Vol. 4(2), pp. 1-11, December 2025
eISSN: 2976-4009

ASEAN Journal of Process Control

Research Article

Comparative Study between Cascade-Forward Neural Network and Feedforward Neural Network Models for Enzymatic Polymerization Process

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Abstract: Enzymatic polymerization has gained attention as a more environmentally friendly alternative to conventional chemical methods, as it avoids the use of fossil fuel-derived monomers and requires milder operating conditions. However, the reaction kinetics and mechanism of the polymerization are complex. The process is also sensitive to the operating conditions, as harsh operating conditions would negatively impact the desired molecular weight of the resulting polymer. To address this, the process model must be developed accurately to predict the molecular weight under suitable conditions. This study compares the effectiveness of two types of artificial neural networks (ANN), namely cascade-forward neural network (CFNN) and feedforward neural network (FFNN) to determine the better-performing ANN type for the enzymatic polymerization process. A total of 84 experimental datasets obtained from laboratory batch study were used, which include the inputs of reaction time, reaction temperature and reactor impeller speed, as well as polymer molecular weight as the output. The effects of these operating inputs on the outputs were also investigated through sensitivity analysis. Twelve training algorithms for both CFNN and FFNN were compared and evaluated in terms of mean square error (MSE), root mean squared error (RMSE), mean absolute error (MAE), mean absolute percentage error (MAPE), regression (R), determinant coefficient (R^2), and accuracy. CFNN model with the Levenberg-Marquardt backpropagation (LM) achieved the highest accuracy of 99.97%, yielding the least MSE, RMSE, MAE, MAPE and R of 2.36, 1.53, 0.80, 0.03%, 1, respectively. The sensitivity analysis showed that both temperature and speed have a significant negative impact on the molecular weight of polymer compared to the time input.

Keywords: Artificial neural network (ANN), feedforward neural network (FFNN), cascade neural network (CFNN), enzymatic polymerization.

1. Introduction

Polymerization process is crucial in the manufacturing industries to produce a wide variety of polymers, such as polyolefins, polyesters and nylons, which can be further used to make valuable products like plastics, packaging films, medical bandages and many more items. However, conventional polymerization methods typically rely on fossil-based monomers, energy-intensive operating conditions, and hazardous catalysts and by-products. Enzymatic polymerization has emerged as a promising and more sustainable alternative to the conventional chemical polymerization methods. Instead of using toxic catalysts, it uses enzymes to synthesize polymers under mild, eco-friendly conditions [1]. Polycaprolactone (PCL) is one of the examples of an artificial polymer with a repeating unit of monomers called caprolactone. PCL is involved in many applications, including medical devices, scaffolds for tissue engineering and biodegradable implants [2].

Enzymatic polymerization has gained attention as a sustainable method, but this process has significant challenges for large-scale implementation. This is because enzymes usually have constraints in their active sites that affect their efficiency, leading to a lower performance when compared to chemical methods [3]. Additionally, the process is considered complex, such that regular optimization, like single-factor analysis, is tedious and difficult to produce results with high accuracy [4]. To overcome these challenges, powerful techniques like machine learning should be applied to evaluate and predict the relationship between the desired output and input of the enzymatic polymerization system.

Johnson et al. [5] developed a mechanistic model for the catalyzed polymerization and degradation of PCL. The results showed that the trends between experimental and the modelled molecular distributions were similar, but the molecular weights were smaller than the experimental data, which was possibly due to the assumptions of the model. Assumptions were made to simplify the mechanistic model developed, including homogenous system model and zero enzyme diffusion in chain length dependence calculations, which removed the effects of enzyme characteristics towards the polymerization process. This will affect the prediction of the actual enzymatic polymerization as these assumptions limit its predictive accuracy at different operating conditions. Therefore, empirical models such as cascade-forward neural network (CFNN) and feedforward neural network (FFNN) can potentially offer a flexible yet effective alternative for process prediction.

Recent advances in machine learning have demonstrated the strong potential of artificial neural networks in modelling enzymatic polymerization processes. Tariq and Arumugasamy [6] developed FANN model based on experimental parameters of reaction time, monomer/solvent ratio, mixing speed and reaction temperature to predict the molecular weight of poly-ε-caprolactone. Results show that Bayesian Regularization (BR) outperformed other training algorithms, such as Levenberg–Marquardt (LM), Scaled Conjugate Gradient (SCG) and Resilient Backpropagation (RP). This study demonstrated the importance of determining suitable training algorithms, the number of hidden neurons, hidden layers and transfer functions. However, this study utilized only 25 datasets. Small datasets are known to exacerbate issues such as overfitting and generalization [7]. This study highlighted the need for further investigation of other modelling techniques. Consequently, the present study aims to evaluate and compare another type of ANN besides FFNN to assess their prediction capabilities.

Mondal et al. [8] demonstrated the use of FFNN and RSM (response surface methodology) to model an enzymatic saccharification process aimed at maximizing the total reducing sugar yields. The study showed that FFNN achieved better predictive performance than RSM, as indicated by a higher coefficient of determination and lower relative mean error. This shows the effectiveness of ANN in capturing complex nonlinear relationships in enzyme-driven processes. Hence, ANN is highly suitable for the enzymatic polymerization modelling.

In this study, CFNN and FFNN were developed and compared to determine the best predictive performance. The data used to develop these models were from experiments carried out by Arumugasamy [9]. Further details on the experimental procedure will be discussed in the Materials and Methods section. The models were trained using a total of 12 training algorithms and evaluated based on statistical performance indicators such as mean square error (MSE), root mean squared error (RMSE), mean absolute error (MAE), mean absolute percentage error (MAPE), regression (R), determinant coefficient (R^2), and accuracy. The results of this study will contribute to identifying the most suitable neural network architecture with training algorithm for accurate molecular weight prediction and better process understanding in enzymatic polymerization systems through sensitivity analysis.

