

# PERFORMANCE CHANGES OF AEROBIC-ANOXIC MEMBRANE BIOREACTOR FOR AZO DYE BIODEGRADATION UNDER DIFFERENT HYDRAULIC RETENTION TIME IN ANOXIC TANK

<sup>1</sup>Puti Sri Komala, <sup>2</sup>Agus Jatnika Effendi, <sup>3</sup>IG. Wenten, <sup>2</sup>Wisjnuaprpto

<sup>1</sup>Department of Environmental Engineering, Andalas University  
Kampus Limau Manis Universitas Andalas, Padang 25163

<sup>2</sup>Department of Environmental Engineering, Institute Technology Bandung

<sup>3</sup>Department of Chemical Engineering, Institute Technology Bandung  
Jalan Ganesha 10, Bandung 40132

Corresponding author: [putisrikomala@ft.unand.ac.id](mailto:putisrikomala@ft.unand.ac.id)

## ABSTRACT

This research investigated the changes in anoxic-oxic membrane bioreactor (AOMBR) performance caused by various hydraulic retention time (HRT) of anoxic tank for Remazol Black-5 azo dye biodegradation. Bioreactor consists of anoxic tank coupled with modified activated sludge process, contact stabilization process combined with an external ultrafiltration membrane. Feed was a mixture of Remazol Black-5 at a concentration of 110-120 mg / L and tempe industry wastewater. The hydraulic retention time of anoxic tank was varied in 3, 3½, 4, 4½, 5, 5½ and 6 hours respectively at constant HRT of contact-, and stabilization tanks ie 2, and 4 hours. The optimum color- and COD removal were obtained at 4 h HRT of the anoxic tank i.e. 77% and 76% on 2 days solid retention time (SRT). The anoxic tank contributed to the highest color- and COD removal of the entirely removal, followed by the stabilization tank. Membrane contributed significant only in color removal of AOMBR performance due to its anoxic microenvironment inside the flocs attached to the membrane. The aerobic condition in contact and stabilization tanks led to high organics removal used microorganisms growth.

**KEY WORDS:** Aerobic-Anoxic Membrane Bioreactor (AOMBR), azo dye, Hydraulic Retention Time (HRT), biodegradation

## 1. INTRODUCTION

One of the largest components in textile industry wastewater are dyes. Among of these dyes, azo dyes are the most widely used in the textile industry. The release of these compounds into the environment is undesirable, not only because of their color, but also because many azo dyes and their breakdown products are toxic or even carcinogenic (Van der Zee, 2002).

Dye decolorization process required an external carbon source as co substrate (Padmavathy et al. (2003). This is also confirmed by Pandey et al. (2007) that decolorization process required either simple organic compounds such as glucose and starch or complex organic compounds such as yeast extract and peptone, or

a combination of organic sources and complex carbohydrates. While Khehra et al. (2005) stated that the yeast extract was one of the most effective supplements to increase the color removal efficiency. However the yeast extract cost is very expensive, especially if used in a continuous experiment or applied in industrial scale. Therefore, it is required less expensive substitutes but with similar content. Wisjnuaprpto et al. (1999) and Wahyuni et al. (2002) used tempe industrial wastewater as co-substrates for azo dye decolorization and shown high color removal efficiency. Tempe is one of the traditional Indonesian food made from fermented soybeans. In addition to cheap tempe also contains highly nutritious. Tempe industrial wastewater easily obtained and

contains organic compounds such as, proteins, vitamins and trace minerals with high concentrations (Sudaryanto, 1998 and Arta, 1999) that are required for microorganisms growth in biological treatment.

Due to its recalcitrant nature, azo dye treatment under conventional activated sludge is difficult. But if aerobic microorganisms are subjected to micro-aerophilic condition they are decolorizing at a faster rate Padmavathy et.al. (2003). Therefore azo dyes treatment in biological processes is generally performed in anaerobic/microaerophilic-aerobic combination.

Wisjnuprpto et al. (1999) used a modified contact-stabilization process for CIRO16 azo dye removal using tempe industry wastewater as co-substrate. Stabilization tank that normally operated in aerobic in this experiment it stirred without aeration, so the condition in the reactor become anaerobic. This modification result in high color removal.

