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A Critical Study and analysis to the Development of Potential Ester-Water Separation Membrane Materials for Ester-Filled Power Transformers

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ABSTRACT
In the present scenario, there has been a continuous search for environmentally friendly alternatives to the consumables used by utilities. One of the consumables being mineral oil conventionally used as dielectric liquid in power and distribution transformers; the proposed biodegradable substitute for which is ester. But the substitute is not effectively compatible with the current operation of passive environmental mechanisms in electrical substations. Therefore, there is an active requirement for a solution to minimize any environmental release of liquids employed.

KEYWORDS
Ester-water separation, membrane materials, pervaporation, Petro-pipe, zeolite, mixed matrix membrane, polyimide

1. INTRODUCTION

A transformer is considered a focal point of any power generation and transmission system. To ensure its proper and efficient functioning, the insulating fluid plays a significant role. Although mineral oil has been used as the insulating fluid in transformers for ages due to its high dielectric strength, cheap cost, and availability, utilities are looking for a less polluting substituting fluid. This is predominantly because mineral oil is non-renewable and non-biodegradable. For the alternative biodegradable oil (Ester) to be successfully incorporated in the active power system, it is required to update the present oil-water separation technologies accordingly while compatibility with the present design of the transformer being a necessity.

The reason behind this active search for an upgrade is that the specific gravity of ester (0.85-0.89) is very close to that of water (1). Therefore, the existing liquid-water separation technologies may not be efficient enough for the new proposed liquid which is ester oil.

This study will help explore and discover potential membrane materials that can be incorporated in present passive oil-water separation devices to efficiently separate ester-water as well.

Rest of this paper is organized as follows: Section 2 contains problem statement followed by proposed methodology in section 3. The next section showcases the results and conclusions.

2. CONVENTIONAL OIL-WATER SEPARATION TECHNOLOGIES

Electrical Figure 1 depicts the basic layout of an oil-water separator (OWS) [2].

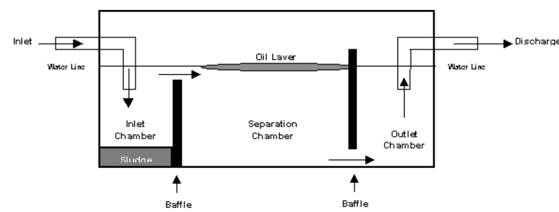

Fig 1: Basic Layout of Oil-Water Separator (OWS)

Gravitation separation method: In this method of separation, oil-water mixture is passed through a series of plates. It is designed such that the oil particles combine and accumulate at the top of the separator tank. The accumulated oil is then transferred to a separate oil containment tank. Separation by gravity is pretty slow, so in order to accelerate the procedure, chemical agents are employed which can be termed as demulsifiers.
This method is not much efficient as it cannot separate emulsified oil from the mixture. Figure 2 depicts oil-water separation through a gravitation separator.

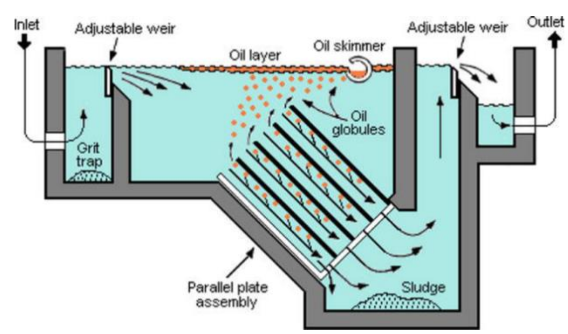

Fig 2: Gravitation separator

API Oil-Water Separator: It is a device that works on the principle of gravity separation. It is working based on Stoke's law, which states that the particles dimension velocity is dependent on the density difference between the solid phase and the liquid phase, viscosity of the liquid, and size of the particle. Figure 3 shows the top and side view of an operating API separator.

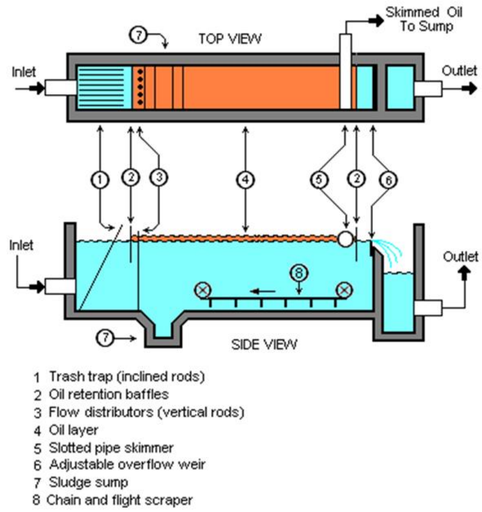

Fig 3: API Separator

Centrifugal separation method: In this method of separation, oil- water mixture is separated by centrifugation as shown in figure 4. The separator is designed with a rotatable cylindrical container inside. In operation, the liquid with higher specific gravity (water) flows and accumulates axially outwards and can be extracted from the boundary of the cylinder, while the liquid with lower specific gravity (oil) piles up at the rotating axis and can be extracted from there only.

