COMPARISON STUDY OF MACROPOLLUTANT REMOVAL IN RIVER WATER USING CONVENTIONAL TREATMENT AND NANOFIBER MEMBRANE-BASED SYSTEM

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Abstract: High levels of nitrate (NO_3) and phosphate (PO_4) concentrations in water sources due to agricultural drainage or municipal disposal can generate eutrophication. It is characterized by blooms of either green or blue-green algae leading to significant drops in dissolved oxygen and frequently renders many fish and zooplankton species unable to survive in the water. In this study, the efforts to reduce total suspended solids (TSS), NO_3, and PO_4 pollutants in river water are evaluated using conventional treatment and membrane filtration systems as a comparison. Conventional water treatment process uses bar screening, flocculation-coagulation, and settling/sedimentation to remove pollutants; while membrane filtration system rejects a wide range of pollutants using pore exclusion. This study assessed electrospinning membrane filtration made of polyacrylonitrile (PAN) and polyacrylonitrile/polyethylene glycol-silver nanoparticle (PAN/PEG-Ag). The effectiveness of removing pollutant levels after going through a conventional type of water treatment and 12% PAN membrane (EM1), and 12% PAN-PEGAg 1% (EM2) showed the reduction level of TSS was 82.76%, 75.86%, 65.52 %, NO_3 reduction was 73.97%, 85.62%, 83.19%, and PO_4 reduction was 77.20%, 59.60%, 53.45%. The removal efficiency using pristine PAN membrane was 81.86%, 73.02% when using the conventional process, and 64.31% when using 12% PAN-1% PEGAg. After the conventional and membrane processes, TSS and nitrate level reductions were adequate, as seen from the set quality standard values, i.e., below 50 mg/L and 10 mg/L. Meanwhile, after both water treatments, the phosphate level did not meet the water quality standard, 0.2 mg/L, as regulated in the Government Regulation Number 22 of 2021.

Keywords: Contaminant levels; Conventional process; membrane separation; water treatment

Abstrak: Konsentrasi nitrat (NO_3) dan fosfat (PO_4) yang tinggi dalam sumber air karena drainase pertanian atau pembuangan kota dapat menghasilkan eutrofikasi. Hal ini ditandai dengan mekarnya ganggang hijau atau biru-hijau yang menyebabkan penurunan oksigen terlarut yang signifikan dan sering membuat banyak ikan dan spesies zooplankton tidak dapat bertahan hidup di air. Dalam studi ini, upaya untuk mengurangi polutan total padatan tersuspensi (TSS), NO_3, dan PO_4 dalam air sungai dievaluasi menggunakan pengolahan konvensional dan sistem filtrasi membran sebagai pembanding. Proses pengolahan air konvensional menggunakan bar screening, flokulasi-koagulasi, dan pengendapan/sedimentasi untuk menghilangkan polutan; sementara sistem filtrasi membran merejeksi berbagai macam polutan menggunakan eksklusi pori. Studi ini menilai filtrasi membran elektrospinning yang terbuat dari *polyacrilonitrile* (PAN) dan *polyacrilonitrile/polyethylene glycol-silver nanoparticle* (PAN/PEG-Ag). Efektifitas penyisihan kadar pencemar setelah melalui jenis pengolahan air secara konvensional dan membran PAN 12% (EM1), dan PAN 12%-PEGAg 1% (EM2) menunjukkan tingkat reduksi dari TSS adalah 82,76%, 75,86%, 65,52%, reduksi NO_3 adalah 73,97%, 85,62%, 83,19%, dan reduksi PO_4 adalah 77,20%, 59,60%, 53,45%. Efisiensi penyisihan menggunakan membran PAN murni adalah 81,86%, 73,02% bila menggunakan proses konvensional, dan 64,31% bila menggunakan 12% PAN-1% PEGAg. Setelah proses konvensional dan membran, penurunan kadar TSS dan nitrat cukup baik, terlihat dari nilai baku mutu yang ditetapkan yaitu di bawah 50 mg/L dan 10 mg/L. Sedangkan setelah dilakukan kedua pengolahan air tersebut, kadar fosfat tidak memenuhi baku mutu air yaitu 0,2 mg/L sebagaimana diatur dalam Peraturan Pemerintah Nomor 22 Tahun 2021.

