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Membrane Separation for Recovery of Umami Compounds: Review of Current and Recently Developed Membranes

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Abstract

The goal of this review was to determine the right membrane materials that were the current test to recovery of umami compounds as guidance for the future. Despite the fact that umami characteristic can influence effect too, the right membrane materials are still needed to understand to avoid increase resulting in fouling during membrane processes or better control of membrane fouling. The results from this study showed for the last 20 years of 18 articles suggested that the major of membrane materials to separate umami compounds which are in hydrolyzed of protein or peptides or as a free amino acid such as glutamic, aspartic or glycine still used conventional pressure-driven filtration membrane. The membrane materials were used such as PES, polyamide and cellulose acetate. We discuss the three membrane materials. The study concludes that membrane materials such as thin-film composite polyamide (PA-TFC) have been promising to the recovery of umami compounds.

Keywords: Umami, Membrane materials, PES, Polyamide, CA

1 Introduction

Membrane technology is one of the future separation methods in which technologies are predicted to be used in industries worldwide. The membrane can replace conventional chemical treatments and also clarifiers using heavy equipment that leads to high operating costs and associated with environmental problems. Even though it cannot completely replace conventional treatment technologies and be a stand-alone treatment option (1). Now many techniques of membrane separation have been developed from conventional to modern; pressure-driven, electrodialysis or liquid membranes could be combined.

One of the varieties of membrane separation techniques can be characterized by their membrane pore size (2). Generally, membranes use the principle of molecular size and pore size distribution to separate different materials even (3) other factors also play a role such as electrostatic, molecular or chemical properties of the sample (4).

Recently, membrane technology was used for water purification and waste management. The special membrane is typically used to obtain pure water. Generally, commercially available membranes for pure water were CTA (cellulose triacetate) or CA (cellulose acetate) or derivate, using reverse osmosis technique (5, 6). With technology development in membrane materials, Connell et al. reported that PVDF (polyvinylidene difluoride) membranes for MF (microfiltration) were the best option on the market in 2017 to treat raw water and mine wastewater (7).

So far, the technology was widely used to recovery nutrition or phytochemicals including condensed milk (8), milk protein separation (9), juice clarification and concentration (10, 11), concentration of whey protein (12), color (13), sugar and polyphenols recovery (14, 15) metals (16, 17). Of course, the membrane-type used for nutrition or phytochemicals are different than that for pure water.

The separation of chemical compounds for different applications has become an important industrial operation. Considerable progress continues to be made in membrane technology, and newer applications for existing systems are being discovered as the trend is to create integrated systems that utilize several different types of the membrane within a process. Aware of the fact that membrane separations have great potential, many scientists are dealing with their

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adjustment to the requirements such as flavor enhancer-industry and pharmaceuticals.

This review will discuss and focus just only on membrane materials type selection which scientifically proven based on current studies and research to recovery nutrition and phytochemicals from natural resources such as umami (flavor enhancer) compounds. Umami contained chemical compounds that the major contribution to food palatability (18, 19, 20). Flavor enhancer industry, particularly in the form of the MSG (monosodium glutamate) was growing so fast especially in East Asia (Japan, China, Taiwan, South Korea) and Indonesia (U.S. International Trade Commission, 2013;). However, the glutamic acid production in the industry still used the conventional manufacture, thus it has not been yet used membrane processes separation in the purification stage (21). Until now, publications about membrane separation of umami compounds are still less discussed. The industrials used membrane technology were fixed and existed, such as the sugar industry (22) and dairy industry (23), but the membrane which compatible with umami compounds is still being discovered and proved by research. To obtain the fixed membranes it is still needed a review of the literature in a thesis, dissertation or research paper as a short guide.

The main objectives of this review focus on obtaining the type of membrane materials which can be used to separation of umami compounds from foods or by-products. This is an early review of membrane processes separation of umami compounds. Moreover, studies that led to the development of novel raw materials to umami production are still needed. Dates syrups and cassava starch have been reported to produce glutamic acid by Ahmed et al. and Nampoothiri & Pandey (24,25) *Albertisia papuana* Becc. leaves have been reported of rich in umami compounds (26). Other raw materials such as tea leaves (27) are rich in umami also by-products from wheat (28) could be utilized for glutamate productions too. It is important to be aware of the fact that there is trend sugarcane production mainly in Java, Indonesia (Directorate General of Estate Crops-Ministry of Agriculture-Republic of Indonesia, 2016) in which molasses (by-product) is the only raw material of MSG production.

In membrane separation processes, fouling is a major problem (29). Factors affecting membrane fouling were membrane properties including membrane materials and surface pore size (11). Moreover, the choice of the membrane depends on the application objective, however commonly used membranes are commercially available. This review was performed to evaluate the disadvantage and advantages of membrane materials to recovery umami in the proposed scheme or to improve the membrane separation technique that was done.

2 Material and Methods

Literature review using a research article conducted in any part of the world from 1987 to 2017. The search terms membranes separation, microfiltration, ultrafiltration, nanofiltration was separately integrated with the savory, umami, flavor enhancer, glutamate or glutamic acid, aspartate or aspartic acid, amino acid polar, nucleotides, hydrolyzed of protein, peptides. All authors analyzed the current state on material membranes specification profiles and evaluation to umami separation. A narrative summary of the results is presented.

2.1 Overview of Umami

Generally, umami well known as a flavor enhancer was used in seasoning mainly in Asia in MSG form. Umami was the fifth basic taste in human which feel as savory, brothy and meaty (18). The main chemical compounds contributed to umami were glutamic acid, although a lot of chemical compounds were in synergy, such as 5'-nucleotides mainly IMP, GMP and AMP (30,31,32). Purwayantie et al. have proved that in *Albertisia papuana* Becc (26). contains the 5'-nucleotides. In addition, it is has been proved by Chaudhari et al., that one of the receptors for umami in humans (T1R1+T1R3) is activated by a broad range of amino acids and displays as a strongly potentiated response in the presence of nucleotides (30). The fact in MSG production (commercially), 1% of 5'-nucleotides were added (survey in 2011 on MSG Factory, Mojokerto, East of Java, Indonesia, unpublished). Another amino acid showing similar behavior to glutamic properties was aspartic acid, both of MSG-like compounds. Yamaguchi et al. reported, the characteristic of aspartic acid possesses only 7% of the efficacy of MSG (100% umami level) (33).

