

**DEGRADATION OF DYE WASTEWATER USING AEROBIC
MEMBRANE BIOREACTOR**

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ABSTRACT

Homemade textile industry is very famous in the East Coast of Peninsular Malaysia. Known locally as Batik Industries, they are traditionally inherited from generation to generation. The Batik Industry makes a big contribution to the economic growth due to high demands locally and from abroad. However, this industry produces wastewater which contributes to water pollution since it utilizes a lot of chemicals. Preliminary studies show that the wastewater from these homemade textile industries contains grease, wax, heavy metal, surfactant, suspended solid, and dyes (organic and inorganic). This study explores the use of aerobic membrane bioreactor processes to remove the suspended solid (mainly due to dyes in the painting and coloring processes) from wastewater of batik industry. Besides that, the effect of dye concentration (color), BOD and COD were studied based on it respect hydraulic retention time. Data on the flux and average values for each of the parameters studied were presented systematically. The results show that the dye concentration, BOD and COD the operating pressure was found to affect the process. The average flux was increased when the applied pressure was increased. The data collected could be used to improve the effectiveness of dye removal from the batik industry wastewater using membrane technology.

ABSTRAK

Industri pembuatan tekstil secara kecil-kecilan terkenal di pantai timur Semenanjung Malaysia. Ia dikenal oleh penduduk tempatan sebagai industri batik dan diwarisi secara turun-temurun. Industri batik banyak menyumbang kepada pembangunan ekonomi negara kerana sambutan yang menggalakkan bukan sahaja dari pasaran tempatan malah dari luar negara. Di sebalik perkembangan ini, industri batik turut menghasilkan air sisa yang mengakibatkan pencemaran air kerana prosesnya yang menggunakan bahan kimia yang banyak. Kajian awal terhadap air sisa industri batik menunjukkan bahawa ia mengandungi gris, minyak, logam berat, surfaktan, pepejal terampai, dan pewarna (organik dan bukan organik). Permasalahan ini telah mendorong penyelidik mengkaji penggunaan proses membran bagi penurasan mikro untuk menyingkirkan bahan pepejal terampai (terutama yang dihasilkan oleh proses pengecatan dan pewarnaan) daripada air sisa industri batik. Penyelidikan ini merungkai penggunaan proses membran bioreaktor aerobik untuk menyingkirkan bahan terampai terutamanya disebabkan oleh proses cantingan dan mewarna batik. Selain itu, kesan daripada kepekatan pewarna, BOD dan COD dikaji berdasarkan waktu retensi hidrolik. Data fluks dan nilai purata untuk setiap parameter yang dikaji ditunjukkan secara sistematik. Keputusan kajian menunjukkan bahawa kepekatan pewarna, dan tekanan operasi mempunyai kesan terhadap proses penapisan. Fluks purata didapati berkurang apabila waktu retensi hidrolik meningkat. Keputusan kajian menunjukkan bahawa fluks meningkat seiring dengan peningkatan tekanan. Data kajian boleh digunakan untuk meningkatkan keberkesanan penyingkiran pewarna yang terdapat dalam air sisa bagi industri batik dengan menggunakan teknologi membran bioreaktor.

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LIST OF ABBREVIATIONS

APHA	American Public Health Association
BOD	Biological Oxygen Demand
CH ₄	Methane
CO ₂	Carbon Dioxide
COD	Chemical Oxygen Demand
DO	Dissolved Oxygen
EM	Effective Microorganism
EQA	Environment Quality Act
HRT	Hydraulic Retention Time
MBR	Membrane Bioreactor
MLSS	Mixed Liquor Suspended Solid
ODI	Oxygen Demand Index
SEM	Scanning Electron Microscopy
SMEWW	Standard Method for Examination of Water and Wastewater
SS	Suspended Solid
UF	Ultrafiltration
UMP	University Malaysia Pahang

