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Gas permeation through H_2-selective mixed matrix membranes: Experimental and neural network modeling

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ABSTRACT

Gas permeability through synthesized polydimethylsiloxane (PDMS)/zeolite 4A mixed matrix membranes (MMMs) were investigated with the aid of artificial neural network (ANN) approach. Kinetic diameter and critical temperature of permeating components (e.g. H_2, CH_4, CO_2 and C_3H_8), zeolite content and upstream pressure as input variables and gas permeability as output were inspected. Collected data of the experimental operation was used to ANN training and optimum numbers of hidden layers and neurons were obtained by trial-error method. The selected ANN architecture (4:10:1) was used to predict gas permeability for different inputs in the domain of training data. Based on the results, the predicted values demonstrate an excellent agreement with the experimental data, with high correlation ($R^2 = 0.9944$) and less error (RMSE = 1.33E−4). Furthermore, using sensitivity analysis, kinetic diameter and critical temperature were found as the most significant effective variables on gas permeability. As a result, ANN can be recommended for the modeling of gas transport through MMMs.

1. Introduction

In the past decade, membrane technology has attracted more attention for gas and liquid separation [1–5]. Some of the more prevalent gas separations by membranes are air separation, hydrogen recovery and natural gas purification [6–10]. Nonporous polymeric membranes give high selectivity but poor permeability. On the other hand, porous inorganic membranes give high permeability but poor selectivity. For better results, both selectivity and permeability in a membrane should be balanced. Mixed matrix membranes (MMMs), comprised of molecular sieving materials embedded in polymer matrixes, have potential to provide high performance gas separation membranes [11,12]. Generally, a combination of sol–gel reaction and polymerization was used to synthesize the hybrid materials [13–16]. The resulting hybrid materials present the advantages of each material; for example, flexibility and selectivity of polymers and thermal stability of ceramics. Among these hybrid membranes, polydimethylsiloxane (PDMS)/zeolite MMMs have been received the most attention for gas permeation studies. Jia et al. were the first to investigate the zeolite-filled rubbery polymer membranes composed of PDMS and silicalite-1 [17]. It was concluded that the silicalite-1 plays the role of molecular sieve in the membrane by facilitating the permeation of smaller molecules but hindering permeation of larger ones, although its pore openings are larger than the size of the permeating gases. To fully understand these phenomena, more systematic studies of important factors (e.g., dispersion state of inorganic nanoparticles into polymer matrix, effect of different kinds of permeating species, upstream pressure and temperature) are needed. Using Artificial Neural Network

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(ANN) approach, it may not be necessary to carry out an entire series of expensive pilot or full-scale tests to collect and verify separation data. The primary advantage of ANN over theoretical models is that, as a black box model, it does not prerequire any governing equation specifically describing the fundamental engineering phenomena. Instead, ANN can be trained the complex transport processes of a system from given observed data, serving as an instrument for universal data approximation. Accordingly, a considerable number of ANN applications for prediction, classification and optimization can be found in various fields such as economics, chemistry and chemical engineering, computer science, water resources engineering etc. [18–25]., which examplify its great predictive potential for transport processes.

Among assorted neural networks, the multi-layer feedforward neural network with backpropagation training algorithm, typically called backpropagation neural network (BPNN), are one of the most widely used networks for information prediction and classification [26,27]. Application of BPNN for membrane research is a great topic because it provides promising results. Extensive research has been performed to investigate the capability of BPNN as a tool for membrane systems modeling [28–30].

To the best of our knowledge, any BPNN application for MMMs processes has not been reported yet. The goal of this research is to study BPNN application for PDMS/zeolite MMMs gas separation process and to explore its predictability for gas permeabilities as function of effective variables.

2. Experimental

2.1. Materials and methods

Zeolite nanoparticles (particle size = 80–250 nm and Si/Al molar ratio = 1) were provided from Research Institute of Petroleum Industries, Iran. Toluene was purchased from Merck and used as received. Crosslinker (polyhydrogenmethylsilane under trade name of V24), silicon oil (RTV 615A en B, density 1.02 g/mL) and catalyst (1,3-divinyl-1,1,1,3-tetramrthyldisiloxane platinum complex under trade name of OL) were supplied by Wacker Silicones Corporation, Adrian; MI.