Kata kunci: Kadar kontaminan; proses konvensional; pemisahan membran, pengolahan air

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Introduction

The quality of river water is influenced by farming, agricultural, and domestic activities (Mateo-Sagasta et al., 2017; Malaj., 2014). High levels of suspended solids, nutrients and intensive disposals of organic waste from the agricultural sector, such as nitrate and phosphate, can contaminate river water and affect its quality (Bashir et al., 2020; Braun et al., 2019). High concentrations of NO_3, NO_2, and PO_4^{3-} in effluents at sewage treatment plants, rivers, wetlands, and lakes are detected (Mishra and Patel, 2009; Costa et al., 2020). The excessive amount of phosphate (PO_4) and nitrate (NO_3) in water can cause eutrophication because aquatic ecosystems cannot fully degrade the pollutants contained in the water bodies (Cai et al., 2013; Ngatia et al., 2019). The eutrophication will decrease dissolved oxygen, which is toxic to native aquatic biota, disturbs the ecological balance and seriously impacts coastal ecosystems (Huang et al., 2010; Devlin and Brodie, 2023). Besides, an increased level of NO_3 in drinking water caused severe health problems, i.e., triggering cancer (Edokpayi et al., 2017; Ward et al., 2005). Nitrates can form N-nitrosamine, a carcinogen that affects cardiovascular, cognitive, and musculoskeletal health (Bondonno et al., 2023). Phosphorus may appear in many forms; among the forms found are orthophosphates, polyphosphates, and organic phosphates. Together, these are referred to as (P) total phosphorus (Davis, 2010). Excessive phosphate accumulation in the human body (hyperphosphatemia) can cause various disorders, such as cardiovascular, aging, chronic kidney disease, hypoparathyroidism, and metabolic acidosis (Komaba and Fukagawa, 2016; Lewis, 2022). Excess phosphate also harms the cardiovascular system and the aging process (Komaba and Fukagawa, 2016).

Changes in water quality due to eutrophication have been reported globally (Chau et al., 2015; Smith et al., 2016). For example, the Yamuna River in India, which supplies drinking water to nearly 20 million people, has been contaminated with high concentrations of NH_3 (Hussain., 2019). Eutrophication has led to negative consequences environmentally and economically, as happened in the Bohai Sea in China, Said Harbor in Egypt, and Indonesia (Wei et al., 2023; Morsy et al., 2022; Yudhistira et al., 2022). Water quality must be maintained to meet the standard in Government Regulation Number 22 of 2021, i.e., the permitted limit for TSS is 50 mg/L, for nitrate is 10 mg/L, and for phosphate is 0.2 mg/L.

Conventional water treatment involves coagulation, flocculation, sedimentation, and filtration processes (Spellman, 2017). Raw water with poor quality requires more advanced and complex treatment, such as using membrane technology, compared to raw water having a quality approaching drinking water.

Low water quality can threaten the availability of secure water and increase the burden of conventional water treatment plants (Balcerzak, 2006). Therefore, assessing the reduction of TSS, NO_3, and PO_4 contaminants from conventional processes is critical. This study evaluated the ability of conventional water treatment to reduce the concentration of TSS, NO_3, and PO_4. Likewise, an electrospinning-type membrane filtration was also used to process similar raw water. The membrane filtration has been widely used to solve water problems in developed countries (Choo and Oh, 2020; Tan et al., 2020). The application of membrane technology in developing countries is currently worth considering for conventional water treatment applications (Othman et al., 2022).

Methodology

Water Sample Collection

The water sampling procedure in clean water treatment plants referred to the SNI 7828:2012 procedure. A total of one liter of water sample was collected by grab sampling at the inlet (raw water) and outlet (treated water) of the water treatment unit, as shown in Figure 1. The samples were stored in glass bottles pre-sterilized with 70% ethanol and then analyzed in the laboratory. Sampling was conducted on sunny days.

