MELT FLOW INDEX ESTIMATION USING NEURAL NETWORK MODELS FOR PROPYLENE POLYMERIZATION PROCESS

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To Allah~ for guiding me to the straight path,

To my other half~ for love, understanding, and support

To my sunshine~ for being with me along this journey

To family members, lecturers, close friends~ for endless care and assistances

and

Those people who have guided and inspired me throughout my journey of education and life

Thank you

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ABSTRACT

One of the major challenges in polymerization industry is the lack of online instruments to measure polymer end-used properties such as melt flow index (MFI). As an alternative to the online instruments and conventional laboratory tests, these properties can be estimated by using a model based-soft sensor. This research presents models for soft sensors to measure MFI in industrial polypropylene loop reactors by using the artificial neural network (ANN), hybrid FP-ANN (HNN) and stacked neural network (SNN) models. The ANN model of the two loop reactors was developed by employing the concept of Feed-Forward Back Propagation (FFBP) network architecture using Levenberg-Marquardt training method. Serial hybrid FP-ANN (HNN) models were developed in this study. The error between actual MFI and simulation MFI from FP model was fed into the HNN model as one of the input variables. To construct the stacked neural network (SNN) model, two layers were needed: 1) level-0 generalizer output comes from a number of diverse ANN models and 2) level-1 generalizer was developed using the results of level-0 generalizer with additional input variables. All models were developed and simulated in MATLAB 2009a environment. The simulation results of the MFI based on the ANN, HNN, and SNN models were compared and analyzed. The HNN model is the best model in predicting all range of MFI with the lowest root mean square error (RMSE) value, 0.010848, followed by ANN model (RMSE=0.019366) and SNN model (RMSE=0.059132). When these three models (ANN, HNN, and SNN) were compared, the SNN model shows the lower RMSE for each type of MFI studied.

ABSTRAK

Salah satu cabaran utama dalam industri pempolimeran ialah kekurangan instrumen dalam talian untuk mengukur sifat-sifat akhir polimer seperti indeks aliran lebur (MFI). Sebagai alternatif kepada instrumen dalam talian dan ujian makmal konvensional, sifat-sifat ini boleh dianggarkan menggunakan model berasaskan *soft sensor*. Kajian ini mempersembahkan model yang digunakan di dalam *soft sensor* untuk mengukur MFI di dalam reaktor gegelung industri pempolimeran propena menggunakan model *Artificial Neural Network* (ANN), model hibrid FP-ANN (HNN) dan model *Stacked Neural Network* (SNN). Model ANN bagi dua reaktor gegelung telah dibangunkan dengan menggunakan konsep *Feed-Forward Back Propagation* (FFBP) melalui kaedah latihan *Levenberg-Marquardt*. Model HNN bersiri telah dibangunkan juga dalam kajian ini. Ralat antara MFI yang dijana oleh model FP dan nilai MFI yang sebenar digunakan sebagai salah satu masukan untuk model HNN. Untuk membangunkan model SNN, dua tahap diperlukan: 1) keluaran dari *level-0 generalizer* datang dari pelbagai jenis model ANN dan 2) *generalizer level-1* dibangunkan menggunakan keputusan dari *level-0* yang dibina. Kesemua model telah dibangunkan dan disimulasikan di dalam persekitaran MATLAB 2009a. Keputusan simulasi MFI dari model ANN, HNN, dan SNN dibandingkan dan dianalisis. Model HNN adalah model yang terbaik jika mengukur semua nilai MFI dengan nilai *root mean square error* (RMSE) sebanyak 0.010848, diikuti dengan model ANN (RMSE=0.019366) dan model SNN (RMSE=0.059132). Jika ketiga-tiga model (ANN, HNN dan SNN) ini dibandingkan, model SNN menunjukkan nilai RMSE yang rendah bagi mengukur setiap jenis MFI yang dikaji.

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LIST OF ABBREVIATIONS

a_i	-	Input vector
ANN	-	Artificial neural network
b_j	-	Output vector
C3	-	Propylene
C_H^i	-	hydrogen concentration in reactor i (kgmol/m^3)
C_M^i	-	monomer concentration in reactor i (kgmol/m^3)
$C_{P_0}^i$	-	catalyst active site concentration in reactor i (kgmol/m^3)
$C_{S_D}^i$	-	catalyst dead site concentration in reactor i (kgmol/m^3)
C_T^i	-	cocatalyst concentration in reactor i (kgmol/m^3)
CSTR	-	continuous stirred-tank reactor
D_r	-	dead polymer chains
E	-	activation energy (J/mol)
$\mathrm{F}_\mathrm{M}^\mathrm{i}$	-	monomer mass flowrate into reactor i (kg/hr)
FP	-	First principle
H_2	-	Hydrogen
ICA	-	independent combining analysis
k_A	-	site activation kinetic constant (m^3/kgmol/s)
k_P	-	polymer propagation kinetic constant (m^3/kgmol/s)
k_{TH}	-	chain transfer to hydrogen kinetic constant (m^3/kgmol/s)
k_{TM}	-	chain transfer to monomer kinetic constant (m^3/kgmol/s)
k_{TT}	-	chain transfer to cocatalyst kinetic constant (m^3/kgmol/s)
k_{TS}	-	spontaneous chain transfer to hydrogen kinetic constant (1/s)
k_D	-	site deactivation kinetic constant (1/s)