The experiment continued by Ananthi (2008) for remazol black 5 (RB5) treatment using addition of anoxic tank which is placed before the contact stabilization process and combined with an external ultrafiltration membrane replacing sedimentation tank, the reactor was called anoxic-oxic membrane bioreactor (AO-MBR). The AO MBR operated at anoxic tank HRT range of 0.5 to 3.5 hours with 0.5 hours increment under constant HRTs contact and stabilization tanks i.e. 2 hours and 4 hours. At a HRT 3 h in anoxic tank under SRT less than a day, a more extensive reductive color biodegradation was observed for RB5 (86%). In this study the HRT in anoxic tank will be extended from previously to optimize die decolorization without affecting microorganisms growth.

The purpose of this study was to evaluate the performance of AOMBR under varied HRT of anoxic tank using aerobic microorganisms. The effect of various HRT of anoxic tank on microorganisms growth, color- and COD removal both in each tanks and membrane were investigated.

## 2. METHODS

### 2.1 Microorganisms

A mixed culture was developed from mixed liquor drawn from a textile and dye industry activated sludge treatment plant. The culture was fed with a mixture of tempe waste water and azo dye then aerated in batch. This culture is used for the continuous experiment.

### 2.2 Co-substrate and dye

Tempe industrial wastewater derived from soybean boiling, was used as co-substrate. Tempe industrial wastewater has a high content of organic carbon and nutrient required for microbial growth. From the previous research (Komala, 2010) obtained, that the optimum co-substrate concentrations ranging from 2.080-2.400 mg COD/L tempe wastewater to the total solution. The dye used in this study was Remazol Black-5 (RB5) reactive azo dye obtained from Dystar Bandung. RB5 with a concentration of 120 mg/l has a wave length of 609 nm.

### 2.3 Anoxic-oxic Membrane Bioreactor

The bioreactor made from acrylic material, consists of an anoxic tank, contact tank and stabilization tank connected to an external ultrafiltration membrane, which was located between the contact- and stabilization tanks (Figure 1).

The feed, a mixture of Remazol Black-5 azo dye and co-substrate tempe industry wastewater was pumped into the anoxic reactor at a rate of 2 l/hour through a screen in the suction section of the pump. The anoxic tank was equipped with a 40 rpm motorized mixer to mix the mixed liquor in the reactor, afterwards the mixed liquor flowed by gravity into the contact tank. At the bottom of the contact- and stabilization tanks a diffuser was mounted to supply air and agitate the liquor. The mixed liquor was then pumped through the membrane with a pressure in range of 0.4-1 bar, generating the permeate as the filtration result and the retentate was concentrated biomass which flowed in the membrane lumen then discharged into the stabilization tank. Biomass from the stabilization tank was recirculated to the anoxic tank and mixed with the incoming feed. Ultrafiltration hollow fiber membrane made from polysulfone (PS) membrane module with 20 kDa MWCO and 1 m<sup>2</sup> surface area was used.

