

## Article

# Effects of Operating Parameters on Direct Contact Membrane Distillation Concentration of Licorice Root, Honeysuckle, and Indigo Woad Root Aqueous Extracts

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**Abstract:** With the rapid development of the traditional Chinese medicine industry, the concentration of the aqueous extract of traditional Chinese medicine has become an important research direction. Among them, the high concentration temperature of traditional Chinese medicine is prone to cause the loss of heat-sensitive components in Chinese medicine. Compared to conventional concentration methods, membrane distillation gives high purity concentrates under mild operating conditions. Therefore, the aim of this study was to evaluate the effects of feed temperature, hot-side flux, cold-side flux, cold-side temperature and different membrane pore sizes on the concentration of aqueous extracts of traditional Chinese medicine. In addition, the contamination of membranes in the MD process and the effect of chemical cleaning on the removal of the contamination were investigated. The results showed that it was feasible to concentrate the aqueous extract of traditional Chinese medicine by DCMD method. Chemical cleaning, especially alkali as a cleaning agent, is effective in removing the dirt layer.

**Keywords:** direct contact membrane distillation; traditional Chinese medicine concentration; membrane contamination; membrane cleaning

## 1. Introduction

Chinese medicine is a valuable resource of the Chinese nation and has played an important role in treating diseases and maintaining the health of the Chinese people for more than 2000 years. Chinese medicine has developed through thousands of years of clinical experience and evolution [1,2]. At present, Chinese medicine has spread to more than 140 countries and regions, providing new methods and theories for treating diseases [3].

Unlike Western medicines, which rely on man-made compounds, herbal medicines are extracted from plants, animals and minerals and contain a number of active and inactive ingredients that require a series of preparatory procedures before they can become usable medicines [4]. The process of Chinese medicine preparation includes extraction, separation, purification, concentration and drying (Figure S1). The continuous progress of pharmaceutical technology has shown that some traditional methods of preparing traditional Chinese medicines can no longer meet the requirements of the modern pharmaceutical industry [5]. For example, aqueous extracts of Chinese medicines usually contain low concentrations of active ingredients and many impurities. Therefore, additional isolation, purification and concentration steps are required to prepare the drugs [6,7]. Traditional separation methods include sedimentation and centrifugation. Purification methods include ethanol precipitation, salting out and adsorption with macroporous resins. Concentration methods include atmospheric pressure concentration and vacuum concentration [8–10]. Traditional methods are high in energy consumption, low



efficiency, high cost, high environmental impact and do not meet the standards of modern green manufacturing. Therefore, the application of new technology in Chinese medicine preparation is of great significance to improve the process of Chinese medicine and ensure the quality of products. At present, the concentration of TCM aqueous extracts and the separation of organic systems in food, chemical and other fields all face common bottlenecks of traditional technologies: thermal concentration methods such as atmospheric pressure and vacuum are prone to cause the loss of heat-sensitive components, while pressure-driven membrane technologies such as ultrafiltration and nanofiltration have the problems of low concentration multiple and severe membrane fouling. As a thermally driven membrane separation technology, membrane distillation (MD) has the advantages of mild operation, high separation efficiency and complete retention of non-volatile components, and has been initially applied in fruit juice concentration, organic wastewater treatment and other fields, becoming a potential technology to solve the separation problems of TCM and organic systems. However, existing studies mostly focus on a single system, and the research on adaptability and pollution control for complex TCM systems remains to be improved.

Membrane distillation (MD) is a thermally driven membrane process in which solvents (gaseous, vapour molecules) can be separated from a hydrophobic membrane driven by the difference in vapour pressure between the two sides of the membrane. Instead, non-volatile components of the feed will be rejected on the feed side of the hydrophobic membrane [11–14]. Therefore, membrane distillation technology has been widely used in food processing, seawater desalination, wastewater treatment, pharmaceutical industry and many other fields (Figure S2). It is highly efficient, relatively low investment, low energy consumption, friendly working environment and easy to use. Membrane distillation technology is considered to be an alternative to many of the traditional methods currently used in the traditional Chinese medicine industry [15–17]. In general, membrane distillation consists of four main configurations: Direct Contact Membrane Distillation (DCMD), Sweep Gas Membrane Distillation (SGMD), Air Gap Membrane Distillation (AGMD) and Vacuum Membrane Distillation (VMD) [18–20]. Among them, direct contact membrane distillation has attracted a lot of attention from researchers due to its simple process flow and large permeate volume. For example, Li et al. [21]. explored the feasibility of direct contact membrane distillation for the treatment of printing and dyeing wastewater, and the results showed that the removal rate of COD of the wastewater was always maintained at about 90%, and the removal rate of chromaticity was as high as 95%, which indicated that the direct contact membrane distillation can be better used for the treatment of printing and dyeing wastewater. Ding et al. [22] used DCMD to concentrate an extract of a traditional Chinese medicine and found that a high concentration rate and concentration quality could be obtained at a feed temperature of 60 °C and a feed flow rate of 0.102 m·s<sup>-1</sup>. As a typical thermally driven membrane separation technology, the trans-membrane flux of MD is dominated by the vapour pressure difference across the hydrophobic membrane, which is essentially different from the core mechanism of pressure-driven membrane processes that achieve separation by overcoming membrane resistance through trans-membrane pressure. Moreover, MD enables efficient concentration under mild temperatures, which can effectively avoid the loss of heat-sensitive active components in traditional Chinese medicines, a key advantage making it more suitable for the concentration of TCM aqueous extracts compared with pressure-driven membrane processes.

