	86,653	87,867	89,097	90,344	91,609	92,891	94,192	95,511	96,848	98,204	99,578	100,972	102,386

While calculating the NPV for the alternatives, the discount rate was assumed to be 10 %. In addition, as it was stated in the cash flow calculations, economic life of the MBBR-MBR system was taken as 15 years. By using the Equation III-4, NPV for MBBR-MBR system is 193,990 €.

The internal rate of return (IRR) is an investment evaluation method, that is, to find out the potential rate of return of the asset. The principle is to use the internal rate of return to discount, and the net present value of the investment is exactly zero [7.37]. IRR is calculated assuming the value of NPV as zero using the same Equation III-4. If the project has high IRR value, then it can be concluded that the project has high financial feasibility. However, if the IRR value is lower than the discount rate, the application of the system would be unattractive. The IRR value calculated is 18%, which is higher than the discount rate assumed.

Both NPV value and IRR suggest that MBBR-MBR system is financially applicable for the implantation of into industrial scale.

7.3.4. LCA study results

7.3.4.1. Inventory results

The inventory results of the MBBR-MBR treating process are shown in Table VII-5. All data are related to the functional unit (1 m^3 treated water). The impact of sludge generation was not taken into account in Simapro software; therefore, the impact of sludge generation could not be quantified in the LCA study. Nevertheless, sludge generated in the treatment was quantified and is presented in Table VII-5.

Table VII-5 Inventory analysis of the MBBR-MBR treatment

Processes Included in LCA	**MBBR**		***Unit/FU***	***Ecoinvent Unit Process***
	Input	**Output**		
COD	2	0.13	kg	
TSS	0.94	0.01	kg	
N	0.055	0.003	kg	
P	0.008	0.001	kg	
Color	700	105	g Pt-co	
Conductivity	6.46	5.42	mS/cm	
Wastewater	1	1	m^3	
Sludge		0.021	kg	
decolorizing agent	0		kg	DTPA, diethylenetriaminepentaacetic acid, at plant/RER U
Electricity	1.12		kWh	Electricity, medium voltage, production ES, at grid/ES U

7.3.4.2. Environmental impact assessment

The environmental impact of MBBR-MBR treatment according to the LCA results using endpoint approach is discussed, and then the results of MBBR-MBBR treatment is compared with the CAS treatment of our previous study using the same criteria with respect to their total environmental impact.

The results of the environmental impact assessment are presented in points (mPt) so that different categories could be compared. During the MBBR-MBR treatment, as can be seen in Table VII-6, there was no consumption of decolorizing agent since the system removed most of the color in the effluents. The consumption of electricity during treatment represents the total environmental impact. The results show that the impact on Ecosystem was much lower, while the major impacts were occurred on Resources and Human Health.

Table VII-6 Environmental impact of MBBR-MBR

	Human Health (mPt)	***Ecosystems (mPt)***	***Resources (mPt)***
Electricity (kWh/m^3)	20.7	1.7	28.8
Decolorizing agent(kg/m^3)	0	0	0
TOTAL	20.7	1.7	28.8

Figure VII-1 shows the impact of electricity consumption of MBBR-MBR treatment on the detailed categories related to Human Health, Ecosystem, and Resources. Climate change human health and Particulate matter formation categories were the main factors that had an impact on Human health of electricity consumption. In the meantime, the impact on Ecosystems mainly was due to Agricultural land occupation and Climate change ecosystem, while Terrestrial ecotoxicity, Natural land transformation, Urban land occupation, and Terrestrial acidification had minor impacts on the Ecosystem category. Furthermore, the major impact on Resources came from Fossil depletion category, while the Metal depletion category had almost no impact.

