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# Experimentally Guided Neural Network and Statistical Forecasting of Membrane Water/Salt Selectivity with Minimal Mean Errors

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## ABSTRACT

Membrane life and performance are crucial factors in adopting membrane-based processes for water treatment and separations. This study investigated various time series models using hold-out validation of experimentally generated water vapor flux and salt rejection rates. Membrane properties were optimized by incorporating carbon-based nanomaterials to enhance anti-wetting and porosity, developing correlations between membrane characteristics and high fluxes. Fine-tuned Autoregressive Integrated Moving Average (ARIMA), Prophet, Exponential Smoothing, and Neural Prophet models were trained on an experimental dataset (N=434) collected over 36 hours to forecast performance for 72 hours. Results demonstrate the superiority of the Exponential Smoothing statistical model in predicting and forecasting membrane performance, yielding the lowest root mean square error (RMSE) of 0.006 and mean absolute error (MAE) of 0.007. This outperformance is attributed to its non-linear data fitting approach, which employs weighted averages to mitigate non-stationary behavior in time series data, a characteristic often observed in membrane performance over time. While other models showed promise, they did not match the accuracy of Exponential Smoothing in this context. The proposed modeling approach offers a more efficient alternative to traditional experimental studies, potentially leading to significant cost and time savings in the research and development phase of membrane distillation processes. This method's applicability to various membrane types and operational conditions warrants further investigation.

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## 1. Introduction

Membrane based separation processes such as membrane distillation (MD), reverse osmosis [1], filtration [2], electrodialysis [3] remains a popular method for water desalination and treatment of produced water [4, 5]. MD remains a versatile desalting technique that offers advantages such as low energy input via lower operational temperature and pressure and theoretically has nearly 100% rejection of non-volatile feed streams compared to the conventional methods [6-10]. Despite an increase in the need for clean water and the potential of MD as an emergent technology, its conventional use is limited by low membrane potency specific to high performing permeate flux [5]. To address this, polymer membranes are being modified via top-down approach of surface coating with nanomaterials or functional groups or bottom-up approach of creating mixed polymer membrane/nanomaterial matrix to synthesize membranes with tunable properties specific to membrane distillation [11-15]. Another critical factor for industrial scalability is the membrane performance and life under MD operating conditions, which remain a challenge due to the time and labor-intensive nature of MD experiments [16-19]. However, due to the recent surge in data training and predictive analysis techniques, it is possible to develop experimentally informed models that can forecast membrane performance and fluxes for longer duration of runs. Such approaches will further support effective MD process design and the development of membrane cleaning procedures to enable membrane reuse for an extended duration.

The use of an artificial neural network (ANN) platform to accelerate the design of efficient nanocomposite membranes through material discovery and select process parameters for membrane distillation operation has been reported in handful of studies. For instance, Dudchenko *et al.* [20] developed an ANN model to improve the accuracy of empirical methods by estimating the heat transfer and membrane permeability, while Fetanat *et al.* [21] fitted a feed-forward ANN model with Bayesian regularization for predicting nanocomposite membrane performance. Binger *et al.* [22] investigated ANN to predict the effect of spacer geometries on pressure loss and concentration polarization in membrane channels. However, few studies have reported time-dependent membrane performance and distillation studies based on experimental datasets. Although ARIMA models have been reported for the evaluation of the long-term performance of MD desalination on Polyvinylidene fluoride (PVDF) membrane [23] and the prediction of membrane fouling of ultrafiltration membranes based on the volumetric flux prediction [24], these studies predicted the MD performance based single time-series model that does not account for the type of data generated such as linear vs nonlinear or their combination and the resultant forecast failed to recognize the nonlinear time series (TS) of MD performance data.