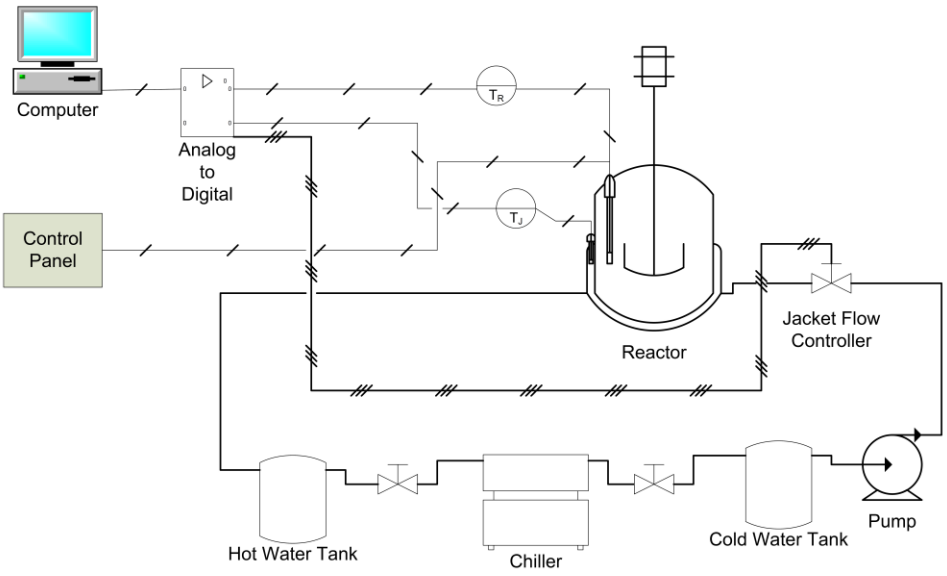
2. Materials and Methods

2.1. Polycaprolactone Production ε-Caprolactone *from in a Batch Reactor*

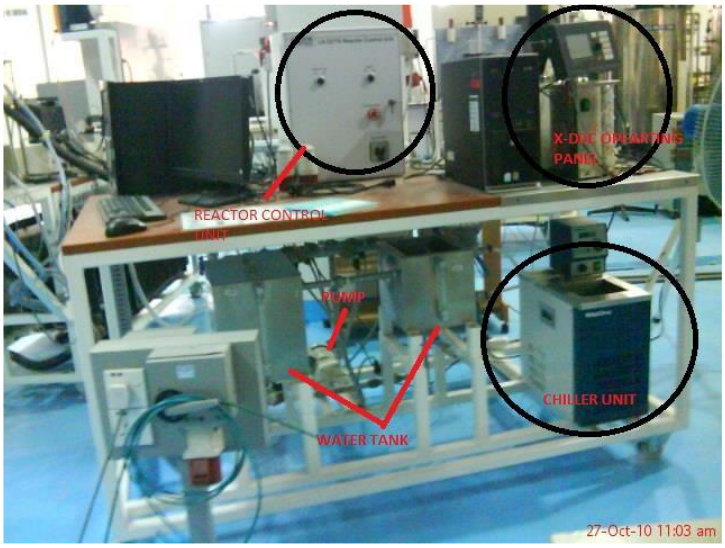
ε-caprolactone (ε-CL) was used as the monomer to produce polycaprolactone (PCL), while toluene was used as the solvent for the polymerization process to occur. Both chemicals were purchased from Merck Pte. Ltd. Candida Antarctica lipase B (CALB) enzyme was used as the reaction catalyst. Chloroform was used to stop the polymerization process. This chemical was purchased from Science Technics Pte. Ltd.

Infors-HT Labfors bioreactor was utilized to scale up the lab-based production of PCL from ε-CL in a round-bottom flask. Three parameters were studied, namely reactor temperature, reactor impeller speed, and reaction time. Experiments were conducted at different temperatures (60 °C, 70 °C, and 80 °C) and impeller speeds (250 rpm, 500 rpm, 750 rpm, and 1000 rpm) to collect data and model the polymerization process.

The rig was positioned on a dry level floor, and its 3-phase plug was connected to the laboratory 3-phase power supply. The chiller power cables were connected to the power supply located beside the control panel. The tank covers were removed, and the chiller, cold water tank and hot water tank were filled with water. The water inlet and outlet valves were opened, while the hoses of the water inlet and outlet were connected to the jacketed glass. The water flow rate to the chiller was regulated using a control valve, after which the water pump was turned on from the control panel. The chiller unit was switched on and set to the desired temperature, followed by switching on the heater and setting the heating temperature. The set-up apparatus is shown in Figure 1.



(a)



(b)

Figure 1: Bio-reactor set up (**a**) Schematic diagram; (**b**) Actual set-up in laboratory.

The reaction feed was prepared using ε-caprolactone (monomer), toluene (solvent), and CALB (enzyme), in a ratio of solvent to monomer to catalyst of 2:1:10 (v/v/wt) based on Kumar et al. (2000). The chemicals and enzyme were fed into the reactor with an effective volume of 2 litres containing the reaction feed. The experiment start time was at t = 0 when the reactor was switched on, and the desired reactor temperature was entered into the computer connected to the reactor. Once the set temperature was achieved, the stirrer was activated using the control panel and set to the desired impeller speed. Samples were collected at one-hour intervals from the start of the reaction until the 7th hour. The collected samples were analyzed using Gel Permeation Chromatography (GPC) to determine their molecular weights and Gas Chromatography (GC) to determine the residual monomer concentrations. A total of 84 experimental datasets were collected and used for modelling.

