

**Research Article**

## OPTIMIZATION THE PERFORMANCE OF MULTI-STAGE BNR REACTORS BASED ON MBBR REACTOR FOR REMOVAL OF NITROGEN AND PHOSPHORUS

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### ABSTRACT

Nowadays, rural, urban and different industries' wastewaters should be treated to the best way possible to prevent sanitary and environmental dangers. Bio processes is an environment friend more effective than physical and chemical processes since: 1. Lower expenses 2. No chemical sludge production and less sludge produced 3. No limitation for denitrification process 4. Better quality of formed sludge from biological removal and possible use as fertilize. The purpose of this research is to discuss optimized biological treatment of nitrogen and phosphor which are two kinds of dangerous environment which we need them for running biological system. In first step, multi-stages systems based on Moving Bed Biofilm Reactor is discussed and then a system of optimum condition is selected and detailed design is done. The origin for designed system for treatment wastewater is A<sup>2</sup>O method, since this method is able to remove total nitrogen and phosphor well and also from economic view and environmental view, we don't need more complex methods to remove more of these two pollutants.

**Keywords:** Biotreatment, BNR Multistage Methods, BioFilm Moving Bed Reactors

### INTRODUCTION

Organic and non-organic Nitrogen are in different forms of chemicals and oxides in environment. Some of these materials are protein, ammonia, nitrite, nitrate and nitrogen gas. Table 1 shows different forms of nitrogen.

**Table 1: Shows different forms of nitrogen[ Ergas and Aponte-Morales, 2014]**

Composition	Formula	Capacity
Organic nitrogen	Org-N	-3
Ammonium / ammonia	NH <sub>3</sub> /NH <sub>4</sub> <sup>+</sup>	-3
Total nitrogen	TKN	-3
Nitrogen gas	N <sub>2</sub>	0
Nitrous oxide	N <sub>2</sub> O	+1
Nitric Oxide	NO	+2
Nitrite	NO <sub>2</sub> <sup>-</sup>	+3
Nitrogen dioxide	NO <sub>2</sub>	+4
Nitrate	NO <sub>3</sub> <sup>-</sup>	+5

Sources of nitrogen in wastewater are diverse. These resources lead to introduction of nitrogen to pure water and Potable water. Origins of this nitrogen are outflows of contaminated tanks, water flows of agricultural irrigation, residual of chloramine from disinfection and sterilization processes. Municipal wastewaters enter large amounts of nitrogen to wastewater, 60-70 percent of entered nitrogen is ammonia nitrogen and 30-40 percent is organic nitrogen. Industrial wastewaters specially chemical industries which produce Nitrogen-containing chemical substances like fertilizer, nitric acid and ammonia are important resources of nitrogen. Other industries which produce nitrogen entering to wastewaters are paper production plants and food industry. In addition refineries enter nitrogen to wastewater but its amount is low compared to other industries but still need to remove nitrogen and phosphorous is maintained because

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for start-up biological system in refinery we need to add phosphor and nitrogen and finally this added nitrogen and phosphor which has been used as feed to micro-organisms should be treated in order to remove its additional because nitrogen and phosphor are assumed as environmental pollutants. Entering sources of phosphor to wastewater consists of: 1. Different industries like refineries. 2. Non point water flows 3. Detergent mixtures which include phosphor 4. Phosphor-containing fertilizer

### Phosphor and Nitrogen's Harmful Effects

#### Effects on Water

The main problems which nitrogen and phosphor cause in water is eutrophication in which water is enriched with nutritious materials and growth of alga and aquatic plants increases. Quality of water decreases. This phenomenon affects beauty of sea and leads to death of sea animals. Determinative nutritious substance in sea waters is nitrogen and limiting nutrition in alga growth in fresh water is phosphors. Sun light reduced into deep water which resulted in the death of aquatic plants. With eradication of sea plants carbon and energy needed for bacteria growth is provided so oxygen concentration decreases and because of the decrease of dissolved oxygen in solution, anoxic areas increase which lead to high death rate of sea animals. Additional nitrogen increases the expense of water treatment and also acid production in fresh water ecosystem. Nitrogen in from of nitrate and nitrite and ammonia can be too toxic and dangerous so that the ability of sea animals to growth and reproduction is impaired. Eutrophication happens only in lakes, stabilization pools and sometimes in rivers with low velocity [United States Environmental Protection Agency, 2007]. Eutrophication is also named as algal bloom. Moreover, high growth of algae which covers the surface of water, causes taste and smell originated from algae and limits access to water. Figure 1 show the eutrophication phenomenon.