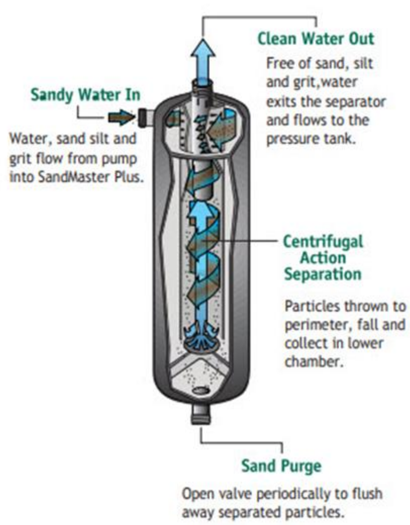

Fig 4: Centrifugal oil-water separator

Flotation: It is based on the fact that oil is lighter than water and piles up atop the water. This method introduced bubbles into the procedure to accelerate the rate of oil accumulation on the top. Various methods of flotation separation include dissolved air flotation (DAF), froth flotation, dissolved gas flotation (DGF), etc. [17]

Coagulation- Flocculation: Coagulation means an addition of chemicals also known as coagulants, which are broadly based on either aluminium or iron, into the contaminated mixture. These coagulants carry opposite charge from the non-settleable substance in the mixture, hence neutralizing them to support sedimentation [16].

Flocculation is the process of gently stirring the mixture to further assist with the filtration process.

Passive Oil Water Separator (OWS) devices are to be classified [4][11]as:

i. Petro pipe
ii. HFF oil stop valve

Table I shows the classification [21] of oil-water separation techniques based on materials used:

Table I: Oil-Water Separation

Filtration Materials	Absorbent Materials
a. Mesh	a. Porous Media
b. Membrane	b. Powder
c. Filter	c. Gel
d. Film	d. Nano-composite

The passive OWS devices mentioned in this report operate on membrane-based separation.

3. PETRO-PIPE

The purpose of a Petro-pipe is to allow rainwater to flow through and in case of an oil spill, filter the oil sheen out. As soon as the spill becomes larger, the media inside the Petro-pipe gets locked.

Petro pipes come with male or female fitting and can be used with or without ball valves. It is designed to work passively [5] throughout day and night.

Schematic: Petro-pipe is attached to the housing pipe with the help of 5 screws made of stainless steel and it performs best when installed on a 20°-25° mounting position. It is primarily employed in outdoor oil-contaminated areas and has to be changed every 5 years.

Petro pipes have a removable retaining ring which after being removed, grants access to the dirt filter inside. This filter needs to be cleaned at least once a year.

Figure 6 displays the typical dimensions of a PIFH-616 Petro-pipe.

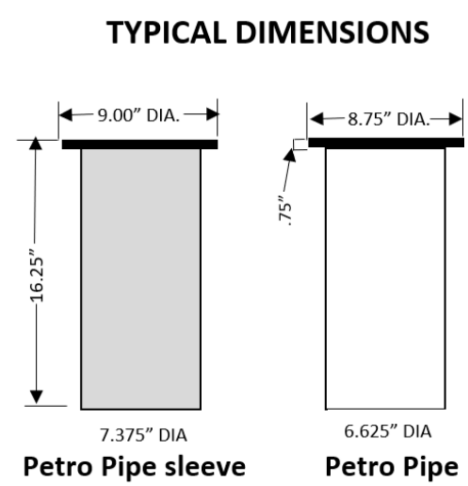

Fig 6: PIFH-616 Petro-Pipe

A pre-filter cage is used inside the oil containment tank to filter out dirt and prevent it from entering the Petro-pipe. Figure 7 shows the layout of a Petro-pipe [5] on a concrete floor.

