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Pervaporative Desulfurization: A Comprehensive Review of Principles, Advances, and Applications

Pervaporative desulfurization is a potential method for removing sulfur compounds from liquid hydrocarbon streams, having many advantages over existing methods. This overview covers the fundamentals, recent breakthroughs, and different applications of pervaporative desulfurization. The paper begins by explaining pervaporative desulfurization's core concepts, focusing on how polymeric membranes selectively separate sulfur molecules from liquid hydrocarbons. It explores membrane properties, operating conditions, and feed composition in pervaporative separation. It highlights how pervaporative desulfurization may reduce sulfur content to ultra-low levels and meet strict environmental and fuel quality standards. Finally, this comprehensive review paper concludes with a discussion on the future prospects and research directions in the field of pervaporative desulfurization. It highlights the need for continued innovation in membrane materials, module design, and process optimization, as well as the importance of addressing challenges related to scale-up and industrial implementation. Overall, this review paper provides valuable insights into the advancements, challenges, and potential applications of pervaporative desulfurization, offering a comprehensive understanding of this technology for researchers, engineers, and all parties involved in the development and implementation of sustainable sulfur removal processes.

Keywords: pervaporative desulfurization, membrane desulfurization, trace sulfur removal, thiophene removal, dibenzothiophene, acid rain, sulfur-containing oils, hydrodesulfurization, oxidative desulfurization.

Contents

Review plan

The present review is devoted to pervaporative desulfurization and principles, membrane fabrication techniques, process optimization of pervaporative desulfurization, storing sulfur-containing oils after desulfurization, limitations of pervaporative desulfurization and conclusion. Each section covered necessary information and data to be easily understood by the readers. Online available references from trustworthy sources were used. This review has focused on all aspects of PVD, including its optimization, the storage of sulfur-containing oils, and the limitations of PVD, which were not fully discussed in other review papers.

Introduction

Sulfur compounds in crude oil can cause serious environmental and health problems, such as acid rain [1–5], respiratory diseases [6, 7] and cancer [8]. Therefore, the removal of sulfur from crude oil has become a critical issue in the petroleum industry. Conventional desulfurization technologies, such as hydrodesulfurization (HDS) and oxidative desulfurization (ODS), have been widely used to remove sulfur compounds from crude oil [9–16]. However, these methods have some limitations, such as high energy consumption, high capital and operating costs, and low selectivity for certain sulfur compounds. Therefore, alternative desulfurization technologies, such as pervaporative desulfurization (PVD), have attracted increasing attention in recent years.

Compared to other desulfurization methods, such as extractive desulfurization, oxidative desulfurization, bio-desulfurization, and adsorptive desulfurization, pervaporative desulfurization is considered a promising method for desulfurization of fuels due to its low cost, low energy consumption, high yield, and easy processing [17, 18]. It offers advantages such as higher selectivity, lower investment and operating costs, and scalability [19]. Therefore, pervaporative desulfurization has various applications in different industries. One of the main applications is in gasoline desulfurization. The stringent regulations on sulfur content in gasoline have led to a growing interest in the use of pervaporation for this purpose [20]. Pervaporation is a promising separation technology for removing sulfur compounds from gasoline while maintaining its octane number [21]. Compared to traditional hydrodesulfurization methods, pervaporation can be carried out at normal pressure and temperature without the need for hydrogen and catalysts, resulting in lower operating costs [21]. Pervaporation is also used for the separation of azeotropic mixtures, solutions with similar boiling points, and thermally sensitive compounds. It is an effective method for removing dilute organics from aqueous solutions [20].

Pervaporative desulfurization has several applications in environmental protection. One of the main applications is in the removal of sulfur dioxide (SO_2) from flue gas. Wet flue gas desulfurization using phosphate rock slurry has been employed successfully in industrial applications for the elimination of SO_2 from phosphorus chemical processes. This technology offers an efficient and cost-effective method for SO_2 abatement. The use of phosphate rock as the raw material reduces the desulfurization cost [22]. Pervaporation is also used for the desulfurization of transport fuels, such as gasoline.