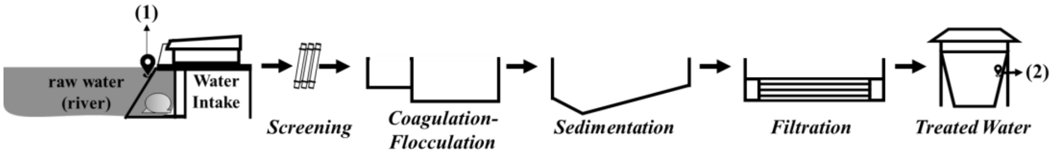

Figure 1. Conventional Water Treatment Position of Sample Collection

Analysis of the Chemical Quality of Water

The total suspended solids (TSS) in water samples were analyzed using a gravimetric technique, referring to the National Standardisation Agency's (2004) SNI 06-6989.3-2004. The quality parameters of nitrate (NO_3) were analyzed using the cadmium reduction method, HACH 8039; the detection limit was 0.3 - 30 mg/L $NO_3{}^{-}N$ (HR). The phosphate (PO_4) parameter was measured using PhosVer 3 with UV oxidation, HACH 8007; the detection limit was 0.02 - 125 mg/L PO_4^{3-}.

Water Treatment using Conventional Processes and Membrane Separation

As shown in Figure 1, the conventional water treatment process consists of screening, coagulation, flocculation-sedimentation, and filtration. The raw water and treated water samples in a conventional water treatment system of ± 1 L volume were conducted using a rinsed glass bottle with the grab sampling technique. The water samples were put into a container (a cooling box) filled with ice cubes at a temperature of ± 4°C, and then the water samples that have been collected are taken to the laboratory for analysis.

The water sample from the inlet of the conventional water treatment system was directly passed through a cross-flow filtration type flat sheet membrane module, as illustrated in detail in Figure 2. The filtration was carried out at room temperature with an air pressure of 0.2 bar. Two types of membranes as described in Table 1 were used as a filter media for water contaminant removal. TSS, NO_3, and PO_4 levels were analyzed from the permeate stream after a 30-minute filtration.

Water Quality Determination

The reductions of TSS, NO_3, dan PO_4 levels conventionally and by membrane filtration were calculated using Equation 1: (Lun et al. 2022).

$$\text{Reduction (\%)} = \frac{C_i - C_f}{C_i} \times 100 \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (1)$$

Remarks:

Ci : Initial concentration of TSS, NO_3, and PO_4

Cf : Concentration of TSS, NO_3, and PO_4 after treatment.

The data from the conventional and membrane filtration processes were compared to the quality standard in the Government Regulation Number 22 of 2021.

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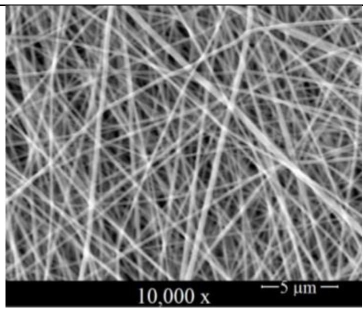
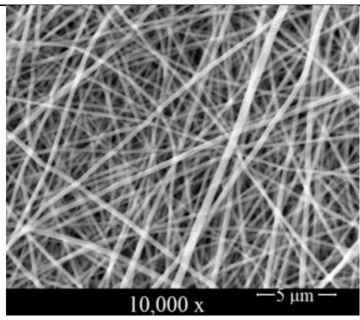
Table 1. Membrane Specification

Code	Composition	Mean pore size (µm)	Smallest pore size (µm)	Morphology
EM1	PAN	1,034	0,749	
EM2	PAN-PEGAg	1,536	0,992	