2.2. Development of FFNN and CFNN Models

2.2.1. Modelling Using FFNN

FFNN is a type of artificial neural network where the data flows in a single direction without loops or feedback. FFNN was used to model the enzymatic polymerization process by predicting the molecular weight of the PCL based on the inputs of temperature, impeller speed and time. FFNN consists of 3 neuron layers, namely the input layer, hidden layers and output layer. The FFNN model was developed using MATLAB 2025a™ version. The MATLAB "feedforwardnet" function includes built-in data normalization and denormalization procedures; therefore, external steps of data normalization and denormalization were not required. The 84 samples obtained during experiments were replicated 3 times to ensure high accuracy of the modelling. A total of 252 data sets were divided for training (70%), validation (20%) and testing (10%) using the "divideint" function in MATLAB.

Hidden layers are intermediate layers between input and output layers that enable the network to learn complex patterns of a system. Additional layers are implemented to potentially improve the model's accuracy. However, for this study, one hidden layer would suffice to obtain high accuracies of more than 90%. Hidden neurons are the processing units in the hidden layers to process the data from the input layer. The hidden neurons for the model were determined through trial and error with the highest correlation coefficient, R. The transfer functions were also determined through trial and error, which was later confirmed to be tan-sigmoid for the hidden layer, while purelin (linear transfer function) for the output layer.

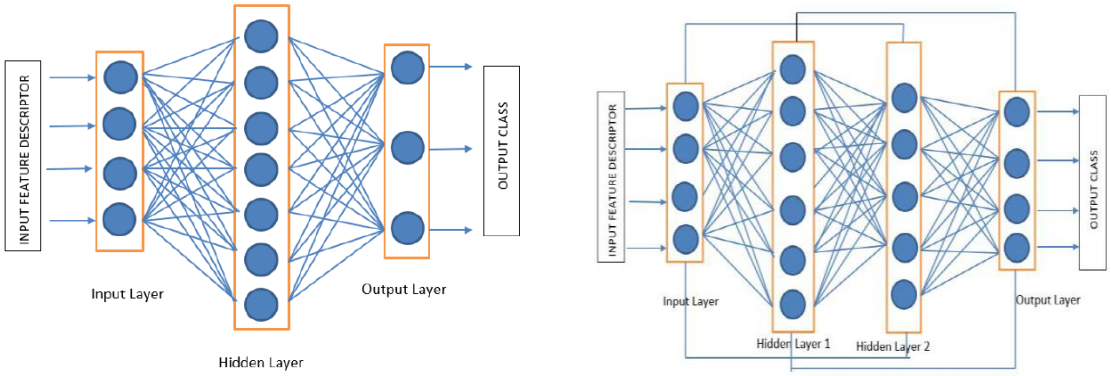
In ANN, training algorithms determine weights during training, while considering the data given and the loss function optimized [10]. In this study, 12 training algorithms, as shown in Table 1, were compared and evaluated.

Table 1. Training algorithms used in this study [11].

Acronym	Algorithm	Description
LM	trainlm	Levenberg-Marquardt
BFG	trainbfg	BFGS Quasi-Newton
RP	trainrp	Resilient Backpropagation
SCG	trainscg	Scaled Conjugate Gradient
CGB	traincgb	Conjugate Gradient with Powell/Beale Restarts
CGF	traincgf	Fletcher-Powell Conjugate Gradient
CGP	traincgp	Polak-Ribiére Conjugate Gradient
OSS	trainoss	One Step Secant
GDX	traingdx	Gradient descent with momentum and adaptive learning rate backpropagation
GDM	traingdm	Gradient descent with momentum backpropagation
GDA	traingda	Gradient descent with adaptive learning rate backpropagation
RP	trainrp	Resilient backpropagation
BR	trainbr	Bayesian regularization backpropagation

2.2.2. Modelling Using CFNN

CFNN is similar to FFNN, except that it has an extra weighted connection from the input layer to each hidden layer and from each hidden layer to the successive layer as shown in Figure 2. To ensure fair comparison between CFNN and FFNN, the parameters used in tuning both the neural network architectures are the same. The modelling procedure is summarized in Figure 3.



(**a**) (**b**)

Figure 2: Architecture of neural networks for (**a**) FFNN; (**b**) CFNN [12].

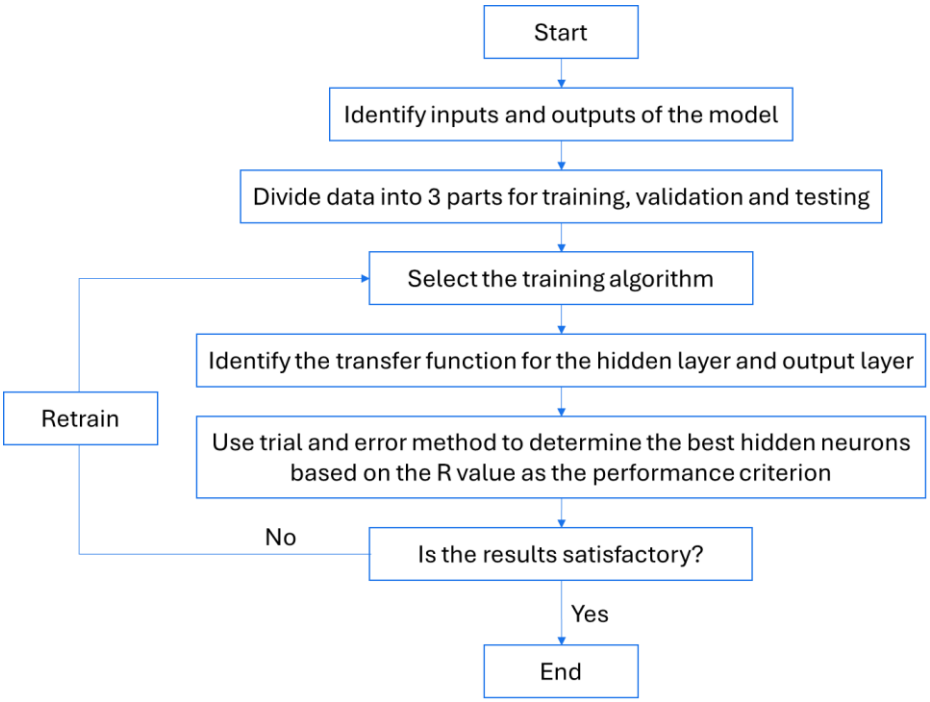


Figure 3: Flowchart of developing CFNN and FFNN models.