This article aims to provide a comprehensive review of the principles, mechanisms, and applications of pervaporation in desulfurization (PVD). While many other review articles focus on pervaporation characteristics and membranes, this review article also presents membrane fabrication methods and the optimization process of pervaporation desulfurization. The study on the optimization process of PVD enables readers to gain a better understanding of the procedure and to overcome obstacles for large-scale applications in industries.

Principles of Pervaporation

Pervaporation (PV) is a membrane-based separation process that utilizes the differences in the solubility, diffusivity, and vapor pressure of different components in a liquid mixture to selectively permeate through a membrane. PV involves the following steps (Fig 1.):

The liquid mixture is fed to the upstream side of a membrane module.

The membrane selectively permeates the more volatile component (usually water or organic solvents) from the liquid mixture through the membrane pores by evaporation or diffusion. The permeated component is condensed or adsorbed on the downstream side of the membrane. The non-permeated component is collected as the retentate [23, 24].

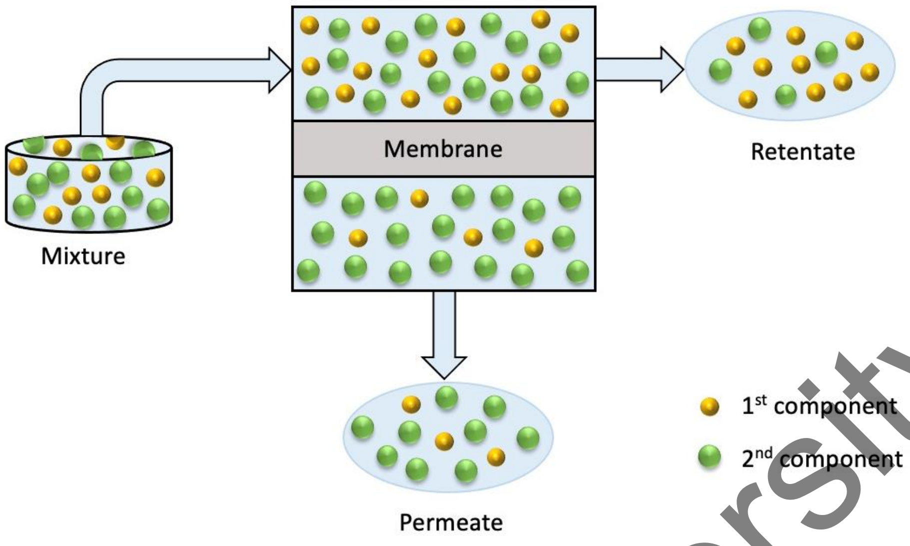

Figure 1. Schematic illustration of pervaporation process

PV is a promising separation technology for various applications, such as dehydration, concentration, and purification of organic and aqueous solutions. PV has several advantages over conventional separation methods, such as low energy consumption, high selectivity, and high recovery. PV membranes can be classified into two main categories: dense membranes and porous membranes. Dense membranes are made of polymer or ceramic materials that have low permeability but high selectivity for certain components. Porous membranes are made of materials that have high permeability but low selectivity for all components. The performance of PV membranes depends on various factors, such as membrane material, membrane structure, membrane morphology, membrane surface properties, feed composition, operating temperature, and pressure [24-28].

Mechanisms of PVD

Pervaporative desulfurization (PVD) is a combination of PV and selective permeation to remove sulfur compounds from crude oil. PVD involves the following steps:

The crude oil is mixed with a solvent that can dissolve the sulfur compounds but not the hydrocarbons.

The mixture is fed to the upstream side of a pervaporation membrane module.

The membrane selectively permeates the solvent and the dissolved sulfur compounds from the mixture through the membrane pores by evaporation or diffusion.

The permeated solvent and sulfur compounds are condensed or adsorbed on the downstream side of the membrane.

The retentate, which contains the hydrocarbons, is collected.