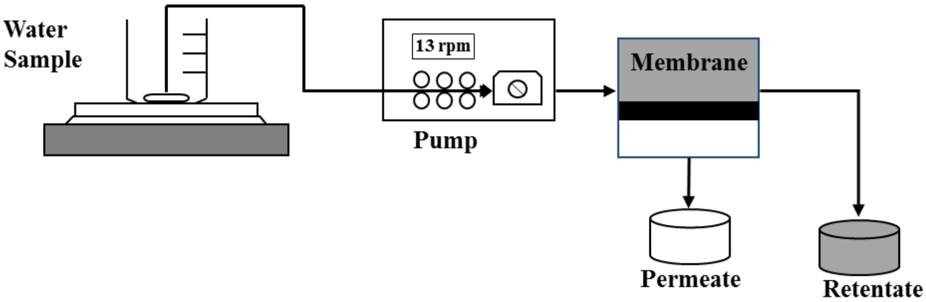


Figure 2. Membrane Filtration Process

Result and Discussion

Membrane Permeability Performance

The membrane performance in filtration can be observed from the water flux and rejection results. Membrane flux is obtained from the volume of permeate that can pass through the membrane per unit surface area of the membrane and unit of time (Fathanah et al., 2019). Figure 3 shows an overview of the water flux on both types of membranes.

The flux performance of the EM2 membrane was slightly higher than that of the EM1 membrane, shown both when filtration used deionized water and river water. This observation is in line with the specifications of the two membranes. As presented in Table 1, the EM2 membrane had a larger average pore size than the EM1 membrane. Therefore, the amount of water passing through the EM2 membrane layer was more significant than the EM1 membrane.

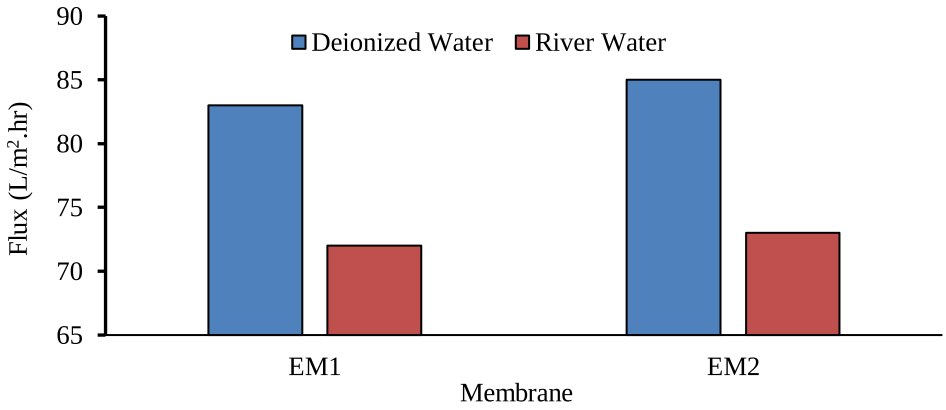


Figure 3. Water Flux Profile of Membrane

The mean pore size of nanofibers EM1 was 0,749 µm, and EM2 was 0,992 µm, which is categorized as a microfiltration membrane (Bilad et al., 2011). In this study, the bigger pore size of the EM2 membrane was due to the involvement of the pore-forming agent, polyethylene glycol (PEG).

Total Suspended Solid (TSS)

Table 2 shows the results of TSS analysis from conventional and membrane techniques. The raw water analysis shows that TSS levels in Table 2 met the drinking water quality standards per Government Regulation Number 22 of 2021, i.e., ≤ 50 mg/L. It is similar to a previous study demonstrating that the TSS in North Aceh waters (Krueng Aceh River) was 4-14 mg/L and did not pass the threshold of the decree of the state minister for the environment number 51 of 2004 concerning seawater quality standards (Khairunna et al., 2021). The result also proves that the river water sampled in the study had an adequate TSS parameter. Water treatment using conventional and membrane electrospinning (EM1 and EM2) techniques could reduce the TSS level significantly, with the conventional process showing higher TSS reduction than the membrane filtration technique (Figure 4).