2.2.3. Performance Evaluation of FFNN And CFNN Models

The neural networks for FFNN and CFNN were evaluated by comparing their mean square error (MSE), root mean squared error (RMSE), mean absolute error (MAE), mean absolute percentage error (MAPE), regression (R), determinant coefficient (R^2), and accuracy. The equations to calculate the performance parameters are shown below. Note that Pt is the predicted molecular weight at time t, At is the actual molecular weight at time t, and n is the number of data points.

$$MAE = \frac{1}{n}\sum_{t=1}^{n}|P_t - A_t| \quad (1)$$

$$MAPE = \frac{1}{n}\sum_{t=1}^{n}\left|\frac{A_t - P_t}{A_t}\right| \times 100 \quad (2)$$

$$MSE = \frac{1}{n}\sum_{t=1}^{n}(P_t - A_t)^2 \quad (3)$$

$$RMSE = \frac{1}{n}\sum_{t=1}^{n}(P_t - A_t)^{0.5} \quad (4)$$

$$SMAPE = \frac{1}{n}\sum_{t=1}^{n}\frac{|A_t - P_t|}{\frac{|A_t| - |P_t|}{2}} \times 100 \quad (5)$$

$$Accuracy\ (\%) = 100 - SMAPE \quad (6)$$

Additionally, sensitivity analysis was conducted to study the impact of the temperature, impeller speed and time inputs on the molecular weight of polymer. As described by Mohammadi et al.[13], the higher the relevancy factor (RF), the higher the impact of the input towards the output. Note that x is the input of temperature, time, or impeller speed.

$$RF\ (x, P_t) = \frac{\sum_{t=1}^{n}((x - \bar{x})(P_t - \bar{P}_t))}{\sqrt{\sum_{t=1}^{n}(x - \bar{x})^2 \sum_{t=1}^{n}(P_t - \bar{P}_t)^2}} \quad (7)$$

3. Results and Discussion

3.1. Comparison of Neural Network Algorithm Results

Tables 2 and 3 summarize the performance of 12 training algorithms for CFNN and FFNN models, respectively. Among all algorithms, both CFNN and FFNN trained with LM algorithms achieved the best predictive performance. The CFNN with LM model yielded an R of 1 and R^2 of 1, with an RMSE of 1.53 and an accuracy of 99.97%. For FFNN with LM model, it achieved R of 0.9997, R^2 of 0.9995, RMSE of 8.07 and accuracy of 99.81%. These results indicate a good correlation between predicted and experimental molecular weights, suggesting that LM algorithm is effective in solving non-linear least squares problems with faster convergence characteristics [14]. Gradient descent-based algorithms such as GDM and GDA demonstrated poor results, with R values below 0.93 for both models. Notably, GDM had the lowest accuracy of less than 0%, poor R values of around 0.3, and high statistical errors, indicating its inefficiency for complex non-linear systems.

Table 2. The best performance of 12 training algorithms with optimized neurons for CFNN.

Training algorithms	Optimized neuron	R	R^2	RMSE	Accuracy (%)	MAE	MAPE (%)	MSE
LM	18	1.0000	1.0000	1.53	99.97	0.80	0.03	2.36
BFG	19	0.9592	0.9200	97.83	96.83	110.82	3.19	20260.82
SCG	19	0.9925	0.9850	85.33	98.63	31.63	1.37	1806.41
CGB	19	0.9856	0.9715	58.48	98.08	44.91	1.93	3419.43
CGF	20	0.9867	0.9735	56.63	98.38	38.77	1.62	3206.41
CGP	20	0.9843	0.9688	0.97	98.23	42.22	1.79	3757.60
OSS	19	0.9941	0.9883	37.43	98.90	24.54	1.10	1401.25
GDX	19	0.9242	0.8542	132.21	95.77	97.88	4.25	17480.02
GDM	19	0.3388	0.1148	1795.37	-25.05	2016.65	88.09	5303882.38
GDA	19	0.9195	0.8456	136.19	95.42	108.06	4.58	18546.57
RP	19	0.9933	0.9866	40.03	98.74	29.89	1.27	1602.46
BR	16	0.9234	0.8527	132.87	95.69	102.69	4.35	17653.17

Table 3. The best performance of 12 training algorithms with optimized neurons for FFNN.

Training algorithms	Optimized neuron	R	R2	RMSE	Accuracy (%)	MAE	MAPE (%)	MSE
LM	19	0.9997	0.9995	8.07	99.81	4.36	0.19	65.08
BFG	20	0.9713	0.9434	82.70	97.57	56.59	56.59	6839.03
SCG	19	0.9948	0.9896	35.28	98.95	24.08	1.05	1244.34
CGB	20	0.9967	0.9934	28.43	99.44	13.33	0.57	808.12
CGF	18	0.9590	0.9198	97.96	96.70	75.97	3.31	9597.10
CGP	18	0.9966	0.9933	28.37	99.05	21.82	0.94	804.98
OSS	19	0.9849	0.9700	59.93	97.98	45.64	2.02	3591.95
GDX	19	0.9610	0.9234	95.71	96.90	72.11	3.11	9160.97
GDM	19	0.3127	0.0978	2219.86	-3.13	1867.03	82.62	4927786.54
GDA	20	0.9136	0.8347	141.00	95.60	102.28	4.41	19879.59
RP	17	0.9963	0.9926	30.01	99.01	22.75	0.99	900.32
BR	18	0.9996	0.9991	10.32	99.86	3.46	0.15	106.43

