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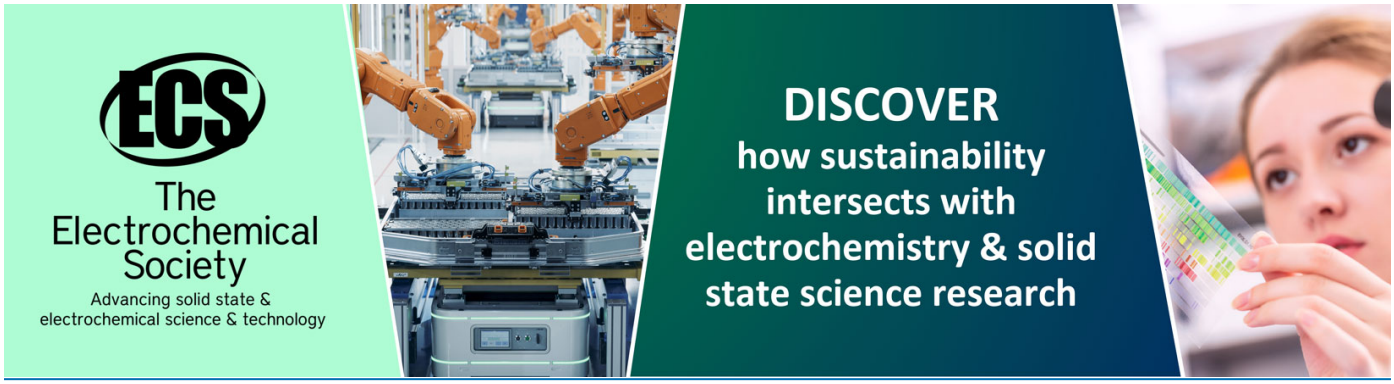

IOP Conf. Series: Materials Science and Engineering **358** (2018) 012009 doi:10.1088/1757-899X/358/1/012009

Simulation of CO_2 Solubility in Polystyrene-b-Polybutadiene-b-Polystyrene (SEBS) by artificial intelligence network (ANN) method

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Abstract. This study reports on the integration of Artificial Neural Network (ANNs) with experimental data in predicting the solubility of carbon dioxide (CO_2) blowing agent in SEBS by generating highest possible value for Regression coefficient (R^2). Basically, foaming of thermoplastic elastomer with CO_2 is highly affected by the CO_2 solubility. The ability of ANN in predicting interpolated data of CO_2 solubility was investigated by comparing training results via different method of network training. Regards to the final prediction result for CO_2 solubility by ANN, the prediction trend (output generate) was corroborated with the experimental results. The obtained result of different method of training showed the trend of output generated by Gradient Descent with Momentum & Adaptive LR (traingdx) required longer training time and required more accurate input to produce better output with final Regression Value of 0.88. However, it goes vice versa with Levenberg-Marquardt (trainlm) technique as it produced better output in quick detention time with final Regression Value of 0.91.

1. Introduction

Rubber and elastomer foams have a wide variety of applications like in transportations, packaging, construction and building insulation. Thermoplastic elastomers, often called thermoplastic rubbers are consist of a soft midblock and hard end-block which behaves both thermoplastic and elastic properties. SEBS is one type of thermoplastic elastomer that has captured attention in recent years due to its unique characteristics compared to natural rubber. The advantage of synthetic rubber is it has good resistance in temperature and oil. Nowadays, thermoplastic elastomer is being foamed to prepare light weight polymeric material which possesses many applications such as thermal insulation, shock absorber, automotive, aerospace and biomedical. However, there are many factors to be controlled like processing conditions and its physical properties in order to prepare fine cellular structure of thermoplastic elastomer foam. According to Rahida et al. [1] stable dimension of SEBS foam can be prepared when optimum rigidity, CO_2 solubility and diffusivity were achieved. They claimed that controlling diffusion of CO_2 through SEBS during foaming is of great importance. In their study they found that the shrinkage problem in elastomer foaming can be controlled by increasing the hard-polystyrene end-blocks or blending with thermoplastic polystyrene. This is because greater styrene contents increased the storage modulus and reduced the CO_2 solubility and diffusivity which then led to stabilized elastomer cell foam. In recent years, most sophisticated magnetic suspension balance machine was used to measure the CO_2 solubility and diffusivity in polymers. This experimental