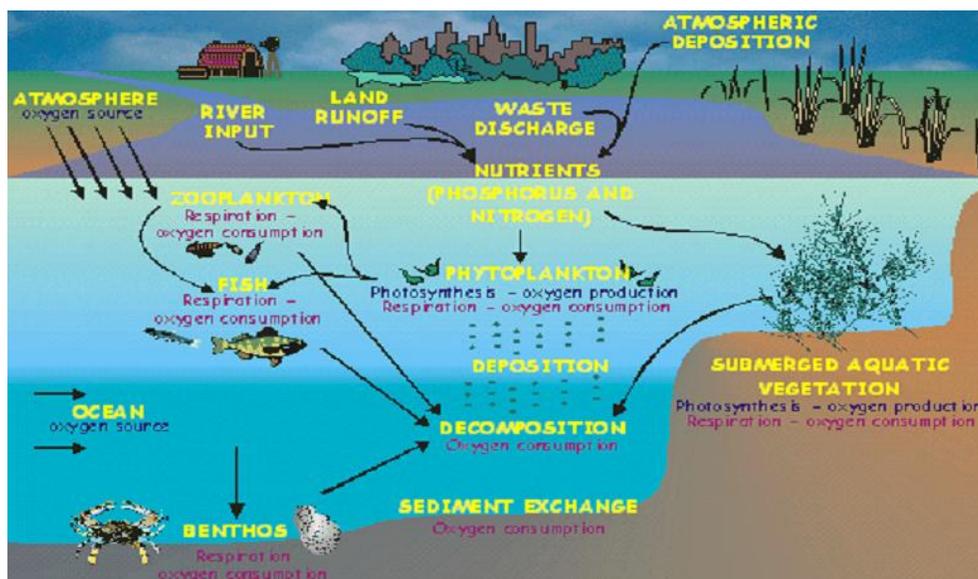


Figure 1: Show the eutrophication phenomenon

#### Effects on Human Health

Presence of nitrogen (nitrate) in potable water threatens human health. Nitrate, oxides iron available in blood's hemoglobin and produce metmogolobines (combination of hemoglobin and oxygen). In some people, body again converts metmoglobine to hemoglobin but children cannot perform this process fast and this phenomenon cause inability of their blood to carry oxygen and skin disease appears in children. For human health guaranty, United States Environmental Protection Agency announces maximum allowed amount of nitrate and nitrite in drinking water as 10 mg/L and 1 mg/L respectively (Ergas and Aponte-Morales, 2014). To prevent these kind of cases to happen, the amount of nitrogen and phosphor in wastewater treatment units should be limited. Conventional Secondary treatment processes are not able

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total nitrogen and phosphorus removal within permissible limit for discharge the water, In these cases, we should use BNR (Biological Nutritious Removal) system. This system removal nitrogen and phosphor by use of micro-organisms in different biological conditions. Now, there are several BNR processes some of which is only used for total nitrogen removal and some of them only are to removal total phosphor and some of them are designed to removal both. Appropriate process is selected according to the quality of wastewater outflow, analysis operator, quality of entering wastewater and available treatment processes (if treatment performance enhance is intended). Different forms of BNR processes change according to sequence of living conditions (aerobic, anaerobic and anoxic) and time.

### Background

Nitrate removal first was discussed in 19<sup>th</sup> century. But after invention of active sludge by Ardene and Locket in 1914 highly changed (Arden and Locket, 1914). One of the original and extensive works on nitrification and Denitrification was performed by Sawyer and bradney in which they discuss the problem of sludge increase. Also great work of duning in 1964 showed that residence time of solid substance or more sludge is needed not only for *Nitrosomonas* bacteria growth rate but also for prevention of autotrophs (Downinng *et al.*, 1964). Verman in 1962 stated that the construction of Denitrification region after nitrification in a sludge system for Denitrification of high organic bases. Ludzak in 1962 introduced the process of active aerobic sludge which has been simulated in a channel system.

In this method active sludge first enters the aerobic part and then enters to a channel name semi aerobic part. Mixture of liquids with air which includes nitrate in this part of semi aerobic operation once again enters aerobic system. According to work of showing in 1974 and Parker in 1975 for separation of nitrogen with low concentration, these methods are not suitable and one 3-stage system is used. In the first stage, organic carbon is separated and nitrification intensity in the second stage increases and in the third stage methanol is added for denitrification. In 1969 Mccarthy proposed that for removal of nitrates from water a anaerobic filter with methanol is used and results showed that for nitrate reduction, methanol is needed, In 1971 nitrogen removal by using extensive aerobic increases from 20% to 80%. Bernard in 1973 utilized anaerobic filter designed by Mccarthy. He looked for ways to reduce nitrate and examined the methods purposed by Rishnan and Eknefelder and solved nitrate in second stage liquid was used in first stage for direct use of entering carbon and their removal in final clarifier. Then residual nitrate in final clarifier was removed and returns solved nitrate in liquid to the anaerobic stage. Process designed by Rishnan and Aknfelder is called MLE method (James *et al.*, 1973). In 1970 Luin suggested phostrip method in which active sludge in a digestive ferment and then phosphor is released. Upper surface full of phosphor is treated by lime and chemical sediments are separated. According to Milbury report in 1967 all of the treatment unit in which phosphor removal is by plug flow and ... Denitrification is used, phosphor releases in the entrance region of anaerobic region has been observed (Milbury *et al.*, 1967). In 1974, Bernard observed the removal of phosphor from 8 mg/lit to less than 0.2 mg/ lit in 6 weeks in a pilot unit.

In the research time, nitrogen separation process had 4 stages in which phosphor releases in about 30 mg/lit in the second stage of anoxic happens and only during this release, phosphor separates well. If this special condition changes good phosphor separation is not obtained (James *et al.*, 1973). Bernard observed that a common feature in the treatment, release of phosphorus, So it is obvious that regions devoid of oxygen and nitrate(anaerobic) after on aerobic stage, in order to get biologic phosphor separation in high amount is needed.

