Prospect and Challenges of Tight Ultrafiltration Membrane in Drinking Water Treatment

P T P Aryanti¹, A N Hakim², S Widodo³-⁴, I N Widiasa⁴, I G Wenten²,*

¹Chemical Eng. Dept., Jenderal Achmad Yani University, Jl. Terusan Jend. Sudirman PO BOX 148, Cimahi, Indonesia.
²Department of Chemical Engineering, Institut Teknologi Bandung, Jl. Ganesha 10, Bandung 40132, Indonesia.
³PPPTMGB LEMIGAS, Balitbang KESDM, Jl. Ciledug Raya, Kav. 109, Cipulir, Kebayoran Lama, Jakarta, 12230, Indonesia.
⁴Chemical Engineering Department, Diponegoro University, Jl. Prof Sudarto-Tembalang, Semarang 50239, Indonesia.

*Corresponding author: igw@che.itb.ac.id

Abstract. Ultrafiltration (UF) membrane for drinking water treatment has been consistently modified to improve the selectivity towards trace organic compounds (TrOCs) and maintain the flux stability for long-term application. Currently, the tight UF membrane has been developed to answer the challenges of conventional UF membranes. With small pore size (300 - 5000 Da), the tight UF showed higher TrOCs rejection and lower irreversible fouling than the conventional UF membranes. Two methods have been proposed to fabricate the low molecular weight cut off (low-MWCO) UF membranes, i.e. blending membrane solution with fillers (nanoparticles or volatile solvent) and modifying membrane surface layer by chemical or plasma grafting method. Low water flux and high operating pressure are some challenges to be solved during the application of the tight UF membrane. Hence, the development of tight UF membrane with low-MWCO pore size has focused on improving flux while maintaining high selectivity. In this paper, the development and prospect of tight UF membrane will be reviewed. Challenges during the application of low-MWCO UF membrane for water treatment and methods to solve the challenges will be discussed.

Keywords: drinking water, high selectivity, low-MWCO, trace organic, ultrafiltration.

1. Introduction

In drinking water treatment, pressure driven membrane technologies achieved extraordinary levels of process intensification, substantially decreasing the equipment-size/production-capacity ratio, energy consumption, and waste production, which result in lower cost and sustainable technical solutions [1-3]. In spite the advantages, the application of pressure driven membrane in water treatment is limited by fouling phenomenon, which reduces the production efficiency [4-8]. Details of the theoretical background of the pressure-driven membrane processes including strategies to prevent and mitigate membrane fouling have been reported in several literature [9-16]. Reverse osmosis (RO) is the most used membranes in surface water treatment and desalination to produce fresh (pure) water with low content of soluble organics and ions [17-19]. However, the demineralized water by RO has not been recommended as drinking water for long-term consumption related to the World Health Organization.
(WHO) regulation [20]. The possible health consequences of low mineral content water consumption have been discussed in the literature [21, 22]. Hence, remineralization of NF and RO water product to reduce the water corrosivity is required before the water is distributed to the customer [23]. The standard for drinking water quality in terms of total dissolved solids (TDS) and mineral content has been summarized in the WHO guideline [23-25]. UF is an attractive alternative in water and drinking water treatment, which exhibits some advantages compared to NF and RO, i.e. lower investment cost, higher permeate flux, and lower energy consumption. From an economic point of view, the installation of UF membranes in water treatment plant may significantly reduce the operating cost by eliminating coagulation, flocculation, and sedimentation processes.

The UF membranes are generally characterized by their pore size between 0.01 µm (1 nm) – 0.1 µm (100 nm) or its molecular weight cut-off (MWCO) between 10,000 - 100,000 Dalton (Da) [25]. Due to its small pore size, the UF membrane provides an effective barrier for suspended particles, colloids, and pathogens while keeping the mineral content in water intake. Up to this time, the modification of UF membranes is mostly focused on improving the hydrophilicity, to reduce energy consumption and enhance productivity [26-30] and enhance an anti-fouling property of the membrane to maintain the membrane flux for long-term use [31]. Numerous review and research studies related to the UF membranes for water and drinking treatment have been published, as shown in Figure 1 [32-38]. Simultaneously, development of UF membranes with innovative modules is also growing significantly to fulfill the requirement of fresh or drinkable water [39-42]. The development is not only for industrial and urban areas, but also for a rural and remote area that have inadequate sanitation, poor hygiene habits, and lack of adequate quantities of water for hygiene [43-46].

