Optimal Control of A Batch Emulsion Copolymerisation Reactor Based on Recurrent Neural Network Models

Yuan Tian, Jie Zhang and Julian Morris
Centre for Process Analytics and Control Technology
University of Newcastle, Newcastle upon Tyne NE1 7RU, U. K.
Phone: +44-191-2227240; Fax: +44-191-2225292
E-mail: {yuan.tian, jie.zhang, julian.morris}@newcastle.ac.uk

ABSTRACT: A recurrent neural network based non-linear dynamic modelling and optimal control strategy for a batch emulsion copolymerisation reactor is proposed. To avoid the excessive effort and time associated with the development of a detailed mechanistic model, recurrent neural networks are used to build an empirical model to represent the complex polymerisation process. Based on the developed neural network model, the sequential quadratic programming (SQP) method was used to calculate the optimal temperature profile leading to a polymer product with a desired number average molecular weight, desired copolymer composition and the highest conversion. By changing the constraint in the objective function, investigation of the process physics has been made to demonstrate the effect of temperature control on the overall polymer conversion. This scheme exploits the advantages of neural network model based optimal control policies and is tested by simulation showing good performance.

KEYWORDS: Optimal control, batch processes, neural networks, long range prediction, emulsion polymerisation.

INTRODUCTION

Polymer reaction engineering is a fast growing, multi-disciplinary field which combines a number of science and engineering subjects, including polymer chemistry, polymer physics, colloidal and interfacial phenomena, thermal dynamics, advanced analytical characterisation methods, reaction engineering, numerical analysis, optimisation and control (Kiparissides, 1996). Polymerisation reactors play a key role in polymer engineering, with the great importance of different polymer products. A major feature of polymerisation reactors is their highly complex and non-linear characteristics. Modelling of polymerisation processes is therefore very consequential as many approaches have given considerable effort on it (e.g. Richards and Congalidis, 1989).

Since the molecular and morphological properties of a polymer product strongly influence its physical, chemical, thermal, rheological, mechanical properties as well as the polymer's end-use applications, the importance and complexity of the polymer quality control have to be underscored (e.g. Kiparissides, 1996). The requirement of an accurate process model for the optimal quality control is becoming more and more important (e.g. Guntern et al., 1998; Thomas and Kiparissides, 1984; Chen and Weigand, 1994; Soroush and Kravaris, 1992). Development of comprehensive mathematical models to predict the polymer quality in terms of process operating conditions in a polymer reactor has shown to be a very important methodology to the efficient control of product quality (e.g. Richards and Congalidis, 1989; Urquiola et al., 1993; Dube et al., 1996; Vega et al., 1997; Saldivar et al., 1998). However, due to the excessive effort and time associated with the development of detailed mechanistic models, empirical models, which need to have sufficient representation ability to enable the underlying system characteristics to be approximated with an acceptable accuracy, are promising alternatives of mechanistic models. Several types of data based empirical models have been applied to polymerisation reactors including mutlivariate statistic approach (MacGregor et al., 1994) and neural network approach (e.g., Zhang et al., 1998b).

In this study, neural network modelling scheme is the focus on dealing with the highly non-linear polymerisation processes. Neural network have been shown to be able to approximate any continuous non-linear functions (e.g. Cybenko, 1989; Girosi and Poggio, 1990; Park and Sandberg, 1991) and have been applied to process modelling and control (e.g. Bulsari, 1995; Morris et al., 1994; Zhang et al., 1998a). Neural networks for non-linear process modelling can be broadly divided into two categories: static networks, including multi-layer feed forward neural networks and radial basis function networks, and dynamic networks, which include globally recurrent neural networks (Su et al., 1992), locally recurrent neural networks (Zhang et al., 1998a), Elman networks (Elman, 1990), and dynamic filter networks (Morris et al., 1994). Static neural networks are suitable for situations where short-term predictions are the main focus and are relatively easy to develop and train. Dynamic neural networks, used in this study, are more appropriate for the building of long term prediction models (e.g. Su et al., 1992; Zhang et al., 1998a) and usually require much longer training times than static networks.