Table 1: The Properties of Amino Acid Based Umami Compounds

Properties/ amino acid	glutamic acid (MSG-like)	aspartic acid (MSG-like)	Glycine (sweet)	Serine (sweet)	Threonine (sweet)	Alanine (sweet)
MW (Dalton)	129.12*	114.11*	57.05*	87.08*	101.11*	71.09*
	147**	133**	75**	105**	119**	89**
Solubility (pH):						
pK1	2.19	1.88	3.24	2.21	2.09	2.34
pK2	9.67	9.60	9.60	9.15	9.10	9.69
pKR	4.25	3.65	-	-	-	-
polarity	polar	polar	Non polar	Polar	polar	Non polar
charge	negative	negative	neutral	Neutral	neutral	neutral
Side chains	COOH-group	COOH-group	-	OH-group	OH- group	-
Bonding form	Salt bridge	Salt bridge		H-bond	H-bond	
Vol. classes (A^o)	138-154 (medium)	108-117 (small)	60-90 (very small)	60-90 (very small)	108-117 (small)	60-90 (very small)

Source: Nelson et al. (70)

*data obtained from mas spectrum analysis https://www.seas.upenn.edu/~cis535/.../GCB535HW6b.pdf

** https://www.genomics.agilent.com/files/Mobio/Amino%20Acids_Abbrv_MolWeight_Classifications_2pgs.pdf

Based on molecular and functional points of view both of them have a similarity of non-essential amino acids (34) and neurotransmitters. Glutamate and aspartate were the two neurotransmitters in the central nervous system (CNS) of the brain (35,36), but Herring et al. explained that the aspartate has not been an excitatory neurotransmitter, just the only glutamate was prominent (37).

The solubility of both amino acids is very effected by pH. The pKa values of glutamic and aspartic at acid pH (a carboxyl group), are of 2.19 and 1.88, at base pH (an-ammonium ion) are of 9.67 and 9.60, at side chain group pH are of 4.25 and 3.65, meanwhile the pI (isoelectric point) occurs at pH of 3.22 and 2.77 (34). Thus, both amino acids were very polar and more acidic than other amino acids. Other umami compounds were glycine, threonine, and serin, also contribute to umami benefit in which they act modulator on the glutamic receptor of umami taste (38, 39, 40). The characteristics of umami compounds are presented in Table 1.

Membrane separation of amino acids has been started since 1980, include glutamic acid. The majority of these work aims to recover savory fraction that umami-rich such as peptides. On the other hand, most of umami-rich foods linked with high protein foods or fermented foods (41) such as fermented fish (42) fermented mung bean (43), soy sauce (44), fermented shrimp products in Southeast Asia (45), cheese (46), fermented meats (47), wine (48), sake (49), fermentation of MSG production (50). During fermentation, processes proteolysis or autolysis occurs that generates the predominant tastants of amino acids. Zhao et al. and Y. Zhang et al. explained, the major contribution of taste-active of fermented foods was glutamate, amino acid derivate, and peptides (51,52). Peptides particularly impart umami taste such as α-glutamyl peptides especially pyrutamyl-Pro-X peptides which also produced by pGlu cyclase. Moreover, a number of bioactive peptides find their potential applications in the food or pharmaceutical industry (53, 54).

3 Result

By research methods, we find of 18 research articles in past 20 yr (Table 2) The finding showed and proved about umami separation such as savory fraction, protein hydrolyzed or peptides, the free amino acid (glutamate, aspartate, glutathione, glycine, glutamine) can separate by using membrane filtration. There is three of membrane separation technology for a long time, majority using pressure-driven conventional membrane filtration processes; only two using the technique of Electro Membrane Process or electrodialysis (EMP) by M. Kumar et al. and Doyen et al. and only one using Emulsion Liquid Membranes (ELM) technique by Bhuvaneswari et al. (55, 56, 57).

Table 2: Umami Fraction of Membranes Separation Reported

Objectives	**Membrane Separation Technic**	**Findings**
Selective membranes filtration integrated into a pilot process to obtain the hydrolyzed of spent brewer's yeast (peptides) with different MW and with improving the chemical and nutritional Amorim et al. (58)	Methods: UF and NF Type of membrane: UF (Hydranautics model 3838-30; MWCO 10kDa; Lenntech) NF (PTI; Advanced Filtration, NF 3838/30-FF; MWCO 3kDa Filtration area: UF 7.4m^2 and NF 6.9m^2	The nutrition obtained: 1. Protein and sugar (30-69%) and 20-48% 2. The major minerals: Na and K 3. The freest amino acids: glutamate, glutamine, and alanine 4. Peptides profiles: hydrophilic and hydrophobic usually associated with biological activities
Recover savory fraction of fermented mung bean from two autolysate (Rhizopus and Aspergillus) (43)	Methods: diafiltration-nanofiltration (DF-NF), continuous mode. Membrane separation/pressure-driven operations: MF of 0.2 μm and NF 45PE (MWCO 300Da; Dow Film Tech, USA) Membrane modules: sheet (plate and frame) diameter 20 cm, effective area of 0,036 m^2 Membrane processes: pump motor frequency of 20 Hz (flow rate 7.5L/minute), 23-25^oC; 20bar, Pre-filter in 200 μm filter	DF-NF method on autolysate of Rhizopus/Aspergillus was able to reduce salt at dial volume 0.2 with a resulted composition of concentration of salt 0 and 0.14%, glutamic acid (total protein) 0.64 and 0.32%
Separation taste compounds of *Albertisia papuana* Becc. leaves extract (26)	Methods: stepwise filtration (MF-UF-NF) Type of membrane: MF (PES) UF (PES; MWCO 10kDa, 5kDa; Nadire, Germany) NF (PES; MWCO 4Da; Nadire, Germany) NF (PES; MWCO 1kDa; Sartorius, Germany System membrane: dead end Membrane modules: plate and frame Diameter: 47 mm in diameter	No glutamic acid showed in HPLC analysis when filtration conducted at pH 8.0 (Tris HCL buffer as a solvent)