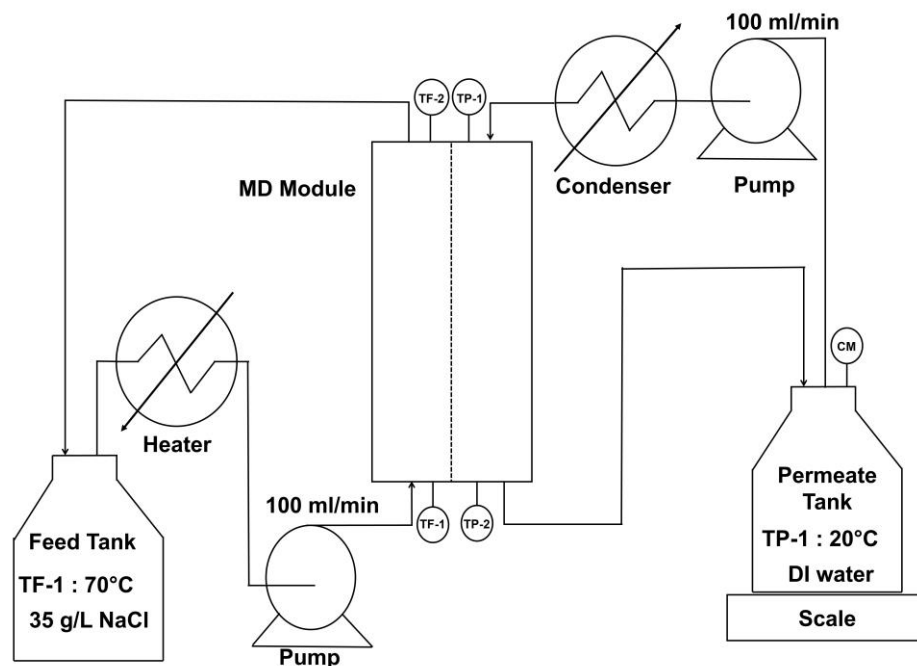
To overcome the limitations of the studies mentioned earlier, this work evaluated a range of time-series forecasting models, including statistical forecasting methods such as Autoregressive Integrated Moving Average (ARIMA) and Exponential Smoothing (ES), as well as machine learning models like Prophet and Neural Prophet. The models were trained on an experimental dataset of water vapor flux and salt rejection rates with respect to time. Prophet and Neural Prophet were chosen specifically because both frameworks are designed for time-series forecasting tasks that may exhibit non-linear trends, seasonality, and irregular sampling intervals. The aim of this work was to evaluate whether these neural-network-based (or hybrid) models—known for capturing complex patterns and for their relative ease of hyperparameter tuning—could outperform more traditional statistical methods. Given the inherent ability of neural network-based machine learning (ML) models to capture complex non-linear relationships and temporal dependencies, it was hypothesized that these models would outperform traditional statistical models in time-series forecasting tasks [25]. Moreover, Neural Prophet extends Prophet by incorporating an auto-regression component (AR-Net), which can in principle capture longer-term dependencies [26, 27]. Specifically, it was anticipated that the neural network-based ML models would demonstrate superior prediction accuracy and generalization to unseen data. This hypothesis was tested using an experimental dataset, with model performance evaluated based on appropriate metrics such as Mean Absolute Error (MAE) and Root Mean Squared Error (RMSE). Contrary to the hypothesis, findings of this study indicate that the statistical exponential smoothing model yielded the best prediction with the lowest mean errors when comparing model prediction and experimental results. Despite the potential of deep learning algorithms in different fields, these models underperformed in forecasting water vapor flux and salt rejection rates. Therefore, this study emphasizes the importance of choosing the most suitable machine learning model tailored to data training, testing, and prediction to achieve the lowest possible error rates. This is especially useful in material discovery and process design, which is expensive and laborious but can potentially benefit from predictive multivariate time series

analysis. The findings of this work further highlight the importance of evaluating multiple forecasting methods, as the best-performing model can vary depending on the unique characteristics of the dataset and the specific problem being addressed.