### Process Types OF BNR System

Conventional Processes for this method include:

1. MLE process for total nitrogen removal (Continuous flow with the initial anoxic and next step aerobics)
2. A<sup>2</sup>O process for total nitrogen and phosphor removal (A anaerobic stage following by MLE process)
3. Step feeding for total nitrogen removal (changing anoxic and aerobic stage)
4. Four-stage Bardenpho process for total nitrogen removal(Continuous flow with 4 step anoxic/aerobic/anoxic/aerobic)

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5. Modified Bardenpho process for total nitrogen and phosphor removal
6. University of Cape Town process or Virginia Initiative Process for total nitrogen and phosphor removal.
7. Modified University of Cape Town Process for total nitrogen and phosphor removal(A<sup>2</sup>O process with a secondary anoxic process in which internal reflux of nitrate is performed)
8. Rotating Biologic contacts for total nitrogen removal(Continuous Process with a consequential stage of anoxic/aerobic)
9. Oxidation ditch for total nitrogen and phosphor removal (Continuous flow by using annular channel in order to make anoxic, aerobic and anaerobic in a chronological form)
10. Phoredox process or A/O for total nitrogen omission
11. Johannesburg(JHB) for total nitrogen and phosphor omission
12. Biological Chemical Flexible System(BCFS) for total nitrogen and phosphor omission
13. Phostrip process for total nitrogen omission

Figure 2 to 12 show some of these processes.

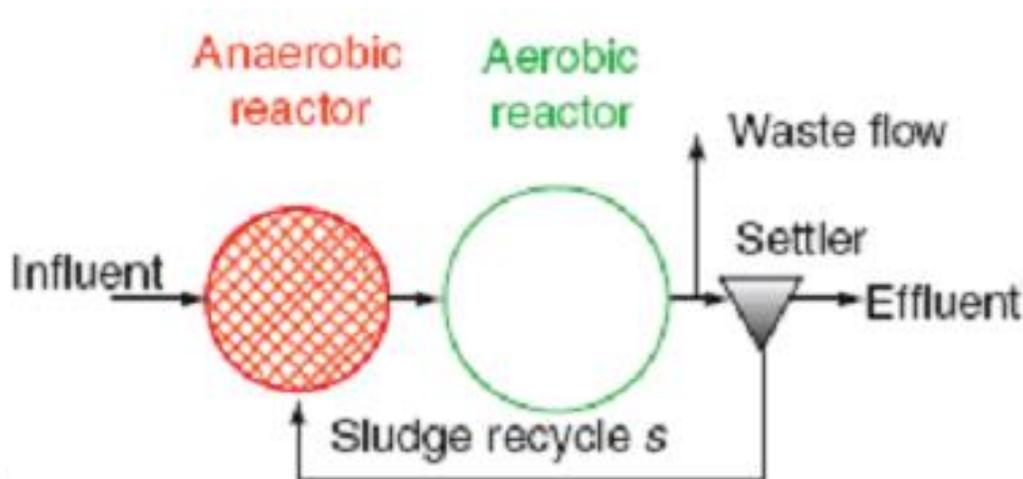


Figure 2: A/O process (Xiaoxia et al., 2014)

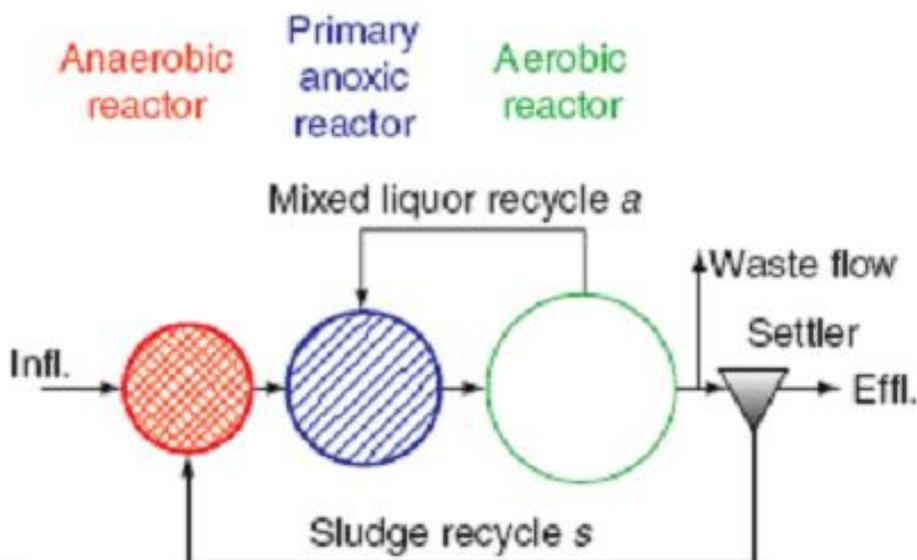


Figure 3: A2O process [Wei Zeng et al., 2011]

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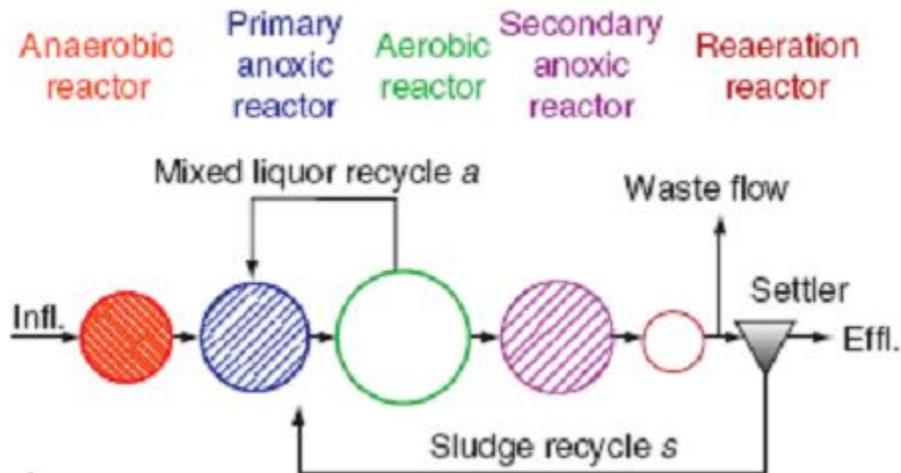