technique has made possible to measure CO_2 solubility and to estimate diffusion coefficient in any kind of polymers at various temperature and pressure. Some studies have been done on solubility measurement of different gases in polymers. However, this method is highly expensive, time consuming, and highly affected by pressure-volume-temperature (PVT) data of polymers. Due to these limitations, there are some efforts to predict the CO_2 solubility and diffusivity in polymers by intelligence methods as reported elsewhere. Golzar et. al. [2] for example, predicted the solubility of CO_2 and nitrogen (N_2) in common polymers via linear and non-linear models. They reported on structure-properties relationship via intelligence methods like ANN, ANFIS and GA by using set of solubility data for CO_2 and N_2 in different polymers from literatures.

This study reported on the ability of ANN to predict combination of experimental and interpolated data of CO_2 solubility and diffusivity in SEBS in the temperature range of 61.76 to 101.9°C and pressure range of 5 to 20 MPa without repeating such costly and time consuming experimental measurement by MSB. In addition, to the best authors' knowledge, no study has been done on verification of interpolated experimental data of CO_2 solubility in SEBS by ANN. ANN has become one of the most promising techniques in dealing with complex and non-linear problem relations that cannot be solved by simple electronic calculator. The operation of the ANN is likely the operation of the nervous system containing neurons that functional to solve engineering- based problem, scientific or no-scientific based problem. For example Taskinen et al.[3] predicted physicochemical properties of organic compounds which focusing on pharmaceutical research and Fatehi et al. [4] studied on estimation of viscosities of pure ionic liquids using an artificial neural network based on only structural characteristics. In addition, the ability of ANN in processing the complex system even in critical condition makes the ANN as one of the extremely powerful methods for prediction process. Therefore, the intention of this study is to predict and further validate the interpolated data that might be useful for reducing the need of repeating MSB measurement as well as facilitating the control of SEBS cell stabilization. The idea behind is to predict the optimum CO_2 solubility at varying temperature and pressure using ANN that could possibly control the foam shrinkage in SEBS.

2. Methodology

2.1 Data input

Solubility of CO_2 in SEBS data was collected from previous MSB measurement in the temperature range of 61.76 to 101.9°C and the pressure range of 5 to 20 MPa. The selected data were divided into three sections which are the most concerned part in making a perfect ANN model. Randomly, those data were separated according to the following ratio 70% used for training and learning session, and 30% will equally divided into two parts which are validation purpose and for test sets. According to Huang et al.[5], the appropriate distribution of data set is very important in order to obtain the accurate results. Besides, the percentage for test set was in the range of 5 to 35%.

2.2 Neural Network Structure Used

There is various type of neural network structures can be used for prediction. In this study, Multilayer Perceptrons (MLP) with a basic backpropagation training and universal theorem was used as it could work with limited sources of inputs. This network is the most recommended ANNs to handle a simple task. It is classified as feedforward network that could approximate generic functions (continuous and integrable). In this study, feedforward backpropagation (FFBP) was chosen to be used in predicting the CO_2 solubility in SEBS. In feed forward neural network, the process happens in forward direction and the neuron is connected from input to output. The three layers must be connected with each other in order for the process to be done. For feed forward neural network, the connection of the layer is from input to hidden and lastly to output. The direction cannot be reversed to the opposite direction and it is so called as feedforward backpropagation.

After the training, new output data is obtained and further been compared with the actual output. The output data that was keyed in at the starting of the training is known as actual output while the new output is known as expected output data. The adjustment on the data from output to hidden followed by input was made by utilizing the backward propagation together with feedforward for obtaining better result. In this study, the most effective and suitable method of feedforward

backpropagation method was used because the variables in this study; temperature, pressure and CO_2 solubility have no linear relationship between one another.

