Membranes for separation of ethanol-water mixtures via pervaporation: a systematic review

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Dedicatoria

Dedicado a Leticia Bayona Jaime y a Harvey Enrique Criado Angarita, cuyo profundo e inmarcesible amor me ha enseñado que aún de las nubes negras caen aguas limpias.

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Resumen

Título: Separation of ethanol-water mixtures via pervaporation using membrane technology: A systematic review [*]

Autor: Harvey Criado Bayona[**]

Palabras Clave: Pervaporación, Membranas, Separación etanol-agua.

Descripción: En este trabajo se realizó una revisión sistemática acerca de las membranas más adecuadas para su uso en separación de mezclas etanol-agua por pervaporación. La definición de la pregunta de investigación se realizó mediante la estrategia Population/Outcomes de dos elementos. La búsqueda y clasificación de la información se realizó de acuerdo con el protocolo PRISMA. Se realizó un análisis bibliométrico para obtener un panorama acerca del número de artículos publicados y citados por año, autores más relevantes, países y revistas con mayor número de publicaciones. Se analizaron los avances, desarrollos y desafíos en materiales para síntesis de membranas utilizadas para deshidratación de etanol mediante pervaporación. Se pudo concluir que el uso de materiales orgánicos con adición de inorgánicos, ya sea en matriz compuesta o híbrida, es el enfoque de síntesis a futuro. Se estudiaron los efectos de los parámetros de operación en el rendimiento de las membranas, clasificándolas según su mecanismo de afinidad con el agua, concluyendo que la concentración de etanol en la alimentación afecta tanto al flux como al factor de separación, y lo hace según el mecanismo de afinidad con el agua. Por otra parte, se concluyó que el aumento de la temperatura favorece el flux de la membrana, pero reduce su factor de separación tanto para membranas hidrofílicas como hidrofóbicas.

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[*] Trabajo de Grado

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Abstract

Title: Separation of ethanol-water mixtures via pervaporation using membrane technology: A systematic review [*]

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Key Words: Pervaporation, Membranes, ethanol-water separation.

Description: In this work, a systematic review was carried out about the most suitable membranes for use in separating ethanol-water mixtures by pervaporation. The definition of the research question was based on the Population/Outcomes strategy with two elements. The search and classification of the information was realized according to the PRISMA protocol. A bibliometric analysis was carried out to obtain an overview of the number of articles published and cited per year, the most relevant authors, countries and journals with the highest number of publications. The advances, developments and challenges in materials for the synthesis of membranes used for dehydration of ethanol by pervaporation were analyzed. It was possible to conclude that the use of organic materials with the addition of inorganics, whether in a composite or hybrid matrix, is the synthesis approach for the future. The effects of the operating parameters on the performance of the membranes were studied, classifying them according to their affinity mechanism with water, concluding that the concentration of ethanol in the feed affects both the flux and the separation factor, and does so according to the affinity mechanism with water. On the other hand, it was concluded that the increase in temperature favors the membrane flux, but reduces its separation factor for both hydrophilic and hydrophobic membranes.

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Introduction

The production of bioethanol reaches a strategic interest in Colombia, due to the great potential for producing it based on the sugar cane crop, one of the strongest sectors of the colombian economy since its cultivation can occur throughout the whole year. This natural availability has been the basis for the increasing production of bioethanol in the last ten years in the country (Portafolio, 2011). Nevertheless, for the most profitable uses of the ethanol, such as in gasoline blended fuels to improve octane ratings (Berlanga et al., 2011) or in pharmaceutical synthesis (Uyazán et al., 2004), a high level of purity is required.

The conventional distillation is the most used process for ethanol-water separation. However, owing to physico-chemical restrictions, through this route it is not possible to obtain ethanol with a purity level higher than 95.6%. For overcoming these limitations, extractive or azeotropic distillation are usually employed, which are very expensive processes, that usually involve dangerous solvents and high energy costs due to the use of multiple distillation units. Therefore, other alternatives are being sought from the technical and economic perspective (Wee et al., 2008).

In these last years, the pervaporation has been considered as a promising candidate to overcome the azeotropic limitations in the separation of ethanol-water mixture. It is a unit operation in which two liquid species are separated (Peng et al., 2011c). Due to their selective adsorption and diffusion across the membrane. It generates great technically advantages, highlighting that the permeate flux is independent of the feed pressure (Noble et al., 2004).

On the other hand, there are no limitations by the thermodynamic liquid-vapor equilibrium of the species (L. Wang et al., 2020). The latter allows getting a high separation degree at low temperatures and at atmospheric pressure. Consequently, it is possible to obtain a reduction of 40-60% of the associated energy costs of the separation when compared to the traditional distillation routes (Wee et al., 2008). Further, pervaporation stands as an interesting alternative from the ecological point of view since the dangerous solvents, as the benzene and toluene, are avoided, allowing ethanol to be used in the pharmaceutical production and as ingredient or precursor in the food industry Additionally, the membranes can be recycled through a desorption cleaning operation making the process more sustainable (Alamaria et al., 2016).

Nevertheless, it is should be mentioned that the performance of the pervaporation is determined by the membrane design through the controlling of synthesis conditions and the choice of the material (Achiou et al., 2018). Particularly, the adequate synthesis parameters can increase the selectivity and the permeate flux, as well as prevent the formation of the structural defects that affects the membrane performance.

Based on the above premises, the objective of this work was to present a systematic review of the distinct scientific developments in this field in the last ten years, based on the PRISMA protocol, (Moher et al., 2009) with the purpose to approach the advances in the development of membranes for ethanol-water separation via pervaporation and establish the challenges and future perspectives.

1. Objectives

1.1 General Objective

To perform a systematic review related to the development of membranes for using in ethanol-water separation systems via pervaporation.