However, MD technology faces several limitations in practical application, including membrane fouling, membrane wetting and temperature polarization. Membrane fouling refers to the physical or chemical adsorption and deposition of organic substances in the feed liquid on the membrane surface or inside the pores, leading to the reduction of membrane pore size and the decrease of permeate flux [23]; membrane wetting is a secondary problem caused by fouling, which damages the gas-liquid interface of the hydrophobic membrane due to the decrease of membrane surface hydrophobicity, resulting in the irreversible loss of separation characteristics; scaling refers to the crystalline deposition of inorganic ions, and no obvious scaling phenomenon was observed in this study, with fouling dominated by organic adsorption [24,25]. Among them, membrane fouling is the key factor restricting the long-term stable operation of DCMD for the concentration of traditional Chinese medicine (TCM) extracts. Therefore, understanding the causes and mechanisms of membrane contamination is important for adopting appropriate strategies to reduce membrane contamination (Figure S3).

## 2. Experimentation

### 2.1. Water Samples

The aqueous extracts in this study were self-extracted. The decoction of 25 g of Bozhou Chinese honeysuckle, liquorice root and indigo woad root was carried out using 1000 mL of deionised water for 1 h respectively, filtered using gauze and then added with another 1000 mL of deionised water to continue the decoction for another hour and filtered. The two filtrates were combined to obtain 2000 mL of herbal aqueous extract, which was stored in a refrigerator at 2 °C. All operations were performed at room temperature.

## 2.2. Membrane Materials

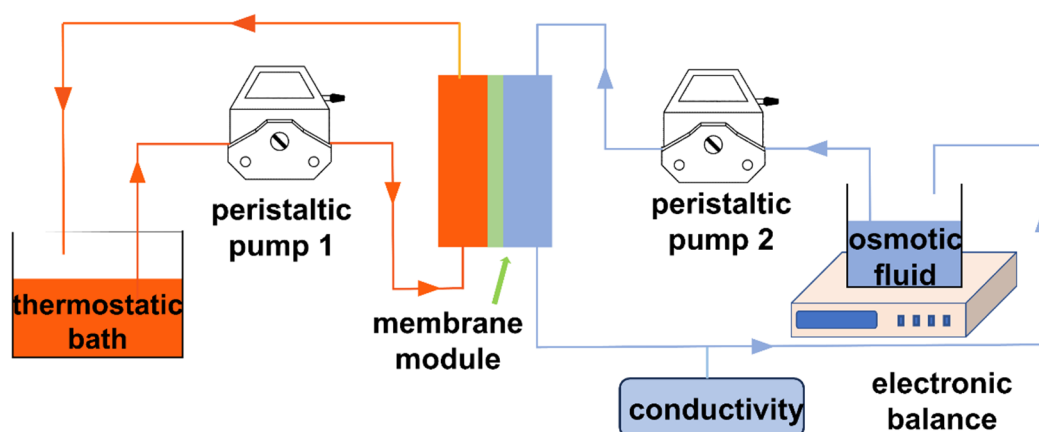
In this paper, two pore sizes of polytetrafluoroethylene (PTFE) hydrophobic membranes are investigated. The nature of the membranes and related information is shown in Table 1.

**Table 1.** Membrane-related parameters for DCMD.

Membrane Materials	Membrane Pore Size ( $\mu\text{m}$ )	Membrane Thickness ( $\mu\text{m}$ )	Contact Angle ( $^\circ$ )	Availability of Support Layer	Producers
polytetrafluoroethylene	0.45	230	121.8	Yes	Shanghai Furnace
polytetrafluoroethylene	0.2	230	121.8	Yes	Shanghai Furnace

## 2.3. MD Settings

The MD experimental setup established for this experiment is shown in Figure 1. After being heated by a thermostatic magnetic stirrer (Lichen Scientific Instrument Co., Ltd. (Shanghai, China), SZCL-1000), the feed was sent through a peristaltic pump (LEADFLUID Co., Ltd. (Baoding, China), BT-600 L) to the hot side of the flat membrane module. The cold side liquid is cooled by a cold well (Zhengzhou Kotai Experimental Equipment Co., Ltd. (Zhengzhou, China), DLSK-3/15) and sent to the cold side of the module by another peristaltic pump. The permeate was measured in real time by an electronic balance (Shanghai Yaoxin Electronic Technology Co., Ltd. (Shanghai, China), LQ-A2001). The effective membrane area of the membranes used in this study was  $0.004 \text{ m}^2$ .



**Figure 1.** Diagram of the DCMD experimental setup.

## 2.4. Optimisation of MD

In this part, the effects of feed temperature ( $45\sim 65 \text{ }^\circ\text{C}$ ), hot-side flow rate ( $200\sim 1000 \text{ mL}\cdot\text{min}^{-1}$ ), cold-side flow rate ( $200\sim 1000 \text{ mL}\cdot\text{min}^{-1}$ ), condensate temperature ( $5\sim 25 \text{ }^\circ\text{C}$ ), and membrane pore size on the efficiency of the MD process were investigated. At the end of each experiment, conductivity was measured using a conductivity meter (Shanghai Jingmagnet Instrument Co., Ltd. (Shanghai, China), DDS-307A). The concentration of the concentrate was also measured using a high performance liquid chromatograph (Shimadzu Corporation (Kyoto, Japan), LC-20AOXR). All experiments were performed three times to evaluate the stability of the results.