## 2. Methods

### 2.1. Membrane Distillation Setup and Experiments

PVDF membranes were modified using the bottom-up wet phase inversion approach by adding carbon nanofibers (CNF) and copper (Cu) nanoparticles decorated CNF fabricated using top-down thermal in-situ growth approach [28], into PVDF polymer matrix. The fabricated membranes were characterized using scanning electron microscopy and ImageJ analysis (pore size distribution), and tensiometer (static water contact angle). All the experiments were triplicated using sodium chloride solution at inlet and outlet flow rates of 100 ml/min. Fig. (1) represents the flow diagram of MD experiments with inlet and outlet streams. The hot and cold feed temperatures were maintained at  $70 \pm 2^\circ\text{C}$  and  $20 \pm 2^\circ\text{C}$  respectively. Water vapor flux  $J_v$  ( $\text{kg}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ ) and salt rejection  $R$  (%) were recorded. The water vapor flux,  $J_v = m/(At)$  (Equation 1) correlates  $m$ , the mass of the overflow collected,  $A$  the active area of the membrane, and  $t$  duration of MD run. While salt rejection (%) efficiency represented as  $R(\%) = (1 - C_p/C_f) \times 100$  (Equation 2) compares the conductivities of the permeate ( $C_p$ ) and the feed ( $C_f$ ). Salt rejection and water vapor flux were recorded for three membranes for 36 hours at a 5 min sampling time resulting in a total of 434 data points, out of which 85% of the experimental data until 32<sup>nd</sup> hour was used for training and the remaining dataset for 4 hours were used to test the accuracy of statistical and machine learning models.



**Figure 1:** Process Flow Diagram for Direct Contact Membrane Distillation (DCMD) used in study. TF-1 = Temperature of Feed in, TF-2 = Temperature of feed out, TP-1 = Temperature of permeate in, TP-2 = Temperature of permeate out, and CM = conductivity meter.

### 2.2. Statistical Forecasting: ARIMA and Exponential Smoothing (ES)

Autoregressive Integrated Moving Average (ARIMA) was used in this study because it is a linear statistical model that recognizes non-zero correlation between the consecutive data points for time-series data fitting for higher accuracy forecasting [29, 30]. ARIMA protocol involved determining whether the time series is stationary or nonstationary. The application of autocorrelation function to water vapor flux data revealed non-stationarity or

fluctuation with time for all the experimental data. The details of non-stationarity analysis of the experimental data are provided in SI as Fig. **S1** and Table **S1**. Based on this analysis the ARIMA parameters (p, d, f.) for data training were optimized and summarized in Table **S2**. The Akaike Information Criterion and size of ARIMA are reported in Table **S3**. Exponential smoothing was chosen as an alternative study due to its capability to provide better predictions compared to other statistical models [31]. The exponential smoothing method gives reliable forecasts based on the weighted average of the past observations with highest weight associated to the most recent observation [32, 33]. The summary of the parameters used for Exponential smoothing model optimization are summarized in Table **S4**.

### 2.3. Time Series Based Machine Learning Models: Prophet and Neural Prophet

Prophet and Neural Prophet, open-source forecasting specialized tools for modeling nonlinear time series were also evaluated. The model is represented by  $Y_t = g_t + S_t + h_t + \epsilon_t$  (Equation 3) where parameters  $S_t$  corresponds to periodic changes,  $h_t$  to irregularities, and  $\epsilon_t$  as the error to account for any idiosyncratic changes. One advantage of Prophet over traditional statistical methods is its ability to handle missing and irregularly spaced data values, a common occurrence during large-scale MD operations [26]. To optimize the Prophet algorithm, a grid search method with an absolute error accuracy metric was used. The grid search parameters included the number of change points and the changepoint prior scale. Shorter MD runs of 36h showed higher sensitivity to the model parameters, particularly to `changepoint_prior_scale`, impacting the prediction significantly. [34].

Neural Prophet, an improved Prophet neural network time-series data modeler includes Auto-Regressive (AR) similar to ARIMA but models the dynamics of an AR (Auto-Regressive) process using a feed-forward neural network approach suitable for sequential MD experimental data [27]. Another advantage is its scalability to estimate long-range dependencies in case of MD processes that were trained on data collected at fine granularities, this this case, minutes. Neural prophet architecture consisted of six components represented as:  $Y_t = g_t + S_t + h_t + f_t + A_t + L_t$  (Equation 4) where, additional parameters  $f_t$  represent the regression effects for future-known exogenous variables,  $A_t$  corresponds to auto-regression effects based on past observations, and finally,  $L_t$  is the regression effects at time  $t$  for lagged observations of exogenous variables. Incorporating these elements into a model can help capture the dynamics of a membrane process more accurately. It incorporated additional parameters for future-known exogenous variables such as feed water composition and temperature. Auto-regression to predict the water vapor flux and salt rejection rate values based on its previous values. Regression effects for lagged observations of exogenous variables such as the effect of process parameters on the current water vapor flux. Furthermore, these parameters were tuned using grid search process to obtain the combination that produced the best model performance.