2.3 Artificial Neural Network Training

The most important part in using ANNs is to find the almost perfect set of weight, *w* parameters. To achieve that, abundant training is required. In the training process, theoretically the output from neural model is compared with the desire outputs for all of the training samples using the following formula where the error could be defined.

$$E = \frac{1}{2}\sum_{k\in T_r}\sum_{j=1}^{m}(y_j(x_k, w) - d_{jk})^2 \quad (1)$$

Where,

d_{jk} = j^{th} element of d_k
$y_j(x_k, w)$ = j^{th} neural network output for input x_k
T_r = Index set of training data

Foremost, the value of weight is initialized. During training session, *w* is updated with the following formula,

$$= w - n\frac{\partial E}{\partial w},$$

until the value of *E* is small enough. Value of *n* is called learning rate. Per-sample of E_k is,

$$E_k = \frac{1}{2}\sum_{j=1}^{m}(y_j(x_k, w) - d_{jk})^2 \quad (2)$$

The E_k value is used and been updated as

$$= w - n\frac{\partial E_k}{\partial w}.$$

Next, the value of $\frac{\partial E_k}{\partial w}$ is computed by using basic differentiation,

$$\frac{\partial E_k}{\partial w_{ij}^l} = \frac{\partial E_k}{\partial z_i^l}.\frac{\partial z_i^l}{\partial w_{ij}^l} \quad (3)$$

And,

$$\frac{\partial z_i^l}{\partial w_{ij}^l} = \frac{\partial \sigma}{\partial \gamma_i^l}.z_j^{l-1} \quad (4)$$

The gradient $\frac{\partial E_k}{\partial z_i^l}$ can be initialized at output layer as,

$$\frac{\partial E_k}{\partial z_i^L} = (y_i(x_k, w) - d_{ik}) \quad (5)$$

The error is then calculated between both outputs from ANNs and the desired outputs. For a network that using sigmoid equation, the back propagation process is then give by the following formula:

$$\delta_i^L = (y_i(x_k, w) - d_{ik}) \quad (6)$$

$$\delta_i^l = \sum_{j=1}^{N_{l+1}}\left(\delta_j^{l+1}w_{ji}^{l+1}\right)z_i^l(1 - z_i^l), l = L-1, L-2, \ldots 2 \quad (7)$$

And the derivative with respect to weights is,

$$\frac{\partial E_k}{\partial w_{ij}^l} = \delta_i^l z_j^{l-1} l = L, L-1, \ldots, 2 \tag{8}$$

3. Results and Discussion

3.1. Comparison of CO_2 solubility interpolated data between trainlm and traingdx methods

After interpolating solubity result at various ranges of temperature and pressure, a set of CO_2 solubity data was divided into three sections for training and learning by ANN model. The input data was then normalized in the range between 0 and 1. The biggest value of solubility from experimental data is marked as 1 while the smallest value is noted as 0. For the purpose of verifying the ability of ANN to predict the interpolated data, two types of training functions; trainlm and traingdx are employed. The data was imported to ANN database to run the simulation process. Figure 1(a) and (b) shows the training results from trainlm and traingdx networks, respectively. The prediction values are clearly corroborated with experimental values for both networks where it can be seen that the predicted values of training set, validation set and testing set are closed to the experimental value line. However, the predicted value for trainlm is more intense and closer than traingdx. This result is further supported by the values of squared correlation coefficients (R^2) for trainlm and traingdx of 0.911 and 0.886, respectively. The R^2 values for both networks verified are very close to the unity and it shows that ANN model can be used for predicting interpolated CO_2 solubility data. However, trainlm has higher ability to predict CO_2 solubility because the value of R^2 is higher than traingdx.