CHAPTER 1

INTRODUCTION

1.1 Title:

DEGRADATION OF DYE WASTEWATER USING AEROBIC MEMBRANE BIO-REACTOR

1.2 Synopsis

Wastewater reclamation and reuse is a great interest and a viable option for many industrial sectors and countries which suffer from water scarcity problems [36]. Current conventional treatment processes are often not sufficient enough to meet the requirements of wastewater reuse, even with stringent regulations. In recent years due to stringent government legislation and strict environmental regulation, dye degradation is a deeply researched topic worldwide. Both new and modified methods were proposed primarily aiming at effectively removing one or more recalcitrant dyes from synthetic or industrial wastewater. These methods include physicochemical, biological and advanced chemical oxidation treatment based strategies. [2,35] As a result, membrane technology is being considered as a reliable option for wastewater reclamation, since it can selectively eliminate physical, chemical, and microbiological contaminants from wastewater MF or ultrafiltration (UF) processes; however, appear to be attractive only for the separation of particulate contaminants. Therefore, by integrating different conventional treatment processes; by using membrane bioreactor, along with emerging membrane filtration technology can provide an efficient and yet cost effective treatment option for synthetic dye wastewaters. [35, 13]

1.3 Background of the Study

Wastewater from textile industries constitutes a threat to the environment in large parts of the world, as the degradation products of textile dyes are often carcinogenic. In addition, light absorption hindered by textile dyes creates problems to photosynthetic aquatic plants and algae. The main important pollutants in textile effluent are recalcitrant organic compounds, color, toxicant and inhibitory compounds, surfactants and chlorinated compounds. During processing, 5-20% of the used dyestuffs are released into the process water and dye is the most difficult constituent to treat by conventional biological wastewater treatment. In addition to their visual effect and their adverse impact in terms of chemical oxygen demand, many synthetic dyes are toxic, mutagenic and carcinogenic. The current existing techniques for the treatment of wastewater containing dyes have high cost, formation of hazardous by-products or intensive energy requirement. Therefore in order to get the desired throughput, a membrane bioreactor system can be introduced and the effectiveness of this system will be determined throughout this project.

A membrane is defined as a material that forms a thin wall capable of selectively resisting the transfer of different constituents of a fluid and thus effecting a separation, of the constituents. While, bioreactors are reactors that convert or produce materials using functions naturally endowed to living creatures. Reactors using immobilized enzymes, microorganisms, animal, or plant cells and those applying new methodologies such as genetic manipulation or cell fusion are typical bioreactors. Bioreactors differ from conventional reactors as living organisms present in the reactors operate under milder conditions of temperature and pressure. The ranges of operating conditions within bioreactors are usually determined by the biocatalyst (organism) and are usually small. In general, Membrane Bioreactor (MBR) systems essentially consists of combination of membrane and biological reactor systems. These systems are the emerging technologies, currently developed for a variety of advanced wastewater treatment processes.

1.4 Problem Statement

Homemade textile industry is very famous in East Coast of Peninsular Malaysia. Known locally as Batik Industries, they are traditionally inherited from generation to generation. The Batik Industry makes a big contribution to the economic growth due to high demands locally and from abroad. However, this industry produces wastewater which contributes to water pollution since it utilizes a lot of chemicals. Preliminary studies show that the wastewater from these homemade textile industries contains grease, wax, heavy metal, surfactant, suspended solid, and dyes (organic and inorganic).

Wastewaters from printing and dyeing units in textile plant are often rich in color, containing residual of dyes and chemical, and needs proper treatment before releasing into the environment [9]. The color of textile effluent is unacceptable under Malaysian Environmental Regulation besides the other parameter such as Chemical Oxygen Demand (COD), Biological Oxygen Demand (BOD), total iron, etc [36]. Due to these factors, the textile industry faces the challenge of balancing the environmental protection, its economic viability and sustainable development. There is an urgent need to find a way to preserve the environment while keeping the economic growing.