The selection of solvents is crucial for PVD because the solvent should have high solubility and selectivity for sulfur compounds but low solubility and selectivity for hydrocarbons. The choice of membrane materials is also important for PVD because the membrane should have high selectivity for sulfur compounds but low selectivity for hydrocarbons and solvent. The performance of PVD depends on various factors, such as solvent composition, membrane material, membrane structure, membrane morphology, membrane surface properties, feed composition, operating temperature, and pressure.

Adsorptive Pervaporative Desulfurization

Adsorptive Pervaporative Desulfurization (APD) is a combination of adsorption and pervaporation techniques used for oil desulfurization. The mechanisms involved in APD include:

Adsorption: The oil is passed through an adsorbent material, such as hydrophobic membrane [29], which selectively adsorbs sulfur-containing compounds. The adsorbent or membrane act as a molecular sieve, capturing the sulfur compounds from the oil stream. Metal organic fillers inside the membrane play important role in adsorbing organic sulfur. The interaction between metal ions and polymers and the interaction between sulfur and metals share some similarities, particularly in terms of coordination chemistry and bonding [30–33]. In both cases, the interaction involves the coordination of atoms or ions with specific binding sites on the polymer or metal surface. Metal ions can coordinate with functional groups on the polymer, forming coordination complexes, sulfur of while can interact with metal surfaces through coordination bonding. The following are possible mechanisms for the adsorption of aromatic sulfur compounds onto adsorbents [34–39]:

1. π-Complexation:

– This mechanism includes the transfer of electrons from the π-orbitals of the sulfur compounds, like thiophene, in the fuel to the metal atom's vacant s-orbitals (orbitals with available electron density).

– Alternatively, electrons can be given from the metal atom's d-orbitals to the antibonding π-orbitals of aromatic sulfur compounds.

– π-Bond creation or electron donation from the metal atom's d-orbitals results in chemical bonding between the sulfur and the metal.

2. Acid-Base Interaction:

– In this process, metal sites on the adsorbent, such as iron, chromium, aluminum, copper, zinc, cobalt, and so on, operate as Lewis acidic sites.

– Lewis acidic sites in hydrocarbon structures interact with sulfur atoms via Lewis acid-base interactions.

– Lewis acid-base interactions are chemical interactions in which electron-rich Lewis bases (in this example, sulfur atoms) interact with electron-deficient Lewis acids (metal sites), resulting in chemical bond formation.

3. Direct Sulfur-Metal Interaction:

– The direct creation of a σ-bond between the sulfur in the fuel and the metal atoms in the adsorbent is involved in this mechanism.

– The bond is created by the donation of lone pair electrons from the hydrocarbon structure's sulfur atom to a metal atom.

– As a result, the sulfur forms a strong chemical bond with the metal surface.

These mechanisms demonstrate the various ways in which aromatic sulfur compounds can be adsorbed. One or more of them may be engaged in the adsorption process, depending on the individual metal and the chemical characteristics of the sulfur compounds. These mechanisms are critical in understanding how and why specific adsorption processes occur.

However, despite its increased sulfur content, application to heavy oil is impractical due to limited accessibility of large molecules such as dibenzothiophene and its derivatives in small pores and steric hindrance, which lowers adsorption efficacy.

Pervaporation: Pervaporation is a membrane-based process where a semi-permeable membrane selectively allows certain molecules (in this case, hydrocarbons) to pass through while blocking others (sulfur compounds and water).

Selective Permeation: The pervaporation membrane has a high affinity for hydrocarbons, allowing them to permeate through the membrane and exit as the purified oil product. At the same time, the sulfur compounds, which have a lower affinity for the membrane, are retained and concentrated on the feed side of the membrane. During the selective permeation process, the pervaporation membrane aids in the removal of sulfur-containing compounds from oil. This membrane exhibits a high affinity for hydrocarbons, allowing them to permeate through the membrane and exit as purified oil. The mechanisms of desulfurization involve interactions between the sulfur compounds and the active sites of the membrane material, such as hydrogen bonding and acid-base complexation. These interactions enable the preferential removal of sulfur compounds, such as thiophene, from the oil phase [37, 38].