Figure 4: Bardenpho process [Georgine Grissop PE, 2010]

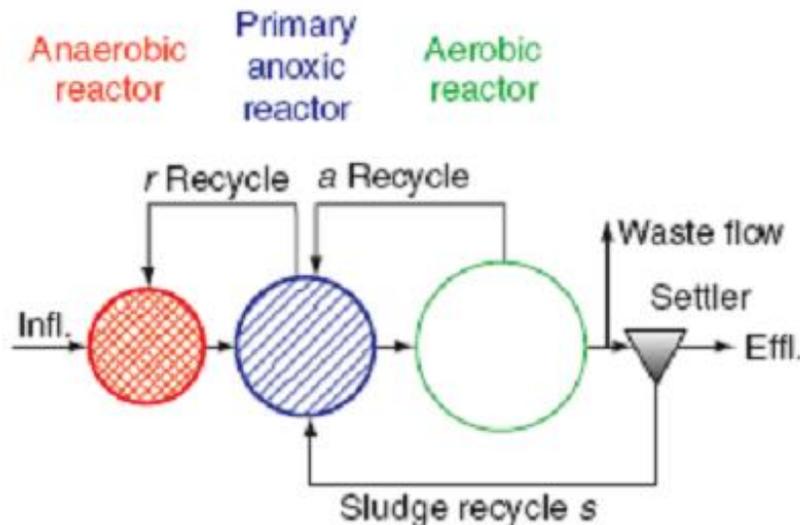


Figure 5: University of Cape Town(UCT) [Ekama, 2011]

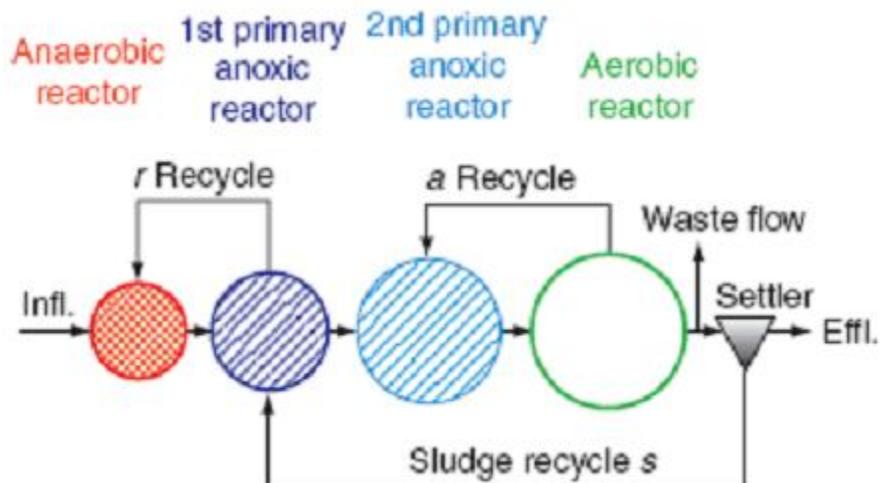
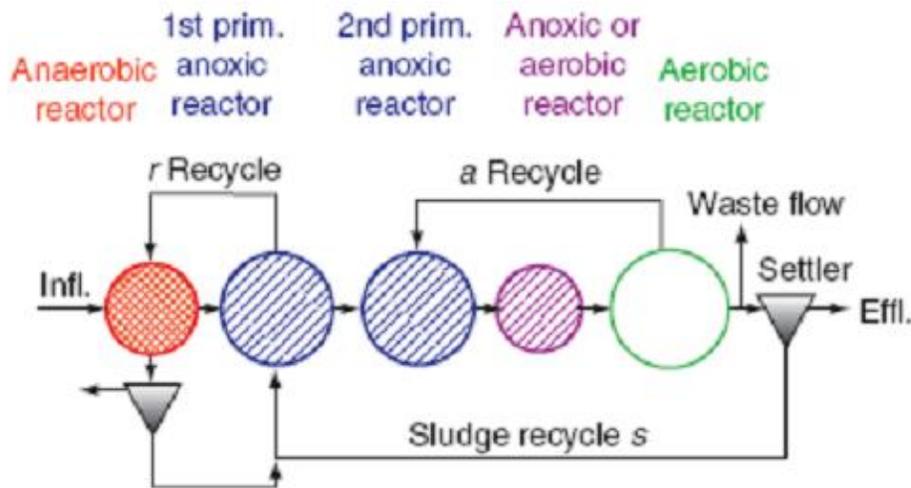


Figure 6: Modified University of Cape Town(UCT) [Ekama, 2011]

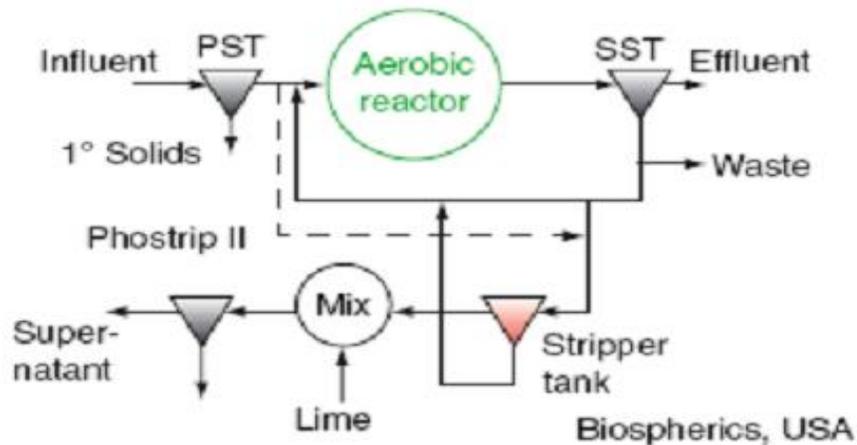
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**Figure 7: Johannesburg (JHB)[Ekama, 2011]**