Figures 4 and 5 show the distribution and accuracy of the predicted molecular weight using all the training algorithms except GDM in the CFNN and FFNN models, respectively. GDM is excluded because its accuracy was significantly different compared to the other algorithms, as shown in Figure 6. Overall, the accuracy for the 11 training algorithms in both models was high, with accuracy values of more than 90%. This is possible because the datasets were duplicated three times, making the data sufficient for training. FFNN models were relatively more accurate than CFNN models, despite the CFNN model with LM having the best performance when comparing the accuracy values in Tables 2 and 3. This may be due to the increased complexity in the CFNN model as its input layer is connected to more layers in the architecture network, which increases the chance of overfitting, leading to the reduction of overall accuracy [12]. GDM presented the worst results for both models; therefore, this training algorithm is unsuitable for predicting the polymerization process. This is reflected in the literature, as its low accuracy may be attributed to the inability to determine the molecular weight trend due to neglected error surface and gradient difference update [15].

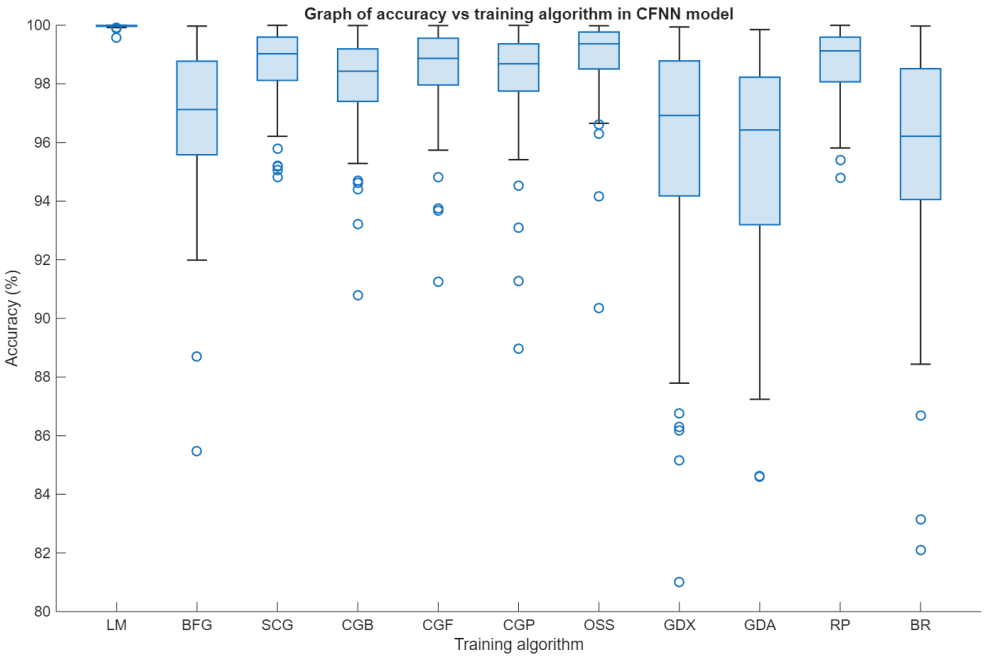


Figure 4: Boxplot of accuracy versus the 11 training algorithms in CFNN model.

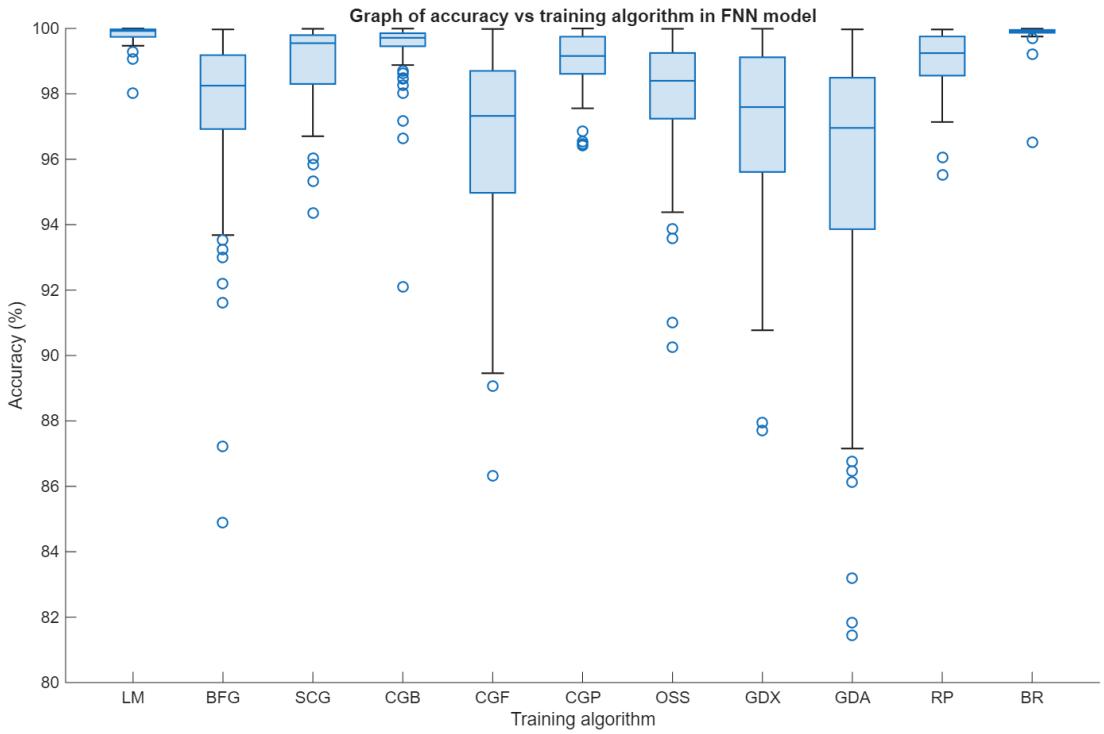


Figure 5: Boxplot of accuracy versus the 11 training algorithms in FFNN model.