Affiliated works have been done based on the emulsion copolymerisation batch processes and different methods were used for the modelling of polymerisation processes (e.g., Penlidis et al., 1985; Rawlings and Ray, 1988; Vega et al., 1997). However, there are not many works on the optimisation of batch emulsion copolymerisation processes, especially with the neural network methodology. In this paper, one specific polymerisation process is studied and it is a batch emulsion copolymerisation reactor (Richards and Congalidis, 1989). Simulation of the batch emulsion copolymerisation reactor was used to represent the real reactor. With the experimental data sets from the simulation programme, recurrent neural network models were built. Based on the neural network models, different optimisation objectives were realised in conformance with the industrial need and economic requirement.

A BATCH EMULSION COPOLYMERIZATION REACTOR

In general, polymerisation processes can be broadly classified into homogeneous and heterogeneous processes. The homogeneous process comprises polymerisation which is carried out in a single phase. In heterogeneous systems, either the polymer is insoluble in the monomer phase or the polymerisation involves the presence of different phases (Kiparissides, 1996). More explicitly, polymerisation processes can be categorised into the following processes: 1) Bulk, 2) Solution, 3) Precipitation, 4) Suspension, 5) Emulsion, 6) Solid catalysed polymerisation, 7) Interfacial polycondensation, and 8) Solid state.

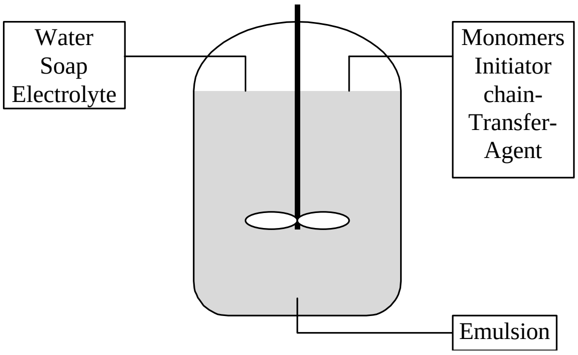

Figure 1. A batch emulsion copolymerisation reactor

The production of polymers by emulsion copolymerisation processes is of significant industrial importance (e.g. Saldivar et al., 1997). Emulsion copolymers offer a unique mix of properties compared with homo-polymers. Emulsion copolymerisation is a heterogeneous process and the physical characteristics of emulsion copolymerisation are very complex due to the presence of multiple phases, multiple monomers, radical species, and other ingredients: complex particle formation mechanisms, and the possibility of many modes of reactor operation. In contrast to the other polymerisation methods, the primary advantage of the emulsion copolymerisation process is that it is possible to obtain polymers of very high molecular weight at a relatively fast rate.

Figure 1 shows the batch emulsion reactor and the polymerisation recipe. A typical recipe always includes a dispersion medium (usually water), monomer(s), emulsifier (surfactant), and initiator. Normally, a water-soluble initiator is used and the monomers are either virtually insoluble, or else scatteredly soluble in the dispersion phase. With the decomposition of the initiator, new particles appear and consist of a number of polymer molecules of high degree of polymerisation, stabilised by surfactant and swollen with monomer.

According to the kinetic mechanism, a mathematical model (in the form of a set of differential equations) has been developed to simulate the copolymerisation process (Richards and Congalidis, 1989). The model covers material balances, the particle number concentration balance, phase volume balances, energy balance, and the calculation of important secondary variables. Some key variables representing polymer quality and production include conversion, copolymer composition and number average molecular weight.

For a batch reactor, the monomers, water and emulsifier are loaded into the reactor at the beginning. Then the initiator is loaded when the reactor temperature reaches its set-point. The initial conditions used in this study are given in Table 1.