Table 2. The Levels of Total Suspended Solids (TSS) in Raw Water and Processed Water

TSS Concentration (mg/L)				Remark
Raw Water	**After Processing**		**Threshold**	
29	Conventional	5	50	Meets Requirement
	EM1	7		Meets Requirement
	EM2	10		Meets Requirement

The study shows that conventional processing resulted in a TSS reduction of 82.76%. Meanwhile, TSS reductions in water samples after undergoing treatment with membrane filtration techniques using EM1 and EM2 were 75.86% and 65.52%, respectively. The results indicate that TSS reduction using the conventional process was better than membrane filtration. This condition was related to the pore size of both membranes used in this study. EM1 and EM2 had pores in the range of microfiltration membranes, so some suspended particles in water could pass through the membrane layer. Therefore, conventional processing resulted in better TSS reduction than the membrane technique. Conventional processes for the reduction of TSS involve a series of physical or chemical treatment steps to separate and remove these solid particles from the water (Al Bazedi & Abdel-Fatah, 2020).

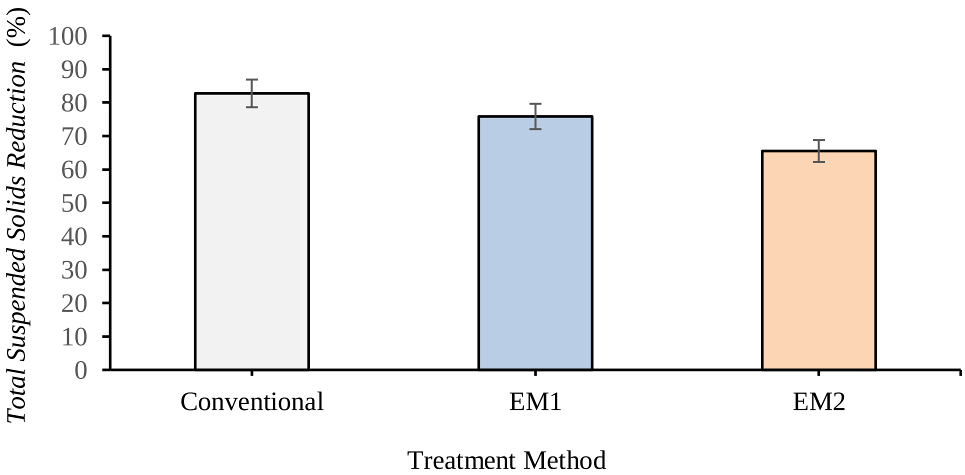


Figure 4. Total Suspended Solid (TSS) Reductions

Nitrate (NO_3)

Excess nitrate levels in water can lead to low dissolved oxygen levels (hypoxia), which is toxic to animals at concentrations ≥ 10 mg/L. Table 3 shows the nitrate (NO_3) concentrations in water before and after conventional and membrane filtration treatments. Nitrate levels in raw water exceeded the threshold set in the Government Regulation Number 22 of 2021, greater than 10 mg/L. Therefore, the outcomes of water treatments using conventional and membrane techniques follow the technical standards stipulated in Government Regulation Number 22 of 2021, namely ≤ 10 mg/L. Figure 5 presents the comparison of NO_3 reduction using conventional and membrane techniques.

Table 3. The Concentrations of Nitrate (NO_3) in Raw Water and Processed Water

Nitrate Concentration (mg/L)				Remark
Raw Water	**After Processing**		**Threshold**	
26,449	Conventional	6,884	10	Meets requirement
	EM1	3,804		Meets requirement
	EM2	4,445		Meets requirement