1.2 Specific Objectives

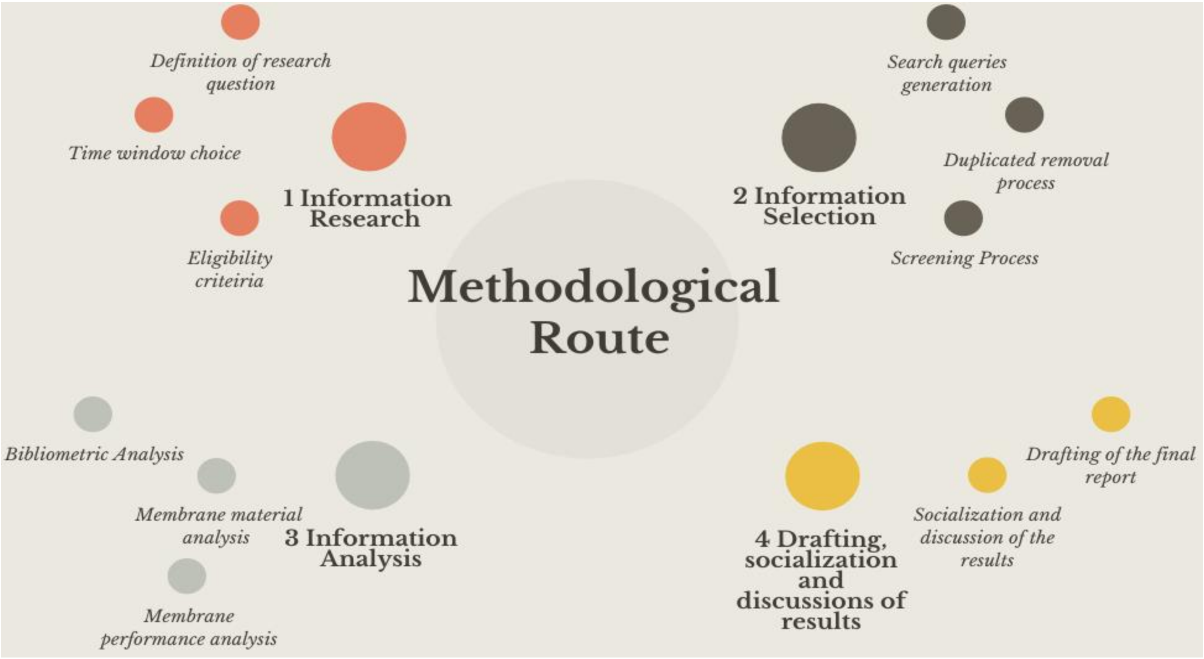
1. To perform a bibliometric analysis of scientific publications related to the employment of membranes for ethanol-water separation systems via pervaporation.
2. To identify the scientific and technological advances in the field of study.
3. To establish the challenges in the field of study that will serve as the basis for planning future research proposals.

2. Materials and Methods

The methodological route is disclosed in the flowchart of Figure 1. It presents the step-sequence for making the systematic review. The review was carry out in accordance with the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) statement (Moher et al., 2009). The PRISMA is a 27 adaptable items protocol that establishes the methodological sequence for making an investigation. It allows obtaining outcomes without biases or blind points based on the selection of the information by means of an eligibility criteria, the screening classification process and the critical analysis of the results obtained, and guarantees that topics of interest are addressed in a wide, comprehensive, and impartial way.

Figure 1

Flowchart of the methodological route employed



2.1 Information Research

The research question was formulated through the Population/Phenomena (PO) two elements format strategy (Munn et al., 2018) that is shown in Table 1. The research question was affirmed as follows: *What are the suitable membranes for ethanol-water separation via pervaporation?*

Table 1

The Population/Phenomena-Outcomes (PO) framework for research question

P (Population/Phenomena)	Membranes
O (Outcomes)	Membranes with better performance to use in ethanol-water separation via pervaporation with a viable synthesis process for industrial implementation.

This systematic review covered from 2011 to June 6th, 2021, since the last literature reviews carried out on this subject were published in 2011 (Bolto et al., 2011a; Peng et al., 2011a). Table 2 displays the eligibility criteria used to include the papers for the systematic review according to the PRISMA protocol. The search strategies were also restricted to english-language publications.

Table 2

Selection criteria employed for the systematic review

Inclusion Criteria	Exclusion Criteria
Studies about synthesis of membranes for ethanol-water separation via pervaporation.	Patents, letters, editorials, thesis, dissertations, and congress blogs.
Studies concerning the pervaporation performance of membranes for ethanol-water separation.	Studies not related to the pervaporation or not focused on ethanol-water separation.

2.2 Information Selection

The databases chosen for the information selection were Web of Science (WOS), Scopus, Springer, ACS, and Taylor & Francis because they are the most relevant ones for engineering and physical-chemistry research (Badia, 2018). The research equation was defined in the corresponding format in every database. The studies must include the following words in the title: *membrane, ethanol,* and *pervaporation.* Later, all documents were downloaded and imported into Mendeley, that is a reference management software, to exclude the duplicated studies. The filtered articles were then introduced into a free software named Rayyan (Johnson & Phillips, 2018; Ouzzani et al., 2016) for a screening process that consists in a systematic selection based on the title and the abstract of the papers; the objective was to discard the papers that did not comply with the following exclusion criteria: i) studies with insufficient data or information; ii) studies with wrong labeling in the title, abstract or keywords; iii) studies not focused on water-ethanol mixture; iv) studies not focused on pervaporation process; v) studies with non-compatible approach about the research question.

2.3 Information Analysis

The information collected was analyzed by means of a bibliometric analysis. The data were organized by number of publications per year, countries, authors, number of citations per year and journal name employing Excel and VOS Viewer. Once the information was classified, the analysis of the membranes was carried out, from their material, as well as their performance, for identifying the scientific and technological advances in the field of study, as well as to pose the future challenges that will serve as the basis for planning future research proposals.

3.Results and Discussion

3.1 Information Selection: PRISMA Protocol Results

The results of the computerized systematic search are shown in Table 3.