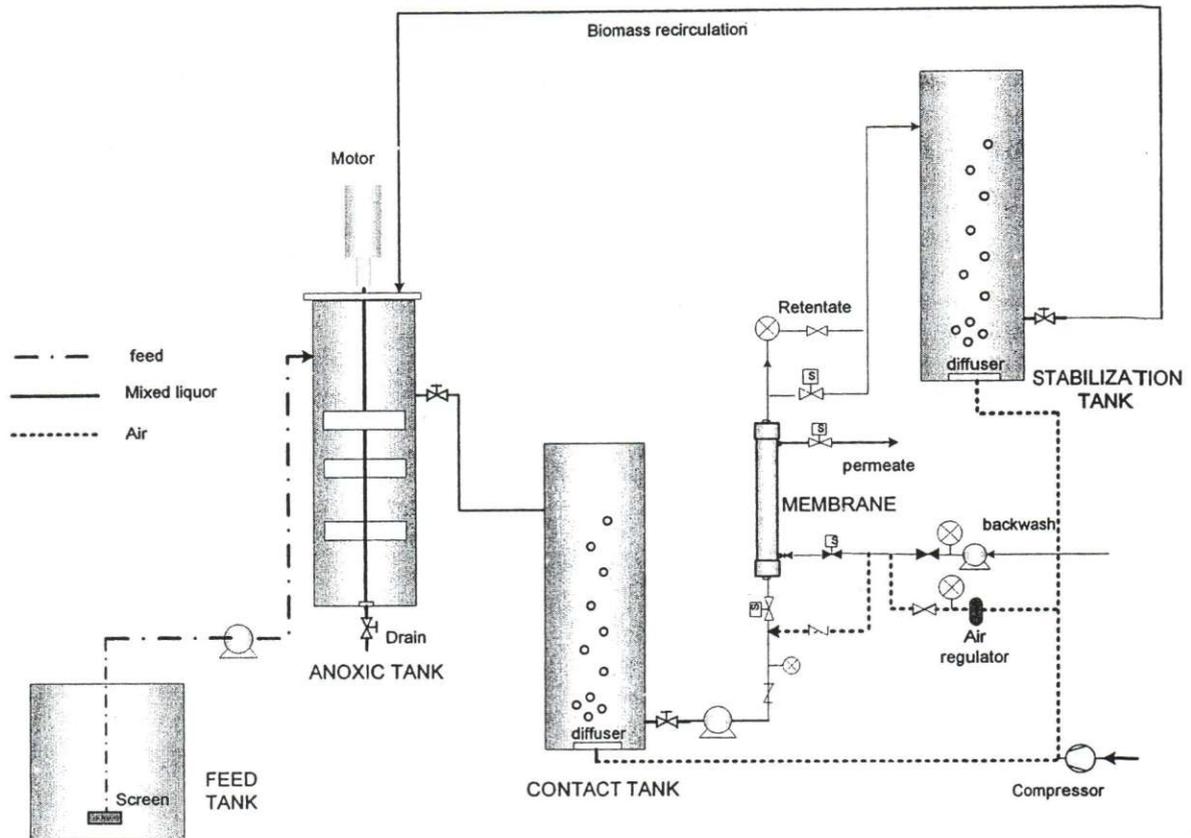


Figure 1. Scheme of Aerobic-Anaerobic Membrane Bioreactor set-up

Membrane operated in crossflow, air is supplied into the membrane feed and adjusted using an air regulator located at the pressure gauge allowed aeration pressure to be controlled. Periodically backwash water flowed in the opposite direction of the normal flow, in out-in mode from shell to lumen. Backwash intervals were controlled by the timer and solenoid valves. An adjustment valve was mounted each at retentate- and permeate line to regulate the flow and release the pressure. This valve was set, so that the retentate pressure was not too high which resulted in small retentate stream and large permeate flow.

The excess sludge was wasted daily from the stabilization tank by taking into account the amount of suspended solids in the permeate as well. Amount of sludge waste removed based on the desired SRT in bioreactor as expressed in Eqs. 1. The bioreactor was operated at different HRT of anoxic tank in range of 3 h to 6 h HRT with half hour increment under constant HRT of

contact- and tabilization tanks i.e. 2 h and 4 h respectively. Dye concentration of feed, anoxic, contact-, and stabilization tanks and membrane permeate daily observed. The steady state condition is achieved characterized by the constant COD and color concentration for three times measurement consecutively.

$$SRT = \frac{X_a V_a + X_k V_k + X_s V_s}{Q_w X_s + (Q - Q_w) X_e} \quad (1)$$

where:

SRT = solid retention time (day)

Q = influen flow (L/h)

$X_a$  = biomass concentration in anoxic tank (mg/L)

$V_a$  = anoxic tank volume (L)

$X_k$  = biomass concentration in contact tank (mg/L)

$V_k$  = contact tank volume (L)

$X_s$  = biomass concentration in stabilization tank (mg/L)

$V_s$  = stabilization tank volume (L)

$X_e$  = biomass concentration in permeate ( mg/L )

$Q_w$  = sludge waste ( L/h )

#### 2.4. Analytical methods

Samples from the feed, anoxic, contacts, stabilization tanks, and membrane permeate were collected daily for laboratory measurement. Samples were centrifuged, and then the supernatant was measured for COD by closed reflux method and colors with UV-vis spectrophotometer, while the filtrates for VSS measurement by gravimetry method. The measurement method used in accordance with the Standard Method of Examination of Water and Waste (APHA, 1995).