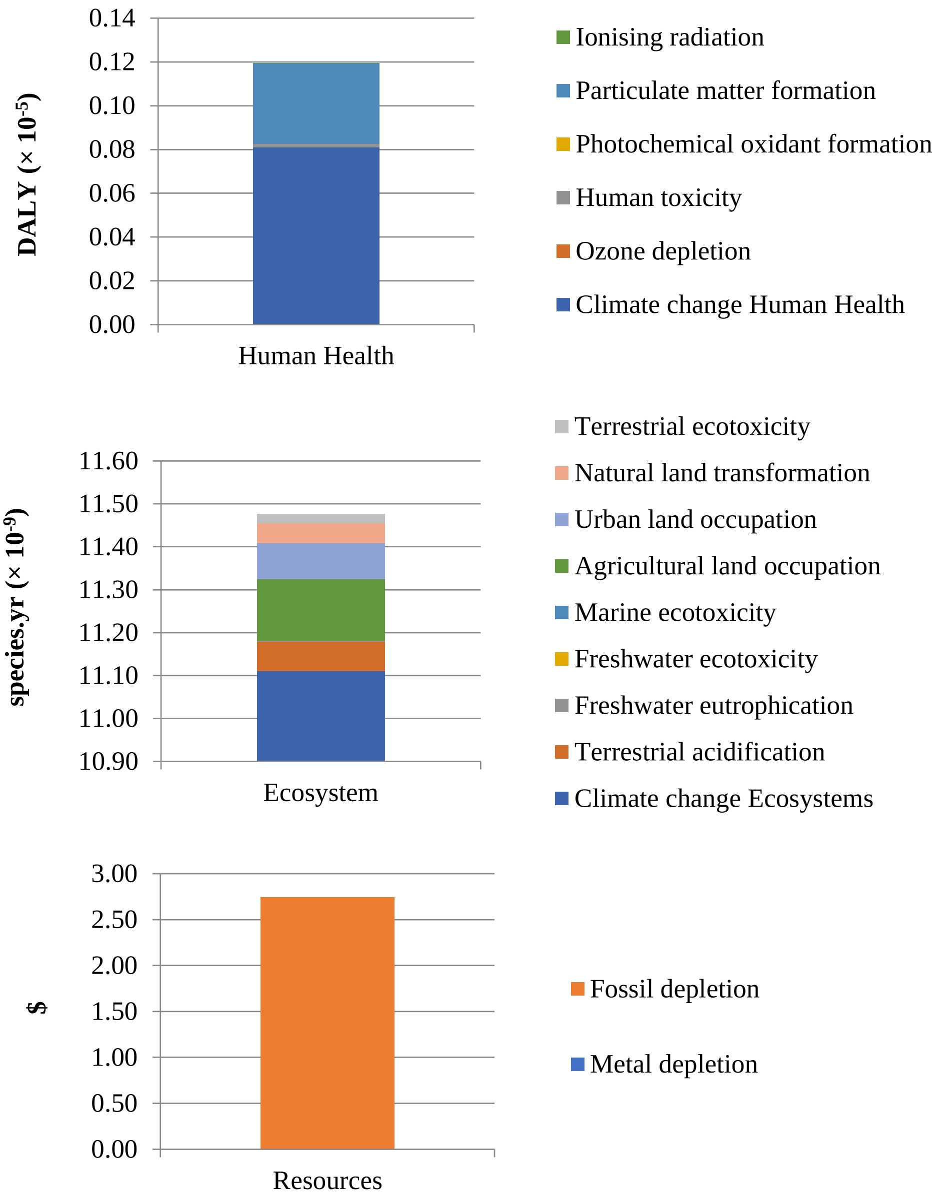

Figure VII-1 Analysis of the effect of electricity consumption on the impacted categories of MBBR-MBR

The environmental impact of MBBR-MBR process was compared with the environmental impact of CAS of our previous study [7.24]. The comparison of environmental impacts is demonstrated in Table VII-7.

As shown, the MBBR-MBR system had a lower impact on all three categories. Although, according to Table VII-2 and Table VII-3, MBBR-MBR had a slightly higher energy consumption of 1.12 kW/m^3 compared with 0.96 kW/m^3 of CAS, the endpoint results have shown that avoiding the use of decolorizing agent fully compensated for the environmental impact due to high energy consumption.

Table VII-7 Environmental impacts of the three processes

	Human Health (mPt)	**Ecosystems (mPt)**	**Resources (mPt)**
CAS	57.2	5.3	113.0
MBBR-MBR	20.7	1.7	28.8

If endpoint methods are helpful for decision-making because they can compare results in points, then midpoint analysis can help identify issues of specific environmental concern [7.38]. The results of midpoint assessment of MBBR-MBR were also compared with the previous CAS study, shown in Table VII-8. It can be observed clearly that MBBR-MBR had more environmental advantages since its impacts were the lower in all the categories, especially in Climate change Human Health, Marine eutrophication, Terrestrial ecotoxicity, Freshwater ecotoxicity and Marine ecotoxicity, thanks to the high quality of the effluent treated by MBBR-MBR and the avoiding of using extra decolorizing agent.