Table 3

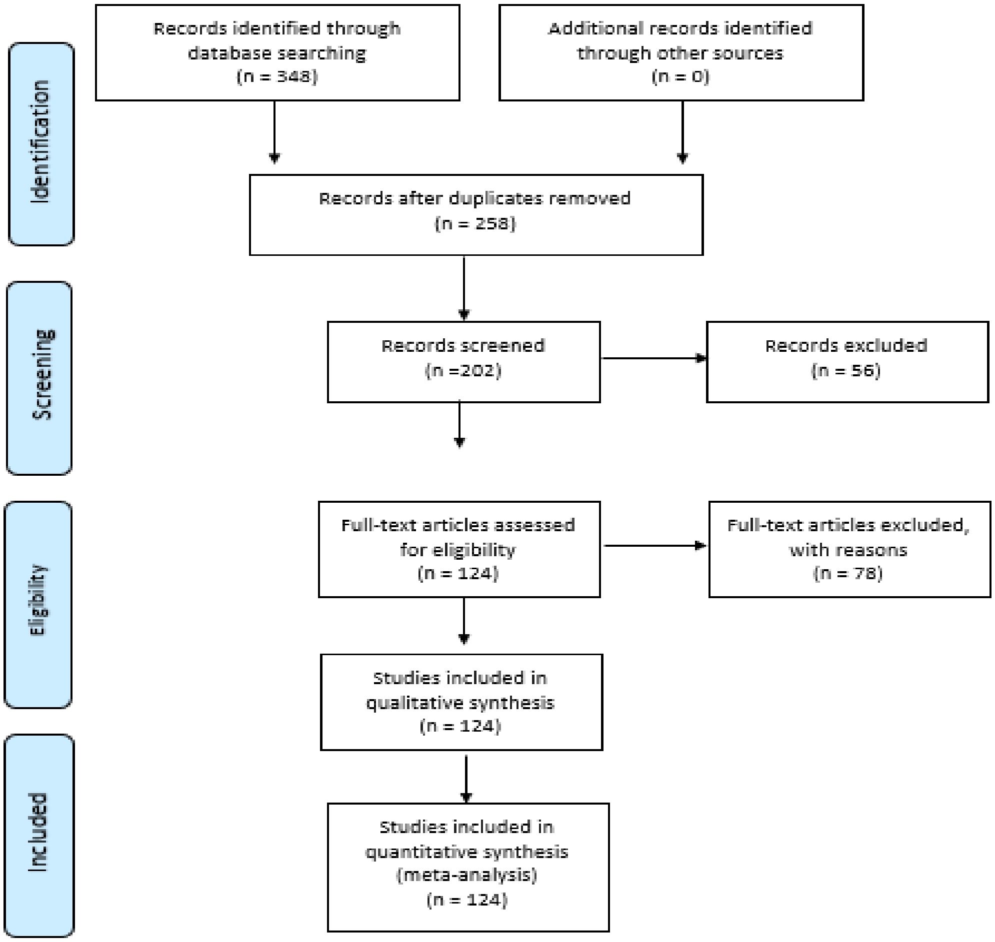
Database search strategy and results

Database	Search Queries	Results
Scopus	(TITLE ("pervaporation" AND "membrane") AND TITLE ("ethanol"))	191
Web of Science	TI=("pervaporation"and"ethanol"and"membrane")	68
ACS	[Publication Title: ethanol] AND [Publication Title: pervaporation] AND [Publication Title: membrane]	11
Springer	[Publication Title: ethanol] AND [Publication Title: pervaporation] AND [Publication Title: membrane]	69
Taylor & Francis	[Publication Title: ethanol] AND [Publication Title: membrane] AND [Publication Title: pervaporation] AND [Publication]]	9
Total		**348**

A total of 348 articles were found from the search strategy and after removing the duplicates ones, the number of studies was reduced to 258. Afterwards, 134 articles were excluded according with the selection criteria of the screening process, so that 124 articles were finally selected for making the bibliometric analysis. To sum up, Figure 2 presents a flowchart that summarizes the search and selection process developed according to PRISMA protocol. Consequently, a total of 124 peer-reviewed papers were selected from 348 originally compiled through the search queries

Figure 2

Flowchart of the article selection process.



3.2 Bibliometric Analysis

From the selected articles a bibliometric analysis of the scientific literature was performed. The articles were completely read and analyzed to provide a clear outlook of the research progress in membranes for ethanol-water separation by pervaporation.

The countries with more articles published in research of membranes for ethanol-water separation via pervaporation are listed in Table 4. It is clear the influence of the Asian countries, where China is consolidated as the country with the largest number of publications in this field, with a total of 51 works published by Chinese researchers, which represents 41.3% of the total reviewed.

Table 4

Countries with more articles published about ethanol-water separation via pervaporation

Country	Number of Papers	Country	Number of Papers
1-China	51	6-USA	5
2-Thailand	6	7-Japan	4
3-India	6	8-Brazil	4
4-Taiwan	5	9-Singapore	4
5-Iran	5	10-Belgium	4

Furthermore, Table 5 provides a list of the most relevant authors according to the number of publications. As expected, Asian countries are the most productive. Li, J. is the most published author with 12 papers followed by Zhang, Y. and Xiao, Z. with 10 and 9 publications each, respectively.

Table 5

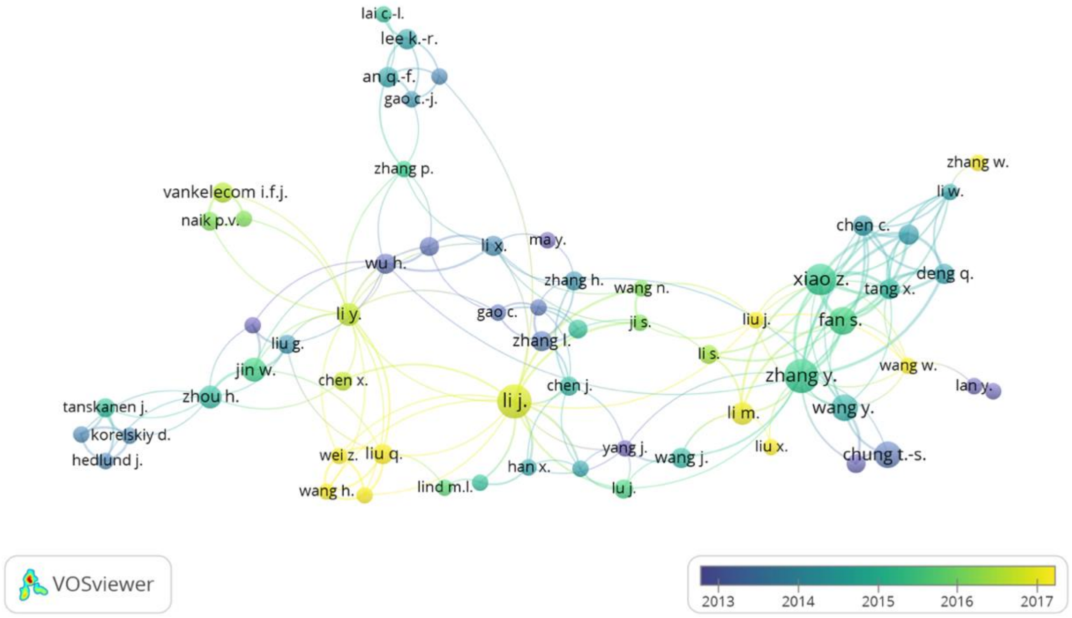
List of authors with the highest number of publications related to membranes for ethanol-water separation via pervaporation

Author	Number of Papers	Author	Number of Papers
Li, J.	12	Chung, T.-S.	7
Zhang, Y.	10	Zhou, H.	6
Xiao, Z.	9	Li, Y.	6
Wang, Y.	7	Li, M.	6
Fan, S.	7	Jin, W.	6

Moreover, Figure 3 illustrates the link between the authors through an overlay visualization generated from a meta-data analysis of the papers using VOS Viewer free software. In this figure, the size of each node is proportional to the co-occurrence of the citation between the author, and the color of each node represents the year of highest citation for each author. It shows that Zhang, Y., Xiao, Z., and Fan, S. have been closely participating in membrane research for ethanol-water separation by pervaporation. At the same time, Li, J. has made significant contributions in recent years. All three are consolidated as the most influential author in the research field.

Figure 3

Co-authorship overlay visualization map during 2011 - 2021



In particular, the study published by L. Le *et al*. in 2011, titled “*Pebax/POSS mixed matrix membranes for ethanol recovery from aqueous solutions via pervaporation”* (Le et al., 2011), was the most cited paper with 142 citations. Secondly, the articles titled “A review of membrane selection for the dehydration of aqueous ethanol by pervaporation ” (Bolto et al., 2011b) and “*A Review of Membrane Materials for Ethanol Recovery by Pervaporation”* (Peng et al., 2011a)*”* follow as the next most referenced with 126 and 115 citations, respectively, as it is shown in Table 6.

