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Comparison of Melt Flow Index of Propylene Polymerisation in Loop Reactors using First Principles and Artificial Neural Network Models

Nur Fazirah Jumari, Khairiyah Mohd-Yusof*

Center for Engineering Education, Universiti Teknologi Malaysia, Johor Bahru, Malaysia
khairiyah@cheme.utm.my

The inability to measure product quality in polymerisation industries on-line causes major difficulties. There are no on-line instruments to measure resin characteristics that define polymer quality, such as melt flow index (MFI) and density. MFI always often have to be evaluated in a time consuming and manpower intensive lab analysis. In most plants, MFI is measured only several times a day using a manual analytical test. An on-line MFI measurement is essential in fulfilling customer requirements and preventing losses. This paper presents models for soft sensors to measure MFI in industrial polypropylene loop reactors using first principle (FP) model and artificial neural network (ANN) model. For the FP model, two industrial interconnected loop reactors for propylene polymerisation are modelled as two continuous stirred tank reactors (CSTRs) in series. The mathematical models of nonlinear differential equations which describe the polymerisation process were solved numerically. The ANN model of the two loop reactors are developed by employing the concept of Feed-Forward Back Propagation (FFBP) network architecture using Levenberg-Marquardt training method. The ANN model act as estimator to predict the polymer MFI. Both models are developed and simulated in MATLAB. The simulation results of the MFI between FPM and ANN model are compared and analysed. The prediction of the ANN model is found to be more accurate compare to the MFI calculated by the FP model. The ANN model prediction is good within the range of training data. The CPU time recorded that ANN model is much faster than FP model.

1. Introduction

The major problem faced by the polymerisation industry is that the resin characteristics that define polymer quality, such as melt flow index (MFI) and density cannot be measured on-line. Properties, such as MFI, are difficult to measure and usually unavailable in real time since it requires close human intervention (Wang et al., 2016). They can only be measured off-line in the laboratory, which leads to difficulty in controlling product quality in polymerisation processes because of the delay involved before the product quality is known. In most plants, MFI is measured only several times a day using a manual analytical test. Products which do not meet the specifications must either be sold off at a reduced price or wasted. This does not only cause loss of revenue, but also resources, such as raw material, production time and energy.

An on-line product quality measurement like MFI is essential in fulfilling customer requirements and preventing losses. Since sensors are not available to measure MFI, developing a soft sensor is the next best alternative. Soft sensors are inferential estimators, drawing conclusions from process observations when hardware sensors are unavailable or unsuitable. A suitable, fast and robust process model for the polymerisation reactor is required so that the MFI can be estimated from the model. A typical soft sensor consists of three main pillars: the process model, the variables used by the model and an update technique. Soft sensor for inferential measurement can be developed using first principle method, multivariate statistical method, or a hybrid of both methods. A first principle model is very knowledge-intensive and is referred to as a “white box”, while multivariate method is data-driven and is usually named as a “black box” model. Development of “white-box” soft sensor is a complex process and computationally intensive for real time applications. It provides rich information about process behaviour as well relationship between process variables. In contrast, development

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of “black-box” soft sensor is simpler. It is a goal-driven approach in which only inferential measurement is required while all the comprehensive information about the process is hidden within the database.

1.1 First principle model for polymerisation

Zacca and Ray (1993) were the first to develop and simulate a comprehensive process model of loop reactors for propylene polymerisation. They modelled the loop reactors as two tubular sections interconnected by perfectly mixed inlet and outlet zones, using a distributed parameter system. It was found that at high recycle ratios, the ratio of volumetric flow rates between recycle stream and outlet stream, the loop reactors exhibit the behaviour of CSTRs. Luo et al. (2007) studied the molecular weight distribution and the weight average molecular weight of polypropylene produced in a single industrial loop reactor. The loop reactor modelled as a CSTR with temperature control and covered both steady-state and dynamic performance of the reactor. The comparison between simulation and industrial data was also presented. Lucca et al. (2008) modelled a single industrial loop reactor for liquid phase propylene polymerisation. The loop reactor was presented in the form that is similar to the one reported by Zacca and Ray (1993), and also modelled as a distributed parameter system. Modelling and simulation studies were conducted from an industrial perspective. The particular emphasis was placed on modelling the end-use properties, such as MFI, of the polymer resins. Method of moments that described the number of live and dead polymer chains in the polymerisation medium were applied to allow for calculation of average molecular weights, which was then used to predict the value of MFI and xylene soluble (XS).