Several innovative treatment technologies have been developed and applied in order to treat textile effluent. The most widely used treatment systems are conventional activated sludge. This system poorly removes the widely used dyes, and is clearly ineffective in decolorizing textile effluent, even when mixed and treated together with sewage. Activated Carbon is the most commonly used and most successful adsorbent. However, it is expensive and the level of color removal depends on the dye type. Also, 100% color removal is rarely achieved. Ozone has been shown to have the ability to breakdown most dyes. However, even high doses of ozone do not completely mineralize the organic dye to carbon dioxide and water due to the decolourisation rate which is decreasing with increasing initial dye color [6]. Treatment of textile effluent nowadays requires a sound and efficient system in facing the current challenges. Membrane technology could be one of such promising technology which able to treat textile effluent in a more beneficial way. Although

filtration techniques require an initial high setup cost, it is outweighed by the significant cost saving achieved through reuse of permeate

1.5 Objectives of the Study

1. To characterize batik waste water before and after treatment process.
2. To determine the effect of Hydraulic Retention Time (HRT) of batik waste water quality.
3. To determine the efficiency of the dye degradation by using aerobic membrane bioreactor.

1.6 Scope of Study

1. The dye wastewater sample will be collected in textile industry which is batik wastewater.
2. The HRT will be varies to 24, 48 and 72 hours.
3. The study area will be restricted by East Coast of Peninsular Malaysia, known as batik industries region.
4. The parameters that will be observed are Biological Oxygen Demand (BOD), Chemical Oxygen Demand (COD), Color and Suspended Solid (SS).

1.7 Significance of Study

Dye in batik wastewater contains high concentration of pollutants such as high color, carcinogenic dyes and toxic heavy metals. Due to low cost operation required as well as high selectivity achieved, separation using membrane technology has becoming promising alternatives to treat colored wastewater. This project is worth to study since it can give contribution to existing data on water quality in study area for the future research. Besides, this process can establish the best water purification technique and clarify the batik wastewater to be reuse or treat.

CHAPTER 2

LITERATURE REVIEW

2.1 Wastewater

Wastewater is the spent water after homes, commercial establishments, industries, public institutions, and similar entities have used their waters for various purposes. It is synonymous with sewage, although sewage is more general term that refers to any polluted water (including wastewater), which may contain organic and inorganic substances, industrial wastes, groundwater that happens to infiltrate and to mix with the contaminated water, storm runoff and other similar liquids. Certain sewage may not be a spent water or wastewater.

The keyword in the definition of wastewater is “used” or “spent.” That is the water has been used or spent and now it has become wastewater. On the other hand, to become sewage, it is enough that water becomes polluted whether or not it had been used. When one uses the word wastewater, however, the meaning of the two words is blended such that they now often mean the same thing.

The term "wastewater" is a broad, descriptive term. Generally it includes liquids and waterborne solids from domestic, industrial or commercial

uses as well as other waters that have been used (or "fouled") in man's activities, whose quality has been degraded, and which are discharged to a sewage system. The term "sewage" has been used for many years and generally refers to waters containing only sanitary wastes. However, "sewage" technically denotes any wastewaters which pass through a sewer. Two general categories of wastewaters, not entirely separable, are recognized: domestic wastewaters and industrial wastewaters. The details will be separately described in below section.

2.1.1 Domestic Wastewater

Domestic wastewaters originate principally from domestic, household activities but will usually include waters discharged from commercial and business buildings and institutions as well as ground water. Surface and storm waters may also be present. Domestic wastewaters are usually of a predictable quality and quantity.

2.1.2 Industrial Wastewater

Industrial wastewater is wastewater that results from industrial processes and manufacturing. It may either be disposed of separately or become part of the sanitary or combined sewage. Industrial wastewaters, on the other hand, originate from manufacturing processes, are usually of a more variable character, and are often more difficult to treat than domestic wastes. Industrial wastewaters are the discharges of industrial plants and manufacturing processes. It can represent, collectively, an important part of community wastewaters and must be considered for successful wastewater treatment plant operation. In some locations industrial wastewater discharges are collected together with other community wastewaters and the mixed wastes are treated together. In other instances, the industry may provide some pretreatment or partial treatment of its wastewaters prior to discharge to the municipal sewers. In still other situations, the volume and character of the industrial waste is such that separate collection and disposal is necessary.