Table 6

Articles about ethanol-water separation via pervaporation with more citations

Author	Citations	Ref
Lieu Le et al.	142	(Le et al., 2011)
Bolto et al.	126	(Bolto et al., 2011b)
Peng et al.	115	(Peng et al., 2011a)
Zhang et al.	71	(Chai et al., 2015a)
Kudashev et al.	70	(Kudasheva et al., 2015a)
Khan et al.	68	(Khan et al., 2018a)
Castro-Muñoz et al.	58	(Castro-Muñoz et al., 2019)

Finally, Table 7 summarizes the top five of journals with the highest number of publications related to membranes for ethanol-water separation by pervaporation. "*Separation and Purification Technology*" and "*Journal of Membrane Science*" are the main journals based on the number of published papers and the citations of them.

Table 7

Main journals for publishing about membranes for ethanol-water separation via pervaporation

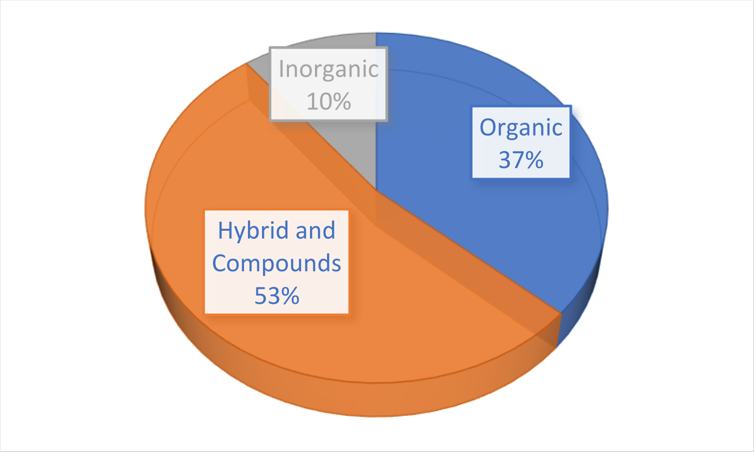
Journal	Citations	Published Papers
Journal of Membrane Science	692	15
Separation and Purification Technology	469	20
Desalination	252	6
Journal of Applied Polymer Science	116	6
RSC Advances	96	7

3.3 Membrane Materials Analysis

The membranes can be classified in three categories according to the nature of their precursors: i) organic membranes, ii) inorganic membranes and iii) hybrid and compound membranes. Figure 4 exposes the distribution of the kind of membranes used in the papers published over the past 10 years. The results indicate a preference for using hybrid/compound membranes compared to the organic and inorganic ones. Indeed, this reflects the preference for the polymeric sources as exclusive membrane material or in a hybrid or compound matrix.

Figure 4

Distribution of membranes created according to their nature of the synthesis materials



The organic membranes are based on polymeric structures and are very attractive due their low fabrication cost (Namboodiri & Vane, 2007), and their suitable qualities for their use in pervaporation i.e. great selectivity and good permeate flux (Y. L. Liu et al., 2007). The most common examples of source for these membranes are polydimethylsiloxane (PDMS) (Han et al., 2016a; Naik, Bernstein, et al., 2016; Yadav et al., 2013), polyvinyl alcohol (PVA) (Chaudhari et al., 2021; Choudhury & Ray, 2021a; Inthavee et al., 2011; Namboodiri & Vane, 2007), polyacrylic acid (PAA)(Ang et al., 2020; P. Zhang et al., 2015), polyurethane (Lishui et al., 2017; K. Zhang et al., 2017), chitosan (Fini et al., 2018a; J. Wang et al., 2015), among others.

However, the organic membranes present important defects for their implementation in a continuous process. Firstly, the polymeric chains utilized for their fabrication present a reduced thermal stability in comparison with the inorganic materials (Van Veen et al., 2001). Consequently, the feed and the operational temperature must be lower than that for the ceramic membranes. On the other hand, the chemical stability of the membrane can be a serious trouble because, if the polymer chains losses their morphology and structure in the presence of organic solvents like ethanol, the membrane can present critical cracks in the continuous operation and spoil the whole process (Peng et al., 2011a).

Swelling is another critical problem. It consists in an obstruction that occurred in the interstitial spaces when the sorption rate is higher than the diffusion one. This effect is exclusive of the polymeric matrix due, partially, to their flexible structure compared with the ceramics (Ma et al., 2012). Swelling can deform the membrane through the creation of large defects in the structure. Therefore, a non-controlled flow tries to pass across the membrane provoking an increase of the total flux but with an important selectivity loss (Kaewkannetra et al., 2011a)(Ma et al., 2012). Further, in critical cases, the structure can break with catastrophic consequences for the entire process.

For the measure of the swelling degree (DS), the membrane is immersed into a specific ethanol-water solution for a time lapse and is dried. Afterwards, comparing the mass before and after the immersion-drying process, is possible to establish the amount of liquid inside the membrane owing to the formation of clusters in the polymeric structure (Jafarinasab et al., 2015a; L. Ji et al., 2015a; P. Zhang et al., 2015). The test outcomes the DS of the membrane by the Equation 1 (L. Ji et al., 2015a; Rokhati et al., 2016):

$$Degree\ of\ Swelling\ (DS) = \frac{W_s - W_d}{W_d}\ x\ 100 \qquad (1)$$

where W_d and W_s refers to the weight of the membrane dried and swollen respectively.

The saturation of the immersion solution simulates the features of the feed stream in the real pervaporation conditions (C.-H. Ji et al., 2016). Thus, the maximum desirable degree of swelling is relative to the specific composition of the selected solution. For this reason, in some situations a DS around 20% is admissible (C.-H. Ji et al., 2016) and, in other cases, the maximum acceptable value is less than 5% (Rokhati et al., 2016).

The swelling phenomena are induced by membrane structure, but also by the operating conditions. The feed stream features play an essential role, due to that the temperature and the solute concentration directly affects the probabilities of occurring swelling in the membrane. A high feed temperature can trigger in an excessive evaporation of the solute by means of the diffusion (Magalad et al., 2011a). The non-controlled phase change causes and increase of the occupied volume and hinders the correct transition of the solute across the membrane. The excessive concentration in the feed also produces swelling because the flux induced is oversized for the permeate capacity. When this occurs, solute clusters are formed, in vapor or liquid phase, they force the membrane for passing through on it, generating voids that induce a non-desirable increase in the mean pore size resulting in loss of selectivity (Le et al., 2012), (Sunitha et al., 2012; Zhan et al., 2012).

On the other hand, the intensity of the pervaporation process also affects the swelling. The increases of the numbers of cycles (batch process) or in the operation time (continuous process), generates wear, resulting in a decrease in the stability of the structure and inducing the formation of solute clusters (C.-H. Ji et al., 2016).