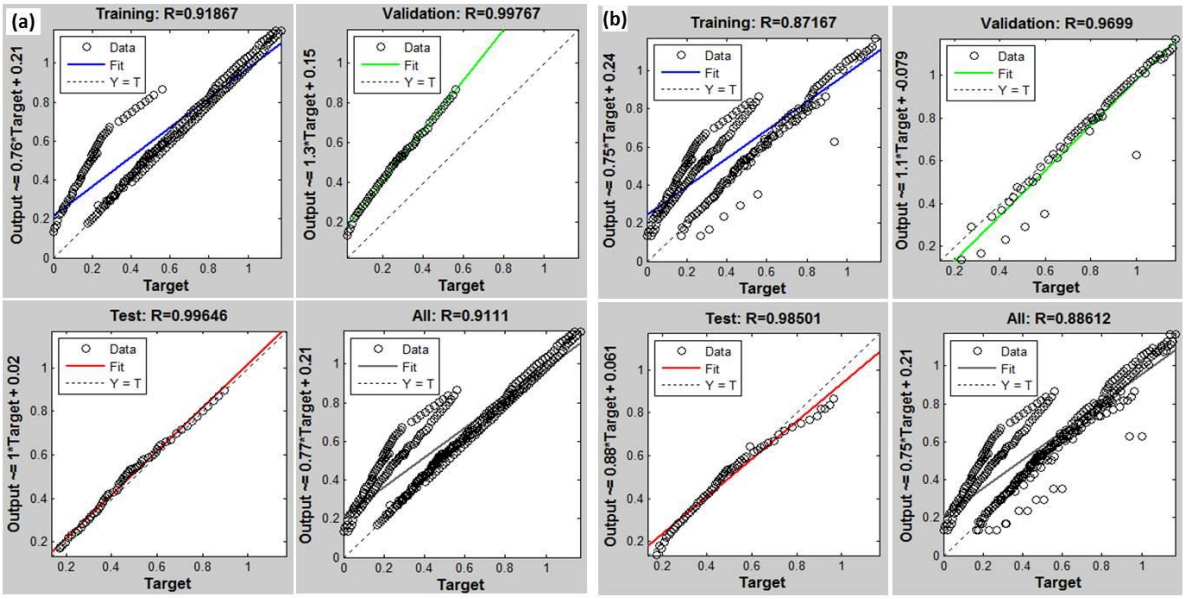

Figure 1. Training results; (a) trainlm technique and (b) traingdx technique

3.2. Error Yield between Input Data and Output Generated

The reliability of ANN model with trainlm and traingdx transfer functions was further examined by measuring the variance between predicted and experimental data. The CO_2 solublity data was trained at fix temperature and varying the pressure to show the trend of trained output (predicted) with the input (combination result of experimental and interpolated data) as a reference in a plot form. Figure 2 and 3 show the error plot obtained from the differences of solubility between input and output by trainlm and traingdx, respectively. For traingdx, at temperature of 61.76 and 62.18°C, the trend was appeared unsmoothed due to some data fluctuated and deviated from the input data.