Table VII-8 Comparison of CAS and MBBR-MBR: midpoint analysis.

Impact Category	**Unit**	**CAS**	**MBBR-MBR**	**Impact reduction of MBBR-MBR**
Climate change Human Health	kg CO_2-eq	1.29	0.08	94%
Ozone depletion	kg CFC-11 eq	1.39×10^{-7}	2.89×10^{-8}	79%
Human toxicity	kg 1.4-DB eq	0.12	0.03	79%
Photochemical oxidant formation	kg NMVOC	3.89×10^{-3}	2.30×10^{-3}	41%
Particulate matter formation	kg PM10 eq	1.95×10^{-3}	1.40×10^{-3}	28%
Ionising radiation	kg U235 eq	0.16	0.11	33%
Terrestrial acidification	kg SO_2-eq	6.46×10^{-3}	4.83×10^{-3}	25%
Freshwater eutrophication	kg P-eq	7.84×10^{-5}	2.79×10^{-5}	64%
Marine eutrophication	kg N-eq	2.24×10^{-3}	1.76×10^{-4}	92%
Terrestrial ecotoxicity	kg 1.4- DB eq	2.96×10^{-4}	6.69×10^{-5}	76%
Freshwater ecotoxicity	kg 1.4- DB eq	7.40×10^{-3}	1.02×10^{-4}	99%
Marine ecotoxicity	kg 1.4- DB eq	1.04×10^{-3}	2.14×10^{-4}	79%
Agricultural land occupation	m^2 year	5.51×10^{-3}	5.22×10^{-3}	5%
Urban land occupation	m^2 year	2.16×10^{-3}	1.79×10^{-3}	17%
Natural land transformation	m^2 year	1.25×10^{-5}	1.10×10^{-5}	12%
Water depletion	m^3	1.12×10^{-2}	3.33×10^{-3}	70%
Metal depletion	kg 1Fe eq	2.02×10^{-3}	8.57×10^{-4}	58%
Fossil depletion	kg oil eq	5.17×10^{-7}	1.70×10^{-7}	67%

7.4. Conclusions

The experimental study of a hybrid MBBR-MBR showed an efficient removal of COD (93%), color (85%), TSS (99 %) with 1 day of HRT. The HRT reduction of 50% for the application of industrial scale is very attractive resulting in space and energy saving. The MBBR-MBR treatment allows new dyeings to be made with 100% treated water, representing 70% of the textile industry's total water consumption.

LCA study suggested that the hybrid MBBR-MBR system generated much lower environmental impact than CAS treatment, mainly because no extra decolorizing agent was used. The values of CAPEX and OPEX showed that MBBR-MBR had smaller expenses than CAS. Higher efficiency of MBBR-MBR system resulted in lower discharge tax and without cost for decolorizing agent. Although the energy consumption was higher, it was compensated by the mentioned advantages. Additionally, in the NPV and IRR study, water reuse after the treatment played an important role leading to the cost saving of water consume and discharge tax. The 18% of IRR calculated demonstrated that MBBR-MBR has great economic feasibility for textile wastewater treatment on the industrial scale.

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CHAPTER VIII. CONCLUSIONS AND RECOMMENDATIONS

8.1. Conclusions

The core of the thesis has been the development and investigation of the hybrid system Moving Bed Biofilm Reactor coupled with Membrane Bioreactor (MBBR-MBR) in treating textile wastewater, which is a novel process for the treatment in such wastewater.

This process allows the efficient removal of organic compounds, suspended solids and color in textile wastewater, while reducing the hydraulic retention time and avoiding the use of decolorizing agents. Moreover, the treated water can be reused in new dyeing processes, which is beneficial for such big water-consuming industry.

As the specific conclusions related to each chapter:

In Chapter I, after giving an overview of the previous literature on MBR and MBBR applied in textile wastewater treatment, the conclusions from the literature review can be summarized as follows:

- MBR was found a well-developed technology in the treatment of textile effluent, it was effective in removal of organic compounds of textile wastewater, but in many cases, the combination with other technologies is needed to eliminate color from the water.
- MBBR process used in textile wastewater treatment showed that they can operate with high concentrations of biomass but with the need of extra coagulation for better decantation.
- MBBR-MBR can work at high organic loading rates and could reduce the energy consumption comparing with MBR. Few studies had investigated the application of MBBR-MBR in textile wastewater treatment. Therefore, development of the application of MBBR-MBR should be attractive to textile wastewater treatment as a reliable and effective method.