To improve the swelling resistance many strategies were studied. The most extended is modifying the structure of the membrane incorporating rigid elements into the polymeric matrix as zeolites nanoparticles (Khan et al., 2018a; Mao et al., 2019; Yin et al., 2017) or MOFs (Jiang et al., 2021; Marti et al., 2015; S. Wang et al., 2017); or increasing the crosslinking in the membrane (Ahmed et al., 2011; Choudhury & Ray, 2021a; C. Liu et al., 2021). Materials as the graphene

(Choudhury & Ray, 2021b; Dharupaneedi et al., 2014; Hieu & Duy, 2017) and the polybenzoxazine (Chuntanalerg et al., 2015; Pakkethati et al., 2011; Pulyalina et al., 2015) presents excellent performance in terms of selectivity and total flux and helps with the swelling resistance improvement by improving the crosslinking.

Another main group comprises, the inorganic membranes, also called ceramics (Peng et al., 2011c). The ceramic materials have a more defined structure pattern in comparation with the polymers due to their crystalline structure, getting a more symmetry in the morphology. Therefore, they have a better mechanical resistance, which is a required feature for the implementation of the membranes in the industry due the high fluxes involved (Achiou et al., 2018). Additionally, the ceramic membranes do not present the phenomenon of swelling, thanks to their rigid structure so that the solute cannot deform the interstitial spaces in the permeate process in normal operational conditions (K. Li, 2007). In addition, many studies prove that pressure gradients used in pervaporation are not enough to induce deformations in ceramic membranes (Achiou et al., 2018; Leppajarvi et al., 2015; Subaer et al., 2020). Another advantage is that the inorganic membranes are not sensitive to the presence of organic solvents (Subaer et al., 2020). Thus, it is possible to use these membranes for a longer period of time, resulting in cost saving due to greater spacing between maintenances. Furthermore, in terms of thermal stability, the ceramic membranes exhibit better performance than the polymeric ones (K. Li, 2007) making the operation more secure in high feed temperatures range.

Unfortunately, the inorganic membranes have some important disadvantage. The most evident is their lower flexibility, hence it is indispensable the use of a support (Y. Liu et al., 2015). Consequently, it can affect the permeate capacity and decrease the total flux of the pervaporation system (Zhou et al., 2012). Another trouble derived from the support is their possible separation of the membrane leading to a critical failure during operation. For this reason, the adhesion between support and membrane is an important issue to consider in the synthesis process. In this regard, the correct adhesion of the membrane depends on the chemical affinity with the support. Generally, similar nature structures present better results (Karp et al., 2018a).

The other crucial drawback is the elevated production cost of the inorganic membranes. This is associated with the high temperature requirements in the fabrication process to obtain the characteristic crystalline structure (Achiou et al., 2018; Wee et al., 2008). Indeed, the majority of the process requires temperatures between 400ºC and 1000ºC (Leppäjärvi et al., 2015). Likewise, the cost of the support and its functionalization increase the production costs. Some are made from inexpensive precursors as the alumina based (Fini et al., 2018a; Ma et al., 2012; Zhou et al., 2012) but other materials as stainless steel (Gao et al., 2020) or sintered structures (Peng et al., 2011a) can be very expensive. For these reasons, the use of pure inorganic precursors for membrane manufacturing is not the preferred option. In fact, their use as upgrade material in the hybrid membranes is more common and viable, especially to avoid swelling phenomenon.

The last group includes the hybrid and the compounds that are the most attractive category of the membranes, but also the most complex in their fabrication. These membranes pretend to modify the approach of the materials study for pervaporation, making a synergy between organic and inorganic sources for improving the advantages of each one and minimize the limitations. In recent years, many studies were done to create interesting combinations (Magalad et al., 2011b; Mokhtarzadeh et al., 2020; Naik, Wee, et al., 2016) utilizing inorganic complements in organic supports for overcoming the support limitation of the ceramic membranes and improve the mechanical and chemical resistance of the polymeric ones (Khan et al., 2018a; Kudasheva et al., 2015b; Vinu et al., 2018).

However, the main trouble of the compound membranes is the complicated interaction between the organic and inorganic materials. Usually, ceramics and polymers do not have a good chemical affinity owing to their different nature. In terms of structure, the crystalline pattern of the ceramics is very different from the amorphous morphology of the polymers. This entails in adhesion problems and therefore, ruptures and failings in the operation. In order to improve the interaction, many studies suggest the use of mediums, as guar gum, for creating a more consistent structure that guarantees the non-separation during a large exposition time (Han et al., 2016b; Wu et al., 2015a; Zhan et al., 2012). Nonetheless, another method highly suggested is the crosslinked technology that creates a more stable union of the inorganic and the polymeric components (Bolto et al., 2011a; Borisov et al., 2018; Strunck et al., 2020a). The use of nanoparticles that serve of "bridge" is also a trend topic on the search for solving the adhesion problem (Lan et al., 2016; Le et al., 2011; Q. Liu et al., 2019). All the methods used for improving the union of the organic and inorganic components search the same outcome: guarantee the non-separation of the parts of the compound membranes. This is shown in the chemical stability in presence of organic solvent, tolerance of mechanical effort and the thermal stability.

3.4 Membrane Performance Analysis

The membrane performance was assessed in terms of both total flux and separation factor. In this work, these parameters were analyzed as a function of the operating conditions such as temperature and ethanol feed concentration. However, the effect of hydrophilic/hydrophobic nature of the membrane was firstly discussed for an appropriate approach of the membrane performance according to the water-affinity mechanism.

3.4.1 Effect of the membrane water affinity on separation performance of ethanol-water mixtures

The choice of a hydrophobic or hydrophilic membrane depends on the composition of the mixture to be treated and on the design separation process. If the feed is rich in ethanol, a hydrophilic membrane is adequate to use for selective sorption of water since there is less amount of mass to permeate across membrane. On the contrary, the use of a hydrophobic membrane is recommended to separate a solution with a high concentration of water. This selection criteria is supported by many experimental designs presented in the literature (Chai et al., 2015a; De Guzman et al., 2019; Mao et al., 2019; Shi et al., 2015). There is no evidence to establish that either of the two affinity mechanisms is more efficient than the other. In consequence, the hydrophobic and hydrophilic membranes were analyzed separately.

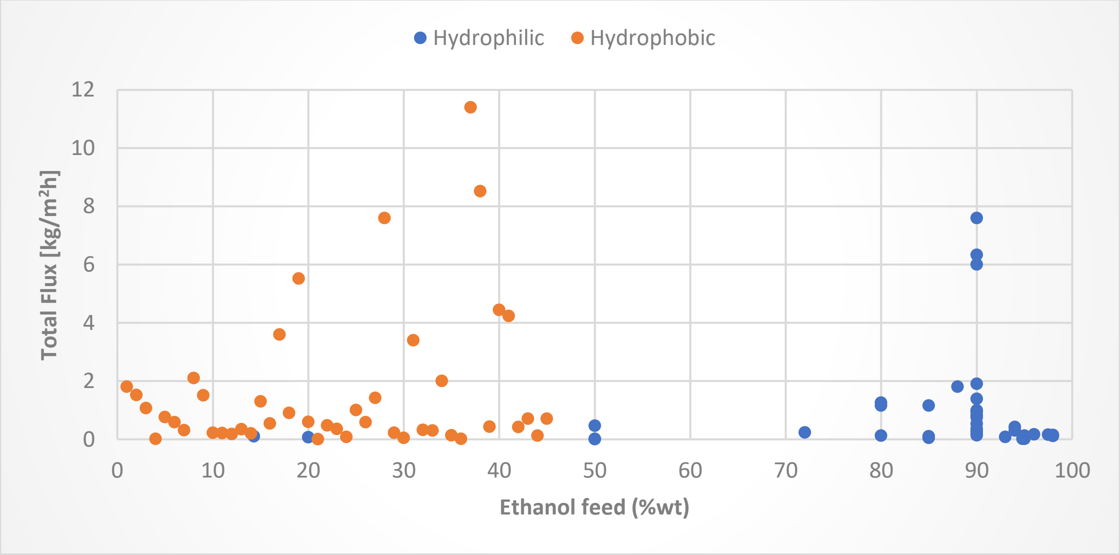
Firstly, the membranes were listed according to their permeate capacity and selectivity. Appendix A summarizes the hydrophilic and the hydrophobic membranes with higher total flux, while Appendix B resumes the hydrophilic and the hydrophobic membranes with higher separation factor.