### 3. RESULTS

It is well known that the anoxic conditions are the expected condition where the decolorization lasted. Based on this reason, every half hour HRT increase of the tank ranging from 3 hours to 6 hours was observed. Color and COD removal efficiency at different HRT are shown in Figure 2 and 3, while the growth of biomass in each tank during the experiment were presented in Figure 4.

The maximum color removal was achieved at 5.5 h HRT anoxic tank i.e. 80% but COD removal efficiency was only 43%. Whereas at 4 h HRT maximum COD removal 77% was obtained, while color removal was 76%. At HRT higher than 4 h despite the higher color removal obtained, otherwise the lower COD removal. Biomass growth in entire reactors reached the largest concentration at 4 h HRT in the anoxic tank, at higher HRT the biomass tend to decrease. This indicated that most of aerobic bacteria died, due to lack of dissolved oxygen in anoxic reactor under extend HRT so that the reactor become anaerobic. The survived facultative bacteria decolorized dyes better in anaerobic condition, but its ability to reduce organics compounds smaller than aerobic microorganisms. The optimum color- and COD removal was decided at 4 h HRT in anoxic tank by considering that the color removal was quiet high while the biomass growth was not hampered by the HRT in the reactor. Of the entire reactors, the anoxic tank had the largest contribution on color removal i.e. 66% followed

by membrane (28%) and stabilization tank (22%), while color removal in the contact tank was not too significant (5%) (Figure 5).

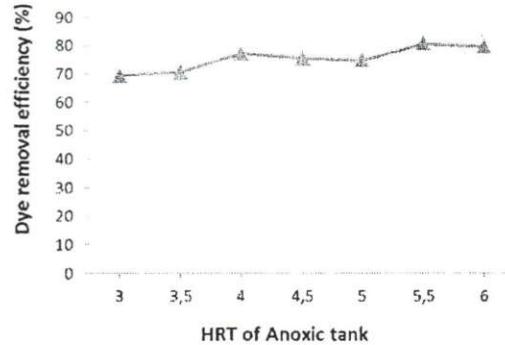


Figure 2. Dye removal efficiency vs HRT in anoxic tank (at HRT in contact stabilization 2 and 4 hrs)

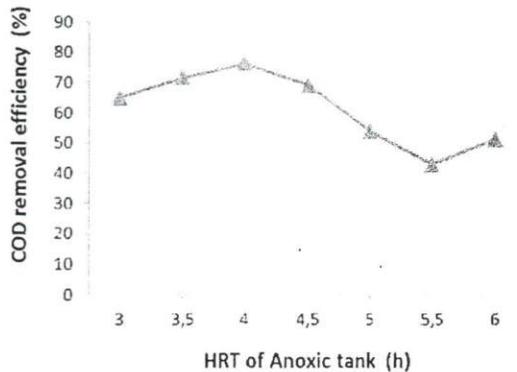


Figure 3. COD removal efficiency vs HRT in anoxic tank (at HRTs of contact stabilization 2 and 4 hrs)