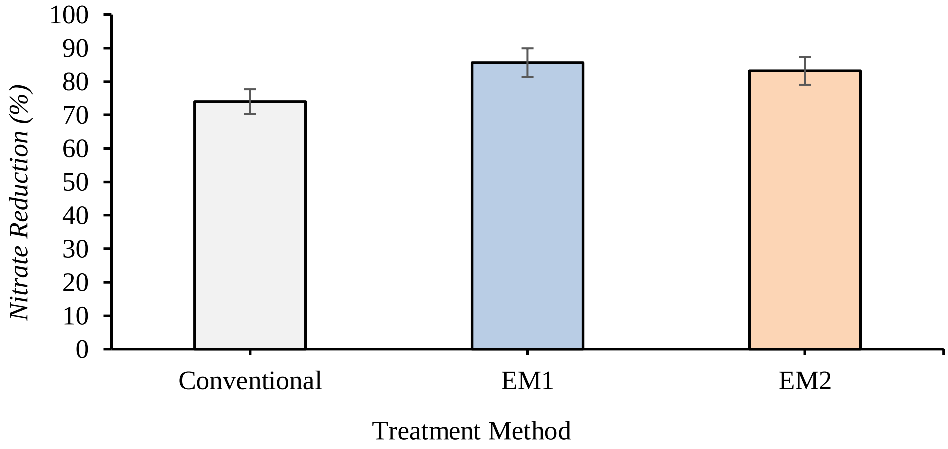


Figure 5. Nitrate (NO_3) Reductions

Coagulation, flocculation, and filtration in a conventional process can reduce nitrate by up to 73.97%. The rapid sand filter media in a conventional process consisting of silica sand, gravel, and sand have not been able to reduce nitrate optimally. Nitrate reduction with membrane technology was better than separation using a conventional technique. The reductions of nitrate (NO_3) levels using the EM1 and EM2 membranes were 85.62% and 83.19%, respectively. The EM1 membrane resulted in a more significant reduction than the EM2 membrane because the EM1 membrane had a smaller pore size (0.749 µm) than EM2 (0.992 µm). The Yekrang Research Team reported that electrospun nanofiber membrane technology could be used in water treatment plants (WTP). PVC/TPU/PC nanofiber membranes with 50 wt.% PC content had excellent filtration performances and could be used for water treatment (Yekrang et al., 2023). Electrospinning membrane technology can effectively reduce harmful contaminants, even in large quantities, and address the increasing problem of water pollution (Li et al., 2023). Moreover, electrospinning membranes were used due to several advantages, such as higher porosity and associated pore structure, which provided higher permeability and separation performance (Zeytuncu et al., 2023).

Phosphate (PO_4)

Table 4 presents the results of phosphate analysis in raw water and processed water samples. The PO_4 level in raw water was far above the allowed threshold according to the Government Regulation Number 22 of 2021. This condition shows that it is not yet at the safe limit to be in the environment. Table 4 shows conventional and membrane filtration water treatments can reduce PO_4 concentrations. However, the effluent concentrations of the two processes have yet to reach a value below the 0.2 mg/L quality threshold. Figure 6 shows the concentration reduction in samples after processing using conventional and membrane filtration techniques.

Table 4. Phosphate (PO_4) Concentrations in Raw Water and Processed Water

Phosphate Concentration (mg/L)				Remark
Raw Water	**After Processing**		**Threshold**	
3,416	Conventional	0,779	0,2	Does Not Meet Requirement
	EM1	1,38		Does Not Meet Requirement
	EM2	1,59		Does Not Meet Requirement