3.4.2 Effect of ethanol feed concentration on separation performance of ethanol-water mixtures

The influence of the ethanol feed concentration on the total flux was illustrated in Figure 5 from data reported in the articles reviewed. It is evident that for hydrophilic membranes a higher ethanol concentration in the feed will result in an increase of total flux (Selim et al., 2019). It has been reported that the higher ethanol concentration in the feed provided a higher driving force via the increase in the ethanol effective partial pressure (Jyoti et al., 2015). While for hydrophobic membranes, increasing the ethanol feed concentration leads to reduce the amount of water for selective permeation across the membrane, and therefore, to increase of total flux (Fini et al., 2018b; Karp et al., 2018b). This is due to the rise of feed concentration that promotes the interaction possibilities of the solute with the membrane surface, increasing the partial pressure of the ethanol and its driving force for the permeation, and consequently the total flux increases (Zhou et al., 2014).

Figure 5

Total flux vs ethanol feed concentration



Otherwise, the effect of the ethanol feed concentration on the separation factor is disclosed in Figure 6 and 7 for hydrophilic and hydrophobic membranes respectively. In the case of hydrophilic membranes, the separation factor improves as the ethanol concentration in the feed increases (J. Liu & Bernstein, 2017; Q. Liu et al., 2017a), whereas in the case of hydrophobic membranes, the relation between these parameters is inversely proportional (Han et al., 2016b; Kamelian et al., 2019a). In the literature has been explained that the increase of the ethanol saturation increases the flux, thus, the mass absorbed is greater and the free volume is larger, which facilitates the pass of the small-sized molecules like water (Jafarinasab et al., 2015b). For the hydrophilic membranes, it favors the separation factor in contrast to hydrophobic membranes.

Figure 6

Separation factor vs ethanol feed concentration for hydrophilic membranes

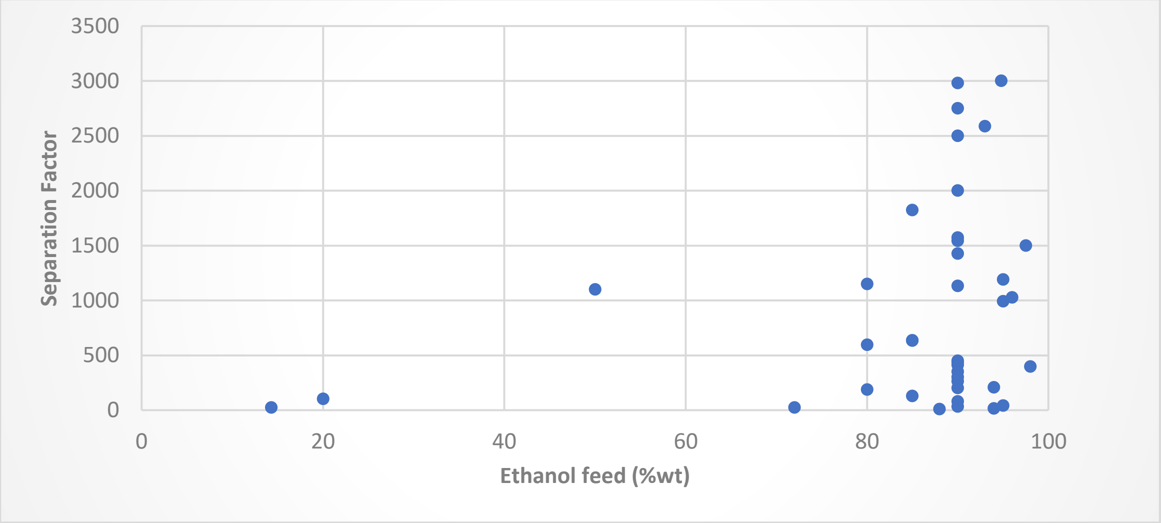
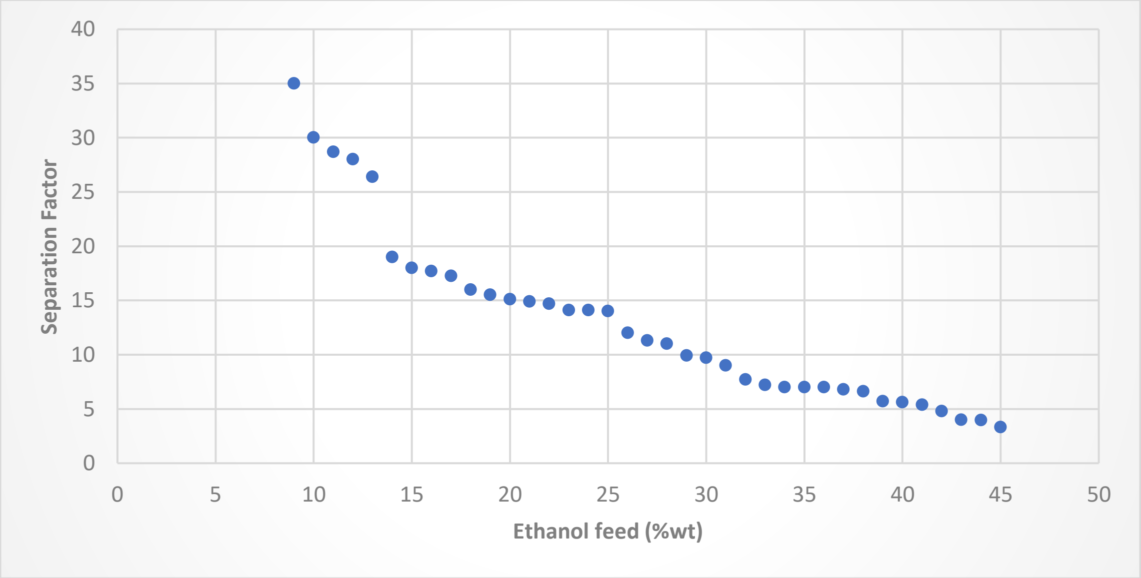