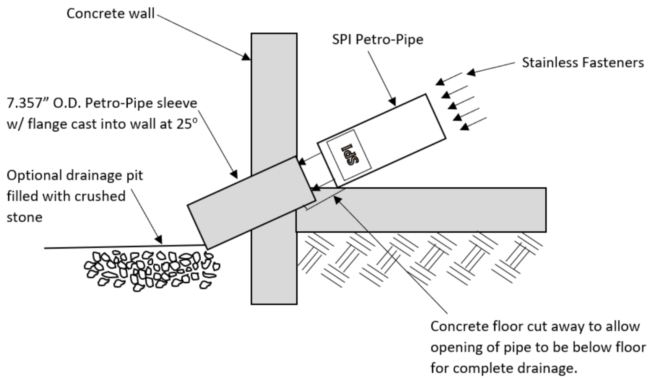

Fig 7: Layout of Petro Pipe

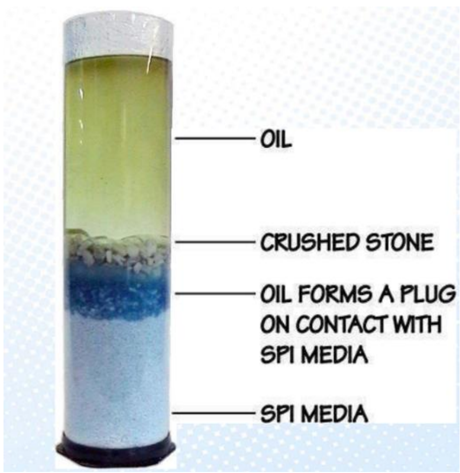

Fig 8: Oil stopping in Petro-pipe

Oil stopping in Petro-pipe as shown in Figure 8, in layers inside the pipe resides the filtration media which swells and clogs when coming in contact with oil. The filtration media used in Petro-pipe is Alum.

Working Mechanism of Petro-Pipe: A mixture of spilled oil and rainwater resides in the accidental oil containment tank wherein the oil aligns on top of the water as depicted in figure 9.

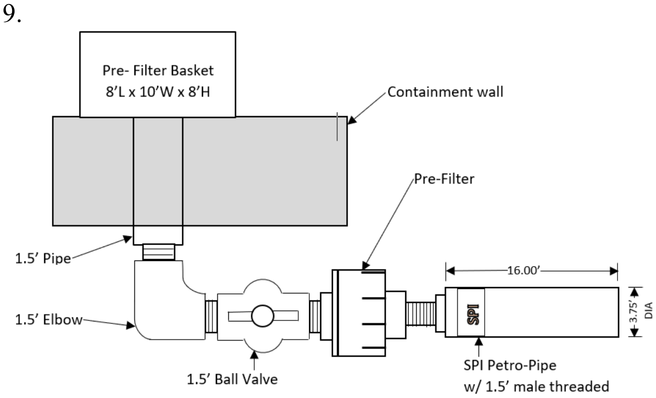

Fig 9: Petro-Pipe Plan

The Petro-pipe is connected to the pipe coming out of the oil containment tank. The water at the bottom of the tank starts draining out of the Petro-pipe.

As the level of water goes down in the containment area, the oil atop starts entering the Petro-pipe thereby slowing down the flow of water. The Petro-pipe will continue to drip the water out for several minutes after the oil gets in contact with the pipe media. This is due to the pressure exerted by the oil in the unused media.

Petro-Pipe Alternative: HFF Oil Stop Valve: This valve is another hydrocarbon filtering device that filters out water while stopping hydrocarbons from flowing out through it. Figure 10 is a display of Justrite oil stop valve.

Fig 10: Justrite oil stop valve

The solidifying media used inside the valve is composed of Justrite oil solidifying polymers, a proprietary blend of USDA food-grade polymers. It is capable of blocking any oil, ester, diesel, and many more organic hydrocarbons when needed. HFF oil stop valve is capable of filtering all organic hydrocarbons to less than 5ppm. The maintenance cost can be reduced further with the help of an additional accessory that is a pre-filter.

4. ESTER AND WATER SEPARATION TECHNOLOGIES

Out of many ester-water separation techniques, the following techniques are of main focusing this report.

Membrane Separation Method: Membrane separation technology has been conventionally used in wastewater treatment processes. A membrane is a selective barrier that allows certain things to pass while restricting others. Figure 11 shows the separation of a mixture through a permeable membrane.