![Figure 1](image_url)

**Figure 1.** Number of publication related to UF membrane application in water treatment indexed by Scopus (TITLE+ABS+KEY: ultrafiltration membrane and water treatment).

Beside intrinsic fouling phenomenon, another limitation of UF membrane in water treatment, particularly for drinking water, is low rejection against soluble contaminants (up to 50%), such as trace organic compounds (TrOCs) with an approximate molecular weight below 1000 Da. The TrOCs, such as endocrine disrupting compounds (EDCs) and pharmaceuticals or personal care products (PPCPs), have been identified as harmful pollutants in drinking water treatment since they are suspected to be carcinogenic [47]. Tight UF membranes have been developed to overcome the limitation in conventional UF membranes. This membrane has a tight structure in the skin layer, with pore sizes between 300 and 5000 Da [48-52]. Due to its tight skin layer, the UF membrane has a high rejection against soluble compounds in water, such as humic substances and dyes [53, 54]. Up to this time, there exists little literature that discusses the tight UF membrane and its application for water treatment. In this paper, the low-MWCO UF membrane will be reviewed comprehensively including
the state of the art and preparation method. In addition, challenges during the application of low-MWCO UF membrane for water treatment and methods to solve the challenges will be discussed.

2. The Application of Tight UF Membrane in Water Treatment

Several studies have been conducted to investigate the performance of tight UF membranes when it was applied in water treatment. One of the studies was conducted by Kramer et al. [54]. A tight UF membrane, with a pore size of 3000 Da, was used as pre-treatment of RO membrane to produce demineralized water from raw municipal sewage. It was found that the tight UF has an ability to reject chemical oxygen demand (COD) by 80%. Lin et al. [55] investigated the selectivity of a tight UF membrane, with a pore size of 4700 Da, to separate mixtures of dyes and Na₂SO₄. The tight UF membrane showed high rejection of dyes up to 98%. Similar research was carried out by Ma et al. [56], where the tight UF was used to separate dyes and mixed salts (both NaCl/Na₂SO₄). High rejection of dyes was also achieved (>98%). The pure water permeability of tight-UF membrane was found at least 6 times that of NF membrane. These results showed that the tight UF membrane can be used as an alternative to NF membranes.

Efligenir et al. [57] used two types of UF tight membrane (1000 Da), i.e. flat-sheet polyamide and TiO₂ tubular ceramic membrane, to remove metal salts from industrial discharge water, and then compared the performance of two membranes. They found that the TiO₂ ceramic membrane showed higher rejection of some metal ions, i.e. above 80% at operating pressure of 8 bar, while the polyamide membrane rejection was only 40%. It suggested that the negative surface charge of TiO₂ ceramic membrane played an important role to the metal ion rejection. Shang et al. [58] also used TiO₂ tight ceramic UF membrane (3000 Da) as pre-treatment of RO membrane to remove phosphate in water. They investigated the influence of membrane charge on the separation efficiency of phosphate by regulating the feed water condition, such as pH and salt concentration. The results pointed out that the ceramic membrane was more negatively charged in higher pH condition, which resulted in higher rejection of phosphate. Almost 87% rejection of phosphate can be achieved at pH 8.5. Meanwhile, the presence of Na⁺ and Ca²⁺ ions in solution decreased the zeta potential of the membrane and led to the increase of natural organic matter (NOM) or effluent organic matter (EFOM) on the membrane surface [59]. The phosphate rejection was found to be correlated to the zeta potential of the organics in the feed water. The application of tight UF in water treatment is summarized in Table 1. Due to its high selectivity at low energy consumption as well as low irreversible fouling, certainly, the tight UF membrane can be considered as a future drinking water treatment technology.
Table 1. The applications of tight UF membranes in water treatment