These conclusions lead us to a better understanding of the motivation and objective of the thesis.

In Chapter IV, three different biological methods: Conventional Activated Sludge (CAS) system, MBR, and MBBR, were compared in treating textile wastewater from a local industry. The main conclusions obtained during this investigation are presented below:

- Technically, MBR was the most efficient technology, of which the chemical oxygen demand (COD), total suspended solids (TSS), and color removal efficiency were 91%, 99.4%, and 80%, respectively, with a hydraulic retention time (HRT) of 1.3 days. MBBR, on the other hand, had a similar COD removal performance compared with CAS (82% vs. 83%) with halved HRT (1 day vs. 2 days) and 73% of TSS removed, while CAS had 66%.
- Economically, MBBR was a more attractive option for an industrial-scale plant since it saved 68.4% of the capital expenditures (CAPEX) and had the same operational expenditures (OPEX) as MBR.
- Environmentally, MBBR system had lower environmental impacts compared with CAS and MBR processes in the life cycle assessment (LCA) study, since it reduced the consumption of electricity and decolorizing agent with respect to CAS.
- According to the results of economic and LCA analyses, the water treated by the MBBR system was reused to make new dyeings. The quality of new dyed fabrics was within the acceptable limits of the textile industry.

In Chapter VI, the application of the hybrid MBBR-MBR system in textile wastewater treatment developed in this thesis allowed for the following conclusions:

- A great performance was observed during the MBBR-MBR treatment in COD removal (93%), color removal (to 85%), TSS (99 %).
- The hydraulic retention time (HRT) was 1 day during the MBBR-MBR operation which it used to be 2 days in industrial CAS treatment. The reduction of 50% in practical cases is promising that can result in space and energy saving.
- COD removal of 93% is almost the maximum for a biological process in this type of wastewater because the water is not completely biodegradable.
- 85% of color removed without a tertiary treatment is an attractive option for textile wastewater treatment from the perspective of economic and environmental impact. The results show that the hybrid MBBR-MBR system is an effective process for textile wastewater treatment.
- The treated water from MBBR-MBR process is 100% reusable for making new dyeings. If the textile industry installs an MBBR-MBR plant to treat all the wastewater after the production and wants to reuse all of the treated water to make the dyeing procedure a

circular process, it could reduce its consumption of tap water up to 70%.

In Chapter VII, the application of the hybrid MBBR-MBR system in textile wastewater treatment developed in this thesis allowed for the following conclusions:

- LCA study suggested that the hybrid MBBR-MBR system generated much lower environmental impact than CAS treatment, mainly because no extra decolorizing agent was used.
- The values of CAPEX and OPEX showed that MBBR-MBR had smaller expenses than CAS. Higher efficiency of MBBR-MBR system resulted in lower discharge tax and without cost for decolorizing agent.
- Additionally, in the NPV and IRR study, water reuse after the treatment played an important role leading to the cost saving of water consume and discharge tax. The 18% of IRR calculated demonstrated that MBBR-MBR has great economic feasibility for textile wastewater treatment on the industrial scale.

8.2. Recommendations

Based on the results obtained during the development of this thesis, the following lines of work are proposed for future research:

- Due to the limitation of the transportation and storage of the wastewater, the thickness (width) of the hybrid MBBR-MBR reactor was designed to be as small as possible without affecting the movement of carriers. If conditions permitted, future studies could increase the reactor thickness to see whether it had a positive impact on the carrier's movement by aeration.
- Also due to limitations in the transportation and storage of water, the treatment flow was not increased to the maximum flowrate that the membrane could withstand. The maximum working flow could be determined while maintaining the good performance and good functioning of the membranes and minimize HRT. It would be advisable to install the plant in the industry itself so as not to have water supply problems.
- Because of the complexity and the variability of textile wastewater depending on each textile industry, future studies could use different type of textile wastewater to verify the treating efficiency of the hybrid MBBR-MBR system.

- Future studies could try other types of carriers with higher specific surface area that allow higher concentrations of sludge.

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