Determination of Operating Conditions of Ethylene/1-Octene Copolymerization Using Artificial Neural Network (ANN)

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Summary: An artificial neural network (ANN) is applied to determine appropriate parameters in copolymerization of ethylene and 1-octene via metallocene catalytic system for producing a copolymer with desired chain microstructures. The polymerization parameters of interests are polymerization temperature, ethylene pressure, and the amount of hydrogen used. The ANN used is a feed-forward network with a back propagation learning method and has a 5-6-6-3 architecture. When comparing with both training and testing experimental data sets, it was found that ANN can provide a good guesstimation of polymerization parameters.

Keywords: artificial neural network; copolymerization; polyethylene (PE)

Introduction

Ethylene/1-olefin copolymer is a versatile commodity polymer widely used because its properties can be tuned over a wide range by controlling the chain microstructures (i.e., average molecular weight, molecular weight distribution, average comonomer content, and chemical composition distribution). Metallocene catalytic system is an efficient process in producing such a polymer with well-defined chain microstructures.[1] The mathematical model of such a copolymerization system to determine the chain microstructures for a given set of operating conditions is quite complex but still available.[2] The practical inverse problem of finding appropriate operating conditions to obtain a copolymer with desired chain microstructures is an extremely complex problem. Typically, this problem may be solved via a number of trial experiments. Insights from the computational work could provide the guidance and help reduce unnecessary number of experiments.

Artificial neural network (ANN) is a branch of artificial intelligence (AI) that is designed to mimic the way biological nervous systems (e.g., brain) process information. It is composed of a large number of highly interconnected processing elements (i.e., neurons) working in union to solve specific problem.[3] Like people, ANN learns from examples. While learning, ANN will develop an internal model representing the real process model. Thus, it is capable of solving highly non-linear complex problems without prior knowledge on detailed mathematical model of the system by capturing relationships between input and output variables from a given patterns. Recently, ANN has been widely used to solve various specific problems in polymer engineering,[4–8] especially when the phenomenological models are extremely complex, if at all possible, to develop.

In this work, ANN is applied to estimate three copolymerization parameters (i.e., polymerization temperature, ethylene pressure, and amount of hydrogen used) for producing ethylene/1-octene copolymers with desired chain microstructures (i.e., number average molecular weight and

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1-octene incorporation) under specified conditions (i.e., catalyst activity, 1-octene/ethylene ratio, and activity/support ratio). The ANN is trained and tested using experimental data sets reported earlier by Li Pi Shan et al.[9]

Artificial Neural Network

Artificial neural network (ANN) is a data driven black-box model for solving complex problems by capturing relationships between input and output variables. ANN learns from examples (i.e., an archive of data sets) by finding the optimal network-connection-weights that gives an output vector close to a target vector for a specific input vector. These optimal weights, which can be found by minimizing the error function, are the critical information storing the relationships between input and output variables.

Four-layer ANN model with a 5-6-6-3 architecture (i.e., five input neurons, two hidden layers each with six neurons, and three output neurons) was used in this study (see Figure 1). This ANN is a feed-forward neural network with back propagation training algorithm.

The training was carried out using 26 data sets from Li Pi Shan *et al.*,[9] who thoroughly investigated the ethylene/1-octene copolymerization with *in situ* supported metallocene catalysts. The other 5 data sets from the same work were used for testing. Each data set consists of 8 variables, three of which are considered ANN output variables (i.e., polymerization temperature, ethylene pressure, amounts of hydrogen used) and the other five of which are ANN input variables (i.e., number average molecular weight, 1-octene incorporation, catalyst activity, 1-octene/ethylene ratio, and activity/support ratio). Table 1 and 2 summarize the information in training and testing data sets.

To minimize the effect of differences in parameter magnitude, the reduced variables were used for training and testing ANN. All variables are reduced to the dimensionless variables within a range of 0–100 using the following relationship: $X_{i,\ reduced} = (X_i - X_{i,\ min})/(X_{i,\ max} - X_{i,\ min})^* 100$. $X_{i,\ reduced}$ is a reduced variable, X_i is an original variable, $X_{i,\ min}$ is the minimum value of variable X_i, and $X_{i,\ max}$ is the maximum value of variable X_i.