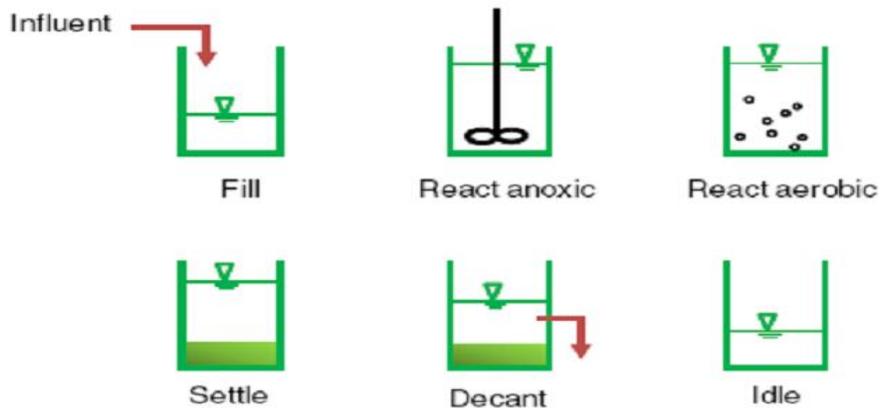


**Figure 8: Biological Chemical Flexible System (BCFS)[Ekama, 2011]**

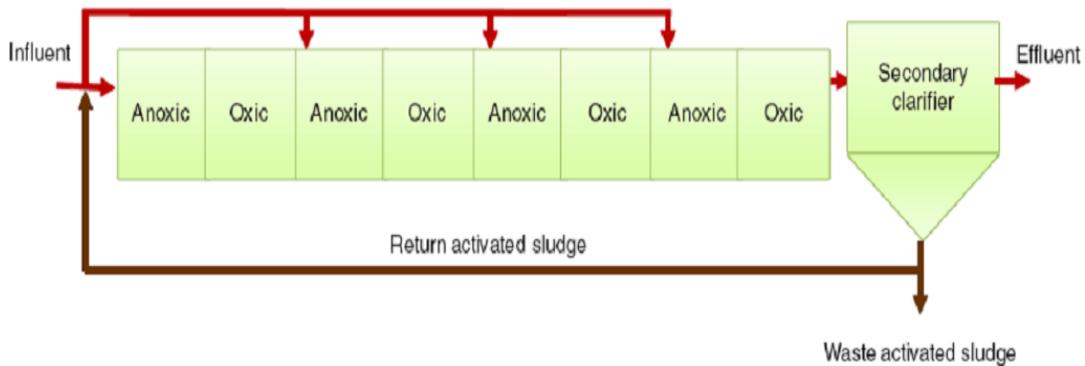


**Figure 9: Phostrip [Ekama, 2011]**

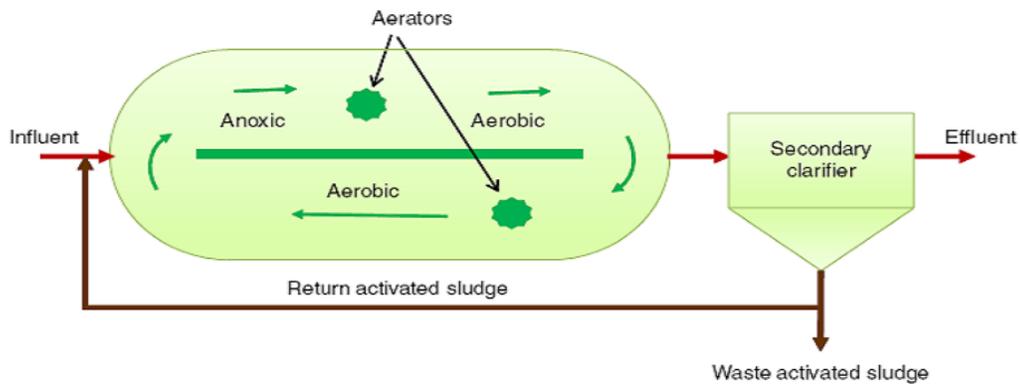
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**Figure 10: Sequencing Batch Reactors (SBR) (Ergas and Aponte-Morales, 2014)**



**Figure 11: Step Feed [Ergas and Aponte-Morales, 2014]**



**Figure 12: Oxidation Ditch[Ergas and Aponte-Morales, 2014]**

Systems designed for total nitrogen removal should have an aerobic stage for nitrification and an anoxic stage for Denitrification and those designed for total phosphor removal a anaerobic stage which is devoid of dissolved oxygen and nitrogen is needed. If low extent of nitrogen and phosphor is intended, a sand bed for filtering and particle removal should be used.

More suitable system design is based on responses to the following questions:

1. What is the final concentration?
2. For reaching strict limits for final nitrogen and phosphor concentration. Should we design systems from basis or we can upgrade previous systems with new modern systems?

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New designs are more flexible and we can choose the BNR applicable to our use. Because it is not limited with available treatment unit and sludge maintenance process.

#### **System Performance**

All of the mentioned processes are related to active sludge system. Active sludge process is the most conventional suspended growth process for municipal wastewater's treatment. In suspended growth process, micro organisms are responsible for treating the mixture fluid. A lot of Suspended growth processes used for municipal and industrial wastewater treatment are proper concentration of dissolved oxygen utilization (aerobic) but High concentrations of suspended organic matter used anaerobic reactions (Moshtagh, 2010).