Regeneration: Periodically, the adsorbent material becomes saturated with sulfur compounds. To maintain the desulfurization efficiency, the adsorbent is regenerated by heating or using other methods to remove the sulfur compounds and restore its adsorption capacity.

Through these mechanisms, APD can achieve efficient desulfurization, producing low-sulfur content oil with reduced environmental impact and meeting stringent fuel quality standards. APD is an attractive option for specific applications where other desulfurization methods may not be feasible or cost-effective.

Selecting Membrane Materials for pervaporative desulfurization

Pervaporative desulfurization (PVD) is a membrane-based separation process (Fig 2.) that has shown promise in the removal of sulfur compounds from crude oil and other industrial waste streams. Membrane materials play a crucial role in the performance of PVD, and the selection of appropriate materials depends on various factors, such as the chemical compatibility with the feed stream, the selectivity towards sulfur compounds, the mechanical strength, and the cost-effectiveness [39–41].

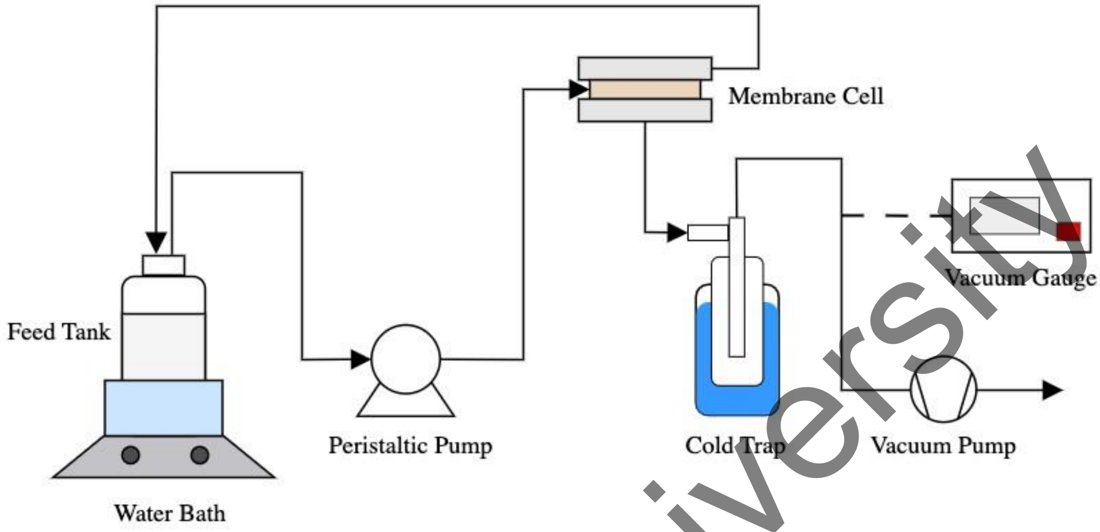

Figure 2. The schematic illustration of pervaporation process

Polymeric membranes, such as polydimethylsiloxane (PDMS) [42–44], polyvinylidene fluoride (PVDF) [45, 46], and cellulose acetate (CA) [47, 48], are the most commonly used membrane materials for PVD. PDMS is a highly permeable and flexible material that has shown excellent selectivity towards sulfur compounds due to its high affinity towards polar molecules. PVDF is a hydrophobic and mechanically robust material that has shown good selectivity towards sulfur compounds. CA is a hydrophilic and biocompatible material that has shown high selectivity towards small sulfur compounds.

Composite membranes, which are composed of two or more polymers or inorganic materials, have also been developed for PVD. Composite membranes have the advantage of combining the properties of different materials to achieve higher selectivity and permeability towards sulfur compounds. For example, Cao et al. (2011) designed PDMS/silica composite membranes that have shown higher selectivity towards sulfur compounds than pure PDMS membranes due to the presence of silica nanoparticles, which enhance the surface affinity towards sulfur compounds [41].