In this work, the loop reactors are modelled as two ideal CSTRs where polymerisation follows the simplified kinetics according to single site mechanism. The process model is simulated and validated using industrial data. Dynamic inputs are utilised to demonstrate the great potential of the process model to be used for process monitoring and troubleshooting purposes in a polypropylene plant. As this process model has the ability to predict polymer slurry density, production rate and MFI, it can also be used as a computer-aided instrument to perform back-up online inferential measurements, in case of instrumentation failure (Jamaludin, 2012).

1.2 Artificial neural network model for polymerisation

In polymerisation, ANN was applied by Hincliffe et al. (2003) to develop a detailed mechanistic model of a polyethylene production process including material and energy balance to predict the reactor temperature, conversion and molecular weight distribution (MWD) and Gonzaga et al. (2009) focused on measuring viscosity as one of the most important product quality of polyethylene terephthalate (PET). In propylene polymerisation process, the model was developed to predict the end-used property of the final product. A virtual soft sensor to infer MFI of polypropylene was developed by Jian Shi et al. (2006) using neural network architecture that integrates independent combining analysis (ICA) and multi-scale analysis (MSA). The proposed method reported can provide prediction reliability and accuracy, which is capable of learning the relationships between process variables and the target MFI. Xia and Pan (2010) studied about inferential estimation of polypropylene MFI using stacked neural network based on absolute error criteria. The estimation errors can be reduced by using single neural network model and can be further reduced using stacked neural network model.

There are many researches that focus on developing the soft sensor using either first principle model or artificial neural network model as mention above. But no comparison of effectiveness has been done on both models. In this paper, FP model and ANN model were developed to predict the MFI of polypropylene in industrial loop reactors. Using these models, the MFI can be estimated from the measured process variables instantly. The objective of this work was to develop the Himont Spheripol Process model for propylene polymerisation to predict MFI value. Three main steps are required to accomplish the objectives of this work. These were model development (FP and ANN model), MATLAB simulation (FP and ANN model) and model comparison. The FP model was simulated from the model developed based on Zacca and Ray (1993) and Lucca et al. (2008) model. The ANN model used Feed-Forward Back Propagation (FFBP) network architecture and Levenberg-Marquardt as training method. The performance of the model was measured by comparing the value of root mean square error (RMSE) and correlation (R^2). The effectiveness of the two models is also being compared.

2. Process description

It is important to understand the process of polymerisation, especially the mechanism to produce FP model. FP model was to be considered not only the overall flow rates and compositions of the streams to and from the reactor system but also the kinetics of the reactions within the reactor vessels. Understanding the process also helps in determined the variables and parameters involved in developing ANN model.

Figure 1 shows the industrial loop reactors being modelled in this study. This figure illustrated the typical Petrochemical Plant in Malaysia. The reaction is a liquid phase propylene polymerisation which is a part of the Spheripol process. The Spheripol process comprises three steps, namely catalyst and raw material feeding, polymerisation and finishing. The fourth-generation of Ziegler Natta catalyst is used due to its high activity and stereospecificity (Albizzati et. al, 1996). The pre-polymer is created when the catalyst and propylene monomers are injected into the baby loop reactor. The pre-polymers are injected into the loop reactor along with monomers (propylene and ethylene) and chain transfer agent (hydrogen). Hydrogen is used to control the average molecular weights (consequently the MFI) of the polymer resins. Homopolymer and random copolymer are produced through the bulk polymerisation. Discharge from the first loop reactor is continuously fed into the second loop reactor. Fresh monomers and chain transfer agent are also injected into the second loop reactor. Discharge from the second loop reactor is flashed to separate the polymer particles from the non-reacted monomers. The stream of polymer particles is then sent to finishing processes.