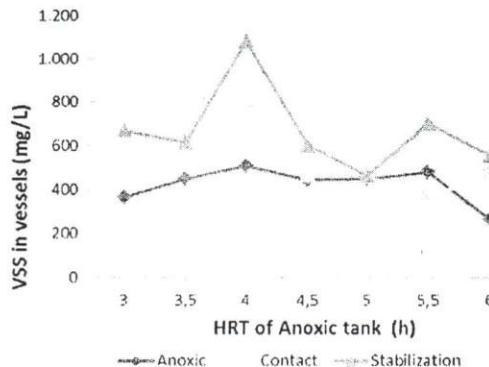
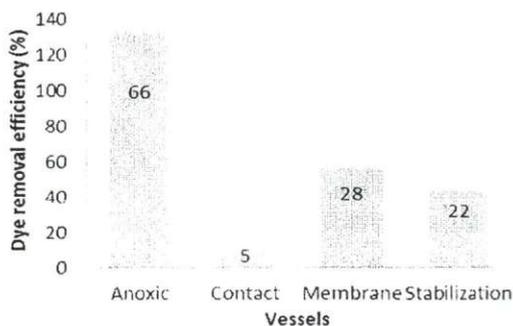
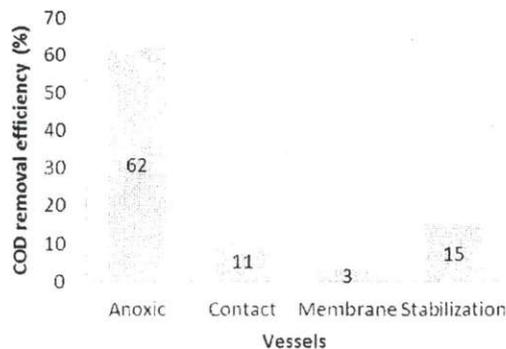


Figure 4. Biomass concentration in each tanks vs HRT in anoxic tank (at HRTs of contact stabilization 2 and 4 hrs)

The anoxic tank also played significant role to organics removal i.e. 62% then the stabilization tank (15%), followed by the contact tank (11%) and the latter was the membrane (3%) (Figure 6). It was confirmed by Padmavaty (2003), that aerobic microorganisms decolorize at a faster rate in micro-aerophilic conditions. It occurred both in the anoxic tank and in the membrane. Membrane played a role in color removal of AOMBR performance. There was transition from aerobic to anaerobic condition in membrane resulted in decreased DO. This also revealed by Gong et al. (2007) that aerobic and anoxic condition could occur as a consequence of DO concentration gradients within microbial flocs or biofilm attached to the membrane. Based on this, anoxic microenvironments could develop inside the flocs/biofilm, whereas aerobic condition prevails on the outer surface layer. The anoxic condition in the membrane led to azo chromophore cleavage.



**Figure 5.** Contribution of the reactors on Dye removal (at HRTs of anoxic, contact stabilization 4, 2 and 4 hrs respectively)



**Figure 6.** Contribution of the reactors on COD removal (at HRTs of anoxic, contact stabilization 4, 2 and 4 hrs respectively)

The organics source was required in decolorization process, so the organics removal significantly took place in anoxic tank. Similarly, organics reduction although not significantly occurred in membrane to support the dye decolorization. Incoming flow into the membrane was divided to two streams namely retentate and permeate. So, the organics removal was also separated in these both sections, small part of which occurred in the permeate. Both aerobic and facultative microorganisms lived rather in aerobic condition than anaerobic environment, in this situation organics substance was used as electron donor while oxygen acted as electron acceptor rather than dye. Therefore color removal was not occurred in the contact-tanks. In the stabilization tank, concentrated biomass existed here allowed to significant organics removal. Also anoxic microenvironment developed between the flocs led to color removal. This condition led to simultaneously carboneously- and color removal in the stabilization tank.

The bioreactor was operated at low SRT (two days) which resulted color- and COD removal 77% and 76%. It suggested to study decolorization by AOMBR at the higher SRT. The high SRT known to increase the decolorization and COD removal (Sponza and Işik, 2002; Lorenzo et al., 2001).

#### 4. CONCLUSIONS

The results of this study showed that azo dye RB5 with addition of textile industry wastewater as co-substrate could be treated effectively by AOMBR at different anoxic HRTs varying between 3 and 6 h. Color removal efficiencies tend to increase with increasing anoxic HRT from 76% to 80%, on the contrary COD removal decrease with increasing HRT from 77% to 43%. This showed that increasing HRT changed the reactor environment becomes anaerobic, led to most of aerobic microorganisms died. Despite the remaining facultative bacteria could decolorized color better, however their ability to reduce organics were smaller. So, the optimum color- and COD removal were obtained at 4 h HRT of the anoxic tank i.e. 77% and 76% on 2 days solid retention time (SRT). Anoxic tank contributed to the highest color- and COD removal of the entirely

removal, followed by the stabilization tank. Membrane contributed significant only in color removal of AOMBR performance due to anoxic microenvironment inside the flocs attached to the membrane. The aerobic condition in contact and stabilization tanks led to higher organics removal for both aerobic and facultative microorganisms.