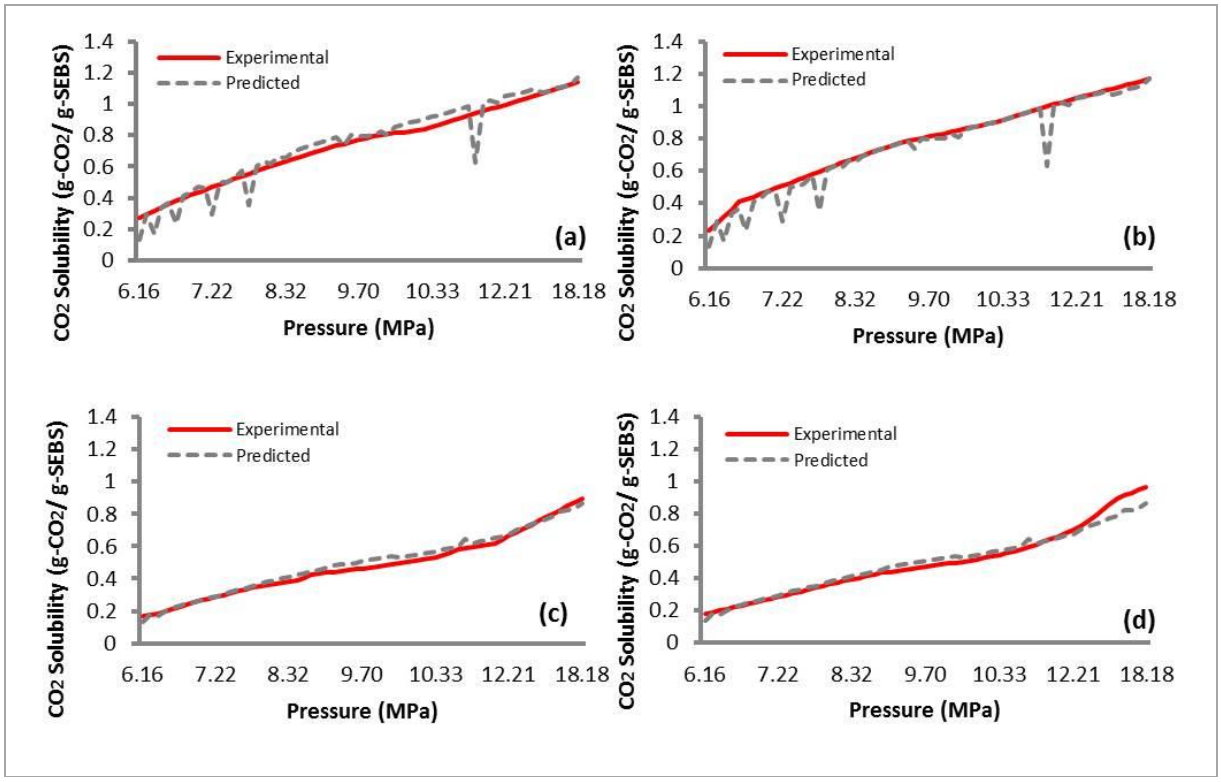

Figure 2. Error yield for traingdx at various temperatures; (a) 61.76°C, (b) 62.18°C, (c) 101.5°C, and (d) 101.9°C

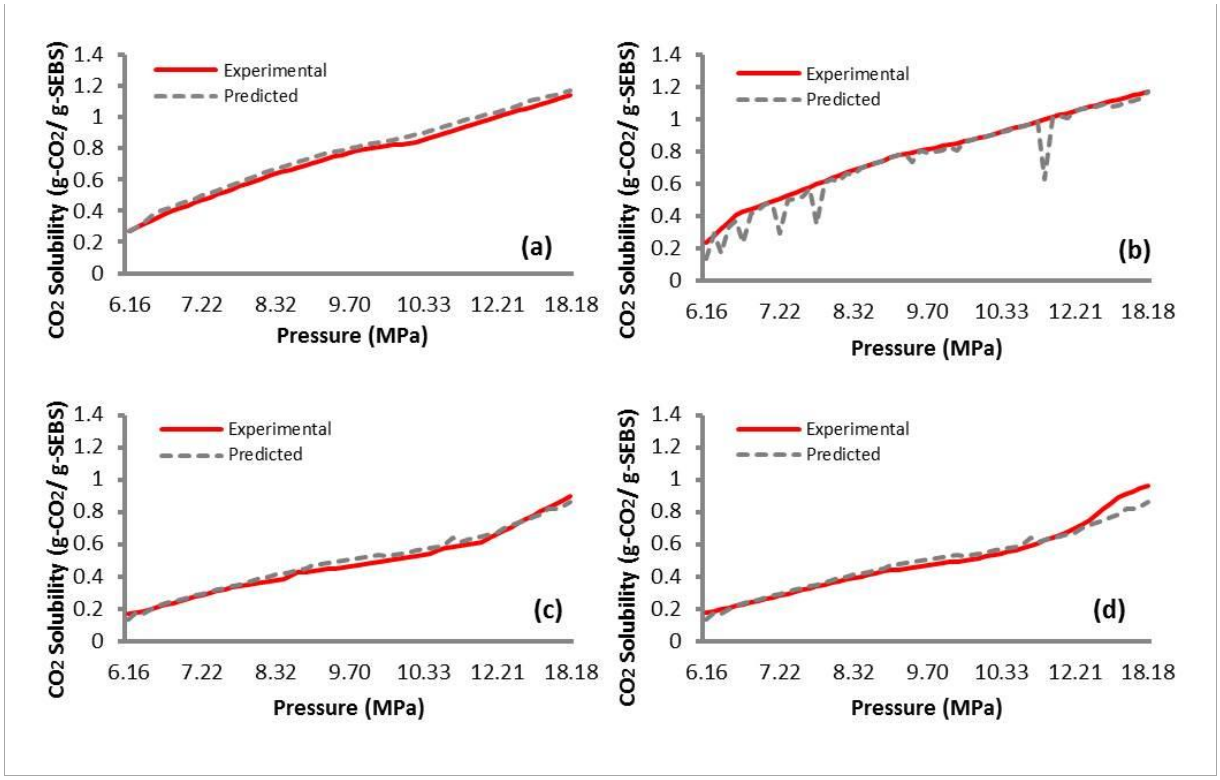

Figure 3. Error yield for trainlm at various temperatures; (a) 61.76°C, (b) 62.18°C, (c) 101.5°C, and (d) 101.9°C

However, at 101.5°C the trend became smooth and corroborated with the input. It shows that the model is reliable and able to train the interpolated data at 101.9°C by traingdx. The similar trend was observed for the predicted data at temperature of 101.9°C which is indicated the reliability of network to predict. For trainlm prediction at similar temperature and pressure, as expected the trends look better at 61.76°C compared to traingdx. It was observed that data fluctuation was reduced and the predicted data line was near to the experimental one, which related to the decreased of the error yield. The smaller gap between predicted and experimental lines by trainlm indicated higher accuracy of prediction is achieved, and thus confirm the ability of ANN model to predict a set of interpolated data. As the function of trainlm algorithm is usually considered for optimizing data, the trend obtained therefore smoother than that of traingdx. Furthermore, as the traingdx algorithm is very sensitive to the learning rate settings, it can oscillate during training and thus the output data became unstable.