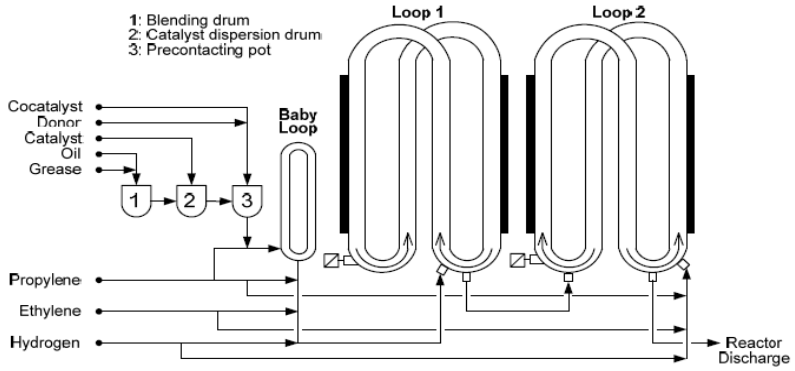

Figure 1: Process flow diagram of industrial loop reactor

3. Models development

3.1 First principle model

The loop reactors are modelled as two ideal CSTRs in this work where polymerisation follows the simplified kinetics according to single site mechanism. The process model is simulated and validated using industrial data. Dynamic inputs are utilised to demonstrate the great potential of the process model for process monitoring and troubleshooting purposes in a polypropylene plant.

The baby loop and other preliminary processes prior to the first loop reactor are not considered. Feed stream of catalyst into dispersion drum, co catalyst and donor into pre-contacting pot, and propylene into the baby loop are modelled as if they are fed directly into the first loop reactor. Feed stream of oil and grease into blending drum is not considered at all. This is justifiable since their flow rate is very small and they do not play any role in the polymerisation reaction.

Propylene polymerisation kinetics with Ziegler-Natta catalysts can be quite complex. A kinetic model that is similar to the one used by Lucca et al. (2008) is used in this paper while the corresponding kinetic parameters are tabulated in Table 1. Process outputs from the dynamic model are input variables for the static model. The output of this model will be dynamic as well. The static models developed are polymer WAMW and MFI since both are not a function of time. NAMW and WAMW of polymer resins are calculated from polymer moments and are given by Soares et al. (2007) where MW_M is the molar mass of the repeating unit (MW_M = 42 g/mol for propylene). Estimation of the polymer MFI is developed through a power-law model of the polymer WAMW and thus makes it valid in a limited range of WAMW. The correlation is developed for each batch and each range of polymer production. Figure 2 below shows the example of correlation of predicted MFI using the power-law of correlation.

3.2 Artificial Neural Network Model

The first step to develop ANN model is gaining information on the selected input data and requirements to build the network architecture. A thorough understanding of the reactor operation and process description is required. Before the network could be developed, the inputs and outputs must be clearly selected. The inputs variables were selected depending on the factor of dynamic behaviour of the chemical process and kinetic reaction of the polymerisation.

According to Fernandes and Lona (2005), for neural networks that have more inputs than output variables, one hidden layer is enough in most cases. In this paper, there are 12 input variables and one output variable. One hidden layer is enough for better prediction. ANN model in this work was developed in MATLAB environment and utilised MATLAB neural network toolbox. The models were trained using backpropagation algorithm. The data used are gained from the industrial Petrochemical Plant in Malaysia. The data are divided into two sets: set A consist of 38 data sets from industry, divided into 28 training data sets and 10 testing data sets and set B, 45 data sets from industry are divided into 33 training data sets and 12 testing data sets. Training set is used to train a neural network. The error of this data set is minimised during training. The test set is used to determine the performance of a neural network on patterns that are not trained during learning and for finally checking the overall performance of the neural network. The transfer function to be trained in the hidden layer is tansig-pureline. The Levenberg-Marquadt, trainlm, was chosen as the training algorithm due to the fastest back-propagation algorithm in the toolbox and it is highly recommended as a first choice algorithm.

Evaluations of the models are based on root mean square error (RMSE). The output of the network was compared to the target value of the testing data set by calculating the RMSE and the correlation coefficient (R^2). For each network structure that is being developed, the initial weights and biases which give the smallest RMSE were recorded. If the small RMSE value is not achieved, the network need to be recreated again using other architecture.