Industrial wastewaters vary widely in composition, strength, flow and volume, depending on the specific industry or manufacturing establishment in the community. The specific composition and volume of the industrial waste will, of course, depend on the use to which the water has been put. Typical industries which produce significant volumes of wastewaters include paper and fiber plants, steel mills, refining and petrochemical operations, chemical and fertilizer plants, meat packers and poultry processors, vegetable and fruit packing operations and many more. Industrial discharges may consist of very strong organic wastewaters with a high oxygen demand, or contain undesirable chemicals which can damage sewers and other structures. They may contain compounds which resist biological degradation or toxic components which interfere with satisfactory operation of the wastewater treatment plant. Less obvious source which must be considered an industrial waste; is thermal discharge since it lowers dissolved oxygen values. Many industries use large quantities of cooling water, with the electric power industry being the largest user. However, the primary metal and chemical industry also use substantial quantities of cooling waters.

2.2 Wastewater Characteristics

Wastewater is composed of many materials that are broken down into three general areas. These areas are the physical, chemical, and biological characteristics of wastewater. [3]

2.2.1 Physical characteristic

The physical characteristics of wastewater include those items that can be detected using the physical senses. They are temperature, color, odor, turbidity, taste, and solids. And those of the constituents which discussed in turn in the below section are based on the parameter that will be determined.

i. Color

Color is the perception registered as radiant of various wavelengths strikes the retina of eyes.

ii. Odor

Odors in wastewater usually are caused by gases produced by the decomposition of organic matter or by other substances added to the wastewater. Fresh domestic wastewater has a musty odor. If the wastewater is allowed to go septic, this odor will significantly change to a rotten egg odor associated with the production of hydrogen sulfide (H₂S).

iii. Solid

Wastewater is normally 99.9 percent water and 0.1 percent solids. If a wastewater sample is evaporated, the solids remaining are called total solids. The amount of solids in the drinking water system has a significant effect on the total solids concentration in the raw sewage. Industrial and domestic discharges also add solids to the plant influent. The most common types are dissolved, suspended, settleable, floatable, colloidal, organic, and inorganic solids. [3]

iv. Temperature

The temperature of wastewater is commonly higher than that of the water supply because of the addition of warm water from households and industrial plants. However, significant amounts of infiltration or storm water flow can cause major temperature fluctuations.

2.2.2 Chemical Characteristic

The chemical characteristics of wastewater of special concern to the Utilitiesman are pH, DO (dissolved oxygen), oxygen demand, nutrients, and toxic substances. [3]

i. Oxygen Demand

Oxygen demand is the amount of oxygen used by bacteria and other wastewater organisms as they feed upon the organic solids in the wastewater. Chemical tests such as the BOD (biochemical oxygen demand), the COD (chemical oxygen demand), the ODI (instantaneous oxygen demand or oxygen demand index), and the TOC (total organic carbon) measure the “strength” of wastewater. [3]

ii. Biological Oxygen Demand (BOD)

BOD is a measure of the oxygen used by microorganisms to decompose this waste. If there is a large quantity of organic waste in the water supply, there will also be a lot of bacteria present working to decompose this waste. When BOD levels are high, dissolved oxygen (DO) levels decrease because the oxygen that is available in the water is being consumed by the bacteria. [16]

iii. Chemical Oxygen Demand (COD)

Chemical oxygen demand (COD) is a measure of the capacity of water to consume oxygen during the decomposition of organic matter and the oxidation of inorganic chemicals such as ammonia and nitrite. [17] Another term that refers to COD is a measure of organic materials in a wastewater in terms of the oxygen required to oxidize the organic materials chemically. [28]

iv. pH

This is a method of expressing the acid condition of the wastewater. pH is expressed on a scale of 1 to 14. For proper treatment, wastewater pH should normally be in the range of 6.5 to 9.0 (ideally 6.5 to 8.0).