Objectives	Membrane Separation Technic	Findings
Green process of glutamic acid production (59)	Methods: membrane integrated hybrid reactor system Type of membrane: NF (polyamide; Sepro Co. (USA) Membrane modules: flat sheet, cross flow	High selectivity by using NF helped achieve over 97% product purity of glutamic acid without any need for pH 5 adjustment in a fully membrane-integrated fermentation process
Objectives	**Membrane Separation Technic**	**Findings**
A pilot plant test on the desalination of soy sauce by nanofiltration (60)	Methods: nanofiltration with pretreatment: UF Type of membrane: NF270-4040 (polyamide thin film composite (Dow Filmtech) and Desal-5 DK-4040 (GE Osmonics) Membrane modules: UF tubular modules and NF spiral wound modules (cross flow)	The membrane of NF270 the most suitable for desalination of soy sauce Glutamic and aspartic acid had the highest retention by NF270
To compare the impact of PES and CA materials on peptides selective migration from snow crab by-product hydrolysate (57)	Methods: Electrodialysis with Ultrafiltration Membranes (EDUF) Type of membrane: UF PES MWCO of 20kDa (GE, France) and CA (Spectrum Laboratories Inc., Rancho Dominguez, CA).	1. The most abundant population in a compartment located near the anode for the recovery of anionic/acid peptide fractions and near the cathode for the recovery of cationic/basic peptide fractions were peptides with MWCO ranging from 300 to 700 Da. 2. Peptides with MWCO ranging from 700 to 900Da did not migrate during the EDUF treatment. 3. The recovery of high MWCO (900-20000Da) in compartments located near the anode and cathode only from CA. 4. Peptides desorbed from PES and CA UFM after 6h of EDUF separation had low MWCO and belonged mainly to the 600-700 Da
Amino acid (glutamate and lysine) separation (56)	Methods: electro-membrane processes (EMP) Type of membrane: PES	The separation of GLU–LYS mixture was possible at pH 8.0 and 85% recovery of glutamic acid.
Nanofiltration of concentrated amino acid solution (diprotic amino acid: glycine and glutamine) (61)	Methods: UF and NF Tight UF and commercial polymeric NF Type of membrane: DK (NF 150-300Da; GE (G-5) (UF 1000Da), GH (G-10) (UF 2.5 kDa), NP030 (NF permanently hydrophilic PES 150-300Da, NP010 (NF (NF permanently hydrophilic PES 1kDa) Membrane modules: plate and frame; effective surface membrane area 350cm^2	1. Higher rejection and flux drop over the concentration was observed in alkaline, where the amino acid is present in dissociated form. 2. At low concentration (< 0.2 mol/L) higher rejection for charged.amino acids and more stresses decrease in flux with increasing concentration occurs for negatively charged amino acids 3. At higher concentration, lower rejection for anionic amino acids
Objectives	**Membrane Separation Technic**	**Findings**
The impact of a two-step UF/NF to produce fractionation of fish hydrolysate on two industrial process (62)	Methods: UF and NF Type of membrane: UF (ESP04; modified PES; MWCO 4kDa) NF (PA coated on PES); AFC40NF (Polyamide film; MWCO 3Da) Membrane modules: tubular membranes, diameter 12 mm, surface area 0.033m2. Spec of ESP04: pH range 1-14; max temperature 65oC; max pressure 30 bar; hydrophilicity relative low (PCI Membrane). Spec of AFC40NF: pH range 1.5-9.5; max temperature 60oC; max pressure 60 bar; apparent retention character 60% CaCl2; hydrophilicity relative high (PCI membrane)	The UF fractionation produces a permeate enriched with respect to the FPH smaller than a molecular weight of about 600–750 Dalton, and a retentate enriched in large peptides (above the same MW). Similar behavior is found for the NF fractionation