Prior to the training, the weights were given a random value between -1 and 1. During the training, the difference between the target outputs (from archival data) and the calculated model outputs of the training data set is minimized by adjusting the weights through back propagation algo-

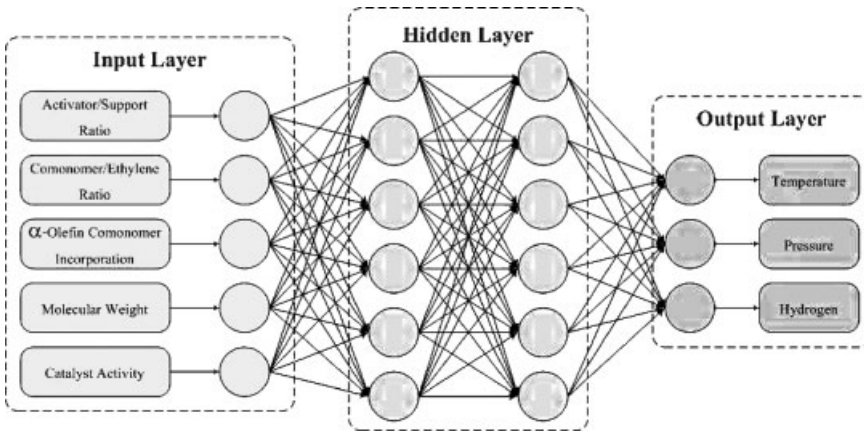

Figure 1.
ANN with a 5-6-6-3 architecture used in this study.

Table 1.
Training data sets.

Temperature (°C)	Pressure (psig)	Hydrogen (mL)	Comonomer/ ethylene ratio	Activator/ support ratio	Activity (kg of PE/mol of cat-hr)	% 1-octene content	M_N (g/mol)
40	100	0	0.07	15.0000	2,107	0.41	354,900
70	100	0	0.07	5.3350	8,448	1.00	114,000
40	200	0	0.07	5.3350	6,713	0.29	373,200
70	200	0	0.07	15.0000	10,880	0.87	213,800
40	100	50	0.07	5.3350	392	2.21	26,000
70	100	50	0.07	15.0000	1,589	2.11	33,600
40	200	50	0.07	15.0000	2,381	1.58	41,800
70	100	0	0.21	15.0000	5,440	3.98	57,500
40	200	0	0.21	15.0000	10,880	3.02	427,600
70	200	0	0.21	5.3350	26,560	3.50	463,800
40	100	50	0.21	15.0000	1,387	7.06	32,400
40	200	50	0.21	5.3350	6,320	2.78	46,600
70	200	50	0.21	15.0000	5,520	3.93	64,100
55	150	25	0.14	10.1675	5,216	1.91	67,100
55	150	25	0.14	10.1675	5,824	1.59	64,300
25	150	25	0.14	10.1675	1,296	1.29	13,800
85	150	25	0.14	10.1675	26,560	2.30	104,500
55	50	25	0.14	10.1675	427	3.88	32,700
55	200	25	0.14	10.1675	6,640	1.86	88,000
55	150	0	0.14	10.1675	15,160	1.74	280,900
55	150	75	0.14	10.1675	1,156	2.80	105,100
55	150	25	0	10.1675	4,267	0.36	63,500
55	150	25	0.28	10.1675	13,200	6.64	95,000
55	150	25	0.14	0.5025	42,480	2.35	204,300
55	150	25	0.14	19.8325	1,680	1.15	63,100
55	150	25	0.14	10.1675	5,200	1.23	71,800
40	100	0	0.07	15.0000	2,107	0.41	354,900
70	100	0	0.07	5.3350	8,448	1.00	114,000
40	200	0	0.07	5.3350	6,713	0.29	373,200
70	200	0	0.07	15.0000	10,880	0.87	213,800
40	100	50	0.07	5.3350	392	2.21	26,000
70	100	50	0.07	15.0000	1,589	2.11	33,600
40	200	50	0.07	15.0000	2,381	1.58	41,800
70	100	0	0.21	15.0000	5,440	3.98	57,500
40	200	0	0.21	15.0000	10,880	3.02	427,600
70	200	0	0.21	5.3350	26,560	3.50	463,800
40	100	50	0.21	15.0000	1,387	7.06	32,400
40	200	50	0.21	5.3350	6,320	2.78	46,600

rithm.[3] In this work, a sigmoid function ($f(x) = 1/(1 - \exp(x))$) is used as an activation function during the training. The objective function is defined as the average error between the target output vector and the calculated model output vector of both training and testing data sets. The objective function at each training epoch is monitored to help avoid overtraining of the network.

Table 2.
Testing data sets.