2.2.3 Biological characteristic

Bacteria, viruses, and parasites make up the biological characteristics of wastewater. Wastewater contains vast quantities of bacteria and other organisms that originate in discharged wastes. The feeding activities of these organisms assist in decomposing wastewater. Aerobic bacteria decompose organic matter in the presence of free oxygen. Anaerobic bacteria decompose organic matter that is shut off from free oxygen, such as in the interior of a mass of feces or a dead body. The products of anaerobic decomposition have an extremely unpleasant odor. Matter in which this condition exists is said to be septic. A large number of the bacteria in wastewater are coliform bacteria - those found in the digestive tract of normal humans. While most of these bacteria are harmless, pathogens will usually be present in wastewater containing the discharges of many persons. It is these relatively few pathogenic organisms that pose the greatest public health hazard. Wastewater that is not properly treated may eventually find its way into a community water source and spread waterborne diseases.

2.3 BATIK

Since the project is to treat the batik wastewater, this subchapter will brief about the batik history, the process of batik production and all. By definition, batik is a cloth which traditionally uses a manual wax-resist dyeing technique. Due to modern advances in the textile industry, the term has been extended to include fabrics which incorporate traditional batik patterns even if they are not produced using the wax-resist dyeing techniques. [18]

2.3.1 Process of Making Batik

Homemade textile industry is very well known in Malaysia especially in the East Coast of Peninsular Malaysia and Sarawak. This industry is traditionally inherited from generation to generation. High skills coupled with the right equipments and tools are needed in order to produce high quality of batik. Today, this industry has become very commercialized and contributed positively to the economic growth for some states such as Kelantan and Terengganu. Moreover, this industry has become the main attraction of foreign and local tourists to visit these states apart from other attractions such as an Islamic ruled government and many more. [24] Batik process consists of several processes before the final outcome look attractive and eye catching. Generally, the batik painting can be categorized to three processes including cloth preparation, dyeing and de-waxing. [19] The detail will be discussed in the lower section.

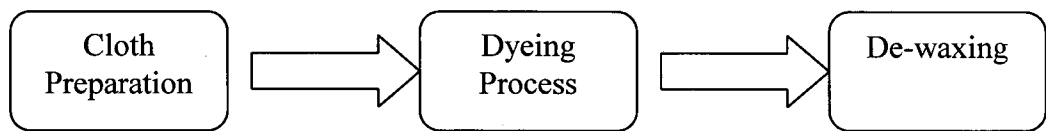


Figure 2.1: diagram of batik process

2.3.1.1 Waxing

The first step includes giving the cotton cloth a thorough washing and to remove the starch. The cloth is then dried, ironed and stretched on a wooden frame. The next step includes making a rough charcoal sketch which serves as a guide for the painting. The design develops gradually with each coat of waxing. The next step is a pretty lengthy one. It includes the application of the wax that is a protective agent in the dyeing process. The wax is a melted mixture of paraffin, resin and yellow beeswax and is poured into a brass shaped pen called Tjanting. The Tjanting is a brass pen shaped is like a small cup with single or double spout. This pen is used in coloring the negative way

by filling the space where the painter does not want the color to appear. The process is quite tedious and has to be done with care. Thus for each and every different color to be used on the fabric the painter has to use the Tjanting pen and the protective wax ink. [19]

2.3.1.2 Dyeing Process

After the first process is done, dyeing process can be applied based on required dyes to color the area that have been sketched. When that area has been adequately colored and dried, the fabric can be gently washed with a special liquid and followed by lukewarm water. This process of waxing, dyeing and de-waxing can be repeated multiple times till the entire batik design or fabric has been completed. Ideally, this process should begin with light colors and end with dark colors. [19]