Objectives	Membrane Separation Technic	Findings
Isolate of umami or savory taste of soy sauce (63)	Methods: step wise of UF (MWCO 10kDa; 3kDa; 5Da) Spec of the membrane: unpublished	Umami and sweet taste-free amino acids and sodium salt are the key compounds of the intense savory taste
Concentration and purification peptide hydrolysates of fish on UF and NF (64)	Methods: UF and NF Type of membrane: UF (MT68; PS, MWCO 8 kDa; PCI) UF (MTP04; modified PES, MWCO 4kDa; PCI) NF (MT04; polyamide/PES, MWCO 300Da Membrane modules: tubular Spec of the membrane: surface area of 0.033 m2 surface area of 0.033 m^2	NF (polyamide/PES, MWCO 300Da) was good for both fluxes and recovery rates to peptides concentration.
Comparative Evaluation of Ultrafiltration Membranes for Purification of Synthetic Peptides (65)	Method: UF (Diafiltration in a cross-flow thin-channel device) Type of membranes: Eight polymer materials membranes with MWCO ranging from 500 to 800 Da 1. Aliphatic alcohol (Zenon Environmental Inc., Burlington, Ontario, Canada) 2. CA (DDS, Nakskov, Denmark); Osmonics Inc. (Minnetonka, Minnesota), Amicon Corp. (Danvers, Massachusetts) and Millipore Corp. (Bed ford, Massachusetts) 3. Regenerated cellulose disks (Amicon and Spectrum Medical Industries (Los Angeles, California) 4. PES 5. PS (Dr. C. Bouchard, Dept. of Chemical Engineering, Ecole Polytechnique de Montreal, Canada) 6. Modified PS flat sheets (proprietary modification) came from Bio-Recovery Inc., Northvale, New Jersey. 7. Fluoropolymer 8. Teflon Membrane modules: plate and frame Spec of membrane: 90 mm diameter disks (effective membrane area = 40 cm^2)	The cellulosic membranes proved to be successful and reliable for the purification of synthetic peptides (hexapeptide (MW 844); insulin (MW 5730). and cytochrome c (MW 12,384) in 5% acetic acid
Amino acid separation of a hydrolysate of Lumbricus rubellus protein	Method: NF Type of membranes: NF (MWCO 1kDa); Millipore, USA The membrane was cleaned with NaOH 0,1N Membrane modules: plate and frame (cross flow)	The best condition for amino acid filtration was obtained on 9 psi, concentration 1 g/L, with amino acid rejection 27-30%
Separation of glutathione and its related amino acids by nanofiltration (66)	Method: NF with pretreatment of MF CA 0.45µm Type of membranes: NTR-7450 (PES, MWCO 1kDa); Nitto Electric Industrial Co. The diameter of the membrane 4.3 cm Membrane modules: dead end	1. NTR-7450 rejected of the electrolytes corresponded to the ratio of their anionic species varying with pH 2. At pH 7.4, glutamic acid rejected almost 100%. 3. In the presence of divalent metal ions, the rejection of glutamic decreased with increasing the concentration of the metal.
Glutamic acid separation by extraction membrane (55)	Method: emulsion liquid membrane Solvent: kerosene Carrier: oleic acid	Using emulsifier tri-ethanol amine to separate glutamic acid more feasible than using HCL and Indian glycol with

Objectives	Membrane Separation Technic	Findings
		condition processes M/E ratio of 0.25 at 150 rpm, external phase pH of 4.0 obtained maximum solute recovery of 63.5%
Glutamine separation from broth fermentation (67)	Method: NF Type of membranes: NTR7450 Spec NTR7450: PES, MWCO 600-800 Da, pH range 1-12; temperatures up to 90oC; max 50bar (Nitto Electric Industrial Co., Japan)	The separation selectivity of Glutamin and glutamic acid was affected greatly by the pH, transmembrane pressure and broth concentration.
Separation of peptides and amino acid with nanofiltration membranes (68)	Method: NF Type of membranes: NF-40 (Film Tech Corporation); Desal-5 (Desalination systems); G-20 (UF; Thin film; MWCO 3500 Da; Desalination systems; GE Osmonics (USA); NTR-7450 (PES; MWCO 600-800 Da; Nitto Electric Industrial Co., Japan); UTC20 and UTC60 (Toray Industries)	Separation of amino acids and peptides was suitable on NF NTR-7450 and G-20 (MWCO 2000-3000Da) but the charged amino acids and peptides were rejected meanwhile peptides and the neutral amino acids permeated through the membranes
Separation of racemic glutamic using cellulose acetate (69)	Method: molecular imprinting technique Type of membranes: CA	The molecularly imprinted CA membranes are applicable to separate between D-glutamic and L-glutamic
Concentration and separation of aspartic acid and phenylalanine in an organic solvent (70)	Method: NF Type of membranes: CA (NTR-1698; Nitto Denko Corporation, Tokyo, Japan), PA-PPSO (Toray Industries, Inc., Tokyo, Japan), polyamide composite (PI-COM; NTGS-2100; Nitto Denko Corporation, Tokyo, Japan) Membrane module: dead-end ((with a diameter of 7.5 cm and an exposed surface area of 32 cm') Membrane process: operated at 40°C; 500 rpm by a magnetic stirrer.	The PA-PPSO membrane with methanol as solvent appeared the most promising to separate aspartic acid meanwhile CA suitable too but the stability was very poor.
The concentration of amino acid (71)	Method: liquid emulsion membrane	A precursor of lubricant (S-GONR) was used as an organic solvent, DBEHPA as a carrier, and Paradox 100 as a surfactant. A 1.5 M H2SO4, a solution was used as an internal aqueous phase.
Recovery of glutamic acid from the fermentation broth (72)	Method: UF, DF (diafiltration) and RO Alat: module-20 UFlRO unit (De Dnaske Sukkerfabrikker (DDS) (Copenhagen, Denmark) Type of membranes: UF (DDS GRIOPP; MWCO 500kDa RO (DDS HR-98); Membrane processes: 0.576 m2 of membrane area, were used for UF and DF; 0.144 m2 of membrane area, were used for RO. cutoff of 500,000 Da was used for UF and DF. The recommended maximum operating pressure and temperature for the membrane were 10 bar and 8O" C, respectively. The pressure and temperature limits for the RO membrane were 80 bar and 8O" C	Separation of glutamic acid from bacterial cells by membrane processing could improve the efficiencies of subsequent evaporation and crystallization processes

Generally, it is not all of the stages in pressure-driven conventional membrane filtration processes was done such as MF first, following by UF and NF, including pore size too, from large to small ones. In fact, by author experience who is worked in membrane filtration in 7 past yr, it has to be done to all of the stage processes, especially the sample test came from the crude extract of plants. If not done, it will cause an increase in fouling seriously, so that it will need more membranes, even

more, if done without cleaning membrane processes, will impact to high costs. The stage of pre-treatment has to done too such as clarification in couple days at lower temperatures or by high-speed centrifuges. Some articles state that MF or UF as a pre-treatment (60,66). We think all of the pre-treatment for the same aims to remove total dissolved solids (TDS) which can affect the effectiveness of membrane used.

Based on Table 2 too, this review only analyzed three kinds of majority membrane materials, a natural polymer such as CA; synthetic membranes as a PES and polyamide-based membranes to the recovery of umami compounds. The membranes present some differences in their performance and structures.