## 2.5. Cleaning of Contaminated Membranes

In this section, the cleaning effect of different cleaning agents on contaminated membranes is examined. The test membranes were first subjected to a 6-h membrane distillation experiment to accelerate their contamination. The contaminated membranes were cleaned at room temperature for 20 min at a flow rate of  $800 \text{ mL}\cdot\text{min}^{-1}$  either alone or with different acids and bases as feed solution. After cleaning, rinse with ultrapure water for 5 min to remove residual dirt and cleaning agent. The optimal cleaning method was selected by comparing the membrane surface contamination and flux recovery after cleaning with different reagents. Reagents such as HCl and NaOH were purchased from Sinopharm Chemical Reagent Co. (Shanghai, China). Scanning electron microscopy analyses confirmed the contamination patterns on untreated, contaminated and clean membrane surfaces. Images of the film samples were obtained by scanning electron microscopy (Zeiss (Oberkochen, Germany), SIGMA 500) after sputter plating of the air-dried sample surfaces with gold. In order to analyse the changes in hydrophobicity of the two membranes before and after the experiments, the dried membranes were tested by a contact angle meter (Zhongchen Digital Technology Apparatus Co., Ltd. (Shanghai, China), JC2000D) using the contact angle as an indicator.

## 2.6. Performance Indicators

In order to evaluate the performance of DCMD, the variation of key performance indicators with operating parameters and membrane materials was investigated. In this paper, permeate flux was used as the main performance index of DCMD.

$$J = \frac{m}{A * \Delta t} \quad (1)$$

where  $J$  is the permeate flux,  $\text{kg} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$ ,  $m$  is the permeate mass,  $\text{kg}$ ,  $A$  is the membrane area,  $\text{m}^2$ , and  $\Delta t$  is the operation time,  $\text{h}$ .

The change in contact angle of the PTFE membrane was used in the experiments in this chapter to measure the degree of membrane contamination and the cleaning effect. Where the cleaning efficiency (CE) was calculated using 2 [26].

$$CE = \frac{A_2 - A_1}{A_0 - A_1} \times 100\% \quad (2)$$

Where,  $A_0$ -contact angle of the original membrane, °,  $A_1$ -contact angle of the contaminated membrane, °;  $A_2$ -contact angle of the cleaned membrane, °.

## 2.7. Direct Contact Membrane Distillation Heat Transfer Modelling

In the DCMD process, heat and mass transfer occur simultaneously. As shown in Figure 2, in which the transfer of heat can be divided into three main stages:

- (1) The main body of the heated raw material liquid stream transfers heat to the surface of the feed side film by convective heat transfer,  $T_1$ ;
- (2) One part of the heat is transferred to the permeate side as latent heat of vapour through the volatile components of the feed solution, and the other part of the heat is transferred to the permeate side as sensible heat through the heat conduction of the diaphragm,  $T_2$ ;
- (3) Eventually, the volatile component vapours that pass through the membrane undergo heat exchange with the refrigerant in the external condenser, transferring heat to the refrigerant,  $T_3$ .

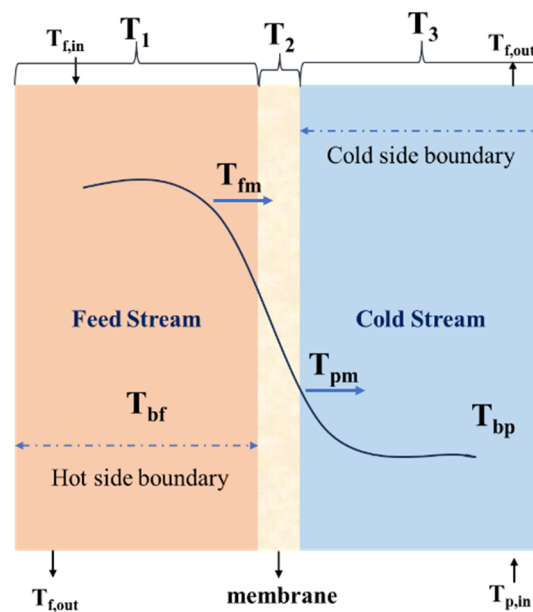


Figure 2. Schematic diagram of heat transfer in the DCMD process.

This leads to Equation:

$$h_f(T_{bf} - T_{fm}) = h_m(T_{fm} - T_{pm}) + J\Delta H_v = h_p(T_{bp} - T_{pm}) \quad (3)$$

Among them:  $h_f$  is the feed heat transfer coefficient,  $T_{bf}$  is the feed temperature, and  $T_{fm}$  is the membrane surface temperature on the hot side;  $h_m$  is the membrane heat transfer coefficient,  $T_{pm}$  is the membrane surface temperature on the cold side,  $J$  is the permeate flux determined by the vapour pressure difference across the

hydrophobic membrane, which is the core driving force of the thermally driven MD process and is essentially different from the trans-membrane pressure driving flux in pressure-driven membrane processes, and  $\Delta H_v$  is the latent heat of evaporation;  $h_p$  is the heat transfer coefficient of condensate, and  $T_{bp}$  is the condensate temperature.