Figure 7

Separation factor vs ethanol feed concentration for hydrophobic membranes



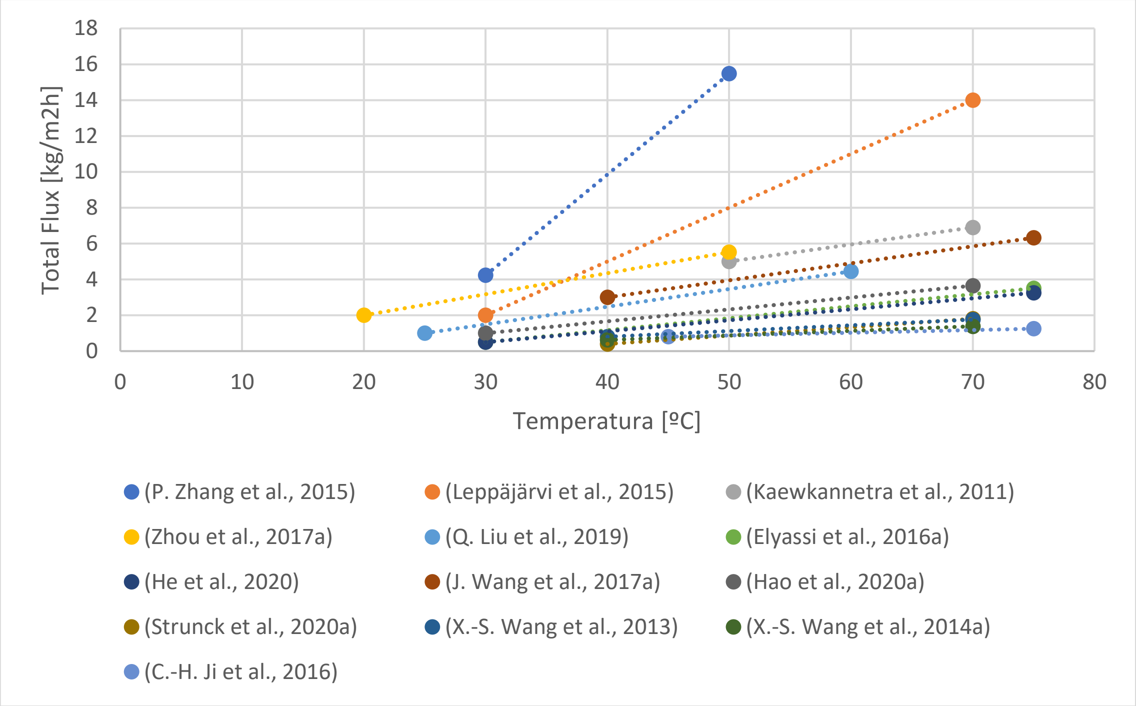
3.4.2 Effect of temperature on separation performance of ethanol-water mixtures

The operating temperature of all the pervaporation experiments reported in the selected papers oscillates between 20ºC and 80ºC. The typical temperature reported for evaluating the membrane was 50ºC that is lower than the boiling point of the ethanol, 78.24ºC at atmospheric pressure (W.M. Haynes; David R. Lide; Thomas J. Bruno, 2017). The heat necessary for the evaporation permeation in the pervaporation process is less than that of required for distillation, thereby, the energy cost is lower.

Figure 8 shows the higher flux for hydrophilic and hydrophobic membranes, as a function of the operating temperature. The increase of the operating temperature increases the total flux (Hao et al., 2020a; Jia et al., 2017; Kaewkannetra et al., 2011b; Kamelian et al., 2019a; Strunck et al., 2020b; J. Wang et al., 2017a; P. Zhang et al., 2015; Zhou et al., 2017a). It has been reported that the heating of the feed favors the change from liquid to vapor of the solute across the membrane, this phase change pushes the feed and the permeated molecules to pass through a large part of the membrane (Fini et al., 2018b), regardless of the water affinity mechanism. Consequently, a high feed temperature in the pervaporation benefits the permeate capacity of the membrane (Elyassi et al., 2016a; Hu et al., 2016a; Leppajarvi et al., 2015; J. Liu & Bernstein, 2017; Q. Liu et al., 2019).

Figure 8

Operation temperature effect in the total flux



On the other hand, the relationship between the temperature and the separation factor is illustrated in Figures 9 and 10, for hydrophilic and hydrophobic membranes respectively. The selectivity decreases as the operating temperature increases regardless of the water affinity mechanism (Cheng et al., 2019a; Q. Liu et al., 2017a; Meireles et al., 2013a).This is owing to the expansion of the free volume provoked by the phase change of the permeate. Consequently, the molecular diffusion is easier for both components hindering the selective sorption (L. Ji et al., 2015b). Thus, to improve the quality of the separation process it is adequate to use lower values of operating temperature range (Chai et al., 2015b; Sun et al., 2013a; Zhu et al., 2017a). Overall, the temperature is directly proportional to the total flux, but has the opposite effect in the separation factor.