4 Discussion

The membrane materials that using to obtain umami compounds at the MF stage can be from PES (26) or CA (66) with a pore size of 0.2 µm in (43). At the UF stage, the majority of membrane material was PES (20, 78) or by modified PES (62, 64). In fact, in comparison to other membrane materials such as CA, regenerated cellulose, PCS, PES, modified PS, fluoropolymer and Teflon, the best to obtain the purified peptides were cellulosic-based membrane material (65). The Cellulose acetate membrane including produces a lot of peptides which has a molecular weight range 600 to 700 Dalton and 900-20,000 Da when compared with PES (57).

There are three articles using diafiltration (43, 72, 73). This technique as a conventional process to achieve high purification of macro solutes with an economically acceptable flux. Diafiltration in Limayem et al. mean adding continuously pure water to feed volume to make constant (44). Paulen et al., explained that between UF and DF were different techniques and generally (74), UF often combined with DF (75, 35). The last one in NF stage majority were used polyamide-base membrane (26, 59, 60, 61, 62, 70, 76, 78).

4.1 PES (Polyether Sulphone) Membrane

This material has become the most popular membrane for MF, UF or NF stage. Almost of membrane researchers said that the PES membrane is good mechanical strength, excellent thermal and pH stabilities, high flux and reasonable cost compared to the other membrane materials. Unfortunately, almost of the research reported that higher rejection of amino acid (especially of negatively charged) such as umami compounds; glutamic acid, aspartic acid or peptides which umami-rich by using PES (26, 60, 66, 76, 79). Polyethersulfone membrane has low hydrophilic.

It is proved that even PES have excellent hydrophobicity, it fouled more seriously. The hydrophobicity of membranes means the lower hydrophilic properties than other membrane materials such as polyacrylonitrile, CA, polyamide, polyamide-imide (77). Unfortunately, PES and PS have a problem in the fouling of polymeric membranes because of that properties. Alsvik & Hägg also explained that the properties between PES and PS are quite the same (80). It is different from the length of units, PS has longer repeating units than PES and the structure of membranes (Fig 1.).

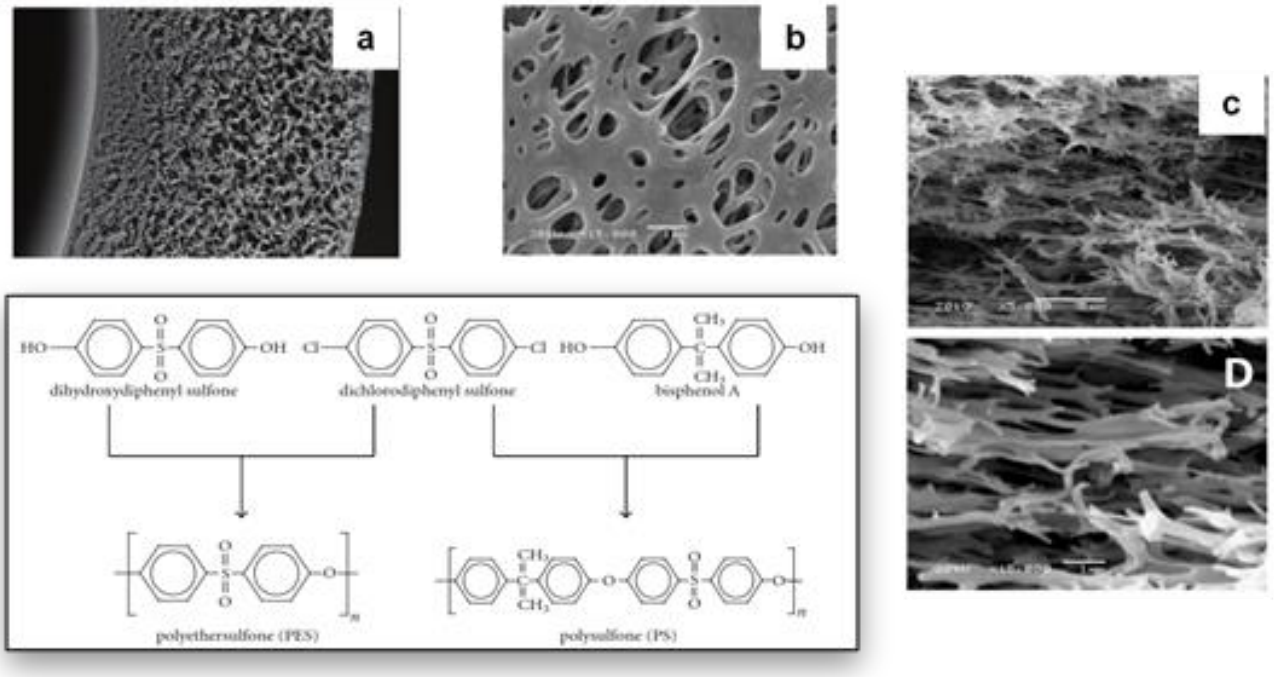

Figure 1: a. The SEM cross-sectional structure of PES (81); b. PES (blank); c. 5,000x; d. 15,000x (82) and the structural formula of PES and PS synthesis (81)