Inorganic membranes, such as ceramic and metal membranes, have also been investigated for PVD. Inorganic membranes have the advantage of high thermal and chemical stability, which makes them suitable for harsh environments. Ceramic membranes, such as alumina and zeolite membranes, have shown promising results in the removal of sulfur compounds from model oil and real crude oil [49, 50]. Metal membranes, such as palladium and silver membranes, have shown high selectivity towards hydrogen sulfide due to their ability to catalytically oxidize sulfur compounds to form sulfates [51, 52].

The choice of membrane material depends on the specific application and the available resources. Polymeric membranes are generally more cost-effective and easier to fabricate than inorganic membranes, but they may suffer from low mechanical strength and chemical compatibility issues. Inorganic membranes are more expensive and difficult to fabricate, but they may offer higher selectivity and stability in harsh environments.

Process Optimization of Pervaporative Desulfurization

In terms of materials, various types of membranes have been investigated for pervaporative desulfurization. For example, a study explored the use of a polyethylene glycol (PEG) membrane and found that it exhibited excellent desulfurization properties [53]. Another study incorporated a unique two-dimensional structure material, graphene egg yolk-shell nanostructure (GYSNs), into a polyurethane matrix to fabricate mixed matrix membranes (MMMs) for pervaporative desulfurization [24]. Additionally, the incorporation of SiO_2 nanoparticles in polyvinyl butyral (PVB) membranes improved the performance of pervaporation for gasoline desulfurization [22]. Additionally, the incorporation of metal-organic frameworks (MOFs) such as CuBTC (copper benzene-1,3,5-tricarboxylate) into polyethylene glycol (PEG) membranes has been shown to improve desulfurization performance [54]. The use of novel thin film nanocomposite membranes based on chitosan succinate modified with Fe-BTC (iron benzene-1,3,5-tricarboxylate) has also been explored for enhanced pervaporation desulfurization. Furthermore, the incorporation of inorganic particles, such as silver oxide (Ag_2O), into polydimethylsiloxane (PDMS) membranes has been investigated to enhance the desulfurization properties. The interaction between the filler and thiophenes in the mixed matrix membrane improves the sulfur removal efficiency of the pervaporative desulfurization process [43].

Farsi et al. optimized PDMS, PEG, PES, and PAN composite membranes by changing cross-linking concentration, time, temperature, and concentration of TEOS. The composite membrane of PEG + PDMS had the highest overall flux (0.7732 L/h) and lowest sulfur backflow (1780 ppm). The utilization of tetraethyl orthosilicate (TEOS) resulted in a decline in the overall flux, accompanied by a reduction in the quantity of sulfur present in the recycled flow. The increase in the concentration of tetraethyl orthosilicate (TEOS) from 8 to 22 wt% in composite membranes consisting of polydimethylsiloxane (PDMS) combined with polyacrylonitrile (PAN), polyethersulfone (PES), polyethylene glycol (PEG), and PDMS alone resulted in a decrease in flux from 0.5412 to 0.5217 L/h, 0.6215 to 0.6033 L/h, 0.7583 to 0.6211 L/h, and 0.6813 to 0.6314 L/h, respectively. In a similar way, the sulfur content in the backflow decreased from 2035 to 1982 parts per million (ppm), 1933 to 1921 ppm, 1825 to 1811 ppm, and 2011 to 1972 ppm, correspondingly [55].

Membrane Fabrication techniques for Pervaporative Desulfurization

Membrane fabrication is a critical step in the development of pervaporative desulfurization (PVD) systems, as the performance of the membranes greatly influences the efficiency and selectivity of the process. Several membrane fabrication techniques have been developed for PVD, including solution casting, electrospinning, phase inversion, and layer-by-layer assembly. Each technique has its advantages and disadvantages, and the choice of the appropriate technique depends on various factors, such as the desired membrane properties, the target application, and the available resources.

Solution casting is a common membrane fabrication technique that involves the casting of a polymer solution onto a substrate and the subsequent evaporation of the solvent to form a thin film (Fig. 3). This technique is simple, cost-effective, and versatile, and can produce membranes with different thicknesses and compositions. However, solution casting may suffer from poor reproducibility and limited scalability, and the resulting membranes may have low mechanical strength and poor permeability.