Temperature (°C)	Pressure (psig)	Hydrogen (mL)	Comonomer/ ethylene ratio	Activator/ support ratio	Activity (kg of PE/mol of cat-hr)	% 1-octene content	M_N (g/mol)
70	200	50	0.07	5.3350	5,040	0.93	49,300
40	100	0	0.21	5.3350	15,387	5.71	153,200
70	100	50	0.21	5.3350	3,000	5.89	47,000
55	150	25	0.14	10.1675	3,740	1.79	76,500
55	150	25	0.14	10.1675	4,640	1.77	95,800

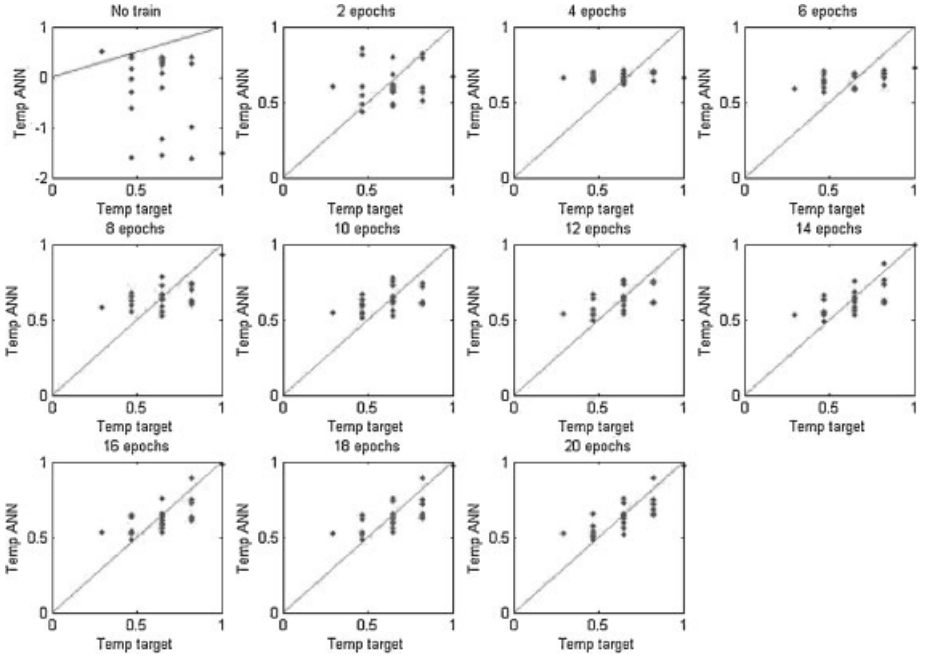

Figure 2.
Comparison between reduced temperature predicted from ANN and reduced temperature obtained from experiment for training data sets at various training epochs.

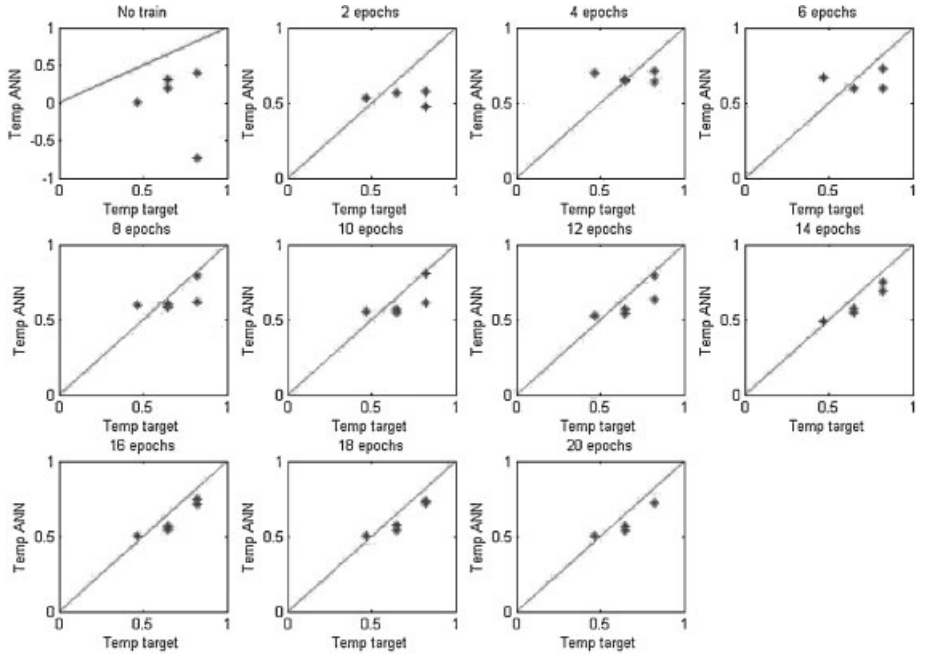

Figure 3.
Comparison between reduced temperature predicted from ANN and reduced temperature obtained from experiment for testing data sets at various training epochs.