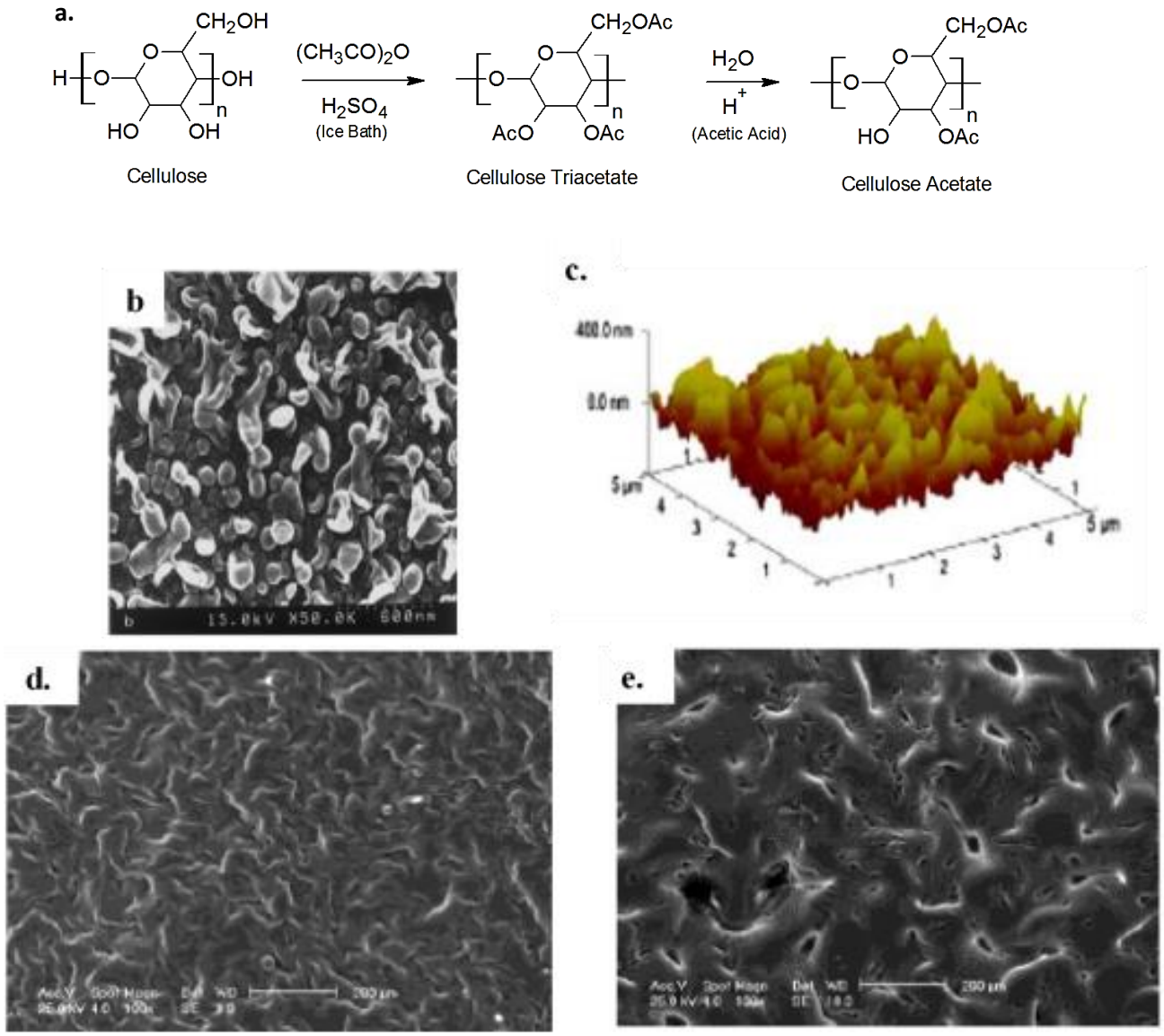

Figure 2: (a) Chemical structure of CA; (b) SEM images of pure CA and the 3D surfaces of CA (83); (c) and (d) SEM of the active surface of CA and SEM of the porous surface of CA (17)

4.2 Cellulose Acetate Membrane

On the contrary, CA membrane properties were different from the PES membrane. This membrane is a green polymer that is produced from raw materials generally from plants. Cellulose acetate or cellulose triacetate is a kind of cellulose-based membrane which is synthesized by a reaction of natural polymer cellulose and acetic acid (42), having properties of higher transparency and toughness among thermoplastics (81) and uncharge membrane properties (84). It is called xylonite sometimes or acetylated cellulose (42). This membrane provided high salt rejection and high fluxes at moderate hydrostatic pressure. The structure of CA is shown in Fig. 2.

4.3 Polyamide-Based Membrane

The membranes of polyamide (PA) being hydrophilic properties material (2); the removed salt and flux effective (85). One of PA membranes is thin-film composite polyamide (PA-TFC) generally using at NF and has been widely applied in many food industrial applications. Polyamide is materials of high tensile strength, abrasion and fatigue resistance, low friction coefficient and good toughness (86). The properties are owing to its better combination between the flux of water and rejection than to other asymmetric membranes. The profile of the PA-TFC membrane consists of three different layers made of different materials (87). However, there is two kind polyester backing substrate, one from PES and the other from PS. Composition of PA-TFC with PES as a polyester backing substrate shown in Fig. 3.