| Membrane material                     | UF pore size | Application                          | Operating condition                          | Results                                                | Ref. |
|---------------------------------------|--------------|--------------------------------------|----------------------------------------------|--------------------------------------------------------|------|
| Polysulfone (PSf)                     | -            | Humic substance removal in peat water| Operating pressure: 1 bar                      | Humic rejection: >80%; Flux: 126 LMH; Low irreversible fouling | [52] |
| Ceramic membrane                      | 3000 Da      | Sewage water treatment                | Operating temp.: 20°C Operating pressure: 8 bar| Organic matter rejection: 81%                         | [54] |
| Polyethersulfone (PES)                | 4700 Da      | N\textsubscript{2}SO\textsubscript{4}/dye separation| Operating temp.: 25°C cross-flow rate: 60 L/h process mode: filtration-diafiltration Operating pressure: 4 bar| Na\textsubscript{2}SO\textsubscript{4} rejection: >98% | [55] |
| Ceramic Membrane (TiO\textsubscript{2}/ZrO\textsubscript{2} active layer) | 8800 Da      | Separation of dyes and mixed salts (NaCl/Na\textsubscript{2}SO\textsubscript{4}) | Flow rate: 3 m/s at 25°C Operating pressure: 2 bar | Na\textsubscript{2}SO\textsubscript{4} rejection: >98% NaCl rejection < 10% | [56] |
| Ceramic membrane (TiO\textsubscript{2})  | 3000 Da      | Separation of phosphate               | pH of solution: 6 – 8.5                        | 87% of phosphate rejection at pH 8.5                  | [58] |
| Regenerated cellulose acetate (RC)    | 5000 Da      | 2,4,6-trichloroanisole (TCA) removal| Cross flow rate: 0,7 ml/min Operating pressure: 4 bar| TCA rejection: > 80%                                  | [60] |
| Charge ceramic membrane               | 3000 Da      | Phosphate removal during WWTP secondary effluent | pH 7.0 ± 0.5 cross-flow velocity: 1 m/s flux: 30 Lm-2h-1 | Phosphate rejection: 62%                              | [59] |
| Ag-doped TiO\textsubscript{2}/Ti composite membrane | 3000 Da      | Dye removal (chlorazol fast pink, chlorazol black, and titan yellow) | Operating pressure: 1-7 bar Operating temp.: 25°C | Dye rejection: 99%                                    | [61] |
| Modified poly(vinyl butyral) (PVB) hollow fiber membranes with carbon nanotubes (CNTs) | -            | Humic substance removal               | -                                            | Humic substance rejection: >90%                       | [62] |

3. Tight UF Membrane Preparation
A few studies have been focused on the tight UF membrane preparation techniques, including the dispersing fillers (polymeric or inorganic) in membrane structure [63,64] and membrane surface modifications by chemical grafting or pore-filling method [65-67]. Among these methods, blending membrane materials with hydrophilic and anti-fouling nanoparticles has attracted much attention in tight UF membrane manufacturing along with refusing fouling from the membrane surface [68-70]. Another method has been proposed by filling the membrane pore with a high cross-link polymer through in-situ polymerization [71,72] or with nanoparticles by vacuum filtration [73]. In the following subchapter, the preparation methods of tight UF membrane will be discussed.
3.1. **Tight UF polymeric membrane preparation by dispersing fillers in membrane structure**

There are two methods to disperse the fillers in membrane structure, namely ex-situ and in-situ method. In ex-situ method, the fillers are dispersed by blending into the membrane solution, while in in-situ method, the fillers are directly generated in membrane solution through a chemical reaction. Blending polymer solution with hydrophilic or anti-fouling additives are widely used in the manufacture and modification of membranes used in UF membrane modification due to its simple and inexpensive method. The frequent additives used in membrane modification are hydrophilic polymer (e.g. polyvinyl pyrrolidone or PVP and polyethylene glycol or PEG), organic compounds (e.g. glycerol, alcohol), inorganic salts (e.g. LiNO₃), inorganic compound (e.g. zeolite, TiO₂, Ag), water, and recently modified polymeric additive (e.g. sulfonated polyether ether ketone or SPEEK and PVP- co-styrene). In tight UF membrane preparation, some of these additives have been used and will be discussed in this sub-chapter.

Sadrzadeh et al. [74] manufactured UF membrane with a pore size of 2000 Da by blending 15%wt of PES and 5% wt of PVP (1300 Da). Further addition of PVP to 10%wt reduced the membrane pore size to 1000 Da and changed the membrane morphology from finger type to sponge type. Aryanti et al. [75] prepared a low-MWCO UF membrane, which had a high rejection to humic substances (up to 80%). They mixed PSf solution with acetone to obtain tight surface layer and PEG400 to increase membrane hydrophilicity. The rapid loss of acetone from the membrane solution lead to a higher polymer concentration in the membrane skin layer and produced tight pore structures [76]. As shown in Figure 2, the presence of acetone in membrane solution improved the humic substance rejection [77]. Furthermore, a tight skin layer of the membrane weakens the interaction between organic matters and the membrane surface, which is easily cleaned by flushing method and contribute to higher flux recovery ratio (FRR) [52].