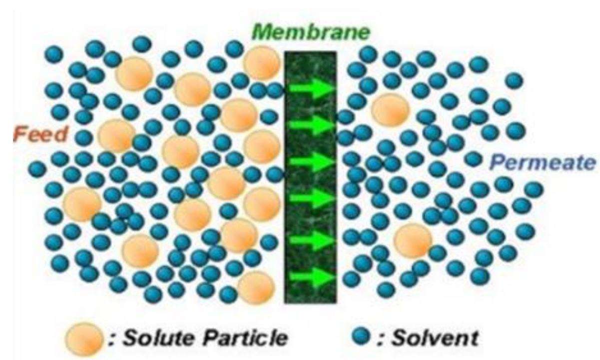

Fig 11: Membrane Separation Process

Based on the material, a membrane can be organic or inorganic. Figure 12 depicts the classification of membrane separation techniques into equilibrium and non-equilibrium.

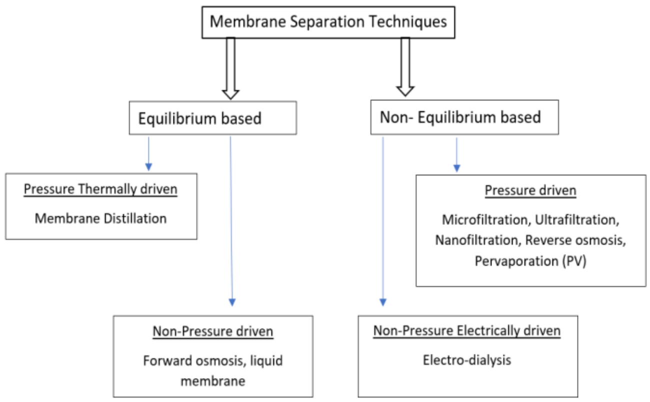

Fig 12: Membrane Separation Process

Of the aforementioned, pressure-driven membrane processes are the most widely employed techniques in wastewater treatment. As the name suggests, these processes utilize hydraulic pressure for the separation of a given mixture. The types of pressure-driven processes are mentioned in the table above, which are differentiated by pore size. These types can be used in different combinations for different wastewater treatment applications. Microfiltration, Ultra filtration, or Nano filtration [30] are usually treated as pre-treatment steps before the reverse osmosis (or hyperfiltration). [9] [28] [29] [31]

Pervaporation Technique: In a pervaporation separation process, membrane permeation and evaporation of a specific liquid in a mixture takes place simultaneously as shown in figure 13. In the pervaporation technique, an isothermal mixture of liquid is passed through a membrane in which, the more permeable liquid gets absorbed [22] [26]. This absorbed component of the mixture evaporates at the downstream phase of the membrane. This vapor can be then condensed and hence, separation is achieved. Although this technique is usually applied in alcohol-water separation, it is also being explored to be utilized in wastewater treatment [1].

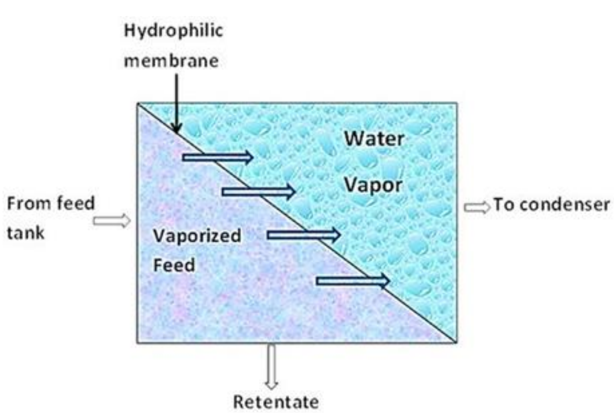

Fig 13: Pervaporation

5. POTENTIAL MEMBRANE MATERIALS

Pervaporation membrane Polydimethylsiloxane (PDMS): Because of its flexible structure, a PDMS membrane offers high selective permeability for organic compounds; hence can be used to separate organic components from a given liquid mixture. [27] Therefore, an Ester-permeable PDMS membrane can be explored in wastewater management techniques for ester-water separation.

CuBDC-GO composite incorporated PDMS Membrane: Several studies have proven that even small modifications in a PDMS membrane have the potential to enhance separation processes. For instance, an advanced copper terephthalate metal-organic framework (CuBDC) introduced between graphene oxide (GO) layers have proven to improve the separation process of ester and water by a significant margin [7].