Table 1. Initial conditions of the batch emulsion copolymerisation reactor

Initial Conditions			
Monomer 1	1.862 mol/m^3	Electrolyte	1.d-3 mol/m^3
Monomer 2	1.25 mol/m^3	Chain Transfer Agent	0.011 mol/m^3
Initiator	3.90d-3 mol/m^3	Emulsifier	0.033 mol/m^3
Emulsion Volume	6.23d-3 lit	Water Volume	0.642 lit

NEURAL NETWORK MODELLING

In this study, recurrent neural networks were used to model the batch emulsion copolymerisation process. Eight batches of data were produced through simulation. The sampling time is 120 seconds. Two of them were used as validation data. The simulated data were used as if they were obtained from a real reactor since the simulation of the process is credible. According to the previous knowledge of the emulsion copolymerisation process, the number average molecular weight, copolymer composition, polymer conversion, also control policy - temperature profile are the most necessary parameters in the quality control of this polymerisation reactor. The aim here is to optimise the temperature profile to get the highest polymer conversion with desired number average molecular weight and copolymer composition.

A multi-neural-network-modelling method is used to model the key quality variables in this process. In order to satisfy the objectives, three network models have been built to model the polymer conversion, number average molecular weight and copolymer composition. Quality specification such as average molecular weight and particle size, copolymer composition as well as production are closely related to the monomer conversion in the reactor. It has been tested that introducing monomer conversion as one of the input of the other two neural networks is necessary. Tables 2 to 4 show the comparisons of the sum of squared errors (SSE) among different network architectures.

Table 2. SSE for different network architectures of the *Conv* model

Neural network structures		SSE
***Temp*(*t*-1)**	***Conv*(*t*-1)**	**0.0264**
Temp(*t*-1) (*t*-2)	*Conv*(*t*-1)	0.4652
Temp(*t*-1) (*t*-2)	*Conv*(*t*-1) (*t*-2)	0.4601
Temp(*t*-1) (*t*-2)(*t*-3)	*Conv*(*t*-1)	276.9688

Table 3. SSE for different network architectures of the *Mn* model

Neural network structures			SSE
Temp(*t*-1)	*Conv*(*t*-1)	*Mn*(*t*-1)	18.6208
Temp(*t*-1)(*t*-2)	*Conv*(*t*-1)	*Mn*(*t*-1)	17.3899
Temp(*t*-1)(*t*-2)(*t*-3)	*Conv*(*t*-1)	*Mn*(*t*-1)	8.0507
Temp(*t*-1)(*t*-2)	*Conv*(*t*-1)(*t*-2)	*Mn*(*t*-1)	17.4817
***Temp*(*t*-1)(*t*-2)**	***Conv*(*t*-1)**	***Mn*(*t*-1)(*t*-2)**	**7.2343**

Table 4. SSE for different network architectures of the *Com* model

Neural network structures			SSE
	Conv(*t*-1)(*t*-2)	*Com*(*t*-1)(*t*-2)	97.4571
Temp(*t*-1)(*t*-2)	*Conv*(*t*-1)	*Com*(*t*-1)	8.2838
Temp(*t*-1)(*t*-2)(*t*-3)	*Conv*(*t*-1)	*Com*(*t*-1)	6.7376
***Temp*(*t*-1)(*t*-2)**	***Conv*(*t*-1)(*t*-2)**	***Com*(*t*-1)**	**3.6059**
Temp(*t*-1)(*t*-2)	*Conv*(*t*-1)	*Com*(*t*-1)(*t*-2)	6.8472

The best representations of the three network models can be summarised as the following forms:

$$\hat{Conv}(t) = f(Temp(t-1), \hat{Conv}(t-1)) \quad (1)$$

$$\hat{Com}(t) = f(Temp(t-1), Temp(t-2), \hat{Conv}(t-1), \hat{Conv}(t-2), \hat{Com}(t-1)) \quad (2)$$

$$\hat{Mn}(t) = f(Temp(t-1), Temp(t-2), \hat{Conv}(t-1), \hat{Mn}(t-1), \hat{Mn}(t-2)) \quad (3)$$

where *Conv*, *Mn*, *Com*, and *Temp* represent polymer conversion, number average molecular weight, copolymer composition, and reactor temperature respectively.