### 2.4. Performance Evaluation Metrics

The accuracy of each of the constructed predicting model for water vapor flux and salt rejection dataset was evaluated by Root-mean-square error (RMSE) and mean absolute error (MAE) [35, 36] using ( $y$ ) experimental test dataset values and ( $\hat{y}$ ) are the predicted value based on  $N$  data points. RMSE represents the standard deviation of the difference between the forecast and actual data, while MAE is the absolute mean error. Lower values of MAE and RMSE imply higher accuracy, indicating the model's predictions closer to the actual values on average.

$$RMSE = \frac{\sum_{i=1}^N (y_i - \hat{y}_i)^2}{N} \quad (\text{Equation 5})$$

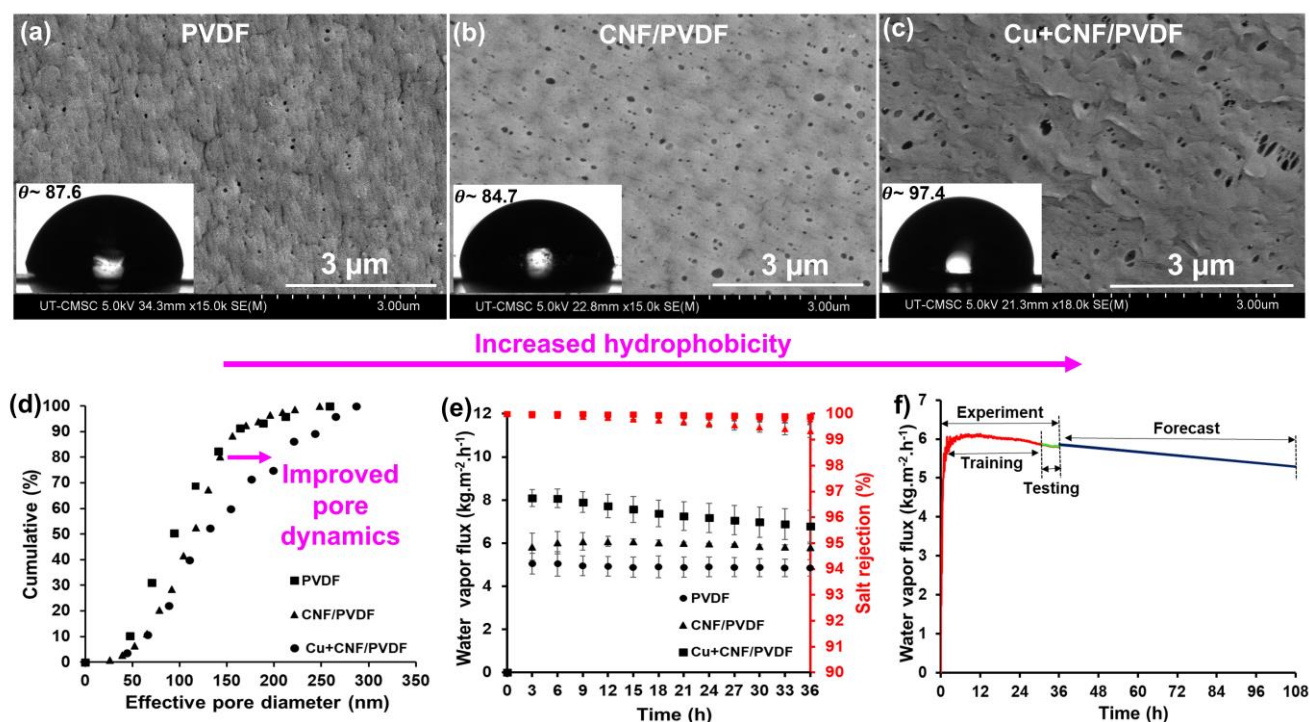
$$MAE = \frac{\sum_{i=1}^N |y_i - \hat{y}_i|}{N} \quad (\text{Equation 6})$$