![Figure 2](image.png)

**Figure 2.** Effect of acetone concentration to humic substances removal [77].

Recently, blending polymer solution with inorganic nanoparticles becomes a choice of numerous researchers to produce a smaller membrane pore size with a good pore distribution along the thickness of UF membrane. TiO₂ has been widely used as an additive due to its properties and good compatibility with organic solvent during the preparation of UF membrane. It has been reported that composite UF membrane with TiO₂ is able to provide high rejection of low MWCO organic matter and good anti-fouling performance. Hamid et al. [78] found that polysulfone (PSf)/TiO₂ composite membrane was able to perform up to 90% rejection of low molecular humic acid. However, the pore size of the resulted membrane was beyond the pore range of Tight UF. Leo et al. [79] found that almost 80% permeate flux can be maintained during filtration process of humic solution, due to the presence of zeolite in membrane structure. Another hydrophilic nanoparticles, such as ZrO₂ [80], SiO₂ [81], zeolite [82], ZnO [83, 84], and Ag [85, 86], are also considered as an alternative modifier to enhance the performances of UF membranes [79]. By increasing the nanoparticle additive concentration, the smaller finger-like pore is formed due to the exchange rate between solvent and
nonsolvent has been delayed thus resulted in a tight pore structure and higher rejection of foulants. Kotte et al. [72] prepared a tight PVDF UF membrane by in-situ generation of crosslinked PEI micro/nanoparticle (with diameters ranging from 400 nm to 3 μm) in membrane casting solution using epichlorohydrin as crosslinker.

3.2. Surface modification of porous membrane by chemical reaction

Pore-filling method by chemical reaction is another strategy to produce a tight structure on the membrane surface (Figure 3). This method has been used in commercial membrane fabrication to modify the characteristic of membrane or covers the defective pore in membrane surfaces structure [87, 88]. A porous membrane support is immersed in a polymer solution containing other type of polymer or particles. Further thermal treatment is performed to form cross-link structure and then dried at room temperature until all solvents are evaporated. The filled membrane is immersed in a solution to extract the unwanted component from the surface structure. Lan et al. [71] filled polyvinylidene fluoride (PVDF) microfiltration membranes with phenolic resins (PRs)/block copolymers (BCPs). They dipped the PVDF membrane in a homogenous solution containing resol and F127 polymer. The coating membrane was heated to cross-link the resol and generate thermosetting PRs, while F127 was dispersed in PRs structure. The pore-filled membrane was immersed in an acid solution (H2SO4) to remove the F127, thus a mesopores surface layer was formed with a pore size of 2350 Da.

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**Dipping in solution**

**Heat treatment**

**Leaching method**

Porous membrane

Homogenous solution

**Figure 3.** Schematic illustration of surface modification of porous membrane by pore-filling method.

UV-initiated graft copolymerization of poly(ethylene glycol) methacrylate (PEGMA) onto a commercial tight UF PES membranes has been performed for humic acid removal [89]. Smaller membrane pore size was produced when the energy of UV irradiation was between 5 to 9 J/cm². Further increase of UV irradiation energy resulted in a degradation of membrane pore leading to a higher membrane pore size. Another method was proposed by Chen et al. [90] to modify the surface of α-alimina UF by TiO2 nanoparticle doping sol-gel method. The resulted UF membrane had a pore size with pore size approaching the tight UF range. Several techniques for tight UF preparations is summarized in Table 2.