Due to the dynamic nature of recurrent neural networks, they are able to provide very accurate multi-step-ahead predictions. It is obvious from Figures 2 to 4, the multi-step-ahead predictions of copolymer composition, conversion and number average molecular weight are quite accurate.

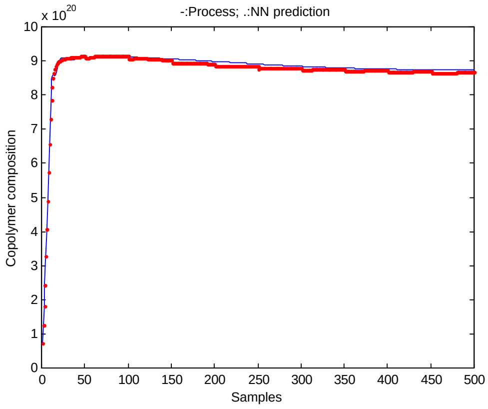

Figure 2. Multi-step ahead predictions of copolymer composition

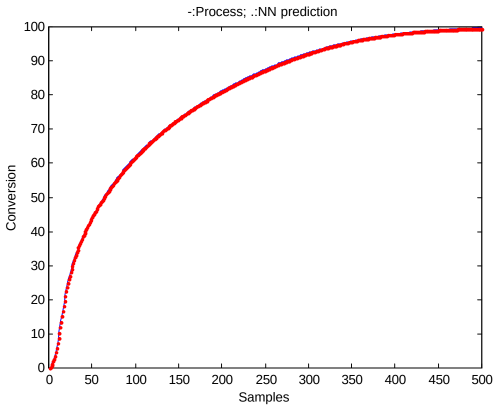

Figure 3. Multi-step ahead predictions of polymer conversion

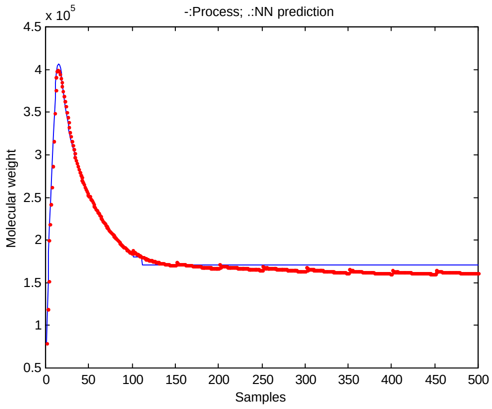

Figure 4. Multi-step ahead predictions of number average molecular weight

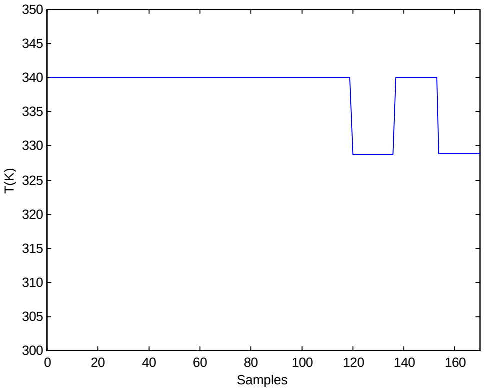

Figure 5. Optimal temperature profile when the batch ending time is set to 340 minutes

OPTIMIZATION CONTROL OF A BATCH COPOLYMERISATION REACTOR

Process optimisation and control can have a significant strategic impact on polymer plant operability and economics. Polymer production facilities face increasing pressures for production cost reductions and more stringent quality requirements. However, product quality is a much more complex issue in polymerisation than in more conventional short chain reactions. Because the molecular architecture of the polymer is so sensitive to reactor operating conditions, upsets in feed conditions, mixing, reaction temperature, can alter critical molecular properties such as molecular weight distributions, copolymer composition distribution, etc. The ultimate goal in operating a batch polymerisation reactor is to produce a final product with certain chemical or mechanical properties. The optimisation objectives could be decided according to: 1), molecular property specifications; 2), safety; 3), energy consumption; 4), reactor and environmental constraints; and 5), economic objectives.