#### **Active Sludge System Limitations**

Active sludge system has some limitations some of which are (Sheng *et al.*, 2008):

1. Low load
2. Sensibility to low temperatures and toxic materials
3. Shortage of active biomass
4. Need of more appliances for accumulating sludge
5. Unstability to Against shock
5. Unstability to flow fluctuation
6. Treating more of active sludge

Because of mentioned limitations, nowadays lots of researchers are attracted to biofilm systems like trickling filters, submerged aerobic biological filters. In stucked growth process, micro organisms are expected to convert organic material or nutritious substance to a bed of inert materials. Organic materials and nutritious material from wastewater flow which move through a biofilm and deleted. Used material in bed includes stones, sand, wood, plastics and other artificial materials. Sticked growth can be used aerobic or anaerobic. Bed material can be submerged in liquid. One biofilm, is a frame of microb bulk which is used widely in nature and biotechnology.

#### **Bio Film System Advantages**

Biofilms advantages include:

1. Active biomass with higher concentration due to the presence of sticky and static micro organisms
2. Lower side effects for nitrification in lower temperature
3. Smaller system in comparison with active sludge system
4. Less problems originated from Ridge sludge
5. Resistance to Against shock
6. Most of the COD load
7. Sohulat behbud tas hilat
8. Lower need of the system to cleaning and back washing
9. Decreasing the string bacteria which impedes suitable sedimentation
10. More flexibility in PH changes and nutrient and toxic concentrations in comparison with membrane systems (which have both constant and sticky micro organisms) (Jennifer *et al.*, 2012)

#### **MBBR (Moving Bed Biofilm Reactor) Systems Characteristics**

MBBR is the most efficient biological treatment. It is based on traditional active sludge and developed fluidized bed reactor. One biofilm reactor is fully mixed with a continuous flow which bio mass on small carriers (their density is a little lower than water density so that they can be suspended in reactor and can be carried in water flow) grow. Floatation of carriers can be due to Aeration in aerobic reactor and in anoxic and anaerobic reactor by mechanical mixers.

Some of the characteristics of MBBR systems are as follows (Ashrafi, 2010)

1. High resistance Resistance to Against High tolerance organic load
2. Small size for process design
3. More flexibility in process design
4. More flexibility in reactor design
5. Low residence time

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6. Low hydrolic loss
7. Low sensibility to low temperature and temperature change tolerance
8. No accumulation of sludge and ease of maintenance
9. Compatibility for decomposition of the substrate that hardy decomposes.
10. Fully mixed hydrolic regime

**Nitrogen and Phosphor Remove**

*Nitrogen*

Biologic processes to remove nitrogen are nitrification and denitrification. In nitrification, ammonia is oxidated to nitrite by autotroph bacteria. Then nitrite is converted to nitrate by other bacteria called *Nitrobacter*. Both autotroph and heterotroph bacteria are capable of Denitrification. Most conventional bacteria in Denitrification are pseudomonas which utilize hydrogen, methanol, carbohydrates, organic acids, alcohols, benzoates and other organic combinations in this process.

Nitrification happens in aerobic environment  $\text{NH}_4^+ \rightarrow \text{NO}_2^- \rightarrow \text{NO}_3^-$

Denitrification happens in anoxic environment.  $\text{NO}_3^- \rightarrow \text{NO}_2^- \rightarrow \text{NO} \rightarrow \text{N}_2\text{O} \rightarrow \text{N}_2$

In BNR systems, nitrification are controlling reactions, because strict considerations are needed for bacteria growth and these bacteria which oxidize ammonia are sensitive to environment conditions. Nitrification solely can not removal nitrogen from waste water. So Denitrification for converting oxidized nitrogen (nitrate) to nitrogen gas is needed. Nitrification is in presence of oxygen in aerobic condition and Denitrification is in devoid of oxygen and happens in anoxic conditions.

**Phosphor**

Biological removal of phosphor is based on its consumption by aerobic heterotroph bacteria that are capable of saving orthophosphate more than needed for their growth. Treatment process can be designed so as to fasten growth of phosphate gathering organisms. First orthophosphate is accumulated and poly phosphate is formed. Then under anaerobic condition, it is accumulated by degradation of poly phosphates and by using obtained energy organic materials are converted to carbon substances which are called poly hydroxyl alkanate. By breaking poly phosphates and PHA production, phosphor is released. In aerobic condition, PAO uses produced PAH as energy to consume phosphor released in anaerobic conditions. (PAO use oxygen to oxidize PAH and consume phosphor. They can also use nitrate instead of oxygen, this way phosphor is removal in anoxic zone instead of aerobic zone.)

Phosphor can also be deleted by using chemical sedimentation. Chemical sedimentation is mainly performed by using coagulants like aluminum and iron or lime to form bulks of phosphor. Chemical removal in comparison with biological removal is more expensive and produces more sludge and leads to higher levels of chemical in sludge.

For decreasing total phosphor concentration to 0.1 mg/lit a combination of biological and chemical processes from economical point of view, can be cheaper than solely process (United States Environmental Protection Agency, 2007).

**Research Methods**

First we compare BNR systems with each other, and then we choose our system. Performance comparisons of different BNR processes in nitrogen and phosphor removal are in table 2.