## 3. Results and Discussion

### 3.1. Membrane Distillation Experiment and Data Training

Our comprehensive experimental approach involved adopting a bottom-up phase inversion approach to incorporate a wide range of concentration of copper nanoparticles and carbon nanofibers in the PVDF polymer

matrix to tune the porosity, crystallinity, and wettability of the membranes. The superior performing membranes comprised of multidimensional Cu and CNF particles incorporated in PVDF membranes to improve their permselectivity via induced chemical heterogeneity, improved pore size distribution, contact angle hysteresis and hierarchical morphologies confirmed by electron microscopy, ImageJ analysis, and tensiometry. Fig. (2) demonstrates the variable porosity from circular pores in (a) PVDF membranes, to ellipsoidal pores in (b) CNF/PVDF membranes to larger ellipsoidal pores in Cu+CNF/PVDF membranes. Additionally, static contact angles increased with the addition of multidimensional Cu and CNF nanomaterials in PVDF membrane resulting in increased hydrophobicity. ImageJ image processing technique confirmed the large pore size distribution of Cu+CNF/PVDF membranes in Fig. (2d). The water vapor flux and salt rejection rate studies demonstrated a 64% increase in water vapor flux and a salt rejection rate of over 99.8% with just 1wt% loading of Cu+CNF in the PVDF matrix as seen in Fig. (2e). While Fig. (2f) schematically summarizes the time-series modeling steps that involved training on data obtained for first 32 hours, subsequently, data obtained by the models between 32<sup>nd</sup> and 36<sup>th</sup> hours were used for testing, followed by forecasting for 72 hours.



**Figure 2:** a-c) Electron Microscopy and static contact angle of PVDF, CNF/PVDF, and Cu+CNF/PVDF membranes, d) pore size distribution, e) water vapor flux and salt rejection rates of PVDF, CNF/PVDF and Cu+CNF/PVDF membranes, f) schematic representation of time-series modeling steps

Preliminary dataset analysis yielded negative for missing values and evaluated z scores for outlier detection were insignificant as seen in Fig. (SI-S2) [37]. To preserve the authenticity of the data, no further resampling or other preprocessing steps were performed. Salt rejection and water vapor flux were recorded during MD runs for 36 hours at a 5 min sampling time resulting in a total of 434 data points, out of which 85% of the experimental data (or until 32<sup>nd</sup> hour) was used for training the statistical and neural forecasting models while the remaining dataset for 4 hours were used to measure the accuracy of statistical and machine learning models.