Organic polymers: The various types of polymers used for Easter-Water membrane separation

i. Poly Vinyl Alcohol (PVA)
ii. Cellulose polymer
iii. Polyimides
iv. Other polymers that are hydrophilic in nature but are not commercially available (yet) despite decades of research include PVA/hydroxyethyl cellulose, PVA/CMC, PVA/sodium alginate, and PVA/poly(allylamine)-hydrochloride, PVA/poly (acrylic acid).
v. Hydrophobic polymer: Amorphous Perfluoro polymers

The aforementioned products show a significant tendency to absorb water from the mixture, causing the membrane to swell. It may lead to unwanted changes in the permeability and selectivity of the membrane [6]. Amorphous Perfluoro polymers (APFP) are a considerably newer group of polymers that exhibits minimal swelling in water.

The other polymers that are used are of Carbon-based materials such as Carbon molecular sieves (CMSs), Graphene and Polymers of intrinsic micro-porosity (PIMs). These materials are usually utilized as polymer fillers rather than being used on their own. Membranes of the above-mentioned organic materials are often compositely layered to improve the selectivity of the membrane.

Inorganic materials:

Zeolite-based membranes**:** These are usually used to remove water from bio-ethanol but have a good shot at separating esters as well. Although making zeolite-based membranes may come out as a challenge, their durability to chemical and thermal changes makes them a stable and worthy membrane material to be tested further. [12] [24]

Linde Type A (LTA) Zeolites: The most commonly used zeolite in PV membranes; zeolite is composed of aluminium and silicon with a 1:1 ratio. Any increase in the aluminium tends to strengthen the hydrophilic nature of the membrane as well as degrade its hydro-thermal and acid resistances. The striking quality of these membranes is that they do not swell during hydro sorption. Factors affecting the selectivity of zeolite membranes are Molecular sieving, sorption and diffusion. Zeolite membranes are highly durable, stable, and have a long lifetime; still, zeolite membranes are not economical as they are known to be approximately 10 times more expensive than a regular polymeric membrane [12].

T-type and Chabazite Zeolites: To widen the operating range of A-type zeolites, steadier frameworks have been developed. Of them are T-type and chabazite (CHA) which are available in the market. The T-type zeolite has silicon to aluminium ratio of 3:1 to 4:1, which makes it less hydrophilic and more stable to acidity as compared to LTA zeolites. The CHA zeolite membranes are more of a recent development and are manufactured by Mitsubishi Chemical Corporation (Japan) and Hitachi Zosen Corporation (Japan). These zeolites can be evolved into several different forms with different pore sizes by exchanging the ion deployed. They can also be constructed with a wide range of silicon to aluminium ratios going as high as 11:1.

ii. Hybrid silica membranes: Also known as organo-silica, these membranes can be recognized as an advancement to microporous silica membranes and are formed by introducing an organic linkage in microporous hydrophilic silica membrane structure or by using bis (triethoxysilyl) ethane (BTESE).The extent of water and solvent separation in hybrid silica membranes is lower as compared to LTA zeolites. These are tested for a wide range of stability yet are considered sensitive to pH, with the operating pH being 2-8.

iii. Mixed matrix membranes: Mixed matrix materials are the latest development to be implemented in membrane-making technology. These membranes were designed with the sole purpose to utilize the best of molecular sieving materials and polymeric materials. This is due to the fact that manufacturing permeation-selective membranes using molecular sieving materials like zeolites is quite challenging. MMMs consist of a polymer matrix and some filler material (either porous like zeolite or non-porous like graphene). The most prominent downside of this seemingly promising membrane is that it has limitations of both the filler material and polymer used. Polyether sulfone (PES) membrane is a popular hydrophilic membrane employed in separation processes with high flow rates. MMMs of dual nanofillers have experimentally proven to improve the permeability of PES membranes. Although the studies on MMM are consistent till date, there is still no commercial MMM available for wastewater treatment.

6. COMPARATIVE ANALYSIS

Vane LM [1] has proposed that the problem of separating azeotropic mixtures with water is addressed along with a comprehensive study of pressure- driven membrane separation like pervaporation (PV) in this field. This literature provides a state-of-the-science analysis of potential materials that can work in correspondence with PV/VP membranes. The motivation behind the study was intensive energy consumed in distillation-based separation processes. Non-polar solvents like n-Hexane, cyclohexane and 1,2,4-Trimethylbenzene as well as poplar solvents like Methyl tert-butyl ether (MtBE) and 1-Butanol are some of the solvents that form a zeotropic mixture with water and are the focal point of this study. Membranes based on materials like NaA zeolite, CHA, hybrid silica etc. have the potential to separate water from practically any solvent. Although promising, they still have their limitations. According to the paper, the most prominent limitation for hydrophilic polymers is the maximum water content of the feed mixture. And to tackle that Amorphous Perfluoro-polymers (APFP), a modified hydrophobic polymer is introduced.