Table 1: Kinetic parameters (Zacca and Ray, 1993)

Kinetic parameters	Arrhenius equations
k_A (m^3 $kmol^{-1}$ s^{-1})	$7.94 \times 10^7 \exp(-12{,}000/RT)$
k_P (m^3 $kmol^{-1}$ s^{-1})	$4.97 \times 10^7 \exp(-12{,}000/RT)$
k_{TH} (m^3 $kmol^{-1}$ s^{-1})	$4.40 \times 10^6 \exp(-12{,}000/RT)$
k_{TM} (m^3 $kmol^{-1}$ s^{-1})	$6.16 \times 10^3 \exp(-12{,}000/RT)$
k_{TT} (m^3 $kmol^{-1}$ s^{-1})	$7.04 \times 10^2 \exp(-12{,}000/RT)$
k_{TS} (s^{-1})	$3.85 \times 10^2 \exp(-12{,}000/RT)$
k_D (s^{-1})	$7.92 \times 10^3 \exp(-12{,}000/RT)$

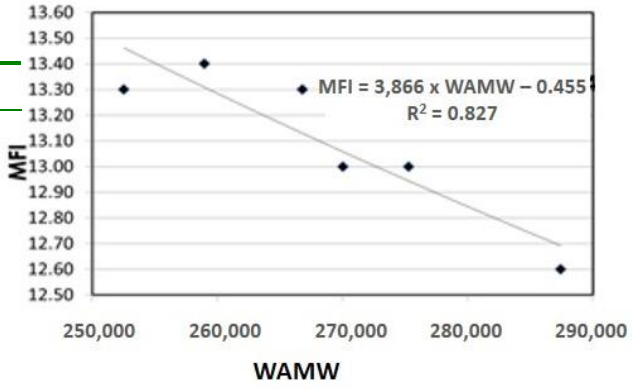

Figure 2: The example of MFI-WAMW relationship as a power-law-type model

4. Result and discussion

The result will be presented into profile data and random data. Profile data is the data that follow the sequence of the plant reading. Random data is the data that selected randomly without follow the order of the plant reading. The result shows the simulation from FP model, followed by ANN model. The diamond point in the Figure 3 and 4 stands for actual value and the circle point stand for predicted value.

4.1 FP Model as a soft sensor to predict MFI

The result on predicted MFI using FP model is discussed. Figure 3 shows the comparison between predicted MFI using FP model and measured value. From the figures, there is a small deviation between simulation result and plant data. It is mainly due to the complexity of coordination reaction in polymerisation process. In the reaction, the chain length of the polymer is depending on transfer reaction. The one of transfer agent in this reaction is hydrogen. Besides acting as transfer agent, hydrogen can also influence the polymerisation rate significantly. Most polymers contain chains of different lengths and they are characterised by the molecular weight distribution (MWD). MWD strongly affects the properties of the polymers. MWD is often characterised by the WAMW. WAMW of a polymer reflects inversely to its MFI, for instance, high MFI corresponds to low molecular weight of polymers.

The empirical model of MFI used in this study is obtained by developing a power-law-typed correlation between measured and simulated values. As lab test is not really accurate and is associated with error, the resulting empirical model would not be so accurate as well. The empirical model (Figure 2) is only valid in

certain batch of polymer production and certain range of MFI. The predicted MFI still lies within the specification of the product grade, and thus indicates reliability of the process model.

4.2 Artificial neural network model as a soft sensor to predict MFI

To develop the ANN model, the number of nodes in the hidden layer for estimating MFI is 6. Figure 4 illustrated the comparison result between predicted MFI using ANN model and measured data. The ANN model developed is a simple ANN model, but it enough to contribute for a better prediction. Jumari and Mohd-Yusof (2016) provides the details about ANN models development.