The temperature polarisation coefficient (*TPC*) equation can also be obtained as:

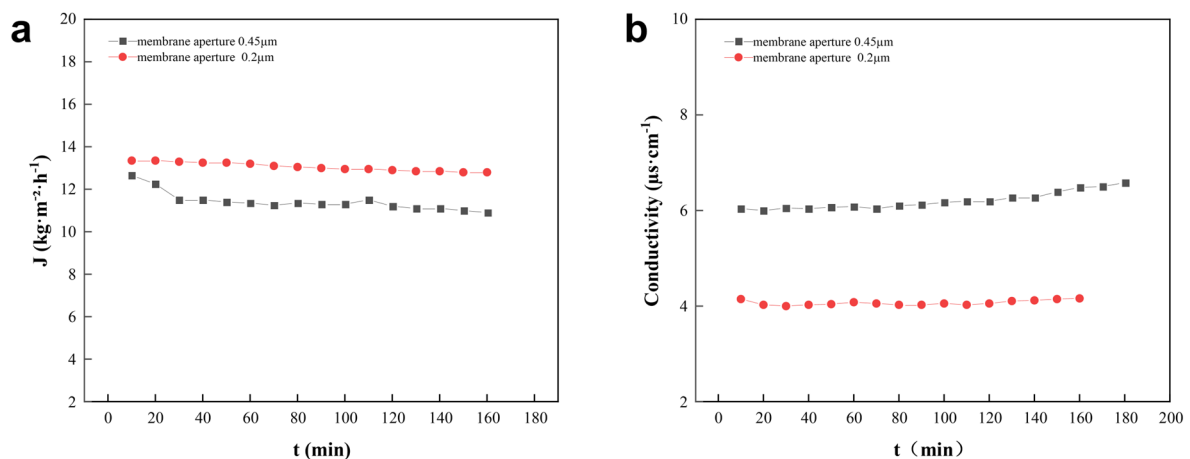
$$TPC = \frac{T_{fm} - T_{pm}}{T_{bp} - T_{bp}} \quad (4)$$

### 3. Results and Discussion

#### 3.1. Effect of PTFE Membranes with Different Pore Sizes on the Experiments

In order to evaluate the stability of PTFE membranes with different membrane pore sizes in the concentration process of traditional Chinese medicine, Panlangen was selected as the raw material liquid, and a continuous experiment was carried out for 180 min, and the results are shown in Figure 3. As shown in a, along with the increase of time, the membrane flux of PTFE membranes with different membrane pore sizes showed a decreasing trend. This is due to the long experimental process, along with the Chinese medicine aqueous extract concentration multiples increase, the viscosity of the material liquid also gradually increased, resulting in a decrease in the saturated vapour pressure of water, so the temperature polarisation of the hot side of the phenomenon is aggravated, so that the membrane flux appeared to be a slow decline in the trend. At the same time, contaminants in the feed solution accumulate on the membrane surface, which also causes a reduction in membrane flux [27].

Figure 3b then shows the change in conductivity during the experiment for both. The figure clearly shows that the conductivity of the 0.2 aperture is far more stable than that of the 0.45 aperture in the later stages of the experiment. The latter has shown an increasing trend in conductivity, indicating a decrease in the retention performance of its membrane and wetting of the membrane surface. The significant increase in conductivity of the 0.45  $\mu\text{m}$  membrane can be mainly attributed to its relatively large pore size, which is more prone to interact with saponin components in the Chinese medicine extract. The larger pore size facilitates the adsorption and penetration of surface-active saponins, which further accelerates the loss of membrane surface hydrophobicity and the occurrence of membrane wetting. The saponin components with surface activity in TCM extracts are the main organic pollutants causing membrane fouling, and their adsorption on the membrane surface further leads to the decrease of membrane hydrophobicity and the occurrence of membrane wetting, which is the core reason for the increase in conductivity [28].

