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Carboxylic acid concentration by forward osmosis processes: Dynamic modeling, experimental validation and simulation

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HIGHLIGHTS

- Acid permeability coefficients were determined by pressure-retarded osmosis model.
- The dynamic process model of forward osmosis was proposed and validated.
- The model could quantify the performance of FO process as a function of time.
- The optimal initial condition region could be provided by 3D simulation.

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ABSTRACT

Driven by osmotic pressure, forward osmosis has attracted growing attention in desalination, water purification and wastewater reuse application. The performance behavior of FO process relies upon not only the membrane properties but also the process operation parameters. In addition, the desired process optimization can be defined from these operation parameters. To profoundly comprehend the forward osmosis performance, related to the process operation parameters during concentrating carboxylic acid, the mathematical model was developed by thoroughly enumerating and integrating each single logical phenomenon equation with pertinent variables during the operation of FO process, which aims to emulate a large-scale FO process with a plate-and-frame module configuration. By means of Levenberg-Marquardt algorithm, the 51 precise dependent process variables were simultaneously determined, as functions of time (Dynamic Simulation Model). FO experimental process was carried out to verify the developed model under the same operating conditions by using Thin-Film Composite (TFC) FO membrane whereas feed solution was varied types of carboxylic acids. The good agreement between model predictions and experimental data was observed. The concentration performances of FO process at 30 h system operation for acetic, butyric, valeric and lactic acid were 1.65, 2.2, 2.3 and 2.5 fold increase, respectively, quantified by developed model. As 3D simulation, not only is the developed model aimed to forecast the optimum initial conditions of draw solution for the best concentration performance of FO process, but the results can also be used as guidelines to select the effective regions of initial conditions to achieve the nearby maximum concentration performance.

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1. Introduction

Carboxylic acids are widely used as basis of chemical in many industries, such as food beverages, pharmaceutical and chemical industries. Acetic acid is an important chemical in many industries, such as synthesis of cellulose acetate, printing, textile, dyeing and also food industries [1]. On the other hand, petrochemical and wood pulping industries usually discharge wastewater, polluted with carboxylic acids. The acid waste streams discharged from these manufacturing processes, generally contain low amount of organic acids such as acetic acid concentration less than 5% from terephthalic acid process and valeric acid concentration in the range of 0.5 to 2.5 g/l from waste stream of nylon manufacturing [2,3].

The membrane processes have been recognized as effective processes to successfully treat organic acid waste streams and to retrieve the diluted acid organic pollutants [3–5]. Based on desalination technology, forward osmosis (FO) becomes more globally recognized and continuously developed as a result of the depletion

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of fossil fuel leading to the escalating of fuel price [6,7]. Unlike the typical pressure-driven membrane processes, forward osmosis requires less energy because it can operate under low hydraulic pressure. The driving force is developed by differential osmotic pressure between opposite sides of the membrane which is intrinsically generated by the difference in concentration gradient between low concentration of feed solution side and high concentration of draw solution side. The forward osmosis process requires no osmotic agent separation or recovery. This will draw more attentions with regard to less resource and energy consumptions. In some remote irrigated areas, where fresh water is shortage, the forward osmosis process can possibly extract water from feed solution and also produces the diluted fertilizer which can be directly utilized in irrigation [8]. To simulate this application of forward osmosis process, therefore, this research selects one molar of NH_4Cl as draw solution to represent the fertilizer.

The other advantages of the forward osmosis are high contaminant rejection and low membrane fouling, compared to the traditional pressure-driven membrane processes [9]. The treatment of nutrient-rich liquid stream, anaerobic digester centrate from dewatering of digested biomass, has been researched both batch and continuous operations of FO process [10]. In food processing, forward osmosis process for concentrating a variety of liquid foods has promoted high potentials of forward osmosis membrane technology for food industries application [11,12]. Both effects of pH and temperature on forward osmosis membrane flux by using rainwater as the makeup water source for cooling water dilution have been investigated [13]. Forward osmosis desalination is also applied in oil and gas manufacturing wastewater which has been addressed in the impacts of membrane selection and operating conditions (hydrodynamic) on the performance of FO process [14]. As a result of their large specific membrane area, the performance of CTA HF membranes has been evaluated with regard to various operating conditions such as draw solution concentration, membrane orientation, cross flow velocity and temperature [15].

