Estimation of ethylene/1-butene copolymerization conditions using the autoencoder model

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Abstract Chain microstructures of ethylene/1-butene copolymers produced at specific polymerization conditions can be calculated using the copolymerization kinetic model. However, this kinetic model cannot be solved inversely to estimate polymerization conditions from desired microstructures. In this work, the autoencoder (AE) model, the machine learning techniques based on artificial neural network (ANN) concept, was developed to help estimate polymerization conditions to produce polymers with desired microstructures (e.g., molecular weight distribution (MWD), chemical composition distribution (CCD), and number and weight average molecular weight). Two models were developed in this work: one with only microstructural distributions (MWD and CCD) and the other with additional information on average microstructures and polymer yield. The results showed that the proposed AE models can adequately estimate polymerization conditions from desired microstructures with acceptable mean square error (MSE). More specific microstructures lead to better estimation of polymerization conditions with lower MSE.

1. Introduction
Ethylene/1-butene copolymers with desired properties can be produced by tailor-making chain microstructures (i.e., molecular weight distribution (MWD), chemical composition distribution (CCD), number average molecular weight (Mₙ), weight average molecular weight (Mₜ), and average comonomer content (CC)). Polymer microstructures obtained at specific polymerization conditions can be calculated using the polymerization kinetic model [1-3]. However, this kinetic model cannot be solved inversely to give polymerization conditions from desired microstructures.

Artificial neural network (ANN) models have been developed to help estimate polymerization conditions required to produce polymers with desired microstructural distributions (MWD and CCD), average properties (Mₙ, Mₜ, and CC) and yield (Y) [4, 5]. The previous results showed that multiple solutions may occur because different sets of polymerization conditions can be used to produce polymers with similar microstructures. This can lead to large discrepancies in estimated conditions and key operating conditions must be specified to ensure the unique solution.

An autoencoder (AE) model, which is also ANN based model, can potentially be used to solve this problem and to help estimate the most likely conditions. Autoencoder is an interconnected network, consisting of two parts: encoder and decoder. The network attempts to generate output data similar to the feed input data through the training session. An encoder is a part of the network that compresses the
input data to the point of maximum compression called the latent space, while a decoder reconstructs the data [6, 7]. Once trained, this model can be used to help suggest the missing information from the feed dataset, which are operating conditions in this case.

In this work, autoencoder model was developed to determine appropriate polymerization conditions to produce polymers with desired microstructures. Two models were investigated: one with only microstructural distributions (MWD and CCD) and the other with both microstructural distributions, averages polymer properties, and polymer yield.

2. Model Development

2.1 Copolymerization kinetic model

In this work, the ethylene/1-butene copolymerization kinetic model was used to calculate microstructural data from given polymerization conditions. The training and testing datasets were generated using the previously proposed kinetic model [1-3], which is based on the population balance concept with pseudo-kinetic rate constants for the copolymerization in a continuous stirred tank reactor (CSTR) at the steady-state condition. The kinetic parameters from the previous work [8] were used. The ranges of polymerization conditions used in this study are summarized in table 1.

| Polymerization Condition | Unit       | Maximum | Minimum |
|--------------------------|------------|---------|---------|
| Ethylene, [Et]           | mol L⁻¹    | 0.2     | 3.74    |
| 1-Butene, [Bu]           | mol L⁻¹    | 0.03    | 1.05    |
| Hydrogen, [H₂]           | mol L⁻¹    | 0.0001  | 0.01    |
| Cocatalyst, [A]          | mol L⁻¹    | 0.0003  | 0.0143  |
| Temperature, T           | °C         | 60      | 90      |
| Molar flow rate catalyst, Cᵐ | Mol s⁻¹ | 0.00001 | 0.02    |

2.2 Autoencoder (AE)

Autoencoder (AE) is ANN based model used to learn a representation for dataset with interconnected network. An encoder is the part of the network that compressed the input data for reduction side called the latent space. Along with encoder, a decoder is learned to reconstruct the data from encoder as close as possible to the input data. The general architecture of autoencoder is shown in figure 1 [6, 9].

Figure 1. General architecture of autoencoder (AE) network.

An autoencoder (AE) model has learned to make an output data similar to an input as shown in equation (1) [7, 10].
\[ X' = f_{w,b}(X) \approx X \]  

where \( X' \) is output from the model, the function \( f_{w,b} \) can be any non-linear function, and \( X \) is input data.

The autoencoder passes input data through the encoding section of the model that consists of hidden layers and encoding the input data into some representation output \( Y \) as shown in equation (2). Then, \( Y \) is forwarded to the decoding section to reconstruct the data \( X' \), which is similar to input data \( X \), by equation (3). The model is optimized by adjusting the model parameters \((w, w', b, b')\) to minimizing the reconstruction error between output from the model \( (X') \) and input data \( (X) \) of the training data.

\[ Y = \emptyset(wX + b) \]  
\[ X' = \emptyset(w'Y + b') \]

where \( \emptyset \) is non-linear activation function, \( b \) and \( w \) are biases and connection weight in the encoding section, and \( b' \) and \( w' \) are biases and connection weight in the decoding section.