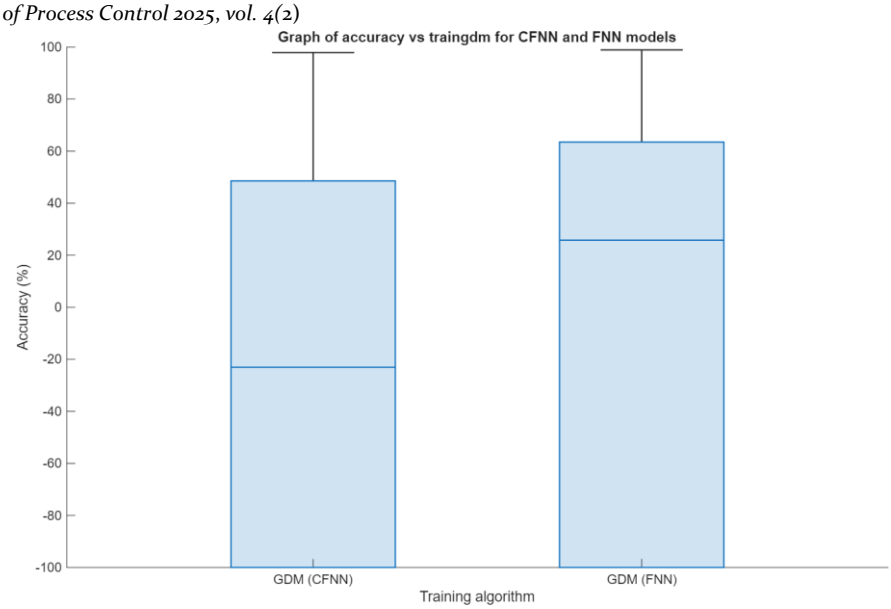


Figure 6: Boxplot of accuracy versus the GDM in CFNN and FNN models.

Figures 7 and 8 show the differences between predicted and actual values to observe the deviation of the modelled polymerization process from the experimental data. FFNN and CFNN models were assessed using the four best-performing training algorithms identified from Tables 2 and 3. Overall, both Figures 7 and 8 showed similar trends, with less significant deviations from the actual values. This suggests that CFNN and FNN models have the potential to represent the actual polymerization process. In contrast, Figure 9 shows a large deviation from the true value when using GDM algorithm. This demonstrates the importance of selecting an appropriate training algorithm. This also confirms that the GDM algorithm is unsuitable for predicting the polymerization process accurately.

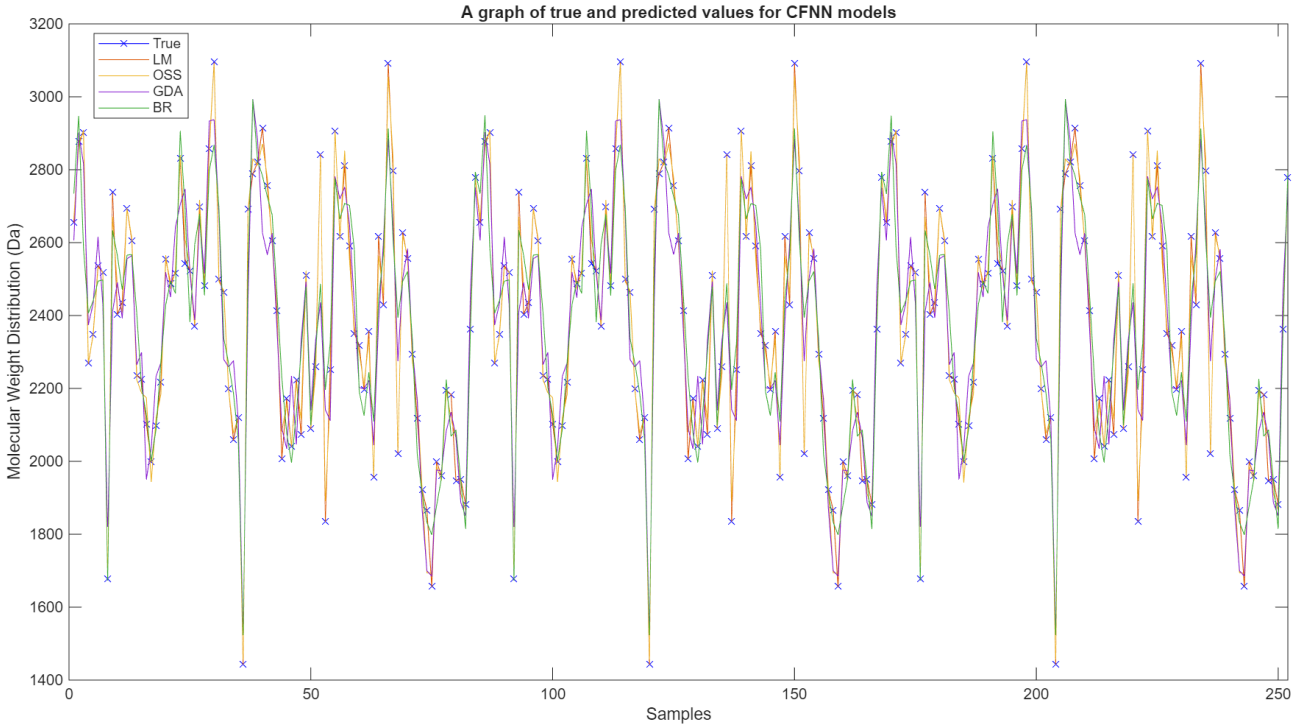


Figure 7: Actual and predicted values using CFNN.