In this study, the main objective is to achieve a high conversion with satisfactory polymer quality through reactor temperature control. The performance index for this batch reactor can be defined as:

$$J = a_1(Conv - 100)^2 + a_2 T_{end} \tag{4}$$

where $a_1 = 0.01, a_2 = 0.03$ are the weighting factors and T_{end} is the batch ending time in minutes. Temperature control policy is divided into 10 stages of equal time length. Let $U = [u_1\ u_2\ \ldots\ u_{10}]^T$ be the vector of control actions and the optimisation problem was formulated as:

$$\min_U J = a_1(Conv - 100)^2 + a_2 T_{end}$$

subject to:

$$290 \le Temp \le 340$$

$$1.7\times10^5 \le Mn \le 2.0\times10^5$$

$$8.5\times10^{20} \le Com \le 10\times10^{20}$$

To solve this optimisation problem, Sequential Quadratic Programming (SQP) methodology was employed. This optimisation scheme mimics Newton's method for constrained optimisation in that at each major iteration an approximation is made of the Hessian of the Lagrangian function using a quasi-Newton updating method. This is then used to generate a QP sub-problem whose solution is used to form a search direction for a line search procedure. In order to show clearly the optimisation method, several different batch ending time were chosen to account for the optimal temperature profile that could satisfy the multi-objectives, as well as time concerns. Table 4 shows the different optimal results based upon the various batches ending time. Figures 5 to 8 demonstrate the temperature control policies for different batch ending time.

Table 4. Comparison of the performance index under different batch ending time (Sample time = 2min)

Batch ending time (min)	340	**360**	380	400
Temperature profile (K)	Figure 5	**Figure 6**	Figure 7	Figure 8
Conversion (%)	78.2351	**80.0181**	81.5012	82.9631
Performance index (*J*)	14.9371	**14.7928**	14.8220	14.9026

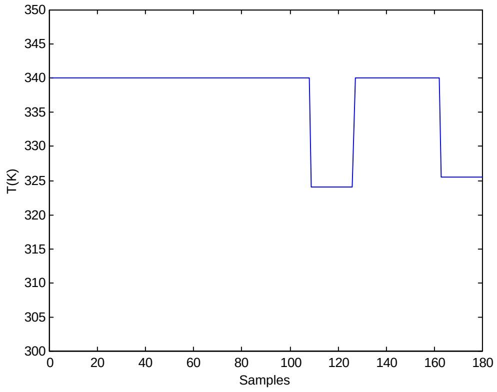

Figure 6. Optimal temperature profile when the batch ending time is set to 360 minutes

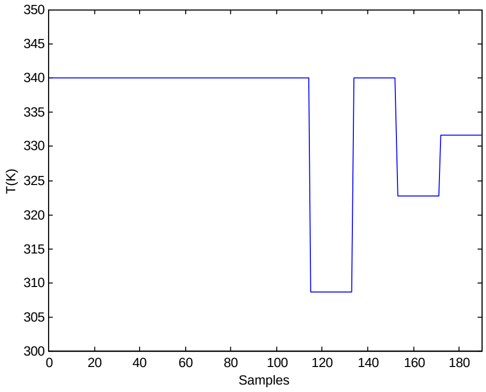

Figure 7. Optimal temperature profile when the batch ending time is set to 380 minutes

Compare to the best isothermal emulsion copolymerisation at 327.3K which requires a batch reaction time of 388 minutes to attain a conversion of 80% under the same initial conditions, the optimal temperature control policy could save about 30 minutes to achieve the same conversion. By fixing the batch ending time to 400 minutes, it is pragmatic to produce polymers with different desired molecular weight characteristics. By varying the desired number average molecular weight *Mn*, different temperature control policies were obtained. Temperature profiles are plotted as a function of time in Figure 9. All of the optimal profiles have been tested back to the simulation programme and could get almost the same quality properties results.