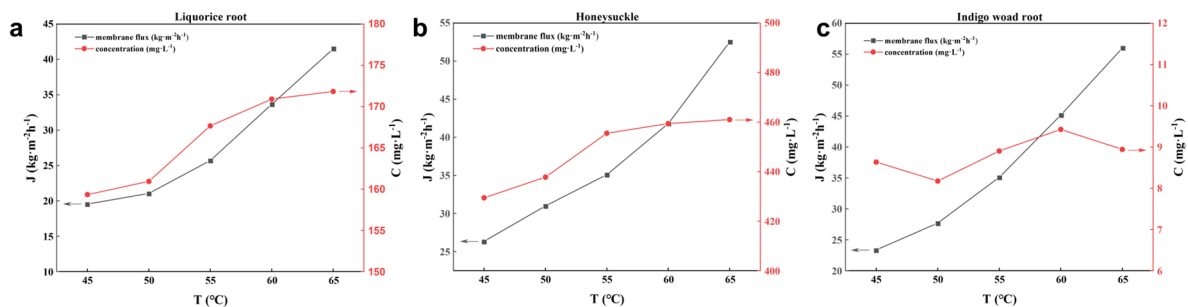


**Figure 3.** Effect of different membrane pore sizes on membrane fluxes.

#### 3.2. Influence of Feed Liquid Temperature on the Aqueous Extract of Traditional Chinese Medicine

Figure 4 shows the trends of membrane flux and material concentration of the aqueous extracts of different traditional Chinese medicines under different temperature conditions. The three herbs, from left to right, were: liquorice root, honeysuckle and indigo woad root, and temperatures of 45  $^{\circ}\text{C}$ , 50  $^{\circ}\text{C}$ , 55  $^{\circ}\text{C}$ , 60  $^{\circ}\text{C}$  and 65  $^{\circ}\text{C}$  were chosen as the operating conditions. Membrane flux increases exponentially with increasing temperature. In previous studies, the cause of this phenomenon has been attributed to the reduction in feed viscosity due to high temperatures, but the rapid increase in the saturated vapour pressure of the liquid on the feed side should be the underlying cause of the phenomenon. According to the Antoine equation [29], the vapour pressure of the feed

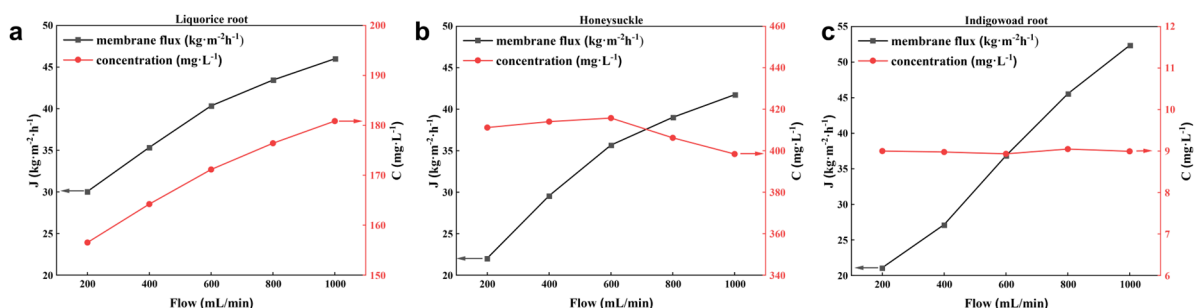
liquid is exponentially related to the temperature on the feed side. At the same time, due to the complexity of the composition of traditional Chinese medicine, in which there is no lack of a variety of heat-sensitive components, so the figure can clearly see that the effect of temperature on the concentration of the material liquid is extremely significant. Generally after 55 °C, the increase in material concentration tends to equilibrate, which is due to the thermal instability of the active ingredients in Chinese medicine.



**Figure 4.** Effect of hot side temperature on membrane flux and aqueous extract concentration.

### 3.3. Effect of Hot-Side Flow Rate on the Aqueous Extract of Traditional Chinese Medicine

Figure 5 demonstrates the trends of membrane flux and feed solution concentration of DCMD-concentrated aqueous extract of traditional Chinese medicine with different feed solution flow rates at the hot side. The data of the three kinds of Chinese medicine extracts show three obviously different trends, indicating that the differences in herb species and extract composition directly affect the concentration variation during membrane distillation. When the hot-side flow rate was increased from 200 mL·min<sup>-1</sup> to 600 mL·min<sup>-1</sup>, the membrane flux appeared to increase substantially, but when the flow rate continued to increase, the rise in membrane flux appeared to decrease. With the increase of hot-side flow rate, the feed solution shows a slight downward trend, which is mainly attributed to the alleviation of concentration polarization on the membrane surface. Higher flow rate enhances convective mass transfer and reduces the accumulation of retained components near the membrane surface, resulting in a relatively low concentration in the bulk feed solution. It is always known that increasing the hot-side flow rate increases the shear force on the membrane surface, so most of the contaminants that were brought to the membrane surface will revert back to the main body of the feed. At the same time, increasing the flow rate can increase the transmembrane flux and intensify the deposition of contaminants on the membrane surface. When the flow rate exceeds 600 mL·min<sup>-1</sup>, the deposition of organic pollutants on the membrane surface cannot be completely eliminated, and mild membrane fouling leads to a slowdown in the growth rate of permeate flux, which is consistent with the organic fouling mechanism of the DCMD process. The different trends among the three Chinese medicine systems are mainly due to the differences in the types and contents of organic components such as saponins and polysaccharides in the extracts, resulting in different degrees of membrane fouling, polarization and separation behavior. Therefore, in the later stages of the experiment, the flux decreased due to the factor of membrane contamination.