Phase inversion is a membrane fabrication technique that involves the precipitation of a polymer solution by immersion in a non-solvent bath. This technique can produce membranes with a range of morphologies, such as asymmetric and porous structures, and can be used to tailor the membrane properties by controlling the composition and conditions of the precipitation bath. Phase inversion has been widely used in PVD, as it can produce membranes with high selectivity and permeability towards sulfur compounds. However, phase inversion may suffer from limited reproducibility and the use of toxic solvents.

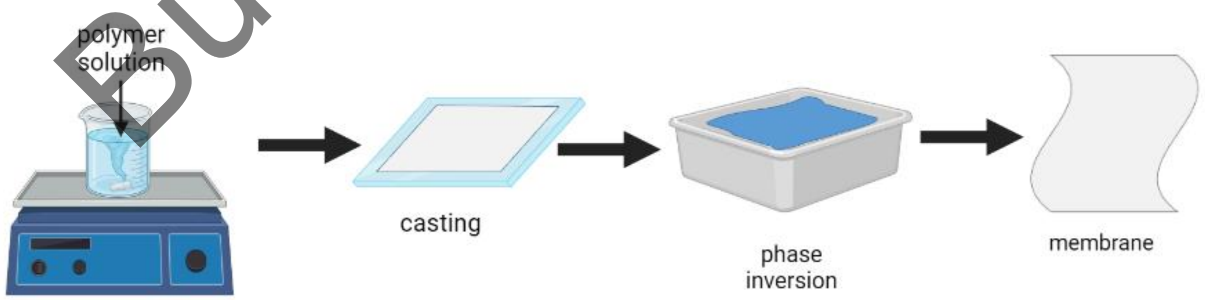

Figure 3. The schematic illustration of membrane fabrication by solution casting method

Layer-by-layer assembly is a membrane fabrication technique that involves the deposition of alternating layers of polyelectrolytes onto a substrate (Fig. 4). This technique can produce membranes with precise control over the thickness and composition of the layers, and can be used to tailor the membrane properties by selecting different polyelectrolytes. Used layer-by-layer assembly which has shown promising results in

PVD, as it can produce membranes with high selectivity and stability towards sulfur compounds. However, layer-by-layer assembly is a complex and time-consuming process that may require specialized equipment and expertise [53].

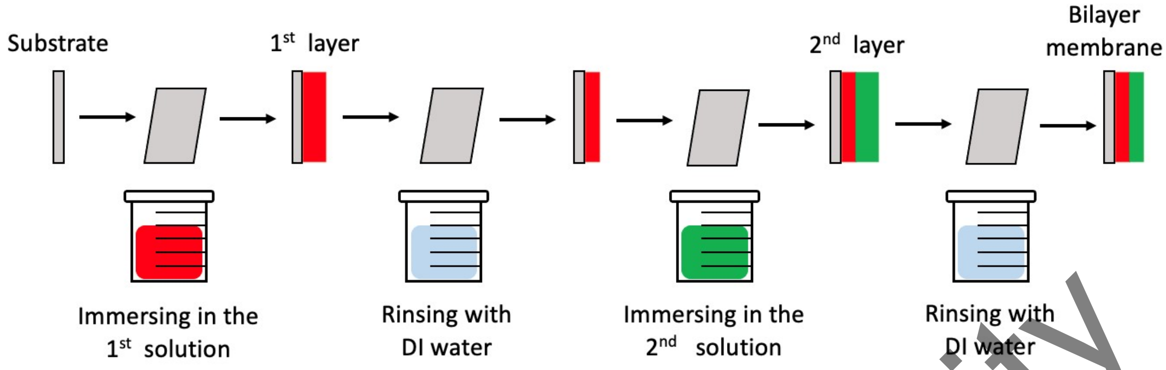

Figure 4. The schematic illustration of membrane casting by layer-by-layer technique

Storing sulfur-containing oils after desulfurization

Storing sulfur-containing oils requires careful consideration and proper handling to ensure safety, maintain oil quality, and comply with environmental regulations.