**Table 2: Performance comparison of different BNR processes in nitrogen and phosphor removal**

Process	Nitrogen removal	Phosphorus removal
MLE	Good	-
A <sup>2</sup> O	Good	Good
Step Feed	Average	-
Bardenpho	Excellent	-
Modified Bardenpho	Excellent	Good
Sequencing Batch Reactors(SBR)	Average	-
Univercity of Cape Town(UCT)	Good	Excellent
Oxidation Ditch	Excellent	Good

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We should choose our system based on table 2. Since we choose this system for oil refinery design and respect to complexity and expenses for construction and needs to be cost effective in energy use point of view, we choose  $A^2O$  system.

This system can removal total nitrogen and phosphor well, also from economical point of view and environmental regulations we don't need more complex method for further removal of these two dangerous pollutants.

By combination of MLE and A/O processes,  $A^2O$  process is obtained which can removal both nitrogen and phosphor. The process is described in figure 13.

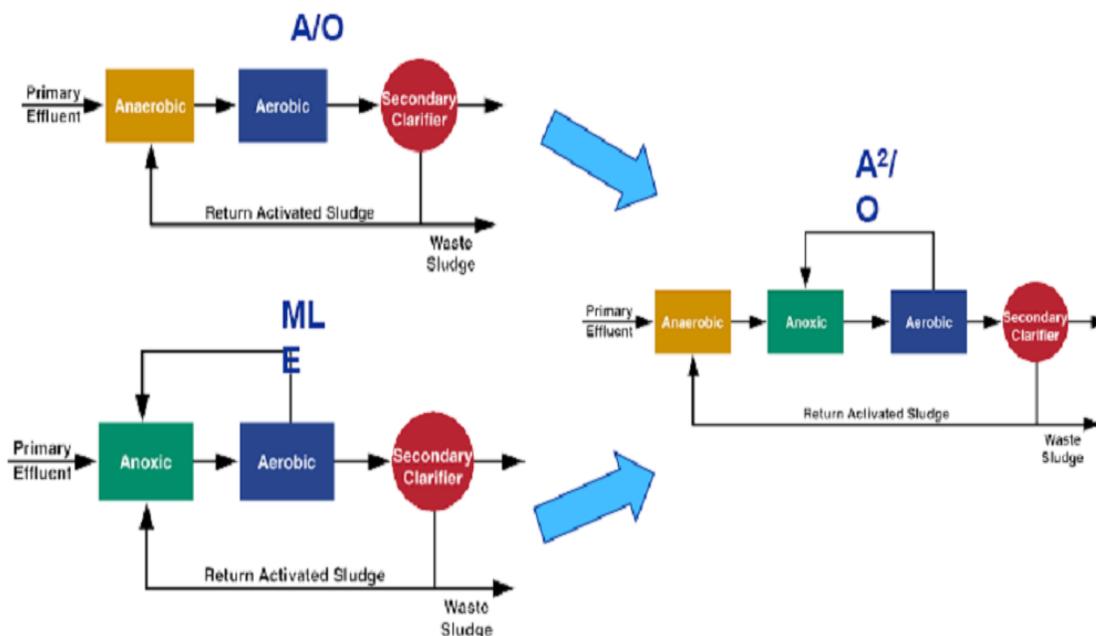


Figure 13:  $A^2O$  process [Georgine Grissop PE,2010]

In  $A^2O$  system, anoxic stage is before aerobic stage, mixture containing nitrate returns from anoxic stage to aerobic stage.

In these systems, total removal of nitrogen is impossible. Conversely, systems in which anoxic stage is after aerobic stage, reflux of mixture is not necessary, but because of decrease in organic substrate in aerobic reactor, Denitrification is not performed and we need external source for carbon which enters system and cause extra expenses. Also added carbon should be controlled precisely that COD concentration in outflow does not increase.

For this reason, interest to find new solutions to provide carbon source for these systems and effective consumption of carbon is obtained which can decrease operational expenses and widely used for waste water treatment system is used (Gang *et al.*, 2013).

### MATERIALS AND METHODS

Parsian refinery (A refinery located in south of Fars, Iran) is discussed.

Total biologic removal of phosphor and nitrogen from waste water treatment is the problem of all refineries. Parsian refinery waste water is measured in one year and wastewater analysis is as follow in table 3.

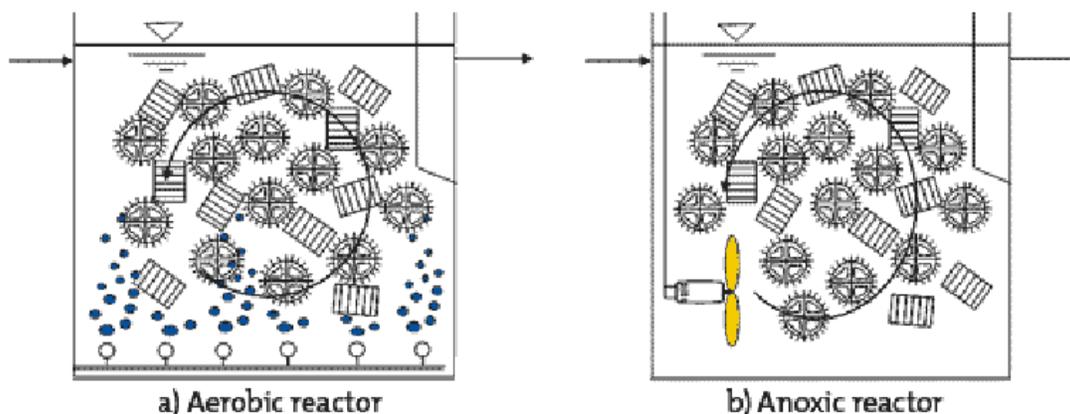
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**Table 3: Parsian refinery waste water is measured in one year and wastewater analysis**