Figure 9

Operating temperature effect in the separation factor in hydrophilic membranes

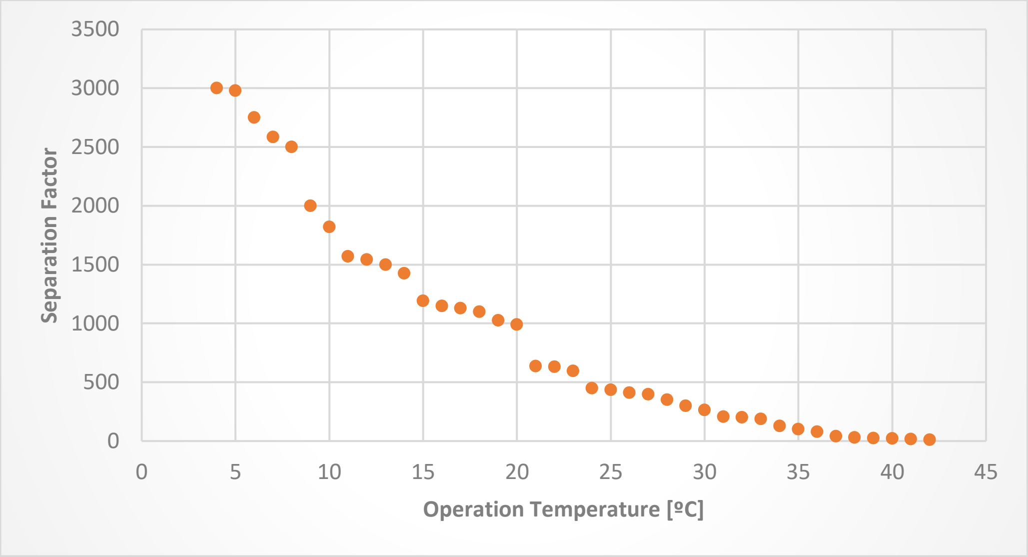
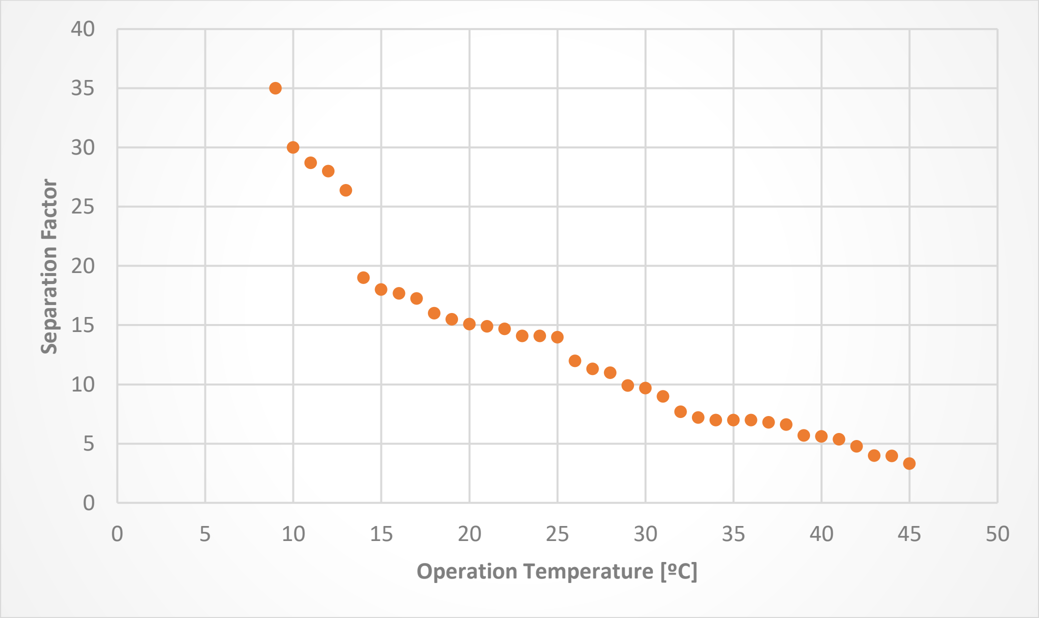


Figure 10

Operating temperature effect in the separation factor in hydrophobic membranes



4. Conclusions

From this systematic review it was possible to determine that the improvements in membrane technology for ethanol-water separation via pervaporation is a relevant research topic. In this regard, China has a remarkable contribution to the field, accounted for 42% of the total amount of papers published in the period under review.

In terms of membrane materials, hybrid/compound alternatives represent 53% of the research, organics 37%, and inorganics 10%, of the total membranes reviewed. These results reflect that the preference for the polymeric sources as the exclusive membrane material or in a hybrid or compound matrix. However, the polymeric sources present some drawbacks, such as the swelling and the chemical stability. Hence, the addition of inorganic materials and hybrid/compounds helps to overcome these difficulties. Nonetheless, it is still necessary to follow improving the adhesion between layers for compound membranes, and enhancing the interaction of inorganics and polymers for increasing the performance of the hybrid membranes

The ethanol feed concentration improves the total flux in hydrophilic and hydrophobic membranes, while the separation factor enhances with the increase of ethanol mass fraction for hydrophilic membranes while having the opposite effect on hydrophobic ones. The operating temperature is directly proportional to the total flux, not depending on the water-affinity mechanism, and in turn, is inversely proportional to the separation factor for both hydrophobic and hydrophilic membranes.

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Appendices

Appendix A: Hydrophilic and hydrophobic membranes reported in the literature with higher total flux

Hydrophilic membranes reported in the literature with higher total flux

Type	Operation Temperature [ºC]	Ethanol (%wt)	Flux [$\frac{kg}{m^2h}$]	Reference
Hybrid	50	90	7,6	(J. Liu & Bernstein, 2017)
Hybrid	75	90	6,332	(J. Wang et al., 2017b)
Inorganic	75	90	6	(Hu et al., 2016a)
Organic	70	90	3,65	(Hao et al., 2020b)
Hybrid	60	90	1,903	(J. Li et al., 2018)
Organic	55	88	1,8	(Strunck et al., 2020a)
Organic	70	90	1,76	(X.-S. Wang et al., 2013)
Hybrid	70	90	1,385	(X.-S. Wang et al., 2014a)
Hybrid	75	80	1,25	(C.-H. Ji et al., 2016)
Organic	50	85	1,16	(Shi et al., 2015)

Hydrophobic membranes reported in the literature with higher total flux

Type	Operation Temperature [ºC]	Ethanol (%wt)	Flux [$\frac{kg}{m^2h}$]	Reference
Organic	30	5	15,47	(P. Zhang et al., 2015)
Inorganic	50	7	14	(Leppäjärvi et al., 2015)
Organic	65	9	8,523	(Jia et al., 2017)
Organic	60	15	6,9	(Kaewkannetra et al., 2011a)
Hybrid	50	5	5,53	(Zhou et al., 2017b)

Hybrid	60	5	4,446	(Q. Liu et al., 2019)
Hybrid	50	10	3,6	(Kamelian et al., 2019a)
Inorganic	60	5	3,5	(Elyassi et al., 2016b)
Hybrid	37	6	3,4	(He et al., 2020)
Organic	40	5,1	2,428	(Chen et al., 2014)

Appendix B: Higher separation factor hydrophilic and hydrophobic membranes reported for ethanol-water separation

Higher separation factor hydrophilic membranes reported for ethanol-water separation

Type	Operation Temperature [ºC]	Ethanol (%wt)	Separation Factor	Reference
Organic	70	98	4281	(Cheng et al., 2019b)
Organic	30	94,8	3000	(Meireles et al., 2013b)
Hybrid	30	90	2980	(Q. Liu et al., 2017b)
Organic	30	90	2750	(Thakur et al., 2013)
Hybrid	37	93	2585	(Cai et al., 2020)
Hybrid	80	90	2500	(Wu et al., 2015b)
Inorganic	75	90	2000	(Hu et al., 2016b)
Hybrid	40	85	1821	(H. Zhang & Wang, 2016)
Hybrid	70	90	1571	(X.-S. Wang et al., 2014b)
Hybrid	60	90	1542	(J. Li et al., 2018)

Higher separation factor hydrophobic membranes reported for ethanol-water separation

Type	Operation Temperature [ºC]	Ethanol (%wt)	Separation Factor	Reference
Inorganic	52,5	5	35	(Chai et al., 2015c)
Organic	60	5	30	(Sun et al., 2013b)
Hybrid	50	5	28,7	(Zhu et al., 2017b)
Organic	30	80	28	(Villagra Di Carlo & Habert, 2013)
Hybrid	30	5	26,4	(Samanta & Ray, 2015)
Hybrid	40	5	19	(Peng et al., 2011b)

Hybrid	55	5,5	18	(Mao et al., 2019)
Organic	50	18	17,7	(Zhan et al., 2020)
Hybrid	50	10	17,25	(Kamelian et al., 2019b)
Hybrid	60	6	16	(Khan et al., 2018b)