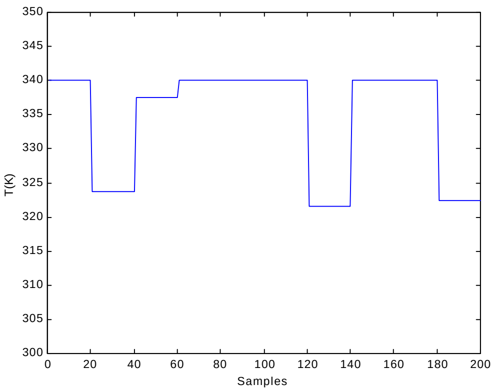

Figure 8. Optimal temperature profile when the batch ending time is set to 400 minutes

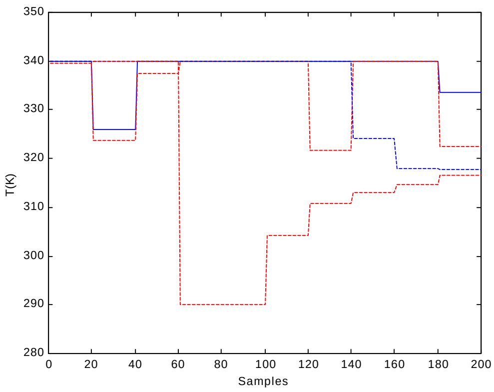

Figure 9. Optimal temperature profiles based on different number average molecular weight objectives (–: 1.4×10^5 =Mn =2.0×10^5; —: 1.5×10^5 =Mn =2.0×10^5; –.: 1.6×10^5 =Mn =2.0×10^5; …: 1.7×10^5 =Mn =2.0×10^5)

Analysis of this emulsion copolymerisation batch reactor exhibits that with the decreasing of the lower constraint of the number average molecular weight, the optimal temperature profile tends to be higher to achieve the highest polymer conversion. Reactor temperature has a great effect upon the polymer conversion. When other conditions permitted, higher temperature will result in higher conversion but at the same time lower final number average molecular weight.

CONCLUSION

The neural network modelling methodology gives a very good representation of the emulsion copolymerisation process. Time optimal control of this reactor based on the neural network model holds a very promising aspect of the model based optimal control, designing control policies for batch emulsion copolymerisation processes that produce polymers with properties satisfying multiple objectives. In this paper, the methodology was introduced to produce a copolymer with desired composition and desired number average molecular weight. The neural network model gives perfect representation of the real process, and provides an efficient and simple way of time optimal control of the emulsion copolymerisation process, not only in this batch emulsion reactor, but also applicable to any systems with high non-linear characteristics. Further studies will be carried out on hybrid modelling of this reactor which combines a simplified mechanistic model and a neural network model. Optimal monomer/initiator addition policies will also be studied in the future.

Acknowledgements – The research is financially supported by a European Union ESPRIT Project - MONNET (No.22416). The authors would like to thank colleagues in CPACT for their constructive comments and Yuan Tian acknowledges CPACT and the MONNET project for supporting his Ph.D. study.

REFERENCE

Bulsari, A. B., *Computer-Aided Chemical Engineering: Vol. 6, Neural Networks for Chemical Engineers*, Elsevier, Amsterdam, 1995.

Chen, Q., Weigand, W.A., “Dynamic optimization of nonlinear processes by combining neural net model with UDMC”, *AIChE Journal*, Vol. 40, No. 9, 1994, pp. 1488-1497.

Cybenko, G., “Approximation by superposition of a sigmoidal function”, *Math. Control Signal Systems*, Vol.2, 1989, pp303-314.

Dube, M.A., Penlidis, A., Mutha, R.K., Cluett, W.R., “Mathematical modelling of emulsion copolymerization of acrylonitrile/butadiene”, *Industrial & Engineering Chemistry Research*, Vol.35, No.12, Dec 1996, pp.4434-4448.

Elman, J. L., “Finding structures in time”, *Cognitive Science*, Vol.14, 1990, pp179-211.

Girosi, F. and Poggio, T., “Networks and the best approximation property”, *Biological Cybernetics*, Vol.63, 1990, pp169-179.