property	Sampling									Unit
	7-6-2010	1-8-2010	25-10-2010	15-11-2010	6-12-2010	27-12-2010	17-1-2011	22-5-2011	19-8-2011	
<b>PH</b>	7.4	6.2	6.5	6.0	6.4	6.2	6.7	5.7	7.4	-
<b>Turbidity</b>	123	332	-	326	260	258	240	-	63	NTU
<b>TDS</b>	120	16760	11700	11465	6700	12886	22814	1350	3450	mg/l
<b>Cl<sup>-</sup></b>	646	9446	6019	5161	3004	6482	9260	710	1401	mg/l
<b>Na<sup>+</sup></b>	418	6120	3900	3344	1946	4200	6000	-	-	mg/l
<b>COD</b>	504	1310	827	747	767	823	2150	684	388	mg/l
<b>BOD<sub>5</sub></b>	220	640	-	-	94	88	213	354	185	mg/l
<b>Oil and Grease</b>	-	-	14	19	59	<10	23	-	-	mg/l
<b>Benzene</b>	-	-	-	-	7	29	-	-	-	mg/l
<b>Toluene</b>	-	-	-	-	5.8	26	-	-	-	mg/l
<b>Xylenes</b>	-	-	-	-	1.8	14	-	-	-	mg/l

Other research has shown that refinery’s waste water has dangerous pollutants like phenols and we should perform biological treatments. For this refinery we should enter phosphor and nitrogen manually at the first place. This is for feeding to micro organisms and their correspondence to the environment. And finally we should removal nitrogen and phosphor which are dangerous environmental pollutants. For this purpose we should pay attention to these condition for this process:

1. An artificial feed is used for this research
2. In feed urea is as ammonia source and  $(NH_4)_3PO_4$  is a source for nitrogen and phosphor.
3. Carbon source is BTX
4. MBBR is used (Figure 14)



**Figure 14: MBBR system in Aerobic and Anoxic [Nakhli, 2011]**

5. Bioreactor is made of glass and their volumes are 15, 7.5 and 7.5 lit for aerobic, anaerobic and anoxic reactor respectively.
6. Sludge is from parsian refinery

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7. Sludge volume is about 5 percent of active volume of each reactor.
8. Sludge reflux is available ( $R=2$ )
9. Coldness carriers in bio reactor with 50% volum is used.
10. First, we use batch feeding so that micro organisms correspond with new conditions and then we use continuous feeding.
11. In anaerobic and anoxic reactors we use mechanical stirrer.
12. In aerobic reactors, aeration is via air stone which can enter compressed air into the system.
13. Flow moves through reactor via gravity force
14. MLSS reflux from aerobic to anoxic reactor is performed by using pump.
15. MLSS reflux provides needed nitrate for anoxic zone.

### RESULTS AND DISCUSSION

We selected system was  $A^2O$  since it's cost effective and it also provides standard condition for nitrogen and phosphor release into the environment. We have applied many changes to this system as it is presented in figure 16. In this system sedimentation tank is used to deposit the wastewater and stabilizer controls dissolved oxygen and reflux flow rate. This system is designed for nitrogen and phosphor elimination in refinery's wastewater for the first time we can extend this research in the following areas:

1. Study on all systems used for nitrogen and phosphor elimination from refinery's feed.
2. Optimum number of stages for anoxic/ aerobic processes
3. Study the effect of of increasing the number of stages in anoxic and aerobic and Change in efficiency system performance.

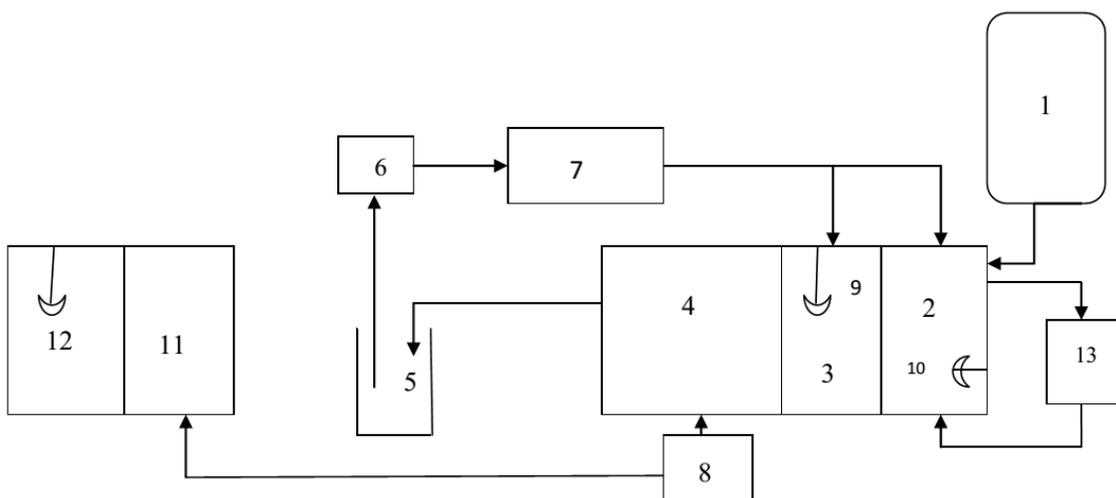


Figure 15: designed system's scheme

1. The feed tank
2. Anaerobic Reactor
3. Reactor anoxic
4. Aerobic reactor
5. Sedimentation tank
6. Pump (backflow)
7. stabilizer
8. air pump
9. mechanical stirrer
10. mechanical stirrer
11. aerobic reactor for safety
12. Anoxic reactor for safety
13. collecting tank of gas produced from the anaerobic reactor

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