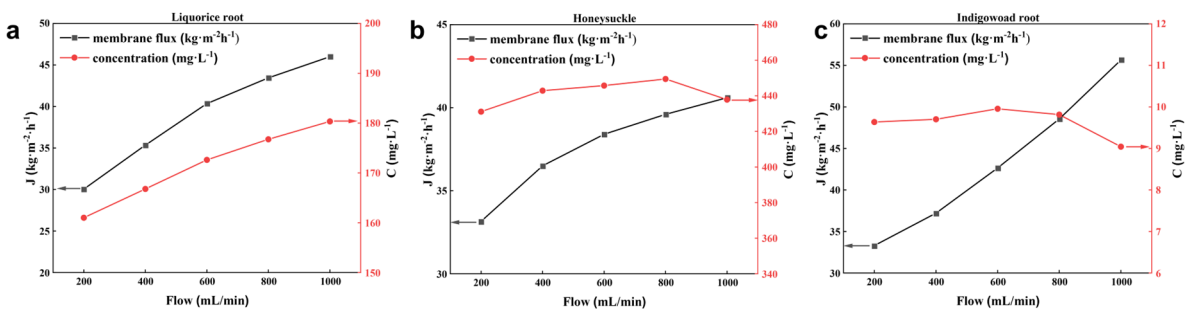


**Figure 5.** Effect of hot-side flow rate on membrane flux and aqueous extract concentration.

### 3.4. Effect of Cold-Side Flow Rate on the Aqueous Extract of Traditional Chinese Medicine

In Figure 6, it can be seen that the membrane flux shows an exponential increase when the condensate flow rate is elevated from 200 to 1000 mL·min<sup>-1</sup>, but the change of condensate flow rate does not have a great effect on the concentration of the feed solution. According to the classical boundary layer and heat-mass transfer theory of

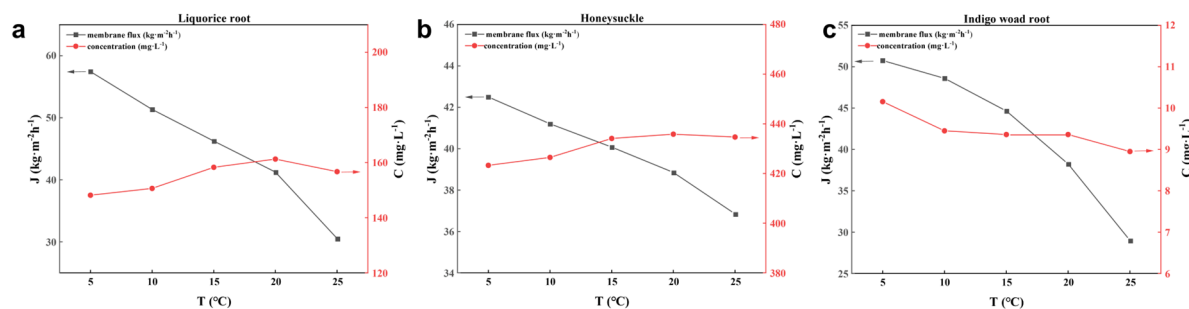
membrane distillation, higher flow rate enhances the turbulence intensity near the membrane surface, which can effectively reduce the thickness of the thermal and mass boundary layers. This theoretical mechanism supports that the concentration polarization and temperature polarization on the membrane surface can be significantly weakened with the increase of flow rate, which is also verified by the obvious increase of membrane flux in Figure 6. During the experiment, as the condensate flow rate increases, it causes the average temperature of the condensate inside the membrane module to decrease. At the same time, the increase in flow rate can effectively reduce the thickness of the boundary layer of the membrane, the concentration polarisation and temperature polarisation on the membrane surface are weakened, and the water production is effectively increased.



**Figure 6.** Effect of cold-side flow rate on membrane flux and aqueous extract concentration.

### 3.5. Effect of Condensate Temperature on the Aqueous Extract of Traditional Chinese Medicine

Figure 7 shows the effect of condensate temperature on membrane flux and feed solution concentration while keeping the feed solution temperature constant. For the experiments, temperatures of 5 °C, 10 °C, 15 °C, 20 °C and 25 °C were selected. With the other three conditions unchanged, the membrane flux tends to decrease as the condensate temperature increases in a linear relationship with temperature. The effect on the concentration of the material does not change much.



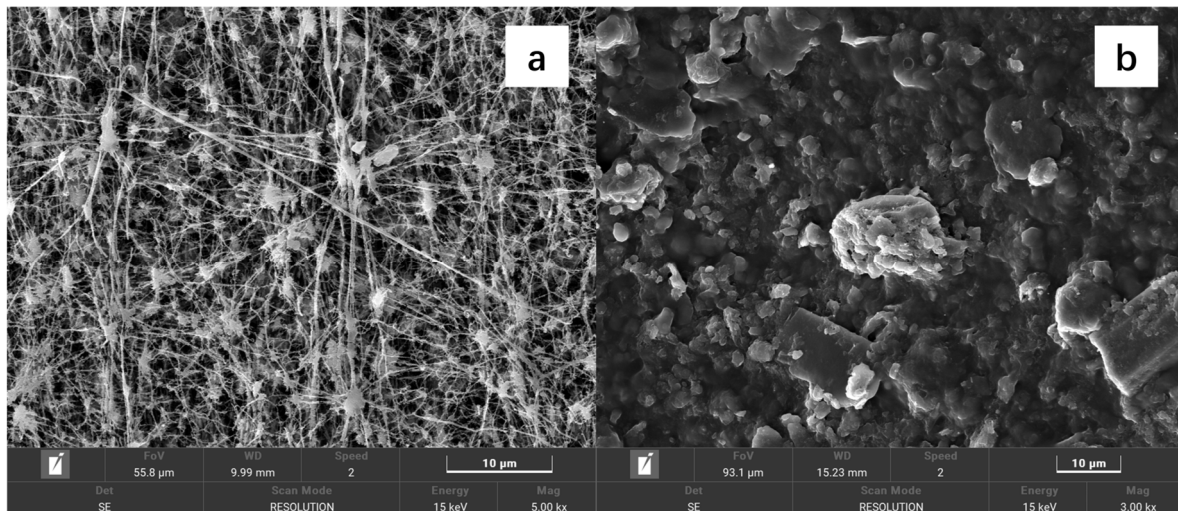
**Figure 7.** Effect of cold side temperature on membrane flux and aqueous extract concentration.