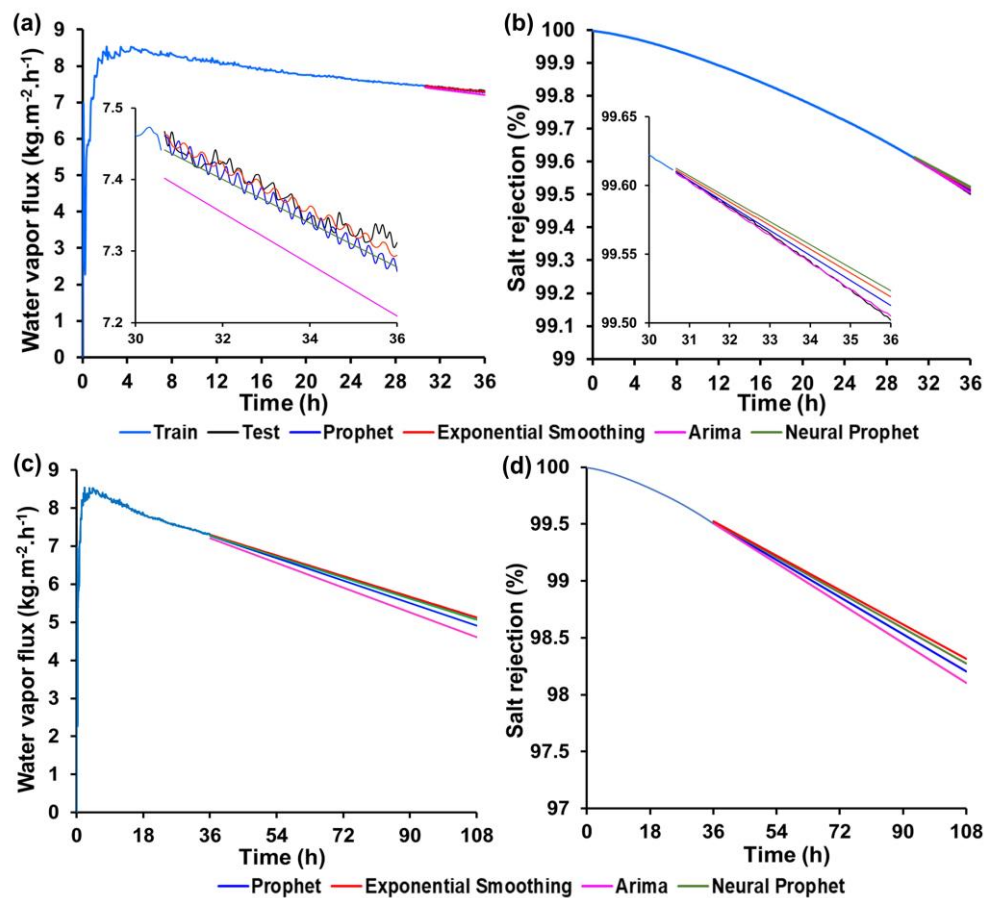
### 3.2. Performance Evaluation of Statical and Neural Network Models for Data Fitting and Time Series Forecasting

The statistical and machine learning model that provides a good fit to the historical data does not necessarily result in accurate forecasts because of the changing data patterns over time that a model may fail to capture. Therefore, ARIMA, Exponential Smoothing, Prophet and Neural Prophet models were evaluated based on their experimental data training and testing accuracies of water vapor and salt rejection rates obtained for best

performing Cu+CNF/PVDF membranes. This study adopted the hold-out validation approach based on its simplicity and speed, both water vapor flux and salt rejection data sets were split into a training set and a test (or hold-out) set. Each statistical and ML model was trained on the training set and their performance were evaluated on the unseen test set. Alternatively, cross-validation involving computing averages overall all iterations of testing and training can be adopted. Fig. (3a) and (3b) illustrate the performance of the models on 4-hours test dataset. Meanwhile, Fig. (3c) (water vapor flux) and (3d) (salt rejection rate) depicts the forecasting performance of the models for the subsequent 72 hours.

Among all the tested models, exponential smoothing yielded the best prediction with the lowest error rate, while the predictive performance of Neural Prophet model was the poorest on test dataset. Neural Prophet performance drops dramatically and predict an increase in PVDF water vapor flux for the next 72 hours where the trend of water vapor flux is clearly downward. In terms of comparison between Prophet vs Neural Prophet, our results show no significance difference between these two models. Both models have the lowest accuracy metrics in predicting salt rejection and water vapor flux in three experimental MD.

In MD studies, the exponential smoothing models demonstrated superior performance by generating forecasts based on the weighted averages of past observations of water vapor flux and salt rejection over the initial 32 hours. The model assigned exponentially decreasing weights to older observations, thereby giving more importance to recent observations. The flexibility of exponential smoothing, which involves adjusting parametric values to disregard older experimental observations during analysis, enabled the model to adapt more effectively to the experimental data. The optimal performance of exponential smoothing can be attributed to its non-linear data fitting approach, which employs weighted averages to mitigate fluctuations in the data [38].