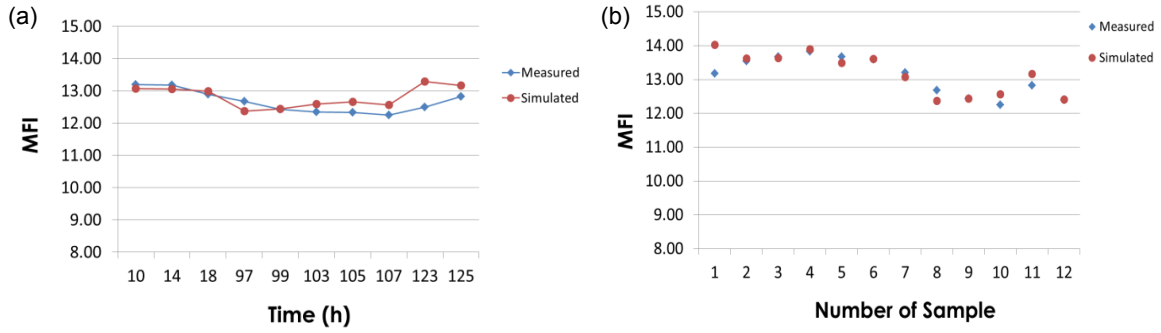

Figure 3: Predicted MFI using FP model a) Profile data, b) Random data

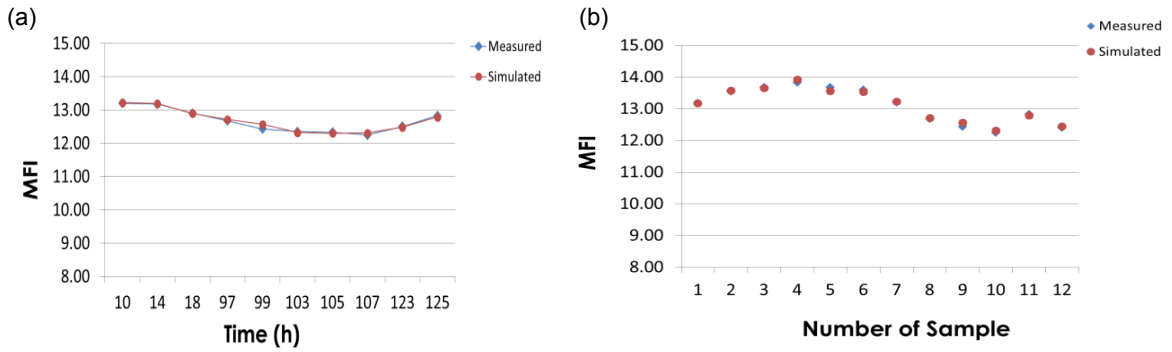

Figure 4: Predicted MFI using ANN model a) Profile data, b) Random data

4.3 Comparison between FP model and ANN model

The objective of this study is to develop the models and find the best model to predict accurate end used product quality, melt flow index (MFI), using first principle FP model and ANN model. The model develop will serve as a soft sensor to estimate melt flow index (MFI). To find the best model, FP model and ANN model is compared. Table 2 shows the R^2 and RMSE values for both models. In term of CPU time, ANN model is much better compare to FP model. To simulate both models, the CPU time recorded is 28.13 min for FP model and 0.32 min for ANN model. The result of this study proved that the ANN model is able to estimate the product quality value with the tolerable error. The best model developed to estimate the MFI value generated the lowest RMSE value and the highest R^2 value compared to FP model. ANN model has the ability to deal with complex and poorly understood behaviour. This model does not use any structure that reflects the physical structure of the system. The ANN model gives an input-output relation of the process and useful in understanding of the system in absent or not relevant for the purpose of the models.

Table 2: Comparison between FP model and ANN model

Model	Types of Data	R^2	RMSE
FP	Profile	0.331181	0.336571
	Random	0.756933	0.302930
ANN	Profile	0.942804	0.007517
	Random	0.978005	0.007630

FP model give a physical insight of the system in term of mathematical description of the model. The effort needed to build these models is high, certainly for complex chemical system. The FP model was developed based on accurate assumptions to simplify the equations. Some of the details in the process system might be neglected due to the complexity. The kinetic model has the limit in certain range of MFI. The model developed approximately accurate with the real process system. The probability to get the best prediction is not high. The results obtained still in the range of the actual value. FP model are not as quick and easy to build, but they have many advantages. In terms of simulation, first-principle models provide extrapolation in addition to the interpolation provided by ANN models. They also can be used for monitoring, control and optimisation. ANN models should be limited within the ranges of data used in their development. The measurement errors captured in the ANN model will lead to huge errors when the data is extrapolated outside the range of the training data. In this work, both models were tested with the out-range data. The FP and ANN model The RMSE for FP and ANN model are 0.380037 and 0.706215 when the out-range profile data is used.

5. Conclusion

In FP model, the MFI correlation was only valid for a certain range of MFI since it depends on the power of law of WAMW. The result obtained is acceptable since the predicted MFI still lies within the specification of the product grade. Compared to FP model, ANN model showed a better result based on lower RMSE values and CPU time. The model: FP model and ANN model output showed good agreement with industrial data. ANN model shows better performance compared to FP Model. In terms of simulation, first-principle models provide extrapolation and generate good performance compare to the ANN models. The study shows that the ANN model is better estimator within the estimation range.

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