K		buffer factor of the reactors
M	-	number of points used in the moving average
MFI	-	melt flow index (g/10min)
MSA	-	multi-scale analysis
MW_M	-	molecular weight of monomer (kgmol/kg)
MWD	-	molecular weight distribution
$NAMW$	-	number average molecular weight
PET	-	polyethylene terephthalate
P_0/P_{0f}	-	Outlet/ inlet active catalyst site concentration
PP	-	polypropylene
PR_{PP}^i	-	production rate of polypropylene in reactor i (kg/hr)
P_r	-	live polymer chains
PSD	-	Particle size distribution
Q_{in}^i	-	inlet volumetric flowrate into reactor i (m^3/hr)
Q_{out}^i	-	outlet volumetric flowrate from reactor i (m^3/hr)
R_{AE}	-	Rate of site activation by electron donor
R_{AM}	-	Rate of site activation by monomer
R_{AS}	-	Rate of spontaneous site activation
R_{P0}	-	Rate of initiation
R_{PR}	-	Rate of propagation
R_{TM}	-	Rate of chain transfer to monomer
R_{TH}	-	Rate of chain transfer to hydrogen
R_{TS}	-	Rate of spontaneous chain transfer
R_D	-	Rate of site deactivation
R_{TC}	-	Rate of chain transfer to co-catalyst
R_{TE}	-	Rate of chain transfer to electron donor
R_D	-	Rate of spontaneous site decativation
R_{DH}	-	Rate of site deactivation by hydrogen
S_P	-	Pure catalyst site
TEAL	-	Triethylaluminium
ρ^i	-	polymer slurry density inside reactor i (kg/m^3)
ρ_{in}^i	-	inlet polymer slurry density inside reactor i (kg/m^3)

ρ_M^i	-	density of monomer in reactor i (kg/m^3)
ρ_{PP}^i	-	density of polypropylene in reactor i (kg/m^3)
γ_{PP}^i	-	specific gravity of polypropylene in recator i
ψ_0^i	-	zeroth moment of live polymer chains in reactor i
ψ_1^i	-	first moment of live polymer chains in reactor i
ψ_2^i	-	second moment of live polymer chains in reactor i
λ_0^i	-	zeroth moment of dead polymer chains in reactor i
λ_1^i	-	first moment of dead polymer chains in reactor i
λ_2^i	-	second moment of dead polymer chains in reactor i
i	-	reactor, i = 1, 2
j	-	polymer moments, $j = 0^{th}, 1^{st}, 2^{nd}$
H	-	hydrogen
in	-	inlet
M	-	monomer
out	-	outlet
PP	-	polypropylene
T	-	cocatalyst

CHAPTER 1

INTRODUCTION

1.1 Project Background

Product quality is an important feature in the globally competitive polymer industry (Jianli *et. al*, 2002). With the skyrocketing cost of raw material and energy, stringent measures must be taken to meet quality requirements to remain competitive. According to Ray (1986), product quality is a much more complex issue in polymerization than in more conventional short reactions. Polymers are produced in various grades, according to customer needs. To meet customer requirements at an economically attractive cost, product quality measurement is therefore a necessity. However, not all variables can be measured directly. Due to the limitations of measurement device, it is often difficult to estimate important process variables such as product concentration and melt flow index (MFI). Polymer qualities often have to be evaluated in a time consuming and manpower intensive lab analysis. Thus, there is a need to find methods to estimate the product quality in real time.

Polymerization reactors are very well known for their complexity especially with regards to reactor design, polymerization recipes, uncertain reaction kinetics, and highly exothermic reaction, which lead to difficulty in product quality control. Developing suitable models are therefore necessary to estimate the essential variables necessary for measurement, control or optimization. For example, polypropylene (PP) which is normally produced in the loop reactors is one of the most complex process and varied because it involves the types of catalyst, the kind of reactor, and the effect of the recycle loop (M. Kim *et al*., 2005). The articles reported by Uvarov and Tsevetkova (1974), Lepski and Inkov (1977) and a number of other authors (Zacca and Ray, 1993; Debling *et al*., 1994; Soares, 2001; Jiang *et al*., 2002; Wei *et al*., 2002; Reginato *et al*., 2003; Luo *et al*., 2007; and Lucca *et al*., 2008) are some the significant researches in the polymerization modeling for loop reactors.

The Himont Spheripol process is one of the most widely used polymerization process that produce polyolefin using the loop reactors. This process takes place in the loop reactors filled with liquid propylene. A small loop reactor is used to prepolymerize the catalyst; the main polymerization, for homopolymer or random copolymer, takes place in one or two loop reactors. For impact copolymer production a gas phase reactor is requested after the loop reactors.