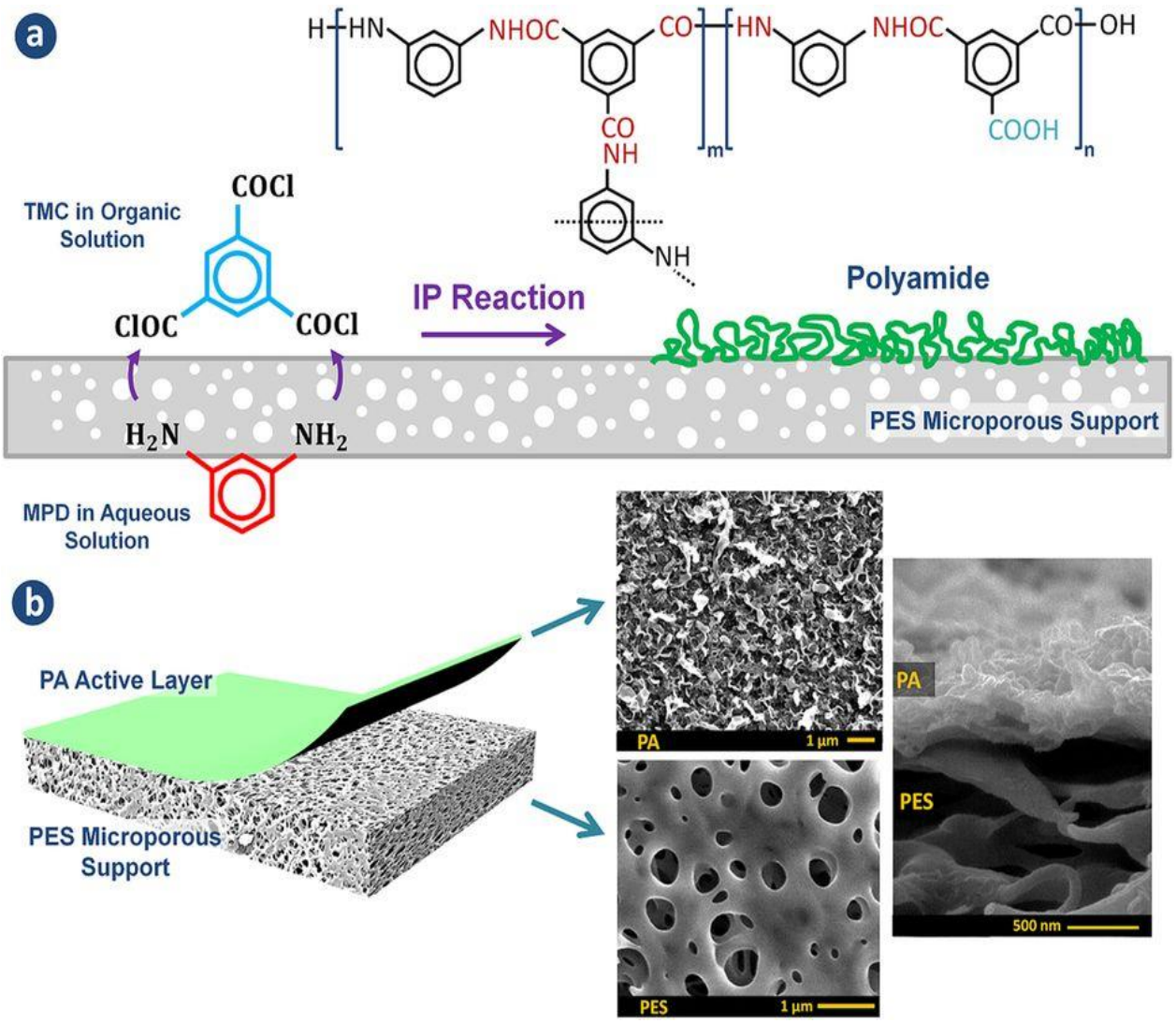

Figure 3: Schematic diagram of PA-TFC (PES) Membrane Synthesis and The Structure with the top and cross-sectional morphologies (83)

Some variant of polyamides membranes were PA-TFC on polyester backing with a PS substrate (78); PA-TFC on polyester backing with a PES substrate (62); aromatic polyamide (60); polyamide polyphenylene sulfone composite; PA-PPSO (70), etc. In accordance with the report of Nady et al., due to intrinsic hydrophobic properties of PES (82), relatively low surface energy and high water contact angle, the PES membrane is vulnerable to adsorptive of fouling. Jeon et al. were reported about the effect of membrane materials and surface pore size on the fouling on PES (11). The fouling of the membrane decreased sharply with increased pore size. Biofouling was occurred on the PES membrane surface due to the interaction between protein or microorganisms (hydrophobic foulants) and the hydrophobic membranes itself. The charge of the membrane such as PES can form the electrostatic interactions between charged proteins and charged membrane (25, 64).

The major of foulants on PES could be microbial metabolites, bacterial cell lysis and un-metabolized wastewater components (88), including proteins, nucleic acids, polysaccharides and other polymers (36, 88, 89). In the same words, Suwal et al. said too, generally the major of foulants were protein, amino acid, and peptide (50). One of the evidence was reported by Doyen et al., the foulants at the surface and or into the pores of the PES were a peptide (57). According to Luo et al. , besides that compounds, the major of foulants could be other chemical compounds than expected due to combined fouling, such as saccharide, organic acid, NaCl too (60). Besides amino acid, glucose could also block the membrane pores by adsorption, or protein adsorption takes place on the membrane surface and due to the inherent hydrophobic characteristics of the membrane.

Generally, one of the causes in fouling was depended on the pH of feed. We agree with Kattan Readi et al. statements

that pH-change as important to electrodialysis process or membrane filtration (90). Both methods when the separation of amino acid, pH-changes play an important role in terms of the efficiency of the process. Due to the zwitterionic character of amino acids, small pH changes may result in significant changes in the charge of the amino acids. pH effect occurs when the separation of l-glutamine from fermentation broth which umami-rich (glutamic acid) that reported by Li et al., (76). In single amino acid solution especially for glutamic acid showed that rejection of glutamic acid increased slightly with the rise of the pH from 4 to 9, and held at about 90% when the pH was higher than 6. Almost all of the glutamic acid in the solutions dissociated into monovalent anions when the pH was higher than 6 and into bivalent anions when the pH was higher than 11. The rejection of glutamic acid was similar to what was expected according to the percentage of glutamic acid monovalent anions at a pH of 6–8. Since the PES membrane (NTR7450) pore size is larger than the Stokes radius of glutamic acid (meanwhile glutamine which has a Stokes' radius of 0.28 nm (91), the results indicated that the rejection of glutamic acid was mainly governed by the charge effect when the pH varies from 6 to 8. The glutamic acid phenomena are different from glutamine so that the PES membrane (NTR7450) can reject more than 90% of glutamic acid and permit almost 85% of glutamine to pass through the membrane when pH is adjusted to about 7. The separation selectivity of glutamine and glutamic acid by the PES membrane as a function of the pH. Glutamine was polar and uncharged as like other umami compounds etc, threonine and serine, meanwhile, glycine was nonpolar, but was different from glutamic or aspartic of amino acid which is polar and having a negative charge.