Comparing Figures 4–7, it is found that increasing the hot-side feed temperature has a huge impact on both membrane flux and feed concentration compared to the other three. This is mainly due to the fact that the fraction of matter at a given kinetic energy is exponentially dependent on temperature, and therefore the saturated vapour pressure increases exponentially with temperature. However, the phenomenon of membrane contamination is a non-negligible problem in the experimental process, and how to mitigate membrane contamination is an important issue in the study of membrane distillation.

### 3.6. Membrane Contamination Analysis and Characterisation

Combined with the definition of the membrane fouling mechanism in the MD process, membrane fouling in this study was dominated by the adsorption of organic components in TCM aqueous extracts on the surface of PTFE membranes. The following characterization and analysis of membrane surface morphology and element composition were carried out to verify the characteristics of the organic fouling and its influence on membrane performance. Figure 8 shows the scanning electron micrographs of the original PTFE membrane as well as the contaminated membrane. As can be seen from the figure, the porous structure formed by the interweaving of membrane fibres in the original membrane is clearly visible, but there is a porous contamination layer on the surface of the membrane after contacting with the extract of traditional Chinese medicine. It is clear that the

contamination layer is formed by the accumulation of particles. The continuous deposition of particles leads to a gradual increase of the scaling layer and a gradual increase of the thermal resistance, resulting in a decrease of the observable flux and the original pore structure is no longer visible.



**Figure 8.** (a) Scanning electron micrograph of primary membrane. (b) Scanning electron micrographs of contaminated membranes.

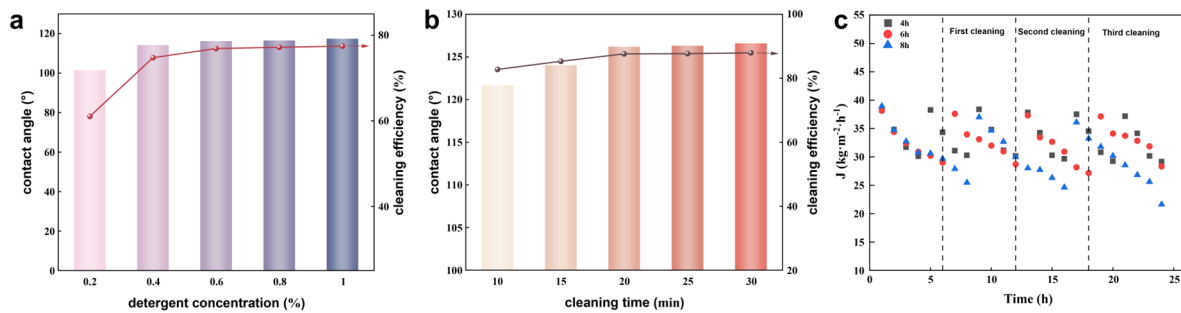
Figure S4, corresponding to Figure 8, shows elemental analyses of the surface of the original and contaminated membranes. As shown in the figure, essentially only two elements, C and F, are present on the surface of the original membrane. As for the contaminated membrane, the content of element F on the membrane surface decreased greatly from 73.4 wt% to 1.0 wt%, and the content of element O increased from 1.1 wt% to 55.1 wt%, as well as the content of element C and element N increased differently, which could indicate that the contaminated membrane surface accumulated a large amount of organic matter, which is consistent with the characteristics of the traditional Chinese medicine extracts that contain a large amount of organic matter. The significant increase in the contents of oxygen and nitrogen elements on the surface of the fouled membrane further confirmed that the fouling layer was composed of organic substances in TCM extracts, which was consistent with the organic fouling mechanism of the MD process defined in this study.

### 3.7. Selection of Cleaning Agents

Cleaning methods for membrane contamination can be divided into physical and chemical cleaning [30]. In this experiment, the contact angle as well as the cleaning efficiency were used as measures to clean the contaminated membrane. According to Figure S5, the cleaning effect of different cleaning agents on the contaminated membrane is demonstrated. It can be seen that no matter what kind of cleaning agent for the contamination of the membrane has a certain cleaning effect, in which the cleaning effect of alkaline cleaning agent is much greater than the other kinds. Based on previous studies, it is known that acidic detergents have excellent removal capabilities for  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ , while alkaline detergents such as NaOH are more effective for organic contaminants [31,32]. And in Figure S4, it can be seen that the fouling layer on the contaminated membrane is mainly composed of organic compounds, which can also explain why NaOH cleaning is optimal. Therefore, NaOH was used as the cleaning agent in the subsequent experiments.

### 3.8. Optimisation of Cleaning Agent Concentration

As can be seen in Figure 9a, the cleaning efficiency increases as the concentration of the cleaning agent increases. When the concentration reaches 0.6, the concentration continues to increase and the cleaning effect rises insignificantly. This is due to the fact that the negative charge on the surface of the pollutant may have already reached its maximum, so the structural looseness of the dirt layer can no longer continue to increase, so the cleaning efficiency of the membrane is no longer significantly increased [27,33]. At the same time, it should also be taken into account that when the concentration of the cleaning agent is too high, it can cause damage to the fibre structure of the membrane and reduce its service life. In summary, 0.6 wt% as the cleaning agent concentration has achieved better results.