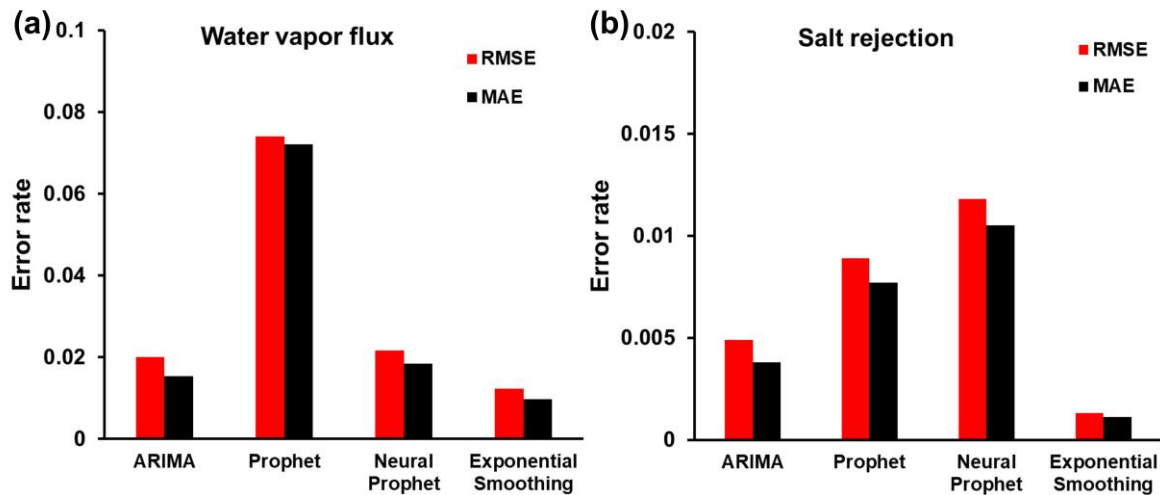
**Figure 3:** Comparison of experimental data training and testing splits for (a) Water vapor flux and (b) Salt rejection rate using ARIMA, prophet, neural prophet, and exponential smoothing models. Comparison of models' performance for forecasting 72 hours for (a-c) Water vapor flux of PVDF, CNF, Cu+CNF, and (d-f) Salt rejection of PVDF, CNF, Cu+CNF for ARIMA, prophet, Neural Prophet, and exponential smoothing.



Additionally, the models were also compared for their accuracy by computing RMSE and MAE as summarized in Fig. (4). Exponential Smoothing demonstrated the least errors, attributable to its intrinsic property of employing a weighted average of the training data and prioritizing recent observations over older ones for forecasting. This approach proved particularly effective in this study, given the consistent temporal trend observed in the water vapor flux and salt rejection data

In the context of this study, the Root Mean Square Error (RMSE) values serve as a measure of the average discrepancy between the model's predictions and the actual observations. Consequently, the minimal errors of 0.012 for water vapor flux and 0.0013 for salt rejection rate, as yielded by the exponential smoothing model, signify its superior performance in comparison to the other models examined in this study. Since RMSE values are sensitive to outliers, the lowest RMSE values corroborate with the smaller z-score obtained during outlier computation (Refer Fig. S2). The largest RMSE was obtained for Prophet and Neural Prophet models for water vapor flux and salt rejection rate. This is attributed to several factors, for instance the addition of AR-Net2 in Neural Prophet as an extension of the basic Prophet model adds complexity to the model, thereby increasing the learnable parameters dramatically which could result in overfitting. The computed smoothing coefficients  $\alpha$ ,  $\beta$  and  $\gamma$  used to adjust the influence of different components of the time series vapor flux and salt rejection data are summarized in Table S4 of SI.

The lower values of the smoothing coefficients represent the suitability of exponential smoothing model to capture the underlying patterns in the data, leading to more accurate forecasts and yielding lower MAE and RMSE values [39]. However, the performance of these models can vary depending on the specific characteristics of the data and the tuning of the model parameters. It's also worth noting that while MAE and RMSE are useful for comparing different models, they don't provide a complete picture of a model's performance. Other factors, such as the interpretability of the model and its ability to capture uncertainty, should also be considered when evaluating and comparing forecasting models.