H_2, CO_2 and CH_4 gases with purity of 99.5% supplied by Technical Gas Service, Inc., and C_3H_8 gas with purity of 99.9% supplied by Air Product and Chemicals, Inc., were used as feed gases.

Gas permeability was determined according to our previous study [31,32].

2.2. Membrane preparation

Initially, zeolite 4A nanoparticles were activated at 200 °C for 12 h in vacuum condition. Adequate zeolite nanoparticles were dispersed in toluene and the mixture was stirred for 1 h (S1). Ultrasonic was then employed for 10 min to produce a homogenized mixture. Crosslinker was added to the mixture. Thereafter, the obtained mixture was stirred and sonicated for 30 and 10 min, respectively.

Autonomously, a solution (S2) of silicon oil and crosslinker was prepared as reported in our previous works [33,34].

The conventional solution-casting method was employed for PDMS/zeolite 4A MMMs synthesis. The final mixture (S3) was obtained by appending the solution to the mixture (S1 + S2) and stirred for 2 h. Finally, catalyst was added to the final mixture (S3). The casting mixture composition was 10:0.1:0.5:100 weight ratios of silicon oil, crosslinker, catalyst and solvent, respectively.

Casting was carried out using a glassy casting die supported by a Teflon plate. The cast film was dried in a vacuum oven for 36 h at 25 °C. In order to eliminate the voids between the zeolite nanoparticles and the polymer phase, high processing temperature was used. Therefore, residual solvent removing and polymer crosslinking process completing were performed for 2 h at 80 °C. Finally, the homogenous PDMS/zeolite 4A MMMs films were separated from the glassy die whereas they were transparent.

3. ANN modeling

3.1. Artificial neural networks

Artificial neural networks (ANNs) are numeric techniques able to capture and represent complex input–output relationships [35]. They have the ability to learn linear, as well as non-linear correlative patterns between sets of input data and corresponding target values, directly from the data set

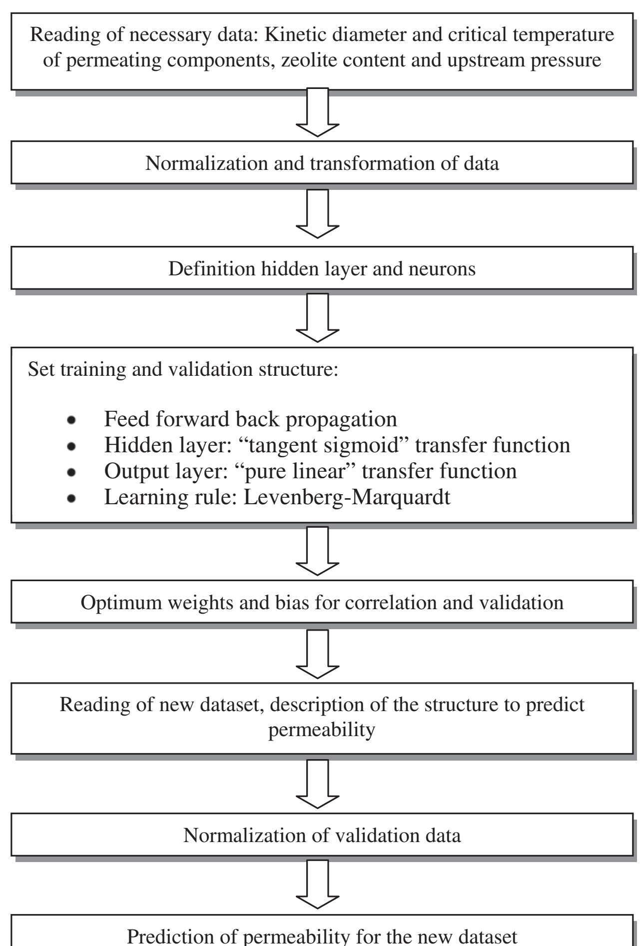

Fig. 1 – Flow diagram for the ANN modeling.