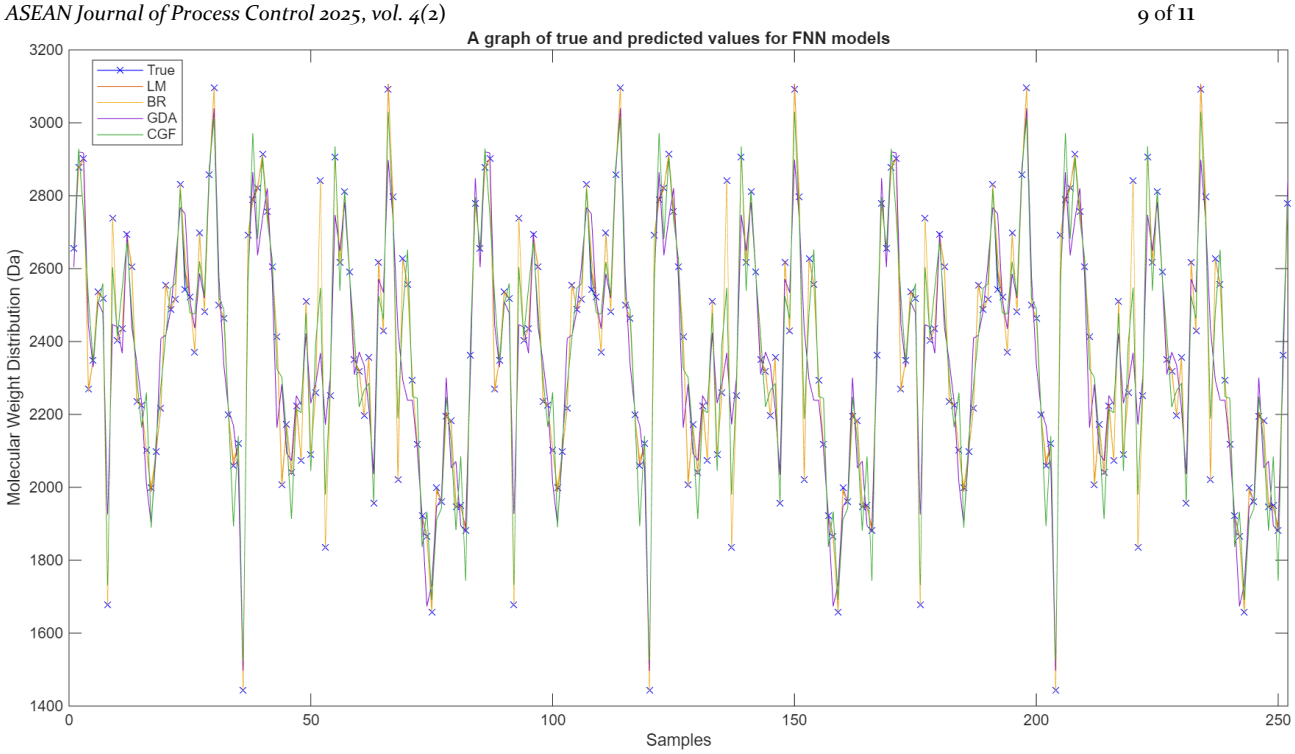


Figure 8: Actual and predicted values using FNN.

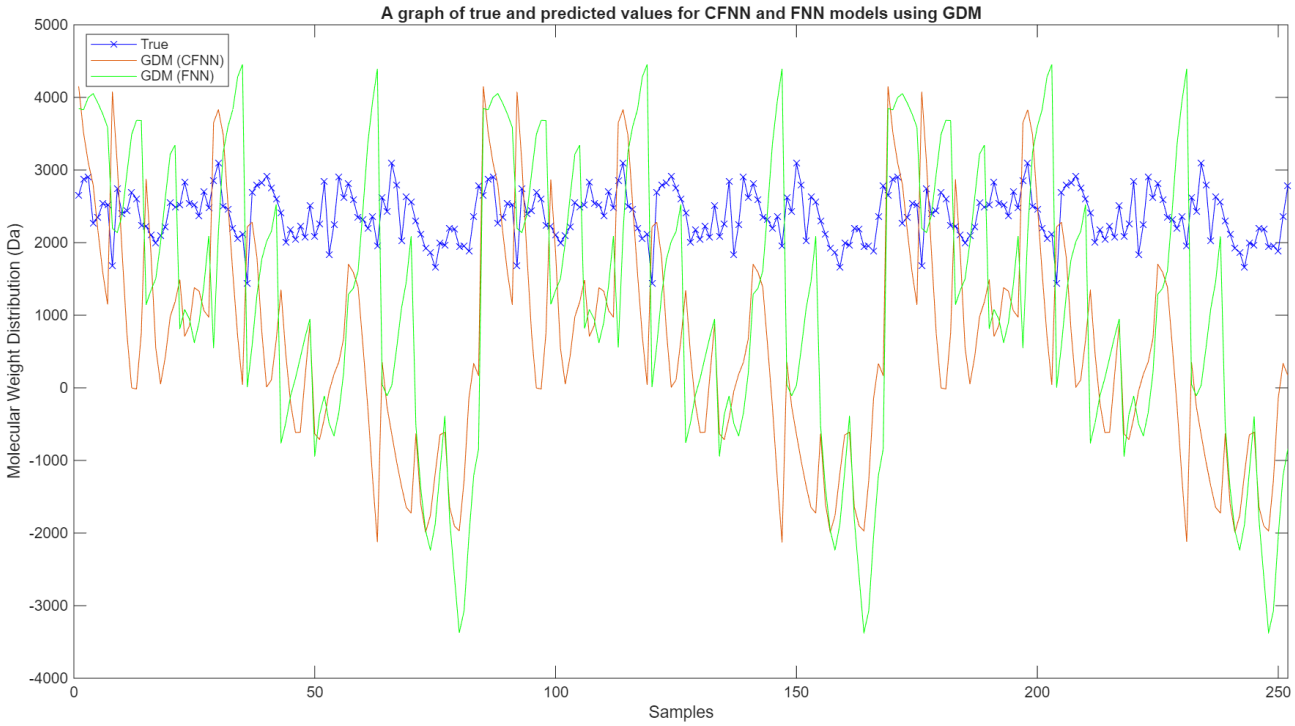


Figure 9: Actual and predicted values using CFNN and FFNN with GDM.