The ability to produce polymer resins which meet customer quality demands is the primary aim of the polymer industry. To meet customer requirements at an economically attractive cost, product quality measurement is therefore a necessity. Soft sensor is one of the possible solutions where the values of the desired variables which cannot be measured are inferred from the measured variables using the process model. Development of inferential systems for polymer properties is a very active research area in polymerization reactor control (Chan et al., 1993; Chan and Nascimento, 1994; McAuley *et al*.,1990; McAuley and MacGregor, 1991; Kiparissides *et al*., 1993; Zabisky *et al*., 1992; Skagerberg *et al*., 1992, Gonzaga *et.al*., 2009 etc.).

According to R. Sharmin *et. al*.,(2006), the model used to estimate polymer properties can be roughly categorized into three groups: (1) mechanistic model developed from first principles (for example, Chan *et al*., 1993; McAuley et al.,1990, Kiparissides et al., 1993; Zabisky *et al*., 1992), (2) black-box model using neural networks (Bhat and McAvoy, 1990; Chan and Nascimento, 1994; Qin and McAvoy, 1992; Rallo *et al*., 2002; Zhang *et al*., 1997, etc.), and (3) statistical model using multivariate statistical tools (Jaeckle and MacGregor, 1998; Kiparissides *et al*., 1993; MacGregor *et al*., 1994; Martin *et al*., 1999; Skagerberg *et al*., 1992).

M. Kim *et al*., (2005) developed a soft sensor in polypropylene process based on hybrid modelling of novel clustering and black-box as well as mechanistic models. Meanwhile, T. Yiagopoulos *et al*., (2004) also developed a soft sensor with a neural network approach for monitoring polymer melt flow index (MFI) at the Basell Spheripol process.

1.2 Problem Statement

The major problem faced by the polymerization 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 (R. Sharmin *et al*., 2006). They can only be measured off-line in the laboratory, which leads to difficulty in controlling product quality in polymerization processes because of the delay involved before the product quality is known. Consequently, 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.

Therefore, an on-line product quality measurement, such as 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 polymerization reactor is required so that the MFI can be estimated from the model.

1.3 Research Objective

The purpose of this research is to develop the Himont Spheripol Process model for propylene polymerization that can be a model as a soft sensor. The Himont Spheripol process model is developed to predict end-use product quality, melt flow index (MFI), by using the artificial neural network (ANN) model, hybrid neural network (HNN) model and stacked neural network (SNN) model.

1.4 Research Scope

The study in developing the models for a soft sensor for polypropylene loop reactors to produce homopolymer of Himont Spheripol Process has been identified as follows:

a) Develop and simulate the different types of neural network models (ANN,HNN and SNN) for polyproplylene loop reactors in MATLAB 2009a environment using industrial data collected from polypropylene plant in Johor.

b) Use the models developed (ANN, HNN, and SNN model) to estimate the different grades of MFI.
c) Validate the simulation results with MFI from industrial polypropylene plant data.
d) Compare the performance of the models developed (ANN, HNN, and SNN model) in predicting the MFI values.

1.5 Research Contribution

Major contribution in this study is the development of the models (ANN, HNN and SNN model) for a soft sensor to measure melt flow index of propylene polymerization process in loop reactors. The development of a comprehensive mathematical model is an essential task in understanding the polymerization processes and translating the best of our knowledge about the interactions of different factors that affect the system. The developed, programmed and simulated process model for the soft sensors provides a basic study that can be used and improved in further study which related to bulk propylene polymerization, such as process optimization and process control. The process model also provides an understanding of the dynamic behaviour of propylene polymerization in loop reactors from an industry perspective.

Another contribution is to compare and determine the best models for a soft sensor that can be utilized in describing the propylene polymerization process. The models are very useful as a guide in industry to help the engineers and operators understand the process clearly for the purpose of plant monitoring and troubleshooting as well as operator training.

1.6 Summary of Chapter 1

Producing polymer is a complex and difficult process. The polymer is produced in various grades according to customer requirement at an economically attractive cost. However, it is often difficult to estimate or measure important process variables such as melt flow index (MFI). MFI always often have to be evaluated in a time consuming and manpower intensive lab analysis. They can only be measured off-line in the laboratory, which leads to difficulty in controlling product quality in polymerization processes because of the delay involved before the MFI value is known. Consequently, 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. Thus, there is a need to find methods to estimate the melt flow index in real time. Therefore, an on-line MFI measurement is essential in fulfilling customer requirements and preventing losses. The purpose of this research is to develop the Himont Spheripol Process model for propylene polymerization that can be a model as a soft sensor. The Himont Spheripol process takes place in the loop reactors filled with liquid propylene. The Himont Spheripol process model is developed to predict end-use product quality, melt flow index (MFI), by using artificial neural network (ANN) model, hybrid neural network (HNN) model and stacked neural network (SNN) model.

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