4. Challenges of Tight UF Membrane for Drinking Water Treatment

4.1. Trade-off between selectivity and permeability.

The most problem in tight UF membrane fabrication is trade-off between selectivity and permeability. These tight UF membranes had too low permeability and higher operating pressures than conventional UF membranes. Ariono et al. [65] found that higher and tighter of membrane pore size was obtained when the polymer concentration was increased, while the permeability was sharply reduced (Figure 4). The increase of polymer concentration increased the viscosity of the membrane solution, which inhibited the growth of membrane pore and reduced the pure water flux. In contrast, the tight membrane skin structure increased the membrane rejection. Further developments are continued to improve the membrane permeability while retaining tight pores [91].
Table 2. Several techniques in tight UF membrane preparation.

| Modification Techniques | Methods | Membrane pore size | References |
|-------------------------|---------|-------------------|------------|
| Blending                | Blending polyethersulfone with PVP (1300 Da) | 2000 Da | [74] |
| Blending                | Blending polysulfone with PEG400 and acetone | 0.01 μm or 10 nm | [75] |
| In-situ generation of fillers | In-situ generation of crosslinked PEI in PVDF membrane structure | - | [72] |
| Dip-coating followed by leaching method | Supported PVDF filled with resin/polymer solution and followed by acid treatment using H2SO4 solution | 2350 ± 200 | [71] |
| Nanoparticle doping sol-gel method | α-alumina microfiltration filled with TiO2 nanoparticle (average particle size: 25 nm) | 14.700 | [90] |
| UV-initiated graft copolymerization | Graft copolymerization of poly(ethylene glycol) methacrylate (PEGMA) onto a commercial tight UF PES membranes. | 4 – 10 | [89] |

Figure 4. Trade-off between selectivity and permeability during tight UF membrane preparation [64].

4.2. Fouling of organic matter during drinking water treatment
Fouling phenomena have hindered the application of tight UF polymeric membranes for the proposed one-step drinking water disinfection. Although negatively charged tight UF membranes have shown some fouling resistance to colloids and humic materials, these membranes are particularly susceptible to chemical degradation due to chlorine and another oxidant attack. Therefore, developments of commercially viable fouling resistant polymeric membranes have been conducted to achieve one-step disinfection without the need for chemical pretreatment. Several techniques to produce anti-fouling UF membranes have been proposed, from passive anti-fouling to active anti-fouling [91-92]. The passive anti-fouling strategies are developed by manipulating the physicochemical or topological structure of the membrane surface to reduce the interaction between foulant and the membrane surface. Introducing hydrophilic polymers (for examples: PEG-based, PVP-based, and zwitterionic) through hydrogen bonding or ionic solvation is considered as an effective method to prevent the adsorption of foulants on the membrane surface [93,94].
In addition to the hydrophilic polymers, some anti-microbial inorganic nanoparticles (NPs) are also used to avoid the adsorption of foulants to the membrane surface, such as metals (silver/Ag) and metal oxides (TiO₂, SiO₂, Fe₂O₃, and ZnO). Among these NPs, Ag NPs are commonly used as anti-fouling agent in the membrane structure due to its ability to disrupt the cell of bacteria and kill the bacteria when attached on the membrane surface. When using NPs in UF membrane modification for drinking water treatment, the most problems that need to be addressed is the release of NPs from the membrane structure to the environment. Up to this time, a few strategies have been proposed to improve the entrapment of NPs in membrane structure, such as PVP or PEG addition into the membrane solution during membrane preparation and modification of the membrane surface before depositing silver on the surface.

5. Conclusions
Some techniques for preparation of tight UF membrane have been developed to answer the challenges of conventional UF membranes. The tight UF membrane which has a tight structure in the skin layer showed a high rejection towards the TrOCs and low-reversible fouling compared to the conventional UF membranes. In some cases, the tight UF membrane can be used as an alternative to NF membranes.

Two methods have been proposed to fabricate the low-MWCO UF membranes, i.e. blending nanoparticles or volatile solvent into membrane solution and modifying membrane surface by chemical or plasma grafting method. Low water flux and high operating pressure are challenges of the tight UF membrane application in water treatment which need to be solved. Hence, the development of tight UF membrane has focused on improving flux while maintaining high selectivity.

Introducing hydrophilic polymers or anti-fouling agents onto the membrane surface is considered as an effective method to improve the membrane permeability as well as to avoid the adsorption of foulants to the membrane surface. Recently, nanoparticles (NPs) with anti-microbial properties are increasingly used in membrane modification to reduce fouling formation on the membrane surface. When using the nanoparticles in UF membrane modification for drinking water treatment, the main problem is the release of NPs from the membrane structure to the environment. Therefore, a few strategies should be considered to improve the entrapment of NPs in membrane structure.

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