Guntern, C., Keller, A. H., Hungerbuhler, K., “Economic optimization of an industrial semibatch reactor applying dynamic programming”, *Ind. Eng. Chen. Res.*, Vol. 37, 1998, pp. 4017-4022.

Kiparissides, C., “Polymerisation reactor modelling: a review of recent developments and future directions”, *Chemical Engineering Science*, Vol. 51, No. 10, 1996, pp. 1637-1659.

MacGregor, J.F., Jaeckle, C., Kiparissides, C., and Koutoudi, M., “Process monitoring and diagnostics by multiblock PLS methods”, *AIChE Journal*, Vol.40, 1994, pp826-838.

Morris, A.J., Montague, G.A., and Willis, M.J., “Artificial neural networks: Studies in process modelling and control”, *Trans. IChemE*, Vol.72, Part A, 1994, pp3-19.

Park, J., Sandberg, I. W., “Universal approximation using radial basis function networks”, *Neural Computation,* Vol3, 1991, pp246-257.

Pendilis, A., MacGregor, J.F., Hamielec, A.E., “Dynamic modeling of emulsion polymerisation reactors”, *AIChE Journal*, Vol. 31, 1985, pp. 881.

Rawlings, J.B., Ray, W.H., “The modelling of batch and continuous emulsion polymerisation reactors”, *Polym. Eng. Sci*., Vol. 28(5), 1988, pp. 237.

Richards, J. R., Congalidis, J. P., “Mathematical modelling of emulsion copolymerisation reactors”, *Journal of Applied Polymer Science*, Vol. 37, 1989, pp.2727-2756.

Saldivar, E., Dafniotis, P., Ray, W.H., “Mathematical modelling of emulsion copolymerisation reactors. I. Model formulation and application to reactors operating with micellar nucleation”, *Journal of Macromolecular Science - Reviews in Macromolecular Chemistry and Physics*, Vol.C38, No.2, May 1998, pp.207-325.

Saldivar, E., Ray, W.H., “Control of semicontinuous emulsion copolymerization reactors”, *AIChE Journal*, Vol. 43, No. 8, 1997, pp. 2021-2033.

Soroush, M., Kravaris, C., “Nonlinear control of a batch polymerization reactor: an experimental study”, *AIChE Journal*, Vol. 38, No. 9, 1992, pp. 1429-1448.

Su, H. T., McAvoy, T. J. and Werbos, P., “Long-term prediction of chemical processes using recurrent neural networks: a parallel training approach”, *Industrial & Engineering Chemistry Research*, Vol.31, 1992, pp1338-1352.

Thomas, I. M., Kiparissides, C., “Computation of the near-optimal temperature and initiator policies for a batch polymrization reactor”, *The Canadian Journal of Chemical Engineering*, Vol. 62, 1984, pp. 284-291.

Urquiola, M.B., Sudol, E.D., Dimonie, V.L., ElAasser, M.S., “Emulsion polymerisation of vinyl acetate using a polymerisable surfactant. III. Mathematical model”, *Journal of Polymer Science, Part A: Polymer Chemistry*, Vol.31, No.6, May 1993, pp.1403-1415.

Vega, J.R., Gugliotta, L.M., Bielsa, R.O., Brandolini, M.C., Meira, G.R., “Emulsion copolymerization of acrylonitrile and butadiene. Mathematical model of an industrial reactor”, *Industrial & Engineering Chemistry Research*, Vol.36, No.4, Apr 1997, pp.1238-1246.

Zhang, J., Morris, A. J., Martin, E. B., “Long-term prediction models based on mixed order locally recurrent neural networks”, *Computers & Chemical Engineering*, Vol.22, 1998a, pp1051-1063.

Zhang, J., Morris, A.J., Martin, E.B., Kiparissides, C., “Prediction of polymer quality in batch hpolymerisation reactors using robust neural networks”, *Chemical Engineering Journal*, Vol. 69, 1998b, pp135-143.