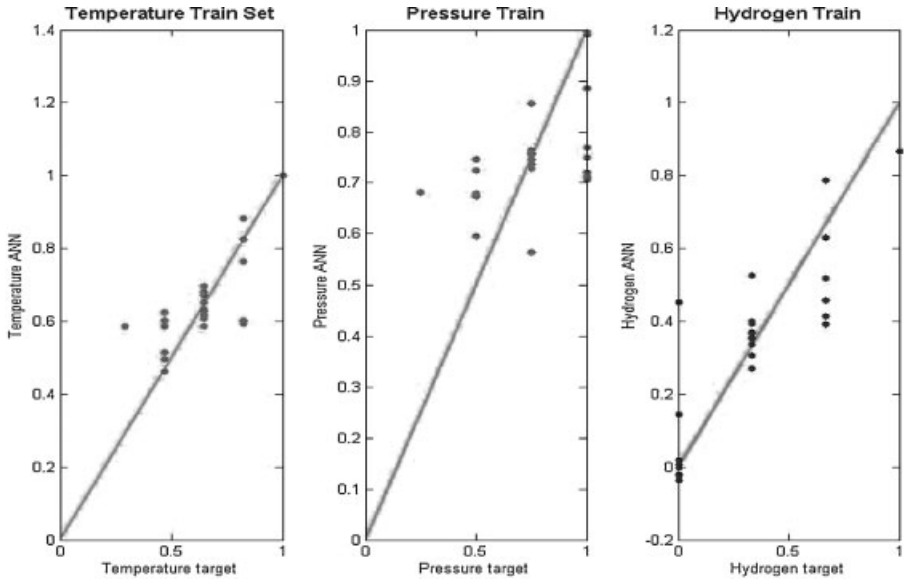

Figure 4.
Comparison between reduced parameters predicted from ANN and reduced parameters obtained from experiments.

Results and Discussion

Figure 2 plots between reduced temperatures obtained from ANN and reduced temperatures obtained from experiment for the training data sets. The scattering of data that is close to the diagonal line indicates good estimation of parameters (i.e., the predicted parameters are close to the experimental data). It can be clearly observed that as the training epoch increases, the ANN model can predict more accurately.

Figure 3 shows similar plot for the testing data sets. Note that these data sets had not been used for training so the ability of ANN to predict parameters simply comes from learning through the training data sets.

Figure 4 shows the comparison between the target output parameters and the ANN predicted output parameters at the optimal training epoch, which gives the minimum average error between the target output vector and the calculated model output vector of both training and testing data sets. Most data are found to scatter near the diagonal lines for all polymerization parameters. Some discrepancies can still be observed, however. This is because for such a complex ANN used in this study a larger number of data sets might be required to train ANN adequately. Typically, ANN has potential to be improved if a number of training sets (i.e., archival experimental data) could be expanded. The results from the model are still useful, however, for providing a good guesstimation that can drastically reduce the number of unnecessary experiments to determine the desired polymerization conditions.

Conclusion

Artificial neural network (ANN) is applied to determine the copolymerization parameters for producing ethylene/1-octene copolymers with desired chain microstructures under specified conditions. It was found that the ANN predicted parameters are close to the experimental data for both

training and testing data set. Thus, the proposed approach can be efficiently used to provide a good guesstimation of copolymerization parameters.

[1] A. E. Hamielec, J. B. P. Soares, *Prog. Polym. Sci.* **1996**, *21*, 651.
[2] J. B. P. Soares, A. Penlidis, in: "*Preparation, Properties & Technology of Metallocene-Based Polyolefins*", J. Scheirs, W. Kamisnky, Eds., John Wiley & Sons, Chichester **2000**, p. 237–267.
[3] N. Sivanandam, S. Sumathai, S. N. Deepa, "*Introduction to Neural Networks using MATLAB 6.0*", McGraw-Hill, New Delhi **2006**.
[4] E. Demirhan, F. Kandermirli, M. Kandermirli, V. Kovalishyn, *Mater. Des.* **2007**, *28*, 1737.
[5] Z. Jiang, Z. Zhang, K. Friedrich, *Compos. Sci. Technol.* **2007**, *67*, 168.
[6] A. T. Seyhan, G. Tayfur, M. Karakurt, M. Tanoglu, *Comput. Mater. Sci.* **2005**, *34*, 99.
[7] G. Padmavathi, M. G. Mandan, S. P. Mitra, K. K. Chaudhuri, *Comput. Chem. Eng.* **2005**, *29*, 1677.
[8] H. Ni, D. Hunkeler, *Polymer.* **1997**, *38*, 667.
[9] C. Li Pi Shan, J. B. P. Soares, A. Penlidis, *J. Polym. Sci. Polym. Chem.* **2002**, *40*, 4426.