3.3. Renormalized Output Data

The presence of large errors in both transfer functions at high temperatures, each of the solubility values were renormalized and tabulated in standard solubility range. By using the value of error generated from the training, two graphs were plotted to show the true pattern of CO_2 solubility predicted by ANNs as depicted in Figure 4. It was observed that the data generated by traingdx was not stable at some temperatures as it might be due to traingdx transfer function required more training as compared to trainlm. In addition, traingdx is quite sensitive to the learning rate setting where when the learning rate is too high, the algorithm might oscillate and became unstable. Whereas the trend obtained by trainlm algorithm appeared smooth and stable as it is usually serves for optimization process. Obviously, this method cannot be used in order to generate output with limited parameter and input data. In contrast, Figure 4(b) clearly shown that the generated pattern by trainlm was smooth and in agreement with the experimental data flow pattern. However, more training is required to produce better R^2 hence, give the best solubility prediction data with lower error.

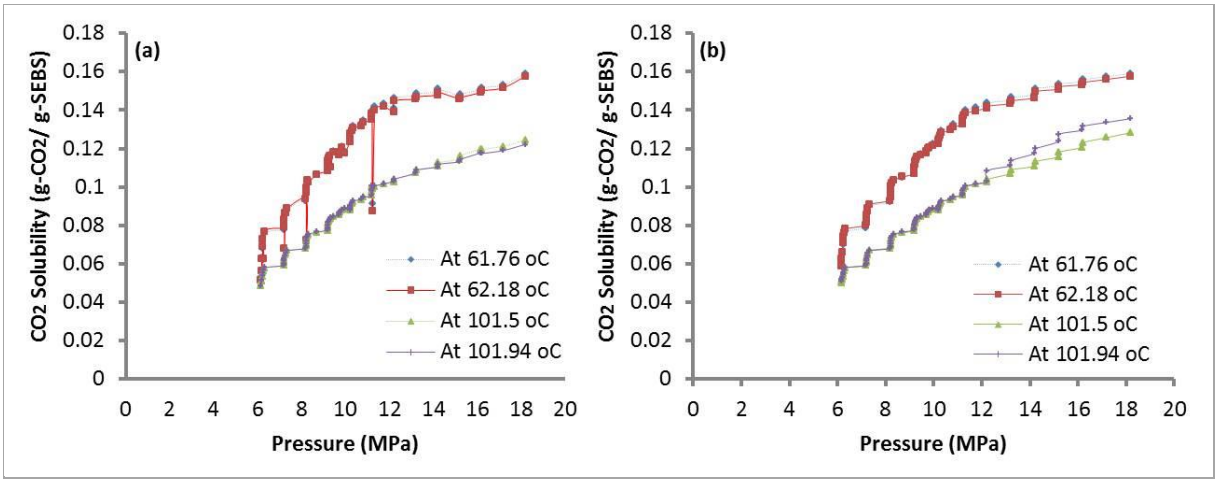

Figure 4. Error yield at various temperatures for technique; (a) traingdx and (b) trainlm.

4. Conclusion

CO_2 solubility control in elastomer foam is vital in producing high performance insulator. Stable dimension with uniformly distributed pores in microcellular elastomer foam promises good heat transfer and perhaps reduce heat loss from insulated system. Thus, the appropriate level of CO_2 solubility needs to be control to help in retaining the stable-shape foam dimension. This can be achieved by measuring the optimum CO_2 solubility prior to foaming process. In this study, the prediction of optimum CO_2 solubility in SEBS by ANN method is proposed as an alternative for direct high cost and time consuming MSB measurement. The simulation results showed the reliability of ANN model with trainlm transfer function to simulate the interpolated CO_2 solubility data from the experimental one. The validated simulation results of R^2 close to the unity reveals the trainlm transfer function is applicable for prediction of interpolated solubility data at temperature range of 61.76 to 101.9°C at various pressure compared to traingdx transfer function.

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