Table II: Manufacturers Details

Material	Manufacturer
Organic Materials	
PVA	
→ Reverse Osmosis Membrane	Bluestar (Hangzhou) Membrane Industries Co, Ltd
→ PVA 2688	
→ Kuraray Povaland Elvanol	Easthony Kuraray
Cellulose	
→ Cellulose Filter Membrane	Merk
→ Arbocel: Highly Purified Cellulose	JRS
→ Whatman 7182-001 Cellulose Nitrate Membrane Filter	Whatman
Polyamide Membrane Filters / Type 25007	Sartorius
CMS220 Carbon Molecular Sieve	Acuro Organics Limited
Graphene membrane	Nematiq
Teflon™ AF Resins	Teflon
Inorganic Materials	
LTA/ NaA Zeolite	
→ Adsorption Column Price Zeolite Catalyst 2- 4 mm	Gongyi City Meiqi Industry & Trade CO., Ltd
→ Zeolite NaA	Heartychem
→ HFS (R) Molecular Sieve Zeolite 8X12 Mesh	HFS
→ Nanozeolites	Zeovation
CHA	
→ Chabazite Zeolites	Clariant
→ ZEBREX	Mitsubishi Chemical
Hybrid Silica	
→ GE&R silica nanoparticles	General Engineering and Research
→ Precipitated Silica/ Hydrophilic Fumed Silica/ Hydrophobic Silica	Shandong Haochuang Material Co., Ltd.
NFX Nanofiltration membrane	Synder Filtration

Table II shows the manufacturers (column 2) of the materials (column 1). This study focuses on the analysis of the materials mentioned in this paper to find which of these can be suitable to be implemented in passive protective mechanisms of power and distribution transformers. According to the paper, hybrid silica membrane can breakdown under alkaline conditions (pH limit 8.5), therefore its potential reduces. Although in my research, I've found that GE&R silica nanoparticles manufactured by General Engineering and Research as well as Precipitated Silica manufactured by Shandong Haochuang Material Co. Ltd claims to offer much more resistance to alkalinity than the mentioned limit. Although NaA zeolites have the greatest number of manufacturers, they are susceptible to damage in acidic conditions, but a little modification goes a long way. The advancement is being the CHA zeolites. Substantially in experiment and research phase, ZEBREX developed by Mitsubishi Chemical promises to deliver an efficient membrane material in the field of wastewater treatment. This review paper analyzed the properties of several polymers which was the motivation of my study to analyze these polymers if used in passive OWS devices that can be used in protection of transformers

7. CONCLUSIONS

This paper systematically reviews the properties of potential membrane materials that can be used in industrial waste-water management, particularly in ester-filled transformers. The following are the chief points summarizing this work-

1. Semi-permeable reverse osmosis membranes manufactured by Bluestar (Hangzhou) Membrane Industries Co, Ltd have a solute rejection rate of 99.3%, which can be combined with other membrane materials to make wastewater treatment more effective.
2. Regenerated Cellulose filters are organic and hydrophilic in nature and have high flow rates due to their symmetric structure. Due to their wide range of pore sizes (0.2-8 µm), they can be easily adjusted to assist with ester-water separation.
3. LTA zeolites offer great durability but are approximately 10 times more expensive than a regular polymeric membrane. But combining it with other ultrafiltration membranes can be effective in ester-water separation.
4. More of a recent development, CHA zeolites have a wide range of silicon to aluminium ratio going as high as 11:1, making it less hydrophilic and more stable to acidity as compared to LTA zeolites.
5. Recent advancements in 2D materials like graphene show great potential in effective ester-water separation. Although it is still not commercialized. [13]
6. NFX Nanofiltration membrane and Ultrafiltration PAN (polyacrylonitrile) membrane manufactured by Synder Filtration offer an optimal operating pH range of 3-9.5 at a maximum temperature of 50°C.
7. Amorphous Perfluoro-polymers (APFP) is a hydrophobic polymer that exhibits minimal swelling in water. This will increase the life expectancy of the employed OWS device.
8. Although in the research phase, Mixed Matrix membranes composed of dual nanofillers have proven to improve water flux and surface properties of Polyether sulfone (PES) membrane, which is a popular hydrophilic separation membrane with high flow rates.

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