**Figure 4:** MAE and RMSE results for models used in this study on salt rejection and vapor flux test dataset.

These findings suggest that while deep learning has merits, traditional statistical-based time series models remain a viable and sometimes preferable alternative, particularly for specific use cases such as the one explored in this study. Evaluations of time series models, guided by experimental data, have confirmed that deep learning does not always provide superior results [40-42]. On the contrary, conventional TS models may not be optimal for datasets spanning extensive durations since models such as ARIMA and Exponential Smoothing require careful tuning of their parameters, making optimizing parameters time-consuming. Meanwhile, models like Neural Prophet can be complex and computationally intensive. This could pose computational resources and time challenges, especially for large datasets. Despite these challenges associated with each forecasting model, with careful implementation and consideration of these potential issues, these models can still serve as valuable tools in predicting membrane life and stability of membrane distillation processes.

It's important to acknowledge that the dataset, consisting of 434 carefully collected data points under controlled laboratory conditions, may present fewer irregularities than typically encountered in industrial settings. This relatively clean and consistent data allowed conventional statistical models to perform strongly. However, in real-world applications where data often contain more noise, outliers, or missing points, advanced machine learning approaches might demonstrate clearer advantages. Additionally, the work focused on water vapor flux and salt rejection as the primary indicators of membrane distillation performance, it is recognized that additional parameters could provide a more comprehensive evaluation of MD systems. Future studies could expand the model to include other relevant metrics such as thermal efficiency, energy consumption, permeate quality beyond salt rejection, and long-term membrane stability indicators. Furthermore, incorporating additional input variables, such as feed properties, membrane characteristics, and operational parameters, could potentially highlight the advantages of machine learning methods in handling multivariate complexity. This expanded approach would provide a more robust comparison between ML and statistical methods, potentially yielding different results than those observed in our current simplified model. As evidenced by large-scale forecasting competitions, the performance of different modeling approaches can vary depending on the specific characteristics of the dataset and the nature of the problem being addressed [43]. Therefore, while the findings in this work demonstrate the effectiveness of statistical methods for this particular dataset, further research with more complex, multivariate datasets is necessary to fully explore the potential of ML methods in membrane distillation performance prediction. Future research could extend the work by adapting the ML models for inverse design, potentially enabling the identification of optimal membrane design parameters for target water vapor flux and salt rejection rates. This approach could provide valuable insights for membrane material development and process optimization. Additionally, comparing the performance of ML and traditional statistical methods in such optimization tasks could further elucidate their respective strengths and limitations in the context of membrane distillation research.

## 4. Conclusion

This study presents a systematic approach to experimentally designing high-performing membranes for separation processes, coupled with an evaluation of suitable forecasting models to assess membrane life and stability over extended periods. The focus on experimentally guided time series (TS) model evaluation demonstrated the effectiveness of statistical time series forecasting models in predicting membrane distillation process performance over longer durations, yielding minimal mean errors. Error analysis comparing predicted and experimental data revealed smaller discrepancies for the exponential smoothing statistical model. The findings of this work highlight the importance of meticulous model selection, considering data characteristics and study objectives. Future research could explore alternative validation approaches and additional models to enhance prediction accuracy. This may include applying advanced deep learning models for multivariate time series analysis in membrane design, focusing on interfacial membrane characteristics such as chemical structure, pore dynamics, wettability, and surface area. The methodology presented here can be extended to time series-based design and discovery of membranes, potentially revolutionizing the approach to membrane development and optimization in various separation processes.

## Conflict of Interest

The authors declare that there are no non-financial or financial conflicts that could have potentially impacted the findings presented in this paper.

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## Availability of Data and Materials

The data, code, and information of materials used in this article are available upon request.



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## Author Contributions

JA conceptualized the study, developed the code for machine learning models, and was a major contributor in writing, reviewing, and editing the original manuscript. SM provided the experimental data, performed a formal analysis, curated the data, and contributed to writing, reviewing, and editing the manuscript. AG acquired the funding required for this research, administered the project, supervised the investigation, and conceptualization of this study, and contributed to writing, reviewing, and editing the manuscript.

## Supporting Information

Experimental data acquisition, Statistical Forecasting: ARIMA and Exponential Smoothing (ES), Outliers Detection, Akaike Information Criterion (AIC) and size of ARIMA, Computation of Mean Absolute Error (MAE) and its parameters.

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