#### ACKNOWLEDGEMENTS

This study was supported by the Research Group Grant 2010 from Institute Technology Bandung (ITB), Indonesia.

#### REFERENCES

- American Public Health Association. (1995). *Standard Methods for the Examination of Water and Wastewater*. A.D. Eaton, L.S. Clesceri, A.E.Greenberg (Eds.), 19th ed., Washington D.C.
- Ananthi, N. (2008). *Pengaruh waktu detensi hidrolis dalam tangki anoxic terhadap kinerja rangkaian bioreaktor membran kontak stabilisasi dalam penyisihan zat warna azo Remazol Black V*. Master Thesis, Department of Environmental Engineering, Institute Technology Bandung.
- Arta, W. (1999). *Kinerja bioreaktor kontinu modifikasi kontak stabilisasi dalam penyisihan zat warna CIRO16*. Tugas Akhir S-1 Program Studi Teknik Lingkungan Institut Teknologi Bandung.
- Gong, Z., Yang, F., Liu, S., Bao, H., Hu, S., and Furukawa, K. (2007). Feasibility of a membrane-aerated biofilm reactor to achieve single-stage autotrophic nitrogen removal based on Anammox. *Chemosphere*, 69, 776–784.
- Khehra, M.S., Saini, H.S., Sharma, D.K., Chadha, B.S. and Chimni, S.S. (2005). Decolorization of various azo dyes by bacterial consortia. *Dyes and Pigments*, 67, 55–61.
- Komala, P.S., Effendi, A.J., Wenten, I.G., and Wisjnuaprpto. (2010). *Perbandingan Ko-substrat Optimum pada Sistem Batch dan Bioreaktor Membran Anoksik-oksik Kontinu dalam Biodegradasi Zat Warna Azo*. Seminar Teknik Kimia Soehadi Reksowardojo, Bandung, 25-26 Oktober 2010.
- Lorenço, N. D., Novais, J. M., and Pinheiro, H. M. (2001), Effect of Some Operational Parameters on Textile Dye Biodegradation in a Sequential Batch Reactor, *Journal of Biotechnology*, 89, 163-174.
- Padmavathy, S., Sandhya, S., Swaminathan, K., Subrahmanyam, Y.V., and Kaul, S.N. (2003). Comparison of decolorization of reactive azo dyes by microorganisms isolated from various sources. *Journal of Environmental Science (China)*, 15 (5), 628–633.
- Pandey, A., Singh, P., and Iyengar, L. (2007). Review Bacterial decolorization and degradation of azo dyes. *International Biodeterioration & Biodegradation*, 59, 73–84.
- Sponza, D.T., and Işik, M. (2002) Decolorization and azo dye degradation by anaerobic/aerobic sequential process. *Enzyme and Microbial Technology*, 31, 102–110.
- Sudarjanto, G. (1998). *Penyisihan zat warna CIRO 16 serta kinetiknya menggunakan kombinasi proses kontak stabilisasi dan adsorpsi karbon aktif*. Tesis S-2. Teknik Lingkungan ITB.
- Van der Zee, F.P. (2002). *Anaerobic azo dye reduction*. Doctoral Thesis. Wageningen University, 142 pages.
- Wahyuni, S., Mubiarti, E., and Yunita, R. (2002). Pengukuran gas pada pengolahan limbah warna CIRB 5 dengan konsentrasi 60 mg/L dalam tahapan reaksi *sequencing batch biofilm reactor* (SBBR). *Infomatek*, 5 (4), 153-160.
- Wisjnuaprpto, Kardena, E. and Artha, W. (1999). Penyisihan zat warna azo CIRO-16 dalam modifikasi proses kontak-stabilisasi menggunakan limbah cair industri tempe. *Jurnal Biosains*, 4 (1), 26-30.