2.3.1.3 De-waxing

After that, the painter completes this process for each and every color that he has used; he can wash the waxed piece in boiling water. This process can be repeated often till the entire wax has been removed from the fabric. This can be followed by application of colors or dyes to the painting using a brush. In case of very large areas, the cloth is removed from the frame and dipped in a basin of dye. The final stage includes the removal of wax residue and the fixing of colors, immersing the cloth into boiling water with a small amount of detergent. The cloth is then rinsed in cold water. The complete painting is then dipped in a diluted solution of sulphuric acid to give the colors permanency. Several handmade Batik tapestries are made of various kinds of hand woven threads to give it a specific quality. This art form is three-dimensional and has features of depth and texture making the entire fabric an attractive piece of art. [19]

2.4 Textile Wastewater, Characteristics and Environmental Impact

Normal textile dyeing and finishing operations are such that the dyestuffs used can vary from day to day and sometimes even several times a day because of the batch wise nature of the processes. Frequent changes of dyestuff employed in the dyeing process and finishing process cause considerable variation in the waste water characteristic particularly the pH, color and COD. Strong color is another important component of the textile wastewater which is very difficult to deal with (Lin and Chen, 1997). Color is noticed in the wastewater effluent and the presence of small concentrations of dyes in water is highly visible, and may affect their transparency and aesthetics (Banat et al., 1996).

Dye wastewater from textile industry is a serious pollution problem because it is high in both color and organic content. A dye is a colored substance that can be applied in solution or dispersion to a substrate in textile manufacturing, thus giving a color appearance to textile materials. Discharging of dyes into water resources even in a small amount can affect the aquatic life and food web. Dyes can also cause allergic dermatitis and skin irritation. One of the main problem regarding textile wastewaters is the colored effluent. The colored effluent contains visible pollutants. The primary concern about effluent color is not only its toxicity but also its undesirable aesthetic impact on receiving waters. Non-biodegradable nature of most of the dyes reducing aquatic diversity by blocking the passage of sunlight through the water represents serious problems to the environment.

In some cases, dyes in low concentration are harmful to aquatic life. Since many dyes have adverse effect on human beings, the removal of color from the effluent or process has appeared of importance for ensuring healthy environment. Hence, it is imperative that a suitable treatment method should be applied. The color of the effluent discharges into receiving waters affects the aquatic flora and fauna and causes many water borne diseases. Some of dyes are carcinogen and others after transformation or degradation yield

compound such as aromatic amines, which may be carcinogenic or otherwise toxic. In addition, dyes accumulate in sediments at many sites, especially at locations of wastewater discharge, which has an impact on the ecological balance in the aquatic system. These pollutants, because of leaching from soil, also affect the groundwater system (Namasivayam and Sumithra, 2005).

Textile finishing wastewater, especially dye house effluents, contain different classes of organic dyes, chemicals, and auxiliaries; thus, they are colored and have extreme pH, COD, and BOD values and they contain different salts, surfactants, heavy metals, mineral oils, and others (Golob et al., 2005). A composite wastewater from an integrated textile plant consists of the following materials: starches, dextrin, gums, glucose, waxes, pectin, alcohol, fatty acids, acetic acid, soap, detergents, sodium hydroxide, carbonates, sulfides, sulfites, chlorides, dyes, pigments, carboxymethyl cellulose, gelatin, peroxides, silicones, flourcarbons, resins, etc. Textile wastewater is known to exhibit strong color, a large amount of suspended solids, high fluctuating pH, high temperature, and high COD concentration.

Reactive dyes in both ordinary and hydrolyzed form are not easily biodegradable and thus even after treatment, color may be present in the effluent. The conventional processes such as coagulation, flocculation, and biological methods adopted for decoloration of effluent containing reactive dyes are no longer able to achieve adequate color removal (Santhy and Selvapathy, 2006).

The discharge of organic pollutants, either BOD or COD, to the receiving stream can lead to the depletion of dissolved oxygen and thus creates anaerobic conditions (Al-Degs et al., 2000). Under anaerobic conditions, foul-smelling compounds such as hydrogen sulfides may be produced. This will consequently upset the biological activity in the receiving stream.

Textile effluent contains dyestuffs, which are visible, even at low concentrations (Prado et al., 2004). These colored effluents are aesthetically