In each forward osmosis application, the process performance which is evaluated by water recovery, permeate water flux and solute rejection are clearly governed by the inherent membrane properties itself, anti-ICP and anti-fouling along with water permeability, solute rejection and the structure parameter of the membrane [16]. The another essentially external factor is process operation parameters that definitely affect to system efficiency such as the characteristics of feed and draw solutions (type, concentration and capacity), the liquid temperature in the process and the flow velocity in the membrane module as well as the module design and the operation time of the process. These parameters also play the significant role in the performance of FO process. However, no such a research has specifically described these parameters in aspect of dynamic modeling to express or model their behaviors to the system performance over operating time. For this reason, the current research is intended to develop dynamic process model as the useful tool that can help to predict how the system efficiency or flux behavior will change over time by varying operating conditions and therefore assist to optimize performance with no experiment time consuming. This dynamic process model can also be implemented as a guideline to practically configure the initial operating parameters of the up-scale FO process with flat sheet membrane used in plate-and-frame modules so that the optimal performance can be obtained by considering both operational and economical aspects as a whole.

The main objective of this research is to develop the dynamic process model, developed by systematically investigating and compiling each single logical phenomenon equation and associated variables during the operation of forward osmosis process, simulated by using low concentration of single carboxylic acid (10 mM of acetic acid, butyric acid, valeric acid and lactic acid) as feed solution and 1 M NH_4Cl as draw solution. By means of Levenberg-Marquardt algorithm, the developed model is able to determine all precise dependent process variables varying over time during simulating the carboxylic acid concentration in forward osmosis process. In order to achieve this goal, the relevant performance membrane constants which are water permeability coefficient, NH_4Cl permeability coefficient and structure parameter, have to be determined and evaluated from corresponding water flux and solute flux models, derived from FO mode configuration (AL-facing-FS) [17] while acid permeability coefficients are defined from proposed water flux and solute flux models which are derived from PRO mode configuration (AL-facing-DS). After accomplishment of model development, the model is validated against the well-controlled FO experiment under the same operating conditions where pH of feed solution and weight change of draw solution are employed as experimental data for model validation. The developed model of this research can also forecast the behaviors of other carboxylic acids and thus offers the further profound understanding and advancing knowledge in this area which can be contributed to future application of wastewater reuse for a large-scale FO process in more pragmatic manner.

2. Process modeling of forward osmosis

Fig. 1 illustrates carboxylic acid and salt concentration gradients across a semi-permeable membrane and depicts water transport, acid flux and salt flux operating in FO mode at any moment during filtration. As natural physics phenomena, osmosis generates the osmotic pressure driving the water molecules through a semi-permeable membrane from diluted feed solution to highly concentrated draw solution. The feed solution consequently becomes more concentrated while the draw solution is diluted over the period of time.

2.1. Permeate water permeability

Referred to classical solution-diffusion model, the water flux, J_w, across the active layer in a Fig. 1 relies on the different osmotic pressure, expressed as

$$J_w = \sigma A(\pi_i - \pi_{mf}) \tag{1}$$

where A is the water permeability coefficient of the membrane, π_i is the osmotic pressure at interface of support layer-active layer, π_{mf}

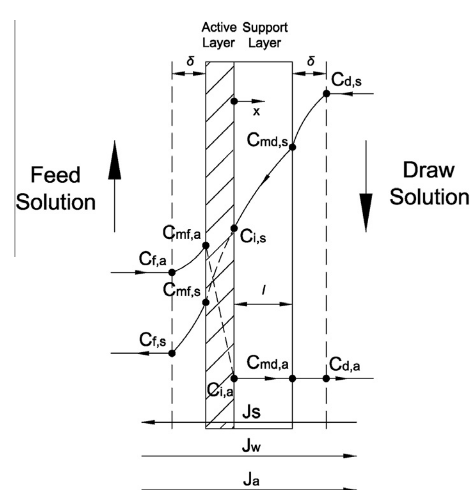

Fig. 1. Schematic of water transport, acid and salt concentration gradients across a semi-permeable membrane for acid concentration by FO process.

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