In this work, a specific type of autoencoder called denoising autoencoder (DAE) model was used to estimate ethylene/1-butene copolymerization conditions from the required microstructures. The development of this model was inspired by the human ability to recognize objects, even if they were partially obscured or damaged. The operation of DAE model is similar to that of autoencoder, but different types of input data are required, such as partially corrupted input data \((X)\) with randomly missing data. The DAE model was used to estimate the missing value through interpolation by learning other data with similar characteristics [9, 11, 12].

The mean squared error (MSE) between (1) the output data \( (X') \) from the model with corrupted input data \((X)\) and (2) input data \( (X) \), as shown in equation (4), was used as the reconstruction loss function to evaluate the model performance. This function was minimized by adjusting weights and biases in the network.

\[ MSE = \frac{1}{N} \sum_{i=1}^{N} (X - X')^2 \]  

This work developed two DAE models to determine polymerization conditions to yield desired microstructures: Model A for determination of polymerization conditions from only desired microstructural distributions (MWD and CCD) and Model B for determination of polymerization conditions from desired microstructural distributions (MWD and CCD), average microstructures \((M_n, M_w, \text{and} \ CC)\) and polymer yield \( (Y) \). The estimated conditions include ethylene concentration, \([Et]\); 1-butene concentration, \([Bu]\); hydrogen concentration, \([H_2]\); cocatalyst concentration, \([A]\); polymerization temperature, \(T\); and the total molar flow rate of catalyst, \(C^{\text{in}}\).

A total of 3,125 datasets was used: 2,500 or 80% as training datasets and 625 or 20% as testing datasets. Each dataset contains corrupted input data \((X)\) and input data \((X)\) as target data, both consisting of polymerization conditions and microstructures. The difference is that some polymerization conditions in the corrupted input data were randomly set to zero. This corrupted input data is then fed into the model to reconstruct the data \( (X') \) similar to input data.
3. Results and discussion

3.1 Model A: DAE model with only microstructural distributions
The model was developed to determine polymerization conditions ([Et], [Bu], [H₂], [A], and T) to produce polymers with desired MWD and CCD. The model consists of 7 layers with a topology of 45-40-30-20-30-40-45 (i.e., 45 neurons in the input layer; 40, 30, 20, 30, and 40 neurons in the subsequent hidden layers; and 45 neurons in the output layer). 45 neurons in the input and output layers consist of (1) 15 neurons for MWD, (2) 15 neurons for CCD, and (3) 5 neurons for conditions. Figure 2 and 3 showed the comparison between polymerization conditions obtained from kinetic model and the model A in the form of diagonal plots for training and testing datasets, respectively. The results showed that the predictions are close to diagonal lines, which indicates good estimation.

![Figure 2](image1.png)  
**Figure 2.** Comparison between polymerization conditions obtained from the polymerization kinetic model and from the model A for the training dataset (MSE = 0.000351).

![Figure 3](image2.png)  
**Figure 3.** Comparison between polymerization conditions obtained from the polymerization kinetic model and from the model A for the testing dataset (MSE = 0.000667).
3.2 Model B: DAE model with microstructural distributions, average microstructures, and yield

This model determined polymerization conditions ([Et], [Bu], [H_2], [A], T, and C^in) for specified microstructural distributions, average microstructures, and polymer yield. The model is for the case when controls of average microstructures and polymer yield are critical. Note that one additional condition, a total molar flow rate of catalyst C^in, is added in the network. This model also consists of 7 layers with a topology of 50-40-30-20-30-40-50 (i.e., 50 neurons in the input layer; 40, 30, 20, 30, and 40 neurons in the subsequent hidden layers; and 50 neurons in the output layer). 5 additional neurons in the input and output layers are for M_n, M_w, CC, Y, and C^in. Figure 4 and 5 showed the diagonal plots comparing between polymerization conditions obtained from kinetic model and DAE model for training and testing simulation dataset, respectively. The results again showed the good prediction for both training and testing datasets.

Figure 4. Comparison between polymerization conditions obtained from the polymerization kinetic model and from the model B for the training dataset (MSE = 0.000266).

Figure 5. Comparison between polymerization conditions obtained from the polymerization kinetic model and from the model B for the testing dataset (MSE = 0.000299).
The value of mean square error of Model B was found to be relatively lower compared to Model A. Therefore, additional constraints introduced from specified microstructures and yield help provide more accurate estimation of polymerization conditions.

4. Conclusion
In this work, two denoising autoencoder models were developed to determine appropriate polymerization conditions to produce polymers with desired microstructures. Model A used only MWD and CCD, while model B specified additional information on average microstructures and polymer yield. Both models estimated polymerization conditions close to those used in the kinetic model with acceptable MSE values. Model B was found to provide slightly better estimation of polymerization conditions with lower MSE.

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