3.2 Sensitivity Analysis

As shown in Figure 10, impeller speed and temperature have a significant impact on the molecular weight of polycaprolactone during polymerization. The negative relevancy factors for both temperature and impeller speed inputs indicate that increasing these inputs would lead to a reduction in molecular weight inputs. Among the three inputs, impeller speed has the most significant impact, suggesting that mixing intensity is the most dominant factor in the molecular weight compared to speed and time. Reaction time shows minimal effect on the molecular weight, as it is hardly visible in Figure 10. This shows that the time input is less dominant in the molecular weight. These findings are consistent with the experimental study carried out by Arumugasamy [9], as lower temperatures are more effective in synthesizing higher-quality polymers. In the same study, the effect of impeller speed on molecular weight showed fluctuations across different temperatures, indicating a possible interaction between

speed and temperature that affects the molecular weight of polycaprolactone. Nevertheless, a general trend was observed that lower impeller speed increases the molecular weight, excluding the anomalously high value of molecular weight at 1000 rpm [9]. Furthermore, it is observed in the study that from the 1st and 7th hour, the molecular weight fluctuated at different conditions, with several experiments showing decreasing trends, increasing trends, or a combination of both. This inconsistency may suggest that time does not have a strong effect on molecular weight; instead, the molecular weight is strongly affected by other conditions, such as temperature and speed.

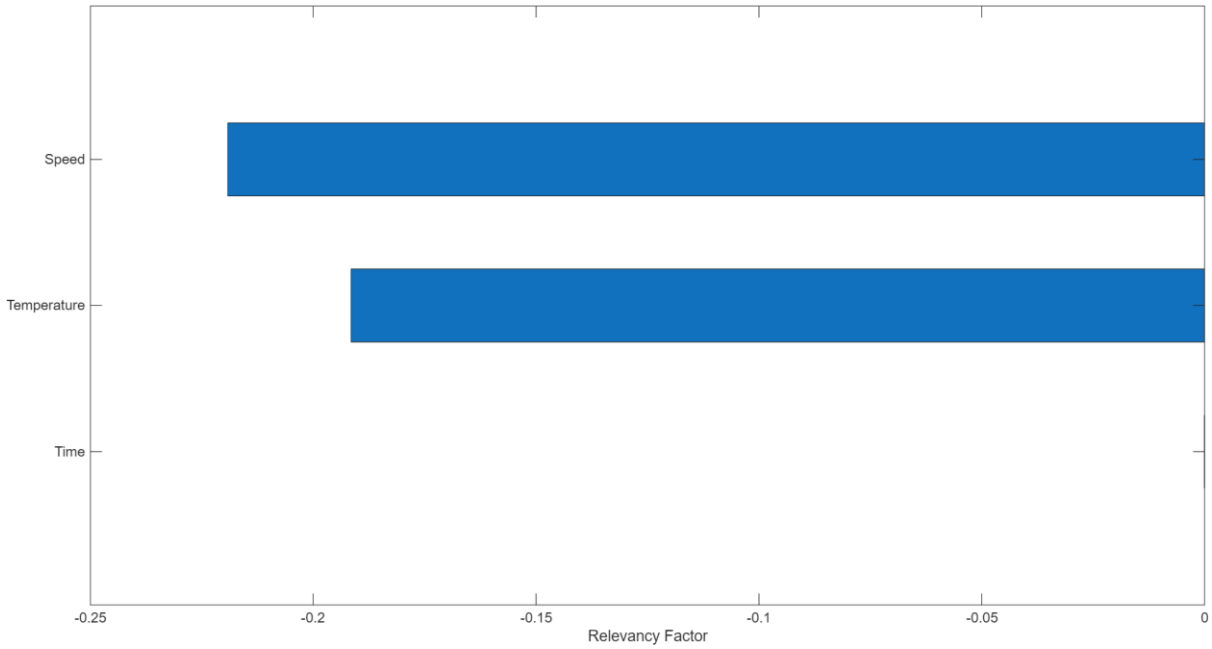


Figure 10: Sensitivity analysis using relevancy factor for CFNN with LM.

4. Conclusions

This study successfully developed and compared Cascade-Forward Neural Network (CFNN) and Feedforward Neural Network (FFNN) models for empirical modelling of enzymatic polymerization. Using 84 sets of experimental data, CFNN and FFNN were developed and evaluated across twelve training algorithms. The comparative results revealed that the CFNN model trained with the Levenberg–Marquardt backpropagation algorithm (LM) achieved the best performance, as it obtained the lowest MSE, RMSE, MAE, and MAPE values, as well as the highest regression coefficient (R = 1) and overall accuracy of 99.97%. In contrast, GDM and GDA performed poorly, with low regression values and large errors. Comparatively, FNN produced more stable results across algorithms compared to CFNN due to its overall higher accuracy. Overall, the findings confirmed that LM algorithm is highly suitable for non-linear modelling, where CFNN with LM showed the best performance in terms of the highest accuracy and the least predictive errors. Sensitivity analysis showed that temperature and impeller speed both heavily contributed to the molecular weight negatively. Future work should focus on optimizing hyperparameters, such as learning rate and iteration number, to minimize overfitting in complex networks and further enhance the accuracy and generalization of the CFNN models.

Acknowledgements:

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