**Figure 9.** (a) Effect of different detergent concentrations on contaminated membranes; (b) Effect of different cleaning times on contaminated membranes; (c) Effect of different cleaning cycles on contaminated membranes.

### 3.9. Optimisation of Cleaning Time

The optimisation of the cleaning time also has a greater impact on the removal of membrane contamination, the longer the cleaning time, the longer the reaction time of the reagent and the pollutant, the greater the removal of contamination, but too long a period of time will exacerbate the destruction of the reagent on the membrane material, which affects the service life of the membrane. Different cleaning time under the membrane flux changes with time as shown in Figure 9b, cleaning time for 20, 25 and 30 min cleaning efficiency is similar, which indicates that the cleaning time is set to 20 min, the reaction between the reagents and pollutants has been saturated, and then continue to extend the cleaning time not only can not enhance the effectiveness of the treatment of membrane contamination, but also reduces the service life of the membrane. Therefore, it is most appropriate to set the cleaning duration to 20 min.

### 3.10. Optimisation of Cleaning Cycles

After determining the cleaning method, the choice of cleaning cycle on the removal of membrane contamination efficiency also has a greater impact, if the cycle is longer, the pollutants gathered on the surface of the membrane will gradually be compacted under the action of the pressure of the water inlet and even enter the membrane holes, thus increasing the difficulty of cleaning, while the cleaning cycle is too short, the chemical reagents of the frequent rinsing will result in the destruction of the nature of the membrane material, affecting the service life of the membrane [34]. The changes of membrane flux with time under different cleaning cycles are shown in Figure 9c, the most obvious decrease of membrane flux was observed in the case of 8 h as a cleaning cycle, and the decrease of membrane flux was closer to that in the case of 4 h and 6 h as a cleaning cycle, and the cleaning effect of 4 h as a cleaning cycle was slightly better than that of 6 h, but the comprehensive consumption of reagents and the service life of the membrane were taken into consideration, and the membrane flux decreased in the case of 6 h as a cleaning cycle. However, considering the reagent consumption and membrane service life, a 6-h cleaning cycle is more cost-effective.

## 4. Conclusions

In this study, we mainly investigated the concentration effect of DCMD with PTFE planar membrane material as the core on the extract of traditional Chinese medicine under different conditions, and verified the universality of DCMD in the concentration process of traditional Chinese medicine. The main conclusions are as follows:

- (1) Different pore size PTFE membrane although all have excellent retention effect, but 0.2 pore size PTFE membrane has higher permeate flux, lower energy consumption and stronger wetting resistance.
- (2) Hot-side temperature and cold-measurement temperature have a significant effect on the permeation flux of the DCMD process. For flow, it is important to note that an increase in flow rate increases the infiltration flux because the thermal boundary resistance observed at high flow rates decreases.
- (3) In the long-term operation need to consider the problem of membrane pollution. The use of different methods to clean the contaminated membrane can extend the service life of the membrane and reduce operating costs. The main pollutants in the Chinese medicine processing and cleaning wastewater are organic matter, and chemical cleaning with alkaline solution can effectively remove the dirt layer.

This study innovatively verified the applicability of DCMD in complex TCM organic systems, clarified the regulatory mechanism of membrane fouling and wetting, and optimized a targeted cleaning system, laying a crucial experimental and theoretical foundation for the industrial application of MD technology in TCM concentration. Future research will focus on process scale-up, membrane surface modification and energy-saving system design,

and expand the technology to the concentration of compound TCM extracts to improve its industrial application system in TCM and other organic separation fields.

### Supplementary Materials

The additional data and information can be downloaded at: <https://media.sciltp.com/articles/others/2604131622297428/MH-26010060-SM.pdf>. Figure S1: Flow chart of Chinese medicine preparation. Figure S2: Application of membrane distillation. Figure S3: Membrane contamination mechanism diagram. Figure S4: Surface elemental analysis of raw (a) and contaminated (b) membranes. Figure S5: Contact angle of PTFE film with different cleaning agents. Figure S6: SEM images of the surface and cross-section of the original membrane and the membrane after different cleaning cycles.

### Author Contributions

Y.M.: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Visualization, Writing—original draft. L.W.: Conceptualization, Data curation, Formal analysis, Investigation, Visualization. J. F.: Methodology, Conceptualization, Data curation, Formal analysis, Visualization. L.Z.: Data curation, Formal analysis. H.W.: Project administration, Resources. C.L.: Conceptualization, Funding acquisition, Methodology, Writing—review & editing, Funding acquisition, Project administration, Supervision, Writing—review & editing. All authors have read and agreed to the published version of the manuscript.

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### Institutional Review Board Statement

Not applicable.

### Informed Consent Statement

Not applicable.

### Data Availability Statement

All data supporting the findings are included within the article and its Supplementary Materials files.

### Conflicts of Interest

The authors declare no conflict of interest.

### Use of AI and AI-Assisted Technologies

No AI tools were utilized for this paper.

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