Gotoh et al. reported that at a pH of 7.4 conditions, glutamic acid rejected almost 100% by using PES (66). This membrane is negatively charged due to the fixed sulfonic anions of the separation layer. So, the pH of a feed solution is expected to affect the rejection of the membrane for the amphoteric electrolytes. Kovacs & Samhaber, conclude that higher rejection and flux drop over the concentration was observed in higher pH range, where diprotic amino acids are present in dissociated from (61). One of the diprotic amino acid tests was glycine, amino acids that contribution to umami taste (6). Purwayantie et al. reported too that when has been done separation of taste compounds using PES membrane in buffer Tris HCl of pH 8.0 in a crude extract of *A. papuana* Becc., it has not been saw of the glutamic acid detected by HPLC. The research conclusion Kovacs & Samhaber could explain the phenomena of research results from Purwayantie et al. (26, 61). The higher rejection and flux drop over the concentration was observed in alkaline, where the amino acid is present in dissociated form. It could happen when glutamic acid in alkaline (pH 8.0) in dissociated form (the net charge is -1). The fact, it is nature characteristic because severe amino acid, protein, and peptide (especially having amino acid negative charge residue) were umami-rich.

One of the foulants according to (92) were metals and one of the metals as main fouling (10%) detected on membrane autopsies was Fe (67.7%). Umami compound's behavior especially glutamic and aspartic acid which can be complexed with metal especially on pH alkaline was clear (28, 93). So that, if the feed were umami-rich and contained metal ions which separation on the PES membrane in alkaline pH, the complexes could be blocking the pore of the membrane and could not be through the pore of the membrane.

Besides of PES membrane, the CA membrane was used for a long time ago (32,70). CA membrane always used to obtained peptides fractions and protein on biomedical application, mainly to recovery hemoglobin and BSA including alanine of amino acid (17); bioactive peptides fractions such as an anticancer peptide from snow crab by-product hydrolysate (57), synthetic peptides (73), acid and basic peptides (26). In accordance with (73) statements that cellulose-based membranes are good compatibility with peptides or proteins. The cellulosic membranes were proved to be successful and reliable to the purification of peptides with having MW 12,384Da. Interestingly reported by Yoshikawa et al., that glutamic acid from racemic amino acids could be separated (resolution) by using the CA membrane (32). It means that the glutamic suitable separated using CA.

Jeon et al. reported that the hydrophilic CA membranes to have a lower fouling potential than other hydrophobic membranes (e.g., PES) (11). The air contact angles of CA and PES were 121° and 115°, respectively. Thus, the hydrophobicities of the materials increase in the order CA > PES. So, hydrophilic membrane materials are preferable for reducing membrane fouling. Sun et al., indicating that the foulant in the CA membrane when separated protein staining dyes and BSA, only smaller aggregates protein formation (94). When using DLS (dynamic light scattering) test, a large protein aggregate was confirmed but using SEC filtration and native-PGE analyses showed BSA dimmer did not play in the fouling. It is clear that CA was not significant fouling by protein. The fouling occurs in the isoelectric pH of BSA (4.8). The BSA and the small aggregates being uncharged, would have the highest tendency to reversibly associated with more strongly held foulant. When in pH 6.9 the foulant easily is removed. It is indicating that the foulant in the CA membrane was pH-dependent too, but no report that one of the foulants on CA was umami compounds. The conclusions, the hydrophilic of CA showed good mitigation of membrane fouling and the membrane pore size had no significant effect on fouling mitigation.

Compared by polyamide-base membranes that are being reported commonly used for RO (95) especially to obtain pure water (96) showed umami productivity better used than cellulose-based membranes. (70) have been done concentration and separation aspartic acid using PA-PPSO composite compared with CA membranes. The results conclude that the PA-PPSO membranes combined with methanol are the most performance of membrane separation. It is proved too by Vikramachakravarthi et al., who has been obtained the umami compounds as glutamic acid achieved over 90% by using PA-TFC (PS) (78). The result of glutamic acid in Vikramachakravarthi et al. was 0.95g/g compared with the result of amino Nitrogen by Lou was 0.008g/ml by using aromatic polyamide membranes (60, 78). Glutamic acid is one of the amino nitrogen. Reported by Bourseau et al., (62) that PA-TFC (PES) still forming fouling while membrane cut off (MWCO) is well affected to obtain fish protein hydrolysates. In line with PA-TFC (PES) to purification blue whiting of fish

peptide hydrolysates using a membrane having MWCO 300Da (60) while MWCO 4000-8000 to fractionating and MWCO 20kDa to the recovery of non-hydrolyzed protein and enzymes (59).

5 Conclusion and Outlook

In 20 past years, still, the majority using pressure-driven conventional membrane filtration as a green process of umami production. The processes were done with a combination of stage pre-treatment (clarification or centrifugation) with stepwise MF-UF/DF-NF. The material was used for years was PES but generally was high rejected for umami compounds (glutamic, aspartic acid, peptides rich umami). It means that the umami compounds could be as a foulants adsorption in the membrane. The pH effect could be trigger building a form of fouling on PES was clear. The effect of pH can trigger interaction between umami compounds that having some functional groups with metals too. Their anions of glutamic acid, other amino acids or derivatives are versatile ligands, and the leading idea to protect the N- or C-terminus of amino acids by metal ions or by metal complexes (55). Although PES can be considered as a model membrane material, it is widely used for commercial MF and UF (97). Now, PES is extensively used for special applications when protein adsorption is not a significant problem (73). In contrast, the CA membrane still rarely uses separation in umami, still focuses on peptides separation with high of WM. Although any foulants on the CA membrane are pH-dependent too, the future need for evaluation or autopsy studies varying of the cellulose-based membrane. However today, the polyamide-based membranes have been used to replace of CA membrane. We conclude that the suitable to obtain the umami compounds was PA-TFC but the variant of polyamide could affect the productivity of chemical compounds from membrane processes with especially using the suitable MWCO of membrane.

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