Storing sulfur-containing oils in superhydrophobic materials offers several benefits due to their water-repellent nature and unique surface properties. Superhydrophobic materials can effectively prevent water and moisture ingress into the stored oils, which is essential for preserving oil quality and preventing degradation [54]. Additionally, the use of superhydrophobic materials can minimize corrosion and oxidation of metal containers used for oil storage, as water contact with the container surfaces is reduced [55]. This leads to extended storage life and improved overall stability of the stored sulfur-containing oils.

Superhydrophobic coatings or linings applied to storage tanks, pipelines, and containers can also facilitate easier and cleaner oil transfer, preventing residual water from mixing with the oil during handling and transportation [56]. However, it is essential to consider the compatibility of the superhydrophobic materials with the specific sulfur-containing oils to ensure there are no adverse chemical interactions that may affect the oil's quality or the material's integrity [57]. Proper testing and selection of suitable superhydrophobic materials are crucial for successful and efficient storage of sulfur-containing oils [58].

Limitations of pervaporative desulfurization:

1. Membrane Stability: The stability of pervaporation membranes is crucial for long-term operation. Polymeric membranes, in particular, may suffer from limitations such as low stability under harsh operating conditions, including high temperatures and chemical exposure [23].

2. Fouling and Membrane Degradation: Fouling, deposition of contaminants on the membrane surface, and membrane degradation can occur during pervaporative desulfurization. Sulfur compounds or other impurities in the feed stream can lead to fouling, reducing the membrane performance over time [54].

3. Aggregation and Interface Defects: The incorporation of fillers or nanoparticles into membranes, such as metal-organic frameworks (MOFs), can enhance the separation performance. However, the aggregation of particles and interface defects can limit the efficiency and transport properties of the membranes [53].

4. Process Optimization: Achieving optimal operating conditions, including temperature, pressure, and feed composition, is crucial for maximizing the efficiency of pervaporative desulfurization. Process optimization requires a comprehensive understanding of the separation mechanism and transport phenomena [23].

5. Cost and Scalability: The cost of membrane materials, fabrication procedures, and overall process design can be a limitation for pervaporative desulfurization. Additionally, scaling up the process to industrial levels while maintaining efficiency and cost-effectiveness can be challenging [23].

6. Removal of Trace Sulfur: Pervaporative desulfurization may face limitations in achieving complete removal of trace sulfur compounds. Achieving ultra-low sulfur levels required for certain applications, such as in the production of high-purity chemicals or pharmaceuticals, may require additional purification steps [59, 60].

While pervaporative desulfurization offers advantages in sulfur removal, it also faces limitations and challenges. These include selectivity, membrane stability, fouling, specific sulfur compounds, aggregation and interface defects, process optimization, cost, scalability, and the removal of trace sulfur. Overcoming these limitations will require further research and development efforts to enhance the efficiency and applicability of pervaporative desulfurization in various industries.

Conclusions

In conclusion, this comprehensive review paper has provided a deep understanding of the principles, recent advances, and diverse applications of pervaporative desulfurization. By exploring the fundamental mechanisms, recent advancements in membrane materials and module design, and the integration of pervaporative desulfurization with other processes, the paper highlights the potential of this technology for efficient and selective sulfur removal from liquid hydrocarbon streams.

The review emphasizes the significance of novel polymeric membranes, nanocomposite membranes, and hybrid membranes in enhancing separation performance, selectivity, and durability. Additionally, surface modification techniques and tailoring membrane morphology have been identified as key factors in improving the overall efficiency of pervaporative desulfurization systems.

Finally, the review identifies future prospects and research directions, including the development of novel membrane materials, optimization of module design, process integration, scaling up for industrial implementation, and a focus on environmental and energy sustainability. These research directions will pave the way for the widespread adoption and commercialization of pervaporative desulfurization, addressing the challenges associated with sulfur removal and contributing to a cleaner and more sustainable energy landscape.

Overall, this review paper serves as a valuable resource for researchers, engineers, and stakeholders interested in the advancement and application of pervaporative desulfurization technology, facilitating further innovation and optimization in this field.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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