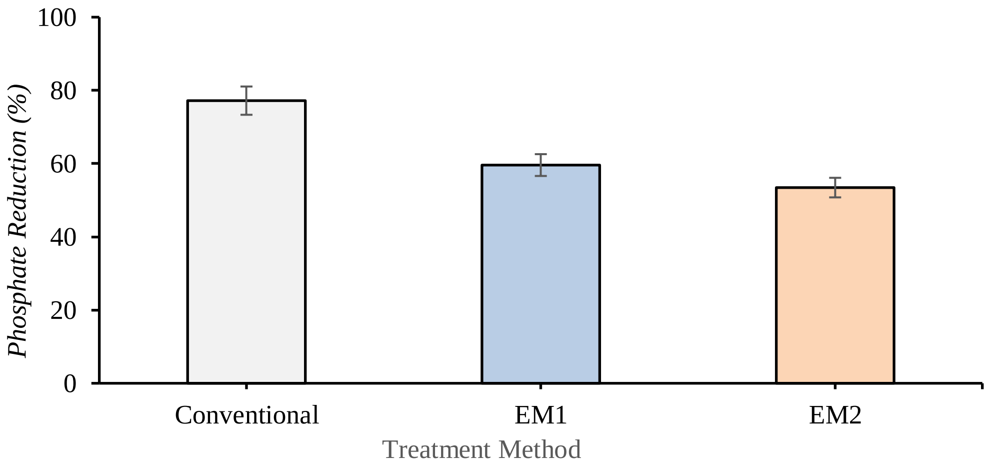


Figure 6. Phosphate (PO_4) Reductions

The highest phosphate (PO_4) reduction, reaching 77.2%, was obtained after conventional processing. The treatment with the membrane filtration technique only reduced phosphate to 59.60% for the EM1 membrane and 53.45% for the EM2 membrane. This result indicates that the type of electrospinning membrane in this study was inadequate to remove phosphate levels in raw water. Further research can be developed by choosing the membrane type within the nanofiltration category to achieve maximum removal efficiency. The elimination of various contaminants in water can be carried out by a physical process through filtration membranes (Cevallos-Mendoza et al., 2022). The mechanisms of reduction of NO_3, PO_4, and TSS by membranes illustrated in Figure 7 by Shen et al., (2020).

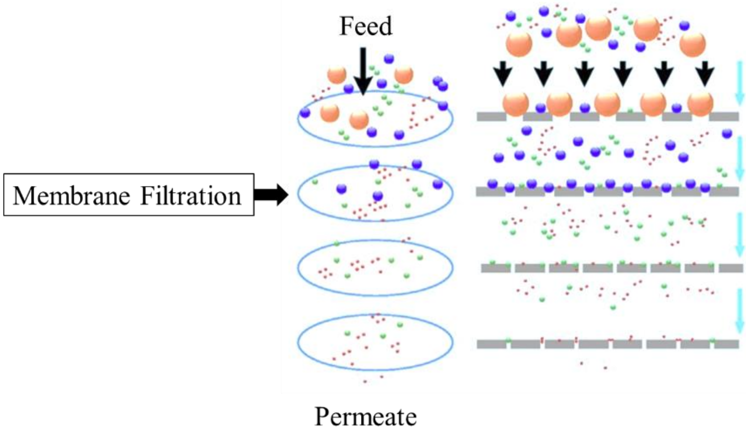


Figure 7. Membrane Filtration

The primary mechanism is through ion rejection based on the size of the membrane pores (Figure 7). Membranes provide significant advantages as a key technology in air purification, gas separation, and water treatment, encompassing the removal of suspended or soluble solids (Barhoum et al., 2023). As water is forced through the membrane, NO_3 ions are unable to pass through the small pores and are left behind in the reject stream, resulting in reduced nitrate concentration in the treated water (Majidi et al., 2022).

The membranes can remove phosphate by size exclusion, where the larger phosphate ions are rejected by the smaller membrane pores. Additionally, some membranes may have surface properties or coatings that allow for the adsorption of phosphate onto the membrane surface, effectively reducing phosphate concentrations in the treated water (Chee et al., 2022). As illustrated in Figure 6, it shows that PO_4 reduction in conventional processes is more effective than membrane types, this is due to the stages of adding coagulant material in the coagulation process where the coagulant will dissolve with phosphate (Davis, 2010). The research results of Owodunni et al., (2023) showed that the removal of phosphate ranged from 65 to 99.6% in coagulation-flocculation.

It's important to note that while membranes are effective in reducing NO_3, PO_4, and TSS, the overall performance can depend on various factors such as membrane material, pore size, operating conditions, and the nature of the contaminants in the water especially in preventing eutrophication treatment (Zeng et al., 2023). The combination of mechanisms and efficiency will vary depending on the specific membrane type used and the water quality being treated.

Conclusion

The highest contaminant reduction using conventional water treatment was in the TSS parameter, 82.76%, while the lowest reduction was nitrate, 73.97%. The 12% PAN type membrane (EM1) had the most significant removal percentage of 85.62% for nitrate, and the lowest was 59.60% for phosphate. Meanwhile, for the 12%PAN- 1%PEGAg (EM2), the highest reduction was for nitrate at 83.19%, and the lowest was for phosphate at 53.45%. Based on the Government Regulation Number 22 of 2021, contaminant levels of TSS and nitrate have met the technical standards set, namely less than 50 mg/L for TSS and 10 mg/L for nitrate from each water treatment process. However, the phosphate level was above 0.2 mg/L and, therefore, did not fulfill the technical criteria.

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